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Photoluminescence Investigations of AlGaN on GaN Epitaxial Films

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AlGaN on GaN epitaxial films with Al contents between 6% and 76% were investigated by stationary photoluminescence experiments which allows to determine the dependence of the energy gap on alloy composition. The observed increase of the luminescence linewidth as a function of the Al molar fraction can be explained by alloy broadening. The localization energy of the bound exciton increases considerably and reaches a value of 53 meV for 61% Al. It could imply that the donor binding energy would markedly deviate from its effective mass value, an unexpected result if the residual donor is Si.

For all nitride based opto-electronic and electronic devices the understanding of the ternary alloys InGaN and AlGaN is an essential point. Optical experiments conclude (at least for small Al-contents) that the non-linear contribution on the dependence of the energy gap on alloy composition is small in AlGaN [1 to 3] and thus AlGaN behaves different than InGaN where the bowing is large. A key issue is to understand why the free carrier concentration in Si doped $Al_xGa_{1-x}N$ for $x > 0.15$ drops significantly while at the same time the thermal activation energy of the conductivity increases considerably.

The films were grown by MOVPE on sapphire (0001) substrates. In a first step a 2 μm thick GaN film was grown on a 30 nm low temperature deposited AlN buffer. On this GaN layer $Al_xGa_{1-x}N$ films with thicknesses between 350 and 650 nm were grown. The AlN molar fraction varied between 0 and 0.76. For films with an Al-content higher than 22% an additional buffer layer was introduced between GaN and AlGaN [4]. More details can be found in [1, 5]. An excimer laser working at 194 nm was used as excitation source in the optical investigations.

In Fig. 1 the photoluminescence spectra of $Al_xGa_{1-x}N$ epilayers with Al contents from 6 to 76% are shown. The dependence of bandgap on alloy composition in the whole range from $x = 0$ to $x = 1$ can be fitted with a small bowing parameter of $b = (0.7 \pm 0.1)$ eV (see Fig. 2).

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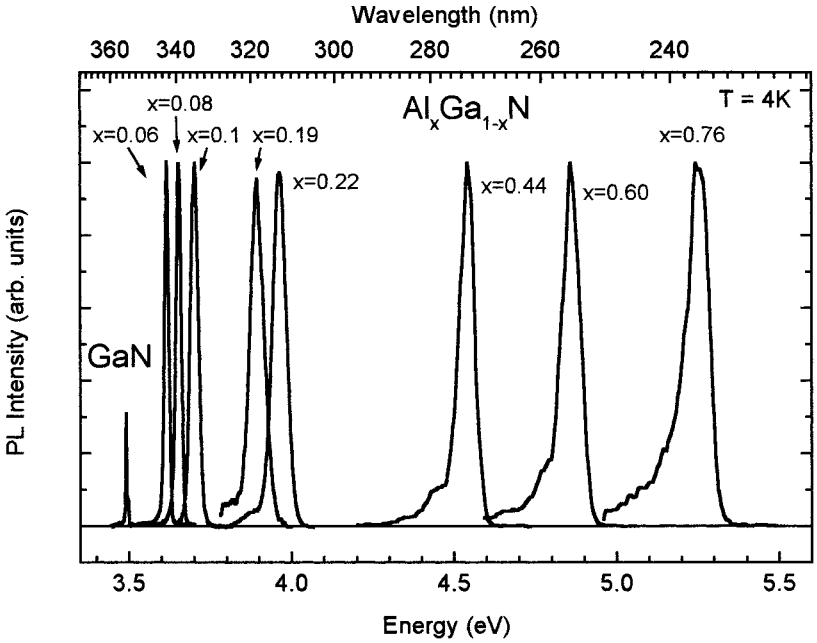

Fig. 1. Photoluminescence spectra of a series of AlGaN on GaN epitaxial films

Besides the energetic shift of the excitonic emission with increasing Al-content the linewidth of the bound exciton luminescence line increases from 14 meV for $x = 0.06$ to 50 meV for $x = 0.76$ which is explainable by alloy broadening. In the inset in Fig. 2 we compare the experimental values of PL and cathodoluminescence (CL) measurements with a calculation based on a model by Schubert et al. [6]. The broadening is caused by a statistical occupation of the cation places Ga by Al and this alloy disorder is seen by the exciton. The main parameters which enter into the model calculation are the exciton volume and its Bohr radius. The exciton volume is calculated from the Bohr radius for which we took 28 Å in GaN and 17 Å in AlN. The free exciton broadening is given by the bold line in the inset of Fig. 2. The calculated linewidth is always larger than the measured one. This difference can be explained by the fact that we are dealing with bound excitons and not with free excitons for which the model of Schubert et al. was developed. The discrepancy can be diminished by introducing an effective Bohr radius accounting for the different binding situation. A very critical parameter is also the dielectric constant of AlN, wich was changed from $\varepsilon = 6.3$ in [7] to $\varepsilon = 8.5$ resulting in a reduction of the calculated linewidth of 30 meV. For further details see [7]. Nevertheless, our experimental results confirm that alloy broadening within this statistical model can account for the linewidth and hence clustering is negligible in line with the results of Zubrilov et al [8].

Temperature dependent measurements were carried out between 4.2 and 300 K. At temperatures between 100 and 300 K dependening on the Al-content the transition from bound to free exciton recombination occurred. Fig. 3 shows the result of a temperature dependent measurement on the sample with 44% Al-content. The tempera-

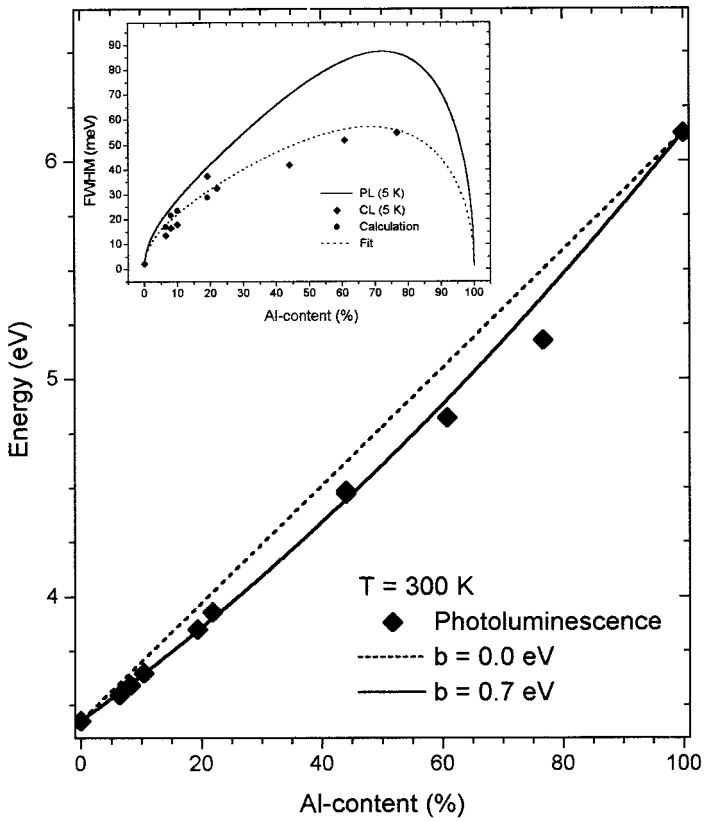

Fig. 2. Luminescence line position of the free exciton at 300 K as a function of Al-content in AlGaN on GaN epitaxial films. The dashed line is a calculation neglecting bowing, and the fully drawn line includes a bowing parameter of $b = 0.7$ eV according to the formula: $E_g(x) = 3.43\,\text{eV} + (3.43\,\text{eV} - 6.13\,\text{eV} - b)\,x + bx^2$. The inset shows the luminescence linewidth as a function of Al-content as obtained by photo- and cathodoluminescence experiments. The dashed and fully drawn lines are calculated, for details see text

ture dependence of the excitonic bandgap could be fitted by the following formula [9, 10]:

$$E(T) = E(0) - \kappa/\{\exp(\theta_E/T) - 1\} \tag{1}$$

with $\kappa = 0.30 \pm 0.2$ eV and θ_E varying from $\theta_E = (550\ \text{K} \pm 50)$ K in GaN to $\theta_E = (300\ \text{K} \pm 50)$ K in AlN. One notes that the data points deviate at low temperatures from the fitted line (bold line in Fig. 3) since the excitons are not free but localized at neutral donors, and there is an energetic difference between free and bound exciton emission – the localization energy. In order to obtain the free exciton line position at low temperatures we extrapolated from the high temperature behaviour to the low temperature behaviour using equ. (1). The first decrease in the line position of the bound excitons is due the fact that the bandgap is already reduced by the influence of temperature although the excitons are still bound to shallow donors. According to Hayne's rule [11] the excitonic localization energy scales linearly with the donor bind-

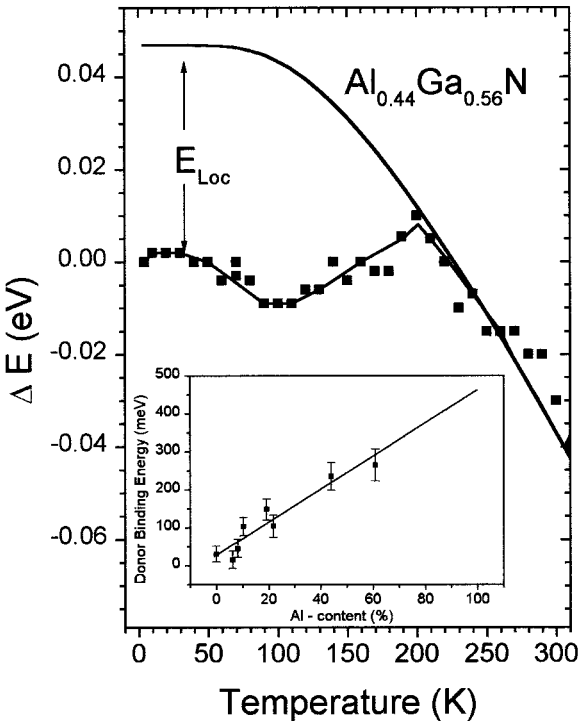

Fig. 3. Change in the luminescence line position as obtained by temperature dependent measurements (for further details see text). In the inset the increase in donor binding energy as a function of the Al-content is shown assuming Hayne's rule is valid in AlGaN

ing energy of the defect $E_{\mathrm{Loc}} = \alpha E_{\mathrm{Bind}}$ with the constant α in GaN being $\alpha = 0.19$ [12]. An increase of the localization energy from 6 meV in GaN to 53 meV in AlGaN with 61% Al-content would hence imply an increase of the donor binding energy from 26 meV in GaN to 470 meV in AlN. It is certainly not in line what one would expect from a simple effective mass approach, but explains the decrease in free carrier concentration and increase in the activation energy of the conductivity as reported by [13, 14].

As shown in Fig. 1, besides the luminescence of the AlGaN layers always the PL signal from the underlaying 2 μm GaN layer can be observed. Because of the high quality of these layers a well resolved donor bound and free A-exciton emission could be detected at low temperatures. In calorimetric measurements even the B-exciton could be observed. The energetic positions of the A- and B-excitons in the GaN layers varied as a function of the Al-content (see Fig. 4). The inset summarizes the results from the CAS and PL measurements. There is a clear blue shift for both excitons, the energetic distance from A to B increases from 9 meV for $x = 0.06$ to 11 meV for $x = 0.22$. From calorimetric reflection and luminescence experiments one can conclude that the top $Al_xGa_{1-x}N$ layer adds additional compressive strain to the underneath GaN film. In Raman spectroscopy the E_2(high) mode can be used for the determination of this strain. The shift of the mode is given by $\Delta\omega_{E_2(\mathrm{high})} = 4.2\ \mathrm{cm}^{-1}$ /GPa $\times\ \sigma_{xx}$ with σ_{xx} the respective component of the strain tensor. We used the room temperature intrinsic value of $\omega(E_2(\mathrm{high})) = 567\ \mathrm{cm}^{-1}$, which was determined for free-standing GaN films. There is a shift of the Raman line from the intrinsic value as a function of Al molar fraction. It is around 0.8 wavenumbers for $x = 0.06$ and around 1.4 wavenumbers for $x = 0.218$. These experimental findings could be quantified by using a two layer

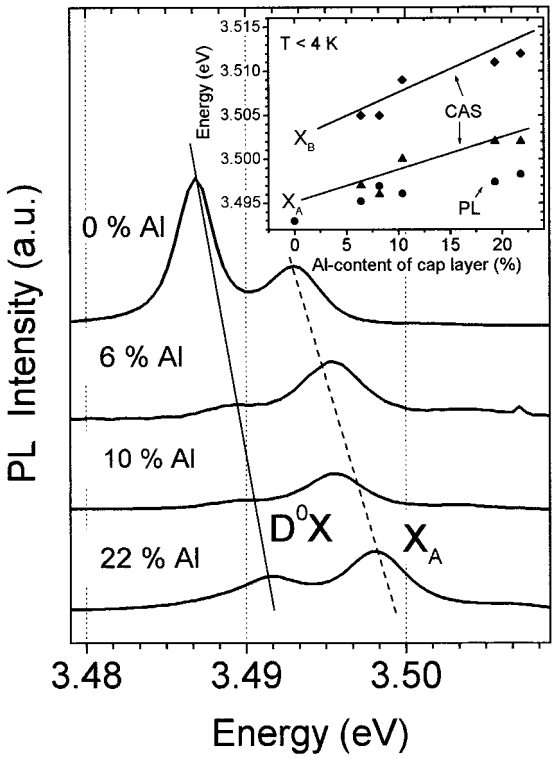

Fig. 4. Blue shift of the donor bound (D^oX), free A-exciton (X_A) and B-exciton (X_B, see inset) line positions in the underlying GaN layer of the AlGaN on GaN films as function of the Al-content of the AlGaN cap layer. In the inset the shift of the X_A and X_B line positions are shown as obtained by photoluminescence (PL) and calorimetric reflection (CAS)

model (neglecting thermal mismatch) and taking into account the elastic constants of $Al_xGa_{1-x}N$ (interpolating between GaN and AlN). Details are published in [15].

In conclusion, AlGaN on GaN epitaxial films show free and bound exciton recombinations with linewidths typical for random alloy broadening. The dependence of energy gap on alloy composition could be extendend up to Al-contents of 76% with a small bowing of $b = 0.7$ eV.

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