ABSTRACT: The search of novel topological states, such as the quantum anomalous Hall insulator and chiral Majorana fermions, has motivated different schemes to introduce magnetism into topological insulators. A promising scheme is using the magnetic proximity effect (MPE), where a ferromagnetic insulator magnetizes the topological insulator. Most of these heterostructures are synthesized by growth techniques which prevent mixing many of the available ferromagnetic and topological insulators due to difference in growth conditions. Here, we demonstrate that MPE can be obtained in heterostructures stacked via the dry transfer of flakes of van der Waals ferromagnetic and topological insulators (Cr$_2$Ge$_2$Te$_6$/BiSbTeSe$_3$), as evidenced in the observation of an anomalous Hall effect (AHE). Furthermore, devices made from these heterostructures allow modulation of the AHE when controlling the carrier density via electrostatic gating. These results show that simple mechanical transfer of magnetic van der Waals materials provides another possible avenue to magnetize topological insulators by MPE.

KEYWORDS: magnetic proximity effect, topological insulator, 2D ferromagnetic insulators, van der Waals heterostructures, anomalous Hall effect
plane magnetic field (current) selective areas of the top surface of a topological insulator gate-tunable bottom surface. The ability to magnetize local/involving a magnetized top surface with an AHE in parallel to a constraints and limitations found in previous works. The gate approaches) is also important in various proposed schemes to realized in a fully vdW heterostructure bypassing growth-based schemes to manipulate and measure Majorana fermions in superconductor/topological insulator hybrids. We use thin flakes mechanically exfoliated from the vdW layered Cr$_2$Ge$_2$Te$_6$ (CGT) as the ferromagnetic insulator in our heterostructure. Single crystals of this material were grown via a self-flux technique as described in a previous work. For the topological insulator, we employ flakes exfoliated from a single crystal of BiSbTeSe$_2$ (BSTS) grown by the Bridgman technique as described in ref 23. Taking advantage of the layered nature of these crystals, it is possible to assemble heterostructures while avoiding those defects and irregularities in the interface that might occur in heterostructures assembled via growth due to lattice mismatch and chemical diffusion (both issues can be detrimental for AHE observation). We chose topological insulator flakes with a thickness in the range of 40–70 nm, which maintain the intrinsic insulating bulk and conduct mainly through the surface states. For the ferromagnetic insulator, we chose flakes with a thickness in the range of 4–10 nm, whose low temperature magnetization shows a more rectangular hysteresis loop with a clear coercivity and higher remnant magnetization compared to the smooth magnetization behavior for bulk CGT and the more complicated hysteresis loops with softer magnetic behavior in thicker CGT flakes (>10 nm). The latter are usually attributed to the formation of labyrinth type domains (see Figure S1 for an evolution of hysteresis loops of CGT as thickness is increased, measured using Magneto Optics Kerr Effect). This distinct magnetic hysteresis loop behavior was expected to be inherited into the topological insulator and would make the AHE more distinguishable from other nonlinear Hall effects. In Figure 1a and b, a schematic of the heterostructures and the microscope image of a typical device are shown, respectively. As can be seen, the CGT flake did not fully cover the topological insulator. This was done to ease the contact fabrication, as CGT is highly insulating at low temperatures (see Figure S2) and a full coverage of the BSTS flakes would lead to poor contacts. In Figure 1c, an ambipolar field effect in the longitudinal resistance measured by the four-probe method can be observed, showcasing that the heterostructure still allows similar gate tuning of the resistance as in BSTS-only devices. An out-of-plane magnetic field is applied and swept to extract the Hall resistance in the device. As seen in the figure, a clear AHE with a rectangular hysteresis loop with an amplitude of a few ohms and a coercive field of ~0.035 T is observed in the $R_{xy}$ vs $V_{BG}$ curve, indicating that a magnetization in the conducting carriers is introduced by interfacing the topological insulator BSTS with the magnetic insulator CGT. Furthermore, the longitudinal resistance ($R_{xx}$) of the device shows a minimum in the magnetoresistance around zero field, typically attributed to the weak antilocalization (WAL) behavior due to the strong spin orbit coupling in the topological insulator. Phenomenologically, the magneto-resistance can be fitted to the Hikami–Larkin–Nagaoka equation used to describe the WAL, and how the fitting parameters vary with the back gate (see Figure S3) is similar to the WAL behavior reported in the literature.
To further study the hysteresis loop (attributed to AHE) in $R_{xy}$, the main focus of this paper, we measure $R_{xy}$ as the function of the gate voltage ($V_{BG}$, applied to the silicon substrate as a global back gate), shown in Figure 2a. As can be seen, there is a change in the magnitude of the hysteresis loop along with a changing slope of the linear background in the Hall resistance for changing $V_{BG}$. After removing a smooth polynomial background, the step size $\Delta R_{xy}$ (assigned as the amplitude of AHE) can be obtained. The result is plotted as a function of $V_{BG}$ in Figure 2b, showing a tunability in the AHE amplitude from $\sim 4 \Omega$ to $\sim 11 \Omega$. The longitudinal resistance $R_{xx}$ (measured at the minimum of the magnetoresistance) is also plotted as a function of $V_{BG}$ in the same figure, and a correlation with $\Delta R_{xy}$ is observed. The extracted Hall coefficient (linear slope of $R_{xy}$ vs magnetic field, reflecting the ordinary Hall effect) is plotted in Figure 2c and is clearly modulated (both the sign and amplitude) by the back gate. The change in the sign indicates the tunability of the dominant carrier type in the device from holes to electrons (in the trivial Hall effect scenario the hysteresis or $\Delta R_{xy}$ would also change sign when the main carrier type or Hall slope changes sign). Furthermore, we have estimated that the strength of the fringe magnetic field in such devices (no more than 1 mT) is too weak to produce the observed $\Delta R_{xy}$ (of $\sim 10 \Omega$, which would require a change in magnetic field on the order 0.1 T, as seen in Figure 1d).

We also studied the temperature dependence of the AHE. Figure 3a shows the evolution of $R_{xy}$ as the temperature is increased, where the hysteresis loop (AHE amplitude $\Delta R_{xy}$)
doping in BSTS (no more than our own measurements, also suggest a similar moderate hole consistent with the Curie temperature (Figure 3b) is seen to vanish between 38 and 60 K. This is from CGT (equivalently, electron transfer from TI to CGT).

Finally, to further shed light onto the gate tunability of the AHE, we present a simple model of two parallel channels of conduction that can largely reproduce key features of our results (the details are left to the Supporting Information). The model consists of two conduction channels representing a gate tunable bottom surface without AHE and a top surface with fixed AHE (not gate-tunable, motivated by the experimental situation that the top surface is much less affected by the metallic surface state at such temperatures).\(^{25}\) It is worth noting that our BSTS flake is only partially covered by CGT on the top surface; therefore the unmagnetized bottom surface and unmagnetized portion of the top surface are expected to make an important contribution to \(R_{xy}\).

Our measurement in CGT shows that it becomes highly insulating with negligible conductance at low temperatures (Figure S2). While the AHE could in principle also arise from CGT that becomes conductive after charge transfer from BSTS, we believe this is a less likely scenario to explain our observations. Previous studies on TI/CGT have noted moderate hole doping in the TI due to charge (hole) transfer from CGT (equivalently, electron transfer from TI to CGT).\(^{35,36}\) All showed that the CGT is p-doped itself. Therefore, for the expected hole doping in BSTS after interfacing with CGT (therefore n-doping or electron transfer on CGT), the charge transfer would make CGT more intrinsic and even more insulating (as indeed shown in previous field-effect studies in CGT such as\(^{35}\)) thus unlikely to become a conducting channel exhibiting its own AHE. Finally, a very large charge transfer from our TI to CGT to make it conductive would heavily dope the TI, and this is not consistent with the actual observed field effects in our devices.

In conclusion, we demonstrated that thin flakes of the topological insulator BSTS can show an anomalous Hall effect when interfaced with thin flakes of CGT using simple mechanical dry transfer methods for the assembly. The observed AHE shows a rectangular hysteresis loop with a coercive field of \(\sim 0.035\) \(T\) at 2 K and vanishes close to the \(T_C\) of thin CGT flakes. Furthermore, the AHE can be tuned from \(\sim 4\) to \(\sim 11\) \(\Omega\) and follows the longitudinal resistance of the device as a back gate is applied. These results indicate that simple mechanical transfer of van der Waals materials allows for a magnetic proximity effect in the surface state of a topological insulator, a basis to realize novel magnetic topological phases such as QAHI and to implement various proposed devices schemes for measuring and manipulating Majorana fermions in topological insulator/superconductor hybrid structures. Our work may further facilitate interfacing the growing inventory of layered materials with different magnetic or electrical properties.

**EXPERIMENTAL METHODS**

**Device Fabrication.** BSTS and CGT thin flakes were exfoliated on SiO\(_2\)/Si substrates inside a glovebox, where the oxygen and water concentration were less than 5 ppm. We chose appropriate CGT thin flakes (thickness between 4–10 nm) and BSTS flakes (thickness between 40–70 nm) based on the optical contrast with the thickness later confirmed by atomic force microscopy. We transferred the freshly exfoliated CGT flakes using the dry transfer technique on top of fresh BSTS flakes. Briefly, we used a PDMS/poly carbonate (PC)
stamp to transfer the CGT flake from its initial substrate to the top of the chosen BSTS flake, the CGT flake is dropped along with the PC carrier film. To remove the PC film the sample is rinsed with chloroform for 10 min followed by acetone and isopropyl alcohol solvent cleaning. Then, the heterostructure is patterned using electron beam lithography. Finally, 5 nm of Cr and 95 nm of Au are deposited as contacts using e-beam evaporation. The prepared devices are then mounted for electrical transport measurements as quickly as possible after liftoff, usually with no more than half an hour to an hour of exposure to air to avoid degradation of CGT.

**Electrical Transport Measurements.** Electrical transport measurements are performed in a variable temperature insert (VTI) system, which allows temperatures from 1.8 K to 300 K and an applied magnetic field of up to 6 T. The longitudinal resistance and Hall resistance were measured using a four probe method with a Stanford Research SR830 lock-in amplifier with a low frequency (∼13 Hz) excitation current of 100 nA to 1 μA or by applying 1–10 μA of DC current with a Keithley 2400 source meter and measuring the voltage drop with a Keithley 2182A nanovoltmeter. The gate control was achieved by applying a DC voltage to the p-doped silicon substrate with a Keithley 2400 source meter and measuring the voltage drop with a Keithley 2182A nanovoltmeter. The gate control was achieved by applying a DC voltage to the p-doped silicon substrate using a Keithley 2400 source meter and measuring the voltage drop with a Keithley 2182A nanovoltmeter. The gate control was achieved by applying a DC voltage to the p-doped silicon substrate using a Keithley 2400 source meter and measuring the voltage drop with a Keithley 2182A nanovoltmeter.

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**Notes**
The authors declare no competing financial interest.

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**ASSOCIATED CONTENT**

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Thickness dependence of the hysteresis loops of the magnetization of CGT (as probed with MOKE, Figure S1), temperature dependence of current in bare CGT indicating its insulating nature at low temperatures (Figure S2), the HLN equation fits to the magneto-resistance (Figure S3), AHE in other devices and their temperature dependence (Figure S4), comparison of MOKE and AHE measured on the same CGT/TI device (Figure S5), and the temperature dependence of $R_{xx}$ (Figure S6); a description of the simple model used for Figure 4a and b detailed along with the parameters used (PDF)
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