Photoluminescence spectra from quantum dots coupled to structured photonic reservoirs

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The spontaneous emission rate of a quantum dot coupled to a structured photonic reservoir is determined by the frequency dependence of its local density of photon states. Through phonon-dressing, a breakdown of Fermi’s Golden rule can occur for certain photonic structures whose photon decay time become comparable to the acoustic phonon decay times. We present a polaron master equation model to describe photoluminescence spectra from a quantum dot coupled to a structured photonic reservoir. We consider specific examples of a semiconductor microcavity and a coupled cavity waveguide and show clear photoluminescence signatures that contain unique signatures of the interplay between phonon and photon bath coupling. © 2014 Optical Society of America

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Quantum dots (QDs) behave as artificial atoms in a solid-state medium and are promising for scalable quantum information processing [1]. Quantum dots can also be coupled to structured photonic reservoirs like photonic crystals, which alters their light-matter interaction dynamics. However, lattice phonon interactions cause QDs to behave quite differently from simple atoms [2], manifesting in various effects such as damping and frequency shifts of driven Rabi oscillations [3–5], excitation induced dephasing of Mollow side-bands [6], off-resonant cavity feeding [7] and asymmetric vacuum Rabi doublets [8, 9]. Recently [10], it has been shown that for certain photonic reservoirs, a break-down of Fermi’s Golden rule can occur when determining the spontaneous emission (SE) rate of an embedded QD. Specifically, for reservoir decay times that are comparable to the phonon relaxation times, a large bandwidth of the local photon density of states (LDOS) dictates how SE occurs. Thus the SE rate is not only determined by the LDOS at the emitter frequency.

In this Letter we develop a polaron ME approach [10] that includes both a photon reservoir and a phonon reservoir, and explore the influence of the phonon-modified SE rate on the photoluminescence spectra (PL) of a coherently driven QD. The PL spectra is a useful experimental technique for determining emission properties of a QD, and recent experiments already show clear signatures of the phonon bath [2, 6] in absence of any photon reservoir coupling. Here we show theoretically how the PL spectra of a QD excited by a weak coherent drive, changes in presence of a structured photonic reservoir. We highlight several non-trivial PL signatures that arise due to broadband frequency dependence of the QD SE rate and perform a systematic study of these effects as a function of temperature. Our theory can be applied to any general LDOS medium, but we focus on typical structures that are currently being explored experimentally, including microcavities and coupled cavity waveguides, as shown schematically in Fig. 1(a-b).

The QD is modelled as a two-level system interacting with an inhomogeneous photonic reservoir and an acoustic phonon bath [11] (Fig. 1(c)). To model PL spectra, we consider a QD that is weakly driven by a cw (continuous wave) pump laser of Rabi frequency \( \eta_x \). The total Hamiltonian of the system in a frame rotating at the laser frequency \( \omega_L \), is [12]

\[
H = \hbar \int dr \int_0^{\infty} d\omega \, f^\dagger(r, \omega) f(r, \omega) + \hbar \Delta_{zL} \sigma^+ \sigma^- - \left[ \sigma^+ e^{i\omega_L t} \int_0^{\infty} d\omega \, d \cdot E(r_d, \omega) + H.c. \right] + \hbar \eta_x (\sigma^+ + \sigma^-) + \Sigma_q \hbar \omega_q b_q^\dagger b_q + \sigma^+ \sigma^- \Sigma_q \hbar \lambda_q (b_q^\dagger + b_q),
\]

(1)
where $\sigma^+/\sigma^-$ are the Pauli operators of the exciton (electron-hole pair), $d = \Delta \mathbf{n}_0 \bar{q}$ is the dipole moment of the QD at spatial position $\mathbf{r}_d$, $\Delta_x L = \omega_r - \omega_L$ is the exciton-laser detuning, $b_q (\bar{b}_q^\dagger)$ are the annihilation (creation) operators of the acoustic phonons, and $\lambda_0$ is the exciton-phonon coupling strength; $f/f^\dagger$ are the boson field operators of the photon reservoir, and we have used the dipole and the rotating wave approximation in describing the interaction between the QD and photonic reservoir. The electric-field operator $E(\mathbf{r}, \omega)$ is related to the medium Green function $G(\mathbf{r}, \mathbf{r}', \omega)$ [12].

The Hamiltonian $H$ is polaron transformed as $H' \rightarrow e^{\phi} H e^{-\phi}$ where $P = \sigma^+ \sigma^- \Sigma_{q} \frac{\Delta \mathbf{n}_0}{\pi} (\bar{b}_q - b_q^\dagger)$ [13], which includes electron-phonon interaction to all orders. For simplicity, a polaron definition of $\Delta_x$ [11] represents incoherent excitation and radiative decay of the QD at spatial position $\mathbf{r}_d$, respectively [17], and the cross-dephasing term of the form $\gamma_{\text{cd}} = \frac{1}{2} \sigma^+ \sigma^-$ is implicitly included in the definition of $\Delta_x$. For such measurement [11] and photon variables [15] is performed in deriving Eq. (2) where the phonon and photon reservoirs are assumed to be in thermal equilibrium and statistically independent [16].

The incoherent interaction of the QD with the photon and the phonon reservoirs are described by the super-operator terms $\mathcal{L}_{\text{phot}}$ and $\mathcal{L}_{\text{phon}}$, respectively; $\mathcal{L}_{\text{phon}}$ is not influenced by the photonic reservoir and in the limit of small pump rates can be approximated as $\mathcal{L}_{\text{phon}} = \Gamma_{\text{ph}} L \sigma^+ + \Gamma_{\text{ph}} L \sigma^- - \gamma_{\text{cd}} (\sigma^+ \rho_{\sigma^+} \sigma^- + \sigma^- \rho_{\sigma^+} \sigma^-)$, where $L[O] = \frac{1}{2} (2 \rho_{O} - O - O^\dagger)$. The phonon scattering terms, $\Gamma_{\text{ph}} = 2 \langle B \rangle^2 \eta_2^2 \text{Re} \int_0^\infty d\tau e^{i \Delta_x L \tau} (e^{i \phi} - 1)$ represent incoherent excitation and radiative decay of the QD, respectively [17], and the cross-dephasing term $\gamma_{\text{cd}} = 2 \langle B \rangle^2 \eta_2^2 \text{Re} \int_0^\infty d\tau e^{i \Delta_x L \tau} (1 - e^{-i \phi})$ influences the PL spectral line-shape through dephasing of off-diagonal density matrix elements [18]. The interaction with the photonic reservoir is however modified by phonons, and is given by $\mathcal{L}_{\text{phot}}(\rho) = \int_0^\infty d\tau \int_0^\infty d\omega J_{\text{phot}}(\omega) \left[ -C_{\text{phot}}(\tau) \sigma^+ \sigma^- e^{i \Delta_x L \tau} \rho + C_{\text{phot}}(\tau) \sigma^- \rho e^{i \Delta_x L \tau} + C_{\text{phot}}^*(\tau) \sigma^+ \rho e^{i \Delta_x L \tau} + C_{\text{phot}}^*(\tau) \sigma^- \rho e^{i \Delta_x L \tau} \right]$, [10] where $\Delta_x = \omega_r - \omega$, $J_{\text{phot}}(\omega) = \frac{d}{d\omega} |G_{\mathbf{r}_d \mathbf{r}_p}(\omega)|^2$ is the photon reservoir spectral function, while $C_{\text{phot}}(\tau) = e^{i |\mathbf{\phi}| - i \mathbf{\phi}(0)}$ is the phonon correlation function. The time-dependent operator $e^{+i \Delta_x L \tau} = e^{i \Delta_x L \tau} \rho e^{i \Delta_x L \tau} + e^{i \Delta_x L \tau} \rho e^{i \Delta_x L \tau}$ results in pump field dependent scattering terms. For weak drives however, such dependence is negligible and $\text{Re} \{\mathcal{L}_{\text{phot}}\}$ gives rise to phonon-modified spontaneous emission decay $\langle \hat{\gamma} L^\dagger \sigma^- \rangle$, where the phonon-modified SE decay rate is $\tilde{\gamma} = 2 \int_0^\infty \text{Re} \{C_{\text{phot}}(\tau) J_{\text{phot}}(\tau)\} d\tau$, (3) and $J_{\text{phot}}(\tau) = \int_0^\infty d\omega J_{\text{phot}}(\omega) e^{i (\omega_r - \omega) \tau}$ is the photon correlation function. As shown elsewhere [10], this phonon-modified SE rate bears contribution from the broadband photonic LDOS sampled by the bandwidth of the phonon bath, which is in contradiction with the well known Fermi’s golden rule for SE. A measurement of SE rate as a function of photonic LDOS frequency can demonstrate the influence of such non-local LDOS contribution [10]. Such measurements are however difficult to obtain experimentally. Alternatively, a PL measurement from a coherently excited QD can be used to verify the non-local frequency dependence of SE rate. Indeed, such measurements have been used to clearly probe the phonon sidebands in PL emission from a single excited QD, but without any photon reservoir coupling [18]. Thus it is of practical relevance to observe the effects of reservoir coupling on experimentally accessible PL.

The QD PL intensity ($I_x$) from a QD is directly proportional to exciton population $n_x = \langle \sigma^+ \sigma^- \rangle = \text{Tr}(\sigma^+ \sigma^- \rho)$, where $\text{Tr}$ denotes the trace. For the current problem, an analytical form of $n_x$ is derived using the Bloch equations [18], yielding

$$n_x = \frac{1}{2} \left[ 1 + \frac{\Gamma_{\sigma^+} - \Gamma_{\sigma^-} - \tilde{\gamma}}{\Gamma_{\sigma^+} + \Gamma_{\sigma^-} + \tilde{\gamma} + \frac{1}{2} (\frac{\eta_2^2}{\eta_2^2 + \eta_2^2} \langle B \rangle^2 \eta_2^2 \text{Re} \int_0^\infty d\tau e^{i \Delta_x L \tau} (e^{i \phi} - 1))} \right]$$

(4)

where $\Gamma_{\text{pol}} = \frac{1}{2} (\Gamma_{\sigma^+} + \Gamma_{\sigma^-} + \tilde{\gamma} + \gamma')$. A temperature dependent pure dephasing term of the form $\gamma' = 3 + 0.95(T - 1) \mu eV$ [9,19] is also included in the ME (Eq.(2)) for calculation of the PL.

To observe the effects of reservoir coupling on PL spectra, we choose a Lorentzian single mode PC cavity (Fig. 1(a)) and a PC coupled-cavity waveguide (Fig. 1(b)), both in the weak coupling regime. The PC defect cavities (Fig.1(a)) are important for investigating fundamental aspects of cavity-QED in solid-state [20], with potential applications for quantum information processing [1] and low power optoelectronics [21]. Photonic crystal waveguides (Fig.1(b)) are useful for slow-light propagation [22] and for manipulating the emission properties of embedded QDs for on-chip single photon emission [23, 24]. The bath function for a single cavity mode is $J_{\text{phot}}(\omega) = g^2 \frac{1}{2} \frac{\pi}{\pi^2 - (\omega - \omega_0 - i \kappa)^2}$, where $g$ is the QD-cavity coupling rate, and $\kappa$ is the cavity decay rate. For the PC coupled cavity waveguide [25], the bath function is evaluated using an analytical tight-binding technique, where nearest neighbor coupling is assumed between adjacent cavities of mode volume $V_{\text{eff}}$ [26] and is $J_{\text{phot}}(\omega) = \frac{-d^2 \omega}{2 \kappa \hbar} \frac{1}{\sqrt{(\omega - \omega_0)(\omega - \omega_0 - i \kappa)}}$, where
The PF is defined as $PF = \tilde{\gamma} / \gamma$ (top panels, Fig. 2(a), 3(a)) for these two representations. In contrast, in regions where the photonic LDOS is weak, then phonons enhance the SE rate through non-local contributions of the photonic LDOS.

For our PL calculations (Fig. 2, 3(b)), the QD is held a fixed detuning from the photonic LDOS and is excited with a weak drive laser ($\eta_c = 0.4 \text{meV}$), where the laser frequency is varied to obtain the broadband PL. The light solid line in Fig. 2 (2, 3) (b) is the PL in the absence of any structured reservoirs ($\gamma_b = 1 \text{meV}$) and $\Delta_{x,L} = 0 \text{meV}$ marks the position of the zero phonon line (ZPL). The cavity (Fig. 2(b)) and the waveguide upper mode-edge (Fig. 3(b)) is located approximately 1 meV to the right of the ZPL (dark dashed line). In the absence of a structured photonic reservoir, the PL is enhanced around this region at low temperatures [2].

Phonon assisted incoherent excitation process $\Gamma^{\sigma^+}$ increases with laser detuning $\Delta_{x,L}$ due to higher phonon emission probability and the phonon-induced decay rate $\Gamma^{\sigma^-}$ reduces [17]. This leads to larger population excitation $n_b$ (Eq.(4)) and hence stronger PL emission on blue side of the ZPL. However, the presence of the structured reservoirs reduces the PL (light dashed line). For weak driving, the SE rate $\gamma$ samples the photonic LDOS at the drive laser frequency ($\omega_L$) when the Markov approximation is valid [12, 28, 29]. Hence $\gamma$, following the photonic LDOS, is enhanced in this region. Thus the PL shows a dip (Fig. 2(b) left inset and Fig. 3(b)). When we include a phonon-modified SE rate ($\tilde{\gamma}$), the size of this dip reduces (dark solid line). It should be noted at this point, that in the case of a cavity, $\tilde{\gamma}$ can also be estimated using phonon-mediated cavity scattering rates [17], in the mean-field approximation [10]. However for the cavity parameters used, there is negligible difference in the PL spectra above, calculated using this mean field approximation [10]. For smaller Q cavities (i.e., $Q < 1000$), these two ME techniques result in substantially different SE rates $\tilde{\gamma}$, though the PL dip is smaller. Thus cavities with Q factors greater than a few thousands are
better to capture the non-local LDOS effects through PL measurements. However any departure from a symmetric Lorentzian LDOS would require the full reservoir calculation of $\gamma$ (Eq. 3) to understand the PL spectra.

In the case of the cavity, the broadband frequency dependence of SE can be directly observed in the from the non-Lorentzian line-shape of the PL dip in Fig. 2(b) left inset. However the dependence of PL-dip on temperature bears a clearer signature of this effect. The PL dip is estimated from the difference in intensity between the points marked by the circles in Fig. 2(b) and 3(b). The circles mark the PL dip and the highest intensity on the right of the dip. When the SE is not influenced by phonons, the intensity dip increases as a function of temperature as the PL intensity of the phonon side-band increases (Fig. 2(b), 3(b)) inset, dashed line). When phonon effects to the SE rate are included, then the size of this PL dip reduces (Fig. 2, 3 (b) inset, thick solid line). This is caused by a phonon-induced reduction of SE close to peak LDOS and an enhancement away from the peak LDOS (dark solid line). Such behavior is more discernable in the case of a waveguide (Fig. 3(b) inset, thick solid line), due to the asymmetric nature of its mode-edge LDOS [10]. Note that a symmetric Lorentzian line-shape of the PL dip in Fig. (2(b) left inset). However the dependence of PL-dip on temperature arises from the difference in intensity between the points marked by the circles in Fig. 2(b) and 3(b). The circles mark the PL dip and the highest intensity on the right of the dip. When the SE is not influenced by phonons, the intensity dip increases as a function of temperature as the PL intensity of the phonon side-band increases (Fig. 2(b), 3(b)) inset, dashed line). When phonon effects to the SE rate are included, then the size of this PL dip reduces (Fig. 2, 3 (b) inset, thick solid line). This is caused by a phonon-induced reduction of SE close to peak LDOS and an enhancement away from the peak LDOS (dark solid line). Such behavior is more discernable in the case of a waveguide (Fig. 3(b) inset, thick solid line), due to the asymmetric nature of its mode-edge LDOS [10]. Note that a symmetric Lorentzian LDOS would require the full reservoir calculation of $\gamma$ (Eq. 3) to understand the PL spectra.

In conclusion, we have introduced a theory to describe photoluminescence spectra from a weakly excited QD coupled to a structured photonic reservoir in the presence of electron-phonon coupling. Using this theory, we have demonstrated how the frequency dependence of the phonon-modified SE rate influences the PL, causing clear spectral signatures that should be observable in related experiments. Our theory can be used to model the QD PL embedded in any photon reservoir system.

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