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Amplified spontaneous emission from a surface-modified GaN film fabricated under pulsed intense UV laser irradiation

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We propose a simple method for fabricating random structures directly on Mg-doped GaN thin films. The process is relatively simple, involving only irradiation with strong UV pulses from a fabrication laser on the thin-film surface. After intense UV laser pulses ($>400 \text{ MW/cm}^2$) are irradiated on the flat GaN film, the surface is roughened and quasi-periodic structures form. When the roughened surface is excited with laser light of intensity about 10 times smaller than the fabrication laser intensity, emission increases around 367 nm, and spectral narrowing and threshold behavior are observed. Because such behaviors are not observed without the application of intense laser irradiation, we conclude that amplified spontaneous emission is induced in the modified GaN surface by the intense UV laser pulse irradiation. This method offers the possibility of easy and direct fabrication of microscale random-laser devices on semiconductor substrates. *Published by AIP Publishing.* <https://doi.org/10.1063/1.5040551>

Gallium nitride (GaN) is an attractive material for various semiconductor applications because of its high emission efficiency, high electrical conductivity, high binding energy, transparency to visible light, and its wide emission range.^{1–3} GaN has been widely applied in UV-blue light emitting diodes (LEDs) for use in sterilization, optical recording, and laser displays. These GaN-based LEDs typically feature a Fabry–Perot cavity structure formed from the end facets of a cleaved or plasma-etched GaN film grown on a hard substrate.^{4,5} However, due to the hardness of the substrate, the cleaving process remains a manufacturing challenge in terms of complexity and consistency. Furthermore, although micro-pillars, nano-columns, and micro-disks have also been adopted as laser-cavity structures and exhibit high-performance laser characteristics,^{6–8} these also require complex fabrication processes with high fabrication costs. In contrast, random structures can be produced with relative ease and low cost. These structures are typically composed of nanoparticle assemblies or surface roughness, and are also enabled to induce laser oscillations due to random optical feedback in the structures caused by multiple light scattering, so they are referred to as random lasers.^{9–12} These random lasers have recently been studied as unique speckle-free laser light sources that are particularly well-suited for use in sensors and full-field imaging.¹² Thus, such a random structure could be one of the promising structures for the fabrication of GaN-based laser and/or LED sources;^{13,14} e.g., Zhu *et al.* proposed a simple and low-cost GaN-film laser, in which random lasing in GaN epitaxial films is induced by weak coherent feedback that is caused by randomly distributed defect pits acting as scatterers.

In our previous studies,^{15–18} we proposed fabrication methods for random lasers that allow control of the lasing modes by selecting the size of spherical nanoparticles or nanorod arrays. These methods were intended to produce random lasers with a low excitation threshold. Considering

the potential applications such as LEDs and photovoltaic devices, improving the electrical-drive properties and lowering the excitation threshold of random lasers and LEDs are crucially important. The cost and complexity of the method for directly fabricating nanostructures on semiconductor substrates also need to be reduced. As an approach to fabricating structures on a substrate, we have paid attention to the fabrication technique for laser-induced periodic structures,^{19–22} in which clear periodic surface structures can be fabricated on various material surfaces, such as glasses, metals, and semiconductors, directly and simply with irradiation by a pulsed intense linearly polarized laser. This method offers the advantages of direct fabrication on the material surface, which is advantageous for the electric-drive properties of the lasing device. In addition, the laser-induced periodic structures offer the chance to control the structure by adjusting the fabrication conditions, resulting in lowering thresholds.

In this study, we propose a simple and low-cost method for fabricating a GaN-based light emitting device, by which a steep increase in UV emission was observed after only irradiating a GaN film with a strong laser to roughen the surface. This technique may be applicable to produce GaN-film lasers at a very low cost. In addition, the results have important implications for fabrication processes with wider-bandgap materials such as BN and diamond, as a similar method may be used to fabricate short-wavelength ultraviolet random lasers from these materials.

In the experiments, a commercially available Mg-doped GaN film (4- μm thickness on a sapphire substrate, MTK Co., Ltd.) was placed on a microscope stage. To fabricate the roughened GaN surface, intense UV laser pulses (355 nm, 1 kHz, 300 ps, spot diameter $\sim 50 \mu\text{m}$, linearly polarized) were irradiated through an objective lens (60 $\times$, NA = 0.85). When the irradiated intensity was above $\sim 400 \text{ MW/cm}^2$, we observed structures fabricated on the surface [Fig. 1(a)]. Figures 1(b) and 1(c) show SEM images of the GaN surface after the irradiation of intense pulses (intensity: $\sim 550 \text{ MW/cm}^2$, irradiation time: 10 s). The polarization is indicated by

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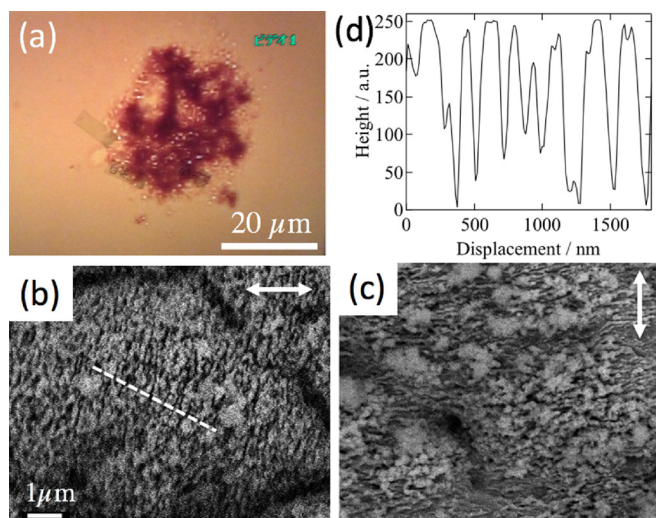


FIG. 1. GaN surfaces after surface treatment with intense UV laser irradiation pulses. (a) Microscopy image. (b) and (c) SEM images of the GaN surface after intense laser irradiation with different polarizations, indicated by arrows. Irradiated UV laser intensity and time in all cases were $\sim 500 \text{ MW/cm}^2$ and 10 s. (d) Line profile along the white dashed line marked in (b).

the arrows in the figure. From the results, we found that the smooth surface of the GaN film was roughened by the intense irradiation pulses. Quasi-periodic structures with a periodicity of 250–300 nm were confirmed [Fig. 1(d)], and they were formed perpendicular to the polarization direction. Similar results have been reported with the use of intense femtosecond-laser irradiation to produce laser-induced periodic structures.^{19–22} After fabricating the modified GaN surfaces with intense pulses, we measured the emission spectra of the samples using the same laser at an intensity of 100 MW/cm^2 as an excitation source. Hereafter, we refer to the UV pulsed laser used for surface modification as a fabrication laser and when used for emission spectral measurements, we refer to it as an excitation laser. Note that we attempted to confirm the lasing using an Mg-doped (p-type) GaN in the UV region (near-band-edge emission) because lasing oscillation using non-doped (n-type) GaN had been reported in various kinds of resonators. Because Mg ions generate other radiative transitions (450 nm), there is a possibility that it may prevent lasing at the near-band-edge.

Figure 2 shows the broadband emission spectra of the GaN films before and after surface modification, as induced by weak excitation. The laser intensity during the surface modification process was fixed at $\sim 550 \text{ MW/cm}^2$, which is the same intensity that was used to fabricate the samples shown in Fig. 1. Figure 2(a) shows the emission spectra from the untreated sample with excitation intensity 0.13–18.2 MW/cm^2 . A broad peak appears around 450 nm, which is typically attributed to doped Mg ions.²³ If the excitation intensity is increased, the near-band-edge emissions at $\sim 370 \text{ nm}$ and visible emissions around 550–600 nm increase. Comparing this result with the emission spectra after surface modification with intense UV irradiation pulses, slight changes can be observed in Fig. 2(b). The emission intensity at 450 nm slightly decreases and the intensities of the UV and visible emissions slightly increase. These changes occur because the evaporation of Mg ions increases UV emissions,

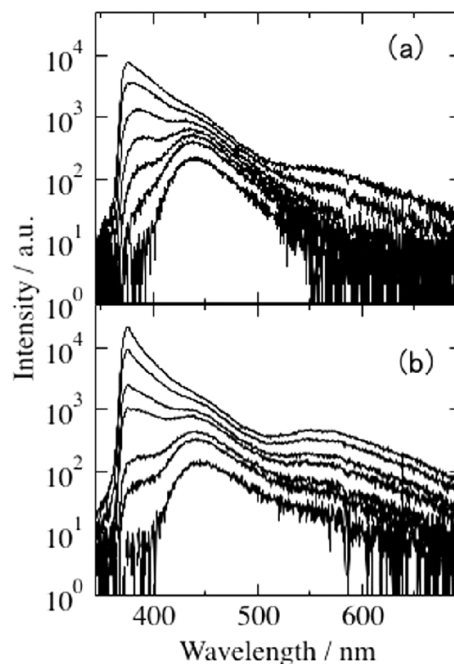


FIG. 2. Emission spectra (a) before and (b) after intense pulsed UV laser irradiation (550 MW/cm^2 and 10 s) under weak excitation. Excitation intensities were 0.13, 0.55, 1.1, 2.7, 5.3, 12.0, and 18.2 MW/cm^2 , respectively, which are much smaller than the thresholds discussed below.

and the increase in structural defects because of the strong laser irradiation also increases visible emissions.

We measured the dependence of the emission spectra on the excitation intensity to confirm emission behaviors induced by the surface-modified GaN film. Figure 3 plots the integrated emission intensities and emission spectra versus excitation intensity. The excitation intensities were varied from 9 to 225 MW/cm^2 , which were sufficiently lower than

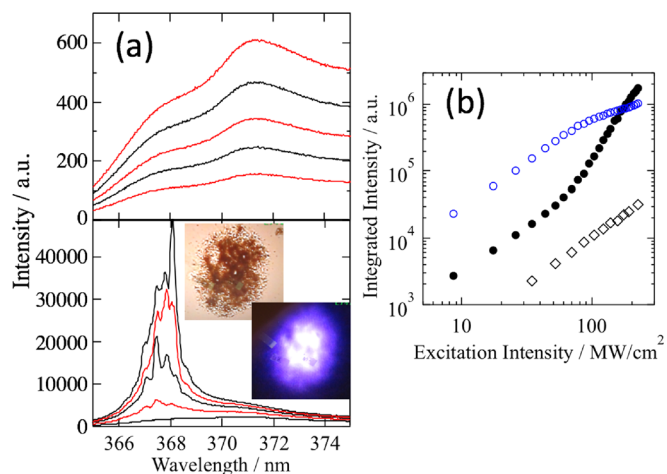


FIG. 3. (a) Emission spectra from a non-treated and modified GaN surfaces (upper and lower panels, respectively) under strong excitation. The excitation intensities are 52, 121, 156, 190, and 225 MW/cm^2 from bottom to top. The insets show the modified surface and its emission images. (b) Integrated emission intensities versus excitation intensity. Solid circles: wavelengths from 366 to 369 nm, open circles: wavelengths from 371 to 385 nm emitted from the modified GaN surface. Diamonds: wavelengths from 366 to 369 nm for the non-treated flat GaN surface. Note that the maximum excitation intensity is still about 3 times smaller than that used for the fabrication of surface-modified GaN surfaces and no damage was caused to the flat GaN surface by the emission-measurement procedure.

the fabrication laser intensity ($\sim 560 \text{ MW/cm}^2$), but much higher than those used for the results shown in Fig. 2. For comparison, first, we measured the emission spectra from a flat GaN surface (before surface modification) and found only broad and weak emissions around 368 nm, which is the near-band-edge emission from GaN. However, no spectral change and threshold behavior was observed in this range of excitation intensity [shown in the upper panel in Fig. 3(a) and marked with diamonds in Fig. 3(b)]. However, after surface modification, the emission spectrum measured at the same position indicated an increase in UV emission around 368 nm with narrowing spectral shape [lower panel in Fig. 3(a)]. The insets in Fig. 3(a) show the modified surface and its emission images. We found that strong UV emission was induced from the modified region (black region in the left image) and threshold behaviors could be observed from these bright spots, while from untreated surfaces, we only observed weak spontaneous emission. These results imply that the threshold behavior is strongly related to the surface modification. Figure 3(b) plots emission intensities against excitation intensity. Emission intensities integrated around the broad emission peak from 371 to 385 nm (open circles) show linear growth until saturation, while the intensities integrated from 366 to 369 nm (solid circles) exhibit a steep increase at the excitation intensity of $\sim 70 \text{ MW/cm}^2$, which is about 8 times smaller than the fabrication laser intensity. However, from the flat GaN surface, the UV emission intensity is relatively small and increases linearly with increasing excitation intensity. Note that we observed no damage to the surface caused by the excitation measurements of the flat surface. Also note that similar behaviors could be observed from the undoped GaN thin film only after the intense fabrication laser irradiation. Although the threshold behaviors were observed, peak fluctuations that have generally been observed in random lasers could not be confirmed when we changed the excitation intensity. We note that it may suggest the possibility of amplified spontaneous emission rather than the possibility of random lasing with coherent random feedback. On the other hand, in the experiments, such threshold behavior and increase in emission intensity could not be confirmed from a flat GaN substrate and observation was carried out in the direction perpendicular to the substrate with the small excitation spot diameter of $50 \mu\text{m}$. Therefore, we considered that, due to the surface modification, the observed phenomena could be induced by the increase in the optical path lengths of excitation light or emission due to the surface modification.

To investigate the influence of surface modifications, we measured the emission spectra and took SEM images of modified Mg-doped GaN surfaces with different irradiation intensities. Figure 4(a) shows the emission spectra from GaN surfaces fabricated with fabrication laser intensities from 347 to 644 MW/cm^2 and a fixed irradiation time (10 s). The excitation laser intensity for these measurements was fixed at 248 MW/cm^2 , which was sufficiently higher than the observed thresholds shown in Fig. 3. If the fabrication laser intensity was relatively weak, less than $\sim 500 \text{ MW/cm}^2$, only a broad emission peak was observed even though the UV emission increased as the fabrication laser intensity increased. When the fabrication laser intensity was $\sim 550 \text{ MW/cm}^2$, the shoulder of the broad emission spectrum

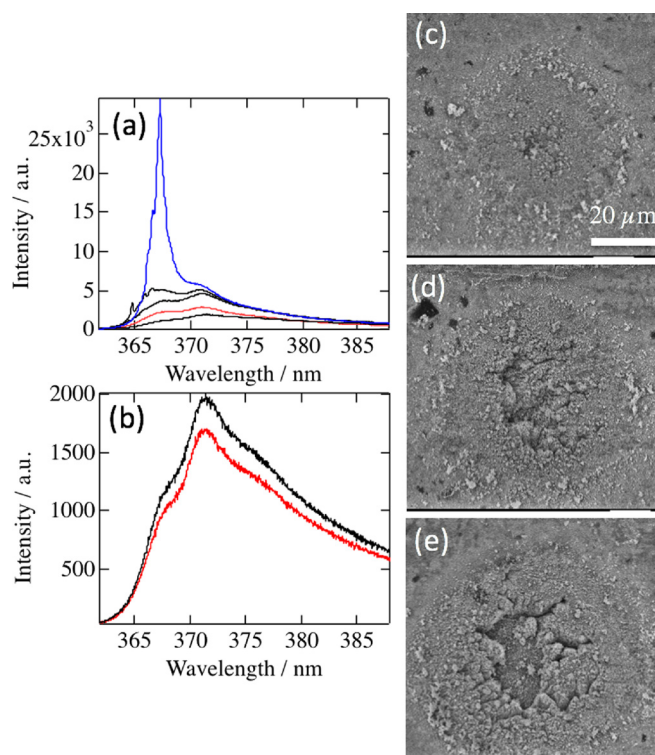


FIG. 4. Emission spectra from the modified GaN surfaces with different UV pulse irradiation conditions. (a) Fabrication irradiation intensities were 347, 495, 545, 594, and 644 MW/cm^2 , while the irradiation time was fixed at 10 s. The excitation intensity for measuring emission spectra was fixed at 248 MW/cm^2 , which is sufficiently higher than typical threshold values in Fig. 3. The blue and red lines highlight the results from samples made at irradiation intensities of 594 and 644 MW/cm^2 , which correspond to the SEM images in (d) and (e). (b) The black and red lines plot the emission spectra of samples fabricated with different irradiation times (black line: 10 s, red line: 2 min.) and fixed fabrication laser intensity at 495 MW/cm^2 . (c–e) SEM images of the samples made with fabrication laser intensities of 347, 594, and 644 MW/cm^2 .

grew around 367 nm. When the fabrication laser intensity was increased to $\sim 600 \text{ MW/cm}^2$, a steep increase in the emission intensity and spectral narrowing at 367 nm were clearly observed [blue line in Fig. 4(a)]. This result is marked with a blue line in the figure. However, further increases of the fabrication laser intensity led to a sudden decrease in UV emission that only showed a broad emission, the result of which is marked with a red line in the figure. These phenomena were observed repeatedly at other positions on the modified GaN surface. In addition, Fig. 4(b) indicates the emission spectra of the samples prepared with different irradiation times. The fabrication laser intensity was set at 495 MW/cm^2 , which is slightly lower than the fabrication laser intensity that we could observe from the threshold behavior of the samples, and the irradiation time was set to be either 10 s (black line) or 2 min (red line). Even with the longer irradiation time, only broad emissions were observed. This means that irradiation intensity is the dominant factor in fabricating surface structures that initiate the observed phenomena.

Figures 4(c)–4(e) show SEM images of the samples used in Fig. 4(a), for which the fabrication laser intensities were (c) 347, (d) 545, and (e) 644 MW/cm^2 , respectively. From these SEM images, we find that increasing the fabrication laser intensity increased the surface roughness.

However, when the fabrication laser intensity was too strong [644 MW/cm^2 , marked with the red line in Fig. 4(a)], the irradiating laser ablated the GaN thin film so that the sapphire substrate is visible from the surface [Fig. 4(e)]. From the above results, we conclude that, with weak fabrication intensities, the surface is not modified enough to provide multiple light scattering and, at stronger fabrication intensities, the thin GaN layer is nearly ablated under the irradiation spot so that insufficient amplification is produced. Thus, the optimal irradiation intensity for fabricating the surface roughness exists. In this study, to fabricate a modified GaN surface that undergoes multiple light scattering, we found that the optimal irradiation conditions were $\sim 550 \text{ MW/cm}^2$, samples of which are shown in Fig. 3.

As quasi-periodic structures on the GaN surface were also fabricated by intense fabrication laser irradiation [see Figs. 1(b) and 1(c)], this periodicity may have been responsible for lasing feedback. However, the emission spectral measurements revealed no clear influence of the periodic structures (different peak wavelengths were observed at different positions). In addition, although we tested the detection polarization dependence, we also observed no clear difference with different polarizations. Thus, in contrast with the previous studies of distinct periodic structures fabricated by femtosecond-pulsed laser irradiation,^{19–22} our experimental results could be explained as an effect of multiple light scattering within the modified surface, rather as an effect of periodic structures. This effect would be explained by the low profile quality of the picosecond-pulsed laser, which is evidenced by the modified surface shown in Fig. 1. Our findings may suggest the possibility of a simple method to directly fabricate LEDs or distributed-feedback lasers on a GaN surface at a greatly reduced cost, if we use a femtosecond laser with a good profile, as used in previously reported studies.^{19–22}

In summary, we demonstrated the fabrication of UV light emitting devices from GaN thin films in a process that uses only strong UV laser pulses ($\sim 550 \text{ MW/cm}^2$). After irradiating intense UV pulses on the GaN thin film, the surface was roughened and near-band-edge emissions increased. When the excitation intensity exceeded 70 MW/cm^2 , which is still about 8 times less than the fabrication laser intensity, we found the spectral narrowing and steep increase in the emission intensity at around 367 nm, although these behaviors were not observed in samples not treated with strong UV irradiation pulses. We found the optimal fabrication laser intensity for fabricating optimal modified GaN surfaces, at $\sim 550 \text{ MW/cm}^2$ by varying the fabrication laser intensity. Because the method is quite simple, needing solely the irradiation of intense lasers on a material surface, it may be extended to use with a wide range of other semiconductor materials. Furthermore, the microscale random structures fabricated by this method may find application in photonic materials for use in photo-catalysis, in photoelectric conversion, and in light harvesting.

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