Anticrossing in the PL spectrum of light-matter coupling under incoherent continuous pumping

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Abstract

We compare the observed splitting in the PL spectrum of a strongly coupled light-matter system, with the splitting of its dressed modes. In the presence of non-negligible decoherence, the two may differ considerably. Whereas the dressed mode splitting has a simple expression, the observed splitting has no general analytical expression in terms of radicals of the system parameters.

Key words: quantum dots, microcavities, strong coupling, PL spectrum

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There is now a large number of reports of strong light-matter coupling in zero dimensional semiconductor systems (one quantum dot in a microcavity). The successful theoretical description of some of these experiments relies on properly taking into account dissipation and decoherence \cite{1}. In these approaches, light-matter coupling is described with the linear model Hamiltonian $H = \omega_a a^\dagger a + (\omega_a - \Delta)b^\dagger b + g(a^\dagger b + ab^\dagger)$ where $a$ and $b$ are the cavity photon and material excitation (bosonic) field operators, respectively, with bare mode energies $\omega_a$ and $\omega_a - \Delta$, coupled with strength $g$. The photoluminescence (PL) spectrum of the cavity emission, analyzed in the steady state (SS), is greatly affected by decoherence. On the one hand, the system loses photons and matter excitations at rates $\gamma_a$ and $\gamma_b$, respectively. On the other hand, it is driven by a cw off-resonant pumping or, equivalently, by a continuous electronic injection in the wetting layer. This is effectively modelled by two excitation rates, $P_a$ and $P_b$. The dynamics of the system is given by a standard master equation. In Ref. \cite{2}, we investigated in detail the SS dynamics and PL spectrum of this system and obtained general analytical

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expressions for the lineshapes:

\[ S(\omega) = \frac{1}{2\pi} \Re \left\{ \frac{i-W}{\omega-\Omega_-} + \frac{i+W}{\omega-\Omega_+} \right\} = \Re \{ A(\omega) \}, \]  

where \( A(\omega) \) is a complex function of the real frequency \( \omega \), with:

\[ \Omega_{\pm} = \omega_a - \Delta/2 - i\Gamma_{\pm} \pm R \in \mathbb{C} \quad \text{(complex frequencies)}, \]

\[ W = [\Gamma_- + i(\Delta/2 + gD)]/R \in \mathbb{C} \quad \text{(dimensionless)}, \]

\[ \Gamma_{\pm} = (\Gamma_a \pm \Gamma_b)/4 \in \mathbb{R}, \quad \Gamma_{a,b} = \gamma_{a,b} - P_{a,b} \in \mathbb{R} \quad \text{(decoherence rates)}, \]

\[ R = \sqrt{g^2 - (\Gamma_- + i\Delta/2)^2} \in \mathbb{C} \quad \text{(half-Rabi frequency)}, \]

\[ D = \frac{\langle a^\dagger b \rangle_{SS}}{\langle a^\dagger a \rangle_{SS}} = \frac{\gamma_a P_b - \gamma_b P_a (i\Gamma_- - \Delta/2)}{g^2 \Gamma_+ (P_a + P_b) + P_a \Gamma_b (\Gamma_-^2 + (\Delta/2)^2)} \in \mathbb{C} \quad \text{(dimensionless)}. \]

With these definitions, the strong coupling (SC) regime is achieved when the condition \(|\Gamma_-| < g\) is satisfied. In such a regime, the so-called dressed modes (DM) appear at the eigenfrequencies \( \Re \{\Omega_{\pm}\} \), overtaking bare modes (of the weak coupling (WC) regime) at the frequencies \( \omega_a \) and \( \omega_b - \Delta \).

The observed spectrum, on the other hand, results from contributions from the leading modes (bare in WC or dressed in SC), so its splitting naturally does not correspond to that of the DM. Emission of each mode is not Lorentzian but also has a dispersive part, that can result in an increase or decrease of the apparent splitting, or even in its appearance in weak coupling or disappearance in strong coupling. It is therefore crucial, experimentally, to have an expression for the observed splitting that can be directly compared with the observed data.

It is possible to fit the lineshapes with the expression (1). In this text, we focus on the alternative practice of fitting the maxima of these spectra, that yield the characteristic crossing or anticrossing behaviors. The advantages are in the simplicity (two lines rather than a whole series of curves) and the robustness against noise (a parasitic second dot shifts only slightly the maxima of the strongly-coupled system whereas its influence on the whole lineshape can be much more deleterious for the fitting, calling for outliers or similar techniques). We need, however, to consider the observed splitting, not the dressed mode splitting. We shall see however that this approach is hindered by the Abel-Ruffini theorem that prohibits solutions in terms of radicals of the parameters.

For this purpose, we consider the equation \( dS/d\omega = 0 \), that gives the local extrema of the spectrum. There exists either one or three real solu-
Figure 1: Dressed modes (blue) and observed PL splitting (red) as a function of detuning. We fix $\gamma_b = 0.1g$ and $P_a = 0.5g$. (a) $\gamma_a = 3.8g$ and $P_b = g$, (b) $\gamma_a = 3.8g$ and $P_b = 0.5g$, (c) $\gamma_a = 3.8g$ and $P_b = 0.1g$, and (d) $\gamma_a = 4.8g$ and $P_b = 0.1g$.

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expression for the observed splitting, given by, in both SC or WC:

\[ \Delta \omega_O = 2g \Re \left\{ \sqrt{\left( 1 + \frac{P_b}{P_a} \right)^2 - 4 \frac{\Gamma_+}{g} \left( \frac{P_b}{P_a} \frac{\Gamma_-}{g} \right) - \frac{P_b}{P_a} - \left( \frac{\Gamma_b}{2g} \right)^2} \right\}. \]  

(3)

This should be contrasted with the expression for the DM splitting:

\[ \Delta \omega_{DM} = 2 \Re \{ R(\Delta = 0) \} = 2g \Re \left\{ \sqrt{1 - \left( \frac{\Gamma_-}{g} \right)^2} \right\}. \]  

(4)

Our main statement in this text is that the counterpart of Eq. (3) at nonzero detuning does not exist in this form and should be computed numerically. This is in stark contrast with the DM case, Eq. (4), which always has an equally simple expression, that in general does not, however, describe accurately experimental data.

Expression (3) can be used to predict the pumping rates at which the splitting will be more visible for a given configuration. The counterpart expression for the splitting observed in the direct exciton emission can be obtained by simply exchanging indexes \( a \leftrightarrow b \). It is also interesting to note that, for the integrated spontaneous PL emission (instead of the SS emission), the formula for the observed splitting is found by only removing \( P_{a,b} \) from the \( \Gamma \)'s and substituting \( P_b/P_a \) by the ratio of initial state populations \( \langle b^\dagger b \rangle^0 / \langle a^\dagger a \rangle^0 \) (under the assumption that initially \( \langle a^\dagger b \rangle^0 = 0 \)).

In conclusion, we have contrasted the crossing and anticrossing of the PL spectrum lines as detuning is varied, with the actual behavior of the dressed modes, which truly quantify the strength of the light-matter coupling. We showed that a careful analysis is required to properly assess an experiment that is not in very strong coupling. No analytical formula in radicals of the parameters of the system exists in the general case, supporting the global fitting of lineshapes as the most convenient description of experimental data.

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References

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