

## Photoluminescence determination of well depth of $\text{Ga}_{0.47}\text{In}_{0.53}\text{As}/\text{Al}_{0.48}\text{In}_{0.52}\text{As}$ in an ultrathin single quantum well

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The photoluminescence spectra were measured from 4 to 90 K for single-quantum-well structures of  $(\text{Ga},\text{In})\text{As}/(\text{Al},\text{In})\text{As}$  with well widths  $L_z = 14.5$  and  $19.3$  Å. The optical transitions were observed from  $n = 1$  electrons in the conduction-band well to  $n = 1$  heavy holes (hh) and light holes (lh) in the valence-band well. Using the observed energy separation between hh and lh subbands, the well depth  $\Delta E_v = 120$  meV for holes in the valence band and the corresponding well depth  $\Delta E_c = 740$  meV for electrons in the conduction band were determined. This gives the relation of  $\Delta E_c = 0.85\Delta E_g$  which agrees with Dingle's rule for  $\text{GaAs}/(\text{Al},\text{Ga})\text{As}$  system.

### INTRODUCTION

It is crucial for device design and applications to determine the well depths of a single-quantum well (SQW) in the conduction band (CB) and valence band (VB). Band discontinuities of semiconductors hetero-interface have been determined by photoemission spectroscopy, absorption spectroscopy, and capacitance-voltage measurements. In the latter two methods, there are many adjustable parameters in determining the conduction-band well energy  $\Delta E_c$  and the valence-band well energy  $\Delta E_v$ . In photoemission, the value of  $\Delta E_v$  is directly obtained, however, the energy resolution is poor.<sup>1</sup> Optical methods offer convenient ways to measure these discontinuities. Dingle and his co-workers<sup>2</sup> have determined a value of  $0.15\Delta E_g$  for well depth in the VB from absorption measurements on  $(\text{Al},\text{Ga})\text{As}/\text{GaAs}$  quantum wells where  $\Delta E_g$  is the difference in the band gaps at the interface. This method in turn gave a value of  $0.85\Delta E_g$  for the well depth in the CB. This value has been accepted for years and has become known as "Dingle rule." Recently, this relationship has become a subject of controversy. The reported values differ using various methods. Even for the most widely studied interface of  $(\text{Al},\text{Ga})\text{As}/\text{GaAs}$  consistent and accurate values have not been determined. Some researchers determined values of  $\Delta E_c$  in range of  $0.60\Delta E_c$  to  $0.66\Delta E_g$  to explain their data.<sup>3,4</sup> Arnold *et al.*<sup>5</sup> deduced  $\Delta E_c = 0.65\Delta E_g$  for the  $(\text{Al},\text{Ga})\text{As}/\text{GaAs}$  system using a current-voltage technique.

This paper reports on the first photoluminescence (PL) spectroscopy determination of well depths  $\Delta E_c = 740$  and  $\Delta E_v = 120$  meV in the CB and VB for an  $(\text{Al},\text{In})\text{As}/(\text{Ga},\text{In})\text{As}$  ultrathin single-quantum well. The measured optical transitions for ultrathin wells which are separated by a large energy difference  $\Delta E$  for the transitions from  $n = 1$  electron subband in the CB to both  $n = 1$  heavy hole (hh) and light hole (lh) subbands in the VB are

the key behind this determination. The relationship for  $\Delta E_c = 0.85\Delta E_g$  has been determined from these PL measurements. A theoretical calculation of  $\Delta E$  versus  $L_z$  is presented to support the measurements of  $\Delta E_v$  and  $\Delta E_c$ .

### EXPERIMENTAL METHOD

The samples were grown using a Varian 360 molecular-beam epitaxy (MBE) machine.<sup>6</sup> The structures were grown on a thermally cleaned InP substrate consisting of a 2500-Å-thick  $(\text{Al},\text{In})\text{As}$  layer followed by the  $(\text{Ga},\text{In})\text{As}$  single-quantum well of widths 14.5 and 19.3 Å, respectively, and a 100-Å cap layer of  $(\text{Al},\text{In})\text{As}$ . Each sample was attached to an aluminum plate by "super" glue and contained in a liquid Dewar. The sample was excited by a 488-nm argon-ion laser from 10 to 160 mW. The excitation area was about 200  $\mu\text{m}$  in diameter. The photoluminescence was collected by a double  $\frac{1}{2}$ -m Spex spectrometer, detected by Hamamatsu R632 S-1 photomultiplier, and analyzed by a lock-in amplifier and a recorder. The temperature of sample was measured by a silicon diode connected to the cold finger on which the sample-aluminum plate combination was placed.

### RESULTS

Various photoluminescence spectra were obtained as a function of lattice temperature  $T_L$  from 4 to 90 K for two single-quantum wells (SQW's) of well widths  $L_z = 14.5$  and 19.3 Å. A set of typical spectra for  $L_z = 14.5$  Å are shown in Fig. 1. The peaks are denoted by  $P_1$ ,  $P_2$ ,  $P_3$ , and  $P_4$ . The higher-energy set  $P_1$  and  $P_2$  are identified as arising from the radiative recombination of carriers in the barrier  $(\text{Al},\text{In})\text{As}$ . The lower-energy set  $P_3$  and  $P_4$  are identified as luminescence from the well  $(\text{Ga},\text{In})\text{As}$ , which are shifted above the bulk band gap  $E_{g,w} = 0.81$  eV, due to confinement of carriers in the quantum well.

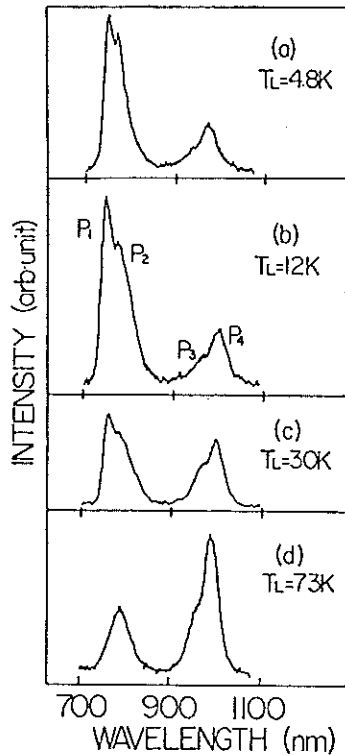


FIG. 1. Typical photoluminescence spectra of a (Al,In)As/(Ga,In)As SQW at well width  $L_z = 14.5 \text{ \AA}$  as a function of lattice temperature  $T_L$ . Excitation density power was about  $10^2 \text{ W/cm}^2$ . The peaks  $P_1$ ,  $P_2$ ,  $P_3$ , and  $P_4$  denoted in this figure are explained in the text.

In order to determine precisely the band gap of (Al,In)As, the origin of the emission peaks  $P_1$  and  $P_2$  must be discussed. From spectra displayed in Fig. 1, it is quite clear that  $P_1$  is stronger than  $P_2$  at lower  $T_L$  and disappears when  $T_L$  is above 80 K. This temperature corresponds to thermal energy  $k_B T_L = 7 \text{ meV}$ . The high-energy tail of  $P_1$  displayed in Fig. 1(a) is fitted to  $E^2 \exp(-E/k_B T_c)$  yielding a carrier temperature  $T_c = 90 \text{ K}$  ( $k_B T_c = 8 \text{ meV}$ ).  $T_c$  is higher than  $T_L$  due to the heating from photoexcited energetic carriers. The three-dimensional exciton's binding energy in (Al,In)As is about  $9 \text{ meV}$  using the parameters  $m_e = 0.118 m_0$ ,  $m_{hh} = 0.677 m_0$ ,  $K(\text{AlAs}) = 10.1$ , and  $K(\text{InAs}) = 14.6$ . The disappearance of  $P_1$  is caused by the lattice vibrational energy ionizing the exciton in the barrier. Thus,  $P_1$  emission most likely arises from the annihilation of acceptor-bound exciton. The  $P_2$  emission is broader than the  $P_1$  and is weak at  $T_L = 90 \text{ K}$ . This peak probably arises from the electron transitions from conduction-band edge to the shallow acceptor states. Furthermore, the intensity dependence of luminescence on pump intensity of the high-energy set ( $P_1$  and  $P_2$ ) is plotted in Fig. 2. The luminescence intensity dependence varies as  $I_e^{1.2}$ . This confirms our speculation that the high-energy set is related to either impurity or exciton emission.<sup>7</sup> From the above identification of  $P_1$  and  $P_2$ , the band gap of barrier (Al,In)As is determined to be  $E_{gb} = E_{P_1} + E_B = 1.66 + 0.009 = 1.67 \text{ eV}$  at  $T_L = 4 \text{ K}$ , where  $E_{P_1}$  is

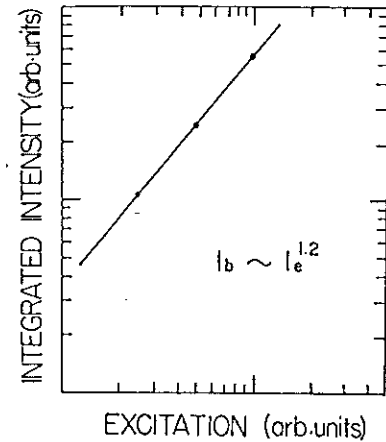


FIG. 2. Intensity dependence of emissions from barriers of (Al,In)As/(Ga,In)As SQW of well width  $L_z = 14.5 \text{ \AA}$  with respect to excitation intensity  $I_e$ .

the peak energy of  $P_1$  and  $E_B$  is binding energy of acceptor-bound exciton.

The luminescence spectra of  $P_3$  and  $P_4$  emission for  $L_z = 14.5$  and  $19.3 \text{ \AA}$  are shown in Fig. 3. These arise from transitions in quantum-well region for both well widths. The peaks  $P_3$  and  $P_4$  are separated by energy differences  $\Delta E = 47$  and  $57 \text{ meV}$  for  $L_z = 14.5$  and  $19.3 \text{ \AA}$ , respectively. The following argument is presented to support the assignment of the two peaks  $P_3$  and  $P_4$  arising from the radiative transitions from  $n=1$  electron states confined in the CB well to  $n=1$  hh and  $n=1$  lh states confined in the VB well. The excitation-power

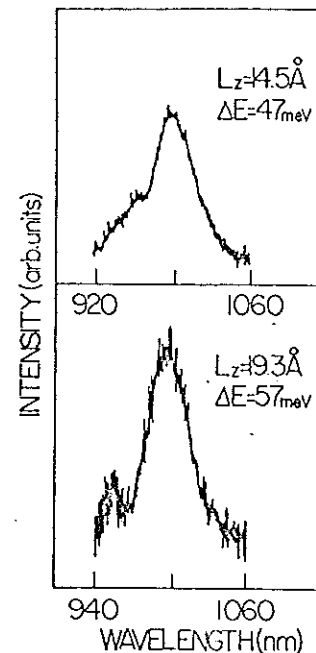


FIG. 3. SQW photoluminescence spectra from well GaInAs of two narrow wells. At lattice temperature  $T_L = 7 \text{ K}$ . The energy separations  $\Delta E = 47$  and  $57 \text{ meV}$  between hh and lh subbands for  $L_z = 14.5$  and  $19.3 \text{ \AA}$ , respectively, are indicated on each curve.

dependence of luminescence intensity of  $P_3$  and  $P_4$  emission from  $L_z = 14.5$ -Å quantum well is displayed in Fig. 4. The ratio of intensity of peaks  $P_3$  and  $P_4$  is defined by  $R_p = I_3/I_4$ , where  $I_3$  and  $I_4$  are the peak intensity for  $P_3$  and  $P_4$ , respectively. The electron temperature  $T_c$  from each spectrum is determined by fitting the high-energy tail of  $P_3$  to  $E^2 \exp(-E/k_B T_c)$ . The most salient feature of spectra shown in Fig. 4 is that the ratio  $R_p$  increases as electron temperature  $T_c$  increases. The  $R_p$  versus  $k_B T_c$  is plotted in Fig. 5. The cross dots are the peak intensity ratios measured from Fig. 4. The solid curve is fitted to  $R_p = C \exp(-\Delta E/k_B T_c)$ , where  $\Delta E$  is energy separation depicted in the inset of Fig. 5. A value of  $C = 5.2$  takes care of the overlap of transitions from electron subband to hh and lh subband tails. The value of  $C$  depends on the well width  $L_z$ , lh, and hh intersubband scattering and other factors. The value of  $\Delta E$  extracted from this fit is 47 meV. Thus, the energy  $\Delta E$  determined by this method is the same as the energy separation between lh and hh subbands in the quantum well  $L_z = 14.5$  Å measured from the photoluminescence spectra. This implies that holes in the VB are nondegenerate (Maxwell-Boltzmann). The populations of holes in the two subbands change with the electron temperature  $T_c$ . The inset of Fig. 5 shows this feature. This argument supports the model involving transitions from  $n = 1$  electron subband to two  $n = 1$  hole subbands.

In this section, the energy separation  $\Delta E$  between hh and lh subbands as a function of well width  $L_z$  are calculated to support the above data for the two thin wells as long as well depth  $\Delta E_v$  in the VB is set to be 120 meV. The following assumptions are used in the model: (1) the quantum well is simulated by an finite square well with width  $L_z$ ; (2) the well depth  $\Delta E_v$  is used as an adjustable parameter to be determined by fitting the observed energy separation  $\Delta E$  between hh and lh subbands at different well width  $L_z$ , and (3) the effective kinetic energy operator is obtained from three-dimensional effective-mass approximation, i.e., to take hh and lh effective mass

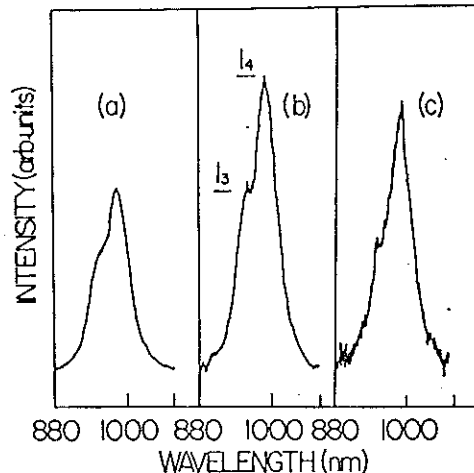


FIG. 4. SQW photoluminescence spectra from well region for  $L_z = 14.5$  Å with different excitation power  $P_e$ . The peak intensity are denoted as  $I_3$  and  $I_4$ , (a)  $P_e = 160$  mW, (b)  $P_e = 80$  mW, (c)  $P_e = 40$  mW.

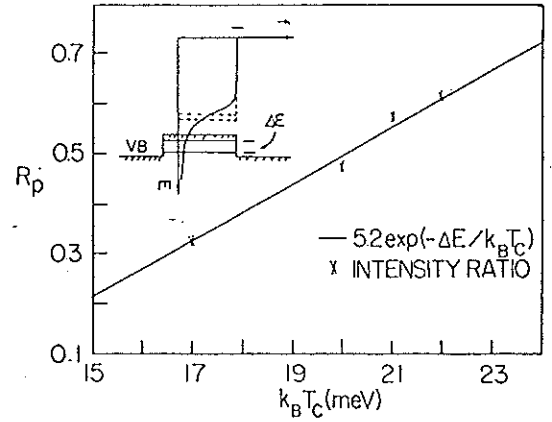


FIG. 5. Ratio of peak intensity of  $I_3$  and  $I_4$  with respect to the energy of carriers  $k_B T_c$ .  $T_c$  is the carrier temperature obtained by fitting each high-energy tail to  $E^2 \exp(-E/k_B T_c)$ . Inset shows that nondegenerate holes populate in hh and lh subbands in the VB of the SQW, where  $f$  is Fermi-Dirac function.

( $m_{hh}, m_{lh}$ ) from the band edge of (Al,In)As and (Ga,In)As compounds. This is a reasonable approximation because the quantum well for localizing holes in the valence band is shallow. Therefore, the energy dependence on the effective mass of holes is negligible. However, for an electron in the conduction band one must consider the energy dependence on the effective mass.

Due to the continuity of wave functions and their spatial derivatives at the boundaries, the eigenenergy value  $E_n$  in the valence band well is determined for a finite well by following equation:<sup>8</sup>

$$\tan p \sigma_n = \frac{2\sigma_n (m_w/m_b)^{1/2} (1 - \sigma_n)^{1/2}}{[(m_w/m_b + 1)\sigma_n^2 - 1]}, \quad (1)$$

where  $\sigma_n = E_n/\Delta E_v$  and  $p = L_z(2\Delta E_v m_w)^{1/2}/\hbar$  are two dimensionless variables;  $m_w$  and  $m_b$  are the effective mass in well and barrier, respectively, and  $n$  is the number of the localized states in the well. The value  $n$  is controlled by value of  $p$ . For  $L_z = 14.5$  and  $19.3$  Å, only one state ( $n = 1$ ) exists in both the CB and VB wells. The values of masses<sup>9</sup> used in this calculation are  $m_{whh} = 0.6094m_0$ ,  $m_{wlh} = 0.0491m_0$ ,  $m_{bhh} = 0.6768m_0$ , and  $m_{blh} = 0.0860m_0$ .

Various values of parameter  $\Delta E_v$  have been used in Eq. (1) to calculate  $\Delta E$  versus  $L_z$ . The calculated results  $\Delta E$  versus  $L_z$  for parameters  $\Delta E_v = 100, 120,$  and  $140$  meV are plotted in Fig. 6. The inset shows the schematic energy-band diagram of a SQW. The dots in Fig. 6 locate the position of the measured  $\Delta E$  for the two wells. The best fit to observed energy separations between hh and lh of 47 and 57 meV at well width  $L_z = 14.5$  and  $19.3$  Å is for the parameter  $\Delta E_v = 120$  meV. The fit of the theoretical calculation to experimental data for the well widths of  $L_z = 14.5$  and  $19.3$  Å demonstrates that well depth in the VB is  $\Delta E_v = 120$  meV. The corresponding well depth for the CB is  $\Delta E_c = 740$  meV which is  $0.86\Delta E_g$ . The results of this photoluminescence method agree well with Dirigle's relationship  $\Delta E_c = 0.85\Delta E_g$  for

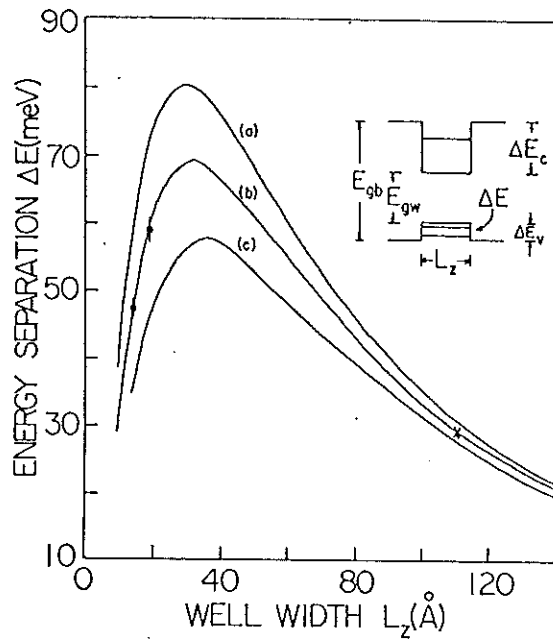


FIG. 6. Calculated energy separation  $\Delta E$  between hh and lh subbands with respect to well width  $L_z$  with  $\Delta E_v$  as a parameter. (a)  $\Delta E_v = 140$  meV, (b)  $\Delta E_v = 120$  meV, (c)  $\Delta E_v = 100$  meV. Inset shows the approximate band diagram of a SQW. The dots are our data for PL and X is the absorption data given in Ref. 13.

(Al,Ga)As/Ga(As) system obtained by absorption. However, it deviates from the result of  $0.73\Delta E_g$  at room temperature obtained by Morgan *et al.*<sup>10</sup> for (Al,In)As/(Ga,In)As who used the current-voltage method. The methods used to determine the well depths may account for this discrepancy. The current-voltage method may have systematic errors due to the background-carrier profile, concentration density of states, and the thermal energy of the free carriers.<sup>11</sup> The photoluminescence method requires the knowledge of hh and lh masses and interaction of carriers among the hh and the lh in subbands. However, the energy separation  $\Delta E$  between hh and lh subbands is rather insensitive to the precise values of masses. For an ultrathin quantum well, the energy separation  $\Delta E$  is large enough to be observed in photoluminescence spectra and the coupling between hh and lh could be neglected. Our results agree well with Harrison's linear-combination of atomic orbitals (LCAO) approach<sup>12</sup> and photoemission studies of Katnani and Margaritondo<sup>1</sup> within their limited accuracy ( $\pm 100$  meV). By using interpolation method, the absolute position of the valence-band maximum can be obtained from the table given in Ref. 12. This yields a value of  $\Delta E_v = 22$  meV. The large uncertainty in theoretical approach allows the agreement with our value of  $\Delta E_v = 120$  meV.

From the result of theoretical calculation shown in Fig. 6, three salient features appear. First, the value of  $\Delta E$  is extremely sensitive to the energy discontinuity  $\Delta E_v$  for an ultrathin SQW of thickness  $L_z = 14.5$  and  $19.3$  Å. This enables us to determine  $\Delta E_v$  as well as  $\Delta E_c$  very accurately ( $\pm 3$  meV). Second, for thick wells of  $L_z > 80$  Å, the value of  $\Delta E$  is insensitive to the choice of  $\Delta E_v$ . Therefore, it is difficult in practice to determine  $\Delta E_v$  for thick wells. A recent paper<sup>13</sup> reported the value of  $\Delta E_v = 290$  meV for (Ga,In)As/(Al,In)As system using the absorption spectra of a quantum well with  $L_z$  varying from 85 to 165 Å. The energies measured for the optical transitions for  $L_z = 110$  Å well given in Ref. 13 can also be fitted well to the absorption transition using the value of  $\Delta E_v = 120$  meV determined by us. The thickness of quantum wells studied by Weiner *et al.*<sup>13</sup> is in an insensitive range for determination of  $\Delta E_v$  by fitting the measured optical transitions (see Fig. 6). This will result in an inaccurate determination of  $\Delta E_v$ . Furthermore, they measured  $\Delta E_g = 29$  meV for  $n=1$  hh and lh subbands of  $L_z = 110$ -Å well which exactly agrees with our results shown in Fig. 6. This adds further support to our determination using a PL method. Third, there is a maximum energy separation  $\Delta E_m$  which occurs at well width  $L_{zm}$  for a given  $\Delta E_v$ . This feature may explain maximum quantum efficiency measured by Welch, *etc.*<sup>6</sup> Moreover, the value of  $\Delta E_m$  increases and  $L_{zm}$  decreases with increasing  $\Delta E_v$ .

In conclusion, the well depths  $\Delta E_c$  and  $\Delta E_v$  in the CB and VB for the SQW of (Al,In)As/(Ga,In)As have been determined to be 740 and 120 meV, respectively. This has been accomplished by fitting the transitions from  $n=1$  electron states to hh and lh states for both  $L_z = 14.5$  and  $19.3$  Å SQW's. It should be pointed out that the (Al,In)As/(Ga,In)As structure may possess new characteristics which are not displayed in the (Al,Ga)As/GaAs system, which arise from the much deeper well depth in the CB for the (Ga,In)As/(Al,In)As system by a factor of over 2 than the AlGaAs well. Some of these characteristics are the following: increase of the electron effective mass in both well and barrier region; the hot-carrier temperature dependence on the well width  $L_z$ ; the larger degeneracy and mobility of two-dimensional electron-hole system and the enhanced interaction strength of electrons and phonons due to the reduced dimensionality and more effective localization.

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