Supplementary Information

Conventional ESR spectral analyses and quantum chemical calculations of high-spin cobalt(II) complexes with large ZFS tensors revisited: Exact **g**- and hyperfine tensors' relationships between fictitious spin-1/2 and true spin Hamiltonian approaches

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Double Perturbation Treatment for Degenerate States

In this section, in order to obtain the energies of the spin Hamiltonian having the sizable zero-field splitting (ZFS), electron-Zeeman and hyperfine terms, we consider the Rayleigh–Schrödinger perturbation for the energies and states of a Hamiltonian ($\mathcal{H}$) composed of a non-perturbed term (H_0: a rank-2 ZFS Hamiltonian in this work) and two perturbing terms (H_1 and H_2: the electron-Zeeman and hyperfine structure Hamiltonians, respectively, in this work). Note that the non-perturbed term is not the electron-Zeeman term which is regarded as a non-perturbed one in putative and conventional high-field approximation treatments. The current approach is termed a Zeeman perturbation treatment, which is not common but has a big advantage over the putative ones in many aspects, exemplifying the derivation of the analytical expressions for bridging the gap between a true spin Hamiltonian and fictitious spin-1/2 Hamiltonian relationships, as described in this work.

The Hamiltonian $\mathcal{H}$ is in the following form of

$$\mathcal{H} = H_0 + \lambda H_1 + \mu H_2 \tag{1}$$

$$\mathcal{H}\left|\Psi_{n,\alpha}\right\rangle = E_{n,\alpha}\left|\Psi_{n,\alpha}\right\rangle \tag{2}$$

where the energy eigenvalues of the non-perturbed Hamiltonian $\varepsilon_n^{(0)}$ are assumed to be g_n-th degenerate.

$$H_0\left|\varphi_{n,\alpha}^{(0)}\right\rangle = \varepsilon_n^{(0)}\left|\varphi_{n,\alpha}^{(0)}\right\rangle \ (\alpha = 1, 2, \dots, g_n) \tag{3}$$

This assumption is underlain by the Kramers' degeneracy for the electronic spin systems with half-integer spin quantum numbers. The wavefunctions and the energies can be expanded as follows:

$$\left|\Psi_{n,\alpha}\right\rangle = \left|\varphi_{n,\alpha}^{(0)}\right\rangle + \lambda\left|\varphi_{n,\alpha}^{(10)}\right\rangle + \mu\left|\varphi_{n,\alpha}^{(01)}\right\rangle + \lambda^2\left|\varphi_{n,\alpha}^{(20)}\right\rangle + \lambda\mu\left|\varphi_{n,\alpha}^{(11)}\right\rangle + \mu^2\left|\varphi_{n,\alpha}^{(02)}\right\rangle + \cdots \tag{4}$$

$$E_{n,\alpha} = \varepsilon_n^{(0)} + \lambda\varepsilon_{n,\alpha}^{(10)} + \mu\varepsilon_{n,\alpha}^{(01)} + \lambda^2\varepsilon_{n,\alpha}^{(20)} + \lambda\mu\varepsilon_{n,\alpha}^{(11)} + \mu^2\varepsilon_{n,\alpha}^{(02)} + \cdots \tag{5}$$

In the equations above, a factor of 1/2 is omitted from the quadratic perturbation parameter. Supposing an application to the spin Hamiltonian, the matrix elements of H_1 expanded by the wavefunctions $\left|\varphi_{n,\alpha}^{(0)}\right\rangle$ $(\alpha = 1, 2, \dots, g_n)$, having the same eigenenergy $\varepsilon_n^{(0)}$, are assumed as

$$\left\langle\varphi_{n,\beta}^{(0)}\middle|H_1\middle|\varphi_{n,\alpha}^{(0)}\right\rangle = 0 \tag{6}$$

for any $\beta \neq \alpha$. As described below, the matrix elements of the electron-Zeeman Hamiltonian have non-zero values between the eigenstates belonging the different eigenvalues. Degenerate perturbation treatment has to be applied to the second perturbing term (H_2).

The eigenstates of the non-perturbed Hamiltonian $\left|\varphi_{n,\alpha}^{(0)}\right\rangle$ is orthonormal and complete, as given in the following.

$$\left\langle\varphi_{n,\alpha}^{(0)}\middle|\varphi_{m,\beta}^{(0)}\right\rangle = \delta_{nm}\delta_{\alpha\beta} \tag{7}$$

$$\sum_{n=0}^{\infty}\sum_{\alpha=1}^{g_n}\left|\varphi_{n,\alpha}^{(0)}\right\rangle\left\langle\varphi_{n,\alpha}^{(0)}\right| = 1 \tag{8}$$

Also, the projection operator to the n-th eigenstate is defined as follows:

$$P_n = \sum_{\alpha=1}^{g_n} \left|\varphi_{n,\alpha}^{(0)}\right\rangle \left\langle\varphi_{n,\alpha}^{(0)}\right| \tag{9}$$

From eqs. (8) and (9), we obtain

$$\sum_{n=0}^{\infty} P_n = 1 \tag{10}$$

The new basis set $\left|\phi_{n,\alpha}^{(0)}\right\rangle$ is made from $\left|\varphi_{n,\alpha}^{(0)}\right\rangle$ by using a unitary transformation.

$$\left|\phi_{n,\alpha}^{(0)}\right\rangle = \sum_{\beta=1}^{g_n} c_{n\alpha}{}^{\beta} \left|\varphi_{n,\beta}^{(0)}\right\rangle \tag{11}$$

The coefficients $c_{n,\alpha}{}^{\beta}$ is the α-th row and β-th column matrix elements of $g_n \times g_n$ unitary matrix. The new basis $\left|\phi_{n,\alpha}^{(0)}\right\rangle$ is also the eigenstate of the non-perturbed Hamiltonian:

$$H_0 \left|\phi_{n,\alpha}^{(0)}\right\rangle = \varepsilon_n^{(0)} \left|\phi_{n,\alpha}^{(0)}\right\rangle \ (\alpha = 1, 2, \dots, g_n) \tag{12}$$

Also, $\left|\phi_{n,\alpha}^{(0)}\right\rangle$ satisfies the orthonormality condition in the following,

$$\left\langle\phi_{n,\alpha}^{(0)}\middle|\phi_{m,\beta}^{(0)}\right\rangle = \delta_{nm}\delta_{\alpha\beta} \tag{13}$$

and the completeness.

$$\sum_{n=0}^{\infty}\sum_{\alpha=1}^{g_n} \left|\phi_{n,\alpha}^{(0)}\right\rangle\left\langle\phi_{n,\alpha}^{(0)}\right| = 1 \tag{14}$$

Then, the wavefunctions can be rewritten by using $\left|\phi_{n,\alpha}^{(0)}\right\rangle$:

$$\begin{aligned}\left|\Psi_{n,\alpha}\right\rangle = &\left|\phi_{n,\alpha}^{(0)}\right\rangle + \lambda\left|\varphi_{n,\alpha}^{(10)}\right\rangle + \mu\left|\varphi_{n,\alpha}^{(01)}\right\rangle + \lambda^2\left|\varphi_{n,\alpha}^{(20)}\right\rangle + \lambda\mu\left|\varphi_{n,\alpha}^{(11)}\right\rangle + \mu^2\left|\varphi_{n,\alpha}^{(02)}\right\rangle + \lambda^3\left|\varphi_{n,\alpha}^{(30)}\right\rangle \\ &+ \lambda^2\mu\left|\varphi_{n,\alpha}^{(21)}\right\rangle + \lambda\mu^2\left|\varphi_{n,\alpha}^{(12)}\right\rangle + \mu^3\left|\varphi_{n,\alpha}^{(03)}\right\rangle + \cdots\end{aligned} \tag{15}$$

Substituting eqs. (1), (5) and (15) to the time-independent Schrödinger equation eq. (2) yields

$$\begin{aligned}(H_0 + \lambda H_1 + \mu H_2)\Big(&\left|\phi_{n,\alpha}^{(0)}\right\rangle + \lambda\left|\varphi_{n,\alpha}^{(10)}\right\rangle + \mu\left|\varphi_{n,\alpha}^{(01)}\right\rangle + \lambda^2\left|\varphi_{n,\alpha}^{(20)}\right\rangle + \lambda\mu\left|\varphi_{n,\alpha}^{(11)}\right\rangle + \mu^2\left|\varphi_{n,\alpha}^{(02)}\right\rangle \\ &+ \lambda^3\left|\varphi_{n,\alpha}^{(30)}\right\rangle + \lambda^2\mu\left|\varphi_{n,\alpha}^{(21)}\right\rangle + \lambda\mu^2\left|\varphi_{n,\alpha}^{(12)}\right\rangle + \mu^3\left|\varphi_{n,\alpha}^{(03)}\right\rangle + \cdots\Big)\end{aligned} \tag{16}$$

$$= \left(\varepsilon_n^{(0)} + \lambda\varepsilon_{n,\alpha}^{(10)} + \mu\varepsilon_{n,\alpha}^{(01)} + \lambda^2\varepsilon_{n,\alpha}^{(20)} + \lambda\mu\varepsilon_{n,\alpha}^{(11)} + \mu^2\varepsilon_{n,\alpha}^{(02)} + \lambda^3\varepsilon_{n,\alpha}^{(30)} + \lambda^2\mu\varepsilon_{n,\alpha}^{(21)} + \lambda\mu^2\varepsilon_{n,\alpha}^{(12)} + \mu^3\varepsilon_{n,\alpha}^{(03)} + \cdots\right)\left(\left|\phi_{n,\alpha}^{(0)}\right\rangle + \lambda\left|\varphi_{n,\alpha}^{(10)}\right\rangle + \mu\left|\varphi_{n,\alpha}^{(01)}\right\rangle + \lambda^2\left|\varphi_{n,\alpha}^{(20)}\right\rangle + \lambda\mu\left|\varphi_{n,\alpha}^{(11)}\right\rangle + \mu^2\left|\varphi_{n,\alpha}^{(02)}\right\rangle + \lambda^3\left|\varphi_{n,\alpha}^{(30)}\right\rangle + \lambda^2\mu\left|\varphi_{n,\alpha}^{(21)}\right\rangle + \lambda\mu^2\left|\varphi_{n,\alpha}^{(12)}\right\rangle + \mu^3\left|\varphi_{n,\alpha}^{(03)}\right\rangle + \cdots\right)$$

The coefficients of both the sides of eq. (16) are compared in the order of λ and μ, giving the following

zeroth
$$H_0\left|\phi_{n,\alpha}^{(0)}\right\rangle = \varepsilon_n^{(0)}\left|\phi_{n,\alpha}^{(0)}\right\rangle \tag{17}$$

λ
$$H_0\left|\varphi_{n,\alpha}^{(10)}\right\rangle + H_1\left|\phi_{n,\alpha}^{(0)}\right\rangle = \varepsilon_n^{(0)}\left|\varphi_{n,\alpha}^{(10)}\right\rangle + \varepsilon_{n,\alpha}^{(10)}\left|\phi_{n,\alpha}^{(0)}\right\rangle \tag{18}$$

μ
$$H_0\left|\varphi_{n,\alpha}^{(01)}\right\rangle + H_2\left|\phi_{n,\alpha}^{(0)}\right\rangle = \varepsilon_n^{(0)}\left|\varphi_{n,\alpha}^{(01)}\right\rangle + \varepsilon_{n,\alpha}^{(01)}\left|\phi_{n,\alpha}^{(0)}\right\rangle \tag{19}$$

λ^2
$$H_0\left|\varphi_{n,\alpha}^{(20)}\right\rangle + H_1\left|\varphi_{n,\alpha}^{(10)}\right\rangle = \varepsilon_n^{(0)}\left|\varphi_{n,\alpha}^{(20)}\right\rangle + \varepsilon_{n,\alpha}^{(10)}\left|\varphi_{n,\alpha}^{(10)}\right\rangle + \varepsilon_{n,\alpha}^{(20)}\left|\phi_{n,\alpha}^{(0)}\right\rangle \tag{20}$$

$\lambda\mu$
$$H_0\left|\varphi_{n,\alpha}^{(11)}\right\rangle + H_1\left|\varphi_{n,\alpha}^{(01)}\right\rangle + H_2\left|\varphi_{n,\alpha}^{(10)}\right\rangle = \varepsilon_n^{(0)}\left|\varphi_{n,\alpha}^{(11)}\right\rangle + \varepsilon_{n,\alpha}^{(10)}\left|\varphi_{n,\alpha}^{(01)}\right\rangle + \varepsilon_{n,\alpha}^{(01)}\left|\varphi_{n,\alpha}^{(10)}\right\rangle + \varepsilon_{n,\alpha}^{(11)}\left|\phi_{n,\alpha}^{(0)}\right\rangle \tag{21}$$

μ^2
$$H_0\left|\varphi_{n,\alpha}^{(02)}\right\rangle + H_2\left|\varphi_{n,\alpha}^{(01)}\right\rangle = \varepsilon_n^{(0)}\left|\varphi_{n,\alpha}^{(02)}\right\rangle + \varepsilon_{n,\alpha}^{(01)}\left|\varphi_{n,\alpha}^{(01)}\right\rangle + \varepsilon_{n,\alpha}^{(02)}\left|\phi_{n,\alpha}^{(0)}\right\rangle \tag{22}$$

The relationship of the zeroth-order, eq. (17), is equivalent to eq. (12).

λ: We calculate the energy $\varepsilon_{n,\alpha}^{(10)}$. $\left\langle\varphi_{m,\beta}^{(0)}\right|$ is multiplied to both the sides of eq. (18) from the left.

$$\left\langle\varphi_{m,\beta}^{(0)}\middle|H_0\middle|\varphi_{n,\alpha}^{(10)}\right\rangle + \left\langle\varphi_{m,\beta}^{(0)}\middle|H_1\middle|\phi_{n,\alpha}^{(0)}\right\rangle = \varepsilon_n^{(0)}\left\langle\varphi_{m,\beta}^{(0)}\middle|\varphi_{n,\alpha}^{(10)}\right\rangle + \varepsilon_{n,\alpha}^{(10)}\left\langle\varphi_{m,\beta}^{(0)}\middle|\phi_{n,\alpha}^{(0)}\right\rangle \tag{23}$$

Using Schrödinger equation of the non-perturbed Hamiltonian eq. (3) and eq. (24),

$$\left\langle\varphi_{m,\beta}^{(0)}\middle|\phi_{n,\alpha}^{(0)}\right\rangle = c_{n\alpha}{}^{\beta}\delta_{mn} \tag{24}$$

eq. (23) is rewritten as follows,

$$\varepsilon_{n,\alpha}^{(10)} c_{n\alpha}{}^{\beta} \delta_{mn} = \sum_{\gamma=1}^{g_n} c_{n\alpha}{}^{\gamma} \left\langle \varphi_{m,\beta}^{(0)} \middle| H_1 \middle| \varphi_{n,\gamma}^{(0)} \right\rangle + \left(\varepsilon_m^{(0)} - \varepsilon_n^{(0)} \right) \left\langle \varphi_{m,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(10)} \right\rangle \tag{25}$$

When $m = n$, eq. (25) is

$$\varepsilon_{n,\alpha}^{(10)} c_{n\alpha}{}^{\beta} = \sum_{\gamma=1}^{g_n} c_{n\alpha}{}^{\gamma} \left\langle \varphi_{n,\beta}^{(0)} \middle| H_1 \middle| \varphi_{n,\gamma}^{(0)} \right\rangle \tag{26}$$

And if $\beta \neq \gamma$, $\left\langle \varphi_{n,\beta}^{(0)} \middle| H_1 \middle| \varphi_{n,\gamma}^{(0)} \right\rangle = 0$ due to (6), then

$$\varepsilon_{n,\alpha}^{(10)} = \left\langle \varphi_{n,\beta}^{(0)} \middle| H_1 \middle| \varphi_{n,\beta}^{(0)} \right\rangle \tag{27}$$

Next, we consider the first-order correction of the wavefunction. Multiplying $\left\langle \phi_{m,\beta}^{(0)} \right|$ to both the sides of eq. (18) from the left yields

$$\left\langle \phi_{m,\beta}^{(0)} \middle| H_0 \middle| \varphi_{n,\alpha}^{(10)} \right\rangle + \left\langle \phi_{m,\beta}^{(0)} \middle| H_1 \middle| \phi_{n,\alpha}^{(0)} \right\rangle = \varepsilon_n^{(0)} \left\langle \phi_{m,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(10)} \right\rangle + \varepsilon_{n,\alpha}^{(10)} \left\langle \phi_{m,\beta}^{(0)} \middle| \phi_{n,\alpha}^{(0)} \right\rangle \tag{28}$$

Using the orthonormality condition (eq. (13)),

$$\varepsilon_{n,\alpha}^{(10)} \delta_{nm} \delta_{\alpha\beta} = \left\langle \phi_{m,\beta}^{(0)} \middle| H_1 \middle| \phi_{n,\alpha}^{(0)} \right\rangle + \left(\varepsilon_m^{(0)} - \varepsilon_n^{(0)} \right) \left\langle \phi_{m,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(10)} \right\rangle \tag{29}$$

Thus, when $m \neq n$,

$$\left\langle \phi_{m,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(10)} \right\rangle = - \frac{\left\langle \phi_{m,\beta}^{(0)} \middle| H_1 \middle| \phi_{n,\alpha}^{(0)} \right\rangle}{\varepsilon_m^{(0)} - \varepsilon_n^{(0)}} \tag{30}$$

This is one of the coefficients of the perturbed wavefunctions in the first order.

μ: Multiplying $\left\langle \varphi_{m,\beta}^{(0)} \right|$ to both the sides of eq. (19) from the left yields

$$\left\langle \varphi_{m,\beta}^{(0)} \middle| H_0 \middle| \varphi_{n,\alpha}^{(01)} \right\rangle + \left\langle \varphi_{m,\beta}^{(0)} \middle| H_2 \middle| \phi_{n,\alpha}^{(0)} \right\rangle = \varepsilon_n^{(0)} \left\langle \varphi_{m,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(01)} \right\rangle + \varepsilon_{n,\alpha}^{(01)} \left\langle \varphi_{m,\beta}^{(0)} \middle| \phi_{n,\alpha}^{(0)} \right\rangle \tag{31}$$

Using Schrödinger equation of the non-perturbed Hamiltonian eq. (3) and eq. (24), eq. (31) is given as

$$\varepsilon_{n,\alpha}^{(01)} c_{n\alpha}{}^{\beta} \delta_{mn} = \sum_{\gamma=1}^{g_n} c_{n\alpha}{}^{\gamma} \left\langle \varphi_{m,\beta}^{(0)} \middle| H_2 \middle| \varphi_{n,\gamma}^{(0)} \right\rangle + \left(\varepsilon_m^{(0)} - \varepsilon_n^{(0)} \right) \left\langle \varphi_{m,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(01)} \right\rangle \tag{32}$$

When $m = n$, eq. (32) is

$$\varepsilon_{n,\alpha}^{(01)} c_{n\alpha}{}^{\beta} = \sum_{\gamma=1}^{g_n} c_{n\alpha}{}^{\gamma} \left\langle \varphi_{n,\beta}^{(0)} \middle| H_2 \middle| \varphi_{n,\gamma}^{(0)} \right\rangle \tag{33}$$

Equation (33) can be expanded as follows:

$$\varepsilon_{n,\alpha}^{(01)} c_{n\alpha}{}^{1} = c_{n\alpha}{}^{1} \left\langle \varphi_{n,1}^{(0)} \middle| H_2 \middle| \varphi_{n,1}^{(0)} \right\rangle + c_{n\alpha}{}^{2} \left\langle \varphi_{n,1}^{(0)} \middle| H_2 \middle| \varphi_{n,2}^{(0)} \right\rangle + \cdots + c_{n\alpha}{}^{g} \left\langle \varphi_{n,1}^{(0)} \middle| H_2 \middle| \varphi_{n,g}^{(0)} \right\rangle$$

$$\varepsilon_{n,\alpha}^{(01)} c_{n\alpha}{}^{2} = c_{n\alpha}{}^{2} \left\langle \varphi_{n,2}^{(0)} \middle| H_2 \middle| \varphi_{n,1}^{(0)} \right\rangle + c_{n\alpha}{}^{2} \left\langle \varphi_{n,2}^{(0)} \middle| H_2 \middle| \varphi_{n,2}^{(0)} \right\rangle + \cdots + c_{n\alpha}{}^{g} \left\langle \varphi_{n,2}^{(0)} \middle| H_2 \middle| \varphi_{n,g}^{(0)} \right\rangle \tag{34}$$

$$\vdots$$

$$\varepsilon_{n,\alpha}^{(01)} c_{n\alpha}{}^{g} = c_{n\alpha}{}^{g} \left\langle \varphi_{n,g}^{(0)} \middle| H_2 \middle| \varphi_{n,1}^{(0)} \right\rangle + c_{n\alpha}{}^{2} \left\langle \varphi_{n,g}^{(0)} \middle| H_2 \middle| \varphi_{n,2}^{(0)} \right\rangle + \cdots + c_{n\alpha}{}^{g} \left\langle \varphi_{n,g}^{(0)} \middle| H_2 \middle| \varphi_{n,g}^{(0)} \right\rangle$$

where g_n is represented as just g. Equation (34) can be represented in the following matrix form:

$$\begin{pmatrix} \left\langle \varphi_{n,1}^{(0)} \middle| H_2 \middle| \varphi_{n,1}^{(0)} \right\rangle & \left\langle \varphi_{n,1}^{(0)} \middle| H_2 \middle| \varphi_{n,2}^{(0)} \right\rangle & \cdots & \left\langle \varphi_{n,1}^{(0)} \middle| H_2 \middle| \varphi_{n,g}^{(0)} \right\rangle \\ \left\langle \varphi_{n,2}^{(0)} \middle| H_2 \middle| \varphi_{n,1}^{(0)} \right\rangle & \left\langle \varphi_{n,2}^{(0)} \middle| H_2 \middle| \varphi_{n,2}^{(0)} \right\rangle & \cdots & \left\langle \varphi_{n,2}^{(0)} \middle| H_2 \middle| \varphi_{n,g}^{(0)} \right\rangle \\ \vdots & \vdots & \ddots & \vdots \\ \left\langle \varphi_{n,g}^{(0)} \middle| H_2 \middle| \varphi_{n,1}^{(0)} \right\rangle & \left\langle \varphi_{n,g}^{(0)} \middle| H_2 \middle| \varphi_{n,2}^{(0)} \right\rangle & \cdots & \left\langle \varphi_{n,g}^{(0)} \middle| H_2 \middle| \varphi_{n,g}^{(0)} \right\rangle \end{pmatrix} \begin{pmatrix} c_{n\alpha}{}^{1} \\ c_{n\alpha}{}^{2} \\ \vdots \\ c_{n\alpha}{}^{g} \end{pmatrix} = \varepsilon_{n,\alpha}^{(01)} \begin{pmatrix} c_{n\alpha}{}^{1} \\ c_{n\alpha}{}^{2} \\ \vdots \\ c_{n\alpha}{}^{g} \end{pmatrix} \tag{35}$$

In this equation, the matrix elements of the left-hand side are the second perturbing Hamiltonian expanded by the *original* wavefunctions with the eigenvalue $\varepsilon_n^{(0)}$. That is, $\left\langle \varphi_{n,\beta}^{(0)} \middle| H_2 \middle| \varphi_{n,\gamma}^{(0)} \right\rangle$ forms $g_n \times g_n$ Hermite matrix whose eigenvalue is $\varepsilon_{n,\alpha}^{(01)}$. Therefore, $\varepsilon_{n,\alpha}^{(01)}$ is the solution of the secular equation:

$$\begin{vmatrix} \left\langle \varphi_{n,1}^{(0)} \middle| H_2 \middle| \varphi_{n,1}^{(0)} \right\rangle - x & \left\langle \varphi_{n,1}^{(0)} \middle| H_2 \middle| \varphi_{n,2}^{(0)} \right\rangle & \cdots & \left\langle \varphi_{n,1}^{(0)} \middle| H_2 \middle| \varphi_{n,g}^{(0)} \right\rangle \\ \left\langle \varphi_{n,2}^{(0)} \middle| H_2 \middle| \varphi_{n,1}^{(0)} \right\rangle & \left\langle \varphi_{n,2}^{(0)} \middle| H_2 \middle| \varphi_{n,2}^{(0)} \right\rangle - x & \cdots & \left\langle \varphi_{n,2}^{(0)} \middle| H_2 \middle| \varphi_{n,g}^{(0)} \right\rangle \\ \vdots & \vdots & \ddots & \vdots \\ \left\langle \varphi_{n,g}^{(0)} \middle| H_2 \middle| \varphi_{n,1}^{(0)} \right\rangle & \left\langle \varphi_{n,g}^{(0)} \middle| H_2 \middle| \varphi_{n,2}^{(0)} \right\rangle & \cdots & \left\langle \varphi_{n,g}^{(0)} \middle| H_2 \middle| \varphi_{n,g}^{(0)} \right\rangle - x \end{vmatrix} = 0 \tag{36}$$

Supposing the degeneracy is removed by the perturbation in the first order. Then, $c_{n,\alpha}{}^{\beta}$'s are determined and thus $\left|\phi_{n,\alpha}^{(0)}\right\rangle$ is also determined. $\left\langle\phi_{m,\beta}^{(0)}\right|$ is multiplied to both the sides of eq. (18) from the left.

$$\left\langle \phi_{m,\beta}^{(0)} \middle| H_0 \middle| \varphi_{n,\alpha}^{(01)} \right\rangle + \left\langle \phi_{m,\beta}^{(0)} \middle| H_2 \middle| \phi_{n,\alpha}^{(0)} \right\rangle = \varepsilon_n^{(0)} \left\langle \phi_{m,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(01)} \right\rangle + \varepsilon_{n,\alpha}^{(01)} \left\langle \phi_{m,\beta}^{(0)} \middle| \phi_{n,\alpha}^{(0)} \right\rangle \tag{37}$$

Using the orthonormality conditions (eq. (13)), eq. (37) is described as

$$\varepsilon_{n,\alpha}^{(01)} \delta_{nm} \delta_{\alpha\beta} = \left\langle \phi_{m,\beta}^{(0)} \middle| H_2 \middle| \phi_{n,\alpha}^{(0)} \right\rangle + \left(\varepsilon_m^{(0)} - \varepsilon_n^{(0)} \right) \left\langle \phi_{m,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(01)} \right\rangle \tag{38}$$

When $m \neq n$,

$$\left\langle \phi_{m,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(01)} \right\rangle = -\frac{\left\langle \phi_{m,\beta}^{(0)} \middle| H_2 \middle| \phi_{n,\alpha}^{(0)} \right\rangle}{\varepsilon_m^{(0)} - \varepsilon_n^{(0)}} \tag{39}$$

$\left|\Psi_{n,\alpha}\right\rangle$ is represented as follows by using the perturbation in the first order, with the completeness (eq. (14)):

$$\left|\Psi_{n,\alpha}\right\rangle = \left|\phi_{n,\alpha}^{(0)}\right\rangle + \lambda \left|\varphi_{n,\alpha}^{(10)}\right\rangle + \mu \left|\varphi_{n,\alpha}^{(01)}\right\rangle + \text{(higher order)}$$

$$= \left|\phi_{n,\alpha}^{(0)}\right\rangle + \lambda \sum_{m=0}^{\infty}\sum_{\beta=1}^{g_m} \left|\phi_{m,\beta}^{(0)}\right\rangle \left\langle\phi_{m,\beta}^{(0)}\middle|\varphi_{n,\alpha}^{(10)}\right\rangle + \mu \sum_{m=0}^{\infty}\sum_{\beta=1}^{g_m} \left|\phi_{m,\beta}^{(0)}\right\rangle \left\langle\phi_{m,\beta}^{(0)}\middle|\varphi_{n,\alpha}^{(01)}\right\rangle$$

$$+ \text{(higher order)}$$

$$= \left|\phi_{n,\alpha}^{(0)}\right\rangle + \lambda \left[\sum_{\beta=1}^{g_n} \left|\phi_{n,\beta}^{(0)}\right\rangle \left\langle\phi_{n,\beta}^{(0)}\middle|\varphi_{n,\alpha}^{(10)}\right\rangle + {\sum_{m=0}^{\infty}}' \sum_{\beta=1}^{g_m} \left|\phi_{m,\beta}^{(0)}\right\rangle \left\langle\phi_{m,\beta}^{(0)}\middle|\varphi_{n,\alpha}^{(10)}\right\rangle\right]$$

$$+ \mu \left[\sum_{\beta=1}^{g_n} \left|\phi_{n,\beta}^{(0)}\right\rangle \left\langle\phi_{n,\beta}^{(0)}\middle|\varphi_{n,\alpha}^{(01)}\right\rangle + {\sum_{m=0}^{\infty}}' \sum_{\beta=1}^{g_m} \left|\phi_{m,\beta}^{(0)}\right\rangle \left\langle\phi_{m,\beta}^{(0)}\middle|\varphi_{n,\alpha}^{(01)}\right\rangle\right] \tag{40}$$

$$+ \text{(higher order)}$$

Here, ${\sum_m}'$ means that the summation will be calculated except $m = n$. $\left\langle\phi_{m,\beta}^{(0)}\middle|\varphi_{n,\alpha}^{(10)}\right\rangle$ and $\left\langle\phi_{m,\beta}^{(0)}\middle|\varphi_{n,\alpha}^{(01)}\right\rangle$ in eq. (40) have been determined from eqs. (30) and (39), respectively, however, $\left\langle\phi_{n,\beta}^{(0)}\middle|\varphi_{n,\alpha}^{(10)}\right\rangle$ and $\left\langle\phi_{n,\beta}^{(0)}\middle|\varphi_{n,\alpha}^{(01)}\right\rangle$ will have been calculated the relationships in the second order described below.

λ^2: Multiplying $\left\langle\phi_{m,\beta}^{(0)}\right|$ the both hands of eq. (20) from the left yields

$$\left\langle\phi_{m,\beta}^{(0)}\middle|H_0\middle|\varphi_{n,\alpha}^{(20)}\right\rangle + \left\langle\phi_{m,\beta}^{(0)}\middle|H_1\middle|\varphi_{n,\alpha}^{(10)}\right\rangle$$
$$= \varepsilon_n^{(0)} \left\langle\phi_{m,\beta}^{(0)}\middle|\varphi_{n,\alpha}^{(20)}\right\rangle + \varepsilon_{n,\alpha}^{(10)} \left\langle\phi_{m,\beta}^{(0)}\middle|\varphi_{n,\alpha}^{(10)}\right\rangle + \varepsilon_{n,\alpha}^{(20)} \left\langle\phi_{m,\beta}^{(0)}\middle|\phi_{n,\alpha}^{(0)}\right\rangle \tag{41}$$

Using the zeroth-order relationship (eq. (12)) and the orthonormality condition (eq. (13)),

$$\varepsilon_{n,\alpha}^{(20)}\delta_{nm}\delta_{\alpha\beta} = \left\langle\phi_{m,\beta}^{(0)}\right|\left(H_1 - \varepsilon_{n,\alpha}^{(10)}\right)\left|\varphi_{n,\alpha}^{(10)}\right\rangle + \left(\varepsilon_m^{(0)} - \varepsilon_n^{(0)}\right)\left\langle\phi_{m,\beta}^{(0)}\middle|\varphi_{n,\alpha}^{(20)}\right\rangle \tag{42}$$

Supposing $m = n$, then

$$\varepsilon_{n,\alpha}^{(20)}\delta_{\alpha\beta} = \left\langle\phi_{n,\beta}^{(0)}\right|\left(H_1 - \varepsilon_{n,\alpha}^{(10)}\right)\left|\varphi_{n,\alpha}^{(10)}\right\rangle \tag{43}$$

Supposing $m = n$ in eq. (29), eq. (44) is obtained.

$$\varepsilon_{n,\alpha}^{(10)}\delta_{\alpha\beta} = \left\langle\phi_{n,\beta}^{(0)}\middle|H_1\middle|\phi_{n,\alpha}^{(0)}\right\rangle \tag{44}$$

Multiplying $\left\langle\phi_{n,\alpha}^{(0)}\right|$ to the both hands of eq. (44) from right and summation is taken for α, we have

$$\sum_{\alpha=1}^{g_n} \varepsilon_{n,\alpha}^{(10)}\delta_{\alpha\beta}\left\langle\phi_{n,\alpha}^{(0)}\right| = \sum_{\alpha=1}^{g_n} \left\langle\phi_{n,\beta}^{(0)}\middle|H_1\middle|\phi_{n,\alpha}^{(0)}\right\rangle\left\langle\phi_{n,\alpha}^{(0)}\right| \tag{45}$$

$$\Leftrightarrow \varepsilon_{n,\beta}^{(10)} \left\langle \phi_{n,\beta}^{(0)} \right| = \left\langle \phi_{n,\beta}^{(0)} \right| H_1 P_n \tag{46}$$

The right-hand side of eq. (43) can be written as

$$\left\langle \phi_{n,\beta}^{(0)} \right| \left(H_1 P_n - \varepsilon_{n,\alpha}^{(10)} \right) \left| \varphi_{n,\alpha}^{(10)} \right\rangle + \left\langle \phi_{n,\beta}^{(0)} \middle| H_1 (1 - P_n) \middle| \varphi_{n,\alpha}^{(10)} \right\rangle \tag{47}$$

And using eq. (46), the first term of eq. (47) is given as

$$\left(\varepsilon_{n,\beta}^{(10)} - \varepsilon_{n,\alpha}^{(10)} \right) \left\langle \phi_{n,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(10)} \right\rangle \tag{48}$$

Notice that

$$1 - P_n = \sum_{m=0}^{\infty}{}' P_m \tag{49}$$

and using eq. (30), the second term of eq. (47) is given as

$$\begin{aligned} \sum_{m=0}^{\infty}{}' \left\langle \phi_{n,\beta}^{(0)} \middle| H_1 P_m \middle| \varphi_{n,\alpha}^{(10)} \right\rangle &= \sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \left\langle \phi_{n,\beta}^{(0)} \middle| H_1 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \left\langle \phi_{m,\gamma}^{(0)} \middle| \varphi_{n,\alpha}^{(10)} \right\rangle \\ &= - \sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \frac{\left\langle \phi_{n,\beta}^{(0)} \middle| H_1 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \left\langle \phi_{m,\gamma}^{(0)} \middle| H_1 \middle| \phi_{n,\alpha}^{(0)} \right\rangle}{\varepsilon_m^{(0)} - \varepsilon_n^{(0)}} \end{aligned} \tag{50}$$

It is worth mentioning that the denominator is not zero because the summation for m eliminates $m = n$. Replacing the right-hand side of eq. (43) to eqs. (48) and (50) yields

$$\varepsilon_{n,\alpha}^{(20)} \delta_{\alpha\beta} = \left(\varepsilon_{n,\beta}^{(10)} - \varepsilon_{n,\alpha}^{(10)} \right) \left\langle \phi_{n,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(10)} \right\rangle - \sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \frac{\left\langle \phi_{n,\beta}^{(0)} \middle| H_1 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \left\langle \phi_{m,\gamma}^{(0)} \middle| H_1 \middle| \phi_{n,\alpha}^{(0)} \right\rangle}{\varepsilon_m^{(0)} - \varepsilon_n^{(0)}} \tag{51}$$

$\varepsilon_{n,\alpha}^{(20)}$ can be obtained supposing $\alpha = \beta$ in eq. (51):

$$\varepsilon_{n,\alpha}^{(20)} = \sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \frac{\left| \left\langle \phi_{n,\alpha}^{(0)} \middle| H_1 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \right|^2}{\varepsilon_n^{(0)} - \varepsilon_m^{(0)}} \tag{52}$$

Supposing $\alpha \neq \beta$ in eq. (51) yields

$$\left\langle \phi_{n,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(10)} \right\rangle = \frac{1}{\varepsilon_{n,\beta}^{(10)} - \varepsilon_{n,\alpha}^{(10)}} \sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \frac{\left\langle \phi_{n,\beta}^{(0)} \middle| H_1 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \left\langle \phi_{m,\gamma}^{(0)} \middle| H_1 \middle| \phi_{n,\alpha}^{(0)} \right\rangle}{\varepsilon_m^{(0)} - \varepsilon_n^{(0)}} \tag{53}$$

Equation (53) is the one of the coefficients of the perturbed wavefunctions in the first order, which have not been determined in eq. (40).

μ^2: Multiplying $\left\langle \phi_{m,\beta}^{(0)} \right|$ to the both hands of eq. (22) from the left yields

$$\left\langle \phi_{m,\beta}^{(0)} \middle| H_0 \middle| \varphi_{n,\alpha}^{(02)} \right\rangle + \left\langle \phi_{m,\beta}^{(0)} \middle| H_2 \middle| \varphi_{n,\alpha}^{(01)} \right\rangle$$
$$= \varepsilon_n^{(0)} \left\langle \phi_{m,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(02)} \right\rangle + \varepsilon_{n,\alpha}^{(01)} \left\langle \phi_{m,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(01)} \right\rangle + \varepsilon_{n,\alpha}^{(02)} \left\langle \phi_{m,\beta}^{(0)} \middle| \phi_{n,\alpha}^{(0)} \right\rangle \tag{54}$$

Using the zeroth-order relationship eq. (12) and the orthonormality condition eq. (13), eq. (54) is

$$\varepsilon_{n,\alpha}^{(02)} \delta_{nm} \delta_{\alpha\beta} = \left\langle \phi_{m,\beta}^{(0)} \middle| \left(H_2 - \varepsilon_{n,\alpha}^{(01)} \right) \middle| \varphi_{n,\alpha}^{(01)} \right\rangle + \left(\varepsilon_m^{(0)} - \varepsilon_n^{(0)} \right) \left\langle \phi_{m,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(02)} \right\rangle \tag{55}$$

Supposing $m = n$,

$$\varepsilon_{n,\alpha}^{(02)} \delta_{\alpha\beta} = \left\langle \phi_{n,\beta}^{(0)} \middle| \left(H_2 - \varepsilon_{n,\alpha}^{(01)} \right) \middle| \varphi_{n,\alpha}^{(01)} \right\rangle \tag{56}$$

An equation is obtained supposing $m = n$ in eq. (38):

$$\varepsilon_{n,\alpha}^{(01)} \delta_{\alpha\beta} = \left\langle \phi_{n,\beta}^{(0)} \middle| H_2 \middle| \phi_{n,\alpha}^{(0)} \right\rangle \tag{57}$$

Multiplying $\left\langle \phi_{n,\alpha}^{(0)} \right|$ to the both hands of eq. (57) from right and summation is taken for α, we have

$$\varepsilon_{n,\beta}^{(01)} \left\langle \phi_{n,\beta}^{(0)} \right| = \left\langle \phi_{n,\beta}^{(0)} \right| H_2 P_n \tag{58}$$

The right-hand side of eq. (56) can be rewritten as

$$\left\langle \phi_{n,\beta}^{(0)} \middle| \left(H_2 P_n - \varepsilon_{n,\alpha}^{(01)} \right) \middle| \varphi_{n,\alpha}^{(01)} \right\rangle + \left\langle \phi_{n,\beta}^{(0)} \middle| H_2 (1 - P_n) \middle| \varphi_{n,\alpha}^{(01)} \right\rangle \tag{59}$$

Using eq. (58), the first term of eq. (59) is

$$\left(\varepsilon_{n,\beta}^{(01)} - \varepsilon_{n,\alpha}^{(01)} \right) \left\langle \phi_{n,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(01)} \right\rangle \tag{60}$$

Using eq. (39) with attention to eq. (49),

$$\sum_{m=0}^{\infty}{}' \left\langle \phi_{n,\beta}^{(0)} \middle| H_2 P_m \middle| \varphi_{n,\alpha}^{(01)} \right\rangle = \sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \left\langle \phi_{n,\beta}^{(0)} \middle| H_2 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \left\langle \phi_{m,\gamma}^{(0)} \middle| \varphi_{n,\alpha}^{(01)} \right\rangle$$
$$= - \sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \frac{\left\langle \phi_{n,\beta}^{(0)} \middle| H_2 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \left\langle \phi_{m,\gamma}^{(0)} \middle| H_2 \middle| \phi_{n,\alpha}^{(0)} \right\rangle}{\varepsilon_m^{(0)} - \varepsilon_n^{(0)}} \tag{61}$$

Substituting the right-hand side of eq. (56) to eqs. (60) and (61) yields

$$\varepsilon_{n,\alpha}^{(02)} \delta_{\beta\alpha} = \left(\varepsilon_{n,\beta}^{(01)} - \varepsilon_{n,\alpha}^{(01)} \right) \left\langle \phi_{n,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(01)} \right\rangle - \sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \frac{\left\langle \phi_{n,\beta}^{(0)} \middle| H_2 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \left\langle \phi_{m,\gamma}^{(0)} \middle| H_2 \middle| \phi_{n,\alpha}^{(0)} \right\rangle}{\varepsilon_m^{(0)} - \varepsilon_n^{(0)}} \tag{62}$$

We obtain $\varepsilon_{n,\alpha}^{(02)}$ supposing $\alpha = \beta$ in eq. (62).

$$\varepsilon_{n,\alpha}^{(02)} = \sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \frac{\left| \left\langle \phi_{n,\alpha}^{(0)} \middle| H_2 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \right|^2}{\varepsilon_n^{(0)} - \varepsilon_m^{(0)}} \tag{63}$$

Supposing $\alpha \neq \beta$ in eq. (62) yields

$$\left\langle \phi_{n,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(01)} \right\rangle = \frac{1}{\varepsilon_{n,\beta}^{(01)} - \varepsilon_{n,\alpha}^{(01)}} \sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \frac{\left\langle \phi_{n,\beta}^{(0)} \middle| H_2 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \left\langle \phi_{m,\gamma}^{(0)} \middle| H_2 \middle| \phi_{n,\alpha}^{(0)} \right\rangle}{\varepsilon_m^{(0)} - \varepsilon_n^{(0)}} \tag{64}$$

Equation (64) is also one of the coefficients of the perturbed wavefunctions in the first order, which have not been determined in eq. (40). Using the completeness (eq. (14)), the wavefunction $\left|\Psi_{n,\alpha}\right\rangle$ is in first order:

$$\begin{aligned}
\left|\Psi_{n,\alpha}\right\rangle &= \left|\phi_{n,\alpha}^{(0)}\right\rangle \left(1 + \lambda \left|\phi_{n,\alpha}^{(0)}\right\rangle \left\langle \phi_{n,\alpha}^{(0)} \middle| \varphi_{n,\alpha}^{(10)} \right\rangle + \mu \left|\phi_{n,\alpha}^{(0)}\right\rangle \left\langle \phi_{n,\alpha}^{(0)} \middle| \varphi_{n,\alpha}^{(01)} \right\rangle \right) \\
&\quad + \lambda \left[\sum_{\beta=1}^{g_n}{}' \left|\phi_{n,\beta}^{(0)}\right\rangle \left\langle \phi_{n,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(10)} \right\rangle + \sum_{m=0}^{\infty}{}' \sum_{\beta=1}^{g_m} \left|\phi_{m,\beta}^{(0)}\right\rangle \left\langle \phi_{m,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(10)} \right\rangle \right] \\
&\quad + \mu \left[\sum_{\beta=1}^{g_n}{}' \left|\phi_{n,\beta}^{(0)}\right\rangle \left\langle \phi_{n,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(01)} \right\rangle + \sum_{m=0}^{\infty}{}' \sum_{\beta=1}^{g_m} \left|\phi_{m,\beta}^{(0)}\right\rangle \left\langle \phi_{m,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(01)} \right\rangle \right] \\
&\quad + \text{(higher order)} \\
&= \exp\left(\lambda \left\langle \phi_{n,\alpha}^{(0)} \middle| \varphi_{n,\alpha}^{(10)} \right\rangle + \mu \left\langle \phi_{n,\alpha}^{(0)} \middle| \varphi_{n,\alpha}^{(01)} \right\rangle \right) \Bigg\{ \left|\phi_{n,\alpha}^{(0)}\right\rangle \\
&\quad + \lambda \left[\sum_{\beta=1}^{g_n}{}' \left|\phi_{n,\beta}^{(0)}\right\rangle \left\langle \phi_{n,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(10)} \right\rangle + \sum_{m=0}^{\infty}{}' \sum_{\beta=1}^{g_m} \left|\phi_{m,\beta}^{(0)}\right\rangle \left\langle \phi_{m,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(10)} \right\rangle \right] \\
&\quad + \mu \left[\sum_{\beta=1}^{g_n}{}' \left|\phi_{n,\beta}^{(0)}\right\rangle \left\langle \phi_{n,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(01)} \right\rangle + \sum_{m=0}^{\infty}{}' \sum_{\beta=1}^{g_m} \left|\phi_{m,\beta}^{(0)}\right\rangle \left\langle \phi_{m,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(01)} \right\rangle \right] \Bigg\} \\
&\quad + \text{(higher order)}
\end{aligned} \tag{65}$$

where the prime (′) on the summation for β means that the summation will be calculated except $\beta = \alpha$. Applying the normality for the wavefunction in the first order $\left\langle \Psi_{n,\alpha} \middle| \Psi_{n,\alpha} \right\rangle = 1 + \text{(higher order)}$, we have

$$\left\langle \phi_{n,\alpha}^{(0)} \middle| \varphi_{n,\alpha}^{(10)} \right\rangle = 0 \tag{66}$$

$$\left\langle \phi_{n,\alpha}^{(0)} \middle| \varphi_{n,\alpha}^{(01)} \right\rangle = 0 \tag{67}$$

Therefore, the normalized wavefunction in the first order can be obtained by using eqs. (30), (39), (53), (60), (66), and (67):

$$|\Psi_{n,\alpha}\rangle = \left|\phi_{n,\alpha}^{(0)}\right\rangle + \lambda\left[\sum_{\beta=1}^{g_n}{}'\left|\phi_{n,\beta}^{(0)}\right\rangle\frac{1}{\varepsilon_{n,\beta}^{(10)}-\varepsilon_{n,\alpha}^{(10)}}\sum_{m=0}^{\infty}{}'\sum_{\gamma=1}^{g_n}\frac{\left\langle\phi_{n,\beta}^{(0)}\middle|H_1\middle|\phi_{m,\gamma}^{(0)}\right\rangle\left\langle\phi_{m,\gamma}^{(0)}\middle|H_1\middle|\phi_{n,\alpha}^{(0)}\right\rangle}{\varepsilon_m^{(0)}-\varepsilon_n^{(0)}}\right.$$
$$\left.-\sum_{m=0}^{\infty}{}'\sum_{\beta=1}^{g_m}\left|\phi_{m,\beta}^{(0)}\right\rangle\frac{\left\langle\phi_{m,\beta}^{(0)}\middle|H_1\middle|\phi_{n,\alpha}^{(0)}\right\rangle}{\varepsilon_m^{(0)}-\varepsilon_n^{(0)}}\right]$$
$$+\mu\left[\sum_{\beta=1}^{g_n}{}'\left|\phi_{n,\beta}^{(0)}\right\rangle\frac{1}{\varepsilon_{n,\beta}^{(01)}-\varepsilon_{n,\alpha}^{(01)}}\sum_{m=0}^{\infty}{}'\sum_{\gamma=1}^{g_n}\frac{\left\langle\phi_{n,\beta}^{(0)}\middle|H_2\middle|\phi_{m,\gamma}^{(0)}\right\rangle\left\langle\phi_{m,\gamma}^{(0)}\middle|H_2\middle|\phi_{n,\alpha}^{(0)}\right\rangle}{\varepsilon_m^{(0)}-\varepsilon_n^{(0)}}\right.$$
$$\left.-\sum_{m=0}^{\infty}{}'\sum_{\beta=1}^{g_m}\left|\phi_{m,\beta}^{(0)}\right\rangle\frac{\left\langle\phi_{m,\beta}^{(0)}\middle|H_2\middle|\phi_{n,\alpha}^{(0)}\right\rangle}{\varepsilon_m^{(0)}-\varepsilon_n^{(0)}}\right] + \text{(higher order)} \tag{68}$$

$\lambda\mu$: Multiplying $\left\langle\phi_{m,\beta}^{(0)}\right|$ to the both hands of eq. (21) from the left yields

$$\left\langle\phi_{m,\beta}^{(0)}\middle|H_0\middle|\varphi_{n,\alpha}^{(11)}\right\rangle + \left\langle\phi_{m,\beta}^{(0)}\middle|H_1\middle|\varphi_{n,\alpha}^{(01)}\right\rangle + \left\langle\phi_{m,\beta}^{(0)}\middle|H_2\middle|\varphi_{n,\alpha}^{(10)}\right\rangle$$
$$= \varepsilon_n^{(0)}\left\langle\phi_{m,\beta}^{(0)}\middle|\varphi_{n,\alpha}^{(11)}\right\rangle + \varepsilon_{n,\alpha}^{(10)}\left\langle\phi_{m,\beta}^{(0)}\middle|\varphi_{n,\alpha}^{(01)}\right\rangle + \varepsilon_{n,\alpha}^{(01)}\left\langle\phi_{m,\beta}^{(0)}\middle|\varphi_{n,\alpha}^{(10)}\right\rangle \tag{69}$$
$$+ \varepsilon_{n,\alpha}^{(11)}\left\langle\phi_{m,\beta}^{(0)}\middle|\phi_{n,\alpha}^{(0)}\right\rangle$$

Using the zeroth-order relationship eq. (12) and the orthonormality conditions, eq. (13), eq. (69) is

$$\varepsilon_{n,\alpha}^{(11)}\delta_{nm}\delta_{\alpha\beta} = \left\langle\phi_{m,\beta}^{(0)}\middle|\left(H_1-\varepsilon_{n,\alpha}^{(10)}\right)\middle|\varphi_{n,\alpha}^{(01)}\right\rangle + \left\langle\phi_{m,\beta}^{(0)}\middle|\left(H_2-\varepsilon_{n,\alpha}^{(01)}\right)\middle|\varphi_{n,\alpha}^{(10)}\right\rangle$$
$$+\left(\varepsilon_m^{(0)}-\varepsilon_n^{(0)}\right)\left\langle\phi_{m,\beta}^{(0)}\middle|\varphi_{n,\alpha}^{(11)}\right\rangle \tag{70}$$

Supposing $m = n$, eq. (70) is described as

$$\varepsilon_{n,\alpha}^{(11)}\delta_{\beta\alpha} = \left\langle\phi_{n,\beta}^{(0)}\middle|\left(H_1-\varepsilon_{n,\alpha}^{(10)}\right)\middle|\varphi_{n,\alpha}^{(01)}\right\rangle + \left\langle\phi_{n,\beta}^{(0)}\middle|\left(H_2-\varepsilon_{n,\alpha}^{(01)}\right)\middle|\varphi_{n,\alpha}^{(10)}\right\rangle \tag{71}$$

The right-hand side of eq. (71) can be divided as follows:

$$\left\langle\phi_{n,\beta}^{(0)}\middle|\left(H_1-\varepsilon_{n,\alpha}^{(10)}\right)\middle|\varphi_{n,\alpha}^{(01)}\right\rangle = \left\langle\phi_{n,\beta}^{(0)}\middle|\left(H_1P_n-\varepsilon_{n,\alpha}^{(10)}\right)\middle|\varphi_{n,\alpha}^{(01)}\right\rangle + \left\langle\phi_{n,\beta}^{(0)}\middle|H_1(1-P_n)\middle|\varphi_{n,\alpha}^{(01)}\right\rangle \tag{72}$$

$$\left\langle\phi_{n,\beta}^{(0)}\middle|\left(H_2-\varepsilon_{n,\alpha}^{(01)}\right)\middle|\varphi_{n,\alpha}^{(10)}\right\rangle = \left\langle\phi_{n,\beta}^{(0)}\middle|\left(H_2P_n-\varepsilon_{n,\alpha}^{(01)}\right)\middle|\varphi_{n,\alpha}^{(10)}\right\rangle + \left\langle\phi_{n,\beta}^{(0)}\middle|H_2(1-P_n)\middle|\varphi_{n,\alpha}^{(10)}\right\rangle \tag{73}$$

Using eqs. (46) and (48), the first term of the right-hand side of eqs. (72) and (73) is

$$\left\langle\phi_{n,\beta}^{(0)}\middle|\left(H_1P_n-\varepsilon_{n,\alpha}^{(10)}\right)\middle|\varphi_{n,\alpha}^{(01)}\right\rangle = \left(\varepsilon_{n,\beta}^{(10)}-\varepsilon_{n,\alpha}^{(10)}\right)\left\langle\phi_{n,\beta}^{(0)}\middle|\varphi_{n,\alpha}^{(01)}\right\rangle \tag{74}$$

$$\left\langle\phi_{n,\beta}^{(0)}\middle|\left(H_2P_n-\varepsilon_{n,\alpha}^{(01)}\right)\middle|\varphi_{n,\alpha}^{(10)}\right\rangle = \left(\varepsilon_{n,\beta}^{(01)}-\varepsilon_{n,\alpha}^{(01)}\right)\left\langle\phi_{n,\beta}^{(0)}\middle|\varphi_{n,\alpha}^{(10)}\right\rangle \tag{75}$$

Using eqs. (30) and (39) with attention to eq. (49), the second term in the right-hand side of eqs. (72) and

(73) are

$$\left\langle \phi_{n,\beta}^{(0)} \middle| H_1 (1-P_n) \middle| \varphi_{n,\alpha}^{(01)} \right\rangle = \sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \left\langle \phi_{n,\beta}^{(0)} \middle| H_1 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \left\langle \phi_{m,\gamma}^{(0)} \middle| \varphi_{n,\alpha}^{(01)} \right\rangle$$
$$= -\sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \frac{\left\langle \phi_{n,\beta}^{(0)} \middle| H_1 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \left\langle \phi_{m,\gamma}^{(0)} \middle| H_2 \middle| \phi_{n,\alpha}^{(0)} \right\rangle}{\varepsilon_m^{(0)} - \varepsilon_n^{(0)}} \tag{76}$$

$$\left\langle \phi_{n,\beta}^{(0)} \middle| H_2 (1-P_n) \middle| \varphi_{n,\alpha}^{(10)} \right\rangle = \sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \left\langle \phi_{n,\beta}^{(0)} \middle| H_2 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \left\langle \phi_{m,\gamma}^{(0)} \middle| \varphi_{n,\alpha}^{(10)} \right\rangle$$
$$= -\sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \frac{\left\langle \phi_{n,\beta}^{(0)} \middle| H_2 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \left\langle \phi_{m,\gamma}^{(0)} \middle| H_1 \middle| \phi_{n,\alpha}^{(0)} \right\rangle}{\varepsilon_m^{(0)} - \varepsilon_n^{(0)}} \tag{77}$$

Replacing the terms of eq. (71) to eqs. (74)–(77) yields

$$\varepsilon_{n,\alpha}^{(11)} \delta_{\beta\alpha} = \left(\varepsilon_{n,\beta}^{(10)} - \varepsilon_{n,\alpha}^{(10)} \right) \left\langle \phi_{n,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(01)} \right\rangle - \sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \frac{\left\langle \phi_{n,\beta}^{(0)} \middle| H_1 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \left\langle \phi_{m,\gamma}^{(0)} \middle| H_2 \middle| \phi_{n,\alpha}^{(0)} \right\rangle}{\varepsilon_m^{(0)} - \varepsilon_n^{(0)}}$$
$$+ \left(\varepsilon_{n,\beta}^{(01)} - \varepsilon_{n,\alpha}^{(01)} \right) \left\langle \phi_{n,\beta}^{(0)} \middle| \varphi_{n,\alpha}^{(10)} \right\rangle - \sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \frac{\left\langle \phi_{n,\beta}^{(0)} \middle| H_2 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \left\langle \phi_{m,\gamma}^{(0)} \middle| H_1 \middle| \phi_{n,\alpha}^{(0)} \right\rangle}{\varepsilon_m^{(0)} - \varepsilon_n^{(0)}} \tag{78}$$

$\varepsilon_{n,\alpha}^{(11)}$ can be obtained supposing $\alpha = \beta$ in eq. (78).

$$\varepsilon_{n,\alpha}^{(11)} = \sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \frac{\left\langle \phi_{n,\alpha}^{(0)} \middle| H_1 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \left\langle \phi_{m,\gamma}^{(0)} \middle| H_2 \middle| \phi_{n,\alpha}^{(0)} \right\rangle}{\varepsilon_n^{(0)} - \varepsilon_m^{(0)}} + \sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \frac{\left\langle \phi_{n,\alpha}^{(0)} \middle| H_2 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \left\langle \phi_{m,\gamma}^{(0)} \middle| H_1 \middle| \phi_{n,\alpha}^{(0)} \right\rangle}{\varepsilon_n^{(0)} - \varepsilon_m^{(0)}} \tag{79}$$

Eventually, the perturbed energies can be obtained in the second order with substituting eqs. (27), (52), (63) and (79) to eq. (14).

$$E_{n,\alpha} = \varepsilon_n^{(0)} + \left\langle \varphi_{n,\alpha}^{(0)} \middle| H_1 \middle| \varphi_{n,\alpha}^{(0)} \right\rangle + \varepsilon_{n,\alpha}^{(01)} + \sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \frac{\left| \left\langle \phi_{n,\alpha}^{(0)} \middle| H_1 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \right|^2}{\varepsilon_n^{(0)} - \varepsilon_m^{(0)}}$$
$$+ \sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \frac{\left\langle \phi_{n,\alpha}^{(0)} \middle| H_1 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \left\langle \phi_{m,\gamma}^{(0)} \middle| H_2 \middle| \phi_{n,\alpha}^{(0)} \right\rangle}{\varepsilon_n^{(0)} - \varepsilon_m^{(0)}} \tag{80}$$
$$+ \sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \frac{\left\langle \phi_{n,\alpha}^{(0)} \middle| H_2 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \left\langle \phi_{m,\gamma}^{(0)} \middle| H_1 \middle| \phi_{n,\alpha}^{(0)} \right\rangle}{\varepsilon_n^{(0)} - \varepsilon_m^{(0)}} + \sum_{m=0}^{\infty}{}' \sum_{\gamma=1}^{g_m} \frac{\left| \left\langle \phi_{n,\alpha}^{(0)} \middle| H_2 \middle| \phi_{m,\gamma}^{(0)} \right\rangle \right|^2}{\varepsilon_n^{(0)} - \varepsilon_m^{(0)}}$$
$$+ \text{(higher order)}$$

here, $\varepsilon_{n,\alpha}^{(01)}$ is an eigenvalue of the matrix given below

$$
\begin{pmatrix}
\left\langle \varphi_{n,1}^{(0)} \middle| H_2 \middle| \varphi_{n,1}^{(0)} \right\rangle & \left\langle \varphi_{n,1}^{(0)} \middle| H_2 \middle| \varphi_{n,2}^{(0)} \right\rangle & \cdots & \left\langle \varphi_{n,1}^{(0)} \middle| H_2 \middle| \varphi_{n,g}^{(0)} \right\rangle \\
\left\langle \varphi_{n,2}^{(0)} \middle| H_2 \middle| \varphi_{n,1}^{(0)} \right\rangle & \left\langle \varphi_{n,2}^{(0)} \middle| H_2 \middle| \varphi_{n,2}^{(0)} \right\rangle & \cdots & \left\langle \varphi_{n,2}^{(0)} \middle| H_2 \middle| \varphi_{n,g}^{(0)} \right\rangle \\
\vdots & \vdots & \ddots & \vdots \\
\left\langle \varphi_{n,g}^{(0)} \middle| H_2 \middle| \varphi_{n,1}^{(0)} \right\rangle & \left\langle \varphi_{n,g}^{(0)} \middle| H_2 \middle| \varphi_{n,2}^{(0)} \right\rangle & \cdots & \left\langle \varphi_{n,g}^{(0)} \middle| H_2 \middle| \varphi_{n,g}^{(0)} \right\rangle
\end{pmatrix} \tag{81}
$$

Exact Analytical Energies for S = 3/2

The full spin-Hamiltonian are composed of the ZFS, the electron-Zeeman and the hyperfine structure terms in the following:

$$H = \boldsymbol{S} \cdot \mathbf{D} \cdot \boldsymbol{S} + \beta \boldsymbol{S} \cdot \mathbf{g} \cdot \boldsymbol{B} + \boldsymbol{S} \cdot \mathbf{A} \cdot \boldsymbol{I}$$

where **D**, **g** and **A** are the zero-field, **g-** and hyperfine tensors, respectively, S and I are the electron and the nuclear spin operator, respectively, and β is the Bohr magneton. Through the discussion, the **D-**, **g-** and **A-**tensors are assumed collinear. Additionally, the external magnetic field is parallel to the z-axis of the principal coordinate system. Under these assumptions, the spin-Hamiltonian H can be represented as

$$H = D\left[{S_z}^2 - \frac{1}{3}S(S+1)\right] + E\left({S_x}^2 - {S_y}^2\right) + \beta S_z g_z B + S_x A_x I_x + S_y A_y I_y + S_z A_z I_z$$

where D and E are the ZFS parameters. When the external magnetic field is parallel to the x- or y-axis of the principal coordinate system, the corresponding energies can be obtained by the cyclic permutation relationships for D and E as given in the following [1-4].

$$D \to \frac{1}{2}(3E - D), E \to -\frac{1}{2}(E + D) \text{ if } \boldsymbol{B} \parallel x$$

$$D \to -\frac{1}{2}(3E + D), E \to \frac{1}{2}(E + D) \text{ if } \boldsymbol{B} \parallel y$$

1. I = 1/2 case

a) Exact diagonalization treatment

The full spin Hamiltonian for the spin quartet state with one I = 1/2 nucleus and the static magnetic field along the z-axis can be represented as a matrix in terms of the {|M_S, M_I>} basis set as follows:

$$H_{\text{full}}^{S=\frac{3}{2}, I=\frac{1}{2}} =$$

	|+3/2, +1/2>	|+3/2, –1/2>	|+1/2, +1/2>	|+1/2, –1/2>	|–1/2, +1/2>	|–1/2, +1/2>	|–3/2, +1/2>	|–3/2, +1/2>
<+3/2, +1/2|	$D + \frac{3}{2}g_z\beta B + \frac{3}{4}A_z$	0	0	$\frac{\sqrt{3}}{4}(A_x - A_y)$	$\sqrt{3}E$	0	0	0
<+3/2, –1/2|	0	$D + \frac{3}{2}g_z\beta B - \frac{3}{4}A_z$	$\frac{\sqrt{3}}{4}(A_x + A_y)$	0	0	$\sqrt{3}E$	0	0
<+1/2, +1/2|	0	$\frac{\sqrt{3}}{4}(A_x + A_y)$	$-D + \frac{1}{2}g_z\beta B + \frac{1}{4}A_z$	0	0	$\frac{1}{2}(A_x - A_y)$	$\sqrt{3}E$	0

<+1/2, –1/2|	$\frac{\sqrt{3}}{4}(A_x - A_y)$	0	0	$-D + \frac{1}{2}g_z\beta B$ $- \frac{1}{4}A_z$	$\frac{1}{2}(A_x + A_y)$	0	0	$\sqrt{3}E$
<–1/2, +1/2|	$\sqrt{3}E$	0	0	$\frac{1}{2}(A_x + A_y)$	$-D - \frac{1}{2}g_z\beta B$ $- \frac{1}{4}A_z$	0	0	$\frac{\sqrt{3}}{4}(A_x - A_y)$
<–1/2, +1/2|	0	$\sqrt{3}E$	$\frac{1}{2}(A_x - A_y)$	0	0	$-D - \frac{1}{2}g_z\beta B$ $+ \frac{1}{4}A_z$	$\frac{\sqrt{3}}{4}(A_x + A_y)$	0
<–3/2, +1/2|	0	0	$\sqrt{3}E$	0	0	$\frac{\sqrt{3}}{4}(A_x + A_y)$	$D - \frac{3}{2}g_z\beta B$ $- \frac{3}{4}A_z$	0
<–3/2, –1/2|	0	0	0	$\sqrt{3}E$	$\frac{\sqrt{3}}{4}(A_x - A_y)$	0	0	$D - \frac{3}{2}g_z\beta B$ $+ \frac{3}{4}A_z$

The matrix can be divided into two 4 × 4 matrices. Exploiting two particular basis sets of {|+3/2, +1/2>, |+1/2, –1/2>, |–1/2, +1/2>, |–3/2, –1/2>} ($M_S + M_I = \pm 2$ or 0) and {|+3/2, –1/2>, |+1/2, +1/2>, |–1/2, –1/2>, |–3/2, +1/2>} ($M_S + M_I = \pm 1$) give the two 4 × 4 matrices, both of which can be analytically and exactly solved. The basis sets are spin-conjugate each other, which are intrinsic to the properties of half-integer spins. The former basis set gives the following 4 × 4 matrix denoted by "full, 1":

$$H_{\mathrm{full},1}^{S=\frac{3}{2},I=\frac{1}{2}} = \begin{pmatrix} D + \frac{3}{2}g_z\beta B + \frac{3}{4}A_z & \frac{\sqrt{3}}{4}(A_x - A_y) & \sqrt{3}E & 0 \\ \frac{\sqrt{3}}{4}(A_x - A_y) & -D + \frac{1}{2}g_z\beta B - \frac{1}{4}A_z & \frac{1}{2}(A_x + A_y) & \sqrt{3}E \\ \sqrt{3}E & \frac{1}{2}(A_x + A_y) & -D - \frac{1}{2}g_z\beta B - \frac{1}{4}A_z & \frac{\sqrt{3}}{4}(A_x - A_y) \\ 0 & \sqrt{3}E & \frac{\sqrt{3}}{4}(A_x - A_y) & D - \frac{3}{2}g_z\beta B + \frac{3}{4}A_z \end{pmatrix}$$

The corresponding secular equation is given as

$$x^4 + a_3x^3 + a_2x^2 + a_1x + a_0 = 0$$

with

$$a_3 = -A_z$$

$$a_2 = -2(D^2 + 3E^2) - \frac{5}{2}(g_z\beta B)^2 - \frac{5}{8}{A_x}^2 + \frac{1}{4}A_xA_y - \frac{5}{8}{A_y}^2 - 2DA_z - \frac{1}{8}{A_z}^2$$

$$a_1 = -4D(g_z\beta B)^2 + \frac{1}{2}D{A_x}^2 - \frac{3}{2}E{A_x}^2 + DA_xA_y + \frac{1}{2}D{A_y}^2 + \frac{3}{2}E{A_y}^2 + D^2A_z + 3E^2A_z - \frac{3}{4}A_z(g_z\beta B)^2$$
$$+ \frac{9}{16}{A_x}^2A_z + \frac{3}{8}A_xA_yA_z + \frac{9}{16}{A_y}^2A_z + D{A_z}^2 + \frac{3}{16}{A_z}^3$$

$$a_0 = (D^2+3E^2)^2 - \frac{5}{2}D^2(g_z\beta B)^2 + \frac{9}{2}E^2(g_z\beta B)^2 + \frac{9}{16}(g_z\beta B)^4 + \frac{1}{8}D^2{A_x}^2 + \frac{3}{2}DE{A_x}^2 - \frac{9}{8}E^2{A_x}^2$$

$$+\frac{9}{32}{A_x}^2(g_z\beta B)^2 + \frac{9}{256}{A_x}^4 - \frac{5}{4}D^2A_xA_y + \frac{9}{4}E^2A_xA_y + \frac{27}{16}A_xA_y(g_z\beta B)^2 - \frac{9}{64}{A_x}^3A_y$$

$$+\frac{1}{8}D^2{A_y}^2 - \frac{3}{2}DE{A_y}^2 - \frac{9}{8}E^2{A_y}^2 + \frac{9}{32}{A_y}^2(g_z\beta B)^2 + \frac{27}{128}{A_x}^2{A_y}^2 - \frac{9}{64}A_x{A_y}^3$$

$$+\frac{9}{256}{A_y}^4 + 2D^3A_z + 6DE^2A_z - \frac{3}{2}D(g_z\beta B)^2A_z + \frac{9}{8}E{A_x}^2A_z - \frac{3}{2}DA_xA_yA_z$$

$$-\frac{9}{8}E{A_y}^2A_z + \frac{11}{8}D^2{A_z}^2 + \frac{9}{8}E^2{A_z}^2 - \frac{9}{32}{A_z}^2(g_z\beta B)^2 - \frac{9}{128}{A_x}^2{A_z}^2 - \frac{27}{64}A_xA_y{A_z}^2$$

$$-\frac{9}{128}{A_y}^2{A_z}^2 + \frac{3}{8}D{A_z}^3 + \frac{9}{256}{A_z}^4$$

In order to eliminate the x^3 term as usual, replacing x to $x + A_z/4$ yields

$$x^4 + p_0x^2 + q_0x + r_0 = 0 \qquad (*)$$

with

$$p_0 = -2(D^2+3E^2) - \frac{5}{2}(g_z\beta B)^2 - \frac{5}{8}{A_x}^2 + \frac{1}{4}A_xA_y - \frac{5}{8}{A_y}^2 - 2DA_z - \frac{1}{2}{A_z}^2$$

$$q_0 = -4D(g_z\beta B)^2 + \frac{1}{2}D{A_x}^2 - \frac{3}{2}E{A_x}^2 + DA_xA_y + \frac{1}{2}D{A_y}^2 + \frac{3}{2}E{A_y}^2 - 2A_z(g_z\beta B)^2 + \frac{1}{4}{A_x}^2A_z$$

$$+\frac{1}{2}A_xA_yA_z + \frac{1}{4}{A_y}^2A_z$$

$$r_0 = (D^2+3E^2)^2 - \frac{5}{2}D^2(g_z\beta B)^2 + \frac{9}{2}E^2(g_z\beta B)^2 + \frac{9}{16}(g_z\beta B)^4 + \frac{1}{8}D^2{A_x}^2 + \frac{3}{2}DE{A_x}^2 - \frac{9}{8}E^2{A_x}^2$$

$$+\frac{9}{32}{A_x}^2(g_z\beta B)^2 + \frac{9}{256}{A_x}^4 - \frac{5}{4}D^2A_xA_y + \frac{9}{4}E^2A_xA_y + \frac{27}{16}A_xA_y(g_z\beta B)^2 - \frac{9}{64}{A_x}^3A_y$$

$$+\frac{1}{8}D^2{A_y}^2 - \frac{3}{2}DE{A_y}^2 - \frac{9}{8}E^2{A_y}^2 + \frac{9}{32}{A_y}^2(g_z\beta B)^2 + \frac{27}{128}{A_x}^2{A_y}^2 - \frac{9}{64}A_x{A_y}^3$$

$$+\frac{9}{256}{A_y}^4 + 2D^3A_z + 6DE^2A_z - \frac{5}{2}D(g_z\beta B)^2A_z + \frac{1}{8}D{A_x}^2A_z + \frac{3}{4}E{A_x}^2A_z$$

$$-\frac{5}{4}DA_xA_yA_z + \frac{1}{8}D{A_y}^2A_z - \frac{3}{4}E{A_y}^2A_z + \frac{3}{2}D^2{A_z}^2 + \frac{3}{2}E^2{A_z}^2 - \frac{5}{8}{A_z}^2(g_z\beta B)^2$$

$$+\frac{1}{32}{A_x}^2{A_z}^2 - \frac{5}{16}A_xA_y{A_z}^2 + \frac{1}{32}{A_y}^2{A_z}^2 + \frac{1}{2}D{A_z}^3 + \frac{1}{16}{A_z}^4$$

The quartic equation (*) can be rewritten by using a parameter u ($\neq 0$) as

$$\left(x^2 + \frac{p_0+u}{2}\right)^2 - u\left(x - \frac{q_0}{2u}\right)^2 = 0$$

Comparing the coefficients of the two quartic equations gives us the resolvent cubic equation about u.

$$\frac{{p_0}^2}{4} - \frac{{q_0}^2}{4u} + \frac{p_0u}{2} + \frac{u^2}{4} = r_0$$

$$u^3 + 2p_0u^2 + ({p_0}^2 - 4r_0)u = q^2$$

In order to eliminate the u^2 term, replacing u with $u - 2p_0/3$ yields

$$u^3 = \frac{1}{3}({p_0}^2 + 12r_0)u + \frac{1}{27}(2{p_0}^3 - 27p_0r_0 + 72{q_0}^2)$$

According to the Viete's method [5, 6], one of the solutions of the cubic equation above is

$$u_0 = 2s_0\cos\left(\frac{1}{3}\arccos\frac{t_0}{2s_0}\right) - \frac{2p_0}{3}$$

with

$$s_0 = \frac{1}{3}\sqrt{{p_0}^2 + 12r_0}$$

$$t_0 = \frac{2{p_0}^3 - 72p_0r_0 + 27{q_0}^2}{3{p_0}^2 + 36r_0}$$

Then, the quartic equation can be factorized to two quadratic equations as follows;

$$\left\{\left(x^2 + \frac{p_0 + u_0}{2}\right) + \sqrt{u_0}\left(x - \frac{q_0}{2u_0}\right)\right\}\left\{\left(x^2 + \frac{p_0 + u_0}{2}\right) - \sqrt{u_0}\left(x - \frac{q_0}{2u_0}\right)\right\} = 0$$

Finally, the exact analytical solutions of the quartic equation are

$$x = \frac{1}{2}\left(\pm_1\sqrt{u_0}\pm_2\sqrt{-2p_0 - u_0\mp_1\frac{2q_0}{\sqrt{u_0}}}\right) + \frac{A_z}{4}$$

The double signs $\pm_1$ and $\pm_2$ can be taken freely, affording four eigenvalues. Therefore, the energy eigenvalues are given in the following;

$$E_1 = \frac{1}{2}\left(-\sqrt{u_0} + \sqrt{-2p_0 - u_0 + \frac{2q_0}{\sqrt{u_0}}}\right) + \frac{A_z}{4}$$

for the |+3/2, +1/2>-dominant state,

$$E_2 = \frac{1}{2}\left(-\sqrt{u_0} - \sqrt{-2p_0 - u_0 + \frac{2q_0}{\sqrt{u_0}}}\right) + \frac{A_z}{4}$$

for the |–3/2, –1/2>-dominant state

$$E_3 = \frac{1}{2}\left(\sqrt{u_0} + \sqrt{-2p_0 - u_0 - \frac{2q_0}{\sqrt{u_0}}}\right) + \frac{A_z}{4}$$

for the |+1/2, –1/2>-dominant state, and

$$E_4 = \frac{1}{2}\left(\sqrt{u_0} - \sqrt{-2p_0 - u_0 - \frac{2q_0}{\sqrt{u_0}}}\right) + \frac{A_z}{4}$$

for the |–1/2, +1/2>-dominant state when $D < 0$.

Now we analytically derive the exact expressions for the eigenfunctions composed of the four spin conjugate states. According to Denton and co-workers [7], the square of the jth element of the eigenvector corresponding to the eigenvalue E_i of the Hermitian matrix can be described as follows

$$|v_{i,j}|^2 \prod_{k=1;k\neq i}^{n} (E_i - E_k) = \prod_{k=1}^{n-1} (E_i - x_{j,k})$$

where v_{ij} is the coefficient to determine, and $x_{j,k}$ is the eigenvalues of the minor M_j of the Hermitian formed by removing the jth row and column. In the case of the spin Hamiltonian, the spin conjugate eigenfunctions Ψ_i corresponding to the eigenvalue E_i can be described as

$$\Psi_i = \alpha_i \left|+\frac{3}{2},+\frac{1}{2}\right\rangle + \beta_i \left|+\frac{1}{2},-\frac{1}{2}\right\rangle + \gamma_i \left|-\frac{1}{2},+\frac{1}{2}\right\rangle + \delta_i \left|-\frac{3}{2},-\frac{1}{2}\right\rangle$$

where α_n, β_n, γ_n and δ_n correspond to v_{n1}, v_{n2}, v_{n3} and v_{n4}, respectively. By using the formula, we calculate α_1, β_2, γ_3 and δ_4, which are the diagonal element of the unitary matrix for diagonalizing the spin Hamiltonian matrix.

$$|\alpha_1|^2 = \frac{(E_1 - x_{1,0})(E_1 - x_{1,1})(E_1 - x_{1,2})}{(E_1 - E_2)(E_1 - E_3)(E_1 - E_4)}$$

$$|\beta_2|^2 = \frac{(E_2 - x_{2,0})(E_2 - x_{2,1})(E_2 - x_{2,2})}{(E_2 - E_1)(E_2 - E_3)(E_2 - E_4)}$$

$$|\gamma_3|^2 = \frac{(E_3 - x_{3,0})(E_3 - x_{3,1})(E_3 - x_{3,2})}{(E_3 - E_1)(E_3 - E_2)(E_3 - E_4)}$$

$$|\delta_4|^2 = \frac{(E_4 - x_{4,0})(E_4 - x_{4,1})(E_4 - x_{4,2})}{(E_4 - E_1)(E_4 - E_2)(E_4 - E_3)}$$

In order to determine the element of the eigenvectors, we define the following matrixes,

$$M_1 = \begin{pmatrix} -D + \frac{1}{2}g_z\beta B - \frac{1}{4}A_z & \frac{1}{2}(A_x + A_y) & \sqrt{3}E \\ \frac{1}{2}(A_x + A_y) & -D - \frac{1}{2}g_z\beta B - \frac{1}{4}A_z & \frac{\sqrt{3}}{4}(A_x - A_y) \\ \sqrt{3}E & \frac{\sqrt{3}}{4}(A_x - A_y) & D - \frac{3}{2}g_z\beta B + \frac{3}{4}A_z \end{pmatrix}$$

$$M_2 = \begin{pmatrix} D + \frac{3}{2}g_z\beta B + \frac{3}{4}A_z & \sqrt{3}E & 0 \\ \sqrt{3}E & -D - \frac{1}{2}g_z\beta B - \frac{1}{4}A_z & \frac{\sqrt{3}}{4}(A_x - A_y) \\ 0 & \frac{\sqrt{3}}{4}(A_x - A_y) & D - \frac{3}{2}g_z\beta B + \frac{3}{4}A_z \end{pmatrix}$$

$$M_3 = \begin{pmatrix} D + \frac{3}{2}g_z\beta B + \frac{3}{4}A_z & \frac{\sqrt{3}}{4}(A_x - A_y) & 0 \\ \frac{\sqrt{3}}{4}(A_x - A_y) & -D + \frac{1}{2}g_z\beta B - \frac{1}{4}A_z & \sqrt{3}E \\ 0 & \sqrt{3}E & D - \frac{3}{2}g_z\beta B + \frac{3}{4}A_z \end{pmatrix}$$

$$M_4 = \begin{pmatrix} D + \frac{3}{2}g_z\beta B + \frac{3}{4}A_z & \frac{\sqrt{3}}{4}(A_x - A_y) & \sqrt{3}E \\ \frac{\sqrt{3}}{4}(A_x - A_y) & -D + \frac{1}{2}g_z\beta B - \frac{1}{4}A_z & \frac{1}{2}(A_x + A_y) \\ \sqrt{3}E & \frac{1}{2}(A_x + A_y) & -D - \frac{1}{2}g_z\beta B - \frac{1}{4}A_z \end{pmatrix}$$

and the secular equation corresponding to each matrix above is given by the following cubic equation;

$$x^3 + p_i x^2 + q_i x + r_i = 0\ (i = 1,2,3,4)$$

with

$$p_1 = D + \frac{3}{2}g_z\beta B - \frac{1}{4}A_z$$

$$q_1 = -(D^2 + 3E^2) - \frac{1}{4}(g_z\beta B)^2 + 3Dg_z\beta B - \frac{3}{2}DA_z + \frac{3}{4}g_z\beta BA_z - \frac{7}{16}{A_x}^2 - \frac{1}{8}A_xA_y - \frac{7}{16}{A_y}^2 + \frac{3}{4}{A_z}^2$$

$$\begin{aligned} r_1 = {} & -D(D^2 + 3E^2) - \frac{3}{8}(g_z\beta B)^3 + \frac{1}{4}D(g_z\beta B)^2 + \frac{3}{2}(D + E)(D - E)g_z\beta B - \frac{9}{32}g_z\beta B{A_x}^2 \\ & + \frac{1}{16}(D - 12E){A_x}^2 - \frac{15}{16}g_z\beta BA_xA_y + \frac{7}{8}DA_xA_y - \frac{9}{32}g_z\beta B{A_y}^2 + \frac{1}{16}(D + 12E){A_y}^2 \\ & + \frac{3}{16}(g_z\beta B)^2A_z + \frac{3}{4}Dg_z\beta BA_z - \frac{1}{4}(5D^2 + 3E^2)A_z + \frac{9}{64}{A_x}^2A_z + \frac{15}{32}A_xA_yA_z \\ & + \frac{9}{64}{A_y}^2A_z + \frac{13}{32}g_z\beta B{A_z}^2 - \frac{7}{16}D{A_z}^2 - \frac{3}{64}{A_z}^3 \end{aligned}$$

$$p_2 = -D + \frac{1}{2}g_z\beta B - \frac{5}{4}A_z$$

$$q_2 = -(D^2 + 3E^2) - \frac{9}{4}(g_z\beta B)^2 - Dg_z\beta B - \frac{1}{2}DA_z - \frac{3}{4}g_z\beta BA_z - \frac{3}{16}{A_x}^2 + \frac{3}{8}A_xA_y - \frac{3}{16}{A_y}^2 + \frac{3}{16}{A_z}^2$$

$$\begin{aligned} r_2 = {} & D(D^2 + 3E^2) - \frac{9}{8}(g_z\beta B)^3 - \frac{9}{4}D(g_z\beta B)^2 + \frac{1}{2}(D + 3E)(D - 3E)g_z\beta B + \frac{9}{32}g_z\beta B{A_x}^2 + \frac{3}{16}D{A_x}^2 \\ & - \frac{9}{16}g_z\beta BA_xA_y - \frac{3}{8}DA_xA_y + \frac{9}{32}g_z\beta B{A_y}^2 + \frac{3}{16}D{A_y}^2 - \frac{9}{16}(g_z\beta B)^2A_z + \frac{3}{4}Dg_z\beta BA_z \\ & + \frac{1}{4}(7D^2 + 9E^2)A_z + \frac{9}{64}{A_x}^2A_z - \frac{9}{32}A_xA_yA_z + \frac{9}{64}{A_y}^2A_z + \frac{9}{32}g_z\beta B{A_z}^2 + \frac{15}{16}D{A_z}^2 \\ & + \frac{9}{64}{A_z}^3 \end{aligned}$$

$$p_3 = -D - \frac{1}{2}g_z\beta B - \frac{5}{4}A_z$$

$$q_3 = -(D^2 + 3E^2) - \frac{9}{4}(g_z\beta B)^2 + Dg_z\beta B - \frac{1}{2}DA_z + \frac{3}{4}g_z\beta BA_z - \frac{3}{16}{A_x}^2 + \frac{3}{8}A_xA_y - \frac{3}{16}{A_y}^2 + \frac{3}{16}{A_z}^2$$

$$r_3 = D(D^2+3E^2) + \frac{9}{8}(g_z\beta B)^3 - \frac{9}{4}D(g_z\beta B)^2 - \frac{1}{2}(D+3E)(D-3E)g_z\beta B - \frac{9}{32}g_z\beta B{A_x}^2 + \frac{3}{16}D{A_x}^2 + \frac{9}{16}g_z\beta BA_xA_y - \frac{3}{8}DA_xA_y - \frac{9}{32}g_z\beta B{A_y}^2 + \frac{3}{16}D{A_y}^2 - \frac{9}{16}(g_z\beta B)^2A_z - \frac{3}{4}Dg_z\beta BA_z + \frac{1}{4}(7D^2+9E^2)A_z + \frac{9}{64}{A_x}^2A_z - \frac{9}{32}A_xA_yA_z + \frac{9}{64}{A_y}^2A_z - \frac{9}{32}g_z\beta B{A_z}^2 + \frac{15}{16}D{A_z}^2 + \frac{9}{64}{A_z}^3$$

$$p_4 = D - \frac{3}{2}g_z\beta B - \frac{1}{4}A_z$$

$$q_4 = -(D^2+3E^2) - \frac{1}{4}(g_z\beta B)^2 - 3Dg_z\beta B - \frac{3}{2}DA_z - \frac{3}{4}g_z\beta BA_z - \frac{7}{16}{A_x}^2 - \frac{1}{8}A_xA_y - \frac{7}{16}{A_y}^2 + \frac{3}{4}{A_z}^2$$

$$r_4 = -D(D^2+3E^2) + \frac{3}{8}(g_z\beta B)^3 + \frac{1}{4}D(g_z\beta B)^2 - \frac{3}{2}(D+E)(D-E)g_z\beta B + \frac{9}{32}g_z\beta B{A_x}^2 + \frac{1}{16}(D-12E){A_x}^2 + \frac{15}{16}g_z\beta BA_xA_y + \frac{7}{8}DA_xA_y + \frac{9}{32}g_z\beta B{A_y}^2 + \frac{1}{16}(D+12E){A_y}^2 + \frac{3}{16}(g_z\beta B)^2A_z - \frac{3}{4}Dg_z\beta BA_z - \frac{1}{4}(5D^2+3E^2)A_z + \frac{9}{64}{A_x}^2A_z + \frac{15}{32}A_xA_yA_z + \frac{9}{64}{A_y}^2A_z - \frac{13}{32}g_z\beta B{A_z}^2 - \frac{7}{16}D{A_z}^2 - \frac{3}{64}{A_z}^3$$

The cubic equation (i = 1, 2, 3, 4)

$$x^3 + p_ix^2 + q_ix + r_i = 0 \; (i = 1, 2, 3, 4)$$

is transformed as follows;

$$x^3 = \left(\frac{{p_i}^2 - 3q_i}{3}\right)x - \frac{2{p_i}^3 - 9p_iq_i + 27r_i}{27}$$

by substituting x to $x - p_i/3$. Then, the solutions of the cubic equations are represented as follows;

$$x_{i,m} = 2s_i\cos\left[\frac{1}{3}\arccos\left(\frac{t_i}{2s_i}\right) + \frac{2m\pi}{3}\right] - \frac{p_i}{3} \; (m = 0, 1, 2)$$

with

$$s_i = \frac{\sqrt{{p_i}^2 - 3q_i}}{3}$$

$$t_i = -\frac{2{p_i}^3 - 9p_iq_i + 27r_i}{3{p_i}^2 - 9q_i}$$

For $n = 1$ ($M_S = +3/2$ and $M_I = +1/2$), $\alpha_1 = r_{\alpha 1} = \sqrt{|v_{1,1}|^2}$ (real).

$$\begin{cases} \left(D+\frac{3}{2}g_z\beta B+\frac{3}{4}A_z-E_1\right)r_{\alpha 1}+\frac{\sqrt{3}}{4}\left(A_x-A_y\right)\beta_1+\sqrt{3}E\gamma_1=0 \text{ — (A1)} \\ \frac{\sqrt{3}}{4}\left(A_x-A_y\right)r_{\alpha 1}+\left(-D+\frac{1}{2}g_z\beta B-\frac{1}{4}A_z-E_1\right)\beta_1+\frac{1}{2}\left(A_x+A_y\right)\gamma_1+\sqrt{3}E\delta_1=0 \text{ — (B1)} \\ \sqrt{3}Er_{\alpha 1}+\frac{1}{2}\left(A_x+A_y\right)\beta_1+\left(-D-\frac{1}{2}g_z\beta B-\frac{1}{4}A_z-E_1\right)\gamma_1+\frac{\sqrt{3}}{4}\left(A_x-A_y\right)\delta_1=0 \text{ — (C1)} \\ \sqrt{3}E\beta_1+\frac{\sqrt{3}}{4}\left(A_x-A_y\right)\gamma_1+\left(D-\frac{3}{2}g_z\beta B+\frac{3}{4}A_z-E_1\right)\delta_1=0 \text{ — (D1)} \end{cases}$$

(B1) $\times \frac{1}{4}\left(A_x-A_y\right)$

$$\frac{\sqrt{3}}{16}\left(A_x-A_y\right)^2 r_{\alpha 1}+\frac{1}{4}\left(A_x-A_y\right)\left(-D+\frac{1}{2}g_z\beta B-\frac{1}{4}A_z-E_1\right)\beta_1+\frac{1}{8}\left({A_x}^2-{A_y}^2\right)\gamma_1 + \frac{\sqrt{3}}{4}E\left(A_x-A_y\right)\delta_1=0 \text{ — (E1)}$$

(C1) $\times E$

$$\sqrt{3}E^2 r_{\alpha 1}+\frac{1}{2}E\left(A_x+A_y\right)\beta_1+E\left(-D-\frac{1}{2}g_z\beta B-\frac{1}{4}A_z-E_1\right)\gamma_1+\frac{\sqrt{3}}{4}E\left(A_x-A_y\right)\delta_1=0 \text{ — (F1)}$$

(E1) – (F1)

$$\frac{\sqrt{3}}{16}\left[\left(A_x-A_y\right)^2-16E^2\right]r_{\alpha 1}+\frac{1}{4}\left[\left(A_x-A_y\right)\left(-D+\frac{1}{2}g_z\beta B-\frac{1}{4}A_z-E_1\right)-2E\left(A_x+A_y\right)\right]\beta_1 + \left[\frac{1}{8}\left({A_x}^2-{A_y}^2\right)-E\left(-D-\frac{1}{2}g_z\beta B-\frac{1}{4}A_z-E_1\right)\right]\gamma_1=0 \text{ — (G1)}$$

(A1) $\times \sqrt{3}\left[\left(A_x-A_y\right)\left(-D+\frac{1}{2}g_z\beta B-\frac{1}{4}A_z-E_1\right)-2E\left(A_x+A_y\right)\right]$

$$\sqrt{3}\left(D+\frac{3}{2}g_z\beta B+\frac{3}{4}A_z-E_1\right)\left[\left(A_x-A_y\right)\left(-D+\frac{1}{2}g_z\beta B-\frac{1}{4}A_z-E_1\right)-2E\left(A_x+A_y\right)\right]r_{\alpha 1} + \frac{3}{4}\left(A_x-A_y\right)\left[\left(A_x-A_y\right)\left(-D+\frac{1}{2}g_z\beta B-\frac{1}{4}A_z-E_1\right)-2E\left(A_x+A_y\right)\right]\beta_1 + 3E\left[\left(A_x-A_y\right)\left(-D+\frac{1}{2}g_z\beta B-\frac{1}{4}A_z-E_1\right)-2E\left(A_x+A_y\right)\right]\gamma_1=0 \text{ — (H1)}$$

(G1) $\times 3\left(A_x-A_y\right)$

$$\frac{3\sqrt{3}}{16}\left(A_x-A_y\right)\left[\left(A_x-A_y\right)^2-16E^2\right]r_{\alpha 1} + \frac{3}{4}\left(A_x-A_y\right)\left[\left(A_x-A_y\right)\left(-D+\frac{1}{2}g_z\beta B-\frac{1}{4}A_z-E_1\right)-2E\left(A_x+A_y\right)\right]\beta_1 + 3\left(A_x-A_y\right)\left[\frac{1}{8}\left({A_x}^2-{A_y}^2\right)-E\left(-D-\frac{1}{2}g_z\beta B-\frac{1}{4}A_z-E_1\right)\right]\gamma_1=0 \text{ — (I1)}$$

Applying (H1) – (I1) yields

$$\gamma_1=\frac{numer(\gamma_1)}{denom(\gamma_1)}r_{\alpha 1}$$

where

$$numer(\gamma_1) = 3\sqrt{3}(A_x - A_y)\left[(A_x - A_y)^2 - 16E^2\right]$$

$$- 16\sqrt{3}\left(D + \frac{3}{2}g_z\beta B + \frac{3}{4}A_z - E_1\right)\left[(A_x - A_y)\left(-D + \frac{1}{2}g_z\beta B - \frac{1}{4}A_z - E_1\right)\right.$$

$$\left. - 2E(A_x + A_y)\right]$$

$$denom(\gamma_1) = 48E\left[(A_x - A_y)\left(-D + \frac{1}{2}g_z\beta B - \frac{1}{4}A_z - E_1\right) - 2E(A_x + A_y)\right]$$

$$- 6(A_x - A_y)\left[{A_x}^2 - {A_y}^2 - 8E\left(-D - \frac{1}{2}g_z\beta B - \frac{1}{4}A_z - E_1\right)\right]$$

From (A1) we obtain

$$\beta_1 = -\frac{4\sqrt{3}\left(D + \frac{3}{2}g_z\beta B + \frac{3}{4}A_z - E_1\right) + 12E\frac{numer(\gamma_1)}{denom(\gamma_1)}}{3(A_x - A_y)}r_{\alpha 1}$$

Finally, from (D1) we obtain

$$\delta_1 = \frac{-4\sqrt{3}E\beta_1 - \sqrt{3}(A_x - A_y)\gamma_1}{4D - 6g_z\beta B + 3A_z - E_1}$$

For the basis sets of {|+3/2, –1/2>, |+1/2, +1/2>, |–1/2, –1/2>, |–3/2, +1/2>} ($M_S + M_I = \pm 1$), the corresponding spin Hamiltonian is written as

$$H_{\mathrm{full},2}^{S=\frac{3}{2},I=\frac{1}{2}} = \begin{pmatrix} D + \frac{3}{2}g_z\beta B - \frac{3}{4}A_z & \frac{\sqrt{3}}{4}(A_x + A_y) & \sqrt{3}E & 0 \\ \frac{\sqrt{3}}{4}(A_x + A_y) & -D + \frac{1}{2}g_z\beta B + \frac{1}{4}A_z & \frac{1}{2}(A_x - A_y) & \sqrt{3}E \\ \sqrt{3}E & \frac{1}{2}(A_x - A_y) & -D - \frac{1}{2}g_z\beta B + \frac{1}{4}A_z & \frac{\sqrt{3}}{4}(A_x + A_y) \\ 0 & \sqrt{3}E & \frac{\sqrt{3}}{4}(A_x + A_y) & D - \frac{3}{2}g_z\beta B + \frac{3}{4}A_z \end{pmatrix}$$

The secular quartic equation above, which is the counterpart Hamiltonian based on the spin conjugate function, can be established by replacing A_y and A_z to $-A_y$ and $-A_z$, respectively, of the counterpart of $H_{\mathrm{full},1}^{S=\frac{3}{2},I=\frac{1}{2}}$. Thus, the eigenvalues E_i (i = 5,6,7,8) are given as

$$E_1 \xrightarrow{A_y=-A_y, A_z=-A_z} E_5$$

for the |+3/2, –1/2>-dominant state,

$$E_2 \xrightarrow{A_y=-A_y, A_z=-A_z} E_6$$

for the |–3/2, +1/2>-dominant state.

$$E_3 \xrightarrow{A_y=-A_y, A_z=-A_z} E_7$$

for the |+1/2, +1/2>-dominant state, and

$$E_4 \xrightarrow{A_y=-A_y, A_z=-A_z} E_8$$

for the |–1/2, –1/2>-dominant state.

b) Double perturbation approach

The perturbation approach for the case of the spin quartet state with $I = 1/2$ does not necessarily encourage to merit attention, but it is worth comparing the accuracy of the double perturbation treatment with the exact analytical treatment. In this context, we note that our Zeeman perturbation approach applied to the cases without hyperfine interactions affords extremely accurate analytical expressions for the eigenvalues and functions. The spin Hamiltonian for the ZFS terms, which come from the spin–spin and spin–orbit interactions in the cases of $S \geq 1$, is taken as the non-perturbed Hamiltonian. The matrix representation of the rank-2 ZFS Hamiltonian in terms of the basis set ($|M_S, M_I\rangle = |+3/2, +1/2\rangle, |+1/2, +1/2\rangle, |-1/2, +1/2\rangle, |-3/2, +1/2\rangle$) is given as

$$H_{\mathrm{zfs}}^{S=\frac{3}{2}} = \begin{pmatrix} D & 0 & \sqrt{3}E & 0 \\ 0 & -D & 0 & \sqrt{3}E \\ \sqrt{3}E & 0 & -D & 0 \\ 0 & \sqrt{3}E & 0 & D \end{pmatrix}$$

The eigenvalues and the corresponding eigen spin wavefunctions are given, respectively in the following:

$$\varepsilon_{\pm\frac{3}{2},+\frac{1}{2}}^{(0)} = \Delta \quad \varphi_{\pm\frac{3}{2},+\frac{1}{2}}^{(0)} = \cos\theta \left|\pm\frac{3}{2}, +\frac{1}{2}\right\rangle + \sin\theta \left|\mp\frac{1}{2}, +\frac{1}{2}\right\rangle$$

$$\varepsilon_{\pm\frac{1}{2},+\frac{1}{2}}^{(0)} = -\Delta \quad \varphi_{\pm\frac{1}{2},+\frac{1}{2}}^{(0)} = \cos\theta \left|\pm\frac{1}{2}, +\frac{1}{2}\right\rangle - \sin\theta \left|\mp\frac{3}{2}, +\frac{1}{2}\right\rangle$$

where

$$\Delta = \sqrt{D^2 + 3E^2}$$

and

$$\tan 2\theta = \frac{\sqrt{3}E}{D}$$

It is noted that the half-integer spin $S = 3/2$ results in the degeneracy in the absence of the static magnetic field and this gives complexity in treating the hyperfine structure terms, which is underlain by the spin conjugate properties.

First, $\left\{\left|\varphi_{M_S,M_I}^{(0)}\right\rangle\right\}$ are divided to two subspaces corresponding to the eigenvalues;

$$\left\{\left|\varphi_{+\frac{3}{2},+\frac{1}{2}}^{(0)}\right\rangle, \left|\varphi_{+\frac{3}{2},-\frac{1}{2}}^{(0)}\right\rangle, \left|\varphi_{-\frac{3}{2},+\frac{1}{2}}^{(0)}\right\rangle, \left|\varphi_{-\frac{3}{2},-\frac{1}{2}}^{(0)}\right\rangle\right\} \text{ (eigenvalue } \Delta)$$

$$\left\{\left|\varphi_{+\frac{1}{2},+\frac{1}{2}}^{(0)}\right\rangle, \left|\varphi_{+\frac{1}{2},-\frac{1}{2}}^{(0)}\right\rangle, \left|\varphi_{-\frac{1}{2},+\frac{1}{2}}^{(0)}\right\rangle, \left|\varphi_{-\frac{1}{2},-\frac{1}{2}}^{(0)}\right\rangle\right\} \text{ (eigenvalue } -\Delta)$$

For simplicity, we rewrite the notation of the eigenfunctions as follows;

$$\left\{\left|\varphi_{+\frac{3}{2},+\frac{1}{2}}^{(0)}\right\rangle, \left|\varphi_{+\frac{3}{2},-\frac{1}{2}}^{(0)}\right\rangle, \left|\varphi_{-\frac{3}{2},+\frac{1}{2}}^{(0)}\right\rangle, \left|\varphi_{-\frac{3}{2},-\frac{1}{2}}^{(0)}\right\rangle\right\} \to \{|+,1\rangle, |+,2\rangle, |+,3\rangle, |+,4\rangle\}$$

$$\left\{\left|\varphi_{+\frac{1}{2},+\frac{1}{2}}^{(0)}\right\rangle, \left|\varphi_{+\frac{1}{2},-\frac{1}{2}}^{(0)}\right\rangle, \left|\varphi_{-\frac{1}{2},+\frac{1}{2}}^{(0)}\right\rangle, \left|\varphi_{-\frac{1}{2},-\frac{1}{2}}^{(0)}\right\rangle\right\} \to \{|-,1\rangle, |-,2\rangle, |-,3\rangle, |-,4\rangle\}$$

The matrix elements of the hyperfine structure Hamiltonian in the basis of $|M_S, M_I\rangle$ are given as

$$\langle M_S', M_I' | H_{\mathrm{hfs}} | M_S, M_I \rangle = \begin{cases} \delta_{M_S M_S'} \delta_{M_I M_I'} M_S M_I A_{zz} \\ \dfrac{1}{16} \delta_{M_S M_S' \mp 1} \delta_{M_I M_I' \mp 1} \sqrt{15 - 4 M_S M_S'} \sqrt{3 - 4 M_I M_I'} \left(A_{xx} - A_{yy}\right) \\ \dfrac{1}{16} \delta_{M_S M_S' \mp 1} \delta_{M_I M_I' \pm 1} \sqrt{15 - 4 M_S M_S'} \sqrt{3 - 4 M_I M_I'} \left(A_{xx} + A_{yy}\right) \end{cases}$$

According to the degenerate perturbation theory, the first-order corrections for the energy of the hyperfine structure Hamiltonian ($\varepsilon_{+,\alpha}^{(01)}$ $(\alpha = 1,2,3,4)$) which belong to the group of the positive energy eigenvalue are given as the eigenvalues of the following matrix:

$$H_{\mathrm{hfs}}^{+} = \begin{pmatrix} \langle +,1 | H_{\mathrm{hfs}} | +,1 \rangle & \langle +,1 | H_{\mathrm{hfs}} | +,2 \rangle & \langle +,1 | H_{\mathrm{hfs}} | +,3 \rangle & \langle +,1 | H_{\mathrm{hfs}} | +,4 \rangle \\ \langle +,2 | H_{\mathrm{hfs}} | +,1 \rangle & \langle +,2 | H_{\mathrm{hfs}} | +,2 \rangle & \langle +,2 | H_{\mathrm{hfs}} | +,3 \rangle & \langle +,2 | H_{\mathrm{hfs}} | +,4 \rangle \\ \langle +,3 | H_{\mathrm{hfs}} | +,1 \rangle & \langle +,3 | H_{\mathrm{hfs}} | +,2 \rangle & \langle +,3 | H_{\mathrm{hfs}} | +,3 \rangle & \langle +,3 | H_{\mathrm{hfs}} | +,4 \rangle \\ \langle +,4 | H_{\mathrm{hfs}} | +,1 \rangle & \langle +,4 | H_{\mathrm{hfs}} | +,2 \rangle & \langle +,4 | H_{\mathrm{hfs}} | +,3 \rangle & \langle +,4 | H_{\mathrm{hfs}} | +,4 \rangle \end{pmatrix}$$

where each matrix element can be calculated in the following;

$$\langle +,1 | H_{\mathrm{hfs}} | +,1 \rangle = \langle +,4 | H_{\mathrm{hfs}} | +,4 \rangle = \frac{A_z}{4}(1 + 2\cos 2\theta)$$

$$\langle +,2 | H_{\mathrm{hfs}} | +,2 \rangle = \langle +,3 | H_{\mathrm{hfs}} | +,3 \rangle = -\frac{A_z}{4}(1 + 2\cos 2\theta)$$

$$\langle +,1 | H_{\mathrm{hfs}} | +,4 \rangle = \langle +,4 | H_{\mathrm{hfs}} | +,1 \rangle = \frac{1}{2}\sin\theta \left[\sqrt{3}\left(A_x - A_y\right)\cos\theta + \left(A_x + A_y\right)\sin\theta\right]$$

$$\langle +,2 | H_{\mathrm{hfs}} | +,3 \rangle = \langle +,3 | H_{\mathrm{hfs}} | +,2 \rangle = \frac{1}{2}\sin\theta \left[\sqrt{3}\left(A_x + A_y\right)\cos\theta + \left(A_x - A_y\right)\sin\theta\right]$$

The other elements are zero. Thus, the matrix can be divided to two 2×2 matrixes with the basis of {|+, 1>, |+, 4>} and {|+, 2>, |+, 3>}. The secular determinants are as follows; for the {|+, 1>, |+, 4>} basis set,

$$\begin{vmatrix} \dfrac{A_z}{4}(1 + 2\cos 2\theta) - x & \dfrac{1}{2}\sin\theta \left[\sqrt{3}\left(A_x - A_y\right)\cos\theta + \left(A_x + A_y\right)\sin\theta\right] \\ \dfrac{1}{2}\sin\theta \left[\sqrt{3}\left(A_x - A_y\right)\cos\theta + \left(A_x + A_y\right)\sin\theta\right] & \dfrac{A_z}{4}(1 + 2\cos 2\theta) - x \end{vmatrix} = 0$$

and for the {|+, 2>, |+, 4>} basis set,

$$\begin{vmatrix} -\dfrac{A_z}{4}(1 + 2\cos 2\theta) - x & \dfrac{1}{2}\sin\theta \left[\sqrt{3}\left(A_x + A_y\right)\cos\theta + \left(A_x - A_y\right)\sin\theta\right] \\ \dfrac{1}{2}\sin\theta \left[\sqrt{3}\left(A_x + A_y\right)\cos\theta + \left(A_x - A_y\right)\sin\theta\right] & -\dfrac{A_z}{4}(1 + 2\cos 2\theta) - x \end{vmatrix} = 0$$

From the first secular equation, we obtain

$$\begin{aligned} &\varepsilon_{+,1}^{(01)} \\ &= \frac{1}{4}\Bigg\{A_z + 2A_z\cos 2\theta \\ &+ 2\sqrt{\sin^2\theta \left[2\left({A_x}^2 - A_x A_y + {A_y}^2\right) + \left({A_x}^2 - 4A_x A_y + {A_y}^2\right)\cos 2\theta + \sqrt{3}\left({A_x}^2 - {A_y}^2\right)\sin 2\theta\right]}\Bigg\} \end{aligned}$$

$$\begin{aligned} &\varepsilon_{+,4}^{(01)} \\ &= \frac{1}{4}\Bigg\{A_z + 2A_z\cos 2\theta \\ &- 2\sqrt{\sin^2\theta \left[2\left({A_x}^2 - A_x A_y + {A_y}^2\right) + \left({A_x}^2 - 4A_x A_y + {A_y}^2\right)\cos 2\theta + \sqrt{3}\left({A_x}^2 - {A_y}^2\right)\sin 2\theta\right]}\Bigg\} \end{aligned}$$

From the second secular equation, we obtain

$$\varepsilon_{+,2}^{(01)}$$

$$= \frac{1}{4}\Bigg\{-A_z - 2A_z\cos 2\theta$$

$$+ 2\sqrt{\sin^2\theta\left[2\left({A_x}^2 + A_xA_y + {A_y}^2\right) + \left({A_x}^2 + 4A_xA_y + {A_y}^2\right)\cos 2\theta + \sqrt{3}\left({A_x}^2 - {A_y}^2\right)\sin 2\theta\right]}\Bigg\}$$

$$\varepsilon_{+,3}^{(01)}$$

$$= \frac{1}{4}\Bigg\{-A_z - 2A_z\cos 2\theta$$

$$- 2\sqrt{\sin^2\theta\left[2\left({A_x}^2 + A_xA_y + {A_y}^2\right) + \left({A_x}^2 + 4A_xA_y + {A_y}^2\right)\cos 2\theta + \sqrt{3}\left({A_x}^2 - {A_y}^2\right)\sin 2\theta\right]}\Bigg\}$$

Similarly, the first-order corrections for the energy of the hyperfine structure Hamiltonian ($\varepsilon_{-,\alpha}^{(01)}$ $(\alpha = 1,2,3,4)$) which belong to the group of the negative energy eigenvalue are the eigenvalues of the following matrix:

$$H_{\mathrm{hfs}}^{-} = \begin{pmatrix} \langle -,1|H_{\mathrm{hfs}}|-,1\rangle & \langle -,1|H_{\mathrm{hfs}}|-,2\rangle & \langle -,1|H_{\mathrm{hfs}}|-,3\rangle & \langle -,1|H_{\mathrm{hfs}}|-,4\rangle \\ \langle -,2|H_{\mathrm{hfs}}|-,1\rangle & \langle -,2|H_{\mathrm{hfs}}|-,2\rangle & \langle -,2|H_{\mathrm{hfs}}|-,3\rangle & \langle -,2|H_{\mathrm{hfs}}|-,4\rangle \\ \langle -,3|H_{\mathrm{hfs}}|-,1\rangle & \langle -,3|H_{\mathrm{hfs}}|-,2\rangle & \langle -,3|H_{\mathrm{hfs}}|-,3\rangle & \langle -,3|H_{\mathrm{hfs}}|-,4\rangle \\ \langle -,4|H_{\mathrm{hfs}}|-,1\rangle & \langle -,4|H_{\mathrm{hfs}}|-,2\rangle & \langle -,4|H_{\mathrm{hfs}}|-,3\rangle & \langle -,4|H_{\mathrm{hfs}}|-,4\rangle \end{pmatrix}$$

with each matrix element represented as follows:

$$\langle -,1|H_{\mathrm{hfs}}|-,1\rangle = \langle -,4|H_{\mathrm{hfs}}|-,4\rangle = -\frac{A_z}{4}(1 - 2\cos 2\theta)$$

$$\langle -,2|H_{\mathrm{hfs}}|-,2\rangle = \langle -,3|H_{\mathrm{hfs}}|-,3\rangle = \frac{A_z}{4}(1 - 2\cos 2\theta)$$

$$\langle -,1|H_{\mathrm{hfs}}|-,4\rangle = \langle -,4|H_{\mathrm{hfs}}|-,1\rangle = \frac{1}{2}\cos\theta\left[\left(A_x - A_y\right)\cos\theta - \sqrt{3}\left(A_x + A_y\right)\sin\theta\right]$$

$$\langle -,2|H_{\mathrm{hfs}}|-,3\rangle = \langle -,3|H_{\mathrm{hfs}}|-,2\rangle = \frac{1}{2}\cos\theta\left[\left(A_x + A_y\right)\cos\theta - \sqrt{3}\left(A_x - A_y\right)\sin\theta\right]$$

The other elements are zero. Thus, the matrix also can be divided to two 2×2 matrixes with the basis of {|–, 1>, |–, 4>} and {|–, 2>, |–, 3>}. The secular determinants are as follows; for the {|–, 1>, |–, 4>} basis set,

$$\begin{vmatrix} -\frac{A_z}{4}(1 - 2\cos 2\theta) - x & \frac{1}{2}\cos\theta\left[\left(A_x - A_y\right)\cos\theta - \sqrt{3}\left(A_x + A_y\right)\sin\theta\right] \\ \frac{1}{2}\cos\theta\left[\left(A_x - A_y\right)\cos\theta - \sqrt{3}\left(A_x + A_y\right)\sin\theta\right] & -\frac{A_z}{4}(1 - 2\cos 2\theta) - x \end{vmatrix} = 0$$

And for the {|–, 2>, |–, 4>} basis set,

$$\begin{vmatrix} \frac{A_z}{4}(1 - 2\cos 2\theta) - x & \frac{1}{2}\cos\theta\left[\left(A_x + A_y\right)\cos\theta - \sqrt{3}\left(A_x - A_y\right)\sin\theta\right] \\ \frac{1}{2}\cos\theta\left[\left(A_x + A_y\right)\cos\theta - \sqrt{3}\left(A_x - A_y\right)\sin\theta\right] & \frac{A_z}{4}(1 - 2\cos 2\theta) - x \end{vmatrix} = 0$$

From the first secular equation, we obtain

$$\varepsilon_{-,1}^{(01)}$$
$$= \frac{1}{4}\Bigg\{-A_z + 2A_z \cos 2\theta$$
$$+ 2\sqrt{\cos^2\theta \left[2\left(A_x{}^2 + A_xA_y + A_y{}^2\right) - \left(A_x{}^2 + 4A_xA_y + A_y{}^2\right)\cos 2\theta - \sqrt{3}\left(A_x{}^2 - A_y{}^2\right)\sin 2\theta\right]}\Bigg\}$$
$$\varepsilon_{-,4}^{(01)}$$
$$= \frac{1}{4}\Bigg\{-A_z + 2A_z \cos 2\theta$$
$$- 2\sqrt{\cos^2\theta \left[2\left(A_x{}^2 + A_xA_y + A_y{}^2\right) - \left(A_x{}^2 + 4A_xA_y + A_y{}^2\right)\cos 2\theta - \sqrt{3}\left(A_x{}^2 - A_y{}^2\right)\sin 2\theta\right]}\Bigg\}$$

From the second secular equation, we obtain

$$\varepsilon_{-,2}^{(01)}$$
$$= \frac{1}{4}\Bigg\{A_z - 2A_z \cos 2\theta$$
$$+ 2\sqrt{\cos^2\theta \left[2\left(A_x{}^2 - A_xA_y + A_y{}^2\right) - \left(A_x{}^2 - 4A_xA_y + A_y{}^2\right)\cos 2\theta - \sqrt{3}\left(A_x{}^2 - A_y{}^2\right)\sin 2\theta\right]}\Bigg\}$$
$$\varepsilon_{-,3}^{(01)}$$
$$= \frac{1}{4}\Bigg\{A_z - 2A_z \cos 2\theta$$
$$- 2\sqrt{\cos^2\theta \left[2\left(A_x{}^2 - A_xA_y + A_y{}^2\right) - \left(A_x{}^2 - 4A_xA_y + A_y{}^2\right)\cos 2\theta - \sqrt{3}\left(A_x{}^2 - A_y{}^2\right)\sin 2\theta\right]}\Bigg\}$$

The second-order correction for the energy of the hyperfine structure Hamiltonian ($\varepsilon_{\sigma,\alpha}^{(02)}$ ($\sigma = +,-;\ \alpha = 1,2,3,4$)) can be written as

$$\varepsilon_{\pm,\alpha}^{(02)} = \sum_{\beta=1}^{4} \frac{|\langle \pm,\alpha|H_{\mathrm{hfs}}|\mp,\beta\rangle|^2}{\varepsilon_{\pm}^{(0)} - \varepsilon_{\mp}^{(0)}}$$

Both the upper and lower signs should be chosen in the double sign. The non-zero matrix elements of H_{hfs} expanded to the basis belonging different eigenspaces are as follows:

$$\langle +,1|H_{\mathrm{hfs}}|-,2\rangle = \langle -,2|H_{\mathrm{hfs}}|+,1\rangle = \frac{\sqrt{3}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{1}{4}\left(A_x + A_y\right)\sin 2\theta$$

$$\langle +,1|H_{\mathrm{hfs}}|-,3\rangle = \langle -,3|H_{\mathrm{hfs}}|+,1\rangle = -\frac{A_z}{2}\sin 2\theta$$

$$\langle +,2|H_{\mathrm{hfs}}|-,1\rangle = \langle -,1|H_{\mathrm{hfs}}|+,2\rangle = \frac{\sqrt{3}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{1}{4}\left(A_x - A_y\right)\sin 2\theta$$

$$\langle +,2|H_{\mathrm{hfs}}|-,4\rangle = \langle -,4|H_{\mathrm{hfs}}|+,2\rangle = \frac{A_z}{2}\sin 2\theta$$

$$\langle +,3|H_{\mathrm{hfs}}|-,1\rangle = \langle -,1|H_{\mathrm{hfs}}|+,3\rangle = \frac{A_z}{2}\sin 2\theta$$

$$\langle +,3|H_{\mathrm{hfs}}|-,4\rangle = \langle -,4|H_{\mathrm{hfs}}|+,3\rangle = \frac{\sqrt{3}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{1}{4}\left(A_x - A_y\right)\sin 2\theta$$

$$\langle +,4|H_{\mathrm{hfs}}|-,2\rangle = \langle -,2|H_{\mathrm{hfs}}|+,4\rangle = -\frac{A_z}{2}\sin 2\theta$$

$$\langle +,4|H_{\mathrm{hfs}}|-,3\rangle = \langle -,3|H_{\mathrm{hfs}}|+,4\rangle = \frac{\sqrt{3}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{1}{4}\left(A_x + A_y\right)\sin 2\theta$$

Therefore,

$$\begin{aligned}
\varepsilon_{+,1}^{(02)} &= \frac{|\langle -,2|H_{\mathrm{hfs}}|+,1\rangle|^2 + |\langle -,3|H_{\mathrm{hfs}}|+,1\rangle|^2}{2\Delta} \\
&= \frac{1}{2\Delta}\left\{\left[\frac{\sqrt{3}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{1}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \frac{{A_z}^2}{4}\sin^2 2\theta\right\} \\
\varepsilon_{+,2}^{(02)} &= \frac{|\langle -,1|H_{\mathrm{hfs}}|+,2\rangle|^2 + |\langle -,4|H_{\mathrm{hfs}}|+,2\rangle|^2}{2\Delta} \\
&= \frac{1}{2\Delta}\left\{\left[\frac{\sqrt{3}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{1}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2 + \frac{{A_z}^2}{4}\sin^2 2\theta\right\} \\
\varepsilon_{+,3}^{(02)} &= \frac{|\langle -,1|H_{\mathrm{hfs}}|+,3\rangle|^2 + |\langle -,4|H_{\mathrm{hfs}}|+,3\rangle|^2}{2\Delta} \\
&= \frac{1}{2\Delta}\left\{\frac{{A_z}^2}{4}\sin^2 2\theta + \left[\frac{\sqrt{3}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{1}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2\right\} \\
\varepsilon_{+,4}^{(02)} &= \frac{|\langle -,2|H_{\mathrm{hfs}}|+,4\rangle|^2 + |\langle -,3|H_{\mathrm{hfs}}|+,4\rangle|^2}{2\Delta} \\
&= \frac{1}{2\Delta}\left\{\frac{{A_z}^2}{4}\sin^2 2\theta + \left[\frac{\sqrt{3}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{1}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2\right\} \\
\varepsilon_{-,1}^{(02)} &= \frac{|\langle +,2|H_{\mathrm{hfs}}|-,1\rangle|^2 + |\langle +,3|H_{\mathrm{hfs}}|-,1\rangle|^2}{-2\Delta} \\
&= -\frac{1}{2\Delta}\left\{\left[\frac{\sqrt{3}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{1}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2 + \frac{{A_z}^2}{4}\sin^2 2\theta\right\} \\
\varepsilon_{-,2}^{(02)} &= \frac{|\langle +,1|H_{\mathrm{hfs}}|-,2\rangle|^2 + |\langle +,4|H_{\mathrm{hfs}}|-,2\rangle|^2}{-2\Delta} \\
&= -\frac{1}{2\Delta}\left\{\left[\frac{\sqrt{3}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{1}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \frac{{A_z}^2}{4}\sin^2 2\theta\right\} \\
\varepsilon_{-,3}^{(02)} &= \frac{|\langle +,1|H_{\mathrm{hfs}}|-,3\rangle|^2 + |\langle +,4|H_{\mathrm{hfs}}|-,3\rangle|^2}{2\Delta} \\
&= -\frac{1}{2\Delta}\left\{\frac{{A_z}^2}{4}\sin^2 2\theta + \left[\frac{\sqrt{3}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{1}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2\right\}
\end{aligned}$$

$$\varepsilon_{-,4}^{(02)} = \frac{|\langle +,2|H_{\mathrm{hfs}}|-,4\rangle|^2 + |\langle +,3|H_{\mathrm{hfs}}|-,4\rangle|^2}{2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\frac{{A_z}^2}{4}\sin^2 2\theta + \left[\frac{\sqrt{3}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{1}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2\right\}$$

In order to calculate the first-order corrections for the energy of the electron-Zeeman Hamiltonian, $H_{\mathrm{eZ}}^{S=\frac{3}{2}} = g_z \beta S_z B$ is also expanded by $|\sigma,\alpha\rangle$ ($\sigma = +,-;\ \alpha = 1,2,3,4$):

$$\langle +,1|H_{\mathrm{eZ}}|+,1\rangle = \langle +,2|H_{\mathrm{eZ}}|+,2\rangle = \frac{g_z\beta B}{2}(3\cos^2\theta - \sin^2\theta)$$

$$\langle +,3|H_{\mathrm{eZ}}|+,3\rangle = \langle +,4|H_{\mathrm{eZ}}|+,4\rangle = -\frac{g_z\beta B}{2}(3\cos^2\theta - \sin^2\theta)$$

$$\langle +,\beta|H_{\mathrm{eZ}}|+,\alpha\rangle = \langle +,\alpha|H_{\mathrm{eZ}}|+,\beta\rangle = 0\ (\alpha \neq \beta)$$

$$\langle -,1|H_{\mathrm{eZ}}|-,1\rangle = \langle -,2|H_{\mathrm{eZ}}|-,2\rangle = \frac{g_z\beta B}{2}(\cos^2\theta - 3\sin^2\theta)$$

$$\langle -,3|H_{\mathrm{eZ}}|-,3\rangle = \langle -,4|H_{\mathrm{eZ}}|-,4\rangle = -\frac{g_z\beta B}{2}(\cos^2\theta - 3\sin^2\theta)$$

$$\langle -,\beta|H_{\mathrm{eZ}}|-,\alpha\rangle = \langle -,\alpha|H_{\mathrm{eZ}}|-,\beta\rangle = 0\ (\alpha \neq \beta)$$

Thus, the first-order corrections for the energy of the electron-Zeeman Hamiltonian are as follows:

$$\varepsilon_{+\frac{3}{2},M_I}^{(10)} = \frac{g_z\beta B}{2}(3\cos^2\theta - \sin^2\theta)$$

$$\varepsilon_{+\frac{1}{2},M_I}^{(10)} = \frac{g_z\beta B}{2}(\cos^2\theta - 3\sin^2\theta)$$

$$\varepsilon_{-\frac{1}{2},M_I}^{(10)} = -\frac{g_z\beta B}{2}(\cos^2\theta - 3\sin^2\theta)$$

$$\varepsilon_{-\frac{3}{2},M_I}^{(10)} = -\frac{g_z\beta B}{2}(3\cos^2\theta - \sin^2\theta)$$

In order to calculate the second-order correction for the electron-Zeeman Hamiltonian, the non-diagonal components of the electron-Zeeman Hamiltonian are obtained.

$$\langle +,1|H_{\mathrm{eZ}}|-,3\rangle = \langle -,3|H_{\mathrm{eZ}}|+,1\rangle = -2g_z\beta B\sin\theta\cos\theta$$

$$\langle +,2|H_{\mathrm{eZ}}|-,4\rangle = \langle -,4|H_{\mathrm{eZ}}|+,2\rangle = -2g_z\beta B\sin\theta\cos\theta$$

$$\langle +,3|H_{\mathrm{eZ}}|-,1\rangle = \langle -,1|H_{\mathrm{eZ}}|+,3\rangle = 2g_z\beta B\sin\theta\cos\theta$$

$$\langle +,4|H_{\mathrm{eZ}}|-,2\rangle = \langle -,2|H_{\mathrm{eZ}}|+,4\rangle = 2g_z\beta B\sin\theta\cos\theta$$

The other elements are zero. Therefore,

$$\varepsilon_{+\frac{3}{2},M_I}^{(20)} = \frac{2(g_z\beta B)^2\sin^2\theta\cos^2\theta}{\sqrt{D^2+3E^2}}$$

$$\varepsilon_{-\frac{3}{2},M_I}^{(20)} = \frac{2(g_z\beta B)^2\sin^2\theta\cos^2\theta}{\sqrt{D^2+3E^2}}$$

$$\varepsilon_{+\frac{1}{2},M_I}^{(20)} = -\frac{2(g_z\beta B)^2\sin^2\theta\cos^2\theta}{\sqrt{D^2+3E^2}}$$

$$\varepsilon^{(20)}_{-\frac{1}{2},M_I} = -\frac{2(g_z\beta B)^2\sin^2\theta\cos^2\theta}{\sqrt{D^2+3E^2}}$$

From the double perturbation theory, the correction for the energy in the case of $\lambda = 1$ and $\mu = 1$ can be represented as

$$\varepsilon_n^{(11)} = \sum_{m\neq n}\frac{\langle n|H_1|m\rangle\langle m|H_2|n\rangle + \langle n|H_2|m\rangle\langle m|H_1|n\rangle}{\varepsilon_n^{(0)} - \varepsilon_m^{(0)}}$$

In this case, H_1 and H_2 are the electron Zeeman and hyperfine structure Hamiltonians, respectively. Thus,

$$\varepsilon^{(11)}_{+,1} = \frac{\langle +,1|H_{\mathrm{eZ}}|-,3\rangle\langle -,3|H_{\mathrm{hfs}}|+,1\rangle + \langle +,1|H_{\mathrm{hfs}}|-,3\rangle\langle -,3|H_{\mathrm{eZ}}|+,1\rangle}{\varepsilon_+^{(0)} - \varepsilon_-^{(0)}} = \frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon^{(11)}_{+,2} = \frac{\langle +,2|H_{\mathrm{eZ}}|-,4\rangle\langle -,4|H_{\mathrm{hfs}}|+,2\rangle + \langle +,2|H_{\mathrm{hfs}}|-,4\rangle\langle -,4|H_{\mathrm{eZ}}|+,2\rangle}{\varepsilon_+^{(0)} - \varepsilon_-^{(0)}} = -\frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon^{(11)}_{-,1} = \frac{\langle -,1|H_{\mathrm{eZ}}|+,3\rangle\langle +,3|H_{\mathrm{hfs}}|-,1\rangle + \langle -,1|H_{\mathrm{hfs}}|+,3\rangle\langle +,3|H_{\mathrm{eZ}}|-,1\rangle}{\varepsilon_-^{(0)} - \varepsilon_+^{(0)}} = -\frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon^{(11)}_{-,2} = \frac{\langle -,2|H_{\mathrm{eZ}}|+,4\rangle\langle +,4|H_{\mathrm{hfs}}|-,2\rangle + \langle -,2|H_{\mathrm{hfs}}|+,4\rangle\langle +,4|H_{\mathrm{eZ}}|-,2\rangle}{\varepsilon_-^{(0)} - \varepsilon_+^{(0)}} = \frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon^{(11)}_{-,3} = \frac{\langle -,3|H_{\mathrm{eZ}}|+,1\rangle\langle +,1|H_{\mathrm{hfs}}|-,3\rangle + \langle -,3|H_{\mathrm{hfs}}|+,1\rangle\langle +,1|H_{\mathrm{eZ}}|-,3\rangle}{\varepsilon_-^{(0)} - \varepsilon_+^{(0)}} = -\frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon^{(11)}_{-,4} = \frac{\langle -,4|H_{\mathrm{eZ}}|+,2\rangle\langle +,2|H_{\mathrm{hfs}}|-,4\rangle + \langle -,4|H_{\mathrm{hfs}}|+,2\rangle\langle +,2|H_{\mathrm{eZ}}|-,4\rangle}{\varepsilon_-^{(0)} - \varepsilon_+^{(0)}} = \frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon^{(11)}_{+,3} = \frac{\langle +,3|H_{\mathrm{eZ}}|-,1\rangle\langle -,1|H_{\mathrm{hfs}}|+,3\rangle + \langle +,3|H_{\mathrm{hfs}}|-,1\rangle\langle -,1|H_{\mathrm{eZ}}|+,3\rangle}{\varepsilon_+^{(0)} - \varepsilon_-^{(0)}} = \frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon^{(11)}_{+,4} = \frac{\langle +,4|H_{\mathrm{eZ}}|-,2\rangle\langle -,2|H_{\mathrm{hfs}}|+,4\rangle + \langle +,4|H_{\mathrm{hfs}}|-,2\rangle\langle -,2|H_{\mathrm{eZ}}|+,4\rangle}{\varepsilon_+^{(0)} - \varepsilon_-^{(0)}} = -\frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

The perturbed energies in the second order are:

$$\begin{aligned}
&E_{+\frac{3}{2},+\frac{1}{2}} \\
&= \sqrt{D^2+3E^2} + \frac{g_z\beta B}{2}(3\cos^2\theta - \sin^2\theta) + \frac{2(g_z\beta B)^2\sin^2\theta\cos^2\theta}{\sqrt{D^2+3E^2}} \\
&+ \frac{1}{4}\Bigg\{A_z + 2A_z\cos 2\theta \\
&+ 2\sqrt{\sin^2\theta\left[2\left({A_x}^2 - A_xA_y + {A_y}^2\right) + \left({A_x}^2 - 4A_xA_y + {A_y}^2\right)\cos 2\theta + \sqrt{3}\left({A_x}^2 - {A_y}^2\right)\sin 2\theta\right]}\Bigg\} \\
&\quad + \frac{1}{2\Delta}\left\{\left[\frac{\sqrt{3}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{1}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \frac{{A_z}^2}{4}\sin^2 2\theta\right\} + \frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}
\end{aligned}$$

$$E_{+\frac{3}{2},-\frac{1}{2}}$$

$$= \sqrt{D^2+3E^2} + \frac{g_z\beta B}{2}(3\cos^2\theta - \sin^2\theta) + \frac{2(g_z\beta B)^2\sin^2\theta\cos^2\theta}{\sqrt{D^2+3E^2}}$$

$$+ \frac{1}{4}\Bigg\{-A_z - 2A_z\cos 2\theta$$

$$+ 2\sqrt{\sin^2\theta\left[2\left({A_x}^2 + A_xA_y + {A_y}^2\right) + \left({A_x}^2 + 4A_xA_y + {A_y}^2\right)\cos 2\theta + \sqrt{3}\left({A_x}^2 - {A_y}^2\right)\sin 2\theta\right]}\Bigg\}$$

$$+ \frac{1}{2\Delta}\left\{\left[\frac{\sqrt{3}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{1}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2 + \frac{{A_z}^2}{4}\sin^2 2\theta\right\} - \frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$E_{+\frac{1}{2},+\frac{1}{2}} = -\sqrt{D^2+3E^2} + \frac{g_z\beta B}{2}(\cos^2\theta - 3\sin^2\theta) - \frac{2(g_z\beta B)^2\sin^2\theta\cos^2\theta}{\sqrt{D^2+3E^2}} +$$

$$= \frac{1}{4}\Bigg\{A_z - 2A_z\cos 2\theta$$

$$+ 2\sqrt{\cos^2\theta\left[2\left({A_x}^2 - A_xA_y + {A_y}^2\right) - \left({A_x}^2 - 4A_xA_y + {A_y}^2\right)\cos 2\theta - \sqrt{3}\left({A_x}^2 - {A_y}^2\right)\sin 2\theta\right]}\Bigg\}$$

$$- \frac{1}{2\Delta}\left\{\left[\frac{\sqrt{3}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{1}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \frac{{A_z}^2}{4}\sin^2 2\theta\right\} - \frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$E_{+\frac{1}{2},-\frac{1}{2}}$$

$$= -\sqrt{D^2+3E^2} + \frac{g_z\beta B}{2}(\cos^2\theta - 3\sin^2\theta) - \frac{2(g_z\beta B)^2\sin^2\theta\cos^2\theta}{\sqrt{D^2+3E^2}}$$

$$+ \frac{1}{4}\Bigg\{-A_z + 2A_z\cos 2\theta$$

$$+ 2\sqrt{\cos^2\theta\left[2\left({A_x}^2 + A_xA_y + {A_y}^2\right) - \left({A_x}^2 + 4A_xA_y + {A_y}^2\right)\cos 2\theta - \sqrt{3}\left({A_x}^2 - {A_y}^2\right)\sin 2\theta\right]}\Bigg\}$$

$$- \frac{1}{2\Delta}\left\{\left[\frac{\sqrt{3}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{1}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2 + \frac{{A_z}^2}{4}\sin^2 2\theta\right\} + \frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$E_{-\frac{1}{2},+\frac{1}{2}}$$

$$= -\sqrt{D^2+3E^2} - \frac{g_z\beta B}{2}(\cos^2\theta - 3\sin^2\theta) - \frac{2(g_z\beta B)^2\sin^2\theta\cos^2\theta}{\sqrt{D^2+3E^2}}$$

$$+ \frac{1}{4}\Bigg\{-A_z + 2A_z\cos 2\theta$$

$$- 2\sqrt{\cos^2\theta\left[2\left({A_x}^2 + A_xA_y + {A_y}^2\right) - \left({A_x}^2 + 4A_xA_y + {A_y}^2\right)\cos 2\theta - \sqrt{3}\left({A_x}^2 - {A_y}^2\right)\sin 2\theta\right]}\Bigg\}$$

$$- \frac{1}{2\Delta}\left\{\frac{{A_z}^2}{4}\sin^2 2\theta + \left[\frac{\sqrt{3}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{1}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2\right\} - \frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$E_{-\frac{1}{2},-\frac{1}{2}}$$
$$= -\sqrt{D^2+3E^2} - \frac{g_z\beta B}{2}(\cos^2\theta - 3\sin^2\theta) - \frac{2(g_z\beta B)^2\sin^2\theta\cos^2\theta}{\sqrt{D^2+3E^2}}$$
$$+\frac{1}{4}\left\{A_z - 2A_z\cos 2\theta\right.$$
$$\left.-2\sqrt{\cos^2\theta\left[2\left(A_x{}^2 - A_xA_y + A_y{}^2\right) - \left(A_x{}^2 - 4A_xA_y + A_y{}^2\right)\cos 2\theta - \sqrt{3}\left(A_x{}^2 - A_y{}^2\right)\sin 2\theta\right]}\right\}$$
$$-\frac{1}{2\Delta}\left\{\frac{A_z{}^2}{4}\sin^2 2\theta + \left[\frac{\sqrt{3}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{1}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2\right\} + \frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$
$$E_{-\frac{3}{2},+\frac{1}{2}}$$
$$= \sqrt{D^2+3E^2} - \frac{g_z\beta B}{2}(3\cos^2\theta - \sin^2\theta) + \frac{2(g_z\beta B)^2\sin^2\theta\cos^2\theta}{\sqrt{D^2+3E^2}}$$
$$+\frac{1}{4}\left\{-A_z - 2A_z\cos 2\theta\right.$$
$$\left.-2\sqrt{\sin^2\theta\left[2\left(A_x{}^2 + A_xA_y + A_y{}^2\right) + \left(A_x{}^2 + 4A_xA_y + A_y{}^2\right)\cos 2\theta + \sqrt{3}\left(A_x{}^2 - A_y{}^2\right)\sin 2\theta\right]}\right\}$$
$$+\frac{1}{2\Delta}\left\{\frac{A_z{}^2}{4}\sin^2 2\theta + \left[\frac{\sqrt{3}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{1}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2\right\} + \frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$
$$E_{-\frac{3}{2},-\frac{1}{2}}$$
$$= \sqrt{D^2+3E^2} - \frac{g_z\beta B}{2}(3\cos^2\theta - \sin^2\theta) + \frac{2(g_z\beta B)^2\sin^2\theta\cos^2\theta}{\sqrt{D^2+3E^2}}$$
$$+\frac{1}{4}\left\{A_z + 2A_z\cos 2\theta\right.$$
$$\left.-2\sqrt{\sin^2\theta\left[2\left(A_x{}^2 - A_xA_y + A_y{}^2\right) + \left(A_x{}^2 - 4A_xA_y + A_y{}^2\right)\cos 2\theta + \sqrt{3}\left(A_x{}^2 - A_y{}^2\right)\sin 2\theta\right]}\right\}$$
$$+\frac{1}{2\Delta}\left\{\frac{A_z{}^2}{4}\sin^2 2\theta + \left[\frac{\sqrt{3}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{1}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2\right\} - \frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

2. *I* = 3/2 case

The matrix elements of the hyperfine structure Hamiltonian in the basis of $|M_S, M_I\rangle$ are

$$\langle M_S', M_I'|H_{\mathrm{hfs}}|M_S, M_I\rangle = \begin{cases} \delta_{M_SM_S'}\delta_{M_IM_I'}M_SM_IA_{zz} \\ \frac{1}{16}\delta_{M_SM_S'\mp 1}\delta_{M_IM_I'\mp 1}\sqrt{15-4M_SM_S'}\sqrt{15-4M_IM_I'}\left(A_{xx}-A_{yy}\right) \\ \frac{1}{16}\delta_{M_SM_S'\mp 1}\delta_{M_IM_I'\pm 1}\sqrt{15-4M_SM_S'}\sqrt{15-4M_IM_I'}\left(A_{xx}+A_{yy}\right) \end{cases}$$

The secular equation of the full spin Hamiltonian (including the zero-field splitting, the electron-Zeeman and the hyperfine terms) is factorized into two octic equations, for which we do not have general solutions. Therefore, the extremely exact energies relevant to the equations can be obtained by applying the double perturbation approach.

The matrix representation of the rank-2 ZFS Hamiltonian is the same as that in the case of $I = 1/2$, and thus the energy eigenvalues and the spin eigenstates have already been shown.

Similar to the I = 1/2 case, the set of the spin eigenfunctions, $\left\{\left|\varphi_{M_S,M_I}^{(0)}\right\rangle\right\}$, is divided into two subspaces corresponding to the eigenvalues;

$$\left\{\left|\varphi_{+\frac{3}{2},+\frac{3}{2}}^{(0)}\right\rangle,\left|\varphi_{+\frac{3}{2},+\frac{1}{2}}^{(0)}\right\rangle,\left|\varphi_{+\frac{3}{2},-\frac{1}{2}}^{(0)}\right\rangle,\left|\varphi_{+\frac{3}{2},-\frac{3}{2}}^{(0)}\right\rangle,\left|\varphi_{-\frac{3}{2},+\frac{3}{2}}^{(0)}\right\rangle,\left|\varphi_{-\frac{3}{2},+\frac{1}{2}}^{(0)}\right\rangle,\left|\varphi_{-\frac{3}{2},-\frac{1}{2}}^{(0)}\right\rangle,\left|\varphi_{-\frac{3}{2},-\frac{3}{2}}^{(0)}\right\rangle\right\} \text{ (eigenvalue } \Delta)$$

$$\left\{\left|\varphi_{+\frac{1}{2},+\frac{3}{2}}^{(0)}\right\rangle,\left|\varphi_{+\frac{1}{2},+\frac{1}{2}}^{(0)}\right\rangle,\left|\varphi_{+\frac{1}{2},-\frac{1}{2}}^{(0)}\right\rangle,\left|\varphi_{+\frac{1}{2},-\frac{3}{2}}^{(0)}\right\rangle,\left|\varphi_{-\frac{1}{2},+\frac{3}{2}}^{(0)}\right\rangle,\left|\varphi_{-\frac{1}{2},+\frac{1}{2}}^{(0)}\right\rangle,\left|\varphi_{-\frac{1}{2},-\frac{1}{2}}^{(0)}\right\rangle,\left|\varphi_{-\frac{1}{2},-\frac{3}{2}}^{(0)}\right\rangle\right\} \text{ (eigenvalue } -\Delta)$$

For simplicity, we rewrite the notation of the eigenfunctions as follows for simplicity.

$$\begin{aligned}&\left\{\left|\varphi_{+\frac{3}{2},+\frac{3}{2}}^{(0)}\right\rangle,\left|\varphi_{+\frac{3}{2},+\frac{1}{2}}^{(0)}\right\rangle,\left|\varphi_{+\frac{3}{2},-\frac{1}{2}}^{(0)}\right\rangle,\left|\varphi_{+\frac{3}{2},-\frac{3}{2}}^{(0)}\right\rangle,\left|\varphi_{-\frac{3}{2},+\frac{3}{2}}^{(0)}\right\rangle,\left|\varphi_{-\frac{3}{2},+\frac{1}{2}}^{(0)}\right\rangle,\left|\varphi_{-\frac{3}{2},-\frac{1}{2}}^{(0)}\right\rangle,\left|\varphi_{-\frac{3}{2},-\frac{3}{2}}^{(0)}\right\rangle\right\}\\&\qquad\to\{|+,1\rangle,|+,2\rangle,|+,3\rangle,|+,4\rangle,|+,5\rangle,|+,6\rangle,|+,7\rangle,|+,8\rangle\}\end{aligned}$$

$$\begin{aligned}&\left\{\left|\varphi_{+\frac{1}{2},+\frac{3}{2}}^{(0)}\right\rangle,\left|\varphi_{+\frac{1}{2},+\frac{1}{2}}^{(0)}\right\rangle,\left|\varphi_{+\frac{1}{2},-\frac{1}{2}}^{(0)}\right\rangle,\left|\varphi_{+\frac{1}{2},-\frac{3}{2}}^{(0)}\right\rangle,\left|\varphi_{-\frac{1}{2},+\frac{3}{2}}^{(0)}\right\rangle,\left|\varphi_{-\frac{1}{2},+\frac{1}{2}}^{(0)}\right\rangle,\left|\varphi_{-\frac{1}{2},-\frac{1}{2}}^{(0)}\right\rangle,\left|\varphi_{-\frac{1}{2},-\frac{3}{2}}^{(0)}\right\rangle\right\}\\&\qquad\to\{|-,1\rangle,|-,2\rangle,|-,3\rangle,|-,4\rangle,|-,5\rangle,|-,6\rangle,|-,7\rangle,|-,8\rangle\}\end{aligned}$$

According to the degenerate perturbation theory, similarly to the I = 1/2 case the first-order corrections for the energies of the hyperfine structure Hamiltonian which belong to the group of the positive eigenenergy ($\varepsilon_{+,\alpha}^{(01)}$ $(\alpha = 1,\cdots,8)$) are the eigenvalues of the 8 × 8 matrix, which can be divided into two 4 × 4 matrixes represented as follows:

$$H_{\mathrm{hfs}}^{+,1}=\begin{pmatrix}\langle+,1|H_{\mathrm{hfs}}|+,1\rangle & \langle+,1|H_{\mathrm{hfs}}|+,3\rangle & \langle+,1|H_{\mathrm{hfs}}|+,6\rangle & \langle+,1|H_{\mathrm{hfs}}|+,8\rangle\\ \langle+,3|H_{\mathrm{hfs}}|+,1\rangle & \langle+,3|H_{\mathrm{hfs}}|+,3\rangle & \langle+,3|H_{\mathrm{hfs}}|+,6\rangle & \langle+,3|H_{\mathrm{hfs}}|+,8\rangle\\ \langle+,6|H_{\mathrm{hfs}}|+,1\rangle & \langle+,6|H_{\mathrm{hfs}}|+,3\rangle & \langle+,6|H_{\mathrm{hfs}}|+,6\rangle & \langle+,6|H_{\mathrm{hfs}}|+,8\rangle\\ \langle+,8|H_{\mathrm{hfs}}|+,1\rangle & \langle+,8|H_{\mathrm{hfs}}|+,3\rangle & \langle+,8|H_{\mathrm{hfs}}|+,6\rangle & \langle+,8|H_{\mathrm{hfs}}|+,8\rangle\end{pmatrix}$$

$$H_{\mathrm{hfs}}^{+,2}=\begin{pmatrix}\langle+,2|H_{\mathrm{hfs}}|+,2\rangle & \langle+,2|H_{\mathrm{hfs}}|+,4\rangle & \langle+,2|H_{\mathrm{hfs}}|+,5\rangle & \langle+,2|H_{\mathrm{hfs}}|+,7\rangle\\ \langle+,4|H_{\mathrm{hfs}}|+,2\rangle & \langle+,4|H_{\mathrm{hfs}}|+,4\rangle & \langle+,4|H_{\mathrm{hfs}}|+,5\rangle & \langle+,4|H_{\mathrm{hfs}}|+,7\rangle\\ \langle+,5|H_{\mathrm{hfs}}|+,2\rangle & \langle+,5|H_{\mathrm{hfs}}|+,4\rangle & \langle+,5|H_{\mathrm{hfs}}|+,5\rangle & \langle+,5|H_{\mathrm{hfs}}|+,7\rangle\\ \langle+,7|H_{\mathrm{hfs}}|+,2\rangle & \langle+,7|H_{\mathrm{hfs}}|+,4\rangle & \langle+,7|H_{\mathrm{hfs}}|+,5\rangle & \langle+,7|H_{\mathrm{hfs}}|+,7\rangle\end{pmatrix}$$

where each matrix element can be calculated as

$$\langle+,1|H_{\mathrm{hfs}}|+,1\rangle=\langle+,8|H_{\mathrm{hfs}}|+,8\rangle=\frac{3A_z}{4}(1+2\cos 2\theta)$$

$$\langle+,2|H_{\mathrm{hfs}}|+,2\rangle=\langle+,7|H_{\mathrm{hfs}}|+,7\rangle=\frac{A_z}{4}(1+2\cos 2\theta)$$

$$\langle+,3|H_{\mathrm{hfs}}|+,3\rangle=\langle+,6|H_{\mathrm{hfs}}|+,6\rangle=-\frac{A_z}{4}(1+2\cos 2\theta)$$

$$\langle+,4|H_{\mathrm{hfs}}|+,4\rangle=\langle+,5|H_{\mathrm{hfs}}|+,5\rangle=-\frac{3A_z}{4}(1+2\cos 2\theta)$$

$$\begin{aligned}\langle+,1|H_{\mathrm{hfs}}|+,6\rangle=\langle+,6|H_{\mathrm{hfs}}|+,1\rangle&=\langle+,3|H_{\mathrm{hfs}}|+,8\rangle=\langle+,8|H_{\mathrm{hfs}}|+,3\rangle\\&=\frac{1}{2}\sin\theta\left[3\left(A_x-A_y\right)\cos\theta+\sqrt{3}\left(A_x+A_y\right)\sin\theta\right]\end{aligned}$$

$$\langle +,2|H_{\mathrm{hfs}}|+,5\rangle = \langle +,5|H_{\mathrm{hfs}}|+,2\rangle = \langle +,4|H_{\mathrm{hfs}}|+,7\rangle = \langle +,7|H_{\mathrm{hfs}}|+,4\rangle$$
$$= \frac{1}{2}\sin\theta\left[3\left(A_x + A_y\right)\cos\theta + \sqrt{3}\left(A_x - A_y\right)\sin\theta\right]$$
$$\langle +,2|H_{\mathrm{hfs}}|+,7\rangle = \langle +,7|H_{\mathrm{hfs}}|+,2\rangle = \sin\theta\left[\sqrt{3}\left(A_x - A_y\right)\cos\theta + \left(A_x + A_y\right)\sin\theta\right]$$
$$\langle +,3|H_{\mathrm{hfs}}|+,6\rangle = \langle +,6|H_{\mathrm{hfs}}|+,3\rangle = \sin\theta\left[\sqrt{3}\left(A_x + A_y\right)\cos\theta + \left(A_x - A_y\right)\sin\theta\right]$$

The other elements are zero.

$$H_{\mathrm{hfs}}^{+,1} = \begin{pmatrix} \langle +,1|H_{\mathrm{hfs}}|+,1\rangle & 0 & \langle +,1|H_{\mathrm{hfs}}|+,6\rangle & 0 \\ 0 & \langle +,3|H_{\mathrm{hfs}}|+,3\rangle & \langle +,3|H_{\mathrm{hfs}}|+,6\rangle & \langle +,3|H_{\mathrm{hfs}}|+,8\rangle \\ \langle +,6|H_{\mathrm{hfs}}|+,1\rangle & \langle +,6|H_{\mathrm{hfs}}|+,3\rangle & \langle +,6|H_{\mathrm{hfs}}|+,6\rangle & 0 \\ 0 & \langle +,8|H_{\mathrm{hfs}}|+,3\rangle & 0 & \langle +,8|H_{\mathrm{hfs}}|+,8\rangle \end{pmatrix}$$
$$H_{\mathrm{hfs}}^{+,2} = \begin{pmatrix} \langle +,2|H_{\mathrm{hfs}}|+,2\rangle & 0 & \langle +,2|H_{\mathrm{hfs}}|+,5\rangle & \langle +,2|H_{\mathrm{hfs}}|+,7\rangle \\ 0 & \langle +,4|H_{\mathrm{hfs}}|+,4\rangle & 0 & \langle +,4|H_{\mathrm{hfs}}|+,7\rangle \\ \langle +,5|H_{\mathrm{hfs}}|+,2\rangle & 0 & \langle +,5|H_{\mathrm{hfs}}|+,5\rangle & 0 \\ \langle +,7|H_{\mathrm{hfs}}|+,2\rangle & \langle +,7|H_{\mathrm{hfs}}|+,4\rangle & 0 & \langle +,7|H_{\mathrm{hfs}}|+,7\rangle \end{pmatrix}$$

while the first-order corrections for the energy, which belong to the group of the negative eigenenergy ($\varepsilon_{-,\alpha}^{(01)}$ $(\alpha = 1,\cdots,8)$), are the eigenvalues of the 4×4 matrixes represented as follows:

$$H_{\mathrm{hfs}}^{-,1} = \begin{pmatrix} \langle -,1|H_{\mathrm{hfs}}|-,1\rangle & \langle -,1|H_{\mathrm{hfs}}|-,3\rangle & \langle -,1|H_{\mathrm{hfs}}|-,6\rangle & \langle -,1|H_{\mathrm{hfs}}|-,8\rangle \\ \langle -,3|H_{\mathrm{hfs}}|-,1\rangle & \langle -,3|H_{\mathrm{hfs}}|-,3\rangle & \langle -,3|H_{\mathrm{hfs}}|-,6\rangle & \langle -,3|H_{\mathrm{hfs}}|-,8\rangle \\ \langle -,6|H_{\mathrm{hfs}}|-,1\rangle & \langle -,6|H_{\mathrm{hfs}}|-,3\rangle & \langle -,6|H_{\mathrm{hfs}}|-,6\rangle & \langle -,6|H_{\mathrm{hfs}}|-,8\rangle \\ \langle -,8|H_{\mathrm{hfs}}|-,1\rangle & \langle -,8|H_{\mathrm{hfs}}|-,3\rangle & \langle -,8|H_{\mathrm{hfs}}|-,6\rangle & \langle -,8|H_{\mathrm{hfs}}|-,8\rangle \end{pmatrix}$$
$$H_{\mathrm{hfs}}^{-,2} = \begin{pmatrix} \langle -,2|H_{\mathrm{hfs}}|-,2\rangle & \langle -,2|H_{\mathrm{hfs}}|-,4\rangle & \langle -,2|H_{\mathrm{hfs}}|-,5\rangle & \langle -,2|H_{\mathrm{hfs}}|-,7\rangle \\ \langle -,4|H_{\mathrm{hfs}}|-,2\rangle & \langle -,4|H_{\mathrm{hfs}}|-,4\rangle & \langle -,4|H_{\mathrm{hfs}}|-,5\rangle & \langle -,4|H_{\mathrm{hfs}}|-,7\rangle \\ \langle -,5|H_{\mathrm{hfs}}|-,2\rangle & \langle -,5|H_{\mathrm{hfs}}|-,4\rangle & \langle -,5|H_{\mathrm{hfs}}|-,5\rangle & \langle -,5|H_{\mathrm{hfs}}|-,7\rangle \\ \langle -,7|H_{\mathrm{hfs}}|-,2\rangle & \langle -,7|H_{\mathrm{hfs}}|-,4\rangle & \langle -,7|H_{\mathrm{hfs}}|-,5\rangle & \langle -,7|H_{\mathrm{hfs}}|-,7\rangle \end{pmatrix}$$

$$\langle -,1|H_{\mathrm{hfs}}|-,1\rangle = \langle -,8|H_{\mathrm{hfs}}|-,8\rangle = \frac{3A_z}{4}(-1 + 2\cos 2\theta)$$
$$\langle -,2|H_{\mathrm{hfs}}|-,2\rangle = \langle -,7|H_{\mathrm{hfs}}|-,7\rangle = \frac{A_z}{4}(-1 + 2\cos 2\theta)$$
$$\langle -,3|H_{\mathrm{hfs}}|-,3\rangle = \langle -,6|H_{\mathrm{hfs}}|-,6\rangle = \frac{A_z}{4}(1 - 2\cos 2\theta)$$
$$\langle -,4|H_{\mathrm{hfs}}|-,4\rangle = \langle -,5|H_{\mathrm{hfs}}|-,5\rangle = \frac{3A_z}{4}(1 - 2\cos 2\theta)$$
$$\langle -,1|H_{\mathrm{hfs}}|-,6\rangle = \langle -,6|H_{\mathrm{hfs}}|-,1\rangle = \langle -,3|H_{\mathrm{hfs}}|-,8\rangle = \langle -,8|H_{\mathrm{hfs}}|-,3\rangle$$
$$= \frac{1}{2}\cos\theta\left[\sqrt{3}\left(A_x - A_y\right)\cos\theta - 3\left(A_x + A_y\right)\sin\theta\right]$$
$$\langle -,2|H_{\mathrm{hfs}}|-,5\rangle = \langle -,5|H_{\mathrm{hfs}}|-,2\rangle = \langle -,4|H_{\mathrm{hfs}}|-,7\rangle = \langle -,7|H_{\mathrm{hfs}}|-,4\rangle$$
$$= \frac{1}{2}\cos\theta\left[\sqrt{3}\left(A_x + A_y\right)\cos\theta - 3\left(A_x - A_y\right)\sin\theta\right]$$
$$\langle -,2|H_{\mathrm{hfs}}|-,7\rangle = \langle -,7|H_{\mathrm{hfs}}|-,2\rangle = \cos\theta\left[\left(A_x - A_y\right)\cos\theta - \sqrt{3}\left(A_x + A_y\right)\sin\theta\right]$$
$$\langle -,3|H_{\mathrm{hfs}}|-,6\rangle = \langle -,6|H_{\mathrm{hfs}}|-,3\rangle = \cos\theta\left[\left(A_x + A_y\right)\cos\theta - \sqrt{3}\left(A_x - A_y\right)\sin\theta\right]$$

$$H_{\mathrm{hfs}}^{-,1} = \begin{pmatrix} \langle -,1|H_{\mathrm{hfs}}|-,1\rangle & 0 & \langle -,1|H_{\mathrm{hfs}}|-,6\rangle & 0 \\ 0 & \langle -,3|H_{\mathrm{hfs}}|-,3\rangle & \langle -,3|H_{\mathrm{hfs}}|-,6\rangle & \langle -,3|H_{\mathrm{hfs}}|-,8\rangle \\ \langle -,6|H_{\mathrm{hfs}}|-,1\rangle & \langle -,6|H_{\mathrm{hfs}}|-,3\rangle & \langle -,6|H_{\mathrm{hfs}}|-,6\rangle & 0 \\ 0 & \langle -,8|H_{\mathrm{hfs}}|-,3\rangle & 0 & \langle -,8|H_{\mathrm{hfs}}|-,8\rangle \end{pmatrix}$$

$$H_{\mathrm{hfs}}^{-,2} = \begin{pmatrix} \langle -,2|H_{\mathrm{hfs}}|-,2\rangle & 0 & \langle -,2|H_{\mathrm{hfs}}|-,5\rangle & \langle -,2|H_{\mathrm{hfs}}|-,7\rangle \\ 0 & \langle -,4|H_{\mathrm{hfs}}|-,4\rangle & 0 & \langle -,4|H_{\mathrm{hfs}}|-,7\rangle \\ \langle -,5|H_{\mathrm{hfs}}|-,2\rangle & 0 & \langle -,5|H_{\mathrm{hfs}}|-,5\rangle & 0 \\ \langle -,7|H_{\mathrm{hfs}}|-,2\rangle & \langle -,7|H_{\mathrm{hfs}}|-,4\rangle & 0 & \langle -,7|H_{\mathrm{hfs}}|-,7\rangle \end{pmatrix}$$

The secular quartic equations can be factorized to two quadratic equations. The first-order corrections are given as the solutions of the following quadratic equation:

$$x^2 + a_i x + b_i = 0 \ (i = 1, \cdots, 8)$$

In the equation, i = 1 and 2, 3 and 4, 5 and 6, and 7 and 8 come from $H_{\mathrm{hfs}}^{+,1}$, $H_{\mathrm{hfs}}^{+,2}$, $H_{\mathrm{hfs}}^{-,1}$, $H_{\mathrm{hfs}}^{-,2}$, respectively. The solutions of the quadratic equation above are

$$x = \frac{-a_i \pm \sqrt{{a_i}^2 - 4b_i}}{2} \ (i = 1, \cdots, 8)$$

The coefficients of the quadratic equation are in the following:

$$a_1 = \frac{1}{2}\left(A_x - A_y - A_z\right) - \frac{1}{2}\left(A_x - A_y + 2A_z\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(A_x + A_y\right)\sin 2\theta$$

$$\begin{aligned} b_1 = &-\frac{9}{16}\left({A_x}^2 + {A_y}^2 + {A_z}^2\right) + \frac{3}{8}\left({A_x}^2 + 2A_xA_y + {A_y}^2 - A_xA_z + A_yA_z - 2{A_z}^2\right)\cos 2\theta \\ &+ \frac{3}{16}\left({A_x}^2 - 4A_xA_y + {A_y}^2 + 2A_xA_z - 2A_yA_z - 2{A_z}^2\right)\cos 4\theta \\ &- \frac{3\sqrt{3}}{8}\left({A_x}^2 - {A_y}^2 + A_xA_z + A_yA_z\right)\sin 2\theta \\ &+ \frac{3\sqrt{3}}{16}\left({A_x}^2 - {A_y}^2 - 2A_xA_z - 2A_yA_z\right)\sin 4\theta \end{aligned}$$

$$a_2 = \frac{1}{2}\left(-A_x + A_y - A_z\right) + \frac{1}{2}\left(A_x - A_y - 2A_z\right)\cos 2\theta - \frac{1}{2}\sqrt{3}\left(A_x + A_y\right)\sin 2\theta$$

$$\begin{aligned} b_2 = &-\frac{9}{16}\left({A_x}^2 + {A_y}^2 + {A_z}^2\right) + \frac{3}{8}\left({A_x}^2 + 2A_xA_y + {A_y}^2 + A_xA_z - A_yA_z - 2{A_z}^2\right)\cos 2\theta \\ &+ \frac{3}{16}\left({A_x}^2 - 4A_xA_y + {A_y}^2 - 2A_xA_z + 2A_yA_z - 2{A_z}^2\right)\cos 4\theta \\ &- \frac{3\sqrt{3}}{8}\left({A_x}^2 - {A_y}^2 - A_xA_z - A_yA_z\right)\sin 2\theta \\ &+ \frac{3\sqrt{3}}{16}\left({A_x}^2 - {A_y}^2 + 2A_xA_z + 2A_yA_z\right)\sin 4\theta \end{aligned}$$

$$a_3 = \frac{1}{2}\left(A_x + A_y + A_z\right) - \frac{1}{2}\left(A_x + A_y - 2A_z\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(A_x - A_y\right)\sin 2\theta$$

$$b_3 = -\frac{9}{16}\left({A_x}^2 + {A_y}^2 + {A_z}^2\right) + \frac{3}{8}\left({A_x}^2 - 2A_xA_y + {A_y}^2 + A_xA_z + A_yA_z - 2{A_z}^2\right)\cos 2\theta$$

$$+\frac{3}{16}\left({A_x}^2 + 4A_xA_y + {A_y}^2 - 2A_xA_z - 2A_yA_z - 2{A_z}^2\right)\cos 4\theta$$

$$-\frac{3\sqrt{3}}{8}\left({A_x}^2 - {A_y}^2 - A_xA_z + A_yA_z\right)\sin 2\theta$$

$$+\frac{3\sqrt{3}}{16}\left({A_x}^2 - {A_y}^2 + 2A_xA_z - 2A_yA_z\right)\sin 4\theta$$

$$a_4 = \frac{1}{2}\left(-A_x - A_y + A_z\right) + \frac{1}{2}\left(A_x + A_y + 2A_z\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(-A_x + A_y\right)\sin 2\theta$$

$$b_4 = -\frac{9}{16}\left({A_x}^2 + {A_y}^2 + {A_z}^2\right) + \frac{3}{8}\left({A_x}^2 - 2A_xA_y + {A_y}^2 - A_xA_z - A_yA_z - 2{A_z}^2\right)\cos 2\theta$$

$$+\frac{3}{16}\left({A_x}^2 + 4A_xA_y + {A_y}^2 + 2A_xA_z + 2A_yA_z - 2{A_z}^2\right)\cos 4\theta$$

$$-\frac{3\sqrt{3}}{8}\left({A_x}^2 - {A_y}^2 + A_xA_z - A_yA_z\right)\sin 2\theta$$

$$+\frac{3\sqrt{3}}{16}\left({A_x}^2 - {A_y}^2 - 2A_xA_z + 2A_yA_z\right)\sin 4\theta$$

Actually, the coefficients for $i = 2$, 3 and 4 can be obtained from $\{a_1, b_1, c_1, d_1\}$ with replacing A_x and A_y to $-A_x$ and $-A_y$, A_x and A_z to $-A_x$ and $-A_z$, and A_y and A_z to $-A_y$ and $-A_z$, respectively.

For the negative counterpart:

$$a_5 = \frac{1}{2}\left(-A_x - A_y + A_z\right) - \frac{1}{2}\left(A_x + A_y + 2A_z\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(A_x - A_y\right)\sin 2\theta$$

$$b_5 = -\frac{9}{16}\left({A_x}^2 + {A_y}^2 + {A_z}^2\right) - \frac{3}{8}\left({A_x}^2 - 2A_xA_y + {A_y}^2 - A_xA_z - A_yA_z - 2{A_z}^2\right)\cos 2\theta$$

$$+\frac{3}{16}\left({A_x}^2 + 4A_xA_y + {A_y}^2 + 2A_xA_z + 2A_yA_z - 2{A_z}^2\right)\cos 4\theta$$

$$+\frac{3\sqrt{3}}{8}\left({A_x}^2 - {A_y}^2 + A_xA_z - A_yA_z\right)\sin 2\theta$$

$$+\frac{3\sqrt{3}}{16}\left({A_x}^2 - {A_y}^2 - 2A_xA_z + 2A_yA_z\right)\sin 4\theta$$

$$a_6 = \frac{1}{2}\left(A_x + A_y + A_z\right) + \frac{1}{2}\left(A_x + A_y - 2A_z\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(-A_x + A_y\right)\sin 2\theta$$

$$b_6 = -\frac{9}{16}\left({A_x}^2 + {A_y}^2 + {A_z}^2\right) + \frac{3}{8}\left(-{A_x}^2 + 2A_xA_y - {A_y}^2 - A_xA_z - A_yA_z + 2{A_z}^2\right)\cos 2\theta$$

$$+\frac{3}{16}\left({A_x}^2 + 4A_xA_y + {A_y}^2 - 2A_xA_z - 2A_yA_z - 2{A_z}^2\right)\cos 4\theta$$

$$+\frac{3\sqrt{3}}{8}\left({A_x}^2 - {A_y}^2 - A_xA_z + A_yA_z\right)\sin 2\theta$$

$$+\frac{3\sqrt{3}}{16}\left({A_x}^2 - {A_y}^2 + 2A_xA_z - 2A_yA_z\right)\sin 4\theta$$

$$a_7 = \frac{1}{2}\left(-A_x + A_y - A_z\right) + \frac{1}{2}\left(-A_x + A_y + 2A_z\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(A_x + A_y\right)\sin 2\theta$$

$$b_7 = -\frac{9}{16}\left({A_x}^2 + {A_y}^2 + {A_z}^2\right) + \frac{3}{8}\left(-{A_x}^2 - 2A_xA_y - {A_y}^2 - A_xA_z + A_yA_z + 2{A_z}^2\right)\cos 2\theta$$

$$+\frac{3}{16}\left({A_x}^2 - 4A_xA_y + {A_y}^2 - 2A_xA_z + 2A_yA_z - 2{A_z}^2\right)\cos 4\theta$$

$$+\frac{3\sqrt{3}}{8}\left({A_x}^2 - {A_y}^2 - A_xA_z - A_yA_z\right)\sin 2\theta$$

$$+\frac{3\sqrt{3}}{16}\left({A_x}^2 - {A_y}^2 + 2A_xA_z + 2A_yA_z\right)\sin 4\theta$$

$$a_8 = \frac{1}{2}\left(A_x - A_y - A_z\right) + \frac{1}{2}\left(A_x - A_y + 2A_z\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(-A_x - A_y\right)\sin 2\theta$$

$$b_8 = -\frac{9}{16}\left({A_x}^2 + {A_y}^2 + {A_z}^2\right) + \frac{3}{8}\left(-{A_x}^2 - 2A_xA_y - {A_y}^2 + A_xA_z - A_yA_z + 2{A_z}^2\right)\cos 2\theta$$

$$+\frac{3}{16}\left({A_x}^2 - 4A_xA_y + {A_y}^2 + 2A_xA_z - 2A_yA_z - 2{A_z}^2\right)\cos 4\theta$$

$$+\frac{3\sqrt{3}}{8}\left({A_x}^2 - {A_y}^2 + A_xA_z + A_yA_z\right)\sin 2\theta$$

$$+\frac{3\sqrt{3}}{16}\left({A_x}^2 - {A_y}^2 - 2A_xA_z - 2A_yA_z\right)\sin 4\theta$$

Similarly, the coefficients for $i = 6$, 7 and 8 can be obtained from $\{a_5, b_5, c_5, d_5\}$ with replacing A_x and A_y to $-A_x$ and $-A_y$, A_x and A_z to $-A_x$ and $-A_z$, and A_y and A_z to $-A_y$ and $-A_z$, respectively.

The second-order corrections for the energy $\varepsilon_{\sigma,\alpha}^{(02)}$ $(\sigma = +,-;\ \alpha = 1,2,3,4)$ can be written as

$$\varepsilon_{\pm,\alpha}^{(02)} = \sum_{\beta=1}^{4} \frac{|\langle\pm,\alpha|H_{\mathrm{hfs}}|\mp,\beta\rangle|^2}{\varepsilon_{\pm}^{(0)} - \varepsilon_{\mp}^{(0)}}$$

Both the upper and lower signs should be chosen in the double sign. The non-zero matrix elements of H_{hfs} expanded to the basis belonging different eigenspaces are in the following;

$$\langle +,1|H_{\mathrm{hfs}}|-,2\rangle = \langle -,2|H_{\mathrm{hfs}}|+,1\rangle = \langle +,3|H_{\mathrm{hfs}}|-,4\rangle = \langle -,4|H_{\mathrm{hfs}}|+,3\rangle = \langle -,5|H_{\mathrm{hfs}}|+,6\rangle = \langle +,6|H_{\mathrm{hfs}}|-,5\rangle$$

$$= \langle -,7|H_{\mathrm{hfs}}|+,8\rangle = \langle +,8|H_{\mathrm{hfs}}|-,7\rangle = \frac{3}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{3}}{4}\left(A_x + A_y\right)\sin 2\theta$$

$$\langle +,2|H_{\mathrm{hfs}}|-,1\rangle = \langle -,1|H_{\mathrm{hfs}}|+,2\rangle = \langle +,4|H_{\mathrm{hfs}}|-,3\rangle = \langle -,3|H_{\mathrm{hfs}}|+,4\rangle = \langle -,6|H_{\mathrm{hfs}}|+,5\rangle = \langle +,5|H_{\mathrm{hfs}}|-,6\rangle$$

$$= \langle -,8|H_{\mathrm{hfs}}|+,7\rangle = \langle +,7|H_{\mathrm{hfs}}|-,8\rangle = \frac{3}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{3}}{4}\left(A_x - A_y\right)\sin 2\theta$$

$$\langle +,2|H_{\mathrm{hfs}}|-,3\rangle = \langle -,3|H_{\mathrm{hfs}}|+,2\rangle = \langle -,6|H_{\mathrm{hfs}}|+,7\rangle = \langle +,7|H_{\mathrm{hfs}}|-,6\rangle$$

$$= \frac{\sqrt{3}}{2}\left(A_x - A_y\right)\cos 2\theta + \frac{1}{2}\left(A_x + A_y\right)\sin 2\theta$$

$$\langle +,3|H_{\mathrm{hfs}}|-,2\rangle = \langle -,2|H_{\mathrm{hfs}}|+,3\rangle = \langle -,7|H_{\mathrm{hfs}}|+,6\rangle = \langle +,6|H_{\mathrm{hfs}}|-,7\rangle$$

$$= \frac{\sqrt{3}}{2}\left(A_x + A_y\right)\cos 2\theta + \frac{1}{2}\left(A_x - A_y\right)\sin 2\theta$$

$$\langle +,1|H_{\mathrm{hfs}}|-,5\rangle = \langle -,5|H_{\mathrm{hfs}}|+,1\rangle = \langle -,4|H_{\mathrm{hfs}}|+,8\rangle = \langle +,8|H_{\mathrm{hfs}}|-,4\rangle = -\frac{3}{2}A_z \sin 2\theta$$

$$\langle +,2|H_{\mathrm{hfs}}|-,6\rangle = \langle -,6|H_{\mathrm{hfs}}|+,2\rangle = \langle -,3|H_{\mathrm{hfs}}|+,7\rangle = \langle +,7|H_{\mathrm{hfs}}|-,3\rangle = -\frac{A_z}{2}\sin 2\theta$$

$$\langle +,3|H_{\mathrm{hfs}}|-,7\rangle = \langle -,7|H_{\mathrm{hfs}}|+,3\rangle = \langle -,2|H_{\mathrm{hfs}}|+,6\rangle = \langle +,6|H_{\mathrm{hfs}}|-,2\rangle = \frac{A_z}{2}\sin 2\theta$$

$$\langle +,4|H_{\mathrm{hfs}}|-,8\rangle = \langle -,8|H_{\mathrm{hfs}}|+,4\rangle = \langle -,1|H_{\mathrm{hfs}}|+,5\rangle = \langle +,5|H_{\mathrm{hfs}}|-,1\rangle = \frac{3}{2}A_z \sin 2\theta$$

Therefore,

$$\varepsilon_{+,1}^{(02)} = \frac{|\langle -,2|H_{\mathrm{hfs}}|+,1\rangle|^2 + |\langle -,5|H_{\mathrm{hfs}}|+,1\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{3}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{3}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \frac{9}{4}{A_z}^2 \sin^2 2\theta\right\}$$

$$\varepsilon_{+,2}^{(02)} = \frac{|\langle -,1|H_{\mathrm{hfs}}|+,2\rangle|^2 + |\langle -,3|H_{\mathrm{hfs}}|+,2\rangle|^2 + |\langle -,6|H_{\mathrm{hfs}}|+,2\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{3}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{3}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2 \right.$$

$$\left. + \left[\frac{\sqrt{3}}{2}\left(A_x - A_y\right)\cos 2\theta + \frac{1}{2}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \frac{{A_z}^2}{4}\sin^2 2\theta\right\}$$

$$\varepsilon_{+,3}^{(02)} = \frac{|\langle -,2|H_{\mathrm{hfs}}|+,3\rangle|^2 + |\langle -,4|H_{\mathrm{hfs}}|+,3\rangle|^2 + |\langle -,7|H_{\mathrm{hfs}}|+,3\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{\sqrt{3}}{2}\left(A_x + A_y\right)\cos 2\theta + \frac{1}{2}\left(A_x - A_y\right)\sin 2\theta\right]^2 \right.$$

$$\left. + \left[\frac{3}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{3}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \frac{{A_z}^2}{4}\sin^2 2\theta\right\}$$

$$\varepsilon_{+,4}^{(02)} = \frac{|\langle -,3|H_{\mathrm{hfs}}|+,4\rangle|^2 + |\langle -,8|H_{\mathrm{hfs}}|+,4\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{3}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{3}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2 + \frac{9}{4}{A_z}^2 \sin^2 2\theta\right\}$$

$$\varepsilon_{+,5}^{(02)} = \frac{|\langle -,1|H_{\mathrm{hfs}}|+,5\rangle|^2 + |\langle -,6|H_{\mathrm{hfs}}|+,5\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{9}{4}A_z \sin^2 2\theta + \left[\frac{3}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{3}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{+,6}^{(02)} = \frac{|\langle -,2|H_{\mathrm{hfs}}|+,6\rangle|^2 + |\langle -,5|H_{\mathrm{hfs}}|+,6\rangle|^2 + |\langle -,7|H_{\mathrm{hfs}}|+,6\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{{A_z}^2}{4}\sin^2 2\theta + \left[\frac{3}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{3}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2\right.$$

$$\left. + \left[\frac{\sqrt{3}}{2}\left(A_x + A_y\right)\cos 2\theta + \frac{1}{2}\left(A_x - A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{+,7}^{(02)} = \frac{|\langle -,3|H_{\mathrm{hfs}}|+,7\rangle|^2 + |\langle -,6|H_{\mathrm{hfs}}|+,7\rangle|^2 + |\langle -,8|H_{\mathrm{hfs}}|+,7\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{{A_z}^2}{4}\sin^2 2\theta + \left[\frac{\sqrt{3}}{2}\left(A_x - A_y\right)\cos 2\theta + \frac{1}{2}\left(A_x + A_y\right)\sin 2\theta\right]^2\right.$$

$$\left. + \left[\frac{3}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{3}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{+,8}^{(02)} = \frac{|\langle -,4|H_{\mathrm{hfs}}|+,8\rangle|^2 + |\langle -,7|H_{\mathrm{hfs}}|+,8\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{9}{4}{A_z}^2 \sin^2 2\theta + \left[\frac{3}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{3}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{-,1}^{(02)} = \frac{|\langle +,2|H_{\mathrm{hfs}}|-,1\rangle|^2 + |\langle +,5|H_{\mathrm{hfs}}|-,1\rangle|^2}{-2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{3}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{3}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2 + \frac{9}{4}{A_z}^2 \sin^2 2\theta\right\}$$

$$\varepsilon_{-,2}^{(02)} = \frac{|\langle +,1|H_{\mathrm{hfs}}|-,2\rangle|^2 + |\langle +,3|H_{\mathrm{hfs}}|-,2\rangle|^2 + |\langle +,6|H_{\mathrm{hfs}}|-,2\rangle|^2}{-2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{3}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{3}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2\right.$$

$$\left. + \left[\frac{\sqrt{3}}{2}\left(A_x + A_y\right)\cos 2\theta + \frac{1}{2}\left(A_x - A_y\right)\sin 2\theta\right]^2 + \frac{{A_z}^2}{4}\sin^2 2\theta\right\}$$

$$\varepsilon_{-,3}^{(02)} = \frac{|\langle +,2|H_{\mathrm{hfs}}|-,3\rangle|^2 + |\langle +,4|H_{\mathrm{hfs}}|-,3\rangle|^2 + |\langle +,7|H_{\mathrm{hfs}}|-,3\rangle|^2}{-2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{\sqrt{3}}{2}(A_x - A_y)\cos 2\theta + \frac{1}{2}(A_x + A_y)\sin 2\theta\right]^2 + \left[\frac{3}{4}(A_x + A_y)\cos 2\theta + \frac{\sqrt{3}}{4}(A_x - A_y)\sin 2\theta\right]^2 + \frac{{A_z}^2}{4}\sin^2 2\theta\right\}$$

$$\varepsilon_{-,4}^{(02)} = \frac{|\langle +,3|H_{\mathrm{hfs}}|-,4\rangle|^2 + |\langle +,8|H_{\mathrm{hfs}}|-,4\rangle|^2}{-2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{3}{4}(A_x - A_y)\cos 2\theta + \frac{\sqrt{3}}{4}(A_x + A_y)\sin 2\theta\right]^2 + \frac{9}{4}{A_z}^2\sin^2 2\theta\right\}$$

$$\varepsilon_{-,5}^{(02)} = \frac{|\langle +,1|H_{\mathrm{hfs}}|-,5\rangle|^2 + |\langle +,6|H_{\mathrm{hfs}}|-,5\rangle|^2}{-2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{9}{4}{A_z}^2\sin^2 2\theta + \left[\frac{3}{4}(A_x - A_y)\cos 2\theta + \frac{\sqrt{3}}{4}(A_x + A_y)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{-,6}^{(02)} = \frac{|\langle +,2|H_{\mathrm{hfs}}|-,6\rangle|^2 + |\langle +,5|H_{\mathrm{hfs}}|-,6\rangle|^2 + |\langle +,7|H_{\mathrm{hfs}}|-,6\rangle|^2}{-2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{{A_z}^2}{4}\sin^2 2\theta + \left[\frac{3}{4}(A_x + A_y)\cos 2\theta + \frac{\sqrt{3}}{4}(A_x - A_y)\sin 2\theta\right]^2 + \left[\frac{\sqrt{3}}{2}(A_x - A_y)\cos 2\theta + \frac{1}{2}(A_x + A_y)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{-,7}^{(02)} = \frac{|\langle +,3|H_{\mathrm{hfs}}|-,7\rangle|^2 + |\langle +,6|H_{\mathrm{hfs}}|-,7\rangle|^2 + |\langle +,8|H_{\mathrm{hfs}}|-,7\rangle|^2}{-2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{{A_z}^2}{4}\sin^2 2\theta + \left[\frac{\sqrt{3}}{2}(A_x + A_y)\cos 2\theta + \frac{1}{2}(A_x - A_y)\sin 2\theta\right]^2 + \left[\frac{3}{4}(A_x - A_y)\cos 2\theta + \frac{\sqrt{3}}{4}(A_x + A_y)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{-,8}^{(02)} = \frac{|\langle +,4|H_{\mathrm{hfs}}|-,8\rangle|^2 + |\langle +,7|H_{\mathrm{hfs}}|-,8\rangle|^2}{-2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{9}{4}{A_z}^2\sin^2 2\theta + \left[\frac{3}{4}(A_x + A_y)\cos 2\theta + \frac{\sqrt{3}}{4}(A_x - A_y)\sin 2\theta\right]^2\right\}$$

Noticeably, the first- and second-order corrections for the energy of the electron Zeeman Hamiltonian are the same as for the $I = 1/2$ case. In order to obtain the cross terms, let us remind the non-diagonal elements for the electron Zeeman Hamiltonian.

$$\langle +,1|H_{\mathrm{eZ}}|-,5\rangle = \langle -,5|H_{\mathrm{eZ}}|+,1\rangle = -g_z\beta B\sin 2\theta$$

$$\langle +,2|H_{\mathrm{eZ}}|-,6\rangle = \langle -,6|H_{\mathrm{eZ}}|+,2\rangle = -g_z\beta B\sin 2\theta$$

$$\langle +,3|H_{\mathrm{eZ}}|-,7\rangle = \langle -,7|H_{\mathrm{eZ}}|+,3\rangle = -g_z\beta B\sin 2\theta$$

$$\langle +,4|H_{\mathrm{eZ}}|-,8\rangle = \langle -,8|H_{\mathrm{eZ}}|+,2\rangle = -g_z\beta B\sin 2\theta$$

$$\langle +,5|H_{\mathrm{eZ}}|-,1\rangle = \langle -,1|H_{\mathrm{eZ}}|+,5\rangle = g_z\beta B\sin 2\theta$$

$$\langle +,6|H_{\mathrm{eZ}}|-,2\rangle = \langle -,2|H_{\mathrm{eZ}}|+,6\rangle = g_z\beta B\sin 2\theta$$

$$\langle +,7|H_{\mathrm{eZ}}|-,3\rangle = \langle -,3|H_{\mathrm{eZ}}|+,7\rangle = g_z\beta B\sin 2\theta$$

$$\langle +,8|H_{\mathrm{eZ}}|-,4\rangle = \langle -,4|H_{\mathrm{eZ}}|+,7\rangle = g_z\beta B\sin 2\theta$$

The cross terms are as follows:

$$\varepsilon_{+,1}^{(11)} = \frac{\langle +,1|H_{\mathrm{eZ}}|-,5\rangle\langle -,5|H_{\mathrm{hfs}}|+,1\rangle + \langle +,1|H_{\mathrm{hfs}}|-,5\rangle\langle -,5|H_{\mathrm{eZ}}|+,1\rangle}{\varepsilon_+^{(0)} - \varepsilon_-^{(0)}} = \frac{3g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,2}^{(11)} = \frac{\langle +,2|H_{\mathrm{eZ}}|-,6\rangle\langle -,6|H_{\mathrm{hfs}}|+,2\rangle + \langle +,2|H_{\mathrm{hfs}}|-,6\rangle\langle -,6|H_{\mathrm{eZ}}|+,2\rangle}{\varepsilon_+^{(0)} - \varepsilon_-^{(0)}} = \frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,3}^{(11)} = \frac{\langle +,3|H_{\mathrm{eZ}}|-,7\rangle\langle -,7|H_{\mathrm{hfs}}|+,3\rangle + \langle +,3|H_{\mathrm{hfs}}|-,7\rangle\langle -,7|H_{\mathrm{eZ}}|+,3\rangle}{\varepsilon_+^{(0)} - \varepsilon_-^{(0)}} = -\frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,4}^{(11)} = \frac{\langle +,4|H_{\mathrm{eZ}}|-,8\rangle\langle -,8|H_{\mathrm{hfs}}|+,4\rangle + \langle +,4|H_{\mathrm{hfs}}|-,8\rangle\langle -,8|H_{\mathrm{eZ}}|+,4\rangle}{\varepsilon_+^{(0)} - \varepsilon_-^{(0)}} = -\frac{3g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,1}^{(11)} = \frac{\langle -,1|H_{\mathrm{eZ}}|+,5\rangle\langle +,5|H_{\mathrm{hfs}}|-,1\rangle + \langle -,1|H_{\mathrm{hfs}}|+,5\rangle\langle +,5|H_{\mathrm{eZ}}|-,1\rangle}{\varepsilon_-^{(0)} - \varepsilon_+^{(0)}} = -\frac{3g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,2}^{(11)} = \frac{\langle -,2|H_{\mathrm{eZ}}|+,6\rangle\langle +,6|H_{\mathrm{hfs}}|-,2\rangle + \langle -,2|H_{\mathrm{hfs}}|+,6\rangle\langle +,6|H_{\mathrm{eZ}}|-,2\rangle}{\varepsilon_-^{(0)} - \varepsilon_+^{(0)}} = -\frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,3}^{(11)} = \frac{\langle -,3|H_{\mathrm{eZ}}|+,7\rangle\langle +,7|H_{\mathrm{hfs}}|-,3\rangle + \langle -,3|H_{\mathrm{hfs}}|+,7\rangle\langle +,7|H_{\mathrm{eZ}}|-,3\rangle}{\varepsilon_-^{(0)} - \varepsilon_+^{(0)}} = \frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,4}^{(11)} = \frac{\langle -,4|H_{\mathrm{eZ}}|+,8\rangle\langle +,8|H_{\mathrm{hfs}}|-,4\rangle + \langle -,4|H_{\mathrm{hfs}}|+,8\rangle\langle +,8|H_{\mathrm{eZ}}|-,4\rangle}{\varepsilon_-^{(0)} - \varepsilon_+^{(0)}} = \frac{3g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,5}^{(11)} = \frac{\langle -,5|H_{\mathrm{eZ}}|+,1\rangle\langle +,1|H_{\mathrm{hfs}}|-,5\rangle + \langle -,5|H_{\mathrm{hfs}}|+,1\rangle\langle +,1|H_{\mathrm{eZ}}|-,5\rangle}{\varepsilon_-^{(0)} - \varepsilon_+^{(0)}} = -\frac{3g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,6}^{(11)} = \frac{\langle -,6|H_{\mathrm{eZ}}|+,2\rangle\langle +,2|H_{\mathrm{hfs}}|-,6\rangle + \langle -,6|H_{\mathrm{hfs}}|+,2\rangle\langle +,2|H_{\mathrm{eZ}}|-,6\rangle}{\varepsilon_-^{(0)} - \varepsilon_+^{(0)}} = -\frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,7}^{(11)} = \frac{\langle -,7|H_{\mathrm{eZ}}|+,3\rangle\langle +,3|H_{\mathrm{hfs}}|-,7\rangle + \langle -,7|H_{\mathrm{hfs}}|+,3\rangle\langle +,3|H_{\mathrm{eZ}}|-,7\rangle}{\varepsilon_-^{(0)} - \varepsilon_+^{(0)}} = \frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,8}^{(11)} = \frac{\langle -,8|H_{\mathrm{eZ}}|+,4\rangle\langle +,4|H_{\mathrm{hfs}}|-,8\rangle + \langle -,8|H_{\mathrm{hfs}}|+,4\rangle\langle +,4|H_{\mathrm{eZ}}|-,8\rangle}{\varepsilon_-^{(0)} - \varepsilon_+^{(0)}} = \frac{3g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,5}^{(11)} = \frac{\langle +,5|H_{\mathrm{eZ}}|-,1\rangle\langle -,1|H_{\mathrm{hfs}}|+,5\rangle + \langle +,5|H_{\mathrm{hfs}}|-,1\rangle\langle -,1|H_{\mathrm{eZ}}|+,5\rangle}{\varepsilon_+^{(0)} - \varepsilon_-^{(0)}} = \frac{3g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,6}^{(11)} = \frac{\langle +,6|H_{\mathrm{eZ}}|-,2\rangle\langle -,2|H_{\mathrm{hfs}}|+,6\rangle + \langle +,6|H_{\mathrm{hfs}}|-,2\rangle\langle -,2|H_{\mathrm{eZ}}|+,6\rangle}{\varepsilon_+^{(0)} - \varepsilon_-^{(0)}} = \frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,7}^{(11)} = \frac{\langle +,7|H_{\mathrm{eZ}}|-,3\rangle\langle -,3|H_{\mathrm{hfs}}|+,7\rangle + \langle +,7|H_{\mathrm{hfs}}|-,3\rangle\langle -,3|H_{\mathrm{eZ}}|+,7\rangle}{\varepsilon_+^{(0)} - \varepsilon_-^{(0)}} = -\frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,8}^{(11)} = \frac{\langle +,8|H_{\mathrm{eZ}}|-,4\rangle\langle -,4|H_{\mathrm{hfs}}|+,8\rangle + \langle +,8|H_{\mathrm{hfs}}|-,4\rangle\langle -,4|H_{\mathrm{eZ}}|+,8\rangle}{\varepsilon_+^{(0)} - \varepsilon_-^{(0)}} = -\frac{3g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

Thus, the perturbed energies for the case with $I = 3/2$ for the spin quartet state were explicitly obtained in the second order. To our knowledge the analytical expressions for the energies in terms of the Zeeman perturbation theory are for the first time given in this work, which are extremely accurate.

3. I = 5/2 case

The matrix elements of the hyperfine structure Hamiltonian are as follows:

$$\langle M_S', M_I' | H_{\mathrm{hfs}} | M_S, M_I \rangle = \begin{cases} \delta_{M_S M_S'} \delta_{M_I M_I'} M_S M_I A_{zz} \\ \dfrac{1}{16} \delta_{M_S M_S' \mp 1} \delta_{M_I M_I' \mp 1} \sqrt{15 - 4 M_S M_S'} \sqrt{35 - 4 M_I M_I'} \left(A_{xx} - A_{yy} \right) \\ \dfrac{1}{16} \delta_{M_S M_S' \mp 1} \delta_{M_I M_I' \pm 1} \sqrt{15 - 4 M_S M_S'} \sqrt{35 - 4 M_I M_I'} \left(A_{xx} + A_{yy} \right) \end{cases}$$

The matrix representation of the rank-2 ZFS Hamiltonian is the same as in the case of I = 1/2, and thus the energy eigenvalues and the spin eigenstates also have already been shown.

Similar to the I = 1/2 and 3/2 cases, $\left\{ \left| \varphi_{M_S, M_I}^{(0)} \right\rangle \right\}$ are divided into two subspaces according to the sign of the eigenvalues;

$$\left\{ \begin{matrix} \left| \varphi_{+\frac{3}{2},+\frac{5}{2}}^{(0)} \right\rangle, \left| \varphi_{+\frac{3}{2},+\frac{3}{2}}^{(0)} \right\rangle, \left| \varphi_{+\frac{3}{2},+\frac{1}{2}}^{(0)} \right\rangle, \left| \varphi_{+\frac{3}{2},-\frac{1}{2}}^{(0)} \right\rangle, \left| \varphi_{+\frac{3}{2},-\frac{3}{2}}^{(0)} \right\rangle, \left| \varphi_{+\frac{3}{2},-\frac{5}{2}}^{(0)} \right\rangle, \\ \left| \varphi_{-\frac{3}{2},+\frac{5}{2}}^{(0)} \right\rangle, \left| \varphi_{-\frac{3}{2},+\frac{3}{2}}^{(0)} \right\rangle, \left| \varphi_{-\frac{3}{2},+\frac{1}{2}}^{(0)} \right\rangle, \left| \varphi_{-\frac{3}{2},-\frac{1}{2}}^{(0)} \right\rangle, \left| \varphi_{-\frac{3}{2},-\frac{3}{2}}^{(0)} \right\rangle, \left| \varphi_{-\frac{3}{2},-\frac{5}{2}}^{(0)} \right\rangle \end{matrix} \right\} \text{(eigenvalue } \Delta\text{)}$$

$$\left\{ \begin{matrix} \left| \varphi_{+\frac{1}{2},+\frac{5}{2}}^{(0)} \right\rangle, \left| \varphi_{+\frac{1}{2},+\frac{3}{2}}^{(0)} \right\rangle, \left| \varphi_{+\frac{1}{2},+\frac{1}{2}}^{(0)} \right\rangle, \left| \varphi_{+\frac{1}{2},-\frac{1}{2}}^{(0)} \right\rangle, \left| \varphi_{+\frac{1}{2},-\frac{3}{2}}^{(0)} \right\rangle, \left| \varphi_{+\frac{1}{2},-\frac{5}{2}}^{(0)} \right\rangle, \\ \left| \varphi_{-\frac{1}{2},+\frac{5}{2}}^{(0)} \right\rangle, \left| \varphi_{-\frac{1}{2},+\frac{3}{2}}^{(0)} \right\rangle, \left| \varphi_{-\frac{1}{2},+\frac{1}{2}}^{(0)} \right\rangle, \left| \varphi_{-\frac{1}{2},-\frac{1}{2}}^{(0)} \right\rangle, \left| \varphi_{-\frac{1}{2},-\frac{3}{2}}^{(0)} \right\rangle, \left| \varphi_{-\frac{1}{2},-\frac{5}{2}}^{(0)} \right\rangle \end{matrix} \right\} \text{(eigenvalue } -\Delta\text{)}$$

The division into the two subspaces is due to the symmetry of the spin eigenfunctions involved. We rewrite the notation of the eigenfunctions as follows for simplicity.

$$\left\{ \begin{matrix} \left| \varphi_{+\frac{3}{2},+\frac{5}{2}}^{(0)} \right\rangle, \left| \varphi_{+\frac{3}{2},+\frac{3}{2}}^{(0)} \right\rangle, \left| \varphi_{+\frac{3}{2},+\frac{1}{2}}^{(0)} \right\rangle, \left| \varphi_{+\frac{3}{2},-\frac{1}{2}}^{(0)} \right\rangle, \left| \varphi_{+\frac{3}{2},-\frac{3}{2}}^{(0)} \right\rangle, \left| \varphi_{+\frac{3}{2},-\frac{5}{2}}^{(0)} \right\rangle, \\ \left| \varphi_{-\frac{3}{2},+\frac{5}{2}}^{(0)} \right\rangle, \left| \varphi_{-\frac{3}{2},+\frac{3}{2}}^{(0)} \right\rangle, \left| \varphi_{-\frac{3}{2},+\frac{1}{2}}^{(0)} \right\rangle, \left| \varphi_{-\frac{3}{2},-\frac{1}{2}}^{(0)} \right\rangle, \left| \varphi_{-\frac{3}{2},-\frac{3}{2}}^{(0)} \right\rangle, \left| \varphi_{-\frac{3}{2},-\frac{5}{2}}^{(0)} \right\rangle \end{matrix} \right\}$$

$$\to \{|+,1\rangle, |+,2\rangle, |+,3\rangle, |+,4\rangle, |+,5\rangle, |+,6\rangle, |+,7\rangle, |+,8\rangle, |+,9\rangle, |+,10\rangle, |+,11\rangle, |+,12\rangle\}$$

$$\left\{ \begin{matrix} \left| \varphi_{+\frac{1}{2},+\frac{5}{2}}^{(0)} \right\rangle, \left| \varphi_{+\frac{1}{2},+\frac{3}{2}}^{(0)} \right\rangle, \left| \varphi_{+\frac{1}{2},+\frac{1}{2}}^{(0)} \right\rangle, \left| \varphi_{+\frac{1}{2},-\frac{1}{2}}^{(0)} \right\rangle, \left| \varphi_{+\frac{1}{2},-\frac{3}{2}}^{(0)} \right\rangle, \left| \varphi_{+\frac{1}{2},-\frac{5}{2}}^{(0)} \right\rangle, \\ \left| \varphi_{-\frac{1}{2},+\frac{5}{2}}^{(0)} \right\rangle, \left| \varphi_{-\frac{1}{2},+\frac{3}{2}}^{(0)} \right\rangle, \left| \varphi_{-\frac{1}{2},+\frac{1}{2}}^{(0)} \right\rangle, \left| \varphi_{-\frac{1}{2},-\frac{1}{2}}^{(0)} \right\rangle, \left| \varphi_{-\frac{1}{2},-\frac{3}{2}}^{(0)} \right\rangle, \left| \varphi_{-\frac{1}{2},-\frac{5}{2}}^{(0)} \right\rangle \end{matrix} \right\}$$

$$\to \{|-,1\rangle, |-,2\rangle, |-,3\rangle, |-,4\rangle, |-,5\rangle, |-,6\rangle, |-,7\rangle, |-,8\rangle, |-,9\rangle, |-,10\rangle, |-,11\rangle, |-,12\rangle\}$$

According to the degenerate perturbation theory, the first-order correction for the energy of the hyperfine structure Hamiltonian which belong to the group of the positive eigenenergy ($\varepsilon_{+,\alpha}^{(01)}$ $(\alpha = 1, \cdots, 12)$) are the eigenvalues of the following matrix:

$$H_{\mathrm{hfs}}^{+,1} = \begin{pmatrix} \langle +,1|H_{\mathrm{hfs}}|+,1\rangle & \langle +,1|H_{\mathrm{hfs}}|+,3\rangle & \langle +,1|H_{\mathrm{hfs}}|+,5\rangle & \langle +,1|H_{\mathrm{hfs}}|+,8\rangle & \langle +,1|H_{\mathrm{hfs}}|+,10\rangle & \langle +,1|H_{\mathrm{hfs}}|+,12\rangle \\ \langle +,3|H_{\mathrm{hfs}}|+,1\rangle & \langle +,3|H_{\mathrm{hfs}}|+,3\rangle & \langle +,3|H_{\mathrm{hfs}}|+,5\rangle & \langle +,3|H_{\mathrm{hfs}}|+,8\rangle & \langle +,3|H_{\mathrm{hfs}}|+,10\rangle & \langle +,3|H_{\mathrm{hfs}}|+,12\rangle \\ \langle +,5|H_{\mathrm{hfs}}|+,1\rangle & \langle +,5|H_{\mathrm{hfs}}|+,3\rangle & \langle +,5|H_{\mathrm{hfs}}|+,5\rangle & \langle +,5|H_{\mathrm{hfs}}|+,8\rangle & \langle +,5|H_{\mathrm{hfs}}|+,10\rangle & \langle +,5|H_{\mathrm{hfs}}|+,12\rangle \\ \langle +,8|H_{\mathrm{hfs}}|+,1\rangle & \langle +,8|H_{\mathrm{hfs}}|+,3\rangle & \langle +,8|H_{\mathrm{hfs}}|+,5\rangle & \langle +,8|H_{\mathrm{hfs}}|+,8\rangle & \langle +,8|H_{\mathrm{hfs}}|+,10\rangle & \langle +,8|H_{\mathrm{hfs}}|+,12\rangle \\ \langle +,10|H_{\mathrm{hfs}}|+,1\rangle & \langle +,10|H_{\mathrm{hfs}}|+,3\rangle & \langle +,10|H_{\mathrm{hfs}}|+,5\rangle & \langle +,10|H_{\mathrm{hfs}}|+,8\rangle & \langle +,10|H_{\mathrm{hfs}}|+,10\rangle & \langle +,10|H_{\mathrm{hfs}}|+,12\rangle \\ \langle +,12|H_{\mathrm{hfs}}|+,1\rangle & \langle +,12|H_{\mathrm{hfs}}|+,3\rangle & \langle +,12|H_{\mathrm{hfs}}|+,5\rangle & \langle +,12|H_{\mathrm{hfs}}|+,8\rangle & \langle +,12|H_{\mathrm{hfs}}|+,10\rangle & \langle +,11|H_{\mathrm{hfs}}|+,12\rangle \end{pmatrix}$$

$$H_{\mathrm{hfs}}^{+,2} = \begin{pmatrix} \langle +,2|H_{\mathrm{hfs}}|+,2\rangle & \langle +,2|H_{\mathrm{hfs}}|+,4\rangle & \langle +,2|H_{\mathrm{hfs}}|+,6\rangle & \langle +,2|H_{\mathrm{hfs}}|+,7\rangle & \langle +,2|H_{\mathrm{hfs}}|+,9\rangle & \langle +,2|H_{\mathrm{hfs}}|+,11\rangle \\ \langle +,4|H_{\mathrm{hfs}}|+,2\rangle & \langle +,4|H_{\mathrm{hfs}}|+,4\rangle & \langle +,4|H_{\mathrm{hfs}}|+,6\rangle & \langle +,4|H_{\mathrm{hfs}}|+,7\rangle & \langle +,4|H_{\mathrm{hfs}}|+,9\rangle & \langle +,4|H_{\mathrm{hfs}}|+,11\rangle \\ \langle +,6|H_{\mathrm{hfs}}|+,2\rangle & \langle +,6|H_{\mathrm{hfs}}|+,4\rangle & \langle +,6|H_{\mathrm{hfs}}|+,6\rangle & \langle +,6|H_{\mathrm{hfs}}|+,7\rangle & \langle +,6|H_{\mathrm{hfs}}|+,9\rangle & \langle +,6|H_{\mathrm{hfs}}|+,11\rangle \\ \langle +,7|H_{\mathrm{hfs}}|+,2\rangle & \langle +,7|H_{\mathrm{hfs}}|+,4\rangle & \langle +,7|H_{\mathrm{hfs}}|+,6\rangle & \langle +,7|H_{\mathrm{hfs}}|+,7\rangle & \langle +,7|H_{\mathrm{hfs}}|+,9\rangle & \langle +,7|H_{\mathrm{hfs}}|+,11\rangle \\ \langle +,9|H_{\mathrm{hfs}}|+,2\rangle & \langle +,9|H_{\mathrm{hfs}}|+,4\rangle & \langle +,9|H_{\mathrm{hfs}}|+,6\rangle & \langle +,9|H_{\mathrm{hfs}}|+,7\rangle & \langle +,9|H_{\mathrm{hfs}}|+,9\rangle & \langle +,9|H_{\mathrm{hfs}}|+,11\rangle \\ \langle +,11|H_{\mathrm{hfs}}|+,2\rangle & \langle +,11|H_{\mathrm{hfs}}|+,4\rangle & \langle +,11|H_{\mathrm{hfs}}|+,6\rangle & \langle +,11|H_{\mathrm{hfs}}|+,7\rangle & \langle +,11|H_{\mathrm{hfs}}|+,9\rangle & \langle +,11|H_{\mathrm{hfs}}|+,11\rangle \end{pmatrix}$$

$$\langle +,1|H_{\mathrm{hfs}}|+,1\rangle = \langle +,12|H_{\mathrm{hfs}}|+,12\rangle = \frac{5}{4}A_z(1+2\cos 2\theta)$$

$$\langle +,2|H_{\mathrm{hfs}}|+,2\rangle = \langle +,11|H_{\mathrm{hfs}}|+,11\rangle = \frac{3}{4}A_z(1+2\cos 2\theta)$$

$$\langle +,3|H_{\mathrm{hfs}}|+,3\rangle = \langle +,10|H_{\mathrm{hfs}}|+,10\rangle = \frac{1}{4}A_z(1+2\cos 2\theta)$$

$$\langle +,4|H_{\mathrm{hfs}}|+,4\rangle = \langle +,9|H_{\mathrm{hfs}}|+,9\rangle = -\frac{1}{4}A_z(1+2\cos 2\theta)$$

$$\langle +,5|H_{\mathrm{hfs}}|+,5\rangle = \langle +,8|H_{\mathrm{hfs}}|+,8\rangle = -\frac{3}{4}A_z(1+2\cos 2\theta)$$

$$\langle +,6|H_{\mathrm{hfs}}|+,6\rangle = \langle +,7|H_{\mathrm{hfs}}|+,7\rangle = -\frac{5}{4}A_z(1+2\cos 2\theta)$$

$$\begin{aligned}\langle +,1|H_{\mathrm{hfs}}|+,8\rangle &= \langle +,8|H_{\mathrm{hfs}}|+,1\rangle = \langle +,5|H_{\mathrm{hfs}}|+,12\rangle = \langle +,12|H_{\mathrm{hfs}}|+,5\rangle \\ &= \frac{\sqrt{5}}{2}\sin\theta\left[\sqrt{3}\left(A_x - A_y\right)\cos\theta + \left(A_x + A_y\right)\sin\theta\right]\end{aligned}$$

$$\begin{aligned}\langle +,2|H_{\mathrm{hfs}}|+,7\rangle &= \langle +,7|H_{\mathrm{hfs}}|+,2\rangle = \langle +,6|H_{\mathrm{hfs}}|+,11\rangle = \langle +,11|H_{\mathrm{hfs}}|+,6\rangle \\ &= \frac{\sqrt{5}}{2}\sin\theta\left[\sqrt{3}\left(A_x + A_y\right)\cos\theta + \left(A_x - A_y\right)\sin\theta\right]\end{aligned}$$

$$\begin{aligned}\langle +,2|H_{\mathrm{hfs}}|+,9\rangle &= \langle +,9|H_{\mathrm{hfs}}|+,2\rangle = \langle +,4|H_{\mathrm{hfs}}|+,11\rangle = \langle +,11|H_{\mathrm{hfs}}|+,4\rangle \\ &= \sqrt{2}\sin\theta\left[\sqrt{3}\left(A_x - A_y\right)\cos\theta + \left(A_x + A_y\right)\sin\theta\right]\end{aligned}$$

$$\begin{aligned}\langle +,3|H_{\mathrm{hfs}}|+,8\rangle &= \langle +,8|H_{\mathrm{hfs}}|+,3\rangle = \langle +,5|H_{\mathrm{hfs}}|+,10\rangle = \langle +,10|H_{\mathrm{hfs}}|+,5\rangle \\ &= \sqrt{2}\sin\theta\left[\sqrt{3}\left(A_x + A_y\right)\cos\theta + \left(A_x - A_y\right)\sin\theta\right]\end{aligned}$$

$$\langle +,3|H_{\mathrm{hfs}}|+,10\rangle = \langle +,10|H_{\mathrm{hfs}}|+,3\rangle = \frac{3}{2}\sin\theta\left[\sqrt{3}\left(A_x - A_y\right)\cos\theta + \left(A_x + A_y\right)\sin\theta\right]$$

$$\langle +,4|H_{\mathrm{hfs}}|+,9\rangle = \langle +,9|H_{\mathrm{hfs}}|+,4\rangle = \frac{3}{2}\sin\theta\left[\sqrt{3}\left(A_x + A_y\right)\cos\theta + \left(A_x - A_y\right)\sin\theta\right]$$

while the first-order corrections of the energy which belongs to the group of the negative eigenenergy ($\varepsilon_{-,\alpha}^{(01)}$ $(\alpha = 1, \cdots, 12)$) are the eigenvalues of the following matrix:

$$H_{\mathrm{hfs}}^{-,1} = \begin{pmatrix} \langle -,1|H_{\mathrm{hfs}}|-,1\rangle & \langle -,1|H_{\mathrm{hfs}}|-,3\rangle & \langle -,1|H_{\mathrm{hfs}}|-,5\rangle & \langle -,1|H_{\mathrm{hfs}}|-,8\rangle & \langle -,1|H_{\mathrm{hfs}}|-,10\rangle & \langle -,1|H_{\mathrm{hfs}}|-,12\rangle \\ \langle -,3|H_{\mathrm{hfs}}|-,1\rangle & \langle -,3|H_{\mathrm{hfs}}|-,3\rangle & \langle -,3|H_{\mathrm{hfs}}|-,5\rangle & \langle -,3|H_{\mathrm{hfs}}|-,8\rangle & \langle -,3|H_{\mathrm{hfs}}|-,10\rangle & \langle -,3|H_{\mathrm{hfs}}|-,12\rangle \\ \langle -,5|H_{\mathrm{hfs}}|-,1\rangle & \langle -,5|H_{\mathrm{hfs}}|-,3\rangle & \langle -,5|H_{\mathrm{hfs}}|-,5\rangle & \langle -,5|H_{\mathrm{hfs}}|-,8\rangle & \langle -,5|H_{\mathrm{hfs}}|-,10\rangle & \langle -,5|H_{\mathrm{hfs}}|-,12\rangle \\ \langle -,8|H_{\mathrm{hfs}}|-,1\rangle & \langle -,8|H_{\mathrm{hfs}}|-,3\rangle & \langle -,8|H_{\mathrm{hfs}}|-,5\rangle & \langle -,8|H_{\mathrm{hfs}}|-,8\rangle & \langle -,8|H_{\mathrm{hfs}}|-,10\rangle & \langle -,8|H_{\mathrm{hfs}}|-,12\rangle \\ \langle -,10|H_{\mathrm{hfs}}|-,1\rangle & \langle -,10|H_{\mathrm{hfs}}|-,3\rangle & \langle -,10|H_{\mathrm{hfs}}|-,5\rangle & \langle -,10|H_{\mathrm{hfs}}|-,8\rangle & \langle -,10|H_{\mathrm{hfs}}|-,10\rangle & \langle -,10|H_{\mathrm{hfs}}|-,12\rangle \\ \langle -,12|H_{\mathrm{hfs}}|-,1\rangle & \langle -,12|H_{\mathrm{hfs}}|-,3\rangle & \langle -,12|H_{\mathrm{hfs}}|-,5\rangle & \langle -,12|H_{\mathrm{hfs}}|-,8\rangle & \langle -,12|H_{\mathrm{hfs}}|-,10\rangle & \langle -,12|H_{\mathrm{hfs}}|-,12\rangle \end{pmatrix}$$

$$H_{\mathrm{hfs}}^{-,2} = \begin{pmatrix} \langle -,2|H_{\mathrm{hfs}}|-,2\rangle & \langle -,2|H_{\mathrm{hfs}}|-,4\rangle & \langle -,2|H_{\mathrm{hfs}}|-,6\rangle & \langle -,2|H_{\mathrm{hfs}}|-,7\rangle & \langle -,2|H_{\mathrm{hfs}}|-,9\rangle & \langle -,2|H_{\mathrm{hfs}}|-,11\rangle \\ \langle -,4|H_{\mathrm{hfs}}|-,2\rangle & \langle -,4|H_{\mathrm{hfs}}|-,4\rangle & \langle -,4|H_{\mathrm{hfs}}|-,6\rangle & \langle -,4|H_{\mathrm{hfs}}|-,7\rangle & \langle -,4|H_{\mathrm{hfs}}|-,9\rangle & \langle -,4|H_{\mathrm{hfs}}|-,11\rangle \\ \langle -,6|H_{\mathrm{hfs}}|-,2\rangle & \langle -,6|H_{\mathrm{hfs}}|-,4\rangle & \langle -,6|H_{\mathrm{hfs}}|-,6\rangle & \langle -,6|H_{\mathrm{hfs}}|-,7\rangle & \langle -,6|H_{\mathrm{hfs}}|-,9\rangle & \langle -,6|H_{\mathrm{hfs}}|-,11\rangle \\ \langle -,7|H_{\mathrm{hfs}}|-,2\rangle & \langle -,7|H_{\mathrm{hfs}}|-,4\rangle & \langle -,7|H_{\mathrm{hfs}}|-,6\rangle & \langle -,7|H_{\mathrm{hfs}}|-,7\rangle & \langle -,7|H_{\mathrm{hfs}}|-,9\rangle & \langle -,7|H_{\mathrm{hfs}}|-,11\rangle \\ \langle -,9|H_{\mathrm{hfs}}|-,2\rangle & \langle -,9|H_{\mathrm{hfs}}|-,4\rangle & \langle -,9|H_{\mathrm{hfs}}|-,6\rangle & \langle -,9|H_{\mathrm{hfs}}|-,7\rangle & \langle -,9|H_{\mathrm{hfs}}|-,9\rangle & \langle -,9|H_{\mathrm{hfs}}|-,11\rangle \\ \langle -,11|H_{\mathrm{hfs}}|-,2\rangle & \langle -,11|H_{\mathrm{hfs}}|-,4\rangle & \langle -,11|H_{\mathrm{hfs}}|-,6\rangle & \langle -,11|H_{\mathrm{hfs}}|-,7\rangle & \langle -,11|H_{\mathrm{hfs}}|-,9\rangle & \langle -,11|H_{\mathrm{hfs}}|-,11\rangle \end{pmatrix}$$

$$\langle -,1|H_{\mathrm{hfs}}|-,1\rangle = \langle -,12|H_{\mathrm{hfs}}|-,12\rangle = \frac{5}{4}A_z(-1+2\cos 2\theta)$$

$$\langle -,2|H_{\mathrm{hfs}}|-,2\rangle = \langle -,11|H_{\mathrm{hfs}}|-,11\rangle = \frac{3}{4}A_z(-1+2\cos 2\theta)$$

$$\langle -,3|H_{\mathrm{hfs}}|-,3\rangle = \langle -,10|H_{\mathrm{hfs}}|-,10\rangle = \frac{A_z}{4}(-1+2\cos 2\theta)$$

$$\langle -,4|H_{\mathrm{hfs}}|-,4\rangle = \langle -,9|H_{\mathrm{hfs}}|-,9\rangle = \frac{A_z}{4}(1-2\cos 2\theta)$$

$$\langle -,5|H_{\mathrm{hfs}}|-,5\rangle = \langle -,8|H_{\mathrm{hfs}}|-,8\rangle = \frac{3}{4}A_z(1-2\cos 2\theta)$$

$$\langle -,6|H_{\mathrm{hfs}}|-,6\rangle = \langle -,7|H_{\mathrm{hfs}}|-,7\rangle = \frac{5}{4}A_z(1-2\cos 2\theta)$$

$$\begin{aligned}\langle -,1|H_{\mathrm{hfs}}|-,8\rangle &= \langle -,8|H_{\mathrm{hfs}}|-,1\rangle = \langle -,5|H_{\mathrm{hfs}}|-,12\rangle = \langle -,12|H_{\mathrm{hfs}}|-,5\rangle \\ &= \frac{\sqrt{5}}{2}\cos\theta\left[\left(A_x - A_y\right)\cos\theta - \sqrt{3}\left(A_x + A_y\right)\sin\theta\right]\end{aligned}$$

$$\begin{aligned}\langle -,2|H_{\mathrm{hfs}}|-,7\rangle &= \langle -,7|H_{\mathrm{hfs}}|-,2\rangle = \langle -,6|H_{\mathrm{hfs}}|-,11\rangle = \langle -,11|H_{\mathrm{hfs}}|-,6\rangle \\ &= \frac{\sqrt{5}}{2}\cos\theta\left[\left(A_x + A_y\right)\cos\theta - \sqrt{3}\left(A_x - A_y\right)\sin\theta\right]\end{aligned}$$

$$\begin{aligned}\langle -,2|H_{\mathrm{hfs}}|-,9\rangle &= \langle -,9|H_{\mathrm{hfs}}|-,2\rangle = \langle -,4|H_{\mathrm{hfs}}|-,11\rangle = \langle -,11|H_{\mathrm{hfs}}|-,4\rangle \\ &= \sqrt{2}\cos\theta\left[\left(A_x - A_y\right)\cos\theta - \sqrt{3}\left(A_x + A_y\right)\sin\theta\right]\end{aligned}$$

$$\begin{aligned}\langle -,3|H_{\mathrm{hfs}}|-,8\rangle &= \langle -,8|H_{\mathrm{hfs}}|-,3\rangle = \langle -,5|H_{\mathrm{hfs}}|-,10\rangle = \langle -,10|H_{\mathrm{hfs}}|-,5\rangle \\ &= \sqrt{2}\cos\theta\left[\left(A_x + A_y\right)\cos\theta - \sqrt{3}\left(A_x - A_y\right)\sin\theta\right]\end{aligned}$$

$$\langle -,3|H_{\mathrm{hfs}}|-,10\rangle = \langle -,10|H_{\mathrm{hfs}}|-,3\rangle = \frac{3}{2}\cos\theta\left[\left(A_x - A_y\right)\cos\theta - \sqrt{3}\left(A_x + A_y\right)\sin\theta\right]$$

$$\langle -,4|H_{\mathrm{hfs}}|-,9\rangle = \langle -,9|H_{\mathrm{hfs}}|-,4\rangle = \frac{3}{2}\cos\theta\left[\left(A_x + A_y\right)\cos\theta - \sqrt{3}\left(A_x - A_y\right)\sin\theta\right]$$

The other elements are zero. The secular equation can be factorized into two cubic equations in the form of

$$x^3 + a_i x^2 + b_i x + c_i = 0 \ (i = 1, \cdots, 8)$$

In the equation, i = 1 and 2, 3 and 4, 5 and 6, and 7 and 8 come from $H_{\mathrm{hfs}}^{+,1}$, $H_{\mathrm{hfs}}^{+,2}$, $H_{\mathrm{hfs}}^{-,1}$, $H_{\mathrm{hfs}}^{-,2}$, respectively. In order to eliminate x^2 term, replacing x to $x - a_i/3$ yields

$$x^3 = \frac{1}{3}({a_i}^2 + 3b_i)x - \frac{1}{27}(2{a_i}^3 - 9a_i b_i + 27c_i)$$

According to the Viete's method [6], the solutions of the cubic equation are given as

$$x_n = 2p_i\cos\left[\frac{1}{3}\arccos\left(\frac{q_i}{2p_i}\right) + \frac{2n\pi}{3}\right] - \frac{a_i}{3} \ (n = 0, 1, 2)$$

with

$$p_i = \frac{1}{3}\sqrt{{a_i}^2 + 3b_i}$$

$$q_i = \frac{-2{a_i}^3 + 9a_i b_i - 27c_i}{3{a_i}^2 + 9b_i}$$

The set of the coefficients of the cubic equation $\{a_i, b_i, c_i\}$ is given in the following;

$$a_1 = \frac{3}{4}\left(A_x + A_y - A_z\right) - \frac{3}{4}\left(A_x + A_y + 2A_z\right)\cos 2\theta + \frac{3\sqrt{3}}{4}\left(A_x - A_y\right)\sin 2\theta$$

$$\begin{aligned}
b_1 = &-\frac{39}{16}\left({A_x}^2 + {A_y}^2 + {A_z}^2\right) + \frac{1}{8}\left(13{A_x}^2 - 6A_xA_y + 13{A_y}^2 - 3A_xA_z - 3A_yA_z - 26{A_z}^2\right)\cos 2\theta \\
&+ \frac{1}{16}\left(13{A_x}^2 + 12A_xA_y + 13{A_y}^2 + 6A_xA_z + 6A_yA_z - 26{A_z}^2\right)\cos 4\theta \\
&+ \frac{\sqrt{3}}{8}\left(-13{A_x}^2 + 13{A_y}^2 - 3A_xA_z + 3A_yA_z\right)\sin 2\theta \\
&+ \frac{\sqrt{3}}{16}\left(13{A_x}^2 - 13{A_y}^2 - 6A_xA_z + 6A_yA_z\right)\sin 4\theta
\end{aligned}$$

$$\begin{aligned}
c_1 = &\frac{5}{64}\left(-21{A_x}^3 - 9{A_x}^2A_y - 9A_x{A_y}^2 - 21{A_y}^3 + 9{A_x}^2A_z + 28A_xA_yA_z + 9{A_y}^2A_z - 9A_x{A_z}^2\right. \\
&\left. - 9A_y{A_z}^2 + 21{A_z}^3\right) \\
&+ \frac{45}{64}\left(2{A_x}^3 + 3{A_x}^2A_y + 3A_x{A_y}^2 + 2{A_y}^3 + 3{A_x}^2A_z + 3{A_y}^2A_z + 4{A_z}^3\right)\cos 2\theta \\
&+ \frac{45}{64}\left({A_x}^3 - 3{A_x}^2A_y - 3A_x{A_y}^2 + {A_y}^3 - 3{A_x}^2A_z - 3{A_y}^2A_z + 2{A_z}^3\right)\cos 4\theta \\
&+ \frac{5}{64}\left(-6{A_x}^3 + 9{A_x}^2A_y + 9A_x{A_y}^2 - 6{A_y}^3 - 9{A_x}^2A_z - 28A_xA_yA_z - 9{A_y}^2A_z\right. \\
&\left. + 9A_x{A_z}^2 + 9A_y{A_z}^2 + 6{A_z}^3\right)\cos 6\theta \\
&+ \frac{45\sqrt{3}}{64}\left(-2{A_x}^3 + {A_x}^2A_y - A_x{A_y}^2 + 2{A_y}^3 + {A_x}^2A_z - {A_y}^2A_z - 2A_x{A_z}^2\right. \\
&\left. + 2A_y{A_z}^2\right)\sin 2\theta \\
&+ \frac{45\sqrt{3}}{64}\left({A_x}^3 + {A_x}^2A_y - A_x{A_y}^2 - {A_y}^3 + {A_x}^2A_z - {A_y}^2A_z - 2A_x{A_z}^2 + 2A_y{A_z}^2\right)\sin 4\theta \\
&+ \frac{45\sqrt{3}}{64}\left(-{A_x}^2A_y + A_x{A_y}^2 - {A_x}^2A_z + {A_y}^2A_z - A_x{A_z}^2 + A_y{A_z}^2\right)\sin 6\theta
\end{aligned}$$

$$a_2 = \frac{3}{4}\left(-A_x - A_y - A_z\right) + \frac{3}{4}\left(A_x + A_y - 2A_z\right)\cos 2\theta - \frac{3\sqrt{3}}{4}\left(A_x - A_y\right)\sin 2\theta$$

$$\begin{aligned}
b_2 = &-\frac{39}{16}\left({A_x}^2 + {A_y}^2 + {A_z}^2\right) + \frac{1}{8}\left(13{A_x}^2 - 6A_xA_y + 13{A_y}^2 + 3A_xA_z + 3A_yA_z - 26{A_z}^2\right)\cos 2\theta \\
&+ \frac{1}{16}\left(13{A_x}^2 + 12A_xA_y + 13{A_y}^2 - 6A_xA_z - 6A_yA_z - 26{A_z}^2\right)\cos 4\theta \\
&- \frac{\sqrt{3}}{8}\left(13{A_x}^2 - 13{A_y}^2 - 3A_xA_z + 3A_yA_z\right)\sin 2\theta \\
&+ \frac{\sqrt{3}}{16}\left(13{A_x}^2 - 13{A_y}^2 + 6A_xA_z - 6A_yA_z\right)\sin 4\theta
\end{aligned}$$

$$
\begin{aligned}
c_2 = \frac{5}{64}\big(&21{A_x}^3 + 9{A_x}^2A_y + 9A_x{A_y}^2 + 21{A_y}^3 + 9{A_x}^2A_z + 28A_xA_yA_z + 9{A_y}^2A_z + 9A_x{A_z}^2 + 9A_y{A_z}^2 \\
&+ 21{A_z}^3\big) \\
&+ \frac{45}{64}\left(-2{A_x}^3 - 3{A_x}^2A_y - 3A_x{A_y}^2 - 2{A_y}^3 + 3{A_x}^2A_z + 3{A_y}^2A_z + 4{A_z}^3\right)\cos 2\theta \\
&+ \frac{45}{64}\left(-{A_x}^3 + 3{A_x}^2A_y + 3A_x{A_y}^2 - {A_y}^3 - 3{A_x}^2A_z - 3{A_y}^2A_z + 2{A_z}^3\right)\cos 4\theta \\
&+ \frac{5}{64}\big(6{A_x}^3 - 9{A_x}^2A_y - 9A_x{A_y}^2 + 6{A_y}^3 - 9{A_x}^2A_z - 28A_xA_yA_z - 9{A_y}^2A_z - 9A_x{A_z}^2 \\
&- 9A_y{A_z}^2 + 6{A_z}^3\big)\cos 6\theta \\
&+ \frac{45\sqrt{3}}{64}\big(2{A_x}^3 - {A_x}^2A_y + A_x{A_y}^2 - 2{A_y}^3 + {A_x}^2A_z - {A_y}^2A_z + 2A_x{A_z}^2 \\
&- 2A_y{A_z}^2\big)\sin 2\theta \\
&+ \frac{45\sqrt{3}}{64}\big(-{A_x}^3 - {A_x}^2A_y + A_x{A_y}^2 + {A_y}^3 + {A_x}^2A_z - {A_y}^2A_z + 2A_x{A_z}^2 \\
&- 2A_y{A_z}^2\big)\sin 4\theta \\
&+ \frac{45\sqrt{3}}{64}\left({A_x}^2A_y - A_x{A_y}^2 - {A_x}^2A_z + {A_y}^2A_z + A_x{A_z}^2 - A_y{A_z}^2\right)\sin 6\theta
\end{aligned}
$$

$$
a_3 = \frac{3}{4}\left(-A_x + A_y + A_z\right) + \frac{3}{4}\left(A_x - A_y + 2A_z\right)\cos 2\theta - \frac{3\sqrt{3}}{4}\left(A_x + A_y\right)\sin 2\theta
$$

$$
\begin{aligned}
b_3 = -\frac{39}{16}\left({A_x}^2 + {A_y}^2 + {A_z}^2\right) &+ \frac{1}{8}\left(13{A_x}^2 + 6A_xA_y + 13{A_y}^2 - 3A_xA_z + 3A_yA_z - 26{A_z}^2\right)\cos 2\theta \\
&+ \frac{1}{16}\left(13{A_x}^2 - 12A_xA_y + 13{A_y}^2 + 6A_xA_z - 6A_yA_z - 26{A_z}^2\right)\cos 4\theta \\
&+ \frac{\sqrt{3}}{8}\left(-13{A_x}^2 + 13{A_y}^2 - 3A_xA_z - 3A_yA_z\right)\sin 2\theta \\
&+ \frac{\sqrt{3}}{16}\left(13{A_x}^2 - 13{A_y}^2 - 6A_xA_z - 6A_yA_z\right)\sin 4\theta
\end{aligned}
$$

$$c_3 = \frac{5}{64}\left(21{A_x}^3 - 9{A_x}^2A_y + 9A_x{A_y}^2 - 21{A_y}^3 - 9{A_x}^2A_z + 28A_xA_yA_z - 9{A_y}^2A_z + 9A_x{A_z}^2 - 9A_y{A_z}^2 - 21{A_z}^3\right)$$
$$+ \frac{45}{64}\left(-2{A_x}^3 + 3{A_x}^2A_y - 3A_x{A_y}^2 + 2{A_y}^3 - 3{A_x}^2A_z - 3{A_y}^2A_z - 4{A_z}^3\right)\cos 2\theta$$
$$+ \frac{45}{64}\left(-{A_x}^3 - 3{A_x}^2A_y + 3A_x{A_y}^2 + {A_y}^3 + 3{A_x}^2A_z + 3{A_y}^2A_z - 2{A_z}^3\right)\cos 4\theta$$
$$+ \frac{5}{64}\left(6{A_x}^3 + 9{A_x}^2A_y - 9A_x{A_y}^2 - 6{A_y}^3 + 9{A_x}^2A_z - 28A_xA_yA_z + 9{A_y}^2A_z - 9A_x{A_z}^2 + 9A_y{A_z}^2 - 6{A_z}^3\right)\cos 6\theta$$
$$+ \frac{45\sqrt{3}}{64}\left(2{A_x}^3 + 4{A_x}^2A_y + A_x{A_y}^2 + 2{A_y}^3 - {A_x}^2A_z + {A_y}^2A_z + 2A_x{A_z}^2 + 2A_y{A_z}^2\right)\sin 2\theta$$
$$+ \frac{45\sqrt{3}}{64}\left(-{A_x}^3 + {A_x}^2A_y + A_x{A_y}^2 - {A_y}^3 - {A_x}^2A_z + {A_y}^2A_z + 2A_x{A_z}^2 + 2A_y{A_z}^2\right)\sin 4\theta$$
$$+ \frac{45\sqrt{3}}{64}\left(-{A_x}^2A_y - A_x{A_y}^2 + {A_x}^2A_z - {A_y}^2A_z + A_x{A_z}^2 + A_y{A_z}^2\right)\sin 6\theta$$

$$a_4 = \frac{3}{4}\left(A_x - A_y + A_z\right) - \frac{3}{4}\left(A_x - A_y - 2A_z\right)\cos 2\theta + \frac{3\sqrt{3}}{4}\left(A_x + A_y\right)\sin 2\theta$$

$$b_4 = -\frac{39}{16}\left({A_x}^2 + {A_y}^2 + {A_z}^2\right) + \frac{1}{8}\left(13{A_x}^2 + 6A_xA_y + 13{A_y}^2 + 3A_xA_z - 3A_yA_z - 26{A_z}^2\right)\cos 2\theta$$
$$+ \frac{1}{16}\left(13{A_x}^2 - 12A_xA_y + 13{A_y}^2 - 6A_xA_z + 6A_yA_z - 26{A_z}^2\right)\cos 4\theta$$
$$- \frac{\sqrt{3}}{8}\left(13{A_x}^2 - 13{A_y}^2 - 3A_xA_z - 3A_yA_z\right)\sin 2\theta$$
$$+ \frac{\sqrt{3}}{16}\left(13{A_x}^2 - 13{A_y}^2 + 6A_xA_z + 6A_yA_z\right)\sin 4\theta$$

$$c_4 = \frac{5}{64}\left(-21{A_x}^3 + 9{A_x}^2A_y - 9A_x{A_y}^2 + 21{A_y}^3 - 9{A_x}^2A_z + 28A_xA_yA_z - 9{A_y}^2A_z - 9A_x{A_z}^2 \right.$$
$$\left. + 9A_y{A_z}^2 - 21{A_z}^3\right)$$
$$+ \frac{45}{64}\left(2{A_x}^3 - 3{A_x}^2A_y + 3A_x{A_y}^2 - 2{A_y}^3 - 3{A_x}^2A_z - 3{A_y}^2A_z - 4{A_z}^3\right)\cos 2\theta$$
$$+ \frac{45}{64}\left({A_x}^3 + 3{A_x}^2A_y - 3A_x{A_y}^2 - {A_y}^3 + 3{A_x}^2A_z + 3{A_y}^2A_z - 2{A_z}^3\right)\cos 4\theta$$
$$+ \frac{5}{64}\left(-6{A_x}^3 - 9{A_x}^2A_y + 9A_x{A_y}^2 + 6{A_y}^3 + 9{A_x}^2A_z - 28A_xA_yA_z + 9{A_y}^2A_z \right.$$
$$\left. + 9A_x{A_z}^2 - 9A_y{A_z}^2 - 6{A_z}^3\right)\cos 6\theta$$
$$+ \frac{45\sqrt{3}}{64}\left(-2{A_x}^3 - {A_x}^2A_y - A_x{A_y}^2 - 2{A_y}^3 - {A_x}^2A_z + {A_y}^2A_z - 2A_x{A_z}^2 \right.$$
$$\left. - 2A_y{A_z}^2\right)\sin 2\theta$$
$$+ \frac{45\sqrt{3}}{64}\left({A_x}^3 - {A_x}^2A_y - A_x{A_y}^2 + {A_y}^3 - {A_x}^2A_z + {A_y}^2A_z - 2A_x{A_z}^2 - 2A_y{A_z}^2\right)\sin 4\theta$$
$$+ \frac{45\sqrt{3}}{64}\left({A_x}^2A_y + A_x{A_y}^2 + {A_x}^2A_z - {A_y}^2A_z - A_x{A_z}^2 - A_y{A_z}^2\right)\sin 6\theta$$

We note that the coefficients for i = 2, 3 and 4 can be obtained from $\{a_1, b_1, c_1, d_1\}$ with replacing A_x and A_y to $-A_x$ and $-A_y$, A_x and A_z to $-A_x$ and $-A_z$, and A_y and A_z to $-A_y$ and $-A_z$, respectively.

For the negative counterpart:

$$a_5 = \frac{3}{4}\left(A_x - A_y + A_z\right) + \frac{3}{4}\left(A_x - A_y - 2A_z\right)\cos 2\theta + \frac{3\sqrt{3}}{4}\left(-A_x - A_y\right)\sin 2\theta$$

$$b_5 = -\frac{39}{16}\left({A_x}^2 + {A_y}^2 + {A_z}^2\right) - \frac{1}{8}\left(13{A_x}^2 + 6A_xA_y + 13{A_y}^2 + 3A_xA_z - 3A_yA_z - 26{A_z}^2\right)\cos 2\theta$$
$$+ \frac{1}{16}\left(13{A_x}^2 - 12A_xA_y + 13{A_y}^2 - 6A_xA_z + 6A_yA_z - 26{A_z}^2\right)\cos 4\theta$$
$$+ \frac{\sqrt{3}}{8}\left(13{A_x}^2 - 13{A_y}^2 - 3A_xA_z - 3A_yA_z\right)\sin 2\theta$$
$$+ \frac{\sqrt{3}}{16}\left(13{A_x}^2 - 13{A_y}^2 + 6A_xA_z + 6A_yA_z\right)\sin 4\theta$$

$$c_5 = \frac{5}{64}\left(-21{A_x}^3 + 9{A_x}^2A_y - 9A_x{A_y}^2 + 21{A_y}^3 - 9{A_x}^2A_z + 28A_xA_yA_z - 9{A_y}^2A_z - 9A_x{A_z}^2 + 9A_y{A_z}^2 - 21{A_z}^3\right)$$
$$+ \frac{45}{64}\left(-2{A_x}^3 + 3{A_x}^2A_y - 3A_x{A_y}^2 + 2{A_y}^3 + 3{A_x}^2A_z + 3{A_y}^2A_z + 4{A_z}^3\right)\cos 2\theta$$
$$+ \frac{45}{64}\left({A_x}^3 + 3{A_x}^2A_y - 3A_x{A_y}^2 - {A_y}^3 + 3{A_x}^2A_z + 3{A_y}^2A_z - 2{A_z}^3\right)\cos 4\theta$$
$$+ \frac{5}{64}\left(6{A_x}^3 + 9{A_x}^2A_y - 9A_x{A_y}^2 - 6{A_y}^3 - 9{A_x}^2A_z + 28A_xA_yA_z - 9{A_y}^2A_z - 9A_x{A_z}^2 + 9A_y{A_z}^2 + 6{A_z}^3\right)\cos 6\theta$$
$$+ \frac{45\sqrt{3}}{64}\left(2{A_x}^3 + {A_x}^2A_y + A_x{A_y}^2 + 2{A_y}^3 + {A_x}^2A_z - {A_y}^2A_z + 2A_x{A_z}^2 + 2A_y{A_z}^2\right)\sin 2\theta$$
$$+ \frac{45\sqrt{3}}{64}\left({A_x}^3 - {A_x}^2A_y - A_x{A_y}^2 + {A_y}^3 - {A_x}^2A_z + {A_y}^2A_z - 2A_x{A_z}^2 - 2A_y{A_z}^2\right)\sin 4\theta$$
$$+ \frac{45\sqrt{3}}{64}\left(-{A_x}^2A_y - A_x{A_y}^2 - {A_x}^2A_z + {A_y}^2A_z + A_x{A_z}^2 + A_y{A_z}^2\right)\sin 6\theta$$

$$a_6 = \frac{3}{4}\left(-A_x + A_y + A_z\right) + \frac{3}{4}\left(-A_x + A_y - 2A_z\right)\cos 2\theta + \frac{3\sqrt{3}}{4}\left(A_x + A_y\right)\sin 2\theta$$

$$b_6 = -\frac{39}{16}\left({A_x}^2 + {A_y}^2 + {A_z}^2\right) - \frac{1}{8}\left(13{A_x}^2 + 6A_xA_y - 13{A_y}^2 - 3A_xA_z + 3A_yA_z - 26{A_z}^2\right)\cos 2\theta$$
$$+ \frac{1}{16}\left(13{A_x}^2 - 12A_xA_y + 13{A_y}^2 + 6A_xA_z - 6A_yA_z - 26{A_z}^2\right)\cos 4\theta$$
$$+ \frac{\sqrt{3}}{8}\left(13{A_x}^2 - 13{A_y}^2 + 3A_xA_z + 3A_yA_z\right)\sin 2\theta$$
$$+ \frac{\sqrt{3}}{16}\left(13{A_x}^2 - 13{A_y}^2 - 6A_xA_z - 6A_yA_z\right)\sin 4\theta$$

$$c_6 = \frac{5}{64}\left(21{A_x}^3 - 9{A_x}^2A_y + 9A_x{A_y}^2 - 21{A_y}^3 - 9{A_x}^2A_z + 28A_xA_yA_z - 9{A_y}^2A_z + 9A_x{A_z}^2 - 9A_y{A_z}^2\right.$$
$$\left. - 21{A_z}^3\right)$$
$$+ \frac{45}{64}\left(2{A_x}^3 - 3{A_x}^2A_y + 3A_x{A_y}^2 - 2{A_y}^3 + 3{A_x}^2A_z + 3{A_y}^2A_z + 4{A_z}^3\right)\cos 2\theta$$
$$+ \frac{45}{64}\left(-{A_x}^3 - 3{A_x}^2A_y + 3A_x{A_y}^2 + {A_y}^3 + 3{A_x}^2A_z + 3{A_y}^2A_z - 2{A_z}^3\right)\cos 4\theta$$
$$+ \frac{5}{64}\left(-6{A_x}^3 - 9{A_x}^2A_y + 9A_x{A_y}^2 + 6{A_y}^3 - 9{A_x}^2A_z + 28A_xA_yA_z - 9{A_y}^2A_z\right.$$
$$\left. + 9A_x{A_z}^2 - 9A_y{A_z}^2 + 6{A_z}^3\right)\cos 6\theta$$
$$+ \frac{45\sqrt{3}}{64}\left(-2{A_x}^3 - {A_x}^2A_y - A_x{A_y}^2 - 2{A_y}^3 + {A_x}^2A_z - {A_y}^2A_z - 2A_x{A_z}^2\right.$$
$$\left. - 2A_y{A_z}^2\right)\sin 2\theta$$
$$+ \frac{45\sqrt{3}}{64}\left(-{A_x}^3 + {A_x}^2A_y + A_x{A_y}^2 - {A_y}^3 - {A_x}^2A_z + {A_y}^2A_z + 2A_x{A_z}^2\right.$$
$$\left. + 2A_y{A_z}^2\right)\sin 4\theta$$
$$+ \frac{45\sqrt{3}}{64}\left({A_x}^2A_y + A_x{A_y}^2 - {A_x}^2A_z + {A_y}^2A_z - A_x{A_z}^2 - A_y{A_z}^2\right)\sin 6\theta$$

$$a_7 = \frac{3}{4}\left(-A_x - A_y - A_z\right) + \frac{3}{4}\left(-A_x - A_y + 2A_z\right)\cos 2\theta + \frac{3\sqrt{3}}{4}\left(A_x - A_y\right)\sin 2\theta$$

$$b_7 = -\frac{39}{16}\left({A_x}^2 + {A_y}^2 + {A_z}^2\right) - \frac{1}{8}\left(13{A_x}^2 - 6A_xA_y + 13{A_y}^2 + 3A_xA_z + 3A_yA_z - 26{A_z}^2\right)\cos 2\theta$$
$$+ \frac{1}{16}\left(13{A_x}^2 + 12A_xA_y + 13{A_y}^2 - 6A_xA_z - 6A_yA_z - 26{A_z}^2\right)\cos 4\theta$$
$$+ \frac{\sqrt{3}}{8}\left(13{A_x}^2 - 13{A_y}^2 - 3A_xA_z + 3A_yA_z\right)\sin 2\theta$$
$$+ \frac{\sqrt{3}}{16}\left(13{A_x}^2 - 13{A_y}^2 + 6A_xA_z - 6A_yA_z\right)\sin 4\theta$$

$$c_7 = \frac{5}{64}\left(21{A_x}^3 + 9{A_x}^2A_y + 9A_x{A_y}^2 + 21{A_y}^3 + 9{A_x}^2A_z + 28A_xA_yA_z + 9{A_y}^2A_z + 9A_x{A_z}^2 + 9A_y{A_z}^2 + 21{A_z}^3\right)$$
$$+ \frac{45}{64}\left(2{A_x}^3 + 3{A_x}^2A_y + 3A_x{A_y}^2 + 2{A_y}^3 - 3{A_x}^2A_z - 3{A_y}^2A_z - 4{A_z}^3\right)\cos 2\theta$$
$$+ \frac{45}{64}\left(-{A_x}^3 + 3{A_x}^2A_y + 3A_x{A_y}^2 - {A_y}^3 - 3{A_x}^2A_z - 3{A_y}^2A_z + 2{A_z}^3\right)\cos 4\theta$$
$$+ \frac{5}{64}\left(-6{A_x}^3 + 9{A_x}^2A_y + 9A_x{A_y}^2 - 6{A_y}^3 + 9{A_x}^2A_z + 28A_xA_yA_z + 9{A_y}^2A_z + 9A_x{A_z}^2 + 9A_y{A_z}^2 - 6{A_z}^3\right)\cos 6\theta$$
$$+ \frac{45\sqrt{3}}{64}\left(-2{A_x}^3 + {A_x}^2A_y - A_x{A_y}^2 + 2{A_y}^3 - {A_x}^2A_z + {A_y}^2A_z - 2A_x{A_z}^2 + 2A_y{A_z}^2\right)\sin 2\theta$$
$$+ \frac{45\sqrt{3}}{64}\left(-{A_x}^3 - {A_x}^2A_y + A_x{A_y}^2 + {A_y}^3 + {A_x}^2A_z - {A_y}^2A_z + 2A_x{A_z}^2 - 2A_y{A_z}^2\right)\sin 4\theta$$
$$+ \frac{45\sqrt{3}}{64}\left(-{A_x}^2A_y + A_x{A_y}^2 + {A_x}^2A_z - {A_y}^2A_z - A_x{A_z}^2 + A_y{A_z}^2\right)\sin 6\theta$$

$$a_8 = \frac{3}{4}\left(A_x + A_y - A_z\right) + \frac{3}{4}\left(A_x + A_y + 2A_z\right)\cos 2\theta - \frac{3\sqrt{3}}{4}\left(A_x - A_y\right)\sin 2\theta$$

$$b_8 = -\frac{39}{16}\left({A_x}^2 + {A_y}^2 + {A_z}^2\right) - \frac{1}{8}\left(13{A_x}^2 - 6A_xA_y + 13{A_y}^2 - 3A_xA_z - 3A_yA_z - 26{A_z}^2\right)\cos 2\theta$$
$$+ \frac{1}{16}\left(13{A_x}^2 + 12A_xA_y + 13{A_y}^2 + 6A_xA_z + 6A_yA_z - 26{A_z}^2\right)\cos 4\theta$$
$$+ \frac{\sqrt{3}}{8}\left(13{A_x}^2 - 13{A_y}^2 + 3A_xA_z - 3A_yA_z\right)\sin 2\theta$$
$$+ \frac{\sqrt{3}}{16}\left(13{A_x}^2 - 13{A_y}^2 - 6A_xA_z + 6A_yA_z\right)\sin 4\theta$$

$$c_8 = \frac{5}{64}\left(-21{A_x}^3 - 9{A_x}^2A_y - 9A_x{A_y}^2 - 21{A_y}^3 + 9{A_x}^2A_z + 28A_xA_yA_z + 9{A_y}^2A_z - 9A_x{A_z}^2 - 9A_y{A_z}^2 + 21{A_z}^3\right)$$
$$+ \frac{45}{64}\left(-2{A_x}^3 - 3{A_x}^2A_y - 3A_x{A_y}^2 - 2{A_y}^3 - 3{A_x}^2A_z - 3{A_y}^2A_z - 4{A_z}^3\right)\cos 2\theta$$
$$+ \frac{45}{64}\left({A_x}^3 - 3{A_x}^2A_y - 3A_x{A_y}^2 + {A_y}^3 - 3{A_x}^2A_z - 3{A_y}^2A_z + 2{A_z}^3\right)\cos 4\theta$$
$$+ \frac{5}{64}\left(6{A_x}^3 - 9{A_x}^2A_y - 9A_x{A_y}^2 + 6{A_y}^3 + 9{A_x}^2A_z + 28A_xA_yA_z + 9{A_y}^2A_z - 9A_x{A_z}^2 - 9A_y{A_z}^2 - 6{A_z}^3\right)\cos 6\theta$$
$$+ \frac{45\sqrt{3}}{64}\left(2{A_x}^3 - {A_x}^2A_y + A_x{A_y}^2 - 2{A_y}^3 - {A_x}^2A_z + {A_y}^2A_z + 2A_x{A_z}^2 - 2A_y{A_z}^2\right)\sin 2\theta$$
$$+ \frac{45\sqrt{3}}{64}\left({A_x}^3 + {A_x}^2A_y - A_x{A_y}^2 - {A_y}^3 + {A_x}^2A_z - {A_y}^2A_z - 2A_x{A_z}^2 + 2A_y{A_z}^2\right)\sin 4\theta$$
$$+ \frac{45\sqrt{3}}{64}\left({A_x}^2A_y - A_x{A_y}^2 + {A_x}^2A_z - {A_y}^2A_z + A_x{A_z}^2 - A_y{A_z}^2\right)\sin 6\theta$$

Similarly to the coefficients for $i = 2$, 3 and 4, we note that those for $i = 6$, 7 and 8 can be obtained from $\{a_5, b_5, c_5, d_5\}$ with replacing A_x and A_y to $-A_x$ and $-A_y$, A_x and A_z to $-A_x$ and $-A_z$, and A_y and A_z to $-A_y$ and $-A_z$, respectively.

The second-order correction for the energy of the hyperfine structure Hamiltonian $\varepsilon_{\sigma,\alpha}^{(02)}$ $(\sigma = +,-;\ \alpha = 1,\cdots,12)$ can be written as

$$\varepsilon_{\pm,\alpha}^{(02)} = \sum_{\beta=1}^{4} \frac{|\langle \pm,\alpha|H_{\mathrm{hfs}}|\mp,\beta\rangle|^2}{\varepsilon_{\pm}^{(0)} - \varepsilon_{\mp}^{(0)}}$$

Both the upper and lower signs should be chosen in the double sign. The non-zero matrix elements of H_{hfs} expanded to the basis belonging different eigenspaces are given in the following:

$$\langle +,1|H_{\mathrm{hfs}}|-,2\rangle = \langle -,2|H_{\mathrm{hfs}}|+,1\rangle = \langle +,5|H_{\mathrm{hfs}}|-,6\rangle = \langle -,6|H_{\mathrm{hfs}}|+,5\rangle = \langle -,7|H_{\mathrm{hfs}}|+,8\rangle = \langle +,8|H_{\mathrm{hfs}}|-,7\rangle$$
$$= \langle -,11|H_{\mathrm{hfs}}|+,12\rangle = \langle +,12|H_{\mathrm{hfs}}|-,11\rangle$$
$$= \frac{\sqrt{15}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{5}}{4}\left(A_x + A_y\right)\sin 2\theta$$

$$\langle +,2|H_{\mathrm{hfs}}|-,1\rangle = \langle -,1|H_{\mathrm{hfs}}|+,2\rangle = \langle +,6|H_{\mathrm{hfs}}|-,5\rangle = \langle -,5|H_{\mathrm{hfs}}|+,6\rangle = \langle -,8|H_{\mathrm{hfs}}|+,7\rangle = \langle +,7|H_{\mathrm{hfs}}|-,8\rangle$$
$$= \langle -,12|H_{\mathrm{hfs}}|+,11\rangle = \langle +,11|H_{\mathrm{hfs}}|-,12\rangle$$
$$= \frac{\sqrt{15}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{5}}{4}\left(A_x - A_y\right)\sin 2\theta$$

$$\langle +,2|H_{\mathrm{hfs}}|-,3\rangle = \langle -,3|H_{\mathrm{hfs}}|+,2\rangle = \langle +,4|H_{\mathrm{hfs}}|-,5\rangle = \langle -,5|H_{\mathrm{hfs}}|+,4\rangle = \langle -,8|H_{\mathrm{hfs}}|+,9\rangle = \langle +,9|H_{\mathrm{hfs}}|-,8\rangle$$
$$= \langle -,10|H_{\mathrm{hfs}}|+,11\rangle = \langle +,11|H_{\mathrm{hfs}}|-,10\rangle = \frac{\sqrt{6}}{2}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{2}}{2}\left(A_x + A_y\right)\sin 2\theta$$

$$\langle +,3|H_{\mathrm{hfs}}|-,2\rangle = \langle -,2|H_{\mathrm{hfs}}|+,3\rangle = \langle +,5|H_{\mathrm{hfs}}|-,4\rangle = \langle -,4|H_{\mathrm{hfs}}|+,5\rangle = \langle -,9|H_{\mathrm{hfs}}|+,8\rangle = \langle +,8|H_{\mathrm{hfs}}|-,9\rangle$$

$$= \langle -,11|H_{\mathrm{hfs}}|+,10\rangle = \langle +,10|H_{\mathrm{hfs}}|-,11\rangle = \frac{\sqrt{6}}{2}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{2}}{2}\left(A_x - A_y\right)\sin 2\theta$$

$$\langle +,3|H_{\mathrm{hfs}}|-,4\rangle = \langle -,4|H_{\mathrm{hfs}}|+,3\rangle = \langle -,9|H_{\mathrm{hfs}}|+,10\rangle = \langle +,10|H_{\mathrm{hfs}}|-,9\rangle$$

$$= \frac{3\sqrt{3}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{3}{4}\left(A_x + A_y\right)\sin 2\theta$$

$$\langle +,4|H_{\mathrm{hfs}}|-,3\rangle = \langle -,3|H_{\mathrm{hfs}}|+,4\rangle = \langle -,10|H_{\mathrm{hfs}}|+,9\rangle = \langle +,9|H_{\mathrm{hfs}}|-,10\rangle$$

$$= \frac{3\sqrt{3}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{3}{4}\left(A_x - A_y\right)\sin 2\theta$$

$$\langle +,1|H_{\mathrm{hfs}}|-,7\rangle = \langle -,7|H_{\mathrm{hfs}}|+,1\rangle = \langle -,6|H_{\mathrm{hfs}}|+,12\rangle = \langle +,12|H_{\mathrm{hfs}}|-,6\rangle = -\frac{5}{2}A_z\sin 2\theta$$

$$\langle +,2|H_{\mathrm{hfs}}|-,8\rangle = \langle -,8|H_{\mathrm{hfs}}|+,2\rangle = \langle -,5|H_{\mathrm{hfs}}|+,11\rangle = \langle +,11|H_{\mathrm{hfs}}|-,5\rangle = -\frac{3}{2}A_z\sin 2\theta$$

$$\langle +,3|H_{\mathrm{hfs}}|-,9\rangle = \langle -,9|H_{\mathrm{hfs}}|+,3\rangle = \langle -,4|H_{\mathrm{hfs}}|+,10\rangle = \langle +,10|H_{\mathrm{hfs}}|-,4\rangle = -\frac{A_z}{2}\sin 2\theta$$

$$\langle +,4|H_{\mathrm{hfs}}|-,10\rangle = \langle -,10|H_{\mathrm{hfs}}|+,4\rangle = \langle -,3|H_{\mathrm{hfs}}|+,9\rangle = \langle +,9|H_{\mathrm{hfs}}|-,3\rangle = \frac{A_z}{2}\sin 2\theta$$

$$\langle +,5|H_{\mathrm{hfs}}|-,11\rangle = \langle -,11|H_{\mathrm{hfs}}|+,5\rangle = \langle -,2|H_{\mathrm{hfs}}|+,8\rangle = \langle +,8|H_{\mathrm{hfs}}|-,2\rangle = \frac{3}{2}A_z\sin 2\theta$$

$$\langle +,6|H_{\mathrm{hfs}}|-,12\rangle = \langle -,12|H_{\mathrm{hfs}}|+,6\rangle = \langle -,1|H_{\mathrm{hfs}}|+,7\rangle = \langle +,7|H_{\mathrm{hfs}}|-,1\rangle = \frac{5}{2}A_z\sin 2\theta$$

Therefore, the second-order corrections for the energy, $\varepsilon_{\sigma,\alpha}^{(02)}$ $(\sigma = +,-;\ \alpha = 1,\cdots,12)$ are in the following:

$$\varepsilon_{+,1}^{(02)} = \frac{|\langle -,2|H_{\mathrm{hfs}}|+,1\rangle|^2 + |\langle -,7|H_{\mathrm{hfs}}|+,1\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{\sqrt{15}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{5}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \frac{25}{4}{A_z}^2\sin^2 2\theta\right\}$$

$$\varepsilon_{+,2}^{(02)} = \frac{|\langle -,1|H_{\mathrm{hfs}}|+,2\rangle|^2 + |\langle -,3|H_{\mathrm{hfs}}|+,2\rangle|^2 + |\langle -,8|H_{\mathrm{hfs}}|+,2\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{\sqrt{15}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{5}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2\right.$$

$$\left. + \left[\frac{\sqrt{6}}{2}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{2}}{2}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \frac{9}{4}{A_z}^2\sin^2 2\theta\right\}$$

$$\varepsilon_{+,3}^{(02)} = \frac{|\langle -,2|H_{\mathrm{hfs}}|+,3\rangle|^2 + |\langle -,4|H_{\mathrm{hfs}}|+,3\rangle|^2 + |\langle -,9|H_{\mathrm{hfs}}|+,3\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{\sqrt{6}}{2}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{2}}{2}\left(A_x - A_y\right)\sin 2\theta\right]^2 + \left[\frac{3\sqrt{3}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{3}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \frac{{A_z}^2}{4}\sin^2 2\theta\right\}$$

$$\varepsilon_{+,4}^{(02)} = \frac{|\langle -,3|H_{\mathrm{hfs}}|+,4\rangle|^2 + |\langle -,5|H_{\mathrm{hfs}}|+,4\rangle|^2 + |\langle -,10|H_{\mathrm{hfs}}|+,4\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{3\sqrt{3}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{3}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2 + \left[\frac{\sqrt{6}}{2}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{2}}{2}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \frac{{A_z}^2}{4}\sin^2 2\theta\right\}$$

$$\varepsilon_{+,5}^{(02)} = \frac{|\langle -,4|H_{\mathrm{hfs}}|+,5\rangle|^2 + |\langle -,6|H_{\mathrm{hfs}}|+,5\rangle|^2 + |\langle -,11|H_{\mathrm{hfs}}|+,5\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{\sqrt{6}}{2}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{2}}{2}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \left[\frac{\sqrt{15}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{5}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \frac{9}{4}{A_z}^2\sin^2 2\theta\right\}$$

$$\varepsilon_{+,6}^{(02)} = \frac{|\langle -,5|H_{\mathrm{hfs}}|+,6\rangle|^2 + |\langle -,12|H_{\mathrm{hfs}}|+,6\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{\sqrt{15}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{5}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \frac{25}{4}{A_z}^2\sin^2 2\theta\right\}$$

$$\varepsilon_{+,7}^{(02)} = \frac{+|\langle -,1|H_{\mathrm{hfs}}|+,7\rangle|^2 + |\langle -,8|H_{\mathrm{hfs}}|+,7\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{25}{4}{A_z}^2\sin^2 2\theta + \left[\frac{\sqrt{15}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{5}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{+,8}^{(02)} = \frac{|\langle -,2|H_{\mathrm{hfs}}|+,8\rangle|^2 + |\langle -,7|H_{\mathrm{hfs}}|+,8\rangle|^2 + |\langle -,9|H_{\mathrm{hfs}}|+,8\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{9}{4}{A_z}^2\sin^2 2\theta + \left[\frac{\sqrt{15}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{5}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \left[\frac{\sqrt{6}}{2}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{2}}{2}\left(A_x - A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{+,9}^{(02)} = \frac{|\langle -,3|H_{\mathrm{hfs}}|+,9\rangle|^2 + |\langle -,8|H_{\mathrm{hfs}}|+,9\rangle|^2 + |\langle -,10|H_{\mathrm{hfs}}|+,9\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{{A_z}^2}{4}\sin^2 2\theta + \left[\frac{\sqrt{6}}{2}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{2}}{2}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \left[\frac{3\sqrt{3}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{3}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{+,10}^{(02)} = \frac{|\langle -,4|H_{\text{hfs}}|+,10\rangle|^2 + |\langle -,9|H_{\text{hfs}}|+,10\rangle|^2 + |\langle -,11|H_{\text{hfs}}|+,10\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{{A_z}^2}{4}\sin^2 2\theta + \left[\frac{3\sqrt{3}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{3}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \left[\frac{\sqrt{6}}{2}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{2}}{2}\left(A_x - A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{+,11}^{(02)} = \frac{|\langle -,5|H_{\text{hfs}}|+,11\rangle|^2 + |\langle -,10|H_{\text{hfs}}|+,11\rangle|^2 + |\langle -,12|H_{\text{hfs}}|+,11\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{9}{4}{A_z}^2\sin^2 2\theta + \left[\frac{\sqrt{6}}{2}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{2}}{2}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \left[\frac{\sqrt{15}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{5}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{+,12}^{(02)} = \frac{|\langle -,6|H_{\text{hfs}}|+,12\rangle|^2 + |\langle -,11|H_{\text{hfs}}|+,12\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{25}{4}{A_z}^2\sin^2 2\theta + \left[\frac{\sqrt{15}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{5}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{-,1}^{(02)} = \frac{|\langle +,2|H_{\text{hfs}}|-,1\rangle|^2 + |\langle +,7|H_{\text{hfs}}|-,1\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\left[\frac{\sqrt{15}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{5}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2 + \frac{25}{4}{A_z}^2\sin^2 2\theta\right\}$$

$$\varepsilon_{-,2}^{(02)} = \frac{|\langle +,1|H_{\text{hfs}}|-,2\rangle|^2 + |\langle +,3|H_{\text{hfs}}|-,2\rangle|^2 + |\langle +,8|H_{\text{hfs}}|-,2\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\left[\frac{\sqrt{15}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{5}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \left[\frac{\sqrt{6}}{2}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{2}}{2}\left(A_x - A_y\right)\sin 2\theta\right]^2 + \frac{9}{4}{A_z}^2\sin^2 2\theta\right\}$$

$$\varepsilon_{-,3}^{(02)} = \frac{|\langle +,2|H_{\text{hfs}}|-,3\rangle|^2 + |\langle +,4|H_{\text{hfs}}|-,3\rangle|^2 + |\langle +,9|H_{\text{hfs}}|-,3\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\left[\frac{\sqrt{6}}{2}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{2}}{2}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \left[\frac{3\sqrt{3}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{3}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2 + \frac{{A_z}^2}{4}\sin^2 2\theta\right\}$$

$$\varepsilon_{-,4}^{(02)} = \frac{|\langle +,3|H_{\text{hfs}}|-,4\rangle|^2 + |\langle +,5|H_{\text{hfs}}|-,4\rangle|^2 + |\langle +,10|H_{\text{hfs}}|-,4\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\left[\frac{3\sqrt{3}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{3}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \left[\frac{\sqrt{6}}{2}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{2}}{2}\left(A_x - A_y\right)\sin 2\theta\right]^2 + \frac{{A_z}^2}{4}\sin^2 2\theta\right\}$$

$$\varepsilon_{-,5}^{(02)} = \frac{|\langle +,4|H_{\text{hfs}}|-,5\rangle|^2 + |\langle +,6|H_{\text{hfs}}|-,5\rangle|^2 + |\langle +,11|H_{\text{hfs}}|-,5\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\left[\frac{\sqrt{6}}{2}(A_x - A_y)\cos 2\theta + \frac{\sqrt{2}}{2}(A_x + A_y)\sin 2\theta\right]^2 + \left[\frac{\sqrt{15}}{4}(A_x + A_y)\cos 2\theta + \frac{\sqrt{5}}{4}(A_x - A_y)\sin 2\theta\right]^2 + \frac{9}{4}{A_z}^2\sin^2 2\theta\right\}$$

$$\varepsilon_{-,6}^{(02)} = \frac{|\langle +,5|H_{\text{hfs}}|-,6\rangle|^2 + |\langle +,12|H_{\text{hfs}}|-,6\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\left[\frac{\sqrt{15}}{4}(A_x - A_y)\cos 2\theta + \frac{\sqrt{5}}{4}(A_x + A_y)\sin 2\theta\right]^2 + \frac{25}{4}{A_z}^2\sin^2 2\theta\right\}$$

$$\varepsilon_{-,7}^{(02)} = \frac{|\langle +,1|H_{\text{hfs}}|-,7\rangle|^2 + |\langle +,8|H_{\text{hfs}}|-,7\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\frac{25}{4}{A_z}^2\sin^2 2\theta + \left[\frac{\sqrt{15}}{4}(A_x - A_y)\cos 2\theta + \frac{\sqrt{5}}{4}(A_x + A_y)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{-,8}^{(02)} = \frac{|\langle +,2|H_{\text{hfs}}|-,8\rangle|^2 + |\langle +,7|H_{\text{hfs}}|-,8\rangle|^2 + |\langle +,9|H_{\text{hfs}}|-,8\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\frac{9}{4}{A_z}^2\sin^2 2\theta + \left[\frac{\sqrt{15}}{4}(A_x + A_y)\cos 2\theta + \frac{\sqrt{5}}{4}(A_x - A_y)\sin 2\theta\right]^2 + \left[\frac{\sqrt{6}}{2}(A_x - A_y)\cos 2\theta + \frac{\sqrt{2}}{2}(A_x + A_y)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{-,9}^{(02)} = \frac{|\langle +,3|H_{\text{hfs}}|-,9\rangle|^2 + |\langle +,8|H_{\text{hfs}}|-,9\rangle|^2 + |\langle +,10|H_{\text{hfs}}|-,9\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\frac{{A_z}^2}{4}\sin^2 2\theta + \left[\frac{\sqrt{6}}{2}(A_x + A_y)\cos 2\theta + \frac{\sqrt{2}}{2}(A_x - A_y)\sin 2\theta\right]^2 + \left[\frac{3\sqrt{3}}{4}(A_x - A_y)\cos 2\theta + \frac{3}{4}(A_x + A_y)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{-,10}^{(02)} = \frac{|\langle +,4|H_{\text{hfs}}|-,10\rangle|^2 + |\langle +,9|H_{\text{hfs}}|-,10\rangle|^2 + |\langle +,11|H_{\text{hfs}}|-,10\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\frac{{A_z}^2}{4}\sin^2 2\theta + \left[\frac{3\sqrt{3}}{4}(A_x + A_y)\cos 2\theta + \frac{3}{4}(A_x - A_y)\sin 2\theta\right]^2 + \left[\frac{\sqrt{6}}{2}(A_x - A_y)\cos 2\theta + \frac{\sqrt{2}}{2}(A_x + A_y)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{-,11}^{(02)} = \frac{|\langle +,5|H_{\text{hfs}}|-,11\rangle|^2 + |\langle +,10|H_{\text{hfs}}|-,11\rangle|^2 + |\langle +,12|H_{\text{hfs}}|-,11\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\frac{9}{4}{A_z}^2\sin^2 2\theta + \left[\frac{\sqrt{6}}{2}(A_x + A_y)\cos 2\theta + \frac{\sqrt{2}}{2}(A_x - A_y)\sin 2\theta\right]^2 + \left[\frac{\sqrt{15}}{4}(A_x - A_y)\cos 2\theta + \frac{\sqrt{5}}{4}(A_x + A_y)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{-,12}^{(02)} = \frac{|\langle +,6|H_{\mathrm{hfs}}|-,12\rangle|^2 + |\langle +,11|H_{\mathrm{hfs}}|-,12\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\frac{25}{4}{A_z}^2 \sin^2 2\theta + \left[\frac{\sqrt{15}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{5}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2\right\}$$

The first- and second-order corrections for the energy of the electron Zeeman Hamiltonian are the same as for the I = 1/2 case. In order to obtain the cross terms, let us remind the non-diagonal elements for the electron Zeeman Hamiltonian.

$$\langle +,1|H_{\mathrm{eZ}}|-,7\rangle = \langle -,7|H_{\mathrm{eZ}}|+,1\rangle = \langle +,2|H_{\mathrm{eZ}}|-,8\rangle = \langle -,8|H_{\mathrm{eZ}}|+,2\rangle = \langle +,3|H_{\mathrm{eZ}}|-,9\rangle = \langle -,9|H_{\mathrm{eZ}}|+,3\rangle$$
$$= \langle +,4|H_{\mathrm{eZ}}|-,10\rangle = \langle -,10|H_{\mathrm{eZ}}|+,2\rangle = \langle +,5|H_{\mathrm{eZ}}|-,11\rangle = \langle -,11|H_{\mathrm{eZ}}|+,5\rangle$$
$$= \langle +,6|H_{\mathrm{eZ}}|-,12\rangle = \langle -,12|H_{\mathrm{eZ}}|+,6\rangle = -g_z\beta B\sin 2\theta$$

$$\langle -,1|H_{\mathrm{eZ}}|+,7\rangle = \langle +,7|H_{\mathrm{eZ}}|-,1\rangle = \langle -,2|H_{\mathrm{eZ}}|+,8\rangle = \langle +,8|H_{\mathrm{eZ}}|-,2\rangle = \langle -,3|H_{\mathrm{eZ}}|+,9\rangle = \langle +,9|H_{\mathrm{eZ}}|-,3\rangle$$
$$= \langle -,4|H_{\mathrm{eZ}}|+,10\rangle = \langle +,10|H_{\mathrm{eZ}}|-,4\rangle = \langle -,5|H_{\mathrm{eZ}}|+,11\rangle = \langle +,1|H_{\mathrm{eZ}}|-,5\rangle$$
$$= \langle -,6|H_{\mathrm{eZ}}|+,12\rangle = \langle +,12|H_{\mathrm{eZ}}|-,6\rangle = g_z\beta B\sin 2\theta$$

Therefore, the cross terms can be calculated as follows:

$$\varepsilon_{+,1}^{(11)} = \frac{\langle +,1|H_{\mathrm{eZ}}|-,7\rangle\langle -,7|H_{\mathrm{hfs}}|+,1\rangle + \langle +,1|H_{\mathrm{hfs}}|-,7\rangle\langle -,7|H_{\mathrm{eZ}}|+,1\rangle}{\varepsilon_{+}^{(0)} - \varepsilon_{-}^{(0)}} = \frac{25g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,2}^{(11)} = \frac{\langle +,2|H_{\mathrm{eZ}}|-,8\rangle\langle -,8|H_{\mathrm{hfs}}|+,2\rangle + \langle +,2|H_{\mathrm{hfs}}|-,8\rangle\langle -,8|H_{\mathrm{eZ}}|+,2\rangle}{\varepsilon_{+}^{(0)} - \varepsilon_{-}^{(0)}} = \frac{9g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,3}^{(11)} = \frac{\langle +,3|H_{\mathrm{eZ}}|-,9\rangle\langle -,9|H_{\mathrm{hfs}}|+,3\rangle + \langle +,3|H_{\mathrm{hfs}}|-,9\rangle\langle -,9|H_{\mathrm{eZ}}|+,3\rangle}{\varepsilon_{+}^{(0)} - \varepsilon_{-}^{(0)}} = \frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,4}^{(11)} = \frac{\langle +,4|H_{\mathrm{eZ}}|-,10\rangle\langle -,10|H_{\mathrm{hfs}}|+,4\rangle + \langle +,4|H_{\mathrm{hfs}}|-,10\rangle\langle -,10|H_{\mathrm{eZ}}|+,4\rangle}{\varepsilon_{+}^{(0)} - \varepsilon_{-}^{(0)}} = -\frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,5}^{(11)} = \frac{\langle +,5|H_{\mathrm{eZ}}|-,11\rangle\langle -,11|H_{\mathrm{hfs}}|+,5\rangle + \langle +,5|H_{\mathrm{hfs}}|-,11\rangle\langle -,11|H_{\mathrm{eZ}}|+,5\rangle}{\varepsilon_{+}^{(0)} - \varepsilon_{-}^{(0)}} = -\frac{9g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,6}^{(11)} = \frac{\langle +,6|H_{\mathrm{eZ}}|-,12\rangle\langle -,12|H_{\mathrm{hfs}}|+,6\rangle + \langle +,6|H_{\mathrm{hfs}}|-,12\rangle\langle -,12|H_{\mathrm{eZ}}|+,6\rangle}{\varepsilon_{+}^{(0)} - \varepsilon_{-}^{(0)}} = -\frac{25g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,1}^{(11)} = \frac{\langle -,1|H_{\mathrm{eZ}}|+,7\rangle\langle +,7|H_{\mathrm{hfs}}|-,1\rangle + \langle -,1|H_{\mathrm{hfs}}|+,7\rangle\langle +,7|H_{\mathrm{eZ}}|-,1\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = -\frac{25g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,2}^{(11)} = \frac{\langle -,2|H_{\mathrm{eZ}}|+,8\rangle\langle +,8|H_{\mathrm{hfs}}|-,2\rangle + \langle -,2|H_{\mathrm{hfs}}|+,8\rangle\langle +,8|H_{\mathrm{eZ}}|-,2\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = -\frac{9g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,3}^{(11)} = \frac{\langle -,3|H_{\mathrm{eZ}}|+,9\rangle\langle +,9|H_{\mathrm{hfs}}|-,3\rangle + \langle -,3|H_{\mathrm{hfs}}|+,9\rangle\langle +,9|H_{\mathrm{eZ}}|-,3\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = -\frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,4}^{(11)} = \frac{\langle -,4|H_{\mathrm{eZ}}|+,10\rangle\langle +,10|H_{\mathrm{hfs}}|-,4\rangle + \langle -,4|H_{\mathrm{hfs}}|+,10\rangle\langle +,10|H_{\mathrm{eZ}}|-,4\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = \frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,5}^{(11)} = \frac{\langle -,5|H_{\mathrm{eZ}}|+,11\rangle\langle +,11|H_{\mathrm{hfs}}|-,5\rangle + \langle -,5|H_{\mathrm{hfs}}|+,11\rangle\langle +,11|H_{\mathrm{eZ}}|-,5\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = \frac{9g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,6}^{(11)} = \frac{\langle -,6|H_{\mathrm{eZ}}|+,12\rangle\langle +,12|H_{\mathrm{hfs}}|-,6\rangle + \langle -,6|H_{\mathrm{hfs}}|+,12\rangle\langle +,12|H_{\mathrm{eZ}}|-,6\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = \frac{25g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,7}^{(11)} = \frac{\langle -,7|H_{\mathrm{eZ}}|+,1\rangle\langle +,1|H_{\mathrm{hfs}}|-,7\rangle + \langle -,7|H_{\mathrm{hfs}}|+,1\rangle\langle +,1|H_{\mathrm{eZ}}|-,7\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = \frac{25 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,8}^{(11)} = \frac{\langle -,8|H_{\mathrm{eZ}}|+,2\rangle\langle +,2|H_{\mathrm{hfs}}|-,8\rangle + \langle -,8|H_{\mathrm{hfs}}|+,2\rangle\langle +,2|H_{\mathrm{eZ}}|-,8\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = \frac{9 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,9}^{(11)} = \frac{\langle -,9|H_{\mathrm{eZ}}|+,3\rangle\langle +,3|H_{\mathrm{hfs}}|-,9\rangle + \langle -,9|H_{\mathrm{hfs}}|+,3\rangle\langle +,3|H_{\mathrm{eZ}}|-,9\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = \frac{g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,10}^{(11)} = \frac{\langle -,10|H_{\mathrm{eZ}}|+,4\rangle\langle +,4|H_{\mathrm{hfs}}|-,10\rangle + \langle -,10|H_{\mathrm{hfs}}|+,4\rangle\langle +,4|H_{\mathrm{eZ}}|-,10\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = -\frac{g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,11}^{(11)} = \frac{\langle -,11|H_{\mathrm{eZ}}|+,5\rangle\langle +,5|H_{\mathrm{hfs}}|-,11\rangle + \langle -,11|H_{\mathrm{hfs}}|+,5\rangle\langle +,5|H_{\mathrm{eZ}}|-,10\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = -\frac{9 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,12}^{(11)} = \frac{\langle -,12|H_{\mathrm{eZ}}|+,6\rangle\langle +,6|H_{\mathrm{hfs}}|-,12\rangle + \langle -,12|H_{\mathrm{hfs}}|+,6\rangle\langle +,6|H_{\mathrm{eZ}}|-,12\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = -\frac{25 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,7}^{(11)} = \frac{\langle +,7|H_{\mathrm{eZ}}|-,1\rangle\langle -,1|H_{\mathrm{hfs}}|+,7\rangle + \langle +,7|H_{\mathrm{hfs}}|-,1\rangle\langle -,1|H_{\mathrm{eZ}}|+,7\rangle}{\varepsilon_{+}^{(0)} - \varepsilon_{-}^{(0)}} = -\frac{25 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,8}^{(11)} = \frac{\langle +,8|H_{\mathrm{eZ}}|-,2\rangle\langle -,2|H_{\mathrm{hfs}}|+,8\rangle + \langle +,8|H_{\mathrm{hfs}}|-,2\rangle\langle -,2|H_{\mathrm{eZ}}|+,8\rangle}{\varepsilon_{+}^{(0)} - \varepsilon_{-}^{(0)}} = -\frac{9 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,9}^{(11)} = \frac{\langle +,9|H_{\mathrm{eZ}}|-,3\rangle\langle -,3|H_{\mathrm{hfs}}|+,9\rangle + \langle +,9|H_{\mathrm{hfs}}|-,3\rangle\langle -,3|H_{\mathrm{eZ}}|+,9\rangle}{\varepsilon_{+}^{(0)} - \varepsilon_{-}^{(0)}} = -\frac{g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,10}^{(11)} = \frac{\langle +,10|H_{\mathrm{eZ}}|-,4\rangle\langle -,4|H_{\mathrm{hfs}}|+,10\rangle + \langle +,10|H_{\mathrm{hfs}}|-,4\rangle\langle -,4|H_{\mathrm{eZ}}|+,10\rangle}{\varepsilon_{+}^{(0)} - \varepsilon_{-}^{(0)}} = \frac{g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,11}^{(11)} = \frac{\langle +,11|H_{\mathrm{eZ}}|-,5\rangle\langle -,5|H_{\mathrm{hfs}}|+,11\rangle + \langle +,11|H_{\mathrm{hfs}}|-,5\rangle\langle -,5|H_{\mathrm{eZ}}|+,11\rangle}{\varepsilon_{+}^{(0)} - \varepsilon_{-}^{(0)}} = \frac{9 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,12}^{(11)} = \frac{\langle +,12|H_{\mathrm{eZ}}|-,6\rangle\langle -,6|H_{\mathrm{hfs}}|+,12\rangle + \langle +,12|H_{\mathrm{hfs}}|-,6\rangle\langle -,6|H_{\mathrm{eZ}}|+,12\rangle}{\varepsilon_{+}^{(0)} - \varepsilon_{-}^{(0)}} = \frac{25 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

Thus, the perturbed energies for the case with $I = 5/2$ for the spin quartet state were explicitly obtained in the second order. To our knowledge the analytical expressions above for the energies in terms of the Zeeman perturbation theory are for the first time given in this work, which are extremely accurate.

4. *I* = 7/2 case

a) Double perturbation approach

The matrix elements of the hyperfine structure Hamiltonian in the basis of $|M_S, M_I\rangle$ are given as follows:

$$\langle M_S', M_I'|H_{\mathrm{hfs}}|M_S, M_I\rangle = \begin{cases} \delta_{M_S M_S'}\delta_{M_I M_I'} M_S M_I A_z \\ \frac{1}{16}\delta_{M_S M_S' \mp 1}\delta_{M_I M_I' \mp 1}\sqrt{15 - 4M_S M_S'}\sqrt{63 - 4M_I M_I'}\left(A_x - A_y\right) \\ \frac{1}{16}\delta_{M_S M_S' \mp 1}\delta_{M_I M_I' \pm 1}\sqrt{15 - 4M_S M_S'}\sqrt{63 - 4M_I M_I'}\left(A_x + A_y\right) \end{cases}$$

The matrix representation of the rank-2 ZFS Hamiltonian is the same in the case of $I = 1/2$, and thus the energy eigenvalues and the spin eigenstates also have already been shown.

Similar to the $I = 1/2$, $3/2$ and $5/2$ cases, $\left\{\left|\varphi_{M_S,M_I}^{(0)}\right\rangle\right\}$ are divided into two subspaces according to the sign of the eigenvalues;

$$\left\{\begin{matrix}\left|\varphi^{(0)}_{+\frac{3}{2},+\frac{7}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{3}{2},+\frac{5}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{3}{2},+\frac{3}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{3}{2},+\frac{1}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{3}{2},-\frac{1}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{3}{2},-\frac{3}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{3}{2},-\frac{5}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{3}{2},-\frac{7}{2}}\right\rangle, \\ \left|\varphi^{(0)}_{-\frac{3}{2},+\frac{7}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{3}{2},+\frac{5}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{3}{2},+\frac{3}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{3}{2},+\frac{1}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{3}{2},-\frac{1}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{3}{2},-\frac{3}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{3}{2},-\frac{5}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{3}{2},-\frac{7}{2}}\right\rangle\end{matrix}\right\} \text{ (eigenvalue } \Delta\text{)}$$

and

$$\left\{\begin{matrix}\left|\varphi^{(0)}_{+\frac{1}{2},+\frac{7}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{1}{2},+\frac{5}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{1}{2},+\frac{3}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{1}{2},+\frac{1}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{1}{2},-\frac{1}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{1}{2},-\frac{3}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{1}{2},-\frac{5}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{1}{2},-\frac{7}{2}}\right\rangle, \\ \left|\varphi^{(0)}_{-\frac{1}{2},-\frac{7}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{1}{2},+\frac{5}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{1}{2},+\frac{3}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{1}{2},+\frac{1}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{1}{2},-\frac{1}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{1}{2},-\frac{3}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{1}{2},-\frac{5}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{1}{2},-\frac{7}{2}}\right\rangle\end{matrix}\right\} \text{ (eigenvalue } -\Delta\text{)}$$

We rewrite the notation of the eigenfunctions as follows for simplicity:

$$\left\{\begin{matrix}\left|\varphi^{(0)}_{+\frac{3}{2},+\frac{7}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{3}{2},+\frac{5}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{3}{2},+\frac{3}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{3}{2},+\frac{1}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{3}{2},-\frac{1}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{3}{2},-\frac{3}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{3}{2},-\frac{5}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{3}{2},-\frac{7}{2}}\right\rangle, \\ \left|\varphi^{(0)}_{-\frac{3}{2},+\frac{7}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{3}{2},+\frac{5}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{3}{2},+\frac{3}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{3}{2},+\frac{1}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{3}{2},-\frac{1}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{3}{2},-\frac{3}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{3}{2},-\frac{5}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{3}{2},-\frac{7}{2}}\right\rangle\end{matrix}\right\}$$

$$\rightarrow \left\{\begin{matrix}|+,1\rangle, |+,2\rangle, |+,3\rangle, |+,4\rangle, |+,5\rangle, |+,6\rangle, |+,7\rangle, |+,8\rangle, \\ |+,9\rangle, |+,10\rangle, |+,11\rangle, |+,12\rangle, |+,13\rangle, |+,14\rangle, |+,15\rangle, |+,16\rangle\end{matrix}\right\}$$

and

$$\left\{\begin{matrix}\left|\varphi^{(0)}_{+\frac{1}{2},+\frac{7}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{1}{2},+\frac{5}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{1}{2},+\frac{3}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{1}{2},+\frac{1}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{1}{2},-\frac{1}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{1}{2},-\frac{3}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{1}{2},-\frac{5}{2}}\right\rangle, \left|\varphi^{(0)}_{+\frac{1}{2},-\frac{7}{2}}\right\rangle, \\ \left|\varphi^{(0)}_{-\frac{1}{2},-\frac{7}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{1}{2},+\frac{5}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{1}{2},+\frac{3}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{1}{2},+\frac{1}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{1}{2},-\frac{1}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{1}{2},-\frac{3}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{1}{2},-\frac{5}{2}}\right\rangle, \left|\varphi^{(0)}_{-\frac{1}{2},-\frac{7}{2}}\right\rangle\end{matrix}\right\}$$

$$\rightarrow \left\{\begin{matrix}|-,1\rangle, |-,2\rangle, |-,3\rangle, |-,4\rangle, |-,5\rangle, |-,6\rangle, |-,7\rangle, |-,8\rangle, \\ |-,9\rangle, |-,10\rangle, |-,11\rangle, |-,12\rangle, |+,13\rangle, |+,14\rangle, |+,15\rangle, |+,16\rangle\end{matrix}\right\}$$

According to the degenerate perturbation theory, the first-order corrections for the energy of the hyperfine structure Hamiltonian ($\varepsilon^{(01)}_{+,\alpha}$ ($\alpha = 1, \cdots, 16$)) which belong to the group of the positive energy eigenvalue are the eigenvalues of the following matrix:

$$H^{+,1}_{\mathrm{hfs}} = \begin{pmatrix} \langle +,1|H_{\mathrm{hfs}}|+,1\rangle & \langle +,1|H_{\mathrm{hfs}}|+,3\rangle & \langle +,1|H_{\mathrm{hfs}}|+,5\rangle & \langle +,1|H_{\mathrm{hfs}}|+,7\rangle & \langle +,1|H_{\mathrm{hfs}}|+,10\rangle & \langle +,1|H_{\mathrm{hfs}}|+,12\rangle & \langle +,1|H_{\mathrm{hfs}}|+,14\rangle & \langle +,1|H_{\mathrm{hfs}}|+,16\rangle \\ \langle +,3|H_{\mathrm{hfs}}|+,1\rangle & \langle +,3|H_{\mathrm{hfs}}|+,3\rangle & \langle +,3|H_{\mathrm{hfs}}|+,5\rangle & \langle +,3|H_{\mathrm{hfs}}|+,7\rangle & \langle +,3|H_{\mathrm{hfs}}|+,10\rangle & \langle +,3|H_{\mathrm{hfs}}|+,12\rangle & \langle +,3|H_{\mathrm{hfs}}|+,14\rangle & \langle +,3|H_{\mathrm{hfs}}|+,16\rangle \\ \langle +,5|H_{\mathrm{hfs}}|+,1\rangle & \langle +,5|H_{\mathrm{hfs}}|+,3\rangle & \langle +,5|H_{\mathrm{hfs}}|+,5\rangle & \langle +,5|H_{\mathrm{hfs}}|+,7\rangle & \langle +,5|H_{\mathrm{hfs}}|+,10\rangle & \langle +,5|H_{\mathrm{hfs}}|+,12\rangle & \langle +,5|H_{\mathrm{hfs}}|+,14\rangle & \langle +,5|H_{\mathrm{hfs}}|+,16\rangle \\ \langle +,7|H_{\mathrm{hfs}}|+,1\rangle & \langle +,7|H_{\mathrm{hfs}}|+,3\rangle & \langle +,7|H_{\mathrm{hfs}}|+,5\rangle & \langle +,7|H_{\mathrm{hfs}}|+,7\rangle & \langle +,7|H_{\mathrm{hfs}}|+,10\rangle & \langle +,7|H_{\mathrm{hfs}}|+,12\rangle & \langle +,7|H_{\mathrm{hfs}}|+,14\rangle & \langle +,7|H_{\mathrm{hfs}}|+,16\rangle \\ \langle +,10|H_{\mathrm{hfs}}|+,1\rangle & \langle +,10|H_{\mathrm{hfs}}|+,3\rangle & \langle +,10|H_{\mathrm{hfs}}|+,5\rangle & \langle +,10|H_{\mathrm{hfs}}|+,7\rangle & \langle +,10|H_{\mathrm{hfs}}|+,10\rangle & \langle +,10|H_{\mathrm{hfs}}|+,12\rangle & \langle +,10|H_{\mathrm{hfs}}|+,14\rangle & \langle +,10|H_{\mathrm{hfs}}|+,16\rangle \\ \langle +,12|H_{\mathrm{hfs}}|+,1\rangle & \langle +,12|H_{\mathrm{hfs}}|+,3\rangle & \langle +,12|H_{\mathrm{hfs}}|+,5\rangle & \langle +,12|H_{\mathrm{hfs}}|+,7\rangle & \langle +,12|H_{\mathrm{hfs}}|+,10\rangle & \langle +,12|H_{\mathrm{hfs}}|+,12\rangle & \langle +,12|H_{\mathrm{hfs}}|+,14\rangle & \langle +,12|H_{\mathrm{hfs}}|+,16\rangle \\ \langle +,14|H_{\mathrm{hfs}}|+,1\rangle & \langle +,14|H_{\mathrm{hfs}}|+,3\rangle & \langle +,14|H_{\mathrm{hfs}}|+,5\rangle & \langle +,14|H_{\mathrm{hfs}}|+,7\rangle & \langle +,14|H_{\mathrm{hfs}}|+,10\rangle & \langle +,14|H_{\mathrm{hfs}}|+,12\rangle & \langle +,14|H_{\mathrm{hfs}}|+,14\rangle & \langle +,14|H_{\mathrm{hfs}}|+,16\rangle \\ \langle +,16|H_{\mathrm{hfs}}|+,1\rangle & \langle +,16|H_{\mathrm{hfs}}|+,3\rangle & \langle +,16|H_{\mathrm{hfs}}|+,5\rangle & \langle +,16|H_{\mathrm{hfs}}|+,7\rangle & \langle +,16|H_{\mathrm{hfs}}|+,10\rangle & \langle +,16|H_{\mathrm{hfs}}|+,12\rangle & \langle +,16|H_{\mathrm{hfs}}|+,14\rangle & \langle +,16|H_{\mathrm{hfs}}|+,16\rangle \end{pmatrix}$$

$$H^{+,2}_{\mathrm{hfs}} = \begin{pmatrix} \langle +,2|H_{\mathrm{hfs}}|+,2\rangle & \langle +,2|H_{\mathrm{hfs}}|+,4\rangle & \langle +,2|H_{\mathrm{hfs}}|+,6\rangle & \langle +,2|H_{\mathrm{hfs}}|+,8\rangle & \langle +,2|H_{\mathrm{hfs}}|+,9\rangle & \langle +,2|H_{\mathrm{hfs}}|+,11\rangle & \langle +,2|H_{\mathrm{hfs}}|+,13\rangle & \langle +,2|H_{\mathrm{hfs}}|+,15\rangle \\ \langle +,4|H_{\mathrm{hfs}}|+,2\rangle & \langle +,4|H_{\mathrm{hfs}}|+,4\rangle & \langle +,4|H_{\mathrm{hfs}}|+,6\rangle & \langle +,4|H_{\mathrm{hfs}}|+,8\rangle & \langle +,4|H_{\mathrm{hfs}}|+,9\rangle & \langle +,4|H_{\mathrm{hfs}}|+,11\rangle & \langle +,4|H_{\mathrm{hfs}}|+,13\rangle & \langle +,4|H_{\mathrm{hfs}}|+,15\rangle \\ \langle +,6|H_{\mathrm{hfs}}|+,2\rangle & \langle +,6|H_{\mathrm{hfs}}|+,4\rangle & \langle +,6|H_{\mathrm{hfs}}|+,6\rangle & \langle +,6|H_{\mathrm{hfs}}|+,8\rangle & \langle +,6|H_{\mathrm{hfs}}|+,9\rangle & \langle +,6|H_{\mathrm{hfs}}|+,11\rangle & \langle +,6|H_{\mathrm{hfs}}|+,13\rangle & \langle +,6|H_{\mathrm{hfs}}|+,15\rangle \\ \langle +,8|H_{\mathrm{hfs}}|+,2\rangle & \langle +,8|H_{\mathrm{hfs}}|+,4\rangle & \langle +,8|H_{\mathrm{hfs}}|+,6\rangle & \langle +,8|H_{\mathrm{hfs}}|+,8\rangle & \langle +,8|H_{\mathrm{hfs}}|+,9\rangle & \langle +,8|H_{\mathrm{hfs}}|+,11\rangle & \langle +,8|H_{\mathrm{hfs}}|+,13\rangle & \langle +,8|H_{\mathrm{hfs}}|+,15\rangle \\ \langle +,9|H_{\mathrm{hfs}}|+,2\rangle & \langle +,9|H_{\mathrm{hfs}}|+,4\rangle & \langle +,9|H_{\mathrm{hfs}}|+,6\rangle & \langle +,9|H_{\mathrm{hfs}}|+,8\rangle & \langle +,9|H_{\mathrm{hfs}}|+,9\rangle & \langle +,9|H_{\mathrm{hfs}}|+,11\rangle & \langle +,9|H_{\mathrm{hfs}}|+,13\rangle & \langle +,9|H_{\mathrm{hfs}}|+,15\rangle \\ \langle +,11|H_{\mathrm{hfs}}|+,2\rangle & \langle +,11|H_{\mathrm{hfs}}|+,4\rangle & \langle +,11|H_{\mathrm{hfs}}|+,6\rangle & \langle +,11|H_{\mathrm{hfs}}|+,8\rangle & \langle +,11|H_{\mathrm{hfs}}|+,9\rangle & \langle +,11|H_{\mathrm{hfs}}|+,11\rangle & \langle +,11|H_{\mathrm{hfs}}|+,13\rangle & \langle +,11|H_{\mathrm{hfs}}|+,15\rangle \\ \langle +,13|H_{\mathrm{hfs}}|+,2\rangle & \langle +,13|H_{\mathrm{hfs}}|+,4\rangle & \langle +,13|H_{\mathrm{hfs}}|+,6\rangle & \langle +,13|H_{\mathrm{hfs}}|+,8\rangle & \langle +,13|H_{\mathrm{hfs}}|+,9\rangle & \langle +,13|H_{\mathrm{hfs}}|+,11\rangle & \langle +,13|H_{\mathrm{hfs}}|+,13\rangle & \langle +,13|H_{\mathrm{hfs}}|+,15\rangle \\ \langle +,15|H_{\mathrm{hfs}}|+,2\rangle & \langle +,15|H_{\mathrm{hfs}}|+,4\rangle & \langle +,15|H_{\mathrm{hfs}}|+,6\rangle & \langle +,15|H_{\mathrm{hfs}}|+,8\rangle & \langle +,15|H_{\mathrm{hfs}}|+,9\rangle & \langle +,15|H_{\mathrm{hfs}}|+,11\rangle & \langle +,15|H_{\mathrm{hfs}}|+,13\rangle & \langle +,15|H_{\mathrm{hfs}}|+,15\rangle \end{pmatrix}$$

$$\langle +,1|H_{\mathrm{hfs}}|+,1\rangle = \langle +,16|H_{\mathrm{hfs}}|+,16\rangle = \frac{7}{4}A_z(1 + 2\cos 2\theta)$$

$$\langle +,2|H_{\mathrm{hfs}}|+,2\rangle = \langle +,15|H_{\mathrm{hfs}}|+,15\rangle = \frac{5}{4}A_z(1 + 2\cos 2\theta)$$

$$\langle +,3|H_{\mathrm{hfs}}|+,3\rangle = \langle +,14|H_{\mathrm{hfs}}|+,14\rangle = \frac{3}{4}A_z(1 + 2\cos 2\theta)$$

$$\langle +,4|H_{\mathrm{hfs}}|+,4\rangle = \langle +,13|H_{\mathrm{hfs}}|+,13\rangle = \frac{1}{4}A_z(1 + 2\cos 2\theta)$$

$$\langle +,5|H_{\mathrm{hfs}}|+,5\rangle = \langle +,12|H_{\mathrm{hfs}}|+,12\rangle = -\frac{1}{4}A_z(1+2\cos 2\theta)$$

$$\langle +,6|H_{\mathrm{hfs}}|+,6\rangle = \langle +,11|H_{\mathrm{hfs}}|+,11\rangle = -\frac{3}{4}A_z(1+2\cos 2\theta)$$

$$\langle +,7|H_{\mathrm{hfs}}|+,7\rangle = \langle +,10|H_{\mathrm{hfs}}|+,10\rangle = -\frac{5}{4}A_z(1+2\cos 2\theta)$$

$$\langle +,8|H_{\mathrm{hfs}}|+,8\rangle = \langle +,9|H_{\mathrm{hfs}}|+,9\rangle = -\frac{7}{4}A_z(1+2\cos 2\theta)$$

$$\langle +,1|H_{\mathrm{hfs}}|+,10\rangle = \langle +,10|H_{\mathrm{hfs}}|+,1\rangle = \langle +,7|H_{\mathrm{hfs}}|+,16\rangle = \langle +,16|H_{\mathrm{hfs}}|+,7\rangle$$
$$= \frac{\sqrt{7}}{2}\sin\theta\left[\sqrt{3}\left(A_x - A_y\right)\cos\theta + \left(A_x + A_y\right)\sin\theta\right]$$

$$\langle +,2|H_{\mathrm{hfs}}|+,9\rangle = \langle +,9|H_{\mathrm{hfs}}|+,2\rangle = \langle +,8|H_{\mathrm{hfs}}|+,15\rangle = \langle +,15|H_{\mathrm{hfs}}|+,8\rangle$$
$$= \frac{\sqrt{7}}{2}\sin\theta\left[\sqrt{3}\left(A_x + A_y\right)\cos\theta + \left(A_x - A_y\right)\sin\theta\right]$$

$$\langle +,2|H_{\mathrm{hfs}}|+,11\rangle = \langle +,11|H_{\mathrm{hfs}}|+,2\rangle = \langle +,6|H_{\mathrm{hfs}}|+,15\rangle = \langle +,15|H_{\mathrm{hfs}}|+,6\rangle$$
$$= \sin\theta\left[3\left(A_x - A_y\right)\cos\theta + \sqrt{3}\left(A_x + A_y\right)\sin\theta\right]$$

$$\langle +,3|H_{\mathrm{hfs}}|+,10\rangle = \langle +,10|H_{\mathrm{hfs}}|+,3\rangle = \langle +,7|H_{\mathrm{hfs}}|+,14\rangle = \langle +,14|H_{\mathrm{hfs}}|+,7\rangle$$
$$= \sin\theta\left[3\left(A_x + A_y\right)\cos\theta + \sqrt{3}\left(A_x - A_y\right)\sin\theta\right]$$

$$\langle +,3|H_{\mathrm{hfs}}|+,12\rangle = \langle +,12|H_{\mathrm{hfs}}|+,3\rangle = \langle +,5|H_{\mathrm{hfs}}|+,14\rangle = \langle +,14|H_{\mathrm{hfs}}|+,5\rangle$$
$$= \frac{\sqrt{5}}{2}\sin\theta\left[3\left(A_x - A_y\right)\cos\theta + \sqrt{3}\left(A_x + A_y\right)\sin\theta\right]$$

$$\langle +,4|H_{\mathrm{hfs}}|+,11\rangle = \langle +,11|H_{\mathrm{hfs}}|+,4\rangle = \langle +,6|H_{\mathrm{hfs}}|+,13\rangle = \langle +,13|H_{\mathrm{hfs}}|+,6\rangle$$
$$= \frac{\sqrt{5}}{2}\sin\theta\left[3\left(A_x + A_y\right)\cos\theta + \sqrt{3}\left(A_x - A_y\right)\sin\theta\right]$$

$$\langle +,4|H_{\mathrm{hfs}}|+,13\rangle = \langle +,13|H_{\mathrm{hfs}}|+,4\rangle = 2\sin\theta\left[\sqrt{3}\left(A_x - A_y\right)\cos\theta + \left(A_x + A_y\right)\sin\theta\right]$$

$$\langle +,5|H_{\mathrm{hfs}}|+,12\rangle = \langle +,12|H_{\mathrm{hfs}}|+,5\rangle = 2\sin\theta\left[\sqrt{3}\left(A_x + A_y\right)\cos\theta + \left(A_x - A_y\right)\sin\theta\right]$$

while the first-order corrections for energy which belong to the group of the negative energy eigenvalue ($\varepsilon_{-,\alpha}^{(01)}$ $(\alpha = 1,\cdots,16)$) are the eigenvalues of the following matrix:

$$H_{\mathrm{hfs}}^{-,1} = \begin{pmatrix}
\langle -,1|H_{\mathrm{hfs}}|-,1\rangle & \langle -,1|H_{\mathrm{hfs}}|-,3\rangle & \langle -,1|H_{\mathrm{hfs}}|-,5\rangle & \langle -,1|H_{\mathrm{hfs}}|-,7\rangle & \langle -,1|H_{\mathrm{hfs}}|-,10\rangle & \langle -,1|H_{\mathrm{hfs}}|-,12\rangle & \langle -,1|H_{\mathrm{hfs}}|-,14\rangle & \langle -,1|H_{\mathrm{hfs}}|-,16\rangle \\
\langle -,3|H_{\mathrm{hfs}}|-,1\rangle & \langle -,3|H_{\mathrm{hfs}}|-,3\rangle & \langle -,3|H_{\mathrm{hfs}}|-,5\rangle & \langle -,3|H_{\mathrm{hfs}}|-,7\rangle & \langle -,3|H_{\mathrm{hfs}}|-,10\rangle & \langle -,3|H_{\mathrm{hfs}}|-,12\rangle & \langle -,3|H_{\mathrm{hfs}}|-,14\rangle & \langle -,3|H_{\mathrm{hfs}}|-,16\rangle \\
\langle -,5|H_{\mathrm{hfs}}|-,1\rangle & \langle -,5|H_{\mathrm{hfs}}|-,3\rangle & \langle -,5|H_{\mathrm{hfs}}|-,5\rangle & \langle -,5|H_{\mathrm{hfs}}|-,7\rangle & \langle -,5|H_{\mathrm{hfs}}|-,10\rangle & \langle -,5|H_{\mathrm{hfs}}|-,12\rangle & \langle -,5|H_{\mathrm{hfs}}|-,14\rangle & \langle -,5|H_{\mathrm{hfs}}|-,16\rangle \\
\langle -,7|H_{\mathrm{hfs}}|-,1\rangle & \langle -,7|H_{\mathrm{hfs}}|-,3\rangle & \langle -,7|H_{\mathrm{hfs}}|-,5\rangle & \langle -,7|H_{\mathrm{hfs}}|-,7\rangle & \langle -,7|H_{\mathrm{hfs}}|-,10\rangle & \langle -,7|H_{\mathrm{hfs}}|-,12\rangle & \langle -,7|H_{\mathrm{hfs}}|-,14\rangle & \langle -,7|H_{\mathrm{hfs}}|-,16\rangle \\
\langle -,10|H_{\mathrm{hfs}}|-,1\rangle & \langle -,10|H_{\mathrm{hfs}}|-,3\rangle & \langle -,10|H_{\mathrm{hfs}}|-,5\rangle & \langle -,10|H_{\mathrm{hfs}}|-,7\rangle & \langle -,10|H_{\mathrm{hfs}}|-,10\rangle & \langle -,10|H_{\mathrm{hfs}}|-,12\rangle & \langle -,10|H_{\mathrm{hfs}}|-,14\rangle & \langle -,10|H_{\mathrm{hfs}}|-,16\rangle \\
\langle -,12|H_{\mathrm{hfs}}|-,1\rangle & \langle -,12|H_{\mathrm{hfs}}|-,3\rangle & \langle -,12|H_{\mathrm{hfs}}|-,5\rangle & \langle -,12|H_{\mathrm{hfs}}|-,7\rangle & \langle -,12|H_{\mathrm{hfs}}|-,10\rangle & \langle -,12|H_{\mathrm{hfs}}|-,12\rangle & \langle -,12|H_{\mathrm{hfs}}|-,14\rangle & \langle -,12|H_{\mathrm{hfs}}|-,16\rangle \\
\langle -,14|H_{\mathrm{hfs}}|-,1\rangle & \langle -,14|H_{\mathrm{hfs}}|-,3\rangle & \langle -,14|H_{\mathrm{hfs}}|-,5\rangle & \langle -,14|H_{\mathrm{hfs}}|-,7\rangle & \langle -,14|H_{\mathrm{hfs}}|-,10\rangle & \langle -,14|H_{\mathrm{hfs}}|-,12\rangle & \langle -,14|H_{\mathrm{hfs}}|-,14\rangle & \langle -,14|H_{\mathrm{hfs}}|-,16\rangle \\
\langle -,16|H_{\mathrm{hfs}}|-,1\rangle & \langle -,16|H_{\mathrm{hfs}}|-,3\rangle & \langle -,16|H_{\mathrm{hfs}}|-,5\rangle & \langle -,16|H_{\mathrm{hfs}}|-,7\rangle & \langle -,16|H_{\mathrm{hfs}}|-,10\rangle & \langle -,16|H_{\mathrm{hfs}}|-,12\rangle & \langle -,16|H_{\mathrm{hfs}}|-,14\rangle & \langle -,16|H_{\mathrm{hfs}}|-,16\rangle
\end{pmatrix}$$

$$H_{\mathrm{hfs}}^{-,2} = \begin{pmatrix}
\langle -,2|H_{\mathrm{hfs}}|-,2\rangle & \langle -,2|H_{\mathrm{hfs}}|-,4\rangle & \langle -,2|H_{\mathrm{hfs}}|-,6\rangle & \langle -,2|H_{\mathrm{hfs}}|-,8\rangle & \langle -,2|H_{\mathrm{hfs}}|-,9\rangle & \langle -,2|H_{\mathrm{hfs}}|-,11\rangle & \langle -,2|H_{\mathrm{hfs}}|-,13\rangle & \langle -,2|H_{\mathrm{hfs}}|-,15\rangle \\
\langle -,4|H_{\mathrm{hfs}}|-,2\rangle & \langle -,4|H_{\mathrm{hfs}}|-,4\rangle & \langle -,4|H_{\mathrm{hfs}}|-,6\rangle & \langle -,4|H_{\mathrm{hfs}}|-,8\rangle & \langle -,4|H_{\mathrm{hfs}}|-,9\rangle & \langle -,4|H_{\mathrm{hfs}}|-,11\rangle & \langle -,4|H_{\mathrm{hfs}}|-,13\rangle & \langle -,4|H_{\mathrm{hfs}}|-,15\rangle \\
\langle -,6|H_{\mathrm{hfs}}|-,2\rangle & \langle -,6|H_{\mathrm{hfs}}|-,4\rangle & \langle -,6|H_{\mathrm{hfs}}|-,6\rangle & \langle -,6|H_{\mathrm{hfs}}|-,8\rangle & \langle -,6|H_{\mathrm{hfs}}|-,9\rangle & \langle -,6|H_{\mathrm{hfs}}|-,11\rangle & \langle -,6|H_{\mathrm{hfs}}|-,13\rangle & \langle -,6|H_{\mathrm{hfs}}|-,15\rangle \\
\langle -,8|H_{\mathrm{hfs}}|-,2\rangle & \langle -,8|H_{\mathrm{hfs}}|-,4\rangle & \langle -,8|H_{\mathrm{hfs}}|-,6\rangle & \langle -,8|H_{\mathrm{hfs}}|-,8\rangle & \langle -,8|H_{\mathrm{hfs}}|-,9\rangle & \langle -,8|H_{\mathrm{hfs}}|-,11\rangle & \langle -,8|H_{\mathrm{hfs}}|-,13\rangle & \langle -,8|H_{\mathrm{hfs}}|-,15\rangle \\
\langle -,9|H_{\mathrm{hfs}}|-,2\rangle & \langle -,9|H_{\mathrm{hfs}}|-,4\rangle & \langle -,9|H_{\mathrm{hfs}}|-,6\rangle & \langle -,9|H_{\mathrm{hfs}}|-,8\rangle & \langle -,9|H_{\mathrm{hfs}}|-,9\rangle & \langle -,9|H_{\mathrm{hfs}}|-,11\rangle & \langle -,9|H_{\mathrm{hfs}}|-,13\rangle & \langle -,9|H_{\mathrm{hfs}}|-,15\rangle \\
\langle -,11|H_{\mathrm{hfs}}|-,2\rangle & \langle -,11|H_{\mathrm{hfs}}|-,4\rangle & \langle -,11|H_{\mathrm{hfs}}|-,6\rangle & \langle -,11|H_{\mathrm{hfs}}|-,8\rangle & \langle -,11|H_{\mathrm{hfs}}|-,9\rangle & \langle -,11|H_{\mathrm{hfs}}|-,11\rangle & \langle -,11|H_{\mathrm{hfs}}|-,13\rangle & \langle -,11|H_{\mathrm{hfs}}|-,15\rangle \\
\langle -,13|H_{\mathrm{hfs}}|-,2\rangle & \langle -,13|H_{\mathrm{hfs}}|-,4\rangle & \langle -,13|H_{\mathrm{hfs}}|-,6\rangle & \langle -,13|H_{\mathrm{hfs}}|-,8\rangle & \langle -,13|H_{\mathrm{hfs}}|-,9\rangle & \langle -,13|H_{\mathrm{hfs}}|-,11\rangle & \langle -,13|H_{\mathrm{hfs}}|-,13\rangle & \langle -,13|H_{\mathrm{hfs}}|-,15\rangle \\
\langle -,15|H_{\mathrm{hfs}}|-,2\rangle & \langle -,15|H_{\mathrm{hfs}}|-,4\rangle & \langle -,15|H_{\mathrm{hfs}}|-,6\rangle & \langle -,15|H_{\mathrm{hfs}}|-,8\rangle & \langle -,15|H_{\mathrm{hfs}}|-,9\rangle & \langle -,15|H_{\mathrm{hfs}}|-,11\rangle & \langle -,15|H_{\mathrm{hfs}}|-,13\rangle & \langle -,15|H_{\mathrm{hfs}}|-,15\rangle
\end{pmatrix}$$

$$\langle -,1|H_{\mathrm{hfs}}|-,1\rangle = \langle -,16|H_{\mathrm{hfs}}|-,16\rangle = \frac{7}{4}A_z(-1+2\cos 2\theta)$$

$$\langle -,2|H_{\mathrm{hfs}}|-,2\rangle = \langle -,15|H_{\mathrm{hfs}}|-,15\rangle = \frac{5}{4}A_z(-1+2\cos 2\theta)$$

$$\langle -,3|H_{\mathrm{hfs}}|-,3\rangle = \langle -,14|H_{\mathrm{hfs}}|-,14\rangle = \frac{3}{4}A_z(-1+2\cos 2\theta)$$

$$\langle -,4|H_{\mathrm{hfs}}|-,4\rangle = \langle -,13|H_{\mathrm{hfs}}|-,13\rangle = \frac{1}{4}A_z(-1+2\cos 2\theta)$$

$$\langle -,5|H_{\mathrm{hfs}}|-,5\rangle = \langle -,12|H_{\mathrm{hfs}}|-,12\rangle = -\frac{1}{4}A_z(-1+2\cos 2\theta)$$

$$\langle -,6|H_{\mathrm{hfs}}|-,6\rangle = \langle -,11|H_{\mathrm{hfs}}|-,11\rangle = -\frac{3}{4}A_z(-1+2\cos 2\theta)$$

$$\langle -,7|H_{\mathrm{hfs}}|-,7\rangle = \langle -,10|H_{\mathrm{hfs}}|-,10\rangle = -\frac{5}{4}A_z(-1+2\cos 2\theta)$$

$$\langle -,8|H_{\mathrm{hfs}}|-,8\rangle = \langle -,9|H_{\mathrm{hfs}}|-,9\rangle = -\frac{7}{4}A_z(-1+2\cos 2\theta)$$

$$\langle -,1|H_{\mathrm{hfs}}|-,10\rangle = \langle -,10|H_{\mathrm{hfs}}|-,1\rangle = \langle -,7|H_{\mathrm{hfs}}|-,16\rangle = \langle -,16|H_{\mathrm{hfs}}|-,7\rangle$$
$$= \frac{\sqrt{7}}{2}\cos\theta\left[\left(A_x - A_y\right)\cos\theta - \sqrt{3}\left(A_x + A_y\right)\sin\theta\right]$$

$$\langle -,2|H_{\mathrm{hfs}}|-,9\rangle = \langle -,9|H_{\mathrm{hfs}}|-,2\rangle = \langle -,8|H_{\mathrm{hfs}}|-,15\rangle = \langle -,15|H_{\mathrm{hfs}}|-,8\rangle$$
$$= \frac{\sqrt{7}}{2}\cos\theta\left[\left(A_x + A_y\right)\cos\theta - \sqrt{3}\left(A_x - A_y\right)\sin\theta\right]$$

$$\langle -,2|H_{\mathrm{hfs}}|-,11\rangle = \langle -,11|H_{\mathrm{hfs}}|-,2\rangle = \langle -,6|H_{\mathrm{hfs}}|-,15\rangle = \langle -,15|H_{\mathrm{hfs}}|-,6\rangle$$
$$= \cos\theta\left[\sqrt{3}\left(A_x - A_y\right)\cos\theta - 3\left(A_x + A_y\right)\sin\theta\right]$$

$$\langle -,3|H_{\mathrm{hfs}}|-,10\rangle = \langle -,10|H_{\mathrm{hfs}}|-,3\rangle = \langle -,7|H_{\mathrm{hfs}}|-,14\rangle = \langle -,14|H_{\mathrm{hfs}}|-,7\rangle$$
$$= \cos\theta\left[\sqrt{3}\left(A_x + A_y\right)\cos\theta - 3\left(A_x - A_y\right)\sin\theta\right]$$

$$\langle -,3|H_{\mathrm{hfs}}|-,12\rangle = \langle -,12|H_{\mathrm{hfs}}|-,3\rangle = \langle -,5|H_{\mathrm{hfs}}|+,14\rangle = \langle -,14|H_{\mathrm{hfs}}|-,5\rangle$$
$$= \frac{\sqrt{5}}{2}\cos\theta\left[\sqrt{3}\left(A_x - A_y\right)\cos\theta - 3\left(A_x + A_y\right)\sin\theta\right]$$

$$\langle -,4|H_{\mathrm{hfs}}|-,11\rangle = \langle -,11|H_{\mathrm{hfs}}|-,4\rangle = \langle -,6|H_{\mathrm{hfs}}|-,13\rangle = \langle -,13|H_{\mathrm{hfs}}|-,6\rangle$$
$$= \frac{\sqrt{5}}{2}\cos\theta\left[\sqrt{3}\left(A_x + A_y\right)\cos\theta - 3\left(A_x - A_y\right)\sin\theta\right]$$

$$\langle -,4|H_{\mathrm{hfs}}|-,13\rangle = \langle -,13|H_{\mathrm{hfs}}|-,4\rangle = 2\cos\theta\left[\left(A_x - A_y\right)\cos\theta - \sqrt{3}\left(A_x + A_y\right)\sin\theta\right]$$
$$\langle -,5|H_{\mathrm{hfs}}|-,12\rangle = \langle -,12|H_{\mathrm{hfs}}|-,5\rangle = 2\cos\theta\left[\left(A_x + A_y\right)\cos\theta - \sqrt{3}\left(A_x - A_y\right)\sin\theta\right]$$

The secular octic equations of $H_{\mathrm{hfs}}^{+,1}$, $H_{\mathrm{hfs}}^{+,2}$, $H_{\mathrm{hfs}}^{-,1}$ and $H_{\mathrm{hfs}}^{-,2}$ can be factorized to two quadratic equations, which is represented as follows:

$$x^4 + a_i x^3 + b_i x^2 + c_i x + d_i = 0\ (i = 1, \cdots, 8)$$

In the equation, i = 1 and 2, 3 and 4, 5 and 6, and 7 and 8 come from $H_{\mathrm{hfs}}^{+,1}$, $H_{\mathrm{hfs}}^{+,2}$, $H_{\mathrm{hfs}}^{-,1}$, $H_{\mathrm{hfs}}^{-,2}$, respectively. In order to eliminate the x^3 term, replace x to $x - a_i/4$

$$x^4 + p_i x^2 + q_i x + r_i = 0$$

with

$$p_i = -\frac{3}{8}{a_i}^2 + b_i$$

$$q_i = \frac{1}{8}{a_3}^3 - \frac{1}{2}a_i b_i + c_i$$

$$r_i = -\frac{3}{256}{a_i}^4 + \frac{1}{16}{a_i}^2 b_i - \frac{1}{4}a_i c_i + d_i$$

The resolvent cubic equation is

$$u^3 + 2p_i u^2 + ({p_i}^2 - 4r_i)u - {q_i}^2 = 0$$

In order to eliminate the u^2 term, replace u to $u - 2p_i/3$ yields

$$u^3 = \frac{1}{3}({p_i}^2 + 12r_i)u + \frac{1}{27}({p_i}^3 - 72p_i r_i + 27{q_i}^2)$$

One of the solutions of the cubic can be represented with trigonometric functions by using Viete's method.

$$u_i = 2s_i \cos\left(\frac{1}{3}\arccos\frac{t_i}{2s_i}\right) - \frac{2p_i}{3}$$

with

$$s_i = \frac{1}{3}\sqrt{{p_i}^2 + 12r_i} = \frac{1}{3}\sqrt{{b_i}^2 - 3a_i c_i + 12d_i}$$

$$t_i = \frac{2{p_i}^3 - 72p_i r_i + 27{q_i}^2}{3{p_i}^2 + 36r_i} = \frac{2{b_i}^3 - 9a_i b_i c_i + 27{c_i}^2 + 27{a_i}^2 d_i - 72b_i d_i}{3{b_i}^2 - 9a_i c_i + 36d_i}$$

The quartic equation can be decomposed into two quadratic equations with u_0.

$$\begin{cases} x^2 + \sqrt{u_i}x + \frac{p_i + u_i}{2} - \frac{q_i}{2\sqrt{u_i}} = 0 \\ x^2 - \sqrt{u_i}x + \frac{p_i + u_i}{2} + \frac{q_i}{2\sqrt{u_i}} = 0 \end{cases}$$

Therefore, the solutions of the equation are

$$x = \frac{1}{2}\left(\sqrt{u_i} + \sqrt{-2p_i - \frac{2q_i}{\sqrt{u_i}} - u_i}\right) - \frac{a_i}{4} = \frac{1}{4}\left(-a_i + 2\sqrt{u_i} + \sqrt{3{a_i}^2 - 8b_i - \frac{{a_i}^3 - 4a_i b_i + 8c_i}{\sqrt{u_i}} - 4u_i}\right)$$

$$x = \frac{1}{2}\left(\sqrt{u_i} - \sqrt{-2p_i - \frac{2q_i}{\sqrt{u_i}} - u_i}\right) - \frac{a_i}{4} = \frac{1}{4}\left(-a_i + 2\sqrt{u_i} - \sqrt{3{a_i}^2 - 8b_i - \frac{{a_i}^3 - 4a_i b_i + 8c_i}{\sqrt{u_i}} - 4u_i}\right)$$

$$x = \frac{1}{2}\left(-\sqrt{u_i} + \sqrt{-2p_i + \frac{2q_i}{\sqrt{u_i}} - u_i}\right) - \frac{a_i}{4}$$

$$= \frac{1}{4}\left(-a_i - 2\sqrt{u_i} + \sqrt{3{a_i}^2 - 8b_i + \frac{{a_i}^3 - 4a_i b_i + 8c_i}{\sqrt{u_i}} - 4u_i}\right)$$

$$x = \frac{1}{2}\left(-\sqrt{u_i} - \sqrt{-2p_i + \frac{2q_i}{\sqrt{u_i}} - u_i}\right) - \frac{a_i}{4}$$

$$= \frac{1}{4}\left(-a_i - 2\sqrt{u_i} - \sqrt{3a_i^2 - 8b_i + \frac{a_i^3 - 4a_ib_i + 8c_i}{\sqrt{u_i}} - 4u_i}\right)$$

The set of the coefficients of the quartic equations $\{a_i, b_i, c_i, d_i\}$ is given as follows:

$$a_1 = A_x + A_y + A_z + (-A_x - A_y + 2A_z)\cos 2\theta + \sqrt{3}(A_x - A_y)\sin 2\theta$$

$$\begin{aligned} b_1 = {} & -\frac{51}{8}\left(A_z{}^2 + A_z{}^2 + A_z{}^2\right) + \frac{1}{4}\left(17A_x{}^2 - 10A_xA_y + 17A_y{}^2 + 5A_xA_z + 5A_yA_z - 34A_z{}^2\right)\cos 2\theta \\ & + \frac{1}{8}\left(17A_x{}^2 + 20A_xA_y + 17A_y{}^2 - 10A_xA_z - 10A_yA_z - 34A_z{}^2\right)\cos 4\theta \\ & + \frac{\sqrt{3}}{4}\left(-17A_x{}^2 + 17A_y{}^2 + 5A_xA_z - 5A_yA_z\right)\sin 2\theta \\ & + \frac{\sqrt{3}}{8}\left(17A_x{}^2 - 17A_y{}^2 + 10A_xA_z - 10A_yA_z\right)\sin 4\theta \end{aligned}$$

$$\begin{aligned} c_1 = {} & \frac{1}{16}\left(-133A_x{}^3 - 29A_x{}^2A_y - 29A_xA_y{}^2 - 133A_y{}^3 - 29A_x{}^2A_z + 28A_xA_yA_z - 29A_y{}^2A_z - 29A_xA_z{}^2\right. \\ & \left. - 29A_yA_z{}^2 - 133A_z{}^3\right) \\ & + \frac{3}{16}\left(38A_x{}^3 + 29A_x{}^2A_y + 29A_xA_y{}^2 + 38A_y{}^3 - 29A_x{}^2A_z - 29A_y{}^2A_z\right. \\ & \left. - 76A_z{}^3\right)\cos 2\theta \\ & + \frac{3}{16}\left(19A_x{}^3 - 29A_x{}^2A_y - 29A_xA_y{}^2 + 19A_y{}^3 + 29A_x{}^2A_z + 29A_y{}^2A_z\right. \\ & \left. - 38A_z{}^3\right)\cos 4\theta \\ & + \frac{1}{16}\left(-38A_x{}^3 + 29A_x{}^2A_y + 29A_xA_y{}^2 - 38A_y{}^3 + 29A_x{}^2A_z - 28A_xA_yA_z + 29A_y{}^2A_z\right. \\ & \left. + 29A_xA_z{}^2 + 29A_yA_z{}^2 - 38A_z{}^3\right)\cos 6\theta \\ & + \frac{\sqrt{3}}{16}\left(-114A_x{}^3 + 29A_x{}^2A_y - 29A_xA_y{}^2 + 114A_y{}^3 - 29A_x{}^2A_z + 29A_y{}^2A_z - 58A_xA_z{}^2\right. \\ & \left. + 58A_yA_z{}^2\right)\sin 2\theta \\ & + \frac{\sqrt{3}}{16}\left(57A_x{}^3 + 29A_x{}^2A_y - 29A_xA_y{}^2 - 57A_y{}^3 - 29A_x{}^2A_z + 29A_y{}^2A_z - 58A_xA_z{}^2\right. \\ & \left. + 58A_yA_z{}^2\right)\sin 4\theta \\ & + \frac{29\sqrt{3}}{16}\left(-A_x{}^2A_y + A_xA_y{}^2 + A_x{}^2A_z - A_y{}^2A_z - A_xA_z{}^2 + A_yA_z{}^2\right)\sin 6\theta \end{aligned}$$

$$d_1 = \frac{21}{256}\left(95{A_x}^4 - 20{A_x}^3A_y + 120{A_x}^2{A_y}^2 - 20A_x{A_y}^3 + 95{A_y}^4 - 20{A_x}^3A_z + 88{A_x}^2A_yA_z + 88A_x{A_y}^2A_z - 20{A_y}^3A_z + 120{A_x}^2{A_z}^2 + 88A_xA_y{A_z}^2 + 120{A_y}^2{A_z}^2 - 20A_x{A_z}^3 - 20A_y{A_z}^3 + 95{A_z}^4\right)$$
$$+ \frac{21}{64}\left(-20{A_x}^4 + 35{A_x}^3A_y - 30{A_x}^2{A_y}^2 + 35A_x{A_y}^3 - 20{A_y}^4 - 25{A_x}^3A_z - 22{A_x}^2A_yA_z - 22A_x{A_y}^2A_z - 25{A_y}^3A_z + 15{A_x}^2{A_z}^2 + 44A_xA_y{A_z}^2 + 15{A_y}^2{A_z}^2 - 10A_x{A_z}^3 - 10A_y{A_z}^3 + 40{A_z}^4\right)\cos 2\theta$$
$$+ \frac{21}{128}\left(-25{A_x}^4 - 80{A_x}^3A_y + 30{A_x}^2{A_y}^2 - 80A_x{A_y}^3 - 25{A_y}^4 + 70{A_x}^3A_z + 22{A_x}^2A_yA_z + 22A_x{A_y}^2A_z + 70{A_y}^3A_z - 15{A_x}^2{A_z}^2 - 44A_xA_y{A_z}^2 - 15{A_y}^2{A_z}^2 + 10A_x{A_z}^3 + 10A_y{A_z}^3 + 50{A_z}^4\right)\cos 4\theta$$
$$+ \frac{21}{64}\left(10{A_x}^4 + 5{A_x}^3A_y - 30{A_x}^2{A_y}^2 + 5A_x{A_y}^3 + 10{A_y}^4 + 5{A_x}^3A_z - 22{A_x}^2A_yA_z - 22A_x{A_y}^2A_z + 5{A_y}^3A_z - 30{A_x}^2{A_z}^2 - 22A_xA_y{A_z}^2 - 30{A_y}^2{A_z}^2 + 5A_x{A_z}^3 + 5A_y{A_z}^3 + 10{A_z}^4\right)\cos 6\theta$$
$$+ \frac{21}{256}\left(-5{A_x}^4 + 20{A_x}^3A_y + 60{A_x}^2{A_y}^2 + 20A_x{A_y}^3 - 5{A_y}^4 - 40{A_x}^3A_z + 44{A_x}^2A_yA_z + 44A_x{A_y}^2A_z - 40{A_y}^3A_z - 30{A_x}^2{A_z}^2 - 88A_xA_y{A_z}^2 - 30{A_y}^2{A_z}^2 + 20A_x{A_z}^3 + 20A_y{A_z}^3 + 10{A_z}^4\right)\cos 8\theta$$
$$+ \frac{21\sqrt{3}}{64}\left(20{A_x}^4 + 5{A_x}^3A_y - 5A_x{A_y}^3 - 20{A_y}^4 - 15{A_x}^3A_z + 22{A_x}^2A_yA_z - 22A_x{A_y}^2A_z + 15{A_y}^3A_z + 15{A_x}^2{A_z}^2 - 15{A_y}^2{A_z}^2 - 20A_x{A_z}^3 + 20A_y{A_z}^3\right)\sin 2\theta$$
$$+ \frac{21\sqrt{3}}{128}\left(-25{A_x}^4 + 20{A_x}^3A_y - 20A_x{A_y}^3 + 25{A_y}^4 - 30{A_x}^3A_z + 22{A_x}^2A_yA_z - 22A_x{A_y}^2A_z + 30{A_y}^3A_z + 15{A_x}^2{A_z}^2 - 15{A_y}^2{A_z}^2 - 50A_x{A_z}^3 + 50A_y{A_z}^3\right)\sin 4\theta$$
$$+ \frac{315\sqrt{3}}{64}\left(-{A_x}^3A_y + A_x{A_y}^3 + {A_x}^3A_z - {A_y}^3A_z - A_x{A_z}^3 + A_y{A_z}^3\right)\sin 6\theta$$
$$+ \frac{21\sqrt{3}}{256}\left(5{A_x}^4 + 20{A_x}^3A_y - 20A_x{A_y}^3 - 5{A_y}^4 - 44{A_x}^2A_yA_z + 44A_x{A_y}^2A_z - 30{A_x}^2{A_z}^2 + 30{A_y}^2{A_z}^2 - 20A_x{A_z}^3 + 20A_y{A_z}^3\right)\sin 8\theta$$

$$a_2 = -A_x - A_y + A_z + \left(A_x + A_y + 2A_z\right)\cos 2\theta - \sqrt{3}\left(A_x - A_y\right)\sin 2\theta$$

$$b_2 = -\frac{51}{8}\left(A_x{}^2 + A_y{}^2 + A_z{}^2\right) + \frac{1}{4}\left(17A_x{}^2 - 10A_xA_y + 17A_y{}^2 - 5A_xA_z - 5A_yA_z - 34A_z{}^2\right)\cos 2\theta$$

$$+ \frac{1}{8}\left(17A_x{}^2 + 20A_xA_y + 17A_y{}^2 + 10A_xA_z + 10A_yA_z - 34A_z{}^2\right)\cos 4\theta$$

$$+ \frac{\sqrt{3}}{4}\left(-17A_x{}^2 + 17A_y{}^2 - 5A_xA_z + 5A_yA_z\right)\sin 2\theta$$

$$+ \frac{\sqrt{3}}{8}\left(17A_x{}^2 - 17A_y{}^2 - 10A_xA_z + 10A_yA_z\right)\sin 4\theta$$

$$c_2 = \frac{1}{16}\left(133A_x{}^3 + 29A_x{}^2A_y + 29A_xA_y{}^2 + 133A_y{}^3 - 29A_x{}^2A_z + 28A_xA_yA_z - 29A_y{}^2A_z + 29A_xA_z{}^2\right.$$

$$\left. + 29A_yA_z{}^2 - 133A_z{}^3\right)$$

$$+ \frac{3}{16}\left(-38A_x{}^3 - 29A_x{}^2A_y - 29A_xA_y{}^2 - 38A_y{}^3 - 29A_x{}^2A_z - 29A_y{}^2A_z\right.$$

$$\left. - 76A_z{}^3\right)\cos 2\theta$$

$$+ \frac{3}{16}\left(-19A_x{}^3 + 29A_x{}^2A_y + 29A_xA_y{}^2 - 19A_y{}^3 + 29A_x{}^2A_z + 29A_y{}^2A_z\right.$$

$$\left. - 38A_z{}^3\right)\cos 4\theta$$

$$+ \frac{1}{16}\left(38A_x{}^3 - 29A_x{}^2A_y - 29A_xA_y{}^2 + 38A_y{}^3 + 29A_x{}^2A_z - 28A_xA_yA_z + 29A_y{}^2A_z\right.$$

$$\left. - 29A_xA_z{}^2 - 29A_yA_z{}^2 - 38A_z{}^3\right)\cos 6\theta$$

$$+ \frac{\sqrt{3}}{16}\left(114A_x{}^3 - 29A_x{}^2A_y \sin 2\theta + 29A_xA_y{}^2 - 114A_y{}^3 - 29A_x{}^2A_z + 29A_y{}^2A_z\right.$$

$$\left. + 58A_xA_z{}^2 - 58A_yA_z{}^2\right)\sin 2\theta$$

$$+ \frac{\sqrt{3}}{16}\left(-57A_x{}^3 - 29A_x{}^2A_y + 29A_xA_y{}^2 + 57A_y{}^3 - 29A_x{}^2A_z + 29A_y{}^2A_z + 58A_xA_z{}^2\right.$$

$$\left. - 58A_yA_z{}^2\right)\sin 4\theta$$

$$+ \frac{29\sqrt{3}}{16}\left(A_x{}^2A_y - A_xA_y{}^2 + A_x{}^2A_z - A_y{}^2A_z + A_xA_z{}^2 - A_yA_z{}^2\right)\sin 6\theta$$

$$
\begin{aligned}
d_2 = \frac{21}{256}\big(&95A_x{}^4 - 20A_x{}^3A_y + 120A_x{}^2A_y{}^2 - 20A_xA_y{}^3 + 95A_y{}^4 + 20A_x{}^3A_z - 88A_x{}^2A_yA_z \\
&- 88A_xA_y{}^2A_z + 20A_y{}^3A_z + 120A_x{}^2A_z{}^2 + 88A_xA_yA_z{}^2 + 120A_y{}^2A_z{}^2 + 20A_xA_z{}^3 \\
&+ 20A_yA_z{}^3 + 95A_z{}^4\big) \\
&+ \frac{21}{64}\big(-20A_x{}^4 + 35A_x{}^3A_y - 30A_x{}^2A_y{}^2 + 35A_xA_y{}^3 - 20A_y{}^4 + 25A_x{}^3A_z \\
&+ 22A_x{}^2A_yA_z + 22A_xA_y{}^2A_z + 25A_y{}^3A_z + 15A_x{}^2A_z{}^2 + 44A_xA_yA_z{}^2 + 15A_y{}^2A_z{}^2 \\
&+ 10A_xA_z{}^3 + 10A_yA_z{}^3 + 40A_z{}^4\big)\cos 2\theta \\
&+ \frac{21}{128}\big(-25A_x{}^4 - 80A_x{}^3A_y + 30A_x{}^2A_y{}^2 - 80A_xA_y{}^3 - 25A_y{}^4 - 70A_x{}^3A_z \\
&- 22A_x{}^2A_yA_z - 22A_xA_y{}^2A_z - 70A_y{}^3A_z - 15A_x{}^2A_z{}^2 - 44A_xA_yA_z{}^2 - 15A_y{}^2A_z{}^2 \\
&- 10A_xA_z{}^3 - 10A_yA_z{}^3 + 50A_z{}^4\big)\cos 4\theta \\
&+ \frac{21}{64}\big(10A_x{}^4 + 5A_x{}^3A_y - 30A_x{}^2A_y{}^2 + 5A_xA_y{}^3 + 10A_y{}^4 - 5A_x{}^3A_z + 22A_x{}^2A_yA_z \\
&+ 22A_xA_y{}^2A_z - 5A_y{}^3A_z - 30A_x{}^2A_z{}^2 - 22A_xA_yA_z{}^2 - 30A_y{}^2A_z{}^2 - 5A_xA_z{}^3 - 5A_yA_z{}^3 \\
&+ 10A_z{}^4\big)\cos 6\theta \\
&+ \frac{21}{256}\big(-5A_x{}^4 + 20A_x{}^3A_y + 60A_x{}^2A_y{}^2 + 20A_xA_y{}^3 - 5A_y{}^4 + 40A_x{}^3A_z - 44A_x{}^2A_yA_z \\
&- 44A_xA_y{}^2A_z + 40A_y{}^3A_z - 30A_x{}^2A_z{}^2 - 88A_xA_yA_z{}^2 - 30A_y{}^2A_z{}^2 - 20A_xA_z{}^3 \\
&- 20A_yA_z{}^3 + 10A_z{}^4\big)\cos 8\theta \\
&+ \frac{21\sqrt{3}}{64}\big(20A_x{}^4 + 5A_x{}^3A_y - 5A_xA_y{}^3 - 20A_y{}^4 + 15A_x{}^3A_z - 22A_x{}^2A_yA_z \\
&+ 22A_xA_y{}^2A_z - 15A_y{}^3A_z + 15A_x{}^2A_z{}^2 - 15A_y{}^2A_z{}^2 + 20A_xA_z{}^3 - 20A_yA_z{}^3\big)\sin 2\theta \\
&+ \frac{21\sqrt{3}}{128}\big(-25A_x{}^4 + 20A_x{}^3A_y - 20A_xA_y{}^3 + 25A_y{}^4 + 30A_x{}^3A_z - 22A_x{}^2A_yA_z \\
&+ 22A_xA_y{}^2A_z - 30A_y{}^3A_z + 15A_x{}^2A_z{}^2 - 15A_y{}^2A_z{}^2 + 50A_xA_z{}^3 - 50A_yA_z{}^3\big)\sin 4\theta \\
&+ \frac{315\sqrt{3}}{64}\big(-A_x{}^3A_y + A_xA_y{}^3 - A_x{}^3A_z + A_y{}^3A_z + A_xA_z{}^3 + A_yA_z{}^3\big)\sin 6\theta \\
&+ \frac{21\sqrt{3}}{256}\big(5A_x{}^4 + 20A_x{}^3A_y - 20A_xA_y{}^3 - 5A_y{}^4 + 44A_x{}^2A_yA_z - 44A_xA_y{}^2A_z \\
&- 30A_x{}^2A_z{}^2 + 30A_y{}^2A_z{}^2 + 20A_xA_z{}^3 - 20A_yA_z{}^3\big)\sin 8\theta
\end{aligned}
$$

$$
a_3 = A_x - A_y - A_z - \left(A_x - A_y + 2A_z\right)\cos 2\theta + \sqrt{3}\left(A_x + A_y\right)\sin 2\theta
$$

$$b_3 = -\frac{51}{8}\left(A_x{}^2 + A_y{}^2 + A_z{}^2\right) + \frac{1}{4}\left(17A_x{}^2 + 10A_xA_y + 17A_y{}^2 - 5A_xA_z + 5A_yA_z - 34A_z{}^2\right)\cos 2\theta$$

$$+ \frac{1}{8}\left(17A_x{}^2 - 20A_xA_y + 17A_y{}^2 + 10A_xA_z - 10A_yA_z - 34A_z{}^2\right)\cos 4\theta$$

$$+ \frac{\sqrt{3}}{4}\left(-17A_x{}^2 + 17A_y{}^2 - 5A_xA_z - 5A_yA_z\right)\sin 2\theta$$

$$+ \frac{\sqrt{3}}{8}\left(17A_x{}^2 - 17A_y{}^2 - 10A_xA_z - 10A_yA_z\right)\sin 4\theta$$

$$c_3 = \frac{1}{16}\left(-133A_x{}^3 + 29A_x{}^2A_y - 29A_xA_y{}^2 + 133A_y{}^3 + 29A_x{}^2A_z + 28A_xA_yA_z + 29A_y{}^2A_z - 29A_xA_z{}^2\right.$$

$$\left. + 29A_yA_z{}^2 + 133A_z{}^3\right)$$

$$+ \frac{3}{16}\left(38A_x{}^3 - 29A_x{}^2A_y + 29A_xA_y{}^2 - 38A_y{}^3 + 29A_x{}^2A_z + 29A_y{}^2A_z\right.$$

$$\left. + 76A_z{}^3\right)\cos 2\theta$$

$$+ \frac{3}{16}\left(19A_x{}^3 + 29A_x{}^2A_y - 29A_xA_y{}^2 - 19A_y{}^3 - 29A_x{}^2A_z - 29A_y{}^2A_z\right.$$

$$\left. + 38A_z{}^3\right)\cos 4\theta$$

$$+ \frac{1}{16}\left(-38A_x{}^3 - 29A_x{}^2A_y + 29A_xA_y{}^2 + 38A_y{}^3 - 29A_x{}^2A_z - 28A_xA_yA_z - 29A_y{}^2A_z\right.$$

$$\left. + 29A_xA_z{}^2 - 29A_yA_z{}^2 + 38A_z{}^3\right)\cos 6\theta$$

$$+ \frac{\sqrt{3}}{16}\left(-114A_x{}^3 - 29A_x{}^2A_y - 29A_xA_y{}^2 - 114A_y{}^3 + 29A_x{}^2A_z - 29A_y{}^2A_z - 58A_xA_z{}^2\right.$$

$$\left. - 58A_yA_z{}^2\right)\sin 2\theta$$

$$+ \frac{\sqrt{3}}{16}\left(57A_x{}^3 - 29A_x{}^2A_y - 29A_xA_y{}^2 + 57A_y{}^3 + 29A_x{}^2A_z - 29A_y{}^2A_z - 58A_xA_z{}^2\right.$$

$$\left. - 58A_yA_z{}^2\right)\sin 4\theta$$

$$+ \frac{29\sqrt{3}}{16}\left(A_x{}^2A_y + A_xA_y{}^2 - A_x{}^2A_z + A_y{}^2A_z - A_xA_z{}^2 - A_yA_z{}^2\right)\sin 6\theta$$

$$d_3 = \frac{21}{256}\left(95A_x{}^4 + 20A_x{}^3A_y + 120A_x{}^2A_y{}^2 + 20A_xA_y{}^3 + 95A_y{}^4 + 20A_x{}^3A_z + 88A_x{}^2A_yA_z - 88A_xA_y{}^2A_z - 20A_y{}^3A_z + 120A_x{}^2A_z{}^2 - 88A_xA_yA_z{}^2 + 120A_y{}^2A_z{}^2 + 20A_xA_z{}^3 - 20A_yA_z{}^3 + 95A_z{}^4\right)$$
$$+ \frac{21}{64}\left(-20A_x{}^4 - 35A_x{}^3A_y - 30A_x{}^2A_y{}^2 - 35A_xA_y{}^3 - 20A_y{}^4 + 25A_x{}^3A_z - 22A_x{}^2A_yA_z + 22A_xA_y{}^2A_z - 25A_y{}^3A_z + 15A_x{}^2A_z{}^2 - 44A_xA_yA_z{}^2 + 15A_y{}^2A_z{}^2 + 10A_xA_z{}^3 - 10A_yA_z{}^3 + 40A_z{}^4\right)\cos 2\theta$$
$$+ \frac{21}{128}\left(-25A_x{}^4 + 80A_x{}^3A_y + 30A_x{}^2A_y{}^2 + 80A_xA_y{}^3 - 25A_y{}^4 - 70A_x{}^3A_z + 22A_x{}^2A_yA_z - 22A_xA_y{}^2A_z + 70A_y{}^3A_z - 15A_x{}^2A_z{}^2 + 44A_xA_yA_z{}^2 - 15A_y{}^2A_z{}^2 - 10A_xA_z{}^3 + 10A_yA_z{}^3 + 50A_z{}^4\right)\cos 4\theta$$
$$+ \frac{21}{64}\left(10A_x{}^4 - 5A_x{}^3A_y - 30A_x{}^2A_y{}^2 - 5A_xA_y{}^3 + 10A_y{}^4 - 5A_x{}^3A_z - 22A_x{}^2A_yA_z + 22A_xA_y{}^2A_z + 5A_y{}^3A_z - 30A_x{}^2A_z{}^2 + 22A_xA_yA_z{}^2 - 30A_y{}^2A_z{}^2 - 5A_xA_z{}^3 + 5A_yA_z{}^3 + 10A_z{}^4\right)\cos 6\theta$$
$$+ \frac{21}{256}\left(-5A_x{}^4 - 20A_x{}^3A_y + 60A_x{}^2A_y{}^2 - 20A_xA_y{}^3 - 5A_y{}^4 + 40A_x{}^3A_z + 44A_x{}^2A_yA_z - 44A_xA_y{}^2A_z - 40A_y{}^3A_z - 30A_x{}^2A_z{}^2 + 88A_xA_yA_z{}^2 - 30A_y{}^2A_z{}^2 - 20A_xA_z{}^3 + 20A_yA_z{}^3 + 10A_z{}^4\right)\cos 8\theta$$
$$+ \frac{21\sqrt{3}}{64}\left(20A_x{}^4 - 5A_x{}^3A_y + 5A_xA_y{}^3 - 20A_y{}^4 + 15A_x{}^3A_z + 22A_x{}^2A_yA_z + 22A_xA_y{}^2A_z + 15A_y{}^3A_z + 15A_x{}^2A_z{}^2 - 15A_y{}^2A_z{}^2 + 20A_xA_z{}^3 + 20A_yA_z{}^3\right)\sin 2\theta$$
$$+ \frac{21\sqrt{3}}{128}\left(-25A_x{}^4 - 20A_x{}^3A_y + 20A_xA_y{}^3 + 25A_y{}^4 + 30A_x{}^3A_z + 22A_x{}^2A_yA_z + 22A_xA_y{}^2A_z + 30A_y{}^3A_z + 15A_x{}^2A_z{}^2 - 15A_y{}^2A_z{}^2 + 50A_xA_z{}^3 + 50A_yA_z{}^3\right)\sin 4\theta$$
$$+ \frac{315\sqrt{3}}{64}\left(A_x{}^3A_y - A_xA_y{}^3 - A_x{}^3A_z - A_y{}^3A_z + A_xA_z{}^3 + A_yA_z{}^3\right)\sin 6\theta$$
$$+ \frac{21\sqrt{3}}{256}\left(5A_x{}^4 - 20A_x{}^3A_y + 20A_xA_y{}^3 - 5A_y{}^4 - 44A_x{}^2A_yA_z - 44A_xA_y{}^2A_z - 30A_x{}^2A_z{}^2 + 30A_y{}^2A_z{}^2 + 20A_xA_z{}^3 + 20A_yA_z{}^3\right)\sin 8\theta$$

$$a_4 = -A_x + A_y - A_z + \left(A_x - A_y - 2A_z\right)\cos 2\theta - \sqrt{3}\left(A_x + A_y\right)\sin 2\theta$$

$$b_4 = -\frac{51}{8}\left({A_x}^2 + {A_y}^2 + {A_z}^2\right) + \frac{1}{4}\left(17{A_x}^2 - 10A_xA_y + 17{A_y}^2 + 5A_xA_z + 5A_yA_z - 34{A_z}^2\right)\cos 2\theta$$

$$+ \frac{1}{8}\left(17{A_x}^2 + 20A_xA_y + 17{A_y}^2 - 10A_xA_z - 10A_yA_z - 34{A_z}^2\right)\cos 4\theta$$

$$+ \frac{\sqrt{3}}{4}\left(-17{A_x}^2 + 17{A_y}^2 + 5A_xA_z - 5A_yA_z\right)\sin 2\theta$$

$$+ \frac{\sqrt{3}}{8}\left(17{A_x}^2 - 17{A_y}^2 + 10A_xA_z + 10A_yA_z\right)\sin 4\theta$$

$$c_4 = \frac{1}{16}\left(133{A_x}^3 - 29{A_x}^2A_y + 29A_x{A_y}^2 - 133{A_y}^3 + 29{A_x}^2A_z + 28A_xA_yA_z + 29{A_y}^2A_z + 29A_x{A_z}^2\right.$$

$$\left. - 29A_y{A_z}^2 + 133{A_z}^3\right)$$

$$+ \frac{3}{16}\left(-38{A_x}^3 + 29{A_x}^2A_y - 29A_x{A_y}^2 + 38{A_y}^3 + 29{A_x}^2A_z + 29{A_y}^2A_z\right.$$

$$\left. + 76{A_z}^3\right)\cos 2\theta$$

$$+ \frac{3}{16}\left(-19{A_x}^3 + 29{A_x}^2A_y - 29A_x{A_y}^2 + 19{A_y}^3 - 29{A_x}^2A_z - 29{A_y}^2A_z\right.$$

$$\left. + 38{A_z}^3\right)\cos 4\theta$$

$$+ \frac{1}{16}\left(38{A_x}^3 + 29{A_x}^2A_y - 29A_x{A_y}^2 - 38{A_y}^3 - 29{A_x}^2A_z - 28A_xA_yA_z - 29{A_y}^2A_z\right.$$

$$\left. - 29A_x{A_z}^2 + 29A_y{A_z}^2 + 38{A_z}^3\right)\cos 6\theta$$

$$+ \frac{\sqrt{3}}{16}\left(114{A_x}^3 + 29{A_x}^2A_y + 29A_x{A_y}^2 + 114{A_y}^3 + 29{A_x}^2A_z - 29{A_y}^2A_z + 58A_x{A_z}^2\right.$$

$$\left. + 58A_y{A_z}^2\right)\sin 2\theta$$

$$+ \frac{\sqrt{3}}{16}\left(-57{A_x}^3 + 29{A_x}^2A_y + 29A_x{A_y}^2 - 57{A_y}^3 + 29{A_x}^2A_z - 29{A_y}^2A_z + 58A_x{A_z}^2\right.$$

$$\left. + 58A_y{A_z}^2\right)\sin 4\theta$$

$$+ \frac{29\sqrt{3}}{16}\left(-{A_x}^2A_y - A_x{A_y}^2 - {A_x}^2A_z + {A_y}^2A_z + A_x{A_z}^2 + A_y{A_z}^2\right)\sin 6\theta$$

$$d_4 = \frac{21}{256}\left(95{A_x}^4 + 20{A_x}^3A_y + 120{A_x}^2{A_y}^2 + 20A_x{A_y}^3 + 95{A_y}^4 - 20{A_x}^3A_z - 88{A_x}^2A_yA_z + 88A_x{A_y}^2A_z + 20{A_y}^3A_z + 120{A_x}^2{A_z}^2 - 88A_xA_y{A_z}^2 + 120{A_y}^2{A_z}^2 - 20A_x{A_z}^3 + 20A_y{A_z}^3 + 95{A_z}^4\right)$$
$$+ \frac{21}{64}\left(-20{A_x}^4 - 35{A_x}^3A_y - 30{A_x}^2{A_y}^2 - 35A_x{A_y}^3 - 20{A_y}^4 - 25{A_x}^3A_z + 22{A_x}^2A_yA_z - 22A_x{A_y}^2A_z + 25{A_y}^3A_z + 15{A_x}^2{A_z}^2 - 44A_xA_y{A_z}^2 + 15{A_y}^2{A_z}^2 - 10A_x{A_z}^3 + 10A_y{A_z}^3 + 40{A_z}^4\right)\cos 2\theta$$
$$+ \frac{21}{128}\left(-25{A_x}^4 + 80{A_x}^3A_y + 30{A_x}^2{A_y}^2 + 80A_x{A_y}^3 - 25{A_y}^4 + 70{A_x}^3A_z - 22{A_x}^2A_yA_z + 22A_x{A_y}^2A_z - 70{A_y}^3A_z - 15{A_x}^2{A_z}^2 + 44A_xA_y{A_z}^2 - 15{A_y}^2{A_z}^2 + 10A_x{A_z}^3 - 10A_y{A_z}^3 + 50{A_z}^4\right)\cos 4\theta$$
$$+ \frac{21}{64}\left(10{A_x}^4 - 5{A_x}^3A_y - 30{A_x}^2{A_y}^2 - 5A_x{A_y}^3 + 10{A_y}^4 + 5{A_x}^3A_z + 22{A_x}^2A_yA_z - 22A_x{A_y}^2A_z - 5{A_y}^3A_z - 30{A_x}^2{A_z}^2 + 22A_xA_y{A_z}^2 - 30{A_y}^2{A_z}^2 + 5A_x{A_z}^3 - 5A_y{A_z}^3 + 10{A_z}^4\right)\cos 6\theta$$
$$+ \frac{21}{256}\left(-5{A_x}^4 - 20{A_x}^3A_y + 60{A_x}^2{A_y}^2 - 20A_x{A_y}^3 - 5{A_y}^4 - 40{A_x}^3A_z - 44{A_x}^2A_yA_z + 44A_x{A_y}^2A_z + 40{A_y}^3A_z - 30{A_x}^2{A_z}^2 + 88A_xA_y{A_z}^2 - 30{A_y}^2{A_z}^2 + 20A_x{A_z}^3 - 20A_y{A_z}^3 + 10{A_z}^4\right)\cos 8\theta$$
$$+ \frac{21\sqrt{3}}{64}\left(20{A_x}^4 - 5{A_x}^3A_y + 5A_x{A_y}^3 - 20{A_y}^4 - 15{A_x}^3A_z - 22{A_x}^2A_yA_z - 22A_x{A_y}^2A_z - 15{A_y}^3A_z + 15{A_x}^2{A_z}^2 - 15{A_y}^2{A_z}^2 - 20A_x{A_z}^3 - 20A_y{A_z}^3\right)\sin 2\theta$$
$$+ \frac{21\sqrt{3}}{128}\left(-25{A_x}^4 - 20{A_x}^3A_y + 20A_x{A_y}^3 + 25{A_y}^4 - 30{A_x}^3A_z - 22{A_x}^2A_yA_z - 22A_x{A_y}^2A_z - 30{A_y}^3A_z + 15{A_x}^2{A_z}^2 - 15{A_y}^2{A_z}^2 - 50A_x{A_z}^3 - 50A_y{A_z}^3\right)\sin 4\theta$$
$$+ \frac{315\sqrt{3}}{64}\left({A_x}^3A_y - A_x{A_y}^3 + {A_x}^3A_z + {A_y}^3A_z - A_x{A_z}^3 - A_y{A_z}^3\right)\sin 6\theta$$
$$+ \frac{21\sqrt{3}}{256}\left(5{A_x}^4 - 20{A_x}^3A_y + 20A_x{A_y}^3 - 5{A_y}^4 + 44{A_x}^2A_yA_z + 44A_x{A_y}^2A_z - 30{A_x}^2{A_z}^2 + 30{A_y}^2{A_z}^2 - 20A_x{A_z}^3 - 20A_y{A_z}^3\right)\sin 8\theta$$

The set of the coefficients for i = 2, 3 and 4 can be obtained from $\{a_1, b_1, c_1, d_1\}$ with replacing A_x and A_y to $-A_x$ and $-A_y$, A_x and A_z to $-A_x$ and $-A_z$, and A_y and A_z to $-A_y$ and $-A_z$, respectively.

For the negative counterpart:

$$a_5 = -A_x + A_y - A_z - (A_x - A_y - 2A_z)\cos 2\theta + \sqrt{3}(A_x + A_y)\sin 2\theta$$

$$b_5 = -\frac{51}{8}\left({A_x}^2 + {A_y}^2 + {A_z}^2\right) + \frac{1}{4}\left(-17{A_x}^2 - 10A_xA_y - 17{A_y}^2 - 5A_xA_z + 5A_yA_z + 34{A_z}^2\right)\cos 2\theta$$

$$+ \frac{1}{8}\left(17{A_x}^2 - 20A_xA_y + 17{A_y}^2 - 10A_xA_z + 10A_yA_z - 34{A_z}^2\right)\cos 4\theta$$

$$+ \frac{\sqrt{3}}{4}\left(17{A_x}^2 - 17{A_y}^2 - 5A_xA_z - 5A_yA_z\right)\sin 2\theta$$

$$+ \frac{\sqrt{3}}{8}\left(17{A_x}^2 - 17{A_y}^2 + 10A_xA_z + 10A_yA_z\right)\sin 4\theta$$

$$c_5 = \frac{1}{16}\left(133{A_x}^3 - 29{A_x}^2A_y + 29A_x{A_y}^2 - 133{A_y}^3 + 29{A_x}^2A_z + 28A_xA_yA_z + 29{A_y}^2A_z + 29A_x{A_z}^2\right.$$

$$\left. - 29A_y{A_z}^2 + 133{A_z}^3\right)$$

$$+ \frac{3}{16}\left(38{A_x}^3 - 29{A_x}^2A_y + 29A_x{A_y}^2 - 38{A_y}^3 - 29{A_x}^2A_z - 29{A_y}^2A_z\right.$$

$$\left. - 76{A_z}^3\right)\cos 2\theta$$

$$+ \frac{3}{16}\left(-19{A_x}^3 - 29{A_x}^2A_y + 29A_x{A_y}^2 + 19{A_y}^3 - 29{A_x}^2A_z - 29{A_y}^2A_z\right.$$

$$\left. + 38{A_z}^3\right)\cos 4\theta$$

$$+ \frac{1}{16}\left(-38{A_x}^3 - 29{A_x}^2A_y + 29A_x{A_y}^2 + 38{A_y}^3 + 29{A_x}^2A_z + 28A_xA_yA_z + 29{A_y}^2A_z\right.$$

$$\left. + 29A_x{A_z}^2 - 29A_y{A_z}^2 - 38{A_z}^3\right)\cos 6\theta$$

$$+ \frac{\sqrt{3}}{16}\left(-114{A_x}^3\sin 2\theta - 29{A_x}^2A_y - 29A_x{A_y}^2 - 114{A_y}^3 - 29{A_x}^2A_z + 29{A_y}^2A_z\right.$$

$$\left. - 58A_x{A_z}^2 - 58A_y{A_z}^2\right)\sin 2\theta$$

$$+ \frac{\sqrt{3}}{16}\left(-57{A_x}^3 + 29{A_x}^2A_y + 29A_x{A_y}^2 - 57{A_y}^3 + 29{A_x}^2A_z - 29{A_y}^2A_z + 58A_x{A_z}^2\right.$$

$$\left. + 58A_y{A_z}^2\right)\sin 4\theta$$

$$+ \frac{29\sqrt{3}}{16}\left({A_x}^2A_y + A_x{A_y}^2 + {A_x}^2A_z - {A_y}^2A_z - A_x{A_z}^2 - A_y{A_z}^2\right)\sin 6\theta$$

$$d_5 = \frac{21}{256}\left(95A_x{}^4 + 20A_x{}^3A_y + 120A_x{}^2A_y{}^2 + 20A_xA_y{}^3 + 95A_y{}^4 - 20A_x{}^3A_z - 88A_x{}^2A_yA_z + 88A_xA_y{}^2A_z + 20A_y{}^3A_z + 120A_x{}^2A_z{}^2 - 88A_xA_yA_z{}^2 + 120A_y{}^2A_z{}^2 - 20A_xA_z{}^3 + 20A_yA_z{}^3 + 95A_z{}^4\right)$$

$$+ \frac{21}{64}\left(20A_x{}^4 + 35A_x{}^3A_y + 30A_x{}^2A_y{}^2 + 35A_xA_y{}^3 + 20A_y{}^4 + 25A_x{}^3A_z - 22A_x{}^2A_yA_z + 22A_xA_y{}^2A_z - 25A_y{}^3A_z - 15A_x{}^2A_z{}^2 + 44A_xA_yA_z{}^2 - 15A_y{}^2A_z{}^2 + 10A_xA_z{}^3 - 10A_yA_z{}^3 - 40A_z{}^4\right)\cos 2\theta$$

$$+ \frac{21}{128}\left(-25A_x{}^4 + 80A_x{}^3A_y + 30A_x{}^2A_y{}^2 + 80A_xA_y{}^3 - 25A_y{}^4 + 70A_x{}^3A_z - 22A_x{}^2A_yA_z + 22A_xA_y{}^2A_z - 70A_y{}^3A_z - 15A_x{}^2A_z{}^2 + 44A_xA_yA_z{}^2 - 15A_y{}^2A_z{}^2 + 10A_xA_z{}^3 - 10A_yA_z{}^3 + 50A_z{}^4\right)\cos 4\theta$$

$$+ \frac{21}{64}\left(-10A_x{}^4 + 5A_x{}^3A_y + 30A_x{}^2A_y{}^2 + 5A_xA_y{}^3 - 10A_y{}^4 - 5A_x{}^3A_z - 22A_x{}^2A_yA_z + 22A_xA_y{}^2A_z + 5A_y{}^3A_z + 30A_x{}^2A_z{}^2 - 22A_xA_yA_z{}^2 + 30A_y{}^2A_z{}^2 - 5A_xA_z{}^3 + 5A_yA_z{}^3 - 10A_z{}^4\right)\cos 6\theta$$

$$+ \frac{21}{256}\left(-5A_x{}^4 - 20A_x{}^3A_y + 60A_x{}^2A_y{}^2 - 20A_xA_y{}^3 - 5A_y{}^4 - 40A_x{}^3A_z - 44A_x{}^2A_yA_z + 44A_xA_y{}^2A_z + 40A_y{}^3A_z - 30A_x{}^2A_z{}^2 + 88A_xA_yA_z{}^2 - 30A_y{}^2A_z{}^2 + 20A_xA_z{}^3 - 20A_yA_z{}^3 + 10A_z{}^4\right)\cos 8\theta$$

$$+ \frac{21\sqrt{3}}{64}\left(-20A_x{}^4 + 5A_x{}^3A_y - 5A_xA_y{}^3 + 20A_y{}^4 + 15A_x{}^3A_z + 22A_x{}^2A_yA_z + 22A_xA_y{}^2A_z + 15A_y{}^3A_z - 15A_x{}^2A_z{}^2 + 15A_y{}^2A_z{}^2 + 20A_xA_z{}^3 + 20A_yA_z{}^3\right)\sin 2\theta$$

$$+ \frac{21\sqrt{3}}{128}\left(-25A_x{}^4 - 20A_x{}^3A_y + 20A_xA_y{}^3 + 25A_y{}^4 - 30A_x{}^3A_z - 22A_x{}^2A_yA_z - 22A_xA_y{}^2A_z - 30A_y{}^3A_z + 15A_x{}^2A_z{}^2 - 15A_y{}^2A_z{}^2 - 50A_xA_z{}^3 - 50A_yA_z{}^3\right)\sin 4\theta$$

$$+ \frac{315\sqrt{3}}{64}\left(-A_x{}^3A_y + A_xA_y{}^3 - A_x{}^3A_z - A_y{}^3A_z + A_xA_z{}^3 + A_yA_z{}^3\right)\sin 6\theta$$

$$+ \frac{21\sqrt{3}}{256}\left(5A_x{}^4 - 20A_x{}^3A_y + 20A_xA_y{}^3 - 5A_y{}^4 + 44A_x{}^2A_yA_z + 44A_xA_y{}^2A_z - 30A_x{}^2A_z{}^2 + 30A_y{}^2A_z{}^2 - 20A_xA_z{}^3 - 20A_yA_z{}^3\right)\sin 8\theta$$

$$a_6 = A_x - A_y - A_z + \left(A_x - A_y + 2A_z\right)\cos 2\theta - \sqrt{3}\left(A_x + A_y\right)\sin 2\theta$$

$$b_6 = -\frac{51}{8}\left({A_x}^2 + {A_y}^2 + {A_z}^2\right) + \frac{1}{4}\left(-17{A_x}^2 - 10A_xA_y - 17{A_y}^2 + 5A_xA_z - 5A_yA_z + 34{A_z}^2\right)\cos 2\theta$$

$$+ \frac{1}{8}\left(17{A_x}^2 - 20A_xA_y + 17{A_y}^2 + 10A_xA_z - 10A_yA_z - 34{A_z}^2\right)\cos 4\theta$$

$$+ \frac{\sqrt{3}}{4}\left(17{A_x}^2 - 17{A_y}^2 + 5A_xA_z + 5A_yA_z\right)\sin 2\theta$$

$$+ \frac{\sqrt{3}}{8}\left(17{A_x}^2 - 17{A_y}^2 - 10A_xA_z - 10A_yA_z\right)\sin 4\theta$$

$$c_6 = \frac{1}{16}\left(-133{A_x}^3 + 29{A_x}^2A_y - 29A_x{A_y}^2 + 133{A_y}^3 + 29{A_x}^2A_z + 28A_xA_yA_z + 29{A_y}^2A_z - 29A_x{A_z}^2\right.$$

$$\left. + 29A_y{A_z}^2 + 133{A_z}^3\right)$$

$$+ \frac{3}{16}\left(-38{A_x}^3 + 29{A_x}^2A_y - 29A_x{A_y}^2 + 38{A_y}^3 - 29{A_x}^2A_z - 29{A_y}^2A_z\right.$$

$$\left. - 76{A_z}^3\right)\cos 2\theta$$

$$+ \frac{3}{16}\left(19{A_x}^3 + 29{A_x}^2A_y - 29A_x{A_y}^2 - 19{A_y}^3 - 29{A_x}^2A_z - 29{A_y}^2A_z\right.$$

$$\left. + 38{A_z}^3\right)\cos 4\theta$$

$$+ \frac{1}{16}\left(38{A_x}^3 + 29{A_x}^2A_y - 29A_x{A_y}^2 - 38{A_y}^3 + 29{A_x}^2A_z + 28A_xA_yA_z + 29{A_y}^2A_z\right.$$

$$\left. - 29A_x{A_z}^2 + 29A_y{A_z}^2 - 38{A_z}^3\right)\cos 6\theta$$

$$+ \frac{\sqrt{3}}{16}\left(114{A_x}^3 + 29{A_x}^2A_y + 29A_x{A_y}^2 + 114{A_y}^3 - 29{A_x}^2A_z + 29{A_y}^2A_z + 58A_x{A_z}^2\right.$$

$$\left. + 58A_y{A_z}^2\right)\sin 2\theta$$

$$+ \frac{\sqrt{3}}{16}\left(57{A_x}^3 - 29{A_x}^2A_y - 29A_x{A_y}^2 + 57{A_y}^3 + 29{A_x}^2A_z - 29{A_y}^2A_z - 58A_x{A_z}^2\right.$$

$$\left. - 58A_y{A_z}^2\right)\sin 4\theta$$

$$+ \frac{29\sqrt{3}}{16}\left(-{A_x}^2A_y - A_x{A_y}^2 + {A_x}^2A_z - {A_y}^2A_z + A_x{A_z}^2 + A_y{A_z}^2\right)\sin 6\theta$$

$$d_6 = \frac{21}{256}\left(95{A_x}^4 + 20{A_x}^3A_y + 120{A_x}^2{A_y}^2 + 20A_x{A_y}^3 + 95{A_y}^4 + 20{A_x}^3A_z + 88{A_x}^2A_yA_z \right.$$
$$- 88A_x{A_y}^2A_z - 20{A_y}^3A_z + 120{A_x}^2{A_z}^2 - 88A_xA_y{A_z}^2 + 120{A_y}^2{A_z}^2 + 20A_x{A_z}^3$$
$$\left. - 20A_y{A_z}^3 + 95{A_z}^4\right)$$
$$+ \frac{21}{64}\left(20{A_x}^4 + 35{A_x}^3A_y + 30{A_x}^2{A_y}^2 + 35A_x{A_y}^3 + 20{A_y}^4 - 25{A_x}^3A_z + 22{A_x}^2A_yA_z \right.$$
$$- 22A_x{A_y}^2A_z + 25{A_y}^3A_z - 15{A_x}^2{A_z}^2 + 44A_xA_y{A_z}^2 - 15{A_y}^2{A_z}^2 - 10A_x{A_z}^3$$
$$\left. + 10A_y{A_z}^3 - 40{A_z}^4\right)\cos 2\theta$$
$$+ \frac{21}{128}\left(-25{A_x}^4 + 80{A_x}^3A_y + 30{A_x}^2{A_y}^2 + 80A_x{A_y}^3 - 25{A_y}^4 - 70{A_x}^3A_z \right.$$
$$+ 22{A_x}^2A_yA_z - 22A_x{A_y}^2A_z + 70{A_y}^3A_z - 15{A_x}^2{A_z}^2 + 44A_xA_y{A_z}^2 - 15{A_y}^2{A_z}^2$$
$$\left. - 10A_x{A_z}^3 + 10A_y{A_z}^3 + 50{A_z}^4\right)\cos 4\theta$$
$$+ \frac{21}{64}\left(-10{A_x}^4 + 5{A_x}^3A_y + 30{A_x}^2{A_y}^2 + 5A_x{A_y}^3 - 10{A_y}^4 + 5{A_x}^3A_z + 22{A_x}^2A_yA_z \right.$$
$$- 22A_x{A_y}^2A_z - 5{A_y}^3A_z + 30{A_x}^2{A_z}^2 - 22A_xA_y{A_z}^2 + 30{A_y}^2{A_z}^2 + 5A_x{A_z}^3 - 5A_y{A_z}^3$$
$$\left. - 10{A_z}^4\right)\cos 6\theta$$
$$+ \frac{21}{256}\left(-5{A_x}^4 - 20{A_x}^3A_y + 60{A_x}^2{A_y}^2 - 20A_x{A_y}^3 - 5{A_y}^4 + 40{A_x}^3A_z + 44{A_x}^2A_yA_z \right.$$
$$- 44A_x{A_y}^2A_z - 40{A_y}^3A_z - 30{A_x}^2{A_z}^2 + 88A_xA_y{A_z}^2 - 30{A_y}^2{A_z}^2 - 20A_x{A_z}^3$$
$$\left. + 20A_y{A_z}^3 + 10{A_z}^4\right)\cos 8\theta$$
$$+ \frac{21\sqrt{3}}{64}\left(-20{A_x}^4 + 5{A_x}^3A_y - 5A_x{A_y}^3 + 20{A_y}^4 - 15{A_x}^3A_z - 22{A_x}^2A_yA_z \right.$$
$$\left. - 22A_x{A_y}^2A_z - 15{A_y}^3A_z - 15{A_x}^2{A_z}^2 + 15{A_y}^2{A_z}^2 - 20A_x{A_z}^3 - 20A_y{A_z}^3\right)\sin 2\theta$$
$$+ \frac{21\sqrt{3}}{128}\left(-25{A_x}^4 - 20{A_x}^3A_y + 20A_x{A_y}^3 + 25{A_y}^4 + 30{A_x}^3A_z + 22{A_x}^2A_yA_z \right.$$
$$\left. + 22A_x{A_y}^2A_z + 30{A_y}^3A_z + 15{A_x}^2{A_z}^2 - 15{A_y}^2{A_z}^2 + 50A_x{A_z}^3 + 50A_y{A_z}^3\right)\sin 4\theta$$
$$+ \frac{315\sqrt{3}}{64}\left(-{A_x}^3A_y + A_x{A_y}^3 + {A_x}^3A_z + {A_y}^3A_z - A_x{A_z}^3 - A_y{A_z}^3\right)\sin 6\theta$$
$$+ \frac{21\sqrt{3}}{256}\left(5{A_x}^4 - 20{A_x}^3A_y + 20A_x{A_y}^3 - 5{A_y}^4 - 44{A_x}^2A_yA_z - 44A_x{A_y}^2A_z \right.$$
$$\left. - 30{A_x}^2{A_z}^2 + 30{A_y}^2{A_z}^2 + 20A_x{A_z}^3 + 20A_y{A_z}^3\right)\sin 8\theta$$

$$a_7 = -A_x - A_y + A_z - \left(A_x + A_y + 2A_z\right)\cos 2\theta + \sqrt{3}\left(A_x - A_y\right)\sin 2\theta$$

$$b_7 = -\frac{51}{8}\left({A_x}^2 + {A_y}^2 + {A_z}^2\right) + \frac{1}{4}\left(-17{A_x}^2 + 10A_xA_y - 17{A_y}^2 + 5A_xA_z + 5A_yA_z + 34{A_z}^2\right)\cos 2\theta$$

$$+ \frac{1}{8}\left(17{A_x}^2 + 20A_xA_y + 17{A_y}^2 + 10A_xA_z + 10A_yA_z - 34{A_z}^2\right)\cos 4\theta$$

$$+ \frac{\sqrt{3}}{4}\left(17{A_x}^2 - 17{A_y}^2 + 5A_xA_z - 5A_yA_z\right)\sin 2\theta$$

$$+ \frac{\sqrt{3}}{8}\left(17{A_x}^2 - 17{A_y}^2 - 10A_xA_z + 10A_yA_z\right)\sin 4\theta$$

$$c_7 = \frac{1}{16}\left(133{A_x}^3 + 29{A_x}^2A_y + 29A_x{A_y}^2 + 133{A_y}^3 - 29{A_x}^2A_z + 28A_xA_yA_z - 29{A_y}^2A_z + 29A_x{A_z}^2\right.$$

$$\left. + 29A_y{A_z}^2 - 133{A_z}^3\right)$$

$$+ \frac{3}{16}\left(38{A_x}^3 + 29{A_x}^2A_y + 29A_x{A_y}^2 + 38{A_y}^3 + 29{A_x}^2A_z + 29{A_y}^2A_z\right.$$

$$\left. + 76{A_z}^3\right)\cos 2\theta$$

$$+ \frac{3}{16}\left(-19{A_x}^3 + 29{A_x}^2A_y + 29A_x{A_y}^2 - 19{A_y}^3 + 29{A_x}^2A_z + 29{A_y}^2A_z\right.$$

$$\left. - 38{A_z}^3\right)\cos 4\theta$$

$$+ \frac{1}{16}\left(-38{A_x}^3 + 29{A_x}^2A_y + 29A_x{A_y}^2 - 38{A_y}^3 - 29{A_x}^2A_z + 28A_xA_yA_z - 29{A_y}^2A_z\right.$$

$$\left. + 29A_x{A_z}^2 + 29A_y{A_z}^2 + 38{A_z}^3\right)\cos 6\theta$$

$$+ \frac{\sqrt{3}}{16}\left(-114{A_x}^3 + 29{A_x}^2A_y - 29A_x{A_y}^2 + 114{A_y}^3 + 29{A_x}^2A_z - 29{A_y}^2A_z - 58A_x{A_z}^2\right.$$

$$\left. + 58A_y{A_z}^2\right)\sin 2\theta$$

$$+ \frac{\sqrt{3}}{16}\left(-57{A_x}^3 - 29{A_x}^2A_y + 29A_x{A_y}^2 + 57{A_y}^3 - 29{A_x}^2A_z + 29{A_y}^2A_z + 58A_x{A_z}^2\right.$$

$$\left. - 58A_y{A_z}^2\right)\sin 4\theta$$

$$+ \frac{29\sqrt{3}}{16}\left(-{A_x}^2A_y + A_x{A_y}^2 - {A_x}^2A_z + {A_y}^2A_z - A_x{A_z}^2 + A_y{A_z}^2\right)\sin 6\theta$$

$$
\begin{aligned}
d_7 = \frac{21}{256}\big(&95{A_x}^4 - 20{A_x}^3A_y + 120{A_x}^2{A_y}^2 - 20A_x{A_y}^3 + 95{A_y}^4 + 20{A_x}^3A_z - 88{A_x}^2A_yA_z \\
&- 88A_x{A_y}^2A_z + 20{A_y}^3A_z + 120{A_x}^2{A_z}^2 + 88A_xA_y{A_z}^2 + 120{A_y}^2{A_z}^2 + 20A_x{A_z}^3 \\
&+ 20A_y{A_z}^3 + 95{A_z}^4\big) \\
&+ \frac{21}{64}\big(20{A_x}^4 - 35{A_x}^3A_y + 30{A_x}^2{A_y}^2 - 35A_x{A_y}^3 + 20{A_y}^4 - 25{A_x}^3A_z - 22{A_x}^2A_yA_z \\
&- 22A_x{A_y}^2A_z - 25{A_y}^3A_z - 15{A_x}^2{A_z}^2 - 44A_xA_y{A_z}^2 - 15{A_y}^2{A_z}^2 - 10A_x{A_z}^3 \\
&- 10A_y{A_z}^3 - 40{A_z}^4\big)\cos 2\theta \\
&+ \frac{21}{128}\big(-25{A_x}^4 - 80{A_x}^3A_y + 30{A_x}^2{A_y}^2 - 80A_x{A_y}^3 - 25{A_y}^4 - 70{A_x}^3A_z \\
&- 22{A_x}^2A_yA_z - 22A_x{A_y}^2A_z - 70{A_y}^3A_z - 15{A_x}^2{A_z}^2 - 44A_xA_y{A_z}^2 - 15{A_y}^2{A_z}^2 \\
&- 10A_x{A_z}^3 - 10A_y{A_z}^3 + 50{A_z}^4\big)\cos 4\theta \\
&+ \frac{21}{64}\big(-10{A_x}^4 - 5{A_x}^3A_y + 30{A_x}^2{A_y}^2 - 5A_x{A_y}^3 - 10{A_y}^4 + 5{A_x}^3A_z - 22{A_x}^2A_yA_z \\
&- 22A_x{A_y}^2A_z + 5{A_y}^3A_z + 30{A_x}^2{A_z}^2 + 22A_xA_y{A_z}^2 + 30{A_y}^2{A_z}^2 + 5A_x{A_z}^3 + 5A_y{A_z}^3 \\
&- 10{A_z}^4\big)\cos 6\theta \\
&+ \frac{21}{256}\big(-5{A_x}^4 + 20{A_x}^3A_y + 60{A_x}^2{A_y}^2 + 20A_x{A_y}^3 - 5{A_y}^4 + 40{A_x}^3A_z - 44{A_x}^2A_yA_z \\
&- 44A_x{A_y}^2A_z + 40{A_y}^3A_z - 30{A_x}^2{A_z}^2 - 88A_xA_y{A_z}^2 - 30{A_y}^2{A_z}^2 - 20A_x{A_z}^3 \\
&- 20A_y{A_z}^3 + 10{A_z}^4\big)\cos 8\theta \\
&+ \frac{21\sqrt{3}}{64}\big(-20{A_x}^4 - 5{A_x}^3A_y + 5A_x{A_y}^3 + 20{A_y}^4 - 15{A_x}^3A_z + 22{A_x}^2A_yA_z \\
&- 22A_x{A_y}^2A_z + 15{A_y}^3A_z - 15{A_x}^2{A_z}^2 + 15{A_y}^2{A_z}^2 - 20A_x{A_z}^3 + 20A_y{A_z}^3\big)\sin 2\theta \\
&+ \frac{21\sqrt{3}}{128}\big(-25{A_x}^4 + 20{A_x}^3A_y - 20A_x{A_y}^3 + 25{A_y}^4 + 30{A_x}^3A_z - 22{A_x}^2A_yA_z \\
&+ 22A_x{A_y}^2A_z - 30{A_y}^3A_z + 15{A_x}^2{A_z}^2 - 15{A_y}^2{A_z}^2 + 50A_x{A_z}^3 - 50A_y{A_z}^3\big)\sin 4\theta \\
&+ \frac{315\sqrt{3}}{64}\big({A_x}^3A_y - A_x{A_y}^3 + {A_x}^3A_z - {A_y}^3A_z - A_x{A_z}^3 + A_y{A_z}^3\big)\sin 6\theta \\
&+ \frac{21\sqrt{3}}{256}\big(5{A_x}^4 + 20{A_x}^3A_y - 20A_x{A_y}^3 - 5{A_y}^4 + 44{A_x}^2A_yA_z - 44A_x{A_y}^2A_z \\
&- 30{A_x}^2{A_z}^2 + 30{A_y}^2{A_z}^2 + 20A_x{A_z}^3 - 20A_y{A_z}^3\big)\sin 8\theta
\end{aligned}
$$

$$
a_8 = A_x + A_y + A_z + \left(A_x + A_y - 2A_z\right)\cos 2\theta - \sqrt{3}\left(A_x - A_y\right)\sin 2\theta
$$

$$b_8 = -\frac{51}{8}\left(A_x{}^2 + A_y{}^2 + A_z{}^2\right) + \frac{1}{4}\left(-17A_x{}^2 + 10A_xA_y - 17A_y{}^2 - 5A_xA_z - 5A_yA_z + 34A_z{}^2\right)\cos 2\theta$$

$$+ \frac{1}{8}\left(17A_x{}^2 + 20A_xA_y + 17A_y{}^2 - 10A_xA_z - 10A_yA_z - 34A_z{}^2\right)\cos 4\theta$$

$$+ \frac{\sqrt{3}}{4}\left(17A_x{}^2 - 17A_y{}^2 - 5A_xA_z + 5A_yA_z\right)\sin 2\theta$$

$$+ \frac{\sqrt{3}}{8}\left(17A_x{}^2 - 17A_y{}^2 + 10A_xA_z - 10A_yA_z\right)\sin 4\theta$$

$$c_8 = \frac{1}{16}\left(-133A_x{}^3 - 29A_x{}^2A_y - 29A_xA_y{}^2 - 133A_y{}^3 - 29A_x{}^2A_z + 28A_xA_yA_z - 29A_y{}^2A_z - 29A_xA_z{}^2\right.$$

$$\left.- 29A_yA_z{}^2 - 133A_z{}^3\right)$$

$$+ \frac{3}{16}\left(-38A_x{}^3 - 29A_x{}^2A_y - 29A_xA_y{}^2 - 38A_y{}^3 + 29A_x{}^2A_z + 29A_y{}^2A_z\right.$$

$$\left.+ 76A_z{}^3\right)\cos 2\theta$$

$$+ \frac{3}{16}\left(19A_x{}^3 - 29A_x{}^2A_y - 29A_xA_y{}^2 + 19A_y{}^3 + 29A_x{}^2A_z + 29A_y{}^2A_z\right.$$

$$\left.- 38A_z{}^3\right)\cos 4\theta$$

$$+ \frac{1}{16}\left(38A_x{}^3 - 29A_x{}^2A_y - 29A_xA_y{}^2 + 38A_y{}^3 - 29A_x{}^2A_z + 28A_xA_yA_z - 29A_y{}^2A_z\right.$$

$$\left.- 29A_xA_z{}^2 - 29A_yA_z{}^2 + 38A_z{}^3\right)\cos 6\theta$$

$$+ \frac{\sqrt{3}}{16}\left(114A_x{}^3 - 29A_x{}^2A_y + 29A_xA_y{}^2 - 114A_y{}^3 + 29A_x{}^2A_z - 29A_y{}^2A_z + 58A_xA_z{}^2\right.$$

$$\left.- 58A_yA_z{}^2\right)\sin 2\theta$$

$$+ \frac{\sqrt{3}}{16}\left(57A_x{}^3 + 29A_x{}^2A_y - 29A_xA_y{}^2 - 57A_y{}^3 - 29A_x{}^2A_z + 29A_y{}^2A_z - 58A_xA_z{}^2\right.$$

$$\left.+ 58A_yA_z{}^2\right)\sin 4\theta$$

$$+ \frac{29\sqrt{3}}{16}\left(A_x{}^2A_y - A_xA_y{}^2 - A_x{}^2A_z + A_y{}^2A_z + A_xA_z{}^2 - A_yA_z{}^2\right)\sin 6\theta$$

$$
\begin{aligned}
d_8 = \frac{21}{256}\big(&95{A_x}^4 - 20{A_x}^3A_y + 120{A_x}^2{A_y}^2 - 20A_x{A_y}^3 + 95{A_y}^4 - 20{A_x}^3A_z + 88{A_x}^2A_yA_z \\
&+ 88A_x{A_y}^2A_z - 20{A_y}^3A_z + 120{A_x}^2{A_z}^2 + 88A_xA_y{A_z}^2 + 120{A_y}^2{A_z}^2 - 20A_x{A_z}^3 \\
&- 20A_y{A_z}^3 + 95{A_z}^4\big) \\
&+ \frac{21}{64}\big(20{A_x}^4 - 35{A_x}^3A_y + 30{A_x}^2{A_y}^2 - 35A_x{A_y}^3 + 20{A_y}^4 + 25{A_x}^3A_z + 22{A_x}^2A_yA_z \\
&+ 22A_x{A_y}^2A_z + 25{A_y}^3A_z - 15{A_x}^2{A_z}^2 - 44A_xA_y{A_z}^2 - 15{A_y}^2{A_z}^2 + 10A_x{A_z}^3 \\
&+ 10A_y{A_z}^3 - 40{A_z}^4\big)\cos 2\theta \\
&+ \frac{21}{128}\big(-25{A_x}^4 - 80{A_x}^3A_y + 30{A_x}^2{A_y}^2 - 80A_x{A_y}^3 - 25{A_y}^4 + 70{A_x}^3A_z \\
&+ 22{A_x}^2A_yA_z + 22A_x{A_y}^2A_z + 70{A_y}^3A_z - 15{A_x}^2{A_z}^2 - 22A_xA_y{A_z}^2 - 15{A_y}^2{A_z}^2 \\
&+ 10A_x{A_z}^3 + 10A_y{A_z}^3 + 50{A_z}^4\big)\cos 4\theta \\
&+ \frac{21}{64}\big(-10{A_x}^4 - 5{A_x}^3A_y + 30{A_x}^2{A_y}^2 - 5A_x{A_y}^3 - 10{A_y}^4 - 5{A_x}^3A_z + 22{A_x}^2A_yA_z \\
&+ 22A_x{A_y}^2A_z - 5{A_y}^3A_z + 30{A_x}^2{A_z}^2 + 22A_xA_y{A_z}^2 + 30{A_y}^2{A_z}^2 - 5A_x{A_z}^3 - 5A_y{A_z}^3 \\
&- 10{A_z}^4\big)\cos 6\theta \\
&+ \frac{21}{256}\big(-5{A_x}^4 + 20{A_x}^3A_y + 60{A_x}^2{A_y}^2 + 20A_x{A_y}^3 - 5{A_y}^4 - 40{A_x}^3A_z + 44{A_x}^2A_yA_z \\
&+ 44A_x{A_y}^2A_z - 40{A_y}^3A_z - 30{A_x}^2{A_z}^2 - 88A_xA_y{A_z}^2 - 30{A_y}^2{A_z}^2 + 20A_x{A_z}^3 \\
&+ 20A_y{A_z}^3 + 10{A_z}^4\big)\cos 8\theta \\
&+ \frac{21\sqrt{3}}{64}\big(-20{A_x}^4 - 5{A_x}^3A_y + 5A_x{A_y}^3 + 20{A_y}^4 + 15{A_x}^3A_z - 22{A_x}^2A_yA_z \\
&+ 22A_x{A_y}^2A_z - 15{A_y}^3A_z - 15{A_x}^2{A_z}^2 + 15{A_y}^2{A_z}^2 + 20A_x{A_z}^3 - 20A_y{A_z}^3\big)\sin 2\theta \\
&+ \frac{21\sqrt{3}}{128}\big(-25{A_x}^4 + 20{A_x}^3A_y - 20A_x{A_y}^3 + 25{A_y}^4 - 30{A_x}^3A_z + 22{A_x}^2A_yA_z \\
&- 22A_x{A_y}^2A_z + 30{A_y}^3A_z + 15{A_x}^2{A_z}^2 - 15{A_y}^2{A_z}^2 - 50A_x{A_z}^3 + 50A_y{A_z}^3\big)\sin 4\theta \\
&+ \frac{315\sqrt{3}}{64}\big({A_x}^3A_y - A_x{A_y}^3 - {A_x}^3A_z + {A_y}^3A_z + A_x{A_z}^3 - A_y{A_z}^3\big)\sin 6\theta \\
&+ \frac{21\sqrt{3}}{256}\big(5{A_x}^4 + 20{A_x}^3A_y - 20A_x{A_y}^3 - 5{A_y}^4 - 44{A_x}^2A_yA_z + 44A_x{A_y}^2A_z \\
&- 30{A_x}^2{A_z}^2 + 30{A_y}^2{A_z}^2 - 20A_x{A_z}^3 + 20A_y{A_z}^3\big)\sin 8\theta
\end{aligned}
$$

The set of coefficients for i = 6, 7 and 8 can be obtained from $\{a_5, b_5, c_5, d_5\}$ with replacing A_x and A_y to $-A_x$ and $-A_y$, A_x and A_z to $-A_x$ and $-A_z$, and A_y and A_z to $-A_y$ and $-A_z$, respectively.

The second-order correction for the energies $\varepsilon_{\sigma,\alpha}^{(02)}$ $(\sigma = +,-;\ \alpha = 1,\cdots,16)$ can be written as

$$
\varepsilon_{\pm,\alpha}^{(02)} = \sum_{\beta=1}^{4} \frac{|\langle \pm,\alpha|H_{\mathrm{hfs}}|\mp,\beta\rangle|^2}{\varepsilon_{\pm}^{(0)} - \varepsilon_{\mp}^{(0)}}
$$

Both the upper and lower signs should be chosen in the double sign. The non-zero matrix elements of H_{hfs} expanded to the basis belonging different eigenspaces are,

$$\begin{aligned}\langle +,1|H_{\mathrm{hfs}}|-,2\rangle &= \langle -,2|H_{\mathrm{hfs}}|+,1\rangle = \langle +,7|H_{\mathrm{hfs}}|-,8\rangle = \langle -,8|H_{\mathrm{hfs}}|+,7\rangle = \langle -,9|H_{\mathrm{hfs}}|+,10\rangle \\ &= \langle +,10|H_{\mathrm{hfs}}|-,9\rangle = \langle -,15|H_{\mathrm{hfs}}|+,16\rangle = \langle +,16|H_{\mathrm{hfs}}|-,15\rangle \\ &= \frac{\sqrt{21}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{7}}{4}\left(A_x + A_y\right)\sin 2\theta\end{aligned}$$

$$\begin{aligned}\langle +,2|H_{\mathrm{hfs}}|-,1\rangle &= \langle -,1|H_{\mathrm{hfs}}|+,2\rangle = \langle +,8|H_{\mathrm{hfs}}|-,7\rangle = \langle -,7|H_{\mathrm{hfs}}|+,8\rangle = \langle -,10|H_{\mathrm{hfs}}|+,9\rangle \\ &= \langle +,9|H_{\mathrm{hfs}}|-,10\rangle = \langle -,16|H_{\mathrm{hfs}}|+,15\rangle = \langle +,15|H_{\mathrm{hfs}}|-,16\rangle \\ &= \frac{\sqrt{21}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{7}}{4}\left(A_x - A_y\right)\sin 2\theta\end{aligned}$$

$$\begin{aligned}\langle +,2|H_{\mathrm{hfs}}|-,3\rangle &= \langle -,3|H_{\mathrm{hfs}}|+,2\rangle = \langle +,6|H_{\mathrm{hfs}}|-,7\rangle = \langle -,7|H_{\mathrm{hfs}}|+,6\rangle = \langle -,10|H_{\mathrm{hfs}}|+,11\rangle \\ &= \langle +,11|H_{\mathrm{hfs}}|-,10\rangle = \langle -,14|H_{\mathrm{hfs}}|+,15\rangle = \langle +,15|H_{\mathrm{hfs}}|-,14\rangle \\ &= \frac{3}{2}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(A_x + A_y\right)\sin 2\theta\end{aligned}$$

$$\begin{aligned}\langle +,3|H_{\mathrm{hfs}}|-,2\rangle &= \langle -,2|H_{\mathrm{hfs}}|+,3\rangle = \langle +,7|H_{\mathrm{hfs}}|-,6\rangle = \langle -,6|H_{\mathrm{hfs}}|+,7\rangle = \langle -,11|H_{\mathrm{hfs}}|+,10\rangle \\ &= \langle +,10|H_{\mathrm{hfs}}|-,11\rangle = \langle -,15|H_{\mathrm{hfs}}|+,14\rangle = \langle +,14|H_{\mathrm{hfs}}|-,15\rangle \\ &= \frac{3}{2}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(A_x - A_y\right)\sin 2\theta\end{aligned}$$

$$\begin{aligned}\langle +,3|H_{\mathrm{hfs}}|-,4\rangle &= \langle -,4|H_{\mathrm{hfs}}|+,3\rangle = \langle +,5|H_{\mathrm{hfs}}|-,6\rangle = \langle -,6|H_{\mathrm{hfs}}|+,5\rangle = \langle -,11|H_{\mathrm{hfs}}|+,12\rangle \\ &= \langle +,12|H_{\mathrm{hfs}}|-,11\rangle = \langle -,13|H_{\mathrm{hfs}}|+,14\rangle = \langle +,14|H_{\mathrm{hfs}}|-,13\rangle \\ &= \frac{3\sqrt{5}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{15}}{4}\left(A_x + A_y\right)\sin 2\theta\end{aligned}$$

$$\begin{aligned}\langle +,4|H_{\mathrm{hfs}}|-,3\rangle &= \langle -,3|H_{\mathrm{hfs}}|+,4\rangle = \langle +,6|H_{\mathrm{hfs}}|-,5\rangle = \langle -,5|H_{\mathrm{hfs}}|+,6\rangle = \langle -,12|H_{\mathrm{hfs}}|+,11\rangle \\ &= \langle +,11|H_{\mathrm{hfs}}|-,12\rangle = \langle -,14|H_{\mathrm{hfs}}|+,13\rangle = \langle +,13|H_{\mathrm{hfs}}|-,14\rangle \\ &= \frac{3\sqrt{5}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{15}}{4}\left(A_x - A_y\right)\sin 2\theta\end{aligned}$$

$$\begin{aligned}\langle +,4|H_{\mathrm{hfs}}|-,5\rangle &= \langle -,5|H_{\mathrm{hfs}}|+,4\rangle = \langle -,12|H_{\mathrm{hfs}}|+,13\rangle = \langle +,13|H_{\mathrm{hfs}}|-,12\rangle \\ &= \sqrt{3}\left(A_x - A_y\right)\cos 2\theta + \left(A_x + A_y\right)\sin 2\theta\end{aligned}$$

$$\begin{aligned}\langle +,5|H_{\mathrm{hfs}}|-,4\rangle &= \langle -,4|H_{\mathrm{hfs}}|+,5\rangle = \langle -,13|H_{\mathrm{hfs}}|+,12\rangle = \langle +,12|H_{\mathrm{hfs}}|-,13\rangle \\ &= \sqrt{3}\left(A_x + A_y\right)\cos 2\theta + \left(A_x - A_y\right)\sin 2\theta\end{aligned}$$

$$\langle +,1|H_{\mathrm{hfs}}|-,9\rangle = \langle -,9|H_{\mathrm{hfs}}|+,1\rangle = \langle -,8|H_{\mathrm{hfs}}|+,16\rangle = \langle +,16|H_{\mathrm{hfs}}|-,8\rangle = -\frac{7}{2}A_x\sin 2\theta$$

$$\langle +,2|H_{\mathrm{hfs}}|-,10\rangle = \langle -,10|H_{\mathrm{hfs}}|+,2\rangle = \langle -,7|H_{\mathrm{hfs}}|+,15\rangle = \langle +,15|H_{\mathrm{hfs}}|-,7\rangle = -\frac{5}{2}A_x\sin 2\theta$$

$$\langle +,3|H_{\mathrm{hfs}}|-,11\rangle = \langle -,11|H_{\mathrm{hfs}}|+,3\rangle = \langle -,6|H_{\mathrm{hfs}}|+,14\rangle = \langle +,14|H_{\mathrm{hfs}}|-,6\rangle = -\frac{3}{2}A_x\sin 2\theta$$

$$\langle +,4|H_{\mathrm{hfs}}|-,12\rangle = \langle -,12|H_{\mathrm{hfs}}|+,4\rangle = \langle -,5|H_{\mathrm{hfs}}|+,13\rangle = \langle +,13|H_{\mathrm{hfs}}|-,5\rangle = -\frac{A_x}{2}\sin 2\theta$$

$$\langle +,5|H_{\mathrm{hfs}}|-,13\rangle = \langle -,13|H_{\mathrm{hfs}}|+,5\rangle = \langle -,4|H_{\mathrm{hfs}}|+,12\rangle = \langle +,12|H_{\mathrm{hfs}}|-,4\rangle = \frac{A_x}{2}\sin 2\theta$$

$$\langle +,6|H_{\mathrm{hfs}}|-,14\rangle = \langle -,14|H_{\mathrm{hfs}}|+,6\rangle = \langle -,3|H_{\mathrm{hfs}}|+,11\rangle = \langle +,11|H_{\mathrm{hfs}}|-,3\rangle = \frac{3}{2}A_x \sin 2\theta$$

$$\langle +,7|H_{\mathrm{hfs}}|-,15\rangle = \langle -,15|H_{\mathrm{hfs}}|+,7\rangle = \langle -,2|H_{\mathrm{hfs}}|+,10\rangle = \langle +,10|H_{\mathrm{hfs}}|-,2\rangle = \frac{5}{2}A_x \sin 2\theta$$

$$\langle +,8|H_{\mathrm{hfs}}|-,16\rangle = \langle -,16|H_{\mathrm{hfs}}|+,8\rangle = \langle -,1|H_{\mathrm{hfs}}|+,9\rangle = \langle +,9|H_{\mathrm{hfs}}|-,1\rangle = \frac{7}{2}A_x \sin 2\theta$$

Therefore, the second-order corrections for the energy, $\varepsilon_{\sigma,\alpha}^{(02)}$ $(\sigma = +,-;\ \alpha = 1,\cdots,12)$ are in the following:

$$\varepsilon_{+,1}^{(02)} = \frac{|\langle -,2|H_{\mathrm{hfs}}|+,1\rangle|^2 + |\langle -,9|H_{\mathrm{hfs}}|+,1\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{\sqrt{21}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{7}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \frac{49}{4}{A_z}^2 \sin^2 2\theta\right\}$$

$$\varepsilon_{+,2}^{(02)} = \frac{|\langle -,1|H_{\mathrm{hfs}}|+,2\rangle|^2 + |\langle -,3|H_{\mathrm{hfs}}|+,2\rangle|^2 + |\langle -,10|H_{\mathrm{hfs}}|+,2\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{\sqrt{21}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{7}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2\right.$$

$$\left. + \left[\frac{3}{2}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \frac{25}{4}{A_z}^2 \sin^2 2\theta\right\}$$

$$\varepsilon_{+,3}^{(02)} = \frac{|\langle -,2|H_{\mathrm{hfs}}|+,3\rangle|^2 + |\langle -,4|H_{\mathrm{hfs}}|+,3\rangle|^2 + |\langle -,11|H_{\mathrm{hfs}}|+,3\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{3}{2}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(A_x - A_y\right)\sin 2\theta\right]^2\right.$$

$$\left. + \left[\frac{3\sqrt{5}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{15}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \frac{9}{4}{A_z}^2 \sin^2 2\theta\right\}$$

$$\varepsilon_{+,4}^{(02)} = \frac{|\langle -,3|H_{\mathrm{hfs}}|+,4\rangle|^2 + |\langle -,5|H_{\mathrm{hfs}}|+,4\rangle|^2 + |\langle -,12|H_{\mathrm{hfs}}|+,4\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{3\sqrt{5}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{15}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2\right.$$

$$\left. + \left[\sqrt{3}\left(A_x - A_y\right)\cos 2\theta + \left(A_x + A_y\right)\sin 2\theta\right]^2 + \frac{{A_z}^2}{4}\sin^2 2\theta\right\}$$

$$\varepsilon_{+,5}^{(02)} = \frac{|\langle -,4|H_{\mathrm{hfs}}|+,5\rangle|^2 + |\langle -,6|H_{\mathrm{hfs}}|+,5\rangle|^2 + |\langle -,13|H_{\mathrm{hfs}}|+,5\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\sqrt{3}\left(A_x + A_y\right)\cos 2\theta + \left(A_x - A_y\right)\sin 2\theta\right]^2\right.$$

$$\left. + \left[\frac{3\sqrt{5}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{15}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \frac{{A_z}^2}{4}\sin^2 2\theta\right\}$$

$$\varepsilon_{+,6}^{(02)} = \frac{|\langle -,5|H_{\mathrm{hfs}}|+,6\rangle|^2 + |\langle -,7|H_{\mathrm{hfs}}|+,6\rangle|^2 + |\langle -,14|H_{\mathrm{hfs}}|+,6\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{3\sqrt{5}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{15}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2 \right.$$

$$\left. + \left[\frac{3}{2}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \frac{9}{4}{A_z}^2\sin^2 2\theta\right\}$$

$$\varepsilon_{+,7}^{(02)} = \frac{|\langle -,6|H_{\mathrm{hfs}}|+,7\rangle|^2 + |\langle -,8|H_{\mathrm{hfs}}|+,7\rangle|^2 + |\langle -,15|H_{\mathrm{hfs}}|+,7\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{3}{2}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(A_x - A_y\right)\sin 2\theta\right]^2 \right.$$

$$\left. + \left[\frac{\sqrt{21}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{7}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \frac{25}{4}{A_z}^2\sin^2 2\theta\right\}$$

$$\varepsilon_{+,8}^{(02)} = \frac{|\langle -,7|H_{\mathrm{hfs}}|+,8\rangle|^2 + |\langle -,16|H_{\mathrm{hfs}}|+,8\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\left[\frac{\sqrt{21}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{7}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2 + \frac{49}{4}{A_z}^2\sin^2 2\theta\right\}$$

$$\varepsilon_{+,9}^{(02)} = \frac{|\langle -,1|H_{\mathrm{hfs}}|+,9\rangle|^2 + |\langle -,10|H_{\mathrm{hfs}}|+,9\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{49}{4}{A_z}^2\sin^2 2\theta + \left[\frac{\sqrt{21}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{7}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{+,10}^{(02)} = \frac{|\langle -,2|H_{\mathrm{hfs}}|+,10\rangle|^2 + |\langle -,9|H_{\mathrm{hfs}}|+,10\rangle|^2 + |\langle -,11|H_{\mathrm{hfs}}|+,10\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{25}{4}{A_z}^2\sin^2 2\theta + \left[\frac{\sqrt{21}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{7}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 \right.$$

$$\left. + \left[\frac{3}{2}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(A_x - A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{+,11}^{(02)} = \frac{|\langle -,3|H_{\mathrm{hfs}}|+,11\rangle|^2 + |\langle -,10|H_{\mathrm{hfs}}|+,11\rangle|^2 + |\langle -,12|H_{\mathrm{hfs}}|+,11\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{9}{4}{A_z}^2\sin^2 2\theta + \left[\frac{3}{2}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(A_x + A_y\right)\sin 2\theta\right]^2 \right.$$

$$\left. + \left[\frac{3\sqrt{5}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{15}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{+,12}^{(02)} = \frac{|\langle -,4|H_{\mathrm{hfs}}|+,12\rangle|^2 + |\langle -,11|H_{\mathrm{hfs}}|+,12\rangle|^2 + |\langle -,13|H_{\mathrm{hfs}}|+,12\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{{A_z}^2}{4}\sin^2 2\theta + \left[\frac{3\sqrt{5}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{15}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 \right.$$

$$\left. + \left[\sqrt{3}\left(A_x + A_y\right)\cos 2\theta + \left(A_x - A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{+,13}^{(02)} = \frac{|\langle -,5|H_{\mathrm{hfs}}|+,13\rangle|^2 + |\langle -,12|H_{\mathrm{hfs}}|+,13\rangle|^2 + |\langle -,14|H_{\mathrm{hfs}}|+,13\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{{A_z}^2}{4}\sin^2 2\theta + \left[\sqrt{3}\left(A_x - A_y\right)\cos 2\theta + \left(A_x + A_y\right)\sin 2\theta\right]^2 + \left[\frac{3\sqrt{5}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{15}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{+,14}^{(02)} = \frac{|\langle -,6|H_{\mathrm{hfs}}|+,14\rangle|^2 + |\langle -,13|H_{\mathrm{hfs}}|+,14\rangle|^2 + |\langle -,15|H_{\mathrm{hfs}}|+,14\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{9}{4}{A_z}^2\sin^2 2\theta + \left[\frac{3\sqrt{5}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{15}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \left[\frac{3}{2}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(A_x - A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{+,15}^{(02)} = \frac{|\langle -,7|H_{\mathrm{hfs}}|+,15\rangle|^2 + |\langle -,14|H_{\mathrm{hfs}}|+,15\rangle|^2 + |\langle -,16|H_{\mathrm{hfs}}|+,15\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{25}{4}{A_z}^2\sin^2 2\theta + \left[\frac{3}{2}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \left[\frac{\sqrt{21}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{7}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{+,16}^{(02)} = \frac{|\langle -,8|H_{\mathrm{hfs}}|+,16\rangle|^2 + |\langle -,15|H_{\mathrm{hfs}}|+,16\rangle|^2}{2\Delta}$$

$$= \frac{1}{2\Delta}\left\{\frac{49}{4}{A_z}^2\sin^2 2\theta + \left[\frac{\sqrt{21}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{7}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{-,1}^{(02)} = \frac{|\langle +,2|H_{\mathrm{hfs}}|-,1\rangle|^2 + |\langle +,9|H_{\mathrm{hfs}}|-,1\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\left[\frac{\sqrt{21}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{7}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2 + \frac{49}{4}{A_z}^2\sin^2 2\theta\right\}$$

$$\varepsilon_{-,2}^{(02)} = \frac{|\langle +,1|H_{\mathrm{hfs}}|-,2\rangle|^2 + |\langle +,3|H_{\mathrm{hfs}}|-,2\rangle|^2 + |\langle +,10|H_{\mathrm{hfs}}|-,2\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\left[\frac{\sqrt{21}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{7}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \left[\frac{3}{2}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(A_x - A_y\right)\sin 2\theta\right]^2 + \frac{25}{4}{A_z}^2\sin^2 2\theta\right\}$$

$$\varepsilon_{-,3}^{(02)} = \frac{|\langle +,2|H_{\mathrm{hfs}}|-,3\rangle|^2 + |\langle +,4|H_{\mathrm{hfs}}|-,3\rangle|^2 + |\langle +,11|H_{\mathrm{hfs}}|-,3\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\left[\frac{3}{2}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(A_x + A_y\right)\sin 2\theta\right]^2 + \left[\frac{3\sqrt{5}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{15}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2 + \frac{9}{4}{A_z}^2\sin^2 2\theta\right\}$$

$$\varepsilon_{-,4}^{(02)}=\frac{|\langle +,3|H_{\mathrm{hfs}}|-,4\rangle|^2+|\langle +,5|H_{\mathrm{hfs}}|-,4\rangle|^2+|\langle +,12|H_{\mathrm{hfs}}|-,4\rangle|^2}{-2\Delta}$$

$$=-\frac{1}{2\Delta}\left\{\left[\frac{3\sqrt{5}}{4}\left(A_x-A_y\right)\cos 2\theta+\frac{\sqrt{15}}{4}\left(A_x+A_y\right)\sin 2\theta\right]^2\right.$$

$$\left.+\left[\sqrt{3}\left(A_x+A_y\right)\cos 2\theta+\left(A_x-A_y\right)\sin 2\theta\right]^2+\frac{{A_z}^2}{4}\sin^2 2\theta\right\}$$

$$\varepsilon_{-,5}^{(02)}=\frac{|\langle +,4|H_{\mathrm{hfs}}|-,5\rangle|^2+|\langle +,6|H_{\mathrm{hfs}}|-,5\rangle|^2+|\langle +,13|H_{\mathrm{hfs}}|-,5\rangle|^2}{-2\Delta}$$

$$=-\frac{1}{2\Delta}\left\{\left[\sqrt{3}\left(A_x-A_y\right)\cos 2\theta+\left(A_x+A_y\right)\sin 2\theta\right]^2\right.$$

$$\left.+\left[\frac{3\sqrt{5}}{4}\left(A_x+A_y\right)\cos 2\theta+\frac{\sqrt{15}}{4}\left(A_x-A_y\right)\sin 2\theta\right]^2+\frac{{A_z}^2}{4}\sin^2 2\theta\right\}$$

$$\varepsilon_{-,6}^{(02)}=\frac{|\langle +,5|H_{\mathrm{hfs}}|-,6\rangle|^2+|\langle +,7|H_{\mathrm{hfs}}|-,6\rangle|^2+|\langle +,14|H_{\mathrm{hfs}}|-,6\rangle|^2}{-2\Delta}$$

$$=-\frac{1}{2\Delta}\left\{\left[\frac{3\sqrt{5}}{4}\left(A_x-A_y\right)\cos 2\theta+\frac{\sqrt{15}}{4}\left(A_x+A_y\right)\sin 2\theta\right]^2\right.$$

$$\left.+\left[\frac{3}{2}\left(A_x+A_y\right)\cos 2\theta+\frac{\sqrt{3}}{2}\left(A_x-A_y\right)\sin 2\theta\right]^2+\frac{9}{4}{A_z}^2\sin^2 2\theta\right\}$$

$$\varepsilon_{-,7}^{(02)}=\frac{|\langle +,6|H_{\mathrm{hfs}}|-,7\rangle|^2+|\langle +,8|H_{\mathrm{hfs}}|-,7\rangle|^2+|\langle +,15|H_{\mathrm{hfs}}|-,7\rangle|^2}{-2\Delta}$$

$$=-\frac{1}{2\Delta}\left\{\left[\frac{3}{2}\left(A_x-A_y\right)\cos 2\theta+\frac{\sqrt{3}}{2}\left(A_x+A_y\right)\sin 2\theta\right]^2\right.$$

$$\left.+\left[\frac{\sqrt{21}}{4}\left(A_x+A_y\right)\cos 2\theta+\frac{\sqrt{7}}{4}\left(A_x-A_y\right)\sin 2\theta\right]^2+\frac{25}{4}{A_z}^2\sin^2 2\theta\right\}$$

$$\varepsilon_{-,8}^{(02)}=\frac{|\langle +,7|H_{\mathrm{hfs}}|-,8\rangle|^2+|\langle +,16|H_{\mathrm{hfs}}|-,8\rangle|^2}{-2\Delta}$$

$$=-\frac{1}{2\Delta}\left\{\left[\frac{\sqrt{21}}{4}\left(A_x-A_y\right)\cos 2\theta+\frac{\sqrt{7}}{4}\left(A_x+A_y\right)\sin 2\theta\right]^2+\frac{49}{4}{A_z}^2\sin^2 2\theta\right\}$$

$$\varepsilon_{-,9}^{(02)}=\frac{|\langle +,1|H_{\mathrm{hfs}}|-,9\rangle|^2+|\langle +,10|H_{\mathrm{hfs}}|-,9\rangle|^2}{-2\Delta}$$

$$=-\frac{1}{2\Delta}\left\{\frac{49}{4}{A_z}^2\sin^2 2\theta+\left[\frac{\sqrt{21}}{4}\left(A_x-A_y\right)\cos 2\theta+\frac{\sqrt{7}}{4}\left(A_x+A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{-,10}^{(02)}=\frac{|\langle +,2|H_{\mathrm{hfs}}|-,10\rangle|^2+|\langle +,9|H_{\mathrm{hfs}}|-,10\rangle|^2+|\langle +,11|H_{\mathrm{hfs}}|-,10\rangle|^2}{-2\Delta}$$

$$=-\frac{1}{2\Delta}\left\{\frac{25}{4}{A_z}^2\sin^2 2\theta+\left[\frac{\sqrt{21}}{4}\left(A_x+A_y\right)\cos 2\theta+\frac{\sqrt{7}}{4}\left(A_x-A_y\right)\sin 2\theta\right]^2\right.$$

$$\left.+\left[\frac{3}{2}\left(A_x-A_y\right)\cos 2\theta+\frac{\sqrt{3}}{2}\left(A_x+A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{-,11}^{(02)} = \frac{|\langle +,3|H_{\mathrm{hfs}}|-,11\rangle|^2 + |\langle +,10|H_{\mathrm{hfs}}|-,11\rangle|^2 + |\langle +,12|H_{\mathrm{hfs}}|-,11\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\frac{9}{4}{A_z}^2 \sin^2 2\theta + \left[\frac{3}{2}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(A_x - A_y\right)\sin 2\theta\right]^2 \right.$$
$$\left. + \left[\frac{3\sqrt{5}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{15}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{-,12}^{(02)} = \frac{|\langle +,4|H_{\mathrm{hfs}}|-,12\rangle|^2 + |\langle +,11|H_{\mathrm{hfs}}|-,12\rangle|^2 + |\langle +,13|H_{\mathrm{hfs}}|-,12\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\frac{{A_z}^2}{4}\sin^2 2\theta + \left[\frac{3\sqrt{5}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{15}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2 \right.$$
$$\left. + \left[\sqrt{3}\left(A_x - A_y\right)\cos 2\theta + \left(A_x + A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{-,13}^{(02)} = \frac{|\langle +,5|H_{\mathrm{hfs}}|-,13\rangle|^2 + |\langle +,12|H_{\mathrm{hfs}}|-,13\rangle|^2 + |\langle +,14|H_{\mathrm{hfs}}|-,13\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\frac{{A_z}^2}{4}\sin^2 2\theta + \left[\sqrt{3}\left(A_x + A_y\right)\cos 2\theta + \left(A_x - A_y\right)\sin 2\theta\right]^2 \right.$$
$$\left. + \left[\frac{3\sqrt{5}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{15}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{-,15}^{(02)} = \frac{|\langle +,6|H_{\mathrm{hfs}}|-,14\rangle|^2 + |\langle +,13|H_{\mathrm{hfs}}|-,14\rangle|^2 + |\langle +,15|H_{\mathrm{hfs}}|-,14\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\frac{9}{4}{A_z}^2 \sin^2 2\theta + \left[\frac{3\sqrt{5}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{15}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2 \right.$$
$$\left. + \left[\frac{3}{2}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(A_x + A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{-,15}^{(02)} = \frac{|\langle +,7|H_{\mathrm{hfs}}|-,15\rangle|^2 + |\langle +,14|H_{\mathrm{hfs}}|-,15\rangle|^2 + |\langle +,16|H_{\mathrm{hfs}}|-,15\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\frac{25}{4}{A_z}^2 \sin^2 2\theta + \left[\frac{3}{2}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{3}}{2}\left(A_x - A_y\right)\sin 2\theta\right]^2 \right.$$
$$\left. + \left[\frac{\sqrt{21}}{4}\left(A_x - A_y\right)\cos 2\theta + \frac{\sqrt{7}}{4}\left(A_x + A_y\right)\sin 2\theta\right]^2\right\}$$

$$\varepsilon_{-,16}^{(02)} = \frac{|\langle +,8|H_{\mathrm{hfs}}|-,16\rangle|^2 + |\langle +,15|H_{\mathrm{hfs}}|-,16\rangle|^2}{-2\Delta}$$

$$= -\frac{1}{2\Delta}\left\{\frac{49}{4}{A_z}^2 \sin^2 2\theta + \left[\frac{\sqrt{21}}{4}\left(A_x + A_y\right)\cos 2\theta + \frac{\sqrt{7}}{4}\left(A_x - A_y\right)\sin 2\theta\right]^2\right\}$$

The first- and second-order corrections for the energy of the electron-Zeeman Hamiltonian are the same as for the $I = 1/2$ case. In order to obtain the cross terms, let us remind the non-diagonal elements for the electron-Zeeman Hamiltonian.

$$\begin{aligned}\langle +,1|H_{\mathrm{eZ}}|-,9\rangle &= \langle -,9|H_{\mathrm{eZ}}|+,1\rangle = \langle +,2|H_{\mathrm{eZ}}|-,10\rangle = \langle -,10|H_{\mathrm{eZ}}|+,2\rangle = \langle +,3|H_{\mathrm{eZ}}|-,11\rangle \\ &= \langle -,11|H_{\mathrm{eZ}}|+,3\rangle = \langle +,4|H_{\mathrm{eZ}}|-,12\rangle = \langle -,12|H_{\mathrm{eZ}}|+,4\rangle = \langle +,5|H_{\mathrm{eZ}}|-,13\rangle \\ &= \langle -,13|H_{\mathrm{eZ}}|+,5\rangle = \langle +,6|H_{\mathrm{eZ}}|-,14\rangle = \langle -,14|H_{\mathrm{eZ}}|+,6\rangle = \langle +,7|H_{\mathrm{eZ}}|-,15\rangle \\ &= \langle -,15|H_{\mathrm{eZ}}|+,7\rangle = \langle +,8|H_{\mathrm{eZ}}|-,16\rangle = \langle -,16|H_{\mathrm{eZ}}|+,8\rangle = -g_z\beta B\sin 2\theta\end{aligned}$$

$$\begin{aligned}\langle -,1|H_{\mathrm{eZ}}|+,9\rangle &= \langle +,9|H_{\mathrm{eZ}}|-,1\rangle = \langle -,2|H_{\mathrm{eZ}}|+,10\rangle = \langle +,10|H_{\mathrm{eZ}}|-,2\rangle = \langle -,3|H_{\mathrm{eZ}}|+,11\rangle \\ &= \langle +,11|H_{\mathrm{eZ}}|-,3\rangle = \langle -,4|H_{\mathrm{eZ}}|+,12\rangle = \langle +,12|H_{\mathrm{eZ}}|-,4\rangle = \langle -,5|H_{\mathrm{eZ}}|+,13\rangle \\ &= \langle +,13|H_{\mathrm{eZ}}|-,5\rangle = \langle -,6|H_{\mathrm{eZ}}|+,14\rangle = \langle +,14|H_{\mathrm{eZ}}|-,6\rangle = \langle -,7|H_{\mathrm{eZ}}|+,15\rangle \\ &= \langle +,15|H_{\mathrm{eZ}}|-,7\rangle = \langle -,8|H_{\mathrm{eZ}}|+,16\rangle = \langle +,16|H_{\mathrm{eZ}}|-,8\rangle = g_z\beta B\sin 2\theta\end{aligned}$$

The cross terms can be calculated as follows:

$$\varepsilon_{+,1}^{(11)} = \frac{\langle +,1|H_{\mathrm{eZ}}|-,9\rangle\langle -,9|H_{\mathrm{hfs}}|+,1\rangle + \langle +,1|H_{\mathrm{hfs}}|-,9\rangle\langle -,9|H_{\mathrm{eZ}}|+,1\rangle}{\varepsilon_+^{(0)} - \varepsilon_-^{(0)}} = \frac{49g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,2}^{(11)} = \frac{\langle +,2|H_{\mathrm{eZ}}|-,10\rangle\langle -,10|H_{\mathrm{hfs}}|+,2\rangle + \langle +,2|H_{\mathrm{hfs}}|-,10\rangle\langle -,10|H_{\mathrm{eZ}}|+,2\rangle}{\varepsilon_+^{(0)} - \varepsilon_-^{(0)}} = \frac{25g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,3}^{(11)} = \frac{\langle +,3|H_{\mathrm{eZ}}|-,11\rangle\langle -,11|H_{\mathrm{hfs}}|+,3\rangle + \langle +,3|H_{\mathrm{hfs}}|-,11\rangle\langle -,11|H_{\mathrm{eZ}}|+,3\rangle}{\varepsilon_+^{(0)} - \varepsilon_-^{(0)}} = \frac{9g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,4}^{(11)} = \frac{\langle +,4|H_{\mathrm{eZ}}|-,12\rangle\langle -,12|H_{\mathrm{hfs}}|+,4\rangle + \langle +,4|H_{\mathrm{hfs}}|-,12\rangle\langle -,12|H_{\mathrm{eZ}}|+,4\rangle}{\varepsilon_+^{(0)} - \varepsilon_-^{(0)}} = \frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,5}^{(11)} = \frac{\langle +,5|H_{\mathrm{eZ}}|-,13\rangle\langle -,13|H_{\mathrm{hfs}}|+,5\rangle + \langle +,5|H_{\mathrm{hfs}}|-,13\rangle\langle -,13|H_{\mathrm{eZ}}|+,5\rangle}{\varepsilon_+^{(0)} - \varepsilon_-^{(0)}} = -\frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,6}^{(11)} = \frac{\langle +,6|H_{\mathrm{eZ}}|-,14\rangle\langle -,14|H_{\mathrm{hfs}}|+,6\rangle + \langle +,6|H_{\mathrm{hfs}}|-,14\rangle\langle -,14|H_{\mathrm{eZ}}|+,6\rangle}{\varepsilon_+^{(0)} - \varepsilon_-^{(0)}} = -\frac{9g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,7}^{(11)} = \frac{\langle +,7|H_{\mathrm{eZ}}|-,15\rangle\langle -,15|H_{\mathrm{hfs}}|+,7\rangle + \langle +,7|H_{\mathrm{hfs}}|-,15\rangle\langle -,15|H_{\mathrm{eZ}}|+,7\rangle}{\varepsilon_+^{(0)} - \varepsilon_-^{(0)}} = -\frac{25g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,8}^{(11)} = \frac{\langle +,8|H_{\mathrm{eZ}}|-,16\rangle\langle -,16|H_{\mathrm{hfs}}|+,8\rangle + \langle +,8|H_{\mathrm{hfs}}|-,16\rangle\langle -,16|H_{\mathrm{eZ}}|+,8\rangle}{\varepsilon_+^{(0)} - \varepsilon_-^{(0)}} = -\frac{49g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,1}^{(11)} = \frac{\langle -,1|H_{\mathrm{eZ}}|+,9\rangle\langle +,9|H_{\mathrm{hfs}}|-,1\rangle + \langle -,1|H_{\mathrm{hfs}}|+,9\rangle\langle +,9|H_{\mathrm{eZ}}|-,1\rangle}{\varepsilon_-^{(0)} - \varepsilon_+^{(0)}} = -\frac{49g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,2}^{(11)} = \frac{\langle -,2|H_{\mathrm{eZ}}|+,10\rangle\langle +,10|H_{\mathrm{hfs}}|-,2\rangle + \langle -,2|H_{\mathrm{hfs}}|+,10\rangle\langle +,10|H_{\mathrm{eZ}}|-,2\rangle}{\varepsilon_-^{(0)} - \varepsilon_+^{(0)}} = -\frac{25g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,3}^{(11)} = \frac{\langle -,3|H_{\mathrm{eZ}}|+,11\rangle\langle +,11|H_{\mathrm{hfs}}|-,3\rangle + \langle -,3|H_{\mathrm{hfs}}|+,11\rangle\langle +,11|H_{\mathrm{eZ}}|-,3\rangle}{\varepsilon_-^{(0)} - \varepsilon_+^{(0)}} = -\frac{9g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,4}^{(11)} = \frac{\langle -,4|H_{\mathrm{eZ}}|+,12\rangle\langle +,12|H_{\mathrm{hfs}}|-,4\rangle + \langle -,4|H_{\mathrm{hfs}}|+,12\rangle\langle +,12|H_{\mathrm{eZ}}|-,4\rangle}{\varepsilon_-^{(0)} - \varepsilon_+^{(0)}} = -\frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,5}^{(11)} = \frac{\langle -,5|H_{\mathrm{eZ}}|+,13\rangle\langle +,13|H_{\mathrm{hfs}}|-,5\rangle + \langle -,5|H_{\mathrm{hfs}}|+,13\rangle\langle +,13|H_{\mathrm{eZ}}|-,5\rangle}{\varepsilon_-^{(0)} - \varepsilon_+^{(0)}} = \frac{g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,6}^{(11)} = \frac{\langle -,6|H_{\mathrm{eZ}}|+,14\rangle\langle +,14|H_{\mathrm{hfs}}|-,6\rangle + \langle -,6|H_{\mathrm{hfs}}|+,14\rangle\langle +,14|H_{\mathrm{eZ}}|-,6\rangle}{\varepsilon_-^{(0)} - \varepsilon_+^{(0)}} = \frac{9g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,7}^{(11)} = \frac{\langle -,7|H_{\mathrm{eZ}}|+,15\rangle\langle +,15|H_{\mathrm{hfs}}|-,7\rangle + \langle -,7|H_{\mathrm{hfs}}|+,15\rangle\langle +,15|H_{\mathrm{eZ}}|-,7\rangle}{\varepsilon_-^{(0)} - \varepsilon_+^{(0)}} = \frac{25g_z\beta BA_z\sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,8}^{(11)} = \frac{\langle -,8|H_{\mathrm{eZ}}|+,16\rangle\langle +,16|H_{\mathrm{hfs}}|-,8\rangle + \langle -,8|H_{\mathrm{hfs}}|+,16\rangle\langle +,16|H_{\mathrm{eZ}}|-,8\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = \frac{49 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,9}^{(11)} = \frac{\langle -,9|H_{\mathrm{eZ}}|+,1\rangle\langle +,1|H_{\mathrm{hfs}}|-,9\rangle + \langle -,9|H_{\mathrm{hfs}}|+,1\rangle\langle +,1|H_{\mathrm{eZ}}|-,9\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = \frac{49 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,10}^{(11)} = \frac{\langle -,10|H_{\mathrm{eZ}}|+,2\rangle\langle +,2|H_{\mathrm{hfs}}|-,10\rangle + \langle -,10|H_{\mathrm{hfs}}|+,2\rangle\langle +,2|H_{\mathrm{eZ}}|-,10\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = \frac{25 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,11}^{(11)} = \frac{\langle -,11|H_{\mathrm{eZ}}|+,3\rangle\langle +,3|H_{\mathrm{hfs}}|-,11\rangle + \langle -,11|H_{\mathrm{hfs}}|+,3\rangle\langle +,3|H_{\mathrm{eZ}}|-,10\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = \frac{9 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,12}^{(11)} = \frac{\langle -,12|H_{\mathrm{eZ}}|+,4\rangle\langle +,4|H_{\mathrm{hfs}}|-,12\rangle + \langle -,12|H_{\mathrm{hfs}}|+,4\rangle\langle +,4|H_{\mathrm{eZ}}|-,12\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = \frac{g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,13}^{(11)} = \frac{\langle -,13|H_{\mathrm{eZ}}|+,5\rangle\langle +,5|H_{\mathrm{hfs}}|-,13\rangle + \langle -,13|H_{\mathrm{hfs}}|+,5\rangle\langle +,5|H_{\mathrm{eZ}}|-,13\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = -\frac{g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,14}^{(11)} = \frac{\langle -,14|H_{\mathrm{eZ}}|+,6\rangle\langle +,6|H_{\mathrm{hfs}}|-,14\rangle + \langle -,14|H_{\mathrm{hfs}}|+,6\rangle\langle +,6|H_{\mathrm{eZ}}|-,14\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = -\frac{9 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,10}^{(11)} = \frac{\langle -,15|H_{\mathrm{eZ}}|+,7\rangle\langle +,7|H_{\mathrm{hfs}}|-,15\rangle + \langle -,15|H_{\mathrm{hfs}}|+,7\rangle\langle +,7|H_{\mathrm{eZ}}|-,15\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = -\frac{25 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{-,16}^{(11)} = \frac{\langle -,16|H_{\mathrm{eZ}}|+,8\rangle\langle +,8|H_{\mathrm{hfs}}|-,16\rangle + \langle -,16|H_{\mathrm{hfs}}|+,8\rangle\langle +,8|H_{\mathrm{eZ}}|-,16\rangle}{\varepsilon_{-}^{(0)} - \varepsilon_{+}^{(0)}} = -\frac{49 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,9}^{(11)} = \frac{\langle +,9|H_{\mathrm{eZ}}|-,1\rangle\langle -,1|H_{\mathrm{hfs}}|+,9\rangle + \langle +,9|H_{\mathrm{hfs}}|-,1\rangle\langle -,1|H_{\mathrm{eZ}}|+,9\rangle}{\varepsilon_{+}^{(0)} - \varepsilon_{-}^{(0)}} = -\frac{49 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,10}^{(11)} = \frac{\langle +,10|H_{\mathrm{eZ}}|-,2\rangle\langle -,2|H_{\mathrm{hfs}}|+,10\rangle + \langle +,10|H_{\mathrm{hfs}}|-,2\rangle\langle -,2|H_{\mathrm{eZ}}|+,10\rangle}{\varepsilon_{+}^{(0)} - \varepsilon_{-}^{(0)}} = -\frac{25 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,11}^{(11)} = \frac{\langle +,11|H_{\mathrm{eZ}}|-,3\rangle\langle -,3|H_{\mathrm{hfs}}|+,11\rangle + \langle +,11|H_{\mathrm{hfs}}|-,3\rangle\langle -,3|H_{\mathrm{eZ}}|+,11\rangle}{\varepsilon_{+}^{(0)} - \varepsilon_{-}^{(0)}} = -\frac{9 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,12}^{(11)} = \frac{\langle +,12|H_{\mathrm{eZ}}|-,4\rangle\langle -,4|H_{\mathrm{hfs}}|+,12\rangle + \langle +,12|H_{\mathrm{hfs}}|-,4\rangle\langle -,4|H_{\mathrm{eZ}}|+,12\rangle}{\varepsilon_{+}^{(0)} - \varepsilon_{-}^{(0)}} = -\frac{g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,13}^{(11)} = \frac{\langle +,13|H_{\mathrm{eZ}}|-,5\rangle\langle -,5|H_{\mathrm{hfs}}|+,13\rangle + \langle +,13|H_{\mathrm{hfs}}|-,5\rangle\langle -,5|H_{\mathrm{eZ}}|+,13\rangle}{\varepsilon_{+}^{(0)} - \varepsilon_{-}^{(0)}} = \frac{g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,14}^{(11)} = \frac{\langle +,14|H_{\mathrm{eZ}}|-,6\rangle\langle -,6|H_{\mathrm{hfs}}|+,14\rangle + \langle +,14|H_{\mathrm{hfs}}|-,6\rangle\langle -,6|H_{\mathrm{eZ}}|+,14\rangle}{\varepsilon_{+}^{(0)} - \varepsilon_{-}^{(0)}} = \frac{9 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,15}^{(11)} = \frac{\langle +,15|H_{\mathrm{eZ}}|-,7\rangle\langle -,7|H_{\mathrm{hfs}}|+,15\rangle + \langle +,15|H_{\mathrm{hfs}}|-,7\rangle\langle -,7|H_{\mathrm{eZ}}|+,15\rangle}{\varepsilon_{+}^{(0)} - \varepsilon_{-}^{(0)}} = \frac{25 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

$$\varepsilon_{+,16}^{(11)} = \frac{\langle +,16|H_{\mathrm{eZ}}|-,8\rangle\langle -,8|H_{\mathrm{hfs}}|+,16\rangle + \langle +,16|H_{\mathrm{hfs}}|-,8\rangle\langle -,8|H_{\mathrm{eZ}}|+,16\rangle}{\varepsilon_{+}^{(0)} - \varepsilon_{-}^{(0)}} = \frac{49 g_z \beta B A_z \sin^2 2\theta}{2\Delta}$$

Thus, the perturbed energies for the case with $I = 7/2$ for the spin quartet state were explicitly obtained in the second order. To our knowledge the analytical expressions above with $I = 7/2$ and $S = 3/2$ for the energies in terms of the Zeeman perturbation theory are for the first time given in this work, which are extremely accurate.

b) Perturbed energies in the case that the ZFS, g- and A-tensors are non-collinear

In this subsection, we consider the general case in which all the three magnetic tensors are non-collinear for the spin quartet state. An approach adopted here is not a Zeeman perturbation treatment, but the rank-2 ZFS and electron-Zeeman Hamiltonian are taken as the non-perturbed term and the hyperfine structure interaction as the perturbed one. Thus, first the non-perturbed Hamiltonian gives a 4 × 4 matrix which is expanded in terms of the spin functions and in which the presence of the electron Zeeman terms destroys the spin conjugation symmetry, and thus, first we exactly solve the corresponding quartic secular equation.

The non-perturbed Hamiltonian for the arbitrary direction of the magnetic field can be represented as

$$H = D\left[{S_z}^2 - \frac{1}{3}S(S+1)\right] + E\left({S_x}^2 - {S_y}^2\right) + \left(S_x g_{xx} + S_y g_{yx} + S_z g_{zx}\right)\beta B_x \\ + \left(S_x g_{xy} + S_y g_{yy} + S_z g_{zy}\right)\beta B_y + \left(S_x g_{xz} + S_y g_{yz} + S_z g_{zz}\right)\beta B_z$$

where B_x, B_y and B_z are the elements of the vector $\boldsymbol{B}$. In the polar coordinate-axis system, $B_x = B\sin\theta\cos\varphi$, $B_y = B\sin\theta\sin\varphi$ and $B_z = B\cos\theta$ where $B = |\boldsymbol{B}|$ and θ and φ are the polar and azimuthal angle, respectively. Note that the *x, y* and *z* axes denote the principal axes of the ZFS tensor. In the following discussion, we set $g'_{ij} = \beta g_{ij} B_j$ for simplicity. The Hamiltonian can be written in the matrix representation

$$H = \begin{pmatrix} D + \frac{3}{2}(g'_{zx} + g'_{zy} + g'_{zz}) & \frac{\sqrt{3}}{2}(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz}) & \sqrt{3}E & 0 \\ \frac{\sqrt{3}}{2}(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz}) & -D + \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) & g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz} & \sqrt{3}E \\ \sqrt{3}E & g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz} & -D - \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) & \frac{\sqrt{3}}{2}(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz}) \\ 0 & \sqrt{3}E & \frac{\sqrt{3}}{2}(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz}) & D - \frac{3}{2}(g'_{zx} + g'_{zy} + g'_{zz}) \end{pmatrix}$$

The secular quartic equation of the Hamiltonian matrix is given as

$$x^4 + p_0 x^2 + q_0 x + r_0 = 0$$

with

$$p_0 = -2(D^2 + 3E^2) - \frac{5}{2}{g'_{xx}}^2 - 5g'_{xx}g'_{xy} - \frac{5}{2}{g'_{xy}}^2 - 5g'_{xx}g'_{xz} - 5g'_{xy}g'_{xz} - \frac{5}{2}{g'_{xz}}^2 - \frac{5}{2}{g'_{yx}}^2 - 5g'_{yx}g'_{yy} \\ - \frac{5}{2}{g'_{yy}}^2 - 5g'_{yx}g'_{yz} - 5g'_{yy}g'_{yz} - \frac{5}{2}{g'_{yz}}^2 - \frac{5}{2}{g'_{zx}}^2 - 5g'_{zx}g'_{zy} - \frac{5}{2}{g'_{zy}}^2 - 5g'_{zx}g'_{zz} \\ - 5g'_{zy}g'_{zz} - \frac{5}{2}{g'_{zz}}^2$$

$$q_0 = 2(D - 3E){g'_{xx}}^2 + 4(D - 3E)g'_{xx}g'_{xy} + 2(D - 3E){g'_{xy}}^2 + 4(D - 3E)g'_{xx}g'_{xz} + 4(D - 3E)g'_{xy}g'_{xz} \\ + 2(D - 3E){g'_{xz}}^2 + 2(D + 3E){g'_{yx}}^2 + 4(D + 3E)g'_{yx}g'_{yy} + 2(D + 3E){g'_{yy}}^2 \\ + 4(D + 3E)g'_{yx}g'_{yz} + 4(D + 3E)g'_{yy}g'_{yz} + 2(D + 3E){g'_{yz}}^2 - 4D{g'_{zx}}^2 - 8Dg'_{zx}g'_{zy} \\ - 4D{g'_{zy}}^2 - 8Dg'_{zx}g'_{zz} - 8Dg'_{zy}g'_{zz} - 4D{g'_{zz}}^2$$

$$r_0 = (D^2+3E^2)^2 + \frac{1}{2}(D^2+12DE-9E^2){g'_{xx}}^2 + \frac{9}{16}{g'_{xx}}^4 + (D^2+12DE-9E^2)g'_{xx}g'_{xy} + \frac{9}{4}{g'_{xx}}^3 g'_{xy}$$

$$+ \frac{1}{2}(D^2+12DE-9E^2){g'_{xy}}^2 + \frac{27}{8}{g'_{xx}}^2{g'_{xy}}^2 + \frac{9}{4}g'_{xx}{g'_{xy}}^3 + \frac{9}{16}{g'_{xy}}^4$$

$$+ (D^2+12DE-9E^2)g'_{xx}g'_{xz} + \frac{9}{4}{g'_{xx}}^3 g'_{xz} + (D^2+12DE-9E^2)g'_{xy}g'_{xz}$$

$$+ \frac{27}{4}{g'_{xx}}^2 g'_{xy}g'_{xz} + \frac{27}{4}g'_{xx}{g'_{xy}}^2 g'_{xz} + \frac{9}{4}{g'_{xy}}^3 g'_{xz} + \frac{1}{2}(D^2+12DE-9E^2){g'_{xz}}^2$$

$$+ \frac{27}{8}{g'_{xx}}^2{g'_{xz}}^2 + \frac{27}{4}g'_{xx}g'_{xy}{g'_{xz}}^2 + \frac{27}{8}{g'_{xy}}^2{g'_{xz}}^2 + \frac{9}{4}g'_{xx}{g'_{xz}}^3 + \frac{9}{4}g'_{xy}{g'_{xz}}^3 + \frac{9}{16}{g'_{xz}}^4$$

$$+ \frac{1}{2}(D^2-12DE-9E^2){g'_{yx}}^2 + \frac{9}{8}{g'_{xx}}^2{g'_{yx}}^2 + \frac{9}{4}g'_{xx}g'_{xy}{g'_{yx}}^2 + \frac{9}{8}{g'_{xy}}^2{g'_{yx}}^2$$

$$+ \frac{9}{4}g'_{xx}g'_{xz}{g'_{yx}}^2 + \frac{9}{4}g'_{xy}g'_{xz}{g'_{yx}}^2 + \frac{9}{8}{g'_{xz}}^2{g'_{yx}}^2 + \frac{9}{16}{g'_{yx}}^4$$

$$+ (D^2-12DE-9E^2)g'_{yx}g'_{yy} + \frac{9}{4}{g'_{xx}}^2 g'_{yx}g'_{yy} + \frac{9}{2}g'_{xx}g'_{xy}g'_{yx}g'_{yy} + \frac{9}{4}{g'_{xy}}^2 g'_{yx}g'_{yy}$$

$$+ \frac{9}{2}g'_{xx}g'_{xz}g'_{yx}g'_{yy} + \frac{9}{2}g'_{xy}g'_{xz}g'_{yx}g'_{yy} + \frac{9}{4}{g'_{xz}}^2 g'_{yx}g'_{yy} + \frac{9}{4}{g'_{yx}}^3 g'_{yy}$$

$$+ \frac{1}{2}(D^2-12DE-9E^2){g'_{yy}}^2 + \frac{9}{8}{g'_{xx}}^2{g'_{yy}}^2 + \frac{9}{4}g'_{xx}g'_{xy}{g'_{yy}}^2 + \frac{9}{8}{g'_{xy}}^2{g'_{yy}}^2$$

$$+ \frac{9}{4}g'_{xx}g'_{xz}{g'_{yy}}^2 + \frac{9}{4}g'_{xy}g'_{xz}{g'_{yy}}^2 + \frac{9}{8}{g'_{xz}}^2{g'_{yy}}^2 + \frac{27}{8}{g'_{yx}}^2{g'_{yy}}^2 + \frac{9}{4}g'_{yx}{g'_{yy}}^3 + \frac{9}{16}{g'_{yy}}^4$$

$$+ (D^2-12DE-9E^2)g'_{yx}g'_{yz} + \frac{9}{4}{g'_{xx}}^2 g'_{yx}g'_{yz} + \frac{9}{2}g'_{xx}g'_{xy}g'_{yx}g'_{yz} + \frac{9}{4}{g'_{xy}}^2 g'_{yx}g'_{yz}$$

$$+ \frac{9}{2}g'_{xx}g'_{xz}g'_{yx}g'_{yz} + \frac{9}{2}g'_{xy}g'_{xz}g'_{yx}g'_{yz} + \frac{9}{4}{g'_{xz}}^2 g'_{yx}g'_{yz} + \frac{9}{4}{g'_{yx}}^3 g'_{yz}$$

$$+ (D^2-12DE-9E^2)g'_{yy}g'_{yz} + \frac{9}{4}{g'_{xx}}^2 g'_{yy}g'_{yz} + \frac{9}{2}g'_{xx}g'_{xy}g'_{yy}g'_{yz} + \frac{9}{4}{g'_{xy}}^2 g'_{yy}g'_{yz}$$

$$+ \frac{9}{2}g'_{xx}g'_{xz}g'_{yy}g'_{yz} + \frac{9}{2}g'_{xy}g'_{xz}g'_{yy}g'_{yz} + \frac{9}{4}{g'_{xz}}^2 g'_{yy}g'_{yz} + \frac{27}{4}{g'_{yx}}^2 g'_{yy}g'_{yz}$$

$$+ \frac{27}{4}g'_{yx}{g'_{yy}}^2 g'_{yz} + \frac{9}{4}{g'_{yy}}^3 g'_{yz} + \frac{1}{2}(D^2-12DE-9E^2){g'_{yz}}^2 + \frac{9}{8}{g'_{xx}}^2{g'_{yz}}^2$$

$$+ \frac{9}{4}g'_{xx}g'_{xy}{g'_{yz}}^2 + \frac{9}{8}{g'_{xy}}^2{g'_{yz}}^2 + \frac{9}{4}g'_{xx}g'_{xz}{g'_{yz}}^2 + \frac{9}{4}g'_{xy}g'_{xz}{g'_{yz}}^2 + \frac{9}{8}{g'_{xz}}^2{g'_{yz}}^2$$

$$+ \frac{27}{8}{g'_{yx}}^2{g'_{yz}}^2 + \frac{27}{4}g'_{yx}g'_{yy}{g'_{yz}}^2 + \frac{27}{8}{g'_{yy}}^2{g'_{yz}}^2 + \frac{9}{4}g'_{yx}{g'_{yz}}^3 + \frac{9}{4}g'_{yy}{g'_{yz}}^3 + \frac{9}{16}{g'_{yz}}^4$$

$$- \frac{1}{2}(5D^2-9E^2){g'_{zx}}^2 + \frac{9}{8}{g'_{xx}}^2{g'_{zx}}^2 + \frac{9}{4}g'_{xx}g'_{xy}{g'_{zx}}^2 + \frac{9}{8}{g'_{xy}}^2{g'_{zx}}^2 + \frac{9}{4}g'_{xx}g'_{xz}{g'_{zx}}^2$$

$$+\frac{9}{4}g'_{xy}g'_{xz}{g'_{zx}}^2+\frac{9}{8}{g'_{xz}}^2{g'_{zx}}^2+\frac{9}{8}{g'_{yx}}^2{g'_{zx}}^2+\frac{9}{4}g'_{yx}g'_{yy}{g'_{zx}}^2+\frac{9}{8}{g'_{yy}}^2{g'_{zx}}^2$$

$$+\frac{9}{4}g'_{yx}g'_{yz}{g'_{zx}}^2+\frac{9}{4}g'_{yy}g'_{yz}{g'_{zx}}^2+\frac{9}{8}{g'_{yz}}^2{g'_{zx}}^2+\frac{9}{16}{g'_{zx}}^4-(5D^2-9E^2)g'_{zx}g'_{zy}$$

$$+\frac{9}{4}{g'_{xx}}^2g'_{zx}g'_{zy}+\frac{9}{2}g'_{xx}g'_{xy}g'_{zx}g'_{zy}+\frac{9}{4}{g'_{xy}}^2g'_{zx}g'_{zy}+\frac{9}{2}g'_{xx}g'_{xz}g'_{zx}g'_{zy}$$

$$+\frac{9}{2}g'_{xy}g'_{xz}g'_{zx}g'_{zy}+\frac{9}{4}{g'_{xz}}^2g'_{zx}g'_{zy}+\frac{9}{4}{g'_{yx}}^2g'_{zx}g'_{zy}+\frac{9}{2}g'_{yx}g'_{yy}g'_{zx}g'_{zy}+\frac{9}{4}{g'_{yy}}^2g'_{zx}g'_{zy}$$

$$+\frac{9}{2}g'_{yx}g'_{yz}g'_{zx}g'_{zy}+\frac{9}{2}g'_{yy}g'_{yz}g'_{zx}g'_{zy}+\frac{9}{4}{g'_{yz}}^2g'_{zx}g'_{zy}+\frac{9}{4}{g'_{zx}}^3g'_{zy}$$

$$-\frac{1}{2}(5D^2-9E^2){g'_{zy}}^2+\frac{9}{8}{g'_{xx}}^2{g'_{zy}}^2+\frac{9}{4}g'_{xx}g'_{xy}{g'_{zy}}^2+\frac{9}{8}{g'_{xy}}^2{g'_{zy}}^2+\frac{9}{4}g'_{xx}g'_{xz}{g'_{zy}}^2$$

$$+\frac{9}{4}g'_{xy}g'_{xz}{g'_{zy}}^2+\frac{9}{8}{g'_{xz}}^2{g'_{zy}}^2+\frac{9}{8}{g'_{yx}}^2{g'_{zy}}^2+\frac{9}{4}g'_{yx}g'_{yy}{g'_{zy}}^2+\frac{9}{8}{g'_{yy}}^2{g'_{zy}}^2$$

$$+\frac{9}{4}g'_{yx}g'_{yz}{g'_{zy}}^2+\frac{9}{4}g'_{yy}g'_{yz}{g'_{zy}}^2+\frac{9}{8}{g'_{yz}}^2{g'_{zy}}^2+\frac{27}{8}{g'_{zx}}^2{g'_{zy}}^2+\frac{9}{4}g'_{zx}{g'_{zy}}^3+\frac{9}{16}{g'_{zy}}^4$$

$$-(5D^2-9E^2)g'_{zx}g'_{zz}+\frac{9}{4}{g'_{xx}}^2g'_{zx}g'_{zz}+\frac{9}{2}g'_{xx}g'_{xy}g'_{zx}g'_{zz}+\frac{9}{4}{g'_{xy}}^2g'_{zx}g'_{zz}$$

$$+\frac{9}{2}g'_{xx}g'_{xz}g'_{zx}g'_{zz}+\frac{9}{2}g'_{xy}g'_{xz}g'_{zx}g'_{zz}+\frac{9}{4}{g'_{xz}}^2g'_{zx}g'_{zz}+\frac{9}{4}{g'_{yx}}^2g'_{zx}g'_{zz}+\frac{9}{2}g'_{yx}g'_{yy}g'_{zx}g'_{zz}$$

$$+\frac{9}{4}{g'_{yy}}^2g'_{zx}g'_{zz}+\frac{9}{2}g'_{yx}g'_{yz}g'_{zx}g'_{zz}+\frac{9}{2}g'_{yy}g'_{yz}g'_{zx}g'_{zz}+\frac{9}{4}{g'_{yz}}^2g'_{zx}g'_{zz}+\frac{9}{4}{g'_{zx}}^3g'_{zz}$$

$$-(5D^2-9E^2)g'_{zy}g'_{zz}+\frac{9}{4}{g'_{xx}}^2g'_{zy}g'_{zz}+\frac{9}{2}g'_{xx}g'_{xy}g'_{zy}g'_{zz}+\frac{9}{4}{g'_{xy}}^2g'_{zy}g'_{zz}$$

$$+\frac{9}{2}g'_{xx}g'_{xz}g'_{zy}g'_{zz}+\frac{9}{2}g'_{xy}g'_{xz}g'_{zy}g'_{zz}+\frac{9}{4}{g'_{xz}}^2g'_{zy}g'_{zz}+\frac{9}{4}{g'_{yx}}^2g'_{zy}g'_{zz}$$

$$+\frac{9}{2}g'_{yx}g'_{yy}g'_{zy}g'_{zz}+\frac{9}{4}{g'_{yy}}^2g'_{zy}g'_{zz}+\frac{9}{2}g'_{yx}g'_{yz}g'_{zy}g'_{zz}+\frac{9}{2}g'_{yy}g'_{yz}g'_{zy}g'_{zz}$$

$$+\frac{9}{4}{g'_{yz}}^2g'_{zy}g'_{zz}+\frac{27}{4}{g'_{zx}}^2g'_{zy}g'_{zz}+\frac{27}{4}g'_{zx}{g'_{zy}}^2g'_{zz}+\frac{9}{4}{g'_{zy}}^3g'_{zz}-\frac{1}{2}(5D^2-9E^2){g'_{zz}}^2$$

$$+\frac{9}{8}{g'_{xx}}^2{g'_{zz}}^2+\frac{9}{4}g'_{xx}g'_{xy}{g'_{zz}}^2+\frac{9}{8}{g'_{xy}}^2{g'_{zz}}^2+\frac{9}{4}g'_{xx}g'_{xz}{g'_{zz}}^2+\frac{9}{4}g'_{xy}g'_{xz}{g'_{zz}}^2$$

$$+\frac{9}{8}{g'_{xz}}^2{g'_{zz}}^2+\frac{9}{8}{g'_{yx}}^2{g'_{zz}}^2+\frac{9}{4}g'_{yx}g'_{yy}{g'_{zz}}^2+\frac{9}{8}{g'_{yy}}^2{g'_{zz}}^2+\frac{9}{4}g'_{yx}g'_{yz}{g'_{zz}}^2$$

$$+\frac{9}{4}g'_{yy}g'_{yz}{g'_{zz}}^2+\frac{9}{8}{g'_{yz}}^2{g'_{zz}}^2+\frac{27}{8}{g'_{zx}}^2{g'_{zz}}^2+\frac{27}{4}g'_{zx}g'_{zy}{g'_{zz}}^2+\frac{27}{8}{g'_{zy}}^2{g'_{zz}}^2$$

$$+\frac{9}{4}g'_{zx}{g'_{zz}}^3+\frac{9}{4}g'_{zy}{g'_{zz}}^3+\frac{9}{16}{g'_{zz}}^4$$

The coefficients of the quartic equation are complex as expected from the non-collinearity between the ZFS and **g**-tensors. The exact analytical energy eigenvalues and the corresponding spin substates for $D < 0$ are given in the following:

$$\varepsilon_1 = \frac{1}{2}\left[-\sqrt{u_0} + \sqrt{-2p_0 - u_0 + \frac{2q_0}{\sqrt{u_0}}}\right] \left(\left|M_S = +\frac{3}{2}\right\rangle\text{-dominant state}\right)$$

$$\varepsilon_2 = \frac{1}{2}\left[\sqrt{u_0} + \sqrt{-2p_0 - u_0 - \frac{2q_0}{\sqrt{u_0}}}\right] \left(\left|M_S = +\frac{1}{2}\right\rangle\text{-dominant state}\right)$$

$$\varepsilon_3 = \frac{1}{2}\left[\sqrt{u_0} - \sqrt{-2p_0 - u_0 - \frac{2q_0}{\sqrt{u_0}}}\right] \left(\left|M_S = -\frac{1}{2}\right\rangle\text{-dominant state}\right)$$

$$\varepsilon_4 = \frac{1}{2}\left[-\sqrt{u_0} - \sqrt{-2p_0 - u_0 + \frac{2q_0}{\sqrt{u_0}}}\right] \left(\left|M_S = -\frac{3}{2}\right\rangle\text{-dominant state}\right)$$

where u_0 is one of the solutions of the resolvent cubic equation of the secular equation,

$$u_0 = 2a_0 \cos\left[\frac{1}{3}\arccos\left(\frac{b_0}{2a_0}\right)\right]$$

with

$$a_0 = \frac{1}{3}\sqrt{{p_0}^2 + 12r_0}$$

$$b_0 = \frac{2{p_0}^3 + 27{q_0}^3 - 72p_0 r_0}{3{p_0}^2 + 36r_0}$$

Note that for $D > 0$, E_3, E_4, E_1 and E_2 is the eigenvalue corresponding to the $|M_S = +3/2>$-, $|+1/2>$-, $|–1/2>$- and $|–3/2>$-dominant state, respectively.

According to Denton and co-workers [7], the square of the jth element of the eigenvector corresponding to the eigenvalue ε_i of a Hermitian matrix can be described as follows

$$\left|v_{i,j}\right|^2 \prod_{k=1;k\neq i}^{n} (\varepsilon_i - \varepsilon_k) = \prod_{k=1}^{n-1}\left(\varepsilon_i - x_{j,k}\right)$$

where v_{ij} is the coefficient to be determined, $x_{j,k}$ is the eigenvalues of the minor M_j of the Hermitian formed by removing the jth row and column. In the case of the spin Hamiltonian under study, the spin eigenfunctions corresponding to the eigenvalue ε_n are written in the form

$$|\psi_n\rangle = \alpha_n\left|+\frac{3}{2}\right\rangle + \beta_n\left|+\frac{1}{2}\right\rangle + \gamma_n\left|-\frac{1}{2}\right\rangle + \delta_n\left|-\frac{3}{2}\right\rangle$$

where α_n, β_n, γ_n and δ_n correspond to v_{n1}, v_{n2}, v_{n3} and v_{n4}, respectively. By using the formula, we calculate α_1, β_2, γ_3 and δ_4, which are the diagonal element of the unitary matrix for diagonalizing the spin Hamiltonian matrix.

$$|\alpha_1|^2 = \frac{\left(\varepsilon_1 - x_{1,0}\right)\left(\varepsilon_1 - x_{1,1}\right)\left(\varepsilon_1 - x_{1,2}\right)}{(\varepsilon_1 - \varepsilon_2)(\varepsilon_1 - \varepsilon_3)(\varepsilon_1 - \varepsilon_4)}$$

$$|\beta_2|^2 = \frac{(\varepsilon_2 - x_{2,0})(\varepsilon_2 - x_{2,1})(\varepsilon_2 - x_{2,2})}{(\varepsilon_2 - \varepsilon_1)(\varepsilon_2 - \varepsilon_3)(\varepsilon_2 - \varepsilon_4)}$$

$$|\gamma_3|^2 = \frac{(\varepsilon_3 - x_{3,0})(\varepsilon_3 - x_{3,1})(\varepsilon_3 - x_{3,2})}{(\varepsilon_3 - \varepsilon_1)(\varepsilon_3 - \varepsilon_2)(\varepsilon_3 - \varepsilon_4)}$$

$$|\delta_4|^2 = \frac{(\varepsilon_4 - x_{4,0})(\varepsilon_4 - x_{4,1})(\varepsilon_4 - x_{4,2})}{(\varepsilon_4 - \varepsilon_1)(\varepsilon_4 - \varepsilon_2)(\varepsilon_4 - \varepsilon_3)}$$

In order to determine the element of the eigenvectors, we define the following four 3 by 3 matrixes;

$$M_1 = \begin{pmatrix} -D + \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) & g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz} & \sqrt{3}E \\ g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz} & -D - \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) & \frac{\sqrt{3}}{2}(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz}) \\ \sqrt{3}E & \frac{\sqrt{3}}{2}(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz}) & D - \frac{3}{2}(g'_{zx} + g'_{zy} + g'_{zz}) \end{pmatrix}$$

$$M_2 = \begin{pmatrix} D + \frac{3}{2}(g'_{zx} + g'_{zy} + g'_{zz}) & \sqrt{3}E & 0 \\ \sqrt{3}E & -D - \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) & \frac{\sqrt{3}}{2}(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz}) \\ 0 & \frac{\sqrt{3}}{2}(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz}) & D - \frac{3}{2}(g'_{zx} + g'_{zy} + g'_{zz}) \end{pmatrix}$$

$$M_3 = \begin{pmatrix} D + \frac{3}{2}(g'_{zx} + g'_{zy} + g'_{zz}) & \frac{\sqrt{3}}{2}(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz}) & 0 \\ \frac{\sqrt{3}}{2}(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz}) & -D + \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) & \sqrt{3}E \\ 0 & \sqrt{3}E & D - \frac{3}{2}(g'_{zx} + g'_{zy} + g'_{zz}) \end{pmatrix}$$

$$M_4 = \begin{pmatrix} D + \frac{3}{2}(g'_{zx} + g'_{zy} + g'_{zz}) & \frac{\sqrt{3}}{2}(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz}) & \sqrt{3}E \\ \frac{\sqrt{3}}{2}(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz}) & -D + \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) & g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz} \\ \sqrt{3}E & g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz} & -D - \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) \end{pmatrix}$$

and the secular equation for each matrix is represented as

$$x^3 + p_i x^2 + q_i x + r_i = 0 \ (i = 1,2,3,4)$$

with

$$p_1 = D + \frac{3}{2}g'_{zx} + \frac{3}{2}g'_{zy} + \frac{3}{2}g'_{zz}$$

$$\begin{aligned} q_1 = {} & -(D^2 + 3E^2) - \frac{7}{4}{g'_{xx}}^2 - \frac{7}{2}g'_{xx}g'_{xy} - \frac{7}{4}{g'_{xy}}^2 - \frac{7}{2}g'_{xx}g'_{xz} - \frac{7}{2}g'_{xy}g'_{xz} - \frac{7}{4}{g'_{xz}}^2 - \frac{7}{4}{g'_{yx}}^2 - \frac{7}{2}g'_{yx}g'_{yy} \\ & - \frac{7}{4}{g'_{yy}}^2 - \frac{7}{2}g'_{yx}g'_{yz} - \frac{7}{2}g'_{yy}g'_{yz} - \frac{7}{4}{g'_{yz}}^2 + 3Dg'_{zx} - \frac{1}{4}{g'_{zx}}^2 + 3Dg'_{zy} - \frac{1}{2}g'_{zx}g'_{zy} \\ & - \frac{1}{4}{g'_{zy}}^2 + 3Dg'_{zz} - \frac{1}{2}g'_{zx}g'_{zz} - \frac{1}{2}g'_{zy}g'_{zz} - \frac{1}{4}{g'_{zz}}^2 \end{aligned}$$

$$
\begin{aligned}
r_1 = {} & -D(D^2+3E^2)+\frac{1}{4}(D-12E){g'_{xx}}^2+\frac{1}{2}(D-12E)g'_{xx}g'_{xy}+\frac{1}{4}(D-12E){g'_{xy}}^2 \\
& +\frac{1}{2}(D-12E)g'_{xx}g'_{xz}+\frac{1}{2}(D-12E)g'_{xy}g'_{xz}+\frac{1}{4}(D-12E){g'_{xz}}^2+\frac{1}{4}(D+12E){g'_{yx}}^2 \\
& +\frac{1}{2}(D+12E)g'_{yx}g'_{yy}+\frac{1}{4}(D+12E){g'_{yy}}^2+\frac{1}{2}(D+12E)g'_{yx}g'_{yz} \\
& +\frac{1}{2}(D+12E)g'_{yy}g'_{yz}+\frac{1}{4}(D+12E){g'_{yz}}^2+\frac{3}{2}(D^2-E^2)g'_{zx}-\frac{9}{8}{g'_{xx}}^2g'_{zx} \\
& -\frac{9}{4}g'_{xx}g'_{xy}g'_{zx}-\frac{9}{8}{g'_{xy}}^2g'_{zx}-\frac{9}{4}g'_{xx}g'_{xz}g'_{zx}-\frac{9}{4}g'_{xy}g'_{xz}g'_{zx}-\frac{9}{8}{g'_{xz}}^2g'_{zx}-\frac{9}{8}{g'_{yx}}^2g'_{zx} \\
& -\frac{9}{4}g'_{yx}g'_{yy}g'_{zx}-\frac{9}{8}{g'_{yy}}^2g'_{zx}-\frac{9}{4}g'_{yx}g'_{yz}g'_{zx}-\frac{9}{4}g'_{yy}g'_{yz}g'_{zx}-\frac{9}{8}{g'_{yz}}^2g'_{zx}+\frac{1}{4}D{g'_{zx}}^2 \\
& -\frac{3}{8}{g'_{zx}}^3+\frac{3}{2}(D^2-E^2)g'_{zy}-\frac{9}{8}{g'_{xx}}^2g'_{zy}-\frac{9}{4}g'_{xx}g'_{xy}g'_{zy}-\frac{9}{8}{g'_{xy}}^2g'_{zy}-\frac{9}{4}g'_{xx}g'_{xz}g'_{zy} \\
& -\frac{9}{4}g'_{xy}g'_{xz}g'_{zy}-\frac{9}{8}{g'_{xz}}^2g'_{zy}-\frac{9}{8}{g'_{yx}}^2g'_{zy}-\frac{9}{4}g'_{yx}g'_{yy}g'_{zy}-\frac{9}{8}{g'_{yy}}^2g'_{zy}-\frac{9}{4}g'_{yx}g'_{yz}g'_{zy} \\
& -\frac{9}{4}g'_{yy}g'_{yz}g'_{zy}-\frac{9}{8}{g'_{yz}}^2g'_{zy}+\frac{1}{2}Dg'_{zx}g'_{zy}-\frac{9}{8}{g'_{zx}}^2g'_{zy}+\frac{1}{4}D{g'_{zy}}^2-\frac{9}{8}g'_{zx}{g'_{zy}}^2-\frac{3}{8}{g'_{zy}}^3 \\
& +\frac{3}{2}(D^2-E^2)g'_{zz}-\frac{9}{8}{g'_{xx}}^2g'_{zz}-\frac{9}{4}g'_{xx}g'_{xy}g'_{zz}-\frac{9}{8}{g'_{xy}}^2g'_{zz}-\frac{9}{4}g'_{xx}g'_{xz}g'_{zz} \\
& -\frac{9}{4}g'_{xy}g'_{xz}g'_{zz}-\frac{9}{8}{g'_{xz}}^2g'_{zz}-\frac{9}{8}{g'_{yx}}^2g'_{zz}-\frac{9}{4}g'_{yx}g'_{yy}g'_{zz}-\frac{9}{8}{g'_{yy}}^2g'_{zz}-\frac{9}{4}g'_{yx}g'_{yz}g'_{zz} \\
& -\frac{9}{4}g'_{yy}g'_{yz}g'_{zz}-\frac{9}{8}{g'_{yz}}^2g'_{zz}+\frac{1}{2}Dg'_{zx}g'_{zz}-\frac{9}{8}{g'_{zx}}^2g'_{zz}+\frac{1}{2}Dg'_{zy}g'_{zz}-\frac{9}{4}g'_{zx}g'_{zy}g'_{zz} \\
& -\frac{9}{8}{g'_{zy}}^2g'_{zz}+\frac{1}{4}D{g'_{zz}}^2-\frac{9}{8}g'_{zx}{g'_{zz}}^2-\frac{9}{8}g'_{zy}{g'_{zz}}^2-\frac{3}{8}{g'_{zz}}^3
\end{aligned}
$$

$$
p_2 = -D+\frac{3}{2}g'_{zx}+\frac{3}{2}g'_{zy}+\frac{3}{2}g'_{zz}
$$

$$
\begin{aligned}
q_2 = {} & -(D^2+3E^2)-\frac{3}{4}{g'_{xx}}^2-\frac{3}{2}g'_{xx}g'_{xy}-\frac{3}{4}{g'_{xy}}^2-\frac{3}{2}g'_{xx}g'_{xz}-\frac{3}{2}g'_{xy}g'_{xz}-\frac{3}{4}{g'_{xz}}^2-\frac{3}{4}{g'_{yx}}^2-\frac{3}{2}g'_{yx}g'_{yy} \\
& -\frac{3}{4}{g'_{yy}}^2-\frac{3}{2}g'_{yx}g'_{yz}-\frac{3}{2}g'_{yy}g'_{yz}-\frac{3}{4}{g'_{yz}}^2-Dg'_{zx}-\frac{9}{4}{g'_{zx}}^2-Dg'_{zy}-\frac{9}{2}g'_{zx}g'_{zy} \\
& -\frac{9}{4}{g'_{zy}}^2-Dg'_{zz}-\frac{9}{2}g'_{zx}g'_{zz}-\frac{9}{2}g'_{zy}g'_{zz}-\frac{9}{4}{g'_{zz}}^2
\end{aligned}
$$

$$
\begin{aligned}
r_2 = D(D^2+3E^2) &+ \frac{3}{4}Dg'^{\,2}_{xx} + \frac{3}{2}Dg'_{xx}g'_{xy} + \frac{3}{4}Dg'^{\,2}_{xy} + \frac{3}{2}Dg'_{xx}g'_{xz} + \frac{3}{2}Dg'_{xy}g'_{xz} + \frac{3}{4}Dg'^{\,2}_{xz} + \frac{3}{4}Dg'^{\,2}_{yx} \\
&+ \frac{3}{2}Dg'_{yx}g'_{yy} + \frac{3}{4}Dg'^{\,2}_{yy} + \frac{3}{2}Dg'_{yx}g'_{yz} + \frac{3}{2}Dg'_{yy}g'_{yz} + \frac{3}{4}Dg'^{\,2}_{yz} + \frac{1}{2}(D^2-9E^2)g'_{zx} \\
&+ \frac{9}{8}g'^{\,2}_{xx}g'_{zx} + \frac{9}{4}g'_{xx}g'_{xy}g'_{zx} + \frac{9}{8}g'^{\,2}_{xy}g'_{zx} + \frac{9}{4}g'_{xx}g'_{xz}g'_{zx} + \frac{9}{4}g'_{xy}g'_{xz}g'_{zx} + \frac{9}{8}g'^{\,2}_{xz}g'_{zx} \\
&+ \frac{9}{8}g'^{\,2}_{yx}g'_{zx} + \frac{9}{4}g'_{yx}g'_{yy}g'_{zx} + \frac{9}{8}g'^{\,2}_{yy}g'_{zx} + \frac{9}{4}g'_{yx}g'_{yz}g'_{zx} + \frac{9}{4}g'_{yy}g'_{yz}g'_{zx} + \frac{9}{8}g'^{\,2}_{yz}g'_{zx} \\
&- \frac{9}{4}Dg'^{\,2}_{zx} - \frac{9}{8}g'^{\,3}_{zx} + \frac{1}{2}(D^2-9E^2)g'_{zy} + \frac{9}{8}g'^{\,2}_{xx}g'_{zy} + \frac{9}{4}g'_{xx}g'_{xy}g'_{zy} + \frac{9}{8}g'^{\,2}_{xy}g'_{zy} \\
&+ \frac{9}{4}g'_{xx}g'_{xz}g'_{zy} + \frac{9}{4}g'_{xy}g'_{xz}g'_{zy} + \frac{9}{8}g'^{\,2}_{xz}g'_{zy} + \frac{9}{8}g'^{\,2}_{yx}g'_{zy} + \frac{9}{4}g'_{yx}g'_{yy}g'_{zy} + \frac{9}{8}g'^{\,2}_{yy}g'_{zy} \\
&+ \frac{9}{4}g'_{yx}g'_{yz}g'_{zy} + \frac{9}{4}g'_{yy}g'_{yz}g'_{zy} + \frac{9}{8}g'^{\,2}_{yz}g'_{zy} - \frac{9}{2}Dg'_{zx}g'_{zy} - \frac{27}{8}g'^{\,2}_{zx}g'_{zy} - \frac{9}{4}Dg'^{\,2}_{zy} \\
&- \frac{27}{8}g'_{zx}g'^{\,2}_{zy} - \frac{9}{8}g'^{\,3}_{zy} + \frac{1}{2}(D^2-9E^2)g'_{zz} + \frac{9}{8}g'^{\,2}_{xx}g'_{zz} + \frac{9}{4}g'_{xx}g'_{xy}g'_{zz} + \frac{9}{8}g'^{\,2}_{xy}g'_{zz} \\
&+ \frac{9}{4}g'_{xx}g'_{xz}g'_{zz} + \frac{9}{4}g'_{xy}g'_{xz}g'_{zz} + \frac{9}{8}g'^{\,2}_{xz}g'_{zz} + \frac{9}{8}g'^{\,2}_{yx}g'_{zz} + \frac{9}{4}g'_{yx}g'_{yy}g'_{zz} + \frac{9}{8}g'^{\,2}_{yy}g'_{zz} \\
&+ \frac{9}{4}g'_{yx}g'_{yz}g'_{zz} + \frac{9}{4}g'_{yy}g'_{yz}g'_{zz} + \frac{9}{8}g'^{\,2}_{yz}g'_{zz} - \frac{9}{2}Dg'_{zx}g'_{zz} - \frac{27}{8}g'^{\,2}_{zx}g'_{zz} - \frac{9}{2}Dg'_{zy}g'_{zz} \\
&- \frac{27}{4}g'_{zx}g'_{zy}g'_{zz} - \frac{27}{8}g'^{\,2}_{zy}g'_{zz} - \frac{9}{4}Dg'^{\,2}_{zz} - \frac{27}{8}g'_{zx}g'^{\,2}_{zz} - \frac{27}{8}g'_{zy}g'^{\,2}_{zz} - \frac{9}{8}g'^{\,3}_{zz}
\end{aligned}
$$

$$
p_3 = -D - \frac{3}{2}g'_{zx} - \frac{3}{2}g'_{zy} - \frac{3}{2}g'_{zz}
$$

$$
\begin{aligned}
q_3 = -(D^2+3E^2) &- \frac{3}{4}g'^{\,2}_{xx} - \frac{3}{2}g'_{xx}g'_{xy} - \frac{3}{4}g'^{\,2}_{xy} - \frac{3}{2}g'_{xx}g'_{xz} - \frac{3}{2}g'_{xy}g'_{xz} - \frac{3}{4}g'^{\,2}_{xz} - \frac{3}{4}g'^{\,2}_{yx} - \frac{3}{2}g'_{yx}g'_{yy} \\
&- \frac{3}{4}g'^{\,2}_{yy} - \frac{3}{2}g'_{yx}g'_{yz} - \frac{3}{2}g'_{yy}g'_{yz} - \frac{3}{4}g'^{\,2}_{yz} + Dg'_{zx} - \frac{9}{4}g'^{\,2}_{zx} + Dg'_{zy} - \frac{9}{2}g'_{zx}g'_{zy} \\
&- \frac{9}{4}g'^{\,2}_{zy} + Dg'_{zz} - \frac{9}{2}g'_{zx}g'_{zz} - \frac{9}{2}g'_{zy}g'_{zz} - \frac{9}{4}g'^{\,2}_{zz}
\end{aligned}
$$

$$r_3 = D(D^2+3E^2) + \frac{3}{4}Dg'^{\,2}_{xx} + \frac{3}{2}Dg'_{xx}g'_{xy} + \frac{3}{4}Dg'^{\,2}_{xy} + \frac{3}{2}Dg'_{xx}g'_{xz} + \frac{3}{2}Dg'_{xy}g'_{xz} + \frac{3}{4}Dg'^{\,2}_{xz} + \frac{3}{4}Dg'^{\,2}_{yx}$$
$$+ \frac{3}{2}Dg'_{yx}g'_{yy} + \frac{3}{4}Dg'^{\,2}_{yy} + \frac{3}{2}Dg'_{yx}g'_{yz} + \frac{3}{2}Dg'_{yy}g'_{yz} + \frac{3}{4}Dg'^{\,2}_{yz} - \frac{1}{2}(D^2-9E^2)g'_{zx}$$
$$- \frac{9}{8}g'^{\,2}_{xx}g'_{zx} - \frac{9}{4}g'_{xx}g'_{xy}g'_{zx} - \frac{9}{8}g'^{\,2}_{xy}g'_{zx} - \frac{9}{4}g'_{xx}g'_{xz}g'_{zx} - \frac{9}{4}g'_{xy}g'_{xz}g'_{zx} - \frac{9}{8}g'^{\,2}_{xz}g'_{zx}$$
$$- \frac{9}{8}g'^{\,2}_{yx}g'_{zx} - \frac{9}{4}g'_{yx}g'_{yy}g'_{zx} - \frac{9}{8}g'^{\,2}_{yy}g'_{zx} - \frac{9}{4}g'_{yx}g'_{yz}g'_{zx} - \frac{9}{4}g'_{yy}g'_{yz}g'_{zx} - \frac{9}{8}g'^{\,2}_{yz}g'_{zx}$$
$$- \frac{9}{4}Dg'^{\,2}_{zx} + \frac{9}{8}g'^{\,3}_{zx} - \frac{1}{2}(D^2-9E^2)g'_{zy} - \frac{9}{8}g'^{\,2}_{xx}g'_{zy} - \frac{9}{4}g'_{xx}g'_{xy}g'_{zy} - \frac{9}{8}g'^{\,2}_{xy}g'_{zy}$$
$$- \frac{9}{4}g'_{xx}g'_{xz}g'_{zy} - \frac{9}{4}g'_{xy}g'_{xz}g'_{zy} - \frac{9}{8}g'^{\,2}_{xz}g'_{zy} - \frac{9}{8}g'^{\,2}_{yx}g'_{zy} - \frac{9}{4}g'_{yx}g'_{yy}g'_{zy} - \frac{9}{8}g'^{\,2}_{yy}g'_{zy}$$
$$- \frac{9}{4}g'_{yx}g'_{yz}g'_{zy} - \frac{9}{4}g'_{yy}g'_{yz}g'_{zy} - \frac{9}{8}g'^{\,2}_{yz}g'_{zy} - \frac{9}{2}Dg'_{zx}g'_{zy} + \frac{27}{8}g'^{\,2}_{zx}g'_{zy} - \frac{9}{4}Dg'^{\,2}_{zy}$$
$$+ \frac{27}{8}g'_{zx}g'^{\,2}_{zy} + \frac{9}{8}g'^{\,3}_{zy} - \frac{1}{2}(D^2-9E^2)g'_{zz} - \frac{9}{8}g'^{\,2}_{xx}g'_{zz} - \frac{9}{4}g'_{xx}g'_{xy}g'_{zz} - \frac{9}{8}g'^{\,2}_{xy}g'_{zz}$$
$$- \frac{9}{4}g'_{xx}g'_{xz}g'_{zz} - \frac{9}{4}g'_{xy}g'_{xz}g'_{zz} - \frac{9}{8}g'^{\,2}_{xz}g'_{zz} - \frac{9}{8}g'^{\,2}_{yx}g'_{zz} - \frac{9}{4}g'_{yx}g'_{yy}g'_{zz} - \frac{9}{8}g'^{\,2}_{yy}g'_{zz}$$
$$- \frac{9}{4}g'_{yx}g'_{yz}g'_{zz} - \frac{9}{4}g'_{yy}g'_{yz}g'_{zz} - \frac{9}{8}g'^{\,2}_{yz}g'_{zz} - \frac{9}{2}Dg'_{zx}g'_{zz} + \frac{27}{8}g'^{\,2}_{zx}g'_{zz} - \frac{9}{2}Dg'_{zy}g'_{zz}$$
$$+ \frac{27}{4}g'_{zx}g'_{zy}g'_{zz} + \frac{27}{8}g'^{\,2}_{zy}g'_{zz} - \frac{9}{4}Dg'^{\,2}_{zz} + \frac{27}{8}g'_{zx}g'^{\,2}_{zz} + \frac{27}{8}g'_{zy}g'^{\,2}_{zz} + \frac{9}{8}g'^{\,3}_{zz}$$

$$p_4 = D - \frac{3}{2}g'_{zx} - \frac{3}{2}g'_{zy} - \frac{3}{2}g'_{zz}$$

$$q_4 = -(D^2+3E^2) - \frac{7}{4}g'^{\,2}_{xx} - \frac{7}{2}g'_{xx}g'_{xy} - \frac{7}{4}g'^{\,2}_{xy} - \frac{7}{2}g'_{xx}g'_{xz} - \frac{7}{2}g'_{xy}g'_{xz} - \frac{7}{4}g'^{\,2}_{xz} - \frac{7}{4}g'^{\,2}_{yx} - \frac{7}{2}g'_{yx}g'_{yy}$$
$$- \frac{7}{4}g'^{\,2}_{yy} - \frac{7}{2}g'_{yx}g'_{yz} - \frac{7}{2}g'_{yy}g'_{yz} - \frac{7}{4}g'^{\,2}_{yz} - 3Dg'_{zx} - \frac{1}{4}g'^{\,2}_{zx} - 3Dg'_{zy} - \frac{1}{2}g'_{zx}g'_{zy}$$
$$- \frac{1}{4}g'^{\,2}_{zy} - 3Dg'_{zz} - \frac{1}{2}g'_{zx}g'_{zz} - \frac{1}{2}g'_{zy}g'_{zz} - \frac{1}{4}g'^{\,2}_{zz}$$

$$
\begin{aligned}
r_4 = & -D(D^2+3E^2)+\frac{1}{4}(D-12E){g'_{xx}}^2+\frac{1}{2}(D-12E)g'_{xx}g'_{xy}+\frac{1}{4}(D-12E){g'_{xy}}^2 \\
& +\frac{1}{2}(D-12E)g'_{xx}g'_{xz}+\frac{1}{2}(D-12E)g'_{xy}g'_{xz}+\frac{1}{4}(D-12E){g'_{xz}}^2+\frac{1}{4}(D+12E){g'_{yx}}^2 \\
& +\frac{1}{2}(D+12E)g'_{yx}g'_{yy}+\frac{1}{4}(D+12E){g'_{yy}}^2+\frac{1}{2}(D+12E)g'_{yx}g'_{yz} \\
& +\frac{1}{2}(D+12E)g'_{yy}g'_{yz}+\frac{1}{4}(D+12E){g'_{yz}}^2-\frac{3}{2}(D^2-E^2)g'_{zx}+\frac{9}{8}{g'_{xx}}^2g'_{zx} \\
& +\frac{9}{4}g'_{xx}g'_{xy}g'_{zx}+\frac{9}{8}{g'_{xy}}^2g'_{zx}+\frac{9}{4}g'_{xx}g'_{xz}g'_{zx}+\frac{9}{4}g'_{xy}g'_{xz}g'_{zx}+\frac{9}{8}{g'_{xz}}^2g'_{zx}+\frac{9}{8}{g'_{yx}}^2g'_{zx} \\
& +\frac{9}{4}g'_{yx}g'_{yy}g'_{zx}+\frac{9}{8}{g'_{yy}}^2g'_{zx}+\frac{9}{4}g'_{yx}g'_{yz}g'_{zx}+\frac{9}{4}g'_{yy}g'_{yz}g'_{zx}+\frac{9}{8}{g'_{yz}}^2g'_{zx}+\frac{1}{4}D{g'_{zx}}^2 \\
& +\frac{3}{8}{g'_{zx}}^3-\frac{3}{2}(D^2-E^2)g'_{zy}+\frac{9}{8}{g'_{xx}}^2g'_{zy}+\frac{9}{4}g'_{xx}g'_{xy}g'_{zy}+\frac{9}{8}{g'_{xy}}^2g'_{zy}+\frac{9}{4}g'_{xx}g'_{xz}g'_{zy} \\
& +\frac{9}{4}g'_{xy}g'_{xz}g'_{zy}+\frac{9}{8}{g'_{xz}}^2g'_{zy}+\frac{9}{8}{g'_{yx}}^2g'_{zy}+\frac{9}{4}g'_{yx}g'_{yy}g'_{zy}+\frac{9}{8}{g'_{yy}}^2g'_{zy}+\frac{9}{4}g'_{yx}g'_{yz}g'_{zy} \\
& +\frac{9}{4}g'_{yy}g'_{yz}g'_{zy}+\frac{9}{8}{g'_{yz}}^2g'_{zy}+\frac{1}{2}Dg'_{zx}g'_{zy}+\frac{9}{8}{g'_{zx}}^2g'_{zy}+\frac{1}{4}D{g'_{zy}}^2+\frac{9}{8}g'_{zx}{g'_{zy}}^2+\frac{3}{8}{g'_{zy}}^3 \\
& -\frac{3}{2}(D^2-E^2)g'_{zz}+\frac{9}{8}{g'_{xx}}^2g'_{zz}+\frac{9}{4}g'_{xx}g'_{xy}g'_{zz}+\frac{9}{8}{g'_{xy}}^2g'_{zz}+\frac{9}{4}g'_{xx}g'_{xz}g'_{zz} \\
& +\frac{9}{4}g'_{xy}g'_{xz}g'_{zz}+\frac{9}{8}{g'_{xz}}^2g'_{zz}+\frac{9}{8}{g'_{yx}}^2g'_{zz}+\frac{9}{4}g'_{yx}g'_{yy}g'_{zz}+\frac{9}{8}{g'_{yy}}^2g'_{zz}+\frac{9}{4}g'_{yx}g'_{yz}g'_{zz} \\
& +\frac{9}{4}g'_{yy}g'_{yz}g'_{zz}+\frac{9}{8}{g'_{yz}}^2g'_{zz}+\frac{1}{2}Dg'_{zx}g'_{zz}+\frac{9}{8}{g'_{zx}}^2g'_{zz}+\frac{1}{2}Dg'_{zy}g'_{zz}+\frac{9}{4}g'_{zx}g'_{zy}g'_{zz} \\
& +\frac{9}{8}{g'_{zy}}^2g'_{zz}+\frac{1}{4}D{g'_{zz}}^2+\frac{9}{8}g'_{zx}{g'_{zz}}^2+\frac{9}{8}g'_{zy}{g'_{zz}}^2+\frac{3}{8}{g'_{zz}}^3
\end{aligned}
$$

Cubic equations,

$$x^3+p_ix^2+q_ix+r_i=0\ (i=1,2,3,4)$$

are transformed as follows;

$$x^3=\left(\frac{{p_i}^2-3q_i}{3}\right)x-\frac{2{p_i}^3-9p_iq_i+27r_i}{27}$$

by substituting x to $x-\frac{p_i}{3}$. Then, the solutions of the cubic equations are represented as follows.

$$x_{i,m}=2a_i\cos\left[\frac{1}{3}\arccos\left(\frac{b_i}{2a_i}\right)+\frac{2m\pi}{3}\right]-\frac{p_i}{3}\ (m=0,1,2)$$

with

$$a_i=\frac{\sqrt{{p_i}^2-3q_i}}{3}$$

$$b_i = -\frac{2{p_i}^3 - 9p_i q_i + 27r_i}{3{p_i}^2 - 9q_i}$$

For n = 1 (M_S = +3/2), $\alpha_1 = r_{\alpha 1} = \sqrt{\left|v_{1,1}\right|^2}$ (real).

$$\begin{cases} \left[D + \frac{3}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_1\right] r_{\alpha 1} + \frac{\sqrt{3}}{2}(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz})\beta_1 + \sqrt{3}E\gamma_1 = 0 \text{ — (A2)} \\ \frac{\sqrt{3}}{2}(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz})r_{\alpha 1} + \left[-D + \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_1\right]\beta_1 + (g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz})\gamma_1 + \sqrt{3}E\delta_1 = 0 \text{ — (B2)} \\ \sqrt{3}Er_{\alpha 1} + (g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz})\beta_1 + \left[-D - \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_1\right]\gamma_1 + \frac{\sqrt{3}}{2}(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz})\delta_1 = 0 \text{ — (C2)} \\ \sqrt{3}E\beta_1 + \frac{\sqrt{3}}{2}(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz})\gamma_1 + \left[D - \frac{3}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_1\right]\delta_1 = 0 \text{ — (D2)} \end{cases}$$

(C2) × $2E(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz})$

$$\begin{aligned} &2\sqrt{3}E^2(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz})r_{\alpha 1} + 2E(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz})^2\beta_1 \\ &\quad + 2E\left[-D - \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_1\right](g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz})\gamma_1 \\ &\quad + \sqrt{3}E({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2)\delta_1 = 0 \text{ — (E2)} \end{aligned}$$

(B2) × $({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2)$

$$\begin{aligned} &\frac{\sqrt{3}}{2}({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2)(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz})r_{\alpha 1} \\ &\quad + \left[-D + \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_1\right]({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2)\beta_1 \\ &\quad + ({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2)(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} \\ &\quad - ig'_{yz})\gamma_1 + \sqrt{3}E({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2)\delta_1 = 0 \text{ — (F2)} \end{aligned}$$

(E2) – (F2)

$$\begin{aligned} &\frac{\sqrt{3}}{2}(4E^2 - {g'_{xx}}^2 - {g'_{yx}}^2 - {g'_{xy}}^2 - {g'_{yy}}^2 - {g'_{xz}}^2 - {g'_{yz}}^2)(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz})r_{\alpha 1} \\ &\quad + \Big\{2E(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz})^2 \\ &\quad - \left[-D + \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_1\right]({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2)\Big\}\beta_1 \\ &\quad + \Big\{2E\left[-D - \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_1\right](g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz}) \\ &\quad - ({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2)(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} \\ &\quad - ig'_{yz})\Big\}\gamma_1 = 0 \text{ — (G2)} \end{aligned}$$

(G2)×$\sqrt{3}E$

$$\frac{3}{2}E\left(4E^2 - {g'_{xx}}^2 - {g'_{yx}}^2 - {g'_{xy}}^2 - {g'_{yy}}^2 - {g'_{xz}}^2 - {g'_{yz}}^2\right)\left(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz}\right)r_{\alpha 1}$$

$$+ \left\{2\sqrt{3}E^2\left(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz}\right)^2\right.$$

$$- \sqrt{3}E\left[-D + \frac{1}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_1\right]\left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2\right.$$

$$\left.\left. + {g'_{yz}}^2\right)\right\}\beta_1$$

$$+ \left\{2\sqrt{3}E^2\left[-D - \frac{1}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_1\right]\left(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz}\right)\right.$$

$$- \sqrt{3}E\left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2\right)\left(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz}\right.$$

$$\left.\left. - ig'_{yz}\right)\right\}\gamma_1 = 0 \text{ — (H2)}$$

$$\text{(H2)} \times \left\{2E\left[-D + \frac{1}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_1\right]\left(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz}\right)\right.$$

$$- \left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2\right)\left(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz}\right.$$

$$\left.\left. - ig'_{yz}\right)\right\}$$

$$\left\{2E\left[D + \frac{3}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_1\right]\left[-D - \frac{1}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_1\right]\left(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz}\right.\right.$$

$$\left. + ig'_{yz}\right)$$

$$- \left[D + \frac{3}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_1\right]\left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2\right)\left(g'_{xx}\right.$$

$$\left.\left. - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz}\right)\right\}r_{\alpha 1}$$

$$+ \left\{\sqrt{3}E\left[-D - \frac{1}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_1\right]\left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2\right)\right.$$

$$- \frac{\sqrt{3}}{2}\left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2\right.$$

$$\left.\left. + {g'_{yz}}^2\right)\left(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz}\right)^2\right\}\beta_1$$

$$+ \left\{2\sqrt{3}E^2\left[-D - \frac{1}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_1\right]\left(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz}\right)\right.$$

$$- \sqrt{3}E\left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2\right.$$

$$\left.\left. + {g'_{yz}}^2\right)\left(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz}\right)^2\right\}\gamma_1 = 0 \text{ — (I2)}$$

Applying (H2) – (I2) yields the following equation,

$$\beta_1 = \frac{numer(\beta_1)}{denom(\beta_1)}r_{\alpha 1}$$

where

$$numer(\beta_1) = 2E\left[D + \frac{3}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_1\right]\left[-D - \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_1\right](g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz})$$
$$- \left[D + \frac{3}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_1\right]({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2)(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz})$$
$$- \frac{3}{2}E(4E^2 - {g'_{xx}}^2 - {g'_{yx}}^2 - {g'_{xy}}^2 - {g'_{yy}}^2 - {g'_{xz}}^2 - {g'_{yz}}^2)(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz})$$

$$denom(\beta_1) = 2\sqrt{3}E(D + \varepsilon_1)({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2)$$
$$+ 2\sqrt{3}E^2(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz})^2$$
$$+ \frac{\sqrt{3}}{2}({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2)(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz})^2$$

Next,

(G2) $\times \frac{1}{2}\sqrt{3}(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz})$

$$\frac{3}{4}(4E^2 - {g'_{xx}}^2 - {g'_{yx}}^2 - {g'_{xy}}^2 - {g'_{yy}}^2 - {g'_{xz}}^2 - {g'_{yz}}^2)({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2)r_{\alpha 1}$$
$$+ \Big\{\sqrt{3}E({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2)(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz})$$
$$- \frac{\sqrt{3}}{2}\left[-D + \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_1\right]({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2)(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz})\Big\}\beta_1$$
$$+ \Big\{\sqrt{3}E\left[-D - \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_1\right]({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2)$$
$$- \frac{\sqrt{3}}{2}({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2)(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz})^2\Big\}\gamma_1 = 0 \text{ — (J2)}$$

(A2) $\times \Big\{2E(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz})^2$

$$- \left[-D + \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_1\right]({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2)\Big\}$$

$$\Big\{2E\left[D+\frac{3}{2}\left(g'_{zx}+g'_{zy}+g'_{zz}\right)-\varepsilon_1\right]\left(g'_{xx}+ig'_{yx}+g'_{xy}+ig'_{yy}+g'_{xz}+ig'_{yz}\right)^2$$

$$-\left[D+\frac{3}{2}\left(g'_{zx}+g'_{zy}+g'_{zz}\right)-\varepsilon_1\right]\left[-D+\frac{1}{2}\left(g'_{zx}+g'_{zy}+g'_{zz}\right)-\varepsilon_1\right]\left({g'_{xx}}^2+{g'_{yx}}^2\right.$$

$$\left.+{g'_{xy}}^2+{g'_{yy}}^2+{g'_{xz}}^2+{g'_{yz}}^2\right)\Big\}r_{\alpha 1}$$

$$+\Big\{\sqrt{3}E\left({g'_{xx}}^2+{g'_{yx}}^2+{g'_{xy}}^2+{g'_{yy}}^2+{g'_{xz}}^2+{g'_{yz}}^2\right)\left(g'_{xx}+ig'_{yx}+g'_{xy}+ig'_{yy}+g'_{xz}\right.$$

$$\left.+ig'_{yz}\right)$$

$$-\frac{\sqrt{3}}{2}\left[-D+\frac{1}{2}\left(g'_{zx}+g'_{zy}+g'_{zz}\right)-\varepsilon_1\right]\left({g'_{xx}}^2+{g'_{yx}}^2+{g'_{xy}}^2+{g'_{yy}}^2+{g'_{xz}}^2\right.$$

$$\left.+{g'_{yz}}^2\right)\left(g'_{xx}-ig'_{yx}+g'_{xy}-ig'_{yy}+g'_{xz}-ig'_{yz}\right)\Big\}\beta_1$$

$$+\Big[2\sqrt{3}E^2\left(g'_{xx}+ig'_{yx}+g'_{xy}+ig'_{yy}+g'_{xz}+ig'_{yz}\right)^2$$

$$-\sqrt{3}E\left[-D+\frac{1}{2}\left(g'_{zx}+g'_{zy}+g'_{zz}\right)-\varepsilon_1\right]\left({g'_{xx}}^2+{g'_{yx}}^2+{g'_{xy}}^2+{g'_{yy}}^2+{g'_{xz}}^2\right.$$

$$\left.+{g'_{yz}}^2\right)\Big]\gamma_1=0 \text{ — (K2)}$$

Applyging (J2) – (K2) yields the following equation,

$$\gamma_1=\frac{numer(\gamma_1)}{denom(\gamma_1)}r_{\alpha 1}$$

where

$$numer(\gamma_1)=\left[D+\frac{3}{2}\left(g'_{zx}+g'_{zy}+g'_{zz}\right)-\varepsilon_1\right]\left[-D+\frac{1}{2}\left(g'_{zx}+g'_{zy}+g'_{zz}\right)-\varepsilon_1\right]\left({g'_{xx}}^2+{g'_{yx}}^2+{g'_{xy}}^2\right.$$

$$\left.+{g'_{yy}}^2+{g'_{xz}}^2+{g'_{yz}}^2\right)$$

$$-2E\left[D+\frac{3}{2}\left(g'_{zx}+g'_{zy}+g'_{zz}\right)-\varepsilon_1\right]\left(g'_{xx}+ig'_{yx}+g'_{xy}+ig'_{yy}+g'_{xz}+ig'_{yz}\right)^2$$

$$+\frac{3}{4}\left(4E^2-{g'_{xx}}^2-{g'_{yx}}^2-{g'_{xy}}^2-{g'_{yy}}^2-{g'_{xz}}^2-{g'_{yz}}^2\right)\left({g'_{xx}}^2+{g'_{yx}}^2+{g'_{xy}}^2+{g'_{yy}}^2\right.$$

$$\left.+{g'_{xz}}^2+{g'_{yz}}^2\right)$$

$$denom(\gamma_1)=2\sqrt{3}E(D+\varepsilon_1)\left({g'_{xx}}^2+{g'_{yx}}^2+{g'_{xy}}^2+{g'_{yy}}^2+{g'_{xz}}^2+{g'_{yz}}^2\right)$$

$$+2\sqrt{3}E^2\left(g'_{xx}+ig'_{yx}+g'_{xy}+ig'_{yy}+g'_{xz}+ig'_{yz}\right)^2$$

$$+\frac{\sqrt{3}}{2}\left({g'_{xx}}^2+{g'_{yx}}^2+{g'_{xy}}^2+{g'_{yy}}^2+{g'_{xz}}^2\right.$$

$$\left.+{g'_{yz}}^2\right)\left(g'_{xx}-ig'_{yx}+g'_{xy}-ig'_{yy}+g'_{xz}-ig'_{yz}\right)^2$$

Finally, from (D2) we obtain

$$\delta_1=\frac{-2\sqrt{3}E\beta_1-\sqrt{3}\left(g'_{xx}+ig'_{yx}+g'_{xy}+ig'_{yy}+g'_{xz}+ig'_{yz}\right)\gamma_1}{2D-3\left(g'_{zx}+g'_{zy}+g'_{zz}\right)-2E_1}$$

Similarly, for $n = 2$ ($M_S = +1/2$), we set

$$\beta_2 = r_{\beta 2} = \sqrt{\left|v_{2,2}\right|^2}$$

and

$$\gamma_2 = \frac{numer(\gamma_2)}{denom(\gamma_2)} r_{\beta 2}$$

$$\begin{aligned} numer(\gamma_2) = {} & 2E\left[D - \frac{3}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_2\right]\left[-D + \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_2\right](g'_{xx} - ig'_{yx} + g'_{xy} \\ & - ig'_{yy} + g'_{xz} - ig'_{yz}) \\ & - \left[D - \frac{3}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_2\right]({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2)(g'_{xx} \\ & + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz}) \\ & - \frac{3}{2}E(4E^2 - {g'_{xx}}^2 - {g'_{yx}}^2 - {g'_{xy}}^2 - {g'_{yy}}^2 - {g'_{xz}}^2 - {g'_{yz}}^2)(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} \\ & + g'_{xz} - ig'_{yz}) \end{aligned}$$

$$\begin{aligned} denom(\gamma_2) = {} & \left[D - \frac{3}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_2\right]\left[-D - \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_2\right]({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 \\ & + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2) \\ & - 2E\left[D - \frac{3}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_2\right](g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz})^2 \\ & + \frac{3}{4}(4E^2 - {g'_{xx}}^2 - {g'_{yx}}^2 - {g'_{xy}}^2 - {g'_{yy}}^2 - {g'_{xz}}^2 - {g'_{yz}}^2)({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 \\ & + {g'_{xz}}^2 + {g'_{yz}}^2) \end{aligned}$$

$$\delta_2 = \frac{numer(\delta_2)}{denom(\delta_2)} r_{\beta 2}$$

$$\begin{aligned} numer(\delta_2) = {} & 2\sqrt{3}E(D + \varepsilon_2)({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2) \\ & + 2\sqrt{3}E^2(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz})^2 \\ & + \frac{\sqrt{3}}{2}({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 \\ & + {g'_{yz}}^2)(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz})^2 \end{aligned}$$

$$\begin{aligned} denom(\delta_2) = {} & \left[D - \frac{3}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_2\right]\left[-D - \frac{1}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_2\right]({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 \\ & + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2) \\ & - 2E\left[D - \frac{3}{2}(g'_{zx} + g'_{zy} + g'_{zz}) - \varepsilon_2\right](g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz})^2 \\ & + \frac{3}{4}(4E^2 - {g'_{xx}}^2 - {g'_{yx}}^2 - {g'_{xy}}^2 - {g'_{yy}}^2 - {g'_{xz}}^2 - {g'_{yz}}^2)({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 \\ & + {g'_{xz}}^2 + {g'_{yz}}^2) \end{aligned}$$

$$\alpha_2 = \frac{-\sqrt{3}\left(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz}\right)r_{\beta 2} - 2\sqrt{3}E\gamma_2}{2D + 3\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - 2\varepsilon_2}$$

were obtained. For n = 3 (M_S = –1/2), we set

$$\gamma_3 = r_{\gamma 3} = \sqrt{\left|v_{3,3}\right|^2}$$

and

$$\alpha_3 = \frac{numer(\alpha_3)}{denom(\alpha_3)} r_{\gamma 3}$$

$$\begin{aligned} numer(\alpha_3) = {} & 2\sqrt{3}E(D + \varepsilon_3)\left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2\right) \\ & + 2\sqrt{3}E^2\left(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz}\right)^2 \\ & + \frac{\sqrt{3}}{2}\left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 \right. \\ & \left. + {g'_{yz}}^2\right)\left(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz}\right)^2 \end{aligned}$$

$$\begin{aligned} denom(\alpha_3) = {} & \left[D + \frac{3}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_3\right]\left[-D + \frac{1}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_3\right]\left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 \right. \\ & \left. + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2\right) \\ & - 2E\left[D + \frac{3}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_3\right]\left(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz}\right)^2 \\ & + \frac{3}{4}\left(4E^2 - {g'_{xx}}^2 - {g'_{yx}}^2 - {g'_{xy}}^2 - {g'_{yy}}^2 - {g'_{xz}}^2 - {g'_{yz}}^2\right)\left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 \right. \\ & \left. + {g'_{xz}}^2 + {g'_{yz}}^2\right) \end{aligned}$$

$$\beta_3 = \frac{numer(\beta_3)}{denom(\beta_3)} r_{\gamma 3}$$

$$\begin{aligned} numer(\beta_3) = {} & 2E\left[D + \frac{3}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_3\right]\left[-D - \frac{1}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_3\right]\left(g'_{xx} + ig'_{yx} + g'_{xy} \right. \\ & \left. + ig'_{yy} + g'_{xz} + ig'_{yz}\right) \\ & - \left[D + \frac{3}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_3\right]\left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2\right)\left(g'_{xx} \right. \\ & \left. - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz}\right) \\ & - \frac{3}{2}E\left(4E^2 - {g'_{xx}}^2 - {g'_{yx}}^2 - {g'_{xy}}^2 - {g'_{yy}}^2 - {g'_{xz}}^2 - {g'_{yz}}^2\right)\left(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} \right. \\ & \left. + g'_{xz} + ig'_{yz}\right) \end{aligned}$$

$$denom(\beta_3) = \left[D + \frac{3}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_3\right]\left[-D + \frac{1}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_3\right]\left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2\right) - 2E\left[D + \frac{3}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_3\right]\left(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz}\right)^2 + \frac{3}{4}\left(4E^2 - {g'_{xx}}^2 - {g'_{yx}}^2 - {g'_{xy}}^2 - {g'_{yy}}^2 - {g'_{xz}}^2 - {g'_{yz}}^2\right)\left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2\right)$$

$$\delta_1 = \frac{-2\sqrt{3}E\beta_3 - \sqrt{3}\left(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz}\right)r_{\gamma 3}}{2D - 3\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - 2\varepsilon_3}$$

were obtained.

For $n = 4$ ($M_S = -3/2$), we set

$$\delta_4 = r_{\delta 4} = \sqrt{\left|v_{4,4}\right|^2}$$

$$\beta_4 = \frac{numer(\beta_4)}{denom(\beta_4)}r_{\delta 4}$$

$$numer(\beta_4) = \left[-D - \frac{1}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_4\right]\left[D - \frac{3}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_4\right]\left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2\right) - 2E\left[D - \frac{3}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_4\right]\left(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz}\right)^2 + \frac{3}{4}\left(4E^2 - {g'_{xx}}^2 - {g'_{yx}}^2 - {g'_{xy}}^2 - {g'_{yy}}^2 - {g'_{xz}}^2 - {g'_{yz}}^2\right)\left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2\right)$$

$$denom(\beta_4) = 2\sqrt{3}E(D + \varepsilon_4)\left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2\right) + 2\sqrt{3}E^2\left(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz}\right)^2 + \frac{\sqrt{3}}{2}\left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2\right)\left(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz}\right)^2$$

$$\gamma_4 = \frac{numer(\gamma_4)}{denom(\gamma_4)}r_{\delta 4}$$

$$numer(\gamma_4) = 2E\left[-D + \frac{1}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_4\right]\left[D - \frac{3}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_4\right]\left(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz}\right) - \left[D - \frac{3}{2}\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - \varepsilon_4\right]\left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2\right)\left(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz}\right) - \frac{3}{2}E\left(4E^2 - {g'_{xx}}^2 - {g'_{yx}}^2 - {g'_{xy}}^2 - {g'_{yy}}^2 - {g'_{xz}}^2 - {g'_{yz}}^2\right)\left(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz}\right)$$

$$denom(\gamma_4) = 2\sqrt{3}E(D + \varepsilon_4)\left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2\right) + 2\sqrt{3}E^2\left(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz}\right)^2 + \frac{\sqrt{3}}{2}\left({g'_{xx}}^2 + {g'_{yx}}^2 + {g'_{xy}}^2 + {g'_{yy}}^2 + {g'_{xz}}^2 + {g'_{yz}}^2\right)\left(g'_{xx} + ig'_{yx} + g'_{xy} + ig'_{yy} + g'_{xz} + ig'_{yz}\right)^2$$

$$\alpha_4 = \frac{-\sqrt{3}\left(g'_{xx} - ig'_{yx} + g'_{xy} - ig'_{yy} + g'_{xz} - ig'_{yz}\right)\beta_4 - 2\sqrt{3}E\gamma_4}{2D + 3\left(g'_{zx} + g'_{zy} + g'_{zz}\right) - 2\varepsilon_4}$$

were obtained.

In the case of the non-collinearity between the ZFS and **A**-tensors, the matrix representation of the **A**-tensor is transformed in the basis of the principal axis system of the ZFS tensor by a unitary matrix

$$\mathbf{A} = \begin{pmatrix} A_x & 0 & 0 \\ 0 & A_y & 0 \\ 0 & 0 & A_z \end{pmatrix} \rightarrow \mathbf{A}' = \begin{pmatrix} A'_{xx} & A'_{xy} & A'_{xz} \\ A'_{yx} & A'_{yy} & A'_{yz} \\ A'_{zx} & A'_{zy} & A'_{zz} \end{pmatrix}$$

Then, the matrix elements of the hyperfine structure Hamiltonian in the basis of $|M_S, M_I\rangle$ are as follows.

$$\left\langle \pm\frac{3}{2}, M'_I \middle| H_{\mathrm{hfs}} \middle| \pm\frac{3}{2}, M_I \right\rangle = \begin{cases} \pm 2M_I \delta_{M_I M'_I} A'_{z1} \\ \pm\frac{1}{2}\sqrt{63 - 4M_I M'_I}\,\delta_{M_I M'_I+1} A'_{z2} \\ \pm\frac{1}{2}\sqrt{63 - 4M_I M'_I}\,\delta_{M_I M'_I-1} {A'_{z2}}^* \end{cases}$$

$$\left\langle +\frac{1}{2}, M'_I \middle| H_{\mathrm{hfs}} \middle| +\frac{3}{2}, M_I \right\rangle = \left\langle -\frac{3}{2}, M'_I \middle| H_{\mathrm{hfs}} \middle| -\frac{1}{2}, M_I \right\rangle = \begin{cases} 2M_I \delta_{M_I M'_I} A'_{z3} \\ \frac{1}{2}\sqrt{63 - 4M_I M'_I}\,\delta_{M_I M'_I+1} A'_{1-} \\ \frac{1}{2}\sqrt{63 - 4M_I M'_I}\,\delta_{M_I M'_I-1} A'_{1+} \end{cases}$$

$$\left\langle +\frac{3}{2}, M'_I \middle| H_{\mathrm{hfs}} \middle| +\frac{1}{2}, M_I \right\rangle = \left\langle +\frac{1}{2}, M_I \middle| H_{\mathrm{hfs}} \middle| +\frac{3}{2}, M'_I \right\rangle^*$$

$$\left\langle \pm\frac{1}{2}, M'_I \middle| H_{\mathrm{hfs}} \middle| \pm\frac{1}{2}, M_I \right\rangle = \begin{cases} \pm 2M_I \delta_{M_I M'_I} A'_{z4} \\ \pm\frac{1}{2}\sqrt{63 - 4M_I M'_I}\,\delta_{M_I M'_I+1} A'_{z5} \\ \pm\frac{1}{2}\sqrt{63 - 4M_I M'_I}\,\delta_{M_I M'_I-1} {A'_{z5}}^* \end{cases}$$

$$\left\langle -\frac{1}{2}, M_I' \middle| H_{\text{hfs}} \middle| +\frac{1}{2}, M_I \right\rangle = \begin{cases} 2M_I \delta_{M_I M_I'} A_{z6}' \\ \frac{1}{2}\sqrt{63 - 4M_I M_I'}\delta_{M_I M_I'+1} A_{2-}' \\ \frac{1}{2}\sqrt{63 - 4M_I M_I'}\delta_{M_I M_I'-1} A_{2+}' \end{cases}$$

$$\left\langle +\frac{1}{2}, M_I' \middle| H_{\text{hfs}} \middle| -\frac{1}{2}, M_I \right\rangle = \left\langle -\frac{1}{2}, M_I \middle| H_{\text{hfs}} \middle| +\frac{1}{2}, M_I' \right\rangle^*$$

$$\left\langle -\frac{1}{2}, M_I' \middle| H_{\text{hfs}} \middle| -\frac{3}{2}, M_I \right\rangle = \left\langle -\frac{3}{2}, M_I \middle| H_{\text{hfs}} \middle| -\frac{1}{2}, M_I' \right\rangle^*$$

where

$$\begin{aligned} A_{z1}' &= \tfrac{3}{4} A_{zz}' \\ A_{z2}' &= \tfrac{3}{4}\left(A_{zx}' + iA_{zy}'\right) \\ A_{z3}' &= \tfrac{1}{4}\sqrt{3}\left(A_{xz}' + iA_{yz}'\right) \\ A_{z4}' &= \tfrac{1}{4} A_{zz}' \\ A_{z5}' &= \tfrac{1}{4}\left(A_{zx}' + iA_{zy}'\right) \\ A_{z6}' &= \tfrac{1}{2}\left(A_{xz}' + iA_{yz}'\right) \\ A_{1\pm}' &= \tfrac{1}{4}\sqrt{3}\left(A_{xx}' \mp iA_{xy}' + iA_{yx}' \pm A_{yy}'\right) \\ A_{2\pm}' &= \tfrac{1}{2}\left(A_{xx}' \mp iA_{xy}' + iA_{yx}' \pm A_{yy}'\right) \end{aligned}$$

The upper and lower signs should be chosen in the double sign.

In order to expand the hyperfine structure Hamiltonian, we carry out the calculations based on the tensor product between the spin eigenfunctions and the basis vector of the nuclear spin operator, as given in the following.

$$|\psi_n(M_I)\rangle \equiv |\psi_n\rangle \otimes |M_I\rangle = \alpha_n \left| +\frac{3}{2}, M_I \right\rangle + \beta_n \left| +\frac{1}{2}, M_I \right\rangle + \gamma_n \left| -\frac{1}{2}, M_I \right\rangle + \delta_n \left| -\frac{3}{2}, M_I \right\rangle$$

$$\left(n = 1, M_S = +\frac{3}{2};\ n = 2, M_S = +\frac{1}{2}; n = 3, M_S = -\frac{1}{2}; n = 4, M_S = -\frac{3}{2}\right)$$

Then, the matrix element of the expanded hyperfine structure Hamiltonian can be simply represented.

$$\left\langle \psi_{M_S'}, M_I' \middle| H_{\text{hfs}} \middle| \psi_{M_S}, M_I \right\rangle = \begin{cases} \pm 2M_I \delta_{M_I M_I'} \lambda_{ij} \\ \pm\frac{1}{2}\sqrt{63 - 4M_I M_I'}\delta_{M_I M_I'+1} \mu_{ij} \\ \pm\frac{1}{2}\sqrt{63 - 4M_I M_I'}\delta_{M_I M_I'-1} \nu_{ij} \end{cases}$$

where

$$\begin{aligned} \lambda_{ij} &= \tfrac{1}{4}\left(A_{xz}'\xi_{ij} + iA_{yz}'\eta_{ij} + A_{zz}'\zeta_{ij}\right) \\ \mu_{ij} &= \tfrac{1}{4}\left(A_{xx}'\xi_{ij} + iA_{xy}'\xi_{ij} + iA_{yx}'\eta_{ij} - A_{yy}'\eta_{ij} + A_{zx}'\zeta_{ij} + iA_{zy}'\zeta_{ij}\right) \\ \nu_{ij} &= \tfrac{1}{4}\left(A_{xx}'\xi_{ij} - iA_{xy}'\xi_{ij} + iA_{yx}'\eta_{ij} - A_{yy}'\eta_{ij} + A_{zx}'\zeta_{ij} - iA_{zy}'\zeta_{ij}\right) \\ \xi_{ij} &= \sqrt{3}\beta_i^*\alpha_j + \sqrt{3}\alpha_i^*\beta_j + 2\gamma_i^*\beta_j + 2\beta_i^*\gamma_j + \sqrt{3}\delta_i^*\gamma_j + \sqrt{3}\gamma_i^*\delta_j \\ \eta_{ij} &= \sqrt{3}\beta_i^*\alpha_j - \sqrt{3}\alpha_i^*\beta_j + 2\gamma_i^*\beta_j - 2\beta_i^*\gamma_j + \sqrt{3}\delta_i^*\gamma_j - \sqrt{3}\gamma_i^*\delta_j \\ \zeta_{ij} &= 3\alpha_i^*\alpha_j + \beta_i^*\beta_j - \gamma_i^*\gamma_j - 3\delta_i^*\delta_j \end{aligned}$$

and i (j) = 1 if $M_S'(M_S) = +\frac{3}{2}$, i (j) = 2, if $M_S'(M_S) = +\frac{1}{2}$, i (j) = 3, if $M_S'(M_S) = -\frac{1}{2}$ and i (j) = 4 if $M_S'(M_S) = -\frac{3}{2}$, respectively. Note that

$$\xi_{ij}{}^{*} = \xi_{ji}$$
$$\eta_{ij}{}^{*} = -\eta_{ji}$$
$$\zeta_{ij}{}^{*} = \zeta_{ji}$$

and thus,

$$\lambda_{ij}{}^{*} = \lambda_{ji}$$
$$\mu_{ij}{}^{*} = \nu_{ji}$$

Zeroth-order energies are the energy eigenvalues of the non-perturbed Hamiltonian,

$$\varepsilon_{+\frac{3}{2}} = \varepsilon_1^{(0)} = \frac{1}{2}\left[-\sqrt{u_0} + \sqrt{-2p - u_0 + \frac{2q}{\sqrt{u_0}}}\right]$$

$$\varepsilon_{+\frac{1}{2}} = \varepsilon_2^{(0)} = \frac{1}{2}\left[\sqrt{u_0} + \sqrt{-2p - u_0 - \frac{2q}{\sqrt{u_0}}}\right]$$

$$\varepsilon_{-\frac{1}{2}} = \varepsilon_3^{(0)} = \frac{1}{2}\left[\sqrt{u_0} - \sqrt{-2p - u_0 - \frac{2q}{\sqrt{u_0}}}\right]$$

$$\varepsilon_{-\frac{3}{2}} = \varepsilon_4^{(0)} = \frac{1}{2}\left[-\sqrt{u_0} - \sqrt{-2p - u_0 + \frac{2q}{\sqrt{u_0}}}\right]$$

Since the energies correspond to the same M_S-dominant state, for example, $\left|\psi_{+\frac{3}{2}}, +\frac{7}{2}\right\rangle$ and $\left|\psi_{+\frac{3}{2}}, +\frac{5}{2}\right\rangle$ are degenerate ($\varepsilon_{+\frac{3}{2}}$) and the matrix element of the expanded hyperfine structure Hamiltonian $\left\langle\psi_{+\frac{3}{2}}, +\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}}, +\frac{5}{2}\right\rangle$ is not zero, the first-order energies must be obtained by using the degenerate perturbation theory. The sub-matrixes to be diagonalized are $\left|\psi_{M_S}, M_I\right\rangle\left\langle\psi_{M_S}, M_I\middle|H_{\mathrm{hfs}}\middle|\psi_{M_S}, M_I'\right\rangle\left\langle\psi_{M_S}, M_I'\right|$ ($M_S = \pm\frac{3}{2}$ and $\pm\frac{1}{2}$, $M_I, M_I' = \pm\frac{7}{2}, \pm\frac{5}{2}, \pm\frac{3}{2}$ and $\pm\frac{1}{2}$). Therefore, the first-order energies can be represented as

$$\varepsilon_{M_S,M_I}^{(1)} = \mathrm{sgn}(M_S) 2M_I\sqrt{|\lambda_{ii}|^2 + \mu_{ii}\nu_{ii}}$$

Where $i = 1$ if $M_S = +\frac{3}{2}$, $i = 2$ if $M_S = +\frac{1}{2}$, $i = 3$ if $M_S = -\frac{1}{2}$ and $i = 4$ if $M_S = -\frac{3}{2}$. The function sgn(x) is the sign function defined by

$$\mathrm{sgn}(x) = \begin{cases} 1, & x > 0 \\ 0, & x = 0 \\ -1, & x < 0 \end{cases}$$

The second-order energies are as follows:

$$\varepsilon^{(2)}_{+\frac{3}{2},+\frac{7}{2}} = \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},+\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}}$$

$$= \frac{49\lambda_{12}\lambda_{21} + 7\mu_{12}\nu_{21}}{\varepsilon_1^{(0)}-\varepsilon_2^{(0)}} + \frac{49\lambda_{13}\lambda_{31} + 7\mu_{13}\nu_{31}}{\varepsilon_1^{(0)}-\varepsilon_3^{(0)}} + \frac{49\lambda_{14}\lambda_{41} + 7\mu_{14}\nu_{41}}{\varepsilon_1^{(0)}-\varepsilon_4^{(0)}}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},+\frac{5}{2}} = \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},+\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}}$$

$$= \frac{25\lambda_{12}\lambda_{21} + 19\mu_{12}\nu_{21}}{\varepsilon_1^{(0)}-\varepsilon_2^{(0)}} + \frac{25\lambda_{13}\lambda_{31} + 19\mu_{13}\nu_{31}}{\varepsilon_1^{(0)}-\varepsilon_3^{(0)}} + \frac{25\lambda_{14}\lambda_{41} + 19\mu_{14}\nu_{41}}{\varepsilon_1^{(0)}-\varepsilon_4^{(0)}}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},+\frac{3}{2}} = \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}}$$

$$= \frac{9\lambda_{12}\lambda_{21} + 27\mu_{12}\nu_{21}}{\varepsilon_1^{(0)}-\varepsilon_2^{(0)}} + \frac{9\lambda_{13}\lambda_{31} + 27\mu_{13}\nu_{31}}{\varepsilon_1^{(0)}-\varepsilon_3^{(0)}} + \frac{9\lambda_{14}\lambda_{41} + 27\mu_{14}\nu_{41}}{\varepsilon_1^{(0)}-\varepsilon_4^{(0)}}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},+\frac{1}{2}} = \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}}$$

$$= \frac{\lambda_{12}\lambda_{21}+31\mu_{12}\nu_{21}}{\varepsilon_1^{(0)}-\varepsilon_2^{(0)}} + \frac{\lambda_{13}\lambda_{31}+31\mu_{13}\nu_{31}}{\varepsilon_1^{(0)}-\varepsilon_3^{(0)}} + \frac{\lambda_{14}\lambda_{41}+31\mu_{14}\nu_{41}}{\varepsilon_1^{(0)}-\varepsilon_4^{(0)}}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},-\frac{1}{2}} = \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}}$$

$$= \frac{\lambda_{12}\lambda_{21}+31\mu_{12}\nu_{21}}{\varepsilon_1^{(0)}-\varepsilon_2^{(0)}} + \frac{\lambda_{13}\lambda_{31}+31\mu_{13}\nu_{31}}{\varepsilon_1^{(0)}-\varepsilon_3^{(0)}} + \frac{\lambda_{14}\lambda_{41}+31\mu_{14}\nu_{41}}{\varepsilon_1^{(0)}-\varepsilon_4^{(0)}}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},-\frac{3}{2}} = \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}}$$

$$= \frac{9\lambda_{12}\lambda_{21}+27\mu_{12}\nu_{21}}{\varepsilon_1^{(0)}-\varepsilon_2^{(0)}} + \frac{9\lambda_{13}\lambda_{31}+27\mu_{13}\nu_{31}}{\varepsilon_1^{(0)}-\varepsilon_3^{(0)}} + \frac{9\lambda_{14}\lambda_{41}+27\mu_{14}\nu_{41}}{\varepsilon_1^{(0)}-\varepsilon_4^{(0)}}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},-\frac{5}{2}} = \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}}$$

$$= \frac{25\lambda_{12}\lambda_{21} + 19\mu_{12}\nu_{21}}{\varepsilon_1^{(0)}-\varepsilon_2^{(0)}} + \frac{25\lambda_{13}\lambda_{31} + 19\mu_{13}\nu_{31}}{\varepsilon_1^{(0)}-\varepsilon_3^{(0)}} + \frac{25\lambda_{14}\lambda_{41} + 19\mu_{14}\nu_{41}}{\varepsilon_1^{(0)}-\varepsilon_4^{(0)}}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},-\frac{7}{2}} = \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{3}{2}},-\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{3}{2}}-\varepsilon_{-\frac{3}{2}}}$$

$$= \frac{49\lambda_{12}\lambda_{21} + 7\mu_{12}\nu_{21}}{\varepsilon_1^{(0)}-\varepsilon_2^{(0)}} + \frac{49\lambda_{13}\lambda_{31} + 7\mu_{13}\nu_{31}}{\varepsilon_1^{(0)}-\varepsilon_3^{(0)}} + \frac{49\lambda_{14}\lambda_{41} + 7\mu_{14}\nu_{41}}{\varepsilon_1^{(0)}-\varepsilon_4^{(0)}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},+\frac{7}{2}} = \frac{\left|\left\langle\psi_{+\frac{1}{2}},+\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},+\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},+\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},+\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{1}{2}},+\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},+\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},+\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},+\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}}$$

$$= \frac{49\lambda_{21}\lambda_{12} + 7\mu_{21}\nu_{12}}{\varepsilon_2^{(0)}-\varepsilon_1^{(0)}} + \frac{49\lambda_{23}\lambda_{32} + 7\mu_{23}\nu_{32}}{\varepsilon_2^{(0)}-\varepsilon_3^{(0)}} + \frac{49\lambda_{24}\lambda_{42} + 7\mu_{24}\nu_{42}}{\varepsilon_2^{(0)}-\varepsilon_4^{(0)}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},+\frac{5}{2}} = \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}}, +\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}}, +\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}}, +\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{+\frac{3}{2}}}$$

$$+ \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}}, +\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}}, +\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}}, +\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}}, +\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}}, +\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}}, +\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{3}{2}}}$$

$$= \frac{25\lambda_{21}\lambda_{12} + 19\mu_{21}\nu_{12}}{\varepsilon_2^{(0)} - \varepsilon_1^{(0)}} + \frac{25\lambda_{23}\lambda_{32} + 19\mu_{23}\nu_{32}}{\varepsilon_2^{(0)} - \varepsilon_3^{(0)}} + \frac{25\lambda_{24}\lambda_{42} + 19\mu_{24}\nu_{42}}{\varepsilon_2^{(0)} - \varepsilon_4^{(0)}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},+\frac{3}{2}} = \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}}, +\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}}, +\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}}, +\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{+\frac{3}{2}}}$$

$$+ \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}}, +\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}}, +\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}}, +\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}}, +\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}}, +\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}}, +\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{3}{2}}}$$

$$= \frac{9\lambda_{21}\lambda_{12} + 27\mu_{21}\nu_{12}}{\varepsilon_2^{(0)} - \varepsilon_1^{(0)}} + \frac{9\lambda_{23}\lambda_{32} + 27\mu_{23}\nu_{32}}{\varepsilon_2^{(0)} - \varepsilon_3^{(0)}} + \frac{9\lambda_{24}\lambda_{42} + 27\mu_{24}\nu_{42}}{\varepsilon_2^{(0)} - \varepsilon_4^{(0)}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},+\frac{1}{2}} = \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}}, +\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}}, +\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}}, -\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{+\frac{3}{2}}}$$

$$+ \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}}, +\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}}, +\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}}, -\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}}, +\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}}, +\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, +\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}}, -\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{3}{2}}}$$

$$= \frac{\lambda_{21}\lambda_{12} + 31\mu_{21}\nu_{12}}{\varepsilon_2^{(0)} - \varepsilon_1^{(0)}} + \frac{\lambda_{23}\lambda_{32} + 31\mu_{23}\nu_{32}}{\varepsilon_2^{(0)} - \varepsilon_3^{(0)}} + \frac{\lambda_{24}\lambda_{42} + 31\mu_{24}\nu_{42}}{\varepsilon_2^{(0)} - \varepsilon_4^{(0)}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},-\frac{1}{2}} = \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}}$$

$$= \frac{\lambda_{21}\lambda_{12} + 31\mu_{21}\nu_{12}}{\varepsilon^{(0)}_2 - \varepsilon^{(0)}_1} + \frac{\lambda_{23}\lambda_{32} + 31\mu_{23}\nu_{32}}{\varepsilon^{(0)}_2 - \varepsilon^{(0)}_3} + \frac{\lambda_{24}\lambda_{42} + 31\mu_{24}\nu_{42}}{\varepsilon^{(0)}_2 - \varepsilon^{(0)}_4}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},-\frac{3}{2}} = \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}}$$

$$= \frac{9\lambda_{21}\lambda_{12} + 27\mu_{21}\nu_{12}}{\varepsilon^{(0)}_2 - \varepsilon^{(0)}_1} + \frac{9\lambda_{23}\lambda_{32} + 27\mu_{23}\nu_{32}}{\varepsilon^{(0)}_2 - \varepsilon^{(0)}_3} + \frac{9\lambda_{24}\lambda_{42} + 27\mu_{24}\nu_{42}}{\varepsilon^{(0)}_2 - \varepsilon^{(0)}_4}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},-\frac{5}{2}} = \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{+\frac{1}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}}$$

$$= \frac{25\lambda_{21}\lambda_{12} + 19\mu_{21}\nu_{12}}{\varepsilon^{(0)}_2 - \varepsilon^{(0)}_1} + \frac{25\lambda_{23}\lambda_{32} + 19\mu_{23}\nu_{32}}{\varepsilon^{(0)}_2 - \varepsilon^{(0)}_3} + \frac{25\lambda_{24}\lambda_{42} + 19\mu_{24}\nu_{42}}{\varepsilon^{(0)}_2 - \varepsilon^{(0)}_4}$$

$$
\begin{aligned}
\varepsilon^{(2)}_{+\frac{1}{2},-\frac{7}{2}} = {} & \frac{\left|\left\langle \psi_{+\frac{1}{2}}, -\frac{7}{2} \middle| H_{\text{hfs}} \middle| \psi_{+\frac{3}{2}}, -\frac{5}{2} \right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, -\frac{7}{2} \middle| H_{\text{hfs}} \middle| \psi_{+\frac{3}{2}}, -\frac{7}{2} \right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, -\frac{7}{2} \middle| H_{\text{hfs}} \middle| \psi_{-\frac{1}{2}}, -\frac{5}{2} \right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{1}{2}}} \\
& + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, -\frac{7}{2} \middle| H_{\text{hfs}} \middle| \psi_{-\frac{1}{2}}, -\frac{7}{2} \right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, -\frac{7}{2} \middle| H_{\text{hfs}} \middle| \psi_{-\frac{3}{2}}, -\frac{5}{2} \right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle \psi_{+\frac{1}{2}}, -\frac{7}{2} \middle| H_{\text{hfs}} \middle| \psi_{-\frac{3}{2}}, -\frac{7}{2} \right\rangle\right|^2}{\varepsilon_{+\frac{1}{2}} - \varepsilon_{-\frac{3}{2}}} \\
& = \frac{49\lambda_{21}\lambda_{12} + 7\mu_{21}\nu_{12}}{\varepsilon_2^{(0)} - \varepsilon_1^{(0)}} + \frac{49\lambda_{23}\lambda_{32} + 7\mu_{23}\nu_{32}}{\varepsilon_2^{(0)} - \varepsilon_3^{(0)}} + \frac{49\lambda_{24}\lambda_{42} + 7\mu_{24}\nu_{42}}{\varepsilon_2^{(0)} - \varepsilon_4^{(0)}}
\end{aligned}
$$

$$
\begin{aligned}
\varepsilon^{(2)}_{-\frac{1}{2},+\frac{7}{2}} = {} & \frac{\left|\left\langle \psi_{-\frac{1}{2}}, +\frac{7}{2} \middle| H_{\text{hfs}} \middle| \psi_{+\frac{3}{2}}, +\frac{7}{2} \right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}} - \varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}}, +\frac{7}{2} \middle| H_{\text{hfs}} \middle| \psi_{+\frac{3}{2}}, +\frac{5}{2} \right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}} - \varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}}, +\frac{7}{2} \middle| H_{\text{hfs}} \middle| \psi_{+\frac{1}{2}}, +\frac{7}{2} \right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}} - \varepsilon_{+\frac{1}{2}}} \\
& + \frac{\left|\left\langle \psi_{-\frac{1}{2}}, +\frac{7}{2} \middle| H_{\text{hfs}} \middle| \psi_{+\frac{1}{2}}, +\frac{5}{2} \right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}} - \varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}}, +\frac{7}{2} \middle| H_{\text{hfs}} \middle| \psi_{-\frac{3}{2}}, +\frac{7}{2} \right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}} - \varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}}, +\frac{7}{2} \middle| H_{\text{hfs}} \middle| \psi_{-\frac{3}{2}}, +\frac{5}{2} \right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}} - \varepsilon_{-\frac{3}{2}}} \\
& = \frac{49\lambda_{31}\lambda_{13} + 7\mu_{31}\nu_{13}}{\varepsilon_3^{(0)} - \varepsilon_1^{(0)}} + \frac{49\lambda_{32}\lambda_{23} + 7\mu_{32}\nu_{23}}{\varepsilon_3^{(0)} - \varepsilon_2^{(0)}} + \frac{49\lambda_{34}\lambda_{43} + 7\mu_{34}\nu_{43}}{\varepsilon_3^{(0)} - \varepsilon_4^{(0)}}
\end{aligned}
$$

$$
\begin{aligned}
\varepsilon^{(2)}_{-\frac{1}{2},+\frac{5}{2}} = {} & \frac{\left|\left\langle \psi_{-\frac{1}{2}}, +\frac{5}{2} \middle| H_{\text{hfs}} \middle| \psi_{+\frac{3}{2}}, +\frac{7}{2} \right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}} - \varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}}, +\frac{5}{2} \middle| H_{\text{hfs}} \middle| \psi_{+\frac{3}{2}}, +\frac{5}{2} \right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}} - \varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}}, +\frac{5}{2} \middle| H_{\text{hfs}} \middle| \psi_{+\frac{3}{2}}, +\frac{3}{2} \right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}} - \varepsilon_{+\frac{3}{2}}} \\
& + \frac{\left|\left\langle \psi_{-\frac{1}{2}}, +\frac{5}{2} \middle| H_{\text{hfs}} \middle| \psi_{+\frac{1}{2}}, +\frac{7}{2} \right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}} - \varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}}, +\frac{5}{2} \middle| H_{\text{hfs}} \middle| \psi_{+\frac{1}{2}}, +\frac{5}{2} \right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}} - \varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}}, +\frac{5}{2} \middle| H_{\text{hfs}} \middle| \psi_{+\frac{1}{2}}, +\frac{3}{2} \right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}} - \varepsilon_{+\frac{1}{2}}} \\
& + \frac{\left|\left\langle \psi_{-\frac{1}{2}}, +\frac{5}{2} \middle| H_{\text{hfs}} \middle| \psi_{-\frac{3}{2}}, +\frac{7}{2} \right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}} - \varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}}, +\frac{5}{2} \middle| H_{\text{hfs}} \middle| \psi_{-\frac{3}{2}}, +\frac{5}{2} \right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}} - \varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}}, +\frac{5}{2} \middle| H_{\text{hfs}} \middle| \psi_{-\frac{3}{2}}, +\frac{3}{2} \right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}} - \varepsilon_{-\frac{3}{2}}} \\
& = \frac{25\lambda_{31}\lambda_{13} + 19\mu_{31}\nu_{13}}{\varepsilon_3^{(0)} - \varepsilon_1^{(0)}} + \frac{25\lambda_{32}\lambda_{23} + 19\mu_{32}\nu_{23}}{\varepsilon_3^{(0)} - \varepsilon_2^{(0)}} + \frac{25\lambda_{34}\lambda_{43} + 19\mu_{34}\nu_{43}}{\varepsilon_3^{(0)} - \varepsilon_4^{(0)}}
\end{aligned}
$$

$$\varepsilon^{(2)}_{-\frac{1}{2},+\frac{3}{2}} = \frac{\left|\left\langle\psi_{-\frac{1}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{1}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{1}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{-\frac{1}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{1}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{1}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{-\frac{1}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{1}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{1}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}}$$

$$= \frac{9\lambda_{31}\lambda_{13} + 27\mu_{31}\nu_{13}}{\varepsilon^{(0)}_3 - \varepsilon^{(0)}_1} + \frac{9\lambda_{32}\lambda_{23} + 27\mu_{32}\nu_{23}}{\varepsilon^{(0)}_3 - \varepsilon^{(0)}_2} + \frac{9\lambda_{34}\lambda_{43} + 27\mu_{34}\nu_{43}}{\varepsilon^{(0)}_3 - \varepsilon^{(0)}_4}$$

$$\varepsilon^{(2)}_{-\frac{1}{2},+\frac{1}{2}} = \frac{\left|\left\langle\psi_{-\frac{1}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{1}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{1}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{-\frac{1}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{1}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{1}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{-\frac{1}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{1}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{1}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}}$$

$$= \frac{\lambda_{31}\lambda_{13} + 31\mu_{31}\nu_{13}}{\varepsilon^{(0)}_3 - \varepsilon^{(0)}_1} + \frac{\lambda_{32}\lambda_{23} + 31\mu_{32}\nu_{23}}{\varepsilon^{(0)}_3 - \varepsilon^{(0)}_2} + \frac{\lambda_{34}\lambda_{43} + 31\mu_{34}\nu_{43}}{\varepsilon^{(0)}_3 - \varepsilon^{(0)}_4}$$

$$\varepsilon^{(2)}_{-\frac{1}{2},-\frac{1}{2}} = \frac{\left|\left\langle\psi_{-\frac{1}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{1}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{1}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{-\frac{1}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{1}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{1}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{-\frac{1}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{1}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{1}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}}$$

$$= \frac{\lambda_{31}\lambda_{13} + 31\mu_{31}\nu_{13}}{\varepsilon^{(0)}_3 - \varepsilon^{(0)}_1} + \frac{\lambda_{32}\lambda_{23} + 31\mu_{32}\nu_{23}}{\varepsilon^{(0)}_3 - \varepsilon^{(0)}_2} + \frac{\lambda_{34}\lambda_{43} + 31\mu_{34}\nu_{43}}{\varepsilon^{(0)}_3 - \varepsilon^{(0)}_4}$$

$$\begin{aligned}\varepsilon^{(2)}_{-\frac{1}{2},-\frac{3}{2}} &= \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}}\\
&+ \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{1}{2}}}\\
&+ \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}}\\
&= \frac{9\lambda_{31}\lambda_{13}+27\mu_{31}\nu_{13}}{\varepsilon_3^{(0)}-\varepsilon_1^{(0)}} + \frac{9\lambda_{32}\lambda_{23}+27\mu_{32}\nu_{23}}{\varepsilon_3^{(0)}-\varepsilon_2^{(0)}} + \frac{9\lambda_{34}\lambda_{43}+27\mu_{34}\nu_{43}}{\varepsilon_3^{(0)}-\varepsilon_4^{(0)}}\end{aligned}$$

$$\begin{aligned}\varepsilon^{(2)}_{-\frac{1}{2},-\frac{5}{2}} &= \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}}\\
&+ \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{1}{2}}}\\
&+ \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}}\\
&= \frac{25\lambda_{31}\lambda_{13}+19\mu_{31}\nu_{13}}{\varepsilon_3^{(0)}-\varepsilon_1^{(0)}} + \frac{25\lambda_{32}\lambda_{23}+19\mu_{32}\nu_{23}}{\varepsilon_3^{(0)}-\varepsilon_2^{(0)}} + \frac{25\lambda_{34}\lambda_{43}+19\mu_{34}\nu_{43}}{\varepsilon_3^{(0)}-\varepsilon_4^{(0)}}\end{aligned}$$

$$\begin{aligned}\varepsilon^{(2)}_{-\frac{1}{2},-\frac{7}{2}} &= \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{1}{2}}}\\
&+ \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}} + \frac{\left|\left\langle \psi_{-\frac{1}{2}},-\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{3}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{1}{2}}-\varepsilon_{-\frac{3}{2}}}\\
&= \frac{49\lambda_{31}\lambda_{13}+7\mu_{31}\nu_{13}}{\varepsilon_3^{(0)}-\varepsilon_1^{(0)}} + \frac{49\lambda_{32}\lambda_{23}+7\mu_{32}\nu_{23}}{\varepsilon_3^{(0)}-\varepsilon_2^{(0)}} + \frac{49\lambda_{34}\lambda_{43}+7\mu_{34}\nu_{43}}{\varepsilon_3^{(0)}-\varepsilon_4^{(0)}}\end{aligned}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},+\frac{7}{2}} = \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},+\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}}$$

$$= \frac{49\lambda_{41}\lambda_{14}+7\mu_{41}\nu_{14}}{\varepsilon^{(0)}_4-\varepsilon^{(0)}_1} + \frac{49\lambda_{42}\lambda_{24}+7\mu_{42}\nu_{24}}{\varepsilon^{(0)}_4-\varepsilon^{(0)}_2} + \frac{49\lambda_{43}\lambda_{34}+7\mu_{43}\nu_{34}}{\varepsilon^{(0)}_4-\varepsilon^{(0)}_3}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},+\frac{5}{2}} = \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},+\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}}$$

$$= \frac{25\lambda_{41}\lambda_{14}+19\mu_{41}\nu_{14}}{\varepsilon^{(0)}_4-\varepsilon^{(0)}_1} + \frac{25\lambda_{42}\lambda_{24}+19\mu_{42}\nu_{24}}{\varepsilon^{(0)}_4-\varepsilon^{(0)}_2} + \frac{25\lambda_{43}\lambda_{34}+19\mu_{43}\nu_{34}}{\varepsilon^{(0)}_4-\varepsilon^{(0)}_3}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},+\frac{3}{2}} = \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}}$$

$$= \frac{9\lambda_{41}\lambda_{14}+27\mu_{41}\nu_{14}}{\varepsilon^{(0)}_4-\varepsilon^{(0)}_1} + \frac{9\lambda_{42}\lambda_{24}+27\mu_{42}\nu_{24}}{\varepsilon^{(0)}_4-\varepsilon^{(0)}_2} + \frac{9\lambda_{43}\lambda_{34}+27\mu_{43}\nu_{34}}{\varepsilon^{(0)}_4-\varepsilon^{(0)}_3}$$

$$\begin{aligned}\varepsilon^{(2)}_{-\frac{3}{2},+\frac{1}{2}} &= \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}}\\
&+ \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}}\\
&+ \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},+\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}}\\
&= \frac{\lambda_{41}\lambda_{14}+31\mu_{41}\nu_{14}}{\varepsilon^{(0)}_4-\varepsilon^{(0)}_1} + \frac{\lambda_{42}\lambda_{24}+31\mu_{42}\nu_{24}}{\varepsilon^{(0)}_4-\varepsilon^{(0)}_2} + \frac{\lambda_{43}\lambda_{34}+31\mu_{43}\nu_{34}}{\varepsilon^{(0)}_4-\varepsilon^{(0)}_3}\end{aligned}$$

$$\begin{aligned}\varepsilon^{(2)}_{-\frac{3}{2},-\frac{1}{2}} &= \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}}\\
&+ \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}}\\
&+ \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},+\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{1}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}}\\
&= \frac{\lambda_{41}\lambda_{14}+31\mu_{41}\nu_{14}}{\varepsilon^{(0)}_4-\varepsilon^{(0)}_1} + \frac{\lambda_{42}\lambda_{24}+31\mu_{42}\nu_{24}}{\varepsilon^{(0)}_4-\varepsilon^{(0)}_2} + \frac{\lambda_{43}\lambda_{34}+31\mu_{43}\nu_{34}}{\varepsilon^{(0)}_4-\varepsilon^{(0)}_3}\end{aligned}$$

$$\begin{aligned}\varepsilon^{(2)}_{-\frac{3}{2},-\frac{3}{2}} &= \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}}\\
&+ \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}}\\
&+ \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{1}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{3}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}}\\
&= \frac{9\lambda_{41}\lambda_{14}+27\mu_{41}\nu_{14}}{\varepsilon^{(0)}_4-\varepsilon^{(0)}_1} + \frac{9\lambda_{42}\lambda_{24}+27\mu_{42}\nu_{24}}{\varepsilon^{(0)}_4-\varepsilon^{(0)}_2} + \frac{9\lambda_{43}\lambda_{34}+27\mu_{43}\nu_{34}}{\varepsilon^{(0)}_4-\varepsilon^{(0)}_3}\end{aligned}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},-\frac{5}{2}} = \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{3}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{5}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}}$$

$$= \frac{25\lambda_{41}\lambda_{14}+19\mu_{41}\nu_{14}}{\varepsilon_4^{(0)}-\varepsilon_1^{(0)}} + \frac{25\lambda_{42}\lambda_{24}+19\mu_{42}\nu_{24}}{\varepsilon_4^{(0)}-\varepsilon_2^{(0)}} + \frac{25\lambda_{43}\lambda_{34}+19\mu_{43}\nu_{34}}{\varepsilon_4^{(0)}-\varepsilon_3^{(0)}}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},-\frac{7}{2}} = \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{3}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{3}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}}$$

$$+ \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{+\frac{1}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{+\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{5}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}} + \frac{\left|\left\langle\psi_{-\frac{3}{2}},-\frac{7}{2}\middle|H_{\mathrm{hfs}}\middle|\psi_{-\frac{1}{2}},-\frac{7}{2}\right\rangle\right|^2}{\varepsilon_{-\frac{3}{2}}-\varepsilon_{-\frac{1}{2}}}$$

$$= \frac{49\lambda_{41}\lambda_{14}+7\mu_{41}\nu_{14}}{\varepsilon_4^{(0)}-\varepsilon_1^{(0)}} + \frac{49\lambda_{42}\lambda_{24}+7\mu_{42}\nu_{24}}{\varepsilon_4^{(0)}-\varepsilon_2^{(0)}} + \frac{49\lambda_{43}\lambda_{34}+7\mu_{43}\nu_{34}}{\varepsilon_4^{(0)}-\varepsilon_3^{(0)}}$$

The perturbed energies to the second order are as follows.

$$E_{+\frac{3}{2},+\frac{7}{2}} = \frac{1}{2}\left[-\sqrt{u_0}+\sqrt{-2p-u_0+\frac{2q}{\sqrt{u_0}}}\right] + 7\sqrt{|\lambda_{11}|^2+\mu_{11}\nu_{11}} + \frac{49\lambda_{12}\lambda_{21}+7\mu_{12}\nu_{21}}{\varepsilon_1^{(0)}-\varepsilon_2^{(0)}}$$

$$+ \frac{49\lambda_{13}\lambda_{31}+7\mu_{13}\nu_{31}}{\varepsilon_1^{(0)}-\varepsilon_3^{(0)}} + \frac{49\lambda_{14}\lambda_{41}+7\mu_{14}\nu_{41}}{\varepsilon_1^{(0)}-\varepsilon_4^{(0)}}$$

$$E_{+\frac{3}{2},+\frac{5}{2}} = \frac{1}{2}\left[-\sqrt{u_0}+\sqrt{-2p-u_0+\frac{2q}{\sqrt{u_0}}}\right] + 5\sqrt{|\lambda_{11}|^2+\mu_{11}\nu_{11}} + \frac{25\lambda_{12}\lambda_{21}+19\mu_{12}\nu_{21}}{\varepsilon_1^{(0)}-\varepsilon_2^{(0)}}$$

$$+ \frac{25\lambda_{13}\lambda_{31}+19\mu_{13}\nu_{31}}{\varepsilon_1^{(0)}-\varepsilon_3^{(0)}} + \frac{25\lambda_{14}\lambda_{41}+19\mu_{14}\nu_{41}}{\varepsilon_1^{(0)}-\varepsilon_4^{(0)}}$$

$$E_{+\frac{3}{2},+\frac{3}{2}} = \frac{1}{2}\left[-\sqrt{u_0}+\sqrt{-2p-u_0+\frac{2q}{\sqrt{u_0}}}\right] + 3\sqrt{|\lambda_{11}|^2+\mu_{11}\nu_{11}} + \frac{9\lambda_{12}\lambda_{21}+27\mu_{12}\nu_{21}}{\varepsilon_1^{(0)}-\varepsilon_2^{(0)}}$$

$$+ \frac{9\lambda_{13}\lambda_{31}+27\mu_{13}\nu_{31}}{\varepsilon_1^{(0)}-\varepsilon_3^{(0)}} + \frac{9\lambda_{14}\lambda_{41}+27\mu_{14}\nu_{41}}{\varepsilon_1^{(0)}-\varepsilon_4^{(0)}}$$

$$E_{+\frac{3}{2},+\frac{1}{2}} = \frac{1}{2}\left[-\sqrt{u_0} + \sqrt{-2p - u_0 + \frac{2q}{\sqrt{u_0}}}\right] + \sqrt{|\lambda_{11}|^2 + \mu_{11}\nu_{11}} + \frac{\lambda_{12}\lambda_{21} + 31\mu_{12}\nu_{21}}{\varepsilon_1^{(0)} - \varepsilon_2^{(0)}} + \frac{\lambda_{13}\lambda_{31} + 31\mu_{13}\nu_{31}}{\varepsilon_1^{(0)} - \varepsilon_3^{(0)}} + \frac{\lambda_{14}\lambda_{41} + 31\mu_{14}\nu_{41}}{\varepsilon_1^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{+\frac{3}{2},-\frac{1}{2}} = \frac{1}{2}\left[-\sqrt{u_0} + \sqrt{-2p - u_0 + \frac{2q}{\sqrt{u_0}}}\right] - \sqrt{|\lambda_{11}|^2 + \mu_{11}\nu_{11}} + \frac{\lambda_{12}\lambda_{21} + 31\mu_{12}\nu_{21}}{\varepsilon_1^{(0)} - \varepsilon_2^{(0)}} + \frac{\lambda_{13}\lambda_{31} + 31\mu_{13}\nu_{31}}{\varepsilon_1^{(0)} - \varepsilon_3^{(0)}} + \frac{\lambda_{14}\lambda_{41} + 31\mu_{14}\nu_{41}}{\varepsilon_1^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{+\frac{3}{2},-\frac{3}{2}} = \frac{1}{2}\left[-\sqrt{u_0} + \sqrt{-2p - u_0 + \frac{2q}{\sqrt{u_0}}}\right] - 3\sqrt{|\lambda_{11}|^2 + \mu_{11}\nu_{11}} + \frac{9\lambda_{12}\lambda_{21} + 27\mu_{12}\nu_{21}}{\varepsilon_1^{(0)} - \varepsilon_2^{(0)}} + \frac{9\lambda_{13}\lambda_{31} + 27\mu_{13}\nu_{31}}{\varepsilon_1^{(0)} - \varepsilon_3^{(0)}} + \frac{9\lambda_{14}\lambda_{41} + 27\mu_{14}\nu_{41}}{\varepsilon_1^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{+\frac{3}{2},-\frac{5}{2}} = \frac{1}{2}\left[-\sqrt{u_0} + \sqrt{-2p - u_0 + \frac{2q}{\sqrt{u_0}}}\right] - 5\sqrt{|\lambda_{11}|^2 + \mu_{11}\nu_{11}} + \frac{25\lambda_{12}\lambda_{21} + 19\mu_{12}\nu_{21}}{\varepsilon_1^{(0)} - \varepsilon_2^{(0)}} + \frac{25\lambda_{13}\lambda_{31} + 19\mu_{13}\nu_{31}}{\varepsilon_1^{(0)} - \varepsilon_3^{(0)}} + \frac{25\lambda_{14}\lambda_{41} + 19\mu_{14}\nu_{41}}{\varepsilon_1^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{+\frac{3}{2},-\frac{7}{2}} = \frac{1}{2}\left[-\sqrt{u_0} + \sqrt{-2p - u_0 + \frac{2q}{\sqrt{u_0}}}\right] - 7\sqrt{|\lambda_{11}|^2 + \mu_{11}\nu_{11}} + \frac{49\lambda_{12}\lambda_{21} + 7\mu_{12}\nu_{21}}{\varepsilon_1^{(0)} - \varepsilon_2^{(0)}} + \frac{49\lambda_{13}\lambda_{31} + 7\mu_{13}\nu_{31}}{\varepsilon_1^{(0)} - \varepsilon_3^{(0)}} + \frac{49\lambda_{14}\lambda_{41} + 7\mu_{14}\nu_{41}}{\varepsilon_1^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{+\frac{1}{2},+\frac{7}{2}} = \frac{1}{2}\left[\sqrt{u_0} + \sqrt{-2p - u_0 - \frac{2q}{\sqrt{u_0}}}\right] + 7\sqrt{|\lambda_{22}|^2 + \mu_{22}\nu_{22}} + \frac{49\lambda_{21}\lambda_{12} + 7\mu_{21}\nu_{12}}{\varepsilon_2^{(0)} - \varepsilon_1^{(0)}} + \frac{49\lambda_{23}\lambda_{32} + 7\mu_{23}\nu_{32}}{\varepsilon_2^{(0)} - \varepsilon_3^{(0)}} + \frac{49\lambda_{24}\lambda_{42} + 7\mu_{24}\nu_{42}}{\varepsilon_2^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{+\frac{1}{2},+\frac{5}{2}} = \frac{1}{2}\left[\sqrt{u_0} + \sqrt{-2p - u_0 - \frac{2q}{\sqrt{u_0}}}\right] + 5\sqrt{|\lambda_{22}|^2 + \mu_{22}\nu_{22}} + \frac{25\lambda_{21}\lambda_{12} + 19\mu_{21}\nu_{12}}{\varepsilon_2^{(0)} - \varepsilon_1^{(0)}} + \frac{25\lambda_{23}\lambda_{32} + 19\mu_{23}\nu_{32}}{\varepsilon_2^{(0)} - \varepsilon_3^{(0)}} + \frac{25\lambda_{24}\lambda_{42} + 19\mu_{24}\nu_{42}}{\varepsilon_2^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{+\frac{1}{2},+\frac{3}{2}} = \frac{1}{2}\left[\sqrt{u_0} + \sqrt{-2p - u_0 - \frac{2q}{\sqrt{u_0}}}\right] + 3\sqrt{|\lambda_{22}|^2 + \mu_{22}\nu_{22}} + \frac{9\lambda_{21}\lambda_{12} + 27\mu_{21}\nu_{12}}{\varepsilon_2^{(0)} - \varepsilon_1^{(0)}}$$

$$+ \frac{9\lambda_{23}\lambda_{32} + 27\mu_{23}\nu_{32}}{\varepsilon_2^{(0)} - \varepsilon_3^{(0)}} + \frac{9\lambda_{24}\lambda_{42} + 27\mu_{24}\nu_{42}}{\varepsilon_2^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{+\frac{1}{2},+\frac{1}{2}} = \frac{1}{2}\left[\sqrt{u_0} + \sqrt{-2p - u_0 - \frac{2q}{\sqrt{u_0}}}\right] + \sqrt{|\lambda_{22}|^2 + \mu_{22}\nu_{22}} + \frac{\lambda_{21}\lambda_{12} + 31\mu_{21}\nu_{12}}{\varepsilon_2^{(0)} - \varepsilon_1^{(0)}} + \frac{\lambda_{23}\lambda_{32} + 31\mu_{23}\nu_{32}}{\varepsilon_2^{(0)} - \varepsilon_3^{(0)}}$$

$$+ \frac{\lambda_{24}\lambda_{42} + 31\mu_{24}\nu_{42}}{\varepsilon_2^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{+\frac{1}{2},-\frac{1}{2}} = \frac{1}{2}\left[\sqrt{u_0} + \sqrt{-2p - u_0 - \frac{2q}{\sqrt{u_0}}}\right] - \sqrt{|\lambda_{22}|^2 + \mu_{22}\nu_{22}} + \frac{\lambda_{21}\lambda_{12} + 31\mu_{21}\nu_{12}}{\varepsilon_2^{(0)} - \varepsilon_1^{(0)}} + \frac{\lambda_{23}\lambda_{32} + 31\mu_{23}\nu_{32}}{\varepsilon_2^{(0)} - \varepsilon_3^{(0)}}$$

$$+ \frac{\lambda_{24}\lambda_{42} + 31\mu_{24}\nu_{42}}{\varepsilon_2^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{+\frac{1}{2},-\frac{3}{2}} = \frac{1}{2}\left[\sqrt{u_0} + \sqrt{-2p - u_0 - \frac{2q}{\sqrt{u_0}}}\right] - 3\sqrt{|\lambda_{22}|^2 + \mu_{22}\nu_{22}} + \frac{9\lambda_{21}\lambda_{12} + 27\mu_{21}\nu_{12}}{\varepsilon_2^{(0)} - \varepsilon_1^{(0)}}$$

$$+ \frac{9\lambda_{23}\lambda_{32} + 27\mu_{23}\nu_{32}}{\varepsilon_2^{(0)} - \varepsilon_3^{(0)}} + \frac{9\lambda_{24}\lambda_{42} + 27\mu_{24}\nu_{42}}{\varepsilon_2^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{+\frac{1}{2},-\frac{5}{2}} = \frac{1}{2}\left[\sqrt{u_0} + \sqrt{-2p - u_0 - \frac{2q}{\sqrt{u_0}}}\right] - 5\sqrt{|\lambda_{22}|^2 + \mu_{22}\nu_{22}} + \frac{25\lambda_{21}\lambda_{12} + 19\mu_{21}\nu_{12}}{\varepsilon_2^{(0)} - \varepsilon_1^{(0)}}$$

$$+ \frac{25\lambda_{23}\lambda_{32} + 19\mu_{23}\nu_{32}}{\varepsilon_2^{(0)} - \varepsilon_3^{(0)}} + \frac{25\lambda_{24}\lambda_{42} + 19\mu_{24}\nu_{42}}{\varepsilon_2^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{+\frac{1}{2},-\frac{7}{2}} = \frac{1}{2}\left[\sqrt{u_0} + \sqrt{-2p - u_0 - \frac{2q}{\sqrt{u_0}}}\right] - 7\sqrt{|\lambda_{22}|^2 + \mu_{22}\nu_{22}} + \frac{49\lambda_{21}\lambda_{12} + 7\mu_{21}\nu_{12}}{\varepsilon_2^{(0)} - \varepsilon_1^{(0)}}$$

$$+ \frac{49\lambda_{23}\lambda_{32} + 7\mu_{23}\nu_{32}}{\varepsilon_2^{(0)} - \varepsilon_3^{(0)}} + \frac{49\lambda_{24}\lambda_{42} + 7\mu_{24}\nu_{42}}{\varepsilon_2^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{-\frac{1}{2},+\frac{7}{2}} = \frac{1}{2}\left[\sqrt{u_0} - \sqrt{-2p - u_0 - \frac{2q}{\sqrt{u_0}}}\right] - 7\sqrt{|\lambda_{33}|^2 + \mu_{33}\nu_{33}} + \frac{49\lambda_{31}\lambda_{13} + 7\mu_{31}\nu_{13}}{\varepsilon_3^{(0)} - \varepsilon_1^{(0)}}$$

$$+ \frac{49\lambda_{32}\lambda_{23} + 7\mu_{32}\nu_{23}}{\varepsilon_3^{(0)} - \varepsilon_2^{(0)}} + \frac{49\lambda_{34}\lambda_{43} + 7\mu_{34}\nu_{43}}{\varepsilon_3^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{-\frac{1}{2},+\frac{5}{2}} = \frac{1}{2}\left[\sqrt{u_0} - \sqrt{-2p - u_0 - \frac{2q}{\sqrt{u_0}}}\right] - 5\sqrt{|\lambda_{33}|^2 + \mu_{33}\nu_{33}} + \frac{25\lambda_{31}\lambda_{13} + 19\mu_{31}\nu_{13}}{\varepsilon_3^{(0)} - \varepsilon_1^{(0)}} + \frac{25\lambda_{32}\lambda_{23} + 19\mu_{32}\nu_{23}}{\varepsilon_3^{(0)} - \varepsilon_2^{(0)}} + \frac{25\lambda_{34}\lambda_{43} + 19\mu_{34}\nu_{43}}{\varepsilon_3^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{-\frac{1}{2},+\frac{3}{2}} = \frac{1}{2}\left[\sqrt{u_0} - \sqrt{-2p - u_0 - \frac{2q}{\sqrt{u_0}}}\right] - 3\sqrt{|\lambda_{33}|^2 + \mu_{33}\nu_{33}} + \frac{9\lambda_{31}\lambda_{13} + 27\mu_{31}\nu_{13}}{\varepsilon_3^{(0)} - \varepsilon_1^{(0)}} + \frac{9\lambda_{32}\lambda_{23} + 27\mu_{32}\nu_{23}}{\varepsilon_3^{(0)} - \varepsilon_2^{(0)}} + \frac{9\lambda_{34}\lambda_{43} + 27\mu_{34}\nu_{43}}{\varepsilon_3^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{-\frac{1}{2},+\frac{1}{2}} = \frac{1}{2}\left[\sqrt{u_0} - \sqrt{-2p - u_0 - \frac{2q}{\sqrt{u_0}}}\right] - \sqrt{|\lambda_{33}|^2 + \mu_{33}\nu_{33}} + \frac{\lambda_{31}\lambda_{13} + 31\mu_{31}\nu_{13}}{\varepsilon_3^{(0)} - \varepsilon_1^{(0)}} + \frac{\lambda_{32}\lambda_{23} + 31\mu_{32}\nu_{23}}{\varepsilon_3^{(0)} - \varepsilon_2^{(0)}} + \frac{\lambda_{34}\lambda_{43} + 31\mu_{34}\nu_{43}}{\varepsilon_3^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{-\frac{1}{2},-\frac{1}{2}} = \frac{1}{2}\left[\sqrt{u_0} - \sqrt{-2p - u_0 - \frac{2q}{\sqrt{u_0}}}\right] + \sqrt{|\lambda_{33}|^2 + \mu_{33}\nu_{33}} + \frac{\lambda_{31}\lambda_{13} + 31\mu_{31}\nu_{13}}{\varepsilon_3^{(0)} - \varepsilon_1^{(0)}} + \frac{\lambda_{32}\lambda_{23} + 31\mu_{32}\nu_{23}}{\varepsilon_3^{(0)} - \varepsilon_2^{(0)}} + \frac{\lambda_{34}\lambda_{43} + 31\mu_{34}\nu_{43}}{\varepsilon_3^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{-\frac{1}{2},-\frac{3}{2}} = \frac{1}{2}\left[\sqrt{u_0} - \sqrt{-2p - u_0 - \frac{2q}{\sqrt{u_0}}}\right] + 3\sqrt{|\lambda_{33}|^2 + \mu_{33}\nu_{33}} + \frac{9\lambda_{31}\lambda_{13} + 27\mu_{31}\nu_{13}}{\varepsilon_3^{(0)} - \varepsilon_1^{(0)}} + \frac{9\lambda_{32}\lambda_{23} + 27\mu_{32}\nu_{23}}{\varepsilon_3^{(0)} - \varepsilon_2^{(0)}} + \frac{9\lambda_{34}\lambda_{43} + 27\mu_{34}\nu_{43}}{\varepsilon_3^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{-\frac{1}{2},-\frac{5}{2}} = \frac{1}{2}\left[\sqrt{u_0} - \sqrt{-2p - u_0 - \frac{2q}{\sqrt{u_0}}}\right] + 5\sqrt{|\lambda_{33}|^2 + \mu_{33}\nu_{33}} + \frac{25\lambda_{31}\lambda_{13} + 19\mu_{31}\nu_{13}}{\varepsilon_3^{(0)} - \varepsilon_1^{(0)}} + \frac{25\lambda_{32}\lambda_{23} + 19\mu_{32}\nu_{23}}{\varepsilon_3^{(0)} - \varepsilon_2^{(0)}} + \frac{25\lambda_{34}\lambda_{43} + 19\mu_{34}\nu_{43}}{\varepsilon_3^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{-\frac{1}{2},-\frac{7}{2}} = \frac{1}{2}\left[\sqrt{u_0} - \sqrt{-2p - u_0 - \frac{2q}{\sqrt{u_0}}}\right] + 7\sqrt{|\lambda_{33}|^2 + \mu_{33}\nu_{33}} + \frac{49\lambda_{31}\lambda_{13} + 7\mu_{31}\nu_{13}}{\varepsilon_3^{(0)} - \varepsilon_1^{(0)}} + \frac{49\lambda_{32}\lambda_{23} + 7\mu_{32}\nu_{23}}{\varepsilon_3^{(0)} - \varepsilon_2^{(0)}} + \frac{49\lambda_{34}\lambda_{43} + 7\mu_{34}\nu_{43}}{\varepsilon_3^{(0)} - \varepsilon_4^{(0)}}$$

$$E_{-\frac{3}{2},+\frac{7}{2}}=\frac{1}{2}\left[-\sqrt{u_0}-\sqrt{-2p-u_0+\frac{2q}{\sqrt{u_0}}}\right]-7\sqrt{|\lambda_{44}|^2+\mu_{44}\nu_{44}}+\frac{49\lambda_{41}\lambda_{14}+7\mu_{41}\nu_{14}}{\varepsilon_4^{(0)}-\varepsilon_1^{(0)}}$$

$$+\frac{49\lambda_{42}\lambda_{24}+7\mu_{42}\nu_{24}}{\varepsilon_4^{(0)}-\varepsilon_2^{(0)}}+\frac{49\lambda_{43}\lambda_{34}+7\mu_{43}\nu_{34}}{\varepsilon_4^{(0)}-\varepsilon_3^{(0)}}$$

$$E_{-\frac{3}{2},+\frac{5}{2}}=\frac{1}{2}\left[-\sqrt{u_0}-\sqrt{-2p-u_0+\frac{2q}{\sqrt{u_0}}}\right]-5\sqrt{|\lambda_{44}|^2+\mu_{44}\nu_{44}}+\frac{25\lambda_{41}\lambda_{14}+19\mu_{41}\nu_{14}}{\varepsilon_4^{(0)}-\varepsilon_1^{(0)}}$$

$$+\frac{25\lambda_{42}\lambda_{24}+19\mu_{42}\nu_{24}}{\varepsilon_4^{(0)}-\varepsilon_2^{(0)}}+\frac{25\lambda_{43}\lambda_{34}+19\mu_{43}\nu_{34}}{\varepsilon_4^{(0)}-\varepsilon_3^{(0)}}$$

$$E_{-\frac{3}{2},+\frac{3}{2}}=\frac{1}{2}\left[-\sqrt{u_0}-\sqrt{-2p-u_0+\frac{2q}{\sqrt{u_0}}}\right]-3\sqrt{|\lambda_{44}|^2+\mu_{44}\nu_{44}}+\frac{9\lambda_{41}\lambda_{14}+27\mu_{41}\nu_{14}}{\varepsilon_4^{(0)}-\varepsilon_1^{(0)}}$$

$$+\frac{9\lambda_{42}\lambda_{24}+27\mu_{42}\nu_{24}}{\varepsilon_4^{(0)}-\varepsilon_2^{(0)}}+\frac{9\lambda_{43}\lambda_{34}+27\mu_{43}\nu_{34}}{\varepsilon_4^{(0)}-\varepsilon_3^{(0)}}$$

$$E_{-\frac{3}{2},+\frac{1}{2}}=\frac{1}{2}\left[-\sqrt{u_0}-\sqrt{-2p-u_0+\frac{2q}{\sqrt{u_0}}}\right]-\sqrt{|\lambda_{44}|^2+\mu_{44}\nu_{44}}+\frac{\lambda_{41}\lambda_{14}+31\mu_{41}\nu_{14}}{\varepsilon_4^{(0)}-\varepsilon_1^{(0)}}$$

$$+\frac{\lambda_{42}\lambda_{24}+31\mu_{42}\nu_{24}}{\varepsilon_4^{(0)}-\varepsilon_2^{(0)}}+\frac{\lambda_{43}\lambda_{34}+31\mu_{43}\nu_{34}}{\varepsilon_4^{(0)}-\varepsilon_3^{(0)}}$$

$$E_{-\frac{3}{2},-\frac{1}{2}}=\frac{1}{2}\left[-\sqrt{u_0}-\sqrt{-2p-u_0+\frac{2q}{\sqrt{u_0}}}\right]+\sqrt{|\lambda_{44}|^2+\mu_{44}\nu_{44}}+\frac{\lambda_{41}\lambda_{14}+31\mu_{41}\nu_{14}}{\varepsilon_4^{(0)}-\varepsilon_1^{(0)}}$$

$$+\frac{\lambda_{42}\lambda_{24}+31\mu_{42}\nu_{24}}{\varepsilon_4^{(0)}-\varepsilon_2^{(0)}}+\frac{\lambda_{43}\lambda_{34}+31\mu_{43}\nu_{34}}{\varepsilon_4^{(0)}-\varepsilon_3^{(0)}}$$

$$E_{-\frac{3}{2},-\frac{3}{2}}=\frac{1}{2}\left[-\sqrt{u_0}-\sqrt{-2p-u_0+\frac{2q}{\sqrt{u_0}}}\right]+3\sqrt{|\lambda_{44}|^2+\mu_{44}\nu_{44}}+\frac{9\lambda_{41}\lambda_{14}+27\mu_{41}\nu_{14}}{\varepsilon_4^{(0)}-\varepsilon_1^{(0)}}$$

$$+\frac{9\lambda_{42}\lambda_{24}+27\mu_{42}\nu_{24}}{\varepsilon_4^{(0)}-\varepsilon_2^{(0)}}+\frac{9\lambda_{43}\lambda_{34}+27\mu_{43}\nu_{34}}{\varepsilon_4^{(0)}-\varepsilon_3^{(0)}}$$

$$E_{-\frac{3}{2},-\frac{5}{2}}=\frac{1}{2}\left[-\sqrt{u_0}-\sqrt{-2p-u_0+\frac{2q}{\sqrt{u_0}}}\right]+5\sqrt{|\lambda_{44}|^2+\mu_{44}\nu_{44}}+\frac{25\lambda_{41}\lambda_{14}+19\mu_{41}\nu_{14}}{\varepsilon_4^{(0)}-\varepsilon_1^{(0)}}$$

$$+\frac{25\lambda_{42}\lambda_{24}+19\mu_{42}\nu_{24}}{\varepsilon_4^{(0)}-\varepsilon_2^{(0)}}+\frac{25\lambda_{43}\lambda_{34}+19\mu_{43}\nu_{34}}{\varepsilon_4^{(0)}-\varepsilon_3^{(0)}}$$

$$E_{-\frac{3}{2},-\frac{7}{2}} = \frac{1}{2}\left[-\sqrt{u_0} - \sqrt{-2p - u_0 + \frac{2q}{\sqrt{u_0}}}\right] + 7\sqrt{|\lambda_{44}|^2 + \mu_{44}\nu_{44}} + \frac{49\lambda_{41}\lambda_{14} + 7\mu_{41}\nu_{14}}{\varepsilon_4^{(0)} - \varepsilon_1^{(0)}}$$

$$+ \frac{49\lambda_{42}\lambda_{24} + 7\mu_{42}\nu_{24}}{\varepsilon_4^{(0)} - \varepsilon_2^{(0)}} + \frac{49\lambda_{43}\lambda_{34} + 7\mu_{43}\nu_{34}}{\varepsilon_4^{(0)} - \varepsilon_3^{(0)}}$$

It is worth noting that when at least two of the ZFS, **g-** and **A**-tensors are collinear and/or the magnetic field is aligned to the principal axis the energy representations are much simpler.

Summary and Chart of Procedure for ESR Analyses of High-Spin Metallocomplexes

Step 0: Reproduce the experimental ESR spectra by using the fictitious spin-1/2 Hamiltonian (if needed), noting that not all the principal values of the $\mathbf{A}^{\mathrm{eff}}$-tensor may be determined.

Step 1: Determine the principal values of the $\mathbf{g}^{\mathrm{eff}}$- and $\mathbf{A}^{\mathrm{eff}}$-tensors with relative coordination between the tensors taken from the quantum chemical calculation.

Step 2: Calculate the principal values of the $\mathbf{g}^{\mathrm{true}}$-tensor as a function of the E/D value by using the $\mathbf{g}^{\mathrm{eff}}/\mathbf{g}^{\mathrm{true}}$ relationships if the ZFS and $\mathbf{g}^{\mathrm{true}}$-tensors are collinear.

Step 2': If the ZFS and $\mathbf{g}^{\mathrm{true}}$-tensors are non-collinear, evaluate the principal values of the $\mathbf{g}^{\mathrm{true}}$-tensor by solving the exact analytical energies or spectral simulation by using the full spin-Hamiltonian for variable E/D values.

Step 3: Calculate the principal values of the $\mathbf{A}^{\mathrm{true}}$-tensor by using the $\mathbf{A}^{\mathrm{eff}}/\mathbf{A}^{\mathrm{true}}$ relationships.

Step 0: Construct the experimental spectra in order to compare with the simulated one on screen.

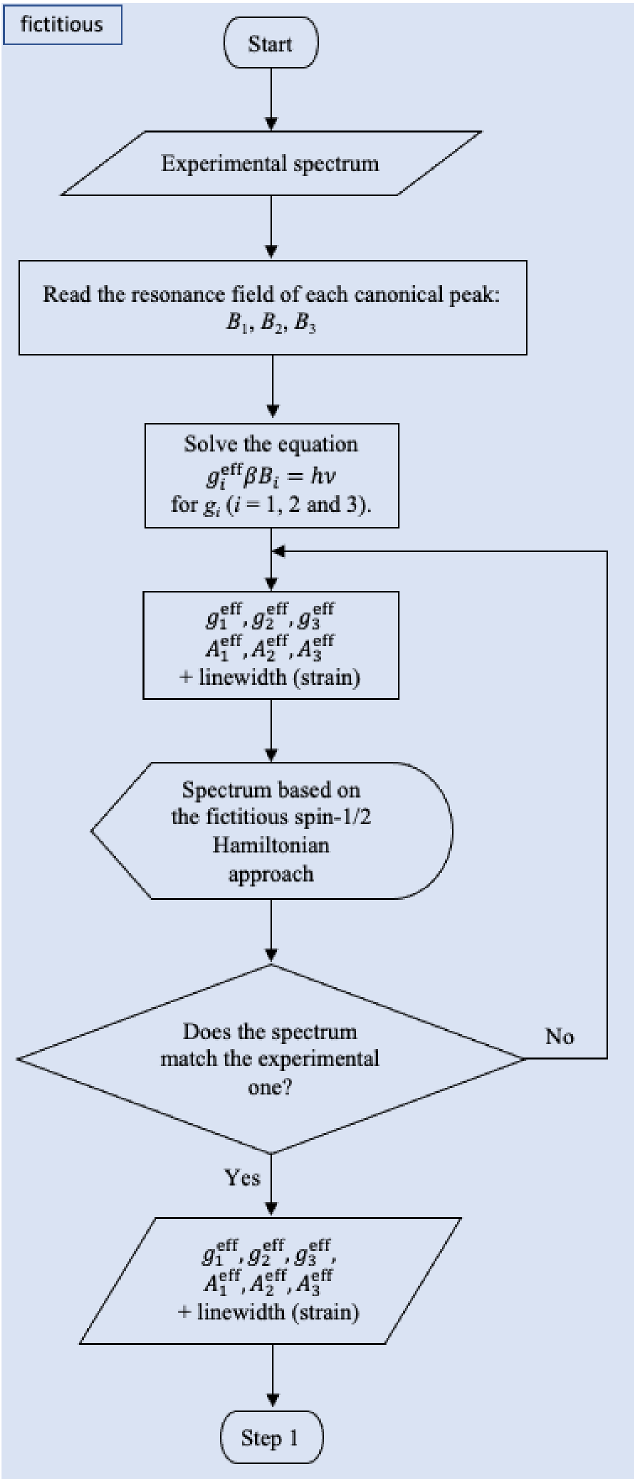

Chart S1. Flow chart of the procedure for reproducing the experimental ESR spectra by using the fictitious spin-1/2 Hamiltonian. The background in blue indicates that the fictitious spin-1/2 Hamiltonian is applied.

Step 1. Introduce the result obtained by the quantum chemical calculations to the effective magnetic tensors (relevant to the fictitious spin-1/2 Hamiltonian approach)

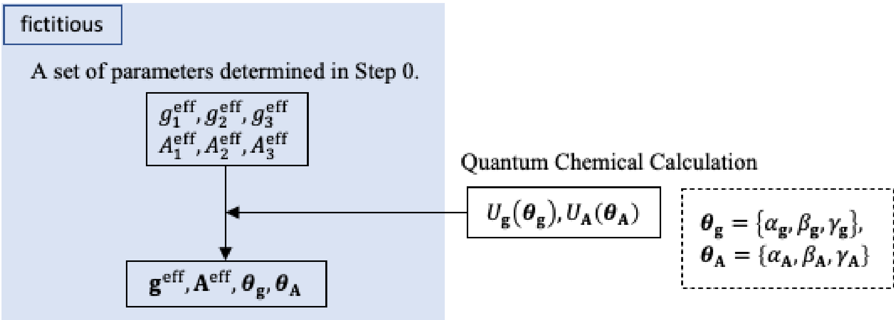

Chart S2. α_{T}, β_{T}, γ_{T} (**T** = **g** or **A**) are the Euler's angle with respect to the molecular principal axis coordination system and $U_{\mathrm{T}}(\boldsymbol{\theta}_{\mathrm{T}})$ is the unitary matrix transforming the $\mathbf{g}^{\mathrm{eff}}$ or $\mathbf{A}^{\mathrm{eff}}$-tensor.

Step 2: Transform the principal values of the $\mathbf{g}^{\mathrm{eff}}$-tensor to those of the $\mathbf{g}^{\mathrm{true}}$-tensor in case of the collinearity between the ZFS and g-tensors.

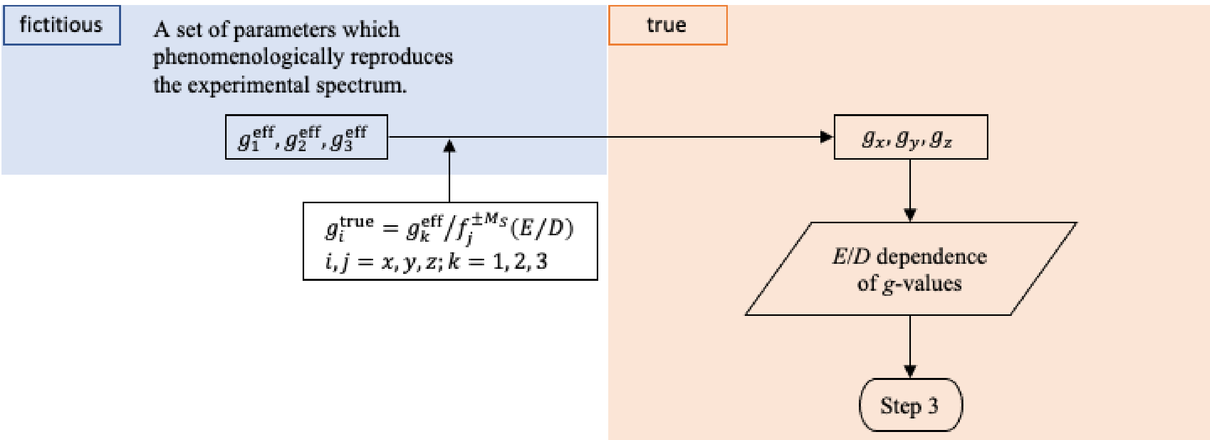

Chart S3. $f_j^{\pm M_S}$ indicates a function of the E/D value used for transforming the g^{eff} values to the g^{true} values. For example, for $S = 3/2$ and for the magnetic resonance transition between the states $|M_S = \pm 3/2\rangle$-dominant transition, $f_x^{\pm\frac{3}{2}}(\lambda) = 1 - \frac{1-3\lambda}{\sqrt{1+3\lambda^2}}$, $f_y^{\pm\frac{3}{2}}(\lambda) = 1 - \frac{1+3\lambda}{\sqrt{1+3\lambda^2}}$ and $f_z^{\pm\frac{3}{2}}(\lambda) = 1 + \frac{2}{\sqrt{1+3\lambda^2}}$ with $\lambda = E/D$. The background in orange indicates that the true spin Hamiltonian is applied.

Step 2': Transform the principal values of the g^{eff}-tensor to those of the g^{true}-tensor in case of the non-collinearity between the ZFS and g-tensors.

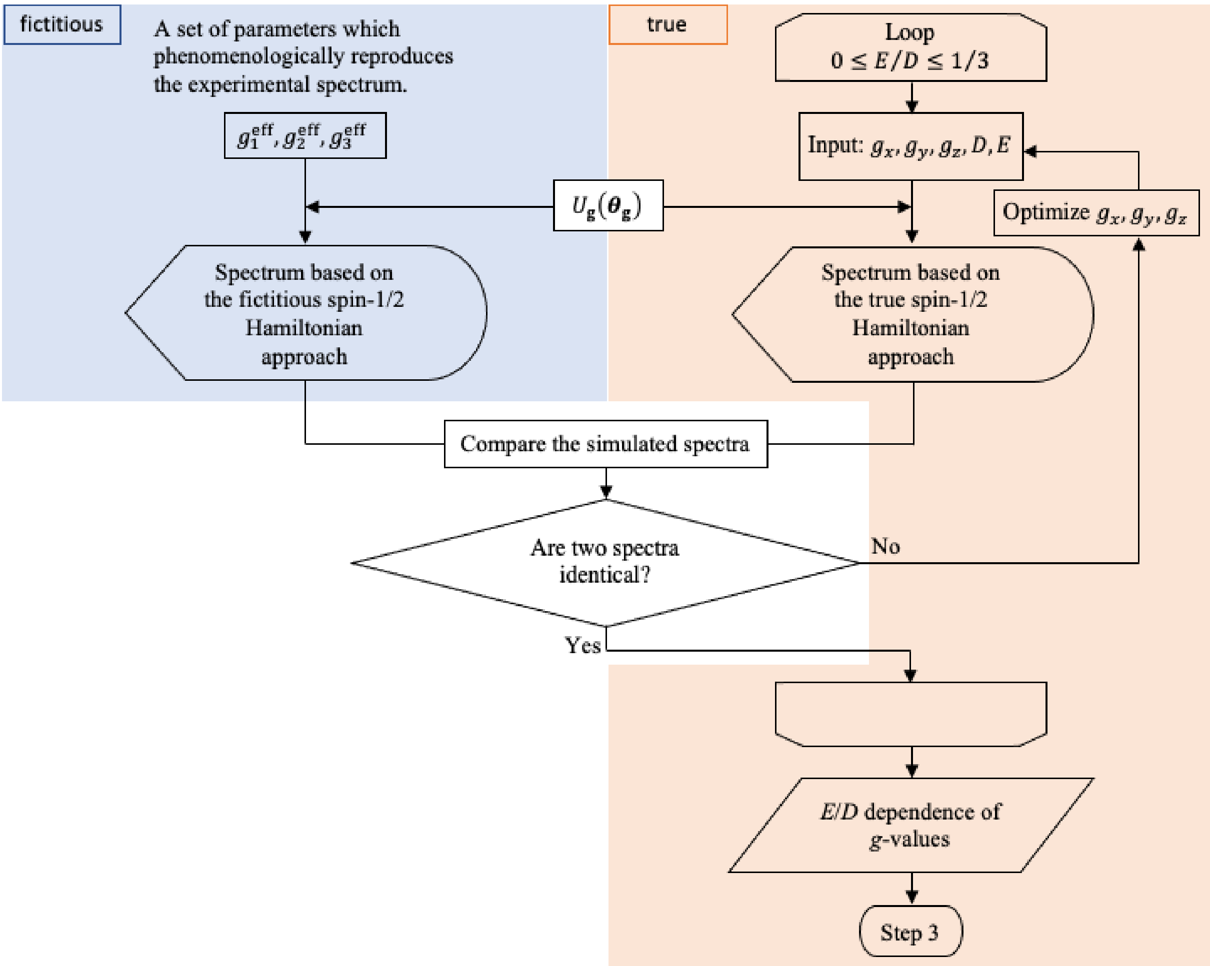

Chart S4. Flow chart of the procedure for obtaining the set of the E/D value and the principal value of the **g**-tensor in the case of the non-collinearity between the ZFS and **g**-tensors.

Step 3: Estimate all the sets of the magnetic parameters and evaluate the accuracy of the principal *A*-values

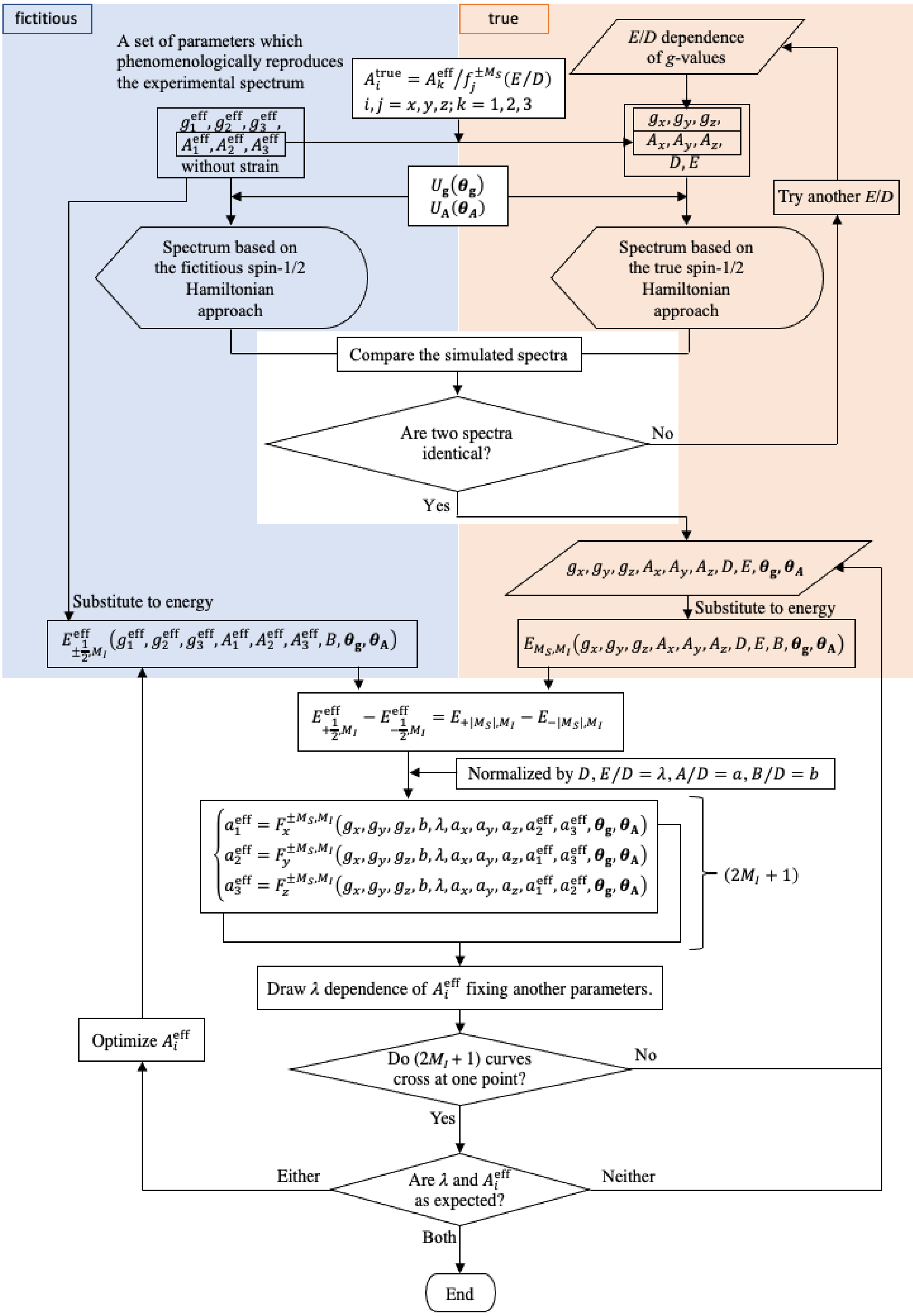

Chart S5. Flow chart of the procedure for obtaining the full set of the magnetic parameters. $E^{\mathrm{eff}}_{\pm\frac{1}{2},M_I}$ and E_{M_S,M_I} denote the analytical energy from the fictitious spin-1/2 and the full spin-Hamiltonian, respectively. In the parameter of the energy, B denotes the resonance field.

There are some remarks relevant to the procedure described above.

1. As mentioned before, not all the principal values of the **A**-tensor may be determined due to the line-broadening appearing in the experimental spectra. Indeed, only the A_Z-value was determined in the complexes under study.
2. Generally, solving the A^{eff}-values is easier than for the A^{true}-values (step 3). Fortunately, the E/D dependence of the A^{true}-values can be obtained for the complexes under study.
3. The possible range of the E/D value can be limited because of the principal values of the **g**-tensor and/or the off-principal-axis extra lines frequently observed in the experimental spectra from high spin systems having sizable ZFS tensors.
4. The D-value can be evaluated from the intensities of small transitions between the other spin substates except the large canonical peaks. For example, both the $|M_S = \pm 3/2\rangle$- and $|M_S = \pm 1/2\rangle$-dominant transitions were observed in the spectra under study, enabling us to determine the D-values.

Applications of the Perturbation Theory to ESR Analyses of Five-Coordinated Cobalt Complexes in Their Quartet Spin State

1. Complex 1

a) Perturbed energies for the case of collinear ZFS, **g-** and **A-**tensors

Here, we derive perturbed energies, which arise only from a hyperfine **A**-tensor ($I = 7/2$), in order to estimate the accuracy of the hyperfine parameters in a quartet spin system ($S = 3/2$) having a sizeable ZFS tensor such as five-coordinated cobalt complexes. Thus, first we solve the problem of the eigenvalue and eigenfunction of the spin Hamiltonian composed of only electron spins in the presence of an external static magnetic field. The approach here for the spin quartet state is not a simple Zeeman perturbation one. The ZFS and electron Zeeman interactions are taken as the non-perturbed term H_0 and the hyperfine ~~splitting~~ interaction is taken as the perturbed term H'. The non-perturbed and perturbed terms are represented as follows.

$$H_0 = \boldsymbol{S} \cdot \mathbf{D} \cdot \boldsymbol{S} + \beta \boldsymbol{S} \cdot \mathbf{g} \cdot \boldsymbol{B}$$
$$H' = \boldsymbol{S} \cdot \mathbf{A} \cdot \boldsymbol{I}$$

For simplicity, the ZFS, **g-** and **A-**tensors are assumed to be collinear. The non-perturbed Hamiltonian H_0 can be rewritten when the static magnetic field $\boldsymbol{B}$ is applied parallel to the z-axis of the principal axis coordinate system of the complex, where the z-axis is the principal z-axis of the ZFS tensor, as follows:

$$H_0 = D\left[{S_z}^2 - \frac{1}{3}S(S+1)\right] + E\left({S_x}^2 - {S_y}^2\right) + g_{zz}\beta B_z S_z$$
$$H' = S_x A_{xx} I_x + S_y A_{yy} I_y + S_z A_{zz} I_z$$

The matrix representation of H_0 on the basis of $|M_S>$ is

$$H_0 = \begin{pmatrix} D + \frac{3}{2}g_{zz}\beta B & 0 & \sqrt{3}E & 0 \\ 0 & -D + \frac{1}{2}g_{zz}\beta B & 0 & \sqrt{3}E \\ \sqrt{3}E & 0 & -D - \frac{1}{2}g_{zz}\beta B & 0 \\ 0 & \sqrt{3}E & 0 & D - \frac{3}{2}g_{zz}\beta B \end{pmatrix}$$

This matrix can be divided into two 2 × 2 matrixes H_1 and H_2 due to the symmetry of the spin functions.

$$H_1 = \begin{pmatrix} D + \frac{3}{2}g_{zz}\beta B & \sqrt{3}E \\ \sqrt{3}E & -D - \frac{1}{2}g_{zz}\beta B \end{pmatrix}$$

$$H_2 = \begin{pmatrix} -D + \frac{1}{2}g_{zz}\beta B & \sqrt{3}E \\ \sqrt{3}E & D - \frac{3}{2}g_{zz}\beta B \end{pmatrix}$$

Thus, the exact energy eigenvalues and the spin eigenfunctions are

$$\varepsilon_{\pm\frac{3}{2}} = \pm\frac{1}{2}g_{zz}\beta B - \sqrt{(D \pm g_{zz}\beta B)^2 + 3E^2}$$

$$\varepsilon_{\pm\frac{1}{2}} = \mp\frac{1}{2}g_{zz}\beta B + \sqrt{(D \mp g_{zz}\beta B)^2 + 3E^2}$$

$$\left|\psi_{\pm\frac{3}{2}}\right\rangle = \cos\theta_\pm \left|\pm\frac{3}{2}\right\rangle + \sin\theta_\pm \left|\mp\frac{1}{2}\right\rangle$$

$$\left|\psi_{\pm\frac{1}{2}}\right\rangle = \cos\theta_\mp \left|\pm\frac{1}{2}\right\rangle - \sin\theta_\mp \left|\mp\frac{3}{2}\right\rangle$$

where

$$\tan 2\theta_\pm = \frac{\sqrt{3}E}{D \pm g_{zz}\beta B}$$

We construct electron-nuclear spin wavefunctions as follows:

$$\left|\psi_{\pm\frac{3}{2}}(M_I)\right\rangle \equiv \left|\psi_{\pm\frac{3}{2}}\right\rangle \otimes |M_I\rangle = \cos\theta_\pm \left|\pm\frac{3}{2}, M_I\right\rangle + \sin\theta_\pm \left|\mp\frac{1}{2}, M_I\right\rangle$$

$$\left|\psi_{\pm\frac{1}{2}}(M_I)\right\rangle \equiv \left|\psi_{\pm\frac{1}{2}}\right\rangle \otimes |M_I\rangle = \cos\theta_\mp \left|\pm\frac{1}{2}, M_I\right\rangle - \sin\theta_\mp \left|\mp\frac{3}{2}, M_I\right\rangle$$

The matrix elements of the hyperfine structure Hamiltonian in the basis of $|M_S, M_I\rangle$ are given in the following:

$$\langle M_S', M_I'|H_{\mathrm{hfs}}|M_S, M_I\rangle = \begin{cases} \delta_{M_S M_S'}\delta_{M_I M_I'} M_S M_I A_{zz} \\ \frac{1}{16}\delta_{M_S M_S'\mp 1}\delta_{M_I M_I'\mp 1}\sqrt{15 - 4M_S M_S'}\sqrt{63 - 4M_I M_I'}\left(A_{xx} - A_{yy}\right) \\ \frac{1}{16}\delta_{M_S M_S'\mp 1}\delta_{M_I M_I'\pm 1}\sqrt{15 - 4M_S M_S'}\sqrt{63 - 4M_I M_I'}\left(A_{xx} + A_{yy}\right) \end{cases}$$

where

$$\delta_{ij} = \begin{cases} 1, & i = j \\ 0, & i \neq j \end{cases}$$

The upper and lower signs should be chosen in the double sign. The matrix representation of the hyperfine structure Hamiltonian expanded by the spin wavefunctions are as follows:

$$\left\langle \psi_{M_S=\pm\frac{3}{2}}(M_I') \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=\pm\frac{3}{2}}(M_I) \right\rangle = \pm\frac{M_I}{2}\delta_{M_I M_I'} A_{zz}(2\cos 2\theta_\pm + 1)$$

$$\left\langle \psi_{M_S=\pm\frac{1}{2}}(M_I') \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=\pm\frac{1}{2}}(M_I) \right\rangle = \pm\frac{M_I}{2}\delta_{M_I M_I'} A_{zz}(2\cos 2\theta_\mp - 1)$$

$$\left\langle \psi_{M_S=+\frac{1}{2}}(M_I') \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=+\frac{3}{2}}(M_I) \right\rangle = \frac{1}{2}\sqrt{63 - 4M_I M_I'}\,\delta_{M_I M_I'\pm 1}\left(C_{+sc}A_{xx} \mp C_{-sc}A_{yy}\right)$$

$$\left\langle \psi_{M_S=-\frac{1}{2}}(M_I') \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=+\frac{3}{2}}(M_I) \right\rangle = -M_I \delta_{M_I M_I'} A_{zz} \sin 2\theta_+$$

$$\left\langle \psi_{M_S=-\frac{3}{2}}(M_I') \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=+\frac{3}{2}}(M_I) \right\rangle = \frac{1}{2}\sqrt{63 - 4M_I M_I'}\,\delta_{M_I M_I'\pm 1}\left(S_{+ss}A_{xx} \mp S_{-ss}A_{yy}\right)$$

$$\left\langle \psi_{M_S=+\frac{3}{2}}(M_I') \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=+\frac{1}{2}}(M_I) \right\rangle = \left\langle \psi_{M_S=+\frac{1}{2}}(M_I) \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=+\frac{3}{2}}(M_I') \right\rangle$$

$$\left\langle \psi_{M_S=-\frac{1}{2}}(M_I') \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=+\frac{1}{2}}(M_I) \right\rangle = \frac{1}{2}\sqrt{63 - 4M_I M_I'}\,\delta_{M_I M_I'\pm 1}\left(S_{+cc}A_{xx} \pm S_{-cc}A_{yy}\right)$$

$$\left\langle \psi_{M_S=-\frac{3}{2}}(M_I') \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=+\frac{1}{2}}(M_I) \right\rangle = M_I \delta_{M_I M_I'} A_{zz} \sin 2\theta_-$$

$$\left\langle \psi_{M_S=+\frac{3}{2}}(M_I') \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=-\frac{1}{2}}(M_I) \right\rangle = -M_I \delta_{M_I M_I'} A_{zz} \sin 2\theta_+$$

$$\left\langle \psi_{M_S=+\frac{1}{2}}(M_I') \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=-\frac{1}{2}}(M_I) \right\rangle = \left\langle \psi_{M_S=-\frac{1}{2}}(M_I) \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=+\frac{1}{2}}(M_I') \right\rangle$$

$$\left\langle \psi_{M_S=-\frac{3}{2}}(M_I') \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=-\frac{1}{2}}(M_I) \right\rangle = \frac{1}{2}\sqrt{63 - 4M_I M_I'}\, \delta_{M_I M_I' \pm 1} \left(C_{+cs} A_{xx} \mp C_{-cs} A_{yy} \right)$$

$$\left\langle \psi_{M_S=+\frac{3}{2}}(M_I') \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=-\frac{3}{2}}(M_I) \right\rangle = \left\langle \psi_{M_S=-\frac{3}{2}}(M_I) \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=+\frac{3}{2}}(M_I') \right\rangle$$

$$\left\langle \psi_{M_S=+\frac{1}{2}}(M_I') \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=-\frac{3}{2}}(M_I) \right\rangle = M_I \delta_{M_I M_I'} A_{zz} \sin 2\theta_-$$

$$\left\langle \psi_{M_S=-\frac{1}{2}}(M_I') \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=-\frac{3}{2}}(M_I) \right\rangle = \left\langle \psi_{M_S=-\frac{3}{2}}(M_I) \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=-\frac{1}{2}}(M_I') \right\rangle$$

where

$$C_{\pm sc} = \frac{1}{4}\left(\sqrt{3}\cos\theta_+ \cos\theta_- \pm 2\sin\theta_+ \cos\theta_- - \sqrt{3}\sin\theta_+ \sin\theta_- \right)$$

$$S_{\pm ss} = \frac{1}{4}\left(\sqrt{3}\cos\theta_+ \sin\theta_- \pm 2\sin\theta_+ \sin\theta_- + \sqrt{3}\sin\theta_+ \cos\theta_- \right)$$

$$S_{\pm cc} = \frac{1}{4}\left(-\sqrt{3}\cos\theta_+ \sin\theta_- \pm 2\cos\theta_+ \cos\theta_- - \sqrt{3}\sin\theta_+ \cos\theta_- \right)$$

$$C_{\pm cs} = \frac{1}{4}\left(-\sqrt{3}\sin\theta_+ \sin\theta_- \pm 2\cos\theta_+ \sin\theta_- + \sqrt{3}\cos\theta_+ \cos\theta_- \right)$$

The upper and lower signs should be chosen in the double sign. We can evaluate the zeroth-order and perturbed energies in the following:

The zeroth-order energies are the energy eigenvalues of the non-perturbed Hamiltonian.

$$\varepsilon^{(0)}_{\pm\frac{3}{2}} = \pm\frac{1}{2} g_{zz}\beta B - \sqrt{(D \pm g_{zz}\beta B)^2 + 3E^2}$$

$$\varepsilon^{(0)}_{\pm\frac{1}{2}} = \mp\frac{1}{2} g_{zz}\beta B + \sqrt{(D \mp g_{zz}\beta B)^2 + 3E^2}$$

The first-order energies are the diagonal elements of the expanded hyperfine Hamiltonian, as given in the following:

$$\varepsilon^{(1)}_{\pm\frac{3}{2},M_I} = \pm\frac{M_I}{2} A_{zz}(2\cos 2\theta_\pm + 1)$$

$$\varepsilon^{(1)}_{\pm\frac{1}{2},M_I} = \pm\frac{M_I}{2} A_{zz}(2\cos 2\theta_\mp - 1)$$

The second-order energies are given in a convoluted manner, as follows;

$$\varepsilon^{(2)}_{+\frac{3}{2},+\frac{7}{2}} = \frac{7\left(C_{+sc}A_{xx} - C_{-sc}A_{yy}\right)^2}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{49}{4}A_{zz}{}^2 \sin^2 2\theta_+}{-2\sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{7\left(S_{+ss}A_{xx} - S_{-ss}A_{yy}\right)^2}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},+\frac{5}{2}} = \frac{7\left(C_{+sc}A_{xx} + C_{-sc}A_{yy}\right)^2 + 12\left(C_{+sc}A_{xx} - C_{-sc}A_{yy}\right)^2}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{25}{4}A_{zz}{}^2 \sin^2 2\theta_+}{-2\sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{7\left(S_{+ss}A_{xx} + S_{-ss}A_{yy}\right)^2 + 12\left(S_{+ss}A_{xx} - S_{-ss}A_{yy}\right)^2}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},+\frac{3}{2}} = \frac{12\left(C_{+sc}A_{xx} + C_{-sc}A_{yy}\right)^2 + 15\left(C_{+sc}A_{xx} - C_{-sc}A_{yy}\right)^2}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{9}{4}A_{zz}{}^2 \sin^2 2\theta_+}{-2\sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{12\left(S_{+ss}A_{xx} + S_{-ss}A_{yy}\right)^2 + 15\left(S_{+ss}A_{xx} - S_{-ss}A_{yy}\right)^2}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},+\frac{1}{2}} = \frac{15\left(C_{+sc}A_{xx} + C_{-sc}A_{yy}\right)^2 + 16\left(C_{+sc}A_{xx} - C_{-sc}A_{yy}\right)^2}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{1}{4}A_{zz}{}^2 \sin^2 2\theta_+}{-2\sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{15\left(S_{+ss}A_{xx} + S_{-ss}A_{yy}\right)^2 + 16\left(S_{+ss}A_{xx} - S_{-ss}A_{yy}\right)^2}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},-\frac{1}{2}} = \frac{16\left(C_{+sc}A_{xx} + C_{-sc}A_{yy}\right)^2 + 15\left(C_{+sc}A_{xx} - C_{-sc}A_{yy}\right)^2}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{1}{4}A_{zz}{}^2 \sin^2 2\theta_+}{-2\sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{16\left(S_{+ss}A_{xx} + S_{-ss}A_{yy}\right)^2 + 15\left(S_{+ss}A_{xx} - S_{-ss}A_{yy}\right)^2}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},-\frac{3}{2}} = \frac{15\left(C_{+sc}A_{xx} + C_{-sc}A_{yy}\right)^2 + 12\left(C_{+sc}A_{xx} - C_{-sc}A_{yy}\right)^2}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{9}{4}A_{zz}{}^2 \sin^2 2\theta_+}{-2\sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{15\left(S_{+ss}A_{xx} + S_{-ss}A_{yy}\right)^2 + 12\left(S_{+ss}A_{xx} - S_{-ss}A_{yy}\right)^2}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},-\frac{5}{2}} = \frac{12\left(C_{+sc}A_{xx} + C_{-sc}A_{yy}\right)^2 + 7\left(C_{+sc}A_{xx} - C_{-sc}A_{yy}\right)^2}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{25}{4}A_{zz}{}^2 \sin^2 2\theta_+}{-2\sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{12\left(S_{+ss}A_{xx} + S_{-ss}A_{yy}\right)^2 + 7\left(S_{+ss}A_{xx} - S_{-ss}A_{yy}\right)^2}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},-\frac{7}{2}} = \frac{7\left(C_{+sc}A_{xx} + C_{-sc}A_{yy}\right)^2}{g_{zz}\beta B - \sqrt{(D+g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D-g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{49}{4}{A_{zz}}^2 \sin^2 2\theta_+}{-2\sqrt{(D+g_{zz}\beta B)^2 + 3E^2}} + \frac{7\left(S_{+ss}A_{xx} + S_{-ss}A_{yy}\right)^2}{g_{zz}\beta B - \sqrt{(D+g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D-g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},+\frac{7}{2}} = \frac{7\left(C_{+sc}A_{xx} + C_{-sc}A_{yy}\right)^2}{-g_{zz}\beta B + \sqrt{(D-g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D+g_{zz}\beta B)^2 + 3E^2}} + \frac{7\left(S_{+cc}A_{xx} + S_{-cc}A_{yy}\right)^2}{-g_{zz}\beta B + \sqrt{(D-g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D+g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{49}{4}{A_{zz}}^2 \sin^2 2\theta_-}{2\sqrt{(D-g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},+\frac{5}{2}} = \frac{7\left(C_{+sc}A_{xx} - C_{-sc}A_{yy}\right)^2 + 12\left(C_{+sc}A_{xx} + C_{-sc}A_{yy}\right)^2}{-g_{zz}\beta B + \sqrt{(D-g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D+g_{zz}\beta B)^2 + 3E^2}} + \frac{7\left(S_{+cc}A_{xx} - S_{-cc}A_{yy}\right)^2 + 12\left(S_{+cc}A_{xx} + S_{-cc}A_{yy}\right)^2}{-g_{zz}\beta B + \sqrt{(D-g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D+g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{25}{4}{A_{zz}}^2 \sin^2 2\theta_-}{2\sqrt{(D-g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},+\frac{3}{2}} = \frac{12\left(C_{+sc}A_{xx} - C_{-sc}A_{yy}\right)^2 + 15\left(C_{+sc}A_{xx} + C_{-sc}A_{yy}\right)^2}{-g_{zz}\beta B + \sqrt{(D-g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D+g_{zz}\beta B)^2 + 3E^2}} + \frac{12\left(S_{+cc}A_{xx} - S_{-cc}A_{yy}\right)^2 + 15\left(S_{+cc}A_{xx} + S_{-cc}A_{yy}\right)^2}{-g_{zz}\beta B + \sqrt{(D-g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D+g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{9}{4}{A_{zz}}^2 \sin^2 2\theta_-}{2\sqrt{(D-g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},+\frac{1}{2}} = \frac{15\left(C_{+sc}A_{xx} - C_{-sc}A_{yy}\right)^2 + 16\left(C_{+sc}A_{xx} + C_{-sc}A_{yy}\right)^2}{-g_{zz}\beta B + \sqrt{(D-g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D+g_{zz}\beta B)^2 + 3E^2}} + \frac{15\left(S_{+cc}A_{xx} - S_{-cc}A_{yy}\right)^2 + 16\left(S_{+cc}A_{xx} + S_{-cc}A_{yy}\right)^2}{-g_{zz}\beta B + \sqrt{(D-g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D+g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{1}{4}{A_{zz}}^2 \sin^2 2\theta_-}{2\sqrt{(D-g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},-\frac{1}{2}} = \frac{16\left(C_{+sc}A_{xx} - C_{-sc}A_{yy}\right)^2 + 15\left(C_{+sc}A_{xx} + C_{-sc}A_{yy}\right)^2}{-g_{zz}\beta B + \sqrt{(D-g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D+g_{zz}\beta B)^2 + 3E^2}} + \frac{16\left(S_{+cc}A_{xx} - S_{-cc}A_{yy}\right)^2 + 15\left(S_{+cc}A_{xx} + S_{-cc}A_{yy}\right)^2}{-g_{zz}\beta B + \sqrt{(D-g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D+g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{1}{4}{A_{zz}}^2 \sin^2 2\theta_-}{2\sqrt{(D-g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},-\frac{3}{2}} = \frac{15\left(C_{+sc}A_{xx} - C_{-sc}A_{yy}\right)^2 + 12\left(C_{+sc}A_{xx} + C_{-sc}A_{yy}\right)^2}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{15\left(S_{+cc}A_{xx} - S_{-cc}A_{yy}\right)^2 + 12\left(S_{+cc}A_{xx} + S_{-cc}A_{yy}\right)^2}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{9}{4}A_{zz}{}^2 \sin^2 2\theta_-}{2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},-\frac{5}{2}} = \frac{12\left(C_{+sc}A_{xx} - C_{-sc}A_{yy}\right)^2 + 7\left(C_{+sc}A_{xx} + C_{-sc}A_{yy}\right)^2}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{12\left(S_{+cc}A_{xx} - S_{-cc}A_{yy}\right)^2 + 7\left(S_{+cc}A_{xx} + S_{-cc}A_{yy}\right)^2}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{25}{4}A_{zz}{}^2 \sin^2 2\theta_-}{2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},-\frac{7}{2}} = \frac{7\left(C_{+sc}A_{xx} - C_{-sc}A_{yy}\right)^2}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{7\left(S_{+cc}A_{xx} - S_{-cc}A_{yy}\right)^2}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{25}{4}A_{zz}{}^2 \sin^2 2\theta_-}{2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{1}{2},+\frac{7}{2}} = \frac{\frac{49}{4}A_{zz}{}^2 \sin^2 2\theta_+}{2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{7\left(S_{+cc}A_{xx} - S_{-cc}A_{yy}\right)^2}{g_{zz}\beta B + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{7\left(C_{+cs}A_{xx} - C_{-cs}A_{yy}\right)^2}{g_{zz}\beta B + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{1}{2},+\frac{5}{2}} = \frac{\frac{25}{4}A_{zz}{}^2 \sin^2 2\theta_+}{2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{7\left(S_{+cc}A_{xx} + S_{-cc}A_{yy}\right)^2 + 12\left(S_{+cc}A_{xx} - S_{-cc}A_{yy}\right)^2}{g_{zz}\beta B + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{7\left(C_{+cs}A_{xx} + C_{-cs}A_{yy}\right)^2 + 12\left(C_{+cs}A_{xx} - C_{-cs}A_{yy}\right)^2}{g_{zz}\beta B + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{1}{2},+\frac{3}{2}} = \frac{\frac{9}{4}A_{zz}{}^2 \sin^2 2\theta_+}{2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{12\left(S_{+cc}A_{xx} + S_{-cc}A_{yy}\right)^2 + 15\left(S_{+cc}A_{xx} - S_{-cc}A_{yy}\right)^2}{g_{zz}\beta B + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{12\left(C_{+cs}A_{xx} + C_{-cs}A_{yy}\right)^2 + 15\left(C_{+cs}A_{xx} - C_{-cs}A_{yy}\right)^2}{g_{zz}\beta B + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{1}{2},+\frac{1}{2}} = \frac{\frac{1}{4}{A_{zz}}^2 \sin^2 2\theta_+}{2\sqrt{(D-g_{zz}\beta B)^2+3E^2}} + \frac{15\left(S_{+cc}A_{xx}+S_{-cc}A_{yy}\right)^2+16\left(S_{+cc}A_{xx}-S_{-cc}A_{yy}\right)^2}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}-\sqrt{(D-g_{zz}\beta B)^2+3E^2}} + \frac{15\left(C_{+cs}A_{xx}+C_{-cs}A_{yy}\right)^2+16\left(C_{+cs}A_{xx}-C_{-cs}A_{yy}\right)^2}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}+\sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{1}{2},-\frac{1}{2}} = \frac{\frac{1}{4}{A_{zz}}^2 \sin^2 2\theta_+}{2\sqrt{(D-g_{zz}\beta B)^2+3E^2}} + \frac{16\left(S_{+cc}A_{xx}+S_{-cc}A_{yy}\right)^2+15\left(S_{+cc}A_{xx}-S_{-cc}A_{yy}\right)^2}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}-\sqrt{(D-g_{zz}\beta B)^2+3E^2}} + \frac{16\left(C_{+cs}A_{xx}+C_{-cs}A_{yy}\right)^2+15\left(C_{+cs}A_{xx}-C_{-cs}A_{yy}\right)^2}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}+\sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{1}{2},-\frac{3}{2}} = \frac{\frac{9}{4}{A_{zz}}^2 \sin^2 2\theta_+}{2\sqrt{(D-g_{zz}\beta B)^2+3E^2}} + \frac{15\left(S_{+cc}A_{xx}+S_{-cc}A_{yy}\right)^2+12\left(S_{+cc}A_{xx}-S_{-cc}A_{yy}\right)^2}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}-\sqrt{(D-g_{zz}\beta B)^2+3E^2}} + \frac{15\left(C_{+cs}A_{xx}+C_{-cs}A_{yy}\right)^2+12\left(C_{+cs}A_{xx}-C_{-cs}A_{yy}\right)^2}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}+\sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{1}{2},-\frac{5}{2}} = \frac{\frac{25}{4}{A_{zz}}^2 \sin^2 2\theta_+}{2\sqrt{(D-g_{zz}\beta B)^2+3E^2}} + \frac{12\left(S_{+cc}A_{xx}+S_{-cc}A_{yy}\right)^2+7\left(S_{+cc}A_{xx}-S_{-cc}A_{yy}\right)^2}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}-\sqrt{(D-g_{zz}\beta B)^2+3E^2}} + \frac{12\left(C_{+cs}A_{xx}+C_{-cs}A_{yy}\right)^2+7\left(C_{+cs}A_{xx}-C_{-cs}A_{yy}\right)^2}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}+\sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{1}{2},-\frac{7}{2}} = \frac{\frac{49}{4}{A_{zz}}^2 \sin^2 2\theta_+}{2\sqrt{(D-g_{zz}\beta B)^2+3E^2}} + \frac{7\left(S_{+cc}A_{xx}+S_{-cc}A_{yy}\right)^2}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}-\sqrt{(D-g_{zz}\beta B)^2+3E^2}} + \frac{7\left(C_{+cs}A_{xx}+C_{-cs}A_{yy}\right)^2}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}+\sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},+\frac{7}{2}} = \frac{7\left(S_{+ss}A_{xx}+S_{-ss}A_{yy}\right)^2}{-g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}+\sqrt{(D+g_{zz}\beta B)^2+3E^2}} + \frac{\frac{49}{4}{A_{zz}}^2 \sin^2 2\theta_-}{-2\sqrt{(D-g_{zz}\beta B)^2+3E^2}} + \frac{7\left(C_{+cs}A_{xx}+C_{-cs}A_{yy}\right)^2}{-g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}-\sqrt{(D+g_{zz}\beta B)^2+3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},+\frac{5}{2}} = \frac{7\left(S_{+ss}A_{xx}-S_{-ss}A_{yy}\right)^2+12\left(S_{+ss}A_{xx}+S_{-ss}A_{yy}\right)^2}{-g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}+\sqrt{(D+g_{zz}\beta B)^2+3E^2}} + \frac{\frac{25}{4}{A_{zz}}^2 \sin^2 2\theta_-}{-2\sqrt{(D-g_{zz}\beta B)^2+3E^2}} + \frac{7\left(C_{+cs}A_{xx}-C_{-cs}A_{yy}\right)^2+12\left(C_{+cs}A_{xx}+C_{-cs}A_{yy}\right)^2}{-g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}-\sqrt{(D+g_{zz}\beta B)^2+3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},+\frac{3}{2}} = \frac{12\left(S_{+ss}A_{xx} - S_{-ss}A_{yy}\right)^2 + 15\left(S_{+ss}A_{xx} + S_{-ss}A_{yy}\right)^2}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{9}{4}{A_{zz}}^2 \sin^2 2\theta_-}{-2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{12\left(C_{+cs}A_{xx} - C_{-cs}A_{yy}\right)^2 + 15\left(C_{+cs}A_{xx} + C_{-cs}A_{yy}\right)^2}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},+\frac{1}{2}} = \frac{15\left(S_{+ss}A_{xx} - S_{-ss}A_{yy}\right)^2 + 16\left(S_{+ss}A_{xx} + S_{-ss}A_{yy}\right)^2}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{1}{4}{A_{zz}}^2 \sin^2 2\theta_-}{-2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{15\left(C_{+cs}A_{xx} - C_{-cs}A_{yy}\right)^2 + 16\left(C_{+cs}A_{xx} + C_{-cs}A_{yy}\right)^2}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},-\frac{1}{2}} = \frac{16\left(S_{+ss}A_{xx} - S_{-ss}A_{yy}\right)^2 + 15\left(S_{+ss}A_{xx} + S_{-ss}A_{yy}\right)^2}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{1}{4}{A_{zz}}^2 \sin^2 2\theta_-}{-2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{16\left(C_{+cs}A_{xx} - C_{-cs}A_{yy}\right)^2 + 15\left(C_{+cs}A_{xx} + C_{-cs}A_{yy}\right)^2}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},-\frac{3}{2}} = \frac{15\left(S_{+ss}A_{xx} - S_{-ss}A_{yy}\right)^2 + 12\left(S_{+ss}A_{xx} + S_{-ss}A_{yy}\right)^2}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{9}{4}{A_{zz}}^2 \sin^2 2\theta_-}{-2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{15\left(C_{+cs}A_{xx} - C_{-cs}A_{yy}\right)^2 + 12\left(C_{+cs}A_{xx} + C_{-cs}A_{yy}\right)^2}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},-\frac{5}{2}} = \frac{12\left(S_{+ss}A_{xx} - S_{-ss}A_{yy}\right)^2 + 7\left(S_{+ss}A_{xx} + S_{-ss}A_{yy}\right)^2}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{25}{4}{A_{zz}}^2 \sin^2 2\theta_-}{-2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{12\left(C_{+cs}A_{xx} - C_{-cs}A_{yy}\right)^2 + 7\left(C_{+cs}A_{xx} + C_{-cs}A_{yy}\right)^2}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},-\frac{7}{2}} = \frac{7\left(S_{+ss}A_{xx} - S_{-ss}A_{yy}\right)^2}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{49}{4}{A_{zz}}^2 \sin^2 2\theta_-}{-2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{7\left(C_{+cs}A_{xx} - C_{-cs}A_{yy}\right)^2}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$

In summary, when the ZFS, **g-** and **A**-tensors are collinear and the magnetic field is aligned to the *z*-axis of the principal axis coordination system, the zeroth-order and perturbed energies in the second order are represented as follows:

$$E_{+\frac{3}{2},+\frac{7}{2}} = \frac{1}{2}g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \frac{7}{4}A_{zz}(2\cos 2\theta_+ + 1) + \varepsilon^{(2)}_{+\frac{3}{2},+\frac{7}{2}}$$

$$E_{+\frac{3}{2},+\frac{5}{2}} = \frac{1}{2}g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \frac{5}{4}A_{zz}(2\cos 2\theta_+ + 1) + \varepsilon^{(2)}_{+\frac{3}{2},+\frac{5}{2}}$$

$$E_{+\frac{3}{2},+\frac{3}{2}} = \frac{1}{2}g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \frac{3}{4}A_{zz}(2\cos 2\theta_+ + 1) + \varepsilon^{(2)}_{+\frac{3}{2},+\frac{3}{2}}$$

$$E_{+\frac{3}{2},+\frac{1}{2}} = \frac{1}{2}g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \frac{1}{4}A_{zz}(2\cos 2\theta_+ + 1) + \varepsilon^{(2)}_{+\frac{3}{2},+\frac{1}{2}}$$

$$E_{+\frac{3}{2},-\frac{1}{2}} = \frac{1}{2}g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \frac{1}{4}A_{zz}(2\cos 2\theta_+ + 1) + \varepsilon^{(2)}_{+\frac{3}{2},-\frac{1}{2}}$$

$$E_{+\frac{3}{2},-\frac{3}{2}} = \frac{1}{2}g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \frac{3}{4}A_{zz}(2\cos 2\theta_+ + 1) + \varepsilon^{(2)}_{+\frac{3}{2},-\frac{3}{2}}$$

$$E_{+\frac{3}{2},-\frac{5}{2}} = \frac{1}{2}g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \frac{5}{4}A_{zz}(2\cos 2\theta_+ + 1) + \varepsilon^{(2)}_{+\frac{3}{2},-\frac{5}{2}}$$

$$E_{+\frac{3}{2},-\frac{7}{2}} = \frac{1}{2}g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \frac{7}{4}A_{zz}(2\cos 2\theta_+ + 1) + \varepsilon^{(2)}_{+\frac{3}{2},-\frac{7}{2}}$$

$$E_{+\frac{1}{2},+\frac{7}{2}} = -\frac{1}{2}g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \frac{7}{4}A_{zz}(2\cos 2\theta_- - 1) + \varepsilon^{(2)}_{+\frac{1}{2},+\frac{7}{2}}$$

$$E_{+\frac{1}{2},+\frac{5}{2}} = -\frac{1}{2}g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \frac{5}{4}A_{zz}(2\cos 2\theta_- - 1) + \varepsilon^{(2)}_{+\frac{1}{2},+\frac{5}{2}}$$

$$E_{+\frac{1}{2},+\frac{3}{2}} = -\frac{1}{2}g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \frac{3}{4}A_{zz}(2\cos 2\theta_- - 1) + \varepsilon^{(2)}_{+\frac{1}{2},+\frac{3}{2}}$$

$$E_{+\frac{1}{2},+\frac{1}{2}} = -\frac{1}{2}g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \frac{1}{4}A_{zz}(2\cos 2\theta_- - 1) + \varepsilon^{(2)}_{+\frac{1}{2},+\frac{1}{2}}$$

$$E_{+\frac{1}{2},-\frac{1}{2}} = -\frac{1}{2}g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \frac{1}{4}A_{zz}(2\cos 2\theta_- - 1) + \varepsilon^{(2)}_{+\frac{1}{2},-\frac{1}{2}}$$

$$E_{+\frac{1}{2},-\frac{3}{2}} = -\frac{1}{2}g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \frac{3}{4}A_{zz}(2\cos 2\theta_- - 1) + \varepsilon^{(2)}_{+\frac{1}{2},-\frac{3}{2}}$$

$$E_{+\frac{1}{2},-\frac{5}{2}} = -\frac{1}{2}g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \frac{5}{4}A_{zz}(2\cos 2\theta_- - 1) + \varepsilon^{(2)}_{+\frac{1}{2},-\frac{5}{2}}$$

$$E_{+\frac{1}{2},-\frac{7}{2}} = -\frac{1}{2}g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \frac{7}{4}A_{zz}(2\cos 2\theta_- - 1) + \varepsilon^{(2)}_{+\frac{1}{2},-\frac{7}{2}}$$

$$E_{-\frac{1}{2},+\frac{7}{2}} = \frac{1}{2}g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \frac{7}{4}A_{zz}(2\cos 2\theta_+ - 1) + \varepsilon^{(2)}_{-\frac{1}{2},+\frac{7}{2}}$$

$$E_{-\frac{1}{2},+\frac{5}{2}} = \frac{1}{2}g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \frac{5}{4}A_{zz}(2\cos 2\theta_+ - 1) + \varepsilon^{(2)}_{-\frac{1}{2},+\frac{5}{2}}$$

$$E_{-\frac{1}{2},+\frac{3}{2}} = \frac{1}{2}g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \frac{3}{4}A_{zz}(2\cos 2\theta_+ - 1) + \varepsilon^{(2)}_{-\frac{1}{2},+\frac{3}{2}}$$

$$E_{-\frac{1}{2},+\frac{1}{2}} = \frac{1}{2}g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \frac{1}{4}A_{zz}(2\cos 2\theta_+ - 1) + \varepsilon^{(2)}_{-\frac{1}{2},+\frac{1}{2}}$$

$$E_{-\frac{1}{2},-\frac{1}{2}} = \frac{1}{2}g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \frac{1}{4}A_{zz}(2\cos 2\theta_+ - 1) + \varepsilon^{(2)}_{-\frac{1}{2},-\frac{1}{2}}$$

$$E_{-\frac{1}{2},-\frac{3}{2}} = \frac{1}{2}g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \frac{3}{4}A_{zz}(2\cos 2\theta_+ - 1) + \varepsilon^{(2)}_{-\frac{1}{2},-\frac{3}{2}}$$

$$E_{-\frac{1}{2},-\frac{5}{2}}=\frac{1}{2}g_{zz}\beta B-\sqrt{(D+g_{zz}\beta B)^2+3E^2}+\frac{5}{4}A_{zz}(2\cos 2\theta_+-1)+\varepsilon^{(2)}_{-\frac{1}{2},-\frac{5}{2}}$$

$$E_{-\frac{1}{2},-\frac{7}{2}}=\frac{1}{2}g_{zz}\beta B-\sqrt{(D+g_{zz}\beta B)^2+3E^2}+\frac{7}{4}A_{zz}(2\cos 2\theta_+-1)+\varepsilon^{(2)}_{-\frac{1}{2},-\frac{7}{2}}$$

$$E_{-\frac{3}{2},+\frac{7}{2}}=-\frac{1}{2}g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}-\frac{7}{4}A_{zz}(2\cos 2\theta_-+1)+\varepsilon^{(2)}_{-\frac{3}{2},+\frac{7}{2}}$$

$$E_{-\frac{3}{2},+\frac{5}{2}}=-\frac{1}{2}g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}-\frac{5}{4}A_{zz}(2\cos 2\theta_-+1)+\varepsilon^{(2)}_{-\frac{3}{2},+\frac{5}{2}}$$

$$E_{-\frac{3}{2},+\frac{3}{2}}=-\frac{1}{2}g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}-\frac{3}{4}A_{zz}(2\cos 2\theta_-+1)+\varepsilon^{(2)}_{-\frac{3}{2},+\frac{3}{2}}$$

$$E_{-\frac{3}{2},+\frac{1}{2}}=-\frac{1}{2}g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}-\frac{1}{4}A_{zz}(2\cos 2\theta_-+1)+\varepsilon^{(2)}_{-\frac{3}{2},+\frac{1}{2}}$$

$$E_{-\frac{3}{2},-\frac{1}{2}}=-\frac{1}{2}g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}+\frac{1}{4}A_{zz}(2\cos 2\theta_-+1)+\varepsilon^{(2)}_{-\frac{3}{2},-\frac{1}{2}}$$

$$E_{-\frac{3}{2},-\frac{3}{2}}=-\frac{1}{2}g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}+\frac{3}{4}A_{zz}(2\cos 2\theta_-+1)+\varepsilon^{(2)}_{-\frac{3}{2},-\frac{3}{2}}$$

$$E_{-\frac{3}{2},-\frac{5}{2}}=-\frac{1}{2}g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}+\frac{5}{4}A_{zz}(2\cos 2\theta_-+1)+\varepsilon^{(2)}_{-\frac{3}{2},-\frac{5}{2}}$$

$$E_{-\frac{3}{2},-\frac{7}{2}}=-\frac{1}{2}g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}+\frac{7}{4}A_{zz}(2\cos 2\theta_-+1)+\varepsilon^{(2)}_{-\frac{3}{2},-\frac{7}{2}}$$

The analytical expressions derived above with the static magnetic field **B** along the principal z-axis for the case that all the three magnetic tensors are collinear for the spin quartet state with a nuclear spin $I = 7/2$ are extremely accurate. Note that the other analytical expressions with **B** applied parallel to the other principal axes can easily be derived by invoking the cyclic transformation of the principal axis coordinates, which includes giving D replaced with $(3E - D)/2$ and E replaced with $-(E + D)/2$ for **B**//x, and similarly D with $-(3E + D)/2$ and E with $(D - E)/2$ for **B**//y.

b) Evaluation of the accuracy of the $A^{\mathrm{eff}}/A^{\mathrm{true}}$ relationships

In this section, we show the accuracy of the A_{ZZ}-value estimated for the spin quartet state with sizable ZFS tensors assuming the $g^{\mathrm{eff}}/g^{\mathrm{true}}$ relationships, noting that the $A^{\mathrm{eff}}/A^{\mathrm{true}}$ relationship is valid. The energies derived in the previous section are fully dependent on g_{ZZ}, D, E, A_{ZZ} and B. If the energies and all the parameters are accurate, the following equation holds, affording the relevant expressions. .

$$E_{+\frac{3}{2},M_I}(g_{zz},D,E,A_{zz},B)-E_{-\frac{3}{2},M_I}(g_{zz},D,E,A_{zz},B)=E^{\mathrm{eff}}_{+\frac{1}{2},M_I}\left(g^{\mathrm{eff}}_{zz},A^{\mathrm{eff}}_{zz},B\right)-E^{\mathrm{eff}}_{-\frac{1}{2},M_I}\left(g^{\mathrm{eff}}_{zz},A^{\mathrm{eff}}_{zz},B\right)\qquad(\dagger)$$

where the difference between the energies from the true spin-Hamiltonian in the left-hand side equates to that between the energies from the fictitious spin-1/2 Hamiltonian in the right-hand side. The values for g_{zz}^{eff}, A_{zz}^{eff} and B can be easily obtained from the experiment and its associated spectral analyses, and the value for g_{zz} and the ZFS parameters are obtained by using the $g^{\mathrm{eff}}/g^{\mathrm{true}}$ relationships as a function of E/D. Then, we solve the equation for A_{zz} and compare the solution with the experimental value obtained from the $A^{\mathrm{eff}}/A^{\mathrm{true}}$

relationship. As shown below, the A^{eff}/A^{true} relationship is valid since the analytical expressions for the energies underlying the relationship and all the necessary parameters are highly accurate. The accuracy of the particular relationship eq (‡), as given below, will be shown in the following sections,

$$\frac{A_{zz}^{eff}}{A_{zz}} = 1 + \frac{2}{\sqrt{1+3\lambda^2}} \quad (‡)$$

where $\lambda = E/D$.

c) ESR Analysis of complex **1**

Table S1. The possible combinations of the g^{eff}/g^{true} relationships.

Case	g^{true}	g^{eff}	g^{eff}/g^{true}	Figure No.
	x	0.82	$1-\frac{1-3\lambda}{\sqrt{1+3\lambda^2}}$	
1	y	1.3	$1-\frac{1+3\lambda}{\sqrt{1+3\lambda^2}}$	Figs. S1, S2, S3, S4 and S5
	z	7.72	$1-\frac{2}{\sqrt{1+3\lambda^2}}$	
	x	1.3	$1-\frac{1-3\lambda}{\sqrt{1+3\lambda^2}}$	
2	y	0.82	$1-\frac{1+3\lambda}{\sqrt{1+3\lambda^2}}$	Figs. 6 and 7 in the main text and Figs. S6, S7, S8, S9 and S10
	z	7.72	$1-\frac{2}{\sqrt{1+3\lambda^2}}$	

* *Case 2*, which is highlighted in yellow, provides the most reasonably simulated spectrum.

Case 1

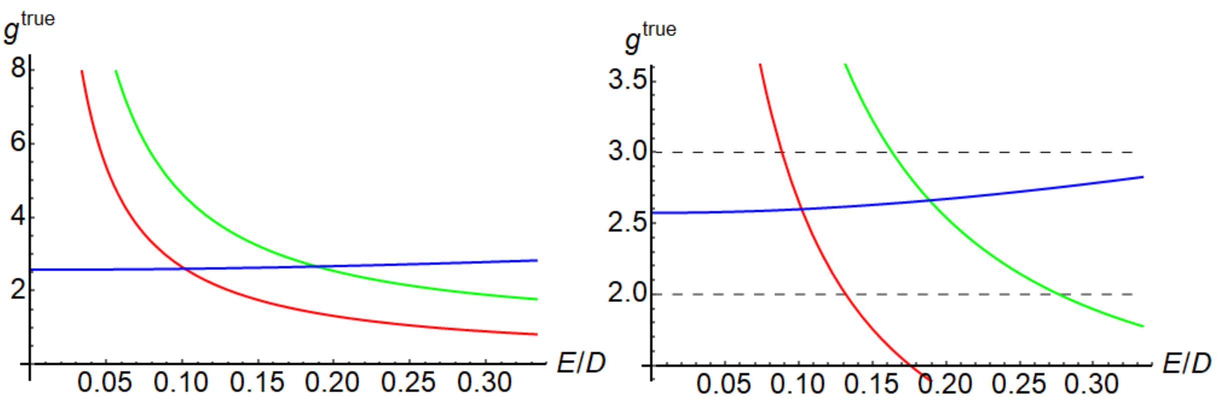

Figure S1. The calculated g^{true} values for complex **1** by use of the g^{eff}-value and g^{eff}/g^{true} relationships. $\frac{0.82}{g_{XX}^{true}} = 1-\frac{1-3\lambda}{\sqrt{1+3\lambda^2}}$, $\frac{1.3}{g_{YY}^{true}} = 1-\frac{1+3\lambda}{\sqrt{1+3\lambda^2}}$, $\frac{7.72}{g_{ZZ}^{true}} = 1+\frac{2}{\sqrt{1+3\lambda^2}}$ with $\lambda = E/D$. The g^{eff}/g^{true} relationships adopted here are for the $|M_S = \pm 3/2\rangle$-dominant transitions.

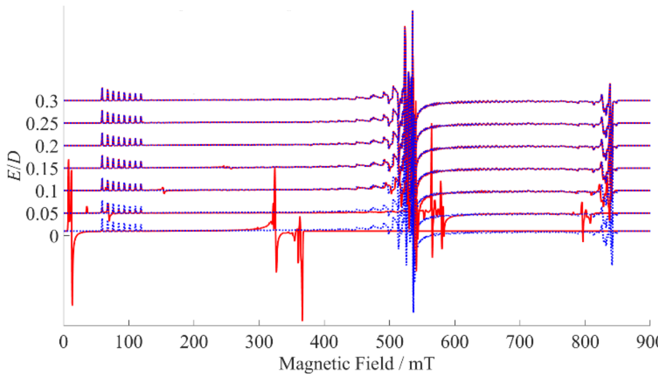

Figure S2. The simulated randomly-oriented X-band ESR spectra of complex **1** ($S = 3/2$) as a function of E/D. The values for E/D used were 0.01, 0.05, 0.1, 0.15, 0.2, 0.25 and 0.3. The spectra in blue and red are based on the fictitious spin-1/2 and true spin Hamiltonian approaches, respectively. Microwave frequency used: 9.625 GHz, the peak-to-peak linewidth: 1.0 mT, the magnetic tensors: $\mathbf{g}^{\mathrm{eff}} = [0.82, 1.3, 7.72]$, $A_x^{\mathrm{eff}}(^{59}\mathrm{Co}) = 919.2$ MHz, $D = -14$ cm^{-1} and $\mathbf{g}^{\mathrm{true}}$ and $A_z^{\mathrm{true}}(^{59}\mathrm{Co})$ for the variable E/D's are calculated by using the value of g^{eff} and the $g^{\mathrm{eff}}/g^{\mathrm{true}}$ relationships shown in Table S2. The **g**-, **A**- and **D**-tensors were assumed to be collinear. Any strain effect of the tensor to the linewidth was not included. The simulated spectra were obtained by using EasySpin (ver. 6.0.6) [8].

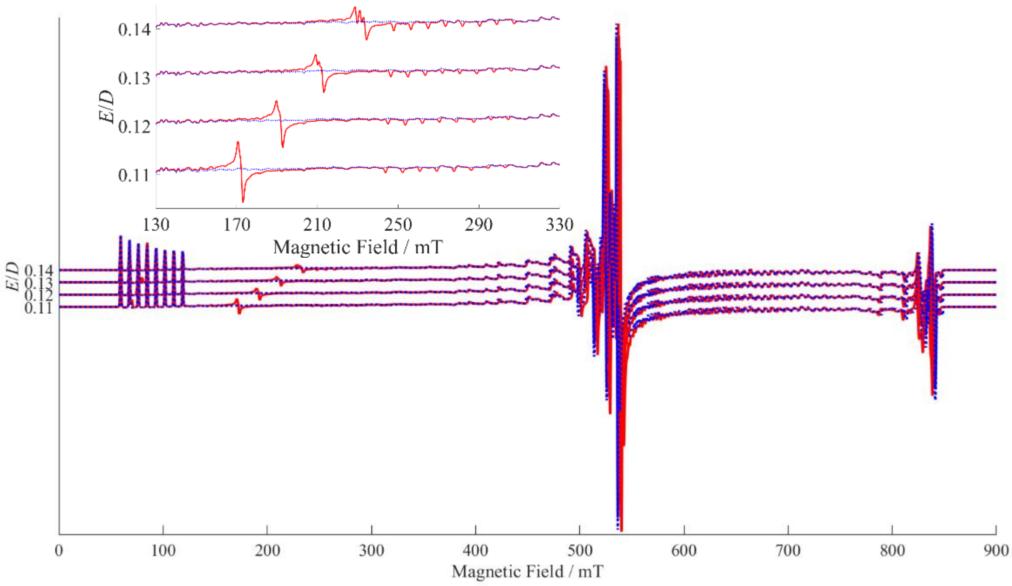

Figure S3. The simulated randomly-oriented X-band ESR spectra of complex **1** ($S = 3/2$) as a function of E/D. The values for E/D used were 0.11, 0.12, 0.13 and 0.14. The spectra in blue and red are based on the fictitious spin-1/2 and true spin Hamiltonian approaches, respectively. Microwave frequency used: 9.625 GHz, the peak-to-peak linewidth: 1.0 mT, the magnetic tensors: $\mathbf{g}^{\mathrm{eff}} = [0.82, 1.3, 7.72]$, $A_x^{\mathrm{eff}}(^{59}\mathrm{Co}) = 919.2$ MHz, $D = -14$ cm^{-1} and $\mathbf{g}^{\mathrm{true}}$ and $A_z^{\mathrm{true}}(^{59}\mathrm{Co})$ for the variable E/D's are calculated by using the value of g^{eff} and the $g^{\mathrm{eff}}/g^{\mathrm{true}}$ relationships shown in Table S2. The **g**-, **A**- and **D**-tensors were assumed to be collinear. Any strain effect of the tensor to the linewidth was not included. The simulated spectra were obtained by using EasySpin (ver. 6.0.6) [8].

Table S2. The principal values of the $\mathbf{g}^{true}$-tensor and $\mathbf{A}_{zz}$-values used in the simulated spectra of complex **1** (Figures S20 and S21). $\frac{0.82}{g_{XX}^{true}} = 1 - \frac{1-3\lambda}{\sqrt{1+3\lambda^2}}$, $\frac{1.3}{g_{YY}^{true}} = 1 - \frac{1+3\lambda}{\sqrt{1+3\lambda^2}}$, $\frac{7.72}{g_{ZZ}^{true}} = 1 + \frac{2}{\sqrt{1+3\lambda^2}}$ with $\lambda = E/D$. The g^{eff}/g^{true} relationships adopted here are for the $|M_S = \pm 3/2\rangle$-dominant transitions.

E/D	g_{XX}	g_{YY}	g_{ZZ}	E/D	g_{XX}	g_{YY}	g_{ZZ}
0.01	27.2	43.6	2.574	0.14	1.879	3.421	2.623
0.10	2.643	4.628	2.599	0.15	1.753	3.223	2.630
0.11	2.399	4.241	2.604	0.20	1.318	2.540	2.671
0.12	2.196	3.921	2.610	0.25	1.064	2.146	2.723
0.13	2.025	3.651	2.616	0.30	0.900	1.895	2.782

One of the possible sets of the magnetic parameters: $g_{XX} = 2.025$, $g_{YY} = 3.651$, $g_{ZZ} = 2.616$, $A_{ZZ}(^{59}Co) = 311.5$ MHz, $D = -14$ cm^{-1} and $E/D = +0.13$. The extra line observed at about 200 mT was simulated when $E/D = +0.18$.

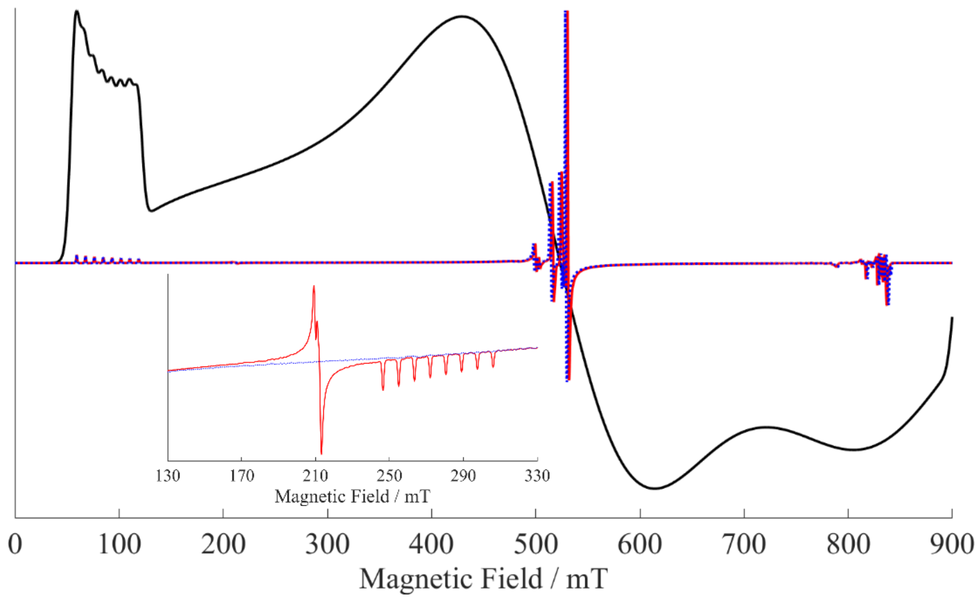

Figure S4. The simulated randomly-oriented X-band ESR spectra of complex **1** ($S = 3/2$). The spectra in blue and red are based on the fictitious spin-1/2 and true spin Hamiltonian approaches, respectively. Microwave frequency used: 9.625 GHz, the peak-to-peak linewidth: 1.0 mT, the magnetic tensors: $\mathbf{g}^{eff} = [0.82, 1.30, 7.72]$, $A_z^{eff}(^{59}Co) = 919.2$ MHz, $\mathbf{g}^{true} = [2.025, 3.651, 2.616]$, $A_z^{true}(^{59}Co) = 311.5$ MHz, $D = -14$ cm^{-1} and $E/D = +0.13$. The **g**-, **A**- and **D**-tensors were assumed to be collinear. Any strain effect of the tensor to the linewidth was not included. The simulated spectra were obtained by using EasySpin (ver. 6.0.6) [8]. Inset: the expanded spectrum from 130 mT to 330 mT. The extralines attributed from the $|M_S = \pm 1/2\rangle$-dominant transitions can be reproduced by using the true spin Hamiltonian (red line) but not by the fictitious spin-1/2 Hamiltonian (blue line).

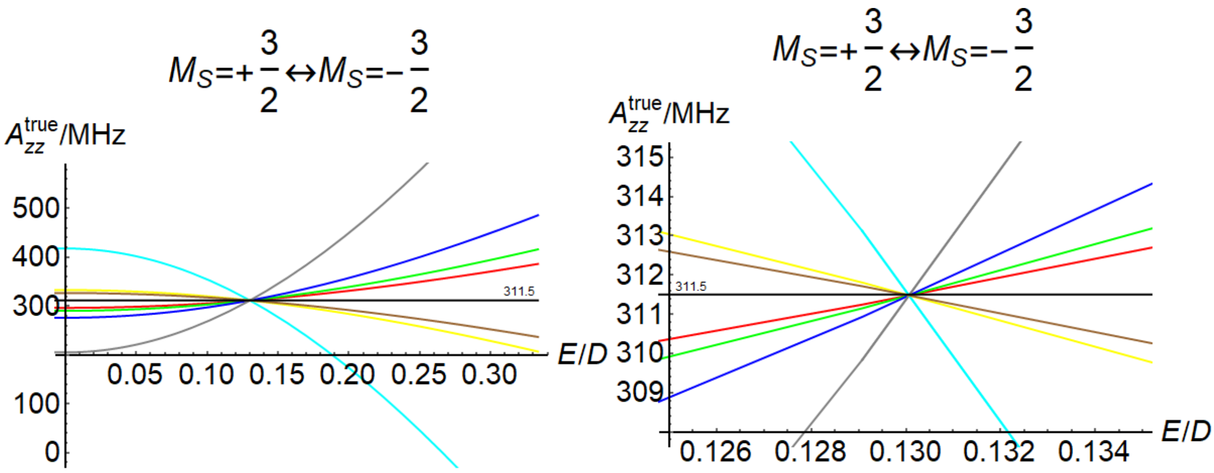

Figure S5. The E/D dependence of the A_Z^{true}-value which is the solution of eq (†) with the parameters used for the simulated spectrum in Figure S22. Red, green, blue, gray, cyan, magenta, yellow and brown curves correspond to the assignments to M_I = +7/2, +5/2, +3/2, +1/2, –1/2, –3/2, –5/2 and –7/2, respectively. Because all the energies and the parameters were accurate, all the curves cross at one point. The black line indicates the A_Z-value (= 311.5 MHz) calculated by using the relationship (eq (‡)) in the case of E/D = 0.13. The figure on the right shows the expanded one in the range of 0.125 ≤ E/D ≤ 0.135.

Case 2

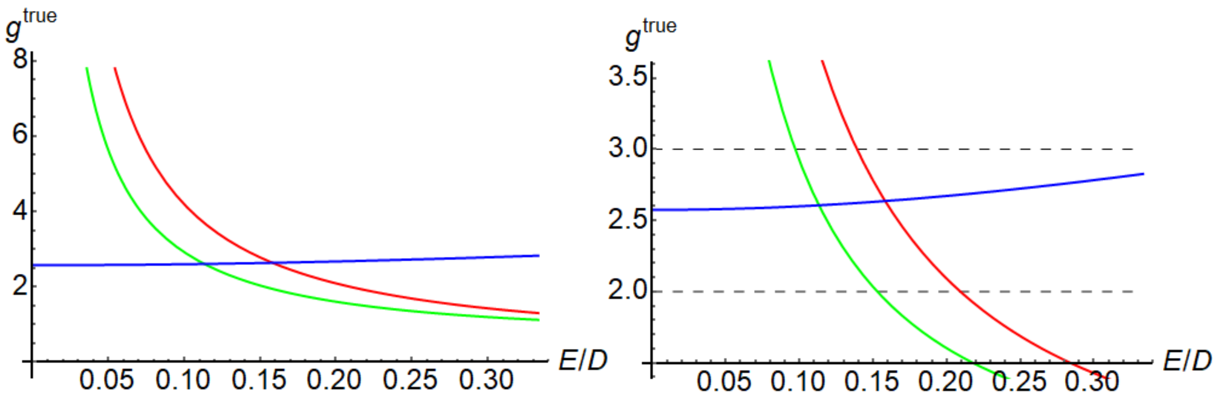

Figure S6. The calculated g^{true}-values by use of the g^{eff}-value and g^{eff}/g^{true} relationships. $\frac{1.3}{g_{XX}^{true}} = 1 - \frac{1-3\lambda}{\sqrt{1+3\lambda^2}}$, $\frac{0.82}{g_{YY}^{true}} = 1 - \frac{1+3\lambda}{\sqrt{1+3\lambda^2}}$, $\frac{7.72}{g_{ZZ}^{true}} = 1 + \frac{2}{\sqrt{1+3\lambda^2}}$ with $\lambda = E/D$. The g^{eff}/g^{true} relationships adopted here are for the $|M_S$ = ±3/2>-dominant transitions.

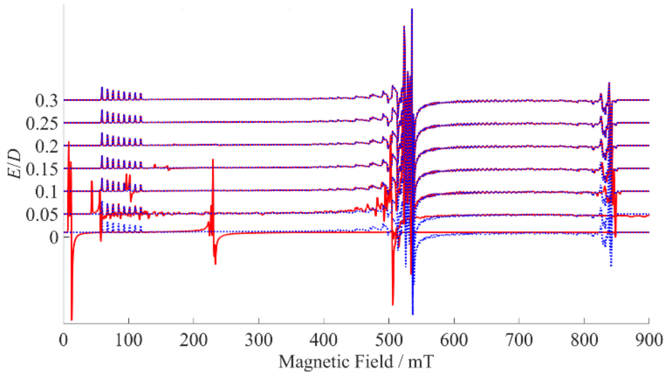

Figure S7. The simulated randomly-oriented X-band ESR spectra of complex **1** ($S = 3/2$). The spectra in blue and red are based on the fictitious spin-1/2 and true spin Hamiltonian approach, respectively. Microwave frequency used: 9.625 GHz, the peak-to-peak linewidth: 1.0 mT, the magnetic tensors: $\mathbf{g}^{\text{eff}} = [0.82, 1.3, 7.72]$, $A_x^{\text{eff}}(^{59}\text{Co}) = 919.2$ MHz, $D = -14\ \text{cm}^{-1}$ and $\mathbf{g}^{\text{true}}$ and $A_z^{\text{true}}(^{59}\text{Co})$ for the variable E/D's are calculated by using the value of g^{eff} and the $g^{\text{eff}}/g^{\text{true}}$ relationships. The **g**-, **A**- and **D**-tensors were assumed to be collinear. Any strain effect of the tensor to the linewidth was not included. The simulated spectra were obtained by using EasySpin (ver. 6.0.6) [8].

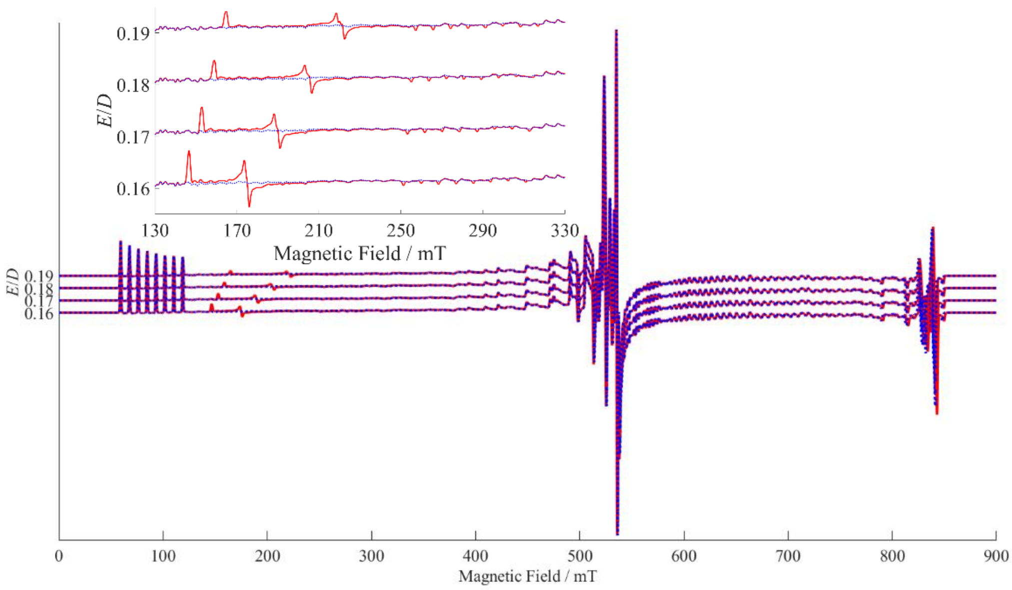

Figure S8. The simulated randomly-oriented ESR spectra of complex **1** ($S = 3/2$) as a function of E/D. The spectra in blue and red are based on the fictitious spin-1/2 and true spin Hamiltonian approaches, respectively. Microwave frequency used: 9.625 GHz, the peak-to-peak linewidth: 1.0 mT; the magnetic tensors: $\mathbf{g}^{\text{eff}} = [0.82, 1.30, 7.72]$, $A_z^{\text{eff}}(^{59}\text{Co}) = 919.2$ MHz, $D = -14\ \text{cm}^{-1}$ and $\mathbf{g}^{\text{true}}$ and $A_Z^{\text{true}}(^{59}\text{Co})$ for the variable E/D's are shown in Table S3. The **g**-, **A**- and **D**-tensors were assumed to be collinear. Any strain effect of the tensor to the linewidth was not included. The simulated spectra were obtained by using EasySpin (ver. 6.0.6) [8]. Note that the resonance peaks marked by a single asterisk *, double asterisk ** and triple asterisk *** appear only for the true spin Hamiltonian approach. Inset: the expanded spectra in the range of 130 mT to 330 mT.

Table S3. The principal values of the $\mathbf{g}^{\text{true}}$-tensor and $\mathbf{A}_{zz}$-values used in the simulated spectra of complex **1** (Figures S25 and S26). $\frac{1.3}{g_{XX}^{\text{true}}} = 1 - \frac{1-3\lambda}{\sqrt{1+3\lambda^2}}$, $\frac{0.82}{g_{YY}^{\text{true}}} = 1 - \frac{1+3\lambda}{\sqrt{1+3\lambda^2}}$, $\frac{7.72}{g_{ZZ}^{\text{true}}} = 1 + \frac{2}{\sqrt{1+3\lambda^2}}$ with $\lambda = E/D$. The $g^{\text{eff}}/g^{\text{true}}$ relationships adopted here are for the $|M_S = \pm 3/2\rangle$-dominant transitions.

E/D	g_{XX}	g_{YY}	g_{ZZ}	E/D	g_{XX}	g_{YY}	g_{ZZ}
0.01	43.1	27.5	2.574	0.18	2.318	1.744	2.654
0.10	4.190	2.919	2.599	0.19	2.198	1.669	2.662
0.15	2.780	2.033	2.630	0.20	2.090	1.602	2.671
0.16	2.606	1.924	2.637	0.25	1.687	1.353	2.723
0.17	2.453	1.828	2.645	0.30	1.427	1.195	2.782

One of the possible sets of the magnetic parameters: $g_{XX} = 2.318$, $g_{YY} = 1.744$, $g_{ZZ} = 2.654$, $A_{ZZ}(^{59}\text{Co}) = 316.0$ MHz, $D = -14$ cm^{-1} and $E/D = +0.18$. The extra line observed at about 200 mT was simulated when $E/D = +0.18$.

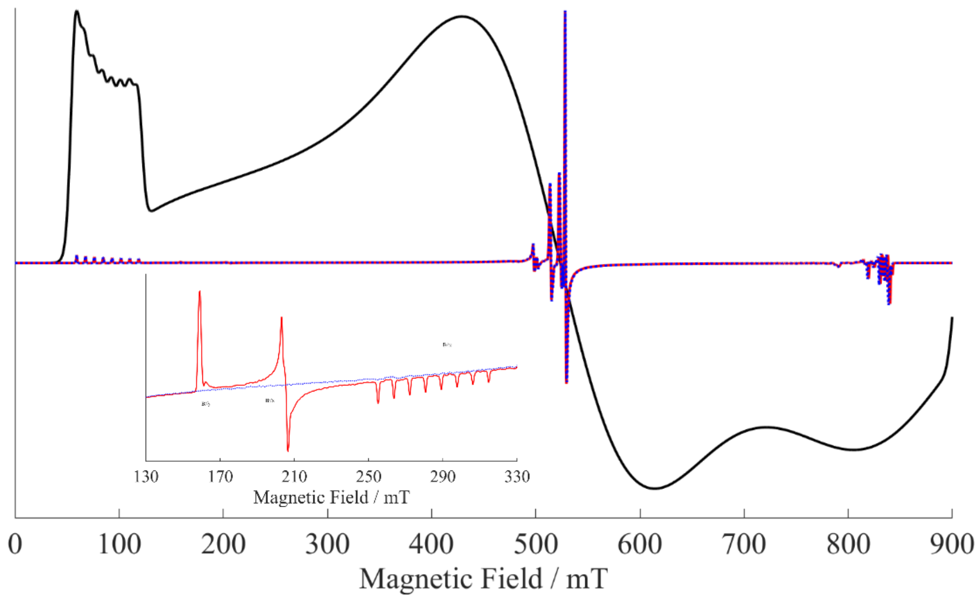

Figure S9. The simulated randomly-oriented X-band ESR spectra of complex **1** ($S = 3/2$). The spectra in blue and red are based on the fictitious spin-1/2 and true spin Hamiltonian approaches, respectively. Microwave frequency used: 9.625 GHz, the peak-to-peak linewidth: 1.0 mT, the magnetic tensors: $\mathbf{g}^{\text{eff}} = [1.30, 0.82, 7.72]$, $A_z^{\text{eff}}(^{59}\text{Co}) = 919.2$ MHz, $\mathbf{g}^{\text{true}} = [2.318, 1.744, 2.654]$, $A_z^{\text{true}}(^{59}\text{Co}) = 316.0$ MHz, $D = -14$ cm^{-1} and $E/D = +0.18$. The **g**-, **A**- and **D**-tensors were assumed to be collinear. Any strain effect of the tensor to the linewidth was not included. The simulated spectra were obtained by using EasySpin (ver. 6.0.6) [8].

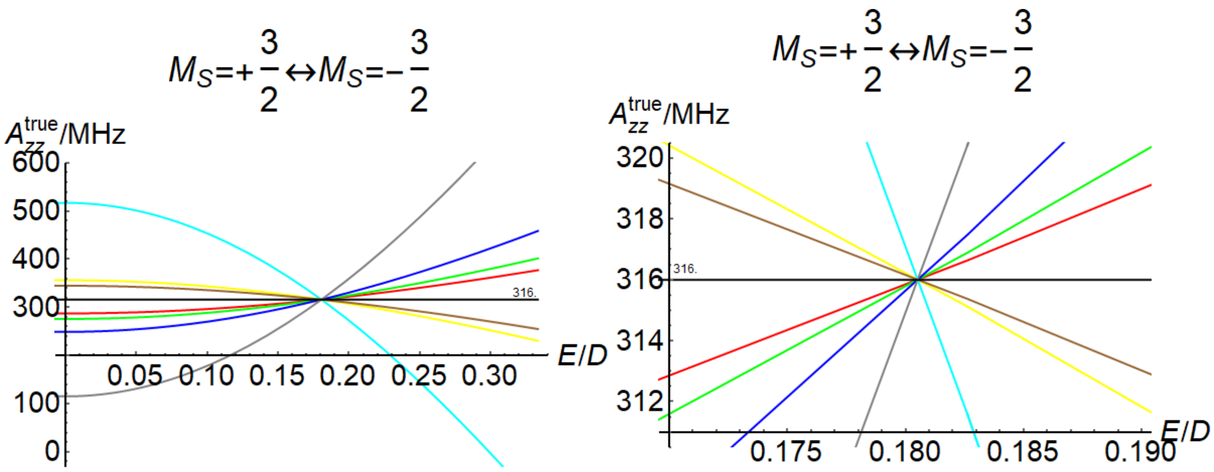

Figure S10. The E/D dependence of the A_Z^{true}-value which is the solution of eq (†) with the parameters used for the simulated spectrum in Figure S1-5. Red, green, blue, gray, cyan, magenta, yellow and brown curves correspond to the assignments to M_I = +7/2, +5/2, +3/2, +1/2, –1/2, –3/2, –5/2 and –7/2, respectively. Because all the energies and the parameters were accurate, all the curves cross at one point. The black line indicates the A_Z-value (= 316.0 MHz) calculated by using the relationship (eq (‡)) in the case of E/D = 0.18. The right figure shows the expanded one in the range of $0.17 \le E/D \le 0.19$.

Table S4. Comparison of the theoretical and the experimental magnetic parameters. Theoretical values were taken from the result of DFT calculations.

	Theor.	Expl. (*Case 1*)	Expl. (*Case 2*)
g_x	2.0671	2.025	2.318
g_y	2.0550	2.616	2.654
g_z	2.1119	3.651	1.744
A_z/MHz	84.49	311.5	316.0
D/cm^{-1}	+9.9705	–14	–14
E/D	+0.1666	+0.13	+0.18

It is worth noting that the principal values of the $\mathbf{g}^{true}$-tensor are not permutable.

Table S5. Permutation of the obtained principal values of the $\mathbf{g}^{true}$-tensor.

Case	g_{XX}	g_{YY}	g_{ZZ}
	2.025	3.65	2.616
(a)	2.025	2.616	3.65
(b)	3.65	2.616	2.025
(c)	3.65	2.025	2.616
(d)	2.616	2.025	3.65
(e)	2.616	3.65	2.025

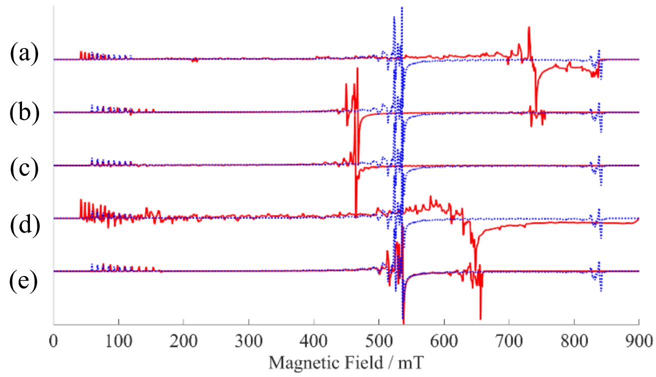

Figure S11. The randomly-oriented X-band ESR spectra of complex **1** ($S = 3/2$), as simulated based on the magnetic parameters in the literature [6] The spectra in blue and red are based on the fictitious spin-1/2 and true spin Hamiltonian approaches, respectively. Microwave frequency used: 9.625 GHz, the peak-to-peak linewidth: 1.0 mT, the magnetic parameters: $g_1^{eff} = 7.72$, $g_2^{eff} = 1.30$, $g_3^{eff} = 0.82$, $A_1^{eff}(^{59}Co) = 919.2$ MHz, $\mathbf{g}^{true}$ are shown in Table S5, $A_z^{true}(^{59}Co) = 459.6$ MHz, $D = –14$ cm^{-1} and $E/D = +0.13$. The **g**-, **A**- and **D**-tensors were assumed to be collinear. Any strain effect of the tensor to the linewidth was not included. The simulated spectra were obtained by using EasySpin (ver. 6.0.6) [8].

Expected ZF (Zero-Field)-Frequency domain spectra for complex **1**:

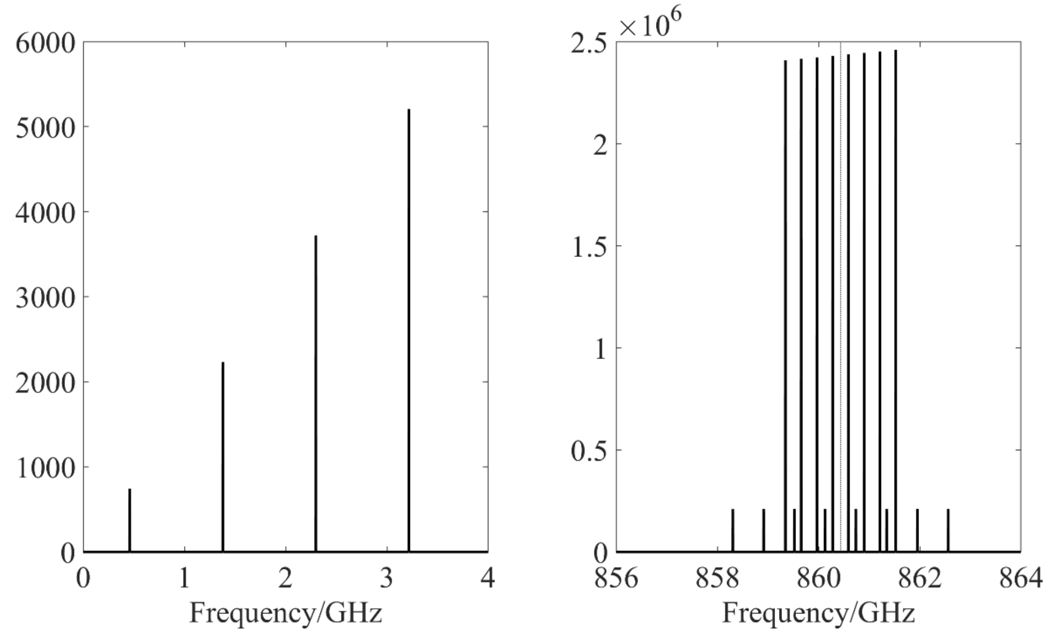

Figure S12. The simulated zero-field frequency-domain spectra of complex **1**. The magnetic tensors: $\mathbf{g}^{\mathrm{true}}$ = [2.025, 2.616, 3.651], $A_z^{\mathrm{true}}(^{59}\mathrm{Co})$ = 311.5 MHz, D = –14 cm^{-1} and E/D = +0.13 and the linewidth was set to 1 MHz. The **g**-, **A**- and **D**-tensors were assumed to be collinear. Any strain effect of the tensor to the linewidth was not included. The simulated spectra (stick) were obtained by using EasySpin (ver. 6.0.0-dev.29) [8]. The low-frequency region was attributed to hyperfine transitions. The broken vertical line on the right indicates the value of $\sqrt{D^2 + 3E^2} = 860.44\ \mathrm{GHz}$ in units of GHz.

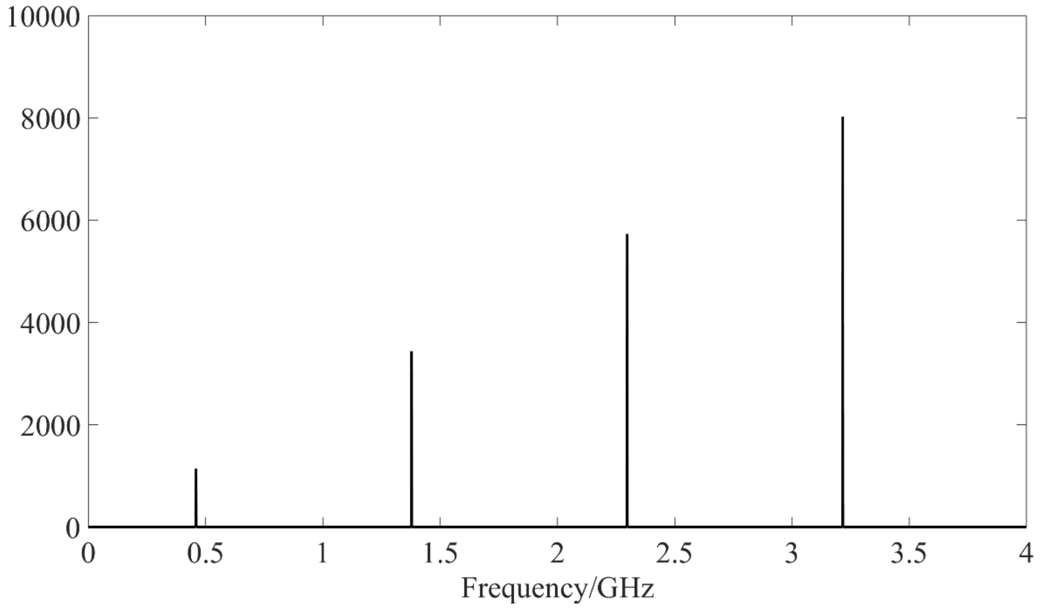

Figure S13. The simulated frequency-domain zero-field Fourier-transform spectrum of complex **1** (S = 3/2). The magnetic tensors: $\mathbf{g}^{\mathrm{eff}}$ = [1.30, 0.82, 7.72], $A_z^{\mathrm{eff}}(^{59}\mathrm{Co})$ = 919.2 MHz and the linewidth was set to 1 MHz. The $\mathbf{g}^{\mathrm{eff}}$- and $\mathbf{A}^{\mathrm{eff}}$-tensors were assumed to be collinear. Any strain effect of the tensor to the linewidth was not included. The simulated spectra (stick) were obtained by using EasySpin (ver. 6.0.0-dev.29) [8]. No peak was expected to appear above 4 GHz, and the spectral pattern is rather simple.

2. Complex 2

a) Perturbed energies for the case of non-collinear ZFS and **A**-tensors

Here, we derive the perturbed energies of the spin Hamiltonian including the ZFS, electron Zeeman and hyperfine interactions in the case of non-collinear ZFS and **A**-tensors with a nuclear spin of $I = 7/2$, in which the D_z- and A_z-axes are parallel. The zeroth-order energies and eigen-functions from the non-perturbed spin Hamiltonian are the same as the previous section since the ZFS and **g**-tensors are assumed to be collinear. The matrix representation of the hyperfine tensor transformed by a unitary matrix is given as follows,

$$\mathbf{A} = \begin{pmatrix} A_x & 0 & 0 \\ 0 & A_y & 0 \\ 0 & 0 & A_z \end{pmatrix} \to \mathbf{A}' = \begin{pmatrix} A'_{xx} & A'_{xy} & 0 \\ A'_{yx} & A'_{yy} & 0 \\ 0 & 0 & A'_{zz} \end{pmatrix}$$

Then, the perturbed Hamiltonian H' can be denoted as follows.

$$H' = H_{\mathrm{hfs}} = \boldsymbol{S} \cdot \mathbf{A}' \cdot \boldsymbol{I} = S_x A'_{xx} I_x + S_x A'_{xy} I_y + S_y A'_{yx} I_x + S_y A'_{yy} I_y + S_z A'_{zz} I_z$$

The matrix elements of the hyperfine structure Hamiltonian in the basis of $|M_S, M_I>$ are

$$\langle M'_S, M'_I | H_{\mathrm{hfs}} | M_S, M_I \rangle = \delta_{M_S M'_S} \delta_{M_I M'_I} M_S M_I A'_{zz}$$

$$\langle M_S - 1, M'_I | H_{\mathrm{hfs}} | M_S, M_I \rangle = \frac{1}{16} \delta_{M_I M'_I \pm 1} \sqrt{15 - 4M_S(M_S - 1)} \sqrt{63 - 4M_I M'_I} \left(A'_{xx} \pm iA'_{xy} + iA'_{yx} \mp A'_{yy} \right)$$

$$\langle M_S + 1, M'_I | H_{\mathrm{hfs}} | M_S, M_I \rangle = \langle M_S - 1, M_I | H_{\mathrm{hfs}} | M_S, M'_I \rangle^*$$

where

$$\delta_{ij} = \begin{cases} 1, & i = j \\ 0, & i \neq j \end{cases}$$

The upper and lower signs should be chosen in the double sign. The matrix representation of the hyperfine splitting Hamiltonian expanded by the spin wavefunctions are given as follows.

$$\left\langle \psi_{M_S = \pm\frac{3}{2}}(M'_I) \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S = \pm\frac{3}{2}}(M_I) \right\rangle = \pm \frac{M_I}{2} \delta_{M_I M'_I} A_{zz} (2\cos 2\theta_\pm + 1)$$

$$\left\langle \psi_{M_S = \pm\frac{1}{2}}(M'_I) \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S = \pm\frac{1}{2}}(M_I) \right\rangle = \pm \frac{M_I}{2} \delta_{M_I M'_I} A_{zz} (2\cos 2\theta_\mp - 1)$$

$$\left\langle \psi_{M_S = +\frac{1}{2}}(M'_I) \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S = +\frac{3}{2}}(M_I) \right\rangle = \frac{1}{2} \sqrt{63 - 4M_I M'_I} \delta_{M_I M'_I \pm 1} \left[C_{+sc} \left(A'_{xx} \pm iA'_{xy} \right) + C_{-sc} \left(iA'_{yx} \mp A'_{yy} \right) \right]$$

$$\left\langle \psi_{M_S = -\frac{1}{2}}(M'_I) \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S = +\frac{3}{2}}(M_I) \right\rangle = \left\langle \psi_{M_S = +\frac{3}{2}}(M'_I) \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S = -\frac{1}{2}}(M_I) \right\rangle = -M_I \delta_{M_I M'_I} A_{zz} \sin 2\theta_+$$

$$\left\langle \psi_{M_S = -\frac{3}{2}}(M'_I) \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S = +\frac{3}{2}}(M_I) \right\rangle = \frac{1}{2} \sqrt{63 - 4M_I M'_I} \delta_{M_I M'_I \pm 1} \left[S_{+ss} \left(A'_{xx} \pm iA'_{xy} \right) + S_{-ss} \left(iA'_{yx} \mp A'_{yy} \right) \right]$$

$$\left\langle \psi_{M_S = +\frac{3}{2}}(M'_I) \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S = +\frac{1}{2}}(M_I) \right\rangle = \left\langle \psi_{M_S = +\frac{1}{2}}(M_I) \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S = +\frac{3}{2}}(M'_I) \right\rangle^*$$

$$\left\langle \psi_{M_S = -\frac{1}{2}}(M'_I) \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S = +\frac{1}{2}}(M_I) \right\rangle = \frac{1}{2} \sqrt{63 - 4M_I M'_I} \delta_{M_I M'_I \pm 1} \left[S_{+cc} \left(A'_{xx} \pm iA'_{xy} \right) - S_{-cc} \left(iA'_{yx} \mp A'_{yy} \right) \right]$$

$$\left\langle \psi_{M_S = -\frac{3}{2}}(M'_I) \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S = +\frac{1}{2}}(M_I) \right\rangle = \left\langle \psi_{M_S = +\frac{1}{2}}(M'_I) \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S = -\frac{3}{2}}(M_I) \right\rangle = M_I \delta_{M_I M'_I} A_{zz} \sin 2\theta_-$$

$$\left\langle \psi_{M_S=+\frac{1}{2}}(M_I') \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=-\frac{1}{2}}(M_I) \right\rangle = \left\langle \psi_{M_S=-\frac{1}{2}}(M_I) \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=+\frac{1}{2}}(M_I') \right\rangle^*$$

$$\left\langle \psi_{M_S=-\frac{3}{2}}(M_I') \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=-\frac{1}{2}}(M_I) \right\rangle = \frac{1}{2}\sqrt{63 - 4M_I M_I'}\,\delta_{M_I M_I' \pm 1}\left[C_{+cs}\left(A'_{xx} \pm iA'_{xy}\right) + C_{-cs}\left(iA'_{yx} \mp A'_{yy}\right)\right]$$

$$\left\langle \psi_{M_S=+\frac{3}{2}}(M_I') \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=-\frac{3}{2}}(M_I) \right\rangle = \left\langle \psi_{M_S=-\frac{3}{2}}(M_I) \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=+\frac{3}{2}}(M_I') \right\rangle^*$$

$$\left\langle \psi_{M_S=-\frac{1}{2}}(M_I') \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=-\frac{3}{2}}(M_I) \right\rangle = \left\langle \psi_{M_S=-\frac{3}{2}}(M_I) \middle| H_{\mathrm{hfs}} \middle| \psi_{M_S=-\frac{1}{2}}(M_I') \right\rangle^*$$

where

$$C_{\pm sc} = \frac{1}{4}\left(\sqrt{3}\cos\theta_+ \cos\theta_- \pm 2\sin\theta_+ \cos\theta_- - \sqrt{3}\sin\theta_+ \sin\theta_-\right)$$

$$S_{\pm ss} = \frac{1}{4}\left(\sqrt{3}\cos\theta_+ \sin\theta_- \pm 2\sin\theta_+ \sin\theta_- + \sqrt{3}\sin\theta_+ \cos\theta_-\right)$$

$$S_{\pm cc} = \frac{1}{4}\left(-\sqrt{3}\cos\theta_+ \sin\theta_- \pm 2\cos\theta_+ \cos\theta_- - \sqrt{3}\sin\theta_+ \cos\theta_-\right)$$

$$C_{\pm cs} = \frac{1}{4}\left(-\sqrt{3}\sin\theta_+ \sin\theta_- \pm 2\cos\theta_+ \sin\theta_- + \sqrt{3}\cos\theta_+ \cos\theta_-\right)$$

The zeroth-order energies, as shown below, are the same ones as in the previous case.

$$\varepsilon^{(0)}_{\pm\frac{3}{2}} = \pm\frac{1}{2}g_{zz}\beta B - \sqrt{(D \pm g_{zz}\beta B)^2 + 3E^2}$$

$$\varepsilon^{(0)}_{\pm\frac{1}{2}} = \mp\frac{1}{2}g_{zz}\beta B + \sqrt{(D \mp g_{zz}\beta B)^2 + 3E^2}$$

The first-order energies are also the same ones as in the previous case.

$$\varepsilon^{(1)}_{\pm\frac{3}{2},M_I} = \pm\frac{M_I}{2}A'_{zz}(2\cos 2\theta_\pm + 1)$$

$$\varepsilon^{(1)}_{\pm\frac{1}{2},M_I} = \pm\frac{M_I}{2}A'_{zz}(2\cos 2\theta_\mp - 1)$$

where the upper and the lower signs should be chosen in the double sign.

The second-order energies are given as follows;

$$\begin{aligned}
\varepsilon^{(2)}_{+\frac{3}{2},+\frac{7}{2}} &= \frac{7\left|C_{+sc}\left(A'_{xx} + iA'_{xy}\right) + C_{-sc}\left(iA'_{yx} - A'_{yy}\right)\right|^2}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{49}{4}{A'_{zz}}^2 \sin^2 2\theta_+}{-2\sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} \\
&\quad + \frac{7\left|S_{+ss}\left(A'_{xx} + iA'_{xy}\right) + S_{-ss}\left(iA'_{yx} - A'_{yy}\right)\right|^2}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} \\
&= \frac{7\left[\left(C_{+sc}A'_{xx} - C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} + C_{-sc}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{49}{4}{A'_{zz}}^2 \sin^2 2\theta_+}{-2\sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} \\
&\quad + \frac{7\left[\left(S_{+ss}A'_{xx} - S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} + S_{-ss}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}
\end{aligned}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},+\frac{5}{2}} = \frac{7\left[\left(C_{+sc}A'_{xx} + C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} - C_{-sc}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D+g_{zz}\beta B)^2+3E^2} - \sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$
$$+ \frac{12\left[\left(C_{+sc}A'_{xx} - C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} + C_{-sc}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D+g_{zz}\beta B)^2+3E^2} - \sqrt{(D-g_{zz}\beta B)^2+3E^2}} + \frac{\frac{25}{4}{A'_{zz}}^2 \sin^2 2\theta_+}{-2\sqrt{(D+g_{zz}\beta B)^2+3E^2}}$$
$$+ \frac{7\left[\left(S_{+ss}A'_{xx} + S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} - S_{-ss}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D+g_{zz}\beta B)^2+3E^2} + \sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$
$$+ \frac{12\left[\left(S_{+ss}A'_{xx} - S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} + S_{-ss}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D+g_{zz}\beta B)^2+3E^2} + \sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},+\frac{3}{2}} = \frac{12\left[\left(C_{+sc}A'_{xx} + C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} - C_{-sc}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D+g_{zz}\beta B)^2+3E^2} - \sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$
$$+ \frac{15\left[\left(C_{+sc}A'_{xx} - C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} + C_{-sc}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D+g_{zz}\beta B)^2+3E^2} - \sqrt{(D-g_{zz}\beta B)^2+3E^2}} + \frac{\frac{9}{4}{A'_{zz}}^2 \sin^2 2\theta_+}{-2\sqrt{(D+g_{zz}\beta B)^2+3E^2}}$$
$$+ \frac{12\left[\left(S_{+ss}A'_{xx} + S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} - S_{-ss}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D+g_{zz}\beta B)^2+3E^2} + \sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$
$$+ \frac{15\left[\left(S_{+ss}A'_{xx} - S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} + S_{-ss}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D+g_{zz}\beta B)^2+3E^2} + \sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},+\frac{1}{2}} = \frac{15\left[\left(C_{+sc}A'_{xx} + C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} - C_{-sc}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D+g_{zz}\beta B)^2+3E^2} - \sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$
$$+ \frac{16\left[\left(C_{+sc}A'_{xx} - C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} + C_{-sc}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D+g_{zz}\beta B)^2+3E^2} - \sqrt{(D-g_{zz}\beta B)^2+3E^2}} + \frac{\frac{1}{4}{A'_{zz}}^2 \sin^2 2\theta_+}{-2\sqrt{(D+g_{zz}\beta B)^2+3E^2}}$$
$$+ \frac{15\left[\left(S_{+ss}A'_{xx} + S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} - S_{-ss}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D+g_{zz}\beta B)^2+3E^2} + \sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$
$$+ \frac{16\left[\left(S_{+ss}A'_{xx} - S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} + S_{-ss}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D+g_{zz}\beta B)^2+3E^2} + \sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},-\frac{1}{2}} = \frac{16\left[\left(C_{+sc}A'_{xx} + C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} - C_{-sc}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D+g_{zz}\beta B)^2+3E^2} - \sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$
$$+ \frac{15\left[\left(C_{+sc}A'_{xx} - C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} + C_{-sc}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D+g_{zz}\beta B)^2+3E^2} - \sqrt{(D-g_{zz}\beta B)^2+3E^2}} + \frac{\frac{1}{4}{A'_{zz}}^2 \sin^2 2\theta_+}{-2\sqrt{(D+g_{zz}\beta B)^2+3E^2}}$$
$$+ \frac{16\left[\left(S_{+ss}A'_{xx} + S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} - S_{-ss}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D+g_{zz}\beta B)^2+3E^2} + \sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$
$$+ \frac{15\left[\left(S_{+ss}A'_{xx} - S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} + S_{-ss}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D+g_{zz}\beta B)^2+3E^2} + \sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},-\frac{3}{2}} + \frac{12\left[\left(C_{+sc}A'_{xx} - C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} + C_{-sc}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{9}{4}{A'_{zz}}^2 \sin^2 2\theta_+}{-2\sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{15\left[\left(S_{+ss}A'_{xx} + S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} - S_{-ss}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{12\left[\left(S_{+ss}A'_{xx} - S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} + S_{-ss}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},-\frac{5}{2}} = \frac{12\left[\left(C_{+sc}A'_{xx} + C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} - C_{-sc}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{7\left[\left(C_{+sc}A'_{xx} - C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} + C_{-sc}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{25}{4}{A'_{zz}}^2 \sin^2 2\theta_+}{-2\sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{12\left[\left(S_{+ss}A'_{xx} + S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} - S_{-ss}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{7\left[\left(S_{+ss}A'_{xx} - S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} + S_{-ss}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{3}{2},-\frac{7}{2}} = \frac{7\left[\left(C_{+sc}A'_{xx} + C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} - C_{-sc}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{49}{4}{A'_{zz}}^2 \sin^2 2\theta_+}{-2\sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{7\left[\left(S_{+ss}A'_{xx} + S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} - S_{-ss}A'_{yx}\right)^2\right]}{g_{zz}\beta B - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},+\frac{7}{2}} = \frac{7\left[\left(C_{+sc}A'_{xx} + C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} - C_{-sc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{7\left[\left(S_{+cc}A'_{xx} + S_{-cc}A'_{yy}\right)^2 + \left(S_{+cc}A'_{xy} - S_{-cc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{49}{4}{A'_{zz}}^2 \sin^2 2\theta_-}{2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},+\frac{5}{2}} = \frac{7\left[\left(C_{+sc}A'_{xx} - C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} + C_{-sc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{12\left[\left(C_{+sc}A'_{xx} + C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} - C_{-sc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{7\left[\left(S_{+cc}A'_{xx} - S_{-cc}A'_{yy}\right)^2 + \left(S_{+cc}A'_{xy} + S_{-cc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{12\left[\left(S_{+cc}A'_{xx} + S_{-cc}A'_{yy}\right)^2 + \left(S_{+cc}A'_{xy} - S_{-cc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{25}{4}{A'_{zz}}^2 \sin^2 2\theta_-}{2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},+\frac{3}{2}} = \frac{12\left[\left(C_{+sc}A'_{xx} - C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} + C_{-sc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{15\left[\left(C_{+sc}A'_{xx} + C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} - C_{-sc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{12\left[\left(S_{+cc}A'_{xx} - S_{-cc}A'_{yy}\right)^2 + \left(S_{+cc}A'_{xy} + S_{-cc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{15\left[\left(S_{+cc}A'_{xx} + S_{-cc}A'_{yy}\right)^2 + \left(S_{+cc}A'_{xy} - S_{-cc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{9}{4}{A'_{zz}}^2 \sin^2 2\theta_-}{2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},+\frac{1}{2}} = \frac{15\left[\left(C_{+sc}A'_{xx} - C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} + C_{-sc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{16\left[\left(C_{+sc}A'_{xx} + C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} - C_{-sc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{15\left[\left(S_{+cc}A'_{xx} - S_{-cc}A'_{yy}\right)^2 + \left(S_{+cc}A'_{xy} + S_{-cc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{16\left[\left(S_{+cc}A'_{xx} + S_{-cc}A'_{yy}\right)^2 + \left(S_{+cc}A'_{xy} - S_{-cc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{1}{4}{A'_{zz}}^2 \sin^2 2\theta_-}{2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},-\frac{1}{2}} = \frac{16\left[\left(C_{+sc}A'_{xx} - C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} + C_{-sc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{15\left[\left(C_{+sc}A'_{xx} + C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} - C_{-sc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{16\left[\left(S_{+cc}A'_{xx} - S_{-cc}A'_{yy}\right)^2 + \left(S_{+cc}A'_{xy} + S_{-cc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{15\left[\left(S_{+cc}A'_{xx} + S_{-cc}A'_{yy}\right)^2 + \left(S_{+cc}A'_{xy} - S_{-cc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{1}{4}{A'_{zz}}^2 \sin^2 2\theta_-}{2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},-\frac{3}{2}} = \frac{15\left[\left(C_{+sc}A'_{xx} - C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} + C_{-sc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{12\left[\left(C_{+sc}A'_{xx} + C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} - C_{-sc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{15\left[\left(S_{+cc}A'_{xx} - S_{-cc}A'_{yy}\right)^2 + \left(S_{+cc}A'_{xy} + S_{-cc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{12\left[\left(S_{+cc}A'_{xx} + S_{-cc}A'_{yy}\right)^2 + \left(S_{+cc}A'_{xy} - S_{-cc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{9}{4}{A'_{zz}}^2 \sin^2 2\theta_-}{2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},-\frac{5}{2}} = \frac{12\left[\left(C_{+sc}A'_{xx} - C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} + C_{-sc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{7\left[\left(C_{+sc}A'_{xx} + C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} - C_{-sc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{12\left[\left(S_{+cc}A'_{xx} - S_{-cc}A'_{yy}\right)^2 + \left(S_{+cc}A'_{xy} + S_{-cc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{7\left[\left(S_{+cc}A'_{xx} + S_{-cc}A'_{yy}\right)^2 + \left(S_{+cc}A'_{xy} - S_{-cc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{25}{4}{A'_{zz}}^2 \sin^2 2\theta_-}{2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{+\frac{1}{2},-\frac{7}{2}} = \frac{7\left[\left(C_{+sc}A'_{xx} - C_{-sc}A'_{yy}\right)^2 + \left(C_{+sc}A'_{xy} + C_{-sc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{7\left[\left(S_{+cc}A'_{xx} - S_{-cc}A'_{yy}\right)^2 + \left(S_{+cc}A'_{xy} + S_{-cc}A'_{yx}\right)^2\right]}{-g_{zz}\beta B + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{49}{4}{A'_{zz}}^2 \sin^2 2\theta_-}{2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{1}{2},+\frac{7}{2}} = \frac{\frac{49}{4}{A'_{zz}}^2 \sin^2 2\theta_+}{2\sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{7\left[\left(S_{+cc}A'_{xx} - S_{-cc}A'_{yy}\right)^2 + \left(S_{+cc}A'_{xy} + S_{-cc}A'_{yx}\right)^2\right]}{g_{zz}\beta B + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{7\left[\left(C_{+cs}A'_{xx} - C_{-cs}A'_{yy}\right)^2 + \left(C_{+cs}A'_{xy} + C_{-cs}A'_{yx}\right)^2\right]}{g_{zz}\beta B + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{1}{2},+\frac{5}{2}} = \frac{\frac{25}{4}{A'_{zz}}^2 \sin^2 2\theta_+}{2\sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{7\left[\left(S_{+cc}A'_{xx} + S_{-cc}A'_{yy}\right)^2 + \left(S_{+cc}A'_{xy} - S_{-cc}A'_{yx}\right)^2\right]}{g_{zz}\beta B + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{12\left[\left(S_{+cc}A'_{xx} - S_{-cc}A'_{yy}\right)^2 + \left(S_{+cc}A'_{xy} + S_{-cc}A'_{yx}\right)^2\right]}{g_{zz}\beta B + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{7\left[\left(C_{+cs}A'_{xx} + C_{-cs}A'_{yy}\right)^2 + \left(C_{+cs}A'_{xy} - C_{-cs}A'_{yx}\right)^2\right]}{g_{zz}\beta B + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{12\left[\left(C_{+cs}A'_{xx} - C_{-cs}A'_{yy}\right)^2 + \left(C_{+cs}A'_{xy} + C_{-cs}A'_{yx}\right)^2\right]}{g_{zz}\beta B + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{1}{2},+\frac{3}{2}} = \frac{\frac{9}{4}{A'_{zz}}^2 \sin^2 2\theta_+}{2\sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{12\left[\left(S_{+cc}A'_{xx} + S_{-cc}A'_{yy}\right)^2 + \left(S_{+cc}A'_{xy} - S_{-cc}A'_{yx}\right)^2\right]}{g_{zz}\beta B + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{15\left[\left(S_{+cc}A'_{xx} - S_{-cc}A'_{yy}\right)^2 + \left(S_{+cc}A'_{xy} + S_{-cc}A'_{yx}\right)^2\right]}{g_{zz}\beta B + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{12\left[\left(C_{+cs}A'_{xx} + C_{-cs}A'_{yy}\right)^2 + \left(C_{+cs}A'_{xy} - C_{-cs}A'_{yx}\right)^2\right]}{g_{zz}\beta B + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{15\left[\left(C_{+cs}A'_{xx} - C_{-cs}A'_{yy}\right)^2 + \left(C_{+cs}A'_{xy} + C_{-cs}A'_{yx}\right)^2\right]}{g_{zz}\beta B + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$

$$
\begin{aligned}
\varepsilon^{(2)}_{-\frac{1}{2},+\frac{1}{2}} = {} & \frac{\frac{1}{4}{A'_{zz}}^2 \sin^2 2\theta_+}{2\sqrt{(D+g_{zz}\beta B)^2+3E^2}} + \frac{15\left[\left(S_{+cc}A'_{xx}+S_{-cc}A'_{yy}\right)^2+\left(S_{+cc}A'_{xy}-S_{-cc}A'_{yx}\right)^2\right]}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}-\sqrt{(D-g_{zz}\beta B)^2+3E^2}} \\
& + \frac{16\left[\left(S_{+cc}A'_{xx}-S_{-cc}A'_{yy}\right)^2+\left(S_{+cc}A'_{xy}+S_{-cc}A'_{yx}\right)^2\right]}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}-\sqrt{(D-g_{zz}\beta B)^2+3E^2}} \\
& + \frac{15\left[\left(C_{+cs}A'_{xx}+C_{-cs}A'_{yy}\right)^2+\left(C_{+cs}A'_{xy}-C_{-cs}A'_{yx}\right)^2\right]}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}+\sqrt{(D-g_{zz}\beta B)^2+3E^2}} \\
& + \frac{16\left[\left(C_{+cs}A'_{xx}-C_{-cs}A'_{yy}\right)^2+\left(C_{+cs}A'_{xy}+C_{-cs}A'_{yx}\right)^2\right]}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}+\sqrt{(D-g_{zz}\beta B)^2+3E^2}}
\end{aligned}
$$

$$
\begin{aligned}
\varepsilon^{(2)}_{-\frac{1}{2},-\frac{1}{2}} = {} & \frac{\frac{1}{4}{A'_{zz}}^2 \sin^2 2\theta_+}{2\sqrt{(D+g_{zz}\beta B)^2+3E^2}} + \frac{16\left[\left(S_{+cc}A'_{xx}+S_{-cc}A'_{yy}\right)^2+\left(S_{+cc}A'_{xy}-S_{-cc}A'_{yx}\right)^2\right]}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}-\sqrt{(D-g_{zz}\beta B)^2+3E^2}} \\
& + \frac{15\left[\left(S_{+cc}A'_{xx}-S_{-cc}A'_{yy}\right)^2+\left(S_{+cc}A'_{xy}+S_{-cc}A'_{yx}\right)^2\right]}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}-\sqrt{(D-g_{zz}\beta B)^2+3E^2}} \\
& + \frac{16\left[\left(C_{+cs}A'_{xx}+C_{-cs}A'_{yy}\right)^2+\left(C_{+cs}A'_{xy}-C_{-cs}A'_{yx}\right)^2\right]}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}+\sqrt{(D-g_{zz}\beta B)^2+3E^2}} \\
& + \frac{15\left[\left(C_{+cs}A'_{xx}-C_{-cs}A'_{yy}\right)^2+\left(C_{+cs}A'_{xy}+C_{-cs}A'_{yx}\right)^2\right]}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}+\sqrt{(D-g_{zz}\beta B)^2+3E^2}}
\end{aligned}
$$

$$
\begin{aligned}
\varepsilon^{(2)}_{-\frac{1}{2},-\frac{3}{2}} = {} & \frac{\frac{9}{4}{A'_{zz}}^2 \sin^2 2\theta_+}{2\sqrt{(D+g_{zz}\beta B)^2+3E^2}} + \frac{15\left[\left(S_{+cc}A'_{xx}+S_{-cc}A'_{yy}\right)^2+\left(S_{+cc}A'_{xy}-S_{-cc}A'_{yx}\right)^2\right]}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}-\sqrt{(D-g_{zz}\beta B)^2+3E^2}} \\
& + \frac{12\left[\left(S_{+cc}A'_{xx}-S_{-cc}A'_{yy}\right)^2+\left(S_{+cc}A'_{xy}+S_{-cc}A'_{yx}\right)^2\right]}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}-\sqrt{(D-g_{zz}\beta B)^2+3E^2}} \\
& + \frac{15\left[\left(C_{+cs}A'_{xx}+C_{-cs}A'_{yy}\right)^2+\left(C_{+cs}A'_{xy}-C_{-cs}A'_{yx}\right)^2\right]}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}+\sqrt{(D-g_{zz}\beta B)^2+3E^2}} \\
& + \frac{12\left[\left(C_{+cs}A'_{xx}-C_{-cs}A'_{yy}\right)^2+\left(C_{+cs}A'_{xy}+C_{-cs}A'_{yx}\right)^2\right]}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}+\sqrt{(D-g_{zz}\beta B)^2+3E^2}}
\end{aligned}
$$

$$
\begin{aligned}
\varepsilon^{(2)}_{-\frac{1}{2},-\frac{5}{2}} = {} & \frac{\frac{25}{4}{A'_{zz}}^2 \sin^2 2\theta_+}{2\sqrt{(D+g_{zz}\beta B)^2+3E^2}} + \frac{12\left[\left(S_{+cc}A'_{xx}+S_{-cc}A'_{yy}\right)^2+\left(S_{+cc}A'_{xy}-S_{-cc}A'_{yx}\right)^2\right]}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}-\sqrt{(D-g_{zz}\beta B)^2+3E^2}} \\
& + \frac{7\left[\left(S_{-cc}A'_{xx}-S_{+cc}A'_{yy}\right)^2+\left(S_{-cc}A'_{xy}+S_{+cc}A'_{yx}\right)^2\right]}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}-\sqrt{(D-g_{zz}\beta B)^2+3E^2}} \\
& + \frac{12\left[\left(C_{+cs}A'_{xx}+C_{-cs}A'_{yy}\right)^2+\left(C_{+cs}A'_{xy}-C_{-cs}A'_{yx}\right)^2\right]}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}+\sqrt{(D-g_{zz}\beta B)^2+3E^2}} \\
& + \frac{7\left[\left(C_{+cs}A'_{xx}-C_{-cs}A'_{yy}\right)^2+\left(C_{+cs}A'_{xy}+C_{-cs}A'_{yx}\right)^2\right]}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}+\sqrt{(D-g_{zz}\beta B)^2+3E^2}}
\end{aligned}
$$

$$\varepsilon^{(2)}_{-\frac{1}{2},-\frac{7}{2}} = \frac{\frac{49}{4}{A'_{zz}}^2 \sin^2 2\theta_+}{2\sqrt{(D+g_{zz}\beta B)^2+3E^2}} + \frac{7\left[\left(S_{+cc}A'_{xx}+S_{-cc}A'_{yy}\right)^2+\left(S_{+cc}A'_{xy}-S_{-cc}A'_{yx}\right)^2\right]}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}-\sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$
$$+\frac{7\left[\left(C_{+cs}A'_{xx}+C_{-cs}A'_{yy}\right)^2+\left(C_{+cs}A'_{xy}-C_{-cs}A'_{yx}\right)^2\right]}{g_{zz}\beta B+\sqrt{(D+g_{zz}\beta B)^2+3E^2}+\sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},+\frac{7}{2}} = \frac{7\left[\left(S_{+ss}A'_{xx}+S_{-ss}A'_{yy}\right)^2+\left(S_{+ss}A'_{xy}-S_{-ss}A'_{yx}\right)^2\right]}{-g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}+\sqrt{(D+g_{zz}\beta B)^2+3E^2}} + \frac{\frac{49}{4}{A'_{zz}}^2 \sin^2 2\theta_-}{-2\sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$
$$+\frac{7\left[\left(C_{+cs}A'_{xx}+C_{-cs}A'_{yy}\right)^2+\left(C_{+cs}A'_{xy}-C_{-cs}A'_{yx}\right)^2\right]}{-g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}-\sqrt{(D+g_{zz}\beta B)^2+3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},+\frac{5}{2}} = \frac{7\left[\left(S_{+ss}A'_{xx}-S_{-ss}A'_{yy}\right)^2+\left(S_{+ss}A'_{xy}+S_{-ss}A'_{yx}\right)^2\right]}{-g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}+\sqrt{(D+g_{zz}\beta B)^2+3E^2}}$$
$$+\frac{12\left[\left(S_{+ss}A'_{xx}+S_{-ss}A'_{yy}\right)^2+\left(S_{+ss}A'_{xy}-S_{-ss}A'_{yx}\right)^2\right]}{-g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}+\sqrt{(D+g_{zz}\beta B)^2+3E^2}} + \frac{\frac{25}{4}{A'_{zz}}^2 \sin^2 2\theta_-}{-2\sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$
$$+\frac{7\left[\left(C_{+cs}A'_{xx}-C_{-cs}A'_{yy}\right)^2+\left(C_{+cs}A'_{xy}+C_{-cs}A'_{yx}\right)^2\right]}{-g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}-\sqrt{(D+g_{zz}\beta B)^2+3E^2}}$$
$$+\frac{12\left[\left(C_{+cs}A'_{xx}+C_{-cs}A'_{yy}\right)^2+\left(C_{+cs}A'_{xy}-C_{-cs}A'_{yx}\right)^2\right]}{-g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}-\sqrt{(D+g_{zz}\beta B)^2+3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},+\frac{3}{2}} = \frac{12\left[\left(S_{+ss}A'_{xx}-S_{-ss}A'_{yy}\right)^2+\left(S_{+ss}A'_{xy}+S_{-ss}A'_{yx}\right)^2\right]}{-g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}+\sqrt{(D+g_{zz}\beta B)^2+3E^2}}$$
$$+\frac{15\left[\left(S_{+ss}A'_{xx}+S_{-ss}A'_{yy}\right)^2+\left(S_{+ss}A'_{xy}-S_{-ss}A'_{yx}\right)^2\right]}{-g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}+\sqrt{(D+g_{zz}\beta B)^2+3E^2}} + \frac{\frac{9}{4}{A'_{zz}}^2 \sin^2 2\theta_-}{-2\sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$
$$+\frac{12\left[\left(C_{+cs}A'_{xx}-C_{-cs}A'_{yy}\right)^2+\left(C_{+cs}A'_{xy}+C_{-cs}A'_{yx}\right)^2\right]}{-g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}-\sqrt{(D+g_{zz}\beta B)^2+3E^2}}$$
$$+\frac{15\left[\left(C_{+cs}A'_{xx}+C_{-cs}A'_{yy}\right)^2+\left(C_{+cs}A'_{xy}-C_{-cs}A'_{yx}\right)^2\right]}{-g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}-\sqrt{(D+g_{zz}\beta B)^2+3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},+\frac{1}{2}} = \frac{15\left[\left(S_{+ss}A'_{xx}-S_{-ss}A'_{yy}\right)^2+\left(S_{+ss}A'_{xy}+S_{-ss}A'_{yx}\right)^2\right]}{-g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}+\sqrt{(D+g_{zz}\beta B)^2+3E^2}}$$
$$+\frac{16\left[\left(S_{+ss}A'_{xx}+S_{-ss}A'_{yy}\right)^2+\left(S_{+ss}A'_{xy}-S_{-ss}A'_{yx}\right)^2\right]}{-g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}+\sqrt{(D+g_{zz}\beta B)^2+3E^2}} + \frac{\frac{1}{4}{A'_{zz}}^2 \sin^2 2\theta_-}{-2\sqrt{(D-g_{zz}\beta B)^2+3E^2}}$$
$$+\frac{15\left[\left(C_{+cs}A'_{xx}-C_{-cs}A'_{yy}\right)^2+\left(C_{+cs}A'_{xy}+C_{-cs}A'_{yx}\right)^2\right]}{-g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}-\sqrt{(D+g_{zz}\beta B)^2+3E^2}}$$
$$+\frac{16\left[\left(C_{+cs}A'_{xx}+C_{-cs}A'_{yy}\right)^2+\left(C_{+cs}A'_{xy}-C_{-cs}A'_{yx}\right)^2\right]}{-g_{zz}\beta B-\sqrt{(D-g_{zz}\beta B)^2+3E^2}-\sqrt{(D+g_{zz}\beta B)^2+3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},-\frac{1}{2}} = \frac{16\left[\left(S_{+ss}A'_{xx} - S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} + S_{-ss}A'_{yx}\right)^2\right]}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{15\left[\left(S_{+ss}A'_{xx} + S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} - S_{-ss}A'_{yx}\right)^2\right]}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{1}{4}{A'_{zz}}^2 \sin^2 2\theta_-}{-2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{16\left[\left(C_{+cs}A'_{xx} - C_{-cs}A'_{yy}\right)^2 + \left(C_{+cs}A'_{xy} + C_{-cs}A'_{yx}\right)^2\right]}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{15\left[\left(C_{+cs}A'_{xx} + C_{-cs}A'_{yy}\right)^2 + \left(C_{+cs}A'_{xy} - C_{-cs}A'_{yx}\right)^2\right]}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},-\frac{3}{2}} = \frac{15\left[\left(S_{+ss}A'_{xx} - S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} + S_{-ss}A'_{yx}\right)^2\right]}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{12\left[\left(S_{+ss}A'_{xx} + S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} - S_{-ss}A'_{yx}\right)^2\right]}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{9}{4}{A'_{zz}}^2 \sin^2 2\theta_-}{-2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{15\left[\left(C_{+cs}A'_{xx} - C_{-cs}A'_{yy}\right)^2 + \left(C_{+cs}A'_{xy} + C_{-cs}A'_{yx}\right)^2\right]}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{12\left[\left(C_{+cs}A'_{xx} + C_{-cs}A'_{yy}\right)^2 + \left(C_{+cs}A'_{xy} - C_{-cs}A'_{yx}\right)^2\right]}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},-\frac{5}{2}} = \frac{12\left[\left(S_{+ss}A'_{xx} - S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} + S_{-ss}A'_{yx}\right)^2\right]}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{7\left[\left(S_{+ss}A'_{xx} + S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} - S_{-ss}A'_{yx}\right)^2\right]}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{25}{4}{A'_{zz}}^2 \sin^2 2\theta_-}{-2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{12\left[\left(C_{+cs}A'_{xx} - C_{-cs}A'_{yy}\right)^2 + \left(C_{+cs}A'_{xy} + C_{-cs}A'_{yx}\right)^2\right]}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{7\left[\left(C_{+cs}A'_{xx} + C_{-cs}A'_{yy}\right)^2 + \left(C_{+cs}A'_{xy} - C_{-cs}A'_{yx}\right)^2\right]}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$

$$\varepsilon^{(2)}_{-\frac{3}{2},-\frac{7}{2}} = \frac{7\left[\left(S_{+ss}A'_{xx} - S_{-ss}A'_{yy}\right)^2 + \left(S_{+ss}A'_{xy} + S_{-ss}A'_{yx}\right)^2\right]}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} + \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}} + \frac{\frac{49}{4}{A'_{zz}}^2 \sin^2 2\theta_-}{-2\sqrt{(D - g_{zz}\beta B)^2 + 3E^2}}$$
$$+ \frac{7\left[\left(C_{+cs}A'_{xx} - C_{-cs}A'_{yy}\right)^2 + \left(C_{+cs}A'_{xy} + C_{-cs}A'_{yx}\right)^2\right]}{-g_{zz}\beta B - \sqrt{(D - g_{zz}\beta B)^2 + 3E^2} - \sqrt{(D + g_{zz}\beta B)^2 + 3E^2}}$$

In summary, when the ZFS and **g**-tensors are collinear but the ZFS and **A**-tensors are non-collinear and the magnetic field is aligned to the *z*-axis of the principal axis coordinate system, the perturbed energies in the second order are represented in a similar way as in the previous section, except for substituting only the second-order energies. The expressions explicitly given above for the case including the hyperfine interactions as the perturbation are for the first time derived and they are extremely accurate for the sizable ZFS tensor.

b) ESR Analysis of complex **2**.

Table S6. The possible combinations of the $\mathbf{g}^{\mathrm{eff}}/\mathbf{g}^{\mathrm{true}}$ relationships

Case	g^{true}	g^{eff}	g^{eff}/g^{true}	Figure #
1	x	0.92	$1-\frac{1-3\lambda}{\sqrt{1+3\lambda^2}}$	Figs. S14, S15, S16 S17 and S18
	y	1.21	$1-\frac{1+3\lambda}{\sqrt{1+3\lambda^2}}$	
	z	8.8	$1-\frac{2}{\sqrt{1+3\lambda^2}}$	
2	x	1.21	$1-\frac{1-3\lambda}{\sqrt{1+3\lambda^2}}$	Figs. 2 and 3 in the main text and Figs. S19, S20, S21, S22 and S23
	y	0.92	$1-\frac{1+3\lambda}{\sqrt{1+3\lambda^2}}$	
	z	8.8	$1-\frac{2}{\sqrt{1+3\lambda^2}}$	

* Case 2, which is highlighted in yellow, provides the most reasonably simulated ESR spectrum.

Case 1 (see Table S6 for the $g^{\mathrm{eff}}/g^{\mathrm{true}}$ relationship)

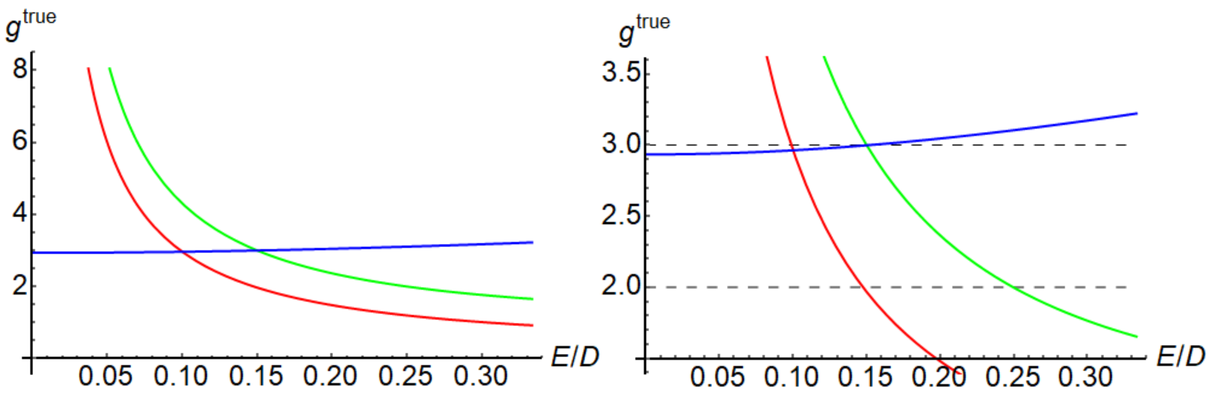

Figure S14. The calculated g^{true}-values by use of the g^{eff}-value and $g^{\mathrm{eff}}/g^{\mathrm{true}}$ relationships. $\frac{0.92}{g_{XX}^{\mathrm{true}}}=1-\frac{1-3\lambda}{\sqrt{1+3\lambda^2}}$, $\frac{1.21}{g_{YY}^{\mathrm{true}}}=1-\frac{1+3\lambda}{\sqrt{1+3\lambda^2}}$, $\frac{8.8}{g_{ZZ}^{\mathrm{true}}}=1+\frac{2}{\sqrt{1+3\lambda^2}}$ with $\lambda = E/D$. The $g^{\mathrm{eff}}/g^{\mathrm{true}}$ relationships adopted here are for the $|M_S = \pm 3/2\rangle$-dominant transitions. The inset on the right shows the expanded figure in the range of $1.5 \le g^{\mathrm{true}} \le 3.5$. The broken lines indicate $g^{\mathrm{true}} = 2.0$ and 3.0.

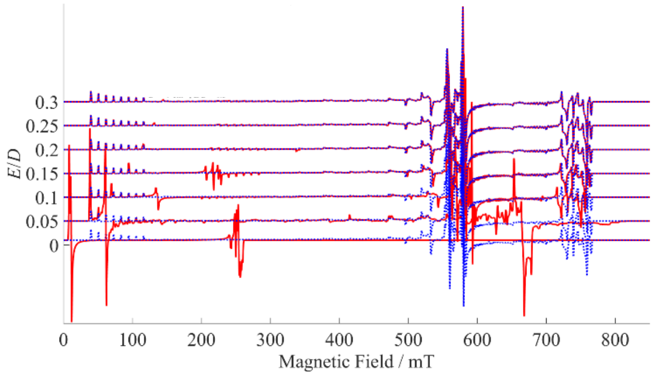

Figure S15. The simulated randomly-oriented X-band ESR spectra of complex **2** ($S = 3/2$). The spectra in blue and red are based on the fictitious spin-1/2 and true spin Hamiltonian approaches, respectively. Microwave frequency used: 9.625 GHz, the peak-to-peak linewidth: 1.0 mT, the magnetic tensors: $\mathbf{g}^{\mathrm{eff}}$ = [0.92, 1,21, 8.8], $A_x^{\mathrm{eff}}(^{59}\mathrm{Co})$ = 1345.2 MHz, D = –9 cm^{-1}, and $\mathbf{g}^{\mathrm{true}}$ and $A_z^{\mathrm{true}}(^{59}\mathrm{Co})$ for the variable E/D's are calculated by use of the g^{eff}-value and the $g^{\mathrm{eff}}/g^{\mathrm{true}}$ relationships. The ZFS and **g**-tensors were assumed to be collinear. A set of rotation angles (Euler angles) of the $\mathbf{A}^{\mathrm{true}}$-tensor with respect to the ZFS tensor were α = –90, β = 90, and γ = 36 degrees. (Note: Because only A_y-value was considered, of which the principal axis was parallel to the D_z-axis, it seemed that there was no effect on the spectra.) Any strain effect of the tensor to the linewidth was not included. The simulated spectra were obtained by using EasySpin (ver. 6.0.6) [8].

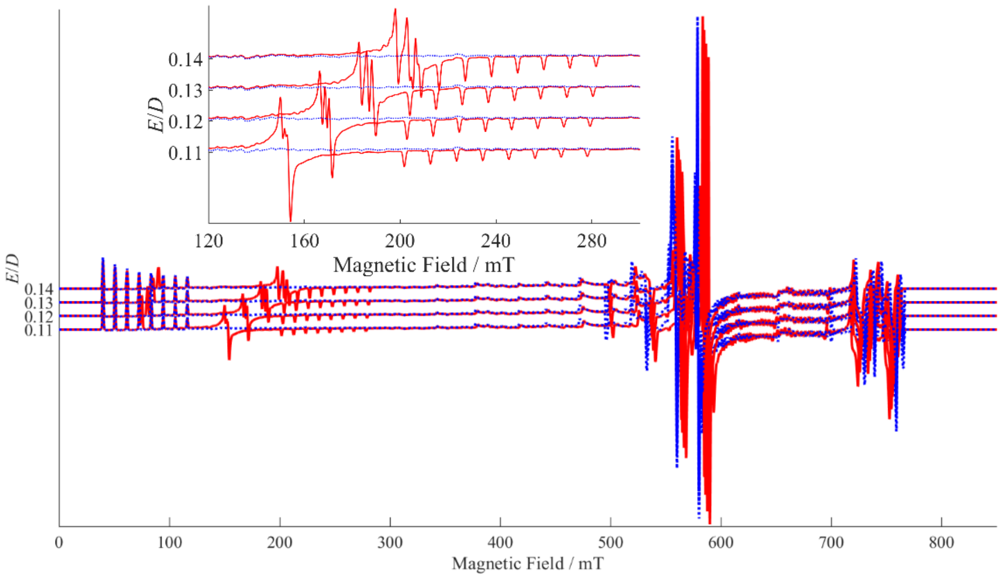

Figure S16. The simulated randomly-oriented ESR spectra of complex **2** ($S = 3/2$) as a function of E/D. The spectra in blue and red are based on the fictitious spin-1/2 and true spin Hamiltonian approaches, respectively. Microwave frequency used: 9.625 GHz, the peak-to-peak linewidth: 1.0 mT; the magnetic tensors: $\mathbf{g}^{\mathrm{eff}}$ = [0.92, 1.21, 8.80], $A_Z^{\mathrm{eff}}(^{59}\mathrm{Co})$ = 1345.2 MHz, D = –9 cm^{-1} and $\mathbf{g}^{\mathrm{true}}$ and $A_Z^{\mathrm{true}}(^{59}\mathrm{Co})$ for the variable E/D's are shown in Table S2-1. The ZFS and **g**-tensors were assumed to be collinear. A set of rotation angles (Euler angles) of the $\mathbf{A}^{\mathrm{true}}$-tensor with respect to the ZFS tensor were α = –90, β = 90, and γ = 36 degrees. (Note: Because only A_y-value was considered, of which the principal axis was parallel to the D_z-axis, it seemed that there was no effect on the spectrum.) Any strain effect of the tensor to the linewidth was not included. The simulated spectra were obtained by using EasySpin (ver. 6.0.6) [8]. Note that the resonance peaks marked by a single asterisk *, double asterisk ** and triple asterisk *** appear only for the true spin Hamiltonian approach. The inset shows the expanded spectra in the range of 120 mT to 300 mT.

Table S6. The principal values of the $\mathbf{g}^{true}$-tensor and A_{zz}-values used in the simulated spectra of complex **2** (Figures S17 and S18).

E/D	g_{XX}	g_{YY}	g_{ZZ}	E/D	g_{XX}	g_{YY}	g_{ZZ}
0.01	30.5	40.5	2.934	0.14	2.108	3.184	2.989
0.10	2.965	4.307	2.962	0.15	1.967	2.999	2.998
0.11	2.691	3.948	2.968	0.20	1.479	2.364	3.045
0.12	2.464	3.650	2.975	0.25	1.194	1.997	3.014
0.13	2.272	3.398	2.982	0.30	1.010	1.764	3.172

One of the possible sets of the magnetic parameters: $g_{XX} = 2.108$, $g_{YY} = 3.184$, $g_{ZZ} = 2.989$, $A_{ZZ}(^{59}Co) = 456.98$ MHz, $D = -9$ cm^{-1} and $E/D = +0.14$. The extra line observed at about 200 mT was simulated by using the parameters.

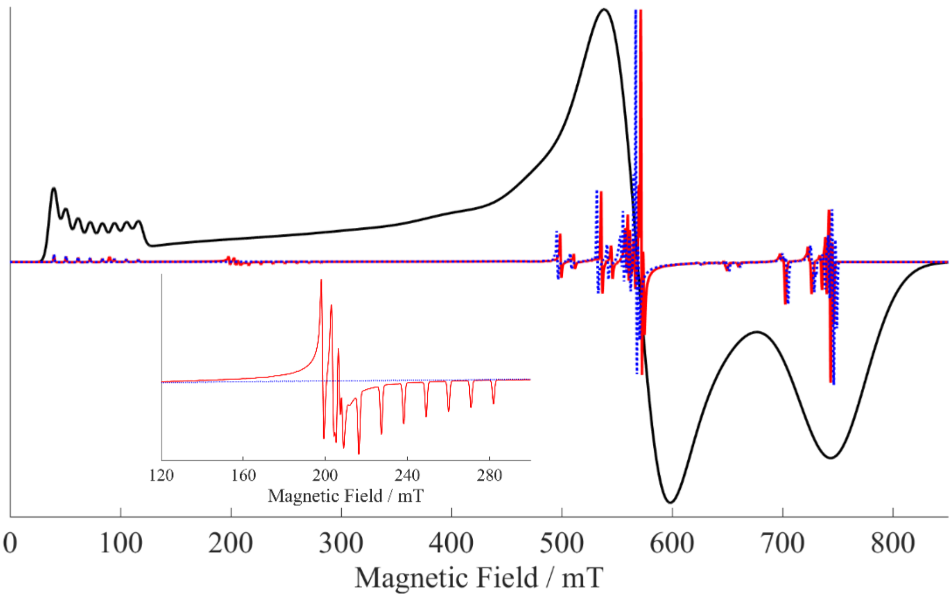

Figure S17. The simulated randomly-oriented X-band ESR spectra of complex **2** ($S = 3/2$). The spectrum in blue and red are based on the fictitious spin-1/2 and true spin Hamiltonian approaches, respectively. Microwave frequency used: 9.625 GHz, (the spectrum in black) the magnetic tensors: $g_1^{eff} = 8.8$, $g_2^{eff} = 1.21$, $g_3^{eff} = 0.92$, $A_1^{eff}(^{59}Co) = 1345$ MHz, the peak-to-peak linewidth: 8.0 mT, strain of the linewidth: [0, 900, 900] MHz; (the spectrum in blue and red) the peak-to-peak linewidth: 1.0 mT, the magnetic tensors: $g_{XX}^{eff} = 0.92$, $g_{YY}^{eff} = 1.21$, $g_{ZZ}^{eff} = 8.8$, $A_{ZZ}^{eff}(^{59}Co) = 1345$ MHz; $g_{XX}^{true} = 2.1085$, $g_{YY}^{true} = 3.18412$, $g_{ZZ}^{true} = 2.9895$, $A_{ZZ}^{true}(^{59}Co) = 456.98$ MHz, $D = -9$cm^{-1} and $E/D = +0.14$. Any strain effect of the tensor to the linewidth was not included in the spectra in blue and red. The simulated spectra were obtained by using EasySpin (ver. 6.0.6) [8]. Inset: The expanded spectra from 120 mT to 300 mT. The extralines attributed from the $|M_S = \pm1/2\rangle$-dominant transitions can be reproduced by using the true spin Hamiltonian (red line) but not by using the fictitious spin-1/2 Hamiltonian (blue line).

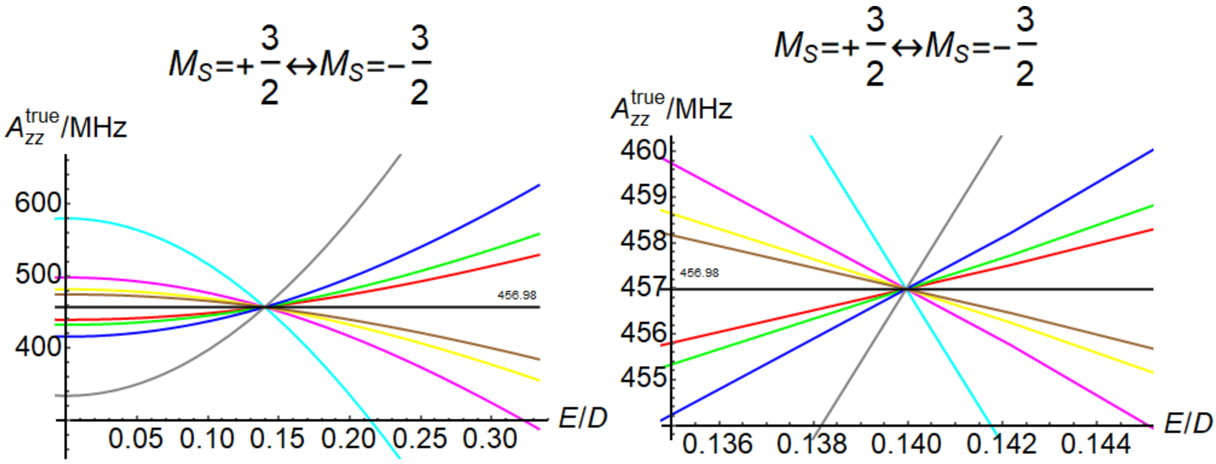

Figure S18. The E/D dependence of the A_Z^{true}-value which is the solution of eq (†) with the parameters used for the simulated spectrum in Figure S2-5. Red, green, blue, gray, cyan, magenta, yellow and brown curves correspond to the assignments to M_I = +7/2, +5/2, +3/2, +1/2, –1/2, –3/2, –5/2 and –7/2, respectively. Because all the energies and the parameters were accurate, all the curves cross at one point. The black line indicates the A_Z-value (= 456.98 MHz) calculated by using the relationship (eq (‡)) in the case of E/D = 0.14. The figure on the right shows the expanded one in the range of 0.135 ≤ E/D ≤ 0.145.

<u>*Case 2*</u> (see Table S6 for the g^{eff}/g^{true} relationship)

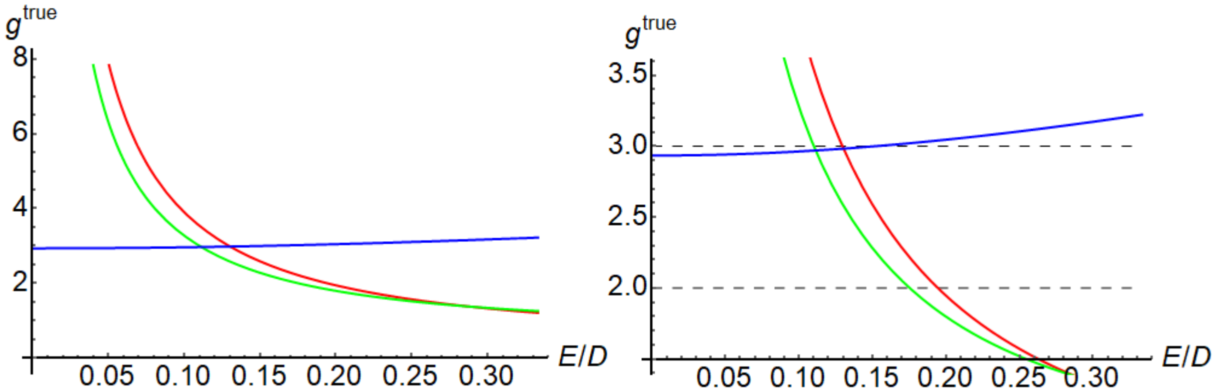

Figure S19. The calculated g^{true} values by using the g^{eff}-value and g^{eff}/g^{true} relationships. $\frac{1.21}{g_x^{true}} = 1 - \frac{1-3\lambda}{\sqrt{1+3\lambda^2}}$, $\frac{0.92}{g_y^{true}} = 1 - \frac{1+3\lambda}{\sqrt{1+3\lambda^2}}$, $\frac{8.8}{g_z^{true}} = 1 + \frac{2}{\sqrt{1+3\lambda^2}}$ with $\lambda = E/D$. The g^{eff}/g^{true} relationships adopted here are for the $|M_S$ = ±3/2>-dominant transitions.

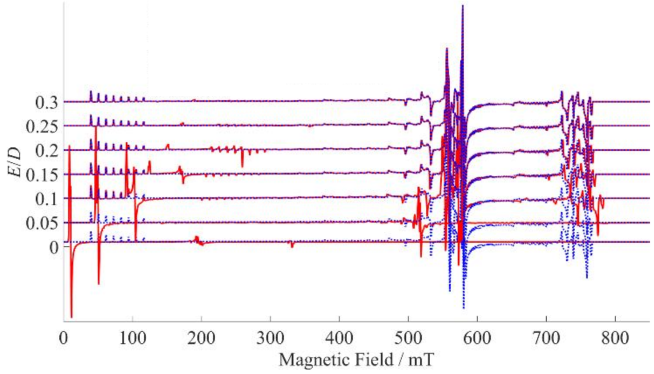

Figure S20. The simulated randomly-oriented ESR spectra of complex **2** (S = 3/2) as a function of E/D. E/D used were 0.01, 0.05, 0.1, 0.15, 0.2, 0.25 and 0.3. The spectra in blue and red are based on the fictitious spin-1/2 and true spin Hamiltonian approaches, respectively. Microwave frequency used: 9.625 GHz, the peak-to-peak linewidth: 1.0 mT; the magnetic tensors: $\mathbf{g}^{\mathrm{eff}}$ = [1.21, 0.92, 8.80], $A_Z^{\mathrm{eff}}(^{59}\mathrm{Co})$ = 1345.2 MHz, D = –9 cm^{-1} and $\mathbf{g}^{\mathrm{true}}$ and $A_Z^{\mathrm{true}}(^{59}\mathrm{Co})$ for the variable E/D's are shown in Table S7. The ZFS and **g**-tensors were assumed to be collinear. A set of rotation angles (Euler angles) of the $\mathbf{A}^{\mathrm{true}}$-tensor with respect to the ZFS tensor were α = –90, β = 90, and γ = 36 degrees. (Note: Because only A_y-value was considered, of which the principal axis was parallel to the D_z-axis, it seemed that there was no effect on the spectrum.) Any strain effect of the tensor to the linewidth was not included. The simulated spectra were obtained by using EasySpin (ver. 6.0.6) [8].

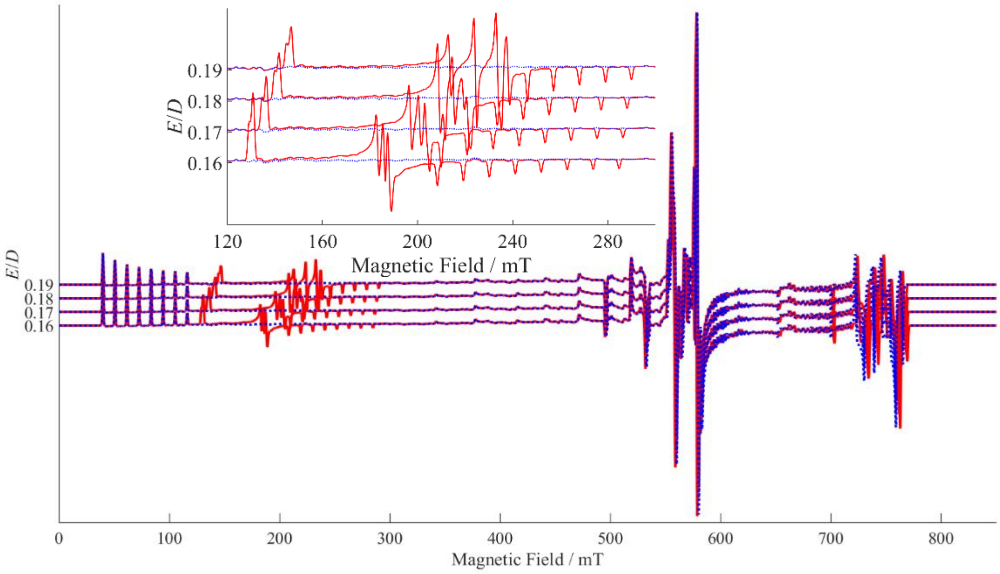

Figure S21. The simulated randomly-oriented ESR spectra of complex **2** (S = 3/2) as a function of E/D. The spectra in blue and red are based on the fictitious spin-1/2 and true spin Hamiltonian approaches, respectively. Microwave frequency used: 9.625 GHz, the peak-to-peak linewidth: 1.0 mT, the magnetic tensors: $\mathbf{g}^{\mathrm{eff}}$ = [1.21, 0.92, 8.80], $A_Z^{\mathrm{eff}}(^{59}\mathrm{Co})$ = 1345.2 MHz, D = –9 cm^{-1} and $\mathbf{g}^{\mathrm{true}}$, and $A_Z^{\mathrm{true}}(^{59}\mathrm{Co})$ for the variable E/D's are shown in Table S7. The ZFS and **g**-tensors were assumed to be collinear. A set of rotation angles (Euler angles) of the $\mathbf{A}^{\mathrm{true}}$-tensor with respect to the ZFS tensor were α = –90, β = 90, and γ = 36 degrees. (Note: Because only A_y-value was considered, of which the principal axis was parallel to the D_z-axis, it seemed that there was no effect on the spectrum.) Any strain effect of the tensor to the linewidth was not included. The simulated spectra were obtained by using EasySpin (ver. 6.0.6) [8]. Note that the resonance peaks marked by a single asterisk *, double asterisk ** and triple asterisk *** appear only for the true spin Hamiltonian approach. The inset shows the expanded spectra in the range of 120 mT to 300 mT.

Table S7. The principal values of the $\mathbf{g}^{true}$-tensor and A_{zz}-values used in the simulated spectra of complex **2** (Figures S22 and S23).

E/D	g_{XX}	g_{YY}	g_{ZZ}	E/D	g_{XX}	g_{YY}	g_{ZZ}
0.01	40.1	30.8	2.934	0.18	2.157	1.957	3.025
0.10	3.900	3.275	2.962	0.19	2.045	1.873	3.035
0.15	2.587	2.281	2.998	0.20	1.945	1.797	3.045
0.16	2.425	2.158	3.006	0.25	1.570	1.518	3.014
0.17	2.283	2.051	3.015	0.30	1.328	1.341	3.172

One of the possible sets of the magnetic parameters: $g_{XX} = 2.283$, $g_{YY} = 2.051$, $g_{ZZ} = 3.015$, $A_{ZZ}(^{59}Co) = 460.91$ MHz, $D = –9$ cm^{-1} and $E/D = +0.17$. The extra line observed at about 200 mT was simulated by using the parameters.

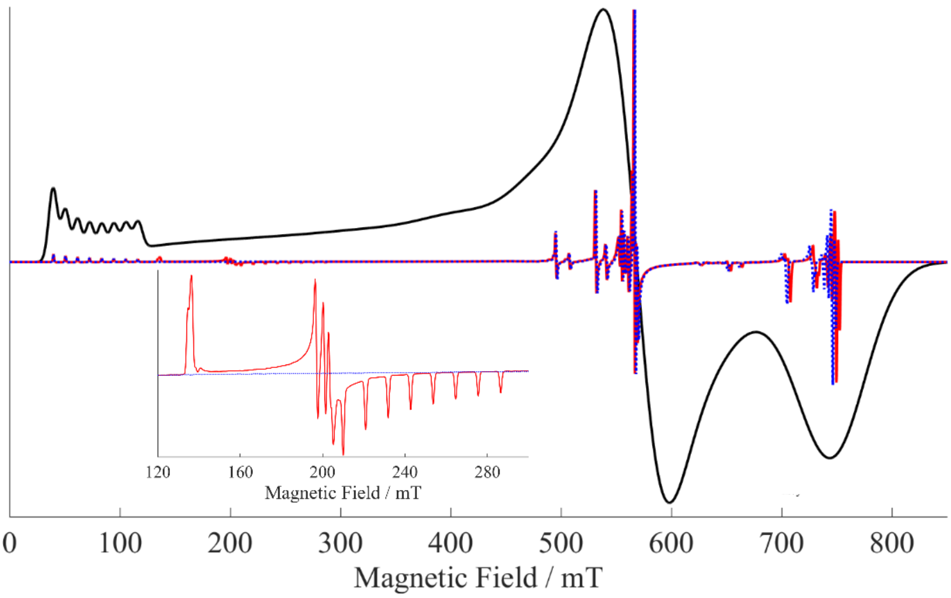

Figure S22. The simulated randomly-oriented X-band ESR spectra of complex **2** ($S = 3/2$). The spectrum in blue and red are based on the fictitious spin-1/2 and true spin Hamiltonian approaches, respectively. Microwave frequency used: 9.625 GHz, (the spectrum in black) the magnetic tensors: $g_1^{eff} = 8.8$, $g_2^{eff} = 1.21$, $g_3^{eff} = 0.92$, $A_1^{eff}(^{59}Co) = 1345$ MHz, the peak-to-peak linewidth: 8.0 mT, the strain parameters of the linewidth: [0, 900, 900] MHz; (the spectrum in blue and red) the peak-to-peak linewidth: 1.0 mT, the magnetic tensors: $g_{XX}^{eff} = 1.21$, $g_{YY}^{eff} = 0.92$, $g_{ZZ}^{eff} = 8.8$, $A_{ZZ}^{eff}(^{59}Co) = 1345$ MHz; $g_{XX}^{true} = 2.2832$, $g_{YY}^{true} = 2.0512$, $g_{ZZ}^{true} = 3.0152$, $A_{ZZ}^{true}(^{59}Co) = 460.91$ MHz, $D = –9 cm^{-1}$ and $E/D = +0.17$. Any strain effect of the tensor to the linewidth was not included in the spectra in blue and red. The simulated spectra were obtained by using EasySpin (ver. 6.0.6) [8]. Inset: The expanded spectra from 120 mT to 300 mT. The extralines attributed from the $|M_S = \pm 1/2\rangle$-dominant transitions can be reproduced by using the true spin Hamiltonian (red line) but not by using the fictitious spin-1/2 Hamiltonian (blue line).

In the simulated spectrum in Figure S22, $A_{ZZ}^{true} = 460.91$ MHz was calculated by using the A^{eff}/A^{true}-relationship for the principal z-axis because $\vec{A}_{ZZ}^{true}$ is parallel to $\vec{g}_{ZZ}^{true}$. The E/D dependence of A_{ZZ}^{true} obtained by putting the magnetic parameters to the derived perturbed energies crosses at a point with $E/D = 0.17$ and $A_{ZZ}^{true} = 460.91$ MHz (Figure S23).

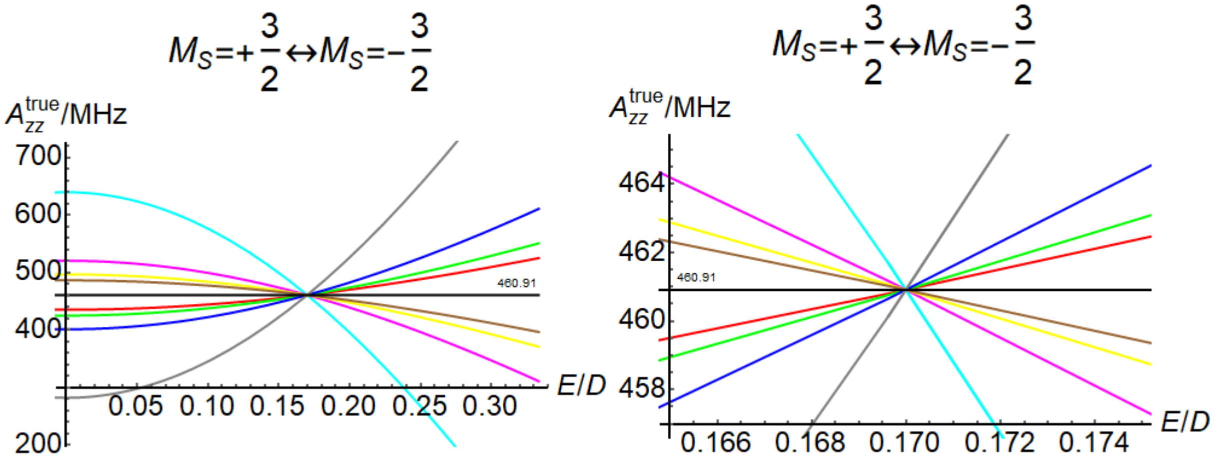

Figure S23. The E/D dependence of the A_Z^{true}-value which is the solution of eq (†) with the parameters used for the simulated spectrum in Figure S22. Red, green, blue, gray, cyan, magenta, yellow and brown curves correspond to the assignments to M_I = +7/2, +5/2, +3/2, +1/2, –1/2, –3/2, –5/2 and –7/2, respectively. Because all the energies and the parameters were accurate, all the curves cross at one point. The black line indicates the A_Z-value (= 460.91 MHz) calculated by using the relationship (eq (‡)) in the case of E/D = 0.17. The inset on the right shows the expanded figure in the range of 0.165 ≤ E/D ≤ 0.175.

Table S8. Comparison of the theoretical and experimental magnetic parameters for complex **2**. The theoretical values were taken from the result of DFT calculations.

	Theor.	Expl. (*Case 1*)	Expl. (*Case 2*)
g_x	2.0705	3.18412	2.0512
g_y	2.0469	2.9895	3.0152
g_z	2.1176	2.1085	2.2832
A_y/MHz	72.37	456.98	460.91
D/cm^{-1}	+9.9705	–9	–9
E/D	+0.1666	+0.14	+0.17

3. Complex 3

a) Perturbed energies in the case of non-collinearity between the ZFS and **g**-tensors for $S = 3/2$

Here, we derive the perturbed energies of the spin-Hamiltonian including the ZFS, electron Zeeman and hyperfine interactions in the case of non-collinearity between the ZFS and **g**-tensors, in which the D_z- and g_z-axes are parallel. The non-perturbed terms include the ZFS and electron Zeeman interaction tensors:

$$H_0 = \boldsymbol{S} \cdot \mathbf{D} \cdot \boldsymbol{S} + \beta \boldsymbol{S} \cdot \mathbf{g} \cdot \boldsymbol{B}$$

where the **g**-tensor is represented in the matrix form as

$$\mathbf{g} = \begin{pmatrix} g_{xx} & g_{xy} & 0 \\ g_{yx} & g_{yy} & 0 \\ 0 & 0 & g_{zz} \end{pmatrix}$$

Using 2nd-order parameters of ZFS (D and E) and with the magnetic field parallel to the z-axis of the ZFS tensor, the non-perturbed Hamiltonian is described as

$$H_0 = D\left[S_z^2 - \frac{1}{3}S(S+1)\right] + E\left(S_x^2 - S_y^2\right) + g_{zz}\beta S_z B_z$$

which is the same formula as in the collinear case. Therefore, the zeroth-order energies are the same as the previous ones. Since the ZFS and **A**-tensors are collinear, the first- and the second-order perturbed energies are already derived.

b) ESR analysis of complex **3**

Figure S24 shows the simulated ESR spectra of complex **3** as a function of E/D in the case of non-collinearity between the ZFS and **g**-tensors. The principal values of the **g**-tensor were optimized for each value of E/D. Since the **A**-tensor was assumed to be collinear with the ZFS tensor, A_Z^{true} in the right-most column in Table S9 was calculated by using the A_Z^{eff}/A_Z^{true} relationship.

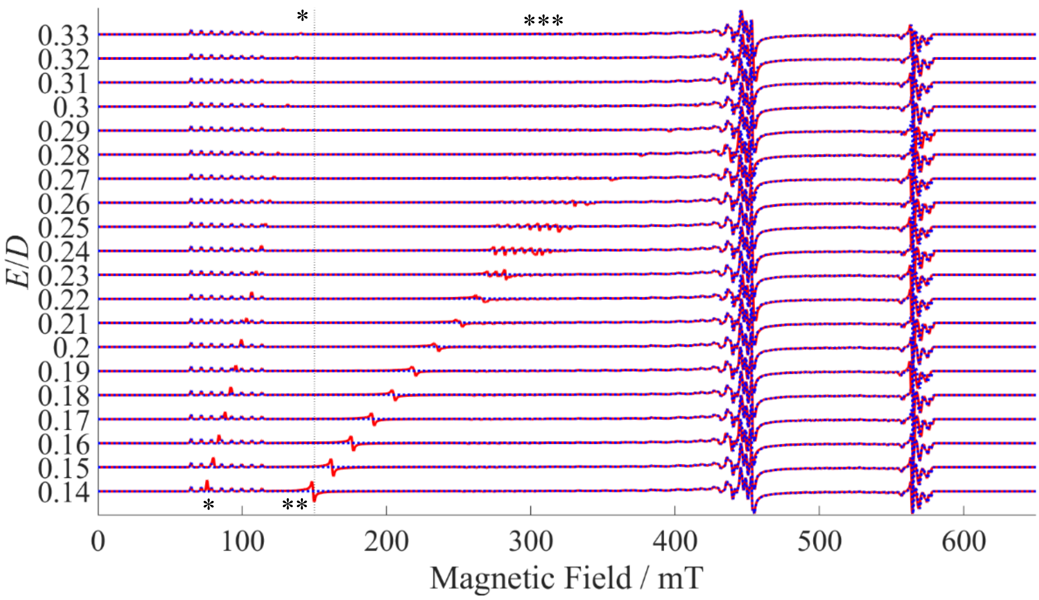

Figure S24. The simulated randomly-oriented X-band ESR spectra of complex **3** (S = 3/2). The spectra in blue and red are based on the fictitious spin-1/2 and true spin Hamiltonian approaches, respectively. Microwave frequency used: 9.482 GHz, the peak-to-peak linewidth: 1.0 mT, the magnetic tensors: g^{eff} = [1.51, 1.19, 7.6], $A_z^{eff}(^{59}Co)$ = 745.5 MHz, $D = -10$ cm^{-1} and g^{true} and $A_z^{true}(^{59}Co)$ for the variable E/D's are in Table S9. The **D**- and **A**-tensors were assumed to be collinear. A set of rotation angles (Euler angles) of the $\mathbf{g}^{true}$-tensor with respect to the **D**-tensor were $\alpha = -113$, $\beta = 180$, and $\gamma = 0$ degrees. Any strain effect of the tensor to the linewidth was not included. The simulated spectra were obtained by using EasySpin (ver. 6.0.6) [8]. The resonance peaks marked by an asterisk *, double asterisk ** and triple asterisk *** appear only for the true spin Hamiltonian approach. The broken line at 150 mT indicates the resonance position of the extra line appearing in the experimental spectrum.

Table S9. The principal values of the $\mathbf{g}^{true}$-tensor and A_z^{true}-values used in the simulated spectra of complex **3** in Figure S24.

$E/D = \lambda$	g_X^{true}	g_Y^{true}	g_Z^{true}	A_Z^{true}/MHz
0.14	3.86	2.82	2.582	253.2
0.15	3.62	2.64	2.589	253.9
0.16	3.42	2.48	2.596	254.7
0.17	3.23	2.34	2.604	255.4
0.18	3.07	2.22	2.612	256.2
0.19	2.93	2.12	2.621	257.1
0.20	2.80	2.02	2.630	258.0

0.21	2.68	1.93	2.640	258.9
0.22	2.57	1.86	2.649	259.8
0.23	2.48	1.79	2.659	260.8
0.24	2.39	1.72	2.670	261.9
0.25	2.31	1.67	2.681	262.9
0.26	2.24	1.61	2.692	264.0
0.27	2.17	1.57	2.703	265.1
0.28	2.11	1.52	2.714	266.3
0.29	2.14	1.48	2.727	267.5
0.30	1.98	1.45	2.739	268.7
0.31	1.93	1.42	2.752	269.9
0.32	1.87	1.39	2.764	271.1
0.33	1.82	1.37	2.777	272.4

We consider two cases, E/D = +0.14 (*Case 1*) and +0.33 (*Case 2*), where the extraline was simulated, appearing at about 150 mT (indicated by the broken line in Figure S24).

Case 1

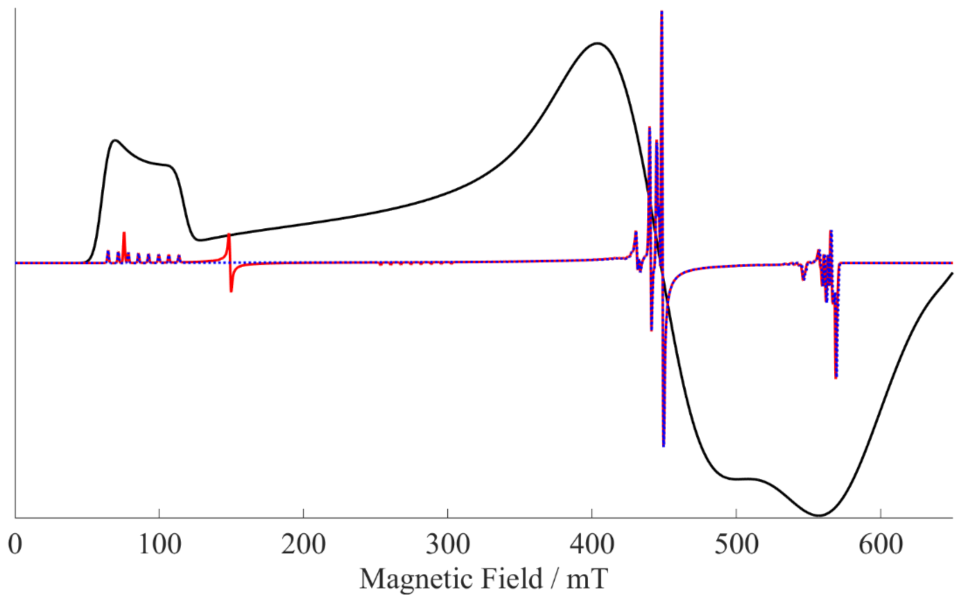

Figure S25. The simulated randomly-oriented X-band ESR spectra of complex **3** (S = 3/2). The spectra in black and blue are the same as given in Figure 9 in the manuscript. The spectrum in red is based on the true spin Hamiltonian approach. Microwave frequency used: 9.4715 GHz, (the spectrum in red) magnetic tensors: g_X^{true} = 3.86, g_Y^{true} = 2.82, g_Z^{true} = 2.5818, $A_Z^{true}(^{59}Co)$ = 253.2 MHz, D = –10 cm^{-1} and E/D = +0.14, peak-to-peak linewidth: 1.0 mT. The ZFS and **A**-tensors were assumed to be collinear. A set of rotation angles (Euler angles) of the $\mathbf{g}^{\mathrm{true}}$-tensor with respect to the ZFS tensor were $\alpha = -113$, $\beta = 180$, and $\gamma = 0$ degrees. Any strain effect of the tensor to the linewidth was not included in the spectrum in red. The simulated spectra were obtained by using EasySpin (ver. 6.0.6) [8]. The resonance peaks marked by an asterisk *, double asterisk ** and triple asterisk *** appear only for the true spin Hamiltonian approach.

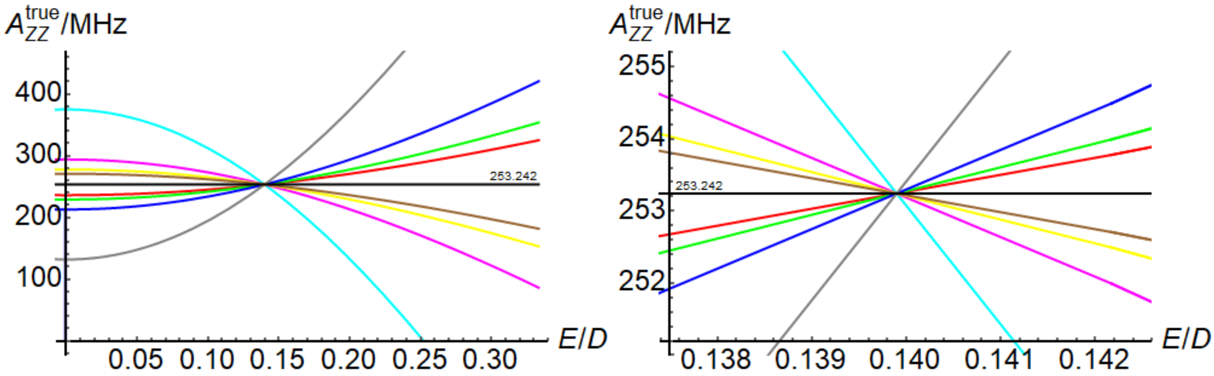

Figure S26. The E/D dependence of the A_Z^{true}-value which is the solution of eq (†). Red, green, blue, gray, cyan, magenta, yellow and brown curves correspond to the assignments to $M_I = +7/2, +5/2, +3/2, +1/2, -1/2, -3/2, -5/2$ and $-7/2$, respectively. Because all the energies and the parameters were accurate, all the curves cross at one point. The black line indicates the A_Z-value (= 253.242 MHz) calculated by using of the relationship (eq (‡)) in the case of $E/D = 0.33$. The right figure shows the expanded one in the range of $0.138 \leq E/D \leq 0.142$.

Case 2

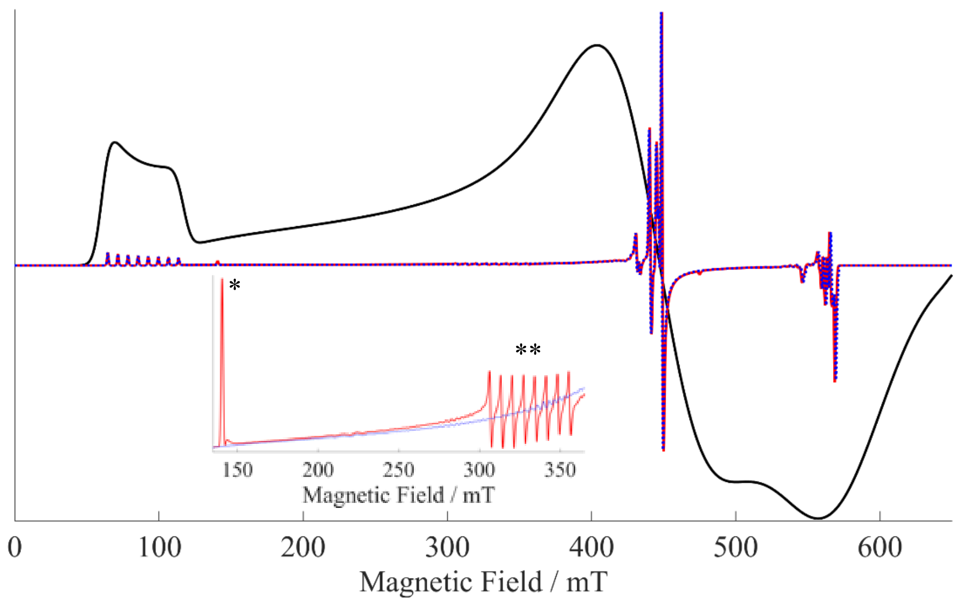

Figure S27. The simulated randomly-oriented X-band ESR spectra of complex **3** ($S = 3/2$). The spectra in black and blue are the same as given in Figure 9 in the manuscript. The spectrum in red is based on true spin Hamiltonian approach. Microwave frequency used: 9.4715 GHz, (the spectrum in red) the magnetic tensors: $g_X^{\mathrm{true}} = 1.82$, $g_Y^{\mathrm{true}} = 1.37$, $g_Z^{\mathrm{true}} = 2.7774$, $A_Z^{\mathrm{true}}(^{59}\mathrm{Co}) = 272.4$ MHz, $D = -10\ \mathrm{cm}^{-1}$ and $E/D = +0.33$, the peak-to-peak linewidth: 1.0 mT. The **D**-, and **A**-tensors were assumed to be collinear. A set of rotation angles (Euler angles) of the $\mathbf{g}^{\mathrm{true}}$-tensor with respect to the **D**-tensor were $\alpha = -113$, $\beta = 180$, and $\gamma = 0$ degrees. Any strain effect of the tensor to the linewidth was not included in the spectrum in red. The simulated spectra were obtained by using EasySpin (ver. 6.0.6) [8] The inset shows the expanded spectrum in the range from 135 mT to 365 mT. The resonance peaks marked by an asterisk * and double asterisk ** appear only for the true spin Hamiltonian approach.

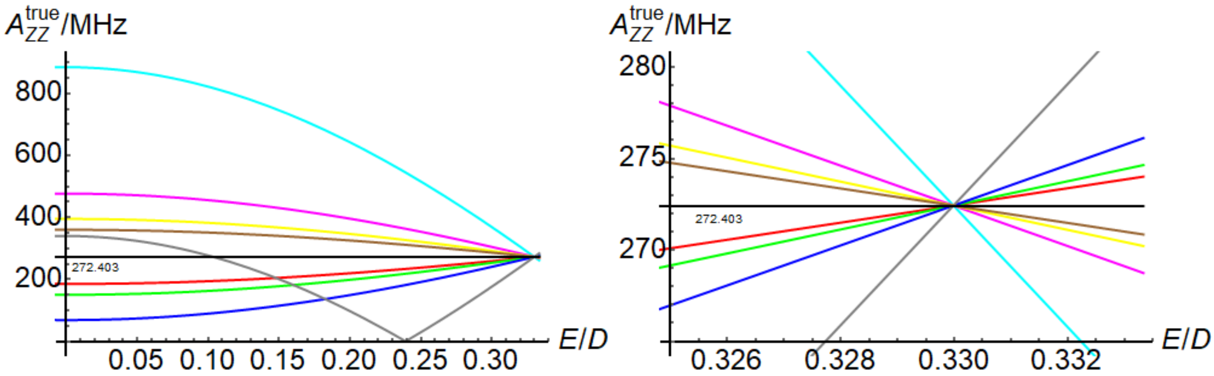

Figure S28. The E/D dependence of the A_Z^{true}-value which is the solution of eq (†). Red, green, blue, gray, cyan, magenta, yellow and brown curves correspond to the assignments to M_I = +7/2, +5/2, +3/2, +1/2, –1/2, –3/2, –5/2 and –7/2, respectively. Because all the energies and the parameters were accurate, all the curves cross at one point. The black line indicates the A_Z-value (= 272.403 MHz) calculated by using the relationship (eq (‡)) in the case of E/D = 0.33. The figure on the right shows the expanded one in the range of $0.325 \leq E/D \leq 1/3$.

4. Discrepancy in the experimental g^{eff}-tensors between the conventional X-band and high-field ESR spectroscopies for complex 4.

We have examined three possible cases below to evaluate the discrepancy in the experimental $\mathbf{g}^{eff}$-tensors between the conventional X-band and high-field ESR spectroscopies for complex **4**.

Case 1: Assuming that the experimental $\mathbf{g}^{eff}$-tensor from the X-band spectroscopy is correct and $E/D = 0.26$. The g^{eff}/g^{true} relationships give g^{true} = [2.46, 2.25, 2.54] (g_{ave} = 2.417) or [1.75, 3.16, 2.54] (g_{ave} = 2.483). The former choice gives the X-band simulated spectrum (Fig. 15) based on the true spin Hamiltonian approach. The g_z-value of 2.51 obtained from the high-field spectroscopy is close to 2.54, while both the g_x- and g_y-values from the high-field spectroscopy disagree with those from the X-band spectroscopy with the help of the relationships.

Case 2: Assuming that the experimental $\mathbf{g}^{true}$-tensor from the high-field spectroscopy is correct and $E/D = 0.26$. The g^{eff}/g^{true} relationships give g^{eff} = [1.76, 1.30, 7.09] or [1.66, 1.37, 7.09], both of which don't seem reasonable.

Case 3: Assuming that both the $\mathbf{g}^{eff}$- and $\mathbf{g}^{true}$-tensor (from the high-field spectroscopy) are correct. The three g^{eff}/g^{true} relationships give greatly different values for the ratio of E/D. We have also invoked the relationships for the possible occurrence of the non-collinearity between the **D**- and **g**-tensors, and there is no such possibility enabling us to reproduce the X-band spectrum.

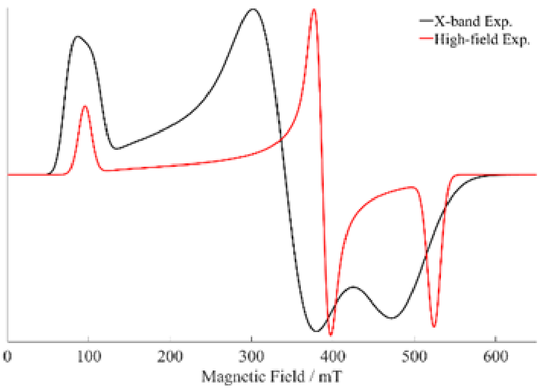

Fig. S29 (black) The reproduced randomly-oriented X-band ESR spectrum of complex **4** (S = 3/2) shown in the literature [10] and (red) the simulated ESR spectrum of complex **4** (S = 3/2) calculated by using the magnetic parameters from the high-field experiment shown in the literature [11]. Microwave frequency used: 9.474 GHz; (the spectrum in black) the magnetic parameters: g_1^{eff} = 7.2, g_2^{eff} = 1.97, g_3^{eff} = 1.4, $A_1^{eff}(^{59}Co)$ = 560.5 MHz, the peak-to-peak linewidth: 16.0 mT, the strain parameters of the linewidth: [0, 1850, 1600] MHz; (the spectrum in red) the magnetic parameters: $\mathbf{g}$ = [2.20, 2.08, 2.51], $D = -38.7$ cm^{-1} and $E = -10.0$ cm^{-1} (E/D = 0.26), the peak-to-peak linewidth: 16 mT. The **D**-, **g**- and **A**-tensors were assumed to be collinear. Any strain effect of the tensor to the linewidth was not included in the spectrum in red. The simulated spectra were obtained by using EasySpin (ver. 6.0.6) [8].

Quantum Chemical Calculations

1. Cartesian coordinates of complex 1 - 3 optimized at the UB3LYP/6-31G* method with PCM (CH_2Cl_2 solvent)

Cartesian coordinates of complex **1**

Co	1.123826	0.490574	-0.010374
S	2.965976	1.884343	-0.126014
O	4.883328	-2.635118	0.671629
O	6.078034	-1.576772	-0.939381
N	-1.734345	1.426148	-0.253925
N	-0.515978	2.014018	-0.049327
N	-1.211109	-0.907482	-1.082154
N	0.045033	-0.552557	-1.484883
N	-1.283233	-0.412126	1.433060
N	0.069572	-0.319422	1.609306
N	2.603479	-1.146266	0.055054
C	-2.664815	2.362564	-0.605923
C	-2.022244	3.593753	-0.613008
C	-0.687991	3.334682	-0.252148
C	-1.707009	-1.874947	-1.909060
C	-0.740435	-2.130543	-2.877237
C	0.336661	-1.281374	-2.580825
C	-1.855935	-0.977152	2.536527
C	-0.833582	-1.268268	3.434377
C	0.351638	-0.830267	2.824231
C	-4.084394	2.098417	-0.911242
C	-4.487495	1.071818	-1.780970
C	-5.836542	0.884945	-2.080938
C	-6.805366	1.724754	-1.526458
C	-6.415726	2.756457	-0.669091
C	-5.067262	2.941931	-0.364159
C	0.348681	4.365007	-0.039747
C	1.108034	4.401404	1.139602
C	2.007354	5.442072	1.372329
C	2.161905	6.462415	0.431329
C	1.410477	6.436636	-0.746359
C	0.507558	5.399087	-0.977467
C	-3.029035	-2.518683	-1.789589
C	-3.792264	-2.725215	-2.952355
C	-5.025670	-3.373185	-2.887751
C	-5.516375	-3.826204	-1.660686
C	-4.762090	-3.633312	-0.501142
C	-3.526325	-2.990031	-0.564194
C	1.588593	-1.131930	-3.344723
C	2.223203	-2.265023	-3.881820
C	3.386990	-2.133898	-4.639312
C	3.932529	-0.869007	-4.874792
C	3.304871	0.264038	-4.351044
C	2.139952	0.135863	-3.593668
C	-3.301341	-1.181250	2.750563
C	-3.741659	-2.382762	3.332626
C	-5.094524	-2.588067	3.604133
C	-6.028957	-1.594866	3.300751
C	-5.600250	-0.393390	2.731325
C	-4.248167	-0.184165	2.462513
C	1.704526	-0.858377	3.414792
C	2.145796	-2.018360	4.075288

C	3.397735	-2.054280	4.688671
C	4.226586	-0.930217	4.657258
C	3.794931	0.229871	4.009482
C	2.542506	0.269187	3.394380
C	4.308976	0.635946	0.062850
C	3.906179	-0.741501	-0.495505
C	4.988614	-1.770826	-0.178861
C	7.220943	-2.444679	-0.701188
C	8.092052	-1.911712	0.424726
B	-1.911540	-0.080795	0.043072
H	2.746176	-1.470750	1.013741
H	2.257855	-1.959268	-0.454391
H	-2.463532	4.553797	-0.837701
H	-0.814416	-2.832920	-3.694409
H	-0.943833	-1.666588	4.432150
H	-3.742481	0.426698	-2.233829
H	-6.128470	0.081625	-2.751631
H	-7.855748	1.577431	-1.762300
H	-7.161472	3.415726	-0.233152
H	-4.768013	3.737971	0.311764
H	0.977833	3.618268	1.879252
H	2.585214	5.457116	2.292555
H	2.861923	7.273174	0.614712
H	1.526182	7.225092	-1.485296
H	-0.076175	5.381429	-1.893926
H	-3.419037	-2.363046	-3.906063
H	-5.604253	-3.520074	-3.795658
H	-6.478673	-4.328020	-1.609104
H	-5.132744	-3.984372	0.457717
H	-2.940270	-2.863999	0.339331
H	1.807148	-3.250807	-3.692551
H	3.869581	-3.019979	-5.042477
H	4.838978	-0.767047	-5.464985
H	3.718493	1.251608	-4.534885
H	1.647651	1.019401	-3.201577
H	-3.018249	-3.160378	3.561375
H	-5.417563	-3.524486	4.050693
H	-7.083095	-1.754503	3.509909
H	-6.318475	0.387643	2.498260
H	-3.923935	0.760662	2.039578
H	1.505032	-2.895538	4.096367
H	3.726314	-2.961227	5.188507
H	5.201016	-0.957041	5.136958
H	4.428231	1.112591	3.991516
H	2.208084	1.179649	2.906853
H	5.204965	0.982476	-0.458052
H	4.558700	0.536400	1.125926
H	3.806806	-0.658813	-1.579921
H	6.854921	-3.450752	-0.485515
H	7.753952	-2.450066	-1.653998
H	8.422373	-0.889965	0.211942
H	8.979646	-2.545082	0.532093
H	7.549045	-1.917900	1.374306
H	-3.062750	-0.362336	0.059984

Cartesian coordinates of complex **2**

Co	-0.043558	0.582647	-0.035243

S	-1.494225	2.394014	-0.247271
O	-4.461155	-1.533024	-0.583094
O	-5.433943	-0.041637	0.822919
N	2.953527	0.817266	-0.111857
N	1.859018	1.644042	-0.139001
N	2.004779	-1.003592	1.352062
N	0.773144	-0.449675	1.583813
N	1.945580	-1.231336	-1.192731
N	0.708344	-0.712083	-1.467510
N	-1.919977	-0.611601	0.134957
C	4.098858	1.541185	-0.208362
C	3.732949	2.876911	-0.300583
C	2.327259	2.897147	-0.253066
C	2.418839	-1.682763	2.452641
C	1.428697	-1.556524	3.419929
C	0.417727	-0.776004	2.836417
C	2.292723	-2.125173	-2.154110
C	1.252589	-2.174832	-3.075263
C	0.280797	-1.275094	-2.608161
C	5.470676	0.938464	-0.208871
C	1.438343	4.102869	-0.309053
C	3.727344	-2.406488	2.540938
C	-0.876128	-0.323032	3.443370
C	3.586910	-2.879232	-2.159523
C	-1.042377	-0.927002	-3.221622
C	-3.091390	1.491031	-0.403655
C	-3.107314	0.200728	0.439550
C	-4.394294	-0.575692	0.166444
C	-6.745713	-0.630324	0.592403
C	-7.408269	-0.029744	-0.636472
B	2.759477	-0.708439	0.024319
H	-2.105637	-1.125572	-0.727999
H	-1.801119	-1.332312	0.846321
H	4.395854	3.726616	-0.389538
H	1.443348	-1.969096	4.419105
H	1.209802	-2.779150	-3.970644
H	-3.911600	2.137476	-0.080793
H	-3.267684	1.236254	-1.455685
H	-3.078156	0.478745	1.495772
H	-6.629786	-1.712330	0.501087
H	-7.302599	-0.401260	1.502947
H	-7.483196	1.058593	-0.546292
H	-8.419921	-0.436857	-0.742414
H	-6.844482	-0.272052	-1.542064
H	3.819852	-1.257568	0.048489
H	-1.667406	-1.076189	3.332612
H	-0.752464	-0.145712	4.516099
H	-1.222590	0.607257	2.983954
H	3.823576	-3.171732	1.763141
H	4.578325	-1.724522	2.435684
H	3.806557	-2.899029	3.513745
H	3.609982	-3.557980	-3.016222
H	4.449712	-2.208224	-2.234480
H	3.714716	-3.475071	-1.249343
H	-1.387508	0.055166	-2.884691
H	-0.962598	-0.899950	-4.312942
H	-1.815589	-1.667424	-2.977484
H	0.855179	4.220019	0.611264

H	2.043260	5.004506	-0.443413
H	0.722829	4.041290	-1.135292
H	5.671190	0.375168	0.709379
H	5.617107	0.252105	-1.050540
H	6.217854	1.732916	-0.287909

Cartesian coordinates of complex **3**

Co	0.651550	0.036957	-0.204214
O	-0.973842	1.160152	-0.174056
N	3.281109	-1.177174	-0.999643
N	1.989548	-1.095052	-1.441692
N	3.420554	1.145133	-0.027405
N	2.140823	1.567047	-0.294169
N	2.930794	-0.816287	1.493073
N	1.570666	-0.676612	1.522521
N	-0.859668	-1.522602	-0.262936
C	-2.143032	0.526189	-0.129668
C	-2.167185	-0.886939	-0.167116
C	-3.344154	-1.630822	-0.119864
C	-4.582359	-0.994213	-0.029880
C	-4.562650	0.413788	0.007223
C	-3.406366	1.197824	-0.037897
C	-3.486590	2.737523	-0.005710
C	-2.938636	3.304385	-1.337666
C	-2.665500	3.285909	1.186389
C	-4.929312	3.257910	0.155987
C	-5.920738	-1.754142	0.023161
C	-6.796278	-1.354804	-1.188745
C	-5.722037	-3.281239	-0.015840
C	-6.673588	-1.403080	1.328779
C	4.025365	-1.910996	-1.868245
C	3.185109	-2.312909	-2.898846
C	1.921572	-1.775677	-2.595030
C	4.275622	2.199385	-0.066794
C	3.529887	3.330787	-0.369894
C	2.201444	2.891559	-0.506336
C	3.373577	-1.289627	2.685275
C	2.263684	-1.458102	3.506113
C	1.155319	-1.058341	2.741232
C	5.486712	-2.185101	-1.684034
C	0.649097	-1.894329	-3.380077
C	5.748185	2.087392	0.186056
C	0.986637	3.695052	-0.851021
C	4.816272	-1.553841	2.989888
C	-0.291411	-1.026800	3.132721
B	3.717217	-0.353373	0.239755
H	-0.786411	-2.071501	-1.119873
H	-0.715316	-2.189252	0.495993
H	-3.268374	-2.713941	-0.153675
H	-5.516661	0.923480	0.076249
H	-3.574440	2.995095	-2.176606
H	-2.926763	4.401917	-1.312170
H	-1.924403	2.950382	-1.528690
H	-3.092950	2.941480	2.136543
H	-1.628124	2.952641	1.134245
H	-2.685189	4.383631	1.189151
H	-5.390571	2.910457	1.087941

H	-5.574847	2.958982	-0.678112
H	-4.915786	4.353914	0.183228
H	-7.760907	-1.877228	-1.156566
H	-6.300208	-1.615526	-2.131152
H	-6.999878	-0.278804	-1.204507
H	-5.221644	-3.602948	-0.936553
H	-6.695534	-3.783388	0.024029
H	-5.130543	-3.636719	0.835955
H	-6.084395	-1.688726	2.208161
H	-6.884105	-0.330717	1.401161
H	-7.632398	-1.934853	1.374361
H	3.454669	-2.909779	-3.759150
H	3.900749	4.341064	-0.475481
H	2.262849	-1.815769	4.526447
H	6.080133	-1.264036	-1.700946
H	5.842089	-2.831196	-2.491292
H	5.690917	-2.686859	-0.731835
H	0.115295	-2.828583	-3.159557
H	0.859938	-1.896414	-4.454084
H	-0.025500	-1.057518	-3.173534
H	5.961953	1.690158	1.184482
H	6.207901	3.076505	0.109595
H	6.239081	1.428426	-0.538797
H	1.010333	4.012534	-1.901674
H	0.935181	4.604504	-0.241426
H	0.083257	3.106105	-0.686154
H	5.252355	-2.280005	2.295159
H	4.908660	-1.954867	4.002704
H	5.419501	-0.641264	2.929418
H	-0.878192	-0.427690	2.431256
H	-0.411643	-0.594395	4.131603
H	-0.723190	-2.035345	3.166273
H	4.887251	-0.507771	0.426123

2. Relationship between molecular structures and principal axes of the A, g, and D tensors.

The relationship between the molecular structures and principal axes of the theoretical magnetic tensors for complex **1**–**3** calculated in this study are summarized in Figure S30. The calculations were carried out by two methods, i.e., DFT and CASSCF.

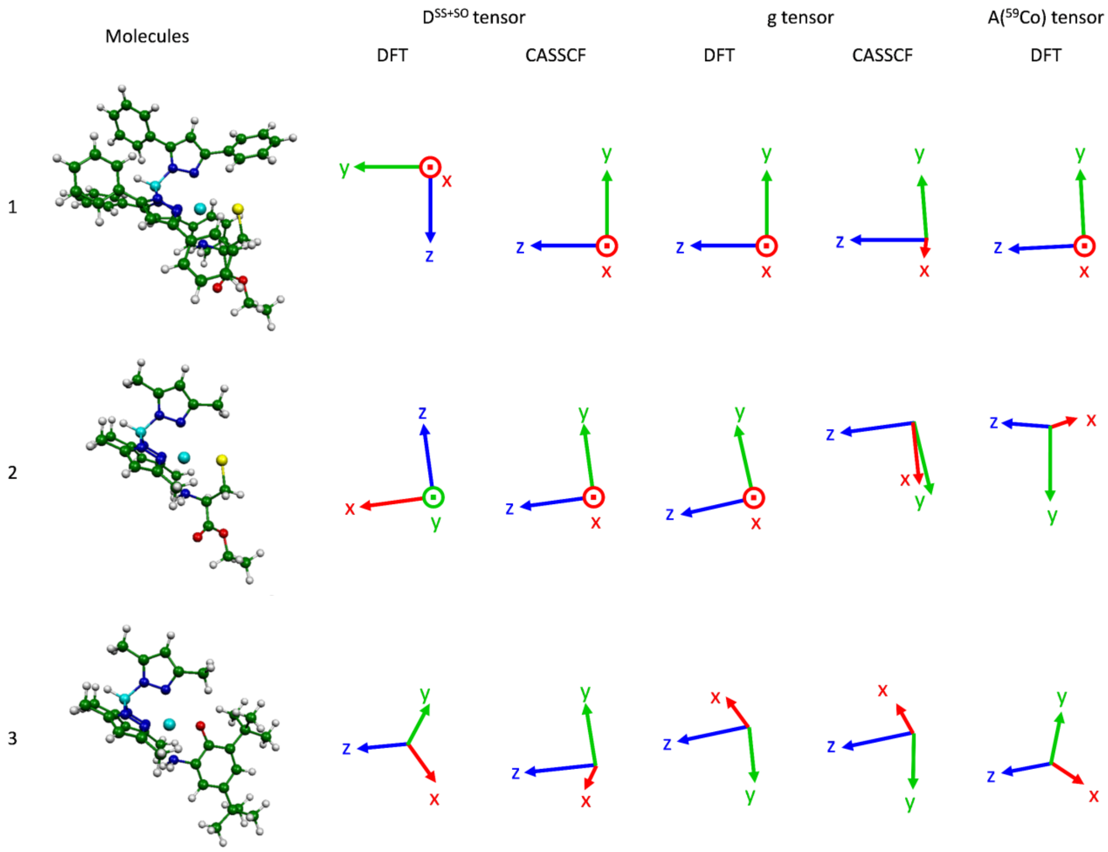

Figure S30. Relationship between the molecular structures and principal axes of the theoretical **D**-, **g**-, and **A**-tensors for complex **1**–**3**. The principal axes of the magnetic tensors obtained by the DFT and CASSCF method are depicted.

3. CASSCF calculations of complex 1–3.

The cobalt complexes **1**–**3** have trigonal bipyramidal structures at the UB3LYP/6-31G* optimized geometries. In the trigonal bipyramidal coordinations, the valence d orbitals split into three groups: d_{xz} and d_{yz} (e″ symmetry in D_{3h} point group), d_{xy} and d_{x2-y2} (e′), and d_{z2} (a_1′) orbitals. The e″ orbitals (d_{xz} and d_{yz}) are the most stable among the three groups, and the d_{z2} orbital is the most unstable. The Co^{II} complexes under study have seven valence d-electrons and therefore the main configuration of the spin-quartet ground state is given by $(d_{xz})^2(d_{yz})^2(d_{xy})^1(d_{x2-y2})^1(d_{z2})^1$. The energy gap between the e″ and e′ orbitals is generally small and thus excited states arising from the electron excitation from the e″ to e′ orbitals have small excitation energies. We note that such a quasi-degeneracy of the electronic states prevents us to predict the **g**- and $\mathbf{D}^{SO}$-tensors by means of DFT methods for those magnetic tensors. Those magnetic tensors can be calculated by using the second-order perturbation theory starting from a non-relativistic wavefunction as the unperturbed wavefunction. Thus, we carried out CASSCF calculations of the electronic ground and excited states of complex **1**–**3**. In the present study, we used the Sapporo-DZP basis set only for the cobalt atom, and the 3-21G basis set for the other nuclei, due to the limitations of our computational resources.

The CASSCF active space of complex **1**–**3** is illustrated in Figure S31. The active space consists of the valence 3d orbitals/electrons of the cobalt atom. We carried out the state-specific (SS) CASSCF for the electronic ground state. Electronic excited states calculations were carried out at the CAS-CI level by using the SS-CASSCF canonical orbitals for the ground state. The CASSCF excitation energies, major electronic configurations having the CI coefficient c greater than 0.3 ($c > 0.3$), and contributions to the principal values of the **g**- and $\mathbf{D}^{SO}$-tensors are summarized in Tables S10–S12. From the Tables S10–S12, the wavefunction of the 1 ^{4}A ground state is well described by the single configuration, justifying the usage of DFT for the ground state geometry optimizations and the **A**-tensor calculations. However, complex **1**–**3** have many low-lying excited states, and the first excited quartet state (2 ^{4}A) is calculated to be ca. 2,000 cm^{-1} above the ground state. From the CASSCF wavefunction analyses, the 2 ^{4}A state contributes significantly to the g_z and D^{SO}_z principal values. The other low-lying states also have non-negligible contributions to the **g**- and $\mathbf{D}^{SO}$-tensors. In the $\mathbf{D}^{SO}$-tensor, the spin-doublet and quartet states have positive and negative contributions, respectively, to the principal value of the $\mathbf{D}^{SO}$-tensor. This is because the absolute sign of the coefficients depending on the difference in spin quantum numbers between the ground and excited states appearing in the sum-over-states formula is different for $\Delta S = 0$ and ± 1 [12]. Note that only the electronic excited states having the same spin quantum number as the electronic ground state can contribute to the **g**-tensor, and therefore the spin-doublet excited states have no contributions to the **g**-tensor of complex **1**–**3**.

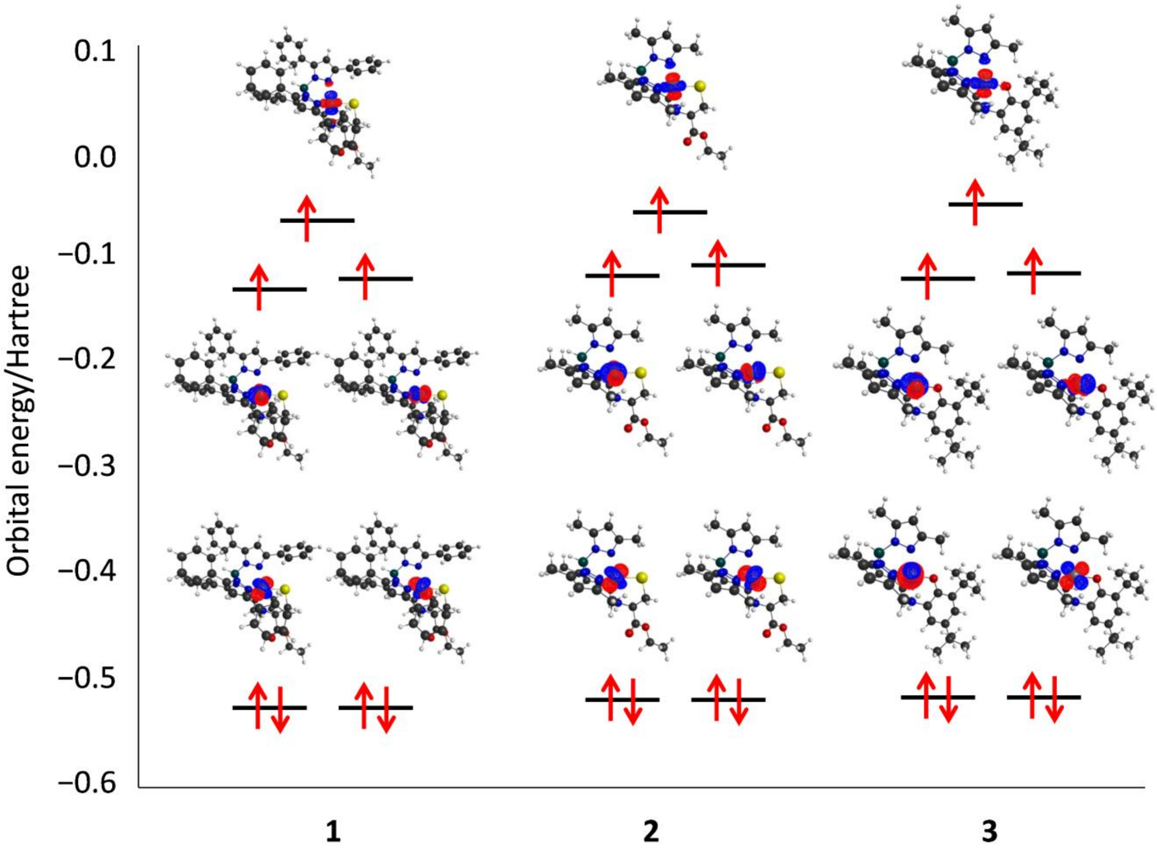

Figure S31. The CASSCF canonical orbitals of the electronic ground state of complex **1**–**3**. Red arrows specify the main electronic configuration of the ground state.

Table S10. The electronic states of complex **1** calculated at the CASSCF(7e,5o)/Sapporo-DZP(Co) and 3-21G(other nuclei) level.

State	Main configurations[a]	ΔE/cm^{-1}	$g_{ii}{}^{b}$			$D^{SO}{}_{ii}{}^{c}$/cm^{-1}		
			$i = x$	$i = y$	$i = z$	$i = x$	$i = y$	$i = z$
1 ^{4}A	0.99 (22uuu)	0						
2 ^{4}A	−0.62 (u22uu) +0.60 (2u2uu) −0.42 (uu22u)	1 862	0.0014	0.0019	0.4745	−0.1523	−0.3854	−46.8702
3 ^{4}A	−0.62 (u22uu) −0.59 (2u2uu) −0.38 (2uu2u)	2 906	0.1025	0.1129	0.0010	−5.5054	−15.9127	−0.1709
4 ^{4}A	−0.73 (2uu2u) −0.44 (uu22u) −0.42 (2uuu2)	6 090	0.0302	0.0391	0.0882	−4.9773	−2.2337	−8.7662
5 ^{4}A	−0.69 (u2u2u) +0.46 (u2uu2)	6 302	0.1187	0.0426	0.0354	−14.5984	−1.5917	−3.8114
6 ^{4}A	−0.82 (uu2u2) +0.49 (u2uu2)	10 477	0.0227	0.0006	0.0003	−2.3709	−0.0107	−0.0461
7 ^{4}A	0.90 (uuu22)	14 193	0.0001	0.0000	0.0002	−0.0064	−0.0001	−0.0221
8 ^{4}A	−0.60 (uu22u) +0.53 (u2u2u) +0.32 (u22uu)	24 160	0.0004	0.0000	0.0001	−0.0505	−0.0006	−0.0165
9 ^{4}A	−0.43 (u2uu2) +0.41 (2uu2u) −0.40 (uu2u2) −0.38 (2uuu2) −0.33 (uu22u)	24 667	0.0016	0.0000	0.0000	−0.1626	−0.0058	−0.0032
10 ^{4}A	0.63 (2uuu2) −0.51 (u2uu2) +0.34 (uuu22)	25 702	0.0000	0.0000	0.0012	−0.0004	−0.0002	−0.1424
1 ^{2}A	0.90 (222u0)	15 806				0.0709	0.0166	1.3954
2 ^{2}A	0.83 (2220u) + 0.34 (u22du)	18 482				0.4716	3.8617	0.1008
3 ^{2}A	−0.76 (22u20) −0.39 (2u220)	18 905				0.2849	0.0031	0.0044
4 ^{2}A	−0.86 (22udu)	20 815				0.0005	0.0004	0.2521

5 ^{2}A	−0.55 (22uud) +0.44 (2u2ud) −0.38 (u22ud)	21 295	0.0000	0.0019	1.7546
6 ^{2}A	0.53 (u22du) −0.31 (u2202)	22 369	0.6478	0.2101	0.9411
7 ^{2}A	−0.55 (2u2du) +0.32 (u22ud) −0.31 (ud22u)	22 444	1.2149	0.5107	0.4749
8 ^{2}A	−0.50 (2uu2d) −0.50 (u22ud) +0.34 (u2u2d) −0.33 (2u2ud)	22 713	0.0067	0.4881	0.0001
9 ^{2}A	0.62 (2ud2u) +0.44 (2202u) −0.37 (u2d2u)	24 513	0.4439	4.4679	0.0019
10 ^{2}A	−0.35 (u22ud) −0.35 (2u220) +0.31 (u2220) −0.31 (u22du)	26 118	0.0617	0.1222	0.0002
11 ^{2}A	−0.66 (22uud) −0.42 (2u2ud) +0.42 (uu22d)	26 660	0.0001	0.0002	0.0418
12 ^{2}A	−0.48 (uu22d) −0.35 (u2u2d) −0.31 (2uu2d) −0.30 (u2220)	27 759	0.0003	0.0023	0.0008
13 ^{2}A	0.52 (2202u) −0.33 (2022u)	28 941	0.5128	0.0567	0.0105
14 ^{2}A	−0.55 (u2d2u) −0.33 (2ud2u) +0.32 (u2202)	29 642	0.0024	0.0001	0.3352
15 ^{2}A	−0.61 (22u02) −0.34 (u2ud2)	30 641	0.3129	0.1998	0.0212
16 ^{2}A	0.46 (u2202) +0.41 (2uud2) +0.34 (2uu2d)	31 374	0.0004	0.0404	0.0412
17 ^{2}A	0.76 (uu2d2)	31 864	0.0016	0.0022	0.0133
18 ^{2}A	−0.58 (ud2u2) +0.45 (u2du2)	32 467	0.0307	0.0031	0.5266
19 ^{2}A	−0.40 (2udu2) +0.38 (2022u) +0.38 (202u2) −0.30 (u2d2u)	32 864	0.1310	0.0980	0.1134
20 ^{2}A	0.51 (022u2) +0.41 (u2du2) −0.39 (2ud2u)	32 897	0.4325	0.2606	0.0108
21 ^{2}A	−0.46 (2u022) +0.38 (u2022)	34 311	0.0247	0.4635	0.0175
22 ^{2}A	−0.57 (udu22) +0.40 (uu2d2) +.31 (ud22u)	34 732	0.5894	0.0919	0.0118
23 ^{2}A	0.43 (220u2) +0.40 (2u022) +0.39 (u2022)	34 898	0.0275	0.0173	0.5097
24 ^{2}A	−0.49 (20u22) +0.45 (02u22) −0.32 (uu22d)	35 717	0.0260	0.0014	0.0175
25 ^{2}A	−0.45 (udu22) +0.40 (u2ud2) +0.38 (2uud2) −0.37 (ud22u)	36 298	0.7669	0.1846	0.0183
26 ^{2}A	0.75 (uud22)	36 557	0.0012	0.0033	0.3389
27 ^{2}A	−0.57 (u0222) +0.55 (0u222)	37 356	0.0006	0.0024	0.1563
28 ^{2}A	0.61 (u0222) +0.60 (0u222)	37 660	0.1560	0.0052	0.0001
29 ^{2}A	0.30 (u22ud)	47 832	0.1514	0.1085	0.0001
30 ^{2}A	−0.39 (u2220) −0.36 (2uud2) −0.35 (2u220) −0.32 (220u2)	48 183	0.0001	0.0007	0.3052
31 ^{2}A	0.34 (2u2ud) −0.33 (u2u2d)	48 892	0.0004	0.0043	0.0689
32 ^{2}A	0.45 (ud22u) −0.43 (22u02) −0.38 (2u202)	49 194	0.0721	0.0001	0.0061
33 ^{2}A	0.40 (uu22d) +0.36 (ud2u2) −0.33 (20u22)	49 327	0.0021	0.0016	0.0003
34 ^{2}A	0.45 (udu22)	49 808	0.0757	0.0294	0.0003
35 ^{2}A	0.49 (2udu2) +0.35 (2022u) −0.34 (0222u)	50 131	0.0021	0.0002	0.0008
36 ^{2}A	0.50 (0222u) +0.46 (2022u)	72 823	0.0175	0.0808	0.0003
37 ^{2}A	−0.37 (u2022) −0.35 (u2220)	73 119	0.0006	0.0062	0.0050

38 ^{2}A	−0.31 (u0222) −0.46 (220u2) +0.41 (u2022) −0.41 (022u2) −0.36 (202u2)	74 198				0.0004	0.0001	0.0324
39 ^{2}A	−0.49 (20u22) −0.41 (02u22) +0.37 (202u2)	74 719				0.0036	0.0223	0.0021
40 ^{2}A	−0.45 (2u022) −0.44 (2u202)	75 076				0.0005	0.0127	0.0022

[a] The CAS-CI configurations having the coefficient $|c| > 0.3$ are listed. 2, 0, u, d represents for doubly occupied, unoccupied, spin up (α), and spin down (β), respectively. [b] Contributions to the **g**-tensor principal values. [c] Contributions to the $\mathbf{D}^{SO}$-tensor principal values.

Table S11. The electronic states of complex **2** calculated at the CASSCF(7e,5o)/Sapporo-DZP(Co) and 3-21G(other nuclei) level.

State	Main configurations[a]	ΔE/cm^{-1}	g_{ii}[b]			D^{SO}_{ii}[c]/cm^{-1}		
			$i = x$	$i = y$	$i = z$	$i = x$	$i = y$	$i = z$
1 ^{4}A	1.00 (22uuu)	0						
2 ^{4}A	−0.71 (2u2uu) +0.49 (u22uu) −0.43 (uu22u)	1 977	0.0003	0.0000	0.4439	−0.0316	−0.0106	−44.2742
3 ^{4}A	−0.69 (u22uu) −0.51 (2u2uu) +0.35 (2uu2u)	3 007	0.0054	0.2529	0.0002	−7.0272	−18.9203	−0.0193
4 ^{4}A	0.70 (u2u2u) −0.43 (u2uu2) −0.33 (2uu2u) +0.30 (uu22u)	6 518	0.1453	0.0007	0.0294	−9.8770	−4.9400	−3.1113
5 ^{4}A	0.66 (2uu2u) +0.53 (uu22u) +0.43 (2uuu2)	6 860	0.0494	0.0017	0.0595	−4.1264	−1.2630	−6.0028
6 ^{4}A	0.75 (uu2u2) −0.52 (u2uu2) −0.40 (2uuu2)	10 740	0.0220	0.0129	0.0000	−3.6012	−0.0043	−0.0054
7 ^{4}A	0.88 (uuu22)	14 958	0.0000	0.0000	0.0001	−0.0056	−0.0001	−0.0148
8 ^{4}A	−0.60 (uu22u) +0.50 (u2u2u) −0.32 (u22uu) +0.31 (2uu2u) +0.31 (uuu22)	24 248	0.0001	0.0001	0.0003	−0.0171	−0.0001	−0.0270
9 ^{4}A	−0.52 (uu2u2) −0.51 (2uuu2) +0.43 (2uu2u) +0.33 (u22uu) −0.31 (u2uu2)	25 270	0.0007	0.0008	0.0000	−0.1587	−0.0031	−0.0024
10 ^{4}A	−0.59 (u2uu2) +0.54 (2uuu2) −0.33 (uuu22) +0.31 (2u2uu)	26 129	0.0000	0.0000	0.0010	−0.0050	−0.0003	−0.1209
1 ^{2}A	−0.90 (222u0)	15 467				0.0263	0.0393	2.1946
2 ^{2}A	0.84 (2220u)	18 290				0.9997	2.9740	0.0355
3 ^{2}A	−0.73 (22u20) −0.42 (2u220) +0.34 (u2220)	19 024				0.2786	0.0039	0.0017
4 ^{2}A	−0.80 (22udu) +0.31 (22uud)	20 944				0.0006	0.0000	0.0449
5 ^{2}A	0.56 (22uud) −0.36 (2u2ud) +0.35 (2u2du)	21 257				0.0030	0.0001	1.8859
6 ^{2}A	−0.49 (2u2du) +0.36 (u22ud)	22 401				1.2926	0.5125	0.3884
7 ^{2}A	−0.53 (u22du) +0.32 (u2202)	22 509				0.4278	0.0923	1.0595
8 ^{2}A	−0.47 (u22ud) +0.43 (2uu2d) −0.41 (2u2ud) −0.39 (u2u2d)	22 896				0.0067	0.6552	0.0158
9 ^{2}A	−0.58 (2ud2u) +0.48 (u2d2u) −0.33 (2202u)	25 001				0.3946	3.8991	0.0004
10 ^{2}A	0.35 (u22ud) +0.35 (u22du) −0.33 (22u20) +0.31 (2u220) −0.31 (u2220) −0.30 (ud22u)	26 152				0.1104	0.1489	0.0171
11 ^{2}A	−0.58 (22uud) −0.46 (2u2ud) −0.37 (uu22d)	26 873				0.0104	0.0079	0.0549
12 ^{2}A	0.56 (uu22d) +0.32 (u2u2d)	27 925				0.0001	0.0000	0.0002
13 ^{2}A	−0.56 (2202u) +0.33 (2022u)	29 177				0.5921	0.0427	0.0003
14 ^{2}A	0.50 (u2d2u) +0.35 (2ud2u) +0.31 (u2202) −0.30 (0222u)	30 028				0.0287	0.0016	0.3716

15 ^{2}A	−0.61 (22u02) +0.34 (u2ud2)	30 829				0.1929	0.3658	0.0011
16 ^{2}A	0.44 (2uud2) −0.38 (u2202) −0.33 (2u202) +0.31 (u2u2d)	31 733				0.0002	0.0081	0.0075
17 ^{2}A	−0.75 (uu2d2)	32 148				0.0086	0.0524	0.0082
18 ^{2}A	−0.47 (ud2u2) −0.44 (2udu2) +0.39 (u2du2)	32 661				0.0169	0.0003	0.8967
19 ^{2}A	0.47 (u2du2) +0.44 (022u2) −0.39 (2ud2u)	33 234				0.4783	0.1363	0.0167
20 ^{2}A	−0.42 (2udu2) −0.38 (u2d2u) +0.36 (202u2) +0.34 (2022u) +0.32 (ud2u2)	33 318				0.3034	0.0604	0.0378
21 ^{2}A	0.45 (2u022) −0.36 (2202u) +0.33 (ud22u) −0.32 (u2022)	34 777				0.4990	0.3616	0.0014
22 ^{2}A	0.45 (udu22) +0.42 (uu2d2) +0.31 (u2u2d)	35 350				0.3930	0.0217	0.0333
23 ^{2}A	0.48 (u2022) −0.45 (220u2)	35 449				0.0627	0.0055	0.6004
24 ^{2}A	0.49 (20u22) −0.42 (02u22)	36 234				0.0300	0.0127	0.0448
25 ^{2}A	0.47 (udu22) +0.46 (u2ud2) +0.33 (02u22) −0.32 (ud22u) +0.32 (2uud2)	36 896				0.5533	0.2555	0.0013
26 ^{2}A	0.79 (uud22)	37 286				0.0017	0.0025	0.2781
27 ^{2}A	−0.65 (u0222) +0.52 (0u222)	37 929				0.0011	0.0005	0.0218
28 ^{2}A	0.66 (0u222) +0.56 (u0222)	38 429				0.0720	0.0027	0.0006
29 ^{2}A	−0.34 (2u220) L0.31 (u22ud) −0.31 (022u2)	48 105				0.1054	0.1623	0.0056
30 ^{2}A	0.45 (u2220) −0.36 (220u2)	48 394				0.0102	0.0021	0.2768
31 ^{2}A	0.33 (u2u2d) −0.33 (2uud2) +0.31 (2u2ud)	49 138				0.0015	0.0001	0.0610
32 ^{2}A	0.42 (ud22u) +0.38 (2u202) +0.37 (22u02) −0.33 (u2202)	49 520				0.0908	0.0000	0.0129
33 ^{2}A	0.37 (uu22d) −0.34 (02u22) +0.31 (ud2u2) +0.31 (20u22)	49 714				0.0000	0.0000	0.0041
34 ^{2}A	−0.52 (udu22) +0.33 (uu2d2) +0.30 (u2ud2)	50 242				0.0625	0.0284	0.0000
35 ^{2}A	0.42 (2udu2) −0.35 (ud2u2) −0.35 (u2du2) +0.30 (uud22)	50 556				0.0002	0.0012	0.0015
36 ^{2}A	−0.50 (0222u) −0.47 (2022u) −0.33 (2202u)	73 080				0.0304	0.0609	0.0015
37 ^{2}A	−0.38 (u2022) −0.36 (u2220) −0.31 (u0222)	73 478				0.0002	0.0031	0.0029
38 ^{2}A	−0.37 (220u2) −0.36 (u2022)	74 598				0.0001	0.0082	0.0310
39 ^{2}A	0.54 (20u22) +0.42 (202u2) +0.35 (02u22) −0.30 (22u02)	74 969				0.0018	0.0114	0.0197
40 ^{2}A	−0.51 (2u022) −0.43 (2u202)	75 492				0.0003	0.0060	0.0042

[a] The CAS-CI configurations having the coefficient $|c|$ > 0.3 are listed. 2, 0, u, d represents for doubly occupied, unoccupied, spin up (α), and spin down (β), respectively. [b] Contributions to the **g**-tensor principal values. [c] Contributions to the $\mathbf{D}^{SO}$-tensor principal values.

Table S12. The electronic states of complex **3** calculated at the CASSCF(7e,5o)/Sapporo-DZP(Co) and 3-21G(other nuclei) level.

State	Main configurations[a]	ΔE/cm^{-1}	g_{ii}[b]			$D^{SO}{}_{ii}$[c]/cm^{-1}		
			$i = x$	$i = y$	$i = z$	$i = x$	$i = y$	$i = z$
1 ^{4}A	−0.99 (22uuu)	0						
2 ^{4}A	−0.87 (u22uu) −0.40 (uu22u)	2 107	0.0006	0.0021	0.4802	−0.0145	−0.5993	−47.1967
3 ^{4}A	−0.85 (2u2uu) +0.47 (u2u2u)	2 971	0.0131	0.2198	0.0009	−14.7149	−8.6068	−0.1838
4 ^{4}A	0.69 (2uu2u) +0.59 (uu22u)	6 117	0.0077	0.0019	0.1020	−0.1673	−0.5887	−10.4763
5 ^{4}A	0.72 (u2u2u) +0.46 (2uuu2)	7 542	0.1591	0.0081	0.0011	−6.5924	−10.6713	−0.0418

6 ^{4}A	−0.75 (uu2u2) +0.60 (2uuu2)	11 242	0.0113	0.0121	0.0033	−0.0177	−2.3647	−0.4181
7 ^{4}A	0.84 (uuu22) −0.33 (2uu2u) +0.32 (u2uu2)	14 915	0.0000	0.0001	0.0002	−0.0077	−0.0062	−0.0248
8 ^{4}A	0.54 (2uuu2) −0.45 (u2u2u) +0.42 (uu2u2) −0.41 (2u2uu) −0.35 (u2uu2)	23 951	0.0009	0.0013	0.0001	−0.0001	−0.2476	−0.0043
9 ^{4}A	−0.57 (uu22u) +0.54 (2uu2u) +0.45 (uuu22) +0.36 (u22uu)	25 363	0.0000	0.0000	0.0001	−0.0012	−0.0017	−0.0062
10 ^{4}A	0.76 (u2uu2) +0.33 (uu2u2)	28 242	0.0000	0.0001	0.0011	−0.0062	−0.0018	−0.1288
1 ^{2}A	−0.89 (222u0)	14 956				0.1541	1.0736	1.3118
2 ^{2}A	0.74 (22u20) −0.48 (u2220)	18 397				1.1163	0.0010	0.0101
3 ^{2}A	−0.81 (2220u) −0.34 (2u2du)	18 835				1.6437	1.1612	0.0535
4 ^{2}A	−0.66 (22udu) +0.45 (22uud) +0.40 (u22ud)	21 072				0.0328	0.0000	0.0652
5 ^{2}A	−0.55 (22udu) −0.43 (22uud) −0.34 (u22ud) +0.33 (u22du)	21 273				0.0026	0.1043	1.0538
6 ^{2}A	0.52 (u22du) +0.47 (2u2ud)	22 157				0.1195	0.4488	1.7226
7 ^{2}A	0.57 (u2u2d) −0.53 (2u2ud)	22 779				0.6743	0.0383	0.1218
8 ^{2}A	−0.49 (2u2du) −0.41 (u22du)	22 908				0.1988	1.3383	0.5310
9 ^{2}A	0.67 (u2d2u) −0.42 (2202u)	25 271				4.5578	0.0296	0.0008
10 ^{2}A	0.45 (22uud) −0.36 (2u2du) −0.32 (u2220)	25 737				0.0005	0.2453	0.0951
11 ^{2}A	0.49 (uu22d) +0.48 (u22ud) −0.44 (22uud)	26 791				0.0020	0.0160	0.0218
12 ^{2}A	−0.48 (uu22d) −0.43 (2uu2d) +0.35 (u22du)	27 882				0.0009	0.0364	0.0136
13 ^{2}A	−0.52 (2202u) +0.41 (0222u) +0.35 (2u2du)	29 731				0.0198	0.6789	0.0450
14 ^{2}A	0.55 (2ud2u) +0.50 (2u202) −0.34 (ud22u)	30 051				0.0302	0.0144	0.2522
15 ^{2}A	0.59 (22u02) +0.33 (2uud2)	31 469				0.1923	0.1796	0.0136
16 ^{2}A	−0.49 (uu2d2)	32 568				0.0066	0.0177	0.0339
17 ^{2}A	−0.46 (u2ud2) +0.35 (202u2) −0.33 (2uu2d) +0.33 (2u202) −0.33 (220u2)	32 586				0.2105	0.0032	0.0026
18 ^{2}A	−0.48 (uu2d2) +0.36 (ud22u) +0.34 (202u2) +0.33 (2ud2u)	33 251				0.0104	0.0185	0.0142
19 ^{2}A	0.52 (2udu2) +0.42 (ud2u2) −0.31 (u2d2u)	33 566				0.0682	0.4600	0.2867
20 ^{2}A	−0.49 (u2du2) −0.40 (ud22u) +0.36 (2udu2)	33 806				0.0006	0.5287	0.4727
21 ^{2}A	−0.49 (u2022) −0.40 (ud2u2) +0.33 (2uud2) −0.32 (2202u)	35 238				0.3823	0.1741	0.0346
22 ^{2}A	0.43 (2u022) +0.42 (220u2) −0.30 (0222u)	35 567				0.0398	0.6308	0.0937
23 ^{2}A	0.46 (02u22) +0.38 (2u022)	35 906				0.0041	0.1197	0.1211
24 ^{2}A	0.40 (20u22) −0.40 (2uud2) +0.39 (uud22)	36 673				0.1957	0.3726	0.0045
25 ^{2}A	0.56 (udu22) +0.31 (2uud2) −0.31 (202u2)	36 784				0.0830	0.1508	0.1354
26 ^{2}A	0.57 (uud22) −0.47 (u0222) −0.36 (20u22)	38 149				0.0227	0.0356	0.1735

27 ^{2}A	0.61 (0u222) −0.55 (u0222)	38 579				0.0087	0.0514	0.0176
28 ^{2}A	0.57 (0u222) +0.41 (u0222) +0.39 (uud22)	39 175				0.0085	0.0045	0.1013
29 ^{2}A	0.41 (ud2u2) +0.37 (u2220) −0.35 (2udu2) −0.35 (2u2ud)	48 229				0.1472	0.0581	0.0501
30 ^{2}A	0.51 (u2ud2) −0.49 (2u220) +0.32 (022u2) −0.32 (220u2)	48 851				0.0001	0.0378	0.2359
31 ^{2}A	−0.35 (u22ud) −0.34 (udu22) −0.34 (2uu2d)	49 661				0.0267	0.0480	0.0267
32 ^{2}A	−0.37 (u2202) +0.33 (2022u) −0.33 (u2ud2)	50 595				0.0001	0.0051	0.0723
33 ^{2}A	0.40 (udu22) −034 (uu22d) −0.33 (022u2)	50 806				0.0192	0.0039	0.0026
34 ^{2}A	0.46 (u2du2) −0.35 (ud22u)	51 295				0.0285	0.0407	0.0281
35 ^{2}A	−0.38 (u2du2) −0.32 (20u22) +0.30 (02u22)	51 448				0.0037	0.0191	0.0000
36 ^{2}A	−0.57 (2022u) −0.35 (0222u) −0.32 (20u22) −0.32 (2202u)	72 257				0.0418	0.0242	0.0044
37 ^{2}A	0.47 (2u022) +0.42 (2u220) −0.39 (0u222) +0.36 (2u202)	73 667				0.0009	0.0001	0.0001
38 ^{2}A	−0.48 (022u2) −0.41 (220u2) −0.38 (202u2)	75 603				0.0005	0.0164	0.0209
39 ^{2}A	0.42 (02u22) +0.34 (20u22) −0.34 (u2022)	76 847				0.0128	0.0007	0.0001
40 ^{2}A	−0.41 (u2022) −0.37 (02u22) −0.33 (u2202)	77 572				0.0296	0.0128	0.0076

[a] The CAS-CI configurations having the coefficient |*c*| > 0.3 are listed. 2, 0, u, d represents for doubly occupied, unoccupied, spin up (α), and spin down (β), respectively. [b] Contributions to the **g**-tensor principal values. [c] Contributions to the $\mathbf{D}^{SO}$-tensor principal values.

4. Quantum chemical calculations on the electronic structure and magnetic tensors of complex 4

The molecular structure of complex **4** and principal axis system of the **D**-tensor calculated by CASSCF is given in Figure S32. The principal values and direction cosines of the **g-** and **A**-tensors of complex **4** in the principal axis system of the **D**-tensor, as calculated by CASSCF, are given in Table S13.

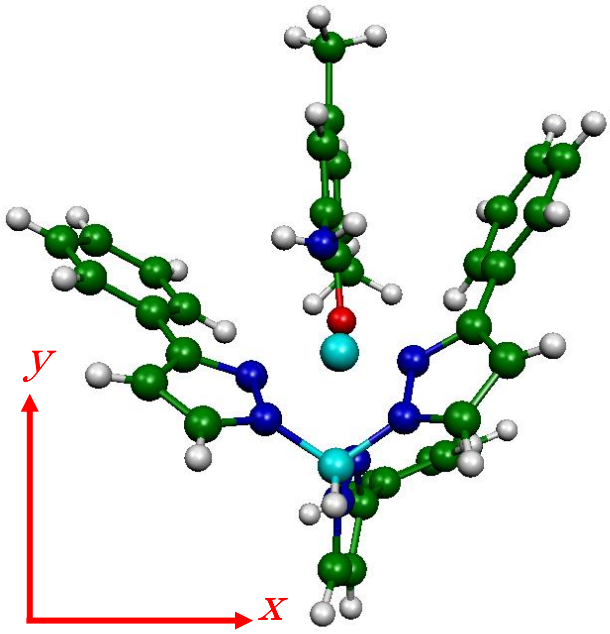

Fig. S32 The molecular structure and principal axis coordinate of the **D**-tensor of complex **4**, as calculated by CASSCF.

Table S13 The principal values and direction cosines of the magnetic tensors of complex **4** in the principal axis system of the **D**-tensor, as calculated by CASSCF.

a) **g**-tensor

	g_{xx}	g_{yy}	g_{zz}
Principal values	**2.2419**	**2.047944**	**2.661085**
X	0.9992315	0.0371124	0.0126125
Y	−0.0368430	0.9991012	−0.0209604
Z	0.0133791	−0.0204796	−0.9997007

b) **A**-tensor

	A_{xx}/MHz	A_{yy}/MHz	A_{zz}/MHz
Principal values	**122.9381**	**63.7055**	**178.7272**
X	0.9658926	−0.2584846	−0.0153994
Y	0.2382587	0.8638740	0.4437956
Z	0.1014112	0.4323279	−0.8959957

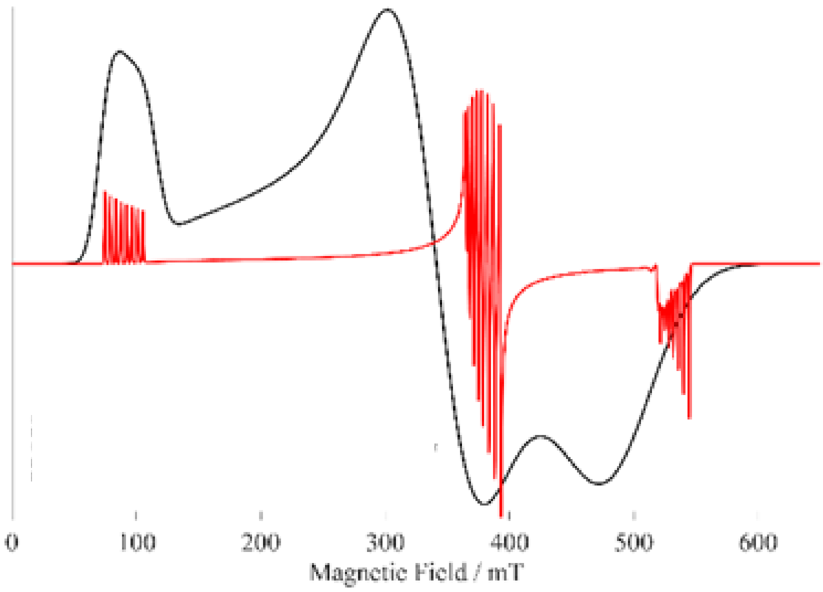

Fig. S33 (black) The reproduced randomly-oriented X-band ESR spectrum of complex **4** (S = 3/2) shown in the literature [10] and (red) the spectrum simulated by use of the theoretical magnetic parameters given in Table S12 including the relative coordination between **D**- and **g**-/**A**-tensors. Microwave frequency used: 9.474GHz; (the spectrum in black) the magnetic parameters: $g_1^{eff} = 7.2$, $g_2^{eff} = 1.97$, $g_3^{eff} = 1.4$, $A_1^{eff}(^{59}Co) = 560.5$ MHz, the peak-to-peak linewidth: 16.0 mT, the strain parameters of the linewidth: [0, 1850, 1600] MHz; (the spectrum in red) the magnetic parameters: **g** = [2.2419, 2.047944, 2.661085], **A**(^{59}Co) = [122.9381, 63.7055, 178.7272] MHz, $D = -48.94$ cm^{-1}, $E = -12.65$ cm^{-1} ($E/D = 0.26$), the peak-to-peak linewidth: 1.0 mT. Any strain effect of the tensor to the linewidth was not included in the spectrum in red. The simulated spectra were obtained by using EasySpin (ver. 6.0.6) [8].

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