PHYSICAL REVIEW B **81**, 121310(R) (2010)

Photon antibunching in strongly coupled exciton–semiconductor cavity systems: Role of off-resonant coupling to multiple excitons

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(Received 28 October 2009; revised manuscript received 1 March 2010; published 29 March 2010)

We present a master equation approach to study the second-order quantum autocorrelation functions for up to two quantum-dot excitons, coupled to an off-resonant cavity in a semiconductor-single quantum-dot cavity system. For a single coupled off-resonant exciton, we observe unusual oscillatory behavior in the early time dynamics of the cavity autocorrelation function, which leads to decreased antibunching relative to the exciton mode. With a second coupled exciton in the system, we find that the magnitude and the lifetime of these oscillations greatly increases, since the cavity is then able to exchange photons with multiple excitonic resonances. These results demonstrate that for incoherent pumping, the ubiquitous two-level atom model completely breaks down and that multiple excitonic resonances act in concert to spoil the antibunching characteristics of the cavity quasimode. We connect these findings to a series of recent surprising experimental results for single quantum dot-semiconductor cavity systems, including photonic crystal systems and micropillar cavities.

DOI: 10.1103/PhysRevB.81.121310 PACS number(s): 78.67.Hc, 42.50.−p, 42.70.Qs, 78.90.+t

I. INTRODUCTION

When a two-level resonance is coupled with a suitable optical cavity mode, the regime of cavity-quantum electrodynamics (cavity-QED) can be realized. Well-known cavity-QED effects include the Purcell effect[1] and vacuum Rabi splitting.[2] Cavity-QED systems that emit *deterministic* single photons are also interesting from an applications viewpoint, opening doors to practical ways of doing quantum information processing.[3,4] Two-level system behavior can occur in a number of material structures, including atoms,[5,6] ions,[7] molecules,[8,9] and color centers.[10] Semiconductor quantum dots (QDs) (Refs. 11 and 12) have also been proposed as an *artificial atom* with strong excitonic levels that mimic two-level behavior and are particularly attractive since they can function as a scalable and compact system for emitting single photons. Direct coupling between QD excitons and semiconductor cavities has recently been demonstrated in a variety of systems such as micropillars,[13–15] microdisks,[16,17] and photonic crystal (PC) cavities.[18,19]

An important difference between modern QD semiconductor structures, and earlier atomic systems, is that the confined photon cavity environment is considerably more complicated than a couple of Fabre-Perot mirrors, frequently exploiting photonic band-gap physics and nanoscale high-index-contrast optical modes. In addition, QDs exhibit rich excitonic structure, hence it is not known to what extent the two-level approximation applies, though it is expected to be suitable for narrowband cavity systems with a well-defined cavity resonance that couples to a *target exciton mode*. Thus, it can be anticipated that, in addition to the familiar cavity-QED phenomena well known to atoms, further complexity unique to these semiconductor cavity-QD systems can be observed. Consequently, the standard two-level model, applied to model strong-coupling phenomenon in semiconductor-dot cavity systems, can be inadequate for explaining higher-order correlation effects such as photon antibunching.

A recent examination of off-resonant coupling between a cavity and a single QD resulted in the observation of a pronounced cavity-mode emission.[19–21] First considered a surprising result, it is now known that this is likely due to the nontrivial coupling between the leaky cavity mode and the exciton mode,[22–26] which is exasperated in a planar PC system.[26] Even more surprising has been the various reports of poor to no antibunching—a measure of the quantum nature of the emitter—even in the strong-coupling regime for an exciton detuned from the cavity resonance.[19] Further experimental studies of the quantum correlation function in the off-resonant regime have provided clues to the nature of this reduction. Specifically, it has been observed that the cavity autocorrelation function shows significantly worse antibunching than the exciton autocorrelation function,[20] leading us to investigate causes related to the differences between the exciton and the cavity emission. An understanding of this mystery is an important step toward the understanding and design of QD-based single photon sources and would provide fresh modeling insight into the nanoscale light-matter interactions.

In this work, we introduce an intuitive quantum optics formalism that allows one to calculate both the exciton and the cavity-mode quantum autocorrelation functions for a single QD-PC cavity system. We provide a framework in which it is possible to interpret recent experimental results, by emphasizing the consequences of the quasimode nature of the cavity, and the significant effect of additional excitons on the quantum autocorrelation functions of the system. Our results highlight the importance of accounting for coupling to additional excitons in the system as they play a qualitatively important role, even if they are far off-resonance.

II. THEORY AND MODEL

Our model system consists of a single QD embedded in a planar PC cavity or a micropillar cavity but is also applicable to other semiconductor cavity systems where the leaky cavity mode dominates the emission. Due to the leaky nature of the cavity mode, both the exciton and the cavity are able to emit photons out of the plane (vertically). In Fig. 1(a), we show the relevant energy level diagram that consists of up to two excitons (both originating from a single QD and assumed to be independent—though they are indirectly coupled through the cavity mode) and the resonant mode of the PC cavity. The uppermost exciton energy level is utilized for exciting the system with a far off-resonant pump pulse. We adopt a master equation approach for the density operator,[27]

$$\dot{\rho} = \frac{1}{i\hbar}[\hat{H}_I,\rho] - \kappa(\hat{a}_c^\dagger\hat{a}_c\rho + \rho\hat{a}_c^\dagger\hat{a}_c - 2\hat{a}_c\rho\hat{a}_c^\dagger) - \sum_{i=1,2}\left[\gamma_i(\hat{\sigma}_{ii}\rho + \rho\hat{\sigma}_{ii} - 2\hat{\sigma}^-\rho\hat{\sigma}^+) - \gamma_i'(\hat{\sigma}_i^z\rho\hat{\sigma}_i^z - \rho)\right] - \sum_{i=1,2}\gamma_{iu}(\hat{\sigma}_{uu}\rho + \rho\hat{\sigma}_{uu} - 2\hat{\sigma}_{iu}\rho\hat{\sigma}_{ui}), \quad (1)$$

with the interaction Hamiltonian $\hat{H}_I = i\hbar\Sigma_{i=1,2}\, g_i(\hat{\sigma}_i^-\hat{a}_c^\dagger - \hat{\sigma}_i^+\hat{a}_c) + \frac{i\hbar\Omega(t)}{2}(\hat{\sigma}_u^+ - \hat{\sigma}_u^-)$, where $\Omega(t)$ is a classical excitation pulse that excites the upper lying exciton level, $|x_u\rangle$, $2\gamma_{iu}$ is the fast nonradiative decay rate, and $\hat{\sigma}_{ii} = \hat{\sigma}_i^+\hat{\sigma}_i^-$, $\hat{\sigma}_i^z = \hat{\sigma}_i^+\hat{\sigma}_i^- - \hat{\sigma}_0^+\hat{\sigma}_0^-$. The boson operators $\hat{a}_c$ and $\hat{a}_c^\dagger$ are the photon creation and annihilation operators for the cavity mode, and $\hat{\sigma}_i^+$ and $\hat{\sigma}_i^-$ are the Pauli raising and lowering operators, respectively. The parameters of the system include g_i—the coupling strength between the cavity and the ith exciton; and $2\gamma_i$, 2κ, and $2\gamma_i'$—the radiative, cavity, and pure dephasing decay rates, respectively. Importantly, both the one- and two-time equations of motion can be calculated from the master equation,[27] and utilizing the quantum regression theorem,[2] we obtain the second-order quantum correlation functions

$$G^{(2)}_{x_i,x_i}(t,t+\tau) \propto \langle\hat{\sigma}_i^+(t)\hat{\sigma}_i^+(t+\tau)\hat{\sigma}_i^-(t+\tau)\hat{\sigma}_i^-(t)\rangle, \quad (2)$$

$$G^{(2)}_{c,c}(t,t+\tau) \propto \langle\hat{a}_c^\dagger(t)\hat{a}_c^\dagger(t+\tau)\hat{a}_c(t+\tau)\hat{a}_c(t)\rangle, \quad (3)$$

for the exciton and the cavity mode, respectively. The time integrated $g^{(2)}(\tau)$ is then obtained by integrating over time t and normalizing to the peak of one of the sideband pulses.[28] The photon indistinguishability and interference can be measured by sending two consecutive generated photon pulses through a beam splitter.[29]

III. CALCULATED SPECTRA AND ANTIBUNCHING CORRELATIONS

We numerically excite the system up to a higher-lying level (x_u) from the ground state via Gaussian excitation pulses, $\Omega(t)$, with full width at half maximum of 10 ps spaced 10 ns apart; the pulse amplitudes correspond closely to π pulses to optimally excite the high-lying level. The system is then allowed to quickly relax from $|x_u\rangle$ to $|x_i\rangle$ via a fast nonradiative decay rate of $2\gamma_{iu} = 0.4$ meV. The total emission spectra $S_T(\omega)$ calculated in the presence of one and two excitons can be found in the lower panel of Fig. 1. It is given by $S_T(\omega) = S_c(\omega) + \Sigma_i S_{x_i}(\omega)$, where $S_c(\omega) = F_c\kappa\langle a^\dagger(\omega)a(\omega)\rangle$ and $S_{x_i}(\omega) = F_{x_i}\gamma_i\langle\sigma_i^\dagger(\omega)\sigma_i(\omega)\rangle$ are the emission spectra for the cavity and the excitons, respectively. We chose a conservative estimate for the ratio of geometric factors, $F_c/F_{x_i} \approx 2$,

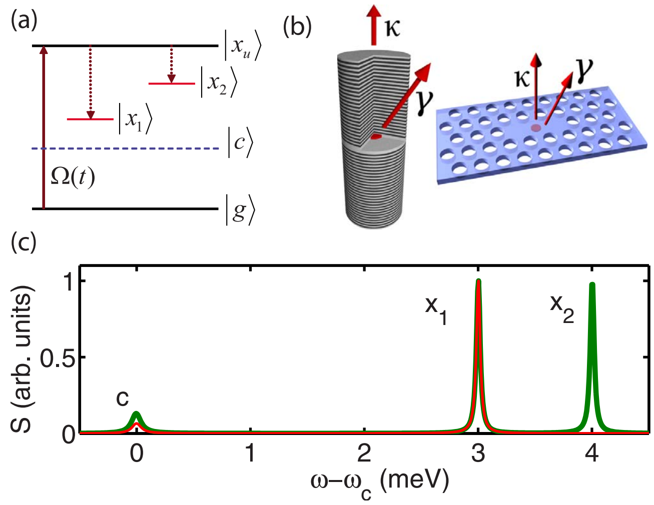

FIG. 1. (Color online) System under investigation. (a) Energy-level diagram for the theoretical model. (b) Schematic of the micropillar cavity and PC cavity with an embedded QD, showing a background radiative decay process and a leaky cavity decay process. (c) Total emission spectra for one exciton (thin red curve with lower peak at c and peak at x_1) and two excitons (thick green curve with peaks at c, x_1, x_2). The two curves completely overlap at peak x_1. Here $\gamma' = 20$ μeV.

which is reasonable for PC cavities but can be much larger for micropillar cavities. This allows proper scaling between excitonic and cavity spectra for the total spectrum calculation. In the spectrum, a single exciton couples to the leaky cavity mode and the dominant cavity emission spectrum consists of both the bare exciton resonance as well as the bare cavity resonance. These one-exciton results are consistent with the results of others.[23–26] As recognized, introducing a second exciton has a small but non-negligible effect on the spectrum. Analogously to the first exciton, the second one also *feeds* the cavity mode, and therefore has the effect of increasing the weight under the peak at the bare cavity resonance as well as introducing a peak at its own bare resonance. We emphasize that the cavity emission spectrum dominates the emission characteristics and contains resonances at both the bare cavity resonance as well as the resonances of the excitonic modes that feed it.

In choosing model parameters, we aim to mirror those determined from related experiments,[19–21] and choose a cavity-exciton coupling of $g_1 = 0.075$ meV, $g_2 = 0.1$ meV and half rates of radiative and cavity decay of $\gamma_{1,2} = 0.1$ μeV and $\kappa = 0.05$ meV, respectively. This leads to equal oscillator strengths for the two excitons at detunings of 3 and 4 meV as seen in Fig. 1, similar to experimental results.[19] The second-order correlation functions are shown in Figs. 2(a) and 2(c). The theory utilized is amenable to projecting onto either the cavity quasimode or the excitonic mode separately—even though the system is on resonance—which is something that is not experimentally accessible. However, since the cavity mode dominates the emission, the on-resonance condition is heavily dominated by the cavity-mode contribution. With other parameters set, we extract a representative pure dephasing rate of $2\gamma' = 4$ μeV, which corresponds to an antibunching dip of 77% on resonance [see

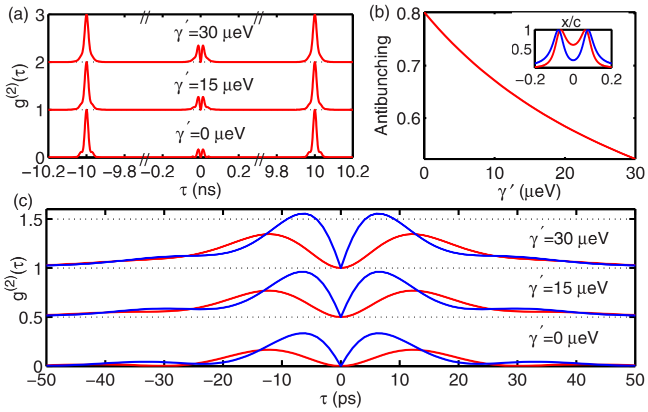

FIG. 2. (Color online) (a) Cavity autocorrelation function. (c) Cavity (red curve with smaller amplitude oscillations) and exciton (blue curve with larger amplitude oscillations) autocorrelation functions. (b) Degree of antibunching—$1-A_0/A$, the ratio of the area under the $\tau=0$ peak, A_0, compared to the area under peaks at other times, A. Inset: associated spectrum—for an exciton on resonance with the cavity in the strong-coupling regime—showing normalized cavity (red curve with more intense emission at $\omega=\omega_0$) and exciton (blue) emission spectra.

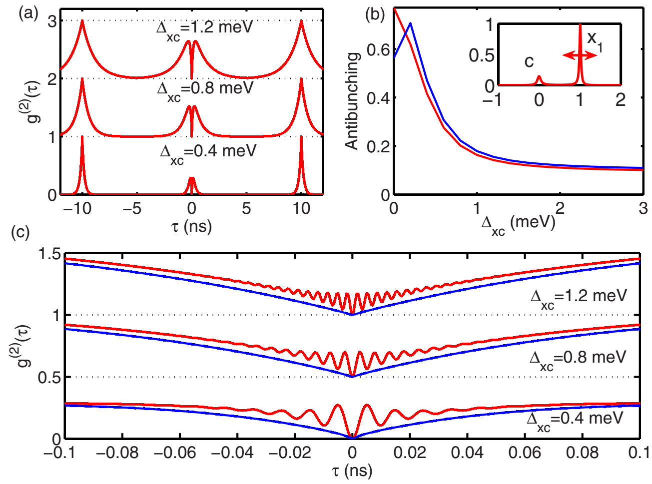

FIG. 3. (Color online) (a) Cavity autocorrelation function. (b) Antibunching for the cavity (red oscillatory curve) and the exciton (blue). Inset: cavity emission spectrum schematic showing the varying detuning of the exciton. (c) Cavity (lower red curve) and exciton (upper blue curve) autocorrelation functions.

Fig. 2(b)]. This represents an intermediate case in the context of the experiments of interest.

We now introduce a detuning between the exciton and the cavity resonance $\Delta_{x_i c}=\omega_c-\omega_{x_i}$, where ω_c and ω_{x_i} are the *bare* cavity and exciton resonances, respectively. We examine a system with a singly coupled, off-resonance exciton $x=x_1$, as well as an upper loading exciton, x_u. The calculated autocorrelation functions are shown in Fig. 3 for three different detunings. In general, we observe an overall broadening of all peaks (as compared to the resonant case) and a reduction in antibunching. This reduction is magnified with increased detuning. However, for all values of detuning seen in Fig. 2(b), we note that $g_{cc}^{(2)}(\tau)$ shows decreased antibunching relative to $g_{xx}^{(2)}(\tau)$. This general trend qualitatively agrees with the experimental observations reported by Press *et al.*,[20] where a similar reduction in antibunching of the cavity mode relative to the exciton was observed in off-resonant systems. For larger detuning, the magnitude of the antibunching levels off to about 11% and 10% for the exciton and the cavity, respectively. We note that the exciton is expected to experience some antibunching even in the absence of the cavity,[28] subject to the amount of pure dephasing, which would explain the limiting case seen for large detuning. A closer examination of the early time dynamics of the autocorrelation functions for a single exciton [see Fig. 3(c)] reveals oscillations in the $g_{cc}^{(2)}(\tau)$ that are not seen for $g_{xx}^{(2)}(\tau)$. These two-photon interference oscillations are caused by the off-resonant coupling of the cavity to the exciton, and are manifestations of the quasimode nature of the cavity mode. The period of the oscillations corresponds to the detuning frequency of the exciton $T_{\mathrm{osc}}\propto 1/f_{\mathrm{detuning}}$, and therefore the frequency of the oscillations increases with increased detuning.

We next turn to describing the effects of other nearby excitons on the ensuing quantum statistics. We now include a second exciton that is essentially identical to the initial one (same γ,γ'). We show a representative plot of $g_{cc}^{(2)}(\tau)$ in the presence of one versus two excitons coupled to the cavity in Fig. 4(a). For the case shown, the second exciton is doubly detuned from the cavity relative to the first exciton. As can be seen in Fig. 4(a), the presence of the additional exciton has a profound influence on $g_{cc}^{(2)}(\tau)$. These extraneous effects consist of magnifying all the features that were previously unique to $g_{cc}^{(2)}(\tau)$. In particular, we find that both the magnitude, as well as the lifetime of oscillations, in the early time dynamics, are significantly increased, thereby magnifying the differences in the antibunching of the cavity versus the exciton mode. The magnitude of these differences varies de-

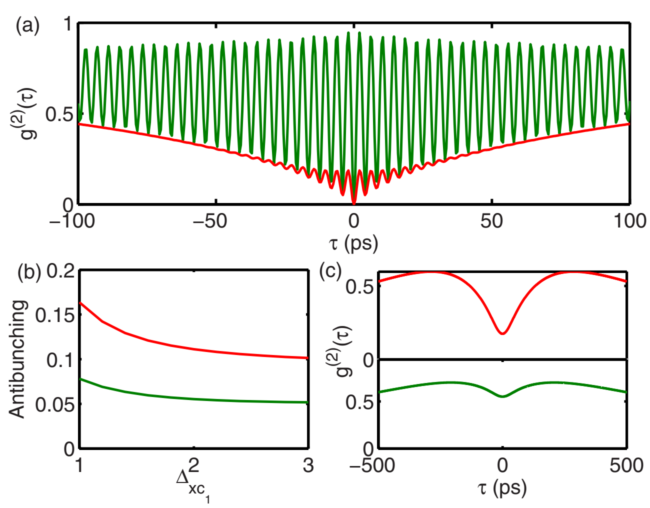

FIG. 4. (Color online) (a) Cavity autocorrelation functions for one (smaller oscillations) and two (larger oscillations) excitons with detunings $\Delta_{x_1c}=1$ meV and $\Delta_{x_2c}=2$ meV. Inset: one (red) and two (green) exciton spectra showing cavity-mode emissions. (b) Antibunching for the cavity mode with one (upper red curve) and two (lower green curve) excitons. The detuning of the second exciton is given by $\Delta_{x_2c}=\Delta_{x_1c}+1.0$ meV. (c) The effect of finite detector time resolution on $g_{cc}^{(2)}(\tau)$ with one (top) and two (bottom) excitons for the same detunings as in (a).

pending on the choice of parameters but the general trend of decreased antibunching in the presence of an additional exciton persists. Specifically, we examined cases with γ and γ' up to an order of magnitude larger, as well as a large range of detunings between the cavity and the initial exciton. In cases where the detuning of the additional exciton is significantly larger than that of the target exciton, we begin to recover the antibunching expected for a single exciton coupled to the cavity.

In Fig. 4(b), we plot the antibunching of the cavity mode coupled to one versus two excitons. We vary the detuning of the excitons, while maintaining a separation of 1 meV between them, and find that the presence of the second exciton spoils the antibunching of the cavity mode by a relative percentage of about 50%, for all detunings for our present parameters. This effect is more pronounced with smaller values of 2γ and $2\gamma'$ the exciton decay rate, and the pure dephasing rate, as well as for smaller detunings from the cavity mode. In cases where the second excitonic line is more intense than the initial one or with more excitons excited (e.g., Ref. 19), we find that the antibunching of the cavity mode further decreases, even to the point of Poissonian statistics, which is again consistent with experimental observations.

Finally, in order to connect more closely with experimental detection, we include the effects of finite detector time resolution, by modeling the detector time response as a Gaussian function with a full width at half maximum of 50 ps and an area of one. We apply this averaging to the output of the $g_{cc}^{(2)}(\tau)$ and $g_{xx}^{(2)}(\tau)$ calculations and obtain the results seen in Fig. 4(c). The primary effect of finite detector response is an averaging out of any behavior that occurs at time scales shorter than the 50 ps response time, such as the quick drop to zero seen on both the cavity and exciton autocorrelation for $\tau \rightarrow 0$, and the oscillatory behavior observed in $g_{cc}^{(2)}(\tau)$. However, this averaging preserves the relative areas under the peaks in the autocorrelations, and therefore has no effect on the magnitude of the predicted antibunching.

IV. CONCLUSION

By utilizing a master equation model to describe the coupling between a leaky cavity mode and one or more QD excitons, we find significant differences between the second-order quantum autocorrelation functions of the exciton modes and cavity modes. These differences are manifestations of the quasimode nature of the cavity and include oscillations in the early time dynamics of $g_{cc}^{(2)}(\tau)$, as well as the reduced antibunching of the cavity mode relative to the exciton. Furthermore, we find that the presence of additional excitons magnifies this difference and largely destroys the antibunching of the cavity mode, which qualitatively agrees with recent experimental observations.[19,20]

ACKNOWLEDGMENTS

This work was supported by the National Sciences and Engineering Research Council of Canada, the Canadian Foundation for Innovation, and the Walter C. Sumner Memorial Foundation.

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