Quantum dot cavity-QED in the presence of strong electron-phonon interactions

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(Received 23 January 2002; published 28 May 2002)

A quantum dot strongly coupled to a single high-finesse optical microcavity mode constitutes a new fundamental system for quantum optics. Here, the effect of exciton-phonon interactions on reversible quantum dot cavity coupling is analyzed without making a Born-Markov approximation. The analysis is based on a polaron operator technique that has been used to study the "spin-boson" Hamiltonian. For bulk acoustic phonons and for a large class of confined phonon models, we find that vacuum-Rabi splitting persists even in the presence of a large Stokes shift and at an appreciable temperature, but its magnitude is exponentially suppressed by the electron-phonon coupling strength.

DOI: 10.1103/PhysRevB.65.235311 PACS number(s): 42.50.Dv, 03.67.Lx

The fundamental system in cavity quantum electrodynamics (cavity-QED) is a two-level atom interacting with a single-cavity mode.[1] If the electric field per photon inside the cavity is sufficiently large, then the single-photon dipole coupling strength (g) between the atom and the cavity mode can exceed the decoherence rates in the system due to cavity losses and dipole dephasing: this corresponds to the strong-coupling regime of cavity-QED whose principal signature is the vacuum-Rabi oscillations.[2] Cavity-QED in the strong-coupling regime has proved to be an invaluable tool in investigating and understanding quantum phenomena.[3] One of the principal applications of cavity-QED techniques has been in the emerging field of quantum information (QI) science: a significant fraction of quantum computation and communication schemes rely on the strong-coupling regime of cavity-QED.[4] Recent developments in semiconductor nanotechnology have shown that excitons in quantum dots (QD's) constitute an alternative two-level system for cavity-QED applications.[5] In contrast to single-atom cavity-QED where reservoir couplings are weak and can be treated using standard quantum optics techniques,[1] the physics of the QD-microcavity system is enriched by the presence of strong electron-electron and electron-phonon interactions. For example, QD absorption and emission spectra in some cases exhibit sidebands and/or appreciable Stokes shifts.[6–8] These features signal nonperturbative electron-phonon interactions[9] and indicate that decoherence due to phonons could present a fundamental limitation to QI processing based on quantum dot cavity-QED.[10]

In this paper, we analyze the effects of electron-phonon interactions on strong electron-hole-photon coupling in cavity-QED, without applying the Born-Markov approximation to the electron-phonon interaction. We consider spectral density functions $J(\omega)$ following a cubic law near $\omega=0$, which characterize for instance the interaction of excitons with bulk acoustic phonons. To obtain the absorption spectrum, we apply a polaron operator technique that has been used to study the "spin-boson" problem.[12] This approach takes into account the relevant multiphonon processes to all orders and the exact solution is recovered when either the electron-phonon or the electron-photon coupling tend to zero. We find that the strong-coupling regime persists even in the presence of a large Stokes shift and at an appreciable temperature ($T>g$). However, the Rabi frequency is exponentially suppressed by the electron-phonon coupling strength. Finally we show that these results also hold for a large class of confined phonon models.

We focus on understanding the dependence of vacuum-Rabi oscillations on electron-phonon interactions. We will therefore assume a simple two-level model for the electronic degrees of freedom of the QD, consisting of the QD electronic ground state $|g\rangle$ and the lowest-energy electron-hole (exciton) state $|e\rangle$. The starting point of our analysis is the Hamiltonian

$$\begin{aligned} H=&\hbar\omega_{eg}\sigma_{ee}+\hbar\omega_c a^\dagger a+\hbar g(\sigma_{eg}a+a^\dagger\sigma_{ge})\\ &+\sigma_{ee}\sum_k \hbar\lambda_k(b_k+b_k^\dagger)+\sum_k \hbar\omega_k b_k^\dagger b_k\\ &+\hbar\Omega_p(\sigma_{eg}e^{-i\omega t}+\sigma_{ge}e^{i\omega t}), \end{aligned} \tag{1}$$

where $\sigma_{eg}=|e\rangle\langle g|$, a and b_k are annihilation operators for the cavity mode, and the kth phonon mode, and Ω_p a weak classical external probe that will be used for obtaining the spectra. We will also allow for Markovian processes, not included in H, to take into account cavity losses (γ_c) and homogeneous Lorentzian broadening of the QD exciton state (γ_{QD}). The validity of this model relies on the adiabatic approximation[13] and the assumption that off-diagonal electron-phonon interaction terms coupling $|e\rangle$ to exciton excited states are sufficiently weak.[14] This assumption is well justified for quantum dots where the energy separation between these states is greater than 20 meV, when the temperature is low enough ($T<50$ K). With these approximations, we can describe the coupled QD-cavity system weakly excited by Ω_p as a three-state system defined by projection of the QD-cavity Hilbert space on the subspace spanned by $|g,n_c=0\rangle$ ($|0\rangle$), $|g,n_c=1\rangle$ ($|1\rangle$), and $|e,n_c=0\rangle$ ($|2\rangle$), where n_c denotes the cavity-mode occupancy . Finally, we transform the Hamiltonian to a rotating frame at the probe frequency.

It is instructive to point out the relations between Hamiltonian (1) and some well-studied problems as depicted in Fig. 1. The first three terms in Hamiltonian (1) give the

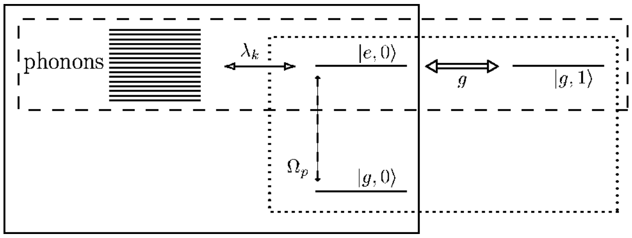

FIG. 1. Level diagram for our quantum dot cavity-QED model depicting its relations with three well studied problems: Jaynes-Cummings model (dotted box), "spin-boson" problem (dashed box), and independent boson model (solid box).

Jaynes-Cummings model in the rotating-wave approximation.[1] If we project the QD-cavity system on any of the $N>0$ manifolds of this model and include the fourth and fifth terms, we obtain up to an irrelevant constant a "spin boson" Hamiltonian.[11] Finally, if we take Hamiltonian (1) and drop the terms involving the cavity, we get the independent boson model[13,15] which has already been used to study phonon effects in small quantum dots.[6–8]

A convenient representation for considering super-ohmic electron-phonon coupling of arbitrary strength is obtained by applying a canonical transformation that exactly diagonalizes the independent boson model:[11–13,15]

$$A' = e^{s} A e^{-s} \quad \text{with} \quad s = \sigma_{ee} \sum_k \frac{\lambda_k}{\omega_k} (b_k^\dagger - b_k). \tag{2}$$

The transformed Hamiltonian reads

$$H' = H'_{sys} + H'_{int} + H'_{bath}$$

with

$$H'_{bath} = \sum_k \omega_k b_k^\dagger b_k ,$$

$$H'_{sys} = \hbar \omega \sigma_{00} + \hbar \omega_c \sigma_{11} + \hbar(\omega_{eg} - \Delta)\sigma_{22} + \langle B \rangle X_g ,$$

$$H'_{int} = X_g \xi_g + X_u \xi_u . \tag{3}$$

Here we have defined the operators

$$X_g = \hbar[g(\sigma_{21} + \sigma_{12}) + \Omega_p(\sigma_{20} + \sigma_{02})],$$

$$X_u = i\hbar[g(\sigma_{12} - \sigma_{21}) + \Omega_p(\sigma_{02} - \sigma_{20})],$$

$$B_\pm = \exp\left(\pm \sum_k \frac{\lambda_k}{\omega_k}(b_k - b_k^\dagger) \right),$$

$$\xi_g = \frac{1}{2}(B_+ + B_- - 2\langle B \rangle),$$

$$\xi_u = \frac{1}{2i}(B_+ - B_-); \tag{4}$$

the mean value $\langle B \rangle = \langle B_+ \rangle = \langle B_- \rangle$ and the polaron shift $\Delta = \Sigma_k \lambda_k^2 / \omega_k$.[15] We consider initial conditions that are factorizable in this new representation ($\rho_{tot} = \rho_{sys} \otimes \rho_{bath}$, with ρ_{bath} a thermal state). The Hamiltonian H'_{sys} includes the coherent contributions of the new interaction terms. In this polaron representation we apply a second-order Born approximation in the residual exciton-photon-phonon coupling H'_{int} (Ref. 12) and trace over the phonon degrees of freedom to obtain an operator master equation for the reduced density matrix $[\rho(t)]$ of the QD-cavity system:

$$\frac{\partial \rho(t)}{\partial t} = \frac{1}{i\hbar}[H'_{sys}, \rho(t)] - \frac{1}{\hbar^2} \int_0^t d\tau \sum_{m=\{g,u\}} \{ G_m(\tau) \times [X_m, e^{-iH'_{sys}\tau/\hbar} X_m \rho(t-\tau) e^{iH'_{sys}\tau/\hbar}] + \text{H.c.} \}, \tag{5}$$

with $G_{g/u}(t) = \langle \xi_{g/u}(t) \xi_{g/u}(0) \rangle$. These polaron Green's functions are given by[11–13,15]

$$G_g(t) = \langle B \rangle^2 \{ \cosh[\varphi(t)] - 1 \}, \quad G_u(t) = \langle B \rangle^2 \sinh[\varphi(t)],$$

with

$$\varphi(t) = \int_0^\infty d\omega \frac{J(\omega)}{\omega^2} [\coth(\beta\hbar\omega/2)\cos(\omega t) - i \sin(\omega t)]. \tag{6a}$$

While $\langle B \rangle$ and Δ can be expressed, respectively, as

$$\langle B \rangle = \exp\left(-\frac{1}{2} \int_0^\infty d\omega J(\omega) \coth(\beta\hbar\omega/2)/\omega^2 \right),$$

$$\Delta = \int_0^\infty d\omega J(\omega)/\omega. \tag{6b}$$

Here we have introduced the following spectral function:

$$J(\omega) = \sum_k \lambda_k^2 \delta(\omega - \omega_k). \tag{7}$$

The polaron shift Δ gives a rough approximation to the Stokes shift (i.e., difference between absorption and emission peaks) and provides us with a convenient measure of the strength of the exciton-phonon interaction.[13,15]

To obtain the absorption spectrum we consider the initial condition $\rho(0) = \sigma_{00}$ and treat the probe Ω_p to lowest order in perturbation theory. We focus on the case where the cavity is on resonance with the zero-phonon line (ZPL) (Refs. 13 and 15) of the QD ($\omega_{eg} - \Delta = \omega_c$). It proves convenient to project Eq. (5) on the basis of eigenstates of $H'_{sys}|_{\Omega_p=0}$: $|0\rangle$, $|+\rangle = (1/\sqrt{2})(|1\rangle + |2\rangle)$ and $|-\rangle = (1/\sqrt{2})(|1\rangle - |2\rangle)$. We expand ρ in powers of Ω_p and let $\rho^{(j)}_{\eta\eta'} = \langle \eta | \rho^{(j)} | \eta' \rangle$ be the corresponding jth-order coefficients. We define $\Delta\omega_\pm = \omega - \omega_c \mp g\langle B \rangle$ and $G_\pm(t) = G_g(t) e^{i\Delta\omega_\pm t} + G_u(t) e^{i\Delta\omega_\mp t}$. It is useful to note that to zeroth order in Ω_p we have $\rho(t) = \sigma_{00}$ at all times and that the products $X_m \rho(t)$ are at least first order in Ω_p. Then it is straightforward to obtain the following set of equations:

$$\frac{\partial \rho^{(1)}_{\pm 0}(t)}{\partial t} = \frac{\mp i \langle B \rangle}{\sqrt{2}} + i \Delta\omega_\pm \rho^{(1)}_{\pm 0}(t) - \frac{g}{\sqrt{2}} \int_0^t d\tau G_\pm(\tau) \times [1 + g\sqrt{2} \rho^{(1)}_{\pm 0}(t-\tau)] \tag{8}$$

and

$$\frac{\partial \rho_{00}^{(2)}(t)}{\partial t} = \sum_{\eta=\{+,-\}} \frac{-\eta i \langle B\rangle}{\sqrt{2}} \rho_{\eta 0}^{(1)}(t) - \frac{1}{2}\int_0^t d\tau G_\eta(\tau)[1+g\sqrt{2}\rho_{\eta 0}^{(1)}(t-\tau)] + \text{c.c.} \tag{9}$$

The absorption spectrum can then be obtained from the asymptotic behavior of the rate of change of the ground-state population $[\lim_{t\to\infty}\partial\rho_{00}^{(2)}(t)/\partial t]$. A simple way to calculate the latter is to Laplace transform Eqs. (8) and (9), and use the relation $\lim_{t\to\infty} f(t) = \lim_{s\to 0} s\hat{f}(s)$. To include the Markovian processes associated with γ_c and γ_{QD}, we modify H'_{sys} to include a non-Hermitian contribution $-i(\gamma_c/2)\sigma_{11} - i(\gamma_{QD}/2)\sigma_{22}$. This is an incomplete description of the dynamics of the system but it is well known that it leads to the correct expression for the absorption spectrum for the standard Jaynes-Cummings model and it can be shown that this fact is not modified by the introduction of exciton-phonon interactions [for example by resorting to a stochastic wave function (MCWF) point of view[16]]. We take $\gamma_c = \gamma_{QD} = \gamma$ and set the frequency origin at ω_c. In this way we obtain

$$A(\omega) = \sum_{\eta=\{+,-\}} \frac{G''_\eta(\omega)\left[\omega^2 + \frac{\gamma}{2}g^2 G''_\eta(\omega) + \frac{\gamma^2}{4}\right] + \frac{\gamma}{2}[\eta\langle B\rangle + gG'_\eta(\omega)]^2}{[\Delta\omega_\eta - g^2 G'_\eta(\omega)]^2 + \left[\frac{\gamma}{2} + g^2 G''_\eta(\omega)\right]^2} \tag{10}$$

for the absorption spectrum, up to an irrelevant prefactor. The dissipative parts $G''_\eta(\omega)$ and the reactive parts $G'_\eta(\omega)$ are given, respectively, by the real and imaginary parts of $\int_0^\infty dt e^{-\gamma t} G_\eta(t)$.

In physical terms the second-order Born approximation we use in deriving Eq. (5) is valid when the energy exchange between the QD and the cavity mode does not have an appreciable effect on the statistical properties of the phonons. Würger has calculated the lowest-order corrections to this approximation for the "spin-boson" problem with a $J(\omega)$ similar to Eq. (11) (presented below) and found them to be negligible provided $g \ll \omega_b$ (where ω_b is the cutoff frequency).[12] For more general spectral functions this latter condition should be generalized to $g \ll \delta_{ph}$, where $\hbar\delta_{ph}$ is the smallest characteristic energy of $J(\omega)$.[11] This condition is satisfied by the spectral functions we consider. Our three-state model differs from the "spin-boson" problem only in the perturbative coupling (Ω_p) to the state $|0\rangle$ which should not alter these considerations. In addition, we focus on the parameter regime defined by $\Delta \lesssim \omega_b$ and $T \ll \omega_b$.

A useful approximation to the spectrum (10) is given by replacing each of the terms $\eta = \{+,-\}$ with the Lorentzian obtained by evaluating the numerator and the function $G''_\eta(\omega)$ at $\omega = \eta\tilde{g} = \eta g\langle B\rangle$ and dropping all contributions of the reactive parts. This treatment is analogous to the pole approximation used in Ref. 12 and is valid for underdamped regimes and frequencies in the neighborhood of the ZPL where the vacuum-Rabi splitting (VRS) is present. Each of these Lorentzians corresponds, respectively, to the lower $(-)$ upper $(+)$ dressed state of the QD-cavity system. We can therefore deduce that when the system presents weakly damped oscillations, good approximations to the splitting, to the compound relative oscillator strength of the two peaks and to the broadenings are given, respectively, by $2g\langle B\rangle$, $\langle B\rangle^2$, and $\gamma + 2g^2 G''_\pm(\pm\tilde{g})$. If we take this formula for the splitting $(2g\langle B\rangle)$ and substitute in it expression (6b) for $\langle B\rangle$, we realize that the Rabi frequency is exponentially suppressed by the electron-phonon coupling strength. An interesting consequence of the g dependence of the broadenings is that the Q value of the corresponding peaks can be degraded if g is increased.

The limit $T\to 0$, $\gamma\to 0$ of the spectrum (10) is of special interest. At zero temperature there are no phonons to be absorbed and so the polaron absorption spectrum [i.e., $A(\omega)$ specialized to $g=0$, $\gamma=0$] is always identically zero for frequencies below the ZPL $(\omega<0)$.[13,15] This implies that in this limit there is a lowest-energy resonance close to $\omega = -\tilde{g}$ with infinite lifetime, for all $J(\omega)$. For underdamped regimes at sufficiently low T and small γ this leads to an asymmetry between the widths of the two resonances (Fig. 2). At $T=0$ the rate $g^2 G''_+(\tilde{g})$ is dominated by one phonon processes.[11,12] It can be shown that under these circumstances if $\gamma=0$ there is VRS for all $\Delta \lesssim \omega_b$.

To study QD coupling to bulk acoustic phonons we use the following spectral function:

$$J(\omega) \propto \frac{\omega^3}{\left(\frac{\omega}{\omega_b}+1\right)^2\left[\left(\frac{\omega}{\omega_b}-1\right)^2+1\right]}. \tag{11}$$

The cubic dependence on ω for $\omega\to 0$ can be deduced for both deformation potential and piezoelectric coupling[14] for a generic QD. The form factor of the exciton provides a natural cutoff at a frequency ω_b set by the size of the dot. The particular frequency dependence of this cutoff is irrelevant for the study of VRS and has been chosen for computational convenience. For systems of interest one would expect: $g \lesssim 0.1$ meV (Ref. 10) and $\omega_b \sim 1$ meV (Ref. 6). Figure 2 shows typical spectra for this model with the cavity $(g\neq 0)$ and without it $(g=0)$. The persistence of VRS for appre-

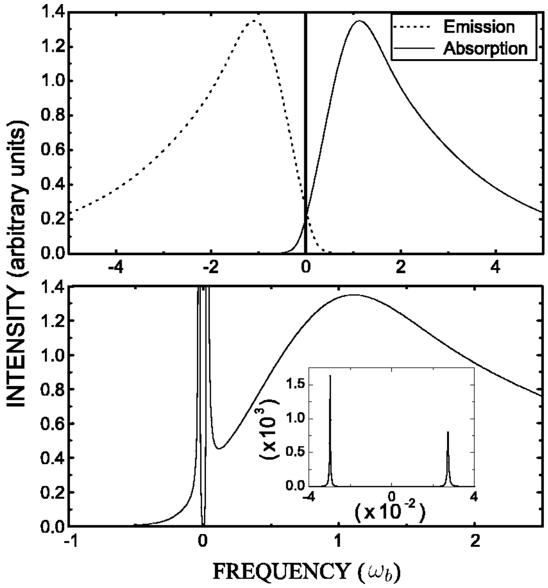

FIG. 2. Calculated spectra for a bulk acoustic phonon model with $\Delta/\omega_b=2$, $T/\omega_b=0.1$, and $\gamma=0$. The frequency origin is set at the zero-phonon line (ZPL). Top shows the polaron spectra ($g=0$) and bottom the QD-cavity absorption spectrum for $g/\omega_b=5\times10^{-2}$. The ZPL of the polaron spectra is given by a δ function with relative oscillator strength $\langle B\rangle^2=0.325$. The inset expands a neighborhood of $\omega=0$ (the asymmetry between the two maxima is explained by the difference between the broadenings).

ciable Stokes shift and temperature ($T>g$) that we find is consistent with results for the "spin-boson" problem.[11,12]

In many of the QD systems of interest strong electron-phonon interactions are associated with phonon confinement.[7] To model these cases we consider coupling to a single-phonon mode at ω_b which is itself broadened by a weak linear (coordinate-coordinate) interaction with a short memory reservoir characterized by a spectral density $\tilde{J}(\omega)$ with ultraviolet cutoff ω_*. As the interaction term is linear there exists an exact transformation to a new set of modes in which the Hamiltonian assumes once again the form given in Eq. (1) and $J(\omega)$ is given as a function of $\tilde{J}(\omega)$.[17] The resulting $J(\omega)$ inherits its ohmic $[J(\omega)\propto\omega]$ or super-ohmic character $[J(\omega)\propto\omega^n, n>1]$ from $\tilde{J}(\omega)$ $[\tilde{J}(\omega)\propto\omega^n]$. Assuming $\tilde{J}(\omega_b)\ll\omega_b$ and $\omega_b\ll\omega_*$ we deduce the following approximate expression for $J(\omega)$:

$$J(\omega)\propto\frac{\tilde{J}(\omega)}{\left(\frac{\omega}{\omega_b}+1\right)^2[(\omega-\omega_b)^2+\pi^2\tilde{J}^2(\omega_b)]}. \quad (12)$$

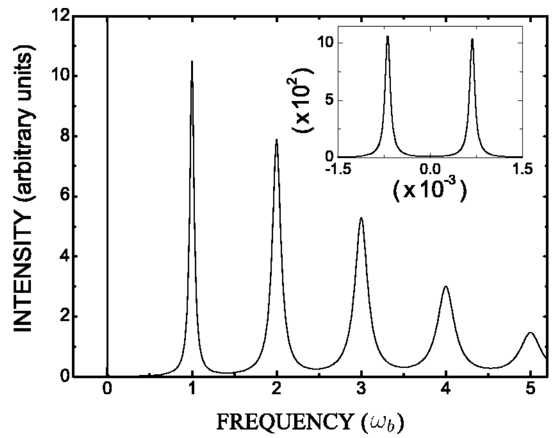

FIG. 3. Calculated QD-cavity absorption spectrum for a confined phonon model with $n=3$, $T/\omega_b=5\times10^{-2}$, $\Delta/\omega_b=3$, $\gamma/\omega_b=10^{-4}$, $\tilde{J}(\omega_b)/\omega_b=3/\pi\times10^{-2}$, and $g/\omega_b=3\times10^{-3}$. The frequency origin is set at the zero-phonon line, and its neighborhood is expanded in the inset.

For this spectral function (12) we have $\delta_{ph}=\tilde{J}(\omega_b)$. We have considered this model for $n=3$ with $g\ll\tilde{J}(\omega_b)$. However, our results should be qualitatively correct for all $n\geqslant3$.[11,12] For confined acoustic phonons ω_b will be again determined by the QD size. Figure 3 shows a representative spectrum for this model exhibiting well-defined sidebands. We find the same results for the dependence of VRS on electron-phonon interactions as in the bulk case.

In summary, we have presented a formalism that allows us to analyze basic cavity-QED effects for QD-microcavity systems with strong exciton-phonon interactions. We find that for super-Ohmic spectral functions the principal role of exciton-phonon interactions is the suppression of the effective QD-cavity coupling strength. A natural extension of this work is the analysis of ohmic spectral functions, which will be presented elsewhere.

Finally, we remark that the analysis presented here would be of interest to all zero-dimensional solid-state-based quantum optical systems, where coupling to lattice vibrations can be relevant. This for example is the case for nitrogen vacancy color centers in diamond nanocrystals coupled to high-Q optical cavities, which could be operated as efficient single-photon sources.[18]

We thank D. Loss for bringing to our attention Ref. 12. I. W. would also like to thank B. Gayral for helpful discussions. This work was supported by the David and Lucile Packard Foundation.

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