Strong Coupling of Quantum Dots in Microcavities

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We show that strong coupling (SC) of light and matter as it is realized with quantum dots in microcavities differs substantially from the paradigm of atoms in optical cavities. The type of pumping used in semiconductors yields new criteria to achieve SC, with situations where the pump hinders, or on the contrary, favors it. We analyze one of the seminal experimental observation of SC of a quantum dot in a pillar microcavity [Reithmaier *et al.*, Nature (London) **432**, 197 (2004)] as an illustration of our main statements.

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The so-called *strong coupling* (SC) regime occurs in a coupled system where the interaction strength overcomes the losses. A case of fundamental interest is that of light (photons) and matter (atoms, electrons, etc.), coupled by the electromagnetic force. That this coupling is usually so weak accounts for the tremendous success of QED, which affords all the required accuracy at the level of perturbation theory. By confining the emitter in a cavity, repeated interactions with the trapped photon(s) occur and SC can thus be obtained, as was demonstrated in pioneering experiments with Rydberg atoms migrating in optical cavities [1]. Every achievement with atoms becomes an objective for semiconductors, which offer unique advantages in terms of integration and scalability, but come with their disadvantages in the form of the overall complexity brought by condensed matter over its fundamental elements. SC was first reported in semiconductors with quantum wells in planar microcavities [2], which launched a new field investigating exotic phases of matter in condensed systems. There is not yet a general consensus that quantum physics rules these systems and that nonlinear optics could not equally or better account for the observed phenomena. A more exact analogue to the atomic case that sticks closer to the quantum regime is provided by zero dimensional (0D) structures—quantum dots (QDs)—where the material excitations—the excitons—are fully quantized. SC in such systems was only recently realized [3–5]. Figure 1 reproduces one of these seminal contributions, by Reithmaier *et al.* [3], with QDs in micropillars. The most striking feature of SC is the splitting of the spectral shape when the system is at resonance: the line of the cavity and that of the emitter, both at the same frequency, do not superimpose but anticross with a splitting related to the coupling strength. The observation of a doublet in Fig. 1 when the two modes are expected to be resonant is the central result of Ref. [3] and of the related works [4,5]. In the ample literature devoted to the description of this spectral shape [6–10], the seminal work of Ref. [6] paid little attention to the excitation (using a coherent state as an initial condition) and neglected dissipation, while works such as [7,9] addressed the case of coherent pumping. Reference [8] described spontaneous emission of an excited state, and thus the initial state was fixed to be the excited state of the atom in an empty cavity. This was repeated in the theoretical work addressing the semiconductor case [10], where, however, a more complicated dynamics enters the picture. In practice, it is not possible to initialize the cavity-emitter system in a semiconductor as it is in the atomic case, where atoms can be singled out, manipulated, and sent one by one into the cavity. Semiconductor QDs in a cavity are typically excited far above resonance and electron-hole pairs relax incoherently to excite the QD in a continuous flow of excitations, establishing a steady state that washes out the coherent Rabi oscillations. Therefore, a Fock state as an initial state does not correspond to the experimental reality. Instead, the system is maintained in a mixed state with probabilities $p(n)$ to realize the nth excited state, which is imposed by the experimental configuration.

In this Letter, we provide the appropriate theoretical model to describe SC with 0D semiconductors. For the sake of illustration, we support our discussion with the

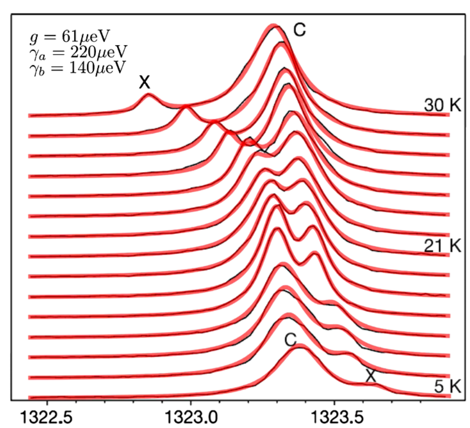

FIG. 1 (color online). Anticrossing of the cavity (C) and exciton (X) photoluminescence lines as reported by Reithmaier *et al.* [3], demonstrating SC in their system. Energies are given in meV. The gray (red) lines are our superimposed fits with the best global fit parameters in the top left corner. Such a good agreement cannot be obtained neglecting pumping.

results of Fig. 1, which our model reproduces with excellent agreement. On the contrary, other models, with their particular initial condition [6,8,10], cannot account for these spectra beyond the mere prediction of the line splitting. The shortcoming of downplaying the importance of the quantum state that is realized in the system owing to pumping has as its worst consequence a misunderstanding of the results, the most likely being the qualification of weak coupling (WC) for a system in SC that cannot be spectrally resolved because of decoherence-induced broadening of the lines. Being blind to the theory makes the track for SC in 0D semiconductors particularly difficult, involving a strong element of chance. Understanding the excitation scheme drastically reduces this element of hazard, as we shall see below. Most importantly, our model unravels the physics behind the experimental result, by spelling out which quantum state has been produced, by providing most-likelihood estimators of the sample parameters, by distinguishing the Bose or Fermi-like character of its excitations, and by predicting results as the excitation is changed, most interestingly, as the pump is increased and the system is brought into the nonlinear regime.

We describe the system with a quantum dissipative master equation $\partial_t\rho = \mathcal{L}\rho$ for the density matrix ρ [11], with the Liouvillian $\mathcal{L}$ defined by its action on any operator O of the tensor product of the light and matter Hilbert spaces $\mathcal{H}_a$ and $\mathcal{H}_b$:

$$\mathcal{L}O = i[O, \omega_a a^\dagger a + \omega_b b^\dagger b + g(a^\dagger b + a b^\dagger)] + \sum_{c=a,b}\Big(\frac{\gamma_c}{2}(cOc^\dagger - c^\dagger c O) + \frac{P_c}{2}(c^\dagger O c - c c^\dagger O) + \text{H.c.}\Big), \quad (1)$$

where g is the interaction strength between the cavity mode—with annihilation operator a at energy ω_a—and the material excitation—with operator b at energy ω_b—and respective decay and pumping rates $\gamma_{a,b}$ and $P_{a,b}$. An important experimental parameter is the detuning between the bare modes, $\Delta = \omega_a - \omega_b$, that can be tuned with temperature. In our case, where the interplay of pumping and dissipation establishes a steady state, the system is ergodic and the cavity emission spectrum follows from the Wiener-Khintchine theorem as $S(\omega) \propto \lim_{t\to\infty} \mathrm{Re}\int_0^\infty \langle a^\dagger(t) a(t+\tau)\rangle e^{i\omega\tau} d\tau$. According to the quantum regression theorem, a set of operators $A_{\{\alpha\}}$ that satisfies $\mathrm{Tr}(A_{\{\alpha\}}\mathcal{L}O) = \sum_{\{\beta\}} M_{\{\alpha\beta\}}\mathrm{Tr}(A_{\{\beta\}}O)$ for all $O \in \mathcal{H}_a \otimes \mathcal{H}_b$ for some $M_{\{\alpha\beta\}}$ yields the equations of motion for the two-time correlators as $\partial_\tau\langle O(t)A_{\{\alpha\}}(t+\tau)\rangle = \sum_{\{\beta\}} M_{\{\alpha\beta\}}\langle O(t)A_{\{\alpha\}}(t+\tau)\rangle$. If b is a Bose operator like a, M is defined by $M_{\substack{nm\\nm}} = -i(n\omega_a + m\omega_b) - n\Gamma_a/2 - m\Gamma_b/2$, $M_{\substack{nm\\n+1,m-1}} = M_{\substack{mn\\m-1,n+1}} = -igm$ and zero otherwise, where we defined as a shortcut the *effective broadenings* $\Gamma_{a,b} = \gamma_{a,b} - P_{a,b}$. We also introduce the following notation:

$$\gamma_\pm = (\gamma_a \pm \gamma_b)/4 \quad \text{and} \quad \Gamma_\pm = (\Gamma_a \pm \Gamma_b)/4. \quad (2)$$

Instead of *ad hoc* initial conditions for the cavity population and off-diagonal coherence, such as those provided by the excited state of the QD [8,10], we use the steady-state values obtained by solving $\mathrm{Tr}(a^\dagger a\mathcal{L}\rho) = 0$ and $\mathrm{Tr}(a^\dagger b\mathcal{L}\rho) = 0$. For instance, the steady-state cavity population $n_a = \lim_{t\to\infty}\langle a^\dagger a\rangle(t)$, reads

$$n_a = \frac{g^2\Gamma_+(P_a + P_b) + P_a\Gamma_b[\Gamma_+^2 + (\frac{\Delta}{2})^2]}{4g^2\Gamma_+^2 + \Gamma_a\Gamma_b[\Gamma_+^2 + (\frac{\Delta}{2})^2]}. \quad (3)$$

The equations are closed and the normalized photoluminescence (PL) spectrum $S(\omega)$ can therefore be expressed analytically:

$$S(\omega) = (\mathcal{L}_1 + \mathcal{L}_2) - \mathrm{Re}(\mathcal{C})(\mathcal{A}_1 - \mathcal{A}_2) - \mathrm{Im}(\mathcal{C})(\mathcal{L}_1 - \mathcal{L}_2), \quad (4)$$

defined in terms of the Lorentzian $\mathcal{L}$ and dispersive $\mathcal{A}$ functions that characterize the emission of the lower (1) and upper (2) eigenstates (*dressed states*):

$$\mathcal{L}_{\substack{1\\2}}(\omega) = \frac{1}{2\pi}\frac{\Gamma_+ \pm \mathrm{Im}(R)}{[\Gamma_+ \pm \mathrm{Im}(R)]^2 + \{\omega - [\omega_a - \frac{\Delta}{2} \mp \mathrm{Re}(R)]\}^2}, \quad (5)$$

$$\mathcal{A}_{\substack{1\\2}}(\omega) = \frac{1}{2\pi}\frac{\omega - [\omega_a - \frac{\Delta}{2} \mp \mathrm{Re}(R)]}{[\Gamma_+ \pm \mathrm{Im}(R)]^2 + \{\omega - [\omega_a - \frac{\Delta}{2} \mp \mathrm{Re}(R)]\}^2}, \quad (6)$$

where the complex coefficient $\mathcal{C}$ is defined as

$$\mathcal{C} = \frac{1}{R}\Big[\Gamma_- + i\frac{\Delta}{2} + \frac{i\frac{g^2}{2}(\gamma_a P_b - \gamma_b P_a)(i\Gamma_+ - \frac{\Delta}{2})}{g^2\Gamma_+(P_a + P_b) + P_a\Gamma_b[\Gamma_+^2 + (\frac{\Delta}{2})^2]}\Big] \quad (7)$$

and the complex *Rabi splitting* as

$$R = \sqrt{g^2 - \Big(\Gamma_- + i\frac{\Delta}{2}\Big)^2}. \quad (8)$$

The spectral shape given by formulas (4)–(8) is that of a coupled (SC and WC) system in a steady state maintained by incoherent pumping with rates $P_{a,b}$, with the QD excitation obeying Bose statistics. This is a valid approximation when the QD is large (it becomes exact in the limit of a quantum well) or in any case when the number of excitations is vanishing. If the QD follows Fermi statistics, the analytical expression for the spectrum is lost. We therefore keep this case out of the present discussion. In the case of Ref. [3], both large QDs and low excitations were used, and we verified numerically that a fermionic model is less appropriate. The basic structure of Eq. (4) is the same as in other descriptions of SC, such as the decay of an initial state. Namely it consists of two peaks, each the sum of a Lorentzian and of a dispersive part. The limit of vanishing pumping is formally equal to the particular case of the decay of an initial state with independent initial populations, whose ratio is the same as that of the pumping rates. The main result is to be found in the way incoherent pumping, even if it is small, affects this intrinsic structure of SC through Eqs. (7) and (8). This is demonstrated by confronting the theory with the experiment. In Fig. 1, we have optimized the *global* nonlinear fit of the results from Ref. [3] with Eqs. (4)–(8). That is, the detuning (ω_a and ω_b) and pumping rates (P_a and P_b) are the fitting parameter from one curve to the other, while g and $\gamma_{a,b}$ have been optimized but kept constant for all curves. We find an excellent overall agreement, that is instructive of many hidden details of the experiment.

First, the model provides more reliable estimates of the fitting parameters than a direct reading of the line splitting at resonance or of the linewidths far from resonance: The best-fitting coupling constant is $g = 61\ \mu\text{eV}$. The value for $\gamma_a = 220\ \mu\text{eV}$ is consistent with the experiment [12], and the value for γ_b, which is the most difficult to estimate experimentally, is reasonable in the assumption of large QDs, as is the case of those that have been used to benefit from their large coupling strength. Our point here is not to conduct an accurate statistical analysis of this particular work but to show the excellent agreement that is afforded by our model with one of the paradigmatic experiments in the field. Such a good global fit cannot be obtained without taking into account the effect of pumping, even when it is small. More interestingly, it is necessary to include both the exciton pumping P_b (expected from the experimental configuration) and also the cavity pumping P_a. The latter requirement comes from the fact that in such samples there are numerous QDs weakly coupled to the cavity in addition to the one that undergoes SC. Beyond this QD of interest, a whole population of "spectator" dots contributes an effective cavity pumping, which looms up in the model as a nonzero P_a. The fitting pumping rates vary slightly with detuning, as can be explained by the change in the effective coupling of both the strongly coupled dot with the cavity (pumping tends to increase out of resonance) and the spectator QDs that drift in energy with detuning. We find as best fit parameters at resonance $P_a \approx 0.12\gamma_a$ and $P_b \approx 0.18\gamma_b$ (the mean over all curves is $\bar{P}_a \approx 0.15\gamma_a$ and $\bar{P}_b \approx 0.28\gamma_b$ with rms deviations of $\approx 10\%$). The existence of P_a in an experiment with electronic pumping is supported by the authors of [3] who observed a strong cavity emission with no QD at resonance. We shall see in the following the considerable importance of this fact to explain the success of their experiment.

From a fundamental point of view, our incoherent pumping model of SC not only fills in a gap in extending the theory to the steady-state case where the excitation is not given (sometimes arbitrarily) as an initial state, it also defines new criteria for SC. The conventional one, from the condition that R be real at resonance, is, neglecting pumping,

$$g > |\gamma_-|. \tag{9}$$

With incoherent pumping, it becomes

$$g > |\Gamma_-|. \tag{10}$$

The full extent of this new criterion can be appreciated in Fig. 2, where it is displayed in shades of blue the region where R is real and in shades of red where it is pure imaginary. This corresponds, respectively, to oscillations or not in the time correlators and therefore to oscillations (SC) or damping (WC) of the fields. In white (delimited by the red frontier) is the region where there is no steady state because of a too-high pumping. The dashed black line delimits the conventional (without incoherent pumping) criterion, Eq. (9). Regions 3 and 4 show how pumping can make a qualitative difference. In region 3, given by $|\Gamma_-| < g < |\gamma_-|$, SC is not expected according to Eq. (9), but holds thanks to pumping, Eq. (10) (in this case, thanks to cavity pumping P_a; in inset, P_a is set to zero and this region has disappeared). In region 4, given by $|\gamma_-| < g < |\Gamma_-|$, where on the contrary SC is expected according to Eq. (9), it is lost because of pumping. In regions 1 and 5, the effect of the pump is quantitative only, renormalizing the broadening and splitting of the peaks, but is still important to provide a numerical agreement with experimental data. Region 2 is that where, although in SC, only one peak is observed in the PL spectrum because of the

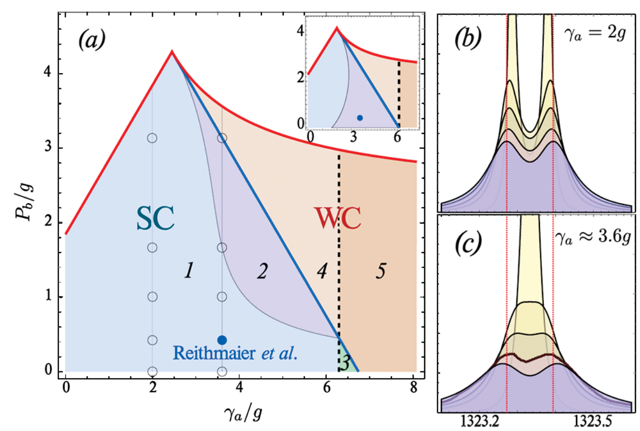

FIG. 2 (color). (a) Regions of strong (blue) and weak (red) coupling at resonance in the space of parameters (γ_a, P_b), with $\gamma_b \approx 2.3g$ and $P_a \approx 0.12\gamma_a$ fitting the experiment of Reithmaier *et al.* [3], marked by a plain blue point. Region 1 exhibits line splitting, while in the darker area 2, although still in SC, the splitting cannot be resolved. The dashed vertical line marks the criterion for SC in absence of pumping, giving rise to region 3, where SC is recovered (with line splitting) thanks to pumping, and region 4, where it is lost because of it. In the inset, the same but for $P_a = 0$, in which case the line splitting of [3] would not be resolved. (b),(c) Spectra of emission with increasing exciton pumping P_b marked by the hollow points in (a). For $\gamma_a = 2g$ in (b), SC is retained throughout and made more visible. For the best fit parameter, $\gamma_a \approx 3.6g$ in (c), line splitting is lost increasing pumping, first because it is not resolved [region 2 of (a)], then because the system goes into WC [region 4 of (a)].

broadening of each peak being too important as compared to their splitting. The position where we estimate the result of Ref. [3] in this diagram validates that SC has indeed been observed in this experiment. In the inset, however, one sees that in the case where the cavity pumping P_a is set to zero keeping all other parameters the same, the point falls in the dark region 2 where, although still in SC, the line splitting cannot be resolved. Even if it is possible in principle to demonstrate SC through a finer analysis of the crossing of the lines, it is obviously less appealing than a demonstration of their anticrossing. This is despite the fact that the case of $P_a = 0$ is equally, if not more, relevant as far as SC is concerned, as it corresponds to the case where only the QD is excited, whereas in the case of Fig. 1, it also relies on cavity photons. With the populations involved in the case of the best fit parameters that we propose—$n_a \approx 0.15$ from Eq. (3)—one can still read in the Reithmaier *et al.* experiment a good *vacuum* Rabi splitting, so the appearance of the line splitting with P_a is not due to the photon-field intensity. Rather, the system is maintained in a quantum state that is more photonlike in character, which is more prone to display line splitting in the cavity emission with the parameters of Fig. 1. One can indeed check that without pumping the spontaneous emission of the state prepared as a photon exhibits a line splitting and does not if prepared as an exciton. This is the same principle that applies here, with the nature of the state (photonlike or excitonlike) resolved self-consistently by pumping. In this sense, there is indeed an element of chance involved in the SC observation, as one sample can fall in or out of region 2 depending on whether or not the pumping scheme is forcing photonlike states.

A natural experiment to build upon our results is to tune pumping. In our interpretation, it is straightforward experimentally to change P_b, but it is not clear how P_a would then be affected, as it is due to the influence of the crowd of spectator QDs, not directly involved in SC. In Fig. 2, we hold P_a to its best fit parameters and vary P_b in two cases: the best fit case $\gamma_a \approx 3.6g$ in 2(c), and $\gamma_a = 2g$ in 2(b), where the system is in SC for all possible values of P_b. Spectra are displayed for the values of P_b marked with points in 2(a). Two very different behaviors are observed for two systems varying slightly in one of their parameters. In 2(b), strong renormalization of the linewidths and splitting results from Bose effects in a system that retains SC throughout. In 2(c), line splitting is lost and transition towards WC then follows. At high pumping, the model breaks down. The most interesting possibility is that the QD becomes Fermi-like. This case also results in the loss of line splitting, but with a smaller decrease in the linewidth at moderate pumpings and a subsequent increase at higher pumpings in the self-quenching regime. On the other hand, in the boson case, linewidths tend to zero as the populations diverge when effects such as particles interaction are neglected. A careful study of pump-dependent PL can tell much about the underlying statistics of the excitons and the precise location of one experiment in the SC diagram.

In conclusion, we obtained a self-consistent analytical expression for the spectrum of emission of a coupled light-matter system in a steady state maintained by a continuous incoherent pumping. Our formalism fully takes into account the effects of the incoherent pump, that, by randomizing the arrival time of the excitation, averages out the Rabi oscillations. The interplay of pumping and decay imposes a quantum steady state that influences considerably the observed spectra. Close to its threshold, SC can be lost or achieved by increasing pumping. Even when SC holds, the decoherence can hinder line splitting. Prospects for applications of SC with semiconductor heterostructures are great, provided that a quantitative understanding of the system can guide the advances now that the qualitative effects have been observed. Our model offers such a fine theoretical description, while still staying at a fundamental level with a transparent physical interpretation. We showed that the experimental reports of SC can be fully explained taking into account conjectures such as the influence of weakly coupled dots and other similar factors that can be better taken advantage of in the future.

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