RESPONSE TO REFEREE REPORTS

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I wish to thank the referees for their reports and the observations made. Text changes have been made in the revised manuscript to address the referees' questions and suggestions. Such changes are explicitly indicated in detail and in context of our responses below. In what follows, original questions and comments from the Referees are in blue, and our responses in black.

Response to Referee 1

The present manuscript applies the formalism of the so-called "dynamical exchange-correlation (xc) field" of Ref.[37] to the single-impurity Anderson model. The authors proposes a rather simple ansatz (Eq.(30)) for this dynamical xc field to obtain the spectral function of the Anderson model in the Kondo regime where the parameters are fixed by using the known peak positions and widths of the spectral peaks. This ansatz seems to work surprisingly well given its simplicity.

While I find the paper interesting in general, I still have a number of points which I would like the author to address:

We wish to thank the referee for the summary of our work. The Referee raises several questions about notations and details in the manuscript, which we believe will help clarify our paper. In the following, the Referee's questions and observations are addressed one at a time.

1. The coefficient $a_{n+,m}$ is defined just after Eq.(14). Shouldn't this also be sigma-dependent?

The Referee's comment is generally true, i.e., the coefficients

$$a_{n_+,m} = \langle n_+|\hat{f}_\sigma|m\rangle \tag{R1}$$

depend on the spin index σ for a general case. For the particle-hole symmetric single-impurity Anderson model without external magnetic field, which is the problem we studied, the weight coefficients are spin-independent, $\langle n_+|\hat{f}_\uparrow|m\rangle = \langle n_+|\hat{f}_\downarrow|m\rangle$. However, since we define the Green's function $G_{ff,\sigma}$ with the spin component, for consistency we should also keep the spin indices explicit for $a_{n_+,m}$. The changes have been made in the manuscript.

2. On Eq. (20): first, I suppose it is only meant to be valid for $t > 0$, no? Second, I am a bit confused about its form: why is there no explicit dependence on the interaction U? Shouldn't it (loosely speaking) be something like $UG^{(2)}(t)/G(t)$ where $G^{(2)}$ is the two-particle Green function? Also, I don't understand the factor $a_{n+,m}\omega_{n^+,m}$ in the denominator. I would have expected this to be $< m|\hat{n}_{-\sigma}f_\sigma|n+ >< n+|f^\dagger_\sigma|m >$. Please clarify!

To the first question, yes, Eq. (20) is valid for positive time. As stated by Eq. (14), we focus on the Green's function (and thus the Vxc) with positive time. The negative time part of G can be obtained with the particle-hole symmetry:

$$G_{ff,\sigma}(t < 0) = -G_{ff,\sigma}(-t). \tag{R2}$$

Then $G(t)$ is used to calculate the spectral function $A(\omega)$. We have added the explanation about the negative time part in the manuscript.

To the second question, the equation of motion of $G_{ff,\sigma}(t)$ is

$$[i\partial_t - \epsilon_f - V^{\mathrm{H}}]G_{ff,\sigma}(t) - \sum_k v_k G_{kf,\sigma}(t) + iUG^{(2)}(t) = \delta(t) \tag{R3}$$

where $V^{\mathrm{H}} = Un_{f\bar{\sigma}}$ is the Hartree term, $G_{kf,\sigma}(t)$ is defined with $\hat{c}_{k\sigma}$ and $\hat{f}^\dagger_\sigma$, and $G^{(2)}(t)$ is, as the Referee pointed out, a two-particle Green's function:

$$G^{(2)}(t>0) = \frac{1}{Z}\sum_{m,n_+} e^{-\beta E_m} e^{-i(E_{n_+}-E_m)t}\langle m|\hat{n}_{f\bar{\sigma}}\hat{f}_\sigma|n_+\rangle\langle n_+|\hat{f}^\dagger_\sigma|m\rangle, \tag{R4}$$

where $Z = \sum_m e^{-\beta E_m}$ is the partition function and $\bar{\sigma}$ denotes a spin opposite to σ. In the dynamical xc field formalism, the $UG^{(2)}$ term can be seen as the product of the Coulomb potential of the dynamical xc hole and the Green's function. The $\sum_k v_k G_{kf,\sigma}(t)$ term represents the hybridization effect. In the manuscript, we incorporate both the Coulomb interaction and the hybridization effect into the dynamical xc field (Vxc) in order to have a simple form of the equation of motion, which is for positive time (the same as setting $t > 0$ for Eq. (15) in the original manuscript),

$$\big[i\partial_t - \epsilon_f - V^{\mathrm{H}} - V^{\mathrm{xc}}(t>0)\big]G_{ff,\sigma}(t>0) = 0. \tag{R5}$$

So according to our definition, $V^{\mathrm{xc}}(t>0)$ can be written with the explicit dependence on U, $G^{(2)}$ and $G_{kf,\sigma}$:

$$V^{\mathrm{xc}}_\sigma(t>0) = \frac{\sum_k v_k G_{kf,\sigma}(t>0)}{G_{ff,\sigma}(t>0)} + \frac{UG^{(2)}(t>0)}{iG_{ff,\sigma}(t>0)}. \tag{R6}$$

Alternatively, using the equation of motion of G, $V^{\mathrm{xc}}(t>0)$ can be expressed with ϵ_f, V^{H} and $G_{ff,\sigma}$. Because of the particle-hole symmetry, $\epsilon_f + V^{\mathrm{H}} = 0$, thus we have

$$V^{\mathrm{xc}}_\sigma(t>0) = \frac{i\partial_t G_{ff,\sigma}(t>0)}{G_{ff,\sigma}(t>0)}, \tag{R7}$$

which is the way we used in Eq. (20). By using the equation of motion, V^{xc} can be expressed with only $G_{ff,\sigma}$ and its time derivative. The $a_{n_+,m}\omega_{n_+,m}$ coefficients in Eq. (20) follow from applying the Lehmann representation of $G_{ff,\sigma}$ to Eq. (R7) of this response. We have added text before Eq. (20) in the revised manuscript to explain the usage of the Lehmann representation of the Green's function and that V^{xc} (Eq. (20)) is solved from the equation of motion. We hope this can improve the readability of our paper.

3.Please give more details on what is actually done in Sec. 3.2 and how, such that interested readers could repeat the calculations. The time-dependent variational principle is used to obtain which quantity, the one-particle Green function of the cluster?

We agree with the Referee that more details will be necessary for repeating the calculation. We have added in the manuscript the relevant information. Here, we give a brief explanation. The time-dependent variational principle algorithm is a part of the Matrix Product States method. We use it to time-evolve the state $\hat{f}^\dagger_\sigma|\Psi_0\rangle$, where $|\Psi_0\rangle$ is the zero-temperature ground state of the finite cluster and is obtained by the density matrix renormalization group algorithm. With $|\psi(t)\rangle = e^{-i\hat{H}t}\hat{f}^\dagger_\sigma|\Psi_0\rangle$, the one-particle Green's function can be calculated:

$$\begin{aligned} iG_{ff,\sigma}(t>0) &= \langle\Psi_0|e^{i\hat{H}t}\hat{f}_\sigma e^{-i\hat{H}t}\hat{f}^\dagger_\sigma|\Psi_0\rangle \\ &= e^{iE_0 t}\langle\psi(0)|\psi(t)\rangle. \end{aligned} \tag{R8}$$

Then the spectral function is calculated from $G_{ff,\sigma}(\omega)$.

4.In Fig.4: could the author plot the NRG results on top of the present results for better comparison? The same applies for Fig.5.

We thank the Referee for this suggestion. Now we add NRG results for both Fig. 4 and Fig. 5.

5.In Eq.(37): I assume that the parameter Ω_T is temperature dependent? How is this parameter determined in practice? Is it used as a fit parameter to reproduce known spectral functions? Please show its evolution as function of temperature!

We thank the Referee for this question, which has inspired us to find theoretical references for our finite-temperature results. The parameter Ω_T is temperature-dependent and is related to the half-width of the Kondo peak at finite temperature. In the original manuscript, we used Ω_T as a fitting parameter to reproduce the broadening of the Kondo peak (and the corresponding decrease in its height) due to thermal fluctuations. However, we recently noticed that some literature (Phys. Rev. Lett. 88, 077205 (2002), Phys. Rev. B 108, L161109 (2023)) provides expressions for the temperature dependence of the Kondo peak half-width (Γ_{K}), based on Fermi-liquid theory and its extension. Using one expression from Phys. Rev. B 108, L161109 (2023), which applies to temperatures much lower than the Kondo temperature T_{K}, we can now express Ω_T as a function of temperature:

$$|\Omega_T| = \frac{\alpha T^2}{T_{\mathrm{K}}}, \tag{R9}$$

where $\alpha \approx 3.44$ is a constant calculated from Fermi-liquid theory and is consistent with NRG numerical results. For higher temperatures, a more complicated relation of $\Gamma_{\mathrm{K}}(T)$ can be used to determine Ω_T. We refer to Eq. (10) of Phys. Rev. B 108, L161109 (2023).

With these expressions of $\Gamma_{\mathrm{K}}(T)$, we can now apply our formalism to a wider range of temperatures. We have added new results and comparison with NRG in the revised manuscript.

6.Finally, I noticed a typo in line 141: it should be "emphasize" instead of "emphasis"

We thank the Referee for pointing out the typo. The word has been corrected in the manuscript.

To summarize, before I can recommend this manuscript for publication in SciPost Physics I would like to see the issues raised above being addressed.

We again thank the Referee's questions and remarks, which help us improve our treatment about Ω_T and the readability and the clarity of the paper. We hope that our response and corrections in the manuscript have addressed the issues raised and that the paper can now be considered as suitable for publication in SciPost Physics.

Response to Referee 2

This paper applies a novel method, the so called "dynamical exchange-correlation field" (Ref. 37), to the Anderson impurity model in order to compute spectral functions in the Kondo regime.

I find the paper quite interesting. But I think the paper lacks clarity in a couple of places. The paper would also benefit from better explanations in some parts, since the dynamical exchange-correlation field is a novel approach. I have a couple of comments and questions that the author should address before I can agree to publication:

We thank the Referee for a thorough, insightful, and very helpful report. We are glad that the Referee gives a positive evaluation of our work. The Referee points out the unclear statements that can be improved, and also asks some detailed questions about our approach. Importantly, some questions from the Referee have helped us improve our original approach and greatly inspired us to gain a deeper understanding of our results. Accordingly, we add explanations for some of the concepts, provide better reasoning for our statements, and answer the Referee's questions one at a time.

(1) I was at first confused by Eq. (11): How could the dynamic exchange correlation hole for $r'' = r$ become time-independent and just equal to the negative density? But this follows from

Eq.(8) and the fact that the second order Green's function $G^{(2)}(r, r', r''; t)$ (using the notation of Ref. 37) vanishes for $r'' = r$, and thus also the correlation function $g(r, r', r; t) = 0$. I think the author should give this explanation after Eq. (11) to help the reader.
The exact constraint of the dynamical xc hole, $\rho^{\rm xc}(r, r', r'' = r; t) = -\rho(r)$ is an important property. As noticed by the Referee, this constraint follows from the fact that the two-particle Green's function vanishes at $r'' = r$. We thank the Referee for pointing out that a brief explanation can avoid confusion and help the reader. We have now added the explanation in the manuscript.

(2) Eqs. (13) and (14) are the Lehmann representations of the Green's function (which should be mentioned), and the denominators are just the partition functions Z. I think the equations would become clearer if Z was introduced and used. In the following equation (20), the denominators cancel anyway.
We agree with the Referee that mentioning the Lehmann representation and introducing the partition function Z can make the equations easier to follow. This change has been made in the manuscript.

(3) To help the reader, it should be explicitly stated that Eq. (20) follows from applying the equation of motion (15) to the Lehmann representation and solving for Vxc.
The Vxc is defined with the second order Green's function and the hybridization function. In Eq. (20), as pointed out by the Referee, the Vxc is directly solved from the equation of motion of the Green's function by plugging in the Lehmann representation of $G_{ff,\sigma}$. We have added the explanation in the manuscript to avoid ambiguity.

(4) Sec. 3.1, after Eq. (22): I am not sure whether it is appropriate to speak of "Kondo regime" in the context of the Anderson dimer. The Kondo effect is usually associated with an impurity coupled to a continuous band of conduction electrons.
We agree with the Referee that it is more appropriate to speak of "Kondo regime" when a continuous bath is involved. Our purpose of using the Anderson dimer is to derive an analytic form of the dimer Vxc, with which we can propose the Vxc of the Anderson model in the Kondo regime. We have changed the word concerning the dimer to "large U regime" in the manuscript.

(5) The last two sentences of Sec. 3.1, p. 8: I think this explanation for the temperature induced broadening follows simply from the Lehmann representation of the GF (13,14) which the author used to obtain the approximation for the dynamic Vxc.
In our approximation, we effectively cutoff some eigenstates of the dimer system to focus on the additional low-temperature peaks in the spectral function. As the referee summarized, these new peaks follow from expanding the Lehmann representation of $G_{ff,\sigma}$ to the order $e^{-\beta\Delta_0}$. We have added text in the manuscript to point out this.

(6) Is the Vxc given by Eq. (30) valid only for $t > 0$? If so, what is the corresponding equation for $t < 0$? I think it would also be interesting to see Vxc in the frequency domain, i.e. the Fourier transform of Eq. (30), which could then be compared to the self-energy for the SIAM. I suspect they must be very similar in the case of the SIAM.
We thank the Referee for this question, which has helped us improve our treatment. We explain the details in the following, and we keep all spin indices implicit.

When we used the ansatz (Eq. (30) in the original manuscript) to derive the approximate form of G_{ff} (Eq. (31) in the original manuscript)

$$G_{ff}(t > 0) = -0.5i\big[\lambda e^{-i(\omega_p+\mathcal{C})t} + (1-\lambda)e^{-i\mathcal{C}t}\big], \tag{R10}$$

we expanded the term $e^{\lambda(e^{-i\omega_p t}-1)}$ to low order assuming the term $\lambda(e^{-i\omega_p t}-1)$ small. This is a reasonable assumption for small positive t since $\lambda \ll 1$. Assuming that Eq. (R10) applies to all positive time, we calculated $G_{ff}(\omega)$ and thus interpreted the parameters as follows: $\mathrm{Im}[\mathcal{C}] \sim -\Gamma_{\mathrm{H}}$, where Γ_{H} is the half-width of the Hubbard side-band, and $\mathrm{Im}[\mathcal{C}+\omega_p] \sim -\Gamma_{\mathrm{K}}$, where Γ_{K} is the half-width of the Kondo peak and is much smaller than Γ_{H} in the Kondo regime. As a result, ω_p has a positive imaginary part at low temperatures, and $e^{-i\omega_p t}$ tends to infinity for large positive time. This means that $\lambda(e^{-i\omega_p t}-1)$ cannot be treated as a small quantity for large positive time, and both Eq. (R10) and our ansatz Eq. (30) should only be valid for small positive t. However, if we only know G_{ff} for small positive t, we cannot calculate $G_{ff}(\omega)$ for $|\omega| \ll 1$.

Here, we explain our revised treatment for large positive time t_l. We know that $G_{ff}(t_l)$ contributes to the Kondo peak (with the half-width Γ_{K}) in the spectral function. Therefore, we propose that the Vxc in the large positive time regime takes the form $V^{\mathrm{xc}}(t_l) \sim -i\Gamma_{\mathrm{K}}$, so that the corresponding Green's function can take the form $G_{ff} \propto e^{-\Gamma_{\mathrm{K}} t_l}$. Note that the Vxc converging to $-i\Gamma_{\mathrm{K}}$ for large time is a direct result of the Kondo effect, and cannot be found in simulations using finite clusters where the bath is not continuous. The complete Vxc approaches different approximate forms in the limits of small time and large time

$$V^{\mathrm{xc}}(t>0) \approx \begin{cases} \lambda\omega_p e^{-i\omega_p t}+\mathcal{C}, & t \text{ small}, \\ -i\Gamma_{\mathrm{K}}, & t \text{ large}. \end{cases} \tag{R11}$$

We notice that an expression

$$V^{\mathrm{xc}}(t>0) = \frac{\lambda(\omega_p+\mathcal{C})+(1-\lambda)\mathcal{C}e^{i\omega_p t}}{\lambda+(1-\lambda)e^{i\omega_p t}} \tag{R12}$$

fulfills the requirement: assuming $\mathrm{Im}[\omega_p] > 0$ and $\omega_p + \mathcal{C} = \Gamma_{\mathrm{K}}$, it reduces to Eq. (30) in the original manuscript for small t, and $V^{\mathrm{xc}}(t) = -i\Gamma_{\mathrm{K}}$ for large t. Using Eq. (R12), one can calculate G_{ff} for all t using $G_{ff} = -\frac{i}{2}e^{-i\int_0^t V^{\mathrm{xc}}(\bar{t})d\bar{t}}$. The calculated $G_{ff}(t)$ takes exactly the same form as Eq. (R10). Using an expression of $G_{ff}(t)$ valid for all t, $G_{ff}(\omega)$ can be calculated, which takes the same form as in in the original manuscript, and no approximation based on low order expansion is required. All the interpretations of the ansatz parameters remain unchanged: $\mathcal{C} = \frac{U}{2} - i\Gamma_{\mathrm{H}}, \omega_p = \frac{U}{2} + i(\Gamma_{\mathrm{H}} - \Gamma_{\mathrm{K}})$, and the whole approach becomes consistent. The asymptotic behavior of V^{xc} (Eq. (R11)) reflects an important and general feature: V^{xc} at small/large $|t|$ determines the spectral function at high/low ω. Admittedly, Eq. (R12) may seem less intuitive. However, as the Referee pointed out in his/her comment on Eq. (20) of the original manuscript, $V^{\mathrm{xc}}(t)$, obtained from the equation of motion and the Lehmann representation of the Green's function, naturally takes a fractional form, with both the numerator and the denominator containing exponential factors of t.

Based on the discussion above, and to ensure better consistency between the ansatz of V^{xc} and the solved G_{ff}, we have added content in the revised manuscript regarding the small-/large-time regime of the Vxc.

To the question about $V^{\mathrm{xc}}(t<0)$, for a particle-hole symmetric system, one can show that $V^{\mathrm{xc}}(-t) = -V^{\mathrm{xc}}(t)$, which means that for $t<0$,

$$V^{\mathrm{xc}}(t<0) = -\frac{\lambda(\omega_p+\mathcal{C})+(1-\lambda)\mathcal{C}e^{-i\omega_p t}}{\lambda+(1-\lambda)e^{-i\omega_p t}} \tag{R13}$$

In principle, we can use Eq. (R12) and (R13) to calculate

$$V^{\mathrm{xc}}(\omega) = \int dt V^{\mathrm{xc}}(t)e^{i\omega t}, \tag{R14}$$

but $V^{\mathrm{xc}}(\omega)$ would have a complicated form. We also want to mention that the Vxc in the frequency domain and the self-energy have the relation

$$\frac{1}{2\pi}\int d\omega' V^{\mathrm{xc}}(\omega-\omega')G_{ff}(\omega') = \big[\Sigma(\omega)+\Delta(\omega)\big]G_{ff}(\omega), \tag{R15}$$

which follows from the equation of motion of G (expressed with the Vxc or the self-energy, and the Hartree term cancels the ϵ_f term for the particle-hole SIAM)

$$i\partial_t G_{ff}(t) - V^{\rm xc}(t)G_{ff}(t) = \delta(t), \tag{R16}$$

$$i\partial_t G_{ff}(t) - \int dt' \big[\Sigma(t-t') + \Delta(t-t')\big] G_{ff}(t') = \delta(t), \tag{R17}$$

where Δ is the hybridization function (in the wide-band limit, $\Delta(\omega) = i\Gamma$).

(7) How did the author arrive at the hyperbolic-tangent form for $R(L)$ fitted to the data in Fig. 3b? Is that based on some theoretical background? Otherwise I think the actual functional form cannot be extrapolated from the calculated data, since the data is still largely in the linear regime. Very different functional forms leading to very different limits $R(L = \infty)$ could be compatible with the data.

We calculated the intermediate quantity R using finite clusters with one interacting site and $L-1$ noninteracting ones ($L \leq 40$ and even, open boundary conditions). The small values of L indicate that the bath in our finite cluster is not continuous. Hence our R—L data does not show a clear converging behavior. We did not use a large cluster ($L \simeq 400$ to approximately approach a continuous bath) to estimate R because large clusters require exponentially increasing computational time, which contradicts the original purpose of using the dynamical xc field formalism to reduce the heavy numerical workload.

The essential idea of the dynamical xc field formalism is to circumvent a complicated numerical problem by finding a suitable reference system, where the numerical efforts can be reduced and known theories can be applied. Following this idea, we analyzed the asymptotic behavior of the Vxc in both the small- and large-time regimes, proposed an ansatz for the Vxc, and determined most of its parameters using Fermi-liquid theory. To estimate the only parameter without theoretical reference, we performed calculations on small clusters and noticed that R showed a linear behavior for $L < 20$ and should converge to a constant when $L \to \infty$. A hyperbolic-tangent function was a convenient way to fit such R—L relation. Surely, using another function (which is also linear for small L and converges for large L) may lead to different value of $R(L = \infty)$. Here, we show a brief result of the spectral function of a symmetric SIAM with $U = 3, \Gamma = 0.5$ (see Fig. 1 in this response), but assuming a 30% deviation in the R value obtained by extrapolation. We can see that the noticeable change in the spectral function is the height of the Hubbard sideband peak. The Kondo resonance peak is almost unchanged since we have theoretical justification related to its height and width. Additionally, the dynamical xc field formalism is formally exact, meaning

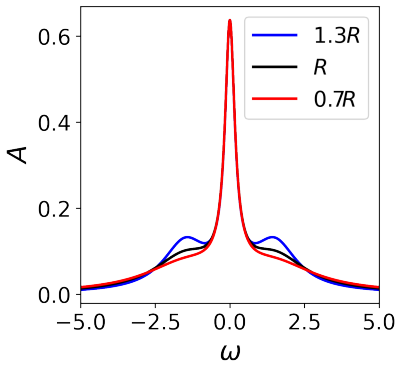

FIGURE 1. Spectral function of a symmetric SIAM with $U = 3, \Gamma = 0.5$. Changing the value of R $\pm 30\%$ has very limited influence on the resonance peak.

that the true $V^{\rm xc}$ (which, according to our definition for the symmetric SIAM, contains $UG^{(2)}$ and hybridization effect) is uniquely determined by, and also determines, the Green's function. The description of the true $V^{\rm xc}$, whether using an ansatz in the form of ours or with additional

parameters, is not necessarily unique. For the SIAM, the constraint in choosing an ansatz is that the behavior of $V^{\rm xc}$ in both the small- and large-time regimes must reflect the features of the Kondo spectral function. We believe that our ansatz captures the essential features with relatively few parameters.

To summarize our answer to the Referee's question, the quality of an ansatz is usually evaluated by comparing the results with available benchmarks, which is satisfactory in our case. Our choice is motivated by the linear behavior of $R(L)$ at small L and the fact that it should converge as $L \to \infty$. Since most parameters of our ansatz are determined with theoretical backgrounds, and the fitting produces qualitatively good results in comparison with benchmarks, we can evaluate our ansatz as a good description of the true Vxc.

(8) It would be nice if in Figs. 4 and 5 the calculated spectra would be directly compared to the NRG spectra of Refs. [28] and [47].

We thank the Referee for this suggestion. We have added those plots for a more direct comparison.

Finally, we wish to conclude our reply by thanking the referee for his/her remarks and criticisms, that have been very valuable and helpful to modifying the manuscript and make it clearer. We hope to have properly and satisfactorily addressed them, and that the paper is now considered suitable for publication in SciPost Physics.