Report of Referee 2

Strengths

a. The two-particle self-consistent approach has proven quite successful in describing the physics of the one-band Hubbard model in two dimensions. It satisfies several exact results. A generalization of this method for general multi-orbital problems is called for. Despite a few previous attempts, the literature has not converged yet towards a unique, benchmarked approach. This paper provides a generalization of the multi-orbital two-particle self-consistent approach, without relying on Dynamical mean filed theory, that it compares with previous works. It constructs 5 different spin-dependent Ansätze and 3 different spin-independent Ansätze that it benchmarks at half-filling.

b. Long derivations that need to be in the literature are presented.

c. They made it possible to close the system of equations for the vertices using sum rules and symmetries.

d. There is an honest declaration of the limitations of the method.

e. The method benchmarked the local vertices with Exact Diagonalisation and Dynamical Mean Field Theory, which are well-known in the field. (See however reservations in the report).

We thank the Referee for this kind summary of our work.

Weaknesses

a. It needs further explanations or reference for some statements.

b. Many calculations remain obscure and even probably incorrect as written (typos?).

c. There is a lack details in the benchmark sections on the correspondence with the model parameters and the values of the D,P and C parameters. This makes the results and conclusions hard to validate.

d. There are some statements/conclusions that are unclear.

e. Some of the notation is unclear or undefined and even inconsistent.

The Referee raised many valid points, which needed clarification. With the help of the very extensive list of recommendations, we hope that we succeeded in improving the readability, accessibility and the clarity of our article. We note that many of the points (2,5,6,7,15,18,19,22) stemmed from us trying to make the paper more accessible by using a compact notation, but which had the opposite effect. In the resubmitted paper, we use a more explicit notation.

Report

This report is for the original submission of this paper. It was discussed with André-Marie Tremblay. The two-particle self-consistent approach (TPSC) satisfies several exact results, like sum-rules, conservation laws and Pauli principle. It also satisfies the Mermin-Wagner theorem in two dimensions. Other methods, such as RPA violate several of these exact results and others like the renormalization group are much more difficult to implement and require more computing power. This makes TPSC a method of choice for the Hubbard model. Its generalization to multi-orbital models is an important and non-trivial next step that this paper intends to achieve. It is appropriate in principle for Scipost. Unfortunately, in its present form, this paper cannot be published. It lacks justifications of some steps, presents benchmarks that are not always relevant, contains many typos and lacks some details that would be necessary for reproducibility. I suggest a partial list of changes and verifications of the derivations to make sure possible mistakes are avoided. After the authors respond to the constructive criticism below and after further review by me or some other referee, I hope publication can be recommended.

Requested changes:

N.B. The changes in notation as one progresses through the paper make it difficult to find out what is a typo and what is a mistake. I have rederived most equations and indicated where I have questions, but I had to stop at some point.

1. In eq. 1 : The interaction term should be 1/4 if you are assuming that the matrix element is antisymmetric, as is shown in eq 57 of Salmhofer and Honerkamp, that you reference. I personally think one should immediately start from the unsymmetrized form of the matrix element, and keep 1/2.

Yes, the Referee is correct that we start implicitly from the anti-symmetrized form. To emphasize the transition from the usual to the anti-symmetrized notation, we rewrote large parts of the corresponding section. Furthermore, we do acknowledge that this formalism is challenging when one is not used to it. To lower this hurdle, we added an alternative derivation starting directly from a Hubbard-Kanamori form of the interaction as an appendix. This alternative derivation is closely linked to earlier approaches and makes comparing the proposed variant with the old variants easier. Nonetheless, we keep in the main text the derivation in terms of an anti-symmetrized interaction, as this allows us to get rid of the spin dependence without the need of introducing a specific interaction form.

2. Also, in Eq.(1), why are you keeping such a general form of the interaction while you are assuming, without saying it explicitly, that the two-body interaction called U here is spin-independent (as follows from Eq.(2)).

We do not assume a spin independent interaction, but instead we assume an interaction fulfilling $SU(2)$ symmetry, as written above equation (2). This spin-symmetry automatically restricts the interaction to the shown form.

3. After eq. 1 : You define the creation (annihilation) operator with position and imaginary time indices, but in eq. 1, all operators are not explicitly imaginary time-dependent since the Hamiltonian, as usual, is imaginary-time independent. Also, you did not define τ_1 as imaginary time. I know you need to define creation (annihilation) operators with all the position, spin, imaginary time and orbital dependences since you need it later.

We apologize for this notational inconsistency and removed the imaginary time dependence. Furthermore, we made sure we stress at the correct point that we switch from time-independent to time-dependent operators.

4. In eq.3, the term C contains $S \cdot S$ which includes $S_z S_z$, so it does not contain only spin flips that are given by $S_x S_x + S_y S_y = (S_+ S_- + S_- S_+)/2$. Shouldn't it be impossible to have same spin for all the creation (annihilation) operators, because then it isn't really a spin-flip? The name seems inappropriate.

The referee is again correct, this identification is not entirely correct and is based on the analogy to the Hubbard-Kanamori type interaction. I.e. the spin-flip Hunds-coupling appears in the C channel, which is why it is sometimes called like this. We rewrote the section by making a connection to the different two-particle irreducible diagrammatic channels the interactions are "native" to, which offers a clear interpretation of the different terms.

5. Throughout the derivations (eq 4 and after) : Why did the order of indices change? Please keep the same order of numbered indices as in previous equations, otherwise it makes the derivation hard to follow.

We performed a renaming of the summation indices during the initial derivation to get a specific index ordering in the result. Following the suggestion of the Referee, we refrained from doing this in the resubmitted version, such that the index order is the same as in the equation of motion in the appendix.

6. You also use two different types of notation : an explicit one (e.g. $\Sigma_{o_1,s_1}(r_1,\tau_1)$) and an abbreviated one (e.g Σ_1). First, I would define what Σ_1 stands for. Second: I would suggest that you keep the same notation throughout, at least within the same eq. I do know how long those equations are, so you try to keep them more concise, but the most important thing is to explain when and how you use a short-hand. (For example, in the appendix, you use both at the same time, and when there are deltas, it is not clear whether these are delta functions for all variables, or just for position and time or otherwise.)

As mentioned above, we compressed redundant indices into these numbered indices. In the introduction, we mentioned once "As a short hand for all non explicitly stated quantum numbers of an object we will use plain numbers as indices". However, as we see this compressed notation led to confusion, we rewrote the paper without using it, with the exception of Appendix A, where we added a clear explanation of the notation.

7. In eq. 6 you seem to change notation from position and imaginary time dependent creation (annihilation) operators to non-explicitly dependent ones: Is this to make the eqs. simpler and easier to follow? Please comment on that.

Yes, the idea was to make the equations more readable by suppressing dependencies which are not changing during the derivation. In accordance with point 6 we explicitly now write these dependencies everywhere.

8. Going from eqs. 5 to 6 : I am not sure how you handled the switch in operators and the indices of U. Could you elaborate on that? When I try to do it, I switch the two annihilation operators of $o'_3 s_1$ and $o'_2 s'_4$, and because they anticommute, only the sign changes. Then I switch o'_2 and o'_3 for both U and the operators. That leaves me with two identical four-point function but a sum of $U_{o'_2 o'_3 o'_4 o_1}$ and $U_{o'_3 o'_2 o_1 o'_4}$. Although, because of the symmetry used, they are equivalent like you said and it cancels the 1/2, but I don't have any δ function or $1-\delta$. I think they should normally come from the model. And, by the way, it would have been easier to start from an interaction matrix element that was not antisymmetrized.

We apologize for the unclear origin of the delta - it is a redundant factor which was added during one point of the derivation (it essentially encapsulates the Pauli-principle since if the indices are identical we have a c^2) for ourselves but forgot to remove again. We removed these redundant factors from all equations and thank the Referee for catching this mistake.

9. Definition of $\bar{\delta}$: "$\bar{\delta}$ is a short hand for $1-\delta$": In eq. 6 I read this as : $\bar{\delta}_{s_1,s'_4}\bar{\delta}_{o'_2,o'_3} = (1-\delta_{s_1,s'_4})(1-\delta_{o'_2,o'_3}$: Which cancels all the aligned spin four-point functions, even when the orbitals are different. Thus, no Hund's coupling of aligned spins on different orbitals. In the benchmark, this is the term on the right in eq 50. Hence, if this above definition is the one you mean, I think it makes the benchmark a lot less accurate, and that this would explain why the case of the Kanamori Hamiltonian works better, because the other terms in the Kanamori model are not neglected by this. Although, when I follow your derivation, I think you mean it in this way $\bar{\delta}_{s_1,s'_4}\bar{\delta}_{o'_2,o'_3} = (1-\delta_{s_1,s'_4}\delta_{o'_2,o'_3}$, so that it respects the Pauli principle, but this should come from the model directly, as in eq 50. The only zero term is the case where two electrons are destroyed on the same orbital with same spin. Was this your intention? Could you clarify, or add a section in the appendix with the whole derivation in details?

We apologize for the unclear definition of the delta. The second interpretation is correct, however as these are redundant factors we removed them from the equations in accordance with comment 8.

10. Another way to see that the product of delta functions that appear in eq.(7) are incorrect (unless redefined as above) is to look at the numerator of eq.(9). If there is δ_{o_1,o_4} as a prefactor of D in eq.(7), it must be a prefactor for the whole numerator of eq.(9). The numerator of eq.(9) is correct I think, but not the delta functions in eq.(7).

See the two points above.

11. In eq. 7 : If we follow your delta, why are there $1-\delta$ for all terms except for the P term? Could you clarify? When I redid this calculation, nothing mathematical would have removed a $1-\delta$ over orbitals.

For the P-channel, the orbital restriction which comes from the definition of the interaction restricts exactly the orbitals which occur in the $1-\delta$ term to be the same, so that the delta is always one and can be ignored. This issue probably arose due to our misleading wording in the definition of $\bar{\delta}$ and we apologize for the confusion this caused.

12. In eq. 7: I think there is a typo : There is a sum over spin s'_4, but primed variables already means that they are summed over in your notation.

This is not a typo and yes, writing out the summation is redundant, but here we wanted to stress that due to this summation, there is an ambiguity in the way one defines the Ansatz equations. To make the origin of the ambiguity more explicit (and since we refer to it in the text) we wrote out the summation explicitly here.

13. Eqs 9 through 14 : I do realize the ambiguity in the spin or spin-independent Ansätze, I have three interrogations upon this realization: the first one is that the SU(2) Symmetry on the interaction U allowed to remove the spin-dependency of the interaction term, why is it ok to reinsert it? The second one is how can you differentiate the two different interaction terms of eq 50 with a spin independent interaction? The third one is that while trying to recover this Ansätze, I came upon the realization that we have to assume that the Ansätze are all decoupled, for D, C and P what is your insight on that?

We thank the Referee for this feedback. In a way the issue here is that there is not only one possible Ansatz but there is a family of Ansätze, which all do lead to formally equal left- and right-hand sides in the equal-time, equal-position, equal-spin limit. There is no a priori right choice. The choice we made is that we renormalize channels independently, as pointed out by the Referee in the third question. They do not necessarily have to have that form. Concerning the first question - the interaction we ultimately obtain from this is again spin independent and has the correct form, which we expect from a general SU(2) symmetric interaction tensor. Thus a posteriori it is allowed to make both choices since neither break the spin symmetry.

The second question focuses on how we decompose a formerly spin dependent interaction, such as the one the Referee refers to in Eq. (50) or a Hubbard-Kanamori interaction. Both of these can be written in the form of Eq. (3), which allows us to extract the spin-independent part we need for all of our calculations.

14. In eq.8 the global sign for P is incorrect if eq. 7 is correct.

We thank the Referee for pointing out this mistake. We fixed it and extended this part of the derivation to make clearer.

15. Why are the indices of D interchanged in going from Eq.(8) to Eq.(9)?

We wanted to bring the Ansätze into the same form for better readability for the following discussion. We undid this rewriting of the index order in the resubmission.

16. Eq. 15: Please comment on why you did not put any (r, τ) dependence.

The self-energy we obtain is local and static, thus we left out the dependence on (r, τ) for brevity. We explicitly added the dependence again in line with comment 6 of the referee.

17. Eqs. 15 and 16 : Only label one number for the whole eq.

Thank you for spotting this mislabeling. We fixed this in the resubmission.

18. Eqs. 21 and 22, you analyze the functional derivative contribution of the Ansätze, but only for one term of D and one of C. What about the P coefficient? Also, is it the same result in the case where the Ansätze is spin-independent?

The P coefficients follow the same structure as the two examples and therefore cancel as well, which is why we initially did not write these again. To make the derivation more transparent we added the complete calculation and all arguments required to Appendix C. The calculation for TPSC3 is identical to the one for TPSC5 but leads to fewer terms.

19. What assumptions are involved in claiming that the functional derivatives on the D Ansätze coefficients of eq. 15 vanish in eq. 21.

Knowing that the base model is SU(2) symmetric, we can rewrite the Ansätze in both cases as symmetric functionals with respect to $G^{\uparrow}$ and $G^{\downarrow}$ in the limit of zero source fields. This is the only ingredient required to argue why these types of contributions do cancel each other. In agreement with point 18, we added the explicit derivation for TPSC5 into the appendix. Furthermore, we removed the parts of the calculation which were in the main text, as they were rather confusing and not presenting the full picture. We thank the Referee for pointing out this possible source of confusion.

20. Eq. 21 : I think the left-hand side of the eq. needs to have a sum over s_4? Also, I think that normally, there should be a sum of the orbital indices o_4 of $D^{s_4,\sigma}_{o4,o1}$, which is not the same index as the one in the Green's function of the functional derivative.

We suppressed the summations as for each summand the argument for why it vanished works identically. The idea was again to keep it as simple as possible to make it easier accessible. Again, this is now part of the newly added appendix and therefore not found in the main text anymore.

21. Eqs. 23 and 24: Following your expression for the self-energy in eqs. 15 and 16 and the definition for Vertex in eq. 20, the sign in both eqs. 23 and 24 should be the opposite.

The Referee is correct, there was a missing sign in the definition of the vertex, as well as a sign mistake in the derivation which together gave the correct result. We rewrote the derivation and also changed the channel in which we are performing the calculation, such that the equivalence of the TPSC3 vertex and the vertex in plain Hartree-Fock becomes more obvious.

22. In eq. 34 through 38, it took me a while to understand that 1/2 meant either 1 or 2 and not spin half. Could you maybe just mention a clarification? Once I understood, the notation made sense, but it would be easier for a first-time reader if you put a small explanation or example.

We are sorry that the notation caused confusion and added an explicit statement above the equation.

23. In Section II.A : You write “We include both the particle-hole symmetrized and the usual version of the Ansätze in our implementation.“ Does usual mean the one you developed above? What is the particle-hole symmetrized Ansätze? Did you take it from another reference? If so, please add the reference and the eq. number, if you symmetrized your Ansätze, please show the final eq. of those.

We symmetrized the Ansätze we obtained before (here referred to as usual, now differently worded) analogously to Ref. [90]. To make the paper self-contained we added their explicit form in the appendix.

24. In Section II.A : You write “Apart from these differences, the sum rules utilized in Ref. [87] and Ref. [88] are a subset of the sum rules employed here, and we checked that by constraining the eqs. we can reproduce the previous results.”. Could you add an appendix on those verifications, what are the constraints that you added? I obtained the same sum rules as you did, but not the ones in the reference you mention.

We added an appendix with an explicit construction on how to reproduce both references. This requires to pick specific sumrules and to restrict both the charge and spin vertex in a specific fashion.

25. After eq 50 : You talk about how U=U’, but you did not mention U’ before, please define it or refer to its definition.

We thank the Referee for pointing out this missing explanation. We added it before introducing the interaction part of the Hamiltonian.

26. Between eqs. 50 and 51, about half-filling : What is the definition of your half-filling? One electron per orbital, or per site? Also, could you explain a little more on how you obtain eq. 52 by explaining the values of nsi,sjoi,oj in the half-filling definition? It could be added in an appendix.

We thank the Referee for pointing out this potential hurdle to understanding the main point of this section. We added more details to the calculation and specified what we mean by half-filling.

27. Eq 49 : It looks like the numbered orbitals are mixed up, because the right-hand side of the eq. does not close a bubble if we follow your definitions in the appendix.

We thank the Referee for pointing out the unclear notation. Here, we performed an index swap to bring it into a matrix product form. We see now that this was badly explained and removed the reordering to be in line with the appendix definition.

28. Eq. 57 has two c† in the hopping term. Is this intended? Is it Hermitian?

Thank you for catching this typo. It should of course be $c^\dagger c$.

29. Section III : You did not mention what was the size of the real-space and reciprocal space used to do TPSC calculations. In figure 3, you seem to be doing TPSC for two site, but even in the single-orbital case, TPSC is not valid below a 64 site x 64 site, and normally it is more used in 128x128 or 256x256, and even 512x512 in the low temperatures regimes. Could you please mention what is the system size that you are using? If it is different for different benchmarks, please mention each time. Also, if it is a 2-site only, then it is expected that TPSC fails, yes.

We thank the Referee for pointing out the missing details on our numerics. We added the information about the numerical setup in the caption of the figure. Here, in the example we indeed do a two-site model and indeed it is expected to perform poorly, but the aim was to illustrate the fundamental issue of the method in this limit. Whether it is single orbital or multi orbital, with this Hamiltonian makes no difference at all. This can be attributed to the underlying Hartree-Fock decomposion. We rewrote the section to make the aim of our test more transparent. Furthermore, we emphasized more strongly that this is not primarily an issue of TPSC, but originates from the underlying Hartree-Fock decomposition.

As a last point let us emphasize that such large momentum meshes are only required at lower temperatures or stronger interactions, i.e., when the correlation length becomes substantial. Since we perform calculations at $\beta = 2$ and relatively weak interactions, we found the calculations to be converged on a 24×24 momentum mesh.

30. Section III: Benchmarks and models : It could be a good thing to recall the different interaction terms of eq. 7 and give the values you used in the calculations. For both eqs. 56 and 50. Also, in the case of spin-independent Ansätze, how did you differentiate the (U-2J) and (U-3J) in eq. 56?

We thank the Referee for this comment. To address it, we added a short section, where we first explain how one obtains these components and then explicitly give them for the studied Hubbard-Kanamori interaction. In short, the idea is to pick a spin component in which only one of the Kronecker delta products in the definition of the interaction tensor is nonzero. In a second step one uses the Kronecker-deltas to split the spin independent interaction tensor into the channels. The $U - 3J$ term is only appearing indirectly – it is not contained in the spin independent tensor but only appears once the full spin dependent tensor is reconstructed. The values of U and J are always given in the plots.

31. Section III.A, page 8, second paragraph : I think you should add a reference on the results of TPSC for the single orbital model.

The Referee is right. We added citations to single-orbital TPSC works at the point where we make the comparison to the single orbital result.

32. Section III.A, page 9, first paragraph : The sentence : "As an illustration, let us considering a dimer at half-filling with a Hubbard- Kanamori interaction." I think you mean "[...] let us consider a dimer [...]".

Thank you for pointing out this typo.

33. Section III.A, page 9, first paragraph : About the illustration of a dimer at half-filling with a Hubbard Kanamori interaction, you mean the Hint of equation 56? If so, I think it would be best if you moved that to section III.B. where eq. 56 is already introduced. If not, could you add the details (eq. number): which interactions exactly you are referencing to? Also, could you show what are the different TPEVs? Which one of the TPEVs is represented by the Hartree-Fock decoupling? TPSC is not expected to be valid in the atomic limit, t=0. So your comment about the fact that the basics ideas of TPSC breaks down is correct, but this was also expected, you could add a reference, from many TPSC papers.

We rewrote the paragraph in question to make it better accessible by explicitly stating the model as well as explicitly deriving the eigenstates and the corresponding eigenenergies. The explicit form of the states also allows one to read off which TPEV's are influenced by these. Here, the central goal is trying to understand where the issues we observed in the analytical calculations stem from. For this, it is instructive to look at the Hartree-Fock decoupling of the dimer in comparison to the exact result. We observe the exact same issues we also found in the periodic case, leading us to conclude that TPSC inherits this shortcoming from Hartree-Fock.

The basic idea of TPSC we are referring to here is the idea of correcting the Hartree-Fock double occupancies by sum rules in a self-consistent way. This idea does (sometimes) not work in multi-orbital systems as the Hartree-Fock is completely off. In this case the self-consistency loop converges to a solution which does not capture the full local structure.

34. In section III.A : after eq 50 : you write "This interaction in combination with the kinetic term leads to a vanishing Σ_0" what is your Σ_0? The non-interacting self-energy? The non-interacting self-energy should be zero, yes, but because the non-interacting green's function is the one that solves the non-interacting Hamiltonian, which solves exactly, thus there is no self-energy in its expression. Please explain what is your Σ_0.

In single-band TPSC one usually redefines the chemical potential such that the lowest order selfenergy diagram (GU) is absorbed. This procedure is often not possible in a multi-orbital system due to off-diagonal components of the generated self-energy, so we have to consider the contribution of this in all our calculation. This is for example very nicely discussed in Ref. [91]. We also added a reference to the Appendix in which the explicit form of this self-energy is given.

35. In section III.B., in the second paragraph, you explain why the higher values of J works better for TPSC while looking at the different groups of spins. You talk about the eigenvalues, but there is not further explanation on how to obtain those eigenvalues. I guess it would come from the diagonalization of the Interaction part only of the Kanamori Hamiltonian, but it would be nice to have more description about that, in an appendix perhaps, or maybe you could add a reference where it is well explained?

We added a more detailed explanation of the diagonalization in Sec. 3.1, as both sections rely on this simplified case to clarify what is happening. We hope that this discussion does give sufficient context, so that this is more easily understandable now.

36. In section III.B. third paragraph, you write " This also indicates that this specific part of TPSC performs better at lower temperatures (since it is basically a ground state targeting approach) - however, the local and static approximation of the vertex becomes more and more inappropriate at lower temperatures which is why TPSC typically fails at low temperatures." TPSC is not "basically a ground state targeting approach". It is actually far from it. It is not valid in 2D deep in the renormalized classical regime, which is a region of the phase diagram in temperature and density that starts at finite temperature and goes to zero temperature. Also, each part of the sentence separated by "-" are two contradicting statements. In general, one would say that TPSC can reach lower values of temperature than DMFT, but one needs to be careful in the grid definition, because the correlation length should be smaller than the lattice size to capture long wavelenght correlations. Many of these explanations can be found in [Gauvin, Phys Rev B 108, 7 (2023). DOI : 10.1103/PhysRevB108.075144].

Indeed, TPSC is not a ground state targeting approach, but Hartree-Fock is (the "it" refers to Hartree-Fock and not TPSC as a whole – we rewrote the sentence top make this clear). The issues which we observe arise from the problems inherited form the Hartree-Fock decomposition, which propagate into the TPSC self-consistency (as we found already in Section 3.1). We rewrote the parts where we explain this issue in more detail, to be more specific. We do not claim that TPSC is unable to reach lower temperatures than DMFT, all that we want to claim is that at very low temperatures the locality approximation becomes usually unsuitable, as the interaction vertex become more and more localized in momentum space, and thus typically long ranged.

37. Appendix A, eq. A23 : Normally, the two derivatives w.r.t. τ_a and τ_b for $G(\tau_a, \tau_b)$ give a different order for ΣG : one is ΣG and the other is $G\Sigma$.

We cleaned up the whole appendix and thank the Referee for pointing out this typo.

38. In eq. A14, what does S0 superscript mean?

S^0 refers to the charge case in which the applied transformation is the identity matrix. We added a sentence clarifying this.

39. The definition of susceptibilities in eq. (A11) does not lead to the matrix structure in eq. 25.

The Referee is correct, we performed a swap of the second and third index, such that we do arrive at a matrix-matrix product. We restructured the main text such that no index reordering is necessary anymore.

Recommendation

Ask for major revision