nature portfolio

Peer Review File

Nearly Perfect Spin Polarization of Noncollinear Antiferromagnets

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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:

Reviewer #1

(Remarks to the Author)
Dear Editor,

The manuscript titled "Nearly Perfect Spin Polarization of Noncollinear Antiferromagnets" presents a study on the spin polarization properties of noncollinear antiferromagnets and their potential applications in spintronics. This work introduces the concept of effective transport spin polarization in noncollinear antiferromagnets, predicting nearly 100% spin polarization in materials such as Mn_3GaN. Additionally, the authors propose an extraordinary tunneling magnetoresistance (ETMR) effect in AFM tunnel junctions using these materials.

However, the proposed mechanism for ETMR in AFMTJs requires further substantiation. The authors should provide a detailed physical mechanism behind this new phenomenon and clarify why a new term, ETMR, is necessary. A comparative analysis with traditional TMR effects in AFM, AM, FM, and other tunnel junctions would be advantageous. Additionally, a thorough symmetry analysis should be included to indicate under what specific conditions the ETMR effect can be expected.

The schematic representation in Fig. 1e, which describes the ETMR effect, appears disconnected from the rest of the manuscript. It lacks integration with TB models or DFT results and is not mentioned that it resembles TMR for p-wave altermagnets, which is also not mentioned or cited.

For the case of Mn_3GaN, the authors calculate the spin component for five bands; however, the band structure or Fermi surface (which are lines in the k-space) of Mn_3GaN is never presented, and the connection with the spin polarization results in Fig. 2 is unclear. Again, these results seem to lack a real connection with Fig. 1e, which describes the ETMR effect.

The manuscript frequently mentions low-decay-rate evanescent states in AFMs. The authors should elaborate on how this quantity was calculated and its impact on the ETMR effect. Furthermore, the effect of SOC on the low-decay-rate evanescent states and the ETMR results should be explained in detail.

The results indicate that the ETMR effect is expected in $Mn_3GaN(1)/SrTiO_3/Mn_3GaN(2)$ AFMTJs. However, in the simulation supercell shown in Figure 3c, periodicity effects of $Mn_3GaN(2)/Mn_3GaN(1)$ interactions may affect the results. The authors should address how they control these unwanted interactions in their DFT calculations and their potential impact on the ETMR results. Additionally, the direction of the Néel vector seems to be missing in this figure.

Figures 1c and 4 could be improved for better readability. A comparative table presenting the ETMR effect in different materials would be helpful for clarity.

The manuscript can be considered for publication once these points are thoroughly addressed.

Sincerely,

Reviewer

Reviewer #2

(Remarks to the Author)
Noncollinear antiferromagnets are ideal material platforms to investigate exotic magnetic/transport phenomena and develop promising applications. In the last decade, various interesting properties have been discovered in this system, such as anomalous Hall effect and Weyl points. Recently. It was found that the nonrelativistic spin texture on the Fermi surface of noncollinear antiferromagnets can result in a strong spin current useful for spintronics. Tunneling magnetoresistance (TMR) has also been demonstrated in antiferromagnetic tunnel junctions (AFMTJs) based on noncollinear antiferromagnets. These achievements without SOC, though exciting, is provoking since the spin is obviously not a good quantum number in noncollinear systems and it's hard to understand how phenomena such as TMR can be realized without the spin conservation.

This manuscript tries to answer this question by introducing an effective spin polarization p_k|| (Eq. 2), which reflects the relative orientations of spins on Fermi surfaces. According to the simple measure, the spins are almost parallel when the |p_k|||=1. Therefore, the spins can be considered as being conserved for such a transverse wavevector, and the associated transport phenomena, such as TMR, can be then well understood at least in the ballistic limit. I therefore consider this is a concept with a fundamental importance for the noncollinear systems.

The authors have performed first principles calculations to show the "perfect" p_k|| exists in many noncollinear antiferromagnets. A AFMTJ based noncollinear AFM Mn3GaN is proposed to host ultra-large TMR. These results indicating the promising application of noncollinear antiferromagnets.

Based on the reason above, I believe this is an important work for understanding the fundamental physics of noncollinear systems and developing the associated applications. I therefore recommend the publication of this work, if the authors can address the comments below:

1. The vertical axis of the Fig. 1c and Fig. S1 is the energy, while the vertical direction of the arrow denotes the y component of the spin. This is confusing and cost me a while to understand the figure. I suggest the authors modify the figure to avoid the confusing.

2. In Fig. 2 and Fig. 4, it looks in some certain areas of the zone, the direction of p_k|| of the adjacent k|| are similar. If this is true, it may indicate these areas host persistent spin texture, which is interesting and important. I suggest the authors confirm it. Maybe they can plot p_k|| using the spherical coordinates, and check whether they are certain areas with similar theta and phi angles.

3. How does SOC influence p_k||? I suggest the authors calculate these in Fig. 2 and Fig. 4 with inclusion of SOC, and present them in SM.

Reviewer #3

(Remarks to the Author)
In this manuscript titled "Nearly Perfect Spin Polarization of Noncollinear Antiferromagnets",
Gautam Gurung et al. discuss the spin polarization in noncollinear magnets, where spin is no longer a good quantum number.
They introduce the spin polarization for noncollinear magnets,
which can be regarded as an extension of the spin polarization discussed in ferromagnets and collinear antiferromagnets.
They calculate the spin polarization using the lattice model to show that a large spin polarization can be realized in noncollinear antiferromagnets.
As an application, they perform TMR calculation with DFT in the Mn3GaN/SrTiO3/Mn3GaN MTJ, where they show a large TMR effect owing to the large spin polarization of a noncollinear antiferromagnet Mn3GaN.

Overall, the realization of a large spin polarization in antiferromagnets is notable, and its application to the TMR effect sounds reasonable.
I will recommend this manuscript for publication after the following points listed below have been clarified.

[1]
As the authors mention, some ferromagnetic materials show a half-metallic electronic structure.
For example, ferromagnetic Heusler compounds will apply to this [e.g., Hong-xi Liu et al., Applied Physics Letters vol. 101, p. 132418 (2012)].
The nearly perfect spin polarization can be understood as the antiferromagnetic version of the half-metallic state,
and thus it will be good to discuss the similarities and differences between the ferromagnetic TMR effect with half-metallic electrode and the antiferromagnetic TMR effect with "Nearly Perfect Spin Polarization".

[2]
How much is the effect of the spin-orbit coupling (SOC) in the TMR effect in the Mn3GaN/SrTiO3/Mn3GaN MTJ?
The authors ignore the effect of the SOC throughout the manuscript.
As they imply for collinear magnets, the SOC will flip the spin, which may largely affect the TMR effect also in the noncollinear magnetic systems.
It would be better if the authors have some discussions on that.

[3]
The in-plane lattice parameters of the MTJ are mentioned,
but how the lattice parameters of the out-of-plane direction, namely, the transmission direction is determined?
The explanation for the out-of-plane lattice parameters, particularly, at the interface between Mn3GaN and SrTiO3, should be given.

[4]
Is the spin polarization in the Mn3GaN/SrTiO3/Mn3GaN MTJ kept at the interface between Mn3GaN and SrTiO3?
The authors show the momentum dependence of the spin-polarization (lowest decay rate) for bulk Mn3GaN (SrTiO3).
The results become more convincing if they also discuss the spin polarization when those materials construct the junction.

[5]
The authors discuss the energy dependence of the TMR effect in the Mn3GaN/SrTiO3/Mn3GaN MTJ.
They show that the transmission at E = E_F - 0.25 eV gives a larger TMR ratio than that at E_F,
where the momentum dependence of the lowest decay rate, "cross pattern", is kept in SrTiO3 as shown in the Supplemental Material.
To claim that the TMR effect at different energies from E_F can be understood in a manner similar to that at E_F,
they should show the spin splitting properties of Mn3GaN at the energy away from E_F as well as the lowest decay rate of insulators.

[6]
This is a minor comment:
sometimes the authors use both of 'noncollinear altermagnet' (e.g., "Apart from GaNMn3, other noncollinear altermagnetic ..."), and 'noncollinear antiferromagnet'.
I understand that there is a recent proposal to extend the concept of altermagnets to noncollinear magnets as the authors also mention in the introduction,
but is there any reason to distinguish these two phrases in this manuscript?
If not, it would be better to use either of two expressions (the latter one is better in my opinion) so that readers clearly understand the content.

[7]
A minor comment: please use either of GaNMn3 or Mn3GaN for the consistency if there are not particular reasons to distinguish these two expressions.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
The manuscript presents one study on the spin polarization properties of noncollinear antiferromagnets, with a particular focus on Mn_3GaN. The authors provide a thorough theoretical framework, supported by density functional theory (DFT) calculations, demonstrating the potential for nearly 100% spin polarization and introducing the concept of extraordinary tunneling magnetoresistance (ETMR) in antiferromagnetic tunnel junctions (AFMTJs). While the manuscript has seen substantial improvements, there are several areas that still require further clarification.

Firstly, the clarity of the figures remains a significant concern. Figures 1c and 1d, in particular, are not sufficiently clear, making it challenging to interpret the data presented. Additionally, Figure 2 illustrates TMR phenomena in both ferromagnetic (FM) and antiferromagnetic (AFM) systems. However, similar TMR effects have already been reported in the literature, including studies published in Physical Review Letters 128.097702, PhysRevX.12.011028, and npj Spintronics2,13(2024). The representation of ETMR in Figure 2 as a novel phenomenon is not entirely convincing, as it appears to resemble a variation of the previously documented d-altermagnetic/d-altermagnetic TMR effect. A more robust argument/representation is needed to clearly distinguish ETMR from these existing effects.

There is also a noticeable disconnection between Figures 3 and 4. Figure 3 presents spin polarization bands for the (111) direction of Mn_3GaN, while Figure 4 discusses transport calculations related to the (001) stacking of the material. The manuscript would benefit from a clearer explanation of how the spin polarization results shown in Figure 3 correlate with the transport properties depicted in Figure 4.

Furthermore, on page 5, the authors mention that some of the “spin textures observed are not symmetry-enforced but are accidental”. This raises concerns about the universality and predictability of the ETMR phenomenon in non collinear AFMs, potentially complicating experimental validation.

Lastly, while the assertion of nearly 100% spin polarization is striking, it may be subject to overestimation under realistic conditions. The authors should consider discussing potential mechanisms (scattering or temperature) that could influence effective spin polarization in practical applications.

Reviewer #2

(Remarks to the Author)
Now I would like to recommend it for publication.

Reviewer #3

(Remarks to the Author)
The authors clearly answered all of my comments. I recommend this revised manuscript for publication.

Version 2:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
The authors have effectively addressed all of my comments.
I recommend this revised manuscript for publication.

RESPONSE TO REVIEWERS

REVIEWER #1

Comment: The manuscript titled "Nearly Perfect Spin Polarization of Noncollinear Antiferromagnets" presents a study on the spin polarization properties of noncollinear antiferromagnets and their potential applications in spintronics. This work introduces the concept of effective transport spin polarization in noncollinear antiferromagnets, predicting nearly 100% spin polarization in materials such as Mn3GaN. Additionally, the authors propose an extraordinary tunneling magnetoresistance (ETMR) effect in AFM tunnel junctions using these materials.

Response: We thank the Reviewer for reading our manuscript and for his/her constructive comments. Below we respond to the comments of this Reviewer.

Comment: However, the proposed mechanism for ETMR in AFMTJs requires further substantiation. The authors should provide a detailed physical mechanism behind this new phenomenon and clarify why a new term, ETMR, is necessary. A comparative analysis with traditional TMR effects in AFM, AM, FM, and other tunnel junctions would be advantageous.

Response: We name the observed effect an *extraordinary* TMR or ETMR, due to its distinctive origin and giant magnitude. For the *ordinary* TMR in conventional MTJs with ferromagnetic (FM) electrodes and in AFMTJs with collinear exchange-split AFM (altermagnetic) electrodes, the spin polarization is defined with respect to the global quantization axis, and the TMR effect can be well understood in terms of spin conservation. In contrast, the ETMR effect relies on the momentum-dependent spin polarization of a non-collinear antiferromagnet, which may have different orientations at different $\boldsymbol{k}_{\parallel}$ (Eq. 2). If such a momentum-dependent spin polarization of conduction channels in a non-collinear antiferromagnet is close to 100%, the ETMR effect in such AFMTJ is expected to become virtually infinite. This is qualitatively similar to TMR in MTJs with ideal half-metallic electrodes, where the fully spin-polarized conduction arises from the presence of only one-spin bands at the Fermi energy, resulting in the global 100% spin polarization of the electric current. However, the mechanism of TMR in half-metallic MTJs is still "ordinary" due to the presence of a global quantization axis. In contrast, in non-collinear antiferromagnets, the fully spin-polarized conduction occurs only within a given $\boldsymbol{k}_{\parallel}$-defined conduction channel with the global spin polarization of electric current being incomplete (or even possibly zero). Therefore, since the mechanism of the giant TMR effect in non-collinear AFMTJs is conceptually different from the ordinary TMR in collinear MTJs and AFMTJs, we dub it "extraordinary."

Following the Reviewer's suggestion, we have provided a comparative analysis of the TMR effects in conventional magnetic tunnel junctions (MTJs), antiferromagnetic tunnel junctions with collinear (altermagnetic) and non-collinear electrodes. Figure R1 illustrates this comparison. Figure R1caption articulates the differences between these effects.

Changes made in the manuscript: Figure R1 has been included in the revised manuscript as Figure 2.

At the end of page 3 and on page 4 (first paragraph), we have added the following statement: "In contrast to the *ordinary* TMR in conventional MTJs with ferromagnetic (FM) electrodes (Fig. 2a,b) and in AFMTJs with collinear exchange-split AFM (altermagnetic) electrodes (Figs. 2c,d), where the spin polarization is defined with respect to the global quantization axis, the predicted TMR effect relies on of the momentum-dependent spin polarization of a non-collinear antiferromagnet, which may have different orientations at different $\boldsymbol{k}_{\parallel}$ (Eq. 2). While such TMR seems qualitatively similar to the ultimately infinite TMR in MTJs with ideal half-metallic electrodes, there is a conceptual difference between the two effects. In nominal half metals, the fully spin-polarized conduction arises from the presence of only one-spin bands at the Fermi energy, resulting in the global 100% spin polarization of the electric current, whereas in non-collinear antiferromagnets, the fully spin-polarized conduction occurs only within a given $\boldsymbol{k}_{\parallel}$-defined conduction channel with the global spin polarization of electric current being incomplete (or even possibly zero). Therefore, since the mechanism of the giant TMR effect in non-collinear AFMTJs is conceptually different from the ordinary TMR in collinear MTJs and AFMTJs, we dub it *extraordinary* TMR (ETMR)."

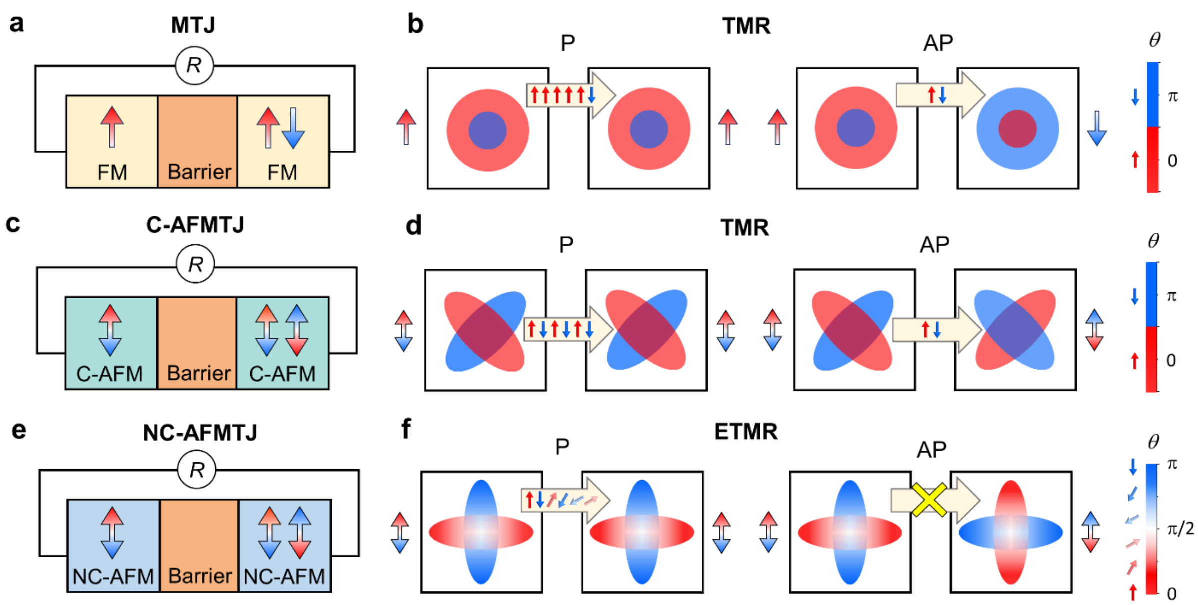


Fig. R1 Schematics of different types of tunnel junctions and TMR effects. (**a**) Schematics of a conventional MTJ where two ferromagnetic (FM) electrodes are separated by a tunnel barrier. (**b**) Mechanism of TMR in a conventional MTJ. Conduction (indicated by block arrows) between left and right FM electrodes for parallel (P) and antiparallel (AP) magnetization (indicated by large arrows). Red and blue circles denote the Fermi surfaces of the ferromagnet for up-spin (small red arrows) and down-spin (small blue arrows) electrons. Angle θ refers to the spin orientation with respect to the magnetization in the left electrode. (**c**) Schematics of an AFMTJ with collinear exchange-split AFM electrodes (C-AFMTJ). (**d**) Mechanism of TMR in C-AFMTJ. Conduction between left and right AFM electrodes for P and AP Néel vectors (indicated by double-arrows). Red and blue ellipses denote the Fermi surfaces of the antiferromagnet for up- and down-spin electrons. Angle θ refers to the spin orientation with respect to the Néel vector in the left electrode. (**e**) Schematics of an AFMTJ with non-collinear AFM electrodes (NC-AFMTJ). (**f**) Mechanism of ETMR. The NC-AFM electrodes have fully spin-polarized conduction channels within the area indicated by crossing ellipses with the spin polarization vector having different orientation θ (indicated by varying color). Electrons in these channels can efficiently tunnel through a tunnel barrier due to its low-decay-rate evanescent states supporting transmission. Matching (mismatching) of the 100% polarized conduction channels in the two electrodes for the P (AP) state produces ETMR.

Comment: Additionally, a thorough symmetry analysis should be included to indicate under what specific conditions the ETMR effect can be expected.

Response: Unfortunately, pure symmetry-based considerations do not allow establishing rules for the emergence of the ETMR effect. This is because, the appearance of nearly 100% conduction channels on the Fermi surface of non-collinear antiferromagnets is controlled by their intricate electronic band structure. The latter is determined by the magnitudes of electrostatic and exchange interactions, which are different for different antiferromagnets, even those which have the same symmetry. For example, from Figure 4, one can see that while Mn_3Ir and Mn_3Pt (and similarly, Mn_3NiN and Mn_3SnN) have the same magnetic space group symmetry, they exhibit very different distribution of the spin polarization in the 2D Brillouin zone. This fact can also be seen from the Kagome lattice model (see Supplementary Figure S1), where the spin polarization changes notably with the parameters of the model, while the symmetry remains the same.

Comment: The schematic representation in Fig. 1e, which describes the ETMR effect, appears disconnected from the rest of the manuscript. It lacks integration with TB models or DFT results and is not mentioned that it resembles TMR for p-wave altermagnets, which is also not mentioned or cited.

Response: In response to this Reviewer's comment, we have revised Figure 1e to make it clearer and be better connected to our DFT results. The revised figure is displayed above as Figure R1f and shows schematically fully spin-polarized conduction channels within the area indicated by crossing ellipses with the spin polarization vector having different orientation θ (indicated by varying color). The angular dependence of the spin-polarization vector distinguishes non-collinear antiferromagnets from altermagnets (where only two spin states, up or down, are available). In this regard, p-wave altermagnets mentioned by the Reviewer are irrelevant to this discussion. This schematic representation is reminiscent to our DFT results where fully spin-polarized conduction channels have different spin orientation depending on the wave vector $\boldsymbol{k}_{\parallel}$ (see, e.g., Figs. 3e,f in the revised manuscript).

Changes made in the manuscript: Figure R1f has been included in the revised manuscript as Figure 2f.

Comment: For the case of Mn3GaN, the authors calculate the spin component for five bands; however, the band structure or Fermi surface (which are lines in the k-space) of Mn3GaN is never presented, and the connection with the spin polarization results in Fig. 2 is unclear. Again, these results seem to lack a real connection with Fig. 1e, which describes the ETMR effect.

Response: The spin-dependent band structure and Fermi surfaces for each of five bands of Mn_3GaN is calculated and plotted in Supplementary Figures S2 and S3. The spin-polarization data shown in Figure 2 (Fig. 3 in the revised manuscript) directly follows from these results.

The connection between the calculated spin polarization in Figure 3 and revised Figure1e (Fig. 2f in the revised manuscript) is now evident. Specifically, the red cross area in Figure 3f displays the region of nearly 100% spin polarization. The related highly spin-polarized area in Figure 2f is represented by two crossing ellipses, whose varying color indicates an angle θ between the polarization vector and a global quantization axis. This angle is $\boldsymbol{k}_{\parallel}$ dependent and thus reminiscent to our DFT results shown in Figure 3e, where the x-, y-, and z-components of spin polarization vector depend on $\boldsymbol{k}_{\parallel}$.

Comment: The manuscript frequently mentions low-decay-rate evanescent states in AFMs. The authors should elaborate on how this quantity was calculated and its impact on the ETMR effect.

Response: Following Reviewer's suggestion, we have added a paragraph in *Methods* to describe details of calculation of the decay rates of evanescent states in $SrTiO_3$. Also, additional information about the complex band structure of $SrTiO_3$ can be found in section G of the revised Supplementary Information.

Changes made in the manuscript: On page 8 of the revised manuscript, a new paragraph is added to *Methods* to describe details of calculation of the decay rates of evanescent states in $SrTiO_3$, as follows: "The decay rates of evanescent states in $SrTiO_3$ are obtained from its complex band structure calculated using PWCOND. An arbitrary wave vector consists of a component parallel to the interface, $\boldsymbol{k}_{\parallel}$, which is conserved during tunneling, and a component perpendicular to the interface, $k_{\boldsymbol{z}}$. For each $\boldsymbol{k}_{\parallel}$, we calculate the dispersion relation $E = E(k_{\boldsymbol{z}})$, allowing complex $k_z = q + i\kappa$. The imaginary part κ is the decay rate, so that the corresponding wave functions decay as $\sim e^{-\kappa z}$."

Comment: Furthermore, the effect of SOC on the low-decay-rate evanescent states and the ETMR results should be explained in detail.

Response: Following the Reviewer's suggestion, we have explored the effects of spin-orbit coupling (SOC) on the spin polarization of bulk Mn_3GaN (001) and ETMR in $Mn_3GaN/SrTiO_3/Mn_3GaN$ (001) AFMTJs using fully relativistic PAW pseudopotentials in the DFT calculations. The results for the spin polarization of bulk Mn_3GaN (001) in the presence of SOC are shown in Figure R2a in comparison to Figure R2b which is obtained without including SOC in the calculation (the same as Fig. 3f in the revised manuscript). We see that the SOC effect on the spin polarization is insignificant and does not qualitatively change the overall picture. Importantly it preserves the cross-pattern feature of $p_{\boldsymbol{k}_{\parallel}}$ which is important for ETMR in $Mn_3GaN/SrTiO_3/Mn_3GaN$ (001) AFMTJs. Figures R2d,f show the results for the $\boldsymbol{k}_{\parallel}$-resolved transmission in $Mn_3GaN/SrTiO_3/Mn_3GaN$ (001) AFMTJs for parallel (Fig. R2d) and antiparallel (Fig. R2f) alignment of the Néel vector in the Mn_3GaN electrodes calculated in the presence of SOC. By comparing these results to those where SOC is neglected (Figs. R2d,f, the same as Figs. 4e,f in the revised manuscript), we see that they look very similar. There is though some enhancement of transmission due to SOC around the $\bar{\Gamma}$ point in the 2DBZ for the antiparallel alignment of the Néel vector (compare Figs. R2f and R2e), which leads to reduction of the ETMR ratio from 1.8×10^4% to 2.6×10^3%. Despite this reduction, the ETMR effect remains huge. We conclude therefore that SOC does not qualitatively change the main conclusions of our work. We note however that the SOC effects may be more pronounced in AFMTJs based on non-collinear antiferromagnets with heavy elements like Mn_3Ir and Mn_3Pt.

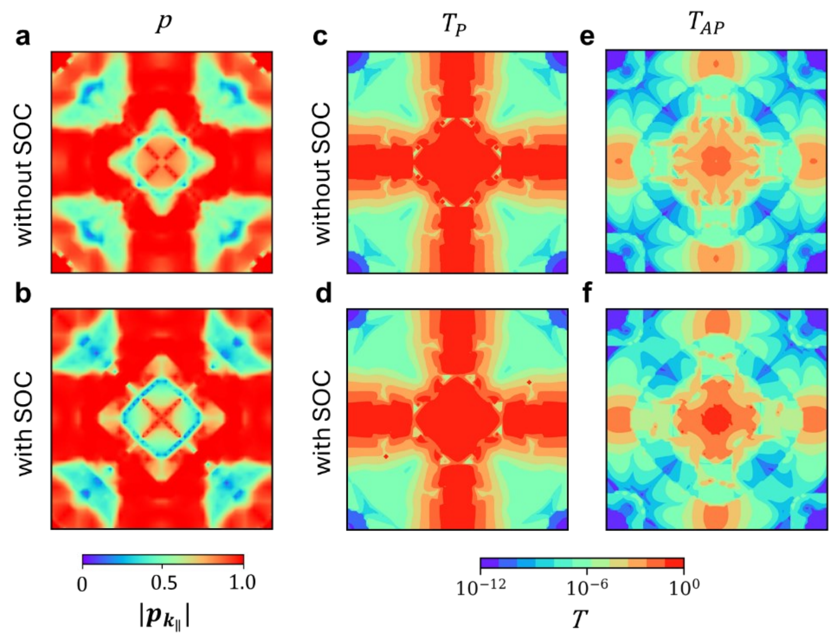


Fig. R2 Effects of SOC on the spin polarization of bulk Mn_3GaN (001) and ETMR in $Mn_3GaN/SrTiO_3/Mn_3GaN$ (001) AFMTJs. (**a**,**b**) $\boldsymbol{k}_\parallel$-resolved spin polarization of bulk Mn_3GaN (001) calculated without (**a**) and with (**b**) SOC. (**c**-**f**) $\boldsymbol{k}_\parallel$-resolved transmission in the 2DBZ for the AFMTJ for parallel (P) (**c**,**d**) and antiparallel (AP) (**e**,**f**) alignment of the Néel vector in the Mn_3GaN electrodes calculated without (**c**,**e**) and with (**d**,**f**) SOC.

Responding for the comment of the Reviewer regarding the effect of SOC on the low-decay-rate evanescent states, we have performed calculations of the complex structure of $SrTiO_3$ from which we extracted the lowest decay rates as a function of the transverse wave vector. The results are shown in Figure R3 both with and without SOC. As is evident, the results are nearly identical. This is not unexpected because SOC is weak in $SrTiO_3$.

Changes made in the manuscript: In the revised manuscript (page 7, the second paragraph), we have included the following: "We also note that our calculations neglect the effects of spin-orbit coupling which may lead to spin mixing affecting the spin polarization and ETMR. We find, however, that taking spin-orbit coupling into account does not change our main conclusions. This is evident from Supplementary Section H, which shows the effects of spin-orbit coupling on the spin polarization of bulk Mn_3GaN (Supplementary Figs. S8a,b) and ETMR in $Mn_3GaN/SrTiO_3/Mn_3GaN$ AFMTJ (Supplementary Figs. S8c-f). While quantitively SOC reduces the ETMR ratio from 1.8×10^4% to 2.6×10^3%, qualitatively the ETMR effect remains huge driven by the nearly perfect spin polarization of Mn_3GaN."

In *Methods*, the following statement has been added: "The effects of spin-orbit interaction on spin polarization of bulk Mn_3GaN (001) and ETMR in $Mn_3GaN/SrTiO_3/Mn_3GaN$ (001) AFMTJs are evaluated using fully relativistic PAW pseudopotentials and discussed in Supplementary Information (Section H), while in the main text, these effects are neglected."

In the revised Supplementary Information, we have added a new section H which discusses the effects of spin-orbit interaction on spin polarization of bulk Mn_3GaN (001) and ETMR in $Mn_3GaN/SrTiO_3/Mn_3GaN/SrTiO_3$ (001) AFMTJs. This section includes a new Figure S8, which is a copy of Figure R2 above.

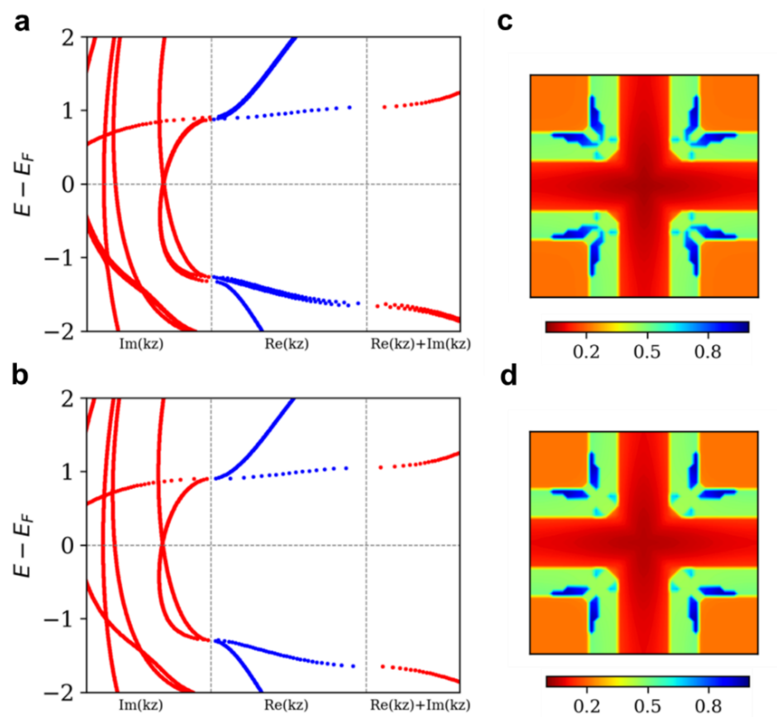


Fig. R3 (**a**,**b**) Complex band structure of $SrTiO_3$ linking real bands along the high-symmetry Γ-Z line and complex bands at the $\bar{\Gamma}$ and $\bar{Z}$ points calculated with (**a**) and without (**b**) spin-orbit coupling. (**c**,**d**) Color maps of the lowest decay rate in the 2DBZ calculated at the Fermi energy of the $Mn_3GaN/SrTiO_3/Mn_3GaN$ AFMTJ with (**b**) and without (**d**) spin-orbit coupling. Fig. R3d is the same as Fig. 4b in the main text but uses a slightly different color scale.

Comment: The results indicate that the ETMR effect is expected in Mn3GaN(1)/SrTiO3/ Mn3GaN(2) AFMTJs. However, in the simulation supercell shown in Figure 3c, periodicity effects of Mn3GaN(2)/Mn3GaN(1) interactions may affect the results. The authors should address how they control these unwanted interactions in their DFT calculations and their potential impact on the ETMR results. Additionally, the direction of the Néel vector seems to be missing in this figure.

Response: For parallel Néel vectors in the Mn_3GaN electrodes, the simulation supercell has periodic boundary conditions such that $Mn_3GaN(2)/Mn_3GaN(1)$ interface represents bulk stacking of Mn_3GaN, i.e., the supercell can be simply represented as $Mn_3GaN/SrTiO_3$. Therefore, there are no additional perturbations associated with this interface. For antiparallel Néel vectors, the simulation supercell is doubled, so that it represents a $Mn_3GaN(1)/SrTiO_3/Mn_3GaN(2)/SrTiO_3$ heterostructure, where $Mn_3GaN(1)$ and $Mn_3GaN(2)$ have antiparallel Néel vectors. Again, there are no additional perturbations associated with this interface. For transmission calculations, the $Mn_3GaN/SrTiO_3/Mn_3GaN$ structure shown in Figure 3c is considered as the scattering region that is ideally attached on both sides to semi-infinite Mn_3GaN leads.

For the convenience of the Referee, in Figure R4, we schematically depict supercells which are used for atomic relaxations and self-consistent calculations of the electronic structure (left panels in Fig. R4) for parallel (Fig. R4(a)) and antiparallel (Fig. R4(b)) Néel vectors and AFMTJ structures that are used for transport calculations (right panels in Fig. R4).

The Néel vectors in this figure lie in the [111] plane and can be parallel or antiparallel. Since it is hard to depict them in the planer plot, we have articulated this in the figure caption.

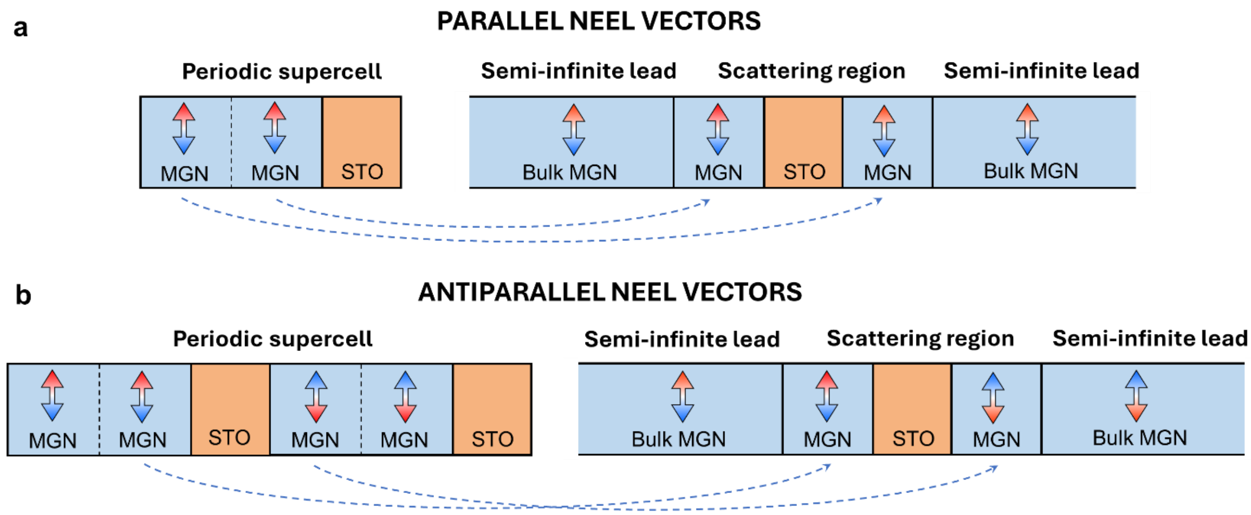


Fig. R4 (**a,b**) Schematic of the supercells which are used for atomic relaxations and self-consistent calculations of the electronic stature for parallel (**a**) and antiparallel (**b**) Néel vectors (left panels) and AFMTJ structures that are used for transport calculations (right panels). Block double arrows indicate Néel vectors. Dashed arrows indicate how the relaxed supercell structures are used to build the scattering region for transport calculations.

Changes made in the manuscript: In *Methods* (in the first paragraph in *DFT calculations*), we have clarified that our calculations involved "$Mn_3GaN/SrTiO_3$ (001) and $Mn_3GaN/SrTiO_3$/ $Mn_3GaN/SrTiO_3$ (001) supercells used for AFMTJs with parallel and antiparallel Néel vector, respectively."

In the Figure 4 (c) caption, we have added the following statement: "The Néel vectors of the Mn_3GaN electrodes lie in the [111] plane (not shown) and can be parallel or antiparallel."

Comment: Figures 1c and 4 could be improved for better readability.

Response: Following the Reviewer's suggestion, we have revised Figure 1c. Instead of using vectors to depict spin orientations, we have employed different color maps for the spin polarization vector magnitude (Fig. 1c in the revised manuscript) and polar angle (Fig. 1d in the revised manuscript) as functions of Fermi energy E_F and wave vector k_y. For the convenience of the Reviewer, the revised Figures are copied below as Figures R5a,b.

Regarding Figure 4 (Fig. 5 in the revised manuscript), we believe that it clearly represents effective spin polarization of different noncollinear antiferromagnets with no ambiguities.

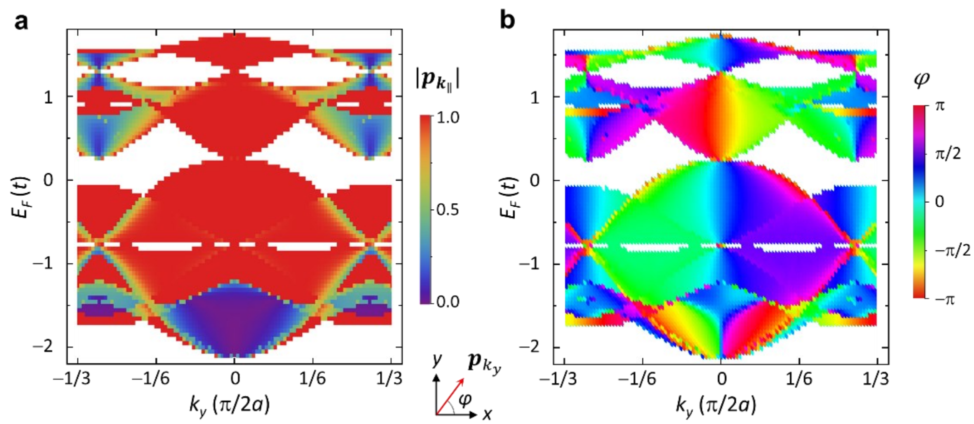


Fig. R5 (**a,b**) Map of the spin polarization vector $\boldsymbol{p}_{k_y}$ as a function of k_y and E_F for the Kagome lattice for $\Delta/t = 1.5$. $\boldsymbol{p}_{k_y}$ vectors lie in the *x-y* plane ($p_z = 0$). Their magnitudes $p_k \equiv |\boldsymbol{p}_k|$ (**a**) and polar angles φ (**b**) are depicted in color.

Changes made in the manuscript: Figures R5a,b have been included in the revised manuscript as Figures 1c,d. The respective text explaining these figures has been added to paragraph 2 on page 3: "Figures 1c,d show magnitudes $p_k \equiv |\boldsymbol{p}_k|$ and polar angles φ..."

Figure 1S in Supplementary Information has been modified accordingly to include the revised color maps for the spin polarization vector magnitude (Supplementary Figs. S1d-f) and polar angle (Supplementary Figs. 1g-i).

Comment: A comparative table presenting the ETMR effect in different materials would be helpful for clarity.

Response: At the end of the manuscript, we discuss spin-polarized properties of different non-collinear antiferromagnets and a possibility of observing ETMR effects using these antiferromagnets. We emphasize the fact that the ETMR effect is not only controlled by the spin polarization of the antiferromagnets, but also by the evanescent states of tunneling barrier and variation of their decay rates as a function of transverse wave vector in the 2D Brillouin zone. As a result, different non-collinear antiferromagnets are expected to have very different magnetoresistive responses depending on the properties of the insulator. Thus, the comparison of the ETMR effect for different materials requires studies of various antiferromagnet-insulator pairs rather than solely antiferromagnets. While these investigations would be interesting, they clearly lie beyond the scope of our manuscript.

Comment: The manuscript can be considered for publication once these points are thoroughly addressed.

Response: We have addressed all comments by the Reviewer as thoroughly as we could and thus hope that the manuscript can now be accepted for publication.

REVIEWER #2

Comment: Noncollinear antiferromagnets are ideal material platforms to investigate exotic magnetic/transport phenomena and develop promising applications. In the last decade, various interesting properties have been discovered in this system, such as anomalous Hall effect and Weyl points. Recently. It was found that the nonrelativistic spin texture on the Fermi surface of noncollinear antiferromagnets can result in a strong spin current useful for spintronics. Tunneling magnetoresistance (TMR) has also been demonstrated in antiferromagnetic tunnel junctions (AFMTJs) based on noncollinear antiferromagnets. These achievements without SOC, though exciting, is provoking since the spin is obviously not a good quantum number in noncollinear systems and it's hard to understand how phenomena such as TMR can be realized without the spin conservation.

This manuscript tries to answer this question by introducing an effective spin polarization p_k|| (Eq. 2), which reflects the relative orientations of spins on Fermi surfaces. According to the simple measure, the spins are almost parallel when the |p_k|||=1. Therefore, the spins can be considered as being conserved for such a transverse wavevector, and the associated transport phenomena, such as TMR, can be then well understood at least in the ballistic limit. I therefore consider this is a concept with a fundamental importance for the noncollinear systems.

The authors have performed first principles calculations to show the "perfect" p_k|| exists in many noncollinear antiferromagnets. A AFMTJ based noncollinear AFM Mn3GaN is proposed to host ultra-large TMR. These results indicating the promising application of noncollinear antiferromagnets.

Based on the reason above, I believe this is an important work for understanding the fundamental physics of noncollinear systems and developing the associated applications. I therefore recommend the publication of this work, if the authors can address the comments below:

Response: We thank the Reviewer for the thorough reading of our manuscript and for his/her constructive comments. We are delighted to learn that the Reviewer finds our work "*important for understanding the fundamental physics of noncollinear systems and developing the associated applications*" and recommends publication of our manuscript. Below we respond to the comments of this Reviewer.

Comment: 1. The vertical axis of the Fig. 1c and Fig. S1 is the energy, while the vertical direction of the arrow denotes the y component of the spin. This is confusing and cost me a while to understand the figure. I suggest the authors modify the figure to avoid the confusing.

Response: Following the Reviewer's suggestion, we have revised Figure 1c. Instead of using vectors to depict spin orientations, we have employed different color maps for the spin polarization vector magnitude (Fig. 1c in the revised manuscript) and polar angle (Fig. 1d in the revised manuscript) as functions of Fermi energy E_F and wave vector k_y. For the convenience of the Reviewer, the revised Figures are copied below as Figures R6a,b.

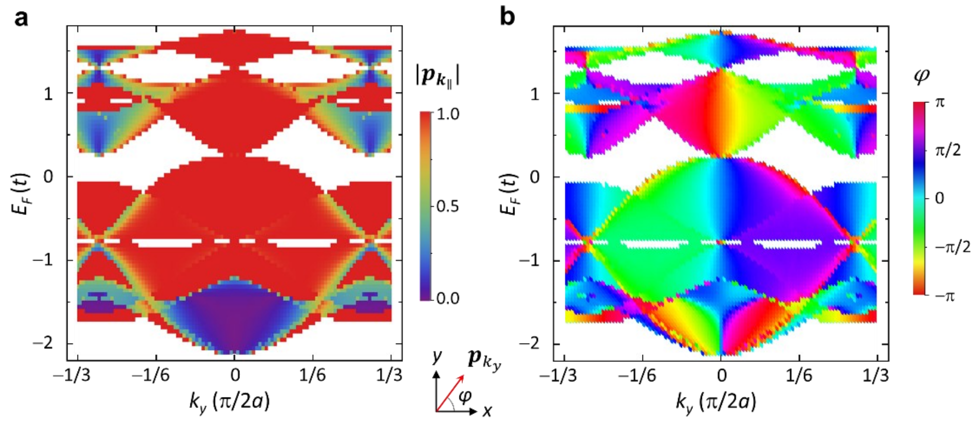


Fig. R6 (**a,b**) Map of the spin polarization vector $\boldsymbol{p}_{k_y}$ as a function of k_y and E_F for the Kagome lattice for $\Delta/t = 1.5$. $\boldsymbol{p}_{k_y}$ vectors lie in the *x*-*y* plane ($p_z = 0$). Their magnitudes $p_k \equiv |\boldsymbol{p}_k|$ (**a**) and polar angles φ (**b**) are depicted in color.

Changes made in the manuscript: Figures R6a,b have been included in the manuscript as Figures 1c,d. The respective text explaining these figures has been added to paragraph 2 on page 3 of the revised manuscript: "Figures 1c,d show magnitudes $p_k \equiv |\boldsymbol{p}_k|$ and polar angles φ ..."

Figure 1S in Supplementary Information has been modified accordingly to include the revised color maps for the spin polarization vector magnitude (Supplementary Figs. S1d-f) and polar angle (Supplementary Figs. 1g-i).

Comment: 2. In Fig. 2 and Fig. 4, it looks in some certain areas of the zone, the direction of p_k|| of the adjacent k|| are similar. If this is true, it may indicate these areas host persistent spin texture, which is interesting and important. I suggest the authors confirm it. Maybe they can plot p_k|| using the spherical coordinates, and check whether they are certain areas with similar theta and phi angles.

Response: We agree that the possibility of the persistent spin texture in non-collinear antiferromagnets is interesting. To check if this is the case, we have color mapped azimuthal θ and polar φ angles of the spin polarization vector of Mn_3GaN in the (001) 2D Brillouin zone and displayed them in Figure R7. It is seen that in the areas indicated by dashed lines, both θ and φ angles remain nearly constant revealing the same orientation of the polarization vector. This is the signature of the persistent spin texture. We note however that the persistent spin texture found for non-collinear antiferromagnets is not symmetry enforced like that predicted by Tao et al., Nat. Comm. 9, 2763 (2018), but rather "accidental," resulting from specific type of interactions intrinsic to a particular antiferromagnet.

Changes made in the manuscript: On page 5 (below Fig. 4) of the revised manuscript the following text has been added: "Interestingly, we observe that some regions in the 2DBZ of Mn_3GaN (001) exhibit a persistent spin texture, where spins in relatively broad regions of the 2DBZ are pointing in the same direction (Supplementary Fig. S4[26]). We note however that this persistent spin texture is not symmetry enforced like that predicted in Ref. 32, but rather "accidental," resulting from specific type of interactions intrinsic to a particular non-collinear antiferromagnet."

New reference 32 was included in the list of references: 32. Tao, L. L. & Tsymbal, E. Y., Persistent spin texture enforced by symmetry. Nat. Commun. 9, 2763 (2018).

New section D has been added to the revised Supplementary Information that discussed the emergence of the persistent spin texture and includes Figure R7 as Figure S4.

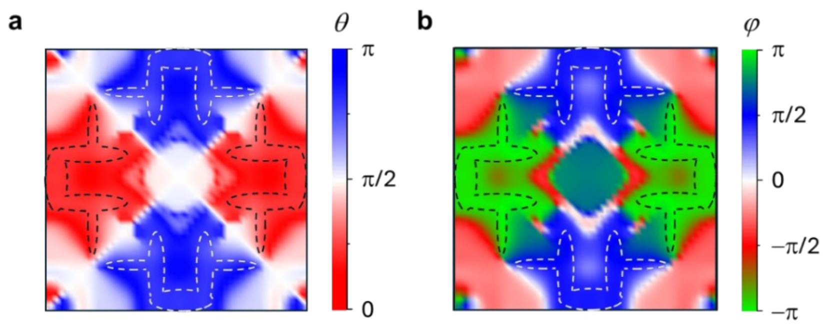


Fig. R7 (a,b) Azimuthal θ (**a**) and polar φ (**b**) angles of the spin polarization vector of Mn_3GaN color mapped in the (001) 2D Brillouin zone. Dashed lines (both black and white) indicate areas with persistent spin texture.

Comment: 3. How does SOC influence p_k||? I suggest the authors calculate these in Fig. 2 and Fig. 4 with inclusion of SOC, and present them in SM.

Response: Following the Reviewer's suggestion, we have explored the effects of spin-orbit coupling (SOC) on the spin polarization of bulk Mn_3GaN (001) and ETMR in $Mn_3GaN/SrTiO_3/Mn_3GaN$ (001) AFMTJs using fully relativistic PAW pseudopotentials in the DFT calculations. The results for the spin polarization of bulk Mn_3GaN (001) in the presence of SOC are shown in Figure R8a in comparison to Figure R8b which is obtained without including SOC in the calculation (the same as Fig. 3f in the revised manuscript). We see that the SOC effect on the spin polarization is insignificant and does not qualitatively change the overall picture. Importantly it preserves the cross-pattern feature of $p_{\boldsymbol{k}_\parallel}$ which is important for ETMR in $Mn_3GaN/SrTiO_3/Mn_3GaN$ (001) AFMTJs. Figures R8d,f show the results for the $\boldsymbol{k}_\parallel$-resolved transmission in $Mn_3GaN/SrTiO_3/Mn_3GaN$ (001) AFMTJs for parallel (Fig. R8d) and antiparallel (Fig. R8f) alignment of the Néel vector in the Mn_3GaN electrodes calculated in the presence of SOC. By comparing these results to those where SOC is neglected (Figs. R8d,f, the same as Figs. 4e,f in the revised manuscript), we see that they look very similar. There is though some enhancement of transmission due to SOC around the $\bar{\Gamma}$ point in the 2DBZ for the antiparallel alignment of the Néel vector (compare Figs. R8f and R8e), which leads to reduction of the ETMR ratio from $1.8\times10^4\%$ to $2.6\times10^3\%$. Despite this reduction, the ETMR effect remains huge. We conclude therefore that SOC does not qualitatively change the main conclusions of our work. We note however that the SOC effects may be more pronounced in AFMTJs based on non-collinear antiferromagnets with heavy elements like Mn_3Ir and Mn_3Pt.

Changes made in the manuscript: In the revised manuscript (page 7, the second paragraph), we have included the following: "We also note that our calculations neglect the effects of spin-orbit coupling which may lead to spin mixing affecting the spin polarization and ETMR. We find, however, that taking spin-orbit coupling into account does not change our main conclusions. This is evident from Supplementary Section H, which shows the effects of spin-orbit coupling on the spin polarization of bulk Mn_3GaN (Supplementary Figs. S8a,b) and ETMR in $Mn_3GaN/SrTiO_3/Mn_3GaN$ AFMTJ (Supplementary Figs. S8c-f). While quantitively SOC reduces the ETMR ratio from $1.8\times10^4\%$ to $2.6\times10^3\%$, qualitatively the ETMR effect remains huge driven by the nearly perfect spin polarization of Mn_3GaN."

In *Methods* (the second paragraph in *DFT calculations*), the following statement has been added: "The effects of spin-orbit interaction on spin polarization of bulk Mn_3GaN (001) and ETMR in $Mn_3GaN/SrTiO_3/Mn_3GaN$ (001) AFMTJs are evaluated using fully relativistic PAW pseudopotentials and discussed in Supplementary Information (Section H), while in the main text, these effects are neglected."

In the revised Supplementary Information, we have added a new section H which discusses the effects of spin-orbit interaction on spin polarization of bulk Mn_3GaN (001) and ETMR in $Mn_3GaN/SrTiO_3/Mn_3GaN/SrTiO_3$ (001) AFMTJs. This section includes the new Figure S8, which is a copy of Figure R8 below.

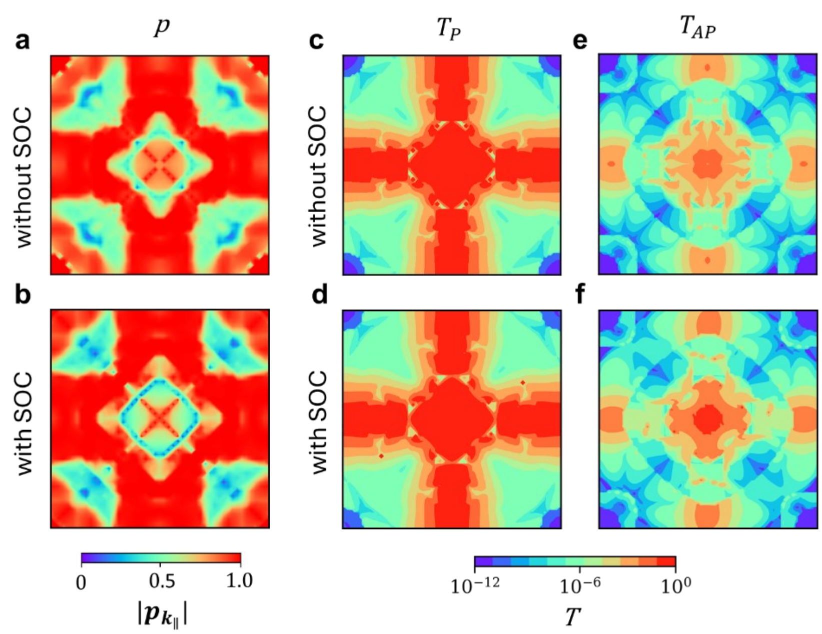


Fig. R8 Effects of SOC on the spin polarization of bulk Mn_3GaN (001) and ETMR in $Mn_3GaN/SrTiO_3/Mn_3GaN$ (001) AFMTJs. (**a**,**b**) $\boldsymbol{k}_\parallel$-resolved spin polarization of bulk Mn_3GaN (001) calculated without (**a**) and with (**b**) SOC. (**c-f**) $\boldsymbol{k}_\parallel$-resolved transmission in the 2DBZ for the AFMTJ for parallel (P) (**c**,**d**) and antiparallel (AP) (**e**,**f**) alignment of the Néel vector in the Mn_3GaN electrodes calculated without (**c**,**e**) and with (**d**,**f**) SOC.

REVIEWER #3

Comment: In this manuscript titled "Nearly Perfect Spin Polarization of Noncollinear Antiferromagnets", Gautam Gurung et al. discuss the spin polarization in noncollinear magnets, where spin is no longer a good quantum number. They introduce the spin polarization for noncollinear magnets, which can be regarded as an extension of the spin polarization discussed in ferromagnets and collinear antiferromagnets. They calculate the spin polarization using the lattice model to show that a large spin polarization can be realized in noncollinear antiferromagnets. As an application, they perform TMR calculation with DFT in the Mn3GaN/SrTiO3/Mn3GaN MTJ, where they show a large TMR effect owing to the large spin polarization of a noncollinear antiferromagnet Mn3GaN.

Overall, the realization of a large spin polarization in antiferromagnets is notable, and its application to the TMR effect sounds reasonable. I will recommend this manuscript for publication after the following points listed below have been clarified.

Response: We thank the Reviewer for the thorough reading of our manuscript and for his/her constructive comments. We are pleased by the fact that the Reviewer finds "*the realization of a large spin polarization in antiferromagnets … notable, and its application to the TMR effect … reasonable*" and conditionally recommends publication of our manuscript. Below we respond to the comments of this Reviewer.

Comment: [1] As the authors mention, some ferromagnetic materials show a half-metallic electronic structure. For example, ferromagnetic Heusler compounds will apply to this [e.g., Hong-xi Liu et al., Applied Physics Letters vol. 101, p. 132418 (2012)]. The nearly perfect spin polarization can be understood as the antiferromagnetic version of the half-metallic state, and thus it will be good to discuss the similarities and differences between the ferromagnetic TMR effect with half-metallic electrode and the antiferromagnetic TMR effect with "Nearly Perfect Spin Polarization".

Response: We name the observed effect an *extraordinary* TMR or ETMR, due to its distinctive origin and giant magnitude. In contrast to the TMR effects in conventional MTJs with ferromagnetic (FM) electrodes and in AFMTJs with collinear exchange-split AFM (altermagnetic) electrodes, where the spin polarization is defined with respect to the global quantization axis, the ETMR effect relies on of the momentum-dependent spin polarization of a non-collinear antiferromagnet, which may have different orientations at different $\boldsymbol{k}_{\parallel}$ (Eq. 2). If such a momentum-dependent spin polarization of conduction channels in a non-collinear antiferromagnet is close to 100%, the ETMR effect in such AFMTJ is expected to become virtually infinite. As the Reviewer correctly notices, this behavior is qualitatively similar to TMR in MTJs with ideal half-metallic electrodes; however, while in nominal half metals, the fully spin-polarized conduction arises from the presence of only one-spin bands at the Fermi energy, resulting in the global 100% spin polarization of the electric current, in non-collinear antiferromagnets, the fully spin-polarized conduction occurs only within a given $\boldsymbol{k}_{\parallel}$-defined conduction channel with the global spin polarization of electric current being incomplete (or even possibly zero).

Changes made in the manuscript: On page 4 (the first paragraph), we have added the following statement: "While such TMR seems qualitatively similar to the ultimately infinite TMR in MTJs with ideal half-metallic electrodes, there is a conceptual difference between the two effects. In nominal half metals, the fully spin-polarized conduction arises from the presence of only one-spin bands at the Fermi energy, resulting in the global 100% spin polarization of the electric current, whereas in non-collinear antiferromagnets, the fully spin-polarized conduction occurs only within a given $\boldsymbol{k}_{\parallel}$-defined conduction channel with the global spin polarization of electric current being incomplete (or even possibly zero)."

Comment: [2] How much is the effect of the spin-orbit coupling (SOC) in the TMR effect in the Mn3GaN/SrTiO3/Mn3GaN MTJ? The authors ignore the effect of the SOC throughout the manuscript. As they imply for collinear magnets, the SOC will flip the spin, which may largely affect the TMR effect also in the noncollinear magnetic systems. It would be better if the authors have some discussions on that.

Response: Following the Reviewer's suggestion, we have explored the effects of spin-orbit coupling (SOC) on the spin polarization of bulk Mn_3GaN (001) and ETMR in $Mn_3GaN/SrTiO_3/Mn_3GaN$ (001) AFMTJs using fully relativistic PAW pseudopotentials in the DFT calculations. The results for the spin polarization of bulk Mn_3GaN (001) in the presence of SOC are shown in Figure R9a in comparison to Figure R9b which is obtained without including SOC in the calculation (the same as Fig. 3f in the revised manuscript). We see that the SOC effect on the spin polarization is insignificant and does not qualitatively change the overall picture. Importantly it preserves the cross-pattern feature of $p_{\boldsymbol{k}_{\parallel}}$ which is important for ETMR in $Mn_3GaN/SrTiO_3/Mn_3GaN$ (001) AFMTJs. Figures R9d,f show the results for the $\boldsymbol{k}_{\parallel}$-resolved transmission in $Mn_3GaN/SrTiO_3/Mn_3GaN$ (001) AFMTJs for parallel (Fig. R9d) and antiparallel (Fig. R9f) alignment of the Néel vector in the Mn_3GaN electrodes calculated in the presence of SOC. By comparing these results to those where SOC is neglected (Figs. R9d,f, the same as Figs. 4e,f in the revised manuscript), we see that they look very similar. There is though some enhancement of transmission due to SOC around the $\bar{\Gamma}$ point in the 2DBZ for the antiparallel alignment of the Néel vector (compare Figs. R9f and R9e), which leads to reduction of the ETMR ratio from 1.8×10^4% to 2.6×10^3%. Despite this reduction, the ETMR effect remains huge. We conclude therefore that SOC does not qualitatively change the main conclusions of our work. We note however that the SOC effects may be more pronounced in AFMTJs based on non-collinear antiferromagnets with heavy elements like Mn_3Ir and Mn_3Pt.

Changes made in the manuscript: In the revised manuscript (page 7, the second paragraph), we have included the following: "We also note that our calculations neglect the effects of spin-orbit coupling which may lead to spin mixing affecting the spin polarization and ETMR. We find, however, that taking spin-orbit coupling into account does not change our main conclusions. This is evident from Supplementary Section H, which shows the effects of spin-orbit coupling on the spin polarization of bulk Mn_3GaN (Supplementary Figs. S8a,b) and ETMR in $Mn_3GaN/SrTiO_3/Mn_3GaN$ AFMTJ (Supplementary Figs. S8c-f). While quantitively SOC reduces the ETMR ratio from 1.8×10^4% to 2.6×10^3%, qualitatively the ETMR effect remains huge driven by the nearly perfect spin polarization of Mn_3GaN."

In *Methods*, the following statement has been added: "The effects of spin-orbit interaction on spin polarization of bulk Mn_3GaN (001) and ETMR in $Mn_3GaN/SrTiO_3/Mn_3GaN$ (001) AFMTJs are evaluated using fully relativistic PAW pseudopotentials and discussed in Supplementary Information (Section H), while in the main text, these effects are neglected."

In the revised Supplementary Information, we have added a new section H which discusses the effects of spin-orbit interaction on spin polarization of bulk Mn_3GaN (001) and ETMR in $Mn_3GaN/SrTiO_3/Mn_3GaN/SrTiO_3$ (001) AFMTJs. This section includes the new Figure S8, which is a copy of Figure R9 below.

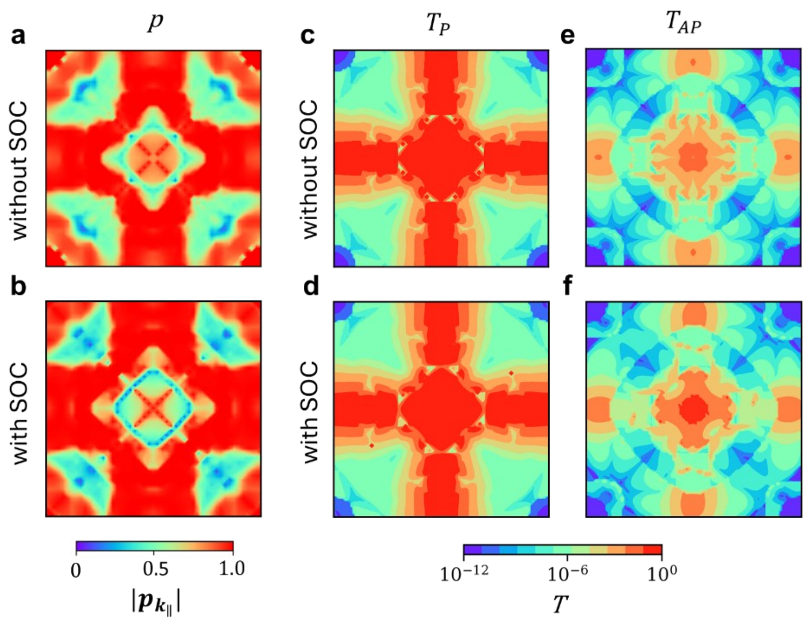


Fig. R9 Effects of SOC on the spin polarization of bulk Mn_3GaN (001) and ETMR in $Mn_3GaN/SrTiO_3/Mn_3GaN$ (001) AFMTJs. (**a**,**b**) $\boldsymbol{k}_{\parallel}$-resolved spin polarization of bulk Mn_3GaN (001) calculated without (**a**) and with (**b**) SOC. (**c-f**) $\boldsymbol{k}_{\parallel}$-resolved transmission in the 2DBZ for the AFMTJ for parallel (P) (**c**,**d**) and antiparallel (AP) (**e**,**f**) alignment of the Néel vector in the Mn_3GaN electrodes calculated without (**c**,**e**) and with (**d**,**f**) SOC.

Comment: [3] The in-plane lattice parameters of the MTJ are mentioned, but how the lattice parameters of the out-of-plane direction, namely, the transmission direction is determined? The explanation for the out-of-plane lattice parameters, particularly, at the interface between Mn3GaN and SrTiO3, should be given.

Response: The structural properties of the $Mn_3GaN/SrTiO_3$ heterostructure have been discussed in detail in Ref. 35 (page 6, second paragraph, in the revised manuscript). Responding to the Reviewer's request, we have included some details from this reference in the *Methods* section.

Changes made in the manuscript: In *Methods* (the first paragraph in *DFT calculations*) we have added the following statement: "Internal coordinates and the c-lattice constant of the supercell were relaxed until the force on each atom was less than 0.001 eV/Å. The resulting bond length between the Mn and O atoms at the Mn_2N/TiO_2 terminated $Mn_3GaN/SrTiO_3$ interface was found to be 2.087Å."

Comment: [4] Is the spin polarization in the Mn3GaN/SrTiO3/Mn3GaN MTJ kept at the interface between Mn3GaN and SrTiO3? The authors show the momentum dependence of the spin-polarization (lowest decay rate) for bulk Mn3GaN (SrTiO3). The results become more convincing if they also discuss the spin polarization when those materials construct the junction.

Response: In response to this Reviewer's comment, we have calculated the layer-resolved spectral density (i.e. layer- and $\boldsymbol{k}_{\parallel}$-resolved density of states), $D^{s_z}(\boldsymbol{k}_{\parallel})$, projected onto the $s_z = \pm½$ spin states for an $Mn_3GaN/SrTiO_3/Mn_3GaN$ AFMTJ. Figure R10a shows the results for the interfacial and bulk-like components of $D^{s_z}(\boldsymbol{k}_{\parallel})$ for the Mn_3GaN electrodes. It is seen that the $s_z = +½$ and $s_z = -½$ spin projections of the spectral density are related by the mirror symmetry $\widehat{M}_{110}$ (or $\widehat{M}_{\bar{1}10}$) transformation similar to the $s^z_{\boldsymbol{k}_{\parallel}}$ spin component shown in Figure 3c of the main text. Importantly, the interfacial and bulk-like $D^{s_z}(\boldsymbol{k}_{\parallel})$ of Mn_3GaN exhibit similar distribution in the 2DBZ, indicating that the spin-dependent features of the band structure of Mn_3GaN do not change much when moving from the bulk to the interface. This behavior is also evident from the spin polarization of the spectral density, $P^z(\boldsymbol{k}_{\parallel}) = \frac{D^{½}(\boldsymbol{k}_{\parallel}) - D^{-½}(\boldsymbol{k}_{\parallel})}{D^{½}(\boldsymbol{k}_{\parallel}) + D^{-½}(\boldsymbol{k}_{\parallel})}$. As seen from Figure R10c, $P^z(\boldsymbol{k}_{\parallel})$ is antisymmetric with respect to diagonals $[110]$ and $[\bar{1}10]$ of the 2DBZ, zero at the diagonals, and have maxima along the $[100]$ and $[010]$ directions. These features are analogous to those exhibited by the *z*-component of the momentum-dependent spin polarization $p^z_{\boldsymbol{k}_{\parallel}}$ in Figure 3e of the main text. Importantly, the interfacial and bulk-like $P^z(\boldsymbol{k}_{\parallel})$ reveal similar patterns, indicating that the non-vanishing spin polarization in the $Mn_3GaN/SrTiO_3/Mn_3GaN$ AFMTJ is preserved at the interface.

Looking at the interfacial and bulk-like components of $D^{s_z}(\boldsymbol{k}_{\parallel})$ in the $SrTiO_3$ barrier layer (Fig. R10b), we observe a pronounced cross pattern reflecting the features of the evanescent states in bulk $SrTiO_3$ reminiscent to that shown in Figure 4b of the main text. Again, we see that the interfacial and bulk-like components of $D^{s_z}(\boldsymbol{k}_{\parallel})$ are qualitatively similar, indicating that the interface does not destroy the properties of the evanescent states followed from the complex band structure of bulk $SrTiO_3$.

Changes made in the manuscript: A new Section E has been added to Supplementary Information that involves the discussion of the layer-resolved spectral density $D^{s_z}(\boldsymbol{k}_{\parallel})$ and its spin polarization $P^z(\boldsymbol{k}_{\parallel})$ and includes Figure R10 as Figure S5.

The following statement has been added to the revised manuscript (page 6, paragraph 3): "We note that, as follows from Supplementary Section E, the interface structure of the AFMTJ maintains bulk-like features of the spectral density (Supplementary Fig. S5a) and its spin polarization (Supplementary Fig. S5b) in Mn_3GaN and the decay rate in $SrTiO_3$ (Supplementary Fig. S5c)."

In *Methods* (the second paragraph in *DFT calculations*), we have explained how the spectral density was calculated: "The layer-resolved spectral density (i.e. the layer- and $\boldsymbol{k}_{\parallel}$-resolved density of states) of the relaxed $Mn_3GaN/SrTiO_3/Mn_3GaN$ (001) structure is calculated using supercell calculations involving periodic boundary conditions with gaussian broadening of 0.01 eV using 50×50 k-points."

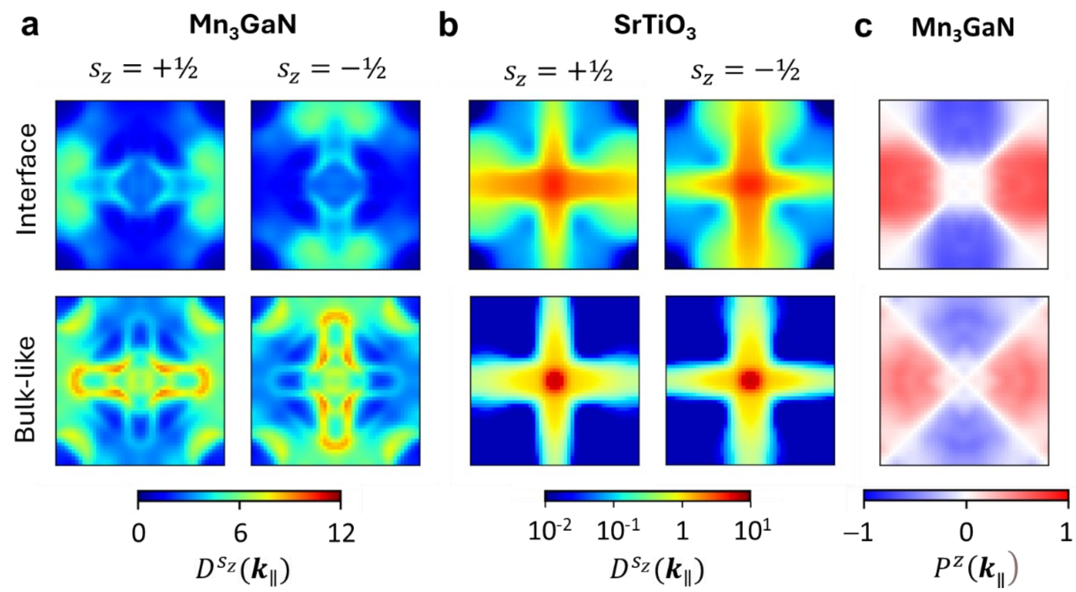


Fig. R10 (a,b) Layer-resolved spectral density $D^{s_z}(\boldsymbol{k}_\parallel)$ projected onto the $s_z = \pm½$ spin states for an $Mn_3GaN/SrTiO_3/Mn_3GaN$ AFMTJ: interfacial (top panels) and bulk-like (bottom panels) components of $D^{s_z}(\boldsymbol{k}_\parallel)$ for Mn_3GaN (**a**) and $SrTiO_3$ (**b**). The interfacial and bulk-like components of the spectral density are calculated by projecting $D^{s_z}(\boldsymbol{k}_\parallel)$ to the interfacial and middle unit cells of the Mn_3GaN or $SrTiO_3$ slabs, respectively. (**c**) Spin polarization of spectral density $P^{z}(\boldsymbol{k}_\parallel)$ for interfacial (top) and bulk-like (bottom) Mn_3GaN.

Comment: [5] The authors discuss the energy dependence of the TMR effect in the Mn3GaN/SrTiO3/Mn3GaN MTJ. They show that the transmission at E = E_F - 0.25 eV gives a larger TMR ratio than that at E_F, where the momentum dependence of the lowest decay rate, "cross pattern", is kept in SrTiO3 as shown in the Supplemental Material. To claim that the TMR effect at different energies from E_F can be understood in a manner similar to that at E_F, they should show the spin splitting properties of Mn3GaN at the energy away from E_F as well as the lowest decay rate of insulators.

Response: In response to this request of the Reviewer, we have calculated the effective spin polarization $\boldsymbol{p}_{k_\parallel}$ for different energies E. The results are shown in Figure R11. It is seen that at $E = E_F - 0.25$ eV, the cross pattern of nearly perfect spin polarization is sustained and even becomes more apparent, while at $E = E_F - 0.5$ eV, it becomes less pronounced. These results are consistent with calculated ETMR value displayed in Figure 4g in the revised manuscript, where it is seen that while at $E = E_F - 0.25$ eV the ETMR ratio is enhanced compared to $E = E_F$, at $E = E_F - 0.5$ eV it is somewhat reduced.

Changes made in the manuscript: We have added a new Section F in Supplementary Information where we included Figure R11 as Figure S6. We have also added a comment on page 7 (the first paragraph) of the revised manuscript as follows: "This enhancement of the ETMR ratio at $E = E_F - 0.25$ eV followed by some reduction at $E = E_F - 0.5$ eV (Fig. 4g) clearly correlates with the appearance of 100% spin polarization in the 2DBZ of Mn_3GaN at different energies (Supplementary Fig. S6)."

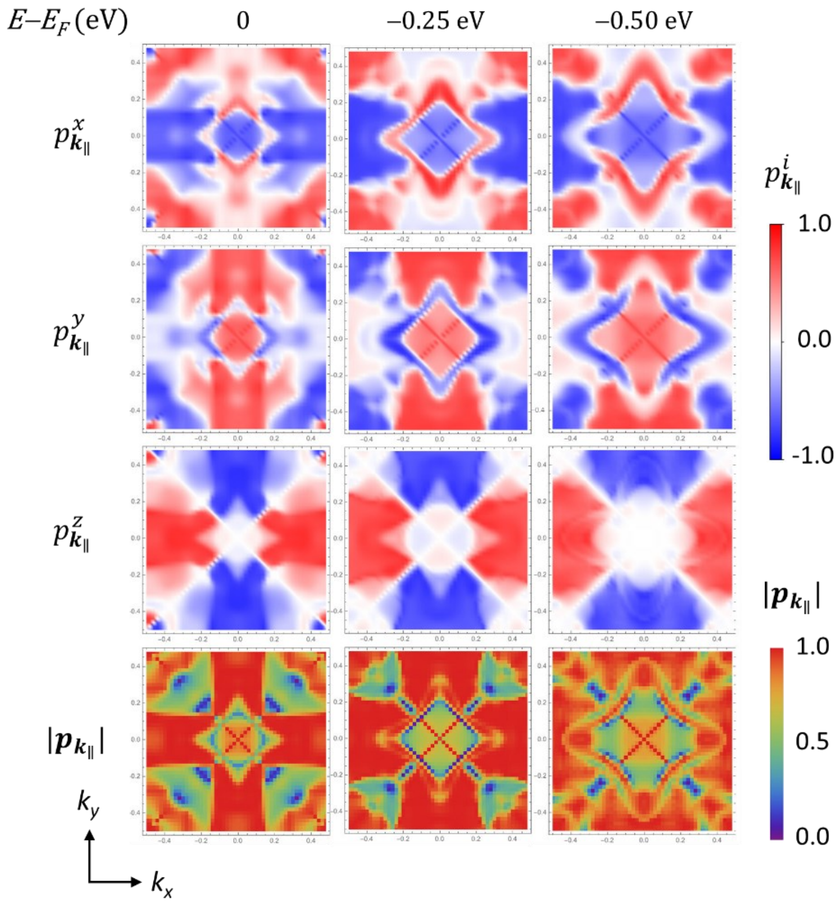


Fig. R11: Components of the effective spin polarization $\boldsymbol{p}_{k_\parallel} = (p^x_{\boldsymbol{k}_\parallel}, p^y_{\boldsymbol{k}_\parallel}, p^z_{\boldsymbol{k}_\parallel})$ (three top panels in each column) and polarization magnitude $p_{\boldsymbol{k}_\parallel} \equiv |\boldsymbol{p}_{\boldsymbol{k}_\parallel}|$ (bottom panels) of bulk Mn_3GaN (001) for different energies E indicated on top.

Comment: [6] This is a minor comment: sometimes the authors use both of 'noncollinear altermagnet' (e.g., "Apart from GaNMn3, other noncollinear altermagnetic ..."), and 'noncollinear antiferromagnet'. I understand that there is a recent proposal to extend the concept of altermagnets to noncollinear magnets as the authors also mention in the introduction, but is there any reason to distinguish these two phrases in this manuscript? If not, it would be better to use either of two expressions (the latter one is better in my opinion) so that readers clearly understand the content.

Response: We agree with the Reviewer.

Changes made in the manuscript: We have made changes in the manuscript to refer to noncollinear antiferromagnets only as "noncollinear antiferromagnets."

Comment: [7] A minor comment: please use either of GaNMn3 or Mn3GaN for the consistency if there are not particular reasons to distinguish these two expressions.

Response: We thank the Reviewer for noticing this inconsistency.

Changes made in the manuscript: We have unified the chemical formula for the antiperovskite to be Mn_3GaN throughout the manuscript.

CHANGES MADE IN THE MANUSCRIPT

Most changes made in the manuscript and Supplementary Information are described in the Response to Reviewers. Other changes include minor corrections, changes in figure numbering and formatting. All changes in the manuscript and Supplementary Information are highlighted.

RESPONSE TO REVIEWERS

REVIEWER #1

Comment: The manuscript presents one study on the spin polarization properties of noncollinear antiferromagnets, with a particular focus on Mn_3GaN. The authors provide a thorough theoretical framework, supported by density functional theory (DFT) calculations, demonstrating the potential for nearly 100% spin polarization and introducing the concept of extraordinary tunneling magnetoresistance (ETMR) in antiferromagnetic tunnel junctions (AFMTJs). While the manuscript has seen substantial improvements, there are several areas that still require further clarification.

Response: We thank the Reviewer for reading our manuscript and for his/her constructive comments. We are pleased that the Reviewer finds "substantial improvements" in our manuscript. Below we respond to the remaining comments of this Reviewer.

Comment: Firstly, the clarity of the figures remains a significant concern. Figures 1c and 1d, in particular, are not sufficiently clear, making it challenging to interpret the data presented.

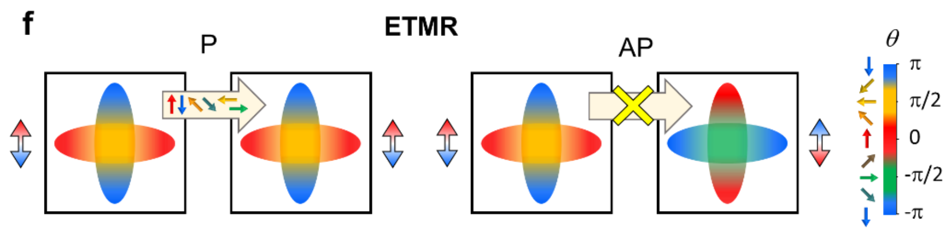
Response: We have positively responded to the Reviewer's same comment in the first round of the review process. We substantially revised Figures 1c and 1d to make them clearer. The data presented in these figures now shows the magnitude and direction of the spin polarization vector, which uniquely define this vector at each momentum and energy point. There is no ambiguity in this presentation. Reviewer #2 who had the same concern about the original Figures 1c and 1d, now is satisfied with our revision. We therefore keep Figures 1c and 1d as they are in the revised version of the manuscript.

Comment: Additionally, Figure 2 illustrates TMR phenomena in both ferromagnetic (FM) and antiferromagnetic (AFM) systems. However, similar TMR effects have already been reported in the literature, including studies published in Physical Review Letters 128.097702, PhysRevX.12.011028, and npj Spintronics2,13(2024). The representation of ETMR in Figure 2 as a novel phenomenon is not entirely convincing, as it appears to resemble a variation of the previously documented d-altermagnetic/d-altermagnetic TMR effect. A more robust argument/representation is needed to clearly distinguish ETMR from these existing effects.

Response: The Reviewer is correct that Figures 2a-d illustrate TMR in ferromagnetic and antiferromagnetic tunnel junctions that have already been reported in the literature. This allows comparing the ETMR to the conventional TMR. In fact, Figure 2 was included in the manuscript in response to this Reviewer, who stated in the first round of review that "A comparative analysis with traditional TMR effects in AFM, AM, FM, and other tunnel junctions would be advantageous." The ETMR effect is illustrated in Figures 2e-f. As seen from Figure 2f, the spin orientation varies across the Brillouin zone, as indicated by arrows and colored ellipses chosen to mimic the mapped z-component of spin polarization shown in Figure 3e. The non-collinear spins in Figure 2f are what make ETMR conceptually different from the conventional TMR shown in Figures 2b and 2d, where the spins are collinear.

This conceptual difference results from the origin of spin conservation. As articulated in the manuscript, for noncollinear antiferromagnetic metals, spin is not a good quantum number and, in general, is not conserved in the tunneling process (even in the absence of spin-orbit coupling). However, we predict that this is not the case for a broad range of wave vectors where our newly defined spin polarization is close to 100%. At these wave vectors, there are no other spin states for electrons to be scattered to, but those that are quantized along the wave-vector dependent spin quantization axis parallel to the spin orientation. This effectively leads to spin conservation as in the collinear case. This unique situation, which has not been discussed in the literature before, is what makes TMR “extraordinary.”

Changes made in the manuscript: To further differentiate the ETMR from the conventional TMR in antiferromagnetic tunnel junctions and to respond to the Reviewer’s comment, we have revised Figure 2f by introducing additional colors into the schematics to reflect noncollinearity of the spins. The revised Figure 2f is reproduced below.



Revised Figure 2f

Comment: There is also a noticeable disconnection between Figures 3 and 4. Figure 3 presents spin polarization bands for the (111) direction of Mn_3GaN, while Figure 4 discusses transport calculations related to the (001) stacking of the material. The manuscript would benefit from a clearer explanation of how the spin polarization results shown in Figure 3 correlate with the transport properties depicted in Figure 4.

Response: The Reviewer might have missed the Figure 3 caption which clearly states that the spin components and spin magnitudes are plotted in the 2D Brillouin zone of Mn_3GaN (001), rather than (111). This fact is also reflected in the main text (the third paragraph on page 5). The real space (111) plane of Mn_3GaN is displayed in Figure 3b just to make it clear for the reader that the magnetic moments of Mn_3GaN lie in the (111) plane where Mn atoms form the Kagome lattice. Therefore, there is no disconnection between Figures 3 and 4.

Comment: Furthermore, on page 5, the authors mention that some of the “spin textures observed are not symmetry-enforced but are accidental”. This raises concerns about the universality and predictability of the ETMR phenomenon in non collinear AFMs, potentially complicating experimental validation.

Response: The text quoted by the Reviewer is, in fact, as follows: “persistent spin texture is not symmetry enforced like that predicted in Ref. 32, but rather “accidental.” This statement was included in the manuscript in response to the comment of Reviewer #2 who was interested in a possibility of persistent spin texture in Mn_3GaN. While the persistent spin texture indeed appears in Mn_3GaNi, its presence is not required for the ETMR effect. The concern of the Reviewer is not therefore relevant.

Changes made in the manuscript: To avoid the confusion, we have removed word "accidental" in the revised manuscript.

Comment: Lastly, while the assertion of nearly 100% spin polarization is striking, it may be subject to overestimation under realistic conditions. The authors should consider discussing potential mechanisms (scattering or temperature) that could influence effective spin polarization in practical applications.

Response: The Reviewer is correct that in experimental conditions, the predicted ETMR effect is expected to be reduced. This reduction can be driven by magnetic moment misalignment due to their thermal fluctuations at finite temperature and/or the presence of defects in the barrier resulting in diffuse scattering of tunneling electrons. These have been articulated in the original version of the manuscript. Two additional requirements for observing the ETMR effect are identical antiferromagnetic electrodes and low bias voltage. All these requirements are now articulated in the revised manuscript.

Changes made in the manuscript: The fifth paragraph on page 7 of the revised manuscript has been modified and now reads as follows: "The are a few important requirements for observing the predicted ETMR effect. AFMTJs must have identical AFM electrodes to provide matching between their spin states in the momentum space. High quality and crystallinity of AFMTJs are required for conservation of transverse momentum $\boldsymbol{k}_{\parallel}$ in the process of tunneling. Defects in the barrier, such as oxygen vacancies, should be avoided because they can lead to diffuse scattering between different $\boldsymbol{k}_{\parallel}$ thus suppressing ETMR (similar to TMR in conventional MTJs[39]). Also, measurements well below the Néel temperature are desirable, due to temperature-driven fluctuations of magnetic moments being detrimental for ETMR. Finally, measurements at a low bias voltage are required, since ETMR is expected to decrease with bias due to the mismatch of spin states at different energies."

REVIEWER #2

Comment: Now I would like to recommend it for publication

Response: We thank the Reviewer for the reading of our revised manuscript. We are pleased by the Reviewer's recommendation to publish it without changes.

REVIEWER #3

Comment: The authors clearly answered all of my comments. I recommend this revised manuscript for publication.

Response: We thank the Reviewer for the reading of our revised manuscript. We are pleased by the Reviewer's recommendation to publish it without changes.

CHANGES MADE IN THE MANUSCRIPT

All changes made in the manuscript are articulated in our response to Reviewer #1 and highlighted.