

## Linewidths of excitonic luminescence transitions in AlGa<sub>N</sub> alloys

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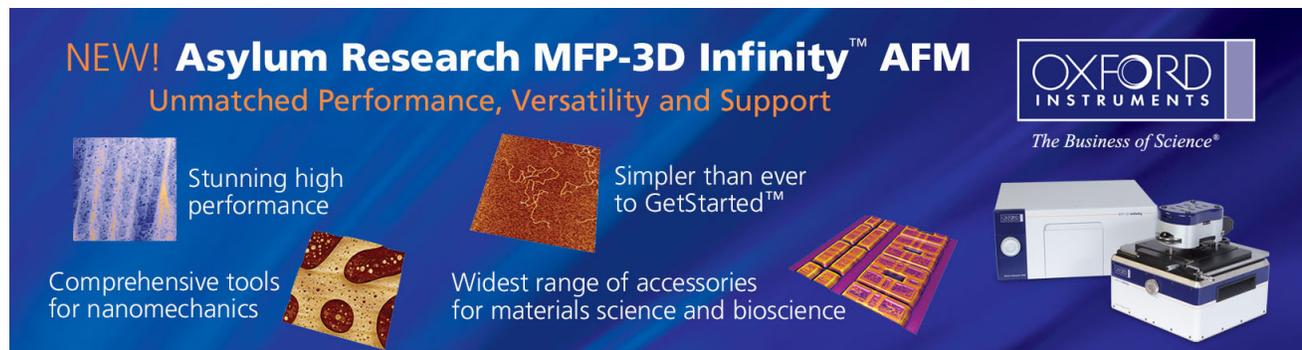
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## Linewidths of excitonic luminescence transitions in AlGa<sub>x</sub>N alloys

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In this work, we present a study of the behavior of linewidths of excitonic photoluminescence transitions measured at 10 K in AlGa<sub>x</sub>N alloys as a function of Al concentration. Samples we have investigated are grown by low-pressure metalorganic chemical vapor deposition on (0001) oriented sapphire substrates with low-temperature GaN buffer layers. The Al composition ranged from 0%–35%. We find that the values of the excitonic linewidth increase as a function of Al concentration and agree very well with those calculated using a model in which the broadening effect is assumed to be due to compositional disorder in semiconductor alloys. The values of the excitonic linewidths measured in our samples are considerably smaller than those reported recently, thus attesting to the high quality of our samples. © 2001 American Institute of Physics.  
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The past few years have witnessed an enormous resurgence of interest in the study of the growth, structural, electronic, and optical properties of III–V nitrides, their alloys, and heterostructures. This interest has been motivated largely by their application in several high power electronic and optoelectronic devices such as light emitting diodes and laser diodes spanning the visible to the ultraviolet spectral range, and by a desire to gain a better understanding of their fundamental properties. A number of these devices have already been fabricated and are commercially available even though a full understanding of their properties is still lacking. In most of these devices, alloys constitute important components and it is therefore essential to obtain a better understanding of their structural, electronic, and optical properties in order to realize devices with improved performance.

An efficient way to investigate the quality of semiconductor alloy systems is by studying the linewidths of their excitonic photoluminescence spectra at liquid helium temperatures. The theoretical prediction of the behavior of the excitonic linewidth associated with the broadening effects of completely random compositional disorder in semiconductor alloys is described well by models presented in several papers,<sup>1–7</sup> as we will briefly discuss in the following. Therefore, the higher the quality of the alloy, the closer the excitonic photoluminescence linewidths are to the theoretically predicted values. Steude *et al.*<sup>8</sup> have recently studied the optical properties of coherently strained Al<sub>x</sub>Ga<sub>1-x</sub>N alloys ( $0 < x < 0.22$ ) grown on GaN by metalorganic chemical vapor deposition (MOCVD) using photoluminescence (PL) and other optical techniques. In particular, they have measured the PL full width at half maximum ( $\sigma$ ) of excitonic transitions as a function of Al composition ( $x$ ) at 4 K and found, as expected, that it increases as a function of  $x$  and reaches a value of about 40 meV at  $x=0.22$ . It was pointed out by Bajaj and Coli<sup>9</sup> that the good agreement observed in Ref. 8 is actually due to use of a theoretical model presented by Schu-

bert *et al.*,<sup>2</sup> which overestimates the broadening effect due to compositional disorder. This model gives values of  $\sigma$  that are about 2.5 times larger than those determined from more sophisticated treatments.<sup>5,6</sup>

In this work we report a systematic study of the variation of  $\sigma$  as a function of Al concentration in Al<sub>x</sub>Ga<sub>1-x</sub>N alloys measured by PL spectroscopy at 10 K. In our samples the value of  $x$  varies from 0 to 0.35, a range considerably larger than that used by Steude *et al.*<sup>8</sup> in their samples. We find that our values of  $\sigma$  increase as a function of  $x$ , as expected, and are considerably smaller than those measured by Steude *et al.*<sup>8</sup>

The 1  $\mu$ m thick Al<sub>x</sub>Ga<sub>1-x</sub>N epilayers were grown by low-pressure MOCVD on sapphire (0001) substrates with 20 nm low-temperature GaN nucleation layers. The growth temperature and pressure were 1060 °C and 100 Torr, respectively. Trimethylgallium (TMG) and trimethylaluminum (TMAI) were used as the metal organic sources. The Al content was determined by TMG and TMAI flow rates as well as from the room temperature PL spectral peak positions. The Al contents for selective samples were also determined by x-ray diffraction and secondary ion mass spectroscopy measurements. The accuracy of the  $x$  values was within  $\pm 0.02$ .

All continuous wave PL measurements were made at 10 K by means of a single photon counting detection system together with a microchannel-plate photomultiplier tube, with a detection capability ranging from 185 to 800 nm and an overall spectral resolution of about 0.2 meV. The excitation wavelength was 292 nm and the PL signals were collected in reflecting mode at an incident angle of about 30°.

In Fig. 1 we display the PL spectra of Al<sub>x</sub>Ga<sub>1-x</sub>N for several values of  $x$  at 10 K. As expected, the PL spectra show excitonic transition energies and linewidths that increase as a function of Al concentration. We also observe a decrease of the PL intensity due to the increase of nonradiative recombination processes with increasing Al concentrations. In order to understand different contributions to the excitonic transitions we have carried out a line shape analy-

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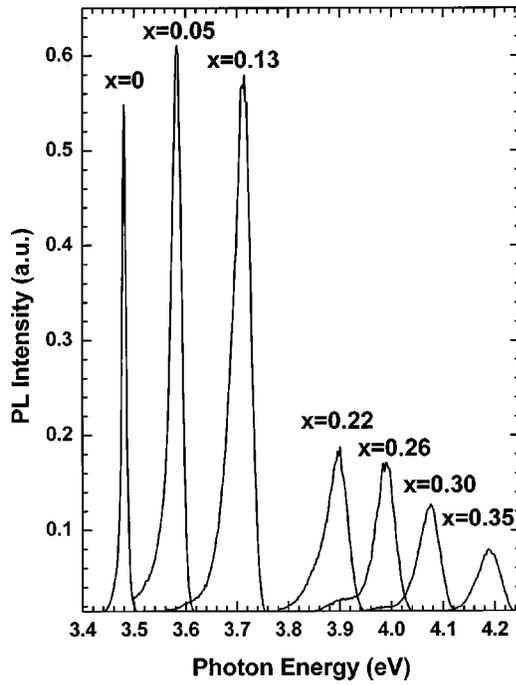


FIG. 1. Photoluminescence spectra of  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  alloys measured at 10 K for several values of  $x$ .

sis of the PL spectra measured using a multiple Gaussian curve fitting procedure based on minimization of the  $\chi^2$  error. For all samples with  $x > 0$  we find that the line shape of the PL signal can be deconvoluted into two distinct contributions due to two different radiative transitions associated with loosely localized excitons.

During the past 20 years a number of groups<sup>1-7</sup> have reported calculations of  $\sigma$  as a function of alloy composition in completely disordered semiconductor alloys, this is briefly reviewed in Ref. 6. In all these calculations it is assumed that excitons are created uniformly in the semiconductor alloy and that they experience slightly different values of the local conduction and valence band edges (assuming the virtual crystal approximation) in the region they occupy. Thus, the values of the emission energies from different parts of the alloy are slightly different from each other, resulting in an inhomogeneously broadened transition. In several of these calculations it is assumed that the volume of the exciton essentially acts as a probe of local compositional disorder and thus the value of  $\sigma$  strongly depends on this quantity. The exciton volume, however, is not a well-defined quantum mechanical quantity, thus it leads to the different values of  $\sigma$  calculated by different groups. For instance, Goede *et al.*,<sup>1</sup> as far as we know, were the first to calculate the dependence of  $\sigma$  on alloy composition using rather simple statistical arguments and defining the exciton volume  $V_{\text{ex}}$  as  $\frac{4}{3}\pi\langle r^3 \rangle$ , where  $\langle r^3 \rangle$  is the expectation value of  $r^3$  and is equal to  $7.5a_{\text{ex}}^3$ . Here  $a_{\text{ex}}$  is the exciton Bohr radius. Singh and Bajaj<sup>3</sup> and Schobert *et al.*<sup>2</sup> defined  $V_{\text{ex}}$  as  $\frac{4}{3}\pi a_{\text{ex}}^3$  in their calculations and obtained values of  $\sigma$  which are about 2.7 times larger than those calculated by Goede *et al.*<sup>1</sup> Later, Zimmerman<sup>5</sup> and Lee and Bajaj<sup>6</sup> avoided the explicit use of the exciton volume and independently calculated the dependence of  $\sigma$  on alloy composition following somewhat similar quantum mechanical approaches. Their results agree rather well with

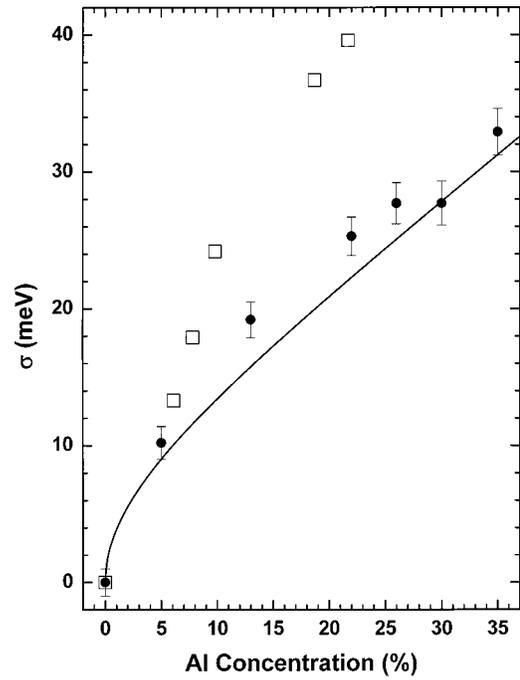


FIG. 2. Variation of the excitonic linewidth ( $\sigma$ ) as a function of Al concentration: Theoretical calculations according to Eqs. (1) and (2) (solid line), determined from the line shape analysis of our PL spectra measured at 10 K (circles), and measured by Steude *et al.* (Ref. 8) (squares) at 4 K.

those determined by Goede *et al.*<sup>1</sup> Lee and Bajaj<sup>6</sup> obtained the following expression for  $\sigma$ :

$$\sigma = 0.41\sigma_0, \quad (1)$$

where

$$\sigma_0 = \frac{dE_g(x)}{dx} \sqrt{8 \ln(2)x(1-x) \frac{V_c}{4\pi a_{\text{ex}}^3/3}}. \quad (2)$$

Here  $V_c$  is the volume of the primitive cell,  $dE_g(x)/dx$  describes the variation of the direct band gap energy with alloy composition,  $a_{\text{ex}} = \epsilon\hbar^2/\mu e^2$ ,  $\mu^{-1} = m_e^{-1} + m_h^{-1}$  is the reduced mass,  $m_e$  and  $m_h$  are the electron and hole masses, respectively, and  $\epsilon$  is the static dielectric constant. To calculate the value of  $\sigma$  in  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  as a function of Al concentration we have used the same values of the physical parameters as those given in Ref. 8, obtained by linear interpolation between the values of GaN and AlN, namely,  $m_e/m_0 = 0.22(1-x) + 0.33x$ ,  $\epsilon = 9.7(1-x) + 6.3x$ . The value of the hole band mass  $m_h = 1.5m_0$  is assumed to be independent of the Al concentration. The volume  $V_c = a_0^3(x)/\sqrt{2}$ , where  $a_0 = 3.160(1-x) + 3.112x$  Å is the lattice constant in the hexagonal plane. The variation of the band gap energy  $E_g(x)$  as a function of the Al concentration ( $x$ ) is given as

$$E_g(x) = (1-x)E_g(\text{GaN}) + xE_g(\text{AlN}) - bx(1-x), \quad (3)$$

where the bowing parameter  $b = 0.6$  eV,<sup>8</sup> and the energy gaps at room temperature  $E_g(\text{GaN}) = 3.42$  eV and  $E_g(\text{AlN}) = 6.20$  eV. It is worth noting that Eqs. (1) and (2) describe the inhomogeneous linewidth broadening due to compositional disorder and do not account for the homogeneous contribution, which is always present at finite temperatures.

In Fig. 2 we display the variation of  $\sigma$  as a function of Al concentration. The solid line is obtained using Eqs. (1) and

(2). Circles represent linewidths of excitonic resonances corresponding to the transition with the highest emission energy obtained from the deconvolution analysis of the PL spectra we measured at 10 K. This is associated with the most loosely bound exciton and therefore is quite close to our model of the free exciton. Squares represent data from Steude *et al.*<sup>8</sup> obtained at 4 K. From our data we have subtracted the constant value for the inhomogeneous broadening at  $x=0$  ( $\sigma_{\text{GaN}}$ ), namely, 7 meV, so as to consider only the effect of the compositional disorder. Although Steude *et al.* did not mention it, we assume that they too have subtracted the value of  $\sigma$  for GaN from their data. Even though there is some scatter in our data, they show in general good agreement with the results of theoretical calculations. Moreover, the fact that subtracting  $\sigma_{\text{GaN}}$  from the derived  $\sigma$  values leads to good agreement with theoretical calculations implies that there is practically no clustering in our alloy samples, which otherwise would increase the linewidth with increasing Al concentration. The values of  $\sigma$  determined by Steude *et al.*<sup>8</sup> are considerably larger than those measured by us. Apparently Steude *et al.* did not perform a line shape analysis and assumed that the transitions they observed were due to radiative decay of only one type of bound exciton. We have accounted only for compositional disorder, but in AlGaN alloys, random electric fields due to additional ionized impurity centers incorporated during growth of alloys may also contribute to broadening excitonic linewidths. It is also important to note that our values of the linewidths are considerably smaller than those determined by Steude *et al.*,<sup>3</sup>

thus indicating the higher quality of our AlGaN alloys. Finally, it should be mentioned that the good agreement between the results of our theoretical calculations and experimental data shows that the excitonic wave function is not significantly perturbed by the localization potential, thus allowing us to use the theory of Lee and Bajaj<sup>6</sup> which is based on a free exciton model.

In summary, we have studied the variation of the excitonic linewidth in  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  alloys as a function of Al concentration, with  $0 \leq x \leq 35$ . We find that the values of the linewidths of excitonic photoluminescence transitions measured at 10 K are considerably smaller than those recently reported by Steude *et al.*<sup>8</sup> and agree very well with those calculated using the formalism of Lee and Bajaj,<sup>6</sup> thus suggesting that random compositional disorder is the main broadening mechanism in our samples.

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