Supplemental Materials for "Nonlinear Orbital and Spin Edelstein Effect in Centrosymmetric Metals"

I. MOYAL-KELDYSH FORMALISM FOR NONLINEAR RESPONSE

We derive the time-reversal-even nonlinear current-induced orbital angular momentum accumulation and current-induced spin accumulation using the Moyal-Keldysh approach [S1], which expands the Dyson equation in static electric and magnetic fields, thus enabling us to evaluate a nonequilibrium Green function up to any required order in this perturbation:

$$\hat{G}(\pi) = \hat{G}_0(\pi) + e\hbar E_i \hat{G}_{E_i}(\pi) + \frac{e^2\hbar^2}{2} E_i E_j \hat{G}_{E_i,E_j}(\pi) + ... \tag{S1}$$

Here, $\pi = (\pi_0,\ \boldsymbol{\pi}) = (\pi_0,\ \hbar\mathbf{k})$, $\hat{G}_0(\pi)$ is the Green function at equilibrium, and E_i is the i-component of the applied electric field. Reference [S1] proposes the matrix form of the Green function and its self-energy as

$$\hat{G}(\pi) = \begin{pmatrix} G^R(\pi) & 2G^<(\pi) \\ 0 & G^A(\pi) \end{pmatrix}, \tag{S2a}$$

$$\hat{\Sigma}(\pi) = \begin{pmatrix} \Sigma^R(\pi) & 2\Sigma^<(\pi) \\ 0 & \Sigma^A(\pi) \end{pmatrix}, \tag{S2b}$$

where $G^R(\Sigma^R)$, $G^A(\Sigma^A)$, and $G^<(\Sigma^<)$ are the retarded, advanced, and lesser Green functions (self-energies) respectively. We use a constant broadening $\eta = \hbar/2\tau$, where τ is the relaxation time, to simplify the self energies and take the $\eta \to 0$ limit [S2]. Then the equilibrium Green functions can be simplified as $G_0^R(\pi) = (\pi_0 - \hat{H}_0(\boldsymbol{\pi}) + i\eta)^{-1}$, $G_0^A = (G_0^R)^\dagger$, and $G_0^< = f(\pi)(G_0^A - G_0^R)/2i$ for the equilibrium Hamiltonian $\hat{H}_0$. The second-order retarded second-order Green function $G^R_{E_i,E_j}(i \neq j)$ can be simplified as

$$\begin{aligned} G^R_{E_i,E_j} = G_0^R\Big[\frac{i}{2}\Big(&\partial_{\pi_i}(G_0^R)^{-1}\partial_{\pi_0}G^R_{E_j} - \partial_{\pi_0}(G_0^R)^{-1}\partial_{\pi_i}G^R_{E_j} \\ &+\partial_{\pi_j}(G_0^R)^{-1}\partial_{\pi_0}G^R_{E_i} - \partial_{\pi_0}(G_0^R)^{-1}\partial_{\pi_j}G^R_{E_i}\Big) \\ &+\frac{1}{4}\partial_{\pi_i}\partial_{\pi_j}(G_0^R)^{-1}\partial^2_{\pi_0}G_0^R\Big], \end{aligned} \tag{S3a}$$

$$\begin{aligned} G^R_{E_i} = G_0^R\Big[\frac{i}{2}\Big(&\partial_{\pi_0}(G_0^R)^{-1}G_0^R\partial_{\pi_i}(G_0^R)^{-1} \\ &-\partial_{\pi_i}(G_0^R)^{-1}G_0^R\partial_{\pi_0}(G_0^R)^{-1}\Big)\Big]G_0^R, \end{aligned} \tag{S3b}$$

and the advanced Green function is $G^A_{E_i,E_j} = (G^R_{E_i,E_j})^\dagger$. The second-order lesser Green function can be written as $\hat{G}^<_{E_i,E_j} = \hat{G}^{<,\mathrm{I}}_{E_i,E_j} f'(\pi_0) + \hat{G}^{<,\mathrm{II}}_{E_i,E_j} f(\pi_0) + \hat{G}^{<,\mathrm{III}}_{E_i,E_j} f''(\pi_0)$ with the Fermi-Dirac distribution function $f(\pi_0)$, where each component is

$$\begin{aligned} G^{<,\mathrm{I}}_{E_i,E_j} = G_0^R\Big[\frac{i}{2}\Big(&\partial_{\pi_i}(G_0^R)^{-1}\partial_{\pi_0}G^{<,\mathrm{I}}_{E_j} + \partial_{\pi_i}(G_0^R)^{-1}G^{<,\mathrm{II}}_{E_j} \\ &-[(G_0^A)^{-1} - (G_0^R)^{-1}]\partial_{\pi_i}G^A_{E_j} - \partial_{\pi_0}(G_0^R)^{-1}\partial_{\pi_i}G^{<,\mathrm{I}}_{E_j} \\ &+(i \leftrightarrow j)\Big) \\ &+\frac{1}{2}\partial_{\pi_i}\partial_{\pi_j}(G_0^R)^{-1}\partial_{\pi_0}(G_0^A - G_0^R)\Big], \end{aligned} \tag{S4a}$$

$$G^{<,\mathrm{II}}_{E_i,E_j} = G^A_{E_i,E_j} - G^R_{E_i,E_j}, \tag{S4b}$$

$$\begin{aligned}G^{<,\mathrm{III}}_{E_i,E_j}=G_0^R\Big[\frac{i}{2}\Big(\partial_{\pi_i}(G_0^R)^{-1}G^{<,\mathrm{I}}_{E_j}+\partial_{\pi_j}(G_0^R)^{-1}G^{<,\mathrm{I}}_{E_i}\Big)\\ +\frac{1}{4}\partial_{\pi_i}\partial_{\pi_j}(G_0^R)^{-1}\partial_{\pi_0}[G_0^A-G_0^R]\\ +\frac{1}{4}[(G_0^A)^{-1}-(G_0^R)^{-1}]\partial_{\pi_i}\partial_{\pi_j}G_0^A\Big],\end{aligned}\tag{S4c}$$

where the first-order Green functions are

$$\begin{aligned}G^{<,\mathrm{I}}_{E_i}=\frac{i}{2}\Big[G_0^R\partial_{\pi_i}(G_0^R)^{-1}[G_0^A-G_0^R]\\ -[G_0^A-G_0^R]\partial_{\pi_i}(G_0^A)^{-1}G_0^A\Big],\end{aligned}\tag{S5a}$$

$$G^{<,\mathrm{II}}_{E_i}=G^A_{E_i}-G^R_{E_i}.\tag{S5b}$$

The second-order kth-components of the nonlinear CIOP and CISP $\langle\hat{X}_k\rangle=\chi^{X_k}_{ij}E_iE_j$ $(X_k=L_k,S_k)$ are evaluated by

$$\chi^{X_k}_{ij}=e^2\hbar^2\int\frac{d\boldsymbol{\pi_0}}{4\pi i}\int\frac{d^d\boldsymbol{\pi}}{(2\pi\hbar)^d}\mathrm{Tr}\Big[\mathrm{\hat{X}_k G^{<}_{E_i,E_j}}\Big],\tag{S6}$$

where d is the dimension of a system. Since the Green function has components dependent on f, f‘, and f“, the tensor $\chi^{X_k}_{ij}$ also has corresponding components such as

$$\begin{aligned}\chi^{X_k}_{ij}=\chi^{X_k,\mathrm{I}}_{ij}+\chi^{X_k,\mathrm{II}}_{ij}+\chi^{X_k,\mathrm{III}}_{ij}\\ =e^2\hbar^2\int\frac{d\boldsymbol{\pi_0}}{4\pi i}f‘(\pi_0)\int\frac{d^d\boldsymbol{\pi}}{(2\pi\hbar)^d}\mathrm{Tr}[\mathrm{\hat{X}_k G^{<,I}_{E_i,E_j}}]\\ +e^2\hbar^2\int\frac{d\boldsymbol{\pi_0}}{4\pi i}f(\pi_0)\int\frac{d^d\boldsymbol{\pi}}{(2\pi\hbar)^d}\mathrm{Tr}[\mathrm{\hat{X}_k G^{<,II}_{E_i,E_j}}]\\ +e^2\hbar^2\int\frac{d\boldsymbol{\pi_0}}{4\pi i}f“(\pi_0)\int\frac{d^d\boldsymbol{\pi}}{(2\pi\hbar)^d}\mathrm{Tr}[\mathrm{\hat{X}_k G^{<,III}_{E_i,E_j}}]\end{aligned}\tag{S7}$$

We are only interested in Fermi surface terms, so we only focus on the components dependent on f‘ and f“, where the latter could be transformed into the former using integration by parts. The other parts are calculated in the Appendix. The intraband term of f“-dependent component $\chi^{X_k,\mathrm{III}}_{ij}$ and some two-band terms of the f‘ component $\chi^{X_k,\mathrm{II,inter}}_{ij}$ correspond to the results from the Boltzmann equation $\chi^{X_k,\mathrm{boltz}}_{ij}$ [S3] using the f-sum rule [S4].

$$\chi^{X_k,\mathrm{III}}_{ij}=e^2\tau^2\sum_n\int\frac{d^d\boldsymbol{k}}{(2\pi)^d}f“(\varepsilon_{n\mathbf{k}})(\hat{X}_k)_n,\tag{S8a}$$

$$\chi^{X_k,\mathrm{II,inter}}_{ij}=e^2\tau^2\sum_{n\neq m}\mathrm{Re}\int\frac{\mathrm{d^dk}}{(2\pi)^\mathrm{d}}\mathrm{f‘(\varepsilon_{n\mathbf{k}})(\hat{X}_k)_n}\frac{1}{\hbar^2}\frac{\partial^2\varepsilon_{\mathrm{n}\mathbf{k}}}{\partial_{\mathrm{k_i}}\partial_{\mathrm{k_j}}},\tag{S8b}$$

where $(\hat{X}_k)_n=\langle u_{n\mathbf{k}}|\,\hat{X}_z\,|u_{n\mathbf{k}}\rangle$, v_i is the ith-component of the velocity operator, and $|u_{n\mathbf{k}}\rangle$ is a periodic part of the Bloch state with the energy eigenvalue $\varepsilon_{n\mathbf{k}}$. The other two-band terms form the time-reversal-even current-induced nonlinear OAM polarization (CIOP) and spin polarization (CISP).

$$\chi^{X_z}_{ij}=-e^2\hbar\tau\sum_{n\neq m}\int\frac{d^d\mathbf{k}}{(2\pi)^d}f‘(\varepsilon_{n\mathbf{k}})(\Omega^{ij}_{nm\mathbf{k}}+\Omega^{ji}_{nm\mathbf{k}}),\tag{S9a}$$

$$\Omega^{ij}_{nm\mathbf{k}}=\mathrm{Im}\Big[\frac{\langle u_{n\mathbf{k}}|\,\hat{X}_z\,|u_{m\mathbf{k}}\rangle\,\langle u_{m\mathbf{k}}|\,v_i\,|u_{n\mathbf{k}}\rangle}{(\varepsilon_{n\mathbf{k}}-\varepsilon_{m\mathbf{k}})^2}(v_j)_n\Big].\tag{S9b}$$

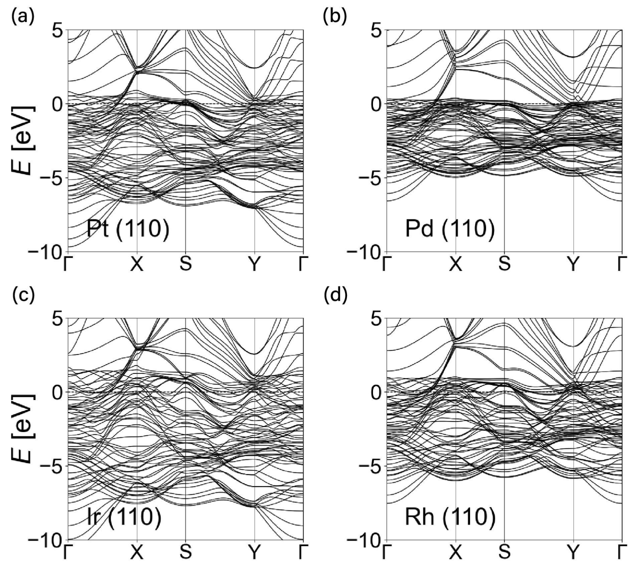

FIG. S1. The band structures of (110) 11-layered films for fcc Pt (a), fcc Pd (b), fcc Ir (c), and fcc Rh (d).

The nonlinear spin accumulation for Eqn. S9 is equivalent to the previous work [S5], which was derived by semiclassical approach. We denote the anomalous OAM dipole polarization α^{L}_{zij} and spin dipole polarization α^{S}_{zij} to $\chi^{X_z}_{ij} = e^2\hbar\tau\alpha^{X}_{zij}$ to evaluate the pure effect from the electronic band structures.

The remaining two-band Fermi-surface and all Fermi-sea terms generate the intrinsic nonlinear angular momentum accumulation in ferromagnets. Note that they are independent of the relaxation time and correspond to the intrinsic nonlinear response from the semiclassical approach:

$$
\begin{aligned}
\chi^{X_k,\mathrm{II}}_{ij} = -e^2\hbar^2 \,\mathrm{Re}\sum_{n}\int \frac{d^d\mathbf{k}}{(2\pi)^d} f(\varepsilon_{n\mathbf{k}}) \\
\Big[\sum_{n\neq m}\frac{3(\langle u_{n\mathbf{k}}|\hat{X}_k|u_{n\mathbf{k}}\rangle - \langle u_{m\mathbf{k}}|\hat{X}_k|u_{m\mathbf{k}}\rangle)\langle u_{n\mathbf{k}}|v_i|u_{m\mathbf{k}}\rangle\langle u_{m\mathbf{k}}|v_j|u_{n\mathbf{k}}\rangle}{(\varepsilon_{n\mathbf{k}}-\varepsilon_{m\mathbf{k}})^4} - \\
\sum_{n\neq m\neq l}\Big\{\frac{\langle u_{n\mathbf{k}}|\hat{X}_k|u_{m\mathbf{k}}\rangle\langle u_{m\mathbf{k}}|v_i|u_{l\mathbf{k}}\rangle\langle u_{l\mathbf{k}}|v_j|u_{n\mathbf{k}}\rangle}{(\varepsilon_{n\mathbf{k}}-\varepsilon_{m\mathbf{k}})(\varepsilon_{n\mathbf{k}}-\varepsilon_{l\mathbf{k}})^3} + (i\leftrightarrow j)\Big\} - \\
\sum_{n\neq m\neq l}\Big\{\frac{\langle u_{m\mathbf{k}}|\hat{X}_k|u_{l\mathbf{k}}\rangle\langle u_{l\mathbf{k}}|v_i|u_{n\mathbf{k}}\rangle\langle u_{n\mathbf{k}}|v_j|u_{m\mathbf{k}}\rangle}{(\varepsilon_{n\mathbf{k}}-\varepsilon_{m\mathbf{k}})^3(\varepsilon_{m\mathbf{k}}-\varepsilon_{l\mathbf{k}})} + (i\leftrightarrow j)\Big\}\Big],
\end{aligned}
\tag{S10a}
$$

$$
\begin{aligned}
\chi^{X_k,\mathrm{I}}_{ij} = -e^2\hbar^2 \,\mathrm{Re}\sum_{n\neq m}\int \frac{d^d\mathbf{k}}{(2\pi)^d} f'(\varepsilon_{n\mathbf{k}})\Big[\frac{\langle u_{n\mathbf{k}}|\hat{X}_k|u_{n\mathbf{k}}\rangle\langle u_{n\mathbf{k}}|v_i|u_{m\mathbf{k}}\rangle\langle u_{m\mathbf{k}}|v_j|u_{n\mathbf{k}}\rangle}{(\varepsilon_{n\mathbf{k}}-\varepsilon_{m\mathbf{k}})^3} - \\
\Big\{\frac{\langle u_{n\mathbf{k}}|\hat{X}_k|u_{m\mathbf{k}}\rangle\langle u_{m\mathbf{k}}|v_i|u_{n\mathbf{k}}\rangle\langle u_{n\mathbf{k}}|v_j|u_{n\mathbf{k}}\rangle}{(\varepsilon_{n\mathbf{k}}-\varepsilon_{m\mathbf{k}})^3} + (i\leftrightarrow j)\Big\} - \\
\Big\{\frac{\langle u_{n\mathbf{k}}|\hat{X}_k q|u_{m\mathbf{k}}\rangle\langle u_{m\mathbf{k}}|v_i|u_{m\mathbf{k}}\rangle\langle u_{m\mathbf{k}}|v_j|u_{n\mathbf{k}}\rangle}{2(\varepsilon_{n\mathbf{k}}-\varepsilon_{m\mathbf{k}})^3} + (i\leftrightarrow j)\Big\}\Big].
\end{aligned}
\tag{S10b}
$$

Equation S10a and the first part of Eqn. S10b is the same as the Fermi sea contribution of the previous work, while the second part of Eqn. S10b differs from the previous work by a factor 1/2 since that paper did not consider the $(i \leftrightarrow j)$ term in their corresponding term. There is also an additional two-band contribution for the nonlinear response, which has not been reported yet.

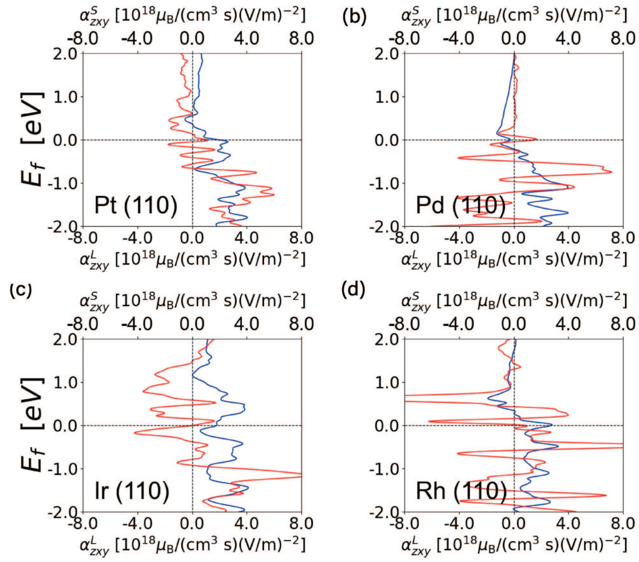

FIG. S2. The nonlinear OAM (blue line) and spin (red line) accumulation in (110) films for fcc Pt (a), fcc Pd (b), fcc Ir (c), and fcc Rh (d) at $T = 300$ K.

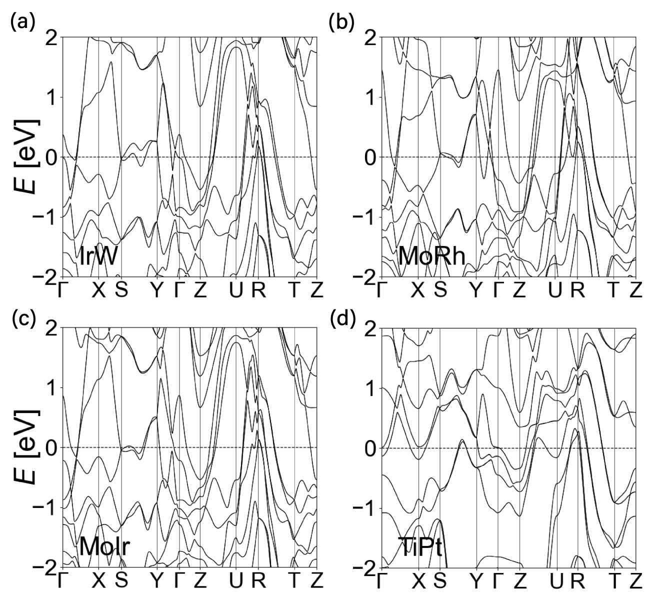

FIG. S3. The band structures for bulk orthorhombic IrW (a), MoRh (b), MoIr (c), and TiPt (d).

II. FERMI ENERGY DEPENDENCE OF THE NONLINEAR OAM AND SPIN ACCUMULATION

The electronic band structures of orthorhombic metals (IrW, MoRh, MoIr, and TiPt) and fcc (110) films made of Pt, Rh, Ir, and Pd with 11 layers are represented in Fig. S3 and Fig. S1 respectively. The nonlinear OAM and spin densities for bulk and film metals are represented in Fig. S4 and Fig. S2, respectively.

By setting the external electric field is set as $E = 10^5$ V/m [S6, S7] and the relaxation time for film elements obtained by their Drude conductivity and electronic concentration from Ref. [S8] (with the assumption that the relaxation time of bulk materials are set to 10 fs), we obtain the nonlinear OAM and spin accumulation at the true Fermi energy $E_\mathrm{f} = 0$ in Table 1.

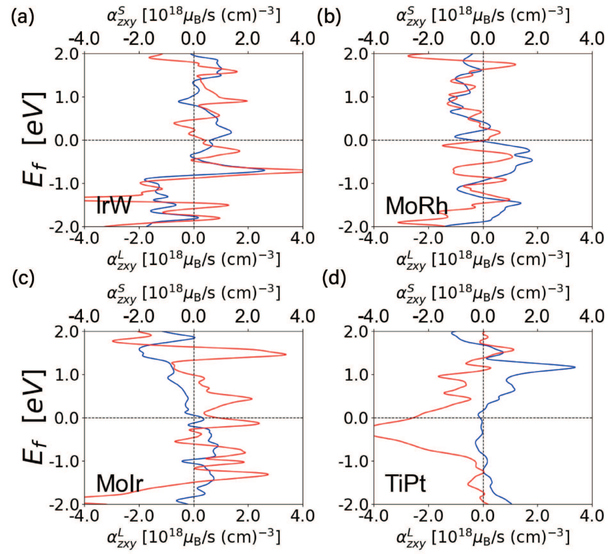

FIG. S4. The nonlinear OAM (blue line) and spin (red line) accumulation for bulk orthorhombic IrW (a), MoRh (b), MoIr (c), and TiPt (e) from DFT calculation at T = 300 K.

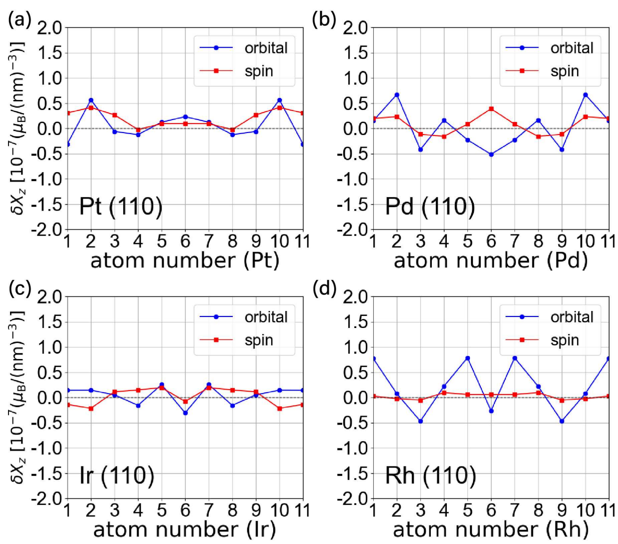

FIG. S5. The nonlinear OAM (blue line) and spin (red line) accumulation in (110) films fcc Pt (a), fcc Pd (b), fcc Ir (c), and fcc Rh (d) at $T = 300$ K.

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