

# EXCITON LOCALIZATION DYNAMICS IN $\text{Al}_x\text{Ga}_{1-x}\text{N}$ ALLOYS

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## ABSTRACT

The optical properties of  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  alloys with  $x$  varied from 0 to 0.35 have been investigated by picosecond time-resolved photoluminescence (PL) spectroscopy. Our results revealed that while the PL intensity decreases with an increase of Al-content, the low temperature PL decay lifetime increases with Al-content. These results can be understood in terms of the effects of tail states in the density of states due to alloy fluctuation in the  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  alloys. The Al content dependence of the energy tail state distribution parameter,  $E_0$ , which is an important parameter for determining optical and electrical properties of the AlGaN alloys, has been obtained experimentally. The PL decay lifetime increases with the localization energy and consequently increases with Al content. The implications of our findings to III-nitride optoelectronic device applications are also discussed.

**Keywords:** GaN, AlGaN alloys, wide bandgap, carrier dynamics, optical transitions, excitons

## 1. INTRODUCTION

In wide bandgap semiconductor optoelectronic devices, such as UV light emitters AND detectors, it is the dynamic processes of the optical transitions that predominantly determine their performance. An understanding of the carrier dynamics provides important information for improving sample quality as well as immense value in designing and optimizing optoelectronic devices based on III-nitrides. However, the investigation and understanding of the dynamic processes of fundamental optical transitions in  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  are rare due to the lack of high-quality samples as well as difficulty in measuring time-resolved optical transitions in the UV region. Important properties, such as the compositional dependence of the optical and electrical properties of the  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  alloys, are not well known despite the fact that AlGaN is a very important material system.

It is well known that the localized exciton transition is the dominant optical process in many semiconductor alloys at low temperatures, including  $\text{CdS}_x\text{Se}_{1-x}$  [1,2],  $\text{GaAs}_x\text{P}_{1-x}$  [3], and  $\text{Zn}_{1-x}\text{Cd}_x\text{Te}$  [4]. A previous calculation [5] has indicated that the amplitude of the potential fluctuation at the band edges caused by the alloy fluctuation is strongly correlated to the energy gap difference between the two semiconductors, e.g. between GaN and AlN for  $\text{Al}_x\text{Ga}_{1-x}\text{N}$ . GaN and AlN form a continuous alloy system whose bandgap ranges from 3.4 to 6.2 eV, giving an energy gap difference  $\Delta E_g$  of 2.8 eV. This is much larger than the typical value of a few tenths of an eV in II-VI semiconductor alloys, in which a strong localization effect is known to exist.

In this paper[6], the compositional dependence of the optical properties of the  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  alloys has been investigated by picosecond time-resolved photoluminescence (PL) spectroscopy. Our results have revealed that the PL intensity decreases with an increase of Al-content. In contrast, however, the low temperature PL decay lifetime increases with Al-content. These behaviors are explained in terms of the effects of tail states due to alloy fluctuations within the  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  alloys.

## 2. EXPERIMENTAL

The 1  $\mu\text{m}$  thick  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  epilayers ( $x < 0.4$ ) were grown by metal-organic chemical vapor deposition (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 metal organic sources. For time-resolved and quasi continuous-wave (cw) PL measurements[6,7], excitation pulses of about 8 ps at a repetition rate of 9.5 MHz were provided by a picosecond laser system with an average power of 20 mW at an excitation wavelength of 292 nm and the PL

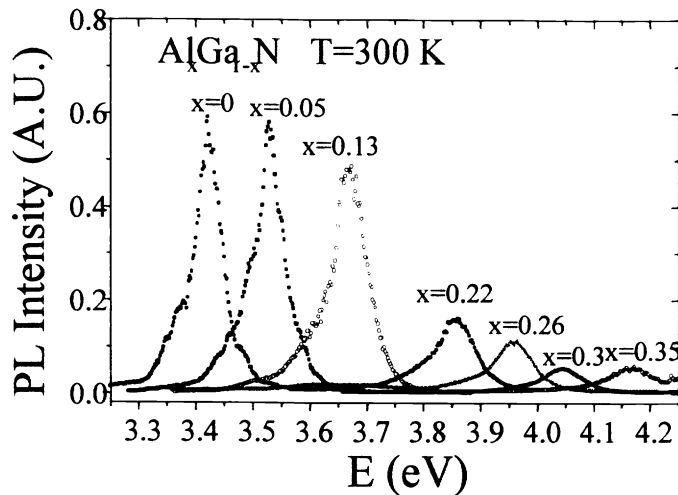


Fig. 1 Room temperature PL spectra of  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  epilayers with different AlN mole fractions produced by our MOCVD reactor at KSU.

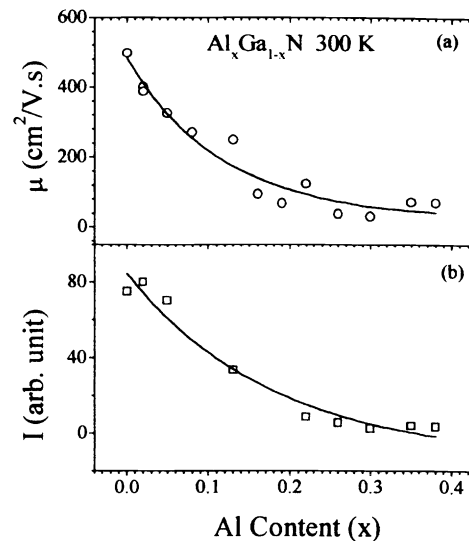


Fig. 2 (a) Electron mobility and (b) relative PL emission intensity of our  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  epilayers as functions of Al-content measured at room temperature ( $T=300\text{ K}$ ).

signals were collected in a reflecting mode at an incident angle of about  $30^\circ$ . The laser beam is polarized along the sample surface (perpendicular to the sample c-axis) and its size on the sample surface was about 0.3 mm in diameter. A single photon counting detection system together with a micro-channel-plate photo-multiplier tube (MCO-PMT) with a detection capability ranging from 185 – 800 nm was used to record time-resolved and cw PL spectra with an overall spectral resolution of about 0.2 meV and time resolution of 25 ps with deconvolution techniques. The variations in PL spectra as well as in temporal responses are negligibly small between run to run, so that an ensemble average of data points was not performed. The Al-content ( $x$ ) was determined by TMG and TMAI flow rates as well as from PL spectra by using equation,

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

with the bowing parameter  $b = 0.98\text{ eV}$  [8]. The energy gaps for GaN and AlN have been assumed as 3.42 eV and 6.20 eV at room temperature, respectively. The Al-contents for selective samples were also determined by x-ray diffraction and secondary ion mass spectroscopy (performed by Charles & Evan) measurements. The accuracy in  $x$  values was within  $\pm 0.02$ .

### 3. RESULTS AND DISCUSSIONS

Room temperature cw PL spectra for  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  alloys with  $0 \leq x \leq 0.35$  are presented in Fig. 1. Apart from the shift of the peak position with increasing Al-content, one also notices a considerable decrease in the PL intensity and increase in the full width at half maximum (FWHM), which is caused by alloy broadening. This alloy broadening also causes a decrease in the electron Hall mobility with an increase of the Al-content. For clear presentation, PL intensity and the electron mobility as functions of Al-content are depicted in Fig. 2. Specifically, the PL intensity (a) as well as the electron mobility (b) at room temperature decrease exponentially with an increase of Al-content.

Figure 3 shows the temperature dependencies of the main emission peak positions ( $E_p$ ) of the  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  alloys with  $x = 0.05, 0.13, 0.22,$  and  $0.35$ , where values of  $E_p$  were determined by fitting the PL spectra near the emission peaks by Gaussian functions. At low temperatures,  $E_p$  increases slightly with temperature in contrast to the expected decrease of the band gap energy.

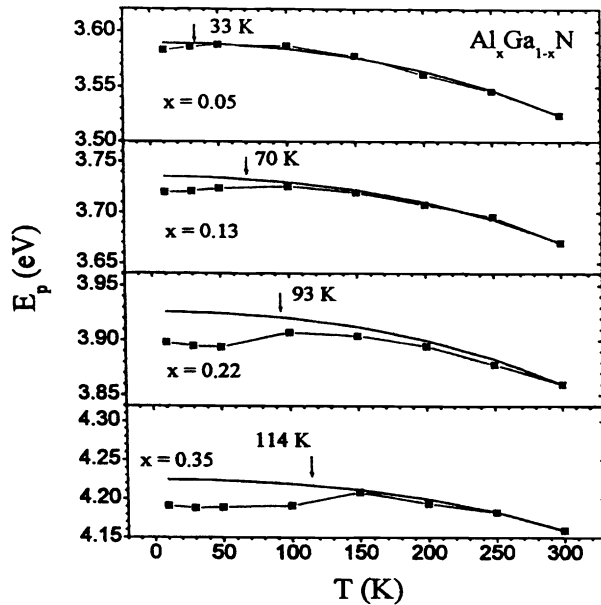


Fig. 3 The temperature dependencies of the main PL emission peak positions  $E_p$ , of the  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  alloys with  $x = 0.05, 0.13, 0.22,$  and  $0.35$ . Arrows indicate the corresponding temperature,  $T_m$ , at which PL behavior switches from localized to non-localized character.

The trend then reverses and the PL peak energy decreases as expected with further increase of temperature. We use Varshni's equation,

$$E(T) = E(T=0) + \alpha T^2 / (T - \beta), \quad [2]$$

with values of  $\alpha = 5.08 \times 10^{-4}$  eV/K and  $\beta = 996$  K[9] to describe the expected temperature dependence of the  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  bandgap. The temperature dependence described by Eq. (2) is plotted with dashed line in Fig. 3. Comparison of this line with the observed temperature dependence of  $E_p$  indicates that the PL peak follows Eq. (2) at higher temperatures and deviates from the expected temperature dependence below a transition temperature,  $T_m$ , by an amount that increases with the Al-content of the sample. The approximate transition temperature ( $T_m$ ) is indicated for each sample in Fig. 3.  $T_m$  increases with the Al-content. A previous work has indicated that stress and defects may also affect the PL spectral shape as well as emission peak positions in AlGaIn [10]. The systematic behavior exhibited by data of Fig. 3, however, may be understood in terms of the localized exciton transition in the tail states due to alloy fluctuation. At low temperatures, exciton localization dominates and the PL peak energy is redshifted relative to the predicted energy (solid line). The redshift is larger for increased Al-contents because the alloy induced fluctuations and the characteristic localization energies are larger. In the low temperature region, however, the PL peak energy slightly increases with temperature because the thermal kinetic energy of the excitons is sufficient to promote the majority of the population to extended states. Finally, at sufficiently high temperatures, the PL emission peak follows the temperature dependence described by Eq. (2). Similar behavior has been reported previously for the temperature-dependent PL emission energy shift in InGaIn/GaN multiple quantum wells[11] and pseudomorphic AlGaIn/GaN heterostructures[12].

The fact that  $T_m$  increases with increased Al-content shows that the characteristic localization energy resulting from

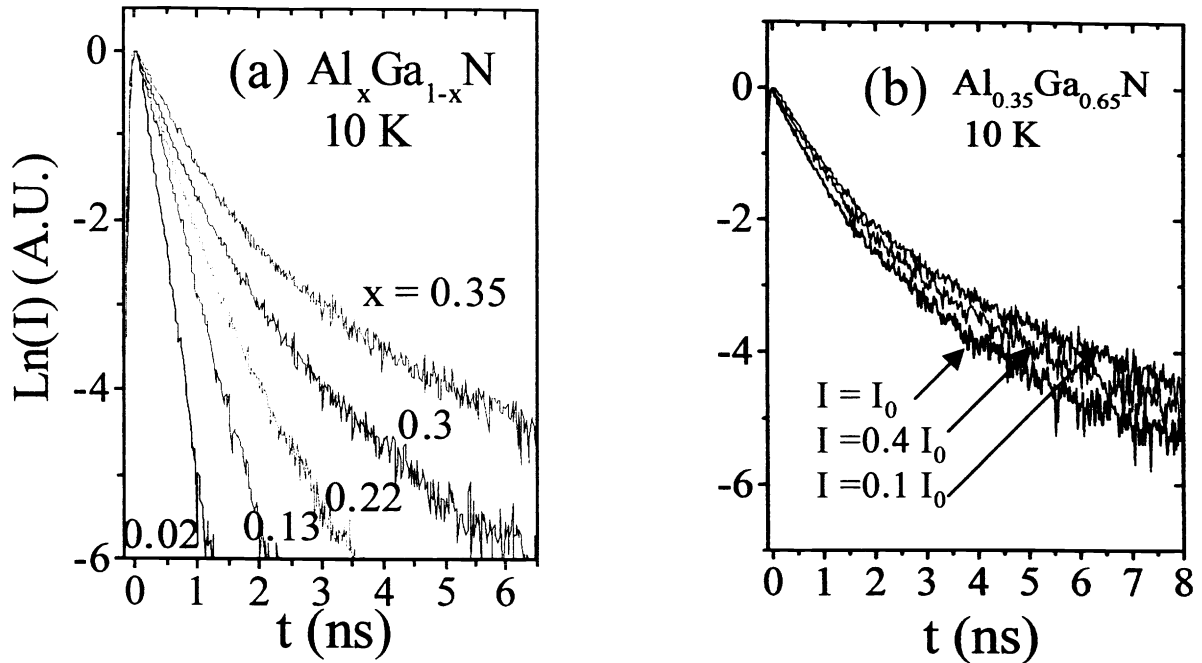


Fig. 4 (a) The semilogarithmic plot of the temporal response for several representative Al contents. (b) The semilogarithmic plot of the temporal response for several representative excitation intensities,  $I$ .

random alloy disorder increases with Al-content in the Ga-rich AlGa<sub>N</sub> alloy. In fact, the temperature dependence of PL peak energy shown in Fig. 3 may be used to estimate the characteristic localization energies,  $E_0$ , for the various AlGa<sub>N</sub> samples. We take  $E_0$  as the deviation at 10 K of the PL peak energy from the energy expected from Eq. 2. Values for  $E_0$  obtained from the data of Fig. (3) increase from 7 to 34 meV as the Al-content increases from 0.05 to 0.35.

PL decay behavior at 10 K was investigated for various AlGa<sub>N</sub> samples. Figure 4(a) shows the PL temporal responses measured at the peak energy for several representative Al-contents. As seen in Fig. 4, the PL decay is single exponential only for the lowest Al- content alloy samples. It is clear from Fig. 4(a) that the effective lifetime increases with increased Al-content. Specifically, the lifetime increases approximately linearly from 0.2 to 0.7 ns as  $x$  varies from 0.02 to 0.35. This behavior is consistent with previous theoretical arguments which predicted that the radiative lifetime of bound excitons increases with binding energy [13,14]. Within the AlGa<sub>N</sub> samples, excitons are energetically and spatially localized due to compositional fluctuations. This localization is analogous to the binding of an exciton to an impurity, so that a larger characteristic localization energy ( $E_0$ ) should result in a longer radiative lifetime. The measured decay time ( $\tau$ ) is related to the radiative lifetime ( $\tau_r$ ) and the nonradiative lifetime ( $\tau_{nr}$ ) by

$$\tau = \tau_r \tau_{nr} / (\tau_{nr} + \tau_r). \quad [3]$$

Therefore, we expect a localization-induced increase of the radiative lifetime, as evidenced by an increase in the measured PL decay time ( $\tau$ ), but the magnitude of the increase in  $\tau$  is not necessarily as great as the increase in  $\tau_r$ .

The excitation intensity dependence of the PL decay was also measured. Figure 4(b) shows the PL temporal responses measured at the peak energy for several representative excitation intensities for an  $\text{Al}_{0.35}\text{Ga}_{0.65}\text{N}$  alloy. As can be seen that the measured lifetime for a given alloy sample decreases with an increase of the excitation intensity. This is also expected for localized excitons because of the band filling effects. It is expected that excitons fill into higher localized states at higher levels of excitation, which effectively increased the exciton nonradiative decay rate (or transfer rate).

#### 4. CONCLUSIONS

In conclusions, we have investigated the optical properties of the  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  alloys ( $0 \leq x \leq 0.35$ ) by using picosecond time-resolved PL spectroscopy. It was found that the PL decay lifetimes at low temperature increase almost linearly with increasing Al-content due to the effect of localization on the exciton radiative lifetime. Conversely, a decrease in PL intensity with increasing Al-content indicates that significant nonradiative carrier loss occurs before carriers thermally relax and become localized. The PL decay lifetimes have been measured and discussed in terms of the effects of tail states due to alloy fluctuation in  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  alloys. The energy tail states distribution parameter  $E_0$ , which is an important parameter describing the effects of the alloy fluctuation on the optical and electrical properties in  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  alloys, has been obtained. It increases monotonically from 7 to 34 meV as  $x$  increases from 0.05 to 0.35. We expect localization of carriers at room temperature for  $\text{Al}_x\text{Ga}_{1-x}\text{N}$ , in particular for large  $x$  alloys since  $E_0$  is larger than the room temperature thermal energy ( $kT = 25$  meV). We believe that while localization may affect minority carrier diffusion in photo-detectors, it should have less effect on heterostructure field-effect transistors (HFET) and laser diodes, where current injection involves majority carrier transport.

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