Dear Editor,

We thank the editor for considering our paper "Spin-lattice couplings and effect of displacements on magnetic interactions of a skyrmion system PdFe/Ir(111)" in the SciPost Physics. We very much appreciate first referee's fruitful suggestions which enhance the readability of the manuscript. With these clarifications following the suggestions of the referee, 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-202409-00026v2

Response to the Editor:-

1. The reviewer has raised several additional comments that should be addressed before publication. From my side, I fully agree with the reviewer's opinion that your work does not explore spin dynamics, so the title should be changed in order to better reflect the content of the manuscript.

   Our reply: We respectfully agreed with both editor and referee's comment about the title of the manuscript. We changed the title as "Spin-lattice couplings and effect of displacements on magnetic interactions of a skyrmion system PdFe/Ir(111)" in the revised manuscript. Below we also addressed other points one by one raised by first referee and revised manuscript accordingly.

Response to the First Referee:-

Report :-

1. Referee comment : The authors partially modified the manuscript according to the referee reports, however there are a few open points (see below).

Our reply: We would like to thank referee for critically reading manuscript and fruitful suggestions. Below we addressed others points one by one and revised manuscript accordingly.

Requested changes :-

1. Referee comment : On page 2, they call "u" now a "thermal displacement". This displacement might be due to thermal motion, but it could also result from other sources, e.g., an electric field. At the moment, it is just a displacement and should be called so.

   Our reply: We fully agreed with the referee and remove the word thermal before displacement in defining u.

2. Referee comment : New table I : since $J^{xx} = J^{yy} = J^{zz}$ and J^{xy}=-J^{yx} etc. only four out of nine tensor components need to be shown or J and D_x, D_y, D_z) .

   Our reply: We would like to thank referee for this comment. $J^{xx} = J^{yy} = J^{zz}$, but J^{ij}=-J^{ji} is not true for few cases. Such as $J^{xy} \neq$-J^{yx} for second nearest neighbour with and without spin-lattice effect, similarly, $J^{zy} \neq$-J^{yz} for first and second nearest neighbour with and without spin-lattice effect. $\boldsymbol{D}_{ij}$ and $\boldsymbol{C}_{ij}$ describe the antisymmetric and symmetric part of $J_{ij}^{\alpha\beta}$ which are calculated from $J_{ij}^{\alpha\beta}$ (see also equation D1 in appendix). For example, the z-component of $\boldsymbol{D}_{ij}$ and $\boldsymbol{C}_{ij}$ are defined as $D_{ij}^{z} = (J_{ij}^{xy} - J_{ij}^{yx})/2$ and $C_{ij}^{z} = (J_{ij}^{xy} + J_{ij}^{yx})/2$ [Physical Review Materials **6(8)**, 084401 (2022)]. We only analyze $\boldsymbol{D}_{ij}$ term further in the present work. Therefore, we keep the full magnetic exchange tensor $J_{ij}^{\alpha\beta}$ for completeness in the revised manuscript.

3. Referee comment : A force constant is given in Appendix E, but it's not clear for which atoms and why the xx, yy, and zz components are identical in a multilayer film.

   Our reply: We would like to thank referee for this fruitful comment. PdFe/Ir(111) contains five layers of Ir, one layer Fe and one layer Pd. The full set of calculated force constants are given in table I.

Atoms	$\phi_{ij}{}^{xx}$	$\phi_{ij}{}^{xy}$	$\phi_{ij}{}^{xz}$	$\phi_{ij}{}^{yx}$	$\phi_{ij}{}^{yy}$	$\phi_{ij}{}^{yz}$	$\phi_{ij}{}^{zx}$	$\phi_{ij}{}^{zy}$	$\phi_{ij}{}^{zz}$
Ir_1-Ir_1 :	10.42	0.00	0.00	0.00	10.42	0.00	0.00	0.00	10.97
Ir_2-Ir_2 :	17.09	0.00	0.00	0.00	17.09	0.00	0.00	0.00	13.89
Ir_3-Ir_3 :	16.39	0.00	0.00	0.00	16.39	0.00	0.00	0.00	4.05
Ir_4-Ir_4 :	14.87	0.00	0.00	0.00	14.87	0.00	0.00	0.00	12.17
Ir_5-Ir_5 :	13.56	0.00	0.00	0.00	13.56	0.00	0.00	0.00	13.77
Fe-Fe :	4.65	0.00	0.00	0.00	4.65	0.00	0.00	0.00	14.08
Pd-Pd :	10.28	0.00	0.00	0.00	10.28	0.00	0.00	0.00	6.14
Fe-Pd :	0.43	0.23	1.25	0.23	0.16	0.72	0.66	0.38	1.69
Ir_1-Fe :	0.02	0.01	0.04	0.01	0.02	0.02	0.01	0.00	0.04
Ir_1-Pd :	0.01	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.02
Ir_2-Fe :	0.02	0.00	0.00	0.00	0.00	0.01	0.00	0.01	0.01
Ir_2-Pd :	0.00	0.00	0.01	0.00	0.00	0.00	0.01	0.01	0.01
Ir_3-Fe :	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Ir_3-Pd :	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Ir_4-Fe :	0.01	0.04	0.05	0.04	0.06	0.03	0.12	0.07	0.31
Ir_4-Pd :	0.02	0.00	0.00	0.00	0.02	0.00	0.00	0.00	0.00
Ir_5-Fe :	0.11	0.00	0.00	0.00	1.42	2.21	0.00	2.29	3.05
Ir_5-Pd :	0.00	0.01	0.02	0.01	0.01	0.01	0.03	0.02	0.11

TABLE I. Force constants ($\Phi_{ij}^{xx} = \Phi_{ij}^{yy} \neq \Phi_{ij}^{zz}$ for same atoms) for PdFe/Ir(111) multilayers in the unit of eV/Å^2.

4. Referee comment : About the title: In an experiment at finite temperatures, magnetic interactions are naturally modified by the dynamics of the system, sure. What is calculated here (eq.2) is the effect of a displacement on a magnetic interaction at T=0, therefore, I don't think that "spin-lattice couplings on dynamical magnetic interactions" is a good description of the content. .

   Our reply: We respectfully agreed with the referee's comment. We changed the title as "Spin-lattice couplings and effect of displacements on magnetic interactions of a skyrmion system PdFe/Ir(111)" in the revised manuscript.

5. Referee comment : About the interlayer distances: since the system is stable and the exchange interactions agree also with other references, I think the results are OK, but maybe the authors give interatomic distances instead of interlayer distances. .

   Our reply: We would like to thank referee for this comment. Here we actually calculated the interatomic distances using Vesta software. (https://jp-minerals.org/vesta/en/) of the relaxed structure. We corrected the word interlayer distances replacing it by interatomic distances in revised manuscript.

6. Referee comment : A reference for the Perdew-Zunger LSDA is missing.

   Our reply: We would like to thank referee for this comment. We added reference [35] for the Perdew-Zunger LSDA.

7. Referee comment : About fig.4: Indeed, panels (b) and (c) show mirror symmetry, but along a plane 60 degrees tilted from the y-axis (for both panels). How does this match with the (in both panels, different) displacement directions? .

   Our reply: The Fig.4(b-c) show the change in J_{ij} with displacements of atoms for first six nearest neighbours (marked in red colour) of Fe atoms as shown in Fig.3(a). The displaced atom is marked as black in Fig.3(a). The displacements along Fe-Ir $[01\bar{1}]$ and Fe-Pd [101] are exactly opposite directions (Here both of them are along the out of plane but exactly opposite directions). Therefore Figures 4(b) and 4(c) have mirror symmetry for J_{ij}.

8. Referee comment : About the 6-fold symmetry: of course, a free monolayer has C_6 symmetry, but the "monolayer of Fe in PdFe/Ir(111)" has not, irrespective of thermal displacements. .

   Our reply: We respectfully disagree referee's comment. The monolayer of Fe in PdFeIr(111) has sixfold symmetry. We checked it during structural relaxation. Structural relaxation was done in two steps. First, we used selective dynamics in VASP and did ionic relaxation along the z axis of PdFe/Ir(111) multilayers. It restored the C_6 symmetry in the plane of Fe, Pd and Ir atoms. In the next step, we used this ionic relaxed structure for the full structural relaxation which included both ionic (for all x,

y, z directions) and volume relaxations. The final relaxed structure has C_6 symmetry in Fe monolayer of PdFe/Ir(111). Absence of negative frequencies in phonon dispersion shows the the dynamical stability of PdFe/Ir(111). We added more discussion on structural relaxation in 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.

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

1. We removed the word thermal before displacement in defining u in page 2.
2. We added list of full set of force constants in page 14.
3. We revised the title of the manuscript in page 1.
4. We added reference [35] for the Perdew-Zunger LSDA. in page 3.
5. We corrected the word interlayer distances as interatomic distances in page 3 of the revised manuscript.
6. We added more discussion regarding structural relaxation in page 12 and 13.