

Direct determination of the band discontinuities in $\text{In}_x\text{Ga}_{1-x}\text{P}/\text{In}_y\text{Al}_{1-y}\text{P}$ multiple quantum wells

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The band structure of $\text{In}_{0.53}\text{Ga}_{0.47}\text{P}/\text{In}_{0.50}\text{Al}_{0.50}\text{P}$ multiple quantum wells grown by molecular-beam epitaxy has been determined from pressure-dependent-photoluminescence measurements at low temperature. The photoluminescence signals from the direct-gap well and the indirect barrier were monitored as a function of pressure up to 4 GPa. High pressure transformed the multiple quantum well from a type I to a staggered aligned, type II at 1.1 GPa. This transition was evidenced by the appearance of a photoluminescence signal due to the recombination of carriers separated in momentum and space. The simultaneous detection of this transition and that of the barrier material, allowed the direct determination of a valence-band offset energy of $(0.24 \pm 0.05)\text{eV}$, without requiring any information on parameters of the bulk materials. Considering that the total band-gap discontinuity for this heterostructure system is 0.50 eV at 20 K, an approximate band-gap splitting of 52:48 is determined to be the band lineup at the $\text{In}_x\text{Ga}_{1-x}\text{P}/\text{In}_y\text{Al}_{1-y}\text{P}$ interface. Variations in the pressure coefficients of the indirect transitions in the barrier indicated that the valence band alignment changes with pressure at a rate of ≈ 18 meV/GPa, due to shifting of the heavy- and light-hole states with biaxial strain induced in the epilayers by applying pressure.

I. INTRODUCTION

The possibility of using quantum confinement to vary the effective band gap of $\text{In}_x\text{Ga}_{1-x}\text{P}/\text{In}_y\text{Al}_{1-y}\text{P}$ heterostructures in the wavelength range 540–650 nm makes this system suitable for the engineering of optoelectronic devices which operate in the visible region of the optical spectrum. $\text{In}_x\text{Ga}_{1-x}\text{P}$ heterostructures have been used as the wide-band-gap component of edge-emitting¹ and vertical-cavity-surface-emitting semiconductor lasers² and have also been employed in the design of heterojunction bipolar transistors.³ In spite of the broad application of this material, basic knowledge of fundamental parameters such as the band discontinuities which are required for the design of efficient devices are still unknown.

We report the first, to our knowledge, direct determination of the band discontinuities in $\text{In}_x\text{Ga}_{1-x}\text{P}/\text{In}_y\text{Al}_{1-y}\text{P}$ multiple quantum wells (MQW's) from low temperature photoluminescence (PL) measurements performed at high pressure. The valence band offset of this heterostructure was directly deduced from the PL signals corresponding to different transitions within the well and barrier obtained by excitation with three different lines of an argon ion laser.

High pressure photoluminescence measurements offer the possibility of determining the band-gap discontinuities in multiple quantum well structures which show a type-I alignment for the direct gap and a type-II alignment for the indirect X minima.⁴ This method uses high pressure to modify the band structure of the heterostructure materials to obtain a band alignment in which the lowest conduction band state and the highest valence band state are separated in space. Under this condition

indirect recombination of the spatially separated carriers is possible. Monitoring the variation with pressure of this indirect transition in addition to the recombination of carriers in the barrier material allows the determination of the energy of the barrier X minima at atmospheric pressure with respect to the well and barrier valence band, respectively. The valence band offset can then be directly calculated as the energy difference between these two transitions. The main advantage of the high pressure technique is that it does not require any assumptions concerning the bulk parameters nor does it require theoretical modeling or a special configuration of the sample as needed in some of the other techniques which have been used to determine the band offsets.^{5–10} Using this technique the band alignment of several heterostructure systems such as $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$,⁴ $\text{GaAs}/\text{In}_x\text{Ga}_{1-x}\text{P}$,¹¹ and more recently $\text{GaAs}/\text{Ga}_x\text{As}_{1-x}\text{P}$ (Ref. 12) have been determined.

The rest of this paper is organized as follows: in Sec. II the sample characteristics and the high pressure photoluminescence experiments are described, the results are presented and discussed in Sec. III and Sec. IV summarizes the main findings of this work.

II. EXPERIMENTAL DETAILS

The $\text{In}_{0.53}\text{Ga}_{0.47}\text{P}/\text{In}_{0.50}\text{Al}_{0.50}\text{P}$ MQW samples were grown by gas-source molecular-beam epitaxy on a semi-insulating (100) GaAs substrate. The MQW's were composed of 30 periods of 8.5 nm wells and 22.5 nm barriers. An In-Al-P capping layer of 210 nm was also deposited. The samples were unintentionally n -doped with a background concentration of 10^{16} cm^{-3} . Transmission elec-

tron microscopy, and photoluminescence measurements indicated that the $\text{In}_x\text{Ga}_{1-x}\text{P}$ material is mostly disordered. A detailed description of the growth and characterization of these samples is given in Ref. 13.

For the high pressure photoluminescence measurements, the GaAs substrate was removed by chemical etching and a small portion ($100 \times 150 \mu\text{m}$) of the epilayer was placed in the gasketed diamond anvil cell (DAC).¹⁴ A small piece of ruby was positioned next to the sample for the measurement of pressure, and a second ruby chip placed in the DAC body outside the pressure chamber was used as the zero pressure reference.¹⁴ A sample of bulk $\text{In}_x\text{Ga}_{1-x}\text{P}$ was also placed outside the pressure chamber for the measurement of the band gap of this material. Argon was used as the pressure transmitting medium. The choice of this pressure medium along with the reduced thickness of the sample were required to guarantee quasihydrostatic conditions at low temperature and at the highest pressure of 4 GPa achieved in these experiments. The DAC was cryogenically cooled to 20 K with a close-cycle He refrigerator. Using a specially designed cryostat chamber the pressure in the cell was changed while still at low temperature. Our ability to vary the pressure at low temperature avoided any hysteresis problems which may be encountered in systems requiring room temperature pressurization subsequent to cooling and measuring.

The PL from the multiple quantum wells was excited using three different lines from an Ar^+ laser, 514, 488, and 458 nm, each with a constant power of 3.5 mW. This variation in the wavelength allowed the excitation of different transitions in the well and barrier materials. The PL was dispersed using a 0.75 m spectrometer equipped with a 1200 g/mm grating and detected by a thermoelectrically cooled photomultiplier tube. Standard chopping techniques and a lock-in amplifier were used to analyze the signal.

III. RESULTS AND DISCUSSION

Typical PL spectra at different pressures obtained by excitation with the different Ar^+ lines are shown in Fig. 1. Curves *a*, *b*, and *c* were obtained with an excitation energy of 2.41 eV (514 nm). At low pressures the main PL feature, labeled E_1 , corresponds to recombination in the well involving the $n = 1$ lowest confined conduction band state and the 1hh heavy hole valence band state. For pressures higher than 1.1 GPa a second much weaker and broader peak (E_2) appears at an energy of 2.09 eV. For pressures higher than 2.3 GPa another transition of characteristics similar to those of the E_2 peak is detected at an energy of 2.2 eV. This peak was labeled E_3 . Both E_2 and E_3 were resolvable even at the highest pressure of 4 GPa. Excitation of the carriers with 2.54 eV (488 nm) photons resulted in PL spectra (not shown) with the same features as those shown in curves *a*–*c*.

When the PL was excited with 2.7 eV (458 nm) photons an additional broad peak at 2.35 eV is also observed. This peak, which was labeled E_4 , is shown in Fig. 1, curve *d*, along with the E_1 peak and a much weaker peak

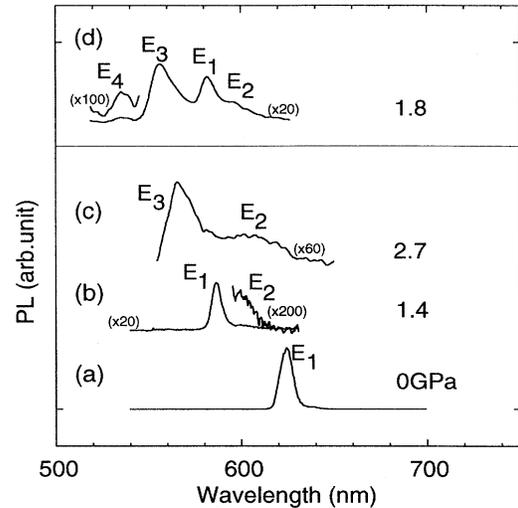


FIG. 1. Typical PL spectra of $\text{In}_x\text{Ga}_{1-x}\text{P}/\text{In}_y\text{Al}_{1-y}\text{P}$ multiple quantum wells at different pressures at 20 K. Curves *a*–*c* were obtained with excitation of the carriers with a photon energy of 2.41 eV (514 nm) and curve *d* was obtained with an excitation of 2.7 eV (458 nm) photons.

at 2.27 eV which corresponds to the E_3 transition. For the highest excitation energy of 2.7 eV, it is possible to excite carriers in the well with sufficient excess energy to scatter to the indirect X level as well as to excite carriers within the barrier. The unique identification of these PL transitions, as shown below, requires the knowledge of the corresponding pressure coefficients.

A weak but resolvable peak at 1.94 eV is also observed in the PL spectra near zero pressure, for all three different excitation energies. Olsthoorn *et al.*¹⁵ have speculated on the nature of this transition in $\text{In}_x\text{Ga}_{1-x}\text{P}$ alloys as an isolated center whose PL was both temperature and excitation independent. The fact that this transition is only observable at very low pressures suggests that this transition is possible only when the $n = 1$ level is resonant with the center. Regardless, the origin of this PL peak remains speculative.

The measured energy variation with pressure of the $E_1 - E_4$ transitions is shown in Fig. 2. The energy of the E_1 transition shows the characteristic direct gap behavior, increasing with pressure at a rate of (92 ± 3) meV/GPa. The remaining transitions, on the other hand, show negative pressure coefficients, a behavior which is characteristic of an X -like band. The highest energy transition E_4 is identified as that corresponding to recombination between electrons in the barrier X level (X_b) and holes in the barrier valence band. The atmospheric pressure value of the energy of these minima agrees very well with previous determination of the band structure of bulk $\text{In}_y\text{Al}_{1-y}\text{P}$.¹⁶ At about 1.1 GPa the onset of the first direct-to-indirect crossover ($E_1 - E_2$) occurs. The transition E_2 is identified as that involving states from the X_b level in the barrier and the 1hh valence band states in the well. This PL is the result of the recombination of carriers which are separated in momen-

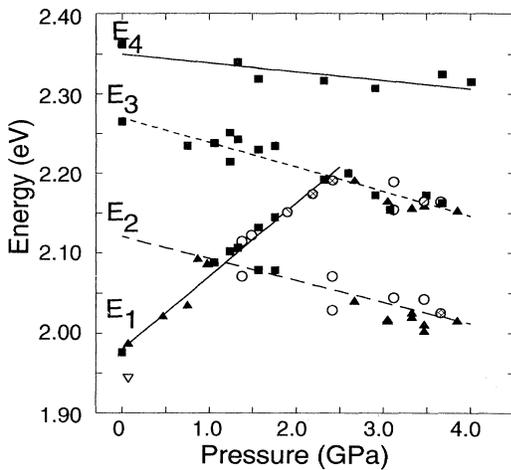


FIG. 2. Variation with pressure of the measured PL transition energies from $\text{In}_{0.53}\text{Ga}_{0.47}\text{P}/\text{In}_{0.5}\text{Al}_{0.5}\text{P}$ multiple quantum wells. Results of different excitations are indicated with different symbols: square, 458 nm; circle, 488 nm; and triangle, 514.5 nm. Four energy bands are clearly identified. The lines correspond to the least squares fit of the data from which the pressure coefficient and the energy at atmospheric pressure of each of the transitions were obtained.

tum and space, that is, at this pressure the type-I MQW is transformed into a type-II staggered aligned system. A second direct-to-indirect crossover ($E_1 - E_3$) is observed at 2.3 GPa. This crossover, which is accompanied by the quenching of the E_1 PL signal, is assigned to the crossing of the $n = 1$ level and the well X minima (X_w). The corresponding crossover in bulk $\text{In}_x\text{Ga}_{1-x}\text{P}$ at low temperatures occurs at 2.6 GPa.¹⁷ The lower transition pressure of this direct-to-indirect crossover in the MQW is a consequence of the larger pressure coefficient of the direct gap of $\text{In}_x\text{Ga}_{1-x}\text{P}$ in a confined structure when compared with bulk material.¹⁷ Thus the transition E_3 is assigned to recombination of carriers from the X_w minima into the well valence band. Figure 3 schematically shows the different PL transitions observed in $\text{In}_x\text{Ga}_{1-x}\text{P}/\text{In}_y\text{Al}_{1-y}\text{P}$ multiple quantum wells in these experiments.

Corroborative evidence of the $E_1 - E_2$ direct-to-indirect crossover was obtained by monitoring the PL intensity as a function of pressure. A sharp reduction in the E_1 PL peak intensity is observed at 1.1 GPa, as shown in Fig. 4. This behavior in the PL intensity is typical of a direct-to-indirect crossover and has been previously observed in other III-V materials.^{18,19} Also shown in this figure is the PL peak intensity corresponding to the E_2 transition, which is approximately constant in the pressure range of these measurements.

Another feature which establishes our assignment of the E_2 transition was obtained by monitoring the dependence of the PL peak intensity of the indirect transitions on excitation power. Figure 5 shows the excitation power dependent PL at 2.4 and 3.12 GPa for the two indirect transitions E_2 and E_3 . It is clear that the PL peak intensity from the indirect transition E_3 within the well is constant with excitation power; however, the indirect

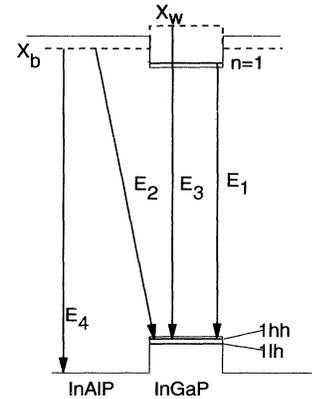


FIG. 3. Schematic diagram of the band structure of $\text{In}_x\text{Ga}_{1-x}\text{P}/\text{In}_y\text{Al}_{1-y}\text{P}$ MQW's showing the PL transitions observed at 20 K at different pressures. The valence band offset can be directly determined from E_2 and E_4 with a small correction due to the heavy hole band confinement energy.

transition E_2 from the barrier into the well is excitation power dependent. The observed blueshift in the PL peak intensity of this transition is attributed to band bending due to an electric field created at the interface by the spatially separated carriers and has been previously observed in the GaAs/AlAs quantum well system²⁰ and in GaAs/ $\text{In}_x\text{Ga}_{1-x}\text{P}$ MQW's at high pressure.²¹ The absence of any redshift in the peak intensity of the E_3 transition indicates negligible lattice heating.

The pressure coefficients of the different transitions were obtained from the slope of the least square fit to the data. The y intercept of these fits yielded the energy of the different electronic levels in the well and barrier at at-

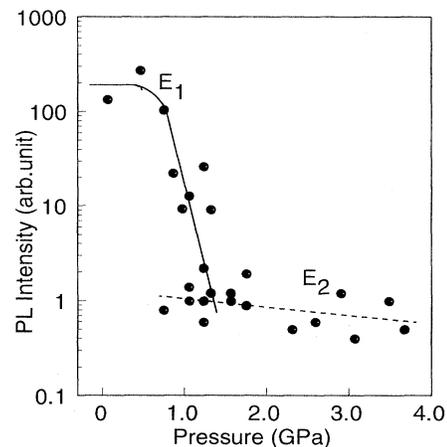


FIG. 4. Pressure dependence of the PL peak intensity from the main direct transition E_1 and that corresponding to the indirect transition E_2 between the barrier and the well. A two-orders-of-magnitude decrease in the peak intensity is observed at about 1 GPa due to the direct-to-indirect transitions between the barrier and the well. Lines through the data are only a guide to the eye.

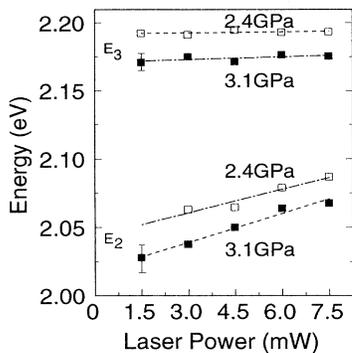


FIG. 5. Excitation power dependent PL at 2.4 and 3.1 GPa. The E_2 PL peak energy from the barrier is blueshifted with excitation power whereas the indirect transition within the well (E_3) is relatively constant with power. The blueshift is a consequence of band bending due to the electric field created by the spatially separated carriers.

mospheric pressure. These parameters and the crossover pressures of the two direct-to-indirect transitions have been summarized in Table I.

Relevant information on the variation with pressure of the valence band offset can be obtained by comparing the pressure coefficients of the E_2 and E_4 transitions. Since both transitions originate at the same level X_b , their pressure coefficient should be the same. However, our results show that the E_2 transition decreases with pressure at about twice the rate of the E_4 transition. This difference in the pressure coefficients indicates that either the 1hh valence band state or the barrier valence band are changing with pressure, possibly due to the presence of biaxial strain.

In two-dimensional systems the effect of hydrostatic pressure results in the generation of a biaxial strain in the growth plane due to the difference in the elastic constants of the well and barrier materials.²² This biaxial strain is responsible for the shifting of the heavy and light hole bands and the splitting of the indirect conduction band minima.²³ We calculated the pressure induced strain for our sample using elastic theory and the corresponding shift of the heavy and light hole bands.²⁴ The elastic constants of both ternary materials were interpolated from the binary constituents²⁵ and were assumed to be constant in the pressure range of these experiments. These calculations show that as pressure is increased a biaxial tensile strain is induced in the $\text{In}_x\text{Ga}_{1-x}\text{P}$ well and a compressive strain, less than half the value of that in the well, is generated in the $\text{In}_y\text{Al}_{1-y}\text{P}$ barrier. This

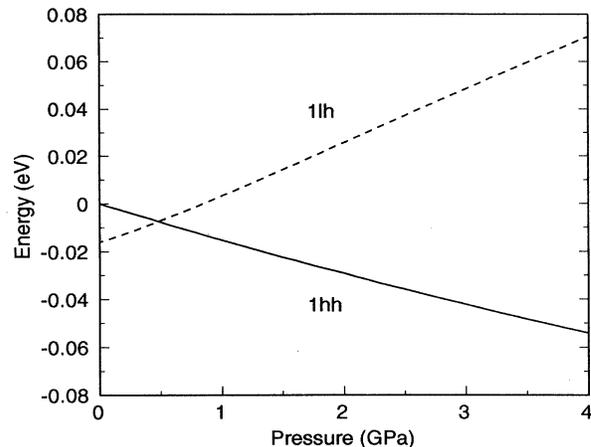


FIG. 6. Energy shift of the heavy and light hole bands in $\text{In}_x\text{Ga}_{1-x}\text{P}$ as a function of pressure. The shifts are calculated with respect to the average valence band energy using the solid-model approach (Ref. 24). Both bands were assumed to be separated by 16 meV at atmospheric pressure.

difference in the magnitude of the strain in the well and barrier indicates that the changes in the band alignment are mostly due to the variation with pressure of the well valence band states. The corresponding shift in energy with pressure of the heavy and light hole well valence band states is shown in Fig. 6. The heavy hole band shifts with pressure at a rate of -13.5 meV/GPa while the light hole energy increases with pressure at a rate of $+21.5$ meV/GPa. Taking into account a separation of 16 meV between the heavy and light hole band as calculated from the envelope-approximation,²⁶ it is found that the two bands cross at 0.5 GPa. Therefore, in almost the entire pressure range of these experiments the light hole band is the top valence band state. Since this band has a positive pressure coefficient and the indirect transition E_2 has a negative pressure coefficient, then the measured rate is larger than that of the E_4 transition. A pressure rate for the light hole band of 18 meV/GPa is obtained from the difference between the measured pressure rates of the E_2 and E_4 transitions, in good agreement with the rate obtained in the calculations. The shifting of the light hole band is also responsible for the increased pressure coefficient of the E_3 transition. The lower crossover pressure of this transition when compared to bulk¹⁷ is due to the splitting of the X_b minima with strain. In calculating the splitting of the well valence band states with strain we have ignored the variation in the confinement energies due to the change of the effective masses

TABLE I. Zero pressure energy band position, pressure coefficient, and crossover pressure of the different transitions observed in $\text{In}_x\text{Ga}_{1-x}\text{P}/\text{In}_y\text{Al}_{1-y}\text{P}$ multiple quantum wells.

	E (eV) [at $P = 0$ GPa]	dE_g/dP (meV/GPa)	Transition pressure (GPa)
In-Ga-P (E_1)	1.984 ± 0.004	92 ± 3	
In-Ga-P/In-Al-P (E_2)	2.12 ± 0.01	-28 ± 5	1.1 ± 0.2 ($E_1 - E_2$)
In-Ga-P (E_3)	2.272 ± 0.007	-31 ± 3	2.3 ± 0.5 ($E_1 - E_3$)
In-Al-P (E_4)	2.35 ± 0.01	-10 ± 4	

with pressure and that due to the change in the well width with pressure. These corrections are very small, have opposite pressure coefficients, and therefore their effect on the confinement energy change with pressure is negligible. From our experimental results and calculations we find that in the $\text{In}_x\text{Ga}_{1-x}\text{P}/\text{In}_y\text{Al}_{1-y}\text{P}$ system the valence band alignment cannot be assumed to remain constant with pressure as has been the case in the $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ system.⁴ While this variation in energy of the valence band states modifies the pressure coefficients of the PL transitions, it does not alter the energy difference between the conduction and valence band states at atmospheric pressure.

The determination of the valence band offset of the $\text{In}_x\text{Ga}_{1-x}\text{P}/\text{In}_y\text{Al}_{1-y}\text{P}$ heterointerface is straightforward knowing the energy of the different indirect transitions at atmospheric pressure. In reference to Fig. 3, the valence band offset is given by

$$\Delta E_v = E_4 - E_2 + E_{1\text{hh}} \quad (1)$$

where $E_{1\text{hh}}$ is the confinement energy of the heavy hole band. In the envelope approximation, $E_{1\text{hh}}$ is found to be equal to 9 meV when an $\text{In}_x\text{Ga}_{1-x}\text{P}$ hole effective mass of $0.46m_0$ is used.¹¹ With this value of $E_{1\text{hh}}$ a valence band offset of 0.24 eV is obtained from Eq. (1). This value of the band offset is smaller than that predicted by the model solid approach of Van de Walle.²⁴

The calculation of the valence band offset using Eq. (1) assumes that the acceptor concentration is negligible in both the well and barrier materials, since our samples are *n*-type. However, in the presence of acceptor levels in either the well or barrier material, this expression needs to be modified to account for the binding energy of the acceptor level. We have calculated the valence band offset in this case considering two limiting situations, one in which an acceptor level with a binding energy of 50 meV is located in the well material, and the other in which the same acceptor level is located in the barrier. In the first case, the valence band offset is found to be 0.19 eV while in the second case a value of 0.29 eV for the valence band offset is obtained. Including the acceptor binding energy and the experimental uncertainties we find $\Delta E_v = (0.24 \pm 0.05)$ eV. To our knowledge, the only other reported measurement of the band offset energies for the $\text{In-Ga-P}/\text{In-Al-P}$ interface has been made by Watanabe and Ohba using capacitance-voltage ($C - V$) carrier profiling.²⁷ They obtained a value at 300 K of 0.11 eV for ΔE_c for an $\text{In}_{0.5}\text{Ga}_{0.5}\text{P}/\text{In}_{0.5}\text{Al}_{0.5}\text{P}$ heterojunction grown by metal-organic chemical vapor epitaxy. Using a band-gap difference of 0.50 eV, their result indicates ΔE_v of 0.39 eV, considerably higher than our value of $\Delta E_v = (0.24 \pm 0.05)$ eV. The reason for this discrepancy may be the precision of the measurement techniques. The ΔE_c obtained by the $C - V$ method is now known to be strongly influenced by the structure (i.e., interfacial charge, doping, and compositional nonuniformities) and measurement conditions (i.e., frequency, temperature), and thus large errors in ΔE_c can easily occur.²⁸ In fact, Watanabe and Ohba note that a simulated carrier profile based on their ΔE_c value did not fit their measured

profile.²⁷ In comparison, the pressure-dependent PL technique employed here measures energy levels directly and does not require additional knowledge of the bulk or interface properties. Thus, the value of $\Delta E_v = 0.24 \pm 0.05$ eV is free of the errors inherent in the $C - V$ technique.

The conduction band offset can be calculated from the measured value of the valence band offset and the band-gap energy difference of the well and barrier materials. The value of the direct band-gap of bulk $\text{In}_x\text{Ga}_{1-x}\text{P}$ was determined from the maximum of the photoluminescence signal from the bulk sample corrected for the exciton binding energy. A value of 1.95 eV was obtained at 20 K. The band-gap of the barrier materials was taken equal to 2.45 eV from previous measurements on the band structure of In-Al-P .¹⁶ The total band discontinuity for this heterostructure system is calculated to be 0.50 eV at 20 K. Therefore, the conduction band offset energy is 0.26 eV and a 52:48 band-gap splitting is determined to be the band lineup in the $\text{In}_x\text{Ga}_{1-x}\text{P}/\text{In}_y\text{Al}_{1-y}\text{P}$ MQW. A schematic of the band structure diagram of this MQW is shown in Fig. 7. The only assumption made in this band diagram involves the energy of the barrier Γ minimum; however, this assumption does not play a role in determining the valence band offset as discussed above. The band diagram also shows the position of the indirect X levels in the well and barrier materials with respect to the well and barrier valence bands as obtained from the measurements. The 136 meV separation between the barrier X level and the lowest confined conduction band state in the well sets a lower limit for the well width of 16 Å for a type-I structure. This result agrees well with the measurements of Hafich *et al.*,¹³ who observed the quenching of the photoluminescence in $\text{In}_x\text{Ga}_{1-x}\text{P}/\text{In}_y\text{Al}_{1-y}\text{P}$ for well widths lower than 18 Å. Therefore the shortest wavelength that can be achieved with this heterostructure system is 563 nm. Shorter wavelengths would result in a type-II MQW which has been shown to have low radiative efficiency and radiative lifetimes up to several μsec .²⁹ These characteristics along with the low mobility of the carriers associated with the X valley are detrimental in device performance.

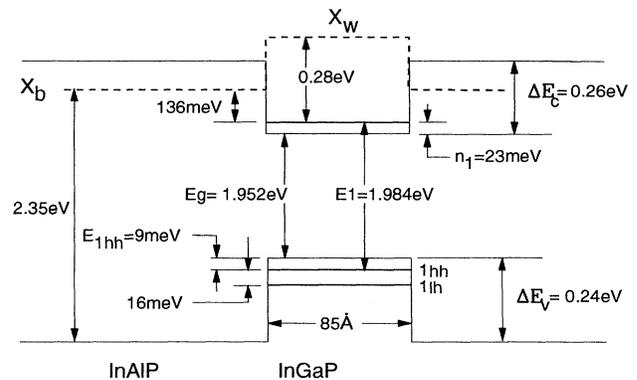


FIG. 7. Energy band diagram of $\text{In}_x\text{Ga}_{1-x}\text{P}/\text{In}_y\text{Al}_{1-y}\text{P}$ at atmospheric pressure and 20 K. The values for the band-gap energies and energies of the barrier and well X minima are all obtained from the experiments.

IV. CONCLUSIONS

The band structure of $\text{In}_{0.5}\text{Ga}_{0.5}\text{P}/\text{In}_{0.5}\text{Al}_{0.5}\text{P}$ multiple quantum wells was determined from low temperature photoluminescence measurements at high pressure. Selective excitation of different transitions in the well and barrier materials allowed, to our knowledge, the first direct determination of the valence band offset at atmospheric pressure. This heterostructure system transformed from a type-I alignment to a type-II staggered-aligned system at 1.1 GPa. This direct-to-indirect crossover was accompanied by a sharp decrease in the intensity of the direct gap photoluminescence signal. The valence band alignment was found to vary with pressure due to pressure induced strains in the epilayers.

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