JOURNAL OF MATERIALS SCIENCE **39** (2004) 3217–3219

Effect of annealing on neutron-transmutation-doped GaN epilayers grown on sapphire substrates

S. H. PARK, T. W. KANG*
Quantum-Functional Semiconductor Research Center and Department of Physics, Dongguk University, 3-26, Pildong, Chungku, Seoul 100-715, Korea
E-mail: twkang@dgu.ac.kr

T. W. KIM
Advanced Semiconductor Research Center, Division of Electrical and Computer Engineering, Hanyang University, 17 Haengdang-dong, Seongdong-gu, Seoul 133-791, Korea

The growth of the GaN epilayer and its related materials has attracted much attention for both scientific and technological reasons [1–5]. Potential applications of the GaN compound semiconductors in optoelectronic devices operating in the visible to ultraviolet spectral regions have driven extensive and successful efforts to grow GaN epitaxial films of high quality on sapphire substrates by using various technique [6–8]. However, GaN epilayers have inherent problems with control of the uniform carrier density; epilayers with high mobility and with uniform carrier density are very important for achieving highly efficient electronic and optoelectronic devices [9, 10]. Furthermore, since thermal treatment is necessary for the fabrication processes of optoelectronic devices, the role of the thermal annealing process is very important in achieving high-performance devices [11].

The neutron-transmutation-doping (NTD) method is a technique using nuclear reactions between isotopes and thermal neutrons in semiconductors [12]. The method can be used to dope impurities uniformly, to control the impurity concentration accurately, and to activate the carriers by using a relatively low-temperature thermal treatment [13, 14]. When neutrons are introduced into a semiconductor, the carrier concentration, the mobility, and the resistivity of the doped semiconductor, as well as the optical properties, are changed. Even though the behavior of the physical parameters of the GaN epilayer after NTD and annealing have been investigated, the electrical and the optical properties of NTD GaN after annealing have not.

This letter reports on the electrical and the optical properties after NTD and annealing treatment of undoped NTD GaN epilayers grown by using plasma-assisted molecular-beam epitaxy (PAMBE). Van der Pauw Hall effect measurements were performed in order to investigate the electronic parameters of the NTD and annealed GaN epilayers, and photoluminescence (PL) measurements were carried out to investigate the optical properties of the samples.

The undoped GaN epilayers used in this study were grown on sapphire substrates by using a PAMBE system. An inductively coupled radio frequency plasma source provided the reactive nitrogen from nitrogen gas with a purity of 99.9999% while Ga with a purity of 99.9999% was evaporated using a conventional effusion cell. Prior to the GaN film growth, the surfaces of the chemically cleaned substrates were exposed to an activated nitrogen beam for 10 min in order to completely cover them with nitridated layers. The deposition of the GaN active layer on the 300-Å-thick GaN buffer layer, which was grown at 550 °C, was carried out at a substrate temperature of 750 °C, and its growth rate was approximately 0.28 μm/h. The typical thickness of the GaN active layer was 600 nm.

To determine the fluences of the thermal neutrons, we inserted a Au-Al wire into the chamber with the GaN epilayers. A Ge-Li radiation detector (EG & G ORTEC, GMX-25190P) was used to identify the neutron-transmutation doping. The fluences of the thermal neutrons related to the doping of impurities into the GaN epilayer were 1.09×10^{19}, 5.29×10^{18}, and 4.15×10^{17} cm^{-2}. The NTD GaN epilayers were annealed at temperatures between 800 and 1100 °C for 30 min in a nitrogen atmosphere.

To determine the carrier concentration, the mobility, and the resistivity, we performed Hall effect measurements. Van der Pauw Hall-effect measurements were carried out at room temperature in a magnetic field of 0.5 T by using a Keithley 181 nanovoltmeter. Ohmic contacts to the samples for the Hall effect measurements were made by diffusing a small amount of indium through the undoped GaN epilayers at 600 °C in a hydrogen atmosphere for approximately 10 min. The PL measurements were carried out using a 75-cm monochromator equipped with an ultraviolet-sensitive photomultiplier tube. The excitation source was the 3250-Å line of a He-Cd laser, and the power density of the laser was 20 mW. The samples were mounted on a cold finger in a cryostat, and the temperature was controlled at 11 K by using a He displex system.

Fig. 1 shows the resistivity as a function of the annealing temperature for undoped NTD GaN epilayers exposed to various fluences. The resistivity of the GaN epilayer decreases with increasing annealing temperature. In particular, the resistivity of the GaN epilayer

---

*Author to whom all correspondence should be addressed.

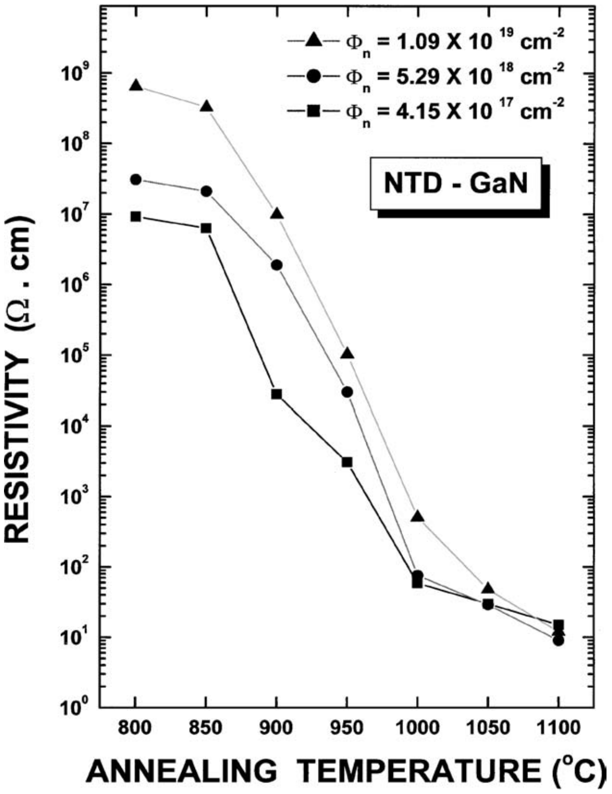

Figure 1 Resistivity as a function of the annealing temperature for undoped NTD GaN epilayers for various fluences.

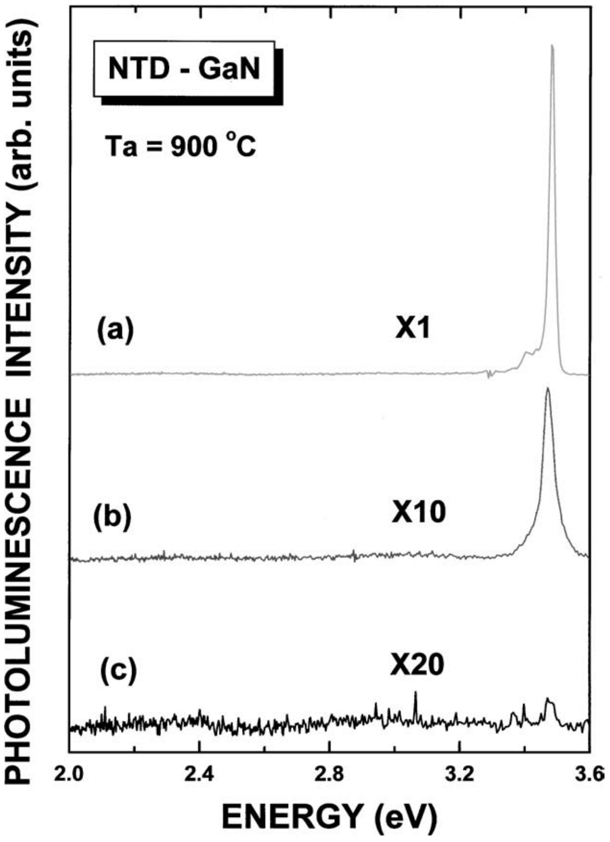

Figure 2 Photoluminescence spectra at 12 K for undoped NTD GaN epilayers annealed at 900 °C for various fluences of (a) 4.15×10^{17}, (b) 5.29×10^{18}, and (c) 1.09×10^{19} cm^{-2}.

dramatically decreases above an annealing temperature of 850 °C, and this behavior originates from the Ga-metal-like behavior resulting from the dissociation of Ga-N bonding.

Fig. 2 shows PL spectra at 12 K for undoped NTD GaN epilayers annealed for 30 min at 900 °C for fluences of (a) 4.15×10^{17} cm^{-2}, (b) 5.46×10^{18} cm^{-2}, and (c) 1.09×10^{19} cm^{-2}. The activated carriers for the undoped NTD GaN epilayers decrease with increasing fluence. The PL spectrum for the undoped GaN epilayer with a doping fluence of 4.45×10^{17} cm^{-2} and annealed at 900 °C shows a free exciton (X) at 3.485 eV while those for the undoped NTD GaN epilayers with doping fluences of 5.46×10^{18} and 1.09×10^{19} cm^{-2} do not show the X peak. A peak at 3.441 eV related to excitons bound to neutral donors (D^{o}, X) and a peak at 3.407 eV attributed to Ge-related transitions are observed in the PL spectrum for the undoped NTD GaN epilayer with a doping fluence of 4.45×10^{17} cm^{-2} and annealed at 900 °C; those peaks are not observed in the PL spectra for the samples with doping fluences of 5.29×10^{18} and 1.09×10^{19} cm^{-2}.

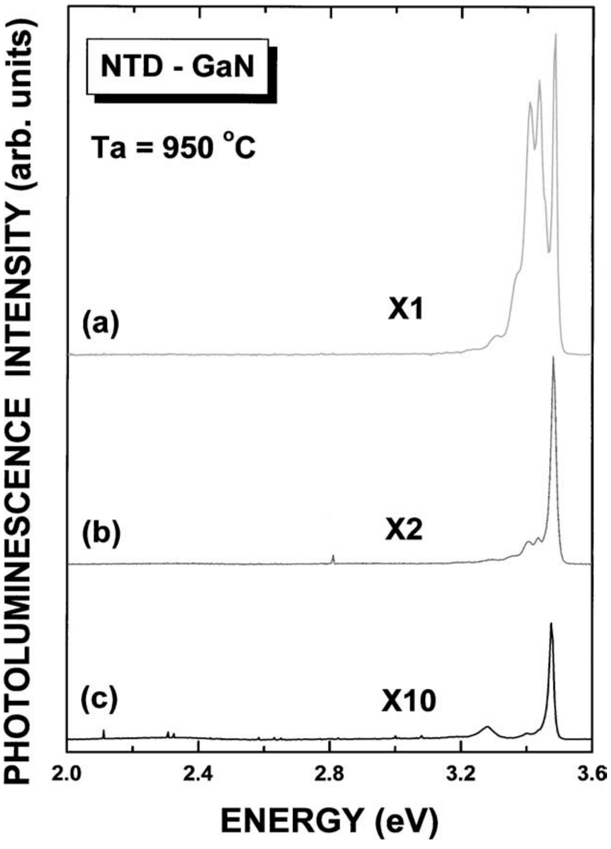

Figure 3 Photoluminescence spectra at 12 K for undoped NTD GaN epilayers annealed at 950 °C for various fluences of (a) 4.15×10^{17}, (b) 5.29×10^{18}, and (c) 1.09×10^{19} cm^{-2}.

Fig. 3 shows PL spectra at 12 K for undoped NTD GaN epilayers annealed for 30 min at 950 °C for fluences of (a) 4.15×10^{17}, (b) 5.46×10^{18} and (c) 1.09×10^{19} cm^{-2}. The PL peak intensity due to the activation energy increases dramatically with decreasing doping fluence, as shown in Fig. 3. The remarkable appearance of the exciton peak related to the Ge atoms, Fig. 3a and b, originates from the neutron transmutation from [Ga^{71} $(n, \gamma)^{72}$] to [Ge$^{72} + \beta$]. However, the PL spectrum for the undoped NTD GaN epilayer with a doping fluence of 4.45×10^{17} cm^{-2}

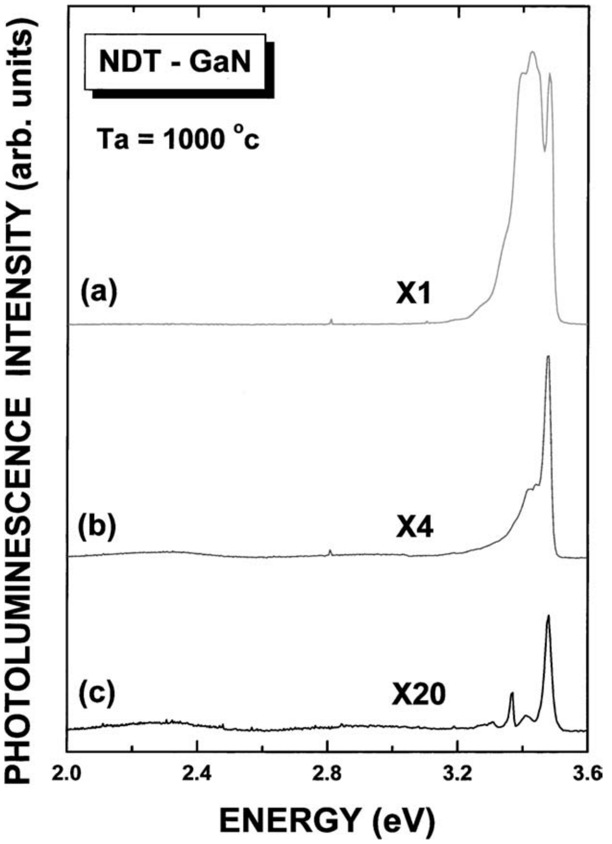

Figure 4 Photoluminescence spectra at 12 K for undoped NTD GaN epilayers annealed at 1000 °C for various fluences of (a) 4.15×10^{17}, (b) 5.29×10^{19}, and (c) 1.09×10^{19} cm^{-2}.

and annealed at 900 °C shows only the yellow band, Fig. 3c.

Fig. 4 shows PL spectra at 12 K for undoped NTD GaN epilayers annealed for 30 min at 1000 °C for fluences of (a) 4.15×10^{17}, (b) 5.29×10^{18}, and (c) 1.09×10^{19} cm^{-2}. The PL peak related to the Ge atom appears more clearly for the undoped NTD GaN epilayers annealed at 1000 °C than it does for the undoped NTD GaN epilayers annealed at 900 and 950 °C. The PL intensities of the yellow bands for undoped NTD GaN epilayers annealed at 1000 °C with doping fluences of 5.46×10^{18} and 1.09×10^{19} cm^{-2} are larger than PL intensity of the yellow band of the undoped NTD GaN epilayer annealed at 950 °C.

The carrier concentrations, mobilities, and resistivities of the undoped NTD GaN epilayers annealed at 1000 and 1050 °C with doping fluences of 4.15×10^{17}, 5.29×10^{18}, and 1.09×10^{19} cm^{-2} are summarized in Table I. The carrier concentration, the mobility, and the resistivity of the undoped NTD GaN epilayers annealed at 1000 °C increase with increasing doping fluence and with increasing annealing temperature.

TABLE I Carrier concentration, mobility, and resistivity, determined from Hall effect measurements at 300 K of the undoped NTD GaN epilayers annealed at 1000 and 1050 °C for various fluence magnitudes

Annealing temperature (°C)	Irradiating fluence (cm^{-2})	Carrier concentration (cm^{-3})	Mobility (cm^2/V·s)	Resistivity (Ω·cm)
1000	4.15×10^{17}	8.09×10^{15}	69	35
1000	5.29×10^{18}	4.24×10^{16}	386	59
1000	1.09×10^{19}	1.89×10^{17}	389	78
1050	1.09×10^{19}	5.35×10^{17}	427	96

In summary, the results of the PL measurements on NTD GaN epilayers showed that the PL intensity related to the Ge atoms increased with increasing annealing temperature. The mobility of the undoped NTD GaN epilayers also increased with increasing annealing temperature. These results indicate that the crystal quality of the NTD GaN is considerably improved after annealing.

Acknowledgment

This work was supported by the Korea Science and Engineering Foundation through the Quantum-functional Semiconductor Research Center at Dongguk University.

References

1. H. AMANO, N. SAWAKI, I. AKASAKI and Y. TOYODA, *Appl. Phys. Lett.* **48** (1986) 353.
2. I. AKASAKI, H. AMANO, Y. KOIDE, K. HIRAMATSU and N. SAWAKI, *J. Cryst. Growth* **98** (1989) 209.
3. S. NAKAMURA, *Jpn. J. Appl. Phys. Part 1* **30** (1991) L1705.
4. B. SANTIC, C. MERZ, U. KAUFMANN, R. NIEBUHR, H. OBLOH and K. BACHEM, *Appl. Phys. Lett.* **71** (1997) 1837.
5. J. FISHER, W. SHAN, J. J. SONG, Y. C. CHANG, R. HORNING and B. GOLDENBERG, *ibid.* **71** (1997) 1981.
6. S. NAKAMURA, *Science* **281** (1998) 956.
7. Y. CHEN, R. SCHNEIDER, S. Y. WANG, R. S. KERN, C. H. CHEN and C. P. KUO, *ibid.* **75** (1999) 2062.
8. P. RAMVALL, Y. AOYAGI, A. KURAMATA, P. HACKE, K. DOMEN and K. HORINO, *ibid.* **76** (2000) 2994.
9. F. REN, M. HONG, S. N. G. CHU, M. A. MARCUS, M. J. SCHURMAN and A. BACA, *ibid.* **73** (1998) 3893.
10. M. S. SHUR, A. D. BYKHOVSKI, R. GASKA, M. A. KHAN and J. W. YANG, *ibid.* **76** (2000) 3298.
11. S. M. SZE, "VLSI Technology" (McGraw-Hill, New York; 1988).
12. H. M. JANUS and O. M. MARMROS, *IEEE Trans. Electron Devices* **23** (1976) 797.
13. J. M. MEESE (ed.), in *Proc. 2nd Internat. Conf. Transmutation Doping in Semiconductors*, Missouri, 1979 (Plenum Press, New York, 1979).
14. J. GULDBUG (ed.), in *Proc. 3rd Internat. Conf. Transmutation Doping of Silicon*, Copenhagen, 1980 (Plenum Press, New York, 1981).

Received 24 June
and accepted 24 November 2003