REPLY TO REFEREE 1

We thank the Referee for carefully reading our manuscript, for the positive evaluation of our work and her/his constructive observations.

The Referee has asked us to consider specific points to be addressed prior to publications. We have considered all of them very thoroughly and included the corresponding changes into the manuscript text, figures and the appendix. Specifically, we report below a detailed reply to all the observations of the report.
The main questions posed by the Referee are the following ones:

The central statement of the paper is that the spin channel provides the most important contribution to the frequency dependence of charge susceptibility, and capable to describe various features of charge susceptibility observed previously in Ref. [22]. To clarify the relative role of vertex corrections and the susceptibility itself, it would be helpful to see in Figs. 3,5,8 the spin contribution with $\lambda_{spin} = 1$.

In order to better clarify the role of the vertex correction to the susceptibility in a coherent way w.r.t. the flow of the paper, we have included a comparison of the spin contribution to the generalized charge susceptibility obtained with and without approximating $\lambda_{\mathrm{sp}} = 1$ in Fig. 12 in Sec. 3.4 and added a corresponding explanation in the text. Panels 2-4 of this figure clearly show the importance of λ_{sp} in the local moment and Kondo regime of the AIM and the HA. The relative role in the local moment regime is an enhancement of the absolute value with respect to the non-interacting limit. This situation is reversed in the Kondo regime, where the spin contribution is largely suppressed in absolute value due to the screening effect of the electronic bath. As described in the text here the Hedin spin-vertex has values smaller than 1 i.e. its non-interacting limit. For the DMFT solutions in the corresponding two regimes (not shown) the very same considerations apply.

To provide a full frequency picture of the effects of this approximation, we have also added the new Appendix C, where a colorplot showing the whole frequency structure (including the frequency off-diagonal elements of the SBE spin contribution) is reported and briefly discussed.

It is not fully clear what is shown by dashed red lines (which are explained as the contribution proportional to χ_s, but not explained in more details).

Regarding the explanation of the red dotted line (note that it was incorrectly indicated as "red dashed" in the main text, which we have corrected in the revised manuscript) in Fig. 3, 5 and 8, we agree that it needs to be extended. Specifically, the spin contribution of Eq. (11) can be formally split into two parts by inserting Eq. (10) i.e. $\frac{3}{2}[G_\nu]^2\lambda^{\rm sp}_{\nu,\nu'-\nu}(-U+U^2\chi^{\rm sp}_{\nu'-\nu})\lambda^{\rm sp}_{\nu,\nu'-\nu}[G_{\nu'}]^2$ (note that $\nu'-\nu$ is a bosonic frequency). Keeping only the latter term one gets $\frac{3}{2}[G_\nu]^2\lambda^{\rm sp}_{\nu,\nu'-\nu}U^2\chi^{\rm sp}_{\nu'-\nu})\lambda^{\rm sp}_{\nu,\nu'-\nu}[G_{\nu'}]^2$, which is precisely the one we indicated with the red dotted line. This specific SBE contribution, being directly proportional to $\chi^{\rm sp}(\omega)$ is of most interest for its transparent link to the physical spin response. In order to better clarify this point, we have now added its explicit expression in the caption of Fig. 3 and the main text.

It might be also useful if the authors provide plots of $\chi_s(T)$ and/or $T*\chi_s(T)$ (possibly in Appendix) to understand evolution of the susceptibilities in different regimes.

We agree that such a plot would be useful and have indeed added it together with the new Appendix D. It is also reproduced here for clarity.

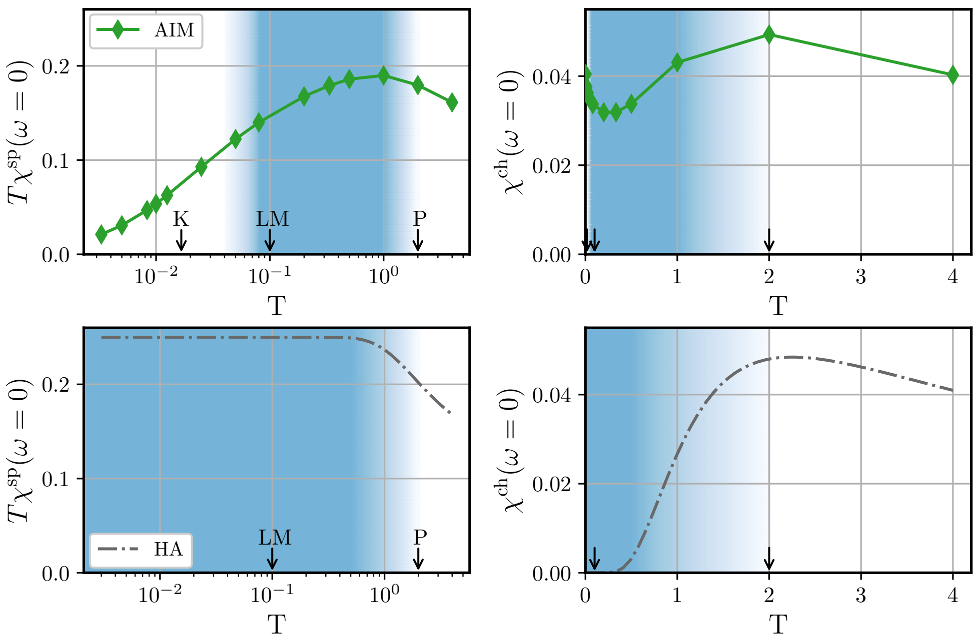

Figure 1: Temperature dependence of $T\chi^{\rm sp}(\omega=0)$ for the AIM (left upper panel) and the HA (left lower panel) plotted on a logarithmic scale and temperature dependence of $\chi^{\rm ch}(\omega=0)$ on a linear scale for the AIM (right upper panel) and the HA (right lower panel). The blue-shadowed areas indicate the parameter regimes associated to the local moment physics in the different models. The black arrows show the location of the respective temperature regimes described in the main text (K=Kondo, LM=Local Moment, P=Perturbative).

Is it also a coincidence that the behaviour of different channels in high-temperature and Kondo regime is somewhat similar (although it is different by magnitude)?

We assume that the question is mostly referring to the comparison of the data plotted in Fig. 3 and Fig. 8, because only there we show the separate contributions of all channels to the diagonal frequency entries of the generalized charge susceptibilities in the perturbative and in the Kondo screened regime. In this case, we should first emphasize that –on a general level– one expects that all contributions of our SBE decomposition display larger intensities for lower frequencies along the diagonal, due to their asymptotic decay at high frequency. Further, their specific signs (positive for the bubble, negative for the spin contribution, etc.) appear to be fixed at half-filling, where special particle-hole symmetric properties hold (such as, e.g., that the on-site Matsubara Green's function is purely imaginary). Hence, to a first glance, the structures of the different contributions along the diagonals (unless they are not completely suppressed) might look qualitatively similar. It is also true, however, that beyond this general observation, additional similarities can be noted between the low-frequency perturbative and Kondo regime, due to the screening effects active in the latter case. These are responsible, for instance, of the low-frequency increase (w.r.t. to the local moment regime) of the bubble term as well as of a moderation of the suppressive contribution of the spin channel, which both drive the (relative) low-T revival of on-site charge response. Obviously, the similarity is not complete. By looking at a more quantitative level, differences also emerge, such as the much smaller/larger contribution of the singlet channel/U_{irr} in the Kondo w.r.t. to the nonperturbative regime, as well as the almost perfect identification of the spin contribution with its component proportional to the physical susceptibility in the Kondo regime. Eventually, even more evident differences between the perturbative and the Kondo regime can be observed when comparing the off-diagonal frequency structures (e.g., by comparing the third column panels of Fig. 4 and Fig. 9, where the corresponding multi-boson contribution is shown).

The title of the paper "Non-perturbative intertwining..." looks to me somewhat misleading. Indeed, the authors consider purely perturbative contributions to the charge channel (apart from the irreducible one, which does not play big role in their results). All the non-perturbative information is therefore hidden in the triangular spin vertex and spin susceptibility, which behaviour the authors almost do not analyze. I suggest the authors also to extend the discussion on the non-perturbative aspects in Conclusion and text of the paper.

This question is of high importance for our work and certainly requires additional clarification (both in the reply and the revised text), as it also touches relevant aspects, which have emerged during the presentation of our results to other colleagues in informal discussions and conferences. Indeed, the Referee is quite right in noticing that one of the pivotal effect we described in the paper, i.e. the sign flip of the diagonal elements of the generalized charge susceptibility is driven by the SBE (and two-particle) *reducible* scattering processes in the spin channel. In the SBE decomposition, however, no perturbative assumption is -a priori- made, and, as the Referee also noted, the two main constituents of the spin SBE-contribution clearly identified as responsible for the systematic suppression of the diagonal entries of $\tilde{\chi}_c^{\nu\nu'}$, namely (i) the (static) physical spin response and (ii) the triangular spin-fermion vertex are the exact ones (for the corresponding case considered) without any a priori restriction to any perturbative approximation.

It is important to emphasize, here, that precisely this clear-cut identification via SBE decomposition, which was missing in previous studies (including ours), allows to unveil the physics underlying the breakdown of the self-consistent many-electron perturbation-expansion. In particular, in previous studies, it was just noticed, essentially on a mere empirical basis, that in several fundamental models for strongly correlation, the suppression of on-site charge response occurring the local moment regime of the corresponding phase-diagrams was mostly driven by a strong suppression of the lowest frequencies diagonal entries of the generalized charge susceptibility and, not, e.g., by a generic/uniform reduction of all its matrix elements (which would have been also possible[1]. This specific feature is the one determining the breakdown of the self-consistent perturbation expansion, as the suppressed (and then even negative) diagonal entries of $\chi_c^{\nu\nu'}$ causes a sign-flip of its eigenvalues, and, hence, whenever one eigenvalue vanishes, the associated divergences of the irreducible vertex function, the non-invertibility of the corresponding Bethe-Salpeter equation (BSE), and the crossing to physical and unphysical solutions in the Luttinger-Ward functional formalism.

A legitimate question posed by many colleagues (as well as by ourself) was then to understand whether the suppression of the on-site charge response associated to a local moment should necessarily occur in this precise fashion (which then unavoidably leads the perturbative breakdown), and, if yes, why this is the case. The identification of the (overall negative!) spin-SBE contribution to $\tilde{\chi}_c^{\nu\nu'}$ as the main suppression mechanism of the on-site charge response, presented in this manuscript, has finally provided a clear-cut answer to these questions, in terms of the two main ingredients of the spin-SBE scattering processes mentioned above. Specifically, in the local

[1]For instance this may indeed happen, in the case of a reduction of the density of states of the non-interacting Hamiltonian

moment regime (i) the long[2] life-time of the on-site spin correlations is directly reflected in a selection rule of the major suppression effects of the local charge-fluctuations for ν ν' (whereas $\nu \equiv \nu'$ in the "perfect" HA case, where the local spin is a conserved quantity) (ii) the spin-fermion coupling (triangular vertex) gets enhanced w.r.t. its perturbative value of 1 at low-fermionic frequencies ν. Evidently, the combination of (i) + (ii) explains why the suppression of the on-site charge response, which is unavoidably associated to the formation of a local moment *must* occur in the precise way observed in the previous work, leading necessarily to a divergence of the irreducible vertex, and, hence, to the breakdown of the self-consistent perturbation expansion. Our analysis, thus, rigorously clarifies the physical nature of the perturbation theory breakdown in all fundamental models considered: The simultaneous enhancement of the on-site magnetic static response and suppression of the on-site charge one, which are both, indeed, intrinsic features of the local moment physics. Hence, any (self-consistent) perturbative approach is bound to fail in describing a proper suppression of the charge fluctuations in the presence of a local magnetic moment, due to the intrinsic impossibility in self-consistent perturbation theory of flipping the sign of any of the eigenvalues of $\tilde{\chi}_c^{\nu\nu'}$, which will remain all positive, as in the corresponding non-interacting case of the model considered. This specific (but relevant!) drawback of self-consistent perturbation approaches has been explicitly observed, e.g. in (truncated) functional renormalization group (fRG) and parquet approximation (PA) calculations, where the local charge response was found to monotonically increase when reducing the temperature even in the local moment regime, reflecting the too weak suppression of $\tilde{\chi}_c^{\nu\nu'}$ for $\nu \sim \nu'$ (indeed the diagonal elements of $\tilde{\chi}_c^{\nu\nu'}$ remain positive in all fRG and PA dataset). This way, one can eventually understand that the breakdown of the perturbative description in Hubbard model systems is intrinsically rooted into the strong communication between the different physical sectors (magnetic vs. charge, but also particle-particle/pairing), which is essential to yield a self-consistently coherent picture of the local moment physics[3] in its entirety, where the enhancement of the static local spin response must consistently occur together with the suppression on-site charge (and pairing) fluctuations. We note in passing that this strong interplay between the different sectors also represents a crucial ingredient for the (indeed nonperturbative in U!) dynamical mean-field theory description of Mott metal-insulator transitions.

We note here -although this is beyond the scope of the present work- that, consistent with our considerations, the "unphysical" solutions[4] obtained in

[2] Actually even infinite in the "perfect realization of the local moment", i.e. the Hubbard Atom

[3] Note that evidently the same consideration will apply, *mutatis mutandis* to the formation of local pairs in the case of an attractive on-site interaction (negative U)

[4] According to several studies bold diagrammatic Monte Carlo resummations do con-

bold (=self-consistent) diagrammatic Monte Carlo after crossing the first vertex divergence line (i.e. in the nonperturbative regime) are precisely characterized by an unphysical metallicity even in the local moment regime, with a too large charge mobility and even a value of double-occupancy increasing with U.

Another point: In the beginning of Sect. 3.2 the authors mention formation of relatively flat part of $T * \chi_s(T)$ and refer to Refs. [38,49]. However, these references refer to Anderson impurity model, where flat part is absent ($T * \chi_s$. monotonously increases). I suggest the authors to cite the papers [22,25] (and possibly others) instead.

Indeed, if one scans the whole temperature range from the high-T perturbative regime down to $T \to 0$, the quantity $T\chi_s(T)$ for the AIM we considered displays a non monotonous behavior with a rather broad maximum at about $T \leq \frac{U}{2}$. We agree, nonetheless, with the Referee, that our statement about a "flat part" of the quantity $T\chi_s(T)$ was rather imprecise and, in general, difficult to be quantified. For that reason, and also in the light of the observation made by the second Referee, in the revised manuscript we have dropped the qualitative statement mentioned above and have refined the corresponding discussion, which also benefited from the additional inclusion of a dedicated figure (showing the behavior of $T\chi_s(T)$ for the HA and the AIM) in the Appendix.

verge also in the nonperturbative regime, albeit not to the correct/physical solution: this is referred to as "misleading convergence" of the self-consistent perturbation expansion, which appears after crossing the first vertex divergence line.

LIST OF CHANGES

1. We added a sentence in Sec. 2.2 to clarify the phase space studied in our DMFT calculations.
2. Sec. 2.3 was modified to clearly explain the aim and importance of this work.
3. In first part of Sec. 3 we added a paragraph to discuss the general features of the crossovers observed in these systems and to specify the reasons for the chosen parameter sets of this work.
4. In the caption of Fig. 3 the explicit formula describing the red dotted line was added.
5. An explicit expression for the red dotted line in Fig. 3 was added in the text of Sec. 3.1 and we corrected the word "dashed" to "dotted".
6. We corrected a typo: we changed the label -5 to -4 in Fig. 3,5,8
7. We added the new Fig. 12 comparing the diagonal part of the spin contribution to $\chi^{\mathrm{ch}}_{\nu\nu'}$ with and without approximating $\lambda^{\mathrm{sp}} = 1$ and a corresponding description at the end of Sec. 3.4.
8. We modified the text of Sec. 4 highlighting the intent of this paper and, in particular, the nonperturbative character of this study. We also added a short statement about the newly shown results for $\lambda^{\mathrm{sp}} = 1$.
9. We added Appendix C with the new Fig. 13, showing the full frequency structure of the spin contribution under the approximation that $\lambda^{\mathrm{sp}} = 1$.
10. Further, we added Appendix D with the new Fig. 14 showing the evolution of the physical spin and charge response function with temperature.