

Pressure dependence of the indirect band gap of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ alloys ($x = 0.70$ and 0.92) at low temperatures

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We have measured photoluminescence and Raman spectra of liquid-phase-epitaxy-grown $\text{Al}_x\text{Ga}_{1-x}\text{As}$ under hydrostatic pressure ($0 < P < 6$ GPa) at low temperatures ($T < 10$ K). The X -point exciton shifts with pressure to lower energies, for an aluminum concentration of $x = 0.70$ at a rate of $-16.2(5)$ meV/GPa, for $x = 0.92$ at $-16.5(5)$ meV/GPa. Taking into account the pressure dependence of the exciton binding energy, we determine the pressure shift of the indirect gap in these two alloys. For $x = 1$ (pure AlAs), an extrapolation leads to a rate of $-15.3(5)$ meV/GPa for the indirect gap. Using this value, data on the pressure shift of the valence-band offset in GaAs/AlAs superlattices are reexamined. This shift approaches a value of 4.7 meV/GPa for large superlattice periods (> 20 monolayers).

I. INTRODUCTION

Although AlAs is widely used as a constituent of semiconductor superlattices and quantum-well structures, comparatively little is known about its physical properties because of the difficulties encountered preparing high-quality bulk material. This situation also holds for the ternary alloy $\text{Al}_x\text{Ga}_{1-x}\text{As}$ with a high aluminum content ($x > 0.5$).

For low aluminum concentrations ($x < 0.42$ at low temperatures) $\text{Al}_x\text{Ga}_{1-x}\text{As}$ is a direct-gap semiconductor with the top of the valence band and the bottom of the conduction band both at the Γ point of the Brillouin zone. At higher concentrations the bottom of the conduction band moves to the X point so that the gap is now indirect. As of now there exist no measurements of the pressure dependence of indirect- and higher-lying direct-band-gap energies under hydrostatic pressure in these materials. Due to the lack of data, it has therefore been assumed that the band gaps of AlAs and $\text{Al}_x\text{Ga}_{1-x}\text{As}$ ($x > 0.5$) have the same pressure dependences as the corresponding band gaps of GaAs.¹⁻³ For the interpretation of measurements on GaAs/ $\text{Al}_x\text{Ga}_{1-x}\text{As}$ and GaAs/AlAs superlattices under pressure,⁴⁻¹⁵ however, it is necessary to know precisely the properties of the bulk constituents. Accurate values for the indirect gap of AlAs are especially important for the determination of the dependence of the valence-band offset^{16,17} in GaAs/AlAs superlattices on pressure.^{6,13} The reason for this is that the pressure dependence of the valence-band offset is determined as the (small) difference between the pressure shifts of the measured transition energy and of the AlAs indirect gap, so that even small errors in the pressure-induced energy shifts result in large relative errors for the valence-band offset.

We report here¹⁸ on photoluminescence measurements of the indirect gap of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ alloys and on Raman measurements at low temperatures ($T \leq 10$ K) under hydrostatic pressures up to 6 GPa. The samples studied had aluminum concentrations of $x = 0.70$ and 0.92 so

that it is possible to obtain the pressure dependence of the indirect band gap of AlAs by extrapolation to $x = 1$. This result is then used to determine the pressure dependence of the valence-band offset in GaAs/AlAs superlattices.

II. EXPERIMENT

The samples were grown by liquid-phase epitaxy 1–2 μm thick on semi-insulating GaAs(100) substrates. Their aluminum concentration was determined by double-crystal x-ray diffraction. By mechanical polishing most of the substrate was removed to a final thickness of ~ 30 μm . Pieces of $\sim 100 \times 100$ μm^2 size were loaded into a gasketed diamond-anvil cell¹⁹ with helium or xenon as the pressure medium. The cell was held in a cryostat at temperatures below 10 K. Although the cryostat used permits changing the pressure at low temperatures, pressure changes were always done with the cell heated to room temperature to ensure hydrostatic conditions. Pressure calibration was done both by the ruby luminescence method using the scale of Mao *et al.*²⁰ and by the shift of the GaAs substrate band gap.^{1,3} These two pressure readings agreed to within 0.03 GPa.

For excitation of the $x = 0.70$ sample the 514-nm line of an argon laser was used. Although the photon energy of this line (2.4 eV) is smaller than the direct gap of this sample (2.51 eV at ambient pressure), the phonon-assisted and alloy-disorder-induced absorption²¹ is large enough to obtain a good signal. For the $x = 0.92$ sample the alloy disorder is much smaller, so that it was necessary to excite above the direct band gap (2.90 eV at ambient pressure). This was done with the 413-nm line (3.0 eV) of a krypton laser. Since the direct gap shifts rapidly to higher energies with pressure, it was only possible to obtain reliable data for pressures below 3 GPa for this sample. The emitted light was dispersed by a 0.85-m-focal-length double monochromator and detected by a photomultiplier with a GaAs cathode in photon-counting mode.

III. PHOTOLUMINESCENCE AND RAMAN MEASUREMENTS

Figure 1 shows photoluminescence spectra of $\text{Al}_{0.92}\text{Ga}_{0.08}\text{As}$ at several pressures. With increasing pressure one observes a shift of the luminescence peaks to the low-energy side and a decrease of luminescence intensity, the latter caused by the increasing direct gap. This in turn reduces the absorption at the laser wavelength and the efficiency of the phonon-assisted luminescence. This efficiency is inversely proportional²¹ to the square of the energy difference between the direct and the indirect band gap (second-order perturbation theory).

To identify the origin of the three peaks seen, we have studied the dependence of the photoluminescence spectra on laser intensity (Fig. 2). The intensities of the peaks labeled D_1 and D_2 stay constant for a fourfold increase in laser intensity, whereas the intensity of the peak labeled E_{lum} increases in proportion to the laser intensity. This identifies D_1 and D_2 as defect-induced peaks (D_1 is probably due to a transition from a shallow donor to the valence band and D_2 is its phonon replica) and E_{lum} as an intrinsic luminescence, in this case the phonon-assisted recombination of $n=1$ (principal quantum number) X -point excitons. The same assignment was found by time-resolved measurements for samples near the direct-indirect crossover.²²

This assignment is further supported by the asymmetric line shape of the luminescence. For the phonon-assisted recombination of excitons one expects a line profile of the form²³

$$I_{\text{lum}}(E) = (\Delta E)^{1/2} \exp(-\Delta E / k_B T_{\text{lum}}), \quad (1)$$

convoluted with the instrumental response (here taken to be Gaussian), with $\Delta E = E - E_{\text{lum}}$, k_B Boltzmann's constant, and T_{lum} the temperature of the exciton gas. One should note that for this line shape E_{lum} is *not* the energy at the peak maximum (compare Fig. 2). The square root in Eq. (1) stems from the density of states (assuming parabolic bands near the band minima), the exponential from assuming Maxwell-Boltzmann statistics for the excitons, which is a very good approximation at low densities.

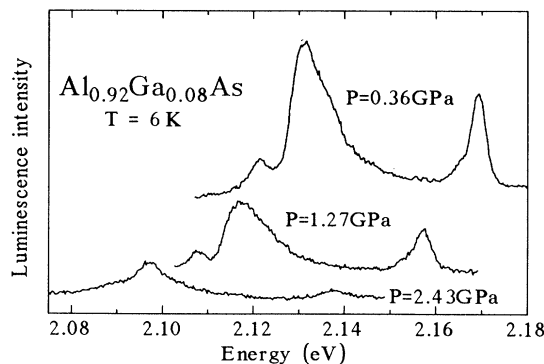


FIG. 1. Low-temperature photoluminescence spectra for $\text{Al}_{0.92}\text{Ga}_{0.08}\text{As}$ at several pressures. The spectra at 1.27 and 0.36 GPa were shifted upwards for clarity.

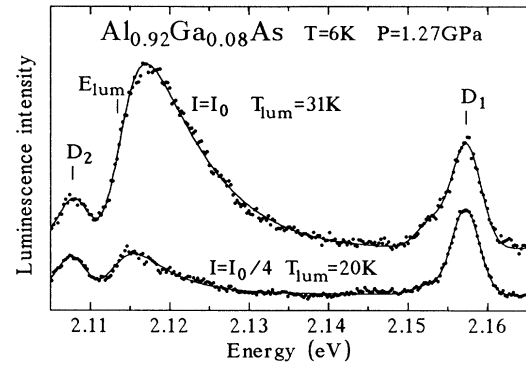


FIG. 2. Photoluminescence spectra for $\text{Al}_{0.92}\text{Ga}_{0.08}\text{As}$ at a pressure of 1.27 GPa and a temperature of 6 K for different laser intensities ($\lambda=413$ nm, $I_0 \approx 10^4$ W cm⁻²).

The line shape of the observed intrinsic luminescence is well described by Eq. (1), as can be seen from Fig. 2. Here, the solid lines represent fits to the experimental spectra, using the luminescence profile of Eq. (1) for the intrinsic luminescence (with the indicated values of T_{lum}) and Gaussian lines for the defect-induced peaks. The energies which were obtained from these fits are shown in Fig. 3. The exciton temperature T_{lum} is higher than the temperature of the diamond cell (6 K), due to two effects, namely, incomplete thermalization of the excitons and the increase of the lattice temperature because of laser heating.

The energy E_{lum} is equal to the exciton energy E_x minus the energy of the phonon involved ($\hbar\omega$), the exciton energy is the difference between the band-gap energy E_g^X and the exciton binding energy R_x .

$$E_{\text{lum}} = E_x - \hbar\omega = E_g^X - R_x - \hbar\omega. \quad (2)$$

Symmetry considerations²⁴ show that in GaAs and AlAs the phonon, which takes part in the phonon-assisted luminescence, is the LO phonon at the X point. For the high aluminum concentrations in our samples only AlAs-like phonons are of importance. To obtain the frequency and pressure dependence of phonons in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ we have also performed Raman measurements under pressure at low temperature. Two spectra for the $x=0.92$ sample are shown in Fig. 4. At a pressure of 2.09 GPa the direct gap is above and at 0.36 GPa below the laser energy ($\lambda=413$ nm $\hat{=} E=3.0$ eV). This results in much smaller absorption of the laser light at the higher pressure, which in turn gives a larger effective volume for the Raman scattering process. Therefore the LO-phonon peak has a much higher intensity in the spectrum at 2.09 GPa compared to the 0.36-GPa spectrum.²⁵ For the $x=0.92$ sample the phonon energies as a function of pressure are given in Fig. 5; results of linear fits for both samples are given in Table I. The pressure coefficients are typical for III-V semiconductors.²⁶ The same applies for the decrease of LO-TO splitting with pressure, which results from a decrease of the effective charge. Raman measurements only give phonon energies at the Γ point. However, in AlAs the LO-phonon dispersion along Γ - X is small²⁷ such that LO phonons at Γ and

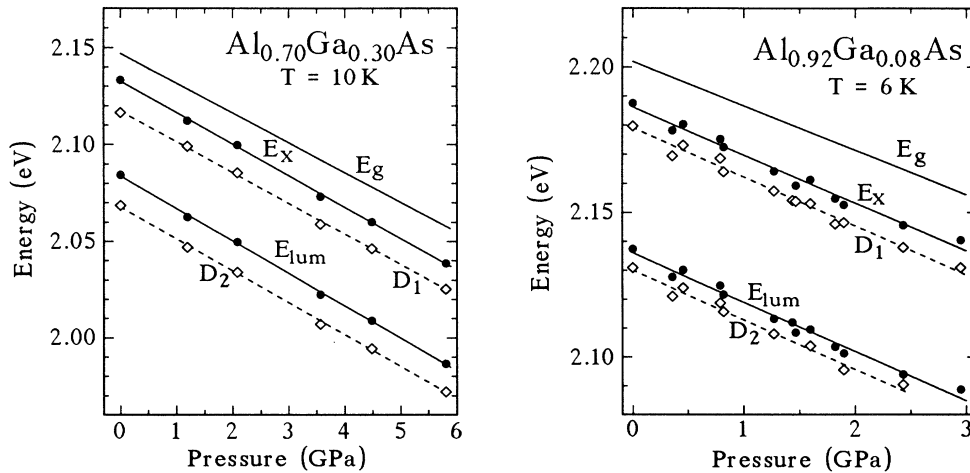


FIG. 3. Indirect gap (E_g), X -point exciton (E_x), intrinsic luminescence energy (E_{lum}), and defect peaks (D_1, D_2) vs pressure for $Al_{0.70}Ga_{0.30}As$ (left) and $Al_{0.92}Ga_{0.08}As$ (right).

X have almost the same energy. This should also hold at higher pressures, since the pressure shift of the X point LO phonons in tetrahedral III-V semiconductors is usually larger than that for Γ -point phonons,^{26,28} which even decreases energy difference between LO(Γ) and LO(X) phonons under pressure.

A second, independent check on the phonon energy is given by the energy difference between D_1 and D_2 , since D_2 is the phonon replica of D_1 . Because D_1 , being a shallow donor, is situated in k space near the X point, the phonon involved is also the LO phonon at the X point. Comparing (see Table I) this difference ($D_1 - D_2$) with the values from Raman scattering ($\hbar\omega$), one finds agreement within the experimental uncertainties. Since the values derived from Raman scattering have much smaller experimental errors, we will use these in the following. The sum of the phonon energy and the luminescence energy gives the energy of the X -point exciton in $Al_xGa_{1-x}As$ (Fig. 3); the results of linear fits and an ex-

trapolation to pure AlAs ($x=1$) are given in Table I.

To obtain the indirect gap from Eq. (2), the last term missing is the exciton binding energy R_x . Since excitons with $n > 1$ were not seen in our measurements, which would have allowed the experimental determination of the exciton binding energy, we have calculated R_x . A hydrogenlike model²⁹ gives

$$R_x = \frac{R_H}{m_0} \frac{\mu}{\epsilon^2}, \quad (3)$$

with R_H the Rydberg energy of hydrogen, m_0 the mass of free electrons, and ϵ the relative dielectric constant. In our case ϵ is equal to the static dielectric constant ϵ_s , since the exciton binding energy is smaller than the LO-phonon energy in all $Al_xGa_{1-x}As$ alloys. The exciton reduced mass μ is given as a function of electron (m_e) and hole (m_h) masses by²⁹

$$\mu^{-1} = m_e^{-1} + m_h^{-1}. \quad (4)$$

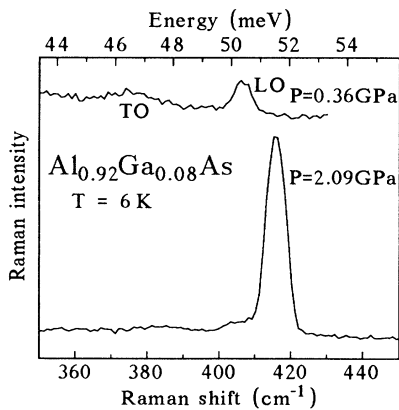


FIG. 4. Raman spectra ($\lambda=413$ nm) for $Al_{0.92}Ga_{0.08}As$ for different pressures.

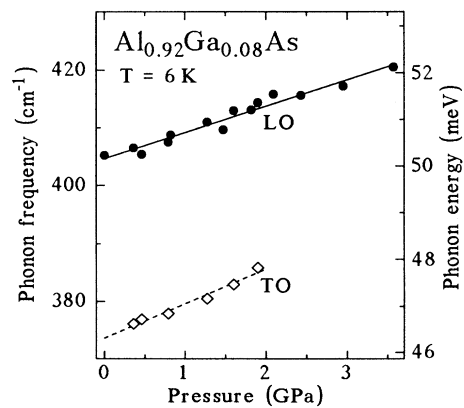


FIG. 5. Pressure dependence of LO and TO phonons in $Al_{0.92}Ga_{0.08}As$.

TABLE I. Intrinsic luminescence (E_{lum}), defect luminescence (D_1, D_2), phonon energies ($\hbar\omega$), X -point excitons (E_X), exciton binding energy [R_x , calculated from Eqs. (3)–(9)], and indirect gap (E_g^X) in $\text{Al}_x\text{Ga}_{1-x}\text{As}$. The values for $x=0.70$ and 0.92 are measured; the values for $x=1$ are obtained from extrapolation. Numbers in parentheses denote the experimental error.

	$x=0.70$		$x=0.92$		$x=1$	
	E (eV)	$\frac{dE}{dP}$ $\left[\frac{\text{meV}}{\text{GPa}} \right]$	E (eV)	$\frac{dE}{dP}$ $\left[\frac{\text{meV}}{\text{GPa}} \right]$	E (eV)	$\frac{dE}{dP}$ $\left[\frac{\text{meV}}{\text{GPa}} \right]$
E_{lum}	2.084(1)	−16.8(5)	2.136(1)	−17.0(5)		
D_1	2.117(1)	−15.9(5)	2.179(1)	−16.9(5)		
D_2	2.068(1)	−16.6(5)	2.130(1)	−17.2(5)		
$D_1 - D_2$	0.049(1)	0.7(5)	0.049(1)	0.3(5)		
$\hbar\omega$ (LO)	0.0488(2)	0.56(5)	0.0502(1)	0.56(4)	0.0507(5)	0.56(5)
$\hbar\omega$ (TO)			0.0464(1)	0.74(7)		
E_X	2.133(1)	−16.2(5)	2.186(1)	−16.5(5)	2.205(7)	−16.6(7)
R_x	0.014	1.2	0.016	1.2	0.017	1.3
E_g^X	2.147(1)	−15.0(5)	2.202(1)	−15.3(5)	2.222(7)	−15.3(7)

Since the conduction band is not spherical at the X point, the electron mass to be used is a combination of longitudinal (m_{\parallel}) and transverse (m_{\perp}) mass:³⁰

$$m_e^{-1} = (m_{\parallel}^{-1} + 2m_{\perp}^{-1})/3. \quad (5)$$

For the valence band one has to take the heavy (m_{hh}) and the light (m_{lh}) hole into account; this results here in³¹

$$m_h^{-1} = (m_{\text{hh}}^{-1} + m_{\text{lh}}^{-1})/2. \quad (6)$$

Following $\mathbf{k}\cdot\mathbf{p}$ theory³² the only pressure-dependent mass in Eqs. (5) and (6) is the light-hole mass m_{lh} with a pressure coefficient of

$$\frac{dm_{\text{lh}}}{dP} = \frac{m_0 m_{\text{lh}} - m_{\text{lh}}^2}{m_0 E_d} \frac{dE_d}{dP}, \quad (7)$$

E_d being the energy of the direct gap. For the pressure dependence of the dielectric constant we use the Lyddane-Sachs-Teller relation

$$\epsilon_s = \epsilon_{\infty} \frac{\omega^2(\text{LO})}{\omega^2(\text{TO})} \quad (8)$$

and the equation by Goñi, Syassen, and Cardona:³³

$$\frac{d\epsilon_{\infty}}{dP} = \frac{5(\epsilon_{\infty} - 1)}{3B_0} (0.9 - f_i). \quad (9)$$

B_0 is the bulk modulus and f_i the ionicity of the material.

From Eqs. (3)–(9) one can now calculate the binding energy of X -point excitons and its pressure dependence. The input parameters needed were taken from Adachi,³⁴ apart from the pressure coefficient for the direct gap, where we used the value^{1,3} for GaAs for all compositions. If necessary, linear interpolation between $x=0$ (GaAs) and $x=1$ (AlAs) was employed. Since the pressure dependence of the exciton binding energy is small, uncertainties in the input parameters (e.g., the pressure shift of the direct gap of AlAs) only slightly affect the final results, which are shown in Table I.

The pressure dependence of the indirect gap of AlAs (−15.3 meV/GPa) is significantly larger than the corresponding value for GaAs (−12.6 meV/GPa, calculated from the measured² pressure dependence of the X -point exciton of −13.4 meV/GPa and the pressure dependence of the exciton binding energy of 0.8 meV/GPa). Although this difference is not very large, it has a significant effect on the pressure dependence of valence-band offsets in superlattices and quantum-well structures, if the standard procedure^{16,17} for the evaluation of the valence-band offset is applied. This makes it necessary to reexamine values for valence-band offsets in GaAs/AlAs superlattices obtained from photoluminescence measurements.

IV. PRESSURE DEPENDENCE OF THE VALENCE-BAND OFFSET IN GaAs/AlAs SUPERLATTICES

The valence-band offset is an important parameter for all types of quantum wells and superlattices. As shown first by Venkateswaran *et al.*,¹⁶ photoluminescence measurements under hydrostatic pressure provide a simple way to measure this parameter. Contrary to first expectations, the valence-band offset is not independent of well width³⁵ and pressure.⁶

Figure 6 shows an energy-band diagram noting the relevant energies in a type-II superlattice. The valence-band offset ΔE_V is given by

$$\Delta E_V = E_g^X(\text{AlAs}) - E_{\text{hh}}^{\text{II}} + \Delta_X + \Delta_{\text{hh}} - R_{\text{SL}}. \quad (10)$$

$E_g^X(\text{AlAs})$ is the indirect gap of AlAs, $E_{\text{hh}}^{\text{II}}$ is the observed photoluminescence energy (zero-phonon line), Δ_{hh} and Δ_X are the confinement energies for a heavy hole and an electron in the X valley, and R_{SL} is the superlattice exciton binding energy. This binding energy is a joint property of electron and hole; in Fig. 6 it is drawn only for convenience in the conduction band.

The Kronig-Penney model³⁶ shows that the confinement energy of a particle in a superlattice depends on its mass, the barrier, and well widths and the well depth. Under pressure the masses of X -point electrons

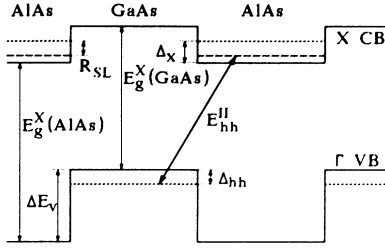


FIG. 6. Band structure of a type-II GaAs/AlAs superlattice. E_{hh}^{II} is the type-II photoluminescence transition, E_g^X is the indirect gap for the respective bulk component, ΔE_V is the valence-band offset, Δ_{hh} and Δ_X are the confinement energies for the heavy hole and the electron in the X valley, and R_X is the exciton binding energy.

and of heavy holes stay constant, as can be seen from $\mathbf{k}\cdot\mathbf{p}$ theory.³² The widths of barrier and well decrease because of hydrostatic compression, thus increasing confinement energies. For the X-point electrons the well depth V_X is given by (see Fig. 6)

$$V_X = E_g^X(\text{GaAs}) - E_g^X(\text{AlAs}) + \Delta E_V. \quad (11)$$

The well depth V_h for the holes is equal to the valence-band offset ΔE_V . Since the indirect gaps of GaAs and AlAs have different pressure coefficients and ΔE_V

TABLE II. Type-II emission energy E_{hh}^{II} and its pressure dependence for several GaAs/AlAs superlattices at low temperatures ($T \leq 80$ K) and the pressure dependence of the valence-band offset ΔE_V , calculated with the new value for the pressure coefficient for the indirect gap of AlAs. m and n are the numbers of monolayers of GaAs and AlAs, respectively, and a and b are the layer thicknesses.

$m \times n$	a, b (nm)	E_{hh}^{II} (eV)	$\frac{dE_{hh}^{II}}{dP}$ ($\frac{\text{meV}}{\text{GPa}}$)	$\frac{d\Delta E_V}{dP}$ ($\frac{\text{meV}}{\text{GPa}}$)
4×4	1.1,1.2	2.012 ^a	-12 ^a	-5.1
6×6	1.5,1.6	1.854 ^a	-17 ^a	1.8
6×6	1.7,1.7	1.935 ^b	-17.0 ^b	1.6
7.2×5.2	2.0,1.5	1.87 ^c	-18 ^c	2.7
10×3	2.8,0.85	1.869 ^d	-17.5 ^d	3.1
8×7	2.3,1.9	1.799 ^a	-18 ^a	2.4
9×9	2.5,2.5	1.805 ^e	-20 ^e	4.2
12×8	3.4,2.3	1.78 ^f	-21 ^f	5.2
11×11	3.1,3.0	1.751 ^a	-20 ^a	4.0
12×12	3.4,3.4	1.761 ^b	-20.1 ^b	4.0
7×22	2.0,6.4	1.824 ^d	-20.9 ^d	5.0
15×15	4.2,4.2	1.701 ^b	-20.4 ^b	4.1
17×17	4.8,5.0	1.694 ^a	-22 ^a	5.7
10.6×24	3.0,6.8	1.75 ^g	-24 ^g	7.9

^aReference 15.

^bReference 13.

^cReference 9.

^dReference 14.

^eReference 15.

^fReference 10.

^gReference 6.

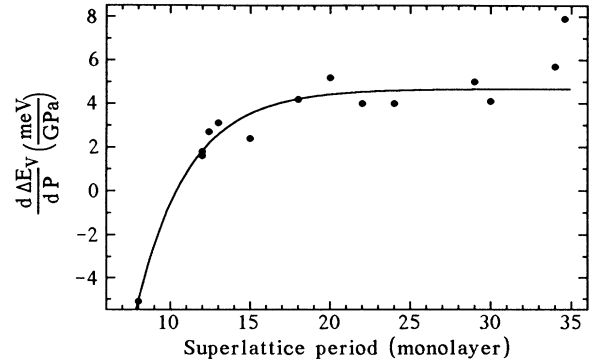


FIG. 7. Pressure coefficient of the valence-band offset in GaAs/AlAs superlattices as a function of the superlattice period. The dots are experimental points (for references see Table II); the solid line is a guide to the eye.

changes with pressure, the well depths V_X and V_h are pressure dependent, too. Using the Kronig-Penney model³⁶ one can now calculate the confinement energies for X-point electrons and heavy holes. Because the confinement energies depend on the well depths and therefore on the valence-band offset, Eq. (10) is an implicit equation for ΔE_V , which can be solved by iteration.

To our knowledge there exist no measurements or calculations³⁷ on the value of the type-II exciton binding energy R_{SL} , so we shall use in the following the pressure dependence of the bulk exciton energy instead, since we expect both binding energies to have the same dependence on masses and dielectric constants.

Table II lists values for the pressure dependence of the type-II photoluminescence energy of GaAs/AlAs superlattice from the literature together with the pressure shifts of the valence-band offset, calculated [Eq. (10)] with the value for the pressure shift of the indirect gap of AlAs as determined in the present article. If one assumes (wrongly), that the indirect gaps of AlAs and GaAs have the same pressure dependence, one gets for the pressure shift of the valence-band offset for a 15×15 superlattice¹³ 7.4 instead of 4.1 meV/GPa. For shorter periods the difference is even larger; for a 6×6 superlattice¹³ one gets 5.1 instead of 1.6 meV/GPa. These large differences show the importance of using precise values for the bulk components in evaluating superlattice properties.

One can see from Table II that the valence-band offset is not only pressure dependent, but that the pressure coefficient also depends on the superlattice structure. Figure 7 clearly shows that there is a correlation between the superlattice period and this pressure shift. The pressure shift increases with increasing period, tending toward a value of 4.7 meV/GPa for long periods.³⁸ For very-short-period superlattices the valence-band offset may even decrease with pressure. The reason for the decrease of the pressure shift of the valence-band offset with decreasing period length is not known. One possibility could be that the concept of a valence-band offset in superlattices with very short periods is not meaningful any more.

V. CONCLUSIONS

We have measured the pressure dependence of the indirect gap in aluminum-rich $\text{Al}_x\text{Ga}_{1-x}\text{As}$ at low temperature. We find that this pressure dependence is markedly larger for AlAs compared to GaAs, so that the pressure dependence of the valence-band offset in GaAs/AlAs superlattices is smaller than previously assumed. Further, this pressure shift shows a dependence on the superlattice

period. For large periods (≥ 20 monolayers) it approaches a constant value of 4.7 meV/GPa, for shorter periods it decreases and may even become negative for very short periods (< 11 monolayers).

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