Semiconductor Optical Bloch Equations Explain Polarization Dependent Four Wave Mixing Quantum Beats in Bulk Semiconductors

W. Wang[1], J. Zhang[2] and D. Lenstra[1]

Department of Physics and Astronomy, Vrije Universiteit Amsterdam,
De Boelelaan 1081, 1081HV, Amsterdam, The Netherlands[1], IMR, Salford University, Manchester, U.K.[2]

We re-examined the semiconductor Bloch equations (SBE) and found that, excluding the many-body electron Coulomb interaction, SBE do not reduce to the optical Bloch equations (OBE). By applying OBE on semiconductors straightforward, we propose an explanation for the polarization dependent four wave mixing (FWM) quantum beats, which could not be explained by the conventional SBE but attributed to bi-exciton processes. We suggest that this effect is simply a coherent process rather than due to electron-electron Coulomb interaction; the coherence transfer between the excited states accounts for the FWM in the configuration with pump and probe in orthogonal polarization states.

Introduction

Polarization-dependent four wave mixing (FWM) quantum beats [1–4] have been observed after simultaneous excitations of two optical transitions, i.e. heavy-hole and light-hole. The signal magnitude and its beat phase depend on the polarization of the pump and probe with respect to each other. This phenomenon has been analyzed with applying semiconductor Bloch equations (SBE) for excitations in a six-band model by broad spectrum pump-probe pulses. [1, 2] However this theory predicts *identical* FWM intensities for the two polarization configurations: pump and probe have either parallel or perpendicular linear (circular) polarizations. Since then, a great effort has been devoted to theoretically explain this phenomenon. One of the successful explanations is the bi-exciton theory. [3, 4] However, why the optical Bloch equation (OBE) can do very well in atomic optics, but the SBE fails in this specific investigation is an interesting question, and it should not be, without constructive considerations, simply answered by the fact that SBE formalism does not include exciton-exciton processes or a Hartree-Fock approximation is not capable to describe quasi-stationary of the excited semiconductor. [5]
The SBE is developed as the similar basis as the OBE. [6, 7] The main extra efforts of the former theory are its focusing on the electron many-body effects, such as semiconductor band gap renormalization, excitonic effects, phase-space filling, etc, which do not exist in OBE. However, in the present work we will completely neglect the Coulomb interaction between the carriers, which means that no excitonic effects are considered. Based on this assumption, we compare the SBE and OBE formalism. We find that the SBE do not reduce to the OBE formalism, contrary to what one would expect.
In an electron two-level system with vacuum state is defined as $|0_a 0_b\rangle$, i.e. an electron can be in two states: $|a\rangle$ and $|b\rangle$. In case of the resonant light excitation, the *system* of the electron can be inverted from the electron system ground state $|g\rangle$ which gives $|0_a 1_b\rangle$, to the system excited state $|e\rangle$ which gives $|1_a 0_b\rangle$. Correspondingly, in the SBE, the states of the electron system $|e\rangle$ and $|g\rangle$ will be given as $|1_c 0_v\rangle$ and $|0_c 1_v\rangle$, in case that an electron in semiconductors can only be in two states: $|c\rangle$ and $|v\rangle$.

In the normal formalism of SBE, what one finds are a coupled equations between polarization P_{cv} and the occupation probabilities for the electron in the state $|c\rangle$ and $|v\rangle$: n_c and n_v. [7, 8] However, to be consistent with the OBE formalism, the coupled equations should be between the electron system polarization P_{eg} and the occupation probabilities of the *electron system* in the state $|e\rangle \equiv |1_c 0_v\rangle$ and $|g\rangle \equiv |0_c 1_v\rangle$: n_e and n_g. Although n_c (n_v) incidently equals n_e (n_g) in the electron two-level system, the physical meaning is completely different and will result in errors in case that complicated many-level system is considered.

In this work, we strictly follow OBE formalism, discard the electron many-body effects. By this semiconductor OBE, we derive the polarization dependent FWM quantum beats in semiconductors without introducing any bi-exciton interaction effects. In this way we suggest that FWM quantum beats might be a purely coherent light-matter interaction effect, rather than due to something, such as electron-electron interaction process.

Modelling

In this work we do not consider the electron Coulomb interaction, we simply treat the semiconductor as a many-level atomic model with considering electron spin states. The ground state of the heavy-hole electron system, as well as those of the light-hole electron system, are described as

$$|hg\rangle = a^{\dagger}_{h1} a^{\dagger}_{h2} |0_h\rangle \, , |lg\rangle = a^{\dagger}_{l1} a^{\dagger}_{l2} |0_l\rangle \, . \tag{1}$$

Next, by Eq.(1) we derived the excited stats $|ex\rangle$ and $|ey\rangle$ of the semiconductor in case of x and y polarized light excitation, only resonant excitation are taken into account

$$\begin{aligned}
|ex\rangle &= \sum_{\theta,\phi} \Bigg\{ \Big[-a^{\dagger}_{c1} a^{\dagger}_{h2} bu - a^{\dagger}_{c1} a^{\dagger}_{h1} (wR_h - cu) + a^{\dagger}_{c2} a^{\dagger}_{h2} (uc^* - wR_h) + a^{\dagger}_{c2} a^{\dagger}_{h1} ub^* \Big] |0_h\rangle \\
&\quad + \Big[a^{\dagger}_{c1} a^{\dagger}_{l2} (uR_l + wc^*) + a^{\dagger}_{c1} a^{\dagger}_{l1} wb - a^{\dagger}_{c2} a^{\dagger}_{l2} wb^* + a^{\dagger}_{c2} a^{\dagger}_{l1} (wc + uR_l) \Big] |0_l\rangle \Bigg\} + c.c. \\
|ey\rangle &= \sum_{\theta,\phi} i \Bigg\{ \Big[-a^{\dagger}_{c1} a^{\dagger}_{h2} bu + a^{\dagger}_{c1} a^{\dagger}_{h1} (wR_h + cu) - a^{\dagger}_{c2} a^{\dagger}_{h2} (wR_h + uc^*) - a^{\dagger}_{c2} a^{\dagger}_{h1} ub^* \Big] |0_h\rangle \\
&\quad + \Big[a^{\dagger}_{c1} a^{\dagger}_{l2} (uR_h - wc^*) - a^{\dagger}_{c1} a^{\dagger}_{l1} wb - a^{\dagger}_{c2} a^{\dagger}_{l2} wb^* - a^{\dagger}_{c2} a^{\dagger}_{l1} (uR_l - wc) \Big] |0_l\rangle \Bigg\} + c.c.
\end{aligned} \tag{2}$$

with $u = -\sqrt{1/2}$ and $w = \sqrt{1/6}$; b, c, R_h and R_l are defined as in [9]. Here, the summation of θ and ϕ gives the total transition magnitude, or the full transition matrix, from electron valence state $|v\rangle$ to electron conduction state $|c\rangle$. From Eq.(2), one can find the excited states essentially are pairs of electrons in both conduction bands and valence bands but with difference phase.

The system Hamiltonian is

$$H_0 = \varepsilon_{ex} a^{\dagger}_{ex} a_{ex} + \varepsilon_{ey} a^{\dagger}_{ey} a_{ey} + \varepsilon_{hg} a^{\dagger}_{hg} a_{hg} + \varepsilon_{lg} a^{\dagger}_{lg} a_{lg} . \tag{3}$$

The energy ε_{ex}, ε_{ey}, ε_{hg} and ε_{lg} are defined for a single electron. For simplicity, we assume the energy $\varepsilon_{ex} = \varepsilon_{ey} = 0$, thereby ε_{hg} and ε_{lg} are the photon resonant energies but with minus sign. The states energy difference of $|hg\rangle$ and $|lg\rangle$ is ε_{lh} defined as $\varepsilon_{hg} - \varepsilon_{lg}$.

The dipole coupling to a laser field is described as

$$H_I = -\mathcal{E}(t)\left(a^{\dagger}_{ex}a_{hg}e^{i\alpha_1} + a^{\dagger}_{ey}a_{hg}e^{i\alpha_2} + a^{\dagger}_{ex}a_{lg}e^{i\beta_1} + a^{\dagger}_{ey}a_{lg}e^{i\beta_2} + h.c.\right). \qquad (4)$$

The modulus of the optical transitions between $|g\rangle \rightarrow |ex\rangle$ and $|g\rangle \rightarrow |ey\rangle$ are equal in both $|hg\rangle$ and $|lg\rangle$ cases, but the phase, α_1, α_2, β_1, and β_2 are different. We derived a relation between the phases of the optical transition matrixes,

$$\exp[i(\alpha_1 - \alpha_2)] = -\exp[i(\beta_1 - \beta_2)]. \qquad (5)$$

After some straightforward algebra, we obtain for the third-order nonlinear optical polarization, origin of the FWM signal. For polarization configurations of pump and probe parallel and orthogonal.

$$\begin{aligned}\mathcal{P}^{(3)}_{\parallel}(t) &= -4i\Theta(t)\Theta(-\tau)E_pE_pE_t\left(e^{-i(\varepsilon_h - i\gamma_2)t} + e^{-i(\varepsilon_l - i\gamma_2)t}\right)\left(e^{i(\varepsilon_h - i\gamma_2)\tau} + e^{i(\varepsilon_l - i\gamma_2)\tau}\right)\\ \mathcal{P}^{(3)}_{\perp}(t) &= -i\Theta(t)\Theta(-\tau)E_pE_pE_t\left(e^{-i(\varepsilon_h - i\gamma_2)t} + e^{-i(\varepsilon_l - i\gamma_2)t}\right)\Big(e^{i(\varepsilon_h - i\gamma_2)\tau}e^{i(\alpha_1-\alpha_2)} +\\ &\qquad e^{i(\varepsilon_l - i\gamma_2)\tau}e^{i(\beta_1-\beta_2)}\Big) \end{aligned} \qquad (6)$$

By the relation in Eq.(5), one can easily find that the two cases in Eq.(6) are different by a sign in the last term and different magnitudes with a factor 1/4, thereafter the ratio of the intensities of FWM signal in the two cases will be 1/16. In case of $\tau = 0$, $\mathcal{P}^{(3)}_{\parallel}$ is on the peak of the beats, but $\mathcal{P}^{(3)}_{\perp}$ on the bottoms of the beats. There is an exact phase difference π between the two cases as shown in Fig.(1).

The calculation of Fig.(1) are based on some parameters: the phase relaxation time $\gamma_2 = 4$ ps; the resonant optical transitions of $|hg\rangle \rightarrow |e\rangle$ is at wavelength 810 nm. The difference of the energy between $|hg\rangle$ and $|lg\rangle$ is 4.2 meV. The beats modulation depth is adjusted by a parameter f_{lh}. In the idea case, the optical transitions are equally taken place at $|hg\rangle$ and $|lg\rangle$, namely $f_{lh} = 1$, the modulation depth will between modulated 1 and 0. In this calculation, we adopt $f_{lh} = 0.75$ to avoid zero point. However, we do not attribute this unbalance to the heavy-hole and light-hole optical transition matrixes. [1, 2] As we have shown that the modulus of the optical transition matrixes are just equal. The reason might be the state-density dependent optical excitations of heavy-hole and light-hole bands. Also the detuning optical excitations and the summation of the optical transitions with different positions $\vec{k}$ could contribute to this by phase interference to mitigate the beats modulation depth. However it is not within the aim of this work in this stage.

Conclusion

In conclusion, we have re-examined the semiconductor Bloch equations (SBE) and found that, excluding the many-body electron Coulomb interaction, SBE do not reduce to the optical Bloch equations (OBE). We applied OBE on bulk semiconductors straightforward, neglecting electron-electron Coulomb interaction, and proposed an explanation for the polarization dependent four wave mixing (FWM) quantum beats, which could not be

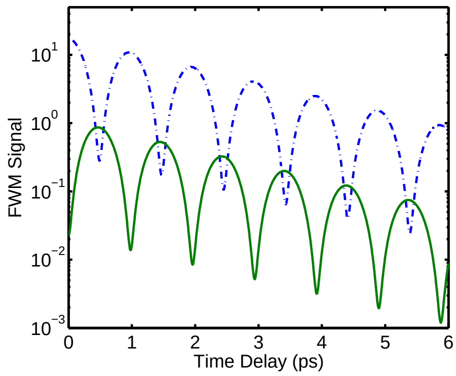

Figure 1: The FWM signal quantum beats as a function of pump-probe time delay. The dash-dotted and solid lines for parallel and orthogonal polarized pump-probe configurations.

explained by the conventional SBE but attributed to bi-exciton processes. We suggest that this effect might be simply a coherent process rather than due to electron-electron Coulomb interaction.

In case of a configuration with pump and probe in orthogonal polarization states, FWM can be generated due to the coherence transfer between the two excited states. In the investigates of polarization dependent FWM quantum beats, this coherence transfer could make the main role in the reproducing experimental results.

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