Nonclassical correlation of polarization-entangled photons in a biexciton–exciton cascade

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Abstract

We develop a theoretical model to study the intensity–intensity correlation of polarization-entangled photons emitted in a biexciton–exciton cascade. We calculate the degree of correlation and show how polarization correlations are affected by the presence of dephasing and energy-level splitting of the excitonic states. Our theoretical calculations are in agreement with the recent observation of polarization-dependent intensity–intensity correlations from a single semiconductor quantum dot (Stevenson et al 2006 Nature 439 179). Our model can be extended to study polarization-entangled photon emission in coupled quantum dot systems.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Polarization correlations of photons emitted in cascade emission are a well-known phenomenon and numerous theoretical and experimental studies exist in the literature on this subject since the early days of quantum optics [1–6]. Some of the earlier studies were motivated in testing generalized Bell’s inequalities [2], the existence of hidden variables and whether quantum mechanics was a non-local theory or not [3, 4], following the question raised by Einstein, Podolsky and Rosen [7]. In recent times, polarization-correlated photon pairs have become important in the field of quantum information science due to their entangled nature. Moreover many applications of quantum information, such as quantum key distribution [8], efficient optical quantum computing [9], long-distance quantum communication using quantum repeaters [10] and implementation of quantum telecommunication schemes [11] require single-photon pairs per cycle. This requirement of entangled photon pairs per cycle of excitation could be satisfied by cascade emission from a single-atom or atom-like systems such as semiconductor quantum dots, provided one gets over the inherent asymmetries. Recently, such cascade emission has been reported for semiconductor quantum dots [12–15]. It was further seen that polarization entanglement of the emitted photon pairs was degraded by the presence of energy-level splitting of the intermediate excitonic states and any incoherent process that leads to a population transfer between the two intermediate excitonic states [14–16]. Moreover, dephasing arising due to interaction of the quantum dot with its solid-state environment can also degrade the entanglement [17]. Some recent studies have also shown how the fidelity of entanglement depends on excitonic-level splitting [18] and the dynamics of the incoherent dephasing [19]. Different methods have been proposed to reduce and control the incoherent dephasing and energy-level splitting of the excitonic states thereby preserving the entanglement in the system [14–16, 18, 20–22]. Further methods to enhance the generated entanglement by coupling the quantum dot to a micro-cavity have also been proposed [23, 24]. As quantum dot systems are of great importance for future applications in quantum information science, a clear yet simple model for understanding the effects of all these different decoherence mechanisms on the dynamics of the system is required. Thus we develop, in this paper, a simple theoretical model to analytically study the influence of different decoherence mechanisms and the intermediate state splitting on the generation of polarization-entangled photon pairs in cascade emission.
Figure 1. Schematic diagram of a four-level cascade system. Here H and V refer to horizontally and vertically polarized photon emission. $\Delta$ is the energy-level separation of the intermediate states and $\gamma$’s are the spontaneous emission rates given by $\gamma = 2\alpha_0|\langle \alpha_0 | l \rangle|^2/3\hbar c^3$. The incoherent dephasing rates of the intermediate states are given by $2\gamma_{\alpha\beta}$ and $2\gamma_{\beta\alpha}$ respectively.

2. Model

We consider a four-level system undergoing cascade emission as our model. We show a schematic diagram of such a cascade in figure 1. The excited state |i⟩ and the intermediate states |α⟩, |β⟩ would correspond to the biexcitonic and optically active excitonic states respectively in a quantum dot. Further, |j⟩ is taken to be the ground state. Here $2\gamma = 2(\gamma_1 + \gamma_2)$ is the total spontaneous emission rate of the state |i⟩. $2\gamma_1$ and $2\gamma_2$ are the spontaneous emission rates of the states |α⟩ and |β⟩ respectively and $2\gamma_{\alpha\beta}(2\gamma_{\beta\alpha})$ is the incoherent dephasing rate of the state |α⟩(|β⟩). The energy-level splitting of the intermediate state is given by $\Delta$. In this type of four-level cascade scheme there are two decay paths for the excited state, |i⟩ → |α⟩ → |j⟩ and |i⟩ → |β⟩ → |j⟩. The generation of entanglement in these schemes is attributed to the fact that these decay paths can become indistinguishable. The eigenbasis of this system is formed by the four states \{|i⟩, |α⟩, |β⟩, |j⟩\}. In this basis, the radiative transition from the excited state generates collinearly polarized photons with linear polarizations along two orthogonal directions denoted by H (horizontal) and V (vertical). When the states |α⟩ and |β⟩ are degenerate, the decay paths become indistinguishable and we get a maximally entangled two-photon state [7, 12]

$$|E⟩ = \frac{1}{\sqrt{2}}(|H⟩_{i} |V⟩_{j}) + (|V⟩_{i} |H⟩_{j}).$$

In practical systems such as atoms and quantum dots these levels are usually non-degenerate and hence the entanglement of the emitted photon pairs depends completely on the degree of degeneracy and dynamics of these intermediate states. In our model, we have taken them to be non-degenerate and study the effect of such intermediate-level splitting on the correlation of the emitted photon pairs. To understand the effect of incoherent dephasing and energy-level splitting of the excitonic state on the dynamics of emitted photon pairs from the cascade, we need to study the two-time second-order correlations. This is given by

$$\langle I(\theta_1, t + \tau)I(\theta_1, t) \rangle = \frac{e^{i\phi_1}}{c} 8 \frac{1}{2r^2} D_1^\dagger D_2^\dagger \sum \langle i | i \rangle \langle i | i \rangle$$

$$\times \{ e^{-2i\gamma_\tau} + \cos 2\theta_1 \cos 2\theta_2AT \} - 2e^{-2i\gamma_\tau/2\gamma_\alpha\beta} (\gamma_\alpha\beta \Delta)^2 \cos \Delta \tau \}.$$
where \( D_1 = |\vec{d}_{\alpha i}| = |\vec{d}_{\beta j}| \) and \( D_2 = |\vec{d}_{\alpha i}| = |\vec{d}_{\beta j}| \). The above simple form of the second-order correlation has been derived to match our theoretical analysis to that of the experimental results [15]. For details of the mathematical analysis leading to the generalized form of the two-time intensity-intensity correlation the reader is referred to section 4 of this paper. One can clearly see from equation (4) that the second-order correlation is profoundly influenced by both the incoherent dephasing rates as well as the energy-level splitting of the intermediate states. Note further that in the presence of small \( \Delta \) this becomes equivalent to the second-order correlations measured in [3, 4]. Next, we define a quantity the degree of correlation \( c_{\mu} \) as

\[
c_{\mu} = \frac{\langle I_{\mu} I_{\mu} \rangle - \langle I_{\mu} I_{\mu'} \rangle}{\langle I_{\mu} I_{\mu} \rangle + \langle I_{\mu} I_{\mu'} \rangle},
\]

(5)

where \( \mu, \mu' \) stands for mutually orthogonal polarization basis. The degree of correlation varies between +1 and -1, where +1 represents perfect correlation (−1 for anti-correlation) and 0 represents no polarization correlation.

### 3. Results and discussion

#### 3.1. Effect of excitonic-level splitting on the correlation

In figure 3(a), we show how the time-averaged degree of correlation varies with the basis angle for different values of splitting \( \Delta \) of the excitonic levels. Note that here the excitonic-level dephasing \( \gamma_0 \) has been taken to be zero. We see that the degree of correlation is independent of the polarization basis when \( \Delta = 0 \) and takes a value \( c_\mu = 1 \). This corresponds to a perfect polarization correlation among the emitted photons. From the expression of \( c_\mu \), it is clear that this can happen only when the cross-polarized correlations vanish and the emitted photons are perfectly co-polarized. One can even see this explicitly from equation (4) by putting the values of \( \theta_1, \theta_2 = \theta_1 + \pi/2 \) for H–V, D–D’ and V–H basis, where H, V, D and D’ stands for horizontal, vertical, diagonal and orthodiagonal polarization basis, respectively. Further, as the cross-polarized correlations are absent the pair of photons emitted in one excitation cycle can take either of the two paths \( |i\rangle \rightarrow |\alpha\rangle \rightarrow |j\rangle \) or \( |i\rangle \rightarrow |\beta\rangle \rightarrow |j\rangle \) thus making these paths indistinguishable. As a consequence, we do not get the ‘Welcher Weg’ or which path information thereby making the final state of the emitted photon pair entangled in both the linear and diagonal polarization basis. The generated entangled states can hence be written as \( 1/\sqrt{2}(|HH\rangle + |VV\rangle) \) and \( 1/\sqrt{2}(|DD\rangle + |D'D'\rangle) \) for the rectilinear and diagonal basis respectively. Note further that in this case perfect anti-correlation \( (c_\mu = -1) \) is expected for measurement in the circularly polarized basis with the entangled state given by \( 1/\sqrt{2}(|RL\rangle + |LR\rangle) \). Thus one should get perfectly cross-polarized photons as the co-polarized correlations vanish in this basis. This is exactly what we get from the general expression of equation (4) (see section 4, equation (13)) and is shown by the solid curves in figure 3(b). Further in figure 3(a) we see that the degree of correlation is practically independent of the excitonic-level splitting \( \Delta \) in the rectilinear basis. As we change our polarization basis the effect of \( \Delta \) becomes significant. In the diagonal basis, for example, with the increase in level splitting the degree of correlation gradually decreases and eventually vanishes. In the presence of \( \Delta \), the cross-polarization does not vanish and we have path information for the emitted photons when we measure the second-order correlations, thus destroying any entanglement in the system. The behaviour of the correlations in the circular basis in the presence of large excitonic-level splitting is shown by the broken curves in figure 3(b). One can clearly see that there is no polarization correlation at all for large \( \Delta \). The sinusoidal behaviour of \( c_\mu \) for nonzero value of \( \Delta \) as seen in figure 3(a) is in agreement with the classical linear
polarization correlation behaviour. Note that our theoretical results are in agreement to experimentally observed data \[15\]. It should be noted that in our analysis we have concentrated on the calculation of the quantum correlation \(c_\mu\). This was also measured in the experiment of Stevenson et al. We have not examined measures of entanglement like concurrence. This is because if \(\Delta\) the intermediate state exciton splitting is nonzero then horizontal and vertical photons have different frequencies which amounts to saying that we have for nonzero \(\Delta\) quantum states which are characterized by two different parameters and measures of entanglement in such situations do not exist.

### 3.2. Effect of decoherence on the correlation

In figures 4(a) and (b), we show how the incoherent dephasing of the intermediate excitonic states affect the time-averaged degree of correlations \(c_\mu\) when excitonic states are non-degenerate \((\Delta \neq 0)\) and degenerate \((\Delta = 0)\) respectively. Note that here we have assumed that both the intermediate states have same dephasing rates. One can clearly see that the effect is different for different measurement basis. The degree of polarization correlation for example in the rectilinear basis decreases with increasing dephasing irrespective of whether the excitonic states are non-degenerate or degenerate. For large dephasing rates the emitted photon pairs become almost un-correlated in their polarization. This is attributed to the presence of significant cross-polarized correlation for large dephasing rates of the intermediate states. The incoherent dephasing of the intermediate levels causes a incoherent population transfer among the states \(|\alpha\rangle\) and \(|\beta\rangle\) thereby allowing the second photon to be emitted with orthogonal polarization to the first one. In the diagonal basis, on the other hand, the dephasing does not affect the correlation at all for large \(\Delta\) but significantly decreases the correlation when \(\Delta = 0\) for large dephasing rates. So we see that in diagonal basis even when the intermediate levels are degenerate we can still have significant cross-correlation if there is some incoherent relaxation process by which they can get coupled. This in turn spoils the quantum correlation in the system as can be seen clearly from figure 4(b).

In figure 5, we show how the correlations behave in the circular basis in the presence of a large dephasing rate \(\gamma_{\alpha\beta}/\gamma = 10\). The red curve corresponds to co-polarized \((\theta_1 = \theta_2 = \pi/4, \phi_1 = \phi_2 = -\pi/2)\) photons and the blue for cross-polarized \((\theta_1 = \theta_2 = \pi/4, \phi_1 = -\pi/2, \phi_2 = \pi/2)\) ones. The solid curve is for \(\Delta \sim \) large and broken one for \(\Delta = 0\). Here R and L stand for right and left circular polarization. The R–R correlation curves are time shifted for better comparison to the R–L correlation.
4. Detail derivation of the intensity–intensity correlation

Our model consists of a biexcitonic state and two excitonic states labelled as |i⟩ and |α⟩, |β⟩ respectively. The equilibrium state is given by technique [27] under the Born, Markov and rotating wave effects in the system are incorporated via a master equation on substituting equations (6) and (7) into (8) and solving for

Here \(\gamma\) is the total spontaneous emission rate of the biexcitonic state |i⟩. 2γ(1 + γA/A) + 4γ2γβα/A 1 - e^(-αA-2γ)A/(2(α0 - A)) + (A -> -A),

\[D(t) = \left(2\gamma_1 \left(1 + \frac{\Gamma_a}{A}\right) + 4\frac{\gamma_2\gamma_β\alpha}{A}\right) 1 - e^{-\left(\frac{\alpha_0-A}{2\gamma}\right)} + (A -> -A),\]

\[F(t) = \left(2\gamma_2 \left(1 - \frac{\Gamma_a}{A}\right) + 4\frac{\gamma_1\gamma_β\alpha}{A}\right) 1 - e^{-\left(\frac{\alpha_0-A}{2\gamma}\right)} + (A -> -A),\]

\[K(t) = \left(2\gamma_2 \left(1 - \frac{\Gamma_a}{A}\right) + 4\frac{\gamma_1\gamma_β\alpha}{A}\right) 1 - e^{-\left(\frac{\alpha_0-A}{2\gamma}\right)} + (A -> -A).\]

The effect of non-degeneracy of the excitonic states and their incoherent dephasing on the dynamical evolution of the system is given by [27]

\[\tilde{E}^{(\pm)}(\vec{r}, t) = \tilde{E}_0^{(\pm)}(\vec{r}, t) - \left(\frac{\alpha_0}{c}\right) \frac{1}{r} \left[\hat{\varepsilon} \cdot (\hat{\varepsilon} \cdot \vec{d}_{\alpha i})\right] |\alpha_i\rangle |i\rangle,\]

\[+ \left[\hat{\varepsilon} \cdot (\hat{\varepsilon} \cdot \vec{d}_{\beta i})\right] |\beta_i\rangle |i\rangle,\]

\[+ \left[\hat{\varepsilon} \cdot (\hat{\varepsilon} \cdot \vec{d}_{\beta j})\right] |\beta_j\rangle |j\rangle.\]

Finally, using equation (11) in (2) we get the general form of the two-time intensity–intensity correlation

\[\langle II \rangle = \left(\frac{\alpha_0}{c}\right)^2 \frac{1}{r^4} \left[\left(\hat{\varepsilon} \cdot \vec{d}_{\alpha i}\right)^* \cos \theta_i |i\rangle |\alpha_i\rangle + \left(\hat{\varepsilon} \cdot \vec{d}_{\beta i}\right)^* e^{i\phi_i} \sin \theta_i |i\rangle |\beta_i\rangle\right] \times\]

\[\times \left|\hat{\varepsilon} \cdot \vec{d}_{\alpha j}\right|^2 \cos^2 \theta_j |\alpha_j\rangle + \left|\hat{\varepsilon} \cdot \vec{d}_{\beta j}\right|^2 \sin^2 \theta_j |\beta_j\rangle |\beta_{r+j}\rangle + \left|\hat{\varepsilon} \cdot \vec{d}_{\alpha j}\right| e^{i\phi_j} \sin \theta_j \left|\hat{\varepsilon} \cdot \vec{d}_{\beta j}\right|^2 |\alpha_j\rangle |\beta_{r+j}\rangle\]

\[\times \left|\hat{\varepsilon} \cdot \vec{d}_{\alpha i}\right| \cos \theta_i |i\rangle |\alpha_i\rangle + \left(\hat{\varepsilon} \cdot \vec{d}_{\beta i}\right) e^{i\phi_i} \sin \theta_i |\beta_i\rangle |i\rangle.\]

The two-time correlation function that appears in equation (12) is evaluated by invoking the quantum regression theorem [28] and equation (9). Finally, we get
In the special case of time intensity–intensity correlations for arbitrary polarization, Equation (13) gives the most general form of the two-dots in the micro-cavity. As a future prospect it would be interesting to extend the results found in context to such cascade emissions in quantum theoretical calculation is in agreement with the experimental shown how these effects are important in determining whether the excitonic states can affect polarization entanglement of the emitted photon pairs are classically correlated or entangled under how the dephasing and energy-level splitting of the excitonic states can affect polarization entanglement of photons emitted in a biexciton–exciton cascade. We have also shown that these effects are important in determining whether the emitted photon pairs are classically correlated or entangled in different polarization basis. Further, we have shown that our theoretical calculation is in agreement with the experimental results found in context to such cascade emissions in quantum dots. As a future prospect it would be interesting to extend the method of the present paper to a system of coupled dots or a dot in the micro-cavity.

Equation (13) gives the most general form of the two-time intensity–intensity correlations for arbitrary polarization directions and for any system undergoing a cascade emission. In the special case of \(\phi_1 = \phi_2 = 0\), \(\gamma_{\beta} = \gamma_{\alpha}\) and \(\gamma_2 = \gamma_4\) this reduces to the simplified result (4) of section (2).

5. Conclusions

In conclusion, we have developed a simple theory to understand how the dephasing and energy-level splitting of the excitonic states can affect polarization entanglement of photons emitted in a biexciton–exciton cascade. We have also shown that these effects are important in determining whether the emitted photon pairs are classically correlated or entangled in different polarization basis. Further, we have shown that our theoretical calculation is in agreement with the experimental results found in context to such cascade emissions in quantum dots. As a future prospect it would be interesting to extend the method of the present paper to a system of coupled dots or a dot in the micro-cavity.

\[
\langle II \rangle = \left( \frac{\alpha_0}{c} \right)^8 \frac{1}{4\pi^4} D_1^2 D_2^2 \langle |i_i| \langle j_j | \left[ f_1(\tau) + w_1(\tau) + f_2(\tau) + w_2(\tau) \right.ight.
\]

\[
+ (\cos 2\theta_1 + \cos 2\theta_2)(f_1(\tau) - w_2(\tau))
\]

\[
+ (\cos 2\theta_1 - \cos 2\theta_2)(w_1(\tau) - f_2(\tau))
\]

\[
+ \cos 2\theta_1 \cos 2\theta_2(1) - (f_1(\tau) - w_2(\tau) - f_2(\tau) - w_1(\tau))
\]

\[
+ \sin 2\theta_1 \sin 2\theta_2 (e^{-i(\phi_1 + \phi_1)} u(\tau) + e^{i(\phi_1 + \phi_1)} u^*(\tau)) \right].
\]

Here \(D_1 = |d_{\alpha}| \) and \(D_2 = |d_{\beta}|\). The \(f\)'s, \(w\)'s and \(u\) are found from the solutions of the density matrix equations (9) and are given by

\[
f_1(\tau) = e^{-\alpha_0 \tau} \left( \cosh(\Delta \tau) + \frac{\gamma_{\alpha}}{A} \sinh(\Delta \tau) \right),
\]

\[
f_2(\tau) = 2 e^{-\alpha_0 \tau} \frac{\gamma_{\beta}}{A} \sinh(\Delta \tau),
\]

\[
w_1(\tau) = 2 e^{-\alpha_0 \tau} \frac{\gamma_{\alpha}}{A} \sinh(\Delta \tau),
\]

\[
w_2(\tau) = e^{-\alpha_0 \tau} \left( \cosh(\Delta \tau) - \frac{\gamma_{\alpha}}{A} \sinh(\Delta \tau) \right),
\]

\[
u(\tau) = e^{-(\alpha_0 - i\Delta \tau)}.
\]

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