Lasing action in gallium nitride quasicrystal nanorod arrays

Shih-Pang Chang,[1] Kuok-Pan Sou,[1] Chieh-Han Chen,[1] Yuh-Jen Cheng,[1,2,*] Ji-Kai Huang,[3] Chung-Hsiang Lin,[3] Hao-Chung Kuo,[1] Chun-Yen Chang,[4] and Wen-Feng Hsieh[1]

[1]*Department of Photonics, National Chiao Tung University, 1001 Ta Hsueh Rd. Hsinchu 300, Taiwan*
[2]*Research Center for Applied Sciences, Academia Sinica, 128 Sec. 2, Academia Rd, Nankang, Taipei 115, Taiwan*
[3]*Luxtaltek corporation, No. 38, Keyih Road, Chunan, Miaoli County, Taiwan*
[4]*Department of Electronic Engineering, National Chiao Tung University, 1001 Ta Hsueh Rd., Hsinchu 300, Taiwan*
[*]*yjcheng@sinica.edu.tw*

Abstract: We report the observation of lasing action from an optically pumped gallium nitride quasicrystal nanorod arrays. The nanorods were fabricated from a GaN substrate by patterned etching, followed by epitaxial regrowth. The nanorods were arranged in a 12-fold symmetric quasicrystal pattern. The regrowth grew hexagonal crystalline facets and core-shell multiple quantum wells (MQWs) on nanorods. Under optical pumping, multiple lasing peaks resembling random lasing were observed. The lasing was identified to be from the emission of MQWs on the nanorod sidewalls. The resonant spectrum and mode field of the 12-fold symmetric photonic quasicrystal nanorod arrays is discussed.

OCIS codes: (140.3580) Lasers and laser optics; (140.7240) UV, EUV, and X-ray lasers; (160.3380) Laser materials; (160.6000) Semiconductor materials; (290.5890) Scattering.

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1. Introduction

There have been great research interests in gallium nitride material due to its promising applications in UV to blue optoelectronic devices. Conventionally, the devices are built in two-dimensional thin film structure, where emission sources are from the 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. The fabricated nanostructures have shown quantum confinement effect and interesting light emission properties [4–6]. Stimulated emission from single free standing lying nano wire and 2-D periodic nanorod arrays have been observed [7,8], where lasing modes are the Fabry-Perot modes of the wire end surfaces or photonic crystal band edge modes. Recently, lasing action due to the feed back from the multiple scatterings of optically pumped disordered nanorods was also reported [9]. This phenomenon is called random lasing [10], which is often analyzed in terms of statistics.

The statistical nature of random lasing is interesting. However, it puts a limitation to device applications due to the lack of control on the lasing modes and frequency locations from sample to sample. Recently, there are great interests in studying photonic quasicrystals, which may look random at first glance yet have well defined patterns. Their optical properties are fascinating and lie somewhere between those of periodic and random structures [11–14]. It therefore may provide a way to design a pseudo random laser with deterministic properties, which could be important for practical applications. Here we report the observation of lasing action from room temperature optically pumped GaN quasicrystal nanorod arrays. The lasing phenomenon resembles that of a random laser.

The quasicrystal nanorod array sample was fabricated from a GaN epitaxial wafer by nano imprint patterned etch, followed by epitaxial regrowth. The imprint was a 12-fold symmetric quasicrystal pattern [12]. The regrowth formed hexagonal facets on the nanorod sidewalls and hexagonal pyramids on the top. The regrowth also grew InGaN/GaN MQWs on the sidewalls and pyramid facets. The use of MQWs allows us to investigate the optical properties of QPC at a designed wavelength. Under optical pumping, multiple lasing peaks were excited. The lasing was identified to be from the MQWs on the nanorod sidewalls. The peak distribution did not show obvious regularity and behaved like a random lasing action. The linewidth of laser peaks was in the range of 0.2-0.3 nm, indicating a strong resonant feedback oscillation. The lasing threshold pump intensity decreases with increasing pump area. The lasing mechanism is attributed to the feedback of close loop multiple scatterings among nanorods.

2. Fabrication and measurements

The fabrication process of nanorods is as follows. A 0.5 μm SiO_2 thin film was deposited by plasma-enhanced chemical-vapor deposition on a 3 μm GaN substrate grown by metal organic 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 quasicrystal pattern is a two-dimensional pattern filled with squares and equilateral triangles whose edges have the same length (superlattice constant a), as shown in Fig. 1(a). The distance a between every two nearest neighboring points is the same. The circular disks were placed at vortices. Such a pattern does not have translational symmetry but has long-range order and scaling similarity. The SiO_2 disks were used as hard masks in inductively coupled plasma reactive ion etching (ICP-RIE), which etched the exposed GaN and formed GaN nanorods. The height of nanorods was about 1 μm. The SiO_2 disks were subsequently removed by buffer oxide etching. The fabricated GaN nanorods were put back to MOCVD regrowth. Crystalline facets were formed on the etched nanorod

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surfaces. Multiple InGaN/GaN QWs were subsequently grown on the crystalline facets, leading to a core-shell nanorod structure. The plane-view SEM image of the fabricated nanorods is shown in Fig. 1(b), which shows the 12-fold symmetric quasicrystal pattern of nanorod arrays with superlattice constant *a* ~800 nm. The sidewall length of hexagonal nanorod is ~360 nm. The quasicrystal pattern shown in Fig. 1(a) is overlaid on the SEM image to show its quasicrystal pattern. Despite the original cylindrical shape of nanorods created by ICP-RIE, crystalline facets were formed on the regrown nanorods, as shown in Fig. 1(b). The side walls are {10-10} M-planes and the facets of the hexagonal pyramid are {10-11} semi-polar planes, as shown in the upper right inset of Fig. 1(b).

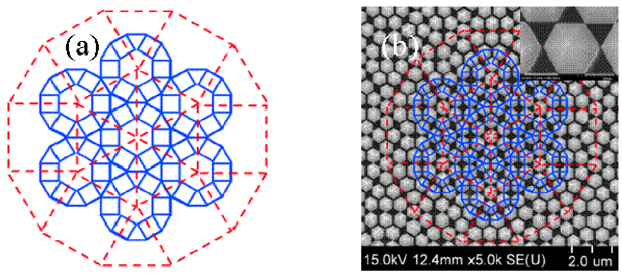

Fig. 1. (a) 12 fold symmetric quasicrystal pattern. (b) SEM plane-view image of the fabricated GaN quasicrystal nanorod arrays. The upper right inset image shows the crystalline facets.

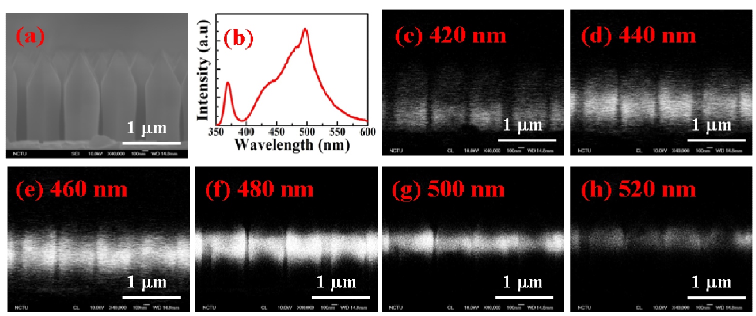

Fig. 2. (a) GaN nanorod SEM side view. (b) Spatially integrated CL spectrum. (c)-(h) Spectrally resolved CL images showing the location dependent emission wavelength.

The SEM cross-section image of nanorods is shown in Fig. 2(a), which shows the pyramid tips and the crystalline sidewall facets. The total nanorod height is about 1.5 um. The SEM scattering electron detection was switched to cathodoluminescent (CL) detection under the same magnification. The spatially integrated CL spectrum of the cross section image is shown in Fig. 2(b). The first peak near 365 nm is the band edge emission of GaN. The large broad spectrum on the right is due to the emission from the InGaN/GaN core-shell MQWs surrounding the nanorod facets. The full width at half maximum linewidth (FWHM) is almost 100 nm. To analyze the emission locations of the broad spectrum, the spectrally resolved CL images were taken at several wavelengths, as shown in Fig. 2(c)-2(h). They show that the emission wavelength red shifts as the scanned location moves from the bottom toward top of nanorods. This red shifted emission is attributed to the increase of In incorporation in MQWs toward the top portion of nanorods. The CL cross-section images also show that the gain is mainly distributed over the wavelength range 460-480 nm, where CL intensity is high.

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 delivered to the sample surface in normal direction by a 15X UV microscope objective. The pump spot at sample surface had a Gaussian intensity profile with $1/e^2$ diameter of 50 µm, verified by a knife-edge measurement. The photoluminescent (PL) spectrum was collected by the same UV objective and coupled into an optical fiber connected

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to the input of a spectrometer (Jobin Yvon IHR320). The PL spectra of nanopillar sample at various pump intensities are shown in Fig. 3(a), where the legends are pump intensities indicated in Fig. 3(b). As pump intensity increases, multiple emission peaks with narrow linewidth in the range of 0.2-0.3 nm emerge from the broad emission background. The integrated intensity within the spectral range of these emission peaks, from 455 nm to 470 nm, versus pump intensity is shown in Fig. 3(b). It indicates an onset of lasing action at threshold pump intensity of ~5 MW/cm^2. This wavelength range corresponds to the emission of the MQWs located from the middle to upper part of the nanorods, as shown in Fig. 2(e)-2(f). The existence of lasing peaks between 455nm and 470nm range is mainly because the gain is high in this wavelength range. The wavelengths of these lasing peaks do not show obvious regularity. The lasing modes are unlikely due to whispering gallery modes of individual nanorod or any regular optical feedback paths. 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 mode competitions among lasing peaks. This could be due to the spatial overlap of lasing modes that are competing for the same gain region. We did not observe any lasing peaks corresponding to the GaN 365 nm transition. The reason is because the pumping energy is mostly absorbed by the MQWs, which are located at the nanorod surfaces.

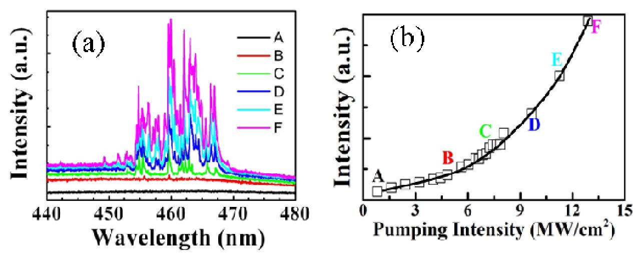

Fig. 3. (a) PL spectra at various pump power densities labeled in graph (b). (b) The integrated intensity versus pump intensity.

Discussion

Given the rather irregular lasing peaks, what we have observed here is likely a random lasing action. A random laser is a lasing action in a disordered active medium due to the existence of multiple scatterings [10,15]. The multiple scatterings can increase the dwell time of light inside the gain medium. When the light scattered inside the gain medium picks up enough gain to compensate the loss of light leaving the pump region, gain saturation occurs and a lasing threshold is reached. In another scenario, the multiple scatterings may bring the light back to a starting point forming a loop, which serves as the optical feedback for a conventional laser cavity. The former one is called incoherent feedback, while the latter one is called coherent feedback. The coherent feedback random lasing often has much narrower lasing linewidth. The nanorods in our sample play the roles of scattering sites and providing gain. The lack of short-range order of the quasicrystal pattern makes the sample a pseudo disordered medium. From the observed rather narrow lasing peaks, the lasing is attributed to the coherent scattering feedback from nanorods. Aside from the irregular lasing peaks, another random laser characteristic is that the lasing threshold pump intensity decreases with increasing pump spot area in a power law relation [10,15,16]. We have measured the threshold pump intensity versus pump spot area A_p. The experimental data points are shown in Fig. 4 along with a red power-law fitting curve. The power law fits well to the experimental data and gives a threshold pump intensity I_{th} ~$1/A_p^{0.27}$ dependence. The reason of this behavior in a simplified picture is because the scattered photons have better chance to accumulate gain in a larger gain area without leaving the pumped region and returning back, which leads to a lower threshold pump intensity. There were some theoretical studies on the power-law dependence [10,17]. The observed coefficient 0.27 is different from the reported

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studies, which might be related to the unique quasicrystal pattern and requires further investigation.

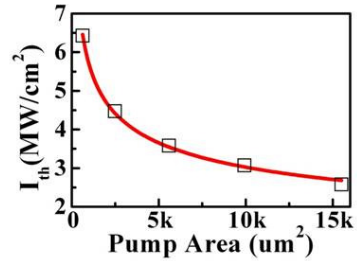

Fig. 4. Threshold pump intensity versus pump area.

We investigated the resonant modes of this pseudo random lasing by performing two-dimensional (2D) finite difference time domain (FDTD) simulation. The 2D model was chosen for simplicity and believed to be a reasonable approximation because the gain was predominantly distributed in 2D direction perpendicular to nanorod axis. The lasing mode field would therefore mainly follow the gain in the 2D direction. The finite nanorod length in the vertical direction will certainly modify the mode frequency and profile, but we consider it as a secondary effect. The simplified model is meant to explain qualitatively the observed lasing behavior. Figure 5(a) shows the quasicrystal model used for simulation. The nanorod and quasicrystal pattern dimensions measured from SEM image were used to construct the model. The over all pattern size is about 16 μm across. The finite pattern size was used due to the limit of computation power and time. An optical pulse with a center wavelength at 470 nm and a linewidth of 30 nm was launched at a randomly chosen location near nanorod sidewall to simulate the emission from MQWs. The pulse spectral width was chosen according to the observed lasing spectral range. Both transverse electric and magnetic field were calculated. After a long enough propagation time, the shape of spectrum became steady. The spectrum contained distinct resonant peaks, which were the resonant modes excited by the broad-band pulse. The same calculation was repeated for different pulse launching locations. The quality factors of these modes are in the range of 500. The spectrum varied from location to location and had many or just a few resonant peaks. All these resonant peaks can be regarded as the potential lasing modes. For illustration purpose, three spectra were shown in Fig. 5(b)-5(d). The corresponding launching locations are the points labeled (b), (c), and (d) in Fig. 5(a). Points (b) and (c) are deliberately at locations away from center. Point (d) is at the center rod.

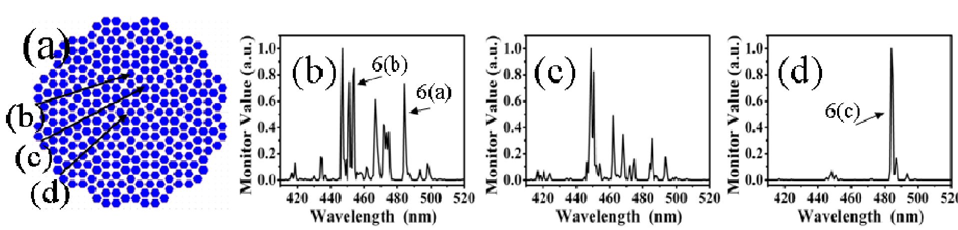

Fig. 5. (a) The quasicrystal model used in FDTD simulation. (b)-(d) The resonant spectra obtained by launching broad band pulses at locations labeled correspondingly in Fig. 5(a)

The calculated resonant spectra show irregular peak locations, similar to what was observed in the experiment. The calculated resonant peaks do not match to the observed spectrum in peak to peak locations. This can be attributed to the fact that the model used in simulation is an overly simplified approximation. The lasing peaks should be ideally deterministic and can be engineered by design since the QPC pattern is deterministic. This could be important in practical applications.

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We then calculated the resonant mode profiles. It was calculated by choosing one of the peaks in the resonant spectrum and launching it at the same location. After propagating for a reasonable long time, a distinctive pattern was developed. We regarded it as the mode profile of the specific peak. Ideally, one can repeat the calculation for all the peaks to obtain all the mode profiles. We have calculated a few of them. The mode profiles could be rather irregular or have certain distinctive feature. For illustration purpose, the mode profiles of the resonant peaks labeled 6(a) and 6(b) in Fig. 5(b), and 6(c) in Fig. 5(c) are shown in Fig. 6(a)-6(c), respectively. The mode field in Fig. 6(a) shows that certain part of nanorods can couple together to provide guiding effect and form various propagation loops. Figure 6(b) on the other hand shows a more scattering like coupling pattern. Figure 6(c) is the field pattern for a source launched at the center rod. The mode field propagates outward and forms triangular shape resonant loops. These patterns show that the intricate property of quasicrystal structure has varieties of mode field patterns, which require a further investigation.

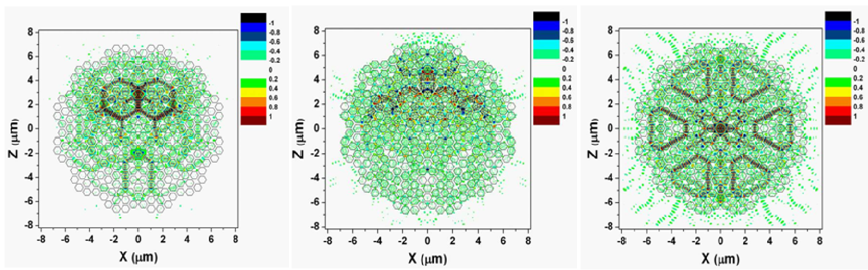

Fig. 6. (a)-(c) Mode field patterns correspond to the resonant peaks labeled as 6(a), 6(b), and 6(c) in Fig. 5.

Summary

We have observed a lasing action in an optically pumped 12-fold symmetric quasicrystal nanorod arrays. The sample was fabricated from a GaN epitaxial substrate by nano patterned etching and epitaxial regrowth. The regrowth grew core-shell MQWs and crystalline facets on nanorods. Under optical pumping, multiple lasing peaks emerged from broad emission background. The irregular multiple lasing wavelengths and the inverse dependence of threshold pump intensity on pump spot area resembles the characteristics of random lasing. The irregularity of resonant peaks is qualitatively explained by a simplified FDTD simulation.

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