Mollow quintuplets from coherently excited quantum dots

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Charge-neutral excitons in semiconductor quantum dots (QDs) have a small finite energy separation caused by the anisotropic exchange splitting. Coherent excitation of neutral excitons will generally excite both exciton components, unless the excitation is parallel to one of the dipole axes. We present a polaron master equation model to describe two-exciton pumping using a coherent continuous wave pump field in the presence of a realistic anisotropic exchange splitting. We predict a five-peak incoherent spectrum, namely a Mollow quintuplet under general excitation conditions. We experimentally confirm such spectral quintuplets for In(Ga)As QDs and obtain very good agreement with theory.

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Quantum light-matter interaction in semiconductor nanostructures is a topic of considerable interest. An understanding of the fundamental light-matter interactions is important for future quantum devices, such as single photon emitters [1,2] and entanglement-based sources [3,4]. Due to efficient semiconductor growth technology and natural scalability, semiconductor quantum dots (QDs) offer an excellent opportunity to explore rich light-matter interaction in a solid state environment. It is well known that under high-field coherent optical pumping, the fluorescence spectrum of a two-level system develops a symmetric "Mollow triplet" structure [5] with two outer sidebands split by twice the Rabi frequency. While driven QDs, also referred to as "artifial atoms," do show behavior similar to a driven two-level atom [6,7], due to the coupling of the exciton to a phonon reservoir [8–13], both the position and width of the Mollow triplets can change significantly [14–17]. Moreover, real QDs have many exciton states, and e.g., charge-neutral excitons are split by a small anisotropic exchange energy. Dressed-states in the presence of an anisotropic exchange splitting have also been observed experimentally [18,19].

In this Letter we develop and exploit a polaron master equation to include two neutral excitons and model the ensuing incoherent spectrum. We generalize the well known Mollow triplet to a *Mollow quintuplet* regime caused by a sum of two separate Mollow triplets with a similar central resonance. Mollow quintuplets have already been measured in the context of spin-resolved resonance fluorescence [7]. A useful analytical expression for the incoherent spectrum is given and compared to experiments, where we find very good qualitative agreement.

Figure 1 shows a schematic of the excitation geometry. Neglecting QD zero-phonon-line (ZPL) decay mechanisms, and working in a rotating frame with respect to the pump laser frequency ω_L, the two-exciton Hamiltonian is given by $H=\sum_{\mathbf{q}}\hbar\omega_{\mathbf{q}}b_{\mathbf{q}}^{\dagger}b_{\mathbf{q}}-\sum_j\hbar\Delta_j\sigma_j^+\sigma_j^-+\sum_j(\hbar\Omega_j^0/2)(\sigma_j^++\sigma_j^-)+\sum_{j,\mathbf{q}}\hbar\lambda_{j\mathbf{q}}\sigma_j^+\sigma_j^-(b_{\mathbf{q}}^{\dagger}+b_{\mathbf{q}})$, where $\Delta_j\equiv\omega_L-\omega_{x_j}$ $(j=1,2)$ are the laser-exciton detunings, $b_{\mathbf{q}}$ $(b_{\mathbf{q}}^{\dagger})$ are the annihilation (creation) operators of the phonon bath, σ_j^+,σ_j^- are the Pauli operators of the jth exciton, $\lambda_{j\mathbf{q}}$ are the exciton–phonon coupling strength, and Ω_j^0 are the Rabi frequencies of the pump. We consider two excitons coupled to a common phonon bath.

Adopting a polaron transformation $H'=e^SHe^{-S}$, with $S=\sum_{j\mathbf{q}}\sigma_j^+\sigma_j^-(\lambda_{j\mathbf{q}}/\omega_{\mathbf{q}})(b_{\mathbf{q}}^{\dagger}-b_{\mathbf{q}})$, the exciton–phonon coupling can be taken into account nonperturbatively [11,12]. To second-order in the polaron-transformed exciton–phonon interaction, the polaron master equation can be written as $\partial\rho(t)/\partial t=(-i/\hbar)[H'_{\mathrm{sys}},\rho(t)]+\sum_j(\gamma_j/2)\mathcal{L}[\sigma_j^-]+\sum_j(\gamma'_j/2)\mathcal{L}[\sigma_j^+\sigma_j^-]-(1/\hbar^2)\int_0^\infty d\tau([H_I,H_I(-\tau)\rho(t)]+\mathrm{H.c.})$, where the upper time limit of the time integral, $t\to\infty$, since the relaxation time of acoustic phonon bath is very fast (a few ps). The polaron-modified system Hamiltonian, $H'_{\mathrm{sys}}=-\sum_{j=1,2}\hbar(\Delta_j+\Delta_p)\sigma_j^+\sigma_j^-+$

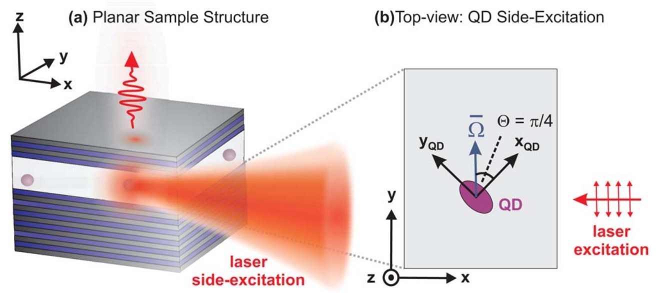

Fig. 1. Schematic of the QD under continuous wave pumping with a field that is polarized along, e.g., $\theta=(\pi/4)$ in a weakly coupled planar cavity. This QD can be excited with electric dipole along the x or y axis; the double arrows show the polarization of the laser.

$\sum_j(\hbar\Omega'_j/2)(\sigma_j^+ + \sigma_j^-)$, where the renormalized Rabi frequency is given by $\Omega'_j = \Omega_j^0\langle B\rangle$, with $\langle B\rangle = \exp[(1/2)\int_0^\infty d\omega(J(\omega)/\omega^2)\coth(\hbar\omega/2k_bT)]$. We chose a phonon spectral function that accounts for exciton-LA interactions from the deformation potential, $J(\omega) = \alpha_p\omega^3\exp(-\omega^2/2\omega_b^2)$ [20], where ω_b is the phonon cutoff frequency and α_p characterizes the strength of the exciton–phonon interaction. We also include Lindblad scattering terms, $\mathcal{L}[D] = (D\rho D^\dagger - D^\dagger D\rho) + \text{H.c.}$, which describe ZPL radiative decay and ZPL pure dephasing. For convenience, we will include the polaron shift $\Delta_p = \int_0^\infty d\omega(J(\omega)/\omega)$ into Δ_j below.

For Rabi fields much less than ω_b, one can derive an *effective phonon master equation* [13,14],

$$\frac{\partial\rho}{\partial t} = \frac{-i}{\hbar}[H'_{\text{sys}},\rho] + \sum_j\frac{\gamma_j}{2}\mathcal{L}[\sigma_j^-] + \sum_j\frac{\gamma'_j}{2}\mathcal{L}[\sigma_j^+\sigma_j^-] + \sum_j\frac{\Gamma_{\text{ph}}^{\sigma_j+}}{2}\mathcal{L}[\sigma_j^+] + \sum_j\frac{\Gamma_{\text{ph}}^{\sigma_j-}}{2}\mathcal{L}[\sigma_j^-] - \sum_j\Gamma_{\text{ph},j}^{\text{cd}}S, \quad (1)$$

where $S = (\sigma_j^+\rho\sigma_j^+ + \sigma_j^-\rho\sigma_j^-)$. This model is applicable to two excitons coupled to the same phonon bath, but the generalization to model two QDs or two excitons coupled to different phonon baths is straightforward. The phonon-induced scattering rates are derived as follows [13,14]: $\Gamma_{\text{ph}}^{\sigma_j\pm} = (\Omega_j'^2/2)\text{Re}[\int_0^\infty d\tau e^{\pm i\Delta_j\tau}(e^{\phi(\tau)} - 1)]$ and $\Gamma_{\text{ph},j}^{\text{cd}} = (\Omega_j'^2/2)\text{Re}[\int_0^\infty d\tau\cos(\Delta_j\tau)(1 - e^{-\phi(\tau)})]$, where $\phi(\tau) = \int_0^\infty d\omega(J(\omega)/\omega^2)[\coth(\beta\hbar\omega/2)\cos(\omega\tau) - i\sin(\omega\tau)]$. For the one exciton limit, the above model has been used to describe the spectra for driven In(Ga)As QDs [14], where phonon-mediated processes are found to play a significant role. Consistent with these experiments, we will use $\omega_b = 1$ meV and $\alpha_p = 0.15/(2\pi)^2$ ps^2.

The emission from the QD is experimentally detected through a planar cavity (which spatially separates the pump field from the emitted spectrum), and the incoherent spectrum of the system is given by [14] $S(\mathbf{r},\omega) \equiv F(\mathbf{r})S(\omega)$. Here $F(\mathbf{r})$ is a geometrical factor and $S(\omega)$ can be calculated by the quantum regression theorem.

From our two-exciton polaron master equation, the analytical spectrum is calculated to be

$$S(\omega) = \sum_j\alpha_j\left\{\text{Re}\left[\frac{ih_j(0)C_j(\omega)D_j(\omega) - f_j(0)D_j(\omega)}{(D_j(\omega) + i2\Delta_j)D_j(\omega) - E_j(\omega)^2}\right] - \text{Re}\left[\frac{E_j(\omega)[g_j(0) + ih_j(0)C_j(\omega)]}{(D_j(\omega) + i2\Delta_j)D_j(\omega) - E_j(\omega)^2}\right]\right\}, \quad (2)$$

where $C_j(\omega) = \Omega'_j/[2(i\delta\omega - \gamma_{\text{pop}}^j)]$, $D_j(\omega) = i\delta\omega - \gamma_{\text{pol}}^j - i\Delta_j + \Omega_j'^2/[2(i\delta\omega - \gamma_{\text{pop}}^j)]$, and $E_j(\omega) = \Gamma_{\text{ph},j}^{\text{cd}} + \Omega'_1C_j(\omega)$. The steady-state functions are [14] $f_j(0) = \frac{1}{2}[1 + \langle\sigma_j^z\rangle_{\text{ss}} - 2\langle\sigma_j^-\rangle_{\text{ss}}\langle\sigma_j^+\rangle_{\text{ss}}]$, $g_j(0) = -\langle\sigma_j^+\rangle_{\text{ss}}^2$, and $h_j(0) = -\langle\sigma_j^+\rangle_{\text{ss}}[1 + \langle\sigma_j^z\rangle_{\text{ss}}]$, where the steady-state inversion ($\langle\sigma_j^z\rangle_{\text{ss}}$) and polarization ($\langle\sigma_j^+\rangle_{\text{ss}}$) are given in Ref. [14]. The parameters α_j are scaling terms that adjust the strength of the exciton emission through cavity decay; these are proportional to the exciton dipole moments and they may differ depending upon the amount of asymmetry in the QD.

Due to Coulomb interactions and orientational inhomogeneities of the QDs, there are usually two neutral excitons polarized along the x and y axes, respectively [c.f. Fig. 1]. Thus for various pump polarization angles, θ, there will be different Rabi frequencies, $\Omega'_1 = \Omega_1\cos\theta$ and $\Omega'_2 = \Omega_2\sin\theta$, for the two excitons. Here $\Omega_j = \langle B\rangle\,\Omega_j^0$ is the maximum Rabi frequency. For clarity, we will first assume that $\Omega_{1,2} = \langle B\rangle\,\Omega^0 \equiv \Omega'$, though in general these will differ. Figure 2(a) shows an example calculation of the incoherent spectrum for various θ, using $\Delta_1 = -\Delta_2 = -6$ μeV. We assume a phonon bath temperature of $T = 6$ K, and use parameters for In(Ga)As QDs [21]. The main parameters are given in the figure caption. The red dashed lines shown in the same figure are the results obtained without phonon scattering (apart from ZPL decay), which show that phonon scattering plays a qualitatively important role; in particular, we see phonon-mediated spectral broadening and a coherent reduction of the Rabi frequency. For $\theta = 0$, only one exciton is excited, and the spectrum shows the expected Mollow triplet; note that the spectrum is asymmetric because of the off-resonant pump field and the coupling to phonons; only in the limit of no pure dephasing and the neglect of phonon scattering, a symmetric Mollow triplet is obtained [14]. As θ increases, then the other orthogonal exciton is gradually excited, as can be clearly seen at $\theta = \pi/8$—where the Mollow triplet evolves into a *Mollow quintuplet*. At $\theta = \pi/4$, the quintuplet merges into a triplet again if the two excitons share symmetric parameters. Figure 2(b) shows the spectrum at $\theta = \pi/8$ for various Rabi fields ($\Omega' = 20, 30, 40$ μeV from bottom to top), which show how the spectral quintuplet evolves with pump strength. From the analytical spectrum result, we conclude that there will be six peaks for the incoherent spectrum in general. However, both of the central peaks lie at the laser pump frequency and merge one peak, so there are actually five resolvable peaks.

Next we turn our attention to experiments. The planar cavity sample under investigation is grown by

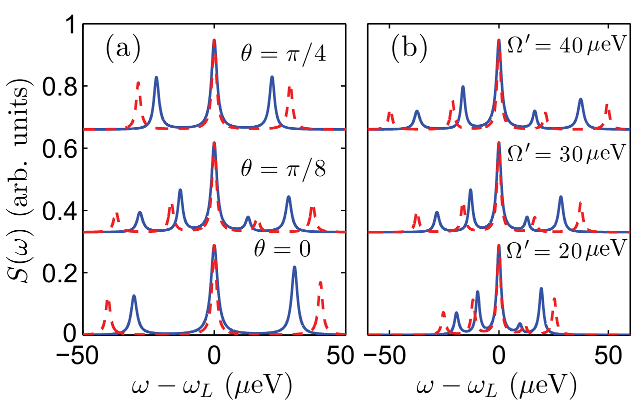

Fig. 2. Incoherent spectrum $S(\omega)$ with (blue, solid) and without (red, dashed) phonon scattering. The Rabi frequencies are $\Omega'_1 = \Omega'\cos\theta$ and $\Omega'_2 = \Omega'\sin\theta$, and $\gamma_{1,2} = \gamma'_{1,2} = 1$ μeV, $\delta_{12} = -12$ μeV, and $\alpha_{1,2} = 1$. (a) $\Omega' = 30$ μeV, with different θ. (b) $\theta = \pi/8$, with different Ω'.

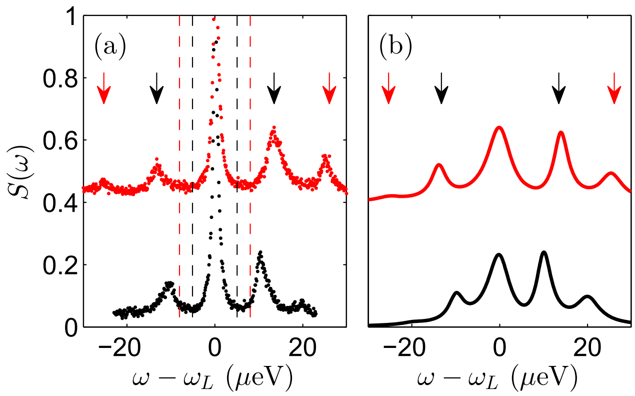

Fig. 3. Experimental data for a driven In(Ga)As QD and corresponding theoretical simulations, both at a phonon bath temperature of 6 K. (a) Experimental results with pump powers 50 μW (dark, lower) and 100 μW (red, upper). (b) Theoretical fits, with the following parameters: $\gamma_1 = 0.55$ μeV, $\gamma_2 = 0.85$ μeV, $\gamma_1' = 5.93$ μeV, $\gamma_2' = 7.13$ μeV, $\delta_{12} = 10$ μeV, $\alpha_1 = 1$ and $\alpha_2 = 0.4$. The Rabi frequencies are $\Omega_1' = 13.66/\sqrt{2}$ μeV, $\Omega_2' = 21.96/\sqrt{2}$ μeV (dark, lower), and $\Omega_1' = 13.66$ μeV, $\Omega_2' = 21.96$ μeV (red, upper).

metal-organic vapor epitaxy. Self-assembled In(Ga)As QDs are embedded in a GaAs λ-cavity, sandwiched between 29 (4) periods of $\lambda/4$-thick AlAs/GaAs layers as the bottom (top) distributed Bragg reflectors. For the investigations, the sample is kept in a Helium flow cryostat providing high temperature stability $T = 6 \pm 0.5$ K. Laser stray-light suppression is achieved by use of an orthogonal geometry between QD excitation and detection in combination with polarization suppression and spatial filtering via a pinhole. Resonant QD excitation is achieved by a narrow-band (≈500 kHz) continuous wave Ti:Sapphire ring laser. For high-resolution spectroscopy of microphotoluminescence we employ a scanning Fabry–Pérot interferometer with $\Delta E_{\text{res}} < 1$ μeV as described earlier [14,21].

Figures 3(a) and 3(b) show the experimental results and theoretical simulations, respectively. Due to the fact the experimental central peak includes both coherent and incoherent field, we focus on reproducing the experimental data around the Mollow sidebands. Figure 3(a) shows the experimental data under pump strength 50 μW (dark, lower) and 100 μW (red, upper). The dark (inner) arrows and red (outer) arrows show the inner sideband and outer sideband of the quintuplet, respectively. Figure 3(b) shows the theoretical simulations with fitting parameters given in the caption, which are consistent with earlier experiments [14,21]. We obtain a very good qualitative agreement between experiments and theory. Moreover, we have observed these spectral quintuplets from many of our In(Ga)As QDs.

In summary, we have introduced a theory to describe resonance fluorescence of coherently excited QD neutral excitons and predict the emergence of a spectral quintuplet. The theory uses a polaron master equation from which an analytical spectrum is derived and presented. Using data for In(Ga)As QDs, we obtain excellent agreement between theory and experiment.

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