Observation of Zeeman effect in topological surface state with distinct material dependence

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Manipulating the spins of the topological surface states represents an essential step towards exploring the exotic quantum states emerging from the time reversal symmetry breaking via magnetic doping or external magnetic fields. The latter case relies on the Zeeman effect and thereby we need to estimate the g-factor of the topological surface state precisely. Here, we report the direct observations of the Zeeman effect at the surfaces of Bi2Se3 and Sb2Te2Se by spectroscopic-imaging scanning tunnelling microscopy. The Zeeman shift of the zero mode Landau level is identified unambiguously by appropriately excluding the extrinsic effects arising from the nonlinearity in the band dispersion of the topological surface state and the spatially varying potential. Surprisingly, the g-factors of the topological surface states in Bi2Se3 and Sb2Te2Se are very different (±18 and −6, respectively). Such remarkable material dependence opens up a new route to control the spins of the topological surface states.
The helical Dirac fermions on the surfaces of topological insulators (TIs) host novel relativistic quantum phenomena in solids. When the time reversal symmetry (TRS) of the topological surface state (TSS) is broken, a gap opens at the Dirac point. This brings about novel topological excitations, such as the magneto-electric effect and the quantum anomalous Hall effect, and the magnetic monopole effect. Magnetic doping has proven to be an effective way to break the TRS via magnetic exchange interactions, thereby enabling the experimental observation of the quantum anomalous Hall state. However, magnetic dopants may introduce charge inhomogeneity and weaken the spin–orbit coupling of TI compounds.

The Zeeman effect, the coupling of spins with a magnetic field, offers an alternative way to break the TRS of the TSS without changing its slope. In graphene, the Zeeman splitting of the LLs observed in graphene and conventional two-dimensional (2D) electron systems. Such a Zeeman effect is most pronounced for the zeroth LL (LL0). The Zeeman effect, the coupling of spins with a magnetic field, is crucial to know its g-factor.

Under B, Landau levels (LLs) are formed as a result of the cyclotron motion of the electrons. Inclusion of the Zeeman effect can lead to an energetic shift of the LLs as the corresponding TSS lack spin degeneracy. Such a Zeeman effect is most pronounced for the zeroth LL (LL0), and decreases dramatically with increasing Landau index n (Supplementary Note 1).

Understanding the Zeeman shift behaviour and determining the g-factor of the TSS have entailed intense research investigations primarily by the means of quantum oscillation measurements. However, the reported data are still largely controversial. The inconsistency is due to the fact that the host materials are inevitably doped by defects. This prevents the lower LLs, which exhibit prominent Zeeman shifts, from reaching the Fermi level and contributing to quantum oscillations in the accessible B (ref. 16). Furthermore, the sign of g0 cannot be determined from the Zeeman shift of the nonzero LLs. A recent tunnelling spectroscopy study appears to be successfully probing the LLs of the TSS formed at an interface with a conventional semiconductor. However, the method used depends on specific details of band-bending in such heterostructures, and therefore cannot be readily applied to other TI compounds. Spectroscopic imaging scanning tunnelling microscopy (SI-STM) can access electronic states in a wide energy range with high spatial and energy resolutions. Thus, it can be used to study any LL, regardless of the doping level of the TI compounds.

Using this technique, we have developed a methodology to unambiguously observe the Zeeman shift of LL0. This allows us to determine the g-factors of the TSSs in Bi2Se3 and Sb2Te2Se precisely, which turn out to be very different.

**Results**

**Modelling the Zeeman effect of the TSS.** In principle, the Zeeman shift of the LL0 energy (E0) of the TSS is linear with B, and its slope determines g0. In practice, however, more factors are involved, hindering its direct observation. On one hand, a finite curvature is superimposed on the linear dispersion of the TSS in actual compounds. This induces an extra B-linear change in E0 that is irrelevant to the Zeeman effect. On the other hand, there exists spatial potential variations in the TSS originating from the inhomogeneously distributed charged defects. This introduces an extrinsic B dependence of LL energies as the spatial extension of the LL wave functions shrinks with increasing B.

To quantify the effects of nonlinear dispersions and the potential variations, we consider the following model Hamiltonian.

\[
H = \frac{1}{2m^*m_e} (\Pi_x^2 + \Pi_y^2) + \nu (\sigma_x \Pi_x - \sigma_y \Pi_y) + \frac{1}{2} g_0 \mu_B B \sigma_z + V(x,y)
\]

Here, \(\sigma\) are the Pauli matrices and \(\Pi\) are the canonical momenta. The first term denotes the nonlinearity of band dispersions, with \(m^*\) being the effective mass relative to that of the free electron (\(m_e\)). The second term represents the helical Dirac component of the TSS, with \(\nu\) being the electron velocity. The third term is the Zeeman term and the last term represents the potential variation. The energy of the \(n\)-th LL, \(E_n\), has been given in ref. 20, albeit without contribution from the last term of equation (1).

To include the effect of this term, we consider a 2D parabolic potential model \(V(x,y) = E_0 + \alpha_x x^2 + \alpha_y y^2\) to approximate the shape and location of the potential extremes, where \(E_0\) is the Dirac-point energy. At the potential extreme, the \(B\)-dependence of \(E_0\) can be calculated using a first-order approximation (Supplementary Note 1). Including the effects of nonlinear dispersions and potential variations, the \(E_0(B)\) is accordingly given as

\[
E_0(B) = E_0 + \frac{1}{2} \left( \frac{2}{m^*} - g_0 \right) \mu_B B + \left( \alpha_x + \alpha_y \right) \frac{\hbar}{|e|B}
\]

Note that \(m^*\) renormalizes \(g_0\), and the potential variation introduces an additional \(1/B\) dependence term. To determine the intrinsic \(g_0\), we have developed an analysis scheme for our SI-STM data, which corrects these extrinsic factors, \(m^*\) and \(\alpha_x + \alpha_y\), and applied it to two different TI materials, Bi2Se3 and Sb2Te2Se.

**SI-STM study of the Zeeman effect of the TSS.** As the first step, we evaluate \(m^*\) using momentum-resolved LL spectroscopy.
Figure 2a,b display the LL spectra of Bi$_2$Se$_3$ and Sb$_2$Te$_2$Se (Supplementary Note 2, Supplementary Figs 1 and 2), respectively, as measured at a fixed location at various $B$. In contrast to the electron-doped Bi$_2$Se$_3$, Sb$_2$Te$_2$Se is hole-doped and thus its Dirac point is in the empty state. For each material, the corresponding $E_n$ exhibits a quasi-linear scaling relation with a varying scaling variable $(nB)^{1/2}$ (Fig. 2c), which represents the energy–momentum dispersion of the TSS$^{22}$. The potential effect on $E_n$ is more significant for the LLs with small $n$ exposed to high $B$ than those with large $n$ exposed to low $B$ (ref. 21). In this regard, the observed scaling with $(nB)^{1/2}$ demonstrates that such potential effect has negligible influence on it at the measured location. For both compounds, a finite curvature is evidently seen in the dispersions indicating that $m^*$ is finite, as also demonstrated by angle-resolved photoemission spectroscopy measurements and band calculations$^{2,23}$. Remarkably, both compounds show a very

**Figure 2** | LL spectroscopy of Bi$_2$Se$_3$ and Sb$_2$Te$_2$Se. Tunnelling spectroscopy showing the LLs of the TSS measured at a fixed location of (a) Bi$_2$Se$_3$ and (b) Sb$_2$Te$_2$Se surface at 1.5 K. The spectra were acquired at different $B$ from 0 T to 11 T with an interval of 1 T and are offset vertically for clarity. The red lines mark the energies of the Dirac points. The data shown in a are the same data used in ref. 22. Measurement conditions of b: $V_i = -100$ mV, $I_l = 50$ pA and $V_{	ext{mod}} = 1.4$ mV$_{\text{rms}}$. (c) Scaling analysis of $E_n(B)$ based on the data of Sb$_2$Te$_2$Se shown in b and a comparison with that of Bi$_2$Se$_3$. The $E_n$ values were obtained by fitting the LL spectra with multiple Lorentz functions. The black curves depict the fitting to the low-energy parts of the scaling relations.

**Figure 3** | Surface $g$-factor measurement on Bi$_2$Se$_3$. (a) Potential landscape of Bi$_2$Se$_3$ obtained by mapping $E_0$ at 11 T showing a potential minimum. The centre of the potential minimum is determined by the 2D parabolic potential fitting and is marked as a cross. The scale bar corresponds to 10 nm. Measurement conditions: $V_i = 50$ mV, $I_l = 50$ pA, $V_{\text{mod}} = 2.8$ mV$_{\text{rms}}$ and $T = 1.5$ K. (b) Tunnelling spectra taken at the potential minimum centre at fields from 3 to 11 T with 1 T intervals. The spectra have been shifted for clarity. Measurement conditions: $V_i = -220$ mV, $I_l = 100$ pA, $V_{\text{mod}} = 1.4$ mV$_{\text{rms}}$ and $T = 1.5$ K. (c) $E_0$ at different $B$ obtained by fitting the data of b with a Lorentzian line shape and plotted with black symbols (left axis). The error bars are the standard deviation of the fitting analysis. The effects of the potential and the non-ideal dispersions on the LL$_0$ energies are represented by $E_V = (x_s + x_h) \frac{B}{B_0}$ and $E_W = \frac{E^2}{\Delta B}$, respectively. Their influences can be excluded by subtracting their contributions. The red symbols denote the LL$_0$ energies after subtracting the effect of the potential ($E_0 - E_V$). The blue symbols correspond to the LL$_0$ energies after subtracting the effects from both the potential and the non-ideal dispersions ($E_0 - E_V - E_W$) (right axis, in the relative energy scale). The black curve denotes the fitting to $E_0$ with $B$ according to equation (2). The red and blue lines show the linear fitting of $E_0 - E_V$ and $E_0 - E_V - E_W$ with $B$, respectively. (d-f) Similar data and analysis as a-c for a potential maximum of Bi$_2$Se$_3$. Measurement conditions of d and e: $V_i = -200$ mV, $I_l = 165$ pA, $V_{\text{mod}} = 1.8$ mV$_{\text{rms}}$ and $T = 1.5$ K.
similar band curvature, despite their different constituent elements. We thus expect $m'$ to be nearly the same for both the compounds. To evaluate this, $m'$ is extracted from the scaling function, while ignoring the potential variations. For each compound, we have performed three measurements at different locations/samples. On the basis of these measurements, $m'$ is found to be $0.12 \pm 0.03$ for Bi$_2$Se$_3$ and $0.13 \pm 0.02$ for Sb$_2$Te$_2$Se with negligible inhomogeneity.

Next, we assess the impact of potential variations on $E_0(B)$. The spatial variation of $E_0$ represents the potential landscape, although it is smeared out by the magnetic length $l_B$ (refs 19,21). We start with Bi$_2$Se$_3$ and map out the potential landscape by performing a spectroscopic imaging of $E_0$ at a high $B$ of 11 T, where $l_B \sim 7.7$ nm (Fig. 3a,d). We focus on areas around the potential extremes (Fig. 3a,d) and fit their shapes with the 2D parabolic potential model introduced above (Supplementary Fig. 3). After positioning the tip at the fitted potential centre (Fig. 3a,d, cross point), the $B$ dependence of the LL$_0$ peak is measured (Fig. 3b,e). The measured $E_0(B)$ is plotted in Fig. 3c (left axis). The error bars are the standard deviation of the fitting analysis. The effects of the potential and the non-ideal dispersions on the LL$_0$ energies are represented by $E_{\text{g-factors}}$ (Fig. 3c,f, blue symbols). Subsequently, $g_s$ of Bi$_2$Se$_3$ is determined to be $+18 \pm 4$. (The error in $g_s$ is the propagation of uncertainty evaluated from the errors of $m'$ and $g_m$, which are extracted above and measured at different potential extremes, respectively.) This differs significantly from its corresponding bulk value $+32$ as indicated by magneto-transport and nuclear magnetic resonance measurements$^{22,23}$.

The same methodology is applied to Sb$_2$Te$_2$Se (Fig. 4; Supplementary Figs 4 and 5). Distinct from Bi$_2$Se$_3$, the LL$_0$ state measured at the potential minimum (Fig. 4b) exhibits a nonmonotonic $B$ dependence (Fig. 4c) and increases with $B$ at high fields. Given that the effective masses of both compounds are almost the same, $g_s$ in Sb$_2$Te$_2$Se must differ significantly from that of Bi$_2$Se$_3$. Indeed, we evaluate $g_s$ in Sb$_2$Te$_2$Se to be $-6 \pm 2$. While both systems share very similar band dispersions in their TSSs, their $g$-factors turn out to be strikingly different in both size and sign.

**Discussion**

We elucidate the above results under the framework of $k \cdot p$ theory. For narrow gap semiconductors, the conduction bands are coupled to the spin–orbit-split valence bands through a second-order perturbative term. Such a coupling substantially enhances the orbital sector of the $g$-factor. As a result, the total $g$-factor of such electrons/holes can significantly deviate from that of the free electrons in both magnitude and sign$^{26}$. In the case of TIs, the strong atomic spin–orbit coupling creates a symmetry-inverted band gap. Thus, the atomic orbital characters of the wave function undergo a strong variation at and in the

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**Figure 4 | Surface g-factor measurement on Sb$_2$Te$_2$Se.** (a) Potential landscape of Sb$_2$Te$_2$Se obtained by mapping $E_0$ at 12 T showing a potential minimum. The centre of the potential minimum is determined by the 2D parabolic potential fitting and marked by a cross. The scale bar corresponds to 10 nm. (b) Tunnelling spectra taken at the potential minimum centre at fields from 3 to 12 T with 1 T intervals. The spectra have been shifted for clarity. (c) $E_0$ values at different $B$ obtained by fitting the data of b with a Lorentz line shape and plotted with black symbols (left axis). The error bars are the standard deviation of the fitting analysis. The effects of the potential and the non-ideal dispersions on the LL$_0$ energies are represented by $E'_{\text{g-factors}}$ (right axis, in the relative energy scale). The black curve denotes the fitting to $E_0$ with $B$ according to equation (2). The red and blue lines represent the linear fitting of $E_0 - E'_{\text{g-factors}}$ and $E_0 - E'_{\text{g-factors}}$ with $B$, respectively. (d-f) Similar data and analysis as a-c for a potential maximum of Sb$_2$Te$_2$Se. Measurement conditions of a and d: $V_x = 215$ mV, $I_x = 50$ pA, $V_{\text{mod}} = 2.8$ mV, $T = 4.4$ K and $B = 12$ T. Measurement conditions of b and e: $V_x = 210$ mV, $I_x = 50$ pA, $V_{\text{mod}} = 1.8$ mV, and $T = 4.4$ K.
vicinity of the inverted band edges. Such a variation manifests itself in the orbital character of TSS around the Dirac point as well. As $\text{Sb}_2\text{Te}_3\text{Se}$ and $\text{Bi}_2\text{Se}_3$ comprise different elements, their wave functions must have different orbital characters. Such a difference further implies that the $g$-factors of the two compounds should be different too. Note that a considerable energy dependence of the $g$-factor is expected, since the orbital character of the wave function changes notably in these systems. Our measurement is merely around the Dirac point, which is directly relevant to the gap opening of the TSS via $B$. In this regard, the $g$-factor of the TSS may be different from that of the bulk since they are measured at different energies. Further theoretical investigations regarding those factors are needed to develop a general theory that describes the $g$-factor.

Considering the significant material dependence of $g_0$, we envision an interesting possibility to tailor the $g$-factor of the TSS by controlling the chemical composition of the chalcogenide TI materials in the form of solid solutions. This provides a new knob in manipulating the TSS for its spin-related applications.

Methods

Evaluation of the finite curvature of LL scaling. We evaluated $m^*$, which characterizes the finite curvature, using the scaling analysis of the LL energies shown in Fig. 2c. Since only the Zeeman shift of the LL$_0$ is prominent, Supplementary equation (10) (Supplementary Note 1) can be approximated by $E_n = 0.5B |n|B + \frac{\hbar^2v_n^2}{2m^*}|n|B$. This indicates that the scaling of $E_n$ with $(nB)^{1/2}$ still applies even in the case of finite $m^*$. Regarding $E_n^*$, its energy at 3 T was used for the scaling analysis, because its shift is negligible at low $B$. By fitting the low-energy part of the scaling relation, the $m^*$ value can be obtained. It must be noted that the low-energy fitting considerably deteriorated when high-energy data were also included. Since the effect of $m^*$ on $E_n^*$ is determined by the electronic states around the Dirac point, we only fitted the low-energy parts.

Fitting the potential extremes with 2D parabolic potential model. First, two sections across the potential extremes were drawn and fitted with a one-dimensional parabolic potential to estimate its shape and location. The obtained parameters were subsequently input as the initial guess of the 2D parabolic fitting. The 2D fitting provided the shape and location of the fitted potential, whose equipotential lines were superimposed on the potential map. The generated fitting error was small, demonstrating that the 2D parabolic potential model fitted the measured potential well. This conclusion was further augmented by evaluating two sectional lines extracted along the major eclipse axes of both the fitted potential and the measured $E_n$ map (Supplementary Figs 3c-f and 4c-f). Since the 2D parabolic potential merely applies to potential extremes, it cannot be used at low $B$, where the LL$_0$ state spatially expands beyond the potential extremes. For example, the effect of the potential variations at low $B$ is discussed in Supplementary Note 3, Supplementary Figs 6 and 7. Therefore, for the fitting of Figs 3c-f and 4c-f, we did not include the data below the critical $B$ value, where the potential-corrected $E_n$ begins to deviate from the $B$-linear behaviour. The size of the LL$_0$ state [2(B)] at the critical $B$ value, where the potential-corrected $E_n$ begins to deviate from the $B$-linear behaviour. The size of the LL$_0$ state [2(B)] at the critical $B$ value, where the potential-corrected $E_n$ begins to deviate from the $B$-linear behaviour.
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