Large anomalous Hall effect in ferromagnetic insulator-topological insulator heterostructures

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We demonstrate the van der Waals epitaxy of the topological insulator compound Bi$_2$Te$_3$ on the ferromagnetic insulator Cr$_2$Ge$_2$Te$_6$. The layers are oriented with (001)Bi$_2$Te$_3$∥(001)Cr$_2$Ge$_2$Te$_6$ and (110)Bi$_2$Te$_3$∥(100)Cr$_2$Ge$_2$Te$_6$. Cross-sectional transmission electron microscopy indicates the formation of a sharp interface. At low temperatures, bilayers consisting of Bi$_2$Te$_3$ on Cr$_2$Ge$_2$Te$_6$ exhibit a large anomalous Hall effect (AHE). Tilted field studies of the AHE indicate that the easy axis lies along the c-axis of the heterostructure, consistent with magnetization measurements in bulk Cr$_2$Ge$_2$Te$_6$. The 61 K Curie temperature of Cr$_2$Ge$_2$Te$_6$ and the use of near-stoichiometric materials may lead to the development of spintronic devices based on the AHE.

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Introducing a magnetic exchange gap into the Dirac spectrum of a topological insulator (TI) surface generates circumferential, half-integer quantum Hall states. Recently, independent experimental groups have detected the quantum anomalous Hall effect (QAHE) in thin films of magnetically doped topological insulators, in which the upper and lower surfaces are ferromagnetically coupled. However, the host TI materials are disordered due to the extreme concentration of magnetic dopants, which may contribute to the milli-Kelvin temperature scale at which the QAHE is observed and the reported sensitivity of the effect to fabrication details. In contrast, introducing ferromagnetism into the TI surface by proximity to a ferromagnetic insulator (FI) leaves the TI ordered, and can break time reversal symmetry without directly alloying the TI with magnetic elements. To date, epitaxy of FI-TI systems has been investigated using the FI materials EuS, GdN, and Y$_3$Fe$_5$O$_{12}$. As compared to the layered TI crystal structure, these FI compounds possess 3D structures (e.g. the NaCl-type structure in the case of EuS) producing radicals at a TI-FI interface. To create a pristine interface, a FI compound is sought with a stable, lattice-matched surface. The strong anisotropy of the Bi$_2$Te$_3$ crystal structure causes films to grow in the [001] direction, further constraining the problem by requiring a match to that lattice plane.

Here we employ the ferromagnetic insulator Cr$_2$Ge$_2$Te$_6$ (CGT) as a substrate for the growth of Bi$_2$Te$_3$, a standard TI material. Like Bi$_2$Te$_3$, the CGT structure consists of layered units terminated with hexagonal Te planes. While Bi$_2$Te$_3$ consists of quintuple layers (QL) made of five alternating layers of Te and Bi, the layered units of CGT are narrower triple layers (TL), in which two hexagonal Te planes surround a layer of Cr ions and Ge pairs. In both crystals, van der Waals bonds join adjacent planes of Te [see Fig. 1(a)]. The hexagonal lattice of Te in CGT has a lattice constant of 3.94 Å (=1/2 the lattice constant of the full cell), which we show below to be a good match to that of the Te surface of Bi$_2$Te$_3$ when rotated by 30 degrees (4.39 Å), and similarly to the Se surface of Bi$_2$Se$_3$ (4.14 Å). In addition to this fortunate crystal structure, CGT has a relatively high Curie temperature of 61 K, a resistivity greater than 10$^4$ Ω·cm below 77 K, and an easy axis of magnetization that points along the c-axis in bulk crystals. Taken together, these factors favor the study of the Cr$_2$Ge$_2$Te$_6$-Bi$_2$Te$_3$ (CGT-BT) system for the introduction of ferromagnetic order in TIs. Through Hall measurements, we show that the magnetization of CGT has a large impact on electrical transport through the Bi$_2$Te$_3$ layer.

To form CGT-BT heterostructures, we first grow large single crystals of CGT. We mix high purity Cr (99.99%), Ge (99.999%), and Te (99.999%) in a molar ratio of 2:6:36, where the Ge and Te excess acts as flux. The mixture is heated to 700 °C for 20 days and cooled to 500 °C over 36 hours, followed by centrifugation to separate the CGT crystals from the flux. The CGT crystals are then annealed in vacuum to remove any residual Te flux. Flat single crystals with sizes ranging from 0.5 to 3 mm can be manually isolated from the product. X-ray diffraction, transport measurements, and magnetization measurements confirm that the samples are single crystal (with space group R3), insulating ($E_g = 0.2$ eV), and ferromagnetic ($T_C = 61$ K).

We grow the Bi$_2$Te$_3$ layer using metal organic chemical vapor deposition (MOCVD). CGT is prepared for epitaxy using two different methods. We either adhere ~ 500 μm CGT squares to a SiO$_2$ carrier chip using polyimide or exfoliate mesoscopic crystals of CGT onto a SiO$_2$ chip using the scotch tape method. In both cases, the process prepares a freshly cleaved CGT surface prior to Bi$_2$Te$_3$ deposition. To initiate growth of Bi$_2$Te$_3$, the CGT substrate is heated to 280 °C in a 600 sccm flow of H$_2$ (6N) at 100 Torr. Tellurium and bismuth are introduced into the growth chamber by flowing hydrogen gas through diisopropyl telluride (DiPTe) and trimethyl bismuth (TMBi) bubblers, resulting in partial pressures $p_{TMBi} = 7 \times 10^{-6}$ atm and $p_{DiPTe} = 2.8 \times 10^{-5}$ atm. The precursors thermally decompose at the sample, producing atomic Bi and Te. After 2700 s, the TMBi flow is stopped, terminating growth, and the sample is cooled to 125 °C under continued DiPTe flow.
Cross-sectional transmission electron microscope (TEM) imaging allows for a direct characterization of the heterostructure (see Fig. 1). In cross-section, the layered units of the two crystal structures are directly visible, indicating that the Te layers are stacked along the growth direction. The TEM images displayed in Figs. 1(b–c) show that the interface between the two structures occurs over a distance of less than 1 nm. We attribute the high quality interface to the similarity of the two crystal structures.

Electron diffraction is used to determine the relative orientation of the Bi$_2$Te$_3$ and CGT layers. The indexed diffraction pattern is shown in Fig. 2(b). By comparing with simulated diffraction patterns, we confirm precise alignment of the Bi$_2$Te$_3$ and CGT (001) axes. Additionally, Bi$_2$Te$_3$ (110) is parallel to CGT (100). As illustrated in Fig. 2(a), this orientation corresponds to an alignment of the interfacial Te planes. Although TEM demonstrates the alignment locally, electron backscatter diffraction (EBSD) can be used to compare the Bi$_2$Te$_3$ orientation across the entire film surface. An illustration of the method is shown in Fig. 2(c), where EBSD patterns from a micron-sized Bi$_2$Te$_3$ island and the surrounding CGT are automatically indexed, showing a 30º relative rotation of the unit cells, consistent with the TEM diffraction results. Similar analysis at many points on the Bi$_2$Te$_3$ film surface shows this registry to be a global feature of the CGT-BT films.

Transport measurements are performed on the CGT-BT heterostructures. We electrically contact two types of heterostructures: (a) macroscopic Bi$_2$Te$_3$ films grown on squares of CGT ~500 µm wide and ~10 µm thick (similar to those studied with TEM) and contacted with silver paint following the van der Pauw method, and (b) mesoscopic CGT-BT bilayers formed by exfoliation of CGT on a SiO$_2$ substrate followed by Bi$_2$Te$_3$ deposition, which produces a ~150 µm thick layer of Bi$_2$Te$_3$ on a ~50 µm thick CGT layer, with lateral dimensions of tens of microns. We use electron beam lithography (EBL) to define six Ti/Au contacts to each mesoscopic sample for measurements of the longitudinal and Hall resistance. Control devices consisting of EBL-contacted exfoliated CGT on SiO$_2$ and MOCVD Bi$_2$Te$_3$ on SiO$_2$ are studied in parallel with the CGT-BT heterostructures. The Bi$_2$Te$_3$ control samples were fabricated from Bi$_2$Te$_3$ islands that grew near the CGT-BT platelets (on the same chip) and measured in the same cryostat to exclude any extrinsic origin to the AHE measured in the CGT-BT samples.

Longitudinal resistance $R_{xx}$ is measured as a function of temperature to distinguish metallic (insulating) contributions from Bi$_2$Te$_3$ (CGT). Whereas all Bi$_2$Te$_3$ control samples studied show metallic $R_{xx}(T)$ characteris-
The resistances of the CGT control samples (pre- and post-EBL) show thermally activated behavior with an energy gap of 0.2 eV, as shown in Fig. 3(a). For the CGT-BT bilayer, we construct a parallel resistor model by summing the $\text{Bi}_2\text{Te}_3$ and CGT conductivities:

$$R_{\text{CGT-BT}}(T) = \left[ (R_{0,\text{BT}} + aT)^{-1} + (R_{0,\text{CGT}}e^{E_g/\alpha T})^{-1} \right]^{-1}. \quad (1)$$

From the sample of Fig. 3(b), we extract $R_{0,\text{BT}} = 2140 \, \Omega$, $a = 1.4 \, \Omega/K$, $R_{0,\text{CGT}} = 65 \, \Omega$, and gap energy $E_g = 0.14 \, \text{eV}$. The corresponding room temperature resistivities of the two layers of this sample (width = 200 nm, length = 500 nm, $\text{Bi}_2\text{Te}_3$ thickness = 30 nm, CGT thickness = 10 $\mu$m) are $\rho_{\text{CGT}} = 40 \, \text{m}\Omega\cdot\text{cm}$, $\rho_{\text{BT}} = 0.3 \, \text{m}\Omega\cdot\text{cm}$, in agreement with typical values for both materials in the literature. The model gives a $\text{Bi}_2\text{Te}_3$ conductivity that is more than 1000 times greater than CGT at temperatures below 78 K in this sample. All other samples have thinner CGT and therefore smaller contributions of the CGT to the conductivity. At 4.2 K, the three $\text{Bi}_2\text{Te}_3$ control samples show resistivities ranging from $0.40 - 1.1 \, \text{m}\Omega\cdot\text{cm}$, while the five CGT-BT samples show resistivities in the range $0.3 - 6.2 \, \text{m}\Omega\cdot\text{cm}$, with a mean resistivity $2 \, \text{m}\Omega\cdot\text{cm}$. The similarity of the resistivity of the CGT-BT bilayers to $\text{Bi}_2\text{Te}_3$ control samples fabricated on SiO$_2$ indicates that transport in the CGT-BT samples occurs primarily in the $\text{Bi}_2\text{Te}_3$ layer.

We study magnetotransport in one CGT-BT film and in four mesoscopic CGT-BT samples, with similar results in all cases. As shown in Fig. 3(c), the $\text{Bi}_2\text{Te}_3$ control samples (and $\text{Bi}_2\text{Te}_3$ samples generally) show either a linear Hall resistance ($R_{xy}$) or a linear Hall resistance with superimposed Shubnikov de Haas (SdH) oscillations in the case of high mobility samples. In contrast, the heterostructures all show a nonlinear Hall resistance, but without large oscillations. The nonlinearity decreases with increasing temperature, as shown in Fig. 3(d). Whereas the $\text{Bi}_2\text{Te}_3$ control samples show sharp antilocalization features, the CGT-BT magnetoresistance $R_{xx}(B)$ does not. The inset of Fig. 4(a) shows a typical example of $R_{xx}(B)$. Strong magnetic dephasing may suppress localization effects in the bilayer samples.

In addition to the high field nonlinearity, the CGT-BT heterostructures exhibit a hysteretic AHE at low fields, as shown in Fig. 4. The hysteresis is absent in all control samples consisting only of CGT or $\text{Bi}_2\text{Te}_3$. The hysteresis observed in the CGT-BT samples is suggestive of a ferromagnetic origin. As shown in Fig. 4(b), the magnetoresistance, $R_{xx}$, also displays hysteresis, with peaks at the coercive fields. Tilted field measurements show that the coercive field of the hysteresis loop increases and the background Hall contribution falls off linearly as the field is rotated into the plane of the CGT-BT interface. The dependence is consistent with the easy axis of the ferromagnet pointing along the c-axis, as observed in several layered ferromagnets.

The coercive field of the hysteresis observed here, which is $\sim 0.1 \, \text{T}$ at low temperatures, is similar to the field required for saturation of bulk CGT (0.26 T). However, bulk measurements found the width of the magnetization loop to be narrower than 0.01 T, while we observe a substantial hysteresis loop in the AHE of CGT-BT heterostructures. The apparent hardening of the magnetization is consistent with recent measurements of EuS-$\text{Bi}_2\text{Se}_3$ heterostructures, where the authors attributed the increase in the observed perpendicular anisotropy to exchange coupling between the TI and FI layers. The persistence of the hysteresis to $\sim 110 \, \text{K}$, which is considerably above the known Curie temperature of bulk CGT (61 K), is also surprising.

We consider two origins of the observed high field nonlinear Hall effect. The first model considers the AHE, with the nonlinearity due to a magnetic origin. The second picture relies on a two-band model of transport, with multiple conduction channels of differing mobility. Both models result in nearly identical predictions for $R_{xy}(B)$.

The AHE, as observed in magnetic conductors, correlates strongly with the magnetization, leading to the conventional representation $R_{xy} = R_s M_\parallel + R_B B/\mu_0$ where $M_\parallel$ is the out-of-plane component of the magnetization, $R_s$ is the anomalous Hall coefficient, $B$ is the external applied field, and $R_B$ is the ordinary Hall coefficient. In magnetic conductors, nonlinearity originates in $M(B)$ which often exceeds the linear component ($R_s \gg R_0$) to the extent that $R_{xy}$ appears to saturate at accessible fields, as shown in Fig. 3(c). Studies of EuS-$\text{Bi}_2\text{Se}_3$ ascripe a similar, but smaller ($R_s \ll R_0$) Hall nonlinearity to a ferromagnetic AHE, since it correlates clearly with the

![FIG. 3. Transport in control samples and CGT-BT devices.](image-url)
Brillouin dependence of the magnetization of EuS. In contrast, the nonlinear Hall effect in CGT-BT does not reflect the typical $M(B)$ of CGT. Magnetization measurements show that CGT saturates at fields below 0.3 T, while here we observe saturation at ~ 6 T. However, it is possible that ferromagnetic order within the CGT is strongly modified near the interface, where it would contribute most to the Hall effect.

Alternatively, the presence of parallel conduction channels with differing mobilities but comparable overall conductance will produce a similar nonlinearity, as observed in many heterojunction systems. The CGT bulk is insulating at low temperatures, so any conduction channels lie in the Bi$_2$Te$_3$ bulk or at the CGT-BT interface. In the two-band model, the measured Hall resistance depends on the magnetic field as

$$R_{xy}(B) = \left(\frac{B}{e}\right) \left[\frac{n_1 \mu_1^2 + n_2 \mu_2^2}{n_1 \mu_1 + n_2 \mu_2} + \frac{(B \mu_1 \mu_2)^2 (n_1 + n_2)}{(n_1 \mu_1 + n_2 \mu_2)^2 + (B \mu_1 \mu_2)^2 (n_1 + n_2)^2}\right],$$

where $n_i$ are the two-dimensional carrier densities, $\mu_i$ are the mobilities, and $e$ is the carrier charge. This model nicely fits the data [see fits plotted in Fig. S3(c)], and implies a high mobility, low density (HMLD) channel and a low mobility, high density (LMHD) channel of the same sign (n-type) in all samples. Due to variations in film thickness and roughness, $n_i$ and $\mu_i$ vary within one order of magnitude from sample to sample, and we consider the mean values for discussion. At 4.2 K, the mean 2D carrier densities are $n_{\text{HMLD}} = 5 \times 10^{15} \text{ cm}^{-2}$, $n_{\text{LMHD}} = 6 \times 10^{13} \text{ cm}^{-2}$ and the mean mobilities are $\mu_{\text{HMLD}} = 200 \text{ cm}^2/\text{V s}$, $\mu_{\text{LMHD}} = 7,000 \text{ cm}^2/\text{V s}$. The samples with the highest $\mu_{\text{LMHD}}$ obtained from the fits (40,000 cm$^2$/V s) far exceed the highest known mobility for Bi$_2$Te$_3$. However, large SdH oscillations are not observed in any of the CGT-BT samples.

One of the channels is consistent with bulk transport through the Bi$_2$Te$_3$ layer. The fit parameters of the LMHD channel are similar to those extracted from the Bi$_2$Te$_3$ control samples. The mean carrier density of the LMHD channel is $n_{\text{3D}} = 3 \times 10^{20} \text{ cm}^{-3}$, a value typical of strongly doped Bi$_2$Te$_3$ and close to the control samples $n_{\text{3D}} = 4 \times 10^{19} \text{ cm}^{-3}$. The mobility of this channel (200 cm$^2$/V s) is also similar to the control samples (400 cm$^2$/V s). While the LMHD channel is consistent with bulk Bi$_2$Te$_3$ conduction, the second high-mobility channel suggested by the two-band model could only originate from the interface between the two materials, since it is absent in either type of control sample. In principle, a LMHD channel could be present at the interface, but this does not explain the noted absence of SdH oscillations.

We demonstrate ferromagnetic coupling in a TI-FI heterostructure with a high level of crystalline order. To date, hysteretic AHE has not been observed in TI-FI heterostructures. The large magnitude of both the hysteretic AHE and the high field nonlinearity in this system make this an exciting development for realizing more complex experiments based on TIs. The hysteretic AHE reflects the bulk properties CGT, while the high-field nonlinearity implies that either an additional type of magnetic order impacts transport through the Bi$_2$Te$_3$, or that a high mobility state arises at the interface. Tuning the Fermi energy of the Bi$_2$Te$_3$ by applying a gate potential through the CGT layer would do much to clarify the properties of the AHE and reduce bulk conduction in the Bi$_2$Te$_3$. It is likely that heterostructures with more insulating Bi$_2$Te$_3$ and therefore greater sensitivity to the interface, will display still further enhanced AHE.

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