nature portfolio

Peer Review File

Transport phase diagram and anomalous metallicity in superconducting infinite-layer nickelates

Corresponding Author: Dr Yu-Te Hsu

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 their manuscript, "Transport phase diagram and anomalous metallicity in superconducting infinite-layer nickelates" Y.-T. Hsu et al present a detailed study of the magnetotransport properties of a new generation of nickelate thin films. These measurements bring the overall shape the nickelate phase diagram into sharper focus, and as such I believe they will be of interest to the broad community interested in superconductivity and nickelates in particular. The patterns observed are significant and the methodology is clearly laid out and the analysis is thoroughly presented. However, there are few matters of interpretation that I believe should be considered before it is published.

Perhaps the central claim of the paper is the consistent T^1.5 power law thar appears in the resistivity of all the over-doped samples. The authors scrupulously report the variation in effectiveness of fit for several functional forms, and the consistency of the 1.5 exponent is striking. However, in Figure one they present this power law as being characteristic of the superconducting samples, with a \rho ~T^2 power law taking over at dopings higher than x = 0.3, where the superconducting dome ends. This would be a striking result, but I don't think it is warranted by the fits they present in the supplemental information. The temperature region over which a T^2 form fits the data is limited to the cusp of the downturn in d\rho/dT that appears to be related to the low-temperature upturn in \rho. One could similarly fit the data on the superconducting samples in this way. Do the authors believe there is something that clearly distinguishes the data on the superconducting samples from those on the non-superconducting samples? This is an important issue because affects how we should understand the relationship between the observed anomalous transport properties and the pairing interaction.

Relatedly, I think the authors should clarify what they think the T^1.5 law is telling us about these nickelate systems. In their discussion they seem to settle on a possible metallic spin glass as the best interpretation of the T^1.5 power law, but this would appear to undercut their claim that their measurements reveal an intrinsic anomalous metallicity that is important for understanding the superconductivity. If a spin glass explains the observed power law, is the conclusion of anomalous metallicity still valid?

Finally, the authors note that, although the power low of \rho(T) is consistent across the superconducting dome, the prefactor of the T-dependent part of the resistivity actually scales with \rho_0 more closely than it does with the doping level. It seems to me that this suggests a difficulty in determining the geometric factor for these samples, which might indicate the presence of residual extended defects. If the authors believe this is not the case, or that it is unlikely to be, it would be helpful to have clearly stated why. This is particularly important as the improved crystallinity of these samples is a major selling point for the paper.

In summary, Hsu et al have performed a very careful and comprehensive evaluation of the patterns in the transport properties of these new, improved Nickelate films. I agree with their estimation of these data as "laying important groundwork" for understanding anomalous metallicity in the nickelates, and their work certainly goes a long way towards establishing the existence of a distinctive metallic state in these materials. As such I think this paper is worth publishing in Nature Communications if the authors can address the interpretational issues I outlined above.

(Remarks to the Author)

The manuscript reports the results of the experimental studies of the infinite-layer-nickelate films Nd{1-x}Sr{x}NiO2 (NSNO). As I have inferred from reading the introductory section the manuscript, the authors motivate this study by trying to uncover differences and/or similarities between the transport properties of NSNO and 'high-Tc' cuprate superconductors. The special attention is given to the temperature dependence of resistivity and, consequently, magneto transport in the magnetic fields up to 54 Tesla (!) It seems the authors are basically following the avenues of the experiments in cuprates as well as more exotic URu2Si2 system where such high magnetic fields have been used to suppress the superconducting or hidden order phase correspondingly. Overall, I am quite impressed by the scope of this work. At the same time, the sheer scope of the work apparently renders some of the comments by the authors sound a bit superficial.

- (1) Indeed, let us look at Fig. 1, where the main findings of the manuscript are summarized. In Fig. 1 the authors present the system's phase diagram in the T-x plane based on the transport data. Looking at the overdoped region, the reader notices that superconductivity is suppressed by disorder at around x=0.30. In the region to the right the authors claim that resistivity shows T^2 temperature dependence. I actually find this statement extremely surprising for the following [fairly basic] reason: x=0.30 formally signifies the quantum critical (T=0) pairbreaking critical point. This means that there will be fluctuations associated with it which in two-dimensional systems give logarithmic temperature dependence to conductivity [see e.g. PRB 76, 094511 (2007) and references therein]. Thus, I would very much appreciate if the authors would comment on this apparent contradiction between their experimental results and well established theoretical predictions. The same argument can also be applied to transport in magnetic field both in-plane and out-of-plane.
- (2) Minor comment: in line 100 (second half of page 4) the authors write that "... suggesting that these upturns are caused by some form of competing electronic order." This statements is extremely vague. I suggest that it should be either removed or unpacked: there are not that many types of electronic order especially for disordered two-dimensional systems (i.e. films).

Overall, despite some of the shortcomings, I think this is an experimental work of high quality and is certainly worthy of publication in Nature Communications. However, before I can recommend this manuscript for publication, I would like the authors to address my critique above.

Reviewer #3

(Remarks to the Author)

The manuscript titled "Transport phase diagram and anomalous metallicity in superconducting infinite-layer nickelates" by Y. Hsu et al. presents a compelling investigation into the transport properties and anomalous metallicity observed in superconducting infinite-layer nickelates. Through systematic measurements of electric transport under magnetic fields up to 54 and over a wide temperature range down to 0.3 K on a series of NSNO thin films ranging from underdoped to highly overdoped, the authors propose the existence of non-Fermi-liquid behavior over an extended doping range within the superconducting dome. This topic is both timely and intriguing. Moreover, exploring the phase diagram and anomalous metallicity of infinite-layer nickelates holds significance for the condensed matter physics community. However, before publication, it is essential to address the following comments and revise the manuscript accordingly:

- 1. The infinite-layer nickelates show the resistive upturn for a wide range of doping in the low temperature region. According to the author's previous paper, this resistive upturn cannot be directly ascribed to disorder or localization effects (with or without interaction corrections), nor to Kondo physics. What is the mechanism for this resistive upturn? This is a serious issue that hinders author reveal the resistivity behavior inside the quantum critical region and comprise reliability of this research. To explore the quantum criticality and the anomalous/strange metallicity, it needs to go to very low temperature. For example, strange-metal behavior in a ferromagnetic Kondo lattice (Nature 579, 51–55 (2020)) or La2-xCexCuO4 (Nature 476, 73, 2011) appears from 5K to 40mK or 20K to 20mK for over two or three decades. The linear-T resistivity can extend to 40mK or 20 mK. By comparison, the linear-T resistivity quantum the quantum critical point near xopt in IFN only extends to around 40K. Electron-phonon interaction could also contribute to linear-T resistivity at that temperature. The manuscript should address how other trivial scattering channels at higher temperatures, such as electron-phonon interaction, are ruled out.
- 2. The resistance exhibits a plateau in the over-doped region at low temperatures. Could saturating phase coherence from short-lived superconducting puddles be responsible for this anomalous metallicity? Discussion of references related to anomalous metals (e.g., Rev. Mod. Phys. 91, 011002 (2019), Science 366, 1450 (2019)) would be valuable.
- 3. The manuscript should specify the lowest temperature at which $\Delta\rho \propto T1.45$ power is observed from x=0.175 to x=0.275. It appears that this behavior only extends to a few Kelvin, raising a similar concern despite the weaker insulating behavior beyond xopt Furthermore, the anomalous metallicity observed from x=0.175 to x=0.275 extends to 50 K, with the upper temperature being independent of doping, while T-linear resistivity near xopt extends to 300 K. This suggests that these two regions may not have same origin or can evolve gradually. The author claims that the non-FL form of $\Delta\rho(T)$ over an extended doping range beyond xopt is more reminiscent of the quantum critical phase observed in hole-doped cuprates. However, the behavior observed in the nickelates differs significantly from that in hole-doped cuprates, as evidenced by previous research (Science 323, 603-607(2009)). It can be clearly seen from figure 3 in this paper, the component of power evolves smoothly from 1 to 2 for a wide region doping and temperature in the whole over-doped phase diagram. Therefore, the anomalous metallicity observed from x=0.175 to x=0.275 may represent a distinct "phase" or phenomenon unrelated to quantum criticality at xopt.
- 4. The Magnetoresistance in ILNs near optimal doping show distinct behavior from cuprates. There are no H-linear MR and MR fails to follow the quadrature form or Kohler's scaling, which is intriguing. More discussion and analysis are suggested in comparison to cuprates (e.g., Science 361, 479–481 (2018), Nature 601, 205-210 (2022), Nature 595, 661–666 (2021)).

Version 1:		
Reviewer comments:		
Reviewer #1		

(Remarks to the Author)

In their revised manuscript, Y.-T. Hsu and coauthors have made several helpful revisions that I think make the overall message of the manuscript clearer and make the analysis that they used more accessible. I particularly appreciate their inclusion of the new section A.2 on the quantitative evaluation of the resistivity; this is a topic that frequently gets short shrift in papers on electrical transport. additionally, their explanation of the contrast is between superconducting and non-superconducting films is greatly aided by the addition of their data on non-superconducting samples to figure four as well as the new figure in the supplemental information. As such I think that the manuscript is now ready for publication in Nature Communications.

If I could be permitted one final recommendation for the authors it would be to expand somewhat their discussion of the significance of strong magnetic scattering as indicated by n~1.5 power law. The authors do a nice job of articulating the reasons for preferring the spin-glass interpretation of this power law, and I appreciate the added sentence stating that this conclusion indicates the presence of dominant magnetic scattering. However, I think that for the broad readership of nature communications it would be useful to unpack the significance of this observation a bit more, particularly with respect to the absence of the observed static magnetic order in the phase diagram, the comparison between cuprates and nickleates, and the nature of the pairing mechanism.

Reviewer #2

(Remarks to the Author)

I have read the author's responses to my comments and to the comments and critique expressed by other referees. The authors have addressed the concerns that I have mentioned in my report. I also find the revised version of the manuscript and the corresponding discussion much improved compared to the previous version. Overall, I think that this manuscript presents important and interesting transport results which will be of interest to broader condensed matter community. I support the publication of this manuscript in the present form.

Open Access This Peer Review File is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made.

In cases where reviewers are anonymous, credit should be given to 'Anonymous Referee' and the source.

The images or other third party material in this Peer Review File are included in the article's Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain

permission directly from the copyright holder.

To view a copy of this license, visit https://creativecommons.org/licenses/by/4.0/

Reviewer #1 (Remarks to the Author):

In their manuscript, "Transport phase diagram and anomalous metallicity in superconducting infinite-layer nickelates" Y.-T. Hsu et al present a detailed study of the magnetotransport properties of a new generation of nickelate thin films. These measurements bring the overall shape the nickelate phase diagram into sharper focus, and as such I believe they will be of interest to the broad community interested in superconductivity and nickelates in particular. The patterns observed are significant and the methodology is clearly laid out and the analysis is thoroughly presented. However, there are few matters of interpretation that I believe should be considered before it is published.

> We thank the Reviewer for the positive feedback on the significance and presentation of our work. We also thank the Reviewer for pointing out key interpretational issues that required further clarifications.

(#1.1) Perhaps the central claim of the paper is the consistent T^1.5 power law that appears in the resistivity of all the over-doped samples. The authors scrupulously report the variation in effectiveness of fit for several functional forms, and the consistency of the 1.5 exponent is striking. However, in Figure one they present this power law as being characteristic of the superconducting samples, with a \rho ~T^2 power law taking over at dopings higher than x = 0.3, where the superconducting dome ends. This would be a striking result, but I don't think it is warranted by the fits they present in the supplemental information. The temperature region over which a T^2 form fits the data is limited to the cusp of the downturn in $d \cdot d \cdot d$ that appears to be related to the low-temperature upturn in \rho. One could similarly fit the data on the superconducting samples in this way. Do the authors believe there is something that clearly distinguishes the data on the superconducting samples from those on the non-superconducting samples? This is an important issue because affects how we should understand the relationship between the observed anomalous transport properties and the pairing interaction.

> We thank the Reviewer for pointing out that, as shown in the original manuscript, the form of low-temperature (T) resistivity in the very overdoped, non-superconducting (non-SC) samples ($x \ge 0.30$) does not appear manifestly different than that found in the overdoped, superconducting (SC) samples ($0.175 \le x \le 0.275$). To better illustrate the key difference in the form of low-T resistivity across x = 0.275, we compare in the following figure (Fig. R1) the low-T resistivity and the corresponding $d\rho/dT$ measured at zero and large applied magnetic fields (>10 T). Samples of similarly low T_0 and ρ_0 are chosen for this illustration. As can be seen in Fig. R1b and R1f, $d\rho_{ab}/dT$ in the field-induced normal state of the x = 0.275 and 0.25 films show a downward deviation from the higher-T behaviour at $\approx 3T_0$ (grey shaded region), indicating that the apparent T^2 behaviour below $3T_0$ is most likely caused by the onset of the resistivity upturn. In contrast, for the x = 0.30 and 0.3125 films, $d\rho_{ab}/dT$ shows a kink at $T_2 \approx 20$ K (arrows in Fig. R1d and R1h), below which $d\rho/dT$ exhibits T-linear behaviour with a near zero intercept, consistent with $\rho(T) = \rho_0 + A_2T^2$. The fact that the T_2 values in $T_2 = 0.30$ and T_2

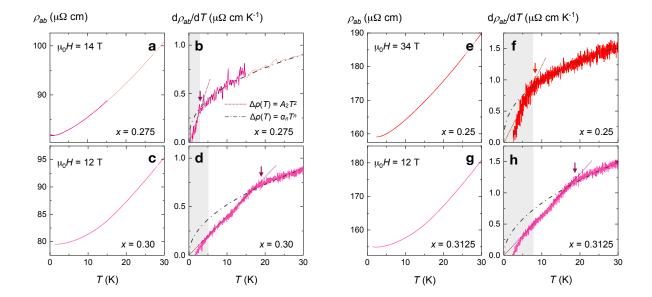


Figure R1: (a, c, e, g) Normal-state resistivity $\rho_{ab}(T)$ and (b, d, f, h) temperature-derivative $d\rho_{ab}/dT$ measured at large applied magnetic field for $0.25 \le x \le 0.3125$; for x = 0.275, data above 15 K are measured without applied field. Grey shadings mark the region below $3T_0$ (measured in high magnetic fields). Temperature at which a kink in $d\rho_{ab}/dT$ is found is marked by the arrows. For x = 0.30 and 0.3125, $d\rho_{ab}/dT$ shows a T-linear behaviour with zero intercept over a considerable temperature range ($2T_0 \le T \le 20$ K; d & h), indicating $\rho = \rho_0 + A_2T^2$ within this temperature range.

We find further evidence demonstrating the difference in the form of the low-T resistivity between the SC and non-SC films by tracking the extracted temperature scales. As shown in Fig. R2d, both the upper and lower temperature bounds of the T^2 resistivity for the SC samples closely track T_0 , indicating that the apparent T^2 resistivity at the lowest T is caused by the resistive upturn. By contrast, in the non-SC films, $T_{2, \text{ upper}}$ is much higher than T_0 and shows no correlation. This observation suggests that while the lower bound of the T^2 resistivity is limited by the magnitude of T_0 , the upper bound of T^2 resistivity in the non-SC samples is not influenced by the resistivity upturn.

Based on these distinctive features of the low-T resistivity in the non-SC films, including (i) a kink in $d\rho/dT$ at $T\gg T_0$, (ii) an extended temperature range in which T^2 resistivity is found, and (iii) the absence of a correlation between $T_{2, \text{ upper}}$ and T_0 , we conclude that the low-T resistivity of non-SC NSNO is best described by $\rho(T) = \rho_0 + A_2T^2$, contrasting with the $\rho(T) = \rho_0 + \alpha_n T^{1.45}$ form found in the SC films. We have now made the following revision in the existing figures to clarify this point:

- Fig. 4: added 12-T data on x = 0.30 and 0.3125 in inset of panel e
- Fig. S13: added 12-T data for $x \ge 0.30$
- Fig. S14: updated panel d for non-SC films
- Fig. R2: added to the Supplementary Information (SI) as new Fig. S15

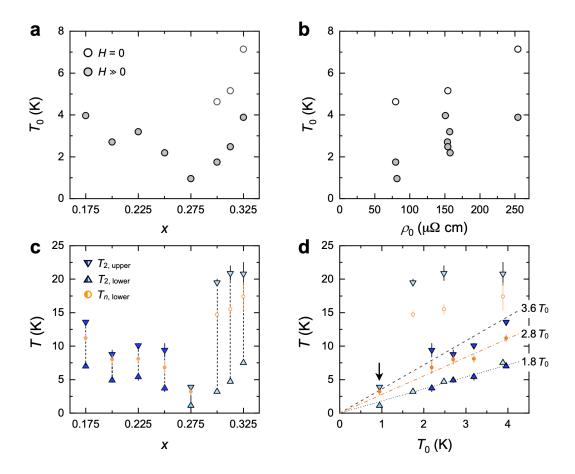


Figure R2: (a, b) T_0 of overdoped NSNO films as a function of (a) Sr doping x and (b) residual resistivity ρ_0 . (c, d) Upper and lower temperature bounds of T^2 resistivity as a function of (c) x and (d) T_0 , as well as the lower bound of $T^{1.45}$ resistivity ($T_{n, lower}$). Vertical arrow in (d) marks the corresponding temperature scales for x = 0.275, while dash, dotted-dash, and dotted line corresponds to $T = 3.6T_0$, $2.8T_0$, and $1.8T_0$, respectively.

(#1.2) Relatedly, I think the authors should clarify what they think the T^1.5 law is telling us about these nickelate systems. In their discussion they seem to settle on a possible metallic spin glass as the best interpretation of the T^1.5 power law, but this would appear to undercut their claim that their measurements reveal an intrinsic anomalous metallicity that is important for understanding the superconductivity. If a spin glass explains the observed power law, is the conclusion of anomalous metallicity still valid?

> The observation of a $T^{1.5}$ power law strongly points to a mechanism of magnetic scattering. The Reviewer correctly inferred that we regard a spin-glass ground state as the most compatible scenario with currently available data. While it is debatable whether a spin glass state represents an "intrinsic" property of the nickelates, as it is realised by the presence of strong disorder, we wish to point out that the superconducting nickelates are intrinsically disordered as a doped Mott-like system. We note that evidence for a spin-glass state has also been found in La_{2-x}Sr_xCuO₄ (LSCO) below p^* [Nat. Phys. 16, 1064; ref. 56 in manuscript], hinting at the magnetic nature of the cuprate pseudogap. Therefore, our results serve as indirect evidence for an intrinsic magnetism associated with the NiO₂ lattice (as $T^{\approx 1.5}$ resistivity

is also found in $La_{1-x}Sr_xNiO_2$ and $Pr_{1-x}Sr_xNiO_2$) and reaffirm recent findings from μSR [Nat. Phys. 18, 1043; ref. 15 in manuscript].

To emphasize the significance of the $T^{1.5}$ resistivity observed in NSNO, we have now added the following sentence in the main text [lines 250-253]:

"Importantly, the manifestation of T^{1.5} resistivity power law provides evidence for a dominant magnetic scattering in the normal ground state, and supports the existence of a fluctuating magnetic order intrinsic to the Ni-O lattice under hole doping."

(#1.3) Finally, the authors note that, although the power low of \rho(T) is consistent across the superconducting dome, the pre-factor of the T-dependent part of the resistivity actually scales with \rho 0 more closely than it does with the doping level. It seems to me that this suggests a difficulty in determining the geometric factor for these samples, which might indicate the presence of residual extended defects. If the authors believe this is not the case, or that it is unlikely to be, it would be helpful to have clearly stated why. This is particularly important as the improved crystallinity of these samples is a major selling point for the paper. > In our previous work conducted on the same set of samples [Nature 619, 290], we demonstrated that $d\rho_{ab}/dT$ above 100 K in NSNO and LSCO near respective optimal doping have nearly identical values ($\approx 1.1 \ \mu\Omega$ cm K⁻¹). The striking similarity in $d\rho_{ab}/dT$ between these two systems indicates that the level of residual disorder does not play a dominant role in determining the prefactor of the T-dependent resistivity. We also find that $d\rho_{ab}/dT$ at high T for films with $0.15 \le x \le 0.30$ increases as x increases (Fig. S2c), while ρ_0 decreases slightly as x increases in this doping range. This observation demonstrates that the magnitude of the Tdependent resistivity is not dictated by the residual resistivity of our films. Instead, we believe that the correlation between ρ_0 and a_n most likely implies that the physical process leading to the $T^{1.5}$ resistivity is enhanced by the presence of residual disorder, consistent with magnetic scattering in a spin glass.

To clarify that the level of residual order does not determine the prefactor of the *T*-dependent resistivity, we have expanded the SI with an additional section **[A.2 Quantitative evaluation of in-plane resistivity]** and added the following sentence to the main text (lines 258-261):

"The fact that the present NSNO thin films exhibit nearly identical dpab/dT values as found in high-quality LSCO single crystals strongly suggests that neither extended defects – known to affect the transport properties of earlier NSNO films – and/or dominant electron-phonon scattering are playing a major role in quantifying the in-plane resistivity values of these next-generation films; see SI Sec. A.2 and F for further discussions."

Note that this added sentence simultaneously addresses comment #3.1 from Reviewer #3.

In summary, Hsu et al have performed a very careful and comprehensive evaluation of the patterns in the transport properties of these new, improved Nickelate films. I agree with their estimation of these data as "laying important groundwork" for understanding anomalous

metallicity in the nickelates, and their work certainly goes a long way towards establishing the existence of a distinctive metallic state in these materials. As such I think this paper is worth publishing in Nature Communications if the authors can address the interpretational issues I outlined above.

> We thank the Reviewer for their positive evaluation of our work and for recommending its publication in Nature Communications, following our consideration of these interpretational issues. We hope the Reviewer will find our response to the raised points satisfactory.

Reviewer #2 (Remarks to the Author):

The manuscript reports the results of the experimental studies of the infinite-layer-nickelate films Nd{1-x}Sr{x}NiO2 (NSNO). As I have inferred from reading the introductory section the manuscript, the authors motivate this study by trying to uncover differences and/or similarities between the transport properties of NSNO and 'high-Tc' cuprate superconductors. The special attention is given to the temperature dependence of resistivity and, consequently, magneto transport in the magnetic fields up to 54 Tesla (!) It seems the authors are basically following the avenues of the experiments in cuprates as well as more exotic URu2Si2 system where such high magnetic fields have been used to suppress the superconducting or hidden order phase correspondingly. Overall, I am quite impressed by the scope of this work. At the same time, the sheer scope of the work apparently renders some of the comments by the authors sound a bit superficial.

> We thank the Reviewer for their positive appraisal of the scope of our work. Below we address the Reviewer's specific comments in the order in which they appeared.

(#2.1) Indeed, let us look at Fig. 1, where the main findings of the manuscript are summarized. In Fig. 1 the authors present the system's phase diagram in the T-x plane based on the transport data. Looking at the overdoped region, the reader notices that superconductivity is suppressed by disorder at around x=0.30. In the region to the right the authors claim that resistivity shows T^2 temperature dependence. I actually find this statement extremely surprising for the following [fairly basic] reason: x=0.30 formally signifies the quantum critical (T=0) pairbreaking critical point. This means that there will be fluctuations associated with it which in two-dimensional systems give logarithmic temperature dependence to conductivity [see e.g. PRB 76, 094511 (2007) and references therein]. Thus, I would very much appreciate if the authors would comment on this apparent contradiction between their experimental results and well established theoretical predictions. The same argument can also be applied to transport in magnetic field both in-plane and out-of-plane.

> We thank the Reviewer for bringing the theoretical paper on the conductivity correction upon approaching a pair-breaking quantum critical point (QCP) to our attention. However, we do not share the view that the absence of a logarithmically divergent conductivity in the x=0.30 film to be fundamentally surprising. Firstly, we do not think that x=0.30 presents a disorder-induced pair-breaking QCP in the present case of NSNO. In fact, ρ_0 of x=0.275 and 0.30 films are very similar ($\approx 80~\mu\Omega$ cm) in magnitude and substantially lower than that of the optimally-doped films ($\rho_0=120-150~\mu\Omega$ cm). This is not consistent with the hypothesis that x=0.30 is a pair-breaking QCP, for which T_c should be monotonically suppressed with increasing disorder

(and thus higher ρ_0). Secondly, while the logarithmic conductivity corrections have been demonstrated in various superconducting nanowires (i.e. quasi-1D systems), we are unaware of existing experimental reports on logarithmic conductivity correction upon approaching a pair-breaking QCP in thin films (n = 2). Thirdly, a recent theoretical study has shown that the conductivity corrections upon approaching a pair-breaking QCP in a Q2D multiband superconductivity can be of either sign [PRB 108, 184513]. Finally, we note that in the LSCO cuprate family, no logarithmic correction to the conductivity has been reported at the edge of the SC dome near x = 0.27. It is thus likely that the model referred to by the Reviewer is not directly applicable to NSNO, and the lack of a logarithmic conductivity correction in x = 0.30 film does not lead to a clear contradiction of theoretical expectations.

No further changes are made in the main text in response to this point.

(#2.2) Minor comment: in line 100 (second half of page 4) the authors write that "... suggesting that these upturns are caused by some form of competing electronic order." This statement is extremely vague. I suggest that it should be either removed or unpacked: there are not that many types of electronic order especially for disordered two-dimensional systems (i.e. films). > We agree with the Reviewer that the wording here is vague. As mentioned in the Introduction, the ILNs are known to host (short-range) charge and spin order as well as predicted to host stripe order. Given that charge order has so far only been observed in the underdoped regime in NSNO, we think it is a likely candidate for such a state-removing electronic order as discussed here.

For clarification, we have made the following revision (lines 100-101):

"...suggesting that these upturns are caused by a competing electronic order, such as the charge order recently found in underdoped ILNs [16–18]."

Overall, despite some of the shortcomings, I think this is an experimental work of high quality and is certainly worthy of publication in Nature Communications. However, before I can recommend this manuscript for publication, I would like the authors to address my critique above.

> We thank the Reviewer for the positive comments on the quality of our work. We hope the Reviewer considers that our responses have clarified the issues that were raised.

Reviewer #3 (Remarks to the Author):

The manuscript titled "Transport phase diagram and anomalous metallicity in superconducting infinite-layer nickelates" by Y. Hsu et al. presents a compelling investigation into the transport properties and anomalous metallicity observed in superconducting infinite-layer nickelates. Through systematic measurements of electric transport under magnetic fields up to 54 and over a wide temperature range down to 0.3 K on a series of NSNO thin films ranging from underdoped to highly over-doped, the authors propose the existence of non-Fermi-liquid behavior over an extended doping range within the superconducting dome. This topic is both

timely and intriguing. Moreover, exploring the phase diagram and anomalous metallicity of infinite-layer nickelates holds significance for the condensed matter physics community. However, before publication, it is essential to address the following comments and revise the manuscript accordingly:

> We thank the Reviewer for the positive comments on the methodology and significance of our work. We also thank the Reviewer for identifying the outstanding issues that require further clarifications.

(#3.1a) The infinite-layer nickelates show the resistive upturn for a wide range of doping in the low temperature region. According to the author's previous paper, this resistive upturn cannot be directly ascribed to disorder or localization effects (with or without interaction corrections), nor to Kondo physics. What is the mechanism for this resistive upturn? This is a serious issue that hinders author reveal the resistivity behavior inside the quantum critical region and comprise reliability of this research.

> We acknowledge that the persistence of the resistive upturn in NSNO complicates the interpretation of the low-T resistivity. For the reasons discussed in the main text [lines 98-113], we believe that the resistive upturns in the underdoped and overdoped regimes have different origins. In the underdoped side, the resistive upturn is likely caused by a competing electronic order as inferred from our previous works [Phys. Rev. Research 3, L042015; Front. Phys. 10, 846639; Nature 619, 290]; in the overdoped side, the resistive upturn is caused, at least in large part, by the presence of disorder as evidenced by its effective suppression under moderate magnetic fields. This can also be seen in Fig. R2, which shows T_0 is substantially suppressed by applied magnetic field (Fig. R2a) and T_0 correlates more strongly with ρ_0 than x (Fig. R2b). While it is possible that part of the resistive upturn beyond $x_{\rm opt}$ is unrelated to disorder, we believe the impact of the remaining resistivity upturn should not persist to $T >> 3T_0$, which is below 10 K for all the overdoped films (except x = 0.325). Our main conclusions, which are largely based on the form of normal-state resistivity between 10 and 300 K, are thus robust against the any potential impact of very low-T resistive upturn.

To clarify that our main conclusions on the normal-state resistivity are robust in the presence of resistive upturns, we have made the following revision (lines 174-176):

"We note that varying the lower T-limit for the power-law fitting between $2T_0$ and $5T_0$ does not affect the extracted n, as $n\alpha_n T^{n-1}$ is constrained to go through the origin, **demonstrating that** the extraction of the resistivity power-law parameters is robust in the presence of the small resistive upturns in OD-NSNO."

(#3.1b) To explore the quantum criticality and the anomalous/strange metallicity, it needs to go to very low temperature. For example, strange-metal behavior in a ferromagnetic Kondo lattice (Nature 579, 51–55 (2020)) or La2-xCexCuO4 (Nature 476, 73, 2011) appears from 5K to 40mK or 20K to 20mK for over two or three decades. The linear-T resistivity can extend to 40mK or 20 mK. By comparison, the linear-T resistivity quantum the quantum critical point near xopt in IFN only extends to around 40K. Electron-phonon interaction could also contribute to linear-T resistivity at that temperature. The manuscript should address how other trivial scattering channels at higher temperatures, such as electron-phonon interaction, are ruled out.

> We agree with the Reviewer that, in order to establish the existence of a quantum critical

point (QCP), one needs to study the normal-state behaviour down to very low temperatures. (This condition is currently precluded by the presence of the anomalous upturns in the present series of NSNO, despite a substantial improvement in the film crystallinity compared to the previous generation of samples.) Nonetheless, we note that the impact of quantum criticality is in fact more prominent at intermediate temperatures in which both thermal and quantum fluctuations are significant. The confluence of thermal and quantum fluctuations would lead to a fan-shaped region of T-linear resistivity on its temperature-tuning parameter (in our case the Sr doping x) phase diagram. While at present we cannot conclusively demonstrate that x_{opt} represents a QCP (nor do we claim that is the case), we note that the asymmetrical region of Tlinear resistivity on its T-x phase diagram strongly resembles that found in other quantum critical metals. Furthermore, we believe it is highly unlikely that a dominant electron-phonon scattering would be responsible for the high-T T-linear resistivity due to the doping evolution of T_1 . As mentioned in SI Sec. F, the Debye temperature Θ_D of LaNiO₂ single crystal is ≈ 340 K [Phys. Rev. Research 4, 023093; ref. 64 in manuscript], meaning that a T-linear resistivity due to electron-phonon scattering typically sets in around 100 K (i.e. $1/3 \Theta_D$; see e.g. Rev. Mod. Phys. 94, 041002]. As Θ_D is not expected to vary strongly with x, one expects T_1 to be largely doping independent in such a scenario. The discrepancy between T_1 (≈ 50 K near x_{opt}) and $\Theta_D/3$ and the strong x-dependence of T_1 (varying between 50 K and 200 K for $0.05 \le x \le 0.16$) thus do not support a dominant electron-phonon scattering behind the T-linear resistivity found in NSNO.

We have now added a sentence in the main text (lines 250-253) to clarify this point as well as a related point #1.3 from Reviewer #1 and refer the readers to the relevant SI section:

"The fact that the present NSNO thin films exhibit nearly identical dpab/dT values as found in high-quality LSCO single crystals strongly suggests that neither extended defects – known to affect the transport properties of earlier NSNO films – and/or dominant electron-phonon scattering are playing a major role in quantifying the in-plane resistivity values of these next-generation films; see SI Sec. A.2 and F for further discussions."

(#3.2) The resistance exhibits a plateau in the over-doped region at low temperatures. Could saturating phase coherence from short-lived superconducting puddles be responsible for this anomalous metallicity? Discussion of references related to anomalous metals (e.g., Rev. Mod. Phys. 91, 011002 (2019), Science 366, 1450 (2019)) would be valuable.

> We thank the Reviewer for bringing these literature on anomalous metallicity in 2D metals to our attention. According to one of the referred articles [Rev. Mod. Phys. 91, 011002], the defining signatures of the anomalous metallicity in a phase-incoherent superconductor is an anomalous low resistivity (far below the normal-state value) coupled with a large positive magnetoresistance (MR). In non-SC NSNO films ($x \ge 0.30$), the resistivity in the 'plateau region' (as described by the Reviewer) is in fact comparable or higher than the normal-state resistivity at 20 K (above T_{c0}). Moreover, the MR in these non-SC films is negative (Fig. R1), inconsistent with the expectations of short-lived superconducting puddles. Therefore, we do not regard the type of anomalous metallicity as discussed in the referred articles to be relevant for the low-T

resistivity behaviour in non-SC films. Nonetheless, we agree with the Reviewer that a reference to the anomalous metallicity in 2D superconductors would be valuable here.

We have now added a sentence and two new references to the main text (lines 224–227):

"We note here that the low-T resistivity plateau found in the T^2 resistivity regime in $x \ge 0.30$, with a small negative MR, is unlikely to be associated with short-lived superconducting puddles as found in other superconductors in the 2D limit [Rev. Mod. Phys. 91, 011002; Science 336, 1450], for which a large positive MR is expected."

(#3.3) The manuscript should specify the lowest temperature at which $\Delta \rho \propto T1.45$ power is observed from x=0.175 to x=0.275. It appears that this behavior only extends to a few Kelvin, raising a similar concern despite the weaker insulating behavior beyond xopt. Furthermore, the anomalous metallicity observed from x=0.175 to x=0.275 extends to 50 K, with the upper temperature being independent of doping, while T-linear resistivity near xopt extends to 300 K. This suggests that these two regions may not have same origin or can evolve gradually. The author claims that the non-FL form of $\Delta \rho(T)$ over an extended doping range beyond xopt is more reminiscent of the quantum critical phase observed in hole-doped cuprates. However, the behavior observed in the nickelates differs significantly from that in hole-doped cuprates, as evidenced by previous research (Science 323, 603-607(2009)). It can be clearly seen from figure 3 in this paper, the component of power evolves smoothly from 1 to 2 for a wide region doping and temperature in the whole over-doped phase diagram. Therefore, the anomalous metallicity observed from x=0.175 to x=0.275 may represent a distinct "phase" or phenomenon unrelated to quantum criticality at xopt.

> We thank the Reviewer for pointing out that the lower bound for the $T^{1.45}$ resistivity behaviour should be specified. Previously this temperature scale was shown implicitly in Fig. S14. **We have now added an additional Figure to the SI (new Fig. S15) to show the lower bound of** $T^{\approx 1.5}$ resistivity and added a line to the caption of Fig. 1 to refer to Fig. S15. We wish to mention that, the lower bound of the $T^{1.45}$ resistivity region (i.e. $T_{n, lower}$) is likely affected by the presence of resistive upturn, as hinted by the close correspondence between $T_{n, lower}$ and T_0 (Fig. S15d). As such, we chose not to include $T_{n, lower}$ in Fig. 1, which shows the characteristic temperature scales that are robust against disorder/defects.

With regards to the non-Fermi liquid transport found in the nickelates and cuprates, we in fact share the Reviewer's view that the manifestation of the non-FL behaviour are distinct in these two systems. We also interpreted the non-FL resistivity in overdoped NSNO between ≈ 5 and 50 K to be caused by an interaction that is distinct from that giving rise to the T-linear resistivity at high temperatures (lines 78-81, 264-277, and SI Sec. F). No further changes are made here.

(#3.4) The Magnetoresistance in ILNs near optimal doping show distinct behavior from cuprates. There are no *H*-linear MR and MR fails to follow the quadrature form or Kohler's scaling, which is intriguing. More discussion and analysis are suggested in comparison to cuprates (e.g., Science 361, 479–481 (2018), Nature 601, 205-210 (2022), Nature 595, 661–666 (2021)).

> We share the Reviewer's view that the MR behaviour in ILNs near x_{opt} is intriguing and

distinct from the cuprates. Given the absence of a H-linear MR in the high-field limit, most of the analyses described in the literature as mentioned by the Reviewer is not immediately applicable. Instead, we have performed a series of analyses to (1) identify the appropriate functional form to describe normal-state MR (Fig. S5), (2) study the temperature evolution of the MR magnitude in the high-H limit (Figs. S9, S10), and (3) investigate whether a scenario of anisotropic scattering can lead to such an unconventional MR (Fig. S11). These analyses are complemented by accompanying discussions on the distinct MR behaviour and inference for a strongly T-dependent mean-free-path anisotropy near $x_{\rm opt}$ in NSNO (SI Sec. B & C, Table S1). In our view the current extent of analysis and discussion on the nickelate MR is balanced in terms of experimental data, numerical simulations, and interpretational extrapolations. Therefore, we do not think it would be beneficial to further expand the MR analyses/discussion given the existing amount of MR-related materials in the current manuscript (including the SI).

No further changes are made to address this point.

Reviewer #1:

Remarks to the Author:

In their revised manuscript, Y.-T. Hsu and coauthors have made several helpful revisions that I think make the overall message of the manuscript clearer and make the analysis that they used more accessible. I particularly appreciate their inclusion of the new section A.2 on the quantitative evaluation of the resistivity; this is a topic that frequently gets short shrift in papers on electrical transport. additionally, their explanation of the contrast is between superconducting and non-superconducting films is greatly aided by the addition of their data on non-superconducting samples to figure four as well as the new figure in the supplemental information. As such I think that the manuscript is now ready for publication in Nature Communications.

If I could be permitted one final recommendation for the authors it would be to expand somewhat their discussion of the significance of strong magnetic scattering as indicated by n~1.5 power law. The authors do a nice job of articulating the reasons for preferring the spin-glass interpretation of this power law, and I appreciate the added sentence stating that this conclusion indicates the presence of dominant magnetic scattering. However, I think that for the broad readership of nature communications it would be useful to unpack the significance of this observation a bit more, particularly with respect to the absence of the observed static magnetic order in the phase diagram, the comparison between cuprates and nickleates, and the nature of the pairing mechanism.

> We thank the Reviewer for encouraging us to stress the significance of magnetic scattering interpretation associated with the n~1.5 power law. We would like to mention that, with regards to the absence of static magnetic order in the nickelates and the comparison with cuprates, we have quite extensively discussed these issues in the Discussion (page 13) and Supplementary Note F. Therefore, we do not see a strong need to expand the discussion on these points further. With regards to the nature of pairing mechanism, as it was only briefly mentioned in the Introduction (last paragraph), we have now added a sentence to the concluding paragraph in the main text to highlight the implication of our findings on understanding the pairing mechanism in the nickelates.