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Lattice disorder in neutron irradiated GaN: Nuclear reaction analysis and Rutherford backscattering studies

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Abstract

Disorders of N and Ga lattice in GaN, introduced by fast neutron irradiation with a fluence of 6.7×10^{18} cm^{-2}, are investigated by nuclear reaction analysis (NRA) using $^{14}N(d,p)^{15}N$ reaction with 2.6 MeV D_2^+ ions and by Rutherford backscattering (RBS) with 1.5 MeV $^4He^+$ ions. The $\langle 0001 \rangle$ aligned NRA yield measured in as-irradiated GaN slightly increases compared with that of un-irradiated crystals, indicating that primary knock-on (PKO) produced by the neutrons results in $\sim 7.2 \times 10^2$ displaced N atoms. The slight increase in the aligned RBS yield for as-irradiated samples relative to that of un-irradiated ones indicates that the $\sim 1.8 \times 10^2$ displaced Ga atoms are produced by PKO. The displacement of N atoms is four times larger than that of Ga atoms, reflecting the lighter weight of N than Ga, although both the displacements are recovered by annealing over 1000 °C. The displacement of Ga atoms in GaN is two times smaller than that in GaP ($\sim 3.0 \times 10^2$ atoms/PKO), showing the stronger bonds in GaN than GaP.

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1. Introduction

Gallium nitride (GaN) has unique electrical, optical and thermal properties, which make it a promising material for optoelectronic and high-power devices. However, GaN based devices would be affected by deep level defects, which act as carrier killer centers. Neutron-transmutation-doping (NTD) [1] is used to homogeneously introduce impurity atoms into semiconducting materials such as GaAs [2], GaP [3] and GaN [4]. In GaN [4], Ge and O impurities are transmuted from Ga and N atoms by (n,γ) reactions, respectively. The concentration of the neutron transmuted O atoms is negligibly small because of the very small thermal neutron capture cross section of nitrogen isotopes. After the transmutation reactions, the transmuted atoms are not usually located at their original positions but are at interstitial sites due to recoils by the γ- and β-rays produced through the nuclear reactions. Fast neutrons in a nuclear reactor mainly cause radiation damages in samples, while NTD is achieved by using thermal neutrons [2–4]. In NTD processes, large amounts of fast neutrons would induce various defects in GaN lattice such as vacancies, interstitial atoms and defect complexes [4–6], which is one of the disadvantages in the NTD process as observed in the other semiconductors. On the other hand, for space-based applications, GaN-based devices have to be able to

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operate in radiation environment. In other words, the device operation would be affected by deep level defects introduced by various cosmic rays. Therefore, it is important to study the lattice disorder introduced by fast neutron irradiation into GaN during NTD processes. In our previous study, we found that residual strain around Ga sites in neutron irradiated GaN is correlated with Ga vacancy-transmuted Ge complex defects by combining Rutherford backscattering (RBS) and Raman scattering spectroscopy [7]. In this paper, we apply nuclear reaction analysis (NRA) to obtain the direct evidence of N lattice disorder in GaN, since conventional RBS could only analyze Ga atoms in GaN. The conventional RBS measurement is complementarily shown to compare the N lattice disorder with the Ga lattice disorder.

2. Experimental

Samples used in this study were undoped n-type wurtzite GaN layers grown on a 40 nm AlN buffer layer on a sapphire substrate by metalorganic-vapor-phase epitaxy (MOVOP). Neutron irradiations were performed using the Kyoto University Reactor [2–4], which is a light-water-moderated reactor. The samples were irradiated with thermal and fast neutrons at fluxes of 8.15×10^{13} and $3.90 \times 10^{13}\ \mathrm{cm^{-2}\,s^{-1}}$, respectively. The fluences of thermal and fast neutrons were 1.41×10^{19} and $6.74 \times 10^{18}\ \mathrm{cm^{-2}}$, respectively. The Ge concentration transmuted by the irradiation for 48 h is calculated to be $1.77 \times 10^{18}\ \mathrm{cm^{-3}}$. Nuclear reaction (NR) analysis was performed here using $^{14}\mathrm{N(d,p)}^{15}\mathrm{N}$ reaction with 2.6 MeV deuter molecular ions (D_2^+) using a net beam accumulation of ~30 μC, as used for detecting interstitial carbons in C-doped GaN [8]. The total amount of the incident D_2^+ was $\sim 2.0 \times 10^{14}$ ions. The detection angle θ of the ejected proton was 165° with respect to the direction of the incident D_2^+ ion beam. In order to clearly separate the ejected protons from the backscattered deuterons, a mylar absorber film of 12 μm in thickness was placed in front of a detector. D^+ ions with an energy of 1.3 MeV in samples were chosen to give the maximum NR differential cross-section for $^{14}\mathrm{N}$ [9]. Conventional RBS and channeling techniques were complementarily used to measure Ga in GaN with a 1.5 MeV $^{4}\mathrm{He}^+$ ion beam using a Van de Graaff accelerator.

3. Results and discussion

Fig. 1 shows NR spectra taken in the random and aligned directions for neutron irradiated GaN, (a) un-irradiated, (b) as-irradiated, (c) annealed at 600 °C and (d) annealed at 1100 °C. The thicknesses of the samples (a)–(d) were estimated to be 3.3, 2.3, 2.3, 2.1 and 4.2 μm, respectively, from an energy loss of D^+ in the samples. Although a $^{14}\mathrm{N}(\mathrm{d},\alpha_1)^{13}\mathrm{C}$ reaction appears at around 1000–1200 channel (4.2–5.3 MeV), the present NRA was performed below 950 channel (4.2 MeV). Spectra arising from $^{14}\mathrm{N}(\mathrm{d},\mathrm{p}_{1,2})^{15}\mathrm{N}$, $^{12}\mathrm{C(d,p)}^{13}\mathrm{C}$, $^{14}\mathrm{N}(\mathrm{d},\mathrm{p}_3)^{15}\mathrm{N}$, $^{14}\mathrm{N}(\mathrm{d},\mathrm{p}_{4,5})^{15}\mathrm{N}$

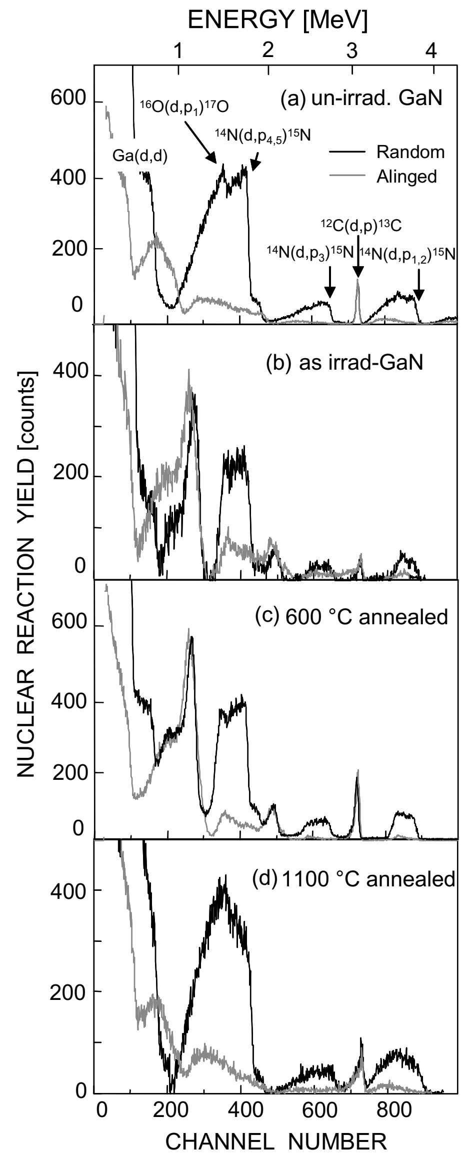

Fig. 1. Random and aligned NR spectra for (a) un-irradiated (thickness: 3.3 μm), (b) as-irradiated (2.3 μm), (c) 600 °C annealed (2.1 μm) and (d) 1100 °C annealed (4.2 μm) GaN. The nuclear reaction for each element is indicated in the figures.

and $^{16}\mathrm{O}(\mathrm{d},\mathrm{p}_1)^{17}\mathrm{O}$ reactions were observed at around 890, 740, 660, 430 and 320 channels, respectively. The increased yield in the aligned spectrum after neutron irradiation indicates the increased dechanneling from the ⟨0001⟩ direction due to the damage originated from the primary knock-on (PKO) by the fast neutron irradiation. $^{16}\mathrm{O}(\mathrm{d},\mathrm{p}_0)^{17}\mathrm{O}$ reaction appeared around 575 channel is superimposed on the spectrum from $^{14}\mathrm{N}(\mathrm{d},\mathrm{p}_3)^{15}\mathrm{N}$ reaction observed around 600 channel. For thinner GaN films as shown in spectra (b) and (c), $^{16}\mathrm{O}(\mathrm{d},\mathrm{p}_1)^{17}\mathrm{O}$ reaction was observed around 290 channel, separated from $^{14}\mathrm{N}(\mathrm{d},\mathrm{p}_{4,5})^{15}\mathrm{N}$, although these two reactions for the thicker samples (a) and (d) overlapped in around 400 channel. Moreover, in the samples (b) and (c), the shoulders beside $^{14}\mathrm{N}(\mathrm{d},\mathrm{p}_{4,5})^{15}\mathrm{N}$ were observed around

440 channel. The shoulder would be attributed to the NR from O in Al_2O_3 substrate, since the protons resulting from the NR suffer the less energy loss in thinner GaN films. Rutherford backscattering from Ga atoms in GaN due to a deuter molecular ion beam was also observed at around 110 channel (Ga(d, d)). Accordingly, $^{14}N(d, p_{1,2})^{15}N$ reaction is suitable for analyzing the N lattice disorder in neutron irradiated GaN, since no other reactions are overlapped. $^{14}N(d, p_{1,2})^{15}N$ reaction for the samples (a)–(d) is summarized in Table 1. The ratio between aligned and random yields (χ_{min}) from $^{14}N(d, p_{1,2})^{15}N$ reaction was evaluated for the reaction region (860–900 channels), because the scattering from greater depth requires knowledge of the probability of dechanneling by displaced atoms. Therefore, data near the surface are of quantitative use only. The observed χ_{min} (NRA) values (χ_{min} for NRA) were (a) 4.4%, (b) 6.6%, (c) 6.7%, (d) 4.0% for the samples (a)–(d), respectively. These values contain the uncertainty of ±0.5%. The χ_{min} (NRA) for as-irradiated GaN showed a slight increase relative to un-irradiated GaN, indicating that the N lattice disorder was introduced by fast neutron irradiation. The lattice disorder remained in the 600 °C annealed sample. However, the χ_{min} (NRA) has become comparable to the un-irradiated sample by annealing at 1100 °C, suggesting that the N lattice displacement in neutron irradiated GaN recovers by annealing at 1100 °C. On the other hand, the Ga lattice displacement [4,7] evaluated by the conventional RBS also recover by annealing at 1000 °C. In photoluminescence (PL) spectra for neutron irradiated GaN reported in our previous study [7,10], PL emissions relating to a DX-like center of Ge and a yellow band were enhanced by annealing above 1000 °C, although the band edge emission of GaN disappeared under the influence of these intense emissions. Therefore, the PL experiments support the present NR results.

Near the surface, the number of atoms displaced from a channel direction is approximately related to the minimum yield by [11]

$$N_D = n(\chi_{min} - \chi^0_{min})/(1 - \chi^0_{min}), \tag{1}$$

where n is the N or Ga concentration ($4.38 \times 10^{22}\ cm^{-3}$) in GaN. Using χ_{min} (NRA) values, the N atoms displaced from the $\langle 0001 \rangle$ channel are $\sim 1.0 \times 10^{21}\ cm^{-3}$ and $\sim 1.1 \times 10^{21}\ cm^{-3}$ for the samples (b) and (c), respectively. We also have complementarily applied conventional RBS and channeling techniques to measure the Ga lattice disorder in (A) un-doped, (B) as-irradiated, (C) 600 °C annealed

Table 1
Summary of NRA and RBS results for neutron irradiated GaN

Annealing temperature (°C)	χ_{min} (%)		N_D ($10^{20}\ cm^{-3}$)	
	NRA	RBS	NRA	RBS
Un-implanted	4.4	2.2	–	–
As-implanted	6.6	2.8	10	2.6
600 °C	6.7	2.6	11	1.7
>1000 °C	4.0	2.1	–	–

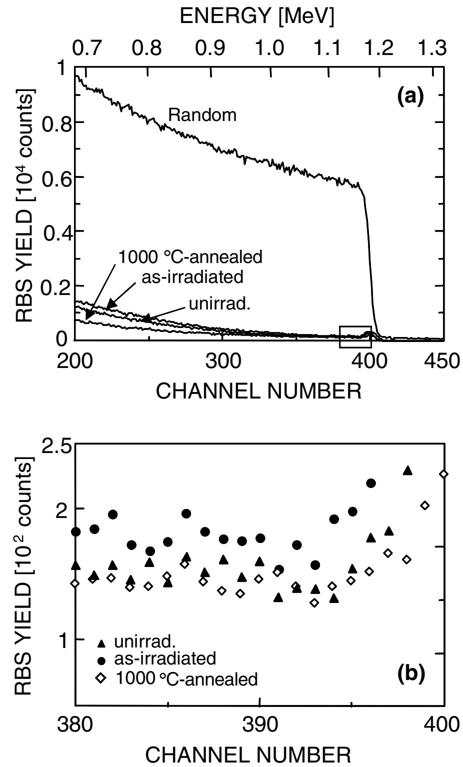

Fig. 2. (a) RBS spectra for 1.5 MeV He ions from GaN films. (b) Magnification for RBS aligned yields near the surface along the $\langle 0001 \rangle$ axis before and after fast neutron irradiation with a fluence of $6.7 \times 10^{18}\ cm^{-2}$.

and (D) 1000 °C annealed GaN, as shown in Fig. 2. The χ_{min} (RBS) values evaluated for the evaluation region (383–393 channels), were 2.2%, 2.8%, 2.6% and 2.1% for the samples (A), (B), (C) and (D), respectively, which also suggests the recovery of the Ga lattice disorder at 1000 °C. Using Eq. (1), the displaced Ga atoms in the as-irradiated samples are evaluated to be $\sim 2.6 \times 10^{20}\ cm^{-3}$, showing that the displacement of N atoms is about four times larger than that of Ga atoms.

In order to obtain more detailed information on the fast neutron induced damages, we discuss radiation damages in terms of the PKO. Using a mean free path of $\Lambda = 4.86$ cm [12] for neutrons with an energy of 2 MeV in GaN, the concentration of PKO is estimated to be $N_{PKO} = \Phi_f/\Lambda = 1.39 \times 10^{18}\ cm^{-3}$, where Φ_f is fast neutron fluence, $6.74 \times 10^{18}\ cm^{-2}$. Therefore, using the RBS and NRA data, each PKO produces 1.9×10^2 and 7.2×10^2 displacement atoms for the Ga and N lattice, respectively. The number of displaced Ga atoms was smaller than that of N, which would be attributed to the lighter weight of N than Ga. In our previous report [7], we made a simple numerical error on the calculation of N_D for Ga, which was one order of magnitude larger than that evaluated by the present study. Therefore, the Ga displacement is two times smaller than that of GaP ($\sim 3.0 \times 10^2$) [4]. These results confirm that the bonds in GaN are stronger in comparison with GaP.

4. Conclusion

In conclusion, we evaluated the disorder of the Ga and N lattices introduced into GaN during neutron irradiation. Fast neutron irradiation with a fluence of 6.7×10^{18} cm^{-2} approximately produced 1.9×10^2 and 7.2×10^2 displaced atoms per PKO for the Ga and N lattice, respectively. The displaced Ga atoms were smaller than those of N, which was attributed to the lighter weight of N than Ga. The Ga displacement was two times smaller than that of GaP ($\sim 3.0 \times 10^2$), confirming that the bonds in GaN are stronger in comparison with GaP.

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