Subpicosecond photon-echo spectroscopy on GaAs/AlAs short-period superlattices

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We present subpicosecond time-resolved four-wave-mixing studies on GaAs/AlAs short-period superlattices having different miniband widths. Photon echoes are observed as a consequence of the inhomogeneous broadening of the excitonic resonance. In addition, the photon-echo signal exhibits periodic modulations as a function of the time delay between the excitation laser pulses. Free excitons and excitons bound to neutral acceptors are identified as the underlying three-level system leading to the observed photon-echo quantum beats. From the beat period we can directly infer the dissociation energy of the bound exciton. In accordance with theory, we find that the dissociation energy decreases with increasing miniband width, i.e., when the excitons become less confined. In addition, we find that the dephasing rate of bound excitons is about one order of magnitude less than that of free excitons and is mainly determined by energy relaxation.

I. INTRODUCTION

The optical properties associated with excitonic transitions in semiconductor microstructures like quantum wells and superlattices have been studied extensively over the last years by means of linear and nonlinear spectroscopy.[1,2] These studies have revealed pronounced differences between the behavior of excitons in bulk and quantum-well semiconductors with regard to binding energy, oscillator strength, screening, bleaching, electric-field effects, and relaxation dynamics.

Yet, in linear optical spectroscopy, the intrinsic properties of excitons in quantum structures are often obscured by inhomogeneous broadening inherent to these microstructures. One of the most prominent examples in this respect is the inhomogeneous linewidth broadening of exciton transitions in quantum wells and superlattices due to fluctuations of the thickness of the respective semiconductor layer by one atomic monolayer. Linear optical spectroscopy in the frequency domain is then unable to probe the intrinsic properties of the excitation. In particular, different excitonic states and their respective homogeneous widths, which are determined by scattering, generally cannot be resolved. This is the case where the unique power of coherent nonlinear optical spectroscopy in the time domain comes to pay. Yet, in these experiments a temporal resolution better than the inverse of the homogeneous linewidth is required. In the case of semiconductors, this means that ultrafast laser spectroscopy with time resolution in the picosecond and femtosecond regime is necessary.

Initially, transient four-wave-mixing (FWM) experiments have been primarily used to investigate the dephasing rate, i.e., the homogeneous part of the inhomogeneously broadened linewidth of excitons in $GaAs/Al_xGa_{1-x}As$ quantum wells.[3,4] Whereas the time constant T_2 for the loss of phase coherence amounts to about 7 ps for resonantly excited heavy-hole (hh) excitons, it is about three orders of magnitude shorter for energetically higher band-to-band transitions.[5] In these transient FWM studies, the diffracted signal had not been time resolved, i.e., it could not be decided whether the nonlinear signal was emitted as a photon echo, as expected for inhomogeneously broadened transitions, or as a free-induction decay, as expected for homogeneously broadened transitions.[6] However, this information is important in order to precisely extract the dephasing time T_2 from the decay of the transient FWM curve, i.e., to determine the homogeneous and inhomogeneous parts of the overall linewidth.

By time resolving the FWM signal using cross-correlation techniques, Schultheis, Sturge, and Hegarty[7] observed a photon-echo behavior from two-dimensional excitons in $GaAs/Al_xGa_{1-x}As$ quantum wells with a time resolution of about 13 ps, which is, however, longer than the respective dephasing time T_2. Noll *et al.*[8] could time resolve the photon echo from localized excitons in CdS_xSe_{1-x} mixed crystals by using a streak camera with a time resolution of about 20 ps, since dephasing times up to 500 ps are observed for this system. Recently, Webb, Cundiff, and Steel[9] found dephasing times as long as 70 ps when exciting in the low-energy part of the lowest excitonic resonance of a $GaAs/Al_xGa_{1-x}As$ multiple quantum well (MQW) and using low laser intensities. Time-resolved measurements on a picosecond time scale showed that the diffracted signal consists of both a free-induction

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decay as well as a photon echo. The authors attribute the photon echo to excitons localized by interface roughness, whereas the origin of the free-induction decay signal has remained unclear.

Time-resolved nonlinear optical spectroscopy is not only able to determine the homogeneous linewidth of inhomogeneously broadened transitions, but, in principle, can also resolve distinct transitions, which are hidden under the inhomogeneous broadening and thus cannot be seen in linear optical spectra. In recent years, several groups observed quantum beats and polarization beats[10] by applying transient coherent spectroscopy like resonance fluorescence[11] as well as transient (but not time-resolved) FWM (Refs. 12–16) on different semiconductor systems. In each of these papers, the observed beat period T_B could always be compared and thus easily identified with the energetic splitting of some distinct transitions already observable in the corresponding linear optical spectra. Consequently, transient quantum-beat spectroscopy is now well established in semiconductors and can be applied to study semiconductor systems, where closely spaced energetic transitions cannot be resolved otherwise.

In this paper we report subpicosecond time-resolved FWM studies on GaAs/AlAs short-period superlattices (SPS), where the excitonic transitions are considerably broadened due to potential fluctuations arising from interface roughness. The temporal resolution of photon echoes allows one to determine the homogeneous as well as the inhomogeneous broadening of the excitonic resonance. From the occurrence of quantum beats in the photon-echo intensity we can directly infer that a three-level system is hidden under the unstructured broad exciton emission band. These quantum beats originate from the simultaneous coherent excitation of excitons localized in potential fluctuations and excitons bound to acceptor impurities. We find that the dissociation energy of the bound exciton decreases with increasing miniband width as expected from theory. In addition, we have performed three-pulse transient FWM experiments using picosecond pulses which show that the dephasing time T_2 of bound excitons is about ten times longer than that of free excitons and is determined by spectral diffusion.

II. EXPERIMENT

We have studied two GaAs/AlAs SPS samples consisting of 100 periods of (i) 41-Å GaAs and 30-Å AlAs (sample A) and (ii) 42-Å GaAs and 14-Å AlAs (sample B). These SPS were grown by molecular-beam epitaxy and the individual layer thicknesses have been determined by double-crystal x-ray diffraction. The samples are slightly p doped due to a carbon concentration of about 1×10^{16} cm^{-3}. Both samples have been further characterized by standard low-temperature (T=5 K) absorption and photoluminescence measurements.

We have used a tunable mode-locked Ti-sapphire laser generating bandwidth-limited pulses with 120-fs duration at full width with half maximum (FWHM) at a repetition rate of 80 MHz as an excitation source for the subpicosecond time-resolved photon-echo spectroscopy. The time-resolved FWM experiments are performed in the two-beam configuration, which in the case of inhomogeneously broadened transitions corresponds to a spontaneous photon-echo experiment.[6] This configuration is schematically illustrated in Fig. 1. The sample is excited by two subsequent laser pulses with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$. The coherent nonlinear signal emitted into the direction $2\mathbf{k}_2 - \mathbf{k}_1$ can be detected either time integrated by measuring the FWM signal with a photomultiplier (PM1) as a function of the time delay τ between the pump pulses or time resolved by up-converting the FWM signal with a third reference pulse in a 1-mm LiB_3O_5 crystal. For a certain time delay τ between the pump pulses, the up-converted signal is monitored by a second photomultiplier (PM2) as a function of the time delay t of the reference pulse. This time resolved FWM technique provides a temporal resolution of about 120 fs. It is important to note that the pulse widths of the pump pulses behind the cryostat of about 140 fs are longer than the pulse width of the reference pulse of 120 fs. This is due to the fact that the pump pulses pass through more dispersive media like lenses, attenuators, and cryostat windows as compared to the reference beam and thus exhibit a larger chirp.

In addition, we have performed three-pulse FWM experiments using a synchronously pumped dye-laser system, which provides laser pulses of about 1-ps duration. In this three-beam configuration, stimulated photon echoes are created provided that the inhomogeneous broadening of the excitonic transition is sufficiently large.[8] These experiments not only allow the determination of the dephasing time T_2 but, in addition, provide quantitative information about energy relaxation rates (spectral diffusion).[17–19] All experiments are performed at a sample temperature of T=5 K.

III. LINEAR OPTICAL PROPERTIES

The low-temperature absorption and photoluminescence spectra of sample A are depicted in Fig. 2(a). The absorption peak at 1.705 eV corresponds to the n=1 hh exciton transition. The absorption resonance shows a tail to lower energies indicating pronounced inhomoge-

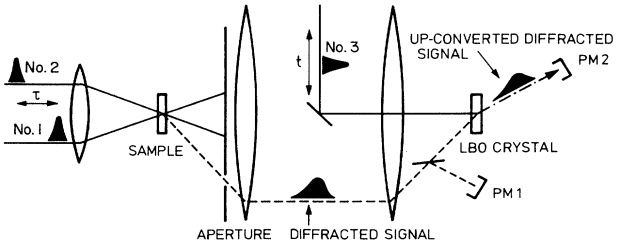

FIG. 1. Experimental setup for the subpicosecond photon-echo experiment. The diffracted FWM signal can be detected either time integrated by measuring the overall intensity as a function of the time delay τ between the two excitation pulses with a photomultiplier PM1 or time resolved by measuring the up-converted diffracted signal at any given τ as a function of the real time t with PM2.

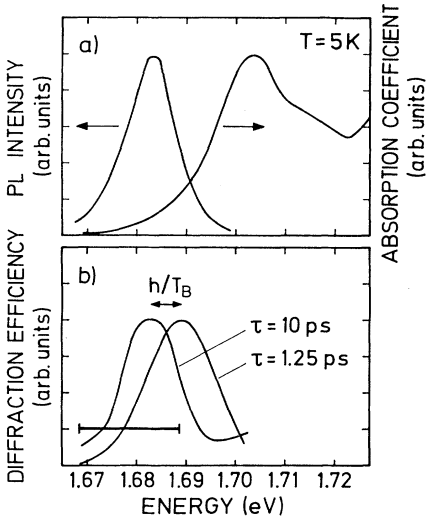

FIG. 2. (a) Low-temperature absorption and photoluminescence spectra of the GaAs/AlAs SPS sample A. (b) Efficiency of the diffracted FWM signal as a function of photon energy for different time delays τ between the pump laser pulses of 1.25 and 10 ps. The horizontal arrow shows the energetic spacing h/T_B obtained from the experimentally determined value of the quantum-beat period T_B.

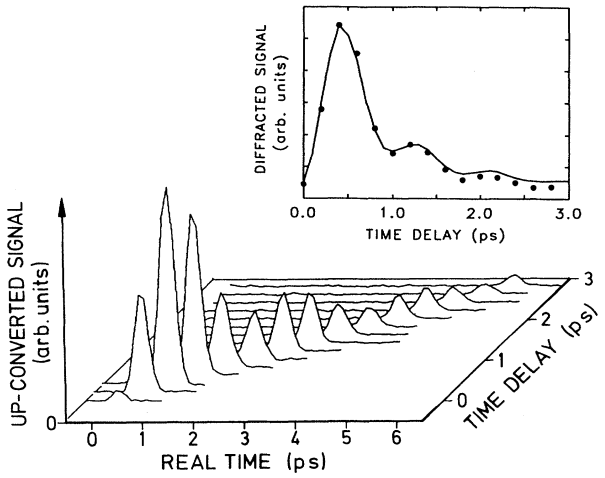

FIG. 3. Up-converted FWM signal as a function of both the time delay τ between the pump laser pulses and the time position t of the third reference pulse. A beating photon echo along the $t = 2\tau$ diagonal is clearly observed. The corresponding time-integrated measurement is shown as the solid line in the inset. The points in the inset represent the result of a numerical integration of the measured time-resolved photon echo over t.

neous broadening due to intrawell and interwell thickness fluctuations.[20,21] The photoluminescence spectrum is located at the low-energy side of the absorption tail and shows a broad unstructured band with an inhomogeneous broadening of about 12 meV (FWHM). The Stokes shift between the maximum of the hh exciton and the luminescence maximum amounts to 22 meV. The hh exciton peak of sample B is at 1.695 eV, otherwise the absorption and photoluminescence spectra are very similar to those of sample A. Calculations based on the Kronig-Penney model[22] show that a change of the GaAs layer thickness by only one monolayer induces energetic shifts of the n=1 hh excitonic transition of about 19 and 16 meV for samples A and B, respectively. This demonstrates that the pronounced inhomogeneous broadening as well as the large Stokes shift are mainly a consequence of intra- and interwell thickness fluctuations in these GaAs/AlAs short-period superlattices.

IV. TIME-RESOLVED FOUR-WAVE MIXING

We now shall discuss the results of the subpicosecond two-beam FWM experiments with time-resolved detection of the diffracted FWM signal (taken with PM2 in Fig. 1). The horizontal bar in Fig. 2(b) indicates the spectral position and the linewidth (FWHM) of the excitation laser pulses. In Fig. 3 the coherent FWM signal for sample A is depicted as a function of both the time delay τ between the excitation laser pulses as well as the "real" time t, which is the time delay of the third reference pulse with respect to the first pump pulse. We clearly observe that the FWM signal is emitted as a spontaneous photon echo at a time $t = 2\tau$, which proves the inhomogeneous nature of the exciton transition.[6,23] The observation of a clear photon-echo signal further implies that the inhomogeneously broadened excitonic transitions can be viewed as an ensemble of noninteracting two-level systems, where, e.g., dipole-dipole interactions are neglected. In this case the temporal width of this spontaneous photon echo is determined by the inverse of the inhomogeneous excitonic broadening, allowing an independent and precise determination of the inhomogeneous broadening provided the laser spectrum is broader than the inhomogeneous width.

In Fig. 4, the up-converted photon-echo signal is plotted as a solid line for one particular time delay τ between the pump pulses. The dashed curve in Fig. 4 represents the cross correlation of the third reference pulse with one pump pulse. Obviously, the width of the up-converted photon echo is larger than that of the cross correlation with the pump pulses. Accordingly, the inhomogeneous broadening can be directly determined from the width of the echo. Knowing the temporal duration of the pump and reference pulses, we can deconvolute the cross-correlated FWM signal. We then obtain a temporal width of 200 fs (FWHM) for the photon echo. Assuming a Gaussian inhomogeneous broadening with spectral width ΔE for the excitonic transition, we have computed the up-converted photon-echo signal taking into account the experimentally determined finite pulse widths of the pump and reference pulses. The dotted curve is a result of such a calculation. It represents the best data fit assuming an inhomogeneous broadening of ΔE=25 meV. This value is in good agreement with the experimentally determined Stokes shift of about 22 meV for the photoluminescence spectrum. However, it is about twice as large as the spectral linewidth of 12 meV observed in the lumi-

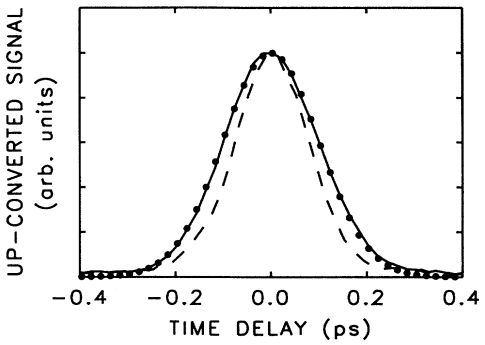

FIG. 4. Experimental cross-correlation curves of the photon echo (solid line) and of one pump pulse (dashed line). The dotted line represents a calculation of the up-converted FWM signal assuming an inhomogeneous broadening of ΔE=25 meV.

nescence spectrum in Fig. 2(a) showing that appreciable spectral relaxation into energetically lower-lying states takes place before the excitons recombine.

The time-resolved photon-echo spectrum of Fig. 3 shows an additional striking feature, namely a periodic modulation of the overall intensity of the echo along the $t = 2\tau$ line. This pronounced beating is also seen when the FWM signal is measured time integrated by using PM1 for detection. The respective transient FWM trace is shown as a solid line in the inset of Fig. 3. The dots represent the result of a numerical integration with respect to the t axis of the time-resolved photon-echo signal and the obtained curve coincides with the experimentally determined transient FWM curve as expected. The time period T_B for the observed beating amounts to about 800 fs for this particular sample. The FWM curves of sample B (not shown here) also exhibit quantum beats; however, in this case the time period T_B is determined to about 1.1 ps. The fact that the photon-echo intensity at $t = 2\tau$ is beating as a function of time delay τ excludes the possibility that the beating arises from the interference of two independent sets of inhomogeneously broadened two-level systems, i.e., that we observe a so-called polarization interference.[24] Opposite to the case of a three-level system, the third-order dipole moments of individual two-level systems always rephase at $t = 2\tau$, i.e., a beating of the echo at $t = 2\tau$ is not possible in the case of independent two-level systems.

Consequently, the underlying electronic structure responsible for the periodic modulation of the photon-echo signal must correspond to a three-level system. From the time period T_B we can infer the energetic splitting $\Delta E = h/T_B$ of the two excited states of the three-level system. We thus obtain ΔE=5.2 and 3.8 meV for the SPS samples A and B, respectively. The two distinct transitions energetically separated by ΔE, which are coherently excited in the FWM experiment, are not resolved in the linear optical spectra of Fig. 2(a). Thus the identification is more subtle. However, knowing the measured $\Delta E = h/T_B$ values for the two SPS structures, we can unambiguously exclude the following potential candidates.

Light- and heavy-hole excitons are separated energetically by 60 and 45 meV for samples A and B, respectively.

In principle, a three-level beating between the 1s excitonic ground state and the 2s excitonic excited state should be possible. However, for the present GaAs/AlAs SPS samples, exciton binding energies of about 16 meV are expected,[25–27] leading to a 1s-2s splitting of about 12 meV.

The characteristic feature of semiconductor superlattices is the formation of minibands for electronic motion in growth direction. The Coulomb interaction then leads to excitonic transitions at the lower and upper edges of the miniband.[28,29] Quantum beats between these M_0 and M_1 excitons should also be possible. However, Kronig-Penney band-structure calculations show that the sums of miniband widths for electrons in the conduction band and heavy holes in the valence band amount to $\Delta = 1$ and 21 meV for samples A and B, respectively.

Recently, quantum beats have been observed between exciton states and biexciton states excited by resonant two-photon absorption.[30] The observed time period T_B=3.7 ps corresponds to an energetic splitting of 1.1 meV and represents the binding energy of the biexciton in a GaAs quantum well. Since the energetic splittings observed in the present GaAs/AlAs superlattice structures are much larger, we can also exclude that biexcitonic states are responsible for the quantum beating of the photon-echo intensity.

In the following we demonstrate that we can attribute the quantum beats observed for the GaAs/AlAs SPS samples to the simultaneous coherent excitation of "free" excitons localized by energetic disorder and excitons bound to neutral acceptors (carbon). First of all, the observed energetic intervals $\Delta E = h/T_B$ coincide with typical dissociation energies E_D of acceptor bound excitons in GaAs quantum wells. Miller *et al.*[31] determined the dissociation energy E_D for acceptor bound excitons in a 46-Å-wide GaAs well to be 6.5 meV by performing photoluminescence studies. The results were in accordance with Hayne's rule[32] in the form $E_D = 0.133E(A^0)$, where $E(A^0)$ is the ionization energy of the acceptor. In addition, it was found that $E(A^0)$ and thus also E_D decrease with increasing well width[31,33] as predicted theoretically.[34] The dissociation energy E_D decreases from 6.5 meV for a 46-Å-wide quantum well to the three-dimensional value of about 2.9 meV for thick GaAs wells. From the beating period T_B, we find energetic intervals of ΔE = 5.2 meV for the SPS sample A and ΔE = 3.8 meV for sample B. Taking into consideration that the exact ionization energy of an impurity depends also on its position within the quantum well, the beating frequency found for sample A agrees well with the E_D value expected for that particular GaAs quantum-well width. The lower beating frequency found in case of the SPS sample B can be easily explained by the fact that the dissociation energy E_D decreases when going from a two-dimensional "multiple-quantum-well" structure (sample A with an overall miniband width of Δ = 1 meV) to a

more three-dimensional superlattice structure (sample B with an overall miniband width of $\Delta = 21$ meV).

In Fig. 5 the time-integrated FWM signal is plotted versus the time delay τ between the pumping laser pulses for a slightly lower photon energy as compared to the situation in Fig. 2 and on a longer, picosecond time scale. It is seen that after the damping of the quantum beats has occurred a long-living and unstructured FWM signal survives. In order to decide whether the long-living component is due to free excitons or due to excitons bound to acceptors, we have measured the diffracted FWM intensity as a function of photon energy for a time delay $\tau = 10$ ps and for a time delay $\tau = 1.25$ ps, where the modulated signal has a maximum. The results are shown in Fig. 2(b). For $\tau = 10$ ps the FWM signal has its maximum at low photon energy, namely at the spectral position corresponding to the peak of the luminescence spectrum. Consequently, we attribute this long-living FWM signal to optically excited excitons bound to acceptors. The FWM spectrum for a time delay of $\tau = 1.25$ ps also peaks in the low-energy part of the n=1 hh exciton absorption resonance, however, at slightly higher energy. The horizontal arrow labeled h/T_B in Fig. 2(b) indicates the experimentally determined beating frequency. It coincides with the energetic spacing between the maxima of the two FWM spectra. This means that the FWM spectrum centered at higher photon energy is most probably due to "free" excitons, which are localized by potential fluctuations induced by interface roughness. From the time constant τ_d for the damping of the quantum beats, we can deduce a dephasing time of $T_2 = 4\tau_d = 2$ ps for the free excitons, i.e., the dephasing rate is higher than previously reported for free excitons in undoped quantum wells.[3,4] We attribute the increased dephasing rate to inelastic scattering, i.e., trapping of free excitons into lower-lying bound exciton states. The reduced dephasing of the excitons bound to acceptors and their spectral diffusion will be discussed in Sec. V.

Finally, we point out that the optical excitation of bound- and free-exciton states indeed represent a three-level system, if the acceptor impurity site lies within the coherence volume[35] of the free exciton. Then both transitions have the same ground state in common, which is the nonexcited crystal state. The transition energy of the free exciton actually marks the continuum edge of the bound exciton. However, at low enough impurity concentration the mean distance between impurities might be larger than the diameter of the excitonic coherence volume. This should lead to a mixture of three-level systems and isolated free-exciton two-level systems and thus to a mixture of quantum beats and polarization interferences. For the present samples, an impurity concentration of about 1×10^{16} cm^{-3} implies that there is one impurity atom per sphere of radius 300 Å. This concentration is sufficient to exclude the presence of any free-exciton two-level systems, which are not coupled to bound excitons.

V. PICOSECOND FOUR-WAVE MIXING

We have argued that the long-living FWM part in Fig. 5 is due to a reduced dephasing rate of bound excitons. From the decay we deduce a dephasing time $T_2 = 4\tau_d = 18$ ps. In order to study the phase relaxation of bound excitons without exciting free excitons at higher energy we have used the spectrally narrow picosecond laser pulses from the synchronously pumped dye laser. In Fig. 6 the results of transient two-beam FWM experiments are depicted for the SPS samples A and B. The respective photon energies have been chosen in the low-energy part of the hh excitonic resonances, i.e., at the spectral positions of the bound excitons. The induced carrier density is estimated to about 5×10^8 cm^{-2}. By determining the time constants τ_d for the decay of the FWM curves we obtain dephasing times $T_2 = 4\tau_d$ of 48 and 80 ps for samples A and B, respectively.

Figure 7 shows the experimentally determined dephasing times T_2 for the SPS sample B as a function of photon energy. For comparison, the photoluminescence spectrum, which is due to the recombination of bound excitons, is also shown. A drastic increase of the dephasing times is found for decreasing photon energy. The T_2 times increase to about 130 ps for photon energies at the low side of the photoluminescence spectrum. The same qualitative result is found for sample A. Accordingly, the dephasing time $T_2 = 18$ ps obtained for bound

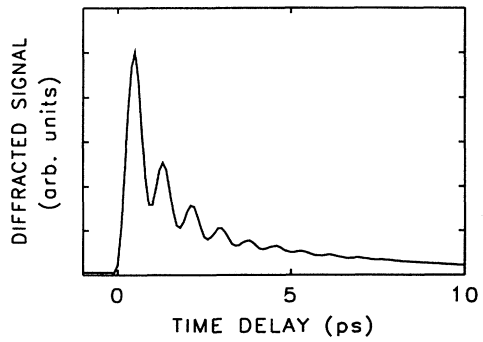

FIG. 5. FWM signal measured time integrated as a function of time delay τ between the pump pulses for the SPS sample A.

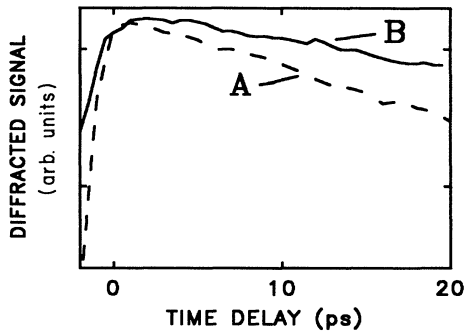

FIG. 6. Temporal evolution of the two-beam FWM signals for samples A and B using picosecond laser pulses. The respective photon energy is chosen to excite dominantly bound excitons.

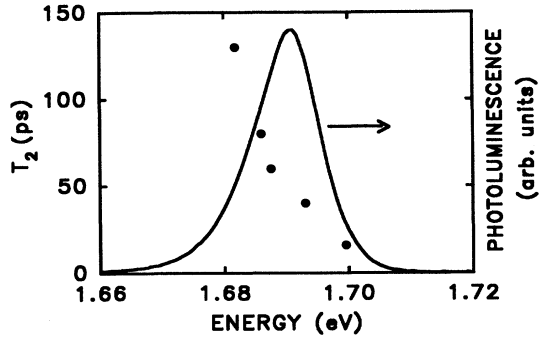

FIG. 7. Experimentally determined dephasing times T_2 (dots) vs photon energy within the spectral range corresponding to the photoluminescence spectrum.

excitons from Fig. 5, where the spectrum of the 120-fs laser pulse encompasses the whole energetic distribution of bound excitons, represents an average value. The pronounced dependence of the dephasing rate on photon energy already suggests that the dephasing is dominated by spectral diffusion within the energetic distribution of the bound excitons. To be more quantitative, we have performed three-beam FWM experiments, which provide a quantitative access to the nature of the phase-breaking scattering processes.

As schematically shown in the inset of Fig. 7, two pump laser pulses with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ impinge onto the sample with time delay τ. Additionally, a third laser pulse with wave vector $\mathbf{k}_3$ then interacts with the optically induced excitation a time period T after the *second* laser pulse. As a consequence, a FWM signal is diffracted into the phase-matching directions $\mathbf{k}_4 = \mathbf{k}_3 \pm (\mathbf{k}_2 - \mathbf{k}_1)$.

For $\tau \neq 0$, this three-beam FWM experiment is equivalent to a stimulated photon-echo experiment.[17–19] In this experiment, the first laser pulse arriving at $t = 0$ excites different two-level systems within the spectral width of the laser pulse. As in the case of the two-beam FWM experiment, the corresponding dipole moments initially are in phase but come out of phase due to the different transition frequencies of the two-level systems. In order to understand the diffraction of the third laser pulse, one has to consider the effect of the second laser pulse, which is to transform the dipole moments into a population ΔN at $t = \tau$. The third laser pulse then impinges on the sample at $t = \tau + T$ and transforms the population ΔN "back" into third-order dipole moments, which then rephase at $t = 2\tau + T$ leading to a macroscopic polarization acting as the source for the stimulated photon echo. However, the rephasing between $t = \tau + T$ and $t = 2\tau + T$ can only take place if the frequencies of the initially (at $t = 0$) excited two-level systems have not changed during the time period between $t = \tau$ and $t = \tau + T$, i.e., if no spectral diffusion has taken place. In other words, if energy-relaxation processes take place between the arrival of the second and third laser pulses, the intensity of the diffracted stimulated photon echo will decrease. Accordingly, in a three-beam experiment the decay of the FWM signal as a function of T for $\tau \neq 0$ is not only determined by recombination (T_1), but also by inelastic scattering processes (T_3). In Fig. 8 the temporal evolution of the FWM signal diffracted into direction $\mathbf{k}_4$ is shown for sample B as a function of T for a fixed time delay $\tau = 15$ ps between the pump laser pulses. The photon energy is chosen to be 1.687 eV, i.e., at a spectral position where spectral diffusion is expected. The time constant τ_d for the decay of the diffracted FWM intensity amounts to 25 ps. This implies that the corresponding decay time for the third-order polarization decays with a time constant $T_{\text{pop}} = 50$ ps and is given by

$$\frac{1}{T_{\text{pop}}} = \frac{1}{T_1} + \frac{1}{T_3}. \tag{1}$$

We have performed a pump-probe experiment (not shown here) in order to determine the recombination lifetime T_1. We find $T_1 = 150$ ps and thus obtain a time constant $T_3 = 75$ ps for the spectral diffusion of the bound excitons excited at this particular photon energy. This means that the decay of the intensity of the stimulated photon echo is mainly determined by energy relaxation and not by recombination.

The dephasing time T_2 can be written as

$$\frac{1}{T_2} = \frac{1}{2T_{\text{pop}}} + \frac{1}{T_{\text{el}}}, \tag{2}$$

where T_{el} is the time constant for all elastic-scattering processes. With the experimentally determined values of $T_2 = 60$ ps and $T_{\text{pop}} = 50$ ps we obtain $T_{\text{el}} = 150$ ps. We can thus conclude that the dephasing rate of bound excitons in the upper part of the luminescence spectrum is neither governed by recombination (T_1) nor by elastic scattering processes (T_{el}), but is mainly given by inelastic scattering (T_3). In particular, we can exclude "elastic" dephasing due to disorder as an important contribution, which implies that the disorder potential cannot be strong on a spatial scale shorter than the exciton Bohr radius.[36,19] In other words, the relative electron-hole motion is not disturbed by the dis-

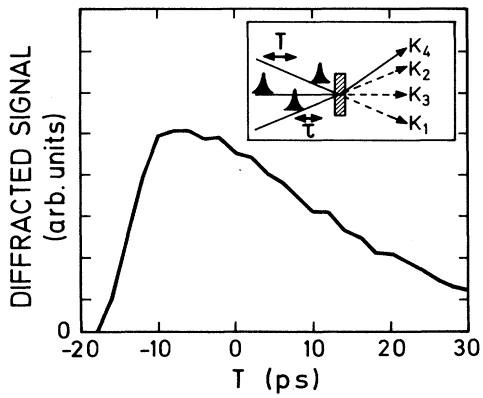

FIG. 8. Temporal evolution of the three-beam FWM signal for sample B as a function of the time delay T of the third laser pulse and for a time delay $\tau = 15$ ps between the first two laser pulses. The experimental setup is illustrated in the inset.

order potential. Altogether, we conclude that phonon-assisted energy-relaxation processes are the most important phase-breaking mechanisms.

To avoid confusion, we would like to point out that our results cannot be directly compared to the FWM data obtained by Webb, Cundiff, and Steel,[9] since they have studied multiple-quantum-well samples which were not carbon doped.

VI. CONCLUSIONS

We have presented transient and time-resolved FWM studies on GaAs/AlAs short-period superlattice structures on a subpicosecond and picosecond time scale. The observed photon-echo behavior allows us to study the homogeneous as well as the inhomogeneous broadening of the excitonic transitions. In addition, the beating of the echo signal shows that a three-level system is hidden underneath the unstructured linear absorption spectrum. We attribute this beating to the simultaneous coherent excitation of excitons bound to acceptor states and excitons localized by interface roughness. From the beat period we directly determine the dissociation energy of the bound excitons. In accordance with theory, we find that the dissociation energy decreases with increasing miniband width, i.e., when the electronic system loses its two-dimensional character. The dephasing rate for the bound excitons is found to be about ten times smaller than the dephasing rate of localized excitons and is determined by energy relaxation due to phonon-assisted phase-breaking processes. In particular, we conclude that the disorder potential on a short spatial scale is not strong enough to disturb the relative electron-hole Coulomb motion considerably.

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*Deceased.

[1] S. Schmitt-Rink, D.S. Chemla, and D.A.B. Miller, Adv. Phys. **38**, 89 (1989), and references therein.

[2] E.O. Göbel and K. Ploog, Prog. Quantum Electron. **14**, 289 (1990), and references therein.

[3] L. Schultheis, J. Kuhl, A. Honold, and C.W. Tu, Phys. Rev. Lett. **57**, 1635 (1986); **57**, 1797 (1986).

[4] A. Honold, L. Schultheis, J. Kuhl, and C.W. Tu, Phys. Rev. B **40**, 6442 (1989).

[5] P.C. Becker, H.L. Fragnito, C.H. Brito Cruz, R.L. Fork, J.E. Cunningham, J.E. Henry, and C.V. Shank, Phys. Rev. Lett. **61**, 1647 (1988).

[6] T. Yajima and Y. Taira, J. Phys. Soc. Jpn. **47**, 1620 (1979).

[7] L. Schultheis, M.D. Sturge, and J. Hegarty, Appl. Phys. Lett. **47**, 995 (1985).

[8] G. Noll, U. Siegner, S.G. Shevel, and E.O. Göbel, Phys. Rev. Lett. **64**, 792 (1990).

[9] M.D. Webb, S.T. Cundiff, and D.G. Steel, Phys. Rev. Lett. **66**, 934 (1991).

[10] The distinction between quantum beats and polarization beats refers to the electronic level structure of the two transitions involved: In the case of quantum beats the underlying electronic system is equivalent to a three-level system; instead, polarization beats are attributed to two independent two-level systems.

[11] V. Langer, H. Stolz, and W. van der Osten, Phys. Rev. Lett. **64**, 854 (1990); H. Stolz, V. Langer, E. Schreiber, S. Permogorov, and W. van der Osten, *ibid.* **67**, 679 (1991).

[12] E.O. Göbel, K. Leo, T.C. Damen, J. Shah, S. Schmitt-Rink, W. Schäfer, J.F. Müller, and K. Köhler, Phys. Rev. Lett. **64**, 1801 (1990).

[13] K. Leo, T.C. Damen, J. Shah, E.O. Göbel, and K. Köhler, Appl. Phys. Lett. **57**, 19 (1990).

[14] B.F. Feuerbacher, J. Kuhl, R. Eccleston, and K. Ploog, Solid State Commun. **74**, 1279 (1990).

[15] K. Leo, T.C. Damen, J. Shah, and K. Köhler, Phys. Rev. B **42**, 11 359 (1990).

[16] S. Bar-Ad and I. Bar-Joseph, Phys. Rev. Lett. **66**, 2491 (1991).

[17] A.M. Weiner, S. de Silvestri, and E.P. Ippen, J. Opt. Soc. Am. B **2**, 654 (1985).

[18] M.D. Webb, S.T. Cundiff, and D.G. Steel, Phys. Rev. B **43**, 12 658 (1991).

[19] U. Siegner, D.Weber, E.O. Göbel, P. Thomas, H. Schwab, C. Klingshirn, J.M. Hvam, and V.G. Lyssenko, Phys. Rev. B **46**, 4564 (1992).

[20] C. Weisbuch, R.D. Miller, R. Dingle, A.C. Gossard, and W. Wiegmann, Solid State Commun. **37**, 219 (1981).

[21] P.S. Koplev, I.N. Uraltsev, A.L. Efros, D.R. Yakovlev, and A.V. Vinokurova, Fiz. Tekh. Poluprovodn. **22**, 424 (1988) [Sov. Phys. Semicond. **22**, 259 (1988)].

[22] H.S. Cho and P.R. Prucnal, Phys. Rev. B **36**, 3237 (1987).

[23] L. Allen and J.H. Eberly, *Optical Resonance and Two-Level Systems* (Dover, New York, 1975).

[24] M. Koch, J. Feldmann, G. von Plessen, E.O. Göbel, P. Thomas, and K. Köhler, Phys. Rev. Lett. (to be published).

[25] D.C. Rogers, J. Singleton, R.J. Nicholas, C.T. Foxon, and K. Woodbridge, Phys. Rev. B **34**, 4002 (1986).

[26] D.D. Smith, M. Dutta, X.C. Liu, A.F. Terzis, A. Petrou, M.W. Cole, and P.G. Newman, Phys. Rev. B **40**, 1407 (1989).

[27] L.C. Andreani and A. Pasquarello, Superlatt. Microstruct. **9**, 1 (1991).

[28] R.H. Yan, R.I. Simes, H. Ribot, L.A. Coldren, and A.C. Gossard, Appl. Phys. Lett. **54**, 1549 (1989).

[29] B. Deveaud, A. Chomette, F. Clerot, A. Regreny, R. Romestain, I.C. Maan, and G. Bastard, Phys. Rev. B **40**, 5803 (1989).

[30] D.J. Lovering, R.T. Phillips, G.J. Denton, and G.W. Smith, Phys. Rev. Lett. **68**, 1880 (1992).

[31] R.C. Miller, A.C. Gossard, W.T. Tsang, and O. Munteanu, Solid State Commun. **43**, 519 (1982).

[32] J.R. Haynes, Phys. Rev. Lett. **4**, 361 (1960).

[33] R.C. Miller, A.C. Gossard, W.T. Tsang, and O. Munteanu,

Phys. Rev. B **25**, 3871 (1982).
[34]G. Bastard, Phys. Rev. B **24**, 4714 (1981).
[35]J. Feldmann, G. Peter, E.O. Göbel, P. Dawson, K. Moore, C.T. Foxon, and R.J. Elliott, Phys. Rev. Lett. **59**, 2337 (1987).
[36]D. Bennhardt, P. Thomas, A. Weller, M. Lindberg, and S.W. Koch, Phys. Rev. B **43**, 8934 (1991).