Effects of surface-bulk hybridization in 3D topological ‘metals’

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Identifying the effects of surface-bulk coupling is a key challenge towards exploiting the topological nature of the surface states in many available three-dimensional topological ‘metals’. Here we combine an effective-model calculation and an ab-initio slab calculation to study the effects of the lowest order surface-bulk interaction: the hybridization. In the effective-model study, we discretize an established low-energy effective four-band model and introduce the hybridization between the surface bands and bulk bands in the spirit of the Fano model. We find that the hybridization enhances the energy gap between bulk and Dirac surface states and preserves the latter’s spin texture qualitatively albeit with a reduced spin-polarization magnitude. Our ab-initio study finds the energy gap between the bulk and surface states to grow upon an increase in the slab thickness, very much in qualitative agreement with the effective model study. Comparing the results of our two approaches, we deduce that the experimentally observed low magnitude of spin polarization can be attributed to a hybridization-type surface-bulk interaction. We discuss evidences for such hybridization in existing ARPES data.

I. INTRODUCTION

Many discrepancies between experimental measurements and theoretical predictions of ideal topological insulators (TI’s) are attributed to the fact that the chemical potential lies in the conduction band and the bulk band interferes with measurements. That is, many available TI materials are actually metallic. Recent developments in thin-film experiments further call for studies of surface-bulk electron interaction in films. However, explicit first-principles calculations on TI films are limited to very thin slabs with thickness less than 10 nm due to the computational cost. Therefore, there is a need for a simple microscopic model which incorporates surface-bulk interactions that can be used to study how physical properties depend on the film thickness.

One important question such a model should address is the effect of surface-bulk interaction on the spin-texture. The surface spin-texture is a key physical characteristic of topological surface states in ideal TIs. While low-energy effective theories guided by symmetries predict perfect spin-momentum locking for topological surface states within the bulk gap, spin- and angle-resolved photoemission spectroscopy (SARPES) data show lower in-plane spin polarization and total spin magnitude. On the other hand, an ab-initio calculation on a few-quintuple-layer (QL) slab found both the spin polarization and the total spin magnitude to be much smaller. Although reduction in spin polarization and total spin magnitude are to be anticipated at large surface Fermi-momenta where hexagonal warping manifests, little is understood about the reduction observed at small Fermi-momenta and how the surface-bulk interaction affects the spin texture. However, such understanding is crucial for pursuing technical applications of spin-momentum locking in thin films of topological insulators in the metallic regime.

Our starting point is the observation made by Bergman and Refael that the lowest order electron-electron interaction term between the surface state and the bulk states can be viewed as a hybridization term in the Fano model. The key effect of hybridization in this low-energy effective theory is to spectroscopically separate surface states localized on the surface from the extended metallic bands. Building on the principles underlying this low-energy effective theory, we study the hybridization effects with a microscopic model of 3D time-reversal invariant strong topological insulators to address the thickness dependence of physical quantities and connect the results to ab-initio slab calculations. Specifically, we study TI slabs with finite thickness in the presence of surface-bulk interaction from two complementary perspectives: a simple microscopic model including the lowest order surface-bulk interaction and an ab-initio calculation of a few-QL Bi$_2$Se$_3$. We focus on how the spectroscopic properties and the spin texture evolve as a function of film thickness.

The paper is structured as follows. In section II we construct a lattice model for a slab with surface-bulk hybridization (S-B hybridization) and study the spectroscopic properties of the model as well as the effects of the S-B hybridization on the spin texture. In section III we present an ab-initio study on 4,5,6-QL Bi$_2$Se$_3$ using density functional theory (DFT) and discuss the insight the simple hybridization model offers in understanding the ab-initio results. We then conclude in section IV with discussions of implications of our results and open questions.
II. LATTICE MODEL FOR A SLAB WITH S-B HYBRIDIZATION

A. The Model

In order to introduce surface-bulk hybridization as a perturbation in the spirit of the Fano mode and study its effects on a slab with finite thickness, we first need a lattice model for a slab. For this, we discretize the effective continuum model by Zhang et al., which is a four band \( k \cdot p \) Hamiltonian guided by symmetries and first-principle-calculation results. We then make the system size finite along the vertical axis, i.e., the film growth direction.

The low-energy effective four-band model in Ref. describes a strong 3D TI with rhombohedral crystal structure such as \( \text{Bi}_2\text{Se}_3 \). In this effective model each QL is treated as a layer since the inter-QL coupling is weak, and the four lowest-lying spin-orbital bands come from the mixing of the two \( P_z \) atomic orbitals from Bi and Se referred to as \( P_1 \) and \( P_2 \) and the two spins \( \uparrow, \downarrow \). We take this effective model written in terms of \( 4 \times 4 \) \( \Gamma \)-matrices and discretize it following the discretization scheme used for 2D TIs in Ref. such that the lattice Hamiltonian reduces to the low-energy effective \( k \cdot p \) Hamiltonian in Ref. in the limit \( |k| \to 0 \). The resulting lattice model in the rotated spin-orbital basis \( \{ |P_1, \uparrow \rangle, -i|P_2, \uparrow \rangle, |P_1, \downarrow \rangle, i|P_2, \downarrow \rangle \} \) is

\[
H_0 = \sum_{k_\parallel, k_z} h_0(k_\parallel, k_z) c_{k_\parallel, k_z}^\dagger c_{k_\parallel, k_z},
\]

where \( c_{k_\parallel, k_z}^\dagger \) and \( c_{k_\parallel, k_z} \) is an operator creating a four spinor in the rotated spin-orbital basis with an in-plane momentum \( k_\parallel = (k_x, k_y) \) and perpendicular momentum \( k_z \), and the lattice Hamiltonian is

\[
h_0(k_\parallel, k_z) = \epsilon_1(k_\parallel, k_z) \Psi_{\downarrow \times \downarrow} + \frac{A_1}{a_z} \sin(k_z a_z) \Gamma^1 \]

\[
+ \frac{A_2}{a_z^2} \sin(k_z a_z) \Gamma^3 + \frac{A_2}{a_y} \sin(k_y a_y) \Gamma^4 + M_1(k_\parallel, k_z) \Gamma^5,
\]

with

\[
\epsilon_1(k_\parallel, k_z) \equiv C + \frac{2D_1}{a_z^2} [1 - \cos(k_z a_z)] + \frac{2D_2}{a_z^2} [1 - \cos(k_z a_z)]
\]

\[
+ \frac{2D_2}{a_y^2} [1 - \cos(k_y a_y)],
\]

\[
M_1(k_\parallel, k_z) \equiv m - \frac{2B_1}{a_z^2} [1 - \cos(k_z a_z)] - \frac{2B_2}{a_z^2} [1 - \cos(k_z a_z)]
\]

\[
- \frac{2B_2}{a_y^2} [1 - \cos(k_y a_y)].
\]

The Gamma matrices are defined as \( \Gamma^1 = \sigma^z \otimes \tau^x \), \( \Gamma^2 = -\sigma^z \otimes \tau^y \), \( \Gamma^3 = \sigma^x \otimes \tau^x \), \( \Gamma^4 = \sigma^y \otimes \tau^x \), and \( \Gamma^5 = \sigma^y \otimes \tau^y \), where \( \sigma^x \) and \( \sigma^y \) are Pauli matrices acting on \( (\uparrow, \downarrow) \) and \( (P_1, P_2) \) spaces, respectively. \( a_x, a_y \) and \( a_z \) are the lattice constants in \( x, y, z \) directions respectively.

We use the parameters for \( \text{Bi}_2\text{Se}_3 \) obtained by fitting the continuum model to ab-initio calculations, \( m = 0.28 \text{ eV}, A_1 = 2.2 \text{ eV \AA}, A_2 = 4.1 \text{ eV \AA}, B_1 = 10 \text{ eV \AA}^2, B_2 = 56.6 \text{ eV \AA}^2, C = -0.0068 \text{ eV}, D_1 = 1.3 \text{ eV \AA}^2 \) and \( D_2 = 19.6 \text{ eV \AA}^2 \).

We model a slab of \( \text{Bi}_2\text{Se}_3 \) by imposing open boundary conditions at the top and the bottom surfaces which break the translational symmetry along the \( z \)-axis. Since \( k_z \) is no longer a good quantum number, while \( k_\parallel \) still is, we substitute \( c_{k_\parallel, j} \equiv \frac{1}{\sqrt{N}} \sum_{j} e^{ik_{\parallel} j a_z} c_{k_\parallel, j} \) in Eq. and label the four-spinor operators by the in-plane momentum \( k_\parallel = (k_x, k_y) \) and the index of layers \( j \) stacking in the \( z \) direction. Now the Hamiltonian for a slab with \( N \) layers is

\[
H_0(N) = \sum_{k_\parallel} H_0(k_\parallel, N),
\]

where

\[
H_0(k_\parallel, N) = \sum_{j=1}^{N} M \epsilon(k_{\parallel, j}) c_{k_\parallel, j} c_{k_{\parallel, j+1}} + \mathbb{T} c_{k_\parallel, j} c_{k_\parallel, j+1} + \mathbb{D} c_{k_\parallel, j} c_{k_\parallel, j+1}
\]

and the \( 4 \times 4 \) matrices \( \mathbb{T} \) and \( \mathbb{M} \) are defined as

\[
\mathbb{T} \equiv -\frac{D_1}{a_z^2} \mathbb{I}_{4 \times 4} + \frac{B_1}{a_z^2} \Gamma^5 - \frac{iA_1}{2a_z} \Gamma^1,
\]

\[
\mathbb{M} \equiv \epsilon(k_{\parallel}) \mathbb{I}_{4 \times 4} + \frac{A_2}{a_z} \sin(k_z a_z) \Gamma^1 + \frac{A_2}{a_y} \sin(k_y a_y) \Gamma^4 + M_2(k_{\parallel}) \Gamma^5
\]

where

\[
\epsilon(k_{\parallel}) \equiv C + \frac{2D_1}{a_z^2} [1 - \cos(k_z a_z)] + \frac{2D_2}{a_z^2} [1 - \cos(k_z a_z)]
\]

\[
+ \frac{2D_2}{a_y^2} [1 - \cos(k_y a_y)],
\]

\[
M_2(k_{\parallel}) \equiv m - \frac{2B_1}{a_z^2} [1 - \cos(k_z a_z)] - \frac{2B_2}{a_z^2} [1 - \cos(k_z a_z)]
\]

For the sake of simplicity, the results presented in the remainder of this section are calculated with \( a_x = a_y = a_z = 1 \text{ \AA} \).

We can diagonalize \( H_0(k_{\parallel}, N) \) as

\[
H_0(k_{\parallel}, N) = \sum_{\alpha=1}^{4N-4} E_{\alpha, k_{\parallel}}^0 (k_{\parallel}) b_{\alpha, k_{\parallel}}^0 d_{\alpha, k_{\parallel}}^0
\]

\[
+ \sum_{\beta=1}^{4} E_{\beta, k_{\parallel}}^0 (k_{\parallel}) d_{\beta, k_{\parallel}}^0 b_{\beta, k_{\parallel}}^0,
\]

where \( b_{\alpha, k_{\parallel}}^0 \) and \( d_{\beta, k_{\parallel}}^0 \) are four-spinor annihilation operators for bulk and surface states (henceforth referred
to as the “Dirac” states) respectively in the absence of hybridization, \( E^0_{B,\alpha} \) and \( E^0_{D,\beta} \) are their corresponding eigenenergies. Here, \( \alpha \) and \( \beta \) label the unhybridized bulk and Dirac states respectively. All energy eigenstates are two-fold degenerate as required by inversion (P) and time-reversal (T) symmetries. For each in-plane momentum \( k_\parallel \), four energy eigenstates with their energies closest to the Dirac point are labeled to be valence (\( \beta = 1, 2 \)) and conduction (\( \beta = 3, 4 \)) Dirac states. \( \alpha \) label the remaining \( 4N - 4 \) bulk states. A natural choice for the labeling is to let \( \alpha = 1, \ldots, 2N - 2 \) denote valence bulk states and let \( \alpha = 2N - 1, \ldots, 4N - 4 \) denote conduction bulk states with the eigenenergies increasing monotonically with \( \alpha \). As the model is derived from a low-energy effective model near the Dirac point, it will break down at large energies. However, we expect qualitatively correct results when it comes to trends of physical properties over the hybridization strength and film thickness which only require knowledge of the low-energy physics near the insulating gap.

Now we introduce the S-B hybridization term that is allowed by symmetries in the spirit of Fano mode\(^\text{[20]}\). The Fano model is a generic model describing the mixing between extended states \( c_k \) with energy \( \epsilon_k \) and a localized state \( b \) with energy \( \epsilon \) through Hamiltonian
\[
H_F = \epsilon b^\dagger b + \sum_k [\epsilon_k c_k^\dagger c_k + A_k (c_k^\dagger b + b^\dagger c_k)],
\]
where \( A_k \) represents scattering strength. Bergman and Refael\(^\text{[20]}\) pointed out that \( H_F \) can be used to describe the lowest order interaction between a helical surface state and a metallic bulk band, i.e. hybridization. They studied effects of hybridization in a field theoretic approach. Here we use a symmetry-preserving form of the surface-bulk hybridization term in the spirit of \( H_F \) for the microscopic model of Bi\(_2\)Se\(_3\) shown in Eq. (11) to study the effects of mixing between Dirac states and bulk states.

For simplicity we consider the case where the hybridization strength preserves in-plane momenta \( k_\parallel \) and is independent of energy and \( k_\parallel \), i.e.
\[
h'(k_\parallel) = \sum_{\alpha,\beta} b_{\alpha,k_\parallel}^0 d_{\beta,k_\parallel} + H.c.
\]
We then impose \( T \) and \( P \) symmetries\(^\text{[23]}\) in the full hybridization perturbation \( H'(k_\parallel) \) by constructing \( H'(k_\parallel) \) through
\[
H'(k_\parallel) = h'(k_\parallel) + P h'(k_\parallel) P^\dagger + T h'(k_\parallel) T^\dagger + P \Theta h'(k_\parallel) (PT)^\dagger,
\]
where the representations for \( T \) and \( P \) symmetry operators with the spatial inversion center at the middle point of the slab in the current \( 4N \) tight-binding spin-orbital basis are \( T = I \sigma_y \otimes I_{2x2} \otimes I_{2N} \) with \( k \leftrightarrow -k \), and \( P = I_{2x2} \otimes \tau^z \) with \( z : [0, N/2] \leftrightarrow [N/2, N] \) and \( k \leftrightarrow -k \), respectively. Here, \( K \) is the usual complex conjugation operator. Finally, the full Hamiltonian including hybridization at a given in-plane momentum \( k_\parallel \) reads
\[
H(k_\parallel) = H_0(k_\parallel, N) + H'(k_\parallel).
\]
After diagonalizing the full Hamiltonian in the tight-binding spin-orbital bases, we can write
\[
H(k_\parallel) = \sum_{B,\alpha} E_{B,\alpha}(k_\parallel) b_{\alpha,k_\parallel}^\dagger b_{\alpha,k_\parallel} + \sum_{D,\beta} E_{D,\beta}(k_\parallel) d_{\beta,k_\parallel}^\dagger d_{\beta,k_\parallel},
\]
where \( b_{\alpha,k_\parallel} \) and \( d_{\beta,k_\parallel} \) are the bulk and Dirac states for \( H(k_\parallel) \), where the terms bulk and Dirac states are defined in the same fashion as for \( H_0(k_\parallel, N) \) in the absence of hybridization.

### B. Topological Metal Regime

We begin our numerical study with no hybridization. In the absence of hybridization, depending on the chemical potential \( \mu \), we now define three regimes: topological insulator (TI), metal (M), and topological metal (TM)(see Fig. (1a)). The familiar topological insulator (TI) regime is where the chemical potential lies within the bulk gap and the system is actually a bulk band insulator, i.e., \( E_c \leq \mu \leq E_v \) with \( E_v \) being the bottom of the conduction band and \( E_c \) the top of the valence band. Within the TI regime, the Dirac states feature Rashba-type spin-momentum locking and a spatial profile localized on the surfaces. Of our particular interest is the distinction we will draw between M and TM regimes based on whether the Dirac states retain the spin-momentum locking and the surface localization when being away from the TI regime.

In order to examine the above two properties of Dirac states, we define \( |\Psi_{D,k_\parallel}(z)|^2 \equiv |\tilde{\psi}_{D,k_\parallel}(z)|^2 + |\tilde{\psi}_{D,k_\parallel}(z)|^2 \) which is a function of \( z \) measured from the bottom of the slab along the finite dimension of the slab. This quantity will show whether the Dirac states are localized on the surfaces or not. Let us identify a particular \( k_\parallel \) of interest for a given value of chemical potential as the in-plane momentum at which the chemical potential \( \mu \) intersects the Dirac branch; we denote such in-plane momentum by \( k_{\parallel,\mu} \). To illustrate the features defining the three regimes, we will now show the spatial profiles and the spin polarizations of the Dirac states in the three regimes in the absence of hybridization. In the next section we will add the hybridization and examine its effects.
within the regime $\mu \gtrsim E_M$ (regime M), wavefunctions for all states at in-plane momentum $k_{\parallel,\mu}$ delocalize. We define the system to behave as a topological metal(TM) when the chemical potential lies within this window, i.e. $E_c < \mu < E_M$, represented by $\mu_{TM}$.

A detectable characteristic of TI and TM regimes is the spin-momentum locking. One measure to quantify spin-momentum locking at the surface is through the so-called “spin polarization” which is the expectation value of the spin component perpendicular to the in-plane momentum of a Dirac state, i.e.,

$$\langle S^z \rangle(k_{\parallel}) \equiv \langle \psi_{D,k_{\parallel}} | S^z | \psi_{D,k_{\parallel}} \rangle$$

with $\mathbf{n} \cdot k_{\parallel} = 0$. Here the $\mathbf{n}$ component of the quantum spin operator is defined as $S^z = \frac{1}{2} \sigma^z \cdot \mathbf{n} \otimes I_{2x2} \otimes I_{NN}$, where $\sigma^z = (\sigma_x, \sigma_y, \sigma_z)$ are Pauli matrices acting on spin, $I_{2x2}$ acts on the orbital degree of freedom, and $I_{NN}$ acts on the layer index. $\langle S^z \rangle(k_{\parallel})$ is evaluated at $k_{\parallel} = k_{\perp,\mu} \hat{x}$ and shown for $\mu = \mu_{TI}, \mu_{TM}, \mu_M$ in Fig. 2(a). We see here that, in the absence of hybridization ($g = 0$), the spin polarization stays maximal in the TI and TM regimes while rapidly dropping upon entering the M regime. Another quantity of experimental interest is the total spin magnitude associated with the Dirac states with in-plane momentum $k_{\parallel}$ defined in terms of spin polarization as

$$S(k_{\parallel}) = \sqrt{\sum_{i=x,y,z} \langle (S_i^z(k_{\parallel}))^2 \rangle}.$$  

Fig. 2(a) and 2(b) show that the spin-momentum locking quantified using these measures clearly distinguishes the TM regime from the ordinary metal regime (M) in the absence of hybridization.

C. Effects of S-B Hybridization

We now turn to the effects of hybridization. One effect of hybridization that is manifest in the experimental
the TM regime, namely how the bulk-Dirac energy gap on the two experimentally accessible characteristics of reduction in the spin polarization.

serves the spin-texture winding despite the quantitative reduction upon hybridization. This is in qualitative agreement with the low values of spin polarization and our DFT results on 3D TIs exhibit a clear energy gap between the Dirac branch and the bulk states at a chemical potential common in 3D TI materials.

Another effect of hybridization is to broaden the Dirac state wavefunctions in the TI and TM regimes. The degree of broadening depends on the chemical potential \( \mu \), hybridization strength \( g \), and the slab thickness \( N \). However, as long as \( g \) is the smallest energy scale in the total Hamiltonian as is the case for Figs. (b-d), the Dirac states in the TI and TM regimes remain localized on the surfaces. A tangible consequence of the wavefunction broadening is the quantitative suppression of the spin-momentum locking. As mentioned earlier, in the absence of hybridization the Dirac states of TI and TM exhibit a maximal degree of spin-momentum locking. However, hybridization rotates the spin vectors of different atomic orbitals and layers away from the direction perpendicular to the in-plane momentum. Hence, both measures of spin-momentum locking shown in Fig. 2 show quantitative reduction upon hybridization. This is in qualitative agreement with the low values of spin polarization and total spin magnitude found in a first-principle calculation of a thin slab in a previous work and our DFT results in the next section. Note that the hybridization still preserves the spin-texture winding despite the quantitative reduction in the spin polarization.

Finally, we study how the effects of hybridization on the two experimentally accessible characteristics of the TM regime, namely how the bulk-Dirac energy gap \( \Delta_{BB}(\mu) \) and the spin polarization \( \langle S_y \rangle(k_x) \) of a Dirac state, vary with the slab thickness. Since the quantized energy spacings due to finite size effects decreases with increasing slab thickness, we consider a dimensionless measure that quantifies the bulk-Dirac energy gap:

\[
r(\mu) \equiv \frac{\Delta_{DB}(\mu)}{\Delta_{BB}(\mu)},
\]

where \( \Delta_{BB}(\mu) \equiv E_{B,2N+1}^{(0)}(k_{\parallel},\mu) - E_{D,3}^{(0)}(k_{\parallel},\mu) \) is the energy spacing in the presence/absence of hybridization between the two lowest lying conduction bulk branches measured at the same in-plane momentum \( k_{\parallel} \). We calculate the electronic structure of Bi\(_2\)Se\(_3\)(111) slabs of 4-6 QLs using the \textsc{Vasp} code with 26,27. A detailed study of thin slabs as shown in Fig. 2(a) to (e), we find that the key effect of hybridization that is spectroscopically detectable is the increase in \( \Delta_{DB}(\mu) \) in both TM and M regimes compared to the TI regime. Otherwise the spectra in the absence or presence of hybridization look similar. Note that most ARPES data on 3D TIs exhibit a clear energy gap between the Dirac branch and the bulk states at a chemical potential that is spectroscopically detectable is the increase in the bulk-Dirac state energy gap. We quantify this energy gap, for a given chemical potential \( \mu \), using the energy difference between a Dirac state above \( \mu \), and its bulk counterpart.

\[
\Delta_{DB}(\mu) \equiv E_{B,2N-1}^{(0)}(k_{\parallel},\mu) - E_{D,3}^{(0)}(k_{\parallel},\mu)
\]

in the presence/absence of hybridization. Comparing Fig. 3(a) to (f), we find that the key effect of hybridization that is spectroscopically detectable is the increase in \( \Delta_{DB}(\mu) \) in both TM and M regimes compared to the TI regime. Otherwise the spectra in the absence or presence of hybridization look similar. Note that most ARPES data on 3D TIs exhibit a clear energy gap between the Dirac branch and the bulk states at a chemical potential common in 3D TI materials.

Another effect of hybridization is to broaden the Dirac state wavefunctions in the TI and TM regimes. The degree of broadening depends on the chemical potential \( \mu \), hybridization strength \( g \), and the slab thickness \( N \). However, as long as \( g \) is the smallest energy scale in the total Hamiltonian as is the case for Figs. 2(b-d), the Dirac states in the TI and TM regimes remain localized on the surfaces. A tangible consequence of the wavefunction broadening is the quantitative suppression of the spin-momentum locking. As mentioned earlier, in the absence of hybridization the Dirac states of TI and TM exhibit a maximal degree of spin-momentum locking. However, hybridization rotates the spin vectors of different atomic orbitals and layers away from the direction perpendicular to the in-plane momentum. Hence, both measures of spin-momentum locking shown in Fig. 2 show quantitative reduction upon hybridization. This is in qualitative agreement with the low values of spin polarization and total spin magnitude found in a first-principle calculation of a thin slab in a previous work and our DFT results in the next section. Note that the hybridization still preserves the spin-texture winding despite the quantitative reduction in the spin polarization.

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\]

where \( \Delta_{BB}(\mu) \equiv E_{B,2N+1}^{(0)}(k_{\parallel},\mu) - E_{D,3}^{(0)}(k_{\parallel},\mu) \) is the energy spacing in the presence/absence of hybridization between the two lowest lying conduction bulk branches measured at the same in-plane momentum \( k_{\parallel} \). We calculate the electronic structure of Bi\(_2\)Se\(_3\)(111) slabs of 4-6 QLs using the \textsc{Vasp} code with 26,27.

**III. DFT CALCULATIONS OF THIN Bi\(_2\)Se\(_3\) SLABS**

Now we turn to an ab-initio study of thin slabs to compare with the simple phenomenological model of hybridization we explored in the previous section. The approach of the previous section is limited, in the sense that it builds on a low-energy effective description of the band structure, and that there is no detailed knowledge of the hybridization strength \( g \) which could in principle be \( k_{\parallel} \)-dependent. On the other hand, the DFT approach on slabs, which does not require calculating surface and bulk separately as in the calculations of semi-infinite systems, is limited to very thin films of several QLs due to computational limits. By combining the two approaches, we extract a more robust understanding of the effects of hybridization in the TM regime and implications on their trends over film thickness.

We calculate the electronic structure of Bi\(_2\)Se\(_3\)(111) slabs of 4-6 QLs using the \textsc{Vasp} code with 26,27.
the projector-augmented-wave method\textsuperscript{28} within the generalized-gradient approximation (GGA)\textsuperscript{29}. Spin-orbit coupling is included self-consistently. We use experimental lattice constants\textsuperscript{30} and an energy cutoff of 420 eV with a $31 \times 31 \times 1$ $k$-point grid. Our DFT calculations are limited up to 6 QLs. For 5-6 QLs, the overlap between top and bottom surface states is already very small yielding an energy gap of the order of meV at $\Gamma$. Expectation values of spin components $\langle S_x \rangle$, $\langle S_y \rangle$, $\langle S_z \rangle$ are calculated from the summation of the expectation values of each atom.

![Diagram](image)

**FIG. 4.** (a) DFT-calculated band structure of a 6-QL slab of Bi$_2$Se$_3$. (b) DFT-calculated spin expectation values of the conduction Dirac state $\langle S_i \rangle(k_x \hat{x})$ for a 6-QL Bi$_2$Se$_3$ slab.

**FIG. 5.** (a) Ratio $r = \Delta_{DB}/\Delta_{BB}$ within the TM regime, calculated at a fixed $k_y$. (b) $\langle S_y \rangle$ of the conduction Dirac state calculated using DFT as a function of slab thickness $N$.

Figure\textsuperscript{3} shows the DFT-calculated band structure and spin expectation values $\langle S_i \rangle(k_x \hat{x})$ of a 6-QL slab. The surface states are doubly degenerate and have a Dirac dispersion and we show five confined states in the bulk conduction band region [Fig. 4(a)]. For small $|k_x|$ values, $\langle S_y \rangle$ of a Dirac conduction state is clearly dominant over other components and exhibits spin-momentum locking [Fig. 4(b)]. As $|k_x|$ increases, a small $z$ component of spin expectation value develops. However, over the entire range of $k_x$, $\langle S_y \rangle$ is much less than the maximal value, in agreement with previous DFT studies.\textsuperscript{13} A comparison between Fig.\textsuperscript{2} and Fig.\textsuperscript{4} indicates that our hybridization model is an effective way to capture the broadening of the Dirac surface state wavefunction and the resulting reduction in the spin polarization and the total spin magnitude.\textsuperscript{31}

Now we discuss the thickness dependence in the bulk-Dirac energy gap measure and the spin polarization. We calculate the dimensionless measure of bulk-Dirac energy gap $r = \Delta_{DB}/\Delta_{BB}$ in the TM regime at the $k_{||}$ point where the Dirac surface state branch has slightly higher energy than the bottom of the conduction band $E_c$, as indicated in Fig.\textsuperscript{4}(a). We find that the ratio $\Delta_{BB}(N_1)/\Delta_{BB}(N_2)$ is close to $(N_2/N_1)^2$ at the $k_{||}$ point of interest as expected of finite-size-effect origin of the scale $\Delta_{BB}(N)$. Surprisingly, despite the small range of thickness accessible to the slab DFT calculation, the dimensionless measure of bulk-Dirac energy gap $r = \Delta_{DB}/\Delta_{BB}$ in Fig.\textsuperscript{4}(a) shows a significant increase upon an increase in the slab thickness. This is qualitatively consistent with observations from the effective model and hybridization effects in Sec. II. On the other hand, the range of thickness in the present calculation appears to be too small to show any change in the $\langle S_y \rangle$ as a function of slab thickness [Fig.\textsuperscript{5}(b)].

**IV. CONCLUSION**

We combined a Fano-type hybridization model calculation with an ab-initio slab calculation to study the lowest order effects of surface-bulk interaction in topological insulators with a particular focus in the TM regime. We defined the TM regime of a topological insulator to be where the Dirac surface states and bulk states coexist and interact, yet the spin-winding is preserved albeit with a reduced spin-polarization magnitude. The hybridization model presented in Sec. II captures the spin-polarization reduction of the Dirac states originating from the hybridization with bulk states. Given the metallic behavior of most TIs, and the experimental evidence of reduced spin polarization, our simple model offers a useful starting point for applications of TIs which need to take real materials in the TM regime into account. Moreover, the hybridization-driven bulk-Dirac energy gap explains why the Dirac branch shows up so well separated from bulk states in ARPES experiments. Note that this energy gap and the suppression of total spin magnitudes are both experimentally observed phenomena that cannot be accessed by the typical approach of coupling a single “surface layer” to a bulk electronic structure to include surface states in semi-infinite systems as in Ref.\textsuperscript{23}

We propose SARPES experiments for films of varying thickness to test our predictions for hybridization-driven suppression of spin polarization for further vindication of the model.

Promising future directions include developing schemes to extend DFT calculations to thicker systems. This would enable us to track thickness dependence in spin polarization and extract more detailed knowledge of $k_{||}$-dependence of the hybridization strength $g$. Preliminary DFT results show that $g(k_y)$ has a significant $k_{||}$-dependence. Another interesting direction will be
to study consequences of the hybridization effect on transport properties. Many puzzling aspects of transport experiments have been attributed to the presence of bulk states or surface-bulk interaction. Our simple model offers a starting point to theoretically address the effects of surface-bulk interaction on transport.

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1 S. Kim, M. Ye, K. Kuroda, Y. Yamada, E. E. Krasovskii, E. V. Chulkov, K. Miyamoto, M. Nakatake, T. Okuda, Y. Ueda, K. Shimada, H. Namatame, M. Taniguchi, and A. Kimura, Phys. Rev. Lett., 107, 056803 (2011).
2 S. R. Park, W. S. Jung, C. Kim, D. J. Song, C. Kim, S. Kimura, K. D. Lee, and N. Hur, Phys. Rev. B, 81, 041405 (2010).
3 M. Bianchi, D. Guan, S. Bao, J. Mi, B. B. Iversen, P. D. King, and P. Hofmann, Nat Commun, 1, 128 (2010).
4 H. Steinberg, J.-B. Laloe, V. Fatemi, J. S. Moodera, and P. Jarillo-Herrero, Phys. Rev. B, 84, 233101 (2011).
5 Y. Zhang, K. He, C.-Z. Chang, C.-L. Song, L.-L. Wang, X. Chen, J.-F. Jia, Z. Fang, X. Dai, W.-Y. Shan, S.-Q. Shen, Q. Niu, X.-L. Qi, S.-C. Zhang, X.-C. Ma, and Q.-K. Xue, Nat Phys, 6, 584 (2010). ISSN 1745-2473.
6 N. Bansal, Y. S. Kim, M. Brahlke, E. Edrey, and S. Oh, Phys. Rev. Lett., 109, 116804 (2012).
7 M. Liu, C.-Z. Chang, Z. Zhang, Y. Zhang, W. Ruan, K. He, L.-1. Wang, X. Chen, J.-F. Jia, S.-C. Zhang, Q.-K. Xue, X. Ma, and Y. Wang, Phys. Rev. B, 83, 165440 (2011).
8 Y. S. Kim, M. Brahlke, N. Bansal, E. Edrey, G. A. Kapilevich, K. Iida, M. Tanimura, Y. Horibe, S.-W. Cheong, and S. Oh, Phys. Rev. B, 84, 073109 (2011).
9 D. Hsieh, Y. Xia, D. Qian, L. Wray, J. H. Dil, F. Meier, J. Osterwalder, L. Patthey, J. G. Checkelsky, N. P. Ong, A. V. Fedorov, H. Lin, A. Bansil, D. Grauer, Y. S. Hor, R. J. Cava, and M. Z. Hasan, Nature, 460, 1101 (2009). ISSN 0028-0836.
10 Z.-H. Pan, E. Vesovo, A. V. Fedorov, D. Gardner, Y. S. Lee, S. Chu, G. D. Gu, and T. Valla, Phys. Rev. Lett., 106, 257004 (2011).
11 C. Joziwiajk, Y. L. Chen, A. V. Fedorov, J. G. Analytis, C. R. Rotunden, A. K. Schmid, J. D. Denlinger, Y.-D. Chung, D.-H. Lee, I. R. Fisher, R. J. Birgeneau, Z.-X. Shen, Z. Hussain, and A. Lanzara, Phys. Rev. B, 84, 165113 (2011).
12 C. Joziwiajk, C.-H. Park, K. Gotlieb, C. Hwang, D.-H. Lee, S. G. Louie, J. D. Denlinger, C. R. Rotunden, R. J. Birgeneau, Z. Hussain, and A. Lanzara, Nat Phys, 9, 293 (2013). ISSN 1745-2473.
13 O. V. Yazyev, J. E. Moore, and S. G. Louie, Phys. Rev. Lett., 105, 266806 (2010).
14 Y. L. Chen, J. G. Analytis, J.-H. Chu, Z. K. Liu, S.-K. Mo, X. L. Qi, H. J. Zhang, D. H. Lu, X. Dai, Z. Fang, S. C. Zhang, I. R. Fisher, Z. Hussain, and Z.-X. Shen, Science, 325, 178 (2009). http://www.sciencemag.org/content/325/5937/178.full.pdf.
15 L. Fu, Phys. Rev. Lett., 103, 266801 (2009).
16 K. Kuroda, M. Arita, K. Miyamoto, M. Ye, J. Jiang, A. Kimura, E. E. Krasovskii, E. V. Chulkov, H. Iwasawa, T. Okuda, K. Shimada, Y. Ueda, H. Namatame, and M. Taniguchi, Phys. Rev. Lett., 105, 076802 (2010).
17 I. Garate and M. Franz, Phys. Rev. Lett., 104, 146802 (2010).
18 T. Yokohama, J. Zang, and N. Nagaosa, Phys. Rev. B, 81, 241410 (2010).
19 M. Fischer, A. Vaezi, A. Manchon, and E.-A. Kim, “Large spin torque in topological insulator/ferromagnetic metal bilayers,” ArXiv:1305.1328.
20 D. L. Bergman and G. Refael, Phys. Rev. B, 82, 195417 (2010).
21 G. D. Mahan, Many-Particle Physics, 3rd ed. (Plenum, New York, N.Y., 2000).
22 A similar discretization was used in other papers, e.g., Rosenberg and Franz, Phys. Rev. B 85, 195119 (2012).
23 H. Zhang, C.-X. Liu, X.-L. Qi, X. Dai, S.-C. Zhang, Nat Phys, 5, 438 (2009). ISSN 1745-2473.
24 M. Konig, H. Buhmann, L. W. Molenkamp, T. Hughes, C.-X. Liu, X.-L. Qi, and S.-C. Zhang, Journal of the Physical Society of Japan, 77, 031007 (2008).
25 The lattice Hamiltonian $h_0(k_x, k_z)$ does not have in-plane three-fold rotational symmetry of the continuum Hamiltonian. However, we do not expect this to change any of our conclusions in a qualitative manner.
26 G. Kresse and J. Furthmüller, Phys. Rev. B, 54, 11169 (1996).
27 G. Kresse and J. Furthmüller, Comp. Mat. Sci., 6, 15 (1996).
28 P. E. Blöchl, Phys. Rev. B, 50, 17953 (1994).
29 J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett., 77, 3865 (1996).
30 S. Nakajima, J. Phys. Chem. Solids, 24, 479 (1963).
31 Our DFT calculations also show evidence of the hexagonal warping effect $2k_z (k_z)$ for $k_z \geq 0.08\pi/a$.