Observation of Internal Transitions of Confined Excitons in GaAs/AlGaAs Quantum Wells

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Internal transitions of confined hydrogenic excitons in type-I multiple-quantum-well structures have been observed and studied as a function of magnetic field (0–15 T) and quantum well width (80–200 Å). A highly sensitive technique that combines visible and far infrared methods, optically detected resonance spectroscopy, was employed, and revealed several internal excitonic transitions, with the $1s \rightarrow 2p_+$ transition dominant. The energies of the $1s \rightarrow 2p_+$ transition vs magnetic field are in very good agreement with variational calculations for the 80 and 125 Å wells. The strength of the intraexcitonic features was observed to be much weaker in the 200 Å well-width samples. [S0031-9007(96)00768-5]

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The Coulomb attraction between electrons and holes in semiconductors results in the formation of hydrogenlike complexes known as excitons. In quantum-well structures which confine the charge carriers, a series of excitons appear which correspond to the various interband transitions among confined electron and hole subbands. These confined excitons have been actively studied in the GaAs/AlGaAs system during the last twenty years [1]. Excitons in GaAs/AlGaAs heterostructures have an energy spectrum similar to that of the hydrogen atom except that the energy scale is greatly reduced by the small effective electron and hole masses and the large dielectric constant of GaAs. Thus the internal energies of the transitions among the various excitonic states all lie in the far-infrared (FIR) region of the spectrum. Information about the various s-like excited states of the lowest confined exciton in the GaAs/AlGaAs system has been obtained from photoluminescence excitation (PLE) [2] and magnetoreflectance [3] experiments; however, the $1s \rightarrow np$ $(n = 2, 3, \ldots)$ transitions cannot be investigated by these spectroscopies.

We report the direct observation of intraexcitonic transitions using a recently developed optically detected resonance (ODR) technique [4–10]. With this method the sample is illuminated by two laser beams simultaneously. A monochromatic FIR laser beam excites electronic transitions whose energies are tuned into resonance with an applied magnetic field. Under resonant conditions, the FIR absorption induces changes in the intensity of the band-edge luminescence which is excited by a separate laser whose photon energy is larger than the effective band gap of the heterostructure under investigation. We have recently reported the observation with this technique of a variety of FIR resonances in donor-doped GaAs/AlGaAs quantum wells, including electron cyclotron resonance (CR), neutral donor $1s \rightarrow 2p_+$ transitions, and singlet and triplet transitions of negative donor ions [11]. In the present work the $1s \rightarrow 2p_+$, $1s \rightarrow 3p_+$, and $1s \rightarrow 4p_+$ (in the hydrogenic notation) internal transitions of the e_1h_1 exciton have been identified in a set of GaAs/AlGaAs multiple-quantum-well (MQW) samples having well widths between 80 and 210 Å. To our knowledge, the only reported study of internal excitonic transitions were carried out on type-II GaAs/AlAs quantum wells by a photoinduced absorption technique [12].

The successful application of the ODR technique to the study of internal transitions of excitons provides information complementary to conventional PL, PLE, or absorption studies of excitons, and permits a determination of the exciton binding energy that is not dependent on separate measurements of the free electron-hole energy, and associated complications such as band-gap renormalization. This provides a number of opportunities to investigate and develop unique information on the energy spectrum of excitons under a variety of conditions. For example, the formation of negatively or positively charged excitons and screening of the excitons in the presence of excess electrons or holes can be explored systematically as a function of carrier density. In addition, studies of the type-I to type-II transition of excitons in ZnSe/ZnFeSe quantum wells can be investigated in detail in the presence of a range of applied magnetic fields which dramatically modifies the exciton binding energy [13,14].

The samples were mounted in the Faraday geometry in helium exchange gas at liquid helium temperature at the center of a 15 T superconducting magnet. The FIR laser light was guided and focused onto the sample by a lightpipe and condensing-cone system. PL was excited with the 6328 Å line of a He-Ne laser via a 600 μm diameter optical fiber at power densities up to 0.75 W/cm^2. The PL signal was collected with a second 600 μm fiber and analyzed with a single grating spectrometer, equipped with a Si-diode detector. A CO_2-pumped FIR laser was used with methanol as the FIR gain medium to generate laser lines at 163.0, 118.8, 96.5, and 70.5 μm. Changes in the PL intensity induced by the FIR pump beam were synchronously detected with a lock-in amplifier referenced to the chopped output of the FIR laser. A dedicated computer was used to record simultaneously the magnetic field values and the corresponding detected changes in the PL, and to control the monochromator drive in order to follow the center of the desired PL feature as it shifted with magnetic field.

The samples used in this study were molecular beam epitaxy–grown GaAs/$Al_{0.3}Ga_{0.7}As$ MQW structures. Sample 1 is an undoped (125 Å well/150 Å barrier)$_{30}$ structure; sample 2 is a (125 Å well/150 Å barrier)$_{30}$ structure doped with Si donors ($n = 1 \times 10^{16}$ cm^{-3}) in the central one-third of the wells; sample 3 is a (80 Å well/150 Å barrier)$_{45}$ structure, also doped with Si donors ($n = 1 \times 10^{16}$ cm^{-3}) throughout the central one-third of the wells. The PL and FIR magnetotransmission from all of these samples have been previously studied with emphasis on the donor-related features [15,16].

A zero magnetic field PL spectrum (I_{PL}) from sample 1 in the vicinity of the gap is shown in Fig. 1(a). The PL spectrum exhibits one dominant feature at 1.5411 eV which is attributed to the ground state e_1h_1 excitonic transition. Recombination associated with excitons bound on residual impurities is responsible for the weak asymmetric tail to low energies. Bound excitons tend to dominate the emission spectra in n-type [15] as well as in p-type samples [17]. Figures 1(b) and 1(c) show similar spectra along with energy scans of the ODR signal, ΔI_{PL}, ($\lambda_{\mathrm{FIR}} = 118.8$ μm) at magnetic fields of 2.4 T, the resonant field for the strongest feature attributed to intraexcitonic transitions (see Fig. 2), and 6.2 T, the resonant field for electron CR, respectively. Note that the ODR signal is entirely negative at 2.4 T and positive at 6.2 T.

In Fig. 2 the change in the luminescence intensity, ΔI_{PL}, of the e_1h_1 feature (the ODR signal) from sample 1 is plotted as a function of magnetic field for the four FIR wavelengths used in this study. At $\lambda_{\mathrm{FIR}} = 118.8$ μm the changes in the electron CR correspond to approximately 1% of the peak PL intensity. A variety of resonances are present in these scans. For example, in the 118.8 μm trace there are five features labeled (a) through (e) at the following field values: 0.92, 1.74, 2.42, 6.20, and 11.32 T. Feature (d) corresponds to cyclotron resonance of electrons [4,5,9]. Feature (e) at 11.32 T is attributed to hole cyclotron resonance, transitions between the highest pair of hole ($m_j = 3/2$) Landau levels [18]. Resonances (a) and (c) are associated with intraexcitonic transitions. In this Letter we concentrate on these internal transitions of the e_1h_1 exciton, and reserve detailed comments regarding the hole CR and other transitions for a longer paper. Features (a) and (c) at 0.92 and 2.42 T are

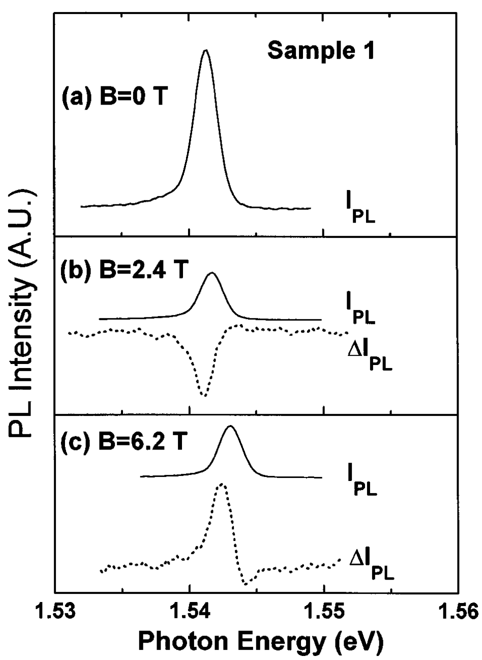

FIG. 1. Photoluminescence intensity I_{PL} (solid lines) and changes ΔI_{PL} (dotted lines) plotted as a function of detected photon energy for sample 1; FIR wavelength = 118.8 μm; $T = 4.2$ K. (a) $B = 0$ T, (b) $B = 2.4$ T (resonance field for the $1s \rightarrow 2p_+$ intraexcitonic transition), (c) $B = 6.2$ T (electron cyclotron resonance).

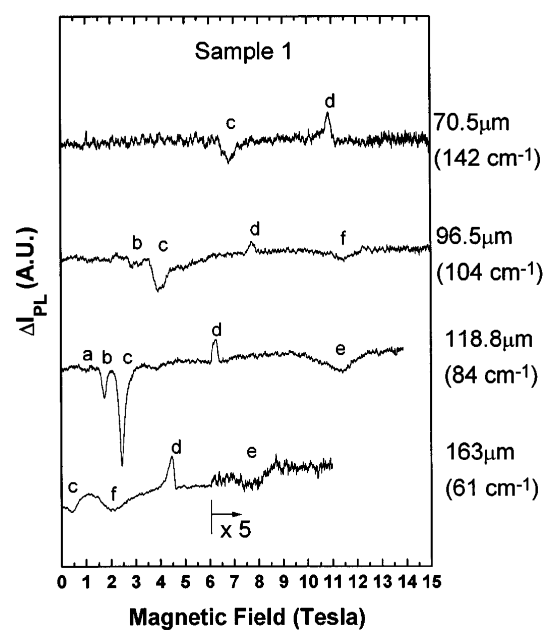

FIG. 2. Changes in the PL signal (ΔI_{PL}) as a function of magnetic field for sample 1 for the following FIR wavelengths: 70.5, 96.5, 118.8, and 163 μm (from top to bottom); $T = 4.2$ K. The wavelength values and their energies in wave numbers are listed to the right of each trace.

identified as the $1s \rightarrow 3p_+$ and $1s \rightarrow 2p_+$ transitions of the e_1h_1 confined exciton, respectively; the origins of features (b) and (f) are uncertain at present. The identification of features (a) and (c) is based primarily on a comparison of the experimental energies of the $1s \rightarrow 2p_+$ transition [feature (c)] with our calculated values, and the energy differences between features (a) and (c). We have used a variational approach to calculate the energies of the $1s$ and $2p_+$ states of an $m_J = \pm 3/2$ (heavy-hole) exciton in $GaAs/Al_{0.3}Ga_{0.7}As$ quantum-well structures in the presence of a magnetic field along the direction of growth as functions of the well width and magnetic field. We use trial wave functions which are products of the electron and hole subband wave functions in a finite quantum well and those which describe the internal motion of the exciton in the presence of a magnetic field. The values of the various physical parameters pertaining to GaAs used in the calculations are [19] $m_e = 0.067$ m, $\epsilon_0 = 12.5$, $m_z^{\mathrm{hh}} = 0.45$ m, $m_{xy}^{\mathrm{hh}} = 0.1$ m, where m_e is the electron effective mass, ϵ_0 is the static dielectric constant, m_z^{hh} is the heavy-hole mass along the growth direction, m_{xy}^{hh} is the heavy-hole mass in the plane perpendicular to the growth direction, and m is the free electron mass. The values of these parameters for the $Al_{0.3}Ga_{0.7}As$ barriers are [20] $m_e = 0.092$ m, $\epsilon_0 = 11.8$, $m_z^{\mathrm{hh}} = 0.54$ m, and $m_{xy}^{\mathrm{hh}} = 0.13$ m. Effects due to hole subband mixing and image charges are not included in these calculations.

The energies of the various ODR features for sample 1 are plotted as functions of magnetic field in Fig. 3(a) and the corresponding features for sample 3 are plotted in Fig. 3(b). The bold solid lines represent the calculated values for the $1s \rightarrow 2p_+$ intraexcitonic transition for these structures. The dotted lines are guides to the eye. As can be seen from Fig. 3(a) and 3(b), the variational calculation describes the data very well for the $1s \rightarrow 2p_+$ transition [features (c)], which lends strong support to the interpretation. The assignment of the weak $1s \rightarrow 3p_+$ transition [feature (a) in Figs. 3(a) and 3(b)] is based on the energy separation between features (a) and (c); the separation is $\gtrsim \hbar\omega_c$. The assignment of the very weak feature (a') observed in sample 3 [Fig. 3(b)] to the $1s \rightarrow 4p_+$ transition is based on similar arguments.

To exclude the possibility that transitions (a), (b), and (c) are associated with residual donors, we have included in this study a MQW structure (sample 2) with dimensions identical to those of sample 1 (grown in the same growth run) but doped with Si donors over the central one-third of the wells. Optically detected resonance spectra from sample 2 contain features (a), (b), and (c), *as well as* the donor-related transitions which (i) are dominant in the ODR spectra, (ii) are clearly well separated in energy, and (iii) give rise to a *positive* ODR signal ($\Delta I_{\mathrm{PL}} > 0$). Furthermore, in the case of sample 1, residual donors are expected to be distributed uniformly throughout the wells, or to accumulate at the heterointerfaces (edges of the wells). A narrow distribution of donors near the interfaces would

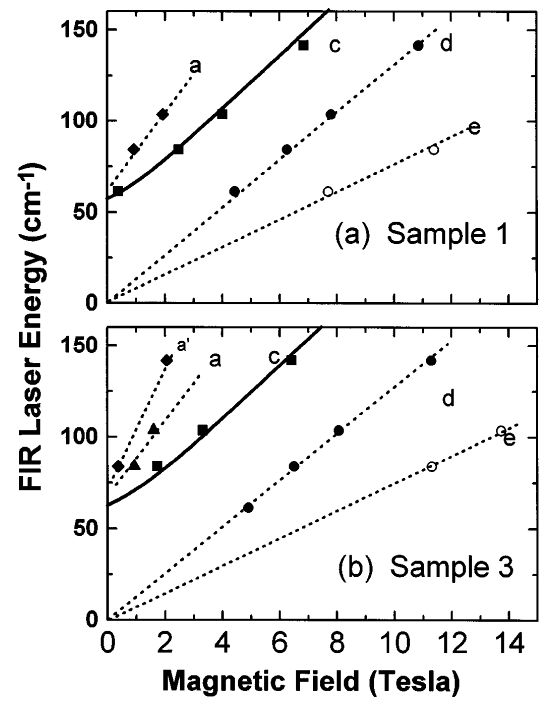

FIG. 3. Energies of the ODR resonances plotted as a function of magnetic field; $T = 4.2$ K. (a) Sample 1. (b) Sample 3. The solid lines represent the results of the variational calculation. The dotted lines are guides to the eye.

lead to lines well below feature (c). A uniform distribution over the wells and barriers leads to a large variation of the donor energies resulting in a line profile with a high energy peak corresponding to impurities near the well centers and a low energy peak corresponding to impurities near the barrier centers [21]. This leads to very broad line profiles for the intraimpurity transitions of uniformly distributed neutral donors; relatively broad lines are observed in the center-one-third-doped wells in the present experiments. In contrast, resonances (a) and (c) are quite sharp, which lends an additional strong argument against assigning them to impurity-related features. Further substantiation of the assignment of features (a) and (c) is provided by comparison of the results from sample 3 (80 Å wells,) which are summarized in Fig. 3(b), with those from sample 1 (125 Å wells) shown in Fig. 3(a). The $1s \rightarrow 2p_+$ transitions for sample 3 are shifted up by approximately 5 – 8 cm^{-1} over the range 6 – 3 T compared with those for sample 1, qualitatively as expected for a narrower well, and in reasonable agreement with the variational calculation.

Returning to the ODR energy scans of Figs. 1(b) and 1(c) at $\lambda_{\mathrm{FIR}} = 118.8\ \mu$m, as noted previously ΔI_{PL} is negative at $B = 2.4$ T [the $1s \rightarrow 2p_+$ intraexcitonic resonance, feature (c) in Fig. 2], while ΔI_{PL} is positive at $B = 6.2$ T [electron CR, feature (d) in Fig. 2]. We interpret the *negative sign* of the intraexcitonic transition as follows: at resonance (2.4 T) the exciton absorbs a FIR laser photon and is promoted to an internal excited state from which it can be easily thermally ionized, thus reducing the population of free excitons available for recombination, increasing the steady state free electron and hole densities, and enhancing the probability of recombination via bound excitons. This process is responsible for the decrease in the e_1h_1 exciton PL intensity. The *positive sign* of the ODR signal or electron CR [Fig. 1(c)] as well as that of the donor-related transition has been discussed previously [11]. The positive change in the e_1h_1 free exciton PL for electron CR is attributed to a resonant heating effect. As the effective temperature of the electrons is increased by resonant absorption of FIR radiation, and this energy is shared among all the carriers and excitons, bound excitons dissociate into free excitons and donors. This carrier heating process increases the free-exciton population, decreases the competing bound exciton recombination channel, and hence leads to an increase in the free-exciton PL intensity (ΔI_{PL}).

The ODR signals (ΔI_{PL}) were observed to be much stronger in the narrower wells (80 and 125 Å) compared to the wider wells investigated. In an undoped 200 Å MQW structure (not included in this Letter due to space limitations), the ODR signals were an order of magnitude weaker than those from the narrowest well studied (80 Å). We have considered two possible explanations for this unexpected behavior. The first concerns the dependence on confinement of the electric dipole matrix elements and oscillator strengths for the internal excitonic transitions We have calculated the electric dipole matrix elements for a strictly two-dimensional (2D) and strictly three-dimensional (3D) hydrogenic system at $B = 0$, and find that the oscillator strengths $\propto \omega_{1s-2p}|\langle 2p|x|1s\rangle|^2$ *decrease* substantially going from 3D to 2D. In addition, a recent calculation of the absorption coefficients of the $1s \rightarrow 2p_+$ transitions of natural donors [21] show a weak *decrease* as the well width is narrowed form 210 to 80 Å. Both calculated results are opposites of the observed behavior, and thus an explanation must be sought elsewhere. Lateral localization by potential fluctuations due to well width variations, which are stronger in the narrower well samples, could provide a possible explanation. For such a situation, excitons dissociated in the presence of larger lateral potential fluctuations would be less likely to reform their excitonic ground state than those experiencing a weakly varying lateral potential landscape, due to the larger probability in the former case of the carriers being localized in spatially separated lateral regions. Further studies are necessary to clarify this point.

In summary, we have used an optically detected resonance technique to study the internal energy structure of e_1h_1 excitons in a series of GaAs/AlGaAs quantum wells. We have observed several of the allowed transitions from the $1s$ ground state to the p-symmetry excited states. The experimental results for the $1s \rightarrow 2p_+$, transition are in good agreement with our calculated values of the transition energies and show the expected increase with decreasing well width. We note that this ODR work complements earlier PLE and magnetoreflectance studies from which the energies of the $1s$ to the s-symmetry excited states can be deduced [22]. The observation of direct intraexcitonic transitions offers the opportunity to study the exciton energy spectrum in semiconductor heterostructures under a variety of circumstances. In addition, the modulation of reflectance or photoconductivity by the FIR laser in related ODR experiments opens the possibility of studying the energies of higher lying confined excitons (e.g., e_2h_2, e_3h_3, etc).

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Note added.—Similar results have recently been obtained independently with a free electron laser as the FIR source [23].

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