I. REPLY TO REPORT 2

The referee writes:

Strengths

1. *Timely and sound topic of research.*
2. *Established state-of-the-art ab initio method.*

Weaknesses

1. *Similar system and methodology presented in previous publications of the author. The new elements present in the current work are not clear enough.*
2. *Experimental realization seems unviable and poorly described.*
3. *Range of validity of the method and results are not discussed.*

Report

The authors investigate the structural properties and the ground-state energy of a system that consists of two distinguishable impurities immersed in a harmonically trapped quasi-1D Bose gas at zero temperature. The highly imbalanced three-component system is characterized by coupling strengths $g_{\sigma,\sigma'}$, where $\sigma = 1, 2, 3$. By tuning different coupling strengths, particularly the impurity-bath ones, the two impurities can correlate and they experience either an attractive or repulsive induced interaction between them mediated by the weakly interacting bath. In particular, a bipolaron state is formed in the strongly interacting regime. The authors also predict the formation of a trimer state in the highly imbalanced three-component mixture.

To quantify these correlations, the authors present a calculation of observables, including the two-body coherence, defined in terms of the one and two-body reduced density matrix. The authors employ the multi-configurational time-dependent Hartree method to compute these observables. The methodology has been extensively used in other systems, such as degenerate quantum mixtures(see references [53-56]). Additionally, the bipolaron is further characterized by measuring the binding energy and the size of the two-body bound state. Finally, the trimer state is characterized using a three-body correlator inspired by the previously defined two-body coherence.

Our answer is:

We thank the referee for their effort to read our work and provide suggestions for improvement and further analysis. In the following, a detailed point-by-point reply to all questions of the referee is given and a list of changes performed in the revised manuscript is provided at the end of the reply letter.

The referee writes:

The authors emphasize the high controllability of a highly imbalanced mixture. They present an example using two isotopes of Rubidium. However, the systematic experimental implementation is poorly discussed thoroughly in the text, rendering the work primarily theoretical without

clear prospects for possible observation. For example, in the calculations, the impurity-impurity coupling strength is set to zero, which is reasonable since the focus is on observing induced interactions rather than direct ones. However, to achieve this, it is necessary to identify a suitable range of magnetic fields with a constant scattering length (zero for impurity-impurity interactions), two adjustable impurity-boson scattering lengths, and a constant and positive boson-boson scattering length. In the particular case of the current draft, the authors propose a ^{85}Rb condensate. The background scattering length is negative (arxiv.org/pdf/1003.4819.pdf) and this may be problematic. The choice of atomic species is quite limiting in experiments and should be discussed carefully.

Our answer is:

We agree with the referee that explicit parameter values e.g. in terms of scattering lengths of specific isotopes or hyperfine states regarding a corresponding future experimental implementation of the used three-component setup is currently challenging. Promising candidates for a corresponding realization are either three different hyperfine states of ^{87}Rb which can feature various Feshbach resonances e.g. the states $|1\rangle \equiv |1,-1\rangle$ and $|2\rangle \equiv |2,0\rangle$ have $g_{11} \approx 1.004$, $g_{22} = 0.9457$, and $g_{12} = 0.9813$, while the states $|2\rangle \equiv |2,0\rangle$ and $|3\rangle \equiv |1,0\rangle$ have $g_{22} \approx 0.9457$, $g_{33} = 1.0086$, and $g_{23} = 0.989$. Otherwise, a mixture of ^{87}Rb and ^{85}Rb may also be used. Recall that an attempt towards proposing an experimental implementation of three-component systems containing impurities was recently made for ^{41}K-^{87}Rb in Ref. [1], while three-component mixtures were realized also in Refs. [2, 3]. It should be also emphasized that since we operate in one-dimension the interactions can also be tuned via confinement induced resonances independently. This fact provides even more flexibility. As such, the physics described in this work should be realizable in forthcoming three-component experiments. We have commented on these issues in Section 2 of the revised manuscript and also hinted to possible experimental challenges (see also the list of changes).

Indeed, the research direction of three-component systems is still at an early stage, but it recently started to gain attention also for future experiments. As such, the corresponding experimental techniques and allowed parameter regions are not yet so matured. Therefore, a fully reliable experimental proposal is not currently a trivial task. Of course, this provides additional motivation for theory works to dictate phenomena and parametric windows that the experiments will try to address in the near future. Let us also comment that this was the situation in the past when the community started to investigate two-component mixtures or few-body settings. We very much hope that a similar route will be followed for three-component systems and also that the referee shares our excitement to investigate unexplored settings even if they are currently experimentally challenging but certainly not out-of-reach.

To conclude, the aim of our work is not to provide an explicit experimental implementation, but rather explore new and intriguing physics being apparently absent in two-component setups and worths to be explored in future three-component experiments. Certainly, we are aware that realizing all the discussed interaction configurations might be tedious experimentally due to the required precise tuning of magnetic fields. Furthermore, as we show in Appendix D the important features of the interspecies correlation behavior, i.e. crossover from positive to negative values, are preserved also for mass-imbalanced setups and, thus, our findings could also be observed in hetero-nuclear experimental settings.

The referee writes:

On the one hand, the work provides an interesting calculation concerning the correlation between impurities and the formation of few-body bound states like bipolarons and trimers, using a state-of-the-art method. On the other hand, I find it challenging to distinguish the current work from previous publications by the authors, specifically PRA 104, L031301 (2021) and PRA 105, 053314 (2022), where a very similar physical system and methodology are employed. Thus, this work represents a new but incremental calculation compared to the previous works, and I cannot identify any significant evidence of groundbreaking theoretical results, apart from the crossover that is not well discussed.

Our answer is:

We thank the referee for their remark but let us respectfully strongly disagree with the statement that our work is an incremental calculation of the previous works. Rather, it should be emphasized that, to the best of our knowledge, our results have not been reported elsewhere and are novel both on the physics but also on the technical side. Specifically, there is almost *zero overlap* between the above-mentioned works and the current one even on the conceptual level. Below, we provide only the main reasons rendering the distinction between the research works clear:

- In Ref. [4] a single impurity coupled to a two-component bath has been studied. Notice already here the *conceptual difference* of the present setup and the one of Ref. [4]. *In the latter case the notion of induced interaction is completely irrelevant.* Indeed, in Ref. [4] the emphasis was placed on the polaron formation with respect to the coupling strengths between the impurity and the two baths. In sharp contrast, here, we consider two distinguishable impurities coupled to the same bath and study the impact of induced impurity-impurity correlations upon varying the impurity-medium interaction strengths. Concluding, *the research questions but also the physical settings of the two works are arguably completely decoupled* apart from the fact that in both cases we use a three-component mixture.

- On the other hand, the setup used in Ref. [5] is already *different* from the one studied in this work *both on the level of the involved components but also in terms of the trap geometries*. Namely, Ref. [5] exploits a two-component mixture with two indistinguishable impurities in a double-well, while in the present work we employ a three-component mixture with two distinguishable impurities trapped in a harmonic oscillator. More importantly, in Ref. [5] we have investigated the collisional dynamics of two indistinguishable bosonic impurities coupled to a bath. Indeed, the impurities were initialized in a double-well whose central barrier was suddenly ramped-down inducing the dynamics. The ground state investigation in Ref. [5] was performed in order to understand the emergent dynamical response and was referring to a different trap geometry and a two-component system. As such, there is clearly *no overlap between the two works even on this level*. In fact, as we also explicate in the main text, for the physics question that we tackle it is crucial to consider a three-component system with two distinguishable impurities such that there is the possibility to achieve impurity-medium couplings of different sign. In other words, an induced repulsion cannot be observed in systems

where the impurities couple with the same strength to the bath, as it is for instance the case in Ref. [5].

We hope that the above argumentation makes clear the *fundamental distinction* of the previous works from the present one *both in terms of the physical system but also on the conceptual research direction.*

The referee writes:

The system may undergo a miscible-immiscible transition when considering a highly imbalanced triple mixture and for a certain range of parameters. Yet, the authors state that the mediated interaction of polarons causes an effective repulsion or attraction. The question is: How can the authors differentiate between these two scenarios?

Our answer is:

We thank the referee for their comment. It should be clarified that these two concepts, namely the “miscible-immiscible phase transition" and the “induced interaction" are different processes which are decoupled from each other. Indeed, two atomic ensembles are immiscible when the employed intercomponent repulsion is larger than the average intracomponent repulsive interactions. In this sense, the interactions enforce the two components to be separated from each other. In the reverse scenario, the two components remain miscible by means that they share a finite spatial overlap. We remark at this point that the impact of interparticle correlations on the miscible-immiscible phase transition in an impurity setting was studied in detail in Ref. [6] and thus it is out of the scope of the present work. On the other hand, the notion of “induced interactions" refers to the effective interaction mediated between the impurities due to the presence of impurity-medium correlations even and in particular for non-interacting impurities.

Focusing on the concept of induced interactions let us clarify that in order to clearly differentiate between an induced repulsion or attraction we have calculated a modified relative distance, see Eq. (11) in the manuscript. The main idea of this measure is to subtract from the impurities relative distance (extracted within the full many-body method) the involved mean-field effects. The latter can be identified using the species mean-field approach which allows to truncate correlations of the many-body wave function at different levels e.g. among the atoms of the bath or between the bath atoms and one impurity (see also the discussion in Section 6.1). Since only within the full many-body calculation the two impurities can become correlated, due to an induced correlation mediated by the bath (and the choice of $g_{BC} = 0$), the modified relative distance captures the presence of induced correlations between the impurities. In particular, we associate a positive (negative) value of the modified relative distance with a correlation-induced repulsion (attraction), see also Figure 4(a) of the manuscript. Complementary, we construct in Section 6.2 of the manuscript an effective two-body model which specifically includes a contact interaction potential modelling the induced interactions. Comparisons between the results obtained from the many-body approach and the effective two-body model with respect to different observables, e.g. the two-body correlation function, allow the identification of the underlying effective interaction strengths. The sign of the latter is in agreement with the observations made in terms of the modified relative

distance, i.e., we find a negative (positive) effective interaction strength when the modified relative distance is negative (positive).

The referee writes:

In this work, typical values for the impurity-boson coupling strength typically range between -3 and 3. Why are the authors constrained within this range? For instance, in PRL 127,103401 (2021), the strongly interacting regime for the double imbalanced mixture is attained with larger values of the impurity-boson coupling strength.

Our answer is:

We thank the referee for their remark. Let us emphasize that already the physics observed in this interaction range is rich and, to the best of our knowledge, completely unexplored. It is our aim to build-up a systematic understanding of the underlying impurities behavior, induced interactions and ground state phases. The considered interaction regimes are indeed sufficient to cover important phases, e.g., phase-separation for impurity-medium interactions larger than the intracomponent ones of the bath, the bunching behavior of the impurities for increasing attraction and the crossover from attractive to repulsive induced interactions. Also, in order to adequately comprehend all these aspects a variety of tools has been exploited/constructed ranging from effective one- and two-body models to full many-body simulations and corresponding reductions of the wave function to control the involved correlations.

However, turning our attention to other interaction regimes will automatically also extend the relevant phenomenology which we believe it deserves separate studies. For instance, a further decrease of the impurity-medium coupling will most probably further facilitate trimer formation [7] or even higher-order bound states such as tetramers. Similarly, increasing the impurity-medium repulsion will lead to the Tonks–Girardeau regime which is interesting on its own since the system will feature fermionization. Both of these interaction regimes are certainly intriguing but lie out of the scope of this work which is already extended as mentioned above. Furthermore, in order to consider the strongly correlated regime, as the referee suggests, it might be better to exploit other setups such as a ring geometry in order to avoid phase-separation among the impurities and the bath and instead facilitate bound state formation.

The referee writes:

Given the previous remarks, especially the close connection with the aforementioned works (PRA 104, L031301 (2021) and PRA 105, 053314 (2022)), which limits the originality of the research, and the absence of a proper experimental protocol, the current work falls short of meeting all four Scipost acceptance criteria. Nevertheless, the methodology is deemed reliable, and the results could be suitable for publication in a more specialized journal. For example,

physical review A or B.

Our answer is:

We hope that the referee through our detailed argumentation above is now convinced about the importance of our results and their novelty as well as the zero connection with the aforementioned works. Regarding the experimental advances in three-component mixtures there are definitely aspects that can be improved since these systems are not exhaustively studied. In the revised version, we comment on these issues and improved the relevant discussion. However, it should also be noted at this point that our work is one of the first theory ones elucidating the rich physics of impurities in three-component systems. For the above-mentioned reasons, we strongly believe that the present manuscript meets the Scipost criteria.

The referee writes:

Other less strong concerns may also be addressed prior to publication in any journal.
The introduction contains the sentence, "In particular, induced interactions are solely attractive as long as the impurities are indistinguishable and thus couple in the same way to their medium." Please provide a reference for such a claim.

Our answer is:

We thank the referee for their comment. Indeed, in previous works only an attractive induced interaction has been observed, see e.g. Refs. [8–12]. This fact is further supported by the analytical expression of the impurities effective interaction potential derived in Refs. [10, 11] which only allows the rise of attractive induced interactions between two (bosonic) impurities coupled with the same strength to the bath. We have accordingly modified the above statement and also provided the respective literature (see also the list of changes).

The referee writes:

If I understood well, in Fig.1, the authors plot the density of each component and the effective potential as well. The density is directly related to the potential via Eq. 6. In order to check the quality of the effective potential model; I encourage the authors to plug the effective potential into a simple Schrödinger equation and obtain its respective wave function. The wave function squared may be compared to the density obtained from the many-body calculation.

Our answer is:

The procedure mentioned by the referee, i.e., solving the one-body Hamiltonian consisting of a kinetic term and the effective potential corresponding to Eq. (6) of the manuscript has been already discussed in Appendix B. For instance, elaborating on the case of weakly interacting impurities we showcase an excellent agreement with the many-body simulations on the one-body density level. Deviations start to occur for increasing interaction where the effective potential picture is conceptually no longer valid. Let us finally mention that we further testify

the validity of the effective two-body model in terms of its wave function predictions.

The referee writes:

Linked to the experimental realization, could the author discuss losses?

Our answer is:

We thank the referee for raising this point. However, let us mention that the inclusion of loss mechanisms stemming, for instance, from two-body collisions or three-body reccombination processes are not an immediate priority for the present work and certainly not an easy task for several reasons that we outline below. First, we examine the ground state properties of the three-component mixture and not corresponding dynamical processes where loss channels might be relevant. Second, the loss coefficients e.g. the corresponding three-body recombination rate is estimated either through explicit scattering calculations (being of course beyond the scope of the present work) or determined experimentally. Third, we should emphasize that the inclusion of loss processes is not a trivial task in every *ab-initio* method, such as the ML-MCTDHX or Quantum Monte Carlo, since they rely on atom number conservation. It is in principle doable to incorporate loss channels but this requires a substantial modification of the fundamental ML-MCTDHX equations of motion, see for instance Ref. [13] for an attempt along these lines. This is arguably a highly demanding task, beyond the scope of the present work, which is interesting on its own right and would definitely require further benchmarks.

The referee writes:

What about the formation of high few-body states on top of bipolarons and trimmers, for instance, tetramers, pentamers, etc?

Our answer is:

A key facet of the ML-MCTDHX method is that it provides access to the total many-body wave function of the system. As such, in general, it is possible to calculate also higher-order correlation functions (see e.g. Ref. [14] for such a calculation with this method) even though they are high-dimensional observables and thus relatively time-consuming. Turning now to the physical system at hand, we have deduced that the magnitude of the participating three-body correlations is suppressed as compared to the two-body ones. For instance, this can be understood by comparing the magnitudes of the two-body correlation functions (see e.g. Figures 2(b1) and (b2) in the manuscript) with the corresponding three-body correlation function (see e.g. Figure 6(a) in the manuscript). It can be seen that the amplitude of the latter is only slightly larger from the amplitude of the former. This is a consequence of the fact that the three-body correlation function still contains two-body correlation effects, see also the projections referring to the two-body correlation functions on the x-y, x-z, and y-z planes in Figure 6(a) of the manuscript. Therefore, pure three-body correlation effects are suppressed compared to the two-body ones.

In a similar vein, it is naturally expected that higher-order correlation effects are further reduced and, therefore, are not of immediate interest at least for the scope of the present work. In this sense, and in view of our already extensive study we believe that such an exploration regarding higher-order correlation functions is better suited for future investigations where it would also make sense to identify conditions in the parameter space which would allow the tuning of the different correlation orders.

The referee writes:

In Fig. 2, the panels a1,b1 and c1 show $G^{(2)}_{AB}(x^A_1, x^B_2)$, however in the horizontal axes reads x^B_1. Vertical units are wrong. Same for of g_{AB} in Fig.3.

Our answer is:

We thank the referee for their remark. We have accordingly modified the horizontal axis of Figure 2 in the manuscript (see also the list of changes). However, the units at the vertical axes in Figures 2 and 3 in the manuscript are correct. Specifically, instead of noting, e.g., "x^A_1 in units of $\sqrt{\hbar/\omega m}$" in Figure 2 we just multiply the parameter x^A_1 with $\sqrt{\omega m/\hbar}$ and mention "$\sqrt{\omega m/\hbar}\, x^A_1$". This is done for better visibility and in order to avoid too many label notations as well as do not make the panels busy. The same holds for the vertical axis in Figure 7 of the manuscript.

The referee writes:

The coherence, as defined in the article, should contain dimensions.

Our answer is:

We thank the referee for their remark. In the revised version we clarify the dimensions of the coherence in the caption of Figures 2, 5 and 6 (see also the list of changes).

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