Photocurrent spectroscopy of a (001)- and a (111)-oriented GaAs/Al₀ 33Ga_{0 67}As quantum-well structure

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Photocurrent (PC) spectroscopy was used to study exciton oscillator strengths in a (001)- and a (111)-oriented GaAs/Al_{0.33}Ga_{0.67}As quantum-well structure (QWS) as a function of the electric field. In the (111) quantum well, an extremely large oscillator strength of the e_1 -hh₂ forbiddentransition exciton was observed that was larger than that of the e_1 -lh₁ exciton, even at low electric fields. [The notation e_n -hh(lh)_m represents a transition between the *n*th electron and the *m*th heavy-(light-) hole subband.] The exciton oscillator strength ratios estimated from the PC spectra were compared with those calculated theoretically with the exciton reduced-mass ratio used as a parameter. The comparison revealed that, whereas the reduced masses of the e_1 -hh₁ and e_1 -hh₂ heavy-hol exciton are lighter than that of the e_1 -lh₁ light-hole exciton in the (001) QWS, reflecting the mass reversal effect in the (001) QWS, it is not so in the (111) QWS; especially, the reduced mass of the e_1 hh₂ exciton was revealed to be as much as 1.5 times larger than that of the e_1 -lh₁ exciton in the (111) QWS. This difference between the (001) and the (111)QWS is discussed, taking valence-band coupling into account; it is attributed to a difference in order between the lh_1 and the hh_2 subband, since a strong repulsion between the two subbands is expected to give either a large or a negative in-plane mass to the upper subband of the two.

I. INTRODUCTION

There has recently been great interest in the electronic and optical properties of quantum-well structures $(QWS's)$ in the presence of electric fields.¹⁻¹⁰ To date, most studies of $GaAs/Al_xGa_{1-x}As QWS's in electric$ fields have concentrated on structures with the growth axis oriented along the [001] crystal direction. $1-9$

On the other hand, several striking features have been found for QWS's with the growth axis oriented along the [111] direction. Hayakawa et al.¹¹ found that in a (111)oriented quantum-well laser structure, the photoluminescence (PL) intensity at room temperature is enhanced by more than 50 times and that the threshold current density is reduced compared with those in (001)-oriented laser structures. They explained these phenomena in terms of an enhancement of the heavy-hole excitonic transitions relative to the light-hole excitonic transitions in (111) oriented quantum wells.¹²

In our previous study¹⁰ the electric-field dependence of excitonic transition energies in a (111) QWS was studied by PL measurements. The energy shift (Stark shift) of the heavy-hole exciton induced by an electric field in the (111) QWS was found to be larger than that in a (001) QWS having the same well width. In addition, relatively strong emission due to the "forbidden" transition between the second heavy-hole $(hh₂)$ and the first electron (e_1) states was observed. This seems also to be related to an enhancement of the heavy-hole excitonic transitions in (111)-oriented quantum wells.

In the present work we study the photocurrent (PC) spectra of the (001) and the (111) QWS in the presence of electric fields; attention was focused especially on the exciton oscillator strengths. A comparison between the peak intensity in the PC spectra and the calculated oscillator strength suggests that the reduced mass of the heavy-hole exciton is heavier than that of the light-hole exciton in the (111) QWS. This relation of the exciton reduced masses which is opposite to that in the (001) QWS is explained by a theory in which valence-band coupling is taken into account.

II. EXPERIMENT

Two samples were grown simultaneously by molecular-beam epitaxy on n^+ -type GaAs substrates. One sample was grown on a (001) substrate and the other on a (111)B substrate. Details of the growth have been described previously.¹⁰ For the purpose of the application of electric fields perpendicular to the quantum wells, the samples had a $p-i-n$ structure with a superlattice embedded in the middle of the i layer. In detail, the structure consisted of 0.5 - μ m-thick Si-doped GaAs followed by 0.3- μ m-thick Si-doped Al_xGa_{1-x}As, 0.25- μ m-thick undoped $Al_xGa_{1-x}As$, a 20-period superlattice composed of alternating 10-nm GaAs wells and 15-nm $Al_xGa_{1-x}As$ barriers, 0.25- μ m-thick undoped $\text{Al}_x\text{Ga}_{1-x}$ As, 0.3- μ m-thick Be-doped $\text{Al}_x\text{Ga}_{1-x}$ As, and finally 5-nm-thick Be-doped GaAs for contact. The Al content x of $Al_xGa_{1-x}As$ was 0.33. Ohmic contacts were formed on the top layer and on the backside of the n^+ substrate.

The PC spectrum was taken with chopped light from a tungsten lamp dispersed by a 0.25-m grating monochromator, incident onto the sample surface perpendicularly. Thus, the polarization of the incident light was parallel to the quantum wells $(xy \text{ polarized})$. A lock-in amplifier was used for standard synchronous detection. Measurements were made at 77 K. The electric field in the quantum

wells was determined by the applied bias voltage plus the built-in voltage $(1.9 V)$ divided by the thickness of the i layer $(1.0 \,\mu\text{m})$.

III. RESULTS AND DISCUSSION

Figures $1(a)$ and $1(b)$ show the PC spectra for the (001) and the (111) QWS at various reverse biases. Exciton transitions are labeled with the notation e_n -hh(lh)_m, which represents the exciton associated with the nth electron and the mth heavy- (light-) hole subband at $k_{\parallel} = 0$. At $k_{\parallel} = 0$, the valence-band states are pure heavy- or light-hole states in both (001) and (111) quantum wells in the envelope-function approximation; 13 it is therefore possible to label the subbands as "heavy hole" or "light

FIG. 1. Photocurrent spectra of (a) (001) QWS and (b) (111) QWS at different applied voltages.

hole" according to their $k_{\parallel} = 0$ character. If not specified, the e_n -hh(lh)_m represents the 1s exciton throughout the text. In our previous study,¹⁰ two peaks of the e_1 -hh₁ and e_1 -lh₁ exciton in the (001) QWS and three peaks of the e_1 -hh₁, e_1 -hh₂, and e_1 -lh₁ exciton in the (111) QWS were observed in the PL spectra. The energies of these peaks in the PL spectra agree with those in the PC spectra in the present study within 1 meV at each bias. Moreover, in the present study, additional peaks in the higher energy region were observed in the PC spectra, as can be seen in Figs. $1(a)$ and $1(b)$.

Comparing Figs. 1(a) and 1(b), one can see remarkable differences between the (001) and the (111) QWS as follows: (1) The energy intervals between the heavy-hole excitons are smaller in the (111) QWS than in the (001) QWS. As a result, the e_1 -hh₂ exciton peak appears at lower energy than the e_1 -lh₁ exciton peak in the (111) QWS. (2) Though the e_1 -hh₂ transition is a forbidden transition at zero electric field, nevertheless, the exciton peak due to it grows so large with increasing electric field that it becomes larger than the e_1 -lh₁ exciton peak in the (111) QWS. This is consistent with the rather strong emission due to the e_1 -hh₂ exciton observed in our previous study.¹⁰ On the other hand, the e_1 -hh₂ exciton peak remains small in the (001) QWS.

In Secs. III A-III C we discuss the transition energies (in III A) and the oscillator strengths (in III B). In III C, the exciton reduced masses estimated from the PC spectra are compared with theories.

A. Transition energies

The experimental transition energies were compared with the theoretical values. For the calculation of the energy levels in the absence of an electric field, the bandgap discontinuity between the GaAs and $AI_xGa_{1-x}As$ was taken to be $\Delta E_g(x) = 1.425x - 0.90x^2 + 1.1x^3$ eV.¹ The depth of the confining potentials for electrons and holes, V_e and V_h , were determined to be $V_e = 0.63 \Delta E_g$ and $V_h = 0.37 \Delta E_g$.¹ Regarding boundary conditions, current-conserving conditions were adopted. The perpendicular effective masses of electrons, heavy holes, and light holes used in the calculation were

$$
m_e^* = (0.0665 + 0.083x)m_0,
$$

\n
$$
m_{hh}^1[001] = (0.34 + 0.41x)m_0,
$$

\n
$$
m_{hh}^1[111] = (0.9 + 0.41x)m_0,
$$

and

$$
m_{\rm 1h}^{\perp} = (0.117 + 0.033x) m_0 ,
$$

respectively.¹² (The value of m_{th}^{\perp} in GaAs wells is somewhat larger than the effective mass of light holes in bulk GaAs, but close to $m_{\text{lh}}^{\perp} = 0.12 m_0$ and 0.125 m_0 used by Yamanaka et al.⁵ and Andrews et al., ⁶ respectively, to fit the experimental energy levels in quantum wells. Andrews et al.⁶ attributed the large value of m_{lh}^{\perp} to the effect of nonparabolicity in the light-hole band.)

For the calculation of the energy shift (Stark shift) induced by an electric field, the "effective infinite-well mod-
el" developed by Miller et al.¹ was used. In this model

FIG. 2. Experimental and calculated energies of the excitonic transitions for (a) (001) QWS and (b) (111) QWS as a function of electric field.

actual quantum wells having finite barriers are approximated by ones having infinitely high barriers and "effective well widths." The effective well widths are chosen to give the correct energies of the actual quantum wells having finite barriers in the absence of an electric field. The Schrödinger equation for a particle in an infinitely deep well in an electric field is solved exactly and the resulting wave functions are Airy functions. For the bindigg energies of excitons, a constant energy (10 meV) was assumed for all Is excitons. The change in the exciton binding energy induced by the electric field is neglected, since it is smaller by an order of magnitude than the change in the band-to-band transition energy. The calculated results for 1s excitons are compared with the experimental data in Fig. 2. One can see good agreement between them. Thus, the 1s exciton energies and their Stark shifts are well explained by using the different perpendicular heavy-hole masses $(0.34m_0$ and $0.9m_0)$ for the (001) and the (111) wells, respectively.

Besides the above-mentioned 1s exciton peaks, an extra small peak appears in the higher-energy side of the e_1 -lh peak for both the (001) and the (111) QWS, as can be seen in Figs. 1(a) and 1(b). As for the additional peak in the in Figs. 1(a) and 1(b). As for the additional peak in the (001) QWS, several published spectra^{5,7,8,14} of (001) QWS's exhibited a similar peak in this energy range; it was attributed to the excited states (2s and 2p) of the e_1 lh_1 exciton. '⁴ As for peak a in Fig. 1(b), this is the first observation of such a peak in this energy range in (111) QWS's. This peak may also be ascribed to the excited states of the e_1 -lh₁ exciton, as in the case of the (001) QWS. However, there is another possibility that peak a may be ascribed to the excited states of the e_1 -hh₂ exciton because not only the 1s e_1 -lh₁ exciton peak but also the 1s e_1 -hh₂ exciton peak appears at lower energy positions than does peak a by several meV.

B. Oscillator strengths

We next discuss the oscillator strengths of excitonic transitions, f_{ex} , in an electric field. Since the integrated areas of the excitonic peaks in the PC spectra are proportional to f_{ex} , we can compare them with the theoretically calculated f_{ex} . The f_{ex} for 1s excitons is expressed $as^{2-4,15}$

$$
f_{\rm ex} \propto \mu^2 |M_{cv}^{h(l)}|^2 F(0) , \qquad (1)
$$

where μ is the in-plane reduced mass of the exciton, $M_{cv}^{h(l)}$ is the optical matrix element between the conduction and the heavy- (light-) hole subbands at $k_{\parallel} = 0$, and $F(0)$ is the electron-hole overlap function at $k_{\parallel}=0$. The electronhole overlap function can be calculated using the wave functions ψ obtained in the "effective infinite-well model" as

$$
F(0) = \left| \int dz \; \psi_e(z) \psi_h(z) \right|^2 \; . \tag{2}
$$

The optical matrix elements at the zone center can be estimated within the envelope-function approximation with the 4×4 Luttinger Hamiltonian in the $|J, M \rangle$ basis.¹³ By taking the quantization axis z of angular momenta along the growth direction, the off-diagonal terms in the 4×4 Luttinger Hamiltonian for both (001) and (111) quantum wells vanish at $k_{\parallel} = 0$. ¹³ Therefore, the hole subband states at $k_{\parallel} = 0$ in both (001) and (111) quantum wells are pure heavy- or light-hole states which are the eigenstates of the z-component of angular momenta M with $M = \pm \frac{3}{2}$ or $\pm \frac{1}{2}$, respectively. Hence, the ratio of the matrix elements at $k = 0$ is

$$
|\mathbf{M}_{cv}^l|^2/|\mathbf{M}_{cv}^h|^2 = \frac{1}{3} \tag{3}
$$

in both (001) and (111) quantum wells for light polarized parallel to the quantum wells $(xy \text{ polarized})$.¹³

Since all values in Eq. (1), except for μ , can be calculated as above, one can estimate the value of μ by comparing the experimental intensity of the exciton peaks in the PC spectra with the calculated value of f_{ex} using Eq. (1), with μ as a fitting parameter.

In Figs. 3(a) and 3(b), the relative oscillator strengths of a heavy-hole exciton to a light-hole exciton in the (001) QWS estimated from the experiments are compared with those calculated using Eq. (1) with a heavy-to-light exciton reduced-mass ratio, μ_{hh}/μ_{lh} , as a parameter. Figure 3(a) shows the relative oscillator strength of the e_1 -hh₁ exciton to the e_1 -lh₁ exciton as a function of the electric field; Fig. 3(b) shows that of the e_1 -hh₂ exciton to the e_1 lh_1 exciton. The calculated results are indicated by solid lines; the experimental results are indicated by closed circles with error bars. The error bars are due to the uncertainty in removing background and in peak deconvolution. From Fig. 3(a) the reduced mass of the e_1 -hh₁ exciton is estimated to be about 0.8 times that of the e_1 -lh₁ exciton. Masumoto et al.¹⁵ also found that the experimental oscillator strength ratio of the e_1 -hh₁ exciton to the e_1 -lh₁ exciton in (001) GaAs/AlAs QWS's in the absence of an electric field roughly agrees with the calculated one using $\mu_{hh}/\mu_{lh}=0.8$ for well widths narrower than 12 nm. These results concerning exciton oscillator strengths are consistent with experimental studies concerning exciton binding energies in (001) QWS's;¹⁶ they revealed that the exciton binding energy is larger for the

FIG. 3. Relative oscillator strengths of (a) the e_1 -hh₁ exciton and (b) the e_1 -hh₂ exciton to the e_1 -lh₁ exciton in the (001) QWS as a function of the electric field. The solid lines indicate the calculated results with μ_{hh}/μ_{lh} as a parameter. The experimental results are indicated by closed circles with error bars.

 e_1 -lh₁ exciton than for the e_1 -hh₁ exciton, since a heavier reduced mass causes a larger binding energy. These ex-'perimental results indicate the "mass-reversal effect;"^{17,1} even though the heavy hole is indeed heavy along the z axis, it is light in the well plane, and vice versa. Figure 3(b) suggests that the reduced mass of the e_1 -hh₂ exciton is even smaller, 0.5 times that of the e_1 -lh₁ exciton.

Figures 4(a) and 4(b) show the relative oscillator strengths of (a) the e_1 -hh₁ exciton and (b) the e_1 -hh₂ exciton to the lh_1 exciton in the (111) QWS. From Fig. 4(a), the reduced-mass ratio of the hh_1 exciton to the lh_1 exciton is estimated at 1.1. Moreover, Fig. 4(b) indicates that the reduced mass of the hh_2 exciton is 1.5 times larger than that of the lh_1 exciton. In this way, the "massreversal" picture does not represent the (111) QWS case.

C. Comparison of exciton reduced masses with theories

We now compare these estimated ratios of the exciton reduced mass with theories concerning the in-plane effective masses of holes, since the exciton reduced mass is given by $1/\mu = 1/m_e^* + 1/m_{\text{hh(lh)}}^*$, where $m_{\text{hh(lh)}}^*$ is the

FIG. 4. Relative oscillator strengths of (a) the e_1 -hh, exciton and (b) the e_1 -hh, exciton to the e_1 -lh₁ exciton in the (111) QWS as a function of electric field. The solid lines indicate the calculated results with μ_{hh}/μ_{lh} as a parameter. The experimental results are indicated by closed circles with error bars.

in-plane effective mass of a heavy (light) hole at the zone center.

We first consider the diagonal approximation of the Luttinger Hamiltonian as a zeroth-order approximation. Neglecting the off-diagonal terms (i.e., valence-band coupling), the Luttinger Hamiltonian gives following inplane masses: 17, 18

$$
m_{\text{hh}}^{\parallel} = m_0 / (\gamma_1 + \gamma_2), \quad m_{\text{lh}}^{\parallel} = m_0 / (\gamma_1 - \gamma_2) \quad (001) \text{ QWS} ,
$$

(4)

$$
m_{\text{hh}}^{\parallel} = m_0 / (\gamma_1 + \gamma_3), \quad m_{\text{lh}}^{\parallel} = m_0 / (\gamma_1 - \gamma_3) \quad (111) \text{ QWS} ,
$$

where γ_1 , γ_2 , and γ_3 are the Luttinger parameters. Since Eqs. (4) give the mass-reversal picture, the diagonal approximation which neglects valence-band coupling seemingly stands well for the (001) QWS and has been used to explain the mass reversal in the exciton reduced mass in explain the mass reversal in the exciton reduced mass in (001) QWS's.^{15,17,18} On the other hand, since the experi ment shows opposite results to mass reversal, it is clear that neglecting valence-band coupling causes in the (111) QWS serious error in the (111) QWS case.

The calculation of the in-plane hole mass which fully takes valence-band coupling (i.e., off-diagonal terms of Luttinger Hamiltonian) into account can be performed analytically if infinitely high barriers are assumed.¹⁹ This infinite-well model seems to be a good approximation for the hole subbands in issue, since the height of an $Al_{0.33}Ga_{0.67}As barrier for holes, ~150 meV, is$ sufficiently high compared with the energies of the hole subbands. A further simplification with little loss of accuracy is obtained by neglecting the difference between γ_2 and γ_3 in the off-diagonal terms of the Luttinger Hamiltonian.²⁰ As a result, the in-plane hole masses of the *n*th heavy- (light-) hole subband are given by²⁰

r

$$
\frac{1}{m \Big|_{\text{hn}}} = \frac{1}{m \Big|_{\text{nh}}} \left[1 + 3 \frac{(-1)^{n+1} + \cos(n \pi \theta^{-1})}{n \pi \theta^{-1} \sin(n \pi \theta^{-1})} \right],
$$
\n
$$
\frac{1}{m \Big|_{\text{hn}}} = \frac{1}{m \Big|_{\text{h}}} \left[1 + 3 \frac{(-1)^{n+1} + \cos(n \pi \theta)}{n \pi \theta \sin(n \pi \theta)} \right],
$$
\n(5)

where $\theta = (m \frac{1}{h} h/m \frac{1}{h})^{1/2}$. θ can be regarded as a parame ter which indicates the relative energy of the lh_1 subband to heavy-hole subbands; when $\theta = n$, the energy of the lh₁ subband coincides with that of the nth heavy-hole subband at $k_{\parallel} = 0$. Note that the masses are independent of the well width in this model. These analytical forms give a good perspective on the relation in hole masses. The calculated in-plane hole masses normalized by m^{\perp}_{lh} are plotted as a function of θ in Fig. 5. From Fig. 5 one can see that, whereas the in-plane mass of hh_1 is always positive, the in-plane mass of hh₂ becomes zero at $\theta=2$ and becomes negative (electronlike) beyond that point. As for the light hole, one can see as follows: The in-plane mass of lh, becomes heavier with an increase of θ , and the lh, subband becomes flat in the vicinity of $\theta = 1.7$. Beyond that point, the lh, subband bends to the opposite side, upward, and the curvature becomes sharper until the inplane mass becomes zero at $\theta=2$. After that, the inplane mass of lh_1 becomes positive again. Thus, while the in-plane mass of hh_1 exhibits a moderate change as a function of θ , the in-plane masses of both \ln_1 and \ln_2 change drastically, depending on the relative energy of lh_1 to hh₂. This is due to a strong coupling between the lh₁ and the hh₂ subband, except at $k_{\parallel}=0$. Especially, when $\theta \gtrsim 1.7$, the strong repulsion between the two subbands gives a negative mass to the upper subband of the two.

In the (001) QWS, since $m_{hh}^{1}[001] = 0.34m_0$ and m_{lh}^{\perp} = 0.117 m_0 result in θ = 1.7, Fig. 5 indicates that the lh_1 subband is almost dispersionless. Therefore, the reduced mass of the lh_1 exciton is almost equal to the electron effective mass. On the other hand, since the hh_1 and the $hh₂$ subband have positive masses, the reduced masses are lighter than the electron effective mass: $\mu_{\text{lh}}(\sim m_e^*) > \mu_{\text{hh}} > \mu_{\text{hh}}$. Thus, in the (001) QWS, the mass-reversal effect in exciton reduced mass is also derived from this model as well as the diagonal approximation. In the case of the (111) QWS, since $m_{hh}^{\perp}[111] = 0.9m_0$ and $m_{lh}^{\perp} = 0.117m_0$ result in $\theta = 2.8$, Fig. 5 indicates that $m_{\text{hh}}^{\parallel} > m_{\text{hh}}^{\parallel} > 0 > m_{\text{hh}}^{\parallel}$, which re-

FIG. 5. Normalized values of in-plane hole masses $m \parallel$ by $m \parallel$ as a function of θ . θ is defined as $(m_{hh}^{\perp}/m_{lh}^{\perp})^{1/2}$.

sults in $\mu_{hh2} > m_e^* > \mu_{hh1} > \mu_{lh1}$. Namely, mass reversal does not occur. In this way, the infinite-well model which takes valence-band coupling into account offers the correct order of magnitude of the exciton reduced mass in both the (001) and the $(111) QWS$ —that is to say, in this model, since the upper of the two subbands (the lh_1 and the hh_2 subband) has a huge, or even a negative mass, due to repulsion from the lower subband, the e_1 hh₂ exciton has a large reduced mass in the (111) QWS in which the hh_2 subband is the upper, while the e_1 -lh₁ exciton has a large reduced mass in the (001) QWS in which the lh_1 subband is the upper.

IV. CONCLUSION

In conclusion, we have studied the PC spectra of a (001) and a (111) QWS in electric fields. Between the (001) and the (111) QWS, in spite of the same well width, differences were observed in the exciton peak energies, their Stark shifts, as well as the exciton oscillator strengths. The observed differences in the exciton peak energies and the Stark shifts could be well explained in terms of the difference in the hole mass normal to the well plane. An extremely large oscillator strength of the e_1 -hh₂ exciton such that it is larger than that of the e_1 -lh₁ exciton, even at low electric fields, was observed in the (111) QWS; the oscillator strength of the e_1 -hh₂ exciton, however, remained small in the (001) QWS. These results were interpreted as representing the difference in exciton reduced mass: $\mu_{\text{lh1}} > \mu_{\text{hh1}} > \mu_{\text{hh2}}$ in the (001) QWS and μ_{lh} $< \mu_{\text{hh}}$ $< \mu_{\text{hh}}$ in the (111) QWS. These relations regarding exciton reduced masses were consistent with the in-plane hole masses theoretically predicted when taking the valence-band coupling into account.

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