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Temperature dependent polarized-piezoreflectance study of GaInP

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Abstract. We report a detailed temperature dependent study of polarized piezoreflectance (PzR) for two GaInP epilayers that exhibit different degrees of ordering in the range between 25 and 500 K. The polarized PzR measurements showed anisotropic character along the [110] and $[1\bar{1}0]$ directions for the ordered sample. The PzR spectra were fitted using the first derivative of a Lorentzian line shape functional form. The direct band-to-band, crystal-field splitting to band and spin-orbit splitting to band transition energies which are denoted as E_0 , $E_0 + \Delta_c$ and $E_0 + \Delta_s$ respectively, at various temperatures, were accurately determined. The temperature dependences of these near band-edge critical point transition energies were analysed by the Varshni expression and an expression containing the Bose–Einstein occupation factor for phonons. The parameters that describe the temperature variation of the transition energies are evaluated and discussed.

1. Introduction

Ga_{0.5}In_{0.5}P (hereafter referred to as GaInP) is a wide band-gap semiconductor grown lattice matched to GaAs. The material is of interest for a variety of device applications such as light emitting diodes [1], semiconductor injection lasers [2, 3], heterojunction bipolar transistors [4, 5] and high electron mobility transistors [5, 6]. GaInP layers grown by organometallic vapour phase epitaxy (OMVPE) typically form the CuPt ordered structure. The Ga and In atoms spontaneously segregated into alternating {111} monolayers during growth, rather than forming a disordered alloy with Ga and In atoms randomly distributed on the group III sublattice. The effects of ordering on the optical band gap of GaInP consist of two major features, namely (1) the band gap E_0 is reduced and (2) at the Γ point, the fourfold degenerate valence-band maximum for the disordered GaInP is split with a crystal-field energy Δ_c [7–11]. The ordering induced shift of the band gap (ΔE_0) and Δ_c are quadratic functions of the long-range order parameter η [10, 11]. The temperature dependence of the energy gap E_0 of GaInP has been investigated by electroreflectance (ER) [12], piezoreflectance (PzR) [13], photoluminescence (PL) [8, 12, 14–16] and excitation photoluminescence (PLE) [16–18]. In most works, the temperature range is restricted to within 4.2 to 300 K. To date no available works can be found for the material at temperature higher than 300 K. In order to have a better understanding of

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the temperature profile of the near band-edge critical point transition features, it would be desirable to have a systematic study of the features over an extended temperature range.

In this paper, we report a detailed temperature dependent study of the polarized PzR spectra for two GaInP epilayers that exhibit different degrees of ordering in the range between 25 and 500 K. In contrast to PL measurements, where at low temperatures there is a selective preference for transitions from conduction band states closer to the valence bands, PzR measurements are sensitive only to the critical points in the joint density of states and are independent of the occupation probability factors [19]. PzR is a technique suitable for this study because it provides sharp first derivative signatures of the relevant transitions and can be applied over a wide temperature range. For the ordered sample, the PzR spectra showed strong polarization dependence characteristics. The PzR spectra were fitted using the first derivative of a Lorentzian line shape function form [20, 21]. By using polarization selection rules, the energies of the direct band-to-band transition, E_0 , crystal-field splitting to band transition, $E_0 + \Delta_c$, and spin-orbit splitting to band transition, $E_0 + \Delta_s$, were determined accurately. The temperature dependences of the near band-edge critical point transition energies were analysed by the Varshni expression [22] and an expression containing the Bose-Einstein occupation factor for phonons [23]. The parameters that describe the temperature variation of the transition energies were evaluated and discussed.

2. Experimental details

The samples of GaInP used in this study were grown on (001) 2° and 15° misoriented Si-doped GaAs substrates by an Aixtron OMVPE system. Trimethylgallium and trimethylindium were used as the column III metalalkyl sources. Phosphine was used as P source. The growth temperature T_g was 670°C and the V/III ratio was kept at a constant value of 200. The layer thickness was $\sim 0.7\ \mu\text{m}$ after 30 min growth. The lattice mismatch between the GaInP epilayer and the GaAs substrate, as checked by double crystal x-ray diffraction, was less than 10^{-3} .

The PzR measurements were achieved by gluing the thin sample on a 0.15 cm thick lead zirconate titanate (PZT) piezoelectric transducer driven by a 200 V_{rms} sinusoidal wave at 200 Hz. In order to enhance the modulation effects, the substrate of the sample was polished with successively finer grades of silicon carbide grinding paper to a thickness of about 100 μm . The alternating expansion and contraction of the transducer subjected the sample to an alternating strain with a typical rms $\Delta l/l$ value of $\sim 10^{-5}$. A 150 W tungsten-halogen lamp filtered by a model 270 McPherson 0.35 m monochromator provided the monochromatic light. The reflected light was detected by an EG&G type HUV-2000B silicon photodiode, and the signal was recorded from an NF model 5610B lock-in amplifier. The Oriel visible-near infrared dichroic linear polarizers were employed for polarization dependent measurements. An RMC model 22 closed-cycle cryogenic refrigerator equipped with a model 4075 digital thermometer controller was used for low temperature measurements. For high temperature experiments the PZT was mounted on one side of a copper finger of an electrical heater, which enable us to stabilize the sample temperature. The measurements were made between 25 and 500 K with a temperature stability of 0.5 K or better.

3. Results and discussion

Figure 1 shows the PzR spectra of the sample with substrate misorientation of 15° for $\mathbf{E} \parallel [110]$ and $\mathbf{E} \parallel [1\bar{1}0]$ at 300 K. The crystal-field splitting between the two polarization directions is very small ($\sim 2\ \text{meV}$), indicating a degree of disordering of the sample. Figure 2(a) shows

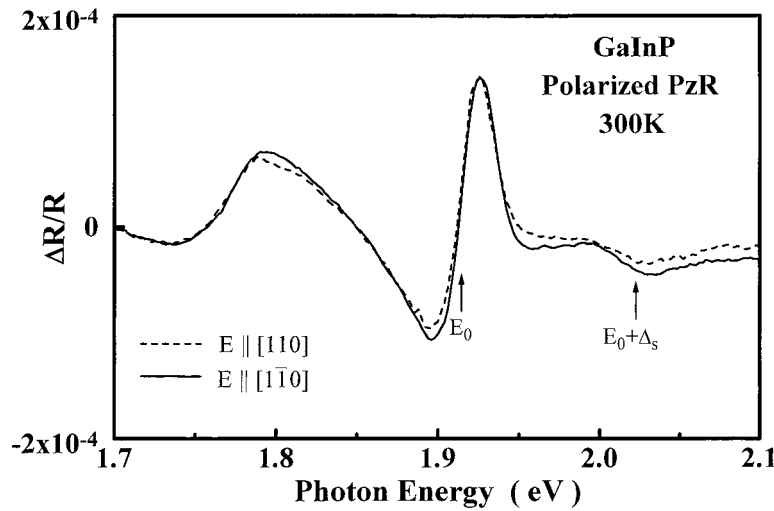


Figure 1. The PzR spectra of a highly disordered GaInP sample at 300 K with the incident light polarized along $E \parallel [110]$ (dashed curve) and $E \parallel [1\bar{1}0]$ (solid curve) directions.

the PzR spectra of the sample grown with 2° misorientation substrate for $E \parallel [110]$ and $E \parallel [1\bar{1}0]$ polarizations. The strongly polarization-dependent dominant features near 1.85 eV correspond to transitions with energy E_0 between the top of the valence band and the bottom of the conduction band, and transitions with energy $E_0 + \Delta_c$ between the crystal field band and the bottom of the conduction band. In order to determine the transition energies accurately, we have performed a theoretical line shape fit (see figures 2(b) and 2(c)). The functional form used in the fitting procedure corresponds to a first derivative Lorentzian line shape function of the form [20, 21]

$$\frac{\Delta R}{R} = \text{Re} \sum_{j=1}^m A_j e^{i\Phi_j} (E - E_j + i\Gamma_j)^{-n} \quad (1)$$

where m is the number of critical points, A_j and Φ_j are the amplitude and phase of the line shape, E_j and Γ_j are the energy and broadening parameter of the transitions and the value of n depends on the origin of the transitions. For the first derivative functional form, $n = 2$ is appropriate for the excitonic transitions. For M_0 type three-dimensional critical point interband transitions, $n = 0.5$ is appropriate [20, 21]. Our experimental signatures for E_0 and $E_0 + \Delta_c$ are more consistent with excitonic line shape ($n = 2$) while the signature for $E_0 + \Delta_s$ has a better fit with $n = 0.5$. We emphasize, however, that the transition energies extracted from such fits are relatively insensitive to the line shape function.

As shown in figure 1, for the highly disordered sample, the dominant feature near $E_0 = 1.916 \pm 0.003$ eV can be associated with the transition from the top of the valence band to the bottom of the conduction band. This value agrees very well with the recent report of 1.915 eV for the disordered sample [24], while the signature at higher energy side located around 2.023 ± 0.003 eV corresponds to the transition between the top of the spin-orbit split-off band and the bottom of the conduction band. Both polarization spectra show similar results. The measurements give a spin-orbit splitting of $\Delta_s = 107 \pm 6$ meV for the highly disordered sample. The polarization dependent PzR spectra for the 2° misoriented partially ordered sample are shown in figure 2(a). As shown in figure 2(b), the $E \parallel [110]$ polarization yields a strong E_0 transition (points) and a weaker $E_0 + \Delta_c$ transition (dashed curve). This is consistent

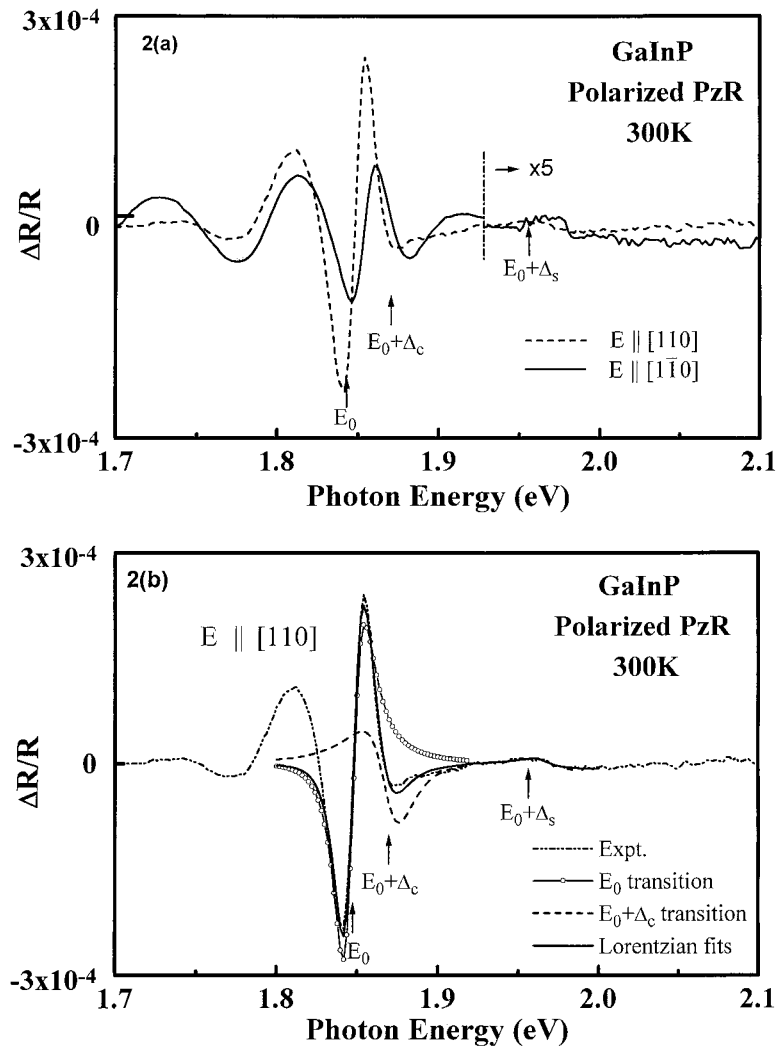


Figure 2. (a) The PzR spectra of a partially ordered GaInP sample at 300 K for $E \parallel [110]$ (dashed curve) and $E \parallel [1\bar{1}0]$ (solid curve) directions, respectively. (b) The $E \parallel [110]$ polarization shows a strong E_0 transition (points) and a weaker $E_0 + \Delta_c$ transition (dashed curve), while (c) $E \parallel [1\bar{1}0]$ polarization yields the $E_0 + \Delta_c$ transition (dashed curve) comparable to the E_0 transition (dotted curve).

with polarization selection rules, which indicate that the intensity of the E_0 signature is about three times stronger than that of the $E_0 + \Delta_c$ signature [10]. On the other hand, the $E \parallel [1\bar{1}0]$ polarization (figure 2(c)) yields the $E_0 + \Delta_c$ transition (dashed curve) comparable to the E_0 transition (dotted curve). The result might indicate that the contribution to the PzR signatures from the momentum-matrix elements associated with these two transitions is roughly the same for light polarized along the $[1\bar{1}0]$ direction. Finite uncertainty in the determination of E_0 also results from the fitting to the complex line shape at the lower-energy side of the E_0 feature, where the spectral structure related to the band-to-band transitions overlaps with the interference oscillations. These oscillations at energies below the E_0 transitions are mainly

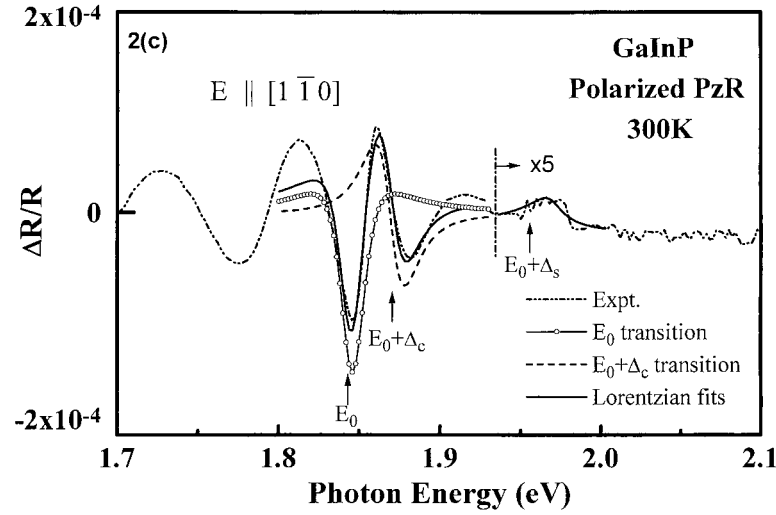


Figure 2. (Continued)

due to interference effects caused by the heterointerface. The deviation of the direct band-to-band transition energy obtained by using different amplitude and phase parameters in the fittings was found to be around 3 meV at room temperature. The transition energies for E_0 and $E_0 + \Delta_c$ were determined to be 1.845 ± 0.003 and 1.869 ± 0.003 eV, respectively. The energy difference between these two signatures yields a crystal-field splitting of $\Delta_c = 24 \pm 6$ meV. The crystal-field splitting is a clear indication of the existence of a significant degree of CuPt ordering. The feature at around 1.955 ± 0.003 eV corresponds to transition involving the spin-orbit split-off band of the GaInP. Another effect of ordering is the band gap reduction. The room temperature results indicate that the E_0 of the partially ordered sample is 70 meV lower than that of the highly disordered one.

The direct experimental measurements of the degree of ordering η are limited [25]. A theoretical relationship between η and the transition energies E_0 and Δ_c has been calculated for ordering in III-V alloys [26]. The ordering parameter η of the 2° misoriented sample was estimated to be 0.47 by using the following relation

$$\eta^2 = \frac{E_0^{(\eta=0)} - E_0^{(\eta)}}{\Delta E_0^{(\eta=1)}} = \frac{\Delta E_0^{(\eta)}}{\Delta E_0^{(\eta=1)}}. \quad (2)$$

Here, we used $E_0^{(\eta=1)} = 1.916$ eV for the band gap of a random alloy ($\eta = 0$) and $\Delta E_0^{(\eta=1)} = 310$ meV as predicted by Wei and Zunger [26].

Displayed by the dotted curves in figure 3 is the PzR spectra in the vicinity of E_0 and $E_0 + \Delta_s$ for the highly disordered GaInP sample and figures 4(a) and 4(b) are the polarized PzR spectra, $E \parallel [110]$ and $E \parallel [1\bar{1}0]$ respectively, in the vicinity of E_0 , $E_0 + \Delta_c$ and $E_0 + \Delta_s$ for the partially ordered GaInP sample for several temperatures between 25 and 500 K. For the highly disordered sample only unpolarized spectra are shown. The line shapes of the PzR spectra do not change significantly as a function of temperature. This is in contrast to PL measurements, where the low-temperature spectra favour the lowest energy transition and emission from the more ordered domains [8, 16]. When the separation between E_0 and $E_0 + \Delta_c$ transitions is less than the experimental linewidth, the usual line shape fit presents large ambiguity in the determination of the splitting energy. We can resolve this difficulty by using polarization

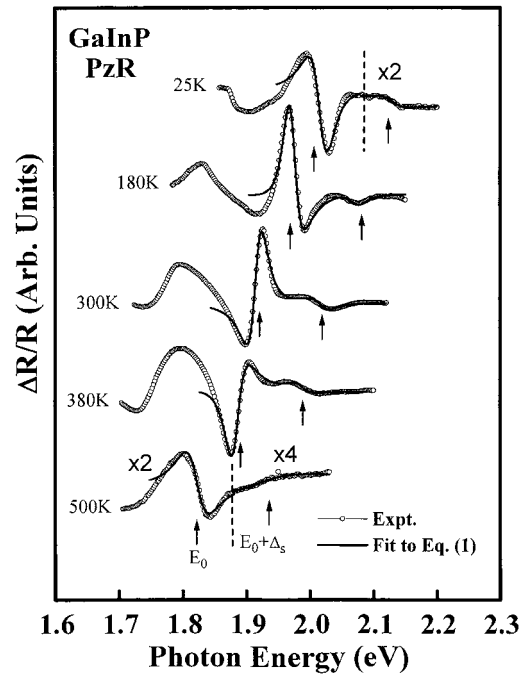


Figure 3. The experimental PzR spectra (points) in the vicinity of E_0 and $E_0 + \Delta_s$ transitions of a disordered GaInP epilayer at 25, 180, 300, 380 and 500 K, respectively. The full curves are least-squares fits to the first derivative of a Lorentzian profile, which yields the near band-edge critical point transition energies indicated by arrows.

selection rules to determine the relative intensity of the two features for both polarizations. These intensity relations have been used in the fitting procedure to obtain a better estimate of Δ_c . The obtained values of E_0 , $E_0 + \Delta_c$ and $E_0 + \Delta_s$ are indicated by arrows in the figures. As expected for semiconductors, the near band-edge critical point transition energies show red shifts with increasing temperature.

The temperature dependences of the E_0 , $E_0 + \Delta_c$ and $E_0 + \Delta_s$ transitions are shown in figure 5. The fitted results for the highly disordered and partially ordered GaInP samples over the temperature range of 25 to 500 K are indicated respectively, by solid circles and open circles in figure 5. Representative error bars are shown. From the results of figure 5, we note the occurrence of the band-gap reduction and the crystal-field splitting in the partially ordered sample. Also noted, within experimental error, the crystal-field splitting and the spin-orbit splitting do not depend on temperature. The full curves are the least-squares fits to the Varshni semi-empirical relationship [22]

$$E_{c_i}(T) = E_{c_i}^V(0) - \frac{\alpha T^2}{T + \beta}. \quad (3)$$

Here, $E_{c_i}^V(0)$ is the critical point transition energy (E_0 , $E_0 + \Delta_c$ or $E_0 + \Delta_s$) at absolute zero; α and β are constants referred to as Varshni coefficients. The constant α is related to the electron-phonon interaction and β is closely related to the Debye temperature [22]. The obtained values of $E_{c_i}^V(0)$, α and β for the near band-edge critical point transitions are listed in table 1.

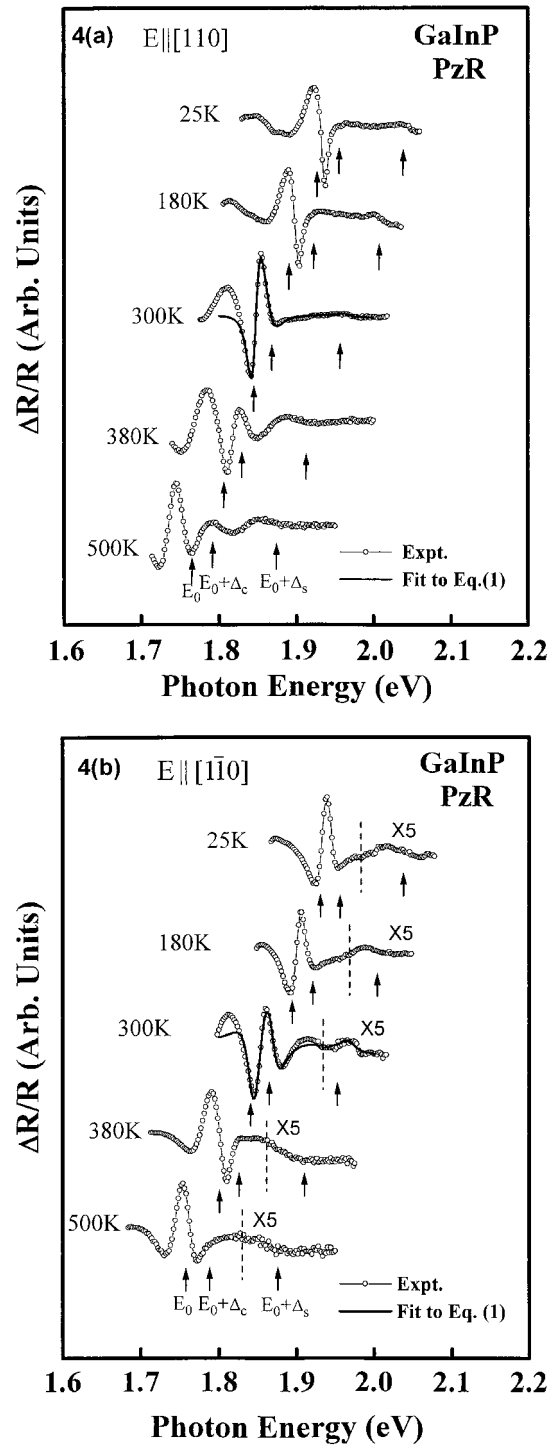


Figure 4. The experimental PzR spectra (points) of a partially ordered GaInP sample at 25, 180, 300, 380 and 500 K for (a) $E \parallel [110]$ and (b) $E \parallel [\bar{1}10]$ directions, respectively. The full curves are representative least-squares fits to the first derivative of a Lorentzian profile at 300 K. The fitted values of the near band-edge critical point transition energies are indicated by the arrows.

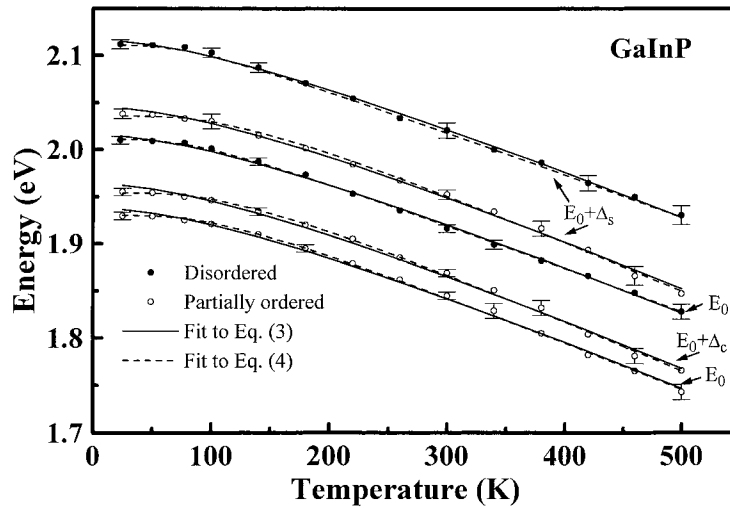


Figure 5. The temperature dependence of E_0 , $E_0 + \Delta_c$ and $E_0 + \Delta_s$ in highly disordered (solid circles) and partially ordered (open circles) GaInP. Representative error bars are shown. The full curves are least-squares fits to equation (3) and the dashed curves are least-squares fits to equation (4).

The temperature dependence of the near band-edge critical point transitions of GaInP can also be described by a Bose–Einstein-type expression of the form [23]

$$E_{ci}(T) = E_{ci}(0) - \frac{2a_B}{[\exp(\Theta_B/T) - 1]} \quad (4)$$

where $E_{ci}(0)$ is the near band-edge transition energy at 0 K, a_B represents the strength of the electron–average phonon interaction and Θ_B corresponds to the average phonon temperature. Shown by the dashed curves in figure 5 is a least-squares fit to equation (4). The obtained values for the various parameters are also listed in table 1.

Included in table 1 are the relevant parameters as determined by fitting the experimental ER [12], PzR [13], PL [15] and PLE [16] spectra for GaInP using equations (3) and (4). The ordering parameter η was estimated by using the relationship shown in equation (2). It is noted that except for [15], all the other works did not give the relevant fitted values of α , β , a_B and Θ_B . The values we have listed here were obtained from the best least-squares fits of the published data of these works. Hence, no estimations of error margins are given for these fitted parameters since we have no knowledge of the experimental errors of their published data.

From a close examination of the parameters in table 1, the values of β as deduced from different measurement techniques are quite similar over all temperature range. The value of α depends on measurement techniques and temperature range. It seems that most of the values of α from other works fall below 0.5 except those of [15] by 't Hooft *et al* where PL measurement technique is used. However low temperature PL spectra were reported to be a rather poor technique for the determination of the band gap of the ordered GaInP [16]. Hence the values extracted from PL measurements of [15] may not be reliable in comparing with other measurement techniques. The low value of α (<0.5) as deduced from other works is probably due to the limited temperature range of these works. In addition, Varshni's relation may not be strictly applicable at low temperature. These together with the smaller variation of the band gap at lower temperature introduce a larger uncertainty for the determination of Varshni coefficients. In view of the aforementioned argument, we are quite confident that the

Table 1. Values of the Varshni- and Bose-Einstein-type fitting parameters, which describe the temperature dependence of GaInP with various degrees of ordering.

Sample condition	Feature	$E_{c_i}^V(0)$ (eV)	α (meV K ⁻¹)	β (K)	a_B (meV)	Θ_B (K)	Temperature range (K)
disordered	E_0^a	2.011 ± 0.003	0.52 ± 0.07	190 ± 100	56 ± 14	237 ± 50	
	$E_0 + \Delta_s^a$	2.115 ± 0.003	0.52 ± 0.07	190 ± 100	50 ± 15	220 ± 50	
ordered ($\eta^b = 0.47$)	E_0^a	1.933 ± 0.003	0.53 ± 0.07	205 ± 100	62 ± 15	255 ± 50	25–500
	$E_0 + \Delta_c^a$	1.953 ± 0.003	0.55 ± 0.07	205 ± 100	74 ± 17	289 ± 50	
	$E_0 + \Delta_s^a$	2.038 ± 0.003	0.54 ± 0.07	205 ± 100	75 ± 17	296 ± 50	
disordered	E_0^c	1.988	0.42	198	57	272	
	$E_0 + \Delta_s^c$	2.093	0.45	205	56	263	
ordered ($\eta^b = 0.48$)	E_0^c	1.930	0.49	255	66	288	6–250
	$E_0 + \Delta_c^c$	1.948	0.47	208	68	285	
	$E_0 + \Delta_s^c$	2.036	0.45	211	64	283	
disordered	E_0^d	2.011	0.56	204			4.2–300
ordered ($\eta^b = 0.46$)	E_0^d	1.937	0.612	204			
weakly ordered ($\eta^b = 0.30$)	E_0^e	1.973	0.5	212	65	263	4.2–200
weakly ordered ($\eta^b = 0.31$)	E_0^e	1.968	0.43	218	61	274	4.2–200
ordered ($\eta^b = 0.58$)	E_0^e	1.897	0.49	239	69	270	4.2–150
ordered ($\eta^b = 0.53$)	E_0^f	1.920	0.41	215	51	263	8–200

^a Present work (polarized piezoreflectance).

^b The ordering parameter was estimated by using equation (2).

^c [13] (polarized piezoreflectance).

^d [15] (polarized photoluminescence).

^e [16] (photoluminescence excitation).

^f [12] (electroreflectance).

values of α and β of the near band-edge critical point transitions as determined in this work over an extended temperature range should offer a much better accuracy.

The parameter α of equation (3) can be related to a_B and Θ_B of equation (4) by taking the high-temperature limit of both expressions. This yields $\alpha = 2a_B/\Theta_B$. Comparison of the numbers shown in table 1 shows that this relation is indeed satisfied. As shown in table 1, our values for a_B and Θ_B are in good agreement with those of earlier works. The temperature shift of interband transition energies is mainly due to the interactions of electrons with relevant acoustic and optical phonons. According to the existing theory this leads to a value of Θ_B significantly smaller than the average LO-phonon temperature Θ_{LO} . The value of $\Theta_{LO} \approx 530$ K is deduced from the Raman spectra of GaInP [27, 28]. From table 1, it can be seen that our fitted value of Θ_B satisfied this theoretical trend.

4. Summary

A detailed temperature dependent polarized PzR study of two GaInP epilayers that exhibit different degrees of ordering was carried out in the range of 25 to 500 K. The polarized PzR

measurements showed anisotropic character along the [110] and $[1\bar{1}0]$ directions for the ordered sample. Using the polarization selection rules and the first derivative Lorentzian line shape functional form, the transition energies of E_0 , $E_0 + \Delta_c$ and $E_0 + \Delta_s$ at various temperatures were accurately determined. The temperature dependences of these near band-edge critical point transition energies were analysed by both the Varshni- and Bose–Einstein-type expressions. The parameters that describe the temperature variation of the transition energies were evaluated. The results indicate that within experimental error, the numbers do not depend on the ordering parameter.

Acknowledgments

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