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Time-resolved photoluminescence studies of $Al_xGa_{1-x}N$ alloys

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The optical properties of $Al_xGa_{1-x}N$ 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 $Al_xGa_{1-x}N$ 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. © *2000 American Institute of Physics.* [S0003-6951(00)01310-3]

In wide-band-gap semiconductor optoelectronic devices, such as UV light emitters, detectors, and UV/blue laser diodes, 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 $Al_xGa_{1-x}N$ 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 $Al_xGa_{1-x}N$ 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 CdS_xSe_{1-x},[1,2] $GaAs_xP_{1-x}N$,[3] and $Zn_{1-x}Cd_xTe$.[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 $Al_xGa_{1-x}N$. GaN and AlN from a continuous alloy system whose band gap ranges from 3.4 to 6.2 eV, giving an energy-gap difference ΔE_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 letter, the compositional dependence of the optical properties of the $Al_xGa_{1-x}N$ alloys have 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 within the density of states due to alloy fluctuations within the $Al_xGa_{1-x}N$ alloys.

The 1-μm-thick $Al_xGa_{1-x}N$ epilayers ($x<0.4$) were grown by metal–organic chemical-vapor deposition 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 (TMAl) were used as metal–organic sources. For time-resolved and quasi-continuous-wavel (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. A single-photon counting detection system together with a microchannel-plate photomultiplier tube with a detection capability ranging from 185 to 800 nm was used to record time-resolved and cw PL spectra. The Al content (x) was determined by TMG and TMAl flow rates as well as from PL spectra by using the equation

$$E_g(x)=(1-x)E_g(\mathrm{GaN})+xE_g(\mathrm{AlN})-bx(1-x), \qquad (1)$$

with the bowing parameter $b=0.98$ eV.[8] The energy gaps for GaN and AlN have been assumed as 3.42 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 accuracies in x values were within ± 0.02.

Room- (300 K) and low-temperature (10 K) cw PL spectra for $Al_xGa_{1-x}N$ alloys with $0 \leqslant x \leqslant 0.35$ are presented in Figs. 1(a) and 1(b). 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. For clear presentation, PL intensity and FWHM as functions of Al content at room and low temperatures are depicted in the insets of Figs. 1(a) and 1(b).

Figure 2 shows the temperature dependencies of the main emission peak positions (E_p) of the $Al_xGa_{1-x}N$ 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

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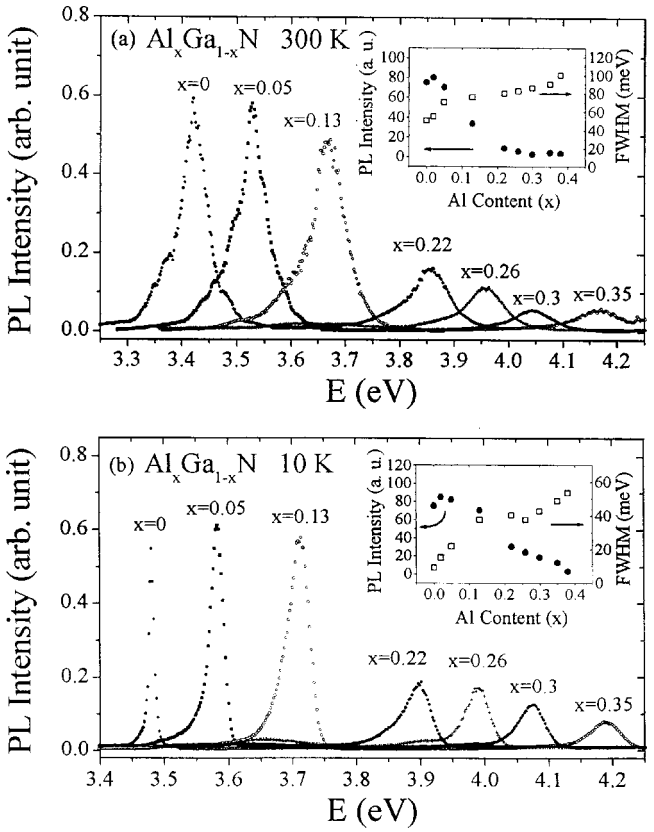

FIG. 1. (a)Room- (300 K) and (b) low-temperature (10 K) cw PL spectra for $Al_xGa_{1-x}N$ alloys with $0 \leqslant x \leqslant 0.35$. The insets show the Al content x dependence of the full width at half maximum (FWHM) and the PL intensity in $Al_xGa_{1-x}N$ alloys.

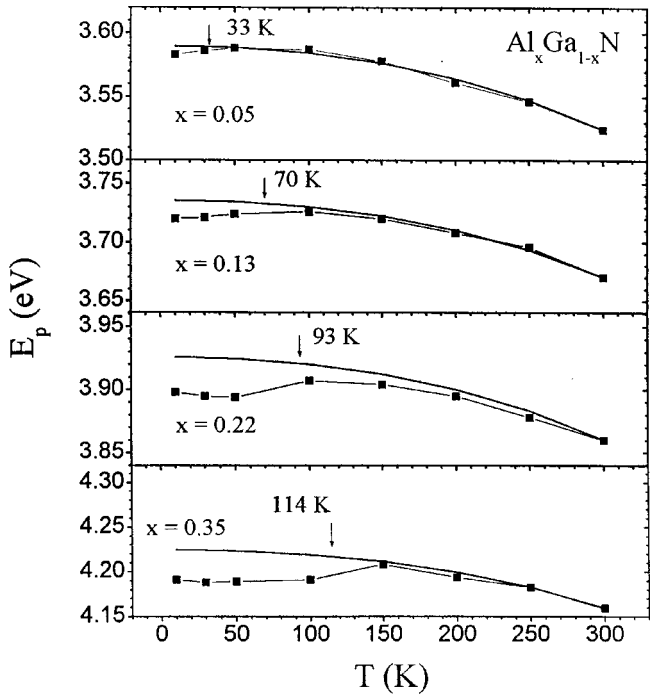

FIG. 2. Temperature dependencies of the main PL emission peak positions E_p of the $Al_xGa_{1-x}N$ alloys with $x=0.05$, 0.13, 0.22, and 0.35. Arrows indicate the corresponding temperature T_m at which PL behavior switches from localized to nonlocalized character.

by Gaussian functions. At low temperatures, E_p increases slightly with temperature in contrast to the expected decrease of the band-gap energy. The trend then reverses and the PL peak energy decreases as expected with further increase of temperature. We use Varshni's equation

$$E(T)=E(T=0)+\alpha T^2/(T-\beta), \tag{2}$$

with values of $\alpha=5.08\times10^{-4}$ eV/K and $\beta=996$ K (Ref. 9) to describe the expected temperature dependence of the $Al_xGa_{1-x}N$ band gap. The temperature dependence described by Eq. (2) is plotted with solid line in Fig. 2. Comparison of this line with the observed PL temperature dependence 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) between the low- and high-temperature behaviors is indicated for each sample in Fig. 2. As shown in Fig. 2, 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 AlGaN.[10] The systematic behavior exhibited by data of Fig. 2, 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 InGaN/GaN multiple quantum wells[11] and pseudomorphic AlGaN/GaN heterostructures.[12]

The fact that T_m increases with increased Al content shows that the characteristic localization energy resulting from random alloy disorder increases with Al content in the Ga-rich AlGaN alloy. In fact, the temperature dependence of PL peak energy shown in Fig. 2 may be used to estimate the characteristic localization energies E_0 for the various AlGaN 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. 2 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 AlGaN samples. Figure 3 shows the effective decay time (τ_{eff}) measured at the PL peak energy plotted as a function of Al content x. Here, τ_{eff} is defined as the time at which the PL intensity decays to $1/e$ of the maximum intensity. This definition for decay time is used because, as seen in the inset of Fig. 3, the PL decay is only single exponential for the lowest Al-content alloy samples. It is clear from Fig. 3 that the effective lifetime increases with increased Al content. Specifically, τ_{eff} 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 AlGaN samples, excitons are ener-

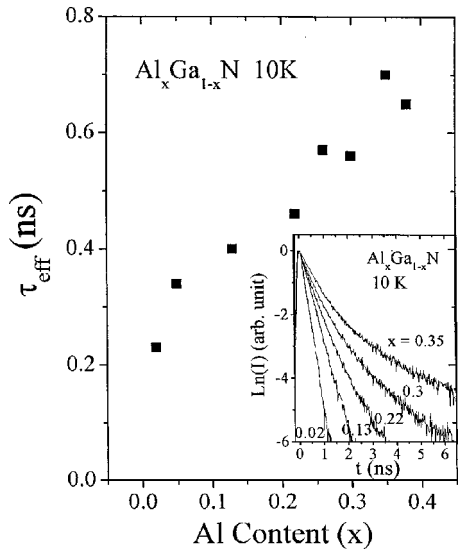

FIG. 3. Effective PL decay time τ_{eff}, as a function of the Al content in $Al_xGa_{1-x}N$ alloys. Decay profiles were measured at the PL emission peak energies at $T=10$ K. The inset shows a semilogarithmic plot of the temporal response for several representative Al contents.

getically 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) result in a longer radiative lifetime.

The emission energy dependence of the PL decay has also been studied at 10 K for the AlGaN samples. As shown in Fig. 4, τ_{eff} increases with decreasing emission energy for all of the represented samples. Such a dependence of decay lifetime on emission energy is a well-known manifestation of a localized exciton distribution within a semiconductor alloy.[15,16] Within the localization model, highly localized (lower-energy) excitons decay primarily via radiative recombination while less localized (higher-energy) excitons exhibit a decreased decay time due to the additional channel of transfer to lower-energy sites. The data of Fig. 4 are qualitatively in agreement with this model.

In summary, we have investigated the optical properties of the $Al_xGa_{1-x}N$ alloys ($0 \leqslant x \leqslant 0.35$) 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 lifetime and its emission energy dependence have been measured and discussed in terms of the effects of tail states due to alloy fluctuation in $Al_xGa_{1-x}N$ 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 $Al_xGa_{1-x}N$ 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 $Al_xGa_{1-x}N$, 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 photodetectors, it should have less effect on heterostructure field-effect transistors and laser diodes, where current injection involves majority-carrier transport.

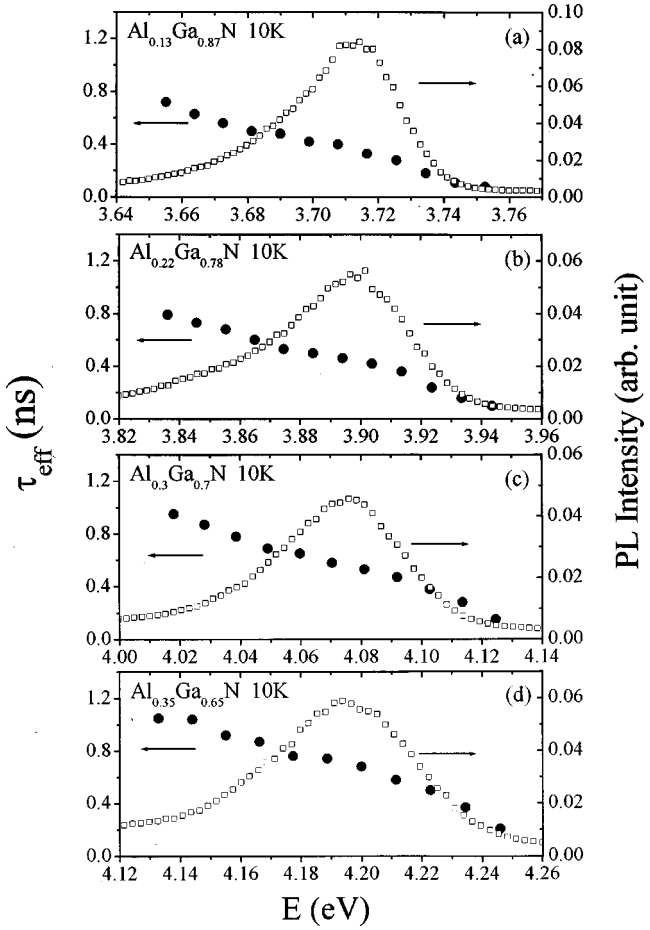

FIG. 4. The emission energy dependence of the effective PL decay lifetime τ_{eff}, for the main emission lines of the $Al_xGa_{1-x}N$ alloys for (a) $x=0.13$, (b) 0.22, (c) 0.3, and (d) 0.35 measured at 10 K.

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