Report of Referee B -- LY15778/Rivera

**The manuscript presents a variational approach to obtain the eigenspectrum of light-matter interaction models in the context of cavity QED. The approach aims at describing the ultrastrong coupling regime limit, in which the coupling strength is sufficiently intense to modify the vacuum properties of the light modes themselves.  With respect to ab-initio calculations based on density functional theory, the proposed approximated approach has the advantage of making it possible to investigate different interesting properties which are not accessible with exact methods. Remarkably, the proposed method allows one to obtain information on the real-space distribution of the cavity modes. The authors apply the method to an example that serves as a benchmark and that elucidates the origin of the saturation of the light-matter coupling strength in the ultrastrong coupling regime.  The proposed method can help to shed light on how fundamental phenomena as the Lamb shift or Casimir-Polder forces are modified in the USC regime. I believe the manuscript can be of interest to a broad audience, both at theoretical and experimental level.**

We thank the reviewer for their positive view of the manuscript.

**However, before recommending publication in PRL, some important issues should be addressed, and various improvement of the manuscript presentation should be implemented.  - The authors demonstrate the effectiveness of the variational method testing it on a simple unidimensional model, for different numbers of discrete levels of the bare matter system. The method predictions for the eigenenergies are in very good agreement with the results of numerical simulations. However, no information is shown about the accuracy of the predictions on any other physical quantity. As one of the main claim is that the proposed method allows one to obtain information about involved observables, I believe it is necessary to discuss whether the approximated eigenstates reproduce faithfully other physical properties. Moreover, as correlations are included only perturbatively, it would be interesting to show if the method can reproduce the amount of correlations or entanglement of the numerically-evaluated eigenstates.**

We thank the reviewer for this very important question. In order to give a partial answer to this question, we take up the estimation of a correlated observable (between the matter and photon), as may become relevant in future experiments on ultrastrong light-matter coupling. Such an observable, a simple example of which being , besides being a nontrivial example of an observable that might feature in a correlated light-matter spectroscopy (proposed in PNAS 114 (12), 3026-3034 or ACS photonics 5 (3), 992-1005), also acts as a measure of correlation or entanglement between the light and matter systems, as it tracks hybridization of the ground state with states that have excitations of both the matter and photon.

In the figure below, we calculate variationally, perturbatively, and via numerical diagonalization, as a function of the coupling, for the case of a two-level system. As can be seen, the perturbative calculation severely overestimates the magnitude of this correlated observable, while the numerical and variational calculations agree much better. Within the variational theory, the “saturation” of is related to the blueshift of the interacting modes, as well as the decrease in the strength of the interacting modes at the location of the emitter. This strong agreement suggests not only that our framework indeed can reproduce other observables besides the total energy. It also suggests that despite treating correlation/entanglement between matter and photons perturbatively, the expectation values calculated differ vastly from the predictions of perturbation theory in the bare modes. On some level, this is also to be expected because despite treating correlation perturbatively, the energies are in extremely good agreement with exact diagonalization. They would not be if these correlations were not added, especially in Fig. 2 of the main text for the two-level system.



Fig. 3: **Expectation value of the correlated observable** ⟨*A* · *p*⟩ **as a function of coupling.** Parameters are identical to those of the top panel of Fig. 2a, and show that despite correlations being treated perturbatively, they are in excellent agreement with exact diagonalization, while in poor qualitative and quantitative agreement with perturbation theory in the bare photonic modes.

We note that other observables, such as the number of photons, or even the squared expectation value of the field, are difficult to compare directly, and are an active subject of investigation. For example, if we take the number of photons as an example of an observable that could be compared, one would find the variational ansatz of Equation (4) would have no photons in the ground state, by virtue of us taking an effective vacuum state (of photons in the “interacting” modes) as our ansatz. Adding correlations perturbatively would lead to some small number of virtual photons (in the “interacting modes”), but this number would be far less than the expected number of photons (in the “bare (sine) modes”) calculated in the ground state using the exact wavefunction numerically (using the “bare” mode creation and annihilation operators). This small number of “interacting photons” in the variational case is quite physical, reflecting that the photons in the ground state are in fact virtual, and would lead for example, to no photodetection signal.



Fig. S1: **Number of virtual photons (bare and interacting) in the ground state calculated variationally, numerically, and through perturbation theory.** Parameters are the same as in Fig. 2 (top panel) of the main text.

The main reason that makes it difficult to compare to field observables such as photon number is that the variational method we propose here, does not prescribe a relation between the variationally obtained modes and the bare modes of the system. This is somewhat analogous to the situation in Hartree-Fock theory, in which the variational orbitals do not have an obvious relation to quantities that would appear in an exact solution (the same phenomenon also appears in DFT, where the Kohn-Sham orbitals do not have an obvious relation to quantities in the exact solution; only the density does).

To summarize the discussion above, we have now added the discussion of the expectation value to the main text, where we discuss the saturation of light matter coupling. The plot showing is now Fig. 3. We write:

“This mechanism is also reflected in Fig. 3 where we calculate a correlated ground state observable such as which is a measure of entanglement between the ground state and excitations of the photon and matter (details shown in Supplemental Materials). Such observables may play a role in correlated spectroscopies, as proposed in Ref. 28. We show predictions of the value of this observable based on the variational method, numerical diagonalization, and perturbation theory in the bare emitter and photon states. As can be seen, perturbation theory overestimates the magnitude of the energy shift, while both numerical and variational methods capture a saturation and then decrease of this expectation value, showing an apparent de-correlation between light and matter in the deep-strong coupling regime. The results of Fig. 2 and Fig. 3 demonstrate not only the accuracy of our ansatz, but provides insight into the mechanisms by which light-matter coupling saturates in the nonperturbative QED regime. The results of Fig. 2 and Fig. 3 also show that despite correlations being treated perturbatively, it remains possible for correlated observables (and energies) to be predicted with high accuracy.”

We have also added details in the Supplementary Information on how the expectation value was calculated within perturbation theory.

**The results of the proposed method are compared with perturbation theory, but no information whatsoever is provided to clarify how the latter is applied. Some more detail should be included also regarding the numerical simulations, at least in the supplementary information.**

In the Supplementary Information, in the section “Derivation of results for one-dimensional cavity model in the main text”, we add a sub-section titled “Perturbative calculation of the energies” where we show the expressions used to perturbatively calculate the energies. Additionally, in the Supplementary Information, in the section titled “Derivation of results for one-dimensional cavity model in the main text” we have added information on how the numerical diagonalization is performed.

**Can the mode profiles shown in Fig2b be reproduced numerically?**

As mentioned in the discussion on other observables, it is not clear how to obtain these mode profiles from the exact solution, because mode profiles strictly speaking are not a quantity which appears within the exact solution. However, an important clue that these mode profiles are a sensible representation of the physics comes from *Physical review letters* 112.1 (2014): 016401. In that work, they perform an exact solution of a harmonic oscillator which is resonant with a cavity (via a simple Bogoliubov transformation). This leads to polaritonic states as the exact eigenstates of the system. They then calculate photodetection signals for upper and lower polaritonic states, and find that the field is strongly reduced at the location of the emitter, with the position-dependent photodetection intensity looking very similar to the modes we present in this manuscript (we have included a figure from their manuscript below). In the limit of infinite coupling, we find the same result that they do, that the modes (in our framework) or the photodetection signals for excited states (in their framework), form a node. Due to the resonant situation they consider, the photodetection signals probe a single mode. It would be of great interest to consider the interaction of a two-level probe weakly coupled to the system we considered in this manuscript, and the resulting photodetection signals.

However, we note that because of the highly non-resonant nature of the situation we consider in this work, the photodetection signals will not feature strong contribution from any one mode, and thus do not look much like the plots in *Physical review letters* 112.1 (2014): 016401. Nevertheless, the similarity of our individual modes to their resonant photodetection signals is a strong, independent test on the validity of the physical content of these mode profiles. Moreover, it is very useful to retrieve this qualitative mode shape without needing to be able to do an analytical diagonalization, which is impossible in the case we consider, and ultimately reveals that these mode shapes are more general than the treatment in the work of de Liberato.

In the manuscript, we now point this out explicitly in discussing Fig. 2, where we write:

“This is a so-called light-matter decoupling effect, which was proposed in Ref. [49]. In Ref. [49], on the basis of photodetection probabilities for exactly-obtained excited polaritonic eigenstates in a Hopfield model, deLiberato obtained ‘effective field mode profiles’ with a strong dip in the location of the emitter, in qualitative agreement with what we report here.”

**- Some key references regarding implementations of USC systems are missing:  T. Niemczyk et al., Nature Physics 6, 772–776 (2010)  P. Forn-Díaz et al. Phys. Rev. Lett. 105, 237001 (2010)  D. Marković et al., Phys. Rev. Lett. 121, 040505 (2018)  - A recent work on the breakdown of gauge-invariance in the USC regime could be mentioned D. De Bernardis et al. Phys. Rev. A 98, 053819 (2018). A reference to introduce the Thomas-Reiche-Kuhn sum rule should be included after equation (11).**

We have included all of these references in the revised manuscript.We have also included a reference (where we discuss the TRK sum rule) to *Atom-Photon Interactions: Basic Processes and Applications,* by C. Cohen-Tannoudji *et al.*

**Right after equation (12) there might be a typo on $E^{(0)}$**

We thank the reviewer for pointing this out, the parentheses in the superscript have been removed.

**One of the main claims is that the method makes it possible to drop the dipole approximation. A detailed example goes probably beyond the reach of the present manuscript, but some further comment on this point could broaden its scope.**

We thank the referee for this point. We now highlight the more general variational equations in the Supplementary Information. In particular, we point out that in these more general equations, no long-wavelength approximation is made, and the field couples to the matter’s full wavefunctions and vice-versa. In particular, we write under Eq (8):

“In the Supplementary Information, we derive a set of equations for the matter orbitals and photonic mode functions which self-consistently takes into account the correlation energy associated with Eq. 8. These equations take into account the spatially varying wavefunctions to the spatially varying mode functions, just like Eqs. 6 and 7, and therefore do not assume the dipole approximation.”

We hope that such comment broadens the scope of the work, and makes it clearer to the reader that our framework can be applied to systems beyond the dipole approximation.

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