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Photoreflectance studies of temperature and hydrostatic pressure dependencies of direct optical transitions in BGaAs alloys grown on GaP

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Abstract
BGaAs layers with boron concentrations of 4.1%, 7.4%, and 12.1% are grown by molecular beam epitaxy on a GaP substrate and studied by optical absorption and photoreflectance (PR) spectroscopy with both temperature and hydrostatic pressure dependence. The direct optical transitions from the bands composing the valence band—namely heavy-hole, light-hole, and spin–orbit split-off—to the conduction band are clearly observed in the PR spectra. For the abovementioned optical transitions, their temperature dependencies are obtained in the range from 20 K to 300 K, and analyzed by Varshni and Bose–Einstein relations. Furthermore, the BGaAs alloys are also studied with hydrostatic pressure up to ∼18 kbar, revealing pressure coefficients of direct optical transitions. The obtained results are discussed within the framework of the band anticrossing model and chemical trends.

Keywords: BGaAs, photoreflectance, hydrostatic pressure, heavy/light-hole transition, spin–orbit splitting

(Some figures may appear in color only in the online journal)

1. Introduction

III–V semiconductor alloys diluted with boron are still one of the least studied III–V compound semiconductors despite increasing interest in recent years. This situation arises from the challenging synthesis of good quality crystals, limiting the achievable boron concentration. For BGaAs alloys, i.e. the most intensively studied boron-based III–V compounds, the majority of previous studies report on the incorporation of boron atoms below 9% [1–14]. For the enhancement of the mentioned boron incorporation on lattice sites during growth by molecular beam epitaxy (MBE), Ptak et al used, for the first time, bismuth atoms as a surfactant [11]. Such an approach allowed BGaAs alloys to be synthesized with boron concentrations up to 17.7%, as reported in [15], together with the optical study results. In that work, based on photoreflectance (PR) measurements, a large band gap

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bowing parameter \((b = 3.6\ \text{eV})\) was obtained, which is consistent with the value of 3.5 eV determined within density functional theory (DFT) calculations \([15]\). Similar values, i.e. 3.39 and 3.5 eV, of the band gap bowing parameter were derived, respectively, by a phenomenological model \([16]\) and the linearized augmented plane wave method \([17]\). It is also worth noting that the band gap changes due to boron incorporation are negligible for boron content below 10\% \([4, 15, 17]\). Moreover, the substitution of gallium with boron atoms leads to the lattice contraction of host materials, which is well established \([2, 5, 9, 11, 14, 18]\). The decrease of lattice constant and insignificant change of energy gap with increasing boron concentration makes BGaAs a perfect candidate for strain engineering, particularly for tensile strain compensation \([19]\).

To date, only photoluminescence (PL) spectroscopy has been applied to study the temperature evolution of optical properties of boron-based structures—BGaAs/GaAs epilayers \([7, 10, 13, 20, 21]\), BGaAs/GaAs single quantum wells (SQWs) \([22, 23]\), and BInGaAs/GaAs SQWs \([24]\). In all of these works, the authors reported the S-shape temperature dependence of PL peak energy, which is typical for highly mismatched alloys (HMAs) \([25–28]\). Due to significant differences in the size and electronegativity of B and Ga atoms, the BGaAs compound can be included in the group of HMAs. The observed S-shape in the temperature dependence of PL peak energy for a boron-based structure arises from the presence of localized states, the source of which can be B–B pairs, clusters of B atoms, and B content inhomogeneities \([3, 8, 11, 21, 29]\). Such imperfections in the crystal lattice stem from the challenging synthesis of B,III–V semiconductors crystals, and other HMAs \([5, 11, 30–32]\). Due to the carrier-localization effect, it is impossible to obtain the temperature dependence of band-to-band optical transitions of boron-based III–V alloys relying on emission-like measurements. For that purpose, PR, which is an absorption- and differential-like technique, is a perfect tool to probe transitions between delocalized states and determine their temperature evolution \([33, 34]\). Furthermore, most of the previously studied boron-based structures were deposited on GaAs, for which the optical features from that substrate can overlap with the signal originating from BGaAs or BInGaAs layers.

In the case of studies of optical transitions with hydrostatic pressure, it was shown, based on photomodulated transmission measurements, that the pressure coefficient of the energy gap for B,Ga\(_{1-x}\)As \((x = 1.2\%–2.3\%)\) is comparable to GaAs \([4]\). Additionally, the pressure dependency of the band gap is almost linear, which may indicate the lack of a strong anticrossing effect in the BGaAs alloy and imply that this alloy can be treated as conventional III–V compound semiconductors up to a few percent of boron \([4]\). However, it is unknown whether the pressure dependence of optical transitions remains linear at a higher boron concentration, for which the strength of the coupling between the boron level and the conduction band of the host material can be more substantial.

In this work, we report PR studies of BGaAs layers, with boron concentrations of 4.1\%, 7.4\%, and 12.1\% grown by MBE on a GaP substrate. We do not expect any PR signal to superimpose on the BGaAs features for that substrate, which could happen for layers deposited on GaAs or other common III–V group substrates. The observed optical transitions, namely from the heavy hole (HH), light hole (LH), and spin–orbit split-off (SO) to the conduction band, were measured in the 20 K–300 K temperature range. The behavior of fundamental transition with temperature was also studied by optical absorption, where usually only one absorption edge can be determined, and additional effects like substrate defect state can distort features related to band structure. Moreover, we have investigated the influence of hydrostatic pressure (up to \(\sim 18\) kbar) on the energy of HH, LH, and SO transitions.

2. Experimental details

The studied BGaAs layers \((\sim 100\) nm\) were grown on a GaP (001) substrate, the temperature of which was maintained at 400°C, by solid-state MBE (with an EPI Mod Gen II MBE system). The growth rate of all BGaAs samples was kept at \(\sim 1.8–2.0\) Å s\(^{-1}\), with a Ga beam equivalent pressure (BEP) of \(1.67 \times 10^{-7}\) Torr and Ga:As BEP ratio \(\sim 18\). The deposition of boron in the MBE chamber by vertical electron beam evaporator was assisted by a Bi overpressure ambient \((5 \times 10^{-8}\) Torr\) to promote the incorporation of boron on the lattice sites. Furthermore, to obtain the boron concentration, high-resolution x-ray diffraction \(\omega–2\Phi\) measurements were performed. Details of the growth procedure and structural characterization were reported in \([15]\).

Optical absorption of the studied alloys was depicted as an optical density \((\text{O.D.})\) calculated on the base of the expression \(\text{O.D.} = \log_{10}(1/T)\). The transmission \(T\) in the denominator of that expression was determined by dividing spectra obtained for the sample by the reference spectra measured through a hole in the same holder. Both of the mentioned spectra were measured by transmitting light from a 150 W tungsten-halogen lamp dispersed by a 300 mm focal length monochromator (with a resolution of the order of 0.2 nm). A silicon photodiode was used to detect the beam intensity. The measurements were performed using the lock-in technique, with a mechanical chopper modulating the light source. For temperature-dependent experiments, samples were attached to a cold finger of a closed-cycle helium cryocooler with a conductive silver paste.

The PR spectra were measured in the so-called ‘bright configuration’ experimental setup \([35]\) with a 150 W tungsten-halogen lamp (probe beam) and continuous-wave laser 405 nm line (pump beam). We used a single grating 0.5 m focal-length monochromator (with a resolution of \(\sim 0.12\) nm) and Si pin photodiode to disperse and detect the probe beam reflected from the sample. The weak changes of the reflectivity coefficient, corresponding to optical transitions, were measured by a lock-in amplifier with a 280 Hz reference frequency, at which the pump beam was modulated. The mentioned lock-in amplifier resolves both signal components, i.e. the AC and DC proportional to \(\Delta R\) and \(R\), respectively. The DC component of the photodetector signal (representing the reflectivity of the sample) was measured with an auxiliary analog-to-digital
converter input of the lock-in amplifier. In the last step, the software performs an operation of the division of the AC by the DC component. To obtain the temperature dependence of the PR spectra, the same procedure as for the optical absorption measurements was used. The measurements of the PR spectra with hydrostatic pressure were performed with a sample mounted on a plug located inside a piston pressure cell equipped with a sapphire window for optical access [36]. The increment of pressure, determined by changes in the resistance of an InSb sensor, was achieved by an inward movement of the piston.

3. Results and discussion

The PR spectra of the BGaAs samples with various boron concentrations, namely 4.1%, 7.4%, and 12.1%, obtained at 20 K and 300 K, are shown in figure 1, together with the results of PR measurements for the bare GaP substrate. It is visible that, in the studied spectral range, the GaP substrate does not give any PR signal, which could superimpose on features originating from BGaAs layers. In contrast, for BGaAs samples, we observed three optical transitions designated as HH, LH, and SO of which nature we have identified in our previous studies [15]. The transitions from heavy and LH bands were split and were consequently distinguishable, especially for the sample with 4.1% B content, due to the presence of compressive strain in the BGaAs layer. Moreover, an increase of boron concentration caused a blueshift of the HH transition and a decrease of the splitting between HH and LH transitions because of reduced lattice mismatch. On the other hand, the SO transition observed for the B_{0.041}Ga_{0.959}As sample at ~2.01 eV redshifts with increasing boron content. All transitions described here, represented by resonances in the PR spectra, were fitted by the Aspnes formula [37]:

\[
\frac{\Delta R}{R}(E) = \text{Re} \left[ \sum_{j=1}^{n} C_j e^{\theta_j} (E - E_j + i\Gamma_j)^{-m_j} \right],
\]

where \( \Delta R/R(E) \) is the energy dependence of the PR signal and \( n \) is the number of transitions to be fitted. Parameters \( C \) and \( \theta \) are the amplitude and the phase of the resonance, whereas, \( E_j \) and \( \Gamma_j \) are the energy and the broadening of the optical transitions, respectively. The \( m \) parameter depends on the type of transition, and for one-electron transition at a three-dimensional critical point (band-to-band transition), we have taken a value equal to 2.5 [38]. The moduli of each PR resonance (red, blue, and green lines in figure 1) have been calculated according to equation (2), with parameters obtained from the fit by Aspnes formula equation (1):

\[
\Delta \rho_j(E) = \frac{|C_j|}{(E - E_j)^2 + \Gamma_j^2}^{m_j/2}.
\]

Comparing the moduli of optical transitions, it is clear that the HH transition is the strongest. Furthermore, the HH transition significantly overlaps with the LH one in the case of the B_{0.121}Ga_{0.879}As sample, making them indistinguishable. That situation originates from decreased splitting between HH and LH bands along with an increase in the broadening of these transitions. The observed PR resonances broaden for the sample with higher boron content due to increased alloy inhomogeneities.

Optical absorption spectra for the studied compounds, shown in figures 2(a)–(c), reveal broad absorption onset red-shifting with temperature, as expected for most semiconductors. The visible increase (in the energy range 1.72–1.8 eV) of the optical density for the samples with a 7.4% and 12.1% boron concentration can be associated with decreased splitting of the HH and LH bands and decreased band curvature of the conduction band resembling the valence band more closely. The energy of a direct allowed transition was determined by extrapolating a linear part of a Tauc plot, of the form \( (\alpha\hbar\omega)^2 \) versus \( \hbar\omega \), where \( \alpha \) represents the absorption coefficient calculated from the optical density as \( \alpha = (\text{O.D.}\log(10))/l \). Furthermore, \( l \) is the length of the absorption medium. To analyze the temperature evolution of the absorption edge presented in figures 2(d)–(f), we fit the energy dependencies by empirical Varshni relation [39] and Bose–Einstein formula, which takes into account electron–phonon interaction [40, 41]. The former relation is given by:

\[
E_0(T) = E_0(0) - \frac{\alpha T^2}{\beta + T},
\]

where \( E_0(0) \) is the energy of the transition at \( T = 0 \) K, whereas \( \alpha \) and \( \beta \) are the so-called Varshni empirical coefficients. The latter formula, i.e., Bose–Einstein, can be written in the form:

\[
E_0(T) = E_0(0) - \frac{2\alpha_0}{\exp \left( \frac{2\alpha_0}{T} \right) - 1},
\]

where \( \alpha_0 \) is the strength of the electron-average phonon interaction and \( \Theta_B \) is the average phonon temperature. Parameters obtained from the fitting procedure, by equations (3) and (4), are summarized in table 1, together with literature data for GaAs. For the 12.1% compound, there is an abrupt change in the determined absorption edge energy, which coincides with a blueshift of the absorption spectra (yellow lines) for temperatures >180 K, followed by an expected redshift. The origin of this feature remains unknown, but its presence has been confirmed by measuring two different samples on two separate setups. For that reason, only experimental points up to 160 K were used for the fitting procedure. That explains relatively large uncertainties of the determined parameter values and their differences when compared to 4.1% and 7.4% alloys. Unlike PR, optical absorption does not allow us to resolve particular optical transitions (HH, LH, and SO). Since the obtained spectra are cumulative, several different absorption processes result in a broad absorption edge. The accuracy of separating the transitions would be intrinsically limited by weak light extinction in the transmission-based experiment for the studied thin layers. In addition, the contribution of defect states from the thick substrate in optical absorption cannot be completely ruled out.
Figure 1. PR spectra (black lines) together with the results of the fitting procedure by equation (1) (gray lines) of GaP reference and BGaAs samples with different boron concentrations measured at (a) 20 K and (b) 300 K. Moreover, at the bottom part of every panel we have plotted the moduli of optical transitions (red, blue, and green lines) obtained by equation (2).

Figure 2. Optical absorption spectra of the BGaAs alloys with (a) 4.1%, (b) 7.4%, and (c) 12.1% boron concentrations measured in the 10 K–300 K range along with obtained temperature dependence of the absorption edge (d)–(f). Solid lines represent fitted curves. The cause of the visible abrupt change of energy for the sample with 12.1% boron content (f) remains unknown but was confirmed for different samples measured on two experimental setups.

For all three samples, we performed PR measurements with temperatures from 20 K to 300 K (see figure 3), which allows us to determine the temperature evolution of optical transitions between delocalized states since the PR technique is insensitive to localized states. It is apparent that all observed transitions redshift and broaden with increasing temperature. We have analyzed both HH and LH transitions only for the sample with the smallest boron content, for which these transitions remained distinguishable in the whole temperature range. Due to the very low amplitude of the SO transition, we have omitted its investigation for the $\text{B}_{0.121}\text{Ga}_{0.879}\text{As}$ sample. The energies of optical transitions were extracted from the fitting procedure.
Table 1. Varshni and Bose–Einstein parameters extracted from the fit by equations (3) and (4) of temperature dependencies of the energy of optical transitions observed for the $B_xGa_{1-x}$As samples.

| Transition  | B content (%) | $\alpha$ ($10^{-4}$ eV K$^{-1}$) | $\beta$ (K) | $\alpha_B$ (meV) | $\Theta_B$ (K) |
|-------------|---------------|----------------------------------|-------------|-----------------|----------------|
| $E_0$ [49]  | 0             | 5.405                            | 204         | 57              | 240            |
| Absorption  | 4.1           | $4.51 \pm 0.13$                  | 204 (fixed) | 53 $\pm$ 8      | 260 $\pm$ 26   |
|             | 7.4           | $4.57 \pm 0.10$                  | 204 (fixed) | 46 $\pm$ 5      | 233 $\pm$ 18   |
|             | 12.1$^a$      | $4.78 \pm 0.26$                  | 204 (fixed) | 36 $\pm$ 12     | 188 $\pm$ 37   |
| HH          | 4.1           | $4.97 \pm 0.16$                  | 204 (fixed) | 51 $\pm$ 7      | 233 $\pm$ 19   |
|             | 7.4           | $4.88 \pm 0.12$                  | 204 (fixed) | 45 $\pm$ 6      | 214 $\pm$ 17   |
|             | 12.1          | $4.18 \pm 0.15$                  | 204 (fixed) | 37 $\pm$ 7      | 213 $\pm$ 26   |
| LH          | 4.1           | $5.38 \pm 0.45$                  | 245 $\pm$ 46| 46 $\pm$ 4      | 221 $\pm$ 12   |
| SO          | 4.1           | $6.31 \pm 0.27$                  | 272 $\pm$ 25| 50 $\pm$ 6      | 215 $\pm$ 17   |
|             | 7.4           | $6.10 \pm 0.20$                  | 218 $\pm$ 17| 48 $\pm$ 5      | 199 $\pm$ 14   |

$^a$ Determined from temperature dependence up to 160 K.

Figure 3. Temperature dependence of PR spectra of the BGaAs alloys with (a) 4.1%, (b) 7.4%, and (c) 12.1% boron concentrations. For a better comparison of spectra obtained at different temperatures, some were multiplied, as marked in the graph.

Figure 4. Relative changes in energy of PR resonances by equation (1) and plotted in figure 4 as relative changes in energy. For comparison, we have also plotted the band gap temperature evolution of GaAs (black line). It is visible that the energy shift between 20 K and 300 K of the HH transition is slightly reduced for the sample containing boron. Moreover, the temperature-induced shift of the SO transition is around 12% and 24% higher than that for the HH one for samples with 4.1% and 7.4% boron content, respectively. The temperature dependencies have been analyzed using Varshni and Bose–Einstein formulas (equations (3) and (4)) and are summarized in table 1, following the procedure shown for the analysis of optical absorption spectra. For both Varshni and Bose–Einstein parameters extracted for the HH transition, their small evolution can be recognized, especially in the case of $\alpha_B$ representing electron-average phonon interaction, for which incorporation of B into GaAs causes its decrease. The slightly reduced sensitivity of transition energy to temperature also impacts the values of the $\alpha$ Varshni coefficient. Our findings cannot be compared with results published to date since the previous band gap dependencies studies were based on the PL method and the observed emission involves localized states [7, 10, 13, 20]. Even though optical absorption mostly resolves one absorption edge, the determined Varshni and Bose–Einstein parameters are in good agreement with those
Figure 4. Temperature-related shift of energies of (a) HH transition and (b) SO transition observed in PR spectra of the $\text{B}_x\text{Ga}_{1-x}\text{As}$ alloys together with the fits by Varshni (thin gray line) and Bose–Einstein (blue dashed line) formula. For comparison, the temperature dependence of energy of the $E_0$ transition for GaAs is also plotted based on the literature data [49].

Figure 5. Room temperature PR spectra measured at various hydrostatic pressures for the $\text{B}_x\text{Ga}_{1-x}\text{As}$ layers with (a) 4.1%, (b) 7.4%, and (c) 12.1% boron concentrations.

obtained for the HH transition from temperature-dependent PR, especially for the Bose–Einstein model where the values lay within their uncertainties, except for the 12.1% alloy which exhibited different absorption edge shift.

The influence of adding boron atoms to the host material, namely GaAs, has also been studied with hydrostatic pressure up to $\sim 18$ kbar. The obtained PR spectra at room temperature under elevated pressure for the samples investigated in this work are shown in figure 5. A strong signal around 1.6 eV corresponding to HH transition is observed for all three samples, whereas the SO one is present only for structures with 4.1% and 7.4% boron concentration. At the same time, the LH transition amplitude is too low to be distinguishable from the HH resonance. A weak and narrow feature observed at $\sim 1.53$ eV is associated with the second-order diffraction of the laser line (405 nm) used as a pumping beam.

The energies of the optical transitions measured at different hydrostatic pressures were obtained by the Aspnes formula (equation (1)) and are plotted in figure 6(a). In the considered
pressure range, both HH and SO transitions blueshift steadily, and experimental results were well fitted by a linear function. Determined pressure coefficients $\alpha_P$ of optical transitions for different samples are given in the legend of the figure. It is apparent that, for boron-containing alloys, the pressure coefficient of the HH transition decreases with increasing boron concentration and is reduced by 30%–40% compared with GaAs (figure 6(b)). The obtained values of $\alpha_P$ parameters are in disagreement regarding chemical trends since that quantity should increase as the cation-atomic number and the ionicity decreases due to substitution of gallium by boron atoms [42]. A smaller discrepancy was found by Shan et al, who reported the value of $\alpha_P$ for $B_{0.012}Ga_{0.988}As$ reduced by 4% relative to GaAs, on the basis of measurement performed up to $\sim 60$ kbar [4]. The results by Shan et al. can be explained within the band anticrossing model (BAC), which assumes anticrossing interaction between the energy level of isovalent impurity and the extended states of the conduction band of the host material [43, 44]. Taking into account the anticrossing interaction, which leads to the splitting of the conduction band into two branches, $E_+$ and $E_-$, the increase of hydrostatic pressure causes the upward shift of the lower $E_-$ branch toward the isovalent level and the change of character of this band from an extended to a localized state [45]. Such behavior is responsible for the reduced value of the pressure coefficient for HMAs like III–V semiconductors containing nitrogen [43, 46], bismuth [47], and that considered in this work, boron [4, 48]. The slight reduction of $\alpha_P$ for the BGaAs alloy with only 1.2% B content observed by Shan et al can be caused by insignificant anticrossing interaction of extended states of GaAs and B level, which is located high above the conduction band edge [4]. Here, we have studied samples with higher boron concentrations, i.e. 4.1%, 7.4%, and 12.1%, for which the coupling strength increases, leading to more significant anticrossing interaction and reduction of pressure coefficients by almost 40% in comparison to GaAs.

4. Conclusions

In this work, we have shown the results of experimental studies of BGaAs alloys with 4.1%, 7.4%, and 12.1% boron content grown by MBE on a GaP substrate, which allows us to unambiguously study the optical transitions for the BGaAs semiconductor. The observed transitions, namely HH, LH, and SO, were investigated by PR spectroscopy with temperature and hydrostatic pressure. To qualitatively analyze the influence of temperature on the energy of transitions, we applied Varshni and Bose–Einstein relation and have shown that the substitution of gallium by boron atoms causes a decrease of electron–phonon interaction for the HH transition and a slight reduction of the $\alpha$ Varshni coefficient. PR has been proven to overcome experimental difficulties characteristic of classic optical absorption, which cannot be used to obtain multiple band-edge transitions and presents additional difficulties. Moreover, we have studied the effect of boron atom incorporation on the optical properties of BGaAs by applying hydrostatic pressure. The reduced value of the pressure coefficient determined for the HH transition has been discussed within the BAC model framework and chemical trends. It can be concluded that, in the BGaAs alloys, the anticrossing interaction between the boron impurity level and the conduction band of GaAs is observed in these studies, which is typical for HMAs.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.
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