Nonlinear photoluminescence spectra from a quantum-dot–cavity system: Interplay of pump-induced stimulated emission and anharmonic cavity QED

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We investigate the power-dependent photoluminescence spectra from a strongly coupled quantum dot-cavity system using a quantum master equation technique that accounts for incoherent pumping, stimulated emission, pure dephasing, and fermion or boson statistics. Analytical spectra at the one-photon correlation level and the numerically exact multiphoton spectra for fermions are presented. Master equation models that neglect stimulated emission processes are shown to lead to unphysical predictions at high powers, such as negative mean photon numbers. We compare to recent experiments on a quantum dot-micropillar cavity system and show that an excellent fit to the data can be obtained by varying only the incoherent pump rates in direct correspondence with the experiments. Our theory and experiments together show convincing evidence for stimulated-emission induced broadening and anharmonic cavity quantum electrodynamics.

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Introduction. Single quantum dot (QD) cavity systems facilitate the realization of solid state qubits (quantum bits) and have applications for producing single photons\textsuperscript{1–3} and entangled photons.\textsuperscript{4,5} Rich in physics and potential applications, the coupled QD cavity has been inspiring theoretical and experimental groups to probe deeper into the underlying physics of both weak and strong coupling regimes of semiconductor cavity quantum electrodynamics (QED). Key signatures of cavity QED include the Purcell effect and vacuum Rabi oscillations. Although a well-known phenomenon in atomic cavity optics,\textsuperscript{6} vacuum Rabi splitting in a semiconductor cavity was only realized a few years ago.\textsuperscript{7,8} Inspired by the recent surge of related experiments, researchers have been working hard to develop new theoretical models to understand the semiconductor cavity QED systems. For example, the persistent excitation of the cavity mode for large exciton-cavity detunings was measured,\textsuperscript{9–12} and qualitatively explained by extended theoretical approaches that account for coupling between the leaky cavity mode and the exciton, and by showing that the main contribution to the emitted spectrum comes from the cavity-mode emission.\textsuperscript{13–17} These formalisms assume an initially excited exciton or an initially excited leaky cavity mode, and they are valid for low-pump powers. However, an interesting question that has been posed recently for the semiconductor systems, e.g., see Refs. 18–20, is what is the role of an incoherent pump on the photoluminescence (PL) spectra, where the pump can excite the exciton and (or) cavity mode? To experimentally investigate the pump-dependent spectra, two recent experiments have been respectively reported by Münch et al.\textsuperscript{21} for a QD-micropillar system, and by Laucht et al.\textsuperscript{22} for a QD-photonic crystal system; these measurements clearly show the pump-induced crossover from strong to weak coupling.

In this Brief Report, we present a straightforward master equation (ME) theory that self-consistently includes incoherent pumping, stimulated emission, and pure dephasing. We derive useful analytical results at the level of one-photon correlations and present numerically exact results for the multiphoton spectra. We reanalyze the Würzburg\textsuperscript{21} experiments directly and show the striking differences with previous models that neglect stimulated emission.\textsuperscript{19,20,22} Accounting for fermion statistics, pure dephasing, and the thermal bath model for the exciton pump, an excellent fit to the data is obtained by only changing the incoherent pump rates in direct correspondence with the experiments.

Cavity System and Model. The system investigated here is shown as a scanning electron microscope (SEM) image in Fig. 1, along with the extended experimental data of Ref. 21. We make the following model assumptions: the cavity is single mode in the frequency of interest; the coupling between the cavity and target QD exciton is described through

\begin{equation}
\omega_x \approx \omega_c
\end{equation}

FIG. 1. (Color online) Typical broadband PL spectrum that is emitted when a target exciton is closely resonant with the cavity mode (near $\omega_0 = 1331.355 \text{ meV}$); away from the target exciton, there are a series of other exciton levels that can also couple, off-resonantly, to the cavity mode. The SEM image shows our micropillar cavity and the QD layer. The emitted photons from the QDs are detected through vertical emission.
a coupling rate g; the decay rate of cavity is $\Gamma_\text{c}$; for the strongly coupled QD, we include only the target exciton as a system operator, and consider both radiative decay, $\Gamma_\text{r}$, and pure dephasing, $\Gamma_\text{c}$. The QD-cavity system is driven simultaneously by an exciton pump, $P_\text{e}$, and a cavity pump, $P_\text{c}$; the former is caused by the incoherent relaxation of electron-hole pairs from the higher energy level, and the latter is due to the cavity coupling with off-resonant excitons (probably coming from other QDs in the cavity layer). To treat the incoherent excitation, we consider a system-reservoir interaction, apply a Born-Markov approximation, and trace over the cavity and target exciton pump reservoirs (bath approximation). One has

$$\frac{dp}{dt} = -\frac{i}{\hbar}[H_\text{e},p] + \mathcal{L}(p),$$

(1)

with the system Hamiltonian $H_\text{e} = \hbar \omega_x \sigma^+ \sigma^- + \hbar \omega_0 \hat{a}^\dagger \hat{a} + \hbar g (\sigma^+ \hat{a}^\dagger + \sigma^- \hat{a})$, where $\hat{a}$ represents the cavity mode operator, $\sigma^\pm$ are the Pauli operators of the target QD exciton (with resonance frequency $\omega_0$), and $\omega_x$ is the eigenfrequency of the leaky cavity mode. The target exciton and cavity mode get pumped incoherently through the corresponding reservoirs. The density operator of the reservoirs can be written as $\rho_{\text{res}}^O = \sum_{\bar{n}_k, \bar{n}_k^O} \vert \bar{n}_k^O \rangle \langle \bar{n}_k^O \vert \otimes \rho_{\text{res}}^k$, for $O = x, c$; where $\rho_{\text{res}}^k$ is the density of reservoir modes and $n_k^O$ is the number of photons in the mode of wave vector $k$. The correlations for the photon reservoir operators $\Delta_k^O$ are given by $\langle \Delta_k^O \rangle = 0$, $\langle [\Delta_k^O]^2 \rangle = n_k^O \delta_{k,0}$, and $\langle \Delta_k^0 \Delta_k \rangle = (n_k^O + 1) \delta_{k,0}$. Defining the average number of photons on the cavity frequency as $\bar{n}_c^O = n_{k_c}^O$, at $k = \omega_x / c$, yields the effective cavity pump rate: $P_c = \Gamma_c \bar{n}_c^O$. This incoherent pump process is consistent with the model of Tian and Carmichael. The superoperator in Eq. (1) becomes

$$\mathcal{L}(p) = \frac{P_\text{e}}{2} \langle 2\hat{a}^\dagger \hat{a} \rho - \hat{a}^\dagger \hat{a} \rho \hat{a}^\dagger \hat{a} \rangle,$$

$$+ \frac{\Gamma_\text{r} + P_\text{c}}{2} \langle 2\hat{a}^\dagger \hat{a} \rho - \hat{a}^\dagger \hat{a} \rho \hat{a}^\dagger \hat{a} \rangle,$$

$$+ \frac{P_{12}}{2} \langle 2\hat{a}^\dagger \rho \sigma^- - \hat{a}^\dagger \sigma^- \rho \hat{a} \sigma^- \rangle + \frac{P_{21}}{2} \langle 2\hat{a}^\dagger \rho \sigma^+ - \hat{a}^\dagger \sigma^- \rho \hat{a} \sigma^- \rangle,$$

$$- \langle \sigma^+ \sigma^- \rho - \rho \sigma^+ \sigma^- \rangle + \frac{\Gamma'_c}{4} \langle \rho, \rho \sigma^- \rangle - \rho,$$

(2)

which is in Lindblad form. For the exciton pump we consider two different models, thermal bath model, $P_{12} = P_c$ and $P_{21} = \Gamma_c + P_e$; and a laser model (heat bath at negative temperatures). The power-dependent PL shows the power-dependent spectra for the thermal bath model (left) and the laser model (right); and in Fig. 2(b), we compare the trend expected from a ME model that neglects stimulated emission processes. The red curves show the one-photon results and the blue curves show the multiphoton case. Although all figures show a similar trend of the doublet becoming a singlet as a function of power, the high-power linewidths are substantially different. In particular, the model with stimulated emission predicts a much larger pump-induced broadening as a function of power. In the absence of stimulated emission, the pump-induced broadening is sup-

BRIEF REPORTS PHYSICAL REVIEW B 81, 033309 (2010)
pressed, and the larger pump rates result in negative exciton and photon densities. The mean exciton number (dashed) and photon number (solid) are shown in Fig. 2(c) using the multiphoton model. Here, we see the drastic influence on the predicted densities if stimulated emission is not included (right), where negative photon densities are predicted in addition to regimes of $n_x > 1$, both of which are obviously unphysical; though we show the thermal bath case here, the laser model gives similar unphysical results. Naturally, with stimulated emission neglected in the model, the regime of $P_c > \Gamma_x$ is phenomenologically not allowed, so the top spectra in Fig. 2(b) are not reliable.

The experimental data is shown in Fig. 2(d), alongside the thermal-bath fermion model, and there is a very good correspondence, even when the only fitting parameter is a proportionality constant with $P_x$. We further remark that even at the one-photon-correlation level, a good fit can also be obtained if one adjusts the proportionality constant; and one of the main points we wish to emphasize here is the importance of including stimulated emission. Although $\Gamma'_x$ may also be pump-dependent, we find that increasing its value by 1–2 orders of magnitude has little influence on our high-power PL, as the stimulated-emission-induced broadening is by far the dominant source of broadening. To have further confidence in the theoretical interpretation, it is important that the models consistently fit the normalized PL on and off-resonance, as well as the integrated PL. In this regard, we obtain very good fits to the spectra when the cavity and exciton are off-resonance (not shown) and for the integrated PL [inset in Fig. 2(d)].

Since our QDs are rather large, e.g., elongated with lengths on the order of 100 nm and widths of about 30 nm, it is natural to present the nonlinear boson PL calculations as well. In Fig. 3, we display the exact boson PL using the two exciton pump models, again with and without stimulated emission terms. Since pure dephasing cannot be included in this boson model, we set the effective exciton decay rate $\Gamma_x = -\Gamma_x' + \Gamma_x''$ to have the same overall broadening. Clearly, none of the PL traces follow the trends of the experiments [cf. Fig. 2(d)], and only the thermal bath models produce net positive densities for all pump rates. Moreover, even the low-power PL have different lineshapes due to the important effect of pure dephasing, which acts to suppress the Rabi oscillations without affecting the envelope of the population decay. While it has been discussed before that the boson model apparently fits well to the same data, since fits were obtained under variation of the coupling constant $g$ and three other free parameters ($\Gamma''_x, \Gamma'_x, P_x$), we believe that having so many free parameters (and a model that neglects stimulated emission) can be detrimental to highlighting the correct underlying physics.

**High-Pump-Power Inversion and Lasing.** Finally, we briefly connect to the prospects for observing one exciton lasing in such a QD-cavity system. It is well known in the field of atomic optics, e.g., see Ref. 25, that the spectral properties of pump-dependent PL can be used to explore the regime of single atom lasing. Characteristic signatures of single state lasing in atomic physics include spectral narrowing, inversion, and a regime of linearly increasing mean photon number as a function of pump power. On the other hand, an incoherent pump of thermal photons will naturally be det-
FIG. 4. (Color online) (a) Mean exciton number versus $P_x$ (with $P_x = 1.6 P_y$, as before): exciton pump thermal-bath model (solid) and laser model (dashed); both cases include stimulated emission. (b) Corresponding mean photon number (left axis: blue) and Fano factor $F$ (right axis: red).

Conclusions. A ME formalism, with incoherent pumping, pure dephasing, and a QD fermion model, has been introduced and used to investigate the power-dependent PL spectrum of a QD exciton under steady-state pumping. We have shown the importance of self-consistently including stimulated emission, and validated our model by directly comparing with recent experimental data on semiconductor microcavity cavities. A very good fit to the data is obtained by only changing the pump rates in direct correspondence with the experiments, showing that we are well into the elusive regime of anharmonic semiconductor cavity QED.

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28We do confirm, however, that Ref. 22 has indeed net positive densities, for their chosen parameters.
29The Fano function, $F = \langle (\hat{a}^\dagger \hat{a} - \langle \hat{a}^\dagger \hat{a} \rangle)^2 \rangle / \langle \hat{a}^\dagger \hat{a} \rangle$, can be used to assess a lasing threshold, if a clear maximum is obtained, e.g., see Ref. 25.