
Supplementary information

**Octupole-driven spin torque switching of
antiferromagnetic tunnel junctions**

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SUPPLEMENTARY INFORMATION

Octupole-driven spin torque switching of antiferromagnetic tunnel junctions

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Supplementary Note 1. Structural and magnetic characterization of PtMn₃ films

To determine the structural and magnetic properties of our PtMn₃ films, we performed X-ray diffraction (XRD) and superconducting quantum interference device (SQUID) magnetometry measurements on the AATJ stack. In the XRD data, shown in Fig. S1, only the Pt (111) and PtMn₃ (111) peaks are observed. This is consistent with the highly textured films observed in the TEM measurements (main text, Fig. 1d) and does not show any evidence for the existence of other Pt-Mn phases. SQUID magnetometry data on the full AATJ stack are shown in Fig. S2, where the magnetic field is swept along both the in-plane and out-of-plane directions up to a magnitude of 7 T. The data show no net magnetization in either direction, indicating the fully compensated antiferromagnetic phase of PtMn₃. In addition, to determine the actual stoichiometry of the PtMn₃ films, X-ray fluorescence (XRF) measurements were performed. The Pt composition was found to range from 23.9 to 25.8 atomic percent (at%) at multiple positions across the 150 mm silicon wafer, which is close to the targeted Pt₂₅Mn₇₅ composition. These results are illustrated in Fig. S3.

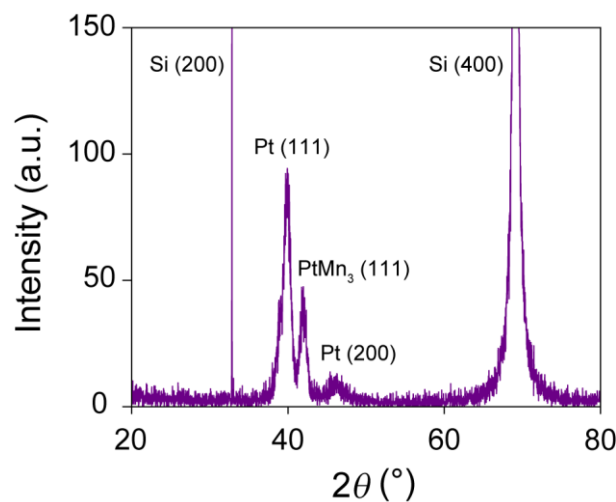


Figure S1| X-ray diffraction measurements. XRD measurements on full Si / SiO₂ / Pt (5 nm) / PtMn₃ (7 nm) / MgO (2 nm) / PtMn₃ (10 nm) / Pt (5 nm) / Ru (10 nm) AATJ material stack.

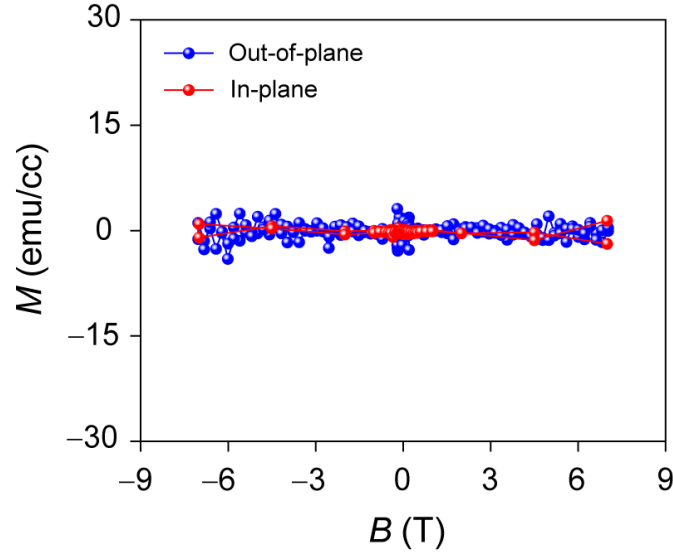


Figure S2| SQUID magnetometry measurements. Magnetization measurements on full Si / SiO₂ / Pt (5 nm) / PtMn₃ (7 nm) / MgO (2 nm) / PtMn₃ (10 nm) / Pt (5 nm) / Ru (10 nm) AATJ material stack, conducted using a superconducting quantum interference device by sweeping the external magnetic field along the in-plane (red) and out-of-plane (blue) directions. The curves were obtained after subtracting the diamagnetic linear background.

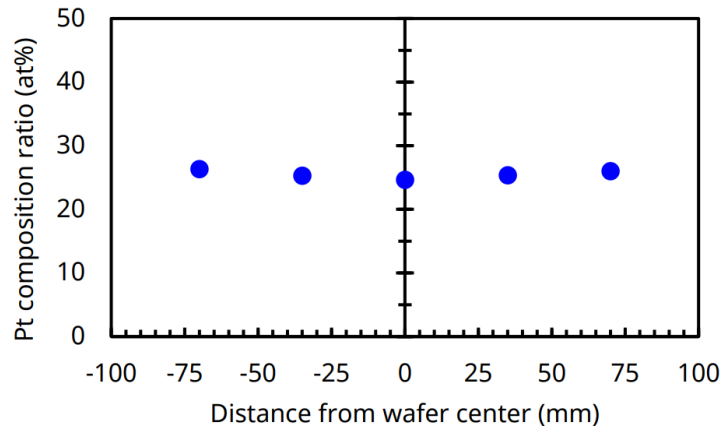


Figure S3| Composition of PtMn₃ across different positions on a 150 mm wafer. The composition was measured by X-ray fluorescence, indicating a uniform stoichiometry.

Supplementary Note 2. Comparison of PtMn₃ (001) and (111) surface terminations

To estimate the intrinsic domain size in sputter-deposited PtMn₃ using NV magnetometry, we used a PtMn₃ (001) film to obtain an uncompensated surface termination with finite stray field. Using the PtMn₃ (111) surface for this purpose was not possible, since it is fully compensated. A comparison of these two surfaces of PtMn₃ is shown in Fig. S4.

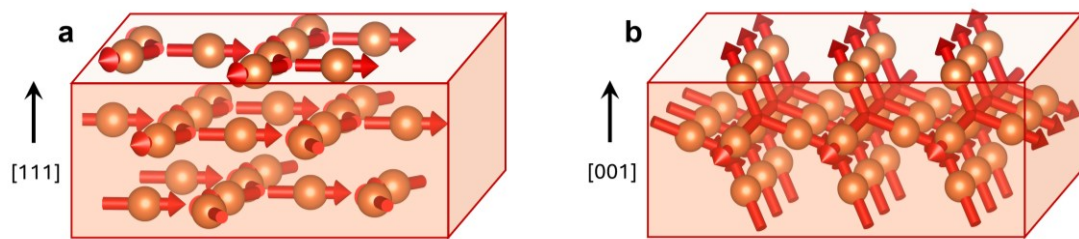


Figure S4| Comparison of (001) and (111) surface terminations of PtMn₃. Only the (001) surface has an uncompensated magnetic moment, which allows its domain structure to be detected in NV microscopy measurements.

Supplementary Note 3. Role of amorphous MgO in TMR of AATJ devices

In crystalline MgO, the barrier is translationally symmetric in-plane, so tunneling is specular, resulting in the conservation of the transverse electron momentum $\mathbf{k}_{\parallel}$. In amorphous MgO, this symmetry is lost. Random fluctuations in atomic positions create local variations of the barrier height experienced by tunneling electrons, generating a scattering potential within the barrier. As a result, tunneling becomes diffusive and the transverse momentum is no longer conserved, $\mathbf{k}_{\parallel} \neq \mathbf{k}'_{\parallel}$. Typically, however, these fluctuations are small compared to the average barrier height, $\Delta \ll V_0$. Specifically, for Fe/MgO/Fe tunnel junctions, the effective barrier height is typically $V_0 \sim 2\text{--}3\text{ eV}$, as inferred from ab-initio band alignment and experimental RA-vs-thickness fits for Fe/MgO/Fe MTJs. The same magnitude of V_0 is expected in PtMn₃/MgO/PtMn₃ junctions. On the other hand, the intrinsic fluctuations of the conduction band minimum (CBM), Δ , in an amorphous oxide layer, as follows from typical magnitudes of band-edge Urbach tail energies in amorphous oxides are of the order of $\Delta \sim 0.3\text{--}0.4\text{ eV}$ [1]. Thus, $\frac{\Delta}{V_0} \sim 0.1\text{--}0.2$, can be considered as a small perturbative parameter.

This physically relevant limit justifies using the Born approximation: fluctuations of the barrier height perturb the evanescent MgO barrier only weakly, allowing tunneling to be treated as a small single-scattering correction on top of the clean barrier. In this case, transmission can then be written as a sum of a coherent and a diffusive part:

$$T(\mathbf{k} \rightarrow \mathbf{k}') = T_0(\mathbf{k}_{\parallel}) \delta_{\mathbf{k}_{\parallel}, \mathbf{k}'_{\parallel}} + \delta T_{\text{diff}}(\mathbf{k} \rightarrow \mathbf{k}').$$

The first term is the usual specular, $\mathbf{k}_{\parallel}$ -conserving tunneling through a clean MgO barrier. The second term describes k -mixing due to disorder. For Gaussian-correlated barrier-height fluctuations with correlation length, ξ , the diffusive contribution can be shown to have the form

$$\delta T_{\text{diff}}(\mathbf{k} \rightarrow \mathbf{k}') \propto \Delta^2 \xi^3 L e^{-\frac{\xi^2}{2} |\mathbf{k}'_{\parallel} - \mathbf{k}_{\parallel}|^2},$$

where Δ is the RMS amplitude of barrier-height fluctuations, ξ is their correlation length, and L is the barrier thickness.

For an amorphous MgO oxide, it is reasonable to take ξ to be of the order of half a lattice parameter, i.e., $\xi \sim 2 \text{ \AA}$. Then the k -space width of the Gaussian is $\sim \frac{1}{\xi} \approx 0.5 \text{ \AA}^{-1}$, which is a substantial fraction of the in-plane Brillouin-zone. In other words, an incoming $\mathbf{k}_{\parallel}$ is scattered into a fairly broad range of $\mathbf{k}'_{\parallel}$.

However, the integrated weight of this broadened, diffuse contribution is controlled by the small ratio Δ/V_0 . Within the Born approximation, integration of $\delta T(\mathbf{k} \rightarrow \mathbf{k}')$ over all final $\mathbf{k}'_{\parallel}$, the diffuse weight scales as

$$\frac{\delta T_{\text{diff}}}{T_0} \sim \kappa_0^2 L \xi \left(\frac{\Delta}{V_0} \right)^2,$$

where κ_0 is the decay rate in MgO, which is $\kappa_0 \sim 0.3 \text{ \AA}^{-1}$. Now, taking into account that $\left(\frac{\Delta}{V_0} \right)^2 \sim 0.01\text{--}0.04$ and $L \sim 20 \text{ \AA}$, we obtain:

$$\frac{\delta T_{\text{diff}}}{T_0} \sim 0.03 - 0.12.$$

We see therefore that only a few percent of the total transmitted weight reside in the diffusive transmission, while the dominant part remains coherent and nearly specular.

Supplementary Note 4. Temperature-dependent switching measurements for additional AATJ device

For all tested devices, there was a complete disappearance of TMR and OTT switching at temperatures ranging from 360 K to 400 K. This process is reversible, i.e., the OTT-induced switching and TMR reappear upon return to room temperature, when the material goes back to its noncollinear order. Figure S5 shows a series of switching curves measured on an AATJ device starting from 300 K (blue circles), gradually increasing to 400 K (red circles) and back to 300 K (green circles). The results show that OTT switching behavior and associated TMR are absent at 360 K and 400 K, and reappear upon return to 300 K. We note that slight changes in the switching current, resistance values and associated TMR were observed in the recovered curve at 300 K. We attribute this to different local domain configurations formed during the heating and subsequent cooling process, as well as to a potential partial shorting of the device during the high-temperature measurements.

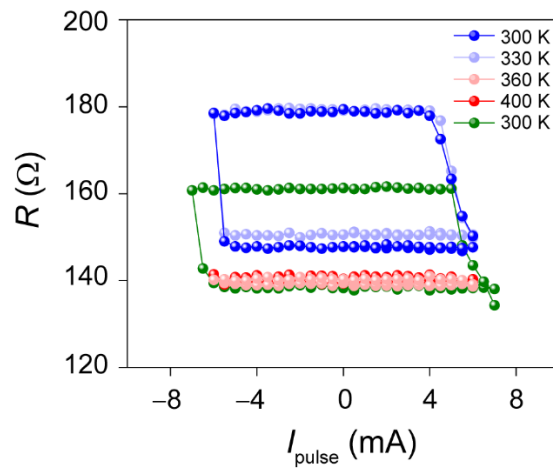


Figure S5| Temperature-dependent current-induced switching measurements. The switching measurements were performed for a temperature sequence starting from 300 K to 400 K and back to 300 K. Current-induced switching reappears upon cooling to room temperature, consistent with the octupolar origin of the OTT and TMR.

Supplementary Note 5. Comparison of two- and three-terminal switching measurements

We performed additional experiments to verify that spin-orbit torque (SOT) from the Pt seed layer is not responsible for the current-induced switching observed in our devices. Figure S6a shows a representative scanning electron microscope (SEM) image of the AATJ devices used for this experiment, prior to the top electrode deposition. The AATJ pillar is located at the center of the narrow Pt channel whose width is slightly larger than the diameter of the pillar. This design allows one to directly compare OTT and SOT switching in the same device, under an identical current density flowing through the Pt layer. Figure S6b shows the current-induced switching experiments in the OTT geometry (two-terminal, with write current flowing vertically through the AATJ) and the SOT geometry (three-terminal, with write current flowing horizontally through the Pt channel). Both OTT and SOT traces are measured in the same device with a pillar diameter of 100 nm. In both cases, the tunneling resistance was measured with a small reading current after each current-induced switching attempt. The results show OTT switching at a current pulse amplitude of approximately 2 mA, whereas switching is absent up to 4 mA of in-plane current in the SOT geometry. These results indicate that the observed switching signatures in the two-terminal OTT geometry do not originate from SOT.

Furthermore, we performed SOT and OTT switching experiments in a second device, after field-cooling the AATJ from 400 K to 300 K under the out-of-plane external magnetic field of ± 0.2 T. The results of this experiment are summarized in Fig. S7. First, we do not observe SOT switching within the applied in-plane current range, consistent with the data shown in Fig. S6. Second, the polarity of the two-terminal OTT switching curves obtained under different field-cooling conditions remains the same, as would be expected from a spin-transfer torque-based switching mechanism [2].

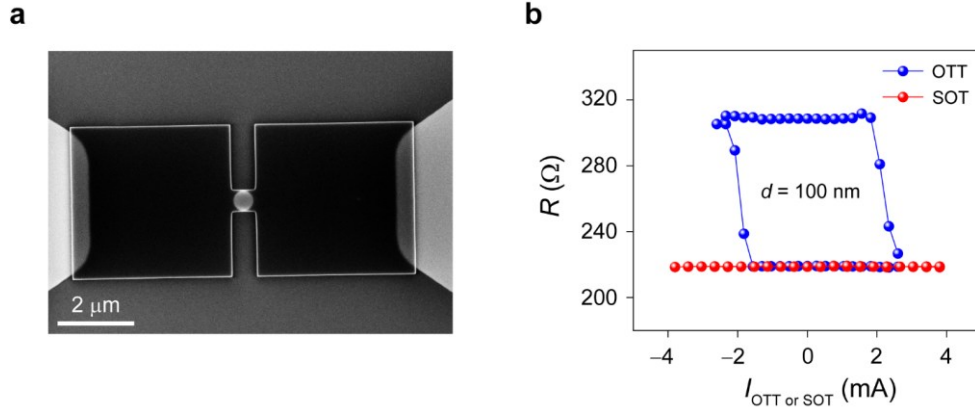


Figure S6| Comparison of two- and three-terminal switching measurements in AATJs. a, A representative SEM image of the AATJ devices used for this experiment, prior to the top electrode deposition. The AATJ pillar is located at the center of the narrow Pt channel whose width is slightly larger than the diameter of the pillar. **b,** Three-terminal SOT and two-terminal OTT switching measurements performed within the same device with a 100 nm pillar diameter. The blue and red curves indicate currents applied vertically through the AATJ, and horizontally through the underlying Pt channel, respectively. The absence of switching, even for in-plane currents that are $\sim 2\times$ larger than the observed OTT switching current, confirms that SOT is not the primary mechanism of the observed two-terminal switching signatures.

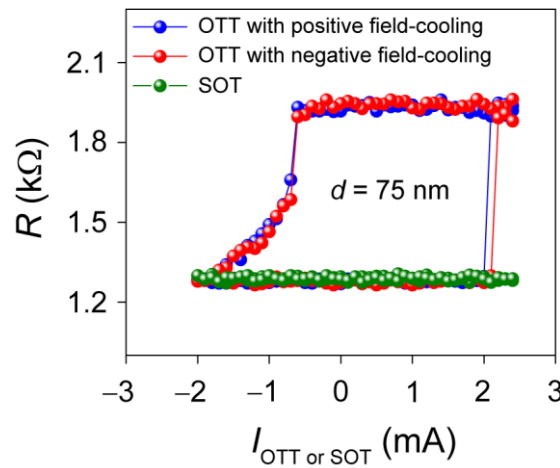


Figure S7| Comparison of two- and three-terminal switching measurements of an AATJ under different field-cooling conditions. Three-terminal SOT and two-terminal OTT

switching measurements performed within the same device with a 75 nm pillar diameter. Prior to the OTT switching, the AATJ was field-cooled from 400 K to 300 K under an out-of-plane external magnetic field of ± 0.2 T, respectively. The blue and red curves indicate write currents applied vertically through the AATJ (i.e., OTT geometry) while the green curve indicates write currents applied horizontally through the underlying Pt channel (i.e., SOT geometry). All measurements were conducted at room temperature.

Supplementary Note 6. Micromagnetic simulations of switching in the presence of domains

To account for domain nucleation, domain wall motion, and their size dependence in AATJs, we developed a micromagnetic model while retaining the same magnetic parameters as those used for the macrospin model of the main text, which was built upon our earlier-developed two-sublattice model [3]. Within the micromagnetic approach, the antiferromagnet is discretized in cuboid cells, each containing three sublattice magnetizations. The magnetization dynamics for each cell are given by three coupled LLG equations, one per each sublattice ($\vec{m}_1$, $\vec{m}_2$, $\vec{m}_3$), as described by Eq. (9) in the main text. Formally, this model is the same as the macrospin model. However, here the effective field, H_{eff} , includes the exchange contribution arising from the coupling between the same sublattices in neighboring cells. This term, equivalent to ferromagnetic exchange, promotes parallel alignment of the spins of one sublattice in different points and is expressed as $\vec{H}_{\text{exch}}^0 = 2A_0 \nabla \vec{m}_i / M_{s,i}$. We have considered each sublattice equivalent and characterized by the exchange constant $A_0 = 5$ pJ/m, even though we verified that there are no significant changes in the results for a wide range of values from 2.5 to 10 pJ/m.

We studied the OTT dynamics for three pillar diameters (50, 100, 200 nm) at a temperature of 300 K. We modeled the free layer directly and considered the effect of the polarizer layer (i.e., fixed PtMn₃ electrode) via the corresponding sublattice polarizations entering the OTT equations, as presented in Eq. (9) of the main text. For each diameter, we first relaxed the magnetic configuration starting from a head-to-head domain configuration along each sublattice anisotropy direction of the Kagome arrangement. We found that for small diameters (50 nm) the magnetic texture remains uniform, while for 100 and 200 nm diameters, domains are nucleated. These are shown in Fig. S8a and Fig. S8b for the diameters of 100 and 200 nm, respectively.

We then investigated the OTT dynamics by applying a 50 ps square-shaped current pulse, and letting the system relax for another 10 ps, in the following three cases:

- I) Both the polarizer and free layer are uniformly magnetized.
- II) The polarizer is uniform, while the relaxed non-uniform states are used for the free layer.
- III) The free layer is uniform, while the relaxed non-uniform states are used for the polarizer.

Case I. The switching is always coherent, independent of the pillar diameter. This case is adequately captured by a macrospin model.

Case II. The switching always yields a uniform final state within the pulse duration (50 ps). This is shown in Fig. S8c and Fig. S8d, where we present the time-dependent x , y and z components of the sublattice magnetization $\vec{m}_3$ for 100 and 200 nm pillars, respectively, and in Fig. S8e and Fig. S8f, where the snapshots at different times are depicted. The magnetizations of the other two sub-lattices are rotated by 120° in the plane. There is an initial domain wall propagation in response to the current, which ultimately leads to a uniform state in the free layer, following the uniform configuration of the polarizer.

Case III. The non-uniform magnetic configurations, depicted in Fig. S9a and Fig. S9b, are now utilized to describe the non-uniform sublattice-resolved spin polarization that exhibits a strong dependence on the pillar diameter. The free layer magnetization, however, is assumed to be initially uniform. For the 100 nm case, the free layer magnetization evolves via coherent rotation, as shown in Fig. S9c and Fig. S9e. Once the current is turned off, the magnetization of each sublattice relaxes towards the equilibrium in-plane Kagome configuration while staying relatively uniform.

For the larger 200 nm diameter case, the dynamics are more complex, as shown in Fig. S9d. In Fig. S9b, we can see regions with different in-plane and out-of-plane directions of the

polarization for each sublattice. Hence, there will be regions where the free layer anisotropy directions for each sublattice and the respective sublattice-dependent spin polarization direction are aligned, and others where they are misaligned. Therefore, the current pulse results in the formation of domains, and domain wall-mediated dynamics associated with a rotation of the magnetic texture induced by the OTT, as observed in Fig. S9f. In this case, we can expect a reduction in TMR due to the partial misalignment of the free layer sublattice magnetizations with respect to those of the polarizer.

Therefore, our results indicate that in general, AATJs with larger pillar diameters will be characterized by non-uniform magnetic textures and non-uniform magnetization dynamics, which can reduce the effective TMR compared to AATJs with uniformly magnetized free and fixed layers.

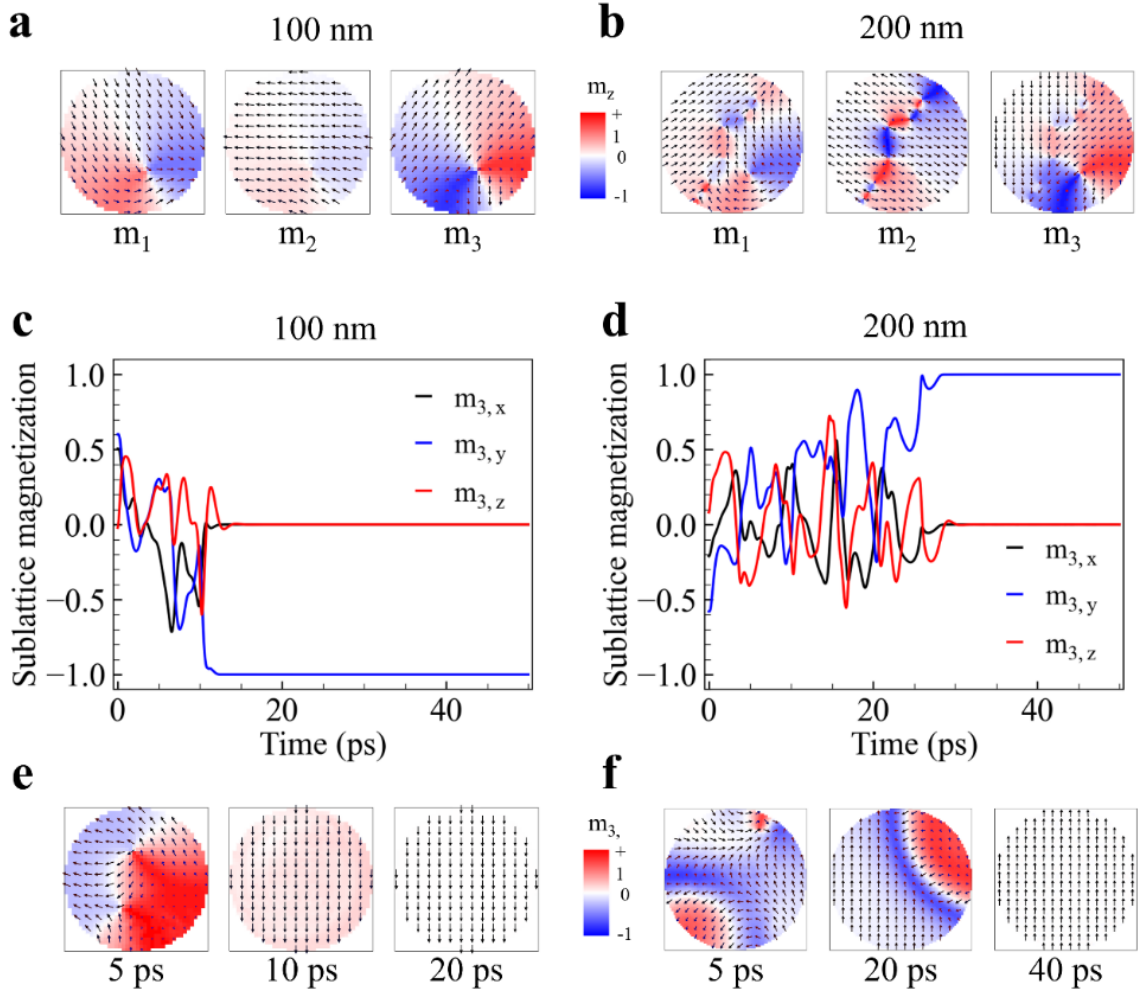


Figure S8| Micromagnetic simulations of the size dependence of OTT dynamics in the case of non-uniform free layer magnetization. a, b, Snapshot of the equilibrium magnetic configuration of the free layer for pillars with diameter of 100 nm (**a**), and 200 nm (**b**). **c, d,** Time-dependent x, y and z components of the magnetization of sublattice 3 in response to a 50 ps current pulse for pillars with diameter of 100 nm (**c**), and 200 nm (**d**). **e, f,** Snapshots taken at different times during the switching dynamics of sublattice 3 for pillars with diameter of 100 nm (**e**), and 200 nm (**f**).

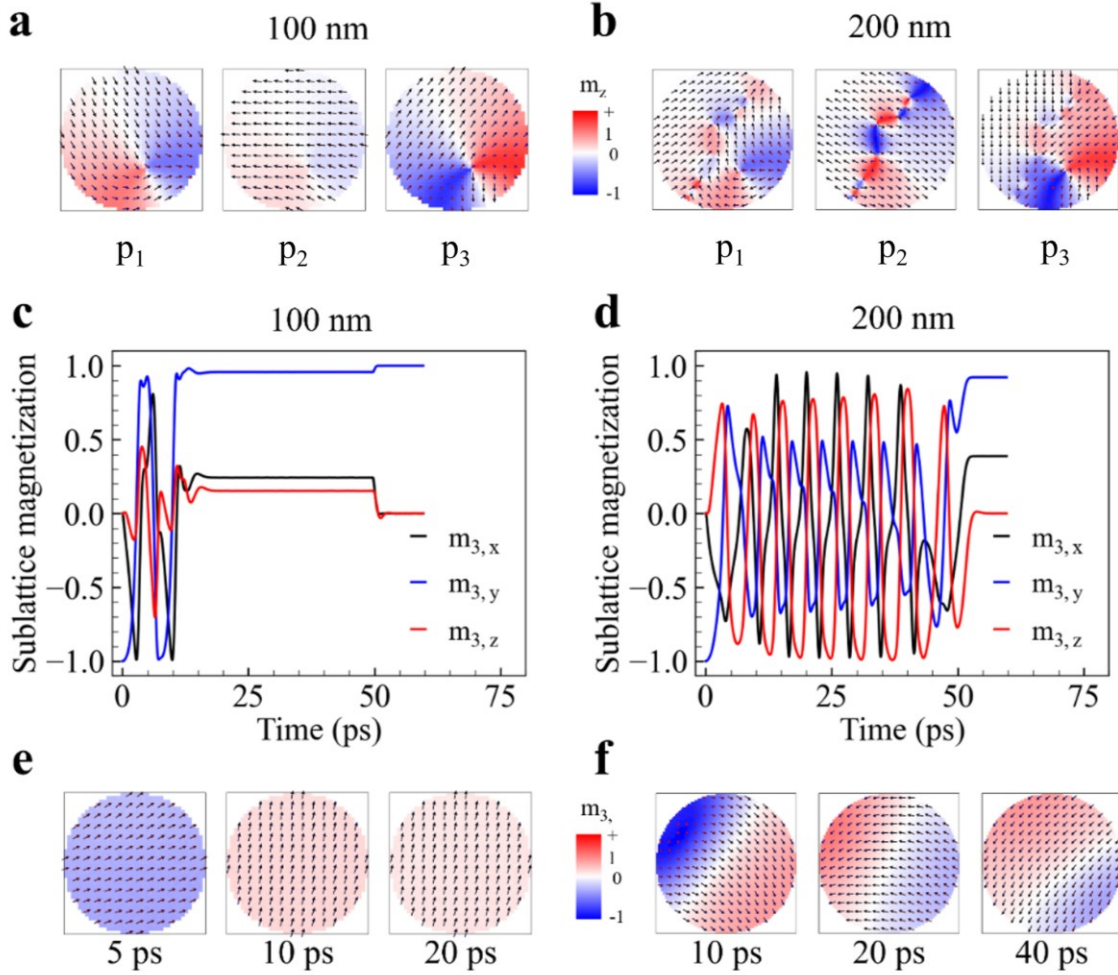


Figure S9| Micromagnetic simulations of the size dependence of OTT dynamics in the case of non-uniform fixed layer polarization. **a, b**, Snapshot of the current-induced polarization directions used for pillars of diameter 100 nm (**a**), and 200 nm (**b**). **c, d**, Here, we have used the same relaxed states as in Fig. S8, but assumed them to represent the equilibrium state of the polarizer. Time-dependent x, y and z components of the magnetization of sublattice 3 of the free layer in response to a 50 ps current pulse for pillars with diameter of 100 nm (**c**), and 200 nm (**d**). **e, f**, Snapshots taken at different times during the switching dynamics of sublattice 3 of the free layer for pillars with diameter of 100 nm (**e**), and 200 nm (**f**). For all the snapshots the palette describes the magnitude of the z -component and the arrows represent the in-plane components.

Supplementary Note 7. Anomalous Hall effect measurements

To investigate the magnetic-field-induced transport response, we performed anomalous Hall effect (AHE) measurements on two sets of devices, the results of which are shown in Fig. S10:

1. **Poly-crystalline PtMn₃:** Full material stacks of Si / SiO₂ / Pt (5 nm) / PtMn₃ (7 nm) / MgO (2 nm) / PtMn₃ (10 nm) / Pt (5 nm) / Ru (10 nm), nominally identical to the AATJ material stacks of the main manuscript, were patterned into Hall-bar structures with a width of 10 μm . We did not observe an AHE signal in these samples, as shown by the blue data points in Fig. S10b. This result is consistent with the absence of a net magnetic moment in this stack, as confirmed by SQUID magnetometry measurements, indicated by blue data points in Fig. S10a.
2. **Epitaxial PtMn₃:** We deposited epitaxial PtMn₃-based stacks of Pt (7 nm) / PtMn₃ (30 nm) on an Al₂O₃ (0001) substrate. The PtMn₃ layer was capped by a layer of AlO_x (2 nm) for protection. The Pt layer was grown at the substrate temperature of 500°C, followed by post-annealing at 850°C for one hour to promote epitaxial growth. Subsequently, PtMn₃ was deposited at the substrate temperature of 550°C. To examine the crystallinity of the material stacks, we performed X-ray diffraction (XRD) as shown in Fig. S10c. The XRD measurement shows a clear peak at $2\theta = 40.2^\circ$ whose asymmetric shape indicates the overlap of the Pt (111) and PtMn₃ (111) peaks. The red data points in Fig. S10b show the AHE signal for this sample. The presence of AHE is consistent with the presence of a small magnetic moment, as confirmed by SQUID measurements, indicated by red data points in Fig. S10a.

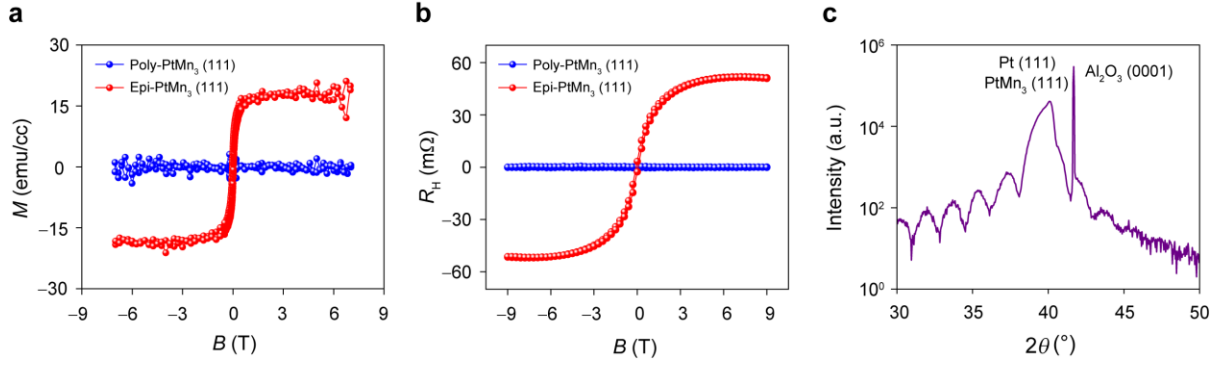


Figure S10| Anomalous Hall measurements in poly-crystalline and epitaxial PtMn₃ (111) films. **a**, SQUID magnetometry results on poly-crystalline (blue) and epitaxial (red) PtMn₃ films. **b**, AHE measurements on Hall bars made from the same samples, indicating a non-zero response in the epitaxial PtMn₃ case. **c**, XRD measurements on the epitaxial PtMn₃ sample, indicating the PtMn₃ (111) peak as well as the Al₂O₃ (0001) peak from the substrate.

Supplementary Note 8. Two-terminal current-induced switching under external magnetic fields

To demonstrate the immunity of the OTT-controlled AATJs to external fields, we performed two-terminal current-induced switching measurements under three different out-of-plane externally applied magnetic fields (B_z). Figure S11 presents the switching results when $B_z = +2$, 0 and -2 T. All three curves exhibit similar switching behavior without any significant changes in switching current values or TMR, indicating the resilience of the OTT switching against external magnetic fields.

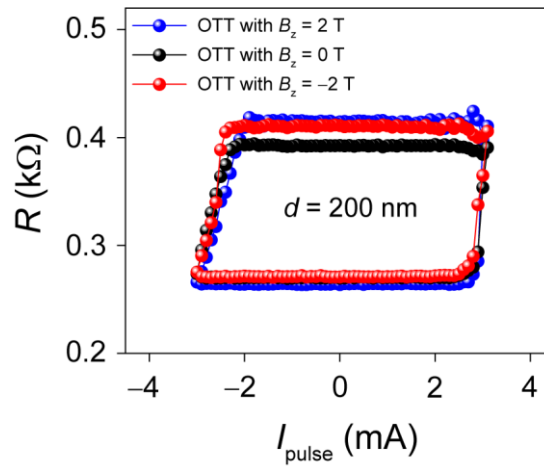


Figure S11| Two-terminal current-induced switching measurements of an AATJ under different magnetic fields. Out-of-plane external magnetic fields of +2, 0, -2 T were applied during the switching measurements on a 200 nm-diameter AATJ at room temperature, corresponding to the blue, black and red curves, respectively.

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