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Magneto-optical determination of exciton binding energy in GaAs-$Ga_{1-x}Al_xAs$ quantum wells

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The binding energy of the exciton in GaAs quantum wells confined within Ga-Al-As is determined by the observation of the different behavior of the ground state and the excited states of excitonic transitions of different subbands with excitation spectroscopy in magnetic fields. An increase in the binding energy with decreasing well thickness is found with values higher than theoretically expected. This discrepancy is explained by an experimentally determined higher reduced mass than that used in the theoretical calculations.

In recent years there has been considerable interest in the properties of the quasi-two-dimensional nature of the excitons in quantum wells of GaAs sandwiched between $Ga_{1-x}Al_xAs$ layers.[1-4] This interest stems from the fact that the well thickness can be made of the same order of the exciton Bohr radius. This unique situation allows the experimental study of excitonic states which in that case behave like a quasi-two-dimensional hydrogen atom; this can be observed as an increase of the exciton binding energy for decreasing quantum-well widths. Several authors have shown this effect theoretically with[1-3] variational calculations of the binding energy of a hydrogenic exciton in a quantum well. The observation of the energy difference of the excitonic ground state and the first excited state has been published,[2] and the ground state has been inferred from these data by comparison with variational calculations. However, no direct observation of the exciton binding energy in quantum wells has been reported yet.

In this paper we report the results of excitation spectroscopy experiments of several GaAs quantum wells with thicknesses between 5 and 12.5 nm in high magnetic fields ($B < 23$ T). Under these conditions the exciton continuum splits into discrete excited excitonic states which are weakly bound to the Landau levels and which can, therefore, be described as free electron and hole states. The lowest bound excitonic states on the other hand experience only a comparatively weak diamagnetic shift. The simultaneous observation of the magnetic field dependence of the bound and the continuum states allows a direct determination of the band edge and the exciton ground-state energies and thereby of the binding energy in the quantum well. In addition, information about the masses of the different hole subbands is obtained from field dependence of the interband transitions.

Four samples have been studied with different GaAs layer thicknesses and Al content (see Table I). The intensity of the luminescence of the lowest-energy transition (heavy-hole exciton ground state) is measured as a function of the excitation intensity at different fixed values of the magnetic field. Both the incident and the emitted radiation were at

TABLE I. Sample parameters and experimentally determined masses.

Sample thickness (nm)	Al content	Reduced masses e-HH	Reduced masses e-LH	Hole masses HH	Hole masses LH
5	0.18	0.084	· · ·	> 1	· · ·
9	0.29	0.079	· · ·	> 1	· · ·
10	0.29	0.077	0.057	> 1	0.2 ± 0.1
12.5	0.21	0.069	0.064	0.7 ± 0.2	0.35 ± 0.1

right angles to the layer plane and parallel to the magnetic field axis (Faraday configuration). The exciting light was left or right circularly polarized with respect to the magnetic field. Magnetic fields up to 23 T were generated with a polyhelix resistive magnet. The radiation with wavelengths between 804 and 740 nm was generated with a Kr laser pumped Cr-599 dye laser with LD700 as a dye.

In Fig. 1 the excitation spectra for one circular polarization are shown for different values of the magnetic field. In this case the spectrometer was placed on the center of the luminescence line and the excitation spectra of the higher-lying transitions are observed. The two lower-lying transitions which partly coincide with the luminescence line and which are due to the two exciton ground states associated with the two lowest hole subbands were measured separately with the spectrometer placed in the low-energy tail of the luminescence. The spectra reveal a very rich structure with several strong and weaker peaks. Figure 2 shows a plot of the transition energies versus magnetic field for the two different polarization directions. The results shown are for the 12.5-nm-thick quantum well. In Fig. 2 several sets of transitions can be distinguished. Two transitions at the lowest energies show only a very weak magnetic field dependence, the next transition at higher energy shows a stronger magnetic field dependence and extrapolates approximately to the same energy as the second transition at zero magnetic field. At higher energies the transitions can be distinguished in two sets, each set extrapolating to the same energy at zero

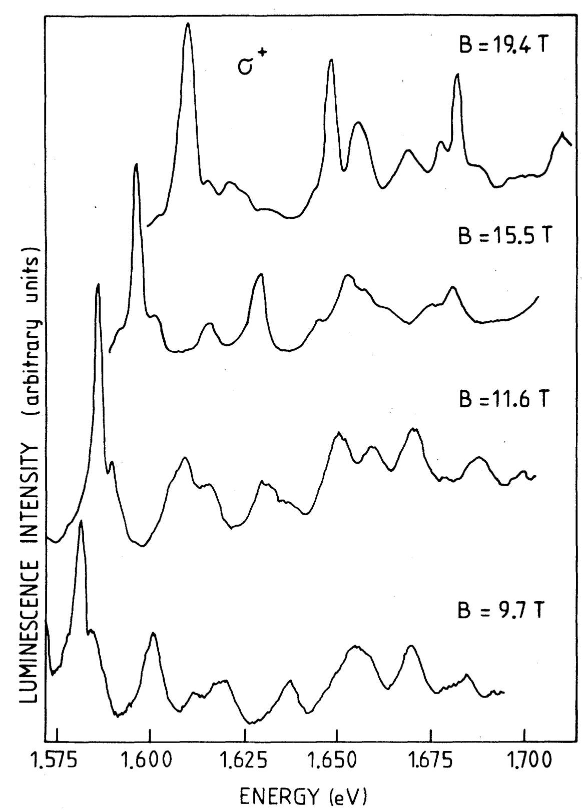

FIG. 1. Excitation spectrum at different values of the magnetic field for the 12.5-nm sample. The spectrometer is placed at the center of the luninescence line and the higher-lying transitions are measured.

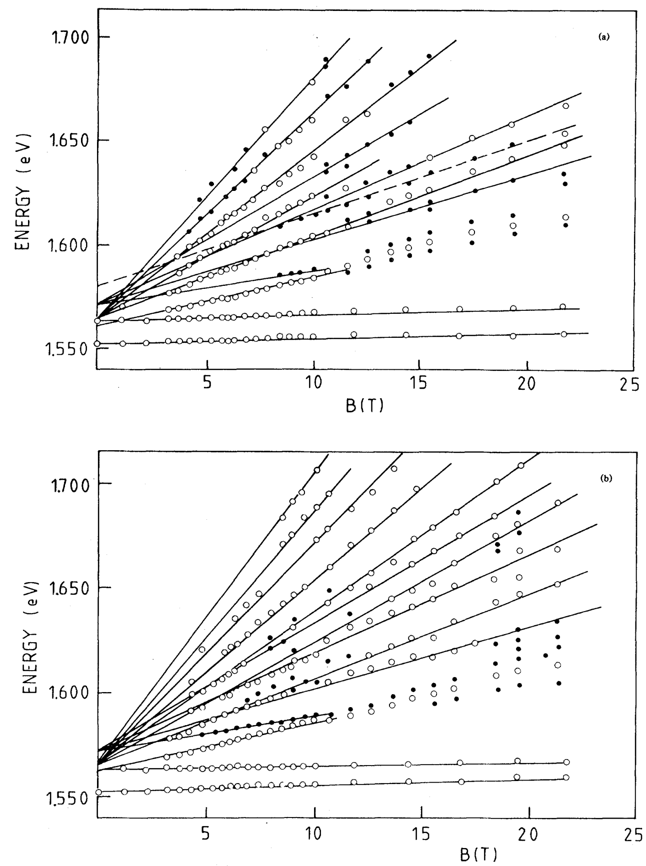

FIG. 2. Energy of the maxima in the excitation spectra as a function of magnetic field for (a) σ^+ and (b) σ^- polarization of the exciting light. The open circles correspond to strong and the full circles to weak transitions. The drawn lines are a guide to the eye.

magnetic field, but to a different energy for both sets. These results clearly show the excitonic character of the absorption process. The two transitions at low energies are interpreted as the exciton ground state for the heavy-hole (HH) and the light-hole (LH) exciton. This ground state is only weakly affected by the magnetic field since the Coulomb energy dominates over the magnetic energy. The higher excited exciton states are much more weakly bound and in this case the magnetic field determines the energy spectrum. Therefore, the two sets of transitions extrapolating to the same energies may be interpreted as essentially free-electron and hole band-to-band transitions. The transition which appears like a splitting of the LH exciton ground state is interpreted as a first excited excitonic state of the HH which for this sample is accidentally degenerate with the LH exciton ground state. It is indeed observed in the thinner samples, where the splitting between the hole subbands is larger, that this transition does not extrapolate to the same energy at zero magnetic field as the LH ground state.

This interpretation of the spectra permits a direct determination of the energy of the continuum and that of the excitonic ground state of different subbands at the same time. Therefore, the experiments measure directly the exciton binding energy. In Fig. 3 the binding energies obtained in this fashion as a function of the well thickness are shown.

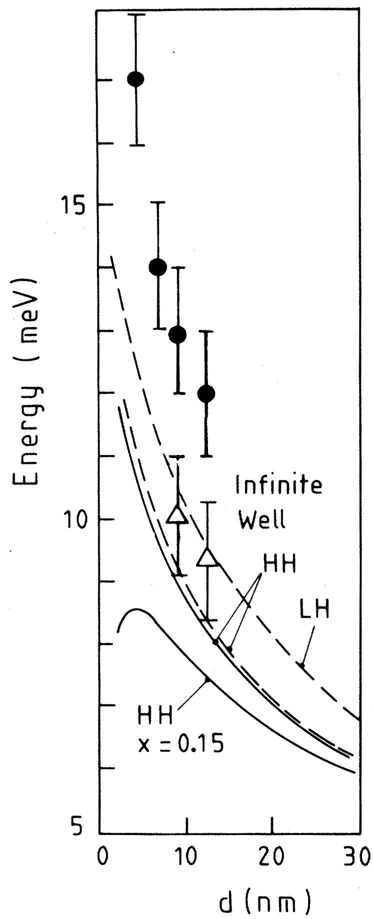

FIG. 3. Exciton binding energy for the HH exciton (closed circles) and the LH exciton (triangles) as determined from magnetic field dependence of the excitation spectra. The lines represent theoretical calculations of the thickness dependence of the binding energy for an infinite well by Miller, Kleinman, Tsang, and Gossard (Ref. 2) (dashed lines) and for infinite and finite height wells by Greene and co-workers (Ref. 3) (solid lines).

Also included in the figure are the results of theoretical calculations by Greene and co-workers and by Miller, Kleinman, Tsang, and Gossard.[2] It can be seen that both experimentally and theoretically an increase in the binding energy is found with decreasing layer thickness. However, the experimental values are much higher than the theoretical ones. In addition, the so-called LH exciton is found to be more weakly bound than the HH exciton, which is contrary to the theoretical predictions. As can be seen from Fig. 2, several Landau-level-like transitions are observed for both subbands. We have analyzed the slopes for the different transitions by plotting them as a function of the Landau-level quantum number and found within experimental error that such a plot yielded a straight line. From the slope of this line a reduced mass for the combined electron and hole Landau-level splitting can be determined and the results are given in Table I. It is clear from this table that the reduced masses for the transitions involving the heavy-hole subband are almost equal to the electron mass, taking nonparabolicity into account, which implies that the heavy-hole mass is much larger than the electron mass. Transitions involving the next hole subband, which can only be observed clearly in two samples, show a reduced mass which is definitely lighter than the electron mass and, therefore, allow a determination of the hole mass of this subband. These experimentally determined reduced masses are strikingly different from those used in the theories to calculate the exciton binding energy, which were $0.04m_0$ for the heavy-hole and $0.51m_0$ for the light-hole exciton. We find that not only the masses are heavier than expected but also that the heavy-hole-electron reduced mass is much heavier than the light-hole-electron mass. It is interesting to note that the experimental findings for the binding energies and the reduced masses are consistent. In a hydrogenic model the binding energy is proportional to the reduced mass. If we, therefore, scale the theoretical curves for the binding energies with a factor which is the ratio of the experimental value of the reduced mass to the theoretical value, both the LH and the HH exciton ground-state energies can be made to agree with the theory within experimental error.

The explanation for the difference in the experimentally determined and theoretically derived hole masses, is believed to be that the theoretical masses are derived from a highly simplified description of the valence band in a quantum well. The bulk valence band in GaAs consists of a degenerate set of $J_z = \pm\frac{3}{2}$ and $J_z = \pm\frac{1}{2}$ bands which are anisotropic in the sense that the mass corresponding to the $J_z = \pm\frac{1}{2}$ state is light in one direction but a mixture of heavy and light in the other two and similarly for the $J_z = \pm\frac{3}{2}$ state. In the theories an average of the light- and heavy-hole mass is determined for the in-plane dispersion relation, leading to a light mass in the heavy-hole subband and a heavier mass in the light-hole subband.[5,6] In this model the two subbands are considered decoupled for any $k_\parallel$ value and, therefore, the two bands can cross at a certain value of $k_\parallel$. However, at finite $k_\parallel$ both bands interact giving rise to strong nonparabolicities. This effect has been shown theoretically by Nedorezov[7] for the valence-band structure of Ge and Si in an infinite potential well, by Bangert, Klitzing, and Landwehr,[8] for p-type silicon inversion layers and recently by Fasolino and Altarelli[9] for the case of a finite well in the GaAs-GaAlAs heterojunction. These more accurate calculations show that the inclusion of the coupling between light- and heavy-hole bands leads to an anticrossing behavior between the bands in the plane of the motion which gives rise to a flattening of the dispersion relation of the heavy-hole subband. In fact, this band is found to be only a few meV wide in a region of k space of order π/d which is the region of interest both for the exciton binding energy and for the magnetic fields used.[9] The masses we deduce from our experiments are in qualitative agreement with this fact. We, therefore, believe that our results provide experimental evidence for this anticrossing behavior and the complex nature of the valence band in quantum wells.

In summary it may be stated that we have measured the exciton binding energies as a function of thickness for GaAs-GaAlAs quantum wells. The experimental binding energies are enhanced with respect to the bulk values and increase with decreasing thickness. The experimental values obtained, however, are higher than theoretical predictions. In addition, we have determined the reduced masses using the magnetic field dependence of the excited states, and have found that these masses are much heavier than those used in the theoretical calculations. Using the experimental values for the reduced exciton masses, theory and experimental results for the binding energies can be reconciled. The observed higher masses are believed to be a direct consequence of the coupling between light- and heavy-hole subbands at finite k values, which has been neglected in the calculations of the binding energies. As such, our results provide experimental evidence for this effect.

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