Peer Review File

Anomalous Nernst effect in the noncollinear antiferromagnet Mn5Si3

Web links to the author's journal account have been redacted from the decision letters as indicated to maintain confidentiality.

28th May 24

Dear Dr Sürgers,

Thank you for submitting your manuscript "Anomalous Nernst effect in the noncollinear antiferromagnet Mn_5Si_3" to Communications Materials. We have now received reports from 3 reviewers and I am delighted to say that their comments are very positive. As you will see from the reports appended to the end of this email, there are still many suggestions or concerns before its acceptance. Such as, the comments related to the correlation between anomalous Hall and Nernst conductivity by the reviewer #1 and #2, and the questions about the magnetic phase by the reviewer #3. As such, we are inviting you to revise your manuscript again to address these comments.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

When submitting your revised manuscript, please include the following:

-A response letter with a point-by-point reply to each of the referee comments and a description of changes made. Please include the complete referee report in the response letter. Please note that the response letter must be separate to the cover letter to the editors.

-A marked-up version of the manuscript with all changes to the text in a different colored font. Please do not include tracked changes or comments. Please select the file type 'Revised Manuscript - Marked Up' when uploading the manuscript file to our online system.

-A clean version of the manuscript. Please select the file type 'Article File'.

-An updated Editorial Policy checklist, uploaded as a 'Related Manuscript File' type. This checklist is to ensure your paper complies with all relevant editorial policies. If needed, please revise your manuscript in response to these points. Please note that this form is a dynamic 'smart pdf' and must therefore be downloaded and completed in Adobe Reader. Clicking this link will download a zip file containing the pdf.

In the event that your manuscript is accepted we will provide detailed guidance on our journal policies and formatting. You may however wish to ensure that the manuscript complies with our house style at this stage. See our style and formatting guide (https://www.nature.com/documents/commsj-phys-style-formatting-guide-accept.pdf) and checklist (https://www.nature.com/documents/commsj-phys-style-formatting-checklist-article.pdf) for reference.

Data availability statements and data citations policy: All Communications Materials manuscripts must include a section titled "Data Availability" at the end of the Methods section or main text (if no Methods). More information on this policy, and a list of examples, is available at http://www.nature.com/authors/policies/data/data-availability-statements-data-citations.pdf.

In particular, the Data availability statement should include:
- Accession codes for deposited data
- Other unique identifiers (such as DOIs and hyperlinks for any other datasets)
- At a minimum, a statement confirming that all relevant data are available from the authors
- If applicable, a statement regarding data available with restrictions
- If a dataset has a Digital Object Identifier (DOI) as its unique identifier, we strongly encourage including this in the Reference list and citing the dataset in the Data Availability Statement.

DATA SOURCES: We strongly encourage authors to deposit all new data associated with the paper in a persistent repository where they can be freely and enduringly accessed. We recommend submitting the data to discipline-specific, community-recognized repositories, where possible and a list of recommended repositories is provided at http://www.nature.com/sdata/policies/repositories.

If a community resource is unavailable, data can be submitted to generalist repositories such as figshare or Dryad Digital Repository. Please provide a unique identifier for the data (for example a DOI or a permanent URL) in the data availability statement, if possible. If the repository does not provide identifiers, we encourage authors to supply the search terms that will return the data. For data that have been obtained from publically available sources, please provide a URL and the specific data product name in the data availability statement. Data with a DOI should be further cited in the methods reference section.

Please refer to our data policies at http://www.nature.com/authors/policies/availability.html.

Please use the following link to submit your documents:

[link redacted]

** This url links to your confidential home page and associated information about manuscripts you may have submitted or be reviewing for us. If you wish to forward this email to co-authors, please delete the link to your homepage first **

We hope to receive your revised paper within three months; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we will close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Materials or published elsewhere in the meantime.

Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Xiaokang Li
Editorial Board Member
Communications Materials

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The paper is an interesting study of anomalous Nernst effect in Mn_5Si_3, a noncollinear antiferromagnet with several magnetic phases. The quality of data is high, the analysis is sound. I recommend its publication and invite the authors to improve the quality of their manuscript, by considering the following points:
i) Looking at Figure 3a, it looks like that the jump in magnetization is larger at H_{c1} than at H_{c2}. However, the opposite is true for Fig. 3b. AN inset zooming around H_{c2} would help.
ii) In page 3, there is a sentence ending with these words: "...in agreement with the universal scaling relation of σ_{xy} and σ_{xx}". Since the way these quantities vary which other depends on where you are in the spectrum going from extrinsic to intrinsic, the authors

should clarify a bit more about what they mean by “agreement with the universal scaling”. They should make a statement about the mean free path of carriers in their sample (from sigma_xx) and compare it with the average non vanishing Berry curvature (from sigma_xy) and tell their readers where they are in the spectrum.
iii) According to Figure 3b, spontaneous magnetization at zero field depends on the prior magnetic field. This suggests that without prior field, the sample is multi-domain. What about the anomalous Hall effect? Have the authors tried to see if prior history plays a role? Is the vanishing of the anomalous Hall conductivity at zero temperature survives in presence of a 5 T prior field?
iv) A consequence of equation 4 is a correlation between the amplitude of anomalous Nernst conductivity (alpha_xy) in It has been noted that the amplitude of anomalous Nernst conductivity (alpha_xy) in various topological magnets peaks to a value set by anomalous Hall conductivity times a fraction of k_B/e (see Fig. 4b in Phys. Rev. B 101, 180404 (2020) & Fig. 4 b Sci. Adv., eabf1467 (2021)) . As far as I can see, in the present data the alpha_xy/sigma_xy (Hc1) ratio peaks to ~0.12k_B/e at ~20K . I think this is worth to be mentioned and discussed.
v) The first reports on Anomalous Nernst effect in Mn3Sn were almost simultaneous Nature Phys 13, 1085 (2017) and Phys. Rev. Lett. 119, 056601(2017). In the case of Mn3Ge, a study of ANE was reported in Sci. Adv. 6, eaaz3522 (2020) a few months after Rev. B 100, 085111 (2019). It is surprising that the authors have chosen to neglect papers whose main defect to include a study of anomalous Thermal Hall effect in addition to ANE.

Reviewer #2 (Remarks to the Author):

The manuscript investigates the anomalous Nernst effect (ANE) in Mn_5Si_3 and highlights a sign change in the Nernst signal below 25 K. Crucially, the observation of a finite ANE below 60 K is incompatible with the proposed and debated magnetic structures, imposing further constraints on the magnetic structure. The study is significant, which suggests that ANE can provide valuable insights into the complex magnetic phase transitions in quantum materials. The manuscript is well-written and logically organized, making it accessible to a broad audience in the field. Thus, I recommend the publication in Communications Materials.

However, before acceptance, few points need to be addressed:
1.The abrupt sign change of the Nernst effect below 25 K is intriguing. Provide a deeper analysis or hypothesis on the microscopic interactions causing this phenomenon would be

appreciated. Is it solely attributed to a subtle magnetic transition, or are there other contributing factors?
2.The correlation between the zero-field Nernst signal and the Hall signal requires more detailed explanation. Clarify how these correlations support the conclusions drawn about the proposed rearrangement of magnetic moments below 25 K.
3.The magnetic characterization provided is extensive, but further details on the anisotropy and domain structures in Mn5Si3 would be beneficial. How do these properties influence the observed Nernst effect?
4.Discuss the reproducibility of the results across multiple samples. This will strengthen the reliability of the findings.

Reviewer #3 (Remarks to the Author):

The authors investigate the anomalous Nernst effect of Mn5Si3 in the non-collinear AFM phase. Mn5Si3 is a very complex compound, with complex magnetic phase transitions at low temperatures. Recently, several works reported the anomalous responses in Mn5Si3 thin films, which are inconsistent with the symmetries determined by the neutron scattering results for bulk Mn5Si3. It means that the neutron scattering results may be incorrectly interpreted. Some researchers also term epitaxially strained Mn5Si3 as altermagnet, which is claimed as a different magnetic phase from AFM and FM. I am pleased to see a work that tackles these discrepancies. This work observes a sign change of temperature dependent ANC below 25 K, which is very interesting. I would recommend publication about handling the following issues.
1. From my understanding, the novelty of this work is observing the “subtle rearrangement of the magnetic moment configuration at low temperature”, which challenges the previously-proposed magnetic structure from neutron scattering. However, the authors did not clearly mention theses points, and the discussion in the main text is too vague. The interesting sign change of ANC, and the inconsistency between the proposed AF1 phase from neutron scattering (tT conserved) and the measured AHE and ANE (tT broken) should be pointed directly in the Abstract part. I understand that it is difficult to perfectly explain these discrepancies. However, it is strongly recommended to express these issues clearly in the Abstract so that other relevant researchers can refer to them.
2. Fig. 1 shows that for T below 60 K, the magnetic phase changes from AF1 to AF2 at large magnetic field. For AF2, only 2/3 Mn2 atoms are magnetic while Mn1 atoms are non-magnetic. However, for AF1, both Mn1 and Mn2 atoms are magnetic. Why a large magnetic field can drive Mn1 atoms from magnetic to non-magnetic? Does the AF2 phase for T below

60 K the same as the AF2 phase for T above 60 K? The authors mentioned “and finally by a transition to the field-induced AF2 phase with nonvanishing Mn1 moments aligned parallel to the direction of the magnetic field.” In Page2, which is self-contradictory.

3. The magnetic structure of AF1’ should also be presented in Fig. 2, to avoid possible confusions from the readers.

4. For epitaxially strained Mn5Si3 layers (which are believed to be altermagnet, or spin-splitting AFM), there are undoubtably non-zero net moments instead of zero net moment, as revealed by arXiv: 2401.02275v1, from the same group of Ref. 31 and Ref. 33. Besides, the net moment and anomalous Nernst effect for altermagnet Mn5Si3 thin film have also been confirmed independently by another research group (Sci. Adv. 10, eadn0479 (2024) and arXiv:2403.13427).

5. In page3, the authors claims “parallel to the easy axis of magnetization c”. However, I did not see any evidence for the easy axis of magnetization in the non-collinear AFM phase.

6. In page6, the authors attribute the AHE and ANE for T below 60 K to “a slight tilting of the magnetic state”. Similar arguments have been used to explain the AHE and ANE in epitaxially strained Mn5Si3 (Ref. 31, Sci. Adv. 10, eadn0479 (2024)). Does the residual strain in Mn5Si3 single crystal lead to this “tilting”? The authors may carry out similar ANE and AHE measurements for samples with no strain.

Response to Reviewer reports

We would like to thank all three Reviewers for their critical and thorough reading of our manuscript and their valuable comments. In our revised manuscript, we have taken into account all points raised by the Reviewers. In the following, we address the Reviewers' comments in a point-by-point fashion. The various changes that have been made to the text are marked in red font in the revised version of the manuscript. Changes that relate directly to the Reviewer´s comments are repeated in red in this response.

Reviewer #1:

The paper is an interesting study of anomalous Nernst effect in Mn5Si3, a noncollinear antiferromagnet with several magnetic phases. The quality of data is high, the analysis is sound. I recommend its publication and invite the authors to improve the quality of their manuscript, by considering the following points:

i) Looking at Figure 3a, it looks like that the jump in magnetization is larger at Hc1 than at Hc2. However, the opposite is true for Fig. 3b. AN inset zooming around Hc2 would help.

Reply 1: The main panel of Fig. 3a clearly shows that the jump in magnetization at Hc2 is roughly ten times larger than at Hc1 in agreement with Fig. 3b (please note the different scaling for the left and right vertical axis). However, we believe that the Reviewer got confused with the two arrows (one single-headed, one two-headed) at the position of Hc2 which might suggest that the jump is very tiny. Therefore, we have exchanged the arrows indicating "Hc1" and "Hc2" by vertical markers for better clarity in a revised Fig. 3a.

------

ii) In page 3, there is a sentence ending with these words: "...in agreement with the universal scaling relation of σxy and σxx". Since the way these quantities vary which other depends on where you are in the spectrum going from extrinsic to intrinsic, the authors should clarify a bit more about what they mean by "agreement with the universal scaling". They should make a statement about the mean free path of carriers in their sample (from sigma_xx) and compare it with the average non vanishing Berry curvature (from sigma_xy) and tell their readers where they are in the spectrum.

Reply 2: We agree with the Reviewer that this statement was somewhat sloppily worded and have now given more details with new references concerning the regime of electron scattering for the present samples.

-Text added: σ_{xy} is affected by extrinsic contributions arising from skew scattering or side-jump mechanism and by the intrinsic Berry-phase contribution [3]. In the dirty and ultraclean regimes of electron scattering the dependence of σ_{xy} on the longitudinal conductivity σ_{xx} can be described by a power law $\sigma_{xy} \propto \sigma^{\alpha}_{xx}$ with $\alpha > 0$. In the intermediate regime of moderate conductvity $3 \times 10^3 - 5 \times 10^5\ \Omega^{-1}cm^{-1}$ the AHE is predominated by the intrinsic Berry-phase contribution where σ_{xy} is independent of σ_{xx} [39, 40]. In the present case, the single crystals with $\sigma_{xy} = 150\ \Omega^{-1}cm^{-1}$ and $\sigma_{xx} = 7400\ \Omega^{-1}cm^{-1}$ fall just within this intermediate regime as discussed earlier [20, 41], similar to the noncollinear antiferromagnets Mn_3Ge and Mn_3Sn [42]. From $\rho_{xx} = 135\ \mu\Omega cm$, a Fermi velocity $v_F = 2 \times 10^6$ m/s [43], and an electron density $n = 1 \times 10^{22}\ cm^{-3}$ [20] we estimate an electron mean freepath of 5.2 nm, larger than several lattice parameters, supporting the classification of this material as a moderately conductive metal.

------

iii) According to Figure 3b, spontaneous magnetization at zero field depends on the prior magnetic field. This suggests that without prior field, the sample is multi-domain. What about the anomalous Hall effect? Have the authors tried to see if prior history plays a role? Is the vanishing of the anomalous Hall conductivity at zero temperature survives in presence of a 5 T prior field?

Reply 3: The Reviewer is right. Before sweeping the magnetic field, the sample is in a multidomain state as inferred from the magnetization and Hall conductivities, see revised Figs. 3a (left inset) and 3g. In the latter we have added data obtained in the first field sweep starting from the virgin magnetic state at zero field to 8 T. In Fig. 3h we plotted the remanent Hall conductivity $\sigma_{xy}(0)$ which is obtained after performing a full field cycle. This is now mentioned in the revised text.

- Text added: Before sweeping the magnetic field, the sample has a magnetization much lower than the remanent magnetization $M_R(0)$, see left inset Fig. 3a. This indicates that the antiferromagnetic structure is not fully compensated in the magnetically pristine state and exhibits weak moments that allow the domain configuration to be changed by applying a magnetic field and generate a net magnetization along the field direction. The hysteresis around zero field is not observed when the magnetic field is oriented perpendicularly to the c axis for all temperatures below 60 K [19].

- Text modified: The absolute value of the transverse resistivity $\rho_{yx}(0)$ in the remanent state at zero field, i.e., after completing a full magnetic field cycle, increases with decreasing temperature down to 25 K and then decreases and even vanishes at T = 5 K while $M_R(0)$ remains non zero.

- Text added to caption Fig. 3g: Open symbols represent data in the virgin magnetic state before sweeping the field from zero to +8 T.

- Text added: In the magnetically virgin state before sweeping the magnetic field, σxy is very small compared to the remanent state σxy(0) observed after completion of a field sweep to high fields, similar to the magnetization M(H), Fig. 3(a). A strong difference between the temperature dependences of ρyx(0) was already reported earlier when the sample was cooled down in zero field from above TN1 and subsequently polarized in a magnetic field of ±3T [20].

- Text modified: The absolute values of the remanent σxy(0) and σxy(Hc1) plotted in Fig. 3(h) gradually...

------ iv) A consequence of equation 4 is a correlation between the amplitude of anomalous Nernst conductivity (alpha_xy) in It has been noted that the amplitude of anomalous Nernst conductivity (alpha_xy) in various topological magnets peaks to a value set by anomalous Hall conductivity times a fraction of k_B/e (see Fig. 4b in Phys. Rev. B 101, 180404 (2020) & Fig. 4 b Sci. Adv., eabf1467 (2021)) . As far as I can see, in the present data the alpha_xy/sigma_xy (Hc1) ratio peaks to ~0.12k_B/e at ~20K . I think this is worth to be mentioned and discussed.

Reply 4: We thank the Reviewer for this comment and have addressed this point in the revised manuscript with new references.

- Text added: In various topological magnets, the ratio |αxy/σxy| increases with temperature from 1 μV/K and often approaches k_B/e = 86 μV/K at room temperature or above [10, 11, 46] and thus follows a universal scaling [47], with the exception of UCo0.8Ru0.2Al which exhibits a colossal ANE [48]. In the present case, maximum values of |αyx(0)/σxy(0)| = 3.5 μV/K and |αyx(Hc1)/σxy(Hc1)| = 5.8 μV/K are obtained for T = 40 K, which are only somewhat smaller when compared to values of other topological materials at this low temperature [10, 11, 46]. The similar ratios obtained in the AF1 and AF1' phases of Mn5Si3 support the correlation between the two properties αxy and σxy that are predominated by intrinsic effects of the Berry curvature in both magnetic phases.

------

v) The first reports on Anomalous Nernst effect in Mn3Sn were almost simultaneous Nature Phys 13, 1085 (2017) and Phys. Rev. Lett. 119, 056601(2017). In the case of Mn3Ge, a study of ANE was reported in Sci. Adv. 6, eaaz3522 (2020) a few months after Rev. B 100, 085111 (2019). It is surprising that the authors have chosen to neglect papers whose main defect to include a study of anomalous Thermal Hall effect in addition to ANE.

Reply 5: It was not our intention to neglect important papers dealing with the thermoelectric effects of Mn3Sn or Mn3Ge. Unfortunately, the two publications mentioned by the reviewer, Phys. Rev. Lett. 119, 056601(2017) and Sci. Adv. 6, eaaz3522 (2020), escaped our attention. They have now been added as Refs. 10 and 11, respectively, in the revised manuscript. We also added a sentence mentioning the published work of the Righi-Leduc effect on Mn3Sn and Mn3Ge. Furthermore, the citations have been reorganised to link the content to the corresponding references in more detail.

- Text added: In addition to the thermoelectric ANE, a sizeable thermal Hall effect (Righi-Leduc effect) generated by the Berry curvature has been reported for these two materials [10, 11].

Reviewer #2:

The manuscript investigates the anomalous Nernst effect (ANE) in Mn5Si3 and highlights a sign change in the Nernst signal below 25 K. Crucially, the observation of a finite ANE below 60 K is incompatible with the proposed and debated magnetic structures, imposing further constraints on the magnetic structure. The study is significant, which suggests that ANE can provide valuable insights into the complex magnetic phase transitions in quantum materials. The manuscript is wellwritten and logically organized, making it accessible to a broad audience in the field. Thus, I recommend the publication in Communications Materials.

However, before acceptance, few points need to be addressed: 1.The abrupt sign change of the Nernst effect below 25 K is intriguing. Provide a deeper analysis or hypothesis on the microscopic interactions causing this phenomenon would be appreciated. Is it solely attributed to a subtle magnetic transition, or are there other contributing factors?

Reply 6: We agree that a deeper analysis of the observed phenomena would be desired. However, this would require a calculation of the electronic band structure and the Berry curvature of Mn5Si3 which cannot be performed without a prior knowledge of the correct magnetic structure not available so far. We therefore restrict the analysis and discussion to a comparison with other noncollinear systems. We have added a discussion of the change of the Fermi surface inferred from the zero Nernst coefficient around 20 K. We also discuss possible factors that could affect the Nernst effect like residual strain, see also our Reply 15 below, or deviations from stoichiometry.

-Text added: We mention that an additional phase has been reported to exist in single crystals grown out of Cu flux which was attributed to strain or stress introduced by Cu inclusions from the growth [31].

-Text added: Possible deviations from the stoichiometry must always be considered. It has been previously reported that large displacements of Mn atoms from their high-symmetry positions lower the structural symmetry in Mn_3SnN or Mn_3GaN resulting in spin canting and allowing the observation of non-vanishing AHE [56]. We cannot completely rule out such effects but emphasize that an x-ray analysis of a pulverized piece from the ingot sample yielded lattice parameters that deviate less than 0.1 % from recently published data of a polycrystal [17, 20].

2.The correlation between the zero-field Nernst signal and the Hall signal requires more detailed explanation. Clarify how these correlations support the conclusions drawn about the proposed rearrangement of magnetic moments below 25 K.

Reply 7: In the revised manuscript we have added Eqs. 5 and 6 to formally show the link between the Nernst effect and the Hall effect for the intrinsic case. Furthermore, we suggest a possible explanation of how the electronic band structure might affect the ANE, being zero around 20 K.

-Text added: In addition, the coercive fields of M(H), ρ_{yx}, and S_{yx} around zero magnetic field are of similar size and continuously decrease from 15 K to 60 K as previously observed for $\rho_{yx}(T)$ [20]. This indicates that the effects of the magnetic field on ρ_{yx} and S_{yx} are of similar origin.

-Text added: Whereas $\sigma_{xy}(0)$ (in phase AF1) and $\sigma_{xy}(H_{c1})$ (in phase AF1') have opposite signs but are of the same magnitude [Fig. 3(h)], $S_{yx}(0)$ also has the opposite sign of $S_{yx}(H_{c1})$ but only reaches half of its value. Both effects, AHE and ANE, do not scale with the change of the magnetization at zero field and Hc1, confirming that they arise from topology and the intrinsic Berry-curvature effect like other antiferromagnets with a nontrivial magnetic structure. (...)

-Text added: The ANE represented by S_{yx} or α_{yx} is a Fermi surface property (Eq. 5). A sign change of these parameters at 20 K inevitably means that they reach zero around 20 K. Hence, from Eq. 4 it follows that $(\partial\sigma_{xy}/\partial E)_\mu = 0$, i.e., $\sigma_{xy}(E)$ has a maximum or minimum at the Fermi surface which is shifting through the Fermi level with increasing temperature from below 20 K to above 20 K. A change of the Fermi surface at low temperatures can be also inferred from the variation of the ordinary Hall effect of Mn_5Si_3 thin films [18]. In that case the ordinary Hall effect $\rho^0_{yx} = R_0\mu_0H$ could be separated from the measured signal, revealing a change of the ordinary Hall constant R_0 or the effective charge carrier density n_H with temperature and a maximum (minimum) of R0 (nH) around 20 K, see Fig. 5(g). At this point, ab initio calculations of the electronic bandstructure and σxy are necessary for a more quantitative analysis of the AHE and ANE following the Berry-phase concept by application of Eqs. 5 and 6. Such calculations involve a knowledge of the correct magnetic configuration in the ground state of bulk Mn5Si3, which has not been determined so far.

Please see also our Replies 2 & 4 to Reviewer #1.

3.The magnetic characterization provided is extensive, but further details on the anisotropy and domain structures in Mn5Si3 would be beneficial. How do these properties influence the observed Nernst effect?

Reply 8: We have added text regarding differences in the magnetization and AHE between the magnetically virgin and polarized state, see also our Reply 3 to Reviewer #1. We argue, why it is unlikely that the magnetic anisotropy is the origin of the anisotropy of the AHE and ANE.

-Text added: Finally, we argue that it is unlikely that the magnetocrystalline anisotropy is the origin of the anisotropic behaviour of both thermal effects. In the AF1 phase, nonzero AHE and ANE at zero field are observed only for H oriented parallel to the c axis but not along the a or b axes, see Supplementary Fig. 1 and Ref. [19], where the latter is the magnetic easy axis in the AF1 phase [30].

4.Discuss the reproducibility of the results across multiple samples. This will strengthen the reliability of the findings.

Reply 9: We have observed the ANE and the sign change on more than one samples. In fact, the data presented in the Supplementary Information were obtained on a different Mn5Si3 single crystal. This crystal also shows the sign change of the zero-field ANE for H applied along the crystallographic c axis, see Supplementary Figure 1 (a-c). We have mentioned this in the main text of the revised manuscript “This behavior is also observed on another single crystal for H applied parallel to the c direction…” and in the revised caption of Supplementary Fig. 1.

-Modified figure caption of Supplementary Fig. 1: Anomalous Nernst effect of another Mn5Si3 single crystal, different from the crystal discussed in the main text, for magnetic fields H and heat flows Q applied along different crystallographic directions a, b, or c. (a-c)

H applied along c, heat flow Q along a over a length of 0.52 mm. (d-e) H applied along the orthorhombic a direction, heat flow Q along b over a length of 1.2 mm. (f) H applied along b, heat flow Q along a over a length of 0.52 mm.

Reviewer #3: The authors investigate the anomalous Nernst effect of Mn5Si3 in the non-collinear AFM phase. Mn5Si3 is a very complex compound, with complex magnetic phase transitions at low temperatures. Recently, several works reported the anomalous responses in Mn5Si3 thin films, which are inconsistent with the symmetries determined by the neutron scattering results for bulk Mn5Si3. It means that the neutron scattering results may be incorrectly interpreted. Some researchers also term epitaxially strained Mn5Si3 as altermagnet, which is claimed as a different magnetic phase from AFM and FM. I am pleased to see a work that tackles these discrepancies. This work observes a sign change of temperature dependent ANC below 25 K, which is very interesting. I would recommend publication about handling the following issues.

1. From my understanding, the novelty of this work is observing the “subtle rearrangement of the magnetic moment configuration at low temperature”, which challenges the previously-proposed magnetic structure from neutron scattering. However, the authors did not clearly mention theses points, and the discussion in the main text is too vague. The interesting sign change of ANC, and the inconsistency between the proposed AF1 phase from neutron scattering (tT conserved) and the measured AHE and ANE (tT broken) should be pointed directly in the Abstract part. I understand that it is difficult to perfectly explain these discrepancies. However, it is strongly recommended to express these issues clearly in the Abstract so that other relevant researchers can refer to them.

Reply 10: We thank the Reviewer for this comment and agree that the abstract was perhaps written too cautiously. We have now extended the abstract by considering the comment of the reviewer.

-Text modified: In the noncollinear phase, we observe an unusual sign change of the zero-field Nernst signal with a concomitant decrease of the Hall signal and a gradual reduction of the remanent magnetization. Furthermore, a symmetry analysis of the proposed magnetic structures shows that both effects should actually vanish. These results indicate a symmetry-breaking modification of the magnetic state with a rearrangement of the magnetic moments at low temperatures, thus questioning the previously reported models for the noncollinear magnetic structure obtained from neutron scattering.

---

2. Fig. 1 shows that for T below 60 K, the magnetic phase changes from AF1 to AF2 at large magnetic field. For AF2, only 2/3 Mn2 atoms are magnetic while Mn1 atoms are non-magnetic. However, for AF1, both Mn1 and Mn2 atoms are magnetic. Why a large magnetic field can drive Mn1 atoms from magnetic to non-magnetic? Does the AF2 phase for T below 60 K the same as the AF2 phase for T above 60 K? The authors mentioned "and finally by a transition to the field-induced AF2 phase with nonvanishing Mn1 moments aligned parallel to the direction of the magnetic field." In Page2, which is self-contradictory.

Reply 11: These are very good questions which often occur when discussing the hitherto known details of the magnetic structure of Mn5Si3 derived from neutron diffraction. From the previous reports it seems to be clear that the field-induced phase in strong magnetic fields and the zero-field AF2 phase have similar magnetic properties. However, it is still not clear, whether the magnetic structures in both phases are really identical or not and whether the Mn1 moment survives in a high field or not. We therefore mention that the field-induced collinear phase at high fields is similar but presumably not identical to the zero-field AF2 phase.

We agree with the Reviewer that the sentence on page 2, as it stands, is confusing because it refers to the different model proposed by Biniskos et al. We have modified this paragraph to make these points clearer to the reader.

-Text added or modified: (...) have been proposed by Biniskos et al. [28], resulting in a ground-state spin arrangement in the AF1 phase that is different from the previous AF1 phase reported by Brown et al. [26]. Here, in zero magnetic field the Mn1 atoms carry a magnetic moment oriented along the orthorhombic b axis while the two thirds of Mn2 atoms carry a magnetic moment with a significant component along the c axis, respectively (Fig. 2c). (...)

Starting from the proposed ground state (Fig. 2c) and increasing the magnetic field applied along the c axis, a "spin flop" phase with coplanar Mn2 moments in the a-b plane is succeeded by an AF1'-like phase, where the Mn1 moments are also predominantly orientated in the a-b plane, but start to have a component along the c axis. At even higher fields, a transition to a field-induced phase is proposed, with collinear and antiparallel oriented Mn2 moments along the b axis and nonvanishing Mn1 moments aligned parallel to the direction of the magnetic field. All models for the magnetic structure reported so far suggest, that the field-induced phase at high magnetic fields has similar properties as the zero-field AF2 phase, although it is not clear, whether the Mn1 moment eventually collapses or not [25, 28].

------

2. The magnetic structure of AF1' should also be presented in Fig. 2, to avoid possible confusions from the readers.

Reply 12: Unfortunately, a model of the magnetic structure for the AF1' phase obtained from, e.g, neutron diffraction in magnetic field has not been reported so far. The field-induced transition from AF1 to an intermediate AF1' phase has been inferred from previous neutron scattering and electronic transport measurements [20,21,25] as mentioned in the manuscript. From modelling of the spinwave spectrum, the proposed model of Biniskos et al. [28] generates several field-induced phases, among them one phase with all magnetic moments, Mn1 and Mn2, oriented in the a-b plane. The authors of Ref. [28] mention that "This phase is possibly associated with the reported AFM1' in several studies …". These points have been considered in the revised version of the manuscript, see our Reply 11. The only magnetic structures determined from neutron diffraction so far are the AF1 and AF2 phases in zero magnetic field, shown in Fig. 2, which we would like to leave unchanged.

----

3. For epitaxially strained Mn5Si3 layers (which are believed to be altermagnet, or spin-splitting AFM), there are undoubtably non-zero net moments instead of zero net moment, as revealed by arXiv: 2401.02275v1, from the same group of Ref. 31 and Ref. 33. Besides, the net moment and anomalous Nernst effect for altermagnet Mn5Si3 thin film have also been confirmed independently by another research group (Sci. Adv. 10, eadn0479 (2024) and arXiv:2403.13427).

Reply 13: We thank the Reviewer for this comment and have added the publications Sci. Adv. 10, eadn0479 (2024) and arXiv:2403.13427 to our list of references. We agree with the Reviewer that the epitaxial films investigated in experiment have a small but finite magnetization and the zero net moment was only obtained in the ab initio calculations. We have rephrased this paragraph accordingly.

-Text added: This gives rise to anomalous Hall and Nernst effects experimentally observed in thin epitaxial Mn5Si3 films with a small net magnetization of 20 - 60 mμB/f.u. [36]. Recently, spinorbit torque switching of the Néel vector has been demonstrated for strained Mn5Si3 films and a strong enhancement of the ANE upon Mn doping of a Mn5Si3 film has been attributed to the shift of the Fermi level [37, 38].

------

4. In page3, the authors claims “parallel to the easy axis of magnetization c”. However, I did not see any evidence for the easy axis of magnetization in the non-collinear AFM phase.

Reply 14: The c axis is the only axis along which a remanent magnetization and a finite Hall effect rho_yx(0) are observed, see Fig. 3a (inset) and Fig. 3c. This is not the case for a field applied perpendicularly to the c axis, see Ref. [19]. A DFT calculation suggests that b and c are the first and second referred easy axes [30]. We have made the appropriate changes in the text.

-Text modified: Inelastic neutron-scattering measurements of the low-energy dynamics reveal a single-anisotropy energy gap in the AF1 phase and a double gap in the AF2 phase arising from a biaxial anisotropy [28, 29]. Concomitantly performed density-functional theory calculations indicate that in the AF1 phase the crystallographic b axis is the easy axis, whereas in the AF2 phase the b and c axes are the first and second preferred magnetic easy axes, respectively, and a is the hard axis [30].

-Text modified: Fig. 3(a) shows a typical magnetization curve of Mn_5Si_3 at T = 35 K with the magnetic field applied parallel to the crystallographic c axis.

-----

5. In page6, the authors attribute the AHE and ANE for T below 60 K to “a slight tilting of the magnetic state”. Similar arguments have been used to explain the AHE and ANE in epitaxially strained Mn_5Si_3 (Ref. 31, Sci. Adv. 10, eadn0479 (2024)). Does the residual strain in Mn_5Si_3 single crystal lead to this “tilting”? The authors may carry out similar ANE and AHE measurements for samples with no strain.

Reply 15: Residual strain could cause a tilting or canting of the magnetic structure. We now discuss this at the end of the discussion. However, we argue that the strong anisotropy of the observed effects and the careful mounting of the sample without clamping makes it unlikely that residual strain - if not uniaxial along the c direction – is the reason for the observed behaviour. We also consider the possibility of deviations from stoichiometry, see our Reply 6. A theoretical (DFT) analysis of the magnetic structure dependence on microscopic details, such as strain, is only possible with a previous knowledge of the ground state of the system. As mentioned in our Reply 6 to Reviewer #2, this has not been determined so far.

-Text added: The question is whether this could be caused by a residual strain in the single crystal similar to epitaxially strain prevailing in Mn_5Si_3 films [33, 36, 37]. Strain caused by Cu impurities was presumably responsible for generating additional magnetic phases in Mn_5Si_3 single crystals [31]. Uniaxial residual stress was reported to cause a change of the antiferromagnetic structure of polycrystalline Mn_3Sn from non-collinear and coplanar to non-coplanar [55]. In the present case, where the different phases observed in the electronic transport measurements agree with the results from neutron-diffraction experiments on different single crystals, it is unlikely that the residual stress in the crystal only evolves along the c axis to generate a nonzero AHE and ANE. The samples were not clamped in the ANE device with pressure [Fig. 4(c)] but only glued to the Cu banks of which the hot end was always freely suspended. ANE measurements shown in Fig. 5 and Supplementary Fig. 1 with the magnetic field along the c axis were done on different samples with c oriented perpendicular to the plane or in the plane of the sample plate, respectively. Moreover, the strong variation of the ANE and the sign change in a small range of temperature around 20 K is even more unlikely as being due to strain. Thus, we believe that residual strain cannot explain the appearance of a finite AHE and ANE in contrast to what is expected from the symmetry analysis.

Summary of changes:

Revised text: marked in red font

Revised figures: Figures 3 and 5

12 new references: [10, 11, 16, 31, 37, 38, 40, 43, 47, 48, 55, 56]

Paragraph "For Mn5Si3, maximum values ISyx(0)I = 0.05 muVK-1 and (...)" **shifted** from Discussion section (p. 7) to Results section (p. 5).

13th Aug 24

Dear Dr Sürgers,

Your manuscript titled "Anomalous Nernst effect in the noncollinear antiferromagnet Mn_5Si_3" has now been seen again by our referees, whose comments appear below. In light of their advice I am delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Materials.

We therefore invite you to edit your manuscript to comply with our journal policies and formatting style in order to maximise the accessibility and therefore the impact of your work.

EDITORIAL REQUESTS
* Your manuscript should comply with our policies and format requirements, detailed in our style and formatting guide (https://www.nature.com/documents/commsj-phys-style-formatting-guide-accept.pdf).

* Please edit your manuscript according to the editorial requests in the attached table, and outline revisions made in the right hand column. If you have any questions or concerns about any of our requests, please do not hesitate to contact me. It is important that each request be addressed in order to avoid delays in accepting your manuscript. Please upload the completed table with your manuscript files as a Related Manuscript file.

* The editorial requests table also includes a full list of the files that must be provided upon resubmission. Please upload your files according to this table.

* An updated editorial policy checklist that verifies compliance with all required editorial policies must be completed and uploaded with the revised manuscript. All points on the policy checklist must be addressed; if needed, please revise your manuscript in response to these points. Please note that this form is a dynamic 'smart pdf' and must therefore be downloaded and completed in Adobe Reader. Clicking this link will download a zip file containing the pdf.

Editorial policy checklist: https://www.nature.com/documents/nr-editorial-policy-checklist.zip

OPEN ACCESS

Communications Materials is a fully open access journal. Articles are made freely accessible on publication. For further information about article processing charges, open access funding, and advice and support from Nature Research, please visit https://www.nature.com/commsmat/open-access

At acceptance, you will be provided with instructions for completing the open access licence agreement on behalf of all authors. This grants us the necessary permissions to publish your paper. Additionally, you will be asked to declare that all required third party permissions have been obtained, and to provide billing information in order to pay the article-processing charge (APC).

Please use the following link to submit your revised files:

[link redacted]

** This url links to your confidential home page and associated information about manuscripts you may have submitted or be reviewing for us. If you wish to forward this email to co-authors, please delete the link to your homepage first **

We hope to hear from you within two weeks; please let us know if the process may take longer.

Best regards,

Xiaokang Li
Editorial Board Member
Communications Materials
orcid.org/0000-0001-8828-5044

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors have addressed the points raised by the referees in a satisfying manner. I recommend publication.

Reviewer #2 (Remarks to the Author):

I have carefully reviewed the revised manuscript and the authors have addressed all the concerns raised during the review process. The revised manuscript is now suitable for publication in Communications Materials.

Reviewer #3 (Remarks to the Author):

The authors address my previous comments and questions very well. The quality of the manuscript is greatly improved. Thus, I recommend its publication as it is.