International Journal of Modern Physics B
Vol. 21, Nos. 8 & 9 (2007) 1621–1625

QUANTUM BEATS IN SEMICONDUCTORS

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Received 30 July 2006

In this paper we present a new method to calculate the Faraday rotation using the Jones calculus applicable to light pulses. This result is used to calculate the quantum beats in a magnetic semiconductor generated by pump-probe experiments in Faraday geometry. Considering a six level system and neglecting band dispersion as well as Coulomb interaction we find the Luttinger parameter of the valence band to be the only band parameter contributing to the quantum beat frequency.

Keywords: Faraday rotation; quantum beats; magnetic semiconductor.

1. Introduction

In today's solid state physics, ultrafast spectroscopy of optically excited semiconductors play an important role. One reason is that diluted magnetic semiconductors provide many parameters to control their properties making them an exellent tool for building up opto-opto and opto-electronical devices that will play an important role in the future of communication technology and spintronics[1,2,3]. Large progress in understanding the interaction and dimensional effects within these materials has been achieved in recent years theoretically[4,5,6,7] as well as experimentally[8,9].
Quantum beats are an exellent method to investigate energy level splitting in external fields. Thus they provide a powerful tool for studying the band parameters which determine the properties of magnetic semiconductors.

2. Faraday Rotation

The bulk semiconductor is illuminated by an electromagnetic wave, which produces an macroscopic polarization. For the electric field and the arising polarization we write $\mathbf{E}(z,t) = \underline{\mathbf{E}}(z,t) + \text{c.c.}$ and $\mathbf{P}(z,t) = \underline{\mathbf{P}}(z,t) + \text{c.c.}$. The propagation of the electric field is given by the reduced wave equation [10]

$$\underline{\mathbf{E}}(z,t) = \underline{\mathbf{E}}(0,t)\,\mathrm{e}^{\mathrm{i}kz} + \mathrm{i}\,\frac{kz}{2}\,\frac{1}{\varepsilon_0\,\varepsilon_\infty}\,\underline{\mathbf{P}}(0,t)\,. \tag{1}$$

In Jones-calculus[11] the position of an analyser is given by the Jones-matrix $\hat{\mathbf{T}}(\theta)$ with θ being the angle between the analysator and the x-axis. The Jones-vector $\mathbf{J}(z,t)$ describes the electric field before passing the analysator. Thus the intensity I of the light beam after the analyser yields

$$I(\theta,z) = \int_{-\infty}^{\infty} \mathrm{d}t\,\left|\hat{\mathbf{T}}(\theta)\mathbf{J}(z,t)\right|^2 = \int_{-\infty}^{\infty} \mathrm{d}t\left|\begin{pmatrix} \cos^2\theta & \sin\theta\cos\theta \\ \sin\theta\cos\theta & \sin^2\theta \end{pmatrix}\cdot\begin{pmatrix} E_x(z,t) \\ E_y(z,t) \end{pmatrix}\right|^2 . \tag{2}$$

The polarization plane of the light beam at a fixed z is given by the intensity maximazing angle $\theta_{\max}$. Thus the Faraday rotation Θ_{F} is given by the difference between the angle $\theta_{\max}$ at the starting point ($z=0$) and the end point ($z=z$). For thin samples the obtained expression for Θ_{F} can be linearized in z

$$\sin(2\Theta_{\mathrm{F}}) = \frac{z}{2}\,\frac{k}{\varepsilon_0\varepsilon_\infty}\,\frac{\mathbf{v}\times\mathbf{p}}{|\mathbf{v}|^2}\cdot\mathbf{e}_z\,, \tag{3}$$

with the vectors

$$\mathbf{v} = \begin{pmatrix} \langle \underline{E}_y(0)|\underline{E}_y(0)\rangle - \langle \underline{E}_x(0)|\underline{E}_x(0)\rangle \\ 2\mathrm{Re}[\langle \underline{E}_x(0)|\underline{E}_x(0)\rangle] \\ 0 \end{pmatrix}\,, \quad \mathbf{p} = \mathrm{Im}\begin{pmatrix} \langle \underline{P}_x(0)|\underline{E}_x(0)\rangle - \langle \underline{P}_y(0)|\underline{E}_y(0)\rangle \\ \langle \underline{E}_x(0)|\underline{P}_y(0)\rangle - \langle \underline{P}_x(0)|\underline{E}_y(0)\rangle \\ 0 \end{pmatrix}$$

using the abbreviation $\langle A(z)|B(z')\rangle = \langle A(z,t)|B(z',t)\rangle = \int_{-\infty}^{\infty}\mathrm{d}t\; A(z,t)B^*(z',t)$. Assuming small Faraday angles Θ_{F} and defining the normalized intensity of the incident beam $I = \langle \underline{E}_x(0)|\underline{E}_x(0)\rangle$ we finally arrive at

$$\Theta_{\mathrm{F}} = -\frac{z}{I}\,\frac{k}{4}\,\frac{1}{\varepsilon_0\varepsilon_\infty}\,\mathrm{Im}\int_{-\infty}^{+\infty} \underline{P}_{\mathrm{y}}(0,t)\underline{E}_{\mathrm{x}}^*(0,t)\,\mathrm{d}t\,. \tag{4}$$

3. Quantum Beats

For analytical investigation of quantum beats we apply the semiconductor Bloch equations [5] in the form

$$\mathrm{i}\hbar\,\frac{\partial}{\partial t}\,N(t) = [\,H(t)\,,\,N(t)\,]\,. \tag{5}$$

For further treatment we neglect the Coulomb interaction between electrons and holes as well as the band dispersion and apply the rotating wave approximation to the minimal coupling Hamiltonian. Changing into Dirac picture leads to the decomposition of the Hamiltonian [5] in the time independent part

$$H_0 = \begin{pmatrix} \frac{g\mu_{\mathrm{B}}}{\mu_0}\,\mathbf{B}\cdot\frac{\boldsymbol{\sigma}}{2} & 0 \\ 0 & -\frac{e\hbar}{m_0}\,\kappa\,\mathbf{J}\cdot\mathbf{B} \end{pmatrix} = \mathrm{diag}(H_{11}, H_{22}) \tag{6}$$

and the time dependent perturbation

$$W(t) = \begin{pmatrix} 0 & -\boldsymbol{\mu}_{\rm cv}\underline{\mathbf{E}}(t) \\ -\boldsymbol{\mu}_{\rm vc}\underline{\mathbf{E}}^*(t) & 0 \end{pmatrix}, \tag{7}$$

with the electric field $\underline{\mathbf{E}}(t) = \underline{\mathbf{E}}_{\rm Pu}(t) + \underline{\mathbf{E}}_{\rm Pr}(t)$ consisting of the pump $\underline{\mathbf{E}}_{\rm Pu}(t)$ and the probe beam $\underline{\mathbf{E}}_{\rm Pr}(t)$. For bound states the dipole matrix elements are connected with the momentum matrix elements via

$$\langle\, {\rm c}\,|\,\hat{\boldsymbol{\mu}}\,|\,{\rm v}\,\rangle = \frac{-e\,\hbar}{{\rm i}m_0 E_{\rm g}}\,\langle\, {\rm c}\,|\,\hat{\mathbf{p}}\,|\,{\rm v}\,\rangle \tag{8}$$

and thus exhibits the same symmetries. In Eq. (8) c denotes the conducting and v the valence band. The angular-momentum matrices $\boldsymbol{\sigma}$ and $\boldsymbol{J}$ as well as the elements of $\langle\, {\rm c}\,|\,\hat{\mathbf{p}}\,|\,{\rm v}\,\rangle$ are given by Glutsch[5]. Bohrs magneton is $\mu_{\rm B} = \mu_0\hbar e/(2m_0)$. Hence interband polarization is given by

$$\underline{\mathbf{P}}(t) = \frac{1}{\Omega}\sum_{m_1 m_2} \boldsymbol{\mu}_{{\rm v}m_2,{\rm c}m_1}\, n_{{\rm c}m_1,{\rm v}m_2}(t)\,, \tag{9}$$

where the symbol $\boldsymbol{\mu}_{\nu_1\nu_2}$ is a compact notation for the matrix $\boldsymbol{\mu}_{\nu_1 m_1,\nu_2 m_2}$ and $n_{\nu_1 m_1,\nu_2 m_2}$ are the off diagonal elements of the density matrix. We assume the external magnetic field and the polarization of light in the z direction (Faraday geometry) i.e., $\mathbf{B} = B\mathbf{e}_z$. Before illumination the semiconductor is assumed to be completely relaxated: $N^{(0)} = {\rm diag}(0,0,1,1,1,1)$. In the Dirac picture the semiconductor Bloch equation takes the form

$${\rm i}\hbar\,\frac{\partial}{\partial t}\,\tilde{N}(t) = [\,\tilde{H}(t)\,,\,\tilde{N}(t)\,]\,. \tag{10}$$

with $\tilde{H}(t) = U\,W(t)\,U^\dagger$ and $\tilde{N}(t) = U\,N(t)\,U^\dagger$ using the unperturbed time evolution operator $U = {\rm e}^{+\frac{\rm i}{\hbar}H_0 t}$. Interested in the Faraday rotation only, we filter out all components parallel to $\mathbf{k}_{\rm Pr}$ and proportional to the intensities of $\underline{\mathbf{E}}^*_{\rm Pu}\,\underline{\mathbf{E}}_{\rm Pu}$, i.e., terms proportional to $|\underline{\mathbf{E}}_{\rm Pu}|^2\underline{\mathbf{E}}_{\rm Pr}$. By solving Eq. (10) in third order perturbation theory and transforming back we may calculate the y-component of the polarization $\underline{P}_y(t)$. Assuming the propagation of light along the x-direction, i.e., $\underline{\mathbf{E}}_{\rm Pr}\,(t) = \underline{E}_{\rm Pr}\,(t)\mathbf{e}_x$ and $\underline{\mathbf{E}}_{\rm Pu}\,(t) = \underline{E}_{\rm Pu}\,(t)\mathbf{e}_x$ we finally arrive at

$$\begin{aligned} \underline{P}_y(t) = -\frac{\rm i}{\Omega\hbar^3}\int_{-\infty}^{t}{\rm d}t_3\int_{-\infty}^{+\infty}{\rm d}t_2\int_{-\infty}^{+\infty}{\rm d}t_1\;\underline{E}_{\rm Pr}(t_3)\Big[&\underline{E}^*_{\rm Pu}(t_2)\underline{E}_{\rm Pu}(t_1)A(t,t_1,t_2,t_3) \\ &+\underline{E}^*_{\rm Pu}(t_1)\underline{E}_{\rm Pu}(t_2)A(t,t_3,t_1,t_2)\Big]\,, \end{aligned} \tag{11}$$

where we used the abbreviations

$$A(t,t_1,t_2,t_3) = {\rm Tr}\big[M_y^\dagger {\rm e}^{\frac{\rm i}{\hbar}H_{11}(t_3-t)}M_x{\rm e}^{\frac{\rm i}{\hbar}H_{22}(t_2-t_3)}M_x^\dagger{\rm e}^{\frac{\rm i}{\hbar}H_{11}(t_1-t_2)}M_x{\rm e}^{\frac{\rm i}{\hbar}H_{22}(t-t_1)}\big] \tag{12}$$

as well as $M_x = \boldsymbol{\mu}_{\rm cv}\mathbf{e}_x$ and $M_y = \boldsymbol{\mu}_{\rm cv}\mathbf{e}_y$.

With the help of Eq. (4) the Faraday rotation in Faraday geometry yields

$$\Theta_{\mathrm{F}} = -\frac{z}{I}\frac{k}{4}\frac{1}{\varepsilon_0\varepsilon}\frac{1}{\Omega\hbar^3}\,\mathrm{Re}\int\limits_{-\infty}^{+\infty}\mathrm{d}t_4\int\limits_{-\infty}^{t_4}\mathrm{d}t_3\int\limits_{-\infty}^{+\infty}\mathrm{d}t_2\int\limits_{-\infty}^{+\infty}\mathrm{d}t_1\;\underline{E}^*_{\mathrm{Pr}}(t_4)\underline{E}_{\mathrm{Pr}}(t_3)$$
$$\times\left[\underline{E}^*_{\mathrm{Pu}}(t_2)\underline{E}_{\mathrm{Pu}}(t_1)A(t_4,t_1,t_2,t_3)+\underline{E}_{\mathrm{Pu}}(t_2)\underline{E}^*_{\mathrm{Pu}}(t_1)A(t_4,t_3,t_1,t_2)\right]. \tag{13}$$

For further calculations we assume nonoverlapping pump and probe pulses of the form $\underline{E}_{\mathrm{Pu}}(t)=\underline{E}^0_{\mathrm{Pu}}\,f(t)$ and $\underline{E}_{\mathrm{Pr}}(t)=\underline{E}^0_{\mathrm{Pr}}\,g(t)=\underline{E}^0_{\mathrm{Pr}}\,f(t-\tau)$ with the amplitude $\underline{E}^0_{\mathrm{Pu}}$, an equal envelope function $f(t)$ and the delay time τ between the pulses. With the help of the envelope function $f(t)$ we can define the following quantities

$$M^{\omega'} := \left|\int_{-\infty}^{+\infty} f(t)\mathrm{e}^{+\mathrm{i}\omega' t}\mathrm{d}t\right|^2 \qquad \text{and} \qquad M_\omega = \frac{1}{\pi}\mathcal{P}\int_{-\infty}^{+\infty}\frac{M^{\omega'}}{\omega-\omega'}\,\mathrm{d}\omega', \tag{14}$$

and for compact notation we introduce

$$M^{\omega'}_{\omega} := M^{\omega'}M_\omega - M^{-\omega'}M_{-\omega}\,. \tag{15}$$

Utilizing the above an simple expression for the quantum beating signal

$$\frac{\Theta_{\mathrm{F}}}{A} = G\sin\left(4\kappa\frac{\mu_B}{\mu_0}B\,\tau\right)+C \tag{16}$$

with the amplitude A and the normalized intensity of the probe pulse I

$$A = -\frac{z}{\hbar^3}\frac{k}{3\Omega\varepsilon\varepsilon_0}\frac{|\underline{E}^0_{\mathrm{Pr}}\underline{E}^0_{\mathrm{Pu}}|^2}{I}\left(\frac{\mu_B}{\mu_0}\frac{P}{E_{\mathrm{g}}}\right)^4, \quad I = |\underline{E}^0_{\mathrm{Pr}}|^2\int_{-\infty}^{+\infty}|g(t)|^2\,\mathrm{d}t \tag{17}$$

can be found, where P is the value of the nonzero momentum matrix elements[5]. In Eq. (16) we take advantage of the abbreviations

$$G = M^m M^n + M^{-m}M^{-n} \quad \text{and} \qquad C = 3M^n_n - \frac{M^m_m}{3} - \frac{M^n_m + M^m_n}{2}, \tag{18}$$

with the parameters m and n being combinations of the splitting of both bands:

$$m = \left(\frac{g}{2}-\kappa\right)\frac{\mu_B}{\mu_0}B \quad \text{and} \quad n = \left(\frac{g}{2}-3\kappa\right)\frac{\mu_B}{\mu_0}B\,. \tag{19}$$

To validate the theory numerical calculations have been performed by solving Eq.(5) using the leap frog algorithm[5] and assuming a gaussian shaped pulse with center frequency ω_s. The results of the comparison are given in Fig. 1.

4. Discussion

Providing a new method for calculating the Faraday rotation it is possible to derive the Faraday angle for a pump-probe experiment in Faraday geometry. This method is not based on the common averaged procedure and extends the theory to arbitrary light pulses. Analytical results compared with numerical solution of the Semiconductor Bloch equation are in excellent agreement as long as the pulses are seperated

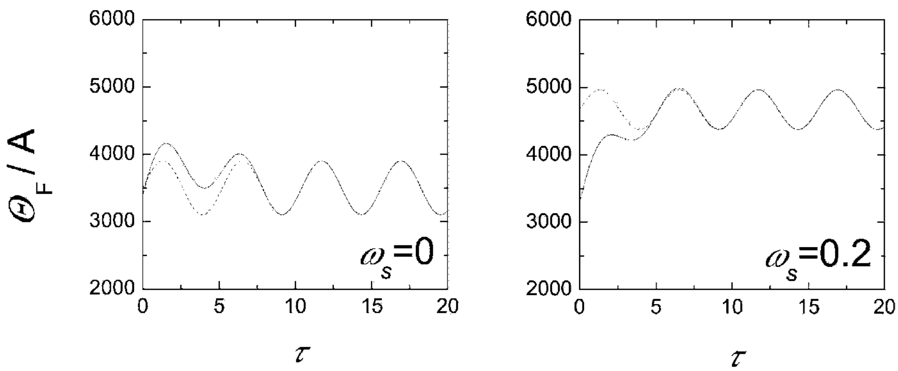

Fig. 1. Quantum beats for different offsets of the center frequency ω_s from the band gap in dependence on delay time τ. The solid line is a numerical solution and the dashed line is the analytical result of Eq.(16). The spectral bandwith of the gaussian envelope is $\sigma = 2$, the magnetic field yields $B = 0.5$ and the band parameters are $g = 0.467$ and $\kappa = 1.206$ (all given in excitonic units).

in time. As expected in this kind of four-wave mixing experiments the Faraday angle is independent of the intensity of the probe but linear in the intensity of the pump pulse. Calculating the Faraday angle for a six level system in the applied approximations it was possible to show that the Quantum beats frequency depends on the Luttinger parameter κ only. This is astonishing since one would expect both the valence as well as the conduction band to contribute. It is very likely that including band dispersion and Coulomb interaction would cause the Quantum beat frequency to depend on all parameters that characterizes conduction as well as valence band.

Acknowledgments

We thank Schering Stiftung and Deutsche Forschungsgemeinschaft (SFB 688) for financial support. The authors are indebted to K. Rönnburg and H. Roskos for participating stimulating discussions.

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