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Unusual magnetic and transport properties in math xmlns="http://www.w3.org/1998/Math/MathML">mrow>ms ub>mi>HoMn/mi>mn>6/mn>/msub>msub>mi>Sn/mi>m n>6/mn>/msub>/mrow>/math> kagome magnet

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Unusual magnetic and transport properties in HoMn₆Sn₆ kagome magnet

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With intricate lattice structures, kagome materials are an excellent platform to study various fascinating topological quantum states. In particular, kagome materials, revealing large responses to external stimuli such as pressure or magnetic field, are subject to special investigation. Here, we study the kagome-net $HoMn_6Sn_6$ magnet that undergoes paramagnetic to ferrimagnetic transition (below 376 K) and reveals spin-reorientation transition below 200 K. In this compound, we observe the topological Hall effect and substantial contribution of anomalous Hall effect above 100 K. We unveil the pressure effects on magnetic ordering at a low magnetic field from the pressure tunable magnetization measurement. By utilizing high-resolution angle-resolved photoemission spectroscopy, Dirac-like dispersion at the high-symmetry point K is revealed in the vicinity of the Fermi level, which is well supported by the first-principles calculations. Our investigation will pave the way to understand the magneto-transport and electronic properties of various rare-earth-based kagome magnets.

I. INTRODUCTION

Topological nontrivial magnetic materials have attracted tremendous attention, and kagome magnets are one of them, which reveal various interesting electronic states such as Dirac fermions [1, 2], intrinsic Chern quantum phases and spin-liquid phases [3–8]. Among these kagome materials, transition-metal-based kagome magnets [1, 2, 9–16] have already appeared to be the distinguished candidates for correlated topological states, as they possess both unusual magnetic tunability and large Berry curvature fields. These materials are also predicted to support intrinsic Chern quantum phases [17, 18] due to their extraordinary lattice geometry and broken timereversal symmetry. In addition, a number of nontrivial magnetic phases have been observed in the rare-earthand-transition-metal-based RMn_6Sn_6 family (R = rare earth) [19–25]. Recently, TbMn₆Sn₆, one of the members of this 166 family, has been identified as a Chern magnet where large anomalous Hall effect (AHE) and the quantized Landau fan diagram featuring spin-polarized Dirac dispersion with a large Chern gap have been observed [7]. In another member of this family, YMn₆Sn₆, a topological Hall effect has been revealed [26–28] and explained by a new chirality mechanism originated from frustrated interplanar exchange interactions and the induced strong magnetic fluctuations. Lately, Gao et al. have reported anomalous Hall effect in RMn_6Sn_6 (R = Tb, Dy, Ho) with clean Mn kagome lattice [29]. Dhakal et al. have recently studied magnetic, magneto-transport, and angle resolved spectroscopic measurement of ErMn₆Sn₆ [30]. Thus, each member of the RMn₆Sn₆ family is catching extensive attention for experimental exploration.

In this article, we explore another member of the ${\rm RMn_6Sn_6}$ family, ${\rm HoMn_6Sn_6}$, which consists of two mag-

netic sublattices: Ho and Mn. Neutron diffraction studies and magnetic measurements [31–34] suggested that Mn and Ho sublattices simultaneously transfer from paramagnetic state to ordered states below a critical temperature of $T_{\rm C}=376$ K. With further lowering of temperature, the easy direction of the ordered state starts to reorient from the basal plane toward the c axis below 200 K [32]. The canting angle with respect to [001] is~49° at 100 K and remains constant down to 2 K [32]. The mechanism of the spin-reorientation (SR) transition of $HoMn_6Sn_6$ has been quantitatively investigated in a molecular field theory [31]. However, detailed magnetic, transport, and angle-resolved spectroscopic measurements along with theoretical investigation of $HoMn_6Sn_6$ have not been reported yet.

Hence, we have performed systematic studies of transport and magnetic behaviors (at ambient and applied pressure) of HoMn₆Sn₆ kagome magnet. By utilizing angle-resolved photoemission spectroscopy (ARPES), we have measured the electronic structure of this compound, which has been supported by first-principles calculations. The electrical transport measurement indicates that the compound is metallic and shows a large anomalous Hall effect and topological Hall effect contribution in HoMn₆Sn₆ at temperatures close to the SR transition temperature, T_{SR} . Furthermore, we measure the magnetizations with external fields applied along the in- and out-of-plane directions when the temperature decreases from 400 K to 2 K. At T=2 K, a well-defined magnetization loop for the field applied along the c-axis suggests a strong out-of-plane magnetization component. Thus, this compound could be a potential candidate for Cherngapped topological material, according to the Haldane model [17, 18, 35–37]. Moreover, magnetization mea-

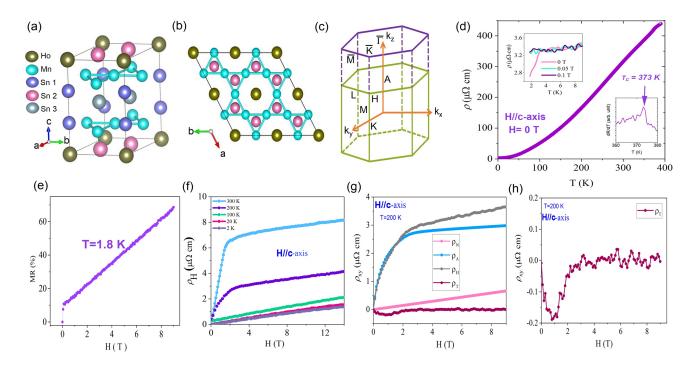


FIG. 1. Crystal structure and sample characterization of $HoMn_6Sn_6$: (a) Crystal structure of $HoMn_6Sn_6$. (b) Top view of crystal structure of $HoMn_6Sn_6$ forming the kagome lattice. (c) Three dimensional (3D) bulk Brillouin zone (BZ) of the crystal with its projection on the [001] surface. High symmetry points are marked on the plot. (d) Temperature variation of the electrical resistivity of $HoMn_6Sn_6$ in zero external magnetic field. The upper inset to the left shows the resistivity at low temperature and magnetic field, while the lower inset to the right indicates the ferrimagnetic transition at T_C =373 K. (e) Magnetoresistance versus magnetic field measured at T =1.8 K. (f) Hall resistivity of $HoMn_6Sn_6$ measured at different temperatures along $H \parallel c$. (g) Magnetic field dependence of total Hall resistivity together with the three different components, ρ^A , ρ^N , and ρ^T (see text for more details), measured with $H \parallel c$, and at T = 200 K. (h) Magnetic field dependence of topological Hall resistivity along $H \parallel c$ (magnified view).

surements under pressure reveal the pressure effects on magnetic ordering at low magnetic fields and below $T_{\rm SR}$ (200 K). ARPES measurements demonstrate the presence of Dirac-like states at the high symmetry point K, close to the Fermi level $E_{\rm F}$. Our exploration of electronic and magnetic properties of HoMn₆Sn₆ will provide an effective way to reveal the magneto-transport behaviors of various rare-earth kagome magnets.

II. METHODS

Single crystals of $HoMn_6Sn_6$ were grown by the Sn flux technique as described in the supplementary materials (SF.1) [38]. DC magnetization and transport measurements were carried out using the physical property measurement system (PPMS). Resistivity and Hall resistivity measurements were performed using the conventional 4-probe method. The resistivity was measured by applying the current (I) along the ab-plane and the field was applied along the c-axis. A large residual resistivity ratio, RRR = 68, indicates a high quality of the single crystals used in this study.

DFT calculations are performed using a full-potential

linear augmented plane wave (FP-LAPW) method, as implemented in WIEN2k [39]. The generalized gradient approximation of Perdew, Burke, and Ernzerhof [40] is used for the correlation and exchange potentials. To generate the self-consistent potential and charge, we employed $R_{\rm MT}K_{\rm max}=8.0$ with muffin-tin (MT) radii $R_{\rm MT}$ = 2.7, 2.4 and 2.5 a.u., for Ho, Mn, and Sn, respectively. The calculations are performed with 264 k-points in the irreducible Brillouin zone (IBZ) and iterated until the total energy difference is lower than 0.01 mRy. Spin-orbit coupling (SOC) is included with the secondvariational method. Besides using the DFT+U method, we also treated the 4f states in an open-core approach to keep the band structures near Fermi level from the influence of the 4f states. The primitive cell contains one formula unit (f.u.). We adopt experimental lattice parameters [33] in calculations.

We construct the TB Hamiltonian by using 118 maximally-localized Wannier functions (MLWFs) [41], 59 for each spin channel, corresponding to d-type orbitals for Ho and Mn atoms, and s- and p-type orbitals for Sn atoms in the unit cell. SOC mixes the spin-up and

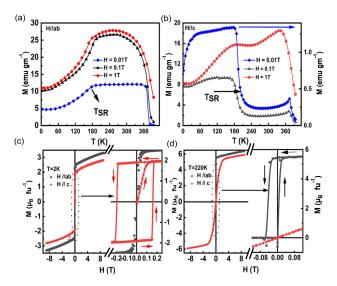


FIG. 2. Magnetic properties of $\operatorname{HoMn_6Sn_6}$ single crystals measured along $H \parallel ab$ and $H \parallel c$ (in and out of plane, respectively): Magnetization as a function of temperature, M(T), measured along (a) $H \parallel ab$ and (b) $H \parallel c$, respectively, at various magnetic fields. Magnetization as a function of magnetic field, M(H), measured along $H \parallel ab$ and $H \parallel c$ at (c) T=2 K and (d) 220 K, respectively. Magnetization at low magnetic field of M(H) for both directions are also shown in the right sides of figures (c) and (d), respectively, for the clarity.

spin-down states. The resulting real-space Hamiltonians $H(\mathbf{R})$ with a dimension of 118×118 accurately represent the band structures within the energy window of interest around E_{F} .

The electronic structure of $HoMn_6Sn_6$ was measured by ARPES at the SLS SIS-X09LA BL at 20 K with a photon energy of 100 eV and under ultra-high vacuum conditions (5×10^{-11} Torr). High quality samples were used for the measurements. The crystals were cut into small pieces and mounted on the sample plate with copper posts on top of them. After that the samples were loaded into the chamber, the chamber was cooled and pumped down for few hours. The samples were cleaved in situ. For the synchrotron measurements, the energy resolution was better than 20 meV and the angular resolution was set to be finer than 0.2° . The measurements were carried out for more than 15 hrs without noticeable decay or damage of the samples.

III. RESULTS

A. Crystal structure and electrical transport measurement

HoMn₆Sn₆ crystallizes into a HfFe₆Ge₆-type structure (space group is P6/mmm, (No. 191)), as shown in Fig. 1(a) with Ho at 1(a) (0, 0, 0), Mn at 6(i) (1/2, 0, z \sim 0.249), Sn at 2(c) (1/3, 2/3, 0), 2(d) (1/3, 2/3, 1/2) and 2(e) (0, 0, z \sim 0.34) [42], which is composed of Ho

layer consisting of Sn atoms and Mn kagome nets, stacked in the sequence -Mn-Ho-Mn-Ho-Mn- along the c-axis [31]. The Ho and Sn₂ atoms lie in the same plane and $Mn-Sn_1-Sn_3-Sn_1-Mn$ atoms are stacked along the c-axis alternatively. The Mn atoms form two kagome layers and the Sn_2 and Sn_3 atoms form a hexagonal structure. The hexagonal structure formed by Sn₂ atoms can be clearly observed from Fig. 1(b), while viewing along the c-axis. The Ho atoms lie at the centre of the hexagons formed by the Sn_2 atoms, clearly visible from Fig. 1(b). Since Sn_3 is below the Sn₂ layers, hence Sn₃ layer is invisible from this top view in Fig, 1(b). The Ho atom lies at the centre of the hexagons surrounded by the Sn_2 atoms. On the other hand, being positioned below or above the Ho atoms, Sn₁ atoms could not be detected from this top view of Fig. 1(b). Figure 1(c) shows the three-dimensional (3D) BZ and its hexagonal shaped projection onto the (001) plane.

The temperature-dependent resistivity measured along $H \parallel c$, as plotted in Fig. 1(d), shows the metallic behavior of the sample over the measured temperature range of 1.8 K to 400 K. The spin reorientation transition at ~ 200 K is not visible in $\rho(T)$ in Fig. 1(d). However, the ferromagnetic transition of Mn sublattice at $T_{\rm C}$ =373 K has been observed, clearly seen in dR/dT(T) (see the inset at the right corner of the Fig. 1(d)). The inset at upper left corner shows an anomaly at low temperature (T=3 K) resistivity at zero magnetic field. The origin of the anomaly is unclear, but it can be suppressed by the application of a magnetic field as weak as 0.05 T. Figure 1(e) presents the magnetoresistance (MR(H)) of $HoMn_6Sn_6$, showing a relatively large value of MR=70%at $\Delta H = 9$ T. Interestingly, the MR at 2K shows a linear behavior as a function of the magnetic field, suggesting the materials hosting Dirac-like states. Figure 1(f) presents the magnetic-field dependence of the Hall resistivity in HoMn₆Sn₆. As can be seen from the figure, there is a strong contribution of anomalous Hall effect for temperatures above 100 K, and the overall behavior of Hall resistivity resembles the M(H) data shown in Fig. 2(d).

In topological magnetic materials, the total Hall resistivity (ρ_H) can be expressed as:

$$\rho_H = \rho^A + \rho^N + \rho^T \tag{1}$$

where, $\rho^A = R_S 4\pi M$ is the anomalous Hall resistivity, $\rho^N(H) = R_0 H$ is the normal Hall resistivity, and ρ^T refers to the topological Hall resistivity. Eq. (1) can be rewritten as $\rho_H = R_0 H + R_S 4\pi M$, in the high field saturation region. We calculate the slope R_0 and intercept $4\pi R_S$ from the linear plot of ρ_H/M versus H/M in the high-magnetic field region. At that region, the ideal linear behavior of the ρ_H/M versus H/M indicates the anomalous Hall resistivity to be the main component of the total Hall resistance. The R_0 derived for $H \parallel c$ and at T=200 K gives positive ρ^N (see figure 1(g)). We derive the topological Hall resistivity by subtracting the normal and anomalous component of the Hall resistivity from the total Hall resistivity.

As can be clearly seen from the magnified view of the

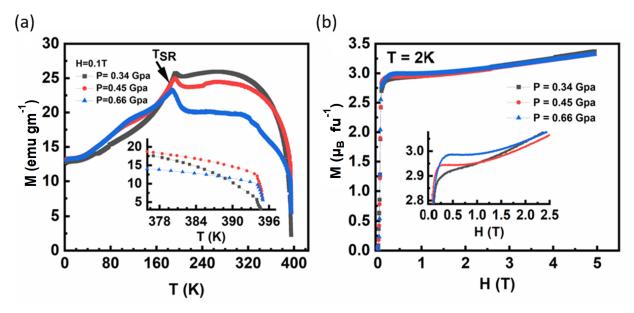


FIG. 3. Pressure-induced magnetization measurements of HoMn_6Sn_6 single crystal measured along the $H \parallel c$. (a) Magnetization as a function of temperature, M(T), measured at various hydrostatic pressure at H = 0.1 T. The inset indicates the magnified view of M(T) near $T_C = 376$ K. (b) Magnetization as a function of magnetic field at various hydrostatic pressure at T = 2K.

topological Hall resistivity (figure 1(h)), the contribution of the topological Hall effect is small compared to anomalous Hall resistivity, but its value is comparable to other topological materials [28, 43]. The magnitude of topological Hall resistivity is maximum ($\sim 0.2~\mu\Omega$ -cm) at a relatively low magnetic field of ~ 1 T, temperature T = 200 K and decreases with further increase in the field and has negligible effect for H ≥ 2.2 T. Similar behavior has been reported in the YMn₆Sn₆ kagome magnet in which the topological Hall effect ρ_{xy}^T reaches $\sim 0.28~\mu\Omega$ -cm at H = 4 T and T = 220K and decreases with further increase in a magnetic field. The change of the topological Hall effect in YMn₆Sn₆ is ascribed to the magnetic structure at the higher magnetic field [43].

B. Magnetic properties and pressure induced magnetization

Figures 2(a) and 2(b) show the iso-field magnetization M(T) measured with different in- and out-of-plane magnetic fields, exhibiting two distinct transitions for both directions. Figure 2(a) presents the M(T) measured at three representative in-plane magnetic fields: $H=0.01,\ 0.1,\ {\rm and}\ 1\ {\rm T.}$ At $H=0.01\ {\rm T}$, the magnetization increases smoothly with temperature until $T\leq 190\ {\rm K}$, becomes plateaued between 200 K to 360 K, finally undergoes phase transition at 374 K (obtained from minima of ${\rm d}M/{\rm d}T$). The high-temperature phase transition at H=0.01 T is close to the previously reported ferrimagnetic-to-paramagnetic-transition temperature of $T_{\rm C}=376\ {\rm K}$ for a polycrystalline sample by Venturini et al. [31].

The obtained transition temperature at T=190 K corresponds to the spin-reorientation temperature, $T_{\rm SR}$, which compares well with the previously reported value of $T_{\rm SR}$ = 200 K, obtained from neutron diffraction. Interestingly, applying external magnetic fields slightly decreases $T_{\rm SR}$ but significantly increases $T_{\rm C}$. ferrimagnetic-to-paramagnetic transition does not complete until 400 K at H > 1 T, which is also shown in the non-linear M(H) measured at T = 390 K (supplementary figure, SF. 2) [38]. It is also noticeable that, along $H \parallel ab$, at H = 1 T, the low-temperature $(T \leq T_{SR})$ magnetization is larger compared to the measurement along $H \parallel c$, and the magnetization difference decreases at $T \geq T_{SR}$. The typical isothermal magnetization for both $H \parallel ab$ and $H \parallel c$ at T = 2 K and 220 K (below and above the T_{SR}) are shown in Figs. 2(c) and 2(d), respectively. At T=20 K and H=9 T, the magnetic moments are 3.14 $\mu_{\rm B}/{\rm f.u.}$ and 2.8 $\mu_{\rm B}/{\rm f.u.}$ for $H \parallel ab$ and $H \parallel c$, respectively (see SF. 2). At T=2 K, the saturation magnetic moments are $M_{\rm S}=3.8$ and $3.2~\mu_{\rm B}/{\rm f.u.}$ for $H \parallel ab$ and $H \parallel c$, respectively, obtained from the extrapolation of M vs. H^{-1} curves. The value of $M_{\rm S}$ is $3.2 \mu_{\rm B}/{\rm f.u.}$, along $H \parallel c$, which is close to $3.26 \mu_{\rm B}/{\rm f.u.}$, found by Clatterbuck et al. [44]. The observed saturation magnetic moment values are significantly lower than the saturation moment of Ho³⁺ ion, which is expected for the ferrimagnetic arrangement of Mn and Ho atoms. The calculated magnetization of the ferrimagnetic configuration is $3.07\mu_{\rm B}/{\rm f.u.}$ in DFT+U, agreeing reasonably well with experiments. The M(H) measured along $H \parallel c$ at T =2 K, shows a square shaped hysteresis loop with a coercivity of 0.18 T, while the M(H) measured with in-plane fields is non-hysteretic. The hysteretic behaviors for two

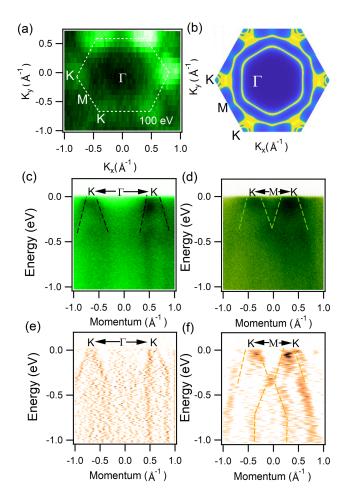


FIG. 4. Fermi surface and dispersion maps of $HoMn_6Sn_6$. (a) Experimentally measured Fermi surface map. (b) Corresponding calculated Fermi surface map. ARPES measured dispersion maps along the (c) K-Γ-K and (d) the K-M-K directions. Second derivative plots along (e) the K-Γ-K and (f) the K-M-K directions.

field directions reverse above $T_{\rm SR}$, suggesting the change of the easy-cone anisotropy at $T < T_{\rm SR}$ to the easy-plane anisotropy at $T > T_{\rm SR}$.

The magnetic properties of HoMn₆Sn₆ were further studied under applied pressure using a Cu-Be mechanical cell with an inner diameter of 2.6 mm, and the lead was used as an internal manometer. The measurements were carried out on the same piece of the sample that has been used for magnetization measurement at ambient pressure (Fig. 2). The pressure calibration from the manometer is presented in SF. 3 [38]. Figure 3 shows magnetization as a function of temperature and magnetic field at pressure up to 0.66 GPa. The spin-reorientation transition is more pronounced with a peak around 190 K, and the ferrimagnetic-to-paramagnetic transition of Mn sublattice is much more sensitive with pressure. The transition temperature shift from 373 K at ambient pressure (Fig. 2(b)) to 394 K at P = 0.34 GPa, with H = 0.1 T (Fig. 3(a)) and the transition is not complete until 395 K

for P>0.34 GPa. The inset of Fig. 3(a) indicates the magnified view of M(T) near $T_{\rm C}=376$ K, which clearly shows the shift of $T_{\rm C}$ for various applied pressure. As shown in the figure, it seems that the critical transition temperature is much higher than 400 K at P>0.34 GPa, however, we do not observe that within our measurement range of 400 K. Considering the value of shift of $T_{\rm C}$ at P=0.34 GPa, the $T_{\rm C}$ shifts to a higher temperature at 53 K/GPa, however, $T_{\rm C}$ does not necessarily increase linearly with the pressure, therefore, a detailed M(T) study at pressure and T>400 K is required.

Even if we could not check to get direct experimental evidence, the hydrostatic pressure can likely compress the lattice anisotropically and result in a lowering of the interlayer and interatomic distances between Ho atoms on the 001 plane [Wyckoff site 1a, in (0, 0, 0)] and Mn atoms on planes 004 [in site 6i $(1/2, 0, \sim 1/4)$], which may help to enhance the saturation moment. One may also argue that the pressure introduces anisotropic lattice change, which changes the crystal field and the easy direction of Ho. A decrease of easy-direction angle from 49° could give a larger z-component magnetization and, therefore, an enhancement of moment.

C. Electronic structure of HoMn₆Sn₆

Figure 4 presents the electronic structure of HoMn₆Sn₆ measured by ARPES and first-principles calculations. We show the Fermi surface (FS) map measured at a temperature of 20 K. The FS map shows the hexagonal symmetry as suggested by the crystal structure. Furthermore, the FS map exhibits the metallic nature, which is accordant with the transport measurements. Six small circles are observed at the BZ corners K, which display a good hexagonal shape (white dotted hexagon). These intense circles at the high symmetry points K possibly denote multiple bulk bands and the complex band structure of this material. However, all features are not visible in the photoemission intensity plots presented in Fig. 4(a). Figure 4(b) shows the calculated FS, which is in very good match with Fig. 4(a) and reveals the hexagonal shape of the FS map as well.

Next, we discuss the dispersion maps along the high symmetry directions. Figure 4(c) presents the dispersion map, in which linear Dirac-like dispersive bands exist along the K- Γ -K path. The 2nd derivative plot

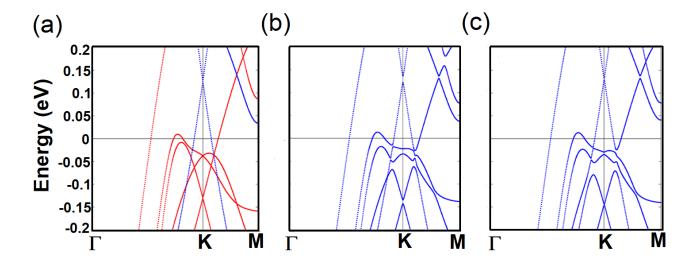


FIG. 5. Band structures of HoMn₆Sn₆ along the Γ -K-M path near $E_{\rm F}$ calculated without and with SOC. The bands are calculated at $k_z = 0.25$ r.l.u. (a) Scalar relativistic bands calculated without SOC. Blue and red lines denote two spin channels. (b, c) SOC bands calculated with the spin-quantization direction along the (b) [001] and (c) [100] directions, respectively.

of Fig. 4(c) shows the clean Dirac-like dispersion map crossing the Fermi level (Fig. 4(e)). Figure 4(d) presents the dispersion map along the K-M-K direction, where electron-like bands exist close to the Fermi level. Corresponding 2nd derivative plot of Fig 4(d) along the K-M-K direction shows some additional bands at M high symmetry point (Fig. 4(f)), where the saddle point of the kagome magnet might exist. Dispersion maps along the K- Γ -K direction at various photon energies are presented in supplementary section 4 (SF. 4). Besides, the dispersion map and corresponding momentum distribution curve (MDC) and energy distribution curve (EDC) along the K- Γ -K path are shown in supplementary section 5 (SF. 5) [38]. Although HoMn₆Sn₆ has been suggested to be a Chern magnet [15], the possible Chern gap above the Fermi level could not be accessed via our ARPES measurement, therefore, further study is required to support this claim.

Finally, we discuss the Dirac crossings and SOC-induced gaps and their dependence on the spin orientations. Figure 5 shows the band structures along the Γ -K-M path at $k_z = 0.25$ r.l.u., calculated with and without SOC. This particular k_z value is chosen, according to the photon energy used in ARPES, to better compare with experiments. As shown in Fig. 5(a), multiple Dirac-like crossings occur near $E_{\rm F}$ at the BZ corners K. Depending on their band characters, different crossings have different k_z dependence. For example, the crossing at ~ 0.12 eV below $E_{\rm F}$ is much less k_z -dependent than the one right above $E_{\rm F}$, showing a more 2D-like band character.

Figure 5(b) shows the SOC band structures calculated with the spin-quantization direction along the out-of-plane direction. As expected, SOC lifts the orbital degeneracy and splits all crossings at K with various gap sizes. The gaps' sizes depend on the orbital characters

of corresponding bands and the SOC Hamiltonian $H_{\rm SO}$; the latter depends on the spin direction. Figure 5(c) shows the SOC band structures calculated with the spin-quantization axis lying in the basal plane. The SOC-induced gaps become negligibly small or eliminated, in comparison to Fig. 5(b), showing the possibility to control the SOC gap size by changing the spin direction. Here, we have shown two extreme cases to illustrate the gap evolution with angle change. Note that, in low temperature, the experimental magnetic ordering direction of HoMn₆Sn₆ is tilted to 49° with respect to [001], resulting in SOC-induced gaps' sizes in between the two cases shown in Figs. 5(b) and 5(c)[45]. Since HoMn₆Sn₆ undergoes an SR transition with increasing temperature, the SOC-induced gap should also evolve with temperature.

IV. CONCLUSIONS

In summary, we have performed systematic transport. magnetic, and spectroscopic measurements in wide temperature, magnetic field, and pressure ranges of $HoMn_6Sn_6$ single crystals. The experimental studies have been supported by first-principles theoretical calculations. Our transport measurement confirms the metallic nature of this compound and shows the signature of a large anomalous Hall effect and contribution of topological Hall effect in this system. From magnetic measurements, we observe a large hysteresis loop and spin reorientation transition below 200 K. We unveil the impact of pressure on magnetic ordering and spin reorientation temperature. Our ARPES measurements reveal the presence of Dirac-like states enclosing the high symmetry point K, supported by the DFT calculations, and suggest the possible existence of a Chern gapped

Dirac-like state in this kagome magnet. Altogether, our detailed studies of the kagome magnet, $HoMn_6Sn_6$, will provide an ideal platform to understand the magnetotransport properties and the electronic structure of various kagome magnets.

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