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Evaluation of Scintillation Properties of GaN*

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Optical and scintillation properties of GaN crystalline film were investigated. It showed 30-50% in-line transmittance at wavelength longer than 400 nm and intense emission appeared at 365 nm under 213 nm excitation in photoluminescence (PL) spectrum. When PL decay time was investigated under 280 and 200 nm excitation, luminescence decay time resulted few ns. In X-ray induced radioluminescence spectra, intense emission appeared at 365, 420, and 550 nm. The former one was ascribed to the exciton emission and the latter two would be due to defects related emission. Scintillation decay time of GaN was quite fast as few ns which was consistent with PL results and no slow component was observed. [DOI: 10.1380/ejssnt.2014.396]

Keywords: Synchrotron radiation photoelectron spectroscopy; Visible/ultraviolet absorption spectroscopy; Luminescence; Photoluminescence; Gallium nitride

I. INTRODUCTION

GaN is well known direct bandgap semiconductor that is used as a blue light emitting device (LED). Up to now, most studies about GaN have been devoted to electro- or photo-luminescence for LED applications. However, there is one possibility for another application, scintillation device. Scintillators are phosphors which convert high energy (keV MeV) ionizing radiation to thousands of visible photons immediately [1] and are widely used in common applications, such as medical imaging (PET and X-ray CT) [2], security systems in ports and airports [3], well-logging [4], astrophysics [5], and particle physics [6]. Direct transition semiconductor materials are potentially good scintillators because a fast decay time can be expected. One of good examples is ZnO. In ZnO, sub-nano seconds fast scintillation decay time due to a free exciton and medium level of scintillation light yield were observed [7]. Thus, similar scintillation properties can be expected in GaN.

GaN was rarely studied for radiation detectors and scintillation properties were reported few times [8-11]. In those reports, GaN was doped with Si or combined with converter materials such as LiF for thermal neutron detection. Almost all previous studies were devoted to investigate other kinds of luminescence, such as photoluminescence (PL) or cathodeluminescence. Very recently after our submission of this work, proton excited scintillation was reported [12] and the main aim of that study was not to grasp scintillation detector properties. Therefore, it means that scintillation responses of GaN are still unclear and there remains a room for study. In most scintillation detectors, majority is bulk materials with few cm to m to detect high energy photons (X-ray or γ-ray) or neutrons efficiently. However, film scintillators are suited

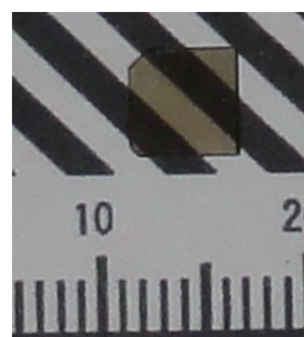

FIG. 1. Appearance of GaN sample, product of Optostar Ltd.

to charged particle (e.g., α-ray) detectors since high energy photons or neutrons become noise for charged particle detection and insensitivity of film scintillators against ionizing radiations with high penetration power are ideal. For this purpose, we [13] and some other groups [14-15] examined ZnO film scintillators for α-ray detectors.

In the present work, we investigated scintillation characteristics of GaN film scintillator for the first time. The sample was prepared by Optostar Ltd. and had a size of $5 \times 5 \times 0.3$ mm^3. Though in some other fields like semiconductor film means a sub-micron thickness, scintillation field recognizes several hundreds micron thick materials as film scintillators so that we call the sample as film in this work. Figure 1 represents an appearance of GaN. In this sample, wide surface areas were polished and oxygen impurity level was investigated by the manufacturer and it was around 10^{19} cm^{-3}. The origin of dark color was blamed for this oxygen impurities. In scintillators, dopant concentrations of famous materials are 1-10 mol% and the oxygen impurity level of the present GaN sample was smaller in several orders.

II. EXPERIMENTAL

In-line transmittance was evaluated by using JASCO V670 spectrometer from 190 to 2700 nm with 1 nm step. PL spectrum under 213 nm excitation was recorded with

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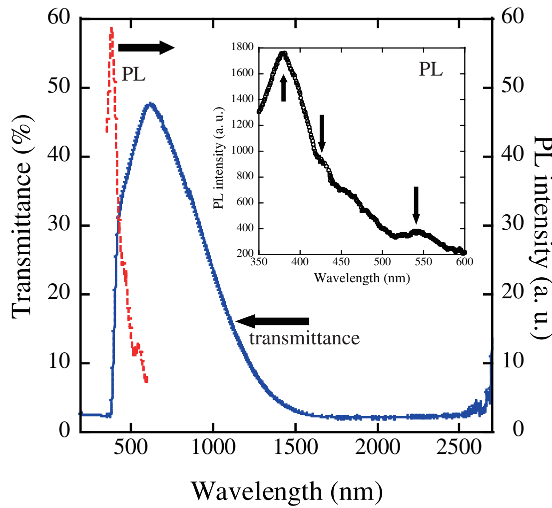

FIG. 2. Transmittance and PL spectra under 213 nm excitation of GaN. The inset shows the expanding image of PL.

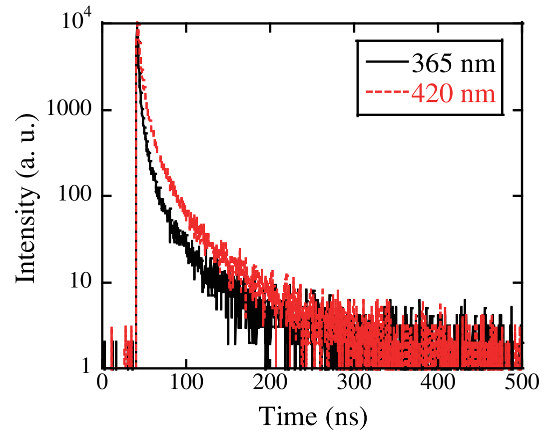

FIG. 3. PL decay time profiles of GaN under 365 and 420 nm excitation.

JASCO FP8600 spectrofluorometer. PL decay time profiles were investigated under 280 nm excitation which was the shortest wavelength of the instrument monitoring at 365 and 420 nm by using Hamamatsu Quantaurus-τ and under 200 nm excitation monitoring to 420 nm at Synchrotron facility (UVSOR BL-7B). The detailed explanation about Synchrotron experiments can be shown in our past report [16].

X-ray induced radioluminescence spectra was evaluated by using our original setup [17-18]. The excitation source was an X-ray generator supplied with 80 kV bias voltage and 2.5 mA tube current. Scintillation photons were fed into a monochromater via optical fiber and into CCD. Radioluminescence was evaluated by a reflection-type measurement as same as general PL measurements to avoid self-absorption of the sample. X-ray was irradiated at an angle of 45° to the sample and readout fiber was also put at an angle of 45° to the sample. Scintillation decay time profiles were evaluated by using pulse X-ray equipped streak camera system for scintillation characterization which was also developed by us [19-20] and became a product of Hamamatsu. The mean and maximum end point energy of X-ray were 20 and 30 keV, respectively. In scintillation spectrum and decay time evaluations, undoped ZnO was also evaluated for comparison. Undoped ZnO was a product of Tokyo Denpa and had a size of $10 \times 10 \times 0.5$ mm^3. All experiments were conducted at room temperature.

III. RESULTS AND DISCUSSIONS

Figure 2 demonstrates in-line transmittance and PL spectra of GaN. In-line transmittance was 30-50% at wavelength longer than 400 nm and reached to 0% around 1500 nm. In PL spectrum, one sharp line at 365 nm due to band-edge (exciton) emission was observed. This exciton emission was reported many times in PL and cathodeluminescence evaluations [21-22]. Compared with scintillation described later, broad lines around 420 and 550 nm were also observed. The origin of 420 emission was mainly ascribed to Ga vacancy (V_{Ga}) [23] and the most plausible explanation about 550 nm emission was unexpected contamination of very low content of Zn or Si [24]. In PL spectrum, exciton emission became quite intense under 213 nm excitation. Such a basic study about PL was investigated so many times (e.g., [23]) so that we did not discuss PL further.

In Fig. 3, PL decay time profiles using Hamamatsu Quantaurus-τ are shown. The instrumental response was deconvoluted and deduced decay times were 4.5 and 7.7 ns for 365 and 420 nm emission, respectively. Compared with ZnO, primary decay time was slightly slower but defect originated emission (420 nm) was faster than the defect emission of ZnO [25]. When 550 nm emission was investigated, significant difference with 420 nm emission was not observed. At least in PL, no slow emission was observed in GaN.

To investigate PL decay time under higher energy (200 nm) excitation, we used Synchrotron radiation (UVSOR). Figure 4 exhibits decay time profile of 420 nm emission under 200 nm excitation. As shown in the figure, main decay time resulted sub-ns which was significantly faster than 280 nm excitation. Sometimes excitation bands of defects emission appears around vacuum ultra violet wavelength in insulator materials [26]. By 200 nm excitation, exciton emission at 365 nm was not efficiently excited in our sample and the surface condition would affect such a PL evaluation.

Scintillation emission spectrum under X-ray irradiation is shown in Fig. 5. Although emission lines appeared around 365, 420, and 550 nm similar to PL one, intensities of each line differed. Exciton emission at 365 nm weakened on a comparison with PL spectrum partially due to a self-absorption in ultra violet wavelength of the sample since X-rays were absorbed not only at the surface. In this X-ray energy region, the X-ray energy is absorbed mainly by photoelectric effects. The penetration depth of several tens keV X-rays to GaN is at least several μm and this penetration depth is larger than that of PL. Though scintillation could be much suffered by self-absorption than PL, the clear detection of exciton emission suggested that GaN would be potentially available for scintillation detectors. In Fig. 5, scintillation spectrum of undoped ZnO

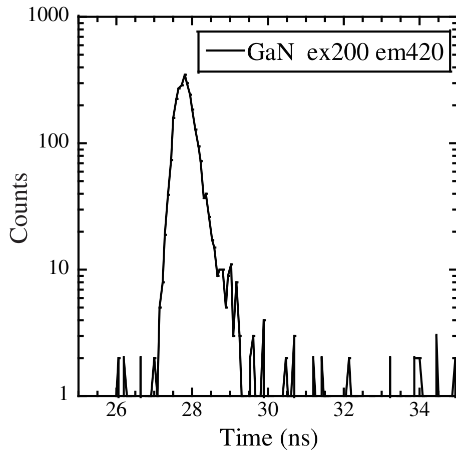

FIG. 4. Decay time profile monitoring at 420 nm under 200 nm excitation in UVSOR.

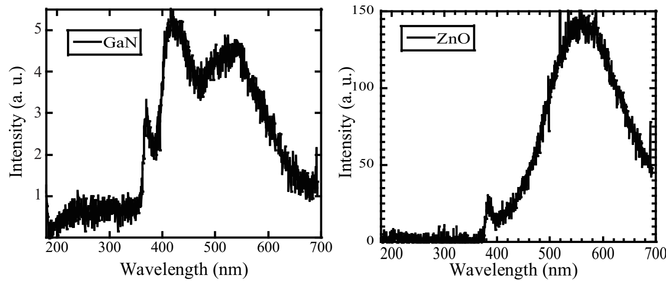

FIG. 5. X-ray induced radioluminescence spectra of GaN and ZnO. Bias voltage and tube current of X-ray generator were 80 kV and 2.5 mA, respectively.

was compared. In ZnO, free exciton emission at 380 nm and defect emission at 550 nm were observed. As same as GaN, exciton emission was not so intense in scintillation.

Figure 6 depicts scintillation decay time profile of GaN. The present data was not resolved in wavelength assuming an actual scintillation detectors so that all emissions (365, 420, and 550 nm) were included. The time profiles was well reproduced by a double exponential assumption. Deduced decay times were 1.1 ns (88%) and 6.5 ns (12%). The impressive point was that no slow component was observed in μs order observation. Scintillation decay time profile of undoped ZnO is also presented in Fig. 6. Obviously, very slow component due to defect was detected as well as fast luminescence. Fast and slow components resulted 3.3 ns and 7.7 μs, respectively. As clearly seen in the figure, most part of scintillation in ZnO was slow emission. In ZnO based scintillators, slow μs component due to oxygen vacancy at visible wavelength [27] sometimes causes a degradation of timing resolution of detectors. It is an advantage of GaN that does not have any slower decay component when compared with the other famous semiconductor scintillator ZnO.

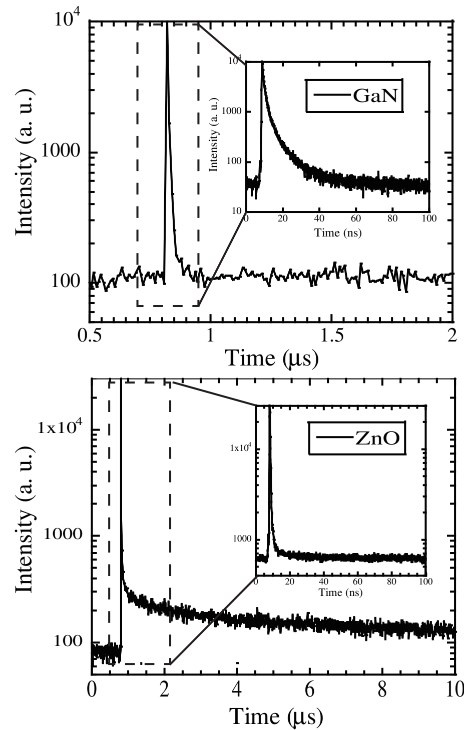

FIG. 6. X-ray induced scintillation decay time profiles of GaN and ZnO. Insets of both panels show expanding image. The mean energy of X-ray was around 20 keV.

IV. CONCLUSIONS

Optical and scintillation properties of GaN crystal were investigated. In-line transmittance showed 30-50% transmittance from visible to near infrared wavelengths. In PL and X-ray induced radioluminescence spectra, emission lines appeared at 365, 420, and 550 nm. The origin of 365 nm line was ascribed to exciton. Origins of 420 and 550 nm emissions were Ga vacancy and very low content of unexpected Zr/Si impurities. PL and scintillation decay times were quite fast and no slow component was observed. The advantage of GaN scintillator was a fast timing property when compared with other conventional semiconductor scintillator, ZnO.

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