Absence of magnetic-proximity effect at the interface of Bi$_2$Se$_3$ and (Bi,Sb)$_2$Te$_3$ with EuS

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We performed x-ray magnetic circular dichroism (XMCD) measurements on heterostructures comprising topological insulators (TIs) of the (Bi,Sb)$_2$(Se,Te)$_3$ family and the magnetic insulator EuS. XMCD measurements allow us to investigate element-selective magnetic proximity effects at the very TI/EuS interface. A systematic analysis reveals that there is neither significant induced magnetism within the TI nor an enhancement of the Eu magnetic moment at such interface. The induced magnetic moments in Bi, Sb, Te, and Se sites are lower than the estimated detection limit of the XMCD measurements of $\sim 10^{-3} \mu_B$/at.

The observation of the quantum anomalous Hall effect (QAHE) in magnetically-doped topological insulators (TI) has raised significant interest for applications in metrology. Test measurements demonstrating QAHE quantization accuracy at a record level of better than 1 part-per-million have recently been reported. However, the magnetic impurities are incorporated randomly in the TI lattice and the resulting disorder presumably reduces the temperature and currents at which the QAHE fully develops. It also leads to the observation of superparamagnetic phases and multidomain magnetization switching, which coincide with the QAHE and are not well understood. In this context, imprinting magnetism by proximity effects in thin film heterostructures appears as a promising alternative that could ensure the preservation of the TI crystalline quality and its bulk insulating properties. Magnetic proximity effects in heterostructures comprising TIs and magnetic insulators (MI) have been intensively investigated using optical methods, electronic transport measurements, polarized neutron reflectivity, and muon spin spectroscopy. Studied heterostructures include Bi$_2$Se$_3$/EuS, (Bi,Sb)$_2$Te$_3$/EuS, Y$_3$Fe$_5$O$_{12}$/(Bi$_x$Sb$_{1-x}$)$_2$Te$_3$, Tm$_3$Fe$_5$O$_{12}$/(Bi$_x$Sb$_{1-x}$)$_2$Te$_3$, Fe$_3$O$_4$/Bi$_2$Te$_3$, Cr$_2$Ge$_2$Te$_5$/Bi$_2$Te$_3$, and (Zn,Cr)Te/(Bi,Sb)$_2$Te$_3$/(Zn,Cr)Te, with the QAHE only reported in the latter. However, the most intriguing results have been arguably observed in Bi$_2$Se$_3$/EuS, where experiments using spin-polarized neutron reflectivity suggest an enhanced magnetic signal within Bi$_2$Se$_3$. They also indicate the persistence of large interfacial ferromagnetism that extends 2 nm into the TI interface up to room temperature, even though isolated EuS orders ferromagnetically only below 17 K. Such results are remarkable and would make Bi$_2$Se$_3$/EuS a leading candidate for the observation of topological magneto-electric phenomena and QAHE at high temperatures.

However, while the number of articles reporting signatures of ferromagnetic behavior in TI/EuS interfaces keeps on growing, topological phenomena have yet to be observed. QAHE has been systematically demonstrated in Cr- and V-doped (Bi,Sb)$_2$Te$_3$ despite that the ferromagnetic state appears to be much weaker than in Bi$_2$Se$_3$/EuS. In addition, first-principles calculations of the electronic band structure and magnetic ordering of the Bi$_2$Se$_3$/EuS interface did not find an induced magnetic moment in the TI or even a significant enhancement of the local magnetic moment of Eu. The discrepancy between experiments and theoretical analysis in combination with the absence of direct experimental proof of locally induced magnetism in the TI underscores the need of further experimental investigations.

Electronic and magnetic information regarding microscopic interactions can be obtained using x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD). These techniques have been successfully implemented to study magnetically-doped TIs and—most importantly—are able to detect elementspecific magnetic moments that are induced in the host TI lattice. In this Letter, we report XAS and XMCD measurements on TI/EuS interfaces, with TIs of the (Bi,Sb)$_2$(Se,Te)$_3$ family. In contrast to previous investigations, our systematic study provides fundamental local and element-selective information of the magnetic moments associated to Eu, Bi, Sb, Se, and Te atoms. In agreement with theoretical reports, we find no indication of proximity-induced magnetism in the TI. This suggests that the magnetic signatures in TI/MI heterostructures which have been reported do not originate from induced ferromagnetic order in the TI atoms at the TI/MI interface.
The TI/EuS heterostructures were grown on single-crystal BaF$_2$(111) substrates by molecular beam epitaxy under ultrahigh-vacuum (UHV) with a base pressure ~ $1 \times 10^{-10}$ Torr. Bi$_2$Se$_3$, Bi$_2$Te$_3$, Sb$_2$Te$_3$, and (Bi,Sb)$_2$Te$_3$ thin films with a thickness of 10 nm were grown by co-evaporation of elemental Bi, Sb, Se, and Te (6N purity), following the procedure in Ref. [28]. After growing the TI layer, the samples were transferred in UHV to a second chamber where a layer of 5-nm thick EuS was deposited at room temperature using an electron-beam evaporator, as in Ref. [7]. A protective 2-nm thick Al capping layer was subsequently deposited in the same chamber and allowed to oxidize in air. Additional Sb$_2$Te$_3$/EuS, Sb$_2$Te$_3$/Bi$_2$Se$_3$/EuS, and Bi$_2$Se$_3$/EuSi(x) with x = 1 and 5 nm samples were capped with a 20-nm thick Se layer grown in-situ, which was desorbed in the UHV preparation chamber for XAS measurements by heating the sample to ~180$^\circ$C. The structural and crystal quality of the films was assessed using in-situ reflection high energy electron diffraction and ex-situ x-ray diffraction and x-ray photoelectron spectroscopy (XPS) revealing epitaxial growth and sharp interfaces [29]. The macroscopic magnetic properties were studied by SQUID magnetometry [29].

XAS was performed using a 6-T magnet on beamline BL-29 (BOREAS) at the ALBA synchrotron (Spain), which provides a UHV sample environment with a base temperature of ~3 K. Measurements were focused on the M$_{4,5}$ edges of Eu (1110-1170 eV), Te (570-590 eV), Bi (2560-2700 eV), and Sb (525-545 eV), and L$_{2,3}$ edges of Se (1420-1460 eV). The XMCD signal was obtained by subtracting XAS spectra with the photon helicity vector antiparallel and parallel to the applied magnetic field. Measurements used total-electron-yield (TEY) detection, where the drain current was taken from the sample to the ground. The magnetic field was applied along the x-ray beam for two different geometries: normal incidence (perpendicular to the sample plane) and grazing incidence (30$^\circ$ off the sample plane).

Figure 1 depicts XAS and XMCD spectra at the Eu M$_{4,5}$ edges for Bi$_2$Se$_3$/EuS, Bi$_2$Te$_3$/EuS, (Bi,Sb)$_2$Te$_3$/EuS, Sb$_2$Te$_3$/EuS, Bi$_2$Se$_3$/EuSi(x) samples and comparison with the spectrum calculated for Eu$^{2+}$ 4f$^7$, as well as (b) EuS and (c) Eu$_2$O$_3$ references. Reference spectra have been vertically shifted for clarity. (d) XMCD spectra for all samples and references. Dashed vertical lines mark the main features for Eu$^{2+}$ (peaks B and D) and Eu$^{3+}$ (peaks A, C, and E) around the M$_5$ edge. Measurements were performed at 3 K under 2 T magnetic field applied in grazing incidence.

FIG. 1. (a) Averaged XAS spectra at the Eu M$_{4,5}$ edges for Bi$_2$Se$_3$/EuS, Bi$_2$Te$_3$/EuS, (Bi,Sb)$_2$Te$_3$/EuS, Sb$_2$Te$_3$/EuS, Bi$_2$Se$_3$/EuSi(x) and Sb$_2$Te$_3$/EuSi/Se samples and comparison with the spectrum calculated for Eu$^{2+}$ 4f$^7$, as well as (b) EuS and (c) Eu$_2$O$_3$ references. Reference spectra have been vertically shifted for clarity. (d) XMCD spectra for all samples and references. Dashed vertical lines mark the main features for Eu$^{2+}$ (peaks B and D) and Eu$^{3+}$ (peaks A, C, and E) around the M$_5$ edge. Measurements were performed at 3 K under 2 T magnetic field applied in grazing incidence.

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TABLE I. Effective magnetic moments for the Eu$^{2+}$ $4f^7$ state in TI/EuS samples as derived from the Eu $M_5$ XMCD asymmetry. Measurements were recorded at 3 K, except for those marked with *, which were recorded at ~10 K. The local spin magnetic moment of Eu in Bi$_2$Se$_3$/EuS for the atomic ground state and as calculated by DFT [18] are included for comparison.

| Sample                         | $\mu_{\text{eff}}$(\(\mu_B\)/atom) |
|--------------------------------|--------------------------------------|
| EuS                           | 5.8 ± 0.3                            |
| Sb$_2$Te$_3$/EuS/Se            | 5.4 ± 0.3                            |
| Bi$_2$Te$_3$/EuS               | 6.1 ± 0.4                            |
| (Bi,Sb)$_2$Te$_3$/EuS          | 5.9 ± 0.3                            |
| Bi$_2$Se$_3$/EuS               | 6.1 ± 0.4                            |
| Sb$_2$Te$_3$/Bi$_2$Se$_3$/EuS/Se| 4.4 ± 0.4                            |
| Bi$_2$Se$_3$/EuS (1 nm)/Se*    | 4.8 ± 0.4                            |
| Bi$_2$Se$_3$/EuS (5 nm)/Se*    | 5.3 ± 0.4                            |
| Atomic Hund’s rule ground state| 7.93                                 |
| DFT calculation [18]           | 6.94                                 |

left- circularly polarized XAS spectra at the Eu $M_5$ edge.

Table I lists the magnetic moments derived from $\mu$ for each sample at 3 K (~10 K for Bi$_2$Se$_3$/EuS (x) with $x = 1$ and 5 nm samples). The calculated XMCD spectrum in Fig. 1(d) yields the theoretical asymmetry $\Delta_{\text{theory}} = -0.497$, which corresponds to the Hund’s rule ground state (GS) value for Eu$^{2+} \ 4f^7$ ($S = 7/2, L = 0, J = 7/2$), with an effective magnetic moment $\mu_{\text{eff}}^{\text{GS}} = 7.93 \ \mu_B$. Because $L = 0$, this is a pure spin moment. Table I shows a reduction of $\mu_{\text{eff}}$ of about 30-35% for all samples with respect to $\mu_{\text{eff}}^{\text{GS}}$, which could be due to the presence of a small crystal field acting on the Eu 4f electrons, as observed for other rare earths [33]. These values are well in agreement with the magnetic moments of Eu in Bi$_2$Se$_3$/EuS calculated by density functional theory [18], also included in Table I. No significant enhancement in the Eu 4f magnetic moments of our TI/EuS samples is found compared to that of the isolated EuS layer. This observation is consistent with theoretical reports [17,18], but contrary to experimental findings using polarized neutron reflectometry [7].

Magnetic hysteresis loops were recorded by fixing the energy at the maximum of the Eu $M_5$ XMCD signal (1125.4 eV) while sweeping the field. Figure 2 shows loops measured for Bi$_2$Te$_3$/EuS, Sb$_2$Te$_3$/EuS/Se and Bi$_2$Se$_3$/EuS in grazing incidence. Values of $\Delta$ at saturation are very similar in all cases. This is consistent with Eu atoms having the same electronic and magnetic state regardless of the TI they are in contact with. The square shape of these hysteresis loops demonstrates that Eu 4f states are ferromagnetic at low temperatures (3 K) but they become paramagnetic at 20 K. This is in agreement with results of SQUID magnetometry, which indicate ferromagnetic ordering below $T_C \approx 15$ K [29].

Therefore, we do not observe signs of room-temperature ferromagnetic behavior in these TI/EuS systems, in contrast to previous reports [7].

The magnetic anisotropy of the Eu layer in the TI/EuS system was analyzed by comparing the XMCD spectra recorded in normal and grazing geometries at 2 T and at remanence (see Fig. S5 in Supplemental Material [29]). At 2 T, curves for normal and grazing geometries overlap for all TI/EuS samples. Without magnetic field, we observe remanence only in samples with the TI underlayer; the isolated EuS layer shows no XMCD signal at zero field. This behavior can be ascribed to the large spin-orbit interaction that is intrinsic to the TI [18]. Moreover, there is a significantly larger remanence in the grazing geometry for all TI/EuS samples, which is indicative of a favorable magnetization axis in the plane of the sample. In-plane anisotropy is also observed macroscopically by SQUID magnetometry (see Fig. S4 in Supplemental Material [29]). Previous experiments on Bi$_2$Se$_3$/EuS demonstrated perpendicular anisotropy in EuS [7]. Theoretical calculations have shown that topological surface states and their large intrinsic spin-orbit coupling may explain this observation [18]. However, the orientation of the magnetization depends strongly on the strain at the interface. When the EuS lattice is relaxed, as is the case for our TI/EuS films (see Fig. S1 in Supplemental Material [29]), it is expected that the magnetization remains in plane [18].

Next we will focus on the magnetic proximity effects. XMCD measurements at accessible absorption edges of Bi, Te, Sb, and Se elements were performed in search of induced magnetism on these non-magnetic atoms. Figure 3 depicts XAS and XMCD spectra at the Bi $M_{4,5}$ and Se $L_3$ edges for Bi$_2$Se$_3$/EuS and at the Sb and Te $M_5$ edges for Sb$_2$Te$_3$/EuS. No XMCD signal is apparent in any of these absorption edges probed, remaining within the noise level.

TEY-detected XAS for soft x rays is a surface sensitive technique with the majority of electrons originating from
at the Bi polarized x-rays. XAS (top panel) and XMCD (bottom panel)

FIG. 3. Experimental spectra for left- and right-circularly
absorption edge.

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the film with a contribution that decreases exponentially
which is in contact with EuS, but also from the bulk of
nal not only arises from the uppermost region of the TI,
at which the XMCD features are expected to appear.

Measurements were performed at 3 K under a magnetic field
of 2 T applied along the normal. Dashed lines mark the energy
included in (b) for comparison (vertically shifted for clarity).

We found values of the spin moment, $S_z \sim 10^{-3} \mu_B$ per
Bi and Se atom in the Bi$_2$Se$_3$/EuS samples and $\sim 10^{-3} \mu_B$
per Sb and $\sim 10^{-4} \mu_B$ per Te in the Sb$_2$Te$_3$/EuS/Se
sample.

The negligible induced magnetic moment on Bi, Sb, Te,
and Se atoms of (Bi,Sb)$_2$(Se,Te)$_3$ in contact with EuS
is a surprising finding. Recent observations suggest
the presence of magnetism within the TI layer of
Bi$_2$Se$_3$/EuS by magnetotransport measurements
polarized neutron reflectometry [7,11], magnetic second-
harmonic generation [10] and low-energy muon spin rotation [12]. Katmis et al. [7] reported values of magnetization
in the TI for a Bi$_2$Se$_3$(20 QL)/EuS(5 nm) sample bet-
between 250 and 300 emu/cm$^3$, which correspond to a mag-
etic moment between 4.4 and 5.3 $\mu_B$ per QL in a unit
cell of Bi$_2$Se$_3$. Assuming that Bi and Se atoms become
equally magnetic, and that magnetism develops over a
unit cell closest to EuS, the magnetic moment amounts
to $\sim$0.88-1.06 $\mu_B$ per (Bi or Se) atom, which is about 2
to 3 orders of magnitude larger than those estimated for
our Bi$_2$Se$_3$/EuS samples from XMCD measurements.

XMCD measurements have already been used to rule
out reported proximity effects using other (non-element
selective) experimental techniques. For example, the
anisotropic magnetoresistance at Y$_3$Fe$_5$O$_{12}$/Pt and fer-
rite/Pt interfaces was attributed to Pt atoms polarized
by proximity effects. However, XMCD measurements at
the Pt edges have shown no evidence of induced mag-
etism [10,12]. Theoretical studies using first-principles
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for Bi$_2$Se$_3$/EuS with ideally sharp interfaces [17] [18,43],
which is consistent with our XMCD results. Instead, Eu
doping on Bi$_2$Se$_3$ could result in local magnetic moments
that are much larger than those induced by the adjacent
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unit cell would account for an observed magnetic mo-
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EuS layer has been theoretically predicted to be energet-
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within the TI and, perhaps, of proximity effects, by sup-
pressing the formation of trivial states [44]. However,
our data does not show evidence of such an intercalation.
Other interfacial effects, including charge transfer

within 3-5 nm [34,36]. This implies that the XAS signal
not only arises from the uppermost region of the TI,
which is in contact with EuS, but also from the bulk of
the film with a contribution that decreases exponentially
with depth. In order to verify the absence of proximity-
induced magnetism, we performed XMCD measurements
at the Se $L_3$ and Bi $M_{4,5}$ edges in a Sb$_2$Te$_3$/Bi$_2$Se$_3$/EuS
heterostructure. The Bi$_2$Se$_3$ layer is 1 nm thick, ensur-
ing that the recorded XAS signal originates from the very
Bi$_2$Se$_3$/EuS interface. XMCD at the Se $L_3$ for this
sample is plotted in Fig. 3(b) (the Bi signal on this sample,
not shown, is very weak). In agreement with the results
on the Bi$_2$Se$_3$/EuS, no XMCD signal is found at this
absorption edge.

Non-negligible Te and Sb XMCD signals have been re-
ported for thin films of Cr-doped Sb$_2$Te$_3$ [26,37] and
bulk Cr-, V-, and Mn-doped Sb$_2$Te$_3$ [26,37] and Cr-
doped (Bi,Sb)$_2$Te$_3$ [25,38]. In all these cases, XMCD
data was recorded in TEY for in-situ cleaved samples.
TEY detected XAS and XMCD signal for our TI/EuS
heterostructures have poor statistics compared to those
reported for in-situ cleaved samples due to the 5 nm EuS
and 2 nm Al on top of the TI. Despite the noise level of
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per Sb and $\sim 10^{-4} \mu_B$ per Te in the Sb$_2$Te$_3$/EuS/Se
sample.
at the TI/MI interface and band bending could explain the absence of induced magnetism in the TI and the lack of observation of the QAHE [44–48]. Another possibility for the absence of proximity effects would be the presence of a magnetic dead layer at the TI/EuS interface. However, XAS and XMCD measurements of ultrathin EuS layers (≤ 1 nm) grown onto Bi₂Se₃ reveal that EuS remains magnetic, which allowed us to exclude such scenario in our TI/EuS samples [29]. This conclusion is further supported by the increase of the in-plane magnetic remanence for the TI/EuS structures, compared to the isolated EuS layer, which can be ascribed to the large intrinsic spin-orbit interaction of the TI at a sharp TI/EuS interface [18].

In summary, we have performed element-selective magnetometry to study proximity effects at the interface of TI/EuS bilayers. A variety of TIs of the (Bi,Sb)₂(Se,Te)₃ family were systematically investigated. The easy-magnetization axis of EuS remains in the plane of the sample and the magnetic remanence increases as a result of its contact with the TI. However, we found no evidence for an enhancement of Eu magnetic moments in the EuS layer, or an increase in the ferromagnetic ordering temperature. The magnetic signals on Bi, Sb, Te, and Se atoms of the TIs in contact with EuS were found to be negligible (below the detection limit of the XMCD technique) at temperatures and conditions where the EuS is magnetic and could polarize these non-magnetic atoms. Overall our XMCD measurements suggest that the observations in TI/EuS interfaces by magnetotransport and optical techniques are not due to proximity-induced magnetism but to a different mechanism, such as magnetic doping by Eu diffusion into the TI.

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