

# Quantum emission dynamics from a single quantum dot in a planar photonic crystal nanocavity

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A theoretical quantum-optical study of the modified spontaneous emission dynamics from a single quantum dot in a photonic crystal nanocavity is presented. By use of a photon Green function technique, enhanced single-photon emission and pronounced vacuum Rabi flops are demonstrated, in qualitative agreement with recent experiments. © 2005 Optical Society of America  
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Photonic crystals (PCs) offer a unique light–matter environment for confining and controlling emission of light. Manipulation of the dynamics of atomic spontaneous emission<sup>1</sup> and single-quantum-dot (QD) strong coupling<sup>2</sup> were recently experimentally demonstrated. The ability to trap photons emitted from excited semiconductor nanostructures has been a subject of great interest over the past decade,<sup>3</sup> and it is because of the high-quality fabrication of PCs and new design insights<sup>4</sup> that single-QD-cavity quantum electrodynamics is now possible. Such an intriguing regime offers many exciting prospects in quantum optics, such as a one-dot laser.<sup>5</sup>

One goal in quantum optics is to maintain the photons in the cavity for times longer than the cavity decay or the optical dephasing time; another is to get the photons out as fast as possible. It follows that an overall figure of merit for improvement is the  $Q/V_m$  ratio, where  $Q$  is the quality factor and  $V_m$  is the effective mode volume of the cavity. Optimization of this ratio, with suitably coupled single QDs, is manifest in emitted photons that are essentially trapped for a finite period of time; these photons may then undergo vacuum Rabi flopping before they escape the cavity.<sup>6</sup> Early pioneering experiments with solids exploited the high-quality one-dimensional (distributed Bragg reflector–like) microcavities,<sup>3</sup> whereas iterative improvements have been attempted to reduce cavity size and shape; microcavity posts and planar photonic crystal (PPC) nanocavities currently show the most promise. Remarkably, both PPCs and posts have shown single-QD strong coupling in just the past year, although the latter exploited the increased optical dipole moments of much larger dots.<sup>7</sup> I believe that the PPCs offer a few strong advantages compared with the pillar cavities, including the possibility of in-plane integration with waveguides and amplifiers as well as significantly higher  $Q/V_m$  ratios<sup>4</sup>; PPC nanocavities have shown impressive enhanced emission factors even at room temperature.<sup>8</sup>

Unlike for one-dimensional planar microcavities, however, a physically intuitive and general theoretical description of QD coupling to a complex PPC environment has not yet been introduced. Moreover, single and several QDs coupled to a nanocavity will

ultimately need the intricate tools of quantum optics because, unlike in microcavities, only a few quantum excitations take place. In the domain of PPCs, typical theoretical attempts employ brute-force numerical simulations (usually without a QD and thus neglecting radiative coupling), e.g., a finite-difference time domain<sup>9</sup>; whereas such techniques may seemingly yield the correct answer, there are two main problems that are frequently ignored. First, these numerical simulations employ the ideal structure and thus typically predict unrealistic  $Q$  values; this is hardly surprising, as nanoscale manufacturing imperfections in these PPCs are known to be important. Second, pure numerical investigations offer little—and sometimes no—scientific insight into the underlying physics of coupling QDs to three-dimensional PPC structures. Finally, finite-difference time domains (and the like) cannot directly describe any quantum optics.

To address some of these issues, I recently presented a simple analysis of enhanced emission rates of single QDs in PPC nanostructures by employing a classical Green function theory and the quantum Dyson equation.<sup>10</sup> In that Letter the importance of QD radiative coupling was emphasized, and intuitive formulas to predict when nonperturbative QD coupling becomes important were presented; it was found that the breakdown of Fermi's golden rule occurs even for modest cavity  $Q$  values of  $\sim 4000$ . In this Letter that work is complimented and extended

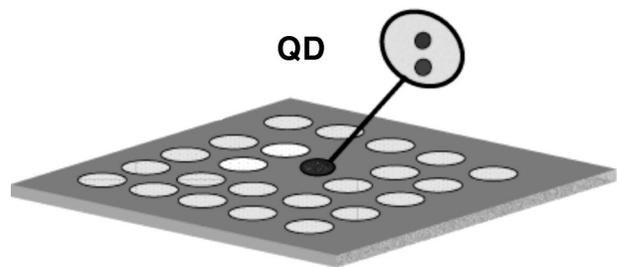


Fig. 1. Schematic of a PPC nanocavity. The light-colored holes represent air, and the deliberately missing holes form a defect nanocavity. The QD (darker-colored hole), containing an excited electron–hole pair (exciton), is situated within this defect.

by presenting a quantum Green function tensor (GFT) formalism that allows one to investigate the quantum dynamics of QDs within PPCs for arbitrary coupling strengths. As before, including manufacturing imperfections is straightforward, as both approaches use the same classical Green functions.

We shall begin by introducing the electric-field photon GFT,  $\vec{\mathbf{G}}(\mathbf{r}, \mathbf{r}'; \omega)$ . The GFT describes the field response at  $\mathbf{r}'$  to an oscillating dipole at  $\mathbf{r}$  as a function of frequency; it can be defined from  $\nabla \times \nabla \times \vec{\mathbf{G}}(\mathbf{r}, \mathbf{r}'; \omega) - \omega^2/c^2 \epsilon_t(\mathbf{r}) \vec{\mathbf{G}}(\mathbf{r}, \mathbf{r}'; \omega) = \omega^2/c^2 \vec{\mathbf{1}} \delta(\mathbf{r} - \mathbf{r}')$ , where  $\vec{\mathbf{1}}$  is the unit tensor and  $\epsilon_t$  is the total relative electric permittivity. The description of spontaneous emission depends largely on the local photon density of states (LDOS), defined as  $\text{LDOS}(\mathbf{r}) \propto \text{Tr}\{\text{Im}[\vec{\mathbf{G}}(\mathbf{r}, \mathbf{r}; \omega)]\}$ . In the regime of cavity QED we are interested in exploiting a large LDOS that can be achieved over a relatively small frequency range. This property is quite natural to PPCs because they can exhibit a photonic bandgap (small LDOS), and one achieves the sudden increase in the LDOS by creating defects. As a PPC is made up of a regular lattice of airholes, defects are created, e.g., by the controlled omission of airholes in a typically high-index semiconductor. One example structure is shown schematically in Fig. 1 for a PPC nanocavity.

It is useful first to make a connection to the classical optical equations and to the expression for photoluminescence measured on a point detector at position  $\mathbf{r}$ . Assuming that one has the GFT of the surrounding environment, the self-consistent field from an embedded QD can be written as  $\mathbf{E}(\mathbf{r}; \omega) = \mathbf{E}^b(\mathbf{r}; \omega) + \int d\mathbf{r}' \vec{\mathbf{G}}^b(\mathbf{r}, \mathbf{r}'; \omega) \cdot \Delta\epsilon_d(\mathbf{r}'; \omega) \mathbf{E}(\mathbf{r}'; \omega)$ , where  $\vec{\mathbf{G}}^b$  is the background GFT and  $\mathbf{E}^b(\mathbf{r}; \omega)$  is a solution before electric permittivity perturbation  $\Delta\epsilon_d$  is added. By exploiting this equation, one can show that the emitted intensity (or photoluminescence) is  $I(\mathbf{r}, \omega) \propto |\vec{\mathbf{G}}^b(\mathbf{r}, r_d; \omega) \cdot \Delta\epsilon_d^r(\omega) \mathbf{E}^b(\mathbf{r}; \omega)|^2$ , where there is a renormalized permittivity (i.e., one that includes radiative coupling)  $\Delta\epsilon_d^r(\omega) = \Delta\epsilon_d(\omega) / [\vec{\mathbf{1}} - V_d \Delta\epsilon_d(\omega) \vec{\mathbf{G}}^b(r_d, r_d; \omega)]$ ,  $V_d$  is the dot volume, and  $r_d$  is the position of the QD. The above notation shows that essentially two GFTs are important for measurements, where  $\vec{\mathbf{G}}^b(r_d, r_d; \omega)$  describes the modified QD dynamics and  $\vec{\mathbf{G}}^b(\mathbf{r}, r_d; \omega)$  accounts for photon propagation from the QD to the detector. We also assume a single QD with a volume and a diameter much smaller than the wavelength of light; thus the local permittivity is  $\Delta\epsilon_d(\omega) = |\mathbf{d}|^2 / [2V_d \epsilon_0 \hbar (\omega_d - \omega - i\Gamma_d)]$ , where  $\Gamma_d$  is the nonradiative decay rate,  $\omega_d$  is the resonance frequency, and  $\mathbf{d}$  is the exciton dipole moment of the lowest-lying exciton that we wish to consider.

Next, the appropriate GFT for the PPC nanocavity is introduced. One can define the fundamental dominant cavity mode normalized from the  $\mathbf{E}$  field of the cavity as  $\mathbf{e}_c = \boldsymbol{\phi}_c / \sqrt{V_m}$ , with  $|\boldsymbol{\phi}_c|^2 = |\mathbf{e}_c|^2 / \max(\epsilon_i |\mathbf{e}_c|^2)$ , where the effective mode volume is  $V_m = \int_{\text{all space}} d\mathbf{r} \epsilon_t(\mathbf{r}) |\boldsymbol{\phi}_c(\mathbf{r})|^2$ . We have  $\vec{\mathbf{G}}^b(\mathbf{r}, \mathbf{r}'; \omega)$

$= \omega_c^2 \mathbf{e}_c(\mathbf{r}) \otimes \mathbf{e}_c^*(\mathbf{r}') / (\omega_c^2 - \omega^2 - i\omega\Gamma_c)$ , where  $\omega_c$  is the cavity resonance frequency and  $\Gamma_c = \omega_c/Q$  is the total cavity linewidth rate; note that the  $\delta(\mathbf{r} - \mathbf{r}')$  contribution to the GFT is neglected because its contribution would merely renormalize the natural exciton transition  $\omega_d$ , which is taken to already contain this quantum correction. Although the GFT above contains only one resonance, it is actually quite typical of a semiconductor PPC defect state that exists deep in the photonic bandgap, and a small finite coupling to radiation modes is contained within the total cavity linewidth rate ( $\Gamma_c = \Gamma_h + \Gamma_v$ , with in-plane decay  $\Gamma_h$  and out-of-plane decay  $\Gamma_v$ ).<sup>11</sup> Dropping the tensor notation, for a QD at the peak antinode field position, the self-energy is  $\Sigma(\omega) = \Omega^2 / (\omega_c^2 - \omega^2 - i\omega\Gamma_c)$ , where  $\Omega^2 = \omega_c |d|^2 / (2\epsilon_0 \epsilon_d \hbar V_m)$  and  $\epsilon_d$  is the background  $\epsilon$  at the QD position. In the limit of small QD dephasing or no dispersion in the surrounding medium, on-resonance vacuum Rabi splitting can occur with a width  $V_R = 2\Omega$ .

We now turn toward a time-dependent quantum-optics approach and take an initially excited level in the QD (with the field in vacuum). Working again with the classical GFT, one can derive the upper-level decay as  $C_u(t) = \int_0^t dt' A_{r_d r_d}(t, t') C_u(t') + 1$ ,<sup>12</sup> where  $A_{r_d r_d}(t, t') = -i / (\hbar \pi \epsilon_0) \int_0^\infty d\omega [1 - \exp[-i(\omega - \omega_d)(t - t')]] \mathbf{d}^* \cdot \text{Im}[\vec{\mathbf{G}}^b(r_d, r_d; \omega)] \cdot \mathbf{d} / (\omega - \omega_d)$ . Note that for the above quantum expressions the role of phonon coupling (or nonradiative decay) has been neglected, which is well justified for low temperatures in high-quality QDs<sup>13</sup>; moreover, in the present case the cavity filters out any interactions with the phonon sidebands,<sup>14</sup> and the cavity results in a significantly faster radiative decay. These expressions can be derived within a quantization scheme that properly includes dispersion and absorption in the medium through the Kramers–Krönig relationship.<sup>12</sup> One can thus solve the entire quantum dynamics for the QD by obtaining the GFT and solving a straightforward Volterra equation of the second kind. In addition, one can derive the quantum emission and spectrum, respectively, as  $I(\mathbf{r}, t) = \epsilon_0 c \langle E^+(\mathbf{r}, t) \cdot E^-(\mathbf{r}, t) \rangle = |(c/\pi) \int_0^t dt' \int_0^\infty d\omega C_u(t') \text{Im}[\vec{\mathbf{G}}^B \times(\mathbf{r}, r_d; \omega)] \cdot \mathbf{d} \exp[-i(\omega - \omega_d)(t - t')]|^2$  and  $I(\mathbf{r}, \omega_s) = \epsilon_0 c \int_0^\infty dt_1 \int_0^\infty dt_2 \{ \exp[i\omega_s(t_2 - t_1)] \langle E^+(\mathbf{r}, t_2) \cdot E^-(\mathbf{r}, t_1) \rangle \}$  for a pointlike photodetector at position  $\mathbf{r}$  and a QD at position  $\mathbf{r}' = r_d$ . It should now be clear that one can describe the QD quantum optical properties to be exploited by the PPC structure semiquantitatively by obtaining the classical GFT and then solving the self-consistent dynamics.

All the required ingredients are now available for investigating single-QD coupling in a PPC nanocavity to arbitrary coupling strengths. Typically, one wishes to maximize the spontaneous emission decay, as this is an important requirement for efficient single-photon emission, and for this we require the relevant classical GFT on-diagonal element. To maximize the coupling, the QD should then be near a field antinode in the nanocavity. The relevant criterion to enhance spontaneous emission enough to achieve

strong coupling is simply  $V_R \gg \Gamma_c, \Gamma_d$ ; this simplified expression also assumes that the QD excitonic transition energy is resonant with the cavity resonance (in practice this is achieved by temperature tuning). Subsequently, by employing typical experimental values of  $\Gamma_t \approx 0.05$  meV ( $Q \approx 15,000$ ),  $\epsilon_d \approx 12$ ,  $V_m \approx 0.05 \mu\text{m}^3$ , and  $d=60$  D, we have the cavity  $V_R \approx 0.3$  meV ( $\gg \Gamma_c, \Gamma_d$ ), which partly explains why single-QD vacuum Rabi splitting was recently observed.<sup>2</sup>

Let us now turn to some concrete numerical examples by solving the quantum dynamic equations above for PPC nanocavities with  $Q=1000, 5000, 15,000$ . In Fig. 2, left, the upper decay level is shown for the QD within the nanocavity; for reference the homogeneous material solution is also shown. Clear vacuum Rabi oscillations are obtained for an increasing  $Q$ , even for the modest  $Q$  values that are now routinely available. Rabi oscillations can also be observed on the corresponding light emission, shown in Fig. 2, center. As strong coupling may in fact not be a desired quantity, e.g., for obtaining a compact single-photon emitter, a theoretical scheme is shown here that allows one to maximize the single photon decay for such an application and thus to explore the full weak-to-strong coupling regimes in a straightforward manner. Remarkably, the enhanced emission factor of the lowest  $Q=1000$  situation already yields impressive Purcell factors of greater than 100. Only a few years ago, *hero* experiments for single QDs in posts were yielding Purcell factors of 5.<sup>15</sup>

Finally, the corresponding spectrum is shown as a function of detuning in Fig. 2, right. The nominal broadening of the single QD in the homogeneous medium is only  $\sim 1 \mu\text{eV}$ , which is close to values for low-temperature measurements.<sup>13</sup> In this noncavity regime one can reliably apply a Markov approximation to the earlier expressions for  $\mathcal{A}(t, t')$  to easily obtain  $|C(t)| = \exp[-(\Gamma + i\delta\omega)t]C(0)$ , where  $\Gamma$  and  $\delta\omega$  are determined from the well-known GFT of a homogeneous medium. Within the cavity, however,  $\Gamma$  increases dramatically through an increase in the LDOS, and eventually the resonance splits into two

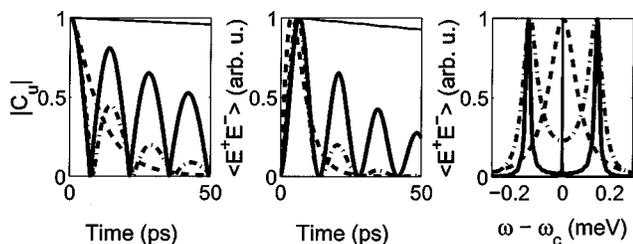


Fig. 2. Left, upper decay of an initially excited quantum dot exciton (on resonance with the cavity) in a photonic crystal nanocavity with  $Q=1000$  (dashed curve), 5000 (dotted-dashed curve), and 15,000 (thick solid curve); the thin solid curve is for a homogeneous medium. Center, corresponding quantum light emission as a function of time at a nearby spatial point. Right, corresponding spectrum (see text).

peaks whose separation is approximately  $V_R$ . Although we have studied the case of  $\omega_d = \omega_c$ , it is also straightforward to explore off-resonant coupling between the QD and the cavity within the above formalism.

Before closing, I remark that it is now possible to obtain the full GFT numerically for arbitrarily shaped nanostructures, such as PPC waveguides posts, as well as systematically accounting for random manufacturing imperfections; the latter effect is actually important for describing quantum optical processes in nanostructures and is already known to play a profoundly important role in the understanding of nanoscale scattering loss.<sup>16</sup> This fact adds further attraction to the methods highlighted here and offers hope of optimizing (and theoretically investigating) such structures in a realistic way.

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