# Supplementary Material
# Double Stokes Polarimetric Microscopy for Chiral Fibrillar Aggregates

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## A Double Stokes vector

Double Stokes vector formalism can be used to describe two electric fields of laser light. For the degenerate SHG case, the double Stokes vector is described by a 9-component vector as follows [1]:

$$S = \begin{bmatrix} S_1 \\ S_2 \\ S_3 \\ S_4 \\ S_5 \\ S_6 \\ S_7 \\ S_8 \\ S_9 \end{bmatrix} = \begin{bmatrix} \sqrt{1/6}(3s_0^2 - s_1^2) \\ \sqrt{1/12}(5s_1^2 - 3s_0^2) \\ s_0 s_1 \\ 1/2(s_2^2 - s_3^2) \\ -s_2(s_1 - s_0) \\ s_2(s_1 + s_0) \\ s_2 s_3 \\ s_3(s_1 - s_0) \\ s_3(s_1 + s_0) \end{bmatrix} \tag{S.1}$$

The double Stokes vector is expressed in terms of the first-order Stokes vector components $s_0$, $s_1$, $s_2$ and $s_3$ of a laser beam, analogous to the Eq. (1). The double Stokes vector is obtained via coherency matrix formalism using Gell-Mann matrices presented in Supplementary Material C [1].

## B Relation between Mueller matrix and molecular susceptibilities

The double Stokes-Mueller formalism (Eq. (2)) enables the calculation of the double Mueller matrix $M$ [1]. The double Mueller matrix elements are related to the second-order susceptibilities $\chi$ in the laboratory coordinate reference frame:

$$M_{\alpha N} = \frac{1}{2} Tr(\tau_\alpha \chi \lambda_N \chi^\dagger), \tag{S.2}$$

where $\tau_\alpha$ and $\lambda_N$ are the Pauli and Gell-Mann matrices, respectively, with index $\alpha = 0...3$, and index $N = 1...9$ (see $\tau_\alpha$ and $\lambda_N$ matrices in the Supplementary Material C). The susceptibilities are presented in the contracted notation $\chi_{IJK} = \chi_{IA}$, where the susceptibility tensor component index $I$ corresponds to the laboratory coordinate system $X$ and $Z$ axes, and $A$ is the contracted index representing $XX$, $ZZ$, and $XZ$ or $ZX$ [2]. Thus, $\chi$ is a $2 \times 3$ rectangular matrix. The superscript † denotes Hermitian conjugation.

The calculated laboratory frame nonlinear susceptibilities $\chi_{IJK}$ can be related to the molecular susceptibilities $\chi_{ijk}$ via tensor rotation [3]:

$$\chi_{IJK} = T_{Ii} T_{Jj} T_{Kk} \chi_{ijk}, \tag{S.3}$$

where $T_{Nn}$ are the rotation matrices, and Einstein summation is assumed. Therefore, the SHG Stokes vector components (Eq. (1)) can be related to the molecular susceptibility tensor elements.

## C Pauli and Gell-Mann matrices

The following Pauli (Eq. (S.4) and Gell-Mann (Eq. (S.5)) matrices are used for obtaining the expressions for the Stokes vector Eq. (1), the double Stokes vector (Eq. (S.1)), and the double Mueller matrix elements (Eq. (S.2)):

$$\tau_0 = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \tau_1 = \begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix} \tau_2 = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \tau_3 = \begin{pmatrix} 0 & i \\ -i & 0 \end{pmatrix}; \tag{S.4}$$

$$\begin{aligned} \lambda_1 &= \sqrt{\tfrac{2}{3}} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} & \lambda_2 &= \sqrt{\tfrac{1}{3}} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -2 \end{pmatrix} & \lambda_3 &= \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 0 \end{pmatrix} \\ \lambda_4 &= \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} & \lambda_5 &= \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix} & \lambda_6 &= \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix} \\ \lambda_7 &= \begin{pmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} & \lambda_8 &= \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ 0 & i & 0 \end{pmatrix} & \lambda_9 &= \begin{pmatrix} 0 & 0 & -i \\ 0 & 0 & 0 \\ i & 0 & 0 \end{pmatrix} \end{aligned} \tag{S.5}$$

## D Relations between the laboratory and the molecular frame nonlinear susceptibility tensor components

The laboratory reference frame nonlinear susceptibility tensor components can be expressed in terms of the molecular achiral $R$ ratio (Eq. (3)), the molecular chiral $C$ ratio magnitude and the phase difference $\Delta$ (Eq. (4)), and the in-plane orientation angle $\delta$ [3]:

$$\frac{\chi_{XXX}}{\chi'_{zxx}} = \sin\delta[(R-3)\sin^2\delta + 3] \tag{S.6}$$

$$\frac{\chi_{XXZ}}{\chi'_{zxx}} = \cos\delta[(R-3)\sin^2\delta + 1] - Ce^{i\Delta}\sin\delta \tag{S.7}$$

$$\frac{\chi_{XZZ}}{\chi'_{zxx}} = \sin\delta[(R-3)\cos^2\delta + 1] - 2Ce^{i\Delta}\cos\delta \tag{S.8}$$

$$\frac{\chi_{ZXX}}{\chi'_{zxx}} = \cos\delta[(R-3)\sin^2\delta + 1] + 2Ce^{i\Delta}\sin\delta \tag{S.9}$$

$$\frac{\chi_{ZXZ}}{\chi'_{zxx}} = \sin\delta[(R-3)\cos^2\delta + 1] + Ce^{i\Delta}\cos\delta \tag{S.10}$$

$$\frac{\chi_{ZZZ}}{\chi'_{zxx}} = \cos\delta[(R-3)\cos^2\delta + 3] \tag{S.11}$$

where $\chi'_{zxx} = \chi_{zxx}\cos\alpha$.

## E Sums of Stokes vector components

The following are expressions of the sums of SHG Stokes vector components in terms of the molecular susceptibility ratios. The equations are obtained for specific incident polarization states that are described by the double Stokes vector (Eq. (S.1)). The Stokes vector is calculated using Stokes-Mueller formalism (Eqs. (2) and (S.2)), and the relations between laboratory and molecular frame susceptibilities for $C_6$ symmetry fibers (Eqs. (S.6), (S.7), (S.8), (S.9), (S.10), (S.11)).

### E.1 0° and 90° incident linear polarizations

$$s_0^{HLP} + s_0^{VLP} = \frac{1}{4}\left((R-3)(R+1)\cos(4\,\delta) - 4\,(R-3)C\,\cos(\Delta)\,\sin(4\,\delta) + 3\,R^2 + 2\,R + 7 + 16\,C^2\right) \tag{S.12}$$

$$s_1^{HLP} + s_1^{VLP} = -\frac{1}{8}\left(\left(7R^2 + 6\,R - 1 - 32\,C^2\right)\,\cos(2\,\delta) + 16(R-1)\;C\,\cos(\Delta)\,\sin(2\,\delta) + (R-3)^2\cos(6\,\delta)\right) \tag{S.13}$$

$$s_2^{HLP} + s_2^{VLP} = \frac{1}{8}\left(-32\,R\,C\,\cos(\Delta)\,\cos(2\,\delta) + \left(5\,R^2 + 2\,R - 3\right)\,\sin(2\,\delta) + (R-3)^2\sin(6\,\delta)\right) \tag{S.14}$$

$$s_3^{HLP} + s_3^{VLP} = -(R-3)C\sin(\Delta)\cos(4\delta) - 3\,(R+1)C\sin(\Delta) \tag{S.15}$$

### E.2 45° and -45° incident linear polarizations

$$s_0^{+45} + s_0^{-45} = \frac{1}{4}\left(-(R-3)(R+1)\,\cos(4\,\delta) + 4(R-3)C\,\cos(\Delta)\,\sin(4\,\delta) + 3\,R^2 + 2\,R + 7 + 16\,C^2\right) \tag{S.16}$$

$$s_1^{+45} + s_1^{-45} = -\frac{1}{8}\left(\left(5R^2 + 2\,R - 3\right)\,\cos(2\,\delta) + 32\,R\,C\,\cos(\Delta)\,\sin(2\,\delta) - (R-3)^2\cos(6\,\delta)\right) \tag{S.17}$$

$$s_2^{+45} + s_2^{-45} = \frac{1}{8}\left(-16\,(R-1)\,C\,\cos(\Delta)\,\cos(2\,\delta) + \left(7\,R^2 + 6\,R - 1 - 32\,C^2\right)\,\sin(2\,\delta) - (R-3)^2\sin(6\,\delta)\right) \tag{S.18}$$

$$s_3^{+45} + s_3^{-45} = (R-3)C\sin(\Delta)\cos(4\delta) - 3\,(R+1)C\sin(\Delta) \tag{S.19}$$

### E.3 Left and right incident circular polarizations

$$s_0^{LCP} + s_0^{RCP} = \frac{1}{2}\left((R-1)^2 + 4 + 8\,C^2\right) \tag{S.20}$$

$$s_1^{LCP} + s_1^{RCP} = -\frac{1}{2}\left((R-3)(R+1)\cos(2\delta) + 4(R-3)C\cos(\Delta)\sin(2\delta)\right) \tag{S.21}$$

$$s_2^{LCP} + s_2^{RCP} = \frac{1}{2}\left((R-3)(R+1)\sin(2\delta) - 4(R-3)C\cos(\Delta)\cos(2\delta)\right) \tag{S.22}$$

$$s_3^{LCP} + s_3^{RCP} = -2(R+1)C\sin(\Delta) \tag{S.23}$$

## F Differences of Stokes vector components

The differences of SHG Stokes vector components are calculated the same way as the sums described in the previous section. The ratios of the differences and sums of the Stokes vector components provide the expressions of DSP polarimetric parameters described in the article.

### F.1 0° and 90° incident linear polarizations

$$s_0^{HLP} - s_0^{VLP} = 2\,(R-1)\,C\,\cos(\Delta)\,\sin(2\,\delta) - \left((R-1)\,(R+1) + 4\,C^2\right)\cos(2\,\delta) \tag{S.24}$$

$$s_1^{HLP} - s_1^{VLP} = \frac{1}{2}\left((R-3)\,(R+1)\,\cos(4\,\delta) + 2\,(R-3)\,C\,\cos(\Delta)\,\sin(4\,\delta) + (R+1)^2 - 8\,C^2\right) \tag{S.25}$$

$$s_2^{HLP} - s_2^{VLP} = \frac{1}{2}\left(2\,(R-3)\,C\,\cos(\Delta)\,\cos(4\,\delta) - (R-3)\,(R+1)\,\sin(4\,\delta) + 6\,(R+1)\,C\,\cos(\Delta)\right) \tag{S.26}$$

$$s_3^{HLP} - s_3^{VLP} = 4\,R\,C\,\sin(\Delta)\,\cos(2\,\delta) \tag{S.27}$$

### F.2 45° and -45° incident linear polarizations

$$s_0^{+45} - s_0^{-45} = \left((R-1)\,(R+1) + 4\,C^2\right)\sin(2\,\delta) + 2\,(R-1)\,C\,\cos(\Delta)\,\cos(2\,\delta) \tag{S.28}$$

$$s_1^{+45} - s_1^{-45} = \frac{1}{2}\left(2\,(R-3)\,C\,\cos(\Delta)\,\cos(4\,\delta) - (R-3)\,(R+1)\,\sin(4\,\delta) - 6\,(R+1)\,C\,\cos(\Delta)\right) \tag{S.29}$$

$$s_2^{+45} - s_2^{-45} = \frac{1}{2}\left(-(R-3)\,(R+1)\,\cos(4\,\delta) - 2\,(R-3)\,C\,\cos(\Delta)\,\sin(4\,\delta) + (R+1)^2 - 8\,C^2\right) \tag{S.30}$$

$$s_3^{+45} - s_3^{-45} = -4\,R\,C\,\sin(\Delta)\,\sin(2\,\delta) \tag{S.31}$$

### F.3 Left and right incident circular polarizations

$$s_0^{LCP} - s_0^{RCP} = -2\,(R+1)\,C\,\sin(\Delta) \tag{S.32}$$

$$s_1^{LCP} - s_1^{RCP} = 2\,(R-3)\,C\,\sin(\Delta)\,\cos(2\,\delta) \tag{S.33}$$

$$s_2^{LCP} - s_2^{RCP} = -2\,(R-3)\,C\,\sin(\Delta)\,\sin(2\,\delta) \tag{S.34}$$

$$s_3^{LCP} - s_3^{RCP} = 2\,(R-1) + 4\,C^2 \tag{S.35}$$

## References

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