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Citation: Applied Physics Letters **85**, 3047 (2004); doi: 10.1063/1.1802379
View online: http://dx.doi.org/10.1063/1.1802379
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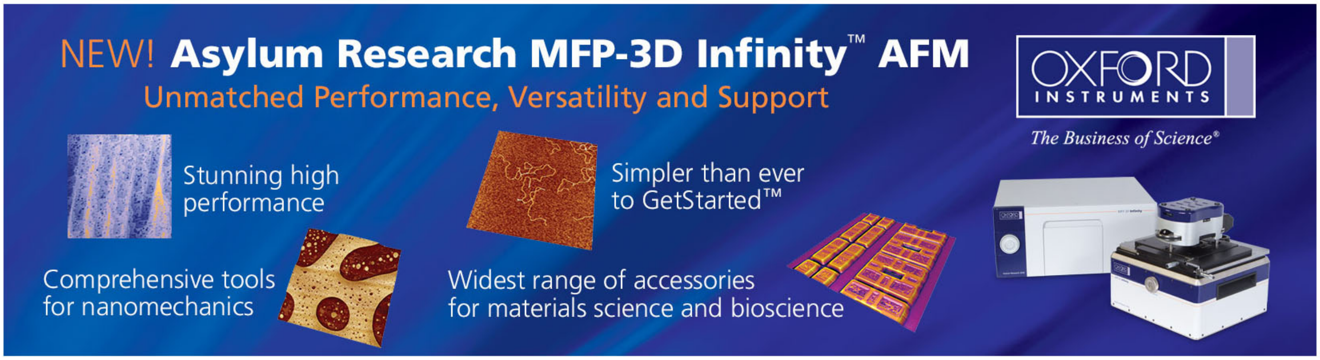
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HTML ABSTRACT + LINKS

APPLIED PHYSICS LETTERS VOLUME 85, NUMBER 15 11 OCTOBER 2004

Microphotoluminescence spectra of hillocks in $Al_{0.11}Ga_{0.89}N$ films

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(Received 31 March 2004; accepted 6 August 2004)

The spatial variation of the optical properties of hillocks in $Al_{0.11}Ga_{0.89}N$ films has been studied by using microphotoluminescence (μ-PL) microscopy. The μ-PL spectrum revealed a strong emission (I_H) at 351 nm from the hillock, besides the near-band-edge emission (I_{nbe}) at 341 nm. Moreover, the I_H intensity increases significantly and its full width at half maximum decreases from ~76 to ~53 meV by probing across the hillock center. These indicated that the hillock structure is a strong emission center. The temperature-dependent μ-PL measurements showed that the I_H also has the S-shape behavior with a transition temperature of ~120 K which is lower than that of I_{nbe}. The redshift of I_H is also smaller than I_{nbe}. Both indicated that the Al composition in hillocks is lower than the surrounding area. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1802379]

AlGaN is a wide band-gap semiconductor with many important applications including high-temperature and high-power electronics, solar-blind photodetectors, blue, and ultraviolet light emitting and laser diodes.[1–3] However, AlGaN grown on sapphire substrate often contains a large amount of dislocations due to lattice mismatch. Even with recent improvements, as due to introduction of low temperature AlN or GaN buffers, AlGaN films grown in this fashion still consist of a mosaic of slightly misoriented domains of microsize with lots of microstructure like dislocations, grain boundaries, V defect, hillocks, pores, etc.[4,5] Thus, the detailed analysis of the spectra and spatial distribution of microstructure is very important to improve material quality. Nevertheless, microstructures sometimes show particular optoelectronic properties. For example, the hexagonal hillocks with dimensions of several microns are often observed on the surface of GaN layers. They are characterized by a high intensity of the band-gap luminescence, while the boundaries of the hillocks are enriched with defects responsible for yellow luminescence at ~2.2 eV.[6,7] So far, the light emission from microstructures is commonly studied by using cathodoluminescence (CL), because the nanoprobe produces secondary electron images in CL with high spatial resolution, as well as point analysis on specific areas. However, no report about microphotoluminescence (μ-PL) studies of the microstructures of III–V nitride semiconductor appeared up to now. In this letter, the spatial variation of μ-PL was studied to characterize the optical properties of AlGaN hillocks. The temperature dependent μ-PL spectra from 10 to 300 K were obtained to show the interesting emission behaviors of the hillock.

The AlGaN films were grown on AlN/sapphire (0001) substrates by low-pressure metalorganic vapor phase epitaxy (MOVPE) system in a horizontal reactor. Trimethylgallium, trimethylaluminum, and ammonia were used as the source precursors for Ga, Al, and N, respectively. Hydrogen was used as the carrier gas. Prior to material growth, the sapphire substrate was annealed to remove any residual impurities on the surface in a H_2 ambient at 1120 °C for 10 min. A nominal 25 nm thick AlN nucleation layer was deposited at 650 °C. The substrate temperature was then raised to 1120 °C to grow a 0.8 μm $Al_{0.11}Ga_{0.89}N$ layer. For μ-PL measurements, a He–Cd laser (Omnichrome 2074) operating at 325 nm was used for above band-gap excitation and was focused to a spot size of 1.5 μm by a microscope objective (100×, 0.5 numerical aperture). The signals were collected by the same objective lens into a monochromator (ARC-500) with both the entrance and exit slits opened to about 50 μm so that the spectral resolution is about 0.2 nm.

Optical examination of the $Al_{0.11}Ga_{0.89}N$ layers revealed particular hillocks with a density of 1×10^6 cm^{-2} covering the substrate surface in Fig. 1(a). The base size of the hexagonal hillock is about 6 μm after 1 h growth. From the three-dimensional (3D) atomic force microscopy (AFM) image (see inset), the hillock has the shape of a regular point-topped pyramid with a height about 200 nm. A series of μ-PL spectra were taken at different locations along a dihedral direction across the hillock as shown in Fig. 1(b). The position label indicates the approximate distance from the hillock center. The spectra are dominated by the near-band-edge emission (I_{nbe}) at 341 nm as the probe spot is far away from the hillock. When it is focused on the hillock, the most significant change in the μ-PL spectra is the appearance of an extra peak (I_H) at 351 nm. Obviously, this strong and prominent emission is related only to the hillock. Note that, although the I_{nbe} is still present at 341 nm, it is so weak that is submerged in the strong and broad 351 nm band. Hoffmann *et al.*[8] reported that CL spectra showed the band-gap gradient from the base to the top of the selective growth GaN pyramids of 5 μm width and 10 μm height. In his report, the emission peak is 355.6 nm at the top of the pyramid and strongly redshifted to 360.6 nm at the pyramid base. The different emission energies reveal the gradual relaxation of strain along the pyramid. However, no peak shift of I_H from the hillock edge to the center was observed in our study. Thus, it suggested that the stress was considerably small in AlGaN hillock which has a rather flat top region in contrast to the steep pyramid.

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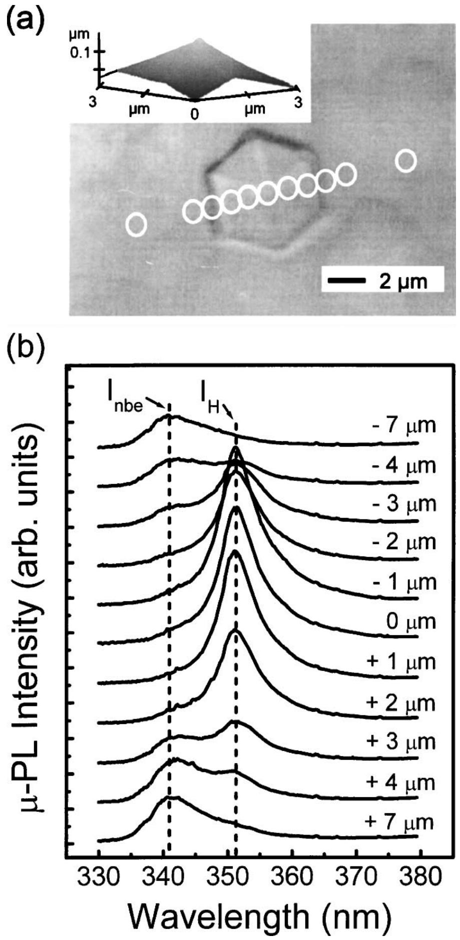

FIG. 1. (a) Optical microscope (OM) image of hillock in AlGaN films; (b) the room temperature μ-PL spectra taken at different locations on OM image. The inset shows the 3D AFM images of hillock.

The spatial μ-PL intensity and full width at half-maximum (FWHM) distribution of the I_{nbe} (341 nm) and I_H (351 nm) are shown in Figs. 2(a) and 2(b), respectively. The I_H intensity at the hillock center is about five times larger than the I_{nbe} intensity far from the hillock. We noticed that the FWHM of I_{nbe} obtained far away from the hillock is ~77 meV that is close to the report of Kim *et al.*[9] Moreover, the FWHM of I_H decreased from ~76 meV at the hillock edge to ~53 meV at the hillock center. The high intensity and narrow FWHM of I_H indicated that the hillock structure is an efficient emission center. From the 3D AFM image, a nipple structure appeared on the top of the AlGaN hillock [see the inset of Fig. 1(a)]. Since the quantum dot (QD)-like structure was observed on the top of AlGaN/GaN selectively grown pyramid,[10] it suggested that the QD-like structure is likely formed on the hillock top.

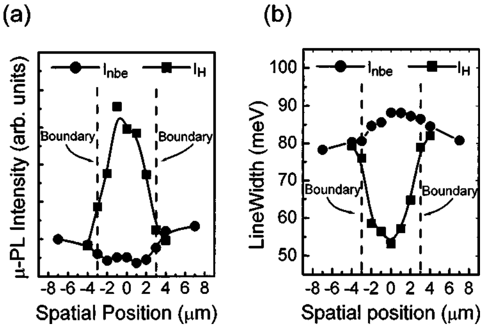

FIG. 2. Spatial μ-PL (a) intensity and (b) full width at half-maximum (FWHM) distribution of I_{nbe} and I_H, respectively.

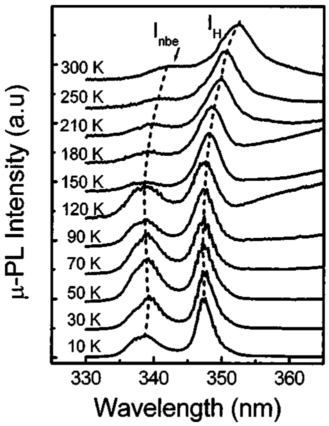

FIG. 3. Temperature dependent μ-PL measurements of AlGaN hillock from 10 to 300 K.

To further examine optical properties of the AlGaN hillock, we also carried out temperature dependent μ-PL measurements from 10 to 300 K. In order to observe both I_{nbe} and I_H peaks, the laser beam was moved to the edge of the hillock to cover both the hillock and the plain region. The temperature dependent μ-PL spectra were shown in Fig. 3. At low temperature, a weak shoulder was observed in the I_{nbe} peak. The energy separation between these two μ-PL structures of the I_{nbe} peak is about 20 meV. Are they donor bound *A* exciton and free *A* exciton? Are they ground state *A* exciton and ground state *B* exciton? No, we excluded both possibilities because, in the case of GaN, the energy separation for both cases is less than 10 meV.[11] We therefore assigned the major peak as a ground state $(n=1)$ free *A* exciton luminescence and the weak shoulder as the excited state $(n=2)$ *A* exciton transition. The FWHM of the ground state *A* exciton peak is so large that it might cover the donor bound exciton and the ground *B* exciton. In the case of GaN, the energy separation between the $n=1$ *A* exciton and $n=2$ *A* exciton is about 20 meV, which is close to our μ-PL data. As the temperature was raised, both transitions merged together. Peak fit procedure was carried out to extract the temperature dependent PL intensity and peak position as shown in Figs. 4 and 5, respectively. In Fig. 3, one clearly sees that the I_{nbe} photon energy increases initially then decreases with temperature and forms an "S shape" curve; as does the I_H. It was

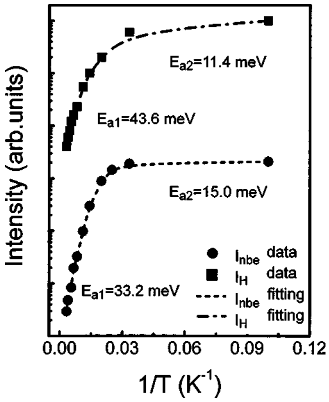

FIG. 4. Integrated PL intensity as a function of temperature for I_{nbe} and I_H.

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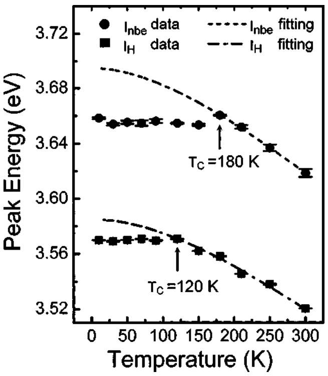

FIG. 5. Temperature dependence of the emission peak of I_{nbe} and I_H. Arrows indicate the transition temperature T_C.

known that the "S shape" is due to localization phenomena induced by alloy inhomogeneous in the AlGaN films.[12] The localization phenomena can be further corroborated by Figs. 4 and 5. In Fig. 4, the integrated PL intensity was plotted versus the inverse temperature, namely the Arrhenius plot, for both I_{nbe} and I_H over the temperature range of 10–300 K. The dashed and dash-dotted curves are fits to the data using the following formula:[13]

$$\frac{I(0)}{I(T)} = 1 + C_1 \exp\left(-\frac{\Delta E_{a1}}{kT}\right) + C_2 \exp\left(-\frac{\Delta E_{a2}}{kT}\right), \qquad (1)$$

where ΔE_{a1} is the free exciton binding energy, ΔE_{a2} is the localization energy, C_1 and C_2 are fitting constant. From the slope, ΔE_{a1} is calculated to be 33.2 meV and ΔE_{a2} is 15.0 meV for I_{nbe}, and ΔE_{a1} is 43.6 meV and ΔE_{a2} is 11.4 meV for I_H, respectively. The localization energies of 15.0 and 11.4 meV result from the localization, due to alloy (potential) fluctuation in AlGaN, of the ground state exciton outside the hillock region and in the hillock region, respectively.

The temperature dependence of the emission energy is then shown in Fig. 5, for analyzing the localization energy. The expected temperature dependence (dashed lines) was calculated by using the Varshni's equation:[14]

$$E_{\mathrm{g}} = E_0 - \frac{\alpha T^2}{\beta + T} \qquad (2)$$

with $\alpha = 6.31 \times 10^{-4}$ eV/K and $\beta = 2584$ K for I_{nbe}, and $\alpha = 21.2 \times 10^{-4}$ eV/K, $\beta = 606$ K for I_H. The observed μ-PL temperature dependence follows Eq. (1) at high temperatures and deviates from it below a transition temperature T_c. The transition temperatures are ~180 and ~120 K for I_{nbe} and I_H, respectively, which are both higher than another report for the $\mathrm{Al_{0.11}Ga_{0.89}N}$ films.[9] Nevertheless, the transition temperatures of ~180 and ~120 K are close to the localization energy of 15.0 and 11.4 meV. They reflect that the localization effect is strong in our sample.

Moreover, the transition temperature of I_{nbe} line was higher than that of the I_H line. The redshift of the I_{nbe} (~35.8 meV) at low temperature relative to the dashed line is larger than that of I_H (~17.6 meV). It has been reported that the increase of transition temperature (T_C) and redshift are due to the localization energy which increases with the Al content.[9] Our experimental results also support that the Al composition in the hillock is smaller than surrounding area in the $\mathrm{Al_{0.11}Ga_{0.89}N}$ films. The recent x-ray energy dispersive spectroscopy observations (not shown) also confirm this. Thus, the hillock microstructure has lower Al composition than the regions free of hillocks in the AlGaN films.

In summary, we have measured the optical properties of hillocks in $\mathrm{Al_{0.11}Ga_{0.89}N}$ films by using the μ-PL microscopy. The large intensity and narrow FWHM of I_H in the hillock structure indicated that it is a strong emission center. The temperature dependent μ-PL spectra showed that the I_H has the *S*-shape behavior with a transition temperature of ~120 K reflecting the strong localization in the hillock. The lower transition temperature and smaller redshift of I_H than that of I_{nbe} suggest that the Al composition is lower in hillock than in other parts of AlGaN films.

The authors would like to acknowledge the support by the National Science Council of the Republic of China under Contract Nos. NSC92-2112-M009-028 and NSC92-2112-M009-022.

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