Topologically protected spin diffusion and spin generator using chalcogenide superlattices

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Spintronics is expected to be the basis for future ultra-low-energy nanoelectronic devices. To operate such devices at room temperature, amplifiers, batteries, capacitors, as well as spin current sources are required. Here we report a chalcogenide superlattice composed of GeTe and Sb$_2$Te$_3$ layers that have a topologically protected spin diffusion length exceeding 100 μm at room temperature. A spin generator is demonstrated by combining magnetic injectors (TbFeCo) with this superlattice. The spin current was found to increase exponentially with the number of superlattice periods. We used this effect to demonstrate a 15-fold increase in the spin current. In addition, spin rectification is possible by growing the superlattice layers with atomic-level thickness accuracy. The reported chalcogenide superlattice spin generators and rectifiers open new opportunities to design low-energy spintronic integrated circuits and quantum computers.

INTRODUCTION
Spintronics is key to realizing ultra-low-power electronics. An example of spintronic device is a magnetoresistive nonvolatile memory. Spintronic devices usually consist of three parts: a spin injector, a spin detector, and a medium to transport spins. The injector and detector are usually made of a ferromagnetic material and a metal with large spin–orbit coupling, whereas the transport medium is generally a metal or a semiconductor. Although injected spin current from a ferromagnetic material diffuses through the transport medium, and is rapidly attenuated due to scattering. The spin diffusion length at room temperature is generally less than 1 μm in metals or semiconductors. A longer spin diffusion length is required for integrating spintronic devices using a single spin source or battery. Therefore, a technology that extends the spin diffusion length is expected to revolutionize low-energy electronic devices.

Spintronics devices can be enhanced by exploiting the discovery of topological insulators (TIs) and other related materials, such as Dirac and Weyl semimetals (DS and WS). Importantly, the spin in TIs is protected from scattering, even at room temperature, by spatial inversion and time-reversal symmetries. However, spin transport in TIs is restricted to edges in two-dimensional structures and to surfaces in three-dimensional (3D) structures. This low dimensionality limits the current that can be transported.

Recently, chalcogenide topological superlattices (SLs) have attracted attention, because they can be used to transport spin using p-electrons. These SLs consist of stacked chalcogenide normal insulator (NI) and TI thin layers. Similar SLs, which consist of GeTe (NI) and Sb$_2$Te$_3$ (TI), have also been applied to nonvolatile phase-change memory. The ferroelectric phase, or simply “Ferro-phase”, of these SLs is sensitive to magnetic fields at room temperature. This is interesting, because it does not contain any magnetic elements. The origin of the magnetic sensitivity is due to the p$_z$-electrons of Ge and Te atoms in the Ge$_2$Te$_2$ layers of the SL. Until now, these SLs have only been applied to nonvolatile phase-change memory devices. However, the fact that the Ferro-phase is a WS implies that they may also be important in spintronics devices and hybrid phase-change spintronics universal memories.

In this study, we show that the spin diffusion length of the Ge$_2$Te$_2$-Sb$_2$Te$_3$ SLs is over 100 μm at room temperature. Moreover, we demonstrate how this topologically protected spin diffusion length can be used in prototype spin generators and rectifiers that operate over hundreds of micrometers. Indeed, the long spin diffusion length allowed these prototypes to be patterned using inexpensive and simple techniques without photo- or electron beam lithography.

RESULTS AND DISCUSSION
Spin transport length of Ge$_2$Te$_2$-Sb$_2$Te$_3$ SL
The SLs used in this work consist of layers of Sb$_2$Te$_3$ and Ge$_2$Te$_2$ atomic units. These layers are alternatively grown as thin films. The Sb$_2$Te$_3$ layer is covalently bonded with a Te-Sb-Te-Sb-Te atomic stacking sequence, whereas the units are weakly held together by the van der Waals (vdW) force. On the other hand, the Ge$_2$Te$_2$ layer has three possible polymorphs with atomic sequences: Ge-Te-Ge-Te, and Te-Ge-Te-Ge (Ge-Te-Ge-Te). These phases are respectively called Petrov, inverted-Petrov, and Ferro. The Ge$_2$Te$_2$-Sb$_2$Te$_3$ layers are bonded by the vdW interaction when both units are terminated with Te atoms. This happens when the Ge$_2$Te$_2$ is in the inverted-Petrov and Ferro-phase. However, in the case of the Petrov phase, the interface has hetero-terminations (Ge and Te), which are covalently bonded.

The Ferro-phase is thermodynamically most stable at more than 500 K and therefore most likely to exist, when the SL is grown at around 520 K. However, as the Ferro-phase is cooled to room temperature, the phase transition converts the Ferro-phase into the Petrov and Inverted-Petrov phases, and the SLs lose magnetic sensitivity. This is due to the appearance of spatial inversion...
symmetry in the Petrov and inverted-Petrov phases, which is not present in the Ferro-phase. The electronic band structure of the Ge$_2$Te$_2$-Sb$_2$Te$_3$ SL in the Ferro-phase has been reported by several groups. Due to the break of the spatial inversion symmetry, the degenerate bands are lifted, except those at the Kramers points in the reciprocal space$^{18,29,30}$. Moreover, it was reported that band splitting induces a large Rashba effect$^{19}$. Therefore, the Ferro-phase is magnetically sensitive, whereas the Petrov and inverted-Petrov phases are not. It is noteworthy that the Ferro-phase is also a W$^{25,26}$.

To increase the amount of the Ferro-phase in the SL the “thermal-annealing under magnetic field (TAUM)” process was proposed$^{22}$. However, TAUM processing requires temperature above the 470 K, which is the above the Curie temperature of many ferromagnetic materials, thus rendering it unsuitable for spin injectors. To solve this problem we have used a new method to grow Ferro-phase SLs (see “Methods”). A high-resolution and atomically resolved transmission electron microscope image of a typical SL in the Ferro-phase is given in Ref.$^{28}$. We used this new SL throughout the present work.

Herein we first demonstrate the spin transport properties of the SL at room temperature. Device-A was fabricated with TbFeCo (80 μm width) and Pt (80 μm width) electrodes, which were directly deposited on the SL through shadow masks. A microscope photograph of the device and the measurement setup are shown in Fig. 1a. TbFeCo is ferromagnetic and it was used as the spin injector. It has perpendicular magnetic anisotropy to the film surface$^{31}$. This particular ferromagnetic alloy was widely used in magneto-optical recording media (disk) in the 1990s. TbFeCo films can be deposited into an amorphous structure by sputtering at room temperature. The lack of grain boundaries in the amorphous structure greatly increases the Kerr rotation signal to noise ratio$^{32}$. For this reason, it can be expected in this work as well that the noise from the grain interface can be reduced, rather than using a crystalline ferromagnetic film. On the other hand, the Pt electrodes are used to convert spin current ($j_s$) into a charge current ($j_c$) by the inverse spin Hall (ISH) effect$^{33–35}$. The anisotropic magneto-resistance (AMR) of the TbFeCo film with a 50 nm thickness is shown in Supplementary Fig. 1. A SL structure composed of six (Ge$_2$Te$_2$)/(Sb$_2$Te$_3$) bilayer periods sandwiched between the upper and lower 5 nm-thick Sb$_2$Te$_3$ layers was used as the spin-transfer layer. The ISH voltages ($V_{ISH}$) obtained at a 100 μm distance are shown in Fig. 1b. In the vicinity of the magnetic domain transition in the spin injector (TbFeCo), $V_{ISH}$ signals are clearly changed (Fig. 1c) and reversed in the opposite magnetic field sweeping. Moreover, the polarity of the $V_{ISH}$ signal is reversed when the direction of the $j_c$ is reversed. The $V_{ISH}$ linearly increases with the $j_c$ (Supplementary Fig. 1).
Spin amplification

Theoretically, it is known that $j_s$ in non-magnetic media, such as semiconductors or metals, relies on the difference between the electrochemical potentials of up-spin ($\mu_\uparrow$) and down-spin ($\mu_\downarrow$) states at the Fermi level, and $j_s$ is given by

$$j_s = \frac{\hbar e}{2\pi} |\nabla \mu| (\mu_\uparrow - \mu_\downarrow)$$  \hspace{1cm} (1)

where $\hbar$, $e$, and $\sigma$ are Planck constant, electron charge, and conductivity, respectively. However, $j_s$ is exponentially reduced by scattering after injection. Therefore, in developing the spin current amplifier, it is important to increase the chemical potential difference (spin accumulation, $\mu_\sigma = \mu_\uparrow - \mu_\downarrow$) at both edges in the injection electrode\textsuperscript{16,31}. On the other hand, in TIs the mechanism of the $j_s$ transport is different from semiconductors and metals due to time-reversal symmetry. As reported by Kim et al.\textsuperscript{23}, the SL with magnetic sensitivity of the SL was also reported in our previous work\textsuperscript{22}.

Electrons. Thus, the WS-SL has two independent spin transport channels (up and down), and electron back scattering is prohibited by the time-reversal symmetry and the chirality of the Weyl nodes.

In TIs, instead, Berry phases and curvatures play a role in the transport. The Berry phases for $j_s$ and $j_c$ are, respectively, described in a simple two-band system with the $S$ as conservation as,

$$A^i(k) = -i\{<u_\uparrow(k)|\nabla_k|u_\uparrow(k)> + <u_\downarrow(k)|\nabla_k|u_\downarrow(k)>\}$$  \hspace{1cm} (2)

and

$$A^{S_z}(k) = -i\{<u_\uparrow(k)|\nabla_k|u_\uparrow(k)> - <u_\downarrow(k)|\nabla_k|u_\downarrow(k)>\}$$  \hspace{1cm} (3)

where $|u_\uparrow(k)>$ and $|u_\downarrow(k)>$ are the wavefunctions in each spin state. Integration of the Berry curvature $\nabla \times A^i(k)$ over the Brillouin zone is zero, whilst that of $\nabla \times A^{S_z}(k)$ is not. This implies that in terms of time-reversal symmetry $j_c$ can be tightly protected in TIs. Moreover, as the $k_z$ is non-zero at the Weyl nodes in the SL, the spin transport may spontaneously induce spin polarization.

When spins are polarized in the z-direction (normal to the SL layers), electrons are injected to the SL by flowing $j_c$ in the TbFeCo electrode (in the x-direction) and $j_s$ flows in the y direction. If the SL is a uniform WS, as discussed above, the $j_c$ polarity is switched with the $j_c$ polarity. Therefore, when the $j_c$ flows in the opposite direction in each of the multiple ferromagnetic electrodes, the $j_s$ alternately switches polarity in the $+y$ and $-y$ directions. If the electrode separations are shorter than the characteristic spin diffusion length, spin accumulation $\mu_\sigma$ can be increased by a factor of two or more. As a result, the increased chemical potential between the electrodes leads to an increase in the $j_s$ flowing out of the electrodes. In other words, such a device could function as a spin amplifier.

To confirm this scenario, we prepared three different samples (Device-B, -C, and -D) and compared the $V_{ISH}$ intensities, as shown in Fig. 3a. In Device-B, three spin injectors were connected in parallel, whereas in Device-C the injectors were connected in series. $V_{ISH}$ intensities of Device-B, -C, and -D were compared against the $j_c$ flowing per injector. As clearly seen in Fig. 3b, the $V_{ISH}$ of Device-C is amplified by a factor of two relative to the control device (D), $V_{ISH}$ (gray). The results are in good agreement with our theoretical prediction. When the electrode distances are changed from 200 to 100 $\mu$m, $V_{ISH}$ is further increased by a factor.
of two (Fig. 3b inset). The $V_{ISH}^C$ hysteresis curve is shown in Fig. 3c. Moreover, when increasing the number of the injectors from three to five in Device-C, $V_{ISH}^C$ increases by a factor of three.

In contrast, the $V_{ISH}^B$ of Device-B is decreased by a factor of three. In Device-B, $j_c$ flows in the same direction as the three parallel electrodes as indicated by the white arrow in Fig. 3a. Assuming that the density of the up-spin is larger than that of the down-spin, the up-spin flows in the $+y$ direction from the left edges in the electrodes, whereas the down-spin flows in the $-y$ direction from the right edges. Therefore, there is a net $j_c$ flow in the $+y$ direction. However, in the spaces between the electrodes, the presence of opposite spins reduces the spin potential. As a result, the overall $j_c$ flow is weakened due to the opposite spin polarities. As a result, $j_c$ direction of the central electrode is opposite to the outer electrode; thus, the same spins with $\uparrow$ or $\downarrow$ polarity are accumulated and this enhances $\mu_s$ between the electrodes. As a result, the $j_c$ flows with a high intensity. B$^1$, SL, $\bigcirc$, and $\otimes$ indicate the magnetic field direction, SL, and $j_c$ directions, respectively. Dotted lines indicate boundaries between injector edges and SLs. Black curves and yellow arrows are $\mu_s$ polarity and $j_c$ flow, respectively.

These experimental results imply that the spins with different polarities are alternatively accumulated between the injectors in the Device-C. If probes were contacted between the injectors, one would extract a much larger $j_c$, corresponding to the $\mu_s$. The device behavior is reminiscent of a spin capacitor, or spin storage device connected in series.

$V_{ISH}$ can further be amplified by increasing the number of the Ge$_2$Te$_2$ layers in the SL, as the Ferro-phase is a WS. The linear bands (Weyl nodes) bridging the bulk gap are mainly composed of $p$-electrons of Sb and Te near the vdW gap space (Supplementary Fig. 3). In contrast, the linear bands (Dirac point) in the inverted-Petrov at the $\Gamma$-point, which is a DS, are composed of $p$-electrons of Ge and Te within the Ge$_2$Te$_2$ layers (Supplementary Fig. 4). In the SL, Ge$_2$Te$_2$, and Sb$_2$Te$_3$ units, and the vdW gap spaces are alternatively stacked in the z-direction with a period of 1.8 nm, which is much shorter than the spin diffusion length shown in Fig. 1d. Therefore, the attenuation of the injected spins from the ferromagnetic electrodes is negligible. In addition, since in the SL, the $k_z$ of the Weyl nodes has an offset, the spin diffusion will increase with the number of the period. These multiple electrons confined within interfaces or the Ge$_2$Te$_2$ layers play a role as spin transport channels. Figure 4a shows this effect using Device-C with three spin injectors. The amplification exponentially increases with the number of the Ge$_2$Te$_2$ layers. Indeed, a five-fold increase in $j_c$ relative to a device with a single injector for a SL with 12 bilayer periods. The SL $V_{ISH}$ amplification gain is compared against a 4.8 nm-thick single layer of GeTe and with single layer of 6.0 nm-thick Sb$_2$Te$_3$ layer. These layer thicknesses correspond to an equivalent SL with $n = 6$ periods. These single layers showed $\leq 100\%$ gain. In the devices, as the transport channels are limited to one (bulk for GeTe) or two (surfaces for Sb$_2$Te$_3$) respectively, both $V_{ISH}$ are not amplified with the thickness. These results clearly imply that the number of the alternatively stacked Sb$_2$Te$_3$ and Ge$_2$Te$_2$ layers, as shown in Fig. 4b are responsible for the $V_{ISH}$ amplification effect. In other words, the spin behavior in the SLs is attributed to the periodicity effect on the electronic band structure. We attribute the long spin diffusion length observed in our devices to the multiple spin transport channels that form due to the periodic arrangement of Ge$_2$Te$_2$ layers and the Ferro-phase SL, which was grown with a high crystal quality.
Inversion symmetry, the Sb2Te3 layer is followed by the Ferro-phase of Ge2Te2, the Ge-Te-Ge-Te changes by that of a single layer of Te or Sb atoms. When the bottom atomic order is stably formed, which corresponds to the yellow panel of Fig. 5. In the "0" state, as the SL breaks the spatial inversion symmetry, $j_i$ flows. In this example we assume, the direction is to the left. Since the Ge-Te-Ge-Te atomic order is ferroelectric, the electric dipole is directed from top to bottom and the vdW gap position moves up to the interface with the second Sb2Te3 layer. When a half layer is formed on the completed Sb2Te3 layer ("1/2" state in the panel), both Sb and Te atoms may terminate the structure. However, the mixture cancels the electric dipoles. When the first layer is formed ("1" state in the panel) next, only the Te layer can break the symmetry, whereas the Sb layer remains unchanged. It is noteworthy that the dipole direction is reversed, the "3/2" state is the same as "1/2" state, and the $j_i$ of the "2" state has the same direction as that of state "0," as the vdW gap is moved up to the top of the Ge2Te2 layer. In the case that the vdW gap position is adjacent to the Ge2Te2 layer, $V_{ISH}$ becomes high. Density functional calculations demonstrated that the gap position and the dipole direction shift the Fermi level up or down near the Weyl points in the band structure.

From these analysis, it is interesting to consider whether the vdW gap plane is terminated using either -Ge-Te or -Sb-Te. As Kim et al. suggested, the surface termination of the surface by -Ge-Te or -Sb-Te determines the Weyl node properties. As the energy level of the Weyl node pairs has an offset in the $\pm k_x$ direction, the interface termination presumably affects the spin polarization and the $j_i$ polarity. This effect remains to be clarified in a detailed analysis of Weyl semimetals.

In summary, Ge2Te2-Sb2Te3 SLs, which consist of an insulator and a TI exhibit a topologically protected spin diffusion length of more than 100 $\mu$m at room temperature. Using TbFeCo electrodes connected in series, a 15-fold increase in the spin current was possible by accumulating spin between the electrodes by using a SL with 12 periods. The results suggest an exponential increase in spin amplification with the number of SL bilayers. Indeed, if the number of SL periods could be stacked to micrometer thicknesses, a large amount of spin could be accumulated not only in the plane of the electrodes but also within the 3D SL, and this will lead to a large spin current. We previously demonstrated that a cross-point device using the same SL can accumulate spins at zero current for a short period after flowing current perpendicular to the surface. This topologically protected long spin diffusion length is very appealing for spintronics device designers only one spin-injection source (battery) is required, and this will substantially simplify spintronics device designs. The short diffusion lengths of existing spin transport media requires each spintronic device, such as MRAM, to have its own spin current source. Thus, a large number of charge current lines need to be integrated into the device and this severely complicates the circuit design. Moreover, this multiple spin source approach wastes energy.
contrast, if one uses the topologically protected long spin diffusion length media, which we demonstrated here, a single spin generator or battery can supply spin current to all spintronic components. This will simplify the circuit design and lower the device energy consumption. For this reason, we believe that Ni-Ti SLs will become indispensable spin transport media and enable the practical and efficient spintronics devices. In addition, the spin current supplied from the spin generator and rectifier, which we demonstrated here, may also open a new field of spintronics in quantum computation.

METHODS

Materials and devices
Spin transport devices were fabricated on sapphire substrates, using a sequence of metal stencil masks. After forming a 40 nm-thick amorphous Si film at room temperature, a 5 nm-thick SbTe, seed layer, GeTe–SbTe, SLS were deposited by RF-magnetron sputtering at 480 K, using a GeTeS (0.45 : 0.52 : 0.03) and a SbTe(0.3 : 0.7) target, respectively. Due to the absence of a SbTe seed layer, the SbTe(0001) axis and GeTe(111) axis were aligned perpendicular to the substrate allowing the SLS to be grown by vdW epitaxy at a couple of hundred degrees above room temperature. Doping (3%-5%) is effective to hold the Ferro-phase stably even at room temperature.28,38. The perpendicular to the substrate allowing the SLs to be grown by vdW epitaxy at khorst.

transport measurements, additional 4QLs were deposited on top of the SL (total 5QLs). Finally, using other metal masks, TbFeCo (22.79 : 67.17 : 10.04) was deposited. The resistances between the electrodes for spin injection and Pt electrodes for detection were separately shown in Supplementary Fig. 2. Impedance contacts were made using polymer-filled gaps and Pt BAR contacts. The volumetric resistances were measured using a four-probe method (0.5 μm thickness). The electrical measurements were performed at room temperature with a current of 0.1 μA.

Device measurements.
ISHE measurements were performed at room temperature (300 K) in vacuum using a Hall resistance measurement system (4800 series) from TOYO Corp. The spin signals were measured ten times and then averaged for every 0.01 Tesla magnetic field step between −0.55 and +0.55 T.

DATA AVAILABILITY
The raw data used in this study are available upon reasonable request to the corresponding author.

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AUTHOR CONTRIBUTIONS

H.A., N.M., S.M., and J.T. coordinated the project. J.T. fabricated the superlattice devices. S.S., H.A., and J.T. measured the spin signals. S.M. contributed the theoretical parts. J.T. wrote the manuscript.

COMPETING INTERESTS

The authors declare no competing interests.