Control of spin-orbit torques through crystal symmetry in WTe$_2$/ferromagnet bilayers

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Supplementary Note 1: Comparison of mechanisms for current-induced switching of magnetic layers with perpendicular magnetic anisotropy (PMA).

In principle, spin-orbit torques with three different symmetries can drive switching of PMA magnetic layers, each associated with different reversal mechanisms and yielding different values for the critical torque required for switching. [Note that in this discussion we will consider all torques per unit magnetization, so that $\bar{\tau}$ has the same units as $d\hat{m}/dt$, where $\hat{m}$ is the magnetic orientation.] (i) If the current can produce an effective field in the vertical (z) direction, yielding a torque of the form $\bar{\tau}_{\text{FL}} = -\gamma H_{\text{FL}} (\hat{m} \times \hat{z})$ where $\gamma$ is the gyromagnetic ratio, then in a macrospin approximation switching will occur at a critical value $H_{\text{FL}} = H_{\text{an}}$, where $H_{\text{an}}$ is the perpendicular anisotropy field. (ii) If the current produces an in-plane antidamping torque of the form $\bar{\tau}_{\text{AD,||}} = \tau_{\text{AD,||}}^0 \hat{m} \times (\hat{m} \times \hat{y})$, then deterministic switching can be achieved if there is also a symmetry-breaking effective field with a component along the current direction (x) \textsuperscript{51,52}, but the switching mechanism in this case is not actually based on a change in the magnetic layer’s effective damping because the antidamping torque is perpendicular to the magnetization. The torque in this case must still overcome the anisotropy field, so that the critical value of the torque in the macrospin limit is $\tau_{\text{AD,||}}^0 = \gamma H_{\text{an}} / 2$ (Refs. S2, S3). In samples larger than a few tens of nm diameter, an in-plane antidamping torque can, alternatively, drive a more efficient non-macrospin reversal process involving current-generated domain wall motion\textsuperscript{54}, but measurements indicate that this becomes ineffective for the highly-scaled PMA devices that are desired for applications\textsuperscript{55}. (iii) If the current produces an out-of-plane antidamping torque of the form $\bar{\tau}_{\text{AD,\perp}} = \tau_{\text{AD,\perp}}^0 \hat{m} \times (\hat{m} \times \hat{z})$, then in this case the direction of the torque is parallel to the magnetization so that it does have the ability to change the effective damping of the magnetic layer. Switching occurs when the effective damping is driven negative, resulting in a critical value of torque $\tau_{\text{AD,\perp}}^0 = \alpha G H_{\text{an}}$, where $\alpha G$ is the Gilbert damping parameter\textsuperscript{56,57}. Because the Gilbert damping is typically on the order of 0.01, an out-of-plane antidamping component has the ability to drive switching of PMA magnetic devices at much lower values of torque than the other two mechanisms, for sample sizes smaller than a few 10’s of nm.

Supplementary Note 2: Analysis of ST-FMR measurements.

We model the ST-FMR measurements by using the Landau-Lifshitz-Gilbert-Slonczewski (LLGS) equation to calculate the precessional dynamics of the magnetization direction, $\hat{m}(t)$, in the macrospin approximation in response to the in-plane and out-of-plane torque amplitudes, $\tau_{\text{||}}(\phi)$ and $\tau_{\text{\perp}}(\phi)$ as defined in the main text\textsuperscript{56,58}. This determines the ST-FMR mixing voltage as

$$V_{\text{mix}} = \left\langle I(t) R \left[ \hat{m}(t) \right] \right\rangle_t = V_s \frac{\Delta}{\left( B_{\text{app}} - B_0 \right)^2 + \Delta^2} + V_A \left( B_{\text{app}} - B_0 \right) \frac{\Delta}{\left( B_{\text{app}} - B_0 \right)^2 + \Delta^2} \quad (S1)$$

where $B_{\text{app}}$ is the applied magnetic field, $B_0$ is the applied magnetic field at ferromagnetic resonance, and $\Delta$ is the linewidth. The $\hat{m}(t)$ dependence of the device resistance, $R$, arises from the anisotropic magnetoresistance (AMR) of the ferromagnet Permalloy. We determine the symmetric and antisymmetric amplitudes, $V_s$ and $V_A$, by fitting Eq. S1 to measurements of the mixing voltage as a function of applied magnetic field. These amplitudes are related to the
torque amplitudes $\tau_\parallel$ and $\tau_\perp$ by Eqs. 1 and 2 in the main text. We note that $\tau_\parallel$ and $\tau_\perp$ are normalized by the total angular momentum of the magnet, and so have dimensions of frequency. We determine torque ratios from the ratio of Eqs. 1 and 2, together with measured values for $B_0$ and $M_{\text{eff}}$. We obtain the value of $B_0$ via fits of the resonance lineshape to Eq. S1, and we estimate $M_{\text{eff}}$ from the frequency dependence of $B_0$ using the Kittel formula

$$2\pi f = \gamma \sqrt{B_0 (B_0 + \mu_0 M_{\text{eff}})} .$$

As we discuss in Supplementary Note 2, $B_0$ and $M_{\text{eff}}$ depend on $\phi$ due to the in-plane magnetic anisotropy of our samples. For our analysis we use angle-averaged values for these quantities; the error in doing so is less than 5% due to the small degree of angular variation.

To obtain quantitative measurements of the individual torque components using Eq. 1 or Eq. 2 (i.e. not just their ratios), it is also necessary to determine $\alpha_G$, $R(\phi)$, and $I_{\text{RF}}$. The Gilbert damping $\alpha_G$ is estimated from the frequency dependence of the linewidth via

$$\Delta = 2\pi f \alpha_G / \gamma + \Delta_0 ,$$

where $\Delta_0$ is the inhomogeneous broadening. To obtain the AMR we measure the device resistance as a function of a rotating in-plane magnetic field (with magnitude 0.08 T) applied via a projected-field magnet. Fitting these data to $R_\alpha + \Delta R \cos^2 (\phi - \phi_0)$ allows calculation of $dR / d\phi$ (Fig. S1). To measure the RF current $I_{\text{RF}}$, we use a vector network analyzer to estimate the reflection coefficients of our devices ($S_{11}$) and the transmission coefficient of our RF circuit ($S_{21}$). These calibrations allow calculation of the RF current flowing in the device as a function of applied microwave power and frequency:

$$I_{\text{RF}} = 2 \sqrt{1 m W \cdot 10^{P_{\text{source}}(\text{dBm}) + S_{21}(\text{dBm})} (1 - |\Gamma|^2)^2} / 50 ,$$

where $P_{\text{source}}$ is the power sourced by the microwave generator and $\Gamma$ is given by

$$\Gamma = 10^{S_{11}(\text{dBm})/20} .$$

The torque conductivity, defined as the angular momentum absorbed by the magnet per second per unit interface area per unit electric field, provides an absolute measure of the torques produced in a spin source/ferromagnet bilayer independent of geometric factors. For a torque $\tau_K$ (where $K = \text{one of the A, B, S, or T indices for the torque components defined in the main text}$) we calculate the corresponding torque conductivity via

$$\sigma_K = M_S I_{\text{RF}} \tau_{\text{magnet}} / \gamma \frac{\tau_K}{(Iw)E} = M_S I_{\text{RF}} \tau_{\text{magnet}} / \gamma \frac{\tau_K}{I_{\text{RF}} \cdot 50(1 + \Gamma)} ,$$

where $M_S$ is the saturation magnetization, $E$ is the electric field, $l$ and $w$ are the length and width of the WTe$_2$/Permalloy bilayer, and $t_{\text{magnet}}$ is the thickness of the Permalloy. The factor $M_S I_{\text{RF}} \tau_{\text{magnet}} / \gamma$ is the total angular momentum of the magnet, which converts the normalized torque into units of angular momentum per second. Due to the unavailability of mm-scale WTe$_2$/Permalloy bilayers, we are unable to measure $M_S$ directly via magnetometry, and instead approximate $M_S \approx M_{\text{eff}}$, which we have found to be accurate in other Permalloy bilayer systems.
Supplementary Note 3: Determination of in-plane magnetic anisotropy.

Figure S2 shows the magnetic field at ferromagnetic resonance as a function of the in-plane magnetization angle for Devices 1 and 2. For Device 1 the current flows nearly parallel to the a-axis ($\phi_{a1} = -3^\circ$), and for Device 2 it is nearly parallel to the b-axis ($\phi_{a1} = 86^\circ$). The data from both samples indicate the presence of a uniaxial magnetic anisotropy within the sample plane, with an easy axis along the b-axis of the WTe$_2$. The angular dependence of the resonance field is described well by the form

$$B_0 = B_{\text{Kinet}} - B_A \cos \left( 2\phi - 2\phi_{\text{Easy-l}} - 2\phi_0 \right)$$

(S5)

where $B_A$ is the in-plane anisotropy field, related to the anisotropy energy $K_A$ via $B_A = 2\mu_0 K_A / M_s$, $B_{\text{Kinet}}$ is the resonance field without any in-plane anisotropy, $\phi_{\text{Easy-l}}$ is the angle from the current direction to the magnetic easy-axis and $\phi_0$ is the angular misalignment extracted from the angular dependence of the mixing voltage as discussed in the main text. This equation also assumes $B_A, B_{\text{Kinet}} \ll \mu_0 M_{\text{eff}}$ which are valid approximations for our experiment.

We find values for $B_A$ of 7 mT and 15 mT for Device 1 and Device 2, respectively. We observe no unidirectional component to the magnetic anisotropy.

We performed similar fits for all of the devices listed in Table S1 (Supplementary Note 4). In all cases the magnetic easy axis was along the b-axis within experimental uncertainty; i.e. $\phi_{a1} = \phi_{\text{Easy-l}} + 90^\circ$. Over all of our devices we find $B_A$ to be in the range 4.9-17.3 mT. Some, but likely not all, of the device-to-device variation may be explained by differences in the sample shape.

To check that the Permalloy has a magnetic anisotropy that is entirely in the sample plane we fabricated a WTe$_2$/Py bilayer Hall bar using the same sample fabrication techniques and Py thickness as our ST-FMR devices. The Hall bar is oriented with the current along the WTe$_2$ a-axis ($\phi_{a1} = -1^\circ$), with a length and width of 26 $\mu$m and 4 $\mu$m respectively. Hall measurements with the magnetic field applied perpendicular to the sample plane are shown in and in Fig. S3a and Hall measurements with the field parallel to the WTe$_2$ b-axis (the in-plane magnetic easy axis) are shown in Fig. S3b. In Fig. S3a, the contribution of the ordinary Hall effect has been removed by subtraction of the linear portion of the curve at large fields. Saturation of the Py moment is achieved in out-of-plane fields above 0.9 T and the extracted peak-to-peak value of the anomalous Hall contribution, $R_{\text{AHE}}$, is 0.62 $\Omega$. If there were any tilting of the anisotropy axis out-of-plane, this should give an antisymmetric signal in the b-axis scan about zero field. Instead, we observe only a very small, approximately-symmetric Hall signal in Fig. S3b ($\sim 1\%$ of the saturated anomalous Hall signal). The small signal that we see has an angular dependence (not shown) consistent with a planar Hall effect, and not an out-of-plane tilt. These results show that the overall magnetic anisotropy is in-plane, without any significant out-of-plane tilt of the equilibrium magnetization direction.

Supplementary Note 4: Data from additional devices.

In Table S1, we provide device parameters, torque ratios, and magnetic anisotropy parameters for 15 WTe$_2$/Permalloy bilayers, and a Pt/Permalloy control device. In Fig. S4, we plot $V_S$ and $V_A$ as a function of $\phi$ for four devices, along with fits to $S \sin(2\phi - 2\phi_0) \cos(\phi - \phi_0)$...
and $\sin(2\phi - 2\phi_i) [B + A\cos(\phi - \phi_i)]$ for the symmetric and antisymmetric data respectively. The sign of the parameter $B$ varies apparently randomly between devices. This is to be expected because Raman scattering does not allow us to distinguish between the $\hat{b}$ and $-\hat{b}$ directions, which are physically distinct for the WTe$_2$ surface crystal structure (a consequence of broken two-fold rotational symmetry). Essentially, the sign of $B$ depends on whether the positive $\hat{b}$ direction lies along $0 < \phi < 180^\circ$ or $180^\circ < \phi < 360^\circ$. Since interchanging the ground and signal leads rotates the definition of $\phi$ by $180^\circ$, the sign of $B$ is determined by the decision of which end of the bilayer is connected to the signal lead.

We carried out calibrated torque conductivity measurements (i.e., using a vector network analyzer to determine $I_{RF}$ as discussed in Supplementary Note 2) for 11 of our devices. The device-averaged torque conductivities for devices with current applied along the a-axis are reported in the main text. The torque conductivity data from all 11 devices is summarized in Fig. S5. In Fig. S5a and Fig. S5b we plot $\sigma_{s}$ and $\sigma_{a}$ respectively as a function of thickness. In Fig. S5c we plot $|\sigma_{a}|$ as a function of thickness for the subset of the 11 devices where current is applied along the a-axis, and in Fig. S5d we plot $|\sigma_{a}|$ as a function of $|\phi_{a}|$ for all 11 devices.

Supplementary Note 5: Symmetry analysis for current generated torques.

The torques acting on an in-plane magnetization can be written as

$$\tau_{||}(\hat{m}, E) = \tau_{||}(\phi, E) \hat{m} \times \hat{c} \quad \text{and} \quad \tau_{\perp}(\hat{m}, E) = \tau_{\perp}(\phi, E) \hat{c},$$

where we have explicitly included the dependence of the torques on the electric field, $E$, in the bilayer. These expressions are generic, since $\hat{m} \times \hat{c}$ and $\hat{c}$ are unit vectors forming a basis for the vectors perpendicular to $\hat{m}$.

The scalar pre-factors, $\tau_{||}(\phi, E)$ and $\tau_{\perp}(\phi, E)$, can be Fourier expanded:

$$\tau_{||}(\phi, E) = E \left( S_0 + S_1 \cos \phi + S_2 \sin \phi + S_3 \cos 2\phi + S_4 \sin 2\phi + S_5 \cos 3\phi + \ldots \right)$$

$$\tau_{\perp}(\phi, E) = E \left( A_0 + A_1 \cos \phi + A_2 \sin \phi + A_3 \cos 2\phi + A_4 \sin 2\phi + A_5 \cos 3\phi + \ldots \right).$$

First, we consider the case of an electric field applied along the WTe$_2$ crystal a-axis. In this case, applying the $\sigma_{v}(bc)$ symmetry operation to the device flips the direction of the electric field (since $E$ is a vector perpendicular to the bc plane) and reverses the component of the magnetization perpendicular to the a-axis (since $\hat{m}$ is a pseudovector). This is equivalent to the transformations $\phi \rightarrow -\phi$ and $E \rightarrow -E$.

The torques must also transform as pseudovectors under $\sigma_{v}(bc)$, which constrains the dependence of $\tau_{||}(\phi, E)$ and $\tau_{\perp}(\phi, E)$ on $\phi$ and $E$. The nature of these constraints can be understood by re-writing $\tau_{\perp}(\phi, E) = \hat{c} \cdot \hat{m} \times \hat{c}$ and $\tau_{||}(\phi, E) = (\hat{m} \times \hat{c}) \cdot \hat{c}$. Since $\hat{c}$ is a vector and $\hat{c} \cdot \hat{m} \times \hat{c}$ transforms as a pseudoscalar (i.e. changes sign under inversion and mirror operations but is invariant under rotations) as the dot product of a vector and a pseudovector is a pseudoscalar. Consistency of the transformations $\phi \rightarrow -\phi, E \rightarrow -E$ and $\hat{c} \rightarrow -\hat{c}$ under $\sigma_{v}(bc)$ then requires that $\tau_{\perp}(-\phi, -E) = -\hat{c} \cdot \hat{m} \times \hat{c} = -\tau_{\perp}(\phi, E)$. One can also show that the cross product of a vector and a pseudovector transforms as a vector, and so $\hat{m} \times \hat{c}$ is a vector. This implies that $(\hat{m} \times \hat{c}) \cdot \hat{c} \rightarrow -\hat{m} \times \hat{c}$ transforms as a pseudoscalar so that
\((\hat{m} \times \hat{c}) \cdot \hat{r}_\parallel \rightarrow -(\hat{m} \times \hat{c}) \cdot \hat{r}_\parallel\) under \(\sigma_i (bc)\), and therefore \(\tau_\parallel (\phi, -E) = -\tau_\parallel (\phi, E)\). We have considered only torques linear in \(E\) so that the symmetry requirement becomes \(\tau_\perp (\phi, E) = \tau_\perp (\phi, E)\). Keeping only the terms in Eq. (S6) that comply with this symmetry requirement leaves

\[
\tau_\parallel (\phi, E) = E \left( S_0 + S_1 \cos \phi + S_2 \cos 2\phi + S_3 \cos 3\phi + \ldots \right)
\]

\[
\tau_\perp (\phi, E) = E \left( A_0 + A_1 \cos \phi + A_2 \cos 2\phi + A_3 \cos 3\phi + \ldots \right) .
\]

The measured angular dependence discussed in the main text for \(E\) along the a-axis can be fit accurately with just the low-order terms \(S_0\), \(A_0\), and \(A_1\). Notably, we do not experimentally observe the term \(S_0\), although it is allowed by symmetry.

For an electric field applied along the b-axis, applying \(\sigma_i (bc)\) to the device flips the projection of the magnetization along the b-axis direction, and leaves the electric field unchanged i.e. \(\phi \rightarrow \pi - \phi\) and \(E \rightarrow E\). From this, one can derive the symmetry constraints \(\tau_\perp (\pi - \phi, E) = -\tau_\perp (\phi, E)\). Therefore the allowed angular dependencies of the torques for an electric field \(E\) along the b-axis are

\[
\tau_\parallel (\phi, E) = E \left( S_1 \cos \phi + S_2 \sin 2\phi + S_3 \cos 3\phi + \ldots \right)
\]

\[
\tau_\perp (\phi, E) = E \left( A_1 \cos \phi + A_2 \sin 2\phi + A_3 \cos 3\phi + \ldots \right) .
\]

In this case, with \(E\) along the b-axis, the lowest order terms \(S_1\) and \(A_1\) dominate our measurements for both the symmetric and antisymmetric amplitudes, although better agreement is obtained when we include the coefficient \(A_2\) as shown in Fig. S6.

**Supplementary Note 6: Higher harmonics in the ST-FMR angular dependence.**

Based on the symmetry analysis in Supplementary Note 5, we may expect that the angular dependence of the in- and out-of-plane torques can be more general than \(\tau_\perp = B + A \cos \phi\) and \(\tau_\parallel = S \cos \phi\). We examined fits of our data to the most general symmetry-allowed Fourier expansion, up to the third harmonic. We find significant values for \(A_2\) (i.e., the term proportional to \(\cos 3\phi\)) with the largest magnitudes occurring for current flowing close to the b-axis direction. Figure S6 shows \(V_s\) as a function of \(\phi\) for two devices, along with fits to

\[
\sin(2\phi - 2\phi_0) \left[ B + A \cos(\phi - \phi_0) \right] \text{ and } \sin(2\phi - 2\phi_0) \left[ B + A \cos(\phi - \phi_0) + C \cos(3\phi - 3\phi_0) \right] ;
\]

the \(\cos 3\phi\) term significantly improves the fit, corresponding to a non-zero value of \(A_2\). We also find significant values for \(S_2\), but \(S_2 / A_1\) is typically similar in magnitude to its value for our Pt/Pt control device \((S_2 / A_1 = -0.10 \pm 0.02)\). All other coefficients up to the third harmonic, except for those discussed in the main text, are zero within our experimental uncertainty.

The \(\cos 3\phi\) term might arise either from a true angular dependence of the torque or from a lack of full saturation for the in-plane anisotropic magnetoresistance \(R(\phi)\) due to in-plane magnetic anisotropy. Our initial analyses suggest that this in-plane anisotropy can account at least partially, but perhaps not completely, for our measured \(\cos 3\phi\) term. This mechanism cannot affect our determination of the \(\tau_\parallel\) torque.
**Supplementary Note 7:** On why there can be no contribution to the out-of-plane antidamping torque from the bulk of a WTe$_2$ layer.

Bulk crystals of WTe$_2$ have a screw symmetry: the crystal structure is mapped onto itself if it is rotated by 180° about an axis normal to the layers (c-axis) and translated by half a unit cell along both the c and a-axis (in the c direction, half a unit cell is one layer spacing). If there is any net bulk spin polarization or spin current with a component perpendicular to the plane, that spin component will be left unaltered by this operation, while the direction of an in-plane charge current will be reversed. This implies that there can be no bulk contribution to the current-induced antidamping spin torque that is linear in the applied in-plane current (see also Supplementary Note 8). This screw symmetry is broken at the WTe$_2$/Py interface, so a surface-generated out-of-plane antidamping torque is allowed by symmetry. This surface contribution might come entirely from a single WTe$_2$ layer at the interface or from imperfect cancellations between more than one WTe$_2$ layer near the interface (e.g., if there is surface-induced band bending).

We have checked that adjacent layers generate $\tau_B$ of opposite sign by studying a sample (Device S1) in which the sample region contains a single-layer step, so that the Permalloy is exposed to two WTe$_2$ surfaces with opposite symmetry (Fig. S8). Device S1 was fabricated with the bar aligned at 3.7° from the a-axis and with a monolayer step dividing the channel into two regions of approximately equal area, as shown by the atomic force microscopy data in Figs. S8a and S8b. The angular dependences of $V_S$ and $V_A$ are shown in Fig. S8c. The non-zero value of $V_S$ implies the existence of spin-orbit torque and a clean WTe$_2$/Py interface. However, we measure $B/A=0.033$ for this device, in contrast to our finding that $|B/A|>0.32$ for all devices measured with $|\phi_s|<10^5$ and an atomically flat channel. We interpret this low value of $B/A$ in device S1 as arising from cancellation of the torques from the two WTe$_2$/Py interface regions of opposite surface symmetry, providing strong evidence that $\tau_B$ arises from an interface effect. Similar results were obtained on two additional devices containing a monolayer step and with the bar direction aligned to the WTe$_2$ a-axis.

**Supplementary Note 8:** Some comments on the microscopic origin of an out-of-plane antidamping torque in WTe$_2$/Py bilayers.

In the main text we have taken a conservative approach to interpreting our data: we demonstrated the consistency of the observed torques with the symmetries of the WTe$_2$ surface, while avoiding speculation regarding microscopic mechanisms. In this section, we discuss a few possible microscopic mechanisms for generation of out-of-plane antidamping torques, with the understanding that these possibilities are not exhaustive. We focus on mechanisms that can generate transport and accumulation of spins polarized in the c-direction, since absorption of c-axis polarized spins is expected to lead to a $\hat{m} \times (\hat{m} \times \hat{c})$ torque. To start, we show that symmetry constraints forbid a nonzero contribution from two well-known effects generating spin-orbit torques: a bulk spin-Hall conductivity, and a bulk-averaged inverse spin galvanic effect. We then consider possible mechanisms for which non-zero contributions are allowed.
To generate a $\hat{m} \times (\hat{m} \times \hat{c})$ torque via the bulk spin-Hall effect, we must have c-axis polarized spins flowing towards the WTe$_2$/Py interface in response to an in-plane electric field. The total current of c-axis polarized spins, $\vec{j}_c$, can be written as $\vec{j}_c = \sigma^c \cdot \vec{E}$, where $\sigma^c$ is the c-axis polarized part of the spin-Hall conductivity tensor. The form of this tensor is constrained by the point group of the crystalS9. For the mm2 point group operations of WTe$_2$, the most general form is:

$$\sigma^c = \begin{pmatrix} 0 & \sigma_{ab} & 0 \\ \sigma_{ba} & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \quad (S9)$$

Notably, the terms $\sigma_{ba}$ and $\sigma_{cb}$, corresponding to c-axis polarized spins flowing in the c-direction (towards the WTe$_2$/Py interface) in response to in-plane electric fields, are zero. Therefore, there can be no contribution to a $\hat{m} \times (\hat{m} \times \hat{c})$ torque from the bulk spin Hall effect in WTe$_2$.

When an electric field is applied to a non-centrosymmetric crystal we expect a nonequilibrium spin-density to be generated in the crystal due to the inverse spin galvanic effect. This spin polarization can also be written in terms of a linear response tensor: $\vec{\sigma} = \chi \cdot \vec{E}$. The tensor $\chi$ must satisfy the relation $\chi = \det(S)S^{-1}\chi S$ for any symmetry operation $S$ in the point group of the crystalS10. The point group rather than the space group is relevant here because we assume the spin density to have a nonzero component that is spatially uniform. For WTe$_2$, the most general form is:

$$\chi = \begin{pmatrix} 0 & \chi_{ab} & 0 \\ \chi_{ba} & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \quad (S10)$$

Since $\chi_{cb}$ and $\chi_{ac}$ are zero, the bulk inverse spin galvanic effect of WTe$_2$ cannot generate a $\hat{m} \times (\hat{m} \times \hat{c})$ torque.

The symmetry of WTe$_2$ does, however, allow for local accumulations of c-axis polarized spins in response to an in-plane electric field, provided these accumulations switch sign between atomic sites related by the screw-axis and glide-plane symmetries. This is similar to recent work on CuMnAs, where the absence of local inversion symmetry allows for current-induced exchange fields that change sign between atomic sites related by the global inversion symmetryS11. The WTe$_2$ crystal can be partitioned into adjacent A and B type layers, where B layers are rotated by 180° with respect to A layers. The symmorphic bc mirror plane maps every layer back onto itself, while the non-symmorphic symmetries (screw-axis and glide-plane) map each layer onto an adjacent one of the opposite type. If we define layer specific spin accumulations $\vec{\sigma}^A = \chi^A \cdot \vec{E}$ and $\vec{\sigma}^B = \chi^B \cdot \vec{E}$ the respective tensors obey:

$$\chi^A = \begin{pmatrix} 0 & \chi_{ab} & \chi_{ac} \\ \chi_{ba} & 0 & 0 \\ \chi_{ca} & 0 & 0 \end{pmatrix}, \chi^B = \begin{pmatrix} 0 & \chi_{ab} & -\chi_{ac} \\ \chi_{ba} & 0 & 0 \\ -\chi_{ca} & 0 & 0 \end{pmatrix}. \quad (S11)$$
Therefore, it is possible to generate local c-axis spin polarizations in the bulk WTe$_2$ crystal via an in-plane current, but the local c-axis spin polarizations change sign between layers. In a real crystal the surface will terminate on either an A or B type layer, leading to a c-axis spin polarization on the surface when current is applied along the a-axis. This mechanism is expected to lead to a $\hat{m} \times (\hat{m} \times \hat{c})$ torque, along with a $\hat{m} \times \hat{c}$ torque due to exchange coupling of the ferromagnet to the WTe$_2$ surface spins.

Another approach is to consider the torques generated in an interface layer formed by hybridization between electronic states of the WTe$_2$ and Py i.e. in a region at the WTe$_2$/Py interface with electronic properties differing from the bulk of either layer. These interface states could generate c-axis polarized spin accumulations via the inverse spin galvanic effect. For example, the spin-orbit coupling Hamiltonian $H_{SOC} \propto \hat{n} \cdot (\vec{k} \times \vec{\sigma})$, where $\hat{n}$ lies in the bc plane, is consistent with the symmetry of the WTe$_2$/Py interface, and leads to a non-zero $\langle \sigma_z \rangle$ in response to electric fields applied along the a-axis. This is a generalization of the usual Rashba-Edelstein effect discussed in the context heavy metal/ferromagnet bilayers, which corresponds to $\hat{n} = \hat{c}$. Such a $\langle \sigma_z \rangle$ can generate both $\hat{m} \times (\hat{m} \times \hat{c})$ and $\hat{m} \times \hat{c}$ torques, with their relative magnitude depending on microscopic details. Magnetic anisotropy associated with this mechanism has been predicted to arise at the interface between ferromagnets and low-symmetry materials with strong spin-orbit coupling$^{512}$.

Recent theoretical work suggests that it may also be possible that the spin-polarized electrons flowing within a metallic ferromagnet layer may generate spin-transfer torque when they scatter from an interface with a material possessing strong spin-orbit coupling, without necessarily requiring charge current flow within the spin-orbit material$^{513,514}$. This mechanism is attractive because it might provide a natural explanation for the apparent lack of dependence on the WTe$_2$ thickness for any of the torque components $\tau_B$, $\tau_A$, and $\tau_S$.

**Supplementary Note 9: Second-harmonic Hall measurements for a WTe$_2$/Py bilayer.**

We are grateful to a Reviewer for pointing out that second-harmonic measurements of Hall voltage as a function of the angle of an in-plane applied magnetic field, $B$, provide an alternative method to measure an out-of-plane antidamping torque independent of the ST-FMR measurements discussed in the main text. We performed this measurement using the Hall bar device discussed in Supplementary Note 3, for which the Permalloy thickness is 6 nm and the WTe$_2$ thickness is 16 nm. The Hall bar has a length and width of 26 $\mu$m and 4 $\mu$m, respectively, and is oriented so that the current is along the WTe$_2$ a-axis ($\phi_{\perp} = -1^\circ$); the voltage probes used for the Hall measurements are 2 $\mu$m wide. The active region of the Hall bar has a uniform WTe$_2$ thickness, with no monolayer steps, over better than 90% of its area. We apply a current $I(t) = I_0 \sin(2\pi ft)$ at a frequency $f$=340 Hz with $I_0 = 0.66$ mA, and measure the Hall voltage at the second harmonic frequency. The angle of the in-plane magnetic field $\phi$ is defined relative to the direction of current flow. Generalizing the argument in Ref. S15 to include the effects of an in-plane uniaxial anisotropy $B_\perp$ with the easy axis parallel to the b-axis of the WTe$_2$ (in addition to the shape anisotropy of the thin film $\mu_0 M_{eff}$), and allowing for in-plane and out-of-
plane current-induced torques with the angular dependence \( \tau = \tau_s \cos \phi_M \) and \( \tau_\perp = \tau_A \cos \phi_M + \tau_B \), the second harmonic signal has the form:

\[
R_{xy}^{2\omega} = \frac{R_{\text{PHE}} \cos 2\phi_M \left( \tau_A \cos \phi_M + \tau_B \right)}{\gamma \left( B - B_\perp \cos 2\phi_M \right)} + \frac{R_{\text{AHE}} \tau_A \cos \phi_M}{2\gamma \left( B + \mu_B M_{\text{eff}} + B_\perp \sin^2 \phi_M \right)} + \frac{V_{\text{ANE}} \cos \phi_M}{\tau_B},
\]

(S12)

where \( R_{\text{PHE}} \) and \( R_{\text{AHE}} \) are the planar and anomalous Hall resistances of the device, and \( V_{\text{ANE}} \) is the anomalous Nernst voltage arising from an out-of-plane thermal gradient proportional to the Joule power \( I^2 R \). This expression neglects terms above first order in \( B_A / B \), which is an accurate approximation over the range of fields studied for our second harmonic measurements. Here \( \phi_M \) is the angle of the magnetization relative to the direction of current flow, which differs from \( \phi \) for low-fields due to the in-plane magnetic anisotropy. To first order in \( B_A / B \), the equilibrium magnetization angle is \( \phi_M = \phi + B_A \sin 2\phi / 2B \). Equation S12 shows that the second harmonic signal associated with \( \tau_B \) has an angular dependence distinct from \( \tau_A \), \( \tau_s \) and the magneto-thermopower voltage \( V_{\text{ANE}} \).

Fig. S9a shows measurements of the second-harmonic Hall voltage in the WTe\(_2\)/Py Hall bar as a function of \( \phi \) for selected magnitudes of applied magnetic field \( B \); the red lines indicate the data, while the black lines are fits to Eq. S13. Even without any fitting, it is clear that the out-of-plane antidamping torque \( \tau_B \) is indeed non-zero, as the magnitude of the second-harmonic signal is significantly different for \( \phi = 180^\circ \) and \( 360^\circ \) [when \( \tau_B = 0 \), Eq. S12 predicts simply that \( R_{xy}^{2\omega} (\phi = 180^\circ) = -R_{xy}^{2\omega} (\phi = 360^\circ) \)]. To fit the data, we use a simplified version of Eq. S12, valid when \( B \ll \mu_B M_{\text{eff}} \):

\[
R_{xy}^{2\omega} = \frac{\cos 2\phi_M}{(B - B_\perp \cos 2\phi_M)} \left( A_0 \cos \phi_M + A_1 \right) + R_0 \cos \phi_M,
\]

(S13)

where \( A_0 = R_{\text{PHE}} \tau_B / \gamma \), \( A_1 = R_{\text{PHE}} \tau_A / \gamma \), and \( R_0 \) is a constant combining the contributions of the in-plane antidamping torque and the anomalous Nernst voltage. For each value of \( B \) we fit the data using the parameters \( A_0 \), \( A_1 \), \( R_0 \), and \( B_A \), along with an additional overall \( \phi \)-independent offset. For the fits, we used the first-order expression for \( \tau_M (\phi) \) discussed above. We find that Eq. S13 fits the data well with \( B_A \approx 3 \text{ mT} \). The torque ratio \( \tau_B / \tau_A \) can then be determined independent of any other sample parameters at each value of the field magnitude, \( A_0 / A_1 = \tau_B / \tau_A \). In figure S9b we plot \( \tau_B / \tau_A \) as a function of \( B \), showing that \( \tau_B / \tau_A \approx 0.20 \rightarrow 0.25 \). These values are similar to, albeit slightly lower than, the values of \( |\tau_B| / \tau_A \) determined by ST-FMR for different devices (|\( \tau_B | / \tau_A = 0.32-0.385 \); see Fig. 4b in the main text or Table S1).

We determine the individual torque conductivities \( \sigma_A \) and \( \sigma_B \) from the second harmonic Hall measurements according to (here the subscript \( K = A \) or \( B \)):

\[
\sigma_k = \frac{M_{\text{in}} \tau_k}{h \gamma / 2e} = \frac{e M_{\text{in}} \tau_k}{\mu_B} \frac{\tau_k}{V \left( 2e \right)}.
\]

(S14)
Using Eq. S13, and $R_{\text{PHIE}} = 0.14 \ \Omega$, for the harmonic Hall measurement with $B = 1000 \ \text{Oe}$ we find \( \tau_A = 8.3 \pm 0.2 \ \text{MHz} \) and \( \tau_B = 2.12 \pm 0.09 \ \text{MHz} \). To estimate the applied electric field we divide the applied voltage (566 mV peak-to-peak) by the length of the Hall device, and to estimate the saturation magnetization \( M_s \approx M_{\text{eff}} \) we fit to the anomalous Hall effect data of Fig. S3 finding \( \mu_0 M_{\text{eff}} = 0.81 \ \text{T} \pm 0.01 \ \text{T} \). From Eq. (S14) we then find \( \sigma_B = (6 \pm 1) \times 10^3 \ (\hbar/2e) \ (\Omega m)^{-1} \) and \( \sigma_A = (25 \pm 4) \times 10^3 \ (\hbar/2e) \ (\Omega m)^{-1} \), where the errors are primarily due to the uncertainty in the thickness of the Permalloy. These values can be compared with the calibrated ST-FMR measurements presented in Fig. S5. The calibrated ST-FMR measurements for devices with $|\phi_s| \leq 10^\circ$ give a range of \( \sigma_B = (3-5) \times 10^3 \ (\hbar/2e) \ (\Omega m)^{-1} \) and \( \sigma_A = (8-14) \times 10^3 \ (\hbar/2e) \ (\Omega m)^{-1} \). The second-harmonic value for \( \sigma_B \) agrees with the ST-FMR measurements within the range of reasonable experimental uncertainty. The value of \( \sigma_A \) as determined from the second-harmonic measurements is approximately twice as large as the typical ST-FMR value. This discrepancy in \( \sigma_A \) is not presently understood but there may be differences in the WTe$_2$ crystal quality or the cleanliness of the WTe$_2$/Py interface, as the Py film used for the Hall bar device was grown in a different round of sputtering depositions than those used for the ST-FMR devices.

We conclude that the second-harmonic Hall measurements confirm the existence of a nonzero out-of-plane antidamping torque \( \tau_B \) and give a value for its strength in agreement with the ST-FMR measurements.
Figure S1: Resistance of Device 1 (red) as a function of applied in-plane magnetic field angle. Measurements are made in a Wheatstone bridge configuration with a static magnetic field of 0.08 T. The fit (black) is used to extract values of $dR/d\phi$.

$$R_0 + \Delta R \cos^2(\phi - \phi_0)$$
Figure S2: Ferromagnetic resonance field as a function of the in-plane magnetization angle for (a) Device 1 and (b) Device 2. The data are represented by red circles and the black lines are the indicated fits. In both cases the applied microwave frequency is 9 GHz and the power is 5 dBm. The blue arrows indicates the values of $\phi$ for which the magnetization lies along the b-axis. Error bars represent estimated standard deviations from the least-squares fitting procedure.
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<th>$l \times w$ (μm) ±0.2 μm</th>
<th>$\tau_B / \tau_A$</th>
<th>$\tau_S / \tau_A$</th>
<th>$\phi_{a-I}$ (Degrees) ±2°</th>
<th>$B_A$ (mT)</th>
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Supplemental Table S1: Comparison of device parameters for the WTe$_2$/Py bilayers discussed in the main text and a Pt/Py control device. $t$ is the thickness of the WTe$_2$ or Pt, $l$ is the sample length, $w$ is the sample width, $\tau_B / \tau_A$ and $\tau_S / \tau_A$ are the torque ratios defined in the main text, $\phi_{a-I}$ is the angle between the a-axis and the applied current, $B_A$ is the anisotropy field within the sample plane (see Extended Data Figure S2), and $\phi_{Easy-I}$ is the angle of the magnetic easy axis with respect to the applied current.
## Supplemental Table S1: Comparison of device parameters for the WTe$_2$/Py bilayers discussed in the main text and a Pt/Py control device.

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<th>Device Number</th>
<th>t (nm)</th>
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<th>l (μm)</th>
<th>$\pm 0.2$ μm</th>
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$\theta$ is the thickness of the WTe$_2$ or Pt, $l$ is the sample length, $w$ is the sample width, $\tau_B / \tau_A$ and $\tau_S / \tau_A$ are the torque ratios defined in the main text, $\phi_{a-I}$ is the angle between the $a$-axis and the applied current, $B_A$ is the anisotropy field within the sample plane (see Extended Data Figure S2), and $\phi_{Easy-I}$ is the angle of the magnetic easy axis with respect to the applied current.

**Figure S3:** Measurements of transverse resistance, $R_{\text{Hall}}$, for a WTe$_2$/Py (6 nm) Hall bar with the magnetic field oriented perpendicular to the sample plane (a) and parallel to the WTe$_2$ b-axis in the sample plane (b) with current directed along the WTe$_2$ a-axis. The contribution of the ordinary Hall effect in (a) has been subtracted. The peak-to-peak anomalous Hall effect contribution to $R_{\text{Hall}}$, $R_{\text{AHE}}$, is 0.62 $\Omega$, as extracted from (a). The inset to (b) shows $R_{\text{Hall}}$ versus the applied field along the b-axis with an expanded vertical scale. The small variation (0.007 $\Omega$) in (b) is consistent with a planar Hall effect.
Figure S4: Plots of the symmetric (blue circles) and antisymmetric (red circles) components of the ST-FMR mixing voltage for (a) Device 3, (b) Device 7, (c) Device 10, and (d) Device 2. The current in Device 3 is applied approximately along the a-axis of the WTe$_2$, with the angle turning increasingly toward the b-axis for Devices 7, 10, and 2. The microwave frequency is 9 GHz and the microwave power is 5 dBm. The solid blue lines are fits of $S \sin(2\phi - 2\phi_0)\cos(\phi - \phi_0)$ to $V_S(\phi)$ and the solid red lines are fits of $\sin(2\phi - 2\phi_0)[B + A\cos(\phi - \phi_0)]$ to $V_A(\phi)$. Error bars represent estimated standard deviations from the least-squares fitting procedure.

Figure S5: a) Torque conductivity $\sigma_S$ as a function of WTe$_2$ thickness for the 11 devices on which we used a vector network analyzer to perform fully-calibrated measurements. The current is applied at various angles to the WTe$_2$ a-axis. b) Torque conductivity $\sigma_A$ as a function of WTe$_2$ thickness for these 11 devices. c) Torque conductivity $\sigma_B$ as a function of WTe$_2$ thickness for 6 fully-calibrated devices with $\phi_{a-I} < 10^\circ$. d) $\sigma_B$ as a function of $\phi_{a-I}$ for the 11 devices used in panels a) and b). e) $\sigma_S$ as a function of $\phi_{a-I}$ for the 11 devices used in panels a) and b). f) $\sigma_A$ as a function of $\phi_{a-I}$ for the 11 devices used in panels a) and b). For the data shown in panels a)-f), the applied microwave power is 5 dBm, and the torque conductivities are averaged over the frequency range 8-11 GHz. Error bars represent estimated standard deviations based on error propagation including uncertainties in calibrating the microwave voltage applied across each device and uncertainties derived from least-squares fits to ST-FMR data.
Figure S5: a) Torque conductivity $\sigma_S$ as a function of WTe$_2$ thickness for the 11 devices on which we used a vector network analyzer to perform fully-calibrated measurements. The current is applied at various angles to the WTe$_2$ a-axis. b) Torque conductivity $\sigma_A$ as a function of WTe$_2$ thickness for these 11 devices. c) Torque conductivity $|\sigma_B|$ as a function of WTe$_2$ thickness for 6 fully-calibrated devices with $|\phi_{a-I}| < 10^\circ$. d) $|\sigma_B|$ as a function of $|\phi_{a-I}|$ for the 11 devices used in panels a) and b). e) $\sigma_S$ as a function of $|\phi_{a-I}|$ for the 11 devices used in panels a) and b). f) $\sigma_A$ as a function of $|\phi_{a-I}|$ for the 11 devices used in panels a) and b). For the data shown in panels a-f, the applied microwave power is 5 dBm, and the torque conductivities are averaged over the frequency range 8-11 GHz. Error bars represent estimated standard deviations based on error propagation including uncertainties in calibrating the microwave voltage applied across each device and uncertainties derived from least-squares fits to ST-FMR data.
Figure S6: Plots of the antisymmetric part of the mixing voltage (red circles) versus the in-plane magnetization angle for (a) Device 2 and (b) Device 7. The microwave frequency is 9 GHz and the microwave power is 5 dBm. The black lines show fits to $\sin(2\phi - 2\phi_0) \left[ B + A \cos(\phi - \phi_0) + C \cos(3\phi - 3\phi_0) \right]$, giving $C/A = -0.24 \pm 0.01$ for Device 2 and $C/A = -0.20 \pm 0.01$ for Device 7. The light grey lines show fits to $\sin(2\phi - 2\phi_0) \left[ B + A \cos(\phi - \phi_0) \right]$. Error bars represent estimated standard deviations from the least-squares fitting procedure.
Figure S7: a) An atomic force microscopy image of the WTe₂ flake used for fabrication of Device 15 after deposition of the Permalloy layer and aluminum oxide cap but before any lithographic processing. The active region used for the device (dashed white box) has a RMS surface roughness < 300 pm. b) A linecut [white line in (a)] from the edge of the WTe₂ flake, showing an average thickness of 5.5 nm.
Fig S8: a) An atomic force microscopy image of the WTe$_2$ flake used for fabrication of Device S1 after deposition of the Permalloy layer and aluminum oxide cap. The dashed white rectangle shows the approximate placement of the device active region (the uncertainty in the lateral location is about 500 nm due to the alignment procedure for the lithography steps). b) A linecut [white solid line in (a)] showing a step height of about 0.7 nm corresponding to a monolayer step in the WTe$_2$ crystal. c) Plot of the symmetric (top, red circles) and antisymmetric parts (bottom, red circles) of the mixing voltage versus the in-plane magnetization angle. The magnitude of the symmetric part indicates a spin-orbit torque comparable to other a-axis aligned WTe$_2$ devices, but the antisymmetric part shows $B/A=0.033$ indicating that $\tau_{ll}$ is much smaller here than in devices without a monolayer step. Error bars represent estimated standard deviations from the least-squares fitting procedure.
Figure S9: a) Second harmonic Hall voltage for a WTe$_2$/Py bilayer (with current along the a-axis) as a function of the angle between the in-plane applied magnetic field and the current flow direction. The data (red) are plotted for different magnitudes of the applied magnetic field (B=0.25 T, 0.1 T, 0.04 T, and 0.02 T, from top to bottom). Data for different values of the applied field have been vertically offset for clarity. The black lines show fits to Eq. S12. b) The torque ratio $\tau_B/\tau_A$ extracted from the angular dependence of the second harmonic Hall voltage, as a function of the magnitude of the applied magnetic field used for the angular sweep. Error bars represent estimated standard deviations from the least-squares fitting procedure.
Figure S10: a) Polarized Raman spectra with the orientation of the electric field of the excitation, \( E \), parallel to the WTe\(_2\) a-axis (black) and parallel to the WTe\(_2\) b-axis (red) for Device 4. Traces are normalized by the silicon substrate peak for ease of comparison (not shown). \( P6 = 165.7 \text{ cm}^{-1} \) and \( P7 = 211.3 \text{ cm}^{-1} \) (as defined in Ref. S12) b) The ratio of intensities for \( P6/P7 \) (blue circles) plotted as a function of angle between the current (lithographically defined bar) direction and the linearly polarized Raman excitation as defined in the inset. The orientation of the WTe\(_2\) a-axis is determined from the angle that maximizes the fit (red) to a \( \cos^2(\phi_{\text{Raman}}) \) type dependence\(^{S16}\). The directions \( \hat{b} \) and \( -\hat{b} \) are not differentiated by Raman scattering.
Supplemental References


