REPLY TO REFEREE 1

We thank the Referee for her/his second review of our manuscript as well as for her/his overall positive assessment on our work, consistent to the positive evaluation of Referee 2.
The Referee has asked us to consider specific points to be addressed prior to publications. We have thoroughly considered all of them (see the detailed reply below) and included the corresponding changes into the manuscript text and the appendix.

I would like to thank to the authors for their efforts in improving the manuscript. Specifically, I appreciate that the authors have incorporated estimates of the Kondo temperature for the Anderson impurity and Hubbard models.

We thank the Referee for her/his overall appreciation of our revision work.

However, I have noticed that my initial and, indeed, primary question has not been adequately addressed. It is possible that the authors may not have fully understood my concern, so let me try to rephrase it:

Already in the abstract ("The suppression effect on the diagonal elements directly originates from the electronic scattering on local magnetic moments, reflecting their increasingly longer lifetime as well as their enhanced effective coupling with the electrons") and in the overview of the results (page 8: "the intermediate temperature regime, where a local moment is formed in all the models considered"), the authors assert that the change in the frequency structure of the generalized charge susceptibility between in the high- and intermediate-temperature regimes is linked to the development of the local magnetic moment. However, this assertion lacks proper justification.

We respectfully disagree with the Referee on this point. As illustrated in our extensive first reply to both Referees, and further discussed below, the link between the local moment physics and the *quite specific* way in which on-site charge fluctuations are suppressed emerges very clearly from our diagrammatic and numerical analysis. It is possible, however, that part of the misunderstanding on the arguments presented in our previous reply stems from the omitted cancellation of a misleading sentence in the revised manuscript, as we discuss in more detail below.

In the beginning of Section 3.2, titled "Intermediate-temperature/local moment regime," the authors state: "As the next step, we focus on intermediate temperatures, for which, in the three cases considered, local magnetic moments are formed as a result of the on-site repulsion U. Their formation is signaled by a relatively flat (Curie) behavior of the quantity $T\chi^{\mathrm{sp}}_{\omega=0}(T)$

in the temperature range under consideration [39, 48] (not shown, see e.g., Fig. 4 in the supplemental of [23])." First, Figure 4 in the supplemental material of Ref. [23] does not depict the spin susceptibility. Rather, it presents the "partial frequency summation" of the generalized charge susceptibility. This quantity does not provide justification for the formation of the local magnetic moment.

We do agree with Referee about her/his critique to the quoted sentence. In fact, as we explicitly wrote in the final part of our first reply, it was our declared intention[1] to remove this sentence by the first revision of the manuscript. Due to a mere oversight, unfortunately, the sentence was eventually not removed. Thanks to the Referee's comment, in the second revision of our manuscript, we could eventually fix this issue. Though no longer relevant, as the whole sentence has been now dropped, we also acknowledge the incorrect referencing to Fig. 4 (instead of Fig. 1) in the supplemental of [23] (the different figure numbering was referring to the arXiv version of that publication).

At the same time, we note that, already in the first revision of the manuscript, we did include, as suggested by both Referees, the full temperature dependence of $T\chi_s(T)$ and $\chi_c(T)$ both for an unscreened (HA) as well as for a screened (AIM) case in final Appendix of our manuscript. The corresponding figures were also supplemented by a brief discussion of the relation between the parameter data sets chosen for the different models and the corresponding regimes.

Second, in my view, if the authors assert that the change in the frequency structure of the generalized charge susceptibility is related to the formation of the local magnetic moment, they should not only perform calculations in the two regimes where the local moment is either present or absent, but also demonstrate that this change occurs as a consequence of the moment's formation.

First, let us note that, the wording "formation of a local moment", though being quite intuitive, lacks a rigorous definition. As we also specify in the revised manuscript, the goal of our paper is not to discuss/compare criteria for delimiting borders between different physical regimes in the models con-

[1] Quoting from our previous reply the part referring to this specific sentence in the text: "*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*".

sidered. In this respect, let us recall that, as mentioned by the same Referee (as well as in her/his suggested work PRB 105, 155151 (2022), now incorporated in our bibliography), no thermodynamic transition is occurring in any of the systems/parameter regimes we considered. On the contrary, the underlying physics is characterized by smooth crossovers between the different regimes. As such, all possible definitions of the corresponding "borders", more or less recently proposed in the literature, while providing insightful guidelines across the parameter space, cannot be regarded as univocally defined criteria for their intrinsic arbitrariness. For instance, we note that, depending on the criterion adopted, the pretty well established evaluation of Kondo Temperature (T_K) does provide estimates which might differ even by a factor of 5. Hence, consistent with the crossover nature of the physical systems we are considering, the evolution of all quantities (including our physical and generalized susceptibilities) will also be (as it should be) smooth.

In this situation, it is quite clear that by presenting additional calculations at the precise parameters, where one or another criterion [2] would set the "border" between two physical regimes, cannot provide insightful information in the specific context of our study.

On the contrary, the main goal of our paper is to identify which scattering processes drive the very specific way (s. PRL 126, 056403 (2021)) in which the suppression of the on-site charge response, necessarily associated to the the physics of the formation of a local moment in strongly correlated systems, takes places in all the models considered, and, thus, triggers the breakdown of the self-consistent many-electron expansion. To this aim, we have exploited the Single Boson Exchange (SBE) decomposition, which, differently from parquet-based decompositions, remains applicable in all (perturbative as well as not perturbative) parameter regimes. Our results demonstrate that the SBE-based inspection of the different processes indeed holds the key for a precise understanding of the scattering mechanisms linking the spin to the charge sector, eventually allowing us to clarify the origin of the empirical observations made in PRL 126, 056403 (2021).

In a nutshell, in the parameter regimes where a sufficiently well-defined

[2] In fact, as we remarked in our previous reply to the second Referee, one could even use the properties of the generalized charge susceptibilities to define additional crossover criteria for these models, similar as the criterion introduced for T_K in PRL 126, 056403 (2021) or to the "fingerprint" criterion tested in PRB 105, 155151 (2022). For instance, one could choose the parameter values where the lowest-frequency diagonal entries of the generalized charge susceptibility drop under a certain value, or become negative, etc. Evidently, this "tautological" choice would not add relevant new information to our analysis. It is worth noticing, however, that such kind of criteria based on the low-frequency property of $\tilde{\chi}^{\mathrm{ch}}_{\nu=\nu'}$ would qualitatively (but of course not quantitatively!) match the (different) crossover borders defined in the recent literature. This is now addressed in a footnote at the end of Sec. 2 of the resubmitted manuscript.

local moment is present due to electron localization, several among all SBE contributions to the generalized charge susceptibility listed in Eq. (1) of our manuscript (precisely: the charge, the pairing-singlet and the double-counting SBE-terms) become negligible. We note, that while this trend can be clearly seen in our data (cf. Fig. 5 and Fig. 6 in the manuscript), its occurrence is unavoidably tied to the intrinsic physics of the local moment formation and, specifically, to the corresponding suppression of on-site charge and pairing fluctuations.

The remaining SBE contributions to the generalized charge susceptibility $\tilde{\chi}^{\text{ch}}_{\nu\nu',\omega=0}$, which need to be considered here, are then:

$$\begin{aligned}
\tilde{\chi}^{\text{ch}}_{\nu\nu',\omega=0} = &-\beta[G_\nu]^2\delta_{\nu\nu'} && \text{bubble}\\
&-[G_\nu]^2\phi^{\text{U-irr}}_{\nu\nu',\omega=0}[G_{\nu'}]^2 && \text{U-irr}\\
&+\frac{3}{2}U^2[G_\nu]^2\lambda^{\text{sp}}_{\nu,\nu'-\nu}\,\chi^{\text{sp}}(\nu'-\nu)\,\lambda^{\text{sp}}_{\nu,\nu'-\nu}[G_{\nu'}]^2 && \text{spin.}
\end{aligned} \tag{1}$$

In particular we observed that, while the SBE-irreducible term (U-irr), originated by multiboson exchange processes of all kinds, features a diffuse and overall positive contribution to the generalized charge susceptibility, the only negative (i.e., damping) terms is represented by the scattering processes involving the exchange of a (on-site) spin fluctuation. In particular, as explicitly discussed in our manuscript as well as in our first reply, the more the magnetic moment gets localized, the more its damping effects on the charge sector get concentrated along the diagonal for $\nu = \nu'$. Hence, the link with the physics of the magnetic moment is clear: The less an on-site magnetic moment is screened, the longer will be its lifetime, so that for a perfect[3] local moment, one has:

$$\chi^{\text{sp}}(\tau) \;= \text{const.} \implies \chi^{\text{sp}}(\nu-\nu') \propto \beta\,\delta_{\nu-\nu'},$$

which corresponds to the Curie-law. The suppressing impact of this increasingly **large** ($\sim 1/T$) and **frequency-selective** ($\sim \delta_{\nu-\nu'}$) spin contribution in Eq. (1) gets further enhanced at low-/intermediate frequencies by the the spin-fermion SBE vertex. In the local moment regime this vertex becomes substantially larger (see Fig. 7 in the manuscript) than its perturbative/high-frequency value. This provides a clear explanation of the scattering mechanism driving the suppression of the on-site fluctuations and the smooth emergence of the local moment physics. Remarkably, the precise identification of the link between the suppression of the diagonal entries of $\tilde{\chi}^{\text{ch}}_{\nu\nu'}$ and the on-site spin fluctuations in the local moment regime has finally allowed us to rigorously explain the origin of quantitative features characterizing the breakdown of the perturbation theory in the HA, see Eq. (12) in the

[3] E.g., where the local magnetic moment becomes a constant of motion of the problem considered.

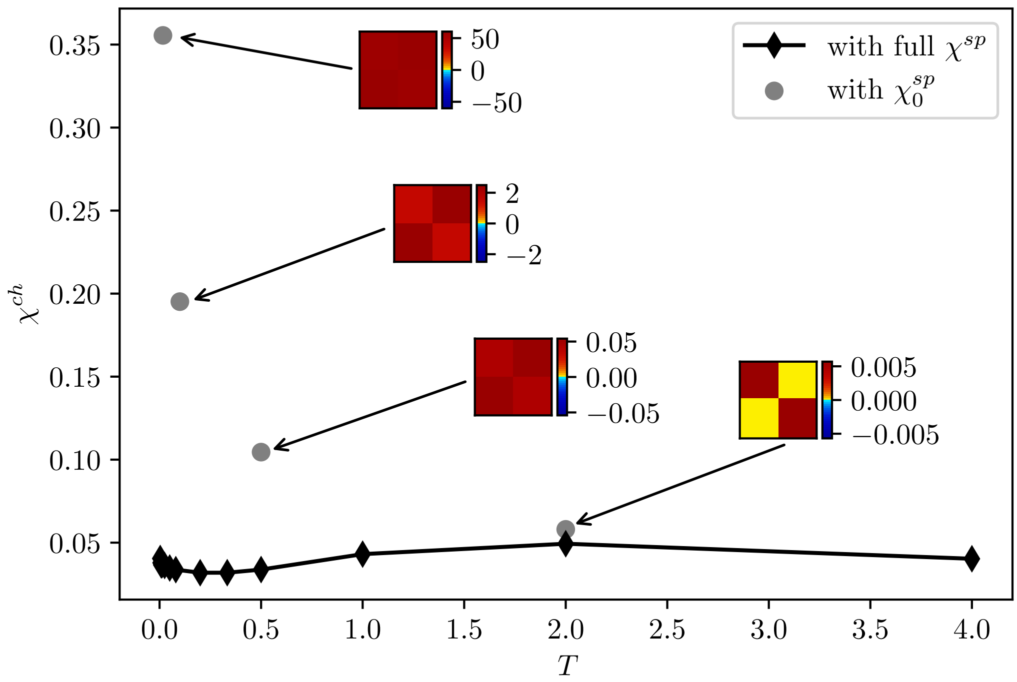

Figure 1: Additional data to the results reported in the upper right panel of Fig. 14 in the manuscript. The black line with diamonds shows the numerically exact on-site charge response (χ^{ch}) for the AIM considered, as a function of T. In comparison, the gray dots show χ^{ch} calculated from Eq. (11) of the manuscript, where in the corresponding spin contribution $\chi^{\text{sp}}(\omega)$ is approximated with $\chi_0^{\text{sp}}(\omega)$, i.e. vertex corrections are neglected. Evidently, the suppression of on-site charge fluctuations in the local moment regime is no longer visible with this approximation. Further, one of the discussed hallmarks of the local moment regime (i.e. the negative diagonal values of $\tilde{\chi}^{\text{ch}}_{\nu\nu'}$ at small frequencies) is also missing, as shown by the four small insets displaying the lowest Matsubara frequencies of $\chi^{\text{ch}}_{\nu\nu'}$ for different T.

manuscript, which have been previously reported in the literature, s. PRL 110, 246405 (2013); PRL 114, 156402 (2015); PRB 94, 235108 (2016); PRB 98, 235107 (2018).

Otherwise, how can one be certain that this change is not simply linked to the effect of temperature, or to a crossover between incoherent and coherent electronic regimes, or some other factor?

The direct link between the physical properties of the local moment and the nonperturbative frequency structure of $\tilde{\chi}^{\text{ch}}_{\nu\nu'}$ has been very clearly demonstrated by our combined SBE analytical/numerical analysis (cf. our discussion above).

To provide further evidence e.g. that the observed behavior of $\tilde{\chi}^{\text{ch}}_{\nu\nu'}$ is not driven by temperature dependent single particle coherent/incoherent effects, we have tested our physical interpretation by performing an **additional analysis**, which we report below.

In particular, we have computed how the temperature dependence of the on-site charge fluctuations for the AIM considered in our study would change, if one turned off the specific SBE scattering term associated to the formation of the local moment, while retaining, at the same time, the other $T-$dependent effects associated to the coherence/incoherence of the electronic systems. In practice, this is realized by neglecting all vertex corrections[4] to the physical spin susceptibility $\chi^{\mathrm{sp}}(\nu-\nu')$ appearing in our SBE expression [Eq. (1) above and Eq. (11) in our manuscript] for $\tilde{\chi}^{\mathrm{ch}}_{\nu\nu'}$, i.e. by replacing $\chi^{\mathrm{sp}}(\nu-\nu')$ with its corresponding (interacting) bubble term[5] $\chi_0^{\mathrm{sp}}(\omega) = -\frac{2}{\beta}\sum_\nu G_{\nu+\omega}G_\nu$.

The results of our additional analysis are reported in Fig. 1 included here. The data shown in the figure convincingly illustrate the validity of our conclusions: Neglecting the mere vertex corrections included in $\chi^{\mathrm{sp}}(\nu-\nu')$ in our SBE decomposition completely cancels any localization effect in the on-site charge response of the system. In fact, this response now displays a monotonous, significant increase when reducing the temperature in the whole T-range considered, in spite of the changes occurring in the corresponding on-site Green's function (G_ν).

Consistently, by looking at the low-frequency behavior of the generalized charge susceptibility, one can immediately see that also the suppression of the diagonal entries of $\tilde{\chi}^{\mathrm{ch}}_{\nu\nu'}$ down to negative values is no longer taking place, thus obliterating one of the main fingerprints of the local moment formation observed in [PRL 126, 056403 (2021)] and discussed in our manuscript.

If the authors determine the formation of the local magnetic moment based on "a relatively flat (Curie) behavior of the quantity $T\chi^{\mathrm{sp}}_{\omega=0}(T)$," I strongly urge them to explicitly present the results for the spin susceptibility at various temperatures for all three models considered and to indicate the specific temperature (or a narrow temperature range due to crossover effects) that they associate with the moment's formation.

As the sentence of our manuscript the Referee is quoting above has been omitted in the newly revised version of the manuscript, we consider this part of her/his observation as resolved.

If the authors further demonstrate that the change in the frequency behavior of the charge susceptibility indeed occurs upon this transition, it

[4]We note here, that the physics of the local moment formation, as well as the associated low-T enhancement of the corresponding on-site response, is fully driven by vertex corrections. In the absence of the latter, the charge and the spin response would become identical, e.g., displaying both a progressively suppressed behavior at lower T even in models with the strongest local moment effects such as the HA.

[5]Keeping the **interacting** bubble terms allows us to keep the coherent/incoherent $T-$dependent effects of the electronic system in our calculation.

would serve as a robust justification. Alternatively, instead of examining the susceptibilities, the authors could apply the criteria introduced in two recent publications [arXiv:2112.02881 (2021), PRB 105, 155151 (2022) to the generalized charge susceptibility]. These relevant works have been referenced in my questions in the previous review round, yet, for some reason, the authors chose not to address them in their manuscript.

On the basis of the considerations made before in our reply, we think that it would be rather improper to regard any of the different criteria existing in the literature as a marker of a definite "*transition*", because the local moment formation, as well as its relevant associated effects on the charge sector are smooth crossovers. While the scope of our work is, as mentioned above, a different one, for the sake of completeness of information, we have now included in the revised manuscript a short focused discussion (cf. footnote 3 in Sec. 2 and the extension of Appendix D) of our choice of parameter for the different regimes w.r.t. the crossover borders proposed in PRB 105, 155151 (2022), now Ref. [48], as well as in PRB 105, L081111 (2022). We think that this addition might provide useful information to the readers, and, at the same time, avoid possible misunderstandings.

In summary, in my opinion, the connection to the local moment in the current version of the manuscript appears more like wishful thinking than a substantiated justification. Without this justification, the authors could simply refer to the intermediate temperature regime as a regime characterized by the freezing of on-site charge fluctuations without mentioning the local magnetic moment.

As explained above, our analytical and numerical analysis fully succeeds in directly linking the emergence of the local moment formation (with its intrinsically associated increasing magnitude and lifetime) to the specific way in which the diagonal entries of the generalized charge susceptibility get gradually suppressed, across the corresponding crossover, triggering the breakdown of the self-consistent perturbation expansion. Hence, we hope that with the additional clarifications provided in our reply and revised manuscript, our work can be considered ready for publication.

As a minor note, there is a typo on page 8, just above Section 3.1, that requires correction: "Fig. ??, s".

We thank the Referee for noticing it: We have fixed the typo in the revised manuscript.