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# Near-field optical spectroscopy and microscopy of self-assembled GaN/AlN nanostructures

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The spatial distribution and emission properties of small clusters of GaN quantum dots in an AlN matrix are studied using high-resolution electron and optical microscopy. High-resolution transmission electron microscopy reveals near vertical correlation among the GaN dots due to a sufficiently thin AlN spacer layer thickness, which allows strain induced stacking. Scanning electron and atomic force microscopy show lateral coupling due to a surface roughness of ~50–60 nm. Near-field photoluminescence in the illumination mode (both spatially and spectrally resolved) at 10 K revealed emission from individual dots, which exhibits size distribution of GaN dots from localized sites in the stacked nanostructure. Strong spatial localization of the excitons is observed in GaN quantum dots formed at the tip of self-assembled hexagonal pyramid shapes with six $[10\bar{1}\bar{1}]$ facets. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1851005]

The successful development of short wavelength light-emitting diodes and the most recent realization of nitride-based quantum dot lasers have stimulated great interest in the application of quantum confined structures for blue and ultraviolet optoelectronic devices.[1,2] In particular, III-nitride-based self-assembled quantum dots (QDs) are very promising for a wide range of commercial applications.[3–7]

The study of self-assembled GaN quantum dots presents a challenge, as the placement of individual dots is difficult to control during the epitaxial growth process, and the dot density can be quite high.[8,9] Thus traditional experimental techniques often only allow simultaneous observation of large ensembles of quantum dots where inhomogeneous broadening washes out many of the interesting features. We have investigated the optical properties of GaN QD and have observed that the built-in strain fields significantly influence the radiative recombination lifetime.[10–12] The role of size distribution of the QDs on the radiative emission process is not yet clear.[12]

Recent reports on the near-field optical properties of GaN QD studied using illumination mode are limited by the low spatial resolution due to carrier diffusion accentuated by a large dot size inhomogeneity.[13,14] The contribution from individual dots or coupled QD clusters exhibiting narrow near-field photoluminescence (PL) line shape (~ few milli-electron-volts) from high-spatial resolution is yet to be reported. The PL line shape of individual dots in the GaN system is expected to be significantly broader than GaAs- or InP-based QDs due to broadening induced by a significantly larger LO phonon scattering rate. In this letter we present the near-field optical emission characteristics from a cluster of a few GaN QDs with very high spatial resolution. This letter will also discuss the lateral and vertical electronic coupling of dots caused by interdot scattering of carriers.

The sample consisted of 40-stacked planes of GaN QDs in AlN matrix, grown on a sapphire substrate by molecular beam epitaxy.[10,11] The buffer layer consisted of alternating layers of AlN and GaN grown on a thin layer of initiation AlN buffer. Quantum dots were formed by growing a GaN layer at just above the critical thickness, which allows it to maintain its pseudo-coherence with the AlN lattice. The QD planes were separated by 2-nm-thick AlN barriers and capped by an AlN layer 3 nm thick. The thin AlN spacer layer in the presence of self-assembly process led to the formation of GaN QDs embedded within two-dimensional GaN/AlN quantum-well-like structures.

The surface morphology of GaN nanostructures was studied using scanning electron (SEM) and atomic force microscopy (AFM). Shown in Fig. 1(a) is a topographical map of an area of $1\times 1\ \mu m^2$ measured using AFM, which exhibits a honeycomb feature at the surface due to modulation of the AlN cap layer by the underlying GaN QDs. Due to the thin cap layer, a high density ($3\times 10^{10}$ dots/cm$^2$) of the QDs as well as a strong inhomogeneity of their lateral dimensions, ranging from 30 up to 50 nm, is clearly evidenced. The height of these capped QDs range from 7 to 10 nm. This nonuniform surface topology induces inhomogeneous broadening in the far-field emission spectrum due to lateral coupling. It was shown by Widmann *et al.*[8] that the QD size varies significantly depending on whether the QDs are allowed to evolve under vacuum before covering with AlN, or not, as a result of a ripening mechanism. This variation in size can lead to a large variation in the piezoelectric effect in the self-assembled GaN layers. Our experiments indicate that ripening leads to reduced footprint and increased height for a larger aspect ratio, as the dots are not spherical.[11]

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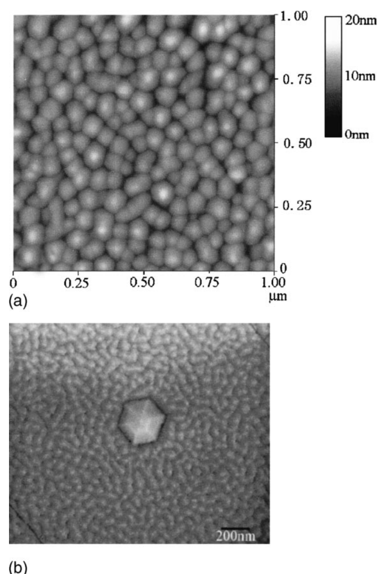


FIG. 1. (Color online) (a) AFM image showing the surface of GaN dots covered with 2 nm AlN cap layer. (b) SEM images showing the surface morphology and self-assembly of a hexagonal pyramid shape GaN structure with ~300 nm diameter.

The modulations at the surface are also observed in the high magnification SEM images [Fig. 1(b)]. The SEM spatial patterns of the capped GaN QDs showing island-like features can be correlated with the sample morphology as measured by AFM. A large hexagonal GaN pyramid is self-assembled on the AlN cap on the surface of the GaN QD layers with a radius of curvature no more than 300 nm. The faces of the pyramids are the $\{10\bar{1}\bar{1}\}$ planes as evidenced by the angle between the inclined edge and the base of the pyramid. The measured angle of around 58°–60° is in good agreement with the calculated angle of 58.4° using the GaN lattice parameters of $c=5.185$ Å and $a=3.189$ Å. The formation of the pyramids indicates that the $\{10\bar{1}\bar{1}\}$ surfaces are self-assembled preferentially compared to the $[000\bar{1}]$ surface. Thus it can be inferred that $\{10\bar{1}\bar{1}\}$ surfaces have the lowest surface potential with respect to the self-assembly process. The tip of the pyramid is very sharp with a diameter measured to be less than 20 nm.

The optical emission properties were investigated by studying the PL characteristics in the far-field and near-field limit. Figure 2 shows the time-integrated far-field PL spectrum of QDs at room temperature, measured using a frequency tripled Ti:sapphire laser delivering pulses of 10 ps duration at 267 nm (photon energy 4.655 eV). The peak emission energy was close to 3.67 eV, with a broad linewidth of ~250 meV arising due to the inhomogeneous strain and also the lateral and vertical coupling amongst the QDs in the various layers. The PL peaks from the QD layers are shifted to a higher energy as compared to the underlying bulk GaN for the wurtzite phase (band gap energy $E_g=3.45$ eV). The inhomogeneously broadened PL line shape can be attributed to the emission from optically pumped carriers thermalized in the statistically distributed ground states of the probed QD, which vary in energy because of small variations in size, composition, and strain. The inset shows the temperature dependence of the PL intensity and emission linewidth. The relatively temperature-insensitive PL emission below 125 K occurs as the radiative decay of excited carriers dominates the recombination process.[12] However, above 125 K the PL intensity decreases more severely with increasing temperature due to increase in nonradiative recombination. The relatively small change in thermally induced PL peak energy shift (0.168 meV/K) is due to strong carrier confinement in the QDs with the redshift at higher temperatures likely due to a reduction of the exciton-Bohr radius that makes the excitons less polar.[9] The photoluminescence excitation spectrum measured using a Xe lamp shows absorption from the GaN nanostructures from higher energies. A large Stark shift exceeding 400 meV is observed to the built-in strain in the QD layers.

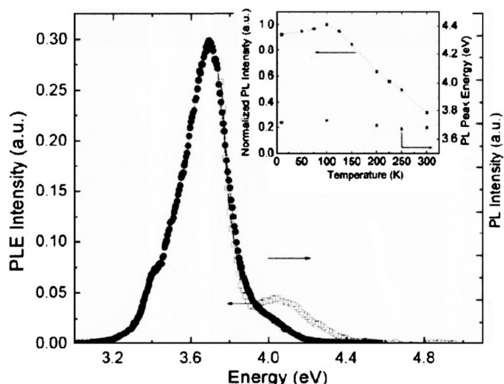


FIG. 2. (Color online) Far-field PL spectrum of GaN QDs with temperature dependence of PL intensity and PL emission energy shown in the inset.

Compared to bulk or GaN/AlN quantum well, a larger PL efficiency has been observed for this QD system despite a relatively shorter radiative lifetime of ~500 ps.[12] The role of inhomogeneity in the far-field PL spectra due to spatial QD distribution has been investigated via near-field PL spectroscopy. We have used a commercial (NSOM)[6] operating in illumination mode at 10 K for measuring the spatially and spectrally resolved PL spectra. A tapered, metal-coated optical fiber having a nominal apical aperture of 30 nm was exploited as the nanosource through which the sample was irradiated with UV light (325 nm delivered by a He–Cd cw laser). Figures 3(a)–3(c) show monochromatic PL images within a 450 nm$^2$ area, in which the detection wavelengths are 343±1, 345±1.5, and 355±1 nm, respectively. The NSOM-PL in Figs. 3(a) and 3(c), which originates from a much smaller number of QDs compared to dot density obtained from surface features in Fig. 1, consists of a number of sharp spectral features of similar amplitude with full width at half maximum ranging from 500 $\mu$eV to 2 meV. We observe that the bright areas in Figs. 3(a) and 3(c) are larger than the dark ones. It may be that the honeycomb-like QD features observed at the surface are not entirely optically active and larger islands or smaller QDs presumably act as nonradiative recombination centers. The nucleation of relatively larger dots emitting at lower energies, i.e., ~3.49 eV [Fig. 3(c)] is more prevalent compared to the smaller dots emitting at higher energies ~3.62 eV [Fig. 3(a)]. The brighter regions A, B, C, E, F in Figs. 3(a) and 3(c) are an indication of strong confinement and a correlation in the vertical direction.

As the optically active part of our sample consists of 40 QD or QW planes, several layers with varying QD spatial distribution are excited simultaneously. So even if the NSOM probe is located above a region of the first QD layer

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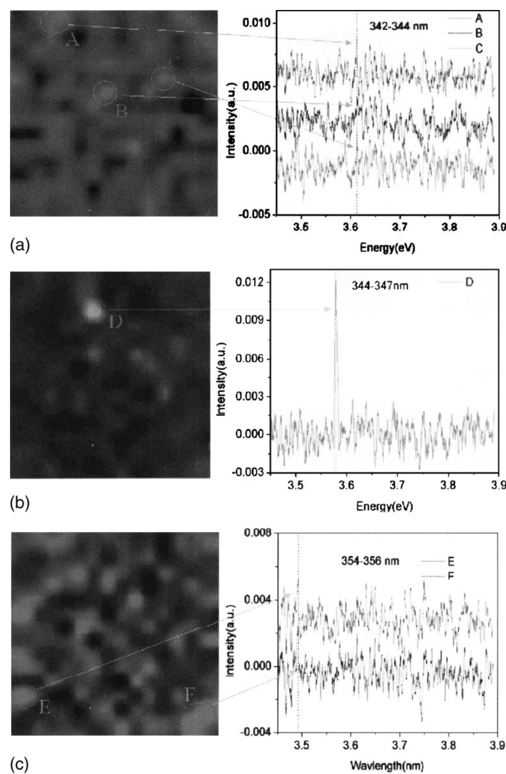


FIG. 3. Near-field luminescence spectra from GaN QD. (a) Spatial and spectrally resolved PL measured at 342–344 nm. (b) Spatial and spectrally resolved PL measured at 344–347 nm. (c) Spatial and spectrally resolved PL measured at 354–356 nm.

containing large dots, luminescence at high energy still originated from the underlying QD planes. This suggests that, while the intense background signal is due to the luminescence of a large part of the active region that cannot be spatially resolved, the localized modulations are due only to the morphology of the dots located on the outermost layer, which can be stronger in the presence of vertical correlation.

To gain insight into the origin of the light emission and the influence of spatial variation of GaN QDs and quantum wells on the intensity, we performed cross-sectional transmission electron microscopy (TEM). Samples were processed in a dual-beam SEM/FIB (FEI Nova 600) using a Ga ion beam accelerating voltage of 5 kV, followed by examination in a Tecnai F20 analytical HRTEM. A near vertical correlation of the GaN dots ~30 nm in width is observed from STEM-HAADF image (not shown here), with some dot assemblies being correlated at an angle slightly off vertical. It is also observed that the width of these dots and their period correspond to the surface texture observed in AFM and SEM images (Fig. 1). A HRTEM image shown in Fig. 4 illustrates that 1.1- to 2-nm-high GaN QD-like clusters are embedded in GaN/AlN QW-like structures.

It is reasonable to assign the high energy PL spectrum (Fig. 2) to the superposition of blueshifted near-band-gap excitonic emissions arising from clusters of dots with size smaller than the excitonic Bohr radius for GaN ($a_B \sim 3$ nm), at least in the growth direction (3 nm). The distribution of the dots in the vertically stacked layers also explains the background emission from spatially unresolved underlying QD and QW layers. The strong room-temperature PL is due to the vertical correlation of the dots, while the lateral coupling at the surface and underlying layers results in nonradiative recombination. Contrast observed in the NSOM images (Fig. 2) may be due to stronger emission from dot clusters correlated more closely to the vertical direction, as opposed to dot clusters correlated off-axis observed in Fig. 4.

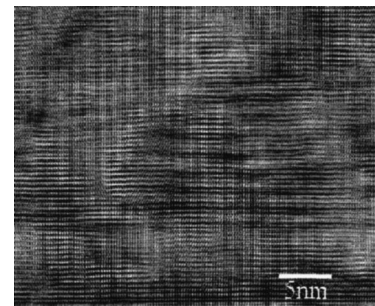


FIG. 4. HRTEM image showing ⟨1−1−1−0⟩ cross section of stacked layers GaN dots.

An intense emission is observed from a 20-nm-diam area "D" at an intermediate energy regime 3.59 eV (~345 nm), with a small background emission [Fig. 3(b)], implying that the source of this strong PL is significantly different from the emission of larger QDs or smaller QDs shown in Figs. 3(a) and 3(c). The emission at 3.39 eV is particularly strong in the vicinity of the hexagonal pyramid structure shown in Fig. 1(b). The emission is likely due to the localization of excitons in GaN QD formed at the tip of the hexagonal pyramid. The strong room-temperature PL is due to the vertical correlation of the dots, while the lateral coupling at the surface and underlying layers results in nonradiative recombination resulting in reduced PL emission at higher temperatures.

In conclusion we have studied the luminescence features of a sample of GaN quantum dots with a spatial resolution of 50 nm. The luminescence spatial patterns near the peak energy were measured, showing island-like features that can be correlated with the sample morphology as measured by AFM and SEM techniques. Cross-sectional TEM investigations revealed 1.1- to 2-nm-thick QD-like GaN layers correlated vertically or slightly off-axis, which may affect the vertical NSOM intensity data.


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