# Absorption Quantum Beats of Magnetoexcitons in GaAs Heterostructures

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(Received 24 January 1991)

We report the experimental observation of a transient oscillatory behavior of magnetoexciton absorption in GaAs/AlGaAs heterostructures. We interpret these oscillations as absorption quantum beats of a coherent superposition of two excitonic spin states. Measurements of the oscillations period may be used as a new technique for accurately measuring the Landé $g$ factor of excitons in semiconductors.

PACS numbers: 71.35.+z, 42.50.Md, 71.70.Ej, 78.65.Fa

The optical-absorption spectrum of semiconductor heterostructures is substantially modified when a magnetic field is applied. It breaks into discrete peaks associated with different Landau levels. Furthermore, the degeneracy between different spin states is removed, similarly to the Zeeman splitting in atoms. Excitonic enhancement manifests itself in the well-known magnetoexciton fanning.[1]

While exciton dynamics has been extensively studied in recent years using time-resolved spectroscopy, the temporal behavior of magnetoexcitons received relatively little attention. Stark *et al.* have recently investigated the change of the magnetoexcitonic absorption at various Landau levels following an excitation by a short optical pulse.[2] Relaxation of spin states at the same Landau level was studied by Potemski *et al.* in a cw photoluminescence experiment.[3] It was suggested in this work that spin-flip processes in quantum-confined structures under a strong magnetic field are substantially weaker than in bulk, and that the excited carriers maintain their spin during the thermalization process.

In this paper we study the dynamics of magnetoexciton spin states using time-resolved optical spectroscopy. We show that deep oscillations characterize the temporal evolution of the excitonic absorption when the system is excited by linearly polarized light. We explain these oscillations as *absorption quantum beats* between two excitonic spin states.

Quantum beats are usually associated with coherence-sensitive experimental methods, such as four-wave mixing, photon echoes, coherent Raman scattering, and resonance fluorescence. When two nondegenerate states are coherently driven by an optical field, the induced polarizations in the medium interfere and give rise to a beating in the radiated or diffracted wave, at a frequency given by the difference between the energies of these states. In semiconductors, the continuous density of states results in a rapid decay of the coherence of the induced polarization.[4] However, the excitonic interaction between optically excited electrons and holes near the band gap causes a collapse of the continuous density of states into discrete energy states, and the transition at the exciton energy may be regarded as a discrete transition of a two-level system. This discreteness makes quantum beats observable also in semiconductors, and indeed they recently had been observed in a four-wave-mixing experiment in semiconductor quantum wells.[4]

Time-resolved absorption saturation measurements, on the other hand, are usually considered as a coherence-insensitive method for investigating decay rates of the population in excited states. In this paper we show that this is not always the case, and quantum beats *can* be observed in the temporal evolution of the absorption in semiconductors. In fact, absorption quantum beats have been reported in the literature for the case of hyperfine splitting of atomic states.[5,6] A similar beating phenomenon arises, in our case, when the degeneracy of magnetoexcitons is removed by an applied magnetic field.

Our experiments were conducted at liquid-helium temperature in a split-coil 5-T magnet, using a standard pump-probe configuration with $\approx$1-ps time resolution. The four windows of the cryostat allowed optical access in two orientations of the magnetic field relative to the sample—normal to the sample ($B_\perp$) and parallel to it ($B_\parallel$).

We have studied two different GaAs/GaAlAs samples: a multiple-quantum-well (MQW) sample and a short-period superlattice. The MQW sample has 100 stepped wells, with adjacent layers of 30-Å GaAs and 100-Å $Al_{0.1}Ga_{0.9}As$, separated by 100-Å $Al_{0.3}Ga_{0.7}As$ barriers. The wave functions associated with the lowest confined electron and heavy-hole energy levels of this structure are confined in the narrow GaAs well. The absorption spectrum of the MQW sample shows a wide ($\approx$6 meV) heavy-hole exciton peak at 1.615 eV, indicating inhomogeneous broadening due to well-width fluctuations. Degenerate four-wave-mixing measurements give a signal which is redshifted by 2 meV relative to the excitonic peak absorption, suggesting that the nonlinear response in this sample is dominated by localized excitons on the lower-energy side of the excitonic absorption peak.[7] The superlattice sample consists of 100 periods of alternating 30-Å layers of GaAs and $Al_{0.3}Ga_{0.7}As$, and shows a somewhat weaker excitonic line at 1.683 eV, with a width similar to that observed in the MQW sample.

Figure 1(a) shows the relative change in transmission,

 

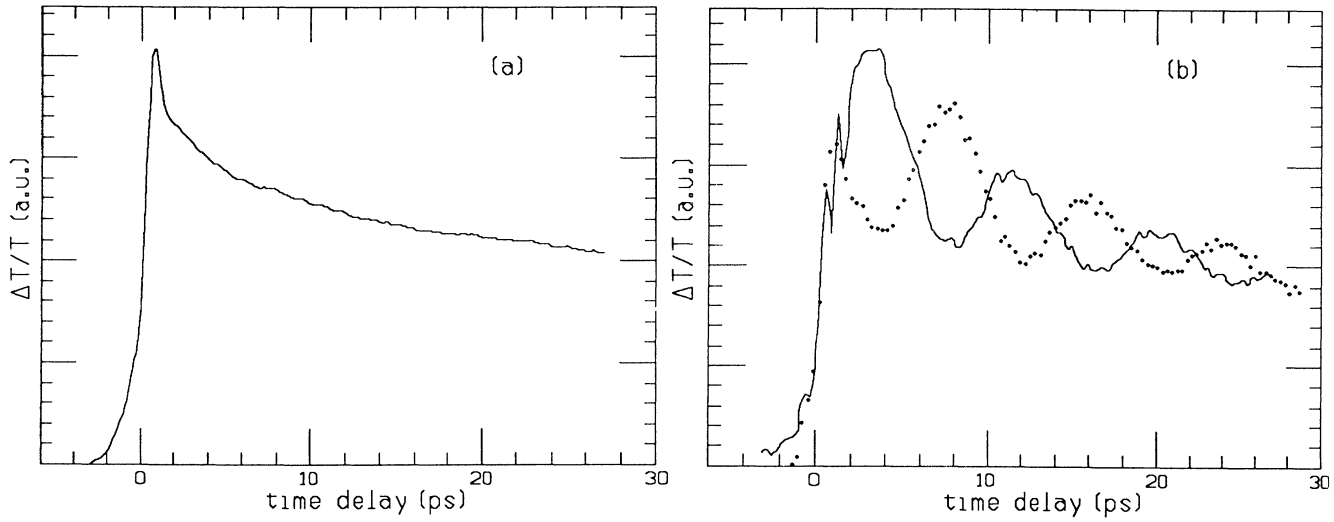


FIG. 1. Temporal evolution of $\Delta T/T$ at the heavy-hole exciton for (a) $B=0$ and (b) $B=4$ T. The solid curves were measured at orthogonal pump and probe polarizations, and the dotted curve was measured as parallel polarizations.

$\Delta T/T$, at $B=0$ for the MQW sample, pumped and probed with roughly equal intensities of approximately 3 W/cm$^2$, slightly below the heavy-hole exciton absorption peak (1.619 eV). Since $\Delta T/T \ll 1$ ($\approx 5\%$) this signal is directly proportional to the exciton absorption change $\Delta\alpha$. The slow decay ($\sim 20$ ps) of the signal reflects the spectral diffusion of the low-energy, localized excitons.[8,9] This behavior is strongly modified as a magnetic field normal to the layers ($B_\perp$) is applied. The solid curve in Fig. 1(b), taken at $B_\perp=4$ T with *orthogonal* linear pump and probe polarizations, exhibits a slow rise (relative to the pulse width), followed by very deep, damped oscillations. These oscillations have a period of 8 ps, with the first minimum occurring a full period after $t=0$. Up to six oscillations were observed at a field of 4 T. The damping time constant, $\sim 20$ ps, is approximately the same as the decay time for the $B=0$ case. The signal then levels off, and decays with a very slow time constant of hundreds of picoseconds, consistent with a recombination process. A similar temporal behavior was observed for the *parallel*-polarization case. The pump-probe signal for parallel polarizations, also at $B_\perp=4$ T, is plotted as the dotted curve in Fig. 1(b). The oscillations are still present, but this time in antiphase to the orthogonal-polarization case. Less pronounced oscillations were observed with the superlattice sample. The phase of the oscillations is similar to that measured in the quantum-well sample, while the oscillation period is slightly longer.

Figure 2 shows the dependence of the oscillations on the magnetic-field strength. The two traces were measured at 2 T (solid curve) and 4 T (dotted curve), with orthogonal polarizations, and demonstrate an oscillation period which is inversely proportional to $B$. We verified this linear dependence of the oscillation period on $B^{-1}$ for magnetic fields from 1 up to 5 T.

The experimental results change dramatically when circular polarizations are used. *Whenever the pump or the probe is circularly polarized the oscillations disappear.* In the particular case of counter-rotating pump and probe polarizations only a weak signal is observed, whose temporal behavior is substantially modified. The time evolution of this signal is currently under investigation.

We also repeated the experiment with the cryostat rotated by 90°, so that the magnetic field was in the plane of the layers ($B_\parallel$) and the laser beams were normal to them. No oscillations were observed in any of the polarizations, in any of the samples.

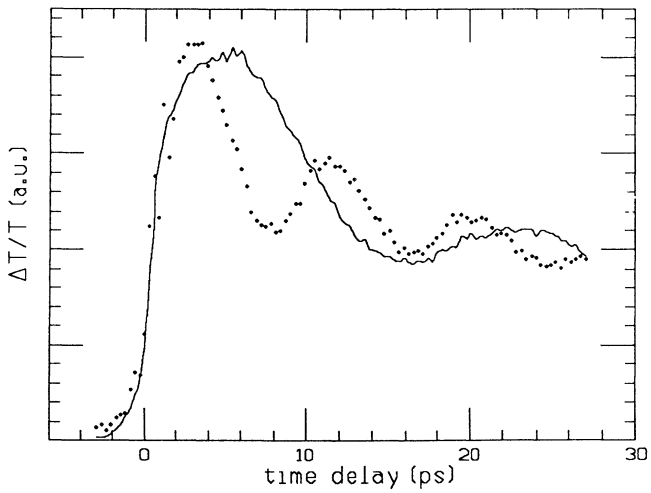


FIG. 2. Pump-probe signal at 2 T (solid curve) and 4 T (dotted curve).

Our interpretation of the experimental results for the $B_\perp$ case is based on the energy-level scheme shown in Fig. 3. The spin splitting between the different $J_z$ levels is given by $g\mu_B B\Delta m_J$, where $m_J$ is the quantum number associated with $J_z$ and $g$ is the effective Landé factor [note that the Landé factor for electrons in the conduction band, $g_e$, is believed to be negative in GaAs (Ref. 10)]. The two allowed transitions for light propagating in the $z$ direction are marked in the figure, corresponding to right-handed ($\sigma_R$) and left-handed ($\sigma_L$) circularly polarized light. Assuming that spin-flip processes are negligible on the time scale of our experiment, we may describe this four-level system as a superposition of two uncoupled two-level systems, which interact with either a $\sigma_R$ or a $\sigma_L$ circularly polarized pump and probe. However, as the splitting between the two spin states ($\sim$ a few tenths of a meV for a 4-T field) is small compared to the bandwidth of our laser (typically $\sim 1$ meV) and to the excitonic linewidth ($\approx 6$ meV), linearly polarized pump and probe interact with both transitions. In other words, a linearly polarized pump excites the two transitions coherently, and the polarizability of the medium seen by a linearly polarized probe is a linear combination of these eigenstates. Based on this picture we can explain the experimental results using the optical Bloch equations.[11]

Let us first consider one of these two-level systems. The total absorbed intensity in the sample can be written in general as[12] $\sim\int dt\, E\, dP/dt$, where $E$ is the time-dependent optical field and $P$ is the induced polarization. Assuming that the relaxation times[11] $T_1$ and $T_2$ are much longer than the duration of the pulse, and that the laser frequency is tuned to resonance, we can write the change in the probe intensity $I_t$, as measured by a slow detector, as

$$\Delta I_t \sim \int dt\, \mathcal{E}_t\, dv(t)/dt \sim \int dt\, \mathcal{E}_t^2 \Delta w(t)\,, \tag{1}$$

where $\mathcal{E}_t$ is the amplitude of the probe, $v(t) = -i[\rho_{12}(t) - \rho_{21}(t)]$, $w(t) = \rho_{22}(t) - \rho_{11}(t)$, $\Delta w(t) = w(t) - w_{eq}$, $w_{eq}$ is the steady-state value of $w$, and $\rho_{ij}(t)$ is the density matrix. [We have assumed in Eq. (1) that the sample is thin and that the change in transmission is small.] The effect of the pump pulse is implicit in this expression through $\Delta w(t)$, which is the change in $w(t)$ due to excitation by the pump. Using the fact that $\mathrm{Tr}\rho = 1$, and taking $w_{eq} = -1$, i.e., the system in steady state is at its ground state, we get $\Delta w = \rho_{22}$. We therefore conclude that $\Delta I_t(t)$ which is measured in a pump-probe experiment is proportional to $\rho_{22}(t)$ induced by the pump pulse. When the pump field is turned off, the induced population $w$ decays exponentially with a relaxation time $T_1$, and the excitonic wave function can be written as

$$\psi(t) = \psi_{R,L} \exp(i\omega_{1,2} t - t/T_1)\,, \tag{2}$$

where $\psi_R$ ($\psi_L$) is the wave function of an exciton created by a $\sigma_R$ ($\sigma_L$) polarization and energy $\hbar\omega_1$ ($\hbar\omega_2$).

The same analysis holds for the case of two uncoupled two-level systems, but since a linearly polarized probe interacts with both systems we should take the *joint density matrix*. To construct this joint density matrix let us consider the case where the pump is polarized in the $x$ direction. The wave function of the system right after the excitation by the pump can be written as $\psi(0) = |\psi_x\rangle = (1/\sqrt{2})(|\psi_R\rangle + |\psi_L\rangle)$ and the time evolution of the system after the pump pulse has left the sample is given by

$$\psi(t) = (1/\sqrt{2})(|\psi_R\rangle e^{i\omega_1 t} + |\psi_L\rangle e^{i\omega_2 t}) e^{-t/T_1}\,. \tag{3}$$

We now rewrite the spin eigenstates in terms of $x$-like and $y$-like wave functions, $|\psi_R\rangle = (1/\sqrt{2})(|\psi_x\rangle - i|\psi_y\rangle)$ and $|\psi_L\rangle = (1/\sqrt{2})(|\psi_x\rangle + i|\psi_y\rangle)$. Inserting these expressions into Eq. (3) we get

$$\begin{aligned}\psi(t) &= \tfrac{1}{2} e^{-t/T_1}(e^{i\omega_1 t} + e^{i\omega_2 t})|\psi_x\rangle \\ &\quad - (i/2) e^{-t/T_1}(e^{i\omega_1 t} - e^{i\omega_2 t})|\psi_y\rangle \\ &= a(t)|\psi_x\rangle + b(t)|\psi_y\rangle\,.\end{aligned} \tag{4}$$

The joint density-matrix elements which are seen by a linearly polarized probe are then given by

$$\rho_{22}^{(x)}(t) = |a(t)|^2 = \tfrac{1}{2}[1 + \cos(\omega_1 - \omega_2)t] e^{-2t/T_1}\,, \tag{5a}$$

$$\rho_{22}^{(y)}(t) = |b(t)|^2 = \tfrac{1}{2}[1 - \cos(\omega_1 - \omega_2)t] e^{-2t/T_1}\,. \tag{5b}$$

Therefore, the resulting change of absorption $\Delta\alpha \sim \rho_{22}$ for linear pump and probe polarizations exhibits an interference term oscillating at the difference frequency $\omega_1 - \omega_2$. This frequency is linearly proportional to $B$, as was indeed observed experimentally. Note that an oscillating term is obtained only for the parallel- and orthogonal-polarization cases. A circularly polarized pump or probe, on the other hand, interacts with one eigenstate only, and the beating disappears. This analysis also explains the phase of the oscillations: The oscillation of the induced absorption in the polarization parallel to the pump ($x$ in our case) starts at a peak and is shifted by $\pi$ with respect to the oscillation in the orthogonal polarization, as indeed observed experimentally [Fig. 1(b)].

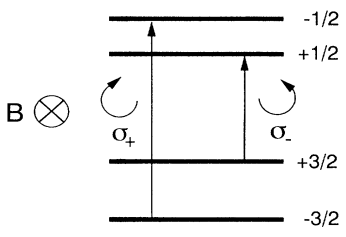


FIG. 3. Electron and heavy-hole energy levels in a normal magnetic field.

The processes which lead to the gradual decay of the oscillations, to which we ascribed the phenomenological time constant $T_1$, are subject to further research. The similarity between the damping time constant of the oscillations and the $B=0$ decay time indicates that the duration of the observed coherent oscillations is limited by the spectral diffusion of the excitons, which apparently preserve their spin orientation as long as they do not experience phonon scattering associated with exciton migration.[9] Preliminary experimental results with circular polarizations indicate that while the localized excitons preserve their spin, spin flip may accompany or follow the spectral diffusion of these low-lying excitons.

The absence of oscillations in the $B_{\parallel}$ case is consistent with measurements of the splitting of sharp photoluminescence lines, related to impurity-bound excitons, in heterostructures in a parallel magnetic field.[13] These measurements show bulklike heavy-hole splitting for very thick quantum wells, gradually decreasing with well width. The splitting is not resolved at well widths below $\approx 150$ Å. Note that when the magnetic field is parallel to the layers and therefore to the quantization axis, the heavy-hole ($J_z = \pm \frac{3}{2}$) splitting vanishes, and the energy diagram in Fig. 3 does not hold anymore.

The absorption oscillations can be used as a novel experimental technique for the measurement of $g_e$ and $g_h$, the electron and hole Landé factors, similarly to a technique which was implemented in gaseous media.[14] The $g$ factors of electrons and holes in GaAs were intensively investigated in both bulk and heterostructures using various techniques. Measurements show a dependence on well width,[15] magnetic-field strength,[16] and orientation.[13] There are, however, inconsistent measurements of $g_e$, ranging over more than 1 order of magnitude. The measured oscillation period of 8 ps at 4 T corresponds to an energy splitting of $\approx 0.5$ meV for the MQW sample, which gives $3g_h - g_e = 2.2$ (Fig. 3). Taking $g_e$ to be the bulk value of $-0.43$, we get $g_h \approx 0.6$. These values are close to those measured for impurity-bound excitons in bulk GaAs.[10] This is in agreement with ESR measurements of $g_e$ in modulation-doped samples, which give a value which is slightly magnetic-field dependent, but very close to the bulk value.[16] On the other hand, our results are inconsistent with the large electron Landé factor, $g_e = 6.8$, deduced from diamagnetic shift measurements in narrow quantum wells.[17] For the superlattice sample we obtain a slightly smaller splitting compared to the MQW sample. This similarity between the MQW and superlattice samples can be explained by the fact that in both structures the lowest electron and heavy-hole wave functions are localized in GaAs layers of similar widths, $\approx 30$ Å.

We are grateful to H. H. Yaffe for the four-wave-mixing data.

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