Transient Linear Birefringence in GaAs Quantum Wells: Magnetic Field Dependence of Coherent Exciton Spin Dynamics

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We use reflective probing of transient linear birefringence to study coherent exciton dynamics in GaAs quantum wells as a function of applied magnetic field at low temperatures. The results show the significance of exchange-enhanced exciton spin relaxation for dephasing at zero field. In applied field the signals show background-free quantum beating which gives the exciton g factor and field-dependent dephasing time. The latter is dominated by spin relaxation which shows for the first time the dramatic slowdown predicted in earlier work. [S0031-9007(96)00078-6]

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Ultrafast dephasing of excitons in semiconductor quantum wells is important because it presents insight into fundamental interactions [1] and possible coherent control of optical properties [2]. A variety of measurement techniques have been used among which are time- and frequency-domain four wave mixing [1,3], resonant Rayleigh scattering [4], and Faraday rotation [5]. In this Letter we report on a different time-resolved technique based on reflective probing of pump-induced transient linear birefringence. This method has various advantages including sensitivity, applicability to opaque as-grown samples, ease of implementation, and freedom from superimposed signals [5]. We have applied the method to GaAs/AlGaAs quantum well samples at low temperatures to investigate heavy-hole exciton dephasing at zero magnetic field and Zeeman beating and spin dephasing in applied field. The data show a dramatic slowdown of spin relaxation with applied field confirming a recent prediction [6–8]. The data also illustrate that, as a result of exchange enhancement, spin relaxation is a significant dephasing process for localized quantum well excitons.

The experiment can be understood using the following semiclassical picture. Our samples are (001)-grown multiquantum wells and magnetic fields are applied along the growth axis (z). The samples are essentially isotropic in the x-y plane [9] and are excited resonantly at the $n = 1$ heavy-hole exciton by a short pump pulse of linearly polarized light propagating along the growth axis; we define the x direction parallel to the pump polarization. This generates equal populations of the two exciton spin orientations ($J_z = \pm 1$) which are σ^+ and σ^- allowed [7,10] each with density of approximately 3.5×10^8 cm^{-2}. In zero magnetic field these states are degenerate and initially the two populations are coherent with the excitation pulse giving a net linear macroscopic dielectric polarization P. Since the leading nonlinear term in the refractive index must be first order in intensity, the polarization P will generate linear birefringence ($n_x - n_y \neq 0$) proportional to $|P|^2$, with optic axis defined by the pump polarization (x). The polarization P and hence the birefringence will decay as the excitons lose phase coherence leaving behind an incoherent population, consisting equally of $J_z = \pm 1$ excitons, which eventually decays by recombination. To measure the pump-induced birefringence a weak delayed probe pulse with linear polarization at 45° to the pump polarization is reflected from the sample and the degree of induced elliptization is measured, that is, the difference of intensities of right and left circularly polarized components in the reflected light. This method of detection is completely insensitive to rotation of the probe polarization azimuth, and so to Faraday rotation, but gives the induced difference of refractive indices ($n_x - n_y$) for light polarized parallel and perpendicular to the pump. Furthermore, effects of pump-induced exciton splitting [11,12] are avoided by use of linearly polarized pulses. Application of a magnetic field removes the degeneracy of the exciton states and causes the pump-induced polarization P and hence the induced optic axis to rotate about the field direction at the Zeeman splitting frequency. This will produce a sinusoidal variation in $n_x - n_y$ and beating in the probe elliptization signal at the Zeeman frequency.

For a more quantitative description allowing for finite frequency width of the pump pulse and assuming inhomogeneous broadening of the transition much greater than the pump, we write the polarization as the sum of contributions P^+ and P^- from σ^+ and σ^- excitons

$$P^{\pm} = \frac{P_0}{2} e^{-t/T_2} \int_{-\infty}^{\infty} e^{i(\omega \pm \Delta)t} G(\omega)\, d\omega\, (\mathbf{x} \pm i\mathbf{y}), \quad (1)$$

$$P = P^+ + P^- = P_0 e^{-t/T_2} \int_{-\infty}^{\infty} e^{i\omega t} G(\omega)\, d\omega\, (\mathbf{x} \cos\Delta t - \mathbf{y} \sin\Delta t), \quad (2)$$

where $G(\omega)$ represents the excitation pulse profile and 2Δ is the Zeeman splitting which is much less than the width of this profile. T_2 is the exciton dephasing time, t the pump-probe delay, and $\mathbf{x}$ and $\mathbf{y}$ are orthogonal unit vectors with $\mathbf{x}$ parallel to the pump polarization. For delays significantly longer than the pulse length the linear

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birefringence and hence the probe signal will then have the form

$$n_x - n_y \propto |P_x|^2 - |P_y|^2 = P_0^2 e^{-2t/T_2} \int_{-\infty}^{\infty} |G(\omega)|^2 \, d\omega \times (\cos 2\Delta t) . \quad (3)$$

This displays a decay with time constant $T_2/2$ and also a beat ($\cos 2\Delta t$) at the Zeeman splitting frequency. (In this semiclassical description the beat arises from independent exciton populations and so is a polarization beat. However, a quantum description in terms of coherent superposition of states of a single exciton is also possible which would lead to identical time dependence of the signal but would be a true quantum beat.)

Two $GaAs/Al_{0.36}Ga_{0.64}As$ 60 period multiple quantum well structures grown by molecular beam epitaxy on (001) substrates and having well widths of 5.6 and 2.75 nm were investigated. The 2.75 nm sample had low-temperature absorption linewidth and Stokes shift of 12 and 5.5 meV, respectively, and for the 5.6 nm sample the corresponding figures were 6 and 3 meV. The experimental setup (Fig. 1) is a conventional pump-probe arrangement based on a mode-locked Ti-sapphire laser. The pump excitation density was approximately 7×10^8 excitons cm^{-2} and the probe intensity was $\sim 10^{-2}$ times that of the pump. Temporal resolution of the system was ~ 2 ps. The probe was horizontally polarized (polarizer $P2$) and the pump linearly polarized at 45° to the horizontal ($P1$). Probe light reflected at 1.5° to normal incidence was detected by a photomultiplier (PMT) after passing through a Soleil-Babinet compensator (SB), a photoelastic modulator (PEM), and a linear polarizer ($P3$). These parts act as a 50 kHz oscillatory filter alternatively transmitting left and right circularly polarized light, so that lock-in 1 referenced at 50 kHz gives a signal proportional to the birefringence. To cancel pump-to-probe leakage and eliminate detector background, pump and probe were chopped at $f_1 = 400$ Hz and $f_2 = 350$ Hz and the signal passed through lock-in 2 referenced to the sum (750 Hz). The output was then normalized to the probe intensity to obtain the reflectivity and to eliminate low frequency laser fluctuations.

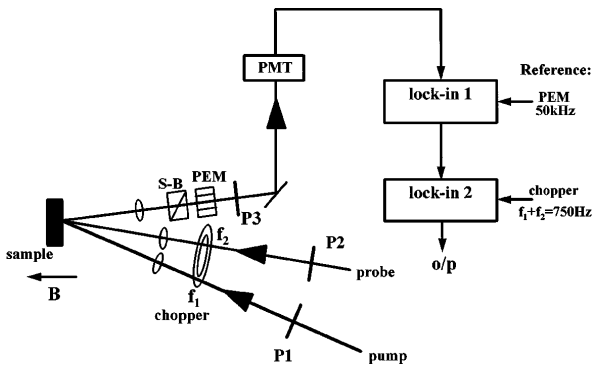

FIG. 1. Experimental setup for transient birefringence. $P1$, $P2$, and $P3$ are linear polarizers, SB a Soleil-Babinet compensator, PEM a photoelastic modulator, and PMT a photomultiplier.

Figure 2 shows typical signals for the $n = 1$ heavy-hole exciton in the 5.6 nm sample at 10 K in zero magnetic field. Rotating the pump polarization through 90° [Fig. 2(a)] should reverse the sign of the birefringence and indeed does result in precise inversion of the signal. No signal was detected for parallel pump and probe polarizations. This gives clear proof that the experimental signal originates from pump-induced birefringence and so is associated with exciton coherence. It also demonstrates that the pump is generating equal populations of σ^+ and σ^- excitons and that the signal is not affected by exciton shifts or splittings of the type observed for high density ($\sim 10^{10}$ excitons cm^{-2}) circularly polarized pump pulses [11,12]. Figure 2(b) shows the observed signal for different excitation wavelengths within the inhomogeneous line profile. The sign and magnitude of the signal varies across the line in a manner which numerical simulation of reflectivity of the structure [13] shows to be a result of pump-induced reduction in exciton oscillator strength as would be expected from Eq. (3) [14]. The decay rate at energies above the line center is considerably greater than at the center and below, consistent with exciton localization at low energies. The

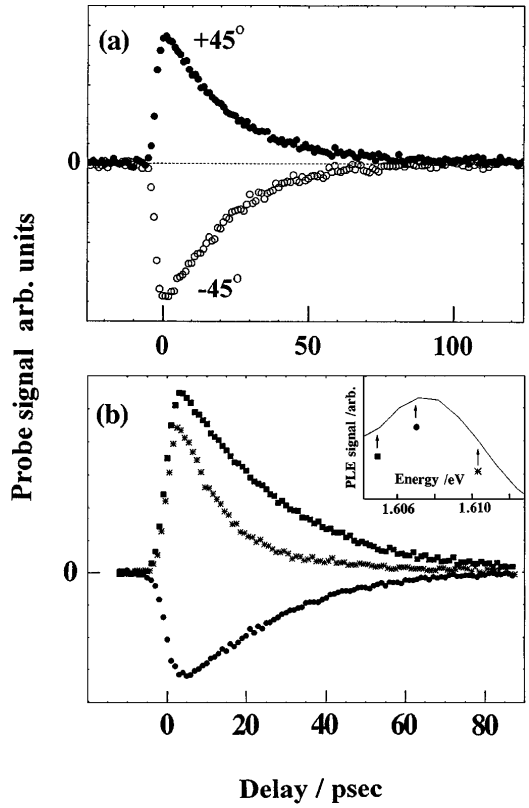

FIG. 2. Transient birefringence for $n = 1$ heavy-hole exciton in a 5.6 nm quantum well sample at 10 K: (a) for two pump polarizations at 90° demonstrating that the signal originates from sample coherence, (b) for different photon energies in the inhomogeneous exciton profile (see inset). Values of T_2 are squares, 50 ± 3 ps; circles, 46 ± 3 ps; and stars, 26 ± 3 ps.

dephasing time can be written [1]

$$\frac{1}{T_2} = \frac{1}{2T_1} + \frac{1}{T'}, \tag{4}$$

where T_1 represents the population decay time due, for example, to exciton recombination, spin relaxation, or spectral diffusion and T' is the pure dephasing time. Measurements using circularly polarized pump pulses at 10 K have given a spin-relaxation time for the 5.6 nm sample of $T_s = 20 \pm 3$ ps while the recombination time T_r is considerably longer [13]. On the low energy side of the line center the measured T_2 is 50 ± 3 ps [see Fig. 2(b)] indicating that dephasing is dominated by spin relaxation, whereas above the line center a shortened T_2 ($=26 \pm 3$ ps) indicates that an additional pure dephasing or diffusion mechanism becomes important. This behavior is fully consistent with previous dephasing investigations in samples of comparable quality [3,4], and illustrates clearly the significance of spin relaxation for dephasing.

Figure 3 shows the birefringence signal from the 2.75 nm sample in a variety of applied fields and at 1.8 K for excitation wavelength close to the line center. At zero field there is exponential decay but as the field is increased beating is observed which can be fitted accurately by Eq. (3). The fit gives the Zeeman splitting from which the exciton g factor 1.52 ± 0.01 is obtained. For the 5.6 nm sample we obtain $g = 0.58 \pm 0.01$ by the same method. These values are consistent with, but far more precise than, our previous measurements using conventional Zeeman spectroscopy [15] where measurements, for the 2.75 nm sample in particular, were strongly affected by level crossing with the $J_z = \pm 2$ exciton dark states. They are also consistent with absorption quantum beat measurements [16].

Figure 4 shows the dephasing rate $(1/T_2)$ for the 2.75 nm sample also obtained by fitting Eq. (3). In this sample the value of T_s obtained using circularly polarized

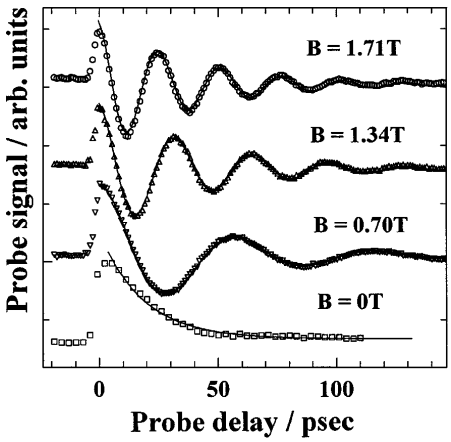

FIG. 3. Transient birefringence from $n = 1$ heavy-hole exciton in a 2.75 nm quantum well sample at 1.8 K for various applied fields. Points are experimental data and curves are fits of Eq. (3).

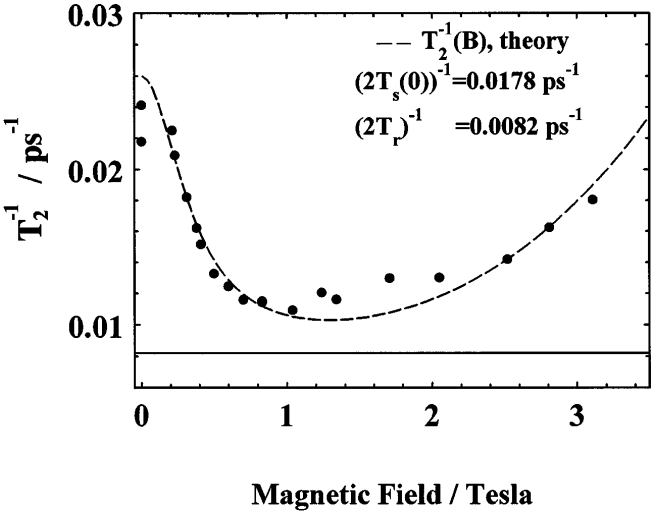

FIG. 4. Dephasing rate for a 2.75 nm sample from fits of Eq. (3) to quantum beat data. The dashed curve is a fit of Eq. (5) to the data taking $[2T_s(B = 0)]^{-1} = 0.0178$ ps^{-1} and $(2T_r)^{-1} = 0.0082$ ps^{-1} (horizontal line).

pump pulses at 10 K in zero field was 21 ± 3 ps and again T_r was considerably longer [13]. It is clear that, in this particular case, pure dephasing and other population lifetime contributions to dephasing are negligible, presumably as a result of the degree of exciton localization indicated by the linewidth and Stokes shift. However, a field of 1 T is sufficient to reduce the spin-relaxation contribution so that the dephasing becomes dominated by recombination. At higher fields the dephasing rate increases once more, probably due to the contribution of "direct" spin-relaxation processes as discussed below.

Measurement of the magnetic field dependence of exciton dephasing in this system thus gives direct information, for the first time, on field-dependent exciton spin relaxation which can be compared with the predicted variation [6–8]. Exciton spin relaxation is dominated by processes involving simultaneous electron and hole spin flips linking the $J_z = \pm 1$ states [6,11,13,17] and is driven by electron-hole exchange interaction which is strongly enhanced by quantum confinement in the GaAs/AlGaAs quantum well system [7]. At zero field it is relatively efficient because the spin states are degenerate but the process is rapidly quenched by application of a small field because energy interchange with a lattice is required. With further increase of field resonant emission and absorption of acoustic phonons at the Zeeman energy ("direct process" spin relaxation [18]) increases in importance due to the increase of the available density of phonon states and can cause recovery of the spin-relaxation rate. To describe this situation we write the exciton dephasing rate in the form

$$\frac{1}{T_2(B)} = \frac{1}{2T_s(B)} + \frac{1}{2T_r}, \tag{5}$$

where we assume T_r to be field independent and [6–8]

$$\frac{1}{T_s(B)} = \frac{1}{T_s(0)} \frac{\Gamma^2}{(2\Delta)^2 + \Gamma^2} + \alpha(2\Delta)^3 \frac{1 + e^{2\Delta/kT}}{|1 - e^{2\Delta/kT}|}. \quad (6)$$

The first term in $T_s(B)^{-1}$ represents zero-phonon spin-relaxation [6] and the second term direct phonon-induced spin-lattice relaxation processes [18]. The zero-phonon relaxation is suppressed when the Zeeman splitting 2Δ significantly exceeds the width parameter Γ, whereas the direct processes become important only at higher fields due to the factor $(2\Delta)^3$. The form of dephasing rate in Eq. (5) can describe the experimental data very well as shown by the curve in Fig. 4. The two undetermined parameters α and Γ were set to 0.6×10^8 ps^{-1} eV^{-3} and 63 μeV, respectively, to obtain a fit. α gives a measure of the exciton-phonon coupling strength and, in an ideal homogeneous system, Γ would be determined by exciton momentum scattering [6] but, in this sample, there is an additional contribution to Γ from inhomogeneous exciton spin splittings which have been measured to be $\sim$20 μeV [7,9]. The values $T_s(0) = 28$ ps and $T_r = 60$ ps used in the fit are consistent with measurements at 10 K [13].

The qualitative field dependence of spin relaxation revealed in Fig. 4 may hold generally in quantum wells where confinement gives significant exchange enhancement. Photoluminescence polarization measurements [7,8] suggest similar behavior for GaAs/AlGaAs wells up to $\sim$10 nm width. For InGaAs/GaAs wells, for which exchange enhancement is estimated to be much less [7], other spin-relaxation mechanisms could be more important leading to different field dependence.

In conclusion, we have demonstrated a novel time-domain method for investigating phase coherence of excitons in semiconductors, making a variety of measurements in GaAs/AlGaAs quantum well samples which reveal both simple phase decay and quantum beating. The measurements greatly improve on the precision of previous g-factor determinations in narrow quantum wells, highlight the importance of spin relaxation in exciton dephasing, and reveal directly the predicted quenching of exciton spin relaxation in applied magnetic field. The technique can readily be extended to shorter pulses and, given the excellent signal-noise ratio (Fig. 3), will be useful for lower excitation densities than used here. In general it can work only in a three-level quantum system since the signal arises from phase coherence of two differently polarized excited populations. The populations investigated in our experiments, namely the two heavy-hole excitons of opposite spin, evolve independently, but the method could also be useful for the study of interacting excitons, for example, biexcitons [19] which might be significant in samples of better quality than those used in this study.

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