

High-field-stabilized reentrant superconductivity in infinite-layer nickelate 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)

The authors have significantly improved their manuscript, both in terms of scientific rigor and in presentation and readability. Below I evaluate their responses to my original comments, as well as the comments of reviewer 2.

- I appreciate that the sample characterization now appears in the manuscript. I have one follow-up question based on the data presented in Figure R3/E1. How is the sample thickness determined? Because if one extracts the Scherrer width from the optimally reduced samples, one obtains thicknesses much lower than the ones reported. It seems quite likely that there is some sample decomposition leading to some amorphous phase.
- What is the difference between the two Eu²⁺ curves on Fig R7/E2 B+D?
- In Fig R6/E4 I understand that the authors have subtracted a background signal that was recorded without the sample. Can the authors confirm if the background signal was field dependent or is it the same background subtraction for all curves?
- Also on Fig R6/E4, it is also not clear how this data correlates with the transport data. Some indication on the plots of what regions are dissipative vs superconducting would be helpful.
- The authors state that the spin-flop transition of Nd results in “secondary features” in the low temperature transport. Can they be clearer on what is meant by that?
- Why does it appear that Fig R8/2 B does not follow the model?

My opinions on the responses to Reviewer 2

1. The authors have responded satisfactorily.
2. The authors have responded satisfactorily.
3. The authors have responded satisfactorily.
4. The authors have decided that a quantitative analysis is beyond the scope of the work. They say that they have a “data-driven” approach, which usually means a high level of quantification, and this is not the case. They also say that they will not conduct DFT calculations, but this is not what reviewer 2 was suggesting. My understanding is that reviewer 2 merely suggested estimating interaction energies from the measurement.
5. The authors have responded satisfactorily. I do have some questions: What is meant by suggesting that the composition modifies “exchange pathways”? I doubt that the type of exchange interaction is strongly modified in the narrow range of compositional space reported here. Also, they mention the “topology” of the reentrant regime. How is the topology relevant?
6. The authors have responded satisfactorily. I would also add that there is precedent for magnetic moments in the superconducting phase of infinite layer nickelates. Reviewer 2 refers to work claiming ferromagnetic order. I understand they refer to the work from Hong Kong. In my opinion, that preprint is mistaken in interpreting magnetic order. They do not have sufficient evidence and my understanding is that those authors are revising their claims. So I believe reviewer 2 was slightly off-base when making this comment.
7. The authors have responded satisfactorily.

Version 1:

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Reviewer #1

(Remarks to the Author)

The authors have responded satisfactorily to my comments.

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Dear Editor and Reviewer,

We thank you for your feedback on the revised version of our manuscript and are pleased to note that the reviewer found that the manuscript is significantly improved, both in terms of scientific rigor and in presentation and readability. In response to reviewer #1's additional comments, we have revised Extended Data Figure E4, and captions of Extended Data Figures E2 and E4. For figure E4, we have shaded the SC region to highlight the SC regime from the transport data. We have also added a sentence in the Method section about the estimation of films thicknesses. Below is our point-to-point response to all additional comments.

Reviewer's comments are in black; our responses are in blue. Minor changes made in the manuscript are highlighted in red here as well as in the manuscript text.

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

The authors have significantly improved their manuscript, both in terms of scientific rigor and in presentation and readability. Below I evaluate their responses to my original comments, as well as the comments of reviewer 2.

Response: We greatly appreciate reviewer #1 for evaluating our response to their comments as well as Reviewer #2 comments. We are happy that the reviewer found that the revised manuscript is significantly improved.

- I appreciate that the sample characterization now appears in the manuscript. I have one follow-up question based on the data presented in Figure R3/E1. How is the sample thickness determined? Because if one extracts the Scherrer width from the optimally reduced samples, one obtains thicknesses much lower than the ones reported. It seems quite likely that there is some sample decomposition leading to some amorphous phase.

Response: We thank the referee for raising this concern. Firstly, we have calculated the Scherrer width using the following equation and X-ray diffraction (XRD) data of sample S3:

$$D_{hkl} = \frac{K \lambda}{\beta_{hkl} \cos \theta}$$

where:

- β_{hkl} is the FWHM of the XRD peak
- K is the shape factor
- θ is the Bragg angle

- λ is the wavelength of the X-ray source

For the (002) peak for sample S3,

$$\theta_{hkl} = 2.16^\circ$$

$K = 0.9$ (which is widely accepted on thin films when detailed information on crystallite morphology is not well define [1-4])

$$\theta = 28.205^\circ$$

$$\lambda = 0.154 \text{ nm (Cu-K}\alpha \text{ line)}$$

The FWHM θ_{hkl} and the Bragg's angle θ values are obtained from the (002) peak in the infinite-layer (IL) XRD spectrum, fitted to a pseudo-Voigt function. The Scherrer width D_{002} could be estimated as:

$$D_{002} = \frac{0.9 * 0.154}{\left(\frac{2.16 * 3.14}{180}\right) \cos\left(\frac{56.41}{2}\right)} \cong 4.175 \text{ nm}$$

The estimated value is not too different from the thickness of sample S3 (6.5 nm). From the small discrepancy of the two values, we agree that partial decomposition or structural disorder might be present, but the effect should be very small. Afterall, the Scherrer width value obtained should be interpreted as a crystalline coherence length instead of the exact physical film thickness.

In addition, it is very common that the XRD peak width could not directly reflect sample thickness in thin films but could be often influenced by substrate-induced strain, as well as defects and structural disorder. For this reason, XRD alone does not always provide a reliable thickness estimate for ultrathin films. Related to this, we also performed cross-sectional scanning transmission electron microscopy (STEM) on several nickelate thin films [5] and representative SECNO samples to independently determine the film thickness occasionally. The STEM-measured thicknesses serve to calibrate the growth rate of the thin film in terms of deposition pulses per unit thickness. Since all samples were grown under the same pulse conditions and within the same energy range, we expect the growth rate to be consistent across all SECNO samples, and therefore the film thickness could be accurately estimated accordingly.

The following sentence is included in the Methods section: *The film thicknesses are estimated from the number of deposition laser pulses, with the growth rate calibrated by cross-sectional scanning transmission electron microscopy (STEM) in terms of deposition pulses per unit thickness [43].*

References:

1. Klug, H. P. & Alexander, L. E. X-Ray Diffraction Procedures 2nd edn, Ch. 9 (Wiley, 1974)
2. Langford, J.I. and Wilson, A.J.C. Scherrer after sixty years: A survey and some new results in the determination of crystallite size. J. Appl. Cryst., 11: 102-113, (1978)
3. Cullity, B. D. Elements of X-ray Diffraction, 2nd edn (Addison-Wesley, 1978)
4. Holzwarth, U., Gibson, N. The Scherrer equation versus the 'Debye-Scherrer equation'. Nature Nanotech 6, 534 (2011)
5. Zeng et al., Origin of a Topotactic Reduction Effect for Superconductivity in Infinite-Layer Nickelates, Phys. Rev. Lett. 133, 066503 (2024)

- What is the difference between the two Eu²⁺ curves on Fig R7/E2 B+D?

Response: We appreciate referee's comment on Fig E2 which helps to bring clarity to this figure.

In Fig. E2B and E2D, the black curves represent the magnetization calculated by considering only the $m_J = \pm 7/2$ states, whereas the orange curves represent the total magnetization obtained by summing contributions from all possible m_J states ($\pm 7/2, \pm 5/2, \pm 3/2$, and $\pm 1/2$). These details have been clarified in the caption of Fig. E2B. The revised caption is as follows:

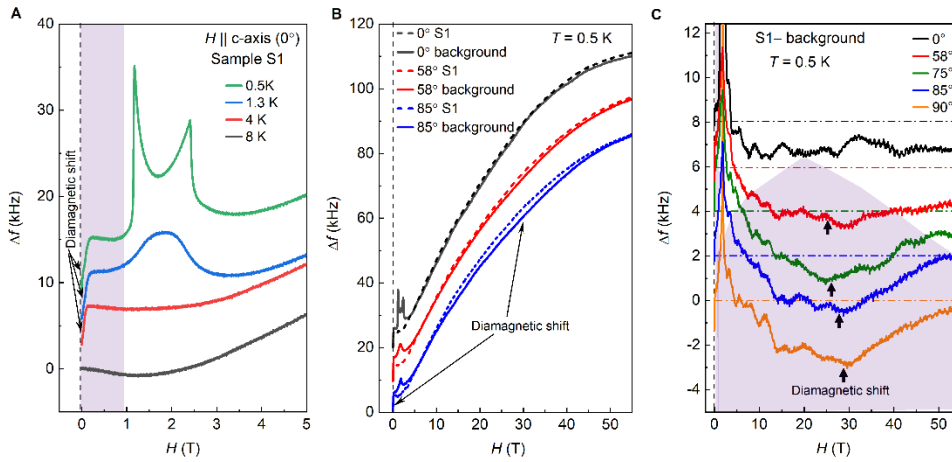
*Extended Data Fig. 2. **Anomalous Hall effect and NdGaO₃ substrate magnetization compared with Brillouin functions.** (A) Anomalous Hall effect normalized to its saturation value at three temperatures above T_c , plotted versus magnetic field H . The total Hall signal is shown in the inset. (B) Brillouin function M/M_s for $m_J = \pm 7/2$ at the same three temperatures. The inset compares this Brillouin function with those expected for the various magnetic ions present in the film or substrate at $T = 13$ K, including all m_J states. (C) Magnetization of the NdGaO₃ substrate measured in pulsed magnetic fields at several temperatures, indicating a spin-flop transition out of the low-temperature antiferromagnetic phase near ≈ 2 T. (D) calculated Brillouin functions at $T = 0.5$ K. **The black curve represents the magnetization calculated by considering only the $m_J = \pm 7/2$ states, whereas the orange curve represents the total magnetization obtained by summing contributions from all possible m_J states ($\pm 7/2, \pm 5/2, \pm 3/2$, and $\pm 1/2$).***

- In Fig R6/E4 I understand that the authors have subtracted a background signal that was recorded without the sample. Can the authors confirm if the background signal was field dependent or is it the same background subtraction for all curves?

Response: In Fig. E4, the background is field dependent as shown as the dashed lines in Fig. E4(B). We measured background at each angle after removing the sample from the coil. We have included this detail in the figure caption of E4.

- Also on Fig R6/E4, it is also not clear how this data correlates with the transport data. Some indication on the plots of what regions are dissipative vs superconducting would be helpful.

Response: We thank the referee for the suggestion to distinguish between the dissipative and superconducting regions in Fig. E4. We agree that this improves the clarity of the figure. We have revised the figure accordingly by adding purple shading to the region where zero resistance is observed in the transport data. A sentence describing this has been included in the figure caption.



Extended Data Fig. 4. Tank-circuit measurements of the superconducting states. (A) Frequency shift versus magnetic field at several temperatures in low fields. For clarity, curves are vertically offset by 2 kHz. Although the midpoint of the resistive transition is at $H \approx 1$ T (see Fig. 1), a diamagnetic shift associated with screening currents (~ 5 kHz) appears at ~ 0.2 T. The broad peak at $H \approx 2$ T and $T = 1.3$ K roughly coincides with the spin-flop transition out of the antiferromagnetic phase of NdGaO₃ (see Extended Data Fig. 2). The inversion of this feature at $T = 0.5$ K coincides with a dip in resistivity at $H = 2$ T for 0.6 K $\leq T \leq 2$ K (see Fig. 1), suggesting some interplay between Eu²⁺ moments in the nickelate film and the NdGaO₃ magnetism over this temperature range. (B) High-field measurements with (solid) and without (dashed) the sample at different angles ϑ . Curves are offset by 10 kHz for clarity. While the low-field diamagnetic shift is visible directly in the raw data, at high fields it becomes apparent only after subtracting a background obtained from measurements without the sample **at particular temperatures and angles**. (C) Frequency shift due solely to the sample, obtained by background subtraction, at several angles ϑ . Curves are vertically offset by 2 kHz; horizontal dash-dot lines indicate baselines. At high fields (≥ 10 T), the frequency shift for $\vartheta = 0^\circ$ remains flat. In contrast, for larger angles, where resistivity vanishes at $T = 0.5$ K (see Fig. 1 and Extended Data Fig. 3), a clear field-induced diamagnetic shift associated with the high-field superconducting state is

observed. Arrows indicate the approximate midpoints of the field ranges over which resistivity vanishes. The purple shaded regions in A and C represent the SC state observed from the transport data.

- The authors state that the spin-flop transition of Nd results in “secondary features” in the low temperature transport. Can they be clearer on what is meant by that?

Response: We apologize for the lack of clarity in the rebuttal letter regarding the connection between the field-dependent resistance feature and the spin-flop transition of Nd. Specifically, we refer to a small dip in resistivity near $H \approx 2$ T for $T < 2$ K, which can be attributed to the spin-flop transition of Nd³⁺ moment.

In the manuscript on page 3, we state:

“Salient features of both samples include a sharp transition from the superconducting to the resistive state at $H \approx 1$ T, followed by a small dip in resistivity near $H \approx 2$ T for $T < 2$ K. This dip coincides with the spin-flop transition of the antiferromagnetically ordered Nd moments in the NdGaO₃ substrate into a polarized state.”

We believe this description is sufficiently clear.

- Why does it appear that Fig R8/2 B does not follow the model?

Response: We agree with the referee that the model does not fully capture the ultra-low-temperature behavior of the high-field superconducting state. We have added a sentence addressing this discrepancy on page 18 of the previous version of the manuscript.

“The steep increase in H_{c2} below 0.6 K remains unexplained using simplified WHH model and, therefore, needs elaborate theory efforts that is beyond the scope of this work. Notably, low-temperature non-saturating H_{c2} has also been reported in other nickelates and cuprates [45, 46] and various mechanisms, including an interplay between superconductivity and a density wave [47] and the existence of multiple bands [48], are proposed to account for this phenomenon.”

My opinions on the responses to Reviewer 2

1. The authors have responded satisfactorily.
2. The authors have responded satisfactorily.
3. The authors have responded satisfactorily.

Response: We thank Reviewer #1 again for providing his feedback on our response to Reviewer #2 comments. We are pleased that most of our responses addressed Reviewer #2's concern.

Below we clarify on the couple of words that were not clear in the rebuttal letter.

4. The authors have decided that a quantitative analysis is beyond the scope of the work. They say that they have a “data-driven” approach, which usually means a high level of quantification, and this is not the case. They also say that they will not conduct DFT calculations, but this is not what reviewer 2 was suggesting. My understanding is that reviewer 2 merely suggested estimating interaction energies from the measurement.

Response: We appreciate the reviewer’s clarification and recognize that we may have misunderstood the original comment regarding quantitative estimation of the exchange interaction. In our analysis, we have already estimated an effective exchange field by fitting the experimental data using a modified WHH model. Our earlier response interpreted the comment as a request for a specific microscopic model like DFT for the exchange interaction in this system.

We now understand that the reviewer’s suggestion was to estimate relevant interaction energy directly from the experimental data. While our current approach, i.e. fitting the Hc₂-T with WHH model provides an effective exchange field, a more direct quantification of the exchange interaction from measurements remains challenging within the scope of this work.

We also note that experimental techniques such as electron spin resonance can provide a more direct measure of exchange interactions; however, such measurements are beyond the scope of the present study and represent a potential direction for future work.

5. The authors have responded satisfactorily. I do have some questions: What is meant by suggesting that the composition modifies “exchange pathways”? I doubt that the type of exchange interaction is strongly modified in the narrow range of compositional space reported here. Also, they mention the “topology” of the reentrant regime. How is the topology relevant?

Response: We apologize for the lack of clarity regarding the terms “exchange pathways” and “topology.”

By “exchange pathways,” we refer specifically to the $4f \leftrightarrow 5d \leftrightarrow 3d\text{Ni}$ exchange path, which links the transition-metal 3d conduction electrons with the rare-earth 4f magnetic moments. We agree that small changes in composition, as in the samples studied here, are unlikely to significantly alter the type of exchange interaction. However, such variations may affect the strength of the interaction by modifying the energy separation between the rare-earth 5d and Ni 3d orbitals.

We also acknowledge that “topology” is not the appropriate term in this context. What we intended to convey is that T_c (determined by composition and thickness) plays a dominant role in governing both the emergence and the field range of the re-entrant phase.

We noticed that we didn’t use these terms in the main manuscript, so we don’t have to make any change in the manuscript text.

6. The authors have responded satisfactorily. I would also add that there is precedent for magnetic moments in the superconducting phase of infinite layer nickelates. Reviewer 2 refers to work claiming ferromagnetic order. I understand they refer to the work from Hong Kong. In my opinion, that preprint is mistaken in interpreting magnetic order. They do not have sufficient evidence and my understanding is that those authors are revising their claims. So I believe reviewer 2 was slightly off-base when making this comment.

Response: We thank Reviewer #1 for noting that Reviewer #2’s comment may have been influenced by the Hong Kong work.

7. The authors have responded satisfactorily.

Response: We are pleased that our response addressed Reviewer #2’s concern.

Dear Editor and Reviewer,

We sincerely thank you for the in-principle acceptance of the manuscript. The manuscript has been revised according to the instructions and the editorial requests.

Below is our response to the reviewer's comment. Reviewer's comments are in black; our responses are in blue. Minor changes made in the manuscript are highlighted in red here as well as in the manuscript text.

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

The authors have responded satisfactorily to my comments.

Response: We greatly appreciate reviewer #1's help and the comments for improving the manuscript.