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Peer Review File

Electronic and magnetic excitations in La$_3$Ni$_2$O$_7$

Corresponding Author: Dr Ke-Jin Zhou

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)
In the present work, the authors employed X-ray absorption spectroscopy (XAS) and resonant inelastic X-ray scattering (RIXS) to detailly study the newly discovered bilayer high Tc superconductor La3Ni2O7, at ambient pressure, by combining the theoretical calculations. Based on the XAS and RIXS measurements, the spin-density-wave order was revealed at (0.25, 0.25) below 150 K. In addition, two possible interesting electronic states (spin-charge stripe and double spin stripe states) were proposed to understand the results obtained from RIXS. Furthermore, they also discussed the low-energy electronic structure using XAS and RIXS. Furthermore, they also found that the inter-layer effective superexchange interaction is an order of magnitude larger than that of the intra-layer. In general, those results look interesting and important for the study of bilayer nickelate superconductors, which deserves to be published somewhere. However, the theory component has some unclear issues.

(1) In the double spin stripe state (panel b of Fig. 3), they said a calculated magnetic coupling J1 to be zero. How can J1 be zero? From the geometrical perspective, the x2-y2 orbital overlaps with the px or py states in the plane and then overlaps with the next x2-y2 orbital, which is simply impossible to be zero. For what physical reason will J1 be 0 here?

(2) To obtain the magnetic coupling Js, the authors mapped DFT energy to the Heisenberg model. Have they tried other states to obtain magnetic coupling Js? The energy differences for those configurations look to be quite large at U = 3 and 4 eV. To map the DFT energy to the Heisenberg model to obtain J, it is better to use the magnetic states with closed energies.

(3) What are the calculated magnetic moments for different Ni sites of those stripes? Furthermore, the energy difference between the Stripe-2 and Stripe-3 states suddenly decreases at U = 4.0 eV. Why? Could they provide some explanations?

(4) The E-phase is well-known for decades in manganates and is also found in iron Te-based superconductors. Could the author briefly discuss the connections between those three families? Is the E-AFM phase discussed in the present work an insulating state here, as FeTe and manganates? What does the band structure of the E-phase (Stripe-2) look like?

Reviewer #2

(Remarks to the Author)
In this work, the authors presented the electronic and magnetic structures of the ambient-pressure normal-state of the recently-discovered La3Ni2O7 high-temperature superconductor. These are probed with a high level of details using x-ray absorption (XAS) and polarimetry-analysed resonant inelastic x-ray scattering (RIXS).

As the main result, the authors found that the magnetic structure of La3Ni2O7 is dominated by a spin-density wave (SDW) order. Interestingly, theoretical simulations of the magnon excitation associated with this order revealed that the out-of-plane 3dz2 orbital linking the bilayer structure of La3Ni2O7 is crucial for the magnetic structure. This is in contrast to the normal state of infinite-layer nickelate and cuprate superconductors, where the in-plane 3dx2-y2 orbital is thought to have a more important role instead.

Therefore, I think this is a unique and valuable contribution to the overall understanding of not only the La3Ni2O7

superconductor itself but also to the study of superconductivity in strongly-correlated materials in general.

However, I think the way that the manuscript is written is still too focused towards fellow experts who are already familiar with the topics of SDW, superconductivity, and x-ray spectroscopy. There are a lot of instances in the manuscript where several specific terms that are important for understanding the results and analysis are not adequately explained and discussed.

Thus, before I can support the publication of this manuscript, I suggest that in general the authors should revise the manuscript to be more suited to the broad audience of Nature Communications, especially in the Introduction where several of these key terms can be more adequately introduced.

For example, below are several places where the manuscript could be improved:

1. The sentence:
"Moreover, the bilayer structure of La3Ni2O7 may suggest a distinct minimal model in comparison to cuprate superconductors."
in the abstract is confusing and hard to understand, especially the term "distinct minimal model". The authors should find a better way to highlight the difference between La3Ni2O7 and the cuprate superconductors.

2. As the SDW is the main result of this paper, the authors should explain the concept of SDW in the introduction section, dedicating perhaps 1-2 paragraphs to introduce this concept to a general audience and its relationship and importance to superconductivity in strongly-correlated materials. This is so that the audience could more easily follow the analysis later in the results and discussion section.

3. I found that the three papers cited by the authors to support prior existence of SDW in La3Ni2O7 (Ref. 25,26,27) to not really be convincing, especially as two of them are still not peer-reviewed. A kink in electrical resistivity can easily be attributed to other explanations, uSR is a local technique and thus cannot distinguish between different types of long-range orders, and the NMR results support CDW scenario but not really SDW. So I think it is a bit disingenous to say that SDW in La3Ni2O7 has been supported by other results, as paragraph 2 seems to imply.

4. When discussing the RIXS results, the authors do not really discuss the two extra dd peaks observed in La3Ni2O7 that are distinct from NdNiO3. I see that these two dd peaks are discussed in more details in the Supplementary when explaining the theoretical simulations. Therefore, I suggest that the authors move this discussion from the Supplementary to the main article, as these two dd peaks seem to be important fingerprints for distinguishing the electronic structure of La3NiO2O7 compared to other nickelate phases.

5. In paragraph 1 of "Magnetic Excitations" section, the authors state that:
"The excitations reach maximal energy of about 70 meV at (0, 0) and (0.5, 0) while soften to zero energy (within the experimental energy resolution) at (0.25, 0.25), suggesting the presence of a quasi-static order."
Why does the softening of magnon implies the existence of a quasi-static order? The authors should explain this in a greatly more details, as this is a key sentence in interpreting the main result of the paper from the RIXS data. This is what I meant when I said that the paper is written more towards fellow experts than towards a broader audience. Fellow RIXS experts may understand this point (in fact, I do), but this might not be immediately intuitive to the broader audience. The authors can for example already included the explanation when they introduce the SDW concept in the Introduction (see point 2 above), or they can also just discuss it here. The authors should also cite ample examples of where this is observed in other systems.

6. When introducing the Stripe-2 model, does this mean the valence of all sites is assumed to be 2.5+? The authors should state this more explicitly in the text.

7. When scanning the magnetic peak against L (Fig. 4e), how broad is the peak (e.g. its HWHM) as a function of L? What is the corresponding out-of-plane correlation length compared to the out-of-plane lattice constant? This quantity can establish the two-dimensionality of the spin order, so the authors should mention it explicitly here.

8. In the Discussion and Conclusion section, the authors mention that "the inter-layer effective magnetic superexchange interaction is much larger than the intra-layer ones." While I agree that this is a very interesting observation, the authors should give this more context by comparing it to the cases of infinite-layer nickelates and superconducting cuprates. For example, the in-plane J for infinite-layer nickelates is ca. 65 meV (H. Lu et al., Science 373, 213 (2021)), interestingly very similar to the Jz of La3Ni2O7 shown here.

9. The authors stipulate that the spin structure of La3NiO2O7 could be either Stripe-1, Stripe-2, or a combination of both. Realistically, how can one distinguish between the two models? The authors should give some suggestions on how these two models could be distinguished and discuss further on how these two models can lead to either a different or a similar effect on the superconductivity of the highly-pressurized state, especially as they lead to a different distribution of charges in the Ni sites.

10. The color scheme in Figure S2c currently makes it rather difficult to distinguish between the different components of the simulated XAS. The authors should change it to a color scheme with more contrast.

Reviewer #3

(Remarks to the Author)
This paper constitutes a first core-level spectroscopic study of the electronic structure and collective excitations of the La3Ni2O7 compound, which was recently synthesized and shown to exhibit high temperature superconductivity under high pressure.
The paper is very well presented and written, and constitute in my opinion a very serious work which should be published rapidly given 1) the importance of the results (e.g. direct proof of magnetic order) and 2) the 'hotness' of the topic which is attracting a lot of attention.
I would fully support the publication of the work in Nature Communications, provided that some relatively minor comments (see below) are addressed upon revision.

The presentation and the discussion of the XAS is very good and convincing, demonstrating on the basis of O K-edge and Ni L3-edge data, backed up with a cluster model calculation that the ground state La3Ni2O7 contains both 3d8 and Ligand states, and allows for a quantitative estimate of the charge-transfer gap amplitude.

The RIXS data confirm this and emphasize the importance of the bilayer structure of the compound in shaping the electronic ground state.
The RIXS data are of high quality and show beautiful dispersing excitations, demonstrated to be magnetic in nature by means of polarimetric analysis (I feel here that more references to previous polarimetric RIXS work could be given - this is not that common). The most spectacular finding is a softening at in-plane momentum of (0.25, 0.25) which indicate the condensation of the magnetic excitation in a SDW.
It is argued that this bears striking similarities with another famous nickelate, namely La3/2Sr1/2NiO4 (as a side remark, it could be useful to include in the XAS discussion a reference XAS spectrum of this material, even from the litterature), but appears to lack the corresponding charge order well known in the 214 system.
The finding is well captured by a Heisenberg model which allows a quantitative discussion of the magnetic interaction parameters. This is not obvious to me that Heisenberg model can be applied for this metallic system. This should probably be discussed. Also I wonder whether there is any information available on the magnetic moment of the Ni.
Finally, I would disagree with the authors that the correlation length of the order is 'relatively short' and would in fact challenge them to find a longer one for similar phases in the literature. Correlation length of 250 \AA correspond for instance to that reported in the stripe phase of cuprates, or in some long-range CDW phases (e.g. magnetic field of strain-induced in these materials). Furthermore it would be interesting to know what the momentum resolution of the measurement actually is.
The L-dependence is intriguing as the maximum does not seem to be at integer L, nor half integer. Could it be incommensurate? Or is this just absorption effect? This could be a bit better discussed.

Minor comments: what are 'electron holes' mentioned in the first paragraph of the main text?

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
In the revised version, the authors did some additional work to reply to my comments and I appreciate their hard efforts and valuable discussion, where the manuscript has been highly improved.

(1) Now, I understand that the magnetic coupling Js provided in the main text are coming from fitting the experimental magnon spectrum, not from DFT calculations. I also thank the authors for adding one sentence to remind the authors of this issue. Although I am still curious about why J_1 was obtained to be zero and much smaller than J_2, I know this point needs more effort and additional theoretical work to demonstrate in another study. Not every mystery can be solved in one paper. Thus, I have no objection to their response to this issue.

(2) Based on the calculated magnetic moments, it seems that their DFT calculations only obtain the tendency of E-AFM order, not the spin-density-wave nature. I understand it needs a very large supercell to get a spin-wave-density state in the E-AFM phase, which is beyond the capability of the DFT calculations in this system. However, I believe the results of the calculated magnetic moments should be put in the supplemental information. In addition, they also need to remind the readers the nature of the spin density wave was not obtained in their present DFT calculations in the portion of those results.

As I said in my previous report, the results discussed here are interesting and important for the study of bilayer nickelate superconductors. In this case, I would like to recommend its publications in Nature Communications. Before its publication, the results of the calculated magnetic moments should be included and discussed in the supplemental information.

Reviewer #2

(Remarks to the Author)
The modifications that the authors have made to the manuscript are sufficient to address the minor issues that I have raised. I have no further comment to make, and I support the publication of this manuscript in Nature Communications in its current

form.

Reviewer #3

(Remarks to the Author)
I am satisfied with the replies to my comments and the modifications made to the manuscript, which I can now recommend for publication in Nature Communications.

Point-by-point reply to peer review comments

Reviewer #1 (Remarks to the Author):

In the present work, the authors employed X-ray absorption spectroscopy (XAS) and resonant inelastic X-ray scattering (RIXS) to detailly study the newly discovered bilayer high Tc superconductor La3Ni2O7, at ambient pressure, by combining the theoretical calculations. Based on the XAS and RIXS measurements, the spin-density-wave order was revealed at (0.25, 0.25) below 150 K. In addition, two possible interesting electronic states (spin-charge stripe and double spin stripe states) were proposed to understand the results obtained from RIXS. Furthermore, they also discussed the low-energy electronic structure using XAS and RIXS. Furthermore, they also found that the inter-layer effective superexchange interaction is an order of magnitude larger than that of the intra-layer. In general, those results look interesting and important for the study of bilayer nickelate superconductors, which deserves to be published somewhere. However, the theory component has some unclear issues.

Reply: We thank the referee for the positive evaluation and endorsement and apologize for the complex description of the theoretical part. We want to emphasize that $La_3Ni_2O_7$ is metallic with a spin density wave. As shown in the main text, we used an effective Heisenberg model for the spin dynamics. The exchange couplings J here should be considered as the Weiss molecular field for a spin density wave. To avoid confusion, we have rephrased this part in the main text and added the explanation at the beginning of Section 6 in Supplemental Material. Formally, we can write down a semiclassical equation for spin dynamics (Gia-Wei Chern et al., Phys.Rev.B.97, 035120, 2018)

$$\frac{\partial S_i}{\partial t} = -S_i \times \frac{\langle \hat{H}_{SDW} \rangle}{\partial S_i} \tag{1}$$

Here, since the $\hat{H}_{SDW}$ dynamics is complicated with two-layer, 4-sites, and 2-orbital model, we replace the $\hat{H}_{SDW}$ by the effective Heisenberg model for simplicity and leave the full consideration in the future.

(1) In the double spin stripe state (panel b of Fig. 3), they said a calculated magnetic coupling J1 to be zero. How can J1 be zero? From the geometrical perspective, the x2-y2 orbital overlaps with the px or py states in the plane and then overlaps with the next x2-y2 orbital, which is simply impossible to be zero. For what physical reason will J1 be 0 here?

Reply: Sorry for the confusion here. We did not calculate any J values. J is obtained from fitting the experimental magnon spectrum, governing the spin dynamics. Strictly speaking, to obtain any J, one should treat them from a local spin perspective. As we stated above, the fitted J values in our work should be considered as the Weiss molecular field. In the revised manuscript, we have added the fitting error bar into Fig.3. We also clarified that J_1S in Stripe-2 should be viewed as negligibly weak exchange interaction in comparison to the dominant inter-layer exchange interaction leading to a similar spin dynamics and magnon dispersion as in Stripe-1.

(2) To obtain the magnetic coupling Js, the authors mapped DFT energy to the Heisenberg model. Have they tried other states to obtain magnetic coupling Js? The energy differences for those configurations look to be quite large at U = 3 and 4 eV. To map the DFT energy to the Heisenberg model to obtain J, it is better to use the magnetic states with closed energies.

Reply: We didn't map the DFT energy to the Heisenberg model. All the couplings are from fitting as we stated above.

(3) What are the calculated magnetic moments for different Ni sites of those stripes? Furthermore, the energy difference between the Stripe-2 and Stripe-3 states suddenly decreases at U = 4.0 eV. Why? Could they provide some explanations?

Reply: We thank the referee for carefully reading the manuscript. The calculated magnetic moments are listed in the table below. Although there are non-magnetic sites (black in Fig.S10(c)) for the initial inputs, the pure charge sites develop small non-zero values after self-consistent calculation for $U > 0$. Therefore, we list them by the slash line in the table. The majority spin moment competes with the minority spin moment in the large U leading to a decreased minority spin moment in U=4. Hence, there is an obvious energy change in U=4. We also want to emphasize that DFT+U always overestimates the magnetic moments. But the tendency of ordering at Q=$(\pi/2, \pi/2)$ is consistent with RIXS observations.
Experimentally, the studies of magnetic moment show some contradictory results. NMR estimate that the magnetic moment at Ni sites, are ∼0.08 μ_B for Ni with inner apical oxygen and 0.018 μ_B for Ni without inner apical oxygen (Dan et al., arXiv:2402.03952). The recent μSR work estimate that the magnetic moment is 0.48-0.67 μ_B if the moment points in the ab-plane, while 0.28-0.31 μ_B if the moment points parallel to the c-axis (Khasanov et al., arXiv:2402.10485). We have added the information of the magnetic moment in the Introduction section.

	U=0	U=1	U=2	U=3	U=4
Stripe-1	0.806	1.08	1.264	1.374	1.493
Stripe-2	0.716	0.941	1.076	1.185	1.282
Stripe-3	0.699/0.0	0.971/0.444	1.163/0.459	1.294/0.473	1.379/0.338
G-AFM	0.517	0.748	0.917	1.061	1.194

(4) The E-phase is well-known for decades in manganates and is also found in iron Te-based superconductors. Could the author briefly discuss the connections between those three families? Is the E-AFM phase discussed in the present work an insulating state here, as FeTe and manganates? What does the band structure of the E-phase (Stripe-2) look like?
Reply: We thank the referee for pointing out the interesting question. In terms of the magnetic structure, both FeTe and some manganites adopt the double-stripe phase known as the E-AFM phase although manganites display more complex phase diagram owing to the coupled spin-charge-orbital degrees of freedom. However, the magnetic moment of the latter two cases are more localized than that found in $La_3Ni_2O_7$ which experimentally is still controversial as stated above. Our calculated band structure of $La_3Ni_2O_7$ (shown in Fig.R1) under both U = 0 and U = 4 eV suggest the system is metallic consistent to the experimental observation. We believe their detailed connection is interesting and deserves further exploration.

Reviewer #2 (Remarks to the Author):

In this work, the authors presented the electronic and magnetic structures of the ambient-pressure normal-state of the recently-discovered La3Ni2O7 high-temperature superconductor. These are probed with a high level of details using x-ray absorption (XAS) and polarimetry-analysed resonant inelastic x-ray scattering (RIXS).

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Therefore, I think this is a unique and valuable contribution to the overall understanding of not only the La3Ni2O7 superconductor itself but also to the study of superconductivity in strongly-correlated materials in general.

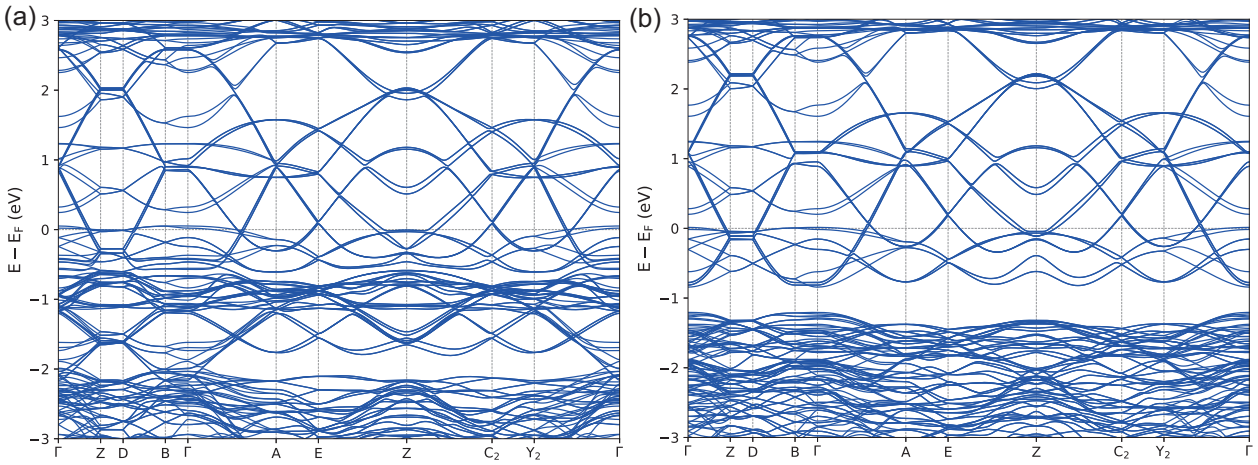

Figure R1: The band structures of Stripe-2 (E-type) phase from DFT+U calculations. (a) U=0 eV. (b) U=4 eV.

Reply: We thank the Referee for their very positive view on the unique and valuable contribution to the understanding of La3Ni2O7 superconductor and the superconductivity in strongly-correlated materials in general.

However, I think the way that the manuscript is written is still too focused towards fellow experts who are already familiar with the topics of SDW, superconductivity, and x-ray spectroscopy. There are a lot of instances in the manuscript where several specific terms that are important for understanding the results and analysis are not adequately explained and discussed.

Thus, before I can support the publication of this manuscript, I suggest that in general the authors should revise the manuscript to be more suited to the broad audience of Nature Communications, especially in the Introduction where several of these key terms can be more adequately introduced.

For example, below are several places where the manuscript could be improved:

1. The sentence: "Moreover, the bilayer structure of La3Ni2O7 may suggest a distinct minimal model in comparison to cuprate superconductors." in the abstract is confusing and hard to understand, especially the term "distinct minimal model". The authors should find a better way to highlight the difference between La3Ni2O7 and the cuprate superconductors.

Reply: This is a valid point. We now change the sentence to "Specifically, the bilayer structure of $La_3Ni_2O_7$ and the mixed valence state of Ni may promote a multi-orbital model instead of a single-orbital model widely adopted in cuprate superconductors.".

2. As the SDW is the main result of this paper, the authors should explain the concept of SDW in the introduction section, dedicating perhaps 1-2 paragraphs to introduce this concept to a general audience and its relationship and importance to superconductivity in strongly-correlated materials. This is so that the audience could more easily follow the analysis later in the results and discussion section.

Reply: We thank the Referee for the useful advice. The introduction section has now been substantially improved to aid the readers for a better understanding of the key results.

3. I found that the three papers cited by the authors to support prior existence of SDW in La3Ni2O7 (Ref. 25,26,27) to not really be convincing, especially as two of them are still not peer-reviewed. A kink in electrical resistivity can easily be attributed to other explanations, uSR is a local technique and thus cannot distinguish between different types of long-range orders, and the

NMR results support CDW scenario but not really SDW. So I think it is a bit disingenous to say that SDW in La3Ni2O7 has been supported by other results, as paragraph 2 seems to imply.

Reply: We thank the Referee for the critical comment and agree that a kink in electrical resistivity reflects a transition involving possibly charge or spin order. The cited NMR work (Ref.27) suggest that the NMR peak broadening can be attributed to the CDW order but cannot be reproduced by SDW alone indicating the possible coexistence of CDW and SDW. A more recent NMR study on La3Ni2O7 single crystal found evidence of SDW with a transition temperature ∼150K. With regards to uSR, we would respectfully highlight that muons carry a S = 1/2 spin and the technique is exclusively sensitive to magnetism. The cited uSR work (Ref.26) has been accepted in Phys. Rev. Lett. A more recent uSR study on La3Ni2O7 also revealed static magnetic order below T ∼150K (R. Khasanov et al. arXiv:2402.10485). Nevertheless, we take the opportunity to modify the relevant text to make a more accurate quotation of the prior works.

4. When discussing the RIXS results, the authors do not really discuss the two extra dd peaks observed in La3Ni2O7 that are distinct from NdNiO3. I see that these two dd peaks are discussed in more details in the Supplementary when explaining the theoretical simulations. Therefore, I suggest that the authors move this discussion from the Supplementary to the main article, as these two dd peaks seem to be important fingerprints for distinguishing the electronic structure of La3NiO2O7 compared to other nickelate phases.

Reply: A brief discussion of the two dd peaks distinct to that in NdNiO3 was shown at the end of the second last paragraph of the Section. To follow the suggestion made by the Referee, we relocated some of the discussion of the dd peaks from the Supplementary to the main text.

5. In paragraph 1 of "Magnetic Excitations" section, the authors state that: "The excitations reach maximal energy of about 70 meV at (0, 0) and (0.5, 0) while soften to zero energy (within the experimental energy resolution) at (0.25, 0.25), suggesting the presence of a quasi-static order." Why does the softening of magnon implies the existence of a quasi-static order? The authors should explain this in a greatly more details, as this is a key sentence in interpreting the main result of the paper from the RIXS data. This is what I meant when I said that the paper is written more towards fellow experts than towards a broader audience. Fellow RIXS experts may understand this point (in fact, I do), but this might not be immediately intuitive to the broader audience. The authors can for example already included the explanation when they introduce the SDW concept in the Introduction (see point 2 above), or they can also just discuss it here. The authors should also cite ample examples of where this is observed in other systems.

Reply: Thanks to Referee for the useful suggestion. We have added some text to elaborate the point with relevant references.

6. When introducing the Stripe-2 model, does this mean the valence of all sites is assumed to be 2.5+? The authors should state this more explicitly in the text.

Reply: For the Stripe-2, we stated that it is without the charge inhomogeneity. To make the statement more explicitly, we change it to 'with homogeneous valence state $Ni^{2.5+}$'.

7. When scanning the magnetic peak against L (Fig. 4e), how broad is the peak (e.g. its HWHM) as a function of L? What is the corresponding out-of-plane correlation length compared to the out-of-plane lattice constant? This quantity can establish the two-dimensionality of the spin order, so the authors should mention it explicitly here.

Reply: We thank the Referee for these valuable suggestions. Here we plot the L dependence of the SDW scattering peak in Figure R2. Its intensity forms a broad peak along L with an HWHM of approximately 0.3 r.l.u., corresponding to a correlation length of about 0.2 nm. Combining with the result of the in-plane correlation length of 27.7 nm, we show that the SDW is two-dimensional like.

8. In the Discussion and Conclusion section, the authors mention that "the inter-layer effective

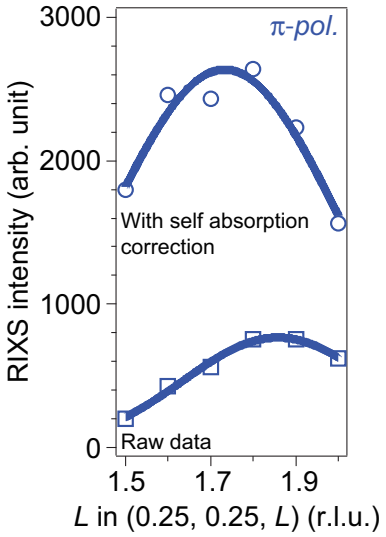

Figure R2: **The L dependence of the SDW scattering peak.** Data were collected using π-polarized photons at 20 K. SDW peak intensity is integrated over an energy window of 36.5 meV (the total energy resolution). The hollow square markers represent the raw data without self-absorption correction, while the hollow circle markers represent the corresponding corrected data. The fitted HWHMs (Γ) are 0.26 r.l.u. and 0.32 r.l.u. for the raw and corrected data, respectively. Correspondingly, the calculated correlation lengths (ξ_L) are 0.23 nm and 0.19 nm for the raw and corrected data, respectively ($\xi_L = 1/\Gamma$).

magnetic superexchange interaction is much larger than the intra-layer ones." While I agree that this is a very interesting observation, the authors should give this more context by comparing it to the cases of infinite-layer nickelates and superconducting cuprates. For example, the in-plane J for infinite-layer nickelates is ca. 65 meV (H. Lu et al., Science 373, 213 (2021)), interestingly very similar to the Jz of La3Ni2O7 shown here.

Reply: We thank the Referee for the useful suggestion. The comparison with multilayer cuprates (*e.g.*, $Y_2BaCu_3O_{6+d}$) and multilayer nickelates with reduced valence states (*e.g.*, $La_4Ni_3O_8$) is provided at the end of the first paragraph in Discussion. With regards to the comparable superexchange strength between Jz of $La_3Ni_2O_7$ and in-plane J of $NdNiO_2$, we view this is rather coincidental. Similar in-plane J is also found in $La_4Ni_3O_8$.

9. The authors stipulate that the spin structure of La3NiO2O7 could be either Stripe-1, Stripe-2, or a combination of both. Realistically, how can one distinguish between the two models? The authors should give some suggestions on how these two models could be distinguished and discuss further on how these two models can lead to either a different or a similar effect on the superconductivity of the highly-pressurized state, especially as they lead to a different distribution of charges in the Ni sites.

Reply: Thanks to the Referee for the excellent question. Elucidating the magnetic structure of 3d transition metal oxides can be done by the resonant soft X-ray scattering. The 3d states are directly probed by the dipole 2p to 3d transitions L edges, leading to very strong enhancements in the scattered intensity. Successful experiments have been done on studying the magnetic and orbital ordering in manganites and nickelates (S.B. Wilkins et al., PRL 90, 187201, 2003; S.B. Wilkins et al., PRL 91, 167205, 2003; K. J. Thomas et al., PRL 92, 237204, 2004; V. Scognoli et al., PRB 73, 100409, 2006). We have planned the experimental verification of the magnetic

structure of $La_3Ni_2O_7$ which is underway. With regards to the effect on the superconductivity, theoretical studies taking the viewpoint of the strong interlayer hybridisation suggest either d-wave or the $(d + is)$-wave pairing symmetry with the latter having a dominant d-wave component (F. Lechermann et al., PRB 108, L201121, 2023; K. Jiang et al., CPL 41, 017402, 2024). We speculate that the Stripe-1, as being close to the strong correlation picture, will likely host d-wave dominant pairing symmetry. While for the Stripe-2, which is close to weaker correlation picture such as iron-base superconductors, random-phase-approximation based studies predict that $La_3Ni_2O_7$ host s±-wave pairing symmetry. Relavant discussions are now added into the Discussion section.

10. The color scheme in Figure S2c currently makes it rather difficult to distinguish between the different components of the simulated XAS. The authors should change it to a color scheme with more contrast.

Reply: We thank the Referee for their good suggestions. Here we display the revised Figure S2c as Figure R3 and which is also updated in the Supplementary Material.

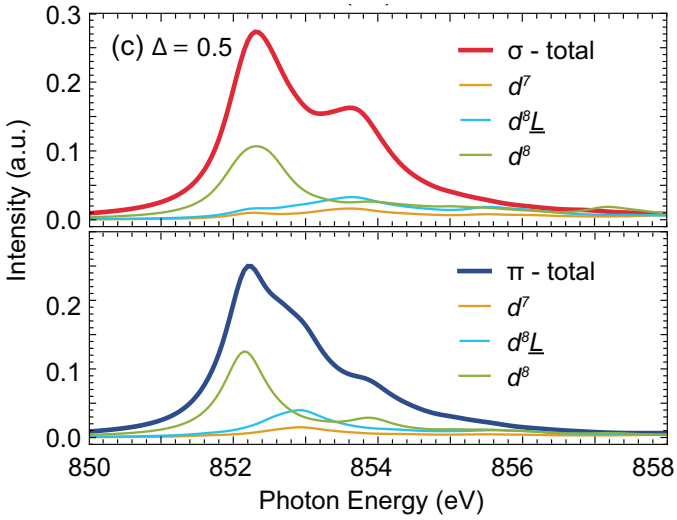

Figure R3: **The revised Fig.S2c.**

Reviewer #3 (Remarks to the Author):

This paper constitutes a first core-level spectroscopic study of the electronic structure and collective excitations of the La3Ni2O7 compound, which was recently synthesized and shown to exhibit high temperature superconductivity under high pressure. The paper is very well presented and written, and constitute in my opinion a very serious work which should be published rapidly given 1) the importance of the results (e.g. direct proof of magnetic order) and 2) the 'hotness' of the topic which is attracting a lot of attention. I would fully support the publication of the work in Nature Communications, provided that some relatively minor comments (see below) are addressed upon revision.

Reply: We thank the Referee for the recognition of our work and the suggestion for a rapid publication in Nature Communications.

The presentation and the discussion of the XAS is very good and convincing, demonstrating on the basis of O K-edge and Ni L3-edge data, backed up with a cluster model calculation that the ground state La3Ni2O7 contains both 3d8 and Ligand states, and allows for a quantitative estimate

of the charge-transfer gap amplitude.

The RIXS data confirm this and emphasize the importance of the bilayer structure of the compound in shaping the electronic ground state. The RIXS data are of high quality and show beautiful dispersing excitations, demonstrated to be magnetic in nature by means of polarimetric analysis (I feel here that more references to previous polarimetric RIXS work could be given - this is not that common).

Reply: We thank the Referee for the useful suggestion. Some extra references of polarimetic resonant X-ray scattering studies are now added to the relevant part in the main text.

The most spectacular finding is a softening at in-plane momentum of (0.25, 0.25) which indicate the condensation of the magnetic excitation in a SDW. It is argued that this bears striking similarities with another famous nickelate, namely La3/2Sr1/2NiO4 (as a side remark, it could be useful to include in the XAS discussion a reference XAS spectrum of this material, even from the litterature), but appears to lack the corresponding charge order well known in the 214 system.

Reply: Following the Referee's suggestion, we extracted and added the O K- and Ni L_3- XAS of $Nd_{3/2}Sr_{1/2}NiO_4$ into the Fig.2d (Z. Hu et al., PRB 61, 3739, 2000) (here displayed in Figure R4) (No published XAS spectra were found about $La_{3/2}Sr_{1/2}NiO_4$ system). Concerning the charge order, an earlier neutron scattering study on $La_{3/2}Sr_{1/2}NiO_4$ reported the observation with an incommensurability twice as large as that of the spin order confirming the spin-charge stripe order (R. Kajimoto et al., PRB 67, 014511, 2003). The reference is now cited.

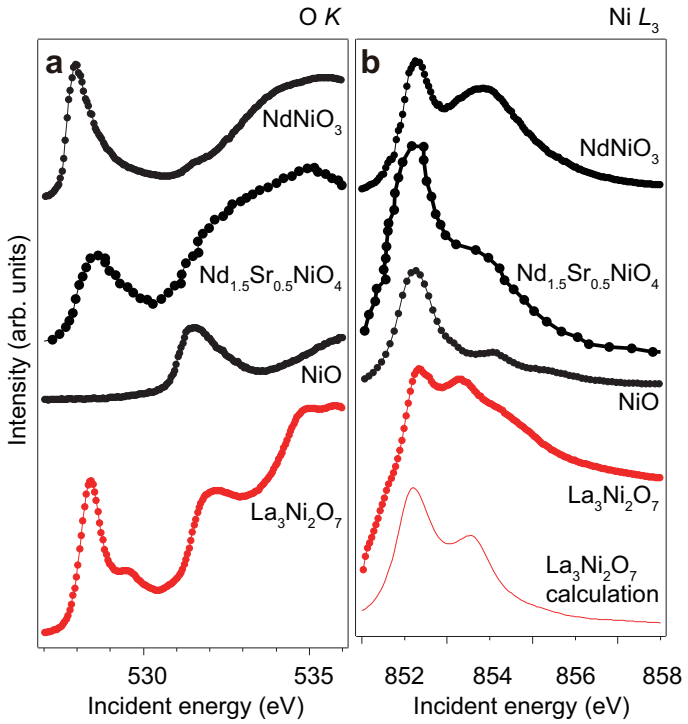

Figure R4: **The revised Fig.1d.**

The finding is well captured by a Heisenberg model which allows a quantitative discussion of the magnetic interaction parameters. This is not obvious to me that Heisenberg model can be applied for this metallic system. This should probably be discussed. Also I wonder whether there is any information available on the magnetic moment of the Ni.

Reply: We thank the referee for this important question. As we stated in the manuscript and

rephrased in the supplementary material, the effective exchange coupling here should be considered as the Weiss molecular field for a spin-density wave governing the spin dynamics for this metallic system. We further emphasize this point in the revised manuscript. On the other hand, this effective Heisenberg approach has provided some qualitative understanding for the fitting of the magnon dispersion of the metallic infinite-layer nickelates (H. Lu et al., Science 373, 213 (2021)) and the doped superconducting cuprates.
Experimentally, the studies of magnetic moment show some contradictory results. NMR estimate that the magnetic moment at Ni sites, are ∼0.08 μ_B for Ni with inner apical oxygen and 0.018 μ_B for Ni without inner apical oxygen (Dan et al., arXiv:2402.03952). The recent μSR work estimate that the magnetic moment is 0.48-0.67 μ_B if the moment points in the ab-plane, while 0.28-0.31 μ_B if the moment points parallel to the c-axis (Khasanov et al., arXiv:2402.10485). We have added the information of the magnetic moment in the Introduction section.

Finally, I would disagree with the authors that the correlation length of the order is 'relatively short' and would in fact challenge them to find a longer one for similar phases in the literature. Correlation length of 250 Å correspond for instance to that reported in the stripe phase of cuprates, or in some long-range CDW phases (e.g. magnetic field of strain-induced in these materials). Furthermore it would be interesting to know what the momentum resolution of the measurement actually is.

Reply: We thank the Referee for pointing out our misstatement. In fact, the correlation of 250 $\mathring{A}$ is comparable to that of long-range charge order in 1/8 doping $La_{7/8}Ba_{1/8}CuO_4$ (∼200 Å) (S.B. Wilkins et al., PRB 84, 195101, 2011). The momentum resolution of the measurement is 0.002 r.l.u. The information is added into the main text and the Methods, respectively.

The L-dependence is intriguing as the maximum does not seem to be at integer L, nor half integer. Could it be incommensurate? Or is this just absorption effect? This could be a bit better discussed.

Reply: We thank the Referee for the insightful comments. To address the concern, we explicitly plot the intensity of the SDW scattering peak as a function of L (Figure. R2). The L-dependence is weak with a correlation length ξ_L of approximately 0.2 nm, showing the two-dimensional nature of the SDW. We also note that the maximum intensity can change with or without considering the absorption effect, meaning it plays a role in the measured intensity. Considering such a weak L-dependence and the strong absorption effect, it is insufficient to claim such SDW is commensurate or incommensurate.

Minor comments: what are 'electron holes' mentioned in the first paragraph of the main text?

Reply: We referred to the doped hole charges in the hole-doped cuprates. The text is superseded now as the Introduction is revised to suit the general readership of Nature Communications.

Point-by-point reply to peer review comments

Reviewer #1 (Remarks to the Author):

In the revised version, the authors did some additional work to reply to my comments and I appreciate their hard efforts and valuable discussion, where the manuscript has been highly improved.

(1) Now, I understand that the magnetic coupling Js provided in the main text are coming from fitting the experimental magnon spectrum, not from DFT calculations. I also thank the authors for adding one sentence to remind the authors of this issue. Although I am still curious about why J_1 was obtained to be zero and much smaller than J_2, I know this point needs more effort and additional theoretical work to demonstrate in another study. Not every mystery can be solved in one paper. Thus, I have no objection to their response to this issue.

(2) Based on the calculated magnetic moments, it seems that their DFT calculations only obtain the tendency of E-AFM order, not the spin-density-wave nature. I understand it needs a very large supercell to get a spin-wave-density state in the E-AFM phase, which is beyond the capability of the DFT calculations in this system. However, I believe the results of the calculated magnetic moments should be put in the supplemental information. In addition, they also need to remind the readers the nature of the spin density wave was not obtained in their present DFT calculations in the portion of those results.

As I said in my previous report, the results discussed here are interesting and important for the study of bilayer nickelate superconductors. In this case, I would like to recommend its publications in Nature Communications. Before its publication, the results of the calculated magnetic moments should be included and discussed in the supplemental information.

Reply: We thank Reviewer #1 for his/her appreciation of our efforts to address the comments.
(1) Following Reviewer #1's advice, We have clarified in the revised manuscript that "All JS values are fitted to the experimental magnon dispersion by solving the semiclassic torque equations." We agree that further theoretical work is required to fully understand the near-zero J1, which may be explored in future studies.
(2) Following Review #2's suggestion, we have added the results of calculated magnetic moments in Supplementary Information, including a reminder that the spin-density-wave nature was not captured in our present DFT calculations due to computational limitations, as well as a brief discussion of the calculated magnetic moments with experimental values.

Reviewer #2 (Remarks to the Author):

The modifications that the authors have made to the manuscript are sufficient to address the minor issues that I have raised. I have no further comment to make, and I support the publication of this manuscript in Nature Communications in its current form.

Reply: We appreciate Reviewer #2's recognition of our efforts to address the comments and are grateful for your recommendation for publication.

Reviewer #3 (Remarks to the Author):

I am satisfied with the replies to my comments and the modifications made to the manuscript, which I can now recommend for publication in Nature Communications.

Reply: We appreciate Reviewer #3's recognition of our efforts to address the comments and are grateful for your recommendation for publication.