Topological Hall effect in bulk ferromagnet \( \text{Cr}_2\text{Te}_3 \) embedded with black-phosphorus-like bismuth nanosheets

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We implement the molecular beam epitaxy method to embed the black-phosphorus-like bismuth nanosheets into the bulk ferromagnet \( \text{Cr}_2\text{Te}_3 \). As a typical surfactant, bismuth lowers the surface tension and mediates the layer-by-layer growth of \( \text{Cr}_2\text{Te}_3 \). Meanwhile, the bismuth atoms precipitate into black-phosphorus-like nanosheets with the lateral size of several tens of nanometers. In \( \text{Cr}_2\text{Te}_3 \) embedded with Bi-nanosheets, we observe simultaneously a large topological Hall effect together with the magnetic susceptibility plateau and magnetoresistivity anomaly. As a control experiment, none of these signals is observed in the pristine \( \text{Cr}_2\text{Te}_3 \) samples. Therefore, the Bi-nanosheets serve as seeds of topological Hall effect induced by non-coplanar magnetic textures planted into \( \text{Cr}_2\text{Te}_3 \). Our experiments demonstrate a new method to generates a large topological Hall effect by planting strong spin-orbit couplings into the traditional ferromagnet, which may have potential applications in spintronics.

Introduction – When it flows in the presence of a static magnetic field in the perpendicular direction, the electron is deflected by the Lorentz force and gives rise to the Hall effect identified by a transverse resistivity [1]. The external magnetic field is not mandatory, however, ‘Berry curvature’ is essential to produce a Hall effect [2]. The spin-orbit coupling in the magnetic system generates the anomalous Hall effect (AHE) associated with ‘THE materials’, it is of significant interests to efficiently insert strong spin-orbit couplings into traditional ferromagnets, which is still challenging so far. Among many typical ferromagnets, the study of chromium tellurides has a long history and can be traced back to 1935 [26]. Depending on the stoichiometric ratio, \( \text{Cr}_{1-x}\text{Te} \) has a rich structural phase diagram with Curie temperatures ranging from 170 K to 340 K [26]. Very recently, the \( \text{Cr}_2\text{Te}_3 \) thin film with a high-quality single crystalline structure has been synthesized using the molecular beam epitaxy (MBE) technique, which shows a strong perpendicular magnetic anisotropy and a large magnetic moment per Cr atom about 2.8 \( \mu_B \) [27, 28]. If strong spin-orbit coupling elements can be doped in an appropriate way, \( \text{Cr}_2\text{Te}_3 \) would exhibit significant THE with the onset temperature close to the Curie temperature. A convenient candidate is bismuth, which is well known not only for its strong spin-orbit coupling, but also as a typical surfactant to achieve the layer-by-layer growth mode in MBE [29, 30]. Furthermore, bismuth has various atom-thick layer structures, such as bismuthene [31] and the very unusual puckered-layer structure [32, 33] similar to black phosphorous [34].

In this Letter, we report a new efficient route towards producing “THE materials” by embedding black-phosphorus-like bismuth nanosheets into the bulk ferromagnet \( \text{Cr}_2\text{Te}_3 \). We reveal a large, high-temperature topological Hall effect together with magnetic susceptibility plateau and magnetoresistivity anomaly. As a con-
FIG. 1. HAADF STEM images of the MBE-grown Bi-embedded Cr$_2$Te$_3$. (a) Cross-sectional HAADF STEM image of a sliced MBE-grown Cr$_2$Te$_3$ sample. The visible white filaments inside the Cr$_2$Te$_3$ layer are bismuth nanosheets. (b) Landscape STEM image of a Cr$_2$Te$_3$ sample. (c) High resolution STEM image of crystalline Cr$_2$Te$_3$. Note that the Cr atoms are invisible in the STEM image. (d) and (e) STEM image and EDS mappings for the distribution of Bi element. (f) STEM image with enlarged scale showing a bilayer bismuth sandwiched between layers of Cr$_2$Te$_3$. (g) Atomic configuration (side view) of a bismuth nanosheet embedded Cr$_2$Te$_3$ structure. (h) High resolution STEM image for several Bi nanosheets embedded in the Cr$_2$Te$_3$ layer.

trol experiment, none of these signals is observed in the pristine Cr$_2$Te$_3$ samples, revealing the critical role of Bi-nanosheets with strong spin-orbit coupling. Therefore, the Bi-nanosheets serve as seeds of spin-orbit couplings planted into Cr$_2$Te$_3$ to generate THE. We also discuss the magnetic skyrmion scenario accounting for our experimental results.

Sample growth and characterizations – The growth of Bi-intercalated Cr$_2$Te$_3$ thin films were performed on semi-insulating epi-ready GaAs(111)B substrates in a home-built molecular beam epitaxy system with a base vacuum of MBE better than $5 \times 10^{-10}$ torr. Prior to the growth of Cr$_2$Te$_3$, the GaAs(111)B substrate was first deoxidized at 580 °C until streaky RHEED patterns appeared, followed by a deposition of 150 nm ZnSe buffer layer for smoothening the voids of GaAs induced by the deoxidation. Cr$_2$Te$_3$ thin film were grown by co-evaporating Bi (99.995%), Cr (99.999%), and Te (99.999%) from Knudsen cells with a flux ratio of 2:1:10 at $T_{\text{substrate}} = 260$ °C. A spherical aberration corrected scanning transmission electron microscopy (STEM) were employed for performing systematic structure and chemical analyses.

The Cr$_2$Te$_3$ thin films were grown on the GaAs(111) substrate with a buffer layer of ZnSe, as shown in the cross-sectional high angle annular dark field (HAADF) STEM image in Fig. 1 (a). During the growing process a concurrent bismuth flux was applied together with Cr and Te fluxes. The participation of bismuth atoms can lower the surface tensions resulting in a surfactant-mediated layer-by-layer growth mode instead of a three-dimensional cluster mode [29, 30]. In fact, we observed the extended streaky reflection high-energy electron diffraction (RHEED) patterns during the entire growing process. The X-ray diffraction measurement also shows that Bi-embedded samples have much better crystalline quality than non-Bi-embedded ones [35].

Figure. 1 (b) shows the landscape STEM image of a Cr$_2$Te$_3$ thin film embedded with Bi nanosheets, where a single crystalline structure is found with a sharp in-
The high-resolution STEM image of Cr$_2$Te$_3$ lattice in Fig. 1(c) shows a typical NiAs-type lattice structure of the pristine Cr$_2$Te$_3$ oriented along [001] direction. The lattice constants of Cr$_2$Te$_3$ are determined to be $a = b = 6.81$ Å and $c = 12.28$ Å. The atom distributions of a Bi nanosheet are revealed in Fig. 1(d) and (e) by the atomically resolved energy dispersive X-ray spectroscopy (EDS) mapping, showing that the Bi atoms occupy Te sites to form a bilayer structure embedded in the Cr$_2$Te$_3$ lattice. The bismuth atoms precipitate into nanosheets with lateral size of several tens of nanometers, corresponding to the luminescent line segments in Fig. 1(a) and (b). The atomically resolved STEM image given in Fig. 1(f) further reveals that the bismuth nanosheet owns a bilayer structure with lattice constants $a = 4.5$ Å and $c = 6.5$ Å, in a striking analogy to the puckered-layer structure of black phosphorus [33]. Fig. 1(g) is the atomic configuration of a Bi-nanosheet embedding in the Cr$_2$Te$_3$ as shown in the high resolution STEM image in Fig. 1(h). The embedded bilayer bismuth exhibits a slight distortion due to the interaction with Cr$_2$Te$_3$.

**Topological Hall effect** – Then Bi-embedded Cr$_2$Te$_3$ samples hybridize strong spin-orbit couplings and ferromagnetism, and we observe a significant THE in the magneto-transport experiments. Fig. 2 is the THE result in a 60-nm-thick sample of Cr$_2$Te$_3$ embedded with Bi nanosheets. The Curie temperature ($T_c$) of the sample is determined to be 180 K from the temperature dependence of resistivity and magnetization [35]. Fig. 2(a) shows the double-sweep measurements of the Hall resistivity $\rho_{yx}(H)$ at various temperatures with the magnetic field $H$ along the [001] direction. Above $T_c$, the Hall resistivity is linearly dependent on $H$, namely, $\rho_{OHE} = R_o H$ as expected for the ordinary Hall effect (OHE). Since the ordinary Hall coefficient $R_o > 0$ as shown in Fig. 2(a), the charge carrier is of $p$-type and the density can also be estimated to be around $10^{21}$ cm$^{-3}$ [35], coming from the unoccupied Cr-3$d$ orbitals. Below $T_c$, a clear hysteresis behavior is observed, indicative of the anomalous Hall effect (AHE). The anomalous Hall resistivity depends on the magnetization $M$ and is generally written as $\rho_{AHE} = R_a M$. There are usually three independent origins of the AHE, including the skew-scattering, side-jump and the Berry curvature in momentum space [2], which may have opposite contributions, thus possibly cancel with one another leading to a vanishing anomalous Hall coefficient $R_a$ [25]. Indeed, we find the sign of $R_a$ is reversed at $T \sim 42$ K with decreasing temperature, and no anomalous Hall resistivity is detected right at this temperature.

The most striking feature of the Hall resistivity curves is the emergence of abnormal peaks in the vicinity of the coercive field ($H_c$), as shown in Fig. 2(a). These peaks show up regardless of the sign of $R_a$, even at $T = 42$ K where the AHE vanishes completely, indicating that these abnormal Hall resistivity peaks should have a different origin from the AHE. In fact these pronounced peaks can be attributed to the THE, which is related to the spin chirality of magnetic skyrmions [3-7]. Similar THE signal
was also observed in 45-nm-thick Bi-embedded Cr$_2$Te$_3$ samples \[35\]. The topological Hall resistivity can be extracted from the total Hall resistivity $\rho_{yx}$ in the form $\rho_{\text{THE}} = \rho_{yx} - R_o H - R_T M$ \[35\]. Fig. 2(b) shows the $H$-dependence of $\rho_{\text{AHE}}$ and $\rho_{\text{THE}}$ at $T = 35$ K, where we also plot $\rho'_{yx} = \rho_{yx} - R_o H$ ($= \rho_{\text{THE}} + \rho_{\text{AHE}}$) for comparison. Obviously, $\rho_{\text{AHE}}(H)$ and $\rho'_{yx}(H)$ almost coincide with each other, except in the region near the coercive field where the THE appears. The resultant $\rho_{\text{THE}}$ and $\rho_{\text{AHE}}$ at 85 K are plotted in Fig. 2(c). Fig. 2(d) is a phase diagram illustrating the strength of THE signals. Obviously, the THE occurs only in the vicinity of the coercive field, implying that the appearance of the non-zero spin chirality is associated with the magnetization reversal. It is worth noting that the maximum $\rho_{\text{THE}}$ in our sample of Bi-embedded Cr$_2$Te$_3$ is around 1300 nΩ·cm, which is larger than that of SrRuO$_3$/SrIrO$_3$ \[3\] and one order of magnitude larger than that of the B20 chiral magnet (e.g., MnSi) \[8\].

**Magnetic susceptibility plateau and magneto-resistivity anomaly** — Fig. 3 shows the temperature dependence of magnetization $M$, magnetic susceptibility $\chi(H) = \partial M(H)/\partial H$, and magneto-resistivity $\rho_{xx}$, respectively. Right in the region near the coercive field, the $M(H)$ curve shows an abnormal magnetization tail, which corresponds to a striking susceptibility plateau in the $\chi(H)$ curve, as indicated by arrows in Fig. 3(a) and (b). Furthermore, as evident in Fig. 3(c), the $\rho_{xx}(H)$ curve also exhibits a hump feature near $H_c$, indicative of a new scattering source for electronic transport. All these abnormal behaviors persist up to 115 K, similar to the THE shown in Fig. 2(c). In the field region where THE appears, the $M(H)$ curve exhibits a tail, indicating the emergence of nontrivial spin texture. The coincidence of the active regions of THE, magnetization tail, susceptibility plateau, and magneto-resistivity hump strongly suggests the same physical origin of them.

**Discussions and conclusions** — As a well established theory \[3\]-\[7\], the magnetic skyrmion can induce the THE owing to its nonvanishing spin chirality. Skyrmions can be identified directly by mapping the spin textures using small angle neutron scattering \[16\], Lorentz transmission electron microscopy \[36\], spin-resolved scanning tunneling microscopy \[37\], resonant x-ray scattering \[38\], or even magneto-optics \[39\]. However, the direct mapping of skyrmions is beyond the scope of this work, and is left to future investigations. We have checked our experimental results very carefully, and found that all our experimental details can be explained in terms of magnetic skyrmion scenario. Furthermore, the skyrmion is also supported in the numerical simulations with the input parameters derived from the first-principles of our system \[10\].

In Cr$_2$Te$_3$ with Bi nanosheet embedded, the magnetization saturates with all the local moments fully polarized in the high fields $H > H_c$, hence the scalar spin chirality $\hat{S}_i \cdot (\hat{S}_j \times \hat{S}_k)$ is zero and no THE occurs. Once the field reverses and exceeds the coercive field $H_c$, the moments in the bulk Cr$_2$Te$_3$ are flipped first, and those near the Bi-nanosheets are not completely flipped yet, but only twisted by the DM interactions (resulting from the hybridization between Bi and Cr orbitals), leading to the formation of magnetic skyrmions in the vicinity of the coercive field. The nonzero spin chiralities of these
skyrmions in turn give rise to the THE. This scenario is consistent with the measurements of magnetization and longitudinal magnetoresistivity as shown momentarily. Skyrmions display the topological paramagnetism \[^{[10]}\], \textit{i.e.}, the susceptibility plateaux and magnetization tails, which is related to the topological stability of the magnetic skyrmions and due to the slowing down of the flipping speed of skyrmion moments near \(H_c\) \[^{[11]}\]. Furthermore, the magnetic texture of skyrmions is also a new scattering source accounting for the magnetoresistivity hump near \(H_c\) in Fig.\[3\](c) \[^{[11]}\].

To the best of our knowledge, it is the first time in the literature to observe all these skyrmion-induced features, THE, topological paramagnetism and magnetoresistivity anomaly, simultaneously in the same system. Therefore, it is very likely that our system hosts the magnetic skyrmions in the THE region. Detailed information of the magnetic skyrmions in Bi-embedded \(\text{Cr}_2\text{Te}_3\) can be also derived from our experimental data \[^{[35]}\]. We estimate the skyrmion size is about 14 nm, comparable with the lateral sizes of Bi nanosheets which may vary from 10 nm to 100 nm (see Fig.\[1\](b)). Therefore, by controlling the diameter of the Bi-nanosheets, a single skyrmion can be obtained and its size may be further reduced in smaller Bi-nanomaterials.

In conclusion, we have grown the bulk ferromagnet \(\text{Cr}_2\text{Te}_3\) with the black-phosphorous-like bismuth nanosheet embedded. It is an efficient experimental approach to insert strong spin-orbit couplings into the traditional ferromagnet. The nanosheet serves as a ‘seed’ for the strong spin-orbit coupling planted into the bulk ferromagnet, and generates the non-trivial magnetic texture, signaled by a large topological Hall effect, topological paramagnetism and magnetoresistivity anomaly. This method can be applied to other bulk ferromagnetic systems, largely expanding the family of topological-Hall-effect materials.

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