Dear Editor,

We thank the editor for considering our paper "Microscopic effect of spin-lattice couplings on dynamical magnetic interactions of a skyrmion system PdFe/Ir(111)" in the SciPost Physics. We very much appreciate positive referee reports and fruitful suggestions. We also would like to thank both the referees for his/her constructive comments which improves the quality of the revised manuscript. We believe that the both the referees found our work interesting and technically sound. Below we convey our response to the points raised by the referees and revised the manuscript accordingly. With these clarifications following the suggestions of the referees, we believe that the revised manuscript is now suitable for publication in SciPost Physics.

Yours Sincerely,

Banasree Sadhukhan,

Anders Bergman,

Johan Hellsvik,

Patrik Thunström,

Anna Delin

Referee Response : scipost-202409-00026v1

Response to the First Referee:-

Strengths :-

1. Referee comment : very original. The coupling between phonon and magnetic interactions is usually not studied because it very complex. Here the authors have used a combination of codes (VASP, Phonopy and home made second principle code) and techniques (MC, DFT) and are able to explain very well the outcome.

2. Referee comment : excellent bibliography. Pd/Fe/Ir(111) is a prototypical skyrmion ultra-thin film. There are a lot of work in this subject and it is sometimes difficult to navigate in all the references. The authors have done a great job in this respect.

3. Referee comment : clearly written

Our reply: We would like to thank referee for his/her valuable time reading

manuscript carefully and thoughtful reviews. In order to ensure that the manuscript reflects the state of the art in the field of modern spintronics, it refers to the most recent and relevant work in the field.

Weaknesses :-

1. Referee comment : the figures. Please put the article references on fig.1 and fig.2 Use thicker dashed lines in panel b like in panel a. Use also smaller symbols on panel a.

   Our reply: We would like to thank referee for useful suggestion. We added references in Fig.1 and Fig.2. If we thicker the lines in Fig.2(b), two successive lines coincide. Therefore, we kept same thickness of dashed lines in Fig.2.

2. Referee comment : the phonon spectra is not really useful here. It is mentioned that it is only here to check the stability of the ultra thin film however, the stability of PdFe/Ir(111) is not a key finding. It would be more interesting to study the substitution and intermixing. With the phonon band structure, one can do more that what is done in the paper i.e. establishing an elastic model for thermal phonon and coupling it to the extended Heisenberg. Since all phonon were calculated, the displacement can also be done along a specific phonon - also not done.

   Our reply We put the phonon spectra in the manuscript to check the stability of PdFe/Ir(111) multilayers and to reflect the message that the we started from the relaxed structure to calculate the spin-lattice couplings, estimation of dynamical magnetic interactions due to thermal vibrations.

3. Referee comment : the coupling between u and J or D is also not mentioned - the model could be refined here.

   Our reply: We would like to thank referee for comment. The couplings between u (thermal displacements) and J or D (magnetic interactions) are the spin-lattice coupling parameters $\Gamma_{ijk}^{\alpha\beta\mu}$ (see equation (2) in Sec. II). We added the word "thermal" before displacement in defining u of equation (2) to avoid confusion.

4. Referee comment : some typos (appendix C - formaliism).

Our reply: We would like to thank referee for pointing out. We corrected it in revised manuscript.

5. Referee comment : the effects of the displacement on the phase diagrams are not

Our reply: We would like to thank referee for comment. Exploring phase diagram using full spin-lattice dynamics (i.e using Hamiltonian in B1 in Appendix B) is beyond the scope of our current study.

We also have a future perspective in our study. In future, will study how the dynamical magnetic interactions affect the skyrmion dynamics i.e thermal stability, lifetime, energy barrier and phase diagram of PdFe/Ir(111) using full spin-lattice dynamics instead of spin Hamiltonian only. For that purpose, we are developing method for solving full spin-lattice dynamics where we used the calculated spin-lattice couplings and dynamical magnetic interactions.

Report :-

1. Referee comment : The article "Microscopic effect of spin-lattice couplings on dynamical magnetic interactions of a skyrmion system PdFe/Ir(111)" by Sadhukhan et al reports on the effect of atomic displacement on the parametrization of an extended Heisenberg Hamiltonian. PdFe/Ir(111) is one of the protypical skyrmion system. This system has been studied extensively since the last ten years and is now a very good test case. It is now being used to explore the coupling between magnetism and phonon displacement. This is an interesting topic which could have implications in the future spintronics and skyrmionics where the stability of skyrmions with respect to temperature is a crucial quantity and is a directly link to thermal phonon.

Our reply: We would like to thank referee for critically reading manuscript and fruitful suggestions.

2. Referee comment : The article is well written and the figure are appropriate but could be improved.

Our reply: We would like to thank referee for comment. We improved Fig.1 and Fig.2 as suggested by referee.

3. Referee comment : Apart from the comment above, I have a two requests. (a) The effect of the phonon is explored but no table summarizes the Hamiltonian parameters is given in the paper. It would be interesting to have these quantities to explore the stability diagram depending on phonon displacement. (b) the force constants obtained from the band structure should be published. As I said before the stability of this ultra thin film is not really interesting because it is obviously stable. The elastic parameters are more important. Even if the authors do not have access to a code with phonon and Heisenberg parameters, other people in the community might be interested by this quantity.

   Our reply: We would like to thank referee for fruitful suggestions. We added (a) Heisenberg parameters with thermal displacements (see table I) and (b) force constants (in appendix E) in the revised manuscript.

Requested changes :-

1. Referee comment : table with the different parameters of the extended Heisenberg

   Our reply: We would like to thank referee for fruitful suggestions. We added extended Heisenberg parameters in the revised manuscript.

Recommendation :-

1. Referee comment : Ask for minor revision

   Our reply: We thank referee for comments on our current work which increase the readability of the revised manuscript. It is our hope that we have satisfactorily responded to the points raised by the referee and revised the manuscript accordingly. We believe that the current study with revised version has enough merit for publication in SciPost Physics.

Response to the Second Referee:-

Strengths :-

1. Referee comment : The manuscript gives a detailed investigation of spin-lattice couplings (SLC) in a system where a skyrmion lattice can be induced by a magnetic field.

2. Referee comment : In principle, this could now be the starting point to check how structural modifications (due to anharmonic effects at finite temperature or at step-edges) change the stability of the skyrmion lattice.

3. Referee comment : Large SLC probably means also that the structure is modified by magnetic pattern. Maybe the authors can at some point estimate these changes for a skyrmion lattice of given size (2nd term in eq. B3)

   Our reply: We would like to thank referee for his/her valuable time reading manuscript carefully and thoughtful reviews. To study the effect of spin-lattice dynamics on the diameter of skyrmion, we need to solve full spin-lattice dynamics instead of spin Hamiltonian only. Therefore exploring the change in diameter of skyrmion using full spin-lattice dynamics (i.e using Hamiltonian in B1 in Appendix B) is beyond the scope of our current study.

   We also have a future perspective in our study. In future, will study how the dynamical magnetic interactions affect the skyrmion dynamics i.e thermal stability, lifetime, energy barrier, diameter and phase diagram of PdFe/Ir(111) using full spin-lattice dynamics instead of spin Hamiltonian only. For that purpose, we are developing method for solving full spin-lattice dynamics where we used the calculated spin-lattice couplings and dynamical magnetic interactions.

Weaknesses :-

1. Referee comment : In the title, 'dynamical magnetic interactions' are mentioned, so I expected to see some impact of SLC on the spin-dynamics. Maybe 'magnetic interactions' would be more clear.

Our reply: At finite temperatures, magnetic excitations are always associated with lattice vibrations. To describe the magneto-mechanical phenomena of a system, we should consider the lattice degrees of freedom in spin dynamics. When the spin and lattice degrees of freedom technically are coupled, it gives an intrinsic spin-lattice coupling, which describes how lattice vibrations microscopically affect the dynamical behaviour of magnetic interactions. With spin-lattice effects, the magnetic interactions are not static no more, they are dynamical. Therefore we used dynamical magnetic interactions instead of magnetic interactions only because here magnetic interactions are depend on lattice displacements (i.e thermal displacements).

2. Referee comment : See list of requested changes below.

Our reply: We would like to thank referee for comment. Below we addressed all points one by one and revised manuscript accordingly.

Report :-

1. Referee comment : The system Pd/Fe/Ir(111) was one of the first, where a skyrmion lattice could be induced via an external magnetic field. The stability depends sensitively on the stacking, and small structural changes can probably have a large effect on the exchange parameters. Here, the authors investigate the dependence of Heisenberg parameters (J) and the Dzyaloshinskii-Moriya interaction (DMI) on small displacements of the Fe positions, termed spin-lattice coupling (SLC). The parameters (up to 3rd nearest neighbors) are evaluated with density functional theory (DFT) and the stability of the skyrmion lattice checked with atomistic spin-dynamics simulations. Compared to systems investigated by some of the authors in other papers (CrI3 and bcc Fe), large valued of the SLC were found.

2. Referee comment : The manuscript is in most parts clearly written and addresses the topic in depth, there are, however, a few points that should be clarified (see below).

Our reply: We would like to thank referee for report and finding our manuscript clearly written with in depth analysis. Below we addressed all points one by one and revised manuscript accordingly.

Requested changes :-

1. Referee comment : The interlayer distances given on page 3 are too large. In an fcc(111) film, the ratio between interlayer distance and in-plane lattice constant should be sqrt(2/3). For Pd/Fe and Fe/Ir one expects them to be even smaller (see ref.[15])

   Our reply: We are aware of the paper given in Ref.[15] where the reported interlayer distances are $d_{Fe-Ir} = 2.06$ Å and $d_{Fe-Pd} = 2.02$ Å. Our calculated relaxed interlayer distances are $d_{Fe-Ir} = 2.63$ Å and $d_{Fe-Pd} = 2.62$ Å. The structural relaxation was done using LSDA as implemented in VASP. To check structural relaxation, we calculated the phonon dispersion spectrum (see Fig. 7(a)-(b) in Appendix). It demonstrates the absence of imaginary modes, i.e., the stabilization of the structure for both stacking. We highlighted the discussion in revised manuscript.

2. Referee comment : The type of exchange-correlation functional should be specified. Probably there are many different LSDA's in the used DFT codes, e.g. Perdew-Zunger or Vosko-Wilk-Nussair.

   Our reply: We would like to thank referee for this comment. We used Perdew-Zunger exchange-correlation functional in our LSDA calculations. We added it in revised manuscript.

3. Referee comment: Specifying the displacements, the authors use directions like [100] or [101]. Probably this does not correspond to the Ir lattice, please introduce the notation or use the bulk Ir directions as reference.

   Our reply: We would like to thank referee for this comment. The displacement of Fe along [100] (as shown in Fig.1(a)), [010] and [110] mean along x, y, xy directions respectively. They are in-plane displacements of Fe atoms. The displacement of Fe along [101] is along Fe-Pd bond length towards Pd layer and $[01\bar{1}]$ is along the Fe-Ir bond length towards Ir layers. We specified the displacement directions in the last paragraph of Sec.IIIC and to avoid confusion about direction of displacements, we added and revised the lines in the paragraph.

4. Referee comment : Related to that, looking at Fig.4, I did not figure out in which direction the Fe was displaced. Assuming that the shift in (b) and (e) is in y-direction, I had expected a (vertical) mirror symmetry in the panels. Maybe the authors can

indicate the shift direction in the panels.

Our reply: We would like to thank referee for comment. We added shift directions in Fig.4. The displacements along Fe-Ir [01$\bar{1}$] and Fe-Pd [101] are exactly opposite directions. Therefore Figures (b) and (c) have mirror symmetry for J_{ij}.

5. Referee comment : In the introduction, the authors write that 'A is the entire plane perpendicular to the propagation direction'. I don't know what propagates here.

   Our reply: We would like to thank referee for comment. We modified the line in revised manuscript for defining the topological charge of skyrmion. A magnetic skyrmion is characterized by an integer winding number or 'topological charge" Q, defined by $Q = \frac{1}{4\pi}\int_A \vec{n}\cdot\left(\frac{\partial\vec{n}}{\partial x}\times\frac{\partial\vec{n}}{\partial y}\right)dxdy$, where $\vec{n}(x,y)$ is the unit vector of the local magnetization, and the integral is taken over the surface area A.

6. Referee comment : On page 6 I read that 'due to thermal displacements' the Fe layer loses its 6-fold symmetry. What are thermal displacements in this connection? Anharmonic vibrations should mainly occur along the surface normal. In-plane, the layer lost it 6-fold symmetry already due to the substrate and overlayer.

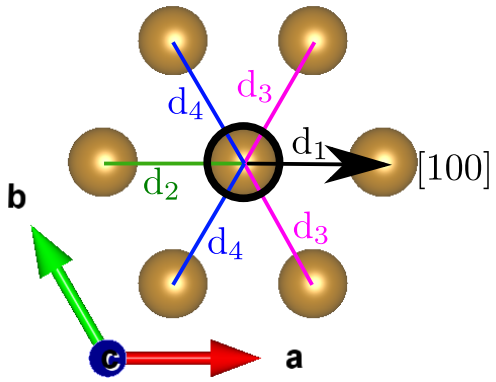

FIG. 1. In-plane displacement of Fe atom along [100] direction where C_6 symmetry of Fe monolayer has broken but still two pairs of atoms (marked by blue and magenta colours) are in same distance from the displaced atom.

Our reply: We would like to thank referee for comment. PdFe/Ir(111) do not have symmetries along out of plane direction (z direction) as due to the substrate and overlayer. But the hexagonal structure Fe monolayer in PdFe/Ir(111) multilayers

has C_6 symmetry (in-plane). Therefore, before considering spin-lattice interactions (i.e without displacements of Fe atoms) in-plane Fe atoms for PdFe/Ir(111) have C_6 symmetry. Now, in plane displacement of Fe atoms in hexagonal plane breaks C_6 but still atoms have few symmetries (see Fig.1) whereas full symmetry of all atoms have completely broken by out of plane displacements of Fe atoms.

7. Referee comment : In the references, please correct the capitalization in the titles (e.g. CrI3 instead of cri3).

   Our reply: We would like to thank referee for pointing out. We corrected it in revised manuscript.

Recommendation :-

1. Referee comment : Ask for major revision

   Our reply: We thank referee for fruitful comments on manuscript which increase the quality of the revised manuscript. We hope that we have satisfactorily responded to the points raised by the referee and revised the manuscript accordingly. We believe that the revised version is now suitable for publication in SciPost Physics.

List of changes:- All list of changes are in blue colour in the revised manuscript.

1. We revised the line in defining the topological charge of skyrmion in page 1.
2. We added the word "thermal" before displacement in page 2.
3. We added citations in Fig.1 and Fig.2 of page 3, 4.
4. We added exchange-correlation for LSDA calculations in page 3.
5. We added displacement directions in page 5.
6. We added table.I in page 6.
7. We corrected typos in revised manuscript.
8. We added the value of force constant in appendix E.
9. We added directions in Fig.4.