Cavity-assisted emission of polarization-entangled photons from biexcitons in quantum dots with fine-structure splitting

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Abstract: We study the quantum properties and statistics of photons emitted by a quantum-dot biexciton inside a cavity. In the biexciton-exciton cascade, fine-structure splitting between exciton levels degrades polarization-entanglement for the emitted pair of photons. However, here we show that the polarization-entanglement can be preserved in such a system through simultaneous emission of two degenerate photons into cavity modes tuned to half the biexciton energy. Based on detailed theoretical calculations for realistic quantum-dot and cavity parameters, we quantify the degree of achievable entanglement.

OCIS codes: (250.5590) Quantum-well, -wire and -dot devices; (270.0270) Quantum optics; (270.5565) Quantum communications.

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1. Introduction

One of the key aspects on our way into the age of quantum-information is the identification of efficient sources of entangled photons [1]. Prototypical in this area are parametric sources based on nonlinear media such as lithium niobate [2]. Of central importance for technological applications, however, is the deterministic emission with high brightness and the possibility of electrical pumping - this is where semiconductors come into play. Recently, entangled-photon sources through direct two-photon emission across the bandgap have been demonstrated [3], and semiconductor quantum dots (QDs) have been utilized as deterministic quantum emitters for single photons [4, 5] and for lasing at the single-photon level [6, 7]. Looking at the emission from QD biexcitons, it is only natural to also consider QDs as deterministic single quantum emitters of entangled photon pairs [8, 9]. However, exciton fine-structure splitting has been shown to destroy polarization-entanglement [10–13]. Great effort has been undertaken to reduce this exciton fine-structure splitting by improving structural properties [14] and growth conditions [15], by exploration of QDs in alternative materials [16], or by applying external electric [17], magnetic [18], or strain fields [19]. In this Letter we explore an alternative route. We demonstrate that for a QD inside a high-quality cavity, a direct two-photon emission process

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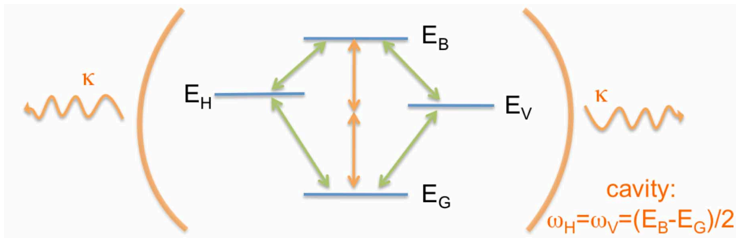

Fig. 1. Sketch of the quantum-dot cavity system. The optical transitions in a single QD are coupled to two orthogonal modes of a high-quality optical cavity with frequencies ω_i. Electronic QD configurations considered are ground-state with energy E_G, the two lowest exciton levels with possible fine-structure splitting with energies E_H and E_V, and the biexciton with energy E_B. The cavity modes are tuned close to half the biexciton energy, $\hbar\omega_V = \hbar\omega_H \approx (E_B - E_G)/2$. The effects of pure dephasing of electronic coherences and the finite lifetime of photons inside the cavity are taken into account.

from the biexciton can be used to render the polarization-entanglement of emitted photons virtually independent of fine-structure splitting. This would allow presently available high-quality QD structures to be used as single deterministic sources of polarization-entangled photon pairs.

2. Theory & methods

We investigate the generation of polarization-entangled photon pairs by two-photon emission from QD biexcitons. To this end, we study a single QD coupled to a high-quality optical cavity as illustrated in Fig. 1 (details of the theoretical model are given below). Electronic QD configurations considered are ground-state, the two lowest excitons with fine-structure splitting, and the biexciton. Two degenerate cavity modes are assumed to be tuned close to half the biexciton energy. A pure dephasing of electronic coherences and a finite lifetime of photons inside the cavity are taken into account. We initialize the system in the biexciton configuration with no photons inside the cavity and follow the biexciton decay until two photons have been emitted from the cavity and the system has fully returned to its ground-state. Generally, the biexciton can decay through different competing channels: (i) the biexciton-exciton cascade by subsequently emitting two photons with H- or V-polarization, or (ii) by simultaneous emission of two photons in a direct two-photon emission process. In the biexciton-exciton cascade decay, an exciton fine-structure splitting big enough to be spectrally resolved, reveals the "which-path" information and polarization-entanglement of the emitted photon pair is lost [10, 11]. The two-photon emission from the QD biexciton to the ground state is a resonant higher-order process, via an intermediate virtual state for which energy matching is not required. When the cavity modes are tuned to half the biexciton energy, the direct two-photon emission can be enhanced by being resonant, as opposed to the cascaded emission which is then detuned. For sufficiently high cavity quality, the Purcell-enhancement of the resonant transition increases the preference of the two-photon emission [20–22], such that it is the dominant process to occur in the light emission from the QD biexciton (preferred over the cascade-decay, which for finite biexciton binding energy is far off-resonant from the cavity modes, and thus suppressed).

The Hamiltonian of the QD biexciton-exciton system interacting with the quantized cavity field is given by

$$\begin{aligned}\mathscr{H} =& E_G|G\rangle\langle G| + E_B|B\rangle\langle B| + E_H|X_H\rangle\langle X_H| + E_V|X_V\rangle\langle X_V| \\ &+ \sum_{i=H,V}\left(\hbar\omega_i b_i^\dagger b_i - \left[g\left(|G\rangle\langle X_i|b_i^\dagger + |X_i\rangle\langle B|b_i^\dagger\right) + \text{h.c.}\right]\right). \qquad (1)\end{aligned}$$

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The first line describes the electronic system with the free energies of the electronic ground state, E_G, of the excitons, E_H, E_V, respectively, and of the biexciton, E_B, in the respective electronic configurations $|G\rangle$, $|X_H\rangle$, $|X_V\rangle$ and $|B\rangle$. The second line represents the free part of the photon field in the two orthogonal cavity modes at frequencies ω_i with photon creation and annihilation operators, $b_i^\dagger$ and b_i, respectively. The excitation and de-excitation of the electronic system through photon absorption or emission takes place with coupling strength g. The coupled biexciton-exciton-photon dynamics obey the following equation of motion for the system density operator ρ_s in Lindblad [23] form:

$$\frac{\partial}{\partial t}\rho_s = -\frac{i}{\hbar}[\mathscr{H},\rho_s] + \mathscr{L}_{\text{cavity}}(\rho_s) + \mathscr{L}_{\text{pure}}(\rho_s)\,. \tag{2}$$

In addition to the system part explicitly described by the Hamiltonian, Eq. (1), we have included a finite lifetime $\hbar/\kappa$ of the photons inside the cavity through $\mathscr{L}_{\text{cavity}}(\rho_s) = \frac{\kappa}{2}\sum_{i=H,V}(2b_i\rho_s b_i^\dagger - b_i^\dagger b_i\rho_s - \rho_s b_i^\dagger b_i)$, and a phenomenological pure dephasing $\mathscr{L}_{\text{pure}}(\rho_s) = -\frac{1}{2}\sum_{\chi,\chi',\chi\neq\chi'}\gamma^{\text{pure}}_{\chi\chi'}|\chi\rangle\langle\chi|\rho_s|\chi'\rangle\langle\chi'|$ of coherences between electronic configurations, with $\chi,\chi' \in \{G, X_H, X_V, B\}$ [11]. For the results shown below we used the same value $\gamma^{\text{pure}}_{\chi\chi'} = \gamma = \hbar/200\,\text{ps}^{-1} \approx 3\,\mu\text{eV}$ for all the electronic coherences, which is a realistic value for pure dephasing of the excitonic coherences at low temperature [24]. We have checked, however, that a slightly different choice of the pure dephasing model following Ref. [25] does not qualitatively change our results. We calculate the system dynamics by directly solving the von-Neumann equation, Eq. (2), in time for the initial condition that the electronic system is in the biexciton configuration and the cavity modes are empty. We note that all the different dynamically competing emission and absorption processes are fully included in this non-perturbative approach.

To analyze the dynamics of the emission of the two photons from the QD biexciton inside the cavity and determine the quantum properties and statistics of the emitted light, we calculate the second-order photon correlation function

$$\begin{aligned} G^{(2)}_{ij,kl}(t,\tau) =& \langle b_i^\dagger(t) b_j^\dagger(t+\tau) b_k(t+\tau) b_l(t)\rangle \\ =& \,\text{tr}\{\rho_s b_i^\dagger(t) b_j^\dagger(t+\tau) b_k(t+\tau) b_l(t)\}\,, \end{aligned} \tag{3}$$

for photons in the two orthogonal and degenerate cavity modes H and V. Using the quantum-regression theorem [26], the full two-time dependence of $G^{(2)}_{ij,kl}(t,\tau)$ is evaluated. The only non-vanishing elements are the diagonal elements $G^{(2)}_{ii,ii}(t,\tau)$ and the off-diagonal elements $G^{(2)}_{ii,jj}(t,\tau)$ with $i \neq j$ [11]. The diagonal elements contain information about the statistics of the emitted light [6], whereas the off-diagonal elements, characterize the polarization entanglement of emitted photons [27]. By taking the double time-average of $G^{(2)}_{ii,jj}(t,\tau)$ we obtain the two-photon density matrix $\rho_{i,j}$, with $i, j = H, V$, which is needed for the quantum-state tomography of the emitted photon pair (ρ is normalized such that $\text{tr}\{\rho\} = 1$). All calculations in this work are performed for typical parameters of present high-quality QD-cavity systems. For the biexciton binding energy we use $E_B^{XX} = 1\,\text{meV}$, the coupling strength of electronic transitions to the photon modes is $g = \hbar/10\,\text{ps}^{-1} \approx 66\,\mu\text{eV}$. The cavity modes are tuned to half the biexciton energy $\hbar\omega_i \approx (E_B - E_G)/2$ and a finite fine-structure splitting δ (varied from -40 to $+40\,\mu\text{eV}$) between exciton levels is included. We note that even for the highest cavity quality studied, no pronounced Rabi-flopping occurs at the ground-state to exciton and exciton to biexciton transitions, as those are far off-resonant from the cavity modes.

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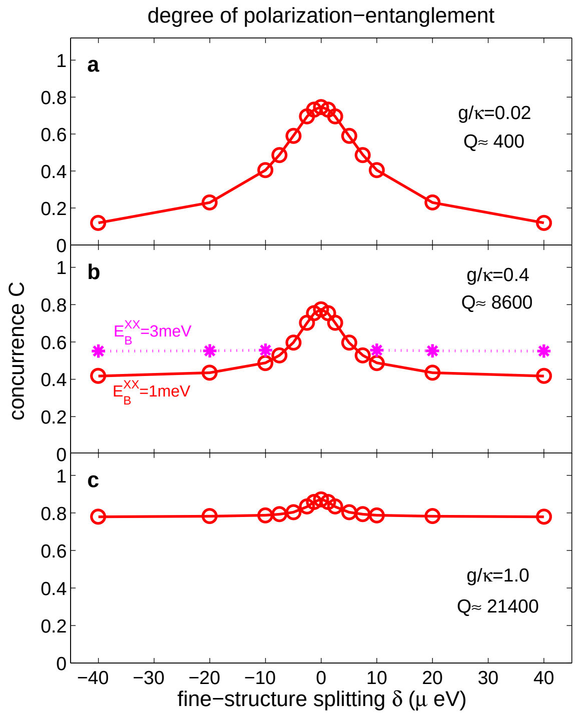

Fig. 2. Dependence of polarization-entanglement on the fine-structure splitting δ for different-quality cavities. The concurrence C is shown (○), which in the system studied is given by $C = 2|\rho_{H,V}|$. Cavity modes are tuned to half the biexciton energy, $\hbar\omega_i = (E_B - E_G)/2$. Results shown are for (a) $\kappa/\hbar = 5\,\mathrm{ps}^{-1}$, (b) $\kappa/\hbar = 0.25\,\mathrm{ps}^{-1}$, and (c) $\kappa/\hbar = 0.1\,\mathrm{ps}^{-1}$ using $E_B^{XX} = 1\,\mathrm{meV}$. In panel (b), the concurrence is also shown for increased biexciton binding energy $E_B^{XX} = 3\,\mathrm{meV}$ ($*$). Clearly visible is that the higher the cavity quality and the larger the biexciton binding energy, the less sensitive the concurrence (and thus the polarization entanglement) is to the fine-structure splitting.

3. Results & discussion

In Fig. 2 we analyze the degree of polarization-entanglement of the emitted photons. As a direct measure of entanglement in the HV-bipartite system, we use the concurrence $C = 2|\rho_{H,V}|$ (defined as in Ref. [11] with $G^{(2)}_{HV,HV}(t,\tau) = 0$ due to the absence of pumping). Results are shown for cavities of different quality by changing the value g/κ (κ is the loss rate of photons from the cavity). Cavity quality-factors Q are given in each panel for wavelength $\lambda = 880\,\mathrm{nm}$, corresponding to InGaAs systems. Note that the cavity is tuned to the two-photon resonance of the biexciton transition unless otherwise noted and, hence, it is detuned from the single-photon biexciton and exciton transitions.

Using this setup with a low-quality cavity, both biexciton-to-exciton and exciton-to-ground-state emission can efficiently couple into the spectrally broad cavity mode and these two cascaded first-order processes are stronger than the direct two-photon emission from the biexciton, which is a second-order process. In this case of the low-quality cavity, the results in Fig. 2(a) clearly reveal that – just as in previous studies – the concurrence quickly decays with increasing fine-structure splitting. Here, the decay of the biexciton is mostly through the biexciton-exciton cascade, as the higher-order direct two-photon emission process is not sufficiently favored. The maximum value of $C = 1$ is not reached for zero fine-structure splitting as our model takes into

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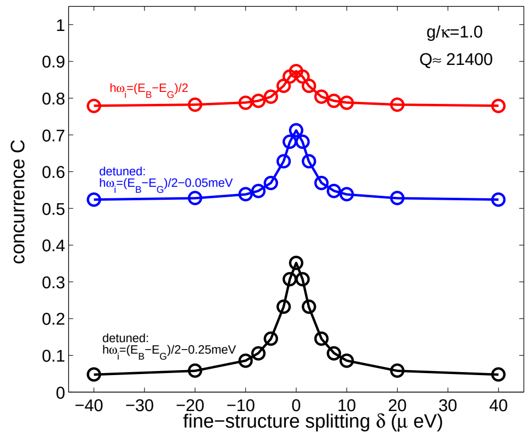

Fig. 3. Sensitivity to detuning from two-photon resonance condition. The concurrence C is shown for different detuning of the cavity modes from the two-photon resonance condition for the high-quality cavity. Red: $\hbar\omega_i = (E_B - E_G)/2$ [same as in Fig. 2(c)]. Blue: $\hbar\omega_i = (E_B - E_G)/2 - 0.05\,\text{meV}$. Black: $\hbar\omega_i = (E_B - E_G)/2 - 0.25\,\text{meV}$.

account a finite lifetime (cross-dephasing [11]) of coherences between the different excitonic configurations $|X_H\rangle$ and $|X_V\rangle$.

For a higher cavity quality, shown in Fig. 2(b), the dependence of the concurrence on the fine-structure splitting is less prominent. In this case the narrower cavity line reduces the coupling to the detuned biexciton-exciton-cascade and favors the direct higher-order two-photon transition through Purcell enhancement. While the different competing processes discussed above are fully included in our (non-perturbative) treatment of the system dynamics, here their relative importance can be deduced from the dependence of the concurrence on the fine-structure splitting.

Finally, in Fig. 2(c) we consider a high-quality cavity with a Q-factor typical for present technology. Our results reveal dominant two-photon emission leading to a concurrence that is virtually independent of the fine-structure splitting and takes an almost constant value of about 80%. This main result of our paper demonstrates that polarization-entanglement can be preserved in systems with finite (and even large) fine-structure splitting via an alternative process through appropriate mode engineering in high-quality cavities.

In Fig. 2(b) we also show the result for a QD with larger biexciton binding energy of 3 meV (see the dotted line). In this case the single-photon biexciton and exciton transitions are further detuned from the cavity resonance and even for the medium-quality cavity the degradation of entanglement with increasing fine-structure splitting gets further reduced. This underscores the potential of systems with higher biexciton binding energy, such as the CdSe system studied in Ref. [28], to further optimize the presented new scheme.

The influence of a finite detuning between the cavity mode and the two-photon resonance is presented in Fig. 3 for the high-Q cavity case of Fig. 2(c). With increasing detuning, the two-photon transition becomes less efficient to the advantage of the cascaded decay, and the level of entanglement achievable for finite fine-structure splitting is reduced. As expected for a high-Q cavity, we observe that a precise tuning of the narrow cavity lines to the two-photon transition is required to achieve a high degree of polarization entanglement. This, however, is less challenging than the precise engineering of the exciton fine-structure splitting of previous approaches [17–19]. The required degeneracy of H- and V-polarized modes should be attainable with current micropillar samples, which offer a sufficient cavity quality to meet the

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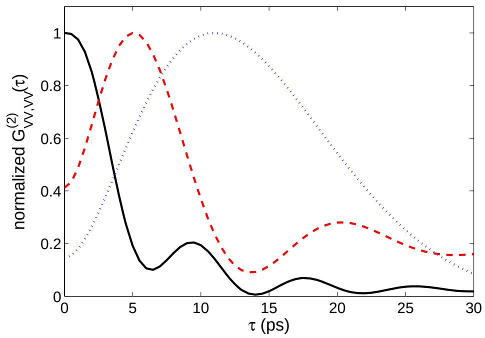

Fig. 4. Statistics of the emitted photons. Second-order photon correlation function $G^{(2)}_{VV,VV}(t,\tau)$ averaged over all emission times t. Results are for $g/\kappa = 0.4$ (medium-quality cavity) and $\delta = 0.0$; each set of data is normalized to its maximum. Strong photon "bunching" is visible for the cavity modes tuned to half the biexciton energy, $\hbar\omega_i = (E_B - E_G)/2$, (black, solid line). Clear "anti-bunching" is observed when the cavity modes are further detuned from the two-photon resonance condition for $\hbar\omega_i = (E_B - E_G)/2 - 0.25\,\text{meV}$ (red, dashed line) and $\hbar\omega_i = (E_B - E_G)/2 - 0.5\,\text{meV}$ (blue, dotted line), respectively.

requirements discussed in this paper. In Fig. 3, we also note that the peak in the concurrence is much narrower for the high-quality cavity than in the low-quality cavity in Fig. 2(a). For low cavity quality, the short lifetime of photons inside the cavity significantly broadens the resonances such that very small fine-structure splittings are not resolved. Thus, they do not give rise to the "which-path" information in the cascade decay. This partly preserves entanglement even at small non-zero splitting. Details of the degradation of the entanglement with increasing detuning from the two-photon resonance condition quantitatively depend on the QD dephasing. Generally, the magnitude of the dephasing rate is determined by the QD parameters, temperature and excitation conditions. Furthermore, two different dephasing models have been suggested in the literature [11,25], for which we have checked in our calculations that the specific choice does not qualitatively change our results. We would like to point out that these calculations have also shown that the main result of our paper, namely the high degree of polarization entanglement for photons emitted through the direct two-photon process, is actually rather insensitive to the specific choice of the pure dephasing.

Figure 4 displays the statistics of photons emitted for intermediate cavity quality, $g/\kappa = 0.4$, for different detuning of the cavity resonance from half the biexciton energy. For $\hbar\omega_i = (E_B - E_G)/2$ a strongly increased probability for simultaneous emission of both photons at $\tau \approx 0$ is clearly visible ("bunching"), whereas for $\hbar\omega_i = (E_B - E_G)/2 - 0.25\,\text{meV}$ and $\hbar\omega_i = (E_B - E_G)/2 - 0.5\,\text{meV}$ it is more likely to have the two photons emitted with a certain time-delay. This is evidence for step-wise decay through the biexciton-exciton cascade. Spectral features of the emitted photons under similar conditions are discussed in detail in Ref. [20].

4. Conclusions

For a realistic high-quality QD cavity system, we have theoretically investigated the generation of polarization-entangled photon pairs by direct two-photon emission from QD biexcitons. Our results demonstrate for two cavity modes with orthogonal polarization tuned to half the biexciton energy, that the simultaneous emission of two identical photons (direct two-photon process) can be dominant over the biexciton-exciton cascaded decay. Calculating the second-order photon correlation function, we show that in this case the polarization-entanglement between emitted photons can be made largely insensitive to exciton fine-structure splitting. This alter-

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native scheme has the potential of a new route in using presently available high-quality QDs as single deterministic quantum emitters for polarization-entangled photon pairs without the need to minimize the exciton fine-structure splitting.

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