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Optics Communications 280 (2007) 457–462
OPTICS COMMUNICATIONS
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Optical Bloch equations with dual photon excitation explain the polarization dependence of four wave mixing quantum beats

Wenfeng Wang [a,b,*], Jun Zhang [c], Daan Lenstra [d]

[a] Faculty of Science, VU University Amsterdam, De Boelelaan 1081, 1081HV Amsterdam, The Netherlands
[b] Faculty of Physics and Electronics, Hubei University, Wuhan, China
[c] School of Computing, Science and Engineering, Salford University, Manchester M54WT, United Kingdom
[d] Faculty of EEMCS, Delft University of Technology, Delft, The Netherlands

Received 20 March 2007; received in revised form 4 July 2007; accepted 6 July 2007

Abstract

We explain the polarization dependence of four wave mixing (FWM) quantum beats for semiconductors as essentially due to the spin phase correlations of photo-excited electrons, rather than to Coulomb interaction between the electrons. A theoretical analysis is given within the framework of optical Bloch equations for the light–semiconductor interactions and the Luttinger–Kohn model for the band structure. Residual Coulomb interactions between charge carriers are ignored. The results suggest that the polarization dependence of FWM quantum beats is a purely coherent effect of dual photon excitations, rather than, e.g., exciton–exciton Coulomb interaction. We show that the coherence transfer between the excited states is responsible for the FWM in a configuration with orthogonally polarized pump and probe.

PACS: 42.50.Md; 78.66.Fd; 42.65.−k

Keywords: Optical transient phenomena; Optical properties of semiconductors; Nonlinear optics

1. Introduction

Polarization dependent four wave mixing (FWM) quantum beats [1–11] have been observed after simultaneous excitations of two optical transitions, associated with heavy-hole (HH) and light-hole (LH). The signal magnitude and its beat phase depend on the relative linear polarization of pump and probe. This phenomenon has been analyzed by applying semiconductor Bloch equations [12,14,13] (SBE) for excitations in a six-band model by broad spectrum pump and probe pulses [1,2]. However the theoretical investigations predicted, in contradiction with experimental observations, *identical* FWM intensities for the two polarization configurations: pump and probe having either parallel or perpendicular linear polarizations. Since then, several attempts have been made to theoretically explain the observed phenomena. The bi-exciton model, with Coulomb interaction between the excitons as the essential ingredient, has been claimed to give a successful explanation [5,6,3,7–11].

In this work, an alternative explanation, within the optical Bloch equation (OBE) formalism, is presented, in which the spin phase correlations of the excited electrons are emphasized, but residual Coulomb interaction between excited carriers is neglected. We suggest that the polarization dependence of FWM quantum beats is a purely coherent light-matter interaction effect, rather than due to electron Coulomb interaction, e.g. in a bi-exciton state.

* Corresponding author. Address: Faculty of Physics and Electronics, Hubei University, College Road 11, 430062 Wuhan, China. Tel.: +86 27 88662513; fax: +86 27 88663390.
E-mail address: w.wang@hubu.edu.cn (W. Wang).

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doi:10.1016/j.optcom.2007.07.023

We assume that the electrons in semiconductors can be completely described within a space with six states: $|1/2,\pm1/2\rangle$, $|3/2,\pm1/2\rangle$, and $|3/2,\pm3/2\rangle$. The spatial properties of these states are defined in the conventional way [13] as

$$\begin{aligned}
&|1/2,1/2\rangle = |S\uparrow\rangle\\
&|1/2,-1/2\rangle = |S\downarrow\rangle\\
&|3/2,3/2\rangle = -\sqrt{1/2}(|X\uparrow\rangle + \mathrm{i}|Y\uparrow\rangle)\\
&|3/2,1/2\rangle = -\sqrt{1/6}(|X\downarrow\rangle + \mathrm{i}|Y\downarrow\rangle) + \sqrt{2/3}|Z\uparrow\rangle\\
&|3/2,-1/2\rangle = \sqrt{1/6}(|X\uparrow\rangle - \mathrm{i}|Y\uparrow\rangle) + \sqrt{2/3}|Z\downarrow\rangle\\
&|3/2,-3/2\rangle = \sqrt{1/2}(|X\downarrow\rangle - \mathrm{i}|Y\downarrow\rangle).
\end{aligned} \tag{1}$$

The band structure, without split-off bands, of III–V semiconductors is denoted with $|c1\rangle$, $|c2\rangle$, $|h1\rangle$, $|h2\rangle$, $|l1\rangle$, and $|l2\rangle$, with c, h and l standing for conduction, HH, and LH bands, which are twofold degenerate. For small Bloch momentum $\vec{k}$, we approximate the eigenstates of $|c1\rangle$, $|c2\rangle$ by just the basis states $|1/2,\pm1/2\rangle$ for conduction bands. For valence bands the wave functions are more complicated. They can be written in a compact matrix multiplication form as [15]

$$\begin{pmatrix} |h1\rangle\\ |h2\rangle\\ |l1\rangle\\ |l2\rangle \end{pmatrix} = \frac{1}{\sqrt{N_i}} \begin{pmatrix} -b & R_h & 0 & -c^*\\ -c & 0 & R_h & b^*\\ R_l & b^* & c^* & 0\\ 0 & c & -b & R_l \end{pmatrix} \begin{pmatrix} |3/2\rangle\\ |1/2\rangle\\ |-1/2\rangle\\ |-3/2\rangle \end{pmatrix} \tag{2}$$

with $R_h = H_h - E_h$, $R_l = -H_l + E_l$, and $N_i = |R_i|^2 + |c|^2 + |b|^2$, $i = h,l$. Here, we use the conventional notations [13], and $\hbar \equiv 1$ is assumed in the following:

$$\begin{aligned}
&b = \sqrt{3}\bar{\gamma}m_0^{-1}(k_x - \mathrm{i}k_y)k_z,\\
&c = \sqrt{3}\bar{\gamma}(2m_0)^{-1}[(k_x^2 - k_y^2) - 2\mathrm{i}k_xk_y],\\
&H_h = -(2m_0)^{-1}[\gamma_1 k^2 - \gamma_2(2k_z^2 - k_\perp^2)],\\
&H_l = -(2m_0)^{-1}[\gamma_1 k^2 + \gamma_2(2k_z^2 - k_\perp^2)].
\end{aligned} \tag{3}$$

As is well known, the optical transitions, for a given $\vec{k}$, between the conduction and valence bands in direct bandgap III–V semiconductors have different energies ϵ_h and ϵ_l for HH and LH bands, respectively. Optical transitions between HH and LH bands are forbidden. Meanwhile, the optical transitions $|h\rangle \to |c\rangle$ and $|l\rangle \to |c\rangle$ are assumed to be independent of each other. This means that the transitions $|h\rangle \to |c\rangle$ involve the electronic states $|h\rangle$ and $|c\rangle$ only, *independent* of the electronic states $|l\rangle$. Therefore, with respect to $|h\rangle \to |c\rangle$ transitions, we have the initial state of this excitation

$$|I\rangle_h = a_{h1}^\dagger a_{h2}^\dagger |0_h\rangle, \tag{4a}$$

with $|0_h\rangle \equiv |0_{h1}0_{h2}0_{c1}0_{c2}\rangle$, and the initial state

$$|I\rangle_l = a_{l1}^\dagger a_{l2}^\dagger |0_l\rangle, \tag{4b}$$

for the transition $|l\rangle \to |c\rangle$, with $|0_l\rangle \equiv |0_{l1}0_{l2}0_{c1}0_{c2}\rangle$.

Only resonant optical transitions will be considered. Therefore, the photon energy $\hbar\omega_0$ is well defined for each band and wave vector k_0. We shall therefore omit in the following the wave vector $\vec{k}$ as the subscript in the notation, e.g. $a_{h1}^\dagger \equiv a_{h1,\vec{k}}^\dagger$.

2. Model

We specifically investigate one type of the FWM quantum beats, the Λ three-level quantum beats [16], in which the excited state is involved in one excitation by dual photons, with different frequencies. This should not be understood as two, one by one, excitations giving two excited states. Analogously, for the case of a semiconductor, the energies of the dual photons are just the resonant energies of the HH and LH electronic states with the same wave vector. The excited state of the dual photons excitation is composed of two parts, one from the initial state $|I\rangle_h$ of the $|h\rangle \to |c\rangle$ transitions, the other from the initial state $|I\rangle_l$ of the $|l\rangle \to |c\rangle$ transitions.

Because the exact forms of the excited state depend on the polarization of the incident light, we consider here for the sake of clarity the case of linearly polarized light, polarized in the x–y plane. Therewith the incident laser field $\mathscr{E}(t)$ propagating in z direction, along the chosen quantization direction. By mathematical transformation of these results, one can obtain those for the circularly polarized case.

First we consider the dipole transition between the basis states in Eq. (1). Note that the z components of right hand side of Eq. (6) do not contribute, since the light polarization lies in the x–y plane. With $\langle S|x|X\rangle = M$ and due to the symmetry of the S state, the x–y plane components of the dipole matrix elements are

$$\begin{aligned}
&\langle 1/2,1/2|\vec{r}_\perp|3/2,3/2\rangle = -\sqrt{1/2}(\hat{x} + \mathrm{i}\hat{y})M\\
&\langle 1/2,1/2|\vec{r}_\perp|3/2,1/2\rangle = -\sqrt{2/3}\hat{z}M\\
&\langle 1/2,1/2|\vec{r}_\perp|3/2,-1/2\rangle = \sqrt{1/6}(\hat{x} - \mathrm{i}\hat{y})M\\
&\langle 1/2,1/2|\vec{r}_\perp|3/2,-3/2\rangle = 0\\
&\langle 1/2,-1/2|\vec{r}_\perp|3/2,3/2\rangle = 0\\
&\langle 1/2,-1/2|\vec{r}_\perp|3/2,1/2\rangle = -\sqrt{1/6}(\hat{x} + \mathrm{i}\hat{y})M\\
&\langle 1/2,-1/2|\vec{r}_\perp|3/2,-1/2\rangle = \sqrt{2/3}\hat{z}M\\
&\langle 1/2,-1/2|\vec{r}_\perp|3/2,-3/2\rangle = \sqrt{1/2}(\hat{x} - \mathrm{i}\hat{y})M.
\end{aligned} \tag{5}$$

Thereby, with the wave functions of valence bands in Eq. (2), the optical transitions between the conduction bands and valence bands can be written in units of M as

$$\begin{aligned}
&\langle c1|\vec{r}_\perp|h1\rangle = -ub(\hat{x} + \mathrm{i}\hat{y})\\
&\langle c1|\vec{r}_\perp|h2\rangle = -uc(\hat{x} + \mathrm{i}\hat{y}) + wR_h(\hat{x} - \mathrm{i}\hat{y})\\
&\langle c2|\vec{r}_\perp|h1\rangle = -wR_h(\hat{x} + \mathrm{i}\hat{y}) + uc^*(\hat{x} - \mathrm{i}\hat{y})\\
&\langle c2|\vec{r}_\perp|h2\rangle = -ub^*(\hat{x} - \mathrm{i}\hat{y})\\
&\langle c1|\vec{r}_\perp|l1\rangle = uR_l(\hat{x} + \mathrm{i}\hat{y}) + wc^*(\hat{x} - \mathrm{i}\hat{y})\\
&\langle c1|\vec{r}_\perp|l2\rangle = -wb(\hat{x} - \mathrm{i}\hat{y})\\
&\langle c2|\vec{r}_\perp|l1\rangle = -wb^*(\hat{x} + \mathrm{i}\hat{y})\\
&\langle c2|\vec{r}_\perp|l2\rangle = -wc(\hat{x} + \mathrm{i}\hat{y}) - uR_l(\hat{x} - \mathrm{i}\hat{y}),
\end{aligned} \tag{6}$$