

# Octupole-driven spin torque switching of antiferromagnetic tunnel junctions

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**This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.**

Version 0:

Reviewer comments:

Referee #1

(Remarks to the Author)

The discovery of the giant magnetoresistance (GMR) effect in the late 1980s significantly increased information storage density in hard disk drives (HDDs) due to the GMR HDD read heads applications in the 1990s, a breakthrough that was later honored with the Nobel Prize. Later in 2004, the development of HDD read heads based on the tunnel magnetoresistance (TMR) effect by Seagate further boosted the HDD storage density. On the other hand, the TMR-based magnetic tunnel junctions (MTJs), characterized by amplified electrical signal outputs, has enabled the magnetoresistive random-access memory (MRAM) technology, which could be a superior alternative compared with the conventional CMOS technology. However, all these exotic effects had been only realized by using ferromagnetic materials before. As a result, the MRAM storage densities could be affected by the stray fields of ferromagnets and the information writing speeds are limited to the GHz spin dynamics of ferromagnets.

In contrast, antiferromagnets possess negligible stray fields and much higher spin dynamics on the THz level, which could significant advance the modern MRAM technology. Consequently, antiferromagnetic spintronics has emerged as a rapidly growing field in the past decade. One of the most important tasks of this hot field is to develop highly integrated all-antiferromagnetic tunnel junctions (AATJs). For this purpose, electrically controllable nonvolatile AATJs with large room-temperature electrical outputs (TMR > 150%) are essential. Although the very first AATJs have been realized two years ago in 2023 by two independent groups: one was based on Mn<sub>3</sub>Pt/MgO/Mn<sub>3</sub>Pt from Beihang University [Nature 613, 485 (2023)] and the other was based on Mn<sub>3</sub>Sn/MgO/Mn<sub>3</sub>Sn from Tokyo University [Nature 613, 490 (2023)], both were operated by external magnetic fields and the room-temperature TMR values were not sufficiently large, i.e., <150%. Therefore, electrically controllable nonvolatile AATJs with large room-temperature electrical outputs (TMR > 150%) are urgently needed to develop the next-generation all-antiferromagnetic MRAM technology.

Indeed, we noticed this work with great excitement soon after it appeared on arXiv in early September, almost ten days before the Nature review invitation. To us, this work is a significant and exciting breakthrough for antiferromagnetic spintronics. That is because the authors have realized the current operated AATJs with substantially enhanced room-temperature TMR values up to 363%. It could be a milestone building block for developing the all-antiferromagnetic MRAM technology. In addition to this point, the work has other major merits. Specifically,

(1) the authors report strikingly reliable switching in Mn<sub>3</sub>Pt-based nanoscale AATJ devices based on an attractive and brand-new mechanism – octupole-driven spin-transfer torque (OTT). It provides a fundamentally new pathway for controlling antiferromagnetic order electronically, moving beyond conventional spin-transfer or spin-orbit torque paradigms. Furthermore, the experimental demonstration is supported by a profound and rigorous theoretical framework. The combination of density functional theory calculations and multi-sublattice micromagnetic simulations does not merely support the data but delivers a deep microscopic understanding of the effect, linking the macroscopic switching to the sublattice-resolved spin current imbalance and the cluster magnetic octupole moment.

(2) The other compelling aspect of this work from an applied perspective is the demonstration of remarkable thermal stabilities. The authors quantitatively evaluate the thermal stability of the switched state through pulse-width-dependent

switching measurements. The extracted energy barrier could enable the data retention time on the order of ten years, meeting the fundamental requirement for non-volatile memory applications. Achieving such retention in a nanoscale AATJ device, while simultaneously maintaining a remarkably high TMR ratio and efficient current-induced switching, represents a significant leap forward. These characteristics firmly establish AATJs as a competitive platform for the next-generation MRAM technology.

(3) Compared with the initially reported AATJs in 2023 which were deposited at high temperatures, the materials here are all fabricated at room temperature. This is a highly critical technical progress, which immediately enables the fabrication compatibility of AATJs with the contemporary silicon-based semiconductor technology.

Therefore, it is our genuine pleasure to recommend it for publication in Nature. The following points could be considered by the authors for improving the manuscript.

1. To differentiate the current device with early tunnel junctions which utilized antiferromagnets as key functional layers but still contained ferromagnetic layers, the term/abbreviation AATJ shall be utilized throughout the entire manuscript including the extended data, in place of AFMTJ. While both are descriptive, AATJ is more concise and has been widely adopted.
2. The temperature-dependent measurements presented in Fig. 2c are compelling and serve as a critical control experiment. The clear extinction of both the TMR and switching behavior between 360 and 400 K provides strong evidence that the observed phenomena are intrinsically linked to the magnetic order in Mn<sub>3</sub>Pt. The authors note that some devices show gradual multilevel switching, which they attribute to magnetic domain behavior. Could the authors comment on whether domain wall motion or nucleation is likely involved? The discussion on how domain size relative to pillar diameter affects switching uniformity would be helpful for future device scaling.
3. The authors suggest that OTT efficiency could be enhanced by band structure engineering. Could they speculate on which kind of band structure features or material systems might exhibit stronger OTT? This could guide further following studies.
4. In Fig. 2a, the switching current densities seem slightly asymmetric for the two polarities. Could the authors comment on this issue in the main text?
5. The statistical distribution of the TMR values across numerous devices presented in Fig. 2b is highly valuable. It effectively demonstrates the robustness and reproducibility of the fabricated AATJs. On the other hand, one would be curious about the origin of the large performance variation. An explanation from the microstructural point of view for nanoscale AATJ devices could be helpful.
6. Given the ultrafast nature of antiferromagnetic dynamics, a paragraph of discussion in the outlook speculating on the potential switching speed limits of OTT—perhaps guided by the authors' theoretical models—would be an attractive addition for further exploring high-speed applications.

Referee #2

(Remarks to the Author)

I co-reviewed this manuscript with one of the reviewers who provided the listed reports.

Referee #3

(Remarks to the Author)

The manuscript by J. Kang and co-authors reports on the all-electrical (read and write) control of all-antiferromagnetic tunnel junctions (AFMTJs) deposited on SiO<sub>2</sub> substrates, which are compatible with industrial processes. The AFMTJs are constructed in the form of PtMn<sub>3</sub>/MgO/PtMn<sub>3</sub>, where MgO serves as the tunnel barrier and PtMn<sub>3</sub> is a non-collinear antiferromagnet, a specific class of AFM materials that can spin-polarize a charge current. Previous studies have shown that such systems based on Mn<sub>3</sub>X can exhibit tunnel magnetoresistance (TMR) due to their unique spin transport properties. While the possibility of polarizing a current in chiral Mn<sub>3</sub>X materials is well-established, the write operation in AFMTJs has not been reported until now. The authors demonstrate in this work the write operation in scaled AFMTJ devices down to 50 nm in diameter. The write current is relatively low (10 MA/cm<sup>2</sup>), and the reported TMR can achieve very high values (up to 350%). The switching mechanism is explained via octupole-driven spin-transfer torque (OTT), with corresponding models for macrospin and micromagnetic simulations proposed in the manuscript.

Realizing all-electrically addressable AFMTJs holds great promise for the spintronics and microelectronics communities, as it could combine the advantages of MRAM (speed, CMOS compatibility, density) with improved immunity to external fields, addressing one of the current drawbacks of MRAM technology that limits its penetration into the consumer market.

The paper is well-written and clear, and the findings are exciting and of utmost importance for memory applications (and beyond), if confirmed. However, several clarifications are needed before I can recommend its publication. Below, I address my main questions and concerns.

**Nature of the AFM :**

• While the measurements clearly show STT and TMR effects, I am skeptical about the reality of Mn<sub>3</sub>Pt layers deposited at room temperature without annealing. The literature (e.g., ref. 11 in the present paper) indicates that obtaining the desired chiral AF phase requires high-temperature deposition and annealing.

The authors should provide evidence that their layers are free of ferromagnetic clusters, such as X-ray diffraction, magnetization measurements, or simple transport measurements (e.g., AHE vs. T to see if the critical temperature corresponds to the expected Néel temperature for Mn<sub>3</sub>Pt). Ferromagnetic clusters in this type of layer are quite common to my knowledge.

• I do not understand why the authors used different deposition methods for fabricating the NV center samples (high substrate temperature, with annealing, as is customary in the literature to obtain the correct phase, and with different substrates) and for fabricating the junctions (room temperature, without annealing).

It is difficult to accept the findings from one sample to support the results of a presumably relatively different sample. Clarifications are needed.

• Additionally, the authors should specify in the methods how the PtMn<sub>3</sub> is made (co-deposition or stoichiometric target) and provide the real stoichiometry of the layers, which is important for interpreting the results.

#### Device properties:

• The TMR values reported can be very high in some cases, but there is also an extremely large dispersion among the measured devices, with very few occurrences of large values. I wonder about the reliability of the TMR, the reason for the wide spread, and whether additional mechanisms could be at play.

The TMR is assessed only using switching curves, and other mechanisms related to “high” current density could be involved. Previous literature examples have shown that thin tunnel barriers can exhibit current-driven resistance changes regardless of magnetic properties (e.g., <https://doi.org/10.1109/TMAG.2003.815727>, <https://doi.org/10.1063/1.1687533>). I would appreciate it if the authors discussed and commented on the reason for the wide distribution of properties.

• I think it would be important to measure these devices under a magnetic field, as done in ref. 11, to correlate the TMR values with those obtained in the switching experiments.

• Can such high TMR values be expected given that the MgO is not crystallized?

• The MgO thickness is indicated to be 2 nm, but the resistance-area (RA) product is in the range of 1-10  $\Omega\cdot\text{cm}^2$ . To my understanding, at 2 nm MgO, the RA should be larger. Can the authors comment on this?

• It seems that each figure is made from different devices with varying properties. Can the authors provide clarification? Do the devices break down easily?

• How many times were the devices cycled to extract switching current, how large are the variations cycle to cycle?

• The manuscript states: “*The result reveals a magnetic domain size range of ~100-170 nm (Extended Data Fig. 2). It is thus reasonable to assume that only some of our devices, which have sizes comparable to or smaller than the expected domain size, would show abrupt and square-shaped switching.*” The nice plateau after switching is shown only once (Fig. 2b); but in the supplementary material, all devices below 100 nm exhibit gradual switching. The influence of the size of magnetic domains versus the size of junctions deserves to be discussed more thoroughly.

• The manuscript also states: “*We found that the current-induced switching and TMR both vanished at temperatures above 360 K. This result is consistent with PtMn<sub>3</sub> undergoing a magnetic phase transition from a non-collinear to a collinear phase at elevated temperature.*” Is this behavior irreversible when cooling down, and was the vanishing of properties above 360 K observed in all devices tested?

#### Referee #4

(Remarks to the Author)

In this work, the authors report on the experimental observation of spin-transfer torque in an all-antiferromagnetic tunnel junction (AFMTJ), the reciprocal effect of the tunnel magnetoresistance (TMR) previously reported in AFMTJs. They use a non-collinear antiferromagnet, Mn<sub>3</sub>Pt, which shows no net magnetization, but a spin-split band structure, leading to a cluster magnetic octupole moment.

The observation of TMR with non-collinear antiferromagnets has been reported recently (Refs. 11,12 and 13) and, as the authors write in the paper, Onsager reciprocity suggests that the sizeable TMR should be accompanied by its reciprocal effect, the analogous to the spin-transfer torque (STT) in ferromagnet. In this regard, the observation of this octupole-driven spin-transfer torque (OTT) is an expected follow-up after the groundbreaking reports in refs. 11-13, which from my point of view limits the impact of the results, taking into account that 2 of these 3 original reports were already published in Nature.

Additionally, I find several issues in the work that could be problematic:

1) The observed switching is assumed to be from the OTT across the AFMTJ. However, the AFMs have adjacent Pt layers, which could be a source of spin-orbit torque (SOT). In fact, the work of Ref. 13 by some of the authors, demonstrates electrical switching by SOT using an adjacent Pt layer. With the experiments shown, and without a proper control experiment, one cannot rule out this origin of the switching.

2) How do the authors know that the grown Mn<sub>3</sub>Pt films have the noncollinear AFM phase and do not have secondary phases with different stoichiometry that are ferromagnetic (FM)?

- 3) The NV magnetometry measurements are performed on different Mn<sub>3</sub>Pt films, sputtered on an MgO (100) substrate to obtain an epitaxial layer. These are different conditions from the full stack used for the AFMTJ, so it is unclear to me that the information obtained (such as the magnetic domain size) in one case can be simply extrapolated to the other case.
- 4) The TMR is only observed from the electrical switching between the two states. As a reference experiment, I miss a standard TMR with applied magnetic field, as in Refs. 11 and 12. This will help compare the magnitudes of the direct (TMR) and reciprocal (OTT) effect.
- 5) It is unclear whether the STEM image in Fig. 1c is from an actual AFMTJ pillar or from the stack before the etching.

Version 1:

Reviewer comments:

Referee #1

(Remarks to the Author)

The authors have clarified all the important questions raised by the referees. It is my pleasure to recommend it for publication at Nature.

In addition, the magnetic-field-dependent TMR measurements may not be applicable to the sample structure in this work. That is because to enable a magnetic-field-induced TMR, the switching fields of the top and bottom magnetic layers have to be distinct, which is not the case here.

Referee #2

(Remarks to the Author)

I co-reviewed this manuscript with one of the reviewers who provided the listed reports.

Referee #3

(Remarks to the Author)

I thank the author for the thorough and comprehensive responses to my and other reviewers questions. These responses have helped highlight some crucial aspects of the work and have strengthened the manuscript.

However, I remain unsatisfied with certain responses, particularly those concerning the AFM and AAMTJ devices measured under a magnetic field. I find it puzzling that there is no magnetic field characterization of TMR and spin transport by AHE, which has been done in similar studies to complete some of the AFM properties. While I recognize the evidence supporting the absence of magnetic moments in the polycrystalline AFM (SQUID curves), the arguments presented do not concern transport properties which was the initial request. Given that magnetic octupole moments can be manipulated by current-induced spin torque, it should be possible to switch them under the influence of a magnetic field. One would expect to observe TMR in a resistance-field loop and an AHE signal.

I believe this is an important aspect to demonstrate, and I will stick to this statement—I want to see it. Beyond gaining insight into the transport properties, this will also be important for analyzing the reversal field of each PtMn electrode, such as knowing which one is reversing in the device. For instance, can the authors explain why some R-I loops have opposite chirality, e.g., as seen in Extended Data Fig. 1?

This is also very important in a context where the resilience to external magnetic field is an important topic for MRAM, and that AAMTJ are supposed to address this issue.

Additionally, although I capture and appreciate the theoretical argumentation provided in the rebuttal letter and extended information, I am not entirely convinced by the claims of large TMR values related to termination of PtMn. To me, they appear to be outlier occurrences—I would expect more statistics to support this. Having measured many MgO-based MTJ devices, both amorphous and crystalline, I have never observed such a distribution.

I also find that the explanation of low RA, despite very large MgO barrier thickness, is opposed to the large TMR explanations. On a personal note, the RA value of 1000 Ohm.μm<sup>2</sup> from [Qin et al., Nature 613, 485 (2023)] is much more reasonable for 2.5 nm of MgO—maybe lower than the seminal paper but in a similar range. More recent papers about VCMA work rather show a RA of 1000 for MgO thickness in the range of 2nm.

In conclusion, while I have reservations about recommending this manuscript with its current form, I acknowledge that this work should hold significant importance. Although I do not request a complete study, I would expect to see some field-dependent data to support and clarify the nature of the AFM and the CMO, as these are crucial for substantiating the work.

Referee #4

(Remarks to the Author)

First, I would like to clarify that the opening statement in my report, “Onsager reciprocity suggests that the sizeable TMR should be accompanied by its reciprocal effect, analogous to spin-transfer torque (STT) in ferromagnets”, is taken literally from the manuscript itself (page 4): “Onsager reciprocity suggests that the sizeable TMR in AFMTJs should be accompanied by a reciprocal effect, analogous to STT in ferromagnet-based MTJs [44–47].” This formulation by the authors implies that the observation of octupole-driven spin-transfer torque (OTT) is, to some extent, expected. While I agree that identifying the theoretical mechanism and providing experimental confirmation for OTT would be novel if unambiguously demonstrated, my view is that this degree of “expectedness,” following the recent Nature reports on TMR, reduces the impact required for another Nature publication. The work may instead be more appropriate for a Nature sister journal (e.g., Nature Materials, Nature Electronics, etc.). However, the authors have not yet provided unequivocal evidence that the observed effect originates from OTT.

Below, I respond point by point to the authors’ replies.

#### 1) Possible spin–orbit torque (SOT) contribution from adjacent Pt layers

The authors’ response to my main concern, namely, that the adjacent Pt layers could generate SOT, is not convincing. While an asymmetry in switching current might be a well-established signature of STT-driven switching in ferromagnets, the expected behavior in noncollinear antiferromagnets is not known. If, as the authors argue, the extrapolation of STT concepts from ferromagnets to OTT in noncollinear antiferromagnets was not “expected”, then the same logic must apply to current asymmetry: one cannot assume that an asymmetry uniquely signals OTT in this system.

Similarly, it remains unclear whether SOT-driven switching in noncollinear antiferromagnets is expected to produce symmetric switching currents. Although this kind of experiment was reported by some of the authors in Advanced Materials (2024), the switching current data were not presented in a way that this can be checked. As a result, the arguments used to exclude an SOT origin are weak.

Importantly, Fig. 1b of the main text suggests a straightforward control experiment using the already fabricated AFMTJ devices is possible: applying current along the bottom Pt layer and measuring the TMR, then comparing the result with measurements using current across the AFMTJ pillar at comparable current densities. This experiment would require no additional fabrication yet is surprisingly absent.

Further control experiments could include replacing Pt with W (which has an opposite spin Hall angle) or, ideally, using a seed layer with a negligible spin Hall effect. This also raises the question of why Pt is specifically chosen as a seed layer, given that Mn<sub>3</sub>Pt is polycrystalline.

The most decisive test to distinguish SOT from OTT would be to reverse the magnetic state of the top “polarizer” layer. If switching with an external magnetic field is not possible, this could be achieved by field cooling from above the critical temperature (~360 K) under opposite field directions, and then examining whether the hysteresis of the current-induced switching reverses sign. Such a sign reversal is expected for OTT but not for SOT. Alternatively, replacing the top Mn<sub>3</sub>Pt “detector” layer with a ferromagnetic metal would also provide a control sample.

Without confirming this most crucial point, I cannot recommend publication of the article in Nature (or in any other journal).

#### 2) Secondary ferromagnetic phases

The concern regarding possible secondary ferromagnetic phases is adequately addressed by the authors.

#### 3) NV magnetometry on differently oriented samples [(001) vs (111)]

I still have reservations regarding the interpretation of the NV magnetometry results. If the film grown along the (001) direction is truly epitaxial, then according to the authors’ own hypothesis (see point 4), it should not be fully compensated. In that case, I would not expect similar magnetic domain sizes in not compensated (001) and fully compensated (111) samples, and any direct comparison between them becomes questionable.

#### 4) TMR measurements under magnetic field

While I understand that the films appear to be fully compensated and therefore not switchable with laboratory-accessible magnetic fields, I find it surprising that the polycrystalline films show no net magnetic moment at all. Previous reports on Mn<sub>3</sub>Pt do observe a finite net moment (e.g., Phys. Rev. Materials 5, 014201 (2021)). This suggests that the authors’ hypothesis — that epitaxial strain (absent in textured polycrystalline films) is responsible for the formation of an uncompensated magnetic moment — may not be valid.

#### 5) Clarification of Fig. 1c

Point 5 regarding Fig. 1c has now been clarified, and this issue is satisfactorily addressed.

Version 2:

Reviewer comments:

### Referee #3

(Remarks to the Author)

I thank the authors for their detailed and clear responses, as well as the complementary measurements they have provided. While the authors have presented squid and AHE measurements demonstrating the absence of net magnetization, the manuscript lacks an attempt to measure TMR under a magnetic field, which could have been conducted. Additionally, performing R-V loops at large fields, even at  $\pm 1$  T, would provide a striking demonstration of the device immunity to external fields. This is crucial for corroborating the work and addressing the resilience to external magnetic fields, a significant topic for MRAM applications.

I also have difficulties regarding the explanation that PtMn/MgO termination significantly impacts the TMR amplitude and sign, although the theoretical explanation is sound. This is particularly concerning in contrast to the polycrystalline nature of the sample, which should largely average these phenomena, as evidenced by the absence of a magnetic signal in the poly sample.

Despite these reservations, the manuscript represents a significant advancement in the field of spintronics. Given the novel contributions and the potential impact of this work, I can recommend the acceptance of this manuscript for publication in Nature.

### Referee #4

(Remarks to the Author)

I thank the authors for their detailed responses and for performing additional control experiments. In particular, the comparison between two-terminal (vertical) and three-terminal (in-plane) current injection is a valuable addition and indeed goes in the right direction toward ruling out a dominant spin-orbit torque (SOT) contribution.

However, a key concern from my previous report remains insufficiently addressed.

The most decisive experiment I suggested, reversing the magnetic state of the top “polarizer” layer (e.g., via field cooling from above the Néel temperature under opposite field directions) and testing whether the sign of the current-induced switching hysteresis reverses, has not been performed, nor meaningfully discussed in the rebuttal. As I previously stated, such a sign reversal would be expected for an octupole-driven spin-transfer torque (OTT) mechanism, but not for SOT. Importantly, this experiment is not only a direct test of OTT vs SOT, but also closely connected to the concerns raised by Referee #3 regarding field-dependent measurements and the ability to distinguish between different magnetic configurations of the two electrodes. The authors argue that conventional field-induced TMR measurements are not feasible in their system due to the absence of switching up to 9 T and (maybe) the lack of different coercive fields between the two  $\text{Mn}_3\text{Pt}$  layers. While this explanation is plausible, it further strengthens the case that the proposed field-cooling protocol is the most natural and accessible alternative to probe the relative orientation of the two layers.

Moreover, beyond SOT, other possible artifacts (for example, thermally driven effects such as transient heating toward or above the Néel temperature, which is around 360 K in this system) are not unambiguously excluded by the newly added experiments. The proposed polarizer-reversal test would also help rule out such scenarios.

Crucially, the requested experiment does not require any redesign of the device structure and should be doable with the already measured devices. Given its decisive nature, the absence of any attempt to address it leaves the central claim of unambiguous OTT switching insufficiently supported.

In summary, while the new SOT-control experiment strengthens the manuscript, I believe that the polarizer-reversal test (or an equally decisive alternative) remains essential to conclusively establish the mechanism of switching. Without this, the attribution to OTT is still not fully convincing.

### Version 3:

Reviewer comments:

### Referee #3

(Remarks to the Author)

I thank the authors for adding the data measured under a magnetic field; this was, in my opinion, an important control experiment to conduct. Personally, I would have included the figure in the main text.

I have no further comments and continue endorsing the paper for publication in Nature.

### Referee #4

(Remarks to the Author)

I thank the authors for their detailed responses and for performing the additional control experiments requested. Although some concerns may remain (such as the use of only  $\pm 0.2$  T for the field cooling process, that might not be large enough to control the octupole direction or, as referee 3 said, the lack of TMR under a magnetic field), there are now also good

evidences in favor of the authors' interpretation. I will thus recommend the paper for publication.

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## **Response to Reviewers:**

Dear Dr. Thomas,

Thank you for the opportunity to submit a revised version of the manuscript “Octupole-driven spin-transfer torque switching of all-antiferromagnetic tunnel junctions” for publication in *Nature*. We appreciate the time and effort that you and the reviewers dedicated to providing feedback on this manuscript.

We have addressed all comments made by the reviewers. Please see below, in [blue color](#), a point-by-point response to the reviewers’ comments, and a list of the corresponding revisions.

### **Reviewer #1 (Remarks to the Author):**

The discovery of the giant magnetoresistance (GMR) effect in the late 1980s significantly increased information storage density in hard disk drives (HDDs) due to the GMR HDD read heads applications in the 1990s, a breakthrough that was later honored with the Nobel Prize. Later in 2004, the development of HDD read heads based on the tunnel magnetoresistance (TMR) effect by Seagate further boosted the HDD storage density. On the other hand, the TMR-based magnetic tunnel junctions (MTJs), characterized by amplified electrical signal outputs, has enabled the magnetoresistive random-access memory (MRAM) technology, which could be a superior alternative compared with the conventional CMOS technology. However, all these exotic effects had been only realized by using ferromagnetic materials before. As a result, the MRAM storage densities could be affected by the stray fields of ferromagnets and the information writing speeds are limited to the GHz spin dynamics of ferromagnets.

In contrast, antiferromagnets possess negligible stray fields and much higher spin dynamics on the THz level, which could significant advance the modern MRAM technology. Consequently, antiferromagnetic spintronics has emerged as a rapidly growing field in the past decade. One of the most important tasks of this hot field is to develop highly integrated all-antiferromagnetic tunnel junctions (AATJs). For this purpose, electrically controllable nonvolatile AATJs with large room-temperature electrical outputs (TMR > 150%) are essential. Although the very first AATJs have been realized two years ago in 2023 by two independent groups: one was based on Mn<sub>3</sub>Pt/MgO/Mn<sub>3</sub>Pt from Beihang University [Nature 613, 485 (2023)] and the other was based on Mn<sub>3</sub>Sn/MgO/Mn<sub>3</sub>Sn from Tokyo University [Nature 613, 490 (2023)], both were operated by external magnetic fields and the room-temperature TMR values were not sufficiently large, i.e., <150%. Therefore, electrically controllable nonvolatile AATJs with large room-

temperature electrical outputs ( $\text{TMR} > 150\%$ ) are urgently needed to develop the next-generation all-antiferromagnetic MRAM technology.

Indeed, we noticed this work with great excitement soon after it appeared on arXiv in early September, almost ten days before the Nature review invitation. To us, this work is a significant and exciting breakthrough for antiferromagnetic spintronics. That is because the authors have realized the current operated AATJs with substantially enhanced room-temperature TMR values up to 363%. It could be a milestone building block for developing the all-antiferromagnetic MRAM technology. In addition to this point, the work has other major merits. Specifically,

(1) the authors report strikingly reliable switching in Mn3Pt-based nanoscale AATJ devices based on an attractive and brand-new mechanism – octupole-driven spin-transfer torque (OTT). It provides a fundamentally new pathway for controlling antiferromagnetic order electronically, moving beyond conventional spin-transfer or spin-orbit torque paradigms. Furthermore, the experimental demonstration is supported by a profound and rigorous theoretical framework. The combination of density functional theory calculations and multi-sublattice micromagnetic simulations does not merely support the data but delivers a deep microscopic understanding of the effect, linking the macroscopic switching to the sublattice-resolved spin current imbalance and the cluster magnetic octupole moment.

(2) The other compelling aspect of this work from an applied perspective is the demonstration of remarkable thermal stabilities. The authors quantitatively evaluate the thermal stability of the switched state through pulse-width-dependent switching measurements. The extracted energy barrier could enable the data retention time on the order of ten years, meeting the fundamental requirement for non-volatile memory applications. Achieving such retention in a nanoscale AATJ device, while simultaneously maintaining a remarkably high TMR ratio and efficient current-induced switching, represents a significant leap forward. These characteristics firmly establish AATJs as a competitive platform for the next-generation MRAM technology.

(3) Compared with the initially reported AATJs in 2023 which were deposited at high temperatures, the materials here are all fabricated at room temperature. This is a highly critical technical progress, which immediately enables the fabrication compatibility of AATJs with the contemporary silicon-based semiconductor technology.

Therefore, it is our genuine pleasure to recommend it for publication in Nature. The following points could be considered by the authors for improving the manuscript.

**Author response:** We thank the reviewer for recommending this work for publication and for their helpful comments, which have helped us improve the quality of the manuscript. In the following, we provide a point-by-point response to the reviewer's comments and a listing of the corresponding revisions.

1. To differentiate the current device with early tunnel junctions which utilized antiferromagnets as key functional layers but still contained ferromagnetic layers, the term/abbreviation AATJ shall be utilized throughout the entire manuscript including the extended data, in place of AFMTJ. While both are descriptive, AATJ is more concise and has been widely adopted.

**Author response:** Following the reviewer's suggestion, we changed the acronym "AFMTJ" to "AATJ" throughout the manuscript.

2. The temperature-dependent measurements presented in Fig. 2c are compelling and serve as a critical control experiment. The clear extinction of both the TMR and switching behavior between 360 and 400 K provides strong evidence that the observed phenomena are intrinsically linked to the magnetic order in Mn<sub>3</sub>Pt. The authors note that some devices show gradual multilevel switching, which they attribute to magnetic domain behavior. Could the authors comment on whether domain wall motion or nucleation is likely involved? The discussion on how domain size relative to pillar diameter affects switching uniformity would be helpful for future device scaling.

**Author response:** We thank the referee for highlighting the compelling nature of our temperature-dependent switching measurements.

The gradual switching observed in some devices can be attributed to the role of domain walls in the switching process. As stated in the main text, we designed the junction diameter (50 to 200 nm) to be within the range of domain sizes measured in NV microscopy on an epitaxial PtMn<sub>3</sub> film. As expected, we therefore observed both single- and multi-domain switching behavior in the devices measured. We note that in our polycrystalline textured PtMn<sub>3</sub> films, grain boundaries can also affect the domain size, and thus the domains in our textured (111) PtMn<sub>3</sub> films (which cannot be measured by NV magnetometry due to their compensated surface) could be smaller than those of our epitaxial (001) PtMn<sub>3</sub> films that were used in the NV microscopy experiment.

We emphasize that abrupt single-domain switching was observed for several devices as shown in **Fig. R1**. Importantly, all these devices have a pillar size of 100 nm or smaller, in agreement with the intrinsic domain sizes expected from NV microscopy. These results confirm that such square-shaped switching behavior is achievable, but its occurrence is constrained by the magnetic domain size and interface termination, as outlined below.

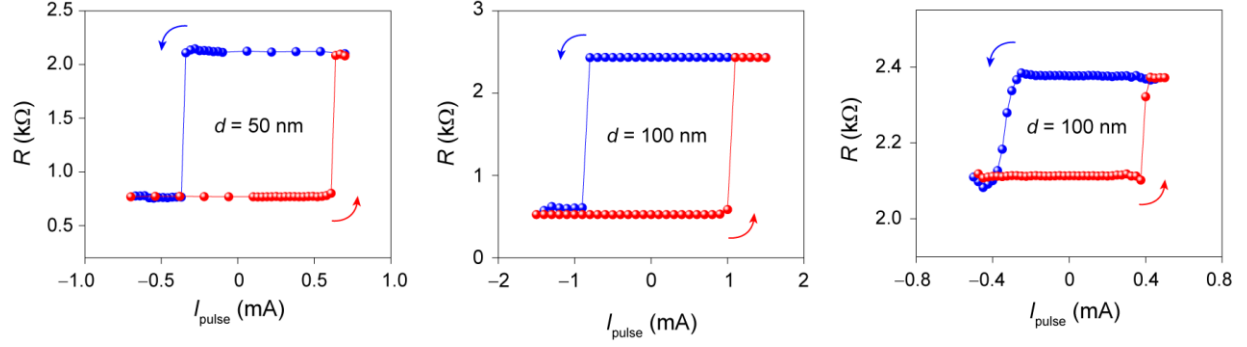


**Figure R1.** Current-induced switching measurements of AATJs showing abrupt transition between the two resistance states, indicating single-domain switching.

While a complete study of the different domain types and their associated dynamics goes beyond the scope of this paper, we theoretically investigated two factors – one *microstructural*, and the other *micromagnetic* – that play a key role in transitioning from multi- to single-domain switching behavior in our AATJs:

1. Effect of the surface termination of PtMn<sub>3</sub>: Even if both the top and bottom PtMn<sub>3</sub> layers contain a single lateral magnetic domain, surface roughness can generate regions with different relative surface termination on the two sides of the tunnel barrier. To account for different surface terminations, we simulated tunnel junctions with an effective translation of the unit cell of one PtMn<sub>3</sub> layer compared to the other, as illustrated in **Fig. R4a**. As discussed in detail in response to Comment #5, these variations in surface termination between the two electrodes lead to local variations of the TMR and hence, OTT efficiency. These local soft spots with a reduced critical switching current could act as nucleation centers for domain walls, which, combined with the local variations of the TMR ratio, account for the gradual switching observed in many of the measured devices.

2. 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 in the previous version of the manuscript. 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 Methods section of the manuscript. Formally, this model is the same as the macrospin model described in the previous version of the manuscript. However, here the effective field,  $H_{\text{eff}}$ , 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 then 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 PtMn<sub>3</sub> 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. R2a** and **Fig. R2b** 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 and results in sharp switching.

Case II. The switching always yields a uniform final state within the pulse duration (50 ps). This is shown in **Fig. R2c** and **Fig. R2d**, 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. R2e** and **Fig. R2f**, 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. R3a** and **Fig. R3b**, 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. R3c** and **Fig. R3e**. 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. R3d**. In **Fig. R3b**, 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. R3f**. 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.

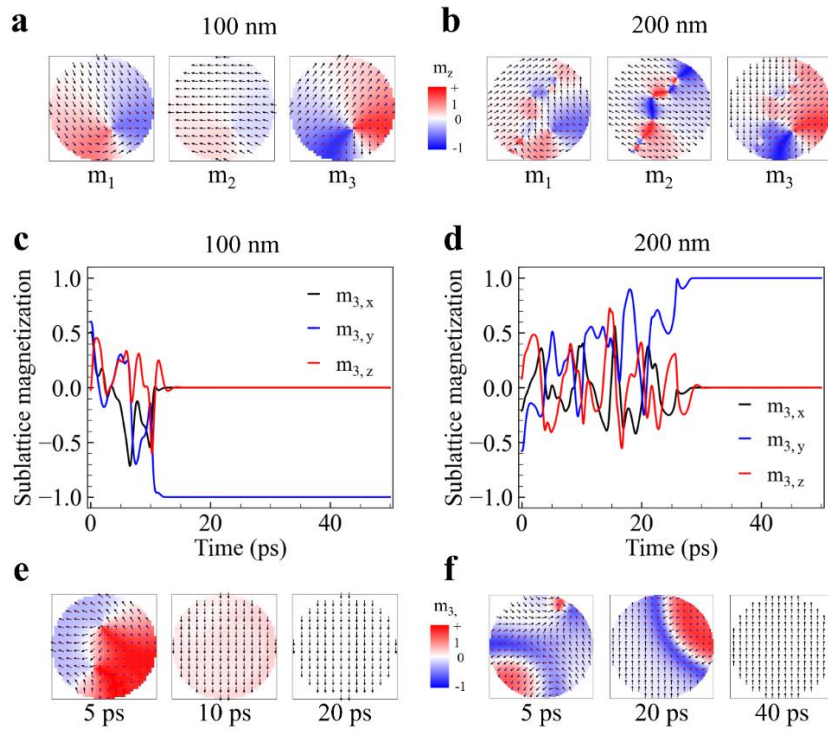


**Figure R2. Micromagnetic simulations of the size dependence of OTT dynamics in the case of non-uniform free layer magnetization.** Snapshot of the equilibrium magnetic configuration of the free layer for pillars with diameter of 100 nm **a**, and 200 nm **b**. Time-dependent x,y and z components of 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**. Snapshots

taken at different times during the switching dynamics of sublattice 3 for pillars with diameter of 100 nm **e**, and 200 nm **f**.

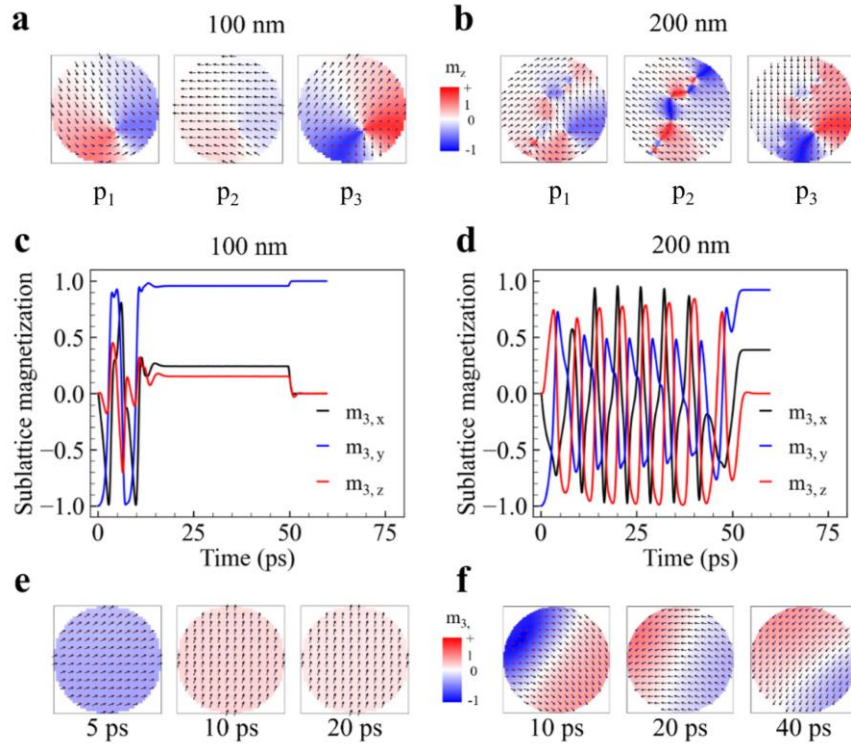


**Figure R3. Micromagnetic simulations of the size dependence of OTT dynamics in the case of non-uniform fixed layer polarization.** Snapshot of the current-induced polarization directions used for pillars of diameter 100 nm **a**, and 200 nm **b**. Here, we have used the same relaxed states as in Fig. R2, 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**. 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.

#### Revisions in the Manuscript:

Following this comment (and the related comment #5), we have added a discussion of the effect of surface termination on gradual switching and TMR distribution, as a new subsection within Methods, titled “Effect of interface termination on the TMR distribution of AATJ devices”. We have also added the micromagnetic simulations of switching in the presence of domain walls as a new Supplementary Note 5 to the revised manuscript.

3. The authors suggest that OTT efficiency could be enhanced by band structure engineering. Could they speculate on which kind of band structure features or material systems might exhibit stronger OTT? This could guide further following studies.

**Author response:** A comprehensive understanding of this question requires a systematic and extensive DFT study, which will be the topic of future work. However, we would like to qualitatively address this important comment as follows: Since the TMR and OTT are intrinsic consequences of the spin-split band structure and momentum-dependent spin polarization of the octupolar antiferromagnets, any material modification that would enhance this spin splitting would result in larger TMR and OTT efficiency. As illustrated in the manuscript, the spin splitting of the band structure is directly related to the local spin splitting on the Mn atoms. For example,  $\text{IrMn}_3$  and  $\text{RhMn}_3$  have the same crystal and magnetic noncollinear spin structure as  $\text{PtMn}_3$  with Néel temperatures of 727°C and 580°C, respectively. They do not show the noncollinear to collinear phase transition, which makes them attractive choices for memory devices in applications such as the automotive industry, which may need to operate at temperatures up to 150°C. Their large Néel temperatures indicate large exchange interactions, which we expect to result in large exchange splitting on the Mn atoms and hence larger octupole polarization. Therefore, both of these compounds may be good starting points for further investigation.

#### **Revisions in the Manuscript:**

Following the reviewer's suggestion, we have added a brief discussion of these potential material options in the revised manuscript, within the "Discussion and Outlook" section.

4. In Fig. 2a, the switching current densities seem slightly asymmetric for the two polarities. Could the authors comment on this issue in the main text?

**Author response:** We thank the reviewer for pointing this out. In all devices tested, the switching current required for the transition from the low resistance (low R) state to the high resistance (high R) state was consistently larger than the opposite direction. This asymmetry of switching current is a well-known signature of STT-driven switching in ferromagnets, where the nonlinear bias dependence of STT efficiency produces different critical currents for positive and negative bias. In fact, this asymmetry also rules out the possibility of the switching being related to spin-orbit torque (SOT). Given that the mechanism of OTT switching originates from staggered spin current acting on each sublattice due to the imbalance between intra- and inter-sublattice spin currents across the AATJ, the resulting switching characteristics naturally resemble those of STT.

#### **Revisions in the Manuscript:**

Per the reviewer's suggestion, we have added the following text in the revised manuscript, at the end of the section titled "Experimental observation of OTT-induced switching":

"It is worth noting that the Pt seed layer may provide a finite spin-orbit torque to the adjacent PtMn<sub>3</sub>. However, the asymmetry of switching currents for opposite switching directions, which was consistently observed in all measured devices, rules out SOT as the primary switching mechanism."

5. The statistical distribution of the TMR values across numerous devices presented in Fig. 2b is highly valuable. It effectively demonstrates the robustness and reproducibility of the fabricated AATJs. On the other hand, one would be curious about the origin of the large performance variation. An explanation from the microstructural point of view for nanoscale AATJ devices could be helpful.

**Author response:** We appreciate the referee's insightful comment and agree that the statistical distribution of TMR values shown in Fig. 2b demonstrates the robustness and reproducibility of our AATJ characteristics. At the same time, the observed spread in TMR values is understandable given the microstructural characteristics of our stacks: while the magnetic electrodes are highly (111)-textured, the tunnel barrier is amorphous. As a result, individual junctions inevitably experience variations in the local atomic configuration at the crystalline–amorphous interface, including differences in interface termination, short-range order within the barrier, and local fluctuations in the effective barrier height or thickness. These factors are expected to affect spin-dependent transmission, and their device-to-device variation naturally leads to a statistical distribution of TMR values.

To illustrate one possible origin of the TMR distribution, we modeled the effect of interface termination on the TMR response. Specifically, we performed theoretical calculations of the electron transmission across AATJs in which two antiferromagnetic PtMn<sub>3</sub> layers are separated by a vacuum gap, and different interface terminations are simulated by introducing a lateral shift  $d_{||}$  of the top PtMn<sub>3</sub> layer relative to the bottom layer (**Fig. R4a**). This approach allowed us to mimic various stacking configurations, including those where the magnetic structures at the two interfaces do not align and cases where the magnetic layer at one interface effectively encounters a layer of different registry on the opposite side. A vacuum barrier was used to simplify the modeling and isolate the effect of interface termination.

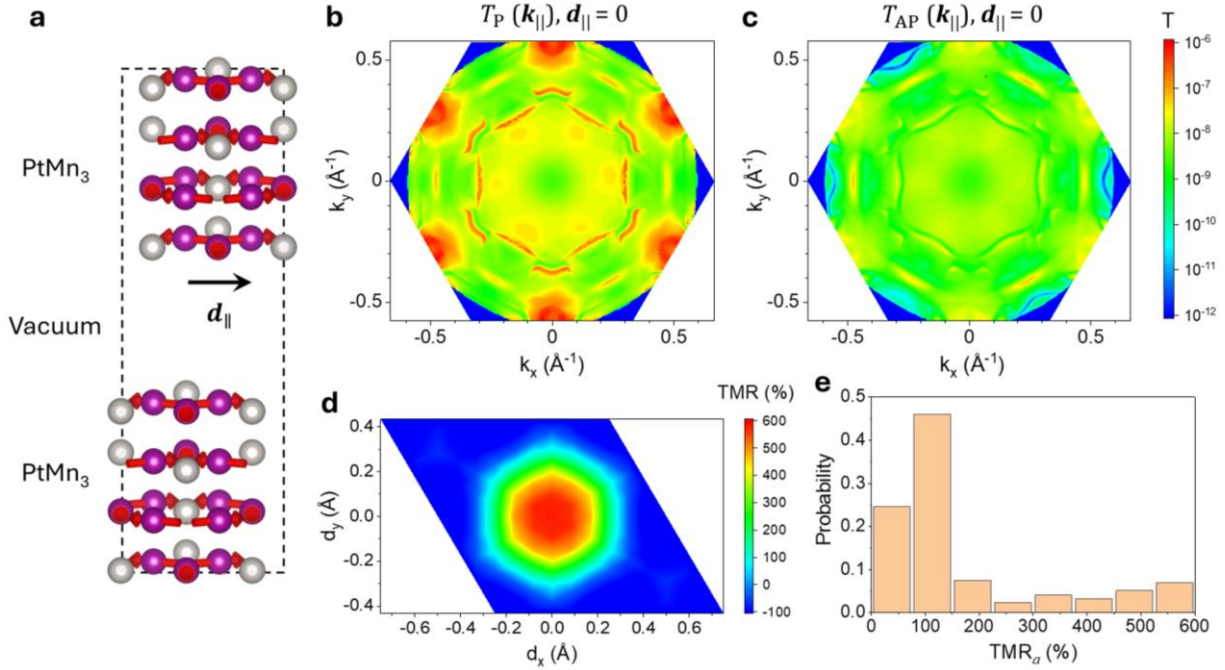


**Figure R4. Theoretical modelling of the effect of interface termination on TMR in PtMn<sub>3</sub>/vacuum/PtMn<sub>3</sub> AATJs.** **a**, Schematic of an AATJ where the top PtMn<sub>3</sub> layer has a lateral shift of  $d_{\parallel}$  with respect to the bottom layer. **b,c**,  $k_{\parallel}$ -resolved transmission for parallel  $T_P(k_{\parallel})$  (b) and antiparallel  $T_{AP}(k_{\parallel})$  (c) alignment of antiferromagnetic PtMn<sub>3</sub> layers for perfect matching of the magnetic interface structure for top and bottom electrodes ( $d_{\parallel} = 0$ ). **d**,  $TMR = (T_P - T_{AP})/T_{AP}$  as a function of lateral shift  $d_{\parallel} = (d_x, d_y)$ . **e**, Histogram of the distribution of TMR ratios defined as  $TMR_{\alpha} = |T_P - T_{AP}|/\min(T_P, T_{AP})$ .

**Figures R4b,c** show the calculated  $k_{\parallel}$ -resolved transmission for parallel  $T_P(k_{\parallel})$  (b) and antiparallel  $T_{AP}(k_{\parallel})$  (c) alignment of antiferromagnetic PtMn<sub>3</sub> layers when  $d_{\parallel} = 0$ , i.e., the magnetic moments of the top interface are perfectly aligned with the magnetic moments of the bottom interface. Such magnetic moment matching at the interfaces, combined with that in the bulk electrodes, makes  $T_P(k_{\parallel})$  much larger than  $T_{AP}(k_{\parallel})$  for almost all  $k_{\parallel}$ , resulting in total  $TMR = (T_P - T_{AP})/T_{AP}$  of about 600%. Shifting the top electrode with respect to the bottom, as described by the parameter  $d_{\parallel}$ , reduces  $T_P$ , while  $T_{AP}$  does not change much. As a result, the TMR decreases but remains positive for most lateral displacements  $d_{\parallel}$  except near shifts of approximately half a unit cell, where the minimum TMR is about -100% (**Fig. R4d**).

An important observation that follows from this analysis is that the TMR is primarily determined by the bulk magnetic structure of the electrodes. Magnetic moment matching in the P alignment, contrasted to the mismatch in the AP alignment, largely determines the sign of the TMR, although the interface termination still plays a role. We would like to

point out that the interface termination is constrained by the underlying bulk structure of PtMn<sub>3</sub> and cannot adopt arbitrary magnetic configurations. Due to bulk PtMn<sub>3</sub> exhibiting globally broken time-reversal symmetry, an exact match of both the magnetic and atomic structures at the two interfaces is not possible for the AP-aligned electrodes. This is why the distribution of TMR in **Fig. R4d** is strongly biased toward positive values.

To enable comparison to our experimentally measured values, we consider the absolute value of the TMR ratio, defined as  $TMR_a = |T_P - T_{AP}| / \min(T_P, T_{AP})$ . In **Fig. R4e**, we plot the distribution of the absolute TMR defined in this manner as obtained from our calculations, where the probability is represented by the normalized density of TMR values. Remarkably, this simple analysis captures the qualitative features of the experimental distribution shown in Fig. 2b of the main text.

We conclude, therefore, that the observed broad distribution of TMR values is not unexpected for junctions lacking full crystallinity in the stack. Improving this behavior would require exploring growth conditions that promote higher crystallinity and better control over surface termination, which will be the subject of future investigations.

#### **Revisions in the Manuscript:**

Following the reviewer's comment, we have added this discussion as a new sub-section within Methods, titled "Effect of interface termination on the TMR distribution of AATJ devices". We have also added Fig. R4 as a new Extended Data Fig. 7 in the revised manuscript.

6. Given the ultrafast nature of antiferromagnetic dynamics, a paragraph of discussion in the outlook speculating on the potential switching speed limits of OTT—perhaps guided by the authors' theoretical models—would be an attractive addition for further exploring high-speed applications.

**Author response:** We thank the referee for this suggestion. It is indeed interesting to consider the possibility of exploiting exchange fields in antiferromagnets for fast OTT-induced switching. For example, in the thermally activated regime of current-induced switching (as in our current experiments) the mean switching time is proportional to the attempt time  $\tau_0$ , following a similar expression as previously studied for ferromagnets [Koch et al., Phys. Rev. Lett. 92, 088302 (2004)]. Recent work has shown that in noncollinear antiferromagnets, large exchange fields can reduce  $\tau_0$  by several orders of magnitude compared to state-of-the-art perpendicular ferromagnets (PMA) [Konakanchi et al., Phys. Rev. Lett. 135, 136704 (2025)]. This suggests that thermally activated reversal via OTT in PtMn<sub>3</sub> may be significantly faster than in PMA ferromagnetic systems.

Additionally, the exchange fields impart a finite “mass” to octupole precessions in noncollinear antiferromagnets—a feature absent in PMA ferromagnets. These inertial dynamics have recently been exploited to lower the threshold current for precessional switching of octupoles in  $\text{Mn}_3\text{Sn}$  via spin-orbit torque using sub-ns pulsed currents [Takeuchi et al., Science, 389, 830 (2025)]. We anticipate that similar physics could enable fast, low-threshold OTT switching in suitably designed  $\text{PtMn}_3$  devices.

### **Revisions in the Manuscript:**

Following the reviewer’s suggestion, we have added a brief discussion of this point in the revised manuscript, within the “Discussion and Outlook” section.

### **Referee #2 (Remarks to the Author):**

I co-reviewed this manuscript with one of the reviewers who provided the listed reports.

**Author response:** We sincerely appreciate the reviewer’s time and careful reading of our manuscript.

### **Reviewer #3 (Remarks to the Author):**

The manuscript by J. Kang and co-authors reports on the all-electrical (read and write) control of all-antiferromagnetic tunnel junctions (AFMTJs) deposited on  $\text{SiO}_2$  substrates, which are compatible with industrial processes. The AFMTJs are constructed in the form of  $\text{PtMn}_3/\text{MgO}/\text{PtMn}_3$ , where  $\text{MgO}$  serves as the tunnel barrier and  $\text{PtMn}_3$  is a non-collinear antiferromagnets, a specific class of AFM materials that can spin-polarize a charge current.

Previous studies have shown that such systems based on  $\text{Mn}_3\text{X}$  can exhibit tunnel magnetoresistance (TMR) due to their unique spin transport properties. While the possibility of polarizing a current in chiral  $\text{Mn}_3\text{X}$  materials is well-established, the write operation in AFMTJs has not been reported until now. The authors demonstrate in this work the write operation in scaled AFMTJ devices down to 50 nm in diameter. The write current is relatively low ( $10 \text{ MA/cm}^2$ ), and the reported TMR can achieve very high values (up to 350%). The switching mechanism is explained via octupole-driven spin-transfer torque (OTT), with corresponding models for macrospin and micromagnetic simulations proposed in the manuscript.

Realizing all-electrically addressable AFMTJs holds great promise for the spintronics and microelectronics communities, as it could combine the advantages of MRAM (speed, CMOS compatibility, density) with improved immunity to external fields, addressing one of the current drawbacks of MRAM technology that limits its penetration into the consumer market.

The paper is well-written and clear, and the findings are exciting and of utmost importance for memory applications (and beyond), if confirmed. However, several clarifications are needed before I can recommend its publication. Below, I address my main questions and concerns.

**Author response:** We thank the reviewer for the positive assessment of this work and for their helpful comments, which have helped us improve the quality of the manuscript. In the following, we provide a point-by-point response to the reviewer's comments and a listing of the corresponding revisions.

#### **Nature of the AFM :**

1- While the measurements clearly show STT and TMR effects, I am skeptical about the reality of Mn<sub>3</sub>Pt layers deposited at room temperature without annealing. The literature (e.g., ref. 11 in the present paper) indicates that obtaining the desired chiral AF phase requires high-temperature deposition and annealing.

The authors should provide evidence that their layers are free of ferromagnetic clusters, such as X-ray diffraction, magnetization measurements, or simple transport measurements (e.g., AHE vs. T to see if the critical temperature corresponds to the expected Néel temperature for Mn<sub>3</sub>Pt). Ferromagnetic clusters in this type of layer are quite common to my knowledge.

**Author response:** The PtMn<sub>3</sub> films in this study show no sign of ferromagnetic clustering. Following the reviewer's suggestion, we performed X-ray diffraction (XRD) and superconducting quantum interference device (SQUID) magnetometry measurements on our AATJ stacks. In the XRD data, shown in **Fig. R5**, only the Pt (111) and PtMn<sub>3</sub> (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 of ferromagnetic clustering. SQUID magnetometry data on the full AATJ stack are shown in **Fig. R6**, where the magnetic field is swept along both the in-plane and out-of-plane directions up to a magnitude of 7 T. The figure shows no net magnetization in either direction, indicating the fully compensated antiferromagnetic phase of PtMn<sub>3</sub> without ferromagnetic clustering. In addition, we have provided the stoichiometry of our PtMn<sub>3</sub> films, measured using X-ray fluorescence (XRF), in response to comment #3 of the reviewer. There we show that we obtained the correct 3 to 1 stoichiometry necessary for the noncollinear phase.

Additionally, we would like to point out that room-temperature TMR values of the size shown here have never been previously observed in any ferromagnetic tunnel junction

that is not based on the CoFe(B)-MgO material system. Therefore, even if Mn- or Mn-Pt-based ferromagnetic clusters had been present in our experiment, they could not have explained the experimental observations of our study.

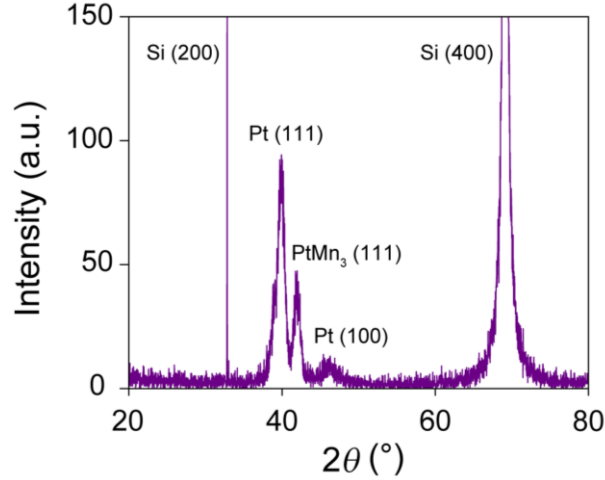


**Figure R5. X-ray diffraction measurements.** XRD measurements on full Si / SiO<sub>2</sub> / Pt (5 nm) / PtMn<sub>3</sub> (7 nm) / MgO (2 nm) / PtMn<sub>3</sub> (10 nm) / Pt (5 nm) / Ru (10 nm) AATJ material stacks.

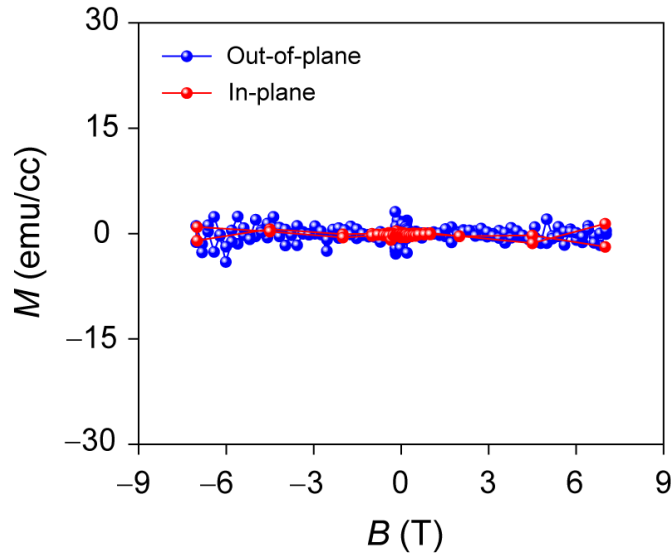


**Figure R6. SQUID magnetometry measurements.** Magnetization measurements on full Si / SiO<sub>2</sub> / Pt (5 nm) / PtMn<sub>3</sub> (7 nm) / MgO (2 nm) / PtMn<sub>3</sub> (10 nm) / Pt (5 nm) / Ru (10 nm) AATJ material stacks, conducted using a superconducting quantum interference device by sweeping 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.

**Revisions in the Manuscript:**

We have added the X-ray diffraction, X-ray fluorescence, and SQUID magnetometry results to the revised manuscript as a new Supplementary Note 1.

2- I do not understand why the authors used different deposition methods for fabricating the NV center samples (high substrate temperature, with annealing, as is customary in the literature to obtain the correct phase, and with different substrates) and for fabricating the junctions (room temperature, without annealing). It is difficult to accept the findings from one sample to support the results of a presumably relatively different sample. Clarifications are needed.

**Author response:** Thank you for pointing out that this was not sufficiently clear in the previous version of our manuscript.

All of our PtMn<sub>3</sub> layers were deposited by co-sputtering of the Pt and Mn targets. To have an educated guess on the suitable pillar sizes that would most likely have a single domain magnetic configuration, we needed to estimate the intrinsic domain size in sputter-deposited PtMn<sub>3</sub>. However, using the (111) PtMn<sub>3</sub> surface for this purpose is not possible, since it is fully compensated. For this reason, to measure the domain size via NV magnetometry, we needed to grow the PtMn<sub>3</sub> in the (001) direction as that would result in an uncompensated surface termination. The comparison of these two surfaces of PtMn<sub>3</sub> is shown in **Fig. R7**. The reason we used an MgO (001) substrate for this experiment was that the growth of PtMn<sub>3</sub> (001) on an amorphous substrate was not possible. By performing the NV magnetometry on the epitaxial single crystalline sample we were able to characterize the intrinsic magnetic domain size determined by the magnetic interactions. This serves as an upper limit to the domain sizes of PtMn<sub>3</sub> in our AATJ devices. As we discussed this in the manuscript, in our AATJ stack, the domain size is also influenced by grain boundaries due to the in-plane polycrystallinity of our textured PtMn<sub>3</sub> films, which further reduces the domain size.

#### **Revisions in the Manuscript:**

We have added a brief discussion of this point in a new Supplementary Note 2, along with Fig. R7 as Supplementary Figure S4.

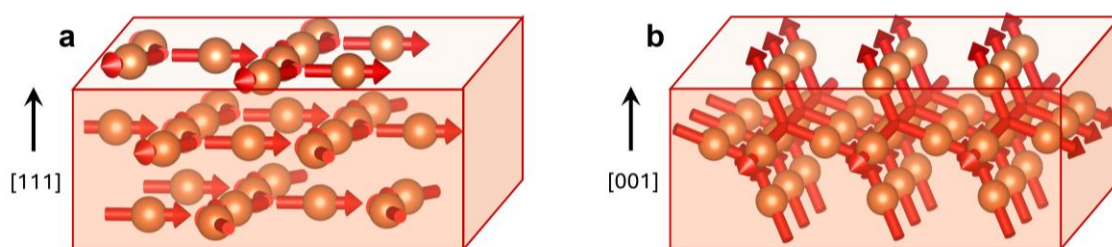


**Figure R7. Comparison of (001) and (111) surface terminations of  $\text{PtMn}_3$ .** Only the (001) surface has an uncompensated magnetic moment, which allows its domain structure to be detected in NV microscopy measurements.

3- Additionally, the authors should specify in the methods how the  $\text{PtMn}_3$  is made (co-deposition or stoichiometric target) and provide the real stoichiometry of the layers, which is important for interpreting the results.

**Author response:** The  $\text{PtMn}_3$  layers were deposited by co-sputtering from Pt and Mn targets. To determine the actual stoichiometry, X-ray fluorescence (XRF) measurements were performed on the samples. 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  $\text{Pt}_{25}\text{Mn}_{75}$  composition. These results are illustrated in **Fig. R8**.

We have added the following text to the Methods section in the revised manuscript:

“The  $\text{PtMn}_3$  layers were deposited by co-sputtering from Pt and Mn targets. X-ray fluorescence measurements showed a Pt composition ranging from 23.9 to 25.8 atomic percent across the wafer.”

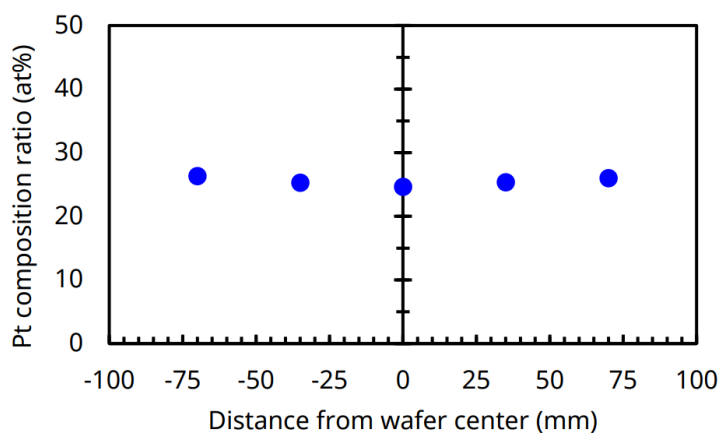


**Figure R8. Composition of  $\text{PtMn}_3$  across different positions on a 150 mm wafer.** The composition was measured by X-ray fluorescence, indicating a uniform stoichiometry.

### **Revisions in the Manuscript:**

We have added the X-ray diffraction, X-ray fluorescence, and SQUID magnetometry results to the revised manuscript as a new Supplementary Note 1.

### **Device properties:**

4- The TMR values reported can be very high in some cases, but there is also an extremely large dispersion among the measured devices, with very few occurrences of large values. I wonder about the reliability of the TMR, the reason for the wide spread, and whether additional mechanisms could be at play. The TMR is assessed only using switching curves, and other mechanisms related to “high” current density could be involved. Previous literature examples have shown that thin tunnel barriers can exhibit current-driven resistance changes regardless of magnetic properties (e.g., <https://doi.org/10.1109/TMAG.2003.815727>, <https://doi.org/10.1063/1.1687533>). I would appreciate it if the authors discussed and commented on the reason for the wide distribution of properties.

**Author response:** We appreciate the reviewer’s comment, which raises two important questions that we will address in sequence:

**1. Evidence that the switching is magnetic in origin:** As the reviewer correctly points out, non-magnetic current-induced resistance changes have been reported previously in ultrathin or defective tunnel barriers. However, several points indicate that such mechanisms cannot account for the behavior observed in our AATJs.

First, in several of our devices, as shown in **Fig. R9**, we observe abrupt switching with two well-defined and saturated resistance values. This is qualitatively different from any non-magnetic current-driven resistance modifications, which could be caused by mechanisms including soft dielectric breakdown, filament formation, or electromigration [Ielmini, IEEE Trans. Electron Devices 58, 4309 (2011), Ielmini, Semicond. Sci. Technol. 31 063002 (2016), Liu et al., IEEE Trans. Electron Devices 58, 3042 (2011), Frankovic et al., J. Appl. Phys. 81, 1604 (1997), Kozlova et al., Ultramicroscopy 158, 74 (2015)].

Second, non-magnetic current-induced resistive switching effects are, in general, thermally activated, and therefore do not disappear at the elevated temperatures considered here. This is in fact illustrated in one of the papers highlighted by the reviewer [Deac et al., J. Appl. Phys. 95, 6792 (2004)], where the authors report a resistance change ratio (i.e., apparent TMR, but not magnetic in origin) of “8% at room temperature, which

is increased to 11% at 75°C, then 30% at 100°C, and over 100% at 125°C.” This is in sharp contrast to the resistive switching signals observed in our work, which completely disappear as temperature increases beyond the noncollinear ordering temperature of PtMn<sub>3</sub>. Furthermore, the TMR and switching in our measurements recover as we cool down the sample down to room temperature. This striking qualitative difference provides strong evidence that the AATJ switching observed in our work is indeed magnetic in origin.

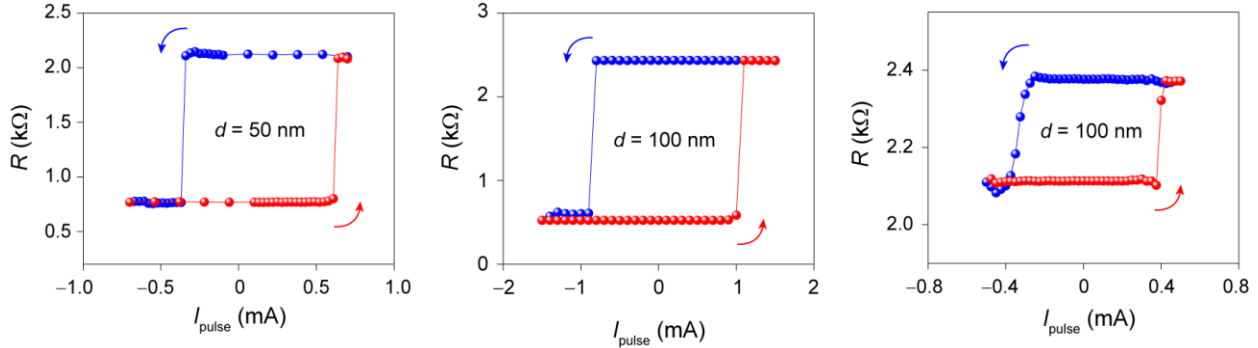


**Figure R9.** Current-induced switching measurements of AATJs showing abrupt transition between the two resistance states, indicating single-domain switching.

**2. Origin of the observed TMR distribution:** The observed spread in TMR values is understandable given the microstructural characteristics of our stacks: while the magnetic electrodes are highly (111)-textured, the tunnel barrier is amorphous. As a result, individual junctions inevitably experience variations in the local atomic configuration at the crystalline–amorphous interface, including differences in interface termination, short-range order within the barrier, and local fluctuations in the effective barrier height or thickness. These factors are expected to affect spin-dependent transmission, and their device-to-device variation naturally leads to a statistical distribution of TMR values.

To illustrate one possible origin of the TMR distribution, we modeled the effect of interface termination on the TMR response. Specifically, we performed theoretical calculations of the electron transmission across AATJs in which two antiferromagnetic PtMn<sub>3</sub> layers are separated by a vacuum gap, and different interface terminations are simulated by introducing a lateral shift  $d_{||}$  of the top PtMn<sub>3</sub> layer relative to the bottom (**Fig. R10a**). This approach allowed us to mimic various stacking configurations, including those where the magnetic structures at the two interfaces do not align and cases where the magnetic layer at one interface effectively encounters a layer of different registry on the opposite side. A vacuum barrier was used to simplify the modeling and isolate the effect of interface termination.

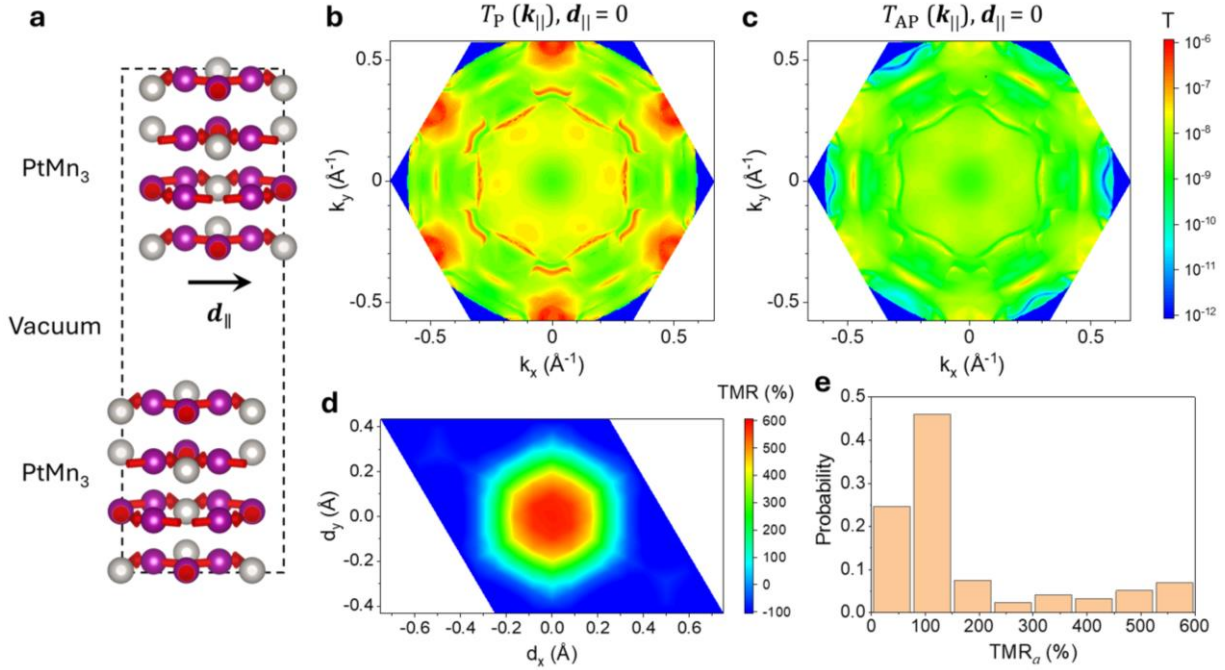


**Figure R10. Theoretical modelling of the effect of interface termination on TMR in PtMn<sub>3</sub>/vacuum/PtMn<sub>3</sub> AATJs.** **a**, Schematic of an AATJ where the top PtMn<sub>3</sub> layer has a lateral shift of  $d_{\parallel}$  with respect to the bottom layer. **b,c**,  $k_{\parallel}$ -resolved transmission for parallel  $T_P(k_{\parallel})$  (b) and antiparallel  $T_{AP}(k_{\parallel})$  (c) alignment of antiferromagnetic PtMn<sub>3</sub> layers for perfect matching of the magnetic interface structure for top and bottom electrodes ( $d_{\parallel} = 0$ ). **d**,  $TMR = (T_P - T_{AP})/T_{AP}$  as a function of lateral shift  $d_{\parallel} = (d_x, d_y)$ . **e**, Histogram of the distribution of TMR ratios defined as  $TMR_a = |T_P - T_{AP}|/\min(T_P, T_{AP})$ .

**Figures R10b,c** show the calculated  $k_{\parallel}$ -resolved transmission for parallel  $T_P(k_{\parallel})$  (b) and antiparallel  $T_{AP}(k_{\parallel})$  (c) alignment of antiferromagnetic PtMn<sub>3</sub> layers when  $d_{\parallel} = 0$ , i.e., the magnetic moments of the top interface are perfectly aligned with the magnetic moments of the bottom interface. Such magnetic moment matching at the interfaces, combined with that in the bulk electrodes, makes  $T_P(k_{\parallel})$  much larger than  $T_{AP}(k_{\parallel})$  for almost all  $k_{\parallel}$ , resulting in total  $TMR = (T_P - T_{AP})/T_{AP}$  of about 600%. Shifting the top electrode with respect to the bottom, as described by the parameter  $d_{\parallel}$ , reduces  $T_P$ , while  $T_{AP}$  does not change much. As a result, the TMR decreases but remains positive for most lateral displacements  $d_{\parallel}$  except near shifts of approximately half a unit cell, where the minimum TMR is about -100% (**Fig. R10d**).

An important observation that follows from this analysis is that the TMR is primarily determined by the bulk magnetic structure of the electrodes. Magnetic moment matching in the P alignment, contrasted to the mismatch in the AP alignment, largely determines the sign of the TMR, although the interface termination still plays a role. We would like to

point out that the interface termination is constrained by the underlying bulk structure of PtMn<sub>3</sub> and cannot adopt arbitrary magnetic configurations. Due to bulk PtMn<sub>3</sub> exhibiting globally broken time-reversal symmetry, an exact match of both the magnetic and atomic structures at the two interfaces is not possible for the AP-aligned electrodes. This is why the distribution of TMR in **Fig. R10d** is strongly biased toward positive values.

To enable comparison to our experimentally measured values, we consider the absolute value of the TMR ratio, defined as  $TMR_a = |T_P - T_{AP}| / \min(T_P, T_{AP})$ . In **Fig. R10e**, we plot the distribution of the absolute TMR defined in this manner as obtained from our calculations, where the probability is represented by the normalized density of TMR values. Remarkably, this simple analysis captures the qualitative features of the experimental distribution shown in Fig. 2b of the main text.

We conclude, therefore, that the observed broad distribution of TMR values is not unexpected for junctions lacking full crystallinity in the stack. Improving this behavior would require exploring growth conditions that promote higher crystallinity and better control over surface termination, which will be the subject of future investigations.

#### **Revisions in the Manuscript:**

Following the reviewer's comment, we have added this discussion as a new sub-section within Methods, titled "Effect of interface termination on the TMR distribution of AATJ devices". We have also added Fig. R10 as a new Extended Data Fig. 7 in the revised manuscript.

5- I think it would be important to measure these devices under a magnetic field, as done in ref. 11, to correlate the TMR values with those obtained in the switching experiments.

**Author response:** Thank you for this suggestion. While we agree that this would be a worthwhile experiment, it is unfortunately impossible to perform in our polycrystalline sputtered PtMn<sub>3</sub>-based AATJs. To illustrate the reason for this, we performed SQUID measurements on our full AATJ stack consisting of Si / SiO<sub>2</sub> / Pt (5 nm) / PtMn<sub>3</sub> (7 nm) / MgO (2 nm) / PtMn<sub>3</sub> (10 nm) / Pt (5 nm) / Ru (10 nm), by sweeping the magnetic field along both the in-plane and out-of-plane directions. **Figure R11a** shows the result of this measurement, in which no net magnetization is observed in either of these directions up to 7 T, indicating a fully compensated antiferromagnetic phase. This is in contrast to the previous report on (001)-oriented epitaxial PtMn<sub>3</sub> [Qin et al., Nature 613, 485 (2023)] in which the presence of a weak ferromagnetic moment due to epitaxial growth enabled magnetic-field-induced switching of the cluster magnetic octupole moments, thereby leading to the magnetic field-induced changes in TMR. To check whether epitaxial PtMn<sub>3</sub> (111) also gives rise to weak ferromagnetic moments, we separately grew Pt (7 nm) /

PtMn<sub>3</sub> (10 nm) / Pt (3 nm) films on a c-plane sapphire substrate. As a result, we obtained fully epitaxial (111)-oriented PtMn<sub>3</sub>. **Figure R11b** presents SQUID measurements on this epitaxial sample, in which a weak ferromagnetic moment is detected whose magnitude is comparable to those reported previously for epitaxial PtMn<sub>3</sub> films [Qin et al., Nature 613, 485 (2023), Liu et al., Nat. Electron. 1, 172 (2018), Zhao et al., Physica E. 138, 115141 (2022)]. Based on this, we infer that magnetic-field-induced switching can be achieved only in epitaxial PtMn<sub>3</sub> films. We hypothesize that epitaxial strain in such films, which is absent in textured polycrystalline films, may have a role in the formation of the uncompensated magnetic moment.

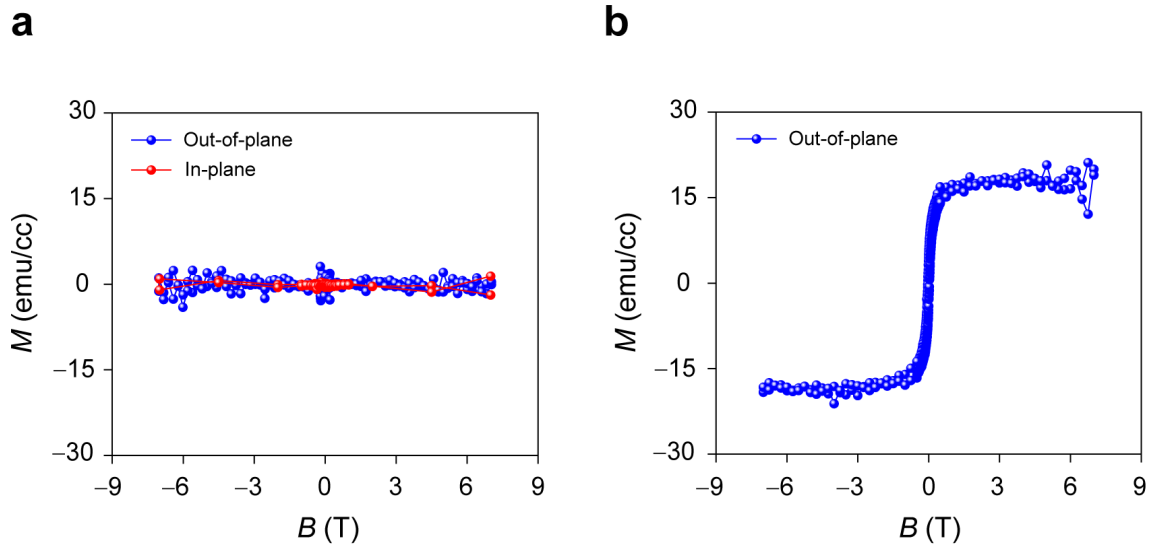


**Figure R11.** Magnetization measurements on **a**, polycrystalline Si / SiO<sub>2</sub> / Pt (5 nm) / PtMn<sub>3</sub> (7 nm) / MgO (2 nm) / PtMn<sub>3</sub> (10 nm) / Pt (5 nm) / Ru (10 nm) films and **b**, epitaxial Al<sub>2</sub>O<sub>3</sub> (0001) / Pt (7 nm) / PtMn<sub>3</sub> (10 nm) / Pt (3 nm) films. The measurements were conducted using a superconducting quantum interference device (SQUID) 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 backgrounds.

6- Can such high TMR values be expected given that the MgO is not crystallized?

**Author response:** Thank you for this important question, which we would like to discuss it in some detail.

In crystalline MgO, the barrier is translationally symmetric in-plane, so tunneling is *specular*, resulting in the conservation of the transverse electron momentum  $k_{\perp}$ . 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  $V_0 \sim 2\text{--}3$  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 PtMn<sub>3</sub>/MgO/PtMn<sub>3</sub> junctions. On the other hand, the intrinsic fluctuations of the conduction band minimum (CBM),  $\Delta$ , in an amorphous oxide layer, as follows from typical magnitudes of band-tail (Urbach) energies in amorphous oxides [see, e.g., Körner et al., Phys. Rev. B 86, 165210 (2012)] are of order  $\Delta \sim 0.3\text{--}0.4$  eV. Thus,  $\frac{\Delta}{V_0} \sim 0.1\text{--}0.2$ , can be considered as a small 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. The second term describes  $k$ -mixing due to disorder. For Gaussian-correlated barrier-height fluctuations with correlation length  $\xi$ , 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  $\Delta$  is the RMS amplitude of barrier-height fluctuations,  $\xi$  is their correlation length, and  $L$  is the barrier thickness.

For an amorphous MgO oxide, it is reasonable to take  $\xi$  to be of 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  $\Delta/V_0$ . 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  $\kappa_0$  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.

### **Revisions in the Manuscript:**

Following the reviewer's comment, we have added this discussion as a new Supplementary Note 3.

7- The MgO thickness is indicated to be 2 nm, but the resistance-area (RA) product is in the range of 1-10  $\Omega \cdot \text{cm}^2$ . To my understanding, at 2 nm MgO, the RA should be larger. Can the authors comment on this?

**Author response:** While a 2 nm MgO barrier in conventional ferromagnet-based MTJs typically results in a larger RA value, the RA value for the same MgO thickness can differ substantially in our PtMn<sub>3</sub>/MgO/PtMn<sub>3</sub> AATJs, for the following reasons.

First, the tunneling resistance strongly depends on the distribution of conduction channels and their transmission coefficients in k-space, which are intrinsically dependent on the electrode materials sandwiching the tunnel barrier. In conventional CoFe (or Fe) / MgO / CoFe (or Fe) MTJs, the large RA value originates from  $\Delta_1$  symmetry filtering enabled by the single-crystalline MgO barrier. The coherent  $\Delta_1$  tunneling selectively allows only slow decaying  $\Delta_1$  states to contribute to conduction, suppressing other channels and naturally leading to high RA values [Butler et al., Phys. Rev. B 63, 054416 (2001)]. In contrast, our MgO does not exhibit well-defined crystallization, and therefore does not support  $\Delta_1$  symmetry filtering. The absence of coherent symmetry-selected tunneling can substantially reduce the RA value at the same nominal thickness.

Second, roughness and morphological nonuniformity of the MgO layer, particularly at the bottom PtMn<sub>3</sub>/MgO interface, can locally reduce the effective barrier thickness. Since RA scales exponentially with barrier thickness, even a modest reduction in the effective thickness can significantly lower the RA value.

It is worth pointing out that the lower RA product for the same MgO thickness observed in our work is consistent with the numbers reported for (001)-oriented PtMn<sub>3</sub>/MgO/PtMn<sub>3</sub>

AATJs in [Qin et al., Nature 613, 485 (2023)], where AATJs with MgO thickness of 2.5 nm were reported to have RA around  $1000 \Omega \cdot \mu\text{m}^2$ . This is  $\sim 30\times$  smaller than what would be expected for a Fe/MgO/Fe MTJ with the same MgO thickness (see, e.g., Fig. 2b in Yuasa et al., Nat. Mater. 3, 868 (2004)).

8- It seems that each figure is made from different devices with varying properties. Can the authors provide clarification? Do the devices break down easily?

**Author response:** Indeed, the figures in the paper show data from a number of different devices. This is primarily because of the limited endurance of the devices, which restricts the number of measurements that could be performed in a single device. We also provide statistics of the TMR and a large number of representative switching loops measured across our wafer in Fig. 2b and Extended Data Fig. 1, respectively. Importantly, while there is a distribution of device characteristics, all key observations, conclusions, and qualitative features discussed in the paper were measured consistently and reproducibly in multiple devices.

9- How many times were the devices cycled to extract switching current, how large are the variations cycle to cycle?

**Author response:** The critical switching currents shown in Fig. 2d were extracted from a single switching measurement obtained at each pulse width, and the error bars represent the entire current range over which the switching occurs. To examine the cycle-to-cycle variations in switching currents, we performed consecutive switching cycles on a representative 100 nm AATJ. Here, we used a fine current step size of  $25 \mu\text{A}$  to reveal the switching transition in more detail. **Figure R12** shows two consecutive switching curves measured in this manner, indicating nearly identical switching currents in both iterations. From these data and similar behavior observed in other devices (e.g., Fig. 2c of the main text), we conclude that cycle-to-cycle variations of switching current are not large enough to affect the findings of this work.

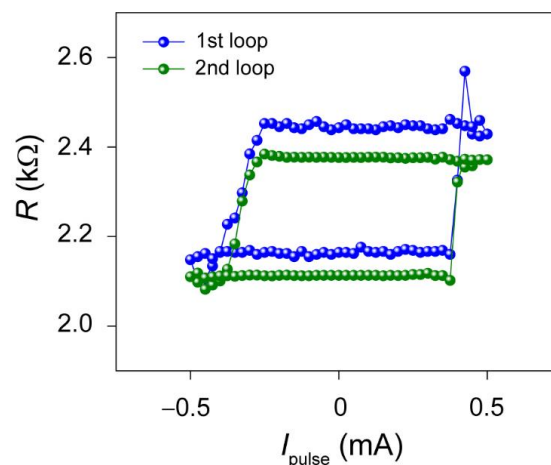


**Figure R12.** Repeated measurement cycles of OTT switching in an AATJ device with a diameter of 100 nm.

10- The manuscript states: "*The result reveals a magnetic domain size range of ~100-170 nm (Extended Data Fig. 2). It is thus reasonable to assume that only some of our devices, which have sizes comparable to or smaller than the expected domain size, would show abrupt and square-shaped switching.*" The nice plateau after switching is shown only once (Fig. 2b); but in the supplementary material, all devices below 100 nm exhibit gradual switching. The influence of the size of magnetic domains versus the size of junctions deserves to be discussed more thoroughly.

**Author response:** The gradual switching observed in some devices can be attributed to the role of domain walls in the switching process. As stated in the main text, we designed the junction diameter (50 to 200 nm) to be within the range of domain sizes measured in NV microscopy on an epitaxial PtMn<sub>3</sub> film. As expected, we therefore observed both single- and multi-domain switching behavior in the devices measured. We note that in our polycrystalline textured PtMn<sub>3</sub> films, grain boundaries can also affect the domain size, and thus the domains in our textured (111) PtMn<sub>3</sub> films (which cannot be measured by NV magnetometry due to their compensated surface) could be smaller than those of our epitaxial (001) PtMn<sub>3</sub> films that were used in the NV microscopy experiment.

While a complete study of the different domain types and their associated dynamics goes beyond the scope of this paper, we would like to highlight one factor that plays a key role in transitioning from multi- to single-domain switching behavior in our AATJs: The surface termination of PtMn<sub>3</sub>. Even if both the top and bottom PtMn<sub>3</sub> layers contain a single lateral magnetic domain, surface roughness can generate regions with different relative surface termination on the two sides of the tunnel barrier. To account for these different surface terminations, we simulated tunnel junctions with an effective translation of the unit cell of one PtMn<sub>3</sub> layer compared to the other, as illustrated in **Fig. R10a**. As discussed in detail

in response to Comment #4, these variations in surface termination between the two electrodes lead to local variations of the TMR and hence, OTT efficiency. These local soft spots with a reduced critical switching current could act as nucleation centers for domain walls, which, combined with the local variations of the TMR ratio, account for the gradual switching observed in many of the measured devices.

Nevertheless, we emphasize that abrupt single-domain switching was observed for several devices as shown in **Fig. R13**. Importantly, all these devices have a pillar size of 100 nm or smaller, which is in agreement with the intrinsic domain sizes expected from NV microscopy. These results confirm that such square-shaped switching behavior is achievable, but its occurrence is constrained by the magnetic domain size, grain size and the surface termination.

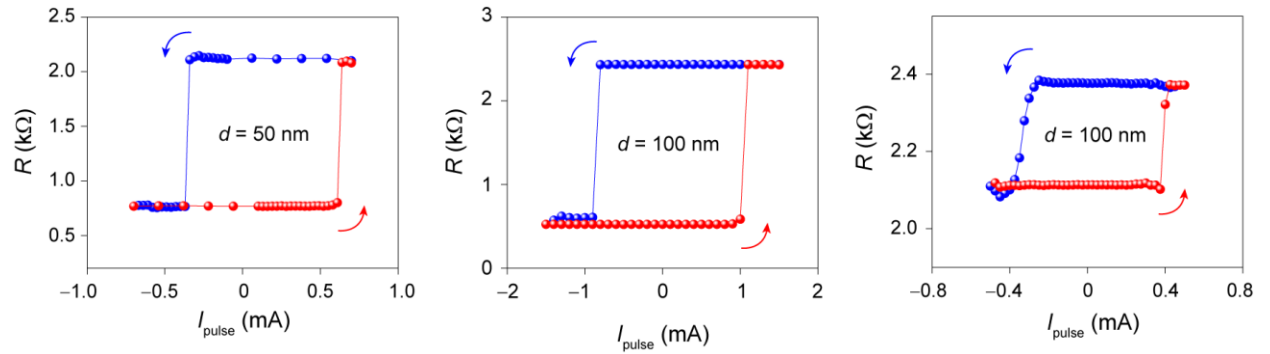


**Figure R13.** Current-induced switching measurements of AATJs showing abrupt transition between the two resistance states, indicating single-domain switching.

#### Revisions in the Manuscript:

Following this comment (and the related comment #4), we have added a discussion of the effect of surface termination on gradual switching and TMR distribution, as a new sub-section within Methods, titled “Effect of interface termination on the TMR distribution of AATJ devices”.

11- The manuscript also states: " We found that the current-induced switching and TMR both vanished at temperatures above 360 K. This result is consistent with PtMn3 undergoing a magnetic phase transition from a non-collinear to a collinear phase at elevated temperature." Is this behavior irreversible when cooling down, and was the vanishing of properties above 360 K observed in all devices tested?

**Author response:** 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 R14** 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.

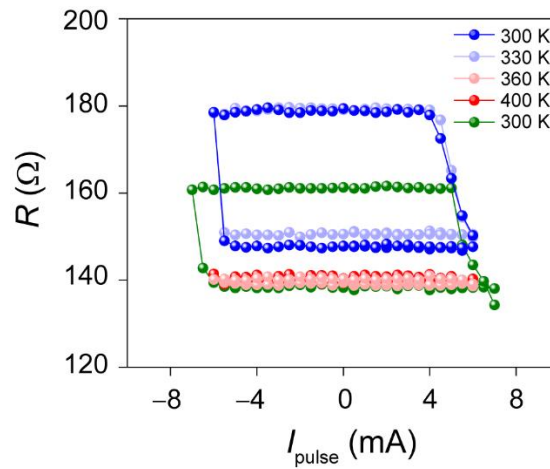


**Figure R14. Temperature-dependent current-induced switching measurements.** The switching measurements were performed for a temperature sequence starting from 300 K to 400 K and back to 300 K. Current-induced switching reappears upon cooling to room temperature, consistent with the octupolar origin of the OTT and TMR.

#### Revisions in the Manuscript:

Following the reviewer's comment, we have added this discussion and Fig. R14 in a new Supplementary Note 4.

#### **Reviewer #4 (Remarks to the Author):**

In this work, the authors report on the experimental observation of spin-transfer torque in an all-antiferromagnetic tunnel junction (AFMTJ), the reciprocal effect of the tunnel magnetoresistance (TMR) previously reported in AFMTJs. They use a non-collinear antiferromagnet, Mn<sub>3</sub>Pt, which shows no net magnetization, but a spin-split band structure, leading to a cluster magnetic octupole moment.

The observation of TMR with non-collinear antiferromagnets has been reported recently (Refs. 11,12 and 13) and, as the authors write in the paper, Onsager reciprocity suggests that the sizeable TMR should be accompanied by its reciprocal effect, the analogous to the spin-transfer torque (STT) in ferromagnet. In this regard, the observation of this octupole-driven spin-transfer torque (OTT) is an expected follow-up after the groundbreaking reports in refs. 11-13, which from my point of view limits the impact of the results, taking into account that 2 of these 3 original reports were already published in Nature.

**Author response:** We thank the reviewer for the careful reading of our manuscript and for their insightful comments, which helped us improve its quality.

Regarding the impact of our work, we respectfully disagree with the reviewer's statement about this work being an "expected follow-up" of previous TMR demonstrations in AATJs. OTT is a fundamentally new physical mechanism for control of antiferromagnetic order. On a fundamental level, the phenomenon of octupole-driven spin-transfer torque, and its underlying mechanism of sublattice-resolved spin-transfer, were neither theoretically predicted nor experimentally demonstrated prior to our work. On the practical level, it is challenging to realize AATJs with sufficiently low resistance-area products required to observe the OTT-induced switching. As such, the impact of the present paper goes far beyond the large TMR values that we report in PtMn<sub>3</sub>-based AATJs, although we do also improve the record TMR observed in AATJs by more than 3x compared to prior reports.

These elements of novelty and their high fundamental and technological impact have been also highlighted in detail by the other reviewers.

Additionally, I find several issues in the work that could be problematic:

1) The observed switching is assumed to be from the OTT across the AFMTJ. However, the AFMs have adjacent Pt layers, which could be a source of spin-orbit torque (SOT). In fact, the work of Ref. 13 by some of the authors, demonstrates electrical switching by

SOT using an adjacent Pt layer. With the experiments shown, and without a proper control experiment, one cannot rule out this origin of the switching.

**Author response:** We appreciate the reviewer's important comment. There is a key experimental signature that enables one to rule out the effect of SOT as our primary switching mechanism. In all devices tested, the switching current required for the transition from the low resistance (low R) state to the high resistance (high R) state was consistently larger than the opposite direction. This asymmetry of switching current is a well-known signature of STT-driven switching in ferromagnets, where the nonlinear bias dependence of STT efficiency produces different critical currents for positive and negative bias. Given that the mechanism of OTT switching originates from staggered spin current acting on each sublattice due to the imbalance between intra- and inter-sublattice spin currents across the AATJ, the resulting switching characteristics naturally resemble those of STT.

On the other hand, SOT switching is known to have symmetric switching current densities for both directions of switching. In fact, this symmetry has been used previously to separate SOT and STT effects in ferromagnet-based SOT-MRAM devices that may exhibit both effects [see, e.g., Sato et al., Nat. Electron. 1, 508 (2018)]. Therefore, while we do not exclude the possibility of a small SOT component in our devices, the universal and reproducible switching asymmetry across all AATJs clearly indicates that the dominant switching mechanism is not arising from SOT.

#### **Revisions in the Manuscript:**

Following the reviewer's suggestion, we have added the following text in the revised manuscript, at the end of the section titled "Experimental observation of OTT-induced switching":

"It is worth noting that the Pt seed layer may provide a finite spin-orbit torque to the adjacent PtMn<sub>3</sub>. However, the asymmetry of switching currents for opposite switching directions, which was consistently observed in all measured devices, rules out SOT as the primary switching mechanism."

2) How do the authors know that the grown Mn<sub>3</sub>Pt films have the noncollinear AFM phase and do not have secondary phases with different stoichiometry that are ferromagnetic (FM)?

**Author response:** The PtMn<sub>3</sub> films in this study show no sign of ferromagnetic clustering. Following the reviewer's suggestion, we performed X-ray diffraction (XRD) and superconducting quantum interference device (SQUID) magnetometry measurements on

our AATJ stacks. In the XRD data, shown in **Fig. R15**, only the Pt (111) and PtMn<sub>3</sub> (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 of ferromagnetic clustering. SQUID magnetometry data on the full AATJ stack are shown in **Fig. R16**, where the magnetic field is swept along both the in-plane and out-of-plane directions up to a magnitude of 7 T. The figure shows no net magnetization in either direction, indicating the fully compensated antiferromagnetic phase of PtMn<sub>3</sub> without ferromagnetic clustering. In addition, to determine the actual stoichiometry, X-ray fluorescence (XRF) measurements were performed on the samples. 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 Pt<sub>25</sub>Mn<sub>75</sub> composition. These results are illustrated in **Fig. R17**.

Additionally, we would like to point out that room-temperature TMR values of the size shown here have never been previously observed in any ferromagnetic tunnel junction that is not based on the CoFe(B)-MgO material system. Therefore, even if Mn- or Mn-Pt-based ferromagnetic clusters had been present in our experiment, they could not have explained the experimental observations of our study.

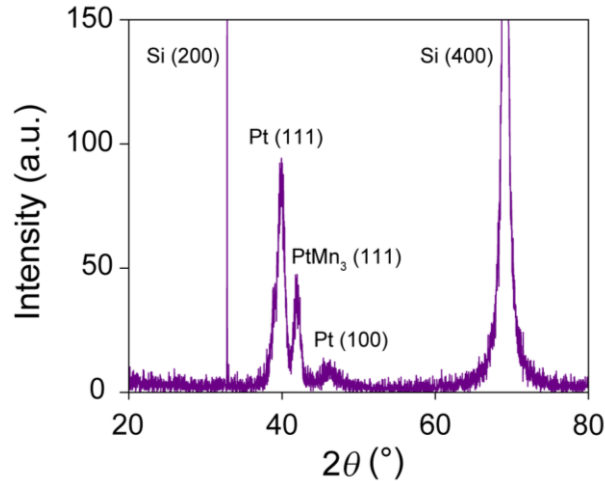


**Figure R15. X-ray diffraction measurements.** XRD measurements on full Si / SiO<sub>2</sub> / Pt (5 nm) / PtMn<sub>3</sub> (7 nm) / MgO (2 nm) / PtMn<sub>3</sub> (10 nm) / Pt (5 nm) / Ru (10 nm) AATJ material stacks.

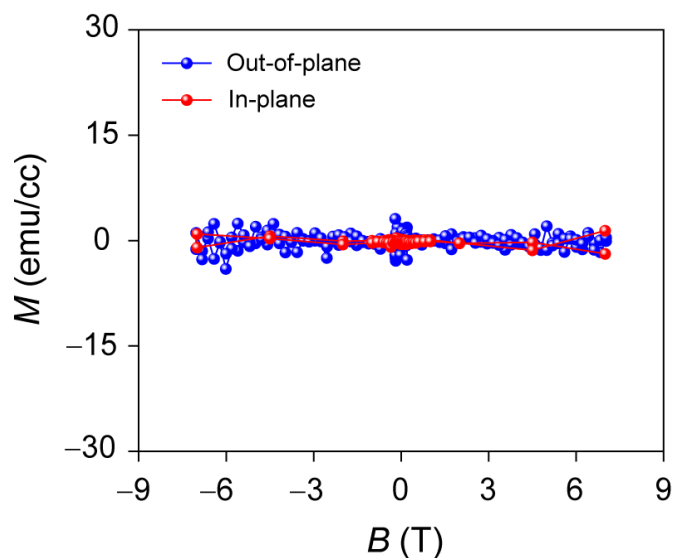


**Figure R16. SQUID magnetometry measurements.** Magnetization measurements on full Si / SiO<sub>2</sub> / Pt (5 nm) / PtMn<sub>3</sub> (7 nm) / MgO (2 nm) / PtMn<sub>3</sub> (10 nm) / Pt (5 nm) / Ru (10 nm) AATJ material stacks, conducted using a superconducting quantum interference device by sweeping 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.

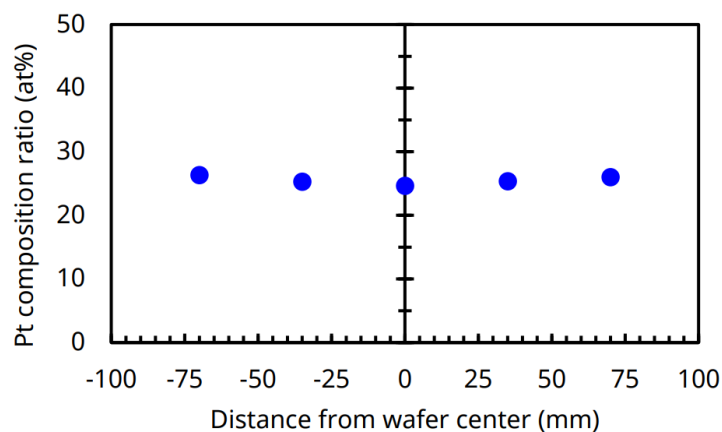


**Figure R17. Composition of PtMn<sub>3</sub> across different positions on a 150 mm wafer.** The composition was measured by X-ray fluorescence, indicating a uniform stoichiometry.

#### Revisions in the Manuscript:

We have added the X-ray diffraction, X-ray fluorescence, and SQUID magnetometry results to the revised manuscript as a new Supplementary Note 1.

3) The NV magnetometry measurements are performed on different Mn<sub>3</sub>Pt films, sputtered on an MgO (100) substrate to obtain an epitaxial layer. These are different conditions from the full stack used for the AFMTJ, so it is unclear to me that the information obtained (such as the magnetic domain size) in one case can be simply extrapolated to the other case.

**Author response:** Thank you for pointing out that this was not sufficiently clear in the previous version of our manuscript.

All of our PtMn<sub>3</sub> layers were deposited by co-sputtering of the Pt and Mn targets. To have an educated guess on the suitable pillar sizes that would most likely have a single domain magnetic configuration, we needed to estimate the intrinsic domain size in sputter-deposited PtMn<sub>3</sub>. However, using the (111) PtMn<sub>3</sub> surface for this purpose is not possible, since it is fully compensated. For this reason, to measure the domain size via NV magnetometry, we needed to grow the PtMn<sub>3</sub> in the (001) direction as that would result in an uncompensated surface termination. The comparison of these two surfaces of PtMn<sub>3</sub> is shown in **Fig. R18**. The reason we used an MgO (001) substrate for this experiment was that the growth of PtMn<sub>3</sub> (001) on an amorphous substrate was not possible. By performing the NV magnetometry on the epitaxial single crystalline sample we were able to characterize the intrinsic magnetic domain size determined by the magnetic interactions. This serves as an upper limit to the domain sizes of PtMn<sub>3</sub> in our AATJ devices. As we discussed this in the manuscript, in our AATJ stack, the domain size is also influenced by grain boundaries due to the in-plane polycrystallinity of our textured PtMn<sub>3</sub> films, which further reduces the domain size.

#### Revisions in the Manuscript:

We have added a brief discussion of this point in a new Supplementary Note 2, along with Fig. R18 as Supplementary Figure S4.

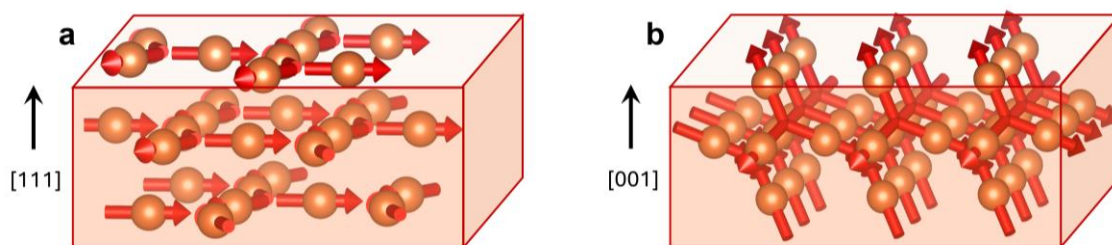


**Figure R18. Comparison of (001) and (111) surface terminations of PtMn<sub>3</sub>.** Only the (001) surface has an uncompensated magnetic moment, which allows its domain structure to be detected in NV microscopy measurements.

4) The TMR is only observed from the electrical switching between the two states. As a reference experiment, I miss a standard TMR with applied magnetic field, as in Refs. 11 and 12. This will help compare the magnitudes of the direct (TMR) and reciprocal (OTT) effect.

**Author response:** Thank you for this suggestion. While we agree that this would be a worthwhile experiment, it is unfortunately impossible to perform in our polycrystalline sputtered PtMn<sub>3</sub>-based AATJs. To illustrate the reason for this, we performed SQUID measurements on our full AATJ stack consisting of Si / SiO<sub>2</sub> / Pt (5 nm) / PtMn<sub>3</sub> (7 nm) / MgO (2 nm) / PtMn<sub>3</sub> (10 nm) / Pt (5 nm) / Ru (10 nm), by sweeping the magnetic field along both the in-plane and out-of-plane directions. **Figure R19a** shows the result of this measurement, in which no net magnetization is observed in either of these directions up to 7 T, indicating a fully compensated antiferromagnetic phase. This is in contrast to the previous report on (001)-oriented epitaxial PtMn<sub>3</sub> [Qin et al., Nature 613, 485 (2023)] in which the presence of a weak ferromagnetic moment due to epitaxial growth enabled magnetic-field-induced switching of the cluster magnetic octupole moments, thereby leading to the magnetic field-induced changes in TMR. To check whether epitaxial PtMn<sub>3</sub> (111) also gives rise to weak ferromagnetic moments, we separately grew Pt (7 nm) / PtMn<sub>3</sub> (10 nm) / Pt (3 nm) films on a c-plane sapphire substrate. As a result, we obtained fully epitaxial (111)-oriented PtMn<sub>3</sub>. **Figure R17b** presents SQUID measurements on this epitaxial sample, in which a weak ferromagnetic moment is detected whose magnitude is comparable to those reported previously for epitaxial PtMn<sub>3</sub> films [Qin et al., Nature 613, 485 (2023), Liu et al., Nat. Electron. 1, 172 (2018), Zhao et al., Physica E. 138, 115141 (2022)]. Based on this, we infer that magnetic-field-induced switching can be achieved only in epitaxial PtMn<sub>3</sub> films. We hypothesize that epitaxial strain in such films, which is absent in textured polycrystalline films, may have a role in the formation of the uncompensated magnetic moment.

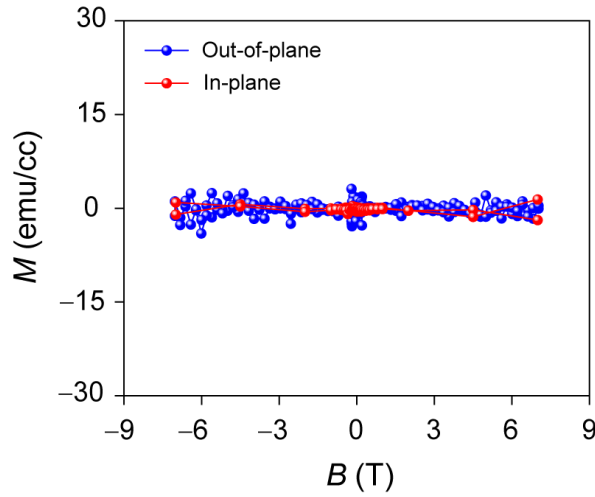
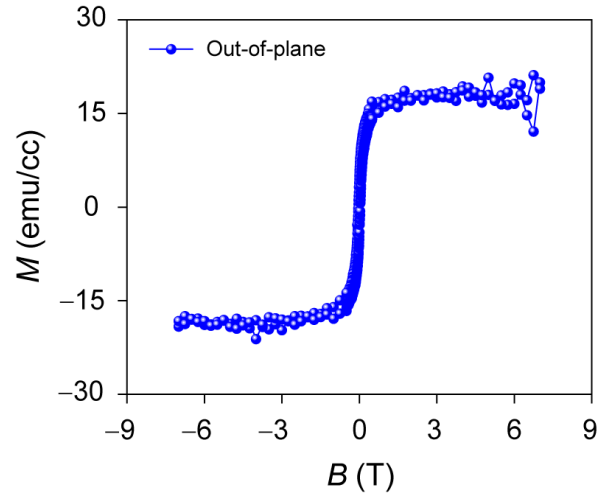
**a****b**

**Figure R19.** Magnetization measurements on **a**, polycrystalline Si / SiO<sub>2</sub> / Pt (5 nm) / PtMn<sub>3</sub> (7 nm) / MgO (2 nm) / PtMn<sub>3</sub> (10 nm) / Pt (5 nm) / Ru (10 nm) films and **b**, epitaxial Al<sub>2</sub>O<sub>3</sub> (0001) / Pt (7 nm) / PtMn<sub>3</sub> (10 nm) / Pt (3 nm) films. The measurements were conducted using a superconducting quantum interference device (SQUID) 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 backgrounds.

5) It is unclear whether the STEM image in Fig. 1c is from an actual AFMTJ pillar or from the stack before the etching.

**Author response:** The STEM image shown in Fig. 1c is from an actual pillar of 100 nm AATJ device.

#### Revisions in the Manuscript:

We have included the following text in the Methods section to clarify this point:

“Cross-sectional lamella samples processed from a 100 nm AATJ device were prepared by focused ion beam (FIB) using the Thermo Scientific Helios 5 Hydra CX DualBeam Plasma FIB/SEM.”

## Response to Reviewers:

Dear Dr. Thomas,

Thank you for the opportunity to submit a revised version of the manuscript “Octupole-driven spin-transfer torque switching of all-antiferromagnetic tunnel junctions” for publication in *Nature*. We appreciate the time and effort that you and the reviewers dedicated to providing feedback on this manuscript.

We have addressed all comments made by the reviewers. Please see below, in [blue color](#), a point-by-point response to the reviewers’ comments, and a list of the corresponding revisions.

### **Reviewer #3 (Remarks to the Author):**

I thank the authors for their detailed and clear responses, as well as the complementary measurements they have provided. While the authors have presented squid and AHE measurements demonstrating the absence of net magnetization, the manuscript lacks an attempt to measure TMR under a magnetic field, which could have been conducted. Additionally, performing R-V loops at large fields, even at  $\pm 1$  T, would provide a striking demonstration of the device immunity to external fields. This is crucial for corroborating the work and addressing the resilience to external magnetic fields, a significant topic for MRAM applications. I also have difficulties regarding the explanation that PtMn/MgO termination significantly impacts the TMR amplitude and sign, although the theoretical explanation is sound. This is particularly concerning in contrast to the polycrystalline nature of the sample, which should largely average these phenomena, as evidenced by the absence of a magnetic signal in the poly sample. Despite these reservations, the manuscript represents a significant advancement in the field of spintronics. Given the novel contributions and the potential impact of this work, I can recommend the acceptance of this manuscript for publication in *Nature*.

**Author response:** [We sincerely thank the reviewer for the careful review, and for recommending the paper for publication.](#)

[We also thank the reviewer for suggesting that we measure current-induced switching of an AATJ under large magnetic fields, as a demonstration of the device’s immunity to external fields. To address this point, we performed two-terminal current-induced switching measurements under three different out-of-plane externally applied magnetic fields \( \$B\_z\$ \). Figure R1 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.

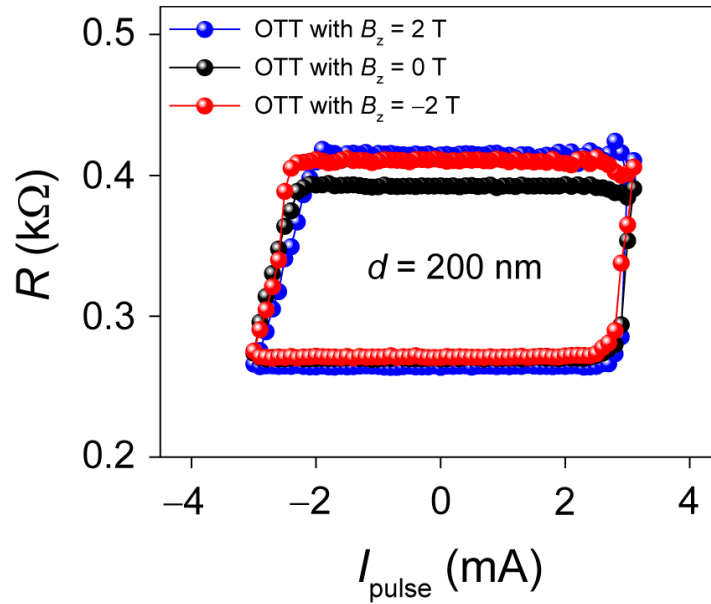


**Figure R1. Two-terminal current-induced switching measurements of an AATJ under different magnetic fields.** Out-of-plane external magnetic fields of +2, 0, -2 T were applied during the switching measurements on a 200 nm-diameter AATJ at room temperature, corresponding to the blue, black and red curves, respectively.

We also appreciate the reviewer's comment regarding the effect of surface termination. As explained in our first response letter, as well as in "Methods: Effect of interface termination on the TMR distribution of AATJ devices", surface termination indeed affects the magnitude and sign of TMR. However, the TMR is primarily determined by the bulk magnetic structure of the electrodes. The interface termination is constrained by the underlying bulk structure of PtMn<sub>3</sub> and therefore cannot adopt arbitrary magnetic configurations. This is why variations in surface termination do not lead to a full cancellation of the TMR, even though they can cause significant device-to-device variations. While our calculated TMR distribution qualitatively captures the experimental observations (see Extended Data Fig. 7), statistics on smaller AATJs, where a larger number of devices remain in a single-domain regime, may help address this point more completely. This will be the subject of future investigation.

#### **Revisions in the Manuscript:**

We have added Fig. R1 and the corresponding discussion to the revised manuscript as a new Supplementary Note 8.

#### **Referee #4 (Remarks to the Author):**

I thank the authors for their detailed responses and for performing additional control experiments. In particular, the comparison between two-terminal (vertical) and three-terminal (in-plane) current injection is a valuable addition and indeed goes in the right direction toward ruling out a dominant spin-orbit torque (SOT) contribution.

However, a key concern from my previous report remains insufficiently addressed.

**Author response:** We sincerely appreciate the reviewer's comments, and confirmation that *"the comparison between two-terminal (vertical) and three-terminal (in-plane) current injection is a valuable addition and indeed goes in the right direction toward ruling out a dominant spin-orbit torque (SOT) contribution."* In the following, we provide a response to the reviewer's remaining comments and a list of the corresponding revisions.

The most decisive experiment I suggested, reversing the magnetic state of the top "polarizer" layer (e.g., via field cooling from above the Néel temperature under opposite field directions) and testing whether the sign of the current-induced switching hysteresis reverses, has not been performed, nor meaningfully discussed in the rebuttal. As I previously stated, such a sign reversal would be expected for an octupole-driven spin-transfer torque (OTT) mechanism, but not for SOT. Importantly, this experiment is not only a direct test of OTT vs SOT, but also closely connected to the concerns raised by Referee #3 regarding field-dependent measurements and the ability to distinguish between different magnetic configurations of the two electrodes. The authors argue that conventional field-induced TMR measurements are not feasible in their system due to the absence of switching up to 9 T and (maybe) the lack of different coercive fields between the two Mn<sub>3</sub>Pt layers. While this explanation is plausible, it further strengthens the case that the proposed field-cooling protocol is the most natural and accessible alternative to probe the relative orientation of the two layers.

**Author response:** We sincerely thank the referee for suggesting this additional experiment.

Before presenting the results of this experiment, we would like to clarify that in fact, the OTT switching polarity is not expected to change as a result of the fixed (i.e., reference) layer direction. This is evident from a comparison to STT switching in a ferromagnetic MTJ: Reversing the magnetization direction of the fixed layer in a ferromagnetic MTJ changes the polarity of its R-H loops, but not that of its R-I loops [see, for example, Table 1 in Grimaldi et al., Nature Nanotechnology 15, 111 (2020)]. The same behavior is expected in the case of two-terminal OTT switching of an AATJ.

Following the reviewer's suggestion, we performed SOT and OTT switching experiments after field-cooling an AATJ from 400 K to 300 K under the out-of-plane external magnetic field of  $\pm 0.2$  T. The results of this experiment are summarized in Fig. R2. First, we do not observe SOT switching within the applied in-plane current range, consistent with the data shown in our previous response letter and Supplementary Note 5 (Fig. S6). Second, the polarity of the two-terminal OTT switching curves obtained under different field-cooling conditions remains the same, as expected from spin-transfer torque switching. Taken together, these results provide further evidence that the mechanism of current-induced two-terminal switching in these devices is indeed OTT, rather than SOT.

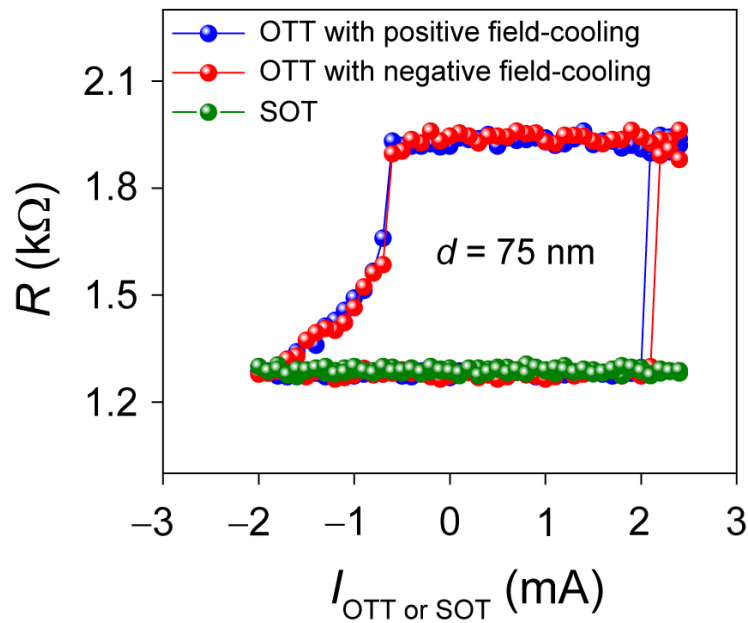


**Figure R2. Comparison of two- and three-terminal switching measurements of an AATJ under different field-cooling conditions.** Three-terminal SOT and two-terminal OTT switching measurements performed within the same device with a 75 nm pillar diameter. Prior to the OTT switching, the AATJ was field-cooled from 400 K to 300 K under an out-of-plane external magnetic field of  $\pm 0.2$  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.

#### Revisions in the Manuscript:

We have added Fig. R2 as a new Supplementary Fig. S7 to the revised manuscript. The corresponding discussion has been added to the revised Supplementary Note 5.

Moreover, beyond SOT, other possible artifacts (for example, thermally driven effects such as transient heating toward or above the Néel temperature, which is around 360 K in this system) are not unambiguously excluded by the newly added experiments. The proposed polarizer-reversal test would also help rule out such scenarios.

Crucially, the requested experiment does not require any redesign of the device structure and should be doable with the already measured devices. Given its decisive nature, the absence of any attempt to address it leaves the central claim of unambiguous OTT switching insufficiently supported. In summary, while the new SOT-control experiment strengthens the manuscript, I believe that the polarizer-reversal test (or an equally decisive alternative) remains essential to conclusively establish the mechanism of switching. Without this, the attribution to OTT is still not fully convincing.

**Author response:** We thank the referee for this important additional comment. To examine the possibility of current-induced Joule heating during the write process, we performed two-terminal current-induced switching measurements at low temperature. Figure R3 shows a current-induced switching curve at 100 K. Notably, a clear current-induced switching behavior is observed at 100 K. Given that this temperature is far below the Néel temperature of PtMn<sub>3</sub>, this result suggests that Joule heating is not the primary mechanism that drives switching.

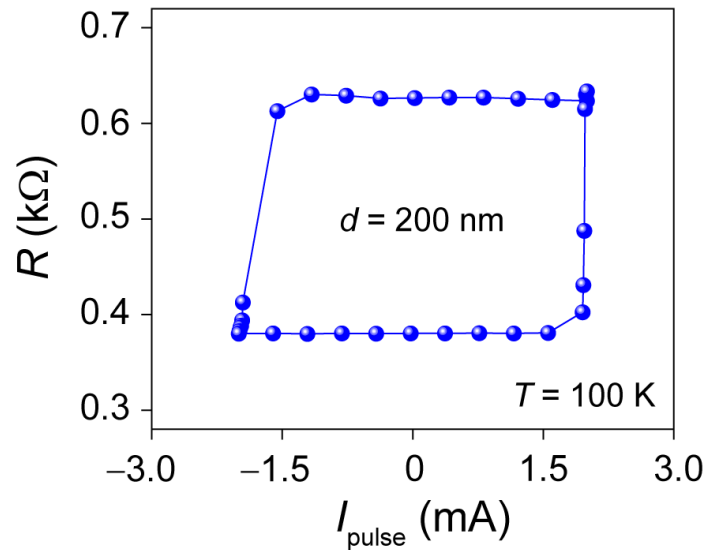


**Figure R3. Two-terminal current-induced switching at low temperature.** The switching measurement was performed at 100 K, without an external magnetic field, for a 200 nm-diameter AATJ.