Nonlinear Temporal Dynamics of Strongly Coupled Quantum Dot-Cavity System

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We theoretically analyze and simulate the temporal dynamics of strongly coupled quantum dot-cavity system driven by a resonant laser pulse. We observe the signature of Rabi oscillation in time domain. Under weak driving, the coupled QD-cavity system follows the same dynamics as a set of two classical linear coupled oscillators. Following this, we describe an improved, non-linear semi-classical model that mimics the quantum optical model very well for both very low and high peak intensity of the driving pulse. However, the non-linear semi-classical model deviates from quantum optical description at intermediate peak intensities of the driving pulse and we show that this discrepancy arises from the coherence present in the quantum optical system. Finally, we also present experimental data showing the signature of the Rabi oscillation in time domain.

A single quantum dot (QD) coupled to a photonic crystal micro-cavity constitutes an integrated nano-photonic platform for probing solid state cavity quantum electrodynamic (QED) effects [1]. The eigenstates of this coupled system form an anharmonic ladder, which results in an optical nonlinearity at a single photon level. In recent years, this nonlinearity has been used to perform all-optical [2, 3] and electro-optic switching [4] as well as to generate non-classical states of photons [5–7].

In this paper, we study the temporal dynamics of the coupled dot-cavity system driven by a short laser pulse (Fig. 1 a) using a full quantum optical numerical simulation. The oscillatory behavior of the cavity output (Fig. 1 b), which is caused by the vacuum Rabi splitting, is analyzed at low, intermediate, and high intensity of the driving laser. Specifically, we derive a linear semi-classical description of the system, and show that under weak driving, the coupled QD-cavity system follows the same dynamics as a set of two classical linear coupled oscillators. Following this, we describe an improved, non-linear semi-classical model, that mimics the quantum optical model very well for both very low and high peak intensity of the driving pulse. However, the non-linear semi-classical model deviates from quantum optical description at intermediate peak intensities of the drive pulse and we show that this discrepancy arises from the coherence present in the quantum optical system. Finally, we present a study of the temporal dynamics as a function of the major parameters describing the cavity-QD system as well as experimental data showing the signature of the Rabi oscillation in time domain. Under rotating wave approximation, the quantum-mechanical Hamiltonian \( \mathcal{H} \) describing the coherent dynamics of the coupled system is given by

\[
\mathcal{H} = \omega_\sigma \sigma^\dagger \sigma + \omega_a a^\dagger a + ig(a\sigma^\dagger - a^\dagger \sigma).
\] (1)

Here, \( \omega_\sigma \) and \( \omega_a \) are, respectively, the resonance frequencies of the cavity and the QD; \( a^\dagger \) is the annihilation operator for the cavity mode; \( \sigma = |g\rangle \langle e| \) is the lowering operator for the QD with excited state \( |e\rangle \) and ground state \( |g\rangle \); \( g \) is the cohering interaction strength between the QD and the cavity mode.

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\[
H = \Delta_c a^\dagger a + \Delta_a \sigma^\dagger \sigma + ig(a\sigma^\dagger - a^\dagger \sigma) + i\Omega(t)(a-a^\dagger). \quad (2)
\]

Here, \( \Delta_c \) and \( \Delta_a \) are the detuning of the cavity and the QD resonance from the laser frequency; \( \Omega(t) \) is the maximum laser strength and \( p(t) \) is proportional to the envelope of the laser electric field. The dynamics of the lossy system is determined by using the Master equation

\[
\frac{d\rho}{dt} = -i[H, \rho] + 2\kappa \mathcal{L}[a] + 2\gamma \mathcal{L}[\sigma] \quad (3)
\]

where \( \rho \) is the density matrix of the coupled QD-cavity system; \( \kappa \) is the cavity field decay rate and \( \gamma \) is the dipole spontaneous emission rate. \( \mathcal{L}[\sigma] \) is the Lindblad operator corresponding to a collapse operator \( D \). This is used to model the incoherent decays and is given by:

\[
\mathcal{L}[D] = D\rho D^\dagger - \frac{1}{2} D^\dagger D\rho - \frac{1}{2}\rho D^\dagger D \quad (4)
\]

The Master equation is solved using numerical integration routines provided in quantum optics toolbox, truncating the photon states to 20 photons [8]. This method is completely quantum mechanical, and no approximation (other than the standard Born-Markov approximation and truncation of Fock state basis) is made.

A semi-classical description of the coupled system [9] can be derived by using the relation

\[
\frac{d\langle D \rangle}{dt} = Tr \left[ D \frac{d\rho}{dt} \right] \quad (5)
\]

valid for any operator \( D \). The mean field dynamical equations for the coupled QD-cavity system can then be written as

\[
\frac{d\langle a \rangle}{dt} = -\kappa \langle a \rangle + g \langle \sigma \rangle - \sqrt{\kappa} \Omega(t) \quad (6)
\]

\[
\frac{d\langle \sigma \rangle}{dt} = -\gamma \langle \sigma \rangle + g \langle a\sigma_s \rangle \quad (7)
\]

\[
\frac{d\langle \sigma_z \rangle}{dt} = -2\gamma(\langle \sigma_z \rangle + 1) - 2g(\langle a^\dagger \sigma \rangle + \langle a\sigma^\dagger \rangle) \quad (8)
\]
where, $\sigma_z = |e\rangle \langle e| - |g\rangle \langle g|$. We note that this set of equations is not complete and to exactly solve this, we need to find the equations describing all the other higher order moments, namely $\langle a\sigma_z \rangle$ and $\langle a\sigma_z^\dagger \rangle$. However, in the low excitation regime (no more than 1 photon in the system) the QD will remain mostly in its ground state and we can approximate $\langle \sigma_z \rangle \approx -1$ and replace $\langle a\sigma_z \rangle = -\langle a \rangle$. The resulting set of equations

$$\frac{d\langle a \rangle}{dt} = -\kappa \langle a \rangle + g \langle \sigma \rangle - \sqrt{\kappa} \Omega(t)$$

(9)

is identical to the set of equations describing the dynamics of two coupled linear classical oscillators (see Appendix A). Although this approximation neglects the nonlinear nature of the QD, it matches the actual output quantitatively at low excitation power. Unfortunately, with increasing drive intensities this model fails completely, as the approximation $\langle \sigma_z \rangle \approx -1$ becomes invalid. For sufficiently high drive intensities though $\langle \sigma_z \rangle \to 0$ and equation (10) simplifies to

$$\frac{d\langle \sigma \rangle}{dt} = -\gamma \langle \sigma \rangle$$

(11)

Alternatively, we can retain the dynamics of the $\sigma_z$, while making the set of equations (6), (7), and (8) complete by using the approximations $\langle a\sigma_z \rangle \approx \langle a \rangle \langle \sigma_z \rangle$ and $\langle a^\dagger \sigma \rangle \approx \langle a^\dagger \rangle \langle \sigma \rangle$ [10]. While this approach neglects the coherence of the system while analyzing the mean-field dynamical equations, the nonlinear behavior of the QD is taken into account.

The temporal cavity outputs at low excitation power match very well for the three different models (Fig. 1b). For the numerical simulation, we used a Gaussian pulse with full-width half-maximum (FWHM) of 5 ps as we want to drive the dot-cavity system with a pulse having bandwidth higher than the coupled system (as shown in the inset of Fig. 1b). An oscillation in the cavity output is observed. This oscillation is due to the coherent Rabi oscillation of the photons between the QD and the cavity. Note that further oscillations are quenched by the decay of the cavity field.

To intuitively understand the origin of the oscillation, we analytically solve the linear semi-classical equations to find the eigenvalues of the lossy coupled system as

$$\omega_{\pm} = \frac{\omega_c + \omega_d}{2} - \frac{\kappa + \gamma}{2} \pm \sqrt{\frac{g^2}{4} + \frac{1}{4} (\delta - i(\kappa - \gamma))^2}$$

(12)

where $\delta = \Delta_c - \Delta_a$ is the dot-cavity detuning. When the real part of the expression under the square root is positive, the system is in strong coupling regime and a split resonance appears in the cavity transmission spectrum (inset of Fig. 1b). Without a coupled QD, a single Lorentzian peak is observed in the cavity transmission. The two peaks are entangled states of the QD and the cavity, known as polaritons. When the cavity is driven with a short pulse, having bandwidth more than the coupled system, the cavity output is modulated at the frequency difference between the polaritons, i.e.,

$$2\sqrt{g^2 + \frac{1}{4} (\delta - i(\kappa - \gamma))^2}.$$

Although the non-linear semi-classical model allows QD saturation, it neglects the quantum mechanical coherence between the QD and the cavity. Fig. 2 compares the semiclassical and quantum optical simulations of the coupled dot-cavity system. We find that the results match well both at low (when the QD excited state population is almost zero and $\sigma_z \sim -1$) and high intensities of the drive (when the QD is saturated and $\sigma_z \sim 0$). As expected, the nonlinear semi-classical approach will deviate for intermediate intensities. We plot the quantity $((\langle a^\dagger \sigma \rangle - \langle a^\dagger \rangle \langle \sigma \rangle)/\Omega_0^2$ integrated over time as a function of the driving strength $\Omega_0$ in the inset of Fig. 2. This quantity is zero in absence of any coherence. We observe that this quantity is smaller for both low and high excitation power, compared to the value at intermediate excitations. Note that the onset of increase in the higher excitation power is due to numerical errors caused by the truncated Fock state basis.

Finally, we analyze the dependence of the temporal cavity output as a function of four quantities: dot-cavity detuning $\delta$, the dot-cavity coupling rate $g$, the cavity field decay rate $\kappa$ and pure QD dephasing rate $\gamma_d$. We observe an increase in oscillation frequency (decrease in the time interval between the two peaks) when we increase $g$ (Fig. 3a). This is consistent with the oscillation.
For all the simulations a low excitation power ($\Omega/2\pi = 2$) is assumed. A long pulse does not have sufficient bandwidth to excite both the polaritons (as shown in the inset of Fig. 1b) and the oscillation frequency in the cavity output deviates more from $2g$. To test the validity of our numerical simulations, we experimentally probed a strongly coupled QD-cavity system. A cross-polarized reflectivity setup was used to obtain the transmission of light through the coupled system and the cavity transmission was monitored with a Hamamatsu streak camera. Details of the fabrication and the experimental setup can be found in Ref. [12] with the experimental parameters of the probed dot-cavity system being $g/2\pi = 25$ GHz and $\kappa/2\pi = 29$ GHz [2]. Unfortunately, we did not observe the predicted oscillations in the initial experiments measuring the transmission of 5 ps pulses through the cavity. This was most likely caused by the limited time resolution of our detector. Subsequently the experiment was performed with a longer pulse (40 ps FWHM). A long pulse does not have sufficient bandwidth to excite both the polaritons (as shown in the inset of Fig. 1b) and the oscillation frequency in the cavity transmission is different from the value $2g$, as shown in Fig. 4. Fig. 5a,b,c show the experimentally obtained cavity output for three different excitation powers. The experimental data match qualitatively the predictions from the numerical simulation and clear oscillation is observed in the cavity output. This oscillation disappears with increasing laser power, as expected from the QD saturation. The oscillation period is estimated to be 25 GHz, corresponding to time difference of 39 ps between the two peaks. We note that the numerically obtained plots in Fig. 4 are

period as predicted by the simple linear analysis. At the same time, the oscillation period depends only weakly on $\kappa$ (Fig. 3b). We note that an increasing cavity output with increasing cavity decay rate $\kappa$. This is due to the increasing overlap between the input pulse and the cavity spectrum. The oscillation frequency increases with increasing detuning between the dot and the cavity and when the QD is detuned too far from the cavity, the oscillation almost disappears. This is expected, as with large enough detuning the input pulse is not affected by the QD (Fig. 3c). An important quantity in solid-state cavity QED is pure QD dephasing, which destroys the coherence of the system, without affecting any population of the quantum dot states. The effect of pure QD dephasing can be incorporated by adding a term $2\gamma_d L(\sigma^\dagger\sigma)$ in the Master equation [11], where $\gamma_d$ is the pure QD dephasing rate. Fig. 3d shows the cavity output as a function of pure QD dephasing rate $\gamma_d$ and we observe that the oscillation eventually disappears with increasing dephasing.

Finally, we analyze the dependence of the cavity transmission on the pulse duration (Fig. 4). The pulse duration is changed from 5 ps to 50 ps. We observe that oscillation frequency is decreasing with increasing pulse length. This can be explained by the reduced overlap between the input pulse and the coupled dot-cavity system with reduction in pulse bandwidth. In other words, a long pulse does not have sufficient bandwidth to excite both the polaritons (as shown in the inset of Fig. 1b) and the oscillation frequency in the cavity output deviates more from $2g$. To test the validity of our numerical simulations, we experimentally probed a strongly coupled QD-cavity system. A cross-polarized reflectivity setup was used to obtain the transmission of light through the coupled system and the cavity transmission was monitored with a Hamamatsu streak camera. Details of the fabrication and the experimental setup can be found in Ref. [12] with the experimental parameters of the probed dot-cavity system being $g/2\pi = 25$ GHz and $\kappa/2\pi = 29$ GHz [2]. Unfortunately, we did not observe the predicted oscillations in the initial experiments measuring the transmission of 5 ps pulses through the cavity. This was most likely caused by the limited time resolution of our detector. Subsequently the experiment was performed with a longer pulse (40 ps FWHM). A long pulse does not have sufficient bandwidth to excite both the polaritons (as shown in the inset of Fig. 1b) and the oscillation frequency in the cavity transmission is different from the value $2g$, as shown in Fig. 4. Fig. 5a,b,c show the experimentally obtained cavity output for three different excitation powers. The experimental data match qualitatively the predictions from the numerical simulation and clear oscillation is observed in the cavity output. This oscillation disappears with increasing laser power, as expected from the QD saturation. The oscillation period is estimated to be 25 GHz, corresponding to time difference of 39 ps between the two peaks. We note that the numerically obtained plots in Fig. 4 are

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FIG. 4: (color online) The normalized cavity transmission for different pulse duration. The pulse duration is changed from 5 ps to 50 ps. We observe oscillation in the cavity output, although the oscillation frequency decreases with increasing pulse-width. This can be explained by the reduced overlap between the pulse and the coupled dot-cavity system in frequency domain.

done with very small excitation power. However, the experiment cannot be performed with such low excitation power as the detected signal is too low. Hence, in the experiment, the coupled system is driven close to the QD saturation, and the oscillations are much less visible.

In summary, we have analyzed the nonlinear temporal dynamics of a strongly coupled QD-cavity system driven by a short laser pulse. We showed that this quantum optical system behaves similar to two coupled classical linear oscillators when the system is driven with a weak pulse and that a signature of the vacuum Rabi oscillations can be observed in the time resolved cavity transmission. For high intensity pulse these oscillations die down due to saturation of the QD. We provided a semi-classical non-linear model and showed that in the actual dynamics the role of quantum coherence is important. Lastly, we presented experimental evidence of those oscillations in the cavity output.

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Appendix A: Dynamics of two classical coupled linear oscillators

The dynamics of two classical coupled oscillators, with resonance frequency $\omega_0$ and decay rates $\Gamma_1$ and $\Gamma_2$, are governed by

$$\frac{d^2 x_1}{dt^2} + \Gamma_1 \frac{dx_1}{dt} + \omega_0^2 x_1 + G(x_1 - x_2) = \Omega(t)e^{i\omega_0 t} \quad (A1)$$

and

$$\frac{d^2 x_2}{dt^2} + \Gamma_2 \frac{dx_2}{dt} + \omega_0^2 x_2 + G(x_2 - x_1) = 0, \quad (A2)$$

where $G$ denotes the coupling strength between the oscillators. One of the oscillators is driven resonantly with driving strength $\Omega(t)$, as the cavity is driven by a laser. We assume the solution of the form $x_1(t) = X_1(t)e^{i\omega_0 t}$ and $x_2(t) = X_2(t)e^{i\omega_0 t}$, where $X_1(t)$ and $X_2(t)$ are slowly varying envelopes of the actual oscillator outputs. Then we can write

$$\frac{dx_1}{dt} = i\omega_0 X_1 e^{i\omega_0 t} + \left(\frac{dX_1}{dt} e^{i\omega_0 t}\right)$$

$$\frac{d^2 x_1}{dt^2} = 2i\omega_0 \frac{dX_1}{dt} e^{i\omega_0 t} - \omega_0^2 X_1 e^{i\omega_0 t} + \left(\frac{d^2 X_1}{dt^2} e^{i\omega_0 t}\right)$$

For $x_2$ we can find similar equations. Using slowly varying envelope approximation ($\frac{dX_1}{dt} << i\omega_0 X_1$ and $\frac{d^2 X_1}{dt^2} << i\omega_0 \frac{dX_1}{dt} \omega_0^2 X_1$), we remove the bracketed terms and get the following equations for the un-driven coupled oscillator system:

$$\frac{dX_1}{dt} = -\left(\frac{\Gamma_1}{2} + \frac{G}{2\omega_0}\right) X_1 + \frac{G}{2\omega_0} X_2 + \Omega(t) \quad (A3)$$

and

$$\frac{dX_2}{dt} = -\left(\frac{\Gamma_2}{2} + \frac{G}{2\omega_0}\right) X_2 + \frac{G}{2\omega_0} X_1 \quad (A4)$$

[1] A. Faraon, A. Majumdar, D. Englund, E. Kim, M. Bajcsy, and J. Vučković, New J. Physics 13, 055025 (2011).
[2] D. Englund, A. Majumdar, M. Bajcsy, A. Faraon, P. Petroff, and J. Vučković, arXiv:1107.2956 (2011).
[3] D. Sridharan, R. Bose, H. Kim, G. S. Solomon, and E. Waks, arXiv:1107.3751v1 (2011).
[4] A. Faraon, A. Majumdar, H. Kim, P. Petroff, and J. Vučković, Phys. Rev. Lett. 104, 047402 (2010).
[5] A. Faraon, I. Fushman, D. Englund, N. Stoltz, P. Petroff, and J. Vučković, Nature Physics 450, 859 (2008).
[6] A. Majumdar, M. Bajcsy, and J. Vučković, arXiv:1106.1926 (2011).
FIG. 5: (color online) Experimentally measured time resolved transmission of 40ps pulses through a strongly coupled dot-cavity system for three different powers (averaged over the pulse repetition period): (a) 0.1 nW; (b) 0.23 nW; (c) 1 nW. The powers are measured in front of the objective lens in the confocal microscopy setup. For this specific system cavity field decay rate $\kappa/2\pi = 29$ GHz and coherent dot-cavity coupling strength $g/2\pi = 25$ GHz. Clear oscillations are observed in the cavity transmission, consistent with the theoretical predictions. We also observe decreasing oscillation with increasing laser power due to QD saturation.

[7] A. Reinhard, T. Volz, M. Winger, A. Badolato, K. J. Hennessy, E. L. Hu, and A. Imamoglu, arXiv:1108.3053v1 (2011).
[8] S. M. Tan, Journal of Optics B: Quantum and Semiclassical Optics 1, 424 (1999).
[9] A. Majumdar, N. Manquest, A. Faraon, and J. Vučković, Optics Express 18, 3974 (2010).
[10] M. A. Armen and H. Mabuchi, Phys. Rev. A 73, 063801 (2006).
[11] A. Majumdar, E. Kim, Y. Gong, M. Bajcsy, and J. Vučković, Phys. Rev. B 84 (2011).
[12] D. Englund et al., Nature 450, 857 (2007).