Gauge-Independent Emission Spectra and Quantum Correlations in the Ultrastrong Coupling Regime of Cavity-QED

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(Dated: February 25, 2021)

A quantum dipole interacting with an optical cavity is one of the key models in cavity quantum electrodynamics (cavity-QED). To treat this system theoretically, the typical approach is to truncate the dipole to two levels. However, it has been shown that in the ultrastrong-coupling regime, this truncation naively destroys gauge invariance. By truncating in a manner consistent with the gauge principle, we introduce master equations to compute gauge-invariant emission spectra and quantum correlation functions which show significant disagreement with previous results obtained using the standard quantum Rabi model, with quantitative differences already present in the strong coupling regime. Explicit examples are shown using both the dipole gauge and the Coulomb gauge.

The intricate interactions between light and matter allow one to observe drastically different behavior depending on the relative magnitude of the light-matter coupling. In the weak-coupling regime, the losses in the system exceed the light-matter coupling strength, and energy in the system is primarily lost before it has the chance to coherently transfer between the matter and the light. Accessing this regime experimentally has allowed for breakthroughs in quantum technologies such as single-photon emitters [1, 2]. Going beyond weak-coupling, the strong-coupling regime is characterized by lower losses in the system, allowing for the observation of vacuum Rabi oscillations: the coherent oscillatory exchange of energy between light and matter. The strong-coupling regime has helped initiate a second generation of quantum technologies [3, 4].

Around 2005, the "ultrastrong-coupling" (USC) regime was predicted for intersubband polaritons [5]. This regime is characterized not by still lower losses, but by a coupling strength that is a comparable fraction of the bare energies of the system. The dimensionless parameter $\eta = g/\omega_0$ (i.e., the cavity-emitter coupling rate divided by the transition frequency) is used to quantify this coupling regime for cavity-QED. Typically, USC effects are expected when $\eta \gtrsim 0.1$, at which point the rotating wave approximation (RWA) used in the weak and strong regimes becomes invalid. Reported signs of USC emerged in 2009 with experiments involving quantum-well intersubband microcavities [6], achieving $\eta \approx 0.11$. Terahertz-driven quantum wells have also demonstrated USC effects [7], and similar effects have been exploited to achieve carrier-wave Rabi flopping with strong optical pulses [8–10]. To date, many different systems have exhibited USC [11, 12]. Recently, using plasmonic nanoparticle crystals, $\eta = 1.83$ has been achieved, with potential to lead to $\eta = 2.2$ [13]. With experiments pushing the normalized coupling strength continuously higher, the interest in USC effects also continues to grow, helping to improve the underlying theories of light-matter interactions. There have also been various predictions made about what novel technologies USC will bring about, including modifications to chemical or physical properties of various systems caused by their USC to light [5, 14], and the potential to create faster quantum gates and gain a high level of control over chemical reactions [11]. To push these advancements forward, it is essential to have a fundamental understanding of the physics involved with these systems and to be able to accurately connect to experimental observables.

The cornerstone model in cavity-QED is constituted by a two-level system (TLS) interacting with a quantized cavity mode. This model has been applied to atoms [15–18], quantum dots [19–22], and circuit QED [23–26]. Rabi initially investigated this system semi-classically in 1936 [27] and developed what is now known as the Rabi model to describe the interactions between a TLS and a classical light field. It took almost 30 years for Jaynes and Cummings to develop the quantized version in 1963 [28]. Their model, the Jaynes-Cummings (JC) model, makes use of a RWA, which has been shown to break down in the USC regime [11, 12, 29]. In this regime, the *quantum* Rabi model (QRM) can be used instead, which does not use the RWA [11]. Notably, emission spectra of coupled qubit-cavity systems are asymmetric without the RWA [30–32].

Gauge invariance.— It has recently been shown that extra care is needed when constructing gauge-independent theories [33] for computing experimental observables for suitably strong light-matter interactions. This development started with a series of papers dealing with so-called gauge ambiguities in the USC regime [34–36]. Gauge in-

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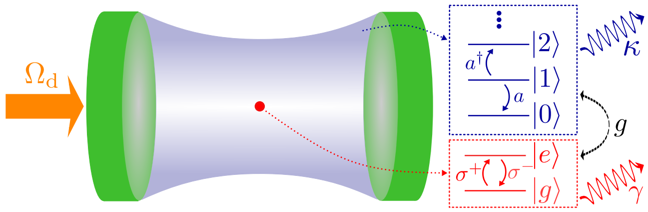

Figure 1. Schematic of a generic cavity-QED system. The optical cavity mode has quantized energy levels (in blue), with a decay rate κ. The matter system is a truncated TLS (in red), with a possible spontaneous emission decay rate γ. The two systems have a coherent coupling strength g. A coherent laser (in orange) drives the system with Rabi frequency Ω_d.

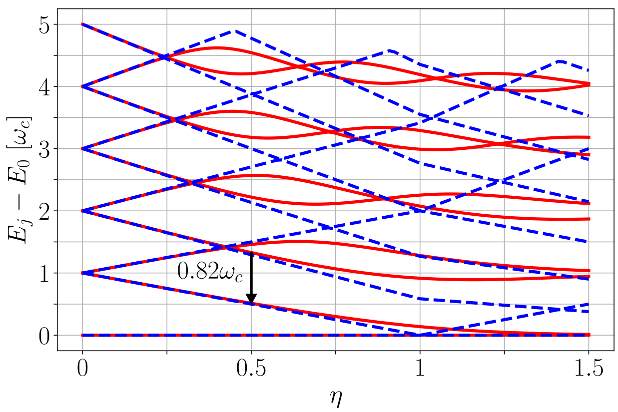

Figure 2. Eigenenergies of the JC Hamiltonian (blue dashed) compared with the QR Hamiltonian (red solid); these begin to disagree significantly around $\eta = 0.1$ (for higher photon numbers). The arrow shows the frequency of the third to second excited state (USC) transition at $\eta = 0.5$, used in the text.

variance is fundamental to QED theory and can be used to reduce the complexity of calculations. Despite visibly different mathematical representations, if a theory is gauge-invariant, all gauges correspond to the same physical laws and physical observables evaluate to the same quantity. Without proper care, gauge invariance of cavity-QED theories can break down when considering USC [37]. This is due to the truncation of the matter system's formally infinite Hilbert space to the two lowest eigenstates in forming the TLS. Only keeping an infinite number of energy levels formally preserves gauge invariance [38]. The impact of these findings is that previous predictions for observables in the USC regime can be ambiguous since the predictions would be impacted by the choice of gauge. This issue was presented as rather insurmountable [37], but has been resolved by using a self-consistent theory at the system Hamiltonian level [36, 39], restoring gauge-invariance to the theory for systems with a finite Hilbert space. Here we extend these works to also ensure gauge-invariance at the master equation level, which is required to treat realistic dissipation, and access experimentally-observable quantities arising from output channels of the cavity.

Model.—In the dipole gauge, we can write the system Hamiltonian, using the QRM and in units of $\hbar = 1$, as

$$H_\mathrm{QR} = \omega_c a^\dagger a + \omega_0 \sigma^+\sigma^- + ig(a^\dagger - a)(\sigma^+ + \sigma^-), \tag{1}$$

where ω_0 is the TLS transition frequency and σ^+ (σ^-) is the raising (lowering) operator for the TLS, ω_c is the frequency of the cavity mode, and $a^\dagger$ (a) is the cavity mode creation (annihilation) operator, which we assume to be a single mode; g is the TLS-cavity coupling strength. In contrast to the Coulomb gauge, straightforwardly truncating the dipole in the light-matter interaction to a TLS subspace does not break gauge invariance in the dipole gauge [36]. Making a RWA on Eq. (1) yields the JC Hamiltonian,

$$H_\mathrm{JC} = \omega_c a^\dagger a + \omega_0 \sigma^+\sigma^- + ig(a^\dagger\sigma^- - a\sigma^+), \tag{2}$$

where the counter-rotating terms $a^\dagger\sigma^+$ and $a\sigma^-$, which do not conserve excitation number, have been neglected. We take $\omega_c = \omega_0$ throughout; the main advantage of this model is that it can easily be solved exactly, and yields the usual JC ladder states that scale with $\pm\sqrt{n}g$, where n is the photon number state. To explore the differences between these models, we can first look at the eigenvalues of the two Hamiltonians, for a range of normalized coupling strengths. The few lowest energy eigenvalues are plotted for both models in Fig. 2. As is now well known, we see that the RWA cannot be used without notable disagreement above $\eta \approx 0.1$, where we enter the USC regime and expect differences between the QRM and the JC model.

For weak to strong-coupling, the usual approach to include dissipation is with a Lindblad master equation [40],

$$\dot{\rho} = -\frac{i}{\hbar}[H_\mathrm{QR}, \rho] + \mathcal{L}_\mathrm{bare}\rho, \tag{3}$$

where ρ is the reduced density matrix. The dissipation term, $\mathcal{L}_\mathrm{bare}\rho = \frac{\kappa}{2}\mathcal{D}[a]\rho$, is the Lindbladian superoperator where $\mathcal{D}[O]\rho = \left(2O\rho O^\dagger - \rho O^\dagger O - O^\dagger O\rho\right)$ and κ is the cavity photon decay rate. Since dissipation is usually dominated by cavity decay, we neglect direct TLS relaxation and pure dephasing [30, 41]; we have checked that our results and conclusions do not change if the TLS relaxation is sufficiently small. The Lindbladian can be derived by following the typical approach in which one neglects the TLS-cavity interaction when considering the coupling of these systems to the environment [24]. However, when moving into the USC regime, this approach fails, and the Lindbladian must be derived while self-consistently including the coupling between the subsystems. For sufficiently strong subsystem coupling, transitions occur between dressed eigenstates of the full Hamiltonian rather than between eigenstates of the individual free Hamiltonians [41]. Indeed, if this dressing is not taken into account, the QRM produces unphysical results, such as system excitation with no pumping [24].

In the USC regime, the system has transition operators $|j\rangle\langle k|$ which cause transitions between the dressed eigenstates of the system $\{|j\rangle, |k\rangle\}$. To obtain these transitions for the cavity mode operator, we use dressed operators [41],

$$x^+ = \sum_{j,k>j} C_{jk}\,|j\rangle\langle k|, \tag{4}$$

and $x^- = (x^+)^\dagger$, where the sum is over states $|j\rangle$ and $|k\rangle$, with $\omega_k > \omega_j$, $C_{jk} = \langle j|\,\Pi_\mathrm{C}\,|k\rangle$, and we neglect thermal excitation effects; Π_C is an operator which couples linearly to dissipation channel modes and, for photon detection, one usually assumes proportional to the cavity electric field operator such that $\Pi_\mathrm{C} = i(a^\dagger - a)$. We then replace $\mathcal{L}_\mathrm{bare}$ in

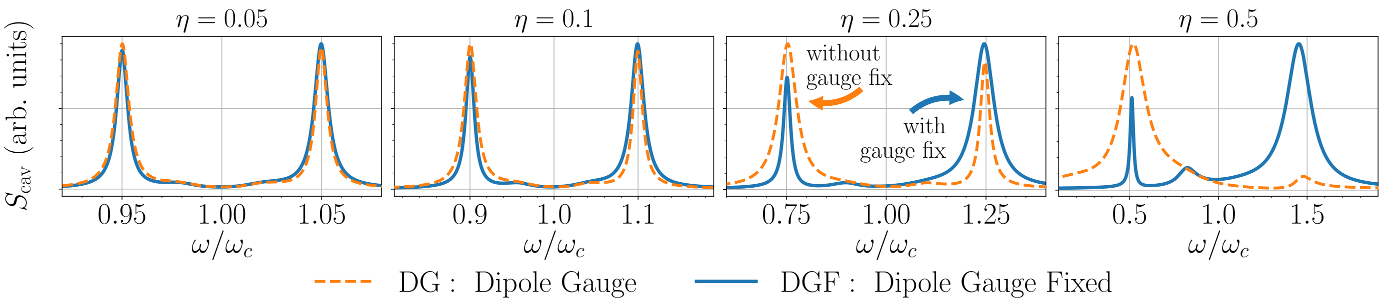

Figure 3. Cavity spectra outside the RWA (QRM) with DG model (orange dashed line), and DGF model (with gauge fix, blue line) for varying η and weak incoherent driving: $P_{\rm inc} = 0.01g$. Spectra are normalized to have the same maxima. Other system parameters are $\kappa = 0.25g$, and $\omega_{\rm L} = \omega_c = \omega_0$. Note a small change with the DSG model even below the USC regime ($\eta = 0.05$).

Eq. (3) with $\mathcal{L}_{\rm dressed}\rho = \frac{\kappa}{2}\mathcal{D}[x^+]\rho$, to arrive at the dressed state (DS) master equation. One can also use a generalized master equation to capture coupling to frequency-dependent reservoirs [41] (see Supplementary Information, Ref 42, for an example of an Ohmic bath).

However, beyond this dressing transformation, it has been shown that there exists a potential gauge ambiguity in the electric field operator which causes further problems when computing observables in the USC regime [36]; namely, $\Pi_{\rm C}$ corresponds to the Coulomb gauge electric field, but the QRM Hamiltonian is derived in the dipole gauge. The gauge transformation from the Coulomb gauge to the dipole gauge is generated by a unitary transformation, which for the restricted TLS subspace is given by the projected unitary operator [36] $\mathcal{U} = \exp(-i\eta(a + a^\dagger)\sigma_x)$. The photon destruction operator transforms as $a \to \mathcal{U}a\mathcal{U}^\dagger = a + i\eta\sigma_x$ [33]. Thus, to "gauge fix" the master equation in the correct dipole gauge, we conduct the dressing operation as above, but with $x^\pm \to x^\pm_{\rm GF} = \sum_{j,k>j} C'_{jk} \left|j\right\rangle \left\langle k\right|$, where we take $C'_{jk} = \left\langle j\right| \mathcal{U}\Pi_{\rm C}\mathcal{U}^\dagger \left|k\right\rangle = \left\langle j\right| \Pi_{\rm D} \left|k\right\rangle = \left\langle j\right| i(a^\dagger - a) + 2\eta\sigma_x \left|k\right\rangle$; see [42] for a derivation of the master equation in the dipole and Coulomb gauges and a proof of their equivalence.

To study the quantum dynamics and spectral resonances, we excite the system with an incoherent pump term, $P_{\rm inc}\mathcal{D}[x^-_{\rm GF}]/2$, or with a coherent laser drive, $H_{\rm drive}(t) = (\Omega_{\rm d}/2)(x^-_{\rm GF}e^{-i\omega_{\rm L}t} + x^+_{\rm GF}e^{-i\omega_{\rm L}t})$, added to $H_{\rm QR}$, where $\Omega_{\rm d}$ is the Rabi frequency and $\omega_{\rm L} = \omega_c$ is the laser frequency; thus, $H_{\rm S} = H_{\rm QR} + H_{\rm drive}$. Note that the QRM with a coherent drive is time-dependent and oscillates around a pseudo-steady-state. In addition, because of the driving laser, the periodic nature of the system Hamiltonian means that in principle the QRM spectra, already quite rich, are modified further to form an infinite manifold of Floquet states; however, we neglect the influence of the coherent drive on the system eigenstates, and use $\Omega_{\rm d} \ll g$.

We define the (incoherent) cavity-emitted spectrum,

$$S_{\rm cav} \propto {\rm Re}\left[\int_0^\infty d\tau e^{i\Omega\tau} \int_0^\infty \left\langle x^-_{\rm GF,\Delta}(t) x^+_{\rm GF,\Delta}(t+\tau)\right\rangle dt\right], \tag{5}$$

where $x^\pm_{\rm GF,\Delta} = x^\pm_{\rm GF} - \left\langle x^\pm_{\rm GF}\right\rangle$ and $\Omega = \omega - \omega_{\rm L}$. Beyond the spectra, which uses a first-order quantum correlation function (CF), we also compute a normalized second-order quantum CF,

$$g^{(2)}(t,\tau) = \frac{G^{(2)}(t,\tau)}{\left\langle x^-_{\rm GF}(t)x^+_{\rm GF}(t)\right\rangle \left\langle x^-_{\rm GF}(t+\tau)x^+_{\rm GF}(t+\tau)\right\rangle}, \tag{6}$$

which quantifies the likelihood of a photon being detected at $(t+\tau)$ if one was detected at t, and $G^{(2)}(t,\tau) = \left\langle x^-_{\rm GF}(t)x^-_{\rm GF}(t+\tau)x^+_{\rm GF}(t+\tau)x^+_{\rm GF}(t)\right\rangle$. We also introduce the time-averaged $g^{(2)}(\tau) = \int_{t_1}^{t_1+T} g^{(2)}(t,\tau)dt/T$, where t_1 is an arbitrary time point at which the system has reached the pseudo-steady-state and T is the period of oscillation [42]. Note without the gauge-fix, we use the unfixed (corresponding to a Coulomb gauge representation) $x^\pm, x^\pm_\Delta$ for computing the observables, and $x^\pm$ for incoherent or coherent driving [42]. All calculations use Python with the QuTiP package [43].

For weak incoherent pumping, Fig. 3 compares the computed spectra with and without the gauge fix (DGF: dipole-gauge fixed and DG: dipole-gauge, respectively), for η ranging from 0.05 (strong coupling) to 0.5 (USC). For relatively small $\eta = 0.05$, the DGF (with gauge fix) spectra already begin to deviate from the DG spectra (usual QRM master equation solution). The *gauge fix evidently introduces some asymmetry even outside the USC regime.*

Next, we increase η to examine the spectra far into the USC regime. Notably, the DGF and DG spectra are now substantially different: the DGF spectra still show a reversed asymmetry, with a significant narrowing of the lower polariton resonance and a broadening of the upper polariton resonance. At $\eta = 0.5$, there is also a profound influence on the oscillator strengths of the resonances with the DGF, and a noticeable resonance around $\omega = 0.8g$, showing a *deep mixing* of the TLS and cavity dynamics in the USC regime. We can identify this energy difference with the transition from the second excited state to the first excited state (cf. Fig. 2, arrow transition).

We have shown how the gauge fix manifests in significant spectral asymmetry, modified damping and drastically different spectral weights in comparison to the usual QRM. For more complex models of dissipation, we also note additional couplings can result in further spectral asymmetry, e.g., electron-phonon interactions for a TLS in a solid-state environment, and pure dephasing [30, 44, 45]. However, the spectral asymmetry we show here is intrinsic and more subtle, and can even result in a complete reversal of the asymmetry predicted from a non-gauge-fixed model (Fig. 3,

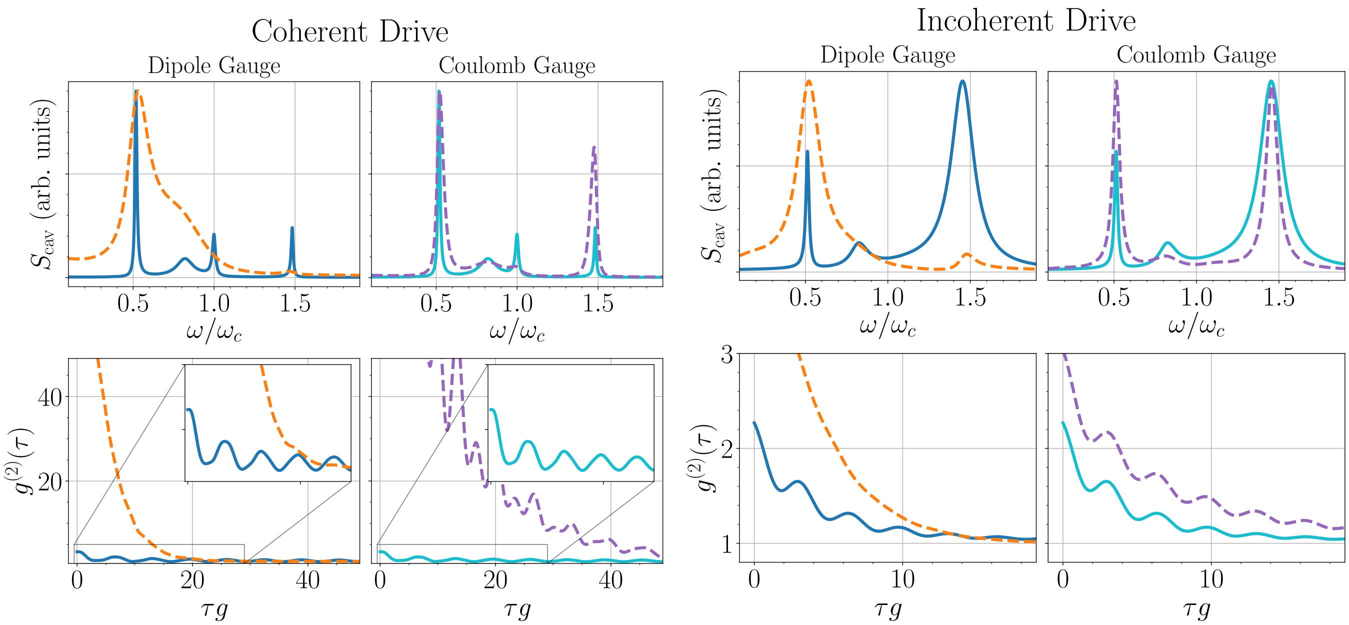

Figure 4. Direct comparison between master equation results using the dipole and Coulomb gauges at $\eta = 0.5$, for both coherent and incoherent excitation, showing the profound effect of gauge-fixing and how this manifests in identical spectra (top) and $g^{(2)}(\tau)$ correlation functions (bottom). Solid and dashed curves are with and without the gauge fix, respectively. For the coherent drive (left), we use $\Omega_{\rm d} = 0.1g$, and the incoherent pumping (right) is the same as in Fig. 3 ($P_{\rm inc} = 0.01g$).

$\eta = 0.5$). Ultimately, this is caused by a gauge-fix modification to the transition matrix elements and emission rates.

Next, we demonstrate how this gauge fix results in truly gauge-invariant observables (shown analytically in [42]). To do this, we will display results for the cavity spectrum and CFs with coherent and incoherent pumping, using the discussed dipole gauge and the Coulomb gauge master equation. The *fixed* Coulomb gauge uses a completely different system Hamiltonian [36],

$$H^{\rm C}_{\rm QR} = \omega_c a^\dagger a + \frac{\omega_0}{2}\left\{\sigma_z \cos(2\eta(a+a^\dagger)) + \sigma_y \sin(2\eta(a+a^\dagger))\right\}, \tag{7}$$

which contains field operators to all orders. In the Coulomb gauge, the gauge-invariant dissipator term is [42]

$$\mathcal{L}^{\rm C}_{\rm dressed}\rho = \frac{\kappa}{2}\mathcal{D}[x^+_{\rm C}]\rho, \tag{8}$$

where $x^+_{\rm C} = \sum_{j,k>j} C^{\rm C}_{jk} |j\rangle\langle k|$ with $C^{\rm C}_{jk} = \langle j|\Pi_{\rm C}|k\rangle$, and we now compute the dressed states in the Coulomb gauge.

Figure 4 (top) shows the coherent and incoherent spectra at $\eta = 0.5$, showing the gauge fix results in either case having a profound effect and agreeing perfectly with each other. For coherent driving, using $\Omega_{\rm d} = 0.1g$, there is a significant sharpening of the resonances, and clearer spectral resonances near $\pm 0.5\,g$. The Coulomb gauge result without the gauge fix corresponds to a minimal coupling Hamiltonian naively truncated to a TLS, which results in incorrect energy levels for the dressed-state master equation [36, 42]. These effects can be even more important at higher pumping strength [42].

Finally, in Fig. 4 (bottom), we examine the second-order coherence, which is important for characterising the generation of non-classical light. In all cases shown, we observe photon bunching at short time-delays. With gauge fixing, there is a significant reduction in the level of bunching, and the usual USC master equations significantly overestimate the bunching characteristics. Moreover, the dynamics are qualitatively different, so clearly *the non-GF master equations results completely fail in these USC regimes.*

While we have shown explicit results for the cavity spectrum and CFs, the gauge fix causes profound effects on *any* observable that is computed from the master equations in the same coupling regimes. The nature of the system-bath coupling is also very important, which must also be related to the coupling to the external fields and the observables to ensure a gauge invariant master equation. For example, it may be more appropriate to use $\Pi_{\rm C} = a + a^\dagger$ (vector potential coupling) rather than $\Pi_{\rm C} = i(a^\dagger - a)$ (electric field coupling) for the interaction and detection (in the Coulomb gauge); this change affects the dissipators, incoherent pumping, and coherent excitation in a way that still yields gauge-independent results (*if* one uses a gauge fix master equation solution), but the observables are different. This is in stark contrast to the JC model, where both these coupling forms yield identical results. It is thus essential to keep the entire master equation theory self-consistent to ensure gauge invariance. Indeed, our theory can also be used to confirm or check the specific form of the system-bath interactions; these various coupling forms, that are widely used in the USC literature and assumed to lead to the same result, in fact differ significantly.

To conclude, we have presented a gauge-invariant master equation approach and calculations for the cavity emission spectra in the USC regime, and shown how the usual QRM in the dipole gauge *fails*, yielding effects that are just as pronounced (or even more pronounced) as counter-rotating wave effects in this regime. We have also shown how the gauge fix significantly affects the cavity CFs.

Apart from having a major influence on the spectra even at $\eta = 0.1$, there are notable quantitative differences at $\eta = 0.05$, which already start to impact standard results in the strong coupling regime. We have also shown the nature of gauge-invariance by explicitly developing and using gauge-invariant master equations in both the dipole gauge and Coulomb gauge, which are shown to yield identical results only with gauge fixing, for the emitted spectrum and for quantum CF dynamics. Apart from yielding new insights into the nature of system-bath interactions, and presenting gauge-invariant master equations that can be used to explore a wide range of light-matter interaction in the USC regime, our results show that currently adopted master equations in the USC regime produce ambiguous results since they do not satisfy gauge invariance.

Acknowledgements.—We acknowledge funding from the Canadian Foundation for Innovation and the Natural Sciences and Engineering Research Council of Canada. F.N. is supported in part by: NTT Research, Army Research Office (ARO) (Grant No. W911NF-18-1-0358), Japan Science and Technology Agency (JST) (via the Q-LEAP program and CREST Grant No. JPMJCR1676), Japan Society for the Promotion of Science (JSPS) (via the KAKENHI Grant No. JP20H00134 and the JSPS-RFBR Grant No. JPJSBP120194828), the Asian Office of Aerospace Research and Development (AOARD), and the Foundational Questions Institute Fund (FQXi) via Grant No. FQXi-IAF19-06. S.S. acknowledges the Army Research Office (ARO) (Grant No. W911NF1910065).

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[42] See Supplemental Material at [URL will be inserted by publisher] for more detailed information, including further numerical details, spectra with an Ohmic DOS, and a pedagogical derivation of the master equations in the main text.

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