Topological magneto-electric effects in thin films of topological insulators

Takahiro Morimoto,1 Akira Furusaki,2,1 and Naoto Nagaosa1,3

1RIKEN Center for Emergent Matter Science (CEMS), Wako, Saitama, 351-0198, Japan
2Condensed Matter Theory Laboratory, RIKEN, Wako, Saitama, 351-0198, Japan
3Department of Applied Physics, The University of Tokyo, Tokyo, 113-8656, Japan

(Dated: August 12, 2015)

We propose that the topological magneto-electric (ME) effect, a hallmark of topological insulators (TIs), can be realized in thin films of TIs in the $\nu = 0$ quantum Hall state under magnetic field or by doping two magnetic ions with opposite signs of exchange coupling. These setups have the advantage compared to previously proposed setups that a uniform configuration of magnetic field or magnetization is sufficient for the realization of the topological ME effect. To verify our proposal, we numerically calculate ME response of TI thin films in the cylinder geometry and that of effective 2D models of surface Dirac fermions. The ME response is shown to converge to the quantized value corresponding to the axion angle $\theta = \pm \pi$ in the limit of the large top and bottom surface area of TI films, where non-topological contributions from the bulk and the side surface are negligible.

I. INTRODUCTION

Topological aspects of the condensed matter systems have been a subject of intensive studies since the discovery of the quantum Hall effect (QHE) [1, 2]. The topological nature of the QHE is encoded in the Hall conductance, which is exactly quantized in units of $e^2/h$ to the Chern number characterizing the global topology of wave functions constituting a manifold in the Hilbert space. The Hall current supporting the quantized Hall conductance flows through the edge channels along the boundary of the sample (bulk-edge correspondence).

The extension of the QHE to time-reversal symmetric systems is realized in topological insulators (TIs) [3, 4]. Here, the $\mathbb{Z}_2$ topological index characterizes the global topology of wave functions of Kramers pairs. Helical edge channels and surface Dirac fermions exist on the boundary of two-dimensional (2D) and three-dimensional (3D) TIs, respectively, as a consequence of nontrivial topology. The spin Hall conductance of 2D TIs is an analog of the Hall conductance in the QHE. However, the spin current is not conserved in the presence of spin-orbit interactions which are indispensable for TIs; the spin Hall conductance is neither a well-defined nor quantized quantity. The charge conductance along the helical edge channels in 2D TIs was shown to be quantized at $2e^2/h$ [5], but the quantization is not so accurate as in the QHE. Furthermore, one cannot relate the charge conductance to the $\mathbb{Z}_2$ topological index in 3D TIs. The quantized quantity characteristic of TIs, corresponding to the Hall conductance in the QHE, has been looked for.

The topological magneto-electric (TME) effect [6–15] has been suggested as a phenomenon that is directly related to the $\mathbb{Z}_2$ topological index for 3D TIs. The TME effect is a quantized response of polarization to applied magnetic fields or a quantized response of magnetization to applied electric fields [6, 7]. The quantized polarization current was proposed to be accessible by transport measurements [6–8]. Alternatively, the TME effect was proposed to be observed by optical measurements of Faraday or Kerr rotation showing a quantized response [10, 11]. However, no experimental observation of the TME effect has been reported so far.

The TME effect of a 3D TI is theoretically described by the axion action (with $c = 1$),

$$ S = \frac{\theta e^2}{32\pi^2 \hbar} \int dt d^3 x \epsilon^{\mu \nu \lambda} F_{\mu \nu} F_{\lambda \delta} = \frac{\theta e^2}{4\pi^2 \hbar} \int dt d^3 x E \cdot B, $$

(1)

where the angle $\theta$ is equivalent to the $\mathbb{Z}_2$ index: $\theta = \pi$ (mod $2\pi$) for TIs and $\theta = 0$ (mod $2\pi$) for trivial insulators [6]. Transforming Eq. (1) to the integral over the surface of a 3D TI yields the Chern-Simons action,

$$ S_{CS} = \frac{\theta e^2}{8\pi^2 \hbar} \int dt d^2 x \epsilon^{\mu \nu \lambda} A_\mu \partial_\nu A_\lambda, $$

(2)

which describes the QHE on the TI surface with the Hall conductance $\sigma_{xy} = (\theta/2\pi)(e^2/h)$. For TIs with $\theta = \pi$, this amounts to $\sigma_{xy} = e^2/2h$. Microscopically, the half-integer QHE ($\sigma_{xy} = \pm e^2/2h$) on the TI surface occurs once surface Dirac fermions acquire a mass and have the Hamiltonian

$$ H = v_F (-p_x \sigma_y + p_y \sigma_x) + M \sigma_z, $$

(3)

where $v_F$ is the Fermi velocity. The mass gap $M$ of the surface Dirac fermions can be introduced, for example, through the exchange coupling $J$ of Dirac fermions to the magnetization $m$ (pointing perpendicular to the surface) of doped ferromagnetic ions: $M = Jm$. In order to realize the TME effect described by Eqs. (1) and (2), two conditions must be satisfied. (i) The magnetization points outward or inward over the whole surface of a 3D TI such that surface Dirac fermions are fully gapped on any surface. (ii) The Fermi energy must be tuned to be inside the gap induced by the magnetization $m$. This magnetic configuration is very difficult to realize experimentally since the external magnetic field favors the same magnetization direction for both top and bottom surfaces. In addition, the fine tuning of the Fermi energy is still very difficult experimentally although some groups...
have succeeded [16, 17]. We note in this regard that localization effect can relax the second condition. That is, when the surface Dirac fermions are localized by magnetic impurities, the Fermi energy can be set in the wider energy window of localized states [14]. Furthermore, the magneto-electric cooling effect can help the magnetization \( m \) align perpendicular to any surface because of the energy gain proportional to the volume which eventually overcome the surface energy [14]. However, there is no experimental study to pursue this possibility up to now.

In this paper we propose alternative routes to realizing the TME effect in thin films of 3D TIs by either applying external magnetic field or doping two kinds of magnetic impurities. The first approach utilizes the QHE of TI thin films [18–20]. In particular, we are interested in the novel \( \nu = 0 \) QHE which was recently observed in the presence of a finite potential difference between the top and bottom surfaces [20]. In this case, the edge channels are gapped on the side surfaces due to the finite thickness effect, which can produce interesting spintronics functions [21]. We point out that this \( \nu = 0 \) quantum Hall system provides an ideal laboratory to realize the TME effect, as discussed in detail below. We also propose that the TME effect can be achieved by doping Cr and Mn to upper and lower halves of TI thin films, respectively.

II. TOPOLOGICAL MAGNETO-ELECTRIC EFFECTS IN TI THIN FILMS: BASIC IDEA

The TME effect described by the actions in Eqs. (1) and (2) predicts that charge polarization \( P \) is induced by applying a magnetic field \( B \),

\[
P = \frac{\theta}{2\pi} \frac{e^2}{\hbar} |B|.
\]

where \( \theta = \pi \) (mod 2\( \pi \)). In this section we explain how this quantized response of charge polarization is obtained for the proposed two setups of TI thin films by discussing charge response of quantum Hall states of Dirac fermions at the top and bottom surfaces.

Let us consider a thin film of a TI in the magnetic field \( B = (0, 0, B) \) which is applied perpendicular to the film; see Fig. 1(a). In this case, Dirac fermions at the top and bottom surfaces form Landau levels (LLs). For simplicity we assume that the Fermi energy is at the Dirac point, \( E_F = 0 \). In this case, important low-energy excitations are states in the LLs at the Fermi energy on the top and bottom surfaces, while the excitations are gapped in the bulk and at the side surface (the latter due to the finite-thickness effect). In addition, we introduce a potential energy difference between the top surface \( (U_0 > 0) \) and bottom surface \( (-U_0) \). For small magnitude of the potential asymmetry \( U_0 \), the LLs at the top and bottom surfaces are occupied up to \( N_T = -1 \) and \( N_B = 0 \) LLs, respectively, as indicated in Fig. 1(a), where \( N_T \) and \( N_B \) are LL indices on the top and bottom surfaces. This situation realizes the \( \nu = 0 \) QHE which was experimentally observed in Ref. [20]. More generally, the \( \nu = 0 \) QHE is realized in TI thin films when LLs at the top and bottom surfaces are filled up to \( N_T = -N - 1 \) and \( N_B = N \) LLs with any integer \( N \), where \( N \) can be controlled by changing the potential asymmetry \( U_0 \). Then the charge densities at the top and bottom surfaces \( (n_T \) and \( n_B) \) become

\[
n_T = \left( N + \frac{1}{2} \right) \frac{e^2}{\hbar} |B|, \quad n_B = -\left( N + \frac{1}{2} \right) \frac{e^2}{\hbar} |B|.
\]

If the top and bottom surfaces are located at \( z = d \) and \( z = -d \), respectively, the charge polarization along the \( z \) direction is given by

\[
P = \frac{1}{2d} |dn_T + (-d)n_B| = \left( N + \frac{1}{2} \right) \frac{e^2}{\hbar} |B|.
\]

The polarization in the \( \nu = 0 \) QHE coincides with the value expected from the TME effect, Eq. (4) with \( \theta = (2N + 1)|\pi| \), for \( B > 0 \). For \( B < 0 \), the axion angle is given by \( \theta = -(2N + 1)|\pi| \). This jump of \( \theta \) at \( B = 0 \) takes place because the surface bands become gapless at \( B = 0 \). In both cases, the axion angle takes a nontrivial value \( \theta = \pi \) modulo \( 2\pi \). We emphasize again that, in the setup with the \( \nu = 0 \) QHE, an arbitrary odd integer \( \theta/|\pi| = 2N + 1 \) can be realized by tuning the potential asymmetry \( U_0 \) with gating the top and bottom surfaces independently such that the highest occupied LLs are set to \( N_T = -N - 1 \) and \( N_B = N \).

Next we consider the second setup in which the upper and lower halves of a TI film are doped with two types...
of magnetic ions, e.g., Cr and Mn. Here a crucial assumption is that the two kinds of magnetic ions have exchange couplings \( J \) of opposite signs with Dirac fermions in the TI film. This is indeed the case with Cr and Mn, as can be seen from the fact that Cr-doped and Mn-doped Bi\(_2\)Te\(_3\)-based TIs show quantized anomalous Hall conductivity of opposite signs when the Fermi energy is tuned within the excitation gap of surface Dirac fermions [16, 17, 22]. Then a uniform magnetization depicted in Fig. 1(b) produces an exchange field pointing outward or inward at the top and bottom surfaces, and the desired constant Dirac mass \( Jm0\sigma \) in Eq. (3) is achieved except for the side surface. In the 3D geometry in Fig. 1(b), massive Dirac fermions show quantum anomalous Hall effect \( \sigma_{xy} = -\text{sgn}(Jm)e^2/2h \) and \( \sigma_{xy} = \text{sgn}(Jm)e^2/2h \) at the top and bottom surfaces, respectively. Since the electron density on each surface is given by \( n = \sigma_{xy}B \), applying a magnetic field induces the charge polarization

\[
P = \frac{1}{2d} \left\{ d \left[ -\text{sgn}(Jm)\frac{e^2}{2h} B + (-d)\text{sgn}(Jm)\frac{e^2}{2h} B \right] \right\} = -\text{sgn}(Jm)\frac{e^2}{2h} B. \tag{7}
\]

Thus the TME effect with the axion angle \( \theta = -\pi \text{sgn}(Jm) \) is realized with a uniform magnetization. Since Dirac fermions on the side surface are gapped because of the finite thickness, the TME effect should be observed when the chemical potential is tuned into the energy gap of the side-surface excitations. Compared with the original proposal with the non-uniform configuration of magnetization [14] which is experimentally difficult to realize, this setup has the advantage that it only requires a uniform magnetization, which facilitates experimental realization.

For both setups the charge polarization can be detected as a charge current in response to time-dependent \( B \).

### III. Non-Topological Contributions from the Bulk and the Side Surface

In general, the axion angle can deviate from \( \theta = 0, \pi \) (mod \( 2\pi \)) when both time reversal symmetry \( T \) and inversion symmetry \( P \) are broken. The \( \nu = 0 \) QHE is achieved in TIs by applying magnetic field, which breaks \( T \), and by inducing potential asymmetry between the top and bottom surfaces, which breaks \( P \). Thus the axion angle in the \( \nu = 0 \) QHE setup is not necessarily quantized.

In the previous section we have shown that polarization corresponding to the quantized value of \( \theta = \pi \) (mod \( 2\pi \)) is obtained when we neglect contributions from the bulk and focus on contributions from the top and bottom surfaces. While the charge polarization in the TME effect is accompanied by a flow of the surface current, non-topological contributions may also arise from the current flowing through the bulk. Furthermore, the side surfaces connecting the top and bottom surfaces in Fig. 1 may give rise to non-topological contributions. Then the natural question is whether charge polarization of TI thin films is really quantized as Eq. (4) predicts with the axion angle \( \theta = \pi \) (mod \( 2\pi \)) when contributions from the bulk and the side surface are taken into account. To address this question, we consider the 3D bulk of TI thin films and calculate the charge polarization and the current distribution induced by increasing \( B \).

Suppose that the TI sample has the cylinder geometry shown in Fig. 2, in which the top and bottom surfaces are located at \( z = \pm d \) and the side surface is at \( r = R_0 \). In order to describe the bulk electronic states of the TI film, we take a simple massive Dirac Hamiltonian

\[
H = H_0 + V, \quad H_0 = v_F(p \cdot \sigma)\tau_x + m(r)\tau_z, \tag{8}
\]

where \( p = (p_x, p_y, p_z) \) is the momentum, and \( \sigma = (\sigma_x, \sigma_y, \sigma_z) \) is the spin Pauli matrices, while \( \tau_x, \tau_z \) are the orbital Pauli matrices. We assume that the chemical potential \( \mu = 0 \). The Dirac mass (i.e., the bulk band gap) \( m(r) = m_{\text{in}} > 0 \) inside the TI film. The Dirac mass \( m(r) \) changes its sign at the surface, and \( m(r) = m_{\text{out}} < 0 \) outside the TI. In the setup of the \( \nu = 0 \) QHE in Fig. 1(a), \( V \) is given by

\[
V(r) = v_{Fe}A \cdot \sigma\tau_x + U(z), \tag{9}
\]

where \( A \) is the vector potential in the symmetric gauge \( A = \frac{1}{2}B(-y, x, 0) \), and the potential energy \( U(z) = U_0 \) for \( z \geq 0 \) and \( U(z) = -U_0 \) for \( z < 0 \). In the setup of a TI doped with Cr and Mn in Fig. 1(b), \( V \) is given by

\[
V(r) = M(z)\sigma_z, \tag{10}
\]

where \( M(z) = M_0 \) for \( z \geq 0 \) and \( M(z) = -M_0 \) for \( z < 0 \). In the cylindrical coordinates \((r, \varphi, z)\), wave functions are written as

\[
\psi(r, \varphi, z) = r^{-\ell/2}e^{(\ell - \frac{\varphi}{2})}\phi(r, z) \tag{11}
\]

for the angular momentum \( \ell \in \mathbb{Z} + \frac{1}{2} \). Then the Dirac equation is reduced to \([H_0(\ell) + V]\phi_\ell = E(\ell)\phi_\ell \), where

\[
H_0(\ell) = v_F \left( p_\ell \sigma_x + \frac{\hbar \ell}{r} \sigma_y + p_z \sigma_z \right) \tau_x + m(r)\tau_z, \tag{12}
\]

and \( \phi(r, z) \) obeys the Dirichlet boundary condition \( \phi = 0 \) at \( r = 0 \).

Small magnetic field \( \delta B(t) \) applied in the \( z \) direction (in addition to the constant magnetic field \( B \) for the \( \nu = 0 \) QHE case) induces a change in the polarization along the \( z \) direction that is proportional to \( \delta B(t) \). The change in the polarization leads to polarization current \( J \) proportional to \( \delta B(t)/dt \). From the linear response theory (see Appendix for details), the induced current density is written as

\[
\frac{\mathbf{J}(r_0)}{\delta B/dt} = \sum_{i \in O} \sum_{j \in U} \frac{2\hbar \text{Im} \left[ \langle \psi_i | j(r_0) | \psi_j \rangle \langle \psi_j | \mathcal{O} | \psi_i \rangle \right]}{(E_i - E_j)^2}, \tag{13}
\]
where \(i\) and \(j\) label occupied states \(\psi_i\) (\(i \in O\)) and unoccupied states \(\psi_j\) (\(j \in U\)) with energies \(E_i\) and \(E_j\), respectively, \(j(r_0) = -\epsilon v_F \delta(\hat{r} - r_0) \sigma_x\), and \(\mathcal{O} = \frac{\hbar}{2} \epsilon v_F (-y \sigma_x + x \sigma_y) \). 

The current density in Fig. 2 shows that the current flows along the surface of the TI film, not through the bulk. Thus the charge polarization in the present setup is indeed a topological phenomenon carried by the Dirac fermions on the surface of TIs. The surface current of topological nature is understood as follows. Time-dependent \(\delta B\) induces an electric field along the azimuthal (\(\varphi\)) direction. Then the Hall current flows along the \(r\)-direction on the top and bottom surfaces. The directions of the Hall current are opposite for the top and bottom surfaces, where \(\sigma_{xy} = \pm e^2/2h\) and \(\sigma_{x y} = \pm e^2/2h\), respectively. On the side surface, the current in the \(z\) direction flows to connect current distributions at the top and bottom surfaces, and it is exactly the polarization current along the \(z\) direction in the TME effect. From the discussions focusing on the top and bottom surfaces in the previous section, the polarization current is expected to be equal to the time derivative of \(P\) in Eq. (4) with the axion angle at \(\theta = \pi\) for the \(\nu = 0\) QHE from Eq. (6) with \(N = 0\), and at \(\theta = -\pi\) for the magnetically doped TI from Eq. (7) with \(Jm = M_0 > 0\). However, the axion angles obtained from the integration of the polarization current plotted in Fig. 2, 

\[
\theta = \frac{\hbar}{e^2} \pi \int r dr d\theta J_z (z = 0),
\]  

are found to be 0.4 and -0.3 for the \(\nu = 0\) QHE and the magnetically doped TIs, respectively, and both deviate from the quantized value \(\theta/2\pi = \pm \frac{\pi}{4}\). We attribute these deviations to finite-size effects of the surface of TI films, considering that the bulk contribution to the polarization current is negligibly small as shown in Fig. 2(a) and (b). Specifically, Dirac fermions on the side surface give non-topological contributions to the polarization current because of broken time-reversal and inversion symmetries.

**IV. ANALYSIS FROM SURFACE WEYL FERMIONS AND THE QUANTIZED AXION ANGLE**

The current distribution in Fig. 2 indicates that the polarization current is negligible in the gapped bulk and flows only at the surface of TI thin films. We now focus on low-energy Dirac fermions at the surface and study their contribution to the magneto-electric effect. In particular, we are interested in finite-size corrections which cause the deviation of \(\theta\) from the quantized value. For this purpose, we introduce an effective 2D model (Fig. 3) for surface Dirac fermions, in which surfaces of a 3D TI thin film are extended on the 2D \((X, Y)\) plane. The whole 2D surface has linear dimensions of \((2L_x + 4d)L_y\) and the \(y\) direction is assumed to be periodic. The original 3D coordinates \((x, y, z)\) of each surface are related to 2D...
coordinates \((X,Y)\) as follows:

\[
(x, y, z) = \begin{cases}
(-X + d + \frac{L_x}{2}, Y, -d), & \text{if } -L_x - d < X < -d, \\
(X - d - \frac{L_x}{2}, Y, d), & \text{if } d < X < L_x + d, \\
(\frac{L_x}{2}, Y, L_x + d - X), & \text{if } L_x + d < X, \\
(\frac{L_x}{2}, Y, -L_x - d - X), & \text{if } X < -L_x - d,
\end{cases}
\]

(15)

see Fig. 3.

For the \(\nu = 0\) QHE setup the effective Hamiltonian for the surface Dirac fermions is given by

\[
H = v_F \{ -p_X \sigma_y + [p_Y + eA_Y(X)]\sigma_x \} + U(X)\sigma_0.
\]

Here, we take the Landau gauge \(A_Y(X) = Bx(X)\). The potential difference between the top and bottom surfaces is given by the potential

\[
U(X) = \begin{cases}
U_0, & d < X < L_x + d, \\
-U_0, & -L_x - d < X < -d, \\
0, & \text{otherwise},
\end{cases}
\]

(17)

as schematically illustrated in Fig. 4(a). Since the 2D model is translationally invariant along the \(Y\) direction under the Landau gauge, the wave number \(k_Y(= k_y)\) is a good quantum number.

The polarization in the \(z\) direction is given by

\[
P = \frac{e}{2dL_x}z(X).
\]

(18)

We note that the spectrum of \(z\) is bounded in our 2D effective model so that the above equation is well defined. From the linear-response theory, the axion angle \(\theta\) determined from the charge polarization \(\theta/2\pi = h\delta P/e^2\delta B\) is given by

\[
\frac{\theta}{2\pi} = \frac{e}{2dL_x} \frac{h}{e^2} \int \frac{dk_y}{2\pi} \sum_{i \in O} \sum_{j \in U} \frac{2}{E_{i,k_y} - E_{j,k_y}} \text{Re} \langle \psi_{i,k_y} | z | \psi_{j,k_y} \rangle \langle \psi_{j,k_y} | eV_F x \sigma_x | \psi_{i,k_y} \rangle,
\]

(19)

where \(i\) and \(j\) label occupied states \(\psi_{i,k_y}\) (\(i \in O\)) and unoccupied states \(\psi_{j,k_y}\) (\(j \in U\)) with momentum \(k_y\) and energy \(E_{i,k_y}\) and \(E_{j,k_y}\), respectively. To compute Eq. (19) numerically, we discretize the 2D model. In order to avoid the fermion doubling on the lattice, we used a regularization scheme in the momentum space. Namely, we used the plane wave basis for the \(X\) direction labeled by \(k_x\) and set a cutoff to the wave number \(k_X\) as \(|k_X| < \Lambda\). The numerical results are obtained by extrapolating the cutoff \(\Lambda \to \infty\).

We show results for the axion angle computed from Eq. (19) in Fig. 4. The axion angle \(\theta/2\pi\) approaches \(1/2\) as \(L_x \to \infty\). This shows that non-topological contributions from the side surface become negligible as the ratio \(L_x/d\) is increased. Indeed the deviation of the computed axion angle \(\theta/2\pi\) from the quantized value \(1/2\) scales with the ratio of the areas of the side surface and the top/bottom surfaces, \(d/L_x\), as shown in Fig. 4(c). Thus the axion angle converges to the quantized value \(1/2\) in the limit where the top/bottom surfaces have much larger area than side surfaces. Since typical values of the thickness \(2d\) and the size \(L_x\) of TI films are \(2d = 100\) nm and \(L_x = 1\) mm [20], the axion angle \(\theta/2\pi\) is expected to show a quantized behavior in practice.

Next we move on to the 2D surface model for the mag-
netically doped TI. The effective Hamiltonian for the surface Weyl fermions is given by

\[ H = v_F (-p_X \sigma_y + p_Y \sigma_x) + M \cdot \sigma, \]

where we assumed no potential asymmetry between the top and bottom surfaces. The upper and lower halves of the TI film are doped with Cr and Mn, respectively. A uniform magnetization along the z direction induces effective exchange fields \( +M_0 \) and \( -M_0 \) along the z direction in the upper and lower halves of the TI film, respectively, because signs of exchange couplings are opposite for Cr and Mn. In the 2D model of surface Weyl fermions, these exchange fields are perpendicular to the top and bottom surfaces

\[ M(X) = (0, 0, M_0) \]

for \( |X \pm (L_x/2 + d)| < L_x/2 \) and produce a mass term \( M_0 \sigma_z \), while they are perpendicular to the side surface

\[ M(X) = (\pm M_0, 0, 0) \]

for \(-d < \pm X + L_x + d < 0 \) and \( 0 < \pm X < d \) and enter as a vector potential \( \pm M_0 \sigma_x \) in Eq. (20). This is schematically illustrated in Fig. 5(a). We numerically obtain the polarization and the axion angle using the linear response theory [Eq. (19)] in the same manner as in the \( \nu = 0 \) QHE case. The result is shown in Fig. 5. We again see asymptotic behavior \( \theta/2\pi \to -1/2 \) with \( L_x \to \infty \) for fixed \( d \). In a similar way to the case of the \( \nu = 0 \) QHE, the deviation of the axion angle from the quantized value \(-1/2\) scales with \( d/L_x \) as shown in Fig. 5(d). Thus there is a non-topological contribution proportional to the area of the side surface, but it diminishes in the thermodynamic limit \( L_x/d \to \infty \) where typical experiments of thin films are performed.

V. DISCUSSIONS

We have proposed that the TME effect should be observed in TI films in the \( \nu = 0 \) QHE and in magnetically doped TI films. We have shown that the polarization current in these setups are topological in that bulk contributions are negligible and show a quantized response in the limit of large top/bottom surface area. The non-topological contribution to the polarization current is proportional to the area of the side surface, which is interpreted as the axion angle \( \theta \) at the side surface deviating from the quantized value \( \pm \pi \) due to the breaking of time-reversal and inversion symmetries. For a thin-film sample of area 1mm\(^2\) and a \( B \)-field sweeping speed of 1T/s, the polarization current is estimated to be 20pA.

This magnitude of the polarization current is accessible in the current state of transport measurements [23]. Application of ac magnetic fields induces a polarization current oscillating in time in the \( z \)-direction. This ac polarization current can be detected, for example, with electrodes capacitively coupled to top and bottom surfaces and measuring currents between two electrodes as schematically shown in Fig. 6. Besides the corrections to the TME effect [Eq. (4)] due to broken \( T \) and \( P \) symmetries in our setup, there can be other corrections from other mechanisms, such as the coupling between top and bottom surfaces. However, the dominant part of the polarization should come from the topological contribution.

In the ideal situation where the axion action in Eq. (1) with \( \theta = \pi \) is realized over the whole sample, the total charge induced by \( B \) is proportional to the cross sectional area of the sample perpendicular to \( B \) as follows.
We suppose that $B$ is parallel to the $z$ direction, and the sample geometry is defined by height functions for upper and lower surfaces, $z = z_u(x,y)$ and $z = z_l(x,y)$, respectively, for $x, y$ within the projected region $S$ of the sample onto the $xy$-plane. The total charge on the upper surface is given by

$$Q = \int_S dx dy \frac{1}{z_u(x,y) - z_l(x,y)} dz \frac{e^2}{2\pi \hbar} B,$$

where $A_S = \int_S dx dy$ is the cross sectional area of the sample in the $xy$-plane. Conversely, the axion angle $\theta$ deviates from the quantized value in our setups for the TME effect, since there is non-topological contribution proportional to the area of the side surface as shown in Fig. 4(c) and Fig. 5(c).

*Note added:* After the completion of this work, a paper on a related topic by Wang et al. [24] has appeared.

**ACKNOWLEDGMENTS**

We thank Y. Tokura and M. Kawasaki for fruitful discussions. This work was supported by Grant-in-Aid for Scientific Research (No. 24224009, No. 26103006, No. 24540338, and No. 15K05141) from the Ministry of Education, Culture, Sports, Science and Technology (MEXT) of Japan and from Japan Society for the Promotion of Science.

**Appendix: Linear response theory for current density and polarization**

We give a brief derivation of Eqs. (13) and (19) from linear response theory. The Hamiltonian

$$H = H_0 + A(t),$$

(A.1)

consists of an unperturbed part $H_0$ and a perturbation $A(t) = A e^{-i\omega t}$. We are interested in the expectation value of an operator $B$ in the ground state of $H$,

$$\langle B \rangle = B_0 e^{-i\omega t},$$

(A.2)

which is expanded in powers of $A$. The Fourier component $B_\omega$ in linear order in $A$ is obtained from the linear response theory,

$$B_\omega = -\frac{i}{\hbar} \int_0^\infty d\tau e^{i\omega \tau} \text{tr} \left[ e^{-iH_0 \tau/\hbar} [A, \rho_0] e^{iH_0 \tau/\hbar} B \right],$$

(A.3)

where the density matrix $\rho_0 = |0\rangle \langle 0|$ is the projection to the many-body ground state $|0\rangle$ of $H_0$. For noninteracting fermions, $\rho_0$ can be written as $\rho_0 = \prod_{i=1}^N |\psi_i\rangle \langle \psi_i|$, in terms of the occupied single-particle states $\psi_i$. The Fourier component $B_\omega$ in Eq. (A.3) is then rewritten as

$$B_\omega = -\frac{i}{\hbar} \int_0^\infty d\tau \sum_{i \in O, j \in U} \left[ \frac{B_{ij} A_{ji} e^{i(E_i - E_j + \hbar \omega)\tau/\hbar}}{E_i - E_j + \hbar \omega} - \frac{B_{ji} A_{ij} e^{i(E_i - E_j + \hbar \omega)\tau/\hbar}}{E_i - E_j - \hbar \omega} \right],$$

(A.4)

where $O$ and $U$ stand for sets of occupied and unoccupied single-particle states in the ground state of $H_0$, and $A_{ij}$ and $B_{ij}$ are matrix elements of the operators $A$ and $B$.

We have used the hermiticity of the operators $A$ and $B$ when passing from the first line to the second line. In the dc limit Eq. (A.4) reduces to

$$B_{\omega \rightarrow 0} = \sum_{i \in O, j \in U} \frac{2 \text{Re}(B_{ij} A_{ji})}{(E_i - E_j)^2 - (\hbar \omega)^2}. $$

(A.5)

The axion angle $\theta$ is defined in Sec. IV as a response coefficient of polarization $P$ to magnetic field $B$. The linear response formula [Eq. (19)] is obtained by setting $B = P$ and $A = e\nu F \times \sigma B$ in Eq. (A.5).

In order to obtain the formula in Eq. (13) for the current density that is proportional to the time derivative of the magnetic field, we need to extract from $\langle B \rangle$ the component that is proportional to the time derivative of the perturbation, $d\langle A(t) \rangle/dt = -i\omega A e^{-i\omega t}$. That component is given by

$$\lim_{\omega \rightarrow 0} \text{Re} \left[ B_{ij}/(-i\omega) \right] = \frac{2h}{E_i - E_j} \text{Im}(B_{ij} A_{ji}).$$

(A.6)

When the magnetic field is varied as $B \rightarrow B + \delta B$, the 3D bulk Hamiltonian $H$ in Eq. (8) is perturbed by $\delta B O = \frac{1}{2} e\nu F (-y\sigma_x + x\sigma_y) \tau_3 \delta B$. Equation (13) is obtained by setting $B = f$ and $A = O$ in Eq. (A.6).
[1] R. E. Prange and S. M. Girvin, eds., The Quantum Hall Effect (Springer-Verlag, New York, 1987).
[2] S. Das Sarma and A. Pinczuk, eds., Perspectives in Quantum Hall Effects (Wiley, New York, 1997).
[3] M. Z. Hasan and C. L. Kane, “Colloquium: Topological insulators,” Rev. Mod. Phys. 82, 3045 (2010).
[4] X.-L. Qi and S.-C. Zhang, “Topological insulators and superconductors,” Rev. Mod. Phys. 83, 1057 (2011).
[5] M. König, S. Wiedmann, C. Brüne, A. Roth, H. Buhmann, L. W. Molenkamp, X.-L. Qi, and S.-C. Zhang, “Quantum Spin Hall Insulator State in HgTe Quantum Wells,” Science 318, 766 (2007).
[6] X.-L. Qi, T. L. Hughes, and S.-C. Zhang, “Topological field theory of time-reversal invariant insulators,” Phys. Rev. B 78, 195424 (2008).
[7] A. M. Essin, J. E. Moore, and D. Vanderbilt, “Magnetoelectric Polarizability and Axion Electrodynamics in Crystalline Insulators,” Phys. Rev. Lett. 102, 146805 (2009).
[8] A. M. Essin, A. M. Turner, J. E. Moore, and D. Vanderbilt, “Orbital magnetoelectric coupling in band insulators,” Phys. Rev. B 81, 205104 (2010).
[9] X.-L. Qi, R. Li, J. Zang, and S.-C. Zhang, “Inducing a Magnetic Monopole with Topological Surface States,” Science 323, 1184 (2009).
[10] W.-K. Tse and A. H. MacDonald, “Giant Magnetooptical Kerr Effect and Universal Faraday Effect in Thin-Film Topological Insulators,” Phys. Rev. Lett. 105, 057401 (2010).
[11] J. Maciejko, X.-L. Qi, H. D. Drew, and S.-C. Zhang, “Topological Quantization in Units of the Fine Structure Constant,” Phys. Rev. Lett. 105, 166803 (2010).
[12] A. Malashevich, I. Souza, S. Coh, and D. Vanderbilt, “Theory of orbital magnetoelectric response,” New J. Phys. 12, 053032 (2010).
[13] R. S. K. Mong, A. M. Essin, and J. E. Moore, “Anti-ferromagnetic topological insulators,” Phys. Rev. B 81, 245209 (2010).
[14] K. Nomura and N. Nagaosa, “Surface-Quantized Anomalous Hall Current and the Magnetoelectric Effect in Magnetically Disordered Topological Insulators,” Phys. Rev. Lett. 106, 166802 (2011).
[15] H. Y. Hwang, Y. Iwasa, M. Kawasaki, B. Keimer, N. Nagaosa, and Y. Tokura, “Emergent phenomena at oxide interfaces,” Nat. Mater. 11, 103 (2012).
[16] C.-Z. Chang, J. Zhang, X. Feng, J. Shen, Z. Zhang, M. Guo, K. Li, Y. Ou, P. Wei, L.-L. Wang, Z.-Q. Ji, Y. Feng, S. Ji, X. Chen, J. Jia, X. Dai, Z. Fang, S.-C. Zhang, K. He, Y. Wang, L. Lu, X.-C Ma, and Q.-K. Xue, “Experimental Observation of the Quantum Anomalous Hall Effect in a Magnetic Topological Insulator,” Science 340, 167 (2013).
[17] J. G. Checkelsky, R. Yoshimi, A. Tsukazaki, K. S. Takahashi, Y. Kozuka, J. Falson, M. Kawasaki, and Y. Tokura, “Trajectory of the anomalous Hall effect towards the quantized state in a ferromagnetic topological insulator,” Nat. Phys. 10, 731 (2014).
[18] C. Brüne, C. X. Liu, E. G. Novik, E. M. Hankiewicz, H. Buhmann, Y. L. Chen, X. L. Qi, Z. X. Shen, S. C. Zhang, and L. W. Molenkamp, “Quantum Hall Effect from the Topological Surface States of Strained Bulk HgTe,” Phys. Rev. Lett. 106, 126803 (2011).
[19] Y. Xu, I. Miotkowski, C. Liu, J. Tian, H. Nam, N. Alidoust, J. Hu, C.-K. Shih, M. Z. Hasan, and Y. P. Chen, “Observation of topological surface state quantum Hall effect in an intrinsic three-dimensional topological insulator,” Nat. Phys. 10, 956 (2014).
[20] R. Yoshimi, A. Tsukazaki, Y. Kozuka, J. Falson, K. S. Takahashi, J. G. Checkelsky, N. Nagaosa, M. Kawasaki, and Y. Tokura, “Quantum Hall effect on top and bottom surface states of topological insulator (Bi_{1-x}Sb_x)2Te3 films,” Nat. Commun. 6, 6627 (2015).
[21] T. Morimoto, A. Furusaki, and N. Nagaosa, “Charge and Spin Transport in Edge Channels of a ν = 0 Quantum Hall System on the Surface of Topological Insulators,” Phys. Rev. Lett. 114, 146803 (2015).
[22] J. G. Checkelsky, J. Ye, Y. Onose, Y. Iwasa, and Y. Tokura, “Dirac-fermion-mediated ferromagnetism in a topological insulator,” Nat. Phys. 8, 729 (2012).
[23] Y. Omori, F. Auvray, T. Wakamura, Y. Niimi, A. Fert, and Y. Otani, “Inverse spin hall effect in a closed loop circuit,” App. Phys. Lett. 104, 242415 (2014).
[24] J. Wang, B. Lian, X.-L. Qi, and S.-C. Zhang, “Quantized topological magnetoelectric effect of the zero-plateau quantum anomalous hall state,” arXiv:1506.03141 (2015).