I. REPLY TO REPORT 1

The referee writes:

Strengths

1. *Relevant and timely topic*

2. *Solid numerical framework*

3. *Well-detailed methodologies*

Weaknesses

1. *Explanation of the limits of the numerical methods could be improved. (See report)*

2. *Direct potential experimental consequence and/or challenges could be explained in some more detail.*

Report

The manuscript titled: "Crossover from attractive to repulsive induced interactions and bound states of two distinguishable Bose polarons," presents a study of a population-imbalanced three-component mixture. The authors focus on a Bose gas coupled to two different species of minority atoms, which are considered single impurities. The different impurity-boson interactions allow for tuning the sign of the mediated impurity-impurity interactions, resulting in a wide range of two-body impurity states. These states are characterized by their energy, as well as two-body and three-body correlations and spatial distribution. The authors employed a recently developed variational multilayer multiconfiguration time-dependent Hartree method for atomic mixtures (ML-MCTDHX) approach in their studies.

The manuscript presents original and interesting results. The numerical approach, based on a method recently developed by the authors for 1D systems, is solid. Finally, the manuscript is clearly written, with well-explained sections and some Appendices which help the understanding of technical points of the manuscript.

In my opinion, the manuscript fulfills at least one of the expected acceptance criteria, in particular: a) detailing a groundbreaking theoretical-computational discovery and b) opening a new pathway in an existing or new research direction, with clear potential for multipronged follow-up work.

After some minor requests, I recommend its publication in Sci. Post. Phys.

Our answer is:

We thank the referee for their appreciation of our work and importantly the comments and further suggestions that helped us to improve our presentation. Below, let us elaborate on all issues raised. A list of changes is also attached at the end of the reply letter.

The referee writes:

Requested changes *1. Please provide details regarding the restrictions of the approach in terms of the strength of impurity-boson and boson-boson interactions for their studies.*

Our answer is:

We thank the referee for their comment. To address this point regarding the limitations of the method we have added a brief discussion in the revised manuscript and also provide two review articles where the constraints of the method are discussed in great detail (see also the list of changes).

The *ab-initio* method employed in this work is in principle capable of calculating the ground state of the three-component system for a wide range of interaction strengths. However, by increasing the impurity-medium interaction strength or the intracomponent coupling of the bath atoms the entanglement and the intracomponent correlations increase respectively. As a consequence, a larger number of orbitals is required in order to ensure numerical convergence. If simultaneously the atom number is large then increasing the number of orbitals becomes computationally costly and very time-consuming since the amount of the involved basis states/particle configurations is substantial. For this reason, in this study, we consider a mesoscopic number of weakly interacting bath particles which suppresses intracomponent correlations and thus allows to use a relatively small number of corresponding orbitals for this component. On the other hand, we vary the impurity-medium interactions to an arguably much larger range since the impurities are two and therefore it is possible to use a larger number of orbitals and ensure numerical convergence, see also Fig. **??** and our response to the third question of the third referee.

The referee writes:

2. Concerning the repulsive impurity-boson interaction, is there an underlying bound state? Can the physics of the attractive branch be captured for repulsive interactions? Please provide your comments on this.

Our answer is:

We thank the referee for this intriguing question. In the repulsive impurity-medium interaction regime while the bipolaron energy is negative it is not possible to obtain conclusive evidences regarding bound state formation, e.g. due to phase-separation.

For simplicity, in order to emulate a system consisting of two bosonic impurities (B and C species) coupled with the same interaction strength to the bath (A species), we set $g_{AB} = g_{AC}$. The corresponding bipolaron energy (see Eq. (13) in the manuscript) within the full many-body treatment (see also the diagonal of Figure 5(a) in the manuscript) and in the species mean-field case (where all interspecies correlations are suppressed) is shown here in Figure 1. It can be readily seen that for $g_{AB} = g_{AC}$ the bipolaron energy is negative for both attractive and repulsive impurity-medium couplings. Specifically, it decreases faster for attractive interactions as compared to repulsive ones. This behavior is attributed to the phase-separation between the impurities and the bath for strong repulsions. Notice also that in the presence of interspecies

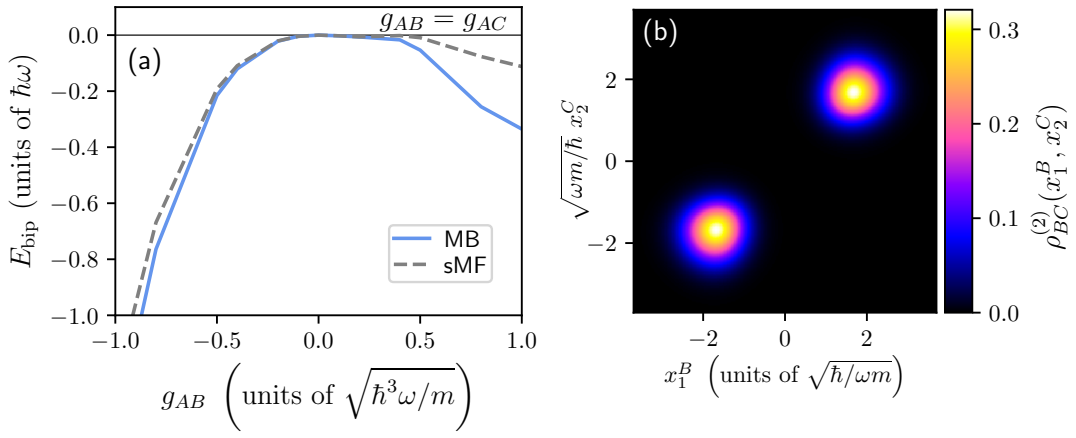

FIG. 1. (a) Bipolaron energy, $E_{\rm bip}$ (as defined in Eq. (13) of the manuscript) for varying $g_{AB} = g_{AC}$. The results obtained withing the full many-body (MB) approach (solid line) and in the species mean-field (sMF) method (dashed line) are depicted. In both cases, $E_{\rm bip}$ takes negative values for either attractive or repulsive couplings and its magnitude increases when intercomponent correlations are included. The black line marks $E_{\rm bip} = 0$. (b) Two-body density $\rho^{(2)}_{BC}(x_1^B, x_2^C)$ of the impurities for $g_{AB} = g_{AC} = 1.0$. Two-humps occur in the diagonal but without an elliptical shape which signalling no bound state formation.

correlations the bipolaron energy is further reduced (cf. solid and dashed lines in Figure 1). The above observations are in accordance with the ones reported in Ref. [1], where a similar model was studied consisting of a bath coupled to two identical bosonic impurities.

However, we do not associate the aforementioned impurity configuration at strong repulsions with a bipolaron for the following two reasons. First, for strong impurity-medium couplings where the impurities phase-separate with the bath the concept of a dressed state becomes ill-defined and thus the notion of a “bipolaron" becomes inappropriate. Second, we require an additional feature to be fulfilled, e.g., the two-body impurities density $\rho^{(2)}_{BC}(x_1^B, x_2^C)$ should exhibit an elliptical shape (i.e. an elongated diagonal and a relatively suppressed anti-diagonal) indicating the bunching of the impurities at relatively small distances as it is discussed and analyzed in the manuscript for attractive impurity-medium coupling strengths. For comparison, here, we present in Figure 1(b) the corresponding $\rho^{(2)}_{BC}(x_1^B, x_2^C)$ in the case of $g_{AB} = g_{AC} = 1.0$. The two-body density exhibits two humps along the diagonal since in this interaction regime, due to the harmonic trap, the impurities form a shell around the density of the bath and reside either both at the left or both at the right edge of the bath. Importantly, both of the two-body density humps are not elongated along the diagonal (i.e. the characteristic elliptical shape is absent) but rather show a Gaussian-like distribution centered around the edges of the cloud of the medium. This holds equally for larger repulsive impurity-medium couplings.

In view of the above analysis it is not viable to characterize the two impurity state as a bipolaron for repulsive impurity-medium interactions. This is also in a agreement with the conclusions of Ref. [1].

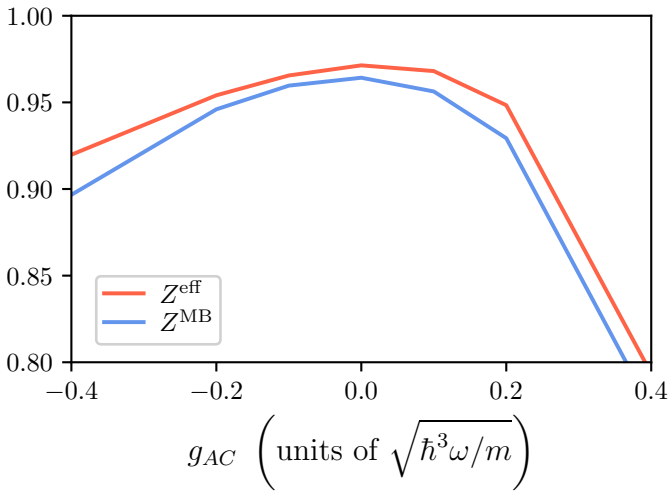

FIG. 2. Impurities residues with respect to g_{AC} and $g_{AB} = 0.2$. The residues are obtained either through the many-body wave function and the effective two-body model (see legend). The predictions of both approaches are in qualitative agreement with the residue in the many-body case being reduced due to correlations.

The referee writes:

3. In Section 6.2, if I understood correctly, this approach directly provides the spatial distribution of the impurities, and the authors manage to derive an effective mass. Can the coherence of the impurity (residue) be extracted in a similar manner?

Our answer is:

We thank the referee for this interesting question. The approach presented in Section 6.2 yields an effective two-body wave function describing two impurities experiencing induced interactions of strength g_{BC}^{eff} and an effective potential created by the bath density and the external trap. Therefore, it is indeed possible to extract the residue of the two-body impurities wave function within this effective model and compare it with the respective outcome using the full many-body wave function. Figure 2 presents the residue exploiting the effective two-body wave function and calculated as $Z^{\mathrm{eff}} = |\langle \Phi_{\mathrm{eff}}^{BC}(g_{AB}, g_{AC})|\Phi_{\mathrm{eff}}^{BC}(0,0)\rangle|^2$ together with the residue obtained from the many-body description, namely $Z^{\mathrm{MB}} = |\langle \Psi^{\mathrm{MB}}(g_{AB}, g_{AC})|\Psi^{\mathrm{MB}}(0,0)\rangle|^2$, for fixed $g_{AB} = 0.2$ and varying g_{AC}. Overall, the qualitative behavior of the two residues in this parameter range is the same, and therefore the effective two-body model provides an adequate estimate of the residue. Notice, however, that the residue Z^{MB} in the many-body case is independently of g_{AC} smaller than Z^{eff} predicted from the effective model. These deviations are expected, since in the calculation of Z^{MB} also the impurity-medium and the intracomponent medium correlations are taken into account, while being disregarded in Z^{eff}.

We have added a respective comment in the main text to discuss the possibility of extracting different impurities observables from the effective two-body model with a qualitative correct behavior (see also the list of changes). We finally remark that the parameter range for the

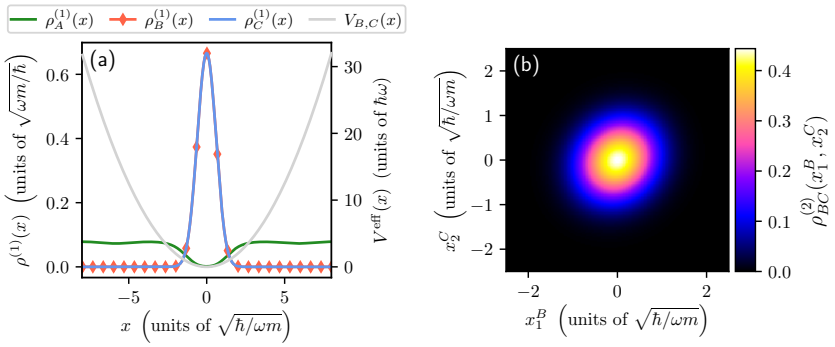

FIG. 3. (a) One-body densities of the impurities (B, C species) and the medium (A species), see legend, for repulsive impurities-medium interactions $g_{AB} = g_{AC} = 3.0$. The medium atoms experience a ring potential, while the impurities reside in a harmonic trap $V_{B,C}(x)$. We consider $N_A = 15$ interacting bath particles with $g_{AA} = 1.0$. A depletion of the density of the bath at the location of the impurities takes place. (b) The impurities two-body density in the same interaction regime as in panel (a). The strong impurity-medium repulsion leads to an elongation of the two-body density indicating a bunching behavior of the impurities at the center.

interaction strength g_{AC} shown in Figure 2 corresponds to the one presented in Figure 4(b) of the manuscript. As argued in the main text, the effective model is not valid for stronger interaction strengths where the impurities probe the edges of the bath, see also the fidelity (Figure 4(c) of the manuscript).

The referee writes:

4. In principle, in a homogeneous system as given in Equation 21, bipolarons are expected to form for both attractive and repulsive interactions (when g_{AB} and g_{AC} have the same sign). Is it because the trap allows for the phase separation of the impurities, preventing the formation of a bound state?

Our answer is:

We agree with the comment of the referee that a bipolaron state is possible to form also for repulsive impurity-medium interactions in a homogeneous system, see also our analysis below. It is indeed the presence of the trap in our setup that prevents bipolaron formation for repulsive interactions. As it was discussed in the answer to the question 2 of the referee, the bipolaron energy is negative for attractive as well as for repulsive impurity-medium couplings, as shown in Figure 1(a), indicating the formation of a bound state. However, in order to strictly infer the creation of a bipolaron and in particular distinguish it from such a clustering tendency of the impurities at the edges of the bath [cf. the two-body density depicted in Figure 1(b)] we need to simultaneously ensure the presence of a polaronic state. For the latter, among others, the minimal requirement is the existence of a finite spatial overlap between the impurities and the bath in order to achieve dressing via the impurity-medium coupling. However, at strong repulsive impurity-medium interactions the impurities phase-separate from their bath and therefore it is not possible to associate the negative energy state in Figure 1(b) with a

bipolaron.

The situation is different in a homogeneous system and a bipolaron can be formed also for repulsive $g_{AB} = g_{AC}$ as the referee suggests. To showcase this phenomenon we performed additional calculations for a bath residing in a ring potential and being coupled to two distinguishable harmonically trapped impurities. The resulting one-body densities of the bath and the impurities are illustrated in Figure 3(a), while the impurities two-body density is presented in Figure 3(b) for the case of $g_{AB} = g_{AC} = 3.0$ and $g_{AA} = 1.0$. As it can be seen, for this strong impurity-medium repulsion the bath density is depleted at the center but still shares a finite overlap with the impurities [Figure 3(a)] and simultaneously there is a bunching of the impurities as indicated by their respective elongated two-body density [Figure 3(a)]. Also, this interaction configuration is associated with a negative bipolaron energy $E_{\rm bip} \approx -0.96$. Therefore, these findings suggest indeed the formation of a bipolaron for repulsive impurity-medium couplings.

However, note that our aim here is to only argue about the possibility of bipolaron formation in a homogeneous setting and for repulsive impurity-medium interactions. Certainly, further analysis is required in order to obtain a systematic understanding of this process. For instance, we found that a bunching of the impurities only appears for sufficiently large interactions among the bath particles and impurity-medium repulsions. As such, studying the role of the ring size and the number of bath particles is an interesting venue for future works. We have added a respective comment in the conclusions of the manuscript (see also the list of changes).

The referee writes:

5. Could you comment on the feasibility and challenges of measuring in the experiment the correlation regimes and the integrated correlation function?

Our answer is:

To date, there are several available experimental techniques which allow the measurement of two-body correlation functions in both the position [2–5] and in momentum space [6]. For instance, the two-body correlation function could be spatially resolved by analyzing the single-shot images as described in Ref. [5]. Of course, for the current setup the fact that the atom number is relatively small implies that a large sample of single-shots would need to be used in order to reproduce the density. On the other hand, in order to measure the integrated correlation function it is not necessary to spatially resolve the two-body correlation function, thus rendering its detection more easy. Instead, it is sufficient to determine whether the two particles reside on the same or opposite sides with respect to the trap center. This procedure provides then information about the bunching or anti-bunching behavior. A corresponding comment has been added in the revised manuscript (see also the list of changes).

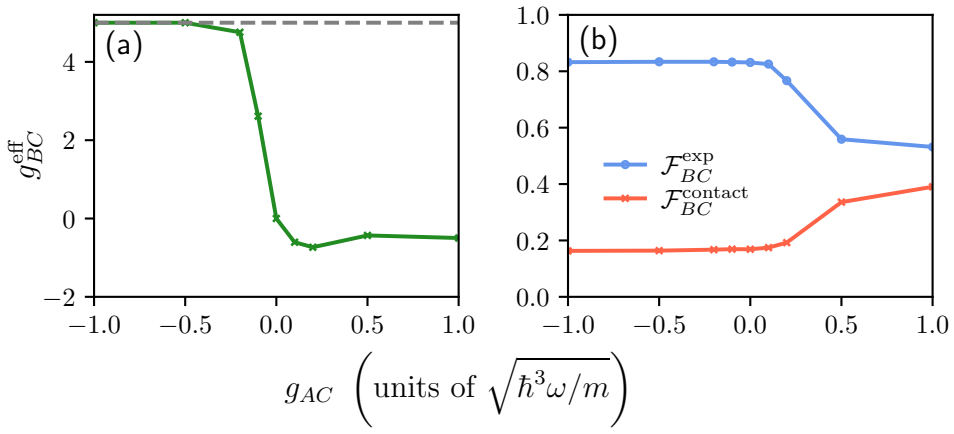

FIG. 4. (a) Induced effective interaction strength, g_{BC}^{eff}, which maximizes the overlap between the two-body correlation functions $\mathcal{G}_{BC}^{(2)}$ predicted within the many-body approach and $\mathcal{G}_{BC}^{(2),\mathrm{eff}}$ obtained from the effective two-body model [Eq. (12)] with a contact potential of strength g_{BC}^{eff}. We restrict our analysis to g_{BC}^{eff} lying in the interval $[-5, 5]$. Thereby, the gray dashed line corresponds to the upper boundary of g_{BC}^{eff}. (b) Fidelities $\mathcal{F}_{BC}^{\mathrm{contact}}$ and $\mathcal{F}_{BC}^{\mathrm{exp}}$ corresponding to the overlap of the impurities two-body wave function obtained from the full many-body approach and the effective model of Eq. (12) provided in the manuscript containing either an exponential or a contact-type effective interaction potential respectively. In all cases, we consider a fixed strongly repulsive $g_{AB} = 2.0$.

The referee writes:

6. Is it realistic to extend their approach to 2D or 3D systems?

Our answer is:

In general the MCTDHB and, therefore, also the ML-MCTDHX methods can also be applied to higher-dimensional problems, see also the review articles [7, 8] summarizing the ranges of applicability and detailing the ingredients of these methods. Relevant works in two-dimensions can be found e.g. in Refs. [9, 10], comparisons with experimental data were showcased in Refs. [5, 11], while the method has also been applied to study three-dimensional few boson systems in Ref. [12]. However, operating in higher-dimensions naturally further increases the complexity of the many-body simulations, e.g. in terms of the grid size and the orbital dimensionality, especially in view of the fact that within this approach the many-body wave function is stored.

Moreover, we believe that the one-dimensional setting provides a nice platform to explore the underlying quasi-particle physics and interplay of correlations as a first step in these three-component mixtures. Future studies could be indeed devoted to higher-dimensional setups. Finally, let us note that correlation effects are expected to be enhanced in one-dimensional setups as compared to higher dimensional ones [13] which makes our system desirable.

The referee writes:

7. The authors mention that a non-local mediated interaction may emerge for strong boson-

impurity interactions with a different sign. Do the authors expect Equation 12 to hold in this case as well?

Our answer is:

We thank the referee for their comment. Indeed, it is expected that the two-body effective model with short-range interactions given by Eq. (12) in the manuscript fails to describe the regime where non-local induced interactions occur. To clarify this issue a relevant comment has been included in the main text (see also the list of changes).

To be more precise, in this non-local regime the impurities have vanishing spatial overlap. As such, it is anticipated that the effective strength of the induced contact interaction becomes relatively large in order to capture the effect of induced interactions. This fact already indicates that the model of contact induced interactions is not an adequate effective description in the non-local induced interaction regime. To testify this prediction, we present in Figure 4(a) the extracted induced interaction strength, $g_{BC}^{\rm eff}$. We obtain this quantity by varying the two-body correlation function $\mathcal{G}^{(2),\rm eff}_{BC}$, obtained from the effective two-body model (Eq. (12) in the manuscript), as a function of the contact interaction strength $g_{BC}^{\rm eff}$ and determine the optimal value of $g_{BC}^{\rm eff}$ which maximizes the overlap with the two-body correlation function $\mathcal{G}^{(2)}_{BC}$ predicted in the many-body approach. Notice that, for simplicity, we restrict $g_{BC}^{\rm eff}$ in the representative interval $[-5,5]$ of strong effective $g_{BC}^{\rm eff}$.

It is evident that for $g_{AC} \lesssim -0.2$ (and $g_{AB} = 2.0$) which corresponds to the non-local induced interaction regime (see also the discussion on page 10, Section 5.2, in the main text), the effective strength $g_{BC}^{\rm eff}$ reaches the upper boundary of the aforementioned interval (see Figure 4(a) and the gray dashed line). Simultaneously, in this interaction regime, the fidelity, i.e., the overlap between the wave function in this effective description and the many-body result, is very small namely below 20% [cf. Figure 4(b)]. Moreover, since by further increasing the permitted value of $g_{BC}^{\rm eff}$ does not lead to a significant increase of the fidelity, we can conclude that a description according to Eq. (12) of the main text, i.e., an effective model consisting of a contact interaction potential, is not sufficient to describe the regime where the spatial impurities' overlap is suppressed. For comparison, we show in Figure 4(b) the fidelity corresponding to the effective model in Eq. (12) of the manuscript but containing an exponential interaction potential (Eq. (21) in the manuscript). Apparently, the fidelity is kept large and thus the exponential effective interaction potential is a much better choice within the non-local induced interaction regime.

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