Strain relaxation induced microphotoluminescence characteristics of a single InGaN-based nanopillar fabricated by focused ion beam milling

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A freestanding nanopillar with a diameter of 300 nm and a height of 2 μm is demonstrated by focused ion beam milling. The measured microphotoluminescence (μ-PL) from the embedded InGaN/GaN multiple quantum wells shows a blueshift of 68 meV in energy with a broadened full width at half maximum, ~200 meV. Calculations based on the valence force field method suggest that the spatial variation of the strain tensors in the nanopillar results in the observed energy shift and spectrum broadening. Moreover, the power-dependent μ-PL measurement suggests that the strain-relaxed emission region exhibits a higher radiative recombination rate than that of the strained region, indicating potential for realizing high-efficiency nanodevices in the UV/blue wavelength range. © 2008 American Institute of Physics. [DOI: 10.1063/1.2965461]

Recent progress in III-nitride nanofabrication technologies have opened up many possibilities for advanced optoelectronic devices in the UV/blue wavelength range, such as nanorod light emitting diode arrays, single nanowire lasers, single nanorod photodiodes, and nanowire sensors. The development of such sophisticated devices relies on the understanding of the electrical and optical properties of III-nitride nanostructures, which can be significantly altered from those of bulk. Although multiple studies have been made on the collective characteristics of GaN-based nanorods or nanowires, few reports have investigated the modified material and optical properties of a single nanostructure due to the limitations in fabrication. A theoretical model that describes the strain variation of GaN/InGaN multiple quantum wells (MQWs) embedded in a nanopillar had yet to be developed. In this work, a freestanding nanopillar with a diameter of 300 nm and a height of 2 μm is fabricated using a focused ion beam (FIB). The measured microphotoluminescence (μ-PL) from the embedded InGaN/GaN MQWs shows an energy shift of 68 meV and a slightly broadened full width at half maximum (FWHM), which are caused by the strain relaxation of MQWs in the nanopillar. Calculations based on the valence force field (VFF) method reveal the correlation between the emission characteristics and the spatially varied polarization field. Moreover, the power-dependent μ-PL measurement suggests that the strain-relaxed emission region of the nanopillar exhibits a higher radiative recombination rate than that of the strained region, making strain-relaxed InGaN-based nanopillars a promising candidate for high-efficiency UV/blue light sources.

As seen in Fig. 1(a), the epitaxial structure of the device consists of a 2 μm thick GaN nucleation layer, a 2 μm thick Si doped n-GaN cladding layer, ten periods of 2.5 nm thick In0.2Ga0.8N MQWs separated by a 10 nm thick GaN barrier, a 100 nm thick AlGaN cladding layer, and a 100 nm thick Mg-doped p-GaN layer. To prepare for the FIB milling, a 100 nm thick SiNx was deposited by plasma enhanced chemical vapor deposition, followed by a 200 nm thick Al layer using electron-beam evaporation. Then, a platinum (Pt) mask with a thickness of 500 nm was formed by the FIB deposition mode in an area of 500×500 nm2 on the Al layer to provide localized protection from the ion beam sputtering. Finally, a 30 keV Ga ion beam with a current of 100 pA was applied to remove the material around the Pt mask in a four-step milling process. Figure 1(b) shows the field-emission scanning-electron micrograph (FESEM) of the fabricated single nanopillar. The fabricated well area was approximately 5×5 μm2, while the dimensions of the fabricated nanopillar were ~300 nm in diameter and ~2 μm in height. The μ-PL characteristics were investigated for the fabricated

FIG. 1. (Color online) (a) Schematic of the epitaxial structure and the deposited materials for nanopillar fabrication using a focused ion beam. (b) The field-emission scanning-electron micrograph (FESEM) image of the fabricated freestanding nanopillar with a diameter of 300 nm and a length of 2 μm.

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The device was first put on a piezoelectric translator stage with a resolution of 2 nm, where the He–Cd laser beam with a wavelength of 325 nm was focused on to a spot size of ~3 μm by a 40× objective lens with a numerical aperture of 0.5. The emission was then collected by the same objective lens, transmitted to a charge-coupled device (CCD) and a 0.32 m spectrometer, and measured at a spectral resolution of 0.1 nm. After collecting the μ-PL spectrum and the scanning image of the nanopillar with the Pt mask, the mask was removed by the metal etcher and the same structure was scanned again.

The measured μ-PL spectra at an excitation power of 10 mW are shown in Figs. 2(a) and 2(b) for the device with and without Pt mask, respectively. The respective CCD images with a scanning area of 5×5 μm² are displayed as the insets. As shown in the inset of Fig. 2(a), the photoluminescence from the freestanding nanopillar is weak, since the Pt mask blocks a significant amount of the excitation on to the nanopillar. Moreover, since the measured spectrum is very similar to that of the as-grown planar structure, we believe that the photoluminescence is mainly contributed by the emission from the periphery of the well. After the mask removal, the photoluminescence from the nanopillar becomes comparable to that from the well region, as shown in the inset of Fig. 2(b). However, the spectrum without the Pt mask shows additional photoluminescence in the shorter wavelength region, which is likely to originate from the freestanding nanopillar. The measured spectra are then analyzed by the Lorentzian curve fitting. As shown in Fig. 2, two and three dominant Lorentzians are found for the device with and without Pt mask, respectively, denoted as Lorentzians 1, 2, and 3. The peak wavelengths and the FWHMs of Lorentzians are also plotted. The color maps show the calculated strain variations in a freestanding, rhombus-shaped In0.2Ga0.8N/GaN nanopillar with a height of Z=300 nm and an edge width of X=180 nm for (a) in-plane strain tensor ε_xx and (b) vertical strain tensor ε_zz. The vertical distributions of ε_xx and ε_zz at the center of the nanopillar, X=95.67 nm, and at the edge, X=1.91 nm, are also plotted.
containing an In0.2Ga0.8N electric field can be estimated using a simplified structure
The energy shift due to the difference in the present piezoelectric tensors in the
relaxation region, as shown in Fig. 2

FIG. 4. (Color online) (a) The measured power-dependent μ-PL spectra of the
device without the Pt mask with excitation powers ranging from 0.88 to 15.83 mW. Lorentzian analyses of the measured spectra plotted for (b) the peak intensities, (c) the emission wavelengths, and (d) the FWHMs of the PL from the strained and the strain-relaxed regions.

\[ X = 1.91 \text{ nm}, \] 
where the change of strain from the pillar center is most significant, there exist substantial strain variations on the QW/barrier interfaces and a slight tensile strain of \( e_{zz} \) in the barriers. Moreover, as shown in Fig. 3(b), the strain relaxation of \( e_{zz} \) takes place within a distance of \( \sim 10 \text{ nm} \), much shorter than that of \( e_{xx} \). We also observe a vertical distribution of \( e_{zz} \) in the \( Z \) direction, mainly arising from the stacking of MQWs. Both in-plane and vertical strain tensors contribute to the polarization field \( P_{e} \) via the expression

\[ P_{e} = e_{31}E_{xx} + e_{33}E_{yy} + e_{33}E_{zz}, \] 

where \( e_{31} \) and \( e_{33} \) are piezoelectric coefficients. Based on the calculated strain shown in Fig. 3, we obtain the piezoelectric filed of \(-0.0276 \text{ C/m}^2\) in the center of the nanopillar and of \(-0.01 \text{ C/m}^2\) at \( X = 1.91 \text{ nm} \) using parameters from Ref. 12. The energy shift due to the difference in the present piezoelectric field can be estimated using a simplified structure containing an In0.2Ga0.8N/GaN single QW with a thickness of 2.5 nm located between a 2 μm thick n-GaN and 100 nm thick p-GaN with doping concentrations of \( 10^{18} \text{ cm}^{-3} \) and \( 10^{12} \text{ cm}^{-2} \). The carrier density \( n_{D} \) is estimated to be \( \sim 7.0 \times 10^{12} \text{ cm}^{-2} \) under an excitation power of 10 mW. An energy shift of \( \sim 210 \text{ meV} \) is calculated, corresponding to \( \lambda \sim 415 \text{ nm} \), which matches the blue edge of the PL from the strain-relaxed region, as shown in Fig. 2(b). Since the center of MQWs still suffers from strain with its magnitude comparable to that of the as-grown planar structure, the photoluminescence from the center of the pillar contributes to the red-edge emission, which is also partially overlapped with the PL from the strained region, as expected. Hence, we believe that the majority of the nanopillar photoluminescence originates from the strain-relaxed region, within a distance of \( \sim 40 \text{ nm} \) near the edge of the nanopillar. Moreover, the variation of strain tensors in the relaxation region results in a spatially varied polarization field, leading to the broadened emission spectrum.

Figure 4(a) shows the measured power-dependent μ-PL spectra of the device without the Pt mask with excitation powers varying from 0.88 to 15.83 mW. The peak intensities, emission wavelengths, and FWHMs of the PL from the strained and the strain-relaxed regions are derived from the Lorentzian fitting of the measured spectra, as plotted in Figs. 4(b)–4(d), respectively. As seen in Fig. 4(b), the PL intensity of the strained region first increases with excitation power, and then gradually saturates for excitation powers larger than 6.05 mW. The saturation can also be observed in the wavelength shift and the FWHM, as shown in Figs. 4(c) and 4(d), respectively. On the other hand, the peak intensity and the FWHM of the PL from the strain-relaxed region are monotonically increasing with the excitation wavelength. The wavelength blueshift is also more significant than that of the strained region. As the carrier density in InGaN MQWs increases with the incident power, the emission wavelength shifts toward smaller wavelengths and the spectrum becomes broader due to band filling and the screening effect of the piezoelectric field.16 However, the radiative recombination process can reduce the band filling effect in MQWs. Figures 4(b)–4(d) suggest that the transition bands are nearly filled in the strained region, where the peak intensity, wavelength shift, and the FWHM vary slowly under high excitation powers. Since the PL from the strain-relaxed region still exhibits the band filling effect under the same excitation condition as the strained region, it is deduced that the radiative recombination rate of the strain-relaxed region is faster than that of the strained region. It is also possible that other processes—including photon extraction and non-radiative recombination—may also be involved, and further studies are necessary to completely clarify the emission mechanism. Nonetheless, we believe that strain-relaxed InGaN-based nanopillars can be a promising candidate for highly efficient UV/blue light sources.

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