Strong-coupling of quantum dots in microcavities

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Abstract

We show that strong-coupling (SC) of light and matter as it is realized with quantum dots (QDs) 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, favours it. We analyze one of the seminal experimental observation of SC of a QD in a pillar microcavity [Reithmaier et al., Nature (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 quantum electrodynamics (QED), that 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 strong-coupling 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, that 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. Strong-coupling was first reported in semiconductors with quantum wells in planar microcavities [2], that launched a new field investigating exotic phases of matter in condensed systems such as Bose-Einstein condensates [3] and superfluids [4]. 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) heterostructures—quantum dots (QDs)—where the material excitations—the excitons—are fully quantised. SC in such systems was only recently realized [5, 6, 7]. Fig. 1 reproduces one of these seminal contributions, by Reithmaier et al. [5], with QDs in micro-pillars. The most striking feature of strong coupling 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 doublet observed in Fig. 1 at 21K, when the two modes are expected to be resonant, demonstrates that they are strongly-coupled, and this observation is the central result of Ref. [5] and of the related works [6, 7]. In the ample literature devoted to the description of this spectral shape [8, 9, 10, 11, 12], the seminal work of Ref. [8] paid little attention to the excitation (using a coherent state as an initial condition) and neglected dissipation, while works such as [9, 11] addressed the case of coherent pumping. Ref. [10] 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 [12], where, however, a more complicated dynamics enters the picture. In practice, it is not possible to initialise
FIG. 1: Anticrossing of the cavity (C) and exciton (X) photoluminescence lines as reported by Reithmaier et al. in Nature (2004), demonstrating SC in their system. Energies are given in meV. The 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 the pump-induced decoherence.

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 \( n \)th excited state, that are 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 results of Fig. 1 that our model reproduces with an excellent agreement. On the contrary, other models, with their particular initial condition [8, 10, 12] cannot account for these spectra beyond the mere prediction of the line splitting. The shortcoming of downplaying the im-
portance 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 of 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 \( \rho \), 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 \( \mathcal{H}_b \):

\[
\mathcal{L} O = i [O, \omega_a a^\dagger a + \omega_b b^\dagger b + g(a^\dagger b + ab^\dagger)] + \sum_{c=a,b} \left( \frac{\gamma_c}{2} (cO c^\dagger - c^\dagger cO) + \frac{P_c}{2} (c^\dagger O c - cc^\dagger O) + \text{h.c.} \right),
\]

where \( g \) is the interaction strength between the cavity mode—with annihilation operator \( a \) at energy \( \omega_a \)—and the material excitation—with operator \( b \) at energy \( \omega_b \)—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 effectively by changing the 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 \( S(\omega) \propto \lim_{t \to \infty} \Re \int_0^\infty \langle a^\dagger(t)a(t+\tau) \rangle e^{i\omega \tau} d\tau \). According to the quantum regression theorem, a set of operators \( A_{\{\alpha\}} \) that satisfy \( \text{Tr}(A_{\{\alpha\}} \mathcal{L} O) = \sum_{\{\beta\}} M_{\{\alpha \beta\}} \text{Tr}(A_{\{\beta\}} O) \) 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 \( a \) (the photon operator), \( M \) is defined by \( M_{nm} = -i(n\omega_a + m\omega_b) - n\Gamma_a/2 - m\Gamma_b/2 \), \( M_{n+1,m-1} = M_{m-1,n+1} = -igm \) 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:

\[
\gamma_\pm = (\gamma_a \pm \gamma_b)/4 \quad \text{and} \quad \Gamma_\pm = (\Gamma_a \pm \Gamma_b)/4.
\]
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 [10, 12], we use the steady state values obtained by solving $\text{Tr}(a^\dagger a \mathcal{L}\rho) = 0$ and $\text{Tr}(a^\dagger b \mathcal{L}\rho) = 0$. For instance, the steady state cavity population $n_a = \lim_{t \to \infty} \langle a^\dagger a(t) \rangle$, reads:

$$n_a = \frac{g^2 \Gamma_+ (P_a + P_b) + P_a \Gamma_+ (\Gamma_+^2 + (\frac{\Delta}{2})^2)}{4g^2 \Gamma_+^2 + \Gamma_+ \Gamma_0 (\Gamma_+^2 + (\frac{\Delta}{2})^2)}.$$ (3)

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

$$S(\omega) = (\mathcal{L}_1 + \mathcal{L}_2) - \Re(C)(A_1 - A_2) - \Im(C)(\mathcal{L}_1 - \mathcal{L}_2),$$ (4)

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

$$\mathcal{L}_1(\omega) = \frac{1}{2\pi} \frac{\Gamma_+ \pm \Im(R)}{(\Gamma_+ \pm \Im(R))^2 + (\omega - (\omega_a - \frac{\Delta}{2} \mp \Re(R)))^2},$$ (5)

$$A_1(\omega) = \frac{1}{2\pi} \frac{\omega - (\omega_a - \frac{\Delta}{2} \mp \Re(R))}{(\Gamma_+ \pm \Im(R))^2 + (\omega - (\omega_a - \frac{\Delta}{2} \mp \Re(R)))^2},$$ (6)

where the complex coefficient $C$ is defined as:

$$C = \frac{1}{R} \left[ \Gamma_- + i\frac{\Delta}{2} + \frac{i\frac{\Delta}{2}(\gamma_a P_b - \gamma_b P_a)(i\Gamma_+ - \frac{\Delta}{2})}{g^2 \Gamma_+ (P_a + P_b) + P_a \Gamma_0 (\Gamma_+^2 + (\frac{\Delta}{2})^2)} \right]$$ (7)

and the complex Rabi splitting as:

$$R = \sqrt{g^2 - \left(\Gamma_- + i\frac{\Delta}{2}\right)^2}.$$ (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. [5], both large QDs and low excitations were used, and we verified numerically that a Fermionic model is less appropriate. The basic structure of eqn (11) 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
FIG. 2: (a) Regions of strong (blue) and weak (red) coupling at resonance in the space of parameters \((\gamma_a, P_b)\), with \(\gamma_b \approx 2.3g\) and \(P_a \approx 0.12\gamma_a\) fitting the experiment of Reithmaier et al. [5], 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 inset, the same but for \(P_a = 0\), in which case the line-splitting of [5] would not be resolved. (b) and (c): Spectra of emission with increasing exciton pumping \(P_b\) marked by the hollow points in panel (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 the line-splitting is not resolved (region 2 of (a)) then because the system goes into WC (region 4 of (a)).

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 eqns (7–8). This is
demonstrated by confronting the theory with the experiment. In Fig. 1, we have optimised the global nonlinear fit of the results from Ref. [5] with eqns (4–8). That is, the detuning ($\omega_a$ and $\omega_b$) 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 optimised but kept constant for all curves. We find an excellent overall agreement, that instructs on many hidden details of the experiment.

First, the model provides more reliable estimations 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 (the authors place it at $180\mu\text{eV}$ but from a Lorentzian fit of the 5K curve in the assumption that the system is not strongly-coupled here, where our model shows this to be a poor approximation), and the value for $\gamma_b$, that 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 experiment 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) but 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 of 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 [5] 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 (some-
times 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:

\[
g > |\gamma_-|.
\]  

(9)

With incoherent pumping, it becomes:

\[
g > |\Gamma_-|.
\]  

(10)

The full extent of this new criterion can be appreciated in Fig. 2, where 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, eqn (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 eqn (9), but holds thanks to pumping, eqn (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 eqn (9), it is lost because of pumping. In the regions 1 and 5, the effect of the pump is quantitative only, renormalising 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 broadening of each peak being too important as compared to their splitting. The position where we estimate the result of Ref. [5] in this diagram validates that SC has indeed been observed in this experiment. In 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 eqn (3)—one can still read in Reithmaier et al.’s 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 photon-like 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 the PL spectrum without pumping for a strongly-coupled state prepared as a photon exhibits a line-splitting whereas the same system prepared as an exciton does not show it. This is the same principle that applies here, with the nature of the state (photon-like or exciton-like) 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 photon-like 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(c), we hold $P_a$ to its best fit parameters and vary $P_b$ in the best fit case $\gamma_a = 3.6g$ on panel (c), then for $\gamma_a = 2g$ on panel (b), where the system is in SC for all possible values of $P_b$. Spectra are displayed for the values of $P_b$ marked by the points in (a). Two very different behaviours are observed for two systems varying slightly in one of their parameters. In one case (b), strong renormalisation of the linewidths and splitting results from Bose effects in a system that retains SC throughout. In the other (c), line-splitting is lost and transition towards WC then follows. At very high pumping, the model breaks down. The most interesting possibility is that the QD becomes Fermi-like, in which case the annihilation operator $b$ should be a two-level system. Loss of line-splitting also results in this case with a smaller decrease in the linewidth than is observed here at moderate pumpings and a subsequent increase at higher pumpings, as the system reaches the self-quenching regime (when $b$ is a Bose operator, linewidths tend to zero as the populations diverge when effects assured to take place at high density, 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 randomising the arrival time of the excitation, averages out the Rabi oscil-
lations and through the interplay of pumping and decay rates, imposes a quantum steady state that influences considerably the observed spectra. Close to its threshold, SC can be lost or imposed by increasing pumping, and 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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