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Spin memory of the topological material under strong disorder

Inna Korzhovska1, Haiming Deng1, Lukas Zhao1, Yury Deshko1, Zhiyi Chen1, Marcin Konczykowski2, Shihua Zhao1, Simone Raoux3 and Lia Krusin-Elbaum

Robustness to disorder is the defining property of any topological state. The ultimate disorder limit to topological protection are still unknown, although a number of theories predict that even in the amorphous state a quantized conductance might yet reemerge. Here we report that in strongly disordered thin films of the topological material Sb2Te3, disorder-induced spin correlations dominate transport of charge—they engender a spin memory phenomenon, generated by the nonequilibrium charge currents controlled by localized spins. We directly detect a glassy yet robust disorder-induced magnetic signal in films free of extrinsic magnetic dopants, which becomes null in a lower-disorder crystalline state. This is where large isotropic negative magnetoresistance (MR)—a hallmark of spin memory—crosses over to positive MR, first with only one e²/h quantum conduction channel, in a weakly antilocalized diffusive transport regime with a 2D scaling characteristic of the topological state. A fresh perspective revealed by our findings is that spin memory effect sets a disorder threshold to the protected topological state. It also points to new possibilities of tuning spin-dependent charge transport by disorder engineering of topological materials.

Introduction

Electronic disorder1 and elementary excitations in quantum condensed matter are fundamentally linked and it is well established that spatially fluctuating potentials tend to promote decoherence and localization of fermions, i.e., the formation of Anderson insulators2. The interplay of interactions and disorder often leads to new quantum behaviors; disorder typically boosts interparticle correlations both in charge and in spin channels, and that could either aid or suppress the motion of charge3. Spin effects related to disorder are particularly important when spin-orbit coupling (SOC) is strong4, and when spin-dependent charge transport can be electrically manipulated for uses, e.g., in spin-based electronics5.

Strong SOC is a hallmark of three-dimensional (3D) topological insulators6, where 2D gapless spin-polarized Dirac surface states are robust against backscattering. Most topological materials are known to contain a natural population of charged defects7 that do not cause a destruction of the topological Dirac states; indeed, they can be compensated8, although Dirac puddle disorders9 may remain. Under weak disorder, a coherent interference of electron waves survives disorder averaging10, and strong SOC enhances conductivity by a quantum weak antilocalization (WAL) correction related to the topological n-Berry phase2 when magnetic impurities are absent. With increasing disorder the 2D WAL channels can be outnumbered by the weak localization (WL) channels11—this is a precursor of Anderson localization2, which occurs at strong disorder. Under strong disorder, theory and numerical simulations12-14 predict an emergence of a new topological state, dubbed ‘topological Anderson insulator’, in which conductance $G_0 = e^2/h$ is quantized. Indeed, recent theoretical demonstrations of topological phases in amorphous systems15 point to promising new possibilities in engineered random landscapes. Strong disorder, however, is not trivial to install, quantify and control, and topological matter under such conditions has not yet been experimentally tested.

Results

Magnetotransport under strong disorder

The experiments were performed on 20-nm thin films of Sb2Te3, in which extreme positional disorder (amorphous state) is possible to obtain without fine-tuning the deposition parameters (Supplementary Note 1). Sb2Te3, in addition to being a canonical topological material16 in a crystalline state, is also a well known phase-change material (PCM)17 (Supplementary Fig. 1), that undergoes amorphous-to-crystalline transformation with the concurrent orders-of-magnitude resistive drop, and hence a huge

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range of disorder and its effect on the topological properties could be controllably explored.

Our first key finding is shown in Fig. 1a. The change in the longitudinal magnetoresistance $\Delta R_{xx}$ in the amorphous Sb$_2$Te$_3$ films at low magnetic fields is with a remarkable fidelity impervious to the tilt of magnetic field. It is relatively large and negative (i.e., charge transport becomes less dissipative with increasing field) up to a field $H_{\text{max}}$ (Fig. 1a and Supplementary Fig. 2). The as-deposited films are highly resistive and obey Anderson scaling $^{1}$ (Fig. 1b) consistent with electron localization (Supplementary Note 2), which can be induced either by increasing system size for fixed disorder or decreasing disorder for fixed system size. Robust in 3D, such scaling is marginal in 2D unless the system is a glassy state. We emphasize that magnetic signal crucially depends on the level of disorder; indeed, it becomes barely detectable in the crystalline phase as disorder in the same film is reduced by thermal annealing (Fig. 1d).

Direct detection of ‘glassy’ magnetic signal

To start we note that in the presence of disorder a finite population of singly occupied states below the Fermi energy $E_F$ has been discussed as long as 20 years back by Sir Neville Mott $^{2}$. Magnetic response from randomly localized spins in such ‘Fermi glass’ state was expected to be weak and, as far as we know, has never been experimentally demonstrated. So our second surprising finding was a very robust magnetic signal from the disordered Sb$_2$Te$_3$ films directly detected using a custom-designed $\mu$-Hall sensors (Fig. 1c, d), with the thin film flakes mechanically exfoliated from their substrates and transferred onto the active sensor area (“Methods”, Supplementary Fig. 3, Supplementary Note 4). We surmise that here the observed effective moment per atom, $m$, measured by a $\mu$-Hall sensor. Left: $m$ in a 0.1 T field under field-cooling (FC) and zero-field-cooling (ZFC) conditions. Right: $m$ of the same flake for three levels of disorder indexed by $T_a$. It onsets at ~200 K, saturates at low $T$, and becomes vanishingly small for $T_a > 160$ °C. e Cartoon of electron hopping to a distant empty site under flowing current, see text. The electron with spin $S$ may reach this site directly or via an intermediate site hosting localized spin $S$. Random disorder landscape induces spatial randomness in the spin $g$-factors and hence the randomness of local fields (gray arrows) controlling the precession of all spins. The indirect channel will depend on the state of $S$ during hopping attempts and the memory of this state is encoded in the transport of charge. These spin correlations are destroyed by magnetic field, and consequently the resistance is reduced.

Spin memory effect

Under strong disorder charge transport is a complex electron hopping process that at low temperatures proceeds via quantum tunneling between localized states assisted by phonons $^{10,16}$. While considerations of magnetotransport have mainly focused on the orbital effects, a recently proposed idea $^{16}$ takes note of putative nonequilibrium spin correlations in the localized regime created by the flowing current when electron hopping times $\tau$ are short relative to the spin relaxation times $\tau_s$. These time scales determine the magnetic field range over which spin-correlation-driven neg-MR (positive magnetoconductance) ought to be present.
Fig. 2 Spin memory effect in the longitudinal conductance $G_{xx}$ of Sb$_2$Te$_3$ in the VRH regime. a $R_{xx}(T)$ of a 20 nm thin Sb$_2$Te$_3$ film under strong disorder increases at low temperatures by orders of magnitude; the strength of disorder is reproducibly controlled by $T_a$. Inset: lithographically patterned Hall-bar and van der Pauw contact configurations used in measurements of $R_{xx}$. b $R_{xx}$ exponential in inverse $\gamma^*$ confirms that below $\sim$10 K (black down-arrow) charge transport is by the 3D VRH$^9$, the regime where dynamic spin correlation are expected. The color code is as in a. Red arrows in a and b point in the direction of increasing disorder. c Field dependence of $G_{xx}$ for two disorder states shown at 1.9 K. A fit (solid lines) to Eq. (1) fully reproduces the non-analytic form of $\Delta G_{xx}(H)/G_{xx}(0)$ arising from spin-memory in the VRH regime. Inset: Zoom of the data at low fields. d Disorder dependence of $t/\tau_s$, the ratio of hopping to spin-relaxation time, is found to closely correlate with the low- $T$ $R_{xx}$. e $t/\tau_s$ increases by nearly two orders of magnitude in the disorder range studied; it is a very sensitive probe of the phase change at crystallization, see inset.

The idea is illustrated in Fig. 1e. When the current is injected, an electron with spin $S$ attempting to hop to an available empty site can do it in two ways: directly or via an intermediate site occupied by a localized spin $s$. It may take several attempts for the indirect hops to succeed and the return probability will depend on history, i.e., on whether the tunneling electron can form a triplet or a singlet state with $s$. For example, in the absence of disorder a triplet state would remain so in the presence of applied magnetic field and no reduction of magnetoresistance (increase of magnetoconductance) is expected. Under strong disorder (such as shown in Fig. 1b), however, spin $g$-factors will be spatially random so that localized spins at different sites will precess incoherently and spin correlations will be destroyed by the field. Accordingly, in a simple model$^{15}$ the change in magnetoeconductance $\Delta G_{xx}$ arising from such spin correlations should follow not a power law$^{25}$ but a unique non-analytic form:

$$\Delta G_{xx}(H)/G_{xx}(0) \sim A \left[ \Gamma \left( \frac{d_s}{2} \right) \right] \sum_{l=1}^{\infty} \left( \frac{llH}{H^{**}} \right)^{d_s/2} \left( \frac{l}{\tau_s} \right)^{d_s/2}.$$

where $A = \frac{G_{xx}(H \to \infty) - G_{xx}(0)}{G_{xx}(0)}$, $d_s = 4/3$ is the spectral dimension of the percolation cluster$^{26}$ (which is the relevant dimension in the hopping process), $\Gamma$ is an index for the diffusing spin, $\Gamma \left( \frac{d_s}{2} \right)$ is the gamma function $\Gamma \left( \frac{d_s}{2} \right)$ is the limiting magnetic field range set by the hopping rate $1/\tau$ and the disorder-induced spread $\Delta g$ of spin $g$-factors.

The strongly localizing behavior we observe in the longitudinal resistance $R_{xx}$ (Fig. 2a) at low temperatures (below $\sim$10 K) follows variable range hopping (VRH) law $R_{xx}(T) = R_0 \exp \left( \frac{T}{T_a} \right)$ of Efros-Shklovskii (E-S) kind$^{27}$, see Fig. 2b. The E-S energy scale $T_a$ characteristic of the hopping process (Supplementary Fig. 1b) is tracked on decreasing disorder by a well controlled thermal annealing schedule (“Methods”); it is inversely proportional to the electron localization length$^{19,28}$ which we will show controls the $g$-factor distribution width $\Delta g$. We remark that in this regime (at low $T$) the detected magnetic moment appears ‘flat’ in temperature (Fig. 1d).

In the VRH regime, the fit of conductance to Eq. (1) for two states of disorder is illustrated in Fig. 2c. As seen in the figure, at low magnetic fields the characteristic non-analytic behavior is accurately followed; here the ratio of hopping time to spin relaxation time $t/\tau$, and the hopping field scale $H^{**}$ were used as fitting parameters (“Methods”). The fits at different disorder levels controlled by the anneals at different temperatures $T_a$ are shown in Supplementary Fig. 4. The ratio $t/\tau_s$ strongly depends on the level of disorder (Fig. 2d), with the hopping and spin relaxation rates, $1/\tau \propto H^{**}$ and $1/\tau_s \propto H_a$, in close correspondence with the disorder dependence of $R_{xx}$ all the way through crystallization transition. The hopping time $\tau$ can be independently extracted from the $E$-$S$ energy (Supplementary Fig. 5a), and, as expected for the hopping conductivity $\tau$ increases exponentially on decreasing temperature (Supplementary Fig. 5b). This allows us to consistently obtain the evolution of spin-relaxation time $\tau_s$ (Fig. 2e) and $\Delta g$ (Fig. 3a) with decreasing disorder (increasing $T_a$), and hence that of the low-field spin-relaxation scale $H_a = \frac{2}{H^{**}}$ (Supplementary Fig. 6); $H_a$ marks a crossover from the concave-up field shape associated with $\tau_s$ to concave-down behavior at higher fields, see Fig. 2c.

An intriguing question arises as to what controls the unexpectedly large (Tesla-range) field scale where neg-MR, the hallmark of dynamic spin memory, is found. For the systems with small spin-orbit coupling where $g$-factor $\sim 2$, the expected field range would be in the $10^{-4}$-$10^{-5}$ Tesla range. In theory$^{18}$, this range is set by the competition between the magnetic energy $g_m H$ and either thermal energy or the exchange $J$ between neighboring spins—it ought to be well below the competing

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effects. Here, however, with large effective $g$ value\textsuperscript{27}, $g\mu_B H/k_B \sim 20\,K$ is comparable to the spin-memory range. This brings us back to disorder-induced spin correlations. Fig. 1d shows that under extreme disorder the onset of magnetic response is rather abrupt and at a remarkably high temperature $T_a \sim 200\,K$. While the details of spin correlations in this Anderson-like-localized glassy state clearly deserve further experimental and theoretical studies, particularly with strong SOC, it is negative\textsuperscript{4}. Large fluctuations of Landé $g$-factors

Let us now consider $g$-factor fluctuations in a strongly disordered state. The $g$ distribution width naturally arising from our magnetoconductance data (Fig. 3a) within the model considered above is spectacularly wide at the highest level of disorder, $\Delta g \sim 40$; it exceeds the effective $g$ value of $\sim 30$ obtained, e.g., directly in the same topological insulator family from the electron spin resonance (ESR) experiments\textsuperscript{27}. Such large $g$ spread is uncommon but not unprecedented. Giant fluctuations of $g$-factors have been reported in, e.g., InAs nanowires where $\Delta g > |g_{\text{eff}}|$ and the effective factor $|g_{\text{eff}}|$ is also large\textsuperscript{24}. Furthermore, in semiconducting quantum wells $|g_{\text{eff}}|$ has been known to increase roughly linearly\textsuperscript{28} with quantum confinement energy $E_Q$ as $|g_{\text{eff}}| \approx g_0 + \beta E_Q$, where $\beta$ is a material-specific constant. Here we propose that in the strongly localized state (Supplementary Note 2), quantum confinement is enforced by the wells constrained by the localization length $\xi$ (Fig. 3b). In this view, a simple particle-in-a-box approximation gives $E_Q \propto \xi^{-2}$ that indeed fully scales with $\Delta g$ (Fig. 3a). The expected linear $|g_{\text{eff}}|$ vs. $E_Q$ would then set $|g_{\text{eff}}| \sim \Delta g^{-\alpha}$ with $\alpha \sim 0.1$, with $|g_{\text{eff}}|$ approaching the ESR-determined value in the unconfined state, see Fig. 3c.

Mapping spin memory phase space

Our experiments reveal how positive $\Delta G_{\text{ax}}(H)$ (neg- $MR$) evolves with decreasing disorder; it persists over a spectacularly large disorder range, all the way through the crystallization process and beyond (Fig. 4a). The large limiting field range set by $H_{\text{max}}$ at strong disorder falls with increasing $T_a$ (Fig. 4b) to pin off $\Delta G_{\text{ax}}(H)$ eventually to null. A clear visual of the field-disorder phase space is shown in Fig. 4c, where the strength of disorder is represented by $E_Q, H \sim E_Q$ diagram shows that spin-memory region is restricted by $H^*$ to relatively low fields, but when the system is less localized the ‘envelope’ of the spin-memory space switches to $H_{\text{max}}$.

The temperature range over which spin memory is evident is set by the VRH process (Fig. 2b), with $\Delta G_{\text{ax}}(H)$ well described by Eq. (1). Above $\sim 10\,K$, outside the VRH regime, $\Delta G_{\text{ax}}(H)$ becomes nearly flat (Fig. 4d); there both $\tau_c$ (Supplementary Fig. 7a) and $|g_{\text{eff}}|$ (see Supplementary Fig. 7b) appear to saturate, and spin-memory phenomenon is not expected. The typical field scale associated with the E-S energy $T_a \sim 10–30\,K$ in the 1–2T range, in close correspondence with $H^*$ at low $T$. As before, $H_{\text{max}}$, and in the low-temperature localized state $H_{\text{max}}$ becomes the limiting crossover field (Fig. 4e). Above $T_a \approx 180\,C$ ($E_Q \approx 60\,meV$), in the strongly disordered crystalline state spin memory phenomenon is not detectable.

Transition to a 2D weak antilocalization regime

The exit from the spin-memory state at $\approx 180\,C$ is clearly evidenced by a transition from the negative MR (neg- $MR$) to a positive MR (pos- $MR$) ‘cusp’ characteristic of the WAL state (Fig. 5a). The WAL cusp\textsuperscript{29} scales with the transverse component of applied magnetic field $H_T = H \cos \beta$ (Fig. 5b), consistent with the 2D (orbital) character expected in a topological insulator\textsuperscript{8} under weak disorder. This 2D scaling should be contrasted with isotropic (3D) scaling of the neg-$MR$ peak in the spin-memory state. Thus, unlike the WL-WAL transition driven by magnetic impurities\textsuperscript{20}, the transition from spin-memory onto a WAL state is also a 2D–2D dimensionality transition at which the electron system rapidly delocalizes. We remark again that WAL onsets at $\approx 180\,C$, way above the crystallization temperature of $\approx 140\,C$ (see inset in Fig. 5c), pointing to a disorder threshold for the gapless topological state.

DISCUSSION

The emergence of WAL in the diffusive transport regime\textsuperscript{3} should be reflected in the evolution of the electron localization length $\xi$. Figure 5c shows that with increasing $T_a$, $\xi$ first experiences a ‘kink’ at the first order crystallization transition\textsuperscript{27}. However, its largest change—an increase by orders of magnitude—initiates at the onset of WAL. We note that the electron mean free path is only limited by the film thickness and the grain size in the crystalline state at high $T_a$ (Supplementary Fig. 8).

To translate $T_a$ into a disorder energy scale $W_Q$\textsuperscript{12} we use the annealing evolution of the full-width-half-max (FWHM) of the in-plane Raman $E_1$ mode (Fig. 5c and Supplementary Fig. 1) as a metric of disorder\textsuperscript{30}. A simple phenomenological scaling is derived by setting maximum disorder in the amorphous state at the energy corresponding to the bulk gap $E_g \sim 200\,meV$\textsuperscript{16} in the crystalline state and minimum disorder (at the highest $T_a$) to zero (Fig. 5d, Supplementary Note 11, and Supplementary Fig. 9). When the angular field-dependence of magnetoresistance $\Delta R$ is plotted...
corresponding to a precision equal to one conductance quantum to 1/2 for a single 2D channel. The plot of Nagaoka (HLN) findings show that spin-memory effect sets a disorder critical point of 0.25. Strong charge screening in Sb2Te3 (large dielectric constant $\varepsilon \sim 170^{33}$) renders the Coulomb energy ($E_C \approx 4$ meV) much smaller than the Fermi energy ($E_F \approx 260$ meV); with $E_C \ll E_F$ this points to the transition to be disorder-driven$^{32}$. Our findings show that spin-memory effect sets a disorder threshold at which topological protection of the surface states is reclaimed, although bulk channels may still contribute. Direct measurements of glassy spin response in materials other than Sb2Te3 are yet to be carried out; it is clear, however, that for spin memory the spin-lifetime $\tau$ has to be much longer than the hopping time $\tau$. We expect spin memory to be visible in a broader family of strong SOC materials; it has been reported in SnSb2Te4$^{33}$, where crystal (and defect) structure is different$^{34}$. The electrical control of this spin-dependent transport can, in principle, be achieved through electrostatic gating$^{35}$, and by locally modifying spin correlations using currently practiced doping techniques.

**METHODS**

Film growth and structural characterization

Films of Sb2Te3 with thicknesses ranging from 20 to 100 nm were sputter-deposited at room temperature in Ar gas at 4 mTorr and a flow of 46 sccm from a nominally stoichiometric target using 15 W DC power on Si3N4 (100 nm)/Si substrates. The stoichiometry was confirmed by Rutherford
For background subtraction. At each temperature an empty twin sensor was used exfoliated from their substrates and placed directly on the SiO₂-passivated with H⁺ magnetization, 50 μm in lateral size thin Sb₂Te₃ films. a Transition from the spin-memory to weak antilocalization (WAL) state in disordered crystalline Sb₂Te₃ films. Upon annealing, ΔRₓₓ undergoes transition from the spin memory neg-MR state to a positive magnetoresistance (pos-MR) WAL regime at Tₑ ≃ 180 °C, when charge transport becomes diffusive. b The WAL pos-MR has a 2D character as evidenced by the scaling with the transverse component of magnetic field. c Localization length $\xi \propto T_c^{-3}$ shows a distinct kink at crystallization and a smooth transition to a 2D WAL state. $T_c$ was obtained from fitting to the E-S VRH formula. Inset: Crystallization at $T_c \geq 140$ °C (yellow dash) is clearly seen as sharp lines in the Raman spectra vs. $T_a$. Transition to WAL at $\theta = 180$ °C is indicated by red dash. d Ratio $r$ of low-field $\delta R$ with the magnetic field out-of-plane ($\theta = 0$) to the one with a field tilt $\theta = 30$ °C vs. disorder parameter $W_D$. In the 2D WAL regime, $r$ is constant, owing to scaling with $H_L = Hcossθ$. Inset: $W_D$ is obtained from parametric scaling of the FWHM vs. $T_a$ of $E_2^\text{g}$ in-plane Raman mode (see Supplementary Note 11). e Longitudinal conductance $G_{xx}$ vs. $W_D$. $G_{xx}$ ∝ $e^2/h$ at the transition to 2D WAL. Inset: The number of quantum channels estimated from the fit to HLN theory. The parameter $a = 0.5$ indicates a single quantum channel.

Backscattering (RBS) and particle induced x-ray emission (PIXE). RBS data were collected at NEC 3UH Pelletron using a Si surface barrier detector with He ions at 3 MeV. PIXE data was collected using a Si-Li detector with H⁺ ions at 1 MeV. Elemental analysis was done at Evans Analytical Group. X-ray diffraction characterization was performed using Bruker D8 Discover system with the da Vinci configuration, with a monochromated beam (λCu ≈ 1.5418 Å) and a scintillator detector with analyzer crystal (HR-XRD). The film morphology was characterized using the FEI Titan Temis 300 transmission electron microscope (TEM), 200 kV, with TEM resolution of 0.9 Å and 4 x 4 k Ceta 16 M CMOS camera. Disorder was characterized by Raman spectra using 633 nm linearly polarized excitation in a back-scattering configuration, with power kept below 2 mW to avoid heating effects.

**Magnetic measurements**

Magnetic measurements were performed using custom-designed on-chip $\mu$-Hall sensors based on In$_{0.15}$Ga$_{0.85}$As heterostructures (Si). To measure magnetization, ~50 μm in lateral size thin Sb₂Te₃ film samples were exfoliated from their substrates and placed directly on the SiO₂-passivated sensor using PDMS. At each temperature an empty twin sensor was used for background subtraction.

**Transport measurements**

Transport measurements were performed in a 14 Tesla Quantum Design PPMS system in 1 mTorr of He gas on many film samples, each subjected to the same annealing protocol used to tune the level of disorder. Lithographically patterned structures combining both Hall bar and van der Pauw electrical contact configurations with Ti/Au metallurgy were used (Fig. 2a). Measurements were performed on as-deposited films and on the same films after each 5 min annealing step in a box furnace in flowing nitrogen in the temperature range across crystallization at $T_a \sim 140$ °C. We used a numerical Monte Carlo technique to fit our transport data to Eq. (1).

**Data Availability**

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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