Gap and spin texture engineering of Dirac topological states at the Cr–Bi$_2$Se$_3$ interface

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The presence of an exchange field in topological insulators reveals novel spin related phenomena derived from the combination of topology and magnetism. In the present work we show the controlled occurrence of either metallic or gapped topological Dirac states at the interface between ultrathin Cr films and the Bi$_2$Se$_3$ surface. The opening and closing of the gap at the Dirac point is caused by the spin reorientation transitions arising in the Cr films. We find that atom-thin layers of Cr adhered to Bi$_2$Se$_3$ surfaces present a magnetic ground state with ferromagnetic planes coupled antiferromagnetically. As the thickness of the Cr film increases stepwise from one to three atomic layers, the direction of the magnetization changes twice from out–of–plane to in–plane and to out–of–plane again. The out–of–plane magnetization drives the gap opening and the topological surface states acquire a circular meron spin structure. Therefore, the Cr spin reorientation leads to the metal-insulator transition in the Bi$_2$Se$_3$ surface and to the correlated modification of the surface state spin texture. Consequently, the thickness of the Cr film provides an effective and controllable mechanism to modify the metallic or gapped nature, as well as the spin texture of the topological Dirac states.

I. INTRODUCTION

The recent discovery of three dimensional (3D) topological insulators (TIs) has led to unique and fascinating physical phenomena, such as the quantum anomalous Hall (QAH) phase, and the topological magnetoelectric effect. The robustness of the surface metallicity under time reversal invariant perturbations and the realization of novel quantized states arising from their peculiar coupling to magnetic fields are distinct characteristics of this new phase of quantum matter. A central feature of TIs is the existence of helical surface states (SSs) with the electron spin locked to the crystal momentum. The presence in the TI of an exchange field, which violates time reversal symmetry (TRS), lifts the Kramers degeneracy and discloses novel spin related phenomena directly derived from the combination of topology and magnetism. The QAH effect has already been observed in three-dimensional magnetic TIs. Nevertheless, the experimental realization of a magnetoelectric topological insulator, which in fact corresponds to a non–integer quantum Hall effect at the surface, still remains a challenge.

To experimentally achieve these topological phases a surface gapped by a TRS breaking perturbation is required. There are three different ways to break TRS in TIs, either by conventional doping with magnetic elements, by proximity to a magnetic field at a TI-magnetic interface, or by an external magnetic field. The effect of magnetic doping with 3d transition metals in the Bi$_2$Se$_3$ family of compounds has been extensively investigated both theoretically and experimentally. It has been shown that the interaction with magnetic impurities modifies the electronic and magnetic ground state of the 3D TIs. However, the changes in the ground state are not universal since they critically depend on the specific magnetic atoms, occupation sites of the magnetic impurity, and experimental conditions. Cr-doped Bi$_2$Se$_3$ is a prototype magnetic TI, and several works have reported magnetically induced effects in this system. First-principles calculations found that substitutional Cr, which is energetically more favorable than interstitial Cr, preserves the insulating character in the bulk and that Cr-doped Bi$_2$Se$_3$ is likely to be ferromagnetic. However, evidence from the experimental observations is so far inconclusive. Both ferro- and antiferromagnetic Cr defects in high quality epitaxial thin films has been observed. Nevertheless, Cr doping of bulk or thin films of Bi$_2$Se$_3$ crystals seems to lead to a gap opening in the Dirac cone, evidencing time–reversal symmetry breaking. In contrast, surface deposition of Cr atoms on the surface of Bi$_2$Se$_3$ up to $\approx 10\%$ monoatomic layer (ML) coverages preserves the metallic surface. The absence of gap opening at the Dirac point indicates that for dilute Cr adatom concentrations there is no long–range out–of–plane ferromagnetic order. Despite these works, the interface between Cr films and the Bi$_2$Se$_3$ surface has not been investigated and thus the spin behavior of the topological SS under the interaction with ultrathin Cr films remains unknown.

In the present work we explore the spin configuration and topological state at the interface of Bi$_2$Se$_3$ surfaces and Cr films in the ultrathin limit, one to three MLs thick. We find that the presence of the Cr magnetic film triggers a double transition, from a Dirac-metal to a gapped system, on the topological SS of Bi$_2$Se$_3$ as a function of the Cr thickness. The gap opening at the Dirac point is induced by the proximity of the Cr film and thus the observed modulation of the gap is associated with the spin reorientation occurring in the magnetic layer. In fact, the magnetization direction in the Cr film evolves from out–of–plane to in–plane and once again to out–of–plane as the Cr thickness increases stepwise from one to two and three MLs. Correlated with the gap, there is
a modulation of the spin texture of the topological SSs, which undergoes a double circular skyrmion to circular meron transition.

II. MODEL AND METHODS

Density Functional Theory (DFT) spin–polarized calculations were carried out with the SIESTA code\cite{27} as implemented in the GREEN package\cite{28,29}, although specific structures were also calculated with the Vienna \textit{ab-initio} simulation package (VASP)\cite{30}. The generalized gradient approximation with Perdew-Burke-Ernzerhof\cite{31} type exchange-correlation functional was used in all cases. In the SIESTA calculations, the spin–orbit coupling is considered via the recently implemented fully-relativistic pseudopotential formalism\cite{28} while the semi-empirical pair-potential approach to van der Waals (vdW) forces of Ortmann \textit{et al.}\cite{32} was employed to correctly account for the weak inter quintuple layer (QL) interaction in the Bi$_2$Se$_3$ crystal. The numerical atomic orbitals basis set was generated according to the double \textit{ζ}-polarized scheme with confinement energies of 100 meV. For the computation of three-center integrals, a mesh cut-off as large as 1200 Ry was used, equivalent to a real space grid resolution below 0.05 Å$^3$. In the VASP calculations plane wave basis set with a kinetic energy cutoff of 340 eV was used. For the Brillouin zone (BZ) integrations a centered $13\times13\times1$ k-sampling was employed, while the electron temperature was set to $k_BT=10$ meV in both calculation schemes.

Bi$_2$Se$_3$ has a rhombohedral crystal structure with space group R$ar{3}$m (D$_3^3_d$). It can be described as a layered compound constituted by QLs along the [0001] direction. A QL contains alternating Se and Bi atomic layers, and within each QL the two Bi layers are equivalent, while the Se in the middle is inequivalent to the external Se. The stacking pattern is fcc-like, -AbCaB-CoCaA-, where capital and small letters stand for Se and Bi, respectively. The Se-Bi bonds within the QLs are mainly covalent, while at adjacent QLs the Se-Se double bonds are only weakly bonded through van der Waals forces. The in-plane lattice parameter is $a_{Bi_2Se_3}=4.14$ Å, while $c=9.54$ Å determines the periodicity along the [0001] direction.

Bulk Cr follows a bcc crystal structure with lattice parameter $a_{Cr}=2.91$ Å. Each atom has 8 nearest neighbors (n.n.s). Surfaces perpendicular to the [111] direction exhibit three-fold C$_3$ symmetry and an open structure, since only 6 out of the 8 n.n.s lie in the adjacent atomic layers, while the remaining 2 n.n.s are located three atomic layers above and below. Along this direction the stacking sequence follows an ...ABCABC... pattern, analogous to that of the Bi$_2$Se$_3$ crystal in the [0001] direction (see Figure 1). Cr is unique among the 3$d$ transition metals, showing an itinerant antiferromagnetic ground state. It exhibits a spin density (SDW) wave along the [100] direction –or, equivalently, along the [111] direction–, with a wave vector almost commensurate with the lattice, being its Néel temperature $T_N=311$ K. Contrary to what happens in bulk crystals, in which all three crystallographic directions are equivalent, in thin Cr films the SDW wave vector is perpendicular to the film surface and the SDW is commensurate with the lattice. Since the Cr–Bi$_2$Se$_3$ systems studied are formed of a maximum of 3 Cr layers, we can consider Cr as a pure antiferromagnet in the ultrathin film regime. Thus, the Cr slabs are expected to show atoms in the same atomic layer coupled ferromagnetically, being the interlayer coupling antiferromagnetic.

We model the Cr–Bi$_2$Se$_3$ interfaces by $1\times1\times1$ and $2\times1\times1$ supercells with the equilibrium in–plane lattice constant of bulk Bi$_2$Se$_3$. We take the [0001] Bi$_2$Se$_3$ direction as $z$ and the (111) plane as the $xy$ plane. Along the $z$ direction the supercells contain the Cr film on top of either 4 or 6 QLs (20 or 30 atomic planes) of Bi$_2$Se$_3$ and a vacuum layer larger than 20 Å to avoid interaction between opposite surfaces. During the structure optimization, the Cr overlayers and the interface most QL of Bi$_2$Se$_3$ were fully relaxed until the residual forces were smaller than 0.02 eV/Å, while the remaining atoms were fixed to the relaxed geometry of the corresponding Bi$_2$Se$_3$ thin film.

III. ATOMIC STRUCTURE AND INTERFACE CHARGE TRANSFER

We consider commensurate Cr films with 1, 2 and 3 ML thicknesses on top of (111) Bi$_2$Se$_3$ surfaces. The atomic structure of the (111) composed slab exhibits three-fold C$_3$ symmetry and three reflection planes perpendicular to the surface –see Fig. 2(a)–. The in–plane lattice parameters of Bi$_2$Se$_3$ (4.14 Å) and Cr (4.12 Å) show a small lattice mismatch of 0.5%. First, we examine different positions for the Cr overlayers, including \textit{fcc} and \textit{hcp} hollow sites, bridge and Se-top sites. As expected, the high
symmetry hollow sites are the energetically most favorable. Figures 2(a) to (d) show the calculated equilibrium structures. For 1 and 2 ML films the interfacial Cr atoms occupy the fcc hollow sites following the Bi$_2$Se$_3$ stacking, ...-BcAbC-A and ...-BcAbC-AB respectively, where bold letters correspond to Cr atoms. However, for the 3 ML film the interface Cr moves into the hcp hollow site on top of the Bi subsurface layer and there is a reversal of the stacking sequence ...-BcAbC-BAC. This spatial self-organization of the Cr film has to be due to the peculiar open structure of the (111) bcc surface in which first n.n.s are in the adjacent layers and in the third layers above and below. In this way, while for the 1 and 2 ML Cr films the interface Cr atoms are almost coplanar to the surface Se and lie on top of the Se in the center of the first QL, for the 3 ML film the Cr-Se interface bond distance increases notably and the Cr at the interface lies on top of the outermost Bi. The relaxed bond lengths are given in Table I. The Cr-Cr distances are close to the bond lengths in bulk Cr, 2.49 Å. Note the increase in the Cr1-Se1 bond length for the 3 ML film. Additionally, the bond distances for the non-equilibrium Cr trilayer in the fcc configuration –see Fig. 2(a)– are presented at the bottom of the Table. In this configuration, similar to the 1 and 2 ML cases, the interface Cr atoms remain almost coplanar to the Se surface at the expense of very large n.n.s Cr-Cr bond distances. The fcc configuration is about 80 meV more energetic than the equilibrium 3 ML Cr-Bi$_2$Se$_3$ structure, well above the energy involved in room temperature fluctuations.

The calculated binding energies are also given in the Table. The binding energy $E_{ads}$ is obtained as

$$E_{ads} = E_{Cr−Bi$_2$Se$_3$} - E_{Bi$_2$Se$_3$} - E_{Cr} \tag{1}$$

where $E_{Cr−Bi$_2$Se$_3$}$ is the total energy for the composed Cr–Bi$_2$Se$_3$ system, $E_{Bi$_2$Se$_3$}$ is the total energy of the isolated 4 QL Bi$_2$Se$_3$ system and $E_{Cr}$ is the total energy of the isolated Cr subsystem in the same ionic and magnetic configuration as it acquires in the composed Cr–Bi$_2$Se$_3$ system. We found a negative value for the adhesion energy for all the Cr films in correspondence with the exothermic character of dilute Cr adsorbed on Bi$_2$Se$_3$ surfaces for submonolayer coverages.

The different atomic configuration of the equilibrium structures is clearly reflected in the interface charge redistribution. We have calculated the Mulliken charges for the Cr–Bi$_2$Se$_3$ systems and for the corresponding isolated slabs, a pristine 4 QL Bi$_2$Se$_3$ slab, and the isolated Cr films of 1, 2 and 3 Cr MLs with the same atomic and magnetic configuration as they present when adhered to Bi$_2$Se$_3$. The differences between the Mulliken charges of the entire Cr–Bi$_2$Se$_3$ systems and those corresponding to the isolated subsystems are displayed in Figure 2. In all the cases the charge transfer is small and mostly confined to the Cr film and the first Bi$_2$Se$_3$ QL. For 1 and 2 Cr ML coverages, the Cr layers acquire charge at the expense of the Se atoms, both at the interface and in the middle of the first QL. In the 3 Cr ML system, on the contrary, the charge transfer is towards the Bi$_2$Se$_3$. The interfacial Cr donates charge, mainly to the n.n.s. Se, which gains electron charge, increasing its ionic radius and consequently increasing the interface bond length. This different behavior can be attributed to the different adsorption site of the first Cr layer (hcp hollow versus fcc hollow for 1 and 2 Cr MLs). Nevertheless, there is always a chemical interaction at the interface. In addition, the Bi$_2$Se$_3$ free surface presents a small charge gain in all the calculated structures.

### IV. MAGNETIC GROUND STATE

To model the magnetic ground state of Cr layers we consider different configurations having parallel and antiparallel collinear Cr magnetizations both between planes and within a plane. We employed an in–plane unit cell with 2 atoms per plane. We find a ferrimagnetic ground state with ferromagnetic Cr planes coupled antiferromagnetically for all the studied Cr film thicknesses.

Due to the C$_3$ symmetry of both Cr and Bi$_2$Se$_3$ layers, the 3$d$ Cr and 4$p$ Se orbitals hybridize, as can be clearly appreciated in the spin–resolved total DOS for three Cr layers adhered to Bi$_2$Se$_3$ shown in Fig. 5. The hybridization drives the Cr states close to the Fermi level, confined in an energy region $\approx 1.5$ eV below $E_F$. In addition, a large energy splitting of about 4 eV between the spin–majority and the spin–minority states is obtained, and the majority Cr states are fully occupied while the minority–spin channel is almost unoccupied. Therefore, the magnetic moments (MMs) of the Cr layers are close to the Hund rule value for isolated Cr atoms. The calculated MMs, shown in Table I, are remarkably large at the surface plane ($\geq 4 \mu_B$/atom) for all the systems, while they decrease for the subsurface Cr layers. For 1 and 2 ML Cr films there is an appreciable induced MM on the Se and Bi topmost planes of 0.2 $\mu_B$, aligned opposite to the Cr MM at the interface, while the induced MMs in the Bi$_2$Se$_3$ for the 3 ML Cr film is almost negligible in correspondence with the different chemical interaction at the interface. Note the larger MMs of the 3 ML Cr fcc structure due to larger interlayer distances.

| Cr3-Cr2 | Cr2-Cr1 | Cr1-Se1 | $E_{ads}$ |
|---------|---------|---------|-----------|
| 1 ML    | -       | -       | 2.39 -2.00|
| 2 ML    | -       | -       | 2.59 -1.98|
| 3 ML    | 2.64    | 2.48    | 2.84 -1.77|
| 3 ML-fc*| 3.11    | 3.42    | 2.39 -1.69|

TABLE I. Relaxed bond lengths in Å between the Cr layers –columns 2 and 3– and between the interface Cr and the interface Se –column 4–. The last row corresponds to the more energetic fcc configuration (see Fig. 2) for the 3 ML Cr system. The adhesion energy $E_{ads}$ is given in eV in the rightmost column. The fcc–like case for the 3 ML Cr is more than 80 meV less stable.
FIG. 2. (a) Top view of Bi$_2$Se$_3$ (111) surface. Dashed lines depict the mirror planes M$_1$, M$_2$ and M$_3$, and the M and K points of the Brillouin zone are also indicated. (b) to (d) show the relaxed geometries for 1 to 3 ML Cr coverages on a 4 QL Bi$_2$Se$_3$ slab, along with arrows indicating the magnetization of the Cr layers for the magnetic ground state in each case. (e) Band structure around the center of the Brillouin zone for a pristine 4 QL Bi$_2$Se$_3$ slab. (f) to (h) display the band dispersion diagrams for 1 to 3 Cr MLs on a 4 QL Bi$_2$Se$_3$ slab in the magnetic ground state configuration shown above. (i) to (k) Depict the band structure of 1 to 3 Cr MLs on a 4 QL Bi$_2$Se$_3$ slab with the Cr MMs perpendicular to that of the magnetic ground state for each system, i.e. along $x$ for (i) and (k) and along $z$ for (j). The projection of the states on the interfacemost QL of Bi$_2$Se$_3$ is shown in red, while the projection on the Cr subsystem is shown in cyan.

Since the spin–orbit coupling is included in the calculations we can determine the direction of the Cr MM relative to the crystal lattice. The preferential orientation of the Cr magnetization vector was obtained by comparing the total energies of in–plane ($M_x$, $M_y$) and out–of–plane ($M_z$) orientations of the total magnetization $\mathbf{M}$ (the z axis is defined normal to the surface). It is noteworthy to point out that in the ground state within the planes the Cr atoms are always coupled ferromagnetically, thus the Cr MMs are aligned within each layer—see
FIG. 3. (a) Side view of the 3 Cr ML on the Bi₂Se₃ (111) surface with the Cr layers following the stacking pattern of Bi₂Se₃, i.e. with the first Cr layer occupying the fcc hollow site. The arrows indicate the magnetic ground state for this ionic configuration. Note that the ionic configuration shown in Fig. 2 (d) is more stable for the Cr trilayer. The band structure of a 4 QL Bi₂Se₃ slab with a Cr trilayer in the ionic and magnetic configuration depicted in (a) is shown in (b).

The projection of the states on the interfacemost QL of Bi₂Se₃ is shown in red, while the projection on the Cr subsystem is shown in cyan.

FIG. 4. Mulliken charge rearrangement of the 1 (cyan), 2 (red) and 3 (orange) Cr ML systems on a 4 QL Bi₂Se₃ slab. The figure displays the atomic charge difference between the isolated Cr and Bi₂Se₃ subsystems and that acquired in the composed Cr–Bi₂Se₃ system. Only the Cr subsystem and the first and last QLs are shown since the charge rearrangement in the inner QLs of Bi₂Se₃ is negligible. The Cr–Se interface is indicated with a dashed line as a guide to the eye.

Table II. Magnetic moments of the Cr layers in Bohr magnetons for 1, 2 and 3 ML coverages. Cr1 (Cr3) corresponds to the interfacemost (farthest from the interface) Cr layer. 

|       | µCr1 | µCr2 | µCr3 | µTot |
|-------|------|------|------|------|
| 1 ML  | -    | -    | -4.3 | 4.0  |
| 2 ML  | -    | 4.2  | -3.1 | 1.4  |
| 3 ML  | -4.2 | 3.6  | -3.7 | -4.3 |
| 3 ML-fcc* | -4.9 | 4.7  | -4.0 | -3.8 |

The easy magnetization axis for the 1 ML Cr–Bi₂Se₃ system lies perpendicular to the surface (out–of–plane), while as the thickness of the Cr film increases a double spin reorientation transition takes place and the magnetization direction changes to in–plane for 2 ML and again to out–of–plane for the 3 ML Cr film. A similar spin reorientation transition has been reported in ultrathin Co films grown on hexagonal Ru (0001) [32]. The magnetic anisotropy for the 1 and 2 ML Cr–Bi₂Se₃ systems is unusually large, of ≈ 25 meV and 35 meV respectively, while for the 3 ML Cr–Bi₂Se₃ system we obtain a smaller value of 5 meV.

V. TOPOLOGICAL SURFACE STATES

We additionally analyze the electronic structure of the Cr–Bi₂Se₃ slabs. Figures 2 (e) to (h) show the corresponding band dispersions around the Γ point and that of the pristine Bi₂Se₃ 4 QL film. The band dispersion of the pristine film shows the topologically protected metallic surface states with the Fermi level located at the Dirac point. However, for all the Cr–Bi₂Se₃ slabs the position of the Fermi level is shifted up between 0.2 and 0.4 eV with respect to the Dirac cone of the free Bi₂Se₃ surface, which persists in the three cases. As a result the free surface topological SSs are always electron doped.

Next, we focus on the SS when a single Cr overlayer is adhered to the Bi₂Se₃ surface. A large Dirac gap opens up, and the gap opening only occurs at the interface with the magnetic film while the Dirac cone at the free Bi₂Se₃ surface remains, evidencing the spatially localized character of the effect. Furthermore, our calculations reveal that the magnetic easy axis is along the out–of–plane direction as shown in Fig. 2 (b). Therefore, the origin of
the gapped Dirac point is the exchange coupling between
the TI SS and the out–of–plane magnetization of the Cr
film, which breaks TRS.

As explained above, in the 2 ML system the Cr lay-
ers present an in–plane magnetization, and we do not
find any appreciable energy difference when the in–plane
magnetization is along or normal to the vertical reflection
planes of the Bi$_2$Se$_3$ thin films –see Figure 2 (a)–. Thus,
we discuss the results for the in–plane magnetization nor-
al to the reflection plane M$_1$. The corresponding band
dispersion around the Γ point is represented in Figure 2
(g). The topological surface state survives and there is
no shift in momentum space of the Dirac point, which
remains at Γ. However, the dispersion is no longer linear
and the SS presents a large anisotropic mass. Only along
the -K$_2$–Γ–K$_2$ line, perpendicular to the mirror plane,
electrons at $k$ and $-k$ have the same energy. The preser-
vation of the Dirac point can be easily understood consid-
ering that although the breaking of TRS occurs for any
non–zero magnetization, the slab is invariant under a re-
fection normal to the in–plane magnetization direction,
thus the reflection symmetry M$_1$ survives. This result is
a clear demonstration that in order to open a gap at the
Dirac cone, breaking the TRS and the three reflection
symmetries M$_{1,2,3}$ of the Bi$_2$Se$_3$ lattice is required. As
in the 1 ML system, the Dirac cone at the free surface of
Bi$_2$Se$_3$ remains unmodified but for an energy shift.

For the system consisting of 3 MLs of Cr on top of the
Bi$_2$Se$_3$ thin film, the magnetization points again along
the out–of–plane direction. Therefore, its behavior is
analogous to that of the 1 Cr ML slab: a gap opens at the
original Dirac point, although the gap is smaller. More-
ever, it is worth to note that for 3 Cr MLs, the Fermi level
lies exactly within the gap of the surface Dirac fermions
gapped by the exchange interaction.

For comparison, we have additionally included the dis-
ersion relations of the 1, 2 and 3 Cr–Bi$_2$Se$_3$ systems with
the magnetization of the Cr layers aligned perpendicular
to that of the corresponding magnetic ground states, i.e.
in–plane along $x$ for the 1 and 3 ML Cr and out–of–plane
along $z$ for the 2 ML Cr case –Fig. 2(i) to (k)–. Now, the
behavior of the topological SS is just the opposite, which
confirms the correlation between the opening of the gap
at the Dirac point and the presence of a perturbation that
breaks both TRS and the invariance of the system un-
der the three reflection symmetries of the Bi$_2$Se$_3$ lattice.
The crossing of the topological SS persists whenever the
magnetization is aligned in–plane and perpendicular to a
reflection plane, as in the 1 and 3 Cr ML systems –Fig. 2
(i) and (k)–. In both cases the reflection symmetry M$_1$ is
preserved. On the contrary, a gap opens for the out–of–
plane 2 ML Cr film, where TRS and the three reflection
symmetries M$_{1,2,3}$ are broken. The mass enhancement
and the induced anisotropy in the topological SS for the
1 and 3 Cr MLs are also clearly appreciable. Moreover,
the origin of the large calculated MAE is evident from
the sharp contrast between the band structures of these
excited states – Fig. 3 (i) to (k)– and their correspond-
ing magnetic ground states – Fig. 2 (f) to (h)–. Finally,
the band structure of the non-equilibrium 3 ML Cr film
with the fcc stacking is shown in Fig. 3(b). As expected,
there is a gap opening due to the out–of–plane magneti-
ization, analogous to that developed in the equilibrium 3
ML Cr–Bi$_2$Se$_3$ structure –see Fig. 3 (h)–.

These results prove that the gap opening of the topo-
logical surface states is exclusively due to the interplay
of the topology and the induced magnetization, and in-
dependent of the chemical behavior. As remarked above
the 1 and 2 ML Cr–Bi$_2$Se$_3$ systems show similar interface
chemical interactions –the charge transfer has the same
sign and similar value– and opposite to the interface in-
teraction in the 3 ML slab (see Figure 4). Nevertheless,
there is a gap in the 1 and 3 ML Cr–Bi$_2$Se$_3$ systems, while
in the 2 ML Cr–Bi$_2$Se$_3$ structure the topological SS at the
Γ point remains.

VI. SPIN TEXTURE OF THE SURFACE STATES

As shown above, the magnetization of the Cr layers at-
tached to the surface of the Bi$_2$Se$_3$ film provides a local
magnetic field, which modifies the degeneracy and topol-
ogy of the SS. Additionally, it induces a spin component
along the magnetization direction and alters the spin tex-
ture of the topological SS. We examine the spin texture
of the SSs in the equilibrium Cr–Bi$_2$Se$_3$ systems close to
Γ by calculating the expected value of the spin operator.
The results are displayed in Figure 6 which also includes
the spin distribution of the Dirac cone states of the pris-
tine Bi$_2$Se$_3$ surface. For the latter the spin is locked per-
pendicular to crystal momentum, showing the distinct
helical spin texture protected by TRS, and $S_z$ vanishes
close to the Dirac point. At large $k$ there is, however, a
finite small $S_z$ component due to the trigonal warping.$S_z$
remains null along the mirror lines Γ–M and reverses
its sign traversing from K to -K, in correspondence with
the trigonal symmetry of the system.

The spin texture of the gapped topological SSs (1 and
3 ML Cr systems) is in sharp contrast to that of the free
surface. In the vicinity of the gapped Dirac point, the
states show an imbalance between $S_z$ and -$S_z$ at a
given energy, and they present a significant net out–of–plane
spin polarization. Only the in–plane components reverse
sign changing from $k$ to -$k$. Furthermore, the upper and
lower Dirac bands have opposite $S_z$, evidencing that
the spin degeneracy is indeed lifted at the Γ point. For larger
$k$, away from Γ, the induced $S_z$ component gradually de-
creases, and the out–of–plane spin distribution results
from the competition between the magnetic order that
aligns the spin along the out–of–plane direction and the
spin texture imposed by the warping term which forces
adjacent K points to have opposite $S_z$. In the 2 ML
Cr slab, the in–plane magnetization exhibited by the Cr
layers in the interfacial plane does not induce observable
spin reorientations of the Dirac state, and its spin texture
FIG. 6. (a) to (d): Side view of the spin texture of the surface state for Cr overlayers of 0 (pristine Bi$_2$Se$_3$ surface) to 3 MLs. (e) to (h): Top view of the hole–like surface state for Cr coverages of 0 to 3 MLs. The expectation value for the spin is shown as an arrow at each $k$–point, while the $S_z$ component is additionally color coded according to the scale at the right, being the limits ±100 (30)% of the modulus of $S=\sqrt{S_x^2 + S_y^2 + S_z^2}$ for the 1 and 3 (0 and 2) Cr MLs. In the 2 Cr ML system –(c) and (g)– three elliptical black solid lines depict constant energy contours. The circular meron texture is patent in the 1 and 3 ML cases, while the spin texture of the 2 Cr MLs on Bi$_2$Se$_3$ is an anisotropic circular skyrmion.

is analogous to that of the free surface Dirac cone. However, due to the large anisotropy of the effective mass, the constant energy lines are no longer circular, but present an elliptical shape. Nevertheless, the SSs exhibit a well defined spin helicity and the total spin cancels in every constant energy contour. TRS breaking is evident from the spin texture of the three Cr–Bi$_2$Se$_3$ systems analyzed.

VII. CONCLUSIONS

In summary, we have found that the structural configuration of ultrathin Cr films attached to the (111) surface of Bi$_2$Se$_3$ is determinant to establish the topological behavior of Bi$_2$Se$_3$ SSs. Due to the coupling between Cr 3$d$ orbitals and the Bi$_2$Se$_3$ electrons, the Cr interface induces simultaneous charge and magnetic doping. However, the properties of the topological SS critically depend on the Cr film thickness and are independent of the specific chemical interaction at the Cr-Bi$_2$Se$_3$ interface. As the thickness of the Cr film increases stepwise from one to three MLs, the magnetization of the Cr layers undergoes two reorientation transitions, and changes from out–of–plane (1 ML) to in–plane (2 ML) and to out–of–plane (3 ML) once again. For the 1 ML and 3 ML Cr–Bi$_2$Se$_3$ interfaces the magnetic overlayer induces a gap at the Dirac point, producing massive fermions at the interface. Moreover, the gap already opens for a single Cr ML, and the value of the gap depends on the absolute value of the exchange interaction. In contrast, for the 2 ML Cr system the gapless Dirac cone is preserved. The complexity of the spin texture of gapped Dirac states signifies a competition between the in–plane helical component of the spin dictated by the spin–orbit coupling and the out–of–plane TRS breaking component induced by the proximity to the magnetic Cr. Our results evidence the importance of the actual structural configuration of the magnetic films and show that the thickness of the Cr film can be used to modify in a controlled way the metallic or gapped nature of topological Dirac states and their associated spin texture.

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