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MODIFICATION OF THE YELLOW LUMINESCENCE IN GAMMA-RAY IRRADIATED GaN BULK SINGLE CRYSTAL

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Abstract.

We report the variation of the yellow luminescence (YL) in GaN bulk single crystals by gamma-ray irradiation. The crystals are irradiated at room temperature with gamma-rays of 1.17 and 1.33 MeV from a cobalt-60 source. Gamma-ray dose is 160 kGy. The resistivity varies from 30 Ωcm for an unirradiated sample to $10^4 \Omega$ cm for gamma-ray irradiated one. The nitrogen displacement in gamma-ray irradiated samples is observed by Rutherford backscattering channeling experiments using proton beam, suggesting the existence of the deep energy level relating to interstitial nitrogen atoms. The YL from the un-irradiated GaN with a peak at 557 nm (2.22 eV) is observed at around 440 nm to 800 nm, whereas that of the gamma-ray irradiated GaN shows a peak at 532 nm (2.33 eV) although the YL spectrum is almost overlapped with un-irradiated ones. Compton electrons emitted by the gamma-ray irradiation induce the shallow donor located at about 50 meV bellow the conduction band. This energy level is close to that of nitrogen vacancy. The modification of YL is attributed to a transition from the shallow donor induced by the gamma-ray irradiation to the native gallium vacancy.

I. Introduction

Examining the defects caused by various radiations to GaN under the space environment is important. In our previous study [1, 2], we reported that the energy levels relating to nitrogen vacancy (V_N) and gallium vacancy (V_{Ga}) were induced by neutron and proton irradiated GaN. The neutron irradiation has been used as the neutron transmutation doping into semiconductors such as GaAs [3], GaP [4], and GaN [5]. Atoms in semiconductors mainly transmute by a (n, γ) reaction. Therefore, to survey the radiation effect of gamma ray alone is meaningful. It is expected that Frenkel pairs on Ga and N sublattices are produced by the Compton electrons having a mean energy of about 700 keV [6]. In the present study, we report the variation of the yellow luminescence (YL) in GaN bulk single crystals by gamma-ray irradiation.

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II. Experiments

GaN bulk single crystals with a thickness of $450 \pm 50 \,\mu\text{m}$ were purchased from Furukawa Co. Ltd. The crystals were irradiated at room temperature with gamma-rays of 1.17 and 1.33 MeV from a cobalt-60 source of Kyoto University Research Reactor Institute. Samples were irradiated with an absorption dose rate of 1.771 KGy/h. Total gamma-ray dose was 160 kGy. The resistivity measured by van der Pauw method varied from 30 Ω cm for an un-irradiated sample to $10^4 \,\Omega$ cm for gamma-ray irradiated one. Photoluminescence (PL) spectra were measured at 77 K using a He-Cd laser. To evaluate the lattice displacement, Rutherford backscattering spectroscopy (RBS) analysis was performed using the Van de Graaff accelerator of Hosei University. 1.5-MeV H⁺ ions were used in the present experiment because the kinematic factor of N atoms for He-ion as a probing beam is very small. The experimental arrangement has already been reported elsewhere [7, 8]. The samples were mounted on a triple axis goniometer and the backscattered ions were collected by a silicon surface barrier detector placed at a scattering angle of $\theta = 150^\circ$. The beam spot and the beam divergence were ~1.0 mm in diameter and ~0.03°, respectively.

III. Result & Discussion

Figure 1 (a) shows the PL emission spectra before and after gamma-ray irradiation. The band edge emission was observed at 356 nm (3.48 eV) in both un-irradiated and gamma-ray irradiated samples. The YL from the un-irradiated GaN with a peak at 557 nm (2.22 eV) is observed at around 440 nm to 800 nm (see Fig.1(b)), whereas that of the gamma-ray irradiated GaN shows a peak at 532 nm (2.33 eV) although the YL spectrum is almost overlapped with un-irradiated ones (see Fig.1(a)). Therefore, the modification of YL observed in the present study would be attributed to a transition from the shallow donor, which is induced by Compton electrons emitted by the gamma-ray irradiation, to a native V_{Ga} . Since the usual YL has been proposed as a transition from a shallow donor to V_{Ga} located at about 1.1 eV above the valence band [9], the gamma-ray induced shallow donor is located at about 50 meV below the conduction band. This energy level is close to a donor level ($64 \pm 10 \text{ meV}$ [10]) relating to V_N . Figure 2 shows the YL model estimated from the photoluminescence at 77 K.



Figure 1. PL emission spectra before and after gamma-ray irradiation measured at 77 K. The PL intensity is normalized by the band edge emission. Fig.1(b) is the expansive spectra of YL.



Figure 2. Yellow luminescence estimated from the photoluminescence. The gamma-ray induced shallow donor relating to nitrogen vacancy is located at about 50 meV below the conduction band.



Figure 3 Random and aligned RBS spectra of un-irradiated and gamma-ray irradiated GaN bulk single crystal.

Figure 3 shows typical random and aligned RBS spectra of as-irradiated and un-irradiated GaN bulk single crystal. The aligned spectra were obtained from scattering along the <0001> channeling directions. The aligned yield increases with increasing the depth within the crystal since the dechanneling probability increases with increasing the depth. Therefore, the minimum yield χ_{min} (the ratio between aligned and random) of Ga and N was evaluated using a width of about 20 channels (120 nm) behind the surface peak. Near the surface, the number of atoms displaced into channels in both Ga and N is approximately related to the minimum yield by [11,12]

$$N_D = N_{Ga \text{ or } N} \left(\chi_{min} - \chi^0_{min} \right) / \left(1 - \chi^0_{min} \right), \qquad (1)$$

where $N_{\text{Ga or N}}$ is the Ga or N density $(4.43 \times 10^{22} \text{ cm}^{-3})$ in GaN, χ^0_{min} the minimum yield for the un-irradiated samples, and χ_{min} the observed minimum yield for irradiated ones. The values of χ_{min} for Ga were 1.5 % for un-irradiated and 2.3 % for gamma-ray irradiated samples. On the other hand, the values of χ_{min} for N were 7.7 % for un-irradiated and 9.5 % for gamma-ray irradiated samples. The number of displaced Ga and N atoms estimated using Eq. (1) were 3.5×10^{20} cm⁻³ and 8.7×10^{20} cm⁻³, respectively. Although the disorder is recognized in Ga and N lattices, the displacement concentration of N atoms is about two times larger than that of Ga atoms. This suggests that N interstitial (N_i) exists in gamma-ray irradiated GaN bulk single crystal. This result is similar to that of the neutron irradiated GaN [13]. Since N_i atoms form the deep acceptor level at 960 meV below the bottom of the conduction band [13], the origin of the high resistivity after the gamma-ray irradiation is attributed to the carrier compensation effect due to the deep level of N_i.

IV. Conclusion

By gamma-ray irradiation, the resistivity of GaN bulk single crystals increased from 30 Ω cm to 10⁴ Ω cm. The nitrogen displacement in gamma-ray irradiated samples was observed by Rutherford backscattering channeling experiments using proton beam. The high resistivity was attributed to the carrier compensation due to the deep acceptor level relating to interstitial nitrogen atoms. The peak energy of yellow luminescence after gamma-ray irradiation varied from 2.22 eV to 2.33 eV. The modification of YL was attributed to a transition from the shallow donor induced by the gamma-ray irradiation to the acceptor of native gallium vacancy. The shallow donor was related to nitrogen vacancy located at about 50 meV below the conduction band.

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