Observation of the 2s state excitons in (111)-oriented GaAs/$Al_xGa_{1-x}As$ quantum-well structures

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Transitions associated with the 2s excited-state heavy-hole excitons have been observed in the low-temperature photoluminescence excitation spectra of (111)-oriented GaAs/$Al_{0.3}Ga_{0.7}As$ multiple quantum wells. The binding energy of the 1s heavy-hole excitons has been found to be about 10% larger in (111)-oriented quantum wells than that in (100)-oriented ones.

The binding energy of the quasi-two-dimensional excitons in quantum-well structures (QWS's) has been investigated theoretically[1-6] and experimentally[1,6-11] for many years. In particular, the observation of 2s excited-state excitonic transition peaks in low-temperature photoluminescence excitation (PLE) spectra[6,9] and photoluminescence (PL) spectra[8] makes it possible to determine the binding energy of the 1s ground-state excitons very accurately. In this Brief Report, we present the observation of a 2s excited-state transition peak in low-temperature PLE spectra of (111)-oriented GaAs/$Al_{0.3}Ga_{0.7}As$ QWS's. Comparison of these spectra with that of (100)-oriented QWS and previously reported results of PLE and PL spectra of (100)-oriented QWS's reveals that the binding energy of $n=1$ electron heavy-hole 1s excitons is slightly ($\sim 10\%$) larger for (111)-oriented QWS's than that for (100)-oriented ones.

GaAs/$Al_{0.3}Ga_{0.7}As$ multiple-quantum-well (MQW) samples in this study were grown by molecular-beam epitaxy on n-type GaAs substrates (the number of Si atoms is approximately 2×10^{18} cm^{-3}) with orientations of (100) and (111)B (As face) misoriented by 0.5° toward (100). The substrate temperature and the group-V to group-III flux ratio during the growth were 720°C and ~2–3, respectively. The substrate holder was continuously rotated by 5 rpm during the growth. Details for the crystal growth have been reported elsewhere.[12-14] MQW samples consisted of 40 periods of GaAs wells (well width $L_z=47$ or 100 Å) and 200-Å thick $Al_{0.3}Ga_{0.7}As$ barriers, which were clad by 1.4-μm-thick $Al_{0.3}Ga_{0.7}As$ layers. All the epitaxial layers were undoped. The epitaxial growth was simultaneously done on both (100)- and (111)-oriented substrates mounted on the same molybdenum block side by side. L_z was determined from the ground-state light-hole excitonic transition energy (E_{11l}) in PLE spectra by the calculation.[15] Since the light-hole band in GaAs is assumed to be isotropic, the E_{11l} is same for both (100)- and (111)-oriented QWS's when L_z is same (see Fig. 1). L_z determined only by using the growth rate of GaAs slightly (within ±5%) differs from L_z determined by the PLE spectrum since the actual growth rate of GaAs changes due to the distribution of Ga flux and the desorption of Ga, which varies according to the spatial distribution of the substrate temperature. PLE spectra were measured at 4.2 K. The light from a 150-W broadband Halogen lamp was dispersed by a monochlomator and was vertically applied onto a sample for excitation.

PLE spectra of (111)- and (100)-oriented MQW's with $L_z=47$ Å are shown in Fig. 1. Transition energies calculated by using the finite square-potential-well model are

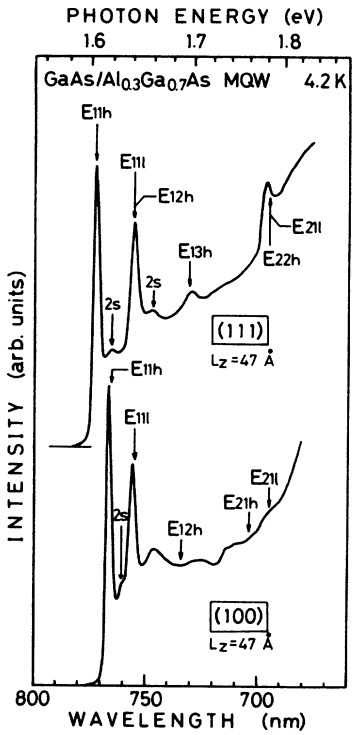

FIG. 1. Photoluminescence excitation spectra measured at 4.2 K on (111)- and (100)-oriented GaAs/$Al_{0.3}Ga_{0.7}As$ multiple quantum wells with the well width of 47 Å. 2s excited-state excitonic transitions are labeled by "2s." It is noted for a (111)-oriented quantum well that the energy of the E_{11l} and E_{22h} transitions almost coincides with that of the E_{12h} and E_{21l} transitions, respectively, since the $n'=2$ heavy-hole subband almost degenerates with the $n'=1$ light-hole subband.

indicated by arrows. Excitonic transitions are labeled by $E_{nn'm}$ where n and n' denote the electron and hole quantum numbers, respectively, and m signifies whether the excitonic transition involves a light or heavy hole, l or h, respectively. Effective masses for holes in GaAs derived from absorption spectra of MQW's at 300 K were used for calculation;[15] $m^*_{hh}[111]=0.9m_0$, $m^*_{hh}[100]=0.34m_0$,[16] and $m^*_{lh}[111]=m^*_{lh}[100]=0.117m_0$. The electron effective mass used was $m_e^*=(0.0665+0.0436E+0.236E^2-0.143E^3)m_0$, where E is in eV.[17] Discrete peaks are clearly observed on the high-energy side of E_{11h} and E_{11l} transition peaks in a (111)-oriented MQW. These peaks as well as a shoulder on the high-energy side of the E_{11h} peak in a (100)-oriented MQW are assigned to the 2s excited-state excitonic transitions.

The splitting energy ΔE_{1s-2s} between the 1s and 2s states of the heavy-hole excitons are plotted as a function of L_z in Fig. 2. For MQW's with $L_z=100$ Å in this study, the 2s excited-state transitions of the heavy-hole excitons was noticed only in a (111)-oriented sample as a shoulder, which is plotted in Fig. 2. Previously reported ΔE_{1s-2s} measured for (100)-oriented GaAs/Al_xGa_{1-x}As MQW's with $x=0.3$–0.4 by PLE (Ref. 1, 6, and 9) and PL (Ref. 8) are also plotted in Fig. 2. The value of ΔE_{1s-2s} is directly read from Fig. 1 in Ref. 1 for $L_z=42$ Å in the way that the position of the 2s transition is at the edge of the observed shoulder, whereas Miller *et al.*[1] took the 2s transition at the midpoint of the rising portion of the shoulder; this difference has been pointed out by Dawson *et al.*[6] As seen in Fig. 2, all the data of ΔE_{1s-2s} independently measured for (100)-oriented MQW's by PLE spectroscopy lie on a single curve within the deviation of ±0.2 meV. This demonstrates that PLE spectroscopy is a very reliable and reproducible method to determine ΔE_{1s-2s}. ΔE_{1s-2s} measured by PL spectroscopy is slightly larger than that measured by PLE spectroscopy. However, the difference in ΔE_{1s-2s} between these two spectroscopies is not greater than 0.4 meV.

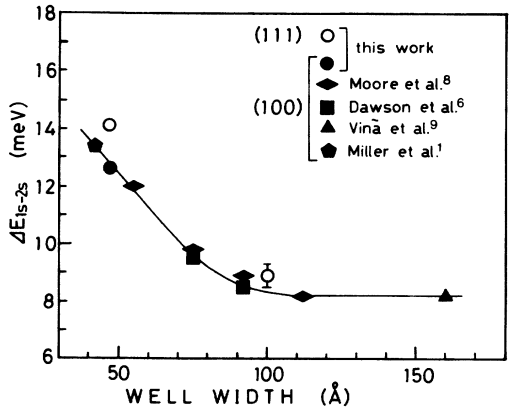

FIG. 2. Energy difference ΔE_{1s-2s} of the 2s excited-state excitonic transition peak from the 1s ground-state transition for heavy-hole excitons as a function of well width.

Therefore, the error in ΔE_{1s-2s} measured by these two methods on high-quality samples is less than 0.5 meV. As seen in Fig. 2, it is clear that the binding energy of 1s excitons in (111)-oriented QWS's is larger than that in (100)-oriented ones. The 1s-exciton binding energy is given as a summation of the experimentally measured ΔE_{1s-2s} and the theoretically calculated 2s exciton binding energy. For GaAs/Al_xGa_{1-x}As QWS's with $x=0.3$–0.4 and $L_z=\sim 40$–100 Å presented in Fig. 2, the binding energy of 2s excitons is in the range of 1.5–2.0 meV, which is about one order of magnitude smaller than the binding energy of 1s excitons.[3,6] The error in the calculated 2s exciton binding energy is not greater than 0.2 meV.[6] Thus the total error in the binding energy of 1s excitons determined in this manner is expected to be less than 0.5 meV. The binding energy of 1s excitons is determined to be 16.1 and 14.4 meV for (111)- and (100)-oriented MQW's with $L_z=47$ Å by using the binding energy of 2s excitons of 2.0 and 1.8 meV for (111) and (100) orientations; these values are calculated as reported in Ref. 6. The binding energy of 1s excitons in the (111)-oriented QWS is about 10% larger than that in the (100)-oriented one. When this difference in the binding energy is assumed to result from the difference in the heavy-hole subbands, the in-plane heavy-hole mass in the (111)-oriented QWS is about 70% larger than that in the (100)-oriented QWS.

It should be noted in Fig. 1 that the enhancement of the E_{11h} transition relative to the E_{11l} transition reported in PLE spectra for $L_z\sim 95$ Å (Ref. 18) is not observed for $L_z=47$ Å. We have observed that the PL efficiency and the threshold current density of (111)-oriented QWS's and QWS lasers with $L_z=40$ and 50 Å are considerably improved as compared with (100)-oriented counterparts.[13,19,20] Therefore, the E_{11h} transition is also considered to be enhanced in a (111)-oriented MQW, shown in Fig. 1. For $L_z=47$ Å, the $n'=2$ heavy-hole subband almost degenerates with the $n'=1$ light-hole subband at $k=0$, which possibly results in the enhancement of E_{11l} transition in a (111)-oriented MQW with $L_z=47$ Å. Here we do not discuss the binding energy of 1s light-hole excitons since the very strong valence-band mixing occurs between the $n'=2$ heavy-hole and the $n'=1$ light-hole subbands.

In summary, we have for the first time observed transitions of the 2s excited-state heavy-hole excitons in the low-temperature PLE spectra of (111)-oriented GaAs/$Al_{0.3}Ga_{0.7}$As MQW's. The binding energy of 1s heavy-hole excitons derived from PLE spectra is about 10% larger in (111)-oriented QWS's than that in (100)-oriented ones. This indicates that the in-plane heavy-hole mass for the ground state is about 70% larger in (111)-oriented QWS's than that in (100)-oriented ones.

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