Antibunching of thermal radiation by a room-temperature phonon bath: an exact solution of a strongly interacting light-matter-reservoir system

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Progress in semiconductor technology introduces a new platform for quantum optics studies in solid state: a quantum dot strongly coupled to a cavity mode. We present an exactly solvable model for the combined electron, photon and phonon dynamics. For a cavity mode prepared in a Fock state, the model reproduces the Jaynes-Cumming solution and interaction with a phonon bath leads higher \( g^{(2)}(0) \). In contrast, for an initial thermal photon distribution, the phonon bath interaction gives a counter-intuitive reduction in \( g^{(2)}(0) \), resulting the classical photon distribution evolving into a nonclassical one.

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Successes in semiconductor nano- and micro-structure fabrication are paving the way for new explorations in quantum optics.[1, 2] Enabling these studies is the capability for placing nanostructures, such as quantum dots, inside high finesse microcavities. Such experimental structures are also attractive for engineering applications, including single-photon and entangled-photon production.[2, 3] Further novel ideas and applications will emerge as we better understand underlying physics, e.g. the quantum many-body physics.

Recent work has incorporated several aspects of many-body phenomena into semiconductor quantum-dot cavity-quantum-electrodynamics (QD-CQED) calculations. Absorption spectra in the presence of electron-phonon interactions via polaron operator technique were derived.[4–6] Exciton dynamics in the presence of LA-phonons was investigated using a perturbation approach based on the Lamb-Dicke approximation.[7] In the many-photon and many-emitter limit, robust resonances of vacuum Rabi splitting were predicted,[8] in agreement with semiconductor microcavity laser experiments.[9] In this paper, we deviate from previous perturbative treatments and present an exactly solvable approach for QD-CQED in the presence of LO-phonons. Also in this paper, we demonstrate an application of our approach by treating the dynamics and correlations in a system consisting of InAs semiconductor quantum dot (QD) interacting with a microcavity photon field and a phonon bath. By describing both interactions on equal footing and under arbitrary coupling conditions, we discovered a surprising phenomenon where the 2nd order correlation, \( g^{(2)}(0) \) for a photon field prepared in a thermal state is reduced by the electron-phonon-bath interaction to a value exhibited by nonclassical (antibunched) radiation.

To derive the working equations, we consider one electron populating a QD in a microcavity. Within the effective mass approximation, [10] the QD is treated as a two-level system with transition energy \( \hbar \omega_{cv} = \hbar (\omega_c - \omega_v) \), where \( \omega_c \) and \( \omega_v \) are the lower- and upper-state energies, respectively. The QD interacts with the radiation field and with LO-phonons of the bulk host material, cf. Fig 1. The radiation field is in a cavity mode of frequency \( \omega \), and calculations were performed assuming an InAs quantum dot in bulk GaAs: \( \hbar \omega_{cv} = 1.0eV \), \( \hbar \omega_{LO} = 36meV \) and \( \hbar M = 80\mu eV \).

Figure 1: QD-CQED problem. The quantities are defined in the discussion surrounding Eq. 1 and calculations were performed assuming an InAs quantum dot in bulk GaAs: \( \hbar \omega_{cv} = 1.0eV \), \( \hbar \omega_{LO} = 36meV \) and \( \hbar M = 80\mu eV \).

\[
H = \sum_{i=v,c} \hbar \omega_i a_i^\dagger a_i + \hbar \omega c^+ c + \hbar \omega_{LO} \sum_q b_q^\dagger b_q - \hbar (M a_{c(\nu)}^\dagger a_{c(\nu)} + \sum_{q,i=v,c} g_q^c a_i^\dagger a_q b_q) + h.a. \tag{1}
\]

where \( a_{c(v)} \), \( a_{c(v)}^\dagger \) are the conduction (valence) electron annihilation and creation operators; \( c, c^\dagger \) are the photon
annihilation and creation operators; $b_q$, $b_q^\dagger$ are the LO-phonon annihilation and creation operators at momentum $\mathbf{q}$. In the above equation, $M$ is the electron-phonon coupling coefficient [1, 2] and $g_q$ is the electron-phonon Fröhlich-coupling element. [11, 12]

Central to our approach are two factors that allow the numerical evaluation of system dynamics to any desired accuracy. First is the recognition that the photon and phonon operators appear exclusively as correlations with electronic operators, which for the one-electron, 2-level system may be given entirely by $a_i^\dagger a_v$, $a_i a_w$ and $a_i^\dagger a_w$. The conjecture is to described the problem completely with the expectation values of dressed (phonon or phonon-assisted) versions of these electronic operator combinations, i.e. with $\mathcal{G}_{n,m}^{p,s} = \langle a_i^\dagger a_v e^{ip_c b_q b_m^\dagger} \rangle$, $E_{n,m}^{p,s} = \langle a_i a_w e^{ip_c b_q b_m^\dagger} \rangle$ and $T_{n,m}^{p,s} = \langle a_i^\dagger a_v e^{ip_c b_q b_m^\dagger} \rangle$, where we introduce $b = \sum_q (g_q^e - g_q^e)b_q$ and its Hermitian adjoint. Each expectation value describes a different order in electron-light and electron-phonon coupling, obtained by raising or lowering the indices $p,s,m,n$, starting with $\langle G_{00}^{00} \rangle$ and $\langle E_{00}^{00} \rangle$, which are the ground- and excited-electronic occupations, respectively, and with $\langle T_{00}^{00} \rangle$, which is the microscopic polarization linking optical transitions between states $c$ and $v$. The second factor is that the equations of motion for $\mathcal{G}_{n,m}^{p,s}$, $E_{n,m}^{p,s}$, and $T_{n,m}^{p,s}$ can be derived completely by working in the Heisenberg picture and applying mathematical induction. These equations display the dynamics of the electronic populations as influenced by photon- and phonon-assisted transitions:

$$\partial_t \mathcal{G}_{n,m}^{p,s} = -i [m - n] \omega_{\text{LO}} + i (p - s) \omega - (p + s) \kappa] \mathcal{G}_{n,m}^{p,s} + i M T_{n,m}^{p+1,s} - i M (T_{n,m}^{s+1,p})^* + isMT_{n,m}^{p-1,s} (2)$$

$$\partial_t E_{n,m}^{p,s} = -i [\omega_{c v} - (p - s) \omega + (m - n) \omega_{\text{LO}}] E_{n,m}^{p,s} - (p + s) \kappa T_{n,m}^{p,s} - iT_{n,m}^{p+1,s} - iT_{n,m}^{s+1,p} - iM(pE_{n,m}^{p-1,s} + E_{n,m}^{p+1,s} - G_{n,m}^{p+1}) + iM^*(pE_{n,m}^{p-1,s} + E_{n,m}^{p+1,s} - G_{n,m}^{p+1}) + iM^*(pE_{n,m}^{p-1,s} + E_{n,m}^{p+1,s} - G_{n,m}^{p+1}) (3)$$

$$\partial_t E_{n,m}^{p,s} = -i [(m - n) \omega_{\text{LO}} + i (p - s) \omega + (p + s) \kappa] E_{n,m}^{p,s} - i M T_{n,m}^{p+1,s} + i M (T_{n,m}^{s+1,p})^* + inE_{n,m}^{p-1,s} - imE_{n,m}^{p+1,s} (4)$$

where $g_i = \sum_q (g_q^e - g_q^e)$ for $i = v,c$ and $\kappa = 10 \mu eV$ considers a cavity loss.

Equations 3 - 4 also provide the equations of motion for the statistical and coherence properties of the radiation field. [2, 13] Together, they represent an infinite hierarchy of coupled differential equations, cf. Fig. 2. Such a situation also occurs in other approaches, e.g. in the cluster expansion method, where each succeeding equation in the hierarchy has to be derived individually and the derivation usually gets more demanding with each higher correlation order. [9, 14, 15] In contrast, our approach provides the equations for all orders in the expansion explicitly as shown in Eqs. (2) - (4) and the iteration to any desired level in the hierarchy is readily implementable in the numeral evaluation. A restriction is that there can only be a finite number of electronic states, which does not effect the applicability of our approach to typical QD-CQED problems. An advantage is that electron-phonon and electron-phonon interactions are treated on equal footing and for arbitrary coupling conditions (e.g. with Rabi frequency $\Omega \approx \sqrt{\gamma_b}$, where $g_{\text{eff}} = \sum_q (g_q^e - g_q^e)^2$ and beyond 2nd Born approximation for electron-phonon-bath interaction).

Now, we will illustrate an application of our approach by studying the influence of an LO-phonon bath on emission dynamics and statistics of a QD prepared in the excited state and residing inside a microcavity, where the single-mode photon field is initially in either a Fock state or a thermal state[16]. The LO-phonon bath and the electronic system are assumed to be initially uncorrelated. At the starting point $t = 0$, a short pulse excites the electronic system on a time scale that the LO-phonon bath remains in equilibrium. The starting conditions are $G_{0,0}^{0,0}(0) = G_{1,1}^{0,0}(0) = 0$, $E_{0,0}^{0,0}(0) = 1$ and $E_{1,1}^{0,0}(0) = n_\omega n_{\text{LO}}$, where $n_\omega$ is the photon number. For the thermal state $n_\omega = [\exp(\beta \omega) - 1]^{-1}$ with $\beta = (k_B T)^{-1}$ chosen to give the desired mean photon number. For the phonon distribution, $n_{\text{LO}} = [\exp(\beta_{\text{LO}} \omega_{\text{LO}}) - 1]^{-1}$, where $\beta_{\text{LO}} = (k_B T_{\text{LO}})^{-1}$ and $T_{\text{LO}}$ is the phonon-bath temperature. The remaining initial conditions are $G_{n,m}^{p,s}(0) = E_{n,m}^{p,s}(0) = 0$ for all other values of $p,s,n,m$ and $T_{n,m}^{p,s} = 0$ for all $p,s,n,m$. For an initial Fock state with photon number $n_\omega = 1$ and $\omega = \omega_{\text{QD}} - \Delta_{\text{LO}}$, Fig. 3 shows the computed time dependences of the photon population for two phonon-bath temperatures. To benchmark to the Jaynes-Cummings model and provide a reference for latter discussions, we use a low photon-bath temperature $T_{\text{LO}} = 3k_B$ so that LO-phonon effects are negligible. In Fig. 3 (a), the dotted curve depicts a photon number oscillating between $1 \leq n_\omega \leq 2$ at Rabi frequency $\Omega = \mathcal{M}/\sqrt{2}$, reproducing exactly the analytical solution for a two-level system interacting with a photon number state in the absence of
The similarities between the solid time traces and spectra in Fig. 3 are surprising for they suggest an insensitivity of QD-CQED to the initial photon statistics, when phonon interaction is included. Such a conclusion is misleading because it is based only on the photon dynamics, without considering other properties of the photon field, in particular, the 2nd order correlation, $g^{(2)}(0)$. Figures 4 (a) and (b) show the computed time traces of $\langle c^\dagger(t) c(t) c(t) c(t) \rangle / \langle c^\dagger(t) c(t) \rangle^2$. The dotted traces are computed for $T = 3K$. Aside from the strong oscillations for both situations, the Fock-state trace indicates values between 0 and 0.5 as expected for nonclassical light in a superposition of 1- and 2-photon Fock states. For the initially thermal-state case, the values lie mainly above 1, as expected for classical light. The solid traces show the corresponding results at phonon-bath temperature $T = 300K$. For the Fock-state case, the phonon interaction leads to a curve with reduced amplitude variation, that overlaps with mostly the upper portion of the $T = 3K$ curve. For the thermal-state situation, the phonon-bath also shrinks the amplitude fluctuations, but the curve now overlaps mostly with bottom portion of the $T = 3K$ one.

The significance of the difference in phonon-bath influence is better illustrated by casting the results in a form resembling what might be measured in an experiment. [20] Figures 4 (c) to 4 (f) plot the occurrences of a given $g^{(2)}(0)$ value, taken from the time traces over a time span of 2 ns and the arrows indicate the average values.

Figure 3: Left column is for initial Fock state and right column is for initial thermal state. (a) and (b) plot the photon population versus time for $T_{LO} = 3K$ and $300K$ (dotted and solid traces, respectively). (b) and (e) are the intensity spectra for $T_{LO} = 3K$. (c) and (f) are the intensity spectra for $T_{LO} = 300K$.

Figure 4: Left column is for initial Fock state and right column is for initial thermal state. (a) and (b) plot the photon correlation versus time for $T_{LO} = 3K$ and $300K$ (dotted and solid traces, respectively). (c) and (d) show the count number for a given $g^{(2)}(0)$ for $T_{LO} = 3K$. (e) and (f) show the results for $T_{LO} = 300K$. The counts are accumulated over a time span of 2 ns and the arrows indicate the average values.

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regions. On the left column are the results for Fock-state case. Comparison of the top and bottom shaded regions indicates that the phonons narrow the range of $g^{(2)}(0)$ values exhibited by the system. The average $g^{(2)}(0)$ increases because of the dissipation via the phonon bath. On the right column are the results for the thermal state, depicting also narrowing of the range of $g^{(2)}(0)$ values when phonons are present. However, the $g^{(2)}(0)$ distribution is skewed towards lower values, so that the average is reduced from 1.1 to 0.8, with a most probable value of 0.5. This is interesting because it indicates conversion of classical to nonclassical light, and counterintuitive in that the photon field is made less bunched or random via interaction with a bath. The antibunching mechanism appears similar to that producing squeezed states with parametric amplification.[10] In our case, the local oscillator is the LO phonon bath [see Figs. 3 (c) and 3 (d)], where the energetically flat phonon dispersion imparts regularity in the beating of the different Rabi oscillations. The significantly longer phonon wavelength compared to the microcavity length may be the reason for an incoherent phonon bath to have the same effect as a coherent laser local oscillator.

In summary, this paper introduces and applies an exactly solvable quantum-dot microcavity quantum electrodynamics model that includes the interaction with a phonon bath. Equations of motion for electron, photon and LO-phonon dynamics are derived in the Heisenberg picture using a mathematical induction scheme and solved with numerical evaluation techniques. The approach was applied to investigate emission from a quantum dot in the high-phonon-temperature, few-photon limit. For a cavity mode prepared in a Fock state, it reproduces the Jaynes-Cummings solution, and including the phonon interaction leads to higher $g^{(2)}(0)$ values. In contrast, for a cavity mode prepared in a thermal state, we discovered a counterintuitive reduction in $g^{(2)}(0)$ with phonon interaction, to an extent that the initial classical (bunched) photon distribution evolves into a nonclassical (antibunched) one. The result occurs at high ($T_{LO} = 300K$) temperature, and is therefore interesting for engineering applications involving nonclassical light. Also, it is relevant for understanding photon antibunching experiments when blackbody radiation from cavity wall is considered.

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[20] Note, the $g^{(2)}(0)$ value refers to an average of several measurements at different times.