

Cavity-QED assisted *attraction* between a cavity mode and an exciton mode in a planar photonic-crystal cavity

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Abstract: The photoluminescence spectra from a quantum-dot exciton weakly-coupled to a planar photonic-crystal cavity is experimentally investigated by temperature tuning. Significant resonance shifts of the cavity mode are observed as the cavity mode spectrally approaches that of the exciton mode, showing the appearance of cavity-to-exciton attraction or mode pulling. Cavity-mode spectral shifts are also found theoretically using a master equation model that includes incoherent pump processes for the coupled exciton and cavity, pure dephasing, and allows for photon emission via radiation modes and the leaky cavity mode. Both experiments and theory show clear cavity mode spectral shifts in the photoluminescence spectra, when certain coupling parameters are met. However, discrepancies between the experimental data and theory, including more pronounced spectral shifts in the measurements, indicate that other unknown mode-pulling effects may also be occurring.

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References and links

1. J. McKeever, A. Boca, A. D. Boozer, R. Miller, J. R. Buck, A. Kuzmich, and H. J. Kimble, "Deterministic generation of single photons from one atom trapped in a cavity," *Science* **303**, 1992 (2004).
2. A. Boca, A. D. Boozer, J. R. Buck, and H. J. Kimble, "Experimental realization of a one-atom laser in the regime of strong coupling," *Nature* **425**, 268 (2003).
3. E. Hagley, X. Matre, G. Nogues, C. Wunderlich, M. Brune, J. M. Raimond, and S. Haroche, "Generation of Einstein-Podolsky-Rosen pairs of atoms," *Phys. Rev. Lett.* **79**, 1 (1997).
4. E. Moreau, I. Robert, J. M. Gérard, I. Abram, L. Manin, and V. Thierry-Mieg, "Single-mode solid-state single photon source based on isolated quantum dots in pillar microcavities," *Appl. Phys. Lett.* **79**, 2865 (2001).
5. M. Pelton, C. Santori, J. Vučković, B. Zhang, G. S. Solomon, J. Plant, and Y. Yamamoto, "Efficient source of single photons: a single quantum dot in a micropost microcavity," *Phys. Rev. Lett.* **89**, 233602 (2002).
6. J. P. Reithmaier, G. Seogonk, A. Löffler, C. Hofmann, S. Kuhn, S. Reitzenstein, L. V. Keldysh, V. D. Kulakovskii, T. L. Reinecke, and A. Forchel, "Strong coupling in a single quantum dot semiconductor microcavity system," *Nature* **432**, 197 (2004).

7. P. Michler, A. Kiraz, L. Zhang, C. Becher, E. Hu, and A. A. Imamoğlu, "Laser emission from quantum dots in microdisk structures," *Appl. Phys. Lett.* **77**, 184 (2000).
8. E. Peter, P. Senellart, D. Martrou, A. Lemaître, J. Hours, J. M. Gérard, and J. Bloch, "Exciton-photon strong-coupling regime for a single quantum dot embedded in a microcavity," *Phys. Rev. Lett.* **95**, 067401 (2005).
9. T. Yoshie, A. Scherer, J. Hendrickson, G. Khitrova, H. M. Gibbs, G. Rupper, C. Ell, O. B. Shchekin, and D. G. Deppe, "Vacuum Rabi splitting with a single quantum dot in a photonic crystal nanocavity," *Nature* **432**, 200 (2004).
10. K. Hennessy, A. Badolato, M. Winger, A. Atatüre, S. Fält, E. L. Hu, and A. Imamoğlu, "Quantum nature of a strongly coupled single quantum dot-cavity system," *Nature* **445**, 896 (2007).
11. E. M. Purcell, "Spontaneous emission probabilities at radio frequencies," *Phys. Rev.* **69**, 681 (1946).
12. See, e.g., "Statistical Methods in Quantum Optics 2," H. J. Carmichael, Springer (2008).
13. M. Kaniber, A. Laucht, A. Neumann, J. M. Villas-Boas, M. Bichler, M.-C. Amann, and J. J. Finley, "Investigation of the nonresonant dot-cavity coupling in two-dimensional photonic crystal nanocavities," *Phys. Rev. B* **77**, 161303(R) (2008).
14. D. Press, S. Götzinger, S. Reitzenstein, C. Hofmann, A. Löffler, M. Kamp, A. Forchel, and Y. Yamamoto, "Photon antibunching from a single quantum-dot-microcavity system in the strong coupling regime," *Phys. Rev. Lett.* **98**, 117402 (2007).
15. S. Hughes and P. Yao, "Theory of the quantum nature of a strongly coupled single quantum dot cavity system," *Opt. Express* **17**, 3322 (2009).
16. G. Cui and M. G. Raymer, "Emission spectra and quantum efficiency of single-photon sources in the cavity-QED strong-coupling regime," *Phys. Rev. A* **73**, 053807 (2006).
17. A. Auffeves, B. Besga, J.-M. Gérard, and J.-P. Poizat, "Spontaneous emission spectrum of a two-level atom in a very-high-Q cavity," *Phys. Rev. A* **77**, 063833 (2008).
18. M. Yamaguchi, T. Asano, and S. Noda, "Photon emission by nanocavity-enhanced quantum anti-Zeno effect in solid-state cavity quantum-electrodynamics," *Opt. Express* **16**, 18067 (2008).
19. A. Naesby, T. Suhr, P. T. Kristensen, and J. Mørk, "Influence of pure dephasing on emission spectra from single photon sources," *Phys. Rev. A* **78**, 045802 (2008).
20. J. Suffczynski, A. Dousse, K. Gauthron, A. Lemaître, I. Sagnes, L. Lanco, J. Bloch, P. Voisin, and P. Senellart, "Origin of the optical emission within the cavity mode of coupled quantum dot-cavity systems," *Phys. Rev. Lett.* **103**, 027401 (2009).
21. T. Tawara, H. Kamada, S. Hughes, H. Okamoto, M. Notomi, and T. Sogawa, "Cavity mode emission in weakly coupled quantum dot - cavity systems," *Opt. Express* **17**, 6643 (2009).
22. M. Winger, T. Volz, G. Tarel, S. Portolan, A. Badolato, K. J. Hennessy, E. L. Hu, A. Beveratos, J. Finley, V. Savona, and A. Imamoğlu, Explanation of photon correlations in the far-off-resonance optical emission from a quantum-dot cavity system, *Phys. Rev. Lett.* **103**, 207403 (2009).
23. P. Yao, P. K. Pathak, E. Illes, S. Hughes, S. Münch, S. Reitzenstein, P. Franeck, A. Löffler, T. Heindel, S. Höfling, L. Worschech, and A. Forchel, "Nonlinear photoluminescence spectra from a quantum-dot-cavity system: Interplay between pump-induced stimulated emission and anharmonic cavity-QED," *Phys. Rev. B* (in press).
24. F. P. Laussy, E. del Valle, and C. Tejedor, "Strong coupling of quantum dots in microcavities," *Phys. Rev. Lett.* **101**, 083601 (2008).
25. A. Laucht, N. Hauke, J. M. Villas-Bôas, F. Hofbauer, G. Böhm, M. Kaniber, and J. J. Finley, "Dephasing of exciton polaritons in photoexcited InGaAs quantum dots in GaAs nanocavities" *Phys. Rev. Lett.* **103**, 087405 (2009).
26. A. Ridolfo, O. Di Stefano, S. Portolan, and S. Savasta, "Photoluminescence from microcavities strongly coupled to single quantum dots," unpublished.
27. E. Kuramochi, M. Notomi, S. Mitsugi, A. Shinya, and T. Tanabe, "Ultrahigh-Q photonic crystal nanocavities realized by the local width modulation of a line defect," *Appl. Phys. Lett.* **88**, 041112 (2006).
28. S. Mosor, J. Hendrickson, B. C. Richards, J. Sweet, G. Khitrova, H. M. Gibbs, T. Yoshie, A. Scherer, O. B. Shchekin, and D. G. Deppe, "Scanning a photonic crystal slab nanocavity by condensation of xenon," *Appl. Phys. Lett.* **87**, 141105 (2005).
29. L. Tian, H.J. Carmichael, "Incoherent excitation of the Jaynes-Cummings system," *Quantum Opt.* **4**, 131 (1992).
30. T. Takagahara, "Theory of exciton dephasing in semiconductor quantum dots," *Phys. Rev. B* **60**, 2638 (1999).
31. B. Krummheuer, V. M. Axt, and T. Kuhn "Theory of pure dephasing and the resulting absorption line shape in semiconductor quantum dots," *Phys. Rev. B* **65**, 195313 (2002).
32. P. Borri, W. Langbein, S. Schneider, U. Woggon, R. L. Sellin, D. Ouyang, and D. Bimberg, "Ultralong dephasing time in InGaAs quantum dots," *Phys. Rev. Lett.* **87**, 157401 (2001).
33. I. Wilson-Rae and A. Imamoğlu, "Quantum dot cavity-QED in the presence of strong electron-phonon interactions," *Phys. Rev. B* **65**, 235311 (2002).
34. F. Milde, A. Knorr, and S. Hughes, "Role of electron-phonon scattering on the vacuum Rabi splitting of a single-quantum dot and a photonic-crystal-nanocavity," *Phys. Rev. B* **78**, 035330 (2008).

1. Introduction

The study of light-matter interactions between an electronic two-level system and a quantized optical field gives rise to interesting regimes of light-matter interaction and has applications in quantum information science. The regime of cavity-quantum electrodynamics (cQED) has been intensively studied in atomic physics for several decades, and has found use for the creation of single photon sources [1], one atom lasers [2], and for demonstrating the violation of Bell's inequalities [3]. Motivated in part by the prospect of developing compact sources for quantum cryptography, the regime of *solid-state* cQED has also recently been carried out in various semiconductor systems, whereby a quantum-dot (QD) exciton serves as a single photon emitter within a suitably designed cavity, e.g. a micropillar [4–6], a microdisk [7, 8], or a planar photonic crystal cavity [9, 10]. The prominent feature of most of these cQED systems is the coupling of a single photon emitter (atom or QD) to a suitably large Q/V_m -ratio cavity, where Q is the cavity quality factor and V_m is the effective mode volume. In the weak-to-intermediate coupling regime, the photon emission rate is enhanced through the Purcell effect [11], and the strong coupling regime [12], vacuum Rabi splitting can be observed in the photoluminescence (PL) spectrum.

In the past few years, there have been several experimental reports on the off-resonant coupling between a single exciton and a cavity mode in semiconductor cavities. Hennessy *et al.* [10] reported on the off-resonant excitation of the cavity mode from an excited exciton using a photonic crystal - single QD system, and Kaniber *et al.* [13] also observe similar cavity coupling mechanisms, as do Press *et al.* [14] for micropost cavities. These works have posed the question of whether the usual simple atomlike models of the QD fail. However, modified theoretical models that account for the leaky nature of the cavity show that such couplings are expected for a planar PC cavity medium. Hughes and Yao [15] introduced a quantization procedure for any arbitrary inhomogeneous dielectric that was utilized to derive simple analytical spectral formulas in terms of the cavity mode emission and the radiation (exciton) mode emission; when used to calculate the QD emission from a typical PC cavity, the leaky cavity quasi-mode emission was found to completely dominate the detected spectra, whereby it contains both the *dressed* exciton resonance and the dressed cavity resonance. Leaky cavity mode spectra have also been reported by Cui and Raymer [16], Auffeves *et al.* [17], Yamaguchi *et al.* [18], and Naesby *et al.* [19], for simple cavities, where the cavity emission can be geometrically separated from the radiation emission. In the case of a planar PC medium, no geometrical separation of radiative decay and cavity decay is possible, and, in general, both exciton (via radiation modes) and cavity mode decay processes contribute to out-of-plane photon detection. Further experimental reports by Suffczynski *et al.* [20] support the model that fast exciton dephasing is responsible for the cavity mode feeding. Tawara *et al.* [21] also suggest that the bright cavity mode emissions with nonzero detuning may be influenced by radiative recombinations of deep-level defects in the barrier layers. Very recently, Winger *et al.* [22] propose that the QD confinement ensures the presence of a quasicontinuum of excitonic transitions, part of which overlaps with the cavity resonance; however, for our samples and excitation powers below, we see absolutely no evidence of a quasicontinuum and we still obtain a very clear cavity mode resonance over a wide range of spectral detunings. Thus, this subject matter is still under debate, and we have found that from a modeling perspective, a simple intuitive master equation approach is convenient to obtain significant off-resonant coupling over a wide spectral range for various QD - cavity systems.

In this work, we experimentally study the detuning dependence between a *weakly-coupled* exciton and a leaky cavity mode, and observe an unusual cQED spectral signature in the PL spectra: *cQED-assisted attraction between a cavity mode and an exciton mode*. This observation is persistent, and is observed in several different samples, and for several different exciton-

cavity couplings. To help explain this novel observation, we adopt a recent master equation model that includes fermion statistics and pure dephasing processes for the target exciton, as well as an incoherent pump terms for the target exciton and cavity mode [23]. This model is similar to previous master equation works applied to semiconductor cavities [24, 25], but also accounts for stimulated emission processes of the incoherent cavity pump [23, 26] and emission via radiation modes as well as the leaky cavity mode. Reasonable qualitative agreement between the experimental data and the theory is obtained, though further discrepancies suggest that other unknown effects may be occurring which lead to more pronounced mode pulling. Our results should help stimulate further theoretical developments, and systematic experimental measurements, in this rapidly growing field.

2. Experiment

A line-defect cavity with a local width modulation in a 2D planar photonic crystal [27] consists of a triangular lattice of air holes in a 200 nm-thick GaAs membrane containing a single InAs/InGaAs dot-in-well layer. The structural parameters of the photonic crystal include a lattice constant (a) of 315 nm and an air hole radius (r) of 81 nm. The air holes of the width modulation area in the line defect, which act as a cavity, were shifted by 6, 4 and 2 nm, respectively, outwards from their original positions. The cavity mode energy is around 1050 meV, and the mode volume of this cavity is about $2.0(\lambda/n)^3 = 0.09 \mu\text{m}^3$. The samples were mounted in

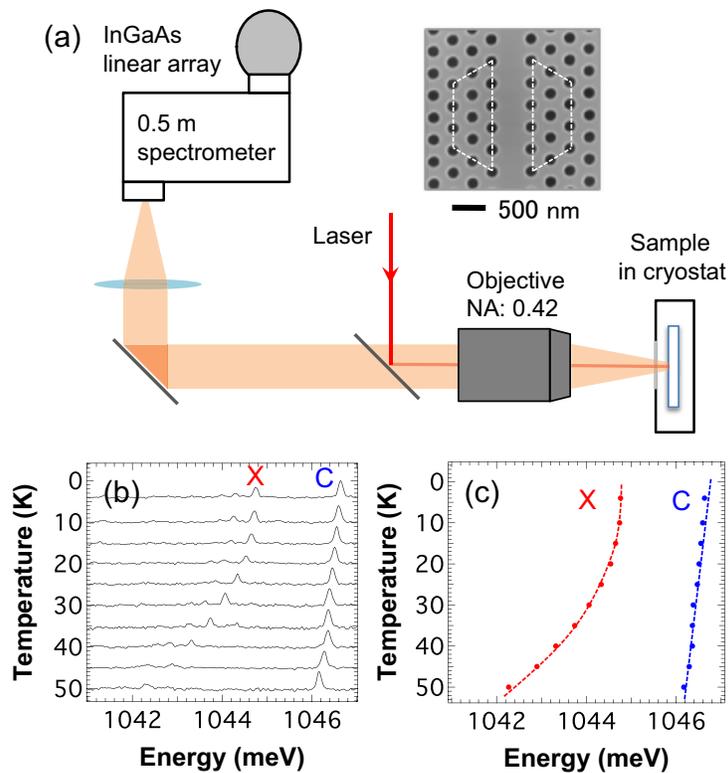


Fig. 1. (a) Schematic of the PL measurement setup and image of the fabricated cavity by scanning electron microscope (SEM). The area enclosed by the dotted line indicates the cavity region with width modulation. (b) PL spectra with temperature scanning and (c) peak plot for exciton 'X' and cavity 'C'. The dotted lines in (c) are guides to the eye.

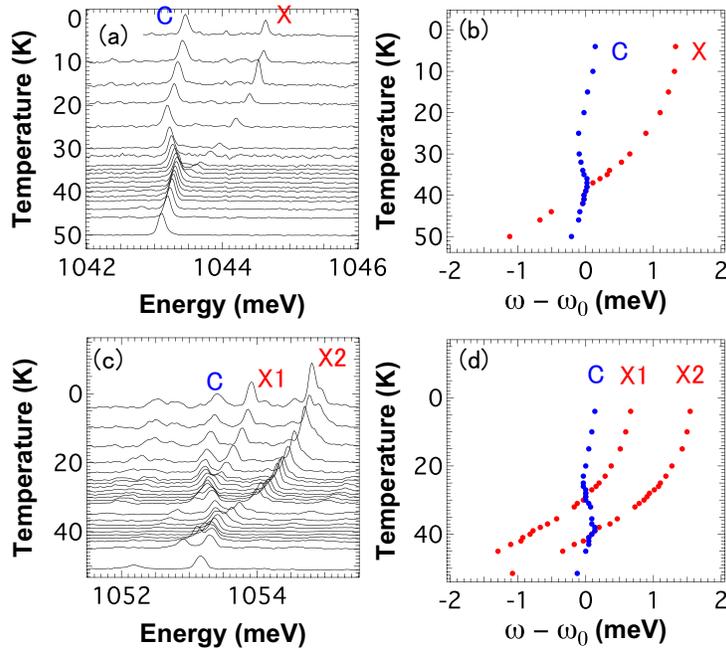


Fig. 2. Temperature dependent PL spectra and spectral peak plot of X and C for two different cavities with $\omega_x > \omega_c$ at 4 K. (a) Spectrum evolution, and (b) peak energies of sample one, and (c) and (d) show another set for a different sample. The crossover energy of the exciton and cavity, ω_0 , is 1043 meV for (a) and (b), and is 1053 meV for (c) and (d).

a continuous flow liquid-helium cryostat. Photoluminescence spectra were obtained by using an excitation source that consist of a YAG (1064 nm = 1.17 eV) continuous-wave laser. The excitation laser was focused to a spot with a $\sim 1 \mu\text{m}$ diameter through an objective lens with a numerical aperture of 0.42. We use the excitation power of $30 \mu\text{W}$, which is weak enough to excite no charging states of an exciton. We note that the contribution of the deep level emission from GaAs Barrier and InGaAs QW can likely be neglected under this excitation condition [21]. Figures 1(b) and (c) show the PL emission from the QD exciton and cavity mode detuned by temperature scanning (4-50 K) with no energy crossing ($\omega_x < \omega_c$). While the cavity-mode continuously redshifts at a rate of about $9 \mu\text{eV/K}$ according to temperature dependence of refractive index of GaAs slab, the exciton energy changes much faster because of the bandgap shift.

In Fig. 2, we display two examples of a temperature-scanning PL, showing temperature-evolutions of the cavity-exciton spectra and peak energies for two different samples, as panels (a) and (b), and (c) and (d). The photon energies are represented as detuning relative to the cavity-exciton crossover ω_0 . In these cavities a relation of $\omega_x > \omega_c$ applies at 4 K. As the temperature raises, these two peaks continuously redshift thereby the relative energy separations decrease. These cavities show no anti-crossing behavior of the vacuum field Rabi splitting near $\omega_x = \omega_c$, inferring that they are only weakly coupled, primarily because the QDs are not positioned at the anti-node positions of cavity fields. However, surprisingly, as the two peaks approach one another near the crossover, the cavity modes clearly blueshift toward the exciton resonances, which is opposite to the trend expected from Fig. 1(c), becoming *attracted* by the exciton resonances for small cavity-exciton detunings. This attraction can be easily distinguished from the normal energy shift by a sudden blueshift and overall temperature scanning

trends as shown in Figs. 1(b-c). We have observed similar *attraction* features in several other samples. A check was undertaken to exclude a probable unwanted gas deposition to the cavity, that may substantially shift the cavity resonance [28]. We raised the temperature up to room temperature once to evaporate the adsorbed gas species, recooled the sample to 4 K, and performed a similar experiment. It was found that every time we carried out a similar measurement, *exactly* the same results were obtained. Thus, the apparent attraction of the cavity-mode energy is not an experimental artifact, but is a universally observed phenomenon when specific exciton-cavity coupling and pump conditions meet.

Below we will introduce a master equation theory that uses the exciton and cavity decay rates, so here we briefly summarize the experimentally determined spectral widths: cavity linewidth $\Gamma_c = 0.10$ meV ($Q = 10\,000$), total exciton linewidth (including radiative and non-radiative contributions), $\Gamma_x^{\text{tot}} = 0.07$ meV (0.14 meV) at 4 K (35 K) [$\omega_0 = 1043$ meV] as obtained from Fig. 2(a) and (b), and $\Gamma_c = 0.18$ meV ($Q = 5800$), $\Gamma_{x1}^{\text{tot}} = 0.14$ meV (0.16 meV) at 4 K (23 K), and $\Gamma_{x2}^{\text{tot}} = 0.13$ meV (0.17 meV) at 4 K (43 K) ($\omega_0 = 1053$ meV) as from Figs. 2(c) and (d), all of which are represented as the full width at half maximum (FWHM). The total radiation lifetime of single QD excitons was determined in the sample (*with a substrate and before cavity fabrication*) by time-domain measurements to be 1.3 ns, which corresponds to a width of 3 μ eV in FWHM. The exciton-cavity coupling constant g is smaller than that expected to satisfy the strong-coupling condition, approximately when $g^2 > (\Gamma_c - \Gamma_x)^2/4$; however, as mentioned earlier, because the both QDs are not placed at anti-node positions of cavity fields, they are only in the weak- or the intermediate- coupling regime. In addition, our sample most likely includes several QDs per cavity system, and the background dots (through background excitons) can act to incoherently excite the cavity mode.

3. Theory

3.1. Formalism

For the theory, we adopt and extend a recent model of Yao *et al.* [23] applied to model pump-dependent strong coupling effects. The basic starting point is similar to the analytical model proposed by Laussy *et al.* [24], but with several notable differences: *i*) we include a thermal bath model for the reservoirs which ensures net positive densities for all pump powers [23]; *ii*) we apply fermion (rather than boson) statistics and include pure dephasing, similar to the work of Laucht *et al.* [25]; and *iii*), we include cavity decay and radiation-mode decay processes in calculating the total emitted spectra, both of which are important contributions for our planar PC cavity system.

Including an *incoherent* pump for exciting both the dot and the cavity, and using a thermal bath model for the Markovian reservoirs, the quantum dissipative master equation of the density matrix ρ is [29]

$$\frac{d\rho}{dt} = \frac{-i}{\hbar}[H_s, \rho] + \mathcal{L}(\rho), \quad (1)$$

where H_s is the system Hamiltonian given by $H_s = \hbar\omega_x\sigma^+\sigma^- + \hbar\omega_c a^\dagger a + \hbar g(\sigma^- a^\dagger + \sigma^+ a)$, and \mathcal{L} is the superoperator

$$\begin{aligned} \mathcal{L}(\rho) = & \frac{P_x}{2} (2\sigma^+\rho\sigma^- - \sigma^-\sigma^+\rho - \rho\sigma^-\sigma^+) + \frac{\Gamma_x + P_x}{2} (2\sigma^-\rho\sigma^+ - \sigma^+\sigma^-\rho - \rho\sigma^+\sigma^-) \\ & + \frac{P_c}{2} (2a^\dagger\rho a - aa^\dagger\rho - \rho aa^\dagger) + \frac{\Gamma_c + P_c}{2} (2a\rho a^\dagger - a^\dagger a\rho - \rho a^\dagger a) \\ & + \frac{\Gamma_x'}{4} (\sigma_z\rho\sigma_z - \rho), \end{aligned} \quad (2)$$

where $P_{x(c)}$ is the incoherent pump rate for *target* exciton (cavity) excitation, Γ_c is the decay rate of the leaky cavity mode, Γ_x is the *radiative* decay rate of the exciton, and Γ'_x is the pure dephasing rate of the exciton. The operators σ^+ , σ^- are the Pauli operators of the exciton (electron-hole pair), $\sigma_z = \sigma^+ \sigma^- - \sigma^- \sigma^+$, and a, a^\dagger are the cavity mode operators. The medium is therefore described in terms of a dominant cavity mode with a decay rate Γ_c and a resonance frequency ω_c (which exists deep inside the photonic bandgap), and the sum of background radiation modes above the light line.

The master equation approach includes dissipation, and necessarily goes beyond a simple dressed-state picture. Although this prohibits a simpler diagonalization of the system Hamiltonian to obtain the dressed-state eigenvalues, it does allow one to obtain the spectrum of the dissipative system, which naturally contains any additional broadening/narrowing and resonant energy shifts due to cavity-exciton coupling. In our semiconductor cavity system of interest, the photons can be emitted via the leaky cavity mode (emitted with a rate Γ_c), or via background radiation modes above the PC light line (emitted with a rate Γ_x). The steady-state spectrum for the cavity-mode emission and the radiation-mode (excitonic) emission, respectively, can be written as $S_{\text{cav}}(R, \omega) = F_{\text{cav}}(R) S_{\text{cav}}^0(\omega)$ and $S_{\text{rad}}(R, \omega) = F_{\text{rad}}(R) S_{\text{rad}}^0(\omega)$, where $F_{\text{cav}/\text{rad}}(R)$ is a geometrical function [15] that depends on the collection/detector geometry. The relevant spectral contributions can be written as $S_{\text{cav}}^0(\omega) = \frac{\Gamma_c}{\pi} \lim_{t \rightarrow \infty} \text{Re} \left[\int_0^\infty \langle a^\dagger(t) a(t + \tau) \rangle e^{i\omega\tau} d\tau \right]$ for emission from the leaky cavity, and $S_{\text{rad}}^0(\omega) = \frac{\Gamma_x}{\pi} \lim_{t \rightarrow \infty} \text{Re} \left[\int_0^\infty \langle \sigma^+(t) \sigma^-(t + \tau) \rangle e^{i\omega\tau} d\tau \right]$, for emission via radiation modes. Using the master equation above, adopting the one photon-correlation approximation $\langle \sigma_z a \rangle = -\langle a \rangle$, and applying fermion statistics $[\sigma^-, \sigma^+]_+ = 1$ (valid for weak excitation), one obtains [23]

$$S_{\text{cav}}(R, \omega) = F_{\text{cav}}(R) \frac{\Gamma_c}{\pi} \text{Re} \left[\frac{i \langle a^\dagger a \rangle_{ss} D(\omega)}{C(\omega) D(\omega) - g^2} + \frac{ig \langle a^\dagger \sigma^- \rangle_{ss}}{C(\omega) D(\omega) - g^2} \right], \quad (3)$$

and

$$S_{\text{rad}}(R, \omega) = F_{\text{rad}}(R) \frac{\Gamma_x}{\pi} \text{Re} \left[\frac{i \langle a^\dagger a \rangle_{ss} C(\omega)}{C(\omega) D(\omega) - g^2} + \frac{ig \langle \sigma^+ a \rangle_{ss}}{C(\omega) D(\omega) - g^2} \right], \quad (4)$$

with $C(\omega) = \omega - \omega_c + \frac{i}{2}\Gamma_c$ and $D(\omega) = \omega - \omega_x + \frac{i}{2}(2P_x + \Gamma_x + \Gamma'_x)$. The total spectrum

$$S_{\text{tot}}(R, \omega) = F_{\text{cav}}(R) S_{\text{cav}}^0(\omega) + F_{\text{rad}}(R) S_{\text{rad}}^0(\omega), \quad (5)$$

where we estimate – by fitting the trends of the experiments below – that $F_{\text{cav}}/F_{\text{rad}} \approx 3/1$, since the detector collects a larger percentage of the cavity-mode emission (as the radiation-mode emission is distributed over a broad range of angles). This estimate partly depends upon the value of Γ_x , since both F_{rad} and Γ_x determine the spectral contribution from the radiation-mode emission (more details are given later).

The steady-state solutions for $\langle \sigma^+ \sigma^- \rangle_{ss}$ (exciton population), $\langle a^\dagger a \rangle_{ss}$ (cavity photon population), and $\langle a^\dagger \sigma^- \rangle_{ss}$, are determined to be [23]

$$\langle a^\dagger a \rangle_{ss} = \frac{g^2 \Gamma (P_x + P_c) + P_c (2P_x + \Gamma_x) \left[\frac{\Gamma^2}{4} + (\omega_c - \omega_x)^2 \right]}{g^2 \Gamma (2P_x + \Gamma_x + \Gamma_c) + \Gamma_c (2P_x + \Gamma_x) \left[\frac{\Gamma^2}{4} + (\omega_c - \omega_x)^2 \right]}, \quad (6)$$

$$\langle a^\dagger \sigma^- \rangle_{ss} = \frac{-ig \left(\langle a^\dagger a \rangle_{ss} - \frac{P_x}{2P_x + \Gamma_x} \right) \left[i(\omega_c - \omega_x) + \frac{\Gamma}{2} \right]}{\frac{\Gamma^2}{4} + (\omega_c - \omega_x)^2 + \frac{g^2}{2P_x + \Gamma_x} \Gamma}, \quad (7)$$

$$\langle \sigma^+ \sigma^- \rangle_{ss} = \frac{P_x + ig \left(\langle a^\dagger \sigma^- \rangle_{ss} - \langle a \sigma^+ \rangle_{ss} \right)}{(\Gamma_x + P_x) + P_x}, \quad (8)$$

where $\Gamma = 2P_x + \Gamma_x + \Gamma'_x + \Gamma_c$. Thus, through Eqs. (3-8), we have a convenient analytical form for the total spectrum and mean photon/exciton numbers, using a model that accounts for fermion statistics, incoherent pump processes that can excite both the QD exciton and the cavity, as well as pure dephasing. Physically, the exciton pump originates from higher lying exciton levels that incoherently relax to excite the exciton; while the cavity mode pump can be due to background excitons (from the same QD or different QDs) [24] that off-resonantly excite the cavity mode (e.g., see Ref. [15]). The above equations apply in the weak excitation regime only, but we have numerically verified that higher-order correlation effects are negligible in the simulations below, since the mean number of cavity photons is much less than unity. Nonlinear processes and anharmonic cavity-QED effects using the same master equation are highlighted elsewhere [23].

3.2. Simulations

To map closely to our experiments described above, we choose a fixed cavity decay rate of $\Gamma_c = 0.14 \text{ meV}$, and a radiative decay rate of the exciton, $\Gamma_x = 0.3 \mu\text{eV}$; this latter rate is about one order of magnitude smaller than for QDs in bulk samples, because the radiation mode decay is significantly suppressed by the slab and the photonic crystal geometry. The pure dephasing is the dominant exciton broadening mechanism and is allowed to vary from $\Gamma'_x = 0.07 - 0.14 \text{ meV}$ as a function of detuning, which occurs in the experiments by temperature tuning. This process is well known and is primarily due to electron – acoustic-phonon interactions [30–32] and possibly spectral diffusion. Although coupling to LA-phonons results in a detuning dependent g and asymmetry in the exciton lineshape [33, 34] (especially at lower temperatures), we have numerically verified that accounting for such coupling does not cause any cavity-to-exciton spectral shifting. We first find a sensible range of pump powers and exciton-cavity coupling constants. We determine $P_x = 0.005 \text{ meV}$ and $P_c = 0.01 P_x$ from a fit that reasonably represents the experiments data, over a broad spectral range. The exciton-cavity coupling constant is varied from a weak-coupling rate $g = 0.002 \text{ meV}$ through to a strong-coupling rate $g = 0.1 \text{ meV}$. For the largest g value, the mean exciton and cavity photon numbers are found to peak at around 0.24 and 0.04, respectively. Thus we are far outside the regime of single exciton lasing, and, as remarked earlier, higher-order photon correlation effects can be safely neglected (at least within the stated limits and assumptions of the model).

We next compute the spectra for the smaller exciton-cavity coupling parameter, $g = 0.002 \text{ meV}$ ($2 \mu\text{eV}$). In Fig. 3(a), we display the normalized spectra, which contain contributions from both the cavity-mode emission and the radiation-mode emission. We highlight that the radiation-mode spectra, on its own (neglecting the cavity mode emission), consistently yields only one exciton peak and no evidence of cavity-like mode emission; this is, in fact, the usual and expected behavior for a weakly coupled PL spectra, but it is not at all like the spectra observed in the experiments. Rather, a double-peak feature is obtained even for exciton and cavity modes that are far off resonance; this behavior has been highlighted before for an initially excited exciton, coupled to a leaky cavity mode [15, 16]. Figure 3(b) show an analysis of the the resonances of the two-peaked spectra for each value of detuning, and cavity shifts of around $\pm 0.02 \text{ meV}$ are observed for the cavity mode. To obtain the total spectra [Eq. (5)], as mentioned earlier, we have used $F_{\text{cav}}/F_{\text{rad}} = 3/1$; specifically, what we can obtain from fitting is the ratio $(\Gamma_c F_{\text{cav}})/(\Gamma_x F_{\text{rad}})$, and so the aforementioned ratio (3/1) is correct if $\Gamma_x = 0.3 \mu\text{eV}$. We also stress that it is certainly not surprising that the spatial dependence of the detector and collection optics will have an influence on the measured spectrum.

Next we investigate the influence of the larger exciton-cavity coupling constant of $g = 0.1 \text{ meV}$ ($100 \mu\text{eV}$). In Fig. 3(c-d) is shown the emitted spectra, where now we are clearly in the strong coupling regime, showing the familiar repulsion behavior between the cavity mode

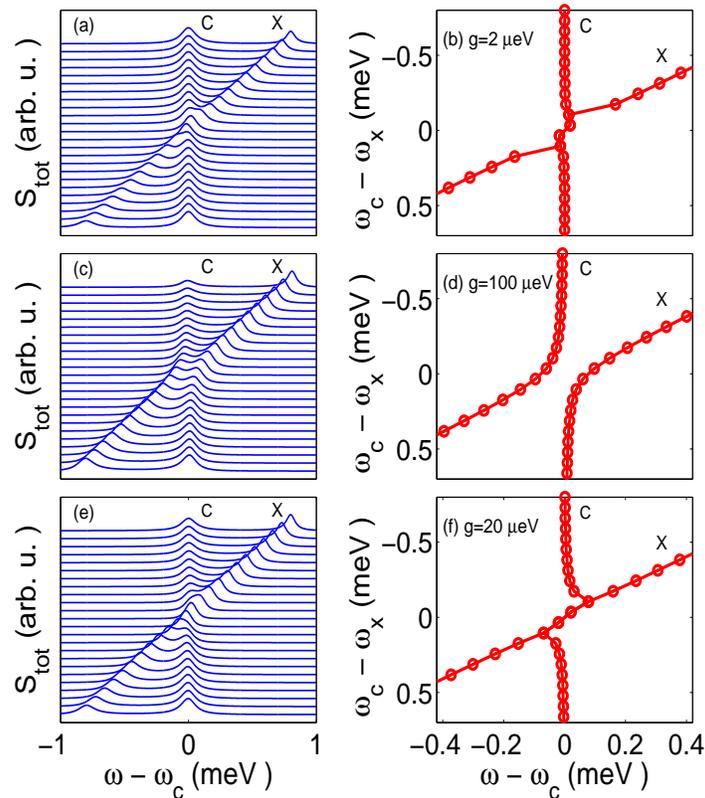


Fig. 3. PL spectra [(a),(c),(e)] and spectral peaks [(b),(d),(f)], as a function of cavity-exciton detuning. The pure dephasing of the exciton is allowed to double as a function of detuning, corresponding to a temperature increase. In the simulations, $P_x = 0.005$ meV, $P_c = 0.01P_x$, $\Gamma_c = 0.14$ meV, $\Gamma_x = 0.3$ μ eV, and $\Gamma'_x = 0.07 - 0.14$ meV. The exciton-cavity coupling rate is $g = 2$ μ eV (a-b), $g = 100$ μ eV (c-d), and $g = 20$ μ eV (e-f).

and exciton mode as they come near resonance.

Finally, we choose an intermediate exciton-cavity coupling constant of $g = 0.02$ meV (20 μ eV). In Fig. 3(e-f) is shown the total emitted spectra, and once again we see attraction and mode pulling features between the cavity mode and exciton mode as they come near resonance, which is the opposite trend to the strong coupling regime. In this intermediate coupling scenario, we obtain cavity shifts of around ± 0.12 meV. Importantly, these spectral shifts are significantly larger than what might be expected from the product of two Lorentzian lineshapes, which would be the case for the cavity-mode emission from only an excited exciton [15]. What is also interesting, is that these calculations also indicate that the experimental spectra must include contributions from both radiation-mode and cavity-mode decay channels, though it is common practise to only compute one of these.

Although a direct fit to the experimental data is difficult because of various temperature dependent parameters, such as the shifting cavity resonance and increasing pure dephasing, we have nevertheless demonstrated that significant and unusual cavity mode shifts can occur when the cavity mode spectrally approaches the exciton mode, even in the weak coupling regime; the details of the total PL spectra depend upon the various linewidths, the incoherent pump rates, and the inclusion of both radiation-mode and cavity-mode emission. We also note that if the

cavity pump is zero, then the total emitted spectrum does not match the experiments well, as the oscillator strength of the cavity emission is not large enough, and the total spectrum becomes dominated by the radiation decay; similarly, if the cavity mode pump is increased by an order of magnitude, then the emission is dominated by only the bare cavity mode. Thus we believe that we have found a qualitatively new and unexpected cavity-QED regime, where the cavity mode emission peak appears to be attracted to the exciton mode, given the right range of parameters. Nevertheless, the experimental data, particularly the data in Fig. 2(b) show even more striking cavity mode shifts, indicating that further processes may also be taking place that we have not yet identified.

4. Conclusions

We have introduced the apparent phenomenon of cQED-assisted *attraction* between a leaky cavity mode and an exciton mode in a photonic crystal cavity. This weak-to-intermediate coupling effect is partly explained by a quantum theory that includes incoherent pumping, pure dephasing, fermion statistics for the QD emitter, and emission from both radiation modes and the leaky cavity mode. Even in the weak coupling regime, the observed emitted spectra, and the calculations, suggest that the cavity mode emission dominates the emission process from the photonic crystal cavity, similar to other observations in the strong coupling regime. However, in the present case, one also has a significant influence for the radiation-mode spectrum, so both decay channels contribute to the observed PL spectra. More surprisingly, is the persistent observation of cavity mode attraction toward the exciton mode as the system sweeps through cavity-exciton resonance, as a function of temperature. In comparison to the theoretical findings, additional mode-pulling phenomena are evident in some of the experimental data, which indicate that some other effects may be occurring in the sample. A more systematic investigation of pump power and possibly the role of multiple QD excitons would be desirable and is left to future work.

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