Anomalous Spin-Orbit Torques in Magnetic Single-Layer Films

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Abstract:

The spin Hall effect couples charge and spin transport¹⁻³, enabling electrical control of magnetization^{4,5}. A quintessential example of SOI-induced transport is the anomalous Hall effect (AHE)⁶, first observed in 1880, in which an electric current perpendicular to the magnetization in a magnetic film generates charge accumulation on the surfaces. Here we report the observation of a counterpart of the AHE that we term the anomalous spin-orbit torque (ASOT), wherein an electric current parallel to the magnetization generates opposite spin-orbit torques on the surfaces of the magnetic film. We interpret the ASOT as due to a spin-Hall-like current generated with an efficiency of 0.053 ± 0.003 in Ni₈₀Fe₂₀, comparable to the spin Hall angle of Pt⁷. Similar effects are also observed in other common ferromagnetic metals, including Co, Ni, and Fe. First principles calculations corroborate the order of magnitude of the measured values. This work suggests that a strong spin current with spin polarization transverse to magnetization can be generated within a ferromagnet, despite spin dephasing⁸. The

large magnitude of the ASOT should also be taken into consideration when investigating spin-orbit torques in ferromagnetic/nonmagnetic bilayers.

The spin Hall effect can convert a charge current into a perpendicular flow of spin angular momentum (spin current)⁹. One of its manifestations in a magnetic conductor is the AHE¹⁰, illustrated in Fig. 1a. Due to the imbalance of electrons with spins parallel and antiparallel to the magnetization, the flow of spin current results in charge accumulation on the top and bottom surfaces. The spin current in this configuration is polarized parallel with the magnetization¹¹⁻¹³. Applying similar considerations to the configuration illustrated in Fig. 1b, in which the electric current is parallel to the magnetization, a spin current can flow between the top and bottom surfaces of the magnetic conductor, except with electron spins transverse to the magnetization. In single-layer ferromagnets with bulk inversion symmetry, the transversely polarized spin current does not give rise to a bulk spin torque (those with broken bulk inversion symmetry have been shown to exhibit a non-zero bulk spin-orbit torque^{14,15}). Instead, we predict that it will result in net anomalous spin-orbit torque (ASOT) on the top and bottom surfaces, where inversion symmetry is broken (see Supplementary Information section S1). It should be noted that the term "anomalous" here does not mean the ASOT has different behavior from conventional spin-orbit torque⁵ – the two have the same symmetry - but rather is used to illustrate its connection with the AHE. Both ASOT and AHE are spin-orbit interaction-induced phenomena that can only be observed in single-layer magnetic conductors, under different current and magnetization configurations, as illustrated in Figs. 1a and 1b.



Figure 1 Illustrations of the anomalous Hall effect and anomalous spin-orbit torque. a, In the anomalous Hall effect (AHE) a charge current I (black arrow) perpendicular to the magnetization \mathbf{m} (yellow arrows) generates a flow of spin current (grey arrows) in the z-direction. Here blue arrows on purple spheres represent spin directions of electrons. Due to the imbalance of majority and minority electrons, the flow of spin current results in spin and charge accumulation on the top and bottom

surfaces. **b**, When a charge current is applied parallel with the magnetization, the AHE vanishes, but spin-orbit interaction generates a flow of transversely polarized spin current that gives rise to anomalous spin-orbit torque (ASOT). The ASOTs (red arrows) are equivalent to out-of-plane fields (green arrows) that tilt the magnetization out of plane. $\tau_{\rm T}^{\rm ASOT}$ ($\tau_{\rm B}^{\rm ASOT}$) and $h_{\rm eff}^{\rm T}$ ($h_{\rm eff}^{\rm B}$) are the ASOTs and equivalent fields at the top (bottom) surfaces, respectively. **c**, Simulated distribution of the out-of-plane magnetization $m_{\rm z}$ in a 32 nm Py film driven by equal and opposite ASOTs on the surfaces, scaled by the maximum value.

Interconversion between transversely polarized spin current and charge current has been recently studied in ferromagnetic multilayers¹⁶⁻¹⁹ with considerable spin-charge conversion efficiency. Due to strong spin dephasing^{8,20}, transversely polarized spin current decays rapidly near the surface of the ferromagnet; therefore, the spin-charge conversion observed in these studies are likely due to interfacial spin-orbit interaction²¹. Transversely polarized spin current generated in the bulk of ferromagnets has yet to be demonstrated. Recently it has been theoretically predicted that transversely polarized spin current, competes with spin dephasing. In this paper, we also show that transversely polarized spin current can exist in ferromagnets in the clean limit, using first-principles calculations. We refer to the mechanism of the current-induced transversely polarized spin current in the bulk ferromagnet as the transverse spin Hall effect (TSHE). We emphasize that the TSHE is different from previously studied spin current generation in the AHE configuration¹², where the spin polarization is necessarily parallel with the magnetization.

Under the assumption that the current-induced ASOT in a ferromagnet results in a small perturbation to the magnetization, the ASOTs are equivalent to effective magnetic fields in the *z*-direction²³ that tilt the magnetization out of plane, as illustrated in Fig. 1b.

The out-of-plane magnetization tilting, m_z^{ASOT} , due to the ASOT at the top (τ_T^{ASOT})

and bottom ($\tau_{\scriptscriptstyle B}^{\scriptscriptstyle ASOT}$) surfaces can be derived as

$$m_{z}^{\text{ASOT}}(z) = \frac{\tau_{\text{T}}^{\text{ASOT}} \cosh \frac{d-z}{\lambda} + \tau_{\text{B}}^{\text{ASOT}} \cosh \frac{z}{\lambda}}{\lambda \sinh \frac{d}{\lambda} (|H_{\text{ext}}| + M_{\text{eff}}) \mu_{0} M_{\text{S}}} m_{x}, \qquad (1)$$

where *d* is the total thickness of the film, λ is the exchange length, H_{ext} is an applied external magnetic field in the *x*-direction, M_{eff} is the effective demagnetizing field, M_{s} is the saturation magnetization, and m_{x} is the projection of the unit magnetization along the *x*-direction. Here, the ASOT is assumed to be located only at the surfaces and the surface anisotropy is neglected. (See Supplementary Information section S4 for the derivation of Eq. (1), a discussion of why ASOT can be treated as a pure surface effect, and a numerical analysis that takes into account the surface anisotropy.)

Because exchange coupling in the magnetic material aligns the magnetization, the spatially-antisymmetric magnetization tilting is expected to be measurable only when the magnetic material is thicker than the exchange length (e.g. 5.1 nm for Ni₈₀Fe₂₀). A simulation of the out-of-plane magnetization distribution due to ASOT in a 32 nm Ni₈₀Fe₂₀ (Py) film is shown in Fig. 1c.

То observe ASOT, fabricate we a sample with structure substrate/AlO_x(2)/Py(32)/AlO_x(2)/SiO₂(3), where the numbers in parentheses are thicknesses in nanometers; the substrate is fused silica, which allows optical access to the bottom of the sample. Py is chosen because it is magnetically soft and widely used for the study of spin-orbit torques. The film is lithographically patterned into a 50 μ m \times 50 µm square and connected by gold contact pads, as shown in Fig. 2(a). When an electric current I of 40 mA is applied directly through the sample, ASOTs at the top $(\tau_{\rm T}^{\rm ASOT})$ and bottom $(\tau_{\rm B}^{\rm ASOT})$ surfaces lead to non-uniform magnetization tilting, as described by Eq. (1). When a calibration current I_{Cal} of 400 mA is passed around the sample, an out-of-plane Oersted field $\mu_0 h_{Cal} \approx 0.85$ mT is generated that uniformly tilts the magnetization out of plane, which is used for calibrating the magnitude of the ASOTs:

$$m_{\rm z}^{\rm Cal}(z) = \frac{h_{\rm Cal}}{\left|H_{\rm ext}\right| + M_{\rm eff}}.$$
(2)

We detect the magnetization changes using the polar magneto-optic Kerr effect (MOKE) by measuring the Kerr rotation θ_k and ellipticity change ε_k of the polarization of a linearly polarized laser reflected from the sample^{24,25}. The penetration depth of the laser in Py is approximately 14 nm, which is less than half the thickness of the 32 nm Py. Therefore, the MOKE response is more sensitive to the ASOT-induced out-of-plane magnetization $m_z^{ASOT}(z)$ on the surface on which the laser is directly incident.

The Kerr rotation due to ASOT as a function of the external field (shown in Figs. 2c and d) resembles a magnetization hysteresis, as can be understood from Eq. (1). The

overall offsets of the Kerr rotation signals are due to a residual, current-induced out-ofplane Oersted field due to imprecision in locating the MOKE probe spot exactly in the center of the $50 \times 50 \ \mu\text{m}^2$ sample, (see Supplementary Information Fig. S4b for MOKE signal dependence on the laser spot position), which does not depend on the inplane magnetization orientation²³. In contrast, when a uniform calibration field h_{Cal} is applied, the Kerr rotation is symmetric as a function of external field H_{ext} (see Fig. 2e and f), consistent with Eq. (2). The Kerr rotation due to ASOT on the top (Fig. 2c) and bottom (Fig. 2d) surfaces are the same sign, in agreement with our phenomenological model (Fig. 1c), which predicts the bottom ASOT has similar magnitude but opposite sign as the top ASOT. In contrast, the Kerr rotation due to the calibration field (Fig. 2e and f) changes sign because h_{Cal} is reversed upon flipping the sample.



Figure 2 Symmetry of the anomalous spin-orbit torque. Diagrams of the measurement configurations with the laser incident on \mathbf{a} , the top and \mathbf{b} , the bottom of the sample. The plots below each diagram correspond to signals measured in that diagram's configuration. **c-d**, The measured Kerr rotation signals for when current is applied through the sample, which arise from ASOTs. **e-f**, The measured Kerr rotation signals for when the calibration field h_{Cal} is applied.

As shown in Fig. 3a, the polar MOKE response due to ASOT is linear with applied electric current, indicating no significant heating-related effects up to 5×10^{10} A/m² current density. As shown in Fig. 3b, the polar MOKE response exhibits a cosine dependence on the relative angle between the electric current and the magnetization, consistent with Eq. (1).

Unlike the Oersted field, which depends on the total current, ASOT should depend current density. To confirm this. series on the we grow a of $AlO_x(2)/Py(t)/AlO_x(2)/SiO_2(3)$ films on silicon substrates with 1 µm-thick thermal oxide, where t varies from 4 nm to 48 nm. For all samples, we apply the same current density of 5×10^{10} A/m², and use MOKE to quantify the ASOT. To fit the measured MOKE results, we use a propagation matrix method²⁴ (see method section and Supplementary Information S5) to numerically simulate the MOKE signal as a function of the Py thickness. As presented in Fig. 3c, the validity of the method is first verified by a thickness-dependent calibration measurement, where a uniform 0.85 mT out-ofplane calibration field is applied to all samples. To extract the ASOT amplitude, the top-surface Kerr rotation and the ellipticity change due to the ASOT is fitted in Fig. 3d.

The only free fitting parameter is the ASOT on the top surface, τ_{T}^{ASOT} , which is assumed to be the same for all Py thicknesses under the same current density and to have equal magnitude and opposite sign as the ASOT on the bottom surface τ_{B}^{ASOT} .

The good agreement between experiment and simulation supports the assumption that ASOT depends on current density. The ASOTs are extrapolated to be $\tau_{\rm T}^{\rm ASOT} = -\tau_{\rm B}^{\rm ASOT} = (-0.86 \pm 0.04) \times 10^{-6} \text{ J/m}^2$ from the fitting*. Relating this torque to a spin current allows us to find the Spin-Hall-angle-like efficiency of the ASOT $\xi = \frac{2e\tau_{\rm B}^{\rm ASOT}}{i\text{ h}} = 0.053 \pm 0.003$, where *e* is the electron charge, $j_{\rm e}$ is the electric current

density and \hbar is the reduced Planck constant; this efficiency is comparable with the effective spin Hall angle of Pt (0.056 ± 0.005) measured in a Pt/Py bilayer⁷. The corresponding ASOT conductivity for 32 nm Py is calculated as

$$\sigma^{\text{ASOT}} = \frac{2e}{h} \frac{\tau_{\text{B}}}{E} = \xi \sigma = 2300 \pm 120 \,\Omega^{-1} \text{cm}^{-1}$$
, where *E* is the applied electric field. In

Fig. 3d, the deviation of the ASOT-induced change in Kerr ellipticity from the model for the 4 nm Py sample can be accounted for if a 1% variation between τ_{T}^{ASOT} and τ_{B}^{ASOT} is assumed, which may be due to a slight difference in spin relaxation at the two interfaces (see Supplementary Information section S6 for further discussion).

^{*} All the uncertainties in this letter are single standard deviation uncertainties. The principle source of uncertainty here is the fitting uncertainty, which is determined by a linear regression analysis by plotting the experimental data as a function of the simulation results.



Figure 3 Dependence of ASOT on current density, angle, thickness and the interface. Kerr rotation change as a function of **a**, current density and **b**, the angle between current direction and magnetization. Kerr rotation (experimental, black squares; fit, black solid line) and ellipticity change (experimental, red circles; fit, red dashed line) **c**, due to the calibration field, and **d**, due to ASOT. **e**, Comparison between total SOT conductivities (σ_{tot}^{SOT}) measured for 4 nm Py with different capping layers,

and the bottom-surface ASOT conductivity (σ^{ASOT}) of 32 nm Py. Error bars indicate single standard deviation uncertainties. In all these samples, the other side of the Py is in contact with AlO_x.

Since ASOT results in magnetization changes near the surface, the extracted ASOT values may be influenced by spin-orbit interaction at the interface with the capping layer, such as Rashba-Edelstein spin-orbit coupling²⁶⁻²⁸. To determine the relative

contribution of such interface effects, we compare the ASOT at the top surface of the $AlO_x(3)/Py(32)/AlO_x(3)$ sample with the total spin-orbit torque (SOT) in a series of control samples, $AlO_x(3)/Py(4)/Cap$, where Cap is varied among $AlO_x(3)$, $AlO_y(3)$, different oxidation time), $SiO_2(3)$, $Cu(3)/SiO_2(3)$ and $Al(3)/SiO_2(3)$. These capping layer materials are often assumed to have weak spin-orbit interaction due to their being light elements, but they will change the electrostatic properties and band structures of the top interface. The bottom surface is the same as for the 32 nm Py sample and thus any interfacial contribution from the bottom surface should have similar ASOT conductivity. Since Py is only 4 nm in these control samples (thinner than the exchange length), the magnetization uniformly responds to the total SOT, which is a sum of the

ASOTs at the top and bottom surfaces $(\tau_{T}^{ASOT} + \tau_{B}^{ASOT})$. Interfacial spin-orbit effects,

like the Rashba-Edelstein effect or interface-generated spin currents, are highly material- and structure-specific^{21,29}. For this reason, if either effect played an important role in the ASOT, we would not expect quantitatively, or even qualitatively, similar results for interfaces with substantially different characteristics. Should there be a significant interface-dependence of the ASOT, a large total SOT will be observed in some of these control samples with asymmetric interfaces. As shown in Fig. 3e, all

samples exhibit total SOT conductivities $\sigma_{\text{tot}}^{\text{SOT}} = \frac{2e}{h} (\tau_{\text{T}}^{\text{ASOT}} + \tau_{\text{B}}^{\text{ASOT}}) / E$ of at most 4%

of the bottom-surface ASOT conductivity of the 32 nm Py sample. This suggests that the top-surface ASOT, which varies less than 4% among Py with different capping layers, does not contain a substantial contribution from the interface of the Py with the capping layers.

The insensitivity of ASOT to the interface implies that it arises from the bulk spinorbit interaction within the magnetic material. ASOT can be phenomenologically understood as the result of the TSHE – a flow of transversely polarized spin current generates ASOT by transferring spin angular momentum from one surface to the other. We evaluate the TSHE conductivity using linear response in the Kubo formalism in the clean limit using density functional theory³⁰ (see Supplementary Information section S7 for technical details). First-principles calculations for Ni, Fe and Co all show significant TSHE conductivities, summarized in Table 1. We also measure the ASOT conductivities of these materials experimentally, provided in Table 1. For comparison, we also calculate and measure the AHE conductivities for these materials. If the ASOT is only due to the TSHE from the intrinsic band structure, the calculated TSHE conductivity should match the measured ASOT conductivity. As shown in Table 1, the conductivities are similar in magnitude as those calculated, indicating that the intrinsic mechanism may significantly contribute to the ASOT. However, the signs for Fe and Co are opposite between measured and calculated values; this may be because the intrinsic mechanism is not the sole source of ASOT and other mechanisms should be taken into account. By analogy with the AHE, we expect that extrinsic mechanisms such as skew scattering^{10,31} can also contribute to generating transversely polarized spin current and hence ASOT (see Supplementary Information Fig. S8).

		Ni	Fe	Со
culation	Structure	FCC	BCC	НСР
	AHE Conductivity	-1.3	0.72	0.45
Ca	TSHE Conductivity	3.92	1.05	-0.24
Experiment	Structure	FCC	BCC	НСР
	Conductivity	56	32	46
	AHE Conductivity	- 0.5± 0.05	0.5 ± 0.05	0.3 ± 0.03
	ASOT Conductivity	3.5 ± 0.1	-1.0 ± 0.2	0.8 ± 0.5

Table 1. Measured and calculated electrical, AHE and ASOT conductivities. All values have units of $10^3 \Omega^{-1}$ cm⁻¹. All experimental data are extrapolated based on 40 nm sputtered polycrystalline films, sandwiched between two 3 nm AlO_x layers. The positive sign for the ASOT conductivity corresponds to the scenario that if the applied electric field is in the *x*-direction, the generated spin current flowing in the *z*-direction has spin moment in the -*y*-direction. Under this choice, the spin Hall conductivity of Pt is positive.

The existence of ASOT may change some of the conventional understanding of spin-orbit torques in magnetic multilayers. For example, an electric current can generate a net spin-orbit torque in a SiO₂/Py/Cu/Pt multilayer acting on the Py magnetization. The net spin-orbit torque is the superposition of spin-orbit torques at the two surfaces of the Py layer. Although Pt or the Pt/Cu interface were often thought to be the source for spin-orbit torque, we find that the spin-orbit torque at the SiO₂/Py interface is much larger than that at the Py/Cu interface, as shown in Fig. 4. This is because the spin-orbit torque at the Py/Cu interface is the superposition of ASOT in Py and the external spin-orbit torque due to spin current generated from Pt, the two of which are in opposite directions. Therefore, although the total spin-orbit torque at the SiO₂/Py interface is in fact greater than that at the Py/Cu interface.

Although the total ASOT equals zero in an isolated magnetic layer with symmetric surfaces, such symmetry is likely broken when the ferromagnet is in contact with a nonmagnetic layer with strong spin-orbit coupling (see Supplementary Information section S9 for more discussion). If there is an asymmetry in the ASOT at the two surfaces of the magnetic layer, a net spin-orbit torque is expected, which contributes to the total spin-orbit torque in magnetic multilayers. This net spin-orbit torque, arising from the spin-orbit interaction of the ferromagnet itself, may have been previously overlooked.



Figure 4 (a) Illustration of the asymmetric SOTs in a SiO₂/Py(32)/Cu(3)/Pt(2)/AlO_x(3) multilayer. Measurement configurations are the same as in Figure 2. The net spin torques at the top surface $\tau_{\rm T} = \tau_{\rm T}^{\rm ASOT} + \tau_{\rm Pt/Cu}$, and at the bottom surface $\tau_{\rm B} = \tau_{\rm B}^{\rm ASOT}$

are probed by MOKE, where $\tau_{Pt/Cu}$ is the spin-orbit torque due to spin current

generated from Pt injected into Py. From the spin current shown in the figure, it can be expected that τ_T is smaller than τ_B , contrary to common understanding. (b) The measured Kerr rotation signals when light probes the top surface with calibration field h_{Cal} applied and current-driven spin-orbit torque applied. (c) The measured Kerr rotation signals when the bottom surface is interrogated with calibration field h_{Cal} applied and current-driven spin-orbit torque applied. While the Kerr rotations due to the calibration signal are similar in magnitude, those due to current-driven spin-orbit torque are much larger at the bottom surface than at the top surface.

Methods

Sample Fabrication

The samples used in this study are fabricated via magnetron sputtering. The AlO_x layers are made by depositing 2 nm Al film and subsequent oxidization in an oxygen plasma.

MOKE Measurement of ASOT

The MOKE measurements are performed with a lock-in balanced detection system²⁵, which is illustrated in Supplementary Information Fig. S3. An alternating current with frequency 20.15 kHz is applied through the patterned sample and the ASOT-induced MOKE response at the same frequency is measured. We use a Ti:sapphire mode-locked laser with \approx 100 fs pulses at 80 MHz repetition rate with center wavelength 780 nm; the detectors used are slow relative to the repetition rate, so the measured signals are averaged over the pulses. The laser beam is focused by a 10x microscope objective into a spot of ~4 µm diameter. Laser power below 4 mW is used to avoid significant heating effects. To eliminate the quadratic MOKE contribution, the

average is taken of the signals for incident laser polarizations of 45° and 135° with respect to the magnetization²⁵. A combination of a second half-wave plate and a Wollaston prism is used to analyze the Kerr rotation signal. For Kerr ellipticity measurements, a quarter-wave plate is inserted before the half-wave plate.

Fitting of the thickness-dependent MOKE signal

In the simulations, the magnetic film is discretized into many sublayers of thickness 0.4 nm. By assuming equal and opposite ASOT, $\tau_T^{ASOT} = -\tau_B^{ASOT}$, at the top and bottom of each sublayer, we calculate the resultant out-of-plane magnetization using numerical methods (see Supplementary Information section S4). For calibration, a constant out-of-plane calibration field h_{Cal} is applied to all sublayers, and the out-of-plane magnetization is calculated using the same numerical methods. Based on the calculated out-of-plane magnetization distribution, the polar MOKE response is determined using the propagation matrix method and taking into account multiple reflections (see Supplementary Information section S5). The above processes provide linear relationships between τ_T^{ASOT} and h_{Cal} with the predicted MOKE response for various film thicknesses. In modelling the thickness-dependent MOKE response for calibration, shown in Fig. 3a, all parameters are measured by other techniques. The good agreement corroborates our numerical model. In the fitting of the thickness-dependent MOKE response for a specific techniques are for all film thicknesses under the same current density. All other parameters are

the same as those used in modelling the calibration result. The good agreement shown in Fig. 3b confirms our assumption that τ_{T}^{ASOT} depends on the current density.

Acknowledgements

The work done at the University of Denver is partially supported by the PROF and by the National Science Foundation under Grant Number ECCS-1738679. W.W., D.G.C. and V.O.L. acknowledge support from the NSF-MRSEC under Award Number DMR-1720633. T.W., Y.W, and J.Q.X acknowledge support from NSF under Award Number DMR-1505192. V.P.A. acknowledges support under the Cooperative Research Agreement between the University of Maryland and the National Institute of Standards and Technology Center for Nanoscale Science and Technology, Award 70NANB14H209, through the University of Maryland. We would also like to thank Mark Stiles and Emilie Jue for critical reading of the manuscript, and Xiao Li for illuminating discussions.

Contributions

X.F. and H.O. conceived the idea; X.F., W.W., and T.W. designed the experiments; T.W. fabricated the sample, T.W., Y.W., W.W., A.R., A.D., T.J.S., D.B. and B.L.Z. patterned and characterized the samples; W.W. performed the MOKE measurements; W.W., X.F., V.O.L., D.G.C. and J.Q.X. analyzed the data; V.P.A. and P.M.H. carried out the first-principles calculations. X.F., W.W., V.O.L., V.P.A. and P.M.H. prepared the manuscript; all authors commented on the manuscript.

Competing interests

The authors declare no competing financial interest.

References

- 1 Hirsch, J. E. Spin Hall effect. *Physical Review Letters* **83**, 1834-1837, doi:10.1103/PhysRevLett.83.1834 (1999).
- 2 Kato, Y. K., Myers, R. C., Gossard, A. C. & Awschalom, D. D. Observation of the spin hall effect in semiconductors. *Science* **306**, 1910-1913, doi:10.1126/science.1105514 (2004).
- 3 Kimura, T., Otani, Y., Sato, T., Takahashi, S. & Maekawa, S. Room-temperature reversible spin Hall effect. *Physical Review Letters* **98**, doi:10.1103/PhysRevLett.98.156601 (2007).
- 4 Mihai Miron, I. *et al.* Perpendicular switching of a single ferromagnetic layer induced by in-plane current injection. *Nature* **476**, 189-U188, doi:10.1038/nature10309 (2011).
- 5 Liu, L. *et al.* Spin-Torque Switching with the Giant Spin Hall Effect of Tantalum. *Science* **336**, 555-558, doi:10.1126/science.1218197 (2012).
- Hall, E. H. On the "Rotational Coefficient" in Nickel and Cobalt. *Proc. Phys. Soc. London*4, 325 (1880).
- 7 Liu, L., Moriyama, T., Ralph, D. C. & Buhrman, R. A. Spin-Torque Ferromagnetic Resonance Induced by the Spin Hall Effect. *Physical Review Letters* **106**, doi:10.1103/PhysRevLett.106.036601 (2011).
- Ghosh, A., Auffret, S., Ebels, U. & Bailey, W. E. Penetration Depth of Transverse Spin Current in Ultrathin Ferromagnets. *Physical Review Letters* 109, doi:10.1103/PhysRevLett.109.127202 (2012).
- Zhang, S. F. Spin Hall effect in the presence of spin diffusion. *Physical Review Letters* 85, 393-396, doi:10.1103/PhysRevLett.85.393 (2000).
- 10 Nagaosa, N., Sinova, J., Onoda, S., MacDonald, A. H. & Ong, N. P. Anomalous Hall effect. *Reviews of Modern Physics* 82, 1539-1592, doi:10.1103/RevModPhys.82.1539 (2010).
- 11 Miao, B. F., Huang, S. Y., Qu, D. & Chien, C. L. Inverse Spin Hall Effect in a Ferromagnetic Metal. *Physical Review Letters* **111**, doi:10.1103/PhysRevLett.111.066602 (2013).
- 12 Taniguchi, T., Grollier, J. & Stiles, M. D. Spin-Transfer Torques Generated by the Anomalous Hall Effect and Anisotropic Magnetoresistance. *Physical Review Applied* **3**, doi:10.1103/PhysRevApplied.3.044001 (2015).

- 13 Wang, H. L., Du, C. H., Hammel, P. C. & Yang, F. Y. Spin current and inverse spin Hall effect in ferromagnetic metals probed by Y3Fe5O12-based spin pumping. *Applied Physics Letters* **104**, doi:10.1063/1.4878540 (2014).
- 14 Chernyshov, A. *et al.* Evidence for reversible control of magnetization in a ferromagnetic material by means of spin-orbit magnetic field. *Nature Physics* **5**, 656-659, doi:10.1038/nphys1362 (2009).
- 15 Ciccarelli, C. *et al.* Room-temperature spin-orbit torque in NiMnSb. *Nature Physics* **12**, 855-860, doi:10.1038/nphys3772 (2016).
- 16 Tian, D. *et al.* Manipulation of pure spin current in ferromagnetic metals independent of magnetization. *Physical Review B* **94**, doi:10.1103/PhysRevB.94.020403 (2016).
- 17 Das, K. S., Schoemaker, W. Y., van Wees, B. J. & Vera-Marun, I. J. Spin injection and detection via the anomalous spin Hall effect of a ferromagnetic metal. *Physical Review B* **96**, doi:10.1103/PhysRevB.96.220408 (2017).
- 18 Humphries, A. M. *et al.* Observation of spin-orbit effects with spin rotation symmetry. *Nature Communications* **8**, doi:10.1038/s41467-017-00967-w (2017).
- 19 Baek, S. H. C. *et al.* Spin currents and spin-orbit torques in ferromagnetic trilayers. *Nature Materials* **17**, 509-513, doi:10.1038/s41563-018-0041-5 (2018).
- 20 Ralph, D. C. & Stiles, M. D. Spin transfer torques. *Journal of Magnetism and Magnetic Materials* **320**, 1190-1216, doi:10.1016/j.jmmm.2007.12.019 (2008).
- 21 Amin, V. P., Zemen, J. & Stiles, M. D. Interface-Generated Spin Currents. *Physical Review Letters* **121**, doi:10.1103/PhysRevLett.121.136805 (2018).
- 22 Pauyac, C. O., Chshiev, M., Manchon, A. & Nikolaev, S. A. Spin Hall and Spin Swapping Torques in Diffusive Ferromagnets. *Physical Review Letters* **120**, doi:10.1103/PhysRevLett.120.176802 (2018).
- 23 Fan, X. *et al.* Quantifying interface and bulk contributions to spin-orbit torque in magnetic bilayers. *Nature Communications* **5**, doi:10.1038/ncomms4042 (2014).
- 24 Qiu, Z. Q. & Bader, S. D. Surface magneto-optic Kerr effect. *Review of Scientific Instruments* **71**, 1243-1255, doi:10.1063/1.1150496 (2000).
- Fan, X. *et al.* All-optical vector measurement of spin-orbit-induced torques using both polar and quadratic magneto-optic Kerr effects. *Applied Physics Letters* **109** (2016).
- 26 Pesin, D. A. & MacDonald, A. H. Quantum kinetic theory of current-induced torques in Rashba ferromagnets. *Physical Review B* **86**, doi:10.1103/PhysRevB.86.014416 (2012).
- 27 Kurebayashi, H. *et al*. An antidamping spin-orbit torque originating from the Berry curvature. *Nature Nanotechnology* **9**, 211-217, doi:10.1038/nnano.2014.15 (2014).
- 28 Baek, S.-h. C. *et al.* Spin currents and spin–orbit torques in ferromagnetic trilayers. *Nature Materials*, doi:10.1038/s41563-018-0041-5 (2018).
- 29 Park, S. R., Kim, C. H., Yu, J., Han, J. H. & Kim, C. Orbital-Angular-Momentum Based Origin of Rashba-Type Surface Band Splitting. *Physical Review Letters* **107**, 156803, doi:10.1103/PhysRevLett.107.156803 (2011).
- Wang, X. J., Yates, J. R., Souza, I. & Vanderbilt, D. Ab initio calculation of the anomalous
 Hall conductivity by Wannier interpolation. *Physical Review B* 74, doi:10.1103/PhysRevB.74.195118 (2006).

31 Hoffmann, A. Spin Hall Effects in Metals. *leee Transactions on Magnetics* **49**, 5172-5193, doi:10.1109/tmag.2013.2262947 (2013).

Anomalous Spin-Orbit Torques in Magnetic Single-Layer Films

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Supplementary Note 1 Symmetry Argument

Curie's principle [1] states that the symmetry of an effect must follow the symmetry of the cause. This heuristic argument provides guidance in searching for new physical phenomena. Here we show how symmetry leads to the types of surface spin and charge accumulation observed in the spin Hall effect, anomalous Hall effect and anomalous spin-orbit torque.

In polycrystalline, amorphous or crystalline thin films with high symmetry, the structure of the thin film is invariant under several symmetry operations, including (1) mirror symmetry operations: σ_{xy} , σ_{xz} , and σ_{yz} , where the subscript denotes the mirror plane, (2) two-fold rotational symmetry operations: C_2^x , C_2^y and C_2^z , where the superscript denotes the rotational axis, and (3) center inversion symmetry: *i*, where the inversion center is any point of the material.

When an electric field is applied through a nonmagnetic thin film along the x-direction, as shown in Supplementary Fig. 1(a), mirror symmetry σ_{yz} , rotational symmetries C_2^y and C_2^z , and inversion symmetry *i* are broken. However, the system still retains the symmetries σ_{xy} , σ_{xz} , and C_2^x . Therefore, on the top and bottom surfaces in the xy plane, there cannot be any net charge accumulation, which would violate symmetries σ_{xy} and C_2^x . However, spin accumulation with a certain spin polarization is allowed by symmetry. Spin polarizations in the x- and z-directions are forbidden, as both violate symmetry σ_{xz} . Spin polarization in the y-direction is allowed, given that the spins are polarized in opposite directions on the top and bottom surfaces to satisfy the symmetries σ_{xy} and C_2^x . These symmetry conditions characterize what is known as the spin Hall effect in nonmagnetic materials.

In a magnetic film, where the existence of magnetization breaks more symmetries, more complicated surface phenomena are expected. As shown in Supplementary Fig. 1(b), the magnetization along the y-direction breaks the mirror symmetry σ_{xy} , σ_{yz} and rotational symmetries C_2^x and C_2^z , leaving the magnetic film only carrying the mirror symmetry σ_{xz} . As a result, charge accumulation as well as spin accumulation with spins in the y-direction on the top and bottom surfaces are allowed by symmetry; these symmetry conditions characterize the anomalous Hall effect. On the other hand, in a magnetic film with magnetization along the x-direction, as shown in Supplementary Fig. 1(c), the system only carries the C_2^x rotational symmetry and breaks all mirror symmetries. Under such symmetry, spin polarizations in both the y- and z- directions are allowed on the top



Supplementary Figure 1: Symmetry-based analysis of the electric-field-induced charge/spin accumulations at the top and bottom surfaces of (a) a nonmagnetic film, (b) a magnetic film with magnetization \mathbf{m} along the y-direction and (c) a magnetic film with magnetization \mathbf{m} along the x-direction.

and bottom surfaces. Due to spin dephasing, the spin accumulations give rise to spin torques in the z- and y- directions, which obey the same symmetry constraints. The symmetry conditions for the case of z-polarization characterizes damping-like anomalous spin-orbit torque, which is the focus of this work. The symmetry condition for y-polarization characterizes a field-like torque, the effect of which is very similar to that of a current-induced Oersted field. It is challenging to distinguish the field-like torque and the Oersted field, the latter of which is likely to dominate the former. Therefore, the field-like torque is not studied in this paper.

The spin flow in Supplementary Fig. 1(b-c) following the spin Hall symmetry can be attributed to two types of spin Hall effects in a magnetic conductor. As illustrated in Supplementary Fig. 2(a-b), these two spin Hall effects are the longitudinal spin Hall effect with spin direction parallel with the magnetization, and the transverse spin Hall effect with spin direction perpendicular to the magnetization. Even though both longitudinal and transverse spin Hall effects result from spin-orbit coupling, we expect the two to have different characteristics. For example, longitudinal spins undergo spin diffusion in a magnetic conductor, while dissipation of transverse spins is governed by both spin diffusion and spin dephasing.

Besides the flow of spin current, which is a common result of the spin Hall effect in magnetic and nonmagnetic materials, the longitudinal and transverse spin Hall effects have their respectively unique manifestations in magnetic materials. The longitudinal spin Hall effect in a magnetic conductor results in the anomalous Hall effect, which is a net charge accumulation at the surfaces. The transverse spin Hall effect, on the other hand, results in an accumulation of anomalous spin-orbit torques at the corresponding surfaces, in a magnetic conductor, as illustrated in Supplementary Fig. 2(c-d). This phenomenological analogy to the anomalous Hall effect is why we use the term "anomalous" to describe the observed anomalous spin-orbit torque.



Supplementary Figure 2: (a) Longitudinal and transverse spin Hall effects (SHE) in a magnetic conductor when magnetization is perpendicular to the applied electric field. Here the red arrows represent spin directions and light vellow arrows represent spin flow directions. (b) When the magnetization is parallel with the electric field, only the transverse spin Hall effect takes place. (c) In the configuration of (a), the longitudinal spin Hall effect leads to the anomalous Hall effect (AHE), while the transverse spin Hall effect leads to anomalous spin-orbit torques (ASOT). Here the red and blue circles represent positive and negative charge accumulations, while the green and blue arrows represent the directions of anomalous spin-orbit torques. (d) In the configuration of (b), the transverse spin Hall effect leads to anomalous spin-orbit torques on all four sides. Arrows near the four sides are all anomalous spin-orbit torque directions.

Supplementary Note 2 Material Characterization

The material parameters for the samples measured in this work are summarized in Table 1.

The saturation magnetizations of the Py films are measured by a superconducting quantum interference device (SQUID). The surface anisotropy is extrapolated from a thickness-dependent ferromagnetic resonance measurement [2]. The total surface anisotropy energy for Py is extrapolated to be about 9.3×10^{-4} J/m², which is the sum of the surface anisotropy energies at the top and bottom surfaces. The exchange constant is extrapolated from a first-order standing spin wave measurement [3]. The electric conductivities of the Py films are determined by measuring the four-probe resistance. We use ellipsometry to determine the index of refraction of all relevant films, where the films are grown to at least 80 nm on a silicon wafer with 1 μ m thermal oxide. The optical penetration depths in nanometers at 780 nm wavelength are calculated with the formula $d_p = \frac{780}{4\pi k}$, where k is the imaginary part of the refractive index. Supplementary Figure 3 shows the x-ray diffraction (XRD) patterns for the sputtered 40 nm Ni,

Supplementary Table 1: Material parameters of the samples studied in this work: saturation magnetization M_s , electric conductivity σ , exchange length λ_{ex} , index of refraction n, and optical penetration depth at 780 nm wavelength d_p . Numbers in parentheses are the thicknesses in nm.

	$\mu_0 M_s(\mathbf{T})$	$\sigma\left(\Omega^{-1}\mathrm{cm}^{-1}\right)$	λ_{ex} (nm)	n	d_p (nm)
Py(32)	1.09	3.34×10^4	5.1	2.38+4.36i	14.24
Ni(40)	0.54	4.42×10^4	8.4	2.10+4.30i	14.47
Co(40)	2.0	$4.58 imes 10^4$	3.9	2.39+4.45i	13.98
Fe(40)	2.24	3.14×10^4	3.1	2.69+3.68i	16.91

40 nm Fe and 40 nm Co films used in the MOKE-SOT measurements. Supplementary Figure 3(a) depicts the XRD pattern of the 40 nm Ni sample, showing formation of face-centered cubic (FCC) structures, as determined by comparing with the Inorganic Crystal Structure Database (ICSD). The 40 nm Fe sample represents a body centered cubic (BCC) structure, and the 40 nm Co film has a dominant hexagonal close-packed (hcp) phase, which has a strong texture with c-axis out of the film plane.



Supplementary Figure 3: Bragg peaks and corresponding diffraction planes from the Inorganic Crystal Structure Database (ICSD) (top) and x-ray diffraction patterns from experiment (bottom) for the sputtered (a) 40 nm FCC Ni (ICSD #53807), (b) 40 nm BCC Fe (ICSD #64795) and (c) 40 nm HCP Co films (ICSD #53806).

Supplementary Note 3 MOKE Measurement Setup

As discussed in the Methods section, we perform our MOKE measurements with a lock-in balanced detection system. An illustration of the experimental setup is shown in Supplementary Fig. 4. The spot size is $\sim 4 \ \mu$ m in diameter and the power of the laser is less than 4 mW. We carried out the same measurement using a larger laser spot size of about 15 μ m diameter, and did not see any difference (Supplementary Fig. 5 (a)). We also scanned the laser spot onto different regions of the 50 μ m × 50 μ m sample, and saw no difference in the ASOT signal, except for an overall offset of the curve due to the current-induced out-of-plane Oersted field, h_{Oe}^{out} (see Supplementary Fig. 5b). [8].



Supplementary Figure 4: Schematic diagram of the lock-in balanced detection system. HWP: half-wave plate; QWP: quarter-wave plate.

Supplementary Note 4 ASOT-induced Magnetization Distribution

A phenomenological model of the ASOT is shown in Supplementary Fig. 6. In a magnetic material with bulk inversion symmetry, the SOI gives rise to a separation of opposite spins. The spins are transverse to the magnetization and thus will be quickly absorbed. The absorption of transverse spins should yield no net spin torque in the bulk due to the bulk inversion symmetry. However, at the top and bottom surfaces, where inversion symmetry is broken, we expect non-zero and opposite damping-like ASOTs that tilt the magnetization out of plane.

Assuming the ASOT is a purely interfacial spin torque and neglecting surface anisotropy, it is convenient to discretize the FM into a multilayer system (shown in Supplementary Fig. 7), with the layers labeled 1 through n. Each layer has a magnetization nearly aligned in the x-direction by H_{ext} , but with slight difference in the z-component magnetization due to the ASOT. The influence of ASOT can be written as an effective field h_z^i in the z-direction for the *i*th layer. Within such a



Supplementary Figure 5: (a) Kerr rotation results of a 32 nm Py film detected with a 4 μ m (red circles) and 15 μ m (black squares) spot size. (Offset is due to the non-zero out-of-plane Oersted field) (b) Kerr rotation results of a 32 nm Py film measured with laser spot on the left (blue triangles), center (black squares) and right (red circles) of the sample, as illustrated in the inset. Besides the ASOT, the current generates an out-of-plane Oersted field that is spatially asymmetric. The effect of this Oersted field on the magnetization tilt is the same as that of the out-of-plane calibration field, as described by Eq. (2) in the main text. It results in an overall shift of the MOKE signal. The signal corresponding to the ASOT is the step-like signal at low fields, due to the magnetizations switching. The ASOT-related signals are within 5% difference for the three positions. The current density used in all measurements is $5 \times 10^{10} \text{ A/m}^2$.

system, the ASOTs only affect the two surface layers:

$$h_{z}^{1} = \frac{\tau_{\rm T}^{\rm ASOT}}{\mu_{0}M_{s}a}m_{x}, h_{z}^{n} = \frac{\tau_{\rm B}^{\rm ASOT}}{\mu_{0}M_{s}a}m_{x},$$
(S1)

and

$$h_z^2 = h_z^3 = \dots = h_z^{n-1} = 0, (S2)$$

where m_x is the projection of unit magnetization vector in the x-direction, which takes +1 or -1 in our experiment depending on the direction of H_{ext} , τ_T^{ASOT} (τ_B^{ASOT}) is the ASOT at the top (bottom) surface, a is the lattice constant, and M_s is the saturation magnetization.

In addition to ASOT, interlayer exchange coupling significantly affects the behavior of the local magnetization. The interlayer exchange energy per unit area is described as

$$E = -J\mathbf{m}^i \cdot \mathbf{m}^{i+1},\tag{S3}$$

where \mathbf{m}^i and $\mathbf{m}^{(i+1)}$ are the unit magnetization vectors of nearest neighbor FM layers *i* and *i* + 1, and *J* is the interface exchange strength. For a uniform magnetic layer, *J* can be calculated as $J = \frac{2A_{\text{ex}}}{a}$, where A_{ex} is the exchange stiffness. Therefore, for magnetic layer *i*, the exchange coupling results in an effective field:

$$\mathbf{H}_{i} = H_{z}^{i} \mathbf{z} + H_{y}^{i} \mathbf{y} = \frac{J(m_{z}^{i-1} + m_{z}^{i+1})}{\mu_{0} M_{s} a} \mathbf{z} + \frac{2J}{\mu_{0} M_{s} a} \mathbf{y},$$
(S4)



Supplementary Figure 6: Phenomenological model of the ASOT. The spin-orbit interaction from the lattice separates electrons with opposite spins. The spin angular momentum is quickly absorbed by the magnetization. Due to bulk inversion symmetry, the only net angular momentum absorption occurs at the surface, leading to ASOT.

where m_z^i is the z-component of \mathbf{m}^i .

All relevant magnetic fields on magnetic atomic layer i are depicted in Supplementary Fig. 7. From the diagram, we can write

$$\frac{h_z^i + H_z^i - M_{\text{eff}} \sin \theta}{H_{\text{ext}} + H_y^i} = \sin \theta.$$
(S5)

For the middle layers (1 < i < n), since $h_z^i = 0$ and $\sin \theta = m_z^i$, by plugging Eq. S4 into Eq. S5, we have

$$m_{z}^{i} = \frac{\frac{J}{\mu_{0}M_{s}a}(m_{z}^{i-1} + m_{z}^{i+1}) - M_{\text{eff}}m_{z}^{i}}{H_{\text{ext}} + \frac{2J}{\mu_{0}M_{s}a}}.$$
(S6)

Knowing a is small and

$$f''(x) = \lim_{h \to 0} \frac{f(x+h) - 2f(x) + f(x-h)}{h^2},$$

Equation S6 can be rewritten as

$$\frac{Ja}{\mu_0 M_s} (m_z^i)'' = (H_{\text{ext}} + M_{\text{eff}}) m_z^i.$$
(S7)

The solution of Eq. S7 should have the form

$$m_z^i = Ae^{-z/\lambda} + Be^{+z/\lambda},\tag{S8}$$

where $\lambda = \sqrt{\frac{Ja}{\mu_0 M_s(H_{\text{ext}} + M_{\text{eff}})}}$ is the exchange length.



Supplementary Figure 7: Illustration of the discrete-layer model and effective magnetic fields applied on the layer *i*. The ASOT is assumed to only exist in the top and bottom layers (blue layers). h_z^i is the effective field due to ASOT, H_z^i and H_y^i are the effective fields from nearest-neighbor exchange coupling, H_{ext} is the external magnetic field, and $M_{\text{eff}} = (M_s - 2K_a/(\mu_0 M_s))$ is the effective field caused by demagnetization, where $K_a = 0$ for i = 2, 3, ..., n - 1, is the surface anisotropy energy density.

On the other hand, for the top layer, we can rewrite Eq. S5 as

$$m_z^1 = \frac{h_z^1 + \frac{J}{\mu_0 M_s a} m_z^2 - M_{\text{eff}} m_z^1}{H_{\text{ext}} + \frac{J}{\mu_0 M_s a}}.$$
(S9)

Eq. S9 can again be rewritten to

$$(H_{\text{ext}} + M_{\text{eff}})m_z^1 + \frac{J}{\mu_0 M_s a}(m_z^1 - m_z^2) = h_z^1.$$
 (S10)

Since a is small, the first term in Eq. S10 is negligible. Therefore, the equation can be simplified into

$$(m_z^1)' = -\frac{\tau_{\rm T}^{\rm ASOT}}{Ja}m_x.$$
(S11)

Similarly, the bottom layer magnetization yields

$$(m_z^n)' = -\frac{\tau_{\rm B}^{\rm ASOT}}{Ja}m_x.$$
(S12)

Using Eq. S11 and S12 as boundary conditions to solve Eq. S8, we get

$$A = \frac{\lambda}{Ja} \frac{\tau_{\rm T}^{\rm ASOT} - \tau_{\rm B}^{\rm ASOT} e^{-d/\lambda}}{1 - e^{-2d/\lambda}} m_x, \tag{S13}$$

and

$$B = \frac{\lambda}{Ja} \frac{\tau_{\rm T}^{\rm ASOT} - \tau_{\rm B}^{\rm ASOT} e^{d/\lambda}}{e^{2d/\lambda} - 1} m_x.$$
 (S14)

Here, d is the thickness of the magnetic film.

Plugging A and B back into Eq. S8, we get an analytic solution of the ASOT-induced magnetization tilt as shown in Eq. (1) of the main text,

$$m_z^{\text{ASOT}}(z) = \frac{\tau_{\text{T}}^{\text{ASOT}} \cosh \frac{d-z}{\lambda} + \tau_{\text{B}}^{\text{ASOT}} \cosh \frac{z}{\lambda}}{\lambda \sinh \frac{d}{\lambda} (|H_{\text{ext}}| + M_{\text{eff}}) \mu_0 M_s} m_x.$$
(S15)

Because of the strong exchange interaction, the effects from the two ASOTs will cancel out if the magnetic film is thin. However, when the magnetic film is much thicker than the exchange length, the magnetizations at the two surfaces will tilt out of plane in response to the respective ASOTs. A simulation curve based on Eq. S15 is shown in Fig. 1(c).

A more general model involving the surface anisotropy field acting on the two surface layers can be numerically calculated. By inserting Eqs. S1, S2 and S4 into S5, we get:

$$\frac{\tau_{\text{ASOT}}^{i}}{\mu_0 M_s a} m_x = (H_{\text{ext}} + M_{\text{eff}} + \frac{2J}{\mu_0 M_s a}) m_z^{i} - \frac{J}{\mu_0 M_s a} m_z^{i-1} - \frac{J}{\mu_0 M_s a} m_z^{i+1}.$$
 (S16)

The ASOT-induced magnetization tilt can then be calculated by solving the following equation:

$$\begin{bmatrix} J+C' & -J & 0 & \dots & 0 \\ -J & 2J+C & -J & \dots & 0 \\ 0 & -J & 2J+C & \dots & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & 0 & \dots & J+C' \end{bmatrix} \begin{bmatrix} m_z^1 \\ \vdots \\ m_z^i \\ \vdots \\ m_z^n \end{bmatrix} = \begin{bmatrix} \tau_T^{ASOT} \\ \vdots \\ 0 \\ \vdots \\ \tau_B^{ASOT} \end{bmatrix} m_x, \quad (S17)$$

where the surface term $C' = \mu_0 M_s a (H_{\text{ext}} + M_{\text{eff}})$ takes into account the surface anisotropy, and $C = \mu_0 M_s a (H_{\text{ext}} + M_s)$ is for the bulk layers. For 32 nm Py used in our study, the total surface anisotropy is measured to be $9.3 \times 10^{-4} \text{ J/m}^2$. We compare the numerical simulation of magnetization tilting with and without considering the surface anisotropy, as shown in Supplementary Fig. 8(a). The numerical result considering the surface anisotropy maintains the signature of the analytic result without considering the surface anisotropy, but with a small correction in the magnitude. The analysis and fitting in the main text is based on the numerical simulation taking into consideration the surface anisotropy of Py.

In the above model, we use an assumption that the ASOT is only applied to the surface layers, due to strong spin dephasing. Here we investigate if this assumption is reasonable by studying the magnetization tilt when the ASOT is distributed through more layers. As shown in Supplementary Fig. 8(b), spreading the ASOT across 5 surface layers does not affect the simulation result significantly. This is because exchange coupling will redistribute the effect of the ASOT over approximately a thickness of the exchange length. As long as the exchange length (5.1 nm for Py) is longer than the distribution length of the ASOT, the distribution of magnetization tilt will simply be dominated by the exchange length. Therefore, our model assuming that the ASOTs only exist at the top and bottom layers is reasonable.



Supplementary Figure 8: (a) Simulated distribution of the out-of-plane magnetization tilt $m_z^{\text{ASOT}}(z)$ in a 32 nm Py film based on the numerical calculation (black solid line) and the analytic expression (red dashed line). (b) Simulation results driven by equal and opposite ASOTs on the first surface layer (red dashed line) and first five surface layers (black solid line), scaled by the maximum value.

Supplementary Note 5 MOKE Response

In the structural model of Supplementary Note 4, a single FM layer is treated as a series of ultrathin magnetic layers with different magnetization orientations. For magnetic layers with thicknesses less than the coherence length of the incident laser, where multiple reflections should be taken into account, we use medium boundary matrices and medium propagation matrices to treat the multiple reflections. Based on Ref. [4], in the polar MOKE geometry, medium boundary matrix A_j for the *j*th layer can be expressed as:

$$A_{j} = \begin{bmatrix} 1 & 0 & 1 & 0 \\ 0 & 1 & 0 & -1 \\ -\frac{in_{j}Q_{j}m_{z}^{j}}{2} & -n_{j} & \frac{in_{j}Q_{j}m_{z}^{j}}{2} & -n_{j} \\ n_{j} & \frac{in_{j}Q_{j}m_{z}^{j}}{2} & -n_{j} & \frac{in_{j}Q_{j}m_{z}^{j}}{2} \end{bmatrix},$$
(S18)

where n_j is the complex refractive index of the *j*th layer, Q_j is the Voigt coefficient of the *j*th layer, and m_z^j is the out-of-plane magnetization magnitude of the *j*th layer, which is solved for in the previous section.

The propagation matrix D_j can be written as:

$$D_{j} = \begin{bmatrix} U\cos\delta^{i} & U\sin\delta^{i} & 0 & 0\\ -U\sin\delta^{i} & U\cos\delta^{i} & 0 & 0\\ 0 & 0 & U^{-1}\cos\delta^{r} & U^{-1}\sin\delta^{r}\\ 0 & 0 & -U^{-1}\sin\delta^{r} & U^{-1}\cos\delta^{r} \end{bmatrix},$$
 (S19)

where

$$U = exp(-i\frac{2\pi}{\lambda}n_jd_j),$$

$$\delta_i = -\frac{\pi n_j Q d_j}{\lambda} m_z^j,$$

and

$$\delta_r = -\frac{\pi n_j Q d_j}{\lambda} m_z^j,$$

where d_j is the thickness of the *j*th layer and λ is the wavelength of the probe light.

To obtain the magneto-optical Fresnel reflection matrix $\begin{bmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{bmatrix}$, one computes the matrix M defined by

$$M = A_0^{-1} A_1 D_1 A_1^{-1} A_2 \dots$$
 (S20)

Then the 4×4 matrix M can be written in the form of a 2×2 block matrix as follows:

$$M = \begin{bmatrix} G & H \\ I & J \end{bmatrix},\tag{S21}$$

and the magneto-optical Fresnel reflection coefficients are further solved from

$$\begin{bmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{bmatrix} = IG^{-1}.$$
 (S22)

The complex Kerr angles for s- and p-polarized incident light are:

$$\Theta_k^p = \frac{r_{sp}}{r_{pp}}$$

and
$$\Theta_k^s = \frac{r_{ps}}{r_{ss}},$$

(S23)

respectively. Here the real part of the Kerr angle is defined as the Kerr rotation, and the imaginary part of the Kerr angle is the Kerr ellipticity. Since Kerr angle is independent of the incident polarization in polar MOKE, we use the *s*-polarization result in our simulation. Following the model described above, the MOKE response for different materials and structures can be simulated.

Specifically, for the calibration field thickness dependence measurements, the uniform out-ofplane calibration field causes a uniform magnetization tilt across the whole magnetic layer. Therefore it is reasonable to treat the entire magnetic film as one layer in the simulation. A structure of $[Air(\infty)/Py(t)/SiO_2(1000)/Si(\infty)]$ is used for the simulation, where the numbers in parentheses are in nanometers. The thin SiO₂ and AlO_x layers are ignored because of their low absorption coefficients at 780 nm. A few nanometers of dielectric material do not affect the MOKE response significantly. With the calculated out-of-plane magnetization tilt and refractive indices measured via ellipsometry, the resulting simulation curve is in good agreement with the experimental data, shown in Fig. 3(a).

For the ASOT measurements, the same model structure and fitting parameters are used except for including the ASOT strength $\tau_T^{ASOT} = -\tau_B^{ASOT}$ and the resulting non-uniform magnetization distribution. By alternating the value of τ_T^{ASOT} in our model, we are able to match the measured MOKE signals, as shown in Fig. 3(b). The extracted τ_T^{ASOT} is then used to calculate the spin-torque efficiency.

Supplementary Note 6 Kerr Ellipticity Signal of 4 nm Py

In the discussion of how the ASOT-induced MOKE signal changes as a function of Py thickness, shown in Fig. 3(b), we point out that the Kerr ellipticity signal of 4 nm Py deviates from the fitting curve. The deviation can be accounted for if we assume there exists a 1% difference between τ_T^{ASOT} and τ_B^{ASOT} , which is within the error and may arise due to slight imbalance of interfacial spin dissipation. In Supplementary Fig. 9, we show the modified fitting curves that assume a 1% ASOT difference. The modified model only changes the fitting curves at small thicknesses (< 8 nm), and the modified Kerr ellipticity curve now fits well with the data point at 4 nm. Furthermore, as demonstrated in Supplementary Fig. 9c, even a tiny (1%) total SOT can cause a MOKE signal at small thicknesses that is comparable in amplitude to the MOKE signal due to a much larger surface ASOT at large thicknesses. The large variation of the simulated MOKE signals when the Py is thin are due to the interference of the laser as it is reflected from the various interfaces; this effect also appears at small sample thicknesses in the MOKE signal from the calibration field (Fig. 3a).



Supplementary Figure 9: Experimental MOKE rotation and ellipticity signals (squares and circles) and simulated rotation and ellipticity signals (solid black lines and dashed red lines) for (a) unbalanced ($\tau_B = -1.01\tau_T^0$) ASOT, (b) balanced ($\tau_B = -\tau_T^0$) ASOT and (c) a tiny bottom (total) torque ($\tau_T = 0, \tau_B = -0.01\tau_T^0$) as a function of Py thickness. The fitting curves in (a) are a combination of the curves in (b) and (c), as described in this section.

Supplementary Note 7 Contribution from Interfacial Spin-Orbit Interaction

Generally speaking, there are two types of interfacial spin-orbit effects that can generate spin-orbit torque: the Rashba spin-orbit coupling [10] and the interface-generated spin current [11]. Both of these effects are sensitive to the materials forming the interface.

The Rashba model results in spin accumulation as an electric current passes near an interface due to the Edelstein effect. The strength of the Rashba-like spin-orbit coupling is the result of two distinct physical considerations: 1. the intrinsic atomic spin-orbit coupling, which is controlled by the Z-value of the material and 2. the degree of inversion symmetry breaking. The symmetry breaking is generically large at the interface between dissimilar materials, but its precise magnitude (and sign) is sensitive to details. For example, the presence of strongly electronegative ions near the interface (e.g. O) results in a more asymmetric charge distribution, which is directly related to the magnitude of the Rashba coefficient [12]. As an example, Ref. [13] uses angle-resolved photoemission spectroscopy and density functional theory calculations to show that the sign of the Rashba coefficient at a Gd(0001) surface changes sign upon oxidation.

Moreover, the sign and size of the effective Rashba coefficient at an interface is very sensitive to details of the electronic structure [14], and therefore quite sensitive to the interface structure. It has been shown that field-like spin-orbit torque in a Pt/Co bilayer rapidly decreases with alloying of the Pt-Co interface [15]. Using density functional theory calculations, Belashchenko *et al.* [16] showed the field-like torque in a Pt/Co bilayer decreases by a factor of 5 with increasing disorder. Haney et al. [17] used density functional theory calculations to show that the value of the field-like torque is quite sensitive to interface structure. These considerations lead to the conclusion that the Rashba spin-orbit coupling parameter at interfaces is highly material- and structure-specific.

The underlying idea behind interface-generated spin currents is that electrons scattering off a ferromagnet/nonmagnet interface will interact with interfacial spin-orbit fields, and this interaction generates spin currents flowing out of plane. The interfacial spin-orbit fields, which can be treated as effective magnetic fields, can filter the electrons based on their spin direction or can induce spin precession. Interfacial spin filtering and precession can lead to a spin current with spin polarization transverse to the magnetization, and experimental evidence suggests that this spin current can have opposite sign when generated at different material interfaces [18]. Interface-generated spin currents, which may apply a spin torque on the magnetization, sensitively depend on the electrical conductivity of both materials [19, 20] as well as details of the electronic structure [11]. Therefore, there is no *a priori* reason to assume that interface-generated spin currents, if relevant to ferromagnet/oxide interfaces, would be insensitive to the substitution of various oxide layers.

Therefore, if interfacial spin-orbit coupling played an important role in our experiments, we would expect different results for interfaces with substantially different characteristics.

Supplementary Note 8 First Principles Calculations

We compute the full spin-current conductivity tensor $\sigma_{\alpha\beta}^{\gamma}$ using the linear response Kubo formalism in the clean limit:

$$\sigma_{\alpha\beta}^{\gamma} = 2\mathrm{Im}\frac{e^2}{\hbar} \int \frac{dk}{(2\pi)^3} \sum_{n \neq m} f_{n,k} \frac{\langle \psi_n | Q_{\alpha\beta} | \psi_m \rangle \langle \psi_n | \psi_\gamma | \psi_n \rangle}{(E_n - E_m)^2}, \tag{S24}$$

where $f_{n,k}$ is the Fermi factor, v_{γ} is the velocity operator along the γ -direction: $v_{\gamma} = dH/dk_{\gamma}$, and $Q_{\alpha\beta}$ is the spin current corresponding to the α -component spin flowing in the β -direction. Its operator form is $Q_{\alpha\beta} = (v_{\alpha}s_{\beta} + s_{\alpha}v_{\beta})/2$, where s_{β} is the β -component of the Pauli spin matrices. The above expression is evaluated within density functional theory. The ground state is computed with the Quantum Espresso package [5], where we use the experimental lattice constants of (0.286, 0.352, 0.2507) nm for Fe (BCC), Ni(FCC), Co(HCP with c-axis perpendicular to the film plane), respectively. In each case, the plane-wave cutoff energy is set to 120 Ryd, and a 12 × 12 × 12 uniform k-point grid is used. We use ultrasoft, fully relativistic pseudopotentials with GGA functional. For Ni, we use the GGA+U method as described in Ref. [7], with U = 1.9 eV and J = 1.2eV. To evaluate Eq. (1) on a fine k-point mesh, we perform Wannier interpolation using Wannier90 [6]. The integral is evaluated with 200³ k-points, and we use an adaptive mesh technique in which k-points with integrand larger than 0.28 nm² are evaluated on a refined grid. The reported values are numerically converged to within 1%. The magnetization is along the x-direction, consistent with Fig. 1b.

Supplementary Note 9 Spin Torque Dipole

Skew scattering can give rise to ASOT, illustrated in Supplementary Fig. 10.



Supplementary Figure 10: (a) Illustration of a skew scattering-induced spin torque dipole. As x-polarized electrons are scattered by an impurity, the spin orbit interaction (SOI) generates an effective magnetic field $B_{\text{eff}} \propto E_{\text{scatter}} \times v$, where E_{scatter} is the electric field due to the impurity and v is the velocity of the electron. Depending on the scattering trajectory, B_{eff} has opposite directions above and below the scattering center, which rotates the electron spin toward the +y and -y directions. The electron spin is soon repolarized to the x-direction due to dephasing. The additional spin angular momentum gained via the SOI is transferred into the magnetization, which leads to an effective spin torque dipole separated by a distance on the order of the spin dephasing length. (b) A collection of spin torque dipoles in a uniform magnetic film gives rise to equal and opposite ASOTs at the two surfaces, shown in Fig. 1c.

Supplementary Note 10 ASOT in Ferromagnetic Metal (FM)/Nonmagnetic Metal (NM) Bilayers

The ASOT is equal and opposite in a single-layer ferromagnet because the transversely polarized spin current that transfers spin angular momentum to the two surfaces is spatially symmetric; i.e., the spin current decays to zero at both surfaces at the same rate. However, if the ferromagnet is in contact with a nonmagnetic layer to form a FM/NM bilayer, and that NM layer exhibits strong spin-orbit scattering, the symmetry of the spin current at the two surfaces of the FM is likely broken. As a result, an asymmetry in the magnitude of the ASOT at the two surfaces may arise. This can also be understood from an argument based on angular momentum conservation. The ASOT can be interpreted as a result of spin angular momentum transferred from the spin current into the magnetization system. But if the NM creates an additional spin scattering channel, a portion of spin angular momentum will be lost to the lattice via spin-orbit scattering near the FM/NM interface. Thus, the ASOT at the FM/NM interface is likely to be smaller than the ASOT at the other FM surface.

Note that the discussion above does not contradict the data presented in Fig. 3e, where the NM only contains light elements with weak spin-orbit scattering. But if the NM contains heavy elements that exhibit strong spin-orbit scattering, a net spin-orbit torque arising from the asymmetry in the ASOT is expected. This net ASOT will have the same symmetry as the conventional spin-orbit torque in the FM/NM bilayers, but with different origins. Conventionally, the spin-orbit torque in FM/NM bilayers is considered to arise from spin current generated from the spin Hall effect of the

NM or interfacial spin-orbit coupling of the FM/NM interface, and the FM is only a receptor of the spin current. But the net ASOT arises from the spin current generated from the FM bulk, with the NM serving as a spin current absorber.

A net ASOT is likely to be present in all previously studied FM/NM bilayers or multilayers when the FM is a conductor. From the discussion above, the net ASOT is only a fraction of the ASOT on each surface. But because the ASOT in ferromagnetic conductors are very large, the net ASOT should be taken into consideration when studying the spin-orbit torque in FM/NM bilayers. We expect the net ASOT to be closely related to the interface spin scattering rate, which will be investigated in our future work.

References

- [1] Chalmers, A. F. Curie's Principle. *The British Journal for the Philosophy of Science* **21**, 133-148 (1970).
- [2] Johnson, M. T., Bloemen, P. J. H., Den Broeder, F. J. A., De Vries, J. J. Magnetic anisotropy in metallic multilayers. *Reports on Progress in Physics* 59, 1409–1458 (1996).
- [3] Samantaray B., *et al.*. Perpendicular standing spin wave and magnetic anisotropy study on amorphous FeTaC films. *IEEE Transactions on Magnetics* **52**, 2003104 (2016).
- [4] You, C.-Y, and Shin, S.-C. Generalized analytic formulae for magneto-optical Kerr effects. *Journal of Applied Physics* **84**, 541 (1998).
- [5] Giannozzi, P., et al. QUANTUM ESPRESSO: a modular and open-source software project for quantum simulations of materials. *Journal of Physics: Condensed Matter* 21, 395502 (2009).
- [6] Mostofi, A. A., et al. Wannier90: A tool for obtaining maximally-localised Wannier functions. Computer Physics Communications 178, 685-699 (2008).
- [7] Fuh, H.-R. and Guo, G.-Y. Intrinsic anomalous Hall effect in nickel: A GGA+U study. *Physical Review B* 84, 144427 (2011).
- [8] Fan, X., Celik, H., Wu, J., Ni, C., Lee, K.-J., Lorenz, V.O., and Xiao, J.Q. Quantifying interface and bulk contributions to spin-orbit torque in magnetic bilayers. *Nature Communications* 5, 3042 (2014).
- [9] Zhang, W., Jungfleisch, M. B., Jiang, W., Pearson, J. E., Hoffmann, A., Freimuth, F. and Mokrousov, Y. Spin Hall Effects in Metallic Antiferromagnets. *Physical Review Letters* 113, 196602 (2014).
- [10] Pesin, D. A. and MacDonald, A. H. Quantum kinetic theory of current-induced torques in Rashba ferromagnets. *Physical Review B* 86, 014416 (2012).
- [11] Amin, V. P., Zemen, J. and Stiles, M. D. Interface-Generated Spin Currents *Physical Review Letters* 121, 136805 (2018).
- [12] Park, Seung Ryong, Kim, Choong H., Yu, Jaejun, Han, Jung Hoon and Kim, Changyoung, Orbital-Angular-Momentum Based Origin of Rashba-Type Surface Band Splitting *Physical Review Letters* 107, 156803 (2011).
- [13] Krupin, O. and Bihlmayer, G. and Starke, K. and Gorovikov, S. and Prieto, J. E. and Döbrich, K. and Blügel, S. and Kaindl, G. Rashba effect at magnetic metal surfaces. *Physical Review B* 71, 201403 (2005).
- [14] Park, Jin-Hong, Kim, Choong H., Lee, Hyun-Woo and Han, Jung Hoon. Orbital chirality and Rashba interaction in magnetic bands. *Physical Review B* 87, 041301(R) (2013).

- [15] Lijun Zhu, Daniel C. Ralph and Robert A. Buhrman. Enhancement of spin transparency by interfacial alloying. *arXiv:1904.05455* (2019).
- [16] Belashchenko, K. D. and Kovalev, Alexey A. and van Schilfgaarde, M. First-principles calculation of spin-orbit torque in a Co/Pt bilayer. *Physical Review Materials* 3, 011401(R) (2019).
- [17] Haney, Paul M. and Lee, Hyun-Woo and Lee, Kyung-Jin and Manchon, Aurélien and Stiles, M. D. Current-induced torques and interfacial spin-orbit coupling. *Physical Review B* 88, 214417 (2013).
- [18] S. C. Baek, Vivek P. Amin, Young-Wan Oh, Gyungchoon Go, Seung-Jae Lee, Geun-Hee Lee, Kab-Jin Kim, M. D. Stiles, Byong-Guk Park and Kyung-Jin Lee, Spin currents and spin–orbit torques in ferromagnetic trilayers. *Nature Materials* 17, 509-513 (2018).
- [19] Amin, V. P. and Stiles, M. D. Spin transport at interfaces with spin-orbit coupling: Formalism *Physical Review B* **94**, 104419 (2016).
- [20] Amin, V. P. and Stiles, M. D. Spin transport at interfaces with spin-orbit coupling: Phenomenology *Physical Review B* 94, 104420 (2016).