

Markovian Approach to N -Photon Correlations beyond the Quantum Regression Theorem

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Multiphoton correlations from quantum emitters coupled to vibrational environments lie beyond the reach of standard tools such as the quantum regression theorem. Here, we introduce a Markovian framework for computing frequency-resolved N -photon correlation functions that overcomes this limitation. Applying our approach to a driven semiconductor quantum dot provides a tractable description of phonon effects on fluorescence beyond the single-photon spectrum. Our method accurately captures the emergence of the phonon sideband, missed by conventional quantum regression theorem treatments, and reveals rich phonon-induced structure in the filtered two-photon spectrum. Strikingly, we find that photons emitted via the phonon sideband inherit second-order coherence properties of the Mollow triplet.

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Introduction—Correlation functions of the electromagnetic field [1] are fundamental tools for characterizing light in quantum optics [2–4]. A central method for computing these functions is the Markovian quantum regression theorem (QRT) [2–6], which extends single-time evolution equations to multitime correlation functions. Despite its widespread use, the QRT has two key limitations: (i) it typically neglects the finite frequency resolution of physical detectors, and (ii) it is formally valid only under the standard assumption of a locally flat environmental frequency response, leading to inaccuracies or even unphysical results outside these conditions [7–13].

The first limitation was addressed by Eberly and Wódkiewicz with the introduction of the physical spectrum [14], and later extended to frequency-resolved higher-order correlation functions [4,15–17]. At higher orders, however, these correlation functions involve solving complex, high-dimensional integrals due to time ordering of field operators, rendering calculations beyond second order practically intractable [16–19]. This changed with the development of the sensor method by del Valle *et al.* [20], which introduces auxiliary two-level systems

that act as frequency-resolved detectors of the emitted light. The sensor method enables computation of correlation functions of arbitrary order in the absence of environmental structure, advancing significantly beyond integral-based techniques [20–26].

In contrast, the restriction of the QRT to flat environments remains a major limitation. For example, in solid-state and molecular emitters, the vibrational environment is structured and plays a central role in determining dynamical and optical properties [27–42]. Direct application of the QRT to the emitter’s output modes fails to describe such scenarios even qualitatively, missing essential physical features such as phonon sidebands (PSBs) [11,13]. It is often assumed that addressing this requires incorporating non-Markovian effects, either through extensions of the QRT [8,9,11], transformations to dressed representations (e.g., polaron) [38,39,43], or sophisticated numerical methods [12,13,44–46].

Here, we show, to the contrary, that the limitations of the QRT can be overcome using only a weak-coupling Markovian master equation when combined with the sensor approach. Our key insight is to trace out the structured (vibrational) environment in the joint system-sensor eigenbasis, so that the sensors respond directly to both environment-induced transitions and intrinsic system dynamics. This yields a unified and fully Markovian treatment of frequency-resolved N -photon correlations in structured vibrational environments. Crucially, our approach retains the conceptual simplicity and flexibility of both the sensor and master equation formalisms, while avoiding numerically intensive computations.

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We apply our method to model electron-phonon interactions in realistic solid-state quantum emitters, where it captures key phonon-induced features in the emission spectrum—including PSBs—missed by conventional weak-coupling QRT calculations [11,13]. Although PSBs in fluorescence are often attributed to non-Markovian effects [11,12], our results show that they can emerge within a weak-coupling Markovian framework. Most importantly, the computational efficiency of our approach makes the study of phonon influences on frequency-resolved multiphoton correlations tractable, a regime previously inaccessible. As an illustration, we compute the emitter two-photon spectrum [21,22], which probes cross-correlations between photons at different frequencies. We uncover a previously unreported phonon-induced feature, showing that signatures of the characteristic correlation structure of the Mollow triplet [15,16,47–49] persist even in photons emitted via the PSB.

The sensor method—We begin by outlining the sensor method of Ref. [20], which applies in the absence of any structured vibrational environment. Suppose a quantum emitter is described by some system Hamiltonian H_S and we are interested in the properties of the radiation it emits. The time evolution of the emitter's reduced density matrix $\rho_S(t)$ is assumed to follow a master equation of the form $(\partial/\partial t)\rho_S(t) = \mathcal{L}_0\rho_S(t)$, with $\mathcal{L}_0\rho_S = -i[H_S, \rho_S] + (\gamma/2)\mathcal{D}_\sigma(\rho_S)$. Here, σ is the lowering operator between the relevant emitter states, $\mathcal{D}_\sigma(\rho) = 2\sigma\rho\sigma^\dagger - \sigma^\dagger\sigma\rho - \rho\sigma^\dagger\sigma$ is a Lindblad dissipator, and γ is the emission rate.

Within the sensor method, frequency-resolved N -photon correlation functions of the emitted light are obtained by introducing N sensors into the system description (see Fig. 1). Sensor m is a two-level system of energy gap ω_m ($\hbar = 1$ throughout), Hamiltonian $H_m = \omega_m\zeta_m^\dagger\zeta_m$, lowering operator ζ_m , and decay rate Γ_m . Each sensor is weakly coupled to the emitter through $H_{S-m} = \epsilon_m(\sigma\zeta_m^\dagger + \sigma^\dagger\zeta_m)$, where ϵ_m is the coupling strength. The master equation describing the composite state of the emitter and sensors, $\rho(t)$, is then

$$\frac{\partial}{\partial t}\rho(t) = \mathcal{L}_0\rho(t) + \sum_{m=1}^N (\mathcal{L}_m\rho(t) - i[H_{S-m}, \rho(t)]), \quad (1)$$

with $\mathcal{L}_m\rho = -i[H_m, \rho] + (\Gamma_m/2)\mathcal{D}_{\zeta_m}(\rho)$. In Ref. [20] it was shown that if the sensor couplings are sufficiently weak not to perturb the emitter's dynamics, the sensors act as detectors with Lorentzian profiles of bandwidth Γ_m centered on ω_m . Frequency-resolved correlation functions of the emitted field then reduce to simple expectation values of sensor operator products. In particular, the steady-state (ss) physical N -photon spectrum is given by [20]

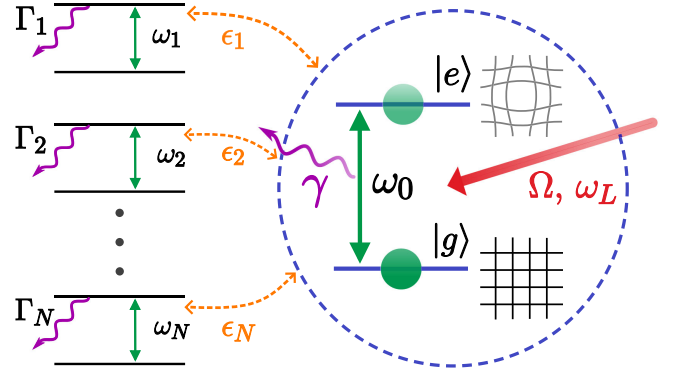


FIG. 1. A schematic of the sensor method. N sensors are weakly coupled to a quantum emitter of energy gap ω_0 and decay rate γ (encircled), with strength ϵ_m for sensor m . Each sensor is a two-level system with energy gap ω_m and decay rate Γ_m . When including vibrations, the quantum emitter is coupled to the host lattice shown to the right. The emitter is externally driven by a laser of frequency ω_L , with Rabi frequency Ω .

$$S_{\Gamma_1, \dots, \Gamma_N}^{(N)}(\omega_1, \dots, \omega_N) = \frac{1}{(2\pi)^N} \frac{\Gamma_1 \cdots \Gamma_N}{\epsilon_1^2 \cdots \epsilon_N^2} \langle :n_1 \cdots n_N: \rangle_{ss}, \quad (2)$$

where $n_m = \zeta_m^\dagger \zeta_m$ is the number operator of sensor m , and $::$ indicates normal ordering [4]. As proved in Ref. [20], this yields results identical to the integral method utilizing the QRT once finite frequency resolution is included in the latter.

Structured environments beyond the QRT—We now move on to the main development of this Letter and describe how the sensor approach can be leveraged to incorporate a structured environment beyond the QRT. For concreteness, we focus on a solid-state quantum emitter interacting with a vibrational environment formed by its host lattice [28,37], modeled as a bath of harmonic oscillators. Each oscillator corresponds to a phonon mode with bosonic annihilation operator b_k and energy ν_k , giving the Hamiltonian $H_E = \sum_k \nu_k b_k^\dagger b_k$. The emitter-phonon interaction is of the form $H_{S-E} = A \sum_k g_k (b_k^\dagger + b_k)$, where the system operator A couples to mode k with strength g_k , characterized by the spectral density $J_{ph}(\nu) = \sum_k |g_k|^2 \delta(\nu - \nu_k)$, which exhibits nontrivial frequency dependence [37].

One could attempt to include this additional environment directly in Eq. (1) by adding a dissipator. However, this fails for structured environments [50–52], since the proof of the original sensor method establishes its equivalence with a QRT-based approach [20]. Such an additive treatment would only be valid if the phonon spectral density was flat and the dissipator of Lindblad form, i.e., under the assumptions of the QRT [6–9]. Key physical features such as PSBs would then be missed.

Instead, we propose an alternative approach: we treat the sensors as part of an extended system and derive a master equation tracing over the phonon environment in the

eigenbasis of the composite emitter-sensor system. Unlike the additive treatment, this ensures the sensors respond to energy exchanges between the emitter and phonons. Moreover, though the original sensor method is equivalent to the QRT, the steady-state expectation value in Eq. (2) is evaluated without invoking it. Our framework therefore enables computation of N -photon spectra without the limitations of the QRT.

To test this idea and demonstrate the power of our approach, we choose to describe the phonon interactions with a weak-coupling Born-Markov theory [6,30,37,53]. While this treatment accurately describes emitter dynamics in the weak phonon coupling and low temperature regime [32,37], it is often assumed to be insufficient for capturing PSBs in the emission spectrum [11,39]. Using our framework, we challenge this assumption and show that the failure to reproduce PSBs arises not from the Markovian weak-coupling theory itself, but from the approximations introduced by the standard QRT approach.

To derive the weak-coupling master equation we move into the interaction picture with respect to the combined emitter-sensor system Hamiltonian $H'_S = H_S + \sum_{m=1}^N (H_m + H_{S-m})$. This gives

$$\tilde{H}_{S-E}(t) = \tilde{A}(t) \sum_k g_k \left(b_k^\dagger e^{i\nu_k t} + b_k e^{-i\nu_k t} \right), \quad (3)$$

where, importantly, the transformed operator $\tilde{A}(t) = e^{iH'_S t} A e^{-iH'_S t}$ acts within the Hilbert spaces of both the emitter and sensors. Following the standard weak-coupling prescription [6,37,53], we obtain a master equation of the form (for further details, see Ref. [54])

$$\frac{\partial}{\partial t} \rho(t) = \mathcal{L}_0 \rho(t) + \sum_{m=1}^N (\mathcal{L}_m \rho(t) - i[H_{S-m}, \rho(t)]) + \mathcal{K}[\rho(t)], \quad (4)$$

where the phonon dissipator $\mathcal{K}(\rho)$ is defined through a rate operator Z as $\mathcal{K}(\rho) = -[A, Z\rho] + [A, \rho Z^\dagger]$. Using the spectral decomposition of the joint Hamiltonian $H'_S = \sum_\alpha \epsilon_\alpha |\psi_\alpha\rangle\langle\psi_\alpha|$, the rate operator is

$$Z = \sum_{\alpha,\beta} \langle\psi_\alpha|A|\psi_\beta\rangle |\psi_\alpha\rangle\langle\psi_\beta| \int_0^\infty d\tau e^{-i(\epsilon_\alpha - \epsilon_\beta)\tau} C(\tau), \quad (5)$$

with phonon correlation function $C(\tau) = \int_0^\infty d\nu J_{\text{ph}}(\nu) [\coth(\beta\nu/2) \cos \nu\tau - i \sin \nu\tau]$ at inverse temperature $\beta^{-1} = k_B T$. We stress again that because H'_S is used rather than H_S in Eq. (3), the phonon dissipator $\mathcal{K}$ acts on the composite emitter-sensor Hilbert space. The N -photon spectrum is obtained by setting the left-hand side of Eq. (4) to zero and solving for the steady state ρ_{ss} of the

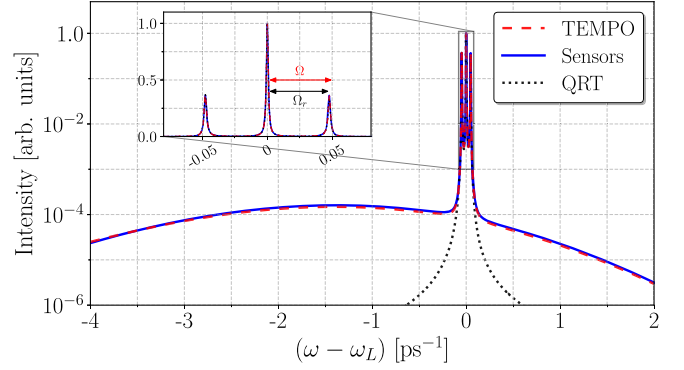


FIG. 2. Emission spectrum of a resonantly driven QD under the influence of a phonon environment calculated using the QRT (dotted, black curves), the sensor-based method introduced in this Letter (solid, blue curves) and the numerically exact TEMPO algorithm (dashed, red curves). The inset shows the Mollow triplet in more detail, where Ω and Ω_r are the original and phonon-renormalized Rabi frequencies, respectively. Parameters used for this plot are $\gamma = 1/700 \text{ ps}^{-1}$, $\Omega = 0.05 \text{ ps}^{-1}$, $\alpha = 0.027 \text{ ps}^2$, $T = 4 \text{ K}$, and $\nu_c = 2.2 \text{ ps}^{-1}$. The sensor width is $\Gamma = 10^{-4} \text{ ps}^{-1}$ and the coupling is $\epsilon = 10^{-6} \text{ ps}^{-1}$.

emitter-sensor system, which is then used in Eq. (2) to compute $S_{\Gamma_1, \dots, \Gamma_N}^{(N)}$.

Phonon influence on QD emission—As an example, we apply our approach to a semiconductor quantum dot (QD) modeled as a two-level system with ground state $|g\rangle$ and excited state $|e\rangle$, separated by an energy gap ω_0 . The QD is driven resonantly by a continuous-wave laser of frequency ω_L . In the rotating frame of the laser, and under the rotating-wave approximation, the system Hamiltonian is $H_S = \delta|e\rangle\langle e| + (\Omega/2)\sigma_x$, where Ω is the Rabi frequency, $\delta = \omega_0 - \omega_L$ is the laser detuning, and $\sigma_x = \sigma + \sigma^\dagger$. Radiative decay with natural linewidth γ is included via the Lindblad dissipator $(\gamma/2)\mathcal{D}_\sigma$ in $\mathcal{L}_0$. This can be included additively, since the spectral density of the electromagnetic environment is well approximated as frequency independent and the dissipator is thus of Lindblad type [34,51,52]. Electron-phonon coupling is through the operator $A = \sigma^\dagger \sigma$, with the phonon environment described by the super-Ohmic spectral density $J_{\text{ph}}(\nu) = \alpha \nu^3 \exp(-\nu^2/\nu_c^2)$, where α is the coupling strength and ν_c is the cutoff frequency [37].

We compute the QD physical emission spectrum $S_\Gamma^{(1)}(\omega)$ using Eqs. (2) and (4) with $N = 1$ sensor. Figure 2 compares results from our approach (solid blue), the standard QRT (dotted black), and the numerically exact TEMPO algorithm [44–46] (dashed red). To obtain the physical spectrum within the QRT and TEMPO methods, the two-time correlation function $\langle \sigma^\dagger(\tau) \sigma(0) \rangle_{\text{ss}}$ is computed and convolved with a Lorentzian of width Γ [14], matching the sensor bandwidth. Importantly, using TEMPO this is generally practicable only for the single-photon spectrum. The

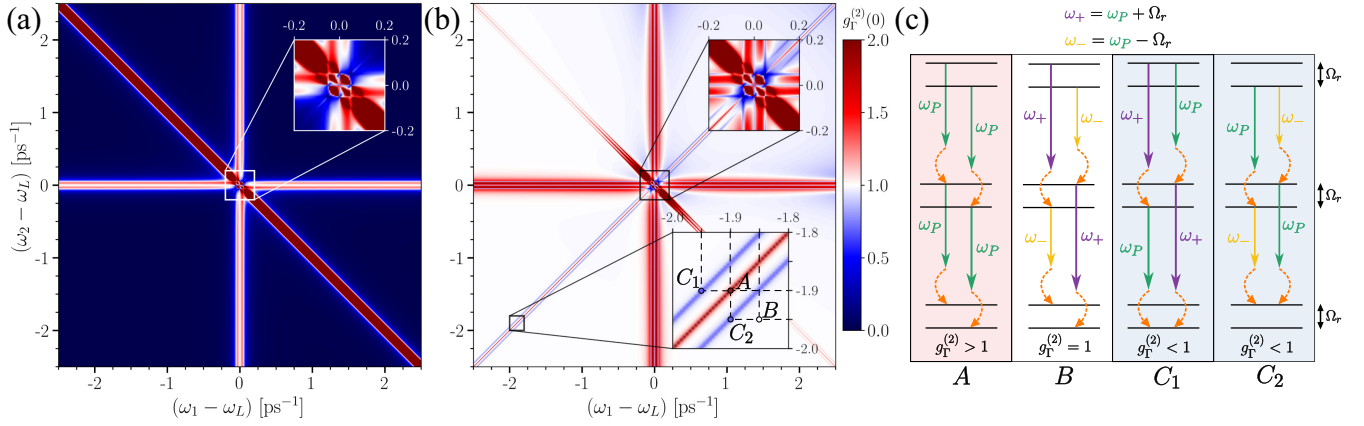


FIG. 3. Two-photon spectrum of a resonantly driven QD calculated (a) without and (b) with phonon interactions, where blue corresponds to antibunching, red to bunching, and white to uncorrelated emission. A new feature appears along the diagonal when phonon interactions are included, namely the additional three parallel lines shown in more detail in the lower inset of panel (b). The upper insets in (a) and (b) present magnifications over a smaller range of frequencies close to resonance. Panel (c) gives ladder diagrams in the driven-dot dressed-state basis showing all the possible photon cascades involving two phonons of the same energy (orange, dashed), where ω_P represents an arbitrary photon energy. The letters below the four boxes correspond to the annotated points in the lower inset of panel (b). All parameters are the same as in Fig. 2 apart from the sensor width, which is $\Gamma = 2\gamma$ for both sensors. The color bar scale only extends up to $g_r^{(2)}(0) = 2$ as this best showcases the phonon-induced triplet structure in the lower inset of (b). For completeness, Supplemental Material [54] includes plots with an extended logarithmic color bar scale.

driving strength is chosen well above saturation ($\Omega = 35\gamma$), fully resolving the Mollow triplet [55].

As shown, our method captures the PSB in quantitative agreement with the TEMPO benchmark, while the QRT completely fails. This resolves a long-standing issue: even in the Markovian weak-coupling regime, the PSB can be recovered once the QRT is avoided. The PSB originates from the Franck-Condon principle [27,38], where photon emission or absorption is accompanied by a lattice vibration. This energy-exchange process is clearly missed by the QRT but captured by our approach. The inset shows the expected phonon-induced renormalization of the Mollow side peaks [56,57], reproduced correctly by all methods for the parameters considered.

It is worth emphasizing that our approach remains conceptually and computationally simpler than TEMPO, which constructs the spectrum from an underlying tensor network, achieved through numerically intensive tensor contraction and compression steps (see Ref. [54] for further details). In contrast, for a two-level system, the single-photon spectrum is obtained in our method by diagonalizing a 16×16 matrix for each frequency. Moreover, this naturally extends to incorporating phonon effects in higher-order photon correlations by simply including additional sensors. To our knowledge, such calculations have not been performed previously, as evaluating the required multidimensional integrals is prohibitively difficult; neither state-of-the-art numerical methods such as TEMPO nor advanced analytical techniques can currently access these frequency-resolved higher-order correlations in the presence of vibrational environments.

To showcase this capability, we compute the normalized two-photon spectrum $g_{\Gamma_1, \Gamma_2}^{(2)}(\omega_1, \omega_2) = S_{\Gamma_1, \Gamma_2}^{(2)}(\omega_1, \omega_2) / [S_{\Gamma_1}^{(1)}(\omega_1)S_{\Gamma_2}^{(1)}(\omega_2)]$ [20–22] by including two sensors with variable frequencies ω_1 and ω_2 . Experimentally, this quantity can be measured in a coincidence setup using a Hanbury-Brown-Twiss arrangement with tunable spectral filters [58,59]. We choose equal sensor widths $\Gamma_1 = \Gamma_2 = \Gamma = 2\gamma$, so that the Mollow peaks remain well separated ($\Gamma \ll \Omega$) while each is fully resolved. Figures 3(a) and 3(b) show the calculated two-photon spectra without and with phonon interactions, respectively. In both panels red corresponds to bunching ($g_r^{(2)} > 1$), blue to antibunching ($g_r^{(2)} < 1$), and white to uncorrelated emission ($g_r^{(2)} = 1$). The frequency range extends beyond that typically considered [22–24,59], since phonon-induced features persist at large detunings. In fact, comparing the two spectra reveals clear signatures of phonon effects across all frequencies shown. For example, well below resonance, the background exhibits antibunching in the absence of phonon interactions [Fig. 3(a)], but becomes mostly uncorrelated with phonon interactions present [Fig. 3(b)]. Without phonons, emission of more than one photon is highly unlikely far from resonance, producing the apparent antibunching. With phonons, however, there is non-negligible intensity at the same frequencies, with no correlations expected between arbitrary parts of the PSB.

Despite this, when the sensors are tuned near each other but detuned from the emitter, a surprising triplet structure emerges in the presence of phonons, highlighted in the lower inset of Fig. 3(b). The three peaks are separated by

exactly the phonon-renormalized Rabi frequency [56,57], Ω_r , matching the side peak separations of the Mollow triplet (see Fig. 2 inset). In the absence of phonons, the two-photon correlations of the Mollow triplet at zero delay are well understood [18,22–24,47–49]. Photon pairs from the central peak are uncorrelated, as are pairs from the opposite side peaks (separated by twice the Rabi frequency). In contrast, photon pairs from the central peak and one side peak (separated by the Rabi frequency), or two from the same side peak, are antibunched.

Remarkably, our results show that signatures of this correlation structure persist within the phonon sideband, indicating that while phonons open additional emission pathways, they do not fundamentally alter the underlying second-order coherence of the Mollow triplet. Specifically, within the PSB, photon pairs differing in frequency by Ω_r appear antibunched, giving rise to blue diagonal lines in Fig. 3(b). Pairs separated by $2\Omega_r$ show no correlation (invisible against the white background), while identical-frequency pairs exhibit strong bunching (red diagonal line) as a consequence of frequency filtering causing the indistinguishability bunching effect [22], where $g_{\Gamma\Gamma}^{(2)}(\omega, \omega) \rightarrow 2$ for $\Gamma \rightarrow 0$, masking any other underlying correlations. Figure 3(c) maps out the corresponding two-photon cascades in the dressed-state basis [60], where each photon emission is accompanied by a fixed-energy phonon emission. Box A depicts identical-frequency pairs, bunched due to indistinguishability. Pairs differing by Ω_r (boxes C_1 and C_2) can occur in two time orderings that interfere destructively, producing antibunching. This interference stems from the finite detector time resolution ($\sim 1/\Gamma$), which introduces uncertainty in emission ordering [18,22,47,61]. Pairs differing by $2\Omega_r$ (box B) appear uncorrelated, consistent with the zero-delay behavior of opposite Mollow side peaks without phonons [18,22,47].

Conclusions—We have introduced a general framework for calculating frequency-resolved N -photon spectra in the presence of structured vibrational environments, overcoming the limitations of the quantum regression theorem. By embedding electron-phonon interactions directly into a weak-coupling master equation for a combined emitter-sensor system, we capture phonon-assisted transitions inaccessible to the QRT [11,13]. Applied to semiconductor quantum dots, our approach reproduces numerically exact results with high accuracy, including the phonon sideband. This shows that the failure of previous QRT-based treatments to account for sidebands stems from the QRT itself, not from the weak-coupling master equation. Leveraging the flexibility of the sensor formalism, we also compute higher-order correlations and, for the first time, investigate the phonon influence on the two-photon spectrum. We uncover a distinct diagonal triplet structure arising from phonon-assisted emission and show that hallmark correlations of the Mollow triplet persist even within the phonon sideband.

Although we have focused on a weak-coupling master equation, our formalism is agnostic to this choice. More advanced treatments, such as the polaron transformation [32,37], could be incorporated to explore stronger coupling regimes. Extending this approach to temporal correlations presents another promising direction. We anticipate that our predictions, particularly the phonon-assisted two-photon features, will motivate experimental efforts to observe and exploit these effects.

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Data availability—The data that support the findings of this article are openly available [62].

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