Subpicosecond four-wave mixing in GaAs/$Al_xGa_{1-x}As$ quantum wells

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We investigate the properties of coherent exciton states in GaAs/$Al_xGa_{1-x}As$ quantum wells using the four-wave-mixing technique with subpicosecond time resolution. In the first part of this work, we show that many-body interactions in a solid manifest themselves in a line shape of the four-wave-mixing signal that is substantially different from the predictions of the generally used simple two-level model. In particular, the coherent interaction of the quasiparticles leads to a signal at negative delay times, in agreement with recent theoretical predictions. In the second part, we report in detail about experiments where energetically close exciton transitions are excited coherently with a short laser pulse. The quantum or polarization interference leads to the observation of quantum beats in the decay of the diffracted signal. These results demonstrate that quantum-beat spectroscopy can be applied to study intrinsic excitations in solids.

I. INTRODUCTION

Four-wave mixing (FWM) is a powerful technique of nonlinear-optical spectroscopy.[1] The development of laser sources creating ultrashort pulses enables one to study the ultrafast dynamics of excitations using *time-resolved* FWM. The time resolution of this method is in principle determined only by the duration of the laser pulses. Therefore this technique is well suited to study the fast (i.e., picosecond to femtosecond) scattering processes of electronic excitations in semiconductors. In III-V compounds, the dephasing of both excitons and free carriers has been investigated by time-resolved FWM. The scattering of excitons with free carriers, excitons, and phonons was studied in bulk GaAs and GaAs/$Al_xGa_{1-x}As$ quantum wells (QW's) by Schultheis and co-workers.[2-7] It was shown[2] that the dephasing time constant T_2 in QW's is of the order of picoseconds and is longer for localized excitons at the low-energy side of the transitions. Detailed studies of the temperature and density dependence[3-7] have shown that for low exciton densities ($<10^9$ cm^{-2} in QW's or $<10^{14}$ cm^{-3} in the bulk) T_2 is a few ps for both high-quality bulk GaAs and GaAs/$Al_xGa_{1-x}As$ QW's. Scattering with acoustic phonons determines T_2 in the temperature range below 30 K. At high densities, scattering with excitons and free carriers reduces T_2.

Becker *et al.*[8] have studied the dephasing time of *free carriers* in bulk GaAs at high excitation densities and room temperature using time-resolved FWM.[9] Dephasing times are determined by carrier-carrier scattering in this regime and are of the order of tens of fs. Similar experiments on QW structures[10] give dephasing times in the same range. The study of the density dependence in both systems indicates that the dephasing is indeed due to carrier-carrier scattering in the density and temperature range where the experiments were performed.

Recently, time-resolved FWM was introduced as a novel technique to study tunneling in semiconductor heterostructures.[11,12] One particular advantage is that FWM tunneling experiments can have higher time resolution than nonresonant luminescence experiments performed so far.[13] In a first experiment,[11] it was shown that the phase relaxation of excitons excited in one well of a double QW structure becomes much faster when they tunnel to the other well and scatter with free carriers. Very recently, the tunneling oscillation of a wave packet in a semiconductor double QW structure was directly observed using time-resolved FWM.[12]

In this paper, we report in detail about FWM studies of excitations in QW's with subpicosecond time resolution. In contrast to the previous studies, which have mainly dealt with the dephasing, i.e., the incoherent interactions of the excitations, we investigate here the properties of the *coherent* states. We report in detail about the recent observation[14] that many-body interactions in a solid lead to a temporal line shape of the FWM signal substantially different from previous assumptions based on noninteracting two-level systems. The *coherent interaction* of excitons leads to a diffracted signal at nega-

tive delay times, in agreement with recent theoretical predictions.[15] In the second part, we report in detail about experiments where energetically close exciton transitions are excited coherently with a short laser pulse. The quantum or polarization interference leads to the observation of quantum beats[16–18] in the decay of the diffracted signal.

The paper is organized as follows: Section II explains the experimental technique and discusses the temporal line shape observed in a self-diffracted degenerate-FWM experiment. In Sec. II A, we discuss the previous models for the line shape and describe the recent theoretical prediction of a signal at negative delay times due to the coherent interaction of the excitations. In Sec. II B, we study the line shape in a $GaAs/Al_xGa_{1-x}As$ QW with a linewidth mainly due to homogeneous broadening. The temperature and density dependence of the FWM line shape are in agreement with the theoretical predictions. In Sec. II C we study the line shape in a $GaAs/Al_xGa_{1-x}As$ QW which is mainly inhomogeneously broadened. At higher densities, the line shape shows dramatic changes and develops a shoulder, in agreement with theory and recent observations in the InGaAs material systems.[19] In Sec. III we study the polarization decay in samples with closely spaced exciton levels, which leads to the observation of quantum beats. In Sec. III A we introduce the topic of quantum beats in solids. In Sec. III B we study quantum beats between the heavy-hole (hh) and light-hole (lh) transitions in a QW. We show that the observation of quantum beats at positive *and* negative delay times confirms the physical model for the exciton-exciton interaction presented in Sec. II. In Sec. III C we present results for a $GaAs/Al_xGa_{1-x}As$ QW where the hh transition is split into three lines due to a spatially varying well width. In Sec. III D we present results for the polarization interference of free and bound excitons. A brief conclusion is given in Sec. IV.

II. LINE SHAPE IN SELF-DIFFRACTED DEGENERATE-FOUR-WAVE-MIXING EXPERIMENTS

A. Description of the technique and line-shape theory

Figure 1 shows a schematic setup of our experiments. We use the so-called two-pulse self-diffracted degenerate FWM technique:[20,21] A first pulse (no. 1) is focused onto the sample by a lens and induces a polarization. A second pulse (no. 2), which is delayed by a time $T=t_2-t_1$, spatially overlaps pulse no. 1 at an angle in the sample. The pulses are polarized perpendicularly, i.e., they create an excitonic-orientational grating with spatially homogeneous exciton density. The amplitude of this grating depends on the time delay between the pulses: With increasing delay between pulse nos. 1 and 2, the macroscopic polarization created by pulse no. 1 decays due to scattering (described by the dephasing time T_2), and the amplitude of the grating becomes smaller. In the self-diffracted FWM technique, pulse no. 2 not only creates the grating, but is also itself diffracted by this grating. The diffracted field has the wave vector

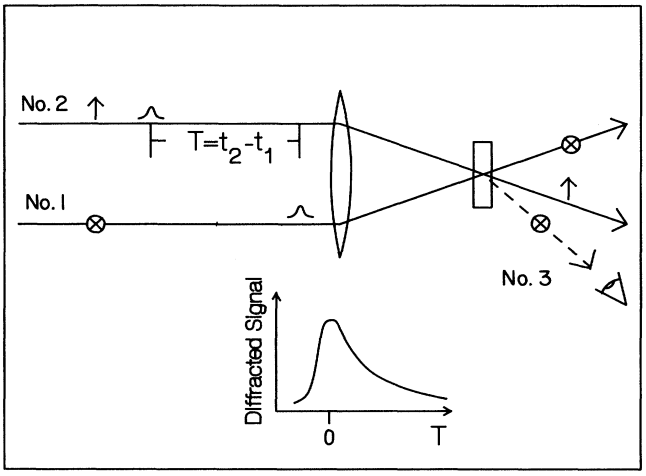

FIG. 1. Schematic of the four-wave-mixing experiment. Measured is the diffracted signal in the direction $\mathbf{k}_3=2\mathbf{k}_2-\mathbf{k}_1$ as a function of the delay time between the pulses $T=t_2-t_1$.

$\mathbf{k}_3=2\mathbf{k}_2-\mathbf{k}_1$ (see Fig. 1) and is detected as a function of the time delay $T=t_2-t_1$. The decrease of the diffracted signal in the direction $\mathbf{k}_3$ with increasing T therefore reflects the decay of the polarization created by pulse no. 1: If T is small, the polarization produced by pulse no. 1 is preserved when pulse no. 2 arrives, and the diffracted signal is large. If T is large, the polarization has decayed [$\sim\exp(-T/T_2)$], and the diffracted signal is weak. In the case of an inhomogeneously broadened transition, the situation is more complicated: In this case, the net polarization decays very quickly due to the destructive interference of excitations with different energies. The second pulse then inverts the phases of the excitations, which leads to the observation of a "photon echo" after a delay time which is identical to the distance between the two pulses. If we only trace the time-integrated diffracted signal as a function of the delay T between the two pulses, we cannot directly observe the echo pulse. Recently, Noll *et al.*[22] have directly observed the photon echo by time resolving the diffracted beam with a streak camera. This observation was made possible by the very long (up to several hundred ps) phase relaxation times of localized excitons in $Cd_xSe_{1-x}S$. Very recently, Webb, Cundiff, and Steel[23] have used the upconversion technique to temporally resolve the diffracted signal in a $GaAs/Al_xGa_{1-x}As$ QW.

Yajima and Taira[24] have calculated the temporal line shape of the third-order diffracted signal in the direction $\mathbf{k}_3$ as a function of the delay time for noninteracting two-level systems. For a homogeneous transition and δ-shaped excitation pulses, they obtained

$$I(T)\sim\begin{cases}0, & T<0\\ \exp(-2T/T_2), & T>0\end{cases} \tag{1}$$

and for an inhomogeneous transition

$$I(T) \simeq \begin{cases} 0, & T<0 \\ \exp(-4T/T_2), & T>0 \end{cases}, \quad (2)$$

i.e., in both cases, the signal for $T<0$ should be *identical to zero* and should decay exponentially for $T>0$, with the decay reflecting the loss of coherence due to the phase relaxation of the excitations. For finite pulse duration, the rise should not be instantaneous, but should closely follow the pulse shape. This theory was used by Schultheis and co-workers[2–7] to analyze their experimental results in bulk GaAs and $GaAs/Al_xGa_{1-x}As$ QW's.

It was recently pointed out by Stafford, Schmitt-Rink, and Schäfer[15] that a more detailed (i.e., microscopic) description of excitons leads to a more complicated temporal line shape. They showed that excitons not only interact with the electric field of the driving laser pulses, but also with the polarization of all other excitons. This interaction leads to the *diffraction of polarization in addition to the diffraction of fields.* In particular, the coherent Coulomb interaction of excitons allows the diffraction of polarization and thus gives rise to a signal at negative delay times ($T<0$), which is not predicted by the noninteracting two-level model.[24] The main difference in the work of Stafford, Schmitt-Rink, and Schäfer[15] is that a microscopic theory of the coherent nonlinear-optical response of excitons is employed. This microscopic theory has been developed earlier by Schmitt-Rink and co-workers[25,26] and has been successfully applied to various problems in nonlinear exciton optics.

In the following, we give a brief account of the application of the microscopic exciton theory to FWM. In a simple picture, one would assume that the electron-hole pair amplitudes ψ_k, which represent vertical transitions at wave vectors $\mathbf{k}$, couple only to the external field E. Then the Rabi frequency Δ_k would be simply

$$\Delta_k = \mu_k E , \quad (3)$$

where μ_k is the interband dipole matrix element. The salient feature of the detailed description[15,25,26] is that it also includes the Coulomb potential of all other electron-hole pairs at wave vectors $\mathbf{k}'$. The total coupling including external *and* Coulomb fields is then

$$\Delta_k = \mu_k E + \sum_{k'} V_{k,k'} \psi_{k'} \quad (4)$$

with the term $\mu_k E$ representing the interaction with the external field and the term $\sum_{k'} V_{k,k'} \psi_{k'}$ representing the Coulomb interaction of the pair amplitudes. If the external field is generated by an ultrashort laser pulse with electric field $E \simeq \delta(t)$, the first term in Δ_k varies like $\sim\delta(t)$ and produces a third-order FWM signal with a time dependence following the noninteracting two-level model of Yajima and Taira,[24] i.e., a signal is only observed for $T>0$. The induced pair amplitude ψ_k and thus the second term in Δ_k, however, exhibit a steplike rise and a $\sim\exp(-t/T_2)$ decay. The associated polarization ($P = \sum_k \mu_k^* \psi_k$) interacts with the polarization grating both for $T<0$ *and* $T>0$. In third order, this interaction involves two polarizations propagating in the direction $\mathbf{k}_2$ and one propagating in the direction $\mathbf{k}_1$ and hence produces a FWM signal $\sim\exp(4T/T_2)$ for negative time delays. For positive time delays, the resulting FWM signal decays as the signal arising from the first term in Δ_k, with a similar magnitude. Therefore the diffracted signal should show a rising wing for $T<0$ and a decay for $T>0$:

$$I(T) \simeq \begin{cases} \exp(4T/T_2), & T<0 \\ \exp(-2T/T_2), & T>0 \end{cases}. \quad (5)$$

In the case of an inhomogeneous transition, the macroscopic polarization $P = \sum_k \mu_k^* \psi_k$ will decay much faster than $\sim\exp(-t/T_2)$. Therefore the signal at negative time delay should disappear for transitions where the broadening is predominantly inhomogeneous. In the case of semiconductors, this means that the signal at negative delay times will be only observable in samples of very high quality.

Recently, Wegener *et al.*[19] have discussed the effects of coherent polarization interactions on FWM in detail. They have also extended the theory to more general situations and used anharmonic oscillators as a simple model to describe the interaction of the polarizations. This approach is particularly helpful to understand the recently reported dramatic changes of the line shape at high excitation intensities, where the microscopic equations can only be treated numerically.[14] Other limits of the noninteracting two-level approach[24] have been discussed by Abram.[27]

B. Experimental results for the line shape at low intensities

The sample used for this experiment contains ten periods of a double QW structure, composed of a 170-Å GaAs QW, a 17-Å $Al_{0.35}Ga_{0.65}As$ barrier, and a 120-Å GaAs QW. The periods are separated by 200-Å-thick $Al_{0.35}Ga_{0.65}As$ barriers. All thicknesses and the Al content are nominal. The sample was grown by molecular-beam epitaxy (MBE) on *n*-doped (100)-oriented GaAs substrate at a (pyrometrically determined) growth temperature of 650 °C without growth interruption at the interfaces. The sample was carefully characterized by cw photoluminescence (PL) and PL excitation (PLE) spectroscopy at a lattice temperature of $T_L = 2$ K. The linewidth of the 170-Å hh transition was about 0.7 meV in PL as well as PLE spectroscopy. The peak shift between PL and PLE ("Stokes" shift) was about 0.1–0.2 meV, which indicates the high quality of this multiple QW sample. Photocurrent spectra of a sample with electrical contacts were used to determine the transitions related to higher subbands. The results indicated a well width slightly (10%) smaller than the design value. A high-resolution transmission electron microscopy (TEM) study gave indeed well widths of 54 ± 1 monolayers (ML), i.e., 153 Å for the wide well (WW), and 39 ± 1 ML $\sim$ 110 Å for the narrow well (NW), in excellent agreement with the PL studies. The barrier thickness determined by TEM was 6 ML $\sim$ 17 Å, i.e., identical to the design value. The TEM studies did not indicate any fluctuation of these values across the ten periods of the sample.

To perform experiments in transmission configuration, the samples were mounted with transparent epoxy glue to sapphire disks. Then, the substrate was removed by selective wet etching. The upper part of Fig. 2 shows the transmission spectrum of the sample in the energy range of the lowest transitions of the 170-Å well, taken with a cw laser at $T_L = 5$ K. Clearly visible are the transitions from the first electron subband to the first hh subband and the first lh subband. The linewidths of the transitions and the splitting between the hh and lh exciton lines are the same as in the PLE data in the unprocessed sample. The energy of the transitions, however, is about 1.5 meV lower than before, indicating homogeneous tensile strain in the QW layers due to the thermal expansion coefficient mismatch between the $GaAs/Al_xGa_{1-x}As$ sample and the sapphire disk.

The laser source used for the time-resolved studies is a tandem-pumped dye laser system: A mode-locked YLF laser equipped with a KTP second-harmonic generator produces 30-ps-long 528-nm pulses with a repetition rate of 76 MHz and an average power of about 2.5 W. This pulse train synchronously pumps a Rhodamin 6G dye laser producing pulses of a few ps length at 580 nm with an average power of about 700 mW. This output is used to pump a LDS-751 (Styril 8) dye laser producing pulses of 450–500-fs duration tunable between 700 and 815 nm with an average power of up to 150 mW. The spectral width of the laser pulses is about 3.6 meV.

In a first experiment, we study the line shape of the FWM signal in the low-intensity regime. We concentrate here on the lowest hh to electron transition of the WW. The fact that we are using a double QW structure has no influence on our results: We excite only the lowest transition of the WW. Electron and hole levels of the two wells are then out of resonance and the wave functions are localized in one well. The excitation intensity was about 160 kW cm^{-2}, which corresponds to an exciton density of about 4×10^8 cm^{-2}.[28] The lower part of Fig. 2 shows the spectrum of the laser. The excitation wavelength (dashed line) was chosen slightly below the hh transition to excite only the hh transition and avoid the simultaneous excitation of lh excitons or free carrier states.

Figure 3 shows the diffracted FWM signal for different lattice temperatures T_L, 5 (top), 40 (middle), and 70 K (bottom). The decay times are independent of the intensity in this range; a further reduction of the intensity yields the same line shapes. The line shape at 5 K shows the expected exponential decay for positive delay times. The time constant of the decay is about 1.2 ps, which corresponds to a phase relaxation time T_2 between 2.4 (in the case of homogeneous broadening) and 4.8 ps (mainly inhomogeneous broadening). In the self-diffracted FWM technique, one cannot discriminate between homogeneous and inhomogeneous broadening. The small Stokes shift between PL and PLE, however, and the approximate agreement of the observed linewidth with the inverse of $T_2/2$ indicate that the hh transition is predominantly homogeneously broadened. The value range obtained for T_2 is in reasonable agreement with the values we have observed for an inhomogeneously broadened QW of 70 Å width and with the values reported by Schultheis *et al.*,[4] who obtained a T_2 of 2 and 3 ps for high-quality 270- and 135-Å QW's, respectively.

For positive delay times, the shorter decay time of the signal for increased lattice temperature is well known[4] and reflects the reduction of the phase relaxation time T_2 due to the increasing scattering of excitons with thermal phonons. The similar behavior of the *signal rise*, however, confirms the theoretical prediction[15] of a rising wing with a rise time related to T_2. Figure 4 shows the rise (dots) and decay (circles) time constants, obtained by fitting an exponential function, for the temperature range from 5 to 70 K.[29] At the lowest temperature of 5 K, the times for rise (1150 fs) and decay (650 fs) are within the error (±50 fs) equal to 2:1, i.e., the theoretical prediction of a rising wing with half of the time constant of the decay [Eq. (5)] is nicely confirmed. With increasing temper-

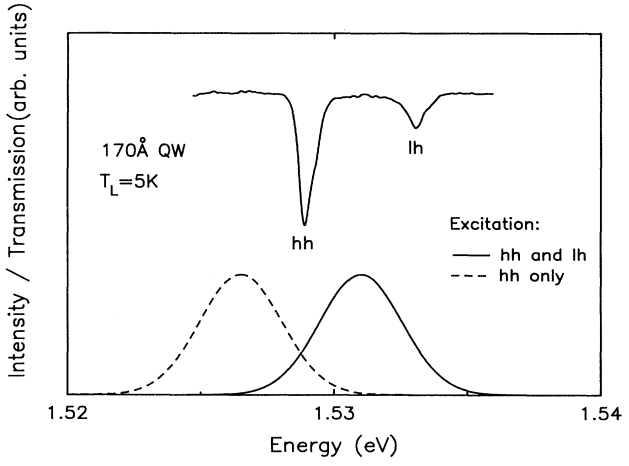

FIG. 2. Upper part: transmission spectrum of the 170-Å quantum-well sample, taken with a cw laser at very low intensity. The heavy-hole and light-hole excitons are marked by the hh and lh symbols, respectively. Lower part: laser spectrum for excitation of hh and lh (solid line) and hh only (dashed line).

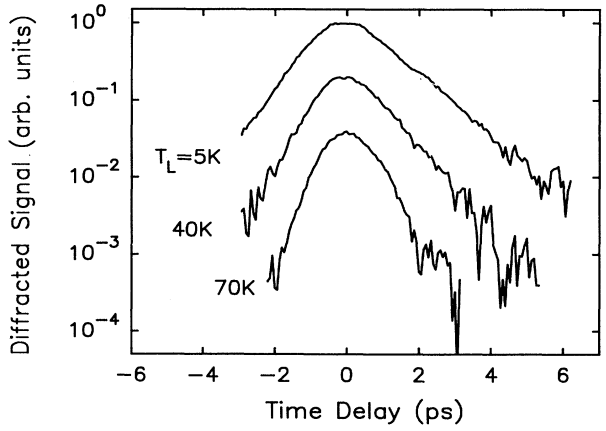

FIG. 3. Four-wave-mixing line shapes for the 170-Å quantum-well sample for various lattice temperatures, taken at very low intensity. Note that both decay and rise time decrease with increasing lattice temperature.

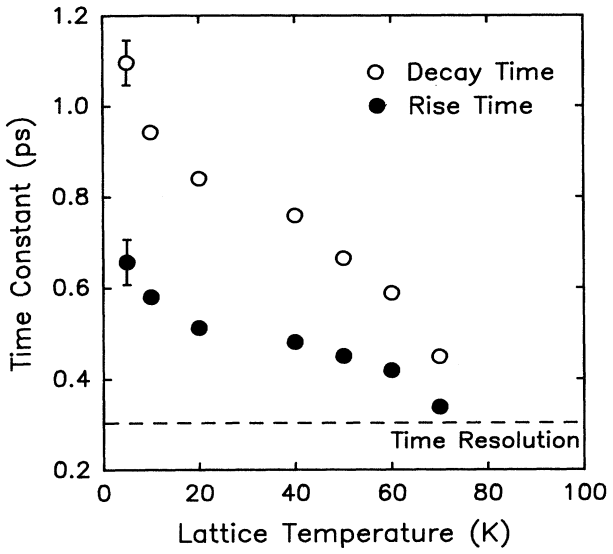

FIG. 4. Decay (circles) and rise (dots) times of the diffracted signal vs lattice temperature. The dashed line indicates the apparative time resolution limit.

ature, however, the ratio between decay and rise times decreases due to the time resolution limit of our experiments.[30]

Another way to study the dependence of the FWM line shape on the dephasing time constant T_2 is a variation of the excitation density: T_2 decreases with increasing exciton density due to exciton-exciton scattering.[4] Figure 5 shows the line shape, measured at $T_L = 5$ K in the same sample as discussed before, as a function of the laser pulse intensity. I_0 is about 160 kW/cm^{-2} and corresponds again to an excitation density of about 4×10^8 excitons cm^{-2}. The dependence of the decay and rise times on intensity shows the same behavior as in the case of temperature variation: With increasing density, the decay becomes faster due to a decrease in T_2. Again, the rise time also decreases, in agreement with the theoretical prediction. This is also visible in Fig. 6, which shows the decay (circles) and rise (dots) times, plotted versus the intensity. Once more, the time constants have approximately the ratio 2:1 and decrease both with increasing intensity. For higher intensities, however, the rise time does not decrease further, and the rise becomes nonexponential. This is caused by the increase of the signal at negative delay time at high intensities, as observed in the GaAs/Al$_x$Ga$_{1-x}$As system (for a discussion of these results, see the next section) and in the InGaAs systems.[19]

The observations for the temporal line shape of the self-diffracted FWM signal at low to medium intensities can be summarized as follows: We observe a pronounced signal at *negative* delay times, in addition to the predictions of the simple model of noninteracting two-level systems,[24] which was generally employed up to now to analyze self-diffracted FWM experiments. The signal rises for negative delay times with half the time constant of the decay for positive times, in agreement with recent theoretical predictions.[15] These new effects are caused by a coherent interaction of the polarizations and should also be important for wave-mixing experiments in media other than semiconductors.[14,19]

We emphasize here once more that the observation of this rising wing is only possible for transitions which are predominantly homogeneously broadened. A very simple qualitative explanation for this effect is that the interaction between the polarization left over by pulse no. 2 and

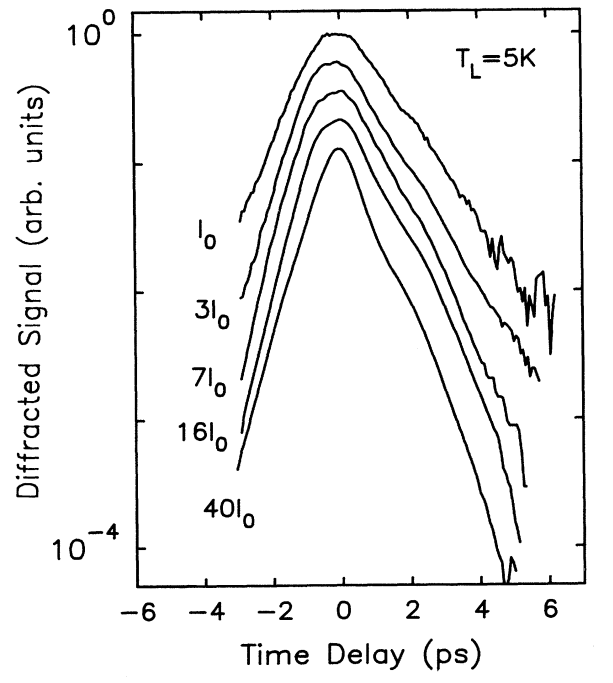

FIG. 5. Four-wave-mixing line shapes for the 170-Å quantum-well sample for various excitation intensities, taken at low lattice temperature ($T_L = 5$ K). I_0 corresponds to an intensity of about 160 kW cm^{-2}.

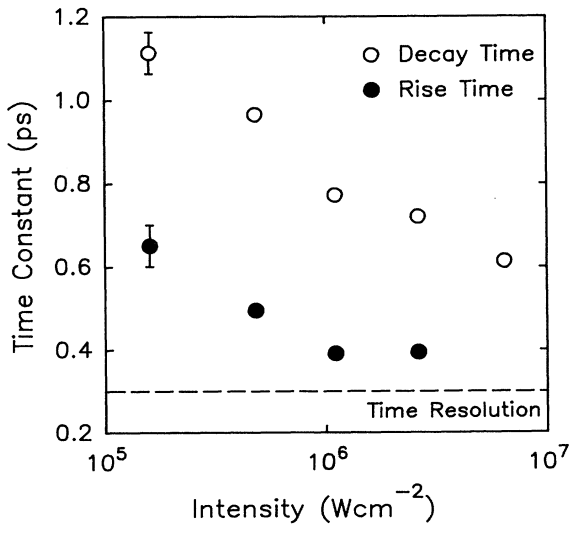

FIG. 6. Decay (circles) and rise (dots) times of the diffracted signal vs excitation intensity.

pulse no. 1 is not possible when the macroscopic polarization of pulse no. 2 quickly decays due to inhomogeneous broadening. The observation of these low-density effects in semiconductors is therefore presently only possible in the GaAs/$Al_xGa_{1-x}As$ material system, where crystal growth methods are advanced enough to produce samples which show predominantly homogeneously broadened transitions. Experiments on samples with inhomogeneously broadened lines did not show a pronounced signal at negative delay times, in agreement with the theoretical prediction.[19]

C. Experimental results for the line shape at high intensities

In the preceding section, we have discussed the line shape in the low-intensity limit. More precisely, we have discussed effects which concern the signal due to the third-order polarization or $\chi^{(3)}$. Recently, it was observed[14,19] that, at high intensities, exciton-exciton interactions lead to a remarkable interference of this term with higher-order terms in the nonlinear polarization. This effect is discussed in Refs. 14 and 19 for the InGaAs material systems.

Here, we briefly discuss the FWM line shape at high intensities in the GaAs/$Al_xGa_{1-x}As$ material system. The sample employed for this study is a ten-period 70-Å QW structure. Details of this structure and its characterization will be presented in Sec. III C. Figure 7 shows the line shape at high intensity ($\sim$20 MW cm^{-2}) for various detunings of the laser from the hh exciton transition. For resonant excitation (detuning of 0 meV), the line shape resembles the low-density case; however, the rise and decay times are much shorter due to efficient exciton-exciton scattering leading to a short phase relaxation time T_2. In this particular experiment, both times are mainly limited by the pulse width of the laser.

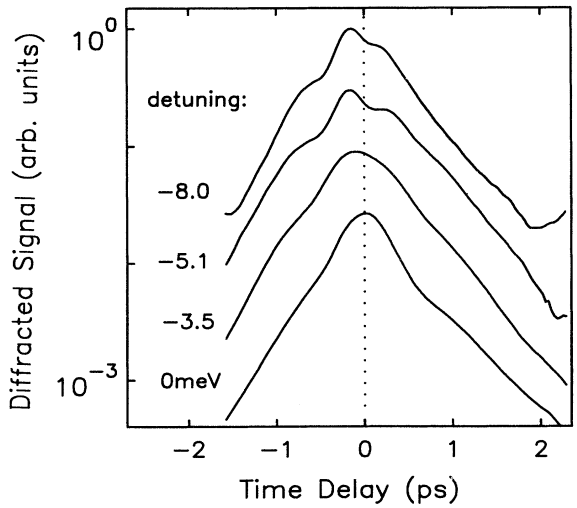

FIG. 7. Four-wave-mixing line shapes for the 70-Å quantum-well sample for various laser detunings from the heavy-hole exciton transition, taken at low lattice temperature (T_L = 5 K) and high excitation intensity ($\sim$20 MW cm^{-2}).

With increasing detuning, however, the line shape changes drastically: A pronounced shoulder appears at small negative delay times. The observation of such a second temporal "peak" is in agreement with the results for the InGaAs material systems[14,19] and is caused by the destructive interference of various orders of the nonlinear polarization. In simple terms, the negative-time-delay signal due to the third-order term $\chi^{(3)}$ is compensated around $T=0$ by the contribution of the fifth-order term $\chi^{(5)}$, which has the opposite sign. Further away from $T=0$, the contribution of $\chi^{(5)}$ disappears because it decays with faster time constants than the $\chi^{(3)}$ term. This effect and a simple physical model are discussed in detail by Wegener *et al.*[19] A sophisticated numerical calculation which treats all orders of polarization[14,19] is also in good agreement with the experimental observations.

Some effects in the experiment are, however, not understood at present: (i) Physically, it is not yet clear why the shape of the "double-peak structure" both in the GaAs and InGaAs material systems depends so strongly on the laser detuning and (ii) the observation of a third temporal "peak" for small positive delay times is not predicted by the theory. The dependence on the excitation energy is similarly observed in the InGaAs systems.[19] The additional "peak" at positive delay times, however, is only observed in the GaAs/$Al_xGa_{1-x}As$ sample. More experimental and theoretical work would be helpful to fully understand these phenomena.

III. QUANTUM BEATS OF EXCITONS IN QUANTUM WELLS

A. Introduction

If a short (and therefore spectrally broad) pulse is used to excite two or more closely spaced optical transitions, a coherent superposition of these states, (i.e., a wave packet) is created. Due to the slightly different energies of the transitions, the macroscopic polarization then shows an interference. In the simplest case of two transitions with energies E_1 and E_2, the macroscopic polarization displays a beating with a time constant of

$$T=h/\Delta E\ , \tag{6}$$

where $\Delta E=E_2-E_1$. This effect is usually called "quantum beats" and is an important technique in nonlinear spectroscopy.[1] Quantum beats have been observed in atoms,[31] molecules,[32] and nuclei.[33]

These observations were made for systems where two excited states with energies E_1 and E_2 have a common ground state with an energy E_0, i.e., the system can be approximately described as a three-level system. However, the observation of interference effects is also possible if two (or more) two-level-like systems with slightly different transition energies are excited coherently. Such interferences of polarizations have also been called quantum beats and were first observed in the vibrational transitions of isomeric molecules by Laubereau, Wochner, and Kaiser.[34] For a detailed discussion, we refer to the article by Zinth and Kaiser in Ref. 35.

Quantum beats from excitations in solids have been ob-

served, e.g., for F centers in CaO (Ref. 36) and molecular crystals.[37] In both cases, however, the quantum beats are not caused by the interference of *extended electronic states*. At first glance, one might think that such an observation is not possible. The macroscopic polarization of a whole band of excited states will disappear very quickly due to the different polarization oscillation frequency of every electronic state. In other words, energy bands can be considered as a very broad inhomogeneous transition in **k** space.[8] Such a fast decay of the macroscopic polarization will then prevent the observation of an interference. However, we have recently pointed out[16] that the existence of *excitons* changes the situation drastically: The electron-hole Coulomb interaction gives rise to sharp excitonic energy levels separated from the energy bands. These excitonic transitions can be predominantly homogeneously broadened, and a coherent superposition of excitonic states should thus enable the observation of quantum beats from extended states in a solid. We have indeed observed quantum beats in solids by studying QW's with split exciton levels.[16] Independently, quantum beats of excitons were observed in $AgBr_2$.[38] Here, the excitons were split by an external magnetic field. Beats in the luminescence of type-II superlattices were also interpreted as quantum beats.[39] Very recently, quantum beats between excitons bound at defects in BI_3 have been observed.[40]

In the following three sections, we will discuss three different observations of quantum beats of excitons in QW's. In a first part, we will discuss our recent observation of quantum beats due to the splitting of lh and hh excitons in a QW.[17] This corresponds approximately to a three-level system, as discussed above. In the second part, we will discuss quantum beats between excitonic levels which are split due to different quantum confinement.[16] In a third part, we will discuss quantum beats between free and bound exciton states.[18] The second case corresponds to quantum beats between two-level-like systems. We want to point out here that the observation of quantum beats is not a specific property of quasi-two-dimensional systems. We only make use of the fact that such QW systems offer well-defined split exciton levels.

In our experiments, we observe the interference in the polarization decay by time-resolved self-diffracted FWM. This technique has been used before to study quantum beats in molecules.[41] One particular advantage of the FWM technique is that the observed signals are emitted in a background-free direction.

B. Quantum beats between light-hole and heavy-hole excitons

In bulk GaAs lh and hh bands are degenerate at the Γ point. This degeneracy is lifted in a QW structure and light holes have a larger confinement energy than heavy holes. This leads to the observation of two distinct excitonic transitions in optical spectra. A typical example was shown in Fig. 2, where hh and lh transitions are clearly visible. The splitting between the two lines in this sample is slightly more than 4 meV.

We have measured the polarization decay in this sample for two different excitation energies: in between the hh and lh transitions (solid line in Fig. 2), and slightly below the hh transition (dashed line in Fig. 2). In the first case, both hh and lh transitions are simultaneously excited, in the second case, the excitation of the lh transition is negligible compared to the hh excitation. Figure 8 shows the diffracted signal for the two different excitation energies: If only the hh transition is excited (dashed line), the signal shows the usual exponential decay reflecting the polarization decay due to dephasing. If both hh and lh transitions are excited simultaneously, the signal shows a beating due to polarization interference. The beat period is 960 ± 50 fs, which is in excellent agreement with the period of 980 fs obtained by inserting the measured splitting between hh and lh excitons of 4.2 meV into Eq. (6).[42] Figure 9 shows the diffracted signal for excitation in between the hh and lh transitions as a function of the excitation intensity. I_0 corresponds to an intensity of about 160 kW cm^{-2}. The periodic modulation of the FWM signal is clearly decreasing with increasing intensity.

A theoretical estimate of the line shape can be easily obtained by calculating the third-order signal for a homogeneously broadened three-level system excited with δ-shaped pulses. One obtains, for the intensity I as a function of the delay time T,[12]

$$I(T)\simeq\Theta(T)\left[\frac{w_1^2}{2\gamma_1}+\frac{w_2^2}{2\gamma_2}+\frac{2w_1w_2(\gamma_1+\gamma_2)}{(\gamma_1+\gamma_2)^2+\Delta E^2}\right]\times[w_1^2e^{-2\gamma_1T}+w_2^2e^{-2\gamma_2T}+2w_1w_2\cos(\Delta ET)e^{-(\gamma_1+\gamma_2)T}], \quad (7)$$

where $\Theta(T)$ is the Heaviside step function, and $\gamma_{1,2}$ and $w_{1,2}$ are the dephasing rates and spectral weights of the two transitions. Note that the signal can be fully modu-

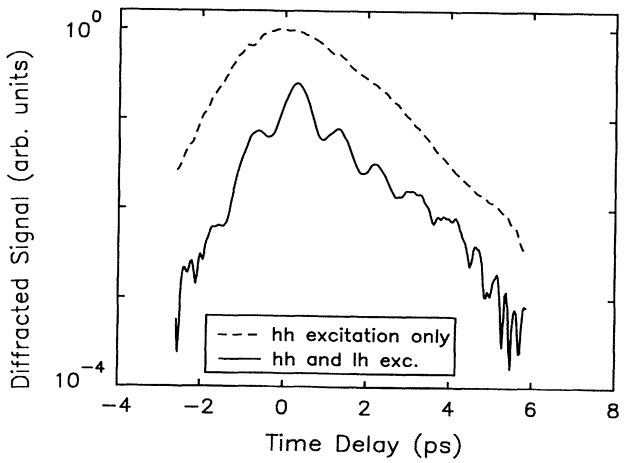

FIG. 8. Four-wave-mixing line shapes for the 170-Å quantum-well sample for excitation of the heavy-hole transition only (dashed line) and simultaneous excitation of heavy-hole and light-hole transitions (solid line). Lattice temperature ($T_L=5$ K) and intensity (~160 kW cm^{-2}) are kept low.

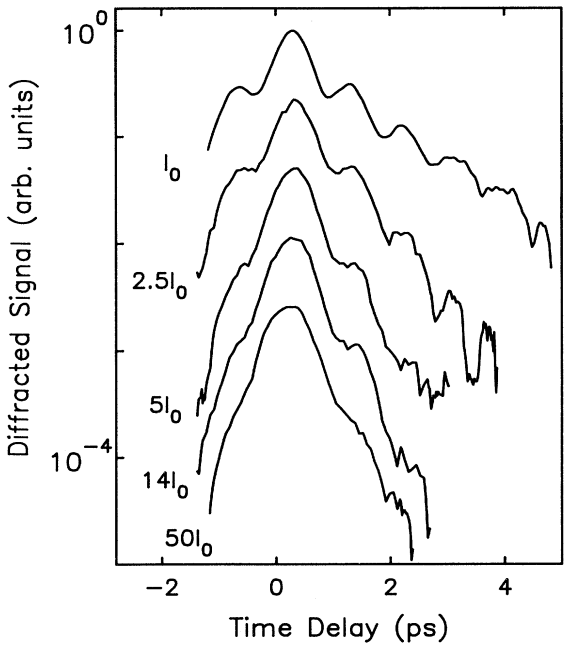

FIG. 9. Four-wave-mixing line shapes for the 170-Å quantum-well sample for simultaneous excitation of the heavy-hole and light-hole transitions and various excitation intensities.

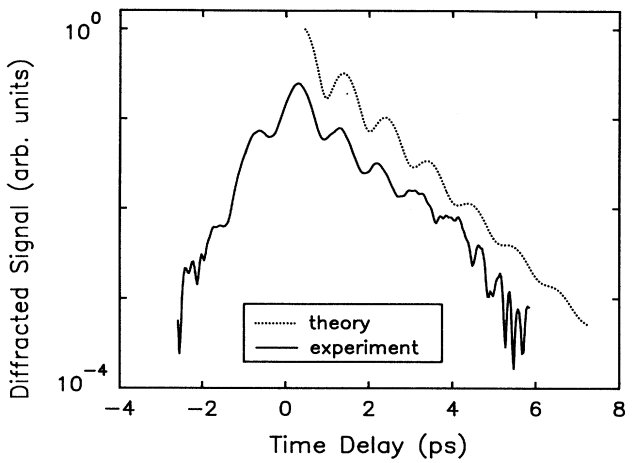

FIG. 10. Comparison of the experimental results (solid line) for heavy-hole light-hole quantum beats and the predictions of a simple three-level model (dashed line).

lated, if the two transitions have the same spectral weight.

Figure 10 shows the theoretical line shape for the following parameters: $w_1=1$, $w_2=\frac{1}{3}$, $\gamma_1=0.35$ meV, and $\gamma_2=0.55$ meV. The values for γ agree well with the experimentally observed linewidths [full width at half maximum (FWHM) of 2γ] if homogeneous broadening is assumed. The values for w_1 and w_2 reflect the different spectral weights of hh and lh excitons, as observed in the experimental spectra and expected from theoretical considerations. The theory (dotted line in Fig. 10) reproduces the observed decay rather well, despite its simplicity.

A striking difference between this simple model and the experiment is again the signal at negative delay time. The theory does not predict any signal for $T<0$, and the inclusion of a finite pulse length would again merely lead to a rising edge at $T=0$. The experimental signal, however, shows clearly the existence of quantum beats *even for negative delay time.*[15] This is yet another confirmation of the importance of exciton-exciton interactions, which were discussed in Sec. II. Quantum beats of hh and lh excitons have also been observed in Ref. 43. These authors have explained signals at negative times by invoking a polarization at twice the excitation frequency. In contrast to our model, this assumption would introduce a pronounced intensity dependence of the signal at negative delay time which is not observed in our experiments.

C. Quantum beats between heavy-hole excitons split due to different confinement

In the preceding section, we discussed quantum beats between hh and lh states, which are split due to the different masses leading to a different confinement. This splitting is an intrinsic property of a QW. However, it is frequently observed[44–49] that the hh exciton transitions of QW's show a splitting due to extrinsic effects. The splitting of the lines is particularly pronounced if the molecular-beam epitaxy growth is interrupted at the interfaces. This splitting is caused by the occurrence of regions with different thickness leading to different confinement energies.

We have investigated the polarization decay in two different samples grown by MBE on *n*-doped [100] GaAs substrate. The basic sequence consists of a 70-Å-thick GaAs QW, followed by a 48-Å-thick $Al_{0.35}Ga_{0.65}As$ barrier, and a 45-Å GaAs QW. Ten periods separated by 150-Å $Al_{0.35}Ga_{0.65}As$ barriers are grown. Sample *B* is grown without growth interruptions, whereas sample *A* is grown with a growth interruption of 60 s at every interface. The samples were processed for transmission experiments in the same way as described in Sec. II B. A careful study of PL, PLE, and photocurrent gave excellent fits of peak positions when well widths about 10% smaller than the design values were used.

In the following, we present a detailed comparison of the FWM line shapes of the two samples. A brief account of these results has been given before.[16] They were, along with Ref. 38, the first to show the existence of quantum beats from extended states in solids. The inset of Fig. 11 shows PLE spectra of the WW hh exciton transition for samples *A* (top) and *B* (bottom). Sample *B* shows a single line of about 3.4 meV width (FWHM). The PL line (not shown) is shifted to lower energies by about 3 meV. This value of the Stokes shift indicates that localization effects of the excitons are important in this sample. Sample *A* shows three clearly resolved lines, split by about 2.7 meV, each with a FWHM of 1–1.5 meV. The PL spectrum (not shown) shows three similar peaks, whose energy agrees within about 0.2 meV with the PLE spectra. This indicates that the excitons associated with the different transitions are not localized. The magnitude of the splitting (2.7 meV) is smaller than the

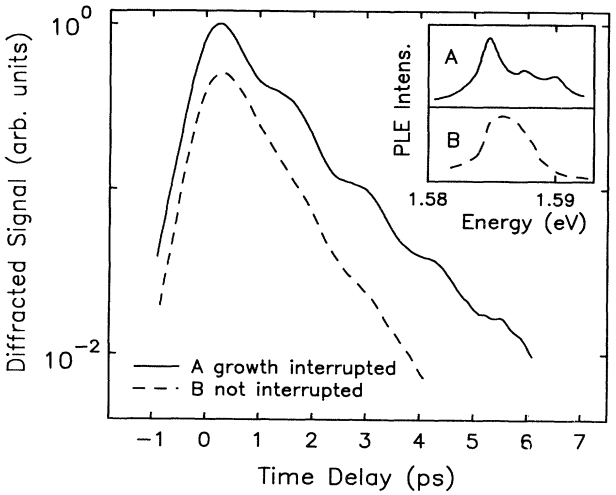

FIG. 11. Four-wave-mixing line shapes for a sample (B) with a single exciton line (dashed line) and a sample (A) with simultaneously excited split exciton lines (solid line). The inset shows the photoluminescence excitation (PLE) spectra for sample A, grown with growth interruption (top), and sample B, grown without growth interruption (bottom).

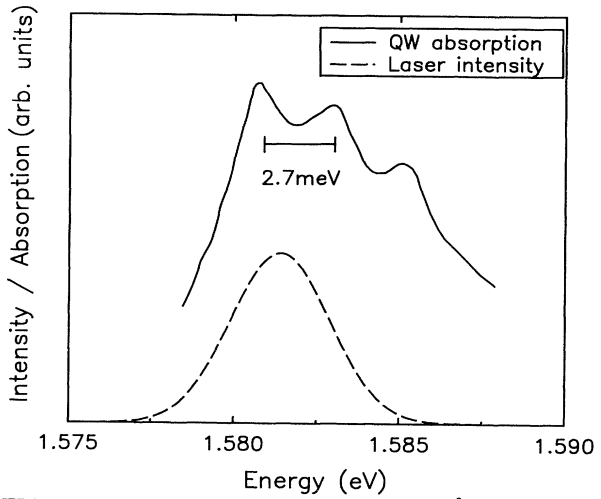

FIG. 12. Absorption spectrum of the 70-Å quantum-well sample (solid line), compared to the spectrum of the laser (dashed line).

expected value ($\sim$4.5 meV) for a monolayer fluctuation of a well of about 63 Å.[50] Splittings smaller than a monolayer fluctuation have been observed previously.[45,49]

Figure 12 shows the absorption spectrum of sample A (top), compared to the spectrum of the laser (bottom). The absorption spectrum shows again three lines, with an identical splitting to PL and PLE spectra. The separation of the peaks is smaller, probably due to some increase in linewidth because of the processing. The relative intensity of the peaks is also different from PL and PLE, probably due to the diffusion of excitons between regions of different thickness.[51,52] The spectrum is also shifted by about 2 meV due to strain.

The decay of the diffracted signal for sample A (solid line) and sample B (dashed line) is shown in Fig. 11. The excitation energy is resonant with the peak for sample B and in between the two lower-energy peaks for sample A, as shown in Fig. 12. The excitation density is about 1×10^9 excitons cm^{-2}. The diffracted signal is *very different for the two samples:* For sample B (single exciton line), the decay is nearly exponential with a time constant of about 0.8 ps, corresponding to a T_2 or 3.2 ps, if we assume an inhomogeneously broadened transition.[24] The coherence decay of sample A with split exciton lines (solid line in Fig. 11), however, shows a different behavior: *an oscillatory structure* is superimposed onto the exponential decay. It is caused by the interference of the two excitonic transitions. This interference can be considered as quantum beats of two separate two-level-like systems with slightly different transition energies, as observed before for molecules containing different isotopes.[34]

The time constant of the decay in sample A (about 1.2 ps) is *larger* than in sample B, which might be caused by the smaller inhomogeneous broadening of the excitonic transition. The period of the quantum beats is about 1.33 ps, which would correspond to an energy splitting between the exciton lines of 3.1 meV. This is slightly larger than the experimentally observed splitting of the lines of about 2.7 meV. The oscillation frequency does not depend on excitation density or temperature; however, the polarization decay becomes faster with increasing temperature and excitation density, and the signal modulation decreases.

We have also studied the narrow wells (45 Å) in the same sample. The hh exciton transition is split into two lines about 10 meV apart, i.e., the splitting is much larger than the spectral width of our laser pulse. Correspondingly, we do not observe quantum beats, simply because a simultaneous excitation of both transitions is not possible. The dephasing time of the narrower well is shorter than that of the wider well grown on the same sample, indicating that dephasing times decrease with decreasing well width.

The observation of split exciton transitions shows that the spatial regions which correspond to the different transitions are larger than the exciton diameter, which is on the order of 30 nm. This indicates that the electromagnetic interference observed in our samples is caused by excitons whose spatial separation is on the order of the laser wavelength (about 200 nm in GaAs). Cathodoluminescence scans have indicated that the regions can even have a size on the order of a few μm.[47]

To model the observed polarization decay, we follow Ref. 24 and consider a two-level-like system with a two-peak inhomogeneous distribution of the form

$$\rho(E)=\sum_{n=1}^{2}\frac{1}{2\sigma\sqrt{\pi}}\exp\left[\frac{-(E-E_n+E_0)^2}{\sigma^2}\right], \quad (8)$$

where σ is the inhomogeneous broadening. The exciton splitting $\Delta E=E_1-E_2$ is finite for sample A and zero for sample B. The spectral weight is assumed to be the same for both transitions. In the short-pulse limit, the time-integrated diffracted signal is given by[16]

$$I(T)\simeq\Theta(T)\exp(-4\gamma T)\int_{-T}^{\infty}dt[1+\cos(\Delta E t)]\times\exp(-2\gamma t-t^2\sigma^2/2), \quad (9)$$

where $\gamma=T_2^{-1}$ is the homogeneous linewidth. For $\sigma\rightarrow\infty$ (strong inhomogeneous broadening), this expression reduces to

$$\lim_{\sigma\rightarrow\infty} I(T)\simeq\Theta(T)\exp(-4\gamma T) \tag{10}$$

while for $\sigma\rightarrow 0$ (homogeneous broadening only)

$$\lim_{\sigma\rightarrow 0} I(T)\simeq\Theta(T)\exp(-2\gamma T)\times\left[1+\frac{\cos(\Delta E T)+(\Delta E/2\gamma)\sin(\Delta E T)}{1+(\Delta E/2\gamma)^2}\right]. \tag{11}$$

As discussed above, strong inhomogeneous broadening suppresses quantum beats in this case, opposite to the three-level case [Eq. (7)]. However, even for a homogeneously broadened system, the modulation of the signal is never complete, again in contrast to the three-level system result Eq. (7).

Figure 13 shows the results of a numerical evaluation of Eq. (10) for the following parameters: $\Delta E=0$ meV, $\gamma=0.21$ meV, $\sigma=1.80$ meV (sample B, dashed line); and $\Delta E=3.1$ meV, $\gamma=0.21$ meV, $\sigma=0.20$ meV (sample A, solid line). The simple theory reproduces the experimental decay remarkably well, except around zero time delay, where the approximation of δ-shaped pulses leads to larger deviations. However, the inhomogeneous broadening of the individual exciton peaks for sample A has to be chosen smaller than the experimental value ($\sigma_{\text{theor}}=0.2$ meV, $\sigma_{\text{expt}}\simeq 0.5$ meV). Theoretical curves with $\sigma_{\text{theor}}=0.5$ meV still show quantum beats with the same period, but the beats are less pronounced compared to the experimental result at large delays. The reason for this difference is not understood at present. One reason might be that the broadening mechanism of the individual lines is not known. Similarly, it is not clear why the oscillation period corresponds to a splitting of 3.1 meV, whereas the experimentally observed splitting in the spectrum is only 2.7 meV. This might be partially explained by the fact that the individual lines are not well separated enough to yield a reliable value for the splitting.

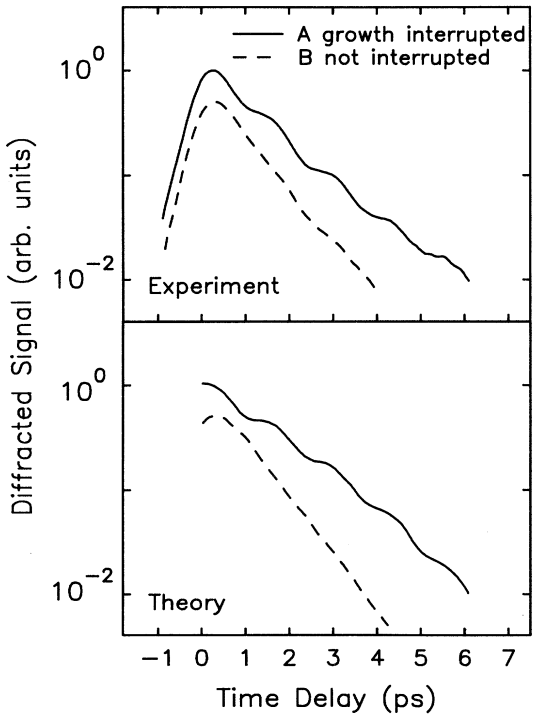

FIG. 13. Predictions of a simple two-level model for the four-wave-mixing line shapes of sample B with a single exciton line (dashed line) and sample A with split exciton lines (solid line) (bottom), compared to the experimental results (top).

D. Quantum beats between bound and free excitons

Up to now, we have reported about the properties of free excitons. In many bulk semiconductors, the luminescence shows not only transitions due to free excitons, but also transitions of excitons bound to impurities. Similarly, bound excitons have also been observed in semiconductors QW's.[53] The splitting between the free and bound excitons is about 1–2 meV, i.e., two separate lines can be observed if the sample is of sufficiently high quality. In samples where the free exciton lines are split due to a spatially varying confinement, the transition corresponding to each well thickness can be accompanied by a bound exciton transition.

In the experiment where we observed beats between lh and hh excitons, the trace for excitation of the hh transition alone (dashed line in Fig. 8) shows also a weak modulation. A more careful study of this modulation reveals a strong dependence of the modulation depth on the location of the laser spot on the sample. Figure 14 shows the diffracted signal for a sample spot where the oscillation is most pronounced. The period of the oscillation is about 2.8 ps, which would correspond to a splitting of 1.47 meV. The luminescence spectrum of the sample shows a free exciton peak and a second transition with

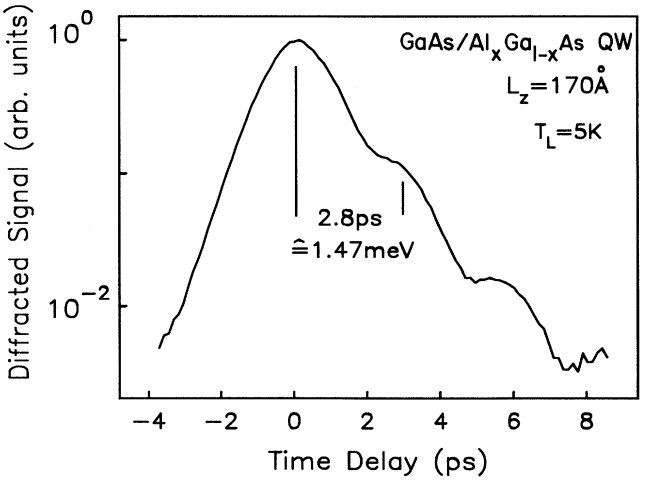

FIG. 14. Quantum beats between free and bound excitons observed in the 170-Å quantum-well sample.

about 1.4 meV lower energy. Temperature- and density-dependent measurements clearly identify the second transition as due to bound excitons. A detailed study of the temperature and density dependence of the FWM signal confirms that the observed modulation is due to quantum beats between bound and free excitons. Details are published elsewhere.[18] Recently, quantum beats between excitons bound to stacking faults in BI_3 have been observed using the FWM technique.[40]

IV. CONCLUSIONS

Degenerate four-wave mixing has been frequently used to study the *dephasing* of electronic transitions in semiconductors. Here, we have reported various results about the properties and interactions of excitons in their *coherent* state. One importance result is that the commonly used picture of excitons as noninteracting two-level systems for analyzing such experiments has to be refined: The *coherent interaction* of excitons leads to important changes of the line shape in time-resolved four-wave-mixing experiments. These observations are in agreement with microscopic theories of excitons. The polarization interaction effects, however, are not limited to excitons in semiconductors, but should be present in virtually all media. It should be very interesting to reconsider earlier wave-mixing measurements in other systems and prove these predictions. Furthermore, more theoretical work is needed to understand the microscopic nature of this effect in other media.

The investigation of quantum wells with several energetically close exciton transitions has demonstrated that quantum beats are observable in solids, despite the broad energy bands which lead to a fast decay of the polarization for the excitation of free carriers. Both interference of independent two-level-like systems and interference in a three-level-like system can be observed. The observation of quantum beats for split levels in coupled wells has also allowed the demonstration of the coherent oscillations of a wave packet in such structures, as reportedly recently.[12]

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