Peer Review File

Handedness manipulation of propagating antiferromagnetic magnons

Corresponding Author: Dr Yoichi Shiota

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

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The authors, Yoichi Shiota et al., reported their work on the experimental and theoretical demonstration of manipulating and electrically reading out propagating magnon handedness (or chirality) in perpendicularly magnetized synthetic antiferromagnets (SAFs). This topic is timely, aligning with the recent growing interest in antiferromagnetic spintronics, which holds great potential for constructing magnonic quantum systems and handedness-based spintronics. Given that the right-handed (RH) and left-handed (LH) precession modes in collinear antiferromagnets can have different resonance conditions, the authors wisely chose a SAF sandwiched by a heavy metal layer with the same sign of spin Hall angle, allowing them to manipulate the coherently propagating magnon handedness in the SAF. Furthermore, the handedness can be easily switched by tuning the frequency of the excitation microwave. From a technical perspective, measuring the propagating characteristics of antiferromagnetic magnons in SAFs is both crucial and challenging, since ISHE is not a phase-sensitive technique. However, to their credit, the authors provide supplementary material that includes extensive data and discussion of these propagating characteristics. Overall, the data and analysis presented in this article are sound and convincing, making this an impressive piece of work that extends the scope of high-frequency antiferromagnetic spintronics. Therefore, I can recommend publication in Nature Communications. I have a few critical comments that the authors should address prior to publication.

Major points:

1. In Figure 2(b), the ANE voltage signal caused by the thermal effect of the microwave antenna has been properly measured and reasonably subtracted. It is worth noting that, in general, the precession of the magnetic moment in metallic films induces time-dependent AMR and AHE signals, which in turn lead to a spin rectification signal. Researchers in this field may wonder whether a spin rectification signal was also observed in this system and whether it was properly subtracted.

2. The authors should avoid drawing overselling conclusions in the discussion part. For instance, from line 216 to 220, the authors claim that their demonstrated method of handedness control and electrical detection of antiferromagnetic magnons is applicable to all antiferromagnets. If that were the case, the authors would not have needed to cleverly design such an artificial antiferromagnetic structure. If the authors believe that it is applicable to all antiferromagnets, even 'in principle', they should include an example, such as a non-collinear antiferromagnetic alloy system like Mn3Sn, which would make the conclusion more solid. Additionally, magnons in the THz range are primarily governed by exchange energy, which significantly differs from the energy range of magnons discussed in the present article, therefore, the term 'THz dynamics' (line 219) should be avoided.

3. From line 231 to 234, the authors found that the experimentally measured magnon decay length was longer than the results of numerical calculations. This finding is crucial for realizing antiferromagnetic magnon-based devices and should be another highlight of this article. It would be better to emphasize and discuss the underlying physical mechanisms in the main text, rather than presenting them as supplementary material (S5).

Minor points:

4. The interface quality between the Ru and the adjacent ferromagnetic layers is crucial for forming the synthetic antiferromagnet, and there is no structural information in the present paper. More information should be provided about the crystallographic structure, interface roughness, and element interdiffusion, which will be important for researchers who might attempt to reproduce the results of this paper.

5. It is recommended to label "m1" and "m2" in Figure 1 (b) and (c). This would help readers quickly establish the connection between Equation 1 on page 3 of the main text and the physical picture described in Figure 1 (b) and (c).

6. For the sample structure, the [Co(0.3)/Ni(0.6)]8.5, usually, the period number of superlattices is an integer. Here, the period number (8.5) is a decimal. What does "half period" mean here?

Reviewer #2

(Remarks to the Author)

The interesting work of Shiota et al, on the manipulation and detection of magnon handedness in synthetic antiferromagnets is well structured, is clear and has key aspects of novelty with respect to previous work by the authors. In my opinion the work deserved publication in Nature Communications, after the authors clarify some aspects especially related to the detection mechanism.

- Regarding the ISHE detection of the magnon handedness. The crucial effect enabling this possibility is the different precession amplitude in the top and bottom layer for the two modes. However, it is not clear from the paper how physically this difference in amplitude is linked to the magnon handedness of the mode, and what is the influence of the measurement configuration (e.g. field angle) on the relative precession amplitudes of the two layers. I suggest the authors provide and discuss e.g. additional calculations / simulations to clarify this aspect, which is crucial for their claim of "direct" handedness detection.

- It appears that the ISHE signal is strongly dependent on the theta field angle, however the authors present only data for theta = 30° . A more complete characterization of the device as a function of the angle theta would be very beneficial to clarify some aspects of the work (e.g. theta = 150°) and of the measurement configuration.

In view of generalizing the work claims (not critical for this work), would the authors' conclusions hold true in case of non-compensated saf, where non-compensation could be another source of different precession amplitudes in the two layers?
Minor point: Horizontal label missing from the inset of Fig. 2b.

- Details on the sample fabrication are missing from the Methods section.

Reviewer #3

(Remarks to the Author)

The manuscript by Yoichi Shiota et al. reports their experimental results on the handedness detection of propagating antiferromagnetic magnons. In synthetic antiferromagnetic (SAF) multilayers, they utilize a microwave to excite magnons and then probe them non-locally using the inverse spin Hall effect (ISHE) in the adjacent heavy metals. By the sign of the ISHE signal, the chirality of the antiferromagnetic magnons can be determined. The paper is well-organized and -written, the experimental results are novel and interesting, and I think it meets the high standards of Nature Communications. Before publication, I think these comments and questions need to be properly addressed.

(1) For uniaxial antiferromagnets, two magnon modes with right-hand and left-hand chirality degenerate at zero-field; however, in the SAF system, the degeneracy happens at the non-zero field and is attributed to the difference of the magnetic anisotropy in the top and bottom ferromagnetic layers. In principle, the shift of the degenerate field indicates there is an effective magnetic field. I notice that in Fig. S2, the shifts of the degenerate field in three samples are different, are they random in samples? Can the shift be controlled?

(2) The following question is that I do not see transparently why the head-to-head (H-H) and tail-to-tail (T-T) configuration of the magnetic moments in the top and bottom ferromagnetic layers could induce the opposite shift of the degenerate field, could the author provide some intuitive picture for this?

(3) I am not very familiar with the SAF; I am curious about whether 0.42 nm thick Ru forms a continuous film or not; did the author check the cross-section? Or adding some references on this is also ok.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The authors have responded appropriately to all of my comments. The achievement presented in this study is significant and interesting for a broad audience devoted to antiferromagnetic spintronics. Overall, I recommend the publication of this work in Nature Communications.

Reviewer #2

(Remarks to the Author)

The authors replied satisfactorily to the reviewers' comments. I believe the paper is now suitable for publication in Nature Communications.

Reviewer #3

(Remarks to the Author) I think the authors properly addressed my questions and comments, so I suggest to publish it now.

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Enclosed please find the manuscript of the paper entitled; "Handedness manipulation of propagating antiferromagnetic magnons" Y. Shiota, T. Taniguchi, D. Hayashi, H. Narita, S. Karube, R. Hisatomi, T. Moriyama, and T. Ono

We would like to thank the editors and the reviewers for their work in considering the above paper for a publication in *Nature Communications*. The reviewer's comments have allowed us to improve our manuscript considerable, and we successfully revised the manuscript based on their remarks. Please find below our response to the comments raised by the reviewers. The modifications are indicated in our responses as well as are highlighted in red in the manuscript. We believe the manuscript is now ready for the publication in *Nature Communications*.

Reviewer comments and responses

Reviewer #1 (Remarks to the Author):

The authors, Yoichi Shiota et al., reported their work on the experimental and theoretical demonstration of manipulating and electrically reading out propagating magnon handedness (or chirality) in perpendicularly magnetized synthetic antiferromagnets (SAFs). This topic is timely, aligning with the recent growing interest in antiferromagnetic spintronics, which holds great potential for constructing magnonic quantum systems and handedness-based spintronics. Given that the right-handed (RH) and left-handed (LH) precession modes in collinear antiferromagnets can have different resonance conditions, the authors wisely chose a SAF sandwiched by a heavy metal layer with the same sign of spin Hall angle, allowing them to manipulate the coherently propagating magnon handedness in the SAF. Furthermore, the handedness can be easily switched by tuning the frequency of the excitation microwave. From a technical perspective, measuring the propagating characteristics of antiferromagnetic magnons in SAFs is both crucial and challenging, since ISHE is not a phasesensitive technique. However, to their credit, the authors provide supplementary material that includes extensive data and discussion of these propagating characteristics. Overall, the data and analysis presented in this article are sound and convincing, making this an impressive piece of work that extends the scope of high-frequency antiferromagnetic spintronics. Therefore, I can recommend publication in Nature Communications. I have a few critical comments that the authors should address prior to publication.

[Response]

We would like to thank the reviewer for his/her careful review of our manuscript and positive evaluation. According to the suggestion from the reviewer, we have revised the manuscript as follows.

Major points:

1. In Figure 2(b), the ANE voltage signal caused by the thermal effect of the microwave antenna has

been properly measured and reasonably subtracted. It is worth noting that, in general, the precession of the magnetic moment in metallic films induces time-dependent AMR and AHE signals, which in turn lead to a spin rectification signal. Researchers in this field may wonder whether a spin rectification signal was also observed in this system and whether it was properly subtracted. [Response]

Since the microwave antenna is electrically isolated from the Hall bar structures by the 50-nmthick SiO_2 layer, the microwave current flows only into the antenna. As a result, spin rectification signal due to AMR and AHE is absent detected in our devices.

To make this point clear, we added explanation in the revised manuscript as follows:

(Page 5 Line 157 in the revised main text)

"It should be noted that the spin rectification signal due to anisotropic magnetoresistance and anomalous Hall effect is absent detected in our devices, because the microwave antenna is electrically isolated from the Hall bar structure by the 50-nm-thick SiO₂ layer (see Supplementary S3)."

2. The authors should avoid drawing overselling conclusions in the discussion part. For instance, from line 216 to 220, the authors claim that their demonstrated method of handedness control and electrical detection of antiferromagnetic magnons is applicable to all antiferromagnets. If that were the case, the authors would not have needed to cleverly design such an artificial antiferromagnetic structure. If the authors believe that it is applicable to all antiferromagnets, even 'in principle', they should include an example, such as a non-collinear antiferromagnetic alloy system like Mn3Sn, which would make the conclusion more solid. Additionally, magnons in the THz range are primarily governed by exchange energy, which significantly differs from the energy range of magnons discussed in the present article, therefore, the term 'THz dynamics' (line 219) should be avoided.

[Response]

According to the reviewer's suggestion, we have revised the manuscript as follows:

(Page 8 Line 253 in the revised main text)

"Our demonstrated method of handedness control and electrical detection of antiferromagnetic magnons is a major step towards harnessing the full potentials of antiferromagnetic magnons, such as the high-speed spin dynamics, robustness against external perturbations, and the polarization degree of freedom."

3. From line 231 to 234, the authors found that the experimentally measured magnon decay length was longer than the results of numerical calculations. This finding is crucial for realizing antiferromagnetic magnon-based devices and should be another highlight of this article. It would be better to emphasize and discuss the underlying physical mechanisms in the main text, rather than presenting them as supplementary material (S5).

[Response]

Thank you for highlighting the importance of measured magnon decay length. To address the

reviewer's suggestion, we have moved the relevant content from the supplementary materials into the newly added section "Propagating magnon decay length" in the revised main text.

Minor points:

4. The interface quality between the Ru and the adjacent ferromagnetic layers is crucial for forming the synthetic antiferromagnet, and there is no structural information in the present paper. More information should be provided about the crystallographic structure, interface roughness, and element interdiffusion, which will be important for researchers who might attempt to reproduce the results of this paper.

[Response]

In the revised manuscript, we have referred prior works (Refs. 35 and 36) that thoroughly examined the crystallographic structure and interface roughness for similar systems. We confirmed that the first oscillation peak of the antiferromagnetic interlayer exchange coupling was obtained at around 0.4-nm-thick Ru spacer layer in our p-SAF structure, which is consistent with that of the prior works.

(Page 4 Line 122 in the revised main text)

"Even though the 0.42-nm-thick Ru spacer layer is atomically thin, the first oscillation peak of the antiferromagnetic interlayer exchange coupling and atomically smooth interfaces have been confirmed in the prior works^{35,36}."

5. It is recommended to label "m1" and "m2" in Figure 1 (b) and (c). This would help readers quickly establish the connection between Equation 1 on page 3 of the main text and the physical picture described in Figure 1 (b) and (c).

[Response]

Thank you for this indication. We have added the labels " \mathbf{m}_1 " and " \mathbf{m}_2 " in Figs. 1b and 1c in the revised manuscript.

6. For the sample structure, the [Co(0.3)/Ni(0.6)]8.5, usually, the period number of superlattices is an integer. Here, the period number (8.5) is a decimal. What does "half period" mean here? [Response]

The use of decimal period number, such as 8.5, indicates that the superlattice structure has a Co layer on both the top and bottom of the film stack, with one side having an additional half period. This design choice is intentional and has been made to optimize the antiferromagnetic interlayer exchange coupling through the Ru spacer layer.

We have clarified this in the revised manuscript to avoid any confusion as follows:

(Page 4 Line 120 in the revised main text)

"The use of decimal period number, such as 8.5, indicates that the superlattice structure has a Co layer on both the top and bottom of film stack, with one side having an additional half period."

Reviewer #2 (Remarks to the Author):

The interesting work of Shiota et al, on the manipulation and detection of magnon handedness in synthetic antiferromagnets is well structured, is clear and has key aspects of novelty with respect to previous work by the authors. In my opinion the work deserved publication in Nature Communications, after the authors clarify some aspects especially related to the detection mechanism. [Response]

We would like to thank the reviewer for his/her positive feedback and for recognizing the novelty of our work. We have addressed the points raised by the reviewer in the revised manuscript as detailed below.

- Regarding the ISHE detection of the magnon handedness. The crucial effect enabling this possibility is the different precession amplitude in the top and bottom layer for the two modes. However, it is not clear from the paper how physically this difference in amplitude is linked to the magnon handedness of the mode, and what is the influence of the measurement configuration (e.g. field angle) on the relative precession amplitudes of the two layers. I suggest the authors provide and discuss e.g. additional calculations / simulations to clarify this aspect, which is crucial for their claim of "direct" handedness detection.

[Response]

We have performed the simulation of the magnetization trajectories and time-evolution of magnetization precession, as shown in Fig. R1. These simulations clearly indicate how the difference in precession amplitudes between the top and bottom layers is linked to the magnon handedness. We believe these additions strengthen our claim "direct" handedness detection.

We have added these results and relevant discussion in S5 of the revised supplementary materials.



Fig. R1. a-d, Calculated magnetization trajectories and time-evolution of magnetization precession for Pt-SAF-Pt structure under external magnetic field $\mu_0 H_{\text{ext}} = 90 \text{ mT}$, $\theta_H = 30^\circ$ with (a,c) $f_{\text{rf}} = 11.5 \text{ GHz}$ and (b,d) $f_{\text{rf}} = 18.1 \text{ GHz}$.

- It appears that the ISHE signal is strongly dependent on the theta field angle, however the authors present only data for theta = 30° . A more complete characterization of the device as a function of the angle theta would be very beneficial to clarify some aspects of the work (e.g. theta = 150°) and of the measurement configuration.

[Response]

We appreciate the comment regarding the angle dependence of ISHE signal. While we agree that a more complete characterization over various θ_H angle would provide additional insights, we currently do not have comprehensive angle-dependent data. However, we have the data at $\theta_H = 150^\circ$ (Fig. R2), which is approximately same with the data for $\theta_H = 30^\circ$. This result is consistent with our physical picture that the ISHE signal depends on the tilted angles of the magnetization. Therefore, it becomes another evidence supporting the present result.

We have included this data in the revised supplementary material and discuss its relevance to the observed phenomena in S4 of the revised supplementary material.



Fig. R2. a,b, ΔV_{ISHE} spectra under the tilted magnetic field $\theta_H = 150^\circ$ for H-H antiparallel magnetization configuration (a) and T-T antiparallel magnetization configuration (b).

- In view of generalizing the work claims (not critical for this work), would the authors' conclusions hold true in case of non-compensated saf, where non-compensation could be another source of different precession amplitudes in the two layers?

[Response]

Thank you for the insightful question. Indeed, in non-compensated SAFs where the thickness or magnetization of the top and bottom ferromagnetic layers differ, the difference in precession amplitudes could be more complicated than in the present case. We will address this issue as a key area for future work.

- Minor point: Horizontal label missing from the inset of Fig. 2b.

[Response]

We have added the horizontal labels in the inset of Figs. 2b

- Details on the sample fabrication are missing from the Methods section.

[Response]

Thank you for pointing out the need for more details on sample fabrication. We have moved the section of "Device fabrication" from the Methods into S3 of the supplementary material, including the detailed fabrication process with the schematic illustration. This will ensure that the sample fabrication is fully transparent.

Reviewer #3 (Remarks to the Author):

The manuscript by Yoichi Shiota et al. reports their experimental results on the handedness detection of propagating antiferromagnetic magnons. In synthetic antiferromagnetic (SAF) multilayers, they utilize a microwave to excite magnons and then probe them non-locally using the inverse spin Hall effect (ISHE) in the adjacent heavy metals. By the sign of the ISHE signal, the chirality of the antiferromagnetic magnons can be determined. The paper is well-organized and -written, the experimental results are novel and interesting, and I think it meets the high standards of Nature Communications. Before publication, I think these comments and questions need to be properly addressed.

[Response]

We would like to thank the reviewer for his/her positive evaluation of our manuscript and for acknowledging the novelty and significance of our experimental results. We have carefully addressed the points raised by the referee in the revised manuscript as detailed below.

(1) For uniaxial antiferromagnets, two magnon modes with right-hand and left-hand chirality degenerate at zero-field; however, in the SAF system, the degeneracy happens at the non-zero field and is attributed to the difference of the magnetic anisotropy in the top and bottom ferromagnetic layers. In principle, the shift of the degenerate field indicates there is an effective magnetic field. I notice that in Fig. S2, the shifts of the degenerate field in three samples are different, are they random in samples? Can the shift be controlled?

[Response]

The perpendicular magnetic anisotropy is influenced by several factors, including the material of the seed and cap layers, the thickness ratio of the Co/Ni layers in supper lattice, the number of repetitions, and whether the ferromagnetic layer is in the top or bottom layer of the SAF structure. By carefully controlling these parameters, we can tune the shift of the degenerate field.

(2) The following question is that I do not see transparently why the head-to-head (H-H) and tail-totail (T-T) configuration of the magnetic moments in the top and bottom ferromagnetic layers could induce the opposite shift of the degenerate field, could the author provide some intuitive picture for this?

[Response]

The degeneracy is resolved when the perpendicular magnetic anisotropy fields of two ferromagnets are different. In such a case, net perpendicular magnetic anisotropy field remains finite and points to the opposite direction between the H-H and T-T configurations, leading to the opposite shift of the degenerate field.

We have added this explanation in the revised supplementary material to clarify the intuitive connection between the magnetization configuration and the shift of the degenerate field as follows:

(Page 4 in the supplementary material)

"The crossing field at which the two precession modes degenerate is shifted from the zero magnetic field owing to the difference in perpendicular magnetic anisotropy between the lower and upper FM layers. In such a case, net perpendicular magnetic anisotropy field remains finite and points to the opposite direction between the H-H and T-T configurations, leading to the opposite shift of the degenerate field."

(3) I am not very familiar with the SAF; I am curious about whether 0.42 nm thick Ru forms a continuous film or not; did the author check the cross-section? Or adding some references on this is also ok.

[Response]

In the revised manuscript, we have referred prior works (Refs. 35 and 36) that thoroughly examined the crystallographic structure and interface roughness for similar systems. We confirmed that the first oscillation peak of the antiferromagnetic interlayer exchange coupling was obtained at around 0.4-nm-thick Ru spacer layer in our p-SAF structure, which is consistent with that of the prior works.

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"Even though the 0.42-nm-thick Ru spacer layer is atomically thin, the first oscillation peak of the antiferromagnetic interlayer exchange coupling and atomically smooth interfaces have been confirmed in the prior works^{35,36}."

Reviewer comments and responses

Reviewer #1 (Remarks to the Author):

The authors have responded appropriately to all of my comments. The achievement presented in this study is significant and interesting for a broad audience devoted to antiferromagnetic spintronics. Overall, I recommend the publication of this work in Nature Communications.

[Response]

We would like to sincerely thank the reviewer for the positive feedback and recommendation for publication. Reviewer's insightful comments have been valuable in improving the quality of the manuscript.

Reviewer #2 (Remarks to the Author):

The authors replied satisfactorily to the reviewers' comments. I believe the paper is now suitable for publication in Nature Communications.

[Response]

Thank you very much for the constructive feedback and for considering our revisions satisfactory. We are grateful for the recommendation to publish the paper in Nature Communications, and we truly appreciate your support throughout the review process.

Reviewer #3 (Remarks to the Author):

I think the authors properly addressed my questions and comments, so I suggest to publish it now. [Response]

We are pleased that the reviewer found our revisions adequate and now recommended the publication of the manuscript. Thank you for the insightful comments, which have helped us enhance the clarity and impact of our work.