

ADVANCED MATERIALS

Supporting Information

for *Adv. Mater.*, DOI 10.1002/adma.202209866

Composite Spin Hall Conductivity from Non-Collinear Antiferromagnetic Order

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Steve Novakov^{1*}, Peter B. Meisenheimer², Grace A. Pan³, Patrick Kezer⁴, Nguyen M. Vu²,
Alexander J. Grutter⁵, Ryan F. Need^{5,6}, Julia A. Mundy³ & John T. Heron^{2*}

¹Department of Physics, University of Michigan, Ann Arbor, MI, 48109, United States

²Department of Material Science and Engineering, University of Michigan, Ann Arbor, MI,
48109, United States

³Department of Physics, Harvard University, Cambridge, MA, 02138, United States

⁴Department of Electrical and Computer Engineering, University of Michigan, Ann Arbor, MI,
48109, United States

⁵NIST Center for Neutron Research, National Institute of Standards and Technology,
Gaithersburg, MD 20899-6102, United States

⁶Department of Materials Science and Engineering, University of Florida, Gainesville, Florida,
32611, United States

[†]Corresponding authors: steveeee@umich.edu, jtheron@umich.edu

Contents:

Supplementary Note 1. Neutron Reflectometry of PtMn₃ Films on MgO and SrTiO₃

Supplementary Note 2. Neutron Diffraction of PtMn₃ Films on MgO and SrTiO₃

Supplementary Note 3. Scanning Transmission Electron Microscopy of PtMn₃ and PtMn₃/Py Films on SrTiO₃

Supplementary Note 4. Current distribution in PtMn₃/Py Heterostructures

Supplementary Note 5. Intrinsic Spin Conductivity in (001)-oriented PtMn₃

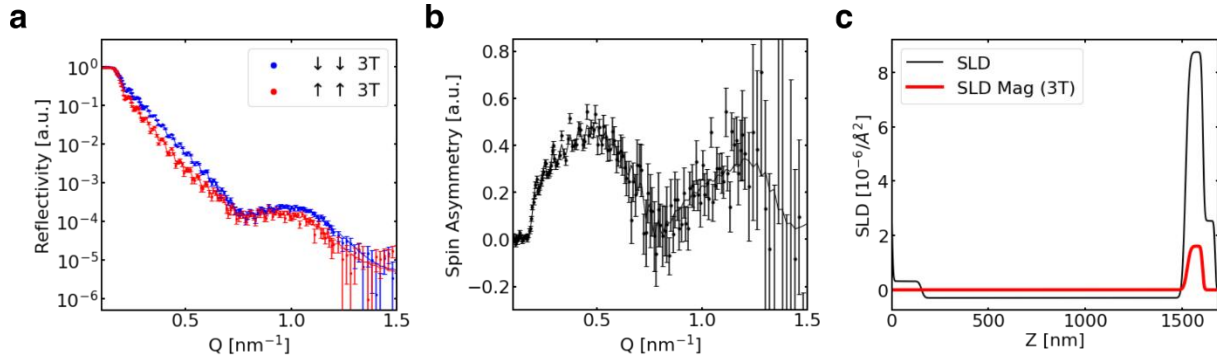
Supplementary Note 6. Harmonic Hall Effect Measurements of PtMn₃/Py

Supplementary Note 1. Neutron Reflectometry of PtMn₃ Films on MgO and SrTiO₃

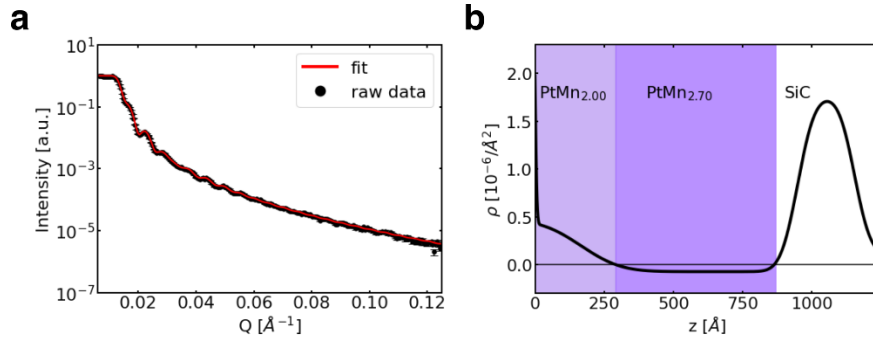
The fitted neutron reflectivity profiles for the MgO/PtMn₃(152 nm)/Py(8.9 nm)/TiO₂(6 nm) sample are shown in Supplementary Figure 1a. There is no evidence for a net magnetization in the PtMn₃, as per the spin asymmetry of the reflectivity profiles and the resulting magnetic profile in Supplementary Figure 1b and 1c, respectively. Obtaining a high-quality fit to the data required breaking up the PtMn₃ into two separate layers, with a 15.5 nm $\pm$ 0.5 nm thick transition growth region (unless otherwise noted, all uncertainties represent ± 1 standard deviation). This transitional growth region exhibits a higher nuclear SLD ($0.317 \cdot 10^{-6} \text{ \AA}^{-2} \pm 0.084 \cdot 10^{-6} \text{ \AA}^{-2}$) than the bulk PtMn₃ layer ($-0.30 \cdot 10^{-6} \text{ \AA}^{-2} \pm 0.02 \cdot 10^{-6} \text{ \AA}^{-2}$). The fitted scattering length density is related to the composition of Pt_{1-x}Mn_{3+x} by $SLD = 10((3+x)b_{Mn} + (1-x)b_{Pt})/a^2c$, where b_{Mn} , b_{Pt} are the nuclear scattering lengths, in fm, and a , c are the lattice constants in $\text{\AA}$ ($a = c = 3.84 \text{ \AA}$ for the cubic system). For $x = 0$, we expect $SLD = -0.282 \cdot 10^{-6} \text{ \AA}^{-2}$ for the bulk cubic system and $SLD = -0.275 \cdot 10^{-6} \text{ \AA}^{-2}$ for our tetragonal films on MgO and SrTiO₃ with the measured a , c from X-ray diffraction. For the MgO sample, we find that $x = -0.258 \pm 0.037$ for the transition layer (Pt_{1-x}Mn_{3+x} $\sim$ PtMn_{2.18}) and $x = 0.010 \pm 0.008$ for the bulk of the film ($\sim$ PtMn_{3.04}). We speculate that lower Mn content near the substrate interface is a method of lattice parameter relaxation, as stated in the main text.

Measurements on CANDOR were performed on a SrTiO₃/PtMn₃(95 nm)/SiC(20 nm) heterostructure in an unpolarized configuration using an incident white beam of neutrons with wavelengths ranging between 4 $\text{\AA}$ – 6 $\text{\AA}$. The unpolarized neutron reflectivity, which is sensitive to the nuclear SLD depth profile, was measured as a function of the momentum transfer vector Q along the film normal. The reflectivity profile, fit, and SLD profile are shown in Supplementary Figure 2. Again, we note a Pt-rich transitional growth region of 18.4 nm $\pm$ 0.3 nm and nuclear

SLD of $0.471 \cdot 10^{-6} \text{ \AA}^{-2} \pm 0.007 \cdot 10^{-6} \text{ \AA}^{-2}$ near the SrTiO₃ interface. The SLD of the thicker, more bulk-like, PtMn₃ layer was $-0.08 \cdot 10^{-6} \text{ \AA}^{-2} \pm 0.02 \cdot 10^{-6} \text{ \AA}^{-2}$. Both SLD values indicate that the film on SrTiO₃ is more Pt-rich than the sample on MgO, with $x = -0.326 \pm 0.003$ for the transition layer ($\sim\text{PtMn}_{2.0}$) and $x = -0.084 \pm 0.019$ for the bulk of the film ($\sim\text{PtMn}_{2.70}$).



Supplementary Figure 1 | Polarized neutron reflectivity of PtMn₃/Py on MgO. **a**, Non-spin flip reflectivities of MgO/PtMn₃(152 nm)/Py(8.9 nm)/TiO₂(6 nm), and **b**, the resulting spin asymmetry. **c**, The extracted magnetic (red) and non-magnetic SLD (black). The magnetic SLD shows a clear lack of any net magnetization in the PtMn₃ layer, with only the Py layer showing a non-zero net magnetization as expected. Error bars represent ± 1 standard deviation.



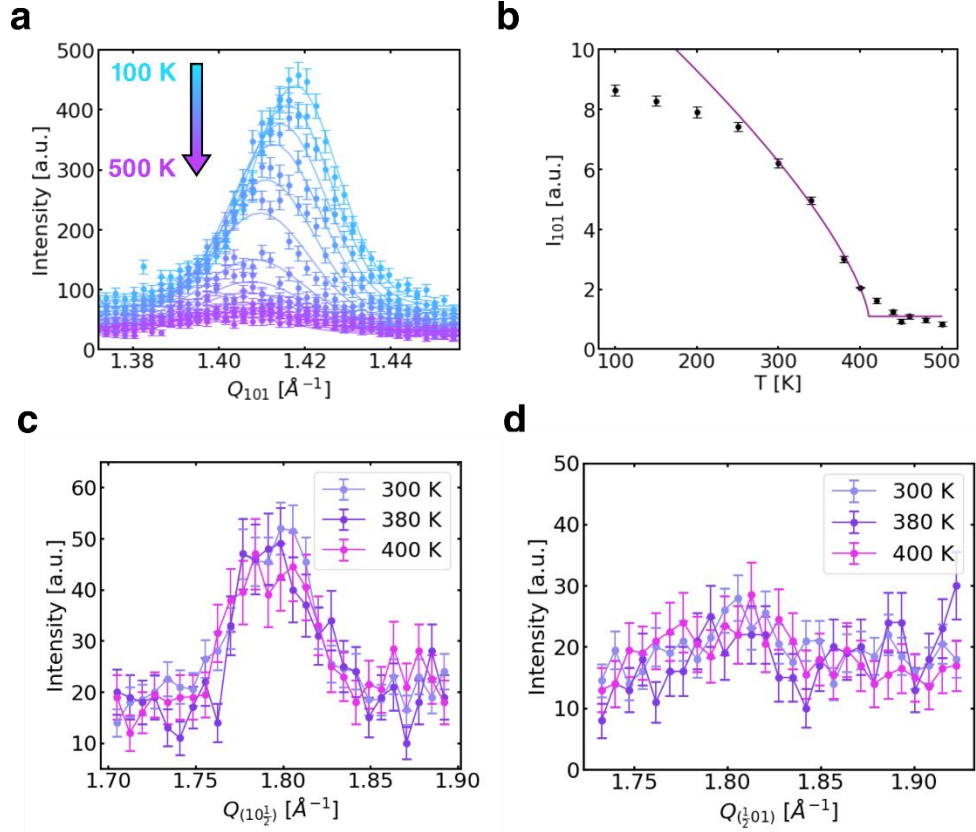
Supplementary Figure 2 | Unpolarized Neutron reflectivity of PtMn₃ on SrTiO₃. **a**, Reflectivity profile of SrTiO₃/PtMn₃(95 nm)/SiC(20 nm) and **b**, the resulting nuclear SLD. Both MgO and SrTiO₃ samples show a ~ 20 nm transition region with reduced Mn stoichiometry, followed by an approximately stoichiometric “bulk” region for the rest of the film.

Supplementary Note 2. Neutron Diffraction of PtMn₃ Films on MgO and SrTiO₃

To probe the AFM order in the PtMn₃ films directly, we perform neutron diffraction measurements of the 101 peak on the BT-7 and BT-4 triple axis spectrometers at the NIST Center for Neutron Research (see Methods). Though the L1₂ ordering on SrTiO₃ is markedly higher, a distinct 101 PtMn₃ peak was not resolved to the high overlap with the substrate. Unlike our XRD RSM measurements, the Q range of higher order peaks (e.g. 024 or equivalent) is out of the measurement range. For this reason, we perform temperature dependent neutron diffraction measurements on MgO/PtMn₃ samples as the in-plane peak separation is higher and MgO does not have 001 and 003 peaks. The temperature dependent neutron diffraction measurements of the 101 PtMn₃ peak are shown in Supplementary Figure 3a. The integrated peak intensities and their standard errors are extracted via gaussian fits to each profile (Methods), and are plotted vs. temperature in Supplementary Figure 3b. The order parameter $M(T)$ was fit to the temperature dependent intensity within 100 K of T_N with $I_{101} = M_{101}^2 \left(\frac{T_N - T}{T_N} \right)^{2\beta} + I_0$. The fit yields $T_N = 403 \pm 2$ K, and $\beta = 0.358 \pm 0.032$, which is within the expected range for a 3D ($\beta \sim 0.37$) Heisenberg AFM. The variance in β could be well explained by the large γ -PtMn₃ content in our MgO samples ($\sim 40\%$ by volume) and the tendency to form isolated planes of Mn in this phase, as per main text Figure 1c, which would result in mixed 2D/3D AFM behavior. Compared to the ideal bulk cubic material ($T_N = 450$ K – 475 K)^[1], the PtMn₃ films on MgO also have a T_N that is approximately 50 K lower.

Unlike the clear second-order behavior of the 101 peak shown in Supplementary Figure 3a-b, the intensity of the $10\frac{1}{2}$ peak, shown in Supplementary Figure 3c is temperature independent, as is the $\frac{1}{2}01$ peak shown in Supplementary Figure 3d. This contrasts with neutron

diffraction measurements of bulk PtMn_3 crystals, which show an obvious first-order transition between non-collinear and collinear phases between 300 K - 400 K depending on the Mn stoichiometry.^[1] For the PtMn_3 films on MgO, the evidence points towards a smooth second order phase transition from the non-collinear phase to a magnetically disordered phase above 404 K. The disappearance of the collinear phase for these highly strained films is not completely surprising, as it also disappears in other alloys in this family such as RhMn_3 ,^[2] suggesting that the emergence of the collinear and non-collinear phases, as well as the associated Néel and transition temperatures are sensitive to chemical composition and lattice parameter. In contrast to films grown on MgO, neutron diffraction on $\text{SrTiO}_3/\text{PtMn}_3$ samples represents a significant challenge, as the films are epitaxially strained in-plane to closely match the SrTiO_3 lattice constant of 3.905 Å. Relative to films on MgO, the substrate/film peak separation is also substantially reduced along the c-axis, with a PtMn_3 lattice constants of 3.82 Å along this direction, as shown in main text Figure 1c. Such separation is generally insufficient to separately resolve a 101 thin film peak using neutron diffraction. Attempting a high-resolution measurement with CANDOR, the white beam reflectometer/diffractometer at the NIST Center for Neutron Research, we find that the tail of the substrate peak overwhelmingly dominates signal in the PtMn_3 101 position even for extremely tight beam collimation, and it was not possible to separate the two for analysis.



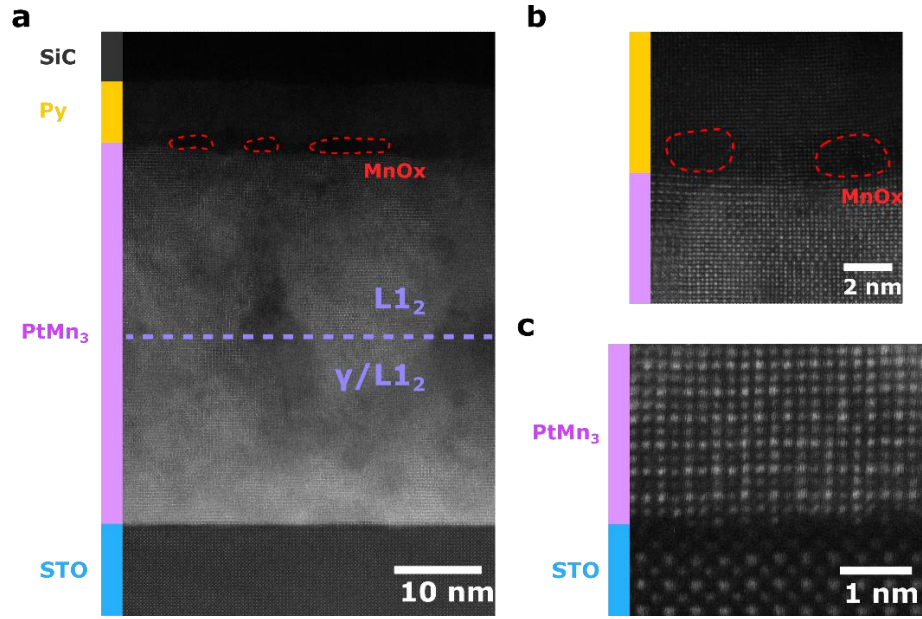
Supplementary Figure 3 | Neutron diffraction of 001 PtMn₃ on MgO. **a**, Temperature dependent neutron diffraction of the 101 PtMn₃ peak for MgO/PtMn₃(152 nm)/Py(8.9 nm)/TiO₂ **b**, with $T_N = 403(2)$ K and $\beta = 0.358(32)$ near T_N , in good agreement with a 3D Heisenberg system. In general, the PtMn₃ films on MgO and SrTiO₃ exhibit tetragonal strain and have lower transition temperatures compared to the bulk cubic film. **c**, $10\frac{1}{2}$ and **d**, $\frac{1}{2}01$ diffraction peaks in the same system on MgO showing no change in amplitude between 3 K - 400 K. The difference in peak height between $10\frac{1}{2}$ and $\frac{1}{2}01$ is attributed to slight misalignment due to temperature dependent tetragonal strain. The lack of Néel or first order transition for these peaks suggests total suppression of the collinear AFM phase in films grown on MgO. Error bars represent ± 1 standard deviation.

Supplementary Note 3. Scanning Transmission Electron Microscopy of PtMn₃/Py Films on SrTiO₃

Detailed HAADF-STEM images of the SrTiO₃/PtMn₃(46.6 nm)/Py(6.8 nm)/SiC sample used for our harmonic Hall effect measurements are shown in Supplementary Figure 4. Overall, the PtMn₃ exhibits high crystallinity persisting through the entire thickness of the thin film. The PtMn₃ film is epitaxially templated to the SrTiO₃, although there is disorder observed at the interface, see Supplementary Figure 4c. We observe a gradient in the manganese composition near the substrate, as discussed in the main text, and in Supplementary Note 1 and Supplementary Figure 2. Additionally, we find that the films have occasional half unit-cell offset dislocations, as well as regions of solid solution face-centered-cubic γ -PtMn₃ with a typical antisite defect of top/bottom face manganese substituted with platinum which is most pronounced near the substrate-film interface, as shown in Supplementary Figure 4c. Antisite defects appear less near the top PtMn₃/Py interface, and we postulate that this is the primary method of strain relaxation due to the substrate lattice mismatch ($\epsilon \sim +1.7\%$) as platinum expands the lattice constant of PtMn₃. On SrTiO₃, the L1₂ ordering fraction for thick (> 50 nm) PtMn₃ films is generally > 0.8 for a variety of temperature (~630-650 °C) and argon pressure setpoints (~10 mTorr, 1 mTorr = 0.133 Pa), as determined by XRD reciprocal space mapping (see Methods). Typically, a post-growth annealing strategy would be employed to improve the ordering fraction and overall crystallinity, as was done in a prior work with PtMn₃ films on BaTiO₃.^[3] Due to the relatively low melting point (~720 °C), and low vapor pressure (~1220 °C) of Mn at high vacuum conditions however, we have found that this leads to the formation of a surface cubic manganese layer with a depth of 2 nm - 5 nm. To reduce possible inhibition of spin current diffusion across the PtMn₃/Py interface from an interfacial manganese layer, the samples

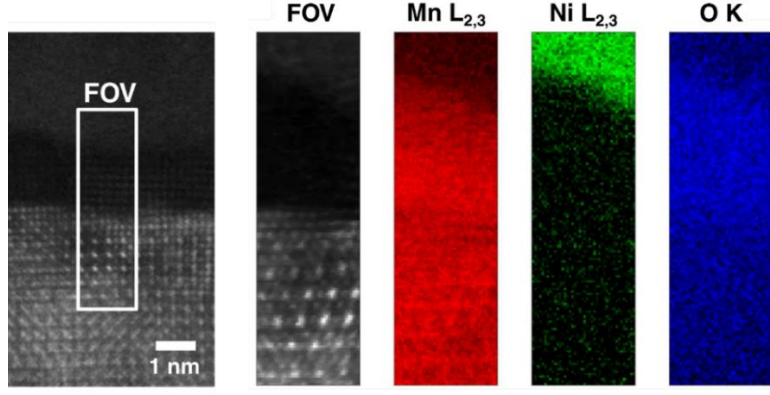
used for the harmonic Hall effect measurements were not subject to a post-deposition anneal. These sample still show discontinuous 1 nm - 2 nm thick surface manganese regions, as shown in Supplementary Figure 4a,b. We note that there is a tradeoff between lowering the growth temperature (and therefore manganese diffusion) and $L1_2$ ordering fraction.

EELS spectroscopic imaging of these surface manganese regions are shown in Supplementary Figure 5. We observe that they are almost exclusively cubic MnO ($T_N \sim 115$ K), and therefore keep all electronic measurements above 120 K to avoid a parasitic antiferromagnetic signal from MnO. As MnO is a good insulator, we expect the effect of these regions on the interfacial spin current diffusion to be minimal, noting that interfacial oxides have been shown to have high interface spin transparency in the kinds of vertical heterostructures employed here^[4]. The source of the oxidized manganese is currently unclear. We acknowledge that this oxidation may happen *ex-situ* during the transfer of the FIB lamella into the STEM. Nevertheless, even if the manganese were to be fully metallic in the as-grown state, we would expect only a fractional suppression of interface spin current, both as per prior studies with continuous 3d transition metal spacers of a similar thickness^[5] and because it is discontinuous.



Supplementary Figure 4 | STEM of PtMn₃/Py film stack for harmonic Hall effect measurements.

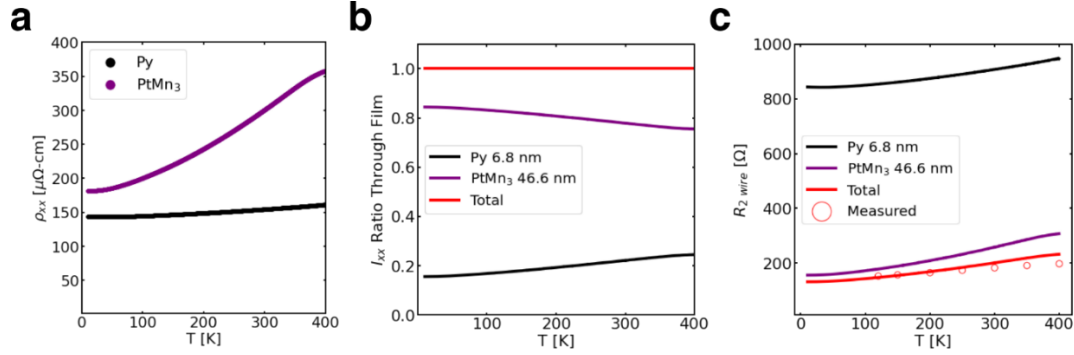
a, Large field-of-view STEM image of SrTiO₃/PtMn₃(46.6 nm)/Py(6.8 nm)/SiC used for harmonic Hall effect measurements. **b**, Top interface between PtMn₃ and Py showing good L₁₂ ordering and interface smoothness. **c**, Bottom PtMn₃ and SrTiO₃ interface, showing increased solid solution phase with typically reduced Mn stoichiometry (~PtMn_{2.0}). We note the presence of MnO_x pockets near the surface, here 1-2 unit cells thick and in disconnected regions.



Supplementary Figure 5 | EELS map of MnO pocket on top PtMn₃ interface. a, EELS map of ~1.5 nm x 6 nm FOV near top interface across MnO cluster from an SrTiO₃/PtMn₃(66 nm)/Py(15 nm) sample showing Mn, Ni and O content. Mn diffusion to the surface is lowered by reducing high temperature exposure and not annealing post-growth. Mn interfacial regions seem to be fully MnO. To avoid MnO antiferromagnetism ($T_N \sim 115$ K), we perform all harmonic Hall effect measurements at $T \geq 120$ K.

Supplementary Note 4. Current Distribution in PtMn₃/Py Heterostructures

We calculate the ratio of total longitudinal current in each film in the SrTiO₃/PtMn₃(46.6 nm)/Py(6.8 nm)/SiC heterostructures by using measured resistivities from isolated films and combining them in a parallel resistor model. Using the calculated resistances for the individual film channels, the ratio of total current in each channel can be estimated, as shown in Supplementary Figure 6b. We calculate the total resistance of the 46 nm PtMn₃ and 6.8 nm Py layers in the Hall channel and combine them in parallel to estimate the 2-wire resistance of the Hall device, as shown in Supplementary Figure 6c. The calculated values of the total device resistance are in excellent agreement with the explicit two wire measurements shown in the same figure (red circles). The respective ratios are used to calculate I_{NM} , I_{FM} at each temperature for the in-plane second Harmonic model and are accounted for in the current-normalized effective fields $h_{DL,FL}^i$ in Oe per 10^7 A cm⁻² (1 Oe = 79.6 A/m).

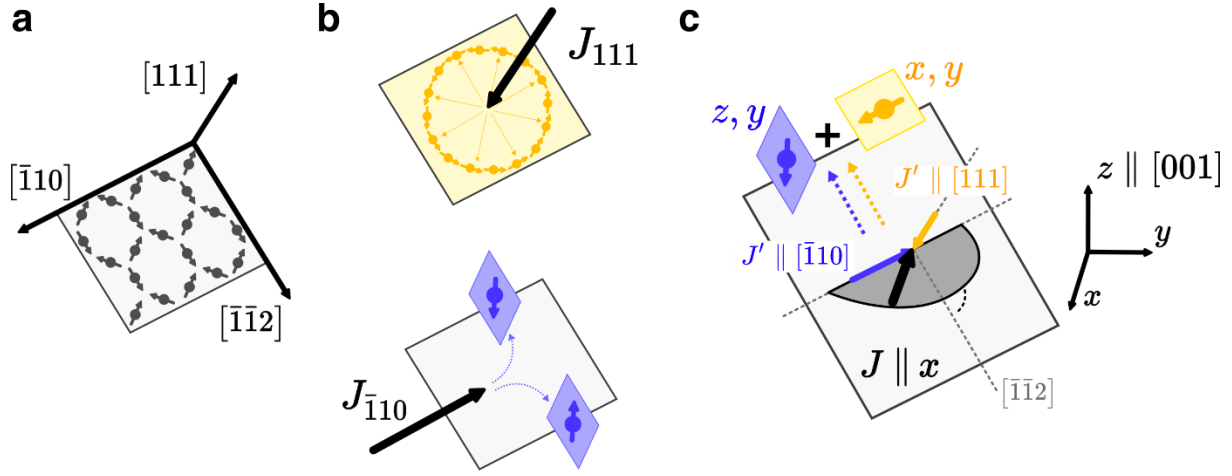


Supplementary Figure 6 | Device resistivity and current splitting in PtMn₃/Py Hall devices. **a**, Resistivities of standalone SrTiO₃/PtMn₃(95 nm) and Al₂O₃/Py(15 nm) films, calculated via longitudinal 4-wire measurements of patterned Hall bar devices. **b**, The resulting current ratio in each film from 10-400 K, assuming a parallel resistor model. **c**, Resistances of each film layer and total Hall channel resistance from 10 K - 400 K in the SrTiO₃/PtMn₃(46.6 nm)/Py(6.8 nm)/SiC devices used for harmonic Hall effect measurements, calculated from resistivity measurements of individual films. The calculated total channel resistance from the 4-wire resistivity scans closely matches the measured 2-wire device resistance (red circles).

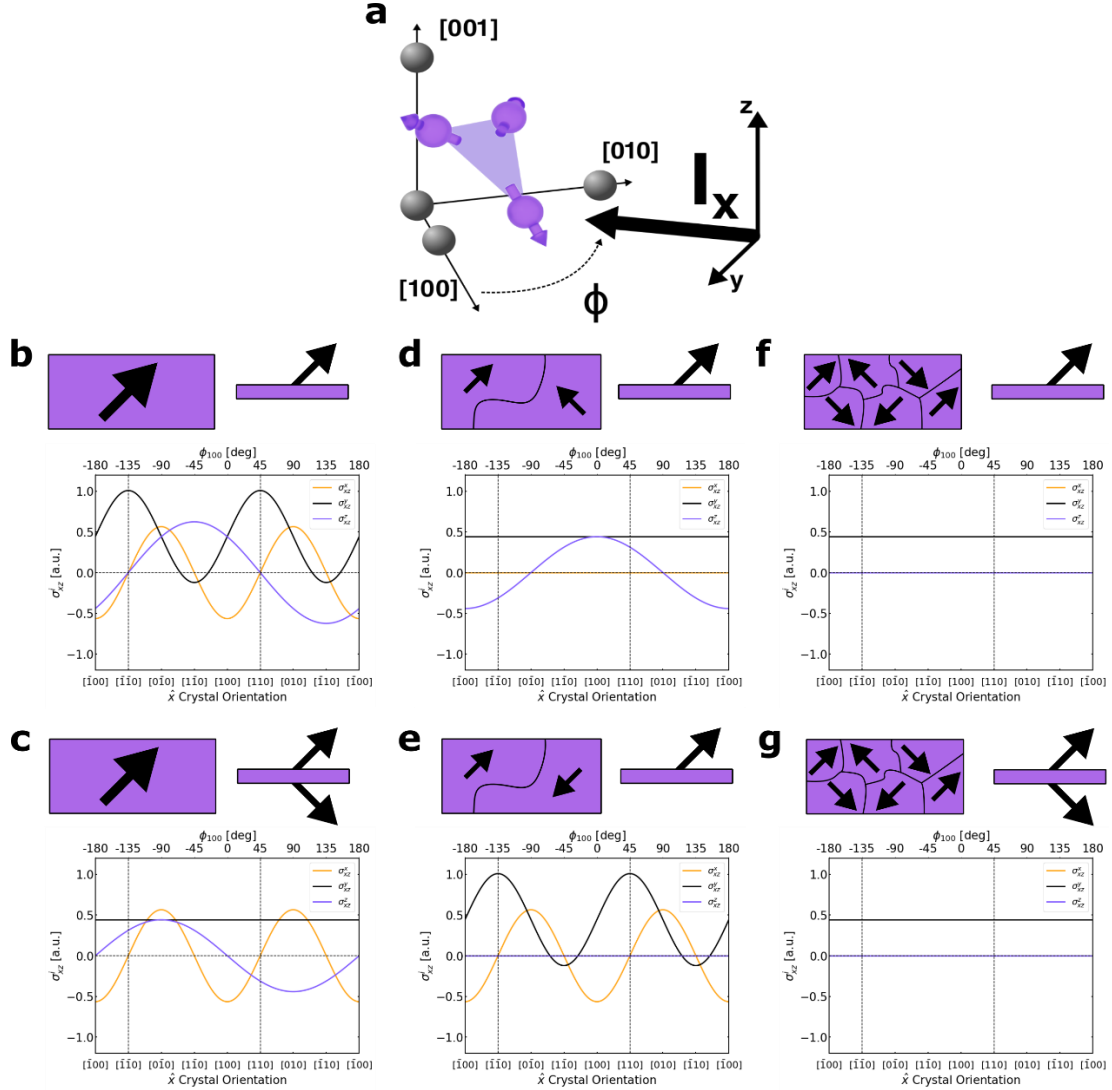
Supplementary Note 5. Intrinsic Spin Conductivity in (001)-oriented PtMn₃

Spin conductivity tensors for the XMn₃ materials (X = Pt, Rh, Ir) have been calculated via density functional theory in prior works in the high symmetry basis where $\hat{x}' = [1\bar{1}0]$, $\hat{y}' = [11\bar{2}]$, $\hat{z}' = [111]$.^[6, 7] The system shows an anisotropy in the spin Hall conductivity that is dependent on charge current incidence to the non-collinear Kagome lattice in the 111 plane. Charge current along [111] creates transverse spin current in the orthogonal plane, equivalent to the conventional spin Hall effect. Charge current within the Kagome lattice creates transverse spin current that has spin polarization orthogonal to the plane, that is nonzero $\hat{p} \parallel \hat{z}'$ (see Fig. 7 in ref. [8]). This behavior is summarized in Supplementary Figure 7a,b. Incident currents partially in and out of the Kagome plane can map to both these conditions. As per

Supplementary Figure 7c, for a current in the 001 plane along direction $\hat{x} \perp [001]$ (all devices in this work) this combined effect can create a composite out-of-plane spin current with $\hat{p} = \hat{x}, \hat{y}, \hat{z}$. To calculate the generated spin currents in some Hall device with coordinates $\hat{x}, \hat{y}, \hat{z}$, as in Supplementary Figure 8a, the applied electric field is projected back into the Hall device basis. For some applied field direction $\hat{E} = e_1\hat{x} + e_2\hat{y} + e_3\hat{z}$, the resulting spin current in the direction $\hat{r} = r_1\hat{x} + r_2\hat{y} + r_3\hat{z}$ with polarization $\hat{p}, p \in (x, y, z)$ is $J_{\hat{E}, \hat{r}}^p = \sum_{p'} (\hat{p} \cdot \hat{p}') [\mathbf{R} \cdot \sigma^{p'} \cdot \mathbf{E}]$, where $\mathbf{R} = \sum_{p'} (\hat{r} \cdot \hat{p}') \hat{p}'$, $\mathbf{E} = \sum_p (\hat{E} \cdot \hat{p}) \hat{p}$ are the projections of the spin current direction and applied electric field into the original basis, and $\sigma^{p'}$ is the spin Hall conductivity tensor for polarization $\hat{p}'$ in the original basis. The $(\hat{p} \cdot \hat{p}')$ term is necessary to account for the difference in spin projection between the two bases. The result for assorted domain degeneracies is shown in Supplementary Figure 8b-g. Generally, increasing the domain degeneracy increases global symmetry and quenches unconventional $\sigma_{xz}^x, \sigma_{xz}^z$, with the fully degenerate case reducing to only the conventional spin Hall effect with isotropic σ_{xz}^y (Supplementary Figure 8g).



Supplementary Figure 7 | Origin of $\hat{x}$, $\hat{z}$ -polarized out-of-plane spin currents in 001 PtMn₃. **a**, The high symmetry basis of the non-collinear phase of L1₂ ordered PtMn₃ ($\hat{x}' = [1\bar{1}0]$, $\hat{y}' = [11\bar{2}]$, $\hat{z}' = [111]$) with the Kagome lattice that exists on the Mn sites in the (111) plane. **b**, Charge currents along $[111]$ result in a conventional spin Hall effect (top) while charge currents within the Kagome plane (bottom) result in transverse in-plane spin current with $\hat{p} = \hat{y}', \hat{z}'$. **c**, For Hall devices of a (001)-oriented film with channel direction $\hat{x}$, the linear combination of these effects is expected to produce an out-of-plane spin current with unconventional $\hat{x}, \hat{z}$ components. The relative amplitude of these components depends on the exact projection of $\hat{x}$ into the Kagome plane, with maximum σ_{xz}^z when $\hat{x} \parallel [\bar{1}10]$, and maximum σ_{xz}^x when $\hat{x} \parallel [100], [010]$. In a real film with multiple antiferromagnetic domains, these amplitudes may be reduced or eliminated entirely, depending on domain size and orientation.



Supplementary Figure 8 | Orientation-dependent spin Hall conductivity in noncollinear PtMn₃ antiferromagnets. **a**, Schematic of noncollinear phase of L1₂ ordered PtMn₃ showing coordinate system for conductivity tensor calculations. **b**, σ_{xz}^i tensor elements for $i = x, y, z$ in the [100], [010], [001] basis for a single AFM domain with Kagome planes with [111] surface normal (black arrow, in-plane projection), along a direction $\hat{x} = [100] + (\cos \phi_{100}, \sin \phi_{100}, 0)$ based on prior DFT results^[6], and rotated into the proper basis. The same for **c** [111], [11 $\bar{1}$], **d** [111], [$\bar{1}$ 11], **e** [111], [$\bar{1}$ $\bar{1}$ 1], **f** [111], [$\bar{1}$ 11], [1 $\bar{1}$ 1], [$\bar{1}$ $\bar{1}$ 1], and **g** all 8 [111]-equivalent domains in equal proportion. In any patterned device, the composition of out-of-plane spin conductivity is highly dependent on both crystal orientation and antiferromagnetic domaining. In general, higher global symmetry reduces or eliminates unconventional tensor elements, with the fully degenerate case in **g** reducing to the conventional isotropic spin Hall effect with only nonzero σ_{xz}^y .

Supplementary Note 6. Harmonic Hall Effect Measurements of PtMn₃/Py

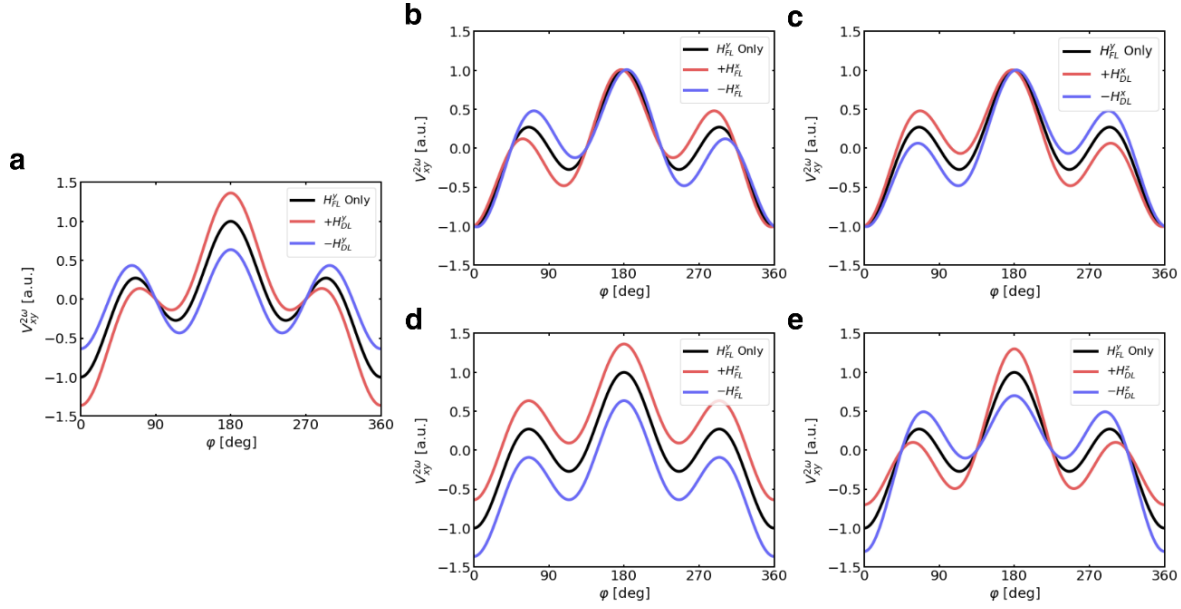
Each σ_{xz}^i component has a distinct effect on the harmonic Hall voltage and can be separated based on symmetry and field dependence. In general, for arbitrary equilibrium orientation, these deflections can have complicated forms due to differences in crystal and shape anisotropy for a given magnet. As is common, we take advantage of the uniaxial in-plane magnetization of thin film Py, and perform second Harmonic scans by orienting the film in-plane ($\theta = \pi/2$) with an external field $H_{ext} \gg H_C, H_a$, where H_C is the in-plane coercive field of the magnet, and H_a is an anisotropy field from patterning. In this regime, the magnet is guaranteed to be saturated in-plane, meaning the equilibrium orientation $\hat{m}(\chi = \frac{\pi}{2}, \varphi) \parallel \mathbf{H}_{ext} = H_{ext}(\cos \varphi \hat{x} + \sin \varphi \hat{y})$. We link this perturbative deflection through the LLGS dynamics by applying a harmonic current $I = I_{total} \sin \omega t \hat{x}$ and measuring the second harmonic Hall voltage $V_{xy}^{2\omega} \propto I_a \nabla_{\theta, \varphi} R_{xy}(\theta, \varphi) \cdot (\Delta\theta, \Delta\varphi)$ which encodes the deflection with high signal-to-noise. The basic form of $V_{xy}^{2\omega}$ has been previously derived in detail^[9, 10, 11, 12], and we review some assumptions behind this model in a prior publication (see Supplementary Note 3 in Ref. ^[13]). This model is readily extended to out-of-plane spin current with $\hat{p} = \hat{x}, \hat{y}, \hat{z}$ (see main text Equation 1). As discussed in the main text, we adopt this current-density based model for the spin torque fields because of the typically small spin diffusion lengths in metals (1 nm - 10 nm)^[14], meaning only the PtMn₃ close to the top interface is contributing. In our measurements, $J_{SHM} < 5 \cdot 10^6$ A/cm² to reduce thermal drift and the baseline thermoelectric contribution. This results in $h_{oe} < 30$ Oe per 10^7 A/cm² for the SrTiO₃/PtMn₃(46.6 nm)/Py(6.8 nm) devices.

Because of the different symmetry and field dependence of all the individual spin torque effective fields, the presence of most components can be visually identified in the $V_{xy}^{2\omega}(\varphi, H_{ext})$ data if they are of sufficient magnitude. To provide a useful visual reference, we plot the addition

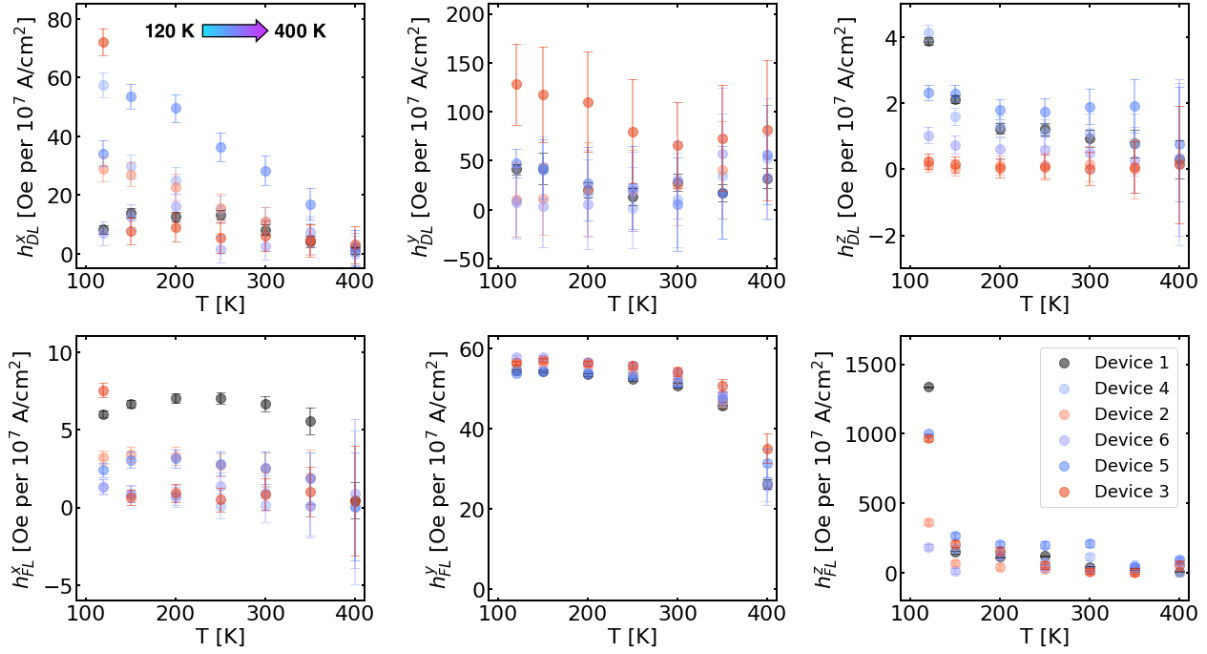
of positive/negative components of these fields to a basic response from a $\hat{p} = \hat{y}$ or Oersted-like field in Supplementary Figure 9. Of the six components, all but the $H_{FL,DL}^x$ are uniquely identifiable due to a distinct effect on the harmonic profile (satellite peak amplitude change, vertical shift, vertical asymmetry). If high precision measurements of $H_{FL,DL}^x$ are desired, then this geometry may not be optimal due to the similarity of the perturbative effect from the field and damping-like torques, as shown in Supplementary Figure 9b-c. In general, care must be taken to fit to visually significant amplitudes and perform a thorough parameter error analysis, as was done here with the DREAM algorithm through the Bumps python package.

The $h_{DL,FL}^i$ for all the SrTiO₃/PtMn₃(46.6 nm)/Py(6.8 nm) devices in this study for each temperature step between 120 K - 400 K are shown in Supplementary Figure 10. As discussed in the main text, the behavior of the field-dependent $R_{xy}^{2\omega}$ offset, attributed to h_{FL}^z , is anomalously large (10-100x higher than h_{FL}^y) and does not generally follow the expected linear behavior with $1/(H_{ext} + H_S)$. Though the exact origin is undetermined, we speculate that the $R_{xy}^{2\omega}$ may be complicated by either an additional thermoelectric contribution from any anomalous spin textures near the interface, or by a spin polarization of the charge current from field-induced canting of the PtMn₃. From the VSM data in main text Figure 2b-c, we see no evidence for an anomalous ferromagnetic component e.g. from a spin glass layer formed by exchange interaction with the PtMn₃. Further, any spin torques would still be expected to follow $1/H_{ext}$, $1/(H_{ext} + H_S)$ field dependencies. There is also a general over-fitting concern with the $h_{DL,FL}^x$ components due to their near identical but opposite polarity contributions to $R_{xy}^{2\omega}$ as per Supplementary Figure 9b-c. For the device highlighted in main text Figure 4 the evolution of the effective field components as the temperature is increased is smooth, as shown in Supplementary Figure 10 for the largest components with $1/H_{ext}$ scaling. It is likely that some amplitude is exchanged

between the two components during the fitting procedure, and that this is responsible for the low-temperature discrepancy between $h_{DL,FL}^x$ amplitudes, as in Supplementary Figure 10a,d.

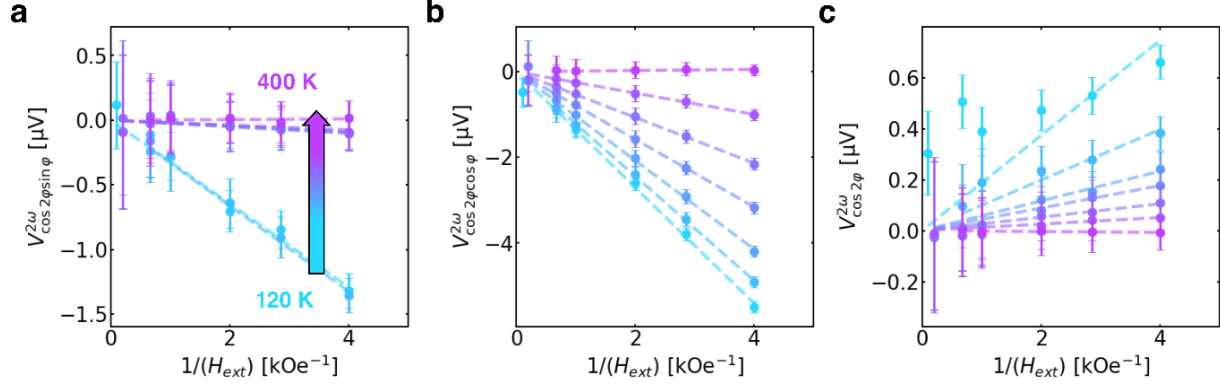


Supplementary Figure 9 | Second harmonic dependence on spin torque effective fields from $\hat{x}, \hat{y}, \hat{z}$ polarized spin currents. **a**, Typical $V_{xy}^{2\omega}(\phi)$ profile for only H_{FL}^y (black, in all subfigures) from SOT or ASOT, and effect of $\pm H_{DL}^y$ (red/blue) from some σ_{xz}^y . **b**, Effect of $\pm H_{FL}^x$ (ϕ asymmetry around 180°), and **c**, $\pm H_{DL}^x$ (opposite ϕ asymmetry around 180°). **d**, Effect of $\pm H_{FL}^z$ (voltage asymmetry), and **e**, $\pm H_{DL}^z$ (voltage shift). Except for $H_{DL,FL}^x$, which could be easily confused if not fit for precisely, any $\hat{p} = \hat{y}, \hat{z}$ spin currents of significant amplitude injected into the Py layer should be visibly apparent based on their distinct effects on $V_{xy}^{2\omega}$.

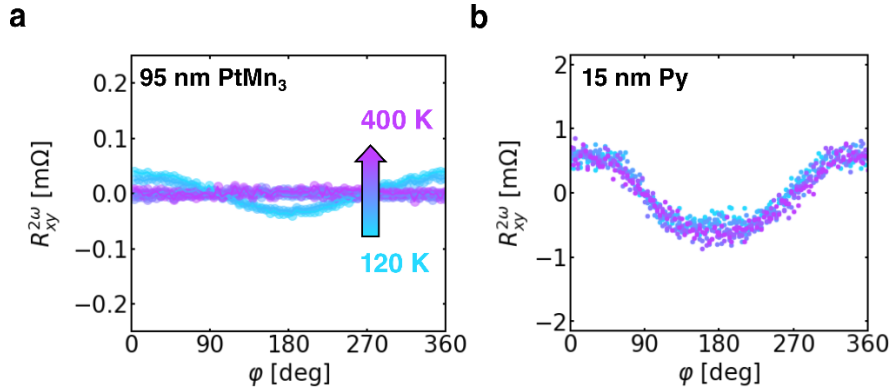


Supplementary Figure 10 | Spin torque effective field components in PtMn₃/Py vs. Temperature.

Extracted spin torque effective fields from the in-plane second harmonic Hall effect measurements of the SrTiO₃/PtMn₃(46.6 nm)/Py(6.8 nm)/SiC devices for damping-like torques from **a**, $\hat{x}$, **b**, $\hat{y}$, and **c**, $\hat{z}$ out-of-plane spin currents, and for the field-like torques from **d**, $\hat{x}$, **e**, $\hat{y}$, and **f**, $\hat{z}$ out-of-plane spin currents (error bars represent 2 standard deviations as reported by the DREAM parameter estimation). In general, the devices exhibit a typical spin Hall effect-like σ_{xz}^y that follows the Néel transition and has an amplitude exceeding typical Pt/Py devices. Components relating to σ_{xz}^x , σ_{xz}^z arise below the first-order transition at 200 K - 250 K, and the σ_{xz}^y diverges as well, which can be attributed to the anisotropic spin Hall conductivity expected in the non-collinear AFM phase.



Supplementary Figure 11 | Inverse field dependence of second harmonic components vs. temperature in PtMn₃/Py. Components from the second harmonic Hall effect signal $V_{xy}^{2\omega}$ from the SrTiO₃/PtMn₃(46.6 nm)/Py(6.8 nm)/SiC device highlighted in main text Figure 4, plotted as a function of inverse external field and temperature corresponding to the **a**, field-like $\hat{x}$ torque, **b**, field-like $\hat{y}$ torque, and **c**, the damping-like $\hat{z}$ torque. At all temperatures these components are visibly linear in $1/H_{ext}$. Compared to the conventional $\hat{y}$ component, the unconventional $\hat{x}$ and $\hat{z}$ components come on more sharply below 200 K, the apparent first-order transition point into the low temperature non-collinear phase.



Supplementary Figure 12 | In-plane harmonic Hall effect control measurements of isolated films. In-plane second harmonic Hall resistance $R_{xy}^{2\omega}$ from 120-400 K for patterned Hall devices of **a**, SrTiO₃/PtMn₃(95 nm)/SiC(20 nm) showing no intrinsic $R_{xy}^{2\omega}$ response except for a slight thermoelectric component $\sim \cos \phi$ below 150 K - 200 K attributed to canting of the noncollinear antiferromagnetic lattice and **b**, Al₂O₃/Py(15 nm)/TiO₂ with $H_{ext} = 250$ Oe showing a consistent thermoelectric component across the full temperature range. Compared to Py, the PtMn₃ self-response at low temperature is approximately 20x lower and is ignored in the bilayer model.

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