REPLY TO REFEREE 2

We thank the Referee for the careful review of our manuscript, for her/his overall positive evaluation, as well as for her/his detailed observations. Below, we detail our Reply to all specific points raised in her/his report:

The manuscript presents a very detailed study and is written in a clear manner. However, before I can recommend this work for publication, I would like the authors to address to the following questions:

My main question concerns the interpretation of the results obtained. The authors relate the transition from the high- to intermediate-temperature regime, and consequently the appearance of the specified frequency structure of the generalised charge susceptibility, to the formation of the local magnetic moment. From my point of view this relation is not very well explained and is not well justified.

If this transition is identified by the divergence of the irreducible vertex function in the charge channel, it can only be related to the breakdown of the many-electron perturbation expansion following Refs. [1,3,7,9,10,13-16,18,19] cited by the authors. In Ref. [22], the divergence of the vertex function was associated with the formation of the local magnetic moment, as in the low-temperature and strong-coupling regime the "divergence curve" aligned with the Kondo temperature. However, there was no justification that the divergence of the vertex function could be connected to the formation/destruction of the local magnetic moment in the high-temperature regime.

If the transition is identified by (quoting the authors) "a relative flat (Curie) behavior of the quantity $T\chi^{sp}_{\omega=0}(T)$," then this condition is imprecise. It would be helpful if the authors show the results for the local susceptibility and explain how they identified the transition point. In fact, the deviation from the Curie behavior of the spin susceptibility is rather smooth (see, e.g., [PRB 99, 165134 (2019)]), which usually does not allow one to accurately pinpoint the formation of the local magnetic moment (see, e.g., [arXiv:2112.02881]). In addition, in [PRB 105, 155151 (2022)] it was argued that the formation of the local magnetic moment cannot be captured by the behavior of the static spin susceptibility, because (quoting the authors of that work) the spin susceptibility "cannot distinguish the fluctuations of the local magnetic moment from the spin fluctuations of the itinerant electrons that also contribute to the susceptibility, especially in the paramagnetic regime."

We thank the Referee for this comment. Indeed, the study of the relation between the spin- and the charge-sector in the different regimes (and especially of the local moment one) is one of the central points of our work. Hence, it is important that this aspect is presented in the most clear and convincing way in our reply and in the revised manuscript.

Let us start by stressing (what we have also done in the revised text) that it is *not* our aim, in this work, to introduce/define or even improve criteria for delimiting the different physical regimes. Indeed, as the Referee points out, this task would be quite hard (if not impossible from a purely rigorous perspective), considering that the different regimes studied in our selected models (HA, AIM, as well as the paramagnetic DMFT solution of the HM on the left side of the MIT) are separated by crossover regions and not by sharp phase transition lines.

Our goal is, instead, to precisely rationalize the mechanisms controlling, on the two-particle level, the physics of charge localization, which arguably is *"the other side of the coin"* of the local moment formation. In particular, we aim at eventually clarifying *how* the specific way in which the charge localization gets encoded in the corresponding generalized susceptibilities is linked to the physics underlying the local moment formation and (where applicable) its Kondo screening.

In fact (cf. Introduction and Sec. II), it was noted in previous works, on an empirical basis, that the freezing of the on-site charge response in the strongly correlated regimes of several basic models was associated to a marked suppression of the diagonal entries of the (corresponding) generalized susceptibility $\chi^{\nu\nu'}$ (which could become quickly negative at low-frequencies) and to a simultaneous slight increase of the off-diagonal elements.

However, the essentially empirical nature of such observations prevented to draw rigorous conclusions, leaving the question open (which was posed to some of us several times in conferences and discussions) *why* the reduction of the on-site charge response driven by correlations was occurring in this precise way rather than, e.g., through an uniform suppression of all matrix elements of $\chi^{\nu\nu'}$, or with a larger suppression of the off-diagonal ones, etc.

As the Referee rightly mentioned, this question is also tightly linked to problem of the breakdown of the self-consistent perturbation theory, since the specific (abovementioned) way in which the suppression of on-site charge fluctuations takes place is primarily responsible for the multiple sign-flips of the eigenvalues of $\chi^{\nu\nu'}$, and hence for all the related consequences (divergences of irreducible vertices, crossings of different solutions of the Luttinger-Ward functional, convergence to unphysical results of the many-electron expansion, etc.). Evidently, if the freezing of the on-site charge fluctuations had occurred in a qualitatively different fashion on the two-particle level than the way described before, it might have been possible for self-consistent perturbative approaches to capture the local moment regime physics (including the associated Mott insulating phase in DMFT).

We think that our diagrammatic decomposition provides a clear-cut answer to the question above. The obtained results precisely identify the scattering processes (i) mostly responsible for the progressive suppression of the low/intermediate diagonal frequency entries of $\chi^{\nu\nu'}$, i.e. those associated to the electronic scattering with a single spin mode, (ii) as well as those causing the slight overall enhancement of the off-diagonal terms, due to *multiple* scattering with collective bosonic excitations. It is important to stress that (i) explains then, in a perfectly natural way, why the freezing of on-site charge fluctuation due to the local moment formation happens in the specific way we observe it: The more well-defined the local magnetic moment will be, the longer will be its lifetime[5]. In (Matsubara) Fourier space this trend gets immediately reflected in a progressive frequency-localization of the suppressive effects originated by the corresponding single spin-exchange processes on the diagonal entries of $\chi^{\nu\nu'}$. At the same time, this lifetime effect, though crucial, would have not been enough alone to allow for a correct transfer of information between the different channels in the local moment regime. The latter is made possible by the simultaneous low-frequency enhancement of the corresponding spin-fermion scattering amplitude (i.e., of the so-called triangular vertex) w.r.t. its perturbative/asymptotic value. This diagrammatic identification, which appears numerically quite solid in the three model considered, provides an clear-cut explanations of the question why the freezing of the on-site charge fluctuations happens in the specific nonperturbative manner observed, and how the relevant information (enhanced on-site magnetic response, suppressed on-site charge response) gets transferred between the different physical sectors.

In this perspective, in the low-temperature limit of the HA (where the physics of the local moment is essentially perfect, up to vanishingly small exponential corrections of order $\sim e^{-\beta U}$), by estimating the minimal magnitude of the spin-fermion vertex to observe a sign-flip on the diagonal entries of $\chi^{\nu\nu'}$, we were able, finally, to clarify why the size of the frequency-region $[-\nu_{max}, \nu_{\max}]$ where (nonperturbative) negative diagonal values of $\chi^{\nu\nu'}$ are observed scales precisely as $\nu_{max} = \frac{\sqrt{3}}{2}U$. Indeed, the previously empirically determined scaling factor of $\frac{\sqrt{3}}{2}$ finds its most natural explanation in the prefactor of the single spin-exchange contribution, further supporting the validity of our analysis.

As we will detail better below, by revising our manuscript we tried to better emphasize the main goal of our study as well as the relevance of the results obtained in this perspective, and to refine/modify imprecise (and, eventually, intrinsically non-conclusive) statements about the borders of the different regimes.

On the contrary, the transition between the low- and intermediate--temperature regimes is clearly defined by the Kondo temperature. Therefore, it would be helpful if the authors specify the values of the Kondo temperature for the systems under consideration and provide an explana-

[5]Lifetime, which becomes infinite in the limiting case of the HA, to be regarded, in this sense, as the *"perfect gas" analog* for the local moment physics.

tion of how they were obtained, as the definition of the Kondo temperature for these systems is not unique. Actually, the issue regarding the formation/destruction of the local magnetic moment is even less clear for the Hubbard atom, which does not have a Kondo regime.

Following the suggestion of the Referee, in the revised manuscript, we have now reported the estimated value of the Kondo temperature (T_K) for the AIM considered, as well as of the "effective" T_K for the DMFT solution of the Hubbard model. The former has been extracted by the temperature dependence of the local magnetic susceptibility $T\chi_m(T)$, namely by matching it to the universal temperature behavior of the Kondo problem, following the procedure detailed in the Appendix A of Phys. Rev. B **97** 245136 (2018) as well as Sec. II in Supplemental Material of Phys. Rev. Lett. **126**, 056403 (2021). We note that this procedure yields, at the U considered, *even on a quantitative level* the corresponding textbook[6] wide-band limit value for T_K [see, Eq. (6.109) and ff. at p. 165–166 of Chap. 6.7 in A. C. Hewson, "The Kondo Problem to Heavy Fermions" (Cambridge University Press, Cambridge, 1993)], except for the (marginally small!) corrections due to the finite (albeit large) bandwidth of the bath electrons. The precise value of T_K in the energy units of our AIM[7] reads $T_K \simeq \frac{1}{65} \simeq 0.015$. As discussed Phys. Rev. Lett. **126**, 056403 (2021), in the strong-coupling regime (which applies to the value of U considered here) this (textbook) value of T_K is extremely well approximated by a specific condition on the lowest Matsubara entries of the generalized charge susceptibility, namely that $\chi_c^{\pi T, \pi T} = \chi_c^{\pi T, -\pi T}$.

As for the DMFT calculations, where the AIM plays an auxiliary role for the self-consistent determination the corresponding dynamical mean field, the procedure described above cannot be straightforwardly followed, as the auxiliary AIM itself (as well as its Kondo temperature) depends (for a fixed U) on the temperature itself. Here, however, by resorting to the criterion based on the lowest Matsubara-frequency mentioned above, one could determine the temperature at which the effective T_K of the corresponding auxiliary AIM is crossed, i.e. $T_K(T) = T$. For our DMFT calculations on the Bethe lattice, with $U = 2.2$ in unit of the half-bandwidth, its estimation yields $T_K \simeq \frac{1}{50}$. Finally, in the Hubbard Atom the local spin operator (as well the local charge operator) is a constant of motion of the problem, allowing to regard this system as an "ideal realization" of the local moment

[6]As T_K marks a crossover scale, as the Referee also mentioned, other definitions/criteria could have been chosen. For instance, by estimating T_K via the width of the corresponding Kondo-peak in the spectral function one typically gets larger estimates (up to five times!) than those obtained from the temperature dependence of $T\chi_m(T)$. The latter criterion represents, however, one of the most common choices made in the literature.

[7]Namely, an AIM with a constant DOS of the bath electrons in the interval $[-D, D]$ with $D = 10$, hybridization amplitude $V = 2$ and impurity on-site interaction $U = 5.75$

physics. The absence of any Kondo screening, yields perfect local moment features in the low-temperature limit, with negligibly small $e^{-\beta U}$ corrections. In that case, the local moment physics can be only be degradated by the thermal activation of the excited states (with 0 and 2 electrons, respectively), which occurs at temperatures of the order of the corresponding energy gap, i.e. $T \sim \frac{U}{2}$. Indeed, a brief glance on the temperature dependence of the susceptibilties, allows to appreciate how, in the case of the HA, both the fulfillment of the Curie behavior of the spin response, as well the corresponding exponential suppression of the charge response, become virtually perfect already at (or below) $T \sim 0.5 < \frac{U}{2}$.

To summarise, it would be helpful if the authors: 1) Provide clear specifications for the transition points between the high-, intermediate-, and low-temperature regimes and elaborate on how these temperatures were calculated. 2) Justify the relation of the mentioned transitions to the formation/destruction of the local magnetic moment, or alternatively, refrain from making such a connection. 3) Demonstrate that the change in the frequency structure is indeed happens at the transition point and not somewhere else in the phase.

Considering the questions raised by the Referee, and consistent with our reply, in our revised manuscript we have now refined the presentation of the aims of the paper, underlying that our main goal is not to provide univocal definitions for the crossover borders between the different regimes (which would be, *a priori*, an unfeasible task, due the lack of sharp phase-transitions in the cases considered), but rather to unambiguously identify the scattering processes responsible for the correct intertwining between different physical sectors, i.e. the processes allowing the large local magnetic response due to electronic localization to be accompained (as it should !) by a corresponding suppression of on-site charge fluctuations. We have also emphasized better in the revised version, how this result has allowed us, eventually, to clarify, why such interplay happens in the way we observe it, relating its manifestations to the intrinsic properties of the local moment formation (such as, e.g., to its characteristic long lifetime). In the revised paper, we also provide a concise justification for the choice of parameters (essentially, as U is kept fixed, for the three selected temperatures), we made to study the physics of the different regimes. In particular, for the HA, we've chosen $T = 2\,\frac{U}{2} (\beta = 0.5)$ for illustrating the behavior of the perturbative regime (of course here, also higher temperatures, but this would have further reduced the "resolution" of our Matsubara susceptibility matrices), and $T = 0.1 << \frac{U}{2} (\beta = 10)$ for describing the local moment regime. For the AIM (for which we used the same interaction value U as in the HA), the choice of the first two "higher" temperatures, representative of the perturbative and of the local moment regime is the same as above (as are both much larger than T_K), while for the screened (Kondo) regime we selected $T = \frac{1}{60} \sim T_K \simeq \frac{1}{65}$ (compare Fig. 1).

Of course, going along the line of thoughts of the Referee, one might indeed try to exploit the sharper sign-structures characterizing in the generalized charge susceptibility to define new /complementary criteria for delimiting the different regimes of the crossover, similarly to what was done in Phys. Rev. Lett. **126**, 056403 (2021) for the Kondo Temperature. For instance, for the high-T border $T^*(U)$ of the local moment region, one could use the sign-flip from positive to negative of the lowest eigenvalue of the generalized susceptibility, which indeed for large coupling display a scaling with $T^* = \frac{\sqrt{3}}{2}U$. The introduction of such sharp criteria in the charge sector might be even more useful out-of-half filling, where the temperature features in the spin-sector might become even more elusive. At the same time, the introduction of any of such crossover criterion represents (intrinsically) an arbitrary choice, hence, not being the main goal of our study, we prefer not to address explicitly this issue in this study.

In addition, I have two small questions: 1) Could the authors comment on why the charge susceptibility was chosen to study the effect of the formation of the local magnetic moment? If this effect "originates from the electronic scattering on the spin susceptibility," can it be observed directly by examining the spin susceptibility?

This represents an important point, indeed.

On the one hand, as discussed also above, the local moment formation and the associated freezing of local charge fluctuations are the two sides of the same coin (*"simul stabunt, simul cadent"*, i.e. one cannot have one effect, without the other). Even, the strong intertwining between the two scattering channels (mediated by the enhanced value of the triangular vertex in the local moment regime) is eventually responsible for the breakdown of the perturbation expansion and all its related manifestations. Hence, it would be reasonable to search for the presence of characteristic features of the local moment formation in the generalized magnetic susceptibility, too.

On the other hand, the two channels are strongly intertwined, though, they are certainly not equivalent, since the on-site spin response is enhanced and the charge response is frozen. This difference is largely reflected in the corresponding generalized susceptibilities. In particular, one observes that the generalized spin susceptibility displays an enhancement at all frequencies in the local moment regime, whereas the low-temperature Curie behavior of the susceptibility would be associated to a rather featureless positive structure extended on all fermionic Matsubara frequencies $|\nu|, |\nu'| \leq U$. This overall strong, but rather diffuse enhancement makes the fingerprints of the local moment formations, in same sense, not so easy to be directly "read" from the overall structure of the generalized magnetic susceptibility. On the contrary, the suppression effects on the generalized charge susceptibility (associated to the different signs of its ↑↑ and ↑↓ counterparts) is reflected in sharp frequency structures of *different signs* in the charge sector, which are very easy to identify, even at a first glance, and to be directly compared to those observed in other regimes. Beyond this practical reason (a much natural identification procedure), it is also worth to stress here a more general point: The freezing of the on-site charge fluctuations is a crucial aspect in strongly correlated electronic models: It plays an essential role in controlling the electronic mobility properties of the systems. In this respect, we note that precisely these nonperturbative suppressive effects in the charge sector, associated to the formation of the local moments, are responsible for the occurrence of Mott-Hubbard metal-insulator transition in DMFT. Indeed, as mentioned above as well as in Phys. Rev. Lett. **126**, 056403 (2021), within (even quite advanced) self-consistent perturbation approach, such as truncated fRG and the parquet approximation, even in the presence of local moment features in the spin sector, the freezing of the on-site charge fluctuations does not take place, due to a too little intertwining between the two channels. These considerations further support the choice of focusing on the generalized charge susceptibility.

2) Could the authors specify the parameters used to obtain the results shown in Fig. 1?

We now remarked the parameters in the figure caption.

Please correct two typos: 1) Page 3 - "(s. below)" 2) Page 5 - "In order to so

We thank the referee for the remarks and have corrected the typos.

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.