Article:

Gonzalez-Tudela, A., del Valle, E., Cancellieri, E. orcid.org/0000-0002-4099-2716 et al. (3 more authors) (2010) Effect of pure dephasing on the Jaynes-Cummings nonlinearities. Optics Express, 18 (7). pp. 7002-7009. ISSN 1094-4087

https://doi.org/10.1364/OE.18.007002

Effect of pure dephasing on the Jaynes-Cummings nonlinearities

A. Gonzalez-Tudela,[1] E. del Valle,[2] E. Cancellieri,[1] C. Tejedor,[1] D. Sanvitto,[3] and F.P. Laussy[2,*]

[1]*Física Teórica de la Materia Condensada, Universidad Autónoma de Madrid, 28049, Spain.*
[2]*School of Physics and Astronomy. University of Southampton, Southampton, SO171BJ, United Kingdom.*
[3]*Física de Materiales, Universidad Autónoma de Madrid, 28049, Spain.*
(Dated: September 30, 2009)

We study the effect of pure dephasing on the strong-coupling between a quantum dot and the single mode of a microcavity in the nonlinear regime. We show that the photoluminescence spectrum of the system has a robust tendency to display triplet structures, instead of the expected Jaynes-Cummings pairs of doublets at the incommensurate frequencies $\pm(\sqrt{n}\pm\sqrt{n-1})$ for integer n. We show that current experimental works may already manifest signatures of single photon nonlinearities.

Strong-coupling of quantum states—whereby bare modes vanish and their quantum superpositions combining their properties take over—is now commonplace in cavity Quantum Electrodynamics (QED) physics. Following the seminal report with atoms [1], it is now realized with circuit QED [2], nano-mechanical oscillators [3] and, last but not least, with quantum dots (QD) (see [4, 5, 6, 7, 8, 9] for some recent reports, and references therein). Regarding the latter, there have been recently rapid progresses in providing an accurate quantitative description of the experiment, thanks to theoretical efforts pointing towards specificities of the semiconductor case, such as geometry of detection, self-consistent states imposed by the incoherent pumping, etc. [10, 11] These efforts culminated with the report by Laucht *et al.* [12], who provided a compelling global fit of their experimental data. This approach allows, in the tradition of a mature field of physics, to discard or validate a theoretical model by confronting it statistically with the experiment. Laucht *et al.* have thus been able to bring out quantitatively which mechanisms matter in the semiconductor strong-coupling case. Confirming the suggestions of many previous works [10, 13, 14, 15], they have shown that a pure dephasing term is involved. Their analysis evidence an exciton dephasing via interactions with phonons [16] at high temperatures, and with carriers outside the quantum dot [17] at high excitation power. In this Letter, we shall not focus on the nature of the dephasing but take for granted that it is not negligible.

The most appealing features of strong-coupling are at the quantum level, when a few quanta of excitations rule the dynamics. A splitting at resonance is a tempting landmark of this regime, but is not, being in no essential way different from the normal mode coupling that is a classical feature of coupled oscillators [18]. To evidence the quantum character of the coupling, photon-counting experiments have been performed, reporting that only one quantum of excitation couples the modes [4, 5]. The next step is to probe nonlinearities and witness their sensitivity at the quantum level.

The most basic and fundamental representation of the QD is that of a two-level atomic-like system [19]. Dressing this fermionic system (i.e., coupling it strongly) with more than one photon yields a splitting of $2\sqrt{n}g$ when n quanta are involved (g is the interaction strength, we take $\hbar = 1$). Transitions between these dressed states provide spectral lines at incommensurate energies $\pm(\sqrt{n}\pm\sqrt{n-1})g$, which are a direct manifestation of full-field quantization, as predicted by one of the most important theoretical model of quantum physics, the Jaynes-Cummings Hamiltonian [20]. Evidencing these nonlinearities is a chief goal of quantum optics. It has been fulfilled with atoms [21] and more recently with superconducting circuits [22], but a direct spectral signature remains elusive for semiconductor QDs, although compelling indirect evidences have been reported [4, 23]. We have predicted that they could be observed with a careful control (or lucky encounter) of the effective quantum state [24]. In this text, in the light of the importance of pure dephasing in semiconductors, we revisit our claims taking it into account. We show that due to dephasing, single-photon nonlinearities manifest through a triplet at resonance in the photoluminescence spectrum, rather than a quadruplet as expected previously.

The Jaynes-Cummings Hamiltonian, that describes the strong coupling of a two-level QD with the single mode of a microcavity, reads $H = \omega_a a^\dagger a + \omega_\sigma \sigma^\dagger\sigma + g(a^\dagger\sigma + a\sigma^\dagger)$ with a the photon annihilation operator (following Bose statistics) and σ the material excitation annihilation operator (following Fermi statistics). The two modes are coupled with interaction strength g and close enough to resonance (with small detuning $\Delta = \omega_a - \omega_\sigma$) to allow for the rotating wave approximation. A Liouvillian $\mathcal{L}$ is used to describe the system in the framework of a quantum dissipative master equation, $\partial_t\rho = \mathcal{L}\rho$, taking into account decay γ_c and incoherent pumping P_c, with $c = a,\sigma$ (referred to as *cavity* and *electronic* pumping, respectively) [24]. Pure dephasing enters as an additional source of decoherence $\mathcal{L}_{\gamma_\sigma^\phi}\rho$: [12]

$$\mathcal{L}\rho = i[\rho, H] + \sum_{c=a,\sigma}\frac{\gamma_c}{2}(2c\rho c^\dagger - c^\dagger c\rho - \rho c^\dagger c) + \sum_{c=a,\sigma}\frac{P_c}{2}(2c^\dagger\rho c - cc^\dagger\rho - \rho cc^\dagger) + \mathcal{L}_{\gamma_\sigma^\phi}\rho\,. \quad (1)$$

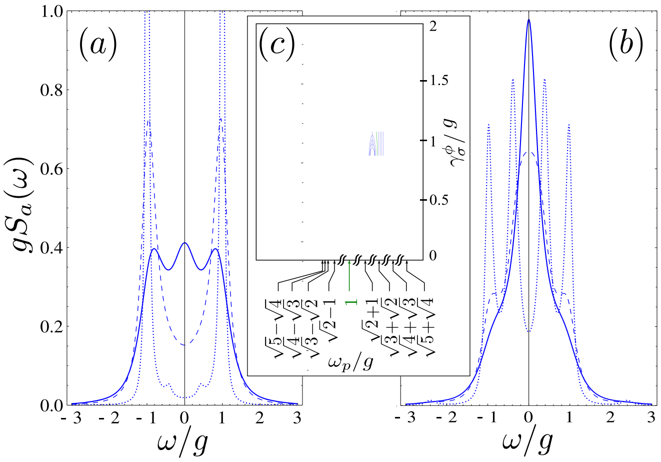

FIG. 1: (Colour online) Loss of the Jaynes-Cummings quadruplet and emergence of a triplet with dephasing, for a system well into strong coupling ($\gamma_a/g = 0.1$ and $\gamma_\sigma/g = 0.001$). Values of dephasing are $\gamma_\sigma^\phi/g = 0$ (dotted), 0.75 (dashed) and 1.5 (solid). Panel (a) [(b)] is for $P_\sigma/g = 0.02$ [0.1]. In inset (c), the dressed states resonances ω_p/g for the parameters of (b), showing the impact of dephasing on strong coupling: inner transitions melt into a common line. In thick green, the vacuum Rabi doublet maintains the satellites of the triplet.

The additional term $\mathcal{L}_{\gamma_\sigma^\phi}\rho$ originates from high excitation powers or high temperatures. It disrupts coherence without affecting directly the populations. It reads $\mathcal{L}_{\gamma_\sigma^\phi}\rho = \gamma_\sigma^\phi(S_z\rho S_z - \rho)$, where $S_z = \frac{1}{2}[\sigma^\dagger, \sigma]$ [25]. As in our previous work [11, 24], we recourse to the quantum regression formula to compute the two-time average $\langle a^\dagger(t)a(t+\tau)\rangle$ which Fourier transform gives the luminescence spectrum S_a. Following the same procedure as we described before, we identify the tensor M that satisfies $\mathrm{Tr}(C_{\{\eta\}}\mathcal{L}\Omega) = \sum_{\{\lambda\}} M_{\{\eta \atop \lambda\}}\mathrm{Tr}(C_{\{\lambda\}}\Omega)$ for any operator Ω in the basis of operators $C_{mn\mu\nu} = a^{\dagger m}a^n\sigma^{\dagger\mu}\sigma^\nu$. The addition of pure dephasing only affects diagonal elements of M and for these, only when pertaining to phase coherence (when $\mu \neq \nu$), where it is acting as a broadening:

$$M_{mn\mu\nu \atop mn\mu\nu} = i\omega_a(m-n) + i\omega_\sigma(\mu-\nu) - \frac{\gamma_a - P_a}{2}(m+n) - \frac{\gamma_\sigma + P_\sigma}{2}(\mu+\nu) - \frac{\gamma_\sigma^\phi}{2}(\mu-\nu)^2\,.$$

Other elements $M_{mn\mu\nu \atop pq\theta\vartheta}$ are as given in Ref. [24]. In the following, we study the effect of nonzero γ_σ^ϕ on the spectral shape of the cavity photoluminescence spectrum, under various cases of particular experimental relevance.

Figure 1 shows the impact of pure dephasing on the most striking landmark of the Jaynes-Cummings nonlinearities, namely, the multiplet structure that corresponds to transitions between rungs of the Jaynes-Cummings ladder [26]. We consider a system as realistic as possible while still good enough to display them unambiguously. A system with $\gamma_a/g = 0.1$ and $\gamma_\sigma/g = 0.001$, for instance (which is still outside the reach of today's technology), produces a Jaynes-Cummings fork (a quadruplet) at resonance [24], as shown in dotted lines in Fig. 1 at lower (a) and higher (b) pumpings. With pure dephasing, the spectra evolve in both cases into a triplet, with melting of the multiplet and emergence of a central peak. The mechanism of this transition is revealed in the inset (c), where the dressed mode resonances ω_p (in units of g) are shown as a function of the dephasing for the parameters of panel (b). These resonances correspond to transitions between the eigenstates of the system, which are the dressed states (polaritons) in the strong coupling regime, and the bare states (exciton and photons) in the weak coupling regime. Dressed states up to five photons are excited for the chosen parameters, and the characteristic $\pm(\sqrt{n} \pm \sqrt{n-1})$ frequencies of the transitions between rungs with such a square root splitting, are indicated at the bottom of the figure (where $\gamma_\sigma^\phi = 0$). The system remains in strong-coupling throughout, for all the states, as is evidenced from the permanence of the outer resonances $\pm(\sqrt{n}+\sqrt{n-1})$ for all $n \geq 2$. The case $n = 1$ (in thick green) corresponds to the vacuum Rabi splitting. Its position is only weakly perturbed by dephasing (as are outer peaks). Inner transitions—when the decay links the same type of states between two Jaynes-Cummings rungs (the two higher or the two lower states)—are more significantly affected. As shown in the figure, these inner resonances, at $\pm(\sqrt{n} - \sqrt{n-1})$ for $n \geq 2$, loose their splitting in succession with increasing dephasing, the sooner the higher the excited state (i.e., the larger the dressing). This loss of inner-splitting does not mean that the system goes to weak coupling, but instead that the corresponding transitions between dressed states are separated by a splitting smaller than the uncertainty due to the dephasing, and as such, these transitions overlap, thereby indeed providing the system with a new common resonance, at the cavity mode. This transition is strong from the accumulation of all the emissions of the system that were previously split from each other (dephasing here acts like a quantum eraser by providing an identical path to many previously distinguishable paths). Further increasing dephasing eventually brings the system into weak coupling, with collapse of the outer resonances as well (not shown). Triplets appear as a robust manifestation of nonlinear strong coupling with dephasing: overlapping an emerging peak at the cavity frequency with the vacuum Rabi doublet that produce satellite peaks. In contrast to previously advanced suggestions [4, 27], our analysis shows that this spectral structure is not attributable to loss of strong coupling. It is also different from the triplet of Hughes and Yao [28] that is due to interferences, and from the Mollow triplet [29], that arises in the classical limit of large number of photons. Instead, our triplet appears as a new regime at the border of the quantum and classical regimes, with dephasing acting as a smoothening agent (rather than a destructive one [30]).

Dephasing is not a parameter that is easy to control directly. In an attempt to probe the nonlinearities of the system, a natural experiment is to increase the pumping power, so as to populate more the higher excited states. The evolution of the Rabi doublet with increasing electronic pumping is shown on Fig. 2(a), for parameters from state-of-the art experimental systems (cf. caption). In this case, a triplet is also formed at resonance, but without any direct manifestation of the Jaynes-Cumming quadruplets, owing to the poor splitting to broadening ratio, even in the best systems available so far. The observation of this trend has been recently reported by Ota *et al.* [27]. We have indeed considered parameters from this work to show that our effect is within the reach of today technology. As the strong-coupling system is pushed more into the nonlinear regime with pumping, a transition to lasing occurs [24, 31]. In panels (b), we follow this evolution in presence of dephasing, from the vacuum Rabi doublet ($P_\sigma/g \approx 0.001$) towards a lasing single peak ($P_\sigma/g \approx 5$) and eventually to a quenched system recovering the bare cavity emission ($P_\sigma/g \approx 500$). This is matched by (c), the cavity population n_a (becoming > 1 with lasing and $\ll 1$ in the quantum/quenched regions), (d), the dot population (showing population inversion with lasing and being empty or saturated in the quantum/quenched regions) and, (e), the two-photon coincidence $g^{(2)}(0)$ (showing poissonian fluctuation at lasing, and antibunching/bunching in the quantum/quenched regions).

In Fig. 3, we display other manifestations of nonlinearities in the luminescence spectrum of a strongly-coupled QD/microcavity system, with the intent of showing the wide range of phenomenologies that are accessible in different configurations, as well as the strong similarities with other experiments that have so far eluded a definite theoretical explanation. In these cases, we focus more on the similar general behavior than on a tight numerical agreement with the experimental values claimed in these works, although our parameters remain within the possible margins for such systems (for instance, we have considered an ideal detector in the cases of Figs. 3, whereas we included the detector resolution of Ref. [27] in our reproduction of their experiment in Fig. 2). In the quest for strong-coupling in semiconductors, one typically performs an anticrossing experiment, where the dot and the cavity are brought to resonance to exhibit level-repulsion (maintaining their line splitting). Figure 3 shows the situation with detuning for two sets of parameters (cf. caption). In the first case, (a), well identified dot and cavity emission lines approach in the expected way but grow a central peak. This situation is similar to the one reported by Hennessy *et al.* [4]. In the second case, (b), a doublet is now produced at resonance and a triplet is observed in its vicinity and only at negative detunings. This situation is similar to the one reported by Sanvitto *et al.* [32] (that has remained unexplained—and unpublished—so far). In the first case, dephasing is constant, the cavity has a higher quality factor and electronic pumping is moderate. The triplet then arises for the same reasons as those explained for the phenomenology of Fig. 1. A slightly better system (either from system parameters or with less dephasing) would grow a quadruplet at resonance, if dephasing is indeed the cause for the central peak in this case as well. These considerations match the experimental situation of a single QD detuned from the cavity by a thin-film condensation technique. In the second case, the experimental situation varies in a few ways that would appear unimportant for the physics investigated, but that turn out to produce very different qualitative results: the dephasing has been correlated with the detuning (with a sigmoid function, to reflect that detuning is tuned with temperature), cavity photon has smaller lifetime and pumping is much stronger. This results in the emergence of a triplet outside of the resonance. In this later case, rather than superimposing a central peak, the non-commensurable transitions placed at $\pm(\sqrt{n} - \sqrt{n-1})$ at resonance produce the multiplet out of resonance, owing to their virtue of being stationary with detuning [24]. The dephasing here serves the

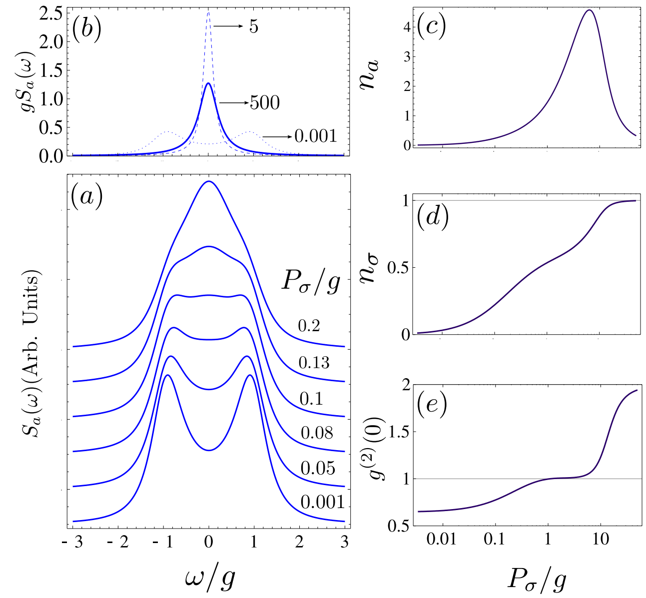

FIG. 2: Evolution of strong-coupling with increasing electronic pumping. Parameters are those of state of the art systems from the literature [6, 8, 27]: $g = 120\mu$eV, $\gamma_a = 38\mu$eV, $\gamma_\sigma = 1\mu$eV, $\gamma_\sigma^\phi = g$, at resonance, P_σ varying as indicated, without cavity pumping and with detector resolution of 46μeV. (a) The system evolves from the vacuum Rabi doublet into a triplet, much like the experiment of Ota *et al.* [27] (that, however, is not strictly at resonance, making their triplet slightly better resolved at possibly smaller values of dephasing). (b) At much higher pumpings, the system goes to lasing then to quenching. (c-e) show these transitions in (c) the cavity population, (d) dot population and (e) $g^{(2)}(0)$.

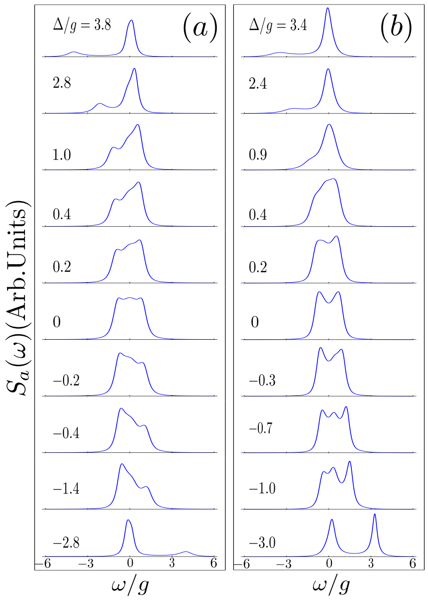

FIG. 3: Strong-coupling in the nonlinear regime in presence of dephasing as detuning is varied. Parameters are $\gamma_\sigma/g = 0.001$ and $P_a/g = 0.011$ for both panels, and for (a) [resp (b)], $\gamma_\sigma^\phi/g = 1$ [sigmoid function of Δ], $\gamma_a/g = 0.35$ [0.5] and $P_\sigma/g = 0.1$ [0.3]. Instead of the usual anticrossing, triplets are observed in slightly varying configurations: (a) A triplet is grown as the dot enters in resonance, much like the experiment of Hennessy *et al.* [4]. (b) As detuning varies with temperature, a triplet is observed *out of resonance*, with an asymmetry with detuning caused by the temperature-dependent dephasing, much like the experiment of Sanvitto *et al.* [32].

purpose of levelling the quadruplet predicted in Ref. [24] for such a structure at nonzero detuning, into a triplet.

In conclusion, we have shown that nonlinearities of the Jaynes-Cummings Hamiltonian—the pinnacle of full-field quantization in cavity Quantum Electrodynamics—have a robust tendency to manifest as triplet structures in presence of a non-negligible dephasing (such as is the case in semiconductors), rather than the expected Jaynes-Cummings quadruplets with no emission at the cavity (central) mode. We have shown that various parameters (corresponding to slightly different experimental situations) result in strong qualitative differences, such as observation of a triplet at—or out of—resonance. Although Jaynes-Cummings nonlinearities in presence of pure dephasing reproduce remarkably various experimental findings, on the basis of a clear physical picture and with the expected experimental parameters, a quantitative analysis is needed to bring a definite proof that this effect is responsible for the observed phenomenology. Experiments typically come with additional complications of their own. For instance, a non-negligible drift in detuning in Ota *et al.*'s experiment is making their triplet markedly more visible even at smaller values of the dephasing. Such a compelling proof, however, is outside the scope of this Letter and a challenge for the microcavity QED community at large.

La Caixa, Newton & RyC programs, Spanish MEC (MAT2008-01555, QOIT-CSD2006-00019) & CAM (S-0505/ESP-0200), and EPSRC funds are acknowledged.

* Corresponding author: fabrice.laussy@gmail.com

[1] S. Haroche and D. Kleppner, Physics Today **42**, 24 (1989).
[2] A. Wallraff, D. I. Schuster, A. Blais, L. Frunzio, R.-S. Huang, J. Majer, S. Kumar, S. M. Girvin, and R. J. Schoelkopf, Nature **431**, 162 (2004).
[3] S. Groeblacher, K. Hammerer, M. R. Vanner, and M. Aspelmeyer, arXiv:0903.5293 (2009).
[4] K. Hennessy, A. Badolato, M. Winger, D. Gerace, M. Atature, S. Gulde, S. Fălt, E. L. Hu, and A. Ĭmamoğlu, Nature **445**, 896 (2007).
[5] D. Press, S. Götzinger, S. Reitzenstein, C. Hofmann, A. Löffler, M. Kamp, A. Forchel, and Y. Yamamoto, Phys. Rev. Lett. **98**, 117402 (2007).
[6] M. Nomura, Y. Ota, N. Kumagai, S. Iwamoto, and Y. Arakawa, Appl. Phys. Express **1**, 072102 (2008).
[7] C. Kistner, T. Heindel, C. Schneider, A. Rahimi-Iman, S. Reitzenstein, S. Höfling, and A. Forchel, Opt. Express **16**, 15006 (2008).
[8] A. Laucht, F. Hofbauer, N. Hauke, J. Angele, S. Stobbe, M. Kaniber, G. Böhm, P. Lodahl, M.-C. Amann, and J. J. Finley, New J. Phys. **11**, 023034 (2009).
[9] A. Dousse, J. Suffczyński, R. Braive, A. Miard, A. Lemaître, I. Sagnes, L. Lanco, J. Bloch, P. Voisin, and P. Senellart, Appl. Phys. Lett. **94**, 121102 (2009).
[10] G. Cui and M. G. Raymer, Phys. Rev. A **73**, 053807 (2006).
[11] F. P. Laussy, E. del Valle, and C. Tejedor, Phys. Rev. Lett. **101**, 083601 (2008).
[12] A. Laucht, N. Hauke, J. M. Villas-Bôas, F. Hofbauer, G. Böhm, M. Kaniber, and J. J. Finley, Phys. Rev. Lett. **103**, 087405 (2009).
[13] A. Naesby, T. Suhr, P. T. Kristensen, and J. Mork, Phys. Rev. A **78**, 045802 (2008).
[14] M. Yamaguchi, T. Asano, and S. Noda, Opt. Express **16**, 118067 (2008).
[15] J. Suffczyński, A. Dousse, K. Gauthron, A. Lemaître, I. Sagnes, L. Lanco, J. Bloch, P. Voisin, and P. Senellart, Phys. Rev. Lett. **103**, 027401 (2009).
[16] P. Borri, W. Langbein, U. Woggon, V. Stavarache, D. Reuter, and A. D. Wieck, Phys. Rev. B **71**, 115328 (2005).
[17] I. Favero, A. Berthelot, G. Cassabois, C. Voisin, C. Delalande, P. Roussignol, R. Ferreira, and J. M. Gérard, Phys. Rev. B **75**, 073308 (2007).
[18] Y. Zhu, D. J. Gauthier, S. E. Morin, Q. Wu, H. J. Carmichael, and T. W. Mossberg, Phys. Rev. Lett. **64**, 2499 (1990).
[19] P. Michler, A. Ĭmamoğlu, M. D. Mason, P. J. Carson, G. F. Strouse, and S. K. Buratto, Nature **406**, 968 (2000).
[20] E. Jaynes and F. Cummings, Proc. IEEE **51**, 89 (1963).
[21] M. Brune, F. Schmidt-Kaler, A. Maali, J. Dreyer, E. Hagley, J. M. Raimond, and S. Haroche, Phys. Rev. Lett. **76**, 1800 (1996).

[22] J. M. Fink, M. Göppl, M. Baur, R. Bianchetti, P. J. Leek, A. Blais, and A. Wallraff, Nature **454**, 315 (2008).
[23] A. Faraon, I. Fushman, D. Englund, N. Stoltz, P. Petroff, and J. Vuckovic, Nature Phys. **4**, 859 (2008).
[24] E. del Valle, F. P. Laussy, and C. Tejedor, Phys. Rev. B **79**, 235326 (2009).
[25] G. S. Agarwal and S. Dutta Gupta, Phys. Rev. A **42**, 1737 (1990).
[26] F. P. Laussy and E. del Valle, AIP Conference Proceedings **1147**, 46 (2009).
[27] Y. Ota, N. Kumagai, S. Ohkouchi, M. Shirane, M. Nomura, S. Ishida, S. Iwamoto, S. Yorozu, and Y. Arakawa, arXiv:0906.4622 (2009).
[28] S. Hughes and P. Yao, Opt. Express **17**, 3322 (2009).
[29] B. R. Mollow, Phys. Rev. **188**, 1969 (1969).
[30] A. Auffèves, J.-M. Gérard, and J.-P. Poizat, Phys. Rev. A **79**, 053838 (2009).
[31] M. Nomura, N. Kumagai, S. Iwamoto, Y. Ota, and Y. Arakawa, arXiv:0905.3063 (2009).
[32] D. Sanvitto, F. P. Laussy, F. Bello, D. M. Whittaker, A. M. Fox, M. S. Skolnick, A. Tahraoui, P. W. Fry, and M. Hopkinson, arXiv:cond-mat/0612034 (2006).