nature portfolio

Peer Review File

Re-entrant unconventional superconductivity induced by rare-earth substitution in $Nd_{1-x}Eu_xNiO_2$ thin films

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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)
Question 1: OK.
Question 2: OK.
Question 3: Mostly OK. I appreciate the authors’ inclusion of additional details on the fitting procedure, which makes the analysis much clearer. One remaining point of confusion concerns the second fitting approach referred to as the d-wave BCS model. I do not see the gap parameters explicitly appearing in the fitting functon(Eq. S5) in the Supplementary Information. I assume these are included implicitly through σ_d (ω). If this is the case, I suggest the authors explicitly show the expression and clarify the role of the gap parameters for readers who may not have a strong spectroscopy background. If this assumption is incorrect, please clarify accordingly.
Question 4: OK.
Question 5: OK.
Question 6: I understand that the authors may not feel comfortable commenting extensively on other groups’ results. However, given that similar samples are studied elsewhere and magnetism can play an important role in superconducting properties, it would be helpful to include a brief acknowledgment of the relevant existing literature.
More comments: The figures would benefit from a careful cleanup to ensure consistent styling. Currently, different fonts are used across figures, and some labels are missing. For example, Fig. 4 lacks panel labels (a) and (b), and Fig. 3 appears to have inconsistent fonts and aspect ratios.

Reviewer #2

(Remarks to the Author)
The authors have significantly improved the manuscript. I have no more comments. I recommend publication as it is.

Reviewer #3

(Remarks to the Author)
The authors have dramatically improved the quality of this manuscript in the previous stages of peer review. My opinion prior to the transfer of this manuscript was that the work is scientifically sound and will be of importance to the Nickelate community. Therefore, I recommend publication of the work without extra modifications.

RESPONSE TO REFEREE REPORTS

We thank the reviewers again for the assessment of the work. With the transfer of the manuscript to Nature Communications, we enclose our point-by-point response to all the remaining comments here, in blue, and provided a revised manuscript and Supplemental Information as separate files with our changes in blue. We believe the manuscript and the attached response address all the comments from the reviewers and that the manuscript is substantively improved and suitable for publication in Nature Communications.

Reviewer #1 (Remarks to the Author):

In my original report, I raised several concerns regarding the impact, interpretation, rigor, and clarity of the manuscript relative to the standards expected by Nature Materials. I note that some of these issues were also highlighted by Referee #3. The revised version shows certain improvements; however, there remain vague and sometimes evasive discussions that fail to adequately address the central scientific issues. While the work has potential, the current manuscript still falls significantly short of Nature Materials standards in terms of impact, clarity, and robustness of results. Below I elaborate on my main points.
1. The reported results represent an incremental advance over the group's previous work (e.g., extending magnetic field from 14 T to 60 T, identifying the Jaccarino–Peter effect, and reporting a large superconducting gap). While these observations are of interest, they largely constitute additional measurements rather than a conceptual breakthrough in materials physics. The DFT calculations appear disconnected from the microscopic physics of the pairing symmetry. As I stated in my initial review, incremental progress could still be acceptable if the data and analysis were rigorous and conclusive; however, I remain concerned about these aspects.

We respectfully do not agree with the description of our work as merely incremental. Our results represent significant experimental evidence that challenges prior results about nickelate superconductivity:

First, the discovery of strong-coupling superconductivity in nickelates directly contradicts the weak-coupling picture established by all prior studies, including the recent Nature Materials paper (*Nature Materials 23, 775–781 (2024)*). These are not just "additional measurements", they fundamentally change our understanding of the pairing mechanism. While it is still inconclusive on exactly how the presence of Eu affects coupling strength, the results will substantially impact future theoretical studies aimed at understanding the microscopic mechanism of superconductivity in nickelates and its relationship to cuprates.

Second, the observation of magnetic-field-induced re-entrant superconductivity via the Jaccarino-Peter mechanism represents a rare and striking phenomenon. Among known superconductors, only a handful of systems exhibit this behavior (e.g., certain heavy fermion compounds). Our work establishes nickelates as a new platform for studying the interplay between magnetism and superconductivity, a central problem in condensed matter physics. The fact that competing groups have now reported similar findings (*arXiv:2508.14666* and *arXiv:2508.16290*) utilizing highly-sought-after resources such as pulsed high magnetic field facilities underscores the significance and timeliness of this discovery.

Regarding the DFT calculations, we note that these provide crucial microscopic support for the Jaccarino-Peter mechanism by demonstrating how Eu substitution introduces the necessary magnetic exchange field. While they do not address effect of Eu substitution on pairing symmetry, they are essential for validating the proposed mechanism.

2. The measurement of the superconducting gap is important, but not sufficiently novel on its own. The data do not convincingly demonstrate the fraction of uncondensed quasiparticles—while the fitting suggests a possible hint, it does not distinguish clearly between s-wave and d-wave symmetry.

The purpose of the optical measurements is to measure the **size of the superconducting gap which represents a major finding**: $\boldsymbol{2\Delta/k_BT_c \simeq 5-6}$**, indicative of strong-coupling superconductivity**. As we have stated explicitly in our previously revised manuscript: Optical spectroscopy, not being a momentum-resolved technique, is unable to provide definitive conclusions about the symmetry of the superconducting gap.

As discussed in the previous rebuttal, the magnitude of the fit parameters remains a significant observation. The s-wave fit requires a normal-state conductivity more than one order of magnitude lower than the value measured via DC transport. The reflectivity spectra can be fitted with reasonable s_{DC} values only if a substantial portion of the spectral weight in σ_1 below the gap persists to the lowest temperatures. This behavior is inconsistent with the complete suppression expected for a fully gapped s-wave superconductor and is naturally explained by the presence of nodes in the superconducting gap, where residual quasiparticle would accumulate. To make it clearer, we tabulated the obtained fit parameters in tables S4, S5 of the Supplemental Information Section S7.

Moreover, the assumption of inhomogeneous or filamentary superconductivity appears ad hoc.

An effective medium approach was employed to consistently fit the reflectivity spectra using the exact value of the normal-state conductivity measured via DC transport.

As discussed in the manuscript, in spite of possible inclusion of normal regions, the observation of robust zero resistance at high magnetic fields, along with a high critical current density and a large diamagnetic screening signal, demonstrates the bulk nature of superconductivity in our samples.

The references cited in response to my earlier comment (#2) show optical conductivity both below and above, which is missing here and is essential for a complete analysis.

The work referred to by the reviewer was carried out with a different technique than ours, namely THz time-domain spectroscopy. This technique allows, after appropriate modeling of the substrate response, to directly extract both the real and imaginary part of the optical conductivity of the thin film.

Our measurements were performed instead with Fourier Transform Infrared Spectroscopy, such as those on LSNO in Cervasio *et al.*, *ACS Applied Electronic Materials* **5**, 4770-4777 (2023). While through reflectivity ratios at various temperatures we are sensitive to very small changes across the superconducting transition, in order to extract the complex optical conductivity, we would have to rely on Kramers-Kronig transformations. This procedure, given the limited measurement range, would introduce considerable uncertainties. To avoid unwanted uncertainties, following the same standard approach as that of Cervasio *et al.*, as well as that of other works using the same technique, we directly fitted the reflectivity ratios with a multilayer model, thus extracting the parameters related to the response of the superconducting film. Overall, the information obtained from our experiment (superconducting gap, normal state conductivity, quasiparticle contribution) is the same as that from the other works mentioned.

3. I remain disappointed by the authors' response to my suggestion for a more systematic presentation of the optical conductivity fitting. At a minimum, the manuscript should list all key equations in a logically consistent manner, include detailed descriptions of fitting parameters, and tabulate the extracted values.

We have followed the referee's suggestion and improved the discussion related to the various fit models in the revised version of Section S7 of the Supplemental Information. We have also included tables S4 and S5 with fit parameters.

Currently, the discussion remains fragmented and overly general, with statements such as: "This approach involves calculating Fresnel reflection and transmission coefficients at each interface..." and "Due to the lack of interference effects...the interfaces were assumed to be incoherent."
These claims raise concerns. For instance, it is unclear how a film only 6 nm thick could reasonably fall into the incoherent Fresnel reflection limit.

There was a misunderstanding here. The absence of interference effects referred to the response of the substrate and in particular to the absence of a reflected signal from its back surface, which was not polished in our sample. In contrast, the coherent effects at the Al_2O_3/NENO and NENO/LSAT interfaces were correctly accounted for in the model.

To clarify, we have modified the sentence in question in section S6 of the Supplemental Information, which now reads:

"Note that the coherent effects at the Al_2O_3/NENO and NENO/LSAT interfaces were taken into account in the model. In contrast, the contribution from the back surface of the substrate (not polished in our sample) was neglected."

4. I strongly recommend additional measurements across different doping levels and Tc values. Given the modest novelty and inconclusive evidence so far, these are essential. For example, the gap size versus temperature (Fig. 4b) shows no systematic variation within experimental uncertainty, suggesting inadequate sampling. The authors dismiss this by stating that the optical measurements were performed at too low a temperature; however, the data shown are at 4 K, whereas other dopings exhibit Tc > 10 K. This rationale is unconvincing and does not meet the level of experimental rigor expected for Nature Materials.

We agree that a broader doping-dependent optical spectroscopy dataset would be good to have and is a clear priority for future work, possibly with an improved experimental setup. We reiterate here how the need to use windows limits the ability to cool the samples to the lowest temperatures to perform optical experiments that can resolve features of the superconducting gap, which are generally harder than transport measurements.

Specifically, optical measurements on samples with lower critical temperatures would have a high chance of failure. A direct measurement of features related to the superconducting gap would be made extremely challenging for both lower and high doping levels (i) by the fact that the gap itself, given the lower T_C, would be smaller and likely fall below our measurement range and (ii) by not being able to reach sufficiently low temperatures, ideally below $\sim T_C/2$, to observe the response of a fully developed order parameter. As a result, we present optical spectroscopy data only for the optimally doped sample with the highest critical temperature.

5. To convincingly demonstrate that the observed superconductivity is of bulk rather than filamentary nature, clear and quantitative evidence of Meissner diamagnetism is essential. This constitutes the bare minimum standard for establishing bulk superconductivity, yet such evidence is currently absent.

As discussed in the Supplemental Information Section S8, the observation of robust zero resistance at high magnetic fields, along with a high critical current density and a large diamagnetic screening signal from mutual inductance measurements, demonstrates the bulk nature of superconductivity in our samples. These multiple independent measurements provide compelling evidence against filamentary superconductivity.

6. Several recent studies have proposed a key role of ferromagnetism in the re-entrant superconducting behavior. For instance, a recent arXiv report (2508.14666) attributes the effect to ferromagnetism of the Eu^{2+} sublattice. In contrast, the present manuscript does not report any ferromagnetic order. It would be desirable, though not strictly essential, for the authors to clarify this apparent inconsistency and discuss how their findings relate to those reports.

While ferromagnetism remains a possibility that warrants further investigation, the magnetic measurements in *arXiv:2508.14666* show features that suggest contributions beyond simple Eu^{2+} ordering. Specifically, that report shows magnetic moments reaching up to 400 μB/Eu in their x=0.22 sample and >10 μB/Eu in x=0.34 and x=0.55 samples, values much higher than the expected saturated moment of 7.94 μB/Eu for Eu^{2+}. This indicates that the magnetic moment may originate from sources other than Eu^{2+}. Additionally, without temperature-dependent magnetization data and given the absence of hysteresis in Hall and magnetoresistance measurements (which would be expected for intrinsic ferromagnetism), we find the evidence for ferromagnetic order inconclusive at this stage. Prior studies have also pointed toward spin-glass states or NiO inclusions as possible sources of magnetic hysteresis. We refrain from claims of magnetic ordering in our system until more definitive experimental evidence is available.

Reviewer #2 (Remarks to the Author):

I have read the modified manuscript by the Ahn team, as well as their descriptive response to each point raised by the reviewers. Going point by point through the response provided by the authors:
1) The removal of kinks is good in Fig.2, but I still don't like this data presentation style as it is not visually clear how many sweeps there are. At a bare minimum, all sweeps should be shown in the SI.

Following the reviewer's suggestion, we now include all sweeps in the Supplemental Information, Section S9.

2) These statements based on models may be plausible, but I still only consider it to be valid if additional thickness samples were investigated due to the large number of parameters used in all fitting procedures.

While the analysis is plausible, it is best to validate using experimental study of thickness dependence, as the reviewer suggests. We thank the reviewer for this idea and will work on that in future studies.

3) Thank you for clarifying this.
4) Thank you for expanding on the discussion on the fitting procedure, which is very helpful. I find it surprising that \lamba_SO is so small, but as the authors note, it is connected to \alpha, which makes the fitting procedure challenging.
5) Thank you for clarifying this section, which is now significantly improved.
6) Thank you for clarifying this point.

We appreciate the reviewer's comments on the improvements and clarifications we have made.

In summary, I think the work is scientifically sound, especially given the challenges associated with accessing the high magnetic field lab for experiments. But there remains the question of why this work is significant beyond the niche case of nickelates.

We thank the reviewer for their careful evaluation and constructive feedback.

Regarding the significance of this work beyond nickelates, recently there has been increasing attention to studies of upper critical fields in different materials, with a special emphasis on Pauli-limit violation.

A few examples include graphene-based superconductors and UTe_2. For example, in UTe_2 there are 3 distinctively different superconducting phases, which behave very differently under the application of the external magnetic field: some superconducting phases are suppressed by the field, some are enhanced, and some exist only in ultra-high magnetic fields. Understanding which mechanisms lead to field-induced superconductivity is a fundamental question with potential for discovering new superconducting materials and states with practical applications.

The Jaccarino-Peter mechanism is among a few known microscopic mechanisms to maintain superconductivity at very high magnetic fields. Our work aims towards a Jaccarino-Peter-like scenario in NENO. This finding establishes a synergetic relationship between local magnetism and superconductivity in a way that parallels heavy-fermion and uranium-based superconductors, but in a chemically and structurally distinct platform. Therefore, beyond the specific case of nickelates, our results contribute to the fundamental understanding of how magnetism and superconductivity, typically antagonistic phenomena, coexist and even reinforce each other, aiding the engineering of new field-tunable or magnetically mediated superconductors across a wide range of materials.

Reviewer #3 (Remarks to the Author):

In the revised version, the authors have significantly improved the quality of the manuscript. Below I provide two more comments:

1. High Tc superconductivity is usually associated with strong coupling and a large superconducting gap. In this manuscript, the authors found that the substitution of Eu strengthens the superconducting gap of Nd-based nickelate superconductors. However, the Tc of these samples are similar to those of Nd1-xSrxNiO2 (15-20 K). They seem inconsistent.

Thank you for the insightful comment. The observation of a larger superconducting gap in Eu substituted Nd-based nickelates, together with a moderate T_C similar to that of $Nd_{1-x}Sr_xNiO_2$, is not inconsistent with unconventional strong-coupling superconductivity. Such a disproportionate increase of gap-over-Tc ratio is a hallmark of strong coupling superconductivity. For example, in $La_{2-x}Sr_xCuO_4$ (LSCO), the gap-to-Tc ratio in underdoped LSCO can reach ~8, with a moderate T_C often below 40 K; the ratio in over doped Bi-2212 can reach ~10 near optimal doping (*Science 362,62-65(2018)*); and this ratio also reaches 4.6 in Pb - an archetypal strong coupling superconductor with modest T_C of 7.2K.

These examples illustrate that strongly coupled superconductors could produce larger gaps at the same Tc of their weaker coupled counterparts. Notably, this is also seen in iron-based superconductors, which is in a similarly intermediate coupling regime to NENO (see Figure 25, *Reviews of Modern Physics 93, 025006 (2021)*).

2. Figures 2g,h need some improvement. A zoom-in figure would help to show if zero resistance is achieved under high fields (re-entrant superconductivity).

Following the reviewer’s suggestion, we revised Figures 2g,h to show the achieved zero resistance under high fields.

RESPONSE TO REFEREE REPORTS

We thank the reviewers again for the assessment of the work. We enclose our point-by-point response to all the remaining comments here, in blue, and provided a revised manuscript and Supplemental Information as separate files. We believe the manuscript and the attached response address all the comments from the reviewers and that the manuscript is suitable for publication in Nature Communications.

Reviewer #1 (Remarks to the Author):

Question 1: OK.
Question 2: OK.
Question 3: Mostly OK. I appreciate the authors' inclusion of additional details on the fitting procedure, which makes the analysis much clearer. One remaining point of confusion concerns the second fitting approach referred to as the d-wave BCS model. I do not see the gap parameters explicitly appearing in the fitting functon(Eq. S5) in the Supplementary Information. I assume these are included implicitly through σ_d (ω). If this is the case, I suggest the authors explicitly show the expression and clarify the role of the gap parameters for readers who may not have a strong spectroscopy background. If this assumption is incorrect, please clarify accordingly.

Response: Following the reviewer's suggestion, we have included the formula for the d-wave Mattis-Bardeen model in the revised manuscript (lines 334-345 in the Supplementary Information) and clarified the role of the gap parameters. This explicit inclusion will assist readers who may not have a strong background in spectroscopy.

Question 4: OK.
Question 5: OK.
Question 6: I understand that the authors may not feel comfortable commenting extensively on other groups' results. However, given that similar samples are studied elsewhere and magnetism can play an important role in superconducting properties, it would be helpful to include a brief acknowledgment of the relevant existing literature.

Response: We have now incorporated our previous response to the reviewer's comment on magnetism in similar samples into the revised manuscript (lines 53-60 in the Supplementary Information) to acknowledge the relevant existing literature.

More comments: The figures would benefit from a careful cleanup to ensure consistent styling. Currently, different fonts are used across figures, and some labels are missing. For example, Fig. 4 lacks panel labels (a) and (b), and Fig. 3 appears to have inconsistent fonts and aspect ratios.

Response: We appreciate the reviewer's attention to detail. We have carefully reviewed and edited the figures to ensure consistent styling and proper labeling. The revised figures are submitted as editable files for editorial purposes.

Reviewer #2 (Remarks to the Author):

The authors have significantly improved the manuscript. I have no more comments. I recommend publication as it is.

Response: We sincerely thank the reviewer for their careful evaluation and constructive feedback, which have significantly improved the quality of the manuscript.

Reviewer #3 (Remarks to the Author):

The authors have dramatically improved the quality of this manuscript in the previous stages of peer review. My opinion prior to the transfer of this manuscript was that the work is scientifically sound and will be of importance to the Nickelate community. Therefore, I recommend publication of the work without extra modifications.

Response: We thank the reviewer for their thoughtful evaluation and constructive feedback. We are pleased that our manuscript has met their expectations and appreciate their support for its publication.