Spin-Flip-Induced Hole Burning in GaAs Quantum Wells: Determination of the Exciton Zeeman Splitting

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A new method of four-wave-mixing spectroscopy in GaAs quantum wells reveals spectral hole burning due to spin relaxation of magnetoexcitons. The measurements resolve the Zeeman doublet of the lowest-energy heavy-hole exciton where the doublet splitting is much less than the exciton inhomogeneous width. The Zeeman splitting depends nonlinearly on the magnetic field and is small compared with that of bulk GaAs. The results reflect effects of the complex band structure of quantum wells. Information on exciton spin relaxation is also provided by the hole-burning measurements.

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The electronic energy spectrum of quantum well structures is fully quantized under a magnetic field parallel to the growth axis. Optical absorption reveals a ladder of magnetoexcitons corresponding to transitions between electron and hole Landau levels [1]. Magnetic fields also lift the Kramers degeneracy with the resultant Zeeman splitting depending on details of the band structure. The removal of this degeneracy is also expected to lead to a substantial increase of the spin relaxation time between Zeeman-split Landau levels since now spin relaxation can only take place via inelastic processes.

The electron g factor in GaAs heterostructures has been extensively studied. Earlier magnetotransport measurements have shown large exchange-induced enhancement of the electron g [2]. Recent electron-spin-resonance studies have revealed the magnetic field dependence of the electron g for different Landau levels [3]. These results were explained in terms of the nonparabolicity of the conduction band [4]. Determination of the exciton Zeeman splitting has proven to be more elusive [5–7]. Earlier magnetoreflectance measurements were able to resolve Zeeman splittings for the light-hole but not the heavy-hole exciton [5]. More recent measurements have inferred the exciton g from nonlinear quantum-beat spectroscopy [6]. A precise determination of the exciton Zeeman splitting in a quantum well using linear optical spectroscopy is difficult since interface disorder leads to exciton localization and subsequent inhomogeneous broadening of the absorption profile [8]; the resultant inhomogeneous broadening varies from 1 meV to several meV, much larger than the splitting in moderate fields.

A related area is relaxation of carrier and exciton spins in semiconductor heterostructures. Polarization-dependent measurements of interband optical transitions have shown an interesting dependence of spin relaxation on growth conditions, carrier confinement, and temperature [9]. Various physical mechanisms for spin relaxation have also been discussed [9,10]. In addition, luminescence measurements at high magnetic fields have revealed a much larger spin relaxation time due to the full quantization of the energy spectrum [11].

In this paper, we report frequency-domain nonlinear optical studies of exciton Zeeman splitting and spin relaxation in GaAs quantum wells. Using selective optical excitation and nonlinear optical methods similar to spectral hole burning (SHB, also referred to as saturation spectroscopy or differential transmission), we are able to probe spin relaxation of magnetoexcitons and measure directly their Zeeman splitting. The measurements reveal a heavy-hole splitting much smaller than that reported for bulk GaAs at low magnetic field and show a nonlinear dependence of the splitting on magnetic field strength. The results reflect effects of the complex band structure of a quantum well.

Nonlinear optical methods such as SHB have the advantage of being able to eliminate inhomogeneous broadening and accurately measure small energy separations as shown in precision measurements in atomic vapors [12]. For GaAs/AlGaAs quantum wells, a nearly monochromatic optical beam with $\sigma-$ circular polarization can be used to excite a narrow spectral hole (say at energy E_- within the inhomogeneous absorption profile) of the lowest heavy-hole (HH1) exciton associated with the $\frac{3}{2}$ to $\frac{1}{2}$ transition (see the inset in Fig. 1 for the energy-level diagram in a magnetic field). The width of the spectral hole is determined by the homogeneous linewidth. Spin flips of electrons and holes associated with these excitons generate a spectral hole at the energy of the $-\frac{3}{2}$ to $-\frac{1}{2}$ exciton transition, designated E_+. The *spin-flip-induced* SHB at E_+ results from the reduced absorption due to the presence of carriers that have flipped their spins from E_-. This SHB can be probed using an optical beam with $\sigma+$ circular polarization. Zeeman splitting can then be obtained by measuring the energy spacing between the spin-flip-induced SHB and the original SHB resonance.

In practice the measurements proposed above are complicated by strong spectral diffusion of the localized excitons. Once created, localized excitons migrate rapidly among localization sites with different energies leading to *a broad quasiequilibrium distribution in energy* as we demonstrated earlier using four-wave mixing [13]. In the normal SHB measurement discussed above, the nonlinear

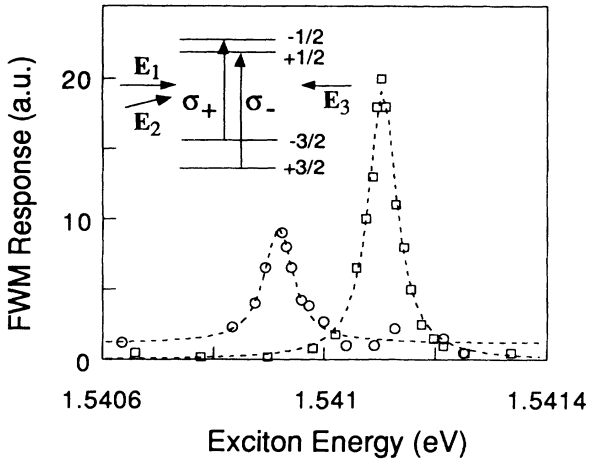

FIG. 1. Hole-burning FWM responses at 4 T. All three beams are circularly polarized with $\mathbf{E}_1$ and $\mathbf{E}_2$ exciting the $\sigma-$ exciton. Squares represent the response when the probe beam $\mathbf{E}_3$ interacts with the $\sigma-$ exciton. Circles represent the response when $\mathbf{E}_3$ interacts with the $\sigma+$ exciton. Dashed lines are Lorentzian fits to the response. Inset: Conduction-band and heavy-hole valence-band energy levels in a GaAs quantum well for a magnetic field parallel to the growth axis.

optical signal may be dominated by this distribution. In the limit that the spin relaxation time is long compared with the spectral diffusion time, nearly all spin-flipped excitons have diffused in energy. Hence, the spin-flip-induced SHB resonance will be overwhelmed by the spectral diffusion process.

To avoid the above complications, we have used a new method of SHB based on nearly degenerate four-wave mixing (FWM) [14,15]. This method can significantly reduce the contribution of the quasiequilibrium exciton distribution to the nonlinear optical response and, hence, allows us to recover the spin-flip-induced SHB resonance. The experimental configuration is based on backward FWM and uses three optical beams. Two nearly degenerate beams designated $\mathbf{E}_1(\omega_1,\mathbf{k}_1)$ and $\mathbf{E}_2(\omega_2,\mathbf{k}_2)$ interact in the sample with a third probing beam designated $\mathbf{E}_3(\omega_3,\mathbf{k}_3)$. The SHB response is obtained by measuring the backward FWM signal (propagating in a direction determined by $\mathbf{k}_1-\mathbf{k}_2+\mathbf{k}_3$) as a function of ω_3. Detailed analytical discussions of frequency-domain FWM spectroscopy have been presented elsewhere [14,15]; however, the underlying physics can be understood as follows: Nearly degenerate beams $\mathbf{E}_1$ and $\mathbf{E}_2$ interfere in the sample to excite a traveling-wave grating which oscillates at a frequency equal to the detuning between the two beams $\delta=|\omega_1-\omega_2|$ ($\ll$homogeneous linewidth). The grating is a spatially modulated pattern of spectral hole burning at ω_1 ($\approx\omega_2$) with amplitude proportional to $(\delta+i\gamma)^{-1}$, where γ is the grating decay rate determined by relaxation and spatial transport of excitons. Measuring the FWM signal as a function of ω_3 probes the spectral profile of the grating within the inhomogeneous profile. In the absence of spectral diffusion, the *spectral* width of the grating is given by the exciton homogeneous linewidth as expected from SHB, and the width of the FWM response is twice the homogeneous linewidth [15]. Note that although both FWM and the traditional SHB (i.e., differential transmission) signals result from the induced nonlinear optical polarization, the FWM signal includes contributions from both the real and the imaginary part of the nonlinear susceptibility while the traditional SHB measures only the imaginary part [12].

In the presence of spectral diffusion, the spectral hole excited by $\mathbf{E}_1\cdot\mathbf{E}_2^*$ diffuses in energy resulting in a spectral redistribution of the excitation. In this case, the FWM response arises from both the spectral hole and the quasiequilibrium distribution of the exciton population as discussed above. The decay of the SHB population is determined by the sum of the exciton spectral diffusion, spin relaxation, and recombination rates. However, the lifetime of the quasiequilibrium distribution is determined by the *recombination time of the exciton* [13]. In the limit that the spectral diffusion rate is much larger than the rate for exciton recombination, setting δ large compared with the recombination rate (but still smaller than or comparable with the spectral diffusion rate) significantly decreases the relative amplitude of the grating associated with the quasiequilibrium distribution. Hence, the FWM response will be dominated by the SHB resonance [14].

The quantum well samples used in our measurements consist of ten periods of 100-Å GaAs wells and 100-Å $Al_{0.3}Ga_{0.7}As$ barriers, grown at 750 °C by molecular-beam epitaxy on semi-insulating (100) GaAs substrates using interrupted growth. The structures show a 1-meV absorption linewidth with a Stokes shift of the luminescence of order 0.3 meV for the HH1 exciton. The nonlinear measurements were carried out at 2.5 K using a split-coil superconducting magnet. δ was set at 140 MHz using two acousto-optic modulators. The exciton density was of order $10^7/cm^2$.

In the first set of measurements, we used three circularly polarized optical beams rotating in the same direction in the laboratory frame. The nonlinear optical response, shown as squares in Fig. 1, involves only the $\sigma-$ excitons associated with the $\frac{3}{2}$ to $\frac{1}{2}$ transition. The width of the response corresponds to a homogeneous linewidth of 0.03 meV. The small linewidth confirms the localized and inhomogeneous broadening nature of the magnetoexciton [8,13].

As discussed earlier, if electrons or holes associated with $\sigma-$ excitons created with $\mathbf{E}_1\cdot\mathbf{E}_2^*$ flip their spin at the same localization site, it will produce SHB at the energy of the $\sigma+$ exciton. Experimentally, spin-flip-induced SHB can be probed by reversing the polarization direction of $\mathbf{E}_3$. The resulting resonance, shown as circles in Fig. 1, clearly shows narrow SHB at a lower energy with

the energy difference being the exciton Zeeman splitting: 0.19 meV at 4 T. The small signal at the original SHB position is most likely due to the residual ellipticity of the circularly polarized beams. The nearly constant background signal in the inset in Fig. 1 is due to excitons that have spectrally diffused. Nonlinear signals due to the spectrally diffused excitons overwhelm the spin-flip-induced SHB when $\delta=0$. Note that spin relaxation of $\sigma+$ excitons requires absorption of acoustic phonons and is slower than spin relaxation of $\sigma-$ excitons. The observed spin-flip-induced SHB is considerably weaker at 4 T when $\mathbf{E}_1\cdot\mathbf{E}_2^*$ excites $\sigma+$ excitons.

In further experiments, we used linearly polarized light for the third beam. With $\mathbf{E}_1\cdot\mathbf{E}_2^*$ exciting only the $\sigma-$ excitons, we can simultaneously probe the SHB and the spin-flip-induced SHB resonance. The FWM response (Fig. 2) shows the well-resolved Zeeman doublet. Because of possible interference between the two resonances, the Zeeman splitting determined from Fig. 2 is less accurate than that from Fig. 1.

Recent measurements have shown that at low and intermediate magnetic field, the electron g factor at the lowest Landau level in GaAs quantum wells is close to the value for bulk GaAs [3,16]. In contrast, the Zeeman splitting of the HH1 exciton obtained above is very small in comparison with that reported for bulk GaAs [17]. Our results seem to be in agreement with the earlier magnetoreflectance measurements where the heavy-hole Zeeman doublet was not resolved. The small Zeeman splitting attributed to the strong valence-band mixing in quantum well structures has been recently predicted by numerical calculations of magnetoexcitons using the Luttinger Hamiltonian [18]. In particular, the mixing of $\sigma-$ excitons with excitons at higher energy pushes the $\sigma-$ exciton to lower energy. The above calculation also predicts an eventual sign change of the Zeeman splitting at higher magnetic fields where the band-mixing effects overcome those of the Zeeman interaction. However, theoretical determination of the magnetic field at which the zero crossing occurs is difficult since the cancellation of the two competing contributions depends strongly on parameters of the model [18]. The sign change of the splitting has not been observed in our measurements up to 6 T. It is interesting to note that disorder-induced localization is expected to enhance the band-mixing effects if the localization scale is smaller than the exciton Bohr radius. Using the recombination rate of the localized exciton, we have estimated the localization scale to be comparable to the exciton Bohr radius.

Figure 3 displays the magnetic field dependence of the Zeeman splitting for the HH1 exciton. The dashed line in the figure represents a quadratic dependence. Clearly the quadratic behavior may not extend into the high-field region. The observed field dependence is somewhat surprising since the calculations predict a field dependence slower than linear. The observed dependence may be due in part to the nonparabolicity of the conduction band [4], which was not included in the calculations. Note that our results differ considerably from those obtained from nonlinear quantum beats in a 30-Å stepwise GaAs quantum well [6]. Zeeman splittings reported in the quantum-beat measurement (0.5 meV at 4 T) are proportional to magnetic fields with a field strength ranging from 1 to 5 T, and are very close to those measured for impurity-bound excitons in bulk GaAs [19]. The sign of the Zeeman splitting cannot be determined in quantum-beat measurements.

The relative amplitude of the two SHB resonances shown in Fig. 2 is determined by the spin relaxation rates as well as the spectral diffusion rate of the exciton (assuming equal oscillator strengths). As expected, exciton spin relaxation rates decrease with increasing magnetic fields, resulting in a decrease in the relative peak height of the spin-flip-induced SHB. The ratio of $\sigma+$ to $\sigma-$ exciton SHB amplitudes decreases approximately by a factor of 4 when the field increases from 2 to 6 T. Using a simple rate equation and a lifetime of 50 ps for the $\sigma+$ exciton SHB (determined independently [20]), we are able to estimate an effective spin relaxation rate of 100 ps for the $\sigma-$ exciton at 4 T. The estimated spin relaxation

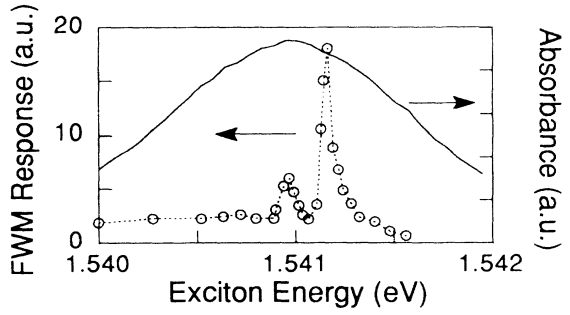

FIG. 2. FWM hole-burning response at 4 T with $\mathbf{E}_3$ linearly polarized. $\mathbf{E}_1$ and $\mathbf{E}_2$ interact only with the $\sigma-$ exciton. The line shape shows the well-resolved Zeeman doublet. The solid line is the exciton absorption spectrum. The dotted line is a guide to the eye.

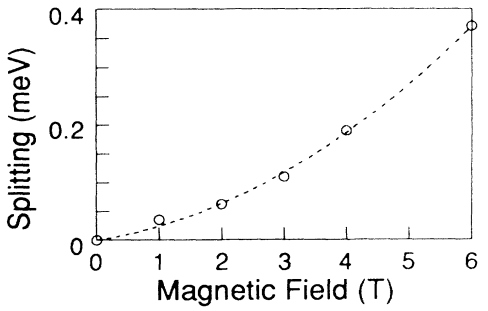

FIG. 3. Magnetic field dependence of the exciton Zeeman splitting. The dashed line is a least-squares fit with a quadratic dependence.

rate is smaller than the spectral diffusion rate, but is much larger than the rate for exciton recombination. Hence, most of the spectrally diffused $\sigma-$ excitons changed their spins before recombination, which explains the negligible nonlinear optical response from the spectrally diffused $\sigma-$ excitons in Fig. 1.

Finally, we note that although spin relaxation of magnetoexcitons in a quantum well is presumably an inelastic process, the relaxation may also proceed via an elastic process due to disorder-induced localization. A localized magnetoexciton with a specific spin orientation may have the same energy as magnetoexcitons at another localization site with different spin orientation. Strong resonant excitation transfer may occur between the two sites. The resonant transfer should depend strongly on the intersite distance as well as on the Zeeman splitting, providing a good probe for the interface disorder. In the SHB measurement, the resonant intersite transfer is characterized by a spin-flip-induced SHB resonance at the energy of the nearly degenerate $\mathbf{E}_1$ and $\mathbf{E}_2$ beams. However, conclusive evidence for the transfer has not been observed in samples used in our measurements.

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