Observation of Time-Resolved Picosecond Stimulated Photon Echoes and Free Polarization Decay in GaAs/AlGaAs Multiple Quantum Wells

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We report measurements of exciton dynamics in GaAs multiple-quantum-well structures at low temperature using three-pulse picosecond four-wave mixing. The *time-resolved measurements of the emission* show both a delayed signal (stimulated photon echo) and a prompt signal (free polarization decay) with different decay times. The results suggest that, in addition to the inhomogeneously broadened resonance, a homogeneously broadened resonance contributes to the optical response of the exciton.

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Transient nonlinear laser spectroscopy methods provide a powerful experimental tool for the study of electronic excitation in solids. In the simplest experiment, optical excitation by a short pump pulse, with wave vector $\mathbf{k}_1$, creates a nonequilibrium state of the system and modifies the absorption, which can be measured as a function of time and frequency with a second beam with wave vector $\mathbf{k}_2$. The decay rates reflect the return to equilibrium and arise from several mechanisms such as radiative decay and scattering due to interactions with other perturbers. The time evolution may be very complex. Using the language developed for simpler systems, this decay can be referred to as longitudinal relaxation. Electronic excitation by coherent optical radiation also creates a macroscopic polarization reflecting the fact that the system is in a superposition of states that give rise to the dipole. This polarization is characterized by a decay that is usually distinct from the longitudinal relaxation and can be designated transverse or dephasing relaxation, corresponding to the homogeneous linewidth in the frequency domain. In the time domain, the dephasing rate can be measured by transient four-wave mixing[1] (FWM) where two beams separated in time (with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$) create a spatial modulation that results in scattering of the incident beams in direction $2\mathbf{k}_1-\mathbf{k}_2$ or $2\mathbf{k}_2-\mathbf{k}_1$. For a homogeneously broadened system, the signal is prompt [called a free polarization decay (FPD)] with respect to the second pulse, while for an inhomogeneously broadened system, the signal emission is delayed [called a two-pulse photon echo (PE)] and the delay time is the interval between the input pulses. In this paper we examine both the longitudinal and transverse relaxation of resonantly excited excitons in GaAs/AlGaAs multiple-quantum-well (MQW) structures using a powerful extension of the above techniques based on time-separated, three-pulse excitation[2] in a backward FWM geometry.

In an ideal quantum well at low temperature, the quasi-two-dimensional excitons are delocalized. For the HH1 exciton (HH denotes heavy hole), phonon and defect scattering result in rapid dephasing [measured using transient FWM in a single high-quality QW, width $=13.5$ nm, to be of order 2 ps corresponding to the homogeneous linewidth of order 0.6 meV (Ref. 3)]. This scattering also gives rise to an initial fast decay in the longitudinal relaxation rate measurement due to scattering to exciton k values away from the initial $k=0$ (i.e., spectral diffusion, defined as the scattering of excitation at energy E to E'); a longer decay component due to exciton recombination follows the rapid decay. However, nonideal growth conditions result in interface roughness leading to inhomogeneous broadening of the absorption spectrum[4] and to localization of low-energy excitons due to fluctuations in the potential. Recent measurements of relaxation and spatial diffusion of these localized excitons in GaAs MQWs show a slower relaxation rate than for the delocalized excitons.[5,6] The rates depend on the excitation energy, increasing near the line center, suggesting the presence of a mobility edge where higher-energy excitons are delocalized. The temperature dependence of the relaxation rates of the lower-energy excitons indicates the origin is due to thermal activation to extended states[5,7] and spectral diffusion of excitons between localization sites is due to phonon-assisted migration.[8-10]

The experimental measurements presented below show this discussion is incomplete. Using transient FWM and *time resolving the emission,* we find both a FPD and a PE signal with strong spectral overlap and different decay times. The results are consistent with the interpretation that both an inhomogeneously broadened resonance and a homogeneously broadened resonance comprise the linear response of the system.

The experimental geometry is illustrated in the upper left inset of Fig. 1(a). The laser produces nearly transformed-limited pulses (1 or 8 ps) at the exciton transition wavelength ($\lambda\approx 800$ nm). Three beams with wave vectors $\mathbf{k}_1$, $\mathbf{k}_2$, and $\mathbf{k}_3$ ($=-\mathbf{k}_2$) are incident on the sample at time t_1, t_2, and t_3, respectively, producing a coherent signal in the direction $-\mathbf{k}_1$. The dephasing (longitudinal) decay rate results in the decay of the signal as a function of t_2-t_1 (t_3-t_2).

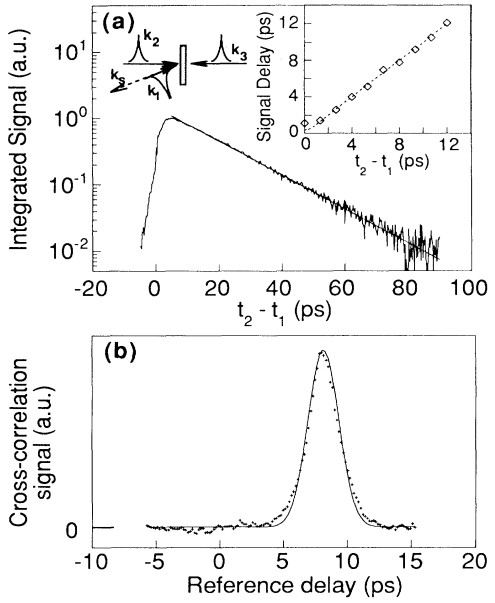

FIG. 1. (a) SPE signal as a function of delay. The solid line is a fit, corresponding to a dephasing time of 68 ps. Upper left inset: Experimental geometry. Upper right inset: Delay of the peak of the photon echo signal. The dashed line is the theoretically calculated delay, assuming a laser pulse width of 1 ps, and a phase coherence time of 68 ps. (b) The SPE signal as a function of time, measured with a 1.6-ps laser pulse width, and a corresponding theoretical fit showing that the inhomogeneous width of the resonance giving rise to the SPE is 2.5 ± 0.25 meV.

Three MBE-grown MQW samples were examined, with similar results. The data presented in this paper were obtained on a sample consisting of 65 periods of GaAs (96 Å) and $Al_{0.3}Ga_{0.7}As$ (98 Å) layers with the substrate removed. At 1.8 K, the HH1 exciton absorption linewidth is 2.35 meV, and the luminescence exhibits a small ($\sim$1 meV) Stoke's shift.

Figure 1(a) illustrates the decay of the integrated signal as a function of $t_2 - t_1$ due to dephasing at $T = 2.0$ K (for $t_3 - t_2 = 0$). The decay rate ($\gamma^{-1} = 68 \pm 2$ ps) (Ref. 11) corresponds to excitons excited 1 meV below the absorption line center, where excitons are expected to be localized, and is independent of excitation density for densities below 8×10^7 excitons/cm^{2}layer. The decay rate implies a homogeneous linewidth of 0.019 meV. This decay is slower than previously reported values,[12,13] most likely due to differences in the microscopic details of interface roughness that determines the phonon-assisted tunneling rate. The narrow homogeneous linewidth demonstrates that the linear absorption spectrum is strongly inhomogeneously broadened and that the signal is a simple PE.

The inhomogeneous broadening of the optical response is confirmed at low excitation densities by demonstrating the response is asymmetric as a function of $\tau = t_2 - t_1$ (i.e., 0 for $t_2 < t_1$) for $t_3 - t_2 > 0$ (Ref. 2) (see Ref. 14 for effects of local-field corrections in semiconductors). For $t_3 - t_2 > 0$, the coherent emission is a stimulated photon echo (SPE).[15] Time resolving the signal by cross correlating with a reference pulse directly verifies the echo. The upper right inset in Fig. 1(a) shows the linear dependence of the echo delay on τ. Numerical integration of the density matrix equations shows that the flattening of the curve near $\tau = 0$ is due to finite-pulse-width effects.

The amplitude of the SPE signal as a function of $t_2 - t_1$ provides a measure of the dephasing rate, but the *temporal evolution of the emission* [Fig. 1(b)] provides a direct measurement of the inhomogeneous broadening. In the simple case of δ-function excitation pulses and strong inhomogeneous broadening by a Gaussian distribution, the FWHM in time of the SPE signal is given by $\Delta t = 2\sqrt{2}\ln 2/\pi\Delta\nu$, where $\Delta\nu$ is the FWHM of the absorption profile. The solid curve in the figure shows the numerically determined SPE signal including finite-pulse-width effects. A best fit shows the linewidth is 2.5 ± 0.25 meV, in good agreement with the linear absorption measurement. The measurement supports the earlier claim that the absorption spectrum is completely inhomogeneously broadened. Small deviations between theory and experiment are most likely the result of a non-Gaussian absorption spectrum. It is important to note that including the wavelength dependence of the dephasing rate[12,16] in our calculations does not affect this conclusion.

The SPE technique enables measurement of lifetime dynamics of the exciton by measuring the signal amplitude as a function of $t_3 - t_2$, in contrast to the ordinary PE. The measured excitation lifetime is 34 ± 1 ps, suggesting that the dephasing time (measured as twice the excitation decay time) is due to decay of the excitation. This short lifetime is in qualitative agreement with earlier transient hole-burning measurements[17] and is clearly not due to recombination ($\tau_{rec} \approx 1$ ns), but rather to phonon-assisted migration (spectral diffusion) between localization sites of excitons localized by disorder.[9] Detailed studies of the time-integrated relaxation measurements of these excitons are presented elsewhere.[16] It is useful to note that with a large, fixed delay in the third pulse, the measurements show a sharp decrease in the phase coherence when the delay between the first and second pulses is greater than the pulse width. This is the standard signature of spectral diffusion.[2] In addition, measurement of the temperature dependence of the decay rate shows that phonon-assisted migration and thermal activation determine the relaxation.[10]

At the higher excitation densities ($> 10^9$ cm^{-2}layer, common in transient FWM), the nonlinear response shows contributions from higher-order terms (as determined by the dependence of the signal strength on exci-

tation beam intensities) and the decay of dynamics become complex. The time evolution of the signal becomes multiexponential and the exponential prefactors depend on intensity and temperature. However, the nonexponential decay of the signal as a function of the delay time t_2-t_1 is not simply the result of complex decay dynamics but rather the unexpected result that *two signals contribute to the time-integrated response.* Figure 2(a) shows the time-resolved emission at a density of 5×10^9 cm^{-2}layer. In addition to the delayed pulse corresponding to the SPE, we observe an additional prompt signal (the FPD signal) that is nearly coincident with the third incident pulse, independent of t_2-t_1. (The FPD signal is observable at this excitation density because the SPE has saturated while the FPD signal continues to increase as the cube of the input intensities. Saturation of the SPE signal is possibly the result of an increase in the dephasing rate due to exciton-exciton interactions[18] or the result that at high excitation, the number of localization sites is significantly depleted.) The physical interpretation of this result based on the usual picture of FWM is

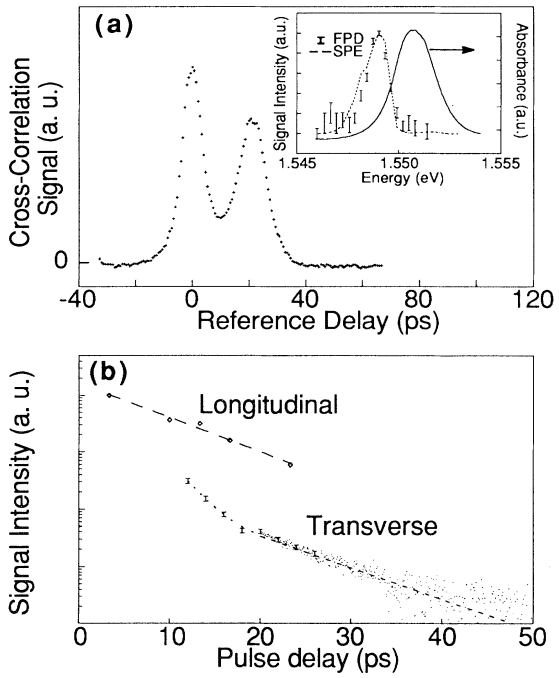

FIG. 2. (a) The cross-correlation signal showing both a SPE signal (at 20-ps delay) and FPD signal (at 0 delay), measured with an 8-ps laser pulse width. Inset: Spectral dependence of both signals. (b) The FPD signal as a function of pulse delay, measured with a 1-ps laser pulse width. The lower curve is a measurement of the dephasing decay as a function of t_2-t_1, corresponding to a two-component decay time of <5 and 16 ps. The upper curve is a measurement of the longitudinal relaxation decay as a function of t_3-t_2 corresponding to a single-component decay time of 14 ps.

that the electronic transition giving rise to the emission is comprised of an inhomogeneously broadened resonance (creating the SPE) and a homogeneously broadened resonance (creating the FPD). Such behavior has been reported in a simpler system.[19]

The spectral dependence of the two signals [Fig. 2(a), inset] was determined using the 8-ps pulse to provide adequate spectral resolution. To avoid saturation effects, the SPE spectral measurement was made at an excitation density near 5×10^7 cm^{-2}layer. t_2-t_1 was set at 20 ps to enable adequate temporal resolution between the FPD and PE signals. Such data by themselves are not complete since both the longitudinal and transverse decay rates depend on wavelength (as indicated earlier for the SPE signal[12]); nevertheless, the data clearly show two resonances with strong spectral overlap of both signals.

In addition to a different saturation intensity, the resonance giving rise to the FPD signal is also characterized by transverse and longitudinal relaxation times which differ considerably from the corresponding time associated with the SPE signal. Figure 2(b) shows the decay of the signal as a function of t_2-t_1 yielding a two-component decay corresponding to dephasing times of 6 and 16 ps.[20] Superimposed on these data is the signal as a function of t_3-t_2 yielding a longitudinal decay time of 14.5 ps (longer delay times are limited by the small signal-to-noise ratio due to the finite t_2-t_1 needed to provide separation between the prompt and delayed signal). Both measurements were shown to be independent of excitation density.

Hence, the data strongly support the presence of two separate resonances contributing to the signal. Numerical calculations of the SPE signals using finite-pulse-width excitation in the presence of different distributions, wavelength-dependent dephasing, and spectral diffusion do not predict the temporal structure seen in Fig. 2(a) unless two separate resonances are assumed where one resonance is inhomogeneously broadened. More formally, the linear absorption spectrum can be described as an inhomogeneously broadened resonance where the decay rates depend on excitation frequency and the distribution function is the sum of a continuous function such as a Gaussian and a weighted δ function.

Excitons localized by potential fluctuations, which also results in inhomogeneous broadening, provide a satisfactory explanation for the origin of the SPE signal and the corresponding relaxation. The short dephasing times of the FPD signal are comparable to those expected for delocalized excitons;[3] however, the FPD resonance would be blueshifted (above the exciton mobility edge) with respect to the SPE resonance, in contrast to the inset in Fig. 2(a). In addition, the measured longitudinal relaxation time is longer than expected for delocalized excitons. (Note that above the line center, our decay rate measurements confirm that both the longitudinal and trans-

verse relaxation times are of order 1 ps and arise from a FPD signal, as expected from earlier work.[3]) The long longitudinal relaxation rate also makes it unreasonable to suggest that the FPD signal arises from delocalized excitons produced in a few high-quality (no interface roughness) QWs within the MQW or from excitons above the mobility edge in QWs which are wider than those QWs giving rise to the SPE. These latter two possibilities are also unlikely since in the first case, we have observed the same spectral structure in different MQW structures; and in the second case, this explanation would produce a low-energy tail in the SPE spectrum not seen in the data. An alternate explanation is based on the fact that exciton-exciton interactions result in a strong exciton density-dependent dephasing. In this sample, the total dephasing rate is measured to be $\gamma_{ph}=\gamma_{ph}^{0}\times(1+n/n_0)$, where $n_0=5\times10^8$ excitons/cm^{2}layer (assuming a uniform distribution of excitons) and the dephasing rate varies linearly with density. This leads to the unlikely result that an average exciton separation much greater than the exciton Bohr radius results in a contribution to the dephasing time due to exciton-exciton interactions of order 70 ps. More likely is that due to variations in interface roughness, some regions are more densely populated with localized excitons, leading to strong dephasing, and regions which are homogeneously broadened giving rise to the FPD signal.

In summary, the time-resolved measurements show the presence of two resonant components in the exciton response. The origin of the SPE is explained by excitons localized by interface roughness. However, while the optical properties of the FPD have been determined, the origin of this signal remains unclear.

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