Entangled photon pairs from the optical decay of biexcitons

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Abstract

We show that it is possible to transfer the exciton–exciton Coulomb correlation to photons, producing thus pairs of near-gap photons with a high degree of quantum entanglement. The photon pairs emerge from the spontaneous optical decay of biexcitons into two polaritons. The pair intensity-correlations, calculated in the low density limit for a CuCl slab, exhibit quantum features which can be observed by coincidence detection.

Keywords: A. Semiconductors; D. Optical properties; E. Nonlinear optics

Entangled particles have been produced in a variety of systems. Photon pairs produced by parametric down conversion [1], cascade two-photon emission [2], pairs of atoms entangled by the exchange of a photon in a high Q cavity [3], are some examples. Entanglement describes a composite quantum system that cannot be factored into a product of single-particle states and thereby has no classical counterpart. It is manifested by the potential to exhibit correlations that cannot be obtained with classical systems [4]. The theory of quantum information processing lies on entanglement. Teleportation of quantum states [5,6], quantum computation steps [7], quantum cryptography [8] are recent striking applications of entanglement.

In this paper, we show that nonlinear correlated excitations in semiconductors can produce entangled near-gap photon pairs. Owing to the possibility of engineering the valence and conduction electronic states of semiconductors opened by modern growth techniques, and owing to the possibility of controlling the exciton–photon interaction in semiconductor microcavities (MCs) [9], the generation of entangled photon pairs in semiconductor systems is expected to be promising towards the realization of integrated quantum-optical devices. In most experiments entanglement has been realized by having the two entangled particles emerging from a common source. In the process here described, the entangled photon pairs emerge from the optical decay of states with two electron–hole (eh) pairs, namely the biexcitons. The quantum process can be schematically described in two steps. First, two incident pump photons, propagating inside the crystal as excitonic polaritons, create a virtual state with two eh pairs. Then the decay of the virtually excited biexcitons can be stimulated by sending an additional light beam (four wave mixing (FWM)) or can be driven by the vacuum fluctuations of the light field (spontaneous hyper Raman scattering (HRS)). The spontaneous optical decay of biexcitons can produce two final polaritons (see Fig. 1) or a longitudinal exciton and a polariton. Energy and

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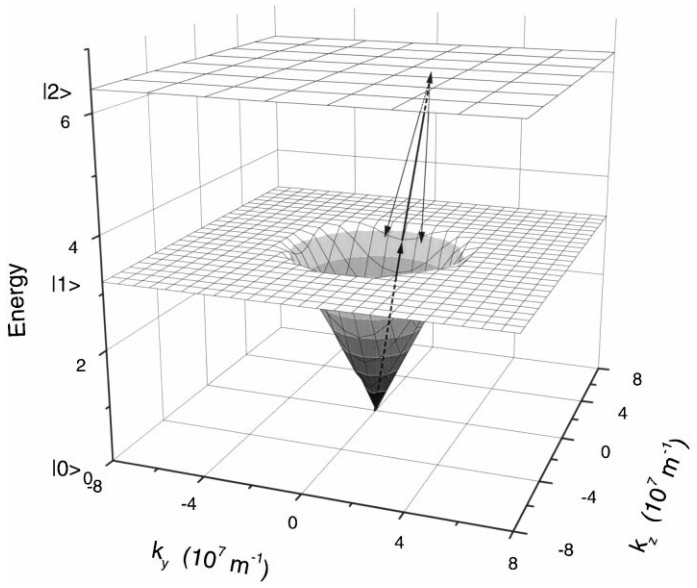

Fig. 1. Resonant Hyper Raman scattering process in CuCl involving the lower polariton branch. $|0\rangle, |1\rangle$ and $|2\rangle$ are, respectively, the ground state, the 1S exciton state and the bound biexciton state.

momentum are conserved in the whole process as indicated in Fig. 1. The detection of the emission lines from one final polariton for different scattering geometries and different energies of incident light has permitted the determination of the polariton dispersion curve in several large-gap bulk semiconductors [10,11]. If the two final quanta are polaritons with a significant photon component, they can escape the semiconductor and can be both detected. We propose coincidence detection of both final polaritons produced by the spontaneous optical decay of biexcitons and analyze their fourth order correlation properties, showing that, despite the presence of dephasing relaxation, they can maintain a high degree of nonclassical correlations. Quantum correlations are determined by the interplay of the exciton–exciton correlation [12], which correlates the polariton pairs and of the vacuum fluctuations of the light field [13] which determine the optical decay of biexcitons.

To theoretically model these effects, we compute the optical response in the $\chi^{(3)}$ limit including the electromagnetic field quantization [13,14]. We consider a two-band semiconductor coupled to a photon field by its electronic polarization [14]. In the absence of the light-matter interaction, the eigenstates of the Hamiltonian $H_c = H_0 + V_{\mathrm{Coul}}$ of the usual semiconductor model [15], can be labelled according to the number N of eh pairs. The $N = 1$ subspace is the exciton subspace. Excitons can be labelled by the additional quantum number n spanning bound and unbound exciton levels. These states can be obtained by applying the exciton creation operator $\hat{B}^{\dagger}_{n,\mathbf{k}} = \sum_{\mathbf{k}'} \Phi^{*}_{n,\mathbf{k}'} \hat{c}^{\dagger}_{\mathbf{k}'+\mathbf{k}/2} \hat{d}^{\dagger}_{-\mathbf{k}'+\mathbf{k}/2}$ to the semiconductor ground state, being $\hat{c}^{\dagger}$ and $\hat{d}^{\dagger}$ the e and h creation operators, while Φ_n is the exciton envelope function solution of the Wannier equation for the semiconductor. The interband electronic polarization of the given wave vector $\mathbf{k}$ can be expressed in terms of exciton operators $\hat{P}^{+}_{\mathbf{k}} = \sum_n M^{*}_n \hat{B}_{n,\mathbf{k}}$, where the matrix element $M_n = \mu \sum_{\mathbf{k}} \Phi_{n,\mathbf{k}}$ is the exciton dipole moment, being μ the interband dipole moment.

In order to analyze the generation and propagation of the scattered polaritons, we have derived the Heisenberg equation of motion for the polarization operator subjected to H_c and interacting with the quantized light field (in the usual dipole and rotating wave approximations). All calculations have been performed in the low-density limit, that includes in the nonlinear optical response those contributions related to the third order nonlinear polarization of the interacting electron system. By expressing the

nonlinear response in terms of the exact eigenstates of the system, it follows that the third order nonlinear polarization depends only on the one eh-pair subspace (excitons) and the two eh-pair subspace (biexcitons) [12,13,16].

As it is well known, excitons in semiconductors are an open quantum system interacting with phonons and other dephasing mechanisms. We include attenuation due to the various scattering mechanisms acting on the polarization waves in a phenomenological way, by introducing the coupling of excitons with a large number of reservoir oscillators at $T = 0$ K temperature. Following the usual Langevin approach, the resulting Heisenberg–Langevin equations for the electronic polarization include a damping term and a quantum-noise Langevin operator. This reservoir model describes only the relaxation of propagating fields but not the effect of thermalization of the eh pairs. We have not included the effect of thermalization of eh pairs since HRS is a below-gap process conserving energy and momentum related to the coherent interaction of waves inside the medium, where thermalized eh populations play a negligible role. Our calculations are performed in this coherent limit [16].

We consider a semiconductor slab of thickness L orthogonal to the z direction. The in-plane component $\mathbf{p} = (k_x, k_y, 0)$ of the wave vector $\mathbf{k}$ is a good quantum number for the whole system. The nonlinear process determining the emission of polariton pairs involves four waves, the two classical input beams (which we chose to be coincident and denoted by I) at energy ω_I and in-plane wave-vector $\mathbf{p}_I$, the scattered field 1 and the *probe* beam 2. The first step of the process is the virtual excitation of biexcitons driven by the pump field. We assume that the pump field propagates into the crystal as a classical coherent beam so that we can replace, in the equation of motion for the nonlinear polarization, the input electric-field operator $\hat{E}_I(z, \omega_I)$ by its corresponding complex-number function $E_I(z, \omega_I)$. By this approximation we assume that the field fluctuations in the input laser beam can be neglected with respect to the strong field expectation value. ¢From the Heisenberg–Langevin equations for the Fourier components of the interband electronic polarization $P_1^+(z, \omega_1) = \sum_{k_z} \mathrm{e}^{ik_z z} P^+_{(\mathbf{p}_1, k_z)}(\omega_1)$, we obtain the relevant nonlinear-polarization operator [13], acting as source term for the field at mode 1,

$$P_1^{nl}(z, \omega_1) = \chi^{(3)} \hat{E}_2^-(z, \omega_2) \mathrm{e}^{2ik_{Iz}z} E_I^2(\omega_I), \tag{1}$$

where E_I is the amplitude of the coherent input field incoming into the semiconductor slab and $k_{Iz} = k_{Iz}^{\mathrm{R}} + ik_{Iz}^{\mathrm{I}}$ is the component of the wave vector, orthogonal to the slab, of the coherent input field. It satisfies the complex polariton dispersion relation $k_z^2 + p^2 = \varepsilon(\omega)\omega^2/c^2$, $\varepsilon(\omega)$ being the complex linear dielectric function. Energy and in-plane momentum are conserved, respectively $\omega_1 + \omega_2 = 2\omega_I$ and $\mathbf{p}_1 + \mathbf{p}_2 = \mathbf{p}_I$. The third order nonlinear susceptibility in Eq. (1) can be written as

$$\chi^{(3)} = \sum_{n,n',n_1,n_2} \chi_n^*(\omega_1)\chi_{n'}^*(\omega_2) \times \left(Y_{n_1,n_2}^{n,n'} - W_{n_1,n_2}^{n,n'}(2\omega_I)\right)\chi_{n_1}(\omega_I)\chi_{n_2}(\omega_I). \tag{2}$$

In Eq. (2), $\chi_n = M_n/(\omega_n - \omega - i\gamma_n)$ are linear susceptibility terms, where ω_n and γ_n are the energy and the damping of the nth exciton level. $Y - W$ in Eq. (2) originates from the Coulomb interaction between two *eh*-pairs. In particular, Y describes exciton–exciton interactions as in the usual Hartee–Fock semiconductor Bloch equations, and $W(2\omega_I)$ is the spectral distribution of the exciton–exciton correlation function [13,17]. It contains resonances due to bound biexciton molecules as well as contributions from the continuum of unbound biexciton states [17]. By inserting Eq. (1) into the operator Maxwell equation for the electric field, we obtain the scattered field operator at mode 1 [13].

We consider a semiconductor slab with ideal antireflection coatings at its sides so that, for the range of frequency, which we will take into account, the photon flux remains unaffected when crossing the interface. In vacuum, the field components $\hat{E}_m^+(z, \omega)$ can be written in terms of photon operators $\hat{E}_m^+(z, \omega) = K_m^0(\omega)\mathrm{e}^{ik_z^0 z}\hat{a}_m(\omega)$, where $k^0 = \omega/c$and $K_m^0(\omega)$ is a normalization coefficient ensuring satisfaction of the commutation rules for the light field. For future use, it is essential to introduce the photon number operator as $\hat{n}_m(\omega) = \int_{\omega-\delta}^{\omega+\delta} \mathrm{d}\omega' \hat{a}_m^\dagger(\omega')\hat{a}_m(\omega')$, where $2\delta > 0$ and small is the bandwidth of the photon-counter.

The electric-field operator calculated within the Heisenberg–Langevin approach allows us to calculate

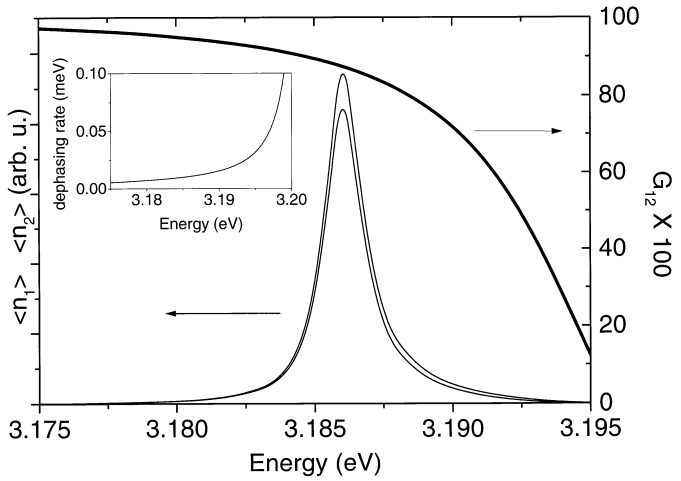

Fig. 2. The mean number of emitted photons $\langle \hat{n}_{1,2} \rangle$ and the degree of entanglement $\mathscr{G}_{12}$ calculated at the phase-matching resonance condition as a function of the incident energy for a CuCl slab of thickness $L = 50$ μm. The inset shows the dephasing rate as a function of energy used for the calculations.

field expectation values as well as higher order field–field correlations [13]. In HRS (in contrast to FWM), there is no input field at mode 2, the expectation value of the nonlinear polarization in Eq. (1) is zero, and as a consequence the expectation value of the scattered field $\langle \hat{a}_1(\omega_1) \rangle$ is equal to zero. Instead the emission spectrum is different from zero owing to vacuum fluctuations at mode 2 which determine the decay of the virtually excited biexcitons. The mean number of emitted photons $\langle \hat{n}_1(\omega_1) \rangle$ is given by

$$\langle \hat{n}_1 \rangle = Q_{1,2}^2 |\chi^{(3)} E_I^2(\omega_I)|^2 g_1(L), \tag{3}$$

where $Q_{1,2} = \omega_1 \omega_2 / (2\varepsilon_0 c^2 \sqrt{k_{z_1}^{\mathrm{R}} k_{z_2}^{\mathrm{R}}})$, and $g_1(z)$ describes the propagation and the phase-matching resonance condition. It is given by

$$g_1(z) = \frac{k_{z_2}^{\mathrm{R}}}{k_{z_2}} \left\{ \frac{\mathrm{e}^{-2k_1^{\mathrm{I}} z}}{2k_{z_I} - k_{z_1} - k_{z_2}} \left(\frac{1 - \mathrm{e}^{-2(2k_{z_I}^{\mathrm{I}} - k_{z_1}^{\mathrm{I}})z}}{2(2k_{z_I}^{\mathrm{I}} - k_{z_1}^{\mathrm{I}})} + i \frac{1 - \mathrm{e}^{-\mathrm{i}(\Delta k_z^*)z}}{\Delta k_z^*} \right) + c.c. \right\}. \tag{4}$$

The phase mismatch Δk_z in Eq. (4) is given by $\Delta k_z = 2k_{z_I} - k_{z_1} - k_{z_2}^*$. The role of modes 1 and 2 can be interchanged. Thus the spontaneous decay of virtually excited biexcitons can give rise to two light beams 1 and 2. They are *sons* of the virtually excited states with two *eh* pairs. Of course, $\langle \hat{n}_2 \rangle$ can be obtained from Eq. (3) simply by exchanging the labels 1 and 2. The intensity of the scattered fields $\langle \hat{n}_{1,2} \rangle$, calculated at the phase-matching resonance condition ($\mathrm{Re}[\Delta k_z] = 0$), are displayed in Fig. 2 as a function of the incident energy ω_I for a CuCl slab of 50 μm. We have considered a forward scattering geometry with incident light orthogonal to the slab and observation angles $\cong 10°$. The two-exciton correlation $W(2\omega_I)$ in the resonant nonlinear susceptibility was calculated by assuming a one bound biexciton level with energy $\omega_2 = 6.372$ eV and with a homogeneous broadening $\Gamma_2 = 1.6$ meV. The exact two-exciton correlation $W(2\omega_I)$ has been calculated numerically for a one-dimensional semiconductor model with long-range Coulomb interaction [17]. The various scattering mechanisms determine an increasing trend of the dephasing rate γ as the energy approaches the exciton resonance. We have used a dephasing rate, displayed in the inset in Fig. 2, fitting quite well the experimental data [18]. The peak at higher energy ($\langle \hat{n}_2 \rangle$) is slightly lower than the other due to a larger degree of dephasing during propagation.

Let us now analyze the correlation properties of the scattered light. We first consider the two-mode intensity correlation functions $\langle \hat{n}_2 \hat{n}_1 \rangle$. and obtain

$$\langle \hat{n}_1 \hat{n}_2 \rangle = \langle \hat{n}_2 \hat{n}_1 \rangle = Q_{1,2}^2 |\chi^{(3)} E_I^2(\omega_I)|^2 g_{1,2}(L), \tag{5}$$

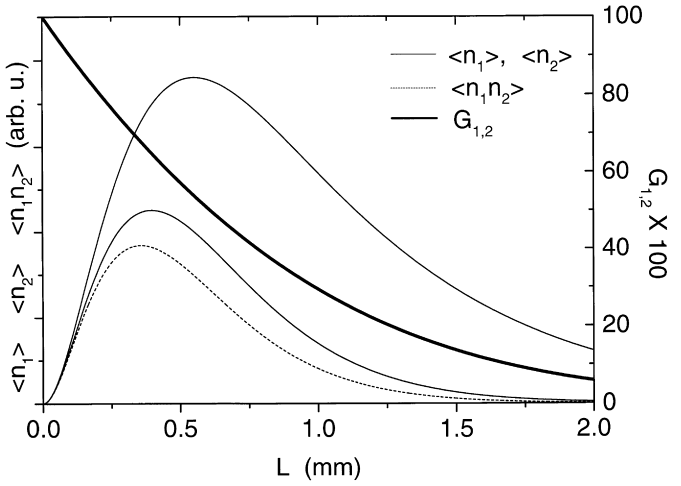

Fig. 3. The mean number of emitted photons $\langle \hat{n}_{1,2} \rangle$, the pair correlations $\langle \hat{n}_1 \hat{n}_2 \rangle$, and the degree of entanglement G_{12} calculated at the phase-matching resonance condition as a function of the slab thickness.

where

$$g_{1,2}(z) = \mathrm{e}^{-2(k^{\mathrm{I}}_{z_1} + k^{\mathrm{I}}_{z_2})z} \left| \frac{1 - \mathrm{e}^{(2k_{z_I} - k_{z_1} - k_{z_2})z}}{2k_{z_I} - k_{z_1} - k_{z_2}} \right|^2 . \qquad (6)$$

As we have already observed, modes 1 and 2 are not independent but are related by the total energy and momentum conservation. For each other mode m, different from mode 2 and from the input mode I, we obtain (in the low density limit) $\langle : \hat{n}_1 \hat{n}_m : \rangle = 0$ ($::$ denotes normal order). This is due to the fact that, at lowest order, photons are created in pairs and, if 1 and m do not belong to the same pair, $\langle : \hat{n}_1 \hat{n}_m : \rangle \neq 0$ implies a higher order scattering process. As a consequence, the single-mode correlation functions, calculated in the low-density limit, are zero. In particular, we obtain $\langle : \hat{n}_1^2 : \rangle = \mathrm{O}(E_I^6)$, where $\mathrm{O}(E_I^6)$ indicates those terms of the order ≥ 6.

The intensity correlation functions in the low-density limit exhibit quantum features as they violate the classical Cauchy–Schwartz inequality, $\langle : \hat{n}_2 \hat{n}_1 : \rangle^2 \leq \langle : \hat{n}_1^2 : \rangle \langle : \hat{n}_2^2 : \rangle$. Entangled photon pairs exhibit fourth order interference in the joint detection probability that cannot be obtained with classical systems [4]. In the classical systems, the visibility of interference fringes cannot exceed 50%. The visibility of interference can be written in terms of the fourth order correlations as $U = 2\langle : \hat{n}_2 \hat{n}_1 : \rangle / (\langle : \hat{n}_1^2 : \rangle + \langle : \hat{n}_2^2 : \rangle + 2\langle : \hat{n}_2 \hat{n}_1 : \rangle)$. By using the correlations calculated above, we obtain $U = 1 - \mathrm{O}(E_I^6)$, i.e. fringe visibility reaching 100 in the low-density limit. Thus, the fourth order interference could be used to observe experimentally the quantum features of light emerging from the spontaneous decay of biexcitons.

In order to quantify the degree of quantum entanglement, we take advantage of the following inequality for quantum fields, $\langle : \hat{n}_2 \hat{n}_1 : \rangle^2 \leq \langle \hat{n}_1^2 \rangle \langle \hat{n}_2^2 \rangle$. Ideal entangled two-particle states realize the maximum violation of the Cauchy–Schwarz inequality compatible with the above inequality ($\langle : \hat{n}_2 \hat{n}_1 : \rangle^2 = \langle \hat{n}_1^2 \rangle \langle \hat{n}_2^2 \rangle$). As a consequence the ratio, $0 \leq \mathscr{G}_{1,2} \equiv \langle : \hat{n}_2 \hat{n}_1 : \rangle / \sqrt{\langle \hat{n}_1^2 \rangle \langle \hat{n}_2^2 \rangle} \leq 1$ gives a measure of the degree of entanglement of the 1–2 pair. By using the correlation functions calculated above in the low-density limit, we obtain

$$\mathscr{G}_{1,2} = \frac{g_{12}(L)}{\sqrt{g_1(L) g_2(L)}} . \qquad (7)$$

In the absence of the attenuation $g_{12}(L) = g_1(L) = g_2(L)$, and we obtain $\mathscr{G}_{1,2} = 1$. Thus, we can conclude that the optical decay of biexciton, for negligible absorption, produces an ideal entangled pair of photons. Of course, the photon reabsorption tends to destroy the ideal entanglement. In particular, the degree of entanglement is affected by those events which, after the biexciton decay, scatter one polariton of the pair. A simple criterium for negligible reabsorption is $k^{\mathrm{I}} L \ll 1$.Thus the degree of

entanglement depends strongly on the energy of the scattered light and on the length of the slab. Fig. 2 displays $G_{1,2}$ as a function of the incident energy. Reabsorption causes $G_{1,2}$ to go rapidly to zero as Ω_I approaches the energy of the 1s exciton level $\omega_1 = 3.2026$ eV. The noticeable biexciton binding energy, determined by the Coulomb interaction between excitons, in CuCl (as well as in other large-gap semiconductors) permits the detection of Hyper Raman lines by using incident light with an energy sufficiently far from the exciton level to prevent strong reabsorption. This permits the polariton pairs to escape the crystal with a high degree of entanglement, as shown in Fig. 2. The pair formation and reabsorption during propagation is shown in Fig. 3, which gives information on the sample thickness more conveniently to observe the photon pairs. As expected pair correlations $\langle \hat{n}_1 \hat{n}_2 \rangle$ are more fragile, with respect to absorption, than signals $\langle \hat{n}_{1,2} \rangle$. We observe that the HRS lines well separated from luminescence bands and corresponding to scattering into two lower polaritons have been observed in CuCl [10] as well in other large-gap bulk semiconductors [11]. These lines, according to the results presented here, appear to be suitable for observing the quantum correlation between the emitted photon pairs.

In conclusion, we have shown that it is possible to transfer the exciton–exciton Coulomb correlation to photons, producing thus pairs of near-gap photons with a high degree of quantum correlation. A small reabsorption after pair emission is the condition to produce output photon pairs with a high degree of quantum correlation. The observation of near-gap entangled photon pairs, due also to the possibility of coherent controlling resonant excitations in semiconductor systems [19,20], is expected to be promising towards the realization of integrated quantum optical devices.

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