Our reply is in unbolded blue, and additions to the manuscript are in unbolded red.

Report of Referee A -- LY15778/Rivera

**Authors attempt to formulate variational approach to quantum electrodynamics. It is condemned to failure from the early beginnings, because at each order of the perturbation theory the parameters like mass or charge need to be renormalized. It will make sense, however, to consider variational approach to study few level atoms interacting with few photon modes. It would resemble the idea of dressed states by Cohen-Tannoudji, see for example his textbook "Photon and Atoms, Introduction to Quantum Electrodynamics".  If Authors see any advantages of their variational method over dressed states approach, they should consider any specific example and obtain new results, that can be experimentally verified. Otherwise, their work does not satisfy PRL criteria of impact and innovation.**

We thank the reviewer for their important question. Our method is specifically useful in regimes of light-matter coupling in which the dressed state approach does not accurately describe the physics.

To summarize what we will argue in depth below, the dressed state approach does not accurately describe light-matter coupling when the rotating wave approximation (RWA) is invalid. The lack of validity of the RWA is critical to recent experiments that have demonstrated the “ultrastrong coupling regime” of quantum electrodynamics (QED), both in superconducting qubit systems and systems in which an ensemble of emitters is collectively coupled to light. See for example Refs. 3-18 of the manuscript, as well as *Nature Physics* 6.10 (2010): 772. These references are also well-summarized in this very recent review: *Nature Reviews Physics* 1.1 (2019): 19.

Beyond extremely simplified models such as the two-level single mode Rabi model, and the Hopfield model, the only methods to study such ultrastrongly coupled QED systems are numerical diagonalization approaches in which one numerically expresses the full light-matter Hamiltonian and diagonalizes it. This approach however is unscalable in that the dimension of the Hilbert space becomes prohibitive if: more than one atom or electron is in the system, or many cavity modes are taken into account. In typical ultrastrongly coupled QED systems, non-resonant contributions from multiple cavity modes become important, and many virtual photons occupy these modes. **The main advantage of our approach over dressed states is that it can describe ground and excited-state energies and observables in ultrastrongly coupled systems. The main advantage of our approach over numerical diagonalization is that it can handle situations with many modes in a scalable way, while also flexible to describe systems with electron-electron interactions.**

At the same time, our approach, as we have shown in the manuscript, leads to results in very good agreement with numerical diagonalization in precisely this ultrastrong coupling regime. Moreover, as we showed in the manuscript, we applied this approach to a multi-level, multi-mode version of the Rabi model, which qualitatively very-well describes experimental ultra-strongly coupled QED systems. **We made explicit experimentally verifiable predictions about the energy spectrum of the system**, as well as about the electromagnetic field fluctuations as they are modified by light-matter coupling, which in principle can be measured through techniques such as those in *Science* 322.5906 (2008): 1357-1360 or *Science* (2015): aac9788.

Another important advantage that our approach has over the dressed state approach, which is more methodological, and thus secondary, is that the dressed state approach, being analytical in nature, only can be profitably used on a QED Hamiltonian describing two atomic levels coupled to a single light-mode. It also indeed can be used on three-level systems although the expressions become considerably more cumbersome. This is excellent for the vast number of systems where a strong laser field populating a single mode is present, and resonant effects dominate the physics. In these regimes, the dressed state approach is certainly one of the best approaches. However, in ultrastrongly coupled QED systems, it is known that multi-mode effects can play an important role as well as multi-level effects, and off-resonant effects, as well as effects when no strong driving field is present (see for example *Nature Physics* 6.10 (2010): 772). Our approach, as exemplified by the results in our manuscript, which pertain to a four-level system coupled to fifty cavity modes, captures all of these effects.

In what follows, we elaborate on some additional important differences in the physics described by the dressed state approach versus our approach are:

1. While a dressed-state approach to describing coupling between an atom and a cavity *in the absence of a driving field* predicts no change in the ground state in the coupled light-matter system, our approach does correctly predict a change in the ground state, due to two mechanisms. The first is the change in the frequencies of the modes of the electromagnetic field, as a result of light-matter coupling (see Equation (12)) of the main text.

The second mechanism is the energy shift of the ground state due to virtual emission and re-absorption of cavity photons. This position-dependent energy shift of the ground state leads to Casimir-Polder forces, which are known to be beyond the rotating-wave approximation. Importantly however, while the typical treatment of Casimir-Polder forces relies on perturbation theory, our theory predicts accurate results for energy shifts and forces in the non-perturbative regime. In particular we find the new result that the Casimir-Polder force in the non-perturbative regime can be understood as virtual emission and re-absorption of photonic quasiparticles whose electromagnetic field fluctuations are very different from those of the bare cavity (see Figure 2b). We briefly note that in the context of dressed states, the dressed states can be used to perturbatively calculate Bloch-Siegert shifts, which are corrections beyond the RWA. However, this approach implicitly assumes that the RWA presents a weak correction to the states and dynamics, which is not the case in ultrastrongly coupled QED systems.

Therefore, if one does spectral measurements on the system in order to measure the excitation energies, one will find that our approach correctly captures the excitation energies, while the dressed state approach does not. Alternatively, if one measures the vacuum forces on emitters in the cavity, one will find a large deviation from the conventional Casimir-Polder theory. Both of these measurements are clear experimental pathways to verify our predictions.

In order to communicate these points to the reader, we have now added a discussion of the dressed state approach in the main text. In particular, we have added a new paragraph in the final paragraph of the first page:

The variational method developed in this manuscript is particularly suited for dealing with QED systems in the ultrastrong coupling regime, in which the rotating-wave approximation no longer holds. Thus, methods based on the Jaynes-Cummings model such as dressed state approaches [42] are no longer accurate in this regime. Moreover, our approach is particularly suited to scalably dealing with systems with many emitter levels, as well as many cavity modes which may have large virtual photon occupation numbers.

[42] Cohen-Tannoudji, C., Dupont-Roc, J., and Grynberg, G. Atom-Photon Interactions. (1992.)

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. Despite correlations being treated perturbatively, this observable is in excellent agreement with exact diagonalization, while in poor 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 light-matter decoupling is also reflected in Fig. 3, where we calculate a correlated ground state observable such as ⟨*A* · *p*⟩, which is a measure of entanglement between the ground state and excitations of the photon and matter (details shown in SM). As shown in Fig. 3, numerical and variational methods capture a saturation and then decrease of this expectation value. The results of Fig. 2 and 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 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 light-matter decoupling effect, which was proposed in Ref. [48], where, on the basis of photodetection probabilities for exactly-obtained excited polaritonic eigenstates in a Hopfield model, "effective field mode profiles" are obtained 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 SM, 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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