nature portfolio

Peer Review File

Evolution from a charge-ordered insulator to a high-temperature superconductor in Bi2Sr2(Ca,Dy)Cu2O8+δ

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

In the manuscript by Zou et al., the temperature dependence of charge order (CO), phonons, and low energy excitations in lightly doped Dy-Bi2212 (p=0.04, 0.06) were measured using high-resolution RIXS and STM. The authors report the observation of CO and phonon modes, which were extracted from RIXS data taken with an instrumental energy resolution of 36 meV. The dispersions of phonons are not found to soften near the CO position. Notably, the manuscript provides experimental evidence for the existence of CO in lightly doped cuprate and pinpoints the origin of CO and its competition with SC.

The origin of CO and its relevance to the superconductivity in cuprates are important issues of current interest. High-resolution RIXS has the potential to unravel new insights into these issues. The experimental data are interesting and up-to-date. However, analyses and discussions on photon dispersion are not fully convincing in understanding electron-phonon coupling (EPC), as commented below. This leads the reviewer to conclude that the present manuscript does not satisfy the condition of a high-profile paper required by Nat. Commun. Therefore, I don't recommend the publication of the present manuscript unless the following major issues are satisfactorily addressed:

(1) Regarding the phonon dispersion and softening, my biggest concern is that the energy resolution of the present work is not good enough to extract a convincing conclusion for low-energy phonons. Several modes of low-energy phonons, including acoustic, A1g and B1g buckling modes, exist in hole-doped cuprates. Their anomalies due to the formation of CO have been observed by inelastic x-ray scattering (e.g., La Tacon et al., Nat. Phys. 10, 52. (2014)) and high-resolution RIXS (e.g., Ref. [32]). The phonon softening and EPC exist in under-doped YBCO and optimally doped LSCO. Interestingly, the present manuscript shows no evidence of phonon anomalies for the wave vector across CO in lightly doped Bi2212. In my viewpoint, the data analysis with only two phonon components to fit the RIXS spectra broadened by an instrument of energy resolution 36 meV does not correctly reveal the phonon softening and EPC. Using the present RIXS data to rule out the dispersion of low-energy phonons buried in the tail of the "broad" elastic peak is not convincing. The

authors also commented similarly on previous works, Ref. [6, 20, 21, 31]. The conclusion of the present manuscript about phonon anomalies requires further support of RIXS data taken with an improved energy resolution. Perhaps it is very challenging to improve the energy resolution of RIXS significantly. Alternatively, the authors are suggested to substantially modify the discussions on the present data to address the issue of phonon anomalies correctly.

(2) The paper reports CO of wave vectors ~ 0.31 r.l.u. and concludes that the CO is incommensurate with the lattice structure. In combining with dispersionless BB phonon, the authors rule out the possibility of the formation of charge-density waves (CDW) induced by lattice distortion. However, observing a non-fractional modulation vector can result from discommensurations or a truly incommensurate order. The discommensurations of CDW have been discussed in many papers, for example, Masros et al., Proc. Natl. Acad. Sci. 113, 12661 (2016), Vinograd et al., Nat. Commun. 12, 3274 (2021), and Ref [32]. The authors need to discuss whether the observed CO is truly incommensurate or appears to be incommensurate due to discommensurations.

In addition, there is one mistake in the caption of Fig. 1b, which uses the phase diagram to show the RIXS measurement strategy. However, the caption incorrectly says, " The orange and purple circles represent our RIXS and STM data, respectively. " In fact, Fig 1b plots neither RIXS nor STM data.

Reviewer #2 (Remarks to the Author):

Zou et al. present a combined resonant inelastic X-ray scattering (RIXS) and scanning tunneling microscopy (STM) data on the cuprate superconductor $Bi2Sr2Ca0.6Dy0.4Cu2O8+\delta$. The aim of their study is to shed light on mechanism of superconductivity close to the insulating regime.

The key results of their study are:

i) Charge order persists to even lower hole concentrations ($p = 0.04$) than previously known.

ii) Charge order is incommensurate and is thus makes a lattice induced distortion unlikely.

iii) Phonon softening near the charge order wave vector is absent

iv) There is no interplay between spin excitations and charge order

v) The intensity of the bond buckling phonon is strongly enhanced in the superconducting state.

From this the authors somewhat conclude that both the electron-phonon coupling and charge order work together to support superconductivity. In particular, they speculate that the onset of charge order at temperatures higher than the superconducting critical temperature, is a sign of Cooper pairs being pre-formed. In my opinion, this conclusion is not borne out by their results.

Although, I appreciate that high-quality of the data, and also the effort to study the same sample with two complementary methods, and also think that the data is quite interesting, I can currently not support publication of the article in its current form in Nature Communications.

I also would like to highlight that there are many English mistakes, use of unsuitable wording, suse of colloquial language, and so. This poses a problem in following what the authors are attempting to say, so it should be fixed.

In addition to the language of the article often not being clear, the conclusions are not presented clearly. I would suggest that the authors carefully go through the article, and both remove English language mistakes, and attempt to make introduction and discussion more concise and less convoluted. What are the main questions being answered and why? What it the impact of this on the $Bi2Sr2Ca0.6Dy0.4Cu2O8+\delta$, the family of cuprates and unconventional superconductivity broadly.

In addition, I would recommend that the authors also look at the following comments:

- Line 15: “How Cooper pairs from and condense have been the main challenges …”: This should be singular. “…has been the main challenge”.

- Line 19: “…the Fermi surface is lacking…”. I assume the authors mean missing.

- Line 23: “We show that an incipient charge order has existed…”. This should be: “We show that incipient charge exists…”.

- Line 34: “evidences”. Evidence is to the best of my knowledge always singular.

- Line 36: “…a different story proposes…”. This is colloquial language, and should be changed.

- Fig. 1b. Why not add the onset of charge order derived from this work? Please add

- Fig. 1b. The symbols are not pentagrams, but stars.

- Fig. 1b. The superconducting region is not labelled, even though it is the most important phenomena the article describes.

- Fig. 2a/b/c. The white dashed lines are barely visible.

- Fig. 2. Caption. “fittings” have nothing to do a fit. I believe you would like to say “fits”. Also, the caption does not introduce what a “PG map” is. This is not a standard expression. It is explained in the main text, but not in the caption. The caption should at least refer to the main text.

- Fig. 5b. Please plot the lines for the fit beyond the recorded data so that the peak shape can be judge better.

- Fig 5 c,d have no numbers on the y-axis. Even for intensity in a.u. it is essential were the zero is to understand how strong the effect is. Please add.

- Fig. 6. It is not clear if how the dI/dV curves in panel c connect to the RIXS data in panel b. Please explain better what you mean.

Note that after going through the first page, I started noting down all language mistakes, because it became somewhat tedious. So the authors should pay attention to that on the later pages.

Reviewer #3 (Remarks to the Author):

Authors have performed the resonant inelastic X-ray scattering (RIXS) and scanning tunneling microscopy (STM) measurements on two different doping levels of the heavily underdoped $Bi2.0Sr2.0Ca0.6Dy0.4Cu2O8+\delta$ samples that are near the onset of the superconductivity. The authors observed charge orders in both insulating and superconducting phases near the boundary of the superconducting dome. The authors also observed an increase of the bond-buckling phonon intensities in the superconducting sample, proposing that the Cooper pairs emerge from the charge-ordered insulating state.

A link between RIXS and STM has been missing and the combined approach in the present manuscript is interesting and highly encouraging for understanding the charge order phenomenon in the cuprates and relationship to the high temperature superconductivity. However, while the RIXS results are new, the manuscript is not benefitted from the STM measurements since similar results have already been reported with much higher quality (Ref. 53). More importantly, the charge order is not properly detected, characterized, and evaluated in the STM data, and I see that there are number of issues (see comments

below). I don't think I can recommend for a publication in the nature communications. My comments and concerns are as follows.

1. The Dy substituted Bi2212 is chemically inhomogeneous such that Tc of the as-grown samples strongly depends on piece by piece. The authors indeed pointed out in the supplementary materials that the Tc of the as-grown sample shows very broad superconducting transition about 20K in the magnetic susceptibility, implying that the doping level is not uniform even within the piece. I wonder if the authors have checked the Tc of an individual piece of the sample before annealing in the same batch. Even if samples are annealed at the same condition, final doping level can be different since as-gown Tc is different from piece to piece. I also wonder how the authors made sure that the doping level of the insulating samples used for the RIXS and STM measurements are the same (assuming that these measurements used different pieces). Also, what's the error bar of the doping level?

2. The authors claim that the doping levels are determined by the empirical formula from the ref. 50. In this formula, there is a "free" parameter of the maximum Tc. What's the maximum Tc did the authors use in this formula? In order to get p~0.06 with Tc~20K, the maximum Tc must be around 115K, which is not true for Bi2212. Similarly, for the as-grown sample, the maximum Tc must be around 67K to get p~0.09 with Tc~40K. First, the maximum Tc in the formula must be the same for all the estimations of the samples in the manuscript since. Second, the maximum Tc used in the formula is incorrect. Third, and more importantly, the maximum Tc strongly depends on where the dopants are in the crystal (H. Eisaki et al., Phys. Rev. B 69, 064512 (2004)). The authors need to identify a maximum Tc for the Dy substituted Bi2212, otherwise one cannot use this formula for the estimation of p. In any cases, the authors need careful examinations of the doping levels.

3. Differential conductance maps, dI/dV at low energies (~30mV) are a mixture of the local density of states modulations (if any) and the charge order modulations at ΔPG due to the setup effect of the STM junction formation in the heavily underdoped cuprates, indicating that dI/dV maps are not a good measure of the charge order. In fact, modulation amplitude and contrast (phase) in dI/dV map strongly depends on the setup condition (see for the systematic analysis in Seamus Davis group's thesis by C. B. Taylor Ph. D Thesis, p. 90-91,Cornell University (2008) and A. Schmidt, Ph. D. Thesis, p. 21, Cornell University (2009)). At least, one cannot evaluate the charge order with such a systematic error in the

measurements. Instead, the R-map (=I(r,+E)/I(r,-E)) or Z-map (=dI(r,+E)/dV/dI/dV(r,-E)), in which the setup effects are effectively mitigated (as the authors presented in the Fig. S5), have been widely used to study electronic modulations (e.g. Ref. 23, T. Hanaguri et al., Nature Phys. 3, 865 (2007), Y. Kohsaka et al., Nature 454, 1072 (2008)). I don't think that the authors can study the charge order from the low energy dI/dV, in which modulation amplitude and contrast (phase) strongly depend on the setup condition. Analyses of the dI/dV would lead to erroneous conclusions.

4. It has also been established that the charge order in Bi2212 appears to be the strongest at the pseudogap energy scale ΔPG. This is the energy scale where the unidirectional d-symmetry charge order is observed without ambiguity. Such an electronic structure can be accessed in the R(r,E) or Z(r,E) (or Z(r,e=E/ΔPG(r)) for e=1). See the paper by Y. Kohsaka et al., Nature 454, 1072 (2008).

5. It has been identified that the symmetry of the unidirectional charge order in Bi2212 (and Na-CCOC) is the d-symmetry (K. Fujita et al., PNAS 111, E3026 (2014)), in which electronic degree of freedom are primarily on the Ox and Oy sites within the CuO2 plaquette and are π out-of-phase. This suggests that, in order to study the charge order, data must be taken with intra-unit-cell spatial resolutions that should be able to distinguish the electronic degree of freedom on Ox and Oy from Cu sites. As far as I can tell in the manuscript, STM images are not taken properly as I don't clearly see unidirectional d-symmetry charge order (Ref. 53). Furthermore, the authors should look at a d-symmetry channel in the phase-resolved fourier transform to obtain correct q-space electronic structure of the charge order (K. Fujita et al., PNAS 111, E3026 (2014)).

6. A. Mesaros et al., PNAS 113, 12661 (2016), has reported that the charge order is highly heterogeneous in real space, exhibiting domain structures of the unidirectional charge order with the discommensurations. They have reported that a peak structure of the charge order in q-space is complicated having multiple satellite peaks due to the discommensurations and is not simply captured by the gaussian and/or Lorentzian line shape to determine the wavevector. They have introduced an optimization technique to estimate wavevectors of the charge order and identified that the wavevectors turned out to be doping independent and are commensurate to the lattice (4a0), as opposed the claims the authors made in the manuscript. This suggests that a peak position of the fourier transform is not the wavevector that characterizes the charge order. Their results have been

reproduced by Yi Zhang et al, Nature 570, 484 (2019), who applied a machine learning technique of a convolutional neural network to judge if the charge order is either commensurate to the lattice (4a0) or not as a function of doping.

7. Fig3b shows the fourier transform of Fig3a, but symmetrized and smoothed, loosing fundamental information of the charge order as described above. I don't think the authors can simply argue a wavevector extracted from the symmetrized/smoothed fourier transform.

8. I also think that if the elastic peak in the RIXS spectrum is probing the charge order, then it should reflect the discommensulations of the charge order such that the peak structure of the elastic peak is expected to have multiple satellites, although it might depend on a size of the beam. However, a resolution in the reciprocal space in the RIXS measurement is not high enough to resolve them. Thus, I am afraid that none of the wavevectors from both RIXS and STM precisely reflect intrinsic wavevector of the charge order. Without proper evaluations of the charge order wavevectors, I don't think one can draw a summary shown in Fig. 6.

9. In line 135, the authors described 'Here the result suggests that the CO in insulating cuprates is generally incommensurate with the underlying lattice, in contrast to previous studies.' I don't think the authors can make this claim without evaluations of the charge order wavevector by using the technique introduced by A. Mesaros et al., PNAS 113, 12661 (2016), in which they claim that QCO is doping independent and commensurate to the lattice.

10. Ref. 29 predicts how A1g mode is going to look like in the RIXS spectra. In this reference, a dispersion of the A1g mode is relatively weak, and a spectral weight is uniform for $0.1<q<0.5$. However, the A1g mode the authors extracted is highly localized, exhibiting strong H-dependent intensities as shown in Fig. 3. While the bond stretching mode the authors observed is consistent with the one predicted in the Ref. 29, A1g mode looks quite different from the predictions. Why are they different?

11. There is a precedent doping dependent study of the RIXS on Bi2212 from Stanford group, H. Lu et al., Phys. Rev. B 106, 155109 (2022), in which phonon anomalies are clearly detected. They use one-phonon fitting to extract a dispersion of the bond stretching mode that exhibit phonon anomalies. Apparently, their raw data and those in the present manuscript are highly similar, exhibiting the elastic peak and bond stretching mode clearly. An energy resolution is almost identical to the present manuscript. I wonder what is different in their studies from those in the present manuscript other than the doping level. The authors also claim an importance of the BB mode for driving or accompanying the condensation of Cooper pairs (line. 279). However, H. Lu et al., didn't observe BB mode even at optimally doping where Tc is the highest. This is paper is also highly relevant to the present manuscript and the authors should cite.

12. In line 271, the authors described that 'From our work, while the CO is indispensable for the formation of Cooper pairs, the CO wavevector QCO is found to continuously change with doping, irrespective of the SC (Fig. 6a).' This statement is apparently inconsistent with recent paper by H. Lu et al., Phys. Rev. B 106, 155109 (2022), in which they claim that QCO is constant around 0.25 above some doping. How do the authors incorporate their results?

13. In line 340, the authors described that 'An auxiliary strategy to uncover the origin of CO may be to consider why the evolution of QCO follows an almost linear trend in a wide range of hole concentration, irrespective of the SC, intertwined orders, and Fermi surface topology'. As I pointed out above, A. Mesaros et al., PNAS 113, 12661 (2016) claims that QCO is doping independent. Without the evaluation of the discommensulations of the charge order, this question doesn't make sense.

14. In line 373, the authors described that 'Similar observations sensitive to the Cooper pairs condensation include the antinodal kink of BB phonon by ARPES [48], and ….'. In the ref. 48, they reported that the B1g phonon is coupled to the single particle excitation spectrum which appear to be strongest near the anti-nodal region, but not A1g mode. The B1g mode is also a bond-buckling phonon, but Ox and Oy motions are out-of-phase and different from A1g symmetry. So, it's confusing that reader gets impression that the 'BB phonon' the authors refer to is the same as those in the ref. 48, but they are different.

-In this letter, we provide a point-to-point response to the reviewer's comments.

The reviewer's original comments are shown in blue italic characters in the following.

Black regular characters show the authors' responses.

Reviewer #1 (Remarks to the Author):

In the manuscript by Zou et al., the temperature dependence of charge order (CO), phonons, and low energy excitations in lightly doped Dy-Bi2212 (p=0.04, 0.06) were measured using high-resolution RIXS and STM. The authors report the observation of CO and phonon modes, which were extracted from RIXS data taken with an instrumental energy resolution of 36 meV. The dispersions of phonons are not found to soften near the CO position. Notably, the manuscript provides experimental evidence for the existence of CO in lightly doped cuprate and pinpoints the origin of CO and its competition with SC.

The origin of CO and its relevance to the superconductivity in cuprates are important issues of current interest. High-resolution RIXS has the potential to unravel new insights into these issues. The experimental data are interesting and up-to-date. However, analyses and discussions on photon dispersion are not fully convincing in understanding electron-phonon coupling (EPC), as commented below. This leads the reviewer to conclude that the present manuscript does not satisfy the condition of a high-profile paper required by Nat. Commun. Therefore, I don't recommend the publication of the present manuscript unless the following major issues are satisfactorily addressed:

Our answer: We thank the reviewer for reviewing our manuscript and recognizing the importance of our work. We also thank the reviewer for their constructive comments and suggestions to improve our manuscript. We have carefully addressed the reviewer's comments on the data analysis and discussions, as elaborate below.

(1) Regarding the phonon dispersion and softening, my biggest concern is that the energy resolution of the present work is not good enough to extract a convincing conclusion for low-energy phonons. Several modes of low-energy phonons, including acoustic, A1g and B1g buckling modes, exist in hole-doped cuprates. Their anomalies due to the formation of CO have been observed by inelastic x-ray scattering (e.g., La Tacon et al., Nat. Phys. 10, 52. (2014)) and high-resolution RIXS (e.g., Ref. [32]). The phonon softening and EPC exist in under-doped YBCO and optimally doped LSCO. Interestingly, the present manuscript shows no evidence of phonon anomalies for the wave vector across CO in lightly doped Bi2212. In my viewpoint, the data

analysis with only two phonon components to fit the RIXS spectra broadened by an instrument of energy resolution 36 meV does not correctly reveal the phonon softening and EPC. Using the present RIXS data to rule out the dispersion of low-energy phonons buried in the tail of the "broad" elastic peak is not convincing. The authors also commented similarly on previous works, Ref. [6, 20, 21, 31]. The conclusion of the present manuscript about phonon anomalies requires further support of RIXS data taken with an improved energy resolution. Perhaps it is very challenging to improve the energy resolution of RIXS significantly. Alternatively, the authors are suggested to substantially modify the discussions on the present data to address the issue of phonon anomalies correctly.

Our answer: We thank the reviewer for the constructive comments and incorporated their suggestions to improve our manuscript.

We agree with the Referee that several low-energy phonons, including acoustic, A1g and B1g buckling modes, exist in hole-doped cuprates. Limited by our resolution of $\Delta E = 36$ meV, we can only detect the buckling mode (30~50 meV) and breathing mode (60~70 meV), which agrees with a previous RIXS study on AF NBCO by Braicovich et al. with an energy resolution of $\Delta E = 40$ meV (ref. 27). Therefore, we concur with the reviewer that our present analysis is insufficient to rule out the softening behavior of lower energy phonons, as observed in previous IXS studies on YBCO and high-resolution RIXS studies in optimally doped LSCO.

Based on the reviewer's suggestion, we have modified the discussion on the phonon softening from our two-phonon fitting model. We have included a discussion on the potential softening of BB and lower-energy phonons and emphasized the importance of considering the limited energy resolution in drawing conclusions. For the same reason, the discussion regarding lattice distortion is deleted from the original manuscript. The corresponding revisions are shown in red (lines 230-235) in the new manuscript, and the IXS study on YBCO has been added as a new reference (ref. 28)

(2) The paper reports CO of wave vectors ~ 0.31 r.l.u. and concludes that the CO is incommensurate with the lattice structure. In combining with dispersionless BB phonon, the authors rule out the possibility of the formation of charge-density waves (CDW) induced by lattice distortion. However, observing a non-fractional modulation vector can result from discommensurations or a truly incommensurate order. The discommensurations of CDW have been discussed in many papers, for example, Masros et al., Proc. Natl. Acad. Sci. 113, 12661 (2016), Vinograd et al., Nat. Commun. 12, 3274 (2021), and Ref [32]. The authors need to discuss whether the observed CO is truly incommensurate or appears to be incommensurate due to discommensurations.

Our answer: We thank the Referee for drawing our attention to these references. We agree with the reviewer that a discussion regarding the discommensurations or a truly incommensurate order should be given, which is highly relevant for distinguishing between k-space Fermi-surface nesting

and r-space electronic strong correlations scenarios. This is significant for understanding the mechanism of charge order in cuprates.

As pointed out by Mesaros *et al.*, Proc. Natl. Acad. Sci. **113**, 12661 (2016), detecting discommensurations by amplitude-measuring scattering probes can be hindered by the disorder, which would broaden the peak intrinsically. Therefore, without phase sensitivity, RIXS cannot distinguish whether the observed CO is truly incommensurate or appears incommensurate due to discommensurations, as pointed out by recent RIXS studies [Phys. Rev. X 11, 041038 (2021) and PRB 106, 155109 (2022)]. The STM work of Mesaros *et al.* is analyzed based on the phase-resolved Fourier-transformed "Z-map" (dI/dV(+E)/dI/dV(-E)) — called "charge density modulation" (CDM) in their study. While our observed CO, revealed by a traditional Fourier transformation on dI/dV (proportional to the density-of-states) channel, differs from their concept, we agree that discommensurations cannot be ruled out as a possible cause of the incommensurate wavevector, and we recognize the importance of considering this perspective.

Change made: In the revised manuscript, we have discussed whether the CO is truly incommensurate or appears to be incommensurate (marked in red). We emphasize that the exact scenario remains to be understood (lines 306-316), and the works raised by the reviewer have been included as new references (refs. 33, 34). In particular, our findings can be reconciled with the discommensurations in a r-space strong correlation scenario (lines 318-331).

In addition, there is one mistake in the caption of Fig. 1b, which uses the phase diagram to show the RIXS measurement strategy. However, the caption incorrectly says, " The orange and purple circles represent our RIXS and STM data, respectively. " In fact, Fig 1b plots neither RIXS nor STM data.

Our answer: We thank the reviewer for this correction. In the revision, we have revised the sentence to "The orange and purple circles denote the specific temperature and doping levels at which the RIXS and STM data were respectively collected".

Reviewer #2 (Remarks to the Author):

Zou et al. present a combined resonant inelastic X-ray scattering (RIXS) and scanning tunneling microscopy (STM) data on the cuprate superconductor Bi2Sr2Ca0.6Dy0.4Cu2O8+δ. The aim of their study is to shed light on mechanism of superconductivity close to the insulating regime.

The key results of their study are:

i) Charge order persists to even lower hole concentrations (p = 0.04) than previously known.

ii) Charge order is incommensurate and is thus makes a lattice induced distortion unlikely.
iii) Phonon softening near the charge order wave vector is absent
iv) There is no interplay between spin excitations and charge order
v) The intensity of the bond buckling phonon is strongly enhanced in the superconducting state.

Our answer: We thank the Reviewer for their careful review, for recognizing the significance of our work, and for providing constructive suggestions to improve the clarity of our manuscript. The reviewer correctly summarized the key results of our manuscript, and we would like to supplement two more findings in our work:

vi) Near the onset edge of the SC dome, CO intensity is enhanced when entering the SC dome (Fig. 5c).

vii) The bond buckling phonon intensity anomaly is likely triggered by the underlying CO phase.

From this the authors somewhat conclude that both the electron-phonon coupling and charge order work together to support superconductivity. In particular, they speculate that the onset of charge order at temperatures higher than the superconducting critical temperature, is a sign of Cooper pairs being pre-formed. In my opinion, this conclusion is not borne out by their results.

Our answer: Our findings, including that the charge order (CO) onset temperature is higher than T_c, and that the CO emerges in the insulating doping (p = 0.04) with its intensity further enhanced within the superconducting dome, provide robust evidence supporting the notion that superconductivity develops from the CO phase (as depicted in path #2 and #3 in Fig. 1b). Furthermore, the key results v) and vii) indicate a collaborative role of electron-phonon coupling (EPC) and charge order in facilitating superconductivity.

Regarding the reviewer's concern, we acknowledge that our experiments do not directly detect Cooper pairs, and our intention was not to propose the existence of pre-formed Cooper pairs through our results. We apologize for any confusion in this regard. The concept of pre-formed Cooper pairs has been suggested by other studies, such as those involving Nernst, STM, transport, and ARPES in the charge-ordered phase (e.g., Refs. [37-41]). Our aim is to emphasize that the pre-formed Cooper pairs condense concurrently with the enhanced EPC.

Change made: We have made the necessary modifications to the discussion to provide a clearer explanation of how electron-phonon coupling (EPC) and charge order (CO) collaborate to support superconductivity (see lines 278-279, 284-286, 291). Additionally, we have revised the paragraph that previously discussed Cooper pair formation (lines 341-346) to eliminate any potential confusion. The revised sections have been highlighted in red for easy identification.

Although, I appreciate that high-quality of the data, and also the effort to study the same sample

with two complementary methods, and also think that the data is quite interesting, I can currently not support publication of the article in its current form in Nature Communications.

I also would like to highlight that there are many English mistakes, use of unsuitable wording, suse of colloquial language, and so. This poses a problem in following what the authors are attempting to say, so it should be fixed.

In addition to the language of the article often not being clear, the conclusions are not presented clearly. I would suggest that the authors carefully go through the article, and both remove English language mistakes, and attempt to make introduction and discussion more concise and less convoluted. What are the main questions being answered and why? What it the impact of this on the $Bi2Sr2Ca0.6Dy0.4Cu2O8+\delta$, the family of cuprates and unconventional superconductivity broadly.

Our answer: We are grateful to the reviewer for acknowledging the high quality of our work and providing valuable comments and suggestions to improve our manuscript. We have carefully incorporated these suggestions and extensively revised the points raised by the referee in our revision. In particular, we have made significant revisions to the introduction and discussion sections to present our main findings, which address the following important questions:

1. What is the lowest hole concentration at which a short-range CO emerges in a doped Mott insulator, and what is its relationship with superconductivity?
2. Is the CO phase bulk or surface, and what is its nature? Does it conform to a weak coupling q-space nesting scenario or a strong coupling r-space scenario?
3. What is the relationship between electron-phonon coupling (EPC) and superconductivity formation in cuprates?

Our results have several important implications:

1. We have extended the existence of CO down to the antiferromagnetic regime ($p = 0.04$), the lowest doping level among all cuprates, where the Fermi surface is missing. This rules out the Fermi-surface nesting scenario as the origin of charge order. Our results suggest that the Cooper pairs emerge from the CO phase, ruling out a simple competing scenario.
2. Our work demonstrates, for the first time, that the CO in the insulating phase has a bulk nature and pervades the cuprate phase diagram down to $p = 0.04$ and temperatures above $T = 273$ K.
3. We have shown that the BB phonon mode, not the BS mode, is critical in connecting to SC. The former is strongly enhanced within the SC dome, while the latter agrees well with a non-interacting model calculation. Significantly, the peak of EPC strength of the BB

mode is induced by an underlying CO, suggesting that the CO and EPC work together to support the SC.

Our findings reveal that the Cooper pairs emerge from a charge-ordered state and possibly condense via an EPC-driving mechanism. This provides important insight into understanding the intertwining phases in cuprates and other novel systems (e.g., Ni-based compounds) where unconventional superconductivity and CO are intimately correlated.

Change made: The introduction (lines 45-72) and the discussion (lines 297-337, 350-354) have been rewritten thoroughly to clarify the above findings.

In addition, I would recommend that the authors also look at the following comments:

Our answer: We thank the reviewer for carefully reading our manuscript and highlighting our language mistakes. We have taken great care to correct these mistakes throughout the revised manuscript.

- Line 15: "How Cooper pairs from and condense have been the main challenges …": This should be singular. "…has been the main challenge".

Our answer: Following the reviewer's suggestion, here we use the singular form of this word.

- Line 19: "…the Fermi surface is lacking…". I assume the authors mean missing.

Our answer: We agree that the latter adjective is more accurate here. We have changed the word "lacking" to "missing" in the abstract (line 20), and to "absent" in the introduction part (line 70).

- Line 23: "We show that an incipient charge order has existed…". This should be: "We show that incipient charge exists…".

Our answer: Following the reviewer's suggestion, we have used "exists" instead of "has existed" in this sentence (line 24).

- Line 34: "evidences". Evidence is to the best of my knowledge always singular.

Our answer: Thanks for the correction. We have corrected this language mistake and the following verb in the revised manuscript (introduction part, line 34).

- Line 36: "…a different story proposes…". This is colloquial language, and should be changed.

Our answer: Following the reviewer's suggestion to make the introduction more concise, we have revised the sentence in lines 34-38.

- Fig. 1b. Why not add the onset of charge order derived from this work? Please add

Our answer: Thanks for the suggestion. However, as the CO exists up to the highest accessible temperature (273 K) in our measurement for both samples, we cannot determine the CO onset temperature.

- Fig. 1b. The symbols are not pentagrams, but stars.

Our answer: Thanks for pointing out this mistake. We have revised the "pentagrams" to "stars" in the caption of Fig. 1b.

- Fig. 1b. The superconducting region is not labelled, even though it is the most important phenomena the article describes.

Our answer: Thanks for the suggestion. We indicated the SC region with "T_c" in Fig. 1b. To make the SC region more apparent, we have added "SC" to the SC dome.

- Fig. 2a/b/c. The white dashed lines are barely visible.

Our answer: Thanks for the comment. We have drawn the white dashed lines thicker to make them more apparent to the readers.

- Fig. 2. Caption. "fittings" have nothing to do a fit. I believe you would like to say "fits". Also, the caption does not introduce what a "PG map" is. This is not a standard expression. It is explained in the main text, but not in the caption. The caption should at least refer to the main text.

Our answer: We thank the reviewer for pointing out our unclear expressions. Following their suggestion, the noun "fittings/fitting" has been corrected to "fits/fit" throughout the revision. The remaining "fitting" in the manuscript is the present continuous tense of the verb "fit". Moreover, we have added a sentence to explain the PG map in the caption of Fig. 2g.

- Fig. 5b. Please plot the lines for the fit beyond the recorded data so that the peak shape can be judge better.

Our answer: Thanks for the suggestion. In the revised Fig. 5b and supplementary Fig. S7a, we have shown the fitted curves beyond the recorded data. All fitted lines are plotted with the same range (H = 0.1~0.5 r.l.u.) to reveal the peak shape better except for that of 215 K. The recorded

data points at 215 K are not enough for a fit with the same range, so its fitted line is plotted in a smaller range. The comparison between the previous and the revised plot is shown in the figure below.

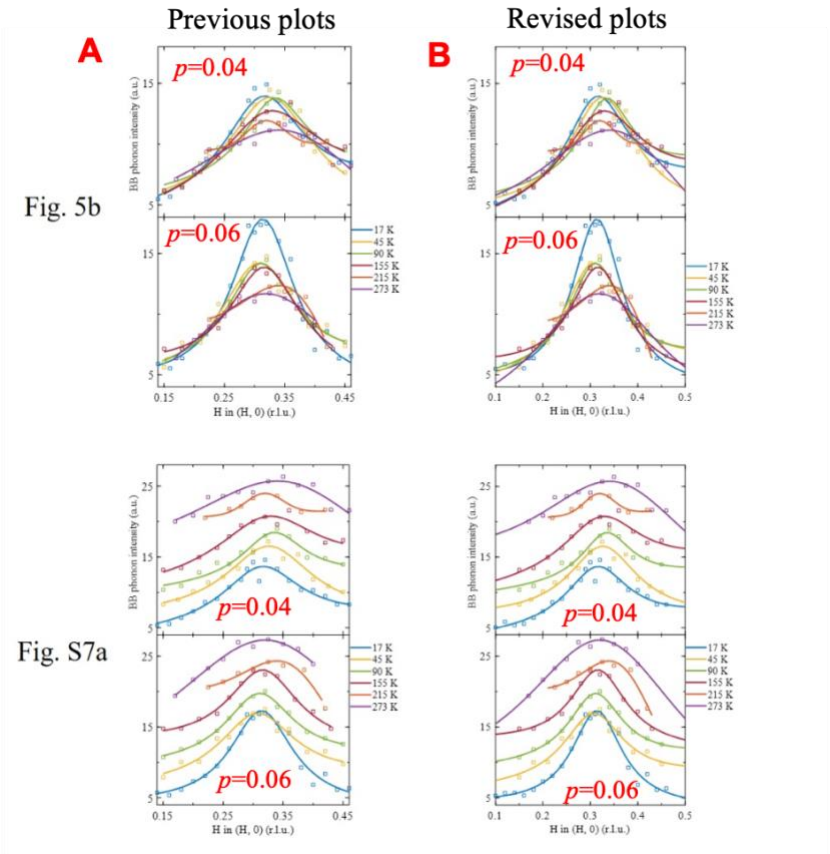


Figure R1: (A) Previous plot of normalized phonon intensity profiles of the p = 0.04 (upper panel) and p = 0.06 (lower panel) samples in Fig. 5b and Fig. S7a (offset for clarity). (B) Revised plot with the fitted curves beyond the recorded data (H = 0.1~0.5 r.l.u.) except for that of 215 K.

- Fig 5 c,d have no numbers on the y-axis. Even for intensity in a.u. it is essential were the zero is to understand how strong the effect is. Please add.

Our answer: Thanks for the suggestion. We have added the numbers to the y-axis in Fig. 5c,d.

- Fig. 6. It is not clear if how the dI/dV curves in panel c connect to the RIXS data in panel b. Please explain better what you mean.

Our answer: We thank the reviewer for this comment. To clarify our idea, we have demonstrated the connection between Fig. 6b and 6c in more detail in the figure caption.

Note that after going through the first page, I started noting down all language mistakes,

because it became somewhat tedious. So the authors should pay attention to that on the later pages.

Our answer: We appreciate the reviewer's thorough reading and constructive comments to improve our manuscript. We have gone through the article to correct the language mistakes. We believe that our revision has corrected the language mistakes.

Reviewer #3 (Remarks to the Author):

Authors have performed the resonant inelastic X-ray scattering (RIXS) and scanning tunneling microscopy (STM) measurements on two different doping levels of the heavily underdoped $Bi2.0Sr2.0Ca0.6Dy0.4Cu2O8+\delta$ samples that are near the onset of the superconductivity. The authors observed charge orders in both insulating and superconducting phases near the boundary of the superconducting dome. The authors also observed an increase of the bond-buckling phonon intensities in the superconducting sample, proposing that the Cooper pairs emerge from the charge-ordered insulating state.

A link between RIXS and STM has been missing and the combined approach in the present manuscript is interesting and highly encouraging for understanding the charge order phenomenon in the cuprates and relationship to the high temperature superconductivity. However, while the RIXS results are new, the manuscript is not benefitted from the STM measurements since similar results have already been reported with much higher quality (Ref. 53). More importantly, the charge order is not properly detected, characterized, and evaluated in the STM data, and I see that there are number of issues (see comments below). I don't think I can recommend for a publication in the nature communications. My comments and concerns are as follows.

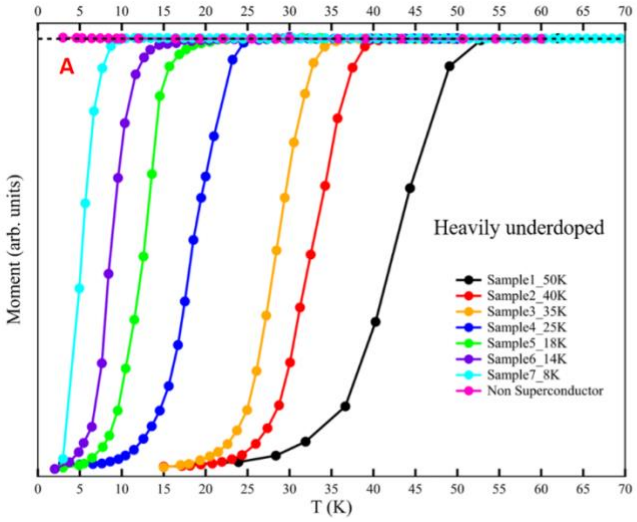
Our answer: We thank the reviewer for their thorough evaluation of our research and for recognizing the importance of our study on cuprates that combined RIXS and STM techniques. While we are grateful for the recognition of the novelty of our RIXS study, we also recognize the concerns raised regarding the novelty and analysis of our STM study. Upon careful examination of the referee's comments, we have identified a misunderstanding in their assessment. The referee may have referred to a different analysis, leading to confusion about our results and their validity. To address their concerns, we would like to clarify the methodology and analysis of our STM study. In particular, we want to emphasize that the CO observed at p = 0.04~0.06 has never been reported before and should not be mixed up with a different phase, such as the "C_{2v}-symmetric clusters" in the previous Ref. 53 (now Ref. 51). We have provided a detailed explanation of the validity of our charge order analysis below and believe that we have addressed their concerns satisfactorily.

1. The Dy substituted Bi2212 is chemically inhomogeneous such that Tc of the as-grown samples strongly depends on piece by piece. The authors indeed pointed out in the supplementary materials that the Tc of the as-grown sample shows very broad superconducting transition about 20K in the magnetic susceptibility, implying that the doping level is not uniform even within the piece. I wonder if the authors have checked the Tc of an individual piece of the sample before annealing in the same batch. Even if samples are annealed at the same condition, final doping level can be different since as-gown Tc is different from piece to piece. I also wonder how the authors made sure that the doping level of the insulating samples used for the RIXS and STM measurements are the same (assuming that these measurements used different pieces). Also, what's the error bar of the doping level?

2. The authors claim that the doping levels are determined by the empirical formula from the ref. 50. In this formula, there is a "free" parameter of the maximum Tc. What's the maximum Tc did the authors use in this formula? In order to get p~0.06 with Tc~20K, the maximum Tc must be around 115K, which is not true for Bi2212. Similarly, for the as-grown sample, the maximum Tc must be around 67K to get p~0.09 with Tc~40K. First, the maximum Tc in the formula must be the same for all the estimations of the samples in the manuscript since. Second, the maximum Tc used in the formula is incorrect. Third, and more importantly, the maximum Tc strongly depends on where the dopants are in the crystal (H. Eisaki et al., Phys. Rev. B 69, 064512 (2004)). The authors need to identify a maximum Tc for the Dy substituted Bi2212, otherwise one cannot use this formula for the estimation of p. In any cases, the authors need careful examinations of the doping levels.

Our answer: The above two questions raised by the reviewer are related, so we address them together. The reviewer mainly concerns with the broadness (thus the inhomogeneity) of our Dy-Bi2212 samples and the uncertainty of the doping levels. We thank the reviewer for pointing out the missing information in our manuscript. We have clarified this point below and added this information in our revised manuscript.

In underdoped cuprates, the SC transition is generally broad mainly for two reasons. The first one is owing to the difficulty in annealing severely underdoped Bi2212. We have carried out a thorough study using different Dy concentrations and annealing conditions for the sample preparation, and the Dy-Bi2212 samples show broad transitions robustly (See Figure R2). The second reason is that compared to the doping range close to the top of the SC dome, the T_c vs. p trend is much steeper near the dome edge, which means that an equal uncertainty range of p will lead to a broader distribution of T_c in this regime.



$Bi_2Sr_2Ca_{1-x}Dy_xCu_2O_{8+\delta}$
(x = 0.3~0.4)

B

Sample	Dy	Condition	Temperature (°C)	Time (day)
1	0.3	air	500	5
2	0.38	air	500	5
3	0.38	air	500	5
4	0.4	0.5 atm O_2	500	5
5	0.38	air	500	5
6	0.4	0.5 atm O_2	500	5
7	0.4	vacuum	500	5

Figure R2: Annealing of Dy-Bi2212 samples to severely underdoped regime. (A) The best obtained Dy-Bi2212 samples at each doping range generally display broad transitions. (B) The corresponding Dy concentration, annealing atmosphere, temperature, and time of each sample in (A). Figure adapted from *Q. Gao*, Ph. D. Thesis, p. 56-57, University of Chinese Academy of Science (2020).

As the reviewer points out, we have taken the widely used formula $T_c/T_c^{max} = 1 - 82.6 * (p - 0.16)^2$ from Ref. 50 (now Ref. 48 in the revised manuscript). In our work, the maximal transition temperature T_c^{max} is set to 96 K, taken from H. Eisaki *et al.*, Phys. Rev. B **69**, 064512 (2004), and the doping levels are estimated with T_c equals the onset transition temperature. For example, the doping of the SC sample is $p = 0.062$ for $T_c = 20$ K (line 563 in the original manuscript). We apologize for mistakenly estimating the doping level of the as-grown sample in our original manuscript, which was estimated as $p \sim 0.09$. This value was obtained by referring to Ref. 16 reporting underdoped Bi2212 samples with similar T_c of 45K/40K as our as-grown sample. However, the accurate doping level of our as-grown sample should be $p = 0.075$ ($T_c = 39$ K) following the same formula above.

From the SC transition width, we estimate that the doping range for the as-grown sample is from $p = 0.059$ to $p = 0.075$, the annealed SC sample is from $p = 0.05$ to $p = 0.062$, and the annealed insulating sample is $0.03 < p < 0.05$. Therefore, the doping levels with error bars for the three samples can be estimated as $p = 0.067 \pm 0.008$ (as-grown sample), $p = 0.056 \pm 0.006$ (annealed SC sample), and $p = 0.04 \pm 0.01$ (annealed insulating sample). For comparison, the doping level of a much sharper transition from $T_c = 96$ K to 93 K is estimated to be $p = 0.15 \pm 0.01$. These T_c and doping ranges are schematically shown in Figure R3 below. The reviewer suggests that we use the T_c^{max} of Dy0.4-Bi2212 for the calculation. This is very difficult in practice as the Dy substitutions are introduced for severe underdoping, and references for "optimally" doping Dy0.4-Bi2212 are unavailable. Therefore, here we follow the same convention of previous works (for example, a recent paper raised by the reviewer PHYSICAL REVIEW B 106, 155109 (2022)) to fix the T_c^{max} to the "optimal" value of Bi2212 materials.

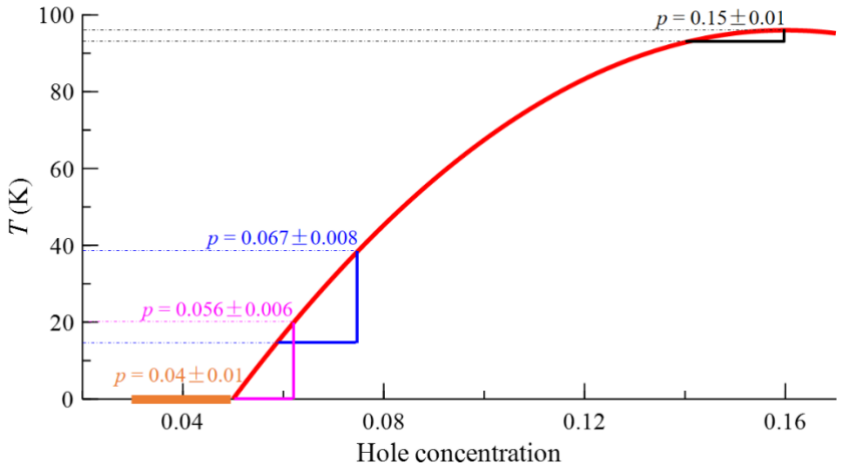


Figure R3: Schematic drawing of the T_c and doping ranges of the studied Dy-Bi2212 samples, compared to the as-grown sample and one near the optimal doing.

The reviewer is correct that the T_c might differ for different pieces before annealing in the same batch. To avoid such issues, we have used the same sample (Supplementary Fig. S1a, denoted as "as-grown") for further annealing. By dividing the as-grown sample into two smaller pieces, we have obtained the p = 0.04 (Fig. S1a, denoted as "Ann2") and p = 0.06 (Fig. S1a, denoted as "Ann1") samples in the manuscript. Notably, the STM measurements were carried out in the *very same* samples after the RIXS experiments (see line 103 of the previous manuscript).

Change made: Following the reviewer's suggestions, we have made the sample preparation and characterization more explicit, and most of the above information is added in the revised supplementary Section 1. In the revised Methods of the main text, we provide the error bar of the doping levels and emphasize that both the RIXS and STM measurements were carried out in the same sample (lines 435-437).

3. Differential conductance maps, dI/dV at low energies (~30mV) are a mixture of the local density of states modulations (if any) and the charge order modulations at ΔPG due to the setup effect of the STM junction formation in the heavily underdoped cuprates, indicating that dI/dV maps are not a good measure of the charge order. In fact, modulation amplitude and contrast (phase) in dI/dV map strongly depends on the setup condition (see for the systematic analysis in Seamus Davis group's thesis by C. B. Taylor Ph. D Thesis, p. 90-91,Cornell University (2008) and A. Schmidt, Ph. D. Thesis, p. 21, Cornell University (2009)). At least, one cannot evaluate the charge order with such a systematic error in the measurements. Instead, the R-map (=I(r,+E)/I(r,-E)) or Z-map (=dI(r,+E)/dV/dI/dV(r,-E)), in which the setup effects are effectively mitigated (as the authors presented in the Fig. S5), have been widely used to study electronic modulations (e.g. Ref. 23, T. Hanaguri et al., Nature Phys. 3, 865 (2007), Y. Kohsaka et al., Nature 454, 1072 (2008)). I don't think that the authors can study the charge order from the low

energy dI/dV, in which modulation amplitude and contrast (phase) strongly depend on the setup condition. Analyses of the dI/dV would lead to erroneous conclusions.

4. It has also been established that the charge order in Bi2212 appears to be the strongest at the pseudogap energy scale ΔPG. This is the energy scale where the unidirectional d-symmetry charge order is observed without ambiguity. Such an electronic structure can be accessed in the R(r,E) or Z(r,E) (or Z(r,e=E/ΔPG(r)) for e=1). See the paper by Y. Kohsaka et al., Nature 454, 1072 (2008).

5. It has been identified that the symmetry of the unidirectional charge order in Bi2212 (and Na-CCOC) is the d-symmetry (K. Fujita et al., PNAS 111, E3026 (2014)), in which electronic degree of freedom are primarily on the Ox and Oy sites within the CuO2 plaquette and are π out-of-phase. This suggests that, in order to study the charge order, data must be taken with intra-unit-cell spatial resolutions that should be able to distinguish the electronic degree of freedom on Ox and Oy from Cu sites. As far as I can tell in the manuscript, STM images are not taken properly as I don't clearly see unidirectional d-symmetry charge order (Ref. 53). Furthermore, the authors should look at a d-symmetry channel in the phase-resolved fourier transform to obtain correct q-space electronic structure of the charge order (K. Fujita et al., PNAS 111, E3026 (2014)).

6. A. Mesaros et al., PNAS 113, 12661 (2016), has reported that the charge order is highly heterogeneous in real space, exhibiting domain structures of the unidirectional charge order with the discommensurations. They have reported that a peak structure of the charge order in q-space is complicated having multiple satellite peaks due to the discommensurations and is not simply captured by the gaussian and/or Lorentzian line shape to determine the wavevector. They have introduced an optimization technique to estimate wavevectors of the charge order and identified that the wavevectors turned out to be doping independent and are commensurate to the lattice (4a0), as opposed the claims the authors made in the manuscript. This suggests that a peak position of the fourier transform is not the wavevector that characterizes the charge order. Their results have been reproduced by Yi Zhang et al, Nature 570, 484 (2019), who applied a machine learning technique of a convolutional neural network to judge if the charge order is either commensurate to the lattice (4a0) or not as a function of doping.

7. Fig3b shows the fourier transform of Fig3a, but symmetrized and smoothed, loosing fundamental information of the charge order as described above. I don't think the authors can simply argue a wavevector extracted from the symmetrized/smoothed fourier transform.

9. In line 135, the authors described 'Here the result suggests that the CO in insulating cuprates is generally incommensurate with the underlying lattice, in contrast to previous studies.' I don't think the authors can make this claim without evaluations of the charge order wavevector by using the technique introduced by A. Mesaros et al., PNAS 113, 12661 (2016), in which they claim that QCO is doping independent and commensurate to the lattice.

13. In line 340, the authors described that 'An auxiliary strategy to uncover the origin of CO may be to consider why the evolution of QCO follows an almost linear trend in a wide range of hole concentration, irrespective of the SC, intertwined orders, and Fermi surface topology'. As I pointed out above, A. Mesaros et al., PNAS 113, 12661 (2016) claims that QCO is doping independent. Without the evaluation of the discommensulations of the charge order, this question doesn't make sense.

Our answer: The above comments (#3-#7, #9, and #13) raise questions about the validity of our STM analysis and the demonstration of the charge order (CO) signal, particularly in comparison to the works of the Seamus Davis group. We acknowledge that these concerns are interrelated and addressed them together. In the following, we present arguments against most of the reviewer's comments by making it clear that it is the dI/dV map of STM, not Z-map or R-map, that is dedicated to CO demonstration and for a direct comparison to the RIXS CO signal. Meanwhile, we concur with the reviewer's idea that discommensurations cannot be ruled out as a possible cause of the incommensurate wavevector. Although this concept is based on Z-map analysis, we recognize the importance of considering this perspective. In our revised manuscript, we have addressed this point more explicitly and included a discussion of the potential influence of discommensurations on the observed results.

Addressing these concerns, we provide answers to three essential questions conveyed by the comments above, aiming to clarify the methodology and analysis of our STM study and the proper demonstration of the CO signal.

1) **Why do we use dI/dV map instead of the R-map/Z-map for CO analysis?**

Our manuscript focuses on the ***bi-directional*** "checkerboard" CO, and for such purposes, the dI/dV map in the low energy range (~±30 meV) is commonly employed in STM studies. This methodology has been widely used in the STM literature, as demonstrated by references [4, 7, 8, 10, 21, 38]. For instance, Ref. 4 presents a systematic STM study that identifies a decreasing trend of $\mathbf{Q}_{CO}$ in superconducting Bi2212, and Ref. 10 corroborates this trend in superconducting Bi2201 by combining STM and REXS methods. From the STM point of view, the defining feature of the CO is an in-phase density-of-states distribution between the particle (-E) and hole (+E) state, distinguishing it from other "electronic modulations" (comment #3) that are studied by the referred works (see comment #3-#6, #9, #13). For example, T. Hanaguri *et al.*, Nat. Phys., **3**, 865 (2007) (comment #3) reveals that the Bogoliubov quasi-particle interference (QPI) is out-of-phase between -E and +E, which is thoroughly demonstrated by K. Fujita *et al.*, PRB, **78**, 054510 (2008).

Different from the dI/dV map, the Z-map (dI/dV(+E) / dI/dV(-E)) is widely used for enhancing other electronic modulations, as mentioned by the reviewer. However, this is achieved by not only "mitigating the setup condition", but also eliminating the "checkerboard" CO (see, for example, comment #3: T. Hanaguri *et al.*, Nat. Phys., **3**, 865 (2007), comment #3, #4: Y. Kohsaka et al.,

Nature 454, 1072 (2008)). Therefore, for our specific analysis of CO, using the Z-map is not a practical approach.

The referred works that use the Z-map and R-map near the pseudogap (PG) energy of cuprates aim to reveal another electronic state characterized by a ***uni-directional*** modulation, which is fundamentally different from our focus on CO (~±30 meV). These electronic modulations likely represent the symmetry of PG (typically >100 meV), as demonstrated in comments #6, #9, and #13: "charge density modulations" in A. Mesaros *et al*., PNAS **113**, 12661 (2016), and comment #5: "d-form factor density wave" in K. Fujita *et al*., PNAS **111**, E3026 (2014), and "electronic nematicity" in Ref. 52, as well as comment #5: "C2v-symmetric clusters" in Ref. 51.

2) **How do we deal with STM set-point effect using dI/dV map**?

The reviewer is correct that the setup condition of STM spectra should be mitigated for more accurate analysis (comment #3). As exemplified in Figure R4 below, the so-called "set-point effect" is introduced if the bias voltage is low, as the tunneling current I (usually a constant) is an integral of the dI/dV from zero to the setup bias voltage. Besides the techniques mentioned by the reviewer, another strategy to mitigate such an effect is to take the dI/dV spectra at a higher setup bias voltage. For example, W. Ruan *et. al.,* Nat. Phys. **14**, 1178 (2018) uses V_s = -0.3 V for the CO analysis of Bi2212 at p = 0.06. In our work, we use V_s = -0.3 V for the p = 0.06 sample and V_s = -0.5 V for the p = 0.04 sample, respectively (see Methods lines 460-464).

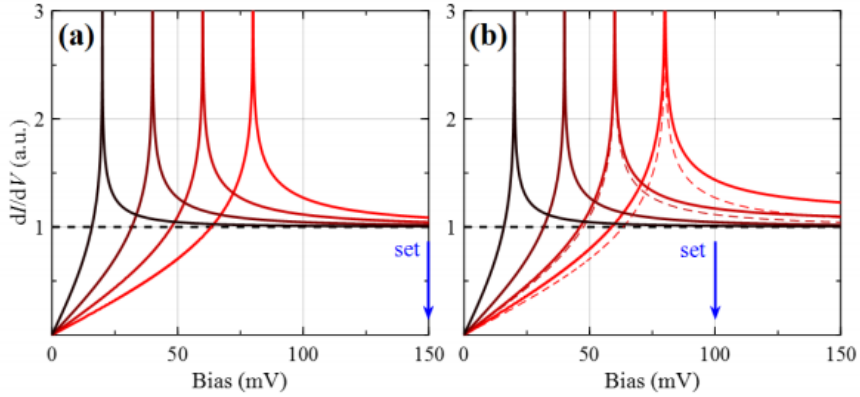


Figure R4: How the setpoint effect influences spectroscopy. (a) Simulated spectra with different d-wave gap, the background overlaps with the setpoint bias voltage V_s = 0.15 V. (b) Same as (a), but with a lower V_s = 0.1 V. The spectral weight deviates from the original value (dashed lines). The set-point effect will be more evident for the spectrum with a larger gap, which characterizes the underdoped cuprates.

3) **Is the CO wavevector incommensurate or due to discommensurations?**

We appreciate the reviewer's insight regarding the symmetrized and smoothed treatments, which may not be appropriate for analyzing ***uni-directional*** electronic modulations (comment #7). However, we would like to clarify that in the case of the CO studied in our work, it is ***bi-directional***,

and as such, a symmetrized treatment is commonly used for its analysis. Additionally, employing a smoothed treatment in the Fourier-transform STM (FT-STM) analysis aids in better revealing the wavevector from the discrete pixels. For reference, please see M. Hamidian *et al*., Nature **532**, 343 (2016).

As we emphasized in the above reply 1), it is crucial to differentiate between CO and uni-directional electronic signals as they represent distinct electronic states. While the analysis by A. Mesaros *et al*., PNAS **113**, 12661 (2016) could be reliable, its conclusions (comments #6, #9, #13) cannot necessarily apply to our study. We agree with the reviewer and A. Mesaros *et al.* that the $\boldsymbol{Q}_{CO} \neq 0.25$ r.l.u. could also be a result of the discommensuations. As pointed out by A. Masros *et al.*, the disorder can hinder the detection of discommensurations by amplitude-measuring scattering probes. Because our RIXS and traditional FT-STM do not have a phase resolution, we cannot rule out the possibility raised there. In the revised manuscript, we have discussed whether the CO is truly incommensurate or appears to be incommensurate. We appreciate the reviewer for drawing our attention to this important reference, as it has enriched our analysis and discussion on this critical aspect of the study.

Changes made: We thank the reviewer for their comments again. In the main text, we have included additional discussions addressing the critical questions about our utilization of the dI/dV map and its comparison with the Z-map (lines 151, and 494-503). Moreover, we have revised the discussion on CO, taking into account the possibility of reconciliation with the discommensurations in a r-space strong correlation scenario (lines 318-331).

8. I also think that if the elastic peak in the RIXS spectrum is probing the charge order, then it should reflect the discommensulations of the charge order such that the peak structure of the elastic peak is expected to have multiple satellites, although it might depend on a size of the beam. However, a resolution in the reciprocal space in the RIXS measurement is not high enough to resolve them. Thus, I am afraid that none of the wavevectors from both RIXS and STM precisely reflect intrinsic wavevector of the charge order. Without proper evaluations of the charge order wavevectors, I don't think one can draw a summary shown in Fig. 6.

Our answer: The momentum resolution ΔQ of the I21 RIXS beamline at Diamond Light Source is estimated to be 0.01 $Å^{-1}$ based on the scattering geometry, with a typical beam size of 2 μm (vertical) x 40 μm (horizontal) [Zhou, K.-J. et al. Journal of Synchrotron Radiation 29, 563–580 (2022)]. For Bi2212, considering that the lattice parameters of a=b=3.83 Å, this corresponds to ΔQ ~ 0.006 r.l.u.. This resolution is notably higher than the observed CO peak position (~0.3 r.l.u.) and width (FWHM of ~0.072 r.l.u.). Thus, the RIXS momentum resolution ΔQ (0.006 r.l.u.) is sufficient to accurately resolve both the CO peak and width. In fact, owing to its exceptional resolution in reciprocal space, RIXS has become a widely utilized technique for investigating CO

in various cuprates, including YBCO in Ref. [3], and Bi2212 in Refs. [6,20], and LSCO in Refs. [21,22]. The STM momentum resolution is estimated to be $\Delta Q = \frac{2\pi}{23.8nm} = 0.026\text{Å}^{-1} \sim 0.016\ r.l.u.$ (23.8 nm is the real-space size of Fig. 3a). Although not as good as RIXS, this resolution is still sufficient to resolve the CO peak and width.

Therefore, the momentum resolutions of both RIXS and STM measurements are sufficient for determining the wavevectors of charge order. However, the lack of phase sensitivity makes detecting phase slips resulting from disorder or topological defects difficult. As highlighted by Masros et al. [Proc. Natl. Acad. Sci. 113, 12661 (2016)], the disorder can hinder the detection of discommensurations by amplitude-measuring scattering probes, leading to the broadening of the charge order peak and wavevector shift. Recent RIXS studies have also pointed out that without phase sensitivity, it is challenging to distinguish between truly incommensurate charge order and apparent incommensurability caused by discommensurations [ref. 29 and PRB 106, 155109 (2022)].

Changes made: In the revised manuscript, we have provided the momentum-space resolutions to demonstrate that the CO peak can be well-resolved by RIXS and STM measurements (lines 413-417, 514-517). And in particular, the instrumental article on the I21 RIXS beamline is included as a new reference (Ref. 19).

10. Ref. 29 predicts how A1g mode is going to look like in the RIXS spectra. In this reference, a dispersion of the A1g mode is relatively weak, and a spectral weight is uniform for 0.1<q<0.5. However, the A1g mode the authors extracted is highly localized, exhibiting strong H-dependent intensities as shown in Fig. 3. While the bond stretching mode the authors observed is consistent with the one predicted in the Ref. 29, A1g mode looks quite different from the predictions. Why are they different?

Our answer: The presence of charge order induces the different behaviors in electron-phonon coupling (EPC) in cuprates. Ref. 29 (now Ref. 26 in the revised manuscript) presents the systematic model calculation of the electron-phonon coupling (EPC) strength in the absence of charge order. As demonstrated by L. Braicovich et al. in Ref. 27, RIXS studies on an insulating NBCO cuprate without charge order have reported that the electron coupling to BB A_{1g} and BS phonons follow the trends predicted by Ref. 29 (now Ref. 26 in the revised manuscript). In our work, we investigate Bi2212 with an incipient charge order. Our findings reveal that the dispersion and EPC strength of the BS phonon agree well with the prediction of Ref. 29 (now Ref. 26 in the revised manuscript), indicating that the charge order has a negligible effect on the BS phonon. However, the coupling to BB A_{1g} mode shows an anomaly near the $\mathbf{Q}_{CO}$, suggesting an interaction between the charge order and BB A_{1g} phonon.

11. There is a precedent doping dependent study of the RIXS on Bi2212 from Stanford group, H. Lu et al., Phys. Rev. B 106, 155109 (2022), in which phonon anomalies are clearly detected. They use one-phonon fitting to extract a dispersion of the bond stretching mode that exhibit phonon anomalies. Apparently, their raw data and those in the present manuscript are highly similar, exhibiting the elastic peak and bond stretching mode clearly. An energy resolution is almost identical to the present manuscript. I wonder what is different in their studies from those in the present manuscript other than the doping level. The authors also claim an importance of the BB mode for driving or accompanying the condensation of Cooper pairs (line. 279). However, H. Lu et al., didn't observe BB mode even at optimally doping where Tc is the highest. This is paper is also highly relevant to the present manuscript and the authors should cite.

Our answer: It is now widely accepted that a typical high-resolution RIXS spectrum of doped cuprates comprises contributions from bond-buckling phonons and bond-stretching phonons (see, for example, Phys. Rev. X 11, 041038 (2021), Phys. Rev. Res. 2, 023231 (2021), Phys. Rev. X 6, 041019 (2016)). Especially, the RIXS study on AF NBCO by Braicovich et al. has demonstrated the capability of Cu L_3-edge RIXS to simultaneously detect bond-buckling phonons and bond-stretching phonons with an energy resolution of $\Delta E = 40$ meV (Phys. Rev. Res. 2, 023231 (2021)). In our study with an energy resolution of $\Delta E = 36$ meV, we observed highly asymmetric lineshapes in the inelastic RIXS spectra, indicating the necessity of considering two phonons in data fitting (Figure 3d,e). In contrast, the paper cited by the reviewer (Phys. Rev. B 106, 155109 (2022)) examined only the BS branch to fit the inelastic part of the RIXS spectra and identify the "softening" of the BS phonon. We believe the BB mode is present in their studies, but it was not considered in their data analysis.

We have also conducted the one-phonon analysis by considering only the BS phonon and explored various lineshapes, including Lorentzian, anti-Lorentzian, and width-variable Gaussian functions, for the fits (see Supplementary Section 3). However, we found that the one-phonon model did not fit the data as well as the two-phonon model due to the presence of additional spectral weight at low energy (see Supplementary FIG. S6). Thus, we consider the two-phonon fitting model more appropriate for our data, and we have elaborated on this point in the revised manuscript. On the other hand, we acknowledge that our energy resolution of 36 meV may not be adequate to rule out the softening behavior of lower energy phonons, as observed in previous IXS studies on YBCO (La Tacon *et al.*, Nat. Phys. 10, 52. (2014)) and high-resolution RIXS studies in optimally doped LSCO (Ref. 29). In the revised version, we have clarified that a two-phonon fitting model is more suitable for our data (lines 224-227, 489-491), and we have added a discussion on the possibility of phonon softening (lines 231-235).

12. In line 271, the authors described that 'From our work, while the CO is indispensable for the formation of Cooper pairs, the CO wavevector QCO is found to continuously change with doping, irrespective of the SC (Fig. 6a).' This statement is apparently inconsistent with recent paper by H. Lu et al., Phys. Rev. B 106, 155109 (2022), in which they claim that QCO is constant around 0.25 above some doping. How do the authors incorporate their results?

Our answer: H. Lu's work reports a constant value of $\mathbf{Q}_{CO}$ above optimal doping. Our manuscript has only presented the doping evolution of $\mathbf{Q}_{CO}$ up to $p = 0.16$, where the decreasing trend of $\mathbf{Q}_{CO}$ is consistent with H. Lu's work. However, beyond the optimal doping level, the quantum critical point may play a dominant role in inducing a constant $\mathbf{Q}_{CO} \sim 0.25$ r.l.u., which is beyond the scope of our study. We agree that our previous statement that "$\mathbf{Q}_{CO}$ is found to continuously change with doping" is inaccurate in a broad doping range, and we have revised our discussion and conclusion accordingly.

Change made: In response to the reviewer's comment, we have limited our discussion to the underdoped regime in the revised manuscript (caption of Fig. 6a), and we have cited H. Lu's work as a new reference (Ref. 32). We appreciate the reviewer's valuable suggestion for this relevant paper.

14. In line 373, the authors described that 'Similar observations sensitive to the Cooper pairs condensation include the antinodal kink of BB phonon by ARPES [48], and ….'. In the ref. 48, they reported that the B1g phonon is coupled to the single particle excitation spectrum which appear to be strongest near the anti-nodal region, but not A1g mode. The B1g mode is also a bond-buckling phonon, but Ox and Oy motions are out-of-phase and different from A1g symmetry. So, it's confusing that reader gets impression that the 'BB phonon' the authors refer to is the same as those in the ref. 48, but they are different.

Our answer: We appreciate the reviewer's comment regarding distinguishing between A_{1g} and B_{1g} modes. As pointed out by Ref. 26, the Cu L_3-edge RIXS is more sensitive to the EPC strength of the A_{1g} mode compared to the B_{1g} mode due to symmetry effects. In contrast, ARPES selectively detects the B_{1g} mode of the bond-buckling phonon. Therefore, we agree that making a direct link between ARPES and RIXS is inaccurate, and we have removed the related discussion from our revised manuscript.

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

In the revised manuscript, Zou et al. have made substantial modifications in response to the comments provided by myself and the other two referees. Some comments collectively addressed similar issues. I am pleased to note that the changes made by the authors are satisfactory, and the paper has now reached its zenith in terms of quality. This work improves our understanding of charge order in cuprates and its interaction with superconductivity. Therefore, I endorse the publication of this work in Nat Commun.

Reviewer #2 (Remarks to the Author):

Following the comments of all three reviewers, Zou et al. have substantially improved their manuscript. For example, the introduction is much clearer and more easy to follow.

Nevertheless, my main concern that the main conclusion of the paper is not supported by the data collected by the author remains. I understand that, as highlighted by the authors in their reply, the concept of pre-formed Cooper pairs has been suggested by other studies. However, the authors fail to establish how such putative pre-formed Cooper pairs would emerge from the CO state. The remaining conclusion that charge order and phonons are coupled have been already observed in other underdoped curates and as such are not novel. Further, reading through the reports by the other referees and the associated replies by the authors, it seems that similarly not all concerns by my fellow referees were entirely addressed.

Therefore, I cannot recommend publication in Nature Communications.

Reviewer #3 (Remarks to the Author):

I am still concerned about a presentation of the STM data, specifically the dI/dV modulations that are regarded as a signature of the charge order. As I pointed out in my comments, low energy dI/dV modulations for the underdoped Bi2212 suffer from the systematic error due to the setup effect. While I appreciate answers to my comments, I respectfully disagree with the authors. The authors must realize that there is a systematic error in the dI/dV modulations for these dopings due to the setup conditions of the STM tip-sample junction formation. All the references [4, 7, 8, 10, 21, 38] don't consider the fact that the so-called bi-directional 'checkerboard' CO seen in dI/dV(r,E) near E=±30 meV is strongly setup dependent.

Let me bring up the thesis by A. Schmidt, Ph. D. Thesis, p. 21, Cornell University (2009) from Davis group once again, in which a systematic study of the setup effect in dI/dV for the underdoped Dy-Bi2212 (Tc=45K) has been presented. Below is a figure copied from the Figure 1.7 in this thesis, showing the topograph, dI/dV(r,-25mV), and Z(r,-25mV) measured at different setpoint bias voltages V0 from 50 to 150mV. In dI/dV(r,-25mV), so-called bi-directional 'checkerboard' CO is obvious for V0=150mV, however, with decreasing V0, the bi-directional 'checkerboard' CO modulation is diminished and is not obvious for V0=50mV. This demonstrates that the bi-directional 'checkerboard' CO is strongly setup dependent, and the dI/dV(r, ~30mV) is not a good observable for a charge order. It is also the fact that the Bogoliubov QPI modulations are scrambled with them. Particularly, the "q1" scattering wavevector of the Bogoliubov QPI is strong and is similar to the one for the CO (Y. Kohsaka et al., Nature 454, 1072 (2008)). Thus, I don't think the authors can use dI/dV(r, ~30mV) as a representative signature of the CO for the underdoped cuprates, and a utilization of the dI/dV(r,~30mV) leads to erroneous conclusions. I don't think that I can accept claims made by ambiguous observables in the manuscript. Because of these reasons, I don't recommend for a publication in Nature Communications.

Copied from Figure 1.7 of the Seamus Davis group thesis, A. Schmidt, Ph. D. Thesis, p. 21, Cornell University (2009)

Additional comments are as follows.

1. While an empirical formular of the Tc(p) can be accepted at some degree, the maximum Tc strongly depends on how materials are disordered. In fact, the maximum Tc of the Bi2212 reported so far is 98K (H. Hobou et al., Phys. Rev. B 79, 064507 (2009)). I also think that the maximum Tc for the Dy-Bi2212 can be different.

2. There is a paper by Z. Du et al., Phys. Rev. X 13, 021025 (2023) that has been published quite recently. They have studied an atomic displacement associated with the cuprate CO using the STM and provided a link to the phonon anomalies observed in the RIXS. They observed that atoms are displaced as the same symmetry as the bond stretching mode for □□0 at the CO wavevector. Their results seem to be highly consistent with those from the Stanford group (H. Lu et al., Phys. Rev. B 106, 155109 (2022)). I think this paper is relevant to the present manuscript and the authors should cite it.

-In this letter, we provide a point-to-point response to the reviewer's comments.

The reviewer's original comments are shown in blue italic characters in the following.

Black regular characters show the authors' responses.

Reviewer #1 (Remarks to the Author):

In the revised manuscript, Zou et al. have made substantial modifications in response to the comments provided by myself and the other two referees. Some comments collectively addressed similar issues. I am pleased to note that the changes made by the authors are satisfactory, and the paper has now reached its zenith in terms of quality. This work improves our understanding of charge order in cuprates and its interaction with superconductivity. Therefore, I endorse the publication of this work in Nat Commun.

Our answer: We thank the reviewer for recommending the publication of our work in Nature Communications. The quality of our work has been significantly improved thanks to the constructive suggestions from all the reviewers.

Reviewer #2 (Remarks to the Author):

Following the comments of all three reviewers, Zou et al. have substantially improved their manuscript. For example, the introduction is much clearer and more easy to follow.

Our answer: We thank the reviewer for reviewing our manuscript again and recognizing the significant improvement of our manuscript.

Nevertheless, my main concern that the main conclusion of the paper is not supported by the data collected by the author remains. I understand that, as highlighted by the authors in their reply, the concept of pre-formed Cooper pairs has been suggested by other studies. However, the authors fail to establish how such putative pre-formed Cooper pairs would emerge from the CO state. The remaining conclusion that charge order and phonons are coupled have been already observed in other underdoped curates and as such are not novel. Further, reading through the reports by the other referees and the associated replies by the authors, it seems that similarly not all concerns by my fellow referees were entirely addressed.

Therefore, I cannot recommend publication in Nature Communications.

Our answer: We thank the reviewer for their further comments, and we are committed to addressing these remaining concerns to enhance the quality of our manuscript.

Firstly, we have established a compelling connection between pre-formed Cooper pairs and CO, both preceding the condensation of Cooper pairs within the superconducting dome and extending beyond it. This is supported by a body of evidence, including references [39, 40, 41, 42] and ref. [38], which elucidate the existence of incoherent Cooper pairs at temperatures above T_c and across a doping range that extends into the insulating regime, where checkerboard CO is present, as demonstrated by our combined STM and RIXS investigation. These findings suggest that a charge-ordered electronic state is energetically favorable for the formation of Cooper pairs, akin to the description provided by the stripe model (e.g., J. Tranquada et al., J. Phys. Soc. Jpn. **90**, 111002 (2021)). This lowest-energy state suggests that two holes are distributed on the peak of the checkerboard plaquette, as visualized by a periodical superconducting coherence peak modulation against the CO (see ref. [38] and W. Ruan et al., Nat. Phys. **14**, 1178 (2018)).

Secondly, in contrast to previous reports, our research reveals that the bond-buckling, rather than the bond-stretching phonon, is intimately coupled to the underlying CO and plays a pivotal role in the superconductivity. The behavior of the stretching phonon aligns well with noninteracting model calculations. Furthermore, our findings offer novel and significant insights into high-T_c cuprate superconductors. Notably, we have extended the presence of CO into the antiferromagnetic insulating regime ($p = 0.04$), which is the lowest doping level among all cuprates, even in the absence of a Fermi surface. This finding disqualifies the Fermi-surface nesting scenario as the origin of charge order and suggests that Cooper pairs emerge from the CO phase, diverging from a simplistic competing scenario. Additionally, we demonstrate, for the first time, that CO in the insulating phase exhibits a bulk nature and pervades the cuprate phase diagram down to $p = 0.04$ and at temperatures exceeding $T = 273$ K.

Lastly, beyond the reviewer's comments, we have effectively addressed all the concerns raised by reviewer #1, leading to their endorsement of our manuscript for publication in Nature Communications. Reviewer #3 has one remaining concern pertaining to our STM methodology for visualizing CO and has made two additional suggestions, which we believe we have addressed comprehensively in our response.

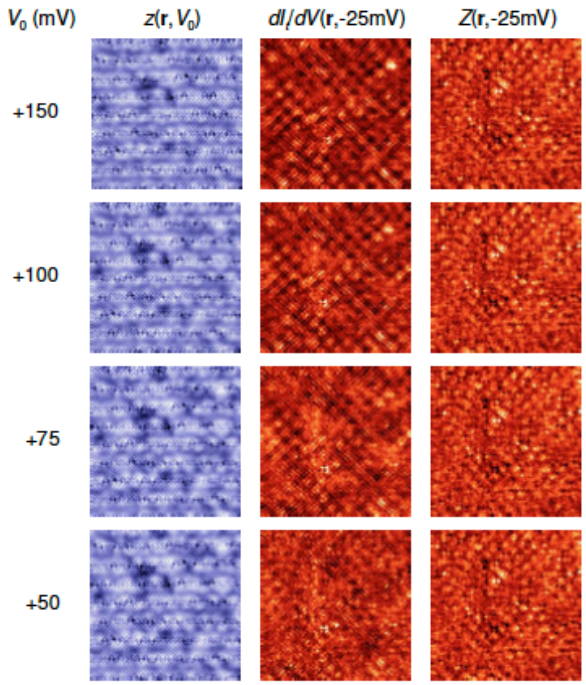
Changes made: To make the connection between pre-formed Cooper pairs and CO state clearer, we have added some discussion in the main text (lines 347-355 marked in red).

Reviewer #3 (Remarks to the Author):

I am still concerned about a presentation of the STM data, specifically the dI/dV modulations that are regarded as a signature of the charge order. As I pointed out in my comments, low energy dI/dV modulations for the underdoped Bi2212 suffer from the systematic error due to the setup

effect. While I appreciate answers to my comments, I respectfully disagree with the authors. The authors must realize that there is a systematic error in the dI/dV modulations for these dopings due to the setup conditions of the STM tip-sample junction formation. All the references [4, 7, 8, 10, 21, 38] don't consider the fact that the so-called bi-directional 'checkerboard' CO seen in dI/dV(r,E) near E=±30 meV is strongly setup dependent.

Let me bring up the thesis by A. Schmidt, Ph. D. Thesis, p. 21, Cornell University (2009) from Davis group once again, in which a systematic study of the setup effect in dI/dV for the underdoped Dy-Bi2212 (Tc=45K) has been presented. Below is a figure copied from the Figure 1.7 in this thesis, showing the topograph, dI/dV(r,-25mV), and Z(r,-25mV) measured at different setpoint bias voltages V0 from 50 to 150mV. In dI/dV(r,-25mV), so-called bi-directional 'checkerboard' CO is obvious for V0=150mV, however, with decreasing V0, the bi-directional 'checkerboard' CO modulation is diminished and is not obvious for V0=50mV. This demonstrates that the bi-directional 'checkerboard' CO is strongly setup dependent, and the dI/dV(r, ~30mV) is not a good observable for a charge order. It is also the fact that the Bogoliubov QPI modulations are scrambled with them. Particularly, the "q1" scattering wavevector of the Bogoliubov QPI is strong and is similar to the one for the CO (Y. Kohsaka et al., Nature 454, 1072 (2008)). Thus, I don't think the authors can use dI/dV(r, ~30mV) as a representative signature of the CO for the underdoped cuprates, and a utilization of the dI/dV(r,~30mV) leads to erroneous conclusions. I don't think that I can accept claims made by ambiguous observables in the manuscript. Because of these reasons, I don't recommend for a publication in Nature Communications.



Copied from Figure 1.7 of the Seamus Davis group thesis, A. Schmidt, Ph. D. Thesis, p. 21, Cornell University (2009).

Our answer: We thank the reviewer for their careful reevaluation of our manuscript. The reviewer concludes that the dI/dV cannot be regarded as a signature of the charge order given its significant variation from V_0 = 50 mV to 150 mV. We respectfully disagree. As demonstrated in our prior response, we acknowledge the influence of setup conditions on the dI/dV signal, a well-recognized aspect within the STM community.

In fact, it is well-known that if the setup voltage (V_0) is too low, the dI/dV signal and the related electronic states will be strongly coupled with the setup condition (as explained in our previous response letter). Therefore, it is such an obvious thing that the CO will depend on the setup voltage if the V_0 is as small as 50 mV, and this is precisely why we use higher setup voltages. To expound upon our viewpoint further, we introduce Figure R1 to provide a more comprehensive clarification. In Panel (a), we present a reference figure from S. Mukhopadhyay *et al.*, PNAS **116**, 13249 (2019). This figure pertains to a Bi2212 sample with T_c = 45 K, as referenced by the reviewer, with an estimated doping level of approximately p ~0.08, calculated from the empirical formula (ref. [48]) $T_c/T_c^{max} = 1 - 82.6 * (p - 0.16)^2$ with T_c^{max} = 96 K. Panel (a) depicts an average gap of $\Delta \sim 75$ meV for this specific doping. Using a d-wave spectral shape with Δ = 75 meV, we proceed to compare the dI/dV spectra at different V_0 in Panels (b) and (c), which simulate and visualize the set-point effect. We focus on the dI/dV (V=30 mV), where the charge order signal predominates. It becomes evident that when V_0 surpasses 100 mV, the spectral weight closely resembles the standard one, free from set-point influence. This observation elucidates why the charge order signal exhibits minimal variation when V_0 is increased beyond 100 mV for an underdoped Bi2212 with $\Delta \sim 75$ meV, as presented in Figure 1.7 of Schmidt's Ph.D. Thesis. In our own study, as detailed in the Methods section (lines 466-472), we took measures to further mitigate the set-point effect by applying V_0 = -0.3 V for the p = 0.06 sample and V_0 = -0.5 V for the p = 0.04 sample.

Therefore, the dI/dV map can be reliably utilized for characterizing the CO, provided that appropriate measures are taken to mitigate the set-point effect. This method has found widespread application in the study of charge order in cuprates, as evidenced by publications such as Science **343**, 393 (2014), Nature **575**, 156 (2019), Nature Physics **19**, 1301 (2023), and others. Furthermore, Davis's group has also employed the dI/dV map to reveal charge order in $Ca_{2-x}Na_xCuO_2Cl_2$ [Nature **430**, 1001 (2004)] and $Bi_2Sr_2CaCu_2O_{8+\delta}$ [Phys. Rev. Lett. **94**, 197005 (2005)].

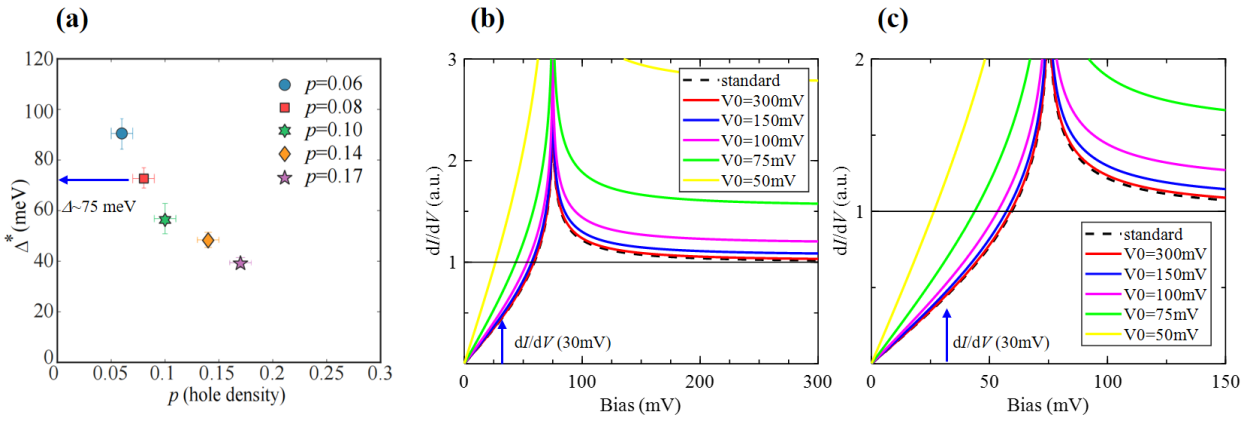


Figure R1: Demonstrating the influence of setpoint effect for underdoped cuprates. (a) To directly compare with the reference given by the reviewer, we show the spatially averaged gap values of differently doped Bi2212 samples, a figure adapted from S. Mukhopadhyay *et al.*, PNAS **116**, 13249 (2019). (b) Simulated spectra for a d-wave gap $\Delta = 75$ meV. The colors represent different set-point bias voltages (V_0). It can be seen the dI/dV background overlaps with $V_0 > 100$ mV. The blue arrow indicates the interested dI/dV spectral weight at $V = 30$ meV, which characterizes the CO signal. (c) Same as (b), but zoomed in for a better observation.

Changes made: To clarify this point, we have explained why we use high set-point bias voltages (V_0) in methods in lines 467-473.

Additional comments are as follows.

1. *While an empirical formular of the Tc(p) can be accepted at some degree, the maximum Tc strongly depends on how materials are disordered. In fact, the maximum Tc of the Bi2212 reported so far is 98K (H. Hobou et al., Phys. Rev. B 79, 064507 (2009)). I also think that the maximum Tc for the Dy-Bi2212 can be different.*

Our answer: We agree with the reviewer that the maximal T_c for Dy-Bi2212 can be different. However, the precise value of the maximum T_c for Dy-Bi2212 is unavailable due to the difficulty in synthesizing optimally Dy-doped Bi2212 samples, so we use the T_c^{max} of 96 K taken from H. Eisaki et al., Phys. Rev. B 69, 064512 (2004). It is important to note that the estimated doping level exhibits only minimal variation when different T_c^{max} values are considered. For instance, in the case of Bi2212 with $T_c = 20$ K, the calculated doping level is approximately $p = 0.0619$ for $T_c^{max} = 98$ K, $p = 0.0621$ for $T_c^{max} = 96$ K, and $p = 0.0628$ for $T_c^{max} = 91$ K. The essential finding of our work lies in two dopings across the onset of the superconducting dome (p=0.05): one is insulating, and the other is superconducting with $T_c = 20$ K. The precise value of T_c^{max} does not alter this fundamental aspect and does not affect our conclusions.

Changes made: We have added the above argument into supplementary section 1 to clarify the issue of maximum T_c.

2. *There is a paper by Z. Du et al., Phys. Rev. X 13, 021025 (2023) that has been published quite recently. They have studied an atomic displacement associated with the cuprate CO using the STM and provided a link to the phonon anomalies observed in the RIXS. They observed that atoms are displaced as the same symmetry as the bond stretching mode for $\omega \rightarrow 0$ at the CO wavevector. Their results seem to be highly consistent with those from the Stanford group (H. Lu et al., Phys. Rev. B 106, 155109 (2022)). I think this paper is relevant to the present manuscript and the authors should cite it.*

Our answer: We appreciate the reviewer's recommendation of this new paper. After a thorough review, we acknowledge that while the paper by Z. Du *et al.* does touch on the subject of CO-related atomic displacements, their research emphasis and investigated aspects substantially differ from the primary focus of our manuscript. Our study focuses on understanding the formation of Cooper pairs close to the insulating regime and relationship with electron-phonon coupling and charge order. We believe that the Stanford group's work by H. Lu *et al.*, "Phys. Rev. B 106, 155109 (2022)," which we have already cited, provides a more direct and relevant comparison to our research. Therefore, we respectfully choose not to include the new paper in our reference list to maintain focus on our research goals.

Reviewers' comments:

Reviewer #3 (Remarks to the Author):

I appreciate the authors for the response to my comments. My new comments are highlighted in green below.

The reviewer's original comments are shown in blue italic characters in the following.

Black regular characters show the authors' responses.

Reviewer #3 (Remarks to the Author):

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CO (Y. Kohsaka et al., Nature 454, 1072 (2008)). Thus, I don't think the authors can use dI/dV(r, ~30mV) as a representative signature of the CO for the underdoped cuprates, and a utilization of the dI/dV(r,~30mV) leads to erroneous conclusions. I don't think that I can accept claims made by ambiguous observables in the manuscript. Because of these reasons, I don't recommend for a publication in Nature Communications.

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I respectfully disagree with the authors' comments above. The figure 1.7 from Schmidt's thesis is a robust experimental demonstration that the low energy CO modulations, which is apparently seen around 30mV, is originated from those at relatively high energies above 50mV. Y. Kohsaka et al, Nature 454, 1072 (2008) has identified and demonstrated in their paper that an intrinsic energy scale of the CO is the pseudogap energy scale Δ, which is about 75mV for p~0.06.

An apparent low energy CO modulation appears in dI/dV because, since dI/dV is given by

$dI/dV(r,E) \sim (eI_0\, N(r,E))/(\int_0^{eV_0} N(r,E)dE)$

where I0 is the setup current and V0 is the setup bias voltage, a dI/dV at $E \neq \Delta$ can be extrinsically modulated through the denominator if a local density of states N(r,E) is strongly modulated at E~Δ(=75mV). For V0=50mV, the setup bias voltage is smaller than the pseudogap energy scale Δ, and thus spatial modulations in the integrated density of states (denominator) become negligible. Indeed, as demonstrated in Fig. 1.7 of Schmidt's thesis, the CO modulations in dI/dV at E=25mV are virtually absent for V0=50mV. However, when V0 is higher than Δ, then the denominator is modulated since N(r,E~Δ) strongly modulated, causing the erroneous modulations in dI/dV at the different energies away from Δ. The authors (and Science 343, 393 (2014), Nature 575, 156 (2019). I don't find this paper: Nature Physics 19, 1301 (2023)) only consider the situation for V0>Δ.

As far as I know, the setpoint effect for the CO was realized around 2006-2007. The papers Ca2-xNaxCuO2Cl2 [Nature 430, 1001 (2004)] and Bi2Sr2CaCu2O8+δ [Phys. Rev. Lett. 94, 197005 (2005)] were published before the setpoint effect was realized.

To conclude, the CO in dI/dV(r,30mV) measured with V0>Δ is erroneous and doesn't provide a correct physics. I don't accept the authors' rebuttals, and thus I don't recommend for a publication.

-This rebuttal letter provides a point-to-point response to the reviewer's comments.

In the following, the reviewer's original comments are shown by blue italic characters.

The authors' responses are shown by normal black characters.

Reviewer #3 (Remarks to the Author):

I respectfully disagree with the authors' comments above. The figure 1.7 from Schmidt's thesis is a robust experimental demonstration that the low energy CO modulations, which is apparently seen around 30mV, is originated from those at relatively high energies above 50mV. Y. Kohsaka et al, Nature 454, 1072 (2008) has identified and demonstrated in their paper that an intrinsic energy scale of the CO is the pseudogap energy scale Δ, which is about 75mV for p~0.06.

Our answers: We express our deep concern regarding the evaluation provided by the referee as they have wrongly ascribed Schmidt's d*I*/d*V* modulation to that above 50 mV. As we have previously explained, the only "high energy" electronic state reported is a uni-directional modulation such as the paper Y. Kohsaka *et al.*, Nature 454, 1072 (2008). This electronic state is a different state from the bi-directional CO, and Y. Kohsaka *et al*. have never assigned it to a bi-directional CO. Therefore, assuming that a low-energy bi-directional CO "originated from" a high-energy uni-directional modulation is logically wrong. The high-energy modulation was actually revealed by Z-map, and we suspect that the referee's preference towards the Z-map method may have influenced their evaluation of our work.

An apparent low energy CO modulation appears in dI/dV because, since dI/dV is given by

$$\frac{dI}{dV}(r,E) \sim \frac{eI_0\, N(r,E)}{\int_0^{eV_0} N(r,E)dE}$$

where I_0 is the setup current and V_0 is the setup bias voltage, a dI/dV at $E \neq \Delta$ can be extrinsically modulated through the denominator if a local density of states N(r,E) is strongly modulated at E~Δ (=75mV). For V_0=50mV, the setup bias voltage is smaller than the pseudogap energy scale Δ, and thus spatial modulations in the integrated density of states (denominator) become negligible. Indeed, as demonstrated in Fig. 1.7 of Schmidt's thesis, the CO modulations in dI/dV at E=25mV are virtually absent for V_0=50mV. However, when V_0 is higher than Δ, then the denominator is modulated since N(r,E~Δ) strongly modulated, causing the erroneous modulations in dI/dV at the different energies away from Δ. The authors (and Science 343, 393 (2014), Nature 575, 156 (2019). I don't find this paper: Nature Physics 19, 1301 (2023)) only consider the situation for V_0>Δ.

Our answers: The referee has made a substantial error in understanding the set-point effect of STM. Different from the society's common wisdom, the referee insists that the setup bias voltage (V_0) should be as low as possible to avoid the set-point effect, which is technically wrong. To illustrate this point, let's bring back the formula referred to by the referee:

$$dI/dV(r,E) = eI_0N(r,E)/\int_0^{eV_0} N(r,E)dE$$

where I_0 and V_0 are the set-point of STM experiment. The set-point effect becomes relevant when

dI/dV(r, E) deviates from the local DOS N(r, E) due to a spatial modulation of the denominator. The denominator is determined by an integral of N(r, E) over the bias voltage from 0 to V_0, not the N(r, E=Δ) value. The referee supposes that a modulation of N(r, E~Δ) will modulate the integral to a similar degree with that in the charge order signal, which is completely wrong. As we have demonstrated, a lower V_0 will induce a stronger dI/dV set-point artifact. At eV_0 much higher than Δ (where the coherence peak locates), the integral will involve the DOS over a broad energy range outside the gap, and the spatially modulated N(r, E~Δ) only affects a negligible energy range in the integral. This is why the common wisdom is to use a high V_0 to mitigate the set-point effect. Indeed, as shown in Fig. R1 in our previous response to Referee #3, set bias V_0 = -100 mV is enough to eliminate the set-point effect in the DOS map measurement.

To make this point more visible, in the figure below we show three dI/dV maps at 30 mV of an underdoped Bi2201 cuprate with different set-point bias, the bi-directional CO signal remains robust with V_0 beyond -100 mV.

The referee mentioned that they could not locate the paper "Nature Physics 19, 1301 (2023)". In response, we would like to point out that this paper, authored by some of our team members, employs the same dI/dV methodology as our manuscript to uncover charge order in cuprates. It is available at https://www.nature.com/articles/s41567-023-02100-9.

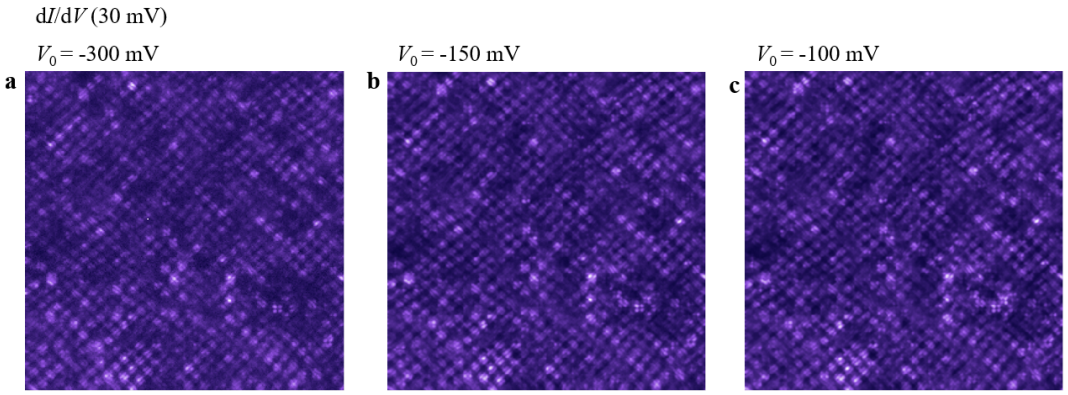

Fig. R1. The CO pattern of an underdoped Bi-2201 with T_c = 10 K with different set bias voltages. a-c, The dI/dV maps at different set bias V_0 with -300 mV, -150 mV and -100 mV, respectively.

As far as I know, the setpoint effect for the CO was realized around 2006-2007. The papers $Ca_{2-x}Na_xCuO_2Cl_2$ [Nature 430, 1001 (2004)] and $Bi_2Sr_2CaCu_2O_{8+\delta}$ [Phys. Rev. Lett. 94, 197005 (2005)] were published before the setpoint effect was realized.

Our answers: Numerous recent STM studies have investigated charge order (CO) using dI/dV after recognizing the setpoint effect, as evidenced by a wealth of publications (e.g., Science 343, 393 (2014); Nature Physics 12, 1047 (2016); Nature Physics 14, 1178 (2018); Nature 575, 156 (2019); Nat. Commun. 12, 1 (2021); Nature Physics 19, 1301 (2023)). This widespread application demonstrates the validity and acceptance of using dI/dV in investigating charge orders in the STM community.

To conclude, the CO in dI/dV(r,30mV) measured with V_0>Δ is erroneous and doesn't provide

a correct physics. I don't accept the authors' rebuttals, and thus I don't recommend for a publication.

Our answers: Referee #3 has recognized the novelty and quality of our resonant inelastic x-ray scattering (RIXS) results. The strong agreement between the charge orders observed in our RIXS and STM studies reinforces the validity of using the dI/dV method to observe CO in cuprates and the accuracy of our results. Our STM methodology, which includes using a higher set bias voltage to mitigate the set-point effect and the adequacy of the dI/dV signal in investigating charge order (CO) physics, is sound. We have addressed the reviewer's concerns and provided evidence supporting the accuracy of our results.

Our study reveals the incipient charge order in strongly correlated insulating cuprates and demonstrates that Cooper pairs emerge from a charge-ordered insulating state and condense with an enhanced interplay between charge excitations and electron-phonon coupling. These findings significantly contribute to our understanding of how superconductivity emerges at the onset of the superconducting dome. Therefore, we respectfully request reconsideration of our manuscript for publication in Nature Communications.

REVIEWER COMMENTS

Reviewer #4 (Remarks to the Author):

I have read the rebuttal carefully several times: Reviewer #3 technical statements and references thereto are scientifically and historically correct, in my opinion.

Reviewer #5 (Remarks to the Author):

Referee #3 has a point in the sense that it is, as a matter of principle, impossible to completely eliminate STM setpoint effects in dI/dV mapping. But it is ludicrous to claim that the charge order observed in this study is purely a setpoint effect: the charge order has been reported in many STM studies of cuprates across various doping levels. In this particular case, the existence of the charge order is corroborated by RIXS measurements on the same samples. In my opinion, the evidence of charge order presented in this work is sufficient to support the conclusions.

That said, the setpoint effect is a very complicated issue that needs to be treated very carefully. The charge order in superconducting Bi-2212 mainly resides within the energy window of 0-50 meV (see, for example, Science 343, 395 (2014) mentioned by the referee). It is therefore not surprising that the charge order at -25 meV, as shown in in Fig. 1.7 of Schmidt's thesis, is most prominent when the setpoint bias is set at large positive values beyond 50 meV. A lower positive setpoint, as suggested by the referee, will not solve the setpoint issue completely, because the setpoint is still within the charge order energy window. Viewed from this perspective, a negative setpoint bias would be optimal to mitigate the setpoint effect. I notice that a negative setpoint bias (-300 meV) was indeed used in this work. It is however possible that the charge order in insulating Bi-2212 behaves differently from that in superconducting Bi-2212. To further eliminate potential setpoint effect in insulating Bi-2212, the authors should consider presenting a bias-dependent FFT map similar to Fig. 2 in Science 343, 395 (2014), or simply a current map at the negative setpoint bias, to show that there are no spectroscopic features at the negative bias to interfere with the dI/dV mapping of the charge order.

-This rebuttal letter provides a point-to-point response to the reviewer's comments.

In the following, the reviewer's original comments are shown by blue italic characters.

The authors' responses are shown by normal black characters.

Reviewer #4 (Remarks to the Author):

I have read the rebuttal carefully several times: Reviewer #3 technical statements and references thereto are scientifically and historically correct, in my opinion.

Our answers: We thank the reviewer for carefully considering our rebuttals. As we emphasized in our reply, although we agree that Reviewer #3's references concerning pseudogap and broken symmetry physics are correct, their critique of the dI/dV map and misunderstanding of the STM set-point effect are technically wrong. This has been corroborated by Reviewer #5, who has pointed out the inaccuracies in Reviewer #3's assessment. Specifically, Reviewer #3's argument does not accurately reflect the established understanding and application of STM set-point biases in our context. The dI/dV mapping technique we employed is well-supported by extensive literature and is a standard method for visualizing charge orders and greatly deepening our knowledge of strong correlated charge order.

Reviewer #5 (Remarks to the Author):

Referee #3 has a point in the sense that it is, as a matter of principle, impossible to completely eliminate STM setpoint effects in dI/dV mapping. But it is ludicrous to claim that the charge order observed in this study is purely a setpoint effect: the charge order has been reported in many STM studies of cuprates across various doping levels. In this particular case, the existence of the charge order is corroborated by RIXS measurements on the same samples. In my opinion, the evidence of charge order presented in this work is sufficient to support the conclusions.

Our answers: We are grateful for the reviewer's insightful comments and clarity, which align with our perspective and substantiate the reliability of our observations. The reviewer is correct that charge order has been consistently reported in numerous STM studies, and the agreement between our STM and RIXS results further reinforces the validity of our conclusions.

That said, the setpoint effect is a very complicated issue that needs to be treated very carefully. The charge order in superconducting Bi-2212 mainly resides within the energy window of 0-50 meV (see, for example, Science 343, 395 (2014) mentioned by the referee). It is therefore not surprising that the charge order at -25 meV, as shown in in Fig. 1.7 of Schmidt's thesis, is most prominent when the setpoint bias is set at large positive values beyond 50 meV. A lower positive setpoint, as suggested by the referee, will not solve the setpoint issue completely, because the setpoint is still within the charge order energy window. Viewed from this perspective, a negative setpoint bias would be optimal to mitigate the setpoint effect. I notice that a negative setpoint bias (-300 meV) was indeed used in this work. It is however possible that the charge order in insulating Bi-2212 behaves differently from that in superconducting Bi-2212. To further eliminate potential

setpoint effect in insulating Bi-2212, the authors should consider presenting a bias-dependent FFT map similar to Fig. 2 in Science 343, 395 (2014), or simply a current map at the negative setpoint bias, to show that there are no spectroscopic features at the negative bias to interfere with the dI/dV mapping of the charge order.

Our answers: Due to the same reasons raised by the reviewer, a negative set-point bias voltage (-300 mV) is applied to mitigate the set-point effect (see Methods of our manuscript). In response to the reviewer's suggestion, we have included a bias-dependent FFT plot along the lattice direction (Fig. R1B, R1D) on the p = 0.04 and 0.06 samples, similar to the one presented in Science 343, 393 (2014) (Fig. R1a). Consistent with the reference, our study shows that the charge order is prominent within the positive energy window (Fig. R1b,c). We note that the CO signal is quantitatively weaker in the p = 0.04 sample, as is also seen in the real-space (Fig. 2f) and momentum-space (Fig. S4A) maps. In our experiment, we have used set-point conditions of -500 mV for the p = 0.04 sample and -300 mV for p = 0.06. As shown in Figs. R1d and R1e, the current map at the set-point bias is featureless, further disputing the claim of a set-point-induced phenomenon.

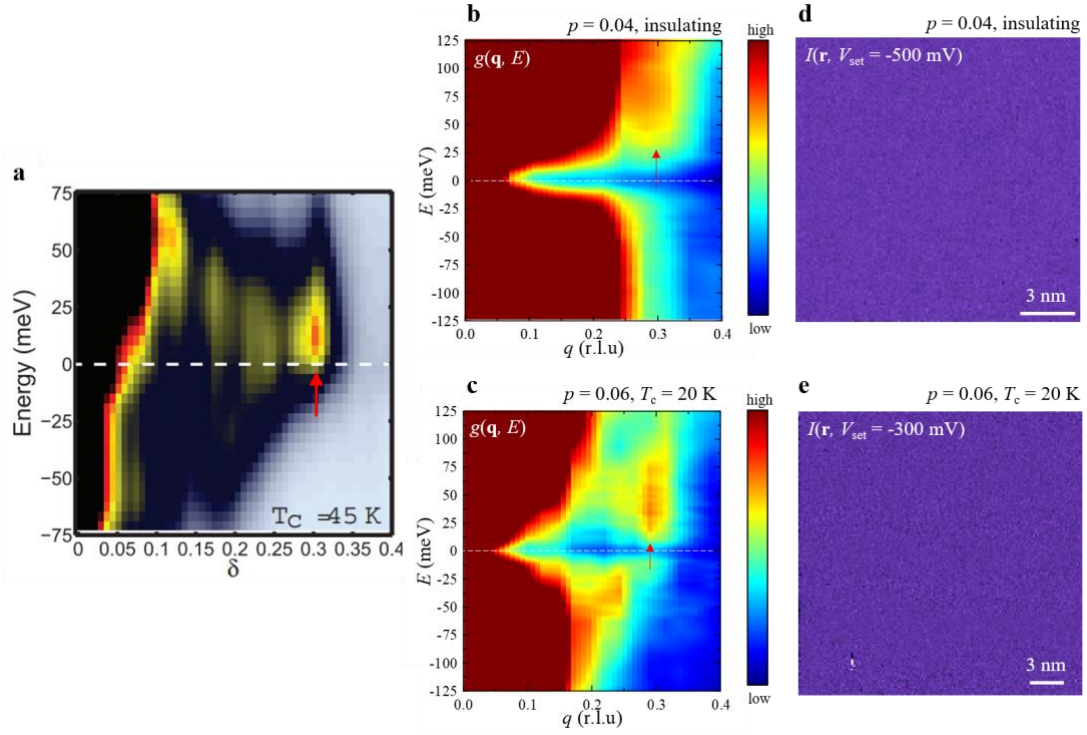

Fig.R1 (a) The energy-dependent FFT plot of an underdoped Bi-2212. The red arrow indicates the prominent charge order signal at the positive energy window. Figure adapted from Science 343, 393 (2014). (b, c) The same plots as (A) but for our p = 0.04 and p = 0.06 samples, displaying the charge order wavevector at $\boldsymbol{Q}$ ~ 0.3 r.l.u.. (d, e) The current map at set-point bias voltages on the p = 0.04 and p = 0.06 samples. The set-point effect is negligible as shown by the featureless map.

Change made: As suggested by Review #5, we include Fig. R1(b-e) in the Supplementary Materials, Fig. S5, to further illustrate that the set-point effect has been mitigated. We also add one paragraph in Supplementary Section 2 to explain the idea.

REVIEWERS' COMMENTS

Reviewer #5 (Remarks to the Author):

I am satisfied with the authors' reply, and I recommend the publication of the manuscript in Nature Communications.