Dynamical magnetoelectric effect in antiferromagnetic insulator Mn$_2$Bi$_2$Te$_5$

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The dynamical axion state is a new state of quantum matter with a dynamical topological axion response, which is fundamentally different from axion insulator with a quantized static axion response. Its exhibits many exotic topological phenomena such as dynamical magnetoelectric effects. However, the experiments are hindered by lacking of realistic materials. Here we propose a simple and concrete criterion for realizing a large dynamical axion field in antiferromagnetic insulating states. We predict the tetradymite-type compound Mn$_2$Bi$_2$Te$_5$ and its related materials host the dynamical axion field. We further propose the optical and transport experiments to detect such a dynamical axion field. Our results could directly aid and facilitate the search for dynamical axion state in realistic materials.

Topological phenomena in physical systems are determined by some topological structure and are thus usually universal and robust against perturbations [1]. The magnetic topological insulators (TIs) [2] brings the opportunity to realize a large family of exotic topological phenomena [3–31]. The electromagnetic response of a three-dimensional insulator is described by the topological term $S_0 = (\theta/2\pi)(e^2/h) \int d^4x dt \mathbf{E} \cdot \mathbf{B}$ [3], together with the ordinary Maxwell action. Here $\mathbf{E}$ and $\mathbf{B}$ are the conventional electromagnetic fields inside the insulator, $e$ is the charge of an electron, $h$ is Planck’s constant, and $\theta$ is the dimensionless pseudoscalar parameter describing the insulator, which refers to axion field in particle physics [32, 33]. Under periodic boundary conditions, all physical quantities depends on $\theta$ only module $2\pi$. While $S_0$ generically breaks time-reversal symmetry $T$ and parity $P$, both symmetries are conserved at $\theta = 0$ and $\theta = \pi$. Therefore, TIs defined by $\theta = \pi$, cannot be by adiabatically connected to trivial insulator defined by $\theta = 0$, without $T$-breaking perturbations. $S_0$ with the static $\theta = \pi$ leads to image monopole [6] and quantized topological magnetoelectric effect (TME) [3, 12–14, 34–37].

When the antiferromagnetic (AFM) long-range order is developed, $\theta$ becomes a dynamical field from magnetic fluctuations and taking continuous values from 0 to $2\pi$. Such magnetic materials with dynamical axion fields (DAFs) leads to many exotic physical effects such as axionic polariton [8], axion instability induced electromagnetic effect [38, 39] and so on. However, the experiments are hindered by lacking of realistic materials [8, 40–44]. The realization of a DAF require a proper coupling between electrons and magnetic fluctuations. The conventional magnetoelectric material Cr$_2$O$_3$ may exhibit DAF [45, 46]. However, the axion field, if exists in Cr$_2$O$_3$, is expected to be quite weak and anisotropic. In this letter, we provide a simple criterion to find materials with a $large$ isotropic axion field, and further predict Mn$_2$Bi$_2$Te$_5$ family materials as potential candidates. A smoking gun signature for DAF is dynamical chiral magnetic effect (CME), where an alternating electric current is generated by static magnetic fields from DAF induced by AFM resonance, which is absent in axion insulator.

Dynamical magnetoelectric effects. In terms of the electromagnetic potential $A_\mu$, the topological axion term in the dynamical axion state is

$$S_\theta = \frac{1}{4\pi} \frac{e^2}{h} \int d^4x dt e^{\mu\nu\sigma\tau} \theta(x,t) \partial_\mu A_\nu \partial_\sigma A_\tau. \quad (1)$$

Here $\theta(x,t) = \theta_0 + \delta\theta(x,t)$, $\theta_0$ and $\delta\theta(x,t)$ is the static and dynamical part of axion field, respectively. $\theta_0$ generically deviates from $\pi$ or 0. $\mu, \nu, \sigma, \tau = t, x, y, z$. $x = (x, y, z)$. The dynamical axion state is essentially different from axion insulator with $\theta_0 = \pi$ and $\delta\theta(x,t) = 0$.

The nonzero $\delta\theta(x,t)$ in DAF leads to the dynamical magnetoelectric effects, which can be seen from the response equation by taking a variation in $S_\theta$ as $j^\mu = (1/2\pi)(e^2/h)\epsilon^{\mu\nu\sigma\tau} \partial_\nu \theta(x,t) \partial_\sigma A_\tau$. Therefore the current density in the spatial space ($\mu = x, y, z$) is

$$j(x,t) = \frac{1}{2\pi} \frac{e^2}{h} [\nabla \theta(x,t) \times \mathbf{E} + \partial_t \theta(x,t) \mathbf{B}]. \quad (2)$$

$\partial_t \equiv \partial/\partial t$. The first term is the anomalous Hall effect (AHE) induced by the spatial gradient of $\theta$, which is essentially different from AHE in ferromagnetic (FM) [47] and frustrated AFM metals [48, 49]. Such a term leads to a half-quantized Hall effect on the surface in TIs, which is the physical origin of TME and quantized AHE [3]. The second term is the dynamical CME [50], where an electric current is generated by magnetic fields from temporal gradient of $\theta$. Such an electric current is a polarization current $j = \partial_t \mathbf{P}$ in insulators, where $\mathbf{P}$ is the charge polarization. In a static uniform magnetic field we have $\partial_t \mathbf{P} \propto \partial_t [\theta(x,t) \mathbf{B}]$, so $\mathbf{P} = (\theta(x,t)/2\pi)(e^2/h) \mathbf{B}$. This is isotropic TME where charge polarization is induced by
a parallel magnetic field. This is different from CME in Weyl semimetals, which only happens in nonequilibrium situations [51–55]. Also, the dynamical CME will not happen in axion insulator with only static θ.

**Axion electrodynamics.** As θ is odd under T and P operation, only T- and P-breaking perturbations can induce a change of θ. So the fluctuations of AFM order with T, P-breaking can induce δθ(x, t) [8]. A large δθ(x, t) is expected when the magnetic fluctuation is strong. However, this is not sufficient. Here we propose a simple criterion for realizing a large DAF, which is generic for any system supporting axionic excitation and does not rely on a specific model. We would like to start with a simple Dirac model describing an insulator for concreteness and show the relation between θ(x, t) and AFM order. The generic effective Hamiltonian is

\[ H_{\text{Dirac}} = c_0(k) + \sum_{a=1}^5 d_a(k) \Gamma^a, \]

where \( d_{1,2,3,4,5}(k) = (Ak_x, Ak_y, Ak_z, m_4(k), m_5) \), and \( m_4(k) = m + Bk^2 \). For simplicity, we neglect the particle-hole asymmetry \( c_0(k) \) and set the velocity \( A \) and curvature \( B \) along three axes to be isotropic, which does not affect the physics we discuss here. \( \Gamma^a \) are Dirac matrices satisfying the Clifford algebra \( \{ \Gamma^a, \Gamma^b \} = 2\delta_{ab} \) and \( \Gamma^5 = \Gamma^1 \Gamma^2 \Gamma^3 \Gamma^4 \). Also \( T \Gamma^a T^{-1} = -\Gamma^a \) and \( PT \Gamma^a P^{-1} = -\Gamma^a \) for \( a = 1, 2, 3, 5 \), and \( TT^4 \Gamma T^{-1} = PT^4 \Gamma P^{-1} = \Gamma^4 \).

\( m_4 \) is the T-invariant mass, and \( m_5 \) is the T, P-breaking mass proportional to AFM order. When \( m_5 = 0 \), the model describes a TI (or trivial insulator) if \( m/B < 0 \) (or \( m/B > 0 \)). The value of \( \theta \) can be calculated from the momentum-space Chern-Simons form [3, 8, 56] as

\[ \theta = (4\pi/1) \int \mathcal{G}(k) \epsilon^{ijkl} d_1 \partial_k d_2 \partial_k d_3 \partial_k d_4, \]

where \( \mathcal{G}(k) = (2|d| + d_4)/\sqrt{|[d_1|d_2|d_3]^2|d_4|^3|}, |d| = (\sum_{a=1}^5 d_a \epsilon^a)^{1/2}, \) and \( i, j, k, l \) take values from 1, 2, 3, 5 and indicates summation. Thus, \( m_5 \) leads to a correction to \( \theta \) to the linear order. Namely,

\[ \delta\theta(x, t) = \delta m_5(x, t)/g, \]

where \( \delta m_5(x, t) \) is proportional to the AFM fluctuations, and \( 1/g = \partial \theta/\partial m_5 \). There are other leading order T, P-breaking terms \( \sum_{i=1}^5 m_i \Gamma^i \) which only give rise to higher-order contributions to \( \theta \), and thus are neglected here [57].

Typical values of \( \theta_0 \) as a function of \( m \) and \( m_5 \) are calculated in Fig. 1(a) and 1(b), respectively. As expected, \( \theta_0 \) deviates from ±π \((m/B < 0)\) or 0 \((m/B > 0)\) when \( m_5 \) is nonzero. \( \theta_0 \) is an odd function of \( m_5 \), this implies \( g \) is an even function of \( m_5 \) as shown in Fig. 1(d), \( 1/|g| \) is largest when \( m_5 = 0 \). From Fig. 1(c), for \( m_5 \) very close to zero, \( 1/|g| \) have a dip (peak) when \( m \rightarrow 0^- \) \((m \rightarrow 0^+)\), and \( 1/|g| \) at the dip is larger than at the peak. Both the dip and peak in \( 1/|g| \) are further suppressed when \( m_5 \) increases, and finally are smearing out when \( m_5 \) is large. In the latter case, \( 1/|g| \) increases when \( m \) becomes inverted.

This suggests that, to get a large \( \theta_0 \), both large \( \delta m_5 \) and \( 1/|g| \) are required, where the latter is achieved when \( m_5 \rightarrow 0 \) and \( m \) approaches zero from the topological nontrivial side. This further indicates a promising strategy for a large DAF: a T, P-broken insulator which is close to topological phase transition with a small inverted \( m \) and is also close to paramagnetic to AFM transition with \( m_5 \approx 0 \), where the magnetic fluctuation is expected to be strong. In general, in a specific AFM material, the magnetic fluctuation and AFM order are determined by the intrinsic screened Coulomb interaction between electrons, and thus the amplitude of \( \delta m_5 \) and \( m_5 \) are fixed. Therefore, to search for the DAF in practice, intrinsic AFM TIs having a weak AFM order and is near the topological phase transition are preferred.

**Material candidates.** The recent discovery of intrinsic magnetic TIs in MnBi2Te4 family materials [21, 58–62] rekindled our hope for searching DAF in intrinsic AFM TIs. The symmetry guiding principle is to find T, P-breaking but PT-conserving TIs. The AFM order in MnBi2Te4 has no contribution to \( \delta\theta(x, t) \) due to conserved P. However, a class of ternary chalcogenides materials X2A2B5, also written as (XB)2(A2B3)1, with \( X=Mn/Eu, A=Sh/Bi, B=Se/Te \), satisfy the symmetry requirement for DAF. In the following, we take Mn2Bi2Te5 as an example. It has a layered rhombohedral crystal structure, shown in Fig. 2(a), with the space group \( P\bar{3}m1 \) (No. 164) if the spin moments of Mn are ignored. It consists of nine-atom layers (e.g., Te1-Bi1-Te2-Mn1-Te3-Mn2-Te4-Bi2-Te5) arranged along the trigonal
z-axis with the ABC-type stacking, known as a nonuple layer (NL). The coupling between different NLs is the van der Waals type. Once the AFM order is considered, $T$, $\mathcal{P}$ are broken.

First-principles calculations are employed to investigate the electronic structures, and the detailed methods can be found in Supplemental Material [57]. We find that each Mn atom tends to have half-filled $d$ orbitals with a magnetic moment $S = 5 \mu_B$, and the orbital moment is quenched $L = 0$. The total energy calculations for different collinear magnetic structures are given in Fig. 2(c). Here the non-collinear magnetic structure is not considered due to vanishing Dzyaloshinski-Moriya interaction from $\mathcal{P}$. In fact, the non-collinear magnetic configuration was previously found to have higher energy [63]. The $A$-type AFM phase with the out-of-plane easy axis, denoted as 001AFM [seen in Fig. 2(b)], is the magnetic ground state for most $X_2A_2B_5$ family materials, except Eu$_2$Bi$_2$Te$_5$. The in-plane $A$-type AFM phase (100AFM) has a slightly higher total energy than that of 001AFM. The FM order is not favored. The magnetic anisotropy of Eu$_2$Bi$_2$Te$_5$ is quite weak because of the local $4f$ orbital, and a small external magnetic field can easily tune 100AFM into 001AFM. The Goodenough-Kanamori rule is the key to have FM order in each Mn layer. Two nearest Mn atoms are connected through Te atom with the bond "Mn-Te-Mn", whose bonding angle is $\sim 95$ degree, so the superexchange interaction is expected to induce the in-plane FM order.

The 001AFM state breaks $T$ and $\mathcal{P}$ but preserves $\mathcal{P}T$, thus DAF is expected to develop. 001AFM Mn$_2$Bi$_2$Te$_5$ is insulating with a full energy gap about 0.1 eV, shown in Fig. 3(a). Fig. 3(b) shows the density of states (DOS) without spin-orbit coupling (SOC). The red states are the projected DOS of Mn $d$ orbitals. We can see that the occupied $3d^5$ states and unoccupied $3d^0$ states are well separated in energy, indicating a high spin state. The band structure calculated through projecting on Te and Bi $p_z$ orbitals can be found in Supplemental Material [57]. We find that the superexchange interaction is expected to induce FM order in each Mn layer. Two near-$100$AFM into $001$AFM. The Goodenough-Kanamori rule is the key to have FM order in each Mn layer. Two nearest Mn atoms are connected through Te atom with the bond "Mn-Te-Mn", whose bonding angle is $\sim 95$ degree, so the superexchange interaction is expected to induce the in-plane FM order.

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SOC effect pushes down $|P1^{+}_x, \uparrow (\downarrow)\rangle$ state and pushes up $|P2^{+}_z, \uparrow (\downarrow)\rangle$ state, leading to the band inversion and parity exchange. The symmetries of the system are threefold rotation symmetry $C_{3z}$ and $PT$ ($Te3$ as the center). In the basis of $\{|P1^{+}_x, \uparrow\rangle, |P2^{+}_z, \uparrow\rangle, |P1^{+}_x, \downarrow\rangle, |P2^{+}_z, \downarrow\rangle\}$, the representation of the symmetry operations is given by $C_{3z} = \exp[i(\pi/3)\sigma_z^x]$ and $PT = i\tau^z\sigma^yK$ ($P = \tau^z$, $T = i\sigma^yK$), where $K$ is the complex conjugation operator. $\sigma^x, \sigma^y, \tau^z$ and $\tau^x, \tau^y, \tau^z$ denote the Pauli matrices in the spin and orbital space, respectively. By requiring these symmetries and keeping only the terms up to quadratic order in $k$, we obtain the following generic form of the effective Hamiltonian $H(k) = \epsilon_0(k) + M_1(k)\tau^z + M_2(k)\tau^y + A_1(k)\tau^x\sigma^x + A_2(k)(k_x\tau^x\sigma^y - k_y\tau^z\sigma^z)$, where $\epsilon_0(k) = C_0 + C_1k_x^2 + C_2(k_x^2 + k_y^2)$, $M_1(k) = M_0 + M_1k_x^2 + M_2(k_x^2 + k_y^2)$, $M_2(k) = M_0 + M_1k_x^2 + M_2(k_x^2 + k_y^2)$, and $A_2(k_x) = A_2 + A_3k_x$. It is very similar to the Dirac model in Eq. (3), where $M_4(k)$ is the $T, P$-conserving mass term responsible for band inversion. $M_5(k)$ is the $P$-breaking mass term from the 001AFM order. To see the microscopic origin of $M_5(k)$, we change the basis to $(|\alpha, \uparrow\rangle, |\alpha, \downarrow\rangle, |\beta, \uparrow\rangle, |\beta, \downarrow\rangle)$ with $|\alpha, \uparrow\rangle = (1/\sqrt{2})(|P1^{+}_x, \uparrow\rangle + |P2^{+}_z, \uparrow\rangle)$ and $|\beta, \uparrow\rangle = (1/\sqrt{2})(|P1^{+}_x, \uparrow\rangle - |P2^{+}_z, \uparrow\rangle)$. We have $P|\alpha, \uparrow\rangle = |\beta, \uparrow\rangle$ and $P|\beta, \uparrow\rangle = |\alpha, \uparrow\rangle$. The sketch of $|\alpha\rangle$ and $|\beta\rangle$ wavefunctions in NLS is shown in Fig. 4(d). Physically, $|\alpha, \uparrow\rangle$ and $|\beta, \uparrow\rangle$ stand for states with the wavefunction majorly localized around B1 and B2 in the unit cell. By transforming $M_5\tau^y$ to the new basis, we see that it represents a staggered Zeeman field that points in the $+z(-z)$ direction on $|\alpha\rangle$ and $|\beta\rangle$. Since $|\alpha\rangle$ and $|\beta\rangle$ majorly overlaps with Mn1 and Mn2, respectively. This staggered Zeeman field is exactly generated by the AFM order on Mn atoms, with electron spins point along opposite $z$ directions on Mn1 and Mn2. Thus the AFM amplitude excitations further induce fluctuations of $\theta(x,t)$. The parameters are determined from first-principles calculations [57]. We notice that $M_0 < 0$ and $M_1, M_2 > 0$, which correctly characterizes the band inversion around $\Gamma$ point. In the nonmagnetic state, $M_5(k) = A_3 = 0$, the system is a TI with a single gapless Dirac surface state. The direct consequence of $M_5(k)$ term is to open a gap of $2B_0$ in the surface-state spectrum, which is independent of the surface orientation. The $\theta_0$ and $1/g$ for 001 AFM $X_2Bi_2Te_5$ are calculated in Fig. 4(c).

Experimental proposals. The DAF $\theta(x,t)$, once realized in $Mn_2Bi_2Te_5$, would induce a nonlinear magnetoelectric effect and can be measured by the nonlinear optical spectroscopy. A static magnetic field $B_0$ and an a.c. electric field $E(t) = E_{ac}\sin(\omega t)$ would excite $\delta\theta(x,t) \propto \sin(\omega t)E_{ac} \cdot B_0$, which further induces a topological magnetization $M_i \propto \theta_0 + \delta\theta(x,t) B \times (a_1\sin(\omega t) + a_2\cos(2\omega t))$. The double frequency $2\omega$ is from the DAF [8]. The Néel order may also induce another harmonic generation. The magnetic field, wavelength and temperature dependence would in principle to distinguish the DAF from static AFM order.

$\theta(t)$ can also be excited by the AFM resonance [64]. For example, a static magnetic field is applied along easy $z$ axis $B = B\hat{z}$ and a microwave is irradiated. The biparticle AFM spins are precessing around $B \hat{z}$, which further induces $\sin(\omega t)E_{ac} \cdot B \times (a_1\sin(\omega t) + a_2\cos(2\omega t))$. In a mean field theory of an interacting Hamiltonian, $M_5 = -(2/3)Um^2$, where $U$ is the on-site repulsion [8]. Therefore, $\delta m^2(t)$ induces $\delta\theta(t)$, and from Eq. (2) one expects the dynamic CME with current $J(t) = -(e^2U)/(3\pi gh)B\sum_{j=\pm} \omega_j \delta m_j^2 \sin(\omega_j t)$. It is worth mentioning that MnBi2Te4, albeit a slightly different material, is an axion insulator with static $\theta = \pi$ [21]. Though AFM resonance could induce AFM spins precessing, as long as the surface-states remain magnetically gapped, $\theta$ is still static to be $\pi$ due to conserved $P$. Therefore no dynamical CME is expected to exist in MnBi2Te4. In summary, the criterion proposed here can be served as a guiding principle for searching DAF. A smoking gun signature is the dynamical CME from a time-dependent axion field in insulators induced by AFM resonance. The dynamical axion state predicted here may help to search for the elementary dark axion particle in high energy physics [65].

FIG. 4. (a) The evolution of energy gap at $\Gamma$ with $\lambda$. (b) The evolution of energy gap at $\Gamma$ with the doping strength. $x$ denotes the element substitution of Bi/Sb and Te/Se. For $x < x_c$ (minimum gap), the system has band inversion. (c) The static $\theta_0$ and $g$ of $X_2Bi_2Te_5$ vs $\lambda$, which are calculated from the effective $k \cdot p$ models. (d) Sketch of the wavefunction of states $|\alpha\rangle$ and $|\beta\rangle$ in each unit cell.
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