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Comparison of (111)- and (001)-grown GaAs-$Al_xGa_{1-x}As$ quantum wells by magnetoreflectance

Weimin Zhou, Doran D. Smith, H. Shen,* J. Pamulapati, and M. Dutta
U.S. Army ETDL, Fort Monmouth, New Jersey 07703

A. Chin and J. Ballingall
General Electric Corporation, Syracuse, New York 13221
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Magnetoreflectance studies on GaAs/$Al_xGa_{1-x}As$ quantum wells grown on (111) and (001) substrates have been performed in magnetic fields from 0 to 9 T at 4.2 K. Excitonic transitions of 1*s*, 2*s*, 3*s*, . . . states are observed and identified for the transitions from the first heavy-hole subband to the first electron subband and from the first light-hole subband to the first electron subband in the well. From the diamagnetic shifts, we have determined the reduced effective masses, $\mu_\perp$, and the binding energies for excitons in the well using several different methods. A comparison of the reduced masses of the excitons in the (111) and (001) samples determined experimentally gives a ratio of $\mu_{111\perp}/\mu_{001\perp}=0.91$. We have also attempted to deduce the heavy-hole and light-hole effective masses perpendicular and parallel to the [111] axis, and the band parameters.

Magnetoexcitons in bulk GaAs and in GaAs-$Al_xGa_{1-x}As$ quantum wells (QWs) have been studied extensively for more than two decades.[1–12] However, because of the complexity of the heavy-hole (hh) and light-hole (lh) valence-band dispersions and their mixing near the zone center of the bulk GaAs, there are still large discrepancies between the experimental results of hole masses and different theoretical approaches. Experimentally, well-defined hh and lh effective masses can be obtained only by decoupling them using stress, high magnetic field, QW structures, etc.; otherwise, errors are added due to the mixing effects. In this work, we present results of high-resolution magnetoreflectance studies on high-quality (111)-grown GaAs-$Al_xGa_{1-x}As$ QWs. For comparison, we have also repeated the measurement on a GaAs-$Al_xGa_{1-x}As$ QW grown on a (001) substrate. Several methods based on improved theories were used for data analysis. Our results provide the complementary valence-band information on the (111)-grown GaAs QW.

Figure 1 shows 4.2 K magnetoreflectance spectra of a GaAs-$Al_xGa_{1-x}As$ multiple quantum well (MQW) grown on (111) substrates at a varying magnetic field from 0 to 9 T, applied perpendicular to the quantum wells (parallel to the growth axis). This sample (No. 1) was grown by molecular-beam epitaxy (MBE) with ten 100 Å (nominal) wells and 100 Å barriers of $x \simeq 0.27$ (x was verified by photoreflectance measurement). A monochromator and lamp combination was used to perform the reflectivity experiments with a resolution of ~ 3 Å for the slit used. An optical fiber was used to get a light beam in and out of the magnetic cryostat. We have also performed photoluminescence (PL) measurements which showed a peak with a ~ 5-meV linewidth at 1.546 ± 0.005 eV, confirming the high quality of this sample.

The energy of reflectance peaks as a function of magnetic field are shown in Fig. 2. Although the peak (or valley) position is not necessarily the exact transition energy, the relative shift of the peak position with magnetic field should be the same as the shift of the exact transition energy.[13] The lowest transition ($\diamond$) is the excitonic transition between the electron at the conduction-band edge and hole at the valence-band edge in the bulk GaAs. Five transitions indicated by squares are identified as 1*s*, 2*s*, 3*s*, 4*s*, and 5*s* exciton states for $n=1$ electron and $n=1$ heavy hole (first confined levels) in the quantum well. The transitions indicated by crosses are interpreted as 1*s*, 2*s*, 3*s*, and 4*s* exciton states of $n=1$ electron and $n=1$ light hole in the quantum well. This interpretation is based on the zero-field calculation of the confinement energies in the quantum well using the three-band Kane model.[14]

We have repeated the magnetoreflectance measurements on a (001)-grown GaAs-$Al_xGa_{1-x}As$ MQW, sample No. 2, with the same well width as sample No. 1. The reflectance peak energies versus the magnetic field are shown in Fig. 3. To determine the exciton binding energy and reduced effective mass, two different analyses, (a) and

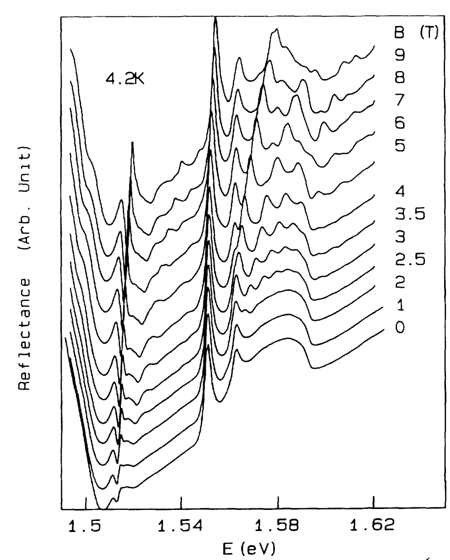

FIG. 1. Magnetoreflectance spectra of the (111)-grown GaAs quantum-well sample No. 1 at 4.2 K. The magnetic field B is applied parallel to the [111] growth direction.

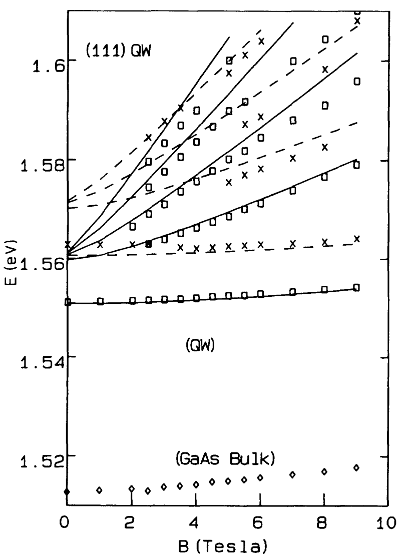

FIG. 2. The plot of the reflectance peak energies vs the magnetic field B for the (111)-grown GaAs QW sample No. 1.

(b), were used.

(a) *Fitting the diamagnetic shifts of ground and excited states with theoretical calculations.* Yang and Sham[11] have pointed out that many earlier studies that have used Landau levels to identify the excited states of the magnetoexciton were incorrect. Our numerical calculation is based on the effective-mass theory of Duggan,[9] which simultaneously accounts for the effects of quantum-well confinement, magnetic field, and the Coulomb potential of the exciton. The details of the calculation and assumptions made for the theory are given in Refs. 9 and 10. Only the reduced effective mass, μ, is used as the free-fitting parameter, all other parameters[9] are fixed. The calculated results for the heavy-hole excitons are shown by solid lines in Fig. 2 for sample No. 1. We can see that the theoretical results are in very good agreement with the experimental data for 1s and 2s states. For higher excited states, due to the interaction with light-hole excitonic states and probably with some forbidden transitions[12] also, the high-field data do not fit the theoretical curve very well. Therefore, we concentrated on curve fitting for the lower states. For comparison, we also fit the (001)-grown sample (No. 2). We obtained from these fittings the binding energies $E_B^{e\text{-hh}}(111) = 10.4 \pm 0.3$ meV and $E_B^{e\text{-hh}}(001) = 11.1 \pm 0.3$ meV, and the in-plane reduced masses $\mu_\perp^{e\text{-hh}}(111) = (0.053 \pm 0.002)m_0$ and $\mu_\perp^{e\text{-hh}}(001) = (0.058 \pm 0.002)m_0$. (These error bars were determined graphically by setting conservative upper and lower bounds.) Since the dependence of the exciton binding energy on the QW well width is small,[15] near 100 Å, the fluctuation of the well widths has little effect on the binding energy; therefore, the difference in binding energies between samples No. 1 and No. 2 is attributed to the different orientations of the samples.

We have also attempted to fit the light-hole exciton using the same theory. The results are shown by the dashed lines in Figs. 2 and 3 giving the binding energies $E_B^{e\text{-lh}}(111) = 11.4 \pm 0.8$ meV, $E_B^{e\text{-lh}}(001) = 11.1 \pm 0.8$ meV, and reduced masses $\mu_\perp^{e\text{-lh}}(111) = (0.060 \pm 0.005)m_0$, $\mu_\perp^{e\text{-lh}}(001) = (0.058 \pm 0.005)m_0$, respectively. Large errors are expected for the light-hole results, due to the mixing with the heavy-hole excited states.

(b) *Comparing the diamagnetic shift of the (111)-grown MQW with that of (001)-grown MQW.* For low magnetic field, the energy shift for a 1s magnetoexciton state can be written[8] as

$$\Delta E_{1s} = \frac{D\epsilon^2\hbar^4}{4c^2e^2\mu^3} H^2 \text{ (cgs units)} . \quad (1)$$

Here ϵ is the dielectric constant, and D is a parameter depending on the dimensionality of the system. The energy shifts parabolically with magnetic field H, and is inversely proportional to the cube of the reduced effective mass μ. Using $\Delta E_{1s} = aB^2$, we have plotted in Fig. 4 ΔE_{1s} against B^2 from 0 to 6 T for both (111) and (001)-grown sam-

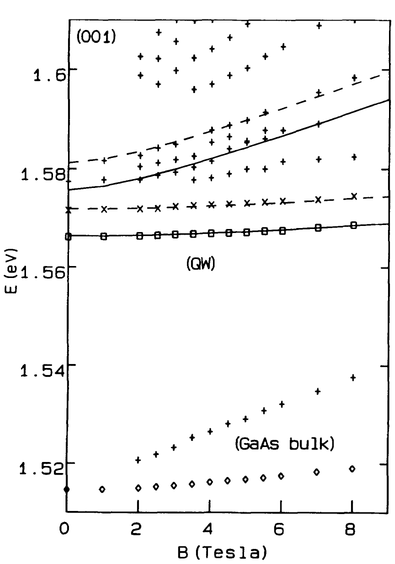

FIG. 3. The plot of the reflectance peak energies vs the magnetic field B for the (001)-grown GaAs QW sample No. 2.

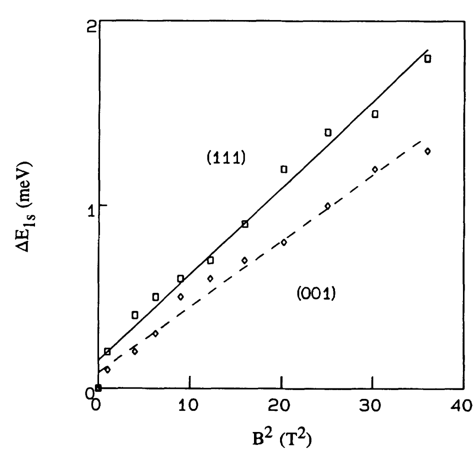

FIG. 4. The diamagnetic shifts, ΔE_{1s}, of the ground-state heavy-hole exciton vs magnetic-field square, B^2. □, sample No. 1; △, sample No. 2. The lines are the corresponding least-squares fits.

TABLE I. Exciton binding energies, E_B, and in-plane reduced masses, $\mu_\perp$, of GaAs/$Al_xGa_{1-x}As$ quantum wells with well width $d_w \sim 100$ Å.

Axis	Transition	d_w (Å)	B field (T)	E_B (meV)	$\mu_\perp$ ($\times m_0$)	Reference
[001]	hh1-e1	110	0–16	···	0.062	8
[001]	hh1-e1	112	1	8.5	0.0455	6
[001]	hh1-e1	112	1.2–1.8	8.5	0.048	6
[001]	hh1-e1	100	3–23	13±1	0.077	7
[001]	hh1-e1	104	0–8	12	0.0586	10
[001]	hh1-e1	100	0–9	11.2	0.058[a]	This work
[111]	hh1-e1	100	0–9	10.56	0.053[a]	This work
[111]	hh1-e1	100	0–9	···	0.054[b]	This work
[001]	lh1-e1	100	0–9	11.2	0.58[a]	This work
[111]	lh1-e1	100	0–9	11.4	0.60[a]	This work

[a]From method (a). [b]From method (b).

ples, and fit them with a straight line using a least-squares procedure. Factor a is the slope. We obtained $a_{111} = 47 \pm 3$ μeV/T^2 and $a_{001} = 36 \pm 3$ μeV/T^2. Since

$$\frac{a_{001}}{a_{111}} = \left[\frac{\mu_{111\perp}}{\mu_{001\perp}} \right]^3 , \tag{2}$$

if the in-plane reduced mass $\mu_{001\perp}$ is well known, we can then obtain $\mu_{111_\perp}$. We use the average value of several reported experimental results of $\mu_{001\perp}$ from independent sources as shown in Table I. Using Eq. (2) we estimate the in-plane reduced mass $\mu_{111\perp} \simeq 0.054 m_0$.

To compare $\mu_{111\perp}$ with $\mu_{001\perp}$ we calculate their ratio for both methods (a) and (b):

$$\frac{\mu_{111\perp}^{e\text{-hh}}}{\mu_{001\perp}^{e\text{-hh}}} = 0.91 \pm 0.05 \ \ (a) ,$$
$$\frac{\mu_{111\perp}^{e\text{-hh}}}{\mu_{001\perp}^{e\text{-hh}}} = 0.91 \pm 0.05 \ \ (b) . \tag{3}$$

This result means that the in-plane (111)-hh mass is slightly lighter than the (001)-hh mass.

The in-plane electron effective mass is isotropic near the bottom of the conduction band. Taking into account the nonparapolicity of the confined electron by using the Kane $\mathbf{k}\cdot\mathbf{p}$ theory,[16–18] we can determine the electron effective mass at the confined level for both (111) and (001)-growth samples in an approximate manner. Using $m_{be}^* = 0.0665$ as the band-edge electron effective mass, we calculated numerically the confinement energy for a well width of 100 Å and a band-offset ratio of 65:35.[19] We obtain the electron effective mass at the first confined level of $m_e^* \simeq 0.069 m_0$.

From the reduced masses found by methods (a) and (b), we can determine the hole effective masses in the plane perpendicular to [111] and [001]. The results are listed in Table II.

The valence-band structure of GaAs is complicated by the interaction between heavy and light holes, and their dispersion relations do not have a simple analytic form. However, under high uniaxial stress, or in a narrow quantum well, the heavy-hole and light-hole bands split. For the case of completely decoupled heavy and light holes, Hensel and Suzuki[20] have derived a simple relationship

TABLE II. Band parameters and heavy-hole and light-hole effective masses ($\perp$, in-plane; $\parallel$, parallel to the axis) in GaAs.

γ_1	γ_2	γ_3	T (K)	Axis	$m_\perp^{hh}/m_0$	$m_\parallel^{hh}/m_0$	$m_\perp^{lh}/m_0$	$m_\parallel^{lh}/m_0$	Reference
6.85	2.10	2.90	5	[001]	0.112[a]	0.377[a]	0.21[a]	0.09[a]	1
				[111]	0.102[a]	0.952[a]	0.253[a]	0.079[a]	1
6.95	2.25	2.86	77	[001]	0.45		0.082[b]		4b,4a
6.8	2.4	1.0	50	[001]	0.475		0.087[b]		3
7.2	2.5	1.1	77	[001]	0.45		0.082[b]		2
				[111]	0.45				2
6.8[a]	1.9[a]		4.2	[001]	0.11[a]	0.34	0.21[a]	0.094	12
			4.2	[001]	0.36±0.13		0.36±0.3		This work
3.3[c]±2.3		1.1[c]±1	4.2	[111]	0.23±0.07	0.83[c]	0.46±0.3	0.19[c]	This work

[a]These values were not provided by the author of the listed reference but they were calculated by using the experimental data provided in the reference and using Eqs. (4) or (5).

[b]The authors of the reference determined these mass values by performing measurement at high B field, approximating the excited state fan as a Landau fan to obtain the mass, then using a $\mathbf{k}\cdot\mathbf{p}$ theory extrapolated to $k = 0$ where they derived the listed values.

[c]The values were determined by using Eq. (5) and our experimental values for in-plane masses. The error bars given are experimental only. Several theoretical approximations and simplifications involving $\mathbf{k}\cdot\mathbf{p}$ theory, a Luttinger-Kohn effective mass description of the valence band, and its results of Ref. 20 make it difficult to determine a theory error bar. Error bars on the parallel masses are greater than 100%; more accurate and direct measurements of parallel masses from a different type of experiment will be presented elsewhere.

between the heavy- and light-hole effective masses involving the well-known Luttinger-Kohn band parameters γ_1, γ_2, and γ_3 as shown below:

Axis	$M_J=\pm\frac{1}{2}$ (light hole)	$M_J=\pm\frac{3}{2}$ (heavy hole)	Equation No.
[001]	$m_0/m_\perp=\gamma_1-\gamma_2$ $m_0/m_\parallel=\gamma_1+2\gamma_2$	$m_0/m_\perp=\gamma_1+\gamma_2$ $m_0/m_\parallel=\gamma_1-2\gamma_2$	(4)
[111]	$m_0/m_\perp=\gamma_1-\gamma_3$ $m_0/m_\parallel=\gamma_1+2\gamma_3$	$m_0/m_\perp=\gamma_1+\gamma_3$ $m_0/m_\parallel=\gamma_1-2\gamma_3$	(5)

These equations are highly simplified so that they may not describe the real QW case exactly. However, they can give us some indications of the relationship between hh and lh masses, and between in-plane and parallel masses in an approximate manner. For example, Eqs. (4) and (5) yield a heavier in-plane light-hole mass than the in-plane heavy-hole mass[5,12,21] which agrees with our experimental results.

We have determined γ_1 and γ_3 for our sample. Using Eq. (5), we have

$$\frac{m_0}{\mu_{111\perp}^{e\text{-hh}}}-\frac{m_0}{\mu_{111\perp}^{e\text{-lh}}}=\frac{m_0}{m_{111\perp}^{e\text{-hh}}}-\frac{m_0}{m_{111\perp}^{e\text{-lh}}}=2\gamma_3\,, \quad (6)$$

and γ_1 can be obtained using our result for $m_{111\perp}$. Using Eq. (5) again, we have estimated the parallel masses $m_{111\parallel}^{\text{hh}}$, and $m_{111\parallel}^{\text{lh}}$ (as shown in Table II). For comparison, we have also listed in Table II, the values of γ_1, γ_2, γ_3, and the effective hole masses for GaAs bulk or QWs from other authors. In order to compare them with our results, we have calculated the heavy- and light-hole effective masses using Eqs. (4) and (5) for the γ_s from Ref. 1. Their values of $m_{111\perp}^{\text{hh}}$ and $m_{001\perp}^{\text{hh}}$ give $\mu_{111\perp}^{e\text{-hh}}/\,\mu_{001\perp}^{e\text{-hh}}=0.964$, which is in agreement with our results shown in Eq. (3). This indicates that $\gamma_3>\gamma_2$. For the parallel masses, we can see from Table II that $m_{111\parallel}^{\text{hh}}>m_{001\parallel}^{\text{hh}}$ for the results of Refs. 1 and 12 and our result. Such results support $\gamma_3>\gamma_2$ also. Notice that the parameters deduced from Eqs. (4) and (5) listed in Table II are subject to large errors due to the approximations and the simplifications; these values are given for comparison purposes only. However, using our (111)-parallel hh mass $0.83m_0$ listed in Table II, we calculate numerically[14] the energy difference between the first electron subband level and the first heavy-hole subband level, yielding 1.554 eV. Then, subtracting from this energy, with the exciton binding energy, 10.4 meV, obtained from our analysis of the magnetoexciton spectrum with method (a), we find a transition energies of 1.5434 eV, which is in reasonable agreement with our experimental PL result for the sample No. 1.

We would like to remark that depending on each experimental condition (stress, QW width, and magnetic field), the distance between the valence-band edge and where the hole is measured is different. Therefore the result for the hole effective masses can be different from experiment to experiment.

In summary, we have performed a magneto-optical experiment on a high-quality (111)-grown GaAs-$Al_xGa_{1-x}As$ quantum well. An in-plane reduced mass $\mu_{111\perp}^{e\text{-hh}}=(0.053\pm0.002)m_0$ and binding energy $E_B^{e\text{-hh}}=10.6\pm0.3$ meV were determined for the 1s heavy-hole exciton, and $\mu_{111\perp}^{e\text{-lh}}=(0.060\pm0.005)m_0$, $E_B^{e\text{-lh}}=11.4\pm0.8$ meV for the 1s light-hole exciton, in the first confined (hh, lh, and electron) subbands of the QW. By comparison with the (001)-grown QW, we obtained $\mu_{111\perp}/\mu_{001\perp}=0.91\pm0.05$. We have also deduced the (111) in-plane effective masses, $m_{111\perp}^{\text{hh}}=(0.23\pm0.07)m_0$ and $m_{111\perp}^{\text{lh}}=(0.46\pm0.3)m_0$.

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*Also at Geo-Centers Inc., Lake Hopatcong, NJ 07849.

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