Large Anomalous Hall Effect in Topological Insulators
Proximitized by Collinear Antiferromagnets

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CrSb is an attractive material for room-temperature antiferromagnetic spintronic applications because of its high Néel temperature ~700 K and semi-metallic character. We study the magnetic properties of CrSb bilayers on few-layer topological insulator thin films using ab initio density functional theory. We find that the intrinsic parts of the total anomalous Hall conductivities of the thin films are non-zero, and approximately quantized. The Néel temperature of CrSb bilayers on few-layer topological insulator thin films is found to be approximately two times larger than that of an isolated CrSb thin film. Due to the low Fermi level density of states of CrSb, Hall quantization might be achievable by introducing disorder. CrSb bilayers on topological insulator surfaces are therefore attractive candidates for high-temperature quantum anomalous Hall effects.

Introduction— The intrinsic contribution to the anomalous Hall effect (AHE) is related to the geometrical phase of Bloch electrons in momentum space [1–2]. When a quasi-two-dimensional anomalous Hall system becomes insulating, its Hall conductivity is quantized at $e^2/h$ times a topological invariant of the occupied Bloch bands, the 1st Chern number. The quantum anomalous Hall effect (QAHE) was first predicted in 1988 [3], but has been realized experimentally only recently, first in topological insulators (TI) thin films doped with transition metal ions [4–7], then in films of the intrinsic magnetic topological insulator MnBi$_2$Te$_4$ [8–11], and then recently in magic-angle twisted bilayer graphene [13–16]. These established QAHE materials exhibit large Hall effects only at relatively low temperatures (< 10 K), and are therefore not suitable for the potential applications imagined in low-dissipation spintronics [17]. High-temperature QAHEs have been proposed in thin films formed by a TI proximally-coupled to a ferromagnetic insulator with a high Curie temperature ($T_C$), such as EuS, yttrium iron garnet, Cr$_2$Ge$_2$Te$_6$ or Tm$_3$Fe$_5$O$_{12}$. Although these films have been realized recently [18–22] and have high temperature magnetic order with $T_C \approx 300$ K and > 400 K respectively, the QAHE has not been observed.

Proximity-induced surface magnetism and anomalous Hall effects can also be achieved by establishing atomically sharp interfaces between a TI and an A-type (consisting of ferromagnetic layers that alternate in orientation) antiferromagnet (AFM) [23–24]. The AFM can locally magnetize the surface of the TI through short-range interfacial exchange coupling. In Refs. 23 and 24 CrSb was used as the AFM, motivated by the material’s high bulk Néel temperature ($T_N$) of around 700 K [25–27]. In this paper we study the magnetic properties and the intrinsic anomalous Hall conductivity (AHC) of CrSb/TI heterojunctions using ab initio density functional theory (DFT). We show that the AHC of a single bilayer (defined below) of CrSb is not only non-zero, as expected because the bilayer does not possess the bulk’s invariance under a combination of time-reversal and translation, but also large - around 0.64 $e^2/h$.

The AHE is traditionally associated with ferromagnetism. Its presence at a AFM/TI heterojunction is reminiscent of recent discoveries of AHE’s in some bulk antiferromagnets, including noncollinear antiferromagnets [28, 29], and also collinear antiferromagnets [30–32] that do not possess time-reversal-like symmetries forbidding the AHE. We find [33] that the AHC increases to 1.2 $e^2/h$ when bilayer CrSb is placed on a 3 quintuple layer (QL) Bi$_2$Te$_3$ TI film, and further to 4 $e^2/h$ if 5 QL of Bi$_2$Te$_3$ are used. Moreover, our DFT results suggest that exchange coupling in bilayer CrSb on TI thin films is twice as strong as in isolated bilayers. Exchange enhancement has also been found experimentally in some ferromagnetic-insulator/TI heterojunctions [18], and can potentially lead to high Néel temperatures. For CrSb bilayers on a TI we estimate using mean-field theory that the Néel temperature is about 1200 K, nearly twice the $T_N$ of bulk CrSb. Since bulk CrSb is a bad metal [34] with a very small Fermi surface, it might be experimentally feasible to open a mobility gap in intentionally disordered devices, and in this way to achieve perfect Hall quantization at high temperatures.

Magnetic properties— Bulk CrSb is an A-type AFM with local moments on the Cr ions and a nickel-arsenide (NiAs) structure [26]. The Cr atoms form triangular lattices with AA stacking along the c axis (see supplemental material for details [35]). When magnetically ordered, each (0001) layer of CrSb contains two Cr atomic layers, and is therefore referred to here as a single bilayer. In our DFT calculations, we use the experimental lattice constants, $a = 4.121\text{Å}$ [27] for the nearest-neighbor Cr-Cr distance in a (0001) plane and $c = 5.47\text{Å}$, which is twice the distance between Cr layers. A single CrSb bilayer is terminated by Cr and Sb atoms respectively at its two surfaces [35]. The two Cr atoms in each 2D unit cell are therefore distinguishable, which implies that the total
magnetization will be non-zero in spite of overall AFM order in bulk. (We expect that this 2D ferrimagnetism will persist for thin CrSb films with only a few bilayers.) As shown in Table I, the net magnetization of an isolated CrSb bilayer is about 0.7 $\mu_B$ per 2D unit cell according to the DFT calculations. The moments decrease in magnitude in both layers when the CrSb bilayer is placed on top of Bi$_2$Te$_3$, but the difference between layers is maintained, and converges as the TI thickness is varied. For comparison we also show results in Table I for the ferromagnetic configuration of the moments, which has a higher energy, and for the case of bulk CrSb in which the two Cr atoms in each unit cell are indistinguishable and have the same local moment magnitude. In all cases the local moment magnitude is $\sim$3–4 $\mu_B$.

In spite of the net magnetization, we find that in CrSb bilayer the local moments prefer an out-of-plane orientation as in bulk CrSb [26]. As shown in Fig. 1 the magnetic anisotropy energy for a CrSb bilayer without a TI substrate, and in a CrSb bilayer on multiple quintuple layers of Bi$_2$Te$_3$, $N$ denotes the number of TI quintuple layers.

![FIG. 1. Magnetic anisotropic energy for a CrSb bilayer on Bi$_2$Te$_3$ substrates with different thicknesses. $E_{in}$ is the ground state energy per Cr atom when the Cr moments are in-plane, while $E_{out}$ is the energy when the Cr moments are out-of-plane. As shown in the supplemental material [35], the easy directions are always either purely in-plane or purely out-of-plane. The green dotted line labels the boundary between in-plane and out-of-plane anisotropy. Above the line the easy direction is out-of-plane, while below the line the easy direction is in-plane. The red and blue curves are for the case of a CrSb bilayer and the case of a Cr$_2$Sb$_2$ layer, in which one Sb atom is removed from the bilayer.](image)

**TABLE I.** Local magnetic moments (in $\mu_B$) of Cr atoms in bulk CrSb, in a CrSb bilayer without a TI substrate, and in a CrSb bilayer on multiple quintuple layers of Bi$_2$Te$_3$. $N$ denotes the number of TI quintuple layers.

| CrSb/(TI)$_N$ | Bulk | $N = 0$ | $N = 3$ | $N = 4$ | $N = 5$ | $N = 6$ |
|---------------|------|--------|--------|--------|--------|--------|
| Cr$_1$(AF)    | 2.977| 3.954  | 3.424  | 3.444  | 3.445  | 3.441  |
| Cr$_2$(AF)    | -2.977| -3.282| -2.960| -2.938| -2.937| -2.935|
| Total         | 0    | 0.672  | 0.464  | 0.506  | 0.506  | 0.506  |
| Cr$_1$(FM)    | 2.956| 4.049  | 3.392  | 3.424  | 3.400  | 3.417  |
| Cr$_2$(FM)    | 2.956| 3.222  | 2.683  | 2.666  | 2.673  | 2.654  |
| Difference    | 0    | 0.827  | 0.709  | 0.758  | 0.727  | 0.763  |

In addition to the magnetic anisotropy energy, the magnetic ordering temperature is also important for applications of magnetized TI surface states. The low Curie temperature of a magnetically doped TI limits its potential for applications. In the past researchers have established [15] that interfaces between a ferromagnetic insulator and a TI can exceed the critical temperature of the isolated ferromagnet, possibly because of strong spin-orbit coupling contributed by the TI [36]. We have calculated the magnetic exchange interactions in CrSb bilayers by mapping the energies of several configurations that are meta-stable in DFT to classical Cr-spin Hamiltonians, with interlayer nearest-neighbor exchange coupling $J_1$ and intralayer nearest-neighbor exchange coupling $J_2$ (The details are provided in the supplemental materials [35]). The truncation of the spin-Hamiltonian is motivated by bulk CrSb calculations that the second nearest neighbor in-plane exchange coupling ($J_2 = 0.29$ meV) is much smaller than the nearest exchange coupling ($J_1 = 7.62$ meV). The values of the exchange couplings obtained in this way are summarized in Table II, where we find that the interlayer and intralayer exchange couplings in CrSb bilayer are similar to those in bulk CrSb. The interlayer exchange coupling $J_i$ in bulk CrSb is around -17.83 meV, vs. -19.14 meV for CrSb bilayer, and roughly doubles in magnitude the intralayer exchange coupling which is 7.62 meV, vs. 8.76 meV for CrSb bilayers. However, when the CrSb bilayer is placed on the top of the TI substrate, both $J_i$ and $J_2$ are significantly enhanced, by approximately a factor of two.

Using the values of the exchange couplings in Table II we estimated the Néel temperatures using mean-field theory. (Details are provided in the supplemental material [35].) In Fig. 2 we plot the quintuple-layer number ($N$) dependence of $T_{MF}$, which shows that the CrSb bilayer has a slightly lower $T_{MF}$ than bulk due to the smaller...
coordination number of each Cr atom, even though the exchange couplings in the CrSb bilayer are slightly larger. (See Table [I]) However, when the CrSb bilayer is placed on a Bi$_2$Te$_3$ thin film, $T_{MF}$ is much larger than that in bulk CrSb, and up to around 2050 K. The Néel temperature estimated from mean-field theory is of course larger than the expected value. In bulk CrSb $T_{MF} \approx 1150K$, which implies a ratio of $T_N/T_{MF} \approx 0.6$ when the experimental value ($\sim 700$ K) is used for the Néel temperature. In comparison, the ratio $T_c/T_{MF} \approx 0.56$ in the case of a 2D Ising model. Quantitatively the ratio of the actual critical temperature to the mean-field critical temperature is expected to increase with space-dimension and with magnetic anisotropy strength. If we assume that we are close to the strong anisotropy Ising limit, the Néel temperatures of the CrSb bilayers on TI substrates would be about 1200 K, which is much higher than room temperature and also higher than the Néel temperature of bulk CrSb.

![Figure 2](image2.png)

**FIG. 2.** Layer dependence of Néel temperature from mean-field theory. The red dot stands for bulk CrSb. The horizontal axis is the number of QLs of Bi$_2$Te$_3$, with $N = 0$ the case of freestanding CrSb bilayer.

**TABLE II.** Magnetic exchange interactions between Cr atoms obtained via the method detailed in supplemental material [35]. $J_0$ is the interlayer nearest-neighbor exchange coupling, and $J_1$ is the intralayer coupling constant.

| Material                        | $J_0$ (meV) | $J_1$ (meV) |
|--------------------------------|------------|------------|
| Bulk CrSb                      | -17.83     | 7.62       |
| Isolated CrSb bilayer          | -19.14     | 8.76       |
| CrSb bilayer/3 QL TI           | -35.22     | 17.68      |
| CrSb bilayer/4 QL TI           | -35.74     | 17.55      |
| CrSb bilayer/5 QL TI           | -35.79     | 17.64      |
| CrSb bilayer/6 QL TI           | -35.86     | 17.52      |

Anomalous Hall Conductivity— Building on the pioneering work by Karpplus and Luttinger, Smit, and Berger [1], it is now widely accepted that there are three main mechanisms contributing to the anomalous Hall effect: the intrinsic contribution due to electric-field-induced inter-band coherence in a perfect crystal [37], skew scattering [38, 39], and side-jump scattering [40]. Moreover, it has become more clear recently that the Karpplus-Luttinger mechanism is a manifestation of momentum-space Berry curvature of Bloch states [2]. The latter, whose integral over 2D Brillouin zone is the 1st Chern number times $2\pi$, is the sole contribution to the Hall conductivity in the case of quasi-two-dimensional insulators, which can therefore exhibit a quantized Hall conductivity. Since the CrSb/TI thin films we study are normally two-dimensional semimetals, we consider only the Karpplus-Luttinger mechanism. To this end we first study the electronic structures of CrSb bilayer.

Transport experiments have shown that bulk CrSb behaves as a semiconductor below the Néel temperature [34] and as a metal above it, although previous DFT calculations find that CrSb is a semimetal [41]. The resistivity $\rho$ for bulk CrSb at room temperature is around $3 \times 10^{-4}$ $\Omega cm$, which is several times larger than that of the classic semi-metal bismuth ($\rho \approx 1.3 \times 10^{-4}$ $\Omega cm$).

![Figure 3](image3.png)

**FIG. 3.** Bandstructure (left) and density of states (DOS, right) of an isolated CrSb bilayer. The left-hand side of the DOS panel corresponds to DOS= 0.

The density of states (DOS) of a freestanding CrSb bilayer in the AFM state has a minimum close to the Fermi energy (shown in Fig. 3), as does bulk CrSb. The Kohn-Sham band structure plot shows four subbands crossing the Fermi level, which are mostly derived from the 3$d$ orbitals of Cr atoms. For the case of CrSb/Bi$_2$Te$_3$ heterojunctions two of the four subbands are pushed below the Fermi level (as shown in the supplemental materials [35]), demonstrating strong hybridization between the Cr $d$ orbitals and the surface states of Bi$_2$Te$_3$, leaving two subbands that cross the Fermi level near the $\Gamma$ point and derive mainly from Bi$_2$Te$_3$ orbitals. The Fermi level DOS is smaller in the CrSb/TI case.

Given the Kohn-Sham bands, we are able to calculate the intrinsic anomalous Hall conductivities of all thin film structures using the Wannier interpolation technique [35, 42]. Fig. 4 presents intrinsic anomalous Hall conductivities for a free-standing CrSb bilayer and for CrSb/TI heterojunctions vs. chemical potential near the Fermi level.
level. The Hall conductivities are large in all cases, and tend to be larger for thicker TI films. In the free-standing CrSb bilayer case, as illustrated in Fig. 4(a), the anomalous Hall conductivity has a minimum near the Fermi level. In Fig. 4(b) and (c), \( \sigma_{xy} \) changes relatively slowly with chemical potential near Fermi level. The Hall conductivities of CrSb/TI heterojunctions have values close to \( e^2/h \) and \( 4e^2/h \) for 3 and 5 quintuple layers TI respectively. The relatively small deviation from quantized value suggests that it may be possible to realize a quantized anomalous Hall effect if a mobility gap can be induced by increasing disorder.

Large intrinsic anomalous Hall conductivities are often associated with hot-spots in momentum space that host weakly split bands states that span the Fermi energy. To identify the dominant contributions to the Brillouin zone integrals we calculated the Berry curvature along the high-symmetry lines together with the band structure [35]. For a free-standing CrSb bilayer the main contribution to Berry curvature comes from the momentum near the K point, while for the case of CrSb/TI heterojunctions the major contribution comes from regions near the \( \Gamma \) point. Even though the 3QL CrSb/TI heterojunction has a Berry curvature between the \( \Gamma \) and K points that is one order of magnitude larger than that in the case of 5 QL \( \text{Bi}_2\text{Te}_3 \), the integrated AHC is larger in the 5QL case. (See Fig. 4.) The Berry curvature hot-spots are not necessarily on high-symmetry lines and are extremely sensitive to electronic structure details.

**Discussion** — In summary, motivated by the large bulk Néel temperature \( \sim 700 \) K of bulk CrSb, we have studied the magnetic properties of free-standing CrSb bilayers and CrSb bilayers on thin TI films. We find that magnetic exchange interactions strengthen substantially when the bilayers are placed on TIs, and estimate critical temperatures that are easily in excess of room temperature. Because the inversion symmetry of bulk CrSb is broken in bilayers, the thin films are ferrimagnetic with a net magnetization around 0.7 \( \mu_B \) per unit cell. The same reduction in symmetry leads to large anomalous Hall effects. We find that the anomalous Hall effect strengthens on TI substrates [23, 24]. Since that CrSb is a semimetal with a small Fermi surface, a bulk mobility gap at the Fermi level can potentially be induced opened by disorder. If so the large intrinsic AHC may survive and would then necessarily be quantized.

**Experimental result from Refs. [23] and [24] on the 2D anomalous Hall resistance (AHR) of CrSb/TI heterojunctions and CrSb/magnetized TI heterojunctions are summarized in Table III.** Many of these observations were made on samples in which the TI has also been made magnetic by Cr doping. Generally speaking, these observations find Hall conductivities that are much smaller than in our calculations. Although this difference could

| Heterojunctions                                      | AHR (kΩ) |
|-----------------------------------------------------|-----------|
| CrSb(22 bilayers)                                   | \( \sim 10^{-5} \) |
| CrSb(22 bilayers)/TI(3 QL)                          | 0.01-0.1 |
| CrSb(13 bilayers)/MTI(3QL)                          | \( \sim 0.2 \) |
| CrSb(13 bilayers)/MTI(9QL)                          | \( \sim 0.8 \) |
| CrSb(44bilayers)/MTI(9QL)                           | \( \sim 0.6 \) |
| Cr-doped (Bi,Sb)\(_2\)Te\(_3\)                     | \( \sim 0.2 \) |
| Cr-doped (Bi,Sb)\(_2\)Te\(_3\)                     | \( \sim 0.6-0.8 \) |

* Cr-doped (Bi,Sb)\(_2\)Te\(_3\)
in principle be related to interface quality, we point out that all observations were made on thin films with many layers of CrSb. As we have emphasized the difference between the environments of antiferromagnetically coupled Cr sites is much larger in the CrSb bilayer case. Our calculations therefore motivate anomalous studies of thin films in which a single CrSb bilayer is grown on TIs.

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