

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The work discusses the influence of electron-phonon coupling on charge order in cuprates. Overall, I think the work is a worthwhile addition to Communications Physics provided that the points below are addressed.

Comments that I would like to see addressed:

1. The text tends to ignore extensive work on La-based cuprates that have rather similar CDW order, but the opposite trend in wavevector. I think that this needs to be addressed.
2. The text seems rather unbalanced about the value of some of the strongly correlated studies, including papers like these:
<https://journals.aps.org/prb/pdf/10.1103/PhysRevB.100.195141>
<https://journals.aps.org/prl/abstract/10.1103/PhysRevLett.113.046402>
These and other related (cited) studies do seem to predict the axial direction of the CDW (and they do so with simpler models, containing fewer assumptions). The text implies that they don't predict this. One difference is that the strongly correlated models do tend to predict wavevectors that increase or stay the same with doping. This is the fact that the current study addresses, but in doing so I think one must state that different cuprates vary a lot in this trend and that this issue likely relates to small changes in parameters. I think it's important to add that many of the strongly correlated models provide good theoretical evidence that different ordering vectors may be separated by very small amounts of energy:
<https://science.sciencemag.org/content/358/6367/1155>
3. I think the title is currently not really correct in terms of what the model shows. "Phonon-Influenced emergent charge order in the cuprates" or similar would be a more accurate reflection of what is argued.
4. Is it true to say that the main instability of the model is still at (π, π) ? It would be better to be more direct about this.
5. The b_{1g} phonon is a sensible phonon to choose to address, but it would be better to provide a clearer explanation of the reasoning. As far as I am aware, I do not think there is a strong empirical case that this mode softens more than other phonons. The text kind-of dances around this issue somewhat and might even imply that the b_{1g} shows especially pronounced softening. It would be better to be clearer and more direct.
6. I find the notation $g(q; k)$ strange. If the quantity is actually integrated over all suitable k_i , I would say the quantity is independent of k and just dependent on q .
7. It is hard to fully appreciate the numbers presented in Figure 2 in its current presentation. Can the colormaps be replotted with Sequential colormap including numerical values for the colorbar.

Some comments, which I would like to hear the authors views on:

8. To what extent could these issues be addressed by DFT, which could likely access the band structure and electron-phonon coupling?
9. Are there any concrete predictions of the current model?

Reviewer #2 (Remarks to the Author):

In this paper, the authors propose to ascribe the CDW charge ordering observed in high-T_c cuprates to a specific electron-phonon interaction. The work is technically sound and the idea is interesting and original.

However, as the authors themselves acknowledge, the results do not allow for a quantitative assessment of the phenomenon being described.

I will give an example: in order

to provide a convincing account of the charge ordering in cuprates, for instance, any proposal should allow for the determination of the boundaries of the region where CDW ordering occurs in the

T vs x phase diagram of these materials. How would the authors address this point?

In summary, this is a nice attempt to describe the charge ordering in cuprates. The technical part is well done. The results, however are not enough to convince the proposed mechanism is in fact responsible for the observed phenomenon.

Reviewer #3 (Remarks to the Author):

The paper addresses the question of the origin of the mysterious charge density wave (CDW) order that has been reported for practically all cuprates. The scenario analyzed in the paper is based on the electron-phonon coupling of the bond-buckling phonon. The authors write interaction Hamiltonian based on standard textbook theory and assume experimental electronic band dispersions. The results of this calculation are interesting to specialists. However, because the paper has many serious shortcomings, I do not recommend it for publication. Perhaps these can be remedied in a future version. Here is the list of areas that need reconsideration in more or less random order.

1. The electronic band structure in the calculations comes from a relatively old paper, Ref. 42. More recent and precise parameterization of experimental data was published that includes not only electronic dispersions but lifetime broadening as well (Li et al. Nat Commun 9, 26 (2018)). The calculation should be redone based on this result or perhaps other experimental work that includes lifetime broadening and bilayer splitting.
2. Work of Devereaux et al has shown that the buckling mode can have strong electron-phonon coupling only in multilayer cuprates. Single layer cuprates like do not show any electron-phonon effects for the buckling branch. There was a recent archive posting on Hg1201 for example showing this. How does this fit into the proposed picture? Are the authors claiming that single layer and multilayer cuprates have completely different CDW mechanisms? Or is Devereaux's work wrong?
3. What about bond-stretching phonons that have much stronger electron- coupling than the buckling phonons? The authors should discuss how these effects fit into their picture and why they think it is OK to ignore these experimental results.
4. The paper cites Ref. 25 that there is a CDW signature in the acoustic phonons of BSCCO. But a subsequent paper (Phys. Rev. B 100, 144502) showed that this is not a signature of a CDW. I recommend either not to cite Ref. 25 because it is not really necessary, or cite both papers.
5. DFT calculations that are also based on electronic structure do not get any phonon anomalies or CDW in cuprates. This is not clear from the paper at all except may be to narrow specialists. The authors should state clearly what they are doing that is different from DFT.
6. What about the anomalies seen by LeTacon group in the acoustic phonon? This should also be discussed.

The work discusses the influence of electron-phonon coupling on charge order in cuprates. Overall, I think the work is a worthwhile addition to Communications Physics provided that the points below are addressed.

We appreciate the reviewer's positive feedback on the value of our work. Below we address in detail the reviewer's questions, comments, and suggestions.

Comments that I would like to see addressed:

1. The text tends to ignore extensive work on La-based cuprates that have rather similar CDW order, but the opposite trend in wavevector. I think that this needs to be addressed.

It is correct that we focus primarily on the charge ordering physics in YBCO and Bi-2201. Rather than ignoring the vast amount of research on La-based cuprates, we take the view that the charge-stripe order in 214 cuprates reflects special, materials specific differences. It is not only the opposite doping trend of the CDW ordering wavevector, but also the appearance of charge and spin stripes. In particular, in the low-temperature tetragonal phase of rare earth doped LSCO and LBCO, the tilt pattern of the oxygen octahedra breaks in-plane xy-isotropy and thereby offers a source for a uniaxial density wave.

We have expanded the introduction in our manuscript to incorporate this so-far only implicit conjecture.

2. The text seems rather unbalanced about the value of some of the strongly correlated studies, including papers like these:

<https://journals.aps.org/prb/pdf/10.1103/PhysRevB.100.195141>

<https://journals.aps.org/prl/pdf/10.1103/PhysRevLett.113.046402>

These and other related (cited) studies do seem to predict the axial direction of the CDW (and they do so with simpler models, containing fewer assumptions). The text implies that they don't predict this. One difference is that the strongly correlated models do tend to predict wavevectors that increase or stay the same with doping. This is the fact that the current study addresses, but in doing so I think one must state that different cuprates vary a lot in this trend and that this issue likely relates to small changes in parameters. I think it's important to add that many of the strongly correlated models provide good theoretical evidence that different ordering vectors may be separated by very small amounts of energy:

<https://science.sciencemag.org/content/sci/358/6367/1155.full.pdf>

We maintain our assessment that - at present - purely electronic model calculations do not satisfactorily explain charge ordering tendencies with incommensurate uniaxial wavevectors. The two papers Ponsioen et al., Phys. Rev. B 100, 195141 (2019) and Corboz et al., Phys. Rev. Lett. 113, 046402 (2014) investigated - with high numerical accuracy - the competition of stripe states and a d-wave superconducting or pair-density wave state in the t-J or the Hubbard model. As is explicitly mentioned in these papers, their work addresses primarily the stripe physics in La-based cuprates. This is also reflected in their choice of rectangular supercells, which are by geometry biased towards stripe formation. Indeed, the remarkable finding in these papers is the near-degeneracy of the candidate phases. This becomes particularly evident in Zheng et al., Science 358, 1155 (2017).

As outlined above, we consider the stripe physics of 214 cuprates different from the incommensurate CDW formation in YBCO and Bi-2201. In that respect, the excellent papers cited above do not provide the missing clue for the structure of the charge order in the latter materials.

In the revised manuscript we have refined - accordingly - the sentence which addresses the purely electronic model calculations.

3. I think the title is currently not really correct in terms of what the model shows. “Phonon-Influenced emergent charge order in the cuprates” or similar would be a more accurate reflection of what is argued.

We acknowledge the reviewer's suggestion to adjust the title of our manuscript. In order to better reflect the essence of our proposal we have chosen the new title "Emergent Charge Order from Correlated Electron-Phonon Physics in Cuprates".

4. Is it true to say that the main instability of the model is still at (π,π) ? It would be better to be more direct about this.

It is correct that the electron-phonon model without quasiparticle renormalization selects a candidate instability wavevector near (π,π) . But even in this case is the the magnitude of the charge susceptibility not sufficiently large to allow for a finite mean-field transition temperature.

We have added a clarifying sentence.

5. The B_{1g} phonon is a sensible phonon to choose to address, but it would be better to provide a clearer explanation of the reasoning. As far as I am aware, I do not think there is a strong empirical case that this mode softens more than other phonons. The text kind-of dances around this issue somewhat and might even imply that the b_{1g} shows especially pronounced softening. It would be better to be clearer and more direct.

The reasoning behind our choice of selecting the B_{1g} bond-buckling phonon for a detailed analysis is twofold. First, to the best of our knowledge there exists only one complete experimental data set for the static lattice-distortion pattern in the CDW state. These data were taken on underdoped YBCO by Forgan et al. (Nat. Commun. 6, 10064 (2015)). The by-far largest displacements were reported for the in-plane oxygen atoms which move out-of-plane in a pattern that closely resembles the B_{1g} normal mode. Second, among the experimental papers that provide evidence for strong electron-phonon coupling and large phonon anomalies near the charge-ordering instability, only one or two track changes in phonon branches upon cooling. Importantly, the data published by Raichle et al. (Phys. Rev. Lett. 107, 177004 (2011)) demonstrates a temperature-dependent softening of the B_{1g} phonon at q_{CO} in stoichiometric, overdoped YBCO. This composition does not support CDW order, and the observed softening of about 6% at the CDW ordering wavevector of the overdoped compound should not be considered small, but rather support the expectation that the very same phonon should soften even more with the onset of CDW order in underdoped YBCO.

The data in the Forgan et al. paper show that also the heavier ions in YBCO distort out of their equilibrium positions in the charge homogeneous phase. This naturally suggests that also other low-energy phonons are involved in the CDW formation, and indeed softening of a low energy optical phonon was observed by Le Tacon (Nat. Phys. 10, 52 (2014)) and Kim et al. Science 362, 1040 (2018). While the softening of this mode does indeed indicate a coupling to the CDW (as must occur in the absence of special symmetries), the small static displacements of the Ba and Y atoms observed by Forgan et al. suggests that ultimately there is a relatively weak participation of this mode in the CDW formation.

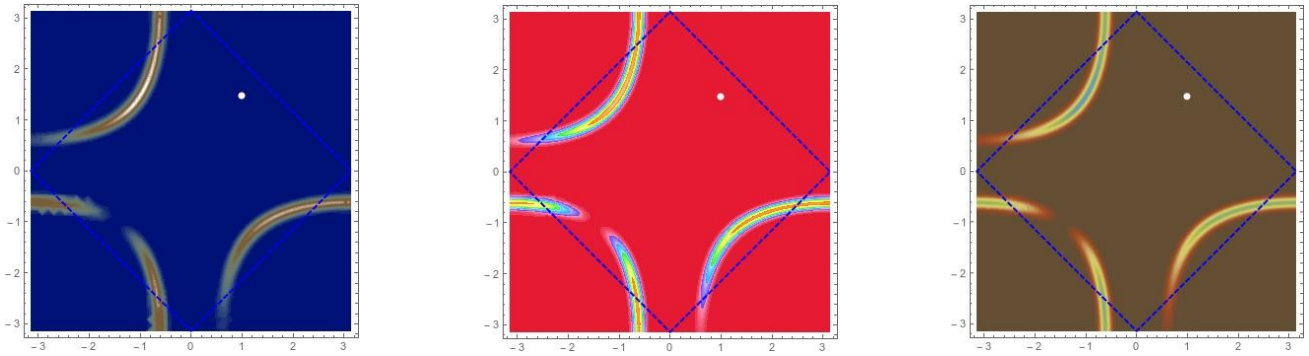
In the revised manuscript we have clarified the reasoning for our focus on the B_{1g} phonon along the lines explained above.

6. I find the notation $g(q;k)$ strange. If the quantity is actually integrated over all suitable k_i , I would say the quantity is independent of k and just dependent on q ?

The electron-phonon coupling necessarily depends on two momenta: k is the momentum of the electron and q is the momentum of the phonon by which the electron is scattered to a state with momentum $k+q$. The electron-phonon coupling $g(q,k)$ by itself is by no means integrated over k . We search for the global maximum with respect to both momenta: the initial momentum of an electron on the Fermi surface (referred to as k_i) and the phonon momentum q which connects k_i to the to the final scattered state with electron momentum also on the Fermi surface. The electron-phonon coupling $g(q,k)$ is only involved in an integral over k as part of the integrand for the evaluation of the susceptibility.

7. It is hard to fully appreciate the numbers presented in Figure 2 in its current presentation. Can the colormaps be replotted with Sequential colormap including numerical values for the colorbar.

As suggested by the reviewer, we have tried various other sequential color maps to test whether the visual impression of the variation of the electron-phonon coupling and the oxygen spectral function along the Fermi surface can be improved. This is a difficult task because both of these quantities vary very weakly along the Fermi surface. We have collected some of our attempts in the plots shown below. None of the versions can really be considered an improvement. We therefore stick to our previous color scheme. But we have added numerical values to the color bar as a replacement of the max/min legend. The maximum numbers in the contour plots Fig. 2a and Fig. 2b match the numbers in the x-y plots Fig. 2b and Fig. 2d, respectively. The dimensionless electron-phonon coupling in Fig. 2a is now in units of 0.01 and the spectral function in Fig. 2b is in units of $0.1/\text{eV}$.



The units for the color bars have been added to the figure caption.

Some comments, which I would like to hear the authors views on:

8. To what extent could these issues be address by DFT, which could likely access the band structure and electron-phonon coupling?

DFT can provide the electronic band structure, the dispersion of the phonon branches, and the electron phonon coupling. In our manuscript we start with a downfolded version of the DFT band structure. The weak dispersion of the optical B_{1g} phonon branch is neglected. Based on a microscopic ansatz we rederived the structure of the electron-phonon coupling to the B_{1g} mode as originally published more than 15 years ago in those papers which we cite in our manuscript. The unknown overall coupling strength is adopted from earlier fits to experiments.

The quasiparticle renormalization is a consequence of the strong electronic correlations in the copper-oxygen planes of cuprates and therefore not accessible by DFT. But it is the quasiparticle renormalization which is the essence of our proposal how the electron- B_{1g} phonon system may develop towards a combined charge and lattice instability. In this respect, a DFT based scheme cannot falsify or verify our proposal.

We have added clarifying sentences to address this point.

9. Are there any concrete predictions of the current model?

Since the submission of our manuscript we continued to think about consequences and predictions of our proposal. We have by now convinced ourselves that that the phonon based mechanism leads to a charge redistribution with predominantly s- symmetry form factor and a subdominant d-symmetry form factor. This means that also the Cu charge is sizeably modulated and the charge on the O_x and O_y atoms inside the unit cell is rather modulated out-of-phase than in-phase. If the charge pattern is only of the latter type on oxygen atoms it is referred to as a d-form factor CDW.

A d-form factor had been reported in early x-ray and STM experiments and subsequently taken for granted in

theoretical analyses. But this initial conclusion has recently been disputed by McMahon et al., arXiv:1904.12929. The resonant x-ray scattering data in the paper rather support a dominant s-wave form factor and are therefore compatible with a prediction from our phononic mechanism.

We have added such a remark to the final discussion chapter of our manuscript.

In this paper, the authors propose to ascribe the CDW charge ordering observed in high- T_c cuprates to a specific electron-phonon interaction. The work is technically sound and the idea is interesting and original. However, as the authors themselves acknowledge, the results do not allow for a quantitative assessment of the phenomenon being described.

We appreciate that the reviewer considers our proposal interesting and original, and we address his/her concerns as follows.

First, the key motivation for this work was the observation by Forgan et al that a B_{1g} -like distortion of the YBCO lattice accompanies CDW formation. Since the CDW necessarily couples to the B_{1g} mode, that experiment does not determine whether the lattice distortion is a consequence or a cause of the CDW instability. The main result of our work is to demonstrate that in a model with no free parameters, the B_{1g} phonon is a plausible mechanism for CDW formation.

Next, we wish to clarify the reviewer's impression that our results do not allow for a quantitative assessment of the CDW formation in cuprates. To start with, our calculations are not meant to offer a quantitative theory for an electronically correlated electron-phonon problem. We employ previously established parameters obtained from the literature and combine them with a phenomenological ansatz for the renormalized anisotropic quasiparticle weight supplemented by experimental (ARPES) input regarding the quasiparticle dispersion and specifically the nodal Fermi velocity.

We dare to claim it remarkable that the minimum input captures quantitatively the magnitude and doping dependence of the incommensurate charge ordering wavevector. The few per cent softening of the B_{1g} phonon may be considered weak, but it provides a magnitude which is compatible with neutron scattering data.

One may argue that a considerably larger charge susceptibility - as an outcome of our calculations - would strengthen the case for our proposed phonon based mechanism. Yet, any theory for the CDW in cuprates has to be reconciled with the prevailing fact that ARPES experiments fail to identify signatures of spectral changes or electronic reconstruction in the CDW phase. We conclude, these signals must be weak. The tendencies we find for a phonon based mechanism are indeed weak. And as we write in our manuscript, achieving moderate planar correlation lengths requires the nucleation by defects. The special role that defects or impurities may have in the CDW formation is supported by the analysis of NMR data (see Refs. [10] and [57]).

In the revised manuscript we have expanded our comments on the quantitatively weak CDW tendencies within the B_{1g} -phonon based proposal.

While we can offer calculations for the maximum $g(q,k)$ at different doping levels (see the plot below) or even use experimental data for the nodal Fermi velocity as input, the quantitative information - in particular with sufficient momentum space resolution - for the anisotropic quasiparticle weight renormalization at different doping levels is not available. The latter is a hard task by itself for correlated electron theory.

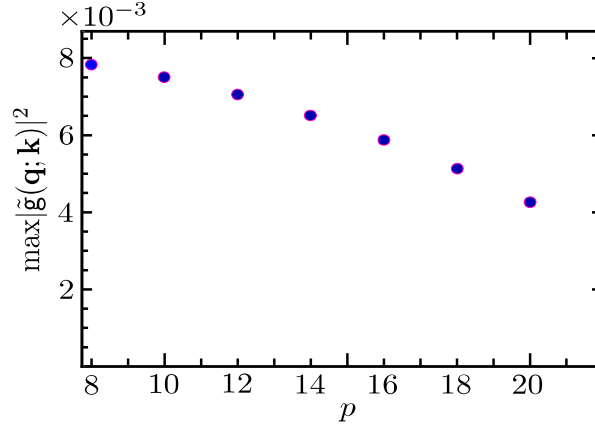
In the revised manuscript we have added an according sentence.

I will give an example: in order to provide a convincing account of the charge ordering in cuprates, for instance, any proposal should allow for the determination of the boundaries of the region where CDW ordering occurs in the T vs x phase diagram of these materials. How would the authors address this point?

The determination of phase boundaries of the CDW in the T vs. x phase diagram is beyond the scope of our current work, but we can comment on the relevant ingredients which determine the variation with respect to hole doping.

In our analysis it is the magnitude of the charge susceptibility at the anticipated ordering wavevector which signifies the tendency towards a combined lattice and charge ordering instability. This quantity is controlled by i) the magnitude of the electron-phonon coupling for initial and scattered electron momenta on the Fermi surface, ii) the nodal Fermi velocity, and iii) the quasiparticle weight at those Fermi surface points where $g(\mathbf{q}, \mathbf{k})$ achieves its largest values in the near nodal regions.

First, the global maximum value of $g(\mathbf{q}, \mathbf{k})$ on the doping dependent Fermi surfaces drops with hole doping, i.e. upon leaving the underdoped region towards the overdoped side. This is shown in the plot attached below. Simultaneously, the nodal Fermi velocity almost doubles (see Ref. [54]). Both of these trends reduce the charge susceptibility.



Our phenomenological ansatz for the quasiparticle weight along the Fermi surface is meant to reflect the Fermi arc formation in underdoped cuprates. In this ansatz, the quasiparticles are assumed intact in the near nodal region with a weak quasiparticle weight reduction. Necessarily we have to expect that the quasiparticle weight - even in the near-nodal regions - will shrink upon lowering the hole doping concentration towards the insulator.

Taken together, these trends naturally suggest a dome shaped CDW region in the T vs. x phase diagram centered around an underdoped composition. These qualitative arguments comply with the experimental findings.

We have added this reasoning to the discussion section in the revised manuscript.

In summary, this is a nice attempt to describe the charge ordering in cuprates. The technical part is well done. The results, however are not enough to convince the proposed mechanism is in fact responsible for the observed phenomenon.

We acknowledge that we have not yet convinced the reviewer that our proposed phonon mechanism is responsible for the charge ordering in cuprates. Our work started from the perception that up to now no purely electronic theory appears to be capable to explain the incommensurate uniaxial charge ordering wavevector. A new route and an expanded ansatz was and is in order to address the complexity of the CDW in cuprates.

At the core of our proposal is that it requires both, correlated electron physics and the coupling to the lattice degrees of freedom. The special momentum-space structure of the electron-phonon coupling has revealed an important ingredient which was not recognized in electronic theories before. This is the variation of the orbital content on the Fermi surface, which dictates for which momenta the coupling, here to the B_{1g} phonon, is strongest. In this respect we consider our results a relevant step forward to elucidate the true complexity of the CDW phenomenon in cuprates.

Strong electronic interactions by themselves develop charge correlations, but we infer that these may lock into an incommensurate charge-ordering pattern only in conjunction with a specific microscopic coupling to the lattice.

In this spirit we have added a concluding remark to our manuscript.

The paper addresses the question of the origin of the mysterious charge density wave (CDW) order that has been reported for practically all cuprates. The scenario analyzed in the paper is based on the electron-phonon coupling of the bond-buckling phonon. The authors write interaction Hamiltonian based on standard textbook theory and assume experimental electronic band dispersions. The results of this calculation are interesting to specialists. However, because the paper has many serious shortcomings, I do not recommend it for publication. Perhaps these can be remedied in a future version. Here is the list of areas that need reconsideration in more or less random order.

We acknowledge the very constructive criticism of the Referee and hope that the following reply clarifies most of the critique raised in the context of our proposal.

1. The electronic band structure in the calculations comes from a relatively old paper, Ref. 42. More recent and precise parameterization of experimental data was published that includes not only electronic dispersions but lifetime broadening as well (Li et al. Nat. Commun. 9, 26 (2018)). The calculation should be redone based on this result or perhaps other experimental work that includes lifetime broadening and bilayer splitting.

The paper by O. K. Anderson et al (Ref. 37), which provides the band structure that forms the basis of our calculation, is invaluable despite its age because it provides an orbitally-resolved model for YBCO. This is essential for our work because the charge density wave involves orbital degrees of freedom that are not accounted for by single-band fits to ARPES experiments, such as the one obtained by Li et al. ARPES experiments, however, do play a role because they establish that band masses are renormalized by a factor of three, and that the spectral weight vanishes towards the antinodes at low temperature.

However, one must be careful about material differences. The vast majority of ARPES results are for BSCCO, including the work by Li et al that was mentioned by the referee. These experiments consistently find a low-temperature quasiparticle broadening (~ 10 meV near the gap node, e.g. PRB 75, 140513) that is orders-of-magnitude larger than what is obtained from transport experiments in YBCO. Notably, microwave measurements in YBCO_{6.5} find transport scattering rates that are of order 0.1 meV (PRB 74 104508, 2006), which is 100 times smaller than typical values quoted for BSCCO. Indeed, STM experiments by Seamus Davis' group and others confirm that BSCCO is highly inhomogeneous, while YBCO is believed to be homogeneous. It seems to us likely that scattering rates obtained from ARPES in BSCCO include significant disorder broadening at low temperature and energy, and are not relevant to YBCO.

The role of bilayer splitting was discussed previously (Maharaj, PRB 90 125108 (2014)), where it was found that it serves principally to determine the relative orientation and phase of the charge density waves in the two layers making up the bilayer. However, it has not been observed to have much effect on the structure of the CDW in the individual layers. In YBCO, the situation is more subtle, as there is evidence that bilayer splitting near the nodal region (where the CDWs form) actually collapses at low doping (Fournier et al, Nat. Phys. 6, 905 (2010)).

In summary, we believe that the approach we have taken is appropriate given what is known about YBCO. We start from a multi-orbital tight-binding model that gives the orbital weights of the different bands. In recognition that DFT calculations are imperfect, we add two missing elements: renormalization of the band mass and quasiparticle weight by strong correlations. Alongside, we have included our reasoning why we disregard bilayer effects.

2. Work of Devereaux et al has shown that the buckling mode can have strong electron-phonon coupling only in multilayer cuprates. Single layer cuprates like do not show any electron-phonon effects for the buckling branch. There was a recent archive posting on Hg1201 for example showing this. How does this fit into the

proposed picture? Are the authors claiming that single layer and multilayer cuprates have completely different CDW mechanisms? Or is Devereaux's work wrong?

The case of single-layer cuprates is more complicated than YBCO. It is true that in pristine Hg1201 and Bi2201, the CuO₂ planes have mirror symmetries that prohibit a linear coupling between the B_{1g} phonon and the electrons. However, both of these materials are doped by large concentrations of interstitial oxygen atoms that sit above the CuO₂ plane. These dopants are unscreened, and generate large electric fields in the CuO₂ planes. Interstitial oxygen atoms, for example, were invoked by Tabis et al (PRB 96, 134510 (2017)) as nucleation sites for short-range CDW patches. Importantly for our work, Johnston et al (EPL 86, 37007 (2009)) showed in the context of Bi2212 that interstitial oxygen generates electric fields at the planar oxygen sites that are of order a few eV/Å. The resultant field strengths are comparable to that obtained for YBCO, and must (by symmetry) produce a linear coupling between the B_{1g} phonon and the CDW. Indeed, Johnston et al showed that the electron-phonon coupling to the B_{1g} mode is strongly enhanced in Bi2212, by up to a factor of 5 near the oxygen impurities. We expect a similar strong enhancement in the single-layer cuprates Bi2201 and Hg1201.

We do note that there is a key difference between this proposal for single-layer cuprates and our calculation for bilayer cuprates; namely, because the electron-phonon coupling is local and inhomogeneous, we expect a network of small CDW patches in the single-layer materials, much as what is seen in experiments.

An important question is the extent to which such inhomogeneous electron-phonon coupling should be visible in the phonon dispersion. Provided the phonons are harmonic, the lattice distortion associated with the local electric fields shouldn't shift the phonon frequencies; rather, the electron-phonon matrix element will be distributed across a range of values leading to a broadening of the phonon dispersion near q*. A proper calculation would be required, however, to determine the scale of the broadening, and in particular how it compares with other sources of apparent broadening (as discussed by the recent work by Ahmadova et al (Phys. Rev. B 101, 184508 (2020)) mentioned by the referee).

In summary, we believe there is a plausible mechanism by which the B_{1g} phonons can play a role in the CDW formation in single-layer cuprates, as well. We have added text to the discussion section at the end of the manuscript to address these issues.

3. What about bond-stretching phonons that have much stronger electron- coupling than the buckling phonons? The authors should discuss how these effects fit into their picture and why they think it is OK to ignore these experimental results.

As we discussed in our response to Referee A, XRD experiments are clear that the lattice distortion has predominantly B_{1g} character, with a small additional in-plane displacement of the oxygen atoms. The justification for neglecting the bond-stretching modes is thus empirical. In the revised manuscript, we have expanded on our empirical arguments for primarily selecting the B_{1g} phonon.

4. The paper cites Ref. 25 that there is a CDW signature in the acoustic phonons of BSCCO. But a subsequent paper (Phys. Rev. B 100, 144502) showed that this is not a signature of a CDW. I recommend either not to cite Ref. 25 because it is not really necessary, or cite both papers.

We thank the referee for pointing out the paper by Merritt et al. (Phys. Rev. B 100, 144502 (2019)). Since this paper falsifies the earlier conclusion of Ref. [25] about a CDW signature in an acoustic phonon branch of B-2212, we chose to no longer cite Ref. [25] and to remove it from our reference list.

5. DFT calculations that are also based on electronic structure do not get any phonon anomalies or CDW in cuprates. This is not clear from the paper at all except may be to narrow specialists. The authors should state clearly what they are doing that is different from DFT.

This is correct, and indeed DFT calculations have long struggled to capture physics related to strong correlations in the cuprates. We have added a short note on the incompleteness of DFT. The key distinction between our work and density functional perturbation theory is the explicit, albeit phenomenological,

inclusion of strong-correlation effects through the renormalization of the band structure. Note that even in our work, the predicted phonon softening is a weak effect.

6. What about the anomalies seen by LeTacon group in the acoustic phonon? This should also be discussed.

Again, we emphasize that our decision to select the B_{1g} mode is empirical: the XRD measurements of Forgan et al show that all atoms in the unit cell are displaced by the CDW, but that the dominant mode in the CDW pattern has B_{1g} character. The question we set out to ask is whether such a mode can be the driver behind the observed CDW in YBCO. In the Le Tacon experiment, the strongest softening is found for a low-energy transverse optical phonon that involves motion of heavy elements (Y and Ba). As we said above: While the softening of this mode does indeed indicate a coupling to the CDW (as must occur in the absence of special symmetries), the small static displacements of the Ba and Y atoms observed by Forgan et al suggests that ultimately there is a relatively weak participation of this mode in the CDW formation.

We have added text addressing the experiments in the Le Tacon group.

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

I appreciate the revisions to the text made by the authors, which improves the manuscript. Most issues are fine. I would, however, like a couple of points to be revisited.

1. The reply and revisions seem to want to assert that YBCO is some sense the canonical example of CO and La-based cuprate are in some sense. a "special, material specific" case. I think it would be better here to just state that the work only addresses a subset of the cuprate charge order phenomenology. The discussion in this section of the introduction is not well supported by the literature overall.

a. Citing just [19] is a bit misleading as there is good quality evidence that this paper is wrong <https://arxiv.org/abs/1904.12929>. I would suggest staying out of such an argument and deleting this statement about orbital symmetry.

b. La-based cuprates have CDWs even without an LTT structure [PRB Phys. Rev. B 89, 224513 (2014); PRB 90, 100510 (2014); PNAS 114, 12430–12435 (2017); Nat. Comm. 10, Article number: 3269 (2019)]. There are even observations of changes in CDW wavevector with temperature PNAS 114, 12430–12435 (2017).

2. I feel that the critical tone of the text that has been added here is not very fair regarding other work.

"Yet, finding the charge-stripe ordered ground states proved elusive due to the near degeneracy of the competing states."

Most calculations determine stripes *are* the ground state. The fact that SC is also predicated at only slightly higher energies is a strength of the work by other groups and a weakness of the current work.

3. I like the new title.

4. Fine.

5. The added explanation improves the paper. I think it would be better to remove the word "clearly" from the statement: "the x-ray experiments [33] clearly that the lattice distortions in the CDW phase have predominant B1g character." [33] has a much more nuanced discussion of this issue. "We note the similarity of some of the displacements to a soft phonon observed in optimally doped YBCO (ref. 28). However, in that mode, the c-motion is in antiphase for the two halves of the bilayer."

6. Fine

7. Fine

8. Fine

9. I had mentioned the arxiv paper earlier in my report.

Reviewer #2 (Remarks to the Author):

The main claim of the paper is the CDW phenomena in cuprates involve both electronic strong correlations and electron-phonon coupling, the latter being unavoidable in the presence of the lattice distortions associated with CDW.

The revised version of the paper made these points more clear.

In my opinion, in the present form the paper can be accepted for publication.

Reviewer #3 (Remarks to the Author):

The authors addressed my comments in the previous round and now the manuscript is significantly improved.

One issue that is still not adequately discussed is the relationship (if any) of the bond-stretching phonon renormalization that is much larger than the softening of the buckling mode. I'd like to see a thorough discussion of where these fit into the picture proposed in the paper with recent references cited.

After that I would recommend the paper for publication.

I appreciate the revisions to the text made by the authors, which improves the manuscript. Most issues are fine. I would, however, like a couple of points to be revisited.

We thank the Referee for providing positive feedback on our work and acknowledge the supportive comments which helped us to improve our paper significantly.

1. The reply and revisions seem to want to assert that YBCO is some sense the canonical example of CO and La-based cuprate are in some sense. a “special, material specific” case. I think it would be better here to just state that the work only addresses a subset of the cuprate charge order phenomenology. The discussion in this section of the introduction is not well supported by the literature overall.

It is certainly not our intention to argue that YBCO is the canonical example for charge order in cuprates. Rather, we point to some apparent differences between the CDW in YBCO and spin- and charge-stripe order in 214 cuprates.

a. Citing just [19] is a bit misleading as there is good quality evidence that this paper is wrong <https://arxiv.org/abs/1904.12929>. I would suggest staying out of such an argument and deleting this statement about orbital symmetry.

We had not recognized that the results and conclusions of Ref. [19] were later overruled by the paper arXiv:1904.12929 (our Ref. [65] in the previous version) of the same group. We follow the reviewer’s suggestion and have removed the sentence on orbital symmetry in the first paragraph. We have also deleted Ref. [19].

b. La-based cuprates have CDWs even without an LTT structure [Phys. Rev. B 89, 224513 (2014), Phys. Rev. B 90, 100510 (2014), PNAS 114, 12430–12435 (2017), Nat. Comm. 10, Article number: 3269 (2019)]. There are even observations of changes in CDW wavevector with temperature PNAS 114, 12430–12435 (2017).

This is correct that charge order is observed also in 214 cuprates without a structural transition to an LTT phase. Indeed, the physics of charge order in 214 cuprates is not addressed in our manuscript. We have therefore dropped the side-remark sentence on the LTT phase. In the revised remaining text of the first paragraph we leave it to point to just two differences between charge order in YBCO and La-based 214 cuprates: i) the different wavevector trends upon doping and ii) the additional spin stripes in 214 cuprates.

2. I feel that the critical tone of the text that has been added here is not very fair regarding other work. “Yet, finding the charge-stripe ordered ground states proved elusive due to the near degeneracy of the competing states.”

Most calculations determine stripes *are* the ground state. The fact that SC is also predicted at only slightly higher energies is a strength of the work by other groups and a weakness of the current work.

We regret that our choice of words conveyed that we do not give high value to the accurate and impressive numerical work which verified stripes as groundstates with competing states extremely close in energy. To avoid any false impression we fixed our wording accordingly.

3. I like the new title.

Thank you. We do, too.

4. Fine.

5. The added explanation improves the paper. I think it would be better to remove the word “clearly” from the statement: “the x-ray experiments [33] clearly that the lattice distortions in the CDW phase have predominant B_{1g} character.” [33] has a much more nuanced discussion of this issue. “We note the similarity of some of the displacements to a soft phonon observed in optimally doped YBCO (Ref. 28). However, in that mode, the c-motion is in antiphase for the two halves of the bilayer.”

Thanks for pointing out that the discussion in Ref. [33] is more cautious and detailed than what is summarized in our sentence containing the word “clearly”. We have now rephrased this sentence as “... the buckling mode, but the out-of-plane components are dominant, and we make the simplifying approximation that the in-plane component can be neglected.”

6. Fine

7. Fine

8. Fine

9. I had mentioned the arXiv paper earlier in my report.

Indeed, in the attempt to provide a prediction or at least to mention a consequence for our phonon-based mechanism to operate we have made contact to the results on the symmetry form factor presented in Ref. [65] of our previous version (arXiv:1904.12929).

Report of the Second Reviewer – COMMSPHYS-20-0083-T

The main claim of the paper is the CDW phenomena in cuprates involve both electronic strong correlations and electron-phonon coupling, the latter being unavoidable in the presence of the lattice distortions associated with CDW. The revised version of the paper made these points more clear.

In my opinion, in the present form the paper can be accepted for publication.

We thank the Referee for his/her positive review of our manuscript.

Report of the Third Reviewer – COMMSPHYS-20-0083-T

The authors addressed my comments in the previous round and now the manuscript is significantly improved.

We thank the Referee for his/her very constructive and thoughtful comments, which helped us to significantly improve our manuscript.

One issue that is still not adequately discussed is the relationship (if any) of the bond-stretching phonon renormalization that is much larger than the softening of the buckling mode. I'd like to see a thorough discussion of where these fit into the picture proposed in the paper with recent references cited.

After that I would recommend the paper for publication.

We added a short discussion on the role of bond-stretching phonons and how it couples to the underlying charge order in underdoped YBCO. The new references added are Refs.~[34-42].

REVIEWERS' COMMENTS:

Reviewer #3 (Remarks to the Author):

The authors incorporated a good discussion of the stretching phonons, which I requested, and the paper can now be published as is.