Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The manuscript by Mohanta et al. reports on computer simulations of spin-waves in skyrmion crystals. The results which are presented might have some value but when I read the methods section a noted a flaw in the algorithm used for the Monte Carlo simulation.

Monte-Carlo algorithms have to be ergodic, which means that it must be possible for the algorithm to cover the whole phase space. The phase space here is the unit sphere. But the authors update spin direction in finite steps of angles phi and theta. With that, there model does not cover the whole phase space, instead they reach only discrete values of phi and theta. It is even worse, with that method the authors make a second mistake since they do not cover the phase space uniformly, since probability densities of random steps in phi and theta scale with cos(theta). This breaks a symmetry condition for Monte Carlo updates, namely that the probability to suggest (not accept) a step from state A -> B must be equal to suggesting the reversed step B -> A. For that reason, Monte Carlo procedures for continues spin models are normally not programmed in spherical coordinates but in cartesian.

I admit that the practical relevance of this flaw might be low, especially since the authors use Monte Carlo only for annealing. However, it must also be in the interest of the authors not to risk their reputation by publishing an algorithm with an (at least for experts in the field) obvious flaw. Hence, I recommend to reject the paper and give the authors the opportunity to revise their calculations.

Report to Manuscript COMMSPHYS-20-0360

In the manuscript "Spin-wave excitations of an emergent skyrmion crystal in manganite-iridate oxide interfaces", the Authors studied the spin-wave excitations in a 2D ferromagnet layer with interfacial Dzyaloshinskii-Moriya interaction (iDMI). Different phases of equilibrium state were considered, i.e. ferromagnetic (FM) phase, spin spiral (SS) phase, skyrmion crystal (SkX) phase, and an intermediate phase between the SkX and FM phases. $La_{1x}Sr_xMnO_3/SrIrO_3$ interface was used as a prototype platform, and possible experimental measurements were proposed. Also this issue has been extensively investigated, the Authors clearly presented abundant numerical results that are directly comparable with experimental data. In this sense, the manuscript is sound and somewhat novel. Scientists working in spintronics should be interested. I think this manuscript can be published in Communications Physics. However, I still have several questions and comments.

1. I think it is not necessary to emphasize the specific material, i.e. the $La_{1x}Sr_xMnO_3/SrIrO_3$ interface. The Hamiltonian used is a quite general one that can well describe many material systems, such as Pt/Co, Fe/Ir, etc. The material only specifies the parameters but should not alter the physics. Instead, I think the Authors should emphasize what new physics is discovered (As already cited in the manuscript, there are a lot of studies on the spin wave excitations in skyrmion crystals).

2. In the introduction, a sentence reads "The SkX phase exhibits spin-wave excitations which are Goldstone modes associated with the chiral spin arrangement of the state". I think a Goldstone mode should refer to the zero-energy mode corresponding to some simultaneous symmetry breaking. The dispersive spectra found here should not be Goldstone modes.

3. In the introduction, SkX phase, SS phase, and FM phase are introduced one after another. But in the **Results** part the sequence changed to FM, SkX, SS. I think it better to unify them.

4. In New J. Phys. **18**, 045015 (2016), spin-wave bands in SkX were calculated, and a topological non-trivial property was found. How the present results compare with theirs?

5. In the Hamiltonian, the dipole-dipole interaction was not included. In ferromagnetic systems, it has an important contribution that is comparable with other terms. It is useful to discuss about its effect.

6. It should be easy to analytically solve the spin wave dispersion in FM phase. It is better to compare the analytical curve with the S results, as well as for the FM modes in SkX and SS phases.

7. For the critical temperature of the formation of SkX, the Authors used "$\lessapprox$", which I suppose means an estimation by eyes. According to Phys. Rev. Lett. **110**, 177207 (2013), the formation of SkX is a first-order phase transition, meaning that the transition point can be precisely determined. Can the critical temperature be exactly determined here? In Fig. 5(a), the total topological number N_{sk} was used to describe the phase transition. Is it used as an order parameter here? In most papers, the average magnetization $\langle M_z \rangle$ is used. It may be useful to plot $\langle M_z \rangle$ together with N_{sk} to have a comparison.

8. About the "diffusive regime", which in my understanding is a transition region between the SkX phase and FM phase, is it a new phase, or the coexistence of SkX and FM phases (like a piece of melting ice)?

9. In the **Methods** part, it was explained that for each Monte-Carlo step the spin angle was updated by $\theta \pm 2^\circ$ or $\phi \pm 2^\circ$. In my understanding, a safe way to update the spin direction is to chose a new direction in a small cone spanned around the initial direction. Since the polar and azimuthal angles θ and ϕ are not equivalent, why is the scheme used here unbiased?

10. In the **Methods** part, Landau-Lifshitz equation was used to describe the spin dynamics without a damping term. Later, a Gaussian broadening was artificially introduced to mimic the effect of finite temporal step size. The damping (for example, well-known Gilbert damping) should also induce a line broadening. Could the Authors comment on the comparison between these two effects, and why the former was neglected?

To summarize, I recommend this manuscript for publication after some revisions. Since the main issue discussed in the manuscript has been studied for a long time, it is necessary to provide deeper understandings to meet the requirements for the journal.

Reviewer #3 (Remarks to the Author):

The paper studies spin wave modes in LaSrMnO/SrIrO system.
The authors use realistic parameters and calculate the state from MC calculations. Furthermore, the authors calculate spin wave spectrum.

It seems that there is not enough new physics to warrant publication in Communications Physics. Some additional justification for method could also be useful. For example, it is not clear why only 50 MC realizations are used for spin wave calculations.

Reviewer #1:

The manuscript by Mohanta et al. reports on computer simulations of spin-waves in skyrmion crystals. The results which are presented might have some value but when I read the methods section a noted a flaw in the algorithm used for the Monte Carlo simulation.

Monte-Carlo algorithms have to be ergodic, which means that it must be possible for the algorithm to cover the whole phase space. The phase space here is the unit sphere. But the authors update spin direction in finite steps of angles phi and theta. With that, there model does not cover the whole phase space, instead they reach only discrete values of phi and theta. It is even worse, with that method the authors make a second mistake since they do not cover the phase space uniformly, since probability densities of random steps in phi and theta scale with cos(theta). This breaks a symmetry condition for Monte Carlo updates, namely that the probability to suggest (not accept) a step from state A -> B must be equal to suggesting the reversed step B -> A. For that reason, Monte Carlo procedures for continues spin models are normally not programmed in spherical coordinates but in cartesian.

I admit that the practical relevance of this flaw might be low, especially since the authors use Monte Carlo only for annealing. However, it must also be in the interest of the authors not to risk their reputation by publishing an algorithm with an (at least for experts in the field) obvious flaw. Hence, I recommend to reject the paper and give the authors the opportunity to revise their calculations.

We thank the reviewer for the comment. We have modified our sampling algorithm and revised our manuscript accordingly. In the Figure 1 below, we compare the coverage of the phase space (unit sphere) in the previous sampling method and the newly implemented method at different number of Monte Carlo steps (N_{MC}). The new method has points spread uniformly while, as the referee pointed out, the previous method was non-uniform.

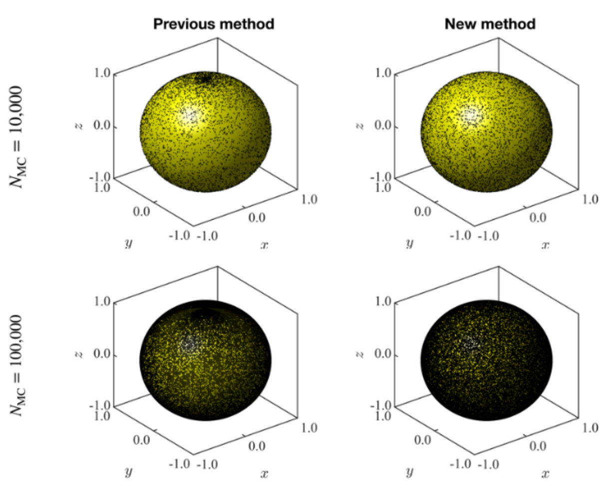
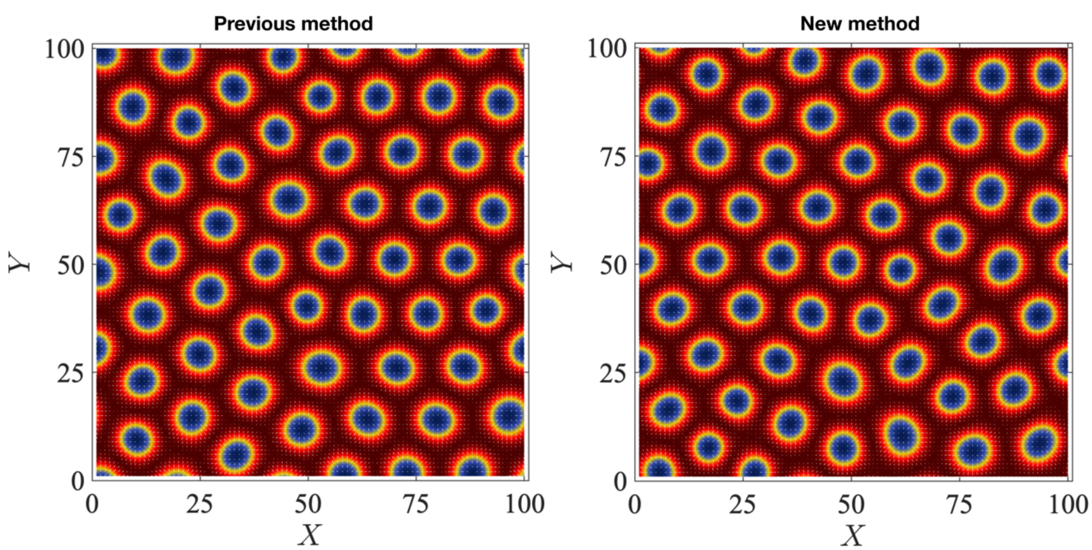


Figure 1: Comparison of the phase-space coverages in the previous and the new Monte Carlo sampling methods.

In the previous version, we used a large number (10^8-10^9) of Monte Carlo steps and small step size (2 degrees) in the coordinate angles which ensured a fairly broad coverage of the unit sphere. With large numbers of Monte Carlo steps, we always found a rotationally-invariant almost bias-free solution for the FM phase with D=0, Hz=0 and A=0. However, it is true that the probability density of the coordinates is denser towards the poles, as in Fig.1, and it violates the ergodicity, at least in principle. We have, therefore, modified our method and revised our manuscript.

In our new sampling method, we start with a random spin configuration and then choose a new spin direction within a small cone spanned around the initial spin direction. This method provides a full and uniform coverage of the phase space spanned by the spin angles, as in Figure 1 above.

We also made a thorough comparison of the Monte Carlo spin configurations obtained using the previous and the new sampling methods and, as the referee anticipated, found no qualitative difference. Below we show the MC spin configurations for the skyrmion crystal obtained using the two methods, for the same set of parameters. The reviewer also correctly pointed out, and we also confirm, that since the Monte Carlo method was used only for annealing to get the spin textures, there are no noticeable changes to the results for the dynamical structure factor presented in the manuscript.



We thank the reviewer again for the valuable comment and hope that the revised version of the manuscript (with corrected Monte Carlo sampling method) will now be suitable for publication in Communications Physics. With the correct Monte Carlo procedure suggested by the reviewer, fortunately none of the conclusions of our study has changed.

Reviewer #2:

In the manuscript “Spin-wave excitations of an emergent skyrmion crystal in manganite-iridate oxide interfaces”, the Authors studied the spin-wave excitations in a 2D ferromagnet layer with interfacial Dzyaloshinskii-Moriya interaction (iDMI). Different phases of equilibrium state were considered, i.e. ferromagnetic (FM) phase, spin spiral (SS) phase, skyrmion crystal (SkX) phase, and an intermediate phase between the SkX and FM phases. $La_{1-x}Sr_xMnO_3/SrIrO_3$ interface was used as a prototype platform, and possible experimental measurements were proposed. Also this issue has been extensively investigated, the Authors clearly presented abundant numerical results that are directly comparable with experimental data. In this sense, the manuscript is sound and somewhat novel. Scientists working in spintronics should be interested. I think this manuscript can be published in Communications Physics. However, I still have several questions and comments.

We thank the reviewer for carefully reading our manuscript and for the insightful comments/suggestions. We have modified our manuscript substantially to incorporate the suggestions and below we respond to the questions/comments.

1. I think it is not necessary to emphasize the specific material, i.e. the $La_{1x}Sr_xMnO_3/SrIrO_3$ interface. The Hamiltonian used is a quite general one that can well describe many material systems, such as Pt/Co, Fe/Ir, etc. The material only specifies the parameters but should not alter the physics. Instead, I think the Authors should emphasize what new physics is discovered (As already cited in the manuscript, there are a lot of studies on the spin wave excitations in skyrmion crystals).

   We thank the reviewer for this suggestion. It is true that the Hamiltonian used is quite general and the presented results are equally applicable to other two-dimensional skyrmion-host material systems. We wanted to draw the attention of the oxide interface community and the neutron-scattering experts about the opportunities for neutron-scattering detection of the spin-wave excitations. As suggested by the reviewer, we have made substantial changes to our manuscript; specifically, we have changed the title, modified abstract and added a discussion on other possible materials in which similar physics is expected to occur.

2. In the introduction, a sentence reads "The SkX phase exhibits spin-wave excitations which are Goldstone modes associated with the chiral spin arrangement of the state". I think a Goldstone mode should refer to the zero-energy mode corresponding to some simultaneous symmetry breaking. The dispersive spectra found here should not be Goldstone modes.

   The skyrmion crystal phase and the spin spiral phase break the translational symmetry in the two-dimensional plane. Therefore, there are zero-energy modes in these two phases associated with the broken translational symmetry. We have emphasized the word "translational" in the revised version. A rigorous analysis of the Goldstone modes in these two phases was done before, for example, in the two references below. We thank the reviewer for the interesting comment.

[43] M. Garst, J. Waizner, and D. Grundler, “Collective spin excitations of helices and magnetic skyrmions: review and perspectives of magnonics in non-centrosymmetric magnets,” Journal of Physics D: Applied Physics 50, 293002 (2017).

[42] O. Petrova and O. Tchernyshyov, “Spin waves in a skyrmion crystal,” Phys. Rev. B 84, 214433 (2011).

3. In the introduction, SkX phase, SS phase, and FM phase are introduced one after another. But in the Results part the sequence changed to FM, SkX, SS. I think it better to unify them.

We thank the referee for the suggestion. We have modified our presentation to bring the description of the SkX phase first, then the SS phase, and after that the FM phase, as discussed in the introduction.

4. In New J. Phys. 18, 045015 (2016), spin-wave bands in SkX were calculated, and a topological non- trivial property was found. How the present results compare with theirs?

We thank the referee for mentioning this reference which we missed in our previous manuscript. In our revised manuscript, we have cited this publication.

The paper New J. Phys. 18, 045015 (2016) discusses topological spin-wave excitations and chiral edge states in a skyrmion crystal. The authors of that paper identify several magnon bands, some of them having a non-zero Chern number. On the other hand, the dynamical structure factor in our realistic calculations captures seven modes which, we believe, are most relevant to neutron-scattering experiments. We have discussed the findings reported in this paper in our revised manuscript but refrained from making a one-to-one comparison since the calculation of the Chern number is not the focus of the present study.

5. In the Hamiltonian, the dipole-dipole interaction was not included. In ferromagnetic systems, it has an important contribution that is comparable with other terms. It is useful to discuss about its effect.

It is true that the dipole-dipole interaction has an important contribution in ferromagnetic materials, particularly in the absence of magnetic fields creating a mosaic of domains. Following the reviewer’s suggestion, we made a literature search to find out the range of the dominant magnetic exchange interaction in Sr-doped manganite systems. One recent publication [J. K. Tiwari et al. J. Phys. Condens. Matter 32, 195803 (2020)] extracted this information and according to their analysis of experimental data, the short-range exchange dominates over the long-range one (dipole-dipole interaction) in establishing the ferromagnetic order in manganites. While the long-range dipole-dipole interaction may be sizeable and help in stabilizing the magnetic phases in bulk manganite systems, we focus on the short-range magnetic exchange interactions which are relevant to the two-dimensional geometry. We have added a discussion in our revised manuscript on this issue and we thank the referee for raising this important comment.

6. It should be easy to analytically solve the spin wave dispersion in FM phase. It is better to compare the analytical curve with the S results, as well as for the FM modes in SkX and SS phases.

   Obtaining the analytical relation of the spin-wave dispersion for the SkX and the SS phases requires linear-spin-wave-theory calculations, which is beyond the scope of the present analysis and we leave it for a possible future study. For the FM phase, we plotted the analytical curve together with the S(q,w) plot and found that the two curves coincide very well, making it a bit difficult to visualize the Monte Carlo S(q,w) plot. We, however, thank the referee for this interesting suggestion.

7. For the critical temperature of the formation of SkX, the Authors used "$\lessapprox$", which I suppose means an estimation by eyes. According to Phys. Rev. Lett. 110, 177207 (2013), the formation of SkX is a first-order phase transition, meaning that the transition point can be precisely determined. Can the critical temperature be exactly determined here? In Fig. 5(a), the total topological number N_{Sk} was used to describe the phase transition. Is it used as an order parameter here? In most papers, the average magnetization $\langle M_z \rangle$ is used. It may be useful to plot $\langle M_z \rangle$ together with N_{Sk} to have a comparison.

   We thank the reviewer for this important question. In Phys. Rev. Lett. 110, 177207 (2013) and in Phys. Rev. B 87, 134407 (2013), the authors explored a fluctuations-induced first order transition from the paramagnet (PM) to the helimagnet (HM) phase (spin spiral phase in our description) at zero magnetic field. In our previous work [Phys. Rev. B 100, 064429 (2019)], we indeed found that the transition from the PM to HM phase at zero magnetic field is a sharp first-order transition. However, the transition from the PM to the SKX phase at finite magnetic fields occurs via an intermediate phase, the skyrmion gas phase, as we showed before [Phys. Rev. B 100, 064429 (2019)] and as was also studied by other groups [e.g., Phys. Rev. B 99, 064435 (2019), arXiv: 1811.01555]. The intermediate weak-crystallization regime between the PM and the SkX phase was also confirmed in experiments which show evidence for precursors of nontrivial topological winding of skyrmions present in the PM phase. The critical temperature is, therefore, not precisely defined. The total skyrmion number Nsk was used to extract the information about the phase transition from the PM phase to the SkX phase, and we carried out a similar analysis by considering Nsk as the order parameter. However, as the reviewer suggested, we have also added the average magnetization $\langle M_z \rangle$ in Fig.5(a) for comparison.

8. About the "diffusive regime", which in my understanding is a transition region between the SkX phase and FM phase, is it a new phase, or the coexistence of SkX and FM phases (like a piece of melting ice)?

The diffusive regime, involving nucleated skyrmions without a long-range order, may appear between the FM and the SkX phase and also between the SkX and the paramagnetic phase at higher temperatures. In our analysis, we explore the transition from the paramagnetic phase, which has an abundance of fluctuations, and the triangular SkX phase. It may be perceived as the melting of ice. In our revised manuscript, we have gathered some more data and turned our focus to the transition from the paramagnetic phase to the SkX phase, which, as also pointed out by previous experiments, resembles the liquid-solid transition.

9. In the **Methods** part, it was explained that for each Monte-Carlo step the spin angle was updated by $\theta\pm2^\circ$ or $\phi\pm2^\circ$. In my understanding, a safe way to update the spin direction is to chose a new direction in a small cone spanned around the initial direction. Since the polar and azimuthal angles θ and ϕ are not equivalent, why is the scheme used here unbiased?

   We thank the reviewer for pointing out this important question. We have implemented a bias-free sampling algorithm, as suggested by the reviewer and compared with the existing results. In the previous version of our sampling algorithm, we used a large number (10^8-10^9) of Monte Carlo steps and small step size in the coordinate angles which ensured a broad coverage of the unit sphere. With large numbers of Monte Carlo steps, we always found a rotationally-invariant almost bias-free solution for the FM phase with D=0, Hz=0 and A=0. However, it is true that the coverage of the unit sphere in the coordinate space is not perfectly uniform; the probability density of the coordinates is denser towards the poles. We have, therefore, updated our Monte Carlo sampling method and revised our manuscript accordingly.

   In our new sampling method, we start with a random spin configuration and then choose a new spin direction within a small cone spanned around the initial spin direction. This method provides a full and uniform coverage of the phase space spanned by the spin angles. However, we did not find any noticeable difference in the spin configurations at the set of parameters we used, between the previous sampling method and the new one; therefore, the results of the spin-wave spectra are kept unchanged.

10. In the **Methods** part, Landau-Lifshitz equation was used to describe the spin dynamics without a damping term. Later, a Gaussian broadening was artificially introduced to mimic the effect of finite temporal step size. The damping (for example, well-known Gilbert damping) should also induce a line broadening. Could the Authors comment on the comparison between these two effects, and why the former was neglected?

    The Gaussian broadening, which is used to minimize small oscillations along the frequency axis appearing due to the finite time step, would be relevant to the resolution of the neutron-scattering spectrometer. As correctly pointed out by the referee, the Gilbert damping may also introduce a broadening of the spin-wave bands. However, the Gilbert damping coefficient (α) for manganite is very low ($\sim 10^{-4}$) [see, for example, Q. Qin et al.

Ultra-low magnetic damping of perovskite $La_{0.7}Sr_{0.3}MnO_3$ thin films, Appl. Phys. Lett. 110, 112401 (2017)]. At these small values of α, only a tiny amount of broadening may be introduced to the higher-energy spin-wave excitations and, therefore, it can be safely ignored. We have added a discussion on this in our revised manuscript.

11. To summarize, I recommend this manuscript for publication after some revisions. Since the main issue discussed in the manuscript has been studied for a long time, it is necessary to provide deeper understandings to meet the requirements for the journal.

    We appreciate the valuable comments of the reviewer that helped us to improve the presentation. We have added a new figure and a discussion to focus on the paramagnet to SkX transition. We have revised our manuscript to take into account the suggestions of the reviewer and hope that the revised version of our manuscript will be suitable for publication in Communications Physics.

Reviewer #3:

The paper studies spin wave modes in LaSrMnO/SrIrO system. The authors use realistic parameters and calculate the state from MC calculations. Furthermore, the authors calculate spin wave spectrum.

It seems that there is not enough new physics to warrant publication in Communications Physics.

We thank the reviewer for carefully reading our manuscript and for the comments. We take this opportunity to mention again the new novelties of our manuscript.

Despite some previous studies on spin-wave excitations in the skyrmion crystal phase, there is no realistic calculation of the dynamical structure factor $S(\mathbf{q},\omega)$ which is the most important physical observable for the neutron-scattering measurements. We also discuss the opportunities for the neutron-scattering detection based on the manganite-iridate oxide interfaces that is expected to host a skyrmion crystal. The analytical results available [Phys. Rev. Lett. 108 (2012), Phys. Rev. B 84, 214433 (2011), 017601 (2012), J. Phys. D: Appl. Phys. 50 293002 (2017)] do not capture the full dispersion relation and our results in the skyrmion crystal reveal a rich structure of seven spin-wave modes that have not been reported before. Also, we extract the spin-wave dispersion in the diffusive regime that appears between the paramagnetic phase and the skyrmion crystal at finite temperatures. Such an analysis in the diffusive-scattering regime is not possible within the analytical frameworks that describe the T=0 case. Our analysis of the transition from the ring-shaped elastic $S(\mathbf{q},\omega)$ profile to the six-peak structure will be indispensable to distinguish a skyrmion crystal from the hexagonal ferrofluids, iron oxide nanoparticles, and hexagonal magnets. Thus, in our opinion there is plenty of new physics in our publication.

We have also revised our manuscript substantially and included new results to better present our main findings. We have now gathered new results to explore the phase transition from the paramagnetic to the SkX phases, that remained a puzzle for years and recent experiments indicate a liquid-crystal transition to the SkX phase.

Some additional justification for method could also be useful. For example, it is not clear why only 50 MC realizations are used for spin wave calculations.

50 MC independent thermalized realizations of the spin configurations were used to improve the spin-wave dispersion curve and also to reduce the noise coming from small statistical imperfections in the Monte Carlo spin configurations. We have added a note on this in our revised manuscript. Our results did not change using more independent configurations. Note that this number 50 should not be confused with the 10^9 MC steps performed to reach a thermalized regime from where the 50 independent configurations were gathered.

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The authors reacted to the criticism I brought forward in my first referee report. They corrected their Monte Carlo algorithm and repeated all calculations that were based on the Monte Carlo method. However, as I already anticipated, this does not change the results qualitatively and the conclusion remain solid. The response to my previous concerns is, hence, satisfactory.

Referee #3 has the opinion that "... there is not enough new physics to warrant publication in Communications Physics". Here, I disagree and I agree with the respone of authors that their numerical results go clearly beyond previous analytical solutions which neccessarily include approximation which have not been made in the submitted work.

Over all, I recommend to accept this work for publication.

Reviewer #2 (Remarks to the Author):

In the revised manuscript, the authors rewrote part of the manuscript to emphasize the spin wave excitations during liquid-crystal transition of skyrmions. The wrong Monte-Carlo method was corrected. The manuscript is better than before. Before publication, I still have several questions.

1, Indeed only when the thickness of the film is much smaller than the skyrmion size, which is true here since only 2D interface is considered here (see, e.g. supplemental materials of Communications Physics 1, 31 (2018) and Scientific Reports 8, 4464 (2018)). The authors only referred to an experiment that is not so convincing.

2, Although the Monte-Carlo algorithm is corrected, the figures used are still the same as the old version. Although the physics may remain the same, figures obtained by correct method should be used.

3, Since the authors intended to emphasize the liquid-solid transition of skyrmions, it is useful to refer to some previous studies, such as PRB 99, 064435 (2019), and discuss the connection and difference between them.

Response to Reviewer 1:

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Over all, I recommend to accept this work for publication.

We thank the reviewer for the remarks and for recommending our work for publication.

Response to Reviewer 2:

In the revised manuscript, the authors rewrote part of the manuscript to emphasize the spin wave excitations during liquid-crystal transition of skyrmions. The wrong Monte-Carlo method was corrected. The manuscript is better than before. Before publication, I still have several questions.

We thank the reviewer for the suggestions which we have incorporated in our revised manuscript.

1, Indeed only when the thickness of the film is much smaller than the skyrmion size, which is true here since only 2D interface is considered here (see, e.g. supplemental materials of Communications Physics 1, 31 (2018) and Scientific Reports 8, 4464 (2018)). The authors only referred to an experiment that is not so convincing.

We thank the reviewer for pointing to the two references which we have now cited in our revised manuscript.

2, Although the Monte-Carlo algorithm is corrected, the figures used are still the same as the old version. Although the physics may remain the same, figures obtained by correct method should be used.

We have performed the calculations again with the revised Monte Carlo method and revised all figures in our revised manuscript. There are, however, no qualitative differences of the new spin-wave spectra from the previous results.

3, Since the authors intended to emphasize the liquid-solid transition of skyrmions, it is useful to refer to some previous studies, such as PRB 99, 064435 (2019), and discuss the connection and difference between them.

We thank the reviewer for the reference which we have cited and discussed in our revised manuscript.

We hope that we have incorporated all the suggestions of the reviewer and modified our manuscript appropriately.