Lasing at exciton transition in optically pumped gallium nitride nanopillars

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Abstract: We report the observation of room temperature lasing action in optically pumped GaN nanopillars. The nanopillars were fabricated by patterned etching and crystalline regrowth from a GaN substrate. When nanopillars were optically excited, a narrow emission peak emerged from the broad spontaneous emission background. The increasing rate is nine times faster than that of the spontaneous emission background, showing the onset of lasing action. The lasing occurs right at the center of spontaneous emission rather than the often reported redshifted wavelength. A spectroscopic ellipsometry analysis indicates that the gain of lasing action is provided by exciton transition.

OCIS codes: (160.0160) Materials; (160.3380) Laser materials; (160.6000) Semiconductor materials; (140.5960) Semiconductor lasers; (140.7240) UV, EUV, and X-ray lasers.

References and links

1. T. Kuykendall, P. Pauzauskie, S. K. Lee, Y. F. Zhang, J. Goldberger, and P. Yang, "Metalorganic chemical vapor deposition route to GaN nanowires with triangular cross sections," Nano Lett. **3**(8), 1063–1066 (2003).
2. F. Qian, S. Gradečak, Y. Li, C.-Y. Wen, and C. M. Lieber, "Core/multishell nanowire heterostructures as multicolor, high-efficiency light-emitting diodes," Nano Lett. **5**(11), 2287–2291 (2005).
3. K. Kishino, H. Sekiguchi, and A. Kikuchi, "Improved Ti-mask selective-area growth (SAG) by rf-plasmaassisted molecular beam epitaxy demonstrating extremely uniform GaN nanocolumn arrays," J. Cryst. Growth **311**(7), 2063–2068 (2009).
4. S. D. Hersee, X. Sun, and X. Wang, "The controlled growth of GaN nanowires," Nano Lett. **6**(8), 1808–1811 (2006).
5. N. Thillosen, K. Sebald, H. Hardtdegen, R. Meijers, R. Calarco, S. Montanari, N. Kaluza, J. Gutowski, and H. Lüth, "The state of strain in single GaN nanocolumns as derived from micro-photoluminescence measurements," Nano Lett. **6**(4), 704–708 (2006).
6. E. Calleja, M. Sánchez-García, F. J. Sánchez, F. Calle, F. B. Naranjo, E. Muñoz, U. Jahn, and K. Ploog, "Luminescence properties and defects in GaN nanocolumns grown by molecular beam epitaxy," Phys. Rev. B **62**(24), 16826–16834 (2000).
7. T. Kouno, K. Kishino, K. Yamano, and A. Kikuchi, "Two-dimensional light confinement in periodic InGaN/GaN nanocolumn arrays and optically pumped blue stimulated emission," Opt. Express **17**(22), 20440–20447 (2009).
8. J. C. Johnson, H.-J. Choi, K. P. Knutsen, R. D. Schaller, P. D. Yang, and R. J. Saykally, "Single gallium nitride nanowire lasers," Nat. Mater. **1**(2), 106–110 (2002).
9. H.-J. Choi, J. C. Johnson, R. He, S.-K. Lee, F. Kim, P. Pauzauskie, J. Goldberger, R. J. Saykally, and P. Yang, "Self-Organized GaN Quantum Wire UV Lasers," J. Phys. Chem. B **107**(34), 8721–8725 (2003).
10. S. Gradečak, F. Qian, Y. Li, H.-G. Park, and C. M. Lieber, "GaN nanowire lasers with low lasing thresholds," Appl. Phys. Lett. **87**(17), 173111 (2005).
11. S. Shokhovets, K. Köhler, O. Ambacher, and G. Gobsch, "Observation of Fermi-edge excitons and exciton-phonon complexes in the optical response of heavily doped n-type wurtzite GaN," Phys. Rev. B **79**(4), 045201 (2009).
12. A. J. Fischer, W. Shan, J. J. Song, Y. C. Chang, R. Horning, and B. Goldenberg, "Temperature-dependent absorption measurements of excitons in GaN epilayers," Appl. Phys. Lett. **71**(14), 1981–1983 (1997).
13. J. F. Muth, J. H. Lee, I. K. Smagin, R. M. Kolbas, H. C. Casey, B. P. Keller, U. K. Mishra, and S. P. DenBaars, "Absorption coefficient, energy gap, exciton binding energy and recombination lifetime of GaN obtained from transmission measurements," Appl. Phys. Lett. **71**(18), 2572–2574 (1997).
14. G. Yu, G. Wang, H. Ishikawa, M. Umeno, T. Soga, T. Egawa, J. Watanabe, and T. Jimbo, "Optical properties of wurtzite structure GaN on sapphire around fundamental absorption edge (0.78–4.77 eV) by spectroscopic ellipsometry and the optical transmission method," Appl. Phys. Lett. **70**(24), 3209–3211 (1997).

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15. M. Tchounkeu, O. Briot, B. Gil, J. P. Alexis, and R.-L. Aulombard, "Optical properties of GaN epilayers on sapphire," J. Appl. Phys. **80**(9), 5352–5360 (1996).
16. O. Gluschenkov, J. M. Myoung, K. H. Shim, K. Kimb, Z. G. Figen, J. Gao, and J. G. Eden, "Stimulated emission at 300 K from photopumped GaN grown by plasma-assisted molecular beam epitaxy with an inductively coupled plasma source," Appl. Phys. Lett. **70**(7), 811–813 (1997).
17. F. Binet, J. Y. Duboz, J. Off, and F. Scholz, "High-excitation photoluminescence in GaN: hot-carrier effects and the Mott transition," Phys. Rev. B **60**(7), 4715–4722 (1999).
18. S. Bidnyk, T. J. Schmidt, B. D. Little, and J. J. Song, "Near-threshold gain mechanisms in GaN thin films in the temperature range of 20–700 K," Appl. Phys. Lett. **74**(1), 1–3 (1999).
19. K. Kazlauskas, G. Tamulaitis, A. Žukauskas, T. Suski, P. Perlin, M. Leszczynski, P. Prystawko, and I. Grzegory, "Stimulated emission due to spatially separated electron-hole plasma and exciton system in homoepitaxial GaN," Phys. Rev. B **69**(24), 245316 (2004).
20. X. Zhang, P. Kung, A. Saxler, D. Walker, and M. Razeghi, "Observation of room temperature surface-emitting stimulated emission from GaN:Ge by optical pumping," J. Appl. Phys. **80**(11), 6544–6546 (1996).
21. Y. C. Chang, Y.-L. Li, D. B. Thomson, and R. F. Davis, "Phonon-assisted stimulated emission from pendeoepitaxy GaN stripes grown on 6H-SiC substrates," Appl. Phys. Lett. **91**(5), 051119 (2007).
22. W. van Roosbroeck and W. Shockley, "Photon-radiative recombination of electrons and holes in Germanium," Phys. Rev. **94**(6), 1558–1560 (1954).
23. C. Vanneste, P. Sebbah, and H. Cao, "Lasing with resonant feedback in weakly scattering random systems," Phys. Rev. Lett. **98**(14), 143902 (2007).
24. M. Sakai, Y. Inose, K. Ema, T. Ohtsuki, H. Sekiguchi, A. Kikuchi, and K. Kishino, "Random laser action in GaN nanocolumns," Appl. Phys. Lett. **97**(15), 151109 (2010).
25. F. Binet, J. Y. Duboz, E. Rosencher, F. Scholz, and V. Harle, "Mechanisms of recombination in GaN photodetectors," Appl. Phys. Lett. **69**(9), 1202–1204 (1996).
26. J. S. Im, A. Moritz, F. Steuber, V. Harle, F. Scholz, and A. Hangleiter, "Radiative carrier lifetime, momentum matrix element, and hole effective mass in GaN," Appl. Phys. Lett. **70**(5), 631 (1997).
27. J. B. Schlager, N. A. Sanford, K. A. Bertness, and A. Roshko, "Injection-level-dependent internal quantum efficiency and lasing in low-defect GaN nanowires," J. Appl. Phys. **109**(4), 044312 (2011).

1. Introduction

GaN is considered as a very promising material for UV and blue optoelectronic devices due to its wide direct bandgap transition and strong emission properties. GaN one-dimensional (1-D) nano structure devices have attracted great research interests for their novel properties and potential applications [1,2]. These nano devices, known as nanopillars or nanowires, can be grown by selective area or self-assembled growth using rf-plasma-assisted molecular beam epitaxy (rf-MBE) [3] or metal organic chemical vapor deposition (MOCVD) [4]. The selective area growth can produce arrays of nanopillars in designed order while self-assembled growth often produces nanowires in random orientation. The fabricated nanowires or nanopillars have shown improved material quality and size confinement effect which can significantly enhance light emitting properties [5–7].

The optical properties of GaN nanowires are usually investigated by photoluminescent (PL) measurement. The reported room temperature stimulated emission peaks are often at wavelengths redshifted from the spontaneous emission peak [8–10] and longer than the reported nominal GaN exciton transition [11–15]. The redshift of stimulated emission peak has also been observed in intensively pumped GaN thin film material [16–21]. The redshift is due to several different mechanisms. At carrier density above Mott transition, the redshift is attributed to the bandgap shrinkage due to many body interaction [18], where excitons are disassociated into electron-hole plasma [10,17]. Other mechanisms involve exciton-exciton or exciton-phonon scattering, which occur mostly at cryogenic temperature. A room temperature stimulated emission without redshift has rarely been reported.

Here we report the observation of lasing action from optically pumped GaN nanopillars, where the lasing wavelength occurs right at the spontaneous emission peak. The nanopillars were fabricated by patterned etching and crystalline regrowth from a GaN epitaxy wafer. When the sample was pumped by a focused 355 nm pulse laser, a narrow lasing peak emerged from broad spontaneous emission background. A spectroscopic ellipsometry analysis suggests that the gain of lasing action is provided by exciton transition. The lasing wavelength maintains at exciton transition throughout the whole pump power range with a slightly blue shift with increasing pump power, which is attributed to exciton band filling.

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2. Fabrication of GaN nanopillars

The schematic of GaN nanopillar fabrication process is shown in Figs. 1 (a)-(d). A 3 μm GaN thin film was grown on a c-plane sapphire (0001) substrate by low pressure metalorganic chemical vapor deposition (MOCVD). A 300 nm Si_3N_4 thin film was subsequently deposited on the GaN thin film by plasma-enhanced chemical-vapor deposition, followed by the electron-beam evaporation of a 100 nm Ni thin film, as illustrated in Fig. 1 (a). The sample was then subjected to rapid thermal annealing (RTA) at 850 °C under nitrogen ambiance for 1 min. to form self-assembled Ni nanomasks, as shown in Fig. 1(b). A reactive ion etching was conducted to etch Si_3N_4 film using a CF_4/O_2 gas-mixture to transfer Ni nanomask pattern down to Si_3N_4 layer. The sample was subsequently etched down to GaN by inductively coupled plasma reactive ion etching system (SAMCO ICP-RIE 101iPH) operated at 13.5 MHz under a gas mixture of Cl_2/Ar = 50/20 sccm for 2 min. to form nanopillars as shown in Fig. 1(c). The sample was dipped into a nitric acid solution (HNO_3) at 100 °C for 5 min. to remove the Ni nano masks. Finally, the etched GaN sample was put back to MOCVD to grow additional GaN on the etched GaN nanopillars, as illustrated in Fig. 1(d). The Si_3N_4 mask was intentionally left on top of nanopillars to prevent the regrowth in c-axis direction. This regrowth process grew crystalline GaN on the etched pillar side walls. It reduces surface defects created during the ICP-RIE pillar etching process.

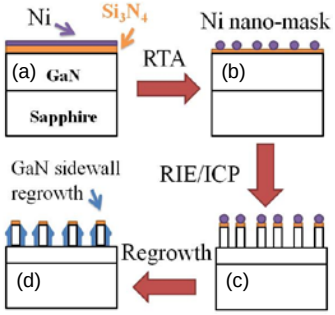

Fig. 1. (a)-(d) Nanopillar fabricating process flow: (a) Si_3N_4/Ni deposition. (b) Rapid thermal annealing (RTA) to form Ni nanomasks. (c) Ni nanomask pattern transferred to Si_3N_4 and GaN by dry etching. (d) Crystalline regrowth to from hexagonal pillars.

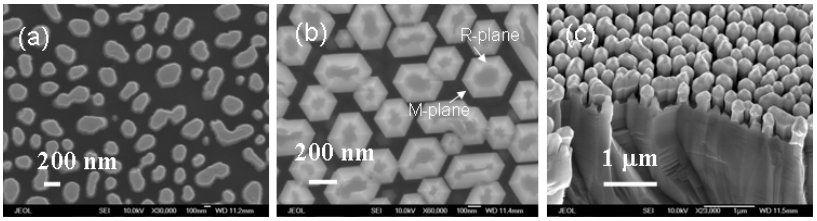

Fig. 2. (a) Top view of the etched nanopillars before regrowth. (b) Top view of nanopillars after regrowth, showing the m-plane {10-10} hexagonal side walls and the top inclined r-plane {1-102} facets. (c) Side view of nanopillars after regrowth.

The SEM images of the fabricated nanopillars are shown in Fig. 2. Figure 2 (a) is the top view of the etched nanopillar sample before regrowth. The irregular shape was originated from the self-assembled Ni nanomasks. Figures 2 (b)-(c) are the top and side views of the nanopillars after regrowth. The regrowth grows hexagonal m-plane {10-10} crystalline sidewall facets. The irregular hexagonal shape is due to the irregular etched nanopillar pattern as shown in Fig. 2 (a). The regrowth also grows inclined r-plane facets {10-12} close to the top of nanopillars with a very slight overgrowth on Si_3N_4 mask. The height of nano-pillars is about 660 nm. The averaged diameter is about 250 nm.

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3. Observation of lasing action and discussion

The optical property of the fabricated GaN nanopillars was probed by time-averaged PL measurement using tripled Nd:YAG pulse laser at 355 nm. The pulse width is 0.5 ns and the pulse repetition rate is 1 KHz. The laser beam was focused on sample surface in normal incidence by a 15X UV microscope objective. The focus spot had a Gaussian beam waist w_0 of 1.8 μm, which is verified by knife edge measurement. The PL signal was collected through the same 15X objective and analyzed by a spectrometer (Jobin Yvon IHR320). The PL spectra are shown in Fig. 3 (a)-(b). The legends in graphs are pump intensity levels in unit of megawatt per centimeter square. The pump power intensity is obtained by dividing the power meter reading measured at sample location by the pulse duty cycle (0.5 ns / 1 ms) and by the pump spot area (πw_0). At low pump intensity, the spontaneous emission has a maximum around 363 nm. This wavelength is in line with the nominally reported exciton transition [11–15], which is believed to be also the case for our sample and will be discussed shortly. The full width at half maximum (FWHM) linewidth is 12 nm, which is about twice of GaN substrate value. The larger linewidth may be due to residual surface defects from fabrication process. As pump intensity increases, a small peak emerges from the center of spontaneous emission, as shown in Fig. 3 (a). It is interesting to see its dramatic increase on top of the broad spontaneous emission background when pump intensity increases, as shown in Fig. 3 (b). This narrow peak has a much faster increasing rate than the spontaneous emission background does, indicating an onset of stimulated emission. The inset in Fig. 3 (b) is a zoom in of the laser emission spectra. It shows a slight blueshift as pump intensity increases.

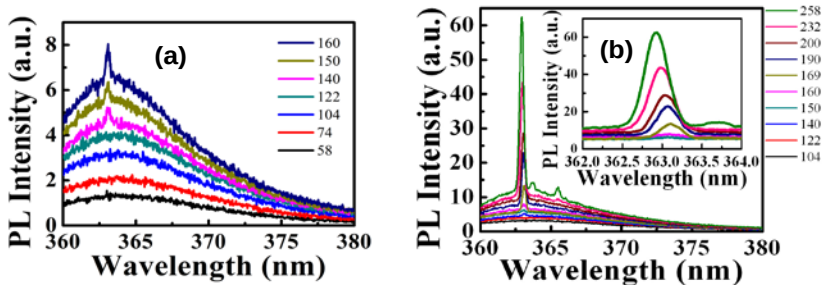

Fig. 3. (a) Spontaneous PL spectra at various pump intensities below and near threshold. (b) PL spectra at various pump intensity levels showing the onset of 363 nm excitonic lasing action. Legend: pump intensity (MW/cm^2).

The peak intensities of the emerging signal and the spontaneous emission versus pump intensity are show in Fig. 4 (a). The spontaneous emission intensity increases linearly with pump intensity (black line). The 363 nm emission peak increases superlinearly around threshold and has slope efficiency 9 times faster than that of spontaneous emission above threshold. This behavior evidently demonstrates the onset of lasing action. The linewidth versus pump intensity for the emerging signal is also shown in Fig. 4 (a). The linewidth significantly reduces across the threshold down to 0.3 nm. The stimulated emission wavelength versus pump intensity is shown in Fig. 4 (b). The wavelength slightly blue shifts from 363.1 nm to 362.9 nm when pump intensity increases from threshold to 1.7 times above threshold. We remark that this slight blueshift of emission wavelength is due to exciton band filling. This is very different from the often reported stimulated emission in GaN nanowires [8,10], where the emission is at longer wavelength and redshifts with increasing pump intensity. The redshift was suggested to be due to electron-hole plasma at high carrier density due to bandgap renormalization. In our sample, the stimulated emission occurs right at the spontaneous emission peak and maintains its position without redshift throughout the pump power range. We have scanned the focused pump spot on the sample and the emission characteristic is fairly consistent except at some locations where the focused spots were damaged before reaching threshold, which might be due to defective nanopillars.

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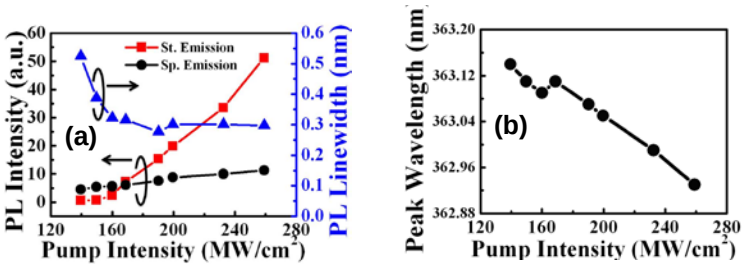

Fig. 4. (a) The 363 nm stimulated (st.) emission intensity, linewidth, and spontaneous (sp.) emission intensity versus pump intensity. (b) The blueshift of stimulated emission wavelength with increasing pump intensity.

To investigate the origin of the 363 nm (3.416 eV) emission in our sample, spectroscopic ellipsometry (J.A. Woollam VASE) was used to analyze the dielectric function of the GaN substrate used to fabricate nanopillars. The measured imaginary part of dielectric function is shown in Fig. 5(a). There is an absorption peak at 3.44 eV near the transition band edge. This spectral characteristic is similar to previous reports [11–14], which identifies this absorption peak as a signature of exciton transition. The measured 3.44 eV value is in good agreement with the reported values ranging from 3.43 eV to 3.45 eV [11–14]. The variations could be due to residual strain from different growth processes. However, the measured spectra are all consistent in having the characteristic exciton absorption peak near the transition edge.

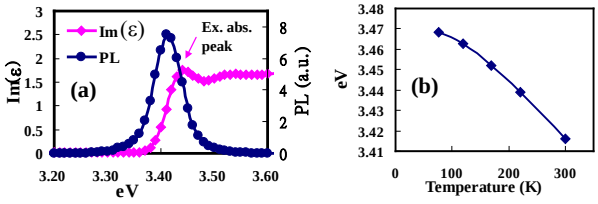

Fig. 5. (a) Imaginary part of dielectric function from ellipsometer measurement and the spontaneous PL emission derived from the measured absorption spectrum using the detailed balance principle. (b) Spontaneous emission peak versus temperature of nanopillars.

By applying the detailed balance principle [22], one can derive the spontaneous emission spectrum $I(\nu)$ from the measured absorption spectrum by

$$I(\nu) = 8\pi n^2(\nu)\alpha(\nu) / \{h^3c^2[\exp(h\nu / kT) - 1]\},$$

where n is the refractive index, α is the absorption constant, h is the Plank constant, k is the Boltzmann constant, T is the temperature, and c is the speed of light. Such an analysis has been reported and shown great agreement between the emission peak derived from absorption spectrum and the measured PL peak [13], which is thus identified as exciton emission. Following the same approach, the derived emission spectrum based on the measured $\alpha(\nu)$ and $n(\nu)$ from spectroscopic ellipsometry measurement is plotted in Fig. 5(a), which has a peak at 3.413 eV (363.3 nm). This number is in close agreement to the measured PL peak at 363.1 nm. From the analysis, it therefore suggests that the origin of 363 nm peak is an excitonic emission. The observed sub-nm blue shift is considered to be due to the band filling of exciton band. In the reported electron-hole plasma observation, the band filling effect was never reported to be significant compared with the redshift from bandgap renormalization effect, which is often in a few nm range. We do not observe redshift in our sample. The emission is thus unlikely due to electron-hole plasma transition. For comparison, the emission peak of nanopillar sample versus temperature was measured and shown in Fig. 5(b). The temperature dependent curve is similar to the reported data [12].

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The lasing linewidth is only about 0.3 nm. The nanopillars in our experiment are short columns about 660 nm in length standing on a GaN substrate. It is unlikely to have lasing modes oscillating in the pillar axis direction since there is only one reflection end surface. For comparison, a sample from the GaN substrate used to fabricate nanopillars was pumped under the same pumping set up. There was no lasing action observed before the sample was damaged. From a simplified view point, the nanopillars are densely populated random scattering sites, which increase photon sojourn time in the pumped region. As a result, photons have more chance to interact with gain due to multiple scattering. When pumping is high enough, lasing action may occur [23]. This may explain why it was hard to reach lasing in GaN substrate, where no scattering feedback is available. From the plane view SEM image, the optical scattering mean free path is longer than wavelength. What we have observed is probably a lasing with weak scattering feedback, where the lasing mode is a quasimode extended over the pump spot region. This is different from a recent report showing multiple lasing peaks from a denser GaN nanocolumns, which is in the strong scattering feedback regime and have multiple lasing peaks [24]. There are small peaks appearing at the highest pump power in Fig. 3(b). It could be due to some scattering feedback paths that reach threshold at much higher pump density.

The measured peak threshold pump power intensity of 140 MW/cm^2 is fairly high. If we assume a hundred percent pump power absorption and consider the absorption coefficient of $1.0\text{x}10^5$ cm^{-1} and radiative recombination coefficient of $1.3\text{x}10^{-8}$ cm^3 s^{-1} from reported studies [13,25], it would give free carrier density of $4\text{x}10^{19}$ cm^{-3}. If we use exciton luminescent decay time of 40 ps [26], the estimated carrier density is $1\text{x}10^{21}$ cm^{-3}. These numbers are significantly higher than the nominally reported Mott density of $2\text{x}10^{18}$ to ~$1\text{x}10^{19}$ cm^{-3} [11,17]. These estimates are upper bound estimates. Other loss mechanisms for example nonradiative recombination, carrier diffusion, and pump scattering loss may lower the carrier density number. On the other hand, due to the temporal and spatial variations of pump pulse, exciton and electron-hole plasma may co-exist during a pump pulse excitation. There have been reports indicating that excitonic effect can still play a substantial role above Mott density [11,27]. The dynamics of excitonic emission under such a coexisting condition is an interesting problem for further study.

4. Summary

We report a room temperature lasing at exciton transition wavelength from optically pumped GaN nanopillars. The nanopillars were fabricated by self-assembled Ni nanomasked etching of an epitaxial GaN substrate, followed by crystalline regrowth to form hexagonal crystalline facets. When the sample was excited by a 355 nm pulse laser with 1.8 μm focused spot size, a lasing action was observed at GaN exciton transition wavelength. The lasing wavelength persists at exciton wavelength throughout the observed pump power range. This is very different from the often reported redshifted electron-hole plasma lasing.

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