March 3, 2025

Response to Referee 1

The manuscript presents a theoretical analysis of the Emery model for high-temperature superconductors using single-site Dynamical Mean-Field Theory (DMFT). DMFT only introduces local self-energy corrections, hence it is usually discarded as a method to study models for high-temperature superconductors, in which non-local correlations are universally considered fundamental. Yet the present work shows that, in contrast with the single-band Hubbard model, the three-orbital Emery model shows important non-local correlations owing to the multi-orbital character. This point is well discussed in the manuscript in general terms and its consequences are explored. In particular, the authors find that the q=0 uniform magnetic susceptibility χ is remarkably different from the single-band Hubbard model (shown in the appendix A) and, remarkably, it reproduces some trends of the experimental data in cuprates, reproducing in particular the qualitative behavior in the pseudo gap region. This is particularly surprising and intriguing, as the DMFT single-particle spectra fail, as expected, to reproduce the pseudo gap. The authors discuss some arguments behind this seeming discrepancy (lack of feedback on single-particle properties of the behavior of the two-particle response given by the spin susceptibility) and the perspective to overcome it by using DΓA.

I find this result very intriguing and I support the publication of the manuscript in SciPost Physics. The use of the very well established single-site DMFT and the straightforward calculation of the uniform susceptibility computing the response to a small static magnetic field make the conclusions very solid and they identify an non trivial effect. I am also intrigued by the simple two-site model described in the appendix B. Indeed this oversimplified model captures the main trend of the DMFT calculation. This success also suggests that the results discussed here can be related to those for the periodic Anderson model (PAM). Indeed in Phys. Rev. B 85, 235110 (2012) the PAM has been solved within DMFT, finding the formation of Zhang-Rice singlets similar to those reported here. In that paper the authors compute the local (impurity) spin susceptibility which however has a Curie-like behavior. So, I am wondering if this is the case also for the Emery model, while the non-Curie behavior is found in the q=0 response, which is notoriously different from the local counterpart in DMFT.

As I detail below, this comparison with the local susceptibility (and possibly with the PAM) could help to strengthen the manuscript.

Another suggestion I have is to consider a title/presentation which makes it more explicit that the two-particle observables mentioned in the title coincide with the uniform susceptibility. The current version of the title gives the incorrect impression that the manuscript would show dynamical two-particle observables which have been computed in a number of papers from the Vienna group and coworkers.

I detail below also other points that I would like the authors to address in order to meet the criteria for publication. A revised version meeting all my points should be published in SciPost Physics.

Response: We appreciate the Referee's comments, their judgement that our results are *"intriguing"* and *"non trivial"* and that our conclusions are *"very solid"*. We welcome their support for publication in SciPost Physics after the requested changes have been implemented.

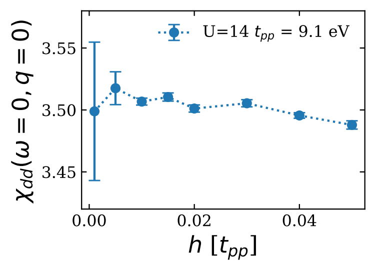

Figure 1: Dependence of the static susceptibility at $\mathbf{q} = 0$ on the applied external field h. The constant behavior around the used value of $H_{\text{ferro}} = 0.01 t_{pp}$ shows that we are in the linear response regime.

Requested Changes

1. I would eliminate the sentence about static mean-field reproducing the undoped system. While an AFM is typically found, the mean-field description is quite far from the actual situation and from DMFT.

   Response: We agree with the referee's comment and have edited the corresponding remark in the manuscript accordingly.

2. At line 10-11 of page 2, I would stress more that the drop of the Knight shift is generally interpreted as resulting from the reduction of spectral weight, while the present calculation finds it without a spectral pseudo gap.

   Response: We followed this suggestion of the referee and added a sentence at the end of the introduction stressing the conclusion that the drop in the spin susceptibility as a function of temperature does not necessarily require the opening of a gap in the single particle spectrum but could be explained with the formation of rather localized oxygen-copper singlet configurations.

3. The authors should discuss what is the choice of the double counting used here and whether also the interactions are extracted from ab-initio calculations. I would also mention the value of the d-p repulsion even if this is neglected, since this is probably the most important simplification introduced in the model

   Response: The double-counting correction and interaction parameters were taken previous research cited in refs [34,35]. This includes an atomic-limit double counting correction, which is chosen to reproduce the charge transfer gap of about 2 eV [$E_{dc} = U_{dd}(n_{d0} - 1/2) = 3.1$ eV with $U_{dd} = 14\, t_{pp} = 9.1$ eV and the density of d-orbital $n_{d0} = 0.84$ obtained from ab-initio calculations].

   Concerning the neglect of U_{pd}: When U_{pd} is treated on a static mean-field level, it can be regarded as a renormalization of the splitting of single particle energies of d- and p- orbitals and can be absorbed into the double-counting term, which was already chosen to reproduce the experimental charge transfer gap.

   We added a comment in the revised version of the manuscript.

4. The authors write that the susceptibility is computed for a value of h≈5 meV. Why ≈ and not just equal? Do I understand correctly that the authors simply use a linear approximation connecting zero field with this value of h? Did they test if this value is small enough for the present calculations? (observing that no change is introduced by reducing h)

   Response: We thank the referee for pointing out this confusing notation which, indeed, was unintentional. The applied field we used was $H_{\text{ferro}} = 0.01 \cdot t_{pp} = 6.5$ meV. We fixed the notation in the new version. Moreover, we indeed made sure to be in the linear regime (see Figure 1).

5. minor: I think that the Clogstone-Jaccarino plot should be explained to the general readership

   Response: We agree with the referee. The linear relation between the night shift and the uniform spin susceptibility was first proposed by Clongston and Jaccarino [PhysRev.134.A650(1964)], with the slope related to the hyperfine coupling constant as proposed by the later research [Physica C: Superconductivity, 157(3)561-570(1989), PhysRevB.42.167(1990)]. This linear relationship can be visualized by plotting the Knight shift as a function of the susceptibility with temperature as an implicit parameter, which is referred to as the so-called Clogston–Jaccarino (CJ) plot. We now added this information to clarify the meaning of the CJ plot.

6. As mentioned above, I would suggest a title/presentation making more explicit that the two-particle observables mentioned in the title coincide with the uniform susceptibility.

   Response: We followed the suggestion of the referee and changed the title to be more specific to our results.

7. I would add if possible one example of the local spin susceptibility to compare with the q=0 response. This can also be connected to a comparison with the periodic Anderson model.

   Response: We thank the referee for this suggestion. Indeed the comparison of the impurity susceptibility to the uniform one is interesting as it shows an inverted doping trend. This is indeed in complete agreement to results on the periodic Anderson model (Amaricci et al. 2012) and indicates that - as a function of doping - the contributions of local copper moments to the full magnetic response is decreasing and that fluctuations around the Fermi-level become increasingly important for the uniform (q=0) response. We added a comment on this observation in the main text and a figure of χ_{imp} in the appendix.

8. In Fig. 2 the x and y range of the panels (a) showing the self-energy are cut in a rather extreme way. I would extend them to highlight better the evolution of the real and imaginary part of Σ_{dd}

   Response: The plot-range was chosen to emphasize the Fermi liquid properties of the self-energy, specifically the linear and quadratic behavior near the Fermi surface as mentioned in the main text. We now added insets showing the self-energy on a larger scale as suggested by the referee.