Optical Spectra of the Jaynes-Cummings Ladder

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Abstract. We explore how the Jaynes-Cummings ladder transpires in the emitted spectra of a two-level system in strong coupling with a single mode of light. We focus on the case of very strong coupling, that would be achieved with systems of exceedingly good quality (very long lifetimes for both the emitter and the cavity). We consider the incoherent regime of excitation, that is realized with semiconductors quantum dots in microcavities, and discuss how reasonable is the understanding of the systems in terms of transitions between dressed states of the Jaynes-Cummings Hamiltonian.

Keywords: Jaynes-Cummings, Strong-Coupling, Microcavity, Quantum Dot, cavity QED, Spectroscopy

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INTRODUCTION

The crowning achievement of cavity Quantum Electrodynamics is the so-called Strong Coupling (SC), a regime dominated by quantum interactions between light (photons) and matter (an atom, or its semiconductor realization, a quantum dot (QD), sometimes called an artificial atom) [1]. In this regime, both the atom and the photon lose their individual identity, and vanish to give rise to new particles—sometimes called polaritons—with new properties. In this text, we shall be concerned with their optical spectral properties.

Strong light-matter coupling originated with atomic cavities [2, 3], and was later reproduced (among other systems) with semiconductor cavities [4], more promising for future technological applications (see for instance [5]). It is only recently, however (c. 2004), that this regime was reached for the zero-dimensional semiconductor case [6, 7, 8] and the number of reports has not been overwhelming ever since [9, 10, 11, 5]. In these systems, the QD excitation—the exciton—strongly couples to the single mode of a microcavity (realized as a pillar, a photonic crystal or other variants). As far as cavity QED is concerned, these systems are in principle superior to their two-dimensional counterpart (where SC is routinely achieved), because only a few excitations enter the problem in an environment with much reduced degrees of freedom, as opposed to planar polaritons whose most adequate description is in terms of continuous fields.

The landmark of SC is the Rabi doublet, where two modes (light and matter) at

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1 Here we must outline that we mean “quantum” in the sense of quantization of the fields, and thus breaking the classical picture. 2D polaritons have demonstrated stunning properties rooted in quantum physics, such as Bose-Einstein condensation or superfluid motion [12, 13]. However, those are manifestations of macroscopic coherence where large numbers of microscopic particles exhibit the behaviour of a continuous field (classical or not).
resonance, do not superimpose but split, each line corresponding to one of the polaritons that have overtaken the bare modes. It was early understood that this splitting is by itself, however, not a proof of quantization \[14\]. Parenthetically, 2D polaritons display most eloquently this splitting (see footnote 1).

Antibunching in the optical emission of a strongly coupled QD-microcavity system has been demonstrated \[9, 10\], further supporting quantization of the fields, but this is not completely conclusive, as although it proves that the dynamics involves a single quantum of excitation between two isolated modes (by itself already a considerable achievement), it does not instruct on the modes themselves (consider the vacuum Rabi problem of two harmonic oscillators, that gives the same result). After all, dimming classical light until single photons remain, would exhibit antibunching, but this says nothing about the emitter itself (which is ultimately quantum anyway; if it’s coming from the sun, say, it originates from the spontaneous emission of an atom, or to much lower probability, from stimulated emission).

A genuine, or quantum, SC \[15\], should culminate with a direct, explicit demonstration of quantization, with one quantum more or less changing the behaviour of the system. The most fundamental model to describe light (bosons)/matter (fermions) interactions is the celebrated Jaynes-Cummings (JC) Hamiltonian \[16\], where such a quantum sensitivity is strongly manifest, and has been observed more or less directly in various systems \[17, 18\]. Recently, direct spectroscopic evidence has been reported for atoms and superconducting circuits, in elaborate experiments \[19, 20\] that remind the heroic efforts of Lamb to reveal the splitting of the orbitals of hydrogen. Even more recently, very clear transitions from up to the fifth step of the ladder have been unambiguously observed in circuit QED in very strong-coupling, with the Rabi splitting more than 260 times the vacuum linewidth \[21\]!

With semiconductor QDs, there has been so far, to the best of our knowledge, no explicit demonstration of JC nonlinearities. In a previous work \[22\], we analyzed the peculiarities of these systems both with respect to their particular physics (involving a steady state under incoherent excitation rather than spontaneous emission or coherent excitation \[23\]) and their parameters. We concluded that even present-day structures could evidence anharmonicity of the quantized levels with particular pumping schemes and a careful analysis of the data. With much better structures but still conceivably realizable in the near future, strong qualitative signatures emerge and there is no need to go further than a simple observation of anharmonically spaced multiplets. Here we take the further step to go towards unrealistically good (as of today!) structures, with negligible exciton decay and quality factors orders of magnitude higher than state of the art system. In this regime of very strong coupling, one expects a priori Lorentzian emission of the dressed states \[24\]. We show with an exact quantum-optical computation of the spectra, the value and limitations of this approximation. Our results below can be seen as the ideal quantum limit of the Jaynes-Cummings model of light and matter, where the quantization of the fields appears in its full bloom.
THE JAYNES-CUMMINGS PHYSICS

The Jaynes-Cummings Hamiltonian is a textbook model, that admits essentially analytical solutions. It reads:

\[ H = \omega_a a^\dagger a + \omega_\sigma \sigma^\dagger \sigma + g (a^\dagger \sigma + a \sigma^\dagger). \]  \hfill (1)

Here, \( \omega_a, \sigma \) are the free energies for the modes \( a \) (boson) and \( \sigma \) (fermion) and \( g \) is their coupling strength. To describe realistically an experiment, one needs at least to include dissipation. Excitation is then typically assumed as an initial state, or with a coherent (Hamiltonian) pumping. To address the semiconductor case, we considered incoherent excitation \(^2\), i.e., a rate \( P_{\sigma} \) of QD excitation. The equation of motion for the density matrix \( \rho \) then reads \( \partial_t \rho = i[H, \rho] + \sum_{\sigma=a,\sigma} \gamma_c (2c^\dagger c \rho - c^\dagger c \rho - \rho c^\dagger c) + P_{\sigma}(2\sigma^\dagger \rho \sigma - \sigma \sigma^\dagger \rho - \rho \sigma \sigma^\dagger) \). So far we have not been able to provide an analytical solution for the steady state density matrix of this equation, but in the tradition of the JC, many exact results can nevertheless be extracted, for instance the eigenvalues of \( H \) in presence of dissipation, in Fig. 1(a). These give the energies of the states that are renormalized by the light-matter interaction, Eq. (1). For a given number \( n \) of excitations, there are two new eigenstates \( |n+\rangle \) and \( |n-\rangle \) that take over the bare modes \( |n \text{ photon}(s), 0 \text{ exciton}\rangle \) and \( |n-1 \text{ photon}(s), 1 \text{ exciton}\rangle \). The polaritons are, in their canonical sense, the states \( |1, \pm\rangle = (|10\rangle \pm |01\rangle)/\sqrt{2} \). For each \( n \), the two new states are splitted by \( 2\sqrt{n}g \). This is the main manifestation of quantization in the JC Hamiltonian: the difference from \( \sqrt{n} \) to \( \sqrt{n+1} \), when \( n \) is small, can be detected in a careful experiment. This structure of renormalized states is called the Jaynes-Cummings ladder. The transitions between its steps account for the lines that are observed in an optical luminescence spectrum. This generates the structure shown on Fig. 1(b).\(^2\)

The transitions between the same kind of dressed states (“same-states transitions”) of two adjacent steps (or manifolds, mathematically speaking), e.g., from \( |n+\rangle \) to \( |n-1, +\rangle \), emit at the energy \( \sqrt{n+1} + \sqrt{n} \) (in units of \( g \)), while the transitions between different kind of dressed states (“different-states transitions”) e.g., from \( |n+\rangle \) to \( |n-1, -\rangle \) emit at \( \sqrt{n+1} - \sqrt{n} \). Different-states transitions emit beyond the Rabi doublet while the same-states ones pack-up in between.

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To obtain the exact emission spectra of this system, one needs to compute two times correlators \( G_a^{(1)}(t, \tau) = \langle a(t+\tau)a(\tau) \rangle \) for the cavity emission and \( \langle \sigma(t+\tau)\sigma(\tau) \rangle \) for the direct exciton emission (in the steady state, the limit \( t \to \infty \) is taken). These two cases correspond to the geometry of detection in an ideal spectroscopic measurement: the cavity spectrum \( S_a(\omega) \) (the \( \tau \)-Fourier Transform of \( G_a^{(1)} \)) corresponds to detection of the cavity mode—this would be along the cavity axis of a pillar microcavity, for

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2 We also considered a rate \( P_\sigma \) of incoherent photon excitation, due to other dots or any other source populating the cavity. For more dissipative cases, this term bears a crucial importance on the spectral shapes. However in the very strong coupling, its role is of a less striking character and we therefore ignore it in the present discussion.

3 This is only the imaginary part. The real part, that corresponds to broadening of the transitions, is given in Ref. \(^{22}\).
FIGURE 1. The Jaynes-Cummings ladder (a) and the positions of peaks in its emitted spectrum (b), obtained from the difference in energy of transitions between any two branches of two consecutive “steps” (or manifolds) of the ladder. Without dissipation, the splitting of the \( n \)th step is \( 2\sqrt{n}g \) and transitions produce an infinite sequence of peaks at \( \pm\sqrt{n} \pm\sqrt{n+1} \), an infinitely countable number of them piling up towards 0 from above and below. With increasing \( |\gamma_a - \gamma_\sigma| \), the steps go one after the other into weak-coupling, producing a complex diagram of branch-coupling in the resonances of the emitted spectrum (b).

instance—while \( S_\sigma(\omega) \) (FT of \( G^{(1)}_\sigma \)) corresponds to direct emission of the exciton, like the top emission of a photonic crystal or the side of a pillar. We present both cases in the results below.

RESULTS

We consider very good cavities with \( g = 1 \) (defining the unit), \( \gamma_a \) of the order of \( 10^{-3} \), \( 10^{-2} \) and \( \gamma_\sigma = 0 \) (results are not significantly modified qualitatively for nonzero values of \( \gamma_\sigma \) of the order of \( \gamma_a \)). We consider small electronic pumping \( P_\sigma \) of the same order of \( \gamma_a \). At higher pumpings, the system goes into the classical regime, with lasing, very high populations, and continuous fields replacing discretization [22]. We therefore focus on small pumping rates, as we are interested in manifestations of quantization. Those are neatly displayed, as seen on Fig. 2 where very sharp (owing to the small decay rates) lines reconstruct the transitions of the JC ladder. In the plot of the cavity emission, Fig. 2(a), we have marked each peak with its corresponding transition between two quantized, dressed states of the JC hamiltonian. These peaks correspond one-to-one with those of the exciton emission, that are, however, weighted differently [22]. A simple argument explains the different weights in the intensity of the lines in the cavity emission \( I_a \) as compared to their counterpart in the exciton emission \( I_\sigma \):
\[ I_a^{(\pm \rightarrow \mp)} \propto |\langle n-1, \mp | a | n, \pm \rangle|^2 = |\sqrt{n} - \sqrt{n-1}|^2 / 4, \]  
(2a)

\[ I_a^{(\pm \rightarrow \pm)} \propto |\langle n-1, \pm | a | n, \pm \rangle|^2 = |\sqrt{n} + \sqrt{n-1}|^2 / 4, \]  
(2b)
i.e., intensity is enhanced for same-peak but smothered for different-peak transitions, while on the other hand, the QD emission is level regardless of the configuration:

\[ I_\sigma^{(\pm \rightarrow \mp)} \propto |\langle n-1, \mp | \sigma | n, \pm \rangle|^2 = 1 / 4, \]  
(3a)

\[ I_\sigma^{(\pm \rightarrow \pm)} \propto |\langle n-1, \pm | \sigma | n, \pm \rangle|^2 = 1 / 4. \]  
(3b)

In both cases, the Rabi doublet dominates strongly over the other peaks. In Fig. 2(a), for instance, the peaks at \pm 1 extend for about 9 times higher than is shown, and already the outer transitions are barely noticeable. This is because the pumping is small and so also the probability of having more than one photon in the cavity (it is in this configuration of about 10% to have 2 photons, see Fig. 5). One could spectrally resolve the window \([-g/2, g/2]\) over a long integration time and obtain the multiplet structure of nonlinear inner peaks, with spacings \{\sqrt{n+1} - \sqrt{n}, n > 1\} (in units of \(g\)), observing direct manifestation of single photons renormalizing the quantum field. Or one could increase pumping (as we do later) or use a cavity with smaller lifetime. In this case, less peaks of the JC transitions are observable because of broadenings mixing them together, dephasing and, again, reduced probabilities for the excited states, but the balance between them is better. In Fig. 3 where \(\gamma_a\) is now \(g/100\), the Rabi doublet (marked \(R\)) is dominated by the nonlinear inner peaks in the cavity emission, and a large sequence of peaks is resolved in the exciton emission.

Going back to the case of Fig. 2, but increasing pumping, we observe the effect of climbing higher the Jaynes-Cummings ladder. Results are shown in Fig. 4 in logarithmic scale, so that small features are magnified. First row is Fig. 2 again but in log-scale, so that the effect of this mathematical magnifying glass can be appreciated. Also, we plot over the wider range \([-15g, 10g]\). Note how the fourth outer peak, that was not visible on the linear scale, is now comfortably revealed with another three peaks at still higher energies. As pumping is increased, we observe that the strong linear Rabi doublet is receding behind nonlinear features, with more manifolds indeed being probed, with their corresponding transitions clearly observed (one can track up to the 19th manifold in the last row). This demonstrates obvious quantization in a system with a large number of photons. The distribution of photons in these three cases is given in Fig. 5 going from a thermal-like, mostly dominated by vacuum, distribution, to coherent-like, peaked distribution stabilizing a large number of particles in the system. At the same time, note the cumulative effect of all the side peaks from the higher manifolds excitations, absorbing all quantum transitions into a background that is building up shoulders, with the overall structure of a triplet. This is the mechanism through which the system bridges from a quantum to a classical system. The new spectra are reminiscent of the Mollow triplet of resonance fluorescence [25]. These are obtained both in the cavity and the exciton emission, but much more so in the latter. With more realistic parameters, it is indeed only visible in the exciton emission.
FIGURE 2. Fine structure of the “light-matter molecule”: emission spectra in the cavity (a) and direct exciton emission (b) of the strongly-coupled system with $(\gamma_a, \gamma_\sigma)/g = (10^{-3}, 0)$ at $P_\sigma/g = 10^{-3}$.

FIGURE 3. Same as Fig. 2 but now with $\gamma_a/g = 10^{-2}$. Less peaks are resolved because of broadening but nonlinear peaks (a, b) are neatly observable. In fact, now inner nonlinear peaks dominate in the cavity emission (the linear Rabi peaks are denoted $R$). In the exciton direct emission, the Rabi doublet remains the strongest.

In the very strong coupling limit, one could content with a basic understanding of the transitions mechanisms (given by Eqs. (2-3)) and the knowledge of the distribution of particles (cf. Fig. 5), to provide a fairly good account of the final result [24]. This is true for the most basic understanding of the transitions only, essentially valuable to identify peaks in terms of transitions in the JC ladder. Some features of the problem, like interferences, are missing completely. Observe for instance the sharp dip that is retained in all the exciton spectra. This is a quantum optical result that can be recovered only with an exact treatment of the emission. The qualitative result remains strong enough to support the underlying physics with little needs of these refinements. In the case of less ideal cavities, however, the evidence is not so strong for field-quantization, and both a proper description as well as an understanding of its specificities is then required to support one’s conclusions.
FIGURE 4. Spectra of emission in log-scales as a function of pumping $P_\sigma/g$, for $10^{-3}$ (upper row), $5 \times 10^{-3}$ (middle) and $10^{-2}$ (lower row).

FIGURE 5. Probability $p(n)$ of having $n$ photon(s) in the cavity, for the three cases shown on Fig. 4. Quite independently of the distribution of photon numbers in the cavity, field-quantization is obvious.
CONCLUSION

We have overviewed the problematic of nonlinearities in optical spectra as it is posed by cavity QED, where both fields (atomic and photonic) are quantized. We explored the very strong coupling regime in a system with very small decay rates. We confirm with an exact quantum optical treatment that a qualitative picture in terms of transitions in the Jaynes-Cummings ladder reasonably accounts for the observations. The successful observations of full quantization of the light-matter field merely require high experimental sensitivity and very good samples. In less ideal conditions, a finer analysis is required, as was discussed elsewhere [22].

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