

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The manuscript presents an ab-initio study of superconductivity in Sr_2RuO_4 (SRO) using a merger of Dynamical Mean-Field Theory and Quasi-particle GW approaches. The approach is certainly state-of-the-art and it provides extremely helpful information.

The results reported are extremely interesting, as they provide an interpretation for the anomalous behavior of the superconducting critical temperature as a function of strain in this material. The authors discuss the evolution of the physics as a function of strain mainly in terms of the competition/interplay between spin and charge fluctuations. In the absence of strain the two channels essentially compete, leading to different superconducting phases with low critical temperature, while strain leads to a synergy between the two kind of fluctuations which cooperate in a given symmetry channel. When the strain is further increased the critical temperature decreases because the contribution of spin fluctuations is reduced as they lose coherence.

These results provide a nice picture which is consistent with several experimental facts. However, I have some reservations about the presentation which do not allow me to suggest publication of the manuscript in the present form

1) I think that the present manuscript does not provide sufficient direct evidence for the main claims I summarized above. The manuscript reports data for the Fermi surface, the charge and the spin susceptibilities and finally a map of the order parameter amplitude for the five largest eigenvalues of the pairing equation.

1a) First of all the definition of the relevant quantities and the procedure to obtain the data of Fig. 4 must be discussed in the supplementary materials and some clearer word must be included in the text besides the very short discussion of page 7 (first half) which can be followed only by experts and still contains some ambiguity about the exact procedure. The reason why showing data for the first eigenvalues should be added once some equations clarify their meaning. An obvious question is why five eigenvalues? Also the discussion about the role of the eigenvalues is very hard to follow and tries to be evocative without delivering a strong message. What does it mean that the first two eigenvalues start to decay with the third or that the fifth makes the spectrum (of what?) dense? I think I can figure it, but the presentation is really far from what is needed for this journal.

1b) I think the authors should make an effort to give convincing arguments about the interplay/competition between spin and charge channels. In the present version, if I understand correctly, the main point is that there is synergy when the fluctuations in the two channels have peak at the same momentum. This is an indication, but not a strong proof. It would be useful to connect this more directly with the information of Fig. 4

2) The introductory sections and the abstract are not well written. For example in the abstract the sentence "Emergence of ... degree of freedom" does not make clear if the emergence the authors talk about is an experimental fact (if so, it should be clear and discussed in the paper) or theoretical, "harmonious" is evocative but of little meaning. The introduction on superconductivity is also weak. The reference to electron-phonon superconductivity is useless and potentially misleading. For example, not all electron-phonon superconductors are BCS-like. In some cases you need Eliashberg theory, in others one can have polaronic effects. Of course this is not a major point, but I would like to convey the message to drop these useless sections to save space for the more relevant discussions related to point 1).

3) I do not understand the reason why the FM signal in the spin response is a consequence of the multiorbital character.

4) Why the information about spin and charge response is reported using two completely different sets of data (dynamical susceptibility vs static)?

5) Finally the language can be improved. I do not think that being "detailed" is the quality we look for in a theory, and there are several expressions like "minimal model Hamiltonian based approaches" which can be simplified.

In my opinion a manuscript which seriously takes into account all these points can be reconsidered for publication in Communications Physics.

Reviewer #2 (Remarks to the Author):

The authors report a GW+DMFT calculation of the Fermi surface properties and of certain aspects of superconductivity in Sr₂RuO₄. This material is one of the few candidates of spin-triplet superconductivity, and has been the subject of intense study, both theoretical and experimental. Despite these studies, a lot of questions are still unanswered about this material.

I think the calculation is certainly an impressive numerical prowess, and useful for the field. Doing calculations in this material is quite challenging, due to the complicated nature of the Fermiology, which includes three active bands, spin-orbit coupling, and so on. Their results about finite strain are also very interesting, given the recent experimental discovery of a van Hove singularity under strain.

The paper has two parts: results about the normal state, and results about superconductivity.

The first part is interesting and well presented. A precise understanding of the normal state, and its dominant fluctuations in the spin and charge channels, is a prerequisite to the understanding of superconductivity, and it is therefore a good contribution to the field of Sr₂RuO₄.

I have a few caveats though:

- * the first panel of fig 1 shows a very wobbly gamma Fermi surface. Isn't this inconsistent with the very well documented quantum oscillation studies in Sr₂RuO₄ [Bergemann et al, Advances in Physics, 2003, Vol. 52, No. 7, 639], which does not show this ``wobbliness'?
- * the authors should give the mathematical definition of χ^s and χ^c that they use, with all the orbital and spin indices. This is non-trivial in a spin-orbit coupled system like SRO.

The second part of the paper, about superconductivity, is too laconic to be useful in my opinion. It feels rushed, and lacks basic information about the calculations done and the results obtained.

- * The authors should explain in more details, with equations, what they mean by "This construction allows us to separate the contributions ... into spin and charge contributions", which is not trivial in a spin-orbit coupled system.
- * The authors should show their leading eigenvalues, instead of talking about them in the text.
- * In which symmetry group representation of the point group D_{4h} are each of these gaps shown in Fig 4? This is a basic piece of information about the results that seems to be missing.
- * Regarding the eigenvalues, what does "nearly degenerate" mean? Compared to what? Does that mean the two eigenvectors form a TRSB state? If yes, in which representation?
- * It would be good to add a sketch of the crystalline axis used in Fig 4, compared to the conventional axis abc used in the literature.
- * The authors seem to put under the rug 2 decades of theoretical study of superconductivity in SRO. Several gap structures have been studied intensively already, and compared to experiments

in various ways (specific heat, ...). The authors should cite these works, like for example [Nomura, Yamada, JPSJ.71.404] and [Scaffidi, Romers, Simon Phys. Rev. B 89, 220510], and contrast their results with the literature.

Instead, the authors show 5 figures of gaps which, as far as I can tell, are completely different than any gap previously proposed in the literature, without even doing a simple quantitative check against experiments like the specific heat.

In conclusion, I think the first part of the paper about the normal state is very useful for the field, but the second part about superconductivity is not publishable as is.

These reports have been very useful for us in improving and clarifying the presentation of our work.

We thank both the referees for noting the originality, scientific value and key ingredients of our manuscript. We realize that the essential objection common to both the reports from the referees, is the need of further clarification and a proper discussion of the superconducting instabilities.

We fully appreciate that the method of computation and discussion related to the superconducting instabilities needed a rigorous revision. The present version includes a thorough revision for all possible singlet and triplet instabilities with and without strain in Sr_2RuO_4 . All queries and suggestion from the referees are addressed in the revised version. We submit, hereby, a revised version in which all of the remarks of the referees have been taken fully into account.

The answer to the comments of the referees and a list of changes made to the manuscript are provided below.

1 Reviewer 1 :

Comment 1 The manuscript presents an ab-initio study of superconductivity in Sr_2RuO_4 using a merger of Dynamical Mean-Field Theory and Quasi-particle GW approaches. The approach is certainly state-of-the-art and it provides extremely helpful information.

The results reported are extremely interesting, as they provide an interpretation for the anomalous behavior of the superconducting critical temperature as a function of strain in this material. The authors discuss the evolution of the physics as a function of strain mainly in terms of the competition/interplay between spin and charge fluctuations. In the absence of strain the two channels essentially compete, leading to different superconducting phases with low critical temperature, while strain leads to a synergy between the two kind of fluctuations which cooperate in a given symmetry channel. When the strain is further increased the critical temperature decreases because the contribution of spin fluctuations is reduced as they lose coherence.

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a map of the order parameter amplitude for the five largest eigenvalues of the pairing equation.

1a) First of all the definition of the relevant quantities and the procedure to obtain the data of Fig. 4 must be discussed in the supplementary materials and some clearer word must be included in the text besides the very short discussion of page 7 (first half) which can be followed only by experts and still contains some ambiguity about the exact procedure.

Answer: In the main paper, we have now added essential method related details and the equations that make the flow of logic and quantities calculated clear. In the supplemental material the diagrammatic construction and the equations for computing spin, charge and superconducting susceptibilities are discussed in greater detail. Hyowon Park, who is an author on our paper originally implemented the scheme and [his PhD thesis](#) (which is cited in the paper) covers the entire methodological details. Additionally, each branch of our implementation is extensively discussed in a recent method paper ([arXiv:1907.06021](#)).

Comments 1 and 1a The reason why showing data for the first eigenvalues should be added once some equations clarify their meaning. An obvious question is why five eigenvalues? Also the discussion about the role of the eigenvalues is very hard to follow and tries to be evocative without delivering a strong message. What does it mean that the first two eigenvalues start to decay with the third or that the fifth makes the spectrum (of what?) dense? I think I can figure it, but the presentation is really far from what is needed for this journal.

Answer: The whole section discussing the superconducting instabilities in different triplet and singlet channels is re-written (please find the revised relevant figures at the end of this document, the revised paper and supplementary material also include them). All the eigenvalues observed and corresponding eigenfunctions, their momentum structure and D_{4h} irreducible representations are thoroughly characterized.

Computation of these eigenfunctions at finite temperatures, within our ab-initio framework, is costly. In the earlier version of the paper, lack of access to significant amount of computing resources led to bad quality of vertex functions from our continuous time Quantum Monte Carlo study. Additionally, our understanding of such intricate multi-orbital superconductivity is also evolving. We thank the referees for leading us to do the calculations in the right direction that allowed us to have a more concrete understanding of different superconducting instabilities in SRO.

Also, in the process, our understanding of the physical consequences of our numerical implementation became more clearer. We compute these eigenfunctions in different intra- and inter-orbital basis which needs to be em-

bedded into band representation to project the superconducting gap on the Fermi surfaces. To embed these normal phase momentum dependent pairing instabilities in orbital basis to band basis further developments are required. Standard embedding mechanism as chalked out in this paper by [Yin et al.](#), has to be developed. Once embedded into the band basis, the subsequent Bogoliubov Hamiltonian has to be diagonalized to extract the spectral functions of the ordered phase. In the present paper, we aim to see how the eigenvalues in singlet and triplet channel evolve with strain and temperature. These conclusions are robust. Nevertheless, to know about the precise momentum structure of the ordered gapped phase, it needs further developments.

Comment 1b I think the authors should make an effort to give convincing arguments about the interplay/competition between spin and charge channels. In the present version, if I understand correctly, the main point is that there is synergy when the fluctuations in the two channels have peak at the same momentum. This is an indication, but not a strong proof. It would be useful to connect this more directly with the information of Fig. 4

Answer We thank the referee for raising this issue. We indeed agree that this is not a trivial issue to address. The fact that under strain the spin and charge fluctuations shift toward singlet-pairing vectors is only suggestive and not conclusive evidence that spin and charge work in symphony to lead a singlet pairing instability under strain. To address the issue conclusively we compute the eigenvalues with and without the charge vertex functions. With both spin and charge vertex functions (as the calculations should be performed), we observe that for $\epsilon_x=0$ triplet eigenvalues are larger than the singlet eigenvalues and as strain is applied $\epsilon_x > 0$, the singlet eigenvalues overcome the triplet eigenvalues. On suppressing the charge vertex, we find that for $\epsilon_x=0$, it does not have any effect on the result; triplet eigenvalues remain larger than the singlet eigenvalues. However, for finite strain, $\epsilon_x > 0$, once the charge vertex is suppressed the triplet eigenvalues become larger than the singlet eigenvalues, which is contrary to the observation including both spin and charge vertex functions. This is strong indication that both spin and charge vertex function work in symphony under strain in the singlet channel. We now include a table in the paper showing the eigenvalues with and without the charge vertex to make a case for this argument.

Comment 2 The introductory sections and the abstract are not well written. For example in the abstract the sentence "Emergence of ... degree of freedom" does not make clear if the emergence the authors talk about is an experimental fact (if so, it should be clear and discussed in the paper) or theoretical, "harmonious" is evocative but of little meaning. The introduction on superconductivity is also weak. The reference to electron-phonon superconductivity is useless and potentially misleading. For example, not

all electron-phonon superconductors are BCS-like. in some cases you need Eliashberg theory, in others one can have polaronic effects. Of course this is not a major point, but I would like to convey the message to drop these useless sections to save space for the more relevant discussions related to point 1).

Answer: The abstract is thoroughly re-written. We note the concerns raised by referee and modify the draft accordingly.

Comment 3 I do not understand the reason why the FM signal in the spin response is a consequence of the multiorbital character.

Answer: Our initial argument stems from the observations that presence of SO coupling and lifting of band degeneracy, lead to weak suppression of spin fluctuations at the Γ point. But we understand, that at least, within our theoretical framework, it can not be concluded unambiguously that the spectral weight at Γ is consequence of multi-orbital character of system. We remove the statement from the revised text.

Comment 4 Why the information about spin and charge response is reported using two completely different sets of data (dynamical susceptibility vs static)?

Answer: We compute both real and imaginary parts of spin and charge susceptibilities by solving the BSE in respective channels. These equations are solved in the matsubara representation with local dynamic vertex functions (which are functions of three matsubara frequencies) and non-local polarization bubble also in the matsubara frequencies. After summing over all internal Fermionic matsubara frequencies and orbital indices, we are left with $\chi_{s,c}(q, i\omega)$. Now it needs to be analytically continued to real bosonic frequencies. One way is to analytically continue the $\chi(i\omega)$ at each momentum, which is tremendously expensive. Precisely at $\omega=0$ the imaginary part of both spin and charge susceptibilities are zero. To understand the precise nature of the spin fluctuations at finite energies, it was imperative in this work that we extract the $\text{Im}\chi(q, \omega)$ for finite ω . For low energies, which is the focus here, the vertex Γ_{loc}^{irr} is analytically continued by a quasiparticle-like approximation. We replace the frequency-dependent vertex with a constant, i.e., $\Gamma_{loc}^{irr}(i\nu, i\nu', i\omega)_{\alpha_1\sigma_1, \alpha_3\sigma_3}^{\alpha_2\sigma_2, \alpha_4\sigma_4} \sim U_{\alpha_1\sigma_1, \alpha_3\sigma_3}^{eff, \alpha_2\sigma_2, \alpha_4\sigma_4}$ which satisfies the constraint that $\chi(q, i\omega=0)=\chi(q, \omega=0)$. This vertex U^{eff} contains all the important spin, orbital dependence. This approximation for analytic continuation works remarkably well for spin susceptibilities as shown by one of the authors of this paper [Hyowon Park in Park et al., PRL 107, 137007 \(2011\)](#) and also by [Z P Yin et al., Nature Physics volume 10, pages 845–850 \(2014\)](#).

However, this approximate method for analytic continuation of charge susceptibility may or may not work, as we know charge susceptibilities can

have significant amount of features at higher energies as well, and to get the susceptibility correctly at all energies approximating the [vertex to be static may not be sufficient](#). This is precisely the reason, we show our computed susceptibilities at $\omega=0$ which is not plagued by any of these approximations for analytic continuation.

Comment 5 Finally the language can be improved. I do not think that being "detailed" is the quality we look for in a theory, and there are several expressions like "minimal model Hamiltonian based approaches" which can be simplified.

Answer: We do a thorough revision of the language in text in lines with the suggestion from the referee.

2 Reviewer 2 :

Comment 1 The authors report a GW+DMFT calculation of the Fermi surface properties and of certain aspects of superconductivity in Sr_2RuO_4 . This material is one of the few candidates of spin-triplet superconductivity, and has been the subject of intense study, both theoretical and experimental. Despite these studies, a lot of questions are still unanswered about this material.

I think the calculation is certainly an impressive numerical prowess, and useful for the field. Doing calculations in this material is quite challenging, due to the complicated nature of the Fermiology, which includes three active bands, spin-orbit coupling, and so on. Their results about finite strain are also very interesting, given the recent experimental discovery of a van Hove singularity under strain.

The paper has two parts: results about the normal state, and results about superconductivity.

The first part is interesting and well presented. A precise understanding of the normal state, and its dominant fluctuations in the spin and charge channels, is a prerequisite to the understanding of superconductivity, and it is therefore a good contribution to the field of Sr_2RuO_4 .

I have a few caveats though: 1. the first panel of fig 1 shows a very wobbly gamma Fermi surface. Isn't this inconsistent with the very well documented quantum oscillation studies in Sr_2RuO_4 [Bergemann et al, Advances in Physics, 2003, Vol. 52, No. 7, 639], which does not show this "wobbliness"?

Answer: We thank the referee for pointing this out. We agree with the referee that, indeed, this particular result presented in the first version of our paper did not agree with experiment. However, we could finally pin down the origin of the discrepancy as the momentum interpolation of the GW self

energy in the tetragonal phase. Under strain once the tetragonal symmetry was lifted this interpolation problem was absent. However, we have sorted this numerical problem out using a new short-ranged basis (see Sec. 3.12 in preprint <https://arxiv.org/abs/1907.06021>) and have revised the main text of the recent version with the QSGW Fermi surface plotted against the recent high resolution ARPES data from Tamai et al. Phys. Rev. X 9, 021048 (2019). We cite the paper by Bergemann et al., suggested by the referee in the relevant section. Additionally a larger high resolution figure for the comparison with ARPES data From Tamai et al. and our computed QSGW Fermi surface is presented in the supplemental material.

Comment 2 the authors should give the mathematical definition of χ^s and χ^c that they use, with all the orbital and spin indices. This is non-trivial in a spin-orbit coupled system like SRO.

Answer: We have now put in all the details regarding the computation of different susceptibilities in the main text and supplemental material. In case of SRO, the hybridization matrix is more diagonal in m_l, m_s basis than J, m_J basis. Additionally, for CT-QMC solver to be sign-problem free, it is desired that hybridization matrix is as diagonal as possible in a certain basis. So we perform all calculations with SO coupling in the m_l, m_s basis as it is both consistent with our physical observations and requirements for a sign-problem free CT-QMC solver. Hence, all the indices present in the equations are consistent with our calculations.

Comment 3 The second part of the paper, about superconductivity, is too laconic to be useful in my opinion. It feels rushed, and lacks basic information about the calculations done and the results obtained. * The authors should explain in more details, with equations, what they mean by "This construction allows us to separate the contributions ... into spin and charge contributions", which is not trivial in a spin-orbit coupled system. * The authors should show their leading eigenvalues, instead of talking about them in the text. * In which symmetry group representation of the point group D4h are each of these gaps shown in Fig 4? This is a basic piece of information about the results that seems to be missing. * Regarding the eigenvalues, what does "nearly degenerate" mean? Compared to what? Does that mean the two eigenvectors form a TRSB state? If yes, in which representation? * It would be good to add a sketch of the crystalline axis used in Fig 4, compared to the conventional axis abc used in the literature. * The authors seem to put under the rug 2 decades of theoretical study of superconductivity in SRO. Several gap structures have been studied intensively already, and compared to experiments in various ways (specific heat, ...). The authors should cite these works, like for example [Nomura, Yamada, JPSJ.71.404] and [Scaffidi, Romers, Simon Phys. Rev. B 89, 220510], and contrast their results with

the literature. Instead, the authors show 5 figures of gaps which, as far as I can tell, are completely different than any gap previously proposed in the literature, without even doing a simple quantitative check against experiments like the specific heat.

Answer: This was our first attempt into computing superconducting eigenvalues and eigenfunctions in a multi-band correlated manifold using a theory for excited states. Earlier studies in [doped one-band Hubbard model](#) this scheme was already shown to work.

However, we fully agree with the referee on his/her reservation about our results and discussion for superconductivity in the first version of the paper. We have thoroughly revised the complete section of superconductivity in the present version and, in the process, have addressed all the issues raised by referee (please find the revised relevant figures at the end of this document, the revised paper and supplementary material also include them).

3 List of changes

- Revised superconducting gap instabilities in triplet and singlet channel are included (Fig. 4). Their irreps in terms of D_{4h} are tabulated. A rigorous analysis with earlier predicted gap instabilities are performed.
- Temperature dependence of the eigenvalue corresponding to leading singlet and triplet instabilities (in supplemental material) and their strain dependence (Fig. 4 in main paper) are shown.
- Essentially the whole section for superconductivity is thoroughly revised, all suggested references and new experimental references are included.
- Abstract and introduction are rigorously revised.
- All details related to calculations of susceptibilities are included.
- Singlet and triplet superconducting instabilities are shown in revised Fig. 4.
- Fermi surface figure is revised showing rigorous benchmarking against recent ARPES data.

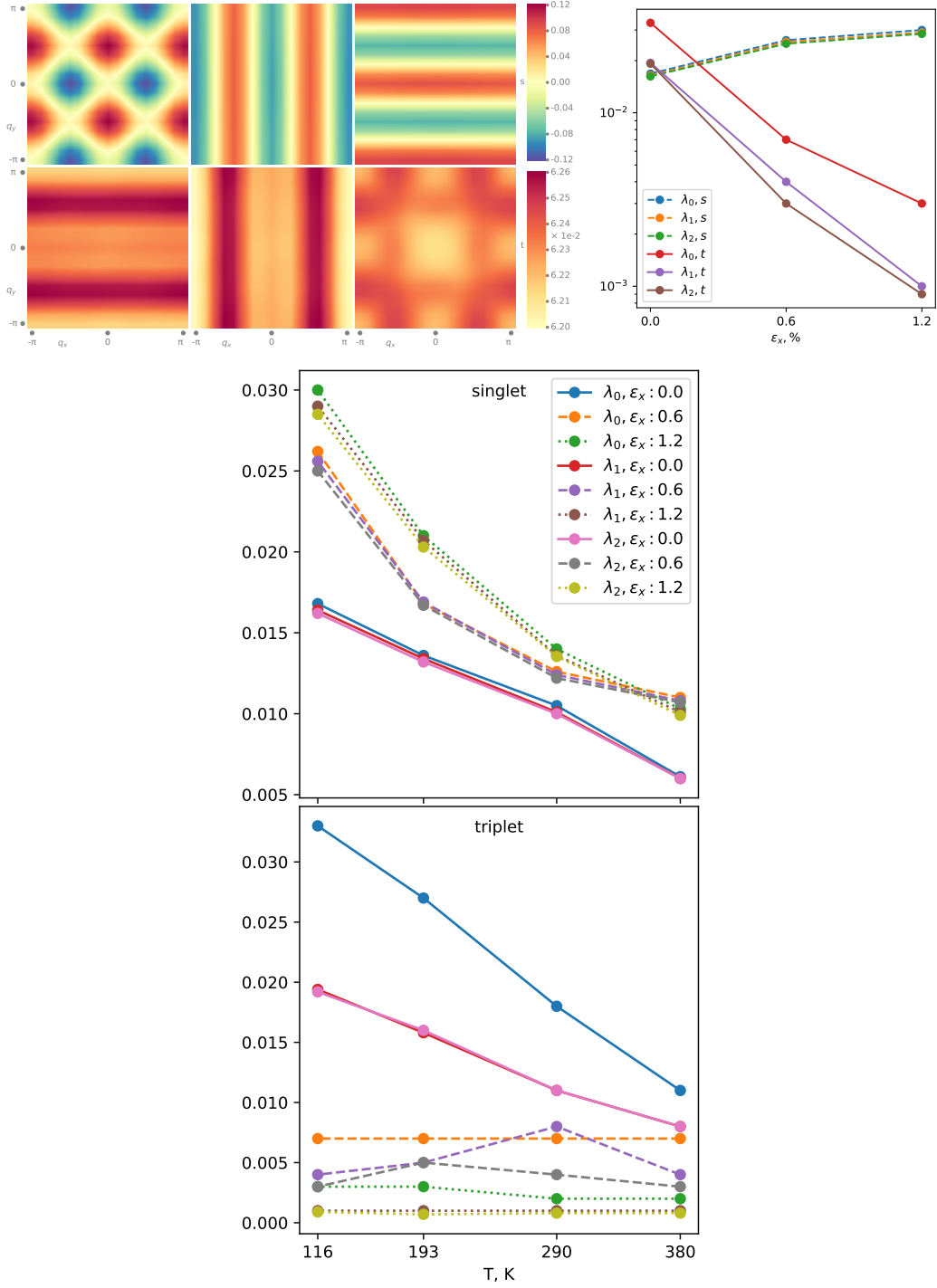


Figure 1: **Superconducting pairing functions and eigenvalues:** (left panel) The superconducting pairing gap symmetries for $\epsilon_x=0$ are shown in the conventional basal plane; eigenfunctions corresponding to first three eigenvalues in singlet (s) symmetries are in the top panel and triplets (t) are in the lower panel. Right panel shows evolution of triplet and singlet eigenvalues under strain. Under strain singlet eigenvalues increase and surpass the triplet eigenvalues. The temperature and strain evolution of singlet and triplet eigenvalues are shown in the bottom panel.

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors have taken into account all the objections raised by myself and by the other referee and they modified the manuscript accordingly. The most important modification is a radical change to the discussion of the main idea about the methodology and the analysis of the response function which allows the author to draw conclusions about the superconducting instabilities. The authors also improved some data (for example the Fermi surface results, which are now much closer to experimental data from ARPES and quantum oscillations).

I believe that these changes significantly improved the manuscript, which is now significantly better. However, I still think that the authors should make a final effort to improve the manuscript.

The introduction section is still too schematic and disordered to convey the message of the manuscript. I give some examples: The sentence about unconventional pairing, though better than before is still uselessly generic (e.g., "It usually involves", what does the sentence mean?). The comment about disorder in line 8 does not seem in the right place.

I think the authors should introduce the problematic starting from SRO, introducing the main ideas and the new experimental evidences challenging then and then introduce their approach.

I do not mean to rewrite the paper according to my taste. My main point is that the paper starts with a premise which is not really fitting with the actual content of the manuscript, which provides certainly important information on SRO, but hardly addresses general questions on unconventional superconductivity.

Reviewer #2 (Remarks to the Author):

The authors have substantially improved the part of the paper about superconductivity since the last version. I believe the manuscript is now in publishable form.

We thank both the referees for reviewing our paper. We sincerely acknowledge that the comments and suggestions from both the referees have helped us significantly improve the content and readability of the paper.

1 Reviewer 1 :

Comment: The introduction section is still too schematic and disordered to convey the message of the manuscript. My main point is that the paper starts with a premise which is not really fitting with the actual content of the manuscript, which provides certainly important information on SRO, but hardly addresses general questions on unconventional superconductivity.

Response: We fully appreciate referee's concern regarding the introduction and rewrite the section accordingly.

2 Reviewer 2 :

Comment: The authors have substantially improved the part of the paper about superconductivity since the last version. I believe the manuscript is now in publishable form.

Response: We thank the referee sincerely for recommending our paper for publication.