Band alignment in GaAs/GaInP heterostructures studied by low temperature photoluminescence under high pressure

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GaAs/GaInP quantum wells were studied at 11 K using excitation energy and intensity photoluminescence coupled with hydrostatic pressure. The study over a range of 0.96 eV and 5 GPa allows comparison to alloy composition changes within the AlGaAs/GaInP system. The nature and origin of a peak at around 1.46 eV, which dominates in most samples over the quantum well emission, is shown to be a spatially indirect transition and the type II nature of the band alignment is revealed using low excitation intensities.

1 Introduction

Heterojunctions in the GaAs/GaInP system have attracted recent attention because of their device applications such as laser diodes and heterojunction bipolar transistors. However, because of the imperfect ordering in currently available GaInP grown by metalorganic vapor phase epitaxy (MOVPE), discrepancies exist between measurements such as band offsets even for similar growth conditions. Theoretical calculations show the band alignment to be dependent on CuPt-type ordering (Froyen *et al.* p.2852, Zhang *et al.* 2002, p.3111). While the band alignment is type I for fully disordered GaInP, the alignment is type II for fully ordered GaInP. The transition from type I to type II occurs at a certain order parameter where the value for the conduction band offset $\Delta E_C = E_C(\mathrm{GaInP}) - E_C(\mathrm{GaAs})$ is zero, but there is currently no consistent experimental data. The insertion of thin layers between the two material interfaces has been shown to drastically improve interface characteristics (Guimarães *et al*. 1992, p.199, Koizumi *et al*. 2003, p.560, Kwok *et al*. 1997, p.3630, Tsai *et al*. 1994, p.786, Uchida *et al*. 1997, p.771) and promote a type-I band alignment (Yu *et al*. 2002, chap. 12). In the GaAs/partially ordered GaInP quantum well (QW) structures without insertion of two thin GaP layers, an undesirable, intense emission band at ~1.46 eV often dominates instead of QW emission (Guimarães *et al*. 1992, p.199, Kwok *et al*. 1997, p.3630, Liu *et al*. 1995, p.1154, Liu *et al*. 1996, p.305, Tsai *et al*. 1994, p.786, Uchida *et al*. 1997, p.771, Yu *et al*. 2002, chap. 12). Due to its strong blueshift with increasing excitation intensity, the ~1.46 eV emission band at a higher excitation intensity is masked by the adjacent, overlapping GaAs related peak at 1.49 eV of a different nature (Kobayashi *et al*. 1999a, p.247, Kobayashi *et al*. 1999b, p.1004).

We report 11 K photoluminescence (PL) measurements of MOVPE grown GaAs/GaInP quantum wells at pressures up to ~5 GPa. We examine the distinct effects of laser excitation photon energy and excitation intensity on the PL properties, with the intention of discussing their band alignment. The use of low temperature allows us to study the nature of this ~1.46 eV emission at very low excitation powers to pressures well above the Γ-X crossover in GaInP. We see a *sublinear* shift with pressure towards higher energy close in behavior to that of the GaInP layer and a consistent Γ-X crossover and associated drop in intensity. Our results clearly show that the true nature of this emission and the interface properties are only revealed at low excitation powers and arise from the GaInP layer. The 1.46 eV emission band observed in this work is a spatially indirect transition, GaInP(Γ_C)–GaAs(Γ_V), of electrons and holes separated at the interface in a type-II band alignment with a rather smaller conduction band offset, which becomes indirect (GaInP(X_C)–GaAs(Γ_V)), both in momentum space and real space for pressures above the Γ-X crossover in GaInP.

2 Experiment

Four types of GaAs/GaInP single QW structures investigated were grown by MOVPE on (100) GaAs substrates at 550°C under the same conditions (Uchida *et al*. 1997, p.771). Sample No. 1 consists of a 100-Å-thick GaAs well and 1100 Å partially ordered GaInP barriers. Sample No. 2 has a 14-Å-thick GaP layer between the lower (i.e., nearer the substrate) GaInP and GaAs layers only, while sample No. 3 has a 28 Å GaP layer between the upper GaInP and GaAs layers. Sample No. 4 contains GaP layers sandwiching the GaAs well. The effects of excitation wavelength and excitation powers on luminescence spectra at 11 K were systematically examined using the 783 nm, 682 nm, 633 nm, 532 nm, and 488 nm lines of lasers. PL measurements at 11 K as a function of pressure up to about 5 GPa were made with a diamond anvil high-pressure cell whose design and operation are described in detail elsewhere (Kobayashi 1985, p.255).

3 Results and discussions

3.1 PL spectra at atmosperic pressure

A below-band gap PL signal with a peak energy (~1.46 eV) less than either band gap of GaAs and GaInP is observed from samples No.1, No.2 and No.3, without the insertion of two thin GaP layers between the GaAs and GaInP, and shows a strong blueshift in energy with increasing laser excitation intensity, while the quantum well emission from the GaAs QW whose peak energy is slightly higher than the sharp excitonic transitions of GaAs at ~1.51 eV is obtained only for sample No.4, where GaP intermediate layers are present at both GaAs/GaInP interfaces. In PL measurements the rather weak emissions from GaInP barriers are also observed at 1.90~1.92 eV in all samples. Their peak energies at 11 K are found to depend weakly on the excitation intensity, while no appreciable PL peak shift is detected at 77 K or higher temperatures. These observations clearly indicate that the GaInP layers used in this study have a smaller band gap than that of disordered GaInP (~1.98 eV) and are partially ordered (Kobayashi and Deol 1991, p.1289, Kobayashi *et al*. 1995, p.311). The application of a thin GaP layer at both the upper and lower interfaces is effective both in suppressing the ~1.46 eV peak and restoring emission from the GaAs QW. The presence of the GaAs/partially ordered GaInP interface with no thin GaP intermediate layer seems to be closely related to the appearance of this intense emission at ~1.46 eV.

Figure 1 shows the comparison of the typical PL spectra of sample No.2 under several different excitation photon energies, measured at 9–11 K and at atmospheric pressure. Excitation intensities are ~0.7, ~1.5 and ~3.0 mW. Note that PL intensity is on a logarithmic scale. Among all the emission peaks in the spectra, only the below-band gap PL spectrum at ~1.46 eV (denoted as Peak A) shows a strong blueshift of its peak energy with excitation laser power. At 2.54 eV and 2.33 eV excitation, whose energies are high enough to excite both the partially ordered GaInP and GaAs layers, it is clear that Peak A totally dominates below the GaAs band gap energy. However, when excited at the lower energy of 1.96 eV, which is rather close to the band gap energy of partially ordered GaInP (~1.9 eV), the excitonic transitions of GaAs at ~1.51 eV (denoted as P2) and the impurity-related peak at 1.49 eV (denoted as P3) apparently become brighter, and are almost comparable to Peak A in intensity. At an energy of 1.82 eV, which is already below the band gap of GaInP but well above that of GaAs, these near band-edge transitions P2 and P3 clearly show a drastic increase in intensity relative to that of Peak A. When excited at 1.58 eV, which is just above that of GaAs, the emission bands P2 and P3 almost dominate below the GaAs band gap energy in the spectra.

These results strongly suggest that the presence of the photoexcited electrons in the GaInP layers is greatly responsible for the observed ~1.46 eV emission (Peak A). In sample No.4, it is found that the QW emission band (denoted as P1) at ~1.53 eV, which has been clearly observed under 2.54 eV or 2.33 eV excitation, is no longer observable. This might indicate that electrons and holes optically generated in the GaInP layers can tunnel through the thin GaP layer and can be trapped by the GaAs QW where they recombine at 1.53 eV. However, when excited at lower energies especially at 1.82 eV or 1.58 eV, the population of electrons in the GaInP layers will be too low for the GaAs QW emission to be detectable at 1.53 eV.

Since the PL properties of samples other than No.4 depend significantly on the excitation intensity, the PL measurements should be made under intentionally lower excitation to investigate the characteristics and the origin of the anomalous peak at ~1.46 eV (Peak A). Otherwise, Peak A is found to be masked by the adjacent, overlapping 1.49 eV band (P3) of a different nature at a higher excitation intensity, an effect clearly investigated in earlier time-resolved photoluminescence (TRPL) measurements (Kobayashi *et al*. 1999a, p.247, Kobayashi *et al*. 1999b, p.1004).

In addition to these changes in PL features below ~1.53 eV, we also obtain the up-converted photoluminescence (UPL) under 1.58 eV excitation, in which samples No.2 and No.3 emit photons with a higher energy ~1.89 eV (Kobayashi et al. 2003, p.277). This UPL peak energy does not show an appreciable shift with excitation intensities, and is closer to that of the normal PL peak of partially ordered GaInP especially when measured at a sufficiently low excitation intensities below 0.1 mW. No UPL, however, can be observed from the sample No.4 with two thin GaP layers, where the ~1.46 eV emission (Peak A) is also absent. Both the UPL and the ~1.46 eV emission observed are made possible by a type II band alignment at the GaAs/partially ordered GaInP interface with no thin GaP intermediate layer.

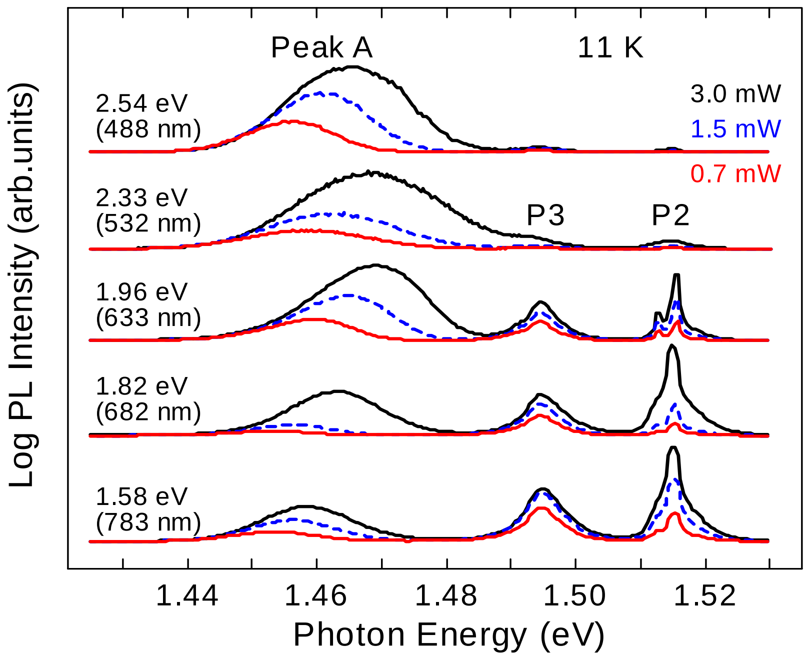

Fig. 1 Comparison of typical PL spectra of sample No.2 at 11 K for five excitation energies 2.54 eV, 2.33 eV, 1.96 eV, 1.82 eV and 1.58 eV, and for three different excitation intensities ~0.7 mW, ~1.5 mW and ~3.0 mW. PL intensity is on a logarithmic scale.

3.2 PL spectra at high pressures

With increasing pressure each spectrum shifts to higher energies. Figure 2 shows some typical PL spectra of sample No.2 at 11 K for two different pressures 3.19 GPa, and 4.33 GPa. PL spectra shown are excited at 2.33 eV with several excitation intensities below ~5 mW. Figure 3 describes the corresponding radiative recombination process responsible for the luminescences near the Γ-X crossover in partially ordered GaInP (3.2–3.5 GPa) or in GaAs (~4 GPa) as well as that for the ~1.46 eV emission (Peak A) at atmospheric pressure (0 GPa), which is based on the type II band alignment with a smaller conduction band offset ($\Delta E_C \leq 0$) (Su *et al*. 1996, p.933).

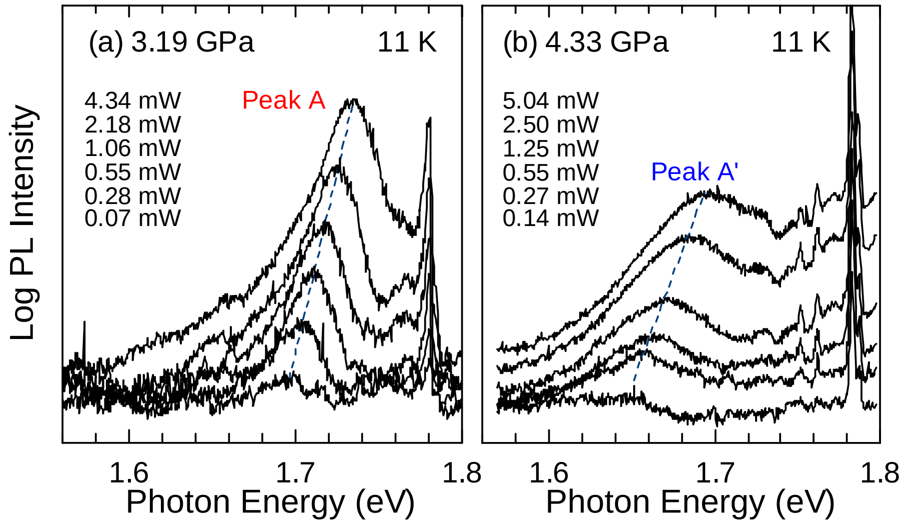

Fig. 2 PL spectra at high pressures a) 3.19 GPa and b) 4.33 GPa of sample No.2 under various excitation intensities at 11 K. Excitation is at 2.33 eV (532 nm). The sharp spikes at ~1.78 eV are from ruby R_1 and R_2 lines used for pressure measurements.

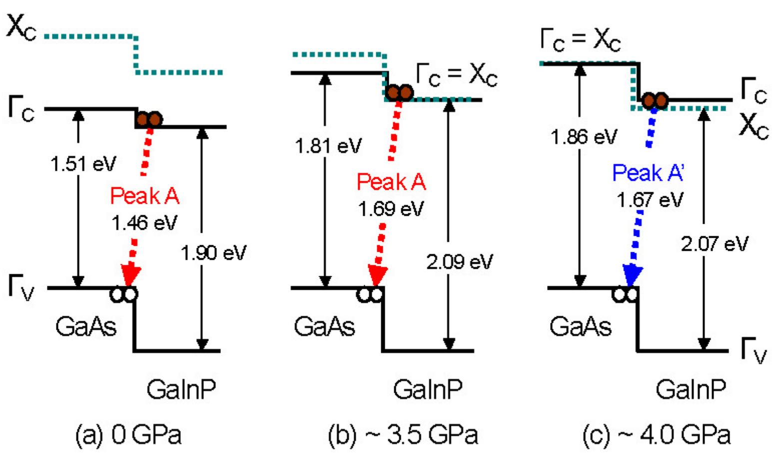

Fig. 3 Band energies at the GaAs/partially ordered GaInP interface for three pressures and radiative recombination processes responsible for Peak A and Peak A'. a) Atmospheric pressure, b) ~3.5 GPa, near the Γ-X crossover in GaInP, and c) ~4.0 GPa, near the Γ-X crossover in GaAs.

In Fig. 2 (a) it is clearly shown that the spectral position of Peak A at 3.19 GPa strongly depends on the excitation intensity. This is a great contrast to the impurity-related GaAs band (P3) in sample No.2 or the QW emission in sample No.4. The sample No.2 has a type II band alignment where the Γ conduction band minimum is still the lowest in GaInP. Peak A is caused by the recombination of the electrons and holes confined at the type II heterojunction, GaInP(Γ_C)–GaAs(Γ_V), in Figs. 3 (a) and (b), and the observed blueshift is explained by the rise of the quasi-Fermi level in the heterojunction with the increase of excitation. In Fig. 2 (b) the emission band at 4.33 GPa, which is just beyond the pressure for the Γ-X crossover in GaAs, shows a similar blueshift with excitation intensity. However, its peak energy is about 20 meV lower than the corresponding spectrum with a similar excitation intensity at 3.19 GPa. Its intensity is at least one order of magnitude weaker than those at lower pressures. This is manifested by the intensity of sharp spikes from ruby R_1 and R_2 lines observed at ~1.78 eV. In addition, we also notice that its spectral width is broader than those for lower pressures. These suggest a change in recombination processes with increasing pressure. These tendencies were not clear in our previous work at 77 K (Kobayashi *et al*. 2001, p.123, Kobayashi *et al*. 2003, p.277) for higher pressures above ~3.5 GPa, but are observable at lower excitation intensities in the present 11 K measurements. Since the Γ-X crossover in partially ordered GaInP in sample No.2 occurs at 3.2–3.5 GPa, the transition GaInP(Γ_C)–GaAs(Γ_V) at lower pressures is replaced by GaInP(X_C)–GaAs(Γ_V) which is denoted as Peak A' in Fig. 3 (c).

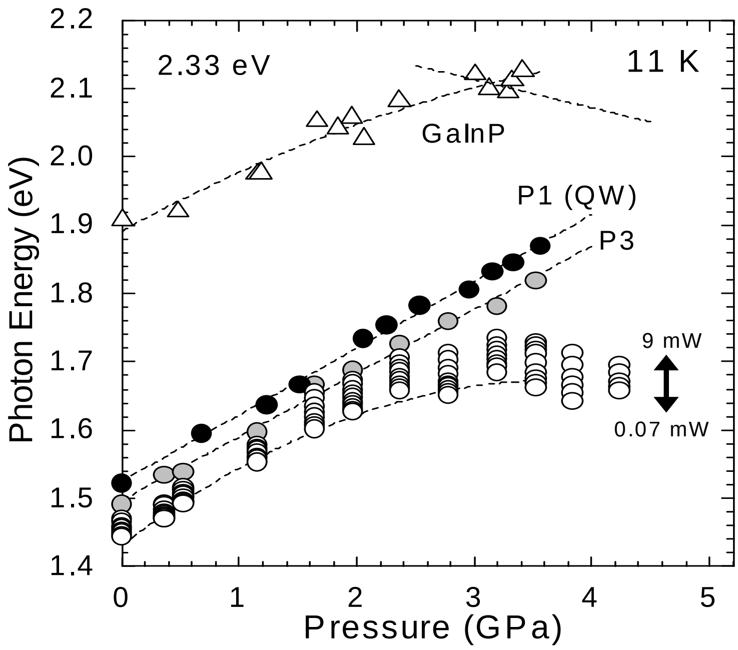

Fig. 4 PL peak energies of sample No.2 at 11 K as a function of pressure. The excitation intensities at 2.33 eV are in the range 0.07~9 mW. Results for QW emission of sample No.4 are also shown for comparison.

We have found that the peak energy of Peak A shows a quite different dependence with pressure when compared to those of P2 and P3. Figure 4 shows the pressure dependence of the PL peak energies of sample No. 2 at 11 K, measured under relatively lower excitation intensities below 9 mW at 2.33 eV. The results for the PL peak of a partially ordered GaInP layer of sample No. 2 and for the QW emission peak (P1) of sample No. 4 are also shown for comparison. Several important characteristics of the PL peaks are immediately apparent. For sample No. 2, the ~1.46 eV emission band (Peak A) shows a *sublinear* shift towards higher energies up to ~3.5 GPa depending strongly on the excitation intensity. It should be noted that its peak shift measured at sufficiently lower excitation is rather similar to that obtained for partially ordered GaInP layers. In addition, it is found that the energy shifts of Peak A with increased excitation intensity at higher pressures are larger than those at lower pressures. These features are in contrast to the QW emission at 1.53 eV in sample No. 4 which shifts almost linearly at a rate of ~103 meV/GPa or the weaker impurity-related emission (P3) at 1.49 eV detected to shift up to ~4 GPa in a manner similar to that observed in bulk GaAs. Beyond ~3.5 GPa, however, the emission peak clearly exhibits a shift towards lower energies as well as a drop of its intensity. A blueshift of transition energy with increased excitation intensity is also observed. This is due to indirect radiative recombinations in both momentum and real space shown by Peak A' in Fig. 3 (c). These PL features above ~3.5 GPa were not so clear in the previous 77 K measurements. Similar observation of the indirect transition GaInP(X_C)–GaAs(Γ_V) was also reported for type I GaAs/disordered GaInP heterojunctions above 2.6 GPa (Chen *et al*. 1990, p.693).

The various PL features at high pressure would imply that the presence of ordered GaInP layers plays an important role in the radiative recombination at ~1.46 eV (Peak A). According to recent theoretical calculations by Zhang *et al*. (2002, p.3111), band alignment between GaAs and partially ordered $Ga_xIn_{1-x}P$ is found to change from type I to type II at the order parameter η=0.46 for a GaAs lattice matched composition. Furthermore, since the pressure coefficient of the GaAs band gap energy is much larger than that of the partially ordered GaInP (Kobayashi and Deol 1991, p.1289, Kobayashi *et al*. 1995, p.311), high pressure may convert the band alignment from type I to type II. Thus, the luminescence due to a spatially indirect recombination of electrons and holes separated at the interface is made possible by a type-II band alignment. Zeman *et al.* (1999, p.239) showed that samples revealing a type I band alignment do not show appreciable UPL at atmospheric pressure, but exhibits up-conversion under a pressure of 1.2 GPa. This fact also supports the type II band alignment in these samples to give efficient UPL at high pressures beyond 1.2 GPa. When a type II heterojunction is photoexcited, the band bending near the interface is enhanced so that the transition energy of the interface luminescence is increased due to the increased quantized energy levels for electrons and holes. As the excitation intensity is further increased, the increased electrons and holes can occupy higher energy levels (band filling effect). This may explain the larger blueshift of the spectral position at higher excitation intensities.

Similar emission peaks below the band gap of each constituent semiconductor are also observed in many other systems in which band alignments are type II and optical transitions observed are spatially indirect. Kim *et al*. (1995, p.1718) investigated PL properties of $Al_xGa_{1-x}As$/GaInP heterojunctions in the range of Al composition x=0–0.4 in both type-I and type-II band alignment regimes, and determined the Al composition for the null conduction band offset (x=0.11). A below-band gap PL spectrum with the peak energy less than either band gap of $Al_xGa_{1-x}As$ and GaInP is observed only from the samples with Al compositions x larger than 0.12. This peak shows a higher blueshift with excitation intensity for the samples with Al compositions above 0.12, and therefore, having a larger conduction band offset (ΔE_C<0) in the type II band alignment. Figure 5 (a) shows the variation of conduction band offset ΔE_C in a type II band alignment with Al mole fraction in $Al_xGa_{1-x}As$/GaInP system. This can be compared with the energy difference between the near band-edge transition P3 and Peak A in sample No. 2 as a function of pressure in Fig. 5 (b), showing that pressures above ~1.2 GPa can greatly affect the conduction band offset in a type II band alignment.

In contrast to our present work the GaInP layers in their $Al_xGa_{1-x}As$/GaInP samples are disordered, which is manifested by the GaInP emission peak energy at about 2.0 eV. Nevertheless, the various PL results for $Al_xGa_{1-x}As$/GaInP heterojunctions with different Al compositions (x=0.12–0.33) are consistent with our present results for GaAs/partially ordered GaInP QWs at various pressures (0–3.5 GPa), with the only exception of abrupt increase in energy difference above ~3.5 GPa. This is due to the emergence of the indirect transition GaInP(X_C)–GaAs(Γ_V), as shown in Fig. 3 (c). These results show the similarity in effects of "pressure" and "alloy composition" on the band structures.

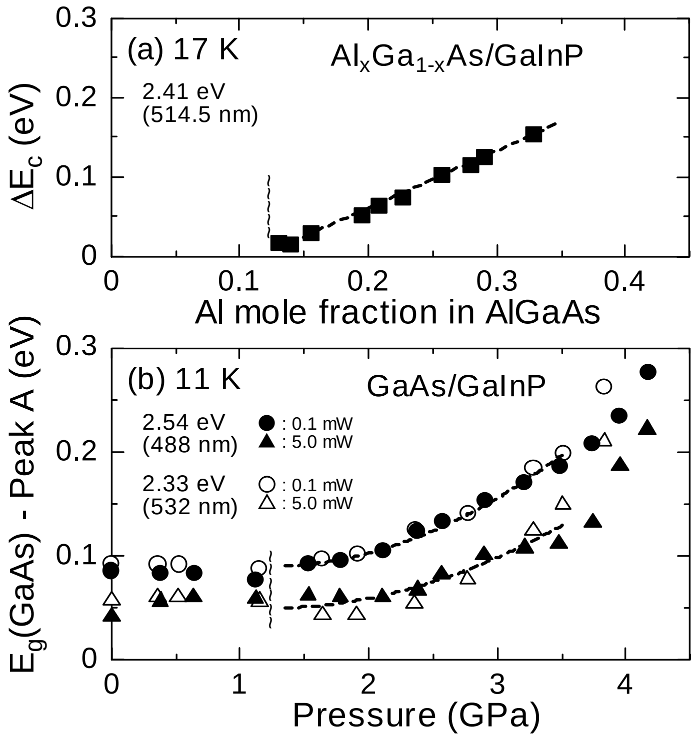

Fig. 5 a) The variation of conduction band offset ΔE_C in a type II and alignment with Al mole fraction in $Al_xGa_{1-x}As$/GaInP system (Kim *et al.* 1995, p.1718). b) Typical energy difference between near band-edge transition P3 and Peak A in sample No. 2 as a function of pressure. Excitation photon energies are 2.33 eV and 2.54 eV.

4 Conclusions

We have studied the PL properties of GaAs/partially ordered GaInP heterostructures at pressures up to ~5 GPa at 11 K enabling us to obtain the high-pressure behaviour of the ~1.46 eV emission (Peak A) in detail at low excitation intensities. This is in contrast to an earlier study at 77 K. Selecting the smaller excitation photon energy below the GaInP band gap but above the GaAs band gap, we have found that the Peak A with an energy (~1.46 eV) less than the band gap of GaAs drastically decreases in intensity while the emission bands from the GaAs layer at 1.49 eV and 1.51 eV dominate. The pressure dependence of Peak A obtained especially at lower excitation intensities is quite different from that of GaAs related transitions but is very similar to those of a partially ordered GaInP layer. These results strongly suggest that the presence of a GaInP layer is closely related to the observation of Peak A. In addition, the shift of this peak toward lower energy with pressure, depending on the excitation intensity, was observed after the Γ-X crossover in a partially ordered GaInP layer. This indicates the change from the spatially indirect transition, GaInP(Γ_C)–GaAs(Γ_V), of electrons and holes separated at the interface in a type-II band alignment between GaInP and GaAs to the indirect transition GaInP(X_C)–GaAs(Γ_V), both in real space and momentum space, which is achieved by the application of high pressure. The effects of pressure on the energy band structure can also be compared to the effects of changing the Al alloy composition x in $Al_xGa_{1-x}As$/GaInP systems.

Acknowledgements

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