High-pressure studies of GaAs-$Ga_{1-x}Al_xAs$ quantum wells of widths 26 to 150 Å

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Photoluminescence spectra of GaAs quantum wells of widths 26 to 150 Å are studied as a function of hydrostatic pressure (0—70 kbar) at 80 and 150 K. The pressure coefficients of both the heavy- and light-hole excitons are found to decrease with decreasing well width. The direct to indirect conduction-band crossover, leading to the formation of type-II heterostructures, occurs at higher pressures for wider wells. A transition associated with the X conduction band in quantum-well structures is observed and its pressure dependence is established. Correlating this transition to barrier-to-well recombination determines the valence-band offset.

I. INTRODUCTION

We present a detailed study of the effect of hydrostatic pressure (0—70 kbar) on the energy levels of GaAs-$Ga_{1-x}Al_xAs$ quantum-well heterostructures at cryogenic temperatures. Several isolated GaAs quantum wells of different widths separated by wide $Ga_{1-x}Al_xAs$ barriers were grown on the same substrate by molecular beam epitaxy (MBE). Such samples enabled us to determine the variations in the pressure coefficients as a function of the well width. The pressure at which the Γ and X conduction subbands cross, leading to the formation of a type-II heterostructure, is also shown to depend on the well width. Correlating the observed indirect transition to recombination between the $Ga_{1-x}Al_xAs$ barrier and the GaAs heavy-hole confined transition, permits the determination of valence-band offsets. The physical origin of these effects is discussed in the light of available theoretical considerations.

It is well known that in the GaAs-$Ga_{1-x}Al_xAs$ multilayer structures, potential wells are formed in the GaAs layers, leading to quantized subbands whose energies are determined by the well width and depth. For spontaneous emission,[1] strongly allowed transitions occur between the quantized levels in the conduction band (CB) and the heavy (h) and light (l) hole subbands of the same quantum number n. These are labeled as E_{nh} (E_{nl}). We also introduce a superscript Γ or X to denote the CB from which they originate (e.g., E_{nh}^{Γ}). The experiments are performed at either 80 or 150 K at which the E_{1h}^{Γ} and E_{1l}^{Γ} are clearly observed. At lower temperatures thermal population effects make E_{1l}^{Γ} very weak. At room temperature, however, the thermal broadening smears out the E_{1l}^{Γ} transition. Most of the data presented in this paper are measured at 80 K. The 150-K pressure data are essentially similar, and are not shown in this paper.

We find that the pressure coefficients of the heavy- and light-hole excitonic transitions E_{1h}^{Γ} and E_{1l}^{Γ} decrease with decreasing well widths. The pressure at which Γ and X conduction subbands cross also decreases for narrower wells. The latter would affect the performance of devices such as quantum-well lasers[2] under pressure, particularly for narrow wells. By identifying the indirect transition observed as arising from recombination between an electron in the $Al_xGa_{1-x}As$ X conduction band and a hole in the GaAs valence subband, we determine the valence-band offset (to within binding energies) to be $Q_v = 0.30 \pm 0.04$.

II. EXPERIMENT

The heterostructures, grown by molecular beam epitaxy, consist of several single GaAs wells separated by wide $Ga_{1-x}Al_xAs$ barriers. Sample 1 consisted of four wells labeled $A-D$ with well widths of 26, 48, 70, and 96 Å, respectively, separated by $Ga_{0.67}Al_{0.33}As$ barriers of 750 Å width. Sample 2 consisted of five wells labeled $E-I$ with widths of 47, 70, 93, 117, and 140 Å, respectively. The aluminum mole fraction of the barrier was 0.3 and the width was 750 Å. Sample 3 consisted of 40 periods of 150-Å GaAs and 100-Å $Ga_{0.75}Al_{0.25}As$. The aluminum mole fractions of samples 1 and 2 were determined by photoluminescence and Auger spectroscopy and are accurate to $\pm 1\%$. The well widths are estimated from the growth parameters and are confirmed from a fit of the photoluminescence data to theory,[3] which determines the well widths to $\pm$ 5 Å. The GaAs substrate was thinned to about 30 μm and the sample placed in a Merrill Bassett[4] gasketed diamond-anvil cell. Argon was used as the pressure transmitting fluid. Fluorescence from the ruby R_1-R_2 lines was used to calibrate the pressure.[5] The cell was attached to the cold finger of a cryostat to obtain data at cryogenic temperatures. Photoluminescence was excited with 10 mW of 5145-Å radiation from an argon-ion laser. The pressure was hydrostatic to at least ± 0.5 kbar, as observed by the linewidths of the ruby fluorescence peaks.[6] From the linewidths of the more sensitive quantum-well emission lines in the direct gap phase, the pressure was seen to be uniform to better than ± 0.1 kbar.

III. RESULTS AND DISCUSSION

A. Pressure coefficients

The photoluminescence (PL) spectra for sample 1 at 80 and 150 K are shown in Fig. 1. Also shown in the figure is a schematic of the conduction band in the quantum wells, labeled A–D. The energy of the lowest level ($n=1$) is indicated by the dashed line inside the wells. In the spectra, the transitions from the $n=1$ level in the CB to the $n=1$ heavy- and light-hole levels in the valence band E_{1h}^{Γ} and E_{1l}^{Γ} are indicated by h and l, respectively, for the wells A–D. The wells are grown sequentially from A to D with the well D closest to the surface. The transition energies for the wells increase with decreasing well width. The h transition is more intense than the l at 80 K. At 150 K, l becomes slightly more intense due to additional thermal population of the hole states in the light-hole subband. The linewidth of the h peak for the 26-Å well is somewhat larger than the others due to its sensitivity to interface defects. The relative intensities of the peaks are not corrected for the absorption length of the exciting radiation or for the spectral response of the monochromator and detector.

Figure 2 shows PL spectra of sample 1 at different pressures. As pressure is applied, all the peaks move to higher energies and their relative intensities change dramatically at higher pressures. Figure 3 shows similar

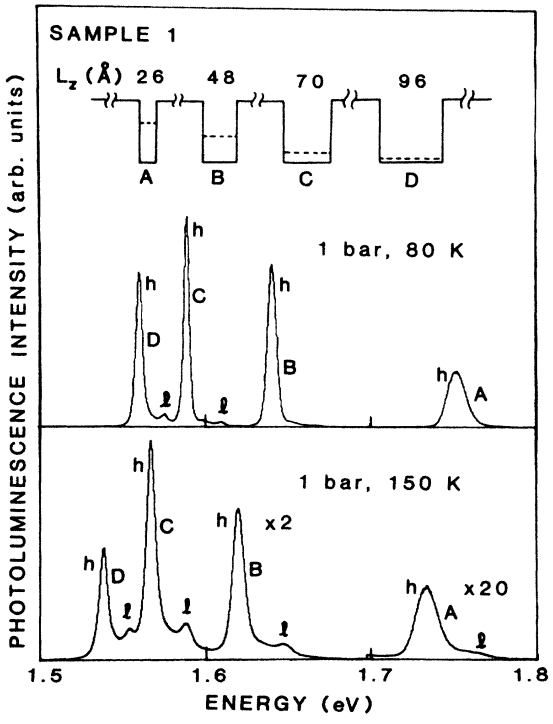


FIG. 1. Photoluminescence (PL) spectrum of sample 1 at 80 and 150 K at atmospheric pressure. The quantum wells A–D had well widths L_z of 26, 48, 70, and 96 Å separated by 750-Å-wide $Ga_{0.67}Al_{0.33}As$ barriers. The peaks labeled h and l for a given well are due to transitions from the $n=1$ electron subband to the heavy- (h) or light- (l) hole $n=1$ subband. The well D was closest to the sample surface.

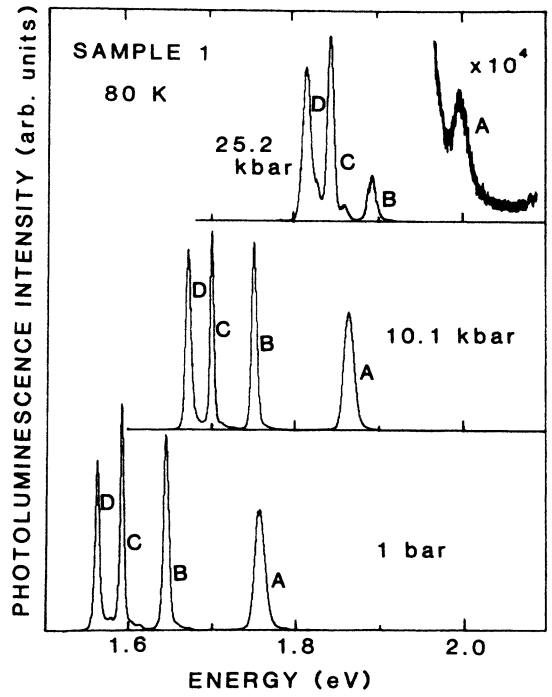


FIG. 2. PL of sample 1 at 80 K at 1 bar, 10.1 kbar, and 25.2 kbar. The peaks due to wells A–D are so indicated. Note the sharp decrease in the intensity of the peaks in wells A and B at 25.2 kbar.

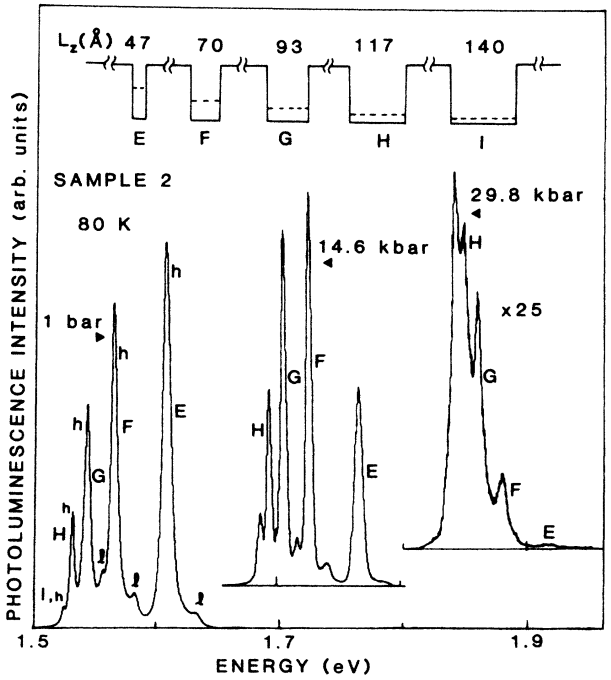


FIG. 3. PL of sample 2 at 80 K at 1 bar, 14.6 kbar, and 29.8 kbar. The quantum wells E–I have well widths L_z of 47, 70, 93, 117, and 140 Å with the well E closest to the sample surface. Note the drop in the intensity of the peaks in the narrow wells at high pressures.

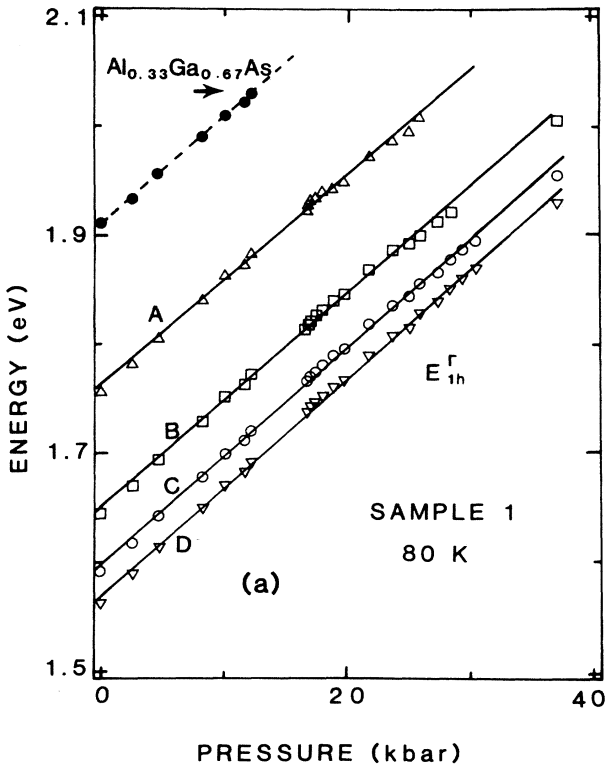
spectra for sample 2 which contains five single wells labeled E–I. In this sample, the well I was the first on top of the GaAs substrate and hence was farthest from the sample surface. The wells were grown in sequence, as shown at the top of Fig. 3. Hence the intensities of peaks farther from the surface of the sample are smaller. The symbols h and l are due to E_{1h}^{Γ} and E_{1l}^{Γ} transitions as in Figs. 1 and 2. With increasing pressure all the peaks move to higher energies. The relative intensities of the peaks in Fig. 3 change dramatically with pressure as in Fig. 2, qualitatively similar to that observed before in a 150-Å well.[7] It is well known[8–10] that the direct (Γ) CB of bulk GaAs increases with pressure and crosses the indirect (X) CB around 40 kbar. The X CB has a small negative pressure coefficient.[8–11] The decrease in intensity of the quantum-well transitions derived from the Γ CB is associated with the Γ-X crossover. Due to the band offsets between well and barrier, the lowest X CB is in the $Al_xGa_{1-x}As$ layer, and it is the barrier X CB that causes the intensity decrease. Details of the pressure dependence of the intensities will be discussed later.

Figures 4(a) and 4(b) show the effect of pressure on the E_{1h}^{Γ} and E_{1l}^{Γ} transitions for sample 1 at 80 K for different well widths. Also shown in Fig. 4(a) is the pressure dependence of the PL from the $Ga_{0.67}Al_{0.33}As$ barrier. The lines through the data are due to least-squares fits to the function.

$$E_{1h}^{\Gamma}(P)=E_{1h}^{\Gamma}(0)+\alpha P\ , \tag{1}$$

where the energies are in eV and the pressure P is in kbar. Similar data are obtained for sample 1 at 150 K (not shown). Figures 5(a) and 5(b) show the pressure dependence of the photoluminescence of sample 2 at 80 K. The pressure dependence of the $Ga_{0.7}Al_{0.3}As$ barrier is also shown in Fig. 5(a). The linear pressure coefficients (α) obtained from the quantum-well data in Figs. 4 and 5 are shown in Fig. 6. Two separate pressure runs were made on each sample to assure the accuracies of the α's. Figure 6 displays α as a function of the transition energy in the quantum well at 80 K and atmospheric pressure. A systematic decrease in α is seen with increasing transition energy (or narrower well widths). The decrease in α observed, in the range of L_z used in samples 1 and 2, is about 5%. The statistical error in each value of α is about $\pm 1\%$. The lines through data are a guide to the eye. The trend of decreasing α is the same for both samples. (The actual pressure coefficients themselves are not the same, due to the difference in aluminum mole fraction in the barrier.)

The well-width dependence of the pressure coefficients, though small, is a real effect outside the experimental uncertainty. We have established this by the following analysis. Samples 1 and 2 are such that in a given sample there are four and five single wells, respectively, separated by wide barriers (see Figs. 1 and 3). For a given sample and pressure, all the wells experience the *same* pressure. One can measure the energy positions of the E_{1h}^{Γ} or E_{1l}^{Γ} transitions accurately to within 0.01%. Hence the *differences* between the energies of E_{1h}^{Γ} (or E_{1l}^{Γ}) transitions for separate wells can be determined to an accuracy of 0.01% at a given pressure.



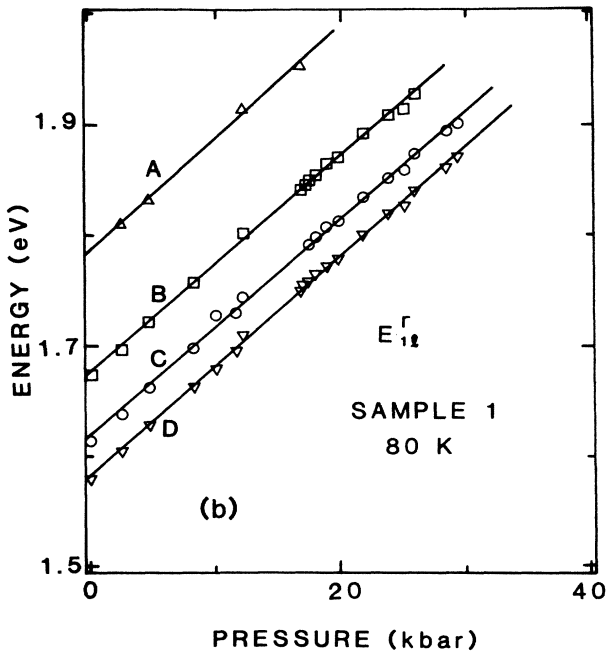


FIG. 4. (a) Energies of E_{1h}^{Γ} transitions ($n=1$ electron to $n=1$ heavy hole) as a function of pressure for wells A–D in sample 1 at 80 K. The pressure dependence of $Al_{0.33}Ga_{0.67}As$ barrier PL is also shown. The solid lines are linear least-squares fits to the data. (b) Same as (a) for the E_{1l}^{Γ} transition in sample 1 ($n=1$ electron to $n=1$ light hole) at 80 K.

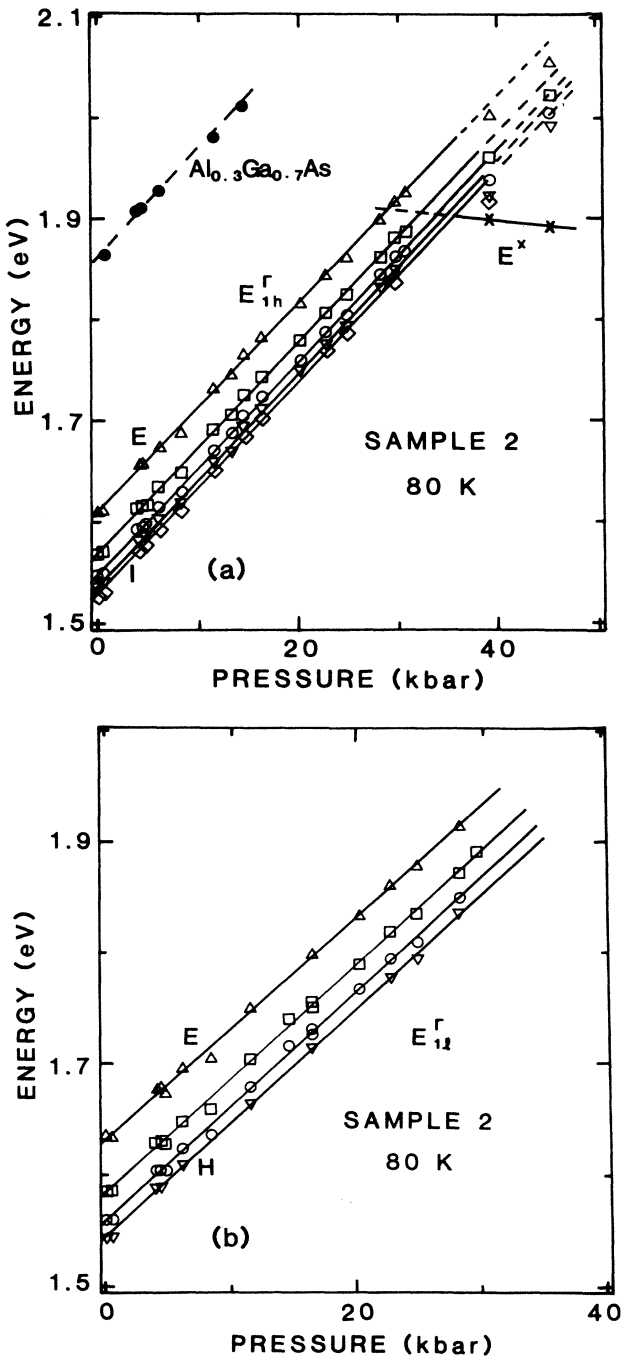


FIG. 5. (a) Pressure dependence of E^{Γ}_{1h} for wells $E-I$ and the $Al_{0.3}Ga_{0.7}As$ barrier PL for sample 2 at 80 K. (b) Same as (a) for the E^{Γ}_{1l} transition at 80 K.

A plot of the energy difference between, say, wells A and D for their E^{Γ}_{1h} transitions, as a function of pressure yields a slope that *directly* measures the difference in α for wells A and D. Instead of fitting E^{Γ}_{1h} energies to linear functions and then subtracting their pressure coefficients to obtain the differences in α, we first subtract the energies of E_{1h} transitions and then fit these difference energies to a linear function, to obtain the difference in α. The advantage is that statistical errors due to the uncertainty in the pressure (± 0.5 kbar), as deduced from the linewidth of the ruby flourescence, are not compounded in the difference fit.

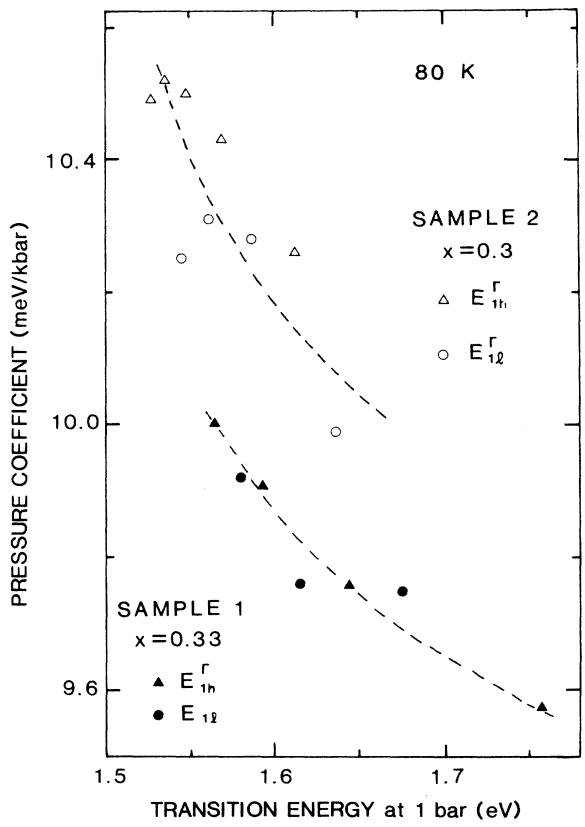


FIG. 6. Pressure coefficient (α) versus transition energies E^{Γ}_{1h} and E^{Γ}_{1l} for different wells in samples 1 and 2. Note the different Al mole fractions. The lines through data points are guides to the eye.

If two wells had the same α, such a difference fit would yield a line of zero slope. If the two had energies that approached one another with pressure, we would get a negative slope. Figure 7 shows the difference analysis for sample 1, at 80 K. The lines through data are due to least-squares fits. It is very clear that the slopes of these lines are not zero, and moreover are larger in magnitude for the pairs of wells of widely different widths and hence widely separated transition energies. Similar difference fits were made for all pairs of wells in samples 1 and 2, confirming the trend seen in Fig. 7. The variation of α with well width is thus established to within 0.1%, due to the nature of our samples which contain many wells in the same sample. Figure 8 shows these differences in α as a function of difference in transition energy, for both samples, at 80 K. It is clear that the data for both samples fall on a smooth curve and are independent of the aluminum mole fraction. The same trend is seen at 150 K. Leburton and Kahen[12] have calculated a similar trend in α from an analysis of the absorption data[13] taken at room temperature and below 10 kbar, with a considerably larger error in α (± 0.4 meV/kbar).

There are many effects which may account for the L_z dependence of α. Pressure decreases the lattice constant

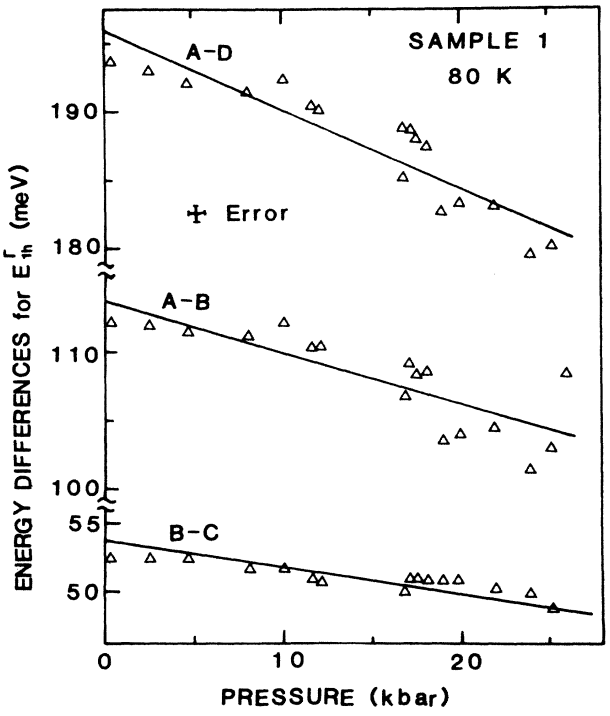


FIG. 7. Energy difference of E^{Γ}_{1h} transition for pairs of quantum wells *A* and *D*, *A* and *B*, and *B* and *C* versus pressure for sample 1 at 80 K. The solid lines are due to linear least-squares fits. Note that the magnitude of the slopes are larger for pairs of wells with larger differences in transition energies, and hence L_z.

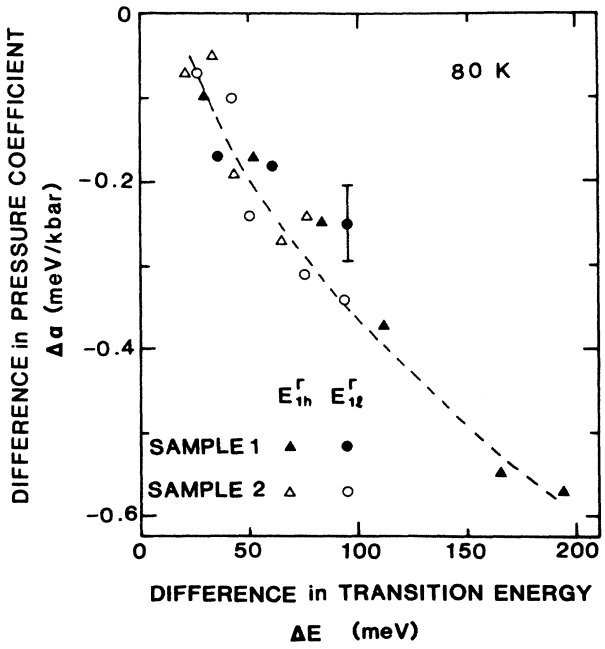


FIG. 8. Change in α versus the change in E^{Γ}_{1h} transition energy for samples 1 and 2 at 80 K. The data are from difference analyses similar to that shown in Fig. 7.

and also narrows L_z in a heterostructure, thus raising the quantum levels. The increase in α due to this effect is countered by the higher rigidity of the bands away from the band edge, which tends to decrease α. These two effects have been included in Ref. 12. In addition, there are several other effects. Due to the different pressure coefficients of the GaAs well and $Ga_{1-x}Al_xAs$ barriers, the height of the barrier itself changes with pressure. The α for $Ga_{1-x}Al_xAs$ is known to depend[14] on x. At 300 K, α increases linearly[14] from 11.4 to 12.2 meV/kbar as x varies from 0 to 0.25. Beyond $x=0.25$, α decreases nonlinearly down to 10.2 meV/kbar for $x=0.4$ and remains more or less constant up to $x=0.5$. The values of α for bulk GaAs (Refs. 8–10), measured by different techniques and at different temperatures, range from 12.3 to 10.7 meV/kbar between 300 and 5 K. We have measured the α for the barriers in samples 1 and 2 to be 9.8 and 10.5 meV/kbar, respectively, at 80 K, in reasonable agreement with Ref. 14. Our measurement of α for bulk GaAs (Ref. 15) at 80 K is 10.7 ± 0.1 meV/kbar, consistent with the low-temperature values.[10,16] Since α for GaAs is higher than that for the barriers, one would expect a decrease in barrier height with pressure resulting in a decrease in α. Changes in exciton binding energies also affect α. Magneto-optic experiments[17] have shown that the exciton binding energy increases with decreasing well width, and that the increase is somewhat faster than that expected from theory. Furthermore, the CB effective mass in the bulk is calculated[18] to increase by about 20% between 0 and 40 kbar, which affects both the subband energies and the exciton binding energies in the quantum wells, which in turn affect α. Theoretical calculations which quantitatively include all these effects are not available at present. The net decrease in α with the decrease in L_z in our samples is a combined effect due to the various competing mechanisms mentioned above.

B. Valence-band offsets

The photoluminescence intensity for the Γ-band-derived transitions (E^{Γ}_{1h} and E^{Γ}_{1l}) decreases dramatically when the Γ-*X* crossover pressure is approached. Weak structures which show a slightly negative pressure coefficient are observed in the crossover pressure region. It is known that the *X*-band edge has a small negative pressure coefficient of -1.34 ± 0.04 meV/kbar (Ref. 10) for bulk GaAs. Figure 5(a) indicates peaks labeled E^X suggesting that they are derived from the *X* band.

We have reported a detailed investigation[7,15] of sample 3, which was a multiple-quantum-well structure with 40 periods of 150 Å GaAs wells separated by 100-Å-thick barriers of $Ga_{0.75}Al_{0.25}As$ grown on a GaAs substrate by MBE. For pressures beyond the Γ-*X* crossover, a weak peak labeled E^X is seen at lower energies than E^{Γ}_{1h} and shifts linearly with $\alpha=-1.3\pm0.1$ meV/kbar (Fig. 9). The negative α and its closeness to the bulk value clearly suggest that it is an *X*-band-derived transition. By extrapolating the data back to atmospheric pressure, the value of E^X is found to be lower than that of the *X* minimum of bulk GaAs at 80 K (E^X_g in Fig. 9). While free exciton binding energies[19] in the *X* band may be small

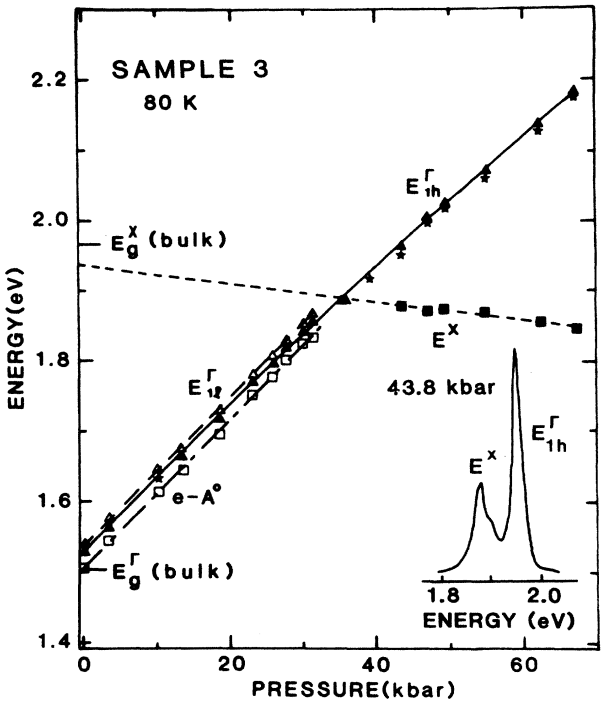


FIG. 9. Pressure dependence of E^{Γ}_{1h}, E^{Γ}_{1l}, E^{X}, and e-A^0 transitions. E^{Γ}_{g} and E^{X}_{g} denote the energies of the Γ and X conduction-band edges in bulk GaAs at atmospheric pressure (Ref. 14). Inset is the spectrum obtained at 43.8 kbar, where the E^{X} band is clearly observed and has a pressure coefficient of -1.3 ± 0.1 meV/kbar.

(5 to 10 meV), donor bound excitons or free to bound transitions can have slightly higher binding energies. The binding energy we obtain is 35 ± 6 meV, using our $P=0$ intercept of 1.936 ± 0.006 eV and the Aspnes value[20] of E^{X}_{g} at 80 K, 1.971 eV. Furthermore, the transition we observe is $\sim$25 meV lower in energy than the D^{0}_{X} observed in bulk GaAs by Wolford and Bradley,[10] after correcting for the differences in temperature between our work and Ref. 10. This large shift below D^{0}_{X} suggests that E^{X} might not be a transition within the GaAs well.

Due to the band offsets between the well and barrier layers, the most likely possibility is the formation of a type-II heterostructure, where recombination occurs between an electron in the X CB of the $Al_xGa_{1-x}As$ barrier and the GaAs well. Such recombination is indirect in **k** space as well as in real space, and is consequently expected to be weak in intensity, as in fact E^{X} is. For valence offsets of $\sim$15% or more, the $Al_{0.25}Ga_{0.75}As$ X CB is lower than the GaAs X CB. Recombination is then possible between the band edge (for wide barriers) or the $n=1$ X CB electron (for narrow barriers) and the GaAs heavy hole. Identifying E^{X} as this type-II recombination and extrapolating E^{X} to $P=0$, one can determine the valence-band offset to be

$$E_v = E^{X}_{g}(Al_xGa_{1-x}As) - E^{X} + h \ ,$$

where h is the heavy-hole confinement energy, about 2 meV for a 150-Å well. E^{X} is known from our data to be 1.936 eV. The X-band energy in $Al_xGa_{1-x}As$, however, varies considerably between different values available in literature.[21] In conjunction with our measurements of bulk $Al_{1-x}Ga_xAs$ under pressure,[15] and other available data, we place E^{X}_{g} ($Al_{0.25}Ga_{0.75}As$) at 2.024 ± 0.012 eV, the error bar indicating the spread in the values. This gives a valence-band offset of 90 ± 12 meV, or as a fraction of the total offset $Q_v=0.30\pm0.04$. Clearly, an accurate value of the X-band energy in $Al_xGa_{1-x}As$ is necessary for this calculation, and this will be discussed in more detail in a later publication.[15]

C. Crossover pressures

It is well known that the pressure-induced Γ-X CB crossover in bulk GaAs is signaled by a decrease in the PL intensity. In the quantum-well structures, the Γ-X subband crossover is similarly indicated by the E^{Γ}_{1h} transition. The intensities of the E^{Γ}_{1h} transitions decrease by about four orders of magnitude near the crossover, as has been previously observed[7] in sample 3 ($L_z=150$ Å). Similar effects are seen in the multiwell samples 1 and 2. The peaks due to the narrowest wells A and E in Figs. 2 and 3 decrease in intensity at much lower pressures than the peaks due to wider wells. For example, the 96-Å well (D) in Fig. 2 hardly changes in intensity from 1 bar to 25.2 kbar, whereas the 26-Å well (A) decreases by a factor of 10^4 in the same pressure range. Figure 10 shows these features for three quantum wells of widths 26, 47, and 150 Å, from samples 1, 2, and 3, respectively. The intensities of these peaks are normalized so that the fractional changes in intensity with pressure can be directly compared.

The crossover between the energies E^{Γ}_{1h} and E^{X} in sample 3 (Fig. 9) occurs at 35.5 kbar, which is the pressure at which the intensity of the E^{Γ}_{1h} transition has decreased to $\sim$10% of its $P=0$ value (see Fig. 10). Using this intensity decrease as the criterion, we obtain the crossover pressures P_c (accurate to ±2 kbar) for the wells in samples 1 and 2 from plots such as Fig. 10. The dependence of P_c

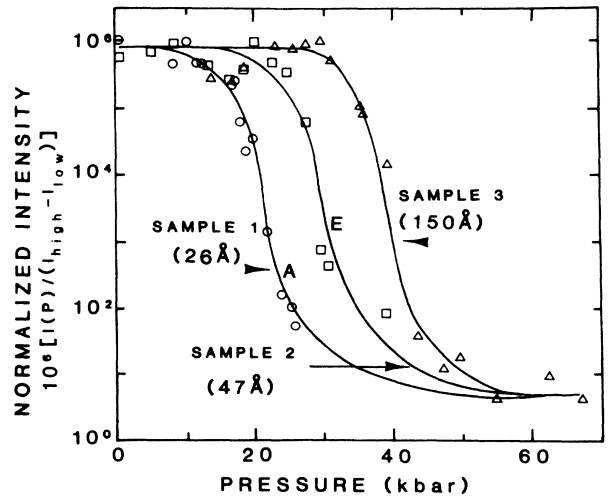


FIG. 10. Normalized intensity of the E^{Γ}_{1h} peaks as a function of pressure.

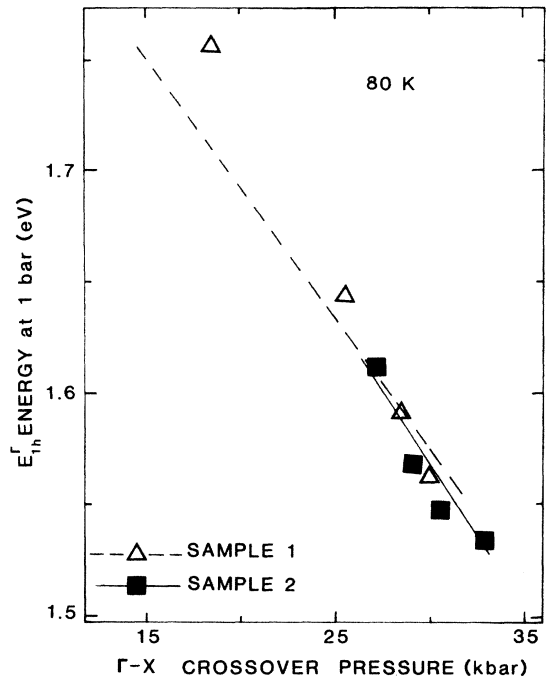


FIG. 11. A plot of the E^{Γ}_{1h} transition energy at 1 bar versus the pressure P_c defined in the text for samples 1 and 2 (data points). The lines are fits to a simple model, described in the text.

on well width is shown in Fig. 11, where we plot the energy of E^{Γ}_{1h} at 1 bar versus P_c. It is very clear from this plot that P_c decreases with increasing E^{Γ}_{1h} transition energy (decreasing L_z).

This effect can be understood by the following argument. The crossover pressure P_c for the Γ and X conduction subbands in quantum wells depends on the difference in their subband energies $E^{X}_{1c}-E^{\Gamma}_{1c}$ and their pressure coefficients α. The measured decrease in α for L_z between 150 and 26 Å is about 10% (see Fig. 6) as compared to a 52% decrease in $E^{X}_{1c}-E^{\Gamma}_{1c}$. Thus, the crossover pressure is mainly governed by $E^{X}_{1c}-E^{\Gamma}_{1c}$. The smaller the difference $E^{X}_{1c}-E^{\Gamma}_{1c}$, the lower the pressure at which the bands cross. Neglecting small effects such as confinement energies in the X CB and the binding energies of the excitons, as they are small compared to E^{Γ}_{1h}, the crossover pressure is given by

$$P_c=\frac{E^{X}-E^{\Gamma}_{1h}(P=0,L_z)}{\alpha_{\Gamma}-\alpha_{X}} . \tag{2}$$

From Eq. (2), we calculate P_c for each sample using $\alpha_X=-1.3$ meV/kbar, the measured values of α_Γ for each well (Fig. 6) and E^X. The values of E^X are estimated to be 1.921 eV for sample 2 ($x=0.3$), and 1.914 eV for sample 1 ($x=0.33$), using $Q_v=0.3$ and the X CB energy of $Al_xGa_{1-x}As$.[15] The calculated values of P_c are shown by the solid (sample 2) and dashed (sample 1) lines in Fig. 11. The calculation is in good agreement with experiment except for the 26-Å well.

The implication of this effect is that the direct transitions which influence the device properties (e.g., the laser energy) can be pressure tuned, at best, up to the crossover energy. The useful tuning pressure range, however, will be smaller as L_z decreases.

Uniaxial strains due to inhomogeneities in pressure, as observed from the linewidths of the quantum-well transition were less than ± 0.1 kbar. While it is possible to get lateral strains due to the different bulk moduli in the well and barrier layers, this difference is small[21] [B (GaAs)=755 kbar and B ($Ga_{0.7}Al_{0.3}As$)=763 kbar]. This translates to an increase in the lattice mismatch from 0.04% at 1 bar to 0.05% at 30 kbar. Any effects due to this added strain would be too small to detect.

IV. CONCLUSIONS

A study of the pressure dependence of GaAs-$Ga_{1-x}Al_xAs$ heterostructures has shown that the pressure coefficients (α) exhibit a small decrease with decreasing well width (L_z). By using samples containing wells of different L_z grown on the same substrate, these differences in α are measured directly. Various mechanisms that lead to the above effect are discussed. From the intensity data it is shown that the direct-indirect crossover of the conduction-subband edges also depends on well width and occurs at lower pressures for narrower wells. A level associated with the X band is seen beyond the crossover pressures. Identifying this transition as due to a recombination between the $Al_xGa_{1-x}As$ X CB and the GaAs valence heavy hole allows us to obtain the valence-band offset to be $Q_v=0.30\pm0.04$, a calculation which is accurate to within the exciton binding energies.

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