Giant tunable spin Hall angle in sputtered Bi$_2$Se$_3$ controlled by an electric field

Qi Lu$^{1,3}$, Ping Li$^{2,3}$, Zhixin Guo$^{2}$, Guohua Dong$^{1}$, Bin Peng$^{1}$, Xi Zha$^{1}$, Tai Min$^{2}$, Ziyao Zhou$^{1}$ & Ming Liu$^{1}$

Finding an effective way to greatly tune spin Hall angle in a low power manner is of fundamental importance for tunable and energy-efficient spintronic devices. Recently, topological insulator of Bi$_2$Se$_3$, having a large intrinsic spin Hall angle, show great capability to generate strong current-induced spin-orbit torques. Here we demonstrate that the spin Hall angle in Bi$_2$Se$_3$ can be effectively tuned asymmetrically and even enhanced about 600% reversibly by applying a bipolar electric field across the piezoelectric substrate. We reveal that the enhancement of spin Hall angle originates from both the charge doping and piezoelectric strain effect on the spin Berry curvature near Fermi level in Bi$_2$Se$_3$. Our findings provide a platform for achieving low power consumption and tunable spintronic devices.
riving magnetization switching by current-induced spin-orbit torque (SOT) is one of the most promising means to achieve magnetic random access memories (MRAM) due to the merits in ultrafast, nonvolatilite, and energy-efficient operation. In a typical SOT-driven device, the in-plane charge current is converted into spin current in the heavy metal layer via the spin Hall effect. The spin current further injects into the adjacent ferromagnetic layer, exerting torque on the magnetic moment and driving magnetization switching. The power consumption of SOT-MRAM devices can be further reduced by improving the charge-spin conversion efficiency in the non-magnetic spin Hall layer or enhancing the spin current transmission at the nonmagnetic/ferromagnet interface. Here, the spin Hall angle defined as the ratio of spin current density to charge current density is crucial in SOT devices. Pursuing materials with a large spin Hall angle is a prerequisite for achieving low power consumption SOT devices. In this context, topological insulators, 2D materials, antiferromagnets, and Weyl semimetals with novel quantum states have been actively investigated. Topological insulators such as Bi₂Se₃ and Bi₂Te₃ show an exceptionally large spin-charge conversion efficiency than conventional heavy metals due to the helical spin-momentum locking in the surface states. Recently, magnetron sputtered Bi₂Se₃ films exhibiting both large spin Hall angle and highly compatible with industrial production, are regarded as optimal materials for the SOT switching. Despite these exciting prospects, topological insulators are problematic because of their relative high resistivities. Exploring interesting approaches to further improve energy efficiency is a challenge and opportunity.

The electric field-induced mechanical strain has been demonstrated to effectively mediate the magnetic anisotropy, exchange coupling, spin structure, and SOT magnitude in the ferromagnetic layer. In addition, the surface charge doping related to the ferroelectric polarization will also contribute to the tuning effect. Due to the dielectric nature of ferroelectric materials, the electric field can be applied without the leak current. Hence the Joule heating is suppressed in electric field controlling, which is suitable for low energy consuming information technology. However, in the spin Hall materials, the electric field control of the charge-spin conversion ratio has been rarely studied. Considering that the intrinsic spin Hall conductivities are strongly dependent on the position of Fermi level, which can be tuned by the charge doping and strain. The strain effect usually shows a symmetric tuning effect with respect to the positive and negative electric fields due to the same lattice deformation, while the charge doping effect always shows an asymmetric tuning effect. Thus building the multifunctional hybrid devices by introducing electric field controlling is particularly attractive to obtaining tunable spin sources and further reducing the power consumption of SOT-MRAM devices.

In this work, the electric field dramatic tuning of spin Hall angle in Bi₂Se₃ films sputtered on the piezoelectric substrates was demonstrated. The values of spin Hall angle under different electric fields are measured using an in situ planar Hall system. With applying an electric field, the charge-spin conversion efficiency was significantly enhanced in a reversible and reproducible manner. The maximum spin Hall angle was determined to be 13.45 at $E = -12$ kV/cm which is six times larger than the initial state. It is interesting to find the spin Hall angle changed asymmetrically while a bipolar electric field was applied across the piezoelectric substrate. By conducting DFT calculation and related control experiments, such great tunability as well as the asymmetrical tuning effect are considered to be originating from the combination effect of the mechanical strain and the interfacial charge doping between Bi₂Se₃ and PMN-PT. It turns out the mechanical strain enhances the spin Hall angle symmetrically, whereas the charge doping changes the spin Hall angle in an asymmetrical way.

Results

The low magnification high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) image gives an overview of the PMN-PT/Bi₂Se₃(8 nm)/NiFe(8 nm)/TaOₓ(1 nm) structure, as shown in Fig. 1a. The thickness of the layers is similar to the nominal ones. The interface of each layer is smooth and continuous, which indicating the uniform growth of all layers in the stack (see Supplementary Fig. 6). Figure 1b shows the high-resolution HAADF-STEM image of the film. The sharp lattice fringes clearly seen in the image reveal the existence of good crystallinity in the Bi₂Se₃. The Bi₂Se₃ film exhibits a mixing of polycrystalline and amorphous state as a whole (see Supplementary Fig. 7). This result is consistent with the previous report on the sputtered Bi₂Se₃ films. We conducted the X-ray photoelectron spectroscopy (XPS) measurement to determine the chemical composition of PMN-PT/Bi₂Se₃ (20 nm) structure. Figure 1c, d shows the Bi 4f core-level spectra and Se 3d core-level spectra, respectively. For Bi 4f core-level spectra, the peaks of Bi 4f½ and 4f½ energy levels are observed at binding energies about 156.36 eV and 161.72 eV. The Bi 4f levels show a spin-orbit splitting of 5.36 eV, which is consistent with the previous report. Here two other components with the binding energy of 157.08 eV and 162.47 eV nearby the original Bi 4f peaks are observed. Those peaks corresponding to the Bi-O bound are due to the little oxidation when measuring the XPS spectra. Two peaks correspond to the Se 3d½ and 3d¾ with binding energies of 51.56 eV and 52.42 eV appear at the Se 3d core-level spectra. The splitting of Se 3d level is determined to be 0.86 eV, comparable to the previous experimental studies. By considering the integrated peak intensity with the sensitivity factor, the surface atomic composition ratio of Se and Bi is estimated to be 1.43 (see Supplementary Fig. 8 and Supplementary Note 7). The composition is close to the target, showing the films are normally stoichiometric. To examine the surface roughness of the samples, we conducted the atomic force microscopy (AFM) measurement on the PMN-PT/Bi₂Se₃(8 nm)/NiFe(8 nm)/TaOₓ(1 nm) sample, as shown in Fig. 1c. The surface roughness Ra is determined to be 0.489 nm, indicating a smooth surface and high film quality which is sufficient for the further SOT study (see Supplementary Fig. 10).

Here we determine the current-induced SOT effective fields by angular field characterization of the planar Hall resistance at positive and negative testing current. The damping-like torque induces the perpendicular effective field $H_{\text{oop}}$, and the field-like torque induces the in-plane transverse effective field $H_T$, as shown in Fig. 2a. According to the Slonczewski equation, the perpendicular effective field $H_{\text{oop}}$ can be expressed as $-\mu H_{\text{oop}} = \mu H_{\text{oop}}(I_B m \times m)$, where $\mu$ is the spin polarization vector, $m$ is the magnetization vector, $I_B$ represent the magnitude of spin current density, and $m$ is the thickness of the ferromagnetic layer. The Hall resistance is directly modified by the small magnetization deviation induced by the SOT effective fields. The direction of the $H_{\text{oop}}$ and $H_T$ relies on the sign of current. Hence the component of SOT effective fields in angular-dependent planar Hall resistance is an odd function of current, whereas other components are even function with respect to the testing current. We take the differential resistance $R_{\text{DH}} = R_{\text{H}}(I_B) - R_{\text{H}}(-I_B)$ to extract the contribution from the SOT effective fields and cancel out other effects such as Joule heating. The inset illustrates the structure of the Hall device and the measurement configuration. The Hall resistance $R_{\text{H}}(I_B)$ is measured using the bipolar input current and rotating the constant 400 Oe in-plane external magnetic field $H_{\text{ext}}$. Here $I$ is the input current along
the $x$-direction, and $\theta$ represents the angle between the current and the magnetic field.

Figure 2b shows the typical planar Hall resistance $R_{\text{H}}$ as a function of $\theta$ measured at $\pm 10$ mA and the corresponding $R_{\text{DH}}$ curve for Bi$_2$Se$_3$(8 nm)/NiFe(8 nm)/TaO$_x$(1 nm) structure on thermally oxidized Si substrate. Here we assume that the top 1 nm Ta capping layer is fully oxidized and has almost no impact on the spin transport properties$^{31,32}$. The value of $H_{\text{OOP}}$ and $H_T$ can be obtained by fitting the following equation

$$R_{\text{DH}}(I, \theta) = \frac{2R_{\text{H}}(H_T + H_{\text{OOP}})}{H_{\text{ext}}} \cos \theta + C$$

where $C$ is the resistance offset, $R_{\text{AHE}}$ is the anomalous Hall resistance, and $H_{\text{perp}}$ is the magnetic field along the $z$ direction when measuring the anomalous Hall resistance. The Oersted field contribution $H_{\text{Oe}}$ is estimated to be $6.66 \times 10^{-3}$ Oe mA$^{-1}$ calculated by Ampere’s law with considering the current shutting effect (see Supplementary Note 9). The $R_{\text{H}}$ is the magnitude of $\sin(2\theta)$ component in the corresponding planar Hall resistance curve. Here the value of $\frac{dR_{\text{AHE}}}{dH_{\text{perp}}}$ is determined to be $3.175 \times 10^{-5}$ Oe by taking the slope of $R_{\text{AHE}}$ versus $H_{\text{perp}}$ at a small 1 mA testing current (see Supplementary Note 3).

After determining the value of $R_{\text{H}}$, $H_{\text{Oe}}$, and $\frac{dR_{\text{AHE}}}{dH_{\text{perp}}}$ parameters, the effective field $H_{\text{OOP}}$ and $H_T$ can be obtained by fitting the angular-dependent $R_{\text{DH}}$ data into Eq. (1). Figure 2c shows the $R_{\text{DH}}$ results and the fitting curves at different testing currents. The fitting curves are in good agreement with the experimental data. The amplitude of $R_{\text{DH}}$ curve increases with increasing the testing current, indicating the contribution from the current-induced SOT effective fields. Figure 2d shows the linear fitting result of SOT effective field $H_{\text{SOT}}$ including $H_{\text{OOP}}$ and $H_T$ with respect to the testing current. The effective spin Hall angle can be calculated by

$$\theta_S = \frac{2eM_sJ_{\text{FM}}H_{\text{OOP}}}{hJ_{\text{NM}}}$$

where $M_s$ is the saturation magnetization and equal to 663.45 emu/cm$^3$ (see Supplementary Fig. 4), $t_{\text{FM}}$ is the thickness of the ferromagnetic layer, and $J_{\text{NM}}$ is the current density in the nonmagnetic layer. The value of $H_{\text{OOP}}$ is obtained by the slope of the linear fitting result. The spin Hall angle of our 8 nm Bi$_2$Se$_3$ is determined to be 2.18, comparable with the previous study on sputtered Bi$_2$Se$_3$ and the ST-FMR analysis (see Supplementary Note 1). This ensures that the measurements and analysis of spin Hall angle are performed precisely and reliable.

We perform the in situ planar Hall measurements at different electric fields to characterize the electric field control spin Hall angle of Bi$_2$Se$_3$. The Hall device with the same multilayer structure is fabricated on a PMN-PT substrate that applies the electric field and generates mechanical strain, as demonstrated in Fig. 3a. The electric field is applied perpendicular to the PMN-PT substrate up to $\pm 12$ kV/cm. The representative $R_{\text{DH}}$ results measuring at $\pm 6$ mA testing current under $0$ kV/cm, $-12$ kV/cm, and $0$ kV/cm back electric field are shown in Fig. 3b. The constant baseline of the results is subtracted to directly compare the amplitude and line shape of the $R_{\text{DH}}$ curves under different electric fields. The $R_{\text{DH}}$ curves under $-12$ kV/cm exhibit a larger amplitude than that of $0$ kV/cm, indicating the increment of the SOT-induced effective fields. Specifically, the peaks around $115^\circ$ and $245^\circ$ nearly disappear under the electric field at $-12$ kV/cm. Such dramatic change in the line shape is due to an increase of the $\cos(\theta)$ component to the total line shape, which corresponds to a remarkable enhancement of perpendicular effective field $H_{\text{OOP}}$. Note that there are no obvious changes in the $R_{\text{DH}}$ curves between $0$ kV/cm and $0$ kV/cm back, implying a reversible switching behavior.

The $H_{\text{OOP}}$ versus the testing current from $0$ kV/cm to $-12$ kV/cm is presented in Fig. 3c. The determined effective fields are
The solid curve represents the difference of the Hall resistances $R_{DH}$ at positive and negative currents. The angular-dependent $R_{DH}$ measured at different input currents. Each solid curve represents the fitting result. d) SOT-induced effective fields $H_{OOP}$ and $H_T$ as a function of applied current. The lines are the linear fitting results.

Proportional to the testing current, and the linear fitting agrees with the data well. The values of $\frac{T_{max}}{T_{min}}$ derived from the linear fitting are $1.227 \times 10^{-9}$ Oe/(A m)$^{-2}$ and $7.976 \times 10^{-9}$ Oe/(A m)$^{-2}$ under 0 kV/cm to $-12$ kV/cm, respectively. Figure 3d demonstrates the spin Hall angles under different electric fields applied across the PMN-PT substrate. Interestingly, it is found that the effective field-dependent spin Hall angle is significantly asymmetric with respect to the zero field. The spin Hall angle of Bi$_2$Se$_3$ has been enhanced by using both positive and negative electric fields. Especially, the spin Hall angle of Bi$_2$Se$_3$ exhibits a significant increment up to 13.45 under $-12$ kV/cm, which is about 6 times larger than the initial state.

Generally speaking, two effects from the PMN-PT substrate are expected to be responsible for the change of the spin Hall angle in Bi$_2$Se$_3$, i.e., strain effect and charge doping effect. In order to clarify the mechanism of the tuning result, it is necessary to directly distinguish the strain effect and charge doping effect. Inserting a charge dissipation layer such as 5 nm Cu between PMN-PT substrate and Bi$_2$Se$_3$ layer is an effective way to separate these two effects. The charge doping effect can be excluded in this condition since the charge will be dissipated in the Cu layer. Here we further perform the same study with structure PMN-PT/Cu(5 nm)/Bi$_2$Se$_3$(8 nm)/NiFe(8 nm)/TaO$_x$(1 nm). Figure 4 gives a comparison of electric field control spin Hall angle between PMN-PT/Bi$_2$Se$_3$/NiFe structure and PMN-PT/Cu/Bi$_2$Se$_3$/NiFe structure. After inserting the Cu dissipation layer between Bi$_2$Se$_3$ and PMN-PT, the electric field control spin Hall angle in Bi$_2$Se$_3$ shows a symmetry behavior at positive and negative electric fields. That is due to the almost equivalent lattice deformation induced by the piezoelectric strain. After separating the strain effect, the higher enhancement of spin Hall angle in PMN-PT/Bi$_2$Se$_3$/NiFe structure at the negative electric field can be attributed to the contribution of the charge doping effect.

To theoretically understand the tuning mechanism, we further investigate the external electric field effect on the spin Hall angle of Bi$_2$Se$_3$ by first-principle calculation. The spin Hall conductivity (SHC) of Bi$_2$Se$_3$ is proportional to the spin Hall angle, as a function of in-plane strain and charge doping have been studied respectively.

We adopt the maximally localized Wannier functions with Wannier90 in combination with the VASP package to calculate the SHC. Firstly, we calculate the SHC of bulk Bi$_2$Se$_3$ with R-3m space group and find its SHC is $38.58 \ (\hbar/e) S/cm$, which is mainly attributed to the electrons around the Fermi level nearby the $\Gamma$ point in the Brillouin zone (Fig. 5a). It is known that the electric field induces biaxial strain in Bi$_2$Se$_3$ via PMN-PT.
substrate\(^4\). Thus, we systematically investigate the relationship between SHC of Bi\(_2\)Se\(_3\) and the biaxial strain. As shown in Fig. 5c, both the tensile and compressive strains can induce the increment of SHC. Moreover, the tensile and compressive strains tune the SHC almost symmetric when the strain is small (<0.25%). When the strain becomes larger, i.e., to the maximum value of our experimental data 0.5%, the asymmetric SHC appears. Nevertheless, the asymmetric difference is less than 20%, much smaller than the experimental results where three times asymmetric is observed (Fig. 3d). This result is consistent with the experimental observation where the charge transfer effect is excluded (Fig. 4).

To further explore the charge transfer effect induced by the PMN-PT substrate, we additionally calculate the electronic structure of a heterostructure composed of Bi\(_2\)Se\(_3\) and PbTiO\(_3\)(001) (a simplified model to PMN-PT). Considering that the optimized lattice constant of Bi\(_2\)Se\(_3\) is 4.16 Å and that of PbTiO\(_3\) is 3.90 Å, the 1 \(\times\) \(\sqrt{2}\) unit cell of Bi\(_2\)Se\(_3\) is commensurate to the 1 \(\times\) 2\(\sqrt{2}\) surface unit cell of PbTiO\(_3\)(001). Figure 5b shows the calculated differential charge density distributions for TiO\(_x\) terminal Bi\(_2\)Se\(_3\)/PbTiO\(_3\)(001) heterostructure. It is seen that obvious electron transfer from PbTiO\(_3\) to Bi\(_2\)Se\(_3\) occurs when PbTiO\(_3\) is under the negative electric field, whereas there is almost no electron transfer when the electric field is switched to positive. Note that the above results have been confirmed by the DFT calculations based on the HSE06 functional\(^{12,43}\), which is well known to give rise to reliable band gaps in semiconductors (see Supplementary Fig. 11). In addition, we have considered the vacancy defect effect on the charge transfer between Ni and Fe elements. The effect on the charge transfer between Ni and Fe elements. The charge transfer process is under the negative electric field. Whereas, when the electric field is reversed, there is almost no charge transfer between PbTiO\(_3\) and Bi\(_2\)Se\(_3\). This result is similar to the case without the metallic covering layer. We have further calculated the averaged charge density difference along the out-of-plane \(z\) direction denoted as \(\Delta p(z)\) by integrating it over the \(x\)-\(y\) plane (see Supplementary Fig. 13). It is found that the Se atoms at the interface have a nature of obtaining electrons from Fe atoms when no external electric field is applied. We should note that the negative external electric field further benefits the electron transfer from the Fe layer to Bi\(_2\)Se\(_3\). These results confirm that the additional electrons transferred from PMN-PT to Bi\(_2\)Se\(_3\) can be preserved in Bi\(_2\)Se\(_3\).
We have then calculated the SHC of bulk Bi$_2$Se$_3$ as a function of the charge doping concentration (-e/unit-cell) under 0.5% biaxial strain. As shown in Fig. 5d, the SHC significantly increases with the increase of doped charge concentration from $-0.1$ to $-0.7$ -e/unit-cell, but it decreases from $0$ to $0.3$ -e/unit-cell. Such a significant increase of SHC with charge doping can be due to the upshift of the Fermi level, which involves the contribution of conduction electrons around the $\Gamma$ point to the spin Berry curvature. Especially, the SHC becomes about three times as that without electron transfer under 0.5% strain when the charge doping is about $-0.7$ -e/unit-cell. These results show that the asymmetry of the spin Hall angle is mainly from the charge transfer effect.

**Discussion**

In conclusion, we have demonstrated a giant (more than 600%) and reversible manipulation of spin Hall angle in sputtered Bi$_2$Se$_3$ films solely by electric fields across the PMN-PT piezoelectric substrate. The spin Hall angle and SOT effective field are evaluated using the angular-dependent planar Hall measurement with the in situ electric field. We find that applying an electric field enhances the spin Hall angle from $2.18$ to $13.45$. Moreover, we conduct the first-principles calculation study on the intrinsic spin Hall conductivity in a relevant system to reveal the modulation mechanism. We find this remarkable modulation of spin Hall angle is achieved by the co-mediating of electric-field-induced strain and surface charge. Our work experimentally realizes a reversible, wide range, low power consumption manner to control and even enhances the spin Hall angle in sputtered Bi$_2$Se$_3$. Such controllable manipulation of spin Hall angle with exceptionally large enhancement utilizing electric field points to

**Fig. 5 Spin Hall conductivity calculation for Bi$_2$Se$_3$ structure.**

a) Band structure projected by spin Berry curvature on a log scale (up panel) and the $k$-resolved spin Berry curvatures (down panel) for the 0.5% biaxial strain bulk Bi$_2$Se$_3$. b) Charge density difference of heterostructure of Bi$_2$Se$_3$ on TiO-terminal PbTiO$_3$(001) surface. The picture on the left shows the application of the negative external electric field (along the $z$ direction) on the heterostructure. Simultaneously, the built-in electric field along the $−z$ direction is generated in PbTiO$_3$. The polarization of PbTiO$_3$ is $P_\uparrow$. The picture on the right is the opposite. The polarization of PbTiO$_3$ is $P_\downarrow$. The red and blue areas indicate loss and gain electrons, respectively. The Pb, Ti, O, Bi, and Se atoms are depicted by gray, light blue, red, magenta, and green balls, respectively. c) SHC as a function of the biaxial strain for bulk Bi$_2$Se$_3$. d) SHC as a function of the charge doping for the bulk Bi$_2$Se$_3$ under 0.5% biaxial strain. The initial state of Bi$_2$Se$_3$ without strain and charge doping is marked with the blue hexagon symbol.
opportunities to enable energy-efficient SOT switching and tunable spintronic devices.

Methods

Film growth. Substrate/Bi$_2$Se$_3$(8 nm)/NiFe(8 nm)/TaO$_x$(1 nm) multilayers were deposited onto thermally oxidized Si and (011)-oriented PMN-PT substrates by DC magnetron sputtering at room temperature with a base pressure less than 1 × 10$^{-7}$ Torr. The background Ar working pressure was 3 mTorr for depositing all the layers. An in situ quartz crystal microbalance monitored the deposition rates.

Device fabrication. The multilayer stacks were patterned into cross Hall bars and rectangular bars for planar Hall measurement and ST-FMR measurement respectively using standard maskless ultraviolet photolithography and lift-off procedure. The second step of photolithography was employed to fabricate the Cr(5 nm)/Au(100 nm) contacts.

Characterization. The planar Hall measurements were performed with Keithley 6220 DC current source and Keithley 2617B electrometer. A cross-sectional TEM sample was prepared by the focused ion beam (FIB, Tescan LYRA 3). The crystal structure of the film was investigated using a probe aberration-corrected scanning transmission electron microscopy (Cs-STEM, Themis Z G2 300 kV, FEI). All of the measurements were performed at room temperature.

DFT calculations. Our calculations are performed using the Vienna ab initio Simulation Package (VASP) based on DFT.$^{37,38}$ The projector augmented wave method and a plane-wave basis set are performed. The electron exchange-correlation functional is described by the generalized gradient approximation of the Perdew–Burke–Ernzerhof functional and the HSE06 hybrid functional.$^{39}$ The plane-wave cutoff energy is chosen to be 500 eV. Moreover, at $\Gamma$-centered $\mathbf{k}$ mesh of 24 × 24 × 3 are adopted in the self-consistent calculations. To better describe the vdW interaction, the optB86b-vdW functional is adopted.$^{40,41}$ Then, DFT wave functions are projected to maximally localized Wannier functions using the WANNIER90 package,$^{39}$ and the Kubo formula is performed to calculate the SHC.

The codes are available from the corresponding author upon reasonable request.

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Author contributions
M.L., Z.Z., and Q.L. conceived and designed the experiments. P.L., Z.G., and T.M. carried out the first-principle calculations. Q.L. fabricated the sample and performed the experiments with help of X.Z. G.D. characterized the film structure with TEM. Q.L. conducted the ST-FMR measurements under the guidance of B.P. Q.L., and P.L. wrote the manuscript with modification from M.L. and Z.G. All authors contributed to the discussion of the results.

Competing interests
The authors declare no competing interests.

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Correspondence and requests for materials should be addressed to Zhixin Guo, Guohua Dong or Ming Liu.

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