Biphotons from Biexcitons

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We analyze the spontaneous and stimulated optical decay of states with two electron–hole pairs virtually excited by optical pumping. We find that the photon pairs from the spontaneous fission in bulk large-gap semiconductors and in semiconductor microcavities exhibit quantum features which can be observed by coincidence detection.

Coherent nonlinear effects and quantum-optical effects in semiconductors and in semiconductor microcavities (SMCs) are attracting growing interest for exploring fundamental light–matter interactions in mesoscopic systems as well as for the future implementation of devices based on quantum correlations [1]. Here we present theoretical results on the spontaneous four-wave mixing (FWM) process in bulk semiconductors and in cavity embedded quantum wells (QWs) obtained by using a fully quantum-mechanical perturbative theory of the nonlinear optical response in semiconductors [2]. This coherent quantum-optical process, also known as hyper-Raman scattering (HRS) was first observed in 1978 by Hönerlage et al. [3] in a CuCl single crystal.

Transient or frequency-resolved FWM are among the most widely used techniques for probing the optical properties of semiconductors. Referring to elementary excitations in semiconductors, the FWM process can be schematically described as follows: two incident pump photons with in-plane wave vector $\mathbf{q}_p$, propagating inside the crystal as excitonic polaritons, drive a virtual state with two electron–hole (e–h) pairs with wave vector $2\mathbf{q}_p$, while a probe beam with wave vector $\mathbf{q}_1$ stimulates the optical decay of the state with two e–h pairs into states with one e–h pair at $\mathbf{q}_1$ and $\mathbf{q}_2 = 2\mathbf{q}_p - \mathbf{q}_1$. The states with two e–h pairs can be true biexcitons as well as two-exciton scattering states. This stimulated fission process thus determines the generation of a new beam at $\mathbf{q}_2$ (FWM) and the amplification of the probe beam at $\mathbf{q}_1$ (parametric gain). Recently, giant optical gain due to this χ^3 process has been observed in a semiconductor microcavity (MC) [4, 5]. If we maintain the pump beam while eliminating the probe beam, we have no coherent emission at $\mathbf{q}_2$, unless we introduce the quantization of the light field [2, 6, 7]. The quantum fluctuations of the light field can play the role of the probe beam stimulating the optical decay of states with two e–h pairs , thus quantum fluctuations at a generic in-plane wave vector $\mathbf{q}_1$ can determine light emission in the direction $\mathbf{q}_2 = 2\mathbf{q}_p - \mathbf{q}_1$ and vice versa [8]. The final result is emission of photon pairs in directions fixed by $\mathbf{q}_1$ and $\mathbf{q}_2$. In order to analyze the generation and propagation of the scattered polaritons we have derived the Heisenberg equation of motion for the electro-

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nic polarization operator interacting with the quantized light field [2]. All calculations have been performed in the low density limit, including in the nonlinear optical response those contributions related to the third-order nonlinear polarization of the interacting electron system. We first consider a semiconductor slab of thickness L orthogonal to the z direction illuminated by a monochromatic pump beam at energy ω_p. The electric-field operator calculated within the Heisenberg-Langevin approach allows us to calculate field expectation values as well as higher-order field–field correlations [8]. The mean number of emitted photons in direction **1** $\langle \hat{n}_1(\omega_1) \rangle$ is given by [8, 9]

$$\langle \hat{n}_1 \rangle = Q_{1,2}^2 \, |\chi^{(3)} E_\mathrm{p}^2(\omega_\mathrm{p})|^2 \, g_1(L) \,, \tag{1}$$

where E_p is the amplitude of the coherent input field incoming into the semiconductor slab, $Q_{1,2} = \omega_1 \omega_2 / (2\varepsilon_0 c^2 \sqrt{k_{1z}^{\mathcal{R}} k_{2z}^{\mathcal{R}}})$, $k_{jz} = k_{jz}^{\mathcal{R}} + i k_{jz}^{\mathcal{I}}$ $(j = 1, 2, \mathrm{p})$ is the component of the wave vector, orthogonal to the slab. It satisfies the complex polariton dispersion relation $k_z^2 + p^2 = \varepsilon(\omega)\, \omega^2/c^2$, with $\varepsilon(\omega)$ being the complex linear dielectric function. Total energy and in-plane momentum are conserved, respectively, $\omega_1 + \omega_2 = 2\omega_\mathrm{p}$ and $\mathbf{q}_1 + \mathbf{q}_2 = \mathbf{q}_\mathrm{p}$. The third-order nonlinear susceptibility $\chi^{(3)}$ in Eq. (1) contains contributions from all the nonlinearities of the interacting electron system [8]. The function $g_1(z)$ describes the propagation and the phase-matching resonance condition [9]. 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 which are sons of the virtually excited states with two e–h pairs. The peak intensities of the scattered fields $\langle \hat{n}_{1,2} \rangle$, calculated at the phase-matching resonance condition, are displayed in Fig. 1 as a function of the incident energy ω_p for a CuCl slab of 50 μm. We have considered a forward scattering geometry with incident light orthogonal to the slab and observation angles $\approx 10°$. The two-exciton correlation in the resonant nonlinear susceptibility has been calculated by assuming one bound biexciton level with energy $\omega_\mathrm{xx} = 6.372$ eV and with homogeneous broadening $\Gamma_\mathrm{xx} = 1.6$ meV. The intensity of the scattered fields exhibit a resonance at $\omega = \omega_\mathrm{xx}/2$. Let us now analyze the correlation properties of the scattered light. We first consider the two-mode intensity correlation functions $\langle \hat{n}_2 \hat{n}_1 \rangle$. We obtain

$$\langle \hat{n}_1 \hat{n}_2 \rangle = \langle \hat{n}_2 \hat{n}_1 \rangle = Q_{1,2}^2 \, |\chi^{(3)} E_\mathrm{p}^2(\omega_\mathrm{p})|^2 \, g_{1,2}(L) \,, \tag{2}$$

where $g_{1,2}(z)$ describes the propagation of the photon pairs. As we have already observed, modes 1 and 2, are not independent but are related by total energy and momentum conservation. For each other mode m, different from mode 2 and from the input mode p, 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 $\langle : \hat{n}_1^2 : \rangle = \mathcal{O}(E_\mathrm{p}^6)$, where $\mathcal{O}\left(E_\mathrm{p}^6\right)$ indicates those terms of order ≥ 6. The calculated intensity correlations exhibit quantum features as they violate the classical Cauchy-Schwartz inequality, $\langle : \hat{n}_2 \hat{n}_1 : \rangle^2 \leq \langle : \hat{n}_1^2 : \rangle \, \langle : \hat{n}_2^2 : \rangle$. In order to quantify the degree of quantum correlation, 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 we can define a quality factor, $0 \leq \mathcal{Q} \equiv \langle : \hat{n}_2 \hat{n}_1 : \rangle / \sqrt{\langle \hat{n}_1^2 \rangle \langle \hat{n}_2^2 \rangle} \leq 1$, which gives a measure of the degree of quantum correlation of the 1–2 pair. In absence of attenuation $g_{12}(L) = g_1(L) = g_2(L)$ and we obtain $\mathcal{Q} = 1$. Thus, we can conclude that the optical decay of biexciton, for negligible absorption, produces an ideal entangled pair of photons. Of course photon reabsorption tends to destroy 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 criterion for negligible reabsorption is $k^{\mathcal{I}} L \ll 1$. Thus, the degree of entanglement depends strongly on the energy of the scattered light and on the length of the slab. Figure 1 displays $\mathcal{Q}$ as a function of the incident energy. Reabsorption causes $\mathcal{Q}$ to go rapidly to zero as Ω_{I} approaches the energy of the 1s exciton level $\omega_0 = 3.2026$ 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 energy sufficiently far from the exciton level to prevent strong reabsorption. We point out that HRS lines well separated from luminescence bands and corresponding to scattering into two lower polaritons have been observed in CuCl [10] as well as in other large-gap bulk semiconductors. These lines, according to the results presented here, appear to be suitable to observe the quantum correlation among the emitted photon pairs. In contrast to bulk materials, the optical properties of semiconductor MCs can be tailored by the independent design of the cavity and of the embedded QWs. As a result, in contrast to bulk materials, it is possible to obtain important levels of quantum correlation even at photon energies close to the excitonic resonance. For reasons of space we do not present here any numerical results on MCs, we only point out that a quantum-noise reduction in the intensity difference of the two emitted beams of more than 80% can be obtained taking into account parameters for realistic MCs [11].

The main limitation of the approach here presented is its applicability to the very low excitation density regime. As a consequence it cannot be directly applied to analyze coherent nonlinear phenomena in the intermediate density regime before dissociation [4]. Thus, we propose a direct extension of the above approach to treat wave-mixing phenomena in MCs in the intermediate density regime. Let us consider the coherent nonlinear wave mixing between a pump (p), a probe (1) and the idler (2) beams in a MC as in the experiment by Savvidis et al. [4] with co-circularly polarized beams. We

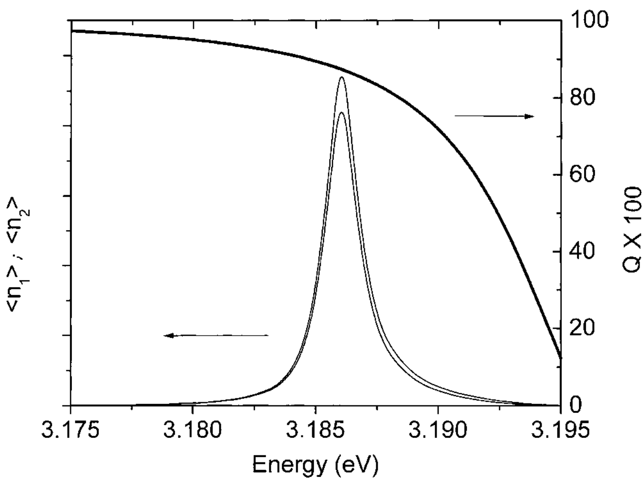

Fig. 1. The mean number of emitted photons $\langle \hat{n}_{1,2} \rangle$ and the degree of entanglement $\mathcal{Q}$ calculated at the phase-matching resonance condition as a function of the incident energy for a CuCl slab of thickness $L = 50$ μm

adopt a direct extension of Eq. (10) of Ref. [2] including multiple polariton scattering. The set of six coupled equations of motion for the intracavity fields E_j (j = 1, 2, p) and the excitonic polarization P_j is given by

$$\frac{\partial}{\partial t} E_j = -(\gamma_c + i\omega_c^{(j)})\, E_j + iVP_j + \sqrt{g_c}\, E_j^{in}\,,$$
$$\frac{\partial}{\partial t} P_j = -(\gamma_x + i\omega_0)\, E_j + iVE_j + \Omega_j^{(NL)}\,, \tag{3}$$

where $\omega_c^{(j)}$, and ω_0, γ_c and γ_0 are the frequencies and dephasing of cavity photons and QW excitons, respectively. E_j^{in} is the coherent input field different from zero only for the pump and probe waves, and g_c is a the mirror coupling coefficient. $\Omega_j^{(NL)}$ is the nonlinear source term determining the wave mixing. It can be written as

$$\Omega_j^{(NL)}(t) = -i \sum_{i,l,m} \delta\left(\mathbf{q}_l + \mathbf{q}_m - \mathbf{q}_i - \mathbf{q}_j\right) \times \left[\frac{\beta}{2} P_i^*(t)\, P_l(t)\, P_m(t) + \frac{1}{n_c}\, P_i^*(t)\, P_l(t)\, E_m(t) - \frac{i}{2}\, P_i^*(t) \int\limits_{-\infty}^{t} F(t-t')\, P_l(t')\, P_m(t')\, \mathrm{d}t'\right]. \tag{4}$$

In Eq. (4) n_c is the exciton saturation density, β describes the mean-field exciton–exciton interaction, and the exciton–exciton correlation is contained in the retarded memory function $F(\tau)$ [12]. Figure 2 displays the reflected probe power spectrum in the absence and in the presence of the pump beam for a MC with $V = 3.8$ meV with the pump and probe beams in the configuration adopted in Ref. [4]. The calculations, although preliminary and performed without including the contributions beyond mean field $F(\tau)$, appear in close agreement with the experimental results [4].

In conclusion, we have analyzed the spontaneous fission processes in semiconductors, and we have shown that the photon pairs emerging from the spontaneous optical decay of two e–h pairs can exhibit a noticeable degree of quantum correlations. We have also proposed a new set of nonlinear equations for the analysis of multiple coherent polariton scattering in semiconductor microcavities.

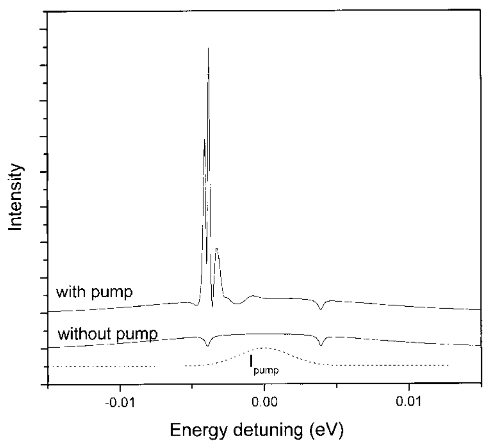

Fig. 2. Reflected probe spectra for pump off and pump on, and spectrum of the input pump beam (dashed curve)

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