Thermally stimulated current studies on neutron irradiation induced defects in GaN

K. Kuriyama,[a] M. Ooi, and A. Onoue
College of Engineering and Research Center of Ion Beam Technology, Hosei University, Koganei, Tokyo 184-8584, Japan

K. Kushida
Department of Arts and Sciences, Osaka Kyouiku University, Kashiwara, Osaka 582-8582, Japan

M. Okada and Q. Xu
Research Reactor Institute, Kyoto University, Kumatori, Osaka 540-0494, Japan

(Received 26 November 2005; accepted 15 February 2006; published online 28 March 2006)

The evaluation of the neutron irradiation induced defects in GaN is studied using a thermally stimulated current (TSC) method with excitation above (below) the energy band gap using ultraviolet (blue, green, red, and infrared) emitting diodes. Annealing at 1000 °C, a broad TSC spectrum for excitation by the ultraviolet light is resolved by five traps, P_1 (ionization energy is 200 meV), P_2 (270 meV), P_3 (380 meV), P_4 (490 meV), and P_5 (595 meV). Infrared illumination shows a remarkable reduction in TSC for the P_2 and P_3 traps, indicating the photoquenching behavior. The possible origins of the observed five traps are discussed. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2190446]

Gallium nitride exhibits unique electrical, optical, and thermal properties, which make it a promising material for optoelectronic and high-power devices. For space-based applications, these devices will have to operate in a radiation environment. In particular, the device operation will be affected by deep level defects. Neutron irradiation is a useful method for the controlled impurity doping[1] by nuclear reactions and homogeneous defect production[1] because of the strong penetration into materials of neutron. Several experiments using deep-level-transient spectroscopy (DLTS),[2–5] isothermal capacitance transient spectroscopy (ICTS),[4] photoemission capacitance transient spectroscopy,[3] capacitance-voltage (*C-V*) measurement,[4] and thermally stimulated current (TSC) spectroscopy[5–7] have been performed to confirm the theoretical calculations of the native defects. In particular, the TSC method is favorable for the high resistive neutron irradiated materials because the TSC technique is applied without the fabrication of Schottky contacts or *p-n* junctions.

In this letter, we report five traps in neutron irradiated GaN by a TSC method with excitation above the energy band gap using an ultraviolet emitting diode. Photoquenching behavior of P_2 and P_3 traps due to the infrared illumination is also reported. Possible candidates of these traps are discussed.

A 5-μm-thick unintentionally doped *n*-type GaN film was grown on a thin (400 Å) AlN buffer layer deposited on a sapphire substrate by metal-organic vapor-phase epitaxy (MOVPE). The unirradiated samples showed the resistivity of 10^5-10^6 Ω cm. The resistivity of the samples irradiated using a neutron irradiation field[8] as described later decreased to 10^4 Ω cm by the 1000 °C annealing. In order to investigate the residual deep defects after the 1000 °C annealing, the TSC method was used in the present study. Al and Au alloy electrodes were fabricated on the four corners of samples of 5×5 cm^2 in area. The bias voltage of 20 V was applied on the electrodes for the TSC measurements using a Van der Pauw technique. Samples were cooled down to 85 K and initialized by illumination for 20 min with an ultraviolet emitting diode [a peak wavelength of $\lambda=365$ nm with a full width at half maximum (FWHM) of 10 nm at 300 K] for the excitation above the energy band gap. In comparison with the illumination above the energy gap, TSC measurements for the excitation below the band gap were also performed using blue ($\lambda=475$ nm, FWHM=25 nm), green (530 nm, 35 nm), red (644 nm, 18 nm), and infrared (950 nm, 50 nm) emitting diodes.

Neutron transmutation doping (NTD) was performed using the center of the core in the Kyoto University Reactor (KUR).[8] Samples were irradiated with fast (fluence equals 1.05×10^{19} cm^{-2}) and thermal neutrons (2.20×10^{19} cm^{-2}) at fluxes of 3.90×10^{13} and 8.15×10^{13} cm^{-2} s^{-1} for each neutron. On the other hand, the neutron irradiation using the Material Controlled Irradiation Facility[9] (MCIF) was performed under 400 °C with the fluence of 1.62×10^{19} cm^{-2} for thermal neutrons and 4.59×10^{18} cm^{-2} for fast ones, and the resistivity of the MCIF samples drastically decreased from 10^5-10^6 to 1–10 Ω cm after the thermal annealing at 1100 °C, showing a significant NTD effect. However, TSC spectra resulting from lattice defects were not observed for the MCIF irradiated samples because of the large amount of electrons and/or holes in the conduction and/or valence bands. Accordingly, the conventional irradiation[8] with the large fluence of fast neutrons decreases NTD efficiency due to the large amount of the introduced defects. Furthermore, after the neutron transmutation reactions,[10] the transmuted atoms are usually not in their original positions but displaced into interstitial positions due to the recoil produced by the γ and β particles in the nuclear reactions.

Figure 1 shows the photoluminescence (PL) spectra for the 1000 °C annealed NTD GaN, together with an unirradiated GaN. PL spectra were taken at 20 K using the 325 nm line of a 35 mW He–Cd laser. Before irradiation, the PL

[a]Electronic mail: kuri@ionbeam.hosei.ac.jp

 © 2006 American Institute of Physics
Downloaded 30 May 2007 to 130.54.110.22. Redistribution subject to AIP license or copyright, see http://apl.aip.org/apl/copyright.jsp

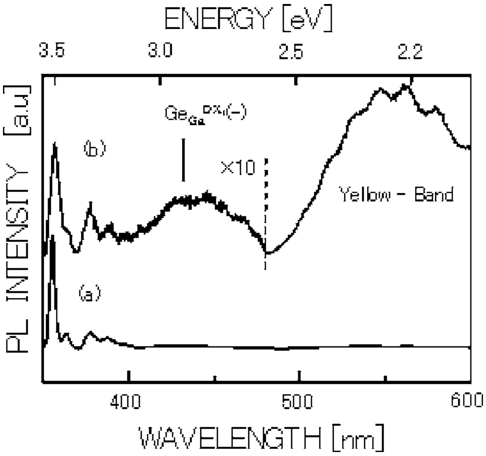

FIG. 1. Photoluminescence spectra at 20 K taken from unirradiated (a) and 1000 °C annealed NTD (b) GaN used for TSC measurements.

spectrum [Fig. 1(a)] is dominated by the donor-bound exciton recombination at 3.490 eV. A broad PL emission with maximum position at ~3.3 eV was also observed, which would be related to a shallow donor-accepter pair.[11] In as-irradiated samples, PL emissions were not observed at all, indicating that the dominant recombination process is a non-radiative one due to the irradiation induced damages. In NTD GaN annealed at 1000 °C for 60 min [Fig. 1(b)], two broad emissions appear at 2.91 eV [blue luminescence (BL)] and 2.25 eV [yellow luminescence (YL)]. Although it is known that the BL observed in undoped GaN is attributed to a V_{Ga} complex such as $V_{Ga}O_N$ and $V_{Ga}H_n$,[11] we propose that the former emission[10] has been related to a DX_1 state[12,13] of the negatively charged Ge_{Ga} [$Ge_{Ga}^{DX}(-)$] introduced by the neutron irradiation. The transformation of the impurity to a *DX* configuration is commonly accompanied by a capture of a second electron by the donor. The YL has also been assigned to a complex defect between the transmuted Ge donor and V_{Ga} acceptor.[10]

Figure 2 shows the TSC spectra for the 1000 °C annealed NTD GaN taken after illumination with the ultraviolet light, together with an unirradiated GaN. The TSC spectra for the samples annealed below 800 °C were not observed at all. In unirradiated samples, four peaks were observed at a peak temperature T_m=200 K (peak P_{01}), 225 K (P_{02}), 250 K (P_{03}), and 280 K (P_{04}). The peak positions of P_{02} and P_{03} are similar to the B_x and C_1 traps observed in unintentionally doped semi-insulating GaN reported by Fang *et al.*,[5] suggesting the same origins. But the thermally stimulated current of unirradiated samples is two orders of magnitude lower than that in NTD GaN. Therefore, the TSC peaks in NTD GaN would additionally result from the neutron irradiation. In neutron irradiated samples, a broad TSC spectrum was observed at a temperature ranging from 100 to 290 K for excitation by the ultraviolet light. A software, the curve fitting toolbox of MATLAB (The Math Works, Inc.), was used to resolve the broad spectrum to optimum Gaussian curves. As a result, the broad spectrum is resolved to five traps, P_1 (T_m=115 K), P_2 (145 K), P_3 (190 K), P_4 (235 K), and P_5 (270 K), as shown in Fig. 2. According to the approximate relationship[14] $E_i \approx kT_m \ln(T_m^4/\beta)$, where E_i is the ionization energy (trap depth), k is Boltzmann's constant, T_m is the TSC peak temperature, and β is the heating rate for the thermal scan (a typical value used here is 0.14 K s^{-1}), the ionization

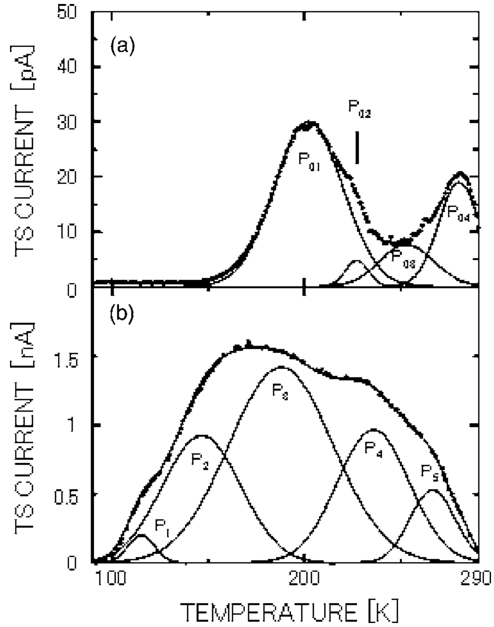

FIG. 2. TSC spectra taken from unirradiated (a) and 1000 °C annealed NTD (b) GaN for the excitation due to ultraviolet light. The spectrum of the NTD GaN is resolved by five traps.

energies, E_i (P_1)=200, E_i (P_2)=270, E_i (P_3)=380, E_i (P_4) =490, and E_i (P_5)=595 meV, for these traps were calculated. E_i values estimated by the present TSC study contain an uncertainty of ±20 meV. Figure 3 shows the TSC spectra in the same GaN sample taken after illumination below the band gap. Thermally stimulated current, peak temperature, and ionization energy for each trap are summarized in Table I for neutron irradiated GaN, together with those of unirradiated GaN. The P_1 trap is responsive to the excitations by blue and green lights, whereas P_1, P_4, and P_5 traps are weakly responsive to the excitation by red and infrared lights. An interesting feature is a remarkable reduction in thermally stimulated current of P_2 and P_3 traps for the 950 nm illumination, suggesting the photoquenching behavior of these traps. Although TSC method usually cannot distinguish the sign of a trap (an electron or a hole trap), possible origins of the observed traps are discussed below.

The P_1 trap is close to a *E* trap (180 meV) observed in 1 MeV electron irradiated GaN.[2] The response to the blue and green light emissions indicates that the electrons fill to P_1 trap via the conduction band from the YL band. Although the origin of the *E* trap has not been clearly identified, this

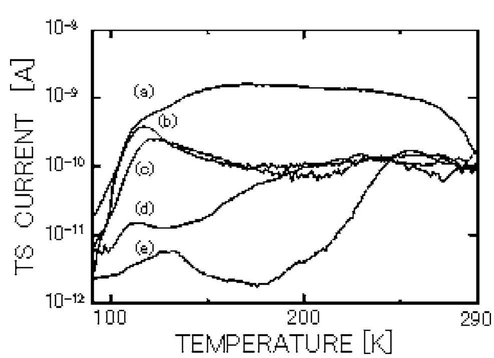

FIG. 3. Logarithm plots of TSC spectra of 1000 °C annealed NTD GaN; (a), (b), (c), (d), and (e) represent the spectra for ultraviolet, blue, green, red, and infrared illuminations, respectively.

Downloaded 30 May 2007 to 130.54.110.22. Redistribution subject to AIP license or copyright, see http://apl.aip.org/apl/copyright.jsp

TABLE I. TSC data for the unirradiated and neutron irradiated GaN.

Sample	Traps	Peaks Current (pA)	Peaks Temperature (K)	Ionization energy (meV)
Unirradited	P_{01}	30	200	400
	P_{02}	5	225	450
	P_{03}	10	250	519
	P_{04}	21	280	630
Neutron irradiated (1000 °C annealed)	P_1	200	115	200
	P_2	900	145	270
	P_3	1400	190	380
	P_4	950	235	490
	P_5	500	270	595

defect center is believed to be a defect complex involving nitrogen vacancy (V_N).[15] On the other hand, a possible origin of the T_5 trap (530 meV) observed in TSC measurements using unirradiated GaN has been proposed to V_N-related defects.[6] We assign the P_1 trap to the defect complex involving V_N at this time, since the trap with energies between 180–200 meV would be effectively produced by neutron irradiation rather than electron irradiation.

The quenching behavior of P_2 and P_3 traps is similar to that of T_3 trap (320±40 meV) (Ref. 7) induced by the excitation using a 690 nm laser diode. In the present study, however, P_2 and P_3 traps do not show a remarkable reduction in thermally stimulated current for the illumination of the 644 nm light-emitting diode (LED). Therefore, the photoquenching behavior observed here may attribute to a different quenching mechanism and/or a different species of defect in comparison with T_3 trap. The activation energies of P_2 and P_3 traps are close to the energy levels of the calculated neutral Ga vacancy.[13] However, since lattice relaxation accompanied by V_{Ga} has been predicted[16] to be negligible, the photoquenching behavior of P_2 and P_3 traps for the infrared illumination may be attributed to the defect complexes involving V_{Ga} rather than the isolated neutral V_{Ga}.

The energy level of the P_4 trap is close to an energy level of *DX*-like center, which introduces a singlet, almost degenerate with the valence band top, and a second singlet about 0.5 eV below the bottom of the conduction band,[12] as observed in photoluminescence measurements. Therefore, this trap is likely to be related to *DX*-like center of Ge_{Ga}.

The energy level of the P_5 trap is close to a defect level labeled D_2 with an activation energy[17] of $E_{D_2}=598\pm20$ meV in 270-keV N^{2+} implanted and annealed MOVPE grown GaN. This defect is coincident with a deep level with an activation energy[4] of $\Delta E2=580\pm17$ meV in unintentionally doped *n*-type metal-organic chemical-vapor deposition (MOCVD) grown GaN. This deep level is believed to be the N_{Ga}.[17] In a theoretical calculation[18] using a model tight-binding Hamiltonian, neutral N_{Ga} in GaN is a deep-hole trap located at about 500 meV below the conduction band edge, while in the computations[12] using *ab initio* molecular dynamics, N_{Ga} introduces a doubly occupied singlet at $E_V+0.4$ eV and an empty doublet at $E_C-0.2$ eV. This empty doublet cannot be occupied even by one electron, due to the large value of the electron-electron repulsion parameter for this level.[12] Although there is a discrepancy between the theoretical calculations, according to the experimental study[17] we assigned the P_5 trap to N_{Ga} at this time.

In summary, we have studied the deep levels in neutron irradiated GaN by TSC measurements. Five traps, P_1–P_5, were observed. P_2 and P_3 traps showed the remarkable reduction of thermally stimulated current by the illumination with infrared light, suggesting the photoquenching behavior of these traps. Most probable candidates of these five traps were also proposed.

Part of this work was carried out under the Visiting Researchers of Kyoto University Research Reactor Institute (KURRI).

[1]M. Satoh, H. Kawahara, K. Kuriyama, T. Kawakubo, K. Yoneda, and I. Kimura, J. Appl. Phys. **63**, 1099 (1988); M. Satoh and K. Kuriyama, Phys. Rev. B **40**, 3473 (1989); M. Satoh, K. Kuriyama, and T. Kawakubo, J. Appl. Phys. **67**, 3542 (1990).
[2]D. C. Look, D. C. Reynolds, J. W. Hemsky, J. R. Sizelove, R. L. Jones, and R. J. Molnar, Phys. Rev. Lett. **79**, 2273 (1997); Z.-Q. Fang, J. W. Hemsky, D. C. Look, and M. P. Mack, Appl. Phys. Lett. **72**, 448 (1998).
[3]W. Götz, N. M. Johnson, H. Amino, and I. Akasaki, Appl. Phys. Lett. **65**, 463 (1994).
[4]P. Hacke, T. Detchprohm, K. Hiramatsu, N. Sawaki, K. Tadatomo, and K. Miyake, J. Appl. Phys. **76**, 304 (1994).
[5]D. C. Look, Z.-Q. Fang, W. Kim, Ö. Aktas, A. Botchkarev, A. Salvador, and H. Morkoç, Appl. Phys. Lett. **68**, 3775 (1996); U. V. Desnica, M. Pavlović, Z.-Q. Fang, and D. C. Look, J. Appl. Phys. **92**, 4126 (2002); Z.-Q. Fang, B. B. Claflin, D. C. Look, T. H. Myers, D. D. Koleske, A. E. Wickenden, and R. L. Henry, Mater. Res. Soc. Symp. Proc. **743**, 749 (2003).
[6]Z. C. Huang, J. C. Chen, and D. Wickende, J. Cryst. Growth **170**, 326 (1997).
[7]H. Witte, A. Krtschil, M. Lisker, E. Schrenk, J. Christen, A. Krost, B. Kuhn, and F. Scholz, Appl. Phys. Lett. **82**, 4083 (2003); H. Witte, E. Schrenk, K. Flügge, A. Krost, J. Christen, B. Kuhn, and F. Sholz, Phys. Rev. B **71**, 125213 (2005).
[8]K. Kuriyama, K. Tomizawa, S. Uematsu, and H. Takahashi, Appl. Phys. Lett. **65**, 746 (1994); K. Kuriyama, J. Takahashi, M. Okada, and M. Uchida, Solid State Commun. **126**, 309 (2003).
[9]T. Yoshiie, Y. Hayashi, S. Yanagita, Q. Xu, Y. Satoh, H. Tsujimoto, T. Kozuka, K. Kamae, K. Mishima, S. Shiroya, K. Kobayashi, M. Utsuro, and Y. Fujita, Nucl. Instrum. Methods Phys. Res. A **498**, 522 (2003).
[10]K. Kuriyama, T. Tokumasu, J. Takahashi, H. Kondo, and M. Okada, Appl. Phys. Lett. **80**, 3328 (2002).
[11]M. A. Reshchikov and H. Morkoc, J. Appl. Phys. **97**, 061301 (2005).
[12]P. Bogusławski, E. L. Briggs, and J. Bernholc, Phys. Rev. B **51**, 17255 (1995); P. Bogusławski and J. Bernholc, *ibid.* **56**, 9496 (1997).
[13]J. Neugebauer and C. G. Van de Walle, Phys. Rev. B **50**, 8067 (1994).
[14]D. C. Look, Semicond. Semimetals **19**, 75 (1983).
[15]Z.-Q. Fang, D. C. Look, W. Kim, and H. Morkoc, MRS Internet J. Nitride Semicond. Res. **5S1**, W11.84 (2000).
[16]I. Gorczyca, A. Svane, and N. E. Christensen, Phys. Rev. B **60**, 8147 (1999).
[17]D. Haase, M. Schmid, W. Kürner, A. Dörnen, V. Härle, F. Scholz, M. Burkard, and H. Schweizer, Appl. Phys. Lett. **69**, 2525 (1996).
[18]D. W. Jenkins and J. D. Dow, Phys. Rev. B **39**, 3317 (1989).

Downloaded 30 May 2007 to 130.54.110.22. Redistribution subject to AIP license or copyright, see http://apl.aip.org/apl/copyright.jsp