# Variational Theory of Nonrelativistic Quantum Electrodynamics

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The ability to achieve ultrastrong coupling between light and matter promises to bring about new means to control material properties, new concepts for manipulating light at the atomic scale, and new insights into quantum electrodynamics (QED). Thus, there is a need to develop quantitative theories of QED phenomena in complex electronic and photonic systems. In this Letter, we develop a variational theory of general nonrelativistic QED systems of coupled light and matter. Essential to our *Ansatz* is the notion of an effective photonic vacuum whose modes are different than the modes in the absence of light-matter coupling. This variational formulation leads to a set of general equations that can describe the ground state of multielectron systems coupled to many photonic modes in real space. As a first step toward a new *ab initio* approach to ground and excited state energies in QED, we apply our *Ansatz* to describe a multilevel emitter coupled to many optical modes, a system with no analytical solution. We find a compact semianalytical formula which describes ground and excited state energies very well in all regimes of coupling parameters allowed by sum rules. Our formulation provides a nonperturbative theory of Lamb shifts and Casimir-Polder forces, as well as suggest new physical concepts such as the Casimir energy of a single atom in a cavity. Our method thus give rise to highly accurate nonperturbative descriptions of many other phenomena in general QED systems.

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Recent years have brought an explosion of progress in the study of light-matter interactions in the nonperturbative regime of quantum electrodynamics (QED) [1–4]. Ultrastrong, and even deep-strong coupling has been observed in systems involving superconducting qubits [5–10], large ensembles of molecules [11–18], Landau level systems [19,20], quantum wells coupled to cavities [21,22], oscillators [23], and even in few-molecule systems [24,25]. Proposals for new platforms of ultrastrong coupling include emitters coupling to highly confined polaritons in metals and polar insulators [26], heavy ions coupled to optical media via the Cerenkov effect [27], and many more. Proposed applications of ultra- and deep-strong coupling of light and matter are similarly broad, including simulation of many-body systems [3], altering chemical reactivity [11,15,18,28–31] and electronic transport properties [32], and realizing analogues of nonlinear optical processes with vacuum fluctuations [33]. Concomitantly with these developments are also theoretical developments in the study of QED systems *ab initio*. Through "reduced quantity theories" such as quantum electrodynamical density functional theory (QEDFT) [34–39], one is now able to calculate observables in large molecules coupled to realistic optical cavities [38–40].

In this Letter, we establish a variational framework to analyze complex light-matter systems from first principles. Although *ab initio* methods such as QEDFT are exact in principle and provide access to all observables, a number of practical difficulties arise related to: the lack of simple exchange-correlation functionals to describe the ground-state energy (as well as more involved observables), the difficulty of obtaining real-space information about the photons as they are affected by light-matter coupling, the difficulty of handling excited state energies, and the common use of the long-wavelength (dipole) approximation. A variational framework, as we show, flexibly allows a real-space description of the electrons and photons as they are modified by the coupling and also beyond the dipole approximation. Beyond these advantages, a variational framework also allows conceptual insights, into a simple nonperturbative theory of Lamb shifts, into a quasiparticle description of QED systems, and into the notion of Casimir forces in the limit of one atom. A variational framework also allows compact semianalytical formulae to describe complex systems which may assist the development of functionals for use in QEDFT.

Motivated by all of these potential advantages, we now develop an *Ansatz* in which the ground state can be considered as a factorizable state of effective matter and effective photon quasiparticles, both in their respective vacuum states. This *Ansatz*—reminiscent of, but qualitatively distinct from, the Hartree-Fock *Ansatz* [41] of electronic structure theory—leads to coupled equations describing ground and excited states of the light-matter system. We apply our *Ansatz* to describe ground and excited states in a multilevel emitter coupled to many

 

photonic modes. We find that for light-matter couplings that respect sum rules, our method yields ground and excited state energies to a remarkable accuracy of up to 99%, even in deeply nonperturbative coupling regimes. In regimes where our results are accurate, we have found the effective quasiparticle description of the ground state of QED. Our findings also furnish a nonperturbative theory of the position-dependent energy (Lamb) shifts of ground and excited states that give rise to Casimir-Polder forces. The variational method developed in this Letter is particularly suited for analyzing QED systems in the ultrastrong coupling regime, in which the rotating-wave approximation no longer holds, and subsequently methods based on the Jaynes-Cummings model such as dressed state approaches [42] are no longer accurate.

In general, the QED Hamiltonian is given by $H = H_{\mathrm{mat}} + H_{\mathrm{em}} + H_{\mathrm{int}}$ where $H_{\mathrm{mat}}$ describes the matter in the absence of the quantized electromagnetic field, $H_{\mathrm{em}}$ describes the photons in the absence of the matter, and $H_{\mathrm{int}}$ describes the coupling between light and matter. The matter Hamiltonian takes the form

$$H_{\mathrm{mat}} = \int d^3x\, \psi^\dagger(\mathbf{x})\left(-\frac{\hbar^2\nabla^2}{2m} + v_{\mathrm{ext}}(\mathbf{x})\right)\psi(\mathbf{x}) + \frac{1}{2}\int d^3x d^3x'\, \psi^\dagger(\mathbf{x})\psi^\dagger(\mathbf{x}')V(\mathbf{x}-\mathbf{x}')\psi(\mathbf{x}')\psi(\mathbf{x}), \tag{1}$$

where $v_{\mathrm{ext}}$ is the one-body external potential, $V(\mathbf{x}-\mathbf{x}')$ is the two-body interaction kernel, and $\psi$ is the second-quantized electron field. Parametrizing the electromagnetic field purely in terms of a vector potential $\mathbf{E} = -\partial_t\mathbf{A}$ and $\mathbf{B} = \nabla\times\mathbf{A}$ renders the free electromagnetic Hamiltonian as

$$H_{\mathrm{em}} = \frac{\epsilon_0}{2}\int d^3x\, \epsilon[\partial_t\mathbf{A}(\mathbf{x})]^2 + \mathbf{A}(\mathbf{x})\cdot[\nabla\times\mu^{-1}\nabla\times\mathbf{A}(\mathbf{x})], \tag{2}$$

where $\epsilon$ and $\mu$ represent a nondispersive and positive dielectric and magnetic background that the matter and photon occupy. For cases we consider in this Letter, these will be taken to be unity.

The interaction Hamiltonian takes the form

$$H_{\mathrm{int}} = \frac{-i\hbar e}{2m}\int d^3x\, \psi^\dagger(\mathbf{x})[\mathbf{A}(\mathbf{x})\cdot\nabla + \nabla\cdot\mathbf{A}(\mathbf{x})]\psi(\mathbf{x}) + \frac{e^2}{2m}\int d^3x\, \psi^\dagger(\mathbf{x})\psi(\mathbf{x})\mathbf{A}^2(\mathbf{x}). \tag{3}$$

The full Hamiltonian $H$, which depends on the fields $\psi$ and $\mathbf{A}$, is parametrized in terms of an orthonormal set of electron single-particle wave functions (orbitals) $\{\psi_n\}$ and in terms of a set of photonic mode functions (orbitals) $\{\mathbf{F}_i\}$. The electron field operator takes the form $\psi(\mathbf{x}) = \sum_n \psi_n(\mathbf{x})c_n$. The $c_n$ is an annihilation operator for an electron corresponding to state $n$. The electromagnetic field operator takes the form $\mathbf{A}(\mathbf{x}) = \sum_i \sqrt{\hbar/2\epsilon_0\omega_i}\times[\mathbf{F}_i(\mathbf{x})a_i + \mathbf{F}_i^*(\mathbf{x})a_i^\dagger]$, where the $a_i^{(\dagger)}$ annihilate (create) a photon in mode $i$. The electromagnetic field operator is parametrized by both mode functions and frequencies. The normalization chosen for the electron wave functions is $\int d^3x\, \psi_m^*\psi_n = \delta_{mn}$ while for the photon mode functions, it is $\int d^3x\, \epsilon\mathbf{F}_i^*\cdot\mathbf{F}_j = \delta_{ij}$ [43]. Assumptions behind the form of the Hamiltonian are stated in the Supplemental Material [44], page 2.

Given an *Ansatz* $|\Omega\rangle$ for the ground state of $H$, the variational theorem ensures that $\langle\Omega|H|\Omega\rangle$ is an upper bound for the ground state energy. We choose as our *Ansatz*

$$|\Omega\rangle = \left(\prod_n c_n^\dagger|0_n\rangle\right)\otimes\left(\bigotimes_i|0_i\rangle\right), \tag{4}$$

where $\prod_n c_n^\dagger|0_n\rangle$ represents a "filled Fermi sea" for effectively noninteracting electrons, and $(\bigotimes_i|0_i\rangle)$ represents a "photonic vacuum" for effectively noninteracting photons (see Fig. 1). Implicitly, this *Ansätz*, once we take the expectation value $\langle\Omega|H|\Omega\rangle$, denotes a family of *Ansätze* labeled by all possibilities for the electron wave functions, photon mode functions, and photon mode frequencies. Thus, we minimize the expectation value with respect to $\psi_n$, $\psi_n^*$, $\mathbf{F}_i$, $\mathbf{F}_i^*$, and $\omega_i$. We enforce that the matter and photon remain normalized by constructing the Lagrange function

$$\mathcal{L}[\{\psi_n,\psi_n^*\},\{\mathbf{F}_i,\mathbf{F}_i^*,\omega_i\}] = \langle\Omega|H|\Omega\rangle - \sum_n E_n\left(\int d^3x\, \psi_n^*\psi_n - 1\right) - \sum_i\frac{\hbar\lambda_i}{2}\left(\int d^3x\, \epsilon\mathbf{F}_i^*\cdot\mathbf{F}_i - 1\right), \tag{5}$$

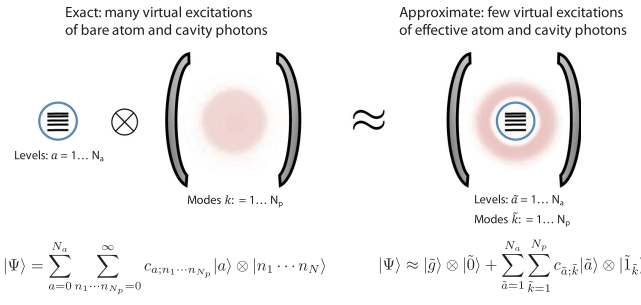


FIG. 1. Ground-state *Ansatz* applied to matter in a cavity: effectively decoupled matter and photons. (Left) Bare description of the coupled light-matter ground state in terms of many virtual excitations of the emitter state and the bare cavity photons. (Right) Quasiparticle description of the coupled system as a factorizable state of an effective emitter in its ground state and the vacuum of an effective photonic degree of freedom.

with the $E_n$ and $\hbar\lambda_i/2$ being the Lagrange multipliers that enforce the normalization conditions. Evaluating the expectation value of the Hamiltonian and minimizing the Lagrange function immediately yields

$$\left(\frac{\mathbf{p}^2}{2m}+v_{\mathrm{ext}}(\mathbf{x})\right)\psi_i(\mathbf{x})+F[\{\psi\}] + \frac{\hbar e^2}{4m\epsilon_0}\left(\sum_n \frac{1}{\omega_n}|\mathbf{F}_n|^2\right)\psi_i(\mathbf{x})=E_i\psi_i(\mathbf{x}), \quad (6)$$

for the electron orbitals and energies, where $F[\{\psi\}]$ represent Hartree-Fock terms (see the Supplemental Material [44]). Here the effect of the QED coupling is to add a one-body ponderomotive potential.

For the photon orbitals and energies, the minimization yields

$$\left[\nabla\times\nabla\times-\frac{\omega_i^2}{c^2}\left(1-\frac{\omega_p^2(\mathbf{x})}{\omega_i^2}\right)\right]\mathbf{F}_i=0, \quad (7)$$

where $\omega_p^2(\mathbf{x})=(e^2/m\epsilon_0)\sum_{n=1}^{N}|\psi_n(\mathbf{x})|^2$ is a position-dependent plasma frequency (squared) which will push the photon orbitals out of the region where the emitter is located. Equations (6) and (7) are main results and can be used to describe ultrastrongly coupled systems in three dimensions, in an arbitrary photonic system, and with multielectron matter. Excited states in this framework can be identified with matter and photon quasiparticle excitations. Taking the divergence of Eq. (7), we see that $\nabla\cdot\{1-[\omega_p^2(\mathbf{x})/\omega^2]\}\mathbf{F}(\mathbf{x})=0$, which is a generalized Coulomb gauge condition on the modes [46]. For more discussion, see the Supplemental Material [44], page 2.

Note that the term in the interaction Hamiltonian linear in the vector potential (the "$A\cdot p$ term") makes no contribution to the expectation value of the ground state of the energy in this *Ansatz*. Physically, this term will mix the factorizable ground state of Eq. (4) with states that have virtual excitations of the matter and the electromagnetic field. The resulting state is nonfactorizable and thus, the $A\cdot p$ term leads to *correlations* in the system, and contributes wholly at lowest order to the correlation energy of QED ground and excited states [47].

We capture the effect of correlations perturbatively. For the ground state, we consider the second-order correction $\delta E$ to the ground-state energy arising from the $A\cdot p$ term. That correction is given by

$$\delta E=\frac{e^2\hbar^2}{8m^2\epsilon_0}\sum_{i=1}^{\infty}\sum_{n=N_\sigma+1}^{\infty}\sum_{m=1}^{N_\sigma}\frac{|\int d^3x\,\mathbf{F}_i^*\cdot\mathbf{j}_{nm}|^2}{\omega_i(\omega_{mn}-\omega_i)}, \quad (8)$$

where $\mathbf{j}_{nm}=\psi_n^*\nabla\psi_m-(\nabla\psi_n^*)\psi_m$, $\omega_{mn}=\omega_m-\omega_n$, and $N_\sigma$ is the number of occupied orbitals, equal to the number of electrons. In a method without self-consistency, the electron and photon orbitals and eigenvalues are those obtained from Eqs. (6) and (7), and then the electron energies and orbitals as well as the photon frequencies and orbitals are plugged into Eq. (8). By considering an *Ansatz* for an excited state, correlation corrections to excited states can also be found. In the Supplemental Material [44], 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 wave functions and the spatially varying mode functions, just like Eqs. (6) and (7), and therefore do not assume the dipole approximation.

In what follows, we provide a proof-of-concept demonstration of the accuracy and content of the variational theory derived here. We consider the QED Hamiltonian corresponding to a single emitter placed at position $z=d$ in a one-dimensional cavity whose axis is along the $z$ direction. As the cavity is considered for simplicity to be one dimensional, the electric field is oriented along a single direction, denoted $x$, while the magnetic field is oriented along a direction transverse to both the electric field and the cavity length, denoted $y$. Working under the long-wavelength (dipole) approximation, the Hamiltonian can then be written as

$$H=H_{\mathrm{matter}}+\frac{\epsilon_0 S}{2}\int dz\,(E^2+c^2B^2)+\frac{q}{m}A(d)p + \frac{q^2}{2m}A^2(d), \quad (9)$$

with q being the emitter charge, $E$ the electric field, $B$ the magnetic field, $A$ the vector potential, and $S$ being a normalization area of the cavity in the $xy$ plane. The fields can be expressed as a mode expansion, where for a cavity of length $L$, the modes are given by $F_n(z)=\sqrt{(2/L)}\sin(n\pi z/L)$ and the corresponding mode frequencies are $\omega_n=n\pi c/L$. We take the matter Hamiltonian to be that of a multilevel system with $N_a$ levels. The matter system we describe can thus be mapped to an $N_a$ site system, which can be considered as a simplified model of a molecule within a tight-binding description. Thus, we parametrize the general family of matter Hamiltonians as $H_{\mathrm{matter}}=\sum_{i=1}^{N_a-1}V_i|i\rangle\langle i|+t(|i\rangle\langle i+1|+|i+1\rangle\langle i|)$. The momentum operator, we write as $p=(-i\hbar/R)\sum_{i=1}^{N_a-1}(|i\rangle\langle i+1|-|i+1\rangle\langle i|)$, where $R$ is a constant with units of length representing roughly the difference in positions between sites. This physical interpretation however is rough: it is also a function of the hopping elements $t$, because we choose $R$ in this Letter such that the Thomas-Reiche-Kuhn (TRK) sum rule is enforced: $(2/m)\sum_{i=2}^{N_a}(|p_{ig}|^2/E_i-E_a)=1$, where $p_{ig}=\langle i|p|g\rangle$ are momentum matrix elements between different matter states [42]. Although the sum rule is based on a full electronic

real-space description, a discrete system which has $(2/m)\sum_{i=2}^{N_a}(|p_{ig}|^2/E_i - E_a) > 1$ cannot exist physically. The TRK sum rule places a bound on how strong the effect of the $A \cdot p$ term can be. The net effect is that the value of $R$ we choose is on the order of $\sqrt{\hbar/2mt}$.

Derivations of the energies of states via the formalism introduced here are shown in the Supplemental Material [44]. Here, we present the main results. Using a one-dimensional version of Eq. (6) and (7), we calculate the electron orbitals, photon orbitals, and photon frequencies in the absence of correlations. In the absence of correlations, we found that the energy of any matter state $a$ with no photonic quasiparticles is given by

$$E_a = E_a^0 + \frac{1}{2}\sum_{n=1}^{\infty}(\hbar\omega_n - \hbar\omega_n^0), \tag{10}$$

where $E_a^0$ is the energy of the matter state in the absence of coupling, $\omega_n$ are found in our framework, $\omega_n^0 = (n\pi c/L)$. The modes found in our framework have their frequencies given by

$$\cot\left(\frac{\omega_n}{c}d\right) + \cot\left(\frac{\omega_n}{c}(L-d)\right) = -\frac{q^2}{m\epsilon_0\omega_n c}. \tag{11}$$

The corresponding "interacting" field mode profiles, shown in Fig. 2(b), are given by compact expressions shown in the Supplemental Material [44]. The result of Eq. (10) says that in the absence of correlations, the energy of the system is the Casimir energy of the system. In particular, it has long been known that when two conducting plates are placed near each other, there is a Casimir energy associated with the fact that the zero-point energy of the nearby plates is different than the zero-point energy of plates infinitely apart. This Casimir energy is simply the difference between the interacting and noninteracting zero-point energies [49,50]. This logic can be applied to any arrangement of macroscopic polarizable objects. What is notable about the result of Eq. (10) is it implies that the same logic about zero-point energy differences can be applied to find the interaction energy of a *single atom* placed near a cavity. In the presence of correlations, we must add to the energy a contribution of the form of Eq. (8), specialized to the case of an emitter in a one-dimensional cavity. We apply the correlation correction to excited states as well, calculating excited-state energy shifts within second-order perturbation theory. In Fig. 2(a), we show the result of this procedure when applied to calculate ground- and excited-state energies for few-level systems coupled to a one-dimensional cavity. The relevant parameters for Fig. 2(a) are listed in the Supplemental Material [44]. For the largest couplings considered here, the magnitude of the energy shift associated with the $A \cdot p$ term predicted from perturbation theory is larger than the energy separation between bare emitter levels, signaling the ultrastrong coupling regime.

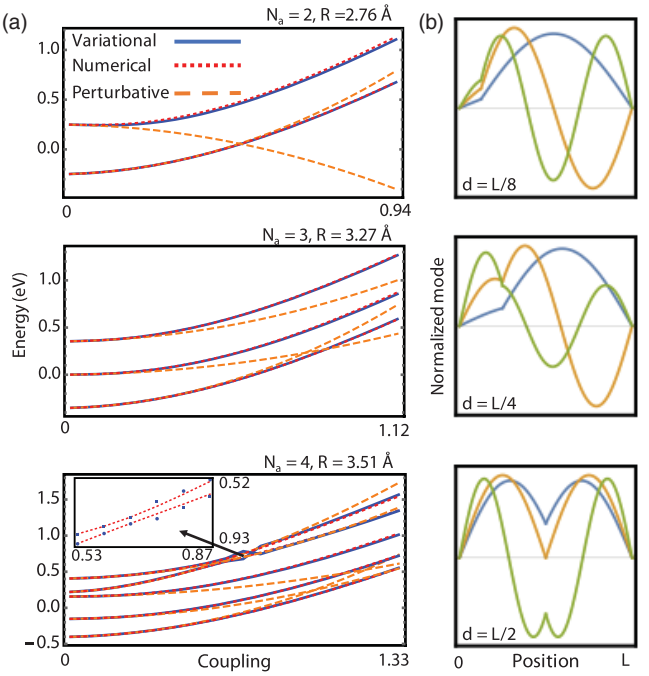


FIG. 2. Variational theory of ground and excited states in nonperturbative QED. (a) Lowest few energy levels of a two- (top), three- (middle), and four- (bottom) level system embedded in the middle of a one-dimensional cavity. The results of the variational method (blue) are compared to perturbation theory (orange), as well as numerical diagonalization (red) with the Fock space truncated to 50 cavity modes with no more than four photons. (Inset) The fourth and fifth energy levels show a weak anticrossing behavior which is reproduced by the variational theory. (b) Mechanism of overestimation of couplings and resonances in perturbation theory: modes derived from the variational theorem are suppressed in the vicinity of the emitter, self-consistently decreasing light-matter coupling.

In all cases, the agreement between our variational approach and numerical diagonalization is excellent, suggesting that our variational method is sufficiently flexible to capture ground states and excited states. The accuracy as a function of number of levels suggests that the breakdown of gauge invariance associated with few-level systems is not crucial to the good agreement between variational and numerical results [51]. Perturbation theory in the bare matter and photon states can both strongly over- and underestimate the energies. Strong disagreement arises in the case of the two-level system (top panel). For the two-level system considered here, the variational result agrees very well with numerical diagonalization, while perturbation theory predicts an energy which evolves with coupling in the wrong direction and is off from the true energy by over 100% for the largest coupling.

Perturbation theory fails for first excited state because the first bare cavity mode is nearly resonant with the transition between ground and excited emitter states,

leading to a very large negative contribution from the $A \cdot p$ term of nearly 2 eV, which is far larger than the spacing of the bare emitter levels. In contrast, no such near resonance is found in the variational framework because the plasma term in Eq. (7) blue shifts all of the photon frequencies. For the largest coupling considered in Fig. 2, we find that the lowest photon frequency is shifted to 0.99 eV (from 0.62 eV), far off resonance from the bare emitter transition. The plasma term also strongly reduces the coupling between light and matter by a mechanism in which the field modes obtained from Eq. (7) are screened out of the emitter, thus self-consistently reducing the strength of the coupling between matter and field [see Fig. 2(b)]. This is a light-matter decoupling effect, which was proposed in Ref. [52], 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.

This light-matter decoupling is also reflected in Fig. 3, where we calculate a correlated ground-state observable such as $\langle A \cdot p \rangle$, which is a measure of entanglement between the ground state and excitations of the photon and matter (details shown in the Supplemental Material [44]). As shown in Fig. 3, numerical and variational methods capture a saturation and then decrease of this expectation value. The results of Figs. 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 Figs. 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.

Our results also demonstrate a nonperturbative theory of the Lamb shift and consequently Casimir-Polder forces.

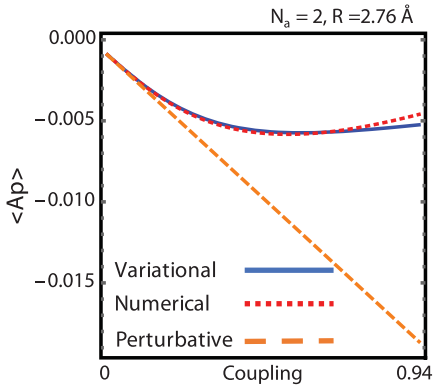


FIG. 3. Expectation value of the correlated observable $\langle A \cdot p \rangle$ as a function of coupling. Parameters are identical to those of the top panel of Fig. 2(a). Despite correlations being treated perturbatively, this observable is in excellent agreement with exact diagonalization, while in poor agreement with perturbation theory in the bare photonic modes.

In particular, it is long known that energy levels of emitters can shift as a result of virtual photon emission and reabsorption. These energy shifts, called Lamb shifts, depend on the particular position of the emitter in the photonic structure it is embedded in. These shifts not only lead to changes in the transition frequencies of the emitter, but the position dependence of these energy shifts also implies forces on the emitter, often called Casimir-Polder forces. Such forces are calculated by applying second-order perturbation theory in the form of Eq. (8) using *bare* atomic and photonic properties [53]. Our calculation of the Lamb shifts via Eq. (8) says that the shifts result from virtual emission and reabsorption of the photonic quasiparticles (the *interacting* modes), which are dependent on properties of the matter. As these interacting photon modes differ greatly from the bare modes and frequencies in the nonperturbative regime, Eq. (8) using interacting modes provides a compact and conceptually simple extension of the theory of Lamb shifts and Casimir-Polder forces to the nonperturbative regime.

The theory posed here could form the basis of an understanding of Lamb shift, Casimir forces, and potentially many other phenomena in the ultrastrong coupling regime for complex QED systems beyond current analytical and numerical methods.

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