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Topological Phase Transition and Texture Inversion in a Tunable Topological Insulator

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The recently discovered three-dimensional or bulk topological insulators are expected to exhibit exotic quantum phenomena. It is believed that a trivial insulator can be twisted into a topological state by modulating the spin-orbit interaction or the crystal lattice, driving the system through a topological quantum phase transition. By directly measuring the topological quantum numbers and invariants, we report the observation of a phase transition in a tunable spin-orbit system, BiTl(S1−xSex)2, in which the topological state formation is visualized. In the topological state, vortex-like polarization states are observed to exhibit three-dimensional vectorial textures, which collectively feature a chirality transition as the spin-momentum–locked electrons on the surface go through the zero carrier density point. Such phase transition and texture inversion can be the physical basis for observing fractional charge ($\pm e/2$) and other fractional topological phenomena.

Topological insulators in three dimensions are nonmagnetic insulators with novel surface states that are a consequence of the nontrivial topology of electronic wave functions in the bulk of the materials ($I$–$5$). Their experimental discoveries in Bi-Sb semiconductors, and in Bi$_2$Se$_3$ and related materials, have led to the exploration of topological quantum phenomena at modest temperatures and without the requirement of any applied magnetic field ($6$–$16$). Current theoretical proposals have suggested such intriguing possibilities as the realization of fractional quasi-particles and the exploitation of the role of spin momentum locking, which leads to quantum phases (such as topological Berry’s phases), in transport ($17$–$24$).

Strong topological insulators are distinguished from ordinary insulators by a finite topological quantum number or an invariant ($v_0$) or, equivalently, an axion angle parameter ($\theta$). The value of $v_0$ or $\theta$ depends on the parity eigenvalues of the wave functions in time reversal–symmetric materials ($I$, $2$) and can be determined from spin texture details of the spin-orbit surface states that form when the bulk is terminated, as demonstrated by Hsieh et al. ($6$–$9$). In particular, a $v_0 = 1 = \theta/\pi$ topology requires the terminated surface to have a Fermi surface that supports a nonzero Berry’s phase (an odd as opposed to an even multiple of $\pi = \theta = \text{axion angle}$), which is not realizable in an ordinary spin-orbit or trivial ($v_0 = 0 = \theta$) insulator. It is believed that a trivial insulator can be twisted into a $v_0 = 1 = \theta/\pi$ topological state by signaling the appearance of unusual spin vortex–like [or skyrmion-like ($I$)] arrangements via increasing spin-orbit interaction or by modulating the lattice parameters, thereby driving the system through a topological quantum phase transition ($1$, $2$). However, the topological insulators Bi$_2$X$_2$ ($X = \text{Se,Te}$) cannot be tuned out from a trivial insulator version without a structural phase transition; in the Bi-Sb semiconductors, the topological phase transition is masked by an intervening band [the “$H$” band ($I$)] and the lack of gating control necessary for its observation.

We demonstrate the existence of a topological phase transition in TIBi(S$_1$–xSe$_x$)$_2$, which, as we show, is a fully tunable topological analog of Bi$_2$Se$_3$. By effectively varying the sulfur/seelenium ratio ($\delta$), both the spin-orbit strength and lattice parameters are effectively tuned ($25$). Because the topological order in TIBi(S$_1$–xSe$_x$)$_2$ originates from Bi and Se atoms, and to illustrate an analogy with the known topological insulator Bi$_2$Se$_3$ ($6$, $8$, $11$), we rewrite the formula as TIBi(S$_1$–xSe$_x$)$_2$. Figure 1A presents systematic photoemission measurements of electronic states that lie between a pair of time reversal–invariant points or Kramers’ points ($\bar{T}$ and $\bar{M}$) obtained for a series of compositions of the spin-orbit material BiTl(S$_1$–xSe$_x$)$_2$. As the selenium concentration is increased, the low-lying bands, separated by a gap of energy $0.15$ eV at $\delta = 0.0$, are observed to approach each other; the gap decreases to less than $0.05$ eV at $\delta = 0.4$. Both bands demarcating the gap show three-dimensional (3D) dispersion where binding energies vary with momentum perpendicular to the surface, $k_z$ [as probed by varying incident photon energy ($25$)], and roughly correspond to the expected position for the valence and conduction bands. The absence of surface states within the bulk gap suggests that the compound is topologically trivial ($v_0 = 0 = \theta$) within a composition range of $\delta = 0.0$ to $0.4$. Starting from $\delta = 0.6$, a linearly dispersive band connecting the bulk conduction and valence bands emerges, which threads across the bulk band gap. Incident photon energy modulation studies support the assignment of these Dirac-like bands as originating from the surface ($25$). Moreover, the band continua in the composition range of $\delta = 0.0$ to $\delta = 0.4$ are degenerate, whereas the Dirac-like bands at $\delta = 0.6$ and beyond are spin-polarized (see Figs. 2, 3, and 4).

The system enters a topologically nontrivial phase upon the occurrence of an electronic phase transition between $\delta = 0.4$ and $\delta = 0.6$ at temperatures below $15$ K. Although the system approaches the transition from the conventional or no-surface-state side ($\delta = 0.4$), both energy dispersion and Fermi surface mapping (Fig. 1, $A$ and $B$, for $\delta = 0.4$) show that at the outer boundary of the bulk conduction band continuum that corresponds to the loci where the Dirac surface states would eventually develop, the spectral weight becomes much more intense; however, the surface remains gapped at $\delta = 0.4$, which suggests that the material is still on the trivial side. Finer control of bulk compositional variation does not allow us to locate a precise value for the transition; this could also be a consequence of an intrinsically broader topological transition. A critical signature of a topological phase transition is that the material turns into an indirect bulk band gap material, as conjectured previously ($I$). As $\delta$ varies from $0.0$ to $1.0$ (Fig. 1C), the dispersion of the valence band evolves from a single-peak to a dual-peak shape with a “dip” at the $\bar{T}$ point ($k = 0$); the $\delta = 0.0$ compound features a direct band gap in its bulk, whereas the $\delta = 1.0$ compound has a slightly indirect gap. The overall experimental evolution of the spin-orbit ground state is shown in Fig. 2A.

Now we systematically study the end product of the transition, TIBi(S$_1$–xSe$_x$)$_2$, and explore its spin and polarization properties far away from the Dirac node where nonlinear spin-orbit terms are also important. Such terms correspond to analogs of cubic and higher-order Dresselhaus effects arising from the symmetry of the crystal potential. Without these effects, the surface states form a cone that is isotropic in momentum space ($k_x$, $k_z$) and hence generates a circular Fermi contour ($2$). Spins tangentially arranged on such a circular Fermi surface lead to a Berry’s phase of $\pi$, which is also a measure of the topological invariant and the axion angle ($v_0 = 1 = \theta/\pi$) of the system ($1$, $4$, $6$). However, the details of the symmetry and bulk crystal potential can strongly deform the surface states from that of a circle. We explored

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the Fermi contour of BiTl(S\textsubscript{0.5}Se\textsubscript{1})\textsubscript{2} over a large energy range. This sample features an almost perfectly hexagonal Fermi surface at its native (as-grown) Fermi level (binding energy $E_B = 0.01$ eV), whereas the constant energy contour reverts to isotropic circular shapes when approaching the Dirac node ($E_B = 0.25$ eV and $E_B = 0.50$ eV). A more extreme example is n-type bulk doped Bi\textsubscript{2}Te\textsubscript{3}, a topological insulator that features a highly warped concave-in snowflake-shaped Fermi surface because of its large $k_F$ and a small band gap ($\Delta\approx 25$). To understand the relationship between these Dirac cone deformations and the topological invariants and axion angles ($n_0$ and $q_0$), we used spin–angle-resolved photoemission spectroscopy (spin-ARPES) (26, 27). Such spin-resolved study is also critical because Hall transport data are now possible that rely heavily on surface state topology and spin configuration data for interpretation (15).

We have carried out spin polarization texture measurements over a large range of binding energies to capture the nonlinear regime that can be accessed by gating for transport measurements. For simplicity, we show results for the hexagonal Fermi surface above the Dirac node and one of the circular Fermi surfaces below the Dirac node of BiTl(S\textsubscript{0.5}Se\textsubscript{1})\textsubscript{2} (Fig. 2, B and E). Figure 2F shows the measured out-of-plane spin polarization ($P_z$) of cuts C and E (binding energy and momentum direction are defined in Fig. 2, D and E, respectively). No significant out-of-plane spin polarization component is observed within the experimental resolution for cuts such as these. The in-plane measurements, on the other hand, show large polarization amplitudes (Fig. 3A), suggesting that the spin texture is mostly 2D. On the basis of these data, we determine the direction of full 3D spin vectors following a two-step routine (25, 28). Because $P_z = 0$ (Fig. 2F), the out-of-plane polar angles are all close to 90°, so only in-plane azimuthal angles are shown in Fig. 3B. On the hexagonal Fermi contour located above the Dirac node (Fig. 3B), the spin vectors obtained from the polarization measurements show that the ground state features a 2D in-plane left-handed chirality spin vortex (a skyrmion in momentum space; Fig. 3, B and C). The direction of the spin is roughly perpendicular to the momentum space track that connects the $\Gamma$ point and the momentum point location of the spin on the Fermi surface, rather than being tangential to the Fermi contour, as would be expected for an ideal Dirac cone. Our data also show that the spin texture below the Dirac node is also vortex-like but features right-handed chirality. Therefore, when the system is chemically tuned through the zero carrier density (Dirac node), the chirality of the spin vortex gets inverted, as seen in the data. A systematic method of surface chemical potential tuning has been demonstrated in (6) and is also applicable here (25). However, it is not physically possible to realize this chirality inversion or chemical tuning in Bi\textsubscript{2}Te\textsubscript{3}, because the node is buried under other bands.

![Fig. 1. Topological phase transition.](http://www.sciencemag.org)
Fig. 2. Evolution of spin-orbit ground state and spin texture. (A) Energy levels of $\Gamma_n^-$ (blue circles) and $\Gamma_n^+$ (green squares) bands, obtained from ARPES measurements as a function of composition $d$. CB, conduction band; VB, valence band. Parity eigenvalues (+ or −) of Bloch states (25) are shown. The topological invariants, $v_n$, obtained from the parity eigenvalues are presented as $[0,\theta,e_n]$, where $\theta = \pi v_0$ is the axion angle and $v_n$ is the strong invariant ($\ell$–$4$). (B) Fermi surface topology evolution of BiTl(S0Se1)$_2$ across the Dirac node. The corresponding binding energies of constant-energy contours are indicated. Observed spin textures are schematically indicated by arrows. (C) Experimental scattering geometry used to measure the spin polarization components shown in (B). (D) ARPES-measured dispersion along the $\Gamma-M$ momentum space cut. The binding energies used for the cuts are as follows: $E_b$ (cuts C and D) = 0.01 eV, $E_b$ (cut E) = 0.50 eV. (E) A map of the momentum space cuts C, D, and E across the Fermi nodes. The hexagonal Fermi surface is located 0.40 eV above the spin degenerate node; the circular Fermi surface is located 0.1 eV below the spin degenerate node. (F) Measured out-of-plane spin polarization profile of cuts C and E, showing only weak modulations.

Fig. 3. Chirality inversion and spin-dependent scattering profiles for charge and spin transport. (A) Measured in-plane spin polarization profiles of cuts C, D, and E (Fig. 2). (B) Fitted in-plane azimuthal angle values of spin vectors measured along cuts C, D, and E on the ARPES-measured Fermi surfaces. Spin rotation handedness or chirality changes from left-handed (LHC) to right-handed (RHC) in passing through the Dirac node toward the higher binding energy. The binding energies are indicated at the lower left corner for the experimental Fermi surfaces presented. (C) ARPES-measured Fermi surfaces are shown with spin directions based on polarization measurements. Photon energy used for spin-resolved measurements is indicated at the upper right corners. Spin texture reveals a 1/2 Dirac gas. (D) Left panels: Spin-independent scattering profiles on Fermi surfaces in (C); right panels: Spin-dependent scattering profiles on Fermi surfaces in (C) relevant for surface quasi-particle transport, which is sampled by the quasi-particle interference (QPI) modes.
The observed chirality inversion of the surface spin texture indicates a 180° turnaround of the spin momentum–locking profile in moving chemical potential across the Dirac node. In the present experiment, this is equivalent to measuring the profile above and below the Dirac node. The profiles above and below the Dirac node are shown in (A) and (B), respectively. (C) A capacitor-like device geometry, using topological insulator thin films of opposite chirality (LHC and RHC) composite, is predicted to host exotic exciton properties where quasi-particle excitation carries fractional charge. (D) Spin texture evolution of topological surface bands as a function of energy away from the Dirac node (left axis) and geometrical warping factor ω (right axis). The warping factor is defined as

\[ \omega = \frac{k_x (\tau - \nu) - k_y (\tau + \nu)}{k_x (\tau - \nu) + k_y (\tau + \nu)} \]

where \( \omega = 0, \omega = 1, \) and \( \omega > 1 \) imply circular, hexagonal, and snowflake-shaped Fermi surfaces, respectively. The sign of \( \omega \) indicates texture chirality for LHC (+) or RHC (–). The insets show out-of-plane 3D spin polarization measurements at corresponding Fermi surfaces.

**References and Notes**

1. M. Z. Hasan, C. L. Kane, Rev. Mod. Phys. 82, 3045 (2010).
25. See supporting material on Science Online.
The origin of superconductivity in the iron pnictides has been attributed to antiferromagnetic spin-ordering transition at $T_N$ and a tetragonal-to-orthorhombic structural transition at $T_S$ (2, 3). Both $T_N$ and $T_S$ decrease in a similar fashion upon ion substitution and lead to an emergent dome-shaped superconducting (SC) phase with a high $T_c$ of up to 55 K (4). Such high $T_S$s are not expected from a conventional electron pairing through lattice vibrations (5). Focusing on the nesting tendency connecting quasi-cylindrical Fermi surfaces (FSs), electron pairing mediated by the AF spin fluctuations has been argued (6, 7).

On the other hand, there is a proposal that orbital ordering (8–10), which causes inequivalence of the Fe 3d partial density of states, occurs at $T_S$. This phenomenon could give rise to C2 symmetric physical properties that cannot be expected from only 1% lattice distortion at $T_S$. In particular, inequivalent occupation of the $xz$ and $yz$ orbitals (x, y, and z represent orthorhombic axes) near the Fermi level ($E_F$), as observed by angle-resolved photoemission spectroscopy (ARPES), indicates the potential role of orbital degrees of freedom (11). Based on the multiorbital nature of iron pnictides, SC pairing through orbital fluctuations has been proposed (12–15).

Experimental evidence has favored different SC pairing symmetries. Sign-reversal superconductivity suggested by scanning tunneling microscopy on FeTeSe (16) and inelastic neutron spectroscopy on (Ba,K)Fe$_2$As$_2$ (17) is consistent with the $s^\pm$ wave symmetry expected from spin fluctuations. The robustness of $T_c$ against the impurities in LaFeAsO (18) suggests the $s^\pm$ wave superconductivity, which may be caused by orbital fluctuations. Such material dependence in pairing symmetry can also arise from the balance of these two pairing mechanisms because a crossover from $s^\pm$ to $s^\mp$ symmetry may occur, depending on the strengths of microscopic electronic parameters (12–15). However, to date there has been no experimental evidence for the role of orbital fluctuations on the superconductivity in the pnictides.

The pairing mechanism will be reflected in the momentum dependence of the SC gap properties. Spin fluctuation mechanism predicts strongly orbital-dependent SC gap magnitude (7, 19), but orbital fluctuations should eliminate such orbital dependence (13–15). Because each FS sheet in iron pnictides has a distinct d-orbital character, probing the FS dependence of the SC gap magnitude will be a crucial test for identifying the contribution of the orbital fluctuations in the electron pairing. Here, we report the FS dependence of the SC gap magnitude of representative iron-pnictide superconductors BaFe$_2$(As$_{0.65}$P$_{0.35}$)$_2$ (AsP122) and Ba$_0.6$K$_{0.4}$Fe$_2$As$_2$ (BaK122) by using bulk-sensitive laser-ARPES (20) [see section 1 of the supporting online material (SOM)].

ARPES, using a fixed photon energy ($h\nu$), probes a specific profile of the in-plane FSs perpendicular to the momentum axis along the z direction $k_z$. Especially for AsP122 with strong FS warping, the orbital character depends on the $k_z$ value (21). Estimation of $k_z$ value corresponding to $h\nu = 7$ eV photons of laser-ARPES is thus necessary for investigating the orbital dependence of the SC gap magnitude. Figure 1B is a schematic of the experimental FS profile along the $k_z$ axis of AsP122 taken from (22). Whereas the inner hole FS is almost cylindrical, the outer one gets enlarged toward $k_z = \pi$, consistent with the band calculations (inset of Fig. 1A) (21). We used a synchrotron radiation photon source of 10 eV and 7 eV to find this characteristic warping in the inner ($k_z^1$) and outer ($k_z^2$) Fermi momenta. As shown in Fig. 1, C and D, separation between $k_z^1$ and $k_z^2$ in C was three-fourths of that in D, which indicates that 7 eV photons probe the FS profile at $k_z \sim \pi$ to 1.2$\pi$. This estimate of $k_z$ for $h\nu = 7$ eV was also quantitatively confirmed by the comparable FS size obtained by the higher photon energy of 63 eV probing $k_z \sim \pi$ to 2.2$\pi$ (Fig. 1E).

High-resolution laser-ARPES $E$-$k$ images for AsP122 (Fig. 2, B and C) are along cut 1 and 2 indicated in Fig. 2A, respectively. Both the inner and middle hole bands were resolved as the two-peak feature in the momentum distribution curve (MDC) near $E_F$ (Fig. 2B), although they are nearly degenerate in Fig. 1, C and D. We could separate three hole bands along cut 1 and 2. The temperature ($T$) dependence of the energy distribution curves (EDCs) at $k_z$ in cut 2 (outer FS) shows the SC gap opening below bulk $T_c$ (Fig. 2D), and the SC gap magnitude $\Delta$ was directly extracted from the fitting procedure using a Bardeen-Cooper-Schrieffer (BCS) spectral function (SOM, section 2). This $\Delta$ value is in excell-