
Second Report of Referee A -- LY15778/Rivera

1: In my previous report I have written:

"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 looks to me that the Authors insist on variational formulation of their non-relativistic version of QED, so let me rephrase my argument. The Hamiltonian in Eqs. (1-3) is ill defined without introducing cut-off (let say \Lambda) in the photon frequencies. If one does so, then the electron mass is apriori an unknown function of \Lambda. Moreover, such a nonrelativistic QED is not a renormalizable theory, and thus one needs to introduce an infinite number of counter-terms into the Hamiltonian to make it independent on \Lambda.

We thank the reviewer for this point. Indeed it is the case that non-relativistic QED is non-renormalizable, and requires an infinite number of counter-terms in order to make the theory independent of the cutoff. For example, charge and mass renormalization in non-relativistic QED, and its consequences for cavity QED are discussed in P.W. Milonni Phys. Rep. **25**, No. 1(1976) and in fact allow for calculations of cutoff dependent quantities such as the Lamb shift in practice. In our work, we analyze only the interaction of the charges with low-energy cavity modes, where there is a well-defined cutoff based on when the underlying cavity material becomes transparent. In some sense, this is analogous to the use of variational methods to analyze electromagnetic interactions of electrons with low-energy optical phonons. We ensure in our work that we only look at these low-energy cavity modes by enforcing boundary conditions on Equation (7) at the cavity walls. We assume that the dynamics of these low-energy cavity modes can be separated from the dynamics of free-space modes, which go on to infinite frequencies.

This assumption is well-justified in cavity-QED experiments, including for example experiments on van der Waals and Casimir-Polder forces on atoms near perfectly conducting walls, where the theoretical calculation of the energy shift due to low-energy modes of the conducting walls is in excellent agreement with experiment (*Physical review letters 68.23 (1992): 3432, Physical review letters* 70.5 (1993): 560). In such calculations, the electron mass is that which has been renormalized by free-space modes. Another important experimental example is in circuit QED, where workers in this field have observed energy shifts arising from many low-energy modes (*Physical review letters* 105.23 (2010): 237001).

In the revised manuscript, after we introduce the vector potential operator and the Hamiltonian of Equations (1-3), we discuss this point (see blue text, as well as Supplementary Materials page 2).

2: In my previous report I have written:

"It would make sense, however, to consider variational approach to study few level atoms interacting with few photon modes." and this holds, since Hamiltonian (1-3) would be well defined self-adjoint Hermitian operator in this case.

3: It looks to me that the ansatz (4) violates the gauge independence, while the complete Hamiltonian Eqs. (1-3) is gauge independent, some comments from the Authors are needed. This gauge independence is probably restored by perturbative treatment of "p A" term.

We thank the reviewer for this very important question. Indeed, from Equation (7) of the manuscript, the Maxwell equations for the modes, which are variational parameters, one finds a condition on the vector potential modes: namely that .

$$\nabla \cdot \left(\left(1 - \frac{e^2}{m\epsilon_0\omega^2} \sum_{n=1}^{N} |\psi_n(\mathbf{r})|^2 \right) \mathbf{F}(\mathbf{r}) \right) = 0 \;.$$

This condition on the modes of the vector potential is essentially the generalized Coulomb gauge condition that is used as the gauge of choice in macroscopic quantum electrodynamics (*Physical Review A* 43.1 (1991): 467), in which one is interested in the interaction of low-energy emitters (like few-level atoms) with electromagnetic fields with optical modes in non-free space systems such as perfectly conducting cavities, dielectric cavities, photonic crystals, plasmonic materials, and many others. In particular, for an emitter interacting with electromagnetic modes of a structure with permittivity epsilon, the generalized Coulomb gauge condition is $\nabla \cdot \epsilon \mathbf{A} = 0$.

While results in macroscopic QED are gauge independent, this generalized Coulomb gauge is the gauge of choice for quantitative analysis of effects such as enhanced spontaneous emission, van der Waals forces of emitters near metallic mirrors, and the strongly-coupled cavity QED interactions we examine in our work. This gauge condition, arising from our variational framework, suggests that the physics of our ansatz is that the matter plays the role of a macroscopic dielectric for the electromagnetic field. For the ansatz of Equation (4), this effective macroscopic dielectric is spatially local ($\epsilon(\mathbf{r}, \mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}')\epsilon(\mathbf{r})$), while for an ansatz that takes into account correlations, such as that of Equation (S8), the gauge condition is $\nabla \cdot \int d\mathbf{r}' \, \epsilon(\mathbf{r}, \mathbf{r}')\mathbf{A}(\mathbf{r}') = 0$ for an effective dielectric which is now spatially non-local. Finally, we note that the reason a gauge arose from the variational analysis is that we assumed from the very start that all interaction with the quantized fields arises only from the vector potential, and that there is no scalar potential contribution.

Here, we have discussed the gauge for the modes of the vector potential. Now, we also remark on the choice of “frame”. We may perform a unitary transformation on the ansatz of Equation (4) as well as the Hamiltonian, encoded by the unitary transformation *U*, which for example, could be considered the Power-Wooley-Zienau (PWZ) transformation $e^{-iq\mathbf{r}\cdot\mathbf{A}(\mathbf{r}_0/\hbar}$, which takes the Hamiltonian from the velocity representation to the length or dipole representation (as discussed for example in “Photons and Atoms” by Cohen-Tannoudji et al.). In this case, the transformed ansatz will have the same energy, as expected by the unitary transformation. Interestingly however, this unitary transformation turns the uncorrelated state in the velocity gauge to a correlated state in the length gauge. Meanwhile, if we used the same ansatz (4) (i.e., with no frame transformation) in the dipolar representation, the energy would certainly be different.

In the revised manuscript, we now add a discussion of these points, after Equation (7) (see blue text, as well as Supplementary Materials page 2).

In summary, I find this paper interesting and worthy of publication, but the Authors should respond to comments (1) and (3).

We thank the reviewer for their positive view of our manuscript, as well as their important comments, which have helped to further clarify the results of our manuscript.

Second Report of Referee B -- LY15778/Rivera

I believe the authors have answered all raised criticism, and I find satisfactory the additional analyses and the modifications implemented on the manuscript.

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