Magneto-optical studies of strain effects on the excitons in $In_x Ga_{1-x} As/Al_v Ga_{1-v} As$ strained quantum wells

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Low-temperature (4.2 K) magnetoreflectance measurements have been performed on a series of $In_xGa_{1-x}As/Al_yGa_{1-y}As$ strained-layer quantum wells (QW's) with the InAs mole fraction x varying from 0 to 0.2, for three different well widths, and for the magnetic-field range of 0–9 T. The diamagnetic shift data of 1s, 2s, 3s,... excitons are fit to a theoretical model to determine the exciton binding energies and the in-plane reduced effective masses, which are presented as a function of x, for excitons in 80, 120, and 340 Å $In_xGa_{1-x}As/Al_yGa_{1-y}As$ QW's. In the case of high-barrier QW's, the binding energies and effective masses appear to decrease linearly with increasing x over the range studied. The rate of change of the effective mass μ as x increases from 0 to 0.2 is experimentally determined, using two different methods, to be $d\mu/dx = -0.057m_0$ and $d\mu/dx = -0.041m_0$, which exhibits a substantial strain dependence when compared with the value $d\mu/dx = -0.07m_0$, from unstrained bulk $In_xGa_{1-x}As$. A theoretical approach taking into account the effect of composition change in the QW and strain on both the conduction and valence bands is made and gives $d\mu/dx = -0.051m_0$, which is in good agreement with our experimental result.

I. INTRODUCTION

 $In_xGa_{1-x}As$ strained quantum-well (QW) heterostructures, grown on GaAs or InP substrates, have demonstrated high performance in high-speed electronic and photonic applications. This QW system has many interesting advantages over GaAs/Al_xGa_{1-x}As QW's including small effective mass, small band gap, large band offset, and lack of DX centers. For opto-electronic device design and modeling, accurate values of crucial parameters, such as effective masses, and exciton binding energy are needed. However, these parameters are typically determined by a semi-empirical interpolation from the values of InAs and GaAs. This interpolation may be invalid due to the effect of strain in a pseudomorphic QW. Recently, many experimental and theoretical works have been done to determine the exciton's binding energies, effective masses, and the g factor, etc., in $In_xGa_{1-x}As/GaAs$ QW systems. I^{1-6} In this system, as in InAs mole fraction x increases, the strain increases along with the effects of changing the barriers height and the well band gap, and all these will affect the exciton binding energy and the effective mass. Especially for small x (x < 0.2), the change in barrier height has a major effect on the exciton's behavior in the very shallow QW which makes the study of the effect of strain more difficult. In this work, high-barrier (y = 0.3 to 1) $In_xGa_{1-x}As/Al_yGa_{1-y}As$ QW's are chosen to determine the effective mass and binding energy as functions of x, from a magneto-optical experiment. The QW's are deep enough to neglect the effect of barrier height change. Therefore, the effect of strain can be estimated using a comparison with theoretical calculation.

II. EXPERIMENT

Three series of unintentionally doped $In_xGa_{1-x}As$ QW $(0 \le x < 0.2)$ samples were grown on GaAs substrates by molecular-beam epitaxy (MBE). As listed in Table I, the first series has 80-Å-wide $In_xGa_{1-x}As$ QW's, with $Al_{0.3}Ga_{0.7}As$ barriers; the second series has 120-Å QW's, and the third series has 340-Å QW's with GaAs barriers.

Low-temperature (4.2 K) magneto-reflectance measurements were taken for each of the samples, with a 0-9 T magnetic field, applied perpendicular to the sample's surface. A monochromator and lamp combination was used to perform the reflectivity experiments with a resolution of ~ 6 Å for the slits used. A divided, multiple-opticalfiber system was used to get the light beams in and out of the magnetic cryostat. Photoluminescence (PL) and xray-diffraction measurements confirmed the sample quality and the InAs mole fraction (within a $\pm 5\%$ error bar). Figure 1 shows the magnetoreflectance spectrum of the 80-Å-wide $In_{0.17}Ga_{0.83}As$ QW, for 0 to 9 T, and is representative of the spectra from all the other samples. The energy of the reflectance peaks as a function of magnetic field is shown in Fig. 2. Five transitions indicated by squares are identified as 1s, 2s, 3s, 4s, and 5s exciton states for the n = 1 electron and n = 1 heavy hole (first confined levels) in the QW. The transitions indicated by triangles are interpreted as 1s exciton states of the n = 1electron and n = 1 light hole in the quantum well. This interpretation is based on the zero-field calculation of the strained quantum well using the three-band Kane model.^{7,8} The peak (or valley) energy is slightly higher than the PL peak energy as expected. Although the peak (or valley) position in the reflectance is not necessarily the

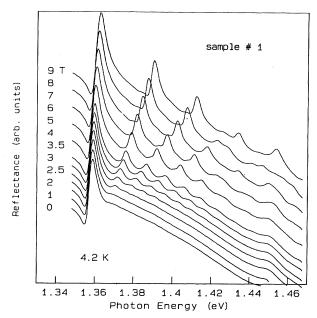


FIG. 1. Magnetoreflectance spectra of $\text{In}_x \text{Ga}_{1-x} \text{As QW sample no. 1}$ (x=0.17) at 4.2 K, for several magnetic fields from 0 to 9 T.

exact transition energy, the relative shift of the peak position with magnetic field should be the same as the shift of an exact transition energy.⁹

III. RESULTS AND DISCUSSIONS

A. Determination of the exciton binding energy and the effective mass by fitting the experimental diamagnetic shift data to a theoretical model (method A)

Yang and Sham¹⁰ have pointed out that many earlier studies using Landau levels to identify the excited states of the magnetoexciton were incorrect. In order to obtain the accurate binding energy and effective mass, we used a model based on the effective-mass theory of Duggan, 11 which simultaneously accounts for the effects of quantum-well confinement, magnetic field, and the Coulomb potential of the exciton, using a separation of variables approximation. The details of the calculation and assumptions are given in Ref. 11. Only the reduced effective mass μ is a free parameter; all other parameters are fixed. The calculated results for the heavy-hole excitons are shown by the solid lines in Fig. 2 for the sample. The theoretical results are in very good agreement with the experimental data for 1s, 2s, and 3s states for the narrow (80 and 120 Å) wells. The high-field data do not fit the theoretical curve very well for higher excited states. This is probably due to the interaction with light-hole excitonic states, or with the transition from higher subband states (n = 2, 3, etc.) for the wide well samples. By concentrating on curve fitting for the lower states, we have determined the binding energy and the in-plane reduced effective mass for each sample. The results are listed in Table I. (The error bars were determined graphically

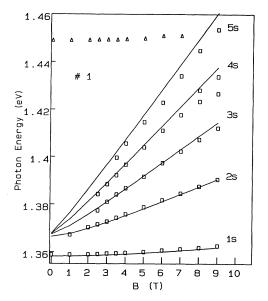


FIG. 2. The plot of the reflectance peak energies of 1s, 2s, ... 5s exciton states vs the magnetic field B for sample no. 1. \Box denotes the data points; the solid lines are the theoretical fits.

with conservative upper and lower bounds.)

The binding energy as a function of the InAs mole fraction x is shown in Fig. 3. The squares are the data points for the 80-Å QW's which have a well width smaller than the exciton Bohr radius (120-130 Å); the diamonds are data from the 120-Å QW's which have a well width comparable to the exciton radius; crosses are data from the 340-A QW's which have a well width much larger than the exciton radius. 12 For the case of $In_xGa_{1-x}As/Al_yGa_{1-y}As$ QW's (80 and 120 Å), the binding energy decreases (near linearly) as the In As mole fraction increases. We obtained $E_B(x) = (-9.33x + 13.0)$ meV for 80-A QW's, and $E_B = (-6.10x + 10.3)$ meV for 120-Å QW's (note that for 340-Å QW's the error bars are too big to give an accurate determination). This result shows a different behavior than that from an $In_xGa_{1-x}As/GaAs$ system reported by Hou et al.² In their results, for small x, the In_xGa_{1-x}As/GaAs QW's are very shallow; the barrier height dependence of the binding energy dominates the change, which gives a rapid increase in binding energy near x = 0, this increase slowing down near x = 0.15, and then decreasing for x > 0.19. In our case, the QW's (with Al_{0.3}Ga_{0.7}As barriers) are deep enough, so that the barrier height dependence is not important, only the change of composition (gap) and strain contribute in the change of binding energy. Therefore, the rate of change in binding energy in our system is similar to that of high In concentration In_xGa_{1-x}As/GaAs QW's calculated by Hou et al.2

The well width dependence of the binding energy has been studied previously for GaAs QW's (Ref. 13) and $In_xGa_{1-x}As$ QW's, ^{1,3} which shows that the binding energy increases for well widths from 0 to \sim 60 Å, then decreases beyond \sim 60 Å until the three-dimensional case is

TABLE I. The list of	$In_xGa_{1-x}As/Al_yGa_{1-y}As$ quantum-well samples	. d_w is the well width, μ is
the reduced mass, and E_1	is the binding energy.	

		d_w		No. of		E_B
Sample No.	x	(A)	у	wells	$\mu(m_0)$	(eV)
1	0.17	80	0.3	1	0.047 ± 0.001	10.4 ± 0.2
2	0.12	80	0.3	1	0.050 ± 0.001	10.8 ± 0.2
3	0.07	80	0.3	1	0.053 ± 0.001	11.3 ± 0.2
4	0.11	80	0.3	10	$0.051 {\pm} 0.001$	11.0 ± 0.2
5	0.025	80	0.3	10	$0.057 {\pm} 0.001$	11.8 ± 0.2
6	0.09	80	0.3	10	0.052 ± 0.001	11.1 ± 0.2
7	0	80	1	10	0.058 ± 0.001	12.0±0.2
8	0.1	90	0	10	0.051 ± 0.001	10.7 ± 0.2
9	0.05	120	0.3	1	0.054 ± 0.001	10.0 ± 0.2
10	0.1	120	0.3	1	0.049 ± 0.001	9.7 ± 0.2
11	0.15	120	0.3	1	0.052 ± 0.001	$9.4{\pm}0.2$
12	0.1	340	0	1	0.051 ± 0.005	6.2 ± 0.6
13	0.05	340	0	1	0.054 ± 0.005	$6.4 {\pm} 0.6$
14	0.084	340	0	1	0.050 ± 0.005	6.1 ± 0.6
15	0.116	340	0	1	0.051 ± 0.005	6.3±0.6

reached. For any given In concentration as shown in Fig. 3 for the case of quasi-two-dimensional (80 and 120 Å) and quasi-three-dimensional (340 Å) systems, a similar behavior can be found.

The reduced effective mass μ as a function of InAs mole fraction x is plotted in Fig. 4 for 80-, 120-, and 340-Å QW's. The effective mass does not depend strongly on the QW well width to first order. As expected all three data sets follow closely one curve showing a near linear decrease of effective mass with increasing x, for x < 0.2. Fitting all the data to a straight line (the solid line) gives an (average) in-plane reduced effective mass as a function of InAs mole fraction for GaAs-based $\text{In}_x\text{Ga}_{1-x}\text{As}$ pseudomorphic QW: $\mu = (-0.057x + 0.057)m_0$, with $d\mu/dx = -0.057m_0$.

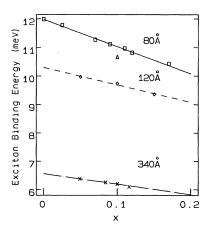


FIG. 3. The plot of the exciton binding energy vs the InAs mole fraction x for 80 Å (\square), 90 Å (\triangle) 120 Å (\diamondsuit), and 340 Å (\times) In_xGa_{1-x}As QW's. The solid lines and dashed line are the least-squares fits.

B. A second method (method B) to determine the decreasing rate $d\mu/dx$ from our experiment

For low magnetic field, the energy shift for a 1s magnetoexciton state can be written as 14

$$\delta E_{1s} = \frac{D\epsilon^2 \hbar^4}{2c^2 e^2 \mu^3} H^2 \tag{1}$$

in cgs units. Here, ϵ is the dielectric constant and D is a parameter depending on the dimensionality of the system. (Other parameters are defined in Ref. 14.) The energy shifts parabolically with the magnetic-field H and is inversely proportional to the cube of the reduced effective mass μ . The diamagnetic shift data for the 1s exciton is fitted to $E_{1s} = \alpha B^2 + E_0$ to obtain the factor α . Figure 5

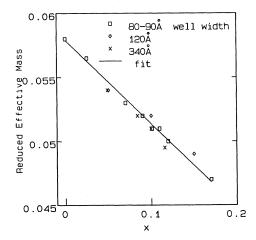


FIG. 4. The plot of the reduced effective masses μ/m_0 as a function of the InAs mole fraction x for the In_xGa_{1-x}As QW's with different well widths. The solid line is the fit of the experimental data.

shows a plot of α versus x for the 80-Å QW's. We can assume that for a given QW width ϵ does not depend on x $(0 \le x < 0.2)$, so $d\mu/(\mu dx) = -d\alpha/(3\alpha dx)$. Using a least-squares fit to the data in Fig. 5, we obtain from the slope $d\alpha/dx = 6.9$, which yields $d\mu/dx = -0.041m_0$.

The difference between the two results of $d\mu/dx$ from methods A and B reflects the different assumptions (approximations) made in each theoretical model. In method A, the errors may come from the difference in finite barrier heights, parabolic band assumption, and the separation of the variable approximation. But all of these should be second-order corrections. The errors for method B may come from the low-field limit approximation, which considers the energy shift to be parabolic and the change in ϵ with x. Since method A fits the experimental data for 1s, 2s, and 3s states from 0-9 T, and method B uses only the 1s state in the low-field data, the result from method A should be more reliable. The rates of change in the effective mass found from both methods are significantly smaller than that of bulk $In_xGa_{1-x}As$ $(d\mu/dx = -0.07m_0$ which will be derived in a later section).

C. A theoretical estimation of the change of the effective mass, and comparison with the experimental result

Since

$$\frac{1}{\mu} = \frac{1}{m_e} + \frac{1}{m_{h\perp}} , \quad \frac{d\mu}{\mu^2} = \frac{dm_e}{m_e^2} + \frac{dm_{h\perp}}{m_{h\perp}^2} , \tag{2}$$

where m_e is the electron effective mass and $m_{h\perp}$ is the heavy in-plane effective mass. Both electron and hole terms need to be evaluated. According to Kane's $\mathbf{k} \cdot \mathbf{p}$ theory, in first-order approximation, the change in effective mass is proportional to the change of band gap E_g , 15

$$\frac{dm_e}{m_e} \simeq \left[\frac{1}{E_g + \Delta} + \frac{2\Delta}{E_g (3E_g + 2\Delta)} \right] dE_g \simeq \frac{dE_g}{E_g}$$
 (3)

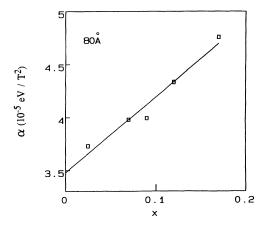


FIG. 5. The plot of α (the diamagnetic shift factor) vs the InAs mole fraction x for the 80-Å $In_xGa_{1-x}As$ QW's.

because for x < 0.2, the spin-orbit split-off energy Δ is much smaller than E_g .

As x increases, the band gap of the bulk $In_xGa_{1-x}As$ decreases, as does the effective mass. For a strained QW, two effects modify the band gap as x changes, the decrease in the band gap due to the composition change, and changes from the deformation potential due to the pseudomorphic strain. Since the strain-induced potential change increases the band gap of the pseudomorphic stained QW with respect to the bulk gap of $In_xGa_{1-x}As$, the rate of decrease of the reduced mass in the QW is expected to be smaller than that of the bulk materials.

Now consider the strain effects on the effective mass. The theoretical work of biaxial strained bulk and superlattice materials has been done by many authors 8,16 using more complex $\mathbf{k} \cdot \mathbf{p}$ theory. For biaxial strain on the $\{001\}$ plane, one can diagonalize a 8×8 matrix of the $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian including the effect of strain 16 and QW confinement. The eigenvalue for the energy gap between the conduction band and heavy-hole band can be written as

$$E_g(e-hh) = E_g^0(x) + \delta E_H^{(00)} + \delta E_S^{(001)} + \delta E_{QW}$$
, (4)

where $E_g^0(x)$ is the band gap for bulk $In_xGa_{1-x}As$, with 17

$$E_{\sigma}^{0}(x) = 1.5192 - 1.47x + 0.375x^{2}$$
 (5)

 $\delta E_H(001)$ is the "hydrostatic" term of the energy shift, and $\delta E_S(001)$ is the "shear" term of the energy shift

$$\delta E_H^{(001)} = a(e_{xx} + e_{yy} + e_{zz}),$$

$$\delta E_S^{(001)} = \frac{b}{2} [2e_{zz} - (e_{xx} + e_{yy})]$$
(6)

where a and b are the deformation potentials; e_{xx} , etc., are the strains with $e_{xx} = e_{yy}$, $e_{zz} = -\lambda e_{xx}$, and $e_{xy} = 0$ for biaxial strain, where $\lambda = 2C_{12}/C_{11}$, C_{11} and C_{12} are the elastic modulus. $\delta E_{\rm QW}$ is the quantum confined energy of the first subbands which depends weakly on x in our case. The rate of change in the electron effective mass can still be found from Eqs. (3) and (6),

$$\frac{dm_e}{m_e dx} \simeq \frac{1}{E_g} \left[\frac{dE_g^0(x)}{dx} + \left[a(2-\lambda) - b(1+\lambda) \right] \frac{de_{xx}}{dx} \right]. \tag{7}$$

For x < 0.2, we can use the parameters¹⁸ a = -9.7 eV, b = -1.7 eV, $\lambda = 0.89$, $m_e = 0.067m_0$, $E_g = 1.5$ eV, and $de_{xx}/dx = -0.072$, and use Eq. (7) where we obtain the rate of change for the electron mass $dm_e/dx = -0.44m_0$.

For the in-plane heavy-hole term, using the commonly accepted value of the heavy-hole mass parallel to the [100] direction $(m_h\|=0.34m_0)$ (Ref. 19) and the Luttinger-Kohn band parameters, ¹⁸ the in-plane heavy-hole mass can be determined as $m_{h\perp}=0.11m_0$, which is not much bigger than the electron mass; ²⁰ thus, the heavy-hole term cannot be neglected. The strain-induced change on the in-plane heavy-hole mass needs to be determined. Since the heavy-hole and light-hole bands are widely separated due to the narrow well confinement and compressive strain, we can neglect the coupling between

the heavy- and light-hole bands near the zone center (Γ point). In this case, the in-plane heavy-hole effective mass can be written as 16

$$\frac{1}{m_{h\perp}^{(001)}} = \left[A' + \frac{P^2}{3E_g} \right] + \frac{1}{2} \left[B' + \frac{P^2}{3E_g} \right],$$
with $P = i\sqrt{2/m_0} \langle s | P_x | X \rangle$, (8)

where A' and B' are the valence-band parameters for the material without strain, P is the matrix element (not dependent on strain). From Eq. (8) we obtain

$$\frac{dm_{h\perp}}{m_{h\perp}dx} = \frac{dE_g(e - hh)}{E_g(e - hh)dx} . \tag{9}$$

Using Eqs. (4)–(9), we find the following rate of change for in-plane heavy hole: $dm_{h1}/dx = -0.073m_0$. The heavy-hole term in Eq. (2) is, therefore, not negligible.

Finally, using Eq. (2) we obtain the theoretically estimated change of the reduced effective mass, $d\mu/dx = -0.051m_0$, which is in agreement with our experimental results, $d\mu/dx = -0.057m_0$ (method A) and -0.041 (method B), for x < 0.2. Using the same estima-

tion without the strain effects, we find for the case of bulk $In_xGa_{1-x}As$, $d\mu/dx = -0.07m_0$. W conclude that the strain effects on both the electron and heavy-hole inplane effective mass are important in strained QW's.

IV. SUMMARY

We have studied the magnetoexciton in strained-layer $In_xGa_{1-x}As/Al_yGa_{1-y}As$ QW's for quasi-two-dimensional and quasi-three-dimensional cases by magnetoreflectance. The experimental data of the exciton binding energies and the effective masses as a function of InAs mole fraction are provided. For small x (<0.2), the rate of change of the effective mass obtained from our experiment is in good agreement with the rate calculated using a $\mathbf{k} \cdot \mathbf{p}$ theory including the effects of strain on both the conduction and valence bands. Substantial strain effects on the effective mass are recognized.

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Bohr radius which is ~ 130 Å. However, according to this reference, the error made for determining the binding energy in the 340-Å wells is only $\sim 10\%$. Also, this error does not affect the rate of decrease of the binding energy with increasing x; G. Bastard, E. E. Mendez, L. L. Chang, and L. Esaki, Phys. Rev. B 26, 1974 (1982).

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¹K. J. Moore, G. Duggan, K. Woodbridge, and C. Roberts, Phys. Rev. B 41, 1090 (1990).

²H. Q. Hou, Y. Segawa, Y. Aoyagi, S. Namba, and J. M. Zhou, Phys. Rev. B 42, 1284 (1990).

³N. J. Pulsford, R. J. Nicholas, R. Warburton, G. Duggan, K. J. Moore, K. Woodbridge, and C. Roberts, Phys. Rev. B 43, 2246 (1991).

⁴D. C. Reynolds, K. R. Evans, C. E. Stutz, and P. W. Yu, Phys. Rev. B 43, 4244 (1991).

⁵M. J. L. S. Haines, N. Ahmed, S. J. A. Adams, K. Mitchell, I. R. Agool, C. R. Pidgeon, B. C. Cavenett, E. P. O'Reilly, A. Ghiti, and M. T. Emeny, Phys. Rev. B 43, 11 944 (1991).

⁶M. J. Lawless, R. J. Warburton, R. J. Nicholas, N. J. Pulsford, K. J. Moore, G. Duggan, and K. Woodbridge, Phys. Rev. B 45, 4266 (1992).

⁷G. Bastard, Phys. Rev. B 12, 7584 (1982), and references cited therein.

⁸D. L. Smith and C. Mailhiot, Rev. Mod. Phys. 62, 173 (1990), and references cited therein.

⁹A. F. Terzis, X. Liu, A. Petrou, B. McCombe, M. Dutta, H. Shen, D. Smith, M. Cole, M. Taysing-Lara, and P. Newman, J. Appl. Phys. 67, 2501 (1990).

¹⁰S. R. Yang and L. J. Sham, Phys. Rev. Lett. 58, 2598 (1987).

¹¹G. Duggan, Phys. Rev. B 37, 2759 (1988).

¹²Remark: the separation of the variable approximation is no longer good for a well width larger than 1.4 times the exciton

¹³G. Bastard and J. A. Brum, IEEE J. Quantum Electron. **QE-22**, 1625 (1986).

¹⁴D. C. Rogers, J. Singleton, R. J. Nicholas, C. T. Foxon, and K. Woodbridge, Phys. Rev. B 34, 4002 (1986).

¹⁵T. Ando, A. B. Fowler, and F. Stren, Rev. Mod. Phys. 54, 437 (1982).

¹⁶T. P. Peasall, F. H. Pollak, J. Y. Marzin, J. M. Gerard, P. Voisin, J. A. Brum, R. People, A. Jackson, and M. Jaros, in *Semiconductors and Semimetals*, edited by T. P. Peasall (Academic, Boston, 1990), Vol. 32.

¹⁷S. H. Pan, H. Shen, and F. Pollak, Phys. Rev. B 38, 3375 (1988).

¹⁸D. Bimberg et al., in Physcis of Group IV Elements and III-V Compounds, edited by O. Madelung, M. Schulz, and H. Weiss, Landolt-Börnstein, New Series, Group III, Vol. 17, Pt. a (Springer, New York, 1986).

¹⁹R. C. Miller, D. A. Kleimen, and A. C. Gossard, Phys. Rev. B 29, 7085 (1984).

²⁰W. Zhou, D. D. Smith, H. Shen, J. Pamulapati, M. Dutta, A. Chin, and J. Ballinggall, Phys. Rev. B 45, 12 156 (1992).