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Non-Markovian Model of Photon-Assisted Dephasing by Electron-Phonon Interactions in a Coupled Quantum-Dot–Cavity System

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We investigate the influence of electron-phonon interactions on the dynamical properties of a quantum-dot–cavity QED system. We show that non-Markovian effects in the phonon reservoir lead to strong changes in the dynamics, arising from photon-assisted dephasing processes, not present in Markovian treatments. A pronounced consequence is the emergence of a phonon induced spectral asymmetry when detuning the cavity from the quantum-dot resonance. The asymmetry can only be explained when considering the polaritonic quasiparticle nature of the quantum-dot–cavity system. Furthermore, a temperature induced reduction of the light-matter coupling strength is found to be relevant in interpreting experimental data, especially in the strong coupling regime.

The emergent field of quantum information technology [1] has spurred major research activities on controlling the fundamental interaction between a semiconductor quantum dot (QD) and a cavity. Solid-state cavity QED (cQED) systems are inherently coupled to the environment, since the emitter is embedded in a solid. This is in contrast to atomic cQED where the atom can be effectively isolated and only few discrete energy levels are sufficient in the description. Remarkably dephasing from solid-state environments cannot simply be seen as a nuisance, but can in fact lead to enhanced coupling of QDs to a detuned cavity mode of importance for efficient single-photon sources [2–4] and nanolasers [5]. Modeling the continuum of reservoir modes of solid-state systems constitutes a considerable challenge. The coupling of the QD-cavity system to its solid-state environment has almost exclusively been described using Markovian theories [2,6], neglecting memory effects of the reservoirs. While the Markovian approximation is well justified for some reservoirs, this is not in general true for the reservoir consisting of quantized lattice vibrations. Such phonon reservoirs dephase the QD-cavity system, whereby the entanglement between light and matter in general is destroyed. Notably the first experimental demonstrations of the strong coupling regime in solid-state cQED [3] revealed features in the emission spectra for large QD-cavity detuning that could not be explained by standard Markovian theory [6]. Since then there has been a lively debate [2,4,7] on the origin of the deviations. We demonstrate that non-Markovian phonon processes play an important role for solid-state cQED.

Here, using a simple physical model we show that photon-assisted dephasing processes are of great importance in describing the effect of phonons in a cQED setting. The underlying physical picture is that the polariton quasiparticle, formed by dressing the QD with the cavity photon, is dephased by phonon processes. We focus on the regime of relatively small QD-cavity detunings and pulsed excitation conditions where dephasing processes mediated by longitudinal acoustic (LA) phonons are expected to be important, and investigate the consequences on the dynamical properties of the cQED system. Pulsed excitation is required for on-demand photon sources emitting indistinguishable single photons and entangled-photon pairs [1], making it an important regime to investigate. Our theory takes into account memory effects of the phonon bath, which are neglected in the usual Markovian Lindblad theory of dephasing processes. The non-Markovian formulation is shown to be vital in interpreting recent experiments [7].

A similar non-Markovian theory has recently been used to describe the field dependent dephasing dynamics of classically driven two-level systems without a cavity [8]. By varying the strength of the applied classical field one can approach a regime where the QD dynamics takes place on a time scale that is near or even below the correlation time of the phonon reservoir, where the usual Markovian Lindblad theory of decay breaks down. In cQED the coupling is mediated by a single photon with a coupling strength g. State-of-the-art samples [7] have coupling strengths up to $\hbar g = 150 \ \mu$eV, translating to a characteristic time scale of about 14 ps, which is considerably longer than the phonon reservoir correlation time of typically 3–5 ps. Importantly, even for these realistic parameters non-Markovian effects are found to play an important role, giving rise to nontrivial detuning dependent dynamics and coupling strength. The QD–LA-phonon interaction and its effects on spectra and dynamics are well understood in the semiclassical regime [9], whereas effects due to

quantized light fields have not received much attention. Initial work on the influence of phonon dephasing on a cQED system has been reported [10]. Common to these works is that little physical insight into the non-Markovian nature of the dephasing processes has been given, which is a central theme of this Letter. We note that the physical processes identified here are also expected to be relevant to other cQED systems coupled to vibrational reservoirs, e.g., quantum wells, organic molecules, nitrogen vacancy centers in diamond, and colloidal QDs [11], implying that the ideas presented here may have wide applications.

We describe the effect of LA phonons on the QD-cavity system using the Jaynes-Cummings model with the addition of the electron–LA-phonon interaction [12,13]. The QD has an excited and a ground state of energies $\hbar\omega_e$ and $\hbar\omega_g$, respectively, while the cavity photon has energy $\hbar\omega_{cav}$. The QD-cavity system space is spanned by the two-level basis $\{|1\rangle = |e, n = 0\rangle, |2\rangle = |g, n = 1\rangle\}$, where n is the cavity occupancy, thus $|1\rangle$ describes the excited QD and $|2\rangle$ describes the excited cavity. The Hamiltonian has three terms, $H = H_s + H_i + H_{ph}$, described below. The Hamiltonian of the QD-cavity system is $H_s = \hbar\Delta\sigma_{11} + \hbar g(\sigma_{12} + \sigma_{21})$, where $\Delta = \omega_e - \omega_g - \omega_{cav}$ is the QD-cavity detuning and $\sigma_{nm} = |n\rangle\langle m|$. The electron-phonon interaction is $H_i = \sigma_{11}\sum_k M^k(b^\dagger_{-k} + b_k)$, where $M^k = M^k_{ee} - M^k_{gg}$ is the effective phonon interaction matrix element [14] and $b^\dagger_k$ creates a phonon in mode k. The free phonon Hamiltonian is $H_{ph} = \sum_k \hbar\omega_k b^\dagger_k b_k$, where $\omega_k = c_s k$ is the phonon dispersion and c_s is the speed of sound.

We apply the time-convolutionless approach [15] for describing the reduced dynamics of the QD-cavity system. The density matrix of the QD-cavity system, $\rho(t)$, is considered to second order in H_i, and assuming that the phonon bath remains in a thermal state. The equations for $s_{pq}(t) = \text{Tr}\{\rho(t)\sigma_{pq}\}$ (representing the excited QD state population, $s_{11}(t)$, the number of photons in the cavity, $s_{22}(t)$, and the photon-assisted polarization, $s_{12}(t)$) are

$$\partial_t s_{11}(t) = -ig[s_{12}(t) - s^*_{12}(t)] - \Gamma s_{11}(t), \quad (1a)$$

$$\partial_t s_{22}(t) = ig[s_{12}(t) - s^*_{12}(t)] - \kappa s_{22}(t), \quad (1b)$$

$$\partial_t s_{12}(t) = i\Delta s_{12}(t) - ig[s_{11}(t) - s_{22}(t)]$$
$$- \frac{1}{2}(\Gamma + \kappa)s_{12}(t) + \partial_t s_{12}(t)|_{ph}. \quad (1c)$$

Without the phonon induced terms represented by $\partial_t s_{12}(t)|_{ph}$ these equations are the standard lossy Jaynes-Cummings model. The losses have been introduced through the Lindblad formalism [15]. These include decay, described by a rate Γ, of the excited QD state to modes other than the cavity and nonradiative channels, and the finite linewidth of the cavity, $\kappa = \omega_{cav}/Q$, where Q is the usual quality factor of the cavity. We take $\Gamma = 1$ ns^{-1} in all simulations to be presented. The phonon induced terms in Eq. (1c) are

$$\partial_t s_{12}(t)|_{ph} = -[\gamma_{12}(t) - i\Delta_{pol}]s_{12}(t) + iG^<(t)s_{22}(t)$$
$$- iG^>(t)s_{11}(t). \quad (2)$$

This term introduces two novel effects compared to standard Markovian cQED models [2]. First, $\gamma_{12}(t)$ enters as a time-dependent pure dephasing rate. Second, the functions $G^{\gtrless}(t)$ renormalize the bare coupling strength g: the effective value of g is changed by the real part of $G^{\gtrless}(t)$, and an additional decay of the polarization is induced by the imaginary part of $G^{\gtrless}(t)$. The long-time polaron shift $\Delta_{pol} = \text{Im}\{\gamma_{12}(\infty)\}$ has been subtracted from $\gamma_{12}(t)$ [15]. Explicitly,

$$G^{\gtrless}(t) = i\hbar^{-2}\int_0^t dt' U^*_{11}(t')U_{21}(t')D^{\gtrless}(t'), \quad (3)$$

$$\gamma_{12}(t) = \hbar^{-2}\int_0^t dt'[|U_{11}(t')|^2 D^<(t') - |U_{21}(t')|^2 D^>(t')]. \quad (4)$$

The phonon bath correlation functions are

$$D^{\gtrless}(t) = \sum_k |M^k|^2[n_k e^{\pm i\omega_k t} + (n_k + 1)e^{\mp i\omega_k t}], \quad (5)$$

with $n_k = 1/[\exp(\hbar\omega_k/k_B T) - 1]$. The operator $U(t) = \exp(-iH_s t/\hbar)$ is the time evolution operator for the QD-cavity system and is the essential ingredient as it introduces the photon-dressed QD into the phonon scattering terms. The Markovian Lindblad formalism is obtained neglecting all memory effects associated with the phonon interaction, i.e. $D^{\gtrless}(t) \propto \delta(t)$ [15]. In this limit $G^{\gtrless}(t) = 0$ and $\gamma_{12}(t) = $ constant, with no dependence on photon properties.

We have numerically solved Eq. (1) with the initial condition of a single excitation on the QD, $s_{11}(0) = 1$, and all other elements set to zero, modeling an experiment where a QD is excited in its discrete states with a short optical pulse [16]. The parameters are chosen similar to recent experiments [7]. The computed excited state populations, $s_{11}(t)$, are shown in Fig. 1 for various detuning values. For $\Delta = 0$ we observe an expected strong enhancement of the decay rate and associated Rabi oscillations, indicating the strong coupling regime. For nonzero detun-
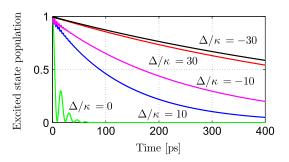

FIG. 1 (color online). Evolution of the excited state population $s_{11}(t)$ for $\hbar g = 140$ μeV, $\hbar\kappa = 100$ μeV, $Q \approx 10^4$, and $T = 4$ K.

ing we observe small Rabi oscillations in the start of the decay curve, again indicative of the strong coupling regime. Interestingly, our theory predicts a shorter lifetime when the cavity is tuned below the QD resonance than when it is tuned above, in stark contrast with standard Markovian theory [17]. The asymmetry is particularly strong for the $\Delta/\kappa = \pm 10$ cases and has recently been observed in experiments [7]. We attribute this pronounced reduction in lifetime to a phonon-assisted Purcell effect, where the QD may couple via phonon emission to the cavity when the cavity is spectrally below it. In the opposite case, where the cavity is spectrally above the QD resonance, phonon absorption is needed for the QD to become resonant with the cavity. This process is suppressed at low temperatures and the phonon mediated coupling between QD and cavity is lost. The asymmetry becomes less pronounced when the detuning is larger than the energy the electron can lose through phonon emission, as seen in the $\Delta/\kappa = \pm 30$ cases. The importance of using the photon-assisted electron-phonon interaction, [i.e., the operator $U(t)$ in Eqs. (3) and (4)], should be emphasized. Setting $g = 0$ only in $U(t)$ we obtain results in quantitative agreement with the $\Delta < 0$ results in Fig. 1, where the phonons do not significantly influence the dynamics, for all values and signs of Δ. Formally this is easily understood, as in this case $\mathcal{G}^{\gtrless}(t) = 0$ and $\gamma_{12}(t)$ loses its dependence on g and Δ, thus the asymmetry is lost in the phonon induced dephasing. One can interpret this approximation as only allowing the phonons to interact with the bare electron and not the electron-photon quasiparticle, the polariton, that is actually present in the system. This illustrates the importance of accounting for the polaritonic quasiparticle nature of the strongly coupled QD cavity and its non-Markovian interaction with the phonon reservoir.

Figure 2 shows the long-time limit [18] of the rates, Eqs. (3) and (4). A strongly asymmetric $\mathcal{G}^{\gtrless}(\infty)$ is apparent versus detuning. Also, $\mathcal{G}^{>}(\infty)$ and $\mathcal{G}^{<}(\infty)$ attain very different values for fixed detuning. As the pure dephasing rate $\gamma_{12}(\infty)$ remains symmetric with respect to detuning,
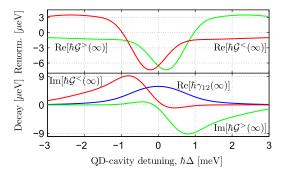
the asymmetry in Fig. 1 is caused by the renormalization rates $\mathcal{G}^{\gtrless}(\infty)$. It is remarkable that despite of their relative weakness, $|\mathcal{G}^{\gtrless}(\infty)|_{\max}/g < 7\%$, such large dynamical effects may occur. The physical origin of this asymmetry can be traced back to the phonon correlation functions given in Eq. (5). Here it is seen that phonon absorption processes are suppressed at low temperatures as they are proportional to the occupation factor n_k, whereas phonon emission processes continue to be possible due to the presence of the phonon vacuum field. In linear QD absorption spectra virtual phonon emission results in asymmetric sidebands centered around an infinitely sharp zero phonon line (ZPL) [19]. However, for the present physical system it is essential that the cavity field is treated nonlinearly. This results in novel effects such as a finite width of the ZPL and the coupling strength renormalization rates $\mathcal{G}^{\gtrless}(t)$.

We next examine temperature effects at zero detuning. For this case: $\mathcal{G}^{\gtrless}(t, \Delta = 0) = \mathcal{G}_R(t) \mp i\mathcal{G}_I(t)$, where $\mathcal{G}_R(t)$ and $\mathcal{G}_I(t)$ are real functions. Figure 3 shows the effect of temperature on the phonon induced rates $\mathcal{G}^{\gtrless}(\infty)$ and $\gamma_{12}(\infty)$, within the low temperature regime typically explored in cQED experiments. As expected, the pure dephasing rate increases with temperature. We also observe an increase as a function of the bare coupling strength g. $\mathcal{G}_R(\infty)$ also increases in magnitude with increasing temperature, but has a negative value. This leads to a lowering of the effective coupling strength entering the equation for $s_{12}(t)$ as temperature is increased. This can be realized by inserting Eq. (2) and $\mathcal{G}^{\gtrless}(\infty, \Delta = 0)$ into Eq. (1c) yielding $g_{\text{eff}}(\infty) = g - |\mathcal{G}_R(\infty)| < g$. The effective coupling strength thus depends significantly on temperature. This mechanism is also relevant for interpreting cQED absorption spectra [13]. We expect this dependence to have a detrimental effect on the possibility of reaching the strong coupling regime. To quantify this prediction we have investigated the transition between the weak and strong coupling regime, while varying the most important parameters in the model, namely g, κ, and T. We define the weak coupling regime as the situation where the initially populated excited state of the QD decays monotonically toward zero. Figure 4 shows the results both for the full
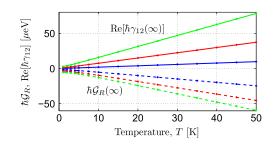
FIG. 2 (color online). $t \to \infty$ limit of $\hbar\gamma_{12}(t)$ (blue [dark gray]) and $\hbar\mathcal{G}^{\gtrless}(t)$ (green [light gray] or red [medium gray]) for the parameters $\hbar g = 140$ μeV and $T = 4$ K. Im$[\hbar\gamma_{12}(\infty)]$ has the constant value 32.7 μeV.

FIG. 3 (color online).   Real part of $\hbar\gamma_{12}(\infty)$ (solid) and $\hbar\mathcal{G}_R(\infty)$ (dashed) for $\hbar g = 50$ (blue [dark gray]), 100 (red [medium gray]), 150 (green [light gray]) μeV. The imaginary parts are independent of temperature and much smaller than the real part and therefore not shown.
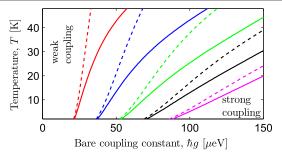
FIG. 4 (color online). Parameter space showing the presence of strong or weak coupling for the full model (solid line) and for $\mathcal{G}^{\gtrless}(t) = 0$ (dashed line), with $\hbar\kappa = 75$ (red [medium gray]), 125 (blue [dark gray]), 175 (green [light gray]), 225 (black), 275 (magenta [light-medium gray]) μeV.

model and for comparison the case where we have artificially put $\mathcal{G}^{\gtrless}(t) = 0$. This is done to emphasize the effect of the temperature induced renormalization of g, by allowing only the pure dephasing rate to be temperature dependent, as is common practice in phenomenological cQED models. As expected, we generally observe the presence of strong coupling in the system for large g and low T, with the parameter space of strong coupling becoming extended as we increase the quality of the cavity. Comparing the results of the full model with those where $\mathcal{G}^{\gtrless}(t) = 0$, we notice a strong effect of the renormalization of the bare coupling constant g. The parameter space where strong coupling is obtained is significantly decreased when including the renormalization of g. This result is relevant in the interpretation of experimental data, as state-of-the-art cQED models [2] do not include the renormalization effects contained in the functions $\mathcal{G}^{\gtrless}(t)$. These effects are of significant importance and therefore cQED models neglecting them can lead to misinterpretation of experimental data.

In conclusion, we have illustrated the importance of applying a dressed-state picture of the polaritonic QD-cavity system when modeling cQED systems interacting with LA phonons. We have shown its relation to recent experiments, explaining the observed asymmetry in lifetimes with respect to the QD-cavity detuning. The asymmetry can only be understood by treating the QD-cavity system as a polaritonic quasiparticle in the phonon induced scattering terms. Furthermore we have investigated a phonon induced lowering of the effective coupling strength with increasing temperature, which was found to change the criterion for strong coupling significantly.

Note added.—Recently, we became aware of related work [20], however, with a focus on modeling experimental emission spectra as opposed to interpretation of the mechanism responsible for the dynamics, which is presented here.

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