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Reconciling quantum and classical spectral theories of ultrastrong coupling: role of cavity bath coupling and gauge corrections

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Cavity quantum-electrodynamics (QED) is a rich area of optical physics, where extreme light–matter coupling can give rise to ultrastrong coupling. The ultrastrong coupling regime presents some fascinating uniquely quantum mechanical effects, such as ground state virtual photons and vacuum squeezing. Focusing on the widely adopted Hopfield model with cavity dissipation, we show how the linear spectrum of an ultrastrong coupled cavity and a dipole can be described either classically or quantum mechanically, but only when the quantum model includes (i) corrections to maintain gauge invariance, and (ii) a specific type of cavity bath coupling, which has so far not been identified. We also show the impact of this bath model on the quantum Rabi model, which has no classical analog in ultrastrong coupling. These results can be used to guide emerging experiments and significantly impact current models and interpretations of ultrastrong coupling between light and matter.

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1. INTRODUCTION

Strong coupling between a single cavity mode and a dipole or two-level system (TLS) [1–3] can be well explained quantum mechanically or classically (or semiclassically) [4–6]. The characteristic signature of strong coupling is a splitting in the emitted spectrum by $2g$, where g is the dipole–cavity coupling rate, which exceeds any losses in the system, e.g., $g^2>\kappa^2/16$ [3] (or more strictly $g^2>\kappa^2/8$ [7]), with κ the cavity decay rate. Quantum mechanically, this is referred to as vacuum Rabi splitting, or in classical physics as normal-mode splitting. Strong coupling has been observed in atoms [2], molecules [8,9], quantum dots [10,11], and circuit quantum-electrodynamics (QED) [12,13], and is often considered a prerequisite for exploring unique quantum effects when one moves beyond a weak excitation approximation or linear response [14,15]. In a quantum description of cavity-TLS coupling, multi-photon effects manifest in an anharmonic response [16–19], which is not captured by the physics of two coupled classical harmonic oscillators (HOs). Nevertheless, a classical description of the emitted spectrum under weak excitation is an adequate description of the system, and one recovers a perfect quantum-to-classical correspondence of the light–matter system, albeit with a different interpretation. Indeed, the quantum interpretation of spontaneous emission can be described in terms of radiation reaction, vacuum fluctuations, or a mixture of both these effects [20,21].

Quantum and classical descriptions of certain light–matter coupling have led to interesting interpretations and insights, including the difference between quantum and classical oscillations in phase qubits [22], classical pseudo-Rabi oscillations in flux qubits [22], and vacuum Rabi splitting as a manifestation of linear-dispersion theory [4].

Beyond strong coupling, recent interest in cavity-QED has turned to ultrastrong coupling (USC) [23–30], where one cannot invoke a rotating wave approximation (RWA), typically when $g \geq 0.1\omega_c$, where ω_c is the cavity resonance frequency. The regime of USC presents some fascinating uniquely quantum concepts such as virtual photons in the ground state [28–31]. Squeezed vacuum states, also with no classical analog, occur in both bosonic and TLS emitter systems in the USC, which are described using the Hopfield model and the quantum Rabi model (QRM), respectively [29]. Exciton and many-emitter Dicke systems also take on the form of the Hopfield model, such as cavity coupling to two-dimensional electron systems including Landau levels in terahertz cavities [32]. In the USC regime, these systems exhibit spectral signatures that reflect the nature of the quantum Hopfield model. On the other hand, the classical theory of coupled oscillators (including collectively coupled TLSs in the dilute thermodynamic limit) does not require a RWA either, and it might be expected that certain observables in the USC regime should also have a quantum-to-classical correspondence, including the effects of cavity losses.

In probing the Hopfield regime, the optical spectrum is typically measured, and most features are argued as being quantum mechanical in nature, e.g., stemming from diamagnetic coupling terms. Yet, the spectral locations of the upper and lower polaritons can be matched with coupled classical HO theory [23,33–35]. In the presence of dissipation, however, a quantum–classical correspondence is generally not known. Although there exists some quantum Langevin approaches for simplified geometries [23], these are not suitable for calculating higher-order quantum correlation functions and arbitrary cavity geometries. Furthermore, the physics of a cavity-coupled TLS is often said to reduce to cavity-HO physics if one neglects saturation effects [36,37]. However, this is not applicable in the USC regime, even with linear response, since saturation effects are unavoidable in the USC regime due to the virtual excitation of the TLS, including the ground state.

The USC regime presents additional challenges for quantum field models, including: (i) gauge corrections because of a truncated Hilbert space [27,38,39], and (ii) the specific form of the system–bath interaction for the cavity mode influences the results [40,41]. Since a classically coupled HO model (expressed entirely in terms of classical electromagnetic fields) has no issues with gauge invariance, it is essential to seek out if and when such a quantum–classical correspondence can be made. This is not just motivated by fundamental physics reasons, but is practically important since many of the emerging USC experiments require some sort of modelling with classical Maxwell solvers [33,42].

In this work, we first show that, for a lossless system, the spectral poles (resonances) of the Hopfield model precisely overlap a classical HO solution, and these deviate from the QRM as soon as one enters the USC regime. We then introduce a spectral theory of the dissipative Hopfield model, using a gauge-invariant master equation theory expressed in the multi-polar gauge (or dipole-gauge in a dipole approximation), and show how it is possible to identify a specific form of the system–bath coupling that matches the classical solution. Identical spectra are obtained in the Coulomb gauge, as must be the case for a gauge-invariant model.

We choose a common and established classical theory, based on a normal-mode expansion of the cavity Green function with phenomenological decay; a more rigorous approach could use quasinormal modes [43,44]. Additionally, we study the impact of this model on both the dissipative Hopfield model and the dissipative QRM, and show the striking differences between these two models for different $\eta \equiv g/\omega_c$. Usually $\eta>0.1$ is the criterion for the USC regime. Figure 1 illustrates a couple of model cavity-QED systems.

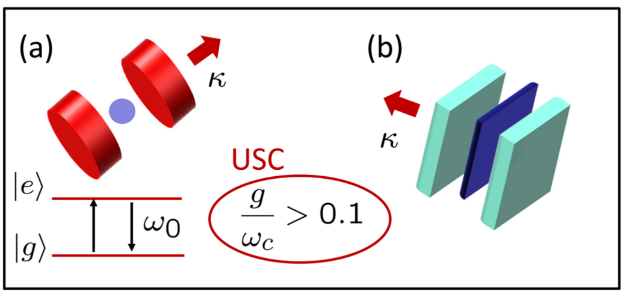

Fig. 1. Simple schematic of two example systems in dissipative cavity-QED that can realize USC, including (a) an atom inside a cavity, which has a decay rate κ, and (b) a planar cavity coupled to a collective emitter system. The emitters can be treated as a bosonic (Hopfield model) or as a TLS (QRM).

2. THEORY

We first consider the interaction between a bosonic cavity mode, with creation (annihilation) operator $a^\dagger, a$, and a bosonic dipole, with creation (annihilation) operator $b^\dagger, b$. Neglecting bath losses for now, in the multi-polar gauge, and with the dipole approximation, the Hopfield model can be written as ($\hbar = 1$)

$$H_{\rm Hop} = \omega_c a^\dagger a + \omega_0 b^\dagger b + ig(a^\dagger - a)(b + b^\dagger) + D(b + b^\dagger)^2, \quad \textbf{(1)}$$

where ω_0 is the atom transition frequency, and $D = \eta g$ is the diamagnetic amplitude [45]. Note that a naïve truncation of the multi-polar gauge Hamiltonian would render this Hopfield D term infinite, and one must account for the gauge invariance of the truncated single-mode model to obtain physical and correct results [39,46].

Using a Bogoliubov transformation, we rewrite this Hamiltonian as $H_{\rm Hop} = \omega_c a^\dagger a + \tilde{\omega}_0 \tilde{b}^\dagger \tilde{b} + i\tilde{g}(a^\dagger - a)(\tilde{b} + \tilde{b}^\dagger) + D$, where $\tilde{\omega}_0 = \omega_0(1 + 4\eta^2)^{0.5}$ and $\tilde{g}^2 = g^2/(1 + 4\eta^2)^{0.5}$. Diagonalization yields two polariton poles [47]: $\omega_\pm^2 = \frac{1}{2}[\tilde{\omega}_0^2 + \omega_c^2 \pm \sqrt{(\tilde{\omega}_0^2 - \omega_c^2)^2 + 16\tilde{g}^2\tilde{\omega}_0\omega_c}]$. Assuming on resonance conditions ($\omega_0 = \omega_c$), then

$$\omega_\pm = \omega_0\sqrt{1 + 2\eta^2 \pm 2\eta(1 + \eta^2)^{1/2}}. \quad \textbf{(2)}$$

To lowest order in the counter-rotating wave effects, i.e., to order η^2 (Bloch–Siegert regime), we obtain $\omega_\pm|_{\rm BS} = \omega_0(1 + \eta^2/2) \pm g$. If one neglects the diamagnetic term, then $\omega_\pm^0 = \omega_0(1 \pm 2\eta)^{0.5}$, which is problematic for $\eta \geq 0.5$, since the lower polariton resonance becomes complex (square root of a negative number). In all our analysis and simulations below, this diamagnetic term is fully included.

We can also compare this solution with the QRM, with

$$H_{\rm QRM} = \omega_c a^\dagger a + \omega_0 \sigma^+\sigma^- + ig(a^\dagger - a)(\sigma^+ + \sigma^-), \quad \textbf{(3)}$$

where σ^+ (σ^-) is the creation (annihilation) operator for the TLS, which has important saturation effects. In this case, we have no diamagnetic term to consider as the relevant TLS term has no effect on energy differences with the TLS operators, since it becomes proportional to the identity operator and only contributes an overall energy shift to the entire system. For the QRM, we have an infinite set of anharmonic eigenenergies, which modifies the Jaynes–Cummings ladder states because of counter-rotating wave terms. Considering again a Bloch–Siegert regime (order η^2 coupling) [30,41], we obtain the poles of the lowest-order polaritons, $\omega_\pm|_{\rm BS}^{\rm QRM} = \omega_0 \pm g(1 + \eta^2/4)^{0.5} \approx \omega_0 \pm g$. These QRM pole resonances differ from the Hopfield model in the Bloch–Siegert regime, even with a linear response.

Linear spectral shifts beyond those in the Jaynes–Cummings model are often termed vacuum Bloch–Siegert shifts [48], but below we quantify why there is nothing uniquely quantum about such resonance shifts in a Hopfield model. This is in contrast to the QRM, which becomes uniquely quantum in nature in the USC regime.

In classical electromagnetism, the bare polarizability volume of an oscillator is $\alpha_0(\omega) = A_0\omega_0/(\omega_0^2 - \omega^2)$, with $A_0 = 2d^2/\epsilon_0$ and d the dipole moment. Considering the emitter position $\mathbf{r}_0$, the photonic Green function, under a single mode expansion (and assuming scalar fields) is [49]

$$G_c(\mathbf{r}_0, \mathbf{r}_0, \omega) = \frac{A_c\omega^2}{\omega_c^2 - \omega^2}, \quad \textbf{(4)}$$

where $A_c = 1/V_{\rm eff}\epsilon_b$, with $V_{\rm eff}$ the effective mode volume and ϵ_b the background dielectric constant. Using the Dyson equation, $G = G_0 + G_0\alpha_0 G = G_0 + G_0\alpha G_0$, where G includes the emitter, one can derive the exact polarizability from $\alpha = \alpha_0 + G_0\alpha$, where α now includes electromagnetic coupling to the environment. With $G_0 = G_c$, we obtain

$$\alpha(\omega) = \frac{A_0\omega_0}{\omega_0^2 - \omega^2 - (\omega_0/\omega_c)4g^2\omega^2/(\omega_c^2 - \omega^2)}, \quad (5)$$

where $4g^2 = d^2\omega_c/(2\epsilon_0 V_{\rm eff}\epsilon_b)$. Since $\omega^2/(\omega_c^2 - \omega^2) = \omega_c^2/(\omega_c^2 - \omega^2) - 1$, we can also write

$$\alpha(\omega) = \frac{A_0\omega_0}{\omega_0^2 - \omega^2 - \frac{2\omega_0 g^2}{\omega_c - \omega} - \frac{2\omega_0 g^2}{\omega_c + \omega} + 4\omega_0 D}, \quad (6)$$

with a co-rotating, counter-rotating, and a static term ($\propto D = g^2/\omega_c$) in the denominator. Note that if a RWA is applied to Eq. (5), then neither the counter-rotating nor the static contributions remain, and so both terms are associated with USC effects, similar to the arguments made from a quantized Hopfield model. Note also that we can replace the atomic polarizability model (α_0) by a Drude–Lorentz model, and obtain classical expressions for the quantum derived vacuum Bloch–Siegert shifts with collective Landau polaritons [32]. In typical Hopfield regime experiments, the interpretation of deviations from a typical RWA Hamiltonian is given explicitly in terms of $H_{\rm CRTs}$ (counter rotating terms) and $H_{\rm dia}$ (diamagnetic coupling), yet here we see a completely classical analogy for both of these contributions, in Eq. (6).

Considering the on-resonance case again, the classical poles are $\omega_\pm^G = \omega_\pm$ [as in Eq. (2)], so it is identical to the solution of the lossless Hopfield model. This is consistent with experimental results on ultrastrong coupled molecular vibrational dipoles in infrared (IR) cavities, where the same classical–quantum correspondence was observed in the oscillator frequencies [50]. In a quantum picture, the blueshift is caused by the polarization-squared term (or the so-called A^2 term in the Coulomb gauge). However, in a classical picture, this blueshift is caused from the poles of the cavity-renormalized polarizability; thus, there is nothing uniquely quantum about this spectral blueshift. Indeed, we have shown above that the classical solution can easily be written to identify precisely the same diamagnetic contribution [Eq. (6)], whose physical original is just a static contribution to the mode sum (Green function).

This correspondence with the poles of the quantum Hopfield model and classical electromagnetism is partly known [33,34], yet sometimes not recovered in quantum models. Moreover, in a linear optical material system, the classical Green function of the hybrid system must have poles in the upper complex plane [51], even with linear gain [52]. The classical correspondence does not mean that there are no unique quantum effects in the Hopfield model, since the ground and excited states are squeezed states [31,53]. Indeed, for $\omega_c = \omega_0$, the quantum ground state, $|0_+0_-\rangle$ has energy $\omega_{0,0} = (\omega_+ + \omega_-)/2 - \omega_0 = \omega_0(1+\eta^2)^{0.5} - \omega_0 > 0$. The ground state contains virtual photons and is an entangled state [54]. Despite this, there appear to be no unique quantum effects that affect the polariton eigenfrequencies.

What is less well known is to what degree the predicted optical spectra agree (or not) between the dissipative quantum and classical coupled mode theories in the USC regime, and how to describe such a regime with open-system master equations. Since all cavity-QED systems have dissipation and input–output channels, it is essential to model them as open quantum systems. Within the RWA, the vacuum Rabi doublets are well described classically or quantum mechanically [4], for both boson and TLS emitters. However, in the USC regime, things are much more subtle, and the quantum models have technical problems related to how to properly include dissipation as well as gauge correction terms (caused by material and cavity mode truncation).

In a classical light–matter model, we can include a heuristic cavity decay rate, κ, in the cavity Green function, and derive the cavity-emitted spectrum as

$$S^{\rm Class}(\omega) = F(\mathbf{R})\left|\frac{E_0\, g^2\omega^2}{(\omega^2 - \omega_c^2 - i\omega\kappa)(\omega^2 - \omega_0^2) - 4g^2\omega^2}\right|^2, \quad (7)$$

where E_0 is the excitation field strength and $F(\mathbf{R})$ is a geometric factor that accounts for light propagation from the system to the particular detection setup. The solution is non-Markovian, causal, and contains no RWA [49]. Also, this phenomenological approach to dissipation ensures a symmetric spectrum outside of the USC regime for a resonant cavity and TLS.

In a quantum picture, to include cavity dissipation, we use an open-system approach [41,55], at the level of a generalized master equation (GME) [41,56–58],

$$\frac{\rm d}{\rm dt}\rho = -\frac{i}{\hbar}[H_{\rm S}, \rho] + \mathcal{L}_{\rm G}\rho + \frac{P_c}{2}\mathcal{D}[X^-]\rho, \quad (8)$$

where $H_{\rm S}$ is the system Hamiltonian (i.e., $H_{\rm Hop}$ or $H_{\rm QRM}$), and P_c is an incoherent pump term, with $\mathcal{D}[O]\rho = 2O\rho O^\dagger - \rho O^\dagger O - O^\dagger O\rho$. The GME cavity dissipator is

$$\mathcal{L}_{\rm G}\rho = \frac{1}{2}\sum_{\omega,\omega'>0} \Gamma_c(\omega)[X^+(\omega)\rho X^-(\omega') - X^-(\omega')X^+(\omega)\rho] + \Gamma_c(\omega')[X^+(\omega)\rho X^-(\omega') - \rho X^-(\omega')X^+(\omega)], \quad (9)$$

where dressed-state operators, $X^\pm$, are defined from

$$X^+(\omega) = \langle j|\Pi_c|k\rangle |j\rangle\langle k|, \quad (10)$$

with $\omega = \omega_k - \omega_j > 0$, $X^- = (X^+)^\dagger$, and Π_c is a cavity operator linear in the photon creation and destruction operators. We neglect atom/emitter decay channels, since these are typically negligible. The cavity decay rates are obtained from $\Gamma_c(\omega) = 2\pi J_c(\omega)$, where $J_c(\omega)$ is the spectral bath density. Below, we use $\Gamma_c = \kappa$, and show that this is sufficient to recover the classical spectral form if the appropriate Π_c operator can be identified.

Recently, it has been shown that the precise form of Π_c matters in the USC regime [41]. For example, one could choose $\Pi_c = i(a^\dagger - a) \equiv P$, or $\Pi_c = a^\dagger + a \equiv Q$, and obtain significantly different predictions, or any linear combination of these two. This is not the case with a RWA. Furthermore, in the USC regime, there a gauge ambiguity for the electric field operator [38], because P represents the Coulomb gauge electric field, and we are using a system Hamiltonian in the dipole gauge. For a restricted TLS subspace, this ambiguity is corrected through the transformation $a' \to \mathcal{U}a\mathcal{U}^\dagger = a + i\eta\sigma_x$ [59], where $\mathcal{U} = \exp(-i\eta(a + a^\dagger)\sigma_x)$ is the projected unitary operator [38,39,60], with $\sigma_x = b + b^\dagger$ (Hopfield model) or $\sigma_x = \sigma^+ + \sigma^-$ (QRM). Thus, one must use a' and $a'^\dagger$ in the computation of the dissipators to ensure gauge invariance. The fact that the Π_c operator should consist only of bosonic a and $a^\dagger$ operators in the Coulomb gauge is a consequence of photon loss being associated only with electromagnetic degrees of freedom, and not the emitter [39].

In the USC regime, the system has transition operators $|j\rangle\langle k|$ which cause transitions between the dressed eigenstates of the system $\{|j\rangle, |k\rangle\}$. For the cavity mode operator, these transitions are obtained from the dressed operators $X^{\pm}$, and again these must be gauge corrected. To make the notation clearer, we can use $X^{\pm}_{\mathrm{GC}}$ to indicate that we are applying Π_c operators with gauge corrections. Thus, the correct cavity-emitted quantum spectrum is obtained from

$$S^{\mathrm{QM}}(\omega) \propto \mathrm{Re}\left[\int_0^{\infty} \mathrm{d}\tau e^{i\omega\tau} \int_0^{\infty} \left\langle X^{-}_{\mathrm{GC}}(t) X^{+}_{\mathrm{GC}}(t+\tau)\right\rangle \mathrm{d}t\right], \quad \textbf{(11)}$$

and calculations without gauge corrections simply use $X^{\pm}$, i.e., without primed cavity operators in the computation of the dissipators and cavity-mode observables. We will show both solutions to better highlight the role of these gauge corrections, and also show how they are required to recover classical correspondence. In all GME calculations below, we use $P_c \ll \kappa$, to ensure weak excitation, and the numerical results are carried out using Python and QuTiP [61,62]. Note this form of time-independent pumping reduces Eq. (11) to a single time integration, since t is evaluated in steady state.

It is important to stress that our gauge-corrected results are necessary to ensure gauge invariance. For example, we could also use a Hopfield model in the Coulomb gauge, where

$$H^{\mathrm{GC}}_{\mathrm{Hop}} = \omega_c a^{\dagger} a + \omega_0 b^{\dagger} b - ig\frac{\omega_0}{\omega_c}(b^{\dagger} - b)(a + a^{\dagger}) + \frac{\omega_0}{\omega_c} D(a + a^{\dagger})^2, \quad \textbf{(12)}$$

and then we could use unprimed operators for the cavity mode, where indicated above, specifically in $X^{\pm}$ and P. Note also that D must be the same in both gauges to ensure gauge invariance, which has been proven also for a dilute Dicke model [34]. For the QRM, the gauge-corrected system Hamiltonian in the Coulomb gauge [38] is $H^{\mathrm{GC}}_{\mathrm{QRM}} = \omega_c a^{\dagger} a + \frac{\omega_0}{2}\{\sigma_z \cos(2\eta(a + a^{\dagger})) + \sigma_y \sin(2\eta(a + a^{\dagger}))\}$, which contains field operators to all orders. For simplicity, we use only the dipole gauge below, but we have checked that all results below are identical in the Coulomb gauge, as must be the case.

3. COMPUTED SPECTRA

Figure 2, shows the classical and quantum solutions for the dissipative Hopfield model, with three types of bath coupling models: (a) $\Pi_c = P$; (b) $\Pi_c = Q$; and (c) $\Pi_c = (P \pm Q)/\sqrt{2}$, where primed cavity operators (discussed above) are used for the gauge-corrected models. We first choose $\eta = 0.5$, which is well into the USC regime, with $\kappa = 0.05g$. As recognized, we find very good agreement with the classical solution only when $\Pi_c = (P \pm Q)/\sqrt{2}$ and only with gauge corrections. To the best of our knowledge, this is the first time that such a solution and correspondence has been made, and these results also demonstrate the significant problem with choosing an arbitrary system–bath interaction form in the USC regime. It should be noted that such a correspondence does not necessarily indicate that this is the correct choice of dissipation model. Rather, this result allows for unambiguous connection and comparison between quantum and classical heuristic models of dissipation, and is further evidence of the limitations of purely phenomenological approaches, when treating losses in open quantum systems.

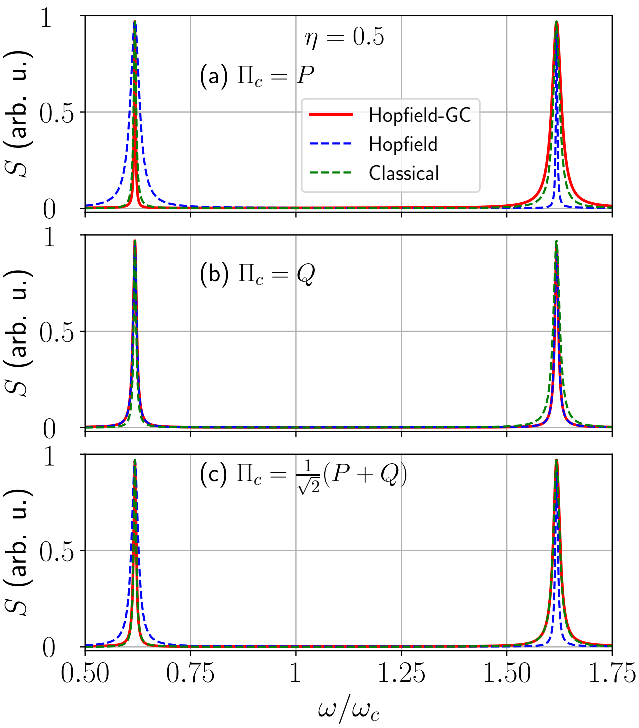

Fig. 2. Hopfield GME results versus the classical model for $\eta = 0.5$, and $\kappa = 0.05g$, with three different bath models, (a) $\Pi_c = P$, (b) $\Pi_c = Q$, and (c) $\Pi_c = (P + Q)/\sqrt{2}$ ($\Pi_c = (P - Q)/\sqrt{2}$ gives identical results). Gauge-corrected results use $X^{\pm}_{\mathrm{GC}}$ and primed cavity operators for Π_c. Only model (c), with gauge corrections ("GC"), overlaps with the classical solution.

Next, we look at the role of this bath coupling model, for different η, using $\Pi_c = (P \pm Q)/\sqrt{2}$, for both the Hopfield model and the QRM. The spectral calculations are shown in Fig. 3, along with the classical solution. We see that the Hopfield model and QRM differ substantially in all USC regimes, and the QRM takes on multiple resonances when η is sufficiently large, even for weak excitation, as well as pronounced spectral asymmetries. Moreover, we find that the dissipative Hopfield model, with $\Pi_c = (P + Q)/\sqrt{2}$, agrees very well with the classical two-oscillator model at all coupling regimes shown. This is clearly not the case for the QRM.

To further support these numerical results, we have derived analytical expressions for the spectral linewidths in the Bloch–Siegert (small η) regime [49]. Specifically, the classical result in Eq. (7) gives linewidths of $\frac{\kappa}{2}[1 \pm \eta]$, for the blue (+) and red (−) peaks, respectively. Using a generic operator $\Pi_c(\theta) = \cos\theta Q + \sin\theta P$, indexed by θ, gives linewidths of

$$\begin{aligned}\Gamma^{\mathrm{Hop}}_{\pm} &= \frac{\kappa}{2}\left[1 \mp 2\eta \sin^2\theta\right], \\ \Gamma^{\mathrm{Hop,\,GC}}_{\pm} &= \frac{\kappa}{2}\left[1 \pm 2\eta \sin^2\theta\right],\end{aligned} \quad \textbf{(13)}$$

showing results without and with ("GC" superscript) gauge corrections, where the latter is the correct result. This shows that a classical correspondence is obtained only when employing gauge corrections and $\Pi_c = (P \pm Q)/\sqrt{2}$ ($\theta = \pm\pi/4$). We stress that the correspondence can only be found when using our gauge-invariant construction, in contrast to other gauge-relative

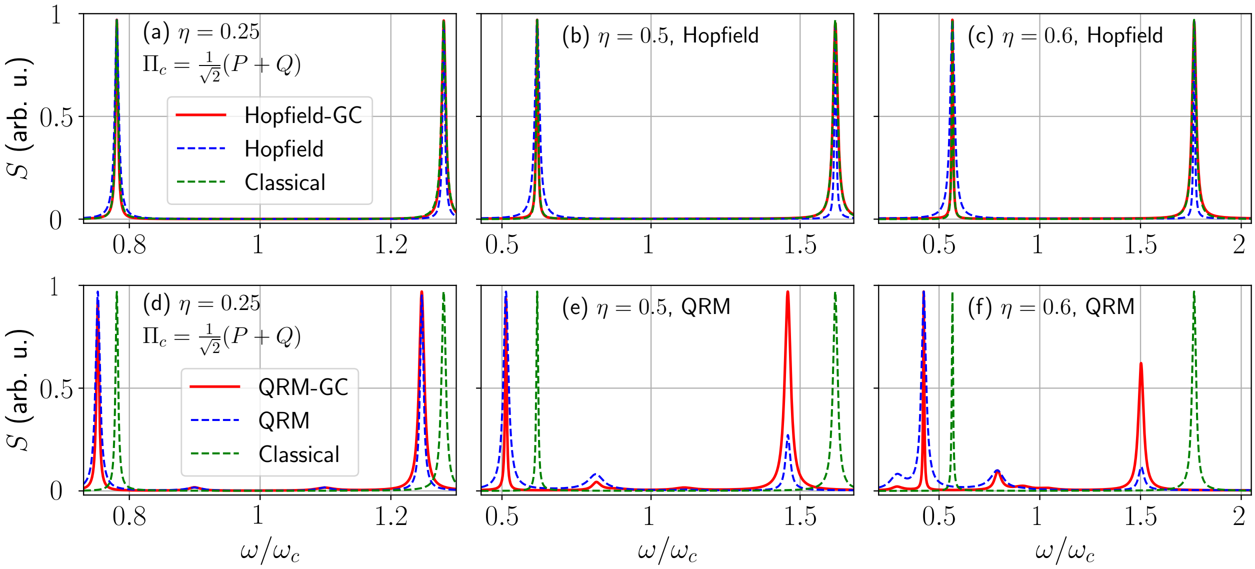

Fig. 3. (a)–(c) Dissipative (GME) Hopfield model and (d)–(f) QRM versus the classical solution, for three values of η. We only present the $\Pi_c = (P+Q)/\sqrt{2}$ bath model and show GME results, with and without gauge corrections. Once again, only the Hopfield model with gauge corrections overlaps with the classical solution. At higher values of η, the QRM also shows multiple resonances, and these are also substantially different with gauge corrections. The QRM fails to recover the classical solution in the USC regime.

approaches [63]. For the QRM, the linewidths are

$$\Gamma_{\pm}^{\text{QRM}} = \frac{\kappa}{2}\left[1 \pm \eta\left(\frac{1}{2} - 2\sin^2(\theta)\right)\right],$$
$$\Gamma_{\pm}^{\text{QRM, GC}} = \frac{\kappa}{2}\left[1 \pm \eta\left(\frac{1}{2} + 2\sin^2(\theta)\right)\right]. \quad \textbf{(14)}$$

In the Bloch–Siegert regime, these analytical decay rates, along with the earlier frequency shifts [to order g^2, $\omega_{\pm}^{\text{Hop}} \approx \omega_0(1 + \eta^2/2) \pm g$ and $\omega_{\pm}^{\text{QRM}} \approx \omega_0 \pm g$], could be used to help compare with experiments with high spectral resolution and increasing η. For sufficiently high η (or strong pumping), differences between the QRM and Hopfield model become substantially different via the emergence of multiple spectral peaks (beyond two) in the spectra (cf. Figs. 2 and 3).

4. DISCUSSION AND CONCLUSIONS

We have shown how the optical spectra for a dissipative Hopfield model in USC can be described quantum mechanically or classically. To achieve correspondence with the classical dissipative result, quantum models must properly respect gauge invariance and implement the appropriate bath coupling operator. Without such a correspondence, any open-system master equations in this regime with *ad hoc* system–bath interactions are ambiguous and can predict wildly differing spectra.

We have also clarified how the dissipative Hopfield model and QRM substantially differ, even for weak excitation, at all USC regimes, including the perturbative Bloch–Siegert regime. Thus, only the Hopfield model yields a classical correspondence under linear response, and this correspondence only occurs with a careful treatment of quantum dissipation and gauge corrections. While we used a normal mode expansion with heuristic broadening, this form is well established for high Q cavities outside the USC regime, and future work could improve such models using classical and quantized quasinormal mode theories [43,44].

One possible clue as to the significance of the $\Pi_c \propto P \pm Q$ coupling can be seen by noting that the classical phenomenological loss model is phase-insensitive. In the quantum loss model, a choice of $P \pm Q$ for the quadrature coupling to the bath is the only choice which gives equal coupling magnitude to each quadrature (i.e., is phase-insensitive in magnitude). Furthermore, we also note that this linear combination of P and Q bath operators also ensures the same linear spectra for cavity pumping or atom pumping (with the same quadrature input coupling for atom pumping), which is not the case with either P or Q models.

Our results also show the different spectral trends of a QRM or a Hopfield model, including cavity dissipation, from the onset of the Bloch–Siegert regime (different linewidth and frequency shifts of the polariton resonances) to USC and deep USC regimes, even with weak, linear excitation fields. Furthermore, they show that non-classical phenomena must be more carefully probed in a Hopfield system, since the typical spectra, including linear reflection and transmission, are exactly the same as a classical field solution. One possibility to explore uniquely quantum field effects could be to excite the system with quantum sources of light.

Broadly, these findings are important for a wide class of light–matter systems now emerging to study the USC regime, including lossy Landau systems and metallic systems [32,33]. Apart from showing a direct classical correspondence for dissipative modes, our results can be used to guide open-system quantum models that are needed when observations are uniquely quantum in nature, e.g., in the QRM for any excitation including coherent excitation, and the Hopfield model excited with non-classical fields.

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Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

Supplemental document. See Supplement 1 for supporting content.

REFERENCES

1. G. S. Agarwal, "Vacuum-field Rabi splittings in microwave absorption by Rydberg atoms in a cavity," Phys. Rev. Lett. **53**, 1732–1734 (1984).
2. A. Boca, R. Miller, K. M. Birnbaum, *et al.*, "Observation of the vacuum Rabi spectrum for one trapped atom," Phys. Rev. Lett. **93**, 233603 (2004).
3. L. C. Andreani, G. Panzarini, and J.-M. Gérard, "Strong-coupling regime for quantum boxes in pillar microcavities: theory," Phys. Rev. B **60**, 13276–13279 (1999).
4. Y. Zhu, D. J. Gauthier, S. E. Morin, *et al.*, "Vacuum Rabi splitting as a feature of linear-dispersion theory: analysis and experimental observations," Phys. Rev. Lett. **64**, 2499–2502 (1990).
5. G. Khitrova, H. M. Gibbs, F. Jahnke, *et al.*, "Nonlinear optics of normal-mode-coupling semiconductor microcavities," Rev. Mod. Phys. **71**, 1591–1639 (1999).
6. S. Hughes and H. Kamada, "Single-quantum-dot strong coupling in a semiconductor photonic crystal nanocavity side coupled to a waveguide," Phys. Rev. B **70**, 195313 (2004).
7. A. Cazé, R. Pierrat, and R. Carminati, "Strong coupling to two-dimensional Anderson localized modes," Phys. Rev. Lett. **111**, 053901 (2013).
8. F. Herrera and F. C. Spano, "Cavity-controlled chemistry in molecular ensembles," Phys. Rev. Lett. **116**, 238301 (2016).
9. J. Flick, M. Ruggenthaler, H. Appel, *et al.*, "Atoms and molecules in cavities, from weak to strong coupling in quantum-electrodynamics (QED) chemistry," Proc. Natl. Acad. Sci. U. S. A. **114**, 3026–3034 (2017).
10. J. P. Reithmaier, G. Sęk, A. Löffler, *et al.*, "Strong coupling in a single quantum dot-semiconductor microcavity system," Nature **432**, 197–200 (2004).
11. T. Yoshie, A. Scherer, J. Hendrickson, *et al.*, "Vacuum Rabi splitting with a single quantum dot in a photonic crystal nanocavity," Nature **432**, 200–203 (2004).
12. X. Gu, A. F. Kockum, A. Miranowicz, *et al.*, "Microwave photonics with superconducting quantum circuits," Phys. Rep. **718-719**, 1–102 (2017).
13. X. Cao, J. Q. You, H. Zheng, *et al.*, "Dynamics and quantum Zeno effect for a qubit in either a low-or high-frequency bath beyond the rotating-wave approximation," Phys. Rev. A **82**, 022119 (2010).
14. I. Schuster, A. Kubanek, A. Fuhrmanek, *et al.*, "Nonlinear spectroscopy of photons bound to one atom," Nat. Phys. **4**, 382–385 (2008).
15. R. Bose, T. Cai, K. R. Choudhury, *et al.*, "All-optical coherent control of vacuum Rabi oscillations," Nat. Photonics **8**, 858–864 (2014).
16. J. M. Fink, M. Göppl, M. Baur, *et al.*, "Climbing the Jaynes–Cummings ladder and observing its nonlinearity in a cavity QED system," Nature **454**, 315–318 (2008).
17. L. S. Bishop, J. M. Chow, J. Koch, *et al.*, "Nonlinear response of the vacuum Rabi resonance," Nat. Phys. **5**, 105–109 (2009).
18. E. Illes, C. Roy, and S. Hughes, "Spectral multiphoton effects and quantum anharmonicities in dissipative cavity-QED systems via off-resonant coherent excitation," Optica **2**, 689 (2015).
19. C. Hamsen, K. N. Tolazzi, T. Wilk, *et al.*, "Two-photon blockade in an atom-driven cavity QED system," Phys. Rev. Lett. **118**, 133604 (2017).
20. P. W. Milonni and W. A. Smith, "Radiation reaction and vacuum fluctuations in spontaneous emission," Phys. Rev. A **11**, 814–824 (1975).
21. P. Milonni, "Semiclassical and quantum-electrodynamical approaches in nonrelativistic radiation theory," Phys. Rep. **25**, 1–81 (1976).
22. A. N. Omelyanchouk, S. N. Shevchenko, A. M. Zagoskin, *et al.*, "Pseudo-Rabi oscillations in superconducting flux qubits in the classical regime," Phys. Rev. B **78**, 054512 (2008).
23. C. Ciuti and I. Carusotto, "Input-output theory of cavities in the ultrastrong coupling regime: the case of time-independent cavity parameters," Phys. Rev. A **74**, 033811 (2006).
24. A. A. Anappara, S. De Liberato, A. Tredicucci, *et al.*, "Signatures of the ultrastrong light-matter coupling regime," Phys. Rev. B **79**, 201303 (2009).
25. F. Beaudoin, J. M. Gambetta, and A. Blais, "Dissipation and ultrastrong coupling in circuit QED," Phys. Rev. A **84**, 043832 (2011).
26. D. Zueco, G. M. Reuther, S. Kohler, *et al.*, "Qubit-oscillator dynamics in the dispersive regime: analytical theory beyond the rotating-wave approximation," Phys. Rev. A **80**, 033846 (2009).
27. D. De Bernardis, P. Pilar, T. Jaako, *et al.*, "Breakdown of gauge invariance in ultrastrong-coupling cavity QED," Phys. Rev. A **98**, 053819 (2018).
28. A. Frisk Kockum, A. Miranowicz, S. De Liberato, *et al.*, "Ultrastrong coupling between light and matter," Nat. Rev. Phys. **1**, 19–40 (2019).
29. P. Forn-Díaz, L. Lamata, E. Rico, *et al.*, "Ultrastrong coupling regimes of light-matter interaction," Rev. Mod. Phys. **91**, 025005 (2019).
30. A. L. Boité, "Theoretical methods for ultrastrong light–matter interactions," Adv. Quantum Technol. **3**, 1900140 (2020).
31. S. D. Liberato, C. Ciuti, and I. Carusotto, "Quantum vacuum radiation spectra from a semiconductor microcavity with a time-modulated vacuum Rabi frequency," Phys. Rev. Lett. **98**, 103602 (2007).
32. Q. Zhang, M. Lou, X. Li, *et al.*, "Collective non-perturbative coupling of 2D electrons with high-quality-factor terahertz cavity photons," Nat. Phys. **12**, 1005–1011 (2016).
33. S. Rajabali, E. Cortese, M. Beck, *et al.*, "Polaritonic nonlocality in light–matter interaction," Nat. Photonics **15**, 690–695 (2021).
34. L. Garziano, A. Settineri, O. Di Stefano, *et al.*, "Gauge invariance of the Dicke and Hopfield models," Phys. Rev. A **102**, 023718 (2020).
35. U. Muniain, J. Aizpurua, R. Hillenbrand, *et al.*, "Description of ultrastrong light-matter interaction through coupled harmonic oscillator models and their connection with cavity-QED Hamiltonians," arXiv, arXiv:2402.11944 (2024).
36. D. Bouchet and R. Carminati, "Quantum dipole emitters in structured environments: a scattering approach: tutorial," J. Opt. Soc. Am. A **36**, 186 (2019).
37. M. A. Jørgensen and M. Wubs, "Quantifying the breakdown of the rotating-wave approximation in single-photon superradiance," J. Phys. B: At. Mol. Opt. Phys. **55**, 195401 (2022).
38. O. Di Stefano, A. Settineri, V. Macrì, *et al.*, "Resolution of gauge ambiguities in ultrastrong-coupling cavity quantum electrodynamics," Nat. Phys. **15**, 803–808 (2019).
39. C. Gustin, S. Franke, and S. Hughes, "Gauge-invariant theory of truncated quantum light-matter interactions in arbitrary media," Phys. Rev. A **107**, 013722 (2023).
40. M. Bamba and T. Ogawa, "Recipe for the Hamiltonian of system-environment coupling applicable to the ultrastrong-light-matter-interaction regime," Phys. Rev. A **89**, 023817 (2014).
41. W. Salmon, C. Gustin, A. Settineri, *et al.*, "Gauge-independent emission spectra and quantum correlations in the ultrastrong coupling regime of open system cavity-QED," Nanophotonics **11**, 1573–1590 (2022).
42. D. G. Baranov, B. Munkhbat, E. Zhukova, *et al.*, "Ultrastrong coupling between nanoparticle plasmons and cavity photons at ambient conditions," Nat. Commun. **11**, 2715 (2020).

43. S. Franke, S. Hughes, M. K. Dezfouli, *et al.*, "Quantization of quasinormal modes for open cavities and plasmonic cavity quantum electrodynamics," Phys. Rev. Lett. **122**, 213901 (2019).
44. S. Franke, M. Richter, J. Ren, *et al.*, "Quantized quasinormal-mode description of nonlinear cavity-QED effects from coupled resonators with a Fano-like resonance," Phys. Rev. Res. **2**, 033456 (2020).
45. S. Savasta, O. D. Stefano, and F. Nori, "Thomas–Reiche–Kuhn (TRK) sum rule for interacting photons," Nanophotonics **10**, 465–476 (2020).
46. M. A. D. Taylor, A. Mandal, and P. Huo, "Resolving ambiguities of the mode truncation in cavity quantum electrodynamics," Opt. Lett. **47**, 1446–1449 (2022).
47. C. Emary and T. Brandes, "Quantum chaos triggered by precursors of a quantum phase transition: the Dicke model," Phys. Rev. Lett. **90**, 044101 (2003).
48. X. Li, M. Bamba, Q. Zhang, *et al.*, "Vacuum Bloch–Siegert shift in Landau polaritons with ultra-high cooperativity," Nat. Photonics **12**, 324–329 (2018).
49. See Supplemental document for more detailed information, including further details on the classical scatteering theory and spectrum, the role of the cavity bath spectrum function, and influence of cavity pump rate.
50. J. George, T. Chervy, A. Shalabney, *et al.*, "Multiple Rabi splittings under ultrastrong vibrational coupling," Phys. Rev. Lett. **117**, 153601 (2016).
51. M. Bamba and T. Ogawa, "System-environment coupling derived by Maxwell's boundary conditions from the weak to the ultrastrong light-matter-coupling regime," Phys. Rev. A **88**, 013814 (2013).
52. S. Franke, J. Ren, M. Richter, *et al.*, "Fermi's golden rule for spontaneous emission in absorptive and amplifying media," Phys. Rev. Lett. **127**, 013602 (2021).
53. T. Makihara, K. Hayashida, G. Timothy Noe II, *et al.*, "Ultrastrong magnon–magnon coupling dominated by antiresonant interactions," Nat. Commun. **12**, 1 (2021).
54. Q. Miao and G. S. Agarwal, "Polaritonic ultrastrong coupling: quantum entanglement in the ground state," Phys. Rev. Res. **5**, 033136 (2023).
55. H. J. Carmichael, *Statistical Methods in Quantum Optics 1: Master Equations and Fokker-Planck Equations* (Springer Science & Business Media, 2013).
56. A. Settineri, V. Macrí, A. Ridolfo, *et al.*, "Dissipation and thermal noise in hybrid quantum systems in the ultrastrong-coupling regime," Phys. Rev. A **98**, 053834 (2018).
57. A. Mercurio, V. Macrì, C. Gustin, *et al.*, "Regimes of cavity QED under incoherent excitation: from weak to deep strong coupling," Phys. Rev. Res. **4**, 023048 (2022).
58. K. Akbari, W. Salmon, F. Nori, *et al.*, "Generalized Dicke model and gauge-invariant master equations for two atoms in ultrastrongly-coupled cavity quantum electrodynamics," Phys. Rev. Res. **5**, 033002 (2023).
59. A. Settineri, O. Di Stefano, D. Zueco, *et al.*, "Gauge freedom, quantum measurements, and time-dependent interactions in cavity QED," Phys. Rev. Res. **3**, 023079 (2021).
60. S. Savasta, O. Di Stefano, A. Settineri, *et al.*, "Gauge principle and gauge invariance in two-level systems," Phys. Rev. A **103**, 053703 (2021).
61. J. R. Johansson, P. D. Nation, and F. Nori, "QuTip: An open-source Python framework for the dynamics of open quantum systems," Comput. Phys. Commun. **183**, 1760–1772 (2012).
62. J. R. Johansson, P. D. Nation, and F. Nori, "QuTiP 2: A Python framework for the dynamics of open quantum systems," Comput. Phys. Commun. **184**, 1234–1240 (2013).
63. A. Stokes and A. Nazir, "Implications of gauge freedom for nonrelativistic quantum electrodynamics," Rev. Mod. Phys. **94**, 045003 (2022).