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Excitonic luminescence linewidths in AlGaN alloys with high aluminum concentrations

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In this work, we report a study of the behavior of linewidths of excitonic photoluminescence transitions measured at 10 K in AlGaN alloys for high Al concentrations of 0.5 and 0.7. Our samples were grown by low-pressure metalorganic chemical vapor deposition on (0001) oriented sapphire substrates. We find that the values of the excitonic linewidths we measure agree very well with those calculated using a model in which the broadening effect is assumed to be due to compositional disorder in completely random semiconductor alloys thus attesting to an excellent quality of our samples even with high Al concentrations. © 2002 American Institute of Physics.

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During the past several years study of the structural, electronic, and optical properties of III–V nitrides, their alloys and heterostructures, has attracted a great deal of interest. Several electronic and optoelectronic devices such as high power field effect transistors, light emitting diodes, and laser diodes, spanning the visible to ultraviolet spectral range, have been fabricated and their performance characteristics have been studied in considerable detail. In addition, a number of theoretical and experimental investigations have been carried out to obtain a better understanding of their fundamental properties. In spite of this flurry of activity, a full understanding of the mechanisms involved in the operation of these devices and of the fundamental physical properties of these material systems is still lacking. In most of these devices, alloys constitute important components and it is, therefore, essential to gain a better understanding of their structural, electronic, and optical properties in order to realize devices with improved performance.

One of the most efficient ways to investigate the quality of semiconductor alloy systems is to study the behavior of the linewidths of their excitonic photoluminescence spectra at liquid helium temperatures. Several groups1–7 have calculated the effect of compositional disorder on the excitonic linewidth in completely random alloys. This work has been motivated by the need to elucidate the effect of compositional disorder on the excitonic linewidth in completely random alloys. This work has been motivated by the need to elucidate the effect of compositional disorder on the excitonic linewidth in completely random alloys. This work has been motivated by the need to elucidate the effect of compositional disorder on the excitonic linewidth in completely random alloys. This work has been motivated by the need to elucidate the effect of compositional disorder on the excitonic linewidth in completely random alloys.

In Fig. 1 we display the PL spectra in Al$_{1-x}$Ga$_x$N alloys (0.02 $\leq x < 0.22$) 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 find, as expected, that it increases as a function of $x$.

Following this work we reported a systematic study of the variation of $\sigma$ as a function of Al concentration in Al$_{1-x}$Ga$_{x}$N alloys, using PL spectroscopy at 10 K. In our samples the value of $x$ varied from 0 to 0.35, a range considerably larger than that used by Steude et al.8 in their samples. We found that our values of $\sigma$ increased as a function of $x$, as expected, and were considerably smaller than those measured by Steude et al.8

In this work we report an extension of our study to higher values of Al concentrations up to $x = 0.7$. It has been suggested that the quality of AlGaN degrades as the value of Al concentration is increased. We, however, find that the values of $\sigma$ we measure are very close to those calculated by Lee and Bajaj9 assuming completely random alloys, thus attesting to the high quality of our samples even in this range of large Al concentrations.

About 1-μm-thick Al$_{1-x}$Ga$_x$N epilayers were grown by low-pressure MOCVD on sapphire (0001) substrates with 20-nm-thick low-temperature GaN nucleation layers. The growth temperature and pressure were 1060 °C and 100 Torr, respectively. Trimethylgallium (TMGa) and trimethylaluminium (TMAI) were used as metalorganic sources and ammonia was used as a source of nitrogen. The Al content in the layers was determined from TMGa 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 in $x$ values was within ±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 samples were irradiated with a deep UV laser system using a wavelength of 292 nm$^9$ and the PL signals were collected in a reflecting mode at an incident angle of about 30°.

In Fig. 1 we display the PL spectra in Al$_{1-x}$Ga$_x$N for $x = 0.3$, 0.5, and 0.7 at 10 K. The values of both the excitonic transition energies and of linewidths increase as a function of Al concentration. We also observe a decrease of the PL intensity due to the enhancement of nonradiative recombina-
tion processes with increasing Al concentration. In order to understand different contributions to the excitonic transitions, we have performed a line shape analysis of the measured PL spectra, using a multiple Gaussian curve-fitting procedure, based on the minimization of the $\chi^2$ error. 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.

As mentioned earlier, during the past 20 years a number of groups\textsuperscript{1-7} have reported calculations of $\sigma$ as a function of alloy composition in completely disordered semiconductor alloys. We compare our measured values of $\sigma$ with those calculated by Lee and Bajaj\textsuperscript{6} using a quantum statistical approach. It should be pointed out that their results agree rather well with those determined by Goede, John, and Henning.\textsuperscript{1,2} Lee and Bajaj\textsuperscript{6} obtain the following expression for

$$\sigma = 0.41 \sigma_0,$$

(1)

where

$$\sigma_0 = \frac{dE_e(x)}{dx} \sqrt{8 \ln(2)x(1-x)} \frac{V_e}{4\pi a_c^3\beta^3}.$$

(2)

Here $V_e$ is the volume of the primitive cell, $dE_e(x)/dx$ describes the variation of the direct band gap energy with alloy composition, $a_{en} = e\hbar^2/m_e e^2$, $\mu^{-1} = m_e^{-1} + m_h^{-1}$ is the reduced mass, $m_e$ and $m_h$ are the electron and hole masses, respectively, and $\varepsilon$ is the static dielectric constant. To calculate the value of $\sigma$ in Al$_{x}$Ga$_{1-x}$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, $m_e/m_0(x) = 0.22(1-x) + 0.33x$, $\varepsilon(x) = 9.7(1-x) + 6.3x$. The values of the hole masses associated with the topmost valence bands in GaN and AlN are not known. We, therefore, assume that the hole mass is independent of Al concentration and take its value to be $1.5 m_0$. The volume $V_e(x) = a_0^3(x)\sqrt{2}$ where $a_0(x) = 3.160(1-x) + 3.112x$ Å is the lattice constant in the hexagonal plane. The variation of the band gap energy $E_e(x)$ as a function of the Al concentration $(x)$ is given as

$$E_e(x) = (1-x)E_e(GaN) + xE_e(AlN) - bx(1-x),$$

(3)

where $b$ is the bowing parameter and the values of the energy gaps at room temperature are $E_e(GaN) = 3.42$ eV and $E_e(AlN) = 6.20$ eV. There are more than 20 articles in literature that report the measurements of the values of $b$ in AlGaN using a variety of optical characterization techniques. These values range from $\pm 1.3$ to $-0.8$ eV.\textsuperscript{10} Lee et al.\textsuperscript{10} have provided an excellent but a brief review of this work. Based on their own PL measurements on AlGaN grown by MOCVD and a detailed analysis of the previous work, they propose a value of $b = 0.62 (\pm 0.45)$ eV. Steude et al.\textsuperscript{8} based on their earlier work, have also suggested $0.60$ eV as the value of $b$. In our calculations we have used this value of $b$. According to Lee et al.\textsuperscript{10} the apparently strong bowing ($b = 1.3$ eV) which was obtained in samples not grown on low-temperature buffer layers was the result of incorrectly attributing the dominant transitions to near band-edge excitonic transitions. For further details the reader is referred to their work. 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 in samples with Al concentration $x = 0.5$ and $x = 0.7$ and correspond to the transitions with the highest emission energy as obtained from the deconvolution analysis of the PL spectra we measured at 10 K. These are, therefore, associated with the most loosely bound excitons and can be described by our model which is based on the free excitons. From our data we have subtracted the constant value for the inhomogeneous broadening at $x = 0$ ($\sigma_{GaN}$), namely 7 meV, so as to consider only the effect of the alloy compositional disorder. For the sake of completeness we have also displayed some values of $\sigma$ for Al concentrations less than $x = 0.5$ from our previous work.\textsuperscript{8} We find that the measured values of $\sigma$ even for larger values of Al concentration, namely $x = 0.5$ and $x = 0.7$, agree

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig1.png}
\caption{Photoluminescence spectra in Al$_{x}$Ga$_{1-x}$N alloys measured at 10 K for Al concentrations $x = 0.3$, 0.5, and 0.7.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig2.png}
\caption{Variation of excitonic linewidth ($\sigma$) as a function of Al concentration: theoretical calculation according Eqs. (1 and 2) (solid line), determined from the line shape analysis of our PL spectra measured at 10 K (triangles). Circles represent our earlier data.}
\end{figure}
rather well with the calculated values, thus suggesting a high quality of our samples. This is somewhat surprising in view of the fact that the samples with larger values of $x$ are expected to be of poorer quality. Our work seems to suggest that it is possible to grow high quality Al$_x$Ga$_{1-x}$N alloys with large values of $x$.

It should be pointed out that we have only accounted for the effect of compositional disorder, but in AlGaN alloys, random electric fields due to additional incorporation of ionized impurity centers during growth also contribute to enhance excitonic linewidths. Finally, it should be mentioned that the good agreement between the results of our calculations and the 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 which is based on a free exciton model.

In summary, we have studied the variation of the PL excitonic linewidth in Al$_x$Ga$_{1-x}$N alloys as a function of Al concentration, $x$ larger that 0.3. We find that the values of the linewidths of excitonic transitions measured at 10 K agree very well with those calculated using a formalism of Lee and Bajaj, thus suggesting that random compositional disorder is the main broadening mechanism in our samples.

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