

High-pressure photoluminescence study of GaAs/GaAs_{1-x}P_x strained multiple quantum wells

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We report low-temperature photoluminescence studies of GaAs/GaAs_{1-x}P_x strained quantum-well samples grown by gas-source molecular-beam epitaxy as a function of pressure. We have found that the transitions between the lowest Γ -confined electron and hole states shift toward higher energy with increasing pressure, and also that the pressure coefficients of the transitions depend on the alloy concentrations and the quantum-well structures. From the observation of the pressure-induced crossover of the lowest Γ -confined electron state in the wells against the conduction-band (001) X minima in the barriers, we are able to determine the valence-band offset for the GaAs/GaAs_{0.68}P_{0.32} heterostructure. At this crossover point, the emission of Γ - Γ transition quenches and the emission with the characteristics of the X minima becomes dominant. We have also observed an emission associated with a deep center which becomes active with pressure above ~ 17 kbar, from the GaAs/GaAs_{0.61}P_{0.39} multiple-quantum-well sample. The pressure dependence of the emission suggests the localized state to be donorlike and resonant above the bottom of the conduction band at ambient pressure.

I. INTRODUCTION

The optical and electronic properties of strained quantum-well (QW) structures have been extensively studied in recent years.¹ These strained structures are of great fundamental interest from both the theoretical and the experimental points of view. There are also many advantages of using strained QW's in device applications. Efficient diode lasers with low lasing threshold and high output power have been fabricated using strained QW's.^{1,2} The strained QW's are also very useful in the fabrications of modulation-doped high-mobility field-effect transistors (MODFET's) with very high cutoff frequency.^{3,4} Currently the GaAs/GaAs_{1-x}P_x QW systems are receiving a great deal of attention since they have wide applications in such diode lasers and MODFET's. These systems are very different from the most widely studied QW structures such as In_xGa_{1-x}As/GaAs and In_xGa_{1-x}AsP/InP, where the well layers are subject to the lattice-mismatch-induced strain. In GaAs/GaAs_{1-x}P_x heterostructures pseudomorphically grown on GaAs substrates, the GaAs_{1-x}P_x alloy barrier layers rather than the GaAs well layers are strained. Although some studies on the optical and electrical properties of GaAs/GaAs_{1-x}P_x QW's have been reported,⁵⁻⁸ these material systems are not well understood. In fact, the conduction- and valence-band offsets at the heterointerface are not available in the literature, despite their crucial importance in device design. The application of hydrostatic pressure is a powerful technique for studying band structures of semiconductors. There have been numerous optical investigations utilizing the effect of different pressure coefficients for different conduction-

band minima to induce the type-I to type-II conversions and to determine the band offsets in some superlattices and quantum-well systems.⁹⁻¹⁹ In this paper, we report the results of low-temperature photoluminescence (PL) studies on three GaAs/GaAs_{1-x}P_x strained multiple-quantum-well (MQW) samples ($x = 0.24, 0.32,$ and 0.39) under hydrostatic pressure using a diamond-anvil cell. We have observed in these samples that the transitions between the lowest Γ -confined electron and hole states shift toward higher energy with increasing pressure and determine their pressure coefficients. The pressure coefficients are found to be significantly smaller than that of the GaAs band gap, and depend on the alloy concentrations and the quantum-well structures. The pressure-induced crossover of the first confined electron state in the GaAs wells against the conduction-band (001) X minima in the GaAs_{1-x}P_x barriers has been observed. From the observation of this crossover, where the emission of Γ - Γ transition quenches and the emission with the characteristics of the X minima takes over, we were able to determine the valence-band offset for the GaAs/GaAs_{0.68}P_{0.32} heterostructure. We have also observed emission arising from a deep center from the GaAs/GaAs_{0.61}P_{0.39} MQW sample. This center becomes active with pressure above ~ 17 kbar. The pressure dependence of the emission suggests that the center is a donorlike localized state and resonant above the bottom of the conduction band at ambient pressure.

II. EXPERIMENTAL DETAILS AND RESULTS

The samples used in this work were fabricated by gas-source molecular-beam epitaxy on an undoped GaAs

buffer layer with a thickness of 1 μm . Semi-insulating (001) GaAs substrates were used. The MQW structures consist of 15 periods of GaAs wells and $\text{GaAs}_{1-x}\text{P}_x$ barriers. Undoped GaAs cap layers of about 400 \AA were grown to cover the multiple quantum wells. The phosphorus concentration and layer thicknesses were determined by computer simulations to x-ray rocking curves.²⁰ Samples approximately $200 \times 200 \mu\text{m}^2$ in size were cut from the wafers which have been thinned to $\leq 25 \mu\text{m}$ by mechanical polishing. Samples were loaded into a gasketed diamond-anvil high-pressure cell by use of the methanol-ethanol liquid mixture as a pressure medium. The pressure cell was attached to a cold finger of a closed-cycle refrigerator and cooled down to 10 K. Photoluminescence measurements were performed on the samples in a backscattering geometry by using the 5145- \AA line of an Ar^+ laser as the excitation source. The PL signal was dispersed by a double-grating spectrometer and detected by a photon-counting system.

Figures 1 and 2 show PL spectra taken from the $\text{GaAs}(76.5 \text{ \AA})/\text{GaAs}_{0.68}\text{P}_{0.32}$ (76 \AA) and $\text{GaAs}(87 \text{ \AA})/\text{GaAs}_{0.61}\text{P}_{0.39}$ (88 \AA) MQW samples at different pressures, respectively. It can be seen from the figures that the PL spectra show typical quantum-well Γ - Γ excitonic emission characteristics in the low-pressure range. The full width at half maximum (FWHM) of the Γ - Γ emission peak is about 6 meV, which indicates the good quality of the strained MQW structures. The strong, sharp luminescence of the Γ - Γ transition shifts toward higher

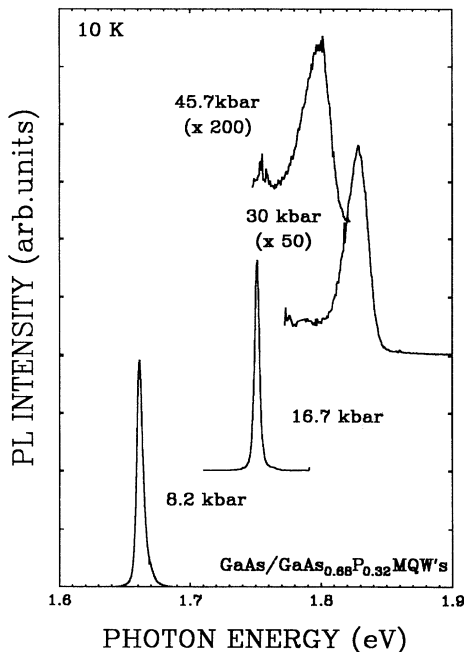


FIG. 1. Photoluminescence spectra (10 K) of the $\text{GaAs}(76.5 \text{ \AA})/\text{GaAs}_{0.68}\text{P}_{0.32}$ (76.5 \AA) MQW's taken at different pressures. The emission from the Γ - Γ confined transition shifts to higher energy with pressure and diminishes around 26 kbar, and the broadened emission from the X - Γ transition takes over and moves down as pressure increases.

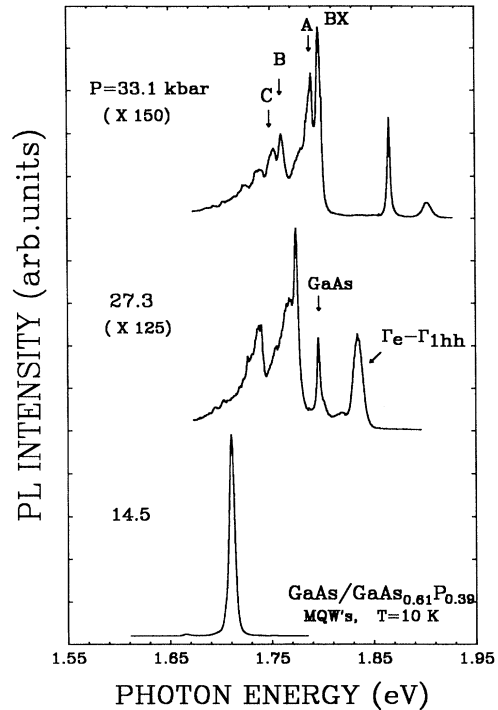


FIG. 2. PL spectra of the $\text{GaAs}(87 \text{ \AA})/\text{GaAs}_{0.61}\text{P}_{0.39}$ (88 \AA) MQW's at different pressures. The emission labeled by BX and its satellite sideband structures marked with A , B , and C appeared after pressure higher than 17 kbar. The emission from the Γ band of the GaAs substrate also could be observed in this sample.

energy with increasing pressure. For the $\text{GaAs}/\text{GaAs}_{0.68}\text{P}_{0.32}$ MQW's, the PL peak starts broadening and the emission intensity decreases sharply at pressures around 26 kbar. At higher pressures, the emission associated with the Γ - Γ transition quenches, and a much weaker and broader emission emerges. This broad peak moves down in energy with pressure, as shown in Fig. 1. It is well known that the conduction band of bulk GaAs at the Γ point rises with pressure with respect to the top of the valence band, and crosses the indirect conduction-band X -point minima, which have a small negative pressure coefficient, at around 40 kbar. The negative pressure dependence of the new peak which we observed is typical of X -like characteristics. The crossover pressure (~ 26 kbar) is much lower than the bulk GaAs Γ - X band crossing and suggests that the applied pressure has caused the lowest confined electron state in the GaAs wells to cross over the X minima in the $\text{GaAs}_{0.68}\text{P}_{0.32}$ barriers and has resulted in a spatially separated indirect transition. A relatively strong emission peak (labeled BX) with a satellite sideband appears as the pressure increases in the $\text{GaAs}/\text{GaAs}_{0.61}\text{P}_{0.39}$ MQW sample, as shown in Fig. 2. The BX emission peak has a sharp high-energy cutoff and shifts to higher energy with pressure at a relatively small rate. However, no emissions with pure X -like characteristics could be observed in this sample, and the $\text{GaAs}(87 \text{ \AA})/\text{GaAs}_{0.76}\text{P}_{0.24}$ (87.5 \AA) MQW sample within the sensi-

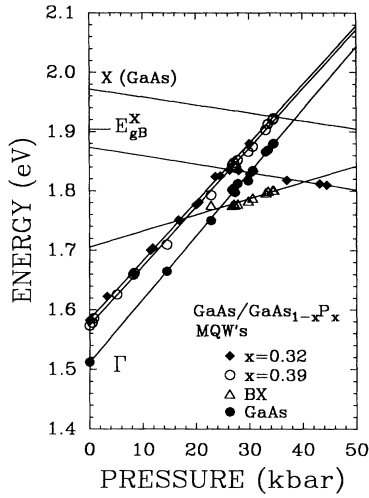


FIG. 3. Energy variations of different emissions observed from GaAs/GaAs_{0.68}P_{0.32} and GaAs/GaAs_{0.61}P_{0.39} MQW's as a function of pressure. The solid lines drawn through the points represent the least-squares fits to the experimental data. Also shown are the pressure dependence of the X conduction-band edges of GaAs. The energy position of the X_z minima of the GaAs_{0.68}P_{0.32} barriers at ambient pressure, as indicated by E_{gB}^X in the figure, is 1.908 eV.

tivity of the detection system used in this work. In Fig. 3, we plot the pressure results collected from the GaAs/GaAs_{0.68}P_{0.32} and GaAs/GaAs_{0.61}P_{0.39} MQW's in conjunction with the dependences of Γ and X conduction-band minima of GaAs on pressure. The onset of the unknown emission structure BX can be derived from the figure to be ~ 17 kbar.

III. ANALYSIS OF RESULTS AND DISCUSSIONS

In the GaAs/GaAs_{1-x}P_x system grown commensurately on GaAs substrates, GaAs_{1-x}P_x layers are sub-

$$E_{\Gamma}(x) = E_{\Gamma}^0(x) + [-2a_{\Gamma}(C_{11} - C_{12})/C_{11} \pm b_{\nu}(C_{11} + 2C_{12})/C_{11}] \epsilon, \quad (1)$$

where the $+$ is for the heavy-hole band and the $-$ for the light-hole band, a_{Γ} is the Γ band-gap hydrostatic deformation potential and b_{ν} is the shear deformation potential for the valence band, C_{ij} are the elastic stiffness constants, and ϵ is the in-plane strain given by $\epsilon = (a_{\text{GaAs}} - a_{\text{GaAs}_{1-x}\text{P}_x})/a_{\text{GaAs}_{1-x}\text{P}_x}$, where a_{GaAs} is the lattice constant of GaAs and $a_{\text{GaAs}_{1-x}\text{P}_x}$ is that for unstrained GaAs_{1-x}P_x alloys. The latter is evaluated by linear interpolation between the lattice constants of GaAs and GaP. The biaxial tension-induced shifts of the X conduction-band minima are given by

$$\Delta E(X_z) = [-2a_x(C_{11} - C_{12})/C_{11} - \frac{2}{3}\Xi_u^x(C_{11} + 2C_{12})/C_{11}] \epsilon \quad ([001]\text{single band}) \quad (2)$$

and

$$\Delta E(X_{xy}) = [-2a_x(C_{11} - C_{12})/C_{11} + \frac{1}{3}\Xi_u^x(C_{11} + 2C_{12})/C_{11}] \epsilon \quad ([100,010]\text{doublet band}), \quad (3)$$

where a_x is the X band-gap hydrostatic deformation potential and Ξ_u^x is the shear deformation potential. This modified band alignment after taking strain effect into account is essential in the determination of the valence-band offset from the experimental data. The alloy depen-

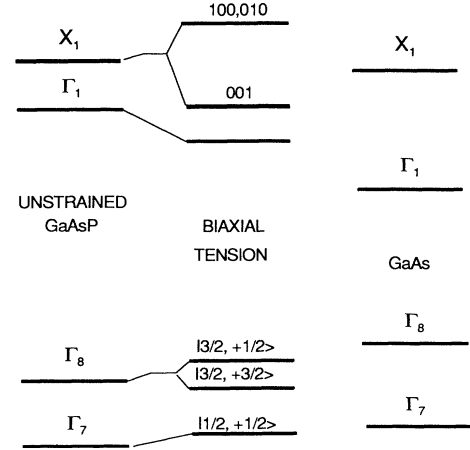


FIG. 4. The schematic diagram of the band edges for the strained GaAs/GaAs_{1-x}P_x heterostructure under the condition of (001) tension.

jected to a biaxial tensile strain due to the larger lattice constant in GaAs. The effect of the built-in biaxial tensile strain on the GaAs_{1-x}P_x band structure at the Γ point is to reduce the direct band gap and lift the degeneracy of the top of the valence band, where the light-hole band lies in energy above the heavy-hole band. The six equivalent X_1 bulk conduction-band minima are split by (001) strain with two equivalent band minima X_z (for k perpendicular to the interface, [001] singlet band) lower than the four equivalent band minima X_{xy} (for k along the interface, [100,010] doublet band). Figure 4 shows the schematical diagram of the band-edge energies for the strained GaAs/GaAs_{1-x}P_x heterostructure under the condition of (001) tension. By using the phenomenological deformation-potential theory,²¹⁻²³ the strain effect on the Γ direct band gap of GaAs_{1-x}P_x can be expressed as

dence of the unstrained band gap of GaAs_{1-x}P_x is formulated as²⁴

$$E_{\Gamma}^0(x) = 1.515 + 1.172x + 0.186x^2 \quad (\Gamma \text{ band}, T = 2 \text{ K}), \quad (4)$$

$$E_x^0(x) = 1.9715 + 0.144x + 0.211x^2$$

(X band, $T = 2$ K). (5)

The confined energy levels in the well are calculated by use of the envelope-function approximation proposed by Bastard.^{25,26} Table I summarizes all the parameters used in the calculations.

As shown in Fig. 3, the pressure-induced crossover of the first confined electron state (Γ_{1e}) in the GaAs wells against the X_z minima in the GaAs_{0.68}P_{0.32} barriers takes place when the applied pressure is higher than 26 kbar. A straightforward determination of the valence-band offset for the multiple quantum wells can be deduced using the formula^{17,19}

$$\Delta E_v = E_{gB}^X - E_{PL} - E_{ex}^b - \Delta E_{1hh}. \quad (6)$$

Here E_{gB}^X is the energy of barrier X_z minima, E_{ex}^b is the exciton binding energy, and ΔE_{1hh} the confinement energy of the lowest heavy-hole state in the wells. Since the pressure coefficient of the X minima of the GaAs_{1-x}P_x alloy is not available at present, we use the values of E_{gB}^X and E_{PL} at ambient pressure for estimating the band offset. Although this method results in some errors in the value of the valence-band offset, it allows us to compare our results with earlier reported values.^{6,8} The energy position of the X_z minima of the GaAs_{0.68}P_{0.32} barriers is calculated to be 1.908 eV at ambient pressure. $E_{PL}(P=0)$ is known from experimental data to be 1.87 eV by extrapolating the X -related transitions back to ambient pressure. The confinement energy of the lowest heavy-hole level (ΔE_{1hh}) is found to be about 12 meV in our calculations. In estimating the exciton binding energy, it must be considered that the binding energies for Γ_{1e} - Γ_{1hh} excitons are about 10 meV and these for the X_z - Γ_{1hh} excitons are even smaller due to spatial separation of the electrons and holes.^{9,27} We therefore take E_{ex}^b to be 5 meV. Using these data, we are able to obtain the band discontinuity between the top of the valence band of the GaAs well and the light-hole valence band of the GaAs_{0.68}P_{0.32} barrier as $\Delta E_v \approx 0.045$ eV, taking into con-

sideration the small Stokes shift (~ 5 meV) detected between the PL emission peak and the PL excitation emission peak. The uncertainty introduced in the calculation of the strain-modified band structure of the GaAs_{0.68}P_{0.32} barrier and the confinement energy of the heavy hole in the GaAs well, as well as in estimating the exciton binding energy, is about 20 meV. As a result, this leads to an unstrained free-standing band lineup with a valence-band offset of 0.09 ± 0.02 eV between GaAs and GaAs_{0.68}P_{0.32}, which is approximately a 75:25 distribution for the energy-gap difference in the conduction and valence bands. The unstrained valence-band offset for GaAs/GaAs_{1-x}P_x was previously presented in Refs. 6 and 8. The unstrained valence-band offset was chosen to be 5 meV per percent phosphorus in Ref. 8, or a linear interpolation with x , within a valence of 600 meV, between GaAs and GaP in Ref. 6. Using these data, one gets an unstrained valence-band offset of ~ 0.16 – 0.19 eV for GaAs/GaAs_{0.68}P_{0.32}. Compared to our result of 0.09 eV, the deviation is rather large. The methods used in Refs. 6 and 8 were to fit photoluminescence excitation (PLE) data by treating the band offset as a parameter or to make an evaluation based on the model-solid theory. Thus the results are indirect and model dependent. The pressure-optical method used in this work is a direct and more accurate measurement without introducing any model-dependent parameters, and has been widely used for the determination of band offsets in a number of III-V compound MQW systems.^{9–11,17,19} It is a direct measurement of the lowest confined state associated with the Γ minimum of the conduction-band edge of GaAs which crosses against the indirect X conduction-band edges of the GaAs_{1-x}P_x barriers. The crossing occurs in a narrow range of energies, and therefore provides more accurate data for deducing the band offset of the MQWs.

The pressure dependences for different PL transition energies observed in each sample can be derived from the experimental data using the best linear-fit function:

$$E_i(P) = E_i(0) + \alpha P. \quad (7)$$

Table II lists the pressure coefficients of all transition energies for all the samples studied in this work, where i denotes the transitions such as Γ - Γ or X - Γ and $\alpha = dE_i/dP$. It is noteworthy that the pressure coefficients of the Γ_{1e} - Γ_{1hh} transitions in all three samples are significantly smaller than that of the Γ point of the GaAs band gap and depend on the phosphorus compositions of barriers and quantum-well structures. Similar results have been reported and discussed in the previous pressure-dependent optical measurements on a variety of III-V compound quantum-well structures.^{10–14,16,17} We note that the PL emission labeled as BX in Fig. 2 for the GaAs/GaAs_{0.61}P_{0.39} MQW's has a small positive pressure coefficient of 2.7 meV/kbar. This pressure dependence does not follow any known conduction-band edge. Since the effect of hydrostatic pressure on acceptor energy levels is negligible, the emission is most likely to be associated with the radiative recombination of excitons bound to donorlike deep centers. This is because the pressure dependence of an energy level of a deep center is

TABLE I. Parameters used in the calculations.

Parameter	GaAs	GaP
a (Å)	5.6533 ^a	5.4505 ^a
C_{11} (10^{11} dyn/cm ²)	12.21 ^a	14.39 ^a
C_{12} (10^{11} dyn/cm ²)	5.7 ^a	6.52 ^a
a (eV)	-8.33 ^b	-8.83 ^b
b_v (eV)	-1.90 ^b	-1.5 ^b
a_X (eV)	1.0 ^b	1.6 ^c
Ξ_u^X (eV)	-8.61 ^a	-6.5 ^c
m_e^* (emu)	0.067 ^a	0.0925 ^a
m_{hh}^* (emu)	0.51 ^a	0.45 ^a
m_{lh}^* (emu)	0.082 ^a	0.12 ^a
γ_1	6.95 ^a	4.05 ^a
γ_2	2.25 ^a	0.49 ^a

^aReference 24.

^bReference 23.

^cReference 22.

TABLE II. Pressure coefficients of various PL transitions in all three samples studied in this work. The pressure coefficients of the Γ and X bands are also listed for comparison. (The units are in meV/kbar.)

Sample no.	$dE_{\Gamma-\Gamma}/dP$	$dE_{X-\Gamma}/dP$	dE_{BX}/dP
GaAs(87 Å)/GaAs _{0.76} P _{0.24} (87.5 Å)	9.5(5)		
GaAs(76.5 Å)/GaAs _{0.68} P _{0.32} (76 Å)	9.9	-1.4(5)	
GaAs(87 Å)/GaAs _{0.61} P _{0.39} (88 Å)	9.9(5)		2.7
GaAs	10.7; 10.73 ^a	-1.34 ^a	

^aD. J. Wolford and J. A. Bradley, Solid State Commun. **53**, 1069 (1985).

mainly determined by the composition of its wave function, whereas the pressure coefficient of a deep donor state is determined mainly by the pressure dependence of the average conduction-band edges of both well and barrier materials.²⁸ The energy separations of the satellite peaks marked by A , B , and C in Fig. 2 relative to the main emission BX peak are about 10, 36.5, and 46.5 meV, respectively, which approximately agrees with the TA^X (9.8 meV), LO^Γ (36.2 meV), and $LO^\Gamma + TA^X$ phonons of GaAs. The slight energy differences could be attributed to the effect of hydrostatic pressure. Pistol and Liu have studied the Raman shift for optical phonons of GaAs_{1-x}P_x in strained quantum-well structures.⁸ The Raman spectra of the GaAs_{1-x}P_x alloys exhibit a two-mode behavior and the frequency of GaAs-like LO phonon decreases with increasing alloy concentration. They found that the GaAs-like LO phonon in the strained-alloy layers is shifted to even lower frequency due to the strain effect. When closely examining the satellite sideband of the BX emission in the PL spectra, we could not observe any significant feature clearly related to the frequency-redshifted GaAs-like LO phonon in the GaAs_{1-x}P_x barrier layers. This suggests that the deep donor centers reside in GaAs wells rather than in GaAs_{0.61}P_{0.39} barriers. By extrapolating the experimental data to the ambient pressure, we find that this donorlike deep center is a resonance about 200 meV above the bottom of the conduction band. The behavior of this deep center is in some ways similar to the so-called DX deep donor center in n -doped GaAs.²⁹⁻³² Where n -type substitutional dopants give rise to a shallow effective-mass level and a much more localized resonant level, an application of about 20-kbar hydrostatic pressure can shift DX from a resonant to a stable state. The pressure coefficient of the DX -like deep donors in S -doped bulk GaAs is reported in Ref. 32 to be 1 meV/kbar compared to 2.7 meV/kbar of BX emission. Such a difference in the pressure coefficients could be attributed to deep centers that originate from different impurity species, with resultant different energy positions and pressure dependences if the centers responsible for the BX emission are DX -like. A detailed secondary-ion-mass spectrometry (SIMS) analysis did reveal that there are carbon and silicon as well as oxygen impurities present in the samples which showed BX emission and its phonon sidebands. The concentration of each species was found to be less than 10^{15} cm⁻³ by the SIMS analysis. They were introduced into the MQW samples mainly from the arsenic gas source (AsH₃) during sample growth, and are most likely re-

sponsible for the present of the resonant donorlike deep centers. The presence of such deep centers could be the fact that accounts for the diminution of the pure PL emission related to X_z - Γ_{1hh} transition in both GaAs/GaAs_{0.76}P_{0.24} and GaAs/GaAs_{0.61}P_{0.39} MQW's at pressures above the Γ - X crossover, despite the fact that the Γ - Γ PL quenches as a result of the pressure induced by the Γ_{1e} state in the wells to cross against the X_z minima of the barriers. The localized deep centers can efficiently capture those cross-interface electrons with X characteristics through nonradiative processes such as multiphonon emission, due to their wave function derived from the contribution of the entire Brillouin zone. This process will increase the k -space overlap of the wave functions of the spatially separated electrons and holes, and results in the recombination via these deep centers having larger oscillator strength compared with the purely cross-interface indirect X - Γ recombination.

IV. CONCLUSIONS

Low-temperature photoluminescence measurements have been performed on GSMBE-grown GaAs/GaAs_{1-x}P_x MQW samples to investigate the effect of pressure on the properties of various transitions using a diamond-anvil high-pressure cell. In general, the transitions between the lowest confined electron and hole levels (Γ_{1e} - Γ_{1hh}) for all the samples studied in this work are found to depend on pressure in a way similar to the GaAs band gap but with lower pressure coefficients. The observation of the pressure-induced Γ_{1e} - X_z crossover allows a direct determination of the valence-band offset for the GaAs/GaAs_{0.68}P_{0.32} MQW sample. The unstrained valence-band offset has been determined to be 0.09 ± 0.02 eV, corresponding roughly to a 75:25 distribution in the differences of the conduction and valence bands for the constituents. We have also observed an emission associated with a deep center in the GaAs/GaAs_{0.61}P_{0.39} MQW sample. By analyzing the pressure dependence of this emission and its phonon sidebands, we suggest that the deep center is a donorlike localized state staying inside the GaAs well layer, with a resonant energy position about 200 meV above the bottom of the conduction band at ambient pressure.

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