Tuning Coupled Toroidic and Polar Orders in a Bilayer Antiferromagnet

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Note 1: Magnetoelectric susceptibility tensor

Starting from the constitutive relation: $P_i = \overleftrightarrow{\alpha}_{ij} H_j$, where P_i is the polarization (polar), H_j is the magnetic field (axial), and $\overleftrightarrow{\alpha}_{ij}$ is an axial tensor. Under a spatial symmetry operation *R*, $\overleftrightarrow{\alpha}_{ij}$ must satisfy Neumann's principle:

$$\overleftrightarrow{\alpha}_{ij} = (detR) R_{ik} R_{jl} \overleftrightarrow{\alpha}_{kl}$$

While under a spatial symmetry combined time-reversal symmetry *T* (primed), $R' = TR$,

$$\overleftrightarrow{\alpha}_{ij} = -(detR) R_{ik} R_{jl} \overleftrightarrow{\alpha}_{kl}$$

to account for

$$H \xrightarrow{T} -H,\, P \xrightarrow{T} P.$$

Therefore, for a magnetic point group of *m'mm*, we enforce invariance under each of three symmetry operations: m'_x, m_y, m_z, with $R_x = diag(-1,1,1)$, $R_y = diag(1,-1,1)$ and $R_z = diag(1,1,-1)$, respectively. Thus, the only nonzero components are α_{yz} and α_{zy}.

Therefore, its magnetoelectric susceptibility tensor $\overleftrightarrow{\alpha}$ has the form of:

$$\overleftrightarrow{\alpha} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & \alpha_{bc} \\ 0 & \alpha_{cb} & 0 \end{pmatrix}.$$

Note 2: Functional forms for ED SHG for point group $m'mm$, $22'2'$, $m'2'm$ and $m2m$

The *c*-type ED SHG nonlinear optical susceptibility tensors for $m'mm$, $22'2'$, and $m'2'm$ are the same. They are polar and of rank-3:

$$\chi^{ED,(c)}_{m'mm/22'2'/m'2'm}(2\omega) = \begin{pmatrix} \begin{pmatrix} \chi^{xxx} \\ 0 \\ 0 \end{pmatrix} & \begin{pmatrix} 0 \\ \chi^{xyy} \\ 0 \end{pmatrix} & \begin{pmatrix} 0 \\ 0 \\ \chi^{xzz} \end{pmatrix} \\ \begin{pmatrix} 0 \\ \chi^{yxy} \\ 0 \end{pmatrix} & \begin{pmatrix} \chi^{yxy} \\ 0 \\ 0 \end{pmatrix} & \begin{pmatrix} 0 \\ 0 \\ 0 \end{pmatrix} \\ \begin{pmatrix} 0 \\ 0 \\ \chi^{zxz} \end{pmatrix} & \begin{pmatrix} 0 \\ 0 \\ 0 \end{pmatrix} & \begin{pmatrix} \chi^{zxz} \\ 0 \\ 0 \end{pmatrix} \end{pmatrix}$$

In case of normal incidence, the resulted functional forms for RA-SHG in the parallel and the crossed channels are:

$$I^{ED,\ (c)}_{parallel}(2\omega) \propto ((\chi^{xyy} + 2\chi^{yxy})cos(\phi)^2 sin(\phi) + \chi^{xxx} sin(\phi)^3)^2$$

$$I^{ED,(c)}_{crossed}(2\omega) \propto (\chi^{xyy} cos(\phi)^3 + (\chi^{xxx} - 2\chi^{yxy})cos(\phi)sin(\phi)^2)^2$$

The $m2m$ supports *i*-type ED SHG radiation with the corresponding susceptibility tensor,

$$\chi^{ED,(i)}_{m2m}(2\omega) = \begin{pmatrix} \begin{pmatrix} 0 \\ \chi^{xxy} \\ 0 \end{pmatrix} & \begin{pmatrix} \chi^{xxy} \\ 0 \\ 0 \end{pmatrix} & \begin{pmatrix} 0 \\ 0 \\ 0 \end{pmatrix} \\ \begin{pmatrix} \chi^{yxx} \\ 0 \\ 0 \end{pmatrix} & \begin{pmatrix} 0 \\ \chi^{yyy} \\ 0 \end{pmatrix} & \begin{pmatrix} 0 \\ 0 \\ \chi^{yzz} \end{pmatrix} \\ \begin{pmatrix} 0 \\ 0 \\ 0 \end{pmatrix} & \begin{pmatrix} 0 \\ 0 \\ \chi^{zyz} \end{pmatrix} & \begin{pmatrix} 0 \\ \chi^{zyz} \\ 0 \end{pmatrix} \end{pmatrix}$$

Under normal incidence, the resulting functional forms for RA-SHG in the parallel and the crossed channels are:

$$I^{ED,(i)}_{parallel}(2\omega) \propto (\chi^{yyy} cos(\phi)^3 + (\chi^{yxx} + 2\chi^{xxy})cos(\phi)sin(\phi)^2)^2$$

$$I^{ED,(i)}_{crossed}(2\omega) \propto ((2\chi^{xxy} - 2\chi^{yyy})cos(\phi)^2 sin(\phi) - \chi^{yxx} sin(\phi)^3)^2$$

Note that the *c*-type and the *i*-type have the complementary nonzero tensor elements, corresponding to orthogonal polarization components rotated by 90°. Therefore, the interference between *c*-type and *i*-type contributions is straightforward, as their polarization components add vectorially in orthogonal bases:

$$I_{parallel}^{ED,interfered}(2\omega) \propto ((\chi^{xyy} + 2\chi^{yxy})cos(\phi)^2 sin(\phi) + \chi^{xxx} sin(\phi)^3 + \chi^{yyy} cos(\phi)^3 + (\chi^{yxx} + 2\chi^{xxy})cos(\phi)sin(\phi)^2)^2$$

$$I_{crossed}^{ED,interfered}(2\omega) \propto (\chi^{xyy} cos(\phi)^3 + (\chi^{xxx} - 2\chi^{yxy})cos(\phi)sin(\phi)^2 + (2\chi^{xxy} - 2\chi^{yyy})cos(\phi)^2 sin(\phi) - \chi^{yxx} sin(\phi)^3)^2$$

Note 3: Group theory analysis of spin terms

Consider the spins in the two layers: $\overrightarrow{S_1} = (S_{1x}, S_{1y}, S_{1z})$ and $\overrightarrow{S_2} = (S_{2x}, S_{2y}, S_{2z})$, where the number in the subscript denotes the top layer (1) and bottom layer (2), and the letter in the subscript denotes the corresponding component in the Cartesian coordinate. The linear combination of the linear spin terms and their corresponding irreducible representation (irrep) of the mmm are tabulated in Table S1. The table is built by constructing linear combinations of the spin terms and examine their transformations under each symmetry operations in the point group mmm to fill in the characters. By comparing the filled characters with the character table, we are able to determine the irrep, the basis and the corresponding magnetic point group of each linear combination. Each linear combination of the linear terms corresponds to a spin configuration shown in the table.

Table S1. ***Character table for linear spin combinations***

Linear spin	Irrep	Basis	Subgroup	m_x	m_y	m_z	2_x	2_y	2_z	i	Spin config
$S_{1x}+S_{2x}$	B_{3g}	R_x	$mm'm'$	1	-1	-1	1	-1	-1	1	FM along a
$S_{1y}+S_{2y}$	B_{2g}	R_x	$m'mm'$	-1	1	-1	-1	1	-1	1	FM along b
$S_{1z}+S_{2z}$	B_{1g}	R_x	$m'm'm$	-1	-1	1	-1	-1	1	1	FM along c
$S_{1x}-S_{2x}$	B_{2u}	y	$mm'm$	1	-1	1	-1	1	-1	-1	AFM along a
$S_{1y}-S_{2y}$	B_{3u}	x	$m'mm$	-1	1	1	1	-1	-1	-1	AFM along b
$S_{1z}-S_{2z}$	A_u	xyz	$m'm'm'$	-1	-1	-1	1	1	1	-1	AFM along c

Next, to identify all the possible bilinear terms, we considered the out productor of the two spins,

$$S_1 \otimes S_2 = \begin{pmatrix} S_{1x}S_{2x} & S_{1x}S_{2y} & S_{1x}S_{2z} \\ S_{1y}S_{2x} & S_{1y}S_{2y} & S_{1y}S_{2z} \\ S_{1z}S_{2x} & S_{1z}S_{2y} & S_{1z}S_{2z} \end{pmatrix}$$

By projecting the linear combinations of the bilinear terms into the irrep of mmm, we constructed Table S2.

Table S2. ***Character table for bilinear spin combinations***

Linear spin	Irrep	Basis	Subgroup	m_x	m_y	m_z	2_x	2_y	2_z	i
$S_{1y}S_{2z}+S_{1z}S_{2y}$	B_{3g}	R_x	$2/m$	1	-1	-1	1	-1	-1	1
$S_{1x}S_{2z}+S_{1z}S_{2x}$	B_{2g}	R_x	$2/m$	-1	1	-1	-1	1	-1	1
$S_{1x}S_{2y}+S_{1y}S_{2x}$	B_{1g}	R_x	$2/m$	-1	-1	1	-1	-1	1	1
$S_{1y}S_{2z}-S_{1z}S_{2y}$	B_{2u}	y	$m2m$	1	-1	1	-1	1	-1	-1
$S_{1x}S_{2z}-S_{1z}S_{2x}$	B_{3u}	x	$2mm$	-1	1	1	1	-1	-1	-1
$S_{1x}S_{2y}-S_{1y}S_{2x}$	A_u	xyz	222	-1	-1	-1	1	1	1	-1
$S_{1x}S_{2x}, S_{1y}S_{2y}, S_{1z}S_{2z}$	E	x^2, y^2, z^2	mmm	1	1	1	1	1	1	1

Note 4: Magnetic field dependence of all tensor elements for B//c case

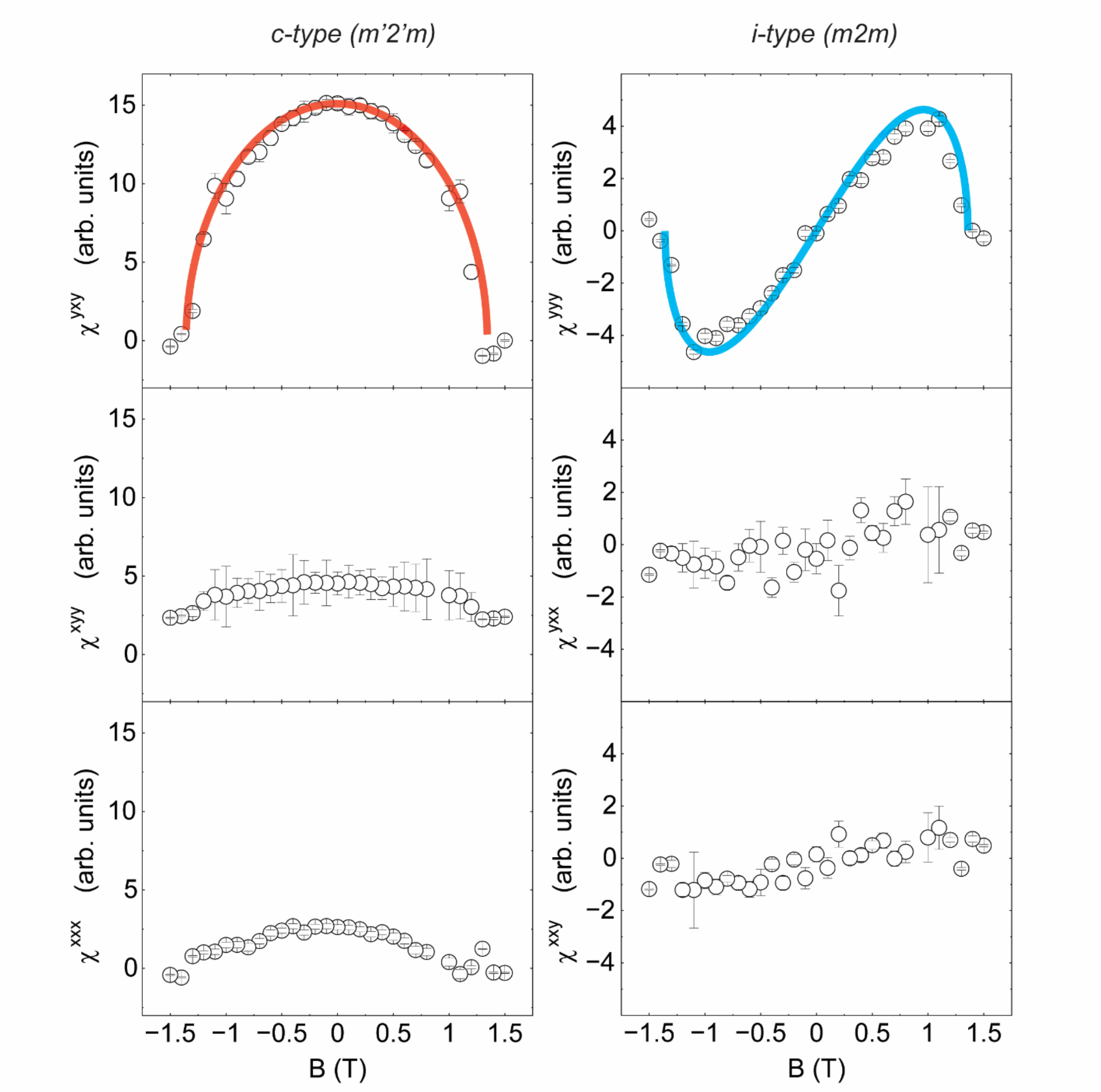

Fig. S1 Fitted tensor elements vs magnetic field. The left column are three tensor elements corresponding to the *c*-type SHG contribution and the right column are three tensor elements corresponding to the *i*-type SHG contribution.

Note 5: Fitting model for field-dependence of nonlinear susceptibility tensor elements

To comprehend these two trends, we build the following fitting model. By defining the angle between $\vec{S}_1$ and $\vec{S}_2$ to be θ, we have $\vec{M} = 2S\cos\frac{\theta}{2}\hat{c}$, $\vec{N} = 2S\sin\frac{\theta}{2}\hat{b}$, and $\vec{P} \propto S^2\sin\theta$, where S represents the magnetic moment magnitude. Knowing that the out-of-plane net magnetization scale linearly with the c-axis B field, i.e., $M = \gamma B$, and that the fully polarized FM magnetic state is achieved at B_c with a net magnetization of $2S$, i.e., $2S = \gamma B_c$, we arrive at $\cos\frac{\theta}{2} = \frac{B}{B_c}$, and thereby, $\vec{N} = 2S\sqrt{1-\left(\frac{B}{B_c}\right)^2}$ and $\vec{P} \propto \cos\left(\frac{\theta}{2}\right)\sin\left(\frac{\theta}{2}\right) = \frac{B}{B_c}\sqrt{1-\left(\frac{B}{B_c}\right)^2}$