Optically Detected Magnetic Resonance Study of a Type-II GaAs-AlAs Multiple Quantum Well

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In a type-II GaAs-AlAs multiple quantum well three optically detected magnetic resonance lines and two level anticrossings were observed. Two of the resonance lines and the two level anticrossings are in agreement with the electronic level scheme of the heavy-hole exciton. The third resonance line is in accordance with a magnetic spin resonance of an unbound electron. These optically detected magnetic resonance measurements open up the possibility to obtain detailed information about the excitons in and the band structure of type-II quantum wells.

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Optically detected cyclotron resonance (ODCR) and optically detected magnetic resonance (ODMR) have proved to be valuable techniques to study carriers and excitons in bulk semiconductors.[1] Recently, these techniques have been applied by Cavenett and co-workers to study the cyclotron and magnetic spin resonance of electrons in quantum well structures.[2-4] However, excitons in quantum wells have not yet been studied by the ODMR technique. In this Letter we present the results of the first ODMR investigation of the excitons in a type-II GaAs-AlAs multiple quantum well. It will be shown that with this technique an identification of the luminescence feature and detailed information about the electronic structure of the excitons can be obtained. Furthermore, it opens up the possibility to obtain information about the band structure of these quantum wells.

Depending on the thickness of the GaAs layer, two types of recombination are possible in a GaAs-AlAs quantum well. For GaAs layers with a thickness larger than $\simeq 35$ Å recombination takes place within the GaAs layer. This recombination is classed as type I. For GaAs layers thinner than $\simeq 35$ Å the lowest Γ conduction-band confined state in the GaAs layer is higher in energy than the X conduction-band confined state in the AlAs layer. Consequently recombination takes place between the electrons in the AlAs and the holes in the GaAs. This recombination between spatially separated carriers is classed as type II. Confirmation of type-II recombination has been provided by photoluminescence studies.[5-8]

As a result of the reduced overlap of the electron and hole wave functions in the type-II systems the luminescence decay rates are several orders of magnitude lower than in the type-I systems.[7-9] The type-II lifetimes are typically in the microsecond range. In an ODMR experiment the microwave-induced transition rate should be higher than or comparable to the optical transition rate to make detection of magnetic resonances feasible. For the microwave powers available for the measurements at liquid-helium temperature, this requirement implies lifetimes of the spin system studied of microseconds or longer. Therefore, in contrast to the type-I systems, the type-II systems can, in principle, be studied by the ODMR technique.

The sample used in this study was grown by molecular-beam epitaxy in a Varian Gen II system. The layers were deposited on a (001)-oriented semi-insulating GaAs substrate and consisted of 1.0 μm of GaAs buffer material. Sixty periods of 25-Å-GaAs/25-Å-AlAs, and finally a capping layer of 0.1 μm of GaAs. The ODMR experiments were carried out at 9.68 GHz with the sample at a temperature of 1.6 K. For the excitation the 514-nm line from an argon-ion laser was used. The luminescence was

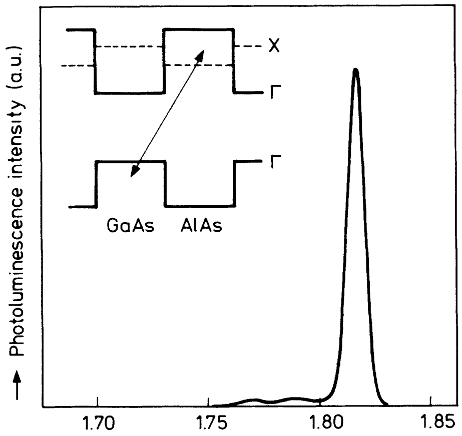

FIG. 1. Low-temperature (1.6 K) photoluminescence spectrum of the 25-Å-GaAs–25-Å-AlAs multiple quantum well. Inset: Valence- and conduction-band structure together with the type-II recombination transition.

observed in a direction parallel to the magnetic field. The microwaves were chopped and synchronous changes in the emission were recorded with a lock-in detector.

The photoluminescence spectrum of the 25-Å-GaAs–25-Å-AlAs multiple quantum well is shown in Fig. 1. For this quantum well the luminescence decay could be fitted with a time constant of 1.5 μs. This time constant is in agreement with the type-II recombination process in this sample. ODMR signals have been observed by the monitoring of microwave-induced changes in the σ^+ and σ^- circularly polarized components of the whole type-II luminescence feature. The ODMR spectrum is presented in Fig. 2(a). The resonant change amounts to about 10^{-3} of the luminescence intensity. Three microwave-induced transitions were observed: two lines with equal linewidths of 5 mT at 351 and 382 mT, respectively, and a line in between at 366 mT with a smaller linewidth of 2 mT.

To obtain more information on the origin of these resonance lines they were studied for various microwave chopping frequencies. The ODMR spectrum shown in Fig. 2(a) was obtained at a chopping frequency of 1 kHz, i.e., the microwave pulses were long compared to the decay time of the luminescence. At this chopping frequency the three resonance lines are observed with equal intensity but opposite polarity for the σ^+ detection with respect to σ^- detection. An increase in the chopping frequency changes the intensity ratio of the σ^+ and σ^- detected signals for the lowest- as well for the highest-field resonance line. At a chopping frequency of 250 kHz, i.e., when the microwave pulse length is comparable to the decay time, the highest-field resonance line is only seen as an increase in the σ^+ emission and the lowest-field resonance line only as an increase in the σ^- emission intensity. The line in between is observed with nearly equal intensity but opposite polarity in the σ^+ and σ^- emission for microwave chopping frequencies up to 250 kHz. From the similar behavior of the lowest- and highest-field resonances as well as the equal linewidths it is clear that these two resonances should belong to the same spin system. The resonance line in between is most likely a resonance in another spin system.

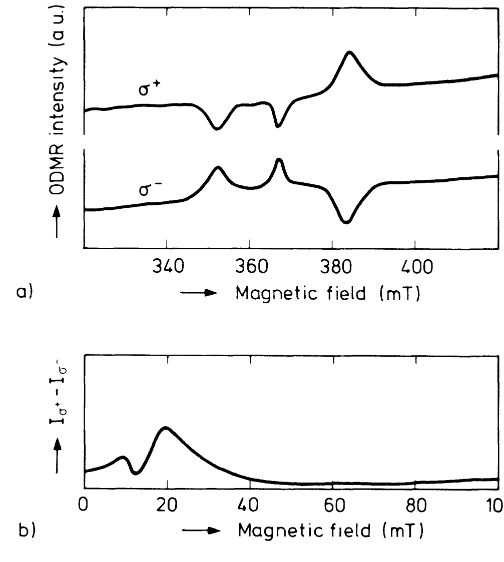

FIG. 2. (a) ODMR signals of the heavy-hole excitons and the unbound electrons obtained at a microwave chopping frequency of 1 kHz and detected as intensity changes in the σ^+ and σ^- circularly polarized components of the emission. (b) Level-anticrossing measurement taken by our monitoring the difference in σ^+ and σ^- emission intensities as a function of the magnetic field.

To determine whether the lowest- and highest-field lines can be assigned to excitons we looked for level anticrossings (LAC's). LAC's are expected for the exciton because of the exchange coupling of the hole and electron and the resulting zero-field splitting of the energy levels. If one of the levels involved decays by emission of circularly polarized light, the LAC can be observed as a change in σ^+ or σ^- emission intensity as a function of the magnetic field. For the LAC measurement the difference of σ^+ and σ^- emission was recorded with a photoelastic modulator operating at 50 kHz. The LAC spectrum showing two overlapping LAC's at about 15 mT is presented in Fig. 2(b).

To ascertain whether the lowest- and highest-field resonance lines and the LAC's are related and to obtain an assignment of the resonance line at 366 mT, the Hamiltonians[10,11] for the electrons,

$$H_e = \mu_B g_{ez} S_z B_z, \tag{1}$$

for the holes,

$$H_h = -D[J_z^2 - \tfrac{1}{3}J(J+1)] - 2\mu_B B_z(\kappa J_z + q J_z^3), \tag{2}$$

and for the excitons,

$$H_{ex} = H_e + H_h + H_{eh} \tag{3}$$

are considered. The exchange coupling of the electron and hole forming the exciton is given by

$$H_{eh} = -a\mathbf{J}\cdot\mathbf{S} - b(J_x^3 S_x + J_y^3 S_y + J_z^3 S_z). \tag{4}$$

In these expressions, S denotes the electron spin, J the effective hole spin including angular momentum, μ_B the Bohr magneton, and B_z the magnetic field strength. The magnetic field is applied along the z direction which corresponds to the [001] crystal axis. In the expressions a potential of low symmetry has been assumed; g_{ez} is the g factor for the electrons, κ and q are the Luttinger parameters[10] for the Zeeman energy splitting of the holes, and a,b are the exchange-coupling constants. The first term in H_h describes the difference in confinement energy for the light hole ($J_z = \pm\frac{1}{2}$) and the heavy hole ($J_z = \pm\frac{3}{2}$).

For the magnetic fields used in the ODMR experiments the Zeeman energy splittings are much smaller than the difference in confinement energy for the light and heavy holes which amounts to 61 meV in the quantum well studied.[6] Therefore, the light-hole exciton ($J_z = \pm\frac{1}{2}$, $S_z = \pm\frac{1}{2}$) and the heavy-hole exciton ($J_z = \pm\frac{3}{2}$, $S_z = \pm\frac{1}{2}$) can be considered separately. At liquid-helium temperature only the heavy-hole exciton energy levels are populated. For the heavy-hole exciton the microwave-induced transitions with $\Delta S_z = \pm 1$, $\Delta J_z = 0$ are allowed. Because of the exchange coupling, these two transitions occur at different magnetic fields. The experimental observation of two related resonances is in agreement with a heavy-hole exciton assignment.

The fact that the LAC's are observed at much lower fields than the ODMR lines implies that the microwave energy $h\nu$ is much larger than the exchange energy. For this situation the heavy-hole exciton energy levels as functions of the magnetic field together with the LAC's and the microwave-induced transitions are shown schematically in Fig. 3.

For the heavy-hole exciton the two resonances with $\Delta S_z = \pm 1$, $\Delta J_z = 0$ occur for $h\nu = \mu_B g_{ez} B_z \pm (1.5a + 3.375b)$. From the exciton resonance fields we obtain $g_{ez} = 1.89$ and the exciton exchange splitting $1.5a + 3.375b = 1.7$ μeV. When we consider the foregoing expression for the exciton resonances, it is obvious that the ODMR line in between the exciton resonances should be a spin resonance of the electrons not bound to holes. In the literature no experimental magnetic electron-spin-resonance g value is reported for AlAs, however, for $Al_{0.8}Ga_{0.2}As$ Böttcher *et al.*[12] report a g value of 1.96 for X-conduction-band electrons. An estimated value for AlAs of $\simeq 1.9$ can be obtained by use of Roth's formula.[13] These values are far off from the g value of 0.4 for Γ-conduction-band electrons in GaAs. We therefore conclude that the electron of the heavy-hole exciton is indeed at the AlAs X point. Preliminary measurements on the anisotropy of the g value indicate that there is an anisotropy of a few percent. This anisotropy and the difference between the 1.96 value of Böttcher *et al.* and the experimental value of 1.89 likely are a consequence of the confinement of the electrons in the AlAs.

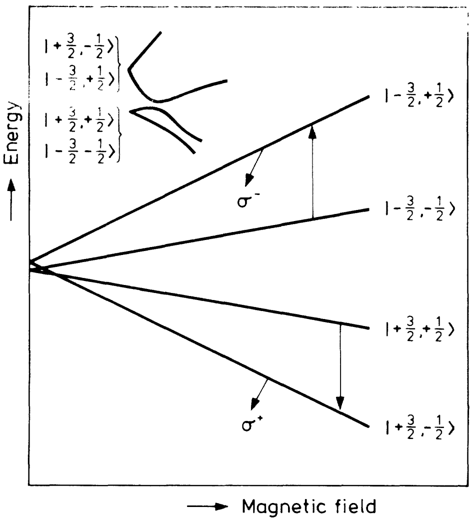

FIG. 3. Energy-level diagram for the heavy-hole excitons. Inset: Schematic of the level anticrossing region. The microwave-induced transitions and the effective population changes are indicated by arrows.

The electron-spin resonance is observed as an increase in the σ^- and a decrease in the σ^+ emission intensity. This can be understood by spin-conserving formation of the excitons.[14] The electron-spin resonance causes the population of the $|+\frac{1}{2}\rangle$ and $|-\frac{1}{2}\rangle$ levels to change in a complementary way. A transfer of population from the $|-\frac{1}{2}\rangle$ to the $|+\frac{1}{2}\rangle$ level at resonance will involve an increase in population of the $|\pm\frac{3}{2},+\frac{1}{2}\rangle$ and a decrease in population of the $|\pm\frac{3}{2},-\frac{1}{2}\rangle$ exciton levels. Because only the $|-\frac{3}{2},+\frac{1}{2}\rangle$ and $|+\frac{3}{2},-\frac{1}{2}\rangle$ levels decay by σ^- and σ^+ radiation, respectively, this electron-spin resonance will increase the σ^- emission and decrease the σ^+ emission which is in agreement with the observation. So in the nonresonant situation the $|-\frac{1}{2}\rangle$ level of the unbound electrons is the most populated. Because the $|-\frac{1}{2}\rangle$ level is the lowest energy level and spin-dependent population of the electron energy levels is not expected with excitation far above the bandgap, the highest population being in the $|-\frac{1}{2}\rangle$ level is most likely a result of thermalization.

The population distribution over the exciton energy levels can also be obtained from the experiment. At a microwave chopping rate comparable to the luminescence decay rate both exciton resonances are detected as an increase in the corresponding circularly polarized emission. This can only be explained by a nonthermalization of the population over the exciton energy levels, with the highest populations in the $|+\frac{3}{2},+\frac{1}{2}\rangle$ and $|-\frac{3}{2},-\frac{1}{2}\rangle$ levels.

From the spin Hamiltonians the level-crossing fields are calculated as

$$B_{LCe} = (1.5a + 3.375b)/\mu_B g_{ez} \tag{5}$$

for the crossing of the $|+\frac{3}{2},-\frac{1}{2}\rangle$ and $|+\frac{3}{2},+\frac{1}{2}\rangle$ levels and

$$B_{LCh} = (a + 2.25b)/(4\mu_B\kappa + 9\mu_B q) \tag{6}$$

for the crossing of the $|+\frac{3}{2},-\frac{1}{2}\rangle$ and $|-\frac{3}{2},-\frac{1}{2}\rangle$ levels. Because of higher-order terms in the spin Hamiltonian, LAC's instead of level crossings will occur. The exchange-coupling strength obtained from the ODMR fields implies $B_{LACe} = 16$ mT. Because the highest populations are in the $|\pm\frac{3}{2},\pm\frac{1}{2}\rangle$ levels, the LAC's should correspond to an increase in the σ^+ emission. This is in accordance with the observation of a LAC region with an overall positive $I_{\sigma^+} - I_{\sigma^-}$. Analysis of the spectrum as consisting of two LAC's gives maxima at 8 and 18 mT. The highest-field maximum most likely corresponds to the crossing of the $|+\frac{3}{2},-\frac{1}{2}\rangle$ and $|+\frac{3}{2},+\frac{1}{2}\rangle$ levels. From the other LAC maximum the sum of the Zeeman energy splitting constants for the hole is obtained as $\kappa+2.25q=0.6\pm0.3$.

The highest population being in the $|+\frac{3}{2},+\frac{1}{2}\rangle$ and $|-\frac{3}{2},-\frac{1}{2}\rangle$ levels is in agreement with the selection rules, forbidding decay from these levels to the ground state. For the population in these levels two ways are possible for indirect decay to the ground state, i.e., either by first making a hole-spin flip or by first making an electron-spin flip. In case of hole-spin flips, the $|+\frac{3}{2},+\frac{1}{2}\rangle$ level decays indirectly by emitting σ^- and $|-\frac{3}{2},-\frac{1}{2}\rangle$ by σ^+ radiation. The opposite is true with respect to σ^- and σ^+ for electron-spin flips. The opposite polarity of the exciton ODMR lines in σ^+ and σ^- detection at low chopping frequencies is a result of these hole-spin flips. For instance, for the highest-field resonance the microwaves couple the $|+\frac{3}{2},+\frac{1}{2}\rangle$ and $|+\frac{3}{2},-\frac{1}{2}\rangle$ levels resulting in an increase of the σ^+ emission. At high enough microwave powers the microwave-induced transition rate will be higher than the hole-spin-flip rate and is the depopulation determining process for the $|+\frac{3}{2},+\frac{1}{2}\rangle$ level. Then, at resonance a simultaneous decrease of the σ^- emission is observed. This effect is not observed at high microwave chopping frequencies because during the lifetime of the exciton only the population of the levels coupled by the microwaves can be changed.

In conclusion, we have observed for the first time ODMR of the excitons in a type-II GaAs-AlAs multiple quantum well. Two of the ODMR lines and the two LAC's are in agreement with the electronic level scheme of the heavy-hole exciton. The luminescence feature in this quantum well is due to the decay of heavy-hole excitons built up of spatially separated electrons and holes. The exciton exchange splitting and the electron and hole Zeeman energy splitting constants are obtained. No thermalization exists among the populations in the exciton energy levels.

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