Supplementary Figures:

Supplementary Figure 1. HAADF-STEM image of the Bi2Se3/NiFe (Py) heterostructures on Al₂O₃ substrate. The uniform and good layered structure in Bi₂Se₃ and a clean and smooth interface between $Bi₂Se₃$ and Py layer are observed. The white scale bar is 2 nm.

Supplementary Figure 2. Bi2Se³ film electrical transport property. Sheet resistance, *R*s, f_{Bise} (QL)
Supplementary Figure 2. Bi2Se3 film electrical transport property. Sheet resista
for different Bi2Se3 thicknesses measured at room temperature.

Supplementary Figure 3. Schematic of energy dispersion in *k***-space and band bending at Bi2Se³ surfaces.** Discrete subbands are formed in 2DEG at the Bi2Se³ surface due to the quantum confinement effect. For clarity the Rashba splitting is not illustrated in the 2DEG subbands.

Supplementary Figure 4. Estimated *n***2D with varying the value of** *V***F. a**, Sheet carrier concentrations of TSS (n_{TSS}), 2DEG (n_{2DEG}) and bulk ($n_{2D-Bulk}$) estimated with $V_F = 4.6 \times 10^5$ m s⁻¹. **b**, with $V_F = 5 \times 10^5$ m s⁻¹. **c**, with $V_F = 5.4 \times 10^5$ m s⁻¹, by fixing $m^* = 0.14$ m₀, E_{SCBM} – $E_{\text{DP}} = 200 \text{ meV}$ and $E_{\text{F}} - E_{\text{CBM}} = 100 \text{ meV}$ for different Bi₂Se₃ thicknesses at room temperature. The data with black squares represent the measured sheet carrier concentrations in Bi2Se³ films at room temperature.

Supplementary Figure 5. Estimated n_{2D} **with varying the value of** m^* **. a, Sheet carrier** concentrations of TSS (n_{TSS}), 2DEG (n_{2DEG}) and bulk ($n_{\text{2D-Bulk}}$) estimated with $m^* = 0.11 m_0$. **b**, with $m^* = 0.13m_0$. **c**, with $m^* = 0.14m_0$. **d**, with $m^* = 0.15m_0$. **e**, with $m^* = 0.17m_0$, by fixing V_F $= 5 \times 10^5$ m s⁻¹, $E_{\text{SCBM}} - E_{\text{DP}} = 200$ meV and $E_{\text{F}} - E_{\text{CBM}} = 100$ meV for different Bi₂Se₃ thicknesses at room temperature. The data with black squares represent the measured sheet carrier concentrations in Bi₂Se₃ films at room temperature.

Supplementary Figure 6. Estimated n_{2D} **with varying the value of** $(E_{SCBM} - E_{DP})$ **. a, Sheet** carrier concentrations of TSS (n_{TSS}), 2DEG (n_{2DEG}) and bulk ($n_{2D-Bulk}$) estimated with (E_{SCBM} $-E_{DP}$) = 150 meV. **b**, with ($E_{SCBM} - E_{DP}$) = 200 meV. **c**, with ($E_{SCBM} - E_{DP}$) = 250 meV, by fixing $V_F = 5 \times 10^5$ m s⁻¹, $m^* = 0.14$ m_0 and $E_F - E_{CBM} = 100$ meV for different Bi₂Se₃ thicknesses at room temperature. The data with black squares represent the measured sheet carrier concentrations in Bi₂Se₃ films at room temperature.

Supplementary Figure 7. Estimated n_{2D} **with varying the value of** $(E_F - E_{CBM})$ **. a, Sheet** carrier concentrations of TSS (n_{TSS}), 2DEG (n_{2DEG}) and bulk ($n_{2D-Bulk}$) estimated with (E_F – E_{CBM}) = 50 meV. **b**, with $(E_{\text{F}} - E_{\text{CBM}}) = 100$ meV. **c**, with $(E_{\text{F}} - E_{\text{CBM}}) = 120$ meV. **d**, with $(E_F - E_{\text{CBM}}) = 150 \text{ meV}$, by fixing $V_F = 5 \times 10^5 \text{ m s}^{-1}$, $m^* = 0.14 \text{ m}_0$ and $E_{\text{SCBM}} - E_{\text{DP}} = 200$ meV for different Bi2Se³ thicknesses at room temperature. The data with black squares represent the measured sheet carrier concentrations in Bi₂Se₃ films at room temperature.

Supplementary Figure 8. Estimated interface SOT efficiency (λ rss and amended λ rss) with varying the value of V_F . a, Interface SOT efficiency estimated with $V_F = 4.6 \times 10^5$ m s⁻¹. **b**, with $V_F = 5 \times 10^5$ m s⁻¹. **c**, with $V_F = 5.4 \times 10^5$ m s⁻¹, by fixing $m^* = 0.14$ *m*₀, $E_{SCBM} - E_{DP} = 0.14$ 200 meV and $E_F - E_{\text{CBM}} = 100$ meV for different Bi₂Se₃ thicknesses at room temperature. The amended λ_{TSS} denoted by red circles represents the interface SOT efficiency from TSS after excluding the opposite 2DEG contribution. The error bars are the standard deviation from three devices at each Bi₂Se₃ thickness.

Supplementary Figure 9. Estimated interface SOT efficiency (λ TSS and amended λ TSS) with varying the value of m^* . a, Interface SOT efficiency estimated with $m^* = 0.11m_0$. b, with $m^* = 0.13m_0$. **c**, with $m^* = 0.14m_0$. **d**, with $m^* = 0.15m_0$. **e**, with $m^* = 0.17m_0$, by fixing V_F $= 5 \times 10^5$ m s⁻¹, $E_{\text{SCBM}} - E_{\text{DP}} = 200$ meV and $E_{\text{F}} - E_{\text{CBM}} = 100$ meV for different Bi₂Se₃ thicknesses at room temperature. The amended λ_{TSS} denoted by red circles represent the interface SOT efficiency from TSS after excluding the opposite 2DEG contribution. The error bars are the standard deviation from three devices at each Bi₂Se₃ thickness.

Supplementary Figure 10. Estimated interface SOT efficiency (λ rss and amended λ rss) with varying the value of $E_{\text{SCBM}} - E_{\text{DP}}$. a, Interface SOT efficiency estimated with $E_{\text{SCBM}} E_{\text{DP}} = 150 \text{ meV}$. **b**, with $E_{\text{SCBM}} - E_{\text{DP}} = 200 \text{ meV}$. **c**, with $E_{\text{SCBM}} - E_{\text{DP}} = 250 \text{ meV}$, by fixing $V_F = 5 \times 10^5$ m s⁻¹, $m^* = 0.14$ *m*₀ and $E_F - E_{CBM} = 100$ meV for different Bi₂Se₃ thicknesses at room temperature. The amended λ TSS denoted by red circles represent the interface SOT efficiency from TSS after excluding the opposite 2DEG contribution. The error bars are the standard deviation from three devices at each Bi₂Se₃ thickness.

Supplementary Figure 11. Estimated interface SOT efficiency (λ rss and amended λ rss) **with varying the value of** $E_F - E_{CBM}$ **. a,** Interface SOT efficiency estimated with $E_F - E_{CBM}$ $= 50$ meV. **b**, with $E_F - E_{CBM} = 100$ meV. **c**, with $E_F - E_{CBM} = 120$ meV. **d**, with $E_F - E_{CBM} = 100$ 150 meV, by fixing $V_F = 5 \times 10^5$ m s⁻¹, $m^* = 0.14$ m_0 and $E_{SCBM} - E_{DP} = 200$ meV for different Bi₂Se₃ thicknesses at room temperature. The amended λ_{TSS} denoted by red circles represent the interface SOT efficiency from TSS after excluding the opposite 2DEG contribution. The error bars are the standard deviation from three devices at each Bi₂Se₃ thickness.

Supplementary Figure 12. MOKE images of SOT driven magnetization switching in Bi2Se3/Py at zero external magnetic field and room temperature. a-e, MOKE images for SOT driven magnetization switching using a pulsed current I along the $+x$ -axis with increasing the current density J_C denoted underneath the corresponding image. $f - j$, MOKE images for SOT driven magnetization switching by *I* along the –*x*-axis. The dark (light) contrast shows the magnetization along the $+y$ ($-y$)-axis, which is also indicated by the white arrows in (a) , (e) , (f) and (j) . The white scale bar is 20 μ m.

Supplementary Figure 13. Current polarity dependent magnetization switching in Bi2Se3/Py at zero external magnetic field and room temperature. a, Initialized state with the Py magnetization along +*y*-axis. **b-c**, Current induced magnetization switching from +*y* to –*y*-axis is only induced by a pulsed current along the +*x*-axis in the absence of an external magnetic field. **d**, Initialized state with magnetization along the –*y*-axis. **e-f**, Current induced magnetization switching from $-y$ to $+y$ -axis is only induced by a pulsed current along the – *x*-axis. The current channel of $Bi₂Se₃/Py$ is 12 μ m wide. The white arrows represent the Py magnetization direction in each case. The white scale bar is $20 \mu m$.

Supplementary Figure 14. MOKE images of SOT driven magnetization switching in Bi2Se3/Cu (1 nm)/Py at zero external magnetic field and room temperature. a-d, MOKE images for SOT driven magnetization switching using a pulsed current *I* along the +*x*-axis with increasing the current density J_C denoted underneath the corresponding image. **e-h**, MOKE images for SOT driven magnetization switching by I along the $-x$ -axis. The dark (light) contrast shows the magnetization along the $+y$ ($-y$)-axis, which is also indicated by the white arrows in (a) , (d) , (e) and (h) . The white scale bar is 20 μ m.

Supplementary Figure 15. MOKE images of SOT driven magnetization switching in Bi2Se3/Cu (2 nm)/Py at zero external magnetic field and room temperature. a-d, MOKE images for SOT driven magnetization switching using a pulsed current *I* along the +*x*-axis with increasing the current density J_C denoted underneath the corresponding image. **e-h**, MOKE images for SOT driven magnetization switching by *I* along the $-x$ -axis. The dark (light) contrast shows the magnetization along the $+y$ ($-y$)-axis, which is also indicated by the white arrows in (a) , (d) , (e) and (h) . The white scale bar is 20 μ m.

Supplementary Figure 16. MOKE images of SOT driven magnetization switching in Bi2Se3/NiO (1 nm)/Py at zero external magnetic field and room temperature. a-d, MOKE images for SOT driven magnetization switching using a pulsed current *I* along the +*x*-axis with increasing the current density J_C denoted underneath the corresponding image. **e-h**, MOKE images for SOT driven magnetization switching by *I* along the $-x$ -axis. The dark (light) contrast shows the magnetization along the $+y$ ($-y$)-axis, which is also indicated by the white arrows in (a) , (d) , (e) and (h) . The white scale bar is 20 μ m.

Supplementary Figure 17. MOKE images for Bi2Se3/NiO (2 nm)/Py at room temperature. a-g, MOKE images with increasing *J*C, showing no current induced magnetization switching with the pulsed current I along $+x$ -axis. The Py magnetization remains along $+y$ -axis. The current channel is 10 μ m wide and the pulsed current width is 500 μ s. **h**, A reference MOKE image of *H* driven magnetization switching from +*y* to -*y*-axis, which is indicated by the change in contrast of the device in (**h**). The white scale bar is 20 μ m.

Supplementary Note 1: HAADF-STEM image of Bi2Se3/NiFe heterostructures

The cross-section of the Bi2Se3/NiFe (Py) heterostructure is characterized with high-angle annular dark-field (HAADF) imaging in an aberration corrected scanning transmission electron microscope (STEM) as shown in Supplementary Fig. 1. We find that the $Bi₂Se₃$ has uniform and good layered structure, and one quintuple layer (QL) is around 1-nm thick. Moreover, the image shows a clean and smooth interface between the Bi_2Se_3 and Py layer.

Supplementary Note 2: Bi2Se³ thickness dependent sheet resistance, *R***^s**

Supplementary Fig. 2 shows the sheet resistance, R_s , for different Bi_2Se_3 thicknesses. We find that R_s is small and remains almost constant at large thicknesses (15 and 20 QL). While R_s shows an abrupt increase as $Bi₂Se₃$ is thinner than 10 QL and it becomes maximum at 5 QL. This trend is similar as other reports 1,2 1,2 1,2 1,2 .

Supplementary Note 3: Evaluation of conventional SOT efficiency (θ_{II}) in Bi₂Se₃ by **ST-FMR**

As shown in Fig. 2c, we obtain the amplitudes of symmetric (V_S) and antisymmetric (V_A) components from the fits of the typical spin torque ferromagnetic resonance (ST-FMR) signal. Subsequently, the spin-orbit torque (SOT) efficiency θ_{TI} can be evaluated from only *V*_S by

$$
V_{\rm S} = -\frac{I_{\rm RF} \gamma \cos \theta_{\rm H}}{4} \frac{dR}{d\theta_{\rm H}} \tau_{\rm DL} \frac{1}{\Delta} F_{\rm S}(H) , \sigma_{\rm S} = J_{\rm S}/E = \tau_{\rm DL} M_{\rm s} t/E \quad \text{and } \theta_{\rm TI} = J_{\rm S}/J_{\rm C} = \sigma_{\rm S}/\sigma^{\text{3,4}} \text{ where}
$$

*I*_{RF} is the RF current flowing through the device, γ is the gyromagnetic ratio, $dR/d\theta_{\rm H}$ is the angular dependent magnetoresistance at $\theta_H = 35^\circ$, Δ is the linewidth of ST-FMR signal, $F_S(H)$ is a symmetric Lorentzian, *H* is in-plane external magnetic field, τ_{DL} is the damping-like spin-orbit torque on unit Co₄₀Fe₄₀B₂₀ (CFB) moment at $\theta_H = 0^\circ$, M_s is the saturation magnetization of CFB, *t* is the thickness of CFB, *J*_S is the measured spin current density with in-plane spin polarizations at the Bi2Se3/CFB interface, which is correlated with the measured symmetric component V_s as shown in Fig. 2c, J_c (A cm⁻²) is the uniform charge current density in the Bi₂Se₃ layer, σ_s is the Bi₂Se₃ spin Hall conductivity, σ is the Bi₂Se₃ conductivity, and *E* is the microwave field across the device.

In the above *conventional* SOT efficiency evaluation method^{[3](#page-27-2)[,4](#page-27-3)}, the dimensionless θ_{II} arising from the Bi₂Se₃ layer are obtained by using the uniform charge current density J_C (unit in A cm^{-2}) in the entire Bi₂Se₃ layer (including carriers from topological surface states, two dimensional electron gas, and bulk states). It can represent the overall charge-to-spin conversion efficiency in the TI/FM device. On the other hand, it is of interest to reveal the *interface* SOT efficiency, λ_{TSS} (nm⁻¹), from only topological surface states (TSS) for which we need to use an interface charge current density $J_{\text{C-TSS}}$ (unit in A cm⁻¹) only in TSS (i.e. two dimensional carriers from TSS). λ _{TSS} from TSS is evaluated in Supplementary Note 6 and Supplementary Note 7.

Supplementary Note 4: Estimation of n_{TSS} **,** n_{2DEG} **,** $n_{\text{2D-Bulk}}$ **,** E_F **and** k_F **at different** t_{BISE}

On the basis of the t_{BiSe} dependent sheet carrier concentration, n_{2D} , in Fig. 1b, we estimate the contributions of three possible conduction channels to the electrical transport, such as surface states (including TSS and two dimensional electron gas, 2DEG) and bulk states (BS). As mentioned in the main text, the thickness of a TSS, t_{TSS} , and 2DEG, t_{2DEG} , in $Bi₂Se₃$ is reported to be \sim 1 nm and 4 nm, respectively. Therefore, no BS are expected if the Bi₂Se₃ thickness is less than ~8 QL (i.e. \approx 8 nm). Consequently, n_{2D} can be writen as n_{2D} = $2 \times n_{TSS} + n_{2DEG}$ for $t_{BISE} \le 8$ QL and $n_{2D} = 2 \times n_{TSS} + n_{2DEG} + n_{2D-Bulk}$ for $t_{BISE} > 8$ QL, where *n*TSS, *n*_{2DEG} and *n*_{2D-Bulk} are the sheet carrier concentration in a TSS layer, total 2DEG and BS, respectively, and the factor of 2 arises due to the bottom and top surfaces in Bi2Se3.

As shown in Supplementary Fig. 3, the TSS shows a linear band dispersion given by $E_F = \hbar V_F k_F + E_{DP}$, where E_F is the Fermi energy, V_F is Fermi velocity ~[5](#page-27-4)×10⁵ m s⁻¹ in TSS,⁵ *h* is reduced Plank constant, k_F is Fermi wave vector of TSS and E_{DP} is the energy of Dirac point (DP). Therefore, the density of states of TSS per surface area is linearly proportional to energy

$$
D(E) = \frac{1}{2\pi} \frac{1}{(\hbar V_{\rm F})^2} (E - E_{\rm DP}).
$$
 (1)

Then we can estimate n_{TSS} by

$$
n_{\rm TSS} = \int_{E_{\rm DP}}^{\infty} D(E)f(E)d(E),\tag{2}
$$

where F $(E) = 1/(1 + e^{\overline{K_{\rm B}T}})$ *E E* $f(E) = 1/(1 + e^{\frac{E-E}{K_B T}})$ $+e^{K_B T}$) is the Fermi-Dirac distribution function, K_B is Boltzmann constant and *T* is 300 K. Consequently, by substituting Supplementary Eq. 1 and *f*(E) into Supplementary Eq. 2, n_{TSS} can be rewritten as

$$
n_{\rm TSS} = \int_{E_{\rm DP}}^{\infty} \frac{1}{2\pi} \left(\frac{K_{\rm B}T}{\hbar V_{\rm F}}\right)^2 \frac{\varepsilon}{1 + e^{\varepsilon - \eta}} d\varepsilon = \frac{1}{2\pi} \left(\frac{K_{\rm B}T}{\hbar V_{\rm F}}\right)^2 F_{\rm I}(\eta),\tag{3}
$$

where $\varepsilon = (E - E_{\text{DP}})/K_B T$, $\eta = (E_{\text{F}} - E_{\text{DP}})/K_B T$ and $F_1(\eta)$ is the Fermi-Dirac integral of order 1. Under the condition of $E_F - E_{DP} \gg K_B T$, Supplementary Eq. 3 is simplified as

$$
n_{\rm TSS} = \frac{1}{4\pi} \left(\frac{K_{\rm B}T}{\hbar V_{\rm F}}\right)^2 \eta^2.
$$
 (4)

From Supplementary Eq. 4, we find that n_{TSS} is proportional to $(E_F - E_{DP})^2$. We can also estimate k_F by $k_F = (E_F - E_{DP})/\hbar V_F$.

The 2DEG is usually formed at the Bi₂Se₃ surface due to band bending. As shown in Supplementary Fig. 3, after band bending, E_{CBM} (*E*_{VBM}) evolves into a surface conduction band minimum, E_{SCBM} (surface valence band maximum, E_{SVBM}) at surface and E_{DP} shifts downwards to a higher binding energy. The quantum confinement along the film normal leads to the quantized subbands in 2DEG enclosed by TSS linear bands as shown in Supplementary Fig. 3. As the Bi₂Se₃ film is in the thick range ($t_{\text{Bise}} > 8$ QL), the TSS and the 2DEG on top and bottom surfaces are separated. The 2DEG subbands on each surface are respectively confined in a triangular quantum well caused by surface band bending^{[1,](#page-27-0)[6](#page-27-5)}. As the Bi₂Se₃ film reduces below 8 QL, the 2DEG states from both surfaces merge, therefore, the square quantum confinement effects between the substrate and capping layer take over the triangular quantum well. It should be noted that, for clarity, the Rashba splitting is not illustrated in 2DEG subbands.

For $t_{\text{BiSe}} \leq 8$ QL, a simple infinite square quantum well is assumed^{[1](#page-27-0)}. The energy minimum of each subband at $k_{\parallel} = 0$ (E_{Cn} , $n = 1, 2...$) gradually moves up with the energy E_{n} $(n = 1, 2...)$ away from E_{SCBM} , where $E_n = \frac{\hbar^2 \pi^2}{2.2 \times 10^{-4} \text{ m}^2 \text{ m}^2} n^2$ n^* 2 m^* t_{BiSe}² $E_{n} = \frac{1}{\sqrt{n}}$ *m t* $=\frac{n \pi}{2m^*}$ $\frac{n^2}{2}$, where m^* is the effective mass of electron. m^* is ~0.14 m_0 for Bi_2Se_3 ,^{[7-9](#page-27-6)} where m_0 is the free-electron mass. The thinner the Bi₂Se₃ film, the more separation of 2DEG subbands, which indicates a decrease of the contribution of 2DEG subbands to the electrical transport as $Bi₂Se₃$ becomes thinner. For $t_{\text{Bise}} > 8$ QL, a simple triangular quantum well is used to describe the confined 2DEG subbands resulting from a band bending at each film surface^{[6](#page-27-5)}. The energy minimum of the allowed 2DEG subbands can be written as $_{\rm 2/3r} \, \hbar^2 (\nabla V)^2 \,$ $_{\rm 1l/3}$ $C_{\rm cn} = E_{\rm n} + E_{\rm SCBM} = \left[\frac{3\pi}{2}(n-\frac{1}{4})\right]^{2/3} \left[\frac{\hbar^2(\nabla V)^2}{2m^*}\right]^{1/3} + E_{\rm SCBM}$ $E_{-} = E_{+} + E_{\text{ccav}} = \left[\frac{3\pi}{2}(n-1)\right]^{2/3} \left[\frac{\hbar^2(\nabla V)^2}{2} \right]^{1/3} + E_{-}$ *m* $\delta \pi = 1$ $\frac{1}{2}$ δ^2 $(\nabla$ $E_n + E_{\text{SCBM}} = \left[\frac{3\pi}{2}(n-\frac{1}{4})\right]^{2/3}\left[\frac{n(n+1)}{2m^*}\right]^{1/3} + E_{\text{SCBM}}$, where the integer *n* represents the

quantum number of allowed 2DEG states and ∇V (meV \AA^{-1}) is the potential gradient of triangular well near the $Bi₂Se₃$ surface, which can be estimated by the ratio of band bending energy (E_{CBM} - E_{SCBM}) and corresponding band bending depth along the film normal (\sim 4 nm).

For each 2DEG subband, n_{2DEG} can be calculated by using the basic 2DEG equation $n_{\text{2DEGn}} = \frac{m}{\pi \hbar^2} K_{\text{B}} T \ln(1 + e^{(E_{\text{F}} - E_{\text{Cn}})/K_{\text{B}}T})$ $=$ $\frac{K_{\rm B} I \ln(1+e^{iE_{\rm F}})}{E_{\rm E}}$ $\int_{\overline{R}}^{\infty} K_{\text{B}} T \ln(1 + e^{(E_{\text{F}} - E_{\text{Cn}})/K_{\text{B}}T})$, where $n = 1$ and 2. We only consider the first and second subbands in our model since the third subband has a negligible contribution to n_{2DEG} . Note that for $t_{\text{BiSe}} \leq 8$ QL, the sheet carrier concentration n_{2DEG} in this model is from one 2DEG layer. However for $t_{\text{Bise}} > 8$ QL, the sheet carrier concentration n_{2DEG} needs to consider two 2DEG layers.

The $n_{2D-Bulk}$ is estimated as follows. As it is known that the Bi_2Se_3 bulk can be regarded as a semiconductor with a band gap $E_g \sim 0.3$ eV as shown in Supplementary Fig. 3. The bulk carrier concentration is calculated by using the basic semiconductor equation^{[10](#page-28-0)} ($E_F - E_{\text{CBM}}$) $3K_B$ T, E_{CBM} is the bulk conductance band minimum)

$$
n_{\text{Bulk}} = \frac{(2m^*)^{3/2}}{2\pi^2\hbar^3} \int_{E_c}^{\infty} \frac{(E - E_{\text{CBM}})^{1/2}}{1 + e^{[(E - E_{\text{F}})/K_{\text{B}}T]}} dE = N_{\text{C}} \frac{2}{\sqrt{\pi}} F_{1/2}(\xi) ,
$$
 (5)

where $N_c = 2(m^* K_B T / 2\pi \hbar^2)^{3/2}$. $F_{1/2}(\xi)$ is the Fermi-Dirac integral of order 1/2, where $\xi = (E_F - E_{CBM})/K_B T$. Then $n_{2D-Bulk}$ can be converted by $n_{2D-Bulk} = n_{Bulk} d$, where *d* is the bulk thickness excluding the TSS and 2DEG thicknesses. From Supplementary Eq. 5, we know that $n_{2D-Bulk}$ is related to $(E_F - E_{CBM})$.

In order to determine n_{TSS} , n_{2D -bulk and n_{2DEG} , we need to know the values of $E_F - E_{DP}$, E_F $-E_{\text{CBM}}$ and $E_F - E_{\text{Cn}}$, accordingly. On the basis of Supplementary Fig. 3, we simply rewrite $E_F - E_{Cn}$ as $E_F - E_{Cn} = (E_F - E_{SCBM}) - E_n = (E_F - E_{DP}) - (E_{SCBM} - E_{DP}) - E_n$. According to the reported ARPES data^{[5,](#page-27-4)[11-13](#page-28-1)} and theoretical band structure calculation^{[14](#page-28-2)}, we adopt $E_F - E_{\text{CBM}}$ \sim 100 meV and E_{SCBM} – $E_{\text{DP}} \sim$ 200 meV. Consequently, E_{F} – E_{DP} , which is determined by the magnitude of band bending induced by electron doping at surface, is the only variable to extract n_{2D} at different t_{Bise} . 2× n_{TSS} , n_{2DEG} , $n_{2D-Bulk}$, $E_F - E_{DP}$ and k_F of TSS as a function of *t*BiSe are quantitatively determined and plotted in Fig. 4.

We have used $V_F = 5 \times 10^5$ m s⁻¹, $m^* = 0.14$ *m*₀, $E_{SCBM} - E_{DP} = 200$ meV and $E_F - E_{CBM} =$ 100 meV for the estimation in Fig. 4 in the main text. Additionally, we have estimated n_{2D} by varying the parameters with the range from literatures, including V_F , m^* , $E_{\text{SCBM}} - E_{\text{DP}}$ and E_F $- E_{\text{CBM}}$, respectively. First, we vary the value of V_{F} from 4.6×10^5 to 5.4×10^5 m s⁻¹,^{[5,](#page-27-4)[15-17](#page-28-3)} and fix $m^* = 0.14$ m_0 , $E_{\text{SCBM}} - E_{\text{DP}} = 200$ meV and $E_F - E_{\text{CBM}} = 100$ meV. The estimated results are shown in Supplementary Fig. 4. We find that as the V_F increases, the n_{TSS} only slightly decreases. Moreover, the n_{2D} keeps the similar trend as that in the Fig. 4a (also see

Supplementary Fig. 4b). Overall, the value of V_F does not affect the estimation results significantly.

Similarly, we varied the value of m^* in the range of $0.11m_0$ to $0.17m_0$,^{[6-9](#page-27-5)[,12](#page-28-4)[,13](#page-28-5)[,18](#page-28-6)} and fixed $V_F = 5 \times 10^5$ m s⁻¹, $E_{SCBM} - E_{DP} = 200$ meV and $E_F - E_{CBM} = 100$ meV as used in the main text. As shown in Supplementary Fig. 5, we find that as m^* increases, the n_{2DEG} and $n_{2D-Bulk}$ only slightly increase. Moreover, the results in Supplementary Fig. 5 are similar to the values shown in the main text.

In addition, we also varied the value of $E_{\text{SCBM}} - E_{\text{DP}}$ from 150 to 250 meV,^{[17](#page-28-7)[,19-21](#page-28-8)} and the value of $E_F - E_{\text{CBM}}$ from 50 to 150 meV.^{[5,](#page-27-4)[11,](#page-28-1)[12](#page-28-4)[,17,](#page-28-7)[19](#page-28-8)} The results are shown in Supplementary Fig. 6 and 7, respectively. We find that different parameters can only weakly affect the values of n_{TSS} , n_{2DEG} and $n_{2D-Bulk}$, and the main features of the results in the main text still hold good.

Supplementary Note 5: Current shunting (I_{TSS}/I_{total}) at different *t*BiSe

The current flowing in each channel (TSS, 2DEG, and BS) can be written as $I_{\text{TSS}} = n_{\text{TSS}} \mu_{\text{TSS}} eEW$, $I_{\text{2DEG}} = n_{\text{2DEG}} \mu_{\text{2DEG}} eEW$ and $I_{\text{Bulk}} = n_{\text{2D-Bulk}} \mu_{\text{Bulk}} eEW$ respectively, where *W* and *E* are the channel width and electric field (same for TSS, 2DEG and BS channels) in each device, and μ_{TSS} , μ_{2DEG} and μ_{Bulk} are the carrier mobility in TSS, 2DEG and BS, respectively. The linear Hall curves we measured at different *t*_{BiSe} indicate the carriers in Bi₂Se₃ have a similar mobility^{[1](#page-27-0)}. Thus, we obtain I_{TSS}/I_{total} at different t_{BiSe} by assuming μ_{TSS} = $\mu_{2DEG} = \mu_{Bulk}$ (Fig. 4c in the main text), where I_{total} is the total current flowing in a Bi₂Se₃ film.

Supplementary Note 6: Evaluation of the interface SOT efficiency, λ **rss, from TSS**

As discussed in Supplementary Note 3, the θ_{TI} versus t_{BiSe} from ST-FMR measurements is obtained by using a uniform charge current density $J_C(A \text{ cm}^{-2})$ in the entire Bi₂Se₃ layer as $\frac{S}{I_{\text{CI}}} = \frac{S_{\text{S}}}{I_{\text{CI}}} = \frac{S_{\text{S}}}{n_{\text{2D}}\mu eE/t_{\text{Bise}}}$ J_{\circ} *J* J_c $n_{\rm m}$ μ eE/t θ . μ $=\frac{J_S}{I}=\frac{J_S}{I}=\frac{J_S}{I}$. The *interface* SOT efficiency from only TSS, λ_{TSS} (nm⁻¹), can be obtained by the *interface* charge current density $J_{\text{C-TSS}}$ (A cm⁻¹) in TSS as $\frac{1}{TSS} = \frac{1}{T} \frac{S}{S} = \frac{1}{T} \frac{S}{S}$ C-TSS 'TSS J_{\circ} *J* J_{arc} n_{res} *µ* λ , μ $\lambda_{\rm TSS} = \frac{J_{\rm S}}{I_{\rm TSS}} = \frac{J_{\rm S}}{I_{\rm TSS}}$. Therefore, we can evaluate $\lambda_{\rm TSS}$ by $\lambda_{\rm TSS} = \frac{n_{\rm 2D}/n_{\rm BISE}}{2} \theta_{\rm TBS}$ TSS $n_{\rm 2D}/t$ $\lambda_{\text{TSS}} = \frac{n_{\text{2D}}/n_{\text{BiSe}}}{n_{\text{TSS}}} \theta_{\text{TI}}$. The

estimated λ _{TSS} is plotted in Fig. 4d.

Supplementary Note 7: Estimation of λ_{2DEG} **in 2DEG and** λ_{inTSS}

According to above analysis, for $t_{\text{BiSe}} \leq 8$ QL, there are only TSS and 2DEG contribution to the SOT efficiency. As mentioned in the main text, the spin current density at the interface arising from only TSS (J_{S-TSS}) is not necessarily equal to the spin current density (J_S) from ST-FMR measurements due to the partial cancellation by the opposite spin polarizations from Rashba splitting in 2DEG. Based on this scenario, we rewrite the λ TSS in Fig. 4d as

$$
\lambda_{\rm TSS} = \frac{J_{\rm S-TSS} - J_{\rm S-2DEG}}{J_{\rm C-TSS}} = \frac{J_{\rm S-TSS}}{J_{\rm C-TSS}} - \frac{J_{\rm S-2DEG}}{J_{\rm C-TSS}}
$$
, where $J_{\rm S-TSS}/J_{\rm C-TSS}$ is the intrinsic interface SOT

efficency from TSS $(\lambda_{\text{intrTSS}})$ which is inversely propotional to V_F and almost remain constant at different $t_{\text{Bis}e}^{22}$ $t_{\text{Bis}e}^{22}$ $t_{\text{Bis}e}^{22}$, and $J_{S\text{-2DEG}}$ is the spin current density from Rashba splitting in 2DEG. Then we get $\lambda_{\text{rss}} = \lambda_{\text{intrTSS}} - \frac{\lambda_{\text{2DEG}}}{I}$ C-TSS *J J* $\lambda_{\text{rss}} = \lambda_{\text{intrijTSS}} - \frac{\lambda_{\text{2DEG}} J_{\text{C-2DEG}}}{I}$, where λ_{2DEG} is the interface SOT efficiency from

Rashba splitting in 2DEG, $J_{C_2DEG} = n_{2DEG} \mu eE$ and $J_{C_TSS} = n_{TSS} \mu eE$. We assume that the difference of surface band bending between 7 and 8-QL $Bi₂Se₃$ films is small, which results in an almost constant λ_{2DEG} . By using the difference of λ_{TSS} between 7 and 8 QL film as shown in Fig. 4d, the λ_{2DEG} is determined and it shows negative value and is ~ -0.4 nm⁻¹. Moreover, the values for $\lambda_{\text{intrITSS}}$ are also estimated for $t_{\text{Bise}} \leq 10$ QL with negligible BS. Interestingly, we find $\lambda_{\text{intriTSS}}$ shows a constant value of ~0.8 nm⁻¹ for 7, 8 and 10 QL Bi₂Se₃ films. This amended interface SOT efficiency is in the similar range of the value of λ_{TSS} $(\sim 0.82 \text{ nm}^{-1})$ at $t_{\text{BiSe}} = 5$ QL as shown in Fig. 4d. This further substantiates our claim of TSS dominated SOT in thinner films and the high SOT efficiency from TSS.

We have used $V_F = 5 \times 10^5$ m s⁻¹, $m^* = 0.14$ m_0 , $E_{SCBM} - E_{DP} = 200$ meV and $E_F - E_{CBM} =$ 100 meV for the estimation in Fig. 4 in the main text. Additionally, similar to Supplementary Note 4, we have also estimated the interface SOT efficiency, λ_{TSS} and amended λ_{TSS} by varying the parameters V_F , m^* , $E_{SCBM} - E_{DP}$ and $E_F - E_{CBM}$ with the range of values from literatures. First, we varied the value of V_F from 4.6×10^5 to 5.4×10^5 m s⁻¹,^{[5,](#page-27-4)[15-17](#page-28-3)} and fixed m^{*} $= 0.14$ *m*₀, $E_{\text{SCBM}} - E_{\text{DP}} = 200$ meV and $E_{\text{F}} - E_{\text{CBM}} = 100$ meV. The estimated results are shown in Supplementary Fig. 8. We find that as the V_F varies, the interface SOT efficiencies do not change much and show similar values with respect to that in the Fig. 4d (also see Supplementary Fig. 8b). Overall, the *V*_F does not affect the estimation results significantly.

Similarly, we varied the value of m^* in the range of $0.11m_0$ to $0.17m_0$,^{[6-9](#page-27-5)[,12](#page-28-4)[,13](#page-28-5)[,18](#page-28-6)} and fixed $V_F = 5 \times 10^5$ m s⁻¹, $E_{SCBM} - E_{DP} = 200$ meV and $E_F - E_{CBM} = 100$ meV as used in the main text.

As shown in Supplementary Fig. 9, we find that as m^* increases, the interface SOT efficiency only slightly increases and the results in Supplementary Fig. 9 are similar to the values shown in the main text.

In addition, we also varied the value of $E_{\text{SCBM}} - E_{\text{DP}}$ from 150 to 250 meV,^{[17](#page-28-7)[,19-21](#page-28-8)} and the value of $E_F - E_{\text{CBM}}$ from 50 to 150 meV.^{[5,](#page-27-4)[11,](#page-28-1)[12](#page-28-4)[,17,](#page-28-7)[19](#page-28-8)} The results are shown in Supplementary Fig. 10 and 11, respectively. We find different parameters can only weakly affect the values of the interface SOT efficiency and the main features of the results in the main text still hold good.

Supplementary Note 8: Reproducible SOT driven magnetization switching in Bi2Se3/NiFe device

The SOT driven magnetization switching by currents is reproducible in other $Bi₂Se₃$ (8) QL)/Py (6 nm) devices. One example is shown in Supplementary Fig. 12. We use the same measurement condition used in Fig. 5 and the current channel is $12 \mu m$ wide. We find that the switching current density is $\sim 6 \times 10^5$ A cm⁻², which is similar as that in the device in Fig. 5.

Supplementary Note 9: SOT efficiency from current induced magnetization switching

For the conventional antidamping spin torque driven magnetization switching, the critical switching current density $J_{\text{C}0}$ for the switching scheme of our Bi₂Se₃/Py device can be described by $23,24$ $23,24$

$$
J_{\rm{CO}} = \frac{2e}{\hbar} \mu_0 M_s t \alpha (H_{\rm{c}} + M_{\rm{eff}}/2) / \theta_{\rm{TI}} ,
$$
 (6)

where *J*C0 is the critical switching current density without thermal fluctuation, *M*s*, t, α, H*^c and *M*eff are the saturated magnetization, thickness, damping constant, coercive field and effective magnetization of Py layer, respectively, and θ_{TI} is the SOT efficiency. The Supplementary Eq. 6 is based on the macrospin model in the absence of thermal fluctuation. In our device, the magnetization switching process can be described by the localized nucleation of reverse domains with an activation volume V_N first, followed by domain wall propagation. We anticipate that the magnetization exhibits coherent reversal inside the activation volume V_N . Therefore, the Supplementary Eq. 6 can apply in our device by introduction of V_N instead of the whole volume of Py layer. In our measurements, the switching current density J_c for the magnetization switching is $\sim 6.2 \times 10^5$ A cm⁻² at room temperature. Then the *J*_{C0} can be obtained by $\frac{C_{\text{C}}}{I} = 1 - \frac{H_{\text{B}}}{K}$ CO \longrightarrow $Py'N$ Y_0 $J_c = 1 - \frac{K_B T}{\hbar^2} \ln \frac{t_p}{\hbar^2}$ $J_{C_0} = 1 - \frac{K_B T}{K_{D} V_N} \ln \frac{t_P}{t_0}$ with thermal fluctuation consideration^{[25-27](#page-28-12)}, where *t*_P is the

current pulse width \sim 500 µs, t_0 is the attempt time \sim 1 ns, the anisotropy energy density K_{Py} is estimated by $H_cM_s/2$ with measured $H_c \sim 6.9$ Oe and $M_s = 6.84 \pm 0.03 \times 10^5$ A m⁻¹. The domain wall width $\delta_{\rm m}$ of Py layer is assumed to be ~220 nm,^{[28,](#page-28-13)[29](#page-28-14)} and *t* is 6 nm, then we can estimate $V_{\text{N}} \approx \delta_{\text{m}}^2 t$. Consequently, we find *J*_{C0} ~5.26×*J*_C. The *M*_{eff} and *α* are ~0.57 T and ~0.01543, respectively, which are obtained from ST-FMR and vibrating sample magnetometer (VSM) measurements. From Supplementary Eq. 6, we determine the SOT efficiency θ_{II} for Bi₂Se₃/Py to be ~1.71. This value is consistent with the value obtained from ST-FMR measurements ($\theta_{\text{TI}} \sim 1$).

The charge to spin conversion efficiency is typically less than one from the definition of $\theta_{\text{TI}} = J_{\text{S}}/J_{\text{C}}$. However, recently, there have been several reports to evaluate θ_{TI} in the topological insulators (TIs) and they have observed θ_{TI} greater than one. Specifically, the θ_{TI} values are reported to be \sim 2–3.5 (in Bi₂Se₃/P_y by ST-FMR measurements at room temperature)³[,](#page-27-2) ~140–425 (in Cr doped BiSbTe/(Bi_{0.5}Sb_{0.5})₂Te₃ bilayer by magnetization switching at 1.9 K)^{[30](#page-28-15)} and ~20 (in $(Bi_{0.5}Sb_{0.5})₂Te₃$ by spin tunneling spectroscopy method below 200 K)^{[31](#page-28-16)}. Therefore, we speculate that the spin-momentum locking at the TSS in TIs plays an important role. Additionally, the recent reported multi-cycle spin transfer scenario^{[32](#page-29-0)} could be also responsible for a large θ_{TI} , which is explained as follows. As a charge current flows within $Bi₂Se₃$ TI film, spin accumulation is generated and the spins flow vertically toward the ferromagnetic Py layer. These spins are absorbed and exert SOTs on the Py magnetization. The θ_{TI} is evaluated by the measured torque in the ST-FMR measurements. Microscopically, in this process, electrons become spin polarized at the TSS due to spin-momentum locking and transfer the spin angular momentum to the Py magnetization at the interface of $Bi₂Se₃/Py$. However, these electrons can again diffuse back into the $Bi₂Se₃$ layer due to no net charge current flowing vertically into the Py layer. This above process repeats multiple times. Therefore, each electron can transfer the spin angular momentum multiple times, as it moves opposite to the charge current direction, and thus exert large torques on the Py layer. As reported recently^{[15,](#page-28-3)[33-36](#page-29-1)}, the electrons can be spin polarized in Bi₂Se₃ TSS with very large spin polarization values ranging from \sim 0.2 to 0.75 due to the spin momentum locking. Therefore, we think the large spin polarization in TSS and the multi-cycle spin transfer scenario are the possible reasons for the θ_{TI} greater than one observed in TIs.

Supplementary Note 10: Current polarity dependent magnetization switching

Supplementary Fig. 13 shows that the current induced magnetization switching depends on the charge current polarity which is a characteristic of SOT driven magnetization switching. For these measurements, we use the same device as in Fig. 5. In Supplementary Fig. 13, the Py magnetization direction in each case is indicated by a white arrow. For the set of measurements in Supplementary Fig. 13a-c, we first saturate the Py magnetization along the +*y*-axis by applying an in-plane external magnetic field (*H*). Then we remove *H* and capture the corresponding MOKE image as the initialized state as shown in Supplementary Fig. 13a. The dark contrast represents the magnetization along the +*y*-axis, which is indicated by the white arrow. Subsequently, we apply a pulsed current I along the $-x$ -axis with current density $J_C = 6.2 \times 10⁵$ A cm⁻² without an external magnetic field (see Supplementary Fig. 13b). We observe that there is no magnetization switching as the magnetization remains along its initial +*y*-axis indicated by the no contrast change. However, as we change the current polarity by applying *I* along the $+x$ -axis with the same value of J_c , we observe that the Py magnetization switches from the +*y* to –*y*-axis indicated by the light contrast (see Supplementary Fig. 13c).

Similarly, for the other set of measurements in Supplementary Fig. 13d-f, we first initialize the Py magnetization along the $-y$ -axis. Then we remove *H* and capture the corresponding MOKE image (see Supplementary Fig. 13d). Subsequently, we apply the current pulses of different polarities with $J_C = 6.2 \times 10^5$ A cm⁻². We find that only the current pulse along the –*x*-axis can realize magnetization switching from the –*y* to +*y*-axis indicated by the dark contrast (see Supplementary Fig. 13f). The dependence of magnetization switching direction on the polarity of current suggests that the switching in our case is primarily driven by the SOT and not by any thermal effects.

Supplementary Note 11: SOT induced magnetization switching with Cu and NiO insertions

In order to further verify the SOT induced magnetization switching in $Bi₂Se₃/Py$, we have performed the MOKE imaging measurements with the same condition used in Fig. 5 on the control devices of $Bi₂Se₃$ (8 QL)/insertion layer/Py (6 nm)/MgO (1 nm)/SiO₂ (4 nm), where the insertion layer is Cu (1 and 2 nm) or insulating layer NiO (1 nm).

It is well known that Cu has a very low spin-orbit coupling strength and a long spin diffusion length. Hence, Cu does not possibly affect the spin generation in the $Bi₂Se₃$ layer and can be a good spin conductor. We follow the recent report^{[22](#page-28-9)} and use Cu insertion to separate the $Bi₂Se₃$ and Py layer. The Cu is sputtered on top of $Bi₂Se₃$ with a low power of 30 W. As shown in Supplementary Fig. 14, the SOT driven magnetization switching is observed in Bi₂Se₃ (8 QL)/Cu (1 nm)/Py (6 nm) devices. We find that the switching current density J_C is ~6.7×10⁵ A cm⁻², which is similar as that in the device without Cu insertion (~6.2×10⁵ A cm^{-2}) in Fig. 5 and Supplementary Fig. 12. Similarly, Supplementary Fig. 15 reveals that the SOT driven magnetization switching by currents can also be observed in $Bi₂Se₃$ (8 QL)/Cu (2 nm)/Py (6 nm) devices. The switching current density slightly increases and is \sim 7.7 \times 10⁵ A cm^{-2} , which might be due to a bit of spin scattering in the Cu insertion layer. The devices with 1–2 nm Cu insertion exhibit almost similar device resistance as that with no Cu insertion from an independent four probe measurements. This indicates that the very thin Cu insertion has a much higher resistivity than the Py layer which is expected and is a usual case for very thin films. Therefore, for simplicity, we do not consider the current shunting in the Cu insertion and take the upper bound of the J_C denoted in the Supplementary Fig. 14 and 15.

In addition, we also perform the MOKE imaging measurements on the devices with insulating NiO insertion between the $Bi₂Se₃$ and Py layer. As shown in Supplementary Fig. 16, the SOT driven magnetization switching is observed in $Bi₂Se₃$ (8 QL)/NiO (1 nm)/Py (6 nm) devices. We find that the switching current density increases compared to that in Fig. 5. The magnetization starts to switch at $J_C \sim 5.4 - 6.7 \times 10^5$ A cm⁻² and fully switches at J_C $\sim 9.1 \times 10^5$ A cm⁻². This is expected since the spins are blocked by an insulator and the transmission will potentially decrease as the insulator thickness increases. As shown in Supplementary Fig. 17, we cannot observe SOT induced magnetization switching with J_C up to $\sim 8 \times 10^5$ A cm⁻² as the NiO insertion layer becomes 2 nm.

By inserting Cu and insulating NiO layers, we can (at least partially) prevent the direct interface between $Bi₂Se₃$ and Py. The SOT induced magnetization switching by $Bi₂Se₃$ is reproducible and robust in different devices. Therefore, we further confirm the highly efficient SOT induced magnetization switching in our Bi2Se3/Py heterostructures.

Supplementary Note 12: Influence of Oersted field on current induced magnetization switching

We perform the MOKE imaging measurements on a control device of $Bi₂Se₃ (8 QL)/NiO$ (2 nm) /Py (6 nm)/MgO (1 nm)/SiO₂ (4 nm) with the same condition used in Fig. 5. Due to the 2-nm NiO insulating insertion layer, the spin currents propagating from $Bi₂Se₃$ into Py is attenuated significantly, which is confirmed by the ST-FMR measurements. Therefore, the current induced Oersted field (H_{OE}) would be the main driving force on the Py layer. At the beginning of this set of measurements, we first saturate the Py magnetization along the +*y*-axis by applying an in-plane external magnetic field (*H*). Then we remove *H* and apply *I* along the $+x$ -axis to the device. When the current density in Bi₂Se₃ (*J*_C) is zero, we capture the MOKE image as shown in Supplementary Fig. 17a. The dark contrast represents the magnetization along the +*y*-axis, which is indicated by the white arrow. Then we gradually increase J_C at room temperature without *H*. As shown in Supplementary Fig. 17b-g, there is no current induced magnetization switching as the magnetization is still along the +*y*-axis indicated by the no contrast change. In these measurements, the applied J_C is up to 8×10^5 A cm⁻² (Supplementary Fig. 17g) which is almost 3 times larger than the ones at which the magnetization switching is triggered in the devices in Fig. 5 and Supplementary Fig. 12. Supplementary Fig. 17h shows a reference MOKE image of magnetization switching from +*y* to –*y*-axis driven by an applied external magnetic field *H*, which is indicated by the change in contrast to light colour. Moreover, the *H*_{Oe} from Bi₂Se₃ layer is estimated to be ~0.12–0.3 Oe by the equation^{[37](#page-29-2)} $H_{\text{Oe}} = J_c t_{\text{Bise}}/2$. We find that even this calculated value of H_{Oe} is much smaller than the required switching field of Py layer. Therefore, the H_{Oe} is not the mechanism for the current induced magnetization switching we observed in the Bi₂Se₃/Py devices.

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