

Axion Insulator States in a Topological Insulator Proximitized to Magnetic Insulators: A Tight-Binding Characterization

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Recent discovery of axion states in materials such as antiferromagnetic topological insulators boosted investigations of magnetoelectric response in topological insulators and their promise towards dissipationless topological electronics. In this paper, we develop a tight-binding methodology to explore the emergence of axion states in Bi_2Se_3 in proximity to magnetic insulators on top and bottom surfaces. The topological protection of the surface states is lifted by a time-reversal-breaking perturbation due to proximity of a magnetic insulator and a gap is opened on the surfaces, giving rise to half-quantized Hall conductance and zero Hall plateau - evidencing an axion insulator state. We developed a real-space tight-binding Hamiltonian for Bi_2Se_3 using first-principles data. Transport properties of the system were obtained within the Landauer-Büttiker formalism, and discuss the creation of axion states through Hall conductance and zero Hall plateau at the surfaces, as a function of proximitized magnetization and corresponding potentials at the surfaces, as well as the thickness of the topological insulator.

I. INTRODUCTION

Axions are hypothetical particles introduced 40 years ago in quantum field theory, to solve the strong charge-parity problem in high-energy physics. However, they remained evasive to any observation in nature to date¹. Many particles discovered or hypothesized in particle physics to resolve a certain contradiction are found by analogy as quasi-particle excitations in condensed matter physics, such as massless Weyl, Dirac fermions, and Majorana fermions, to name a few². Axions were not an exception - weakly interacting quasiparticles in materials such as antiferromagnetic topological insulators (AFM TIs) were shown to exhibit governing dynamics that is similar to axion particles in high-energy physics^{3,4}. Furthermore, the θ term in the topological magnetoelectric effect (TME) was shown to have the same form as the action describing the coupling between an axion particle and a photon⁵. The appearance of axion states in condensed matter systems is accompanied by several interesting phenomena such as the half-quantized anomalous Hall effect (QAHE) and the linear magnetoelectric response¹.

Subsequent studies in particle physics and astrophysics indicated that axions are candidate for dark matter. Identification of such dark axions is a challenging task due to their weak interaction with ordinary materials. Recently, it was proposed that topological insulators with a dynamical axion field can be utilized to detect dark axions, with theoretical prediction of the mass of these particles of 0.7-3.5 meV⁶.

Therefore, condensed matter systems based on topological insulators that can host axion quasi-particles - dubbed axion insulators (AI) - are of prime scientific interest. Several works on axion states in magnetic topological insulator MnBi_2Te_4 have readily been reported in the literature⁷⁻¹⁰. They develop an effective continuum

Hamiltonian close to the Γ point and then approximate the original tight-binding Hamiltonian for low energies before discretizing the continuum Hamiltonian on a cubic lattice. In doing so, they do not address effects related to the symmetry of the structure as well as higher energy outcomes. However, their performance can likely be exceeded by a suitable design of ferromagnet-TI heterostructures. Their magnetization and surface states can be tailored individually as they belong to different constituent materials, to achieve controllable and stable performance even at room temperature. This is the main objective of our theoretical investigation in the present manuscript. The axion insulators are characterized by a half-quantized Hall conductance with opposite sign and chiral currents on the surfaces, the latter being clockwise and counterclockwise on opposite surfaces. The electromagnetic response of a three-dimensional insulator can be expressed by Maxwell's action^{11,12}:

$$S = \frac{8}{\pi} \int d^3x dt (\varepsilon E^2 - \frac{1}{\mu} B^2), \quad (1)$$

in which \mathbf{E} and \mathbf{B} are electric and magnetic field inside the insulator, and ε and μ represent dielectric constant and magnetic permeability, respectively. For insulators with broken time-reversal symmetry (TRS), spatial symmetry or a combination of spatial symmetry and TRS can lead to quantized θ . The axion term should be added to the action as:

$$S_\theta = \frac{\theta}{2\pi} \frac{\alpha}{2\pi} \int d^3x dt \mathbf{E} \cdot \mathbf{B}, \quad (2)$$

in which θ acts as an axion field. Here $\alpha = e^2/\hbar c$ is the fine structure constant. As $\mathbf{E} \cdot \mathbf{B}$ is odd under TRS, S_θ must be invariant when $\theta \rightarrow -\theta$. The axion field (θ) for time-reversal invariant insulators is quantized to 0 for normal insulators or π for strongly topological ones^{1,13}. A schematic classification of TIs is depicted in Fig. 1 in

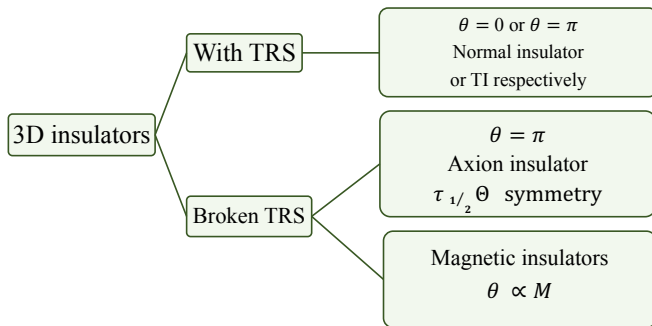


FIG. 1. Schematic classification of 3D insulators based on (broken) TRS and the axion term θ ⁵.

terms of the existence or breaking of TRS. In the presence of TRS, $\theta = 0$ or $\theta = \pi$ correspond to normal insulators or TIs, respectively. For broken TRS, TI can be an axion insulator (AI) or a magnetic insulator. In AI, although TRS is broken, a combination of TRS and lattice translation symmetry is conserved, which is known as effective TRS and $\theta = \pi$. Magnetic insulators can have any arbitrary value of θ proportional to the magnetic order parameter M , i.e. $\theta(r, t) \propto \delta M(r, t)$ ⁵. The axion term states that an electric field induces magnetic polarization and a magnetic field induces electric polarization. θ depends on the band structure of the insulator and can be expressed as follows¹³:

$$\theta = \frac{1}{4\pi} \int d^3k \epsilon^{ijk} \text{Tr}[A_i \partial_j A_k + i \frac{2}{3} A_i A_j A_k], \quad (3)$$

with $A_j = -i \langle u_n | \frac{\partial}{\partial k_j} | u_m \rangle$

The topological magnetoelectric effect can be defined as a linear magnetoelectric polarization¹³:

$$\alpha_{ij} = \frac{\partial M_j}{\partial E_i} \Big|_{B=0} = \frac{\partial P_i}{\partial B_j} \Big|_{E=0} = \tilde{\alpha}_{ij} + \theta \frac{\alpha}{4\pi^2} \delta_{ij}, \quad (4)$$

where the tensor $\tilde{\alpha}_{ij}$ contains nine independent components for different contribution of spin, orbitals, and ions; for materials with inversion and time-reversal symmetry $\tilde{\alpha}_{ij} = 0$. Here we focus on magnetoelectric coupling resulting from the orbital magnetization and polarization. Axion insulator results in a TME in which an energy gap opens on the surfaces and the bulk quantization such that $\theta = \pi$.

A gap is opened on the surfaces when surface states are broken by a time-reversal-breaking perturbation through an external magnetic field, or if TI is placed in proximity to a magnetic insulator, or in presence of magnetic impurities on the 3D TI surfaces. In such cases, the surface Hall conductivity becomes $\sigma_{xy} = \pm \frac{e^2}{2h}$, where sign depends on the specific time-reversal breaking perturbation^{14,15}. If the direction of magnetization of the ferromagnetic (FM) materials which is in proximity to the TI is the same, the quantum Hall conductance is half-

quantized and $\sigma_{xy} = +\frac{e^2}{2h}$ for the top and bottom surfaces; therefore a chiral current emerges at the TI surfaces. For different directions of magnetization on the top and bottom surfaces, $\sigma_{xy} = \pm \frac{e^2}{2h}$ changes sign at surfaces, with no Hall current^{16,17}.

Some applications of axion insulators include antiferromagnetic topological insulators and their axion polaritons to detect dark matter, fabrication of non-reciprocal thermal emitters by Weyl semimetals, and detection of Majorana fermions in quantum anomalous Hall materials for topological quantum computing¹. Faster advances in this field require the development of an efficient and convenient theoretical methodology to design and characterize the axion states in hybrid TI heterostructures, as well as detecting and examining these states at various energy scales. The latter is the ultimate objective of our present manuscript, as exemplified through full characterization of axion insulator states in a ferromagnet-Bi₂Se₃-ferromagnet heterostructure after developing the tight-binding Hamiltonian for Bi₂Se₃ with proximity effect included using the Zeeman term. We are dealing with a three-dimensional system, where developing a tight-binding model is challenging. We started from the microscopic scale and used the DFT data to fit a real-space Hamiltonian with the obtained band structure. Although this model is an effective one, it is well-suited to capture experimental results such as the hybridization gap.

The paper is organized as follows. In Sec. II, we employ the symmetries to present the real-space Hamiltonian based on the parameters of the four-band Hamiltonian for Bi₂Se₃. In Sec. III, the effects of finite thickness and the hybridization gap are considered. The hybridization effect is calculated and the proximity effect on the surface states is detailed. In Sec. IV, we develop the transport methodology and exemplify its use to characterize axion states in a Bi₂Se₃ proximitized to magnetic insulators, by calculating the longitudinal, transverse and Hall conductance as well as the axion term θ . Sec. V summarizes our results and presents our conclusions.

II. THE MODEL HAMILTONIAN

The Bi₂Se₃ family of materials possess a rhombohedral crystal structure with the symmetry group $D_{3d}^5(R\bar{3}m)$ ¹⁸, as shown in Fig. 2. In the layered structure of Bi₂Se₃, atoms in layers are stacked along the z -direction in the ABCABC sequence, with 5 layers in the unit cell; those five atomic layers are therefore usually referred to as a Quintuple Layer (QL)^{19–21}. Each lattice site has 6 nearest neighbors in each layer and 6 next-nearest neighbors in adjacent layers. The vectors \mathbf{a}_i to connect sites within each layer and vectors \mathbf{b}_i to connect sites to adjacent layers can be written as¹⁹:

$$\mathbf{a}_1 = (a, 0, 0), \quad \mathbf{a}_2 = (-a/2, a\sqrt{3}/2, 0), \quad \mathbf{a}_3 = (-a/2, -a\sqrt{3}/2, 0),$$

$$\mathbf{b}_1 = (0, \sqrt{3}a/3, c), \quad \mathbf{b}_2 = (-a/2, -\sqrt{3}a/6, c), \quad \text{and} \quad \mathbf{b}_3 = (a/2, \sqrt{3}a/6, c).$$

The ruling

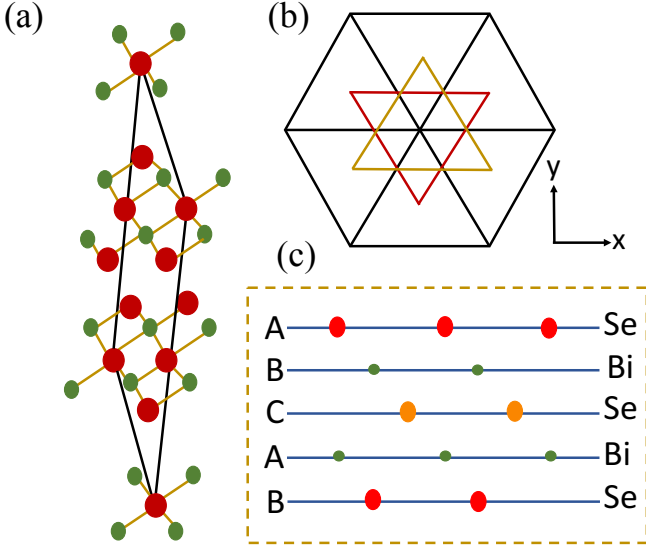


FIG. 2. (a) Bi_2Se_3 crystal structure. (b) Top view of atomic sites. (c) The atoms are stacked in layers in the z -direction in the ABCABC sequence.

| T | P | $\Gamma^{(1)}$ | $\Gamma^{(2)}$ | $\Gamma^{(3)}$ |
|---|---|--|----------------|---|
| | | Dim=1 | Dim=2 | Dim=3 |
| + | + | $\{k_z^2\}$ $\{k_\perp\}$ Γ_5 | | $\{ik_z k_+, -ik_z k_-\}$ $\{k_-^2, k_+^2\}$ |

TABLE I. Polynomials of \mathbf{k} and the Γ matrices under the transformations of the group D_3 , inversion and time reversal symmetry. $\Gamma_5 = \sigma_0 \otimes \tau_3$ and $\Gamma^{(1)}, \Gamma^{(2)}, \Gamma^{(3)}$ are irreducible representations of group D_3 . \mathbf{k} polynomials can only exist in Hamiltonian according to the irreducible representations of group D_3 , as given in the table.

structural symmetries are as follows^{18,19,21}:

- Three rotational symmetry around the z -axis:
 $R_3 = \exp[(i\sigma_3 \otimes 1)/2]\theta$, $\theta = 2\pi/3$.
- Two rotational symmetry around the x -axis:
 $R_2 = [(i\sigma_2 \otimes \tau_3)]$.
- Inversion symmetry: $P = 1 \otimes \tau_3$.
- Time-reversal symmetry: $T = (i\sigma_2 \otimes 1)K$, where K is a complex-conjugate operator, σ and τ are spin and pseudospin Pauli matrices.

D_{3d} is the direct multiplication of the D_3 group and the inverse operator group. Therefore, group D_3 with spatial inversion and time-reversal eigenvalues is considered¹⁸.

Table I represents polynomials of \mathbf{k} and the Γ matrices under the transformations of the group D_3 , inversion and time reversal. By considering the irreducible representation, a four-band Hamiltonian was written as follows²¹:

$$H = \varepsilon'_k \otimes I + \begin{pmatrix} M'_k & A'_1 k_z & 0 & A'_2 k_- \\ A'_1 k_z & -M'_k & A'_2 k_- & 0 \\ 0 & A'_2 k_+ & M'_k & -A'_1 k_z \\ A'_2 k_+ & 0 & -A'_1 k_z & -M'_k \end{pmatrix}, \quad (5)$$

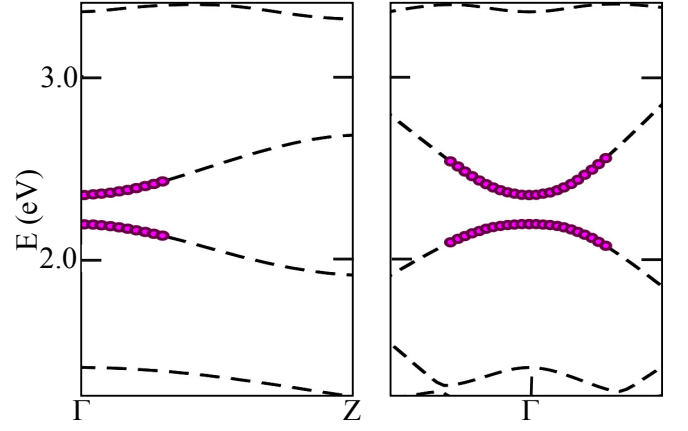


FIG. 3. Band structure of Bi_2Se_3 near the Γ point. Dashed lines represent DFT data and solid dots show the band structure from the four-band Hamiltonian with fitted parameters near the Γ point.

where $\varepsilon'_k = C'_0 + C'_1 k_z^2 + C'_2 k_\perp^2$, $M'_k = M'_0 + M'_1 k_z^2 + M'_2 k_\perp^2$ and $k_\perp^2 = k_x^2 + k_y^2$. By fitting the band structure obtained from this Hamiltonian with the band structure determined from DFT²², we obtain $A'_0 = 5.35 \text{ eV \AA}$, $B'_0 = 4.44 \text{ eV \AA}$, $C'_0 = -0.0063 \text{ eV \AA}$, $C'_1 = 6.65 \text{ eV \AA}^2$, $C'_2 = -1.75 \text{ eV}^2$, $M'_0 = 0.29 \text{ eV \AA}$, $M'_1 = -10.25 \text{ eV \AA}^2$, $M'_2 = -56.6 \text{ eV \AA}^2$ which are similar to the values found in the work of Liu *et al.*²¹.

In this section, a real-space tight-binding Hamiltonian is constructed whose hopping parameters are related to the parameters of effective four-band Hamiltonian near the Γ point, introduced by Chu *et al.*²³. It should also be emphasized that, even though we are dealing with an effective tight-binding model, other effects such as gate voltage, electric field, magnetic field, and strain can be straightforwardly added to the consideration based on the symmetry group of the system. To construct a tight-binding Hamiltonian, each unit cell was considered to contain two atoms (Bi, Se) and spin; giving rise to four states for each unit cell. We considered only the hopping between nearest neighbors. Fig. 4 depicts the structure of Bi_2Se_3 in the x - y plane. Considering the nearest neighbors only, each unit cell will have 6 neighboring unit cells in the x - y plane that are connected by vectors \mathbf{n}_i ($i = 1, 2, 3$) and two unit cells in the z direction connected by vector \mathbf{n}_4 . The real-space Hamiltonian is written as follows:^{23,24}

$$H = \sum_i c_i^\dagger E_{on} c_i + \sum_{i,\alpha} (c_i^\dagger T_\alpha c_{i+\alpha} + H.c), \quad (6)$$

where $\alpha = \mathbf{n}_1, \mathbf{n}_2, \mathbf{n}_3, \mathbf{n}_4$, and the operator c_i^\dagger (c_i) creates (annihilates) an electron at site i . Further we have:

$$E_{on} = (E_0 - 2\sum_\alpha B_\alpha) \sigma_z \otimes \sigma_0, \quad (7)$$

and

$$T_\alpha = C_\alpha \sigma_0 \otimes \sigma_0 + B_\alpha \sigma_z \otimes \sigma_0 - i \left(\frac{A_\alpha}{2} \right) \sigma_x \otimes \sigma \cdot \mathbf{n}_\alpha. \quad (8)$$

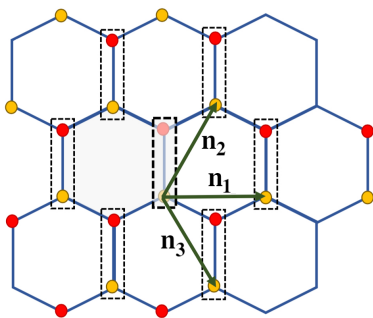


FIG. 4. Lattice structure of Bi_2Se_3 in the x-y plane. Each unit cell has 6 neighboring unit cells in the x-y plane that are connected by vectors \mathbf{n}_i ($i=1,2,3$), and two unit cells in the z direction that are connected by vector \mathbf{n}_4 .

| Hybridization gap (eV) | | | | |
|------------------------|-------|-------|-------|-------|
| | 2 QLs | 3 QLs | 4 QLs | 5 QLs |
| Our model | 0.257 | 0.118 | 0.057 | 0.028 |
| Ref. 26 | 0.252 | 0.138 | 0.070 | 0.041 |

TABLE II. Hybridization gap due to finite-size effect. First-row are obtained from our model and the second-row are from Ref. 26.

E_{on} and T_α are 4×4 matrices corresponding to the hopping parameters and stand for onsite energy and hopping parameters between unit cells respectively. Based on the structure of the system $\mathbf{n}_1 = (1/2, \sqrt{3}/2, 0)$, $\mathbf{n}_2 = (-1/2, \sqrt{3}/2, 0)$, $\mathbf{n}_3 = (1, 0, 0)$, $\mathbf{n}_4 = (0, 0, 1)$. Because of the symmetries of the system, A_{n_1, n_2, n_3} , B_{n_1, n_2, n_3} , and C_{n_1, n_2, n_3} that are related to the x-y plane parameters are isotropic and therefore we use the notation $A_{n_1, n_2, n_3} = A_2$, $B_{n_1, n_2, n_3} = B_2$, $C_{n_1, n_2, n_3} = C_2$, while for n_4 parameters we take $A_{n_4} = A_1$, $B_{n_4} = B_1$, and $C_{n_4} = C_1$. By matching this Hamiltonian with the four-band Hamiltonian near the Γ point, i.e for $k \rightarrow 0$, the parameters of the tight-binding Hamiltonian can be determined.

III. FINITE SIZE AND PROXIMITY EFFECTS ON SURFACE STATES

The Bi_2Se_3 surface Hamiltonian in the basis of top and bottom states with spin \uparrow, \downarrow $\{t \uparrow, t \downarrow, b \uparrow, b \downarrow\}$ can be written as follows^{11,25}:

$$H_{surf}(k_x, k_y) = \hbar v_F (k_y \sigma_x - k_x \sigma_y) \otimes \sigma_z + \Delta_m \sigma_0 \otimes \sigma_x, \quad (9)$$

where v_F is the Fermi velocity, $\Delta_m = \Delta_h + Bk^2$ represents the coupling between the top and bottom states and Δ_h denotes the coupling gap at the surfaces^{26,27}. In the three-dimensional topological insulators family of Bi_2Se_3 , the bulk samples have a gap, while the surface states are gapless and contain a Dirac cone. When the thickness of the sample is reduced to nanoscale, there

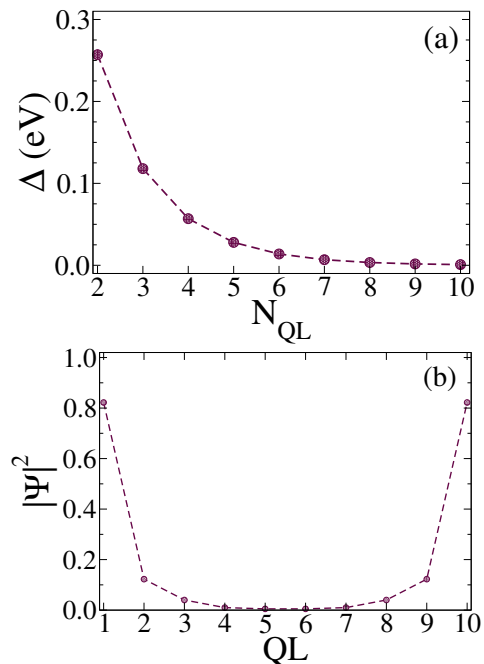


FIG. 5. (a) Hybridization gap due to finite-size effect on surface states, as a function of TI thickness (expressed as a number of quintuple layers (QLs)). (b) Penetration of surface state wave function into the bulk, for a 10 QLs thick sample.

will be an overlap between the wave functions of the top and bottom surface states, and the spin-polarized surface states of one surface combine with the opposite spin components on the other surface, resulting in a gap at the Dirac point. Here, the penetration depth was determined for surface states considering the eigenfunctions of surface states. Then, the hybridization gap was calculated for surface states in several layers along the z-direction. The obtained hybridization gap due to the finite size effect is shown in Fig. 5 and Table II, and is consistent with experimental results²⁶ and DFT data¹¹. One concludes that to prevent surface hybridization, the thickness of the sample should be at least 6 QLs. As mentioned earlier, to investigate the effect of magnetization on the surface states, instead of using a two-dimensional effective surface Hamiltonian, a three-dimensional effective Hamiltonian was employed in real space. Concerning a 2D model, the proximitized magnetization (weak Zeeman field) leads to gaps in the Dirac cones on the surfaces. For a 3D model, however, the application of magnetization results in the formation of a gap in the Dirac cones on the top and bottom surfaces, while the side surfaces remain gapless²³. Fig. 6 illustrates the band structure and gapless single Dirac cone of the surface states at the Γ point. Applying Zeeman splitting term introduces a gap.

Magnetization effect on the top and bottom surfaces can be achieved in two different ways: (i) by doping with magnetic atoms (inclusions) on the top and bottom surfaces, or (ii) by proximitized magnetic material

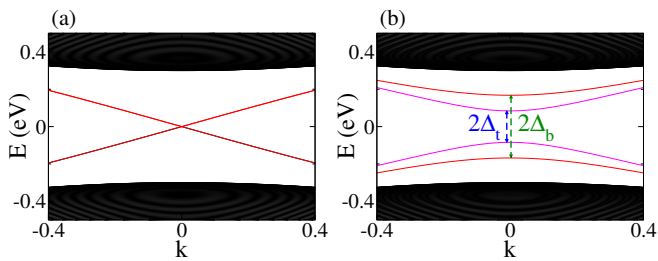


FIG. 6. (a) The band structure and the gapless Dirac cone of the surface states at the Γ point. (b) Applying Zeeman splitting term causes a gap to open; here $\Delta_t = 0.1$ eV and $\Delta_b = -0.2$ eV.

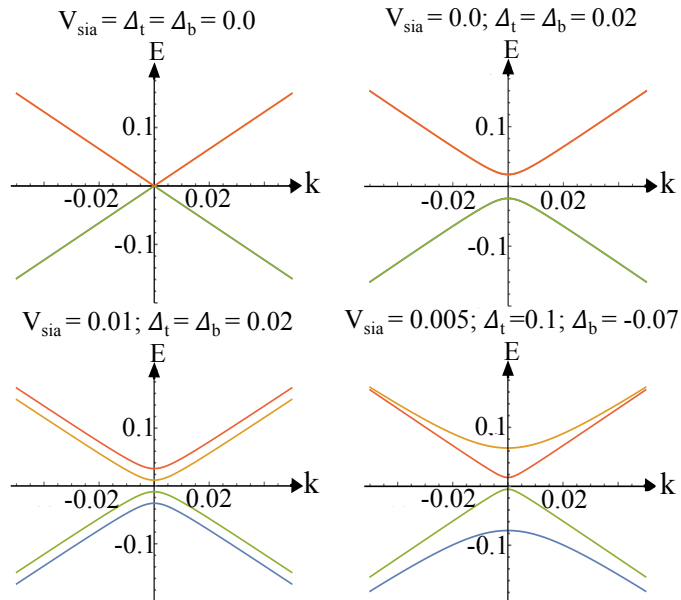


FIG. 7. Effect of magnetization and of structural inversion asymmetry on surface states for different V_{sia} , Δ_t , and Δ_b (in eV).

near those surfaces. Magnetic materials to impose magnetization on the surface states of the TI must be able to establish strong magnetic coupling at the interface while causing no effect on states near the Fermi energy²⁸. Magnetization can be added to the Hamiltonian by the Zeeman term:

$$H_{Zeeman} = \Delta_{t/b}\sigma_0 \otimes \sigma_z, \quad (10)$$

where Δ_t and Δ_b are the exchange fields and represent the gaps opening due to the magnetization applied to the topological surface states in the top and bottom surfaces, respectively^{14,29}. For different magnitudes of the magnetizations on the top and bottom surfaces, the Zeeman term and the gap on the top and bottom surfaces will also be different. Thus, a term related to structural inversion asymmetry (SIA) should be added^{11,25}. This term is a potential difference between two surfaces (V_{sia}) and splits each spin-degenerate band into two subbands. Fig. 7 presents the effect of magnetization and structural

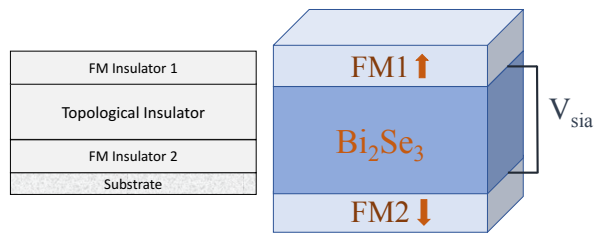


FIG. 8. FM-Bi₂Se₃-FM heterostructure with opposite polarity of magnetization at the two surfaces of the TI. In such a case, the half-quantized Hall conductance will have different sign at the surfaces and the net Hall conductance becomes zero³². Theoretical consideration of the finite thickness of the TI requires additional potential (V_{sia}) to capture the physics due to the structural inversion asymmetry.

inversion asymmetry on TI surface states with a single Dirac cone. Magnetization opens a gap in the Dirac point while V_{sia} tunes the dispersion bands in different directions. The following term should be added to the Hamiltonian as:

$$H_{sia} = V_{sia}\sigma_0 \otimes \sigma_z, \quad (11)$$

IV. AXION INSULATOR STATES IN FM-BI₂SE₃-FM HETEROSTRUCTURE

In what follows, the FM-TI-FM heterostructure shown in Fig. 8 is used to study the emergent axion states and the topological magnetoelectric effect. The essential condition for the topological magnetoelectric effect is lost in the presence of hybridization between the top and bottom surface states. Therefore, the topological insulator should be sufficiently thick to guarantee the absence of hybridization between the top and bottom states³⁰. As discussed in previous section, in case of Bi₂Se₃ the minimum number of quintuple layers to consider is 7. In addition, the exchange fields due to the magnetization applied to the topological insulator at the top and bottom surfaces should have opposite sign (i.e. $\Delta_t\Delta_b < 0$)^{11,31}.

One of the challenges in the fabrication of heterostructures to observe axion states is the proper choice of materials that will be used in proximity of 3D TIs. In addition to structural and magnetic stability, these heterostructures must exhibit robust spin-polarized bandgap in the interface states. A sufficient selection of thin magnetic insulators is nowadays experimentally accessible to sandwich a TI for the purpose of creation of axion states, such as CrI₃, Cr₂Ge₂Te₆, MnBi₂Se₄, and MnBi₂Te₄^{33,34}. For example, for opposite magnetization of the two magnetic layers in a CrI₃-Bi₂Se₃-MnBi₂Se₄ heterostructure, $\Delta_t = 3.2$ meV, $\Delta_b = -26.9$ meV, and $V_{sia} = 9.35$ meV at the Bi₂Se₃ surfaces¹¹. Zero Hall plateau and $\theta = \pi$ are then the main signatures of the axion state.

For characterization of transport/conductance, in this section we consider a four-terminal sample as depicted in Fig. 9. The longitudinal conductance (σ_{xx}) and Hall

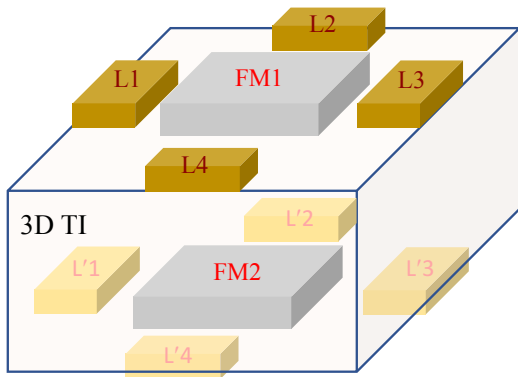


FIG. 9. Schematic illustration of the 3D device with 2D leads to measure the longitudinal (σ_{xx}) and Hall conductance (σ_{xy}) at the surface.

conductance (σ_{xy}) were calculated using the Landauer-Büttiker formalism. This formalism treats conductance in terms of the transmission coefficients of the device. The transmission coefficient between leads i and j can be determined in terms of Green's functions as³⁵:

$$T_{ij} = \text{tr}[\Gamma_i G_{ij} \Gamma_j G_{ij}^\dagger], \quad (12)$$

where Γ_i describes the coupling of the device to the leads and can be expressed in terms of a self-energy Σ_i as $\Gamma_i = i[\Sigma_i - \Sigma_i^\dagger]$. Here Σ_i can be considered as an effective Hamiltonian describing the lead-device interaction³⁶. The Hamiltonian of the complete system can be subdivided as follows:

$$H = H_D + \sum_i (H_L^i + V_{LD}^i + V_{DL}^i), \quad (13)$$

where H_D denotes the device Hamiltonian, H_L^i is the Hamiltonian for the i -th lead, while V_{LD}^i and V_{DL}^i denote left and right Hamiltonian between the i -th lead and device, respectively. In this setup, four identical semi-infinite leads are attached to the sample and the longitudinal and Hall conductance are given by³⁷:

$$\sigma_{xx} = \frac{e^2}{2h} T_{13}, \quad \sigma_{xy} = \frac{e^2}{2h} (T_{14} - T_{12}). \quad (14)$$

A prerequisite for AI states is that the values of $\Delta_{t/b}$ at the surfaces have opposite sign. The localization of the Hall conductance on the surfaces weakly depends on the Zeeman terms as the magnitude of $\Delta_{t/b}$ increases, but we use 2D leads to calculate the Hall conductance, thus the amount of this conductance does not vary³⁸. As explained previously, the Hall conductance is weakly dependent on the Zeeman term chosen, thus we use $\Delta_t = 5$ meV and $\Delta_b = -25$ meV. Our calculated longitudinal and Hall conductance are shown in Fig. 10 for the system with $\Delta_t = 5$ meV and $\Delta_b = -25$ meV. A half-quantized Hall conductance appears upon applying the magnetization to the top and bottom surfaces of a TI due to the

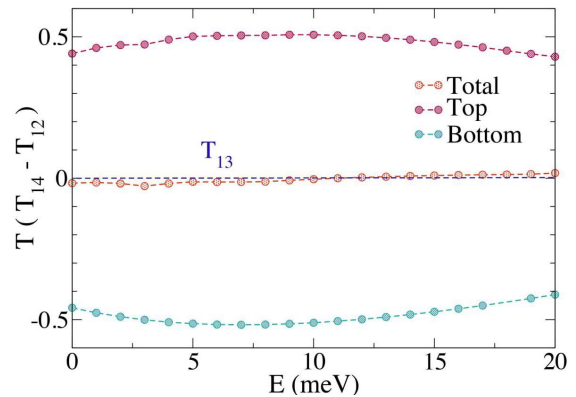


FIG. 10. Hall transmission coefficient for top and bottom surfaces separately and the total Hall conductance, for $\Delta_t = 5$ meV and $\Delta_b = -25$ meV.

configuration of the structure, and can be formulated as^{11,25}

$$\sigma_{xy}^{t/b} = \text{sgn}(\Delta_{t/b}) \frac{e^2}{2h}. \quad (15)$$

It should be noted that calculating/measuring the half-quantization for each surface independently is challenging. As a qualitative indication, Chen *et al.*⁷ recently proposed a nonlocal surface transport device in which the axion insulator can be distinguished from normal insulators without a precise measurement of the half-quantization. Instead, we calculate Hall conductance for the top and bottom surfaces separately as a more definite proof of the axion insulator states. Going beyond the previous work⁷⁻¹⁰, our calculations are more challenging but the results are exceedingly more accurate. For calculation of the quantum transport in this system we employed an original tight-binding Hamiltonian obtained from symmetry considerations and ab-initio calculations, which was a nontrivial task as we had to construct the Landauer-Büttiker formulation for the complex crystal structure of this system.

When the magnetic materials adjacent to the TI are different, a V_{sia} needs to be applied to the top and bottom surfaces of the TI. As discussed also in Ref. 11, V_{sia} causes a phase transition from the axion states insulator to the metallic state as the bandgap closes with increasing the value of V_{sia} . We name the minimum band gap that is induced by the the Zeeman terms ($\min\{\Delta_t, \Delta_b\}$) as the magnetization gap. Based on Fig. 11, when V_{sia} ranges between 0 and $|\Delta_t - \Delta_b|/2$ (0-10 meV in the case considered in Fig. 11) the magnetization bandgap is 10 meV and for V_{sia} values between $||\Delta_t - \Delta_b|/2, ||\Delta_t + \Delta_b|/2$ (10-15 meV), the bandgap linearly decreases. For V_{sia} values above $||\Delta_t + \Delta_b|/2$ (15 meV), the bandgap tends to zero, giving rise to a metallic state. One has axion states as long as V_{sia} is between 0 and $||\Delta_t + \Delta_b|/2$. V_{sia} can only change the size of the magnetization gap, but since we consider E inside the band gap to calculate conductance, the Hall conductance is independent

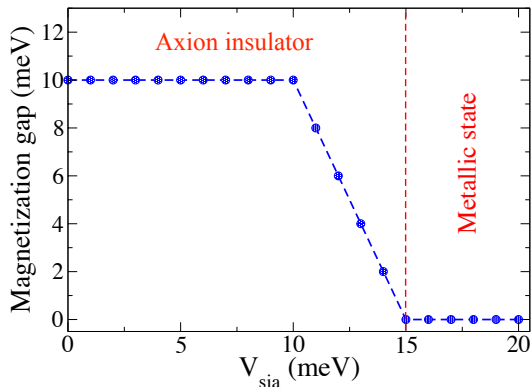


FIG. 11. Phase diagram as a function of V_{sia} . We took $\Delta_t = 5$ meV and $\Delta_b = -25$ meV, and found that V_{sia} between 0 and 15 meV yields axion insulators states, while for $V_{sia} > 15$ meV one finds a metallic state (cf. Ref. 11).

of V_{sia} .

For the same direction of magnetization at the top and the bottom surface we have $\sigma_{xy} = (C_t + C_b)\frac{e^2}{h} = \frac{e^2}{h}$, and if the magnetizations have opposite direction $\sigma_{xy} = 0$ ³⁹. The latter can characterize two different modes, i.e. either the axion insulator state or the normal insulator state²⁴. To determine whether $\sigma_{xy} = 0$ corresponds to the axion insulator or the normal insulator, the electrical transport needs to be calculated independently on the top and bottom surfaces using the Landauer-Büttiker formalism. For axion insulators, transport on the top and bottom surfaces is opposite and leads to net zero transport, while for normal insulators, transport is zero on both surfaces.

As stated in the introduction, in magnetoelectric materials, application of an electric field \mathbf{E} induces magnetization \mathbf{M} and the application of a magnetic field \mathbf{B} induces polarization \mathbf{P} . For 3D insulators, a classification based on TRS and magnetoelectric coupling θ can be introduced. When TRS is broken, the insulators are further subdivided into two categories. If TRS is broken but there is a symmetry as a sum of TRS and translational symmetry (dubbed effective TRS), one deals with an axion insulator and in this case $\theta = \pi$ ^{3,5}. As shown in Fig. 12, we calculated the axion term θ for the FM-TI-FM heterostructure as a function of the TI thickness (in units of quintuple layers). With increasing the number of QLs, $\theta \rightarrow \pi$, indicating formation of an axion insulator for $N_z > 10$ (for other parameters $\Delta_t = 5$ meV, $\Delta_b = -25$ meV, and $V_{sia} = 0$). A moderate electric field can be considered as an on-site energy varying linearly from the bottom to the top of the TI film, such that magnetization becomes linearly dependent on the electric field and the axion angle θ can be computed.

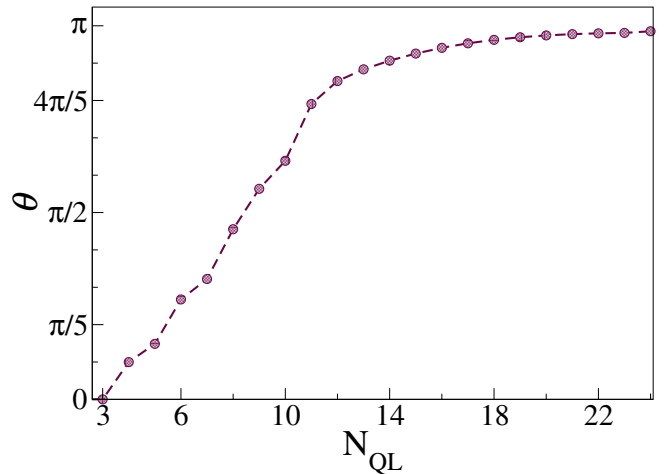


FIG. 12. The axion term θ for different TI thickness (number of QLs) in case of $\Delta_t = 5$ meV, $\Delta_b = -25$ meV, and $V_{sia} = 0$. With increasing thickness, $\theta \rightarrow \pi$, characterizing formation of an axion insulator state for thickness beyond 10 QLs.

V. CONCLUSIONS

In summary, we developed a real space tight-binding Hamiltonian for Bi_2Se_3 taking into account the symmetry of the system and fitting the parameters using the band structure obtained from DFT. This derived tight-binding model is the most important aspect of our study. We show that it is readily well-suited to capture experimental results such as the hybridization gap, but the effects of gating, magnetic field, and strain can also be relatively straightforwardly added to the consideration in our model based on the symmetry group of the system. Since this model is represented in real space, further studies of the effects of doping and magnetic and non-magnetic impurities are facilitated.

In this work, we went on to apply the developed model to a TI sandwiched between magnetic insulators, where the magnetic proximity effect was added through the Zeeman term to create favorable conditions for the appearance of the axion insulator states. In cases when different magnetic materials are considered, that induce exchange fields of opposite sign at their interfaces with the TI, a potential difference due to structural inversion asymmetry V_{sia} was taken into account between the top and bottom surfaces of the TI. In doing so, we demonstrated that a phase transition from the axion state to the metallic state occurs with increasing V_{sia} . To further characterize the axion insulator state, a four-terminal structure was considered to calculate longitudinal and Hall conductance using the Landauer-Büttiker formalism. The total Hall conductance of the axion insulator is expected to be zero, however, the same is expected from a trivial band insulator. Therefore, to distinguish between normal insulator and axion insulator we performed calculation of the Hall conductance for each surface separately, to identify the axion state. Furthermore, by calculating

the axion term θ for a heterostructure in which magnetization gap of opposite sign is induced at the top and bottom surfaces of the TI, we showed that by increasing thickness of the TI beyond a threshold of 11 quintuple layers one reaches $\theta \approx \pi$ - the characteristic feature of an axion insulator. These findings validate the simulation framework approach developed in this work, set the key limits on the stability of the axion insulator state in FM-TI-FM heterostructures, and open a platform for further *in silico* tailoring of the magnetoelectric response in TI

heterostructures by design.

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