Electrically and magnetically switchable nonlinear photocurrent in \( \mathcal{PT} \)-symmetric magnetic topological quantum materials

Hua Wang\(^1\) and Xiaofeng Qian\(^1,2,3\)

Nonlinear photocurrent in time-reversal invariant noncentrosymmetric systems such as ferroelectric semimetals sparked tremendous interest of utilizing nonlinear optics to characterize condensed matter with exotic phases. Here we provide a microscopic theory of two types of second-order nonlinear direct photocurrents, magnetic shift photocurrent (MSC) and magnetic injection photocurrent (MIC), as the counterparts of normal shift current (NSC) and normal injection current (NIC) in time-reversal symmetry and inversion symmetry broken systems. We show that MSC is mainly governed by shift vector and interband Berry curvature, and MIC is dominated by absorption strength and asymmetry of the group velocity difference at time-reversed \( \pm \) points. Taking \( \mathcal{PT} \)-symmetric magnetic topological quantum material bilayer antiferromagnetic (AFM) \( \text{MnBi}_2\text{Te}_4 \) as an example, we predict the presence of large MIC in the terahertz (THz) frequency regime which can be switched between two AFM states with time-reversed spin orderings upon magnetic transition. In addition, external electric field breaks \( \mathcal{PT} \) symmetry and enables large NSC response in bilayer AFM \( \text{MnBi}_2\text{Te}_4 \), which can be switched by external electric field. Remarkably, both MIC and NSC are highly tunable under varying electric field due to the field-induced large Rashba and Zeeman splitting, resulting in large nonlinear photocurrent response down to a few THz regime, suggesting bilayer AFM-\( z \) \( \text{MnBi}_2\text{Te}_4 \) as a tunable platform with rich THz and magneto-optoelectronic applications. Our results reveal that nonlinear photocurrent responses governed by NSC, NIC, MSC, and MIC provide a powerful tool for deciphering magnetic structures and interactions which could be particularly fruitful for probing and understanding magnetic topological quantum materials.

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INTRODUCTION

Magnetic topological quantum materials have attracted a lot of interest as the interplay between magnetic ordering and topology may enable exotic topological quantum states. For example, antiferromagnetic (AFM) topological insulator\(^1\) can host topological axion states with nonzero quantized Chern-Simons magneto-electric coupling\(^2\), which was recently predicted in the class of layered \( \text{MnBi}_2\text{Te}_4 \) materials\(^3-5\). The nontrivial topological surface states have been experimentally verified in \( \text{MnBi}_2\text{Te}_4 \)\(^6-8\). While various experiments have been carried out to study their electronic structure and transport properties, optical probes, especially nonlinear optical spectroscopy, may provide a rich set of alternative tools to probe their topological nature and understand the inherent electronic structure in these exotic quantum materials.

Nonlinear optical responses play a key role in understanding the symmetry, ordering, and topology. Materials with strong nonlinear responses are particularly valuable for ultrafast nonlinear optics\(^9\), efficient generation of entangled photon pairs\(^10,11\), and phase-matching free nonlinear optics\(^12,13\) using 2D materials. Recently, nonlinear photocurrent responses including normal shift current (NSC)\(^14-18\), normal injection current (NIC)\(^17,19\), and Berry curvature dipole-induced current\(^20,21\) have been observed in time-reversal invariant noncentrosymmetric systems. Recently, nonlinear shift current was proposed in centrosymmetric crystals via geometric photon-drag effect\(^22\). In particular, Berry curvature dipole induced photocurrent is closely coupled with ferroelectric orders, offering a unique approach to detect low-energy ferroelectric transition. This was recently theoretically proposed and experimentally verified in ferroelectric few-layer \( \text{WTe}_2 \) semimetal, opening avenues to the development of nonlinear quantum memory\(^24,26\). Compared to few-layer \( \text{WTe}_2 \), layered \( \text{MnBi}_2\text{Te}_4 \) is magnetic with tunable nontrivial topology, which may lead to magnetically and electrically controlled large nonlinear photocurrent responses. The effect of magnetic ordering and intrinsic topology on nonlinear responses in \( \text{MnBi}_2\text{Te}_4 \), however, has been largely underexplored.

Here we provide a microscopic theory of two types of second-order nonlinear current responses, namely magnetic injection current (MIC) and magnetic shift current (MSC), which are the counterpart of the well-known NIC and NSC in time-reversal invariant noncentrosymmetric system. Both MIC and MSC can be induced in \( \mathcal{PT} \)-symmetric systems with space inversion \( \mathcal{P} \) and time reversal \( \mathcal{T} \) being individually broken. We take magnetic topological quantum material bilayer AFM \( \text{MnBi}_2\text{Te}_4 \) as an example of \( \mathcal{PT} \)-symmetric system and predict that MIC can be switched in time-reversal breaking \( \text{MnBi}_2\text{Te}_4 \) upon magnetic transition, namely magnetically switchable nonlinear photocurrent. In addition, vertical electric field breaks \( \mathcal{PT} \) symmetry and enables large electrically switchable NSC. Both MIC and NSC are highly tunable owing to field-induced Rashba and Zeeman splitting. Field induced bandgap reduction not only induces topological phase transition of bilayer AFM \( \text{MnBi}_2\text{Te}_4 \) from zero-plato quantum anomalous Hall insulator into high Chern number quantum anomalous Hall insulator, but also leads to large nonlinear photocurrent response red-shifted down to a few...
Thz regime, suggesting that MnBi$_2$Te$_4$ may serve as a promising platform for terahertz sensing and magneto-optoelectronic applications. The rich nonlinear photocurrent responses in time-reversal and inversion symmetry broken systems are inherently coupled with their crystal structure and magnetic symmetry, offering a promising route for probing and understanding the intrinsic electronic structure and potentially topological physics in magnetic quantum materials.

RESULTS

Microscopic theory of second-order nonlinear photocurrent

General second-order direct current injection (IC) and shift current (SC) under monochromatic electric field $E^0(t) = E^0(\omega) e^{i \omega t}$ with $\omega = \pm \omega$ in the weak limit are given by $^{26,27}$

$$\frac{d E^0}{dt} = -\frac{ne^3}{\hbar^2} \int [d \mathbf{k}] \sum_{m,n} f_m a_m^a b_m^b n_m r_m^c n_m \delta(\omega n_m - \omega) E^b(\omega) E^c(-\omega),$$

(1)

$$\frac{d E^0}{dt} = -\frac{ne^3}{2\hbar^2} \int [d \mathbf{k}] \sum_{m,n} f_m a_m^a b_m^b n_m r_m^c n_m \delta(\omega n_m - \omega) E^b(\omega) E^c(-\omega),$$

(2)

where $a, b, c$ are Cartesian indices, $r_{mn}^{ab} = \frac{\partial \Phi^{ab}}{\partial n_m} - \frac{\partial \Phi^{ab}}{\partial n_n}$ is the gauge covariant derivative. $r_{mn} = i(n)\delta_k(m)$ and $A_n = i(n)\delta_k(n)$ are interband and intraband Berry connection, respectively. $f_m$ is the Fermi-Dirac distribution with $f_m \equiv f_{mn}$. $\delta^{ab}_n \equiv \delta^{ab}_m - \frac{v_n^{ab}}{v_m^{ab}}$ is the group velocity difference of band $m$ and $n$. $\delta^{ab}(\mathbf{2})$ for $d$-dimension. Here $e = -|e|$ carries a negative sign due to the negative charge of electron. We separate the product of the electric field amplitude into the symmetric real and antisymmetric imaginary parts,

$$E^0(\omega) E^0(-\omega) = E^b(\omega) E^c(\omega) = \text{Re}(E^b E^c) + \text{Im}(E^b E^c),$$

(3)

$$E^0(-\omega) E^0(\omega) = E^b(\omega) E^c(\omega) = \text{Re}(E^b E^c) - \text{Im}(E^b E^c).$$

(4)

For linearly polarized light, $E(\omega)$ is real, while for left/right-circularly polarized light $E(\omega)$ is complex with $\text{Im}(E^b(\omega) E^c(\omega)) = -\text{Im}(E^b(\omega) E^c(\omega)) \neq 0$. After symmetrization with $\pm \omega$, we have

$$\frac{d e_m}{dt} = -\frac{ne^3}{\hbar^2} \int [d \mathbf{k}] \sum_{mn} f_m a_m^a n_m b_m^b r_m^c n_m \delta(\omega n_m - \omega) \text{Re}(E^b E^c),$$

$$-\frac{d e_m}{dt} = -\frac{ne^3}{\hbar^2} \int [d \mathbf{k}] \sum_{mn} f_m a_m^a b_m^b n_m r_m^c n_m \delta(\omega n_m - \omega) \text{Im}(E^b E^c),$$

(5)

where $\{r_m^{ab}, r_m^{bc}, r_m^{ac}\} \equiv \{r_m^{ab}, r_m^{bc}, r_m^{ac}\}$ and $\{r_m^{bc}, r_m^{ac}, r_m^{ab}\} \equiv \{r_m^{bc}, r_m^{ac}, r_m^{ab}\}$. We then arrive at

$$\frac{d \eta_{\text{NC}}}{dt} = -\frac{ne^3}{2\hbar^2} \int [d \mathbf{k}] \sum_{mn} f_m a_m^a b_m^b n_m r_m^c n_m \delta(\omega n_m - \omega),$$

(6)

$$\eta_{\text{NC}} = -\frac{ne^3}{2\hbar^2} \int [d \mathbf{k}] \sum_{mn} f_m a_m^a b_m^b n_m r_m^c n_m \delta(\omega n_m - \omega),$$

(7)

$$\eta_{\text{NC}} = -\frac{ne^3}{2\hbar^2} \int [d \mathbf{k}] \sum_{mn} f_m a_m^a b_m^b n_m r_m^c n_m \delta(\omega n_m - \omega),$$

(8)

$\eta_{\text{NC}}$ is the well-known NIC susceptibility, which vanishes under linearly polarized light. $\eta_{\text{NC}}$ is an additional term, MIC susceptibility, arising from our microscopic derivation, which recovers the same formula as Yan's work$^{28}$. It's clear that $\eta_{\text{NC}} = 0$ under time-reversal symmetry. Detailed derivation of nonlinear photocurrent responses using density matrix formalism can be found in Supplementary Note 1.

Similarly, we can separate SC into the well-known MSC and an additional term—MSC:

$$J_0^0 = J_0^0 + J_0^0 = 2ne^3 \text{Re}(E^b E^c) + 2\eta_{\text{NC}} \text{Im}(E^b E^c),$$

(9)

where

$$\eta_{\text{NC}} = -\frac{ne^3}{2\hbar^2} \int [d \mathbf{k}] \sum_{mn} f_m a_m^a b_m^b n_m r_m^c n_m \delta(\omega n_m - \omega) + \delta(\omega n_m - \omega) - \delta(\omega n_m - \omega),$$

(10)

$$\eta_{\text{NC}} = -\frac{ne^3}{2\hbar^2} \int [d \mathbf{k}] \sum_{mn} f_m a_m^a b_m^b n_m r_m^c n_m \delta(\omega n_m - \omega) + \delta(\omega n_m - \omega) - \delta(\omega n_m - \omega).$$

(11)

Under time-reversal symmetry, the MSC and MSC susceptibilities can be rewritten in a compact form as follows,

$$\eta_{\text{NC}} = -\frac{ne^3}{2\hbar^2} \int [d \mathbf{k}] \sum_{mn} f_m a_m^a b_m^b n_m r_m^c n_m \delta(\omega n_m - \omega) - \delta(\omega n_m - \omega),$$

(12)

$$\eta_{\text{NC}} = -\frac{ne^3}{2\hbar^2} \int [d \mathbf{k}] \sum_{mn} f_m a_m^a b_m^b n_m r_m^c n_m \delta(\omega n_m - \omega),$$

(13)

where $R_{mn}^{ab}(\mathbf{r}) = -\frac{\partial \Phi^{ab}}{\partial n_m} + A_{mn}^{ab} - A_{nm}^{ab}$ is the shift vector and $\Phi_{mn}^{ab}(\mathbf{r})$ is the phase factor of the interband Berry connection $R_{mn}^{ab}(\mathbf{r}) = \Phi_{mn}^{ab}(\mathbf{r})$ and $\Omega_{mn}^{ab} = i\Phi_{mn}^{ab} = i(\Phi_{mn}^{ab} - \Phi_{mn}^{ab})$ is the local Berry curvature between band $m$ and $n$. As indicated by Levi-Civita tensor $\epsilon_{abc}$, the direction of light propagation for MSC is along $\mathbf{d}$, i.e., along $(E^b \times E^c)$. The MSC is vanishing in time-reversal invariant system since Berry curvature $\Omega_{mn}^{ab}$ is an odd function and shift vector is even function with $\mathbf{k}$ under time-reversal symmetry. MSC can be induced by circularly polarized light in gyrotropic magnetic materials. During the preparation of this manuscript, we noticed that another paper$^{29}$ by de Juan derived an equivalent equation using the similar density matrix approach and Fei et al.$^{30}$ also proposed giant linearly polarized photogalvanic effect using a diagrammatic approach which is equivalent to MIC. In the present study, bilayer MnBi$_2$Te$_4$ is non-gyrotropic, hence it has vanishing MSC. Nevertheless, MSC in gyrotropic magnetic materials is worth for a further exploration which is closely related to magnetic bulk phototactic$^{31}$. It should be noted that the term of “magnetic” nonlinear current defined in the MSC and MIC in Eqs. (8) and (11) is different from conventional spin current. Instead, it refers to the charge current which is present in magnetic materials but vanishes in intrinsic nonmagnetic materials. Furthermore, we can extend the above nonlinear SC and IC to nonlinear spin current and nonlinear injection spin current by substituting the current operator $\mathbf{J} = -e\mathbf{v}_s - \frac{1}{2} \{s, \mathbf{v}_s\}$, where $s = \frac{1}{2} \sigma_3$ is spin operator with Pauli matrices $\sigma_3$. The extended nonlinear photon spin current responses provide an essential route for exploring spin physics in condensed matter, which is currently under exploration and out of the scope in the present work.

Crystal structure and group theory of bilayer MnBi$_2$Te$_4$

Here we present a magnetic group theoretical analysis of bilayer AFM-z MnBi$_2$Te$_4$(Fig. 1a, c) with interlayer AFM ordering of the Mn atoms aligned along the $z$ direction, while group theoretical analysis of bilayer MnBi$_2$Te$_4$ with different magnetic orderings can be found in Supplementary Note 2. In general, the dichromatic group $M$ can be constructed by $M = G(H) = H \oplus (G - H)^T$, where $G$ is the ordinary geometric point group, $H$ is the subgroup of $G$ of index 2, and $T$ is time-reversal operator. As a result, magnetic point group of bilayer AFM-z MnBi$_2$Te$_4$ is $\mathbb{B}_3 = \mathbb{D}_3(3) = [E, 2C_3, 3C_2, i\mathbb{T}, 2S_5, 3S_5\mathbb{T}$. The character table of point group $\mathbb{D}_3$ is included in Supplementary Table S1.
Without considering the ordering of spin, bilayer MnBi$_2$Te$_4$ is centrosymmetric with vanishing second-order nonlinear current responses. Taking into account the AFM-$z$ structure, $PT$ symmetry is preserved in bilayer MnBi$_2$Te$_4$ with individual $P$ and $T$ symmetry being broken. In addition, as shown in Fig. 1b, bilayer AFM-$z$ MnBi$_2$Te$_4$ holds a twofold rotation symmetry along $x$-axis $C_{2x} = M_x$, and the combination of mirror symmetry and time-reversal symmetry $M_x T$. 

For systems holding magnetic point group $\Gamma_0$ (such as bilayer and bulk AFM-$z$ MnBi$_2$Te$_4$), both normal magnetoelectric coupling and topological axion coupling are symmetry allowed. The polar $i$-vectors (i.e., time-reversal symmetric and space-inversion antisymmetric vector), e.g., electric field $E$ or electric polarization $P$, are represented by $A_{2u}(x, y, z)$ and $E_u(x, y, z)$ in $D_{3d}$ group and they are time-reversal symmetric but space-inversion antisymmetric. $A_{2g}(R_z)$ and $E_g(R_x, R_y)$ represent axial $i$-vectors, which are symmetric under both time reversal and space inversion. The general transformation of polar/axial $i/c$-tensors under space inversion $P$, time-reversal operation $T$, and $PT$ can be found in Supplementary Table S2. We can see the two types of vectors are incompatible in $D_{3d}$ group, e.g., $A_{2u} \otimes A_{2g}$ does not contain total symmetric $A_{1g}$. The magnetic field $B$ or orbital magnetization $M$ are axial $c$-vectors, which are time-reversal antisymmetric but space-inversion symmetric. 

For magnetic point groups, we use Birss’s notation$^{12}$ where $\Gamma_m$ is the total symmetric representation, $\Gamma_m$ is the one-dimensional representation that corresponds to $\mathcal{M}$ in which all elements of $\mathcal{M}$ are represented by $+1$, and $\Gamma_p$ is the pseudovector representation in group $\mathcal{G}$. We shall write the direct product representation $\Gamma_{mip}$ for axial $c$-vector. For magnetic point group $D_{3d}(D_3)$, we have $\Gamma_m = A_{1g}, \Gamma_p = A_{1u}, \Gamma_p = A_{2g} + E_g$. Thus, $\Gamma_{mip} = A_{2u} + E_u$. Consequently, $\Gamma_m \otimes \Gamma_p = \Gamma_p \otimes \Gamma_B = 2A_{1g}, A_{2g} + 3E_g$. Hence, the linear magnetoelectric coupling of $\mathcal{M}$'s structures has two independent nonvanishing components and corresponding orbital magnetoelectric polarizability. Now let’s see how $c$-type second-order current response under linearly polarized light such as MIC is enabled in $D_{3d}(D_3)$ by considering the following direct product $\Gamma_m \otimes \Gamma_p \otimes \Gamma_{BC} = A_{1g} + 3A_{2g} + 4E_u$, indicating only one independent and nonvanishing susceptibility is allowed. For $i$-type second-order current response under circularly polarized light such as NSC, we have the direct product: $\Gamma_j \otimes (\Gamma_p \otimes \Gamma_{BC}) = (A_{2u} + E_u) \otimes (2A_{1g} + E_g) = A_{1u} + 3A_{2u} + 3E_u$. Hence, NSC is not symmetry allowed. In fact, NSC is immune to the time-reversal symmetry and can be determined by nonmagnetic point group. As bilayer/bulk MnBi$_2$Te$_4$ holds nonmagnetic point group $D_{3d}$ with inversion symmetry, both NSC and MIC vanish. Similarly, $c$-type second-order current response under circularly polarized light such as MSC is forbidden in $D_{3d}(D_3)$ by considering the following direct product: $\Gamma_m \otimes \Gamma_p \otimes \Gamma_{BC} = 2A_{1u} + A_{2u} + 3E_u$ which does not contain total symmetric representation. The above analysis can be extended to other higher order optical/photocurrent responses in magnetic materials.

Nonlinear photocurrent in bilayer MnBi$_2$Te$_4$ Ground-state crystal structures of MnBi$_2$Te$_4$ were calculated using first-principles density functional theory$^{35,36}$ implemented in the Vienna Ab initio Simulation Package (VASP)$^{35,36}$ with the projector-augmented wave method$^{37}$. We then performed first-principles calculations of nonlinear photocurrent including MIC and NSC. Specifically, with the fully relaxed ground-state crystal structure, we constructed quasiatomic spinor Wannier functions and tight-binding Hamiltonian from Kohn–Sham wavefunctions and eigenvalues under the maximal similarity measure with respect to pseudatomic orbitals$^{38,39}$. Using the developed tight-binding Hamiltonian, we then computed MIC and NSC susceptibility tensor using a modified WANNIER90 code$^{40}$ with a k-point grid of 1000 $\times$ 1000 $\times$ 1. Spin–orbit coupling was taken into account. More details can be found in the "Methods" section. 

Figures 2a and b present the electronic structure and MIC susceptibility $\eta_{MIC}^{\vec{p}}$ in bilayer MnBi$_2$Te$_4$ with magnetic ordering in AFM-$z$ (↑↑) and time-reversed AFM-$z$ (tAFM-$z$, ↑↓), respectively. The corresponding magnetic ordering is shown in Fig. 2a inset.
The peak MIC conductivity is \(C_{24}\) asymmetric group velocity \(\eta\) respectively. This term arises from the large difference of the \(fi\) aligned along example, FM-\(z\) varying magnetic orders that are different from the above AFM-\(z\) \(\eta\) leads to vanishing \(f_{mn}\) magnetic ordering transition between AFM-\(z\) \(\eta\) is determined by the integral of \(f_{mn}\) \(z\) \(\Delta_{mn}\) at time-reversed \(\pm k\) points, which eventually contributes to the large MIC in bilayer AFM-\(z\) and tAFM-\(z\) MnBi2Te4. The peak MIC conductivity is \(\sim 2 \times 10^{-8} \text{nm} \cdot \text{A} \cdot \text{V}^{-2} \cdot \text{s}^{-1}\), two times the peak NIC in ferroelectric monolayer GeS17. More importantly, it demonstrates that the MIC can be switched upon magnetic ordering transition between AFM-\(z\) \(\uparrow \downarrow\) and its time-reversed tAFM-\(z\) \(\downarrow \uparrow\) configurations. With additional threefold rotation symmetry, we have \(\eta_{\text{MIC}} = -\eta_{\text{MIC}} = -\eta_{\text{MIC}} = -\eta_{\text{MIC}}\). Moreover, the presence of \(M_{x}, M_{y}\) symmetry leads to vanishing \(\eta_{\text{MIC}}\). The switching of MIC for bilayer AFM-\(z\) and tAFM-\(z\) MBT can be understood from a symmetry analysis. The MIC is determined by the integral of \(f_{mn}\) \(\Delta_{mn}\) \(r_{mn}(k)\) \(\delta(\omega_{mn} - \omega)\) in the Brillouin zone. Under time-reversal operation, we have

\[
T \sum_{mnk} f_{mn}(k) \Delta_{mn}(k) \{r_{mn}(k), r_{mn}(k)\} \delta(\omega_{mn} - \omega) = -\sum_{mnk} f_{mn}(k) \Delta_{mn}(k) \{r_{mn}(k), r_{mn}(k)\} \delta(\omega_{mn} - \omega)
\]

\[
= \sum_{mnk} f_{mn}(k) \Delta_{mn}(k) \{r_{mn}(k), r_{mn}(k)\} \delta(\omega_{mn} - \omega).
\]

Hence, MIC switches the sign in the AFM-\(z\) and tAFM-\(z\) MBT.

Van der Waals layered MnBi2Te4 hosts rich topology with varying magnetic orders that are different from the above AFM-\(z\) structure (Supplementary Table S3), subsequently the corresponding nonlinear current responses can be very different. For example, FM-x and FM-z magnetic ordering under external magnetic field (i.e., ferromagnetic ordering with magnetization aligned along x/z direction) holds inversion symmetry, hence has vanishing even-order nonlinear optical and photocurrent responses including SHG, NSC, NIC, MSC, and MIC. Supplementary Fig. S1 shows the band structure for the FM-z magnetic ordering where each band is symmetric with respect to \(\pm k\) points with vanishing MIC. In addition, for bilayer MnBi2Te4 with AFM-x magnetic ordering, the corresponding magnetic point group becomes \(2/m\) \(C_{2h}\), \(T = (\hat{E}, \hat{C}_{2y}, \hat{C}_{2x}, \hat{I}, \hat{T})\), different from \(3m\) for bilayer MnBi2Te4 with AFM-z magnetic structure. Nonetheless, bilayer AFM-x MnBi2Te4 preserves \(PT\)-symmetry and allows for nonvanishing MIC response \(\eta_{\text{MIC}}\), \(\eta_{\text{MIC}}\) and \(\eta_{\text{MIC}}\) as shown in Supplementary Fig. S2. It is worth to point out that van der Waals layered MnBi2Te4 hosts rich topology with varying magnetic ordering. Probing MIC response can, therefore, help directly determine the intrinsic magnetic structure and hence potential topology of magnetic topological quantum materials owing to the intimate coupling between magnetic symmetry and nonlinear photocurrent responses.

NSC in electrically gated bilayer MnBi2Te4

Although both bilayer AFM-\(z\) and tAFM-\(z\) MnBi2Te4 hold the large MIC response as shown above, their Berry curvature \(\Omega_{\alpha}(k)\) and shift vector \(R_{\alpha}(k)\) are vanishing due to \(PT\) \(\Omega_{\alpha}(k) = -\Omega_{\alpha}(k) = \Omega_{\alpha}(k)\) and \(PT R_{\alpha}(k) = -R_{\alpha}(k)\). Hence, no NSC and Berry curvature related physics would be expected in \(PT\)-symmetric system. In fact, one can take the wave functions to be real (due to the \(PT\) transformation). Then the phase effect is absent and the shift vector and Berry curvature become zero in \(PT\)-symmetric system. However, one can apply Berry curvature engineering via electric gating to break the \(PT\) symmetry in the pristine bilayer MnBi2Te4. Here we apply an external electric field along the out of plane direction to bilayer MnBi2Te4. Figure 3a, b show the calculated electronic structure and NSC in bilayer MnBi2Te4 under an external electric field \(E_{z} = 10 \text{mV} \cdot \text{Å}^{-1}\) with magnetic ordering AFM-\(z\) \(\uparrow \downarrow\). Large Rashba and Zeeman spin splitting are indicated in Fig. 3a, which we will discuss soon. The peak NSC conductivity is \(\sim 1000 \text{nm} \cdot \mu \text{A} \cdot \text{V}^{-2}\), which is 50 times larger than that in GeS17. In addition, Fig. 3c-f presents the microscopic distribution of NSC,
The response of MIC is dominated by second-order nonlinear current responses due to the intraband operation. However, for the same magnetic configuration, NSC and MIC in bilayer AFM-\(\text{T}\) have strong electronic gap, thereby shifting the nonlinear photocurrent responses such as MIC and NSC to the THz regime. Figure 4e, f shows field-dependent NSC and MIC responses in bilayer AFM-\(\text{T}\) under varying electric field. It clearly demonstrates that tunable nonlinear photocurrent in bilayer MnBi\(_2\)Te\(_4\).

Both NSC and MIC are highly tunable under varying electric field. Figure 4a shows the electric field-dependent band structure. The bandgap decreases upon increasing vertical electric field until a critical field of \(E_Z = 25 \text{ mV \, Å}^{-1}\). At the same time, it undergoes a topological phase transition from zero-plateau quantum anomalous Hall insulator with Chern number \(C_T = 0\) to high Chern number quantum anomalous Hall insulator (\(C_T = 3\)). The nontrivial topology is evident in the Berry curvature shown in Fig. 4b, which is localized around the center of the Brillouin zone. The integration of Berry curvature yields a high Chern number of \(C_T = 3\). As aforementioned, electric field can introduce large Rashba and Zeeman splitting in bilayer MnBi\(_2\)Te\(_4\). In Fig. 4c, d we show the corresponding spin polarization at the energy of 0.6 eV with AFM-\(\text{T}\) and tAFM-\(\text{T}\) magnetic ordering under a vertical electric field of \(E_Z = 10 \text{ mV \, Å}^{-1}\), which confirms large spin splitting with threefold rotation symmetry under finite electric field. Under the same electric field, spin polarization in AFM-\(\text{T}\) and tAFM-\(\text{T}\) are related by time-reversal \(T\) operation. However, for the same magnetic configuration AFM-\(\text{T}\) or tAFM-\(\text{T}\), the spin polarization under electric field along \(+z\) and \(-z\) are related by PT operation (see Supplementary Fig. S3).

Electric field-induced large spin splitting and distinct spin polarization suggest that bilayer AFM-\(\text{T}\) MnBi\(_2\)Te\(_4\) could serve as a rich platform for developing electrically controlled spintronics.
the strength of NSC response increases under increasing electric field until \( E_z = 23 \text{ mV Å}^{-1} \), and the MIC conductivity remains very large until \( E_z = 23 \text{ mV Å}^{-1} \). More importantly, the low-energy peak of NSC and MIC response linearly decreases to \(-25 \text{ meV} \) upon increasing field up to \( E_z = 23 \text{ mV Å}^{-1} \). It's worth to point out that NSC will switch the sign upon the switching of electric field from \(+z\) to \(-z\), however, the sign of MIC will remain the same. This is another way to distinguish NSC and MIC. We would like to mention that the NSC and MIC calculated here is solely contributed by bulk response. Topologically protected edge states in QAH will also contribute nonlinear current responses, which is outside the scope of this work and needs to be explored in future.

**DISCUSSION**

The distinct directional dependence and switching behavior of NSC and MIC under different electric field and magnetic ordering provide a fundamental basis to distinguish these two current responses in experiment. First, as shown above, under linearly polarized light, MIC can be generated in PT-symmetric system such as bilayer AFM-z MnBi2Te4. NSC, though absent in the PT-symmetric system, can be enabled via electric gating. Moreover, for electrically gated bilayer AFM-z and tAFM-z MnBi2Te4 under linearly \( x \)-polarized light, NSC susceptibility tensor element \( n_{xx}^{MC} \) contributes a large nonlinear photocurrent along \( y \) only, while MIC susceptibility tensor element \( n_{yy}^{MC} \) contributes a large nonlinear photocurrent along \( x \) only. More importantly, nonlinear photocurrent from NSC can be switched by applying vertical electric field along \( +z/-z \) direction, while nonlinear photocurrent from MIC can be reversed by inducing magnetic transition between AFM-z and tAFM-z. Furthermore, different magnetic orderings (e.g., AFM-z, AFM-x, FM-z, and FM-x) yield distinct magnetic point group, leading to different directional dependence of nonlinear photocurrent. Such rich coupling between the magnetic ordering and nonlinear photocurrent responses presents itself a powerful tool for investigating magnetic symmetries, structures, and interactions in magnetically ordered crystals as well as imaging magnetic domain walls.

Bilayer AFM-z MnBi2Te4 possesses large MIC and NSC that are highly tunable under electric field with the first low-energy peak red-shifted down to 25 meV (i.e., \(-6 \text{ THz} \)), offering a promising platform for THz sensing of extreme nonlinear quantum phenomena. The large MIC response is originated from the large absorption strength and asymmetry of the group velocity phenomena. The large MIC response is originated from the large absorption strength and the large shift vector which is related to large inter-band Berry curvature. Topological insulators including the MBT studied here usually have small bandgap, large inter-band transition matrix elements, and large Berry curvature, which is expected for most magnetic topological insulators with symmetry-allowed nonlinear photocurrent. PT symmetry plays an important role in the present case, and it is also crucial for non-Hermitian magnetic structures with balanced gain and loss. In addition to MIC and NSC, MSC presents another interesting second-order photocurrent response, which vanishes in time-reversal invariant systems. MSC can be induced by circularly polarized light in gyrotropic magnetic materials. Furthermore, nonlinear SC and IC can be extended to nonlinear shift spin current and nonlinear injection spin current, yielding nonlinear photo-spin current response. This will provide an essential route for exploring spin physics in condensed matter, which is currently under exploration and out of the scope of the present work.

In summary, the intimate coupling between intrinsic magnetic point group and nonlinear optical/photocurrent responses makes nonlinear spectroscopy and imaging a powerful tool for investigating magnetic symmetries, structures, and interactions in magnetically ordered crystals. In particular, we predicted that PT-symmetric magnetic topological quantum material bilayer MnBi2Te4 possesses large, magnetically switchable MIC in the THz regime. Although NSC vanishes in PT symmetric system, electric field breaks the PT-symmetry in bilayer AFM-z MnBi2Te4 and enables large NSC response at the IR regime. More importantly,
NSC can be switched by vertical electric field and MIC be switched upon magnetic transition, however, NSC (MIC) will not be switched by magnetic transition (electric field). Due to the magnetic group symmetry of bilayer AFM-z MnBi₂Te₄, MIC, and NSC yield photocurrent that are perpendicular to each other upon linearly x/y-polarized light, hence can be distinguished. Very excitingly, electric gating can efficiently tune the large nonlinear photocurrent response down to 6 THz, suggesting bilayer AFM-z MnBi₂Te₄ as an exciting platform for THz and magneto-optoelectronic applications as well as 2D spintronics that are electrically and magnetically tunable. The present work reveals that nonlinear photocurrent responses governed by NSC, NIC, MISC, and MIC provide a powerful spectroscopic/imaging tool for the investigation of magnetic structures and interactions, which could be particularly fruitful for probing and understanding magnetic topological quantum materials.

METHODS

Ground-state crystal and electronic structure

Ground-state crystal structures of MnBi₂Te₄ were calculated using the first-principles density functional theory6,15,16,34 and implemented in the VASP65,36 with the projector-augmented wave method37 and a plane-wave basis with an energy cutoff of 400 eV. We employed the generalized-gradient approximation of exchange-correlation energy functional in the Perdew–Burke–Emzerhof form. A Hubbard U correction with U = 4 eV was applied to Mn atoms to reduce the self-interaction error present in DFT-PBE. We further adopted a Monkhorst-Pack k-point sampling of 11 × 11 × 1 for the Brillouin zone integration and DFT-D3 functional45 to account for weak interlayer dispersion interactions. The convergence criteria for maximal residual force was set to 0.005 eV Å⁻¹, and the convergence criteria for electronic relaxation was set to 10⁻⁶ eV.

First-principles calculations of nonlinear photocurrent

With the fully relaxed ground-state crystal structure, we constructed quasiatomic spinor Wannier functions and tight-binding Hamiltonian from Kohn–Sham wavefunctions and eigenvalues under the maximal similarity measure with respect to pseudomolecular orbitals46,39. Spin–orbit coupling was taken into account. Total 120 quasiatomic spinor Wannier functions were obtained from the projections onto Mn’s s and d pseudomolecular orbitals, Te’s s and p pseudomolecular orbitals, and Bi’s s and p pseudomolecular orbitals for bilayer MnBi₂Te₄. Using the developed tight-binding Hamiltonian, we then computed MIC and NSC susceptibility tensor using a modified WANNIER90 code40. A small imaginary smearing factor η of 5 meV was applied to fundamental frequency. A k-point grid of 1000 × 1000 × 1 was adopted, which is dense enough to reach the convergence as shown in Supplementary Fig. 54. Tight-binding Hamiltonian was not directly symmetrized; however, we carefully inspect the resulted electronic structure by comparing the calculated band structures with the recent publications by Li et al.31 and Du et al.41 and our calculations are in good agreement with their results. Second, the electronic band structure of bilayer AFM-z MBT (Fig. 1d) and the contour plot of the energy difference agree with their results. Second, the electronic band structure of

DATA AVAILABILITY

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

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AUTHOR CONTRIBUTIONS
X.Q. conceived the project. H.W. developed first-principles code for magnetic injection current susceptibility and carried out the calculations. H.W. and X.Q. conducted theoretical analysis, analyzed the results, and wrote the manuscript.

COMPETING INTERESTS
The authors declare no competing interests.

ADDITIONAL INFORMATION
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Correspondence and requests for materials should be addressed to X.Q.

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