Determination of the spin axis in quantum spin Hall insulator candidate monolayer WTe_2

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ABSTRACT

Evidence for the quantum spin Hall effect (QSHE) has been reported in several experimental systems, in the form of edge conductance approaching a quantized value at zero magnetic field. However, another fundamental feature of the QSHE, that of spin-momentum locking in the edge channel, has not been demonstrated. Here, we report that in an applied magnetic field the edge conductance in monolayer WTe_2, a candidate QSHE material, is suppressed by the component of the field perpendicular to a particular axis, implying that the spin in the edge states lies along this axis. Surprisingly, the axis is independent of edge orientation, chemical potential and sample, being fixed relative to the monolayer crystal structure, lying at $40 \pm 2°$ to the layer normal within the mirror plane. This finding is consistent with a theoretical model in which the bulk bands nearest the Fermi energy have the same parity, leading to simple spin texture with a single, momentum-independent spin axis that is inherited by the edge modes. In addition, the results strengthen the case for spin-momentum locking, and therefore that monolayer WTe_2 is a natural two-dimensional topological insulator exhibiting the QSHE.

I. INTRODUCTION

The two-dimensional topological insulator (2D TI) is the original and archetypal example of a topologically nontrivial electronic system with time-reversal symmetry [1-5]. The topology of the electronic states endows a 2D TI with gapless one-dimensional edge modes which are helical. In helical modes the electron's spin is locked to its momentum, pointing in a particular direction for electrons moving one way along the edge and in the opposite direction for electrons moving the other way, and, as a result, elastic backscattering in zero magnetic field is prohibited. In view of this, a single-edge quantized conductance of e^2/h in zero field provides evidence for the QSHE, e being the electron charge and h Planck's constant. However, inelastic and magnetic backscattering are still allowed [6-10] and the quantization is not expected to be accurate, unlike in the quantum Hall effect. Indeed, the leading 2D TI candidate materials, HgCdTe [11] and InAs/GaSb quantum wells [12] and monolayer (1L) WTe_2 [13,14], do not exhibit well quantized conductance. Meanwhile, the other essential feature of the QSHE, that of spin-momentum locking in the edges, has not been experimentally demonstrated.

The natural way to determine the spin axis in a helical mode is to apply a magnetic field $\boldsymbol{B}$ and vary its orientation. Due to mixing of the two opposite-spin branches by the Zeeman coupling term, the conductance should decrease as a result of induced backscattering except when $\boldsymbol{B}$ is perfectly aligned with the spin axis. In the quantum well systems, strong orbital coupling to $\boldsymbol{B}$ unfortunately masks this effect [11]. Here, we

report that in 1L WTe_2 the edge conduction, in both linear and nonlinear response, does exhibit the expected anisotropic dependence on $\boldsymbol{B}$ with respect to a particular axis, which we call $\boldsymbol{d_{so}}$. Importantly, this spin axis is found to be independent of edge orientation, gate voltage, and sample, being fixed relative to the crystal structure. This in turn strongly suggests that the spin-orbit coupling (SOC) is a momentum-independent property of the bulk bands. Our conclusion that the edge modes and the conduction and valence band edges share a single spin axis is illustrated schematically in Fig. 1(a). Fig. 1(b) shows the $1T'$ crystal structure of 1L WTe_2, indicating our choice of axes and the three spatial symmetries: a 2-fold screw axis (C_{2x}) along the tungsten chains; a mirror plane (M_x) perpendicular to the chains (the *y*-*z* plane); and inversion (parity $\mathcal{P}$). The spin axis $\boldsymbol{d_{so}}$ is in the mirror plane, making an angle $\varphi_{so} = 40 \pm 2°$ with the normal *z*-axis.

To interpret this finding theoretically, we consider the low-energy effective models of 1L WTe_2. Within a four-band model based on two orbitals and two spin states, two distinct choices of orbital symmetries are possible. In one [5,15-18], the orbitals are taken to have opposite $\mathcal{P}$ symmetry and the leading-order SOC is momentum-dependent; in the other [19-22], the orbitals have the same $\mathcal{P}$ symmetry and the SOC is then momentum-independent and strictly in the mirror plane. Only in the latter case are the states on arbitrary edges naturally expected to directly inherit the same, unique, spin axis as the bulk, and thus this model is more compatible with our experimental findings, as will be discussed further below.

II. RESULTS

Figure 2 illustrates how the conductance depends on the orientation of $\boldsymbol{B}$ for device MW5. An optical image is shown in Fig. 2(a). The monolayer WTe_2 flake overlies six Pt contacts, encapsulated between hexagonal boron nitride dielectric layers with a graphite gate electrode beneath. Polarized Raman spectroscopy (inset to Fig. 2(a)) was used to determine the orientation of the crystal *x*-axis (see fig. S3) [23]. Here the temperature and gate voltage ($T = 4$ K, $V_g = -2.7$ V) were chosen such that the bulk is insulating and edge conduction dominates [13,14]. Importantly, MW5 has no top gate, allowing microwave impedance microscopy (MIM) to be used to map the in-plane conductivity [24] and thereby establish the precise geography of the edges (Fig. 2(b)) as well as any internal cracks which also conduct. This allows us to investigate the dependence on the direction of the edge relative to the crystal axes.

The sketch in Fig. 2(c) shows the coordinates we use for the field direction: a polar angle θ relative to the *x*-axis and an azimuthal angle φ about this axis, with $\varphi = 0$ along the *z*-axis. Below is a polar plot of the linear conductance G between contacts 1 and 2 versus θ at selected values of φ. We let θ run from $0°$ to $360°$ to parameterize rotating $\boldsymbol{B}$ in a full circle (shaded purple in the sketch). Onsager symmetry requires $G(\boldsymbol{B}) = G(-\boldsymbol{B})$, so the equivalence of the data for θ and $\theta + 180°$ serves as a check on the experiment. The conductance is maximal in a particular direction, $\boldsymbol{d_{so}}$, which is at $\varphi = \varphi_{so} = 38°$ and in the *y*-*z* plane ($\theta = 90°$). In the plane perpendicular to $\boldsymbol{d_{so}}$, specified by $\varphi = 128°$, the conductance is much smaller and relatively independent of θ (red trace). There is a high degree of symmetry about the *y*-*z* (mirror) plane. Figure 2(d) shows the dependence on φ within the *y*-*z* plane for the same contact pair as well as for three others. For every pair the conductance maximum is within a degree of $38°$ (dashed line), even though the edges connecting them have different shapes and orientations with respect to crystal axes (see Figs. 2(a),(b)). Across five different monolayer devices all contact pairs showed similar behavior, with φ_{so} in the range $40 \pm 2°$ (see figs. S4 to S6), implying that orientation and shape of the edge has almost no effect on the magnetic anisotropy of its conductance. Devices made from bilayer WTe_2, which is topologically trivial, did not show any of this anisotropy [25] (Fig. S13).

Figure 3(a) shows the conductance measured as $\boldsymbol{B}$ is rotated in the *y*-*z* plane for a series of temperatures. At $T = 140$ K, bulk conduction dominates and there is no effect of magnetic field. On cooling below about 100 K, as the bulk conduction freezes out, G depends increasingly strongly on φ. Figure 3(b) compares the conductance for $\varphi = \varphi_{so}$, where the temperature dependence is weakest, with that for $\varphi = \varphi_{so} + 90°$. Figure 3(c) shows how G depends on the field strength at 4 K: it drops much faster for $\varphi = \varphi_{so} + 90°$ than for $\varphi = \varphi_{so}$. Figure 3(d) shows that φ_{so} is independent of B, and Fig. 3(e) shows that it is almost independent of V_g, varying by less than $2°$ over the full range (Fig. S7).

III. DISCUSSION

Since it is not yet established what mechanisms control the resistivity and magnetoresistance of helical quantum wires (see Refs [6-10] and references therein), we do not attempt to analyze the dependence of G on temperature or field magnitude, but rather focus on the existence of $\boldsymbol{d_{so}}$ and its interpretation as the spin axis. It should first be noted that, strictly speaking, the minimum mixing of opposite-spin branches (and thus magnetoresistance) is expected when the direction of $\vec{\vec{g}}\boldsymbol{B}$, not $\boldsymbol{B}$, coincides with the spin axis. For a generic edge, the g-tensor $\vec{\vec{g}}$ is not constrained by any symmetry and furthermore can depend on details of the edge. This could explain why the angular separation of the minima and maxima in G is not exactly 90° but ranges down to 83° (in Fig. 3(a), for example, it is 85°). The lack of variation of $\boldsymbol{d}_{so}$ implies either that $\vec{\vec{g}}$ is nearly isotropic or, at least, that $\boldsymbol{d_{so}}$ remains close to a principal axis of $\vec{\vec{g}}$ irrespective of the edge structure.

As mentioned in the introduction, a minimal model of the low-energy physics in bulk monolayer WTe_2 has four bands, built from two orbitals (the bottom of the conduction band and the top of the valence band at Γ) and two spin states. Each orbital must have definite symmetry (even or odd) with respect to $\mathcal{P}$, M_x, and C_{2x}. Two distinct choices are possible for the symmetries. With either choice, the allowed SOC is such that the bulk spin texture near the band edge is described by a canted spin axis near the mirror plane [18]. However, the form of the SOC is quite different in the two cases. In one [5,15-18], the orbitals are assigned opposite $\mathcal{P}$ and the same M_x symmetry (implying opposite C_{2x} symmetry), so that the matrix elements of the $\boldsymbol{k}\cdot\boldsymbol{p}$ Hamiltonian that connect them must be even under M_x and time-reversal $\mathcal{T}$ and odd under $\mathcal{P}$ (and C_{2x}). The leading-order SOC then has the form $H_{SOC}^{\mathrm{I}} = \tau_x(v_x p_y \sigma_x + v_y p_x \sigma_y + v_z p_x \sigma_z)$, where the Pauli matrices τ_i (σ_i) act in the orbital (spin) space, and coefficients v_i have dimensions of velocity. In the other case [19-22], the orbitals are assigned the same $\mathcal{P}$ but opposite M_x (and C_{2x}) symmetry, so that the matrix elements must be even under $\mathcal{P}$ and $\mathcal{T}$ and odd under M_x (and C_{2x}). The leading-order SOC then has the form $H_{SOC}^{\mathrm{II}} = \tau_y(\lambda_y \sigma_y + \lambda_z \sigma_z)$, where coefficients λ_y and λ_z have dimensions of energy.

The edge states are built from bulk states with a range of momentum perpendicular to the edge. In the first case above (H_{SOC}^{I}), the direction of the spin is momentum-dependent, and one expects the spin axis in the edge modes to depend on edge orientation and energy and not to be in the *y*-*z* mirror plane [17]. In the second case, on the other hand, the SOC can be written in the simple form

$$H_{SOC}^{\mathrm{II}} = \tau_y \boldsymbol{D}.\boldsymbol{\sigma}\ ,$$

where $\boldsymbol{D} \equiv (0, \lambda_y, \lambda_z)$ is a constant vector in the *y*-*z* mirror plane making an angle of $\varphi_D = \tan^{-1}|\lambda_y/\lambda_z|$ with the *z*-axis. The equation for H_{SOC}^{II} implies that the projection of the electronic spin on the direction of $\boldsymbol{D}$ is a good quantum number. Each of the doubly-degenerate bulk bands near the Γ-point in the Brillouin zone, having opposite Chern numbers, corresponds to one of the possible values of this quantum number, either positive or negative. In turn, in the presence of a boundary, one expects edge states near E_F to simply inherit the spin quantum number of the bulk bands, and hence the spin axis given by $\boldsymbol{D}$, as long as bands far from E_F can be ignored. (The presence of confining electric fields at the edge does not change this conclusion: even though they can modify the edge dispersion, their contribution to the SOC is small compared with that of the electric fields in the atomic orbitals that govern the bulk SOC.) The edge states with opposite projections of the spin on the $\boldsymbol{D}$ axis propagate in opposite directions, as prescribed by time-reversal symmetry, and form the usual spin-momentum-locked helical pair of the edge states of the quantum spin Hall phase. While the form of the edge dispersion is dependent on a particular edge orientation, the spins of the edge states remain to parallel or antiparallel to $\boldsymbol{D}$, independent of the edge details. The form H_{SOC}^{II}, and choice of equal-parity orbitals, is therefore more consistent with our striking experimental finding that for all edges $\boldsymbol{d_{so}}$ (identified with $\boldsymbol{D}$) is in the mirror plane and independent of edge orientation.

The value of φ_D cannot be calculated accurately because the parameters in the models are not known well enough. However, we note that the form H_{SOC}^{II} can also be derived in a microscopic 8-band tight-binding model [19,22], in which it corresponds to the strongest spin-flip hopping process in the y direction along the Te^1-W bonds (drawn as lines in Fig. 1(b)). In this case the two interfering hopping paths that involve intermediate Te sites on the adjacent Te^1-W bonds give the standard Haldane-Kane-Mele [1,26] contribution to $\boldsymbol{D}$. This turns out to be perpendicular to the Te^1-W bond (see Fig. S8), in the mirror plane making an angle of 29° with the z-axis (indicated by a blue arrow in Fig. 1(b)), not too different from the measured value of φ_{so}, $40 \pm 2°$.

IV. NONLINEAR STUDIES

Helical edge modes are also expected to exhibit a characteristic nonlinear magnetotransport effect [27-29]. In the Taylor expansion of the current-voltage relation, $I(V, \boldsymbol{B}) = G(\boldsymbol{B})V + \gamma(\boldsymbol{B})V^2 + \cdots$, the coefficient γ describes nonreciprocal conduction, allowed because the edge breaks inversion symmetry. Its $\boldsymbol{B}$-odd part, $\gamma_a(\boldsymbol{B}) = [\gamma(\boldsymbol{B}) - \gamma(-\boldsymbol{B})]/2$, is sensitive to the edge spin texture. Two main mechanisms contribute to $\gamma_a(\boldsymbol{B})$ at low temperature (see SI-VII for details). In one, an exchange field proportional to the current-induced spin polarization (CISP) [30], which is directed along $\boldsymbol{d_{so}}$, adds to the applied field to modify the conductance. In the other, nonlinear dispersion and broken inversion symmetry in the edge energy spectrum together lead to lack of cancellation of quadratic-in-voltage currents carried by the two helical branches, similar to the situation on three-dimensional TI surfaces [28]. For both mechanisms, $\gamma_a(\boldsymbol{B})$ should vanish under the same condition that the suppression of G is maximal.

Motivated by this prediction, we measured γ_a by applying an a.c. bias, V_f, at frequency f and measuring the second-harmonic component, I_{2f}, of the current at a fixed phase with V_f chosen such that $|I_{2f}| \ll I_f$, the first-harmonic component. The dependence of $\gamma = 2I_{2f}/V_f^2$ on field amplitude along a particular axis at a given V_g typically has a heart-beat shape, as illustrated in the upper inset to Fig. 4 (see Figs. S9 and S10). The upper part of Fig. 4 shows an example of how γ_a varies as $\boldsymbol{B}$ is rotated in the y-z plane at several temperatures. The lower part of the figure shows $G = I_f/V_f$ measured simultaneously. As T is decreased, γ_a grows in concert with the anisotropy of G. (We note that in the bilayer device, where the anisotropy remained very small, γ_a was unmeasurably small). Just as predicted, γ_a passes through zero at a temperature-independent value of φ very close to the minimum in G, at $\sim 120°$ (blue dotted line) (see Fig. S11). In fact, the CISP mechanism alone quantitatively reproduces the full angular dependence of γ_a when B is not too small (see Fig. S12), and the single fitting parameter provides an estimate of the strength of electron-electron interactions in the helical edge.

V. CONCLUSION

Since the original theoretical prediction that 1L WTe_2 could be a topological insulator [5], it has already been established experimentally [13,14,31] that the edge modes span the full range of accessible chemical potential and have a conductance that can approach e^2/h on a length scale of 100 nm, characteristic of the QSHE. Combined with these observations, our demonstration of the spin polarization in the edge modes, as well as the behavior of γ_a characteristic of a helical mode, verifies the presence of spin-momentum locking and therefore cements the identification of 1L WTe_2 as a natural two-dimensional topological insulator. In addition, the simple spin texture we find for the edges, characterized by a single spin axis independent of edge orientation, implies that this is also the bulk spin axis and is consistent with a particular four-band model in which the band edge orbitals have the same parity. Among other things this has implications for the nature of the superconductivity observed in this system [32,33].

After this manuscript was first submitted, another report of the magnetic isotropy of the edge conduction appeared on arXiv [34]. Those authors' measurements indicate a minimal magnetoresistance for a field oriented at about 31° to the layer normal. We note, however, that the magnitude of the anisotropy in their

data is much smaller; they do not determine the mirror plane or edge orientations or investigate their relationship; they do not consider the same-parity model that matches our findings; and they do not study the nonlinear conductance.

METHODS

A. Device fabrication

hBN crystals were mechanically exfoliated onto thermally grown SiO2 on a highly doped Si substrate. The thickness of hBN flakes used as top and bottom dielectrics are listed in Table S1. For the devices which have a bottom gate, the few-layer graphite is covered by an hBN flake (bottom hBN) by using a polymer-based dry transfer technique. After dissolving the polymer, the hBN/graphite was annealed at 400 °C for 2 h. Next, Pt metal contacts were deposited at ~7 nm thickness on the lower hBN or hBN/graphite by standard e-beam lithography and metallized in an e-beam evaporator followed by acetone lift-off. Then the Pt contacts on hBN or hBN/graphite were annealed at 200 °C for 1 h. WTe2 crystals were exfoliated in a glove box (O2 and H2O concentrations < 0.5 ppm). The monolayer or bilayer WTe2 flake was picked up under another hBN flake (top hBN) or graphite/hBN stack. The stack was then put down on the Pt contacts in the glove box. Finally, another step of e-beam lithography and metallization (Au/V) was used to define wire-bounding pads connecting to the metal contacts and the graphite gate.

B. Electrical measurements

Electrical measurements were carried out in a PPMS DynaCool cryostat (Quantum Design, Inc.) with a base temperature of 2 K and magnetic field up to 14 T and an Oxford He-4 VTI cryostat with temperature down to 1.6 K and magnetic field up to 14 T. A 1mV a.c. excitation at 101 Hz was applied for linear responses. For second-harmonic responses, a 15 mV a.c. excitation at 101 Hz was applied while a 30 μF capacitor was connected in series with the device. The device in the PPMS cryostat can be rotated along two axes. One axis rotator is supplied by the PPMS cryostat and another axis rotator was bought from attocube and is mounted on the chip carrier.

C. Polarized Raman measurements

Raman spectroscopy was performed in vacuum at room temperature. He-Ne laser light at 632.8 nm was focused down to a spot size of ~2 μm by an objective at normal incidence. BragNotchTM filters were used to clean the incident and suppress the Rayleigh scattering. The reflected light was analyzed by an Andor spectrometer with a 1200 groove/mm diffraction grating. For thick WTe2, a laser power of 1 mW and an integration time of 30 seconds was adopted, while monolayer flakes were measured at 150 μW and integrated for 3 minutes. In the polarization dependence measurements (colinear geometry), a linear polarizer and a half-wave plate (HWP) were placed right before the objective, shared by the incident and reflected lights. To determine the crystal axes of monolayer flakes, we used an on-chip needle-shaped thick flake as a reference, where the tungsten chain is known to be along the needle. Based on the relative angle between the polarization patterns of the thick and monolayer flake, we can derive the crystal axes of the monolayer.

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Figure Captions:

FIG. 1. The spin axis of monolayer WTe_2. (a) Cartoon illustrating our key findings: the edge modes on monolayer WTe_2 are spin polarized along an axis $\boldsymbol{d_{so}}$ that is independent of edge orientation and shared with the bulk conduction and valence band edges, sketched here schematically near Γ. (b) Side and top views of the 1T′ monolayer structure indicating the three spatial symmetries (P, M_x and C_{2x}), and showing the direction of $\boldsymbol{d_{so}}$, which is in the *y*-*z* mirror plane making angle $\varphi_{so} = 40 \pm 2°$ with the *z*-axis. Tungsten atoms are shown blue, and the two inequivalent tellurium atoms, Te^1 and Te^2, are different shades of orange. The screw axis (red dotted line) is along the center of a zigzag tungsten chain.

FIG. 2. Sensitivity of edge conduction in monolayer WTe_2 to magnetic field orientation. (a) Optical microscope image of device MW5. The monolayer WTe_2 flake is outlined with a white dashed line. Inset: polar plot of the polarized Raman A_1 mode intensity used to determine the *x* and *y* crystal axes. (b) Image of the device obtained using microwave impedance microscopy to map the local conductivity ($T = 4$ K, $V_g = 0$, $B = 0$), revealing edges and cracks as thin bright lines. (c) Polar plot of conductance G between contacts 1-2 as the magnetic field $\boldsymbol{B}$ (of strength $B = 3$ T) is rotated in the manner indicated in the sketch above ($T = 4$ K, $V_g = -2.7$ V). The sketch also serves to define the angles θ and φ specifying the orientation of $\boldsymbol{B}$ relative to the crystal axes. (d) Polar plot of G for rotation of $\boldsymbol{B}$ in the *y*-*z* (mirror) plane, as indicated the sketch above. Here measurements are plotted for four contact pairs. For each, G is normalized by its minimum value. In all cases the axis of anisotropy (dashed line) is close to $\varphi = 38°$

FIG. 3. Dependence of the anisotropy on temperature, field strength and gate voltage. (All measurements are for contacts 1-2 in device MW5). (a) Two-terminal conductance G measured while rotating $\boldsymbol{B}$ in the *y*-*z* plane (see sketch above), at a series of temperatures ($B = 3$ T; $V_g = -2.7$ V). (b) Corresponding temperature dependence of G at fixed field orientations $\varphi = \varphi_{so}$ and $\varphi = \varphi_{so} + 90°$. (c) Effect of sweeping the field at $T = 4$ K for the same two orientations. (d) Polar plots of G versus φ for different field strengths ($V_g = -2.7$ V, $T = 4$ K). (e) Similar plots for different V_g ($B = 3$ T, $T = 4$ K). (At $V_g = +8$ V, values are divided by 3).

FIG. 4. Anisotropy of the nonlinear conductance. Simultaneous measurements of the field-antisymmetrized nonlinear coefficient γ_a (top) and linear conductance G (bottom) made as $\boldsymbol{B}$ is rotated in the *y*-*z* plane at selected temperatures (contacts 1-2 in device MW5, $V_g = -2.7$ V, a.c. bias 15 mV at f =101 Hz). The field strength $B = 0.3$ T was chosen to maximize γ. Inset: example of the variation of γ with B (see fig. S10).