Polarization dependence of beating phenomena at the energetically lowest exciton transition in GaAs quantum wells

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We report time-resolved degenerate four-wave-mixing experiments on a high-quality single GaAs quantum well. The signals for parallel and cross-polarized exciting pulses show pronounced quantum beats with different frequencies corresponding to the heavy/light-hole splitting and the biexciton binding energy, respectively. Comparison of the experimental data with solutions of the optical Bloch equations for a phenomenological model system shows that these features result from strong contributions of the biexcitonic state. The importance of biexcitons to the nonlinear coherent response of the two-dimensional exciton is further confirmed by the frequency dependence of the time-integrated four-wave-mixing signal amplitudes measured for the two polarization geometries.

I. INTRODUCTION

Theoretical modeling of light-hole (lh) and heavy-hole (hh) excitons in GaAs quantum wells by two independent three-level systems with opposite circular polarization selection rules[1] leads to serious disagreement with many experimental results, which have been observed in polarization-dependent time-integrated (TI) and time-resolved (TR) degenerate four-wave-mixing (DFWM) experiments. For example, changes in the polarization angle Θ between two linearly polarized excitation pulses strongly affect the DFWM signal amplitude and polarization,[2] as well as the dephasing time and the phase of quantum beats.[3–5] Furthermore, a shift of the DFWM signal emission time from $t = 2\tau$ to $t = \tau$ for inhomogeneously broadened transitions has been observed if the polarization of the exciting pulses is switched from parallel to cross-polarized fields.[3,5,6]

In several papers,[3,7–10] it has been shown that most of these discrepancies can be resolved if coupling between σ^+ and σ^- excitons via Coulomb interaction and sample disorder are taken into account. This coupling has been analyzed in the framework of discrete multilevel systems. Solutions of the optical Bloch equations with phenomenologically included terms for the various interaction mechanisms reveal the influence of these mechanisms on TI- and TR-DFWM signals. Coulomb interaction between excitons implies an increase in the phase relaxation rates of the excitonic states, with increasing exciton density (excitation induced dephasing, EID),[11] exchange[12] or local field effects (LFE),[13] and may even lead to the formation of excitonic molecules (biexcitons). Although evidence for biexciton formation (BIF) has been seen in many experiments, e.g., density dependent photoluminescence (PL) measurements,[14,15] pump-probe studies[16,17] and signal modulations observed in TI-DFWM experiments,[18] its importance for the nonlinear coherent optical response of 2D excitons remains controversial.

Recently, we have suggested a different three pulse TI-DFWM configuration, which allows the identification of features characteristic of EID, LFE, and BIF contributions to the nonlinear response.[8] Application of this technique to a high quality GaAs quantum well (QW) revealed that the combination of LFE and BIF is able to explain the polarization dependence of the amplitude and temporal profile of the TI-DFWM signal. It should be noted that the dependence of the signal amplitude and polarization on the polarization angle Θ has also been explained within a full many-body theory, where the symmetry properties of the Luttinger Hamiltonian are discussed for a cubic system.[19]

In this paper, we present polarization-dependent DFWM measurements using a two-pulse, self-diffraction geometry on a homogeneously broadened GaAs quantum well. Systematic studies of the polarization and frequency dependence of the TI- and TR-DFWM signals reveal the contributions of hh and lh excitons, as well as their respective bound and free two-exciton states to the

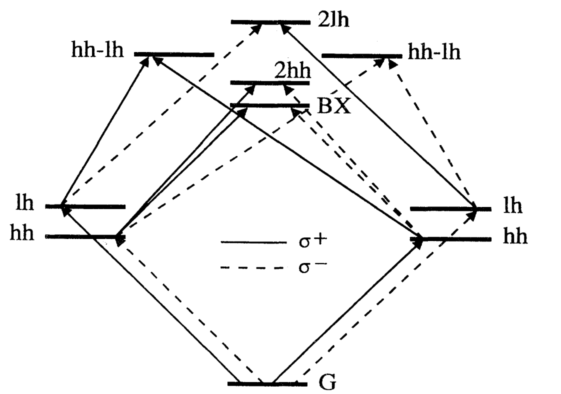

FIG. 1. Ten-level scheme for circularly polarized lh and hh exciton transitions consisting of one ground, four single-exciton, and five two-exciton states including the bound two heavy-hole exciton state BX (biexciton).

coherent nonlinear optical response of the lowest exciton transition of a QW.

These experiments exhibit polarization-dependent features which, so far, to our knowledge, have not been reported: (i) two remarkably different sets of quantum beats are observed in the TR-DFWM signal, corresponding to the hh-lh-exciton energy splitting and the biexciton binding energy, for parallel and cross polarized excitation with 120 fs pulses, respectively; (ii) different beat frequencies for positive and negative time delays in TR-DFWM for parallel polarization; (iii) distinctly longer decay time of the TI signal for negative delays in the case of cross-polarized (CP) as compared to parallel polarized (PP) pulses. In earlier analyses, the experimental data were compared to theoretical calculations based on phenomenological multilevel systems, which included Coulomb interactions.[7,8] For the interpretation of the present experiment, this model has been extended to a ten-level scheme (see Fig. 1), which takes into account third-order processes. Comparisons between experimental results and theoretical calculations prove that many of the surprising observations can be explained as biexcitonic contributions to the nonlinear optical response.

Additionally, we have found that the ratio of the TI signal amplitudes for the CP and PP configurations decreases strongly if the laser frequency is tuned from the low energy side of the hh-exciton line to the high energy side of lh line. This demonstrates that biexciton states are important for the case of two hh excitons, whereas contributions from the remaining two-exciton states, consisting of a hh and a lh exciton or two lh excitons, play a less significant role.

II. EXPERIMENT

All experiments were performed on a high quality 20 nm $GaAs/Al_{0.3}Ga_{0.7}As$ single quantum well, which was mounted in a continuous-flow helium cryostat and maintained at 8 K. For sample characterization, PL and PL excitation (PLE) spectra were measured with a tunable cw dye laser. The hh exciton at 1.5252 eV has a PL linewidth of 0.3 meV and shows no measurable Stokes shift between the PL and PLE spectra. The energy of the lh exciton is 1.5290 eV. A feature in the PL at 1.2 meV below the hh exciton line was identified as a biexciton (BX) by intensity dependent PL measurements.

The excitation pulses for the DFWM experiments were generated by a mode-locked Ti:sapphire laser operating at a repetition rate of 76 MHz and providing transform limited pulses as short as 120 fs. In order to measure the frequency dependence of the DFWM signal, the pulse duration was increased to 720 fs by external spectral filtering. The polarization of the two input pulses was adjusted by Glan-Thompson polarizers and $\lambda/2$ plates providing polarization definition of better than 200:1. The DFWM signal with $\mathbf{k}_s = 2\mathbf{k}_2 - \mathbf{k}_1$ was detected in the self-diffraction backward (reflection) geometry. The diffracted signal was time-resolved via a cross correlation, with a 120 fs reference pulse in a 2 mm thick $LiIO_3$ crystal.

In order to record the DFWM signals, we used CP and PP incident laser pulses at different excitation energies and pulse durations, leading to different exciton densities. In one case, we used a narrow band laser pulse ($\Delta\omega_L = 3$ meV) corresponding to a pulse duration of 720 fs, which was tuned either to 1.5276 eV (denoted N_1, between lh and hh exciton) or to 1.5245 eV (N_2, between hh exciton and biexciton), resulting in an exciton density of 8×10^8 cm^{-2}. In the other case, we used a broadband laser pulse (B, $\Delta\omega_L = 16$ meV) corresponding to a pulse duration of 120 fs, which was tuned to 1.5270 eV resulting in an exciton density of 3×10^8 cm^{-2}.

The TI-DFWM signals recorded for PP and CP incident laser pulses at different excitation energies and pulse durations are depicted in Figs. 2(a) and 2(b), respectively. In the case of excitation condition N_1, a distinct hh/lh beating is observed at positive time delay, which is much stronger in the case of PP than in the case of CP excitation. In particular, the signal at negative time delay decays with the same time constant of 1 ps in both cases. After tuning the laser between the hh exciton and the biexciton (i.e., excitation condition N_2), the hh/lh beating disappears and a weak 2hh-exciton/biexciton beating is observed. The most important feature is the difference in the decay times at negative delay. At PP excitation, the signal at negative time delay decays with a time constant of 1 ps and at CP excitation the decay time is 2.3 ps. This surprising difference in the time constants is also detected using the excitation condition B. Additionally, the PP and CP signals show both lh/hh and 2hh/biexciton beating at positive delay. However, as observed for N_1 excitation, a reduction of the lh/hh beating amplitude is observed for CP excitation. The observed beating clearly indicates the influence of the biexciton under both broadband excitation conditions (B), as well as narrow band excitation (N_2) in the vicinity of the biexciton transition. Similar differences in the slope of the signals at negative delays have also been reported by Wang et al.[17]

The effect of the slightly higher exciton density used in $N_{1,2}$ as compared to B is also observed in the decay times at positive delay in Figs. 2(a) and 2(b). The time

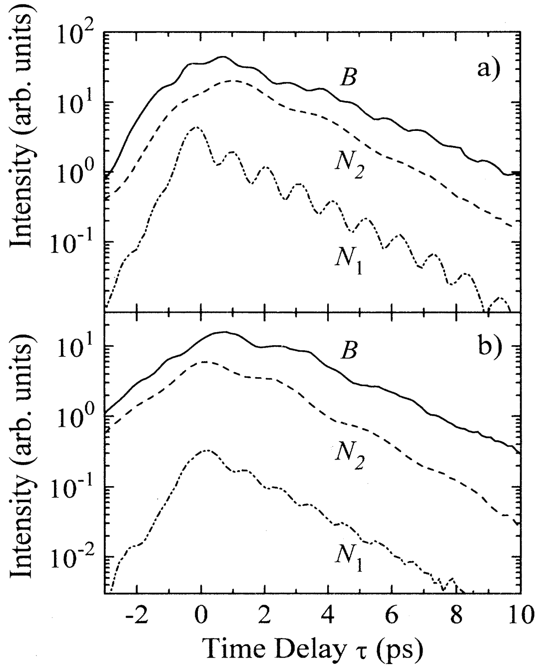

FIG. 2. Time-integrated DFWM intensity versus delay τ for (a) parallel and (b) cross-polarized laser fields. Solid line: excitation B (broadband, between lh and hh); dashed line: excitation N_2 (narrow band, between hh and BX); dotted line: excitation N_1 (narrow band, between lh and hh).

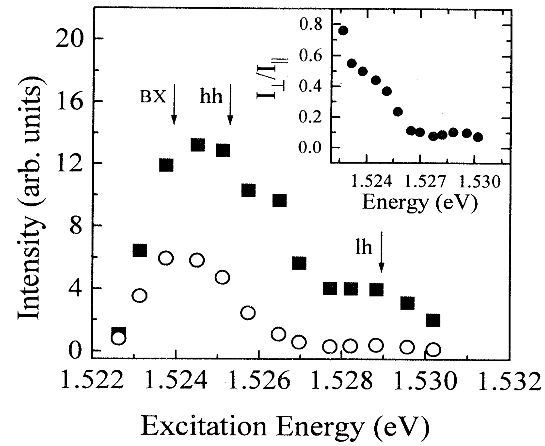

FIG. 3. Peaks of the TI-DFWM signals for PP (closed squares) and CP (open circles) polarizations versus excitation energy. Inset: The ratio of the PP and CP TI-DFWM intensity for different excitation energies (probe delay $\tau = 0$).

constants in N_1 and N_2 are the same, but are slightly faster than in B.

In Fig. 3, the TI-DFWM signal intensity at delay $\tau = 0$ is plotted versus the laser photon energy for PP (closed squares) and CP (open circles) polarization. The measurements were performed under the narrow band excitation condition with an excitation intensity, such that the T_2 time remained nearly constant over the measured range (i.e., in the low density limit). Whereas no shift in the maximum of the light-hole peaks is observed when going from PP to CP polarization, the maximum signal near the hh line is shifted from the hh energy (observed for PP polarization) to the biexciton PL energy. This observed shift indicates that for CP polarization, the response after exciting near the hh line is dominated by the hh biexciton. Since no shift in the light-hole maximum is observed, we conclude that no bound lh two-exciton states exist. This conclusion is further supported by the inset of Fig. 3, where the ratio of the peak signal amplitudes for both polarization configurations are plotted versus photon energy. The curve reaches its maximum value near the position of the biexciton line and decreases rapidly if the laser is tuned across the center of the hh-exciton transition towards the lh-exciton line. This again indicates that the CP signal amplitudes do not differ from the PP amplitudes by a constant factor, but that the CP signal is strongly peaked at the biexciton PL energy.

The PP TI-DFWM signal under excitation condition N_2 [dashed line in Fig. 2(a)] is modulated at a frequency corresponding to the biexciton binding energy. Spectral analysis of the TI-DFWM signal (Fig. 4) shows no phase shift in the modulation when the detection energy is tuned through the biexciton resonance ($\delta = 0$), thus the modulation is due to quantum beating rather than polarization interference.[20,21] It should be noted that spectrally resolving the signal allows observation of the weaker, heavily modulated biexciton contribution since the strong influence of the hh exciton can be excluded.

The result of the TR-DFWM measurements under excitation condition B are presented in the three-dimensional plots of Figs. 5(a) and 5(b). The intensity of the diffracted signal is plotted versus the real time t of the reference pulse and for several fixed delays τ between the two excitation pulses. The two polarization configurations create completely different TR-DFWM signals with rich quantum beat structures. The diffracted signal appears at the same time as the second excitation pulse ($t = \tau$) independent of the delay time τ between the two excitation pulses. This indicates free polarization decay behavior as is expected for a homogeneously broadened transition. The decay time of the PP signal relative to the real time t corresponds to a dephasing time

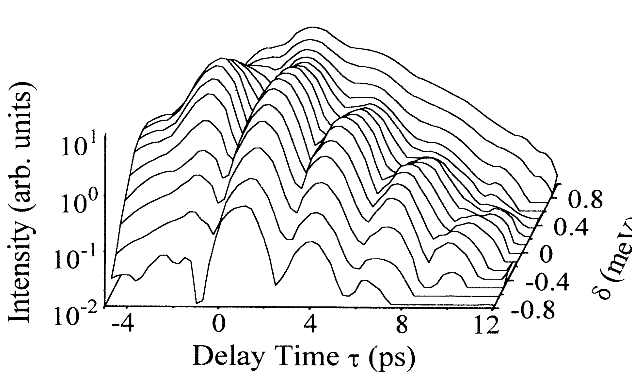

FIG. 4. Spectrally resolved DFWM signal. The detuning δ corresponds to the difference between the detection energy and the biexciton resonance.

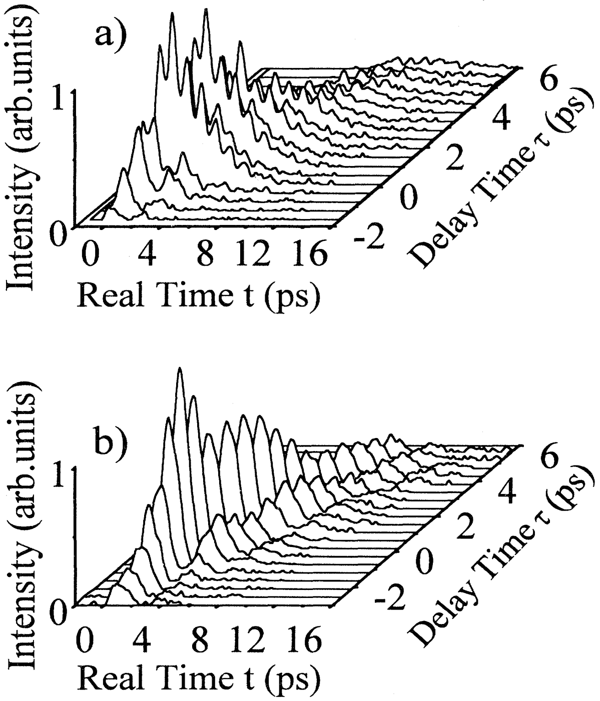

FIG. 5. Measured intensity of the TR-DFWM signal at various probe delays for (a) parallel and (b) cross-polarized laser fields.

of $T_2 = 4.3$ ps, whereas the CP signal gives a dephasing time of $T_2 = 3.7$ ps. Surprisingly, the beat frequencies at positive delays are quite different for the PP and CP configuration. For the PP configuration, the beat period observed for negative delays is much larger than that occurring at positive delays. Additionally, the modulation of the CP signal is quite stronger than that of the PP signal. Analysis of the signals reveals that the beating on top of the PP signal for $\tau \geq 0$ corresponds to the hh/lh-exciton splitting, whereas the beat period for $\tau \leq 0$ is equal to that of the CP signal and corresponds to the biexciton binding energy.

III. THEORETICAL MODELING AND DISCUSSION

The calculation of the DFWM signal is based on a multilevel scheme, in which excitonic and biexcitonic states are taken into account. In previous publications, we have shown that an interacting heavy/light-hole exciton system can be described using a nine-level scheme[7,22] and that biexcitons can be considered by introducing an additional two-exciton state.[8] The discussion belonging to Fig. 3 showed that in the GaAs quantum well, the observed biexciton consists of two hh excitons and that lh excitons do not build biexcitons. Therefore, we introduce only one additional biexcitonic state so that the calculations are based on the ten-level scheme depicted in Fig. 1. The ten-level scheme consists of one ground state (G), four single-exciton states (lh, hh with different selection rules σ^+ and σ^-), and five two-exciton states, which represent either a BX or scattering states (2hh, 2lh, hh-lh). Within this paper, a scattering state represents the energetically lowest state of a continuum of states. All calculations presented here are based on that ten-level system, which is mostly phenomenological, but includes all of the symmetry properties obtained from an exact theory.[19]

The optical Bloch equations for the ten-level scheme were numerically solved to third order in the electric field using the rotating wave approximation. Local field effects, which have an important influence on the DFWM signal, have been considered by introducing a local field parameter L.[8,13,23] All optical transitions are phenomenologically described using optical matrix elements (selection rules) and dephasing times. In the following, we will show that many of these dephasing times exhibit a strong influence on the signal, which enables us to take most of the parameters from experimental data and to use only some of them as fit parameters.

The oscillator strengths of the hh two-exciton state and the biexciton state have to be distributed since the simultaneous creation of a σ^+ and a σ^- hh exciton leads either to a biexciton or to unbound excitons in a two-particle scattering state (2hh in Fig. 1). Therefore, if the optical matrix element of the biexciton is $\mu\,\alpha$, the matrix element of the scattering state must be $\mu\,(1-\alpha)$. In all calculations presented in this paper, the parameters

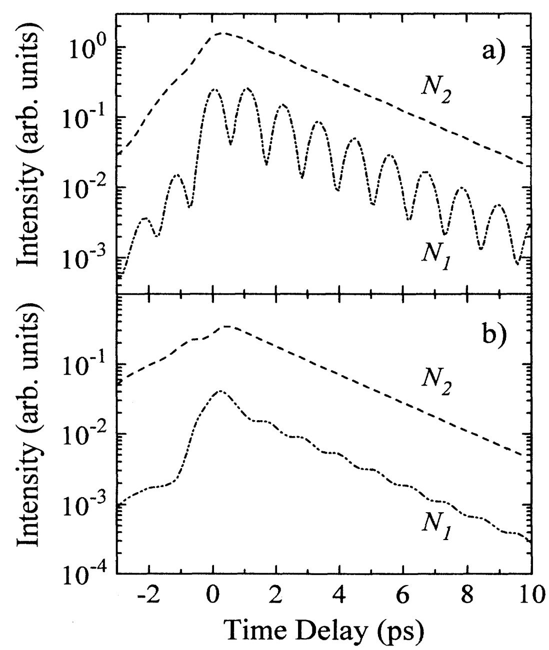

FIG. 6. TI-DFWM signal versus time delay τ calculated using the ten-level scheme for (a) parallel and (b) cross-polarized laser fields and corresponding excitation conditions N_1 and N_2. Parameters used for calculations: local field $L = 0.02$ meV; BX binding energy $\Delta = 1.2$ meV; dephasing times $T_2^{hh/G} = 4.3$ ps, $T_2^{lh/G} = 3.8$ ps, $T_2^{BX/G} = 2.3$ ps, $T_2^{2hh/G} = 1.0$ ps, $T_2^{BX/hh} = 3.7$ ps, $T_2^{2hh/hh} = 3.7$ ps.

are $\mu = 1.5$, $\alpha = 0.5$ (the matrix elements for the hh- and the lh-exciton transitions are 1 and $\frac{1}{\sqrt{3}}$).

The calculated TI-DFWM signals for the excitation conditions N_1 and N_2 are shown in Fig. 6. These curves show the same behavior as the experimental data (Fig. 2): stronger hh/lh beating at positive time delay for PP as compared to CP excitation for N_1, the same decay times for N_1 and N_2 at positive delay, and different slopes at negative delay. Only the 2hh-exciton/biexciton beating does not appear in the calculated curves, the reason for this is discussed below.

The decay at positive delay is determined by the dephasing time $T_2^{\mathrm{hh}/G}$, i.e., the dephasing of the hh exciton state to the ground state (see Fig. 1), and the damping of the lh/hh beating is controlled by the lh dephasing time $T_2^{\mathrm{lh}/G}$.[3] We have taken the values $T_2^{\mathrm{hh}/G} = 4.3$ ps and $T_2^{\mathrm{lh}/G} = 3.8$ ps in order to obtain good agreement with the experimental data. The weaker hh/lh beating in the CP signal compared to the PP case can be reproduced in the calculations by choosing smaller matrix elements for the one-exciton to two-exciton transitions which involve lh states. This is due to the fact that the upper transitions contribute more to the CP signal than to the PP signal. The slopes at negative time delay are due to the dephasing of those off-diagonal elements of the density matrix which describe the dephasing of a two-exciton state to the ground state, e.g., $T_2^{\mathrm{BX}/G}$ or $T_2^{\mathrm{2hh}/G}$. Since the excitation energy for N_2 is closer to the biexciton, the biexcitonic state has a much stronger influence on the DFWM signal than in N_1. Therefore, for CP excitation the slope at a negative delay is dominated by the biexciton to ground state dephasing time $T_2^{\mathrm{BX}/G}$. The slope of the decay for negative time delays under excitation conditions N_1 and B are determined by a combination of the other two-exciton to ground state dephasing times. However, due to the smaller matrix elements of the lh states, the dephasing time is dominated by $T_2^{\mathrm{2hh}/G}$. For the calculations, we have taken the values: $T_2^{\mathrm{BX}/G} = 2.3$ ps, $T_2^{\mathrm{2hh}/G} = 1$ ps.

As stated above, a detailed analysis of the calculations shows that the PP and the CP signals are dominated by the lower and the upper transitions, respectively. Therefore biexcitonic effects should be observed more clearly for CP excitation. The TI as well as the TR calculations and measurements support these conclusions.

Figures 7(a) and 7(b) show the results of the calculation of the TR-DFWM signal under excitation condition B for PP and CP polarized excitation pulses, respectively. Since the difference between the excitation conditions $N_{1,2}$ and B is not only the duration of the laser pulses, but also the exciton density, we increase the local field parameter from $L = 0.02$ meV to $L = 0.1$ meV, in order to take into account the lower exciton density for B. In earlier publications, it has been shown that strong local field effects appear for small exciton densities in a well, whereas their importance decreases with increasing excitation density because of exciton-exciton screening effects.[23] All other parameters remained unchanged.

The theoretical curves in Figs. 7(a) and 7(b) show the same strong beating phenomena as the experimental

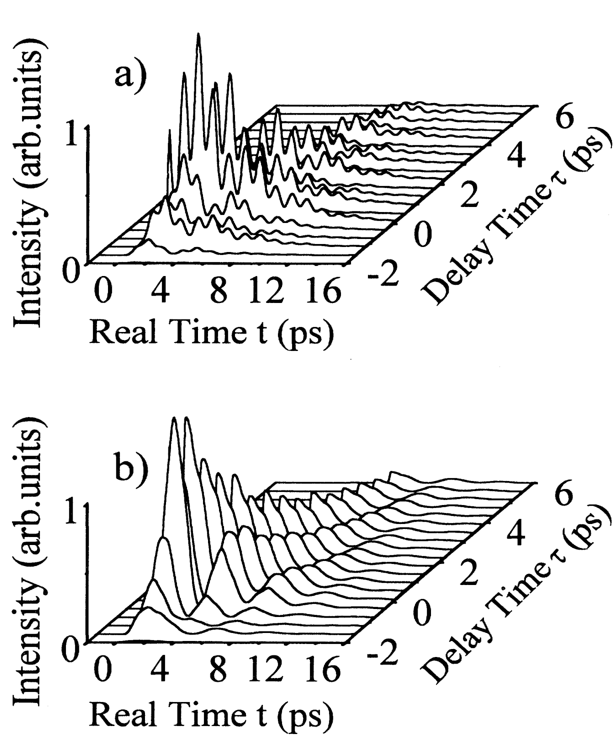

FIG. 7. Calculated intensity of the TR-DFWM signal at various probe delays based on the ten-level model for (a) parallel and (b) cross-polarized fields under excitation condition B and the same parameters as in Fig. 6, except the local field parameter: $L = 0.1$ meV.

data [Figs. 5(a) and 5(b)]. In the case of PP excitation pulses, the beat frequency is related to the heavy/light-hole splitting. The frequency of the CP signal corresponds to the splitting of the 2hh exciton scattering state and the biexciton, i.e., the biexcitonic binding energy.

As stated above, the CP signal is strongly influenced by the upper transitions, whereas the PP signal is dominated by the lower transitions. For this reason, a strong biexcitonic beating can be observed in the CP case, whereas the PP signal shows a slight biexcitonic frequency component only for negative delays. Additionally, the decay with respect to the real time t of the CP signal is determined by the time constants $T_2^{\mathrm{BX/hh}}$ and $T_2^{\mathrm{2hh/hh}}$, and corresponds to the measured value of 3.7 ps from Fig. 5(b). The decay of the PP signal is due to the dephasing of the hh exciton, i.e., $T_2^{\mathrm{hh}/G}$, and corresponds to the measured value of 4.3 ps from Fig. 5(a).

In the perpendicular case, the experimental TR-DFWM data show a distinct biexcitonic beating in the direction $t = \tau$, i.e. a modulation of the maxima, which is absent in the theoretical curve [compare Figs. 5(b) and 7(b)]. The same beating occurs in the experimental TI-DFWM data for PP as well as for CP excitation at positive delay (Fig. 2). This beating is presumably an effect of higher than third order.[8] In the framework of third-order numerical calculations, there is no oscillation belonging to the biexcitonic binding energy at positive time delay ($\tau > 0$), which can also be seen within analytical calculations. An expansion of the model to include fifth-order effects leads to an observation of this frequency at positive delay, i.e., in the mentioned direction of the TR as well as in the TI data at $\tau > 0$. The results are not presented here, because in fifth order, three exciton states also become important but are not accounted for within these calculations.

IV. CONCLUSION

The TR-DFWM signal from a GaAs single quantum well shows completely different beating phenomena for parallel and perpendicular polarized excitation. Whereas parallel excitation produces a signal with distinct hh/lh beating for $\tau > 0$ and 2hh/biexciton beating for $\tau < 0$ as a function of real time, perpendicular excitation shows a clear hh/biexciton beating for both positive and negative τ. In spite of the surprisingly different modulation patterns observed in the TR measurements, the PP and CP polarized TI-DFWM measurements reveal quite similar traces. A distinctly longer decay time of the cross-polarized TI signal is observed for negative delays as compared to parallel polarized signal. The ratio of the TI signal amplitudes for CP and PP polarizations demonstrates the influence of the biexciton state for two-hh excitations.

We also demonstrate that the observed effects can be reproduced by calculations based on a phenomenological ten-level scheme, where Coulomb interaction is introduced via biexcitonic variation and local field effects. The calculations and the measurements are in good agreement and strongly support the conclusion that biexcitons play a significant role in the third-order nonlinear optical response in GaAs quantum wells. We also measured the dephasing time of the biexciton with respect to both the ground state and the single-exciton state. Comparison of our T_2 values with the biexciton lifetime of Ref. 17 demonstrates that the homogeneous linewidth of the biexciton luminescence line is almost lifetime limited.

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