Observation of current-induced, long-lived persistent spin polarization in a topological insulator: A rechargeable spin battery

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Topological insulators (TIs), with their helically spin-momentum–locked topological surface states (TSSs), are considered promising for spintronics applications. Several recent experiments in TIs have demonstrated a current-induced electronic spin polarization that may be used for all-electrical spin generation and injection. We report spin potentiometric measurements in TIs that have revealed a long-lived persistent electron spin polarization even at zero current. Unaffected by a small bias current and persisting for several days at low temperature, the spin polarization can be induced and reversed by a large “writing” current applied for an extended time. Although the exact mechanism responsible for the observed long-lived persistent spin polarization remains to be better understood, we speculate on possible roles played by nuclear spins hyperfine-coupled to TSS electrons and dynamically polarized by the spin-helical writing current. Such an electrically controlled persistent spin polarization with unprecedented long lifetime could enable a rechargeable spin battery and rewritable spin memory for potential applications in spintronics and quantum information.

INTRODUCTION

With an increased interest in spin logic devices compatible with the existing semiconductor technology, manipulating electron spins in nonmagnetic semiconductors has been one of the most active research directions in spintronics (1), promising smaller, faster, less power-consuming information-processing and communication devices. However, several technical challenges have to be overcome in semiconductor spintronics, such as efficient spin injection, long spin lifetime, and efficient spin transport, manipulation, and detection (2, 3). A particularly important goal is to generate a controllable electronic spin polarization that can have a long lifetime, using all-electric approaches that are free from magnetic materials (1–3).

Three-dimensional (3D) topological insulators (TIs) represent a new class of electronic quantum phases with strong spin-orbit coupling, hosting spin-helical topological surface states (TSSs) protected by time-reversal symmetry (4–6). One of the most fundamental and marked properties of TSS is spin-momentum locking (SML), where the electron spin is “locked” in plane and perpendicular to its momentum, making 3D TIs highly promising for applications in nanoelectronics and spintronics (4, 6). Several recent experiments in 3D TIs have demonstrated current-induced helical spin polarization attributed to the SML (7–14). The observed spin polarization reverses upon reversing the current, requires a DC current to maintain it, and vanishes as soon as the current is removed. However, for many applications, a persistent and long-lived spin polarization (sometimes referred to as a spin “battery” or memory) may be desired.

Here, we report spin potentiometric measurements in Bi2Te2Se (BTS221) TI thin flakes that have revealed a new phenomenon not observed previously in TIs: a current-induced persistent electron spin polarization (ESP). A voltage between a ferromagnetic (FM) and one of the nonmagnetic contacts is monitored as a function of an in-plane magnetic (B) field (Fig. 1A) applied to magnetize the FM contact (fig. S1). We observe a hysteretic step-like voltage change when the B field is swept between the opposite directions, resulting in a clear difference in the voltage detected between the opposite FM magnetizations. Such a voltage difference (ΔV) measured by spin potentiometry is a measure of spin chemical potential (μ↑ − μ↓ = −eΔV, where −e is the electron charge), representing an out-of-equilibrium ESP. However, in stark contrast to previous experiments, such a spin signal (ΔV)—both its sign and amplitude—shows little dependence on the sign and magnitude of a relatively small DC detection current (Iw). The Iw-independent spin signal is observed to persist for many hours even without any bias current. A marked “writing” effect on the spin signal is also realized by applying a large DC “writing current” (Iw) for an extended time at zero magnetic field that can reverse the persistent spin signal depending on the direction of Iw (analogous to “charging” a battery and setting its polarity). Although exact mechanisms giving rise to the observed long-lived persistent ESP are not yet clear at this point, we speculate on a possible scenario where nuclear spins—coupled with spin-momentum–locked TSS electrons via hyperfine interaction—can be dynamically polarized by a (sufficiently large) helical spin-polarized current carried by TSS electrons and, in turn, act back on the TSS electron spins to give the observed persistent, out-of-equilibrium ESP.

RESULTS

We performed the spin potentiometric measurements (schematically illustrated in Fig. 1A) on exfoliated BTS221 thin flakes placed on top of SiO2/Si substrates (Fig. 1B). We usually apply a small (detection) current (Iw) between the two “outside” Au contacts and monitor the voltage (V) between an Au contact and a middle FM [permalloy (Py)] contact, as an in-plane B field is applied to magnetize the Py contact (Fig. 1A and fig. S1). The representative results (Fig. 1, C to H, and fig. S2) of a three-terminal spin device (Fig. 1, A and B; a similar setup to what we previously used on other samples to demonstrate a current-induced spin polarization (14)) are measured on a 10-nm-thick flake (device A; Fig. 1B) at T = 0.3 K, with moderate bias current (Ib) ranging from 100 pA to 1 μA [positive current (left) and negative current (right)]. As we sweep...
an in-plane magnetic $B$ field between the $\pm y$ directions (orthogonal to the current direction; note that we define positive $B$ field to be along the $-y$ direction) to switch the magnetization ($M$) of the Py detector, the voltage ($V$) it measures shows a clear hysteretic step-like switching behavior, exhibiting a low (high) voltage state when the Py detector is magnetized to the $-y$ ($y$) direction. It is interesting to note that the same trend of the step-like voltage change is observed regardless of the polarity of $I_d$. This is qualitatively different from previous spin potentiometry measurements where the trend of the step-like change reverses upon reversing $I_d$ that indicates a current ($I_d$)-induced spin polarization due to SML (7, 14). We quantitatively define the spin signal as follows: $\delta V = V_{+M} - V_{-M}$ (labeled in Fig. 1E), reflecting the step-like voltage change near the coercive field. The $\delta V$ versus $I_d$ is plotted in Fig. 11, showing a very weak $I_d$ dependence where $\delta V$ stays around a few negative microvolts even when $I_d$ is varied over four orders of magnitude [inset of Fig. 11, and figs. S2 and S3; the notable increase of $|\delta V|$ between $|I_d| = 10$ and 100 nA in Fig. 11 is more specific to this device and not commonly observed in other devices (for example, fig. S3)]. The above $I_d$-independent spin signal is again in contrast to previous measurements (7, 11, 14), where the spin signal ($\delta V$) is found to be approximately proportional to $I_d$. The direction of the channel (top surface of our sample) spin polarization $S$ can be determined from the sign of $\delta V$ in our spin potentiometric measurements, based on the fact that an FM contact with magnetization $M$ primarily probes the channel spins oriented along the same direction as the FM majority spins (oriented antiparallel to $M$) and thus will measure a lowervoltage (higher electron chemical potential) if $S$ is antiparallel to $M$ (14–16). The determined directions of $S$ (labeled in the insets of Fig. 1, G and H) are the same for both the positive and negative $I_d$. Our observations suggest that the electronic spin polarization revealed by spin potentiometry in this sample is not induced or controlled by the current ($I_d$) we applied in these measurements but rather arise from a different origin that is intrinsic to the sample or setup before these measurements.

We further found that the sign of $\delta V$ in our device can be (re)set by applying a relatively large DC bias current for an extended time (referred to as a writing current, $I_{w}$, to distinguish from the relatively small $I_d$ in Fig. 1 that does not affect $\delta V$). In this experiment, we usually apply a large $I_{w}$ between the two outside Au contacts in a four-terminal device (Fig. 2A) for an extended time at zero $B$ field, and then the spin signal is measured by spin potentiometry (Fig. 2B; the four-terminal configuration is found to give a similar spin potentiometry signal as measured in three-terminal configurations) with a small $I_d$. Figure 2 (C to H) shows the results measured on a 25-nm-thick BTS221 flake (device B) at $T = 1.6$ K. After applying a $I_w = -40 \mu$A for 2 hours, spin potentiometric measurements (Fig. 2B) were performed with $I_d = \pm 0.5 \mu$A (shown in Fig. 2, C and D). We observe a similar step-like voltage change as that in Fig. 1, with a negative $\delta V$, for both the positive and negative $I_d$, indicating the presence of a channel spin polarization whose direction is independent of $I_d$. We then apply a large positive $I_w = 50 \mu$A for 0.5 hours. Afterward, spin potentiometry was performed (Fig. 2, E and F) with $I_d = \pm 0.5 \mu$A, and the sign of the spin signal ($\delta V$) is reversed to positive, which is opposite to that in Fig. 2 (C and D) (after $I_w = -40 \mu$A), indicating that the direction of the spin polarization $S$ has now been reversed after the application of a reversed $I_w$, but still independent of $I_d$. Remarkably, the sign of $\delta V$ can be reversed again and back to the case in Fig. 2 (C and D) after applying a negative $I_w = -40 \mu$A for 3 hours, as shown in Fig. 2 (G and H). Similar effects are also observed when $I_w$ is applied during sample cooling (fig. S4) or at a higher temperature (~26 K; fig. S5). We clearly see that the sign of the spin polarization can be reversed by a current pulse.

**Fig. 1.** Detection of current ($I_d$)-independent spin signal in a Bi$_{2}$Te$_{2}$Se (BTS221) thin flake by spin potentiometry. (A) Schematic 3D device structure used in the potentiometric measurement, showing a three-terminal electrical connection. The Ti surface defines the $x$-y plane, and the surface normal defines the $z$ direction. The two outside nonmagnetic (for example, Au) contacts (the left is grounded) are used to inject a DC bias current, and the middle FM (for example, Py) contact is magnetized by an in-plane magnetic $B$ field (labeled) along its easy axis ($y$ direction, with $-y$ defining the positive $B$ and $M$ direction). The middle FM contact is a tunneling probe (with a thin Al$_2$O$_3$ tunnel barrier underneath in our devices). (B) Optical image of device A used in the potentiometric measurement. (C to H) Voltage measured by the FM spin detector (Py) on device A as a function of in-plane magnetic field for representative bias currents ($I_d$) of 100 pA (C), $-100$ pA (D), 10 nA (E), $-10$ nA (F), 1 $\mu$A (G), and $-1$ $\mu$A (H). The directions of $I_d$ (red arrow), the inferred channel (top surface) spin polarization $S$ (dashed green arrow), and Py magnetization $M$ (black arrow) are labeled in (G) and (H). (I) The voltage change $\delta V = V_{+M} - V_{-M}$ is shown as a function of the applied DC bias current. Here, $\delta V$ is extracted from forward sweep traces (backward sweeps yield similar results). All the measurements were performed at the temperature $T = 0.3$ K.
signal and the direction of the channel spin polarization \( S \) are determined by \( I_w \) (reversing \( I_w \) reverses \( S \)) but are independent of \( I_d \) (as long as \( I_d \) is small enough). We find that the spin signal \( \delta V \) also increases with the increasing writing current \( I_w \) (Fig. 6) and writing time (Fig. 7). Such a current \( (I_w) \)-induced writing effect (where the induced spin polarization persists even after \( I_w \) is turned off) of spin polarization has not been previously observed in 3D TIs. We further note that the direction of \( S \) is consistent with that of the TSS spin polarization induced by \( I_w \) (see arrows indicating the directions of \( S \) and \( I_w \) in Fig. 2, C to H) according to the helicity of SML of the TSS (7, 11, 14).

Figure 3 shows spin potentiometric measurements at \( T = 1.6 \) K in another device (C’); a 30-nm-thick BTS221 flake, with a four-terminal configuration shown in Fig. 2, A and B) at a series of DC bias currents \( I_d \) increasing from relatively small to very large values (\( \pm 80 \) mA), where we found the spin signal transitions from being largely \( I_d \)-independent to linearly dependent on \( I_d \). At a small \( I_d = \pm 0.01 \) mA, the spin signal \( \delta V \) is about \( -0.5 \) \( \mu \)V for both the positive and negative currents (Fig. 3, A and B), and the trend of the signal is qualitatively similar to those presented in Fig. 1, suggesting a channel spin polarization \( S \) along the +y direction and independent of \( I_d \). In contrast, at a large \( I_d \) such as \( \pm 80 \) mA (Fig. 3, I and J), a qualitatively different behavior is observed. Upon reversing \( I_d \) the step-like change in the measured spin potential now reverses its trend, and spin signal \( \delta V \) reverses its sign (\( \delta V \sim 1.8 \) \( \mu \)V for \( I_d = 80 \) mA and \( \delta V \sim -2 \) \( \mu \)V for \( I_d = -80 \) mA). Such a behavior that \( \delta V \) reverses upon reversing \( I_d \) is more similar to that studied in previous spin potentiometric measurements on TIs, indicating an \( I_d \)-induced and reversible helical spin polarization (7, 11, 14). As labeled by the arrows in Fig. 3 (I and J), the direction of \( S \) is locked to such a large \( I_d \) in a way that is consistent
with the spin helicity of TSS. At intermediate positive currents (Fig. 3, C and E), the voltage signal exhibits a “transitional” behavior, where the trend of the step-like change undergoes reversals (sometimes even during measurements taken at the same $I_d$; see some examples marked by the brown arrows in Fig. 3, C and E). The dependence of the spin signal $\delta V$ on $I_d$ is summarized in Fig. 3K, where two distinct behaviors are observed: (i) at $|I_d| < 5 \mu$A, $\delta V$ is always negative and relatively constant, about $-0.5 \mu$V, independent of both the polarity and amplitude of $I_d$; (ii) at $|I_d| > 10 \mu$A, $\delta V$ reverses its sign with reversing $I_d$ and is largely linearly dependent on $I_d$.

Figure 4 shows various studies of the dependences of the $I_d$-independent spin signal (in the small $I_d$ region) on the out-of-plane component of the $B$ field, gate voltage, and temperature. Figure 4A shows the voltage $V$ measured on device A as a function of $B$ field tilted away from the surface at various different angles $\theta$ (depicted in the inset of Fig. 4A). We see that the coercive field ($B_C$ labeled in Fig. 4A) increases as $\theta$ increases, whereas the in-plane component of the coercive field ($B_C \cos \theta$) and the measured spin signal $\delta V$ are independent of $\theta$, as summarized in Fig. 4B. Our results demonstrate that the spin signal $\delta V$ is independent on the out-of-plane component of the magnetic field and also suggest the absence of the Hanle effect in our TI system. It is known that the Fermi level of a bulk-insulating TI sample can be tuned by gating. Figure 4C shows the spin potentiometric measurements at $I_d = \pm 1 \mu$A on device D (a 17-nm-thick flake) at different back-gate voltages. The extracted spin signals $\delta V$ and the corresponding voltage $V_0$ (measured separately at $B = 0 \, \text{T}$) as functions of the back-gate voltage $V_g$ are presented in Fig. 4D. When $V_g$ is tuned from 50 to $-170 \, \text{V}$, $V_0$ measured at $I_d = 1 \, \mu$A exhibits an on-off ratio of $\sim 2$, and the spin signal $\delta V$ is significantly enhanced as the Fermi level is tuned closer to the charge neutrality point of TSS at a negative $V_g$ (Fig. 4, C and D). We further found that the detected $I_d$-independent spin signal $\delta V$ measured on device E (a $\sim 20$-nm-thick flake) decreases with increasing temperature $T$ and is observable up to $\sim 76 \, \text{K}$ (Fig. 4E and fig. S8).

The largely $I_d$-independent spin signal at small $I_d$ suggests that it can be observed even with no $I_d$ applied. Figure 5A shows the open-circuit voltage measured at $T = 1.6 \, \text{K}$ in device F (a 15-nm-thick flake, with a

**Fig. 4.** Dependences of the spin signal on out-of-plane $B$ field component, gate voltage, and temperature. (A and B) Effect of perpendicular (out-of-plane) magnetic field on spin signals. In this experiment, the magnetic $B$ field was tilted away from the sample (device A) surface plane ($x$-$y$) by an angle $\theta$ [depicted in the inset in (A)]. Data are measured at $I_d = -100 \, \text{nA}$ and $T = 0.3 \, \text{K}$. (A) Magnetic field dependence of the voltage at different angles. (B) The coercive field $B_C$ (marked in (A) for the $\theta = 54^\circ$ (trace)), $B_C \cos \theta$, $B_C \cos \theta$, spin signal $\delta V$ as functions of $\theta$. (C and D) Gate dependence of the spin signal measured on device D. (C) The measured voltage $V$ as a function of the in-plane magnetic field measured at $T = 1.6 \, \text{K}$ and $I_d = -1 \, \mu$A (top) and 1 $\mu$A (bottom) at various back-gate voltages $V_g$. (D) The extracted spin signal $\delta V$ (left axis; black symbols) and the voltage $V_0$ (right axis; red dashed line; measured at $B = 0 \, \text{T}$ before performing the spin potentiometry) as a function of $V_g$. The measurements were performed at $T = 1.6 \, \text{K}$. (E) Temperature $T$ dependence of the measured spin signal from device E up to 76 K. Devices D and E have a four-terminal configuration (Fig. 2B).
DISCUSSION

In our study, we have observed a long-lived persistent out-of-equilibrium spin polarization in the TSS conduction electrons even without any bias current. At low temperature, such an electronic spin polarization can persist for at least ~2 days, which is several orders of magnitude higher than the typical lifetime of electron spins [typically shorter than 1 μs (13–17), in rare cases reaching ~1 ms (17)] when directly injected optically (18) or electrically (19) into semiconductors (17). Thus far, nuclear spins are the only known entities that can give an extraordinary long lifetime (capable of reaching many hours or longer, which is close to what we observed) of injected spin polarizations, reflecting the fact that nuclear spins are usually better isolated with much longer relaxation time (in some cases reaching >1 day; Fig. 6 and table S1). Previous studies have also shown that it is possible to extend the electron spin lifetime (reaching thousands of seconds) by coupling to nuclear spins through the hyperfine interaction (20–23). We have compared our measured spin polarization lifetime with previously reported spin relaxation time (T1) of nuclear spin polarizations (NSPs) in a variety of systems (24–41), as shown in Fig. 6 (the same data are also shown in table S1). It is notable that our achieved spin lifetime (~45 hours, even at zero magnetic field) exceeds (by 50% or more) the longest lifetime (~30 hours in B = 5 T) in these previous measurements and is orders of magnitude longer than those lifetimes (where the longest was <1 min) previously measured at zero B field.

It is known that the nuclear spins can be dynamically polarized by spin-polarized electrons through hyperfine interaction (42, 43). Although we do not have direct measurements or evidence of NSP in our experiment, we speculate that nuclear spins (near the surface) hyperfine-coupled to TSS electrons and dynamically polarized by the spin-helical writing current (Fig. 7) may have played important roles and may be part of a possible explanation to our observation (see more detailed discussions in the Supplementary Materials). In our experiment, a large Iw can create an out-of-equilibrium spin polarization in the TSS conduction electrons due to the SML, resulting in a large chemical potential difference (44) between the spin-up and spin-down electrons (for example, Fig. 7, A and B). An electron from the more occupied “majority” spin state that flips its spin and transfers to the less occupied “minority” spin state will induce an opposite nuclear spin flop (Fig. 7A) owing to the hyperfine interaction (without which a full spin reversal of a TSS electron would not be allowed by time-reversal symmetry) (45). Such a hyperfine interaction–enabled electron-nucleus spin flip-flop process results in a dynamic NSP by a current-induced ESP (Fig. 7A). Reversing Iw reverses not only the direction of the TSS

Fig. 5. Persistent spin signal and its time dependence. (A) Open-circuit voltage detected by the FM contact as a function of the in-plane magnetic B field measured on device F with the source-drain electrical connection open, showing a spin signal ΔV ∼1.3 μV. (B) Open-circuit voltage versus B measured after t = 45 hours. (C) Spin signal ΔV versus elapsed time t, where ΔV gradually decreases to ~0.5 μV in the first 3 hours and stays around ~0.6 μV in the next 42 hours. The measurements in (A) to (C) were performed at T = 1.6 K. Inset shows the schematic of the open-circuit measurement in device F. (D) Open-circuit voltage versus B measured at T = 45 K. (E) Open-circuit voltage versus B measured at T = 45 K after t = 10 hours. (F) ΔV versus t measured at T = 45 K. The directions of Py magnetization M (black arrow) and the inferred channel (top surface) spin polarization S (dashed green arrow) are labeled in (A), (B), and (D).
Fig. 6. Comparison of the spin polarization lifetime that we measured with the lifetime of the NSP reported in different systems from previous literature. (A) Spin polarization lifetime as a function of temperature and (B) spin polarization lifetime as a function of magnetic field. The red superscripts refer to the corresponding references. Previous data measured at \( B = 0 \) T are highlighted in green. QD, quantum dot.

ESP via SML but also the induced NSP, as shown in Fig. 7B. We further speculate that the polarized nuclear spins could act back again on the TSS electron spins through the hyperfine interaction and polarize the electron spins, giving rise to a spin potential difference \( (5V) \) in electrons (shown in Fig. 7C and D) measured by spin potentiometry (44).

If the nonequilibrium persistent ESP in our 3D TI samples is indeed maintained by a background NSP as we hypothesized, our observations suggest that both the ESP and NSP in our system are highly persistent and robust (with long lifetime, lack of Hanle effect, among others), possibly coupled (or even effectively locked) to each other in a way that helps stabilize each other (significantly reducing the relaxation rate to give long lifetime) and providing each other with an internal field that far exceeds the external magnetic field. An interesting question calling for further studies would be to investigate whether the spin-helical nature of the TI surface state electrons (with only one spin state at each momentum and thus only half the Fermi surface of the conventional 2D electrons) may bring novel physics and behavior in the coupling with nuclear spins and relaxation of NSP/ESP differently from what are known in conventional metals (46) and semiconductors. In addition, compared with previously studied coupled nuclear-electron spin systems, we speculate that several possible factors in our TI samples might contribute to a highly suppressed nuclear spin relaxation: (i) The 3D TI samples are bulk insulating with few bulk charge carriers at low temperatures, thus largely avoiding what would be an important channel of NSP relaxation (via bulk carriers) in conventional metals and doped semiconductors. (ii) The TSS ESP is concentrated at the surface; this gives rise to a gradient in an effective magnetic field (Knight field due to the ESP) from the surface to the bulk. It has been shown that the spin diffusion could be suppressed in a magnetic field gradient (47). The decrease of the persistent spin signal with increasing temperature in our experiments could arise from a combination of several materials and technical issues: (i) Our BTS221 sample is bulk insulating only at relatively low temperatures. As temperature rises, there will be an increasing amount of thermally activated conducting carriers in the bulk (no longer insulating) that can provide an extra channel to relax nuclear spins. (ii) The spin potentiometric measurement itself can also become less efficient at higher \( T \) because of the increased bulk conduction (loading the surface state spin signal) as well as possible degradation of the Al2O3 tunneling barrier at higher \( T \).

Several recent experiments [including our previous study (14)] performing spin potentiometric measurements (7, 9, 14) in TIs have observed a linearly \( I_w \)-dependent spin potential signal indicating current-induced (electron) spin polarization, but have not observed an \( I_w \)-independent (or open-circuit, persistent) spin signal and a writing current \( (I_w) \) effect, as reported in this work. It is possible that the NSP in the samples studied in those previous experiments (7, 9–14) may not have been sufficiently developed to have notable effects on the ESP. We note that differences in the measurement procedure and history (such as the magnitude and duration of the applied current) can have strong influences on the results (for example, figs. S5 to S7). For example, in cases such as Fig. 1 (where we did not intentionally apply a large writing current), the persistent spin signal is found to develop after

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**Fig. 7. Schematics depicting a proposed mechanism of spin polarization transfer between the electrons and nuclei via the spin flip-flop process due to the hyperfine interaction.** (A and B) Current-induced NSP: A large writing current \( I_w \) (A) induces (via SML of TSS) an ESP, which dynamically polarizes nuclear spins by the hyperfine interaction–enabled electron-nuclear spin flip-flop process. Reversing \( I_w \) (B) can reverse the NSP due to the reversed TSS ESP. (C and D) An existing NSP induces an ESP through hyperfine interaction–enabled spin flip-flop process, depicted as spin-down (C) and spin-up (D) polarized cases. Because of the long lifetime of nuclear spins, the induced ESP (and the measured spin signal) can persist for a long time (even in the absence of any driving current) but can be reversed by a large \( I_w \) (A and B) that reverses the nuclear spins.
performing many measurements (before those in Fig. 1) with a relatively small current but over an extended time (which may have slowly polarized the nuclear spins). Although all the details and factors affecting the NSP remain to be better understood, we note that the samples used in this work, where we have observed $I_D$-independent persistent spin signal (and at larger current a transition to $I_D$-dependent spin signal), are also much smaller (less than a few micrometers in size) than those used in previous studies (7, 11, 14) [tens or hundreds of micrometers in size, even though our previous measurement (14) used BiTS221 flakes exfoliated from the same bulk crystal as in our current work]. We speculate that in smaller-sized samples, it may be easier for the current to drive the TSS electrons into a stronger out-of-equilibrium state (with a larger spin-potential difference), enhancing the electron-nuclear spin flip-flop process. We also speculate that other factors such as disorder and the fabrication process may affect the environment of nuclear spins and effective coupling between the electrons and nuclei, and thus the ability for current to polarize nuclear spins.

In summary, we report the observation of a persistent ESP in a 3D TI with a long time scale. Such a persistent source of ESP and spin potential (even after the injection current is removed) may be used as a spin battery (40) or spin capacitor in spintronics (3, 48). The possibility of electrically controlled and rewritable electronic and nuclear spin memory or storage may also open potential applications in solid-state spin-based quantum computing and information processing (24, 48).

MATERIALS AND METHODS

Device fabrication

The high-quality bulk 3D TI crystals of BTS221 were grown by the Bridgman method (49–51). Thin TI flakes (typical thickness of ~10 to 40 nm and lateral size of up to ~5 μm) were exfoliated from the bulk crystals using the “scotch tape method” (52) and placed on top of standard doped Si substrates with a 285-nm-thick SiO2. To fabricate a thin Al2O3 tunneling barrier (used for spin detection) and protect the sample surface from oxidation, we immediately transferred the exfoliated flakes into a high-vacuum e-beam evaporator and coated them with a 0.7-nm-thick Al film under a pressure of 2.0 × 10−7 Torr. After reexposing the sample in air, the Al film was fully oxidized into Al2O3. The film [50-nm-thick Py (Ni0.80Fe0.20)] and nonmagnetic (70-nm-thick Au) electrodes were fabricated by two rounds of e-beam lithography and e-beam evaporation. Before depositing the Au contacts, the samples were dipped in a BOE (buffered oxide etch) (HF/H2O, 1:6) solution to remove the thin layer of Al2O3 so that Ohmic contacts can be achieved.

Characterization of our magnetic (Py) electrodes

Our magnetic (Py) electrodes were characterized by magnetic force microscopy at room temperature.

Spin potentiometric measurements

The spin potentiometric measurement was performed in a three- or four-terminal configuration involving one FM inner contact (Py) and two or three nonmagnetic outer contacts (Au), as schematically shown in Figs. 1A and 2B, respectively. A DC source-drain bias current can be applied between the two outermost Au electrodes. A voltage (potential difference) was measured between the Py and the adjacent Au contact (to its left) using a high-impedance voltmeter. We define a positive current as flowing from right to left along the –x direction, and a positive in-plane magnetic field as applied in the –y direction along the easy axis (Figs. 1A and 2B) of the FM electrode. All measurements were performed in a He-3 cryostat with the base temperature $T_{\text{base}} = 300$ mK or a variable temperature cryostat with $T_{\text{base}} = 1.6$ K.

SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/3/4/e1602531/DC1

note 51. More discussions on the possible mechanisms for the persistent spin polarization, dynamical spin polarization between TSS electrons and nuclear spins, and the interplay of the SML of TSS electrons and hyperfine interaction between the TSS electron spins and nuclear spins.

fig. 51. Characterization of the magnetic (Py) electrodes.
fig. 52. Complete data set on the current dependence of the measured spin signal in device A. fig. 53. Spin signal largely independent on $I_D$ measured in additional devices.
fig. 54. The writing current effect on the spin signal during cooling.
fig. 55. The writing current effect on the spin signal measured at 26 K.
fig. 56. Effect of the magnitude of writing current $I_w$ on the spin signal.
fig. 57. Effect of current writing time on the spin signal.
fig. 58. Temperature dependence of the measured spin signal.

table 51. Lifetime of the NSP reported in different systems from previous literature. References (33–68)

REFERENCES AND NOTES


Author contributions: I.M. synthesized the TI bulk crystal. J.T. fabricated the devices, performed transport measurements, and analyzed the data with advice from Y.P.C. S.H. and S.D. helped with the theoretical understanding. J.T. and Y.P.C. wrote the paper, with discussions and comments from all other coauthors. Competing interests: The authors declare that they have no competing interests. Data and materials availability: All data needed to evaluate the conclusions in the paper are present in the paper and/or the Supplementary Materials. Additional data related to this paper may be requested from the authors.

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