

Observation of anti-damping spin–orbit torques generated by in-plane and out-of-plane spin polarizations in MnPd_3

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Large spin–orbit torques (SOTs) generated by topological materials and heavy metals interfaced with ferromagnets are promising for next-generation magnetic memory and logic devices. SOTs generated from y spin originating from spin Hall and Edelstein effects can realize field-free magnetization switching only when the magnetization and spin are collinear. Here we circumvent the above limitation by utilizing unconventional spins generated in a MnPd_3 thin film grown on an oxidized silicon substrate. We observe conventional SOT due to y spin, and out-of-plane and in-plane anti-damping-like torques originated from z spin and x spin, respectively, in $\text{MnPd}_3/\text{CoFeB}$ heterostructures. Notably, we have demonstrated complete field-free switching of perpendicular cobalt via out-of-plane anti-damping-like SOT. Density functional theory calculations show that the observed unconventional torques are due to the low symmetry of the (114)-oriented MnPd_3 films. Altogether our results provide a path toward realization of a practical spin channel in ultrafast magnetic memory and logic devices.

Efficient control of magnetization at ultra-high speed has been of prime interest to the spintronics community¹. Spin–orbit torque (SOT) has provided efficient and ultrafast control of magnetization in magnetoresistive random access memory (MRAM) and logic devices^{2,3}. SOT has been observed in magnetic semiconductors⁴ and heavy metals^{5–8}, topological insulators (TIs)^{9–13}, antiferromagnets^{14–18} and semimetals^{19–21} interfaced with a ferromagnetic layer. The charge current injected into the non-magnetic layer (spin channel) along the x direction generates a spin current along the z direction with spin polarization ($\hat{\sigma}$) pointing

along the y direction ($\hat{\sigma}_y$) in heavy metals due to the bulk spin Hall effect. In the case of topological insulators and Weyl semimetals non-equilibrium spin-density is accumulated at the interface due to the time reversal symmetry protected spin-momentum locking²². SOTs exerted on the ferromagnet with in-plane magnetic anisotropy (IMA) are in-plane anti-damping-like ($\tau_{\text{ADL}} \propto \hat{m} \times (\hat{\sigma}_y \times \hat{m})$) and out-of-plane field-like ($\tau_{\text{FL}} \propto \hat{\sigma}_y \times \hat{m}$), where $\hat{m}$ is the magnetization unit vector. Spin current with $\hat{\sigma}$ along the z direction ($\hat{\sigma}_z$) has been observed in transition-metal dichalcogenides^{19,21,23} and in ferromagnets interfaced

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with light metals²⁴ with the charge current flow along the x direction. SOT due to $\hat{\sigma}_z$ exerts torque along the out-of-plane direction ($\tau_{\text{ADL},z} \propto \hat{m} \times (\hat{\sigma}_z \times \hat{m})$), which will enable external-field-free and low-power switching of the out-of-plane magnetization²⁵. Recently, $\hat{\sigma}$ along the x direction ($\hat{\sigma}_x$) has been reported in the uncompensated antiferromagnet Mn₃GaN in addition to $\hat{\sigma}_y$ and $\hat{\sigma}_z$ due to the low magnetic symmetry¹⁸. SOT due to $\hat{\sigma}_x$ exerts torque along the in-plane direction ($\tau_{\text{ADL},x} \propto \hat{m} \times (\hat{\sigma}_x \times \hat{m})$), which will deterministically switch the magnetization along the x direction in the absence of an external magnetic field. The figure of merit of charge to spin conversion is known as the spin torque efficiency $\theta_k \propto \frac{\sigma_{ij}^k}{\sigma_{xx}}$, where σ_{ij}^k (i, j and k refer to the spin current flow, the charge current flow and the spin polarization directions, respectively) and σ_{xx} are spin and charge conductivities, respectively. θ_k needs to be high for efficient control of the magnetization. Furthermore, to avoid current shunting through a conducting ferromagnetic layer, a high σ_{ij}^k is also required²⁶. Another important requirement for the integration of a spin channel into semiconductor integrated circuit technology is the tolerance of SOT materials to thermal annealing at 400 °C. However, no practical spin channel appears to have been reported which can handle postannealing at that temperature, and also possesses high $\tau_{\text{ADL},y}$ along with $\tau_{\text{ADL},x}$ and $\tau_{\text{ADL},z}$, enabling deterministic switching of in-plane magnetization along y , in-plane magnetization along x , and out-of-plane magnetization, respectively, without the need to apply an external magnetic field.

To achieve high-density magnetic memory and logic devices, perpendicular magnetic anisotropy (PMA) is desired²⁷. PMA switching via SOT from heavy metals^{5–8} and TIs^{10,12,13} has been reported in the presence of an external magnetic field. Lateral structural asymmetry^{28–30} and anisotropy tilting of magnetization^{31,32} can switch PMA in the absence of an external magnetic field, but both techniques cannot be used for the large-scale production. Partial PMA switching has been observed on PtMn/[Co/Ni]_x (ref. 14), IrMn/CoFeB (ref. 15) and PtMn/CoFeB/Gd/CoFeB (ref. 17) stack structures, in the absence of an external magnetic field, but with the help of exchange bias. Stray field from an in-plane magnetic layer present above or below the spin channel can facilitate external magnetic field-free PMA switching, but current shunting and magnetic interference between different magnetic layers pose severe design constraints in this approach^{33,34}. An external field is required to prealign the bottom magnetic layer to achieve field-free PMA switching in a trilayer structure²⁴. Novel torque in low-symmetry single-crystal CuPt/CoPt heterostructures can switch field-free PMA; however, it is not compatible with the current technology³⁵. A combination of SOT and spin transfer torque (STT) can also switch PMA in the absence of an external magnetic field, but STT could lead to reduced endurance of the magnetic tunnel barrier and slower magnetization switching³⁶. Fast switching of the magnetization with lower critical current densities (J_{sw}) can be achieved when the charge current flow and the magnetization are collinear; however, this geometry still requires an external magnetic field to achieve magnetization switching⁸. Here we present SOT from sputtered MnPd₃ thin films postannealed at 400 °C that can generate a spin current with $\hat{\sigma}$ along all three axes due to the charge current flow along the x direction, which represents a major advance over the literature and overcomes key limitations of the existing SOT materials.

The thin films with the stack structure Si/SiO₂/MnPd₃ (t nm)/CoFeB (5 nm)/MgO (2 nm)/Ta (2 nm) with IMA were prepared for the SOT characterization with $t = 4, 6, 8, 10, 12, 16, 20$ and 24 nm, respectively. Unless otherwise stated, these films will be labelled MP4–MP24, in which the number denotes the MnPd₃ thickness. All samples were postannealed at 400 °C for 30 min.

Figure 1a shows the tetragonal unit cell of MnPd₃. Grazing incidence X-ray diffraction (GIX) measurements were carried out on an Si/SiO₂/MnPd₃ (50 nm) sample using a Bruker D8 Venture single-crystal diffractometer equipped with a two-dimensional Photon 100 detector. Figure 1b shows the integrated and background-subtracted

experimental profile and the calculated X-ray diffraction profile of the ordered tetragonal MnPd₃ phase. The unit cell was refined from the X-ray diffraction profile using Pawley's method and the obtained lattice parameters for the MnPd₃ phase are $a = b = 3.88$ Å and $c = 15.57$ Å. The (103), (110) and (105) Bragg peaks visible in the experimental X-ray diffraction profile unequivocally demonstrate the existence of the ordered tetragonal MnPd₃ phase. For the disordered cubic MnPd₃ phase these peaks are forbidden. The chemical order parameter, S , was determined as $S = \sqrt{\frac{(I_{110}/I_{114})_{\text{exp}}}{(I_{110}/I_{114})_{\text{calc}}}}$, where the I_{110} reflection is a superstructure

reflection that is only allowed in the ordered MnPd₃ structure, while the I_{114} reflection is a fundamental reflection allowed in both structures. The $(I_{110}/I_{114})_{\text{calc}}$ ratio was obtained to be 0.017. The value for $(I_{110}/I_{114})_{\text{exp}}$ was around 0.020 ± 0.005 , which makes the order parameter number close to 1.0. The relatively large error originates from the substantial texturing of the MnPd₃ thin film, as can be seen from Supplementary Information, section 1 and Supplementary Fig. 1. The cross-section transmission electron microscopy (TEM) bright-field image of the MP10 sample presented in Fig. 1c shows that the MnPd₃ layer grown on thermally oxidized silicon is polycrystalline. The CoFeB and MgO layers are also polycrystalline. The electric and magnetotransport measurements were performed on Si/SiO₂/MnPd₃ (10 nm)/MgO (2 nm)/Ta (2 nm) heterostructures, hereafter termed the MnPd (10 nm) sample (Supplementary Information, section 2 and Supplementary Fig. 2). The resistivity shows metallic behaviour coinciding with the possible transition from a paramagnetic to an antiferromagnetic state below 50 K, as shown in Fig. 1d. The Néel temperature of the MnPd (10 nm) sample was approximately 37 K as measured by temperature-dependent magnetometry (Supplementary Information, section 3 and Supplementary Fig. 3). Polarized neutron reflectometry (PNR) measurements show weak ferromagnetism, persisting up to room temperature, in MnPd₃ films, possibly originating from uncompensated manganese moments, manganese clusters or local ferromagnetic manganese-based compound formation. At room temperature the ferromagnetic component of the magnetization in MnPd₃ was determined to be 9 ± 1.9 kA m⁻¹ by PNR, whereas at 6 K it was -43 ± 3.4 kA m⁻¹ (Supplementary Information, section 4 and Supplementary Fig. 4).

We performed SOT measurements using the second harmonic Hall (SHH) technique on MP4–MP24 samples, control MP (10 nm) and Si/SiO₂/CoFeB (5 nm)/MgO (2 nm)/Ta (2 nm) samples, and a reference Si/SiO₂/Pt (10 nm)/CoFeB (5 nm)/MgO (2 nm)/Ta (2 nm) sample (hereafter termed the Pt10 sample)³⁷. Details of the SHH technique are presented in Supplementary Information, section 5 and Supplementary Fig. 5. The a.c. current injected into the Hall bar induces effective spin–orbit fields, which oscillate the magnetization around its equilibrium position, and as a result, an SHH voltage is induced. In the SHH measurement, the sample is rotated in the x – y plane under constant static magnetic field to keep the magnetization in a single domain state. The spin current with the spin magnetic moment pointing along the negative y axis and positive x and z axes get accumulated between MnPd₃ and CoFeB layers as shown in Fig. 2a. Thus, accumulated spin currents exert $\tau_{\text{ADL},x}$, $\tau_{\text{ADL},y}$ and $\tau_{\text{ADL},z}$, τ_{FL} on the CoFeB layer along the in-plane and out-of-plane directions, respectively. In addition, the Oersted field (H_{Oe}) generated due to the a.c. current flow in the MnPd₃ layer exerts an Oersted torque (τ_{Oe}) on the CoFeB layer. Figure 2b shows R_{xy}^{low} as a function of the angle (φ), where φ is the angle between the current flow and the magnetic field; ω is frequency of input a. c. current. R_{xy}^{low} fits perfectly to $\sin 2\varphi$, indicating that there was no out-of-plane field projection due to imperfect sample mounting. In the MP12 sample the offset in φ was -13.2° . $R_{xy}^{\text{2}\omega}$ as a function of φ is presented in Fig. 2c. Since the torques have different dependencies on φ , we can extract $R_{xy}^{\text{2}\omega}$ due to the different types of torques using the SHH. In the MP12 sample, the extracted spin–orbit fields associated with the $\tau_{\text{ADL},x}$, $\tau_{\text{ADL},y}$, $\tau_{\text{ADL},z}$ and τ_{FL} were (0.0046 ± 0.0004) , (0.10 ± 0.01) ,

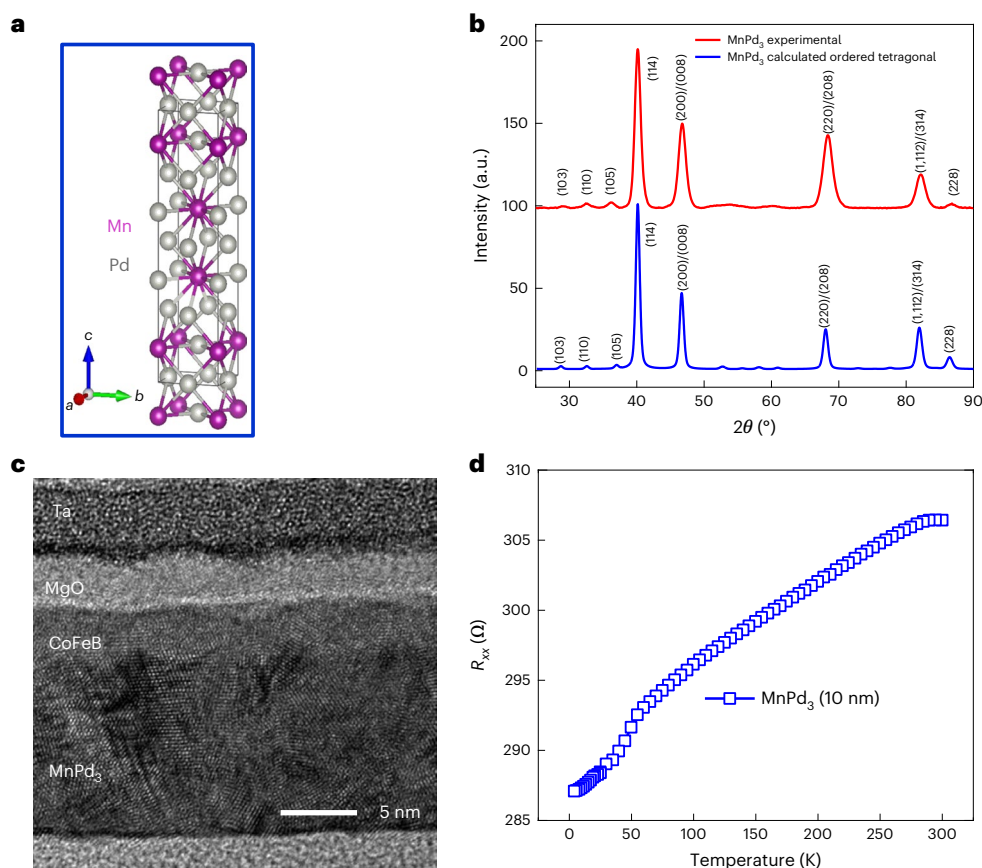


Fig. 1 | Characterization of MnPd₃ thin film. **a**, Schematic diagram of tetragonal MnPd₃ unit cell. **b**, Integrated experimental X-ray diffraction profile of 400 °C 30 min postannealed Si/SiO₂/MnPd₃ (50 nm) sample and the calculated X-ray diffraction profile of ordered tetragonal MnPd₃ phase. **c**, Cross-section TEM

image of an Si/SiO₂/MnPd₃ (10 nm)/CoFeB (5 nm)/MgO (2 nm)/Ta (2 nm) sample. **d**, Four-terminal resistance as a function of temperature for an Si/SiO₂/MnPd₃ (10 nm) sample.

(0.0030 ± 0.0001) and (0.0120 ± 0.0001) mT per 10^6 A cm⁻², respectively (Supplementary Information, section 5). These values of the $\tau_{\text{ADL},y}$ and τ_{FL} are comparable with or better than previous reports on heavy metals/ferromagnet³⁷, TIs/ferromagnet^{12,13} and Weyl semimetal/ferromagnets²⁰. As presented in Fig. 2d–f, θ_x^{eff} , θ_y^{eff} and θ_z^{eff} show MnPd₃ film thickness dependence according to the drift-diffusion model. The observed film thickness dependence of $\theta_{x,y,z}^{\text{eff}}$ clearly shows that the bulk spin Hall effect can originate unconventional SOT in polycrystalline MnPd₃ films.

The simple drift-diffusion model (equation (1)) can be utilized to extract the bulk spin–torque efficiency figure of merit (θ_y ($t \approx \infty$)) and the spin-diffusion length (λ_y),

$$\theta_y^{\text{eff}} = \theta_y(t \approx \infty) \left(1 - \text{sech} \left(\frac{t}{\lambda_y} \right) \right) \quad (1)$$

where t is the MnPd₃ film thickness. This drift-diffusion model considers that the spin current generated by the bulk of thin films is completely absorbed by the ferromagnetic layer without any dissipation at the interface and back-flow of the spin current. The θ_y ($t \approx \infty$) and λ_y obtained by the ordinary drift-diffusion model are 0.34 and 7.11 nm, respectively. Now by considering spin back-flow the drift-diffusion model can be modified into^{38–40}:

$$\theta_y^{\text{eff}} = \theta_y(t \approx \infty) \left(1 - \text{sech} \left(\frac{t}{\lambda_y} \right) \right) \left[\frac{1 + \tanh \left(\frac{t}{2\lambda_y} \right)}{2\rho\lambda_y G_{\uparrow\downarrow}} \right]^{-1} \quad (2)$$

where ρ is the bulk resistivity and $G_{\uparrow\downarrow}$ is the spin-mixing conductivity. The red line in Fig. 2d is a fit to equation (4) with θ_y ($t \approx \infty$) and λ_y as independent fitting parameters. The $G_{\uparrow\downarrow}$ values of the MP4 and MP24 samples were estimated to be 4.54×10^{14} m⁻² and 3.70×10^{15} m⁻², respectively. In thicker MnPd₃ films ($t > \lambda_y$), the damping constant was substantially enhanced because of the large spin current generated by the bulk compared with the thinner films. Thus, $G_{\uparrow\downarrow}$ is large in thicker MnPd₃ films. The extracted values of θ_y ($t \approx \infty$) and λ_y were 0.41 and 6.68 nm, respectively for a $G_{\uparrow\downarrow}$ value of 4.54×10^{14} m⁻². The ρ value used for the fitting was 60 $\mu\Omega\cdot\text{cm}$ obtained by measuring the four-terminal resistance of a Sub/MnPd₃ (20 nm)/MgO (2 nm)/Ta (2 nm) sample. The extracted values of θ_y ($t \approx \infty$) and λ_y were 0.34 and 7.13 nm, respectively for a $G_{\uparrow\downarrow}$ value of 3.70×10^{15} m⁻². The values of σ_{zx}^y corresponding to θ_y (0.34–0.41) were estimated to be $(5.70\text{--}6.80) \times 10^5$ $\hbar/2e$ Ω^{-1} m⁻¹. Similarly, the θ_x and θ_z values range from (0.015–0.017) and (0.0086–0.011), corresponding to $G_{\uparrow\downarrow}$ values of $(3.70\text{--}0.45) \times 10^{15}$ m⁻², respectively. λ_x and λ_z values range from (7.12–6.80 nm) and (7.50–8.04 nm) corresponding to $G_{\uparrow\downarrow}$ values of $(3.70\text{--}0.45) \times 10^{15}$ m⁻², respectively. The σ_{zx}^x and σ_{zx}^z values in the MP samples were as large as 0.28×10^5 $\hbar/2e$ Ω^{-1} m⁻¹ and 0.18×10^5 $\hbar/2e$ Ω^{-1} m⁻¹, respectively. σ_{zx}^z in the MP samples is comparable to recent reports on Mn₃GaN/permalloy (ref. 18) and WTe₂/permalloy (ref. 19). The values of σ_{zy}^y are among the largest reported for antiferromagnets^{16,41}, heavy metals^{6,42,43}, TIs^{9,12} and Weyl semimetals²⁰. $R_{\text{xy}}^{2\omega}$ in the control samples does not show any field or φ dependence, indicating that the SOTs in the MP samples originate from the MnPd₃ layer (Supplementary Information, section 5 and Supplementary Figs. 5 and 6). The values for the spin–orbit fields associated with $\tau_{\text{ADL},y}$ and τ_{FL} of the reference Pt10 sample were estimated to be

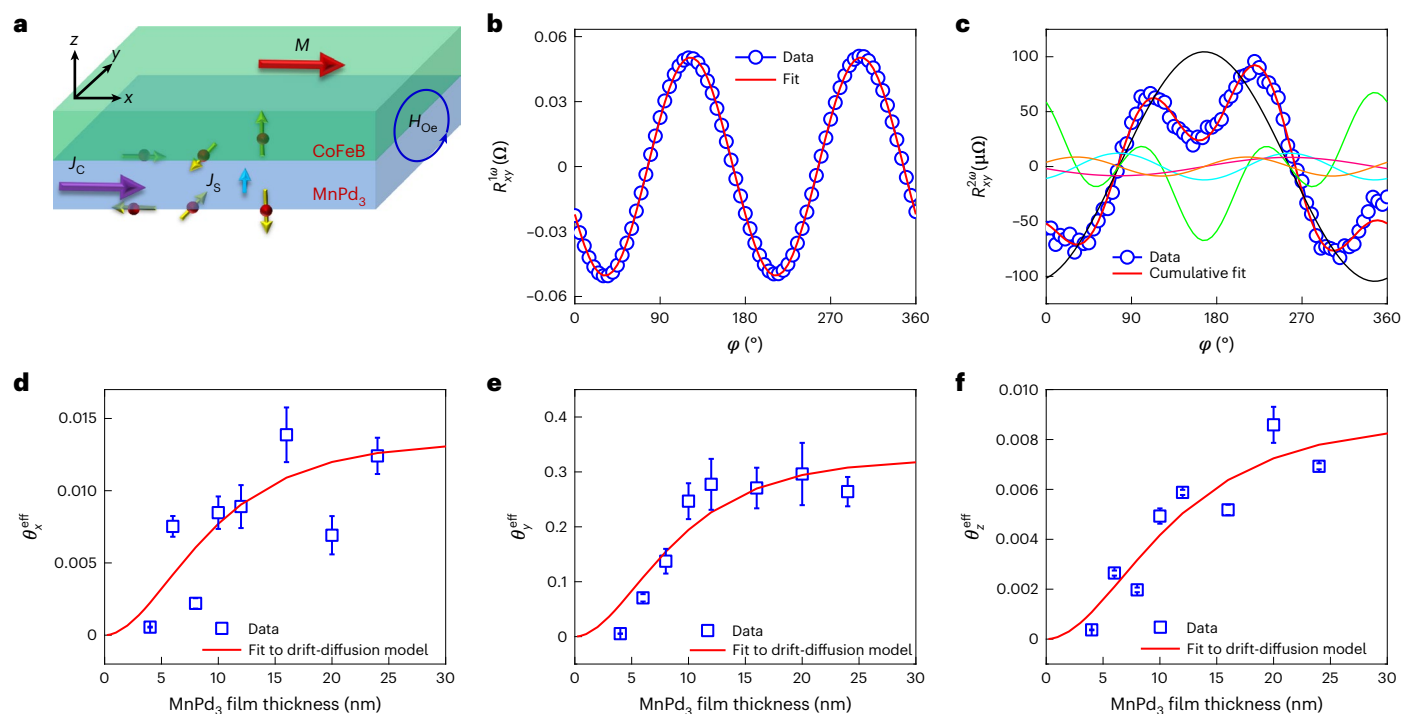


Fig. 2 | SOT characterization using the SHH technique on Si/SiO₂/MnPd₃ (x nm)/CoFeB (5 nm)/MgO (2 nm)/Ta (2 nm). **a**, Schematic diagram showing in-plane charge-current-generated spin current with spin polarizations along three axes. The red spheres represent electrons and yellow arrows represent spin magnetic moment. J_c , J_s , M and H_{Oe} represent charge current density, spin current density, magnetization and Oersted field, respectively. **b, c**, R_{xy}^{ω} (**b**) and $R_{xy}^{2\omega}$ (**c**) as a function of angle (ϕ) between the in-plane magnetic field and the current flow at

a fixed in-plane magnetic field amplitude of ~ 200 mT in the MP12 sample. The pink, black, cyan, green and orange lines are $R_{xy}^{2\omega}$ corresponding to $\tau_{ADL,x}$, $\tau_{ADL,y}$, $\tau_{ADL,z}$, $\tau_{FL} + \tau_{Oe}$, and the planar Nernst effect (PNE), respectively. **d–f**, Effective spin-torque efficiency due to the $\tau_{ADL,x}$ (**d**), $\tau_{ADL,y}$ (**e**) and $\tau_{ADL,z}$ (**f**) as a function of MnPd₃ film thickness. The Hall bar device used for this measurement was 10 μ m wide and 130 μ m long. Error bars are relative errors; some of these are smaller than the size of the symbol.

(0.040 ± 0.003) and (0.012 ± 0.001) mT per 10^6 A cm⁻², respectively (Supplementary Information, section 7 and Supplementary Fig. 7). The estimated value of θ_y^{eff} and θ_{FL}^{eff} are (0.07 ± 0.01) and (0.02 ± 0.002), respectively, in agreement with previous reports^{6,37,44}. After postannealing at 400 °C, the reference Pt10 sample does not show an SHH signal, suggesting that its SOT did not withstand such annealing.

We also performed spin-torque ferromagnetic resonance (ST-FMR) measurements on the MP10 sample to confirm the SOT observed with the SHH technique. The details of the ST-FMR are presented in Supplementary Information, 8 and Supplementary Fig. 8. The θ_y^{eff} estimated by ST-FMR of the MP10 sample is 0.21 ± 0.01 . The θ_y^{eff} of the as-deposited reference platinum and tungsten samples were determined to be 0.06 ± 0.01 and -0.43 ± 0.03 , respectively. The θ_y^{eff} of the tungsten postannealed sample was estimated to be -0.011 (Supplementary Information, section 9 and Supplementary Fig. 9).

To demonstrate external magnetic field-free PMA switching, we prepared Si/SiO₂/MnPd₃ (10 and 12 nm)/Co (1 nm)/MgO (2 nm)/Ta (2 nm) samples (hereafter termed MP10/Co1 and MP12/Co1 samples). The MP10/Co1 and MP12/Co1 samples were annealed at 400 °C for 30 min in a vacuum and subsequently field-cooled under the application of an out-of-plane magnetic field of 0.45 T. Figure 3a shows anomalous Hall resistance (R_{AHE}) as a function of the out-of-plane magnetic field. The hysteretic R_{AHE} loop confirms PMA is present in the cobalt layer. Magnetometry was also used to confirm that PMA is present in MP10/Co1 sample (Supplementary Information, section 3 and Supplementary Fig. 3b). The PMA field (H_k) was estimated to be 500 mT on the blank films. Figure 3b shows current-induced SOT magnetization switching under the presence of an external magnetic field (H_x) along -x direction. The write d.c. current pulse width used for PMA switching

was 20 ms, which was followed by a read current of 0.4 mA. The full switching of magnetization occurs in the MP10/Co1 and MP12/Co1 samples at ~ 10.1 mA (9.57 MA cm⁻²) and ~ 9.5 mA (7.50 MA cm⁻²), respectively. Then, in the absence of an external magnetic field, the d.c. current was swept in pulses from -36 mA to $+36$ mA with a step size of 1.06 mA. For MP10/Co1 as shown in Fig. 3c, partial switching of the magnetization occurs at a positive current of ~ 10.78 mA, and complete switching occurs at ~ 32.4 mA (-29.72 MA cm⁻²). In the subsequent reverse sweep, partial switching of magnetization occurs at ~ -10.87 mA and complete switching occurs at ~ -31.63 mA. For the MP12/Co1 sample as shown in Fig. 3d, partial switching of the magnetization occurs at a positive current of -11.35 mA. Continuously sweeping the d.c. pulses switches the remaining magnetization at -30.92 mA (-24.7 MA cm⁻²). Subsequently, with reverse sweeping of the current pulses, partial switching of magnetization occurs at ~ -9.60 mA and switching of the remaining magnetization occurs at ~ -33.32 mA. Since the R_{AHE} values obtained by field sweeping and current sweeping are similar, we can conclude that full switching of PMA has been observed in both PMA samples. The J_{sw} values observed in our PMA samples without an external magnetic field are comparable with or better than previously reported values in Pt/Co (refs. 5,6,44) (-23 – 100 MA cm⁻²) and Pd_{0.25}Pt_{0.75}/Co (ref. 45) (-22 MA cm⁻²) with an external magnetic field. In Table 1, we summarize the θ_k , σ_{ij}^k , ρ and $\frac{H_k}{J_{\text{sw}}}$ of different spin channels postannealed at different temperatures and measured at room temperature. In our reference Pt (10 nm)/Co (1 nm) sample at -100 mT full switching of PMA was achieved with a J_{sw} of 27.3 MA cm⁻². The SOT switching of magnetization in our PMA samples results from the interplay of $\tau_{ADL,x}$, $\tau_{ADL,y}$, $\tau_{ADL,z}$ and τ_{FL} . In the presence of a relatively weaker $\tau_{ADL,z}$ and $\tau_{ADL,x}$ and strong $\tau_{ADL,y}$ the magnetization was partially switched at a lower current (~ 10 mA), which results in an

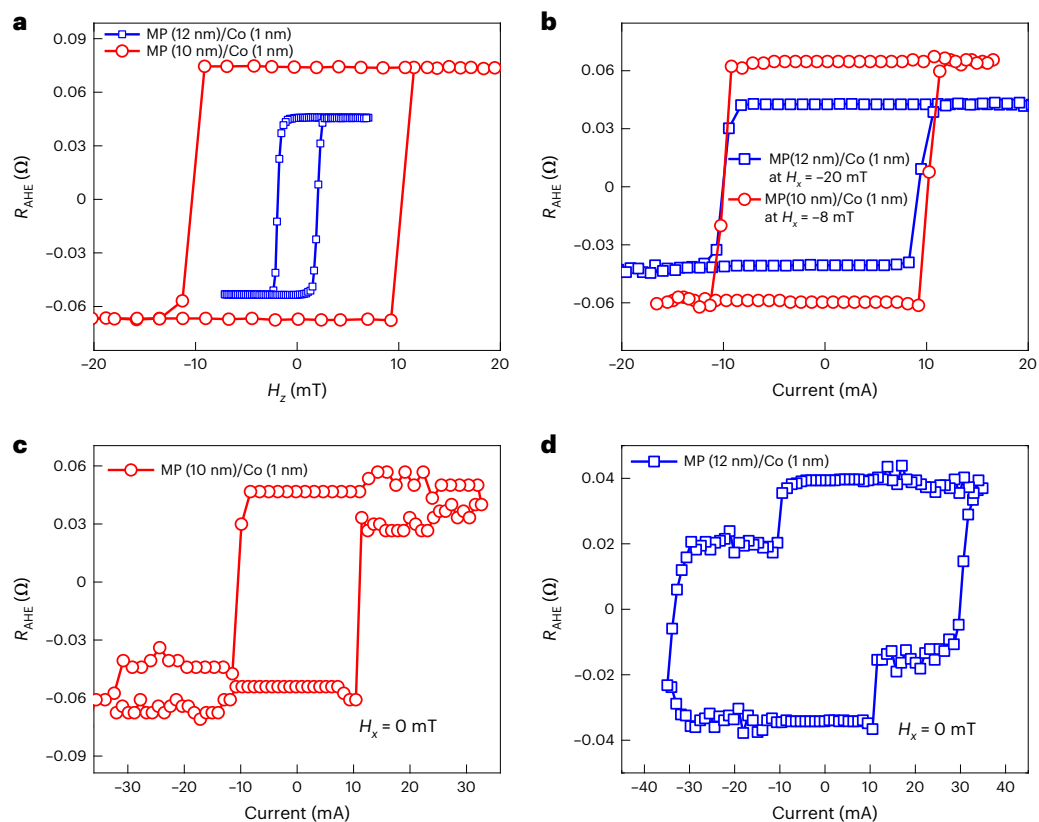


Fig. 3 | Demonstration of external magnetic field-free out-of-plane magnetization switching via z spin polarization generated anti-damping SOT. **a**, The anomalous Hall resistance as a function of the out-of-plane magnetic field. **b**, Switching of perpendicular cobalt layer via SOT under the application of -8 mT and -20 mT magnetic fields along the x direction (H_x) for MP (10 nm)/Co

(1 nm) and MP (12 nm)/Co (1 nm) samples, respectively. **c,d**, MP (10 nm)/Co (1 nm) (**c**) and MP (12 nm)/Co (1 nm) (**d**). Field-free perpendicular Co layer switching via $\tau_{ADL,z}$ generated by z spin polarization. A Hall bar $130\text{ }\mu\text{m}$ long and $10\text{ }\mu\text{m}$ wide was used for the magnetization switching experiment.

Table 1 | Summary of θ_k , σ_{ij}^k , ρ and $\frac{H_k}{J_{sw}}$ of different spin channels postannealed at different temperatures and measured at room temperature

Materials	ρ ($\mu\Omega\cdot\text{cm}$)	θ_x	θ_y	θ_z	θ_{FL}^{eff}	σ_{zx}^y ($\hbar/2e$ $10^5\text{ }\Omega^{-1}\text{m}^{-1}$)	J_{sw} (MA cm^{-2})	$\frac{H_k}{J_{sw}}$ ($\text{mT cm}^2\text{MA}^{-1}$)	H_x (mT)	Postannealing temperature ($^{\circ}\text{C}$)
This work	60–95	0.017	0.41	0.011	0.06	6.80	11–24.7	20.24	Zero	400
Pt (ref. 44)	40	–	0.14	–	0.08	3.5	60	8.33	70	As deposited
W (ref. 42)	188	–	–0.22	–	–0.022	–1.2	11 (ref. 7)	48.20	20	200 (ref. 7) to 300 (ref. 42)
MnGaN (ref. 18)	225	–0.013	0.025	0.018	–0.15	0.11	–	–	–	As deposited
WTe ₂ (ref. 19)	380	–	0.03	0.013	0.034	0.08	–	–	–	As deposited

intermediate state. The intermediate state can occur due to an insufficient external magnetic field, which was unable to completely break mirror symmetry⁴⁶. This behaviour has been observed in our reference sample at a smaller applied field. Current spreading at the Hall crosses can also result in partial switching of magnetization. Previously, intermediate magnetic states were observed below the threshold J_{sw} (ref. 47). The external-magnetic-field-free switching of PMA unambiguously demonstrates that the presence of $\hat{\sigma}_z$ generated $\tau_{ADL,z}$ in the MP samples. These experimental results of PMA switching have been qualitatively reproduced by micromagnetic simulations (Supplementary Information, section 10 and Supplementary Fig. 10). In addition to field-free PMA switching, we also performed field-free magnetization switching of the in-plane CoFeB layer

in the MP24 sample, as detected by using the unidirectional spin Hall magnetoresistance mechanism (Supplementary Information, section 11 and Supplementary Fig. 11). J_{sw} was estimated to be -11.0 MA cm^{-2} using the parallel resistor model.

We explain the appearance of the non-vanishing $\hat{\sigma}_x$ and $\hat{\sigma}_z$ in terms of the contribution from grains of different orientations in our polycrystalline MnPd₃ films. MnPd₃ has the crystal space group $I4/mmm$ (ref. 48). If the film were monocrystalline and (001) oriented with the charge current flowing along the [100] direction (the x direction), only the conventional spin Hall conductivity σ_{zx}^y (where the spin current flows along the [001] direction (the z direction) normal to the charge current and has spin polarization along the [010] direction (the y direction)) would be allowed due to the (001) plane being

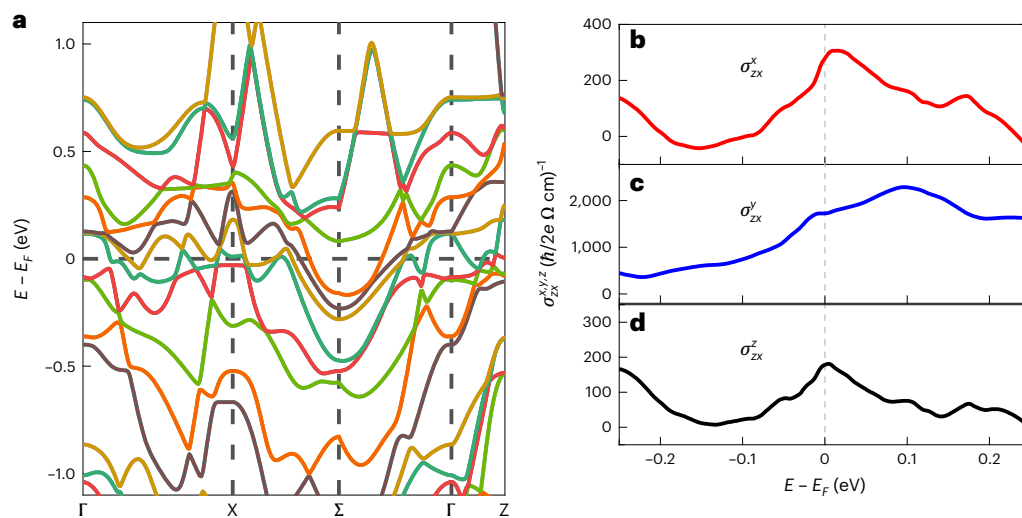


Fig. 4 | Effect of (114) texture on the spin polarization. **a**, The calculated band structure for the energies (E) around the Fermi level (E_F) of stoichiometric MnPd₃ at room temperature. The labels on the x axis denote the high symmetric points

in the Brillouin zone. **b–d**, Spin Hall conductivity as a function of energy. The calculated σ_{zx}^x (**b**), σ_{zx}^y (**c**) and σ_{zx}^z (**d**) as a function of energy for MnPd₃ (114) film, where the x axis is oriented along the [401] direction.

invariant to all symmetry operations of this space group. In a polycrystalline film, however, other crystal orientations with lower symmetries contribute to the spin Hall conductivity. For example, the (114) plane corresponding to the dominant texture of our films (reflected by the strongest X-ray diffraction peak in Fig. 1b) is only invariant with respect to mirror symmetry $M_{[110]}$ and two-fold rotation $C_{[110]}$. This allows the appearance of unconventional components of the spin Hall conductivity tensor, such as σ_{zx}^x and σ_{zx}^z , where the spin polarization is parallel to the direction of the spin current or the charge current. Moreover, in a polycrystalline film, the current direction itself varies with respect to the high-symmetry directions of different grains, which also influences the shape of the spin Hall conductivity tensor. These aspects have been discussed in ref. 16.

To quantitatively evaluate the contribution from this mechanism, we perform first-principles density functional theory (DFT) calculations of spin Hall conductivity of bulk MnPd₃ assuming a room-temperature paramagnetic phase. Figure 4a shows the calculated band structure of MnPd₃. There are several bands crossing the Fermi level (E_F), indicating the metallic ground state. The small gaps between the bands near E_F are favourable for the sizeable spin Hall conductivity⁴⁹, which is given by⁵⁰

$$\sigma_{ij}^k = \frac{e^2}{\hbar} \int \frac{d^3\mathbf{k}}{(2\pi)^3} \sum_n f_{nk} \Omega_{n,ij}^k(\mathbf{k}), \quad (3)$$

$$\Omega_{n,ij}^k(\mathbf{k}) = -2\Im \sum_{n' \neq n} \frac{\langle n\mathbf{k} | J_i^k | n'\mathbf{k} \rangle \langle n'\mathbf{k} | v_j | n\mathbf{k} \rangle}{(E_{n\mathbf{k}} - E_{n'\mathbf{k}})^2}, \quad (4)$$

where f_{nk} is the Fermi–Dirac distribution function for band n and wave vector $\mathbf{k}$, $\Omega_{n,ij}^k(\mathbf{k})$ is the spin Berry curvature, $J_i^k = \frac{1}{2}\{v_i, s_k\}$ is the spin current operator, v_i and s_k are velocity and spin operators, respectively, and $i, j, k = x, y, z$. As expected, for MnPd₃ textured in the (001) plane, only the conventional σ_{zx}^y is non-vanishing (Table 2 in Supplementary Information, section 13). However, for the dominant (114) stacking texture, the unconventional spin Hall conductivities (σ_{zx}^x and σ_{zx}^z) emerge (Table 2 in Supplementary Information, section 13). Figure 4b–d shows the calculated spin Hall conductivities for MnPd₃ (114) film as a function of energy when the charge current flows along the [401] direction (the x direction). We find a high conventional $\sigma_{zx}^y \approx 1,744 \left(\frac{h}{2e}\right) (\Omega \text{ cm})^{-1}$ and sizeable unconventional

conductivities $\sigma_{zx}^x \approx 279 \left(\frac{h}{2e}\right) (\Omega \text{ cm})^{-1}$ and $\sigma_{zx}^z \approx 166 \left(\frac{h}{2e}\right) (\Omega \text{ cm})^{-1}$ at the Fermi energy. It is evident that the σ_{zx}^x and σ_{zx}^z values are approximately an order of magnitude smaller than σ_{zx}^y , which is consistent with our experimental observation. Unconventional SOTs present in the antiferromagnetic regime of the MP10 sample (Supplementary Information, section 14 and Supplementary Fig. 14) confirms that low crystal symmetry is the reason for the origin of $\tau_{\text{ADL},x}$ and $\tau_{\text{ADL},z}$. As we specifically showed by DFT calculations, the spin Hall conductivities generated by the grains with different orientations can have different magnitudes and signs. Therefore, the net spin Hall conductivity of the film might be weakened substantially if these grains have the equivalent sizes and concentrations. However, once a grain orientation is dominating, the net spin Hall conductivity can be finite to generate sizeable SOTs. The control of film orientation and hence the SOT require further work to improve the crystalline texture of the film, which is beyond the scope of the current work.

In summary, we studied anti-damping SOTs generated by the δ_x , δ_y and δ_z in MnPd₃/ferromagnet heterostructures. In both the paramagnetic and the antiferromagnetic phase unconventional SOTs were observed. DFT simulations confirmed the low crystal symmetry present in the (114)-oriented MnPd₃ thin films as the origin of the observed unconventional SOTs. We demonstrated successful growth of conductive MnPd₃ thin films with high σ_{zx}^y and sizeable σ_{zx}^x and σ_{zx}^z after postannealing at 400 °C for half an hour. Complete external-magnetic-field-free switching of both IMA and PMA were realized. The observed SOTs were robust against thermal treatment, and compatible with the low damping constant of CoFeB even after postannealing. All of these are key factors for the integration of a practical spin current source based on antiferromagnetic MnPd₃ into the next generation of SOT-based spintronics devices.

Online content

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Methods

Sample growth and thin film characterization

MnPd₃ thin films for the magnetotransport, SOT characterization and magnetization switching were grown on Si/SiO₂ substrate at room temperature by sputtering a composite MnPd₂ (99.99% pure) target in an AJA sputtering system with a base pressure of 2.0×10^{-8} torr. MnPd₃, CoFeB, platinum and tantalum were d.c. sputtered at 25 W power with deposition rates of 0.27, 0.056, 0.27 and 0.07 Å s⁻¹, respectively. The MgO layer was radiofrequency sputtered at 50 W power with a deposition rate of 0.013 Å s⁻¹. Rutherford backscattering measurements give the atomic composition of manganese and palladium in the MnPd₃ film as 28% and 72%, respectively.

Thin sections for TEM were prepared by a focused ion beam in an FEI Helios Nanolab 660 (ref.). TEM analysis was carried out with a Hitachi HF-5000 equipped with a probe aberration corrector, under 200 kV. Bright field, secondary electron and annular (both medium- and high-angle) dark-field images were captured simultaneously in scanning TEM mode. High-resolution energy-dispersive X-ray spectroscopy mapping and line scans were performed with an Oxford Instruments large solid angle (2.0 sr) dual windowless detector.

Zero field cooled and field cooled magnetization data were collected as a function of temperature using a Quantum Design superconducting quantum interference device magnetometer.

Device fabrication and electrical characterization

The multilayer thin films were patterned into Hall bars for the quantum transport, SHH and magnetization switching experiments using an optical lithography process. The Hall bars used for the experiments were 10 µm wide and 130 µm long. The separation between the Hall crosses in the Hall bar device was 37 µm. For the ST-FMR measurements thin films were patterned into rectangular strips 25–30 µm long and 20 µm wide. After the first photolithography step, an argon ion mill was used to define the device geometry. Ti (10 nm)/Au (150 nm) thick electrical contacts were deposited after the second lithography step using a KJL e-beam evaporator. A physical property measurement system was used to determine the electrical, magnetotransport and SHH measurements. Two Lockins SR830s and a 6221 current source were used to probe the first and second harmonic voltage and supply current, respectively. An Agilent HP 83620B was used to supply the radiofrequency current for the ST-FMR measurements. The d.c. voltage was measured with a Keithley 2000 multimeter.

Data availability

The data that support the findings of this study are available from the corresponding authors on reasonable request.

Code availability

The code used for the findings of this study are available from the corresponding authors on reasonable request.

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Author contributions

M. DC conceived, designed and coordinated the research with contributions from M.M., S.-J.L., W.T. and S.X.W. S.X.W. supervised the study. M. DC grew the thin films, performed the X-ray diffraction measurements and fabricated the Hall bars and the ST-FMR device, and carried out ST-FMR, SHH and switching measurements with contributions from Y.D., X.L., C.B., F.X. and Y.-L.H. D.-F.S. and E.Y.T. performed DFT calculations. V.D.-H.H., A.H. and W.-G.W. carried out TEM and energy-dispersive X-ray spectroscopy studies. A.V. performed grazing incidence X-ray diffraction measurements. M.B.V., S.E. and C.-M.L. performed magnetometry measurements. P.E.Q., B.K. and J.A.B. performed PNR measurements and modelling. Y.-L.H. performed micromagnetic simulations. M. DC performed data analysis and wrote the manuscript with contributions from D.-F.S., P.E.Q., A.V. and S.X.W. All authors discussed the results and commented on the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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