

Exciton binding energy in $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ strained quantum wells

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(Received 12 March 1990)

In this paper we present experimental and theoretical studies of the exciton binding energy in $\text{In}_x\text{Ga}_{1-x}\text{As}$ strained quantum wells confined within GaAs layers, as functions of the well width and the barrier height. Photoluminescence excitation measurements are performed under a steady magnetic field of up to 6 T and a temperature of 10 K. The Landau-level-related transitions have been identified. By extrapolating the photon energies of the transitions to zero magnetic field, the binding energy of the heavy-hole exciton is obtained for samples with well widths ranging from 65 to 100 Å and values of x from 0.07 to 0.13. A model calculation is carried out in a framework of the variation method. A good agreement between the experimental and calculated results is achieved. The exciton binding energy is found to decrease with increasing well width and decreasing barrier height, which is accounted for in terms of the exciton spatial confinement.

I. INTRODUCTION

In recent years considerable effort has been focused on two-dimensional systems.¹ The fact that the well thickness can be of the same order of the exciton Bohr radius allows the experimental study of excitonic states, which behave like a quasi-two-dimensional hydrogen atom. One of the most important questions regarding the effect of spatial confinement on excitons in quantum wells concerns the free-exciton binding energy E_B . Many authors have presented determinations of E_B for excitons confined in $\text{GaAs}/\text{Ga}_{1-x}\text{Al}_x\text{As}$ quantum wells theoretically and experimentally.²⁻⁹ For the experimental work, two methods are currently used for the direct determination of the exciton binding energy. One is to observe the ground and continuum excitonic states by the photoluminescence-excitation (PLE) measurement;⁴⁻⁶ the other is to infer the *subband gap* by the extrapolation of Landau-level transitions using the magneto-optical measurements.⁷⁻¹⁰ The ultrahigh-quality sample is usually needed for the former approach to resolve the 1s exciton state from the continuum, and the accuracy results from the identification of the continuum states. The latter method faces the difficulty of the free-carrier approximation for the Landau-level transitions and the incorporation of the magnetoexciton states.¹¹

The potential interest in strained semiconductor heterostructures has been demonstrated recently.^{12,13} The electronic structure of $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ strained multiple quantum wells (SMQW's) is modulated both by the strain and periodic potential along the growth direction z . As a result, the valence band exhibits less mixing¹³ due to the large splitting of the valence bands generated by the built-in strain. The possibility of growing device-quality samples with rather large lattice mismatches has provided new prospects for modern material science.

The exciton binding energy in an $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ quantum well has been measured recently by us¹⁴ and by Moore *et al.*¹⁵ using the magnetoabsorption and PLE measurement, respectively. However, systematic understanding of the exciton binding energy in this strained heterostructure is still important. In this paper we report the measurement of the exciton binding energy for $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ strained quantum wells with different well sizes and indium fraction using the magneto-PLE method. E_B is also examined theoretically by the variational calculation. The experimental and calculated results are found to be in very good agreement.

II. EXPERIMENTAL DETAILS

The $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ SMQW samples were grown on Cr-doped GaAs substrates with a (100) surface using a Chinese home-made molecular-beam-epitaxy (MBE) machine. They were prepared under optimum conditions¹⁶ without intentional doping. After depositing a 1- μm GaAs buffer layer at a substrate temperature of 600°C, the $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ strained-quantum-well (SQW) structure was grown at 520°C, followed by a 2000 Å GaAs cap layer. Two kinds of samples are used in the present investigation. One is the 15 alternative periods of $\text{In}_x\text{Ga}_{1-x}\text{As}(80\text{ Å})/\text{GaAs}(150\text{ Å})$ multiple quantum wells with indium fraction x ranging from 0.07 to 0.13; the other consists of four $\text{In}_{0.13}\text{Ga}_{0.87}\text{As}$ single quantum wells in a sequence of 100, 80, 65, and 40 Å along the z direction grown on the same substrate; such a structure allows selective excitation and E_B determination for different wells. The indium fraction was determined using the intensity oscillation of reflection high-energy electron diffraction (RHEED) during growth, and the structural parameters for multiple quantum wells were confirmed by the measurement of double-crystal x-ray diffraction. The

uncertainties in indium fraction and layer thickness are inferred to be ± 0.005 and ± 5 Å, respectively.¹⁶ The photoluminescence (PL) measurements show a very sharp exciton line, typically 3.0 meV, at 1.8 K. PLE measurements were carried out at 10 K under magnetic fields up to 6 T, which was provided by a superconducting solenoid immersed in liquid helium. The excitation light from a 50W broadband tungsten lamp went through a 30-cm-focal-length monochromator and was polarized circularly, σ^+ and σ^- , by a linear polarizer and a quarter-plate. The luminescence was dispersed by a 50-cm-focal-length monochromator, detected by a cooled GaAs cathode photomultiplier, and measured by a photon-counting system. The spectral resolution is 3 Å.

III. EXPERIMENTAL DETERMINATION OF EXCITON BINDING ENERGY

Figure 1 shows the typical magneto-PL spectra obtained from an $\text{In}_{0.13}\text{Ga}_{0.87}\text{As}(80 \text{ \AA})/\text{GaAs}(150 \text{ \AA})$ SMQW structure at several values of magnetic field applied parallel to the growth axis. Several transitions can be well resolved at zero field, attributable to the intrinsic transitions between the electron (e), heavy-hole (hh), and light-hole (lh) subbands. They are assigned according to calculations based on the envelope-function model.¹⁷ When a field is applied, a series of Landau-level-related transitions evolve from the $1e-1\text{hh}$ exciton transition. As

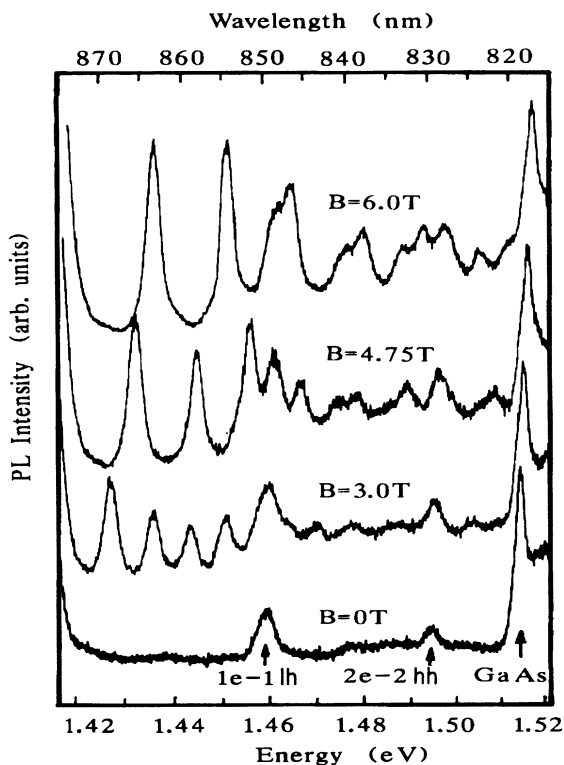


FIG. 1. Typical magneto-PL spectra observed at 10 K with several values of magnetic fields for an $\text{In}_{0.13}\text{Ga}_{0.87}\text{As}(80 \text{ \AA})/\text{GaAs}(150 \text{ \AA})$ SMQW sample. The excitation light is polarized circularly, σ^+ .

seen in Fig. 1, all the peaks move up to the high-energy side rapidly. These structures can be observed at magnetic fields as low as 1.0 T. The $n=0$ Landau-level transition cannot coexist with the $1s$ exciton since unbound electron-hole pairs are not optically active in finite fields. The increase of the PL intensity with the magnetic field is attributed to the enhancement of the density of states by the magnetic field.⁸

In Fig. 2 the transition energies are shown plotted against magnetic fields. All the transitions observed at zero field are slowly shifted up in proportion to the square of the magnetic field due to the diamagnetic effect of the exciton.^{8,10} For the $1e-1\text{hh}$ exciton, the diamagnetic shift (2.1 meV at 6.0 T) is obviously smaller than that of bulk GaAs material. This fact suggests that the transition is due to two-dimensional excitons that experience higher Coulomb binding than three-dimensional ones.^{8,10} On the other hand, the PLE peaks associated with inter-Landau-level transitions show an energy shift almost in proportion to the magnetic field. The least-squares method is used to fit the data of each level to a straight line.

Recently, Rogers *et al.*⁹ analyzed the magneto-optical data of $\text{GaAs}/\text{Ga}_{1-x}\text{Al}_x\text{As}$ quantum wells. They deduced the E_B value by using the semiempirical theory to incorporate the weak Coulomb binding at the high magnetic field and careful linear extrapolation for the low-field data, respectively. They obtained consistent values of E_B by use of these two methods, and also the intercept of the extrapolating lines for low-field data has the same value as the observed onset of the continuum of the exciton.⁹ This means that it is feasible to ascertain the sub-band gap between electron level and heavy-hole level by extrapolation of the low-field data to zero magnetic field. In this case the magnetoinduced states evolving from the

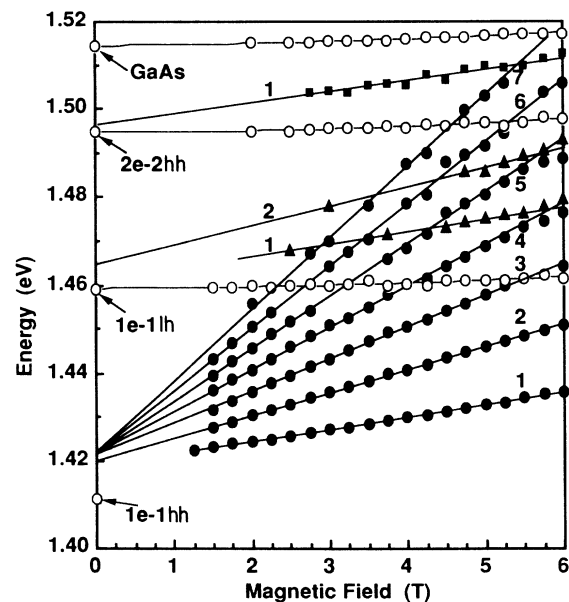


FIG. 2. Transition energies as a function of the magnetic field. The straight lines drawn through the data are the least-squares fitting.

exciton degenerate states are weakly bound to the Landau levels; therefore they can be approximately described as the transitions of free-electron and hole states.

As seen in Fig. 2, the least-squares method gives a good matching of the data to straight lines for all the levels, which are extrapolated from high-energy magnetic fields to 0 T. It is found that the lines from $n \geq 3$ converge at one point with an energy error, namely ± 0.2 meV. An effect of Coulomb interaction can be seen on the energy shifts from $n=1$ and 2, which intercept lower energy than $n \geq 3$ levels do. As previously reported,¹⁴ we interpret that the energy of the convergent point gives the effective band gap between the $1e$ and $1hh$ subbands. Therefore the energy difference between the convergent point and the exciton emission energy at zero field is the binding energy of the free-exciton $1e-1hh$: $E_B = 8.2$ meV. The uncertainties in the experiment typically total an inaccuracy of $E_B \pm 0.4$ meV.

Recent theoretical work^{18,19} has reinterpreted the previous experimental data obtained by Mann *et al.*⁷ and Ossau *et al.*¹⁰ at high magnetic fields in terms of the magnetoexciton states, rather than Landau interband transitions of free carriers. According to their theories, the mixing of the valence band and the magnetoexcitonic nature induce a nonlinear dependence of the transition energy on the magnetic field and anticrossing of excitonic transitions; also, some extra transitions appear in the magneto-optical spectra. As a result, the experiments neglecting the above band effects overestimated the exciton binding energy⁷ compared with calculations^{2,3} and those deduced by other methods.⁴⁻⁶

For $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ strained quantum wells, large lattice mismatch leads to the generation of biaxial compressive strains in the $\text{In}_x\text{Ga}_{1-x}\text{As}$ well layer; the degeneracy of $J = \frac{3}{2}$ valence-band states is therefore lifted by the uniaxial component of the strain into heavy- and light-hole bands. For the above SMQW sample, the layer strain of $\text{In}_x\text{Ga}_{1-x}\text{As}$ is around 1.0% and the splitting caused by the strain is about 60 meV. In this case the light-hole band shifts apart far away from the heavy-hole band, and the light hole is even confined to GaAs layer for higher indium fraction;¹³ hence the valence-band mixing of hh and lh bands is much smaller than that in $\text{GaAs}/\text{Ga}_{1-x}\text{Al}_x\text{As}$ systems within a 6-T field, and is negligible. Moreover, no additional transitions attributable to magnetoexcitons appear in the present measurement because of the special valence-band structure of the $\text{In}_x\text{Ga}_{1-x}\text{As}$ quantum well, and the spectra are identical for the σ^+ and σ^- excitations, so we are quite confident that we shall determine E_B using this method.

For determining the well-width dependence of E_B , the single $\text{In}_{0.13}\text{Ga}_{0.87}\text{As}$ quantum-well sample grown with several different widths (100, 80, 65, and 40 Å) on the same substrate was employed for the magneto-PLE measurement. The observation wavelength was respectively tuned to fit the PL emission from the different quantum wells, and the excitation was performed selectively. Such sample structure and experimental procedure ensure the identity of the indium fraction and experimental conditions, as shown in Fig. 3 for the PL and PLE spectra obtained at 4.0 T. The coupling between the electronic

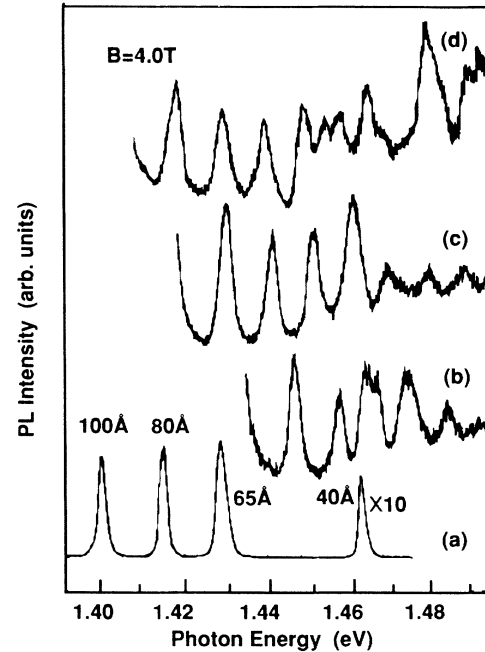


FIG. 3. PL [designated (a)] and PLE [designated (b)–(d)] spectra obtained at 4.0 T for the sample with several single-quantum-well widths (100, 80, 65, and 40 Å) grown on the same substrate. The magneto-PLE spectra were taken by the selective tuning of the observation wavelength of the emission, and they are shown in (b)–(d) for 65, 80, and 100 Å, respectively. The PL efficiency from the 40-Å quantum well is too weak to observe the PLE spectra.

states in the individual quantum wells is prohibited by the wide barrier layer (300 Å). The high quality of the sample allows the observation of PLE spectra from single quantum well even with the lamp excitation. The PLE spectra from the 40-Å QW is very difficult to observe

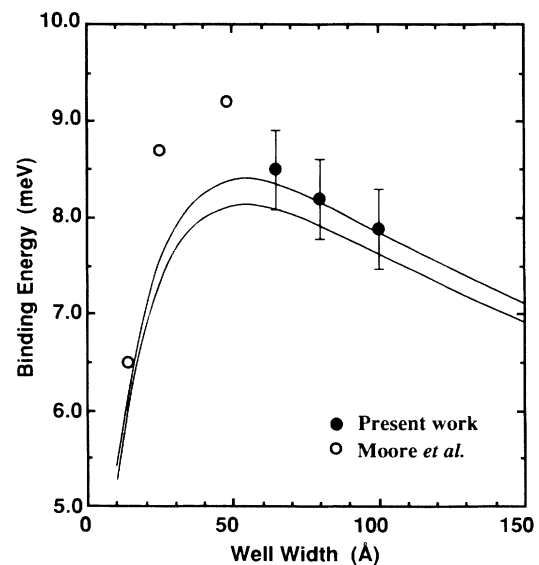


FIG. 4. Exciton binding energy as a function of the well width when the indium fraction is 0.13. The curves are the calculated results using the variational method. The upper curve is for $m_h^|| = 0.22m_0$, the lower for $m_h^|| = 0.18m_0$.

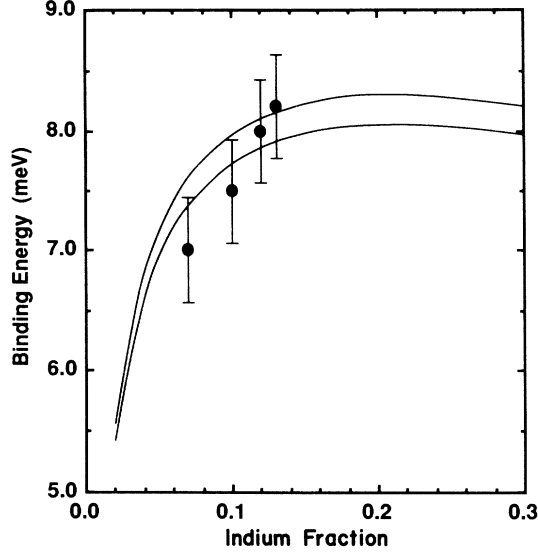


FIG. 5. Exciton binding energy as a function of the indium fraction for 80-Å SMQW's. The curves are the calculated results using the variational method. The upper curve is for $m_h^{\parallel}=0.22m_0$, the lower for $m_h^{\parallel}=0.18m_0$.

since the PL efficiency is 1 order of magnitude lower than the others. The exciton binding energy E_B is deduced by the consideration described above, and the values are plotted against the well width in Fig. 4.

For the $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ system the device-quality material is usually prepared with $x < 0.3$ because of the limitation of the critical layer thickness.¹² Another three 15-period $\text{In}_x\text{Ga}_{1-x}\text{As}(80 \text{ \AA})/\text{GaAs}(150 \text{ \AA})$ multiple-quantum-well samples were measured with an indium mole fraction of $x=0.07, 0.10,$ and 0.12 , respectively. The E_B values for these multiple quantum wells with different indium fractions are plotted in Fig. 5.

IV. THEORETICAL RESULTS AND DISCUSSIONS

The calculation of the exciton binding energy is made in a variational framework in a spirit similar to those reported previously.^{1,2} However, we review some procedures here to include the finite well potential and strain effect. The Wannier exciton in the quantum well is treated in the effective-mass approximation, so the Hamiltonian of the heavy-hole exciton expressed in cylindrical coordinates is³

$$H = \frac{\hbar^2}{2\mu_{\parallel}} \left[\frac{1}{\rho} \frac{\partial}{\partial \rho} \rho \frac{\partial}{\partial \rho} + \frac{1}{\rho^2} \frac{\partial^2}{\partial \phi^2} \right] \left[-\frac{\hbar^2 \partial^2}{2m_e^* \partial z_e^2} - \frac{\hbar^2 \partial^2}{2m_h^* \partial z_h^2} \right. \\ \left. - \frac{e^2}{4\pi\epsilon_0\epsilon_r(\rho^2 + (z_e - z_h)^2)^{1/2}} + V_e(z_e) + V_h(z_h) \right],$$

where m_e^*, m_h^* are the effective masses of the conduction electron and heavy hole along the growth direction (z), respectively, μ_{\parallel} is the exciton reduced mass in the well plane, and ϵ_r is the dielectric constant. $V_e(z_e), V_h(z_h)$ are the quantum potentials for the electron and heavy hole,

respectively. They are assumed to be square wells of width L_z :

$$V_e(z_e) = \begin{cases} 0, & |z_e| < L_z/2 \\ V_e, & |z_e| > L_z/2 \end{cases}$$

$$V_h(z_h) = \begin{cases} 0, & |z_h| < L_z/2 \\ V_h, & |z_h| > L_z/2. \end{cases}$$

Here the origin of the coordinates is chosen to be at the center of the quantum well for the convenience of the calculation. The effect of the biaxial compressive strain in the $\text{In}_x\text{Ga}_{1-x}\text{As}$ layer is included in $V_e(z_e), V_h(z_h)$ in the Hamiltonian. This effect is equivalent to a hydrostatic pressure plus a uniaxial strain along the growth direction, so the heavy- and light-hole bands move away from each other, which can be expressed by

$$\Delta E_{hh} = [-2a(C_{11} - C_{12})/C_{11} + b(C_{11} + 2C_{12})/C_{11}] \epsilon,$$

$$\Delta E_{lh} = [-2a(C_{11} - C_{12})/C_{11} - b(C_{11} + 2C_{12})/C_{11}] \epsilon,$$

where a, b are the deformation potentials. C_{11}, C_{12} are the elastic stiffness constants. ϵ is the strain defined by the free (a_0) and strained (a_1) lattice constants of the $\text{In}_x\text{Ga}_{1-x}\text{As}$ layer, namely $\epsilon = (a_1 - a_0)/a_0$. The GaAs layer is thought to be free of strain since the total thickness is too large compared with $\text{In}_x\text{Ga}_{1-x}\text{As}$ layers.

The band-gap discontinuity for $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ quantum well at 4.2 K is chosen to be $\Delta E_g = 1.5837x - 0.475x^2$ (eV).¹³ The valence- and conduction-band discontinuities are obtained from our recent proposed value $\Delta E_v/\Delta E_g = 0.48$ via magneto-optical measurements.²⁰

In the same spirit as Refs. 2 and 3, we assume that the Hamiltonian is dominated by $V_e(z_e)$ and $V_h(z_h)$, so that the trial wave function for the 1s excitonic state can be constructed as

$$\psi = \varphi_e(z_e) \varphi_h(z_h) g(\rho, \phi, z_e - z_h),$$

where $\varphi_e(z_e), \varphi_h(z_h)$ are the sinusoidal-type wave functions in the finite square quantum well. For instance, the wave function of the first-subband electron level is

$$\varphi_e(z_e) = \begin{cases} \cos k_e z_e, & |z_e| < L_z/2 \\ e^{-\kappa_e |z_e|}, & |z_e| > L_z/2. \end{cases}$$

The wave vectors k_e and κ_e have been determined from the eigenvalue of the first subband calculated in the envelope-function approximation; the boundary conditions used ensured continuity in the particle wave function and current at interfaces.¹⁶ The wave function $g(\rho, \phi, z_e - z_h)$ describes the internal motion of the exciton. We choose the following form for this trial wave function of the 1s state,²

$$g(\rho, \phi, z_e - z_h) = \exp\{-[\alpha^2 \rho^2 + \beta^2 (z_e - z_h)^2]^{1/2}\}.$$

This is a three-dimensional-like hydrogen wave function, where α and β are variational parameters. We then evaluate the expectation value of H , namely E , and,

minimizing E with respect to α and β , the binding energy is obtained by subtracting E from the total first-subband energy of the electron and heavy hole in the quantum wells determined from the above-mentioned subband-level calculation. All the parameters used in the calculation are listed in Table I. The in-plane effective mass of the heavy hole in the $\text{In}_x\text{Ga}_{1-x}\text{As}$ layer, m_h^{\parallel} , is chosen to be $0.18m_0$ and $0.22m_0$, respectively. We have no *a priori* reason to believe that this is correct—support comes from the successful fitting of E_B by Moore *et al.* quoting these values.¹⁵

The exciton binding energy is calculated for the different well widths of $\text{In}_{0.13}\text{Ga}_{0.87}\text{As}$ quantum wells and the various indium fractions of $\text{In}_x\text{Ga}_{1-x}\text{As}(80\text{ \AA})/\text{GaAs}(150\text{ \AA})$ SMQW's. The results are plotted in Figs. 4 and 5 with solid curves.

As shown in Fig. 4, E_B has a strong dependence on the well width. The upper curve is obtained using $m_h^{\parallel}=0.22m_0$ and the lower, $0.18m_0$. The experimental data from Moore's work are also plotted in this figure with open circles. As shown, E_B increases with reduction of the well size and reaches a maximum value at the width of around 55 \AA , which is followed by a rapid drop in the very narrow width limit. This tendency can be accounted for in terms of the spatial confinement of the exciton. As the well width is reduced, the exciton wave function is compressed in the quantum well, which leads to increased binding. However, when the well is too narrow, the delocalization of the exciton wave function into the surrounding barrier (GaAs) layer becomes more important, and this makes the binding energy approach the bulk GaAs value. The penetration of the exciton wave function results especially from the electron, since that of the heavy hole is well confined if the valence-band offset is chosen to be 0.48:0.52. Our experimental data, indicated by solid circles in Fig. 4, agree very well with the calculation.

In Fig. 5 the exciton binding energy is shown to depend on indium fraction from both experimental and theoretical results. This is also easy to understand in terms of the spilling of the exciton wave function into barrier layer. When the indium fraction is small, the well potential is shallow; the exciton, therefore, becomes less confined and approaches the three-dimensional exciton in the GaAs layer when the barrier height diminishes to zero.

It is also notable that the E_B reaches a maximum value at around $x=0.20$ and then drops. In order to under-

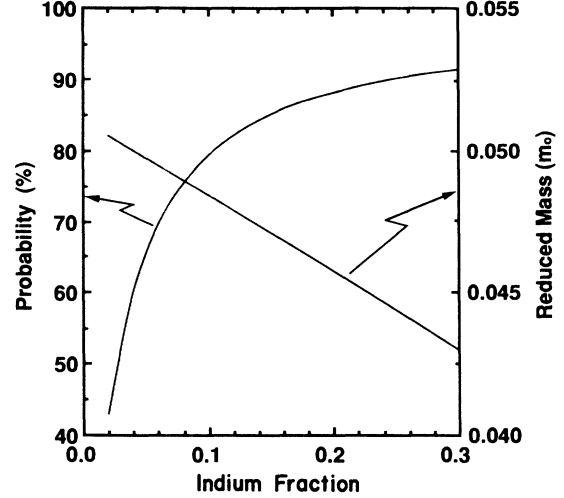


FIG. 6. Probability of finding an electron in the quantum well and the exciton in-plane reduced mass as a function of indium composition.

stand this behavior, both the probability of finding an electron in the quantum well and the exciton in-plane reduced mass are calculated and plotted against the indium fraction in Fig. 6. When the indium fraction is increased, the electron (and therefore the exciton) wave function looks more two dimensional, and the confinement begins to *saturate* towards a value of over 90%. However, the in-plane-exciton reduced mass decreases because the $\text{In}_x\text{Ga}_{1-x}\text{As}$ alloy quantum well samples contain a greater portion of InAs, which has a smaller electron effective mass. These two factors result in a maximum of E_B at $x\sim 0.2$, whereas we can “trust” this dependence in the $\text{GaAs}/\text{Al}_{1-x}\text{Ga}_x\text{As}$ quantum well when the Al fraction is larger than 0.2. Hence, this result seems to be especially important for $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ SQW's because the low-indium-fraction quantum well is usually used. The theoretical results show good quantitative agreement with the experimental determination.

Finally, we briefly comment on our model calculation. Both the changes of the well width and barrier height will affect the electron-subband level significantly, so the stricter consideration should include the nonparabolicity effect of the conduction band. Moreover, the in-plane effective mass of the heavy hole should be taken as a function of x when we calculate the E_B dependence on the indium fraction. Use of smaller m_h^{\parallel} values may lead to reduced E_B for low x , and hence the calculation can be expected to match the experimental data better. Furthermore, the classical-image charges arising from the discontinuity in the dielectric constant at the interfaces is not included in this model calculation.

V. CONCLUSIONS

In summary, we have determined the binding energy of heavy-hole excitons in $\text{In}_x\text{Ga}_{1-x}\text{As}$ strained quantum wells confined in the GaAs layer by magneto-

TABLE I. Physical constants of GaAs and InAs.

	GaAs	InAs
Lattice constant (\AA)	5.6533	6.0583
C_{11} (10^{11} dyn/cm ²)	11.88	8.329
C_{12} (10^{11} dyn/cm ²)	5.38	4.526
a (eV)	-9.8	-5.9
b (eV)	-1.8	-1.7
m_e^*/m_0	0.0665	0.023
m_h^*/m_0	0.34	0.32
ϵ_r	12.5	14.6

photoluminescence-excitation measurements. The special strain-induced band structure in the $\text{In}_x\text{Ga}_{1-x}\text{As}$ well shows that it is feasible to deduce E_B of excitons in terms of Landau interband transitions for low-field data. Therefore the exciton binding energy was measured for samples with different indium fraction and well width, and it is found that E_B depends on both the well size and indium fraction. This is attributed to the delocalization of the exciton wave function as the well width and barrier

height are reduced. A model calculation based on the variational method is carried out, and the result is in good agreement with experimental data.

ACKNOWLEDGMENTS

The authors acknowledge helpful discussions with Dr. G. E. W. Bauer and Dr. J. Kusano.

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