nature portfolio

Peer Review File

Electronic correlations and partial gap in the bilayer nickelate La3Ni2O7

Editorial Note: This manuscript has been previously reviewed at another journal that is not operating a transparent peer review scheme. This document only contains reviewer comments and rebuttal letters for versions considered at *Nature Communications.*

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

I appreciate the authors' effort to improve their manuscript significantly, carefully considering reviewers' comments. Most of their responses to reviewers' questions and comments are reasonable, solid, and concrete. However, one question still requires clarification at this stage.

1. The authors revised the Drude-Lorentz analysis and decomposed the measured optical conductivity into a single Drude component and multiple Lorentz components. Considering the computed electronic structure of La3Ni2O7, which shows the multiorbital character with Ni dz2 and dx2-y2 orbitals crossing the Fermi level, it is expected to observe two Drude components due to intraband transitions within the two orbital characters. I guess that it is a matter of how fitting the authors did. When attempting to fit the measured optical conductivity with two Drude components, it would yield the best match, similar to the single Drude fitting.

Given the distinct correlation strengths for Ni dz2 and dx2-y2, supported by the evidence of a density-wave-like transition being responsible for the Ni dz2, it follows that scattering rates for the two Ni orbitals should differ. Consequently, the use of two Drude components in the fitting process presents a more physically reasonable representation compared to the single Drude fitting.

Except for the aforementioned criticism, the other aspects are clear and transparent. Therefore, if the authors provide reasonable answers to the issue raised, I will recommend the current work for publication in Nature Communications.

Reviewer #2 (Remarks to the Author):

The authors revised their manuscript based on the comments by the three reviewers. Now in the revised version, the two-Drude interpretation is changed, which previously served as evidence for the importance of the Ni-dz^2 orbitals because it is interpreted that this is the orbital that is gapped out below the critical temperature. Now there is only one Drude contribution, which seems to be enough to reproduce the data set. This new interpretation makes the current work even more just an optical characterization of a new compound correlated compound. Pretty much the only conclusion that can be obtained from the study is that the compound is indeed correlated and consistent with the literature and ARPES studies, which compare their band structure with DFT+U suggesting the correlated nature of the compound. Overall, the current study does not provide any new insight into the compound except what is already pointed out by other studies. And I have to say even the confirmation at times is not very convincing. Therefore, I cannot recommend this work for publication in Nature Communications. Below are some of my concerns:

1. Despite the single Drude contribution being given now, the multi-band nature of the compound suggests that this single Drude adds up from the different contributions of hole and electron bands with similar scattering rates. I think this is a reasonable assumption. However, now from this study, it is even more ambiguous which orbital is more important and eventually gapped out below the critical temperature. According to the authors' fat-band plots, potential orbital giving rise to this Drude seems to be equally dominating the S(R)-point.

2. Authors never show the temperature-dependence of the reflectivity and optical conductivity at high-energy regions. I think this might be important to see what are the potentially changing contributions. As one can easily imagine the increase in interband transitions around 1000 cm^-1 is due to the changes in some other high-energy bands and the low-energy region is just the sharpening of the Drude contribution.

3. I still think the density wave behavior in optical conductivity is very weak. What is the error bars in this difference plot? Authors compare the spectra with CsV3Sb5, but there the clear spectral weight transfer is demonstrated with the significant intensity. Furthermore, the gap at the FS around M-point is determined with ARPES along with the modulated 2x2 structure below the critical temperature. I fail to see what constitutes this behavior as a density wave in the current compound. Except ARPES sees that the flat bands around gamma-point are below the Fermi energy (given at 18 K, reference 40 of the manuscript), which might suggest that above a certain temperature it was above the Fermi energy and simply what optics sees is that the shift of the Fermi energy. And of course, once the band disappears from the Fermi surface the carrier density decreases (effect in plasma frequency). The authors also added this discussion as they also pointed out that it is plausible. So what is the distinguishing effect?

Reviewer #3 (Remarks to the Author):

In a previous report, I had offered to review this manuscript again provided there is an independent, peer-reviewed verification of superconductivity at 80 K under high pressure in La3Ni2O7. The authors of the manuscript have now cited three preprints on the arXiv (probably from independent research groups) that report the observation of possible superconductivity in La3Ni2O7 under high pressures. The preprints on the arXiv do not reach the level of a refereed publication. However, given the multiplicity of reports, I am beginning to consider that superconductivity at 80 K under high pressure in La3Ni2O7 may be a real effect. Hence, I have decided to review the present manuscript. Although the manuscript focuses on the ambient pressure, non-superconducting state, the information about electronic correlations and charge dynamics contained in it may contribute towards a better understanding of the superconducting state under high pressure.

The other two referees have given detailed comments on this work and the authors have provided detailed responses. I will let the other referees determine the adequacy of the authors' responses to their comments. Here I will focus on the overall significance of the revised manuscript under consideration, and end with a couple of comments for the authors. Electronic correlations are present in the cuprate and iron-based high-

temperature superconductors. Thus, the presence of electronic correlations in the nickelate La3Ni2O7, as demonstrated in the present work, is of significance. The analysis of the optics data leads to plausible conclusions regarding charge dynamics and a possible (charge) density wave phase transition. In general, analysis of optics data for a multiband system with both intraband and low-lying interband optical transitions is rarely straightforward, and the authors have done a good job of analyzing and interpreting the data. Given the recent interest in this material, the timely nature of this combined experimental and theoretical work, and the importance of the results about this material's electronic properties, I recommend publication in Nature Communications. I suggest the authors consider my comments below and make appropriate changes to the manuscript and/or supplementary information.

1. I find it odd and interesting that this material is so strongly correlated that it is close to the Mott insulating state (as in Fig. 2d). I wonder if the DFT calculations are overestimating the plasma frequency and that the calculated plasma frequency may depend on the "flavor" of DFT being used? On the other hand, if this result is robust, then perhaps the high pressures reduce the strength of correlations allowing superconductivity to occur, as in the cuprates. This may also imply that the CDW is driven by strong correlations and that the CDW is suppressed at high pressures due to relative weakening of strong correlations.

2. If possible, can the authors place data points in Fig. 2d for other superconducting nickelates they mention in the introductory part of the manuscript?

3. Temperature-dependent, optical spectral weight shifts are difficult to quantify accurately with reflectance data alone. In particular, spectral weight should be conserved in the CDW phase transition, and this is not obvious from the presented data. In future, temperature dependent ellipsometry measurements will be needed to accurately quantify spectral weight redistribution due to CDW.

Response to the Report of Reviewer #1 – NCOMMS-23-57046-T

I appreciate the authors' effort to improve their manuscript significantly, carefully considering reviewers' comments. Most of their responses to reviewers' questions and comments are reasonable, solid, and concrete. However, one question still requires clarification at this stage.

We would like to thank Reviewer #1 for the careful review and high evaluation of the revised manuscript and our responses to reviewers' comments. We also thank the referee for pointing out the remaining issue in our manuscript. Let's address it below.

1. The authors revised the Drude-Lorentz analysis and decomposed the measured optical conductivity into a single Drude component and multiple Lorentz components. Considering the computed electronic structure of La3Ni2O7, which shows the multiorbital character with Ni dz2 and dx2-y2 orbitals crossing the Fermi level, it is expected to observe two Drude components due to intraband transitions within the two orbital characters. I guess that it is a matter of how fitting the authors did. When attempting to fit the measured optical conductivity with two Drude components, it would yield the best match, similar to the single Drude fitting. Given the distinct correlation strengths for Ni dz2 and dx2-y2, supported by the evidence of a density-wave-like transition being responsible for the Ni dz2, it follows that scattering rates for the two Ni orbitals should differ. Consequently, the use of two Drude components in the fitting process presents a more physically reasonable representation compared to the single Drude fitting.

We thank the referee for this great suggestion which helped to make our data analysis more physically reasonable. We completely agree with the referee that the fit with two Drude components would not only yield the best match similar to the single Drude fitting, but also presents a more physically reasonable representation compared to the single Drude fitting.

Following this suggestion, we replaced the single Drude with two Drude components to fit the measured optical conductivity $\sigma_1(\omega)$ at 150 K, and also achieved a very good fitting result (Fig. R1a). The application of the two-Drude fitting to the measured $\sigma_1(\omega)$ at all temperatures returns the T dependence of the Drude parameters (Figs. R1b-e). Figs. R1b and R1c depict the T dependence of the weight for D1 ($\omega^2_{p,D1}$) and D2 ($\omega^2_{p,D2}$), respectively. While $\omega^2_{p,D1}$ exhibits no evident anomaly, $\omega^2_{p,D2}$ is suddenly suppressed below $T^* \simeq 115$ K and vanishes quickly with decreasing temperature. This implies that the Fermi surface is partially removed below the transition at T^*. Figs. R1d and R1e plot the quasiparticle scattering rate of D1 ($1/\tau_{D1}$) and D2 ($1/\tau_{D2}$) as a function of T, respectively. It is worth noting that $1/\tau_{D1}$ follows a quadratic T dependence $1/\tau_{D1} \propto T^2$, i.e. Fermi-liquid behavior in a broad temperature range, whereas $1/\tau_{D2}$ varies linearly with temperature $1/\tau_{D2} \propto T$, which is the well-known non-Fermi-liquid behavior. These observations suggest that different Fermi surfaces in $La_3Ni_2O_7$ exhibit distinct electronic properties, and the portion exhibiting non-Fermi liquid behavior is removed due to the transition at T^*, leaving the portion characterized by Fermi liquid behavior to dominate the charge dynamics below T^*.

Note that the use of two Drude components in the fitting process does not change the conclusion, but gives a more physically reasonable description of the system.

Finally, we would like to mention that changing from the single-Drude fit to the two-Drude fit has no influence on the determination of the kinetic energy ratio K_{exp}/K_{band} (determined

above T^*), because $\omega^2_{p,D} = \omega^2_{p,D1} + \omega^2_{p,D2}$. Here, $\omega^2_{p,D}$ is the Drude weight for the single-Drude fitting; $\omega^2_{p,D1}$ and $\omega^2_{p,D2}$ represent the weight of D1 and D2 in the two-Drude fitting, respectively.

In the revised manuscript, we have replaced the single Drude component with two Drude components in the fit and updated all relevant figures and text [Fig. 2a; Fig. 3a-d; Supplementary Table 1; Supplementary Fig. 2 and Supplementary Fig. 6; Page 5, Line 93-94 Page 8, Line 151-162].

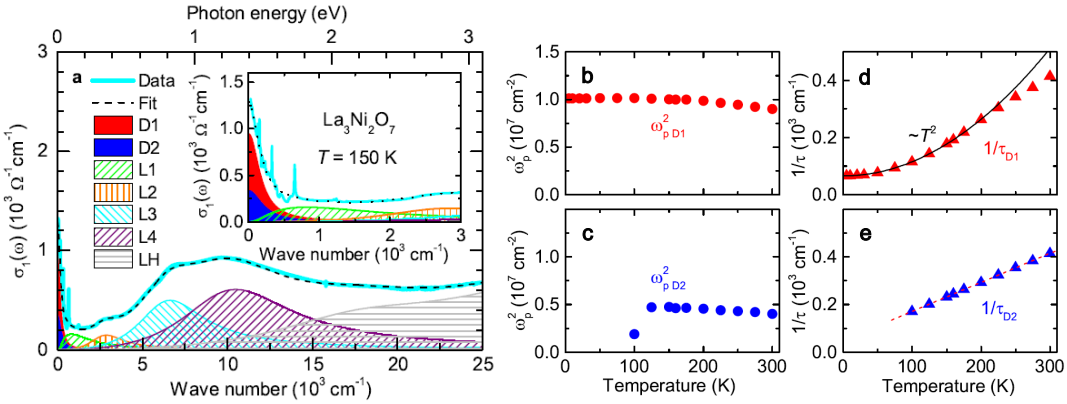

Figure R1. **a** The fit to the measured $\sigma_1(\omega)$ using two Drude components. **b**-**e** The temperature dependence of the Drude parameters extracted from the Drude-Lorentz fit.

Except for the aforementioned criticism, the other aspects are clear and transparent. Therefore, if the authors provide reasonable answers to the issue raised, I will recommend the current work for publication in Nature Communications.

We thank Reviewer #1 again for two rounds of careful reviews of our manuscript and providing so many professional comments and suggestions, which have helped significantly improve the quality of our manuscript. We hope now the referee finds our manuscript ready for publication.

Response to the Report of Reviewer #2 – NCOMMS-23-57046-T

The authors revised their manuscript based on the comments by the three reviewers. Now in the revised version, the two-Drude interpretation is changed, which previously served as evidence for the importance of the Ni-dz^2 orbitals because it is interpreted that this is the orbital that is gapped out below the critical temperature. Now there is only one Drude contribution, which seems to be enough to reproduce the data set. This new interpretation makes the current work even more just an optical characterization of a new compound correlated compound. Pretty much the only conclusion that can be obtained from the study is that the compound is indeed correlated and consistent with the literature and ARPES studies, which compare their band structure with DFT+U suggesting the correlated nature of the compound. Overall, the current study does not provide any new insight into the compound except what is already pointed out by other studies. And I have to say even the confirmation at times is not very convincing. Therefore, I cannot recommend this work for publication in Nature Communications. Below are some of my concerns:

We thank Referee #2 for their careful review of our revised manuscript. Actually, Reviewer

#1 also suggested the use of two Drude components for the fit. Following the reviewers' suggestions, we have replaced the single Drude with two Drude components in the fit (see our reply to Reviewer #1 for details), and found that the use of two Drude components not only yield the best match, but also presents a more physically reasonable representation. The two Drude components exhibit different temperature-dependent behavior (Fermi liquid vs non-Fermi liquid behavior), which allows us to distinguish different bands or orbitals. See our response to the following comment (Comment 1) for details.

In addition, we would like to clarify the novelty and significance of our work.

Novelty: Our work reports ***the first experimental*** determination of the electronic correlations (K_{exp}/K_{band}) in $La_3Ni_2O_7$ (arXiv:2307.02950; 6 Jul. 2023), which is two months earlier than the ARPES paper (arXiv:2309.01148; 3 Sep. 2023). Our study also reveals ***for the first time*** that a large portion of the Fermi surface is removed below the transition at T^*.

To the best of our knowledge, the above important observations are completely new and have never been reported by other experimental studies before our work.

Significance: The determination of K_{exp}/K_{band} (electronic correlations) is of great importance in correlated electronic systems, because it is the basis of theoretical studies. For example, the determinations of K_{exp}/K_{band} in iron pnictides [Nat. Phys. 5, 647 (2009)] and nodal-line semimetals [Nat. Phys. 16, 636 (2020)] represent very important work, providing indispensable information for many theoretical and experimental studies. Our work determines K_{exp}/K_{band} in the important material $La_3Ni_2O_7$, which is also of great significance, particularly to understanding the mechanism of the unconventional superconductivity and other competing orders in nickelates.

1. Despite the single Drude contribution being given now, the multi-band nature of the compound suggests that this single Drude adds up from the different contributions of hole and electron bands with similar scattering rates. I think this is a reasonable assumption. However, now from this study, it is even more ambiguous which orbital is more important and eventually gapped out below the critical temperature. According to the authors' fat-band plots, potential orbital giving rise to this Drude seems to be equally dominating the S(R)-point.

Thank the referee for raising this question. In the revised manuscript, we have followed the suggestion from Reviewer #1, and replaced the single Drude with two Drude components to fit the measured optical conductivity (see our reply to Reviewer #1 for details). We agree with the referee that the use of two Drude components not only yields the best match, but also gives a more physically reasonable description of the system.

The key information learned from optics is that the Fermi surface portion exhibiting non-Fermi liquid behavior is gapped due to the transition at T^*, and the remaining portion shows Fermi liquid behavior. Theoretical work [1,2] has revealed pronounced orbital differentiation in $La_3Ni_2O_7$ with the Ni-$d_{3z^2-r^2}$ orbital being more strongly correlated, thus inducing spin fluctuations and non-Fermi liquid behavior. A recent ARPES study [3] has found that the Ni-$d_{3z^2-r^2}$ derived flat band γ shows much stronger electronic correlations than the Ni-$d_{x^2-y^2}$ derived bands α and β. The combination of our optical results and the above work suggests that the gapped Fermi surface below T^* corresponds to the flat hole-like band

arising from the Ni-$d_{3z^2-r^2}$ orbital, and the remaining Fermi surface consists of the broad electron-like bands near the Γ and S (R) points.

For the electron band near the S (R) point, the Ni-$d_{x^2-y^2}$ orbital has a much larger weight (0.83) than the Ni-$d_{3z^2-r^2}$ orbital (0.47). Moreover, this band is much broader than the flat Ni-$d_{3z^2-r^2}$ band near the Γ point and exhibits weaker electronic correlations [3], so it is reasonable to link it (together with the broad electron band near Γ) to the Drude component showing Fermi liquid behavior (D1).

In the revised manuscript, the above discussions can be found on Page 11.

References

[1] Lechermann et al., arXiv:2306.05121
[2] Shilenko et al., arXiv:2306.06039
[3] Yang et al., arXiv:2309.01148

2. Authors never show the temperature-dependence of the reflectivity and optical conductivity at high-energy regions. I think this might be important to see what are the potentially changing contributions. As one can easily imagine the increase in interband transitions around 1000 cm^{-1} is due to the changes in some other high-energy bands and the low-energy region is just the sharpening of the Drude contribution.

Figure R2a shows the temperature dependence of $\sigma_1(\omega)$ in the high-energy region, which reveals that at 5 K (the blue curve), the low-frequency Drude response is suppressed and the spectral weight is transferred to a broad frequency range (600–6500 cm^{-1}). The $\sigma_1(\omega)$ spectra at different temperatures converge at about 6500 cm^{-1}.

In order to further corroborates the suppression of the Drude weight and the spectral weight transfer, we examine the frequency and temperature dependence of the spectral weight which is defined as $S = \int_0^{\omega} \sigma_1(\omega) d\omega$.

(i) For the sharpening of the Drude component, the S ratio, e.g. $S(200\ \mathrm{K})/S(300\ \mathrm{K})$ (the red curve in Fig. R2b) is higher than 1 in the low-frequency range and decreases smoothly towards 1 with increasing frequency. In sharp contrast, as shown by the blue curve in Fig. R2b, $S(5\ \mathrm{K})/S(125\ \mathrm{K})$ decreases steeply to 0.8 (lower than 1) in the low-frequency range, suggesting that the Drude weight is significantly suppressed at 5 K compared to that at 125 K. In the high-frequency range, $S(5\ \mathrm{K})/S(125\ \mathrm{K})$ increases with increasing frequency and reaches 1 at about 6500 cm^{-1}, indicating that the spectral weight is conserved at about 6500 cm^{-1}.

(ii) For the Drude response sharpening, the low-frequency spectral weight, $S(T)/S(300\ \mathrm{K})$ with ω_c = 600 cm^{-1} (Fig. R2c), increases with decreasing temperature in the high-temperature range (Note: this temperature range is dominated by the sharpening of the Drude response). Below 125 K, $S(T)/S(300\ \mathrm{K})$ is suddenly suppressed, suggesting that the low-frequency Drude weight is suppressed.

In addition, the Drude-Lorentz fitting can also extract the Drude weight accurately, which also reveals a strong suppression of the Drude weight below the transition at T^* (Fig. 3a, b in the manuscript).

All the above results unambiguously suggest that the low-frequency Drude response is suppressed, and the spectral weight is transferred to a broad frequency range (600–6500 cm^{-1}).

In the revised manuscript, the temperature dependence of $\sigma_1(\omega)$ in the high-energy region has been added to Supplementary Fig. 1; the S ratio $S(200\ \mathrm{K})/S(300\ \mathrm{K})$ showing a simple sharpening of the Drude response has been added to Fig. 4a (the red curve); the S ratio $S(5\ \mathrm{K})/S(125\ \mathrm{K})$ and the temperature dependence of the low-frequency spectral weight are shown in Fig. 4a (the blue curve) and Fig. 4c, respectively, which demonstrate the strong suppression of the Drude response and the spectral weight transfer in a broad frequency range. The corresponding discussion is given in the section "Energy scale of the spectral weight redistribution".

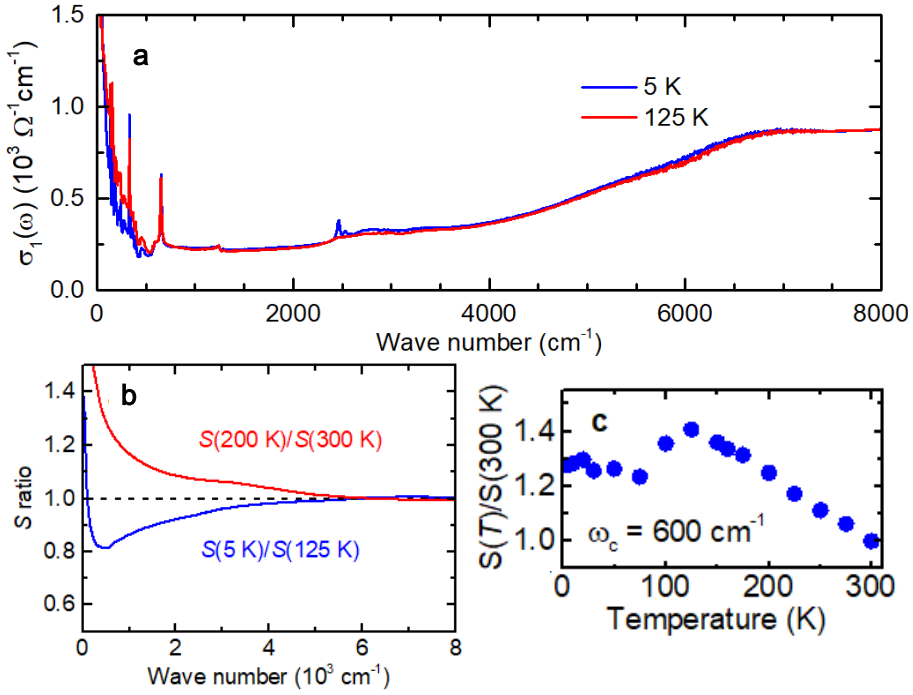

Figure R2. **a** The $\sigma_1(\omega)$ spectra of $La_3Ni_2O_7$. **b** Spectral weight ratio as a function of frequency. **c** The temperature dependence of the low-frequency spectral weight.

3. I still think the density wave behavior in optical conductivity is very weak. What is the error bars in this difference plot? Authors compare the spectra with CsV3Sb5, but there the clear spectral weight transfer is demonstrated with the significant intensity. Furthermore, the gap at the FS around M-point is determined with ARPES along with the modulated $2x2$ structure below the critical temperature. I fail to see what constitutes this behavior as a density wave in the current compound. Except ARPES sees that the flat bands around gamma-point are below the Fermi energy (given at 18 K, reference 40 of the manuscript), which might suggest that above a certain temperature it was above the Fermi energy and simply what optics sees is that the shift of the Fermi energy. And of course, once the band disappears from the Fermi surface the carrier density decreases (effect in plasma frequency). The authors also added this discussion as they also pointed out that it is plausible. So what is the distinguishing effect?

We thank the referee for these questions. Below, we address them in turn.

For our setup, the relative error in the measured reflectivity $R(\omega)$ between different temperatures is less than $\pm 0.1\%$. Based on this information, we multiply $R(\omega)$ at 5 K by 1.001 and 0.999, and then calculate the difference optical conductivity, which shows that the error bars in $\Delta\sigma_1(\omega)$ is smaller than $\pm 0.5\ \Omega^{-1}\text{cm}^{-1}$. We agree that the density-wave-like behavior in $\sigma_1(\omega)$ is very weak for $La_3Ni_2O_7$, but our optical measurements are accurate enough to detect it.

Here, we would like to mention that it is not surprising that the density-wave behavior in $\sigma_1(\omega)$ is weak. As shown in Fig. R3a, for $Lu_5Ir_4Si_{10}$ which exhibits a CDW transition at 83 K, the density-wave behavior in $\sigma_1(\omega)$ is weak and the spectral weight redistribution happens in a very broad frequency range [1], similar to what we observed in $La_3Ni_2O_7$.

After considering the referee's question, we realized that the distinguishing effect lies in the temperature dependence of the Drude weight. For a simple temperature-induced Fermi level shift, the Drude weight is expected to decrease continuously as the temperature is lowered. For example, as shown in Fig. R3b, the temperature-induced Fermi level shift leads to a continuous decrease of the Drude weight in TaAs [2]. However, in $La_3Ni_2O_7$, the Drude weight does not show a continuous decrease upon cooling, but is abruptly suppressed at the density-wave-like transition, in conflict with the temperature-induced Fermi level shift.

In the revised manuscript, we have updated the discussion about the origin of the Fermi surface reduction and spectral weight redistribution [Page 13, Line 250-257]. The reference reporting weak CDW behavior is cited as Ref. 47; the reference reporting the optical response of temperature-induced Fermi level shift is cited as Ref. 60.

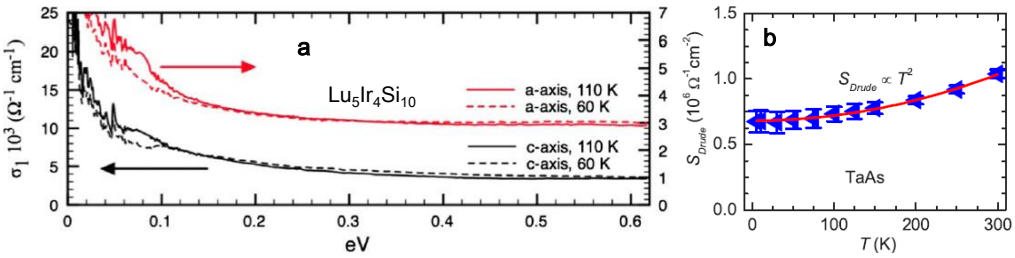

Figure R3. **a** Optical conductivity of $Lu_5Ir_4Si_{10}$ from Ref. [1]. **b** The temperature dependence of the Drude weight in TaAs from Ref. [2].

References

[1] Tediosi et al., Phys. Rev. B **80**, 035107 (2009)

[2] Xu et al., Phys. Rev. B **93**, 121110 (2016)

Response to the Report of Reviewer #3 – NCOMMS-23-57046-T

In a previous report, I had offered to review this manuscript again provided there is an independent, peer-reviewed verification of superconductivity at 80 K under high pressure in La3Ni2O7. The authors of the manuscript have now cited three preprints on the arXiv (probably from independent research groups) that report the observation of possible superconductivity in La3Ni2O7 under high pressures. The preprints on the arXiv do not reach the level of a refereed publication. However, given the multiplicity of reports, I am beginning to consider that superconductivity at 80 K under high pressure in La3Ni2O7 may be a real effect. Hence, I have decided to review the present manuscript. Although the manuscript focuses on the ambient

pressure, non-superconducting state, the information about electronic correlations and charge dynamics contained in it may contribute towards a better understanding of the superconducting state under high pressure.

> We are grateful to Reviewer #3 for making time and efforts to review our manuscript. Here, we would like to mention that one paper from an independent research group confirming the superconductivity in $La_3Ni_2O_7$ under high pressures has been published online [Wang et al., Phys. Rev. X **14**, 011040 (2024)]. We have updated this information in the revised manuscript.

The other two referees have given detailed comments on this work and the authors have provided detailed responses. I will let the other referees determine the adequacy of the authors' responses to their comments. Here I will focus on the overall significance of the revised manuscript under consideration, and end with a couple of comments for the authors. Electronic correlations are present in the cuprate and iron-based high-temperature superconductors. Thus, the presence of electronic correlations in the nickelate La3Ni2O7, as demonstrated in the present work, is of significance. The analysis of the optics data leads to plausible conclusions regarding charge dynamics and a possible (charge) density wave phase transition. In general, analysis of optics data for a multiband system with both intraband and low-lying interband optical transitions is rarely straightforward, and the authors have done a good job of analyzing and interpreting the data. Given the recent interest in this material, the timely nature of this combined experimental and theoretical work, and the importance of the results about this material's electronic properties, I recommend publication in Nature Communications. I suggest the authors consider my comments below and make appropriate changes to the manuscript and/or supplementary information.

> We thank the referee for such a careful review of our manuscript, a clear summary of the main points of work, and the high evaluation of the significance of our work. We particularly appreciate the referee's professional suggestions which were very helpful in further improving our manuscript.

1. I find it odd and interesting that this material is so strongly correlated that it is close to the Mott insulating state (as in Fig. 2d). I wonder if the DFT calculations are overestimating the plasma frequency and that the calculated plasma frequency may depend on the "flavor" of DFT being used? On the other hand, if this result is robust, then perhaps the high pressures reduce the strength of correlations allowing superconductivity to occur, as in the cuprates. This may also imply that the CDW is driven by strong correlations and that the CDW is suppressed at high pressures due to relative weakening of strong correlations.

> We appreciate these helpful questions and suggestions.
>
> Regarding the "flavor" of DFT being used, the ideas we can think of include: (i) the broadening factor; (ii) the Hubbard U; (iii) the DFT implementations; (iv) the doping effect (Fermi level shift). Let's analyze the influence of these "flavors" below.
>
> (i) The plasma frequency in Wien2k is obtained from the integral of the squared band velocities at the Fermi level [1]:
>
> $$\omega_{p,ii}^2 = \frac{4\pi e^2}{\hbar^2} \sum_n \int \frac{d\mathbf{k}}{V} v_{ii,n}^2(\mathbf{k}) \delta(\epsilon_n(\mathbf{k}) - \epsilon_F), \quad (1)$$

where ii represents xx, yy and zz; n is the band index; v and V correspond to the band velocity and the normalization volume, respectively. This equation (without broadening) directly yields $\omega_{p,xx}$ = 3.2186 eV, $\omega_{p,yy}$ = 3.2206 eV, and $\omega_{p,zz}$ = 0.3918 eV for $La_3Ni_2O_7$. Since the optical spectra were measured in the ab planes and $La_3Ni_2O_7$ crystallizes in the orthorhombic $Amam$ structure, we take $\omega^2_{p,\text{cal}} = (\omega^2_{p,xx} + \omega^2_{p,yy})/2$ and get $\omega_{p,\text{cal}}$ = 3.22 eV and K_{band} = 1.17 eV for $La_3Ni_2O_7$. Different broadening factors, such as 0.01 eV, 0.02 eV, and 0.03 eV, were used, but resulted in the same value for $\omega_{p,\text{cal}}$, suggesting that the broadening factor has no influence on the determination of $\omega_{p,\text{cal}}$. Actually, within Wien2k, as long as the band structure details do not change around the Fermi level, the resultant plasma frequency should not change.

(ii) The inclusion of Hubbard U (DFT + U), which allows for the incorporation of local Coulomb interactions for the Ni-3d orbitals, may induce small changes of the band structure, thus changing the value of $\omega_{p,\text{cal}}$. However, for a study of the electronic correlation effect in $La_3Ni_2O_7$, it is more reasonable to compare the experimental results to the calculations without including the Hubbard U. Therefore, we didn't include the Hubbard U in the DFT calculations.

(iii) Different DFT implementations do differ in several aspects when calculating optical properties, but they are expected to agree with each other in general. See Ref. [2] for benchmark results of dielectric constants calculated with different formalisms/codes. Moreover, we have calculated the plasma frequencies for $La_3Ni_2O_7$ using VASP, another widely used DFT code. The resultant values are $\omega_{p,xx}$ = 3.09 eV, $\omega_{p,yy}$ = 3.09 eV, and $\omega_{p,zz}$ = 0.37 eV, in reasonably good agreement with the results from Wien2k.

(iv) In Supplementary Note 3, we have considered a doping of 0.2 electrons per unit cell (the upper limit of self-doping induced by oxygen deficiencies), and calculated the plasma frequency $\omega_{p,\text{cal}}$ = 3.25 eV, which does not show a significant change.

In summary, since both the experimental K_{exp} and calculated K_{band} kinetic energies can be determined with reasonable accuracy, the resultant $K_{\text{exp}}/K_{\text{band}}$ should be robust.

Finally, we agree with the referee on the point that "the density-wave-like order may be driven by electronic correlations, and the high pressures may reduce the electronic correlations and suppress the density-wave-like order, allowing superconductivity to occur".

In the revised manuscript, the influence of DFT "flavors" has been discussed in Supplementary Note 2 and Supplementary Note 3. The discussion about the density-wave-like order and electronic correlations has been added to Page 14, Line 278-281.

References

[1] Ambrosch-Draxl and Sofo, Comput. Phys. Commun. **175**, 1 (2006)

[2] Gajdoš et al., Phys. Rev. B **73**, 045112 (2006)

2. If possible, can the authors place data points in Fig. 2d for other superconducting nickelates they mention in the introductory part of the manuscript?

This is an excellent suggestion. Unfortunately, the value of $K_{\text{exp}}/K_{\text{band}}$ or other experimental information on electronic correlations for other superconducting nickelates is currently

unavailable. This is because the superconducting nickelates we mentioned in the introduction part are thin films. Many spectroscopic studies such as FTIR and ARPES on these films are quite challenging, and have not been successfully carried out so far.

3. Temperature-dependent, optical spectral weight shifts are difficult to quantify accurately with reflectance data alone. In particular, spectral weight should be conserved in the CDW phase transition, and this is not obvious from the presented data. In future, temperature dependent ellipsometry measurements will be needed to accurately quantify spectral weight redistribution due to CDW.

This is a great point. We agree with the referee that "spectral weight should be conserved in the CDW phase transition". Actually, we did a careful spectral weight analysis (Page 9, section "Energy scale of the spectral weight redistribution") and found that the spectral weight is conserved at about 6500 cm^{-1}. On the other hand, we agree with the referee that "In future, temperature dependent ellipsometry measurements will be needed to more accurately quantify the spectral weight redistribution due to the density wave".

The above information has been mentioned in the revised manuscript [Page 10, Line 191–193].

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

I have reviewed the authors' responses to all criticisms raised by three reviewers and found that they adequately addressed all comments.

The novelty of this work lies in its status as the first experimental determination of the origin and nature of the electronic correlations in $La_3Ni_2O_7$, which constitutes a fundamental step toward understanding its superconductivity under pressure. The optical measurements and analysis made by the authors are precise and concrete.

Therefore, I recommend publishing the current work in Nature Communications.

Reviewer #2 (Remarks to the Author):

The authors revised their manuscript based on the comments from the three referees. While I agree with some of the changes made, to my opinion most of the main points remained ambiguous. Therefore, I cannot recommend this work for publication in Nature Communications. Here are my points below:

1. Authors returned back to the two-Drude-interpretation of their data, which could be reasonable. But the first thing one can notice is that their determined Drude parameters differ from the first version of this manuscript (below, I put them side by side in Fig1). This suggests that the decomposition of the optical data can be interpreted in whatever way. According to the authors' description, two different Drude possess different orbital information, in the first version, one type of d-orbitals is gapped due to CDW formation; whereas in the last version the other d-orbital (as different Drude contributions can be adjusted to be gapped).

2.“After considering the referee’s question, we realized that the distinguishing effect lies in the temperature dependence of the Drude weight. For a simple temperature-induced Fermi level shift, the Drude weight is expected to decrease continuously as the temperature is lowered. For example, as shown in Fig. R3b, the temperature-induced Fermi level shift leads to a continuous decrease of the Drude weight in TaAs [2]. However, in La3Ni2O7, the Drude weight does not show a continuous decrease upon cooling, but is abruptly suppressed at the density-wave-like transition, in conflict with the temperature-induced Fermi level shift.”

--> I think this argument is not very reasonable either. Once the band in question shifts completely above the Fermi energy, obviously one should lose the intraband contribution. I really cannot understand why the abrupt change of the Drude weight cannot be seen in this way, either. See the sketch in Fig2.

One minor comment on DFT “flavors”: The broadening factor in Wien2k is just an arbitrary parameter basically to plot the intraband contribution. What the code calculates is only the plasma frequency and based on the chosen broadening it just adds an arbitrary Drude contribution. So I think, we anyways don’t expect to see the plasma frequency changes with different broadening.

Response to the Report of Reviewer #1 – NCOMMS-23-57046A

I have reviewed the authors' responses to all criticisms raised by three reviewers and found that they adequately addressed all comments. The novelty of this work lies in its status as the first experimental determination of the origin and nature of the electronic correlations in La3Ni2O7, which constitutes a fundamental step toward understanding its superconductivity under pressure. The optical measurements and analysis made by the authors are precise and concrete. Therefore, I recommend publishing the current work in Nature Communications.

We thank the referee for reviewing our responses and revised manuscript. We also thank the referee for underlining the novelty and importance of our work, and recommending our work for publication in Nature Communications.

Response to the Report of Reviewer #2 – NCOMMS-23-57046A

The authors revised their manuscript based on the comments from the three referees. While I agree with some of the changes made, to my opinion most of the main points remained ambiguous. Therefore, I cannot recommend this work for publication in Nature Communications. Here are my points below:

1. Authors returned back to the two-Drude-interpretation of their data, which could be reasonable. But the first thing one can notice is that their determined Drude parameters differ from the first version of this manuscript (below, I put them side by side in Fig1). This suggests that the decomposition of the optical data can be interpreted in whatever way. According to the authors' description, two different Drude possess different orbital information, in the first version, one type of d-orbitals is gapped due to CDW formation; whereas in the last version the other d-orbital (as different Drude contributions can be adjusted to be gapped).

Thank Reviewer #2 for this comment. There is some misunderstanding about the two versions of Drude analysis. We are sorry for the misunderstanding. Below, let's clear it up.

First of all, the labels of the components are different for the two versions, so one can NOT compare the Drude parameters side by side. This is also the reason why the Drude parameters are different for the two versions. Figure R1 illustrates the relationship between the two versions. Here, we use $\mathrm{D1}^{old}$ and $\mathrm{D2}^{old}$ for the two Drude components in the first version, and $\mathrm{D1}^{new}$ and $\mathrm{D2}^{new}$ for the two Drude components in the last version.

(i) The calculated interband $\sigma_1(\omega)$ suggests that $\mathrm{D2}^{old}$ is actually an interband component (not d orbitals) as indicated by Arrow4, so $\mathrm{D2}^{old}$ is replaced by the interband L1 in the last version (Arrow3). Actually, taking interband transitions into account was suggested by Reviewer #2 (see the 1st round of response to comments of Reviewer #2 for details).

(ii) Following Reviewer #1's suggestion in the 2nd round of reply, $\mathrm{D1}^{old}$ is decomposed into $\mathrm{D1}^{new}$ and $\mathrm{D2}^{new}$ in the last version (Arrow1 and Arrow2), so $\mathrm{D2}^{new}$ is part of $\mathrm{D1}^{old}$.

Part of $\mathrm{D1}^{old}$ is gapped in the first version; $\mathrm{D2}^{new}$ is gapped in the last version. Note that $\mathrm{D2}^{new}$ is part of $\mathrm{D1}^{old}$, so in the two versions, the same Drude component is gapped. One can not adjust different contributions to be gapped.

Finally, we would like to underline that the first version of the two-Drude analysis was not

reasonable, because it didn't take the low-frequency interband transitions into account. In the last version, L1 describes the low-frequency interband transitions; $D1^{new}$ and $D2^{new}$ take account of the multiband nature of $La_3Ni_2O_7$. At each temperature, we run the least-square fit to make sure the Drude-Lorentz fit converges, so the decomposition of the optical data can not be interpreted in whatever way.

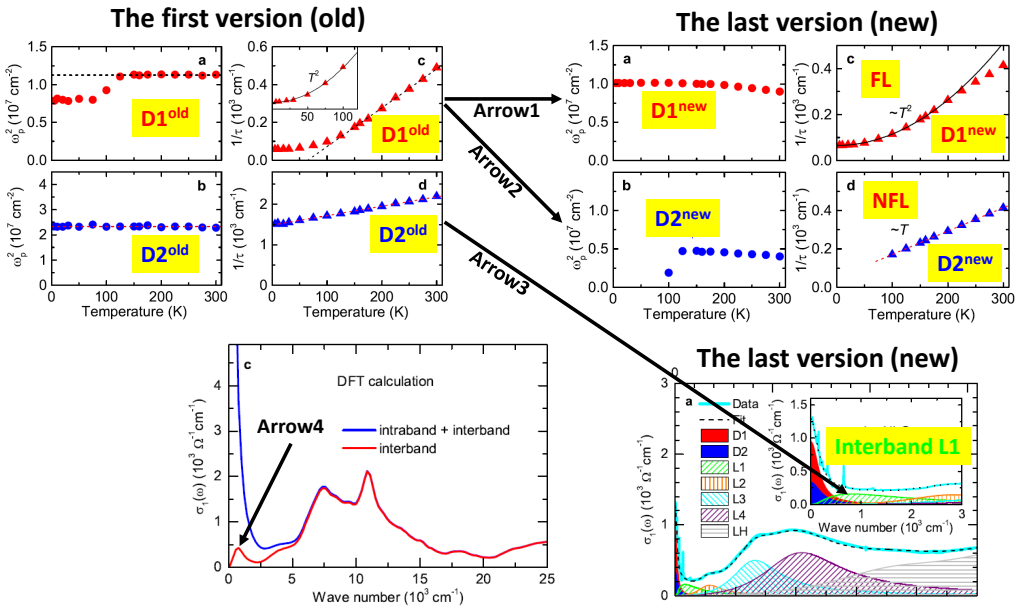

Figure R1. Comparison between the two versions of two-Drude analysis.

2. "After considering the referee's question, we realized that the distinguishing effect lies in the temperature dependence of the Drude weight. For a simple temperature-induced Fermi level shift, the Drude weight is expected to decrease continuously as the temperature is lowered. For example, as shown in Fig. R3b, the temperature-induced Fermi level shift leads to a continuous decrease of the Drude weight in TaAs [2]. However, in La3Ni2O7, the Drude weight does not show a continuous decrease upon cooling, but is abruptly suppressed at the density-wave-like transition, in conflict with the temperature-induced Fermi level shift."

I think this argument is not very reasonable either. Once the band in question shifts completely above the Fermi energy, obviously one should lose the intraband contribution. I really cannot understand why the abrupt change of the Drude weight cannot be seen in this way, either. See the sketch in Fig2.

We thank the referee for this question. In order to answer it clearly, we plot the referee's sketch together with the corresponding Drude weight in Fig. R2.

The Drude weight $\omega_{p,D}^2$ is proportional to the area of the Fermi surface. For a simple temperature-induced band shift, when the band shifts upwards (compare **c** and **d** in Fig. R2), the area of the Fermi surface shrinks, and $\omega_{p,D}^2$ decreases continuously upon cooling, as schematically shown by the red dashed line in Fig. R2. One loses the intraband contribution ($\omega_{p,D}^2$ reaches zero), when the band bottom is just at the Fermi energy (**b** in Fig. R2). When the band shifts completely above the Fermi energy (**a** in Fig. R2), $\omega_{p,D}^2$ remains zero.

Therefore, the temperature-induced band shift leads to a continuous decrease of $\omega_{p,D}^2$ upon

cooling (red dashed line in Fig. R2). In sharp contrast, $\omega^2_{p,D}$ for $La_3Ni_2O_7$ (blue solid circles) is almost temperature independent above T^*, and abruptly drops to zero below T^*, which does not agree with the behavior of the temperature-induced band shift.

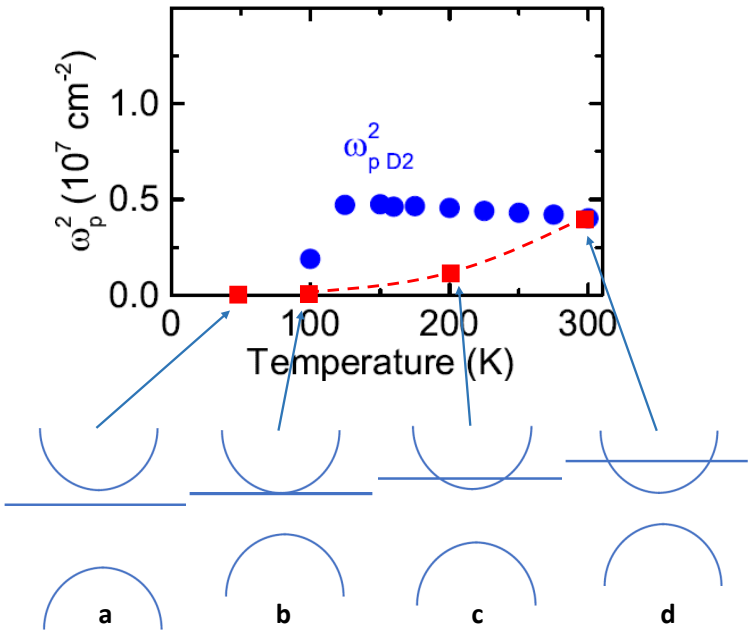

Figure R2. Temperature-induced band shift.

One minor comment on DFT "flavors": The broadening factor in Wien2k is just an arbitrary parameter basically to plot the intraband contribution. What the code calculates is only the plasma frequency and based on the chosen broadening it just adds an arbitrary Drude contribution. So I think, we anyways don't expect to see the plasma frequency changes with different broadening.

We agree with the referee on this point. Actually, this is exactly what we intended to say. The above sentences have been added to Supplementary Note 2, the end of the 1st paragraph.