Two-dimensional ferromagnetic superlattices

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ABSTRACT

Mechanically exfoliated two-dimensional ferromagnetic materials (2D FMs) possess long-range ferromagnetic order and topologically nontrivial skyrmions in few layers. However, because of the dimensionality effect, such few-layer systems usually exhibit much lower Curie temperature ($T_C$) compared to their bulk counterparts. It is therefore of great interest to explore effective approaches to enhance their $T_C$, particularly in wafer-scale for practical applications. Here, we report an interfacial proximity-induced high-$T_C$ 2D FM Fe$_3$GeTe$_2$ (FGT) via A-type antiferromagnetic material CrSb (CS) which strongly couples to FGT. A superlattice structure of (FGT/CS)$_n$, where $n$ stands for the period of FGT/CS heterostructure, has been successfully produced with sharp interfaces by molecular-beam epitaxy on 2-inch wafers. By performing elemental specific X-ray magnetic circular dichroism (XMCD) measurements, we have unequivocally discovered that $T_C$ of 4-layer Fe$_3$GeTe$_2$ can be significantly enhanced from 140 K to 230 K because of the interfacial ferromagnetic coupling. Meanwhile, an inverse proximity effect occurs in the FGT/CS interface, driving the interfacial antiferromagnetic CrSb into a ferrimagnetic state as evidenced by double-switching behavior in hysteresis loops and the XMCD spectra. Density functional theory calculations show that the Fe-Te/Cr-Sb interface is strongly FM coupled and doping of the spin-polarized electrons by the interfacial Cr layer gives rise to the $T_C$ enhancement of the Fe$_3$GeTe$_2$ films, in accordance with our XMCD measurements. Strikingly, by introducing rich Fe in a 4-layer FGT/CS superlattice, $T_C$ can be further enhanced to near room temperature. Our results provide a feasible approach for enhancing the magnetic order of few-layer 2D FMs in wafer-scale and render opportunities for realizing realistic ultra-thin spintronic devices.

Keywords: 2D ferromagnetic material, room temperature, 2-inch Fe$_3$GeTe$_2$ film wafers, proximity effect, (Fe$_3$GeTe$_2$/CrSb)$_n$ superlattice

INTRODUCTION

Two-dimensional (2D) systems involving various functionalities are a central topic in condensed matter physics. Since the discovery of graphene [1,2], the 2D material family has been widely explored in semiconductors [3,4], superconductors [5,6], and ferromagnetic materials (FMs) [7–12]. In particular, spintronic devices based on 2D FMs have attracted significant attention, for example, magnon-assisted tunneling and giant tunneling magnetoresistances were found to possess multiple magnetic states in CrX$_3$ (X = Br and I)-based junctions [13–17]. In 2D FMs, the perpendicular magnetic anisotropy that is partially contributed by spin-orbit coupling plays a more essential role in magnetic order as the thickness reduces [8,11]. Theoretically, because of strong spin-orbit coupling and broken inversion symmetry, Dzyaloshinskii-Moriya interactions [18,19] can provide topological magnetic textures, thus inducing skyrmions. Using Lorentz transmission electron microscopy (TEM), at low temperatures Néel-type skyrmions (magnetic...
bubbles) have been observed in Fe₃GeTe₂ (CrₓGeTe₂) with controllable transitions between skyrmions and magnetic domains [20,21]. However, one unprecedented challenge still exists, that is the suppressed Curie temperature (T_C) as the thickness of 2D FMs decreases [7,11]; this is ascribed to the dimensionality effect of the competing perpendicular magnetic anisotropy energy with thermal fluctuations [11,22]. Modulation of the ferromagnetic properties in few-layer 2D FM s, such as enhancing T_C or the control of the coercive field (H_C), provides a route towards realistic spintronic applications using 2D FM s.

Recent studies have unveiled the gate-controlled ferromagnetic order in 2D FM nanoflakes. As an example, the ferromagnetic parameters of T_C and H_C for monolayer CrI₃ can be tuned via h-BN gating [23,24], and bilayer CrI₃ exhibits a reversible transition between antiferromagnetic (AF) and FM states [25]. Compared to other 2D FM s, Fe₃GeTe₂ is more stable among the recently explored 2D FM s; and by changing the Fe composition [9,26] and applying ionic-liquid gating [11], the ferromagnetism of Fe₃GeTe₂ can be modulated. Complementary to these doping and gating techniques, the proximity effect can induce a stable ferromagnetic order through interface coupling [27,28] and avoid the inconvenience of using dielectric gates for the transient FM states. For example, in Bi₂Se₃/EuS heterostructures, Bi₂Se₃ possesses room-temperature ferromagnetism which is far above the intrinsic T_C of EuS (17 K) as a result of large spin-orbit coupling [29]; the quantum anomalous Hall effect in graphene has been proposed by proximity coupling [30]; and the proximity Cr layer favors the T_C enhancement of Fe₃GeTe₂ films rather than the interfacial strain effect, and that the interfacial Cr layers retain the interlayer AF coupling but have a net magnetic moment. Furthermore, by designing 4-layer Fe-rich Feₓ⁺ₓGeTe₂ and ~1.6 nm CrSb with the same superlattice structure, we have accomplished the highest T_C of 286.7 K in (Feₓ⁺ₓGeTe₂/CrSb)₃ superlattice, which approaches room temperature with a stable ferromagnetic order over 12 months.

RESULTS AND DISCUSSION

Layer structured Fe₃GeTe₂ has a hexagonal structure with a space group P6/ mmc and lattice constants of a = b = 3.991 Å and c = 16.396 Å [34], in which each layer consists of five sublayers with Fe₂Ge slab sandwiched between two Te layers [9,34]; and the A-type antiferromagnetic (A-AF) CrSb [35] is a NiAs-type structure with a space group of P6/ mmc and lattice constants of a = b = 4.108 Å and c = 5.440 Å [36], which serves as an ideal candidate for the epitaxial growth of FM/AF heterostructures. Periodic reflection high-energy electron diffraction (RHEED) intensity oscillations of Fe₃GeTe₂ suggest a layer-by-layer growth mode as shown in Fig. 1a. It typically takes 167±8 s to complete 1-layer Fe₃GeTe₂ growth, guaranteeing the fine controllability of the film thickness. Detailed growth conditions and high-crystalline characterizations are discussed in detail in the Method section and Supplementary Figs 1–2. Figure 1b displays a typical X-ray diffraction (XRD) spectrum for a (FGT/CS)₉ superlattice, in which all diffraction peaks can be exclusively indexed as Fe₃GeTe₂ and CrSb without any mixtures or new compounds generated, with a schematic geometry of FGT/CS (Fig. 1c). The interlayer distance of Fe₃GeTe₂ is half of c-axis of ~0.8 nm (Fig. 1d), consistent with the (002) diffraction peak shown in Fig. 1b and the refinement-XRD value of 8.17 Å [34]. Sharp interfaces between Fe₃GeTe₂ and CrSb are shown in the high-angle-annular-dark-field (HAADF) image (Fig. 1d) with the enlarged part displayed in Fig. 1e, further excluding the possibility of element mixing at these interfaces or nearby layers.
We then examined the thickness-dependent magnetic properties of Fe₃GeTe₂ films by an anomalous Hall effect (AHE) in the Hall-bar geometry with a size of 1.5 × 2 mm². Even down to 4 layers, the easy axis of Fe₃GeTe₂ is still along the c-axis (out-of-plane) as \( H_C \) increases with the angle switching from 0° to 90° (see details in Supplementary Fig. 3a). As displayed in Fig. 1f, the Curie temperature (see details about Arrrott-plots in Supplementary section 1) shows bulk-like behavior with \( T_C \) of ~216.4 K when the thickness is above 8 nm (~10 layers). However, when reducing the thickness, \( T_C \) displays a declined trend and exhibits a dramatic drop below 7-layers. Further reducing to the bilayer, Fe₃GeTe₂ retains a ferromagnetic state with \( T_C \) of 138.4 ± 1.6 K. As the Fe₃GeTe₂ thickness approaches the 2D limit, the thickness-dependent \( T_C(N) \) can be described by a universal scaling law [37–39] written as \( (T_C(\infty) - T_C(N))/T_C(\infty) = ((N_0 + 1)/2N)\lambda \), where \( T_C(\infty) \) denotes the \( T_C \) of the bulk crystal, the critical exponent \( \lambda \) reveals the universality class of the transition, and \( N_0 \) is the critical layer number referring to the mean spin-spin interaction range and separating the boundary between the 2D and 3D magnetism. The scaling law function is fitted to the experimental data yielding \( N_0 \sim 3.52 \pm 0.72 \) and \( \lambda \sim 1.79 \pm 0.38 \). The deduced \( \lambda \) also suggests that Fe₃GeTe₂ belongs to the Ising-type ferromagnet (note that \( \lambda = 1.42 \) is from the Heisenberg model [40], \( \lambda = 1.56 \) from the Ising model [41], and \( \lambda = 1 \) from mean-field theory [38]). A crossover from 3D to 2D Ising ferromagnetism with the thickness decreasing has been reported in Fe₃GeTe₂ nanoflakes [10]. The strong dimensionality effect can be commonly explained by the competition between the magnetic anisotropy energy and prominent thermal fluctuations in thinner samples [7,22].

To enhance the Curie temperature of Fe₃GeTe₂, we geometrically designed FGT/CS superlattices with different periods and thickness. Detailedcharacterizations of A-AF CrSb under in-plane and out-of-plane magnetic fields are presented in Supplementary Fig. 4, with no sign of ferromagnetism in both measurement geometries. Here, we denote Fe₃GeTe₂/CrSb superlattices to be (FGT/CS), where \( n \) is the period. Unless specifically mentioned, hereafter, the thickness of Fe₃GeTe₂ and CrSb is ~3.2 nm (4-layer) and ~1.6 nm, respectively. \( n = 1 \) stands for the single-period structure
of ~3.2 nm Fe₃GeTe₂ and ~1.6 nm CrSb, as schematically shown in Fig. 2a. To confirm the magnetic anisotropy of (FGT/CS)₃ superlattice, angle-dependent AHE was performed (Fig. 2b). The easy axis of (FGT/CS)₃ is still along the out-of-plane direction, sharing the same perpendicular magnetic anisotropy as the pure Fe₃GeTe₂. Here, the temperature-dependent AHE was measured under perpendicular geometry (Fig. 2c inset). At low temperatures such as 2.5 K (Fig. 2c), the AHE presents a resistance switching at ±0.97 T accompanied by another weaker switching behavior at a relatively low field. The origin of such property most likely comes from the FM/AF interface [33,42–44] and we define this phenomenon as double-switching behavior. With the temperature increasing, this behavior becomes inconspicuous at ~55 K, indicating decrease of the interface coupling. Figure 2d presents the switching behavior of the minor loops at low fields. Here, the exchange field (H_EX) is designated to describe the double-switching behavior. Negative H_EX in minor loopO indicates that it is a parallel ferromagnetic coupling between the interfacial Fe₃GeTe₂ and CrSb [42]. This double-switching property can also be observed at low-temperature magnetization hysteresis (M-H curves, Fig. 2e). Accompanied by such a double-switching phenomenon, we uncovered that the T_C of this (FGT/CS)₃ superlattice was raised to 206.3 ± 1.6 K (calculated by Arrott-plots, inset of Fig. 2f), reasonably close to the T_C of ~201 K determined by zero-field-cooled and field-cooled (ZFC-FC) curves (Fig. 2f). Therefore, a dramatic enhancement of T_C over 60 K is achieved when compared to 140.3 K in 4-layer Fe₃GeTe₂. The evolutions of the double-switching AHE and M-H curves at various temperatures are provided in Supplementary Figs 5–8. Conjointly, the T_C modulation and double-switching effect are closely related to the interfacial coupling between the ferromagnetic Fe₃GeTe₂ and antiferromagnetic CrSb.

We further conducted element-specific XMCD at Fe and Cr L₂,₃ absorption edges to probe the local electronic character and investigate how the proximity interfacial interaction evolves in (FGT/CS)₃. In the XMCD measurement, circularly polarized X-ray with 100% left and right polarization was used, denoted as \( \mu^- \) and \( \mu^+ \), respectively. XMCD is defined as the difference of the X-ray absorption spectroscopy (XAS), written as the equation \( X M C D = \mu^- - \mu^+ \). Figure 3a presents a typical pair of XAS and XMCD spectra of Fe L₂,₃ edge.
NotethatthethicknessesofFe\textsubscript{3}GeTe\textsubscript{2} andCrSb are defined in the equation
\[ \beta = \frac{\mu^+ - \mu^-}{\mu^+ + \mu^-} \]
\[ T_C = 208.6 \pm 7.5 \text{ K} \]
by fitting the temperature-dependent Fe XMCD percentage using the empirical equation (1 - \[ T/T_C \])\textsuperscript{y} \[ [46,47] \], consistent with that obtained from the Arrott-plots (206.3 \pm 1.6 K). (e) \[ T_C \] versus the period \( a \), which increases \( \sim 60\% \) from 140.3 \( \pm \) 2.7 K of the pure 4-layer FGT to 230.9 \( \pm \) 1.3 K in (FGT/CS)\textsubscript{10} superlattice. Note that the thicknesses of Fe\textsubscript{3}GeTe\textsubscript{2} and CrSb are \( \sim 3.2 \text{ nm} \) (4-layer) and \( \sim 1.6 \text{ nm} \), respectively.

obtained using total electron yield detection mode. The XAS spectra, in good agreement with Fe\textsubscript{3}GeTe\textsubscript{2} bulks \[ [45] \] in the spectrum shape and energy positions, confirm that the Fe-magnetism originates from the Fe\textsubscript{3}GeTe\textsubscript{2} region. Consistent with the \( T_C \) determined by the AHE measurements (206.3 K), the XMCD signals can be distinguished at 200 K and vanish at 300 K (Fig. 3a). Significantly, Cr \( L_{2,3} \) spectra give a strong XMCD dichroism at 3 K (Fig. 3b), indicating the newly developed Cr magnetic state at the interface. This Cr ferrimagnetic order can be detected at 50 K and becomes much weaker when approaching 100 K (Fig. 3b). By revisiting the AHE measurements, we note that the two magnetic states from intrinsic Fe\textsubscript{3}GeTe\textsubscript{2} and interfacial CrSb can individually contribute to the resistance jump at each \( T_C \), and accordingly, the double-switching behavior occurs. Here, XMCD percentage (\( \beta \)), defined by \[ \beta = \frac{\mu^+ - \mu^-}{\mu^+ + \mu^-} \]
is used to analyze the ferromagnetism. By fitting the temperature-dependent Fe XMCD percentage to the empirical function of (1 - \[ T/T_C \])\textsuperscript{y} \[ [46,47] \], \( T_C \) is calculated to be 208.6 \( \pm \) 7.5 K (Fig. 3d), which is consistent with the magneto-transport measurements. The same positive trends of Cr and Fe spectra as a function of the magnetic field indicate parallel interfacial ferromagnetic coupling between Fe\textsubscript{3}GeTe\textsubscript{2} and CrSb (see details in Supplementary Fig. 10) \[ [47] \], which agrees with the negative \( H_{EX} \) at minor loop\( \Phi \) (Fig. 2d).

Now, we summarize the various \( T_C \) for (FGT/CS)\textsubscript{n} superlattices as a function of period \( n \). Here, \( T_C \) is extracted by Arrott-plots (Supplementary Figs 11–12). As shown in Fig. 3e, a giant improvement of \( T_C \) is observed in the (FGT/CS)\textsubscript{n} superlattice. Once the FGT/CS bilayer is established, denoted as (FGT/CS)\textsubscript{1}, \( T_C \) can be noticeably raised to 178.7 \( \pm \) 2.5 K, \( \sim 40 \) K higher than that of the pure 4-layer Fe\textsubscript{3}GeTe\textsubscript{2}. \( T_C \) increases continuously as \( n \) increases to above 5, above which it saturates at \( \sim 230 \) K. Despite the dimensionality effect on the \( T_C \) of pure Fe\textsubscript{3}GeTe\textsubscript{2} which is given in detail in Fig. 1f, when comparing the \( T_C \) in pure 4-layer FGT with (4-layer FGT/CS)\textsubscript{1}, and \( T_C \) in bulk FGT with (4-layer FGT/CS)\textsubscript{5} where the thickness of FGT in the superlattices is fixed at 4-layer, we believe that besides the dimensionality effect, the FGT/CS interfacial interactions play a
more important role in the $T_C$ increase in these superlattices. The mechanism is proposed in the following section.

**THEORETICAL CALCULATION**

To seek the origin of such a $T_C$ enhancement in the FGT/CS superlattices, we performed DFT calculations within the Generalized Gradient Approximation (GGA) plus U framework. Taking into account the robust FM ground state of Fe$_3$GeTe$_2$ monolayer and the A-AF state of CS (Supplementary section 5), we constructed four different magnetic states (Fig. 4a–d) for the FGT/CS superlattice to address three magnetic couplings at the Fe-Te/Cr-Sb interface (I), at the Fe-Te/Sb-Cr interface (II) and in-between the two FGT van der Waals (vdW) layers (III), corresponding to the exchange constants $J_1$, $J_2$ and $J_3$, respectively (Fig. 4e).

We mapped the calculated energy differences (summarized in Table 1) onto a simple $(J_1, J_2, J_3)$ magnetic exchange model. We found that while the Fe-Te/Cr-Sb interface has a strong FM coupling ($J_1 = 39$ meV), the Fe-Te/Sb-Cr interface is very weakly AF coupled ($J_2 = -1$ meV), and the two FGT vdW layers have moderate FM coupling ($J_3 = 6$ meV). The very weak $J_2$ coupling is a result of large Fe–Cr separation by Te–Sb atoms. The strong $J_1$ exchange is associated with the intact Fe–Te–Cr pathway in which the Cr atom moves closely to Te to remove its otherwise dangling bond. The moderate $J_3$ value of 6 meV here is comparable to the calculated value of 7.5 meV for the FGT bulk, where the experimental vdW interlayer Te–Te distance of 2.94 Å is smaller than the optimized theoretical value of 3.02 Å. Therefore, we suggest that the FGT/CS superlattice has a strong FM Fe-Te/Cr-Sb interface (I) but a weak AF Fe-Te/Sb-Cr interface (II) and moderately FM coupled vdW FGT monolayers (III).

As the Fe-Te/Cr-Sb interface I is strongly FM coupled, it tunes the magnetic behavior of the interfacial FGT monolayer: when this FGT monolayer is changed from the FM ground state to the tri-layered AF state (i.e. up-up-up spins to up-down-up spins as shown in Supplementary Fig. 14c–d, respectively), the total energy rises drastically, from 595 meV/fu for a bare FGT monolayer to 820 meV/fu (per fu of FGT). In contrast, the corresponding energy difference is reduced to 424 meV/fu for the FGT monolayer lying at the Fe-Te/Sb-Cr interface II, which is weakly AF coupled. Obviously, the significant enhancement of FM coupling in the FGT vdW layer at the Fe-Te/Cr-Sb interface I dominates

![Figure 4. DFT calculations for FGT/CS superlattice.](image)

**Table 1.** $(J_1, J_2, J_3)$ magnetic exchange model, relative total energy $\Delta E$ (meV/cell) and total magnetic moments (Ttot, $\mu_B$/fu) of the FGT/CS.

| FGT/CS | Energy* (meV/fu) | $\Delta E$ (meV/fu) | Ttot ($\mu_B$/fu) |
|--------|------------------|--------------------|-------------------|
| FM     | $J_1 - J_3 + J_2$ | 0                  | 7.89              |
| Layered-AF1 | $J_1 + J_2 + J_3$ | 90                 | 0.30              |
| Layered-AF2 | $J_1 + J_3 - J_2$ | 14                 | 0.46              |
| Layered-AF3 | $J_1 - J_2 - J_3$ | 80                 | -7.16             |

* $J_1 = 39$ meV, $J_2 = -1$ meV, $J_3 = 6$ meV are derived.
over the reduction at the Fe-Te/Sb-Cr interface II (caused by negative effects from the tensile strain (see Supplementary section 5) and the weak AF interfacial coupling here). Moreover, it is believed that for the interior FGT vdW layers in the FGT/CS superlattice, their intralayer and interlayer FM couplings should be very similar to their bulk cases.

CONCLUSION

In Fig. 4e, we plot the changes of atomic charge (Δq) and magnetic moments (Δm) of a representative 2-layer FGT/1.6nm-CS superlattice against the FGT monolayer and the CS bulk. It can be observed that the Fe-Te/Cr-Sb interface I has much larger charge/moment changes than those at the Fe-Te/Sb-Cr interface II. More specifically, for the ‘more important’ Fe-Te/Cr-Sb interface I, the Cr atoms donate some electrons to the neighboring FGT vdW layer, and therefore the Cr atoms and FGT monolayer both have increased magnetic moments. Together, these contribute to the above significant enhancement of FM coupling in the FGT/CS superlattice. Moreover, because of this spin-polarized charge transfer, the interlayer AF coupled Cr layers in CrSb become ferrimagnetic and thus have a net magnetic moment, which accounts for the above XMCD observations (Fig. 3c).

Inspired by the $T_C$ tunability in Fe$_{3+x}$GeTe$_2$ via chemical doping [9,26], we created a similar superlattice using Fe-rich Fe$_{3+x}$GeTe$_2$ with CrSb to achieve even higher $T_C$. From the AHE measurements, the hysteresis can be distinguished up to 280 K (Fig. 5a), based on which $T_C$ in (Fe$_{3+x}$GeTe$_2$/CrSb)$_n$ is calculated to be 286.7 ± 5.4 K (Arrrott-plots, Supplementary Fig. 13), in good agreement with the $T_C$ of ~280 K determined from ZFC-FC (Fig. 5b). Considering the evolutions of the $T_C$ in the (FGT/CS)$_n$ superlattice, in these (Fe$_{3+x}$GeTe$_2$/CrSb)$_n$ samples, we plotted the $T_C$ as a function of $n$ from $n = 0$ to $n = 3$ in Fig. 5c. Similar to the period-dependent $T_C$ in the (FGT/CS)$_n$ superlattice, with the period increasing, $T_C$ shows a rising trend up to $n = 3$, with ~70 K increase to 286.7 K ± 5.4 K ($n = 3$). To this point, we achieved a $T_C$ of ~286.7 K in 4-layer 2D Fe$_{3+x}$GeTe$_2$ films via the proximity effect.

In summary, we have developed atomically thin 2D ferromagnetic Fe$_3$GeTe$_2$ films on a large scale even down to bilayer by precisely controlling epitaxial growth rate. Combined with the AF CrSb, a parallel ferromagnetic interface interaction between Fe$_3$GeTe$_2$ and CrSb induces an enormous $T_C$ enhancement up to 286.7 K in the superlattice structure from a low-$T_C$ of 140.3 K in the pure 4-layer Fe$_3$GeTe$_2$. Interestingly, the double-switching behavior is observed for the first time in this system as a result of a proximity effect between FGT and Cr layers. In support of these abundant experiments, our DFT calculations found that the interfacial Cr layers retained their interlayer AF coupling but had a net FM magnetic moment, and that doping the spin-polarized electrons via the interfacial Cr layer gives rise to the $T_C$ enhancement of the Fe$_3$GeTe$_2$ films. Our approach of feasible modulation of $T_C$ enabled by the FM/AF proximity effect, together with the capability of wafer-scale growth, provides a realistic platform for spintronic devices based on 2D FMs.

METHODS

Thin-film synthesis

Thin films were grown on mica and (0001) sapphire in a Perkin Elmer 430 MBE system with a base vacuum of 2.5 × 10$^{-9}$ Torr. The growth substrate temperature for Fe$_3$GeTe$_2$ was ~310°C, with the source temperatures of Fe (99.99%), Ge (99.999%) and Te (99.999%) at 1165°C, 1020°C and 285°C, respectively. They were co-evaporated from standard Knudsen cells. CrSb films were grown at the substrate temperature of 280°C, with Cr (99.99%) and Sb (99.999%) cell temperatures of 1180°C and 400°C, respectively. The flux of each element was calibrated by the crystal monitor. The MBE system was equipped with an in situ RHEED.

Thin-film characterizations

Structural characterizations of Fe$_3$GeTe$_2$ and CrSb samples were carried out by X-ray diffraction (Bruker D8 Discover, Bruker Inc., Billerica, MA, USA) and TEM (FEI Tecnai F20) equipped with EDS. Sample composition and doping concentrations were determined by EDS. Cross-section TEM samples were prepared by Focused ion beam (FEI Scios DualBeam).

Electrical and magnetization characterizations

Magneto-transport measurements were performed with the Physical Properties Measurement System by Quantum Design. The magneto-transport devices were confined to the Hall-bar geometry. Experimental data were collected using lock-in amplifiers (Stanford Research 830, Stanford Research Systems, Sunnyvale, CA, USA). The magnetization measurements were taken using
Figure 5. $T_c$ enhancement in the (Fe$_{3+x}$GeTe$_2$/CrSb) superlattice. (a) Temperature-dependent AHE in (Fe$_{3+x}$GeTe$_2$/CrSb)$_3$. Up to 280 K, hysteresis can still be observed. The inset is the perpendicular geometry for the measurement. (b) ZFC-FC curves for (Fe$_{3+x}$GeTe$_2$/CrSb)$_3$. $T_c$ can be roughly determined to be $\sim$280 K, complying with that of 286.7 $\pm$ 5.4 K calculated by the Arrott-plots (Supplementary Fig. 13). The inset is the ZFC-FC curve for the 4-layer Fe$_{3+x}$GeTe$_2$ with $T_c$ at $\sim$220 K. (c) Period-dependent Curie temperature. As the period increases, $T_c$ can be raised from 217.5 $\pm$ 2.6 K ($n$ = 0, the pure Fe$_{3+x}$GeTe$_2$) to 286.7 $\pm$5.4 K ($n$ = 3, the superlattice). The definition of period $n$ = 1 is a bilayer structure of Fe$_{3+x}$GeTe$_2$ and CrSb, the same as that depicted in Fig. 3d inset. The thickness of Fe$_{3+x}$GeTe$_2$ and CrSb is $\sim$3.2 nm and $\sim$1.6 nm, respectively.

DC-Superconducting-Quantum-Interface-Devices (SQUID) by Quantum Design.

X-ray magnetic circular dichroism measurement

XMCD measurements were performed on Beamline 110 at the Diamond Light Source, UK (100% polarized X-rays), and beamline 6.3.1 at the Advanced Light Source, Berkeley, CA (65% polarized X-rays). During the data acquisition, the polarization of X-ray is switched with a fixed magnetic field at every energy point (Beamline 110), and the field direction is switched without changing the polarization at every energy point (Beamline 6.3.1). Such polarization switching at each energy point ensures identical sample conditions for the measurements.

Density functional theory calculations

We performed DFT calculations using the Vienna Ab initio Simulation Package with a plane wave basis set [48]. The ionic potentials including the effect of core electrons are described by the projector augmented wave method, and the GGA was used as the exchange-correlation functional [49]. To better describe the interactions between CrSb and Fe$_3$GeTe$_2$ in the superlattices, the vdW corrections were considered within Grimme’s approach (DFT-D2) [50]. The plane waves with the kinetic energy up to 400 eV were employed to expand the electronic wave functions. Integration over the first Brillouin zone was carried out using the Monkhorst-Pack grid of $7 \times 7 \times 5$ k-point mesh. The structural relaxations were performed till the Hellmann-Feynman force on each atom was smaller than 0.01 eV/Å. Experimental lattice constants were adopted [34,36]. A 24-atom superlattice consisting of $1 \times 1 \times 1$ unit cell of Fe$_3$GeTe$_2$ (12 atoms) and $1 \times 1 \times 3$ lattice of CrSb (12 atoms) was used to study the interfacial interactions. The Coulomb and exchange parameters $U = 3.5$ (3.0) eV and $J = 0.9$ (0.9) eV were chosen for Fe (Cr) 3d electrons [51,52]. Bader charge analysis was used to identify the interfacial Cr-Fe charge transfer [53].

SUPPLEMENTARY DATA

Supplementary data are available at NSR online.

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