HARVARD
John A. Paulson
School of Engineering
and Applied Sciences

Prineha Narang
Assistant Professor of Computational Materials Science
304 Pierce Hall
29 Oxford St., Cambridge MA 02138
Phone: (617) 496-4710
Email: prineha@seas.harvard.edu

November 4th, 2018

Dr. Michael Thoennessen
Editor in Chief, Physical Review Letters,
American Physical Society,
1 Research Road, Ridge, NY 11961.

Dear Dr. Thoennessen,

We hereby submit our manuscript entitled '**Variational theory of non-relativistic quantum electrodynamics**' by Rivera *et al*. for consideration as a *Letter*.

The ability to achieve ultra-strong coupling between light and matter promises to bring about new concepts for manipulating light at the atomic-scale and, simultaneously, provides fundamentally new insight into quantum electrodynamics (QED) of these systems. It also necessitates new and quantitative theories of QED phenomena in complex electronic and photonic systems. In this work, *we introduce a new paradigm to determine from first principles the dynamics of any system of coupled light and matter in the non-perturbative regime of QED*. Our paper establishes for the first time, an *explicit* set of equations that determines ground-state, excited-state, and real-space properties of an arbitrary multi-electron system interacting with quantized electromagnetic fields in arbitrary photonic structures and in three dimensions. Importantly, we find that despite these equations being variational in origin, these equations give an extremely accurate (> 99%) and semi-analytical account of ground- and excited- state energies in the multi-mode Rabi model, which despite being a very actively studied and highly general model in quantum optics, has no analytical solution.

Interestingly, our general theory reveals unexpected fundamental physics in this model, including screening effects that saturate light-matter coupling in the non-perturbative regime, and blue-shift effects that suppress resonances between photons and electrons. The variational theory advanced in this manuscript also constitutes a non-perturbative theory of Lamb shifts forces felt by atoms in photonic structures. The essential new physics in this non-perturbative theory is that instead of the Casimir-Polder energy shift of an emitter being a result of virtual emission and re-absorption of photons of the bare nanophotonic structure, it results from virtual emission and re-absorption of 'photonic quasiparticles' whose properties are influenced by the emitter itself. This theory reproduces extremely well the energy spectrum of the Rabi model in non-perturbative QED.

Our results represent an important and general advance in physics for many reasons: *1)* Solutions to problems in quantum electrodynamics in the non-perturbative regime are essentially non-existent in literature. *2)* An explicit method to accurately solve such problems are of utmost importance in context of recent experimental advances, and our theory provides just that. *3)* Finally, we expect these equations, being highly general, will provide a new starting point for many researchers analyzing *any* coupled light-matter system in the newly developing field of *ab initio* quantum optics and electrodynamics. We thus expect many additional fundamental contributions to quantum optics and electrodynamics to be derived from this work.

We expect our results will be of great interest to scientists across a broad spectrum of physics including those who study quantum optics, superconducting circuits, cavity QED, chemistry in the strong-coupling regime, condensed matter physics, and first-principles approaches to strongly coupled quantum field theories. For these reasons, we believe that this timely *Letter* is well suited to the broad and interdisciplinary readership of *Physical Review Letters*. Please feel free to contact me at **prineha@seas.harvard.edu** with any additional questions.

Yours Sincerely,

Professor Prineha Narang.

Enclosed is a list of prominent scientists, both theorists and experimentalists, across relevant fields of quantum optics, AMO, condensed matter physics and *ab initio* QED that we recommend as potential reviewers.

Suggested Reviewers & Reviewer Context:

1. **Professor Jeremy J. Baumberg,** Professor of Nanoscience at the University of Cambridge, Email: jjb12@cam.ac.uk (**Picoscale Quantum Optics and Light-matter Interactions**)
2. **Professor Angel Rubio,** Director, Max Planck Institute for the Structure and Dynamics of Matter. Email: angel.rubio@mpsd.mpg.de **(*Ab initio* Methods for Microscopic Dynamics in Optical Materials and Quantum Chemistry)**
3. **Professor Owen Miller,** Professor of Applied Physics, Yale University, Email: owen.miller@yale.edu **(Computational approaches to Light-Matter interactions)**
4. **Professor Aashish Clerk,** Institute of Molecular Engineering, University of Chicago, Email: aaclerk@uchicago.edu **(Quantum Systems)**
5. **Dr. Claudiu Genes,** Max Planck Institute for the Science of Light, Erlangen, *Email:* claudiu.genes@mpl.mpg.de **(Quantum Optics)**
6. **Professor Sir John Pendry,** Faculty of Natural Sciences, Department of Physics, Imperial College, London, *Email:* j.pendry@imperial.ac.uk **(Nonlinear Optics and Photonics)**
7. **Professor Alejandro Rodriguez,** Department of Electrical Engineering, Princeton University, Email: arod@princeton.edu **(Casimir and van der Waals forces)**
8. **Professor Francis C. Spano,** Department of Chemistry, Temple University University, Email: spano@temple.edu **(Strong vibrational light-matter coupling)**
9. **Prof. Gil Refael,** Department of Physics, Mathematics and Astronomy, California Institute of Technology, Email: refael@caltech.edu **(Condensed Matter Physics)**