Alloy composition dependence of defect energy levels in $Ga_xIn_{1-x}P$/InP:Fe and $Ga_xIn_{1-x}P$/InP:S ($x \leqslant 0.24$)

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The defect energy levels in metalorganic chemical vapor deposition (MOCVD) grown $Ga_xIn_{1-x}P$/InP:Fe and $Ga_xIn_{1-x}P$/InP:S epilayers ($x \leqslant 0.24$) have been studied by photoluminescence (PL) and photoconductivity (PC) measurements. To understand the origin of the observed deep levels, we have determined the temperature dependence of the intensity and half-width of the dominant deep-level PL peaks. We find that (1) the dominant deep-level peaks of the samples grown on the same substrate are related to the epilayer composition, and move to higher energies with increasing gallium content; (2) the dominant deep-level peaks of the samples with the same epilayer composition grown on different substrates are different. They are attributed to the impurity in the substrate diffusing into the epilayer during MOCVD growth, forming an impurity-vacancy complex. The following tentative assignments are proposed: the dominant deep-level peaks in $Ga_xIn_{1-x}P$/InP:Fe and $Ga_xIn_{1-x}P$/InP:S are attributed to the emission of a $(V)_P$-$(Fe)_{III}$ complex and a $(V)_{III}$-$(S)_P$ complex, respectively. Comparing the deep level with the near-band-edge emission we show that (1) all deep levels are independent of the band edge as x is varied; (2) the composition dependences of the deep levels associated with such complexes depend on the site occupied by the impurity atom.

I. INTRODUCTION

A considerable effort has been devoted to the study of defect centers in semiconductor materials. The performance characteristics of a semiconductor device can be severely limited by effects which are related to defects in its material. In particular, such defects may produce electronic energy levels which lie in or near the fundamental band gap and these levels can greatly influence the optoelectronic properties of the device.[1]

The physics of the alloy composition dependence of the deep levels associated with the As vacancy in $Al_xGa_{1-x}As$ and with the anion vacancy in $GaAs_{1-x}P_x$ have been predicted by Tang *et al.*[2] and by Ford and Myles.[3] The composition dependence of the defect energy levels associated with vacancy-impurity complexes in $Hg_{1-x}Cd_xTe$ has also been investigated.[4,5] Several workers have observed and studied alloy broadening effects in optical absorption, luminescence, and deep-level transient spectroscopy (DLTS) in III-V and II-VI semiconductor alloys.[5–8]

The ternary compound GaInP has attracted much attention because of its important technological role in the fabrication of high efficiency light-emitting diodes and double-heterostructure visible lasers. GaInP lattice matched to GaAs has been investigated by some authors.[9,10] However, much less attention has been paid to GaInP epitaxial layers on InP substrates, primarily due to difficulties of growth arising from the lattice mismatch. Zn-doped GaInP/InP and undoped GaInP/InP have been successfully grown,[11,12] and experimental results show that these materials can be used for high-speed Schottky photodetectors.[11] It is therefore of interest to study the level structure in the forbidden gap introduced by impurities, native defects, and their complexes.

In this paper we report for the first time the alloy composition dependence of the deep levels associated with $(Fe)_{III}$-$(V)_P$ and $(V)_{III}$-$(S)_P$ complex, i.e., iron substituting in the element-III sublattice $(Fe)_{III}$ combines with a phosphorus vacancy $(V)_P$ and sulfur substituting in the phosphorus sublattice $(S)_P$ combines with a vacancy of the element-III sublattice $(V)_{III}$, in $Ga_xIn_{1-x}P$/InP:Fe and $Ga_xIn_{1-x}P$/InP:S ($x \leqslant 0.24$). In order to determine the possible recombination mechanisms and origins of the deep levels, we have observed the PL spectra for the substrates and epilayers with different Ga content on InP:Fe or InP:S substrates. In addition, the temperature dependence of the PL peaks was studied and the half-width versus temperature plots were also analyzed using a configurational coordinate (CC) model.[13,14]

II. EXPERIMENTAL DETAILS

The samples in this work were prepared by low-pressure metalorganic chemical vapor deposition (MOCVD). They consist of a (100) oriented InP substrate doped with either Fe or S, followed by a 1000 Å undoped InP buffer layer and finally by a nominally undoped (residually n-type) $Ga_xIn_{1-x}P$ film ($x \leqslant 0.24$), whose thickness is about 1 μm. The detailed growth conditions have been described elsewhere.[12] The epilayer compositions were determined with the aid of high-resolution x-ray diffraction.

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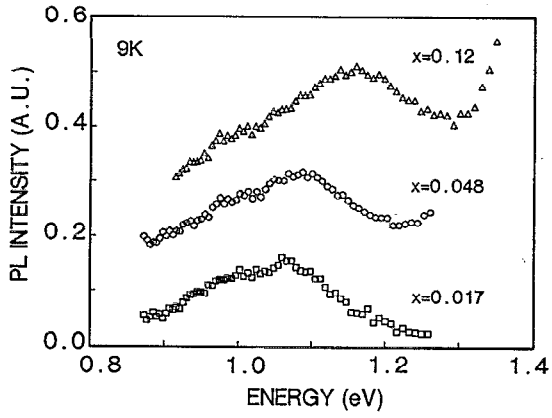


FIG. 1. Representative deep-level PL spectra of $Ga_xIn_{1-x}P$ epilayers on InP:Fe with x equal to 0.017, 0.048, and 0.12, at ~9 K, and laser excitation intensity is ~100 W/cm^2.

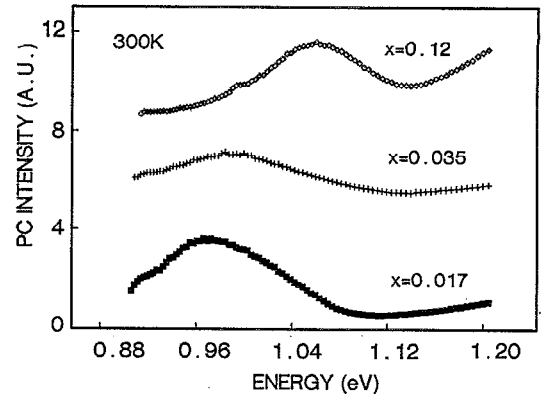


FIG. 2. Representative PC spectra of $Ga_xIn_{1-x}P$/InP:Fe for x equal to 0.017, 0.035, and 0.12 at ~300 K.

The PL measurements were performed using the 5145 Å line of an Ar^+ laser. The signal was dispersed by a 1 m spectrometer and detected by a liquid-nitrogen-cooled Ge *p-i-n* photodiode using conventional lock-in techniques. The sample temperature was maintained in a continuous flow helium cryostat. The photoconductivity was excited using a tungtsen lamp and monochromatic light was obtained through a monochromator. The samples were cut in rectangular shapes, and the ohmic contacts were formed by alloying Au-Ge (88:12)/Ni, followed by a 350 °C anneal in forming gas for 10 s. The samples were mounted on the cold finger of a cryostat and the photocurrent measured by conventional lock-in techniques.

III. RESULTS AND DISCUSSIONS

A. Dependence of deep levels on Ga content

Figure 1 shows representative PL spectra of $Ga_xIn_{1-x}P$/InP:Fe samples observed at a temperature $T\sim9$ K and a laser excitation intensity of ~100 W/cm^2. The deep-level PL peaks are broad and do not have any fine structure. The dominant peak positions shift to higher energies with increasing Ga content. For x equal to 0.017, 0.048, 0.12, and 0.17, corresponding peak maxima have been observed at ~1.066, ~1.092, ~1.158, and ~1.175 eV, respectively. We find that the PL signals of the dominant peaks decrease with increasing Ga content. We have also measured a $Ga_{0.24}In_{0.76}P$/InP:Fe sample but no obvious peak was observed.

Figure 2 shows the PC spectra for a series of $Ga_xIn_{1-x}P$/InP:Fe samples obtained at $T\sim300$ K. They are similar to the low-temperature PL spectra shown in Fig. 1. The dominant PC peak positions shift to higher energies with increasing Ga content. In order to reduce persistent photoconductivity decay,[15] which will influence the PC spectra,[16] the samples were left in the dark for one day before the PC spectra were measured. The differences between the PC peaks of different epilayers are close to those in their PL spectra. The temperature dependence of the energy gap has the general form[17]

$$E_g(T)=E_g(0)-\alpha T^2/(T+\theta), \tag{1}$$

where $E_g(0)$ is the energy gap at 0 K, α is an empirical parameter, and θ is second empirical parameter related to the Debye temperature. Measurement of the energy gap of a $Ga_{0.017}In_{0.983}P$/InP:Fe sample as a function of temperature yields a value for $\alpha=3.74\times10^{-4}$ eV/K, $\theta=170$ K, and 0.078 eV shift of the energy gap from 7 to 300 K. If we assume that the shift of the deep level with temperature follows the energy gap, the dominant PL deep-level peak of $Ga_{0.017}In_{0.983}P$/InP:Fe should move to around 0.994 eV at 300 K, which is close to the peak position of the PC spectrum at 300 K as shown in Fig. 2. The $Ga_{0.017}In_{0.983}P$/InP:Fe samples illustrated in Figs. 1 and 2 are from the same chip. We only measured the temperature dependence of the band-gap energy for $Ga_{0.017}In_{0.983}P$/InP:Fe sample. Since the empirical parameters α and θ are functions of the material composition, therefore, temperature-induced shift of the band gap is different for different samples. Even so, the PC measurements support of the conclusions derived from PL measurements.

B. Level identification

In order to find the origin of the deep levels, the temperature dependence of the integrated emission intensity and the half-width of the dominant PL peak was measured for the $Ga_{0.048}In_{0.952}P$/InP:Fe sample. Figure 3 shows a plot of the integrated intensity, on a logarithmic scale, against 10^3 T^{-1}. The curve increases slightly from 7 to 22 K and then decreases with a single activation energy of 0.078 eV. Above 200 K the PL signal is too weak to be observed. Figure 4 shows the variation of the half-width of the PL dominant peak with temperature together with a fit for this variation using the configuration coordinate model equation[13]

$$W=A[\coth(\hbar\omega/2KT)]^{1/2}, \tag{2}$$

with $\hbar\omega=0.018$ eV. In this equation, A is a constant whose value is equal to the half-width W as the temperature approaches 0 K, and $\hbar\omega$ is the energy of the vibrational mode

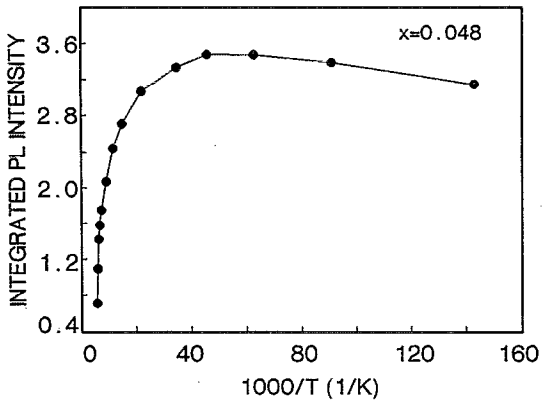


FIG. 3. Variation of the integrated intensity of the dominant PL peak with temperature for $Ga_{0.048}In_{0.952}P$/InP:Fe. Activation energy is derived from the slope of straight (solid) line.

of the excited state. Such a temperature dependence of the PL intensity and the half-width is valid for vacancy-related peaks on the basis of the CC model.[13,14]

We have also investigated samples with the epilayers having the same Ga content $x=0.017$ but grown on different substrates (InP:Fe or InP:S). Both epilayers were simultaneously grown on the two substrates. The observed PL spectra of samples $Ga_{0.017}In_{0.983}P$/InP:Fe and $Ga_{0.017}In_{0.983}P$/InP:S are shown in Fig. 5(a). Both curves were measured at 9 K with a laser excitation intensity of ~ 100 W/cm^2. The dominant PL peaks of samples $Ga_{0.017}In_{0.983}P$/InP:Fe and $Ga_{0.017}In_{0.983}P$/InP:S are at ~ 1.066 and ~ 1.148 eV, respectively. Figure 5(b) illustrates the PL spectra of the InP:Fe and InP:S substrates. For InP:Fe the PL spectrum is characterized by a broad peak centered at ~ 1.068 eV, which is quite different from that of InP:S.

Since the samples are small and since the InP:Fe and InP:S substrates are very close during MOCVD growth, the effect of the temperature difference between two substrates can be neglected. Slight variation of the gas flow influences the sample composition, but we have observed

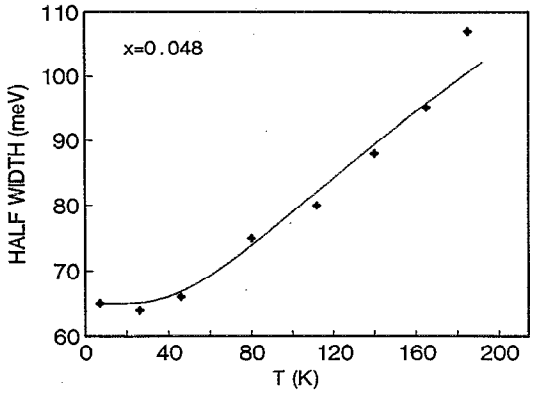
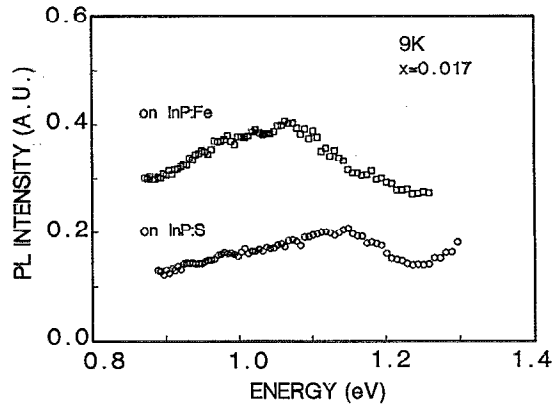


FIG. 4. Variation of the half-width of the dominant PL peak with temperature for $Ga_{0.048}In_{0.952}P$/InP:Fe. "+" is measured data and solid line is theoretical fit using Eq. (2).



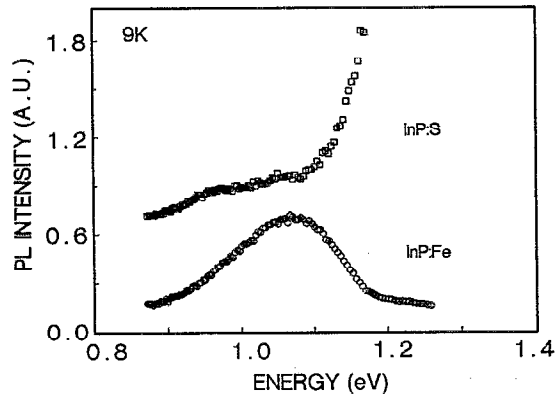


FIG. 5. (a) The deep-level PL spectra of $Ga_{0.017}In_{0.983}P$/InP:Fe, and $Ga_{0.017}In_{0.983}P$/InP:S, at ~ 9 K. (b) The deep-level PL spectra of substrates InP:Fe, and InP:S, at ~ 9 K. Laser excitation intensity is ~ 100 W/cm^2.

that the compositional variation is less than 10%. We have also observed that the energy gaps and the deep levels of different composition epilayers grown on InP:Fe or InP:S evolve in a coherent manner, so the effects of slightly different growth conditions such as gas flow or temperature variations within the growth chamber do not influence our conclusions.

Temkin *et al.*[18] observed a 1.08 eV band in InP and attributed it to emission from a donor and acceptor complex. Huang and Wessels[19] studied the incorporation of Fe into InP grown by organometallic vapor phase epitaxy and found that the intensity of the 1.07 eV peak increased with Fe concentration, indicating that this peak is Fe related. Yu[20] and Lliadis *et al.*[21] reported a 1.1 eV PL emission band in InP:Fe. They attributed this band to a nearest-neighbor molecular-like defect center $(Fe)_{In}$-$(V)_P$ complex. In our case, the temperature dependence of the PL intensity and half-width suggest the presence of native defects. In Fig. 5(a) the energies of the dominant PL peaks are substrate dependent. The origin of the peak in $Ga_xIn_{1-x}P$/InP:Fe is probably due to a $(V)_P$-$(Fe)_{III}$ complex, i.e., a nearest-neighbor molecular-like defect formed with substitutional iron in either gallium or indium sites together with a phosphorus vacancy. Fe in the substrate diffuses into the epilayer during MOCVD growth, as has been observed by Holmes *et al.*[22] It combines with a phos-

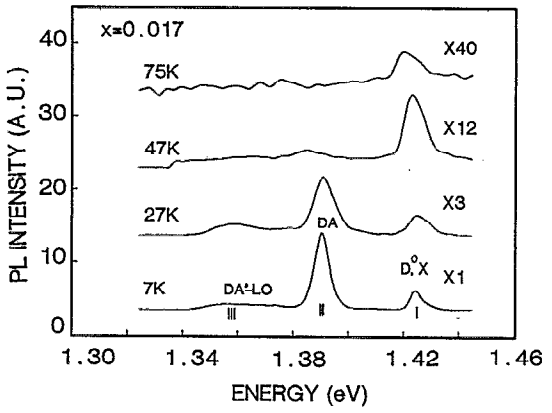


FIG. 6. Evolution of the near-band-edge PL spectra of $Ga_{0.017}In_{0.983}P$/InP:Fe with temperature 7, 27, 47, and 75 K.

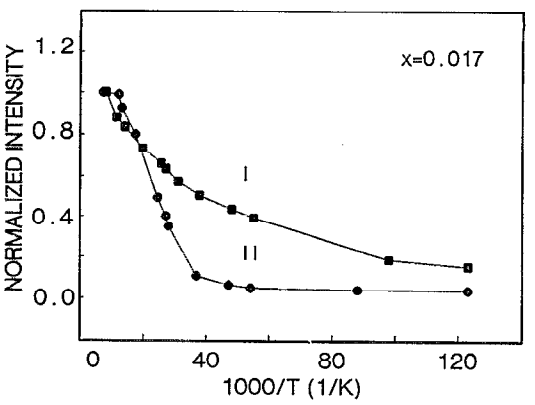


FIG. 7. Variation in the normalized integrated intensity of the near-band-edge PL spectra of $Ga_{0.017}In_{0.983}P$/InP:Fe with temperature for I: 1.424 eV band, and II: 1.39 eV band.

phorus vacancy in the epilayer, forming $(V)_P$-$(Fe)_{III}$ nearest-neighbor pairs. The Fe ions were found to form a substitutional solid solution and occupy sites of the In sublattice of InP:Fe.[23] The decrease in intensity of the dominant PL peaks with increasing Ga content is attributed to the increase in the density of dislocations with increasing Ga content in the epilayers. The dislocations act as efficient carrier trapping and recombination centers. Thus a higher Ga content should be followed by a weaker PL intensity, as observed, due to more nonradiative recombination.

For the $Ga_xIn_{1-x}P$/InP:S samples used in this study the dominant PL peak is probably due to a $(V)_{III}$-$(S)_P$ complex. This is compatible with the well-known fact that S ions from the substrate occupy sites in the P sublattice, the $(V)_{III}$ is assumed to be an acceptor, and the Coulombic force between $(V)_{III}$ and $(S)_P$ should give rise to localized nearest-neighbor pairs. As shown in Fig. 5(a) the 1.066 eV emission in $Ga_{0.017}In_{0.983}P$/InP:Fe is stronger than the 1.148 eV emission in $Ga_{0.017}In_{0.983}P$/InP:S. We have also observed that for epilayers, on the different substrates but with the same Ga content, the PL intensities of the dominant peaks of $Ga_xIn_{1-x}P$/InP:Fe are always stronger than those of $Ga_xIn_{1-x}P$/InP:S grown simultaneously. It is probable that in both samples there are more P vacancies than those of the group III elements. Therefore, the PL emission which is related to a P vacancy is stronger than that associated with a column III element vacancy.

C. Relation between deep levels and band gap

Near-band-edge luminescence has been observed for all our $Ga_xIn_{1-x}P$/InP:Fe samples. We have carried out an investigation on the evolution of the near-band-edge emission with increasing temperature for the $Ga_{0.017}In_{0.983}P$/InP:Fe sample as shown in Fig. 6. At 7 K the strong band-edge luminescence in 1.3–1.5 eV range consists of three bands, which will be referred to as I (1.424 eV), II (1.39 eV), and III (1.356 eV). From 7 to 20 K there is an appreciable change of the relative intensities of the emission bands even though an overall decrease in intensity is observed, as illustrated in Fig. 7. A sudden decrease in band II occurs from 20 to 40 K, and a slower change appears in band I. At the same time, the peaks shift towards lower energies. At high temperatures band I is the only emission present. A similar behavior of the peaks at 1.41, 1.38, and 1.34 eV was observed in n-type InP.[24,25] In the low-temperature PL spectrum of InP, the band at 1.41 eV is attributed to a neutral donor-bound exciton (D^0,X);[25] the bands at 1.38 and 1.34 eV are, respectively, a donor-acceptor (DA) or band to acceptor (BA) transition related to zinc or carbon impurities,[24–30] and the first longitudinal optical (LO) phonon replica of the band at 1.38 eV.[24] From the above results we suggest that in the PL spectra of $Ga_{0.017}In_{0.983}P$/InP:Fe the bands I and II are (D^0,X) and (DA) transitions, respectively. Band III is the phonon replica of the band II. The acceptor may be related to carbon,[28] but its identification remains uncertain. PL spectra of samples with different Ga content are similar to that of $Ga_{0.017}In_{0.983}P$/InP:Fe with features, which shift to the higher energies with increasing Ga content. The emission intensity near the band edge decreases markedly with increasing Ga content, and the linewidth increases with Ga content appearing to saturate above about $x=0.12$. These phenomena can be attributed to the presence of misfit dislocations.[12]

For samples having the same Ga content but grown on different substrates, the near-band-edge emissions differ, both in peak position and structure. It is likely that Fe and S impurities diffuse into the epilayers, and change their optical properties, and these results will be reported elsewhere.

In order to determine the variation of the band-gap energies with the alloy composition $\Delta E_g(x)$ for the $Ga_xIn_{1-x}P$/InP:Fe samples, we have subtracted the 1.416 eV emission energy in InP from the (D^0,X) peaks observed in the $Ga_xIn_{1-x}P$/InP:Fe emission spectra, i.e., $\Delta E_g(x) = E_{DX}(x) - E_{DX}(0)$. A similar method has been used to determine the variation of the deep-level energies with the alloy composition $\Delta E_1(x)$, in which we have subtracted the value of the dominant deep-level energies of InP/InP:Fe from that of $Ga_xIn_{1-x}P$/InP:Fe $(0<x\leqslant 0.24)$.

A least-squares fit to the variation of $\Delta E_g(x)$ and $\Delta E_1(x)$ gives the following expressions:

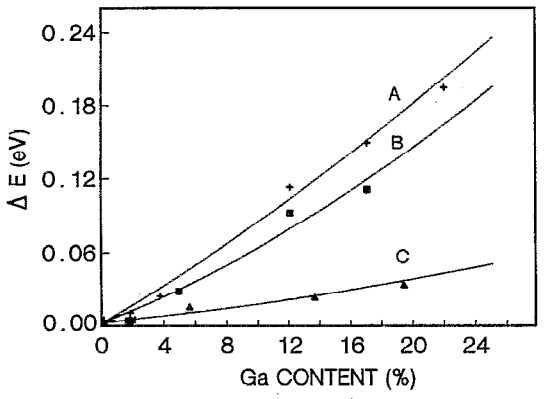


FIG. 8. Composition dependences of the energy levels extracted from the PL data, A: band-gap energies $\Delta E_g(x)$ of $Ga_xIn_{1-x}P$/InP:Fe, B: deep-level energies $\Delta E_1(x)$ of $(Fe)_{III}$-$(V)_P$ complex in $Ga_xIn_{1-x}P$/InP:Fe, and C: deep-level energies $\Delta E_2(x)$ of $(V)_{III}$-$(S)_P$ in $Ga_xIn_{1-x}P$/InP:S. Points are measured data and solid lines are least-squares fit.

$$\Delta E_g(x)=0.765x+0.675x^2 \tag{3}$$

and

$$\Delta E_1(x)=0.52x+0.998x^2. \tag{4}$$

The variation of the band gap with composition for bulk GaInP has been evaluated by Merle *et al.*[31] in the low x region. They find

$$\Delta E_g(x)=0.77x+0.684x^2. \tag{5}$$

Equation (5) is slightly different from Eq. (3). This is because the GaInP epilayers have a different lattice constant than the substrate and a strain is generated which is not completely relaxed.[12]

Equations (3) and (4) clearly show that the deep levels are independent of the conduction-band edge as x is varied. Ford *et al.*[3] have calculated the energies of the vacancy-associated levels and have applied their results to the investigation of the alloy broadening of the deep levels produced by As vacancies in $Al_xGa_{1-x}As$. Their prediction for $Al_xGa_{1-x}As$ is similar to the above result. Our results show that this prediction is valid for $(V)_P-(Fe)_{III}$ complex in $Ga_xIn_{1-x}P$/InP:Fe as well.

We have measured the deep radiative levels in the $Ga_xIn_{1-x}P$/InP:S samples for $x=0$, 0.017, 0.055, 0.12, and 0.194. The deep-level energies of the dominant peaks in the PL spectra, which are associated with $(V)_{III}$-$(S)_P$ complex, have been evaluated using the method described above to obtain the variation of the deep level energies $\Delta E_2(x)$, which are plotted in Fig. 8 together with the data of $Ga_xIn_{1-x}P$/InP:Fe. A least-squares fit to $\Delta E_2(x)$ yields the following equation:

$$\Delta E_2(x)=0.1x+0.25x^2. \tag{6}$$

From Eqs. (4) and (6) we find that the alloy composition dependence of the deep levels which are associated with a particular complex depends on whether the impurity occupies the cation or the anion site of the host crystal. This fact has been predicted by Myles *et al.*[4] and has been observed in PL[4] and in deep-level transient spectroscopy[5] of $Hg_{1-x}Cd_xTe$. Our results show that this prediction is also valid for $Ga_xIn_{1-x}P$/InP.

The uncertainties associated with Eqs. (3), (4), and (6) can be estimated from measurements of the sample composition and homogeneity, the PL spectra and an evaluation of the least-squares fit. In this study, the spectrometer provided resolution and repeatability of $\sim$1 meV, which is smaller than the uncertainty due to the compositional inhomogeneity of the samples. We have repeated the PL measurements at different points on each sample, and uncertainties of $\sim$0.3% for band-gap energies and $\sim$0.8% for deep-level energies have been observed. The larger uncertainty of the deep-level energies is because both observed peak width and noise in the deep-level PL spectra are larger than that in band-gap PL spectra. In order to reduce the error due to the compositional inhomogeneity of the samples, we focused the laser beam on the center of the sample during the PL measurement. The uncertainty of the x-ray compositional measurements is less than 10%, which depends on the Ga content. The larger the Ga content, the more dislocations are found in the epilayer, giving broader x-ray diffraction peaks and increasing the uncertainty. From the above analysis and taking account of the deviation of the least-squares fit, the accuracy estimates of $\sim$12% for band-gap energies and $\sim$15% for deep-level energies are conservative.

IV. CONCLUSIONS

Using PL and PC spectroscopy, we have studied the deep levels in $Ga_xIn_{1-x}P$ epilayers as a function of composition x on InP:Fe or InP:S substrates and compared them to the near-band-edge PL emissions. The deep-level PL and PC spectra consist of broad bands. From a detailed investigation of the temperature, alloy composition, and substrate dependence of these features, we have tentatively identified these deep levels. The positions and PL intensities of the broad bands depend not only on alloy composition, but also on the substrate. We propose that the impurities in the substrate diffuse into the epilayers during MOCVD growth forming impurity-vacancy complexes. The broad bands in the $Ga_xIn_{1-x}P$/InP:Fe are due to nearest-neighbor molecular-like complex defects formed with substitutional iron in either gallium or indium sites together with phosphorus vacancies $[(V)_P-(Fe)_{III}]$. The $(V)_{III}$-$(S)_P$ complex is the origin of the broad bands in the PL spectra of $Ga_xIn_{1-x}P$/InP:S.

A least-squares fit of the experimental data as a function of alloy composition allows us to compare the deep-level and band-gap energies. We find that the deep levels are independent of the conduction band edges, and the composition dependence of the deep levels associated with such complexes depends on the site occupied by the impurity atom.

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