Magnetoelectric Effect in Topological Insulator Films beyond Linear Response Regime

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We study the response of topological insulator films to strong magnetic and electric fields beyond the linear response theory. As a model, we use three-dimensional lattice Wilson-Dirac Hamiltonian where we simultaneously introduce both magnetic field through the Peierls substitution and electric field as a potential energy depending on lattice coordinate. We compute the electron energy spectrum by numerically diagonalizing this Hamiltonian and obtain quantized magnetoelectric polarizability. In addition, we find that the magnetoelectric effect vanishes as the film width decreases, due to the hybridization of surface wavefunctions. Furthermore, applying gate voltage between the surfaces, we observe several quantized plateaus of θ term, which are mainly determined by the Landau level structures on the top and bottom surfaces.

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Introduction.— After a recent discovery of topological insulators (TIs) [1,2], the quest for numerous fascinating effects in response to external perturbations in TIs has started. One of them is the magnetoelectric (ME) effect, which is a long standing issue in multiferroics [3], and has been recently introduced in TIs [1,2,4–8]. The essence of the ME effect is that the external electric or magnetic fields induce magnetization or polarization in a TI, respectively. From the effective topological field theory predictions [4], the ME effect in three-dimensional (3D) TIs can be described by introducing a new term $U_\theta$ in the energy density for the axion electrodynamics [1,2,4–12],

$$U_\theta = -\frac{e^2}{4\pi^2\hbar c}\theta E \cdot B,$$  \hspace{1cm} (1)

where $E$ and $B$ are the electric and magnetic fields respectively. The $\theta$ term, $U_\theta$ [10], characterizes the 3D TIs. In Eq. (1), parameter $\theta$ takes values $\pm \pi$ for TIs, whereas it is 0 for ordinary insulators. The magnetization $M$ and electric polarization $P$ are obtained from the energy density as follows

$$M = -\frac{\partial U_\theta}{\partial B} = \frac{e^2}{4\pi^2\hbar c}\theta E,$$ \hspace{1cm} (2)

$$P = -\frac{\partial U_\theta}{\partial E} = \frac{e^2}{4\pi^2\hbar c}\theta B,$$ \hspace{1cm} (3)

and clearly show the cross-correlated responses [5,12,15].

In a strong magnetic field which breaks time reversal symmetry, however, still a controversy remains in the literature regarding the quantization of ME responses. According to Ref. [8], which considers only TI surface states, the response should be quantized: $\theta/\pi = 2N + 1$ ($N$ being an integer) when the electron densities on the top and bottom surfaces are balanced, i.e. no gate voltage is applied. On the other hand, Ref. [11], considering only bulk states in a magnetic field, concludes that $\theta$ is not quantized but rather arbitrary.

In this Letter, to resolve this controversy, we study the effect of an ultrahigh magnetic field on TIs. To fully treat both bulk and surfaces, we consider a lattice Hamiltonian [16,17] for a 3D TI in a slab geometry, sandwiched between two thin ferromagnets with magnetizations pointing in the opposite directions along $z$ axis, see Fig. 1. The role of ferromagnetic films can also be played by an appropriate doping of the TI with magnetic impurities. We first reproduce the $\theta$ term [1] with $\theta = \pm \pi$, in the balanced case, from the total energy of electrons. This means that the axion electrodynamics remains unchanged as long as the bulk gap is not destroyed, even though time-reversal symmetry which plays essential role in TIs, is broken by the magnetic field. However, as the TI thickness is decreased, we observe that the ME effect starts to vanish. Based on this result, we estimate the critical thickness of TI film capable of displaying the robust ME effect, which can be useful for future experimental studies, especially in the view of recent experimental progress on TI thin films [18,19]. Moreover, we also study the case of both strong electric and magnetic fields. We show that the response to a small change in the electric field can be characterized in this regime by different $\theta$ which are still quantized as long as the Fermi level resides within the bulk gap. We show that the quantization rule of $\theta$ is determined by the Landau level structures on the top and bottom surfaces, while the bulk states do not play an important role in thick TI films.

Model.— To study the ME effect in TIs, we employ the...
Wilson-Dirac tight binding Hamiltonian for 3D lattice TI model \( \text{16 17} \) which is a simplified version of effective 4-band model Hamiltonian for newly discovered 3D TIs such as Bi\(_2\)Se\(_3\), Bi\(_2\)Te\(_3\), and Sb\(_2\)Te\(_3\) \( \text{20 22} \). We introduce external magnetic field \( \mathbf{B} \), applied in \( z \) direction, through the Peierls substitution and set its magnitude \( B \) so that the magnetic flux going through a single plaquette is a rational fraction of the magnetic flux quantum. In other words \( B = (p/q)\Phi_0/a^2 \), where \( p \) and \( q \) are natural numbers with \( p < q, \Phi_0 = hc/e \) is the magnetic flux quantum, and \( a \) is the lattice constant. For convenience we set \( e = c = h = a = 1 \) in the rest of the paper so the magnetic field becomes \( B = 2\pi p/q \). Using Landau gauge \( \mathbf{A} = (0, Bx, 0) \), the Hamiltonian is defined as follows:

\[
H_0 = -\frac{1}{2} \sum_{\mu=x,y,z} (T^\mu + T^{\mu\dagger}) + (m + 3r) \sum_{\mathbf{R}} c^\dagger_{\mathbf{R}} \beta c_{\mathbf{R}}. \tag{4}
\]

Here the translation operators in \( \mu = x, y, z \) directions are

\[
T^\mu = \sum_{\mathbf{R}} c^\dagger_{\mathbf{R} + a_\mu} e^{iA_\mu}(r\beta - it\alpha_{R})c_{\mathbf{R}}, \tag{5}
\]

where \( t \) and \( r \) are the hopping parameters with and without spin flip, respectively, \( m \) is the mass term characterizing the spin-orbit interaction within TI, \( \mathbf{R} = (n_x, n_y, n_z) \) is the lattice coordinate, \( a_\mu \) is the unit lattice vector in \( \mu \)-direction, and \( \alpha_{R}, \beta \) are the standard Dirac matrices:

\[
\alpha_{R} = \begin{pmatrix} 0 & \sigma_\mu \\ \sigma_\mu & 0 \end{pmatrix}, \quad \beta = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix},
\]

where \( \sigma_\mu \) are the Pauli matrices.

To take into account the effect of an external electric field and the interaction of electron spins on top and bottom TI surfaces with the magnetization, we introduce additional terms \( H_E \) and \( H_s \), respectively. Then the total Hamiltonian is

\[
H = H_0 + H_E + H_s, \tag{6}
\]

\[
H_E = \sum_{\mathbf{R}} c^\dagger_{\mathbf{R}} U(n_z) \begin{pmatrix} I & 0 \\ 0 & I \end{pmatrix} c_{\mathbf{R}}, \tag{7}
\]

with the potential energy \( U(n_z) = -E(n_z - L_z/2) \), where \( L_z \) is the TI film thickness. The surface spin contribution, see Fig.\text{[1]} is given by

\[
H_s = \sum_{\mathbf{R}} c^\dagger_{\mathbf{R}} b(n_z) \Sigma^+_{\mathbf{R}} c_{\mathbf{R}}, \quad b(n_z) = \begin{cases} b_s, & n_z = L_z; \\ -b_s, & n_z = 0. \end{cases} \tag{8}
\]

Here \( \Sigma^+ = \text{diag}(\sigma_z, \sigma_z) \) and \( b_s \) is the surface spin constant.

**Discussion.** We investigate the magnetoelectric effect by calculating the energy eigenvalues of electrons. Since our model has translational invariance in \( x \) and \( y \) directions, Eq.\text{[6]} can be written in the form \( \text{23} \)

\[
H = \sum_{k_x, k_y, n_z} c^\dagger_{k_x, k_y, n_z} \mathcal{H}_{n_z, n_z'}(k_x, k_y)c_{n_z'}(k_x, k_y), \tag{9}
\]

where \( \mathcal{H}_{n_z, n_z'}(k_x, k_y) = 4q(L_z + 1) \times 4q(L_z + 1) \) matrix. By numerically performing exact diagonalization of this Hamiltonian matrix, we solve the eigenvalue equation \( \mathcal{H}(k_x, k_y)\lambda|\lambda, k_x, k_y \rangle = \lambda(k_x, k_y)|\lambda, k_x, k_y \rangle \), the magneto-electric effect is described by the energy density of electrons:

\[
U_{\text{tot}} = \frac{1}{V} \sum_{k_x, k_y} \sum_{\lambda} \lambda(k_x, k_y), \tag{10}
\]

where \( V \) is the TI volume and \( \sum_{\lambda} \) is the sum of eigenvalues below the Fermi level which is fixed at \( \epsilon_F = 0 \) in the following. In the presence of both electric \( \mathbf{E} \) and magnetic \( \mathbf{B} \) fields, \( U_{\text{tot}} \) has a term proportional to \( \mathbf{E} \cdot \mathbf{B} \) as expected from the axion electrodynamics, Eq.\text{[1]}.

The values of the energy density for a typical case are plotted in the inset of Fig.\text{[2]} For fixed magnetic field, we change the electric field and plot the energy density as a function of \( \mathbf{E} \cdot \mathbf{B} \). From Eq.\text{[1]}, we estimate the parameter \( \theta \) by \( \theta = -4\pi^2 \tan \gamma \), where \( \tan \gamma \) is the slope of the linear dependence in the inset of Fig.\text{[2]}. In all our calculations, we set the hopping parameters \( t = r = 1 \), the mass \( m = -1 \), and choose the number of layers in \( x \)-direction \( L_z \), so that the number of points in \( k_x \)-direction of the magnetic Brillouin zone is \( L_z/q = 32 \).

First, we study \( \theta \) as a function of TI thickness \( L_z \) for the fixed magnetic field \( \mathbf{B} = \pi/5 \) corresponding to \( p = 1 \) and \( q = 10 \), see the main panel of Fig.\text{[2]} The data shows that \( \theta \) is close to \( \pi \) at large \( L_z \), whereas it decreases notably below \( L_z \approx 10 \). As \( L_z \) approaches 1, the parameter \( \theta \) almost vanishes which means that in very thin TI films the ME effect disappears. When the TI thickness is very small, it is expected that the TI surface wavefunctions hybridize with each other \( \text{5 24} \). To confirm it, we calculate spacial profiles of the wavefunctions in \( z \)-direction for the surface states. At the \( \Gamma \) point \( (k_x = k_y = 0) \) they are calculated for several values of \( L_z \) and shown in Fig.\text{[3]}. We observe that for \( L_z = 30 \) the wavefunctions are strongly localized near the TI surfaces, whereas overlap of these wavefunctions increases.
with decreasing TI thickness. At \( L_z = 4 \) the top and bottom wavefunctions almost completely overlap. This clearly indicates that the reduction of the ME effect is associated with the hybridization of the surface wavefunctions. From Fig. 2 the value of \( L_z \) below which the ME effect start to significantly diminish can be estimated as \( \sim 10 \). For typical TIs \( [20–22, 25, 26] \) this value corresponds to \( \sim 10 \) nm.

So far we have considered a TI in a weak electric field. Experimentally, however, ultrahigh electric fields can be achieved by ionic liquid gating \([18]\), and it is therefore interesting to study a nontrivial response of a TI to both strong electric and magnetic fields. The presence of a strong magnetic field indicates that Landau levels form on top and bottom TI surfaces. The existence of well-defined Landau levels in TIs has been confirmed by recent experimental observations of the quantum Hall effect \([19, 27]\). In our simulation, we fix for simplicity the Landau levels on the bottom surface, whereas the gating induced electric field, \( E_g \), shifts the energy levels on the top surface. As the gate voltage changes from \( V_g \equiv E_g L_z = 0 \), Landau level energies on the top surface move. Figure 4(a) shows that Landau levels cross the Fermi level at the following gate voltages: \( V_{−5}, V_{−4}, V_{−3}, \ldots, V_5 \). In the nonlinear regime of strong electric or magnetic fields, the axion electrodynamics corresponding to \( \theta = \pm \pi \) in general is not expected to be applicable. To probe this regime we vary the electric field by small \( \delta \mathbf{E} \) near \( E_g \), i.e. the total electric field is \( \mathbf{E} = E_g + \delta \mathbf{E} \), and study the response to \( \delta \mathbf{E} \cdot \mathbf{B} \). Then \( \delta \mathbf{E} \) has a role of \( \mathbf{E} \) in Eq. (1) and \( \theta(E_g, B) \) is defined as the overall coefficient describing the ME effect response.

Figure 4(b) shows \( \theta/\pi \) as a function of \( V_g \). One can see that there are several plateaus where \( \theta/\pi \) is approximately quantized in integers \( \{-4, −3, −2, −1, 0, 1, 2, 3, 4, 5\} \). Every new quantization plateau in Fig. 4(b) occurs at the points where Landau levels cross the Fermi level. The exact quantization would be in agreement with the prediction of Ref. [8] for weak electric fields. As we show below, the deviation from the exact quantization is mostly attributed to the asymmetry of TI surface wavefunctions due to the application of the large gate voltage. Thus, Fig. 4(b) shows an important fact that although time-reversal symmetry is broken in a 3D TI by a strong magnetic field, \( \theta \) is not arbitrary as it has been claimed in Refs. [11] and [12], but rather quantized as long as the Fermi level is still within the bulk gap. In accord with this, the two continuous regions of the dependence in Fig. 4(b) at the left and right edges correspond to the cases when the Fermi level enters continuous (valence or conduction) bands. In the two continuous regions, however, our results only qualitatively show the behavior of the parameter \( \theta \), because we do not consider any screening effects coming from conduction and valence bands that may suppress the \( \theta \)-term, and also because beyond the range bounded by \( V_{−5} \) and \( V_5 \), see Fig. 4(a), there are multiple crossings of Landau levels very close to each other.

To investigate the hypothesis that the lack of exact quantization in \( \theta \) is attributed to the asymmetry of TI surface wavefunctions due to gating, we repeat the same calculation for a thicker TI film. The details of the quantization of \( \theta \) as a function of gate voltage \( V_g \) for two different TI film thicknesses.
nless are shown in Fig. 5. The results for $L_z = 10$ are shown by blue circles and for $L_z = 20$ by red triangles. At a fixed gate voltage $V_g (= E_g L_z)$, applied electric field for $L_z = 10$ case is two times stronger than that for $L_z = 20$. The comparison shows that when the TI thickness $L_z$ increases 2 times the slope of each quantization plateau becomes 2 times smaller. This indicates that for thick enough TI films the plateaus should become horizontal, because in this case the asymmetry of surface wavefunctions can be disregarded since their overlap is too small.

Finally, we study the effects of bulk magnetic interaction on the ME effect. The Zeeman type interaction between the electron spin and applied magnetic field is described by

$$\mathcal{H} = \mu_B B_z \begin{pmatrix} g_1 \sigma_z & 0 \\ 0 & g_2 \sigma_z \end{pmatrix},$$

where $g_1$ and $g_2$ are the effective $g$-factors depending on the TI materials. In most of cases, for materials such as Bi$_2$Se$_3$ and Bi$_2$Te$_3$, $g_1$ and $g_2$ have opposite signs [17]. Therefore as a simplified model we consider the case of $g_2 = -g_1$. In addition, we study the influence of the exchange interaction between the electron spins and magnetic impurities that can be introduced by the appropriate doping of the TI bulk. Within the virtual crystal approximation, this interaction has the form

$$\mathcal{H}' = J \mathbf{M} \begin{pmatrix} \sigma_z & 0 \\ 0 & \sigma_z \end{pmatrix},$$

where $J$ is the exchange interaction constant and $\mathbf{M}$ is the mean value of magnetic moments in the bulk. These two interactions, Eqs. (11) and (12), can be covered by two simple model Hamiltonians: $\mathcal{H}' = b_0 \Sigma^\pm$ with $\Sigma^\pm = \text{diag}(\sigma_z, \pm \sigma_z)$ and $b_0$ being the bulk spin constant. The sign “$+$” in $\Sigma^+_z$ indicates bulk electrons coupling with magnetization, while “$-$” sign indicates bulk electrons coupling with the external magnetic field.

The calculation results for magnetic field $B = \pi/10$ corresponding to $p = 1$ and $q = 20$ are shown in Fig. 6 where the red and blue curves correspond to $\Sigma^+_z = \text{diag}(\sigma_z, -\sigma_z)$ case, red is for $L_z = 15$ and blue is for $L_z = 10$. The black and green curves correspond to $\text{diag}(\sigma_z, \sigma_z)$ case, with the black for $L_z = 15$ and green for $L_z = 10$. One can see from Fig. 6 that $\theta/\pi = 1$ persists approximately up to $b_0 = 1$ which corresponds to the TI regime. At $b_0 = |m| = 1$, the system crosses to the Weyl semimetal regime [28, 29] for $\text{diag}(\sigma_z, \sigma_z)$ case and therefore no quantization of $\theta$ beyond $b_0 = 1$ is expected [30]. These results show remarkable robustness of the ME effect in TI phase even for very strong magnetic fields or high concentration of magnetic impurities that affect the bulk structure of the TI. It seems to be irrespective of microscopic details of coupling with the magnetic impurities or field, since it works equally well in TI regime for the toy models with both $\pm$ signs.

Summary.-- We have shown that the ME effect in TIs survives even in both strong magnetic and electric fields even though time-reversal symmetry is broken. We have described the ME effect beyond the linear response regime and have confirmed that it is robust but takes a fascinating form of multiple quantization related to Landau levels crossing of the Fermi level. In addition, the influence of the TI surface hybridization has been studied in very thin TI films, describing how the ME effect vanishes in the limit of very thin TIs. Furthermore, the ME effect is shown to persist even for strong bulk interactions with magnetic field or magnetic impurities.

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Supplementary material for “Magneto-electric Effect in Topological Insulator Films beyond Linear Response Regime”

Model Hamiltonian in momentum space

We present below the details of diagonalization of Wilson-Dirac Hamiltonian \[16, 17\] given by Eqs. (4) and (5) in the main text:

\[
H_0 = -\frac{1}{2} \sum_{\mu=x,y,z} (T_\mu + T_\mu^\dagger) + (m + 3r) \sum_{\mathbf{R}} c_{\mathbf{R}+\mathbf{a}_\mu}^\dagger c_{\mathbf{R}} \beta c_{\mathbf{R}}. \tag{13}
\]

\[
T_\mu = \sum_{\mathbf{R}} c_{\mathbf{R}+\mathbf{a}_\mu}^\dagger e^{i A_\mu} (r \beta - i t \alpha_\mu) c_{\mathbf{R}}. \tag{14}
\]

We use a slab geometry with periodic boundary conditions in \(x\)- and \(y\)-directions and fixed boundary condition in \(z\)-direction, \(0 \leq n_z \leq L_z\) where \(L_z + 1\) is the number of lattice sites in \(z\)-direction. According to our choice of the magnetic field and Landau gauge, there is \(q\) periodicity in the Aharonov-Bohm (AB) phase, i.e. the lattice sites whose \(x\)-coordinate is \((\text{mod}\ q)\) have the same AB phase. Therefore, we can define a new unit cell that is \(q\) times larger than the original one by introducing an additional coordinate \(s\),

\[
n_x = q n'_x + s, \quad 1 \leq s \leq q, \tag{15}
\]

where \(n'_x\) is a coordinate of a one-dimensional unit cell in \(x\)-direction, containing \(q\) lattice points and \(s\) is the coordinate of the lattice site within the unit cell. As a result, the lattice can be represented by \(\mathbf{R} = (s, n'_x, n_y, n_z)\), where \((n'_x, n_y, n_z)\) corresponds to a coordinate of a 3D unit cell and \(s\) corresponds to a coordinate of a lattice point within that unit cell.

Because of periodicity in \(x\)- and \(y\)-directions, it is convenient to introduce a new annihilation (and corresponding creation) operator \(c_{s,n'_x}(k)\) by means of Fourier transformation:

\[
c_{s,n'_x,n_y,n_z} = \sqrt{\frac{q}{L_x L_y}} \sum_{k_x,k_y} e^{i k_x q n'_x} e^{i k_y q n_y} c_{s,n'_x}(k), \tag{16}
\]

where \(L_x\) and \(L_y\) are the numbers of layers in \(x\)- and \(y\)-directions, respectively, and \(k = (k_x, k_y)\) is the wavevector in \(xy\)-plane. In Eq. (9) of the main text we omitted for brevity the subscript \(s\) in \(c_{s,n'_x}(k)\). In terms of these operators the Hamiltonian, Eq. (13), becomes

\[
H_0 = \sum_{\mathbf{k}} c^\dagger(\mathbf{k}) \mathcal{H}_0(\mathbf{k}) c(\mathbf{k}), \tag{17}
\]

where \(c^\dagger(\mathbf{k}) = (c_{10}^\dagger, c_{11}^\dagger, \ldots, c_{q_0}^\dagger, c_{q_1}^\dagger, \ldots, c_{qL_z}^\dagger)\) and the matrix \(\mathcal{H}_0(\mathbf{k})\) is

\[
\mathcal{H}_0(\mathbf{k}) = \begin{pmatrix}
\Delta_1 & \Xi^\dagger & \Omega \\
\Xi & \Delta_2 & \Xi^\dagger \\
\Omega^\dagger & \Xi & \Delta_q
\end{pmatrix}. \tag{18}
\]

Here the diagonal block elements are

\[
\Delta_\lambda = \begin{pmatrix} a_\lambda & b^\dagger \\ b & a_\lambda & b^\dagger \\ \vdots & \ddots & \ddots & \ddots \\ b & \cdots & b & a_\lambda \end{pmatrix}
\]

with \(b = (i t \alpha_\lambda - r \beta)/2\) and \(a_\lambda = t \sin(k_y - 2\pi p \lambda/q) \alpha_y + (m + 3r - r \cos(k_y - 2\pi p \lambda/q)) \beta\). While the nonzero off-diagonal block elements of \(\mathcal{H}_0(\mathbf{k})\) are \(\Xi = (i t \alpha_x - r \beta) I_{L_z+1}/2\) and \(\Omega = (i t \alpha_x - r \beta) e^{-i k_x q} I_{L_z+1}/2\) where \(I_{L_z+1}\) is \((L_z + 1) \times (L_z + 1)\) unit matrix.

To include the external electric field \(H_E\) as well as surface spin interaction with magnetization \(H_s\) terms, we apply the same transformation given by Eq. (16). Then the total model Hamiltonian, \(H = H_0 + H_E + H_s\), is represented as \(\sum_\mathbf{k} c^\dagger(\mathbf{k}) \mathcal{H}(\mathbf{k}) c(\mathbf{k})\) with

\[
\mathcal{H}(\mathbf{k}) = \begin{pmatrix}
\Delta_1 + \Lambda & \Xi^\dagger & \Omega \\
\Xi & \Delta_2 + \Lambda & \Xi^\dagger \\
\Omega^\dagger & \Xi & \Delta_q + \Lambda
\end{pmatrix}. \tag{19}
\]

Here the electric field and overall spin contribution coming from both TI surfaces are given by matrix \(\Lambda = \text{diag}(g_0, g_1, \ldots, g_{L_z})\) with \(g_n = U(n_z) I_4 + b(n_z) \Sigma_n^x\) where \(I_4\) is \(4 \times 4\) unit matrix.