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Peer Review File

Large Spin Polarization from Symmetry-Breaking Antiferromagnets in Antiferromagnetic Tunnel Junctions

Reviewers' Comments:

Reviewer #1:
Remarks to the Author:
This manuscript claims to demonstrate that symmetry-breaking antiferromagnets Mn3Sn provides large spin polarization and in doing so achieve a giant TMR up to 165%. It should be noted that the previously reported highest MR ratio of Mn3Sn MTJ was ~2%, two orders smaller as compared with this work. Therefore, this claim is extremely exciting because it offers the possibility of both efficiently generating spin current via magnets with negligible net magnetization and direct electrical reading of chiral antiferromagnetic structures. This has obvious technological potential in magnetic random-access memory, but also as a tool to study the fundamentals of Weyl magnets. The manuscript is well written, and the conclusions are well supported. I therefore recommend publication in Nature Communications with minor additions/corrections:
1. Line 69 would be better to say “The small TMR could be mixed with the contribution from residual magnetic moment of AFM” as the contribution from residual magnetic moment (~0.1%) and chiral antiferromagnetic structure (~0.3%) can be extracted from the bias-dependent measurements.
2. I would also like to see a slightly extended commentary on the formation of pinned interfacial antiferromagnetic structure and whether it is related to the etching process.
With these minor issues addressed I am very happy to recommend publication.

Reviewer #2:
Remarks to the Author:
The authors reported high TMR achieved in asymmetric AFM-based magnetic tunnel junctions for the first time. They also carried out thorough experiments to characterize structural and magnetic properties of Mn3Sn films and magnetic tunnel junction device performance. The authors are suggested to address the following items to improve the clarity and completeness of their results.
1. The authors clearly stated that the 100% TMR was achieved at 10K and TMR vanishes at 200K in the paper. The fact that the TMR was achieved at 10K only showed up starting from line 126. This should have been stated up in front, in the abstract, when high TMR is mentioned for the first time.
2. Please include a TMR vs Temperature plot in the main text
3. There needs to be more discussions about the blocking temperature of Mn3Sn and what is expected from the theoretical calculations in terms of TMR temperature dep. of Mn3Sn based junctions.
4. The counter electrode chosen for the study is CoFeB, which is commonly used in the practical MTJ devices. When CoFeB and MgO barrier are combined, high TMR can be

achieved since CoFeB crystallizes during the annealing process and develops a grain expitaxy relationship with the MgO tunnel barrier. In this study, there is no annealing process after CoFeB growth. It will be helpful to include a sentence to explain why CoFeB was chosen. If the authors can try other counter electrode, like CoFe, will higher TMR be possible?
5. Both the TMR value and the junction resistance seem to be quite unstable, based on the data provided in Fig. 3 and Fig. 4 from the same device. This needs to be stated and explained in the text.
6. From line 132-136, the authors discussed bias voltage dependence of TMR and included a figure in the supplementary document. Although there is bias voltage dependence of TMR in regular MTJs, the Rmin state is usually flat with weak bias voltage dependence. Here, the voltage dependence of TMR is much stronger. It is worth to comment on it for clarity.
7. Fig. 4 (c) and (d) and the discussions about those two figures are confusing. In Fig. 4 (d), the two arrows indicating magnetic field (H) directions are not defined – it could be the directions of the cooling fields during the field cooling experiments, or the fields that were applied to switch the Mn3Sn after the cooling experiments. This needs to be clarified with one set of figures showing the magnetic configurations right after field cooling experiments and another set to show the magnetic configurations under the influence of the applied field during the MH measurements.

Reviewer #3:
Remarks to the Author:
The original work entitled "Large Spin Polarization from Symmetry-Breaking Antiferromagnets in Antiferromagnetic Tunnel Junctions" from C. Chou et al studied the spin current generation of Mn3Sn in asymmetric MTJ structures obtaining a high TMR ratio (>100%) with a clear dependence on a CoFeB thin film's magnetization orientation.

Antiferromagnetic spintronics has been a hot topic since the discovery of resistance switching in broken inversion symmetry antiferromagnets, CuMnAs. After that, magnetotransport properties have been reported in different antiferromagnetic materials, as the one studied by C. Chou et al, Mn3Sn. This antiferromagnet presents a canted small magnetic moment due to the spin arrangement in their crystal structure. Furthermore, the broken time reversal symmetry, while parity symmetry is present, promotes the appearance of Weyl points in the band structure which electrons show a well-defined helicity (spin-momentum locking). This effect is the origin of a large Anomalous Hall effect, MOKE signal and, recently shown, Tunneling MagnetoResistance, TMR.

The authors results are of importance for the spintronic community, providing interesting and useful results for designing and fabricating new devices based on these materials. I have the following comments to the authors.

• I agree with the authors that for getting a finite TMR in AFM symmetric MTJs both electrodes must present similar crystal structures, to accomplish the same spin-momentum dependence and then distinguish between parallel and antiparallel states. This is reflected in section 2 when the authors claim: “Since the TMR from the asymmetric an AFMTJ is correlated with the net spin polarization in tunneling current, it is expected to be more robust compared with the corresponding effect in symmetric AFMTJs, where in the latter both AFM electrodes have to be crystalized for reaching matched Fermi Surfaces”. However, the origin of the TMR in the asymmetric tunnel junction, explained one sentence before, says: “The momentum dependent spin polarization p(k) in AFM, in combination with the different available tunneling channels into the FM counter electrode at different k locations, leads to finite TMR (Fig.1c)”. I understand the principle that the authors explain in Figure 1c where the rotation of the magnetization in the CFB layer can match (mismatch) with some channels in the momentum space of the antiferromagnet. I think that this point needs to be extended to make their work more accessible to the audiences that are not familiar with these materials, and the main theory behind this phenomenon that explains the main results of the authors, for example including a section in the supplementary material.
• It is clear from the field cooling experiments that there are some surface effects at the interface between the dielectric layer and Mn3Sn, like pinned domains as the authors suggest. Have the authors used other material characterization techniques in addition to TEM and XRD, e.g., XPS to try to identify other MnSn and Mn phases?
• From the hysteresis loops shown in section 3 of the supplementary section, although the magnetization strongly decreases from the 10 K loop, the Mn3Sn still is in the non-collinear phase. Nevertheless, the exchange bias at room temperature and the TMR are loosed at room temperature. For me it is not clear that the blocking temperature of the AFM/FM bilayer are the responsible of the measured TMR or, at least, correlated to the interfacial pinned moments. Are the pinned domains the responsible of the exchange bias?
• If the pinned domains are invariant under field cooling under 70 kOe, the TMR should not be changed as it is illustrated in Figure 4 d, because in Figure 4a the proposed mechanism depends on the direction of the bulk spins. I guess that the authors wanted to represent the magnetic state of the pinned moments of MnSn in figure a.
• Finally, the spin polarization generated by Mn3Sn is just effective at low temperature? Or using thinner layers, where the surface effects could be larger than the bulk, will be operative at larger temperatures? This is important to use the AFM spin polarizers in room temperature applications.

The manuscript is well organized, well written, and clearly explains the main results and the methods than the authors have used. I really think that their work deserves publication at Nature communications after a major revision.

Reviewer #1:

This manuscript claims to demonstrate that symmetry-breaking antiferromagnets Mn3Sn provides large spin polarization and in doing so achieve a giant TMR up to 165%. It should be noted that the previously reported highest MR ratio of Mn3Sn MTJ was ~2%, two orders smaller as compared with this work. Therefore, this claim is extremely exciting because it offers the possibility of both efficiently generating spin current via magnets with negligible net magnetization and direct electrical reading of chiral antiferromagnetic structures. This has obvious technological potential in magnetic random-access memory, but also as a tool to study the fundamentals of Weyl magnets. The manuscript is well written, and the conclusions are well supported. I therefore recommend publication in Nature Communications with minor additions/corrections.

We are grateful to the reviewer for the commendation on our work and the recommendation for publication with revisions. We find that the suggestions from the reviewer highly valuable and have made the changes correspondingly.

1. Line 69 would be better to say "The small TMR could be mixed with the contribution from residual magnetic moment of AFM" as the contribution from residual magnetic moment (~0.1%) and chiral antiferromagnetic structure (~0.3%) can be extracted from the bias-dependent measurements.

Response:

We thank the reviewer for pointing out that both residual moment contribution and chiral AFM structure contribution are present in the previous device. We have revised the manuscript following the reviewer's suggestion (top of page 3):

"The small TMR could be mixed with the contribution from residual magnetic moment of AFM."

2. I would also like to see a slightly extended commentary on the formation of pinned interfacial antiferromagnetic structure and whether it is related to the etching process.

Response:

This is a great question! Firstly, we agree with the reviewer that usually the etching process during MTJ fabrication could modify the interfacial layer property of magnetic electrode, as has been generally observed in standard MTJs. Meanwhile, we found that the pinned domains exist even in as-prepared films that do not experience ion bombardment, as suggested from our *MH* hysteresis loop measurement and from the anomalous Hall effect measurement in [Physical

Review Applied 14, 054037 (2020)], where the 'frozen' portion of magnetization or anomalous Hall effect causes a shift in the hysteresis loop that is not changeable with field. The exact cause of the modified interfacial layer property is not fully understood and requires further study. In the revised manuscript, besides the original discussions on the origins of the interfacial pinned layer, we added the following on the etching induced damage (top of page 9):

"In addition, it is well known that Ar plasma etching process (See Experimental Section for detail) could change the properties of the magnetic electrodes [51], and its influence on the Mn_3Sn interface layer requires further investigation."

Reviewer #2:

The authors reported high TMR achieved in asymmetric AFM-based magnetic tunnel junctions for the first time. They also carried out thorough experiments to characterize structural and magnetic properties of Mn3Sn films and magnetic tunnel junction device performance. The authors are suggested to address the following items to improve the clarity and completeness of their results.

We thank the reviewer for the careful review of our manuscript and providing the valuable suggestions. We have modified our manuscript following the reviewer's suggestion:

1. The authors clearly stated that the 100% TMR was achieved at 10K and TMR vanishes at 200K in the paper. The fact that the TMR was achieved at 10K only showed up starting from line 126. This should have been stated up in front, in the abstract, when high TMR is mentioned for the first time.

Response:

We thank the reviewer for this great comment! To clarify on the temperature limit, in the revised manuscript we have added the temperature information on TMR in abstract and introduction paragraph:

Abstract:

"We observe a high TMR at cryogenic temperature (>100% at 10 K) in these asymmetric AFM tunnel junctions…"

Introduction (bottom of page 3):

"…TMR in AFM/insulator/FM tunnel junctions as large as conventional ferromagnetic MTJs is observed at cryogenic temperatures (>100% at 10 K and vanishes at 200 K and above)."

2. Please include a TMR vs Temperature plot in the main text

Response:

Thanks for this suggestion! In this revision, we move the TMR curves measured at different temperatures to main text as Fig. 3c. The following is the caption of Figure 3:

"…(c) Magnetoresistance of the same AFMTJ measured at different temperatures. The TMR decreases with temperature and vanishes at 200 K and above. The data are shifted vertically for clarity."

In addition, TMR ratio vs. temperature plot is added as supplementary information (section 10). Caption for Supplementary Figure 9:

"Temperature dependence of TMR ratio. The TMR decreases with temperature and vanishes at temperatures above ~200 K. The error bar is defined as the standard deviation of the measured data."

3. There needs to be more discussions about the blocking temperature of Mn3Sn and what is expected from the theoretical calculations in terms of TMR temperature dep. of Mn3Sn based junctions.

Response:

We thank the reviewer's question on the blocking temperature of Mn_3Sn and the suggestion on discussing theoretical temperature dependence of TMR.

First, about the blocking temperature of Mn_3Sn, experimentally in both our experiment (see Supplementary Section 11) and literatures [see for example, Physical Review Materials 2, 051001(R) (2018)], the exchange bias field of Mn_3Sn/FM bilayers reduces to negligible values between 200 K and RT, which is consistent with the temperature (~200 K) under which our TMR magnitude goes to zero and that the field-cooling needs to overcome in order to reset the TMR polarity. In addition, the pinned domain in Mn_3Sn single layer reported in [Physical Review Applied 14, 054037 (2020), now Ref. 36] also shows a similar unpinning temperature below RT.

On the theoretical expectation on the TMR's temperature dependence, at this moment first principles calculation has limited power in predicting the finite temperature performance of MTJ. But using the general power law on magnetism and spin ordering, $m_{AF}(T) = m_{AF}^{(0)}(1 - T/T_N)^{\lambda}$, where m_{AF} represents the ordering parameter of the antiferromagnet, e.g., octupole moment, and $\lambda \sim 0.2 - 0.5$ is the critical index (see, e.g., Physics of Ferromagnetism, 2nd ed. Clarendon, Oxford, 1997, p. 327, and Phys. Rev. B 49, 3962 (1994)), one expects that the TMR decreases with the increase of temperature and vanishes at the Néel temperature (T_N~ 420 K for Mn_3Sn).

In our experiment we find that the TMR reduces to zero at around 200K, before the temperature reaches T_N. We found that this 200K is also the temperature that the field-cooling process needs to overcome in order to reset the TMR polarity and the offset of magnetization (Fig. S8 in the supplementary), suggesting the existence of such a lower-than-T_N unpinning temperature, very similar to the blocking temperature in AFM/FM bilayer structure as discussed above. We note that similar phenomena, including the existence of frozen domains and an unpinning temperature below room temperature, have also been reported in Ref. 36 in the study of anomalous Hall effect of single layer of Mn_3Sn (see Fig. 4d and 4f there). Therefore, the loss of TMR at ~200K is not related to the disappearance of noncollinear antiferromagnetism, but rather the thermally induced fluctuation of AFM domains.

With respect to the modelling on the blocking temperature T_B, in AFM/FM bilayer it can be calculated using the semi-empirical relation of $1 - T_B/T_N = (\xi_0/t_{AF})^{0.5\lambda}$, where t_{AF} is the AFM thickness and ξ_0 is the (pinned) AFM coherence length, dependent on the interfacial coupling strength and parameters like domain sizes (see, e.g., J. Appl. Phys. 92, (2002) 4828) [40]. Currently the relevant parameters such as ξ_0 are not still available, particularly in our Mn_3Sn single layer electrode, therefore it is hard to get an accurate prediction on the temperature dependence of pinned domain behavior, including the TMR. Understanding the microscopic mechanisms of the pinned layer will be one of our future directions.

In the revised manuscript, a description of the Mn_3Sn blocking temperature and its relation to the temperature dependence of TMR is added in main text, page 7:

"Accompanied with the thermally randomized polarity, we see that the magnitude of TMR also decreases with temperature and vanishes above 200 K (Fig. 3c and Supplementary Information section 10), a temperature under which Mn_3Sn is still in the non-collinear antiferromagnetic phase (Néel temperature ~ 420 K). This suggests that the magnetic states of Mn_3Sn contributing to TMR are pinned at low temperature but activated into fluctuated states at RT. The unpinning of Mn_3Sn at a temperature significantly lower than the Néel temperature resembles the blocking temperature of exchange bias in AFM/FM structures [40], where the AFM domains becomes super-paramagnetic due to thermal agitation [41]."

In the mid of page 8:

"Both magnetic moment offset and TMR vanish at temperatures above 200 K. Previously, separations between pinned and rotatable domains have also been observed in the anomalous Hall effect measurement of single layer Mn_3Sn, which exhibits a similar unpinning temperature below RT [36]."

4. The counter electrode chosen for the study is CoFeB, which is commonly used in the practical MTJ devices. When CoFeB and MgO barrier are combined, high TMR can be achieved since CoFeB crystallizes during the annealing process and develops a grain expitaxy relationship with the MgO tunnel barrier. In this study, there is no annealing process after CoFeB growth. It will be helpful to include a sentence to explain why CoFeB was chosen. If the authors can try other counter electrode, like CoFe, will higher TMR be possible?

Response:

This is a great question! The reason that we chose CoFeB as the ferromagnetic counter electrode is two-fold: firstly, as the reviewer pointed out, CoFeB is a very classical ferromagnetic material for MgO based MTJ growth. In our study, although we did not benefit from the MgO templating effect during post deposition annealing, the smooth interface and well-proven chemical stability across MgO/CoFeB is still beneficial for us to get these functional tunnel junction devices. Secondly, as we explain in the introduction paragraph (page 3), the TMR in asymmetric AFMTJ in this work is originated from the spin polarized Fermi surface of Mn_3Sn in reciprocal space, where Mn_3Sn effectively acts like a ferromagnet and generates a net spin polarization when the forward-moving electrons dominate in the tunneling. Therefore, the TMR is expected to be robust against the imperfect crystallization of the barrier or FM counter-electrode, and exists even with amorphous counter-electrode. This is distinct from the previously proposed symmetric AFMTJ scheme where both AFM electrodes must be crystalized and matched with each other [see, ref. 23, *Physical Review Letters* **128**, 197201 (2022)]. Therefore, the usage of CoFeB, which is the most widely employed amorphous ferromagnetic material in their as-grown state, also serves the purpose of illustrating the robustness of the current mechanism in generating spin polarization from AFM.

We agree that it is possible to get as high TMR using alternative counter-electrode materials, such as CoFe, which form polycrystalline structure in the absence of annealing. One potential downside is that the polycrystalline counter-electrode could lead to a rougher interface at MgO/FM due to the lattice/crystalline-orientation mismatch. Additional material screening and optimization will be necessary to optimize such interfaces.

In the revised manuscript, we have added the following to explain the usage of CoFeB as the FM counter-electrode.

Close to the bottom of page 3:

"Since the spin transport from such AFM electrode has a net spin polarization, the TMR is expected to be significant even with amorphous FM counter-electrodes, which is different from previous symmetric AFMTJs which require crystallized and matched electrodes on both sides [23]"

An explanation on why CoFeB was chosen is added to the main text, page 4:

"CoFeB, the widely used ferromagnetic material in regular MTJs, is chosen here because it tends to form amorphous structures when sputtered without post-deposition annealing [32], which results in a smooth interface with MgO. It also yields a simpler physical picture for characterizing the net spin polarization from the Mn_3Sn electrode without the need to consider the exact electronic structure of the FM."

5. Both the TMR value and the junction resistance seem to be quite unstable, based on the data provided in Fig. 3 and Fig. 4 from the same device. This needs to be stated and explained in the text.

Response:

As the reviewer commented, the TMR value of the same device shows variations after each thermal cycle in Fig. 3 and Fig. 4. For the zero-field cooled case (Fig. 3), both the sign and the value of TMR are expected to have a large variation as a result of the randomly aligned pinned domains during the cooling process. For field-cooled case (Fig. 4), we find that despite the polarity of the TMR becomes deterministic among different thermal cycles, the magnitude of TMR can still show variations. This indicates that the cooling field starting from room temperature is still not able to saturate 100% of the interfacial domains, and some of the domains may require even higher in-field annealing temperature.

A description of the unstable TMR magnitude and an explanation is added in main text, page 7:

"We note that despite the polarity of the TMR can be controlled by the cooling field, the magnitude can vary between thermal cycles. This suggests that the cooling field does not saturate the domains and only partially controls the orientation of the pinned domains."

6. From line 132-136, the authors discussed bias voltage dependence of TMR and included a figure in the supplementary document. Although there is bias voltage dependence of TMR in regular MTJs, the Rmin state is usually flat with weak bias voltage dependence. Here, the voltage dependence of TMR is much stronger. It is worth to comment on it for clarity.

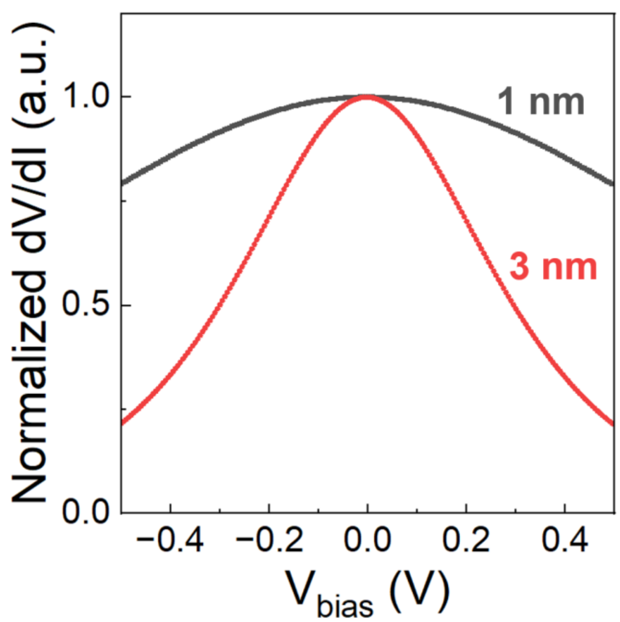
Response:

We thank the reviewer for this nice observation! The reviewer is correct that our MTJs show a stronger bias voltage dependence in their parallel state (R_min) than regular MTJs. Our understanding is that this is related to the larger tunnel barrier thickness and the corresponding higher RA product. In our experiment, we intentionally grow MgO relatively thick (3~4 nm) to avoid pinholes so that we can focus on the pursued physics. Nowadays, in most experiments people study low RA MTJ with thin tunneling barrier (~ 1nm). According to the classical Simmon's theory on tunneling current [J. Appl. Phys. (1963) 34, 1793], the voltage dependence of tunneling resistance sensitively depends on barrier thickness. This is because the current of tunnel junction is an exponential function of the applied voltage and the barrier thickness [see, e.g, Eq. (1) of Materials Today 9, 36–45 (2006)], where the nonlinearity gets stronger with a thicker tunneling barrier. In the figure below, we compared the dV/dI curve for two MTJs with tunneling barrier thickness of 1nm and 3nm, separately, according to Eq. (1) of the Materials Today paper. We can see that the thicker junction has a much stronger voltage dependence. This result is also consistent with the original paper of Simmon (see Fig. 7 of J. Appl. Phys. (1963) 34, 1793, note that the y axis there is in log scale).

In regular low RA MTJs, the intrinsic nonlinear IV characteristic is not very obvious in the dV/dI vs bias voltage measurement. Therefore, R_p is observed to be largely flat and the magnon induced TMR change is dominating, which reflects as a faster variation of R_ap than R_p. In our high RA MTJ, however, the intrinsic IV nonlinearity becomes very significant, which therefore leads to a large R_p change under biasing voltage, on top of the TMR variation.

A description and explanation on the strong bias dependence of R_min is added to the figure caption of Fig. S6 in supplementary:

"Besides TMR, the low resistance state also exhibits a very strong biasing voltage dependence. This can be explained by the large tunneling barrier thickness of our AFMTJ, which leads to strong nonlinearity in the IV curve [S4]."



Response Figure 1. dV/dI calculated from Eq. (1) of the Materials Today paper with tunnel barrier thickness of 1 nm (grey) and 3 nm (red). The resistance is normalized to the zero-bias resistance to illustrate the difference in the bias dependence of the two junctions.

7. Fig. 4 (c) and (d) and the discussions about those two figures are confusing. In Fig. 4 (d), the two arrows indicating magnetic field (H) directions are not defined – it could be the directions of the cooling fields during the field cooling experiments, or the fields that were applied to switch the Mn3Sn after the cooling experiments. This needs to be clarified with one set of figures showing the magnetic configurations right after field cooling experiments and another set to show the magnetic configurations under the influence of the applied field during the MH measurements.

Response:

We thank the reviewer for this important point. Following the reviewer's suggestion, in the revised manuscript we replot Fig. 4d showing all four scenarios: the combination of positive/negative cooling field and positive/negative measurement field. The cooling and measurement fields are also represented with different shapes of arrows.

The caption of Fig 4d is also updated:

"(d) Schematic of the pinned and switchable domains of Mn_3Sn after field cooling. The blue and red arrows of ±FC indicate the cooling fields, and the black arrows of ±H represent the applied field after field cooling. The domains close to interface are pinned as indicated by the TMR results."

Reviewer #3:

The original work entitled "Large Spin Polarization from Symmetry-Breaking Antiferromagnets in Antiferromagnetic Tunnel Junctions" from C. Chou et al studied the spin current generation of Mn3Sn in asymmetric MTJ structures obtaining a high TMR ratio (>100%) with a clear dependence on a CoFeB thin film's magnetization orientation.

Antiferromagnetic spintronics has been a hot topic since the discovery of resistance switching in broken inversion symmetry antiferromagnets, CuMnAs. After that, magnetotransport properties have been reported in different antiferromagnetic materials, as the one studied by C. Chou et al, Mn3Sn. This antiferromagnet presents a canted small magnetic moment due to the spin arrangement in their crystal structure. Furthermore, the broken time reversal symmetry, while parity symmetry is present, promotes the appearance of Weyl points in the band structure which electrons show a well-defined helicity (spin-momentum locking). This effect is the origin of a large Anomalous Hall effect, MOKE signal and, recently shown, Tunneling MagnetoResistance, TMR.
The authors results are of importance for the spintronic community, providing interesting and useful results for designing and fabricating new devices based on these materials.

The manuscript is well organized, well written, and clearly explains the main results and the methods than the authors have used. I really think that their work deserves publication at Nature communications after a major revision.

We thank the reviewer for the confirmation on the importance of our results and the recommendation on publication with revisions. We found that the reviewer's suggestion highly valuable for us to improve the quality of our work. We have revised the manuscript following the reviewer's suggestions:

1. I agree with the authors that for getting a finite TMR in AFM symmetric MTJs both electrodes must present similar crystal structures, to accomplish the same spin-momentum dependence and then distinguish between parallel and antiparallel states. This is reflected in section 2 when the authors claim: "Since the TMR from the asymmetric an AFMTJ is correlated with the net spin polarization in tunneling current, it is expected to be more robust compared with the corresponding effect in symmetric AFMTJs, where in the latter both AFM electrodes have to be crystalized for reaching matched Fermi Surfaces". However, the origin of the TMR in the asymmetric tunnel junction, explained one sentence before, says: "The momentum dependent spin polarization p(k) in AFM, in combination with the different available tunneling channels into the FM counter electrode at different k locations, leads to finite TMR (Fig.1c)". I understand the principle that the authors explain in Figure 1c where the rotation of the magnetization in the CFB layer can match (mismatch) with some channels in the momentum space of the antiferromagnet. I think that this point needs to be extended to make their work more accessible to the audiences that are not familiar with these materials, and the main theory *behind this phenomenon that explains the main results of the authors, for example including a section in the supplementary material.*

Response:

We thank the review for this great suggestion! We totally agree with the reviewer on the mentioned physical picture and the necessity of providing more detailed discussions to make this part accessible to general readers. In the revised manuscript, we have added the following sentences in the main text to further clarify on the difference in the working mechanisms of the two types of tunnel junctions, and included a new section in the Supplementary Information (Section 7) for an extended explanation.

Page 6 of main text:

"The momentum dependent spin polarization $\boldsymbol{p}(\boldsymbol{k})$ in AFM, in combination with the different tunneling probabilities at the center and edge of the projected 2D Brillouin zone, leads to finite TMR (Fig. 1c). The tunneling current in this case is spin polarized and the AFM behaves similar to a FM in regular MTJs. Since the TMR from the asymmetric AFMTJ is correlated with the net spin polarization in tunneling current, it is expected to be less stringent in the crystallization condition of the FM counter-electrode compared with the corresponding effect in symmetric AFMTJs (see more discussion in the Supplementary Information section 8)."

Bottom of page 3:

"Since the spin transport from such AFM electrode has a net spin polarization, the TMR is expected to be significant even with amorphous FM counter-electrodes, which is different from previous symmetric AFMTJs which require crystallized and matched electrodes on both sides [23]."

2. It is clear from the field cooling experiments that there are some surface effects at the interface between the dielectric layer and Mn3Sn, like pinned domains as the authors suggest. Have the authors used other material characterization techniques in addition to TEM and XRD, e.g., XPS to try to identify other MnSn and Mn phases?

Response:

We appreciate the reviewer's suggestion on using additional characterization technique to get a better understanding on the interfacial layer property of the Mn_3Sn electrode. Firstly, we agree that XPS is a great tool in revealing the chemical state of the elements, which could give a lot of useful information on this material. At the same time, it has limited probing depth (a few nanometers). To get information on the buried interface, special treatment and instrumentation such as in-situ etching will be necessary, where the extra etching step might lead to new damage and modify the film's property. A systematic characterization with XPS and other probing tools to further reveal the Mn_3Sn surface property is within our future plan. Although we don't have XPS results currently, we did carry out energy dispersive x-ray (EDX) study using STEM and mapped the elemental distribution near the junction region, where we find no significant Mn or Sn diffusion (see the new Section 10 in the Supplementary Information). Finally, we note that regardless of the specific phase of the interfacial layer, the large coercive field (> 7 Tesla) of the pinned domain is a characteristic to AFM phase, and our main conclusion on the AFM based spin polarization should hold.

In the revised main text, EDX measurements is mentioned in the top of page 5:

"Energy dispersive X-ray spectroscopy is carried out to map the elemental distribution near the junction, and no significant Mn or Sn diffusion is observed (Supplementary Information Section 2)."

Also, we added one sentence close to the bottom of page 8:

"…..which alters the interfacial layer properties including a significant increase of coercivity [48]. Future experiments with characterization tools such as x-ray photoelectron spectroscopy will help to elucidate the exact state of the interfacial layers."

3. From the hysteresis loops shown in section 3 of the supplementary section, although the magnetization strongly decreases from the 10 K loop, the Mn3Sn still is in the non-collinear phase. Nevertheless, the exchange bias at room temperature and the TMR are loosed at room temperature. For me it is not clear that the blocking temperature of the AFM/FM bilayer are the responsible of the measured TMR or, at least, correlated to the interfacial pinned moments. Are the pinned domains the responsible of the exchange bias?

Response:

We thank the review's question on the correlation between the exchange bias in the AFM/FM bilayer and the pinned domains in our TMR measurements.

Firstly, as the reviewer summarized, we did observe a disappearance of TMR around 200K, lower than the Néel temperature ($T_N \sim 420$ K) of Mn_3Sn. We found that this 200K is also the temperature which the in-field cooling process needs to overcome to reset the TMR polarity and the offset of magnetization (Fig. S8 in the supplementary). Similar to our report, an earlier literature, ref. 36 (Physical Review Applied 14, 054037 (2020)) also showed that there is an exchange bias behavior between the pinned and rotatable domains of a single Mn_3Sn layer in the anomalous Hall effect measurement. In ref. 36, the pinned domains also get unpinned at a temperature between 200 K and RT, below Néel temperature, consistent with our TMR measurement.

The existence of such a lower-than-T_N unpinning temperature was the original reason that we chose the term 'blocking temperature', which is usually used to describe the exchange bias phenomena in AFM/FM bilayer. However, here we didn't mean that there is necessarily a one-to-one correspondence in the microscopic mechanism between the exchange bias in AFM/FM and the TMR in our MTJ structure, aside from the coincidence that both the two share low unpinning temperatures. Particularly, it obviously requires more study and evidences to know whether the interfacial pinned layer of the Mn_3Sn electrode as observed in our TMR measurement is directly responsible for the exchange bias effect in AFM/FM bilayer.

Realizing that the usage of the terminology 'blocking temperature' could have caused some unnecessary confusions, in this revision we have changed from 'blocking temperature' to 'unpinning temperature' and added the following sentence on page 7:

"The unpinning of Mn_3Sn at a temperature significantly lower than the Néel temperature resembles the blocking temperature of exchange bias in AFM/FM structures [40], where the AFM domains becomes super-paramagnetic due to thermal agitation [41]."

In supplementary information section 12, we did measure the exchange bias from Mn_3Sn/CoFeB bilayer, whose original purpose was to verify that the Mn_3Sn is in its AFM state at low temperature. This is because people usually regard the exchange bias effect in AFM/FM heterostructure as a strong evidence on the AFM phase. To avoid confusion, we modified the discussion in this section:

"Interestingly, we find that the exchange bias in our Mn_3Sn/CoFeB film vanishes after warming up to RT, suggesting a lower-than-RT blocking temperature in the AFM/FM bilayer, similar to the unpinning of the interfacial layer of Mn_3Sn electrode in the MTJ. Additional studies are necessary to show if there is an internal correlation between these two systems.

4. If the pinned domains are invariant under field cooling under 70 kOe, the TMR should not be changed as it is illustrated in Figure 4 d, because in Figure 4a the proposed mechanism depends on the direction of the bulk spins. I guess that the authors wanted to represent the magnetic state of the pinned moments of MnSn in figure a.

Response:

Thanks for pointing this out! Yes, the reviewer is completely right that we meant to illustrate the pinned magnetic state in the interfacial layer in Fig. 4a. We believe that the color we chose to represent the Mn_3Sn layer in Fig. 4a and 4b might be the main cause of this confusion since it was inconsistent with the schematic of Fig. 4d. In this revision, we've changed the color coding to ensure that the interfacial domain in Fig. 4d match with that in Fig. 4a and 4b. A description is also added in the caption:

"Insets: illustration of the relative orientations between FM magnetic moment and AFM octupole moment near the tunneling interface under different fields."

5. Finally, the spin polarization generated by Mn3Sn is just effective at low temperature? Or using thinner layers, where the surface effects could be larger than the bulk, will be operative at larger temperatures? This is important to use the AFM spin polarizers in room temperature applications.

Response:

Our current devices only shows large TMR at low temperatures (>100% at 10K, vanished at 200K and above). We totally agree that an efficient AFM spin polarizer at higher temperature is critical for room temperature applications. For future work, besides improving the surface effect pinning effect to make the interfacial layer more energetically stable, another possible direction to increase the working temperature is to use AFM with higher Néel temperature (e.g. Mn_3Ir, T_N~730 K) as the Néel temperature and the pinning/unpinning temperature are usually correlated. We add the outlook on increasing the working temperature of AFMTJ in the concluding paragraph on page 9:

"Future studies that continue to increase the TMR ratio and enhance the unpinning temperature of the non-collinear AFM, e.g., using AFM with higher Néel temperature or with stronger pinning at the surface layer, can possibly extend the observed effects to above RT."

Reviewers' Comments:

Reviewer #1:
Remarks to the Author:
The authors have answered the points raised, and the manuscript has been sufficiently improved to recommend publication.

Reviewer #2:
Remarks to the Author:
The authors have thoroughly addressed all my concerns. The results reported in the manuscript are sound and justified to be published in Nature Communications.

Reviewer #3:
Remarks to the Author:
I am glad that the referees answered all my questions and provide more information for their claims. I recommend their work for publication in Nature Communications.