

Lasing action in gallium nitride photonic quasicrystal nanorod arrays

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ABSTRACT

We report the observation of lasing action from optically pumped gallium nitride nanorod arrays in a quasicrystal pattern. The nanorods were fabricated from a GaN substrate by nanoimprint patterned etching, followed by epitaxial regrowth to form crystalline facets. The imprint was a 12-fold symmetric quasicrystal pattern. The regrowth grew a multiple quantum well core-shell structure on nanorods. The cathodoluminescent emission of quantum wells red shifts from the bottom to top region of nanorod. Under optical pumping, multiple lasing peaks were observed. The lasing modes formed by 12-fold symmetric photonic quasicrystal nanorod arrays are discussed.

1. INTRODUCTION

There have been great research interests in gallium nitride material due to its promising applications in UV to blue optoelectronic devices and strong emission properties. Conventionally, devices are built in two dimensional thin film structures, where the emission is from planar quantum wells. Recently, devices with one-dimensional nanostructure have gained substantial attention for their interesting properties and potential applications [1-3]. The one-dimensional structure can be fabricated by top-down patterned etching or bottom-up self assembled growth processes. The fabricated nanostructure has shown size confinement effect and interesting light emission properties [4-6]. Stimulated emission from single free standing lying nano wires and 2-D periodic nanorod arrays have been observed [7,8], where lasing modes are the Fabry-Perot modes of wire end surfaces or photonic crystal band edge modes. Recently, stimulated emission from disordered nanorods was also reported [9,10].

2. EXPERIMENT

Figure 1 shows the fabrication process of nanorods. A 0.5 μm SiO_2 thin film was deposited by plasma-enhanced chemical-vapor deposition on a 3 μm GaN substrate grown by metalorganic chemical vapor deposition (MOCVD) on a c-plane sapphire template. The SiO_2 thin film was fabricated into circular disks arranged in a 12-fold symmetric quasicrystal pattern by nano imprint lithography. The SiO_2 disks were used as hard masks in inductively coupled plasma reactive ion etching (ICP-RIE) to etch down exposed GaN and form GaN nanorods. The SiO_2 disks were subsequently removed by buffer oxide etch (BOE). The fabricated GaN nanorods were put back to MOCVD GaN regrowth at 880°C with a growth rate about 2.8 $\text{\AA}/\text{s}$ on the etched nanorod surfaces, the crystalline GaN facets could be controlled under this relatively low growth temperature. The six pairs multiple InGaN/GaN quantum wells were subsequently grown on the crystalline facets, which leads to a core-shell nanorod structure. The growth temperature of GaN quantum barrier layer is around 800°C and higher 100°C than the InGaN quantum well layer, which is the same as on conventional c-plane GaN. A doubled growth time and flow of group III precursors were applied to achieve blue emission on the side wall of core-shell nanorods. Scanning electron microscopy (SEM) is used to observe the surface morphology. The tilt view corresponding schematic figure is shown in the figure 1, and the cross section SEM image of core-shell nanorod is shown in the figure 1(f). The uniform height and geometry of nanorod arrays can be obtained by this top down method.

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It is a potential approach to prepare large scale and uniform InGaN/GaN core-shell structure comparing with the bottom up method by pulse mode or continuous growth mode in MOCVD system [11-12]. The spectra and spatial luminescence properties of core-shell nanorods were analyzed by cathodoluminescence mapping within the SEM chamber. The nanopillar sample was optically excited by a 355 nm tripled Nd:YAG pulse laser at room temperature. The pulse width was 0.5 ns and the pulse repetition rate was 1 KHz. The laser beam was focused on the sample surface in normal incident by a 15X UV microscope objective.

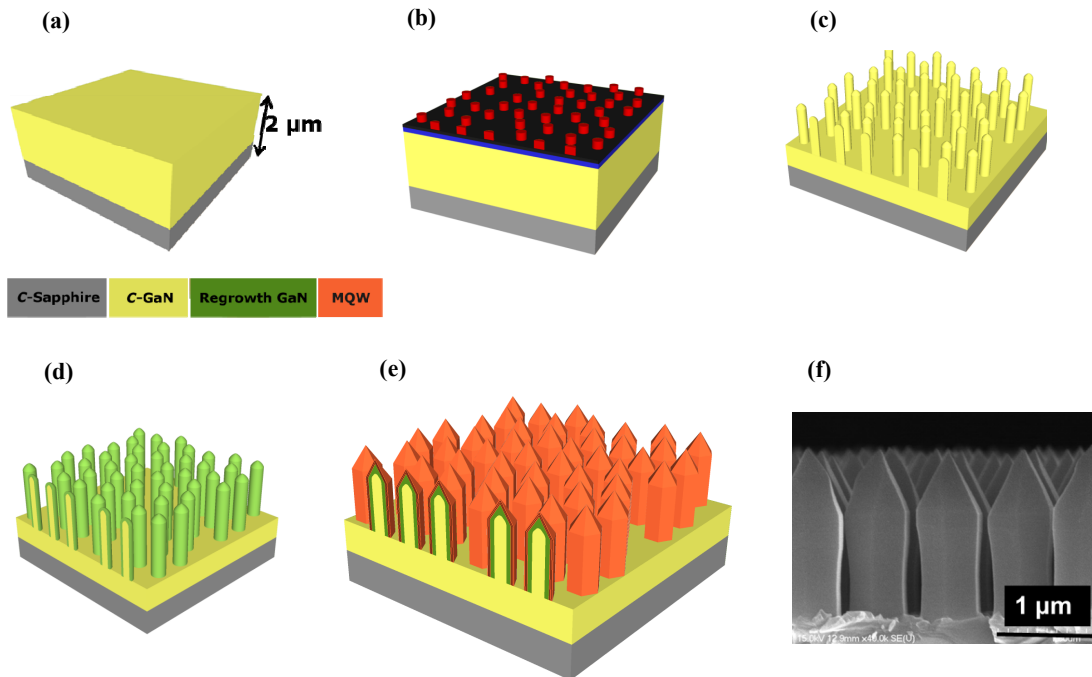


Fig. 1 Fabrication process flow of InGaN/GaN core-shell nanorods from (a) a 3μm thick GaN template growing on c-plane sapphire substrate. (b) after SiO₂ mask layer deposition and nano patterns defined by nano-imprint lithography. (c) after dry etching GaN by reactive ion etcher. (d) after GaN regrowth. (e) after growing 6 pairs InGaN/GaN MQWs on to the GaN nanorod arrays (f) the cross section SEM image of core-shell nanorod arrays.

3. RESULTS AND DISCUSSION

The quasicrystal pattern is a 2-D pattern filled with squares and equilateral triangles whose edges have same length (superlattice constant a), as shown in Fig. 2 (a). The circular disks were placed at vortices. Such a pattern does not have translational symmetry but has long range order and 12-fold rotational symmetry. The plane view SEM image of the fabricated nanorods is shown in Fig. 2 (b), which shows the 12-fold symmetric quasicrystal pattern of nanorod arrays with superlattice constant $a \sim 750$ nm. Despite the original circular nanorods created by ICP-RIE etching, the regrown nanorods have {10-10} M-plane side walls and hexagonal pyramid tips formed by the inclined {10-11} semipolar facets, as shown in the upper right inset image of Fig. 2 (b). The hexagonal side wall length of nanorod is 360 nm.

The photoluminescent (PL) spectra of nanopillar sample at various pump power intensities are shown in the inset of Fig. 3, where the legends are pump intensity levels. As pump intensity increases, narrow emission peaks with line-width in the range of 0.2-0.3 nm emerge from the broad emission background. The integrated power within the spectral range

of these emission peaks versus pump intensity is shown in Fig. 3, which indicates an onset of lasing action at threshold pump intensity of $\sim 5 \text{ MW/cm}^2$. These emission peaks span from 450–470 nm, which corresponds to emission from the quantum wells located from middle to upper part of the nanorod columns by cathodoluminescence images (not shown). The spacing among these lasing peaks do not show obvious regularity. It implies that the lasing modes are unlikely due to whispering gallery modes of individual nanorod. Not all lasing peaks always increase with the increase of pump power. Some of the peaks can decrease due to the increase of other emerging peaks, which indicates that there are lasing mode competitions. Given the rather irregular lasing peaks, what we have observed here is likely a random lasing action.

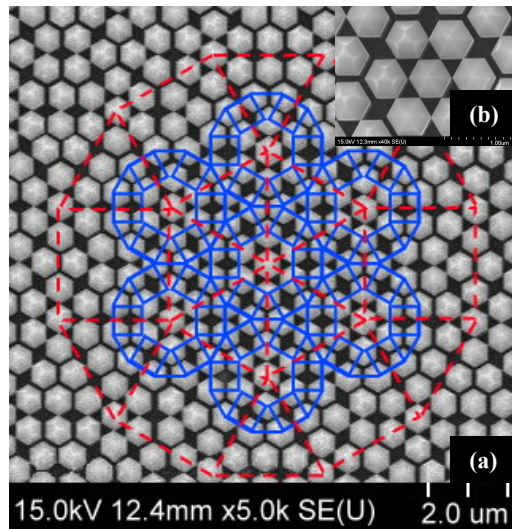


Fig. 2 (a) 12-fold quasicrystal patten. (b) Plane view SEM image of the fabricated nanorod arrays.

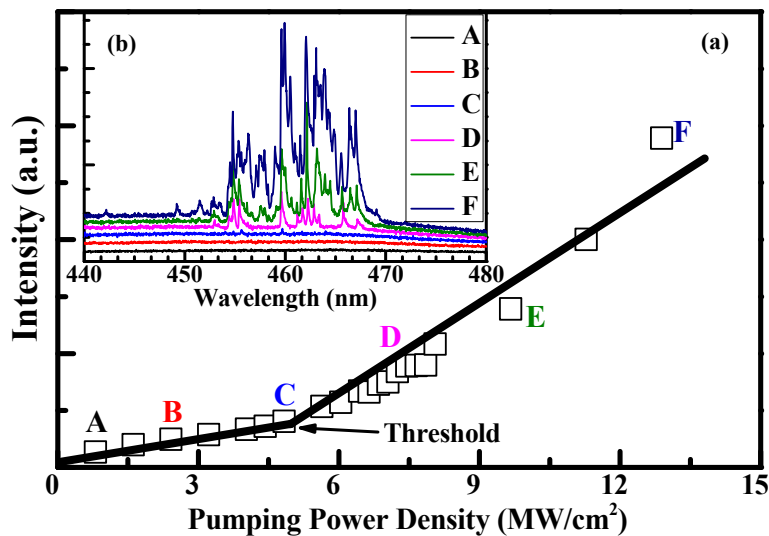


Fig. 3 Integrated emission power versus pump power intensity

Figure 4(a) shows cross section SEM image of core-shell nanorod and the corresponding CL spectrum is shown in fig. 4(b). There are two main emission peaks 450nm and 500nm in the CL spectrum. The emission spectra become red shift from the bottom region to the head of core-shell nanorod as shown in fig. 4(c) to fig. 4(i). The emission wavelength

440nm and 460nm are the dominate signal at the middle portion of the nanorod, which are the lasing wavelength region that we have observed in the fig. 3. There might have some oscillation cavity and path around the nanorod arrays at the middle portion of nanorods. We supposed that may due to the confinement mode of PQC structure which enhances the efficiency of population inversion, and the smooth surfaces at the side wall of nanorod which offer a lower mirror loss oscillation path to achieve lasing action.

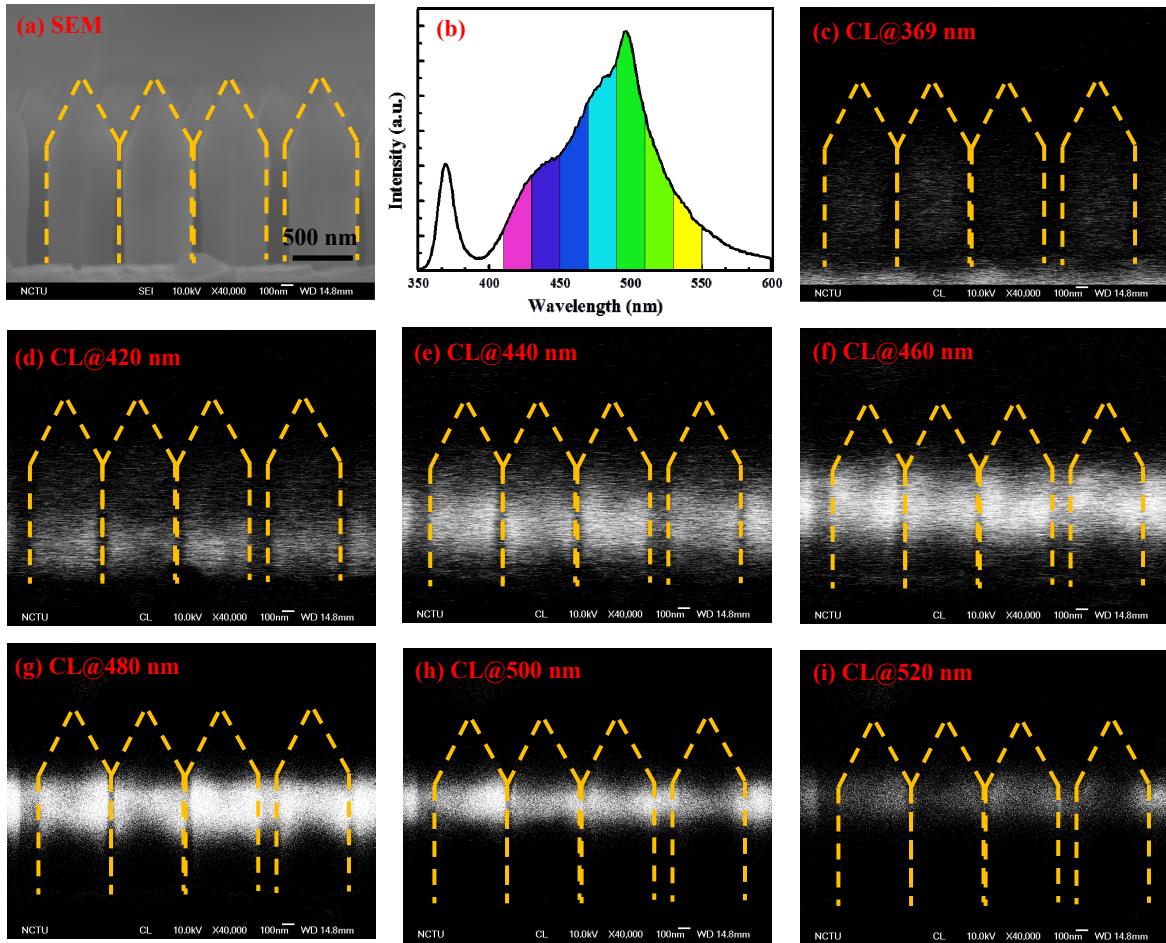


Fig. 4 Spatial luminescence properties of InGaN/GaN core-shell nanorods, (a) cross section SEM image, (b) CL spectrum; specific emission wavelength mapping at (c) 369nm, (d) 420nm, (e) 440nm, (f) 460nm, (g) 480nm, (h) 500nm, (i) 520nm.

A random laser is a lasing action in a disordered active media due the existence of scattering loops where the round trip losses are compensated by gain. The scattering loop serves as the function of optical feedback as the conventional laser cavity. There could also be multiple scattering loops returning to one starting point or overlapping, which can be viewed as randomly distributed feedback. The nanorods in our sample play the roles of scattering sites and providing gain. The lack of short range order of the quasicrystal pattern and the hexagonal nanorod facets together make the sample a pseudo disordered media. One of the random laser characteristic is the lasing threshold pump intensity decreases with increasing pump spot area in a power law relation. The reason in a simplified picture is because there are more multiple recurrent scattering loops that can create tighter confined modes in a larger gain area, which leads to a lower threshold pump intensity. We have measured the threshold pump intensity versus pump spot size. The experimental data points are shown in Fig. 5, along with a power law fitting function. The power law function fits well to the experimental data and gives a threshold pump intensity $I_{th} \sim 1/A^{0.27}$ dependence.

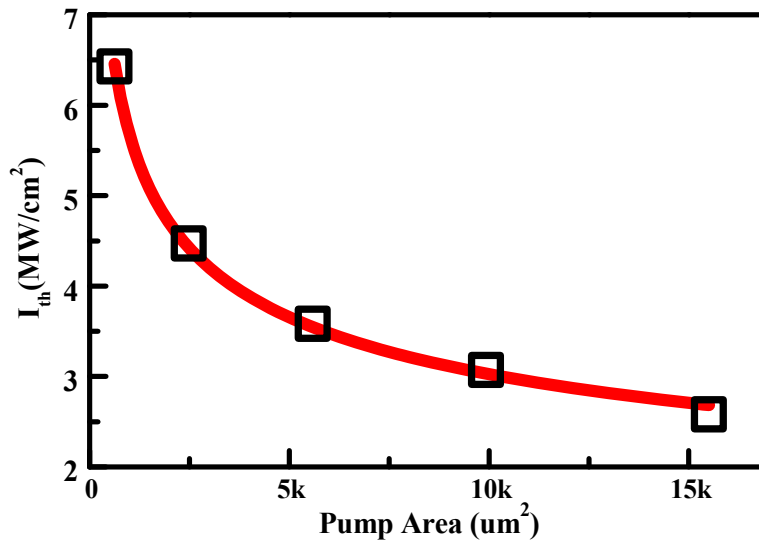


Fig. 5 Threshold pump intensity versus pump spot area and the power law fitting curve showing a threshold pump intensity $P_{th} \sim 1/A^{0.27}$ dependence

4. CONCLUSIONS

We have observed a lasing action in optically pump crystalline nanorod arrays arranged in a 12-fold symmetric quasicrystal pattern. The sample was fabricated from a GaN epitaxial substrate by nano imprint patterned etching and epitaxial regrowth, which formed crystalline facets on nanorods. The regrowth also grew multiple quantum wells on nanorod facets. Under optical pumping, multiple lasing peaks emerged from broad emission background. The irregular multiple lasing peak wavelengths and the inverse dependence of threshold pump intensity on pump spot area shows the characteristics of a random laser.

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