

Hedgehog-like spin texture in Sb-doped MnBi_2Te_4

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We employ spin- and angle-resolved photoemission spectroscopy and circular-dichroism angle-resolved photoemission spectroscopy to systematically investigate the spin texture of Sb-doped MnBi_2Te_4 . Our results display a hedgehog-like spin texture in this system which is signified by reversed-orienting out-of-plane spins at the Dirac gap. Our finding reveals the presence of time-reversal symmetry breaking, implying the possibility for realization of the high-temperature quantum anomalous Hall effect.

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The interplay of topology and magnetism has led to many novel physical phenomena, such as the quantum anomalous Hall effect (QAHE) [1–3], the axion insulator state [4–6], the spin Hall effect [7], and the magnetic topological superconductivity [8]. These effects hold great potential for future applications in topological quantum computing. Breaking the time-reversal symmetry (TRS) and tuning the Fermi level into a magnetic band gap is crucial for realizing QAHE. The former ensures that only one chiral edge state participates in conduction [3], while the latter is key to achieving quantized Hall conductance [2,9]. The realization temperature of QAHE is closely related to the size of the magnetic gap. Enlarging

the magnetic gap can suppress the destruction of the QAH state caused by thermal excitation, enhancing the topological state's stability, and thereby effectively raising the QAHE temperature. The recently discovered magnetic topological insulator MnBi_2Te_4 is an excellent platform for studying QAHE and the axion insulator state. The spontaneous magnetic order in MnBi_2Te_4 breaks TRS, theoretically opening a magnetic gap of the order of 100 meV in the topological surface state (TSS), which provides the possibility of achieving high-temperature QAHE [10]. However, experiments found that the actual gap size of MnBi_2Te_4 is much smaller than theoretically predicted [11–14], and the realization temperature of QAHE is also not as high as expected [15], which poses challenges for future applications.

Recently, Ma *et al.* reported that Sb substitution on the Bi site can simultaneously tune the Fermi level and the TSS gap size in MnBi_2Te_4 [16]. In this system, the bulk energy gap and the TSS gap are two key energy scales that are related to the realization of QAHE. A larger bulk energy gap is more favorable for band inversion to generate TSS, while a larger TSS gap can ease the tuning of the Fermi level into the gap and enhance the resistance of the QAH state to thermal fluctuations. However, whether the TSS gap is linked to magnetism remains a long-standing unresolved issue in this field. On the one hand, angle-resolved photoemission spectroscopy (ARPES) studies have shown no significant change in gap size across the Néel temperature [16], suggesting that bulk magnetic order may not play a dominant role in the gap opening. On the other hand, magnetic and transport measurements demonstrated that Sb doping induces

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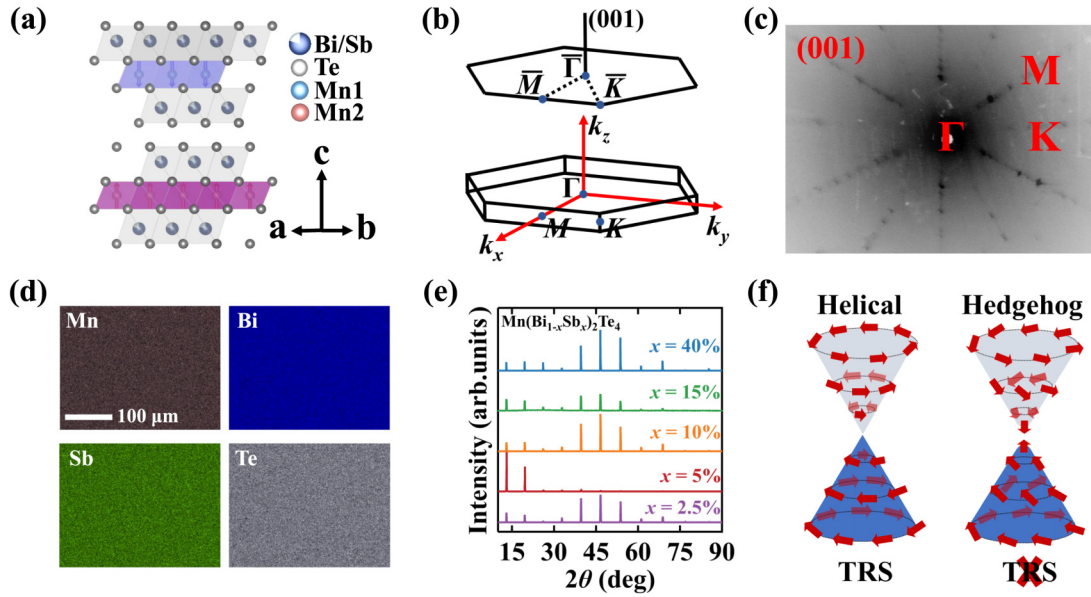


FIG. 1. Basic properties of Sb-doped MnBi_2Te_4 . (a) Crystal structure of Sb-doped MnBi_2Te_4 . The unit cell consists of two septuple layers with different magnetic moments indicated by the arrows. (b) The three-dimensional (3D) and surface-projected BZ of $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$. (c) Laue x-ray diffraction pattern of the (001) surface of $\text{Mn}(\text{Bi}_{0.975}\text{Sb}_{0.025})_2\text{Te}_4$. (d) Energy-dispersive x-ray spectroscopy (EDX) elemental maps of $\text{Mn}(\text{Bi}_{0.9}\text{Sb}_{0.1})_2\text{Te}_4$. (e) Single crystal x-ray diffraction data for the samples on which our data were collected. (f) Schematic of the helical- and hedgehog-like spin textures and their indication on TRS preservation.

short-range magnetic order, which persists up to 80 K—far above the Néel temperature (24 K) [17]. Neutron diffraction and inelastic neutron scattering studies also revealed that antiferromagnetic interactions between Mn_{Sb} antisite defects and Mn atoms survive above the phase transition temperature [18]. These findings imply a possible role of magnetism in the gap opening process. Understanding the nature of the gap is crucial: if the gap is trivial and not magnetically induced, QAHE cannot be realized even if the Fermi level is tuned into the gap [19]. Measuring the spin texture of the gapped TSS could provide information on whether the gap is related to the magnetic breaking of TRS. If the gap is related to magnetism, its spin texture will be hedgehog-like, signified by large and sign-reversed out-of-plane spins at the two sides of the Dirac gap. If the gap is solely induced by chemical disorder on the surface, its spin texture will be helical-like, and no out-of-plane spin is present near the gap [20]. Although many exciting properties of Sb-doped MnBi_2Te_4 were suggested [21–26], the spin configuration of it remains unknown. Investigating the spin texture of Sb-doped MnBi_2Te_4 is not only essential for understanding the nature of its gapped TSS but also provides a reference for future applications in spintronics. In this study, we systematically investigate the spin texture of Sb-doped MnBi_2Te_4 by spin- and angle-resolved photoemission spectroscopy (SARPES) and circular-dichroism ARPES (CDARPES). We observe a hedgehog-like spin configuration in this system, which indicates that the gap is related to the breaking of TRS.

High-quality single crystals of Sb-doped MnBi_2Te_4 were grown using the self-flux method [16]. The samples were characterized by x-ray diffraction (XRD) at room temperature using a Rigaku SmartLab diffractometer with $\text{Cu } K\alpha$ radiation. The diffraction pattern in Fig. 1(e) confirms that the cleaved surfaces of the crystal are parallel

to the (0001) crystallographic orientation. SARPES experiments were performed at Hiroshima Synchrotron Radiation Facilities (HiSOR), Hiroshima, Japan, using the LaserSpin ARPES endstation [27] with a Scienta DA30 analyzer, and the ESPRESSO endstation at BL9B [28] with a VG-SCIENIA R4000 analyzer. A laser light with photon energy $h\nu = 6.3$ eV was used in the former, while synchrotron lights with photon energies $h\nu = 15$ and 28 eV were used in the latter. CDARPES experiments were performed at HiSOR using the μ -ARPES endstation with a VG-SCIENIA R4000 analyzer and a laser with $h\nu = 6.3$ eV [29]. For details of the measurement geometries of the laser ARPES systems; see Fig. S1 in the Supplemental Material [30]. All the samples were cleaved *in situ* along the (0001) crystal plane in an ultrahigh vacuum greater than 7×10^{-11} mbar.

In the doping range we studied, the crystallographic symmetry of Sb-doped MnBi_2Te_4 is found to be the same as that in undoped MnBi_2Te_4 , i.e., a rhombohedral crystal structure with a centrosymmetric space group $R\bar{3}m$. In this quasi-two-dimensional (2D) structure, seven atomic layers are arranged in a Te-X-Te-Mn-Te-X-Te ($X = \text{Bi/Sb}$) sequence, known as a septuple layer (SL). A van der Waals gap exists between adjacent SLs, as shown in Fig. 1(a). Figure 1(b) presents the 3D and surface-projected Brillouin zone (BZ) of $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$, where $\bar{\Gamma}-\bar{M}$ and $\bar{\Gamma}-\bar{K}$ represent two high-symmetry directions of the 2D hexagonal BZ. To identify the high-symmetry directions of the samples, we employed Laue x-ray diffraction, as shown in Fig. 1(c). The elemental maps of all atoms in $\text{Mn}(\text{Bi}_{0.9}\text{Sb}_{0.1})_2\text{Te}_4$ are depicted in Fig. 1(d), revealing a homogeneous distribution of the elements. Figure 1(e) shows the XRD patterns of $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$ for various nominal doping levels x . The minimal shifts in XRD peak positions with varying doping levels are consistent with a previous report [17]. For clarification and better visualization

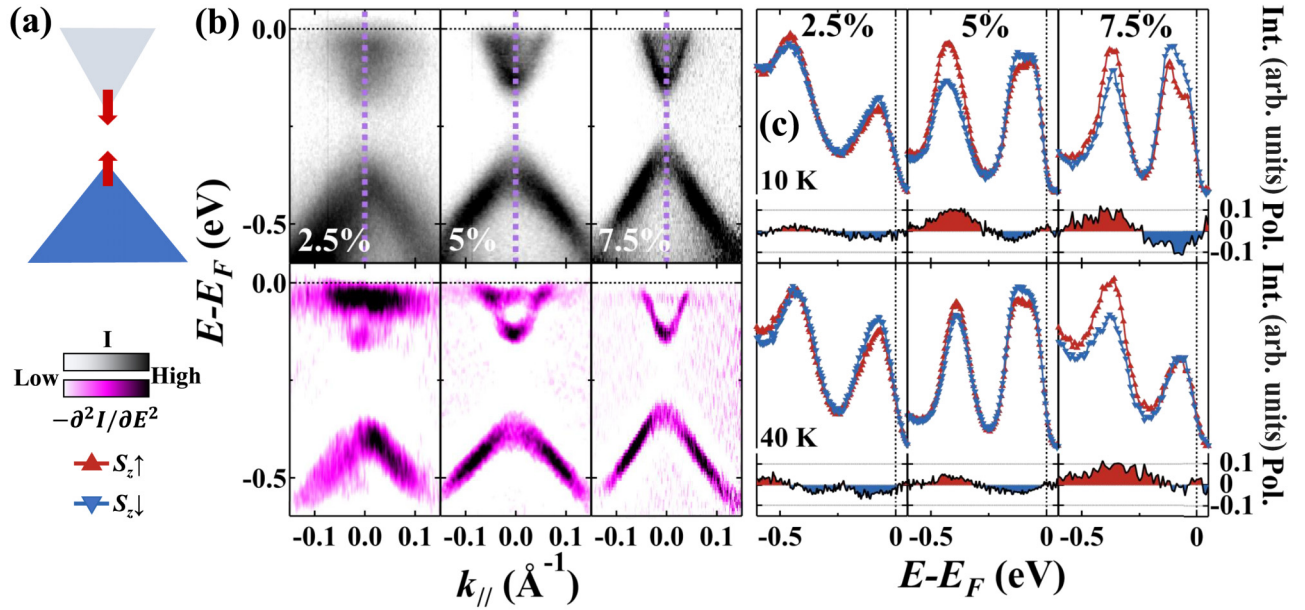


FIG. 2. Spin-integrated and spin-resolved ARPES data acquired at $h\nu = 15$ eV. (a) Schematic of the hedgehog-like spin configuration in the vicinity of the gapped Dirac point. (b) Spin-integrated ARPES data measured using p -polarized light, and the corresponding second derivative analysis along the energy distribution curves (EDCs). The violet dashed lines mark the measurement positions of the spin-resolved EDCs in (c). (c) S_z -resolved EDCs and polarization curves below and above the Néel temperature.

of our main experimental results, schematics of the helical- and hedgehog-like spin textures and their indication on TRS preservation are depicted in Fig. 1(f).

In order to pinpoint experimentally whether the spin texture is helical-like or hedgehog-like, and to study the influence of temperature on the spin configuration, we perform temperature-dependent SARPES measurements at $h\nu = 15$ eV, with the spin direction set to S_z . The results are shown in Fig. 2. Figure 2(a) presents the schematic of the hedgehog-like spin configuration in the vicinity of the gapped Dirac point. Figure 2(b) shows the spin-integrated ARPES E - k maps and the corresponding second derivative analysis. From this panel, we can evaluate the doping levels of the samples based on the conduction band minimum and valence band maximum positions, the same as in our previous study [16]. Both are found to rise from left to right, indicating increasing doping levels. Figure 2(c) shows the S_z -resolved energy distribution curves (EDCs) at $\bar{\Gamma}$ measured at 10 K (below T_N) and 40 K (above T_N). The EDCs for all three samples clearly reveal two peaks: one at $E_B = 0.17$ eV, corresponding to the conduction band minimum [this peak often splits into two nearby peaks, which corresponds to the surface state C_1 and the bulk state C_2 , as discussed in Fig. 3(b)], and another at $E_B = 0.4$ eV, corresponding to the valence band maximum.

For the 5% and 7.5% doped samples, both bands exhibit pronounced out-of-plane spin polarization. The conduction band minimum shows S_z -down polarization, while the valence band maximum shows S_z -up polarization. For the 2.5% doped sample, as shown in Fig. 2(b), the bands appear slightly vague compared to the other two samples, resulting in weaker spin signals. Nevertheless, it can still be discerned that the S_z of the bands above and below the gap are opposite.

Typically, in a topological insulator, TRS enforces spin-momentum locking, resulting in an in-plane helical texture as

illustrated in Fig. 1(f), where the out-of-plane spin component is zero near the zone center. However, in our experiment, we detect a nonzero out-of-plane spin component at $\bar{\Gamma}$, with opposite spin polarizations for the conduction and valence bands. This indicates that TRS is broken in the system, and the spin texture is consistent with the hedgehog-like configuration depicted also in Fig. 1(f). By comparing the spin EDCs at $\bar{\Gamma}$ in Fig. 2(c) which are measured at 10 and 40 K, we observe that the gap size is unchanged across T_N , consistent with our previous report [16], and that the sign of S_z is also temperature insensitive within the binding energy range we measured. The minimal temperature dependence indicates that the gap formation may stem from an interplay between magnetic and nonmagnetic effects.

Besides SARPES, another technique, CDARPES, can also be used to infer the spin information of the material by mapping out the configuration of the orbital angular momentum (OAM). Previous works have shown that in topological materials where spin and momentum are locked, such as Au [31–33], Bi_2Se_3 [34–36], and Bi_2Te_3 [37], CDARPES and SARPES results correspond well with each other [38–40]. Therefore, we also used CDARPES to examine the spin texture of Sb-doped MnBi_2Te_4 , and the results are shown in Fig. 3. To clearly describe the CD pattern, we label the resolved conduction bands and valence bands from top to bottom as C_3 , C_2 , C_1 , V_1 , and V_2 . Based on our previous work [16], C_1 and V_1 correspond to the TSS, and the other three bands correspond to the bulk states. For the four samples measured, the CD signals can be divided into three k -space regions: R_1 ($k < -0.04 \text{ \AA}^{-1}$), R_2 ($-0.04 \text{ \AA}^{-1} < k < 0.04 \text{ \AA}^{-1}$), and R_3 ($k > 0.04 \text{ \AA}^{-1}$). Common characteristics include the following: (1) The TSSs above and below the gap have opposite CD signs: in R_2 , the CD signs of C_1 and V_1 are opposite. This is consistent with the spin ARPES result in Fig. 2, sup-

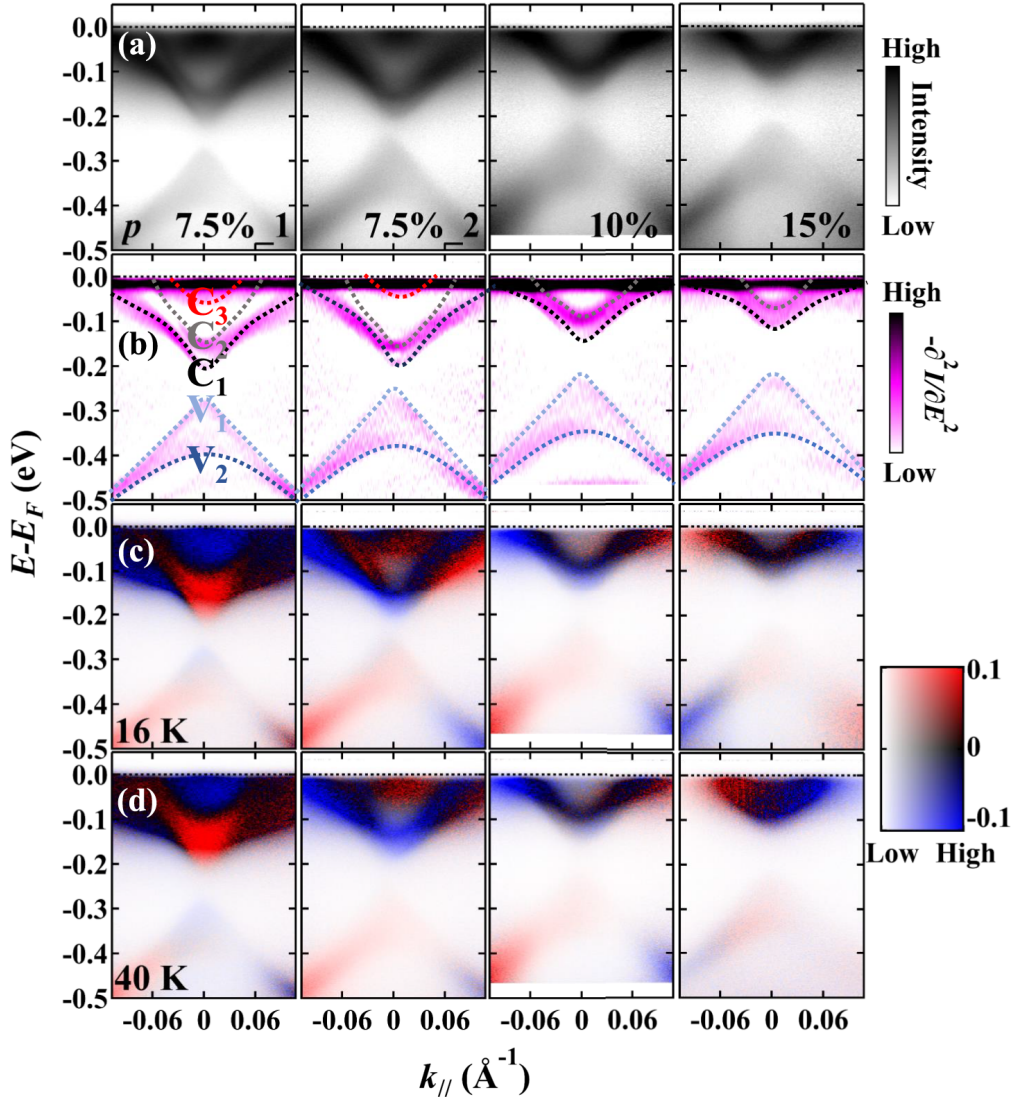


FIG. 3. CDARPES data for $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$ with different x and temperatures, acquired at $h\nu = 6.3$ eV. (a) Spin-integrated ARPES data measured using p -polarized laser radiation. (b) Corresponding second derivative analysis along the EDCs. C_1 - C_3 mark the three conduction bands and V_1 , V_2 mark the two valence bands. (c,d) CDARPES maps below and above the Néel temperature. The intensity of the band structure collected by left- and right-circularly polarized light is defined as I_L and I_R , respectively. The CD intensity is defined as $I_{CD} = (I_L - I_R)/(I_L + I_R)$, where different signs of I_{CD} represent different OAM states of the band structure, defined in the legend at the right.

porting the hedgehog-like spin texture of the gapped TSS. This CD pattern differs from undoped MnBi_2Te_4 , where the TSS shows an “X” shape and an antisymmetric CD sign about $\bar{\Gamma}$ [41,42], indicating that Sb doping alters the OAM near the Dirac point. (2) In R_1 and R_3 , the CD signs of C_1 , V_1 , and V_2 are opposite, consistent with both the hedgehog and the helical spin texture. (3) Temperature has little effect on the CD pattern. Comparing CDARPES results below and above T_N [Figs. 3(b) and 3(c)], the CD pattern remains unchanged, in line with the temperature insensitivity of spin configuration found in Fig. 2 as well as previous results for undoped MnBi_2Te_4 [42]. Sample-dependent signals include the following: (1) the valence bands show three types of patterns: for the $x = 7.5\%$ _1 sample, the pattern is “red-blue-blue” from R_1 to R_3 ; for the $x = 7.5\%$ _2 and 10% samples, it is “red-red-blue”; for $x = 15\%$, it is “blue-red-red.” The conduction band C_2 also shows varying CD patterns: for $x = 7.5\%$ _1, it is mostly

red; for $x = 7.5\%$ _2, it alternates between red (for $k < 0$) and blue (for $k > 0$); for $x = 10\%$, this pattern is opposite to that of $x = 7.5\%$ _2. The CD pattern for the conduction band C_3 is detected only for $x = 7.5\%$ _1 and 7.5%_2. For the former, C_3 is entirely blue; for the latter, C_3 shows a left-blue, right-red pattern. (2) Comparing the CD signs in R_1 and R_3 , the CD sign of the $x = 15\%$ sample is opposite to that of the other samples, indicating that its OAM has inverted. However, the sign of the TSSs in the R_2 region is the same for $x = 7.5\%$ _2, 10%, and 15%, suggesting that doping has little effect on the OAM of TSSs near the Dirac point. We also performed CDARPES constant energy contours measurements, where the CD signal is found to be symmetric along one $\bar{\Gamma}$ - $\bar{M}$ direction and antisymmetric along the other two $\bar{\Gamma}$ - $\bar{M}$ directions (Figs. S2 and S3 in the Supplemental Material [30]). Overall, the CDARPES results shown in Fig. 3, together with the SARPES data in Fig. 2, consistently point to the same

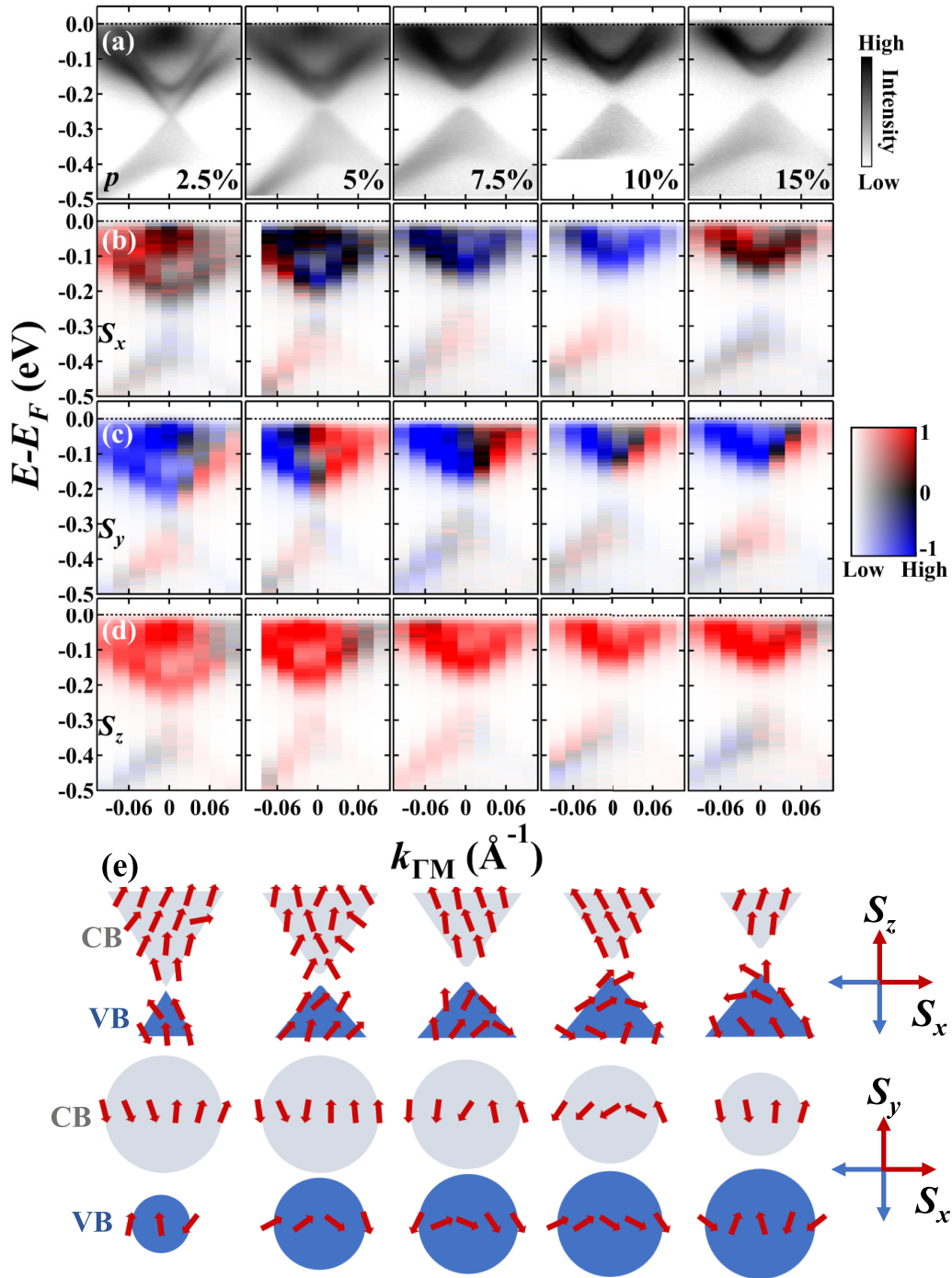


FIG. 4. Spin-integrated and spin-resolved ARPES data for $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$ with different x , acquired at $h\nu = 6.3$ eV. Data was measured using p -polarized laser radiation at a temperature of 10 K (below T_N). (a) Spin-integrated ARPES data. Nominal x values are labeled at the lower right. (b)–(d) Spin-resolved ARPES maps along $\bar{\Gamma}$ – $\bar{M}$ for three spin directions, where the red and blue colors are defined in the legend on the right. (e) Schematic diagrams of the laser-SARPES-measured spin texture for different Sb doping ratio. The gray and blue colors represent the conduction bands (CBs) and the valence bands (VBs), respectively. Directions of red arrows exhibit the spin directions in the S_z – S_x plane (top row) and the S_y – S_x plane (bottom row).

conclusion that the spin configuration of Sb-doped MnBi_2Te_4 is hedgehog-like.

In order to examine the spin texture of Sb-doped MnBi_2Te_4 in three spin directions systematically and thoroughly, we use high-resolution laser-SARPES with 6.3 eV incident photons and p -polarized light (light polarization parallel to the detection plane [43]) to investigate the spin texture with dif-

ferent doping levels. The measurements are conducted at 10 K (below T_N), with the momentum direction parallel to $\bar{\Gamma}$ – $\bar{M}$. Figure 4(a) presents the spin-integrated ARPES results. The TSS gap gradually widens, and the conduction band minimum rises, as the doping level increases, reproducing the results in Ref. [16]. Figures 4(b)–4(d) show the SARPES results along different spin directions (see Figs. S5–S7 in the Supplemental

Material [30] for raw spin EDCs). Figure 4(e) presents the schematic diagrams of the laser-SARPES-measured spin texture for both the conduction and the valence bands.

From the data in Fig. 4, at this photon energy, the spin textures for samples with different doping concentrations have commonalities and sample-dependent characteristics. On the one hand, for the conduction bands, the in-plane spin is approximately tangent to the momentum in all five samples measured, as the S_y component always points out of the page at $k < 0$ and into the page at $k > 0$ [Fig. 4(c)]. Such spin-momentum locking exhibits a helical in-plane spin texture. The radial spin component S_x also exists, which is smaller and has in general an opposite sign between the valence band and the conduction band at the same k [Fig. 4(b)]. Importantly, a substantial out-of-plane spin component (S_z) is observed for both the conduction and the valence band [Fig. 4(d)]. Under the laser light, S_z is found to be always positive near the TSS gap, which seemingly differs from the spin texture of Sb-doped MnBi_2Te_4 found in Fig. 2 as well as that of magnetically doped Bi_2Se_3 and pristine MnBi_2Te_4 , whose S_z changes sign across the topological gap [16,20,44]. Although we primarily attribute such all-up configuration of S_z to final-state effects (discussed below), we emphasize that the very existence of out-of-plane spin polarization on both sides of the gap is evidence for a breaking of TRS.

On the other hand, we observed sample-dependent spin polarization signals across the five samples. As shown in Figs. 4(b)–4(d), S_x for the $x = 5\%$, 7.5% , and 10% samples are opposite to that of the $x = 2.5\%$ and 15% samples. In the valence bands near the TSS gap, S_y points out of the page at $k > 0$ and into the page at $k < 0$ for the $x = 5\%$, 7.5% , and 10% samples, whereas the direction is reversed for the $x = 2.5\%$ and 15% samples. S_z , although primarily positive in the valence bands near the TSS gap for all samples, shows also sample-dependent polarizations farther from the gap, as it mainly points down (up) at $k < 0$ ($k > 0$) for $x = 2.5\%$, 10% , and 15% , while it points up (down) at $k < 0$ ($k > 0$) for $x = 5\%$ and 7.5% .

By comparing the experimental results in Figs. 2(c) and 4(d), we find that at $h\nu = 15$ eV, S_z of the conduction and valence bands have opposite signs. In contrast, at $h\nu = 6.3$ eV, S_z of both the conduction and valence bands is mainly pointing up. The former case is consistent with the reported hedgehog-like spin texture, indicative of the breaking of TRS. We primarily attribute the universal spin-up out-of-plane spin polarization observed in our laser-SARPES results to the final-state effects. Previous works reported that, when using circularly polarized 6 eV photons to probe the spin polarization of Bi_2Se_3 , the final-state spin texture dominates the observed spin polarization, which changes according to the light polarization [45,46]. Although this phenomenon is slightly different from the situation in our experiment—the spin result shown in Fig. 4 is acquired using p -polarized light, whereas Ref. [46] used circularly polarized light—it is possible that p -polarized 6 eV photons produce similar final-state effect, rendering the laser-SARPES result unrelated to the intrinsic spin configuration.

Now we summarize and discuss our experimental results. First, combining all experimental evidence, we stress that the

spin configuration of this system, in particular that of the TSS, is hedgehog-like. By careful band analysis, we attribute the opposite S_z component near the band gap mainly to the TSS. As shown in the spin EDCs of Fig. 2(c), the conduction band often displays two peaks. Comparing these with the E - k map in Fig. 3(b), we found that the peak near the Fermi level corresponds to the bulk band C_2 , and the peak near the Dirac point corresponds to the surface state C_1 . In the valence band, only one peak is resolved, corresponding to the surface state V_1 . At $h\nu = 15$ eV, the S_z component of C_1 is negative, while that of V_1 is positive, consistent with the hedgehog-like spin texture. This is also supported by the CD results in Fig. 3, in which the CD signals of the surface state have opposite signs at the energies near the Dirac point. Far from the Dirac point, the CD signal exhibits a helical OAM texture, consistent with observations in Mn-doped Bi_2Se_3 [20]. In light with the facts that SARPES with higher incident photon energies tend to exhibit intrinsic spin configuration of the samples, and that our CDARPES data are in line with the SARPES results at $h\nu = 15$ eV, we believe that the spin configuration of Sb-doped MnBi_2Te_4 is consistent with the hedgehog-like spin texture.

Next, we discuss the possible reason for this sample-dependent hedgehog-like spin configuration. A relevant reference is the work by Xu *et al.* [20], who investigated the spin texture in Bi_2Se_3 thin films doped with either magnetic (Mn) or nonmagnetic (Zn) elements. SARPES revealed opposite out-of-plane polarizations at $\bar{\Gamma}$ in Mn-doped films, while Zn-doped films preserved the helical spin texture of pristine Bi_2Se_3 . Xu *et al.* interpreted these observations as follows: in topological insulator Bi_2Se_3 , TRS enforces spin-momentum locking, resulting in an in-plane helical texture. Nonmagnetic Zn doping preserves this symmetry, while magnetic doping breaks it, introducing significant out-of-plane spin polarization and leading to the emergence of a hedgehog-like texture. A similar mechanism may also explain the spin configuration in Sb-doped MnBi_2Te_4 . Numerous experiments have demonstrated that MnBi_2Te_4 possesses helical Rashba-type in-plane spin texture [47–49]. Sb substitution may modify the magnetic ground state of MnBi_2Te_4 , breaking the TRS of the surface, which leads to the opposite out-of-plane spin polarization observed in Fig. 2(c). The observation of such hedgehog-like spin texture suggests that Sb doping not only enlarges the gap but also keeps the material TRS broken. However, the absence of significant temperature dependence implies that the gap may arise from a combined effect of magnetic and nonmagnetic mechanisms. While the exact origin remains to be fully resolved, we believe our SARPES data provide valuable experimental insights for future studies.

Regarding the sample dependence of spin helicity we observed primarily in our laser-SARPES measurements, we hypothesize that it may stem from the combination of the following three reasons: (1) Possible hidden spin polarization (HSP) in this system, which arises from local inversion symmetry breaking in centrosymmetric crystals [50,51]: Sb-doped MnBi_2Te_4 satisfies the symmetry conditions for HSP: although its bulk space group is centrosymmetric, the Bi and Te atoms occupy noncentrosymmetric C_{3v} sites, which leads to spatially separated sectors carrying opposite spin

polarizations [50]. Given the small laser spot size ($<10\ \mu\text{m}$ [27]), it is plausible that each measurement captured only a single sector, and differences in cleaved surface conditions across samples could account for the variation in the observed spin polarization. (2) Probing from different magnetic domains: Although the laser beam spot is small enough ($<10 \times 10\ \mu\text{m}^2$) to potentially probe individual domains in this system [52], the varying local magnetization orientations of magnetic domains are expected to give rise to different spin textures across different samples. (3) Unavoidable experimental uncertainties in the measurement process: Jozwiak *et al.* have demonstrated that when probing the spin texture of a topological insulator using a 6 eV laser, the measured spin texture can be manipulated by light polarization [45]. Based on their calculated spin texture, we constructed a model under our experimental conditions (Fig. S8 in the Supplement Material [30]) to qualitatively explain the observed sample-dependent spin helicity. The two distinct S_x polarization signs observed in Fig. 4 may originate from slight differences in measurement positions relative to the actual $\vec{\Gamma}$ - $\vec{M}$ direction, which can reverse the sign of S_x across different scans depending on the measurement direction, while leaving S_y unaffected. Within a single scan, S_x exhibits opposite signs in the conduction and the valence bands.

In conclusion, we examine the spin configuration of Sb-doped MnBi_2Te_4 by both SARPES and CDARPES. We found

sample-dependent hedgehog-like spin configuration in this system, consistent with the existence of TRS breaking. Our results imply the possibility of manipulating the spin states and realizing high-temperature QAHE in Sb-doped MnBi_2Te_4 .

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Data Availability. The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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