nature portfolio

Peer Review File

Charge Stripe Manipulation of Superconducting Pairing Symmetry Transition

Corresponding Author: Professor Bing Huang

This manuscript has been previously reviewed at another journal. This document only contains reviewer comments and rebuttals for versions considered at Nature Communications.

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)
I have completed my review of the revised manuscript "Charge stripe manipulation of superconducting pairing symmetry transition" and the correspondence between myself, the other reviewers, and the authors. In response to the first round of review, the authors have performed several additional simulations of their model, including DQMC simulations for smaller values of U and DMRG simulations on small cylinders. They have also made extensive efforts to address my comments and those from the other referees. Their extensive efforts to improve the manuscript are commendable. However, while these additions strengthen the paper, I still have reservations about recommending publication in its current form, as elaborated below.

1) The original DQMC simulations were restricted to very high temperatures, which prevented the authors from drawing definite conclusions about low-temperature superconducting instabilities. In response to my concerns, the authors have performed additional DQMC simulations for lower U values, allowing them to access lower temperatures. They have also performed zero-temperature DMRG calculations on narrow cylinders. Both sets of additional calculations raise more questions. First, the new DQMC calculations shown in Fig. 3 show rapidly rising correlations in the d-wave pairing channel, which signals the formation of a d-wave superconductor. I accept this. However, the corresponding extended s-wave shows slow growth that one typically sees from trivial thermal effects (i.e., sharpening of the spectral function). The authors state that this growth "could diverge in the thermodynamic limit at sufficiently low temperatures," but this statement is speculation. The corresponding DMRG calculations also have issues. To assess whether the system has dominant superconducting correlations, you must examine the decay of the superconducting, charge, and spin correlations with distance to determine which dominates. Such calculations require very long cylinders, much longer than those considered in this work, and the authors do not examine these competing channels at all. It is thus unclear to me whether the pairing correlations reported represent a superconducting order in the corresponding 2D system.

Related to this point, the authors assert in their reply to me and the other referees that correlation effects can already be seen at T = 0.2t in the Hubbard model. I don't dispute this; one can see the high-energy impact of U (i.e., the opening of the Mott gap) at these temperatures. My point was that extrapolating high-temperature superconducting correlations obtained at this temperature to low-temperature often leads to incorrect conclusions about the actual superconducting instability. This has been demonstrated in DCA studies of the Hubbard model where one can access the superconducting instabilities.

2) I expressed concern over the strength of the periodic potential V0, which is easily the largest energy scale in the problem. The authors point to DMFT calculations about the energy differences between different Ni valence states to justify this choice. However, those energies are related to the intra-atomic interactions and would be present uniformly throughout the system; they don't tell you anything about the inhomogeneous potential in the system. The authors have yet to address this comment to my satisfaction.

3) The other referees also raised the issue of the stripe order being imposed rather than forming spontaneously. For example, the third referee felt that this missed the chance to determine whether the pairing in the nickelates was mediated by spin or charge fluctuations. The authors have attempted to address this point in the context of their model and linking pairing

the former. However, their reply misses what I see as the essential point of this objection: state-of-the-art QMC and DMRG calculations show that the stripe order competes with superconductivity when it forms spontaneously in the homogeneous Hubbard model. The authors are considering a case where some external beyond-Hubbard model potential is imposed on the system that induces charge order. These calculations are interesting, but their relevance to the nickelates remains speculative. I agree that the charge order in these materials may be stabilized by an external interaction but I strongly doubt it is as strong as the one being used by the authors if it is there. And if it's not, it might compete with superconductivity rather than stabalize an s-wave pairing.

4) The other referees and I raised concerns over the use of the single band Hubbard model for the Nickelates. The authors performed additional calculations for a 4-orbital model but neglected the correlations on the second Ni orbital and the interorbital Hubbard and Hund's interactions between the Ni orbitals. Unless I am mistaken, author's model in Eq S2 thus misses important interactions that will strongly influence the spin state of the system. Moreover, recent RIXS work has provided compelling evidence that the nickelates fall closer to the charge transfer insulating regime ($\Delta \approx U$) than previously thought [Y. Shen et al., PRX 13, 011021 (2023); PRX 12, 011055 (2022)]. Therefore, including the oxygen orbitals is necessary and the authors have not adequately justified using a single-band Hubbard model. Their response seems to be more focused on the inclusion of the additional Ni orbitals however they also ignored the potential 3D structure of the system. For example, including the 3dz2-r2 orbital could enhance the interlayer hopping. This issue was raised by the third referee but not really addressed by the authors in their reply. I will also add that just because ARPES sees a single band doesn't mean a multiorbital description is unnecessary to describe the interacting system.

Finally, I would also like to comment that the authors have address comments 3 and 4 of the third referee to my satisfaction.

Overall, this paper would be better conceived as a study of what a strong, imposed charge order does to pairing correlations in the Hubbard model, with the speculative links to the nickelates and cuprates removed. I believe this sentiment is echoed by the third referee, who saw value in these calculations but felt they "did not address the fundamental mechanism of the nickelates."

Reviewer #2

(Remarks to the Author)
I think the authors made substantial improvements over the version I reviewed in the past, both in clarifying the physical importance of the study and in strengthening the methodological approach. I am pleasantly surprised by finding DMRG ground state results together with unbiased determinantal QMC results. I am very satisfied with how the authors addressed the responses of the referees, including mine.
I feel confident recommending this version for publication.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
I have read the author's reply to my second report and their revised manuscript. While I remain unconvinced of the applicability of these results for the nickelates, the revised introduction largely side-steps the material specific concerns I have raised. I am also quite happy to see the new supporting CPQMC calculations at zero temperature. Therefore, I can now recommend publication of the paper in its current form.

I also want to thank the authors for all of their hard work and openness to my comments throughout the process.

Response Letter

We thank the Referee #1 for his/her second-round comments and suggestions on our reply and manuscript. We have carefully considered all the feedback and have made responses accordingly. The changes made to the manuscript are marked in red.

We also sincerely thank the Referee #2 for agreeing to publish our manuscript without any changes.

Response for the Referee #1:

Comment 1: I have completed my review of the revised manuscript "Charge stripe manipulation of superconducting pairing symmetry transition" and the correspondence between myself, the other reviewers, and the authors. In response to the first round of review, the authors have performed several additional simulations of their model, including DQMC simulations for smaller values of U and DMRG simulations on small cylinders. They have also made extensive efforts to address my comments and those from the other referees. Their extensive efforts to improve the manuscript are commendable. However, while these additions strengthen the paper, I still have reservations about recommending publication in its current form, as elaborated below.

Response 1: We greatly appreciate the referee's recognition of our efforts in revising the manuscript. We also thank the referee for his/her additional comments/suggestions, which have helped us to improve the quality of our manuscript. We have addressed the referee's comments, along with major changes made to our manuscript.

Comment 2: 1) The original DQMC simulations were restricted to very high temperatures, which prevented the authors from drawing definite conclusions about low-temperature superconducting instabilities. In response to my concerns, the authors have performed additional DQMC simulations for lower U values, allowing

them to access lower temperatures. They have also performed zero-temperature DMRG calculations on narrow cylinders. Both sets of additional calculations raise more questions. First, the new DQMC calculations shown in Fig.3 show rapidly rising correlations in the *d*-wave pairing channel, which signals the formation of a *d*-wave superconductor. I accept this. However, the corresponding extended *s*-wave shows slow growth that one typically sees from trivial thermal effects (i.e., sharpening of the spectral function). The authors state that this growth "could diverge in the thermodynamic limit at sufficiently low temperatures," but this statement is speculation. The corresponding DMRG calculations also have issues. To assess whether the system has dominant superconducting correlations, you must examine the decay of the superconducting, charge, and spin correlations with distance to determine which dominates. Such calculations require very long cylinders, much longer than those considered in this work and the authors do not examine these competing channels at all. It is thus unclear to me whether the pairing correlations reported represent a superconducting order in the corresponding 2D system.

Response 2: Thank the referee for these comments.

(1) To confirm our statement that "extended *s*-wave could diverge in the thermodynamic limit at sufficiently low temperatures", we have calculated the effective pairing interaction $\overline{P}_\alpha$ using DQMC on a torus of 12×12 lattice at U/t =3, reaching a lower temperature $T=t/16$. Calculating a single data point at $T=t/16$ required *three months of code running*, since much longer measurements are necessary to maintain data quality with $\langle sign \rangle \approx 1$. In Fig. R1, when $T \leq t/12$, we can see that dominant $\overline{P}_\alpha$ for the extended *s*-wave rises more sharply with the decrease of temperature, implying a tendency to diverge in lower temperature. Besides, our calculations indicate that this dominant *s*-wave pairing can be further enhanced by a larger coulomb repulsion U (see Fig. 3(d) in the main text). So, the dominant $\overline{P}_\alpha$ for the *s*-wave pairing symmetry could indeed diverge in the thermodynamic limit at

sufficiently low temperatures.

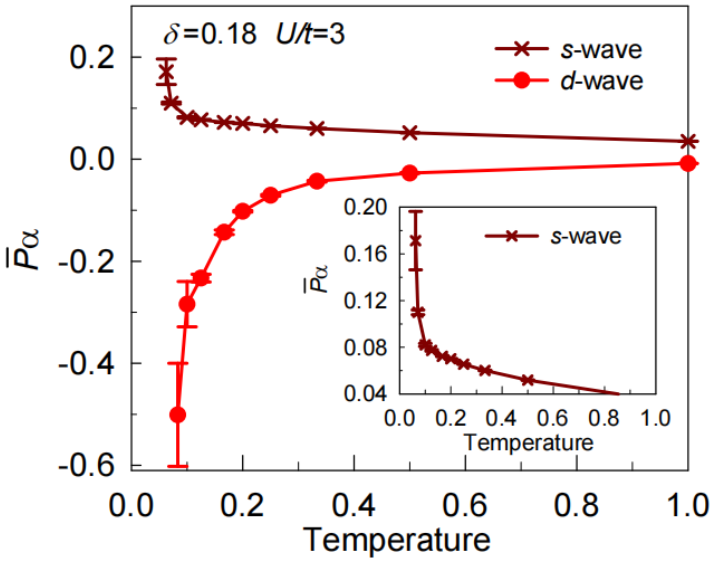

Fig. R1: DQMC-calculated effective pairing interaction $\overline{P}_\alpha$ as a function of temperature for different pairing symmetries at U/t=3, V_0=6 and L=12 under stripe period $\mathcal{P} = 3$ with $\delta = 0.18$. Inset: $\overline{P}_\alpha$ for the s-wave pairing symmetry versus temperature.

(2) We thank the referee for raising the point that "To assess whether the system has dominant superconducting correlations, we must examine the decay of the superconducting, charge, and spin correlations with distance to determine which dominates". Considering the recent numerical studies on the Hubbard model, the above point raised by the referee may be based on the pioneering works of [Jiang *et al*., Science **365**, 1424 (2019)] and [Xu *et al*. Science **384**, 637(2024)]. In these works, the authors computed the decay of the superconducting, charge, and spin correlations with distance to determine which dominates in the ground state of the doped Hubbard model. However, this treatment may be not appropriate for our extended Hubbard model, which contains an additional stripe potential. The charge stripe, with its tunable oscillation strength, is introduced *via* the external stripe potential V_0. Thus, detecting the decay of some correlation functions across the stripe might face some issues, e.g., charge correlations.

To address the referee's concerns about competing orders, we have further employed the constrained-path quantum Monte Carlo (CPQMC) and DMRG to demonstrate that the system may exhibit long-range superconducting correlations for the s-wave pairing in the region $\delta = 0.18$, U/t=4, V_0=6 and L=12 under stripe period $\mathcal{P} = 3$.

To assess whether the system has dominant superconducting correlations, we examine the decay of the superconducting correlations with distance using the CPQMC method (see the computational details in the Method section of our manuscript). The calculated results are summarized in Fig. R2.

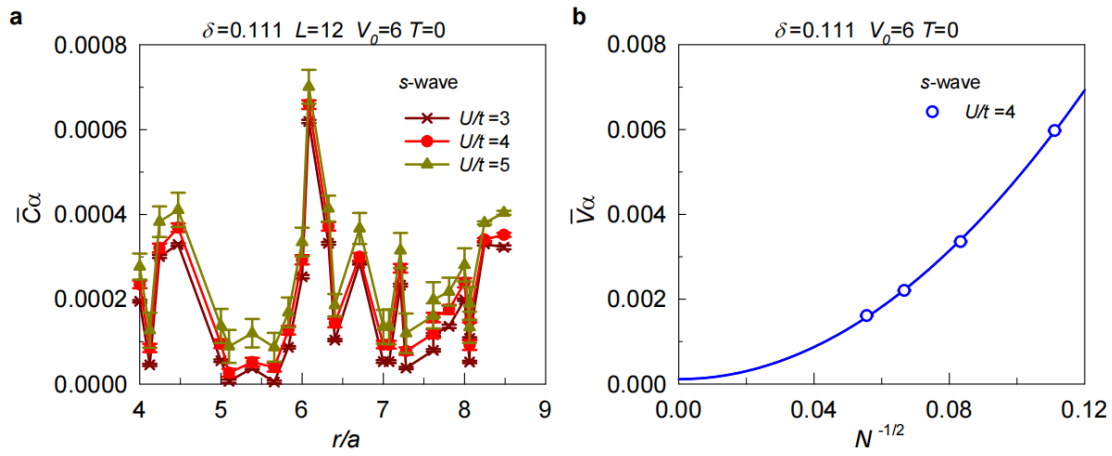

Fig. R2: (a) CPQMC-calculated vertex contributions of *s*-wave pairing symmetry C_α as a function of distance r/a at T=0, V_0 =6 and L=16 under stripe period $\mathcal{P} = 3$ with δ=0.111 for different values of U/t. a represents the lattice spacing in the square lattice. (b) Scaling analysis is based on the averaged long-range C_α typically at U/t=4. The blue solid line represents a fit using a third-order polynomial in $\sqrt{1/N}$.

As shown in Fig. R2a, the long-range superconducting vertex contribution calculated by CPQMC is defined by

$$\bar{C}_\alpha(r) = \frac{1}{NN_r}\sum_{\boldsymbol{i}}\sum_{|\boldsymbol{i}-\boldsymbol{j}|=r}\langle\Delta_\alpha^\dagger(\boldsymbol{i},0)\Delta_\alpha(\boldsymbol{j},0)\rangle,$$

where N_r is the number of sites that are located at a distance r from site $\boldsymbol{i}$. As shown in Fig. R2b, for scaling analysis, we define $\bar{V}_\alpha = \frac{1}{\sqrt{N'}}\sum_{r/a>4}\bar{C}_\alpha(r)$, where N' is the number of sites situated at a given distance $r > 4$. We extrapolate the value of $\bar{V}_\alpha$ to the thermodynamic limit and find that it retains a small but finite value. This suggests the possible existence of long-range superconducting order for the *s*-wave pairing.

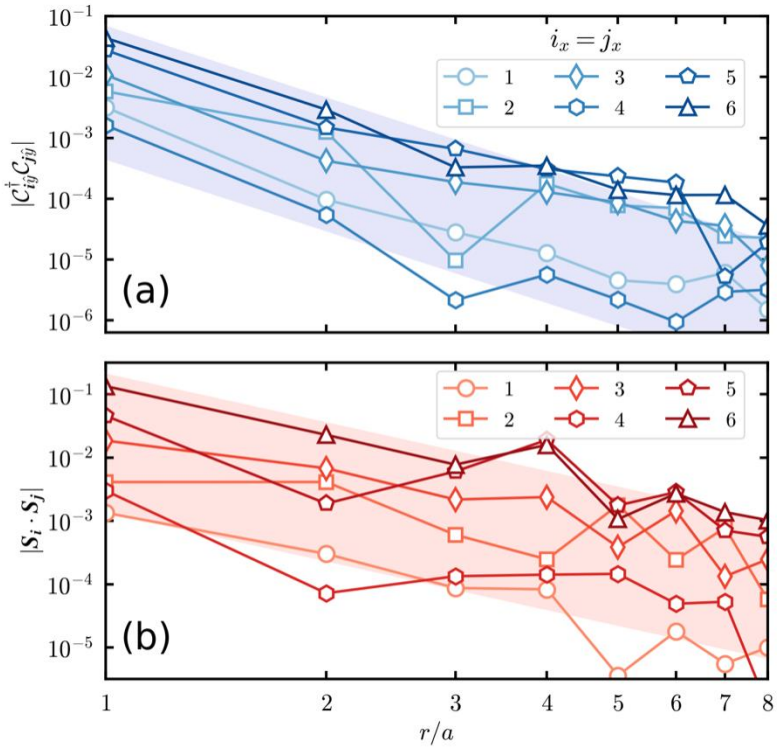

Fig. R3: DMRG-calculated (a) Cooper pair-pair correlation $\left|\mathcal{C}^\dagger_{i\hat{y}}\mathcal{C}_{j\hat{y}}\right|$ and (b) spin-spin correlation $\left|\boldsymbol{S}_{\boldsymbol{i}} \cdot \boldsymbol{S}_{\boldsymbol{j}}\right|$ as a function of distance $r = |\boldsymbol{i} - \boldsymbol{j}|$ in the s-wave state at $\delta = 0.111$, $V_0/t = U/t = 8$ and $P = 3$ on a $L_x \times L_y = 6 \times 18$ lattice. Here, we choose different values of $i_x = j_x = 1, 2, 3, 4, 5,$ and 6. The color-shaded regions indicate the linear tendency of the correlation functions in the double-logarithmic plot. 10240 SU(2) bases are used in DMRG, which is equivalent to 30000 U(1) bases roughly.

We have also paid much afford to directly examine the decaying behavior of correlations with distance, with the aim of determining which, if any, dominates in the s-wave state. In Fig. R3, we illustrate the y-axis of a $L_x \times L_y = 6 \times 18$ lattice as an example. As shown in Fig. R3a, the superconducting correlation $\left|\mathcal{C}^\dagger_{i\hat{y}}\mathcal{C}_{j\hat{y}}\right|$, which is also known as the Cooper pair-pair correlation, displays a quasi-long-range behavior that is dominated by a power-law when different choices of $i_x = j_x$ are made. Additionally, we find that the superconducting correlations are identical for bonds situated along the circumference of the cylinder in comparison to those along the long side, aligning with the anticipated characteristics of the s-wave pairing. As shown in

Fig. R3b, the spin-spin correlations $|\boldsymbol{S_i} \cdot \boldsymbol{S_j}|$ exhibits quasi-long-ranged behavior, corresponding to a gapless spectrum in the spin sector, standing in sharp contrast to the *d*-wave stated in the recent study [Jiang *et al*., Science **365**, 1424 (2019)]. This finding indicates that the antiferromagnetic spin order and the superconducting order coexist.

We would like to emphasize that in the main focus of our current manuscript is to determine the role of charge stripe affecting on the modulation of pairing symmetry. Our current numerical simulations are sufficiently robust to identify the pairing symmetry.

While we have made great efforts to conduct more simulations in response to the referee's comment, we hope the referee could recognize our consistent conclusion, obtained through the combination of multiple numerical methods, despite that each method has its own limitation. We have added Figs. R1-R3 in the Supplemental Material, now labeled as Figs. S4b, S9 and S10, respectively.

Comment 3: Related to this point, the authors assert in their reply to me and the other referees that correlation effects can already be seen at T=0.2t in the Hubbard model. I don't dispute this; one can see the high-energy impact of U (i.e., the opening of the Mott gap) at these temperatures. My point was that extrapolating high-temperature superconducting correlations obtained at this temperature to low-temperature often leads to incorrect conclusions about the actual superconducting instability. This has been demonstrated in DCA studies of the Hubbard model where one can access the superconducting instabilities.

Response 3: We agree with the referee that there is a temperature dependence of the pair-field susceptibility, which may change quantitative details of the phase diagram. In the last manuscript, we identified the presence of a *d*-*s* wave transition based on

DQMC data at low temperatures, and further confirmed it through DMRG simulations at zero temperature for several typical parameter sets. By recently conducting more simulations with DQMC at even lower temperatures $T \leq t/12$, as well as CPQMC and DMRG at zero temperature, detailed in this response and the Supplemental Material, we have verified the long-range superconducting order for both the s-wave and *d*-wave pairings. This rules out the possibility of a qualitative change in pairing symmetry as the temperature decreases. So, we think our conclusion is self-consistent and robust.

Comment 4: I expressed concern over the strength of the periodic potential V_0, which is easily the largest energy scale in the problem. The authors point to DMFT calculations about the energy differences between different Ni valence states to justify this choice. However, those energies are related to the intra-atomic interactions and would be present uniformly throughout the system; they don't tell you anything about the inhomogeneous potential in the system. The authors have yet to address this comment to my satisfaction.

Response 4:

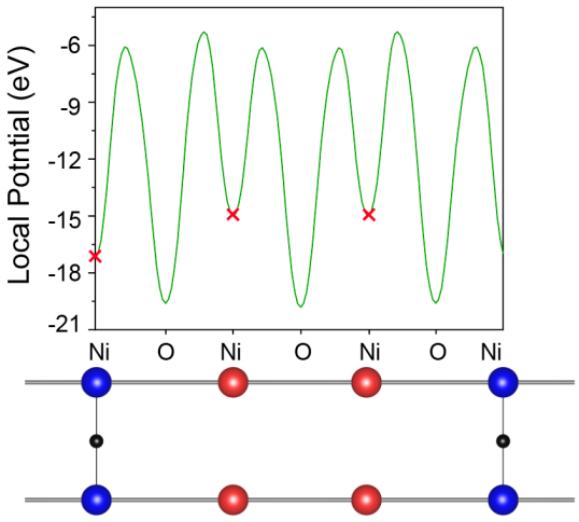

Fig. R4: DFT-calculated local electrostatic potential distribution of the nickelate lattice shows a periodic charge stripe with a period of P=3. The charge stripe is induced by the introduction of H atoms. Blue (red) circles label the sites with (without) the inclusion of H atoms (black circles),

which introduce different Ni valance-states to simulate the periodic charge stripe. Red crosses label the electrostatic potential of Ni ions.

We thank the Referee for this critical question. Currently, the origin of charge orders observed in nickelates remains largely unclear. Some researchers argue that it is an intrinsic phenomenon while others do not. So, there is no widely-accepted atomic structure model for the charge stripe in infinite-layer nickelates. To evaluate the possible charge-stripe amplitude (V_0) induced by the variable valence of the Ni charge-state, we constructed a 3×1×1 superlattice with the incorporation of one hydrogen atom, as illustrated in the lower panel of Fig. R4. Although the introduction of hydrogen may not represent the experimentally observed charge-stripe structure in nickelates, it can effectively lead to the formation of a Ni^{2+}-Ni^{1+}-Ni^{1+} stripe state [Phys. Rev. Lett. **124**, 166402 (2020), Crystals **12**, 656 (2022), Phys. Rev. B **107**, 165116 (2023)]. The DFT-calculated local electrostatic potential in real space is depicted in the upper panel of Fig. R4. Notably, the potential for Ni^{2+} (−17.23 eV) is significantly lower than that for Ni^{1+} (−15.00 eV). Therefore, the charge-stripe amplitude V_0, caused by the variable valence of the Ni charge state, can be roughly estimated as $|(VNi^{2+}-VNi^{1+})/t| = |(-17.23+15.00)/0.37|=6.03$. This value is comparable to the value of ~5 used in our manuscript for realizing *d-s* wave transition.

We also want to mention to the referee that according to your last suggestion, we have made a major change in the main text, *i.e.*, we have removed the arguments related to the real nickelate system and instead focused on the discussion on the interplay between the strong charge stripes and the superconducting pairing symmetries in an extended Hubbard model system. Therefore, the concerns raised in this comment have been resolved.

Comment 5: The other referees also raised the issue of the stripe order being imposed rather than forming spontaneously. For example, the third referee felt that this missed

the chance to determine whether the pairing in the nickelates was mediated by spin or charge fluctuations. The authors have attempted to address this point in the context of their model and linking pairing the former. However, their reply misses what I see as the essential point of this objection: state-of-the-art QMC and DMRG calculations show that the stripe order competes with superconductivity when it forms spontaneously in the homogeneous Hubbard model. The authors are considering a case where some external beyond-Hubbard model potential is imposed on the system that induces charge order. These calculations are interesting, but their relevance to the nickelates remains speculative. I agree that the charge order in these materials may be stabilized by an external interaction but I strongly doubt it is as strong as the one being used by the authors if it is there. And if it's not, it might compete with superconductivity rather than stabalize an *s*-wave pairing.

Response 5: We thank the referee for this critical comment. As discussed in **Response 4**, a strong charge stripe like what we adopted in this study may indeed exist in nickelates, induced by external impurities. We also agree with the referee's suggestion to remove the link between our current model and real nickelate systems, which has been done in our revised manuscript. Therefore, the concerns raised in this comment have been resolved.

Comment 6: 4) The other referees and I raised concerns over the use of the single band Hubbard model for the Nickelates. The authors performed additional calculations for a 4-orbital model but neglected the correlations on the second Ni orbital and the interorbital Hubbard and Hund's interactions between the Ni orbitals. Unless I am mistaken, author's model in Eq S2 thus misses important interactions that will strongly influence the spin state of the system. Moreover, recent RIXS work has provided compelling evidence that the nickelates fall closer to the charge transfer insulating regime ($\Delta \approx U$) than previously thought [Y. Shen et al., PRX 13, 011021 (2023); PRX 12, 011055 (2022)]. Therefore, including the oxygen orbitals is necessary and the authors have not adequately justified using a single-band

Hubbard model. Their response seems to be more focused on the inclusion of the additional Ni orbitals however they also ignored the potential 3D structure of the system. For example, including the 3dz2-r2 orbital could enhance the interlayer hopping. This issue was raised by the third referee but not really addressed by the authors in their reply. I will also add that just because ARPES sees a single band doesn't mean a multiorbital description is unnecessary to describe the interacting system.

Response 6: We thank the Referee for the comment.

In the four-band model used in Eq. S2, we omit the second Ni orbital and include the other two Nd orbitals (Nd $d_{xy}/d_{3z^2-r^2}$). This model, proposed in Communications Physics **3**, 84 (2020), highlights the stronger role of itinerant *s* electrons compared to Nd orbitals, so we can see that the hybridization involving Nd orbitals is weak. Therefore, we did not include the interaction of Nd orbitals, and interactions between Nd and Ni orbitals.

Nickelates have a broad range of compounds. The references [PRX **13**, 011021 (2023), PRX **12**, 011055 (2022)] cited by the referee discuss $La_4Ni_3O_8$, which does not exhibit superconductivity and is not the focus of this paper. In this manuscript, we mainly concentrate on the infinite-layer nickelates RNiO$_2$ (R=Nd, La, Pr), which are superconductors.

Several important experiments suggest that superconducting infinite-layer nickelates behave as a quasi-2D electronic structure. (i) The experiment [Nature **615**, 50(2023)] demonstrates that nickelates are more 2D-like based on the advanced RIXS data. (ii) The experiment [Nature Physics **18**, 869 (2022)] reveals that the rare-earth Fermi-surface pocket rapidly shrinks and eventually disappears with increasing Sr doping, which also supports that nickelates become increasingly like a 2D system under hole-doping. (iii) The experiment [Nature Communications **14**, 7155 (2023)] studied the polar and azimuthal angular-dependent transport behaviors of the

superconducting state in infinite-layer nickelate, supporting the quasi-2D nature of the superconductivity in infinite-layer nickelates. (iiii) Recently, ARPES measurements confirm that the overall band structures are quite similar between RNiO$_2$ and cuprates, exhibiting a dominated $d_{x^2-y^2}$-band feature in a large BZ region [National Science Review **11**, nwae194 (2024); arXiv: 2403.07344(2024)]. So, the infinite-layer nickelates tends to be more two-dimensional after hole-doping, based on the current experimental evidences.

Comment 7: Finally, I would also like to comment that the authors have address comments 3 and 4 of the third referee to my satisfaction.

Overall, this paper would be better conceived as a study of what a strong, imposed charge order does to pairing correlations in the Hubbard model, with the speculative links to the nickelates and cuprates removed. I believe this sentiment is echoed by the third referee, who saw value in these calculations but felt they "did not address the fundamental mechanism of the nickelates."

Response 7: We thank the Referee for this good suggestion to improve the quality of our manuscript. We fully agree with your suggestion. We have made major revisions to the main text. We have removed the arguments related to the real nickelate system and instead focused on the discussion on the interplay between the strong charge stripes and the superconducting pairing symmetries in an extended Hubbard model system.

We wish the referee will be satisfied with our big efforts in replying to the referee's comments and suggestions, along with additional calculations. We believe that our manuscript is now ready for being accepted in its current form.

Response for the Referee #2:

Comment 1: I think the authors made substantial improvements over the version I reviewed in the past, both in clarifying the physical importance of the study and in strengthening the methodological approach. I am pleasantly surprised by finding DMRG ground state results together with unbiased determinantal QMC results. I am very satisfied with how the authors addressed the responses of the referees, including mine. I feel confident recommending this version for publication.

Response 1: We greatly appreciate the referee's agreement with the acceptance of our manuscript.