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

10th Jan 22

Dear Dr Helm,

Thank you for submitting your manuscript, "Antiskyrmions and their electrical footprint in crystalline mesoscale structures.", to Communications Materials. It has now been seen by 4 referees, whose comments are appended below. You will see that while they find your work of potential interest, they have raised substantial concerns that must be addressed. In light of these comments, we cannot accept the manuscript for publication, but are interested in considering a revised version that addresses these serious concerns.

In particular, you will see that Reviewer #2 raises a very significant concern regarding the lack of direct experimental evidence of antiskyrmions in the system, and the fact that previous studies have shown that thick films do not host antiskyrmions but instead bubble domains or possibly other spin textures. Furthermore, all reviewers have raised important questions and concerns about the contributions to the THE data. We believe these are points that need to be addressed in order to further consider the manuscript for publication. We would naturally expect all other comments of the referees to be appropriately responded to, including their requests for further information and discussion.

We hope you will find the referees' comments useful as you decide how to proceed. Should further experimental data or analysis allow you to address these criticisms, we would be happy to look at a substantially revised manuscript. However, please bear in mind that we will be reluctant to approach the referees again in the absence of major revisions. If the revision process takes significantly longer than three months, we will 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.

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

Please use the following link to submit your revised manuscript files:

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We understand that due to the current global situation, the time required for revision may be longer than usual. We would appreciate it if you could keep us informed about an estimated timescale for resubmission, to facilitate our planning. Of course, if you are unable to estimate, we are happy to accommodate necessary extensions nevertheless.

Please do not hesitate to contact me if you have any questions or would like to discuss the required revisions further. Thank you for the opportunity to review your work.

Best regards,

Alannah Hallas, PhD
Editorial Board Member
Communications Materials
orcid.org/0000-0002-0892-0982

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The manuscript „Antiskyrmions and their electrical footprint in crystalline mesoscale structures“ by Winter et al. reports the study of the THE of antiskyrmions (ASKs) in MnPtSn and its tunability by device size, field, and temperature. The authors perform MOKE microscopy in combination with electrical measurements to provide evidence of the THE of ASKs in MnPtSn.

The paper is well written (though the supplementary material might require some proof reading) and reports an interesting result. While I find the topic very exciting, there are several points I would like the authors to address before I can recommend publication:

1) Page 2: The speed of antiskyrmions has been cited misleadingly, incl. several publications on antiferromagnetic (AFM) skyrmions and calculations that use ultra-low damping in the range of YIG. A distinction to AFM skyrmions and concrete examples should be included.

- 2) Page 4: The authors report the simulations to “inevitably” showing AKS. There are also reports of skyrmions in MnPtSn. The statement should be made clearer.
- 3) Page 5, line 2-3: “due to a change in the magnetic texture” is very generic and expected. Try to rephrase or omit.
- 4) Page 5: That the sample stays in a saturated state while the field is decreasing has been seen in many skyrmion materials (there is also a minor typo in ref. 27) and is to be expected also without the anisotropic exchange.
- 5) Why is there no contribution of the THE visible while there are clearly skyrmion pockets forming before the reported onset of the THE? The supplementary material clearly shows these partial lattices.
- 6) If I get it correctly, the simulations were freely tuned to reproduce the experiment. The authors should use experimentally measured input parameters (as far as possible) and/or slightly tuned bulk parameters and check if the simulations still match the experiment.
- 7) Page 6, line 2: is J1 ferromagnetic or weak antiferromagnetic?
- 8) Page 6: “the yielded magnetization hysteresis... is congruent...”. The shown loop would look the same for skyrmions and thus does not contribute to the story, right?
- 9) The authors state that the shallow slope in the Hall resistance right before the saturation is caused by the negative THE of the ASKs. However, while for skyrmions this would be easily visible as a positive contribution, the antiskyrmion contribution is similar to the one of shrinking magnetic textures. While the domains can easily contract, the topological stabilization of the ASKs will counteract this and lead to a longer presence and slower increase in the net magnetization, i.e. a different slope. These effects should be carefully and clearly discussed and disentangled in the main text. The authors should provide direct experimental transport evidence that the signal is indeed a negative THE contribution, such as the transition into skyrmions by an in-plane field (10.1038/s41467-020-14925-6).

Reviewer #2 (Remarks to the Author):

In their work, Winter and co-authors report on the electrical footprint of antiskyrmions in crystalline mesoscale structures. This report, if accurate, would serve as a large advance in the field in topological magnetism. However, I have major concerns that preclude publication at this time, as detailed below.

From the experimental imaging, it is impossible to determine if the observed structures are really antiskyrmions. While antiskyrmions have been clearly observed in Mn_{1.4}PtSn using L-TEM measurements, the techniques here give no information on the actual topological charge of the object, and the signal being seen observed can just as easily be explained by skyrmions, non-trivial bubbles, or even higher order excitations. All previous direct reports of antiskyrmions using imaging techniques have been through thin films (thicknesses on the order of 100 nm, significantly thinner than the system studied here). However, for thick films, neutron scattering measurements recently suggested that “bulk Mn_{1.4}PtSn does not support antiskyrmions” (PHYSICAL REVIEW B 102, 174447 (2020)) and suggests dipole-dipole interactions need to be strongly considered (which is not done in the model used in this current work, as H_{demag} is ignored).

The atomistic spin simulations do not seem to be very relevant to the system being studied. The sample dimensions (50 nm x 50 nm x (3 unit cells) in the vertical axis do not make sense to me. The system being studied has a much larger thickness, and the observed magnetic features from experiment are effectively two orders of magnitude larger than those observed in the results of atomistic spin simulations. While qualitatively producing antiskyrmions, the relevance to the system studied here is questionable, at best. Further, it doesn't appear that demagnetization energies are considered, which may be non-negligible for such thick, bulk-like systems.

The authors should estimate the expected contribution to hall resistivity per anti-skyrmion. Due to the size of the objects, one would expect non-adiabatic contributions to be small. But if purely adiabatic contributions are present, I don't think this magnitude of effect is expected.

If possible, the authors should show a switching loop as measured by i.e. SQUID and then compare the Hall measurements to the M-H curve. This is a common method to measure THE, i.e. as done in Nature Materials 16, 898–904 (2017) and Nature Communications 12, 2758 (2021).

Can the authors exclude effects arising from a non-uniform extraordinary Hall effect due to variations in local microstructure? If so, how do they exclude this effect?

The authors state “therefore, we tuned the classical parameters in atomistic spin-dynamic simulations to clearly show the effects seen in the experiments.” Am I to read this as that the values utilized in atomistic spin simulations were simply varied in order to get good agreement with experiment? Is there a reference to the values being used available from the literature?

The measured domain widths and “antiskyrmion” widths are clearly more than 100 nm in extent and significantly larger than those reproduced in atomistic modeling. Further, as far as I'm aware, antiskyrmions as observed are typically sub-100 nm – again, I'm concerned these are more closely related to a bubble type configuration. At this size, there should be something done to verify the spin structure sketched in Fig. 1a.

The fit in Fig. 3c is not very convincing, considering the uncertainty in the data points. Further, why are MFM measurements for 5 μm and 1.5 μm thick films shown, but not plotted in Fig. 3c, while the images where points in Fig. 3c were obtained are not found in the main text or supplemental. This would be quite useful.

I would be happy to review the work again provided my concerns are addressed. However, many of the above concerns are significant, and preclude publication in the current form.

Reviewer #3 (Remarks to the Author):

This manuscript reports a thorough study of magnetic textures, including antiskyrmions, in patterned hall bar structures of Mn_{1.4}PtSn single crystals using Hall measurements and MOKE imaging. The authors observe clear correlation between the detected topological Hall effect and the imaging of antiskyrmions, demonstrating the existence and phase diagram of antiskyrmions in Mn_{1.4}PtSn microstructures. They also perform atomistic simulations which explain the experimental results by the competition of fundamental interactions including exchange, DMI, and anisotropy. The experimental data are of high quality, which, together with atomistic simulations, convincingly reveal the behavior of antiskyrmions and the phase space of magnetic textures in Mn_{1.4}PtSn and the underlying mechanisms. I recommend its publication in Communications Materials. Below are a few minor comments for the authors to address in the revised manuscript.

1. Figure 1c, especially its inset, should be compared with the magnetization hysteresis loop of the same sample in the main text. I suggest the authors move Figure S6a in the supplement to the main text, in Figure 1c. In addition, the inset of Figure S6a (magnetization hysteresis) shows a two-step switching in the down sweep direction, while the inset of Figure 1c (Hall resistivity) shows a one-step switching for the same sample. What is the reason for this inconsistency?
2. On page 5 of the main text, line 10 from the top, it refers to "as we show in Supplementary Note S5". Should it be either "Supplementary Note S4" or "Supplementary Note Figure S6a"?
3. The MOKE images in Figure 1d of the main text seem to be for the same sample used for the MOKE images in Figure S7 of the supplement. However, the field values for the appearance of similar features are different between the two sets of MOKE images. Can the authors comment on this discrepancy?

Reviewer #4 (Remarks to the Author):

In this manuscript, the authors have built on a significant back-catalogue of research into high quality crystals of tetragonal Heusler alloy Mn_{1.4}PtSn to demonstrate and discuss the electrical signatures of antiskyrmions. The complex magnetic interactions that result in these magnetic textures are of great and ongoing research interest, and electrical detection is one of the key challenges to overcome for developing computing technologies that rely on such textures. Antiskyrmions (ASKs) are a more recent addition to the zoo of magnetic quasiparticles being investigated for these reasons, and the current manuscript is a thorough, well-constructed and original study that will be of immediate interest to, and will benefit, the spintronics and magnetic materials research community. This is the one of very few works that report on the electrical detection of ASKs, and the correlation of MOKE, magnetisation and magnetotransport measurements on carefully microstructured single-crystal devices, with support from both FMR measurements and atomistic simulations, is very convincing.

There are no significant flaws in this paper, and the data and methodology are very well presented. The references, abstract, introduction and conclusions are all appropriate. I am happy to recommend this article be accepted in Communications Materials, however I have

one or two questions for the authors below, as well as several suggested minor corrections and edits.

It has been shown before, in multilayer films containing skyrmions, that ρ_{xy} is made up of both AHE and THE contributions from the skyrmions, and also that the skyrmion AHE contribution is dependent on the size of the skyrmions e.g. Zeissler et al. Nat. Nano. 13, 1161 (2018) and Maccariello et al. Nat. Nano 13, 233 (2018). The authors have discussed the dependence of their measured ρ_{xy} on the density of their ASKs, but there is little mention of the ASK size and how its changes may be relevant. However, it is clear in Fig 3a that the ASK size is changing with thickness, as well as the density. It could be expected then that there is a contribution from the AHE of the ASKs to the total measured ρ_{xy} signal. Can the authors comment on the possible contributions to the thickness-dependent ρ_{xy} they measure? In particular, is there an AHE component being also picked up in the ρ_{xy} , and how significant is it? Could it relate to the large-thickness saturation of the ρ_{xy} signal above ~ 2.4 microns, as in Fig. 3c?

Minor corrections:

Abstract: “its tunability by means of finite size(s), field orientation...” It would be helpful in the abstract to say specifically what the finite size relates to, i.e. the lamella thickness, to distinguish it from the other possibilities that the reader might assume this to mean, such as the ASK size.

Results and Discussion:

Fig 1.a: the V_{xy} is only measured in the presence of a current/electric field (which the authors mention in the text), but there is no current or electric field marked in the figure. Please add this.

Page 4: when the supplementary notes S1-S3 are mentioned, it would be helpful to note in the text that the supplementary notes have further details and images of the samples shown in Figs 1a and 1b including also the other thickness samples that are not shown here, but which have been measured and discussed in the text.

Page 4: can the authors include an equation for the topological Hall effect showing its dependence on the topological charge and other values, for instance similar to equation 3 of Zeissler et al. Nat. Nano. 13, 1161 (2018) ? A suitable equation would link to and make clear the relevance of the fitted dependence of the THE in Fig 3.c.

Page 5: the reference to the magnetisation data being Supplementary Note S5 is incorrect – it should be Supplementary Note S4.

Page 5/supp. Info 5. Here it is explained that there is a correction to the field based on the distance of the sample to the magnet. Has this correction been made for the data in the main text (Figs 1c and 1d)? It would be useful to mention this for readers comparing the supp info data with the main text, since the features of Fig S7 appear at different fields to where they do in Fig 1.

Formatting typos: In-situ and ex-situ should be in italics, and several reference URLs are corrupted or missing, e.g. refs 51, 52, 45

Supplementary info:

Supp. Note 5 refers to Fig 1E, which does not exist – I think this should be Fig 1d.

Fig S9 – the caption refers to a “field range between 1.6 and 2.6”. Can units be added to these numbers?

Supp. Note 7 mentions extracting the THE by first determining the orbital and anomalous components $p_{xy}AHE$ and $p_{xy}THE$, but based on what is written next about ‘the former’ and the ‘latter’ I think these should be $p_{xy}OHE$ and $p_{xy}AHE$, respectively. Also, the text refers to an eq. 1 in the main text which is not there. Near the bottom Figs S12a and S12b are mentioned, but these should be Figs S13a and b.

Fig S11 – should the y axis values for parts c. and d. be negative? Otherwise subtracting the linear slope shown results in positive ρ_{xy} values for parts e and f.

Supp. Note 8 – the reference to Fig 3 should instead say Fig 4d.

Dear Reviewers,

We appreciate your time and efforts spend on reviewing our manuscript. We revised the main text and figures, added additional data and references in order to take the suggestions and comments into account. Changes in the manuscript are highlighted by red color. The revised manuscript consists of 24 pages and 4 main figures. A short video sequence highlights the key experimental aspect of this work. We also provide Supplementary Information consisting of 22 pages including 16 figures.

In the following, we provide a point-by-point response to the referee reports.

Reviewer #1 (Remarks to the Author):

The manuscript „antiskyrmions and their electrical footprint in crystalline mesoscale structures“ by Winter et al. reports the study of the THE of antiskyrmions (ASKs) in MnPtSn and its tunability by device size, field, and temperature. The authors perform MOKE microscopy in combination with electrical measurements to provide evidence of the THE of ASKs in MnPtSn.

The paper is well written (though the supplementary material might require some proof reading) and reports an interesting result. While I find the topic very exciting, there are several points I would like the authors to address before I can recommend publication:

1) Page 2: The speed of antiskyrmions has been cited misleadingly, incl. several publications on antiferromagnetic (AFM) skyrmions and calculations that use ultra-low damping in the range of YIG. A distinction to AFM skyrmions and concrete examples should be included.

We thank the reviewer for this comment. We realized that the citations are misleading and have, therefore, removed the mentioning of antiferromagnetic skyrmions. Even though the ASKs in Mn_{1.4}PtSn have proven higher intrinsic stability to much larger sample thicknesses there are no clear reports of potential speed and motion properties yet.

2) Page 4: The authors report the simulations to “inevitably” showing AKS. There are also reports of skyrmions in MnPtSn. The statement should be made clearer.

The magnetism in Mn_{1.4}PtSn is rather complex it is becoming even more so below T_{SR} , where non-coplanar magnetism sets in. Even though there are reports of skyrmions at low temperatures, our simulations mainly capture the natural connection of the high-temperature magnetism to the emergence of antiskyrmions (ASKs). Likely, the reviewer is referring to the LTEM study [Jena et al., Nat. Commun. 11, 1115 (2020)] that was able to stabilize Bloch-type skyrmions at lower temperatures. As we were not able to obtain a visual imaging of the magnetic textures at low temperatures this goes beyond the scope of the present work.

We, therefore, have changed the sentence in the introduction to: “In contrast to the general approach of micromagnetic simulations we apply atomistic spin dynamic calculations that capture the underlying complex magnetism of Mn_{1.4}PtSn, inevitably linked to the emergence of ASKs at high temperatures.”

3) Page 5, line 2-3: “due to a change in the magnetic texture” is very generic and expected. Try to rephrase or omit.

The sentence has been changed for further clarity “For $d < 8.4 \mu\text{m}$, a noticeable hysteresis appears in the Hall effect before saturation, which details a noncoplanar magnetic and ASK spin texture when the field is parallel to the c axis.”

4) Page 5: That the sample stays in a saturated state while the field is decreasing has been seen in many skyrmion materials (there is also a minor typo in ref. 27) and is to be expected also without the anisotropic exchange.

The typo in the reference has been fixed. The authors agree with the reviewer that with decreasing field the sample stays in a saturated state in many skyrmion systems. The novel feature we see here, is considerably distinct from many systems such as the B20 compounds (isotropic). We show that the hysteresis only can be detected for samples below a certain thickness. This is probably closely related to the emergence of the fractal domains [Zuiniga et al., PRB 103, 184411 (2021)] observed in bulk samples that lead to a more inhomogeneous magnetic distribution and hence the absence of a clear ASK lattice.

5) Why is there no contribution of the THE visible while there are clearly skyrmion pockets forming before the reported onset of the THE? The supplementary material clearly shows these partial lattices.

A THE appears as soon as ASK pockets form. When compared to the maximum effective field induced by a fully crystallized ASK lattice, the individual pockets can have an effect that is 1000 times (or more) less. Nevertheless, we also expect an increase of the THE contribution as soon as individual ASKs are emerging. This, however, stays hidden in the noise.

6) If I get it correctly, the simulations were freely tuned to reproduce the experiment. The authors should use experimentally measured input parameters (as far as possible) and/or slightly tuned bulk parameters and check if the simulations still match the experiment.

The parameters were taken from first principle calculations used by Vir et al. [PRB 99, 140406(R) (2019)] and by Swekis et al. [ACS Applied Electronic Materials 3 (3), 1323-1333 (2021)]. In the Fig. R1 below, we show on the left the experimental T_C values of

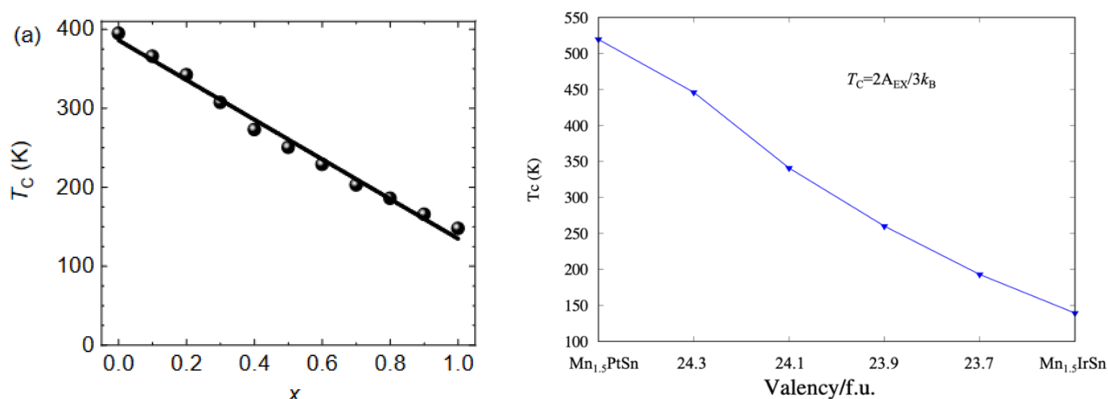


Figure R1 (a) Curie temperature of $\text{Mn}_{1.4}\text{Pt}_{1-x}\text{Ir}_x\text{Sn}$ (T_C) as function of Ir-concentration (x). (b) the first principle calculations of T_C using the exchange parameters [Vir et al. PhD Thesis 2020].

$Mn_{1.4}Pt_xIr_{1-x}Sn$ single crystals [Vir et al. PhD Thesis 2020] and on the right the first principle calculations of T_C using the exchange parameters.

Figure R2a shows the spin-spiral energy for the $Mn_{1.5}PtSn$ calculated from first principle with and without spin-orbit coupling. The spin spiral shows three stable minima depending on the spin-spiral wave vector. There are two minima away from $q = 0$ (i.e., collinear) due to the competition of exchange. When spin-orbit coupling is included, the spin-spiral and ASK size can be calculated, which shows to be approximately 200 nm in the FiM state (see right Fig. R2b). This is consistent with our experiment MOKE/MFM observations. Spin-dynamic calculations would be too expensive to calculate 200 nm size ASKs, which can be captured in micromagnetic simulations. However, micromagnetic simulations cannot capture the physics of the competition of the exchange parameters. Therefore, with spin-dynamic simulations we decrease the magnitudes of the exchange parameters, but keep the ratio the same to take into account the anisotropy effect seen in the hysteresis and the ASK phase. The experimental work shown in Fig. R1 demonstrates the accuracy of the exchange parameters obtained from first principle calculations.

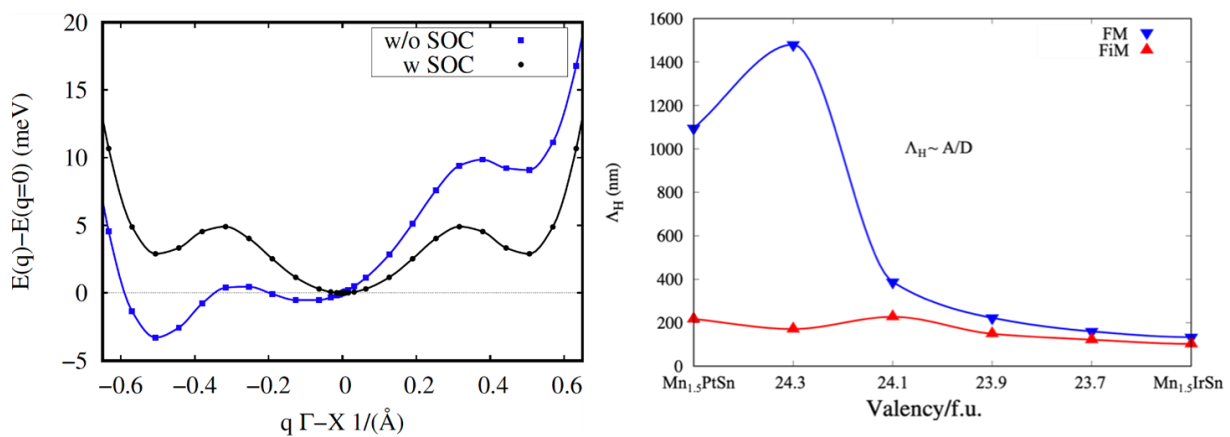


Figure R2: (a) Spin-spiral energy for the $Mn_{1.5}PtSn$ calculated from first principle calculations without and with spin-orbit coupling plotted versus spin-spiral wave vector. (b) Magnetic spiral periodicity, λ_H , which is proportional to the ratio of exchange field and DMI strength plotted against Mn valency from first principle calculation for the FM and FiM sublattices.

7) Page 6, line 2: is J1 ferromagnetic or weak antiferromagnetic?

The interaction J_1 is ferromagnetic. This statement has been added to line 2 page 6.

8) Page 6: “the yielded magnetization hysteresis... is congruent...”. The shown loop would look the same for skyrmions and thus does not contribute to the story, right?

The hysteresis loop shown can be the same for a skyrmion system. However, to the knowledge of the authors, there are no real skyrmion systems that show this behavior with the canting of the spins also giving rise to a topological Hall effect. This actually leads to one of the more interesting features of this story, because most skyrmion systems studied to date have very isotropic and for the most part simple exchange constants on the level of the two-spin Heisenberg interaction. Our system of interest has this “frustration” in the exchange constants, which can lead to multiple magnetic structures in the same system.

9) The authors state that the shallow slope in the Hall resistance right before the saturation is caused by the negative THE of the ASKs. However, while for skyrmions this would be easily visible as a positive contribution, the antiskyrmion contribution is similar to the one of shrinking magnetic textures. While the domains can easily contract, the topological stabilization of the ASKs will counteract this and lead to a longer presence and slower increase in the net magnetization, i.e. a different slope. These effects should be carefully and clearly discussed and disentangled in the main text.

This is a good point. We revisited our magnetization data collected for a 2.4 um thin relatively large lamella sample, used for device C later on. Indeed, we seem to observe a signature similar to what we find in the Hall effect. This confirms the presence of an anomalous Hall contribution due to the individual ASKs. We, therefore, have revised the main text and refer to the ASK Hall-resistivity component instead. Unfortunately, it is impossible to trace it reliably in order to determine the THE directly. This is mentioned in the revised discussion text as well.

The authors should provide direct experimental transport evidence that the signal is indeed a negative THE contribution, such as the transition into skyrmions by an in-plane field (10.1038/s41467-020-14925-6).

We thank the reviewer for this suggestion and are looking forward to such experiment being realized. In order to replicate the conditions in the mentioned LTEM study various challenges need to be overcome. Electron-transparent samples will be needed and the option to perform transport simultaneously while varying the field orientation inside an LTEM setup. The key difference to the field-angle protocol used in the LTEM experiments is that we do not need to apply any in-plane field in order to stabilize the ASK lattice. Most likely, dipole-dipole interactions are at the heart of the transformability of the magnetic textures in very thin samples. As our devices are much thicker, these interactions will be almost negligible. In our simulations, we demonstrate that for micron-sized samples the natural magnetic ground state and a field along the c axis are the most important prerequisites to stabilize ASKs in micron-sized samples. This perfectly matches with our experimental observations.

Reviewer #2 (Remarks to the Author):

In their work, Winter and co-authors report on the electrical footprint of antiskyrmions in crystalline mesoscale structures. This report, if accurate, would serve as a large advance in the field in topological magnetism. However, I have major concerns that preclude publication at this time, as detailed below.

From the experimental imaging, it is impossible to determine if the observed structures are really antiskyrmions. While antiskyrmions have been clearly observed in $\text{Mn}_{1.4}\text{PtSn}$ using L-TEM measurements, the techniques here give no information on the actual topological charge of the object, and the signal being seen observed can just as easily be explained by skyrmions, non-trivial bubbles, or even higher order excitations. All previous direct reports of antiskyrmions using imaging techniques have been through thin films (thicknesses on the order of 100 nm, significantly thinner than the system studied here). However, for thick films, neutron scattering measurements recently suggested that “bulk $\text{Mn}_{1.4}\text{PtSn}$ does not support antiskyrmions” (PHYSICAL REVIEW B 102, 174447 (2020)) and suggests dipole-dipole interactions need to be strongly considered (which is not done in the model used in this current work, as H_{demag} is ignored).

It is true that it is a challenge for MOKE and MFM techniques to resolve the in-plane components of the observed spot-like structures. However, a recent LTEM study demonstrated that ASKs in $\text{Mn}_{1.4}\text{PtSn}$ possess an extraordinarily strong stability to thicknesses of at least up to 250 nm [Saha et al., Nat Commun. 10,5305 (2019)]. We added this reference to the introduction. Unfortunately, this technique loses its sensitivity for larger thicknesses. The investigated samples are thin FIB-cut single-crystal slices. An observation of ASKs in thin films has not been possible to date.

Furthermore, these ASKs scale linearly with the thickness. Additional MFM studies have shown that this can be traced up to at least 1.5 microns. A recent X-ray study by Seki et al. [Nat. Mater. 21, 181 (2022)] visualized these skyrmions in the bulk of a FIB-cut wedge-shaped sample with varying thickness up to a few microns, see Fig. R3. The observed patterns match our MOKE imaging results and provide proof for the presence of skyrmions throughout the bulk in micron-thick samples. A recent MFM study of some of the authors [Zuniga et al., PRB 103, 184411(2021)] confirm our observation that a homogeneous magnetic phase can only be established for thicknesses of a few microns. As we argue below our simulations (different from micromagnetic simulations) further support the

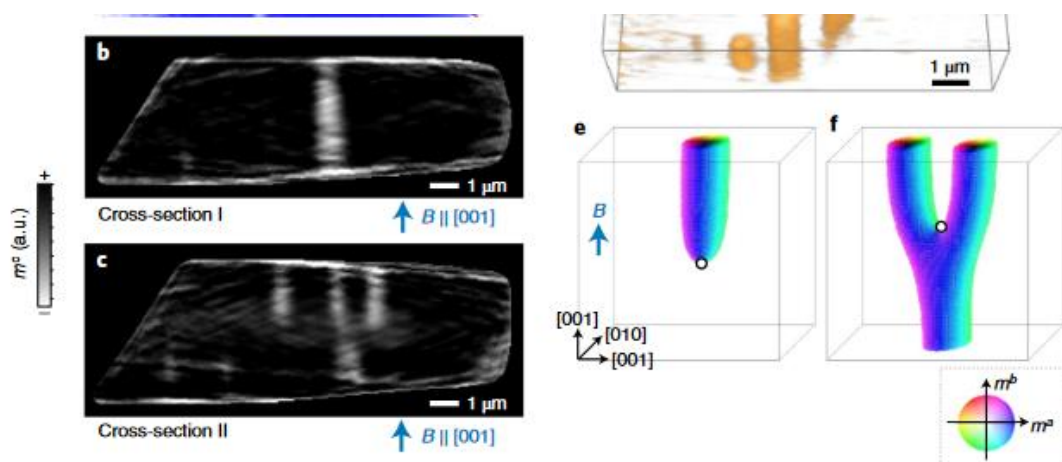


Figure R3: Direct visualization of Skyrmion strings in $\text{Mn}_{1.4}\text{PtSn}$ micron-sized crystals by X-ray tomography [Seki et al., Nat. Mater. 21, 181 (2022)]

establishment of ASKs naturally linked to the complex magnetic ground state of this compound. This all leads us to identifying the observed textures as ASKs.

*We agree that bulk samples do not exhibit clear signatures of ASKs. In the mentioned neutron study, Sukhanov et al. [PRB **102**, 174447 (2020)], reported for bulk crystals with several hundreds of microns if not millimeter sizes the presence of fractal domains without signatures of skyrmions throughout the bulk. However, the mentioned results on bulk samples and the thickness dependence of the domain periodicity of thin micron-sized samples investigated by means of MFM [Zuniga et al., PRB **103**, 184411(2021)] are perfectly in line with what we report. Namely, a critical thickness below which a semi hard-magnetic behavior establishes in field that is directly connected to the emergence of the ASKs.*

Of course, MFM can only identify surface structures. They are not able to distinguish between bubbles and skyrmions. We, however, show that the ASK signature in the Hall resistivity (which is a bulk phenomenon) appears once the samples are tailored below a certain thickness and can directly relate this appearance to the textures observable by MOKE and MFM.

The atomistic spin simulations do not seem to be very relevant to the system being studied. The sample dimensions (50 nm x 50 nm x (3 unit cells) in the vertical axis do not make sense to me. The system being studied has a much larger thickness, and the observed magnetic features from experiment are effectively two orders of magnitude larger than those observed in the results of atomistic spin simulations. While qualitatively producing antiskyrmions, the relevance to the system studied here is questionable, at best. Further, it doesn't appear that demagnetization energies are considered, which may be non-negligible for such thick, bulk-like systems.

*The atomistic simulations are extremely relevant because they include exchange mechanisms that have not been considered in previous works. The exchange interactions are derived from first-principle calculations as also seen in [Meshcheriakova et al., PRL **113**, 087203 (2014)]. Simple micromagnetic simulations cannot capture the subtleties in the exchange interaction. The experimental system being studied does have a much larger thickness, however, having a larger thickness for the simulations does not have an effect when demagnetization energies are not considered. As seen in previous works [Ma et al., Adv. Mater. **32**, 2002043 (2020)] the dipole-dipole interaction reduces its role as the film thickness d is increased as it has a $1/d$ dependence in first order. Furthermore, the dipole-dipole energy is shown in the same work to play a role of increasing the size of the ASKs where the exchange and DMI energies are generally significantly larger for these compounds and are clearly the dominating contributions. The model parameters are taken from first-principle calculations, which accurately reproduce the experimental results. In addition, it is clear from these results that the J_1 , J_2 , J_3 models used must be taken into account in order to explain experimental results across a wide range of similar compounds. The exchange parameters taken from first principles are reduced ("tuned") without the loss of generality allowing us more efficient calculations. The DMI is also increased to decrease the size of the required system (i.e., the ASK size decreases). While qualitatively, reproducing the ASKs, the main result of the model is the extraction of the hysteresis curve and the topological field, which explains the experimental results.*

In summary the dipole-dipole interaction increases the size of the ASKs linearly and is significantly smaller (1/100 times smaller) than the Heisenberg and DMI at a larger thickness (>0.5 micrometer). A model with more unit cells along the c direction would not change the results due to the symmetry of the DMI in the [Saha et al., Nat. Commun. **10**,

5305 (2019)], see Fig. 5 and discussion thereof. The model clearly explains the observed experimental behavior. Please see figures R1 and R2 in our response to reviewer 1 on first-principle calculations.

The authors should estimate the expected contribution to hall resistivity per anti-skyrmion. Due to the size of the objects, one would expect non-adiabatic contributions to be small. But if purely adiabatic contributions are present, I don't think this magnitude of effect is expected.

In the cases of MnSi and FeGe the THE of the skyrmion phases is seen on top of the linear slope of the anomalous Hall effect deep inside the chiral phase. It is, therefore, easily extractable by applying a simple linear fit [Sci. Rep. 8, 15510 (2018); PRL 102, 186602 (2013)]. In our case, the ASK phase only emerges right before or exactly at the turning point of the AHE into saturation. Hence, our subtraction method results in a very rough upper estimate of the Hall-effect contribution.

We agree that in reality, considering the huge spatial extend of the ASKs in Mn_{1.4}PtSn, a much smaller value would be expected. Our ρ_{xy}^{ASK} peaks are in the range of ~[30-60] nΩcm. Somewhat larger but of a similar magnitude as in MnSi and FeGe ~[1-10] nΩcm. In the case of FeGe, a much smaller THE was expected as the skyrmions are larger than in MnSi. One of the reasons why the two materials actually exhibit similar magnitudes in THE can be caused by the differences of the band structures. In the case of Mn_{1.4}PtSn, which is a good metal, many bands are close to the Fermi level and, hence, may contribute to the enhanced THE signature. We, furthermore, need to stress that the linear fit and its subtraction, in our case, leads to a sharp peak that does not resemble the theoretical prediction of a plateau-like THE versus field. Here, a subtraction of the magnetization would be of great help as the transition into the polarized state for sure will not be as discontinuous as we are assuming. Therefore, we can expect a much flatter dependence and, thus, an overall maximum value of the THE closer to what was reported for the other skyrmion materials.

If possible, the authors should show a switching loop as measured by i.e. SQUID and then compare the Hall measurements to the M-H curve. This is a common method to measure THE, i.e. as done in Nature Materials 16, 898–904 (2017) and Nature Communications 12, 2758 (2021).

It was impossible to measure the magnetization for a structured device, because the volume is simply too small. However, in Fig. 2 and Supplementary Note 4 we present a magnetization measurement for the 2.4 μm thick sample before it was structured into the Hall-bar device C. Apparently, the thickness of the huge FIB-cut slice was not perfectly homogeneous. As we show the sample establishes the chiral phase out of the field-polarized state at two different critical field values upon decreasing the magnetic field. With the help of MOKE we saw that the sample has two regions with different onset fields. The SQUID-VSM measurement revealed a double-step transition and a hysteresis similar to what we observe in the local transport signal. An additional shoulder-like feature is discernible that could account for the deviation we observe in the Hall resistivity. However, this feature is close to the noise level and cannot be used to extract the THE directly. We have revised the manuscript and now refer to the signature as ASK Hall resistivity component, which is comprised of an anomalous and topological component. The anomalous component may be the major fraction and, therefore, we can at least provide an upper limit for the magnitude of the actual THE.

Can the authors exclude effects arising from a non-uniform extraordinary Hall effect due to variations in local microstructure? If so, how do they exclude this effect?

We have good metallic, high-quality single crystals and, therefore, it is not clear to us how a sudden change appearing in a particular field range can be associated with a non-uniform extraordinary Hall effect. From our point of view, such a scenario can be ruled out.

The authors state “therefore, we tuned the classical parameters in atomistic spin-dynamic simulations to clearly show the effects seen in the experiments.” Am I to read this as that the values utilized in atomistic spin simulations were simply varied in order to get good agreement with experiment? Is there a reference to the values being used available from the literature?

Please see above our reply on exchange interactions (starting with “The atomistic simulations...”) and to point 6) of reviewer #1.

The measured domain widths and “antiskyrmion” widths are clearly more than 100 nm in extent and significantly larger than those reproduced in atomistic modeling. Further, as far as I’m aware, antiskyrmions as observed are typically sub-100 nm – again, I’m concerned these are more closely related to a bubble type configuration. At this size, there should be something done to verify the spin structure sketched in Fig. 1a.

We understand this concern. We would like to emphasize again that LTEM has proven an enhanced stability to at least 250 nm thicknesses and a linear scaling of the domain periodicity. This has also been confirmed by MFM studies to much larger thicknesses similar to our devices. Please see our reply above.

The fit in Fig. 3c is not very convincing, considering the uncertainty in the data points. Further, why are MFM measurements for 5 μm and 1.5 μm thick films shown, but not plotted in Fig. 3c, while the images where points in Fig. 3c were obtained are not found in the main text or supplemental. This would be quite useful.

We agree that this fit was rather acting as a guide to the eye. We performed an additional thickness dependence study on two separate devices. We etched the two devices in steps and measured transport after each thinning step for device 2.4 μm down to 0.7 μm , and from 4.1 μm down to 2.1 μm , respectively. The new figure 3c presents the wide spread of the data probed locally. A clear enhancement with reducing thickness is visible. This provides solid evidence for it being disconnected from the general anomalous Hall effect that is not expected to change upon periodicity changes of the domains.

Reviewer #3 (Remarks to the Author):

This manuscript reports a thorough study of magnetic textures, including antiskyrmions, in patterned hall bar structures of Mn_{1.4}PtSn single crystals using Hall measurements and MOKE imaging. The authors observe clear correlation between the detected topological Hall effect and the imaging of antiskyrmions, demonstrating the existence and phase diagram of antiskyrmions in Mn_{1.4}PtSn microstructures. They also perform atomistic simulations which explain the experimental results by the competition of fundamental interactions including exchange, DMI, and anisotropy. The experimental data are of high quality, which, together with atomistic simulations, convincingly reveal the behavior of antiskyrmions and the phase space of magnetic textures in Mn_{1.4}PtSn and the underlying mechanisms. I recommend its publication in Communications Materials. Below are a few minor comments for the authors to address in the revised manuscript.

We are very grateful for the recommendation of reviewer #3. Below we discuss the raised concerns and comments.

1. Figure 1c, especially its inset, should be compared with the magnetization hysteresis loop of the same sample in the main text. I suggest the authors move Figure S6a in the supplement to the main text, in Figure 1c. In addition, the inset of Figure S6a (magnetization hysteresis) shows a two-step switching in the down sweep direction, while the inset of Figure 1c (Hall resistivity) shows a one-step switching for the same sample. What is the reason for this inconsistency?

This is a good point. We, however, would like to emphasize that the small volume of the actual transport devices prevents direct magnetization measurements and the suggested analyzes. The two-step transition in the magnetization of the 2.4 μm thin sample was recorded for the unstructured lamella shaped sample with a size of $(200 \times 60 \times 2.4) \mu\text{m}^3$. This volume provided enough material for a SQUID magnetization measurement. In supplementary Note 4 we present MOKE images for that particular sample that show the two distinct regions in the sample with different onset fields for the transition into the spin-spiral phase when the field is reduced starting from the saturated high-field state. The Hall effect is probed locally and, hence, the double-step is somewhat weaker but detectable for the transport device that was structured into the large lamella sample, see Fig. 2g.

2. On page 5 of the main text, line 10 from the top, it refers to "as we show in Supplementary Note S5". Should it be either "Supplementary Note S4" or "Supplementary Note Figure S6a"?

Thank you for this comment. We revised the text accordingly.

3. The MOKE images in Figure 1d of the main text seem to be for the same sample used for the MOKE images in Figure S7 of the supplement. However, the field values for the appearance of similar features are different between the two sets of MOKE images. Can the authors comment on this discrepancy?

The field values were corrected for the reduction of the actual field at the sample due to the separation by the substrate from the pole of the electromagnet. In the supplement, the original set values are shown, as they were recorded during the MOKE experiment. We added a comment to remind the reader of this experimental detail. However, in the new version of the manuscript we refer to this set of images in the supplement. We now show

the images recorded back-to-back with the Hall effect for a 0.8 micron thin device, the same as is presented in the video sequence. We think this way things are much clearer.

Reviewer #4 (Remarks to the Author):

In this manuscript, the authors have built on a significant back-catalogue of research into high quality crystals of tetragonal Heusler alloy $\text{Mn}_{1.4}\text{PtSn}$ to demonstrate and discuss the electrical signatures of antiskyrmions. The complex magnetic interactions that result in these magnetic textures are of great and ongoing research interest, and electrical detection is one of the key challenges to overcome for developing computing technologies that rely on such textures. Antiskyrmions (ASKs) are a more recent addition to the zoo of magnetic quasiparticles being investigated for these reasons, and the current manuscript is a thorough, well-constructed and original study that will be of immediate interest to, and will benefit, the spintronics and magnetic materials research community. This is the one of very few works that report on the electrical detection of ASKs, and the correlation of MOKE, magnetisation and magnetotransport measurements on carefully microstructured single-crystal devices, with support from both FMR measurements and atomistic simulations, is very convincing.

There are no significant flaws in this paper, and the data and methodology are very well presented. The references, abstract, introduction and conclusions are all appropriate. I am happy to recommend this article be accepted in Communications Materials, however I have one or two questions for the authors below, as well as several suggested minor corrections and edits.

We thank the reviewer for the supporting summary. We would like to express our gratitude for pointing us towards the references below. These helped us to revise our manuscript and improve on the overall message. We realized that indeed the THE should be very small, smaller than initially anticipated. Nevertheless, this has no effect on the main outcomes of our work presented here.

It has been shown before, in multilayer films containing skyrmions, that ρ_{xy} is made up of both AHE and THE contributions from the skyrmions, and also that the skyrmion AHE contribution is dependent on the size of the skyrmions e.g. Zeissler et al. Nat. Nano. 13, 1161 (2018) and Maccariello et al. Nat. Nano 13, 233 (2018). The authors have discussed the dependence of their measured ρ_{xy} on the density of their ASKs, but there is little mention of the ASK size and how its changes may be relevant. However, it is clear in Fig 3a that the ASK size is changing with thickness, as well as the density. It could be expected then that there is a contribution from the AHE of the ASKs to the total measured ρ_{xy} signal. Can the authors comment on the possible contributions to the thickness-dependent ρ_{xy} they measure? In particular, is there an AHE component being also picked up in the ρ_{xy} , and how significant is it? Could it relate to the large-thickness saturation of the ρ_{xy} signal above ~ 2.4 microns, as in Fig. 3c?

We have added the two suggested references into the discussion, where we further revised the text according to the comment of the reviewer. We agree that there should be a significant contribution of Hall due to an AHE induced by the presence of ASKs. We now show in Fig. 2 that we indeed are able to see indications of a shoulder feature in the magnetization measured for the relatively large sample with thickness 2.4 μm . However, the situation in $\text{Mn}_{1.4}\text{PtSn}$ might not be as straight forward as it was demonstrated in the mentioned works on Co thin films. It remains an open question if ASKs may also be

created or annihilated by electrical field pulses. Hence, studying the contributions of a single ASK goes beyond of this work. In the case of an AHE of the individual ASKs, we agree that a change in thickness, that changes the magnetic periodicity, and in turn the ASK diameter, would also affect the magnitude of the AHE component. We now mention this in the discussion of the thickness dependence. We removed the quadratic fit and only mention that for a linear increase of the periodicity a strong enhancement of the THE component can be expected. In this work however, we are not able to distinguish these effects in detail and can at least argue to provide an upper limit of the overall magnitudes.

Minor corrections:

Abstract: "its tunability by means of finite size(s), field orientation..." It would be helpful in the abstract to say specifically what the finite size relates to, i.e. the lamella thickness, to distinguish it from the other possibilities that the reader might assume this to mean, such as the ASK size.

The abstract has been modified to read "...finite device thickness..."

Results and Discussion:

Fig 1.a: the V_{xy} is only measured in the presence of a current/electric field (which the authors mention in the text), but there is no current or electric field marked in the figure. Please add this.

We now also mark the current with capital "I".

Page 4: when the supplementary notes S1-S3 are mentioned, it would be helpful to note in the text that the supplementary notes have further details and images of the samples shown in Figs 1a and 1b including also the other thickness samples that are not shown here, but which have been measured and discussed in the text.

We have added a comment to the figure caption as suggested.

Page 4: can the authors include an equation for the topological Hall effect showing its dependence on the topological charge and other values, for instance similar to equation 3 of Zeissler et al. Nat. Nano. 13, 1161 (2018) ? A suitable equation would link to and make clear the relevance of the fitted dependence of the THE in Fig 3.c.

As we have revised the parts of the manuscript concerned with the ASK Hall resistivity component we feel the theoretical descriptions are more comprehensive in the resubmitted version to support our findings. The mentioned works, however, refer to a theory that may need significant modifications in order to be directly applied to $Mn_{1.4}PtSn$ as its magnetism is far more complex. We, however, refer to these works and included a discussion of the possible anomalous component of the observed ASK Hall-resistivity signature.

Page 5: the reference to the magnetisation data being Supplementary Note S5 is incorrect – it should be Supplementary Note S4.

We have corrected this typo

Page 5/supp. Info 5. Here it is explained that there is a correction to the field based on the distance of the sample to the magnet. Has this correction been made for the data in the

main text (Figs 1c and 1d)? It would be useful to mention this for readers comparing the supp info data with the main text, since the features of Fig S7 appear at different fields to where they do in Fig 1.

We have revised Fig. 1. We now show example images recorded for device F with $d = 0.8 \mu\text{m}$ that were simultaneously recorded with Hall resistivity in one and the same setup.

Formatting typos: In-situ and ex-situ should be in italics, and several reference URLs are corrupted or missing, e.g. refs 51, 52, 45

We have corrected these typos

Supplementary info:

Supp. Note 5 refers to Fig 1E, which does not exist – I think this should be Fig 1d.

This sentence was revised.

Fig S9 – the caption refers to a “field range between 1.6 and 2.6”. Can units be added to these numbers?

We have added units to the values.

Supp. Note 7 mentions extracting the THE by first determining the orbital and anomalous components ρ_{xy}^{AHE} and ρ_{xy}^{THE} , but based on what is written next about ‘the former’ and the ‘latter’ I think these should be ρ_{xy}^{OHE} and ρ_{xy}^{AHE} , respectively. Also, the text refers to an eq. 1 in the main text which is not there. Near the bottom Figs S12a and S12b are mentioned, but these should be Figs S13a and b.

We have corrected these typos.

Fig S11 – should the y axis values for parts c. and d. be negative? Otherwise subtracting the linear slope shown results in positive ρ_{xy} values for parts e and f.

We corrected the sign in Fig. S11c and d.

Supp. Note 8 – the reference to Fig 3 should instead say Fig 4d.

We have corrected this typo.

7th Jul 22

Dear Dr Helm,

Thank you for submitting your revised manuscript, "Antiskyrmions and their electrical footprint in crystalline mesoscale structures.", to Communications Materials. It has now been seen again by Reviewer 1, 2 and 4, whose comments are appended below. You will see that they have raised further further points that must be addressed. We remain interested in the possibility of publishing your study in Communications Materials, but would like to consider your response to these concerns in the form of a revised manuscript before we make a decision on publication.

In particular, you will see that both Reviewers 1 and 2 are still raising doubts regarding a clear disentanglement of THE from the AHE, and how each effect is contributing to the ASK texture and its detection. We believe that further discussion of these points, avoiding an overstating of claims and making clear the open challenges, is needed to meet the level of scientific rigour that we are seeking in our journal. We would naturally expect all other comments of the referees to be appropriately responded to, including their requests for further information and discussion.

We therefore invite you to revise and resubmit your manuscript, taking into account the points raised.

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

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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](https://figshare.com/) or [Dryad Digital Repository](http://datadryad.org/). 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.

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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.

We understand that due to the current global situation, the time required for revision may be longer than usual. We would appreciate it if you could keep us informed about an estimated timescale for resubmission, to facilitate our planning. Of course, if you are unable to estimate, we are happy to accommodate necessary extensions nevertheless.

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,

Alannah Hallas, PhD
Editorial Board Member
Communications Materials
orcid.org/0000-0002-0892-0982

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The authors addressed several of my points satisfactorily, however, I still have some major concerns to raise.

Following up on my previous comment 5: I assume it is a much easier explanation that due to the size of the magnetic textures a possible existence of the THE would be too small to see. The authors use this explanation later. If the THE signal lies within the noise I fail to see the presentation of an unambiguous signal specific for ASK structures.

In the methods section the authors explicitly state the objective of their paper to be to “disentangle other contributions to the electrical-transport signature related to the complex magnetism” of their material and to “study the topological Hall signature” as function of various parameters. In the abstract they imply to provide unambiguous signatures of ASK. Such a study would clearly be of major interest to the field. Unfortunately, I have to say that these goals were not achieved. The authors have added extensive explanations of the contribution of the AHE to the observed transport signal, though could not provide a suitable means to disentangle the THE from the AHE. This obviously is difficult but a crucial requirement to actually observe unique ASK signatures. Without this step, any magnetic texture such as skyrmions of varying chirality and even pinned domains should be able to generate a similar Hall response with a change in slope that immediately precedes saturation in an applied magnetic field.

One option to separate the effects of THE and AHE could be to utilize MOKE or MFM loops as function of field to extract the magnetization curve. This should be a crucial step to actually show ASK Hall signatures.

Some minor points:

Following up on my previous comment 4: that the hysteresis only appears for systems below a

certain thickness makes sense since the material is now more affected by the dipolar interaction. However, even in B20 lamellae (10.1103/PhysRevResearch.3.013094) a hysteresis has been seen. I also am a little confused by the author's statement (previous comment 9, second paragraph) that dipolar interactions will not play any role in their samples (also see reviewer 2's previous comment #1). When looking at the paper Ma et al. the authors cited in their response to referee 2 it clearly says "for the range of t of interest here the dipole energy strongly influences λ ". This range corresponds to up to at least $\sim 4\mu\text{m}$ and thus indeed affects the investigated devices?

Page 3: "a huge topological Hall effect" – I assume there will also be quite a lot of AHE present and the two effects will be even more difficult to disentangle. It is not clear to me that this statement can be made here.

Page 5: the extended ASKs in the author's sample appear more bubble-like with a non-zero magnetization, in line with the presented hysteresis data. The statement of ASK having zero magnetization in this context is misleading.

Page 7: I assume the panel "(see Fig. 2c)" is incorrect

Page 7: only a minor wording issue – "only exists in the up sweep" is a bit unprecise. A "finite qT extends to much higher fields in the up sweep as compared to the reoccurrence of topological charge when seeping down" or something similar might better convey the message.

Page 7,8: "Hall effect due to ASK directly" and "Thickness dependent ASK Hall effect" – again, the AHE is here not specifically a signature for ASK.

Page 8: "the THE is expected to grow quadratically with decreasing d " – ok, but again a negative result. This is not in the data. The data is consistent with a slight variation of an AHE of a constant packing density of magnetic textures in 2D

Page 14: The orbital and anomalous components are labeled "AHE" and "THE" (not OHE and AHE).

All in all, at present I do not see that the manuscript fulfills the journals' requirement of "significant advances for a specialized area of research", and especially not the promise of an unambiguous detection mechanism of ASKs. The way this is repeatedly phrased throughout the manuscript is misleading. The authors should try to use MFM or MOKE hysteresis data to properly isolate the THE from the contribution of the AHE. The results of having an AHE caused by large ASK or skyrmion textures and the conclusion that the THE is small are both expected, as the authors state themselves. The imaging remains interesting but the focus of the work lies on the Hall measurements and the effects below T_{sr} are not explained or further investigated and do not really seem to form a core element of the paper. The presented observations would not fulfill the author's claim to offer "a route to room-temperature skyrmionic applications based on multiple distinct types of skyrmionic textures". Given the difficulty to disentangle the different Hall signals the authors could also think about making the explanation of the behavior when crossing the T_{sr} a major focus of the paper but in its current form I cannot recommend publication in Nature Communications Materials.

Reviewer #2 (Remarks to the Author):

See attached

Reviewer #4 (Remarks to the Author):

The authors have responded to my questions and comments, and combined with comments from

the other reviewers, they have modified the paper in a thoughtful way. I am happy that the authors have now taken into careful consideration the contribution of an anomalous Hall component in the total measured transverse voltage signal, and this has meant they are much more circumspect when referring to a possible topological Hall contribution, which I think is highly appropriate. I am happy to recommend this manuscript be accepted, with only some minor points below I wish to make.

There are still some instances where the Supplementary Note number cited in the text does not match where the actual information is. Some of the ones I noted as minor corrections in my previous review have not, in fact, been corrected. For instance, in my previous review, I noted the following minor corrections:

Supp. Note 7 mentions extracting the THE by first determining the orbital and anomalous components ρ_{xy}^{AHE} and ρ_{xy}^{THE} , but based on what is written next about 'the former' and the 'latter' I think these should be ρ_{xy}^{OHE} and ρ_{xy}^{AHE} , respectively. Also, the text refers to an eq. 1 in the main text which is not there. Near the bottom Figs S12a and S12b are mentioned, but these should be Figs S13a and b.

The response letter noted 'We have corrected these typos.', however the mention of subtracting ρ_{xy}^{AHE} and ρ_{xy}^{THE} to find the THE (it should be ρ_{xy}^{OHE} and ρ_{xy}^{AHE} that are subtracted) is still there. The other two corrections have been made.

Similarly, for another minor correction I recommended:

Fig S11 – should the y axis values for parts c. and d. be negative? Otherwise subtracting the linear slope shown results in positive ρ_{xy} values for parts e and f.

The response states 'We corrected the sign in Fig. S11c and d.', however while the text of the caption has changed slightly, the figures are identical to how they were before revision, with no sign change.

Some examples where the cited Supplementary Note is wrong:

Page 6: "...the magnetization, as we show in Supplementary Note S6." Supplementary Note S6 provides details of the atomistic spin-dynamic simulations and it is Supplementary Note S4 that shows the magnetisation.

Page 6: "In Supplementary Notes S4 and S5, we present an additional sequence of MOKE images recorded for device C..."

Note S4 is the magnetisation and shows no MOKE images – these are shown only in S5.

I do not consider these continuing typos to be enough to REQUIRE changes be made, however I strongly urge the authors to check carefully all of their references to Supplementary Notes and Figures before publication to ensure they are correct. I suspect the authors have sometimes used the number for a Supplementary Figure instead of a Supplementary Note.

The authors have provided a detailed response to my concerns. I am still uncertain these measurements are a clear THE signal, but am satisfied with the clarification that this refers to an ASK Hall resistivity possibly containing an anomalous component.

There are a few minor points I would like to see addressed prior to acceptance, however.

- 1.) It would be instructive to calculate the expected antiskyrmion density from the Hall measurements if this were a purely THE effect and compare that to the measured density (i.e. using data similar to the supplementary video), i.e.

$$\rho_{THE} = -P \cdot R'_0 \cdot (n_{ASK} \cdot \Phi_0)$$

where P is the spin polarization of the charge carriers, R'_0 is the Hall coefficient representing the effective charge density contributing to the THE (and is normally assumed to be equal to the classical Hall coefficient), and $n_{ASK} \cdot \Phi_0$ is the magnitude of the emergent field associated with a given antiskyrmions density. n_{ASK} is the antiskyrmion density and $\Phi_0 = h/e$ (see Phys. Rev. Lett. 102, 186602 (2009) or Nat. Comm. 12, 2758 (2021)).

I do think that it is important to show the theoretical estimate for antiskyrmion density from transport measurements to experimentally measured antiskyrmion density.

- 2.) In the supplementary video, I'm assuming the units should be mT, not T.

Response letter

We would like to thank the reviewers for their constructive comments. In the following, we provide a point-by-point response to the raised questions and remarks. Changes in the main text are highlighted by red color.

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The authors addressed several of my points satisfactorily; however, I still have some major concerns to raise.

Following up on my previous comment 5: I assume it is a much easier explanation that due to the size of the magnetic textures a possible existence of the THE would be too small to see. The authors use this explanation later. If the THE signal lies within the noise I fail to see the presentation of an unambiguous signal specific for ASK structures.

In order to help the reader we changed the sentence "...observed feature is hardly distinguishable from the noise background." To: "...observed feature in the magnetization is hardly distinguishable from the noise background."

Again, we demonstrate a route to quantitative detection of the topological Hall effect. Our observations provide an upper limit for the observable effect. For the first time, this has become possible in such systems. Thanks to the scalability the signal should become stronger the lower the thickness and hence the denser the ASK phase.

In the methods section the authors explicitly state the objective of their paper to be to "disentangle other contributions to the electrical-transport signature related to the complex magnetism" of their material and to "study the topological Hall signature" as function of various parameters.

This was a left over from the previous version. We toned it down in the supplement as well. We however still show how to disentangle different Hall contributions. In particular, we show the differences in the AHE components associated with the chiral phase; how the previously reported large topological component can be observed for a particular field orientation; and that it has nothing to do with the actual ASK. The latter point resolves the longstanding riddle of where and how to look for the electronic signature of the ASK phase and why it may have been overlooked to date. Our thickness study further explains the absence of transport signatures in bulk samples.

In the abstract they imply to provide unambiguous signatures of ASK. Such a study would clearly be of major interest to the field. Unfortunately, I have to say that these goals were not achieved. The authors have added extensive explanations of the contribution of the AHE to the observed transport signal, though could not provide a suitable means to disentangle the THE from the AHE. This obviously is difficult but a crucial requirement to actually observe unique ASK signatures. Without this step, any magnetic texture such as skyrmions of varying chirality and

even pinned domains should be able to generate a similar Hall response with a change in slope that immediately precedes saturation in an applied magnetic field.

We modified the abstract in our previous revision in order to lower our claims. We think that there is no overselling here. We do not claim to have found the unambiguous topological Hall signal of ASK. We however, show what is possible at the mesoscale for this compound down to 500 nm. We present unambiguously how the Hall signature emerges in the well-known ASK phase of this exciting and complex compound.

We however, agree that the title may promise a too general result. We now added "... of $\text{Mn}_{1.4}\text{PtSn}$ " to the title. We believe this will help the readers to categorize our work better before reading it. Nevertheless, our approach and observations will be highly beneficial to research on skyrmion compounds in general.

One option to separate the effects of THE and AHE could be to utilize MOKE or MFM loops as function of field to extract the magnetization curve. This should be a crucial step to actually show ASK Hall signatures.

Thank you for this suggestion. This is actually a very challenging endeavor that would go beyond the scope of our work. We agree that a direct detection of the magnetization may be helpful for the subtraction approach. Thanks to our MFM and MOKE experiments we were able to watch while the ASK phase establishes within a narrow field range and measure transport simultaneously. The referee seems to imply that these experiments may be not innovative. Our obtained resolution in both MFM and MOKE experiments are state of the art. Detailed magnetization loops with the useful resolution were not possible in our setups so far. We nevertheless provide a direct electrical measurement of the Hall response in the ASK phase. This will eventually help combined transport and LTEM experiments to identify the THE component quantitatively at thicknesses where the ASK density and size are suitable enough in order to induce a strong emerging field to the electrons.

Some minor points:

Following up on my previous comment 4: that the hysteresis only appears for systems below a certain thickness makes sense since the material is now more affected by the dipolar interaction. However, even in B20 lamellae (10.1103/PhysRevResearch.3.013094) a hysteresis has been seen. I also am a little confused by the author's statement (previous comment 9, second paragraph) that dipolar interactions will not play any role in their samples (also see reviewer 2's previous comment #1). When looking at the paper Ma et al. the authors cited in their response to referee 2 it clearly says "for the range of t of interest here the dipole energy strongly influences λ ". This range corresponds to up to at least $\sim 4\mu\text{m}$ and thus indeed affects the investigated devices?

We fully agree with the referee. Of course, there are dipole-dipole interactions involved to a certain extent. However, it is not the same simple FM magnetic background as in a B20 compound. Hence, dipole-dipole are not able to cause ASK alone or even cannot be the major driving interaction here.

We provide a theoretical model that shows the direct emergence of ASK from the magnetic state in our compound. Our thickness-dependent Hall study demonstrates the emergence of the hysteresis below a certain thickness, which may be associated with a more coherent single-domain state as in the bulk. It furthermore is an indication for an enhanced contribution of dipole-dipole interactions at the mesoscale. Our simulations certainly do not reproduce all experimentally observed features. For example the linear alignments of the band domains may be related to additional components such as the demagnetization fields originating from the sample geometry or the enhanced dipole-dipole interactions for thin samples.

Page 3: “a huge topological Hall effect” – I assume there will also be quite a lot of AHE present and the two effects will be even more difficult to disentangle. It is not clear to me that this statement can be made here.

In principle, the AHE is considered of topological origin too; see the work by Nagaosa, Rev. Mod. Phys. 82, 1539 (2010). The main difference is the real-space variation of the magnetization that is described in terms of momentum Berry curvature. Nevertheless, in the case of the non-coplanar magnetism induced by the in-plane field orientation and the reorientation transition in $\text{Mn}_{1.4}\text{PtSn}$, it was shown for bulk samples, that a huge additional Hall component emerges that cannot be ascribed purely to AHE.

Page 5: the extended ASKs in the author’s sample appear more bubble-like with a non-zero magnetization, in line with the presented hysteresis data. The statement of ASK having zero magnetization in this context is misleading.

The referees critique here is unclear. Only by theoretical definition skyrmions can be mapped to a $\text{SU}(2)$ sphere with all spins summing to zero and only in the cases of $N=\pm 1$ cases. However, the presence of a magnetic field extends magnetization in the core and/or the background region of the magnetic texture—thereby displaying bubble-like features. However, it is well known that transport features are dominated by the magnetic texture variation across the entire texture not purely the core or the background. Of course, all MOKE results showing Skyrmions, Antiskyrmions, topological or non-topological bubbles would have the visual appearance of a point-shaped object with this technique. As we describe in the introduction, X-ray tomography has proven the existence of Skyrmions extending through the bulk of micron-sized crystals. The overall patterns observable at the surface perfectly match those observed in our MOKE experiments. The drop in the overall Hall curve is indeed consistent with a drop in the AHE component. Hence, it is consistent with a reduced overall magnetization.

Page 7: I assume the panel “(see Fig. 2c)” is incorrect

Thank you! We have corrected (see Fig. 2d).

Page 7: only a minor wording issue – “only exists in the up sweep” is a bit unprecise. A “finite qT extends to much higher fields in the up sweep as compared to the reoccurrence of topological charge when sweeping down” or something similar might better convey the message.

We have revised the sentence into:

“In the field-up sweep, q_T in the skyrmion phase exhibits an opposite sign as compared to the net charge in the low-field spin-spiral phase and extends to higher fields as compared to the reoccurring q_T in the down sweep (orange curve).”

Page 7,8: “Hall effect due to ASK directly” and “Thickness dependent ASK Hall effect” – again, the AHE is here not specifically a signature for ASK.

The first mentioning refers to the theoretical calculations. Here we compare the calculated up and down sweeps with each other. In this case, the ASK Hall signature results from the difference between up and down sweep.

The latter mentioning refers to the Hall signature that emerges once the ASK phase establishes. The existence of ASKs has been proven in previous works. We have revised the section title into “Thickness and temperature dependent Hall effect in the ASK phase”. We agree that we hereby cannot unambiguously determine its exact origin. We however reveal this distinct signature once the ASK phase emerges. That, no one else has been able to resolve to date.

Page 8: “the THE is expected to grow quadratically with decreasing d ” – ok, but again a negative result. This is not in the data. The data is consistent with a slight variation of an AHE of a constant packing density of magnetic textures in 2D

We agree. Nevertheless, we do not see a problem in the way we present our results. We demonstrate the thickness dependence and show that the overall topological component remains too small. Nevertheless, it provides the pathway forward to disentangle these components unambiguously if one would eventually be able to achieve nanometer thicknesses (an experiment that is currently underway by other groups, initiated by our observations)

Page 14: The orbital and anomalous components are labeled “AHE” and “THE” (not OHE and AHE).

Thank you. We have corrected these typos in the Supplementary Information.

All in all, at present I do not see that the manuscript fulfills the journals’ requirement of “significant advances for a specialized area of research”, and especially not the promise of an unambiguous detection mechanism of ASKs. The way this is repeatedly phrased throughout the manuscript is misleading. The authors should try to use MFM or MOKE hysteresis data to properly isolate the THE from the contribution of the AHE. The results of having an AHE caused by large ASK or skyrmion textures and the conclusion that the THE is small are both expected, as the authors state themselves. The imaging remains interesting but the focus of the work lies on the Hall measurements and the effects below T_{sr} are not explained or further investigated and do not really seem to form a core element of the paper. The presented observations would not fulfill the author’s claim to offer “a route to room-temperature skyrmionic applications based on multiple distinct types of skyrmionic textures”. Given the difficulty to disentangle the different Hall signals the authors could also think about making the explanation of the behavior when

crossing the T_{SR} a major focus of the paper but in its current form I cannot recommend publication in Nature Communications Materials.

We strongly disagree with this assessment and hope that our responses above may have helped the referee to change some of her/his opinions. Our observation of a Hall signature that can be addressed to the emergence of the ASK phase in this compound is a significant step forward. Our simulations provide the first direct simulation of the process taking into account the complex magnetism. This has not been captured by any of the micromagnetic simulations in the field so far. We solve the discrepancy (that has been around for a while) between experiments performed on either bulk or very thin electron-transparent samples. The ASK phase seems to establish only at the mesoscale in these materials. We agree that this only can be achieved by a significant contribution of dipole-dipole interactions. We provide a thickness dependence and show how the hard-magnetic behavior arises. We show that both MFM and MOKE can resolve the changes at the mesoscale. We combined MOKE with transport in mesoscale structure at various thicknesses. We conclude that we still would need to scale down thicknesses further in order to measure the THE of the ASKs quantitatively. This step is left for future experiments. Our work adds the temperature dependence. We show how the Hall feature disappears below T_{SR} and provide evidence for a significant transformation of the magnetic ground state and its anisotropy.

Reviewer #2

The authors have provided a detailed response to my concerns. I am still uncertain these measurements

are a clear THE signal, but am satisfied with the clarification that this refers to an ASK Hall resistivity possibly containing an anomalous component.

There are a few minor points I would like to see addressed prior to acceptance, however.

1.) It would be instructive to calculate the expected antiskyrmion density from the Hall measurements if this were a purely THE effect and compare that to the measured density (i.e. using data similar to the supplementary video), i.e. $\rho^{THE} = -P \cdot R_0' \cdot (n_{ASK} \cdot \Phi_0)$, where P is the spin polarization of the charge carriers, R_0' is the Hall coefficient representing the effective charge density contributing to the THE (and is normally assumed to be equal to the classical Hall coefficient), and $n_{ASK} \cdot \Phi_0$ is the magnitude of the emergent field associated with a given antiskyrmions density. n_{ASK} is the antiskyrmion density and $\Phi_0 = h/e$ (see Phys. Rev. Lett. 102, 186602 (2009) or Nat. Comm. 12, 2758 (2021)). I do think that it is important to show the theoretical estimate for antiskyrmion density from transport measurements to experimentally measured antiskyrmion density.

The equation of Phys. Rev. Lett. 102, 186602 (2009) approximates a “simple” metal, where the effective skyrmion field is acting only on one present spin state with only a single orbital character. We thank the referee for the response, as in our review we have found that the approximation used in the above equation is not sufficient for the explanation of Hall effect in the antiskyrmion phase of $Mn_{1.4}PtSn$. From the previous work of the authors [see Vir et al., PRB **99**, 140406(2019)] we employ a method that gives a clearer picture in accordance with other works that predict the THE [ref. 44]. In the case of the multiband metal $Mn_{1.4}PtSn$, the topological hall contribution is derived from the subtraction of spin-up and spin-down contributions to the Hall conductivity (addition for the normal Hall conductivity), which is more complex than MnSi. A more detailed approach of the electronic structure is necessary [see Franz et al., PRL 112, 186601 (2014), ref. 44]. This is because in MnSi the spin-up (dominate state) and spin-down are fairly separated at the Fermi energy, whereas in $Mn_{1.4}PtSn$ due to the magnetic-sublattice structure the spin-up and spin-down are mixed. A computational value was already provided by previous calculations of the authors [see Vir et al., PRB **99**, 140406(2019)]. This result is modified $\rho^{THE} = R_{xy}^{THE} \cdot (n_{ASK} \cdot \Phi_0)$, which is equation 16 in ref. 44. R_{yx}^{THE} for $Mn_{1.4}PtSn$ was calculated to be $1.5 \cdot 10^{-8} \Omega cm/T$.

$$|\rho^{THE}| \sim 1.5 \cdot 10^{-8} \cdot (100 \cdot 10^{-9})^2 \cdot (6.626 \cdot 10^{-34} / 1.602 \cdot 10^{-19}) = 62.04 \text{ n}\Omega cm$$

Furthermore, The experimental value of R_0 , is determined from the slope of the normal Hall resistivity in fields above 2 T at room temperature with a value of $\sim 4 \cdot 10^{-8} \Omega cm/T$ [see main text and supplementary information in Vir et al., PRB **99**, 140406(2019)]. Here this R_0 is what is considered by the reviewer. The ASK density for a 1 μm thick device can be approximated to $(100 \text{ nm})^{-2}$. The $Mn_{1.4}PtSn$ crystal structure is due to a Jahn-Teller type distortion, where the

spin-up and spin-down density of states at the Fermi level become flat and nearly equal (but very different for the Fermi sea). The spin polarization is then determined in the polarized state from subtraction of the spin-up and spin-down density of states at the Fermi level. With our phase appearing nearly at the polarized state we approximate an upper bound spin polarization value of ~ 0.1 and we can evaluate the THE from Phys. Rev. Lett. 102, 186602 (2009):

$$|\rho^{\text{THE}}| \sim (0.1) * 4 * 10^{-8} * (100 * 10^{-9})^2 * (6.626 * 10^{-34} / 1.602 * 10^{-19}) = 16.54 \text{ n}\Omega\text{cm}$$

Here, we can clearly see that the methods of Franz et al., PRL 112,186601 (2014) produced a theoretical result of the same order of magnitude. Whereas the equation in Phys. Rev. Lett. 102, 186602 (2009) is significantly smaller. This also confirms the discrepancy in Vir et al., PRB **99**, 140406(2019) and the theoretical calculation, where the R_{xy}^{THE} would require an antiskyrmion field of nearly 10 T much too large for antiskymions of ~ 100 nm scale (~ 0.4 T). Furthermore, the excellent agreement provides further insight when considering thicker samples with expected larger antiskyrmion radius. The bound of the topological Hall effect for antiskyrmion radii between 100 nm and 350 nm is 62.04 n Ω cm and 5.06 n Ω cm. The Hall resistivity measured by our experiments and composed of both anomalous and topological contributions in the suspected antiskyrmion phase lies directly between the bounds of the theoretical results. There is a stronger discrepancy for larger antiskyrmion radii, congruent with the result that the antiskyrmion phase becomes more energetically unfavorable.

We included a brief discussion at the end of Supplementary Note 7, where we provide this estimate for the THE component. We further added the following sentences into the thickness dependence section in the main text:

“In Supplementary Note 7 we provide a theoretical estimation following the previous approaches [13,44] for a known ASK density. For our microscale devices the ASK size is approximately 100 nm, which would lead to a theoretically expected THE component of $|\rho^{\text{THE}}| \sim 50 \text{ n}\Omega\text{cm}$.”

Furthermore, we added the following sentences to the conclusion:

"Our theoretical prediction of the ASK THE magnitude matches the observed Hall signature values, thereby, displaying the importance of the electronic structure for the unexpectedly large Hall signature. However, the disentanglement of the anomalous and topological Hall contributions remains a challenge. In addition, the temperature dependence of the magnetic anisotropy, reflected in our Hall and FMR results, reveals the intimate link to the crystalline, electronic and magnetic structure."

2.) In the supplementary video, I'm assuming the units should be mT, not T.

Thank you for that comment. We have revised the field unit in the video accordingly.

Reviewer #4 (Remarks to the Author):

The authors have responded to my questions and comments, and combined with comments from the other reviewers, they have modified the paper in a thoughtful way. I am happy that the authors have now taken into careful consideration the contribution of an anomalous Hall component in the total measured transverse voltage signal, and this has meant they are much more circumspect when referring to a possible topological Hall contribution, which I think is highly appropriate. I am happy to recommend this manuscript be accepted, with only some minor points below I wish to make.

There are still some instances where the Supplementary Note number cited in the text does not match where the actual information is. Some of the ones I noted as minor corrections in my previous review have not, in fact, been corrected. For instance, in my previous review, I noted the following minor corrections:

Supp. Note 7 mentions extracting the THE by first determining the orbital and anomalous components ρ_{xy}^{AHE} and ρ_{xy}^{THE} , but based on what is written next about ‘the former’ and the ‘latter’ I think these should be ρ_{xy}^{OHE} and ρ_{xy}^{AHE} , respectively. Also, the text refers to an eq. 1 in the main text which is not there. Near the bottom Figs S12a and S12b are mentioned, but these should be Figs S13a and b.

The response letter noted ‘We have corrected these typos.’, however the mention of subtracting ρ_{xy}^{AHE} and ρ_{xy}^{THE} to find the THE (it should be ρ_{xy}^{OHE} and ρ_{xy}^{AHE} that are subtracted) is still there. The other two corrections have been made.

Similarly, for another minor correction I recommended:

Fig S11 – should the y axis values for parts c. and d. be negative? Otherwise subtracting the linear slope shown results in positive ρ_{xy} values for parts e and f.

The response states ‘We corrected the sign in Fig. S11c and d.’, however while the text of the caption has changed slightly, the figures are identical to how they were before revision, with no sign change.

Thank you for pointing these mistakes out to us again. We tried to remove all typos and misleading references. We changed the sign in Fig. S11c and d to negative. This change is necessary because we present the difference between the up and the down sweep.

Some examples where the cited Supplementary Note is wrong:

Page 6: “...the magnetization, as we show in Supplementary Note S6.” Supplementary Note S6 provides details of the atomistic spin-dynamic simulations and it is Supplementary Note S4 that shows the magnetisation.

Page 6: “In Supplementary Notes S4 and S5, we present an additional sequence of MOKE images recorded for device C...”

Note S4 is the magnetisation and shows no MOKE images – these are shown only in S5.

I do not consider these continuing typos to be enough to REQUIRE changes be made, however I

strongly urge the authors to check carefully all of their references to Supplementary Notes and Figures before publication to ensure they are correct. I suspect the authors have sometimes used the number for a Supplementary Figure instead of a Supplementary Note.

This was unfortunate! Some of the errors slipped through. We have now revised the main text and supplementary considering every comment, including the previous ones. We made sure that the references to figures and the supplement are correct.

8th Nov 22

Dear Dr Helm,

Please accept our most sincere apologies for the long duration of this round of review, due to a significant delay in obtaining the final report from one of the referees.

Your manuscript titled "Antiskyrmions and their electrical footprint in crystalline mesoscale structures of $\text{Mn}_{1.4}\text{PtSn}$ " has now been seen again by Reviewers #1 and #2, 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 under the open access CC BY license (Creative Commons Attribution v4.0 International License).

We therefore invite you to revise your paper one last time to address the remaining concerns of Reviewer #1 in relation to semantics and wording, which requires absolute clarity in distinguishing anomalous and topological Hall effects throughout the manuscript. At the same time we ask that you 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.

Please note that it may still be possible for your paper to be published before the end of 2022, but in order to do this we will need you to address these points as quickly as possible so that we can move forward with your paper.

EDITORIAL REQUESTS

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

Best regards,

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Editorial Board Member
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orcid.org/0000-0002-0892-0982

&

Dr Aldo Isidori
Senior Editor
Communications Materials

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors have extensively answered my previous points and we may indeed have a misunderstanding here. I will address this now in more detail and give a few minor points the authors may want to address before publication.

1) To be clear, I did neither expect the authors to change the title to specify the used material

system nor do I doubt the relevance of combined Hall measurements and imaging. However, I think that the focus committed in title/abstract and introduction seems to be partly left over from a previous version of the manuscript and does not describe the intended focus of the paper in an optimal way. Given the history of this review process, coming from the report of a THE signal that was rephrased into a combined THE and AHE signal, I am missing some adjustments to immediately make it clear to a reader that the focus of the work lies within the content the authors conveyed to me in their last response letter and not in the detection of ASKs via electrical means:

<i>“Our simulations provide the first direct simulation of the process taking into account the complex magnetism. This has not been captured by any of the micromagnetic simulations in the field so far. We solve the discrepancy (that has been around for a while) between experiments performed on either bulk or very thin electron-transparent samples. The ASK phase seems to establish only at the mesoscale in these materials. We agree that this only can be achieved by a significant contribution of dipole-dipole interactions. We provide a thickness dependence and show how the hard-magnetic behavior arises. We show that both MFM and MOKE can resolve the changes at the mesoscale. We combined MOKE with transport in mesoscale structure at various thicknesses. We conclude that we still would need to scale down thicknesses further in order to measure the THE of the ASKs quantitatively. This step is left for future experiments. Our work adds the temperature dependence. We show how the Hall feature disappears below TSR and provide evidence for a significant transformation of the magnetic ground state and its anisotropy.”</i>

The wording of e.g., the abstract implied to me that the research goal of the paper was largely unchanged, and the authors still try to obtain an unambiguous ASK Hall signature (as in: “a signal that would suffice to imply ASKs without imaging”, which it does not). The other, now in the manuscript focused aspects seem overly shortened in the abstract. As a result, the core element seemed not reached and thus a negative result. In their response, the authors now provided some explanation that helped to clarify what they want to convey. The authors should consider making it clearer where the actual impact of the work is supposed to be. I strongly suggest revisiting the corresponding sections in abstract and introduction, however, will not fight over semantics and not follow up on this point.

2) Following up on reviewer 2’s point: I also think it is important to stress that the measured result needs to be supported by imaging as (i.e. assuming the measured signal were purely a THE signal, the ASK density would be significantly higher). Maybe I misread their point, but a valuable piece of information would be the prediction of the ASK density based purely on the Hall signal and the subsequent comparison to the imaged density (not the calculated $\rho_{[THE]}$ but $n_{[ASK]}$, which should be way smaller in the images, right?).

3) “Our theoretical prediction of the ASK THE magnitude matches the observed Hall signature values, thereby, displaying the importance of the electronic structure for the unexpectedly large Hall signature.” I recommend wording it as “ASK THE and AHE” making it clear that the simulations model the entire system correctly and to have a better connection to the next sentence.

4) “Even more intriguing, a huge topological Hall effect emerges in the low-temperature region of the phase diagram, just below TSR, exceeding expectations for skyrmions [29].” Do the authors cite here the correct reference?

5) on the discussion following my previous point about the “huge topological Hall effect”:

The AHE has topological origins but in the manuscript the authors still clearly distinguish between AHE and THE. The wording should be clear.

Reviewer #2 (Remarks to the Author):

The authors have provided a thorough response to my concerns, and I feel they are sufficiently addressed.

Although the authors have satisfied my original concerns, I do have some concerns. While I do not think there is anything necessarily wrong with the paper as is, I agree with points raised by Reviewer 1, about the novelty.

The work does very thorough transport measurements, and the authors do clearly see an anomaly that can be of the same order as that estimated for THE, but they lack the detailed imaging data to truly make the claim - much to the authors credit, they are very clear that this is evidence for, and not complete proof, of ASK THE.

Therefore, while I do recommend for publication, this is not without some reservations regarding the novelty of the work.

Dear Editors,

In the following we provide a point-by-point response to the report of reviewer #1.

Reviewer #1 (Remarks to the Author):

The authors have extensively answered my previous points and we may indeed have a misunderstanding here. I will address this now in more detail and give a few minor points the authors may want to address before publication.

1) To be clear, I did neither expect the authors to change the title to specify the used material system nor do I doubt the relevance of combined Hall measurements and imaging. However, I think that the focus committed in title/abstract and introduction seems to be partly left over from a previous version of the manuscript and does not describe the intended focus of the paper in an optimal way. Given the history of this review process, coming from the report of a THE signal that was rephrased into a combined THE and AHE signal, I am missing some adjustments to immediately make it clear to a reader that the focus of the work lies within the content the authors conveyed to me in their last response letter and not in the detection of ASKs via electrical means:

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The wording of e.g., the abstract implied to me that the research goal of the paper was largely unchanged, and the authors still try to obtain an unambiguous ASK Hall signature (as in: “a signal that would suffice to imply ASKs without imaging”, which it does not). The other, now in the manuscript focused aspects seem overly shortened in the abstract. As a result, the core element seemed not reached and thus a negative result. In their response, the authors now provided some explanation that helped to clarify what they want to convey. The authors should consider making it clearer where the actual impact of the work is supposed to be. I strongly suggest revisiting the corresponding sections in abstract and introduction, however, will not fight over semantics and not follow up on this point.

>>

In order to provide an optimized overview about the key points of our work we have modified the abstract slightly. It now reads as follows:

“Skyrmionic materials hold the potential for future information technologies, such as racetrack memories. Key to that advancement are systems that exhibit high tunability and scalability, with stored information being easy to read and write by means of all-electrical techniques. Topological magnetic excitations such as skyrmions and antiskyrmions, give rise to a characteristic topological Hall effect. However, the electrical detection of antiskyrmions, in both thin films and bulk samples has been challenging to date. Here, we apply magneto-optical

microscopy combined with electrical transport to explore the antiskyrmion phase as it emerges in crystalline mesoscale structures of the Heusler magnet $Mn_{1.4}PtSn$. We reveal the Hall signature of antiskyrmions in line with our theoretical model, comprising anomalous and topological components. We examine its dependence on the vertical device thickness, field orientation, and temperature. Our atomistic simulations and experimental anisotropy studies demonstrate the link between antiskyrmions and a complex magnetism that consists of competing ferromagnetic, antiferromagnetic, and chiral exchange interactions, not captured by micromagnetic simulations.”

We tried to stay as brief as possible in order to stay within the allowed length. We understand the points of the reviewer and are grateful for the constructive comments. We think that the main concerns have been considered. We once again want to stress that the observation of a clear feature in the transport in the field and temperature range where the ASK phase shows up is a novel finding for Antiskyrmion compounds and sheds light on discrepancies between thin-film and bulk findings. On top of this, our work reports various firsts that deserve a proper highlighting. We therefore have no doubt on the novelty of the overall work presented in our paper.

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2) Following up on reviewer 2's point: I also think it is important to stress that the measured result needs to be supported by imaging as (i.e. assuming the measured signal were purely a THE signal, the ASK density would be significantly higher). Maybe I misread their point, but a valuable piece of information would be the prediction of the ASK density based purely on the Hall signal and the subsequent comparison to the imaged density (not the calculated $\rho_{\{THE\}}$ but $n_{\{ASK\}}$, which should be way smaller in the images, right?).

>>

As we described in our last response and also in the supplement, that estimates of the THE from the ASK density are less straight forward. The reason is the multiband nature of $Mn_{1.4}PtSn$, with multiple contributions to the spin-up and spin-down channels to be considered for the Hall effect. Hence, the calculation of $R_{0'}$ results in a surprisingly large THE. Moreover, even though n_{ASK} is rather small for thick devices a large THE can be expected in the case of $Mn_{1.4}PtSn$ and the overall detected particle density is less telling as compared to for example the B20 compounds.

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3) “Our theoretical prediction of the ASK THE magnitude matches the observed Hall signature values, thereby, displaying the importance of the electronic structure for the unexpectedly large Hall signature.” I recommend wording it as “ASK THE and AHE” making it clear that the simulations model the entire system correctly and to have have a better connection to the next sentence.

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In this particular sentence we refer solely to the estimated ASK THE. The magnitude of the AHE was not estimated. We therefore will keep this sentence unchanged.

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4) “Even more intriguing, a huge topological Hall effect emerges in the low-temperature region of the phase diagram, just below TSR, exceeding expectations for skyrmions [29].” Do the authors cite here the correct reference?

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We indeed cited the wrong work here. We have corrected this. Thank you for bringing that to our attention.

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5) on the discussion following my previous point about the “huge topological Hall effect”: The AHE has topological origins but in the manuscript the authors still clearly distinguish between AHE and THE. The wording should be clear.

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We have revised this sentence to:

“Even more intriguing, a huge additional component in the Hall effect, associated with a topological origin, emerges in the low-temperature region of the phase diagram, just below T_{SR} , exceeding expectations for skyrmions [33].”

We hope that our brief reply is sufficient to settle the last concerns of reviewer # 1.