Microscopic essence of the second-order nonlinear conductivity in $PT$-symmetric collinear antiferromagnetic metals with magnetic toroidal moment

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A magnetic toroidal moment is a fundamental electronic degree of freedom in the absence of both spatial inversion and time-reversal symmetries and gives rise to novel multiferroic and transport properties. We elucidate essential model parameters of the nonlinear transport in the space-time ($PT$) symmetric collinear antiferromagnetic metals accompanying a magnetic toroidal moment. By analyzing the longitudinal and transverse components of the second-order nonlinear conductivity on a two-dimensionally stacked zigzag chain based on the nonlinear Kubo formula, we show that an effective coupling between the magnetic toroidal moment and the antisymmetric spin-orbit interaction is a source of inducing the nonlinear conductivity. Moreover, we find that the nonreciprocal longitudinal current and nonlinear Hall coefficient are largely enhanced just below the transition temperature of the antiferromagnetic ordering. We also discuss the relevance to the linear magnetoelectric effect. Our result serves as a guide for extracting essential model parameters for various multiferroic and conductive phenomena in noncentrosymmetric antiferromagnetic metals.

Spontaneous time-reversal symmetry breaking has long been attracted much attention, as it leads to intriguing physical phenomena, such as the anomalous Hall effect and the magneto-optical Kerr effect. Modern understanding of these phenomena has been achieved based on the Berry phase mechanism [1, 2]. Although such phenomena were originally studied in the ferromagnetic state, it has recently been recognized that similar phenomena can occur in a certain class of antiferromagnetic (AFM) states without the uniform magnetization [3]. For example, the collinear AFM ordering with the mirror symmetry breaking as the uniform magnetization, results in the anomalous Hall effect [4–7]. Thus, the AFM materials can also exhibit the same physical properties as ordinary ferromagnetic ones, which is advantageous for functional materials without leakage of a magnetic field.

The AFM state also exhibits multiferroic phenomena when both spatial inversion ($P$) and time-reversal ($T$) symmetries are broken simultaneously while their product ($PT$) symmetry is preserved. The typical example is the linear magnetoelectric effect in the AFM insulators, e.g., Cr$_2$O$_3$ [3], Ga$_2$FeO$_4$ [8, 9], LiCoPO$_4$ [11, 12], and Ba$_2$CoGe$_2$O$_7$ [13], and in the AFM metals, e.g., UNi$_2$B$_2$C [14–16] and Ce$_3$TiB$_5$ [17, 18]. Moreover, the nonreciprocal optical and transport properties have been studied [19–22]. Among them, multiferroic phenomena within the linear response theory have been understood by regarding the fact that the AFM states accompany the uniform orderings of the odd-parity magnetic electronic multipoles [14, 22–24], such as the magnetic toroidal (MT) dipole [23, 30–35, 39].

Meanwhile, the microscopic understanding of the nonlinear transports beyond the symmetry argument has not been fully achieved [22, 34]. For example, it remains unclear which model parameters are essentially important to induce nonlinear transports and how the odd-parity magnetic multipoles are related to them. Extracting the essential model parameters in the model Hamiltonian constructed from the ab initio calculations is useful for a deep understanding of nonlinear transports, e.g., it enables us to explore functional AFM materials with a giant nonlinear transport and its efficient bottom-up design.

In this paper, we elucidate the microscopic essential model parameters for the second-order nonlinear conductivity in the $PT$-symmetric collinear AFMs by focusing on the role of the MT moment. By analyzing a minimal model on a two-dimensionally stacked zigzag chain based on the nonlinear Kubo formula, we show that the effective coupling between the MT moment and the antisymmetric spin-orbit interaction (ASOI) plays an essential role in inducing the longitudinal and transverse components of the nonlinear conductivity. Moreover, we find that the nonlinear conductivities are largely enhanced near the transition temperature in the case that the AFM molecular field is comparable to the ASOI. We also discuss the relevance between the transverse nonlinear conductivity and the linear magnetoelectric coefficient by comparing the ASOI and temperature dependences.

In order to capture the essence of the nonlinear conductivity in $PT$-symmetric collinear AFMs with the MT moment, we consider a minimal two-dimensional system where the zigzag chain along the $x$ direction [Fig. 1(a)] is stacked along the $z$ direction [Fig. 1(b)]. The tight-binding Hamiltonian is given by

\[
\mathcal{H} = \mathcal{H}_{\text{hop}}^{\text{AB}} + \mathcal{H}_{\text{hop}} + \mathcal{H}_{\text{ASOI}} + \mathcal{H}_{\text{int}},
\]  

\[
\mathcal{H}_{\text{hop}}^{\text{AB}} = \sum_{k} \sum_{\sigma \sigma'} \left\{ \epsilon^{\text{AB}}(k) c_{k \sigma}^\dagger c_{k \sigma'} + \text{H.c.} \right\},
\]

\[
\mathcal{H}_{\text{hop}} = \sum_{k} \sum_{\sigma} \left\{ \epsilon(k) c_{k \sigma}^\dagger c_{k \sigma} + c_{k \sigma}^\dagger c_{k \sigma} \right\},
\]

\[
\mathcal{H}_{\text{ASOI}} = \sum_{k} \sum_{\sigma \sigma'} g(k) \cdot \sigma' \sigma \left( c_{k \sigma}^\dagger c_{k \sigma} c_{k \sigma'} - c_{k \sigma} c_{k \sigma'} c_{k \sigma'} \right),
\]

\[
\mathcal{H}_{\text{int}} = J_{\text{AF}} \sum_{(ij)} \mathcal{M}_{iA} \mathcal{M}_{jB},
\]

where $c_{k \sigma}^\dagger$ ($c_{k \sigma}$) is the creation (annihilation) operator of electrons at wave vector $k$, sublattice $i = A, B$, and spin $\sigma = \uparrow, \downarrow$. The hopping Hamiltonian $\mathcal{H}_{\text{hop}}^{\text{AB}}$ in Eq. (2) includes the nearest-neighbor hopping between $A$ and $B$ sublattices as $\epsilon^{\text{AB}}(k) = -2t_{1} \cos(k, a/2)$, while $\mathcal{H}_{\text{hop}}$ includes the hoppings...
FIG. 1. (a), (b) Schematic pictures of (a) a two-sublattice zigzag chain and (b) its stacking along the z direction. (c) The temperature \(T\) dependence of the MT moment \(T_{\text{MF}}\) at \(\alpha_1 = 0.4\) and \(\alpha_2 = 0.1\). The AFM structure with the MT moment along the x direction \(T_x\) is shown in the inset. (d) The energy bands measured from the chemical potential \(\mu\) at \(k_z = 0\) for three temperatures.

within the same sublattices along the \(x\) and \(z\) directions as 
\[ e(k) = -2t_z \cos(k_xa) - 2t_1 \cos(k_ca). \] 
\(H_{\text{SOI}}\) in Eq. (4) represents the ASOI that arises from the relativistic spin-orbit coupling as 
\[ g(k) = \{-\alpha_2 \sin(k_ca), 0, \alpha_1 \sin(k_ca)\}. \] 
The ASOI in Eq. (4) has the sublattice-dependent staggered form satisfying 
\[ g(k) = \{0, 0, -\alpha_2 \sin(k_ca), 0, \alpha_1 \sin(k_ca)\}. \]

The model parameters as \((t_1, t_2, t_3, J_{AF}) = (0.1, 1, 0.5, 2.5)\), electron filling as 1/5, and the lattice constant as \(a = c = 1\) in the following discussion; \(t_2\) is set as the energy unit.

The model in Eq. (1) exhibits the MT moment when the global inversion symmetry is broken under the staggered AFM ordering, as shown in the inset of Fig. (1c) [14, 23]. In the present system, the staggered AFM moment along the \(z\) direction is equivalent to the uniform MT moment along the \(x\) direction; 
\[ T_{\text{MF}}^x \equiv \langle (M_x^A + M_x^B)/2 \rangle. \]

The T dependence of \(T_{\text{MF}}^x\) at \(\alpha_1 = 0.4\) and \(\alpha_2 = 0.1\) is shown in Fig. (1c), where \(T_{\text{MF}}^x\) is self-consistently determined for the two-sublattice unit cell by taking over 200\(^2\) grid points in the Brillouin zone. \(T_{\text{MF}}^x\) becomes nonzero below the transition temperature \(T_N\) and saturates below \(T \approx 0.2T_N\). Almost the same behavior is obtained for \(\alpha_1, \alpha_2 \lesssim 0.5\). Reflecting \(T_{\text{MF}}^x \neq 0\), the electronic band structure is asymmetrically modulated along the \(k_z\) direction, as shown in Fig. (1d) [23, 41]. This asymmetric band modulation is understood from the effective coupling between \(T_{\text{MF}}^x\) and the ASOI \(\alpha_1\) in the doubly degenerate bands due to the PT symmetry, i.e., 
\[ e_x(k) = \pm X(k) \] 
with 
\[ X(k) = \sqrt{\left(\alpha_1 s_x - T_{\text{MF}}^x\right)^2 + \alpha_2^2 s_z^2 + 4t_1^2 s_x^2 s_z^2} \] 
where \(s_x = \sin k_x\), \(s_z = \sin k_z\), \(c_1/c_2 = \cos k_x/\sqrt{2}\), and \(T_{\text{MF}}^x = J_{AF}T_{\text{MF}}^x\). The factor \(\left(\alpha_1 s_x - T_{\text{MF}}^x\right)^2\) includes the coupling between \(T_{\text{MF}}^x\) and \(\alpha_1\) with the odd function of \(k_x\). This asymmetric band modulation becomes a source of the nonlinear transport.

For \(T_{\text{MF}}^x \neq 0\), the nonzero components of the second-order nonlinear conductivity \(\sigma_{\nu \mu \lambda}\) in Eq. (6) represents the Drude-type one with the dissipation \(\tau^{-1}\), whose expression eventually coincides with that obtained by the Boltzmann formalism [22, 42, 43]. From Eq. (6), we find the relation \(\sigma_{xx} = \sigma_{zzz}\) by integration by parts. Hence, the present system has two independent components: the longitudinal \(\sigma_{xxx}\) and transverse \(\sigma_{zzz}\). Hereafter, we use the scaled \(\bar{\sigma}_{\nu \mu \lambda} = \sigma_{\nu \mu \lambda}/(e^2 c^2 \hbar^3)\).

We first discuss the longitudinal nonlinear conductivity \(\sigma_{xxx}\). Figure (2a) shows \(\bar{\sigma}_{xxx}\) as a function of \(T\) for various \(\alpha_1 = 0.1\)–0.5 at \(\alpha_2 = 0.1\). The \(T\) dependence for different \(\alpha_1\) is qualitatively similar; \(\bar{\sigma}_{xxx}\) is largely enhanced just below \(T = T_N\), and shows maximum with decrease of \(T\). While further decreasing \(T\), \(\bar{\sigma}_{xxx}\) shows the sign change, and then reaches a negative value at the lowest \(T\).

The nonzero \(\sigma_{xxx}\) is closely related to the formation of the asymmetric band structure under \(T_{\text{MF}}^x \neq 0\), since \(\sigma_{xxx}\) has the same symmetry as \(T_{\text{MF}}^x\). As the asymmetric band modulation is caused by the coupling between \(T_{\text{MF}}^x\) and \(\alpha_1\), they are indispensable for nonzero \(\sigma_{xxx}\). Indeed, by performing the method to extract the essential model parameters for \(\sigma_{xxx}\) [44], we find that \(\sigma_{xxx}\) has the form of \(\alpha_1 T_{\text{MF}}^x F(t_1, t_2, t_3, \alpha_1, \alpha_2, T_{\text{MF}}^x)\), where \(F(t_1, t_2, t_3, \alpha_1, \alpha_2, T_{\text{MF}}^x)\) is an arbitrary function [45]. This analytical approach is consistent with the numerical result in the inset of Fig. (2a), where \(\bar{\sigma}_{xxx}\) vanishes for \(\alpha_1 = 0\), and \(\bar{\sigma}_{xxx}\) is well scaled by \(\bar{\sigma}_{xxx}/\alpha_1\) at low temperatures \(T \lesssim 0.7T_N\).

Meanwhile, \(\bar{\sigma}_{xxx}\) is not scaled by \(\alpha_1\) for \(0.7 \leq T/T_N \leq 1\) in the region where \(\bar{\sigma}_{xxx}\) is drastically enhanced. This is attributed to the rapid increase of \(T_{\text{MF}}^x\) and resultant drastic change of the electronic band structure near the Fermi level.
which affects the two factors $\frac{\partial^2 \epsilon_x(k)}{\partial k^2}$ and $\frac{\partial \epsilon_x(k)}{\partial k}$ in Eq. (6); the small $X(k)$ gives a dominant contribution. Thus, for small constant values of $t_1 = 0.1$ and $\alpha_2 = 0.1$, the enhancement of the $\tilde{\sigma}_{xxx}$ is maximized at the minimum of $X(k)$, i.e., $T_{\text{MF}}^x = \alpha_1 \sin k_F^x$ for the Fermi wave number $k_F^x$, as shown in Fig. 2(a). In short, there are two conditions for large $\tilde{\sigma}_{xxx}$: One is the large essential parameters, such as $\alpha_1$, $T_{\text{MF}}^x$, and $J_{\text{xx}}$, and the other is to satisfy $T_{\text{MF}}^x \approx \alpha_1 \sin k_F^x$. These conditions can be experimentally controlled by electron/hole doping and temperature.

The sign change of $\tilde{\sigma}_{xxx}$ in $T$ dependence is owing to the multiband effect. As shown in Fig. 1(d), the band bottom is shifted in the opposite direction for the upper and lower bands, which means that the opposite sign of $\alpha_1 T_{\text{MF}}^x$ results in the opposite contribution to $\tilde{\sigma}_{xxx}$. This is demonstrated by decomposing $\tilde{\sigma}_{xxx}$ into the upper- and lower-band contributions, as shown in Fig. 2(b). The results indicate that the dominant contribution of $\tilde{\sigma}_{xxx}$ arises from the upper band for $0.9 \leq T/T_N \leq 1$, while that arises from the lower band for $T/T_N \leq 0.9$. The suppression of the upper-band contribution for low $T$ is because it becomes away from the Fermi level by the development of $T_{\text{MF}}^x$.

Next, let us discuss the transverse nonlinear conductivity $\tilde{\sigma}_{xzz}$. Figure 3(a) shows the $T$ dependence of $\tilde{\sigma}_{xzz}$ for $0.02 \leq \alpha_1, \alpha_2 \leq 0.1$ with $\alpha_1 = \alpha_2$. The behavior of $\tilde{\sigma}_{xzz}$ against $T$ is similar to $\tilde{\sigma}_{xxx}$ except for the sign change: $\tilde{\sigma}_{xzz}$ becomes nonzero below $T = T_N$ and shows the maximum near $T_N$. While decreasing $T$, $\tilde{\sigma}_{xzz}$ is suppressed and shows an almost constant value.

Similar to $\tilde{\sigma}_{xxx}$, the origin of nonzero $\sigma_{xzz}$ is the asymmetric band modulation under $T_{\text{MF}}^x \neq 0$ via the effective coupling $T_{\text{MF}}^x \alpha_1$. Besides, we find another contribution from $\alpha_2$ for nonzero $\sigma_{xzz}$ in contrast to $\sigma_{xxx}$. This is owing to an additional symmetry between $k_x$ and $k_x + \pi$ for $\alpha_2 = 0$, which gives the opposite-sign contribution to $\sigma_{xzz}$ so that totally $\sigma_{xzz} = 0$. By performing a similar analysis to $\sigma_{xxx}$, we obtain the essential model parameters of $\sigma_{xzz}$ as $\alpha_1 \alpha_2^2 T_{\text{MF}}^y F(t_1, t_2, t_3, \alpha_1, \alpha_2, T_{\text{MF}}^x)$ [45]. Indeed, $\sigma_{xzz}$ is well scaled by $\alpha_1 \alpha_2^2$ as shown in the inset of Fig. 3(a).

It is noteworthy to comment on the relation between the transverse nonlinear conductivity and other physical quantities. From the symmetry aspect, the second-order transverse conductivity can be related to the combination of the linear Hall and magnetoelectric coefficients [14]. For example, nonzero $\sigma_{xzz}$ consisting of the linear magnetoelectric coefficient $\alpha_1$ and the Hall coefficient $\sigma_{xzz}$ under an external magnetic field $H_x$ is expected when $\sigma_{xzz}$ is finite. Here, we compare these two quantities at the microscopic level, where

FIG. 2. (a) The longitudinal second-order conductivity $\tilde{\sigma}_{xxx}$ for $\alpha_1 = 0.1$–0.5 as a function of $T$ at $\alpha_2 = 0.1$. The inset shows $\tilde{\sigma}_{xxx}/\alpha_1$. (b) The upper- and lower-band contributions to $\tilde{\sigma}_{xxx}$ at $\alpha_1 = 0.4$.

FIG. 3. (a) The transverse second-order nonlinear conductivity $\tilde{\sigma}_{xzz}$ for several $\alpha_1$ and $\alpha_2$ with $\alpha_1 = \alpha_2$. (b) The quantity $\tilde{\sigma}_{xzz}(\alpha_1, \alpha_2)$ with the same parameters as (a). $\tilde{\sigma}_{xzz}$ is calculated by supposing the magnetic field $H_x = 0.01$. The insets of (a) and (b) represent $\tilde{\sigma}_{xxx}(\alpha_1, \alpha_2^2)$ and $\tilde{\sigma}_{xzz}(\alpha_1, \alpha_2^2)$, respectively.
\( \sigma_{zz} \) and \( \sigma_{xz} \) are calculated by the linear response theory:

\[
\alpha_{zz} = \frac{e^2 \mu_B \hbar}{2V_i} \sum_k \sum_{m,n} \frac{f[\varepsilon_{y}(k)] - f[\varepsilon_{m}(k)]}{[\varepsilon_{y}(k) - \varepsilon_{m}(k)]^2 + (\hbar \omega)^2} \epsilon_{ymn} e^{ymn} \langle k | \sigma_{y} \rangle, \tag{7}
\]

\[
\alpha_{xz} = \frac{e^2 \hbar}{V_i} \sum_k \sum_{m,n} \frac{f[\varepsilon_{y}(k)] - f[\varepsilon_{m}(k)]}{[\varepsilon_{y}(k) - \varepsilon_{m}(k)]^2 + (\hbar \omega)^2} \epsilon_{ymn} e^{ymn} \langle k | \sigma_{x} \rangle. \tag{8}
\]

In Eq. (7), \( g \) and \( \mu_B \) are the g factor (\( g = 2 \)) and Bohr magneton, respectively. \( \sigma_{ymn} = (\langle m | \sigma_{y} | n \rangle) \) and \( e^{ymn} \langle k | \sigma_{y} \rangle \) are the matrix elements of spin \( \sigma_y \) and velocity \( v_{yk} = \partial \mathcal{H} / (\hbar \partial k_y) \) for the eigenstate \( | n \rangle \). The inter-band process is important in both tensors. We use the scaled \( \tilde{\alpha}_{zz} = \alpha_{zz} / (e^2 \mu_B \hbar) \) and \( \tilde{\sigma}_{zz} = \sigma_{zz} / (e^2 \hbar H_s) \).

We show the \( T \) dependence of \( \tilde{\sigma}_{zz} \tilde{\alpha}_{zz} \) in Fig. 3(b) for the same parameters in Fig. 3(a). The small magnetic field \( H_s = 0.01 \) is introduced to mimic the induced magnetization in \( \alpha_{zz} \). Compared to the results in Fig. 3(a), one finds the resemblance between the \( T \) dependences of \( \tilde{\sigma}_{zz} \) and \( \tilde{\sigma}_{xz} \tilde{\alpha}_{zz} \), both of which are scaled by \( \alpha_{zz} / \alpha_{zz} \). This result indicates a good qualitative correspondence between them except for the overall scales. Their similar qualitative behavior is explained by the dependence of the essential model parameters in \( \alpha_{zz} \) and \( \sigma_{zz} \); \( \alpha_{zz} \propto \alpha_2 T_s H_s^2 \) and \( \sigma_{zz} \propto \alpha_1 \alpha_2 H_s \). The quantitative difference \( \sigma_{zz} \alpha_{zz} \sim 10^{-2} \) may be ascribed to the magnitude of an internal magnetic field induced by the magnetoelectric effect, that should be used instead of \( H_s \) in \( \sigma_{zz} \). However, it is hard to estimate it in the nonlinear conductivity \( \sigma_{zz} \).

The above results clearly depend on the fact that the essential model parameters are common in \( \sigma_{zz} \) and \( \sigma_{zz} \alpha_{zz} \) more than the symmetry. However, such a correspondence does not always hold by introducing the other model parameters that are not considered here. For example, the interlayer A-B hopping gives the different parameter dependences in \( \sigma_{zz} \) and \( \sigma_{zz} \alpha_{zz} \) obtained by the analytical method [45]. In such a situation, they do not have a simple correspondence.

Finally, we discuss the order estimate of the nonlinear conductivity for \( \alpha_1 = 0.5 \) and \( \alpha_2 = 0.1 \) by the ratio \( \sigma_{zz} / (\sigma_{zz} \alpha_{zz})^2 \) with being independent of the relaxation time in the clean limit. By putting the typical values as \( a \sim 0.5 \) [nm] and \( |z| = 0.2 \) eV, we obtain \( \sigma_{zz} / (\sigma_{zz} \alpha_{zz})^2 \sim 10^{-4} \hbar^2 \alpha^2 |z|^2 \sim 10^{-18} \) [m³ A⁻¹] for \( T \to 0 \) and \( 10^{-17} \) [m³ A⁻¹] near \( T_s \), which is comparable to the value in the 2D nonmagnetic Rashba system under the magnetic field [42]. Further enhancement can be achieved by tuning the model parameters and electron filling [45].

In summary, we investigated the microscopic conditions regarding the essential parameters for the second-order nonlinear conductivity in the \( PT \)-symmetric collinear AFM with the MT moment. Based on the nonlinear Kubo formula in the clean limit, we found that the effective coupling between the ASOI and the MT moment is essential for the nonlinear conductivity. By analyzing both the longitudinal and transverse components of the nonlinear conductivity while changing the ASOI and the temperature, we showed that their large enhancement can be achieved near the transition temperature, provided that the AFM molecular field is comparable to the ASOI. We also showed that the physical phenomena characterized by the same essential model parameters exhibit a similar temperature dependence by analyzing the linear magnetoelectric and Hall coefficients.

The present result elucidates the essential model parameters for MT-related physical phenomena, such as the nonlinear conductivity and the linear magnetoelectric effect, in \( PT \)-symmetric collinear AFMs. The similar analysis to extract essential model parameters can also be applied to any collinear AFMs with the MT moment in the zigzag structure, e.g., CeRu₂Al₁₀ [46, 47], Ce₃TiBi₅ [17, 18], and \( \alpha \)-YbAl₁₋₃Mn₄ [48], and other locally noncentrosymmetric crystal structures, such as Mn₀.₇Au [49], RB₄ ( \( R \) = Dy, Er) [50, 51], CuMnAs citewadley2015antiferromagnetic,wang2021intrinsinc, PrMnSbO [52], NdMnAsO [53], and \( \chi_{Fe₂₋₃Se₂} \) ( \( \chi = K, Tl, Rb \) [54, 56]), once the model Hamiltonian is constructed. The measurements of the linear magnetoelectric effect and the nonlinear conductivity for these materials are also useful for the essential model parameters. Moreover, the analysis is straightforwardly extended to the AFMs with the other odd-parity magnetic multipole moments, such as the magnetic quadrupole, since they are characterized by the same spatial inversion and time-reversal symmetries. Our study will stimulate a further investigation of the multiferroic and conductive phenomena in the \( PT \)-symmetric AFM metals.

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I. ESSENTIAL MODEL PARAMETERS IN RESPONSE TENSORS

In this section, we show the essential model parameters for the asymmetric band modulation, the longitudinal and transverse nonlinear conductivities, and the linear Hall and magnetoelectric coefficients, by using the systematic analysis method in Refs. [1] and [2]. The results are summarized in Table S1.

### Table S1. Essential model parameters for the asymmetric band modulation and response tensors indicated by the checkmark (✓). The last column represents the overall factor of the contributions to the asymmetric band modulation and response tensors.

| Parameter                                      | $t_2$ | $t_3$ | $\alpha_1$ | $\alpha_2$ | $\tilde{T}^{MF}_{xx}$ | $H_x$ | Overall factor |
|------------------------------------------------|-------|-------|-------------|-------------|------------------------|-------|---------------|
| Asymmetric band modulation                     | ✓     | ✓     | ✓           | ✓           | $\alpha_1 T^{MF}_{xx}$ |
| 2nd-order longitudinal conductivity, $\sigma_{xxx}$ | ✓     | ✓     | ✓           | ✓           | $\alpha_1 T^{MF}_{xx}$ |
| 2nd-order transverse conductivity, $\sigma_{xc}$ ($t_y = 0$) | ✓     | ✓     | ✓           | ✓           | $\alpha_1 T^{MF}_{xx}$ |
| $\sigma_{xc}$ ($t_y \neq 0$)                      | ✓     | ✓     | ✓           | ✓           | $\alpha_1 T^{MF}_{xx}$ |
| Linear magnetoelectric coefficient, $\alpha_{xc}$ ($t_y = 0$) | ✓     | ✓     | ✓           | ✓           | $\alpha_1 T^{MF}_{xx}$ |
| $\alpha_{xc}$ ($t_y \neq 0$)                      | ✓     | ✓     | ✓           | ✓           | $\alpha_1 T^{MF}_{xx}$ |
| Linear Hall coefficient, $\sigma_{xc}$ ($t_y = 0$) | ✓     | ✓     | ✓           | ✓           | $\alpha_1 T^{MF}_{xx}$ |
| $\sigma_{xc}$ ($t_y \neq 0$)                      | ✓     | ✓     | ✓           | ✓           | $\alpha_1 T^{MF}_{xx}$ |

A. Asymmetric band modulation

First, we give the essential model parameters for the asymmetric band modulation. Following the method for extracting the essential model parameters in the thermal average of an hermitian operator [1][2], we obtain the momentum distribution of the band modulation and its parameter dependences by analytically evaluating the low-order contributions of the following quantity,

$$\Omega(k) = \text{Tr} \left[ h^{i+1}(k) \right].$$  \hspace{1cm} (S1)

Here $h^{i+1}(k)$ denotes the $(i+1)$-th power of the Hamiltonian matrix at wave vector $k$. The 0th- and 1st-order contributions $\Omega^0(k)$ and $\Omega^1(k)$ are explicitly given by

$$\Omega^0(k) = -8 \left( t_2 \cos k_x + t_3 \cos k_z \right),$$  \hspace{1cm} (S2)

$$\Omega^1(k) = -8 \alpha_1 \tilde{T}^{MF}_{xx} \sin k_x + 4 \left[ \left( \tilde{T}^{MF}_{xx} \right)^2 + \alpha_1^2 \sin^2 k_x + \alpha_2^2 \sin^2 k_z + 2\alpha_2 \left( 1 + \cos k_x \right) + 4 \left( t_2 \cos k_x + t_3 \cos k_z \right) \right].$$  \hspace{1cm} (S3)

The odd function of $k_z$ appears only in the first term of Eq. (S3) in the form proportional to $\alpha_1 \tilde{T}^{MF}_{xx}$, which means that the asymmetric band structure is induced by the coupling between the nonzero $\tilde{T}^{MF}_{xx}$ and $\alpha_1$. It is confirmed at least to the 6th order. Note that the odd functions of $k_z$ included in the higher-order terms in Eq. (S1) are always proportional to $\alpha_1 \tilde{T}^{MF}_{xx}$. Thus, both $\alpha_1$ and $\tilde{T}^{MF}_{xx}$ are the essential model parameters for the asymmetric band structure, and their coupling is also crucial for nonlinear conductivities.
B. Second-order nonlinear conductivity

Next, we elucidate the essential model parameters in the longitudinal and transverse nonlinear conductivities. The essential model parameters in the Drude-type nonlinear conductivities can be extracted by evaluating the following quantity [2],

$$\text{Re} \left[ \Gamma^{ij}_{\mu\nu} \right] = \sum_k \text{Re} \left[ \text{Tr} \left[ \hat{\delta}_{ijk} h'(k) \hat{v}_{ik} h'(k) \hat{\delta}_{ijk} h'(k) \right] \right],$$

(S4)

where $\hat{\delta}_{ijk}$ denotes the $\mu$ component of the velocity operator at $k$.

FIG. S1. (a) Schematic picture of the interlayer hopping $t_4$ between A and B sublattices. (b), (c) The $T$ dependence of (b) $\tilde{\sigma}_{xxxx}$ and (c) $\tilde{\sigma}_{xx}\tilde{\sigma}_{zz}$ for $(t_4, \alpha_2) = (0.1, 0), (0.1, 0.1)$, and $(0.05, 0.1)$.

Here, we introduce the interlayer hopping between the sublattice A and B [Fig. S1(a)]. The effect of the additional hopping is taken into account by replacing $e^{\alpha B}(k)$ as $-2t_1 \cos (k, a/2) \to -2[t_1 + 2t_4 \cos (k, a/2)] \cos (k, a/2)$. The results of the evaluations are given as follows.

- Longitudinal nonlinear conductivity $\sigma_{xxx}$

As the essential model parameters are included in any pairs of $(i, j, k)$ in Eq. (S4), we here show two low-order contributions to Eq. (S4) in the $(i, j, k) = (0, 0, 1)$ and $(0, 1, 3)$ terms as representative examples, which are explicitly given by

$$\text{Re} \left[ \Gamma^{001}_{xxx} \right] = \alpha_1 \tilde{T}_x \left( t_1^2 + 2t_4^2 \right),$$

(S5)

$$\text{Re} \left[ \Gamma^{013}_{xxx} \right] = 4\alpha_1 \tilde{T}_x \left[ t_2 \left\{ \alpha_2^2 \alpha_3^2 + t_1^2 \left( 4 \left( \tilde{T}_x^{MF} \right)^2 + 7\alpha_1^2 + 2\alpha_2^2 + 3\alpha_3^2 \right) \right\} + t_4 \left\{ -4 \left( \tilde{T}_x^{MF} \right)^2 t_1 t_3 + 5\alpha_1^2 t_1 t_3 - \alpha_2^2 t_1 t_3 - 16t_1^2 t_3 - 12t_1 t_2 t_3 - 12t_1^3 + 8 \left( \tilde{T}_x^{MF} \right)^2 t_2 t_4 + 14\alpha_1^2 t_2 t_4 + 2\alpha_2^2 t_2 t_4 + 36\alpha_1^2 t_2 t_4 - 48t_1 t_3^2 + 18t_3^2 \right\} \right].$$

(S6)

Then, the essential model parameters in the longitudinal nonlinear conductivity $\sigma_{xxx}$ are $\alpha_1$ and $\tilde{T}_x^{MF}$, which is consistent with the fact that the nonzero $\sigma_{xxx}$ is closely related to the asymmetric band structure under $T_x^{MF} \neq 0$. Since all the terms in Eq. (S4) are always proportional to $\alpha_1 \tilde{T}_x^{MF}$, $\sigma_{xxx}$ is written in the form:

$$\sigma_{xxx} = \alpha_1 \tilde{T}_x^{MF} \left[ t_2^2 F(t_1, t_2, t_3, t_4, \alpha_1, \alpha_2, \tilde{T}_x^{MF}) + t_4 F'(t_1, t_2, t_3, t_4, \alpha_1, \alpha_2, \tilde{T}_x^{MF}) + t_4 F''(t_1, t_2, t_3, t_4, \alpha_1, \alpha_2, \tilde{T}_x^{MF}) \right].$$

(S7)

By introducing $t_4 \neq 0$, the additional contribution appears, which results in the change of $\sigma_{xxx}$.

- Transverse nonlinear conductivity $\sigma_{xxz}$

Similar to $\sigma_{xxx}$, we show two low-order contributions to Eq. (S4) in the $(i, j, k) = (0, 1, 0)$ and $(0, 1, 1)$ terms for example. The expressions are given by

$$\text{Re} \left[ \Gamma^{010}_{xxz} \right] = -\frac{242}{25} \alpha_1 \tilde{T}_x^{MF} t_4^2,$$

(S8)

$$\text{Re} \left[ \Gamma^{011}_{xxz} \right] = \frac{121}{25} \alpha_1 \tilde{T}_x^{MF} \left[ \alpha_2^2 t_2 + t_4 \left( 4t_1 t_3 + 8t_2 t_4 \right) \right].$$

(S9)
Similar to this result, we find that all the terms in Eq. (S4) are always proportional to $\alpha_1 \tilde{T}_x^{MF}$, then $\sigma_{xzc}$ is expressed as
\[ \sigma_{xzc} = \alpha_1 \tilde{T}_x^{MF} \left[ a_2^2 t_2 F(t_1, t_2, t_3, t_4, \alpha_1, \alpha_2, \tilde{T}_x^{MF}) + t_4 F'(t_1, t_2, t_3, t_4, \alpha_1, \alpha_2, \tilde{T}_x^{MF}) \right], \quad (S10) \]
where the second term proportional to $t_4$ does not vanish even for $\alpha_2 = 0$.

C. Linear responses

We further clarify the essential model parameters for the linear Hall and magnetoelectric conductivities. The essential model parameters in the inter-band contribution of the electric-field induced response tensors can be extracted by evaluating the following quantity $[2]$,\n\[ \text{Im} \left[ \Gamma_{ij}^{0} \right] = \sum_k \text{Im} \left[ \text{Tr} \left[ \hat{A}_{ijk} h'(k) \tilde{\psi}_{ik} h'(k) \right] \right], \quad (S11) \]
where $\hat{A}_{ijk}$ denotes the $\mu$ component of an arbitrary hermitian operator at $k$.

- Magnetoelectric coefficient $\alpha_{xz}$
  The magnetoelectric coefficient $\alpha_{xc}$ corresponds to the case with $\hat{A}_{ijk} = \sigma_x$ in Eq. (S11). Similar to the nonlinear conductivities, the essential model parameters are included in any pairs of $(i, j)$ in Eq. (S11). We show two cases by taking $(i, j) = (0, 2)$ and $(1, 3)$, which are given by
\[ \text{Im} \left[ \Gamma_{12}^{0} \right] = -\frac{44}{5} \alpha_2 \tilde{T}_x^{MF} t_3, \quad (S12) \]
\[ \text{Im} \left[ \Gamma_{13}^{0} \right] = \frac{11}{5} \alpha_2 \tilde{T}_x^{MF} \left\{ t_3 \left[ 4 \left( \tilde{T}_x^{MF} \right)^2 + 6\alpha_1^2 + \alpha_2^2 + 8t_1^2 - 24t_2^2 - 12t_2 \right] + t_4 \left( 16t_4 t_2 + 24t_4 t_4 \right) \right\}. \quad (S13) \]
We also find that all the terms in Eq. (S11) are always proportional to $\alpha_2 \tilde{T}_x^{MF}$, then $\alpha_{xc}$ is expressed as
\[ \alpha_{xc} = \alpha_2 \tilde{T}_x^{MF} \left[ t_3 F(t_1, t_2, t_3, t_4, \alpha_1, \alpha_2, \tilde{T}_x^{MF}) + t_4 F'(t_1, t_2, t_3, t_4, \alpha_1, \alpha_2, \tilde{T}_x^{MF}) \right]. \quad (S14) \]

Therefore, the essential model parameters are $\alpha_2$, $\tilde{T}_x^{MF}$ and $t_3$ or $t_4$.

- Hall coefficient $\sigma_{xc}$
  In order to discuss $\sigma_{xc}$, we introduce the small magnetic field along the $y$ direction $H_y$. Then, we evaluate the essential model parameters for the Hall coefficient $\sigma_{xc}$ with $\hat{A}_{ijk} = \tilde{\psi}_{ij}$ in Eq. (S11). We show two low-order contributions to Eq. (S11) in the $(i, j) = (0, 3)$ and $(1, 3)$ terms for example, which are given by
\[ \text{Im} \left[ \Gamma_{03}^{0} \right] = \frac{44}{5} \alpha_1 \alpha_2 H_y \left( 3 t_2 t_3 + 5 t_4 t_4 \right), \quad (S15) \]
\[ \text{Im} \left[ \Gamma_{13}^{0} \right] = \frac{88}{5} \alpha_1 \alpha_2 H_y \left[ 2 t_2 t_3 + t_3 \left( 8 t_4 t_2 + 7 t_3 t_4 \right) \right]. \quad (S16) \]
All the terms in Eq. (S11) are always proportional to $\alpha_1 \alpha_2 H_y$, then $\sigma_{xc}$ is expressed as
\[ \sigma_{xc} = \alpha_1 \alpha_2 H_y \left[ t_3 F(t_1, t_2, t_3, t_4, \alpha_1, \alpha_2, \tilde{T}_x^{MF}) + t_4 F'(t_1, t_2, t_3, t_4, \alpha_1, \alpha_2, \tilde{T}_x^{MF}) \right]. \quad (S17) \]

Therefore, the essential model parameters are $\alpha_1$, $\alpha_2$, $H_y$ and $t_3$ or $t_4$.

By combining the results, Eqs. (S14) and (S17), $\sigma_{xc}$ has the form:
\[ \sigma_{xc} = \alpha_1 \alpha_2^2 \tilde{T}_x^{MF} H_y \left[ t_3 F(t_1, t_2, t_3, t_4, \alpha_1, \alpha_2, \tilde{T}_x^{MF}) + t_4 F'(t_1, t_2, t_3, t_4, \alpha_1, \alpha_2, \tilde{T}_x^{MF}) + t_4 F''(t_1, t_2, t_3, t_4, \alpha_1, \alpha_2, \tilde{T}_x^{MF}) \right], \quad (S18) \]
which clearly shows that $\sigma_{xc} \propto \alpha_1 \alpha_2^2 \tilde{T}_x^{MF} H_y$, irrespective of the additional parameter of $t_4$.

When $t_4 = 0$, we find that both $\sigma_{xzc}$ and $\sigma_{xc} \alpha_{yz}$ are proportional to $\alpha_1 \alpha_2^2 \tilde{T}_x^{MF}$. On the other hand, such relation does not hold when $t_4 \neq 0$, $\sigma_{xzc} \propto \alpha_1 \tilde{T}_x^{MF}$, whereas $\sigma_{xc} \alpha_{yz} \propto \alpha_1 \alpha_2^2 \tilde{T}_x^{MF}$.
II. EFFECT OF ADDITIONAL INTERLAYER HOPPING

We compare the transverse component of the nonlinear conductivity $\sigma_{xzz}$ and the quantity $\sigma_{xz}\alpha_{yz}$ in the presence of the interlayer hopping $t_4$ between the sublattice A and B.

Figures S1(b) and S1(c) show $\bar{\sigma}_{xzz}$ and $\bar{\sigma}_{xz}\bar{\alpha}_{yz}$ as functions of $T$, respectively, for $t_4 = 0.1, 0.05$ and $\alpha_2 = 0, 0.1$, where $\alpha_1 = 0.4$ is used. As shown by the red dashed line in Fig. S1(b), $\bar{\sigma}_{xzz}$ still remains nonzero even for $\alpha_2 = 0$, while $\bar{\sigma}_{xz}\bar{\alpha}_{yz}$ in Fig. S1(c) vanishes. Furthermore, the nonzero $t_4$ enhances $\bar{\sigma}_{xzz}$, while it suppresses $\bar{\sigma}_{xz}\bar{\alpha}_{yz}$ while increasing $t_4$. This is because the essential model parameters discussed in the previous section are different for $\sigma_{xzz}$ and $\sigma_{xz}\alpha_{yz}$. Indeed, in the presence of $t_4$ and $\alpha_2$, the essential model parameter of $\sigma_{xzz}$ is represented as $\alpha_1 T_4^{MF}[a_2 t_4 F(t_1, t_2, t_3, t_4, \alpha_1, \alpha_2, T_4^{MF}) + t_4 F'(t_1, t_2, t_3, t_4, \alpha_1, \alpha_2, T_4^{MF})]$, which clearly shows that $\sigma_{xzz}$ has the additional contribution from $t_4$ and does not vanish for $\alpha_2 = 0$. On the other hand, the essential model parameters of $\sigma_{xz}$ and $\alpha_{yz}$ does not show the change from $\sigma_{xz} \rightarrow \alpha_1 \alpha_2 H_y F(t_1, t_2, t_3, t_4, \alpha_1, \alpha_2, T_4^{MF}, H_y)$ and $\sigma_{yz} \rightarrow \alpha_2 T_4^{MF} F(t_1, t_2, t_3, t_4, \alpha_1, \alpha_2, T_4^{MF})$; respectively; the hopping $t_4$ is not the essential model parameter for $\sigma_{xz}$ and $\alpha_{yz}$. Thus, there is no simple relation between them. The result of the essential model parameters for $t_4 \neq 0$ is summarized in Table S1.

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