Peer Review File

Superconductivity in an infinite-layer nickelate superlattice

Corresponding Author: Professor Zhaoliang Liao

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
In the manuscript by Xiao et al., the authors report on superconductivity in infinite-layer nickelate superlattices, showing the thickness-dependent structural evolution and a critical thickness for superconductivity. Specifically, after a complete reduction process, the thinner samples exhibit partially reduced and disordered phases, while the thicker samples display a pure infinite-layer structure. The authors also perform X-ray linear dichroism and magnetoelectric transport measurements, with results mostly consistent with what is expected for superconducting infinite-layer nickelate films. While the study observes superconductivity in superlattice samples and documents structural evolution as a function of thickness, it appears more technical in nature and does not contribute significant new insights into the underlying physics of the field. The presence of apical oxygen near the interface, which has already been reported, naturally explains the absence of superconductivity in thinner films. Additionally, the XAS and transport results largely mirror those of other infinite-layer nickelates, offering little in terms of novel physics. Given these considerations, I believe the manuscript would be more appropriately suited for publication in a more specialized journal.

Reviewer #2

(Remarks to the Author)
The manuscript by W. Xiao et al. presents an intriguing study on superconducting nickelate superlattices. The authors demonstrate a well-controlled fabrication of superlattice structures using pulsed laser deposition (PLD) and employ a CaH_2 reduction process to selectively remove apical oxygens. The in-depth structural analysis using iDPC-STEM and X-ray techniques effectively shows the successful realization of nanoscale superlattices with targeted reduction in the nickelate layers. Given that the topotactic reduction in multi-layer structures remains a largely unexplored area, the study of superlattice is critical for advancing the field of nickelate research. While the results represent a meaningful step forward in the study of nickelate thin films, there are several issues related to the results and analysis. Addressing the following points will be necessary to ensure that the manuscript is a suitable candidate for publication in Nature Communications.

1. The authors report a Tc of approximately 12.5 K in the R-N8/S2 structure. How does this compare to their single-layer thin films of R-N8 on $SrTiO_3$? Could the authors provide comments on the difference in Tc between the superlattices and the single-layer films?

2. In Fig. 4f, the superconducting anisotropy, defined by the ratio of Hc2||ab and Hc2||c, appears to be close to one, suggesting a more three-dimensional behavior. Could the authors provide comments on both the superconducting anisotropy and the BKT-like behavior observed in the I-V curves?

3. In Fig. 5c, the authors perform I-V measurements to demonstrate possible BKT transitions in these samples. They show that the superlattice (SL) sample exhibits a significant increase in the exponent and claim that the SL sample is more 2D in nature. However, this argument lacks sufficient evidence. Additionally, the T_BKT is determined to be approximately 0.7 Tc (T_BKT ~ 5.7 K), which is quite low for what would typically be expected in a 2D superconductor, where T_BKT is generally much closer to Tc.

4. Do the authors compare these results with a thicker nickelate version of the SL structure to determine whether the observed behaviors stem from the superlattice architecture or are an intrinsic feature of infinite-layer nickelates?

Reviewer #3

(Remarks to the Author)
In the present work by W. Xiao et al. “Superconductivity in the infinite-layer nickelate superlattice” using soft-chemistry topotactic reduction the authors successfully synthesized nickelate heterostructures (RNiO2+x)n/(SrTiO3)2 with different thickness of the reduced nickelate layer, n. It was shown that above a critical thickness n>5 u.c. the high-quality nickelate heterostructures with infinite-layer crystal structure RNiO2/SrTiO3 can be stabilized. The authors perform a detailed analysis of the electronic state and crystal structure properties of these materials using different experimental techniques, such as x-ray diffraction, x-ray reflectivity, x-ray absorption, and x-ray linear dichroism measurements, in combination with the atomically resolved scanning transmission electron microscopy and resistivity measurements, etc. It was discovered that the nickelate heterostructures with n=8 u.c. display superconductivity with a critical temperature Tc~12 K, in close agreement to that in the hole-doped nickelate thin films. In contrast, the nickelate heterostructures with n<5 u.c. show insulating behavior which is presumably associated with a structural disorder in the reduced nickelates. To the best of my knowledge this is the first report on superconductivity in nickelate heterostructures with the infinite-layer crystal structure.

I read the manuscript with great interest. The results presented in the manuscript are interesting and scientifically sound. The paper is well organized and clearly written. In my opinion, the subject matter of the manuscript is suitable for the publication in the Nature Communications journal. The results discussed in the manuscript are at the high level of present experimental capabilities. I propose that this novel approach to study superconductivity in nickelates compounds will be highly appreciated in the future experimental and theoretical research. In fact, it opens a novel direction for experimental and theoretical research of the microscopic origins of superconductivity in nickelates. In addition, it allows one to use various effects such as quantum confinement and epitaxial strain to control the properties of investigated systems.

I believe that the manuscript meets the criteria for publication in Nature Communications and can be accepted after the authors address the following minor corrections:

1) It was shown that the infinite-layer nickelate heterostructures with n=8 u.c. display superconductivity with a critical temperature Tc~12 K. Is this correct to say that for n>=8 u.c. the infinite-layer nickelate heterostructures show superconductivity? I mean the authors do not discuss (may be I missed this) what happens for n>8. n=8, is this the only point at which this systems superconducts or it does for n>=8 u.c. Is there experimental evidence (support) that, e.g., for n=9 and 10 the reduced nickelate heterostructures are also superconducting? Please clarify this point. It might be great to present extra experimental data for n>8.

2) The authors present a detailed analysis of the electronic structure and structure properties of the synthesized nickelate heterostructures. At the same time, compositions of these materials are discussed very briefly. It was claimed that it is at the optimally hole-doped state with Nd/Sr=0.8/0.2. How the authors determine a composition of the reduced nickelate heterostructures (Nd/Sr=0.8/0.2)? I mean it can be affected upon topotactic reduction of the well-characterized perovskite precursor phase with Nd/Sr=0.8/0.2. I believe that these materials can also be highly inhomogeneous.

3) The authors do not discuss the magnetic properties of these materials. Is there any experimental evidence of a long-range ordering (spin or charge density wave behavior) in these materials? I understand that it is too much to ask the authors to perform such measurements. A brief discussion might be sufficient (if possible).

4) It turns out to me that resistivity data for the R-N8/S2 system show a weak anomaly at about 140 K. It can be related to a charge or spin density wave formation in the reduced nickelate heterostructures, i.e., a long-range ordering stabilized in the infinite-layer nickelates due to quantum confinement. It seems that a similar behavior also appears in the resistivity data of the Ruddlesden-Popper bilayer and trilayer bulk nickelates. It could be interesting to add a brief discussion on this (if possible).

5) In the paper and in the supplemental material the authors discuss somewhat different compositions of the nickelate heterostructures. In the paper these are N_x/S2 and R-N_xS2, while in the SM are N_x/S3 and R-N_x/S3. It looks like that no additional data presented on the N_x/S2 and R-N_xS2 in the SM. It also might be great to make a conclusion how the thickness of the SrTiO3 layer affects the properties of these materials (S2 vs. S3).

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

Reviewer #2

(Remarks to the Author)
W. Xiao et al. have presented additional results that reasonably address the concerns I had in my previous review. I believe the content is significant and beneficial for readers, since the study of superlattices is critical for advancing the field of nickelate research. Therefore, I recommend it for publication in Nature Communications.

Reviewer #3

(Remarks to the Author)
I read the revised manuscript and the authors' correspondence with the reviewers. In my opinion, the authors satisfactorily addressed most of the points and questions raised before. I found the results presented in the manuscript to be interesting, the subject matter of the manuscript is suitable for the publication in Nat. Commun. I believe that this paper opens a promising direction in experimental studies of unconventional superconductivity in nickelates. In my opinion, it is worth to accept the manuscript in its present form for publication in Nature Communications.

Response to the Reviewers' Comments

On behalf of all the contributing authors, I would like to express our sincere appreciations of your letter and reviewers' constructive comments concerning our manuscript. These comments are all valuable and helpful for improving our manuscript. Below, we respond to all reviewer's comments and questions point by point and highlight the corresponding changes in the revised manuscript and Support Information.

For Reviewer #1 (Remarks to the Author):

In the manuscript by Xiao et al., the authors report on superconductivity in infinite-layer nickelate superlattices, showing the thickness-dependent structural evolution and a critical thickness for superconductivity. Specifically, after a complete reduction process, the thinner samples exhibit partially reduced and disordered phases, while the thicker samples display a pure infinite-layer structure. The authors also perform X-ray linear dichroism and magnetoelectric transport measurements, with results mostly consistent with what is expected for superconducting infinite-layer nickelate films. While the study observes superconductivity in superlattice samples and documents structural evolution as a function of thickness, it appears more technical in nature and does not contribute significant new insights into the underlying physics of the field. The presence of apical oxygen near the interface, which has already been reported, naturally explains the absence of superconductivity in thinner films. Additionally, the XAS and transport results largely mirror those of other infinite-layer nickelates, offering little in terms of novel physics. Given these considerations, I believe the manuscript would be more appropriately suited for publication in a more specialized journal.

Response: We sincerely appreciate your comments, and would like to emphasize that our work meaningfully advances research in the field of infinite-layer nickelate superconducting films. The challenges associated with the preparation of nickelate films have historically limited the production to single-layer superconducting nickelate films characterized by simple structures and finite thicknesses. Consequently, the understanding of the underlying physical mechanisms limited, leaving numerous unanswered questions about infinite-layer nickelate superconductivity unanswered. For instance, we seek clarity on whether these materials exhibit bulk/intrinsic superconductivity and how interfacial effects influence their superconducting properties.

Our experimental results demonstrate the successful synthesis of superconducting films within superlattices of complex composition, thereby broadening the material landscape for investigating the physics of nickelate films. In addition, using the interlayer $SrTiO_3$ confinement, we achieved high quality superconducting $Nd_{0.8}Sr_{0.2}NiO_2$ layers at significantly reduced thicknesses (~2.6 nm). This opens avenues for further exploration of the physical properties of these unconventional superconducting materials at the quantum scale. For example, by introducing a magnetic layer between this superlattice interface, the competition between superconductivity and magnetism in infinite-layer nickelate films can be further understood. Alternatively, this platform may also facilitate the development of superconducting Josephson junctions based on infinite-layer nickelates.

While our results with the constructed superlattice fundamentally align with findings from other infinite-layer nickelates, it is vital to highlight that these results were achieved at extremely thin thicknesses. Moreover, the observation that the $SrTiO_3$ interface did not enhance the superconducting transition temperature (T_c) contrasts with trends seen in $FeSe/SrTiO_3$ systems, indicating that the superconducting characteristics of nickelate films are intrinsic and relatively invariant to size or interfacial effects. Therefore, we believe that our work provides a valuable new perspective on the intrinsic superconductivity of infinite-layer nickelates, contributing to a deeper understanding of this fascinating class of materials.

For Reviewer #2 (Remarks to the Author):

The manuscript by W. Xiao et al. presents an intriguing study on superconducting nickelate superlattices. The authors demonstrate a well-controlled fabrication of superlattice structures using pulsed laser deposition (PLD) and employ a CaH_2 reduction process to selectively remove apical oxygens. The in-depth structural analysis using iDPC-STEM and X-ray techniques effectively shows the successful realization of nanoscale superlattices with targeted reduction in the nickelate layers. Given that the topotactic reduction in multi-layer structures remains a largely unexplored area, the study of superlattice is critical for advancing the field of nickelate research. While the results represent a meaningful step forward in the study of nickelate thin films, there are several issues related to the results and analysis. Addressing the following points will be necessary to ensure that the manuscript is a suitable candidate for publication in Nature Communications.

Response: We sincerely appreciate your recognition of our work and valuable suggestions. In response to these issues, we have added relevant experiments and analyses, and revised some of the descriptions in the article. Below are our relevant responses.

1. The authors report a T_c of approximately 12.5 K in the R-N_8/S_2 structure. How does this compare to their single-layer thin films of R-N_8 on $SrTiO_3$? Could the authors provide comments on the difference in T_c between the superlattices and the single-layer films?

Response: Thank you for your insightful question. Previous studies on single $Nd_{0.8}Sr_{0.2}NiO_2$ films of different thicknesses grown on $SrTiO_3$ substrates have shown that the T_c value decreases monotonically with reduced thickness [***Nature Communications*** **13, 743 (2022)**]. Extrapolating from these results, we estimate that the T_c value of the R-N_8 single film would be below 6 K, which is significantly lower than the value we observed in R-N_8/S_2.

We believe that the intercalation of $SrTiO_3$ in the superlattice plays a crucial role in stabilizing the structure of the $Nd_{0.8}Sr_{0.2}NiO_2$ layer, thereby minimizing the occurrence of RP defects during the growth process. This structural enhancement enables the superlattice to achieve a higher T_c value compared to single-layer films of equivalent thickness.

Revision:

We have added above discussion on **Page 4** and **Page 5** of the manuscript. Here are the sentences we changed or added,

On **Page 4**, "This transition temperature aligns with previous observations in 10 nm $Nd_{0.8}Sr_{0.2}NiO_2/SrTiO_3$ single films. It is noteworthy that our superlattice allows a thinner thickness of the nickelate while still maintaining a high T_c value, whereas for thinner single films, the T_c value decreases monotonically with thickness (for a single $Nd_{0.8}Sr_{0.2}NiO_2/SrTiO_3$ film of 4.6 nm, i.e., ~14 uc, the $T_{c,onset}$ is only ~6.5 K)"

On **Page 5**, "In both R-N_3S_2 and R-N_8/S_2, only very few RP defects were observed, demonstrating the stabilizing effect of $SrTiO_3$ interfacial layer on nickelate monostructures, which may be the reason that our superlattices have higher T_c values compared to single films of similar thicknesses."

2. In Fig. 4f, the superconducting anisotropy, defined by the ratio of $H_{c2}||ab$ and $H_{c2}||c$, appears to be close to one, suggesting a more three-dimensional behavior. Could the authors provide comments on both the superconducting anisotropy and the BKT-like behavior observed in the I-V curves?

Response: We thank you for your constructive comments. In the case of unconventional $Re_{1-x}Sr_xNiO_2$ nickelate system, which has been studied extensively, the superconducting anisotropy ratios $\gamma\ (\frac{H_{c2}||\mathrm{ab}}{H_{c2}||\mathrm{c}}, T = 0K)$ are relatively low. As reported by B.Y. Wang et al [***Science Advance* 9, eadf6655 (2023)**], the superconducting anisotropy values for La-, Pr- and Nd-nickelates are 1.6, 1.25, and 1.26, respectively, all of which are much lower than those of well-established conventional two-dimensional superconductors, indicating a more three-dimensional-like behavior. Nontheless, the H_{c2}(T) behavior that satisfies the 2D-Ginzburg-Landau model is consistent with that of many 2D superconductors [***Science*** **350, 409-413 (2015).** ***Nature Physics*** **20, 957-963 (2024).** ***Nano Letters*** **17, 6802-6807 (2017)**], especially in the temperature range between 5 K and 8.2 K, where the curvature of the temperature-dependent H_{c2} curves in the two directions is significantly different. This behavior may represent a unique characteristic of nickelate thin-film superconductors. Therefore, it is indeed insufficient to determine the two-dimensional nature of this system solely based on the temperature-dependent Hc_2 curves for different orientations.

We have provided new data to illustrate the two-dimensional superconducting feature of the R-N_8/S_2 sample. The angular dependence of the upper critical field, analyzed using the 2D-Tinkham formula, serves as significant evidence for the presence of 2D superconductivity [***Science*** **350, 409-413 (2015)**]. As demonstrated in the following results, we have measured the angular dependent-H_{c2} of R-N_8/S_2 at 5 K. The curve of $H_{c2}(\theta)$ exhibited a sharp cusp-like peak when θ approach 90° (μ_0H//ab), which can be described by the 2D-Tinkham formula (red solid curve). Although the fitted curves do not perfectly align with the scatterplot, they exhibit similar trends and shape. In contrast, the curve fitted using the 3D anisotropic G-L model (blue curve) can’t describe this cusp-like curve. The same characteristic was also found in $Nd_{0.8}Sr_{0.2}NiO_2/SrTiO_3$ single films [***Nature Communications*** **14, 7155 (2023). arXiv:2301.07606 (2023)**], which indicate a 2D superconducting feature of $Nd_{0.8}Sr_{0.2}NiO_2$. By correlating this data with relevant literature, we confirm our assertion of the 2D superconducting characteristics of R-N_8/S_2.

Regarding the T_{BKT}-related issues, we will clarify and address them in ***Comment*** **3**.

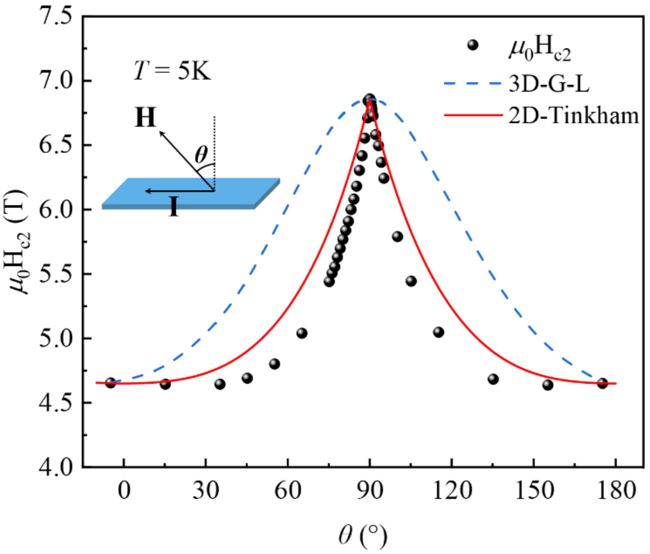


Figure R1. Polar angular dependence of the critical magnetic field $H_{c2}(\theta)$ at $T = 5$ K. The red solid line and the blue dotted line are the fittings with the 2D-Thikham model and the 3D anisotropic G-L model. The inset shows a configuration for measurements.

Revision:

We have changed **Figure 4** by adding the figures related to the above descriptions and put MR into the **Support Information**. Additionally, we have changed and added the following discussion on **Page 7** and **Page 8** at manuscript.

On **Page7**, "This is commonly observed in two-dimensional superconductors. It implies the possible 2D superconducting characteristic of R-N_8/S_2, which is consistent with the square-planar NiO_2 plane geometry. This behaviour can be well described by the 2D Ginzburg-Landau (G-L) formula as

$$H_{c2,\perp}(T) = \frac{\Phi_0}{2\pi\xi_{ab}^2(0)}\left(1 - \frac{T}{T_c}\right) \tag{1}$$

$$H_{c2,\parallel}(T) = \frac{\sqrt{12}\Phi_0}{2\pi\xi_{ab}(0)d_{sc}}\left(1 - \frac{T}{T_c}\right)^{\frac{1}{2}} \tag{2}$$"

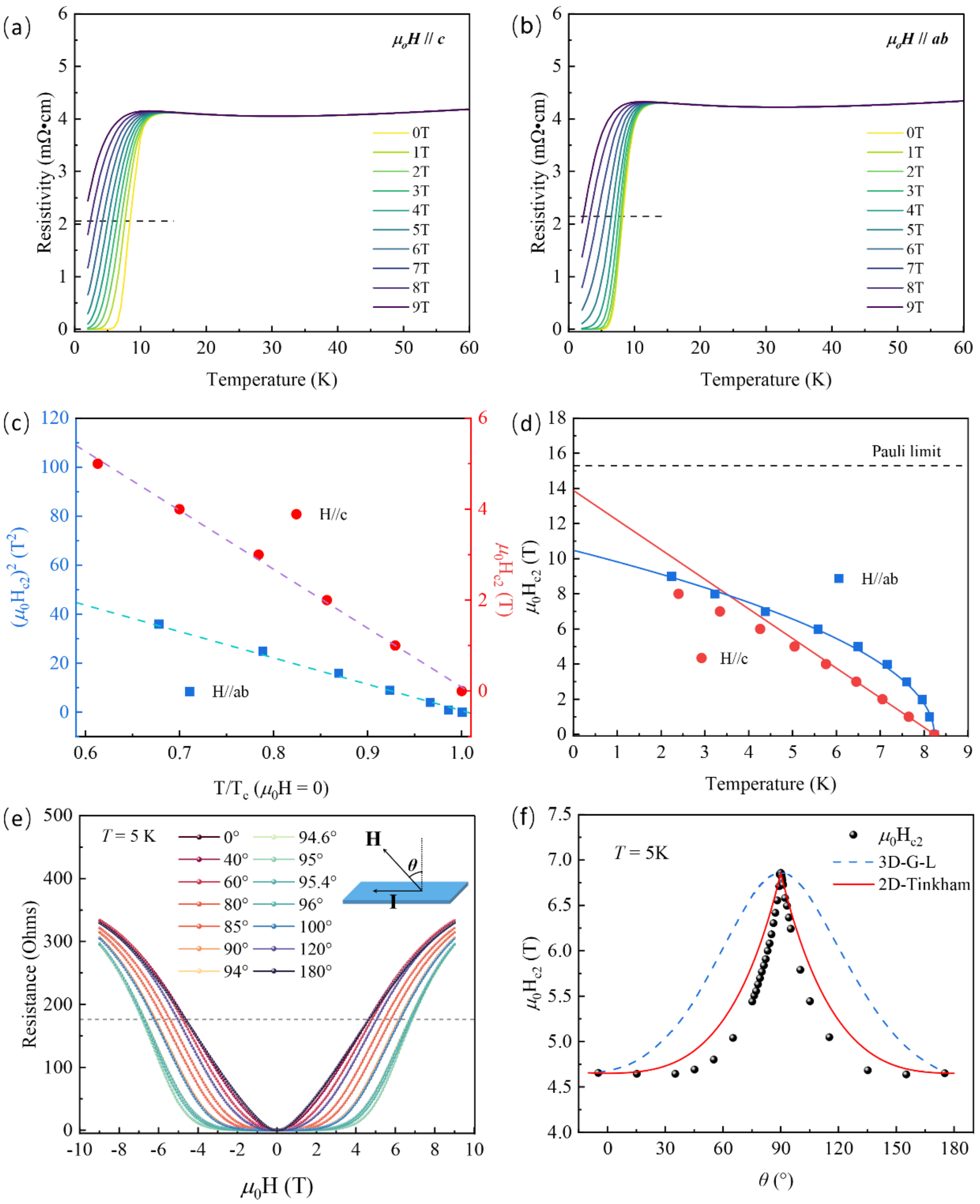
On **Page 7** and **Page 8**, "To confirm the two-dimensional superconducting feature of R-N_8/S_2, we measured the angular dependence of H_{c2} of R-N_8/S_2 at 5 K. Figure 4e shows the MR curves at different θ values, and the inset shows a configuration for measurements. By extracting the values of the critical field at different angles, the angular-dependent $H_{c2}(\theta)$ curves can be obtained, as shown in Figure 4f. A clear cusp-like peak can be observed when θ approach 90° (μ_0H//ab), which can be described by the 2D-Tinkham formula for 2D superconductors (red solid line), which is expressed as

$$\left(\frac{H_{c2}(\theta)sin\theta}{H_{c2}^{\parallel}}\right)^2+\left|\frac{H_{c2}(\theta)cos\theta}{H_{c2}^{\perp}}\right|=1 \qquad (3)$$

In contrast, the peak cannot be reproduced by 3D anisotropic G-L model (blue curve), which is expressed as

$$\mathrm{H}_{c2}(\theta)=\frac{H_{c2}^{\parallel}}{(sin^2\theta+\gamma^2cos^2\theta)^{\frac{1}{2}}} \qquad (4)$$

where anisotropy ratio $\gamma=H_{c2}^{\parallel}/H_{c2}^{\perp}$. These results qualitatively indicate the 2D feature of R-N8/S2. However, unlike the large γ of conventional 2D superconductors, the γ of R-N8/S2 approaches 1 at low temperature. This behavior may represent a unique characteristic of nickelate thin-film superconductors, which was also found in $Nd_{0.8}Sr_{0.2}NiO_2/SrTiO_3$ single films with 2D superconducting feature.”



“Figure 4. Magnetic transport properties and the upper critical fields (H_{c2}) anisotropy of superconducting R-N_8/S_2 superlattice film. Temperature dependence of the resistivity under different magnetic fields from 0 to 9 T **(a)** along *c*-axis and **(b)** in the *a*-*b* plane below 60K. **(c)** Temperature dependence of $(H_c)^2$ ($\mu_0 H$ || *ab*) and H_c ($\mu_0 H$ || *c*) near T_c. The purple and blue dashed lines are the corresponding linear fits. **(d)** Temperature dependence of the upper critical fields for both directions. The solid line is the data fitted around the T_c using the (modified) GL theory, and the dashed lines indicate the Pauli limit of H_{Pauli} = 1.86 T_{c0}. **(e)** Magnetic field dependence of the resistance measured with different magnetic field orientations at 5 K, and the top right inset shows a configuration for measurements. **(f)** Polar angular dependence of the critical magnetic field $H_{c2}(\theta)$ at T = 5 K. The red solid line and the blue dotted line are the fittings with the 2D-Thikham model and the 3D anisotropic G-L model. All the H_{c2} data are extracted at 50% of the normal state resistance or resistivity.**”**

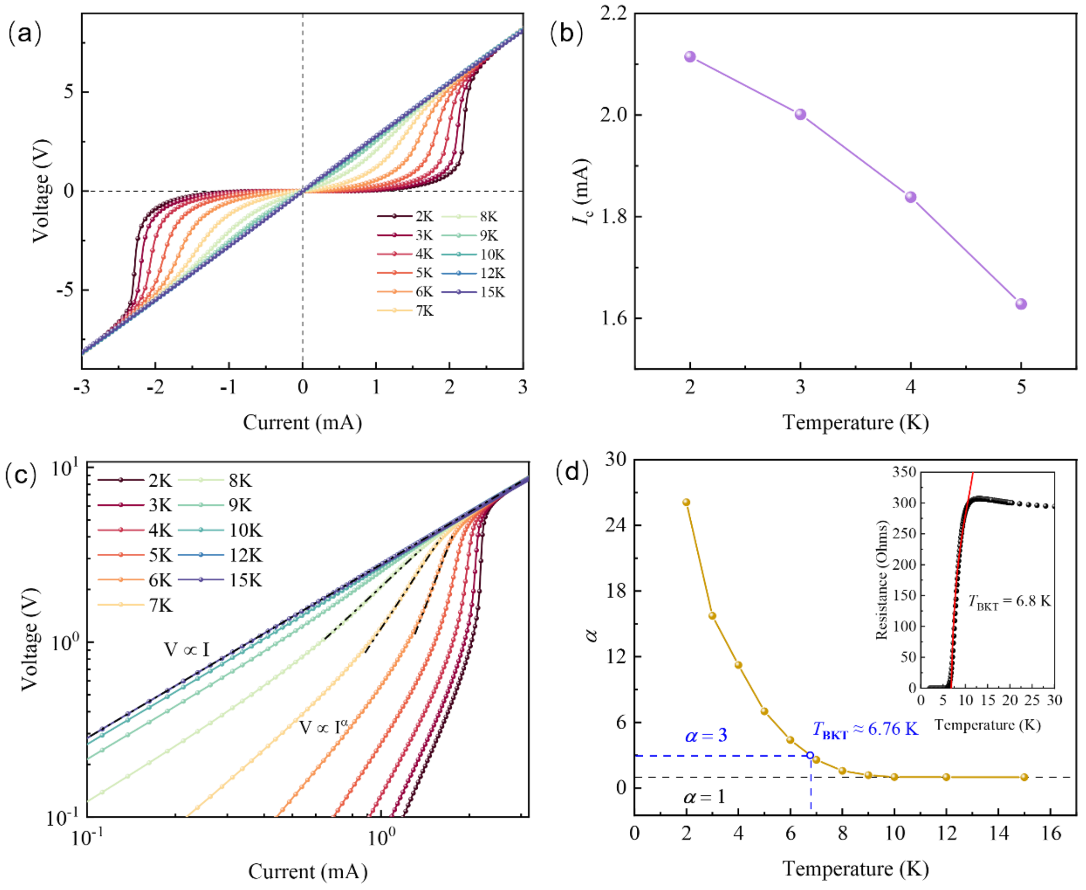
3. In Fig. 5c, the authors perform I-V measurements to demonstrate possible BKT transitions in these samples. They show that the superlattice (SL) sample exhibits a significant increase in the exponent and claim that the SL sample is more 2D in nature. However, this argument lacks sufficient evidence. Additionally, the T_{BKT} is determined to be approximately 0.7 T_c ($T_{\mathrm{BKT}} \sim 5.7$ K), which is quite low for what would typically be expected in a 2D superconductor, where T_{BKT} is generally much closer to T_c.

Response: We appreciate the reviewer's valuable question. In the reply regarding ***Comment 2***, we add a proof of the two-dimensional superconductivity feature of R-N_8/S_2. For the value of T_{BKT}, we are so sorry that we didn't extract the slope near the turning point of the I-V curve [***Science*** **317, 1196-1199 (2007)**], which resulted in a wrong T_{BKT} value in the previous version. In the revised version, we have corrected it and obtained a T_{BKT} of 6.76 K. In addition, we have also fitted the R(T) curve using the Halperin-Nelson equation [***Science*** **350, 409-413 (2015)**] to simulate the BKT temperature value and obtained a T_{BKT} of 6.80 K, which is consistent with the value we obtained from I-V curves. Similar BKT transition temperatures were also observed on $Nd_{0.8}Sr_{0.2}NiO_2$ single films [***Nature Communications*** **14, 7155 (2023).** ***Physical Review Letters*** **133 (2024)**]. Thus, we conclude that the R-N_8/S_2 sample has a 2D superconducting feature with a T_{BKT} of 6.80 K.

Revision:

We have modified **Figure 5** and added an illustration. In addition, the T_{BKT} values were revised and a related discussion of fitting using the Halperin-Nelson equation was added on **Page 8** in the manuscript.

On **Page 8**, "We further extracted the power index values α as a function of temperature from slopes in the log-log scale I-V characteristic curves at different temperatures (Figure 5d), yielding an extrapolated T_{BKT} of ~6.76 K. In addition, the R(T) curve can be reproduced by BKT transition using Halperin-Nelson equation (red solid curve in the inset of Figure 5d), $R = R_0 \exp\left[-2b\left(\frac{T_{c0}-T}{T-T_{BKT}}\right)^{\frac{1}{2}}\right]$, where R_0 and b are material parameters. The fitting results give a BKT transition temperature of $T_{\mathrm{BKT}} = 6.80$ K, which is consistent with the data extrapolated from the I-V curves.”



“Figure 5. Critical current and the BKT transition temperature of R-N$_8$/S$_2$. (a) I-V curves at different temperatures. **(b)** Dependence of critical current with temperature. **(c)** Plots of the I-V curves at different temperatures on a log-scale. The relationship between current and voltage can be represented by the slope of the fitted dotted line. **(d)** Temperature dependence of α in $V \propto I^{\alpha}$. T_{BKT} of R-N$_8$/S$_2$ are indicated by the blue hollow dot. The inset shows the resistance transition at zero magnetic field, and the red solid line represents the BKT transition using the Halperin-Nelson equation.”

4. Do the authors compare these results with a thicker nickelate version of the SL structure to determine whether the observed behaviors stem from the superlattice architecture or are an intrinsic feature of infinite-layer nickelates?

Response: Thanks for this valuable advice. We have prepared a series of superlattices films with different thicknesses and measured their XRD patterns and transport properties. The results are shown below.

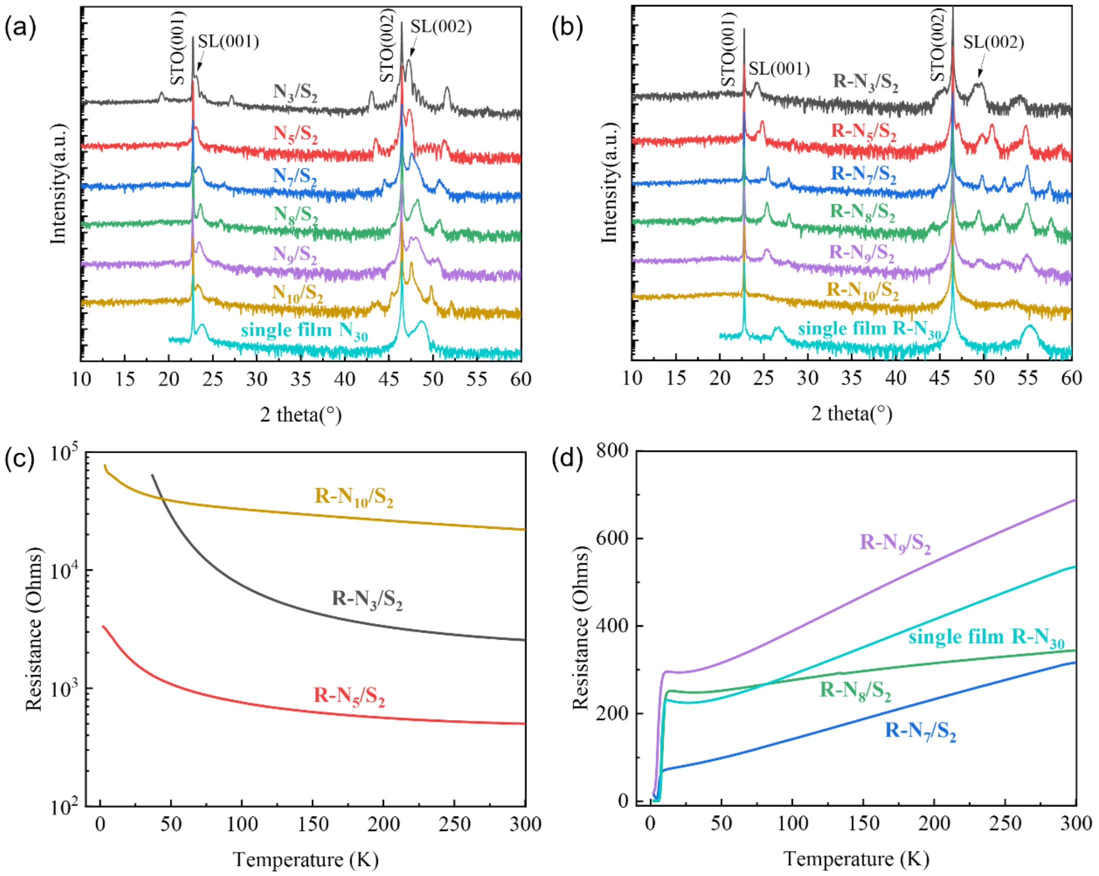


Figure R2. θ-2θ XRD spectra of (a) as-grown (N_n/S_2) and (b) reduced (R-N_n/S_2) superlattices (SLs) with different thickness of nickelates. As $n > 9$, R-N_n/S_2 is hard to obtain a pure infinite-layer structure. Temperature-dependent resistance R(T) of R-Nm/S2 , (c)when $n \leq 5$ and $n > 9$, the R- N_n/S_2 superlattices exhibit insulating behavior, (d) as $5 < n \leq 9$, the R- N_n/S_2 superlattices are superconducting. A single film of 30 u.c. $Nd_{0.8}Sr_{0.2}NiO_{2,3}$ with the same thickness of $SrTiO_3$ capping layers is also included in the **(a)**, **(b)** and **(c)** as comparison data (light blue lines).

Due to the challenges involved in achieving a well-formed infinite-layer phase in thicker superlattice films, superconducting transitions were only observed in the R-N_7/S_2, R-N_8/S_2, and R-N_9/S_2 structures, which exhibited critical temperatures (Tc) in the range of 8 to 12 K. The resistances of R-N_7/S_2 and R-N_9/S_2 could not be completely tuned to zero, possibly as a result of effects encountered during the reduction process [***Nature*** **615, 50-55 (2023)**, ***Physical Review Letters*** **133, 066503 (2024)**].

Although we were unable to achieve superconducting superlattice films with greater thicknesses, we believe that the superconductivity therein stems from the intrinsic nature of the infinite-layer nickelates. Comparing all of our experimental results with infinite-layer nickelates single films, the characteristics are in general agreement. Thus, the superlattice structure does not significantly affect the superconductivity, although it is important to consider the potential impact of quantum effects and interfacial interactions. Our findings further imply, albeit indirectly, that the superconductivity observed in infinite-layer nickelates is an intrinsic property of the material.

Revision: We have added relevant discussions on **Page 2(Abstract)**, **Page 3 (Introduction)**, and **Page 9 (Conclusion)** of the manuscript. The above figures were also added to the Support Information as **"Figure S12"**.

On **Page 2**, "The superconducting superlattice showed a T_c of 12.5 K and a 2D superconducting feature, indirectly indicate the intrinsic superconductivity of infinite-layer nickelates."

On **Page 3**, "The consistent characterization with a single film suggests that the interface has little effect on its superconducting properties, indirectly demonstrating that infinite-layer nickelate superconductivity is intrinsic."

On **Page 9**, "This suggests that the superconductivity in the superlattice originates from the intrinsic properties of the infinite-layer nickelates, it is also confirmed in recently free-standing infinite-layer nickelates." … "Collectively, the findings on the superconducting properties of the infinite-layer nickelate superlattice closely mirror those of $Nd_{0.8}Sr_{0.2}NiO_2$ single films. This suggests that the interface between $SrTiO_3$ and the infinite-layer nickelate has no substantial impact on its superconducting behavior, serving as indirect evidence of the intrinsic superconductivity of infinite-layer nickelates.”

For Reviewer #3 (Remarks to the Author):

In the present work by W. Xiao et al. “Superconductivity in the infinite-layer nickelate superlattice” using soft-chemistry topotactic reduction the authors successfully synthesized nickelate heterostructures $(RNiO_{2+x})n/(SrTiO_3)_2$ with different thickness of the reduced nickelate layer, n. It was shown that above a critical thickness n>5 u.c. the high-quality nickelate heterostructures with infinite-layer crystal structure $RNiO_2/SrTiO_3$ can be stabilized. The authors perform a detailed analysis of the electronic state and crystal structure properties of these materials using different experimental techniques, such as x-ray diffraction, x-ray reflectivity, x-ray absorption, and x-ray linear dichroism measurements, in combination with the atomically resolved scanning transmission electron microscopy and resistivity measurements, etc. It was discovered that the nickelate heterostructures with n=8 u.c. display superconductivity with a critical temperature T_c~12 K, in close agreement to that in the hole-doped nickelate thin films. In contrast, the nickelate heterostructures with n<5 u.c. show insulating behavior which is presumably associated with a structural disorder in the reduced nickelates. To the best of my knowledge this is the first report on superconductivity in nickelate heterostructures with the infinite-layer crystal structure.

I read the manuscript with great interest. The results presented in the manuscript are interesting and scientifically sound. The paper is well organized and clearly written. In my opinion, the subject matter of the manuscript is suitable for the publication in the Nature Communications journal. The results discussed in the manuscript are at the high level of present experimental capabilities. I propose that this novel approach to study superconductivity in nickelates compounds will be highly appreciated in the future experimental and theoretical research. In fact, it opens a novel direction for experimental and theoretical research of the microscopic origins of superconductivity in nickelates. In addition, it allows one to use various effects such as quantum confinement and epitaxial strain to control the properties of investigated systems.

I believe that the manuscript meets the criteria for publication in Nature Communications and can be accepted after the authors address the following minor corrections:

Response: We greatly appreciate your high recognition of our work and constructive suggestions. Based on your questions, we have added relevant experiments and discussions. Below, we will respond to your questions point by point.

1) It was shown that the infinite-layer nickelate heterostructures with n=8 u.c. display superconductivity with a critical temperature T_c~12 K. Is this correct to say that for n>=8 u.c. the infinite-layer nickelate heterostructures show superconductivity? I mean the authors do not discuss (may be I missed this) what happens for n>8. n=8, is this the only point at which this systems superconducts or it does for n>=8 u.c. Is there experimental evidence (support) that, e.g., for n=9 and 10 the reduced nickelate heterostructures are also superconducting? Please clarify this point. It might be great to present extra experimental data for n>8.

Response: We sincerely appreciate your valuable advice. We have added data for both thinner and thicker samples, as shown below in **Figure R2**.

When *n* exceeds the critical thickness of 5 and is less than 10, the resultant reduced superlattices possess a well-defined infinite-layer structure and exhibit superconducting in transport, with $T_{c,\mathrm{onset}}$ values ranging from 8 to12 K. The resistances of R-N_7/S_2 and R-N_9/S_2 could not completely tuned to zero, which may be due to the effect during the reduction process [***Nature*** **615, 50-55 (2023)**, ***Physical Review Letters*** **133, 066503 (2024)**].

However, when n = 9, the intensity of the reduced infinite-layer phase decreases, and when n ≥ 10, the peak position of the reduced film cannot reach 55° and the peak intensity is also weak, despite the fact that the reduction time has been very long (24 h). This is due to the fact that our superlattice has 10 cycles, and as n increases, the stabilizing effect of the spacer layer $SrTiO_3$ on the structure will be gradually weaken. Consequently, some undesirable RP phases begin to emerge, ultimately impacting the results. Thus, it becomes challenging to achieve a pure infinite-layer phase for excessively thick superlattice films due to various influence of miscellaneous factors during the growth process of nickelate, which complicates the realization of thicker ($n \geq 10$) superlattice films with superconductivity.

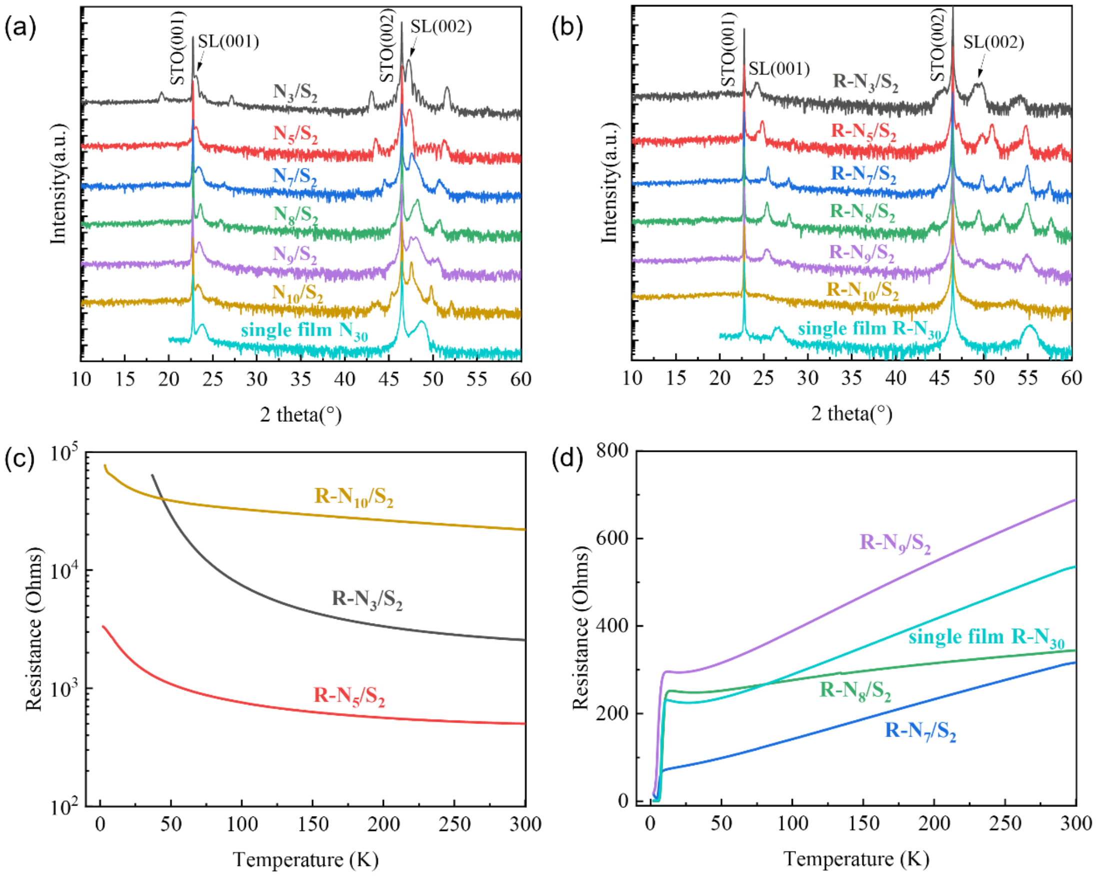


Figure R2. θ-2θ XRD spectra of (a) as-grown (N_n/S_2) and (b) reduced (R-N_n/S_2) superlattices (SLs) with different thickness of nickelates. As n＞9, R-N_n/S_2 is hard to obtain a pure infinite-layer structure. Temperature-dependent resistance R(T) of R-Nm/S2 , (c)when $n \leq 5$ and n＞9, the R- N_n/S_2 superlattices exhibit insulating behavior, (d) as 5＜n ≤ 9, the R- N_n/S_2 superlattices are superconducting. A single film of 30 u.c. $Nd_{0.8}Sr_{0.2}NiO_{2,3}$ with the same thickness of $SrTiO_3$ capping layers is also included in the **(a)**, **(b)** and **(c)** as comparison data (light blue lines).

Revision: The above figures were added to the Support Information as **"Figure S12"**.

2) The authors present a detailed analysis of the electronic structure and structure properties of the synthesized nickelate heterostructures. At the same time, compositions of these materials are discussed very briefly. It was claimed that it is at the optimally hole-doped state with Nd/Sr=0.8/0.2. How the authors determine a composition of the reduced nickelate heterostructures (Nd/Sr=0.8/0.2)? I mean it can be affected upon topotactic reduction of the well-characterized perovskite precursor phase with Nd/Sr=0.8/0.2. I believe that these materials can also be highly inhomogeneous.

Response: Thanks for the reviewer's constructive comments. Topological hydrogen reduction is a difficult process to control, so it is important to characterize the elemental distribution and the degree of homogeneity in the superlattice. However, accurate characterization of the Nd/Sr ratio is difficult due to the presence of Sr elements in the substrate and the interstitial layers of the superlattice. As the uniformity of elemental distribution can indirectly indicate whether the Nd/Sr ratio changes, we further characterized the R-N_8/S_2 sample with EELS elemental mapping resolution. It is shown in the following figures.

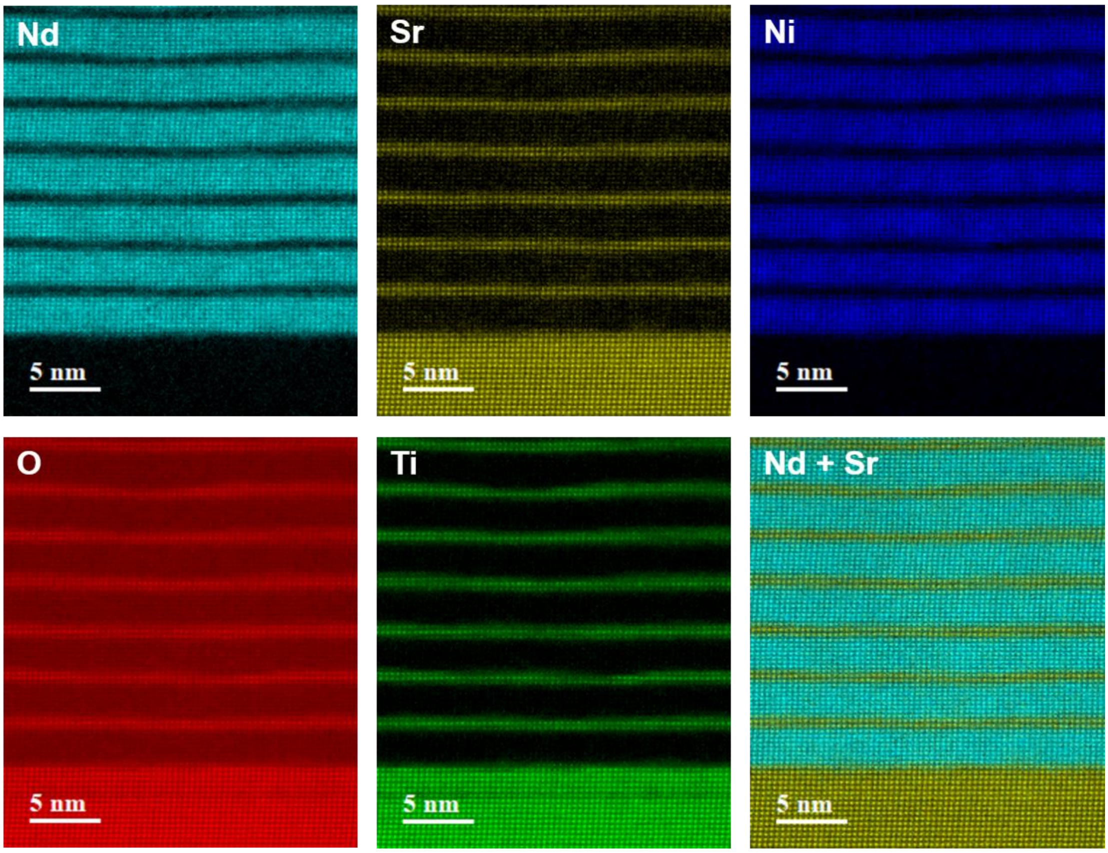


Figure R2. STEM-EELS mappings of R-N_8/S_2.

It can be seen that the distribution of elements in the superlattice is very homogeneous and there is no elemental agglomeration or diffusion. In addition, the topological hydrogen reduction mainly affects the anionic oxygen, and has less effect on the cations in the A-site or B-site. Therefore, the Nd/Sr ratio of the R-N_8/S_2 should be consistent with the epitaxial parent N_8/S_2 film (nominal Nd/Sr = 0.8/0.2).

Revision: We have added the above results to the Support Information as "**Figure S10".**

3) The authors do not discuss the magnetic properties of these materials. Is there any experimental evidence of a long-range ordering (spin or charge density wave behavior) in these materials? I understand that it is too much to ask the authors to perform such measurements. A brief discussion might be sufficient (if possible).

Response: Thank you for your valuable suggestions. Magnetic order in infinite-layer nickelates is currently an underexplored topic. For cuprates, the association between the strong correlation and the antiferromagnetic order leads to a symmetry-breaking lack of order, which produces the corresponding charge density wave or spin density wave. The Q = (0.33, 0) charge density wave observed in a recent study on the infinite-layer nickelate parent phase seems to indicate similar physics in nickelates [***Nature Materials*** **21, 1116-1120 (2022)**, ***Nature Physics*** **18, 869-873 (2022)**]. This charge order has been attributed to the hybridization between the Re-5d and Ni-3d orbitals. This $3a_0$ charge order is also inconsistent with the previously reported RP-phase $Nd_4Ni_3O_8$. The former is oriented along the Ni-O bond direction (h, 0), and the latter suffers from a 45° rotation (h, h). This charge order is also thought to be related to capping $SrTiO_3$ layers [***Physical Review Letters*** **129, 027002 (2022)**]. However, a recent work has shown that this observed charge order is due to an intermediate phase ($Nd_3Ni_3O_7$ or $Nd_3Ni_3O_8$) of the reduction process and that fully reduced $NdNiO_2$ is not characterized by a charge order [***Nature Materials*** **23, 486–491 (2024)**]. The intermediate $Nd_3Ni_3O_7$ or $Nd_3Ni_3O_8$ have similar $3a_0$ space configurations, where the excess apical oxygen atoms (or vacancies) are arranged in rows with $3a_0$ periodicity, forming a 3 × 1 × 3 supermonomer with a superlattice peak at the assumed charge order wave vector Q = (1/3, 0, 1/3) [***Nature Materials*** **23, 486–491 (2024)**]. When Sr doping is introduced, the nickelates show an increase in entropy, rendering them more susceptible to complete reduction. This leads to a lower concentration of $Re_3Ni_3O_{7,8}$, which in turn suppresses the intensity of the Q = (0.33, 0) Bragg peak [***Nature Materials*** **23, 486–491 (2024)**].

In conclusion, the existence of charge density waves in infinite-layer nickelates continues to be a topic of debate. The purity of the infinite-layer phase in the nickelate significantly influences the experimental results, and our superlattice R-N_8/S_2 samples achieved thinner pure infinite-layer phases by interpolated $SrTiO_3$ layers. Building on this, further investigation of the charge density wave order in this superlattice will help to further clarify this issue. In our subsequent work, we will focus on the above question in these samples to deepen our understanding of the physics underlying infinite-layer nickelates.

4) It turns out to me that resistivity data for the R-N_8/S_2 system show a weak anomaly at about 140 K. It can be related to a charge or spin density wave formation in the reduced nickelate heterostructures, i.e., a long-range ordering stabilized in the infinite-layer nickelates due to quantum confinement. It seems that a similar behavior also appears in the resistivity data of the Ruddlesden-Popper bilayer and trilayer bulk nickelates. It could be interesting to add a brief discussion on this (if possible).

Response: We appreciate your constructive suggestions, but we believe that this slight anomaly in the resistance value should be due to instrumental error during the measurement process. To further prove this, we re-measured the temperature dependence resistance of this sample, and at the same time, we re-prepared a new piece of R-N_8/S_2 sample and measured it. The raw data is shown in the figure below.

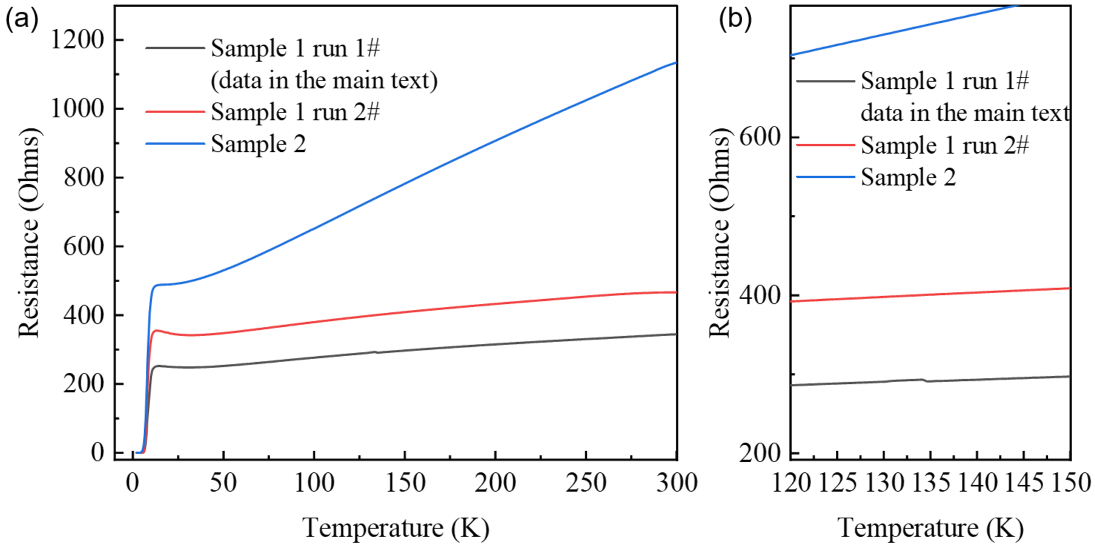


Figure R3. (a) Temperature-dependent resistance *R*(T) of two different R-N_8/S_2. (b) shows an enlarged view of (a) in the temperature range of 120 K-150 K.

It can be seen that after retesting the sample, this slight anomaly disappeared, and another sample also did not show any anomalies. Therefore, it can be concluded that the aforementioned anomaly was caused by errors due to temperature fluctuations of the instrument during the testing process or other factors.

5) In the paper and in the supplemental material the authors discuss somewhat different compositions of the nickelate heterostructures. In the paper these are N_x/S2 and R-N_x/S2, while in the SM are Nx/S3 and R-N_x/S3. It looks like that no additional data presented on the Nx/S2 and R-Nx/S2 in the SM. It also might be great to make a conclusion how the thickness of the $SrTiO_3$ layer affects the properties of these materials (S2 vs. S3).

Response: We appreciate your constructive suggestions. Our N_x/S_3 and R-N_x/S_3 samples were obtained prior to the optimization of the nickelates target, and these data have only enlightened us in terms of the thickness-dependent structure transition as well as the critical thickness. Notably, the N_x/S_3 sample underwent an extended reduction process of nearly 20 hours. We found that using a thinner $SrTiO_3$ spacer layer can help shorten the reduction time and mitigate the effects of the topological reduction process on the film. Following the optimization of the target, we reduced the thickness of the $SrTiO_3$ in the superlattice to just 2 layers. According to our additional data, the structural transition with respect to the nickelate thickness and the critical thickness remain the same even after reducing the thickness of $SrTiO_3$ layers. In addition, after reducing the thickness of $SrTiO_3$ in the superlattice, the reduction time is reduced to 8-10 h, significantly reducing the uncontrollability of the topological reduction process. However, deriving specific effects of the SrTiO3 intercalation layer thickness on its properties from the existing data alone proves challenging. Nevertheless, this remains a significant area for further exploration, and we plan to conduct a systematic investigation on it in our future work.