# Strong Coupling between Single Photons in Semiconductor Microcavities

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We discuss the observability of strong coupling between single photons in semiconductor microcavities coupled by a $\chi^{(2)}$ nonlinearity. We present two schemes and analyze the feasibility of their practical implementation in three systems: photonic crystal defects, micropillars, and microdisks, fabricated out of GaAs. We show that, if a weak coherent state is used to enhance the $\chi^{(2)}$ interaction, the strong coupling regime between two modes at different frequencies occupied by a *single* photon is within reach of current technology. The unstimulated strong coupling of a single photon and a photon pair is very challenging and will require an improvement in mirocavity quality factors of 2–4 orders of magnitude to be observable.

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The experimental realizations of strong coupling between a single mode of an optical cavity and a single atom have made it possible to demonstrate striking predictions of cavity quantum electrodynamics (QED) [1]. Quantum information science has since provided motivation for gaining additional control of such strongly and coherently coupled systems, and quantum dots embedded in monolithic optical cavities have emerged as a promising system for the scalable implementation of cavity QED. Motivated in part by this promise, a large effort has been put into the fabrication of small high quality monolithic microcavities [2].

In parallel, a research effort has begun to make use of the high nonlinearities of semiconductor materials such as GaAs to perform classical frequency conversion, using microcavities to enhance the electric field strength and microstructures to provide the necessary phase-matching conditions [3]. Recently, this approach was extended to parametric down-conversion in nonlinear photonic crystals for the generation of entangled photon pairs [4].

Here we discuss the observability of strong coupling *between single photons* in microcavities coupled by an optical nonlinearity, with an emphasis on the implementation in realistic structures.

We consider two schemes: The first consists of two spatially overlapping single-mode cavities (or a doubly resonant cavity) at frequencies $\omega_a$ and $\omega_b$ such that $\omega_a = 2\omega_b$, coupled by a $\chi^{(2)}$ nonlinearity that mediates the conversion of a photon in cavity $a$ to two photons in cavity $b$ and vice versa. The second consists of three overlapping microcavities with frequencies $\omega_{a,b,c}$ satisfying $\omega_a = \omega_b + \omega_c$, with cavity $c$ occupied by a coherent state $|\alpha\rangle_c$. The effective nonlinearity in this case couples the conversion of a single photon in cavity $a$ to a single photon in cavity $b$ and is enhanced by the coherent state in mode $c$.

The dynamics of the two systems are similar. For the sake of clarity, we will therefore solve the dynamics of the two-mode system and then state the corresponding results for the three-mode system. The Hamiltonian for the two-mode system is given by:

$$\hat{\mathcal{H}} = \hbar\omega_a\hat{a}^\dagger\hat{a} + \hbar\omega_b\hat{b}^\dagger\hat{b} + \hbar\Omega(\hat{a}(\hat{b}^\dagger)^2 + \hat{a}^\dagger\hat{b}^2), \quad (1)$$

where $\hat{a}^\dagger$ ($\hat{a}$), $\hat{b}^\dagger$ ($\hat{b}$) represent creation (annihilation) operators for modes $a$, $b$ and $\Omega$ is the strength of the coupling between the modes:

$$\hbar\,\Omega = \epsilon_0\sqrt{\frac{\hbar\omega_a}{2\epsilon_0 n_a^2 V_a}\frac{\hbar\omega_b}{2\epsilon_0 n_b^2 V_b}} \times \int dV\chi^{(2)}_{ijk}(r)E^i_a(r)E^j_b(r)E^k_b(r), \quad (2)$$

where $\chi^{(2)}_{ijk}(r)$ is the nonlinear susceptibility tensor, $\boldsymbol{E}_{a,b}(r)$ represent the spatial part of cavity modes $a$, $b$ normalized so that their maximum value is 1, $V_{a,b}$ represent the mode volumes defined as in Ref. [5], and we have adopted the repeated index summation convention.

Restricting our attention to the subspace $|a\rangle = |1\rangle_a|0\rangle_b$, $|b\rangle = |0\rangle_a|2\rangle_b$, we obtain the following Hamiltonian:

$$\hat{H} = \hbar\begin{pmatrix} \omega_a & \sqrt{2}\Omega e^{i\Delta t} \\ \sqrt{2}\Omega e^{-i\Delta t} & 2\omega_b \end{pmatrix}, \quad (3)$$

where $\Delta = \omega_a - 2\omega_b$ is the detuning between the cavities. This is the well known Hamiltonian for two states $|a\rangle$ and $|b\rangle$ coupled by an interaction $\hbar\sqrt{2}\Omega(|a\rangle\langle b|e^{i\Delta t} + |b\rangle \times \langle a|e^{-i\Delta t})$ first discussed by Rabi [6]. It is analogous to the Jaynes-Cummings Hamiltonian for an atomic transition coupled to a single cavity mode [7], with the role of the excited atom played by the two photons in mode $b$, the role of the cavity photon played by the photon in mode $a$. The eigenstates are time-dependent superpositions of the uncoupled eigenstates $|a\rangle$ and $|b\rangle$, with energies given by:

$$E_\pm = \frac{\hbar}{2}(\omega_a + 2\omega_b) \pm \frac{\hbar}{2}\sqrt{(2\sqrt{2}\Omega)^2 + \Delta^2}. \quad (4)$$

Just as in the atom-cavity case, if the system is prepared in one state, say $|a\rangle$, and $\Delta = 0$, the time evolution will consist of Rabi flopping between states $|a\rangle$ and $|b\rangle$ at twice the Rabi frequency $2\Omega_R = 2\sqrt{2}\Omega$.

 

In practice, the bare cavities will leak photons. The possibility of observing an oscillation will depend on the ratio of the Rabi oscillation period to the cavity decay time. In the context of atom-cavity systems, if the oscillation is in principle observable, the system is said to be in the strong coupling regime. A precise criterion for the discussion of strong coupling in the present system is afforded by solutions of the following master equation [8]:

$$\dot{\hat{\rho}} = -\frac{i}{\hbar}[\hat{H}_{\rm int}, \hat{\rho}] - \frac{1}{2\tau_a}(\hat{a}^\dagger \hat{a}\,\hat{\rho} + \hat{\rho}\hat{a}^\dagger \hat{a}) + \frac{1}{\tau_a}\hat{a}\,\hat{\rho}\,\hat{a}^\dagger - \frac{1}{2\tau_b}(\hat{b}^\dagger \hat{b}\,\hat{\rho} + \hat{\rho}\hat{b}^\dagger \hat{b}) + \frac{1}{\tau_b}\hat{b}\,\hat{\rho}\,\hat{b}^\dagger, \quad (5)$$

where $\hat{\rho}$ represents the reduced density matrix for the two cavities, $\hat{H}_{\rm int}$ represents the interaction part of the Hamiltonian of Eq. (1), and the second and third terms model the loss of photons from cavities $a$ and $b$. If the system is prepared in the state $|1\rangle_a|0\rangle_b$, the four joint states of the cavities relevant to the time evolution are $|1\rangle = |1\rangle_a|0\rangle_b$, $|2\rangle = |0\rangle_a|2\rangle_b$, $|3\rangle = |0\rangle_a|1\rangle_b$, and $|4\rangle = |0\rangle_a|0\rangle_b$. Expressing $\hat{\rho}$ in this basis, and writing out the master equation for each component separately, the following closed subset can be found:

$$\begin{pmatrix} \dot{\rho}_{11} \\ \dot{\rho}_{22} \\ \dot{V} \end{pmatrix} = \begin{pmatrix} -\frac{1}{\tau_a} & 0 & i\sqrt{2}\Omega \\ 0 & -\frac{2}{\tau_b} & -i\sqrt{2}\Omega \\ i2\sqrt{2}\Omega & -i2\sqrt{2}\Omega & -(\frac{1}{\tau_b} + \frac{1}{2\tau_a}) \end{pmatrix} \begin{pmatrix} \rho_{11} \\ \rho_{22} \\ V \end{pmatrix},$$

where $\rho_{ij} = \langle i|\rho|j\rangle$ and $V = \rho_{12} - \rho_{21}$. The matrix elements $\rho_{33}$ and $\rho_{44}$ are determined in turn by $\dot{\rho}_{33} = \frac{2}{\tau_b}\rho_{22} - \frac{1}{\tau_b}\rho_{33}$ and $\dot{\rho}_{44} = \frac{1}{\tau_a}\rho_{11} + \frac{1}{\tau_b}\rho_{33}$.

The eigenvalues of the matrix tell us whether the solutions have the character of a damped oscillation or a critically damped exponential decay. They are given by:

$$\lambda_0 = -\left(\frac{1}{2\tau_a} + \frac{1}{\tau_b}\right), \quad (6)$$

$$\lambda_\pm = -\left(\frac{1}{2\tau_a} + \frac{1}{\tau_b}\right) \pm \sqrt{\left(\frac{1}{2\tau_a} - \frac{1}{\tau_b}\right)^2 - (2\sqrt{2}\Omega)^2}. \quad (7)$$

The time evolution of two of the eigenstates of the matrix will be oscillatory if $2\sqrt{2}\Omega > |(\tau_b - 2\tau_a)/2\tau_a\tau_b|$. The frequency of the oscillation will be $2\Omega_{\rm R} = \sqrt{(2\sqrt{2}\Omega)^2 - [(1/2\tau_a) - (1/\tau_b)]^2}$, and the $1/e$ time of the decay of the oscillation $1/\tau_{\rm eff} = (1/2\tau_a) + (1/\tau_b)$. In the context of the atom-cavity system, oscillatory behavior is synonymous with strong coupling. In the present context, adoption of such a criterion would not be as restrictive as required for it to be meaningful, since $\lambda_\pm$ has an imaginary part for $\tau_b = 2\tau_a$ regardless of whether the Rabi period is at all comparable to the cavity decay time. Note that the definition in the atomic case is meaningful because the atomic lifetime is always much longer than the cavity lifetime, reducing the strong coupling condition to $\sqrt{2}\Omega > \frac{1}{2}|1/\tau_a|$. We therefore suggest the following criterion for strong coupling in this system: $\tau_{\rm eff} \geq \tau_{\rm R}/2 = 2\pi/2\sqrt{2}\Omega$, illustrated in Fig. 1.

An alternative and somewhat less restrictive criterion is provided by the resolvability of the energy splitting [Eq. (4)]. The width of the split energy levels [Eq. (4)] at nonzero detuning can be obtained by evaluating the master equation [Eq. (5)] in the dressed state basis and taking the Fourier transform of the resulting $e^{\lambda_\pm t}$ [8,9]. The result is $\Gamma_\pm^{\rm eff} = (\Gamma_a/2)\frac{\Omega' \pm \Delta}{2\Omega'} + \Gamma_b \frac{\Omega' \mp \Delta}{2\Omega'}$, where $\Omega' = \sqrt{(2\sqrt{2}\Omega)^2 + \Delta^2}$. Figure 1 (inset) shows a plot of the energy eigenstates as a function of detuning accompanied by the FWHM of the lines obtained by this approach. Applying the Rayleigh criterion (separation = FWHM) for the resolvability of the splitting at zero detuning, we obtain the following "spectral" strong coupling criterion: $\pi\tau_{\rm eff} \geq \tau_{\rm R}/2 = 2\pi/2\sqrt{2}\Omega$, illustrated in Fig. 1 (inset).

We now state the results of a similar calculation for the three-mode system, which is governed by the following effective Hamiltonian:

$$\hat{\mathcal{H}} = \hbar\omega_a \hat{a}^\dagger \hat{a} + \hbar\omega_b \hat{b}^\dagger \hat{b} + \hbar\omega_c|\alpha|^2 + \hbar\Omega|\alpha|(\hat{a}\hat{b}^\dagger + \hat{a}^\dagger \hat{b}),$$

where $\hbar\Omega = \epsilon_0\sqrt{\hbar\omega_a/2\epsilon_0 n_a^2 V_a}\sqrt{\hbar\omega_b/2\epsilon_0 n_b^2 V_b} \times \sqrt{\hbar\omega_c/2\epsilon_0 n_c^2 V_c}\int dV \chi_{ijk}^{(2)}(r)E_a^i(r)E_b^j(r)E_c^k(r)$, and $|\alpha|^2$ is the mean photon number in cavity $c$. The dynamics in a similarly chosen subspace: $|1\rangle = |1\rangle_a|0\rangle_b$, $|2\rangle = |0\rangle_a|1\rangle_b$, $|3\rangle = |0\rangle_a|0\rangle_b$ are identical, with the effective decay rate replaced by $\tau_{\rm eff} = (1/\tau_a + 1/\tau_b)^{-1}$ and $\sqrt{2}\Omega$ of Eq. (1) replaced by $|\alpha|\Omega$. It is important to note that mode $c$ need not be a high quality cavity mode; its main purpose is to enhance the nonlinear interaction between cavities $a$ and $b$ and provide the missing energy for the conversion. An

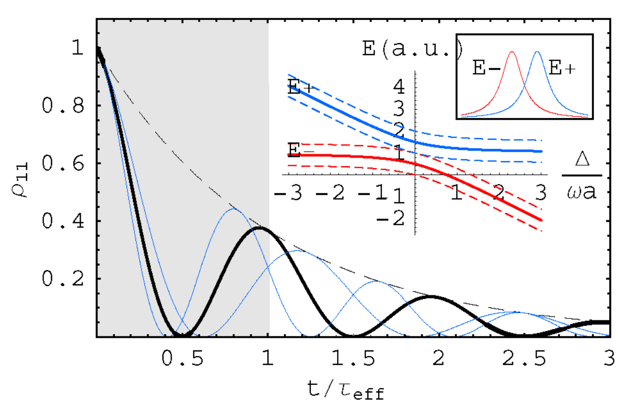


FIG. 1 (color online). Evolution of $\rho_{11}$ for initial conditions $\rho_{11} = 1$, $\rho_{22} = V = 0$. The frequency of the oscillation is $2\Omega_{\rm R} = 2\sqrt{2}\Omega$, corresponding to a period $\tau_{\rm R} = 2\pi/\Omega_{\rm R}$. The $1/e$ time of the decay is given by $\tau_{\rm eff} = (1/\tau_a + 1/2\tau_b)^{-1}$. The strong coupling criterion proposed here consists of requiring that the revival of the oscillation takes place before the $1/e$ time of the decay. Inset: Solid lines: $E_\pm(\Delta)$ for fixed $\omega_a$. Dashed lines: FWHM of $E_\pm$. The energy splitting at zero detuning provides a second, weaker criterion for strong coupling.

implementation of mode $c$ could be a weakly confined laser beam impinging on modes $a$ and $b$.

We now address the role of phase matching, mode overlap, and the tensor nature of $\chi^{(2)}$. In conventional frequency-conversion schemes, the requirement that photons interacting through a $\chi^{(2)}$ nonlinearity phase match can be understood by inspection of the overlap integral for the fields [Eq. (2), for example]. If the eigenmodes correspond to traveling waves, the $\boldsymbol{E}$'s will have the form of complex exponentials in the direction of travel. The overlap is then proportional to a sinc function, leading to the phase-matching requirement $\Delta k \sim 1/L$, where $L$ is the length of the path along which the photons interact. If, however, as is typically the case in the systems considered here, the modes take the form of standing waves, the overlap integral simply takes the form of a spatial overlap of real field amplitudes. Thus, phase matching does not play a role in the systems considered here, but in turn the design of cavities with good overlap becomes of central importance. The polarization of the modes also has to be taken into account, since the $\chi^{(2)}$ interaction is tensorial. This is done by contracting the electric field vectors with the $\chi^{(2)}$ tensor. The effect of this on the value of the overlap integral depends on the detailed geometry of the system and on the symmetry group of the nonlinear material. We will consider, as an example, structures fabricated out of GaAs which has crystalline structure of the $\bar{4}3m$ type. This has the following implications for the values of the components of $\chi^{(2)}$ [10]: $\chi^{(2)}_{xyz} = \chi^{(2)}_{xzy} = \chi^{(2)}_{yxz} = \chi^{(2)}_{yzx} = \chi^{(2)}_{zxy} = \chi^{(2)}_{zyx} = |\chi^{(2)}|$. This makes it comparatively simple to orient the GaAs lattice so that the contraction at any one point does not lead to a reduction in the effective value of the nonlinearity; for example, in the case of all three polarizations being aligned and pointing in the (111) direction, the value of the contraction of the three polarization unit vectors with the $\bar{4}3m$ $\chi^{(2)}$ tensor is $\sim 1.15|\chi^{(2)}|$. The value of $|\chi^{(2)}|$ in GaAs is 200 pm/V at wavelengths of around 1.5 $\mu$m [11], 2 orders of magnitude greater than that of common nonlinear crystals such as $\beta$-barium borate [12]. This high value of the nonlinearity, typical of semiconductor materials, together with the enhancement in the electric field per photon afforded by the microcavity is what makes the proposed schemes viable.

We now turn to a discussion of three systems that could provide a setting for the schemes discussed above, all of which have been used recently to study strong coupling effects between photons and quantum dots [13]; they are photonic crystal defect microcavities [14], microdisks [15], and micropillars [16].

*Photonic crystal defect microcavities (PCDMC).*—PCDMCs are created by removing unit cells from a photonic crystal that has a band gap at the relevant frequencies. They offer an unprecedented ability to control cavity mode volume, polarization, and frequency. They have been demonstrated in photonic crystals consisting of a two-dimensional periodic lattice of holes etched in a thin membrane of GaAs, with confinement in the direction perpendicular to the plane of periodicity provided by total internal reflection. Cavities with quality factors ($Q$) of up to 18 000 [corresponding to confinement times of $\tau(\lambda = 1\ \mu\text{m}) = 9.5$ ps] and mode volumes of $0.7(\lambda/n)^3$ have been demonstrated at a wavelength of 1 $\mu$m [17]. In-and-out coupling can be achieved by integrating waveguides within the photonic crystals [18], through an optical fiber [19], or by free-space optics [17].

The design of PCDMCs with multiple resonances is challenging but does not seem unrealistic. A good starting point is the calculation of the band-gap maps of various defectless lattices, taking into account the finite thickness of the membrane. This is particularly important, since it can lead to a strong modification and in some cases even the closing of higher order band gaps [20,21]. Having found a pattern that has appropriate band gaps, one has to seek high quality defect modes by removing one or more holes. An intuitively interesting class of lattices to pursue are those with two periodicities built into them, such as triangular lattices with a multiple atom unit cell, of which Archimedean lattices [22] are an example. An exciting possibility is also presented by Penrose tiling based photonic quasicrystals, in which a single-frequency cavity has been recently demonstrated [23] and which have been shown theoretically to support modes at widely differing resonant frequencies [24].

To estimate how close the strong coupling regime is to being achievable with current technology, we first consider the three-mode scheme, estimating the Rabi period and comparing it to the cavity lifetime as follows: We assume that a doubly resonant PCDMC can be designed that will support overlapping modes at frequencies $\omega_a$ and $\omega_b$ with $Q$'s similar to those obtained in Ref. [17]. Taking modes $a$, $b$, and $c$ to overlap well, all three polarizations to be the same (TE) and the growth direction to be (111), we estimate the overlap integral in Eq. (2) to be equal to $\frac{1}{2}|\chi^{(2)}_{\text{GaAs}}|V_a$. Taking $V_c = f_c V_a$, where a realistic range for $f_c$ is 1–100, an average of $n$ photons in mode $c$ and $\lambda_b = 1.5\ \mu$m, we then obtain an oscillation period $\tau_{\text{R}}/2 \sim 5\sqrt{f_c/n}$ ns. The corresponding cavity effective lifetime is $\tau_{\text{eff}} = 4.8$ ps. The strong coupling regime is thus within reach with an average of $10^6 f_c$ photons in mode $c$. A similarly constructed estimate in the unseeded case yields $\tau_{\text{R}}/2 \sim 18$ ns, 3 orders of magnitude away according to the spectral criterion.

*Micropillars.*—Micropillars are microscopic cylinders etched out of closely spaced Bragg mirrors, with confinement in the radial direction provided by index contrast. They present clear in-and-out coupling advantages. The design of a doubly resonant Bragg mirror configuration which gives good mode overlap has been studied extensively for the case of parallel mirrors [3] and is readily achievable. A cavity with a $Q$ of 27 700 [$\tau(\lambda = 930\ \text{nm}) = 13.6$ ps] and a mode volume of $100(\lambda/n)^3$ was demonstrated in Ref. [25] at $\lambda = 930$. The corresponding periods are $\tau_R/2 \sim 44\sqrt{f_c/n}$ ns and $\tau_{\rm eff} = 8.0$ ps. The strong coupling regime is thus within reach with an average of $3 \times 10^7 f_c$ photons in mode $c$; in the unseeded case ($\tau_R/2 \sim 177$ ns), it is 4 orders of magnitude away.

*Microdisk resonators.*—Microdisk resonators consist of a thin disk of material, supported by a column. The high-$Q$ modes correspond to whispering gallery modes that hug the outside walls of the resonator. Typically, defects in the microdisks couple counterpropagating modes to create standing wave modes [26]. Doubly resonant ($\omega/2\omega$) microdisks have been demonstrated in Ref. [27]. In-and-out coupling can be achieved by use of a fiber [26]. $Q$'s of 360 000 [$\tau(\lambda = 1.4\ \mu\text{m}) = 267.3$ ps] have been demonstrated in GaAs at a wavelengths of 1.4 $\mu$m [26], with a mode volume of $6(\lambda/n)^3$. Making similar assumptions to those made for the PCDMCs, we obtain $\tau_R/2 \sim 37\sqrt{f_c/n}$ ns and $\tau_{\rm eff} = 95$ ps. The strong coupling regime is thus within reach with an average of only $76 \times 10^3 f_c$ photons in mode $c$, whereas in the unseeded case ($\tau_R/2 \sim 148$ ns), it is only 2 orders of magnitude away.

As a final point, we discuss possible schemes to measure the strong coupling effects presented here. In the spectral domain, one could measure the transmission of the cavities as a function of the detuning between the cavities or as a function of the coherent state intensity. The latter is much simpler but can be implemented only in the three-mode scheme. In the time domain, one could initiate the coupled system with a photon in one of the modes, for example, by sending an appropriately shaped pulse into one of the cavities; one could then wait and measure the photon emission from cavities $a$ and $b$ as a function of time. A simpler alternative that works in the three-mode case is to send a photon into cavity $a$ and then apply a Rabi $\pi$ pulse through cavity $c$ to deterministically convert the photon in mode $a$ to a photon in mode $b$.

In conclusion, we have discussed the observability of strong coupling between single photons in semiconductor microcavities coupled by an optical nonlinearity. We have shown that, if the process is stimulated by a weak coherent state, the strong coupling regime is within reach of current technology. Engineering structures in which the unstimulated process could be observed appears to be a challenging goal for years to come. The observation of such a coupling would constitute a new regime for photons in quantum optical systems. Aside from the design of structures optimized for the implementation of the schemes presented here, an extension of the present work would be the investigation of ways to further enhance the nonlinearities by engineering the material properties, ways to integrate sources, such as quantum dots, with the present schemes, and ways to implement quantum logic gates between strongly coupled single photons.

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