Interface induced states at the boundary between a 3D topological insulator Bi$_2$Se$_3$ and a ferromagnetic insulator EuS

S.V. Eremeev$^{a,b,e}$, V.N. Men’shov$^{c,b,e}$, V.V. Tugushev$^{c,d,b}$, E.V. Chulkov$^{e,f,b,*}$

$^a$Institute of Strength Physics and Materials Science, 634021 Tomsk, Russia
$^b$Tomsk State University, 634050 Tomsk, Russia
$^c$NRC Kurchatov Institute, 123182 Moscow, Russia
$^d$A.M. Prokhorov General Physics Institute, 119991 Moscow, Russia
$^e$Donostia Internation Physics Center (DIPC), 20018 San Sebastián/Donostia, Spain
$^f$Departmento de Física de Materiales UPV/EHU, CFM-MPC, and Centro Mixto CSIC-UPV/EHU, 20080 San Sebastián/Donostia, Spain

Abstract

By means of relativistic density functional theory (DFT) calculations we study electron band structure of the topological insulator (TI) Bi$_2$Se$_3$ thin films deposited on the ferromagnetic insulator (FMI) EuS substrate. In the Bi$_2$Se$_3$/EuS heterostructure, the gap opened in the spectrum of the topological state has a hybridization character and is shown to be controlled by the Bi$_2$Se$_3$ film thickness, while magnetic contribution to the gap is negligibly small. We also analyzed the effect of Eu doping on the magnetization of the Bi$_2$Se$_3$ film and demonstrated that the Eu impurity induces magnetic moments on neighboring Se and Bi atoms an order of magnitude larger than the substrate-induced moments. Recent magnetic and magneto-transport measurements in EuS/Bi$_2$Se$_3$ heterostructure are discussed.

Key words: Topological insulators; Magnetic proximity effect
PACS: 73.20.-r, 75.70.Tj, 85.75.-d

1 Introduction

Since the discovery of three dimensional TIs \cite{Hsieh2008}, magnetic proximity effect (MPE) at the TI/FMI interface has been considered as an effective method to break time-reversal symmetry of topological states by introducing an exchange field from FMI into TI. Such physical phenomena as quantum anomalous Hall effect and topological

* E-mail: waptctce@ehu.es
magneto-electrical effect critically rely on finding a way to open up a magnetic gap at the Dirac point of the topological states. In our recent works [2,3], where the thorough studies of MPE in TI containing heterostructures have been presented, it was argued that the physics of MPE is intricate enough and cannot be naively described in the terms of two-dimensional Dirac-like Hamiltonian with an added massive term. On the other hand, intriguing peculiarities in magnetic and magneto-transport properties of the EuS/Bi$_2$Se$_3$ heterostructures have been recently reported in Refs. [4,5]. It was shown the emergence of a ferromagnetic phase in TI with a significant saturation magnetization [4]; below the Curie temperature, unusual negative magnetoresistance was observed for the Bi$_2$Se$_3$ films thinner than $\sim 4$ quintuple layers (QL) deposited on EuS [5]. Motivated by these important experimental results, below we study the electron band structure of the EuS/Bi$_2$Se$_3$ interface within the DFT method. We focus on the spin polarization of interface states and the spin density redistribution between FMI (EuS) and TI (Bi$_2$Se$_3$) components of the heterostructure.

2 Computational details

For structural optimization and electronic band calculations we use the Vienna Ab Initio Simulation Package (VASP) [6] with generalized gradient approximation (GGA) to the exchange correlation potential. The interaction between the ion cores and valence electrons was described by the projector augmented-wave method [7]. Relativistic effects, including spin-orbit coupling, were fully taken into account. To describe the highly correlated Eu-4f electrons we include the correlation effects within the GGA+$U$ method [8]. The values of $U$ and $J$ parameters were taken to be of 7.4 and 1.1 eV, respectively [9].

To simulate the interface between FMI EuS and TI Bi$_2$Se$_3$ thin film, the lattice constant of the rocksalt EuS in the hexagonal (111) plane is fixed to that of Bi$_2$Se$_3$ which is $\approx 1.8$ % smaller than the experimental lattice parameter of EuS. The EuS(111) substrate was modeled by a slab consisting of 6 EuS bilayers. Eu-terminated side of this slab was chosen as interfacial plane while dangling bonds at the S-terminated side of the slab were passivated by hydrogen atoms. The atomic positions within the four near-interface atomic layers of EuS slab and of Bi$_2$Se$_3$ thin film were optimized. A vacuum spacer of $\sim 15\,\text{Å}$ was included to ensure negligible interaction between opposite sides of the structure. The optimized structure containing interface between EuS(111) slab and 3 QL-thick film of Bi$_2$Se$_3$ is shown in Fig. 1(a). Since selenium and sulfur are isoelectronic elements, the interfacial (first) QL forms a strong bonding with EuS substrate. The equilibrium Se-Eu bondlength at the interface is 3.043 Å that is only slightly larger than S-Eu bond in EuS bulk (2.980 Å).

3 Results

Three dimensional TI (e.g. Bi$_2$Se$_3$) promote the formation the topologically protected metallic states at the surface [1] while when TI is reduced to a film of a few QL thickness, the topological surface states became gapped [10][11][12]. The electronic spectrum of free-standing 3QL Bi$_2$Se$_3$ film shown in Fig. 1(b) demonstrates the gap of
Fig. 1. (Color online) (a) Bi$_2$Se$_3$(3QL)/EuS heterostructure. Dark red and yellow balls indicate Bi and Se atoms, respectively; Dark blue (Eu) and green (S) balls are atoms of the EuS substrate, light blue dots indicate passivation hydrogen layer on the free S-terminated surface of EuS. (b) Electronic spectrum of the free-standing 3QL-thick film of Bi$_2$Se$_3$. Gapped topological state is framed by orange dashed-line rectangle. (c) Charge density distribution along the axis $z$ for double-degenerate gapped state at the $\Gamma$ point; horizontal lines show position of atomic layers, shaded areas mark QLs.

36.4 meV at $\Gamma$. The gap results from spatial overlap of the states from the opposite surfaces of the thin film (Fig. 1(c)), and its size is determined by the film thickness.

The electronic structure of the 3QL Bi$_2$Se$_3$ film deposited on EuS slab is shown in Fig. 2. The spectrum demonstrates similarities with that for a Bi$_2$Se$_3$/MnSe heterostructure [2]. As in the latter case at the boundary between Bi$_2$Se$_3$ and EuS the topological and ordinary interfacial states arise [23]. It happens owing to modification of the electrostatic potential in the near-interface region caused by accumulation of $\sim$ 0.6 electron within the interfacial QL. The modified potential produces trapping of the state localized in the first QL and its shift to the lower energies. In contrast to the Bi$_2$Se$_3$/MnSe case, where the ordinary interfacial state has a gap of 56 meV, the occupied interfacial state in Bi$_2$Se$_3$/EuS, lying at energy of $-0.71$ eV at $\Gamma$, has a tiny $\Gamma$-gap of 7 meV. The small magnetic splitting is explained by the fact that the strongly localized Eu-f orbitals do not contribute to the interfacial state. This is responsible for negligible induced magnetization of the Bi$_2$Se$_3$ film provided via
MPE. So, the magnetic moments on the Se atoms, closest to the interface plane, and on the next Bi atoms are -0.023 and +0.012 \(\mu_B\), respectively, and they are an order of magnitude smaller on the deeper atoms of the first QL. Like in the Bi\(_2\)Se\(_3\)/MnSe case the occupied interfacial state has the maximal localization near the interface plane (Fig. 3a, darkblue line), decaying into the EuS and Bi\(_2\)Se\(_3\). As a function of momentum it extends through the bulk gap, crossing the Fermi level.

The gapped topological state become non-degenerate acquiring a Rashba-like spin splitting since the inversion symmetry in Bi\(_2\)Se\(_3\) film is broken due to the interface [10,11]. The spatial localization of the topological state mainly resided at the outermost QL (Fig. 3a, green line) is almost unchanged as compared to that in the free-standing Bi\(_2\)Se\(_3\) film (Fig. 1(c)). In contrast, spatial localization of the state resided on the opposite side of the Bi\(_2\)Se\(_3\) film, adjacent to EuS substrate, is strongly modified (Fig. 3a, lightblue line). This state is relocated away from the interface plane and its probability maximum lies in the second QL. The similar behavior was found for the topological Dirac state in Bi\(_2\)Se\(_3\)/MnSe [2]. The energy gap in topological state of the 3QL-thick film deposited on EuS substrate increases with respect to the free-standing Bi\(_2\)Se\(_3\) film, up to 64.6 meV, due to the relocation of the topological state away from the interface plane rather than by the MPE which is too small. This explanation is consistent with the fact that in the 2QL-thick Bi\(_2\)Se\(_3\) film with broken inversion symmetry the gap between Rashba-like spin-split topological states was found to be 97.3 meV [11].

In order to confirm that the gap in the topological state in Bi\(_2\)Se\(_3\)/EuS system is controlled by the thickness of the Bi\(_2\)Se\(_3\) film only we increased its thickness up to 5QLs. As one can see in Fig. 3b, the spatial distribution of the ordinary interfacial state (darkblue line) is not sensitive to the increase in the film thickness. The topological state localized at the inner side of the Bi\(_2\)Se\(_3\) film, as in the previous case, is relocated away from the interface plane (lightblue line). At the same time the inner-side- and outer-side-resided (green line) topological states become decoupled owing to decaying overlap with the increasing film thickness. This results in formation of the gapless topological Dirac cones at energies of \(-0.15\) and \(-0.08\) eV, localized at the inner and outer side of the Bi\(_2\)Se\(_3\) film, respectively (Fig. 4). Moreover, the group velocities in these states are almost the same and well comparable with that in a thick Bi\(_2\)Se\(_3\) slab.

The obtained band structure of the 5QL Bi\(_2\)Se\(_3\) film deposited on EuS differs from that of the Bi\(_2\)Se\(_3\)/MnSe heterostructure [2] where the Dirac state is gapped due to the interaction with spin-polarized ordinary interface state. As is stated above, at
Fig. 4. (Color online) Electronic structure of Bi$_2$Se$_3$(5QL)/EuS.

At the ideally sharp Bi$_2$Se$_3$/EuS interface, the MPE is insignificant. But what happens if the magnetic atoms are introduced into the TI film? To study the possible effects of the Eu atom doping on the magnetization of Bi$_2$Se$_3$ we use a free-standing 1QL block with $3 \times 3$ supercell where one of the nine atoms in the outer selenium layer is substituted by an Eu atom. The largest magnetic moments, 0.24 \( \mu_B \), are obtained at the second-layer Bi atoms, closest to the introduced magnetic impurity. The third-layer Se atoms, lying next to the magnetized Bi atoms as well as fourth-layer Bi atom, have smaller induced magnetic moments, 0.05 and 0.06 \( \mu_B \), respectively. The calculated net magnetization induced on the Bi and Se atoms of the supercell is found to be 1.20 \( \mu_B \).

4 Discussion and conclusion

Results of our theoretical study shed light on the interpretation of magnetic and magneto-transport experiments in Bi$_2$Se$_3$/EuS heterostructures [14,15]. Our calculations do not corroborate the idea of the authors of Refs. [14,15] about a crucial role of MPE in Bi$_2$Se$_3$/EuS heterostructures, leading to strong spin polarization of interface electron states in Bi$_2$Se$_3$ film and enhancement of indirect exchange coupling between Eu local moments in EuS layer via these states. On the contrary, the calculations reveal an utterly small magnitude of the induced spin polarization of carriers under ideally sharp boundary between EuS and Bi$_2$Se$_3$. It is quite reasonable to attribute the observed “interface ferromagnetism” [14] to the Eu atoms diffusion from the EuS into the first QL of Bi$_2$Se$_3$ during the heterostructure growth. According to our calculations, the spin magnetization produced by the Eu impurity atom is 1.20 \( \mu_B \) per $3 \times 3$ unit cell of Bi$_2$Se$_3$. Multiplying this quantity by the Landé $g$-factor of $\sim 50$ inherent in Bi$_2$Se$_3$ and presuming that 0-10 at.% of Eu atoms penetrate from the EuS substrate into the first QL of Bi$_2$Se$_3$, we can estimate the effective magnetization due to “interface ferromagnetism” as 7-13 \( \mu_B \) per Eu atom. That corresponds very well to the saturation magnetic moment reported in Ref. [14].

We demonstrated that a weakly gapped ordinary bound state caused by the interface potential arises in the immediate region adjacent to the Bi$_2$Se$_3$/EuS interface. The topological state also exists in the vicinity of the interface, however it is relocated from the interface plane to distant atomic layers of the TI film and the spectrum of this state is remarkably changed with the TI film thickness. Thereby there are two distinct conducting channels near the interface, ordinary and topological, which suggest original character of low-temperature transport properties of the Bi$_2$Se$_3$/EuS system, in particular, unusual
transition from the positive to negative magneto-resistance as the TI film becomes thinner than the critical thickness (\( \sim 4 \) nm [5]). Following our calculations, it can be shown that Eu atoms diffusion can play significant role in magneto-transport properties of the Bi\(_2\)Se\(_3\)/EuS heterostructure, providing the spin polarization of the ordinary bound state at the interface. At large thicknesses of the Bi\(_2\)Se\(_3\) layer, the weakly spin polarized high-mobility topological Dirac-like state relocated from the interface dominates the electron transport properties of the system and thus defines positive sign of magneto-resistance. At small thicknesses of the Bi\(_2\)Se\(_3\) layer, when the topological state becomes gapped due to hybridization effect, only the strongly spin polarized (due to above discussed “interface ferromagnetism” effect) ordinary bound state participates in the electron transport and provides the negative sign of magneto-resistance. The critical thickness of the TI layer observed in [5] testifies to the crossover between contributions of topological and ordinary bound states into the total magneto-resistance.

In summary, in this work we performed the DFT calculations to study interfacial states in the Bi\(_2\)Se\(_3\)/EuS heterostructures. We demonstrated that ordinary and topological bound states co-exist near the interface, thus defining a complex character of magnetic and magneto-transport properties of the system.

**Acknowledgements**

We acknowledge partial support from the Basque Country Government, Departamento de Educación, Universidades e Investigación (Grant No. IT-756-13), the Spanish Ministerio de Ciencia e Innovación (Grant No. FIS2010-19609-C02-01), the Ministry of Education and Science of Russian Federation (Grant No. 2.8575.2013), and the Russian Foundation of Basic Researches (Grant No.13-02-00016-a).

**References**

[1] M. Hasan, C. Kane, Reviews of Modern Physics 82, 3045 (2010).
[2] S.V. Eremeev, et al., Phys. Rev. B 88, 144430 (2013).
[3] V.N. Men’shov, et al, Phys. Rev. B 88, 224401 (2013).
[4] P. Wei, et al., Phys. Rev. Lett. 110, 186807 (2013).
[5] Q.I. Yang, et al., Phys. Rev. B 88, 081407(R) (2013).
[6] G. Kresse, J. Furthmüller, Comput. Mater. Sci. 6, 15 (1996).
[7] G. Kresse, D. Joubert, Phys. Rev. B 59, 1758 (1999).
[8] A.I. Liechtenstein, V.I. Anisimov, J. Zaanen, Phys. Rev. B 52, R5467 (1995).
[9] P. Larson, et al., Phys. Rev. B 75, 045114 (2007).
[10] Y. Zhang, et al., Nat. Phys. 6, 584 (2010).
[11] G. Landolt, et al., Phys. Rev. Lett. 112, 057601 (2014).
[12] I.V. Silkin, et al., JETP Lett. 94, 217 (2011).