Optica Applicata, Vol. XXXVI, No. 2–3, 2006

Investigation of segregation by quantitative transmission electron microscopy

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The segregation effect occuring during molecular beam epitaxy and metalorganic vapour phase epitaxy growth of ternary III-V semiconductor heterostructures was investigated by quantitative transmission electron microscopy (QTEM) and by simulation of optical properties. The concentration distribution of various III-V semiconductor heterostructures was measured by QTEM and averaged along the direction perpendicular to the growth direction. Resulting concentration profiles could be well fitted using the model of MURAKI *et al*. (Muraki K., Fukatsu S., Shiraki Y., Ito R., Appl. Phys. Lett. **61**(5), 1992, p. 557) yielding the segregation efficieny *R*. For the investigation of the effect of segregation on the photoluminescence, concentration profiles for different segregation efficiencies were simulated and photoluminescence peak energies were derived by solving Schrödinger's equation for spatially varying potentials deduced from the measured concentration profiles.

Keywords: quantitative TEM, surface segregation.

1. Introduction

Molecular beam epitaxy (MBE) and metalorganic vapour phase epitaxy (MOVPE) are common methods for deposition of epitaxial layers of high crystal quality on various substrate materials. The deposition of material can be controlled with high accuracy. For instance, sub-monolayer amounts of material can be deposited and the composition of layers can be controlled to about 2%. The perfection of these growth techniques allows fabrication of optoelectronic devices such as light emitting diodes (LEDs) and laser diodes (LDs). Despite the high degree of development of the growth techniques some basic effects occuring during epitaxy are not fully understood, yet.

A very prominent effect is the segregation of an atomic species onto the surface during growth. This effect was first observed by CHIANG *et al*. [1]. They investigated the surface of an AlGaAs layer grown on a GaAs substrate by surface sensitive methods such as X-ray photoemission spectroscopy (XPS) and found that the surface of the AlGaAs consisted of nearly pure GaAs. The observations of CHIANG *et al*. [1] were interpreted by STALL *et al*. [2] as segregation of Ga atoms onto the surface.

First systematic studies were carried out by MASSIES *et al.* [3]. They measured the Ga concentration on the surface and compared it with the composition of the bulk material for different temperatures. MOISON *et al.* [4] explained the observed phenomena as an exchange reaction between the topmost layer ("surface layer") and the layer beneath this layer ("bulk layer"). They also showed that this mechanism leads to a broadening of interfaces of heterostructures and developed a thermodynamic model of segregation to describe the broadening. The model well described the composition of the surface layer for high temperatures [5] and small bulk layer concentrations [6]. In order to apply the exchange model also to lower temperatures DEHAESE *et al.* [5] suggested a kinetic model involving a two level system which leads to the Moison model [4] at high temperatures, but additionally describes the kinetic limitation of the Moison model at lower growth temperatures. However, since both models rely on the assumption of an exchange between the surface layer and the bulk layer, they are both only valid for bulk concentrations smaller than 11%, as was shown by GERARD *et al.* [7] and later by ROSENAUER *et al.* [6]. In reference [6] it is pointed out that the failure of the model at higher bulk concentrations origins from the assumption that the amount of the segregated species in the surface layer cannot exceed one monolayer (ML). But as later investigations showed, this can be the case, indeed. These investigations started with the paper of GERARD [7]. He found a method to measure the amount of segregated In on the growth surface during growth of an InGaAs layer on a GaAs substrate. The 2D–3D transition of the growth mode of an InAs layer grown on GaAs takes place after the deposition of 1.8 ML InAs. The transition is shifted towards smaller depositions if an InGaAs layer was deposited prior to the InAs growth. The difference corresponds to the amount of In on the growth surface after the InGaAs growth. The method of GERARD [7] was used by TOYOSHIMA *et al.* [8] to extensively study the amount of In on the growth surface for different bulk layer compositions. They found that the amount of In on the growth surface can exceed 1 ML and that the 2D–3D transition of the growth mode was invoked, when the amount of In on the growth surface reached 1.6 ML. Based on a recent EFTEM study of WALTHER *et al.* [9], CULLIS *et al.* [10] also developed a segregation based model for the 2D–3D transistion of the growth mode. In agreement with TOYOSHIMA *et al.* [8] they concluded that the transition occurs, when the amount of In on the growth surface reaches a critical value. Applying the model of DEHAESE *et al.* [5] they found a critical amount to be 0.8–0.85 ML.

The investigations of Toyoshima et al. suggest that the structure of the surface layer significantly differs from that of the bulk layer. This was confirmed by several further papers. Firstly, EVANS *et al.* [11] found by temperature programmed desorption experiments that the "surface layer" contained 1.3 ML (1.6 ML) In in steady state growth at a growth temperature of 480°C (530°C) and that the binding energy of In in the surface layer is smaller than in the crystal. Further evidence was given by the investigations of GARCIA *et al.* [12]. They showed that the "surface layer" did

not contribute to the overall stress in the sample. In order to emphasize the differences to the "bulk layer" they called the layer on the growth surface the "floating layer". Further evidence of the mobile state of the In atoms in the floating layer was given by MARTINI *et al.* [13]. They showed that the exponential decrease of the RHEED intensity oscillations observed at the onset of the InGaAs growth as well as the increase of the signal during capping was caused by scattering of the electron beam by mobile adatoms contained in the floating layer.

A phenomenological model for segregation was suggested by MURAKI *et al.* [14]. They assumed that a portion R of the In atoms impinging on the growth surface enter the floating layer, whereas the remaining In atoms are incorporated into the crystal. The In concentration $x(n)$ in monolayer n according to this model is given by the formula

$$x(n) = \begin{cases} 0 & : \quad n < 1 \\ x_0\left(1 - R^n\right) & : \quad 1 \le n \le N \\ x_0\left(1 - R^N\right) R^{n-N} & : \quad n > N \end{cases} \tag{1}$$

where N is the amount of deposited In in monolayer, x_0 the In concentration and n counts the number of monolayer. The amount of In in the floating layer FI(n) can be deduced from the In concentration profile by observing that

$$\mathrm{FI}(n) = x(n)\,\frac{R}{1-R} \tag{2}$$

From Eq. (2) it can be seen that the amount of In in the floating layer can exceed 1 ML, if $x(n) > R/(1-R)$. In the original paper of MURAKI *et al.* [14] this was considered to be physically unrealistic, but as the experiments of TOYOSHIMA *et al.* [8] and EVANS *et al.* [11] showed it can indeed be the case.

Since segregation alters the concentration of the quantum well it can be expected that segregation also affects the photoluminescence of heterostructures. MURAKI *et al.* [14] showed that a correct description of the well width dependence of the PL energy requires the effect of segregation to be taken into account.

In this paper the effect of segregation and its influence on the photoluminescence of heterostructures is investigated. Concentration profiles are accurately measured in a variety of III-V semiconductor heterostructures by quantitative transmission electron microscopy (QTEM). Resulting concentration profiles are consistent with Eq. (1) using R, N and x_0 as fit parameter. The segregation efficiencies deduced are compared with the values from the literature for different materials and different growth

parameters. To study the influence of segregation on the photoluminescence of the heterostructures concentration profiles with various segregation efficiencies are simulated. The peak energy of the photoluminescence intensity as a function of the segregation efficiency was calculated by solving the Schrödinger equation for spatially varying potentials for electrons and holes, where the potentials are deduced from simulated concentration profiles.

2. Quantitative TEM approach

According to this approach concentration profiles are measured from cross-section and cleaved TEM specimens applying the composition evaluation by lattice fringe analysis (CELFA) technique [15]. The CELFA technique exploits the chemical sensitivity of the 002 beam in sphalerite type crystals. In the electron microscope, the TEM specimen is tilted about 5° around an axis parallel to the growth direction and an imaging condition corresponding to a center of the Laue circle of (0, 20, 1.5) was adjusted. Then the central beam and the diffracted 002 beam are selected with the objective aperture. Resulting images exhibit fringes which correspond to the (002) lattice planes of the crystal structure. For the determination of the concentration the 002 Fourier component of the image intensity is measured from the images and compared with theoretically computed 002 Fourier components to obtain the elemental concentration. The 002 Fourier components of the image intensity are theoretically calculated with the Bloch-wave method. Figure 1 shows the 002 Fourier component for InGaAs as a function of the In concentration for different TEM specimen

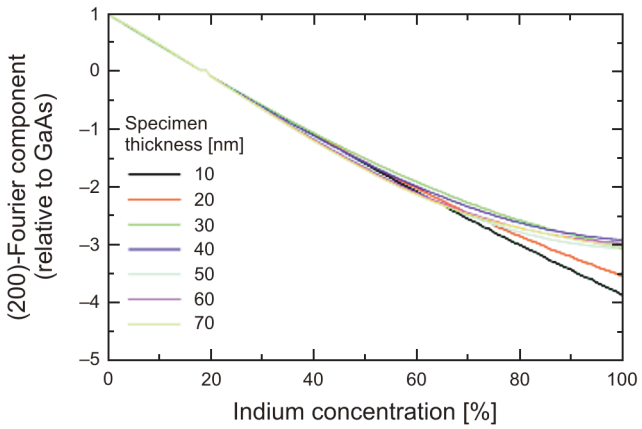

Fig. 1. Calculated relative 002 Fourier component of the image intensity as a function of the In concentration for different TEM specimen thicknesses obtained with the Bloch-wave method. The structure factors used for the Bloch wave calculations were computed within the density functional theory formalism [16].

thicknesses. Bloch-wave calculations rely on the knowledge of crystal structure factors. For the calculations in Fig. 1, the structure factors were computed within the density functional theory formalism [16] to account for redistribution of electrons in bonds. The concentration maps obtained are averaged in planes perpendicular to the growth direction in order to derive concentration profiles.

3. Determination of segregation efficiencies

3.1. Investigation of MBE grown samples

In order to study the effect of segregation on composition profiles several InGaAs/GaAs heterostructures grown at different temperatures and V/III ratios were investigated. As an example, Fig. 2**a** shows an 002 dark field image of a cross-section specimen containing three InGaAs layers being separated by GaAs barrier layers. This sample was grown by MBE at the temperature of 535°C. The quantum wells were nominally 23, 20 and 22 ML thick and the nominal In concentrations of the layers were $x_0 = 0.280$, $x_0 = 0.160$ and $x_0 = 0.245$, respectively. Figure 2**a** shows 3 dark stripes which correspond to the InGaAs layers. Layer 1 contained inhomogeneities indicating the presence of InGaAs islands, whereas layers 2 and 3 were laterally homogeneous. The In concentration profiles measured in layers 2 and 3 are depicted in Fig. 2**b**. They exhibit a strongly asymmetric shape, which is an indication for segregation. The profiles were fitted using Eq. (1) yielding a segregation efficiency of $R = 0.80 \pm 0.01$ for layer 2 and 0.79 ± 0.01 for layer 3. The efficiencies R obtained for a variety of investigated samples are shown in Fig. 3**a** as black triangles, plotted *vs*. the growth temperature.

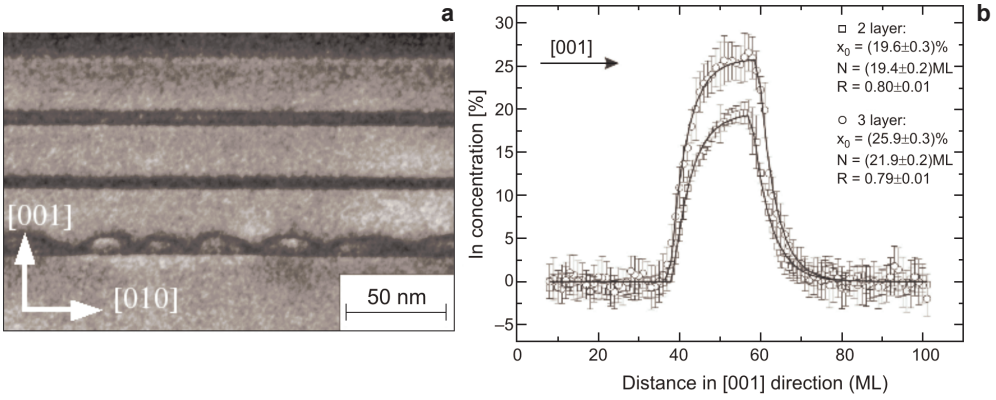

Fig. 2. 002 dark field image of a cross-section sample of an InGaAs/GaAs heterostructure in [100] zone axis orientation (layer 1 exhibits clear indications of islands, whereas layers 2 and 3 are laterally homogenous; the layers are numbered in the order of the growth, where the [001] direction corresponds to the growth direction) – **a**. In concentration profiles measured in layers 2 and 3 – **b**.

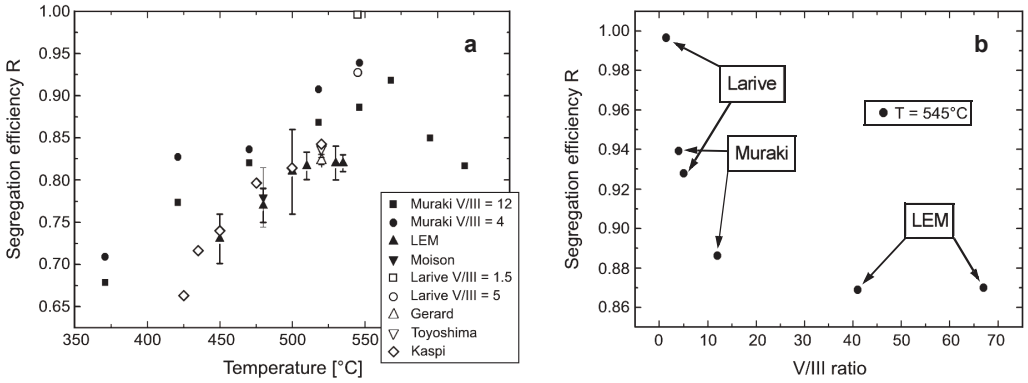

Fig. 3. Segregation efficiency of In in GaAs-on-InGaAs as a function of the growth temperature measured in our group ("LEM") and taken from the literature (the literature segregation efficiencies were deduced from data published in [4, 7, 8, 14, 17, 18]) – **a**. Segregation efficiencies as a function of the V/III ratio at a reference growth temperature of $T = 545$°C. The "LEM" data were linearly extrapolated from data in Fig. 3a – **b**.

Figure 3**a** also gives an overview of segregation efficiencies reported in the literature. As several papers use the model of MOISON *et al.* [4], segregation energies E_s are converted into segregation efficiencies R by

$$R = \frac{1}{\exp\left(E_s / k_B T\right)} \tag{3}$$

where T is the growth temperature and k_B is the Boltzmann constant. Figure 3**a** clearly reveals that the segregation efficiency increases with increasing temperature up to a critical growth temperature of about 550°C. For higher temperatures the segregation efficiency decreases. This can be attributed to the desorption of In atoms from the "floating layer", where the bonding of the In-atoms is weaker than that in a bulk crystal [11]. Altough the general trend of an increasing segregation efficiency is visible in Fig. 3**a**, the values of the segregation efficiencies are scattered. This can be attributed to errors in the determination of the absolute growth temperature or it can be caused by different V/III ratios. In Fig. 3**b** segregation efficiencies are plotted *vs.* of the V/III ratio for a reference growth temperature of $T = 545$°C. The values stemming from our data were obtained by linear extrapolation to 545°C. It can be seen that the segregation efficiency decreases with increasing V/III ratio. However, the segregation efficiency changes more strongly with the temperature than with the V/III ratio.

Since only few reports exist on segregation in material systems other than InGaAs/GaAs, we also investigated segregation of indium in AlAs-on-InAs. A sample consisting of six InAs quantum wells in an AlAs matrix was investigated. The growth temperature was $T = 530$°C. Growth conditions were selected such that high quality InAs/AlAs heterostructures were grown. The nominal thicknesses of the layers were

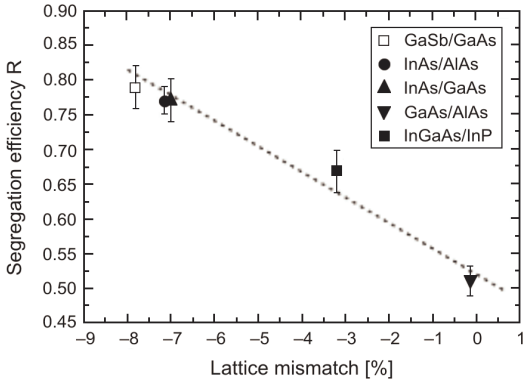

Fig. 4. Segregation efficiency for different types of heterostructures as a function of lattice mismatch between the epitaxial material and the substrate material. The values for the segregation efficiencies of the GaAs/AlAs and the InGaAs/GaAs heterostructures were deduced from references [20] and [4], respectively.

0.5, 0.9, 1.4, 1.9, 2.3 and 2.8 ML. The measured In concentration profiles in the wells with 0.9, 1.4 and 1.9 ML InAs were fitted using Eq. (1) and resulted in a segregation efficiency of 0.77±0.03. A dependence of the segregation efficiency on the thickness of the well could not be detected within the error limits.

To investigate segregation on the non-metal crystal sublattice, a GaSb/GaAs heterostructure containing five GaSb quantum wells in a GaAs matrix was grown by MBE. The structure was grown at a temperature of $T = 530$°C on a GaAs substrate. The thickness of the GaSb layers was varied between 0.5 and 2.5 ML in steps of 0.5 ML. Before and after the growth of each quantum well the growth was interrupted for 30 s in order to remove the excess group V atoms from the growth chamber which also could lead to an asymmetrically broadened concentration profile [19]. Fitting the measured concentration profiles with Eq. (1), we found the segregation efficiency of 0.78±0.03. Also, we did not observe any dependence of the segregation efficiency on the layer thickness.

To compare the segregation efficiencies measured in different types of semiconductor heterostructures, the segregation efficiency R is plotted as a function of the lattice mismatch between the epitaxial layer material and the substrate material in Fig. 4. Note, that Fig. 4 compares segregation efficiencies in samples grown at "optimum" growth conditions which lead to a high crystal quality of the heterostructures. It can be seen that for materials with a lattice mismatch around –7% the segregation efficiencies are close to 0.8, whereas a smaller lattice mismatch results in a lower segregation efficiency. The values for the GaAs/AlAs and InGaAs/InP heterostructure were taken from references [20] and [4], respectively.

3.2. Investigation of MOVPE grown samples

To study the influence of the growth technique on segregation of In in GaAs-on-InGaAs, a MOVPE grown specimen was also investigated. A single InGaAs quantum well was

grown on a GaAs substrate at a growth temperature of 500°C using arsine (F = 2.232 mmol), trimethylgallium (F = 6.146 μmol) and trimethylindium (F = 4.019 μmol) as precursor gases. Fitting the measured concentration profiles using Eq. (1) yields an average value of the segregation efficienies of 0.34±0.04. This value is significantly smaller than the segregation efficiencies found in MBE grown InGaAs/GaAs heterostructures. This appears to be in agreement with the measurements of PISCOPIELLO *et al*. [21], who found the value of $R = 0.65 \pm 0.05$ for the segregation efficiency of In in GaAs at growth temperature of T = 550°C.

4. Simulation of photoluminescence energies

In Section 3 an overview on measured segregation efficiencies and their dependence on material, growth conditions and growth technique was given. In this section we study the influence of segregation on the photoluminescence energy for InGaAs quantum wells embedded in a GaAs matrix.

For this purpose, a procedure was proposed that allows the computation of the photoluminescence energy from a given concentration profile. From the concentration profile, the potential energy of electrons and holes was computed as described in the following. The bandgap energy $E(x)$ for a concentration x is calculated by

$$E_0(x) = E_0^{\mathrm{InAs}} x + E_0^{\mathrm{GaAs}}(1-x) - x(1-x)s \tag{4}$$

where E_0^{InAs} and E_0^{GaAs} are the bandgap energies of InAs and GaAs, and s is the bowing parameter describing deviations from the linear approximation. The energy of the conduction band E_{CB} and the valence band E_{VB} for the heavy hole can be found by noting that

$$E_{\mathrm{CB}} = E_0 + \delta_e \tag{5}$$

$$E_{\mathrm{VB}} = -\delta_h + \frac{1}{2}\delta_s \tag{6}$$

while

$$\delta_e = -2a_c \frac{c_{11} - c_{12}}{c_{11}} \varepsilon \tag{7}$$

$$\delta_h = -2a_v \frac{c_{11} - c_{12}}{c_{11}} \varepsilon \tag{8}$$

$$\delta_s = -2b \frac{c_{11} + 2c_{12}}{c_{11}} \varepsilon \tag{9}$$

where a_c and a_v are the absolute hydrostatic deformation potentials of the conduction band and the valence band, respectively, b is the deformation potential describing

the tetragonal distortion of the crystal, c_{11} and c_{12} are the elastic constants and ε is the strain. The relative alignment of the valence bands in epilayer and substrate is described by the band offset Q_v, which is defined as

$$Q_v = \frac{\Delta E_v}{\Delta E_0} \tag{10}$$

where ΔE_0 is the difference of the bandgaps and ΔE_v is the energy difference of the valence bands. Using the spatially varying potential energy derived from the simulated concentration profiles, the Schrödinger equation was solved using the Numerow algorithm. To calculate the energy of the photoluminescence the difference of the binding energies of the electrons and holes in the well was determined and the binding energy of the exciton, computed by a simple H-atom like model, was substracted from the difference. Such a procedure neglects the effect of confinement-related quenching of the excitons on the PL energy. To have an indication for the dependence of the PL intensity on segregation, the overlap integral B, given by formula

$$B = \int_{-\infty}^{\infty} \mathrm{d}z \left| \Psi_e(z)\, \Psi_e(z) \right| \tag{11}$$

was calculated where Ψ_e and Ψ_h are the wavefunctions of the electron and the hole in the well. The temperature dependence of the PL was taken into account by assuming a temperture dependent bandgap energy according to Varshni's empirical law [23].

To check our approach, the PL of the InGaAs/GaAs heterostructures grown by MBE and MOVPE was measured and the energy at the maximum of the PL intensity was compared with the calculated values of the PL energy taking the concentration profiles determined in Section 3. Figure 5**a** shows a low temperature (5 K) photoluminescence spectrum of the MBE grown InGaAs/GaAs structure. The spectrum exhibits three peaks stemming from the 3 different layers (see Fig. 2**a**), where the broad peak can be attributed to the layer with islands. The other two emission lines were determined to be at 1.315 and 1.385 eV. When the emission energy was computed from the measured concentration profiles, emission energies lying at 1.315 and 1.379 eV were found (material parameter for the bandgap energies and the deformation potentials were taken from reference [22]). Figure 5**b** shows the room temperautre PL spectrum of the MOVPE grown sample. The PL energy is 1.179 eV. From the measured concentration profile we computed an energy of 1.170 eV. The computed energies appeared to be in excellent agreement with measured PL energies.

To study the influence of segregation on the PL energy, concentration profiles were simulated using Eq. (1) varying the segregation efficiency from 0.01 to 0.99 with a stepsize of 0.01 for different quantum well thicknesses N and nominal In-concentrations x_0. For all simulated concentration profiles the respective PL energies and overlap integrals were computed. Figure 6**a** shows the computed PL energy as a function of the segregation efficiency R. For very thin quantum wells the influence of the segregation on the PL energy was negligible. For thicker quantum

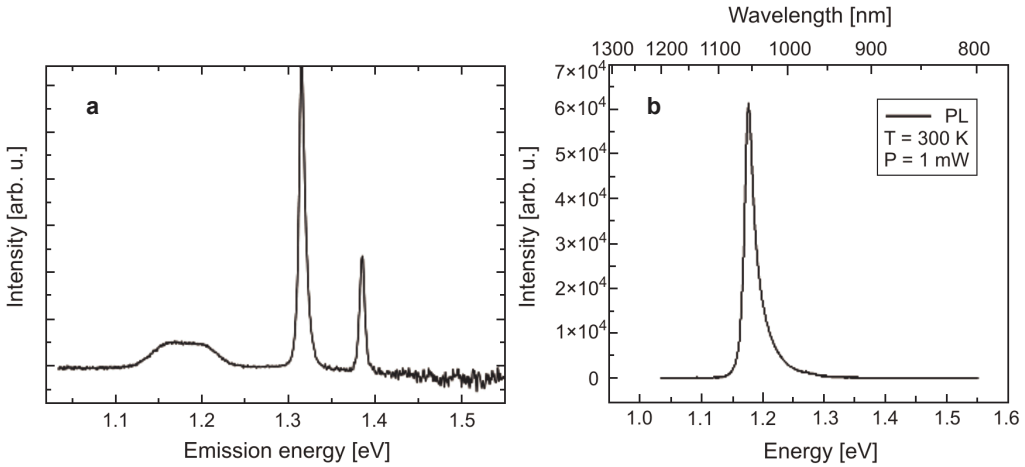

Fig. 5. PL spectra of the MBE grown (**a**) and the MOVPE grown (**b**) InGaAs/GaAs heterostructures.

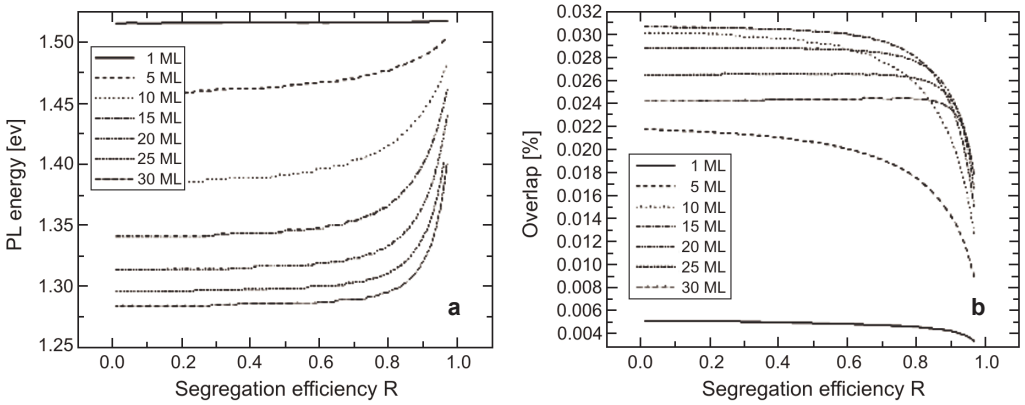

Fig. 6. PL energy as a function of the segregation efficiency *R* for different quantum well thicknesses *N* and an In concentration of 25% – **a**. The corresponding overlap integral – **b**.

wells the PL energy was affected only for segregation efficiencies larger than approximately 0.70. The blue shift of the PL energy was caused by the reduction of the In concentration in the well owing to the segregation of In atoms into the capping barrier layer. Only for small quantum well thicknesses close to 1 ML the effect of segregation becomes small. This can be explained by the combination of two effects. With increasing segregation efficiency, a blue shift is caused by the reduction of the In-concentration. On the other hand, a red shift occurs owing to an increasing quantum well width which is connected with decreasing confinement energies of electrons and holes.

Similar results can be found for the overlap integral B as a function of the segregation efficiency R, which is depicted in Fig. 6**b**. The overlap integral significantly changes only for segregation efficiencies larger than 0.70.

As typical segregation efficiencies for InGaAs/GaAs heterostructures are close to or larger than 0.70, an influence of segregation on the optical properties of the heterostructure can be expected. A blue shift of the PL energy owing to segregation was already reported by MURAKI *et al*. [14] and later also by DE LA CRUZ [24]. The decrease of the overlap integral is in agreement with the experiments of CHIRLIAS *et al*. [25]. They found an increase of the PL intensity when the segregation efficiency was reduced. For very thin quantum wells ($N \approx 1$ ML) DISSEIX *et al*. [26] found that the optical properties were not influenced by segregation, which was confirmed by the calculations performed in this work.

5. Summary

In summary, segregation efficiencies were measured and compared with values from the literature in dependence on different growth parameters for InGaAs/GaAs heterostructures grown by MBE and MOVPE. Values for other types of heterostructures like InAs/AlAs and GaSb/GaAs were also measured. Typical segregation efficiencies were about 0.8 for heterostructures with lattice mismatch of about –7% and significantly smaller for heterostructures with larger lattice mismatch or for heterostructures grown by MOVPE. A significant influence of the segregation on the PL energy was predicted by theoretical calculations for segregation efficiencies larger than 0.70 and for quantum well thicknesses larger than 5 ML.

Acknowledgements – The following people are acknowledged for providing specimen or for discussion on the calculation of the photoluminescence: J. P. Reithmaier, R. Krebs, A. Forchel, D. Schuh, M. Arzberger, M. Bichler, A. Abstreiter, R. Sellin, D. Bimberg, M. Grün, T. Passow, M. Hetterich.

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Received December 15, 2005
in revised form June 8, 2006