Hedgehog spin texture and Berry’s phase tuning in a magnetic topological insulator

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Understanding and control of spin degrees of freedom on the surfaces of topological materials are key to future applications as well as for realizing novel physics such as the axion electrodynamics associated with time-reversal (TR) symmetry breaking on the surface. We experimentally demonstrate magnetically induced spin reorientation phenomena simultaneous with a Dirac-metal to gapped-insulator transition on the surfaces of manganese-doped Bi2Se3 thin films. The resulting electronic groundstate exhibits unique hedgehog-like spin textures at low energies, which directly demonstrate the mechanics of TR symmetry breaking on the surface. We further show that an insulating gap induced by quantum tunnelling between surfaces exhibits spin texture modulation at low energies but respects TR invariance. These spin phenomena and the control of their Fermi surface geometrical phase first demonstrated in our experiments pave the way for the future realization of many predicted exotic magnetic phenomena of topological origin.

Since the discovery of three-dimensional topological insulators1–5 (TI), topological order proximity to ferromagnetism has been considered as one of the core interests of the field6–16. Such interest is strongly motivated by the proposed TR breaking topological physics such as the quantized anomalous chiral Hall current, spin current, axion electrodynamics, and the inverse spin-galvanic effect9–12, all of which critically rely on finding a way to break TR symmetry on the surface and use the unique TR broken spin texture for applications. As quantum coherence is essential in many of these applications, devices need to be engineered into thin films to enhance or de-enhance surface-to-surface coupling or the quantum tunnelling of the electrons. The experimental spin behaviour of surface states under the two extreme limits, namely the doped magnetic groundstate and ultrathin film quantum tunnelling groundstate, is thus of central importance to the entire field. However, surprisingly, it is not known what happens to the spin configuration under these extreme conditions relevant for device fabrication. Fundamentally, TR symmetry is inherently connected to the Kramers’ degeneracy theorem, which states that when TR symmetry is preserved, the electronic states at the TR invariant momenta have to remain doubly spin degenerate. Therefore, the establishment of the TR breaking effect fundamentally requires measurements of electronic groundstate with a spin-sensitive probe. Here we use spin-resolved angle-resolved photoemission spectroscopy (ARPES) to measure the momentum space spin configurations in systematically magnetically doped, non-magnetically doped, and ultrathin quantum coherent TI films17, to understand the nature of electronic groundstates under the two extreme limits vital for magnetic topological devices. These measurements allow us to make definitive conclusions regarding magnetism on topological surfaces, and make it possible to quantitatively isolate the TR breaking effect in generating the surface electronic gap from many other physical or chemical changes also leading to gap-like behaviour18–22 often observed on the surfaces. Spin reorientation measurements and the systematic methodology demonstrated here can be used to probe quantum magnetism on the surfaces of other materials as well.

Topological surface states with magnetic doping

To study the evolution of topological surface states on magnetic doping, magnetically (Mn%) and (chemically similar) non-magnetically (Zn%) doped Bi2Se3 thin films are prepared in high quality using the molecular beam epitaxy (MBE) growth method. A sample layout and a photographic image of a representative MBE-grown film used for experiments are shown in Fig. 1a,b. Using standard X-ray magnetic circular dichroism23,24 (XMCD), we characterize the magnetic properties of doped Bi2Se3 films (Fig. 1c). In Mn-doped Bi2Se3, a hysteretic behaviour in the out-of-plane magnetic response is consistently observed, which suggests a ferromagnetically ordered groundstate. The observation

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of ferromagnetic character and its absence in Zn–Bi$_2$Se$_3$ motivate us to systematically compare and contrast the electronic density of state behaviour in the vicinity of the surface Dirac node of these samples. Figure 1d shows the measured electronic states of Mn(Zn)-doped Bi$_2$Se$_3$ using high-resolution (spin-integrated) ARPES. In the undoped Bi$_2$Se$_3$ film (leftmost panel of Fig. 1d), a map of the spectral density of states reveals a bright and intact Dirac node (signalled by the red spot located at the Dirac crossing in the panel), which indicates that in undoped Bi$_2$Se$_3$ the Dirac node is gapless, consistent with the previous studies of pure Bi$_2$Se$_3$ (ref. 25). In samples where Mn atoms are doped into the bulk (first row of Fig. 1d), we observe that the corresponding bright (red) spot at the Dirac node gradually disappears, revealing a clear systematic spectral weight suppression (SWS) with increasing Mn concentration. In contrast, the spectral intensity at the Dirac node is observed to survive on systematic Zn doping, except for the Zn = 10% sample, where some suppression of intensity is observed. This suggests that the Dirac node remains largely intact on Zn doping. The observed systematic behaviour of spectral evolution motivates us to quantitatively define an energy scale, $E_{\text{SWS}}$, associated with the SWS observed at the Dirac node. The value of $E_{\text{SWS}}$ is taken as the energy spacing between the upper Dirac band minimum and the Dirac node location along the energy axis, as illustrated in Fig. 1e, which roughly corresponds to half of the surface gap magnitude. The value of the energy scale can be quantitatively determined by fitting the ARPES-measured energy-momentum distribution curves (MDCs; method described in Supplementary Section SVI). The doping dependence of $E_{\text{SWS}}$ on samples measured at $T = 20$ K is shown in Fig. 2c. The value of $E_{\text{SWS}}$ is observed to increase rapidly with Mn concentration, but it remains nearly zero with Zn doping. The temperature dependence of $E_{\text{SWS}}$ is shown in Fig. 2d. The temperature induced decrease of $E_{\text{SWS}}$ is consistent with a gradual weakening of magnetism. These observations collectively reveal a direct correlation between XMCD measured ferromagnetic character and the ARPES measured SWS (or gap) on the Mn–Bi$_2$Se$_3$ films.

Although the predominant trend in the doping evolution of surface states suggests a correlation between magnetism and SWS, we do notice a non-negligible $E_{\text{SWS}}$ at high Zn concentration (10%). This is probably due to increasing chemical disorder on the surface of the film, because disorder degrades the surface quality. Similarly, magnetization measurements show that ferromagnetism vanishes before reaching $T = 300$ K (inset of Fig. 2b), but nonzero SWS (Fig. 2d) is still observed, perhaps, in this case, because of thermal disorder of the relaxed film surface at high temperatures. Therefore, the correlation between ferromagnetism and the ARPES gap is only clear at low temperatures in samples with reduced disorder. The momentum width ($\Delta k$) of the surface electronic states can be taken as a rough relative measure of surface disorder.
sample to sample, which is found to significantly increase on both magnetic and non-magnetic dopings (Fig. 2c). Strong spatial fluctuations of the surface electronic states in doped Bi$_2$Se$_3$ have been observed in recent scanning tunnelling microscope (STM) work$^{18}$, where the authors suggest the observation of gap-like feature at the Dirac point without breaking TR symmetry. These ambiguities$^{20}$ associated with the observed gap-like feature across many different experiments strongly call for critically important spin-resolved measurements which also serve as a collective method, as we show, to unambiguously identify the correct nature of the gap.

**Spin configuration of the magnetic groundstate**

To study the evolution of spin texture on magnetic doping, we performed spin-resolved measurements on Mn–Bi$_2$Se$_3$ topological surface states. We present two independent but representative spin-resolved ARPES measurements on Mn(2.5%)–Bi$_2$Se$_3$ film I and film II. Films I and II, both containing the same nominal Mn concentration, are measured and analysed using two different spectroscopic modes, namely, the spin-resolved MDC measurement mode and the spin-resolved energy distribution curve (spin-resolved EDC) measurement mode (see Methods), to exclude any potential systematic error in the spin measurements. Figure 3a–d shows measurements on film I. Our data shows that out-of-plane spin polarization $P_z$ is nearly zero at large momentum $k_z$ far away from the Dirac point energy ($0 < E_B < 0.1$ eV in Fig. 3c,d). On approaching the Dirac point ($0.1$ eV $< E_B < 0.3$ eV), an imbalance between the spin-resolved intensity in $+z$ and $-z$ is observed (Fig. 3c). The imbalance is found to become systematically more pronounced in the data set where scans are taken by lowering the temperature toward the Dirac point. This systematic behaviour observed in the data reveals a net significant out-of-plane spin polarization in the vicinity of the `gapped' Dirac point or near the bottom of the surface state conduction band. More importantly, the out-of-plane spin component $P_z$ does not reverse its sign in traversing from $-k_z$ to $+k_z$. Such behaviour is in sharp contrast to the spin textures observed in pure Bi$_2$Se$_3$ (ref. 26), where spins point to opposite directions on opposite sides of the Fermi surface, as expected from TR symmetry. Therefore, our $P_z$ measurements on film I near the gap edge reveal the TR breaking nature of the Mn–Bi$_2$Se$_3$ sample where magnetic hysteresis was observed using the XMCD technique. To directly measure...
the spin of the surface state at $\Gamma$ (the Kramers’ momentum, $k_z = 0$), we perform spin measurements on Mn–Bi$_2$Se$_3$ film II (same Mn concentration as film I) working in the spin-resolved EDC mode. The measured out-of-plane spin polarization ($P_z$) is shown in Fig. 3g. We focus on the $P_z$ measurement at $\Gamma$, the Kramers’ momentum $k_z = 0$ (red curve); the surface electrons at TR invariant $\Gamma$ are clearly observed to be spin polarized in the out-of-plane direction. The opposite sign of $P_z$ for the upper and lower Dirac band (red curve in Fig. 3g) shows that the Dirac point spin degeneracy is indeed lifted up ($E(k_z = 0, \uparrow) \neq E(k_z = 0, \downarrow)$). Such an observation directly counters the Kramers’ theorem and therefore manifestly breaks the TR symmetry on the surface. Next we analyse $P_z$ measurements at finite $k_z$ (green curves in Fig. 3g) to extract the detailed configuration of the spin texture. In going to larger $k_z$ away from the $\Gamma$ momenta, the measured $P_z$ is found to gradually decrease to zero. Moreover, the constant energy momentum space plane at the Dirac point ($E_\parallel = E_\perp$) is observed to serve as a mirror plane that reflects all of the out-of-plane spin components between the upper and lower Dirac bands. Thus both spin-resolved MDC (film I) and spin-resolved EDC (film II) measurement modes result in consistent conclusions regarding the spin configuration of the films. These systematic measurements, especially at the vicinity of the gap, reveal a hedgehog-like spin configuration for each upper (or lower) Dirac band separated by the magnetic gap, which breaks TR symmetry, as schematically shown in the inset of Fig. 3f (see Supplementary for further data and analysis).

**Spin configurations of the non-magnetic groundstates**

Spin texture measurements on non-magnetically doped films Zn(1.5%)–Bi$_2$Se$_3$ are presented in Fig. 4a–d. The out-of-plane polarization $P_z$ measurements reveal a sharp contrast to the magnetically doped Mn–Bi$_2$Se$_3$ films, specifically, the near absence of a finite $P_z$ component around $\Gamma$ within our experimental resolution (Fig. 4d). A very small $P_z$, however, at large $k_z$ is observed, which is expected because of surface state warping also observed in other TI compounds$^{27}$ (Fig. 4d). That the signal is associated with warping is further confirmed in our data because of their TR symmetric nature with $P_z$ being observed to reverse its sign in traversing from $-k_z$ to $+k_z$. Moreover, our in-plane spin measurements (Fig. 4c) show that Zn–Bi$_2$Se$_3$ film retains the helical spin texture protected by the TR symmetry, as observed in pure Bi$_2$Se$_3$ and Bi$_2$Te$_3$ single crystals$^{26}$. Therefore we conclude that non-magnetic Zn doping does not induce observable spin reorientation on the topological surface. The contrasting behaviour observed between Mn–Bi$_2$Se$_3$ and Zn–Bi$_2$Se$_3$ samples, as
A surface bandgap at the Dirac point can also be generated in Bi$_2$Se$_3$ in its ultrathin film limit. In this case, the top and bottom surfaces couple together and open up a gap as electrons can tunnel from one to the other (Fig. 4e–h). Such a gap in the surfaces is not related to magnetism. It is important to know the spin configuration associated with such a gap. In Fig. 4e–h, we use spin-resolved ARPES to measure the spin configuration on the very top region (within 5 Å) of a Bi$_2$Se$_3$ film whose thickness is three quintuple layers (QL; 3 QL $\approx$ 28.6 Å). At large parallel-momenta far away from $\Gamma$ (for example $-0.10$ Å$^{-1}$ in Fig. 4g), we observe clear spin polarization following a left-handed helical configuration with the magnitude of the polarization around 35–40%. However, in going to smaller $k$, the magnitude of the spin polarization is observed to be reduced. At the TR invariant $\Gamma$ momenta, spin-resolved measurements (Fig. 4g red curve) show no net spin polarization. This reduction of the spin polarization at small momenta near the gap is an intrinsic effect (Supplementary Fig. S19). These observations can be understood by considering the scenario where the surface-to-surface coupling dominates, and the two energetically degenerate surface states from top and bottom, which possess opposite helicities of the spin texture, cancel each other at $\Gamma$ (ref. 17). This results in strong suppression of the spin polarization in the vicinity of this gap, whereas on probing momenta to large $k$ away from $\Gamma$, the finite kinetic energy of the surface states ($\propto v_{k}$) naturally leads to the spatial decoupling of two Dirac cones. These spin measurements on the ultrathin Bi$_2$Se$_3$ film reveal the interplay between quantum tunnelling (coupling) and the spin texture modification, which is of importance in spin-based device design with thin films. The observed spin texture, however, does not break TR symmetry, as the spins remain doubly degenerate at the TR invariant momenta $\Gamma$. This is in clear contrast to the spin texture observed in Mn–Bi$_2$Se$_3$.

**Figure 4 | Spin configurations on non-magnetic samples.** a–d. Spin-resolved measurements on 1.5% non-magnetic Zn–Bi$_2$Se$_3$ film. The in-plane polarization measurements (c) reveal the helical spin configuration, as in pure Bi$_2$Se$_3$ TI (ref. 26), suggesting that non-magnetic impurities do not induce spin reorientation on the topological surfaces. Out-of-plane measurements (d) show that no significant out-of-plane spin polarization $P_{z}$ is induced near the $\Gamma$ point (the TR invariant momenta), leaving the system TR invariant overall. e–g. Spin-resolved ARPES measurements on ultrathin undoped Bi$_2$Se$_3$ film of three QL thickness. The net spin polarization is found to be significantly reduced near the gap edge around the $\Gamma$ momenta. This is consistent with the fact that in ultrathin films electrons tunnel between the top and bottom surfaces. h, i. A schematic of the two types of spin texture observed in our data.

**Magnetic contribution and geometrical phase tuning**

The magnetic contribution to the gap of the Mn–Bi$_2$Se$_3$ film can be quantitatively identified using the spin texture data. The simplest k-p Hamiltonian that describes topological surface states under TR symmetry breaking can be written as $\hat{H} = v (k_{x} \sigma_{y} - k_{y} \sigma_{x}) + b_{z} \sigma_{z}$, where $\sigma$ and $k$ are the spin and momentum operators respectively, $b_{z}$ corresponds to half of the magnetic gap and $v$ is the velocity of the surface Dirac band. We specify the out-of-plane polar angle $\theta$ of the spin polarization vector (inset of Fig. 3g) as $\theta = \arctan P_{z}/P_{\parallel}$. The magnitude of the polar angle $\theta$ reflects the competition between the out-of-plane TR breaking texture ($\propto b_{z}$) and the in-plane helical configuration component ($\propto v k_{z}$). Using the measured spin-resolved data sets ($\theta, k$), we fit the magnetic interaction strength $b_{z}$ within a k-p scenario (see Supplementary Section SII.2). As an example, we fit $b_{z}$ based on spin-resolved data sets in Fig. 3g on Mn$_{2.5}$%–Bi$_2$Se$_3$ film II, as shown in Fig. 5c, and obtain a value of 21 meV. This is a significant fraction of the SWS energy scale observed on the same sample, $E_{SWS} > 50$ meV (see Fig. 2c for Mn$_{2.5}$% obtained from the spin-integrated measurements in Fig. 1c. Thus we identify the magnitude of the magnetic contribution ($b_{z}$) to the observed spectral weight suppression using spin-sensitive measurements, which suggests that the magnetic contribution is significant to $E_{SWS}$ in Mn–Bi$_2$Se$_3$.

As demonstrated recently$^{28}$, the geometrical phase (GP) defined on the spin texture of the surface state Fermi surface$^{3}$ (also known as Berry’s phase) bears a direct correspondence to the bulk topological invariant realized in the bulk electronic band structure via electronic band inversion$^{26,28}$. We experimentally show that a GP tunability can be realized on our magnetic films, which is important to prepare the sample condition to the axion electrodynamics limit. On the Mn–Bi$_2$Se$_3$ film, the spin configuration pattern can be understood as a competition between the out-of-plane TR breaking component and the in-plane helical component of spin. The in-plane spin,
which can be thought of as winding round the Fermi surface in a helical pattern, contributes to a nonzero GP (ref. 26), whereas the out-of-plane TR breaking spin direction is constant as one loops around the Fermi surface and hence does not contribute to the GP. As a result, the GP remains almost π if the chemical potential lies at energies far away from the Dirac point, whereas it starts to decrease and eventually reaches zero as one approaches the TR breaking gap by lowering the chemical potential, as discussed in theory29, at least within the magnetic energy scale $b_0$ (Fig. 5). We show that this theoretical requirement can be experimentally achieved on the Mn–Bi$_2$Se$_3$ surface through surface NO$_2$ adsorption at some specific dosage level. Figure 5a shows the Mn(2.5%–Bi$_2$Se$_3$) surface states with $\textit{in situ}$ NO$_2$ adsorption. The chemical potential is observed to be gradually shifted, and finally placed within the magnetic gap. The associated phase (GP) at each experimentally achieved sample chemical potential is found to gradually change from π to 0. The GP = 0 is the experimental condition for realizing axion electrodynamics with a TI (refs 9,11).

With the chemical potential moved into the magnetic gap, the TR breaking in-gap state features a singular hedgehog-like spin texture (Fig. 5d). Such a spin configuration, simultaneous with the chemical potential placed within the magnetic gap (Fig. 5d), is the fundamental requirement for most of the theoretical proposals relevant to the use of magnetic TIs in novel devices5–12. Furthermore, if the bulk bandgap of the Mn–Bi$_2$Se$_3$ film is tuned to zero at the critical point of the topological phase transition22, a new topologically protected Weyl semimetal phase30,31 with a yet more exotic but undiscovered state of matter is also expected to take place, and is among the most exciting future frontiers to be enabled by our achievement of a sample that features a TR broken hedgehog-like spin texture with a GP = 0 state30,31.

**Methods**

**Sample growth.** The Mn-doped Bi$_2$Se$_3$ thin films were synthesized by MBE using high-purity elemental (5N) Mn, Bi, and Se sources. A thin GaAs buffer layer was first deposited on the epi-ready GaAs 111A substrate after thermal desorption of the native oxide under As pressure. Then the substrate was transferred to another chamber, without breaking the vacuum, where a second buffer layer of ZnSe was deposited for further smoothing the surface. A Mn–Bi$_2$Se$_3$ layer (~60 nm) was then grown with a high Se/Bi beam equivalent pressure (BEP) ratio of ~15. The Mn doping concentration was controlled by adjusting the Bi/Mn BEP ratio in the range from 8 to 60. To protect the surface from oxidation, a thick Se capping layer was deposited on the Mn–Bi$_2$Se$_3$ thin film immediately after the growth. The Zn-doped Bi$_2$Se$_3$ control samples were also synthesized under the same conditions as Mn–Bi$_2$Se$_3$, with the Zn doping concentration controlled by Bi/Zn BEP. An ultrathin Bi$_2$Se$_3$ film was prepared with a thickness of 3 QL, with a typical 1 QL peak to peak variation (2–4 QL). Complete details of film growth are presented in ref. 32 and also detailed in the Supplementary Information.

**Electronic structure measurements.** ARPES measurements (spin-integrated) were performed with 29–64 eV photon energy on beamlines 10.0.1 and 12.0.1 of the Advanced Light Source at Lawrence Berkeley National Laboratory. Spin-resolved ARPES measurements were performed on the 13 beamline at Maxlab in Lund, Sweden, COPEE spectrometer SIS beamline at the Swiss Light Source in Switzerland, and UE112-PGM1 beamline PHOENIXS chamber at BessyII in Berlin, Germany, using photon energies of 8–11 eV, 20–22 eV, and 55–60 eV for the three beamlines respectively. For ARPES measurements, samples were annealed in situ to evaporate the amorphous Se cap layer and reveal the clean surface (see Supplementary Information for further details).

**Spin-resolved measurements.** Spin-resolved measurements were performed using the spin-resolved ARPES beamlines mentioned above with double classical Mott detectors and linearly polarized photons. The spin-resolved measurements in Figs 3 and 4 were performed in two different modes: the spin-resolved MDC
mode and the spin-resolved EDC mode. The spin-resolved EDC mode means each single measurement measures the spin polarization at a fixed binding energy ($E_b$) along a certain momentum cut direction in momentum space (see Fig. 3a-d on Mn(2.5%)–Bi$_2$Se$_3$ film I). The spin-resolved EDC mode means each single measurement measures the spin polarization at a fixed momentum ($k$) along the binding energy axis ($E_b$; see Fig. 3e-g and Fig. 4 on Mn(2.5%)–Bi$_2$Se$_3$ film II, Zn(1.5%)–Bi$_2$Se$_3$ film, and 3 QL undoped Bi$_2$Se$_3$ film). Details of the spin-resolved technique and principles can be found in Supplementary Information.

**Magnetic property characterizations.** To cross-check the ferromagnetism implied by the spin measurements on the Mn–Bi$_2$Se$_3$ surface, XMCD was independently performed on a Mn–Bi$_2$Se$_3$ film surface at the back-endstation of the D1011 beamline at Maxlab in Lund, Sweden, with total electron yield mode at temperatures ranging from 40 K to 300 K. The XMCD measurements were performed on the $L_3$ absorption edge of the Mn atom using standard methods$^{13}$ that are widely used to study the magnetic properties of transition metals and dilute magnetic semiconductor thin films or monolayers$^{14,15}$.

Detailed principles and methods of all techniques used in the experiments, including sample growth, ARPES and spin-resolved ARPES, NO$_2$ surface adsorption, and XMCD are further elaborated in the Supplementary Information.

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**Author contributions**

S.Y.X. performed the experiments with assistance from M.N., C.L., L.A.W., N.A. and M.Z.H.; D.Z., A.R. and N.S. provided samples; M.L., T.B., J.S.-B., O.R., G.L., B.S., J.H.D. and I.O. provided beamline assistance; T.-R.C., H.-T.J., H.L. and A.B. carried out the theoretical calculations; M.Z.H. was responsible for the overall direction, planning and integration among the different research units.

**Additional information**

The authors declare no competing financial interests. Supplementary information accompanies this paper on www.nature.com/naturephysics. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to M.Z.H.