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# Amplified Spontaneous Emission of GaN Nanorods†

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Semiconductor nanowires have been the subject of intense study in recent years. In particular, their ability to confine electrons/holes and photons makes them attractive as potential building blocks of nanoscale electronics and optoelectronic devices.[1-3] For UV/blue light emitting devices, the optical properties of gallium nitride (GaN) nanowires are suitable due to their wide band gap (3.4 eV) and the absence of threading dislocation in the lateral dimensions.[4-7] Therefore, the optical properties of GaN nanostructures have been widely investigated, which include stimulated emission processes such as lasing and amplified spontaneous emission (ASE). These can have a positive impact on the optoelectronic devices based on GaN nanowires. In this Note, we examine the stimulated emission process of GaN nanorods using optical excitation of femtosecond laser pulses.

The shapes and lengths of the GaN nanostructures were characterized by scanning electron microscopy (SEM), whose result is presented in Figure 1(a). The diameters of nanorods were in the range of 150-400 nm and the lengths were 500-1000 nm. The as-grown nanorods were excited with a He-Cd laser (325 nm, Kimmon) to obtain a normal photoluminescence spectrum at room temperature (Figure 1(b)). In the UV region an emission peak was observed at 368 nm (3.4 eV). As the exciton binding energy in GaN is in the range of 20-25 meV,[4,8] emission of the exciton state was not expected due to the thermal energy at room temperature (26 meV). Therefore, the UV emission is attributed to band edge emission (BGE), which indicates the band gap energy.[4-8]

To measure the nonlinear optical response, GaN nanorods were excited by the second harmonics (355 nm) of a cavity-dumped oscillator (Mira/PulseSwitch, Coherent, 1 MHz, 710 nm, 150 fs) using a UV microscope objective.[9,10] The emissions were collected by the same objective, resolved spectrally by a monochromator, and detected by a photomultiplier. The shape of BGE was not changed when a low excitation intensity was used ($< 25$ μJ/cm$^2$), as shown in Figure 2(a). The intensity of BGE increased almost linearly in this low excitation intensity regime, which is presented in Figure 2(b). On the other hand, a superlinear increase in the BGE intensity was observed at an excitation intensity of 30 μJ/cm$^2$. In addition, the bandwidth of BGE decreased with increasing excitation intensity above a threshold of 30 μJ/cm$^2$. For example, the full-width at half maximum (FWHM) of the band was found to be 17 nm at an excitation intensity of 10 μJ/cm$^2$, which decreased to 15 nm at an excitation intensity of 50 μJ/cm$^2$, as seen in the inset of Figure 2(a).

The superlinear increase in the BGE intensity and the narrowing of the bandwidth can be attributed to the waveguiding effect.[4,5] As the diameter of the nanorods is smaller

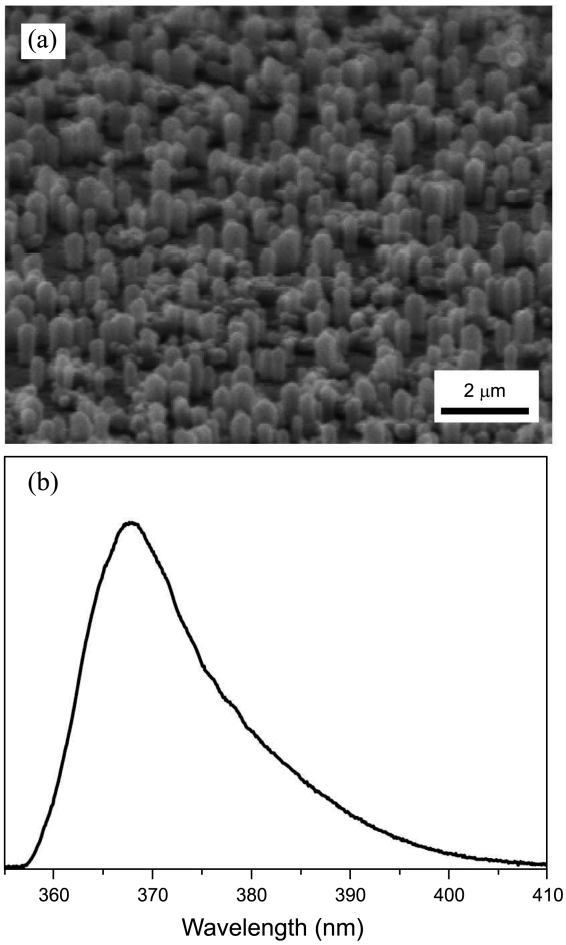


**Figure 1.** (a) Scanning electron microscopy (SEM) image of GaN nanostructures. Scale bar indicates 2 μm. (b) Photoluminescence spectrum of GaN nanorods at room temperature. The peak wavelength was observed at 368 nm, indicating the band gap energy.

†This paper is to commemorate Professor Kook Joe Shin's honourable retirement.

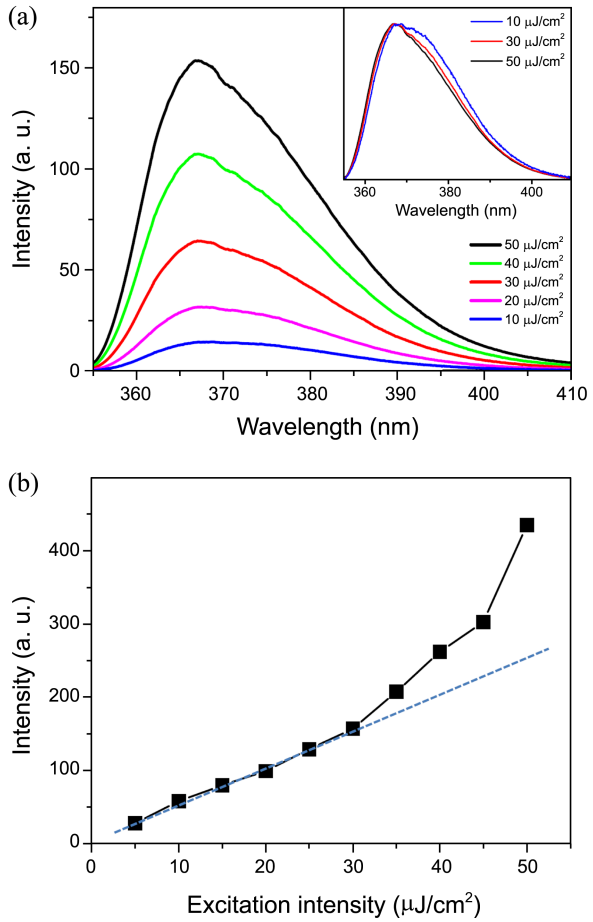


**Figure 2.** (a) The excitation-intensity-dependent emission spectra of GaN nanorods. The shape of the band gap emission (BGE) is nearly identical in the low excitation intensity regime (< 25 μJ/cm$^2$), but decreases above a threshold value (30 μJ/cm$^2$). The inset shows the emission spectra obtained at selected excitation intensities, which indicates that the peak position is slightly blue-shifted with increasing excitation intensity. The intensities are normalized for comparison purposes. (b) The total intensity of BGE as a function of excitation intensity. The intensity increases almost linearly in the low excitation intensity regime, whereas a superlinear increase is observed above the excitation intensity of 30 μJ/cm$^2$.

than (or at least comparable to) the emission wavelength, the electromagnetic waves are guided along the long axis of the nanorods. As a result, stimulated emission processes such as lasing and ASE can be induced. However, when the loss at the end facets of the nanorods is taken into account, the gain length of the nanorods (< 1 μm) may not be sufficient to induce lasing. Assuming the flat end facets, the reflectivity was estimated to be less than 20% due to the small difference in the refractive index between GaN (2.5) and air (1.0).[4,8,11] In previous reports, the gain length for the lasing of GaN nanorods was found to be larger than 5 μm.[4,5] Accordingly, the Fabry-Pérot-type modes, which are a characteristic of lasing, were absent in our GaN nanorods mainly due to their short length and low reflectivity at the end facets. Therefore, we postulate that the ASE process may be responsible for the superlinear increase in the BGE intensity and the narrowing of the bandwidth, while other emissions, such as normal photoluminescence, also contribute to the emission spectrum.

In addition to the narrowing of the bandwidth, after close examination, the peak position becomes slightly blue-shifted with increasing excitation intensity. It is noted that the red-shift is usually observed in the stimulated emission process of GaN, which can be attributed to electron-hole plasma and the resulting band gap renormalization.[4,5] When we assume that incident photons are completely absorbed in the nanorod with a diameter of 150-400 nm due to the large absorption coefficient (~$10^5$ cm$^{-1}$),[4,8] the carrier density in the nanorods is estimated to be in the range of $2 \times 10^{18}$ - $6 \times 10^{18}$ cm$^{-3}$ at an excitation intensity of 50 μJ/cm$^2$. Thus, the carrier density in the nanorods was lower than the Mott density (~$10^{19}$ cm$^{-3}$),[4] which suggests that the band gap renormalization is not highly probable. In addition, the heating effect of the intense excitation is known to induce a red-shift.[4,5] In this regard, the observed blue-shift implies that sample heating was not significant in our experimental setup, mainly because the excitations were carried out by femtosecond pulses. Nevertheless, the peak positions could be affected by other factors especially in high carrier density regime. For example, the band-filling effect changes the bandwidth.[4,12,13] The FWHM of the band was 17 nm at an excitation intensity of 10 μJ/cm$^2$, which is wider than the bandwidth of 15 nm in the normal photoluminescence spectrum (< 1 mW/cm$^2$). In the low carrier density regime, the initially-excited-high-lying states relaxed to lower-lying ones, where normal photoluminescence takes place. Increasing the carrier density, on the other hand, resulted in the saturation of the low-lying states and the occupation of higher-lying states, where the density of states is high.[13] Accordingly, the bandwidth could broaden in the short wavelength region and the peak position also becomes blue-shifted due to the high density of states in higher-lying states. However, due to the stimulated emission process, the bandwidth was also narrowed in both the long and short wavelength region, which induced the whole BGE to appear blue-shifted.

In summary, the optical properties of GaN nanorods were studied by examining BGE using femtosecond laser pulses. The superlinear increase found in the BGE intensity was attributed to an ASE process caused by the waveguiding effect. The band-filling and ASE affected the bandwidth simultaneously, even though their individual contribution could not be determined separately at this point. Below the electron-hole plasma regime, the peak position was observed to be slightly blue-shifted mainly due to the band-filling effect. The observed ASE process suggests that the stimulated emission processes can take place even in the short GaN nanorods, which promises the possible applications for the effective optoelectronic devices based on GaN nanostructures.

## Experimental Section

GaN nanorods were grown on *c*-plane sapphire substrates

using hydride vapor phase epitaxy at a substrate temperature of 500 °C for 20 min. A horizontal reactor was heated by a 5-zone furnace, using $N_2$ as the carrier gas. The flow rates of $NH_3$ and HCl were set to 2 slm and 40 sccm, respectively. The total flow rate and pressure of the reactor (~1 atm) were kept constant during the growth process.

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