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Lasing characteristics of ZnO nanosheet excited by ultraviolet laser beam

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ABSTRACT

The lasing characteristics of ZnO nanosheets were investigated for an application to ultraviolet laser diode. ZnO nanosheets were synthesized on a silicon substrate by a CVD method, and then those ZnO nanosheets were examined by a photoluminescence method with a third-harmonic Nd:YAG laser (355 nm, 5 ns). The observed emission spectra showed the obvious lasing characteristics having mode structure and a threshold for lasing. The threshold power density of a ZnO nanosheet was measured to be 40 kW/cm^2, and it would be low enough to be oscillated by an electrical pumping for ultraviolet laser diode.

Keywords: ZnO nanosheet, laser; ultraviolet; photoluminescence.

Kota Okazaki is a Ph.D. student of information science and electrical engineering (ISEE) from Kyushu University in Japan. He has been working on an evaluation of ultraviolet lasing characteristics from a single ZnO nanocrystal and an application to an ultraviolet laser diode.

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Introduction

Zinc oxide (ZnO) is one of the most promising semiconductor materials in ultraviolet (UV) region. ZnO has a wide band-gap of approximately 3.37 eV at room temperature and a large exciton binding energy of approximately 60 meV that is significantly larger than the thermal energy at room temperature which corresponds to 26 meV. It can ensure an efficient exciton emission at room temperature with low excitation energy. Furthermore, since there is a great deal of resources of ZnO, ZnO is a promising material replacing GaN in UV applications.

Nanostructured ZnO has attracted a great interest due to their importance in both scientific research and potential technological applications. From its excellent crystallization, UV stimulated emission at room temperature from optically pumped ZnO nanocrystals has been reported. In addition, there are various lasing mechanisms with other ZnO nanocrystals such as Fabry-Perot (FP) cavity effect in a ZnO nanowire[**1**] and a nanoribbon[**2**], whispering gallery mode (WGM) in a ZnO nanowire[**3**], a random lasing from ZnO nanoparticles[**4**,**5**] and nanobelts[**6**], lasing inside a ZnO nanosheet[**7**,**8**]. These reports show that ZnO nanocrystals can be candidates for excellent UV laser mediums. However, the concrete lasing mechanisms from ZnO nanocrystals remain to be established, and there are some major challenges to solve. For an UV LD application, it is necessary to clarify the mechanisms and the characteristics of the ZnO nanocrystals. In this study, we report on the further investigations of the lasing characteristics such as an oscillation spectra and a threshold power density from a single ZnO nanosheet under a light excitation.

Experimental

Synthesis of ZnO nanocrystals with carbothermal CVD method

ZnO nanocrystals were synthesized by a carbothermal CVD method. The mixed sample of ZnO powder and Graphite was used, in which Graphite worked as a catalyst. A silicon substrate on which ZnO nanosheets are synthesized was used. In advance, the silicon substrate was coated by gold thin film with a thickness of 1 nm, which was done by use of vacuum deposition equipment. The gold also works as a catalyst promoting the synthesis of ZnO nanocrystals. The sample was placed in an alumina boat, and then the silicon substrate was located above about 10 mm from the sample where the gold-coated surface was opposed to the sample. The prepared alumina boat was inserted to the center of a silica glass vacuum chamber. Next, the air in the camber was evacuated up to several Torr by a vacuum pump, and then the mixed gases of argon (Ar) and oxygen (O_2) were flowed. The amounts of gas flow of Ar and O_2 were 100 sccm and 3sccm, respectively.

ZnO nanosheets were synthesized at the temperature of 1100 °C, the ambient gas pressure of 300 Torr and the synthesis time of 30 min. **Fig. 1** shows the scanning electron microscope (SEM) (KEYENCE, VE-7800S) image of the nanosheets synthesized on a silicon substrate. ZnO nanosheets were synthesized in a form of an island as shown in **Fig. 1** (**a**), and the thickness of the nanosheets in **Fig. 1** (**b**) was about 570 nm.

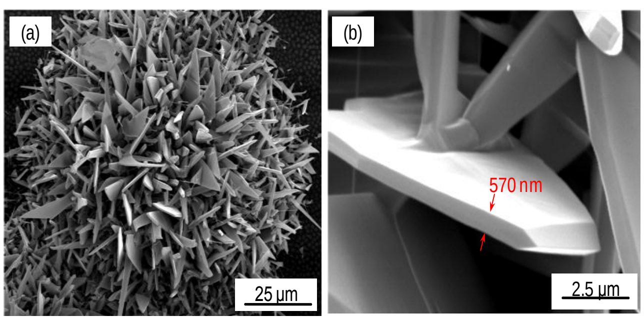

Fig. 1. SEM images of ZnO nanosheets synthesized on a silicon substrate by a carbothermal CVD method. The ZnO nanosheets form an island (**a**) and the width of the single nanosheet was about 570 nm (**b**).

Arrangement for photoluminescence measurements

The ZnO nanosheets were examined by a PL method. The experimental configuration for the PL method is shown in **Fig. 2**. A third-harmonic Nd:YAG laser which has a wavelength of 355 nm and a pulse duration of 5 ns were used as an excitation light for the ZnO nanosheets. In advance, ZnO nanosheets were dispersed on a silica glass substrate, and then the silica glass substrate was fixed on a base which could be controlled by micrometers. The ZnO nanosheets on the silica glass substrate were observed by a conventional optical microscope which was applied for a detail observation of the PL from a single ZnO nanosheet and positioned with the micrometers under the optical microscope. The excitation laser was injected through the silica glass substrate, and then the PL from a ZnO nanocrystal was acquired by the optical microscope and transferred to a spectrometer (Lambda Vision, TC-2000) with a light fiber.

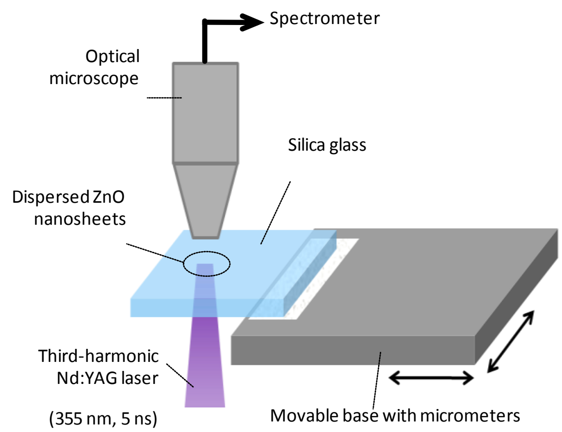

Fig. 2. Experimental configuration of photoluminescence (PL) method.

Results and discussion

The optical microscope image of the single nanosheet on a silica glass is shown in **Fig. 3** (**a**), and the nanosheet was observed by an atomic force microscope (AFM) (KEYENCE, VN-8000M/8010M) as shown in **Fig. 3** (**b**). The thickness of the nanosheet was about 400 ~ 900 nm. Furthermore, the nanosheet under the excitation with the Nd:YAG laser beam is shown in **Fig. 3** (**c**). **Fig. 3** (**d**) shows the emission spectra from the nanosheet in changing the excitation intensities. The peak intensities at *a*, *b* and *c* of the emission spectra against the excitation power densities were shown in **Fig. 3** (**e**). As shown in **Fig.** 3 (d), mode structures due to lasing from the nanosheet was observed. The lasing threshold power density was estimated to be about 40 kW/cm^2 from **Fig. 3** (**e**).

The pulse duration of the excitation laser was 5 ns which was adequately longer than the fluorescence lifetime of ZnO nanocrystals which is several hundreds ps and hence the excitation with a nanosecond pulse laser can be regarded as a continuous excitation. It indicates that the obtained threshold power density can be applied to an electrical pumping. Since the area of the nanosheet was estimated to be 66 µm^2 from the AFM results, threshold power was estimated to be about 26 mW by multiplying the area by the threshold power density. If the voltage of 30 V is applied to the ZnO nanosheet, the current of 0.88 mA is required to be oscillated. From these results, the laser oscillation of a ZnO nanosheet with electrical pumping can be realized by a total excitation from a lateral side.

Concerning ZnO nanosheets, there are only a few reports on lasing. The stimulated emission under the light excitation with a Ti: Sapphire laser (266 nm, 120 fs) from ZnO nanosheets grown by cathodic electrodeposition have been reported by F. Wang et al., and the lasing threshold was about 4.2 GW/cm^2 [**7**]. In addition, E. S. Jang et al. have reported that lasing due to narrowing of PL spectra with increasing excitation power density was observed from ZnO nanosheets prepared by a solution process [**8**]. The nanosheets on Si substrate were excited by a Nd:YAG laser (355 nm, 6 ns), and the lasing threshold was about 25 kW/cm^2. However, the oscillation mechanism whether

WGM or random lasing was not clear because a large number of nanosheets were totally excited. B. Q. Cao et al. shows random lasing from a large number of ZnO nanobelts excited by third harmonic of a Q-switched Nd:YAG laser (355 nm, 5 ns), and the lasing threshold was about 500 kW/cm^2 [**6**]. The nanobelts has similar configuration to nanosheets, even though the nanobelts are thinner (≈200 nm) than nanosheet. H. Yan et al. also shows the lasing spectra having mode-structure from a single ZnO nanoribbon which has a long and thin shape and the width, length and thickness were about 550 nm, 35 µm and 110nm, respectively [**2**]. The nanoribbon was excited by a femtosecond laser (310 nm, 200 fs), and the lasing threshold was 16 MW/cm^2.

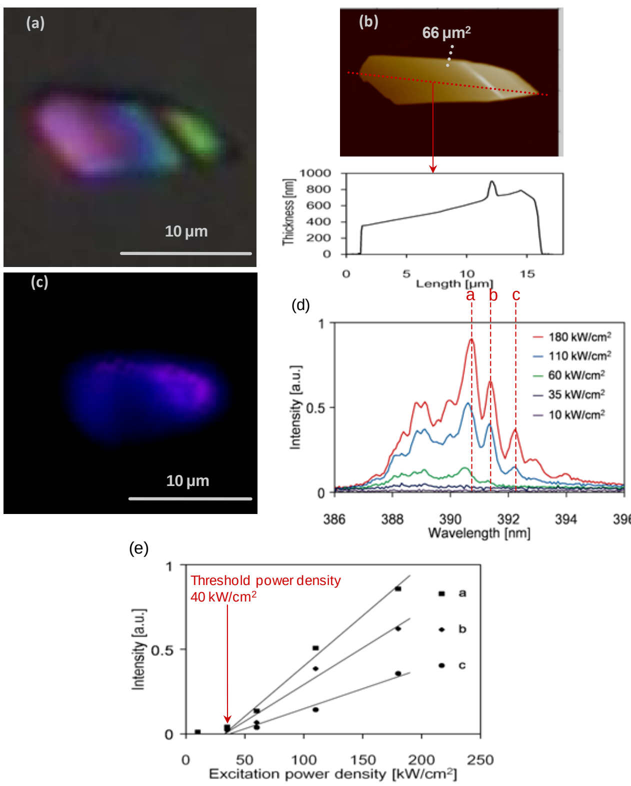

Fig. 3. (**a**) Optical microscope image of a single ZnO nanosheet on a silica glass. (**b**) Atomic force microscope image of the nanosheet which has the size of 400 ~ 900 nm in thickness and 15 µm in length. (**c**) Optical microscope image of the nanosheet under the excitation with third-harmonic Nd:YAG laser (355 nm, 5 ns). (**d**) Emission spectra from the single ZnO nanosheet. The mode structures due to lasing were observed. (**e**) The peak intensities of the spectra plotted at a, b, and c in **Fig. 3** (**d**) as a function of the excitation power densities. The threshold power density was estimated to be about 40 kW/cm^2.

In our study, the lasing from the single nanosheet prepared by a carbothermal CVD method were observed as well, and it should be noted that the oscillation spectra in **Fig. 3** (**d**) showed obviously clear mode-structure indicating the onset of lasing due to the detailed observation from the single ZnO nanosheet, and the lasing threshold was about 40 kW/cm^2 which is considerably lower as well as the report [**8**] than the lasing threshold of ZnO nanowire which is about 200 ~ 400 kW/cm^2 [**1,3,9**]. The lasing spectra having mode-structure from the single ZnO nanosheet indicates that the lasing mechanism was attributed to micro-cavity effect including Fabry-Perot or WGM and not random lasing. The thickness of the nanosheet was about 400 ~ 900 nm from the AFM result. Since the refractive index of ZnO is 2.4 at the wavelength around 390 nm, the ultraviolet lights can propagate inside the nanosheet, and the total reflection angle at the boundary of ZnO/air is calculated to be about 25° by the Snell's law, which is low enough to make the lights confinement easy. Thus, the oscillation route inside the nanosheet would not only be formed by the reflections at lateral side surfaces but also at upper and under surfaces, and WGM-like oscillation routes would be intricately formed inside the nanosheet.

Conclusion

A photoluminescence (PL) and lasing characteristics of a single zinc oxide (ZnO) nanosheet were investigated by an optical pumping with a third-harmonic Nd:YAG laser (355 nm, 5 ns). ZnO nanosheets were synthesized by a carbothermal chemical vapor deposition method. According to the PL spectra, lasing from the ZnO nanosheet was observed, and the lasing threshold power density was about 40 kW/cm^2 which was considerably lower than the reported threshold of ZnO nanowire (200 kW/cm^2 ~). It indicates that ZnO nanosheets can be a superior laser medium to ZnO nanowires. Furthermore, the ZnO nanosheet can be applied to an ultraviolet laser diode because the threshold power density would be low enough to be oscillated by an electrical pumping.

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