PHYSICAL REVIEW B VOLUME 31, NUMBER 6 15 MARCH 1985

Photoluminescence studies of a GaAs-$Ga_{1-x}Al_xAs$ superlattice at 8–300 K under hydrostatic pressure (0–70 kbar)

Umadevi Venkateswaran, Meera Chandrasekhar, and H. R. Chandrasekhar
Department of Physics and Astronomy, University of Missouri-Columbia, Columbia, Missouri 65211

T. Wolfram
Amoco Research Center, Standard Oil Company (Indiana), Warrenville Road, P.O. Box 400, Naperville, Illinois 60566

R. Fischer, W. T. Masselink, and H. Morkoç
Coordinated Science Laboratory, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801

(Received 3 December 1984; revised manuscript received 8 January 1985)

The pressure dependence (0–70 kbar) of the photoluminescence spectra from a GaAs-$Ga_{1-x}Al_xAs$ multiple-quantum-well (MQW) structure is reported at 8–300 K. The pressure coefficients of the E_{nh}^{Γ} and E_{nl}^{Γ} ($n=1,2$) transitions between the Γ conduction subbands and the heavy- (h) and light- (l) hole bands are determined. The energy of E_{1h}^{Γ} has a sublinear pressure dependence for the MQW at 80 K but not at 300 K. The reverse is true for the band gap of bulk GaAs. In the MQW the Γ and X bands cross around 35 kbar and a new transition E_{1h}^{X}, observed for the first time, has a pressure coefficient of -1.3 ± 0.1 meV/kbar at 80 K. the X–Γ band separation at atmospheric pressure in the bulk is deduced to be 45–50 meV lower than the previously accepted value of 0.464 eV. The carrier lifetimes in the Γ and X bands and the Γ-to-X scattering times are deduced.

I. INTRODUCTION

We present the first high-pressure (0–70 kbar) study of the energy levels in a GaAs-$Ga_{1-x}Al_xAs$ multiple-quantum-well (MQW) structure, at temperatures of 8–300 K. While the energy levels of MQW's have been investigated in detail by several optical techniques,[1] the effect of hydrostatic pressure has received much less attention. Previous studies[2,3] were limited to pressures of 10 kbar or less at 300 K. In this work we obtain the pressure coefficients of several of the emission lines. We have also made the first observation of a quantized energy level in the X conduction band.

It is well known that in the GaAs-$Ga_{1-x}Al_xAs$ multilayer structures potential wells are formed in the GaAs layers. The energy levels in the wells are discrete, and when dispersion is included, the levels form subbands whose energies are determined by the well width and depth. The subbands are labeled by their quantum numbers n. The valence band (VB) splits into light- (l) and heavy- (h) hole subbands. For spontaneous emission,[1] strongly allowed transitions occur between conduction band (CB) and heavy- (light-) hole subbands with the same n value, and are labeled E_{nh} (E_{nl}). We also introduce a superscript Γ or X to denote the CB that originates the electron (e.g., E_{nh}^{Γ}).

In this work we report the pressure dependence from 0 to 70 kbar of some of the transitions observed in emission. The lowest energy transition, E_{1h}^{Γ}, is studied at 8, 80, and 300 K. At low temperatures the deeper valence subbands such as the $n=1$ light hole or $n=2$ heavy hole are occupied, and consequently recombination to these bands is weak. As the temperature is increased, thermal depopulation of these deeper states allows transitions to occur, and the E_{1l}^{Γ} is observed at 80 K and E_{2h}^{Γ} at 300 K. We also observe recombination between an electron and a neutral acceptor impurity (carbon), labeled e-A^0 at 8 and 80 K.

We observe that E_{1h}^{Γ}, E_{1l}^{Γ}, E_{2h}^{Γ}, and e-A^0 all increase in energy, with pressure, similar to the E_0 transition in bulk GaAs.[4-7] Around 35 kbar, the Γ and X CB's cross and a new peak due to the $n=1$ subband at X, labeled E_{1h}^{X} is observed, we believe, for the first time. We find that the $X-\Gamma$ energy separation in bulk GaAs at atmospheric pressure[8] is 45–50 meV lower than the previously accepted value of 0.464 eV. These results are discussed in Sec. III.

II. EXPERIMENT

The MQW consisted of 40 periods of 150-Å GaAs–100-Å $Ga_{0.75}Al_{0.25}As$ grown on a GaAs substrate by molecular-beam epitaxy. The substrate was thinned to ~ 30 μm and the sample mounted in a Merrill Bassett[9] gasketed diamond-anvil cell with argon as the pressure transmitting fluid. Fluorescence from the ruby R_1-R_2 lines was used to calibrate[10] the pressure, which remained hydrostatic through 70 kbar. The cell was attached to a cryostat and data were taken at 8, 80, and 300 K. Photoluminescence was excited using 0.4–10 mW of 5145 Å radiation from an Ar^+ laser.

In Figs. 1 and 2 are shown the photoluminescence spectra for different pressures at three temperatures. A sharp line due to the E_{1h}^{Γ} and a weaker peak due to e-A^0 transitions are observed at 8 K (Fig. 1). At 80 K (Fig. 2) we observe the e-A^0, E_{1h}^{Γ}, and E_{1l}^{Γ}, and at 300 K the E_{1h}^{Γ} and E_{2h}^{Γ} transitions. Since all the dominant transitions could be observed at 80 and 300 K we chose these two temperatures for detailed studies over a wide pressure range.

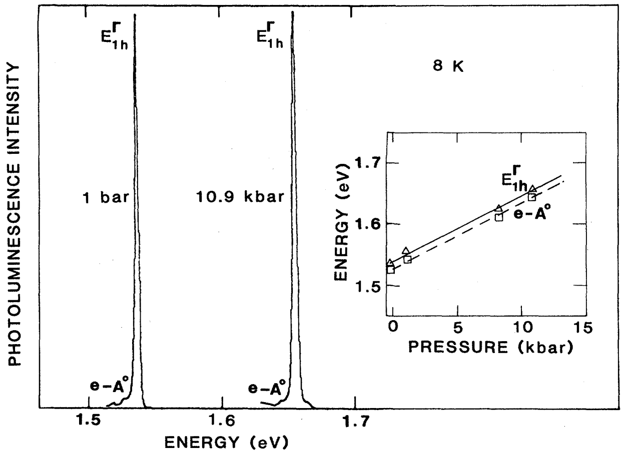

FIG. 1. Luminescence spectra of the GaAs-$Ga_{1-x}Al_xAs$ superlattice at 1 bar and 10.9 kbar at 8 K. In the inset are plotted the peak energies of the e-A^0 (squares) and E^{Γ}_{1h} (triangles), and fits to linear functions by dashed and solid lines, respectively.

The energies of all transitions originating in the Γ CB increase with pressure (Fig. 2), in a manner similar to that observed in bulk GaAs.[4–7] At 35.5 kbar, the Γ and X conduction bands cross, and a new peak E^{X}_{1h}, arising from recombination between the $n=1$ quantized energy level in the X CB and heavy hole at Γ is observed at 80 K (Fig. 2, top panel). At higher pressures, the E^{Γ}_{1h} peak broadens and loses intensity but continues to increase in energy, while the E^{X}_{1h} peak decreases slowly in energy typical of the indirect X CB transition in the bulk.[5]

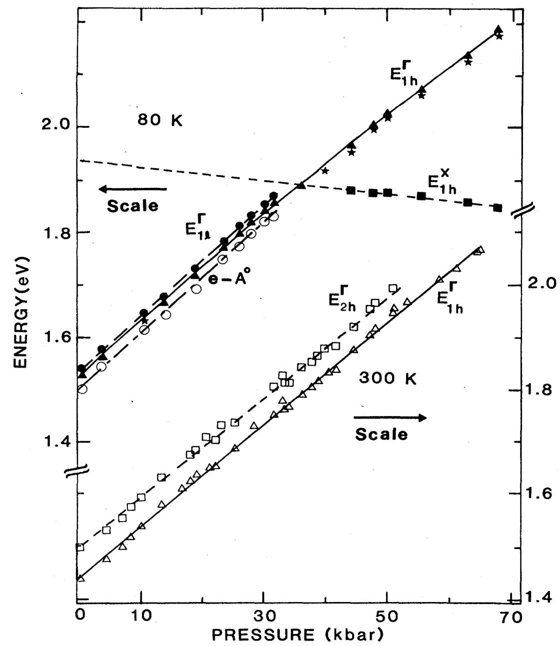

FIG. 3. Energies of the E^{Γ}_{1h} (solid triangles and stars), E^{Γ}_{1l} (solid circles), e-A^0 (open circles), and E^{X}_{1h} (solid squares) as a function of pressure at 80 K. The stars represent the energies of E^{Γ}_{1h} at high laser powers (40–60 mW) when heating effects shifted the peak. The stars were not included in the fits (lines through data points) discussed in the text. The energies of E^{Γ}_{1h} (open triangles) and E^{Γ}_{2h} (open squares) at 300 K, are fit to linear functions.

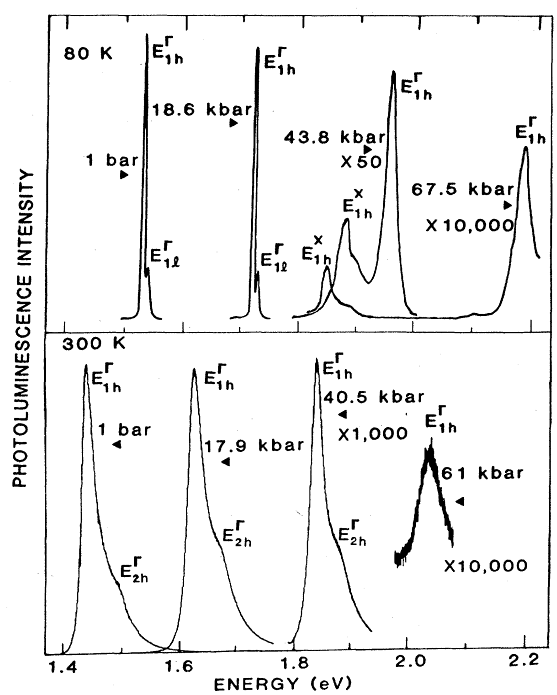

FIG. 2. Luminescence spectra of the superlattice for several pressures, at 80 K (top panel) and 300 K (lower panel). Note the appearance of the E^{X}_{1h} peak at pressures above 40 kbar, coupled with the broadening and decreasing intensity of the E^{Γ}_{1h} peak (80 K).

Figure 3 shows a plot of the peak energies of the e-A^0, E^{Γ}_{1h}, E^{Γ}_{1l}, and E^{X}_{1h} transitions as a function of pressure. The weaker peaks, e-A^0 and E^{Γ}_{1l}, are observed up to the crossover, beyond which the lower intensity and larger linewidth of E^{Γ}_{1h} obscures them. The curves through the data are least-squares fits to the data. The best fits (Table I) were obtained when E^{Γ}_{1h} was fit to the quadratic function

$$E^{\Gamma}_{1h}(P) = E^{\Gamma}_{1h}(0) + \alpha_{\Gamma} P + \beta_{\Gamma} P^2 \ , \qquad (1)$$

where P is pressure in kbar. The other transitions fit best to linear functions, i.e., $\beta_{\Gamma}=0$ for E^{Γ}_{1l} and e-A^0 and

$$E^{X}_{1h}(P) = E^{X}_{1h}(0) + \alpha_{X} P \ . \qquad (2)$$

At 300 K (Fig. 2, bottom panel and Fig. 3) the E^{Γ}_{1h} peaks increase linearly in energy up to the highest pressures employed (Table I). There is no significant broadening of these peaks, despite the lower intensity, which obscures E^{Γ}_{2h} beyond 50 kbar. E^{X}_{1h} is not observed at 300 K, and the only indication of the Γ-X crossover comes from the falling intensity.

III. DISCUSSION

While there are several similarities between the behavior of the energy levels in the MQW and bulk GaAs under pressure, there are also several notable differences.

First, both photoluminescence and absorption measure-

TABLE I. Pressure coefficients of the transitions in MQW superlattices and bulk GaAs. α_Γ (α_X) and β_Γ are the linear and quadratic coefficients, respectively (see text).

Sample	Transition	T (K)	α_Γ (α_X) (meV/kbar)	β_Γ (meV/kbar²)	Pressure (kbar)
150 Å well	E^Γ_{1h}	300	9.7 ±0.09		0–70
(present		80	10.7 ±0.2	−0.016 ±0.003	0–70
work)		8	10.7 ±0.2		0–11
	E^Γ_{2h}	300	9.5 ±0.1		0–50
	E^Γ_{1l}	80	10.45 ±0.07		0–32
	e-A^0	80	10.5 ±0.1		0–32
		8	10.6 ±0.3		0–11
	E^X_{1h}	80	(−1.3 ±0.1)		44–70
120 Å well	E^Γ_{1h}	300	11.5[a]		0–3.5
(laser)			8.5[a]		3.5–10
85 Å well (absorption) (n=1–3)	$E^\Gamma_{nh(l)}$	300	11.5[b]		0–10
Bulk GaAs	E^Γ_g	300	12.6 ±0.1[c]	−0.038 ±0.001	0–180
		300	12.3 ±0.02[d]	−0.031 ±0.001	0–110
		120	9.5[d]		0–100
		77	10.7[e]		0–10 (uniaxial)
		5	10.6 ±0.2[f]		0–80
	E^X_g	380	(−2.7 ±0.5[g])		0–70
		120	(−2.8 ±0.8[d])		40–55
		5	(−1.45 ±0.1[f])		0–80

[a]Reference 2.
[b]Reference 3.
[c]Reference 4.
[d]Reference 5.
[e]Reference 11.
[f]Reference 7.
[g]Reference 6.

ments in the bulk at 300 K show a marked sublinearity[4,5] in the E^Γ_g-vs-P curve (Table I). In contrast, E^Γ_{1h} in the MQW shows a very linear shift with a pressure coefficient about 25% smaller than the bulk value. At lower temperatures (120–5 K), however, the E^Γ_g vs P in the bulk is linear[5,7] while E^Γ_{1h} in the MQW (at 80 K) is sublinear. The coefficient α_Γ for the bulk and MQW at low temperatures are nearly the same,[5,7,11] which is not the case at 300 K.

The E^X_{1h} peak, observed between 44 and 70 kbar at 80 K, has a small linear negative pressure coefficient, somewhat smaller on the average than that observed in the bulk.[5–7] By extrapolating the data back to atmospheric pressure, we obtain $E^X_{1h} - E^\Gamma_{1h} = 0.408 \pm 0.007$ eV. One has

$$E^\Gamma_{1h} = E^\Gamma_g + \Delta E^\Gamma_c + \Delta E^\Gamma_v \tag{3}$$

and

$$E^X_{1h} = E^X_g + \Delta E^X_c + \Delta E^\Gamma_v \ , \tag{4}$$

where E^Γ_g (E^X_g) are the Γ (X) gaps at atmospheric pressure in the bulk,[8] $E^X_g - E^\Gamma_g = 0.464$ eV at 80 K, and ΔE^Γ_c, ΔE^X_c (ΔE^Γ_v) are the energy differences between the $n=1$ subband and the conduction- (valence-) band edges; we obtain $\Delta E^X_c - \Delta E^\Gamma_c = -0.056 \pm 0.007$ eV. ΔE^Γ_c is believed to be $\sim 65\%$ of the difference $E^\Gamma_{1h} - E^\Gamma_g$, i.e., $\Delta E^\Gamma_c \simeq 0.014$ eV (since $E^\Gamma_{1h} - E^\Gamma_g$ is 0.021 eV), so that $\Delta E^X_c = -0.042 \pm 0.007$ eV. This means that the $n=1$ subband in the X band is *below* the band edge. This is clearly unphysical if the X band is considered to be a one-dimensional well. Since all the energies relating to the MQW have come from one consistent set of data, it follows that the Γ–X separation in the bulk should be lower by about 0.045–0.050 eV. This would be consistent with a value of $\Delta E^X_c \sim 0.002$ eV since the X band has a mass about 13 times that of the Γ band.

Comparisons with previous superlattice studies under pressure[2,3] are also interesting (Table I). Kirchoefer *et al.*[2] studied emission from a laser up to 10 kbar, and reported a "kink" in the E^Γ_{1h} vs P curve at 3.5 kbar. This kink was attributed to shears of unknown origin which caused the light-hole band to move at a slower rate than the heavy-hole band, and eventually dominate the slope at 8.5 meV/kbar. An absorption study also by Kirchoefer *et al.*[3] for pressures up to 10 kbar, however, showed no such kink in the curve. In our data, we see no evidence of a changing energy separation between the light- and heavy-hole bands up to 32 kbar, indicating a truly hydrostatic environment, and our linear pressure coefficients α_Γ are similar to those found in Ref. 3 (Table I).

We observe that the intensity of the E^Γ_{1h} peak decreases about four orders of magnitude as the X and Γ bands cross both at 80 and 300 K (Fig. 4). The experimental values were fit to the expression[5]

$$I = I_0\left[[1 + A\exp(\Delta E/kT)]^{-1} + \frac{\tau_{\Gamma-X}}{\tau_{\rm rad}} \right] \ , \tag{5}$$

where I_0 is a constant, $\tau_{\Gamma-X}$ the Γ-to-X scattering time for conduction electrons, and $\tau_{\rm rad}$ is the radiative recombination time for the direct transition. ΔE is the energy difference between Γ and X CB's (a function of P), computed using the pressure coefficients in Table I. In the bulk,

$$A = 6\left(\frac{m_X}{m_\Gamma}\right)^{1/2} \frac{\tau_\Gamma}{\tau_X} \ , \tag{6}$$

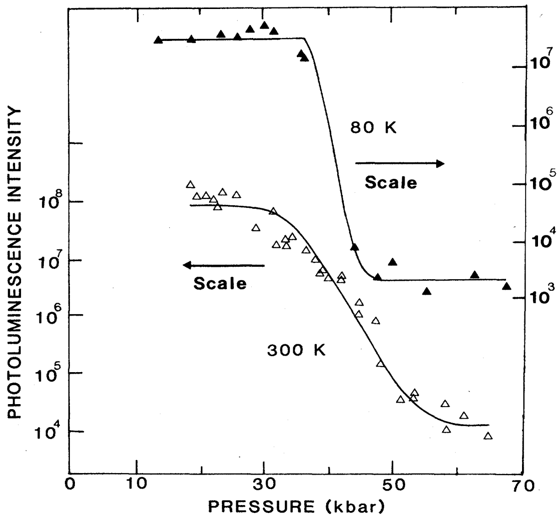

FIG. 4. Photoluminescence intensity of the E_{1h}^{Γ} peak as a function of pressure at 80 K (solid triangles) and 300 K (open triangles) fit to Eq. (5) (solid lines).

where m_X (m_Γ) are the average effective masses and τ_X (τ_Γ) the lifetimes of minority carriers in the X (Γ) bands. The factor of 6 arises from the degeneracy of the equivalent X bands in the bulk, and this is replaced by 2 for the MQW structure.

With I_0, A, and $\tau_{\Gamma-X}/\tau_{\rm rad}$ as parameters the best fit to the data gave the solids lines in Fig. 4. $\tau_{\Gamma-X}$ is found to be $\sim 10^{-17}$ sec while $\tau_{\rm rad} \sim 10^{-13}$ sec at 80 and 300 K. $\tau_{\rm rad}$ is obtained from the half-width of E_{1h}^{Γ} at 60 kbar (i.e., well beyond the $\Gamma-X$ crossover). At 300 K we obtain $A = 1.9 \pm 0.5$, which is consistent with being a third of the bulk value[5,6] of $A = 7$. While the bulk value is unchanged[5] at 120 K, we obtain $A = 0.006 \pm 0.004$ for the MQW at 80 K. Since neither the masses nor τ_Γ change substantially with temperature, Eq. (6) implies that τ_X has increased by a factor of (460 ± 330) between 300 and 80 K.

The linewidth of the E_{1h}^{Γ} peak is constant at about 25 cm^{-1} between 0 and 30 kbar, then increases monotonically to ~ 250 cm^{-1} at 70 kbar, indicating a decrease in the electron lifetime.

IV. CONCLUSIONS

In this study of a MQW under high hydrostatic pressures, we have obtained the pressure coefficients of the E_{1h}^{Γ}, E_{2h}^{Γ}, E_{1l}^{Γ}, and e-A^0 transitions. We report the first observation of a transition involving a quantized energy level in the indirect X CB, E_{1h}^{X}, and obtained its pressure coefficient. We note the following differences between the MQW and the bulk.

(a) At 300 K, E_g^{Γ} vs P is strongly sublinear in the bulk while E_{1h}^{Γ} vs P is linear in the MQW with a smaller coefficient α_Γ.

(b) E_g^{Γ} vs P is linear in the bulk at 120 and 5 K while E_{1h}^{Γ} vs P at 80 K is sublinear in the MQW with the same linear pressure coefficients.

(c) Our data suggest that the $X-\Gamma$ CB separation is 45–50 meV less than the accepted value of 0.464 eV at 80 K.

(d) From the intensity data, the $\Gamma-X$ scattering time $\tau_{\Gamma-X}$ and the carrier lifetime in the CB at Γ, τ_Γ (for $P > 35$ kbar) is found to be almost independent of temperature, while the carrier lifetime in the CB at X, τ_X, is several hundred times smaller at 300 K than at 80 K. In the bulk τ_Γ/τ_X appears to be independent of temperature.

ACKNOWLEDGMENTS

We thank D. A. Schiferl for many helpful discussions and C. R. Holmes and S. L. Potts for their expert help in the construction of the pressure cell and related cryogenic apparatus. The work at the University of Missouri was supported by the Standard Oil Company (Indiana). M.C. also thanks the Research Council of the University of Missouri. The work at the University of Illinois was funded by the Air Force Office of Scientific Research.

[1] R. C. Miller, D. A. Kleinman, W. A. Nordland, and A. C. Gossard, Phys. Rev. B **22**, 863 (1980); R. C. Miller, A. C. Gossard, W. T. Tsang, and O. Munteanu, *ibid.* **25**, 3871 (1982), and references therein.

[2] S. W. Kirchoefer, N. Holonyak, K. Hess, D. A. Gulino, H. G. Drickamer, J. J. Coleman, and P. D. Dapkus, Solid State Commun. **42**, 633 (1982).

[3] S. W. Kirchoefer, N. Holonyak, K. Hess, D. A. Gulino, H. G. Drickamer, J. J. Coleman, and P. D. Dapkus, Appl. Phys. Lett. **40**, 821 (1982).

[4] B. Welber, M. Cardona, C. Kim, and S. Rodriguez, Phys. Rev. B **12**, 5729 (1975).

[5] D. Olego, M. Cardona, and H. Müller, Phys. Rev. B **22**, 894 (1980).

[6] P. Yu and B. Welber, Solid State Commun. **25**, 209 (1978).

[7] D. J. Wolford and J. A. Bradley, Bull. Am. Phys. Soc. **29**, 291 (1984).

[8] D. E. Aspnes, Phys. Rev. B **14**, 5331 (1976).

[9] L. Merrill and W. A. Bassett, Rev. Sci. Instrum. **45**, 290 (1974).

[10] R. A. Noack and W. B. Holzapfel, in *High Pressure Science and Technology*, edited by K. D. Timmerhaus and M. S. Barber (Plenum, New York, 1979), Vol. I, p. 748.

[11] M. Chandrasekhar and F. H. Pollak, Phys. Rev. B **15**, 2127 (1977).