

Supporting Information for Magneto-optical Kerr switching properties of $(\text{CrI}_3)_2$ and $(\text{CrBr}_3/\text{CrI}_3)$ bilayers

Ke Yang,[†] Wentao Hu,[†] Hua Wu,^{*,†,‡} Myung-Hwan Whangbo,^{¶,§,||} Paolo G. Radaelli,[⊥] and Alessandro Stroppa^{*,#}

[†]*Laboratory for Computational Physical Sciences (MOE), State Key Laboratory of Surface Physics, and Department of Physics, Fudan University, Shanghai 200433, China*

[‡]*Collaborative Innovation Center of Advanced Microstructures, Nanjing 210093, China*

[¶]*Department of Chemistry, North Carolina State University, Raleigh, North Carolina 27695-8204, USA*

[§]*State Key Laboratory of Crystal Materials, Shandong University, Jinan 250100, China*

^{||}*State Key Laboratory of Structural Chemistry, Fujian Institute of Research on the Structure of Matter, Chinese Academy of Sciences, Fuzhou, Fujian 350002, China*

[⊥]*Clarendon Laboratory, Department of Physics, University of Oxford, Parks Road, Oxford OX1 3PU, United Kingdom*

[#]*CNR-SPIN, c/o Department of Physical and Chemical Sciences, University of L' Aquila, 67100 Coppito (AQ), L' Aquila, Italy*

E-mail: wuh@fudan.edu.cn; alessandro.stroppa@spin.cnr.it

SI. Selection Rules

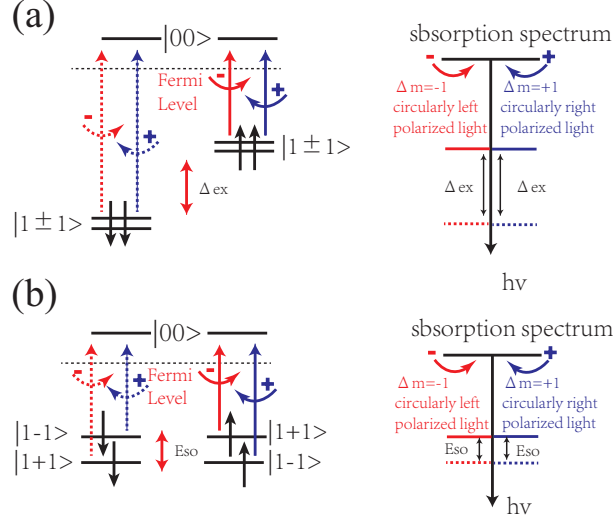


Figure S1: The simplified electronic structure and absorption spectrum. (a) the $p \rightarrow s$ transition without spin-orbit coupling. (b) the $p \rightarrow s$ transition without exchange splitting.

The real part σ'_ω and imaginary part σ''_ω of the conductivity tensor are, respectively, even and odd with respect to ω and are linked by the well-known Kramers-Kronig transformation.¹

$$\sigma'_{ij}(\mathbf{k}, \omega) = \frac{2}{\pi} P \int_0^\infty \frac{\omega' \sigma''_{ij}(\mathbf{k}, \omega')}{(\omega'^2 - \omega^2)} d\omega'$$

$$\sigma''_{ij}(\mathbf{k}, \omega) = -\frac{2}{\pi} \omega P \int_0^\infty \frac{\sigma'_{ij}(\mathbf{k}, \omega')}{(\omega'^2 - \omega^2)} d\omega'$$

In such case, the absorptive parts of optical conductivity tensor (real diagonal and imaginary off-diagonal elements) due to interband transitions can be obtained using Kubo's formula within linear response theory:²⁻⁴

$$\sigma'_{xx}(\omega) = \frac{\lambda}{\omega} \sum_{\mathbf{k}, jj'} \left[|\Pi_{jj'}^+|^2 + |\Pi_{jj'}^-|^2 \right] \delta(\omega - \omega_{jj'})$$

$$\sigma''_{xy}(\omega) = \frac{\lambda}{\omega} \sum_{\mathbf{k}, jj'} \left[|\Pi_{jj'}^+|^2 - |\Pi_{jj'}^-|^2 \right] \delta(\omega - \omega_{jj'})$$

where $\lambda = \frac{\pi e^2}{2\hbar m^2 V}$ is a material specific constant, $\hbar\omega$ is the photon energy, $\hbar\omega_{jj'}$ is the

energy difference between the occupied and unoccupied bands at the same k point, and $\Pi_{jj'}^{\pm} = \left\langle \mathbf{k}j \left| \frac{1}{\sqrt{2}} (\hat{p}_x \pm i\hat{p}_y) \right| \mathbf{k}j' \right\rangle$ are the dipole matrix elements for circularly polarized light with $\hat{p}_+ = \frac{1}{\sqrt{2}}(\hat{p}_x + i\hat{p}_y)$ (left) and $\hat{p}_- = \frac{1}{\sqrt{2}}(\hat{p}_x - i\hat{p}_y)$ (right), respectively. Besides, according to Kubo's formula, the selection rules for electric dipolar transitions must be satisfied, i.e., $\Delta l = \pm 1$, and $\Delta m_l = \pm 1$. The first selection rule implies that only transitions between s and p levels or between p and d levels are allowed. For the second selection rule, the transitions with $\Delta m_l = +1$ and $\Delta m_l = -1$ correspond to left and right circularly polarized light, respectively.^{5,6} More generally, absence of the Kerr effect is due to the fact that the value of the numerator of Eq. (1) off diagonal element of the optical conductivity tensor σ_{xy} is zero, *i.e.*,

$$|\langle \mathbf{k}j \uparrow | \hat{p}_+ | \mathbf{k}j' \uparrow \rangle| = |\langle \mathbf{k}j \downarrow | \hat{p}_- | \mathbf{k}j' \downarrow \rangle|$$

$$|\langle \mathbf{k}j \uparrow | \hat{p}_- | \mathbf{k}j' \uparrow \rangle| = |\langle \mathbf{k}j \downarrow | \hat{p}_+ | \mathbf{k}j' \downarrow \rangle|$$

(corresponding to the case without exchange splitting see FIG.S1(b))

$$|\langle \mathbf{k}j \downarrow | \hat{p}_+ | \mathbf{k}j' \downarrow \rangle| = |\langle \mathbf{k}j \downarrow | \hat{p}_- | \mathbf{k}j' \downarrow \rangle|$$

$$|\langle \mathbf{k}j \uparrow | \hat{p}_+ | \mathbf{k}j' \uparrow \rangle| = |\langle \mathbf{k}j \uparrow | \hat{p}_- | \mathbf{k}j' \uparrow \rangle|$$

(corresponding to the case without spin-orbit coupling see FIG.S1(a))

In other words, both the spin-orbit coupling and exchange splitting have an essential role in the occurrence of MOKE.

SII. Symmetry-adapted form of the optical conductivity tensor σ_{ij} for all the magnetic point groups

It is well known that the magneto-optical Kerr effect is related to off-diagonal components of the optical conductivity tensor. In this section we consider the optical conductivity tensor

rather than the corresponding dielectric tensor. The quantities are related by the identity.⁷

$$\varepsilon_{ij}(\omega) = \delta_{ij} + i \frac{4\pi}{\omega} \sigma_{ij}(\omega)$$

The general form of the Kerr angle in arbitrary symmetry only involves the antisymmetric part of the optical conductivity tensor.⁸

$$\begin{aligned} \tilde{\sigma}_{ij} &= (\sigma_{ij} - \sigma_{ji})/2, i \neq j \\ \tilde{\sigma}_{ji} &= -(\sigma_{ij} - \sigma_{ji})/2, i \neq j \end{aligned} \quad (i, j = 1, 2, 3)$$

Now consider the two kinds of forms of the conductivity tensor:

$\sigma_{ij} = \sigma_{ji}$ ($i \neq j$), there is no antisymmetric part and therefore no MOKE;

$\sigma_{ij} \neq \sigma_{ji}$ ($i \neq j$), there is an antisymmetric part, consequently, MOKE is allowed.

Based on this fact, we list below the conductivity tensor σ_{ij} of total 122 magnetic point groups.⁹ In order for MOKE to be allowed, the magnetic point group should be one of the 31 pyromagnetic point groups which have an antisymmetric part: $(1, -1, 2, 2', m, m', 2/m, 2'/m', 2'2'2, m'm'2', m'm'2, m'm'm, 4, -4, 4/m, 42'2', 4m'm', -42'm', 4/mm'm', 3, -3, 32', 3m', -3m', 6, -6, 6/m, 62'2', 6m'm', -6m'2', 6/mm'm')$. More detail can be seen below, particularly in the tensors marked with red labels.

$$\begin{pmatrix} 1 & \mathbf{1} \\ \sigma_{ij} & 1 & 2 & 3 \\ 1 & \sigma_{11} & \sigma_{12} & \sigma_{13} \\ 2 & \sigma_{21} & \sigma_{22} & \sigma_{23} \\ 3 & \sigma_{31} & \sigma_{32} & \sigma_{33} \end{pmatrix} \begin{pmatrix} 2 & 11' \\ \sigma_{ij} & 1 & 2 & 3 \\ 1 & \sigma_{11} & \sigma_{12} & \sigma_{13} \\ 2 & \sigma_{12} & \sigma_{22} & \sigma_{23} \\ 3 & \sigma_{13} & \sigma_{23} & \sigma_{33} \end{pmatrix} \begin{pmatrix} 3 & \mathbf{-1} \\ \sigma_{ij} & 1 & 2 & 3 \\ 1 & \sigma_{11} & \sigma_{12} & \sigma_{13} \\ 2 & \sigma_{21} & \sigma_{22} & \sigma_{23} \\ 3 & \sigma_{31} & \sigma_{32} & \sigma_{33} \end{pmatrix} \begin{pmatrix} 4 & -11' \\ \sigma_{ij} & 1 & 2 & 3 \\ 1 & \sigma_{11} & \sigma_{12} & \sigma_{13} \\ 2 & \sigma_{12} & \sigma_{22} & \sigma_{23} \\ 3 & \sigma_{13} & \sigma_{23} & \sigma_{33} \end{pmatrix}$$

$$\begin{pmatrix} 117 & -4'3m' \\ \sigma_{ij} & 1 & 2 & 3 \\ 1 & \sigma_{11} & 0 & 0 \\ 2 & 0 & \sigma_{11} & 0 \\ 3 & 0 & 0 & \sigma_{11} \end{pmatrix}
\begin{pmatrix} 118 & m - 3m \\ \sigma_{ij} & 1 & 2 & 3 \\ 1 & \sigma_{11} & 0 & 0 \\ 2 & 0 & \sigma_{11} & 0 \\ 3 & 0 & 0 & \sigma_{11} \end{pmatrix}
\begin{pmatrix} 119 & m - 3m1' \\ \sigma_{ij} & 1 & 2 & 3 \\ 1 & \sigma_{11} & 0 & 0 \\ 2 & 0 & \sigma_{11} & 0 \\ 3 & 0 & 0 & \sigma_{11} \end{pmatrix}
\begin{pmatrix} 120 & m' - 3'm \\ \sigma_{ij} & 1 & 2 & 3 \\ 1 & \sigma_{11} & 0 & 0 \\ 2 & 0 & \sigma_{11} & 0 \\ 3 & 0 & 0 & \sigma_{11} \end{pmatrix}$$

$$\begin{pmatrix} 121 & m - 3m' \\ \sigma_{ij} & 1 & 2 & 3 \\ 1 & \sigma_{11} & 0 & 0 \\ 2 & 0 & \sigma_{11} & 0 \\ 3 & 0 & 0 & \sigma_{11} \end{pmatrix}
\begin{pmatrix} 122 & m' - 3'm' \\ \sigma_{ij} & 1 & 2 & 3 \\ 1 & \sigma_{11} & 0 & 0 \\ 2 & 0 & \sigma_{11} & 0 \\ 3 & 0 & 0 & \sigma_{11} \end{pmatrix}$$

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