nature portfolio

Peer Review File

Enhanced superconductivity in the compressively strained bilayer nickelate thin films by pressure

Corresponding Author: Professor Hai-Hu Wen

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)
Dear Authors,
In this manuscript, the authors report on the effect of pressure on the superconductivity in bilayer nickelate thin films. This study is an important and timely exploration of the connection between the differences between superconductivity in bulk materials under pressure and it in compressively-strained thin films at ambient pressure. I agree that the sentence in discussion page 17 line 356 [The combination of strain engineering and high pressure is an essential tool in the research of nickelate superconducting thin films.] While the authors measured four samples under applied pressure, the results are quite complicated. The authors could improve the presentation of the data to make it easier to understand. Theoretical calculations provide some insight, but the discussion in this manuscript seems not sufficient to explain the complicated results.
To convince the general reader of the claims in this manuscript, it would be better to make major revisions to improve the paper, such as the addition of figures.

1, The definition of the onset superconducting transition temperature should be reconsidered. The definition applied in Fig. 1a in ref. 40, Y. Liu et al., Nature Materials 24, 1221 (2025). is most reliable and common method. Thie method should be applied to estimate the onset transition temperature. This method is also applied in Fig. 1 in ref. 13. To resolve the confusion regarding the transition temperature of bilayer nickelate thin films, it is appropriate to apply a reliable definition. In particular, the temperature values of broad transitions for Fig. 2b, Fig. 3b, suppl. Fig. S1, and Fig. S7b in this manuscript are strongly affected by how to be defined. For example, linear approximation in the transition region can be defined as the range between 80% and 90% of normal resistance values. By adopting this method, it is possible to compare all data through fair analysis. The experimental observation of the enhancement under applied pressure itself is a main claim of this manuscript. The temperature values should be compared by fair definition.
To remove the confusion by the different definition between this manuscript and previous studies ref. 40 and ref. 13, the authors should revise the definition with applying identical method.

2, There is no consistent explanation for the strange behaviour that the transition temperature increases with increasing the resistivity. This observation may come from the artifact by applying inadequate definition method discussed in comment 1. While the increase of the transition temperature may be induced by increase of Fermi surface or other enhancement of interaction, the increase of the resistance likely comes from the reduction of charge carrier density and/or increase of scattering. If these phenomena occurs simultaneously in the sample, how did the authors explain consistently for both the increase of transition temperature and resistance? These phenomena are independently discussed in the present manuscript, but the consistent explanation is necessary to convince the claim to readers.

3, The resistance values at 300 K for four samples in this study should be compared by plotting as a function of pressure to make clear the reproducibility of the pressure effect. Whether superconductivity occurs or not is an important point, but it is also important to compare the reproducibility in the resistivity of the normal state before that occurs. It is expected that the resistance variation by applying pressure should systematically occur via shorten lattice and increase of Fermi surface. If the authors claim the reproducibility, this point should be clearly explained with additional data plots.

4, In addition to the comment 2, can the simultaneous occurrence of increase of resistance and transition to the insulator be interpreted by theoretical band calculation ? These behaviours were not observed in bulk studies. This point may not be

explained in the present manuscript. While four samples in this study show similar behaviour, the difference between bulk results under pressure and that in this study should be discussed.
Intuitively, the applying pressure shrinks the lattice, corresponding to the increase of Fermi surface and in-plane hopping. It is likely that the resistance value decreases with increasing pressure up to a certain critical pressure condition, for example occurrence of structural phase transition. If such transition occurs in bilayer nickelate film, which will be also observed in bulk.

5, In this study, four samples were measured and discussed under pressure. In Fig. 1d, the resistivity values of six films are plotted. But in other figures under pressure, the resistance values are plotted. Were the resistance results, resistivity and resistance, consistent and reproducible? Judging from the high values of resistivities, the highest S6, S4, S2+35 days, S2, S5, S3, S1+35 days, and the lowest S1are plotted. S6 and S1 is the highest and lowest, respectively. However, the values for 600 Ohm of S3 in Fig. 3, 300 Ohm of S4 in Fig. S7, 200 Ohm of S1 in Fig. 2, and 150 Ohm of S2 in Fig. S3 are not consistent to the resistivities shown in Fig. 1. If this difference comes from the sample geometry, the authors should present the photo of all samples or its summary in tables to explain the geometrical difference. If the sample size and geometry are comparable, the authors have to explain the origin for this inconsistent resistivity / resistance shown in the Figures.

6, Is the comparison with infinite layer nickelate meaningful? In page 3 line 49, the authors describe that [This is strongly contrasted to the 3d^9 configuration in cuprates and infinite-layer nickelates.]. Why do the authors compare their results under pressure with infinite layers when they believe that the electronic structure and the origin of superconductivity are clearly different?

7, in page7, line 150, what does the reference 1 and 14 mean? Refs 1 and 14 correspond to bulk studies. In addition, in page 6 line 116-117, 20.75 A is elongated. The authors should describe the bulk value of c-axis length at ambient pressure. What does [abnormal elongation] mean? The comparison with the c-axis in previous film studies may be meaningful.

8, in page 9 line 193, [The slight increase of RH above 2.2 GPa may be induced by the solidification of PTM. Such behavior indicates that the carrier concentration increases upon applying a pressure.] These explanations seem too simplified. How does the solidification of PTM affect the RH of the film? Is this behavior observed in other materials by applying identical PTM? Moreover, as described in page 6 line 135, the RH of the bilayer nickelates is more complicated due to the multiband nature of the electronic structure of bilayer nickelates. The increase of RH does not simply indicate the increase of carrier concentration. The authors should provide a unified explanation throughout the text.

9, in page 10 line 208, honestly speaking, it is rather difficult to agree that the description [Although the temperature dependence of normal-state resistance exhibits a drastic evolution, the superconducting onset transition temperature dependence shows a clear enhancement at relative low pressures.] because the shift of transition is small in Fig. 3a. Moreover, the description in line 216-217 [the R(T) curves display nearly similar evolutions as sample S3 under high pressures, which first shows a decrease from 2 to 4 GPa and then increases gradually with further increasing pressure.] is not fair. The authors have to be fair. For sample S3, the resistance firstly increases from 0 GPa to 2 GPa. Why did the authors only explain the behavior from 2 to 4 GPa not from 0 to 2 GPa?

10, What does this description [As is known, the delicate nickelate thin films can be easily and partially deteriorated at high pressures when the liquid PTM solidifies, which would produce disorders and defects in the thin film that will enhance the carrier scattering.] mean? Probably, no one knows that the bilayer nickelate film is easily deteriorated by applying pressure because there are less studies. Many bulk crystals show reproducible results under pressure. In addition, in ref. 54, there is no similar deterioration. If the authors would like to conclude that the films in this study are easily deteriorated, this would suggest that they were inferior specimens with insufficient crystal quality to withstand the pressure application experiments. The authors may need to reconsider the pressure effect once the quality of thin film samples reaches a level comparable to that of bulk crystals.
Alternatively, the authors should clearly describe the upper limit of pressure at which the intrinsic property of the nickelate films can be discussed in this study. To remove the confusion to general readers, it would be better that the authors describe the observed results above the upper limit come from the extrinsic effect or PTM effect.

11, in page 13 line 277, how does the solidification affect the electronic structure of the film?

12, in page 13 line 288, only c-axis length is defined. How is the a-axis length defined in the calculation model? If the authors discuss the pressure effect, the a-axis should be also compressed as increasing pressure.

13, in page 14 line 303, Fig. 5d is firstly explained in main text. To improve the readability, it would be better that the authors either change the order of the figures or change the order of the explanations in the text.

14, in page 17 line 360-371, it is difficult to agree that the discussion in this study. Are the claims in this discussion firstly found in this manuscript? Because no reference is added, it would be better to check this point again. Moreover, it would be better to discuss here the consistency between film and bulk. If the interplay between interlayer and intralayer electronic correlation induces the reduction of the transition temperature, why this reduction has not been observed in bulk under pressure?

15, in page 18, line 389, although the transition temperature in ref. 54 seems similar, the overall behaviour under pressure seems completely different, no insulating behaviour appear in ref. 54. What is the origin of this difference?

16, in page 20 line 424, a five-probe method is described. The sample photo should be provided to explain the measurement configuration. Was the current uniformly applied to the small (0.6 mm x 0.3 mm) sample with five electrodes ?

17, It would be better to reconsider the offset for the resistance data shown in Fig. 3b and Fig. S7, because the minus value of resistance seems strange. Moreover, the offset value should be explicitly explained if it is added to the data. In general, the equal offset is added to the data.

18, It would be better to reconsider the color of the data shown in Fig. 1d, Fig. 3, and Fig. S7. It is difficult to recognize the correspondence of data.

19, in page 11 line 244, it is difficult to agree with [relatively sharp] for the data of 13 GPa. If the authors quantitatively discuss the sharpness of transition, it would be helpful to understand the evaluation standard.

20, the bilayer heterostructure La2.5Pr0.5Ni2O7 / La2.85Sr0.15Ni2O7 is applied to this pressure study. The basic resistance results of single films of 3 unit-cell La2.5Pr0.5Ni2O7 and 3unit-cell La2.85Sr0.15Ni2O7 are quite helpful to understand the effectiveness of the bilayer heterostructure. It would be better to present those results in supplement.

Reviewer #2

(Remarks to the Author)
This is an excellent manuscript with a significant contribution to the field of high-temperature superconductivity in nickelates La3Ni2O7. The authors successfully demonstrate a pressure-induced enhancement of superconductivity in compressively strained thin film samples, revealing a dome-shaped T-P phase diagram. The combination of high-quality experimental data and insightful theoretical analysis provides a compelling case for the proposed mechanism. I believe this work is highly suitable for publication and recommend its acceptance following a thorough and careful revision based on the following comments.

(1). The observation of a transition from metallic to weak insulating behavior at pressures above the optimal 9 GPa is a critical and intriguing finding. To fully substantiate this phenomenon and provide deeper insight, a more detailed analysis is required. It would be invaluable to see the full resistance-temperature curves at pressures both below and above this transition point. A discussion on whether this behavior is linked to an intrinsic structural or electronic phase transition, such as a charge density wave or Fermi surface reconstruction, would be highly valuable. Are there any other signatures or characteristics of this weak insulating behavior (e.g., a change in the resistance slope, hysteretic behavior, or magnetoresistance behavior) that could provide additional insights? Providing a more detailed picture of the normal state properties in this high-pressure regime will greatly strengthen the manuscript's conclusions.

(2). The manuscript attributes the enhanced superconductivity to a synergy of enhanced orbital hybridization, improved itinerancy, and stronger magnetic fluctuations. While the qualitative argument is sound, a more quantitative or at least qualitative analysis distinguishing the relative importance of these factors would be a significant addition. Furthermore, given the discussion of magnetic fluctuations as the likely pairing glue, a brief discussion on the likely symmetry of the superconducting pairing (e.g., d-wave or s-wave) and how it evolves under pressure, or a future outlook on this topic, would be highly beneficial for a comprehensive understanding.

(3). The theoretical model assumes that applied hydrostatic pressure primarily reduces the out-of-plane lattice constant, with the in-plane constant clamped by the substrate.
However, smaller in-plane lattice constant would also influence the electronic behavior, e.g., leading to enhanced intralayer interorbital hybridization. Could the authors provide a quantitative estimate of the actual strain state of the film under pressure? A more detailed justification for the assumption of a fixed in-plane lattice parameter would strengthen the connection between the theoretical model and experimental conditions.

(4). The Hall effect measurements indicate that the primary charge carriers are holes and that their density slightly increases with pressure. To substantiate this claim, it would be highly beneficial to include specific numerical data or estimations of the carrier concentration at various pressures. Additionally, the manuscript would be significantly enhanced by a qualitative discussion on how the theoretically predicted alpha, beta, and gamma bands, which are believed to cross the Fermi level, evolve under pressure. Specifically, which of these bands is most responsible for the observed hole-like nature of the carriers, and what is its role in contributing to superconductivity? This discussion would offer deeper insights into the microscopic electronic structure.

(5). The manuscript could benefit from a brief discussion on the potential influence of the interlayer Ni-O-Ni bond angle, as this factor has been shown to be crucial in other nickelate studies and may be relevant to the observed interlayer spin fluctuations.

(6). There are minor inconsistencies between some figures and the main text. For example, the peak of the superconducting dome in Figure 4 (nearly 7.5 GPa) appears slightly different from the value stated (9 GPa) in the text. While the regime plotting in Figure 4 is only approximate, it will still be helpful to make the phase diagram more consistent with the text.

Reviewer #3

(Remarks to the Author)
Li et al. presents a comprehensive investigation of enhanced superconductivity in compressively strained bilayer nickelate thin films under hydrostatic pressure. The authors demonstrate that the superconducting transition temperature can be enhanced from ~40 K at ambient pressure to over 61.5 K at 9 GPa, revealing a dome-shaped pressure-temperature phase diagram. The study combines experimental work with theoretical calculations to provide insights into the underlying mechanisms. Overall, the study addresses an important topic in the rapidly evolving field of nickelate superconductivity and the results are generally well presented.

I recommend this manuscript for publication in Nature Communications after addressing the following questions and suggestions:

1. Heterostructure design rationale: The authors employ a heterostructure design with 1.5 UC of $La_{2.5}Pr_{0.5}Ni_2O_7$ and 1.5 UC of $La_{2.85}Sr_{0.15}Ni_2O_7$ to enhance stability, but the specific choice of Sr doping concentration (x=0.15) and layer thicknesses appears somewhat arbitrary. Could the authors provide more detailed justification for these parameters? Were systematic studies conducted to optimize these values, or were they based on prior literature? Please provide more information.

2. Strain characterization limitations: While the authors claim compressive strain based on c-axis elongation from XRD 2θ-ω scans, this characterization is insufficient for a heterostructure containing two different compositions with potentially different lattice parameters. Reciprocal space mapping (RSM) or transmission electron microscopy (TEM) would provide more definitive evidence of the strain state and film quality. The current XRD data quality also appears limited for making strong claims about crystalline quality.

3. Hall coefficient interpretation: The positive Hall coefficients observed in this work contrast with recent literature reports showing negative Hall coefficients in Sr-doped bilayer nickelate films. Given that this heterostructure contains both Sr and Pr, could the authors provide a more detailed discussion of the charge carrier behaviour and how it relates to the superconducting mechanism in their specific system?

4. Data presentation: The resistance vs. temperature data should be converted to resistivity vs. temperature for proper materials characterization and cross-comparison with others' work. Additionally, experimental uncertainties should be included, particularly for the transition temperatures in the phase diagram.

5. Mechanisms: The manuscript describes two different trends in superconducting transition behaviour under pressure but does not clearly explain their origins. Could the authors elaborate on what physical mechanisms drive these different behaviours and how they relate to the theoretical calculations?

6. Reproducibility: While multiple samples are measured, it would be valuable to include a more quantitative discussion of sample-to-sample variation in superconducting properties at ambient pressure, particularly given the complexity of the heterostructure growth and post-annealing process.

Despite these questions, this work represents a significant contribution to understanding pressure effects in nickelate superconductors and successfully demonstrates substantial enhancement of transition temperatures through the combination of strain engineering and pressure tuning.

Reviewer #4

(Remarks to the Author)
I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
Dear Authors,
I appreciate the authors for revisions. However, there are still critical points that need to be revised. Please reconsider following comments.

1, As for comment 1, the response and revisions are not insufficient. The definition of the onset superconducting transition temperature should be reconsidered. It is still not fair to define the onset critical temperature as defined in the revised manuscript. Tc^cross shown in Fig. 2b should be defined as Tc^onset for a fair comparison. In addition, the definition of Tc^cross in supplementary Fig. 8 seems appropriate for applying the definition of Tc^onset as similar as previous studies. The data in Fig. 3b, however, can be interpreted very differently depending on the definition used. When the same definition as other papers is applied, the transition temperature shows almost no increase. Furthermore, the definition of Tc^cross in

Fig. 3b is clearly not valid. The authors should conduct a more fair analysis of the data. There is concern that changing the method of definition could alter the conclusion. For a fair and scientific discussion, it is important to use a comparable definition to that used in previously reported literature, such as ref. 13 and 40. The authors must revise the definition of the onset temperature.

2, The response to the comments 2 and 4, the comments of [Therefore, we may attribute the increase of normal state resistance to the damage to the electric connection of the film caused by the solidification of the pressure-transmitting medium.] and [Some of the conduction paths of the thin films may be partially destroyed due to change of the pressure environment, such as the solidification of the PTM, producing considerable amount of dislocations and defects on the films.] are provided. These comments appear to be an arbitrary interpretation that changes in the pressure medium cause abnormalities in resistance but have no intrinsic effect on the transition temperature. The basis for drawing this conclusion has not been sufficiently explained. If the dislocation is induced in the film by applying pressure, the pressure may not induce the lattice compression because the pressure may be relaxed by inducing dislocation. Under this situation, what does understanding the pressure dependence of the transition temperature lead to? Lattice compression may not be increased as pressure increases above 8 GPa. In Fig. 4, the decrease above the critical temperature of 8 GPa may be attributed to an extrinsic origin unrelated to the increase in pressure.
To explain the pressure dependence of resistivity for general readers, Figure R1 should be added in supplementary information.
If the authors claim this sentence [We may argue that the weak insulating behavior above Tc at high pressures is an intrinsic property of the bilayer nickelate thin films.] in line 281, the results should be compared to the bulk studies of bilayer nickelates although [Such behavior has been reported in the underdoped cuprates48, 49.] is commented.

3, As for comment 12, Applying pressure causes the thin film and substrate to compress isotropically, so it is reasonable to assume that the a-axis length of film is also compressed. The proposed calculation model seems not valid. Failure to explain the lattice deformation in the computational model to readers may lead to misunderstanding. It should be explicitly stated that the calculation assumes a fixed a-axis length and affection of this fixing.

4, Fig. R2 should be included in supplementary information. If the authors claim the stability of superconductivity in air, in addition to the description about the stability in air, the fragility by applying pressure should be also described in main text.

5, The claims in abstract seem not supported by results. The descriptions should be revised fair to the results.

The claim of [Here we report the enhancement of Tc onset from about 40 K to over 60 K] in abstract should be revised. By applying appropriate definition for fair comparison, judging from the data in Fig. 2b, the temperature seems roughly 30 K to over 40 K. In the abstract, the sentence [superconductivity with an onset transition temperature (Tconset) over 40 K was achieved in La3Ni2O7 and (La,Pr)3Ni2O7 thin films at ambient pressure due to in-plane compressive strain.] is also described. However, 40 K in the literature was estimated by an appropriate definition, but the author's definition is not appropriate. In the same abstract, it is a critical problem for comparison the onset temperature under different definitions. Such description misleads the general readers. The authors should revise their method of definition. The sentence at line 29 of the abstract in this manuscript should be revised to [Here we report the enhancement of Tconset from about 30 K to over 40 K].

The claim of [reaching the maximum Tc onset at about 61.5 K.] in abstract should be revised.
By applying appropriate definition for fair comparison, the maximum Tc onset in Fig. 3b seems roughly 50 K.

It would be better to revise the claim of [Our theoretical calculations show that the increase of Tc may be mostly described by the reduction of the c-axis lattice constant, which yields a cooperative enhancement of magnetic fluctuations within and between the layers and increased metallicity under pressure.], because he conclusion that the c-axis length is the cause is not logically valid in calculations where only the c-axis length is varied. Despite the theoretical calculation under hydrostatic pressure, the authors did not calculate any parameters other than the c-axis length in the first place.

Correspondingly to the revisions in abstract, the main text should be appropriately revised.

6, in line 138, [a relativity large size (3 mm x 1.5 mm) as shown in the inset of Fig. 1c] is added. In the inset, the scale bar is necessary to show the sample size.

7, in line 139, [the applied current is supposed to be uniformly distributed on the thin films.] is added in the revision. How did the authors judge uniformity of the current? Since it is rather difficult in general to characterize the current uniformity in sample, when providing such speculation, it is necessary to explain the experimental basis.

8, in line 364-370, the claim of the consistency in the sentence of [This is consistent with the Hall coefficient data shown in Fig. 2d, indicating stronger metallicity of the normal state upon applied pressure.] appropriate? The detection of insulating behavior in experimental results seems more important. In this point, the inconsistency seems more important to be noted in the discussion part.

9, in line 431-438, the extrinsic origin with inducing dislocation etc has to be also discussed.

Reviewer #2

(Remarks to the Author)
The authors have revised the manuscript significantly and its quality is improved. Their replies to all reviewers' questions are satisfactory and I recommend its publication in Nature Communication.

Reviewer #3

(Remarks to the Author)
The authors have adequately addressed all my concerns and made a lot of efforts to improve the quality of the data. I am satisfied with the revisions and recommend this manuscript for publication in Nature Communications.

Reviewer #4

(Remarks to the Author)
I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Version 2:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
Dear Authors,
I would like to thank the authors for their revisions.
The manuscript has been significantly improved, and it is believed that the revised version is quite well for general readers owing to a fair comparison with previous literature.

Responses to the Reviewers' report

(*The words in black color are from the reviewers´ reports, and the blue words give our responses*)

Responses to the Report of Reviewer #1

In this manuscript, the authors report on the effect of pressure on the superconductivity in bilayer nickelate thin films. This study is an important and timely exploration of the connection between the differences between superconductivity in bulk materials under pressure and it in compressively-strained thin films at ambient pressure. I agree that the sentence in discussion page 17 line 356 [The combination of strain engineering and high pressure is an essential tool in the research of nickelate superconducting thin films.] While the authors measured four samples under applied pressure, the results are quite complicated. The authors could improve the presentation of the data to make it easier to understand. Theoretical calculations provide some insight, but the discussion in this manuscript seems not sufficient to explain the complicated results.
To convince the general reader of the claims in this manuscript, it would be better to make major revisions to improve the paper, such as the addition of figures.

Response: We thank this reviewer for the careful reading and constructive suggestions. The reviewer provided several constructive suggestions, and we have modified our manuscript according to these suggestions/questions. We have made sufficient revisions to the manuscript, for example, we have modified Figs. 1-5 and Supplementary Figs. 3, 5, 7; we have added Supplementary Note 1 and 2, Supplementary Figs. 1a, 8, 9, 11 and many new descriptions and discussions. We hope this reviewer will be satisfied with our responses and revisions.

1. The definition of the onset superconducting transition temperature should be reconsidered. The definition applied in Fig. 1a in ref. 40, Y. Liu et al., Nature Materials 24, 1221 (2025). is most reliable and common method. This method should be applied to estimate the onset transition temperature. This method is also applied in Fig. 1 in ref. 13. To resolve the confusion regarding the transition temperature of bilayer nickelate thin films, it is appropriate to apply a reliable definition. In particular, the temperature values of broad transitions for Fig. 2b, Fig. 3b, suppl. Fig. S1, and Fig. S7b in this manuscript are strongly affected by

how to be defined. For example, linear approximation in the transition region can be defined as the range between 80% and 90% of normal resistance values. By adopting this method, it is possible to compare all data through fair analysis. The experimental observation of the enhancement under applied pressure itself is a main claim of this manuscript. The temperature values should be compared by fair definition.

To remove the confusion by the different definition between this manuscript and previous studies ref. 40 and ref. 13, the authors should revise the definition with applying identical method.

Response: We thank this reviewer for the constructive suggestion. We agree with the reviewer that the definition of T_c used in ref. 13 and 40 is good and is also one of the standard methods to define T_c. The standard "cross-point" method in the literatures is more suitable for superconductors with sharp superconducting transitions. In our present data, the superconducting transition width are relatively wide, and there is even no complete superconducting transition under higher pressure in CAC measurements. Therefore, we follow the suggestion of this reviewer, and define the linear approximation of the transition region by using the data between 80% and 90% of normal resistance values, and the T_c^{cross} is defined as the cross-point of this transition region and normal state linear extrapolation (see revised Fig. 2b, Fig. 3b and Supplementary Fig. 8). Then, we successfully get a new set of T_c^{cross} values with pressure dependence of S1 and S3, as shown in the revised Fig. 4. Although the values are lower than the T_c^{onset} in our initial manuscript, it seems that the pressure dependence of T_c^{cross} exhibits similar behavior as T_c^{onset} under pressure. In the revised version, we keep the two sets of data, namely T_c^{onset} and T_c^{cross} in the phase diagram for the readers to have a better judgement.

In the revised manuscript, we have revised Fig. 2b, Fig. 3b and added a new Supplementary Fig. 8 to illustrate the definition of T_c^{cross} and T_c^{onset}, despite the T_c^{cross} values are a bit lower in the revised phase diagram. We also made some necessary discussions for why we use both definitions to get T_c. We hope this reviewer is satisfied with our responses and related reversions to the manuscript.

2. There is no consistent explanation for the strange behaviour that the transition temperature increases with increasing the resistivity. This observation may come from the artifact by applying inadequate definition method discussed in comment 1.

While the increase of the transition temperature may be induced by increase of Fermi surface or other enhancement of interaction, the increase of the resistance likely comes from the reduction of charge carrier density and/or increase of scattering. If these phenomena occurs simultaneously in the sample, how did the authors explain consistently for both the increase of transition temperature and resistance? These phenomena are independently discussed in the present manuscript, but the consistent explanation is necessary to convince the claim to readers.

Response: We thank this reviewer for the careful review. We agree with the reviewer that the increase of T_c may be induced by increase of Fermi surface or the enhancement of some interactions. The better metallicity in the normal state in low pressure region (see Fig. 2a) may be induced by the enhanced itinerancy of the charge carriers. However, we want to point out that the abnormal increase of room temperature resistance ($R_{293\ K}$) may not reflect the intrinsic properties of the bilayer thin films under high pressure. For instance, as shown in Fig. S7, the pressure for the abnormal increase of resistivity (~4 GPa) is much lower than the pressure for the saturation of T_c (~8 GPa). Therefore, we may attribute the increase of normal state resistance to the damage to the electric connection of the film caused by the solidification of the pressure-transmitting medium. The reasons are as follows: (1) For different runs of experiment (S3 in Fig. 3 and S4 in Fig. S7) on our bilayer thin films, the evolution of superconducting transition temperature with pressure is relatively consistent with each other, while the critical pressure for the increase in normal state resistance is quite different. (2) Similar increase of normal state resistance under high pressure were also observed in the infinite layer thin film [see the data in ref. 46: Wang, N. N. *et al.*, Nat. Commun. 13, 4367 (2022)]. However, the T_c in 112 thin films is monotonically increased without saturation, which may indicate that there is no correlation between the change in T_c and absolute value of normal state resistance in thin films for the high-pressure measurement in CAC apparatus. Therefore, we believe that the abnormal increase of resistance in our thin films under pressure are most likely due to the extrinsic reasons. To address these issues more clearly for the readers, we have added some necessary explanations in the main text and a new Supplementary Note 2 in the revised Supplementary Information to discuss the possible reason for the abnormal enhancement of resistance in our high-pressure measurement.

3. The resistance values at 300 K for four samples in this study should be compared by plotting as a function of pressure to make clear the reproducibility of the pressure effect. Whether superconductivity occurs or not is an important point, but it is also important to compare the reproducibility in the resistivity of the normal state before that occurs. It is expected that the resistance variation by applying pressure should systematically occur via shorten lattice and increase of Fermi surface. If the authors claim the reproducibility, this point should be clearly explained with additional data plots.

Response: As we responded to the reviewer for comment 2, the abnormal increase of resistance values at room temperature ($R_{293\ K}$) may not reflect the intrinsic properties of our thin films under pressure. As shown in Fig. R1 below, we plot the pressure dependent room-temperature resistivity ($\rho_{293\ K}$) of the four thin films. It can be clearly seen that in low-pressure region, the room temperature resistivity decreases continuously and shows good reproducibility with each other, indicating a possible consequence of the shorten lattice constant and increase of Fermi surface of our samples under pressures as pointed out by the reviewer in this comment. However, in the high-pressure region, it shows an opposite behavior, we don`t have the intention to attribute the abnormal increase of resistivity with pressure to the intrinsic pressure effect of the bilayer thin films. There are several extrinsic factors that can affect the absolute resistivity of thin films when pressure is high. For example, the change of the pressure environment, the variation of the electrode contact, defects or disorders generated by pressure, etc. For our samples with different PTM (S3 and S4) and different samples with the same PTM (our samples and 112 thin films in ref. 46), the abnormal change of resistivity always occurs and the critical pressure for this phenomenon seems to be quite random. Therefore, in order to clarify this issue, we add a new Supplementary Note 2 and make necessary revisions to the main text in the revised manuscript.

We hope this reviewer will be satisfied with our revised version and the explanations concerning the abnormal change of room temperature resistivity under high-pressures.

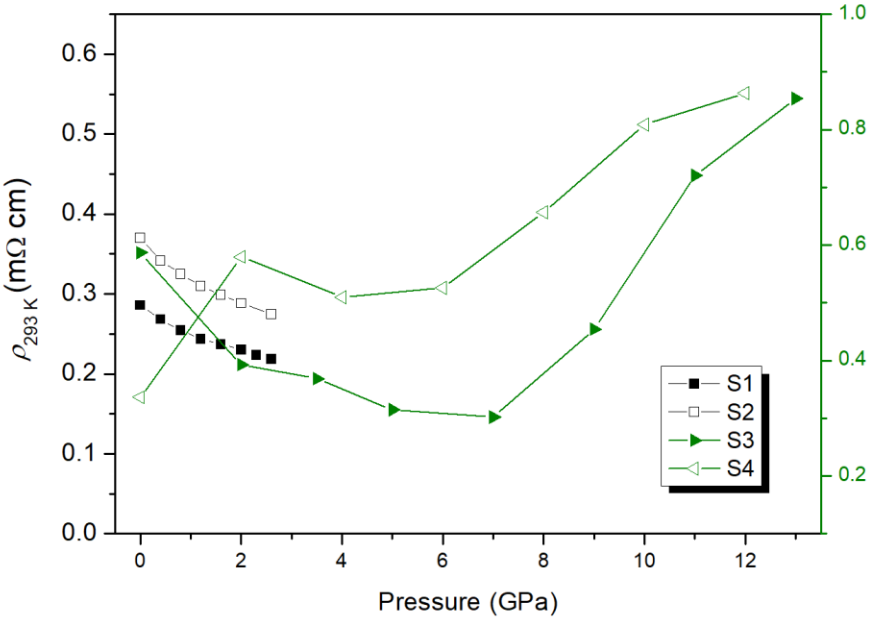

Fig. R1: Pressure dependent room-temperature resistivity ($\rho_{293\ K}$) for different LSP_{327} thin films. The abnormal increase of $\rho_{293\ K}$ from 0 to 2 GPa for S4 might be caused by changes in the electrode contact when applying PTM on samples.

4. In addition to the comment 2, can the simultaneous occurrence of increase of resistance and transition to the insulator be interpreted by theoretical band calculation? These behaviours were not observed in bulk studies. This point may not be explained in the present manuscript. While four samples in this study show similar behaviour, the difference between bulk results under pressure and that in this study should be discussed.

Intuitively, the applying pressure shrinks the lattice, corresponding to the increase of Fermi surface and in-plane hopping. It is likely that the resistance value decreases with increasing pressure up to a certain critical pressure condition, for example occurrence of structural phase transition. If such transition occurs in bilayer nickelate film, which will be also observed in bulk.

Response: We thank this reviewer for the careful review and this comment. As we responded to this reviewer in comments 2 and 3, the increase of resistance in the high-pressure region may be due to extrinsic reasons. Some of the conduction paths of the thin films may be partially destroyed due to change of the pressure environment, such as the solidification of the PTM, producing considerable amount of dislocations and defects on the films. Similar behavior can also be seen in the previous work on the infinite-layer nickelates in ref. 46: Wang, N. N. *et al.*, Nat. Commun. 13, 4367 (2022). For the appearance of weak insulating behavior above T_c in high pressure region, we can indeed get some information from theoretical calculation. Our calculations, however, also show that beyond some pressure values the electronic structure changes are minimal, reaching some kind of saturation. Therefore, we believe the weak insulating behavior in bilayer thin film may be induced by the gradually enhanced correlation effect, associated with the shift of the γ-pocket with a flat band feature, which intimately leads to the formation of the density wave order.

Although such insulating behavior is not observed in bulk samples under pressure, we want to note that, in thin film form of nickelate superconductors, the weak insulating behavior is also observed in Sr-doped $La_3Ni_2O_7$ with dome-like superconducting region [also see ref.39 and 45]. The different behavior between thin films and bulk crystals might be attributed to the completely different situations within the thin film and the bulk. The pressure induced structural disorders and dislocations may be prominent for these ultra-thin films but is less pronounce on the bulk samples. In bulk crystals, there is no initial strain and the superconductivity is induced by suppressing the weak insulating states and a structural phase transition occurs. However, in thin films, it is already superconductive at ambient pressure and the *c*-axis lattice constant is elongated due to the compressive strain given by substrate. Therefore, the electronic structure may be different from the bulk samples.

As for the possibility of structure phase transition, we think there is no direct indication for this transition in our films under pressure. Again, we believe that the increase of resistance in high pressures in our experiment is extrinsic, and it cannot serve as evidence for a possible structural phase transition.

In the revised manuscript, we have modified the related discussions regarding to the origin of weak insulating behavior in the point of theoretical calculations. We hope this reviewer will agree with our related revisions and changes to the manuscript.

5. In this study, four samples were measured and discussed under pressure. In Fig. 1d, the resistivity values of six films are plotted. But in other figures under

pressure, the resistance values are plotted. Were the resistance results, resistivity and resistance, consistent and reproducible? Judging from the high values of resistivities, the highest S6, S4, S2+35 days, S2, S5, S3, S1+35 days, and the lowest S1 are plotted. S6 and S1 is the highest and lowest, respectively. However, the values for 600 Ohm of S3 in Fig. 3, 300 Ohm of S4 in Fig. S7, 200 Ohm of S1 in Fig. 2, and 150 Ohm of S2 in Fig. S3 are not consistent to the resistivities shown in Fig. 1. If this difference comes from the sample geometry, the authors should present the photo of all samples or its summary in tables to explain the geometrical difference. If the sample size and geometry are comparable, the authors have to explain the origin for this inconsistent resistivity / resistance shown in the Figures.

Response: We thank the reviewer for reminding us of this issue. Since the space for high pressure measurement in PCC is much larger than that in CAC, the sample sizes in Fig.2 and Fig. S3 are much larger than that in Fig. 3 and Fig. S7. That is why the resistance value is bigger in CAC measurement. In the revised manuscript, we have converted the resistance data into resistivity in revised Fig.2(a-c), Fig. 3; Supplementary Fig. 3, Fig. 5, Fig. 7-9 for the consistency of data. We can see that the data present in the revised figures are better normalized. The little variations for the values of resistivity in Fig .1d and those measured under high pressure is inevitable. The possible reasons are listed as follows: (1) The inhomogeneity of thin films. since we have remade the electrodes during the high-pressure measurements, the slight variation of the values of resistivity is acceptable if the thin films have some kinds of inhomogeneity. (2) The slight degradation of the film. In CAC measurements, we need to greatly reduce the thickness of the substrate by polishing, which could affect the resistivity of the samples. We have added this information to the main text in revised manuscript.

6. Is the comparison with infinite layer nickelate meaningful? In page 3 line 49, the authors describe that [This is strongly contrasted to the 3d^9 configuration in cuprates and infinite-layer nickelates.]. Why do the authors compare their results under pressure with infinite layers when they believe that the electronic structure and the origin of superconductivity are clearly different?

Response: We agree with the reviewer on this issue and have no intention to have a comparison with infinite layers. Thus, we have removed the related sentence and references in the revised manuscript.

7. in page7, line 150, what does the reference 1 and 14 mean? Refs 1 and 14 correspond to bulk studies. In addition, in page 6 line 116-117, 20.75 A is elongated. The authors should describe the bulk value of c-axis length at ambient pressure. What does [abnormal elongation] mean? The comparison with the c-axis in previous film studies may be meaningful.

Response: We thank the reviewer for these useful suggestions. In the revised manuscript, we have added the *c*-axis lattice constants of bulk crystals at ambient pressure and compare the *c*-axis lattice constant with previous reported films. The expression “abnormal elongation” is also revised since it was improperly used in the old version of the main text.

8. in page 9 line 193, [The slight increase of R_H above 2.2 GPa may be induced by the solidification of PTM. Such behavior indicates that the carrier concentration increases upon applying a pressure.] These explanations seem too simplified. How does the solidification of PTM affect the R_H of the film? Is this behavior observed in other materials by applying identical PTM? Moreover, as described in page 6 line 135, the R_H of the bilayer nickelates is more complicated due to the multiband nature of the electronic structure of bilayer nickelates. The increase of R_H does not simply indicate the increase of carrier concentration. The authors should provide a unified explanation throughout the text.

Response: We thank the Reviewer for reminding us of this issue. As we discussed in the response to comment 2, the solidification of PTM might induce large number of dislocations or defects in the films, which further affects the resistance value of the films. However, this may not have clear effect on the Hall resistivity since the ρ_{xy} is only related to the thickness of thin films. In this sense, we agree with the reviewer that the solidification of PTM may not affect the R_H of the film. Considering the multiband nature of the electronic structure of bilayer nickelates, we also agree with the reviewer that the decrease of R_H should not directly indicate the increase of carrier concentration. In multiband system, the carrier concentration is also related to the change of carrier mobility and the electronic scattering mechanism. Thus, the nonmonotonic change of R_H under pressure in Fig. 2d may be related to the modification of the electronic band structure or the multiband contribution of the electron and hole charge carriers.

In the revised manuscript, we change the related expressions on the pressure dependent R_H and try to give a unified explanation for the R_H data. We hope our related reversions can remove the concerns of the reviewer on this issue.

9. in page 10 line 208, honestly speaking, it is rather difficult to agree that the description [Although the temperature dependence of normal-state resistance exhibits a drastic evolution, the superconducting onset transition temperature dependence shows a clear enhancement at relative low pressures.] because the shift of transition is small in Fig. 3a. Moreover, the description in line 216-217 [the R(T) curves display nearly similar evolutions as sample S3 under high pressures, which first shows a decrease from 2 to 4 GPa and then increases gradually with further increasing pressure.] is not fair. The authors have to be fair. For sample S3, the resistance firstly increases from 0 GPa to 2 GPa. Why did the authors only explain the behavior from 2 to 4 GPa not from 0 to 2 GPa?

Response: We thank this reviewer for the critical reading to the data. As we discussed above, the change of resistance value under pressures may be affected by many extrinsic factors, such as the change of pressure environment due to the solidification of PTM, or the generation of defects in the sample under pressure, *etc.* The unusual change in resistivity of S4 from 0 to 4 GPa may be due to the changes in the electrode contact when applying PTM on samples. Therefore, because there are too many extrinsic factors that can affect the absolute resistivity of thin films, we cannot reach a reconciled conclusion on the nonmonotonic change of the resistance in the present stage.

According to the reviewer`s comments and suggestions, we modified the descriptions in Page 10 line 208 and 216 and also added a new Supplementary Note 2 in the revised version to clarify the related issues concerning the possible reasons for the nonmonotonic change of resistivity of thin films under pressure.

10. What does this description [As is known, the delicate nickelate thin films can be easily and partially deteriorated at high pressures when the liquid PTM solidifies, which would produce disorders and defects in the thin film that will enhance the carrier scattering.] mean? Probably, no one knows that the bilayer nickelate film is easily deteriorated by applying pressure because there are less studies. Many bulk crystals show reproducible results under pressure. In addition, in ref. 54, there is no similar deterioration. If the authors would like to conclude that the films in this study are easily deteriorated, this would suggest that they were inferior specimens with insufficient crystal quality to withstand the pressure application experiments. The authors may need to reconsider the pressure effect once the quality of thin film samples reaches a level comparable to that of bulk crystals.
Alternatively, the authors should clearly describe the upper limit of pressure at which the intrinsic property of the nickelate films can be discussed in this study. To remove the confusion to general readers, it would be better that the authors describe the observed results above the upper limit come from the extrinsic effect or PTM effect.

Response: We thank this reviewer for the careful review and this comment. We are sorry that the word “deteriorated” was improperly used and we have revised it in our revised version. Under high pressures, some defects or disorders can be introduced in the thin films due to the change of pressure environment. At high pressures on a very thin film, some micro-cracks, disorders and dislocations may appear, which can naturally cause the increment of resistance. However, these defects, dislocations and even micro-cracks might not give strong influence on the electronic structures, and thus the appearance of superconductivity.

As we clearly discussed in the new Supplementary Note 2 of the revised manuscript, the PTM induced disorders can only affect the values of resistance as it may disrupt the conducting channels in certain areas of the thin films.

However, the temperature dependence of resistance and the superconducting transition temperature should not be affected, and these properties always reflect the intrinsic behavior of our bilayer thin films in the whole pressure range we measured. Therefore, the evolution of T_cs and weak insulating behavior under pressure seem to be true and repeatable experimental results.

11. in page 13 line 277, how does the solidification affect the electronic structure of the film?

Response: Thanks for the question. We think the solidification of PTM will not affect the electronic structure of the film. It merely introduces defects and micro-cracks on the film surface, thereby affecting the normal state resistance. Therefore, we didn`t discuss this issue in our original manuscript. However, we added one sentence in the revised version to stress on this issue.

12. in page 13 line 288, only c-axis length is defined. How is the a-axis length defined in the calculation model? If the authors discuss the pressure effect, the a-axis should be also compressed as increasing pressure.

Response: We thank the reviewer for pointing out this issue. For our bilayer nickelate films we emulate applied pressure by compressing the out-of-plane lattice parameter *c* while keeping the in-plane lattice constant *a* fixed at its ambient pressure value. This choice reflects the experimental geometry: the film is epitaxially clamped to a rigid substrate so the in-plane lattice is set by the substrate (and is not free to relax under moderate external pressure), while the film is free to change its interlayer spacing. Physically, the dominant pressure-driven changes in the electronic structure of layered nickelates arise from modifications of interlayer coupling, apical-oxygen distances, and Ni-O bond angles — all of which are controlled primarily by *c* and by internal atomic relaxations. Computationally, fixing a while fully relaxing internal coordinates for each *c* gives the correct symmetry and the principal microscopic degrees of freedom affected by pressure, and it also allows straightforward comparison to film experiments where in-plane clamping is present.

13. in page 14 line 303, Fig. 5d is firstly explained in main text. To improve the readability, it would be better that the authors either change the order of the figures or change the order of the explanations in the text.

Response: We thank the Referee for this comment, and followed his/her recommendation, Fig. 5 is updated accordingly. We hope this reviewer will agree with the revised version.

14. in page 17 line 360-371, it is difficult to agree that the discussion in this study. Are the claims in this discussion firstly found in this manuscript? Because no reference is added, it would be better to check this point again. Moreover, it would be better to discuss here the consistency between film and bulk. If the

interplay between interlayer and intralayer electronic correlation induces the reduction of the transition temperature, why this reduction has not been observed in bulk under pressure?

Response: We thank the Reviewer for the comment. Certainly, the referee is right that our formulation was very general and many relevant points were discussed by multiple groups, which we also cited throughout the manuscript. Thus, we have modified now this paragraph by explicitly mentioning difference between the thin films and the bulk. In particular, in strained thin films, external pressure acts on a lattice already clamped by the substrate, so the in-plane lattice constants and the in-plane Ni-O-Ni bond angle remain largely fixed while compression is mostly accommodated along the out-of-plane direction. On the contrary, in the bulk crystals there is a remarkable correspondence between the in-plane Ni-O-Ni bond angle and the T_c value once the structural transition to the *I4/mmm* phase takes place, see for example Fig.1 in B. Samanta, and A.B. Georgescu, arXiv: 2506.11427. Note, in compressed thin films the in-plane bond Ni-O angle is more bent than those achieved under hydrostatic pressure, correlating with a lower T_c. Thus, our hypothesis is that in thin films pressure mostly affects the *c*-lattice constant, while keeping in-plane Ni-O-Ni bond angle fixed. Thus, the effect of the hydrostatic pressure on the bulk and thin films is largely different.

Indeed, the reduction of transition temperature in bulk crystals under high pressure were observed. In ref. 1 and 21, the reported T_c value always decreases with increasing pressure in bulk bilayer nickelate samples. As we mentioned above, the pressurizing effect on the lattice constants in thin films are very different from the bulk samples, thus the T_c dependence of the thin films and bulk samples should have some differences. In the revised Discussion section, we have re-formulated the corresponding sentences in the conclusion.

15. in page 18, line 389, although the transition temperature in ref. 54 seems similar, the overall behaviour under pressure seems completely different, no insulating behaviour appear in ref. 54. What is the origin of this difference?

Response: We thank the reviewer for the careful review and this comment. We think the origin of the difference between our data and that in ref. 54 (ref. 55 in the revised version) is because of the different strains applied on thin films. This will induce significant difference of electronic band structures and transport properties. In ref. 54, the films are grown on substates with tensile strains ($SrTiO_3$: +1.9%; $NdGaO_3$: +0.6%) or slightly compressive strains ($LaAlO_3$: -0.5%) comparing to our present study ($SrLaAlO_4$: -2%). The ground states of $La_3Ni_2O_7$ thin films in ref. 54 is weak insulating with a density wave like transition, while our film has already become superconducting at ambient pressure. Therefore, the different behavior of our films and that in ref. 54 under high pressure are understandable. We believe that by applying higher pressure to the thin films in ref.54, the behavior of weak insulation above T_c might also occur.

16. in page 20 line 424, a five-probe method is described. The sample photo should be provided to explain the measurement configuration. Was the current uniformly applied to the small (0.6 mm x 0.3 mm) sample with five electrodes?

Response: We thank this reviewer for the suggestion. We have provided the sample photo showing standard five-probe electrode configuration as an inset in the revised Fig. 1c. The Hall coefficient measurements are done in the piston–cylinder cell, where the sample size is much bigger than that used in CAC. The typical size for our measurement is 3 mm x 1.5 mm, and the current is supposed to be uniformly applied on the thin films. In the revised version, we have added the sample photo and necessary explanation for the Hall measurement.

17. It would be better to reconsider the offset for the resistance data shown in Fig. 3b and Fig. S7, because the minus value of resistance seems strange. Moreover, the offset value should be explicitly explained if it is added to the data. In general, the equal offset is added to the data.

Response: We thank the reviewer for this comment. According to this suggestion, we have replotted the Fig. 3b and Fig. S7 to avoid the minus value of resistance in the revised manuscript. In order to prevent the curves from intersecting with each other and thereby affecting the visualization of the superconducting transition, we made necessary vertical shifts to the curves except for the 0 GPa one. The offset of the curves under high pressure is non-equal in each increment.

18. It would be better to reconsider the color of the data shown in Fig. 1d, Fig. 3, and Fig. S7. It is difficult to recognize the correspondence of data.

Response: We agree with the reviewer for this suggestion. In the revised version, we have changed the colors in Fig. 1d, Fig. 3 and Fig. S7. We think the new colors are much easier to distinguish from each other. We hope the reviewer is satisfied with these modifications.

19. in page 11 line 244, it is difficult to agree with [relatively sharp] for the data of 13 GPa. If the authors quantitatively discuss the sharpness of transition, it would be helpful to understand the evaluation standard.

Response: We thank the reviewer for the careful review. Due to the pressure induced weak insulating behavior and lose of zero resistance state under high pressures in Fig. 3, it is difficult to quantitatively describe the sharpness of the transition. In our present study, we focus more on the onset transition temperature rather than the sharpness of the transition itself. Therefore, we agree with this reviewer that it is inappropriate to say the superconducting transition is relatively sharp in Fig.3. We think a more accurate description would be: “a clear resistance drop”. In the revised manuscript, we have carefully checked the descriptions concerning the superconducting transitions and make some necessary reversions.

20. the bilayer heterostructure $La_{2.5}Pr_{0.5}Ni_2O_7/La_{2.85}Sr_{0.15}Ni_2O_7$ is applied to this pressure study. The basic resistance results of single films of 3 unit-cell $La_{2.5}Pr_{0.5}Ni_2O_7$ and 3unit-cell $La_{2.85}Sr_{0.15}Ni_2O_7$ are quite helpful to understand the effectiveness of the bilayer heterostructure. It would be better to present those results in supplement.

Response: We thank the reviewer for the suggestion. As we addressed in our manuscript, the bilayer heterostructure $La_{2.5}Pr_{0.5}Ni_2O_7/La_{2.85}Sr_{0.15}Ni_2O_7$ are pretty stable in air conditions, which enables us to conduct subsequent investigation on the transport properties of the bilayer nickelate thin films under pressure in different labs with different high-pressure cells. Indeed, we also fabricated the pure 3-UC $La_{2.85}Sr_{0.15}Ni_2O_7$ and $La_2PrNi_2O_7$ thin films and other heterostructures using the similar growth process, the *R(T)* curves are shown in Fig. R2. We can see that the superconducting transition is absent in those Sr doped sample and heterostructures, and $La_2PrNi_2O_7$ are pretty easy to deteriorate in air. Therefore, we cannot measure the high-pressure resistance on these samples at present stage.

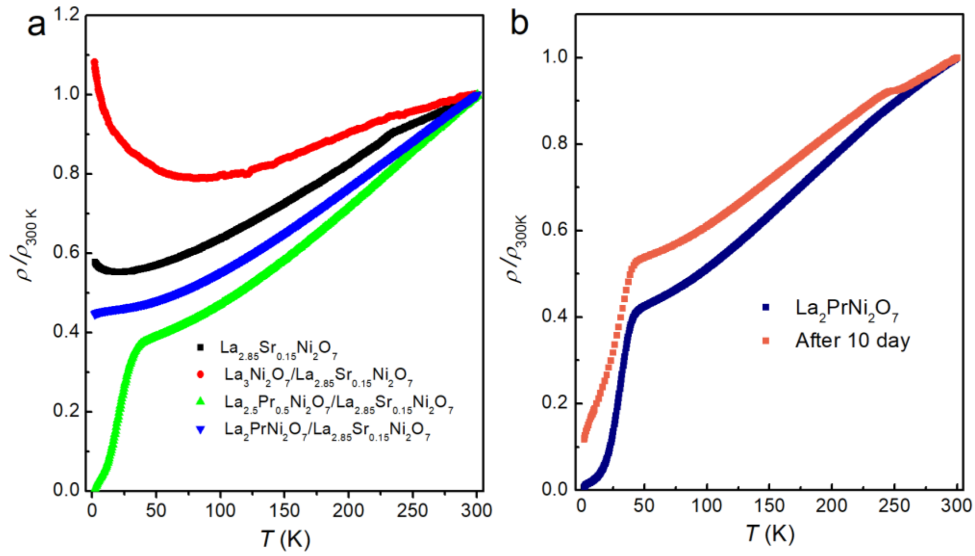

Fig. R2: **a,** The normalized *ρ (T)* curves of pure $La_{2.85}Sr_{0.15}Ni_2O_7$ and other heterostructures. **b,** The normalized *ρ (T)* curves of $La_2PrNi_2O_7$ and after 10 days.

We would like to thank this reviewer for the careful review and the constructive comments/suggestions. With the point-to-point responses and related revisions, we hope this reviewer will make a positive recommendation to our work for publication.

Responses to the Report of Reviewer #2

This is an excellent manuscript with a significant contribution to the field of high-temperature superconductivity in nickelates $La_3Ni_2O_7$. The authors successfully demonstrate a pressure-induced enhancement of superconductivity in compressively strained thin film samples, revealing a dome-shaped T-P phase diagram. The combination of high-quality experimental data and insightful theoretical analysis provides a compelling case for the proposed mechanism. I believe this work is highly suitable for publication and recommend its acceptance following a thorough and careful revision based on the following comments.

Response: We appreciate the careful review and positive remarks to our manuscript by this reviewer. We have revised the manuscript according to his/her constructive comments and suggestions as listed below. The revised manuscript is greatly improved according to these suggestions, and we hope the reviewer will be satisfied with the revised version.

1. The observation of a transition from metallic to weak insulating behavior at pressures above the optimal 9 GPa is a critical and intriguing finding. To fully substantiate this phenomenon and provide deeper insight, a more detailed analysis is required. It would be invaluable to see the full resistance-temperature curves at pressures both below and above this transition point. A discussion on whether this behavior is linked to an intrinsic structural or electronic phase transition, such as a charge density wave or Fermi surface reconstruction, would be highly valuable. Are there any other signatures or characteristics of this weak insulating behavior (e.g., a change in the resistance slope, hysteretic behavior, or magnetoresistance behavior) that could provide additional insights? Providing a more detailed picture of the normal state properties in this high-pressure regime will greatly strengthen the manuscript's conclusions.

Response: This is a very good question. The transition from metallic to weak insulating behavior can be repeatedly observed in different runs of resistive measurements with the high-pressure CAC apparatus. The full temperature dependence of resistance is shown in Fig. 3**a**, one can see that this transition from metallic to weak insulating behavior occurs at about 9 GPa. From these curves, the slope of resistance versus T above T_c can be easily determined. As shown in the Fig. R3 below, the slope of the *R(T)* curves above T_c in samples S3 and S4 changes from positive to negative when entering the weak insulating state after about 9 GPa. We don't know whether this weak insulating behavior is related to the formation of any density wave order, but the phenomena can be easily repeated. We also use a magnetic field to suppress the superconductivity, the “normal state” recovered by suppressing superconductivity consistently show the transition from a metallic to a weak insulating behavior and the magnetoresistance in the normal state is negligible (see Supplementary Fig. 9d-f in the revised version). The general behavior of the semiconducting part looks like that in underdoped cuprates [R1, R2]. We then also fit the low-T data just above T_c, indeed a $\rho \propto -\ln T$ relation is found (see Fig. R4 and Supplementary Fig. S8a-c in the revised version). Thus, we think that the weak insulating behavior may be induced by the gradually enhanced correlation effect.

In addition, as explained in the response to the Reviewer #1, we believe that the observed weakly insulating behavior may be associated an intrinsic density-wave (DW) instability, which becomes increasingly relevant at the Fermi level under compression.

Furthermore, we have performed Hall coefficient calculations within standard Boltzmann theory using the obtained electronic structure (see Fig. R5 and also Supplementary **Fig. 11** in the revised manuscript). In the oxygen-rich (clean) case, the Hall coefficient at ambient pressure is slightly negative, consistent with the results of Ref. [arXiv:2508.15284, Fig. 3]. Upon increasing pressure, however, the Hall coefficient turns to be positive, reflecting the stronger contribution of the γ-band at the Fermi level. Taken together with the possible oxygen deficiency present in the films, potential DW instability associated with the movement of γ-band may account for the results we observed experimentally.

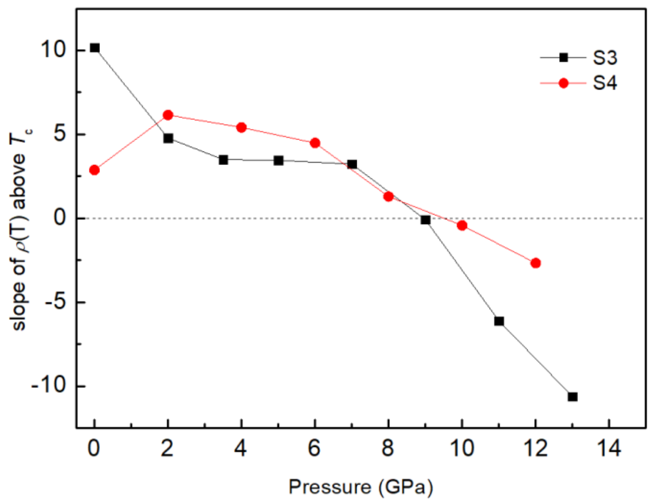

Fig. R3: The slope of ρ(T) curves just above T_c for S3 and S4. We can see that the slope changes from positive to negative at around 9 GPa.

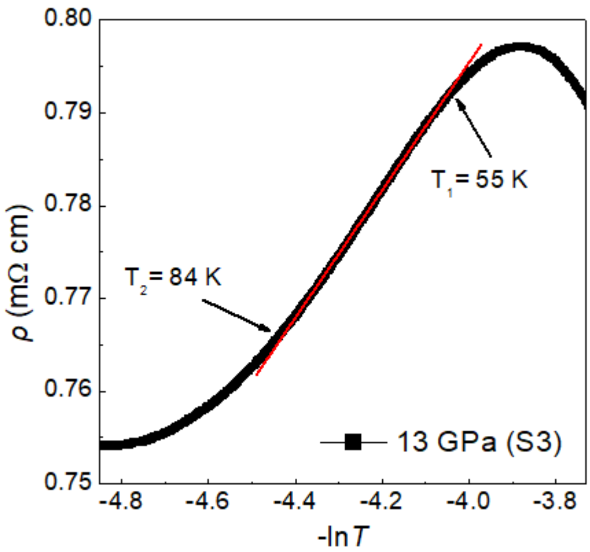

Fig. R4: The ρ versus -ln*T* curve for S3 at 13 GPa. In the temperature range from 55 to 84 K, the plot can be fitted to the linear relationship (red line).

[R1] Ando, Y. *et al.* Logarithmic divergence of both in-plane and out-of-plane normal-state resistivities of superconducting $La_{2-x}Sr_xCuO_4$ in the zero temperature limit. *Phys. Rev. Lett.* **75**, 4662–4665 (1995).
[R2] Ono, S. *et al.* Metal-to-insulator crossover in the low-temperature normal state of $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$. *Phys. Rev. Lett.* **85**, 638–641 (2000).

2. The manuscript attributes the enhanced superconductivity to a synergy of enhanced orbital hybridization, improved itinerancy, and stronger magnetic fluctuations. While the qualitative argument is sound, a more quantitative or at least qualitative analysis distinguishing the relative importance of these factors would be a significant addition. Furthermore, given the discussion of magnetic fluctuations as the likely pairing glue, a brief discussion on the likely symmetry of the superconducting pairing (e.g., d-wave or s-wave) and how it evolves under pressure, or a future outlook on this topic, would be highly beneficial for a comprehensive understanding.

Response: We are thankful to the referee for this comment. The stronger hybridization between the Ni-e_g orbitals can be quantified by the reduced crystal-field splitting as shown in the table below:

$\Delta_{3d_{z^2}-3d_{x^2-y^2}}$ (in meV)	$\Delta c/c_0$ (in %)
451	0
391	1.5
316	3.0
184	5.0

Furthermore, the data is for the DFT electronic structure and this orbital balancing with pressure is further enhanced with correlations. As a result, there is charge flow from Ni-$3d_{z^2}$ to Ni-$3d_{x^2-y^2}$ orbital, enhancing the presence of the gamma pocket at the Fermi level. This strengthens the tendency towards s+- wave Cooper-pairing. We have added the corresponding sentences in the main text.

3. The theoretical model assumes that applied hydrostatic pressure primarily reduces the out-of-plane lattice constant, with the in-plane constant clamped by the substrate.
However, smaller in-plane lattice constant would also influence the electronic behavior, e.g., leading to enhanced intralayer interorbital hybridization. Could the authors provide a quantitative estimate of the actual strain state of the film under pressure? A more detailed justification for the assumption of a fixed in-plane lattice parameter would strengthen the connection between the theoretical model and experimental conditions.

Response: This remark is similar to the comment 12 of the Referee #1. As pointed above, in the bulk crystals there is a remarkable correspondence between the in-plane Ni-O-Ni bond angle and the T_c value once the structural transition to the *I4/mmm* phase takes place, see for example Fig.1 in B. Samanta, and A.B. Georgescu, arXiv: 2506.11427. This means that it is the in-plane Ni-O-Ni bond angle, which largely controls T_c in the bulk crystals. In the strained thin films, the situation is different, external pressure acts on a lattice already clamped by the substrate (especially there are only few unit cells in thin films), so the in-plane lattice constants and the in-plane Ni-O-Ni bond angle remain largely fixed (here we refer again to the arXiv:2506.11427), while compression is mostly accommodated along the out-of-plane direction. Along the *c*-axis, there is the bilayer segment and the LaO spacer segment and therefore much more susceptible to changes with pressure. The smaller, and thus "more homogeneous", *a*-axis with the stiff transition metal-O bonds is not changed by pressure in equal terms. The small changes that still happen should work somewhat against the orbital balancing trend described above. Therefore, we assumed the dominant effect from pressure compared to the ambient state is driven by the *c*-axis change. Thus, the effect of the hydrostatic pressure on the bulk and thin films is largely different. We have re-formulated the corresponding sentences in the conclusion.

4. The Hall effect measurements indicate that the primary charge carriers are holes and that their density slightly increases with pressure. To substantiate this claim, it would be highly beneficial to include specific numerical data or estimations of the carrier concentration at various pressures. Additionally, the manuscript would be significantly enhanced by a qualitative discussion on how the theoretically predicted alpha, beta, and gamma bands, which are believed to cross the Fermi level, evolve under pressure. Specifically, which of these

bands is most responsible for the observed hole-like nature of the carriers, and what is its role in contributing to superconductivity? This discussion would offer deeper insights into the microscopic electronic structure.

Response: We thank the reviewer for the suggestion. We followed the Referee's recommendation and present in Fig. R5 the calculated Hall coefficient, modeled within a quasiparticle approximation to the electronic states, as a function of pressure. At ambient pressure, the Hall coefficient is small and negative, in remarkable agreement with the values reported for oxygen-rich samples by Maosen Wang *et al.* (arXiv:2508.15284). With increasing pressure, the Hall coefficient changes sign and becomes positive, reflecting the enhanced contribution of the hole-like γ pocket at the Fermi level and a stronger tendency toward $s^{\pm}$ superconductivity.

Experimentally, however, we consistently observe a positive Hall coefficient. By having a comparison with Ref. 45 [Maosen Wang *et al.*, arXiv:2508.15284], this suggests the presence of a certain degree of oxygen deficiency (interstitials) in our samples. Since the α and β bands contribute with opposite signs to the Hall coefficient and the γ-band contribution is relatively small, oxygen deficiency may dominate the pressure evolution of the Hall response. Consequently, the experimental Hall data should not be interpreted solely as a direct signature of electronic structure evolution under pressure. To achieve a more conclusive analysis, studies on next-generation, higher-quality films will be required.

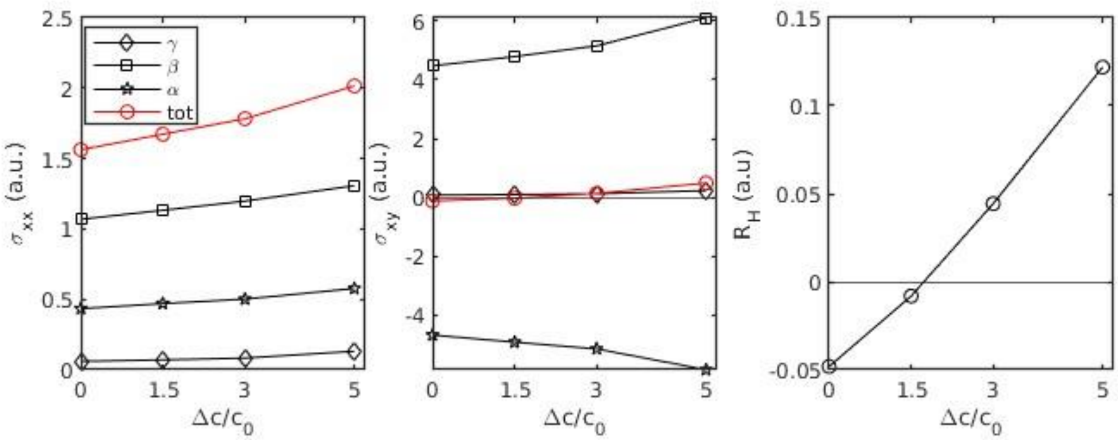

Fig. R5. Calculated quasiparticle contribution to the longitudinal (left panel), transverse (central panel) conductivities, including the individual band contributions and the total Hall coefficient (right panel) at T = 60 K as a function of applied pressure. We employed the Kubo formalism, developed for multiorbital nickelates for the slave-boson renormalized electronic structure, as described in ref. R3.

[R3] S. Bötzel, I.M. Eremin and F. Lechermann, *Phys. Rev. B* **107**, 174526 (2023).

5. The manuscript could benefit from a brief discussion on the potential influence of the interlayer Ni-O-Ni bond angle, as this factor has been shown to

be crucial in other nickelate studies and may be relevant to the observed interlayer spin fluctuations.

Response: We thank this reviewer for raising this interesting point. We agree with the reviewer that the interlayer Ni-O-Ni bond angle is crucial for the occurrence of superconductivity in nickelates. However, at this moment, we cannot do the synchrotron diffraction measurement on the thin films under pressure to determine the Ni-O-Ni bond angle. The in-plane lattice is determined at ambient pressure to be about 3.75 Å, which is about -2% strained compared with the bulk value. The *c*-axis lattice constant is accordingly elongated. Since the thin films are already superconductive at ambient pressure, the Ni-O-Ni bond angle was supposed to be close to 180° as the bulk samples in the tetragonal phase. Under pressures, we may expect that the Ni-O-Ni bond length will shrink.

In theoretical modeling we assume a full tetragonal symmetry of the system. Note that the effects of finite Ni-O-Ni bond angle is discussed extensively by B. Samanta, and A.B. Georgescu, arXiv: 2506.11427 and we include the reference to their paper for consistency.

In the revised manuscript, we have some discussion on the interlayer Ni-O-Ni bond angle under pressure and its possible relations to the interlayer spin fluctuations. We hope the reviewer will satisfied with those reversions and discussions.

6. There are minor inconsistencies between some figures and the main text. For example, the peak of the superconducting dome in Figure 4 (nearly 7.5 GPa) appears slightly different from the value stated (9 GPa) in the text. While the regime plotting in Figure 4 is only approximate, it will still be helpful to make the phase diagram more consistent with the text.

Response: We thank this reviewer for the careful review and this suggestion. In the revised version, we have corrected these inconsistencies.

We hope this Reviewer is satisfied with our point-to-point responses and related revisions, and will make a recommendation to our work for publication in Nature Communications.

Responses to the Report of Reviewer #3

Li et al. presents a comprehensive investigation of enhanced superconductivity in compressively strained bilayer nickelate thin films under hydrostatic pressure. The authors demonstrate that the superconducting transition temperature can be enhanced from ~40 K at ambient pressure to over 61.5 K at 9 GPa, revealing a dome-shaped pressure-temperature phase diagram. The study combines experimental work with theoretical calculations to provide insights into the underlying mechanisms. Overall, the study addresses an important topic in the rapidly evolving field of nickelate superconductivity and the results are generally well presented.

I recommend this manuscript for publication in Nature Communications after addressing the following questions and suggestions:

Response: We thank this Reviewer for the careful review and positive remarks on our work. We also thank the reviewer for the useful and constructive comments/suggestions which have been well addressed in the revised version. We hope the reviewer will be satisfied with our related responses and reversions.

1. Heterostructure design rationale: The authors employ a heterostructure design with 1.5 UC of $La_{2.5}Pr_{0.5}Ni_2O_7$ and 1.5 UC of $La_{2.85}Sr_{0.15}Ni_2O_7$ to enhance stability, but the specific choice of Sr doping concentration (x=0.15) and layer thicknesses appears somewhat arbitrary. Could the authors provide more detailed justification for these parameters? Were systematic studies conducted to optimize these values, or were they based on prior literature? Please provide more information.

Response: We thank this reviewer for the careful review and this comment. The heterostructure designed in our present study are quite important for the investigation of pressure effect on bilayer thin films. We found that with other heterostructures, the thin film samples will either lack superconductivity or the superconductivity will easily deteriorate in air (see the Fig. R6 below). The films with this heterostructure were deposited by using the PLD technique with two different targets $La_{2.5}Pr_{0.5}Ni_2O_7$ and $La_{2.85}Sr_{0.15}Ni_2O_7$. The deposition and post-annealing conditions have been optimized by having better superconducting transition at ambient pressure (see Supplementary Note 1 in the revised version).

Actually, the values of layer thickness and Sr doping concentration in our present study are mainly based on the previous literatures, and we haven`t conducted a systematic study to optimize the thickness and doping concentration yet. We prepared the sample with Sr doping concentration x =

0.15 because the thin films of $La_{3-x}Sr_xNi_2O_7$ exhibited the best superconductivity around this doping level (see ref. 39 in the revised version). Furthermore, in ref. 37 and 39, the superconducting thin films are all fabricated with the thickness of 3-UC. Based on this information, we finally decided to grow the bilayer thin film with a heterostructure of $La_{2.5}Pr_{0.5}Ni_2O_7/La_{2.85}Sr_{0.15}Ni_2O_7$ with a total thickness of 3-UC in our present study. In the revised manuscript, we have given some necessary explanations for the choice of Sr doping concentration and layer thicknesses.

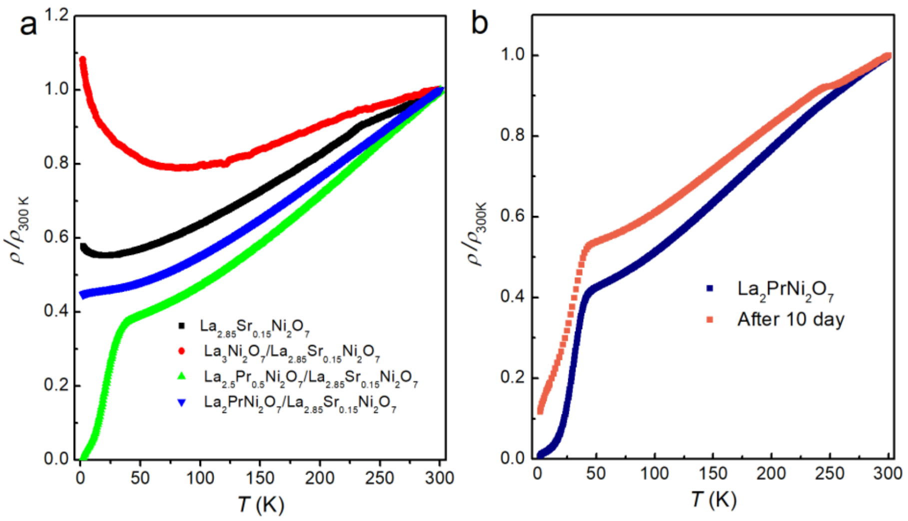

Fig. R6: **a,** The normalized *ρ (T)* curves of pure $La_{2.85}Sr_{0.15}Ni_2O_7$ and other heterostructures. **b,** The normalized *ρ (T)* curves of $La_2PrNi_2O_7$ and after 10 days.

2. Strain characterization limitations: While the authors claim compressive strain based on c-axis elongation from XRD 2θ-ω scans, this characterization is insufficient for a heterostructure containing two different compositions with potentially different lattice parameters. Reciprocal space mapping (RSM) or transmission electron microscopy (TEM) would provide more definitive evidence of the strain state and film quality. The current XRD data quality also appears limited for making strong claims about crystalline quality.

Response: Thanks for the professional comment and suggestion. We agree with the reviewer that the direct evidence of compressive strain is needed. In the revised version, we present the newly measured Reciprocal space mapping (RSM) of our bilayer LSP_{327} thin films on SLAO substrate and present the data in Fig. S1a and Fig. R7 below. The results confirm coherent epitaxial strain between the film and substrate with an in-plane lattice constant of 3.75 Å, and the strain state of the film corresponds to in-plane misfit strain about −2 %. Moreover, we also improve the XRD data of our samples and updated it in Fig. 1b. All these data show that our LSP_{327} film has good crystalline quality with sufficient compressive strain.

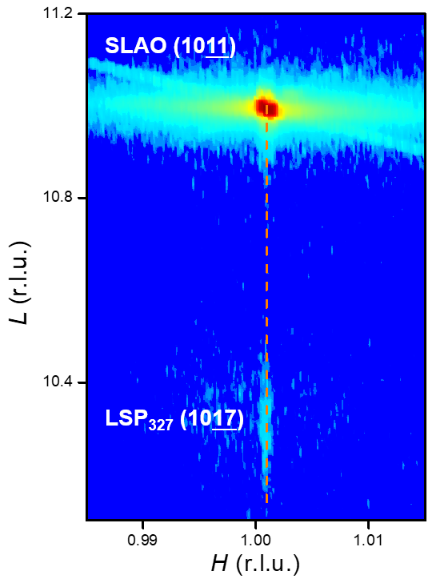

Fig. R7: Reciprocal space mapping around the $SrLaAlO_4$ (1011) and LSP_{327}(1017) reflections. The film is coherently strained to the substrate.

3. Hall coefficient interpretation: The positive Hall coefficients observed in this work contrast with recent literature reports showing negative Hall coefficients in Sr-doped bilayer nickelate films. Given that this heterostructure contains both Sr and Pr, could the authors provide a more detailed discussion of the charge carrier behavior and how it relates to the superconducting mechanism in their specific system?

Response: We thank this reviewer for the careful review. The Hall coefficient (R_H) in bilayer nickelate thin films indeed shows some kind of contrast in different literatures. In bulk crystals under high pressure (see ref. 13), the Hall coefficient is positive and shows clear enhancement when entering the superconducting state. And in superconducting bilayer thin films, the Hall coefficient of $La_3Ni_2O_7$ (see ref. 36) is positive. However, in $La_{2.85}Pr_{0.15}Ni_2O_7$ (see ref. 37) and $La_{2.91}Sr_{0.09}Ni_2O_7$ (see ref. 39), the Hall coefficient are negative. Recently, the researchers report an anomalous electron-hole crossover of the Hall coefficient in superconducting $La_{3-x}Sr_xNi_2O_{7-\delta}$ thin films with different oxygen content (see new ref. 45 in revised version). They found that the R_H can be simultaneously tuned through Sr doping concentration and oxygen content in films. Therefore, the positive R_H in our bilayer LSP_{327} thin film may be due to the special doping concentration and oxygen contents. From our calculations, presented in Fig.R5 and Supplementary Fig. 13 in the revised manuscript, we find that the β and γ bands give hole - like contribution and the α band around Γ point is responsible for the electron-like contribution, and our theoretical calculation show that the largest effect of the pressure for the Fermi surface and the bandwidth occurs for the γ pocket. Within purely quasiparticle picture, the Hall coefficient is expected to be small and negative at ambient pressure, in agreement with oxygen-rich data of ref.45. The Hall coefficient becomes positive as a function of pressure due to increasing volume of the γ-pocket. Therefore, based on our present Hall coefficient data on our samples, we may conclude that dominant hole-like Hall coefficient at ambient pressure may be a result of certain amount of oxygen deficiency in our samples. In the revised manuscript, we have discussed about the charge carrier behavior more carefully and make some discussions concerning the superconducting mechanism. We hope our responses can remove the concerns of this reviewer.

4. Data presentation: The resistance vs. temperature data should be converted to resistivity vs. temperature for proper materials characterization and cross-comparison with others' work. Additionally, experimental uncertainties should be included, particularly for the transition temperatures in the phase diagram.

Response: Thanks for the contractive suggestions. In the revised version, we have converted the resistance data *R(T)* into resistivity *ρ(T)* in Fig.2, Fig.3 and Supplementary Fig. 3, Fig. 5a, Fig. 7. The error bars of T_cs based on the uncertainty of the measured temperature are also included in the phase diagram of Fig. 4.

5. Mechanisms: The manuscript describes two different trends in superconducting transition behaviour under pressure but does not clearly explain their origins. Could the authors elaborate on what physical mechanisms drive these different behaviours and how they relate to the theoretical calculations?

Response: We thank this reviewer for this comment. Our theoretical calculations show a synergic effect of multiband effect in the system by applying a high pressure. There are two main effects that enhance Cooper pairing in the thin films under pressure. First, the reduced *c*-axis lattice constant leads to a stronger presence of the γ-pocket at the Fermi level and its participation in Cooper pairing with an $s^{\pm}$-wave order parameter. Second, pressure increases the hybridization between the $3d_{z^2}$ and $3d_{x^2-y^2}$ orbitals, resulting in greater quasiparticle band coherence. Both effects contribute to the enhancement of T_c. However, the even spin fluctuations are also significantly sensitive to the changes of *c*-axis lattice constant. Especially, a dome like behavior is seen for the q_1^e and q_3^e peaks, which is consistent with the experimental observation.

Therefore, it is likely a synergic effect of increased hybridization between $3d_{z^2}$ and $3d_{x^2-y^2}$ orbitals, resulting in the charge flow between the orbitals and larger itinerancy of the pressurized thin films, together with the enhanced antiferromagnetic spin fluctuations both in the in-plane and out-of-plane channels. Moreover, these effects quickly saturate upon larger pressures explaining why no further increase of T_c and even its reduction is observed.

6. Reproducibility: While multiple samples are measured, it would be valuable to include a more quantitative discussion of sample-to-sample variation in superconducting properties at ambient pressure, particularly given the complexity of the heterostructure growth and post-annealing process.

Response: We thank this reviewer for this pertinent comment. Actually, all the thin film samples in our present study were grown and post-annealed with a similar process, however, the onset of superconducting transition temperature shows some kind of sample dependence. In the revised version, we have added a new Supplementary Note 1 to talk about the details of the synthesis process and post-annealing parameters of the LSP_{327} thin film and give a table (see below) to summarize the post annealing parameters and the transport properties of the six films we used in present work. Based on the limited data in the table, it seems that more annealing time are important for the good metallic behavior and higher T_c.

Samples	Ozone pressure (Pa)	Flow rate (sccm)	Annealing time (h)	T_c (K)	ρ_{200K}	ρ_{200K}/ρ_{50K}
S1	20	10	1.5	38.7	0.163	1.65
S2	20	10	0.5	31.6	0.259	1.57
S3	25	20	1.5	38.8	0.217	1.56
S4	25	20	1	41.4	0.248	1.68
S5	25	20	2	41.9	0.237	1.86
S6	20	10	1	38.1	0.285	1.46

Despite these questions, this work represents a significant contribution to understanding pressure effects in nickelate superconductors and successfully demonstrates substantial enhancement of transition temperatures through the combination of strain engineering and pressure tuning.

Response: We would like to thank this reviewer for the careful review and kind appreciation to our work. The constructive suggestions and comments raised by the reviewer are very helpful for us to improve our manuscript. With the point-to-point responses and related revisions, we hope this reviewer will be satisfied with the revised version.

Responses to the Report of Reviewer #4

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Response: We thank this reviewer for the input in reviewing our work. We hope the comments and suggestions raised by this reviewer have been properly addressed and answered in our responses to the former three reviewers.

We sincerely thank the editor and all four reviewers for their valuable feedback that we have used to improve the quality of our manuscript.

Responses to the Reviewers' report
(*The words in black color are from the Reviewers´ reports, and the blue words give our responses.*)

Responses to the Report of Reviewer #1

I appreciate the authors for revisions. However, there are still critical points that need to be revised. Please reconsider following comments.

Response: We thank this reviewer for the valuable comments/suggestions. We have carefully considered the issues pointed out by this reviewer and revised the related parts. The definition of the transition temperature has been reconsidered. The T_c^{cross} shown in the last version has been defined as T_c^{onset}, this definition has been unified through the whole paper for a fair comparison between our work and the previous published literatures. We hope the reviewers are satisfied with the relevant revisions.

1. As for comment 1, the response and revisions are not insufficient. The definition of the onset superconducting transition temperature should be reconsidered. It is still not fair to define the onset critical temperature as defined in the revised manuscript. Tc^cross shown in Fig. 2b should be defined as Tc^onset for a fair comparison. In addition, the definition of Tc^cross in supplementary Fig. 8 seems appropriate for applying the definition of Tc^onset as similar as previous studies. The data in Fig. 3b, however, can be interpreted very differently depending on the definition used. When the same definition as other papers is applied, the transition temperature shows almost no increase. Furthermore, the definition of Tc^cross in Fig. 3b is clearly not valid. The authors should conduct a more fair analysis of the data. There is concern that changing the method of definition could alter the conclusion. For a fair and scientific discussion, it is important to use a comparable definition to that used in previously reported literature, such as ref. 13 and 40. The authors must revise the definition of the onset temperature.

Response: We thank this reviewer for pointing out this important issue. We agree with the reviewer that the definition of the onset superconducting transition temperature should be reconsidered. In the revised manuscript, we have redefined the T_c^{onset} as the cross-point of the linear extrapolation of normal state and the transition region with resistivity of 80%-90%ρ_n (see Figs. 2, 3, and Supplementary Fig. 8 in the revised version). The transition temperatures in phase-diagram (Fig. 4) and the related statement have also been corrected. This indeed gives a solid base for a more fair comparison between our results and those in previously reported literatures. We hope this reviewer agrees with our new definition in the updated version.

2. The response to the comments 2 and 4, the comments of [Therefore, we may attribute the increase of normal state resistance to the damage to the electric connection of the film caused by the solidification of the pressure-transmitting medium.] and [Some of the conduction paths of the thin films may be partially destroyed due to change of the pressure environment, such as the solidification of the PTM, producing considerable amount of dislocations and defects on the films.] are provided. These comments appear to be an arbitrary interpretation that changes in the pressure medium cause abnormalities in resistance but have no intrinsic effect on the transition temperature. The basis for drawing this conclusion has not been sufficiently explained. If the dislocation is induced in the film by applying pressure, the pressure may not induce the lattice compression because the pressure may be relaxed by inducing dislocation. Under this situation, what does understanding the pressure dependence of the transition temperature lead to? Lattice compression may not be increased as pressure increases above 8 GPa. In Fig. 4, the decrease above the critical temperature of 8 GPa may be attributed to an extrinsic origin unrelated to the increase in pressure.

To explain the pressure dependence of resistivity for general readers, Figure R1 should be added in supplementary information.

If the authors claim this sentence [We may argue that the weak insulating behavior above Tc at high pressures is an intrinsic property of the bilayer nickelate thin films.] in line 281, the results should be compared to the bulk studies of bilayer nickelates although [Such behavior has been reported in the underdoped cuprates48, 49.] is commented.

Response: We thank this reviewer for these comments. As we responded in last run of review, as well as in the illustration of Figure R1 and Supplementary Note 2, for our samples with different PTM (S3 and S4) and different samples with the same PTM (our samples and 112 thin films in ref. 46), the abnormal change of resistivity magnitude always occurs and the critical pressure for this phenomenon seems to be quite random and there is no correlation between the change in Tc and absolute value of normal state resistance in thin films for the high-pressure measurement in the CAC apparatus. Therefore, we believe that the abnormal increase of resistance in our thin films under pressure are most likely due to the extrinsic reasons, for example the creation of some disorders or defects. Accordingly, we have added the Figure R1 to the supplementary information as Supplementary Fig. 9 in the revised manuscript.

These disorders or defects may modify the current flow in some regions, produce new scattering centers, thus the general magnitude of resistivity will be strongly influenced, but the transition temperature itself may not change too much, because it is determined by the onset transition temperature. Thus, concerning the random values of the resistivity under high pressures, here we mean the creation of disorders or defects, instead of the dislocations. We agree with the reviewer that the dislocations induced in the film may release some lattice compression. Here, we further address this situation by the looking at the analog in 112 thin films under high pressures (ref. 46). The authors there found that the infinite-layer structure is partially destroyed, producing considerable amount of disorders or defects in a large scale after the high-pressure measurements. However, the T_c is monotonically increased to above 30 K. For another example, in Sr-doped $La_3Ni_2O_7$ thin films, the weak insulating behavior can also be observed although the quality of the film is very high, which means that the weak insulating behavior may be intrinsic in some doping or pressure region. Therefore, we think that the disorders or defects of the film under high pressures may also induce the broadening of the superconducting transition, but the evolution of T_c and weak insulating behavior above T_c can be interpreted as intrinsic pressure effect of the thin films.

As for the sentence in line 281, we agree that it is important to compare this behavior with other bilayer structure, such as bulk crystals and thin films. However, as we responded in the comment 4 in the last run of review, we still want to emphasis that the electronic structures are quite different in the bulk samples and thin films. Although such insulating behavior is not observed in bulk samples under pressure, we want to note that, in nickelate superconducting thin films, the weak insulating behavior was also observed in Sr-doped $La_3Ni_2O_7$ with dome-like superconducting region [see ref. 39 and 45].

We have added above information to the revised manuscript and we hope this reviewer is satisfied with our responses and related reversions to the manuscript.

3. As for comment 12, Applying pressure causes the thin film and substrate to compress isotropically, so it is reasonable to assume that the a-axis length of film is also compressed. The proposed calculation model seems not valid. Failure to explain the lattice deformation in the computational model to readers may lead to misunderstanding. It should be explicitly stated that the calculation assumes a fixed a-axis length and affection of this fixing.

Response: We thank this reviewer for this pertinent comment. In the manuscript, we have clearly stated that the in-plane lattice constant was fixed during the theoretical calculation in the main text (Line 319-325, 388-390, 415-418, 427-429, 506-511 in version R1). We agree with the reviewer that a more detailed justification for the assumption of a fixed in-plane lattice parameter would strengthen the connection between the theoretical model and experimental conditions as commented by the reviewer #2 in the last run of review.

In the compressively strained thin films, although the thin film and substrate will be compressed simultaneously under pressure, the case is that the external pressure acts on a lattice already clamped by the substrate (especially there are only few unit cells in thin films), so the in-plane lattice constants and the in-plane Ni-O-Ni bond angle remain largely fixed [see for example Fig.1 in B. Samanta, and A.B. Georgescu, arXiv: 2506.11427 (2025)]. In the compressively strained thin films, the c-axis lattice constant has already elongated for 1-2%, thus we assume that the compression is mainly influenced the c-axis lattice constant. In addition, along the c-axis, there is a bilayer block with NiO planes and two rock-salt layers of LaO, therefore it is much more susceptible to the change with pressure. It is anticipated that the in-plane compressing rate is smaller concerning a "more homogeneous" a-axis with the stiff transition metal-O bonds. Therefore, we assume that the dominant effect from pressure compared to the ambient state would be a more stronger shrinkage of c-axis lattice constant.

In the revised manuscript, we have added a new Supplementary Note 3 in the revised manuscript to explain the lattice deformation in the computational model to readers and given more explanations on the fixed in-plane lattice constant during the theoretical calculations.

4. Fig. R2 should be included in supplementary information. If the authors claim the stability of superconductivity in air, in addition to the description about the stability in air, the fragility by applying pressure should be also described in main text.

Response: We thank this reviewer for this comment. In the revised version, we have added the Fig. R2 to Supplementary information as a new Supplementary Fig. 2. We also add some necessary descriptions on the fragility of superconductivity in bilayer thin films without this kind of heterostructure in the revised main text. This reads as: “Without this kind of heterostructure, the films are pretty easy to deteriorate in air, showing fragility of superconductivity, and it is hard to do the subsequent transport measurements especially under high pressures”. We hope our responses can remove the concerns of this reviewer.

5. The claims in abstract seem not supported by results. The descriptions should be revised fair to the results.

The claim of [Here we report the enhancement of Tc onset from about 40 K to over 60 K] in abstract should be revised. By applying appropriate definition for fair comparison, judging from the data in Fig. 2b, the temperature seems roughly 30 K to over 40 K. In the abstract, the sentence [superconductivity with an onset transition temperature (Tconset) over 40 K was achieved in La3Ni2O7 and (La,Pr)3Ni2O7 thin films at ambient pressure due to in-plane compressive strain.] is also described. However, 40 K in the literature was estimated by an appropriate definition, but the author’s definition is not appropriate. In the same abstract, it is a critical problem for comparison the onset temperature under different definitions. Such description misleads the general readers. The authors should revise their method of definition. The sentence at line 29 of the abstract in this manuscript should be revised to [Here we report the enhancement of Tconset from about 30 K to over 40 K].

The claim of [reaching the maximum Tc onset at about 61.5 K.] in abstract should be revised.

By applying appropriate definition for fair comparison, the maximum Tc onset in Fig. 3b seems roughly 50 K.

It would be better to revise the claim of [Our theoretical calculations show that the increase of Tc may be mostly described by the reduction of the c-axis lattice constant, which yields a cooperative enhancement of magnetic fluctuations within and between the layers and increased metallicity under pressure.], because he conclusion that the c-axis length is the cause is not logically valid in calculations where only the c-axis length is varied. Despite the theoretical calculation under hydrostatic pressure, the authors did not calculate any parameters other than the c-axis length in the first place.

Correspondingly to the revisions in abstract, the main text should be appropriately revised.

Response: According to the suggestions proposed by this reviewer, we have revised the abstract and the related main text carefully in the updated version, now the statements in the revised versions (including abstract and main body) are well supported by the results.

6. in line 138, [a relativity large size (3 mm x 1.5 mm) as shown in the inset of Fig. 1c] is added. In the inset, the scale bar is necessary to show the sample size.

Response: The scale bar of the sample size is added.

7. in line 139, [the applied current is supposed to be uniformly distributed on the thin films.] is added in the revision. How did the authors judge uniformity of the current? Since it is rather difficult in general to characterize the current uniformity in sample, when providing such speculation, it is necessary to explain the experimental basis.

Response: We thank the reviewer for this comment. As shown in the Figure R1 below, we use a typical five-probe electrode configuration to measure the R_{xx} and R_{xy} simultaneously. Since the sample size is relatively large with a rectangular shape, and the electrodes for applying the current is parallelly made in the two terminals, therefore, the current is supposed to be uniformly applied on the thin films. We agree with the reviewer that it is difficult to characterize the current uniformity in sample, especially under pressures. In the revised manuscript, we have replaced the image in the inset of Fig. 1c with Fig. R1 and changed the statement, and explained the experimental details for the general reader.

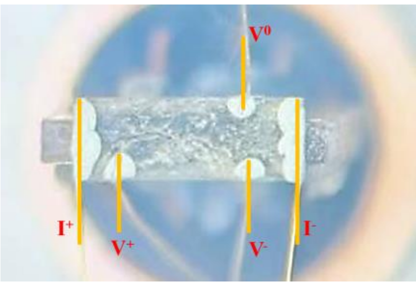

Fig. R1: The standard five-probe electrode configuration of the thin film sample before putting it into the high-pressure cell of PCC. The electrodes for applying the current are put on the two terminals with a uniform distribution covering the entire sample.

8. in line 364-370, the claim of the consistency in the sentence of [This is consistent with the Hall coefficient data shown in Fig. 2d, indicating stronger metallicity of the normal state upon applied pressure.] appropriate? The detection of insulating behavior in experimental results seems more important. In this point, the inconsistency seems more important to be noted in the discussion part.

Response: We thank the reviewer for raising this issue. Under pressure, the enhanced itinerancy of electrons can lead to the increase of the metallicity of the film, and this phenomenon is consistent with the Hall coefficient data in the low-pressure region (< 2 GPa). However, Hall coefficient measurements under higher pressure in CAC is challenging. According to the reviewer`s comment, we realize that the observation of weak insulating state under higher pressures is indeed more important. In the revised manuscript, we have modified the related discussions regarding to the weak insulating behavior based on the theoretical calculations. We hope this reviewer will agree with our related revisions and changes to the manuscript.

9. in line 431-438, the extrinsic origin with inducing dislocation etc has to be also discussed.

Response: Thanks for the comment. As we addressed above, we think the high pressure may induce some disorders or defects, not dislocations. The influence on the magnitude of global resistivity is understandable. It may give weak influence on the onset transition temperature and the weak insulating bahevior, as we argued in comment 2 above. We don`t think the pressure induced disorders or defects on the films will cause the weak insulating behavior in our present work. The reasons are as follow: (1) A large amount of disorders or defects in a large scale after the high-pressure measurements was also observed in 112 thin films under pressure, however, there is no weak insulating behavior observed. (2) The weak insulating behavior is also observed in over-doped region of Sr-doped $La_3Ni_2O_7$ at ambient pressure although the quality of the thin films is pretty high. Therefore, the weak insulating behavior observed in our present work should originate from the intrinsic pressure effect of the sample itself, for example the gradually enhanced correlation effect which provides the tendency for the formation of a density wave order, probably associated with the shift of the γ-pocket with a flat band feature.

We have included some discussion concerning this issue in this paragraph. We hope this reviewer will be satisfied with these revisions.

We hope this reviewer is satisfied with our point-to-point responses and related revisions, and will make a positive recommendation to our work for publication in Nature Communications.

Responses to the Report of Reviewer #2

The authors have revised the manuscript significantly and its quality is improved. Their replies to all reviewers' questions are satisfactory and I recommend its publication in Nature Communication.

Response: We are grateful for the recognition and kind recommendation to publish our work in Nature Communications. The insightful feedback is highly valued and much appreciated.

Responses to the Report of Reviewer #3

The authors have adequately addressed all my concerns and made a lot of efforts to improve the quality of the data. I am satisfied with the revisions and recommend this manuscript for publication in Nature Communications.

Response: Many thanks for the satisfactory assessment and supportive recommendation! We deeply appreciate the professional guidance that strongly enhanced the quality of our manuscript.

Responses to the Report of Reviewer #4

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Response: We sincerely thank this reviewer for co-reviewing the manuscript. His/her involvement is greatly appreciated, and these insights contribute significantly to the review process. We hope the comments and suggestions raised by this reviewer have been properly addressed and answered in this run of review.