Non-local sidewall response and deviation from exact quantization of the topological magnetoelectric effect in axion-insulator thin film

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Topological insulator (TI) thin films with surface magnetism are expected to exhibit a quantized anomalous Hall effect (QAHE) when the magnetizations on the top and bottom surfaces are parallel, and a quantized topological magnetoelectric (QTME) response when the magnetizations have opposing orientations (axion insulator phase) and the films are sufficiently thick. We present a unified picture of both effects that associates deviations from exact quantization of the QTME caused by finite thickness with non-locality in the side-wall current response function. Using realistic tight-binding model calculations, we show that in Bi2Se3 thin films deviations from quantization in the axion insulator-phase are reduced in size when the exchange coupling of tight-binding model basis states to the local magnetization near the surface is strengthened. Stronger exchange coupling also reduces the effect of potential disorder, which is unimportant for the QAHE but detrimental for the QTME, which requires that the Fermi energy lie inside the gap at all positions.

Introduction—In magnetoelectric materials, an applied electric field $\mathbf{E}$ induces magnetization $\mathbf{M}$ and an applied magnetic field $\mathbf{B}$ induces electrical polarization $\mathbf{P}$. Magnetoelectric response is described by the linear magnetoelectric polarizability tensor $\alpha$, whose diagonal components,

$$\alpha_{ii} = \frac{\partial M_i}{\partial E_i} \bigg|_{\mathbf{B}=0} = \frac{\partial P_i}{\partial B_i} \bigg|_{\mathbf{E}=0} = \frac{\theta}{2\pi} \frac{e^2}{\hbar},$$

are pseudoscalars. ($\theta$ in Eq. 1 is dimensionless.) Since $\mathbf{B}$ and $\mathbf{M}$ are odd under time-reversal and $\mathbf{E}$ and $\mathbf{P}$ are odd under space-inversion, magnetoelectric response normally occurs in insulators that break both time-reversal and inversion symmetry (TRS and IS), and is typically characterized by a small value of $\theta$. We are interested here in the orbital magnetoelectric response of three-dimensional (3D) topological insulators (TIs), which is special in the sense that it is non-zero even when TRS is not broken in the sample bulk. The magnetoelectric response is instead related to a non-trivial topological invariant of the bulk bands and is quantized at $\theta = \pi$ ($\alpha_{ii} = e^2/2\hbar$).

The Quantized Topological Magnetoelectric Effect (QTME) is realized only when the TI surface magnetization adopts an axion insulator configuration, one in which all facets and hinges (where facets meet) of the bulk TI crystal surface are insulating. As shown in Fig. 1, when the magnetizations have the same orientation, the film displays the Quantum Anomalous Hall Effect (QAHE) and the side walls (hinges) are not gapped. Both the QAHE and the QTME can be understood qualitatively by considering the limit of weakly-gapped Dirac cone surface states, since these give rise to half-quantized intrinsic anomalous Hall conductances $\sigma_H = \pm e^2/2\hbar$ with a sign determined by the magnetization orientation. The QAHE has been observed in uniformly-doped magnetic TI thin films, in modulation-doped TI films, and have surface magnetism only, and recently also in TI films with proximity-induced 2D magnetism. On the other hand, the QTME not yet been directly confirmed experimentally, even though successful realization of the axion insulator configuration is strongly suggested in some experiments and the absence of a Hall effect in states with oppositely oriented top and bottom surface magnetizations. Novel intrinsic antiferromagnetic TIs, such as the van der Waals layered MnBi2Te4 and Mn4Bi2Te7 families and their heterostructures with nonmagnetic TIs have also been found to display the QAHE. These systems do not suffer from the intrinsic disorder of doped TIs and...

FIG. 1. (a) Schematic of a TI thin film with surface magnetizations at top and bottom in either parallel (P: Chern-insulator phase) or antiparallel (AP: axion-insulator phase) configurations. (b)-(c) Bandstructure of a 15QL Bi2Se3 TI thin film in the P configuration for two choices of the top and bottom surface exchange fields $J_t$ and $J_b$. The gapped Dirac surface states are indicated by red and blue lines. In the symmetric case (b) these states are nearly doubly degenerate, and approach exact degeneracy in the thick film limit.
typically possess relatively larger magnetic gaps at their Dirac points.

In this Letter we employ a unified description of the QAHE and the QTME in TI thin films, by expressing the magnetization in terms of sidewall currents that respond non-locally to electric potentials that vary slowly across the film. In this picture, perfect quantization of the QTME requires sidewall response that is localized near the top and bottom surfaces, whereas the QAHE requires only bulk state localization. We characterize the non-locality of the sidewall response by calculating finite-size corrections to the QTME theoretically using a realistic tight-binding model, demonstrating that they are smaller for stronger exchange coupling \( J(\mathbf{r}) \) between the surface magnetization and tight-binding model basis states localized near the surface. By increasing surface state gaps, stronger exchange coupling not only reduces finite-size corrections but also reduces the effect of disorder, which can be tolerated in QAHE measurements but is deteriorative for the QTME.

**QAHE, QTME, and sidewall response**— We consider the linear response of the \( \hat{z} \)-direction orbital magnetization of a thin film with a quasi-2D bulk gap to an electric potential that varies slowly across the film. Since the bulk is time-reversal invariant and insulating, the magnetization response must originate from changes in currents that circulate around the film side walls: \( M_z = (1/d) \sum_l I_{\text{sw}}(l) \). Here \( d \) is the thickness of the film, and we have anticipated our use of a tight-binding model by specifying the vertical position along the side wall using a discrete layer index \( l \). By allowing the \( l \)-dependent bulk electric potentials to turn on slowly upon entering the sample bulk \( [27] \) so that lateral electric fields are present only near the sidewall, we can relate the current response to bulk Hall conductivity:

\[
I_{\text{sw}}(l) = \frac{1}{e} \sum_{l'} \sigma_H(l, l')(V(l') - \mu)
\]  

where \( V(l) \) is a layer-dependent electric potential, and \( \sigma_H(l, l') \) is the thin-film Hall conductivity generalized \( [28] \) to allow for non-locality in the \( \hat{z} \)-direction \( [29] \):

\[
\sigma_H(l, l') = \frac{2e^2}{h} \sum_{n, n' \neq n} f_n \int \frac{d^2 k}{(2\pi)^2} \frac{\text{Im}[\langle u_{n,k}|P_l v_{n,k}(\mathbf{k})|u_{n',k}\rangle \langle u_{n',k}|P_l' v_{n,k}(\mathbf{k})|u_{n,k}\rangle]}{(E_{n,k} - E_{n',k})^2}.
\]  

In Eq. 2 we have introduced a chemical potential to allow for a unified discussion of the QAHE and QTME. In Eq. 3 \( f_n \) is a band occupation number, \( P_l \) is a layer projection operator, \( |u_{n,k}\rangle \) is a band state of the 2D Bloch Hamiltonian \( H(\mathbf{k}) \), \( E_{n,k} \) is the corresponding band energy \( \partial H(\mathbf{k})/\partial k_i \), \( i = x, y \) is the velocity operator. When summed over \( l \) and \( l' \), \( \sigma_H(l, l') \) yields \( e^2/h \) times the total Chern number \( C \) of all occupied 2D-bands, and is quantized.

The QAHE measures \( [30] \) the response of the total sidewall current to a uniform chemical potential shift, and its quantization is therefore explained simply by the quasi-2D band Chern numbers. The QTME is a zero-temperature property of a state that is fully insulating, and is a response not to chemical potential but to electric potential. Its quantization can nevertheless be understood in terms of Chern quantization by the following argument. Define \( \sigma(l) = \sum_{l', l} \sigma_H(l, l') \), where each layer is classified by proximity as belonging to the top or bottom layer subset. For thick films the side-wall response must be localized where time-reversal symmetry is broken, i.e., near the top or bottom surface. It follows that for any configuration of the surface magnetism the QTME requires sidewall response that is localized near the top or bottom surface. It follows that \( \sigma(l) \) and \( \sigma(l') \) must be separately universal. Since both must change sign when the magnetization is reversed at their surface, \( \pm \sigma_t \pm \sigma_b \) must be quantized. In the special case of an axion insulator (\( \sigma_H = 0 \)) it follows that \( \sigma_b = -\sigma_t = 2\sigma_t = ne^2/h \), and that for \( n = 1 \), \( \delta M = M(l_2) - M(0) \approx (1/ed)\sigma_t (V(l_2) - V(l_1)) = (e^2/2h)E_z \) when an electric field \( E_z \) is applied across the sample in the insulating state. \( V(l_{t(b)}) \) is the electric potential at the top (bottom) layer with layer index \( l_{t(b)} \). (Note that for asymmetric exchange fields at the two surfaces, the magnetization is non-zero even when \( E_z = 0 \).) Because the side-wall response has a finite localization length, the magnetization response has a finite size correction that varies inversely with the number of layers \( N \) in the film and is characterized by the dimensionless number \( m_{\text{corr}}(d) = \langle |\delta M(d = \infty) - \delta M(d)\rangle/\delta M(d = \infty) \rangle = (h/4e^2) \sum_{l(t)} \sigma_H(l(t) - l')/N \).

**Finite Thickness Corrections in Bi2Se3**— Finite-size corrections in thin films distinguish the QTME from the QAHE. To estimate their size in realistic systems we have added a uniform electric field \( E_z \) applied across finite thickness quasi-2D TI films to a realistic tight-binding (TB) model with surface magnetism, and explicitly evaluated the magnetization carried by the distorted bands using \( [31] \)
Note that Eq. 1 and Eq. 2 agree in the case of constant \( V(l) \) since a constant electric potential shifts band energies without changing wavefunctions. Eq. 3 gives the 2D bulk magnetization for an infinite cross-sectional area TI slab with broken TRS at the surfaces. The physical origin of the response of this magnetization to an electric field is the changes in the side-wall currents discussed above. This example of bulk-edge correspondence is closely analogous to that of the QAHE. \(^{30, 33}\)

We focus on Bi\(_2\)Se\(_3\) thin films, whose electronic structure can be described by a sp\(^3\) TB model with parameters obtained by fitting to ab initio electronic structure calculations. \(^{35, 36}\) We apply this TB model to a thin-films with finite numbers of van-der-Waals-coupled quintuple layers (QLs), and model broken time-reversal at the top and bottom surfaces by adding exchange fields of strength \( J_l \) and \( J_b \), oriented orthogonal to the (111) surface that couple to electron spin. We will consider two types of exchange-fields: (i) a homogeneous field applied to the entire first surface QL, modelling magnetic modulation doping; \(^{14, 18}\) and (ii) a homogeneous field applied only to the very top and bottom atomic monolayers (MLs), modelling the exponentially evanescent proximity effect of an adjacent magnetic layer. \(^{10, 21, 32}\)

In Figs. 1(b), (c) we plot the bandstructures of 15QL TI films for two strengths of exchange fields of type (i), both in the parallel (P) QAHE configuration, corresponding to the Chern insulator phase. For symmetric exchange fields \( J_{t(b)} = 0.1 \) eV (smaller than the bulk gap \( \approx 0.3 \) eV), the in-gap states on the two surfaces are essentially degenerate Dirac cones with exchange gaps \( \Delta \approx J \) at the DP. In the following, we will refer to the surface states below the exchange gap as valence-band states and to those above as conduction-band states. An analysis of the wavefunctions \(^{33}\) shows that, around the \( \Gamma \) point, these states are localized either at the top or bottom surface, decaying exponentially within the first two QLs, just like the Dirac surface states of a non-magnetic TI film \(^{35}\). For larger \( k \), the surface state bands merge with bulk bands and the corresponding wavefunctions are delocalized across the film \(^{33}\). On the other hand, when the exchange field at one surface is of the order of the bulk gap, as in Fig. 1(c), the Dirac-cone bandstructure at that surface is strongly modified; in particular the valence band no longer resembles a gapped Dirac cone even near the \( \Gamma \) point. In fact, the band flattens, but remains separated from the bulk bands for most \( k \) values. The corresponding wavefunctions are now localized at the strongly magnetized surface for a larger region of the BZ \(^{33}\). For exchange fields of type (ii) (not shown in the figure), the structure of the gapped Dirac cones is robust and, apart from the increase of the exchange gap with \( J_{t(b)} \), remains unmodified even for \( J_{t(b)} > 0.3 \) eV.

These results demonstrate that the spatial distribution of \( J(\mathbf{r}) \) plays a separate role from its strength in influencing how the Dirac surface state electronic structure is modified by surface magnetism.

We now consider the implications of these electronic structure properties for the magneto-electric response. We compute \( M_z \) numerically as a function of \( E_z \), represented in the TB Hamiltonian as an on-site energy \( \theta \) with varying strength \( \theta \) and position dependence, \( \theta \). That is, the magneto-electric coefficient \( \theta \) extracted from the calculation of the magnetization in Bi\(_2\)Se\(_3\) TI thin films of different thicknesses \( d \) and different values of the top and bottom surface exchange fields. \(^{10, 21, 37}\) By keeping \( eE_zd \) smaller than the surface-state gap \( \Delta \) in such a way that the \( M_z \) depends linearly on \( E_z \), we extract the \( \theta \) parameter defined in Eq. 1. The results are shown in Fig. 2. From Fig. 2(a) we can see that, starting from 5QLs, \( \theta \) versus \( d \) is well described by the relation \( \theta = \theta_{d \to \infty}(1 - w/d) \) \(^{34}\), where \( w \) is a non-universal length scale that can be extracted from this figure, and measures the localization of the side-wall current response. Finite-size corrections are larger than 10% for film thicknesses below \( \sim 20 \) nm. Fig. 2(b) demonstrates that for all choices of the exchange strengths \( J_{t/b} \) and position dependence, \( \theta_{d \to \infty} = \pi \) to within numerical accuracy \( \sim 1\% \). That is, the magneto-electric coefficient is quantized. Importantly, as shown in Fig 2(a), the length scale \( w \) decreases with increasing \( J_{t(b)} \). Finite-size corrections are reduced when the surface magnetization is strengthened.

The quantization of the magneto-electric response is consistent with the properties of the non-local Hall conductivity shown in Fig. 3, where \( \sigma_{H}(l, l') \) is plotted as a function of the QL indices \( l \) and \( l' \) for two values of the exchange constants. As anticipated, \( \sigma_{H}(l, l') \) is localized near the top and bottom surfaces, where TRS is broken. Furthermore, a careful numerical evaluation of \( \sigma(\mathbf{b}) = \sum_{l,l' \in \mathbf{b}} \sigma_{H}(l, l') \) shows that the larger \( J_{t(b)} \) is
the more localized $\sigma_{l(b)}$ is at the surfaces.

\[ C(l) = \frac{1}{2\pi} \int_{BZ} d^2k \Omega_{xy}^{(l)}(k), \]  

where

\[ \Omega_{xy}^{(l)}(k) = -2i \sum_{p, q, m, n} \frac{\langle u_{nk}|v_p(k)|u_{mq}(k)\rangle|\langle u_{np}(k)|v_m(k)\rangle|}{(E_{nk} - E_{pq}(k))^2} W_{nk}^{(l)}. \]  

In Eq. 6 $W_{nk}^{(l)} \equiv \sum_{s \epsilon \{t, b\}} |\langle s|u_{nk}\rangle|^2$ is the weight of $|u_{nk}\rangle$ when projected on orbitals $|s\rangle$ centered at the sites $s$ of the $l^{th}$ QL. The total Berry curvature does not single out the contribution of a particular quasi-2D band. However, when projected on a given QL, $\Omega_{xy}^{(l)}(k)$ and $C(l)$ are a good measure of the contribution of the surface states relative to the contribution of the bulk states. The total $\Omega$ and $C$ obtained by summing Eqs. 4, 5 over $l$, yield $C = 1$ in the Chern insulator state and $C = 0$ in the axion insulator state, regardless of the number of QLS and the value of $J$.

In Fig. 4 we plot $C(l)$ versus the QL index $l$ for a 15QL film for different values of $J_b/J_t$. Consistent with the band-structures in Fig. 3 these results show that for $J_b/J_t$ substantially smaller than the bulk gap, the only states with substantial Berry curvature are those that derive from the non-magnetic state Dirac cones, which are strongly localized in the first three QLS. Note that the first three QLS represent the typical localization region of the evanescent surface states in non-magnetic TI films. In this case, we can operationally define a surface QAHE conductance by $\tilde{\sigma}_{H, l(b)} = e^2/h \sum_{l=t(b)} C(l)$, and find that it is closer to $\pm e^2/2h$: in other words the explanation for the TME in terms of surface localized half-quantized Hall conductivities applies literally. Only under these conditions do the definitions of a top and bottom surface Hall conductivities apply literally. Only under these conditions do the definitions of a top and bottom surface Hall conductivities apply literally.
depends only on $J_t$ and $\sigma_b$ depends only on $J_b$. This locality condition is certainly satisfied in the limit of weak time-reversal symmetry breaking where the exchange gap is considerably smaller than the bulk gap and the Hall conductivity is contributed by weakly gapped Dirac-cone surface states. It is less obvious, perhaps, that the locality condition is satisfied in the strong $J_t, J_b$ limit where we have shown that states with large Berry curvature are extended across the sample - although even here non-local response would be very surprising. The assumption of locality is supported by the fact that, when combined with quantization of the total Hall conductivity and time-reversal symmetry properties, it naturally accounts for the expected quantization value of the QTME $\sigma_t = \sigma_b = e^2/2h$, which we confirm numerically, and for the $(1 - w/d)$ form of finite-size corrections, with $w$ being a non-locality length of the side-wall response and $d$ the film thickness.

We have established the $(1 - w/d)$ finite-size law using explicit calculations of magnetization in the presence of an electric field applied across a Bi$_2$Se$_3$ topological insulator which yield $w \sim 2nm$. If this is correct, the part in $10^6$ quantization accuracy routinely achieved for quantum Hall systems would require films of $\sim \mu m$ thickness. Stronger surface magnetism generates larger quasi-2D gaps, which in turn imply greater robustness of the quantized response against potential disorder that is inevitable and can invalidate quantization by inducing surface electron or hole puddles. It is therefore encouraging for QTME measurement efforts that finite-size corrections to the QTME are smaller for surface magnetism that is stronger. This can be achieved either in the sense of coupling more strongly to electron spins near the Fermi level or in the sense of being present over more near-surface layers of the film.

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