Entanglement dynamics and multiple Mollow triplets between two coupled quantum dots in a nanowire photonic crystal system

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We introduce a nanowire-based photonic crystal waveguide system capable of deterministically mediating the photon coupling between two quantum dots that are macroscopically separated. Using a rigorous Green-function-based master equation approach, this two dot system is shown to provide a wide range of interesting quantum dynamical regimes. In particular, we demonstrate the formation of long-lived entangled states and study the resonance fluorescence spectrum which contains clear signatures of the coupled quantum dot pair. Depending upon the operating frequency, one can obtain a modified Mollow triplet spectrum or a Mollow nonuplet, namely nine spectral peaks. These multiple peaks are explained in the context of photon-exchange-mediated dressed states. Results are robust with respect to loss, and spectral filtering via propagation allows for each dot’s emission to be observed individually.

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The ability to mediate coupling and entanglement between qubits is important for optical quantum computation systems [1, 2]. In particular, it is desirable that future quantum information systems are scalable, and should operate on-chip, where, e.g., photons are manipulated in the plane of a waveguide. In addition, the ability to produce and maintain entanglement between spatially separated qubits is required, both for measurement purposes and to permit individual control of separated qubits.

Photonic crystal (PC) slabs [3–5] with embedded quantum dots (QDs) are a strong candidate for on-chip quantum information systems [6], since they have the ability to modify the local optical density of states (LDOS) through integrated cavities and waveguides. Systems containing a single QD coupled to a PC antinode can operate as a single photon source and facilitate the strong coupling regime [6–8]. However, the traditional slab PC platform, comprised of a periodic array of holes in a semiconductor slab, and other semiconductor cavity systems, has yet to demonstrate systems coupling multiple QDs in a controlled way. This is largely due to the Stranski-Karatanow growth technique, where self-assembly of QDs results in limited control over their position and emission frequency and poor coupling to PC waveguide modes [6–8], such that coupling has so far been demonstrated between QDs only in a shared cavity [6, 10]. Systems that couple individual QDs via an arbitrary length PC waveguide mode [11] are desirable, offering the ability to excite and probe individual QDs. Coupling QDs using plasmonic waveguides has been proposed [12], though metallic waveguide systems suffer from material losses and Ohmic heating.

Photonic crystal structures comprised of arrays of dielectric rods [5] offer an alternative to the traditional slab design [13, 14]. Moreover, semiconductor nanorod and nanowire (NW) fabrication techniques have seen dramatic improvements in recent years [15, 16] and the ability to produce QDs of deterministic position and optical properties in NWs has been demonstrated both during MBE growth [16, 17] and via post-process [18]. Deterministic emitter placement has also been shown for NV centers in diamond NWs [19].

In this Letter, we introduce a chip-based system comprised of a finite-size nanowire PC waveguide with a pair of embedded QDs at opposite ends. Figure 1 shows a schematic of our proposal, which can be fabricated using current growth techniques. The geometry exploits the large spontaneous emission (SE) enhancements and a near lossless waveguide mode of the NW PCs [20]. Using a quantum master equation formalism centered on the photonic Green function, we demonstrate that this system can strongly couple a pair of qubits. We also study the fluorescence spectrum emitted from the device, which displays signatures of nonlinear coupling via photon transport. In particular, we introduce a unique regime of cavity QED, where a “Mollow nonuplet” (i.e., with nine spectral peaks) is obtained via significant photon exchange.

The coupling dynamics of N QDs (treated as two-level artificial atoms in the dipole approximation) in an arbitrary medium with permittivity ε, is governed by the Hamiltonian [21]:

\[ H = \int d^3 r \int_0^\infty d \omega \omega F^\dagger(r, \omega) F(r, \omega) + \sum_n \hbar \omega_n \hat{\sigma}_n^+ \hat{\sigma}_n - \sum_n \int_0^\infty d \omega (\hat{d}_n \hat{E}(r_n, \omega) + H.c.), \]

where the
nth QD is at position \( r_n \), with resonance \( \omega_n \), and the dipole operator \( d_n = d_n (\hat{\sigma}_n^- + \hat{\sigma}_n^+) \), where \( d_n \) is the dipole moment of QD \( n \); \( \hat{f} \) is a vectorial bosonic field annihilation operator, related to the electric field operator via \([21]\)

\[
\hat{E}(r, \omega) = i \sqrt{\frac{n_c}{\pi \epsilon_0}} \int d^3r' \sqrt{\epsilon_f} (r', \omega) \hat{G}(r, r'; \omega) \hat{f}(r', \omega),
\]

where \( \hat{G}(r, r'; \omega) \) is the electric-field Green function, describing the system response at \( r \) to a point source at \( r' \). Working in a rotating frame with respect to a laser frequency \( \omega_L \), we derive the quantum master equation for a system by applying the second-order Born and Markov approximations to the dipole operator \([21]\):

\[
\dot{\rho} = -i \sum_{n} \Delta \omega_n [\hat{\sigma}_n^+ \rho \hat{\sigma}_n^- - \hat{\sigma}_n^- \rho \hat{\sigma}_n^+] - i \sum_{n \neq n'} \delta_{n,n'} \{ \hat{\sigma}_n^+ \hat{\sigma}_n^- \rho - \rho \hat{\sigma}_n^+ \hat{\sigma}_n^- \}

+ \Gamma_{n,n'} \{ \hat{\sigma}_n^+ \rho \hat{\sigma}_n^- - \hat{\sigma}_n^- \rho \hat{\sigma}_n^+ \} - \frac{i}{\hbar} \{ H_{\text{drive}}, \rho \} + \sum_n \gamma_n \{ \hat{\sigma}_n^+ \rho \hat{\sigma}_n^- - \hat{\sigma}_n^- \rho \hat{\sigma}_n^+ \},
\]

where \( \Delta \omega_n = (\omega'_n - \omega_L) \), \( \omega'_n = \omega_x + \Delta_n \), and \( \Delta_n = \frac{\hbar}{\epsilon_0} d_n \cdot \text{Re} \{ \hat{G}(r_n, r_n; \omega^2) \} \cdot d_n \) is the photonic Lamb shift; the inter-QD coupling terms are \( \delta_{n,n'} \) and \( \Gamma_{n,n'} = \frac{\hbar}{\epsilon_0} d_n \cdot \text{Im} \{ \hat{G}(r_n, r_n; \omega^2) \} \cdot d_n \). The pump term \( H_{\text{drive}} = \sum_n \frac{\hbar^2}{\epsilon_0} \{ \hat{\sigma}_n^+ + \hat{\sigma}_n^- \} \) represents the external coherent drive applied to each QD at laser frequency \( \omega_L \), where the effective Rabi field \( \Omega_{R,n} = |E_{\text{pump},n}(r_n) \cdot d_n|/\hbar \). [22]

In the above derivation, the Rabi fields and coupling terms are smaller than the scale over which an appreciable change in the LDOS occurs so that the scattering rates are essentially pump independent \([22]\) and the Born and Markov approximations are rigorously valid \([22]\). We use the scattered part of the Green function and thus subtract off the divergent homogeneous vacuum Lamb shift, which is already included in \( \omega_x \). To better highlight the radiative coupling dynamics, we also neglect pump-induced dephasing effects (e.g., through phonon-induced interactions). However, the final term in Eq. \([11]\) accounts for pure dephasing via the standard Lindbladian superoperator \( \mathcal{L}(\hat{O}) = (\hat{O} \rho \hat{O}^\dagger - \frac{1}{2} \{ \hat{O}^\dagger \hat{O}, \rho \}) \), with \( \gamma_n \) the pure dephasing rate of QD \( n \). Importantly, Eq. \([11]\) allows one to analyze the radiative coupling dynamics of a system of QDs in an arbitrary dielectric bath medium such as a PC waveguide, where all of the coupling depends explicitly on the medium Green functions.

Figure \([1\text{a}]\) shows a schematic of the proposed PC NW waveguide. In our specific design, the waveguide has a length and width of \( 41a \) and \( 7a \), with lattice constant \( a = 0.5526 \mu \text{m} \) to produce a waveguide mode edge near the telecom wavelength of 1550 nm. As described previously \([28]\), a waveguide is formed by reducing the radius of a single row of NWs, from \( r_h = 0.180a \) to \( r_d = 0.140a \). Light remains confined to the higher index (GaAs, \( \epsilon = 13 \)) upper portion (height \( 2.27a \)) of the NWs, and the lower index portion (AlO, \( \epsilon = 3.1 \) height \( 2a \)) separates the NWs from the substrate below. We consider a pair of QDs that are embedded post-process at the top of selected NWs, where they efficiently couple into the waveguide Bloch mode anti-node as depicted schematically in Fig. \([1\text{b}]\). Each QD resides on top of a NW 10 unit cells from the center of the structure (separated by \( 21a \), 10.6 \( \mu \text{m} \)). The relevant \( \hat{G} \) components for the two QDs indicated, found using a numerical finite-difference time-domain (FDTD) approach \([6, 20]\), are shown in Fig. \([2\text{b-d}]\) through the waveguide band. We note that the largest LDOS peak corresponds to the quasi mode formed at the mode edge of a slow-light waveguide mode \([20]\), whereas the lower frequency peaks are Fabry-Pérot ripples due to facet reflections \([20]\). Optimal coupling is achieved by choosing the mode which maximizes the symmetric photon exchange terms, \( \text{Im} \{ \hat{G}(r_{1/2}, r_{1/2}; \omega) \} \). The photonic mode \( \lambda_j \) which best achieves this is shown in Fig. \([2\text{a}]\), containing anti-nodes at the symmetric QD positions.

We first consider the initial condition of a single excited QD (QD 1) and no external drive. We also include a pure dephasing rate of 1 \( \mu \text{eV} \) in all calculations, similar to experimental numbers on InAs QD at 4 K \([23]\). Both QDs were taken to have a dipole moment of \( d = 30 \) Debye (0.626 \( \mu \text{eV} \)) oriented in the vertical direction, and a renormalized exciton line at \( \omega'_1 = \omega_{\lambda_1} = 793.40 \text{meV} \). The calculated SE rates and exchange terms, in units of \( \text{Im} \{ \hat{G}(r,r;\omega'_j) \} = \omega'_j n^3_{\text{R}}/(6\pi e^2) \) \([26]\), where \( n^3_{\text{R}} \) is the refractive index at the QD position, are 131.7 and 129.8, respectively, for the chosen positions and frequencies. This indicates that the SE rate of the QDs has been increased by two orders of magnitude relative to a homogeneous medium, with 98.6\% of emitted photons propagating to the other QD (via the \( \Gamma_{n,n'} \)). This is quite remarkable given the openness of the structure and the large spatial separation of the QDs. Having obtained the Green functions of the photon system, we solve the master equation (Eq. \([11]\)) to obtain the density matrix \( \rho(t) \) \([27]\), which is used to obtain the population of each QD from \( \langle n_{\text{R}}(t) \rangle = |\text{Tr}[\hat{\sigma}_n^+ \rho \hat{\sigma}_n^-]| \). We also calculate the system concurrence \( C \) \([28]\), to obtain a measure of the entanglement between the two QDs. The results are shown in Fig. \([3\text{a}]\), with a long-lived entangled state clearly forming as the QDs couple resonantly to the waveguide mode and exchange their single excitation, extending its lifetime dramatically.

We next examine the dynamics of the symmetric and asymmetric entangled states \( |\psi_{\pm}\rangle = \frac{1}{\sqrt{2}}(|0,1\rangle \pm |1,0\rangle) \), where the first (second) quantum number refers to the first (second) QD. For this system, \( |\psi_{\pm}\rangle \) populations decay at \( \Gamma_{\pm} = \Gamma_{1,1} \pm \Gamma_{1,2} \) \([11,21]\). Due to the phase difference in the effective Bloch mode between the two QD positions, \( \text{Im} \{ \hat{G}(r_2,r_1;\omega) \} \), and thus \( \Gamma_{1,2} \), is negative. The symmetric state \( |\psi_{+}\rangle \) thus has a longer lifetime than \( |\psi_{-}\rangle \) or even \( |1,0\rangle \), as shown in Fig. \([1\text{D}]\). We note that by changing the positions or resonance of the QDs, one can invert this relationship such that the asymmetric state will have the dramatically longer lifetime, a feature that has useful quantum information applications.

To investigate the nonlinear coupling dynamics, we consider a cw pump field applied to one of the QDs, and
calculate the resulting spectrum. In order to determine the emitted spectra, we assume a detector \( D \) is placed \( 1 a (0.5526 \mu m) \) from the terminus of structure, along \( y = z = 0 \) (see Fig. 2(a)), on the side closer to QD 2. The incoherent spectrum from QD \( n \) at a point detector at position \( r_D \), including the effects of spectral filtering via light propagation, is \( S_{D,n}(\omega) = \frac{2}{\epsilon_0} |G(r_D, r_n; \omega) \cdot d_n|^2 S_{0,n}(\omega) \), where \( S_{0,n}(\omega) = \lim_{t \to \infty} \text{Re}\left\{ \int_0^\infty d\tau (\hat{\sigma}_n^+ (t + \tau) \hat{\sigma}_n^- (t)) e^{i(\omega - \omega_0)\tau} \right\} \). The total spectrum is simply \( S_D = S_{D,1} + S_{D,2} \).

Figure 2(a) shows the dynamics for QD 1 driven by a \( \Omega_R = 0.025 \text{ meV} \) pump at \( \omega_L = \omega_{\lambda_1} + \delta_{1,2} \) (with both QDs initially in the ground state). The dipole moment has been increased to an experimentally accessible 60 Debye to better highlight exchange effects, but all other parameters remain the same. It can be seen that a highly entangled state is formed with steady state \( \langle n_1 \rangle, \langle n_2 \rangle \), and \( C(\rho) \) of 0.27, 0.23, and 0.45 respectively. We note that the strong medium-assisted photon exchange leads to Rabi oscillations and steady-state populations in the unpumped QD 2 almost identical to that of QD 1, and the chosen \( \omega_L \) maximizes the steady-state \( C(\rho) \). Figure 2(b) displays the spectra emitted from both QD 1 and QD 2, as well as that emitted from an identical system containing only QD 1. The Mollow triplet is clearly observed in both QDs, despite the lack of external Rabi field on QD 2. The dynamics are dominated by the \( \Gamma \) exchange terms, with \( \Delta_{R,2} = 0.0882 \text{ meV} \), and \( \Omega_R = 0.025 \text{ meV} \). In addition the sideband splitting has been reduced from the traditional separation of \( \Omega_R \) due to this resonant photon exchange, with \( \Delta_{R,2} = 0.701\Omega_R \) and \( \Delta_{R,1} = 0.704\Omega_R \). In particular, the Rabi field seen by QD 2 is due entirely to photons emitted from QD 1 via \( \Gamma_{1,2} \), and the Rabi filed at QD 1 is similarly dominated by the \( \Gamma_{2,1} \) process, although has been increased slightly by the laser pump. The asymmetry in \( |G(r_D, r_n; \omega) \| \), which increases with \( \omega \) near \( \omega_L \) and is stronger for \( n = 2 \), leads to the slight asymmetry in sideband height and enhancement in \( S_{D,2} \). As the position and intensity Mollow sidebands are directly dependent on the strength of the pump and exchange term, one can experimentally study the coupling dynamics of this system by measuring the spectra emitted from each QD when the other one is pumped.

Lastly, we study a system where in contrast with the previous cases, we work in a regime with \( \delta_{1,2} >> \Gamma_{1,1} \), by choosing \( \omega_L = \omega_{\lambda_1} = 794.19 \text{ meV} \) and \( d = 60 \text{ Debye} \). This allows exchange splitting to dominate the system.
FIG. 5. (Color online) (a) Energy levels and expected transitions for system evolving under $H_{\text{eff}}$. Unprimed transitions are from the interaction picture, and primed are found when one considers the full, time-dependent Hamiltonian. The four-fold degenerate transition between identical levels at $\omega_L$ is not labeled. (b) The detected spectrum from QD 1 (2) in solid blue (dashed red) with dynamics in the inset. Population of QD 1 (2) is again in dashed red (solid blue) and concurrence is in green (light). All expected transitions are evident, and the unique dynamics of each QD can be observed.

dynamics, with $\delta_{1,2} = -9.68 \mu eV$, $\Gamma_{1,1} = 0.64 \mu eV$, $\Gamma_{1,2} = 0.41 \mu eV$. We note that this exchange splitting is on the order of that reported for neighboring QDs $|2\rangle$, despite the large spatial separation in our device. In this regime, the system largely evolves under $H_{\text{eff}} = \hbar \omega_L \sigma_z + \hbar \delta_{1,2} \sigma_y + \hbar \delta_{1,1} \sigma_x + \hbar \Delta \sigma_z + \hbar \omega$. With no pump, the eigenstates of $H_{\text{eff}}$ are simply $|0,0\rangle$, $|\psi_+\rangle$, and $|1,1\rangle$ with energies $0$, $\hbar \omega_L$, $\hbar \delta_{1,2}$, and $2 \hbar \omega_L$ respectively, mimicking a biexcitonic cascade system with level splitting. For non-zero pump, we find a Stark-shifted level structure, $E_i/\hbar = \Delta \omega_L \pm \sqrt{A \pm B}$, where $A = 2 \delta_{1,2}^2 + 2 \Delta \omega_L^2 + \Omega_R^2$, and $B = 2 \sqrt{\delta_{1,2}^4 + \delta_{1,2}^2 \delta_{1,1}^2 - \delta_{1,2}^2 \Delta \omega_L^2 - \Omega_R^2 \Delta \omega_L^2 + \Delta \omega_L^4}$. As $\Omega_R \rightarrow 0$, it can easily be shown that one recovers the exchange-splitting result. The temporal periodicity of the original Hamiltonian allows one to treat it in the Floquet picture, resulting in an infinite sequence of the interaction picture energy levels centered at $n \hbar \omega_L$, where $n$ is an integer. Since only QD 1 is driven, we can truncate this sequence to the $n = 0$ and $n = 1$ sets, corresponding to the absorption of 0 or 1 photons from the laser. This spectrum is shown in Fig. 4(b) where the four unique transitions of the interaction picture, labeled $a$–$d$, are seen to lead to a nine-peaked observable spectrum of the full Hamiltonian, with the ninth peak being a four-fold degenerate transition at $\omega_L$.

We solved the dynamics of the above system, with $\Omega_R = 10 \mu eV$ and $\Delta \omega_L = 0$, and the resulting detectable spectrum, populations, and concurrence is shown in Fig. 4(b). We calculate interaction energy levels of $E_i = \pm 1.18 \Omega_R, \pm 0.212 \Omega_R$, indexed in increasing energy, and thus clear signatures of all the expected transitions are observed. $|\psi_+\rangle$ and $|\psi_\downarrow\rangle$ correspond to asymmetric and symmetric combinations of the biexciton and vacuum state, yielding for zero pump correlated exciton states $|1,1\rangle + |0,0\rangle$. As such, transitions $b$ and $c$ correspond to transitions between correlated and anti-correlated exciton states, with $b$ maintaining parity, whereas transitions $a$ and $d$ are between symmetric and asymmetric states, which are anti-correlated for $a$ and correlated for $d$. We note that the $a$ and $d$ transitions do not appear in the QD 2 spectrum, as they have no effect on the state of QD 2. These peaks are robust with respect to pure dephasing; we stress that we are using an experimentally viable $\gamma' = 1 \mu eV$ in the above work, and a numerical study has indicated that these peaks remain resolvable up to $\gamma' \approx 5 \mu eV$.

Of particular importance for this specific system is the spectral filtering of respective QDs. Specifically, we define a second detector $D'$ at the mirror position of $D$, i.e., closer to QD 1. Throughout the frequency range of interest, we find $\left|G(r_D, r_1; \omega)\right| = \left|G(r_D, r_2; \omega)\right| \approx 8\left|G(r_D, r_1; \omega)\right| = 8\left|G(r_D, r_2; \omega)\right|$. In consequence, the spectra of QDs 1 and 2 in Fig. 4(b) correspond almost exactly with the total spectra observed at $D$ and $D'$ respectively, indicating that QDs can be observed individually by taking advantage of the inherent structural spectral filtering. We stress that one cannot isolate individual QD spectra in a comparable cavity structure, with the strong spectral filtering originating from the rich LDOS of the finite-sized waveguide. Furthermore, this exchange splitting regime is wholly inaccessible in a simple cavity structure, as $\text{Re} \{G\}$ falls off rapidly away from the peak of a Lorentzian LDOS, resulting in dynamics which are unavoidably dominated by the more rapid $\Gamma$ processes. As such, the multiply-peaked spectra of Fig. 4(b) is unique to our proposed PC waveguide structure. The ability to model an effective four-level system and separately observe each component makes these structures promising for applications in quantum information science.

In conclusion, we have analyzed and explored the quantum dynamics of a pair of QDs embedded in a finite-sized NW PC. Our theoretical formalism fully includes factors such as finite-size effects, and radiative loss, through the use of the numerically exact structural Green function. By operating at a spectral resonance of the waveguide, we showed the ability to produce a long-lived entangled state and a photon-exchange-dependent Mollow triplet in the spectrum of a unpumped QD. We then tuned the pump and QD resonances to a broad waveguide band region and showed how the system evolves largely under the dynamics of the controllable system Hamiltonian. Under certain excitation conditions, we can observe nine spectral peaks which are explained using a dressed-state picture. Spectral filtering, naturally provided by the waveguide structure, allows the spectrum and dynamics of each QD to be observed independently. This versatility makes these structures attractive for use as quantum simulators or to study quantum dynamics in lab.

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