Photoluminescence of Single Quantum Dots in Microcavities

A. Ridolfo\(^1\), O. Di Stefano\(^1\), S. Portolan\(^2\), S. Savasta\(^1\), R. Girlanda\(^1\)

\(^1\) Dipartimento di Fisica della Materia e Ingegneria Elettronica, Università di Messina Salita Sperone 31, I-98166 Messina, Italy
\(^2\) Institute of Theoretical Physics, EPFL, CH-1015 Lausanne, Switzerland
E-mail: ssavasta@unime.it

Abstract. We study theoretically the photoluminescence of a single quantum dot in a microcavity under incoherent excitation. Analytical results including pure dephasing show that strong coupling and linewidths are largely independent on the pumping intensity (until saturation effects come into play). We show the reliable predicting character in the analysis of some experiments.

1. Introduction

Cavity quantum electrodynamics (CQED) studies the interaction between a quantum emitter and a single radiation-field mode. When an atom is strongly coupled to a cavity mode [1], it is possible to realize important quantum information processing tasks [2]. Indeed semiconductor quantum dots provide nanoscale electronic confinement resulting in discrete energy levels, and an atom-like light-matter interaction. CQED addresses properties of atomlike emitters in cavities and can be divided into a weak and a strong coupling regimes. When the photon-emitter interaction strength overcomes losses, the system enters the so-called strong coupling (SC) regime [3, 4]. In this case the usual irreversible spontaneous emission dynamics changes into a reversible exchange of energy between the emitter and the cavity mode. Commonly, these solid state systems in the SC regime are characterized with respect to their behaviour under optical incoherent pump excitation [2, 3, 4, 5, 6, 7, 8]. The specific interplay of photon, exciton pumping and decay results in a mixed quantum steady state that influences considerably the observed spectra as we are going to show in detail (see Fig. 1). Hence, appropriate theoretical modeling of SC with semiconductor QDs under incoherent excitation is very important in order to fulfill the great expectations nowadays attended from future implementations of SC in QD systems. In this work we provide a theoretical model able to describe SC of a single QD in a semiconductor microcavity under incoherent excitation. In order to avoid any inconsistencies or artifacts, we start from a microscopic description of the pumping and decay mechanisms of the QD and the cavity mode. Cavity mode and quantum dot excitations are considered as two strongly coupled subsystems, each in interaction with two independent reservoirs providing both damping and pumping mechanisms. The \(P_x\) represents the incoherent pump contribution due to exciton-exciton scattering mechanism mediated by phonons. At the same time, the cavity mode is weakly coupled to others various electronic transitions of the system which would contribute in feeding the cavity mode as well, resulting in a second incoherent pumping channel \((P_a)\). In
the limit of weak excitation density the resulting dynamics coincides with that of two strongly coupled harmonic oscillators in the presence of reservoirs. In particular, at low excitation regime we obtain a very simple analytical expression for the PL spectrum, that do not display any amplification effect as well as any change of Rabi splitting or broadening as function of the pump intensities. For higher pump densities saturation effects and even lasing come into play.

2. Theoretical Model and Results

The master equation for this strongly interacting system can be written as

$$\dot{\rho} = i[\rho, H_S] + \mathcal{L}_C^R + \mathcal{L}_QD^R,$$

where the system Hamiltonian reads

$$H_S = \omega_a a^\dagger a + \omega_j \sigma_+ \sigma_- + g(a^\dagger \sigma_- + a \sigma_+),$$

with $g$ being the interaction strength between the cavity mode (with annihilation operator $a$) at energy $\omega_a$ and the lowest energy ($\omega_j$) quantum dot exciton with transition operator from the ground state to the exciton level $\sigma_+ = |e\rangle \langle g|$ ($\sigma_- = \sigma_+^\dagger$). The superoperators $\mathcal{L}_C^R$ and $\mathcal{L}_QD^R$ describe the interaction of the cavity mode and of the QD with the reservoirs providing both damping and pumping mechanisms. Following e.g. Ref. [9] we model transmission and diffraction cavity losses with a quasimode picture introducing an effective coupling $\gamma^c$. The cavity pumping mechanism takes into account that in these samples there may be QDs or more generally electronic transitions weakly coupled to the cavity, in addition to the one that undergoes SC, determining an effective pumping of the cavity mode ($P_a$). For the sake of simplicity we describe these weakly coupled transitions as a continuum of harmonic oscillators coupled to the microcavity with $H_{P_a} = \sum_i g_i b_i a^\dagger + H.c.$. By applying the usual Born-Markov and rotating-wave approximations, we obtain,

$$\mathcal{L}_C^R = \frac{P_a + \gamma_a}{2} (2a^\dagger \rho a - a^\dagger a \rho - \rho a^\dagger a) + \frac{P_a}{2} (2a^\dagger \rho a - \rho a^\dagger a - \rho a^\dagger),$$

where $\gamma_a = \gamma^c + \gamma^p$ contains contributions from both the reservoirs and $P_a$ is the total pumping rate depending on the populations of the electronic levels weakly coupled to the cavity mode. Assuming only direct and weakly pumped electronic transitions we obtain $P_a = \sum_i \gamma_i \langle n_i \rangle$ and $\gamma_i = \gamma_i^d$, where $\langle n_i \rangle$ is the population of the $i$-th level and $\gamma_i^d = 4g_i^2/\omega_i^2 ([\omega_i - \omega_a]^2$, being $\omega_i$ the inverse of the dephasing time of the $i$-th transition. The material excitation strongly coupled to the cavity mode is also under the influence of two different reservoirs:

$$\mathcal{L}_QD^R = \mathcal{L}_{QD}^{se} + \mathcal{L}_{QD}^P + \mathcal{L}_{QD}^d. \text{ The first term describes spontaneous emission in all the available light modes except the cavity one, } \mathcal{L}_{QD}^{se} = (\gamma_x/2)(2\sigma_- \rho \sigma_+ - \sigma_+ \rho \sigma_- - \rho \sigma_+ \sigma_-), \text{ where } \sigma_+ = |e\rangle \langle g| \text{ and } \sigma_- = \sigma_+^\dagger. \text{ The second term } \mathcal{L}_{QD}^P = \sum_j (\alpha_j/2)(2\sigma_x \rho \sigma_- - \sigma_+ \rho \sigma_-) \text{ is responsible for the incoherent excitation of the QD (e.g. via phonon induced relaxations) from the } j \text{-th levels at higher energy that get populated by optical pumping. The } \alpha_j \text{ are temperature-dependent phonon-assisted scattering rates from the } j \text{-th excitonic levels to the state } |e\rangle \text{ [10]. The latter, } \mathcal{L}_{QD}^d = (\gamma_d/2)(\sigma_z \rho \sigma_z - \rho), \text{ describes pure dephasing, where } \sigma_z = [\sigma_+, \sigma_-].$$

This master equation induces an open hierarchy of dynamical equations. The inclusion of one-photon states only gives linear optical Bose-like dynamics. On the other hand, once also 2-photon states are taken into account, saturation and lowest order ($\chi^{(3)}$) nonlinear optical effects arise. Within our model we are able to go beyond 2-photon nonlinear dynamics, thus presenting non-perturbative calculations (see e.g. Fig. 3).
In the following we calculate the steady-state emission spectrum: \( S(\omega) = \lim_{t \to -\infty} 2 \text{Re} \int_0^\infty \langle a^\dagger (t) a(t + \tau) \rangle e^{i\omega \tau} d\tau \). In the weak excitation regime (linear dynamics) \( \langle \sigma_{\text{ee}} \rangle \ll \langle \sigma_{\text{gq}} \rangle \simeq 1 \), we obtain analytical results:

\[
S(\omega) = 2 \text{Re} \left[ \frac{i}{\sqrt{2\pi}} \frac{\omega - \tilde{\omega}_x}{(\omega - R_1)(\omega - R_2)} \right],
\]

where

\[
n_a = \langle a^\dagger a \rangle = \frac{P_a}{\gamma_a} + \frac{g^2}{\gamma_a} \frac{\gamma_a + \gamma_x + 2\gamma_d}{\gamma_a + \gamma_x}(\gamma_a P_x - \gamma_x P_a)
\]

and

\[
C = \langle a^\dagger \sigma_- \rangle = \frac{g}{\tilde{\omega}_a - \tilde{\omega}_x} (n_a - n_x);
\]

in which \( \tilde{\omega}_a = \omega_a - i\gamma_a/2 \) and \( \tilde{\omega}_x = \omega_x - i(\gamma_x + 2\gamma_d)/2 \) and \( n_x \) can be obtained from Eq. (5) just exchanging the labels \( a \) and \( x \), and \( P_x = \sum \gamma_i \langle n_i \rangle \); the complex polariton energies determining the spectrum resonances are given by \( R_{1,2} = (\tilde{\omega}_a + \tilde{\omega}_x)/2 \mp \sqrt{4\gamma^2 + (\tilde{\omega}_a - \tilde{\omega}_x)^2}/2 \) . experimental features are well reproduced by our master equation (1). Despite of its analytical simple form, eq. (4) shows a nontrivial dependence of the system on the ratio between the cavity and dot pumping rates. Fig.1 displays one PL spectrum (thin continuous line) from the data reported by Reithmaier et al. [4] together with the corresponding fit we obtain using Eq. (4) (thicker continuous line). Beside the nearly coincidence of the two curves in this situation, Eq. (4) is also in very good agreement with the experimental spectra of Ref. [4] at different detunings. We gather from the fit the ratio \( P_a/P_x = 0.86 \). We also obtain \( g = 65 \mu eV, \gamma_a = 200 \mu eV, \gamma_x = 106 \mu eV \). In order to evidence the impact that the pumping mechanism can have on SC emission lines even at very small detuning, we plotted in Fig. 1 two spectra with the same parameters as the fit but with \( P_x = 0 \) (dotted line) and \( P_a = 0 \) (dashed line). These plot shows that, close to the SC threshold, incoherent pumping of the exciton level can even hinder the line splitting. This is the case of the system investigated by Reithmaier et al. [4]. In fig. 2 we shows how an increment of dephasing causes a progressive lost of Rabi spitting although the sum of two contributions is constant i.e. \( 2\gamma_d + \gamma_x = 106 \mu eV \).

Another situation for a similar system has been recently reported in Ref. [2]. Some first results based on the present multi-photon model (1) are depicted in Fig. 3. It displays emission...
spectra at different detunings reproducing one experimental observation of the SC of a QD-cavity system reported in Ref. [2]. Although the parameters have been chosen in order to fit just one of the spectra, our model provides results which agree very well with all the spectra presented in Fig. (2a) of Ref. [2] changing only the cavity energy.

Our model is also well suited to describe situations beyond that of linear or lowest order nonlinear effects. Fig. 4 displays emission spectra obtained at different pumping intensities. These results have been obtained after a calculation of the SC quantum dynamics including all the photon number states up to a given number $N$. We truncate only when including larger photon numbers does not produce any change in the emission spectrum. We used the same broadenings of Fig. 3, a coupling $g = 76 \, \mu\text{eV}$ and we set $P_a = 0$ and $\Delta = 0$. Increasing the excitation a reduction of the Rabi splitting due to saturation effects is clearly observable. We notice how as soon as a small population inversion ($n_x > 0.5$) is reached an important narrowing of the linewidth appears indicating a lasing behaviour.

In conclusion, we have given a simple theoretical framework with a reliable predicting character for the analysis of photoluminescence properties under incoherent excitation of single quantum dot microcavity devices in a steady state maintained by a continuous incoherent pumping. In particular we provided an analytical expression for the PL spectrum of QD-cavity sistem including pure dephasing. Our model showed a great flexibility proposing simple but sensible descriptions of various experiments in the strong-coupling regime.

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