Polarization dependence of heavy- and light-hole quantum beats

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Simultaneous excitation of heavy- and light-hole quantum-well excitons with linearly polarized ultrashort laser pulses results in oscillating four-wave-mixing and pump-probe signals. These are elliptically polarized, except for exactly parallel and perpendicular incident polarizations, for which they are also linearly polarized. In the latter case, the heavy- and light-hole components are in phase or out of phase, respectively.

Recently, quantum beat measurements have been introduced as a tool in time-resolved nonlinear optical semiconductor spectroscopy.[1–8] In these experiments, an ultrashort spectrally broad laser pulse excites a nonstationary coherent wave packet of several optical transitions, the temporal evolution of which is subsequently monitored using time-resolved four-wave mixing[2–7] or pump-probe[7,8] techniques. For excitation of two optical transitions, one observes an oscillating modulation of the signal, with a time period determined by their energy splitting. In addition, the damping of the coherent oscillations contains explicit information on relaxation processes beyond that contained in "conventional" measurements.

The most simple theoretical description of these phenomena considers a three-level model consisting of the ground state and two excited states from which the wave packet is formed.[6,7] However, this approach ignores the details of the crystal structure and hence cannot describe the polarization dependence of the effect. In the present paper, we address this issue for the specific case of quantum beats of heavy-and light-hole excitations in semiconductor quantum well.[3,5,6] We consider a six-level model, describing $J=\frac{1}{2}$ and $\frac{3}{2}$ conduction and valence electrons, respectively, as studied previously in the context of the nonresonant optical Stark effect.[9–11]. Within the framework of this simple model and for linearly polarized ultrashort laser pulses, we show analytically that the resonant time-resolved third-order nonlinear optical response depends sensitively on the incident laser polarizations. (i) For parallel or perpendicular incident polarizations, the pump-probe and four-wave mixing signals are also linearly polarized (along the probe pulse), but heavy-and light-hole components are in phase or out of phase, respectively. (ii) For all other incident polarizations, the pump-probe and four-wave-mixing signals are elliptically polarized. Point (i) is also verified experimentally, but the data show in addition a strong polarization dependence of the dephasing rate which we are unable to explain at this time. Other experimental evidence for the latter effect has been presented previously.[12,13]

We consider a laser field $\mathbf{E}(t)$ propagating (nearly) along the growth (z) direction of a cubic semiconductor quantum-well structure, such as $GaAs/Al_xGa_{1-x}As$. Within the rotating-wave approximation, its dipole interaction with the $J=\frac{3}{2}$ valence (v) to $J=\frac{1}{2}$ conduction (c) electron transitions reads

$$H_{\mathrm{int}}=-[\mathbf{P}\cdot\mathbf{E}^*(t)+\mathbf{P}^\dagger\cdot\mathbf{E}(t)]\ , \tag{1}$$

where the polarization operator $\mathbf{P}$ obeys the usual circular selection rules,[14] which are included in the dipole matrix elements $\boldsymbol{\mu}_{nm}=<c,n|e\mathbf{r}|v,m>$,

$$\mathbf{P}=\sum_{n,m} v_m^\dagger c_n \boldsymbol{\mu}_{nm}^*\ . \tag{2}$$

Here c_n destroys a conduction electron with moment $n=\pm\frac{1}{2}$ and $v_m^\dagger$ creates a valence electron with moment $m=\pm\frac{3}{2}$ (heavy hole, h) or $m=\pm\frac{1}{2}$ (light hole, l). Introducing the Rabi frequency $R_{mn}(t)=\boldsymbol{\mu}_{nm}\cdot\mathbf{E}(t)$, the optical Bloch equations for the polarization $p_{mn}=<v_m^\dagger c_n>$, conduction electron population $c_{nn'}=<c_n^\dagger c_{n'}>$, and valence electron population $v_{mm'}=<v_m^\dagger v_{m'}>$ read

$$\partial_t p_{mn} + i(\varepsilon_n^c - \varepsilon_m^v)P_{mn} + \gamma_{nm}^{cv} P_{mn} = i\sum_{m'} v_{mm'} R_{m'n}(t) - i\sum_{n'} R_{mn'}(t) c_{n'n}$$

$$\partial_t v_{mm'} + (i\varepsilon_{m'}^v - i\varepsilon_m^v + \gamma_{m'm}^{vv})(v_{mm'} - \delta_{mm'}) = i\sum_n [p_{mn} R_{m'n}^*(t) - R_{mn}(t) p_{m'n}^*] \quad (3)$$

$$\partial_t c_{n'n} + i(\varepsilon_n^c - \varepsilon_{n'}^c)c_{n'n} + \gamma_{nn'}^{cc} c_{n'n} = -i\sum_m [R_{nm'}^*(t) p_{mn} - p_{mn'}^* R_{mn}(t)] .$$

Here, $\varepsilon_{n(m)}^{c(v)}$ are the conduction (valence) electron energies and we have added phenomenological relaxation rates γ to the equations.

In order to describe time-resolved two-pulse experiments, we split the laser field into two parts,

$$\mathbf{E}(t) = \mathbf{E}_{\mathbf{k}_1}(t) e^{i\mathbf{k}_1\cdot\mathbf{r}} + \mathbf{E}_{\mathbf{k}_2}(t) e^{i\mathbf{k}_2\cdot\mathbf{r}} , \quad (4)$$

and calculate the third-order optical polarization $\mathbf{P}^{(3)}(t) = \sum_{nm} \boldsymbol{\mu}_{nm}^* p_{mn}^{(3)}$ radiating into the directions $\mathbf{k}_1$ and $2\mathbf{k}_2 - \mathbf{k}_1$, assuming that pulse 1 is weaker than pulse 2. The former $\mathbf{P}_{\mathbf{k}_1}^{(3)}(t)$ describes pump-probe (PP) experiments, i.e., the modification of the linear transmission of pulse 1 because of pulse 2, while the latter, $\mathbf{P}_{2\mathbf{k}_2-\mathbf{k}_1}^{(3)}(t)$, describes four-wave-mixing (FWM) experiments, i.e., the diffraction of pulse 2 off the transient grating created by interference with pulse 1. For an optically thin sample and a slow detector, the respective signals are

$$I_{\mathrm{PP}} \propto -\,\mathrm{Im} \int dt\, \mathbf{P}_{\mathbf{k}_1}^{(3)}(t) \cdot \mathbf{E}_{\mathbf{k}_1}^*(t) , \quad (5)$$

$$I_{\mathrm{FWM}} \propto \int dt\, |\mathbf{P}_{2\mathbf{k}_2-\mathbf{k}_1}^{(3)}(t)|^2 . \quad (6)$$

It is instructive to evaluate the above equations in the ultrashort-pulse limit $\mathbf{E}_{\mathbf{k}_1}(t) = \mathbf{E}_{\mathbf{k}_1}\delta(t-\tau)$ and $\mathbf{E}_{\mathbf{k}_2}(t) = \mathbf{E}_{\mathbf{k}_2}\delta(t)$, where τ is the time delay between the incident pulses. After some straightforward algebra, we obtain for the third-order nonlinear optical polarization

$$\mathbf{P}_{\mathbf{k}_1}^{(3)}(t) = -i\Theta(t-\tau)\Theta(\tau) \sum_{n,mn',m'} \boldsymbol{\mu}_{nm}^* e^{-i(\varepsilon_n^c - \varepsilon_m^v - i\gamma_{nm}^{cv})(t-\tau)} \Big[(\boldsymbol{\mu}_{nm'} \cdot \mathbf{E}_{\mathbf{k}_1}) e^{-i(\varepsilon_{m'}^v - \varepsilon_m^v - i\gamma_{m'm}^{vv})\tau} (\boldsymbol{\mu}_{n'm} \cdot \mathbf{E}_{\mathbf{k}_2}) + (\boldsymbol{\mu}_{n'm} \cdot \mathbf{E}_{\mathbf{k}_1}) e^{-i(\varepsilon_n^c - \varepsilon_{n'}^c - i\gamma_{nn'}^{cc})\tau} (\boldsymbol{\mu}_{nm'} \cdot \mathbf{E}_{\mathbf{k}_2}) \Big] (\boldsymbol{\mu}_{n'm'} \cdot \mathbf{E}_{\mathbf{k}_2})^* , \quad (7)$$

$$\mathbf{P}_{2\mathbf{k}_2-\mathbf{k}_1}^{(3)}(t) = -i\Theta(t)\Theta(-\tau) \sum_{n,m,n',m'} \boldsymbol{\mu}_{nm} e^{-i(\varepsilon_n^c - \varepsilon_m^v - i\gamma_{nm}^{cv})t} (\boldsymbol{\mu}_{nm'} \cdot \mathbf{E}_{\mathbf{k}_2})(\boldsymbol{\mu}_{n'm} \cdot \mathbf{E}_{\mathbf{k}_2}) e^{i(\varepsilon_{n'}^c - \varepsilon_{m'}^v + i\gamma_{n'm'}^{cv})|\tau|} (\boldsymbol{\mu}_{n'm'} \cdot \mathbf{E}_{\mathbf{k}_1})^* . \quad (8)$$

These expression are written in a time-ordered fashion. In the case of PP ($\tau > 0$), pulse 2 acts first (at time 0) and generates valence and conduction electron populations which propagate for a time interval τ until pulse 1 arrives (at time τ). In the case of FWM ($\tau < 0$), pulse 1 acts first (at time - $|\tau|$) and generates a polarization which propagates for a time interval $|\tau|$ until pulse 2 arrives (at time 0).

To proceed, we assume (without loss of generality) that pulse 2 is linearly polarized along the x direction ($\varphi = 0$), while the polarization direction of pulse 1 is rotated by an angle φ. We also adopt the usual 3:1 ratio for *c-h* to *c-l* transition probabilities.[14] Denoting the dipole matrix element for *c-h* transition by μ, we find for the PP and FWM polarizations

$$\mathbf{P}_{\mathbf{k}_1}^{(3)}(t) = \frac{-i}{2}\Theta(t-\tau)\Theta(\tau)|\mu|^4 |E_{\mathbf{k}_2}|^2 E_{\mathbf{k}_1} \left[\begin{pmatrix} \cos\varphi \\ \sin\varphi \\ 0 \end{pmatrix} \left[\tfrac{4}{3} e^{-\gamma^{cc}\tau} \left(e^{-i(\varepsilon^c - \varepsilon_h^v - i\gamma_h^{cv})(t-\tau)} + \tfrac{1}{3} e^{-i(\varepsilon^c - \varepsilon_l^v - i\gamma_l^{cv})(t-\tau)} \right) + e^{-\gamma_{hh}^{vv}\tau} e^{-i(\varepsilon^c - \varepsilon_h^v - i\gamma_h^{cv})(t-\tau)} + \tfrac{1}{9} e^{-\gamma_{ll}^{vv}\tau} e^{-i(\varepsilon^c - \varepsilon_l^v - i\gamma_l^{cv})(t-\tau)} \right] + \begin{pmatrix} \cos\varphi \\ -\sin\varphi \\ 0 \end{pmatrix} \left(\tfrac{1}{3} e^{-i(\varepsilon_h^v - \varepsilon_l^v - i\gamma_{hl}^{vv})\tau} e^{-i(\varepsilon^c - \varepsilon_l^v - i\gamma_l^{cv})(t-\tau)} + \tfrac{1}{3} e^{-i(\varepsilon_l^v - \varepsilon_h^v - i\gamma_{hl}^{vv})\tau} e^{-i(\varepsilon^c - \varepsilon_h^v - i\gamma_h^{cv})(t-\tau)} \right) \right] \quad (9)$$

$$\mathbf{P}_{2\mathbf{k}_2-\mathbf{k}_1}^{(3)}(t) = \frac{-i}{2}\Theta(t)\Theta(-\tau)|\mu|^4 E_{\mathbf{k}_2}^2 E_{\mathbf{k}_1}^* \left[\begin{pmatrix} \cos\varphi \\ -\sin\varphi \\ 0 \end{pmatrix} \left(e^{-\gamma_h^{cv}(t-\tau)} e^{-i(\varepsilon^c - e_h^v)(t+\tau)} + \tfrac{1}{9} e^{-\gamma_l^{cv}(t-\tau)} e^{-i(\varepsilon^c - \varepsilon_l^v)(t+\tau)} \right) + \begin{pmatrix} \cos\varphi \\ \sin\varphi \\ 0 \end{pmatrix} \left(\tfrac{1}{3} e^{-i(\varepsilon^c - \varepsilon_h^v - i\gamma_h^{cv})t} e^{-i(\varepsilon^c - \varepsilon_l^v + i\gamma_l^{cv})\tau} + \tfrac{1}{3} e^{-i(\varepsilon^c - \varepsilon_l^v - i\gamma_l^{cv})t} e^{-i(\varepsilon^c - \varepsilon_h^v + i\gamma_h^{cv})\tau} \right) \right] . \quad (10)$$

These expressions can now be substituted into Eqs. (5) and (6) to (analytically) obtain the detected PP and FWM signals in the ultrashort-pulse limit.

In Eqs. (9) and (10), two types of terms appear: Those that are polarized along pulse 1, with angle φ relative to pulse 2, and those that have polarization $-\varphi$. Both evolve differently in time, so that, in general, the signals are elliptically polarized. The experimentally detected signal depends then sensitively on whether one places a polarizer behind the sample or not. In the following, we will assume that this is the case for PP measurements (with polarizer along probe pulse 1), but not for FWM measurements, as already implied by Eqs. (5) and (6).

For parallel and perpendicular incident polarizations, the induced PP polarization is polarized parallel to probe pulse 1, and the two terms in Eq. (9) add in phase or out of phase, respectively. This translates into a phase difference of π for the respective PP quantum beats and is illustrated in Fig. 1 for the following dephasing rates: $\gamma^{cc\,-1}=\gamma_{hh}^{vv\,-1}=\gamma_{ll}^{vv\,-1}=20\,\text{ps}, \gamma_{h}^{cv\,-1}=\gamma_{l}^{cv\,-1}=\gamma_{hl}^{vv\,-1}=6$ ps. For an angle of 45° between the incident polarizations, only the first term in Eq. (9) contributes to the PP signal and quantum beats are absent.

For parallel and perpendicular incident polarizations, much the same behavior is found for the FWM signal, as illustrated in Fig. 2 for the same parameters. The only difference is that the induced FWM polarization for perpendicular incident polarizations is now polarized antiparallel to pulse 1. This behavior also arises in the case of selective c-h or c-l excitation and is well known from previous studies of impurity transitions in solids.[15] It is a direct consequence of circular selection rules.

We have also experimentally investigated the polarization dependence of h-l quantum beats in a ten-period 170-Å GaAs/Al_xGa_{1-x}As quantum-well sample using time-resolved four-wave mixing. The laser source is a 100-fs Kerr-lens mode-locked Ti-sapphire laser. The incident pulses are tuned between the first c-h and c-l transitions, which are split by about 4.2 meV. The spectral width of the laser pulses is about 20 meV. Excitation density ($< 10^9$ cm^{-2}) and lattice temperature ($T_L \approx 12.5$ K) are kept low to avoid fast dephasing due to exciton-exciton and exciton-phonon scattering.[16] Figure 3 shows the experimental FWM results for parallel (solid line) and

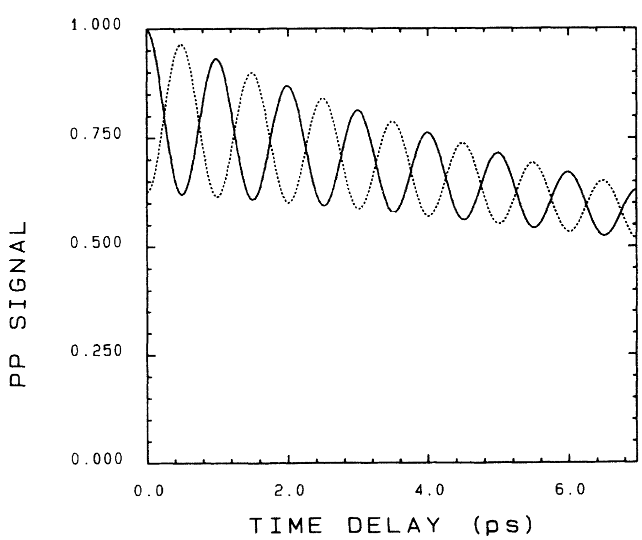

FIG. 1. Pump-probe signal in the ultrashort-pulse limit for parallel (solid) and perpendicular (dashed) polarizations.

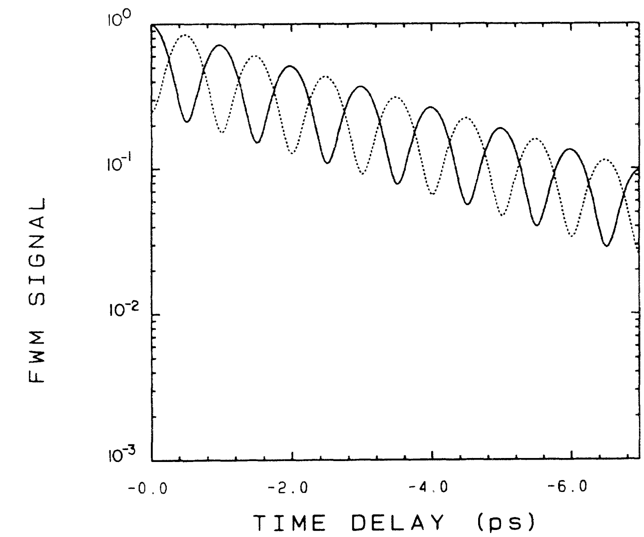

FIG. 2. Four-wave-mixing signal in the ultrashort-pulse limit for parallel (solid) and perpendicular (dashed) polarizations.

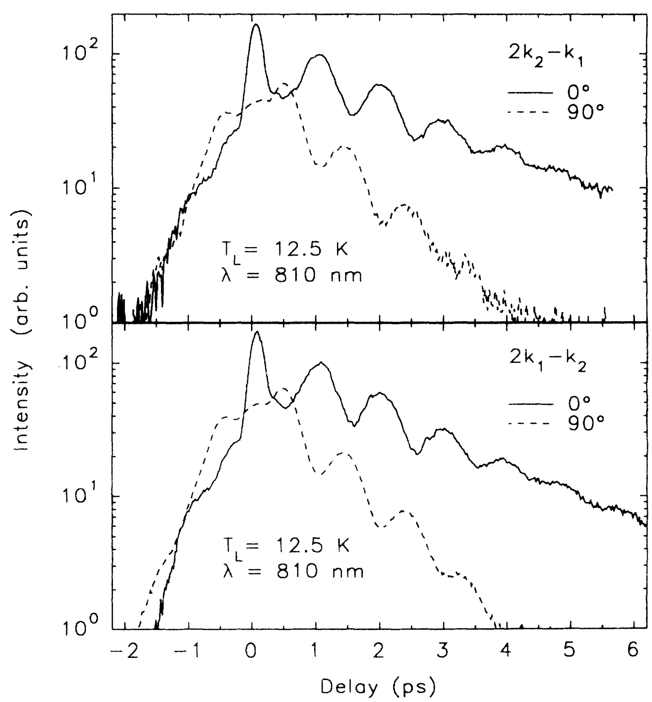

FIG. 3. Experimental four-wave-mixing signal in GaAs/Al_xGa_{1-x}As for parallel (solid) and perpendicular (dashed) polarizations.

perpendicular (dashed line) incident polarizations, measured both in the direction $2\mathbf{k}_2-\mathbf{k}_1$ (upper part) and $2\mathbf{k}_1-\mathbf{k}_2$ (lower part). All traces show pronounced quantum beats with a period of about 950 fs, in good agreement with the *h-l* splitting. The quantum beats for parallel and perpendicular polarizations have a phase difference of π, as predicted by Eq. (10). The experimental data also confirm the prediction that the signal for parallel polarizations has its maximum near zero delay. The decay time of the FWM signal is much slower for parallel than for perpendicular polarizations, as reported previously for the decay of *c-h* FWM signal.[12,13] This effect is not understood at present, but appears to be sample dependent. Also note that the peak intensities are larger for parallel polarizations, consistent with the slower dephasing.

Note that there is also a pronounced rising FWM signal for positive delay (i.e., pulse 2 preceding pulse 1) which has been predicted[17] and observed[5,6,18–20] previously. This rising FWM signal is a consequence of exciton-exciton interactions and not described by our simplified model. For perpendicular polarizations, the rise and decay times are close to the predicted ratio 1:2.[17] For parallel polarizations, however, the ratio is much larger, about 1:5. Again, at present, we do not have an explanation for this effect.

In conclusion, we have studied heavy-hole–light-hole quantum beats in the third-order nonlinear optical response of semiconductor quantum wells. Our theoretical and experimental results clearly demonstrate a pronounced dependence on incident laser polarization. Part of this can be readily accounted for by the symmetry of the conduction and valence Bloch states. However, the data reveal additional polarization dependencies, in particular, of the dephasing rate, which are not understood at present. Since, for example, the four-wave-mixing signal intensity decreases with dephasing, it appears entirely possible that, in samples with very fast dephasing for perpendicular incident polarizations, no significant signal can be detected at all.[21]

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[21]J. Kuhl *et al.* (private communication) have also studied the polarization dependence of *h-l* quantum beats using backward FWM and different samples. Contrary to the results reported here, they find results characteristic of an isotropic medium, i.e., the signal simply vanishes as the polarization angle of the incident pulses is turned from 0° to 90°.