Random Laser Action in GaN nanocolumns

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Abstract

We report observations of random laser action in self-organized GaN nanocolumns. We have measured three samples with different filling fractions and investigated the dependence of the lasing property on the random configuration of nanocolumns. Numerical calculations based on a finite-difference time-domain method have also been performed and the comparison with the experimental results shows a clear relationship between the strength of light localization and the occurrence of random laser action.

In a random disordered system, a suitable combination of multiple light scattering and optical interference leads to the localization of light. Furthermore, localized light may form closed loop paths, and if the system has an optical gain, a lasing action may build up at each loop path. This phenomenon is called random lasing [1]. The first experimental observation of random lasing employed laser dye solutions with scattering particles [2]. Later observations of random lasers have been reported in semiconductors, such as ZnO powder [3,4], ZnO nanorods [5], GaAsN [6], GaAs [7], SnO_2 [8] and ZnSe [9]. In recent years, electrically pumped random laser action was reported in ZnO [10] and various applications using random lasers were proposed [11]. The physics of random lasers has been discussed both theoretically and experimentally in terms of photon statistics [12], chaotic behavior [13], mode structures [14], and in the context of prelocalized modes [15]. However the occurrence of random lasing due to a random structure has not been fully analyzed and explained. In this paper, we report observations of random laser action in self-organized GaN nanocolumns [16]. Using three samples with different filling fractions of nanocolumns, we have investigated the dependence of the lasing property on the random structure. This is also the first report of random laser action in GaN. We have performed numerical calculations based on a two-dimensional (2D) finite-difference time-domain (FDTD) method [17], and show the relationship between the strength of light localization and the occurrence of random lasing.

The samples are self-organized GaN nanocolumns fabricated on a Si substrate by an rf-plasma assisted molecular beam epitaxy (rf-MBE) technique. Figure 1 shows bird's-eye and top views of scanning electron microscope (SEM) images of the samples. The configuration characteristics of all samples are also summarized in Fig. 1. The photo-excited emission spectra from the samples were measured at room temperature using the experimental setup shown schematically in Fig. 2. We used the frequency-tripled output (355 nm) of a Nd:YAG pulse laser as a pump pulse. The pulse width and the repetition rate are 5 ns and 20 Hz, respectively. The pump pulse was focused onto the sample using a cylindrical lens to achieve a pump stripe of 10-mm-length and 120-μm-width. We observed emission spectra of the output light radiated from the side facet, perpendicular to the *c*-axis [0001] of the

GaN-nanocolumns. An objective (40 ×) and a spatial filter were used to collect the emission with a spatial resolution of approximately 1.3 μm. The polarization behavior of the output was also observed by inserting a Glan laser polarizer before the spectrometer; the definitions of TE and TM polarization are shown in Fig. 2.

Figures 3 (a), (b) and (c) show the single-shot emission spectra for different pump intensities of Sample-1, Sample-2 and Sample-3, respectively. At low excitation intensity all samples show the same spectra consisting of a broad spontaneous emission from the bandedge (or excitons) of GaN. Since the spontaneous emission intensities of the three samples were approximately proportional to the filling fraction of nanocolumns, the emission intensity from each single nanocolumn is almost the same for all samples. This result proves that there is no significant difference in crystal quality among the three samples, which means that we can unambiguously study the effects due to the random configurations of nanocolumns.

As shown in Fig. 3(a), Sample-1 shows spontaneous emission spectra at all excitation densities (180–800 kW/cm^2). The integrated intensity of the spontaneous emission was linear in the excitation density. On the other hand, Sample-2 and Sample-3 show drastic changes in their spectra at higher excitation densities.

In Sample-2, when the pump intensity exceeded a threshold value (320 kW/cm^2), impressive narrow emission peaks emerged from the broad spontaneous emission. As the pump intensity was increased, multiple strong emission peaks were detected at wavelengths between 362 nm and 370 nm. The width of the narrowest peak spectrum was less than the spectral resolution of ~ 0.1 nm. These multiple lasing modes are attributed to random lasing with coherent feedback. The single-shot spectrum shape, such as the positions of lasing peaks, changed randomly from one pump pulse to another. This behavior is known as a typical characteristic of random lasing [13].

In Sample-3, an increase of the pump intensity (350–700 kW/cm^2) produced a clear peak, rising in intensity, whose center wavelength shifted to the red. This behavior is often observed for the emission from an electron-hole plasma (EHP) under high excitation conditions. The red shift is attributed to the

bandgap renormalization effect.

It should be noted that the position of the EHP peak and the random lasing peaks are quite different. The EHP peak occurs at the low energy side of the spontaneous emission, which is normal behavior at high excitation. On the other hand, the random lasing peaks occur on the high energy side of the spontaneous emission, in contrast to the characteristics of ordinary lasers. Although the forms of the random lasing spectra change shot by shot, most of the lasing peaks are on the high energy side. This strange phenomenon may be a unique and an important property of random lasers. Further study is necessary to clarify this behavior.

The inset in Fig. 3(b) shows the polarization dependence of random lasing emission in Sample-2 at an excitation density of 770 kW/cm^2. The filled squares shows the integrated intensity of the random lasing emission. This result shows that the random lasing in GaN nanocolumns is mainly TE-polarized. According to calculations for 2D random system, the light localization effect should be stronger for the TM mode than for the TE mode [18]. In the case of GaN, however, the TE-polarized emission becomes dominant because of the polarization selection rule due to the symmetry of the wave functions of its band structures.

In order to elucidate the dependence of the random lasing on the sample structure, numerical calculations based on the FDTD method have been performed. Since the height of the GaN nanocolumns is much larger than both their diameter and the luminescence wavelength of GaN, we performed 2D calculations to simulate the light localization property. Taking account of the experimental results of the polarization behavior, the TE mode ($E \perp z$) was chosen for the calculation. The configurations of the nanocolumns used in the calculations were directly translated from the SEM images of the samples. The upper panel of Fig. 4(a) shows SEM images with 5-nm-pixel pitch, and the lower panel of Fig. 4(a) shows the sample models obtained in this way. Black areas denote the nanocolumns and white areas denote air, and the dielectric constants were set to 7.29 and 1.00, respectively. The FDTD cell size is 5 nm for both x and y axes. For the boundary condition, we have adopted the perfect matched layer (PML) absorbing boundary condition. In the simulation, a pulse was

incident on the center of the area and the remaining light energies in the area were calculated. For the incident light, we used a pulse with a center wavelength of 365 nm and a width of 14 fs, which was irradiated from t=-70 to t=70 fs and was peaked at t=0 fs, as shown in Fig. 4(b). The area irradiated by the incident light was a disk with a diameter of 100 nm at the center of the system. The spectral region of the incident pulse, given by its Fourier transform, is about 355–375 nm, which covers the spectral range of the excitation pulse and the spontaneous emission of GaN. The FDTD time increment was 10 as (attoseconds), and total calculation time was 2.2 ps. The calculations were done with high accuracy of more than 13 digits.

Figure 4(b) shows the total electromagnetic energy that remains in the system as a function of time. The total energy for each sample is normalized to the charged energy. As shown in Fig. 4(b), the energy decay rates are quite different among the samples. We can see that Sample-2 shows the strongest localization effect for the observed wavelength region, while Sample-1 is the most diffusive system. This is the reason why Sample-2 shows clear random lasing and Sample-1 shows no change in its spectrum. It should be noted that the strength of the light localization depends on several parameters of the structure. Sample-3 has the largest filling fraction, but the smallest average diameter, indicating that the scattering efficiency is lowest. In our experiment, therefore, Sample-2 is the system with the optimum parameters for random lasing. Since the sample qualities are almost the same for the three samples, this result clearly shows that the buildup of the random lasing is closely related to the strength of light localization in the system.

In summary, we have observed random laser action in self-organized GaN nanocolumns. As far as we know, this is the first detailed report of such random lasing connected to the strength of the light localization. Comparing the numerical calculations with the experiments has confirmed the close relationship between the appearance of random laser action and the localization of light. So far we have not studied details of the deflection properties such as direction and polarization dependences of emission. These are important subjects left in the future.

Acknowledgements

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Figures:

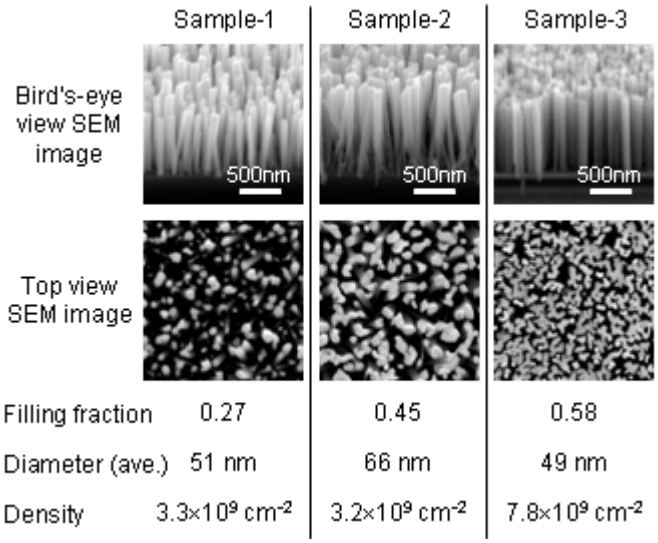

Figure 1: Bird's-eye view and top view SEM images of GaN nanocolumn samples. Three samples typically differ on filling fraction.

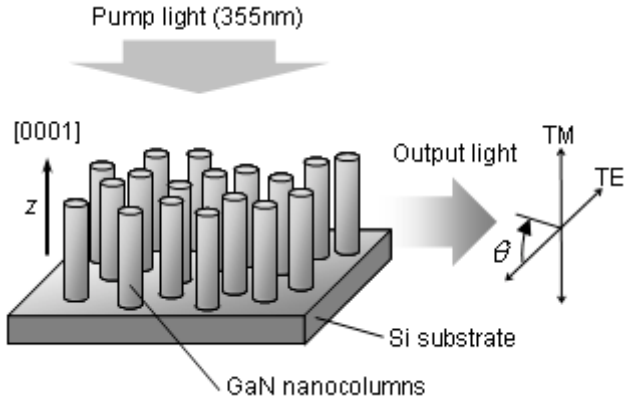

Figure 2: Schematic of experimental setup.

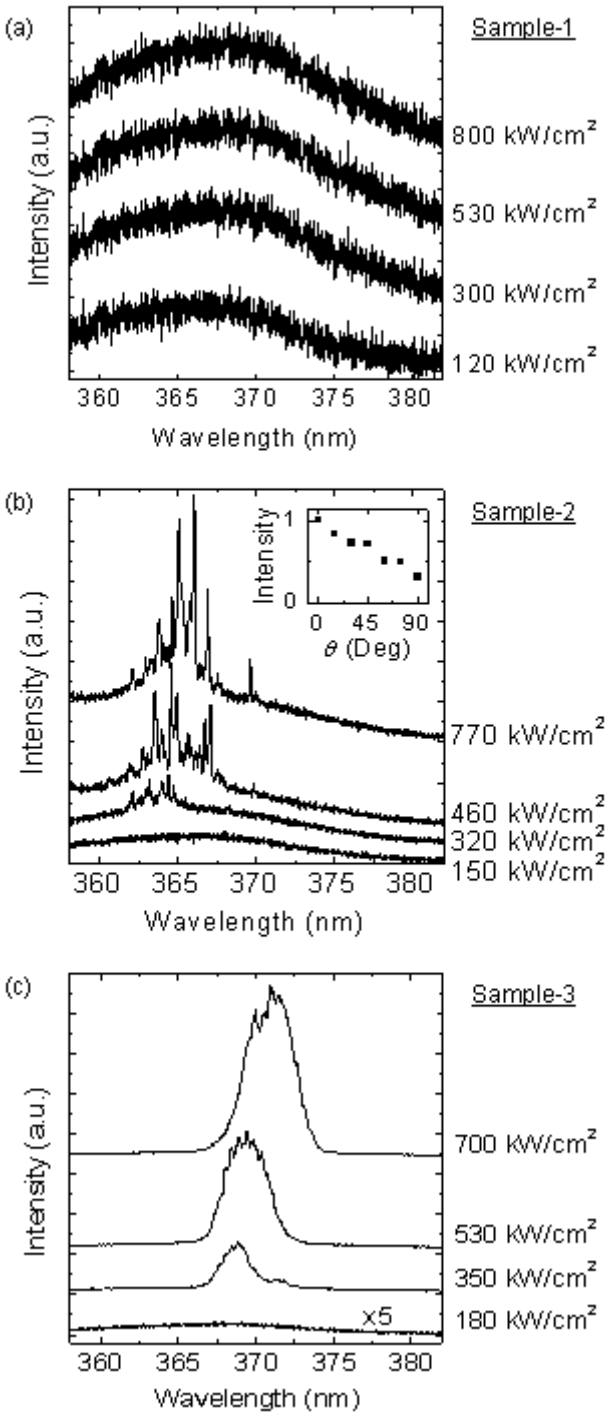

Figure 3: Spectra of photo-excited experimental results under various excitation densities for (a) Sample-1, (b) Sample-2 and (c) Sample-3. Random laser was clearly observed in Sample-2. Inset of (b): polarization behavior of random laser observed in Sample-2. Random laser in GaN is mostly polarized to TE.

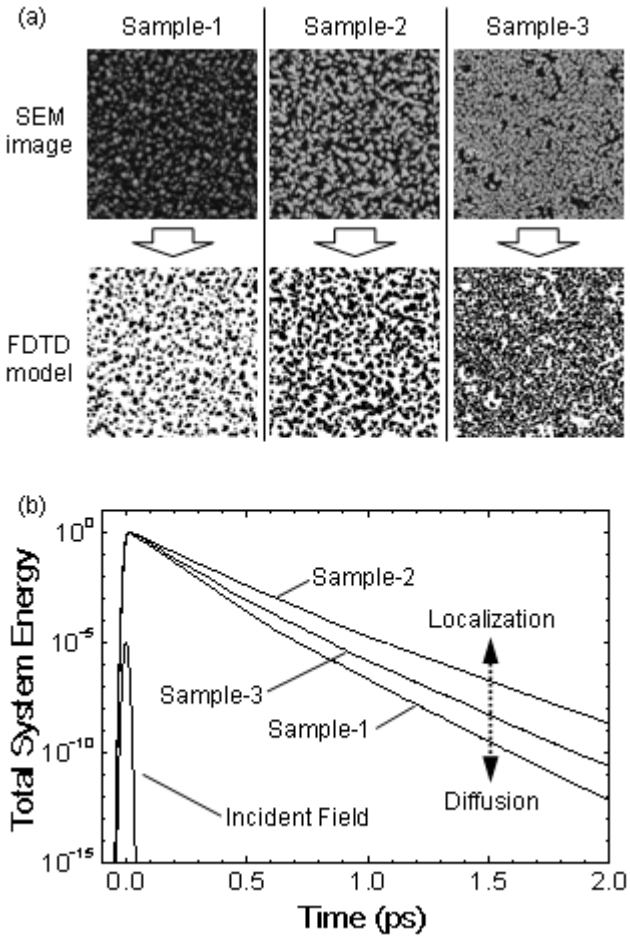

Figure 4: (a) Images of sample model used in 2D-FDTD calculation. FDTD models were directly translated by SEM image of samples used in optical experiment. Black area denotes GaN and white area denotes air in FDTD model. (b) Total system energy as a function of calculation time. After 2.0 ps, localization of light is strongest in Sample-2.