On the spectroscopy of quantum dots in microcavities

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Abstract. At the occasion of the OECS conference in Madrid, we give a succinct account of some recent predictions in the spectroscopy of a quantum dot in a microcavity that remain to be observed experimentally, sometimes within the reach of the current state of the art.

Light-matter coupling of a single quantum dot in a microcavity has recently enjoyed considerable activity. Led by sustained technological progress, various groups now control strong coupling (see Refs. [1–4] for some recent reports, and references therein). From a “naive” reading of the figures in the literature, one can establish a rough picture of the state of the art, positioning various systems (and various groups) in terms of the cavity, $\gamma_a$, and exciton, $\gamma_b$, decay rates, in units of the coupling strength $g$. This is shown in the inset of Fig. 1. The closer the system to the origin, the better it is to exhibit quantum phenomena. We shall consider in this text (and its supporting media animations) three sets of parameters:

(i) $\gamma_a/g = 0.55$, $\gamma_b/g = 0.014$,
(ii) $\gamma_a/g = 0.25$, $\gamma_b/g = 0$,
(iii) $\gamma_a/g = 0.01$, $\gamma_b/g = 0$.

The first one is the best system claimed in the literature, by Nomura et al. [5], from Arakawa’s group. As we shall shortly discuss, there should be no great confidence entrusted in the accuracy of our inset, established on the basis of the estimations of various authors, whose estimations are not always consistent for a direct comparison nor, in many cases, strictly consistent in the absolute, for lacking a quantitative analysis. Point (ii) is a slightly better system, within the reach of experiments in the immediate future. Point (iii) is an unrealistically good system, considered for its illustration of very marked quantum features.

We recently studied the regime of strong light-matter coupling between a two-level system (the quantum dot) and a single cavity mode, under an incoherent pumping, both for the quantum dot itself, $P_\sigma$, and for the cavity, $P_a$, [6] extending our treatment of the linear (and bosonic) regime [7]. We summarize and extend our main findings, and formulate desirable goals (from the point of view of the authors), both for experimentalists and for theoreticians.

The investigation itself is of a very fundamental character, since it assumes a Jaynes-Cummings hamiltonian. In the limit of vanishing excitation, the case of spontaneous emission is recovered. This case also matches that of the linear regime, which is also that of coupling of two boson modes [8], and is therefore solvable exactly. In this case, peculiarities that can be attributed to the semiconductor case are the effective quantum state realized in the system, that can be, beyond the mere excited state of the quantum dot, an initial state prepared as a
photon or, even more likely, an arbitrary mixed state (a density matrix) of light and matter. This initial quantum state affects drastically the spectral shape [7; 8].

When pumping is increased, if the system is indeed described by the Jaynes-Cummings model, dressed states of the light-matter system are excited, with potential applications for quantum devices, since they are ruled by single-photon nonlinearities. As pumping is further increased, a transition into lasing is observed, that still maintains strong-coupling, and is therefore closely related to the one-atom laser. This transition is shown in Fig. 1 for the parameters of Nomura et al. (see also the animation lasing-exciton-pumping.mov). In qualitative agreement with the actual experiment, the Rabi doublet collapses, is followed by a narrow (lasing) line, that is ultimately broadened due to quenching by the incoherent pumping [9]. Direct and explicit manifestation of a few quanta is not directly apparent in case (i) with exciton pumping only. Carrying out a similar experiment but increasing cavity pumping instead, is more prone to give away the nonlinear features (see the animation lasing-cavity-pumping.mov). Such nonlinear features, if they are of a quantum character, should exhibit the characteristic signature of a dressed states square-root splitting, resulting in resonances at $\pm (\sqrt{n} \pm \sqrt{n-1})$ (with $n$ the number of excitations in the system). This is strikingly manifest in system (iii) (Fig. 1, see also the animation jc-mollow-transitions.mov). In such a case, one must observe the system in a suitable rescaled framework (e.g., linewidth are vanishing). The results are plotted in log-scale on the figure (the Rabi doublet sets the scale). A neat transition from the vacuum Rabi doublet (two peaks) into quenching (Lorentzian line) is observed. It goes through the quantum regime with multiplets at anharmonic frequencies, and the classical regime with a Mollow triplet that results from melting together the distinguishable transitions between the quantum dressed states. For the case of point (ii), we provide a more detailed picture by supplementing the spectral shape evolution, with the main observables of interest (see also the animation realistic-jc-structure.mov). These are the cavity population $n_a$, the probability of the quantum dot to be in its excited state $n_\sigma$ and the two-photon counting probability at zero delay $g^{(2)}(0)$. We also plot the trace of $\rho^2$ ($\rho$ being the total density matrix), that gives the order of coherence (or purity) of the system, and the probabilities $p_{g(e)}(n)$ of the system to be found with $n$ photons dressing the ground (excited) state (stacked on top of each other in the figure, so that the total height is their sum, that represents the cavity photon statistics).

**Challenges for experimentalists and theoreticians:**

We list a short series of goals that we regard as open, realistic given currently available systems and important results, whether obtained or infirmed. The first three “challenges” are experimental and the last two theoretical:

(i) Alter the effective quantum state realized in the system.
(ii) Compare cavity and direct exciton emission.
(iii) Evidence Jaynes-Cummings features in the spectral shape.
(iv) Solve exactly the Jaynes-Cummings model with incoherent pumping.
(v) Provide a statistical description of data.

(i) We already commented on the sensibility of the spectral shape on the effective quantum state of the system. Beside, the lasing of the strongly-coupled light-matter system bears some differences whether driven by the exciton pumping or by the cavity pumping (that we shall discuss somewhere else). For these two reasons, it is a desirable goal to be able to control the photon-like or exciton-like character of the state, ideally up to an arbitrary ratio of the photon versus the exciton component (both in the spontaneous emission and nonlinear regimes). A striking manifestation of this effect would be to unravel the inner Jaynes-Cummings peaks by bringing the system from exciton-like to photon-like.
Figure 1. An overall picture of the spectroscopy of microcavity quantum electrodynamics. In inset, positions of various systems. Photoluminescence spectra for increasing pumping from (for all purposes) vanishing to infinite are shown for, (i), a representative of the best currently available systems (as reported by Nomura et al. [5]), (ii), a realistic near-term device exhibiting explicitly nonlinear quantum features, and (iii), an unrealistically good system, probing directly the hamiltonian spectrum. Case (i) is in agreement with the experimental results. Case (iii)—displayed in log-scale—exhibits neatly transitions from vacuum (Rabi doublet) to quantum (Jaynes-Cummings peaks) and lasing (Mollow triplet) before being quenched by saturation of the dot. Case (ii) shows the counterpart of such a transition in a realistic system, as of today. Quadruplets are observed and a transition to lasing follows the emergence of two inner peaks, melting into a single line. Below each spectrum, the probability $p_g(e)$ of the state $|g(e), n\rangle$ [for ground(excited) state dressed by $n$ photons] is shown (see also the animation excitation-statistics.mov), showing the transition from a Fock state (with strong antibuching) to a coherent state and ultimately a thermal state for the cavity while the dot is maintained in its excited state. Cavity population, $n_a$, quantum dot population, $n_\sigma$, $g^{(2)}(0)$ and the trace of $\rho^2$ are also shown for cavity pumping, from $P_a/g = 0$ (blue) to 0.05 by steps of 1/180. All spectra are without cavity pumping.
(ii) In the linear regime, there is a symmetry between the effective quantum state and the channel of emission (through the cavity mode or the direct exciton emission). In the nonlinear regime, such an equivalence breaks down, and is replaced by richer spectroscopic structures. In particular, the direct exciton emission is usually more prone to exhibit quantum phenomena, in particular the Jaynes-Cummings ladder or the Mollow triplet (that is seen on Fig. 1 mainly because for such exceedingly good systems, one probes directly the Hamiltonian spectrum rather than the cavity or the exciton optical spectra). For this reason, it is desirable to be able to compare spectral shapes of both the cavity and direct exciton emission.

(iii) Obviously, a direct observation of the nonlinear Jaynes-Cummings structure would be an important achievement. It can be helped relying on the two previous points, or it could be probed by resonant excitation. If the coupling is dephased at a similar rate as the coherent light-matter coupling, the characteristic Jaynes-Cummings multiplet gives rise to a triplet [10]. Maybe Jaynes-Cummings nonlinearities have been already observed, in this disguise [11, 12].

(iv) One of the interesting feature of the Jaynes-Cummings model is that it admits analytical solutions. To the best of our knowledge, none is known yet in the presence of an incoherent pumping (results in Fig. 1 are obtained numerically). Beside the interest for its own sake, the exact solution would allow convenient optimization, e.g., to obtain the best antibunching to signal ratio. It would also explain features such as the existence of a fixed point for $n_\sigma(P_a, P_\sigma)$, a possible criterion to define lasing in such systems. Last but not least, if solutions can be found for two-time correlators, it would greatly help in fitting experimental data.

(v) So far, most reports have been based on qualitative features, such as observation of an anticrossing (but see Refs. [4] and [13] for some notable, and laudable, exceptions). Beside the possibility to be even qualitatively incorrect, this approach has the serious shortcoming of great inaccuracy, that impedes understanding and biases estimates of the systems and how far they are of reaching predicted features. We expect that the inset of Fig. 1 would change significantly if positioning was made after statistical inference rather than reading of linewidths in a Lorentzian fitting, or fitting of maxima (which is impossible [14]). A statistical analysis of the data, providing intervals of confidences for the fitting parameters, is not trivial in the nonlinear case, where conventional nonlinear regression methods do not apply, for lacking a closed-solution of the model. This would however demonstrate the possibility of such systems to be accounted for exactly (within standard deviates), and entitle the community to speak of microcavity quantum electrodynamics.

References

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Animations
Animations accompany this text as QuickTime movies.

Figure 2. lasing-exciton-pumping.mov (1.7MB): Cavity/exciton populations $n_a/\sigma$, two-photon coincidences $g^{(2)}(0)$ and cavity emission spectrum $S(\omega)$, with increasing exciton pumping, for the parameters of Nomura et al. (i).

Figure 3. realistic-jc-structure.mov (1.7MB): Same as Fig. 2 but for slightly more favorable parameters, (ii), evidencing explicitly the Jaynes-Cummings nonlinearities.

Figure 4. lasing-cavity-pumping.mov (255KB): Cavity emission spectrum for the parameters of Nomura et al. (i) when increasing cavity pumping.

Figure 5. jc-mollow-transition.mov (701KB): Cavity emission spectrum for a system in very strong coupling (iii) when increasing exciton pumping.

Figure 6. excitation-statistics.mov (551KB): Probabilities $p_{g(e)}(n)$ of a system in very strong coupling, (ii), to have $n$ photons dressing the ground (excited) state, when increasing exciton pumping.