

Hydrostatic-pressure dependence of band offsets in GaAs/Al_xGa_{1-x}As heterostructures

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We have determined the pressure dependences of the band offsets in the GaAs/Al_xGa_{1-x}As hetero-junctions by measuring the pressure dependences of the harmonic-oscillator-like electron-energy-level spacings $\hbar\omega_c$ of two Al_xGa_{1-x}As parabolic quantum wells at hydrostatic pressures up to ~ 26 kbar at 2 K using photoluminescence-excitation spectroscopy. We found that the conduction-band offset for $x \approx 0.3$ increases with pressure at a rate of 0.73 ± 0.25 meV/kbar. Using this result and the finding that the pressure coefficient of the direct-band-gap energy of Al_xGa_{1-x}As is independent of x for $x = 0-0.4$, we found that the fractional conduction- (valence-) band offset Q_c (Q_v) of GaAs/Al_xGa_{1-x}As increases (decreases) with pressure at a rate of 0.0020 ± 0.0007 kbar⁻¹. From this result, we conclude that the previous determinations of the band-offset values of GaAs/Al_xGa_{1-x}As by high-pressure measurements overestimated (underestimated) the fractional conduction- (valence-) band offset by 0.06 ± 0.02 , and the corrected Q_c value at atmospheric pressure should be 0.63 ± 0.04 .

I. INTRODUCTION

Among the most important parameters for understanding semiconductor heterostructures are the conduction- and valence-band offsets ΔE_c and ΔE_v at the junction between the constituent materials. There have been many efforts to understand the origin of the offsets theoretically and to determine the offset values experimentally. However, theories for the band offsets are not yet capable of predicting the offset values with desired accuracy (e.g., the thermal energy at room temperature, ~ 25 meV), and the experimental determinations have large uncertainties for most heterostructure systems, although there has been some success in determining the band offsets for the GaAs/Al_xGa_{1-x}As system. For this system, there have been numerous measurements of the band offsets using a wide variety of methods, and the majority give values in the range $Q_c = \Delta E_c / \Delta E_g = 0.60-0.65$,¹ where ΔE_g is the difference in the band-gap energies of GaAs and Al_xGa_{1-x}As. However, most of these methods are indirect, involving fits to models.

The band offsets can be determined *directly* by the use of hydrostatic pressure. In this method one exploits the known changes in the band structures of the constituent materials due to pressure, and measures the critical pressure at which the photoluminescence (PL) intensity of a quantum well drops dramatically as the energy of the X states in the barrier (Al_xGa_{1-x}As) drops below the lowest-energy electron level in the quantum well. Using this technique, Venkateswaran *et al.*² and Wolford *et al.*³ obtained $Q_c = 0.70 \pm 0.04$ and 0.69 ± 0.03 , respectively, for GaAs/Al_xGa_{1-x}As ($x \sim 0.3$). However, they made one critical assumption in these determinations, namely that the band offsets are independent of pressure. If this assumption is incorrect, their values represent only

the offset values at the critical pressure, and the correct offset values at atmospheric pressure will be different. Therefore, it is necessary to determine the pressure dependences of the offsets and correct their offset values for these dependences to obtain the correct atmospheric-pressure offset values. Also, the determination of the pressure dependences of the band offsets would provide a compelling test of the band-offset theories as these are refined further to be able to predict the offsets and their pressure dependences with better accuracy, and would give insights leading to a better understanding of the offsets in strained layer superlattices.

There have been several attempts to determine the pressure dependences of the offsets in GaAs/Al_xGa_{1-x}As. In most cases,⁴⁻⁶ the pressure dependences of ΔE_v in type-II superlattices [$x \approx 1$ (Refs. 4 and 5) or in materials with $x \sim 0.3$ at pressures higher than the Γ - X crossover pressure⁶] were determined from those of their PL energies, and small positive pressure dependences (e.g., $\sim 0.5-1.1$ meV/kbar for $x = 1$) of ΔE_v were found. For the type-I case, tunneling-current measurements in GaAs/Al_{0.4}Ga_{0.6}As double-barrier heterostructures at 77 K by Mendez, Calleja, and Wang⁷ suggested that ΔE_c decreases with pressure at a rate as high as 3 meV/kbar, while ΔE_v is pressure independent, which implies that the pressure coefficients of the direct-band-gap energies of GaAs and Al_{0.4}Ga_{0.6}As differ by as much as 3 meV/kbar. They obtained these results by comparing their data with the calculations of the energy of the confined electron level as a function of pressure in the presence of the electric field due to the bias voltage. However, their analysis was complicated by the existence of several different tunneling mechanisms, and their overall pressure range was limited to ~ 8.5 kbar.

Another way to determine the pressure dependences of

the offsets, which can be used up to higher pressures in the type-I regime, is to measure the pressure dependences of the energy-level spacings of quantum wells. Square quantum wells are not very useful here, since for this structure the energy-level spacings are affected by changes in the band offsets only indirectly through their effects on the barrier heights, and the energy-level spacings are fairly insensitive to small fractional changes in these. For parabolic well structures, however, the harmonic energy-level spacings are strongly dependent on the band offsets, since the harmonic-oscillator spacings are directly determined by the parabolic curvatures and these are proportional to the offsets, as shown below.⁸

Parabolic wells are fabricated in such a way to produce parabolic band-edge profiles in the conduction and valence bands, as shown in Fig. 1. At a given pressure, in a simple model where the electrons and the holes are assumed to have constant masses and the parabolic well is assumed to be effectively infinitely deep, the energy levels for electrons and holes are evenly spaced, and the electron-energy-level spacing is given by

$$\hbar\omega_e = \hbar \left[\frac{K_c}{m_e^*} \right]^{1/2} = \hbar \left[\frac{8\Delta E_c}{L_z^2 m_e^*} \right]^{1/2} = \hbar \left[\frac{8Q_c \Delta E_g}{L_z^2 m_e^*} \right]^{1/2}, \quad (1)$$

where K_c is the curvature of the parabolic conduction-band-edge profile, L_z is the width of the parabolic well, ΔE_c is the conduction-band-offset energy between the center and the edge of the well, and m_e^* is an average electron effective mass to be discussed later. The energy-level spacings for the light and heavy holes have similar expressions. This model, despite its apparent simplicity, has been demonstrated⁹⁻¹¹ to quantitatively describe the energies of the parabolic quantum-well states fairly well for energy levels near the bottom of the wells.¹²

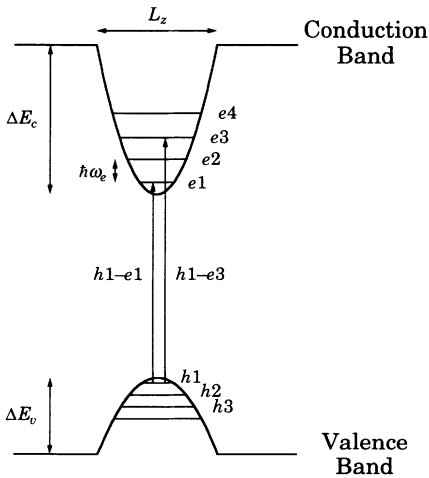


FIG. 1. Schematic of the parabolic band-edge energy profiles in the conduction and valence bands, with the resulting energy levels and several optically allowed transitions. Only the heavy-hole levels are shown for the valence band. Optically allowed transitions occur only between conduction- and valence-band states with quantum numbers differing by an even number.

In principle, Eq. (1) can be used to determine ΔE_c directly from the energy-level spacings obtained from absorption, photoluminescence excitation (PLE), or light-scattering measurements. However, since the uncertainty in L_z due to the molecular-beam epitaxy (MBE) growth variation across the wafer is quite large, it is impractical to determine the offset in this way with the needed accuracy.^{9,11,13} Nevertheless, the pressure dependence of ΔE_c can be obtained from the observed pressure dependence of $\hbar\omega_e$ if ΔE_c for zero pressure is known with reasonable accuracy, since Eq. (1) then “fixes” the value for L_z : the band-offset values determined using the high-pressure technique can be used initially to fit the observed $\hbar\omega_e$ to Eq. (1), using L_z as the fitting parameter. Then the pressure dependence of ΔE_c determined through pressure measurements of $\hbar\omega_e$ can be used to correct ΔE_c for zero pressure, and this corrected offset value can be used to refine the zero-pressure value and the pressure dependence of ΔE_c iteratively.

In principle, the pressure dependence of the valence-band offset ΔE_v also can be determined in this way if the energy-level spacings for the holes can be determined experimentally. In practice, these cannot be obtained directly with high accuracy because, as will be discussed later, the needed light-hole transitions are not usually observed and the energy-level spacings for the heavy holes have large relative uncertainties since the spacings are much smaller than those for the electrons. Instead, since the valence-band offset is given by $\Delta E_v = \Delta E_g - \Delta E_c$, the pressure dependence of the valence-band offset can be determined from that of the conduction-band offset if the pressure dependence of the gap energy difference ΔE_g is known.

In this paper, we report the determinations of the pressure dependences of the band offsets in the GaAs/ $\text{Al}_x\text{Ga}_{1-x}\text{As}$ heterojunctions by measuring the pressure dependences of the energy-level spacings in parabolic wells.

II. EXPERIMENT

Two parabolic well samples with nominal widths $L_z = 1000$ (sample 1) and 500 \AA (sample 2) were measured. The samples were grown on GaAs substrates by molecular-beam epitaxy at the University of California at Santa Barbara, as described by Sundaram *et al.*¹⁴ The parabolic band-edge profiles were generated by alternate deposition of thin layers of GaAs and $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ with varying thicknesses. The relative thicknesses of the $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ layers increased quadratically with distance from the well center, with the total thickness of each pair of GaAs and $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ layers kept constant at w_0 (20 and 10 \AA for samples 1 and 2, respectively). This resulted in an average Al content x varying quadratically from x_0 (≈ 0) at the center to 0.3 at the edges, generating the parabolic band-edge profiles. These sample parameters are summarized in Table I.

The samples were thinned down to thicknesses of $\sim 25\text{--}40 \mu\text{m}$ by chemically etching the GaAs substrates in a 3:1:1 mixture of H_2SO_4 :30% H_2O_2 : H_2O , and then

TABLE I. Growth parameters for the two samples measured.

	L_z	w_0	x_0
Sample 1	1000 Å	20 Å	0.005
Sample 2	500 Å	10 Å	0.02

cleaved to $\sim 125 \times 125 \mu\text{m}^2$ squares. In separate experiments a cleaved piece of each sample was loaded into a cryogenic diamond anvil cell (DAC) (Ref. 15) together with $\sim 20\text{-}\mu\text{m}$ ruby chips. Since this experiment monitored very small changes (~ 1 meV) in the PLE peak spacings, it was extremely important to achieve nearly perfect hydrostaticity inside the pressure chamber of the DAC. To this end, we used He as the pressure medium¹⁶ and warmed up the DAC to room temperature, where He is liquid in the pressure range used,¹⁷ for each pressure change. The pressures were determined with an accuracy of ± 0.3 kbar at 2 K using the luminescence of the ruby chips excited by the 5145-Å line of an Ar⁺ laser. The PLE spectra were taken at 2 K using as the tunable-energy excitation source either a Ti:sapphire laser or a dye laser, scanned by an IBM PC. The illumination power at the sample was ~ 2 mW, focused to a diameter of $\sim 50 \mu\text{m}$. The shapes of the PLE spectra were found to be independent of power up to this level. The luminescence was dispersed by a double monochromator and detected with a cooled GaAs photomultiplier tube. The data were acquired using conventional photon-counting electronics and recorded by the PC. The spectral resolution, limited by the linewidth of the tunable lasers, was $\lesssim 0.1$ meV.

III. RESULTS

A typical PLE spectrum of sample 1, here taken at 7.1 kbar, is shown in Fig. 2. The calculated transition energies are indicated by the vertical lines below, with the identifications of the peaks shown above the peaks using the notation of Fig. 1. Only the transitions with the quantum numbers of the initial and final states differing by an even number are observed, as expected from the parity selection rule. For a given pressure the calculated transition energies include the conduction-band parabolic well energy levels using Eq. (1), the heavy- and light-hole valence-band parabolic well energy levels using similar equations, the exciton binding energies (~ 6.5 meV, approximated by those for a $\sim 250\text{-}\text{Å}$ square well, which has similar zero-point energies¹⁸), and the GaAs band gap, which has a pressure dependence of 10.7 ± 0.1 meV/kbar. (The lowest calculated transition energy is aligned with the lowest-energy peak, by shifting the calculated transition energies rigidly by ~ 3 meV, which is within the uncertainty of the band-gap energy due to the uncertainty in pressure.) The heights of the vertical lines give the calculated relative peak intensities based on the square of the dipole matrix elements, involving the envelope function overlap integrals and the atomiclike dipole matrix elements. The best fit, shown here, required a 4% increase in the nominal L_z , within the uncertainty of this parameter. Comparison of the peak energies and

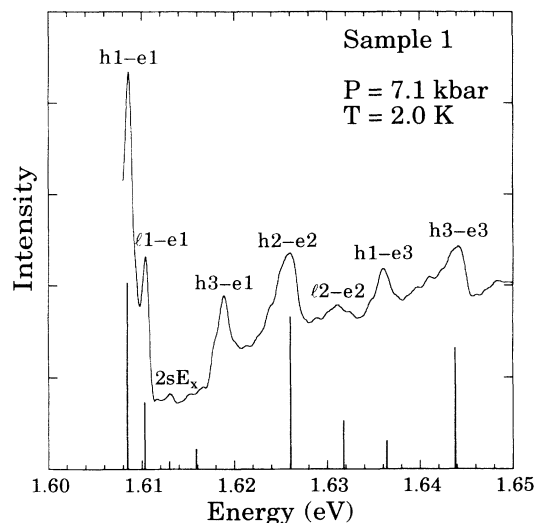


FIG. 2. PLE spectrum of a 1000-Å-wide $\text{Al}_x\text{Ga}_{1-x}\text{As}$ parabolic well (sample 1) at 2.0 K and 7.1 kbar. The vertical lines below give the calculated transition energies, and their heights give the calculated relative intensities. The labels above the peaks give the assignments of the peaks to the transitions using a notation indicated in Fig. 1. The peak labeled $2sE_x$ corresponds to the first excited state of the $h1-e1$ exciton.

intensities with these calculations gives unambiguous assignments of the peaks, except that labeled $h3-e1$. Various other authors have observed that this peak, in both square and parabolic wells, often does not fit the $h3-e1$ transition energy calculated using the parameters that give good fits for other peaks.¹⁹ However, it has been pointed out that peculiarities of the $h3-e1$ exciton transition make the calculation of its energy especially unreliable.²⁰

From the spectrum shown, the electron-energy-level spacing can be determined from the energy difference between the $h1-e3$ and $h1-e1$ peaks by:²¹

$$\hbar\omega_e = \frac{1}{2}(E^{h1-e3} - E^{h1-e1}), \quad (2)$$

where E^{h1-e3} is the energy of the $h1-e3$ peak. The light-hole energy-level spacing cannot be determined directly from this spectrum since such light-hole transitions as $l3-e1$ or $l4-e2$ are not observed. In principle, the heavy-hole energy-level spacings can be determined directly from $E^{h3-e1} - E^{h1-e1}$. However, the narrow spacing in the heavy-hole levels, the difficulty in fitting the $h3-e1$ peak energy, and the uncertainty in the pressure dependence of the heavy-hole effective mass result in large relative uncertainties, making the direct determination of the energy spacing unreliable.

Figure 3 shows the pressure dependence of the electron-energy-level spacing of sample 1, as determined by Eq. (2). The pressure dependence of the conduction-band offset ΔE_c can be determined from these data using Eq. (1) if the pressure dependences of L_z and m_e^* are known. For the pressure dependence of L_z , we assumed a compressibility of 1.325×10^{-3} /kbar for $\text{Al}_x\text{Ga}_{1-x}\text{As}$,²² independent of x . For the pressure dependence of the electron effective mass, we used²³

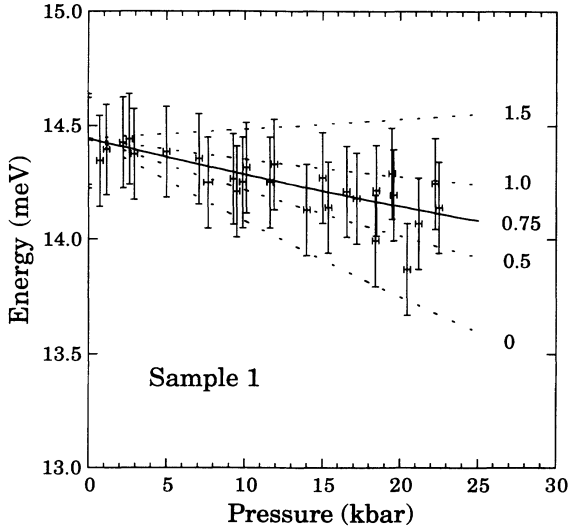


FIG. 3. Pressure dependence of the electron-energy-level spacing of a 1000-Å-wide parabolic well (sample 1), determined from Eq. (2). The curves give the calculated pressure dependences of the energy spacing, using Eq. (1), for various assumed values for the linear pressure coefficient α_c of the conduction-band offset, indicated to the right of the curves. The solid curve represents the best fit.

$$\frac{m_e^*(P)}{m_e^*(0)} = 1 + 6.15 \times 10^{-3} P, \quad (3)$$

where P is the pressure in kbar. $m_e^*(0)$ is an average²⁴ of the x -dependent $\text{Al}_x\text{Ga}_{1-x}\text{As}$ electron effective mass at atmospheric pressure, given by 0.069, only slightly different from the pure GaAs effective mass of 0.067. The value of Q_c used for the initial calculation was 0.69. Using these parameters and assuming a linear pressure dependence of the band offset, $\Delta E_c(P) = \Delta E_c(0) + \alpha_c P$, we obtained a least-squares fit to the data:

$$\alpha_c = \frac{d\Delta E_c}{dP} = 0.75 \pm 0.25 \text{ meV/kbar}. \quad (4)$$

For this value the calculated pressure dependence of the electron-energy-level spacing is given by the solid curve in Fig. 3. For comparison, the calculated pressure dependences of $\hbar\omega_e$ for other assumed values of α_c are shown by the dotted curves. It should be noted that a more rigorous theory for the energy levels than Eq. (1), including band nonparabolicity and the x dependence of the electron effective mass,¹² gives corrections to Eq. (4) much smaller than the error bars. The corrections can therefore be ignored.

The pressure dependence of the conduction-band offset can be written as

$$\frac{d\Delta E_c}{dP} = \frac{d}{dP}(Q_c \Delta E_g) = Q_c \frac{d\Delta E_g}{dP} + \Delta E_g \frac{dQ_c}{dP}. \quad (5)$$

The first term is due to a change in the band-gap energy difference ΔE_g , and the second term due to a relative shift in the band lineups. $d\Delta E_g/dP$ can be obtained from the x dependence of the pressure coefficient dE_g/dP of

the direct-band-gap energy of $\text{Al}_x\text{Ga}_{1-x}\text{As}$: for example, if dE_g/dP is independent of x for the x values of interest, ΔE_g would not change with pressure. Therefore, it is necessary to know the x dependence of dE_g/dP , in order to further analyze the data. To the best of our knowledge, there has been only one systematic set of measurements of dE_g/dP for $\text{Al}_x\text{Ga}_{1-x}\text{As}$ alloys reported in the literature. Lifshitz *et al.*²⁵ obtained dE_g/dP for $\text{Al}_x\text{Ga}_{1-x}\text{As}$ alloys with $x=0-0.5$ for pressures less than 10 kbar by measuring the pressure shift of the iso-transmission energy of optical transmission spectra at room temperature. They found a strongly nonlinear and nonmonotonic x dependence of dE_g/dP , with a cusplike maximum at $x \approx 0.25$. There has been no report of such measurements at low temperatures. However, our recent measurements²⁶ of PL and PLE of 200-Å $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{AlAs}$ square quantum wells at 2 K at pressures up to 30 kbar found that dE_g/dP is independent of x for $x=0-0.4$. This method takes advantage of the high luminescence efficiency of the quantum-well structure and the fact that the pressure coefficient of the lowest-energy transition of a wide (>100 Å) square well is the same as that of the *bulk* band gap of the well material ($\text{Al}_x\text{Ga}_{1-x}\text{As}$, in this case) within the experimental uncertainties. Our result directly contradicts one of the conclusions of the aforementioned tunneling measurements by Mendez, Calleja, and Wang⁷ that dE_g/dP for GaAs and $\text{Al}_{0.4}\text{Ga}_{0.6}\text{As}$ differ by as much as 3 meV/kbar, which amounts to about 25% of dE_g/dP for GaAs (10.7 meV/kbar). It is possible that this contradiction is a result of large error bars in their determinations of $d\Delta E_v/dP$ and $d\Delta E_c/dP$ due to the limited pressure range or the complications in their analysis.

Using our result of the x independence of dE_g/dP , we

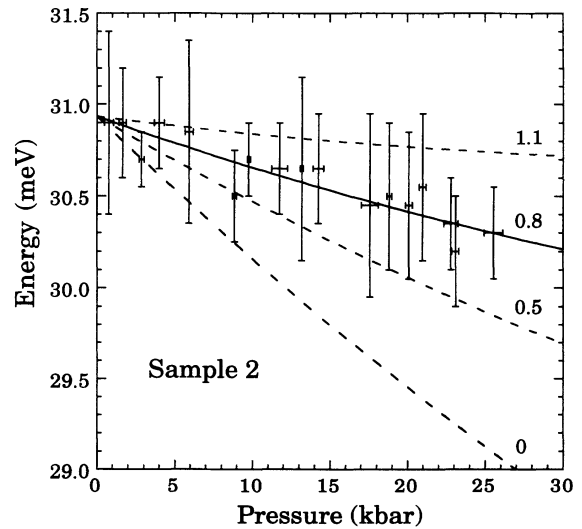


FIG. 4. Pressure dependence of the electron-energy-level spacing of a 500-Å-wide parabolic well (sample 2). The curves give the calculated pressure dependences of the energy spacing, for various assumed values for the linear pressure coefficient of the conduction-band offset, indicated to the right. The solid curve represents the best fit.

obtain $d\Delta E_g/dP=0$, as discussed above. Then from Eq. (5), we obtain

$$\frac{dQ_c}{dP} = \frac{1}{\Delta E_g} \frac{d\Delta E_c}{dP} = 0.0020 \pm 0.0007 \text{ kbar}^{-1}. \quad (6)$$

These results were confirmed by measurements of sample 2, which has energy-level spacings that are twice as large. Figure 4 shows the pressure dependence of $\hbar\omega_e$ for this sample, with curves indicating the calculated pressure dependences of ΔE_c for several assumed values of α_c . The value of α_c obtained by a least-squares fit was 0.8 ± 0.3 meV/kbar, corresponding to $dQ_c/dP = 0.0023 \pm 0.0009 \text{ kbar}^{-1}$, which is well within the error bars of the result for sample 1.

IV. DISCUSSION

Since the results for the two samples are virtually identical, we will use the result for sample 1, which has smaller error bars, for discussion. Using Eq. (6), we can correct the previous determinations of the band offsets using the high-pressure technique. For example, Wolford *et al.* obtained $Q_c = 0.69$ for a GaAs/Al_{0.28}Ga_{0.72}As sample by observing a Γ - X crossover at ~ 30 kbar. Our result indicates that this determination overestimated the conduction-band offset at atmospheric pressure by $\sim 0.0020 \text{ kbar}^{-1} \times 30 \text{ kbar} \approx 0.06$. Therefore, the corrected conduction-band offset value is $Q_c \approx 0.69 - 0.06 = 0.63$. This Q_c value for atmospheric pressure can be used to further refine our fitting of the data. When $Q_c = 0.63$ was used in fitting the data to Eq. (1), we obtained $\alpha_c = 0.73$ meV/kbar, virtually the same as the initial fit. This indicates that the choice of the initial Q_c value is not critical in determining the *pressure dependence* of Q_c , and the iterative procedure described earlier is unnecessary.

Our corrected Q_c value 0.63 is more in line with the values 0.60–0.65 obtained in the majority of other determinations by less direct methods. In comparison, if we use the results for type-II GaAs/Al_xGa_{1-x}As superlattices, e.g., $d\Delta E_v/dP \approx 1.1x$ meV/kbar for $x \sim 1$ obtained by Lambkin *et al.*,⁴ the corrected zero-pressure Q_c would be ~ 0.71 . It is conceivable that the difference between these two results is due to differences in the materials properties of Al_xGa_{1-x}As for different x : for example, the slight biaxial strain due to the lattice mismatch between GaAs and Al_xGa_{1-x}As, albeit small, may affect $d\Delta E_v/dP$ differently for different x . If this is the case, it would be inappropriate to use the result for GaAs/AlAs to obtain the correct zero-pressure offsets for Al_xGa_{1-x}As ($x \sim 0.3$).

It should be pointed out that we have determined dQ_c/dP rigorously only for the range of $x \lesssim 0.2$. Since the $n = 3$ electron wave function for sample 1 (sample 2) extends only ~ 600 Å (~ 400 Å), corresponding to

$x \lesssim 0.11$ ($x \lesssim 0.2$), our measurements probe the curvatures of the parabolic wells only for this range. The above application of this result to the correction of the band-offset values is based on an assumption that our result also applies to x values out to $x = 0.3$, which we think is reasonable in light of the fact that the pressure coefficient of the band gap is also independent of x for that range.

In principle, our rigorous result for $x \lesssim 0.2$ can be used to test the existing theories on the band offsets. While several theories may predict correctly the band offsets at atmospheric pressure, a valid theory should also be able to predict the effects of pressure on these offsets if the pressure-induced changes in the constituent materials are appropriately included. Unfortunately, the existing theories are not accurate enough to calculate such small pressure coefficients of the offsets.²⁷ However, as the accuracies of these theories are further improved, the pressure dependences we have found will provide a powerful test of their validities.

V. CONCLUSIONS

By measuring the harmonic-oscillator-like electron-energy-level spacings $\hbar\omega_e$ of two Al_xGa_{1-x}As parabolic quantum wells at pressures up to ~ 26 kbar at 2 K using PLE spectroscopy, we have determined that the conduction-band offset ΔE_c of the GaAs/Al_xGa_{1-x}As heterojunctions has a pressure dependence of 0.73 ± 0.25 meV/kbar for $x \approx 0.3$. Using this result and the finding that the pressure coefficient of the direct-band-gap energy of Al_xGa_{1-x}As is independent of x , we have found that the fractional conduction- (valence-) band offset Q_c (Q_v) of GaAs/Al_xGa_{1-x}As increases (decreases) with pressure by about $0.0020 \pm 0.0007 \text{ kbar}^{-1}$. From this result, we conclude that the previous determinations of the band-offset values of GaAs/Al_xGa_{1-x}As by high-pressure measurements overestimated (underestimated) the fractional conduction- (valence-) band offset by 0.06 ± 0.02 . The corrected Q_c value at atmospheric pressure should be 0.63 ± 0.04 , which is in line with 0.60–0.65, obtained in the majority of other determinations by less direct methods. Further, the pressure dependences of the offsets obtained in these measurements provide a compelling test of the validity of any band-offset theory.

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