



Irradiation Induced Defects in a Si-doped GaN Single Crystal by Neutron Irradiation

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Abstract : The local structure of defects in undoped, Si-doped, and neutron irradiated free standing GaN bulk crystals, grown by hydride vapor phase epitaxy, has been investigated by employing electron magnetic resonance(EMR), Raman scattering and cathodoluminescence. The GaN samples were irradiated to a dose of 2×10^{17} neutrons in an atomic reactor at Korea Atomic Energy Research Institute. There was no appreciable change in the Raman spectra for undoped GaN samples before and after neutron irradiation. However, a forbidden transition, $A_1(\text{TO})$ mode, appeared for a neutron irradiated Si-doped GaN crystal. Cathodoluminescence spectrum for the neutron irradiated Si-doped GaN crystal became much broader or was much more broadened than that for the unirradiated one. The observed EMR center with the g value of 1.952 in a neutron irradiated Si-doped GaN may be assigned to a Si-related complex donor.

Keywords : GaN, Neutron irradiation, defect, Electron magnetic resonance, Raman spectroscopy, Cathodoluminescence

INTRODUCTION

The recent achievement in GaN-based blue-light-emitting devices has the potential for a wide range of applications including the blue laser diode for high density optical storage, full-color displays, and ultra violet detectors.¹ However, defects associated with nonradiative or radiative recombination may be detrimental to the device efficiency and the residual defects or dopants may have a crucial role on properties of its opto-electronic

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devices, especially, for high temperature and power applications. For these reasons and possibilities of the usage in extreme environmental conditions, the origins of the defects have been widely studied by the artificial irradiation such as electron²⁻⁴, photon⁵, gamma-ray⁶, proton⁷, and neutron⁸.

In this study, the damages caused by neutron irradiation in GaN single crystals, grown by the hydride vapor phase epitaxy (HVPE) technique,⁹ have been investigated by employing Raman and cathodoluminescence (CL) and electron magnetic resonance (EMR) spectroscopy.

EXPERIMENTALS

The growth techniques and the preparation of the samples were reported in our previous reports.⁸⁻¹⁰ Undoped and Si-doped HVPE-grown free standing GaN single crystals were irradiated to a dose of 2×10^{17} neutrons in an atomic reactor at Korea Atomic Energy Research Institute. The color of transparent GaN samples was changed to dark brown with naked eyes after the irradiation. Raman spectra were obtained using the excitation by the 514.5 nm line of an Ar⁺ ion laser and a double monochromator under the backscattering geometry of $z(x-\bar{z})$ at room temperature. CL measurements were performed at 85 K with a Gatan monoCL attached to a Hitachi S-4700 scanning electron microscope. EMR measurements were carried out by employing a Bruker ESP300 X-band spectrometer with an Oxford ESR900 cryostat for the low temperature operation. The EMR spectra were obtained after the c-axis of the wurtzite crystal was aligned to be parallel to the external magnetic field.

RESULTS and DISCUSSION

Fig. 1 shows the Raman spectra for undoped ((a) and (c)) and Si-doped GaN ((b) and (d)) before ((a) and (b)) and after ((c) and (d)) neutron irradiation. For the unirradiated undoped GaN the well-known allowed Raman modes, E_2 and $A_1(\text{LO})$, are observed in (a).¹¹

This is nearly the same for the irradiated undoped GaN, except for a little broader full-width at half maximum (FWHM) of the peaks, as shown in (c). However, we can observe the appearance of the forbidden mode, $A_1(\text{TO})$, under the backscattering geometry of $z(x-\bar{z})$ in the irradiated Si-doped GaN sample. This observation of the new mode may well be possible when the local structure is deformed from the single crystalline wurtzite structure. Therefore, we suggest that the local environment including the doped Si atom(s) in the crystal is structurally deformed, e. g. tilting off the c-axis of the wurtzite structure, by the neutron irradiation.

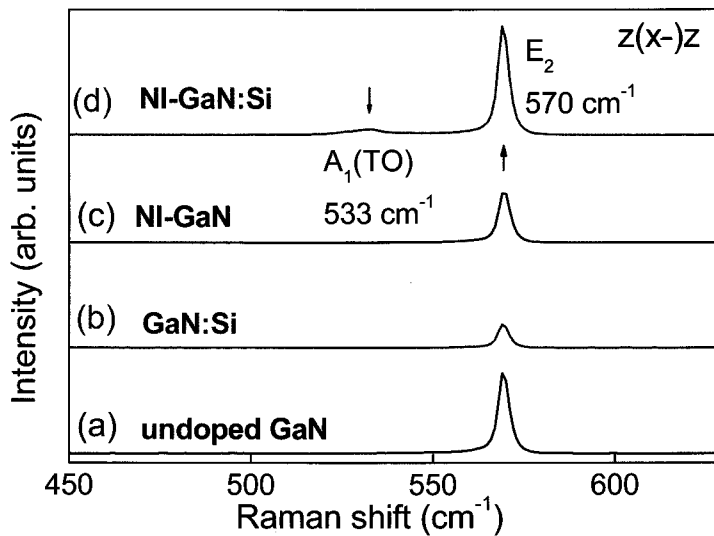


Fig. 1. Raman spectra of GaN samples. NI is the abbreviation of “neutron irradiated”.

Fig. 2 shows the comparison of the CL spectra for the Si-doped GaN crystal before and after the irradiation. After the irradiation a broad peak at the center of about 3.30 eV with the FWHM of 0.5 eV was obtained. This broad band means that the local energy bandgap of the semiconductor fluctuates up to about 4.0 eV. This may happen in the semiconductor with the locally deformed structures such as a tilting, the change of the local strain, the generation of vacancies and/or interstitials, and some others. Therefore, the Raman scattering and CL measurements indicate the presence of structurally deformed defect phases such as a tilted ones containing silicon atoms(s) off the c-axis and locally

strained ones in the neutron irradiated Si-doped GaN.

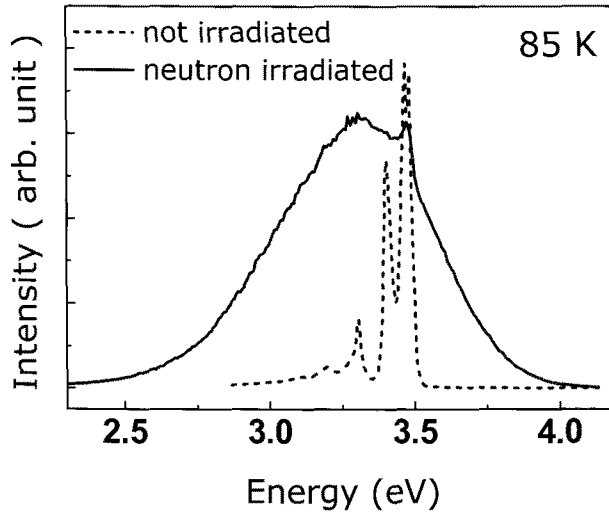


Fig. 2. CL spectra of Si-doped GaN samples.

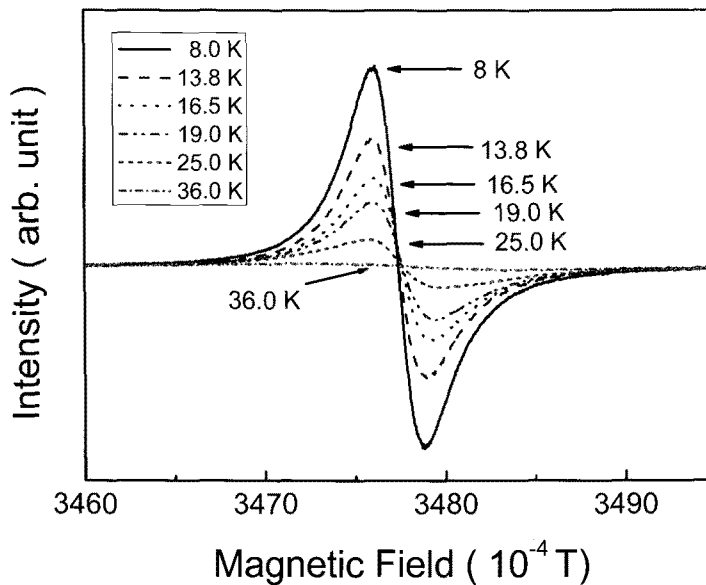


Fig. 3 Temperature dependence of EMR spectra for the neutron irradiated GaN:Si.

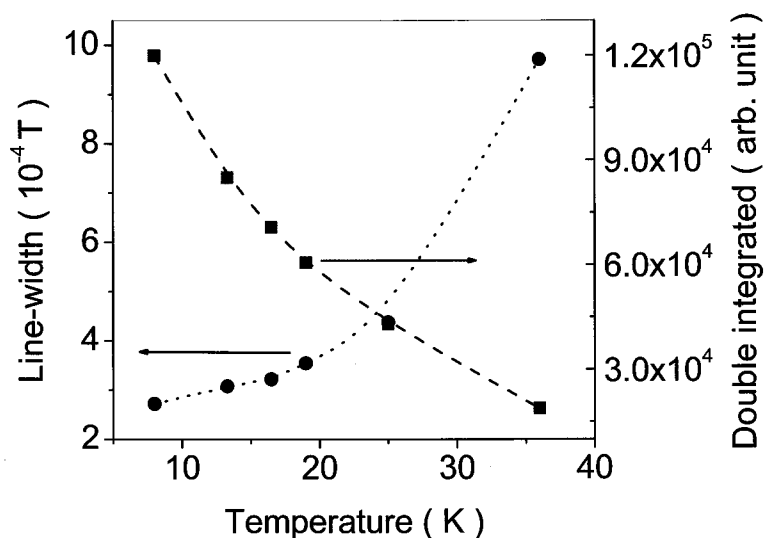


Fig. 4 Temperature dependence of the peak-to-peak linewidths and the double integrated values for the EMR center.

Fig. 3 shows the comparison of the EMR spectra for the neutron irradiated Si-doped GaN crystal at measured temperatures. The observed signal at each temperature shows a symmetric line shape, which means that the paramagnetic center in the irradiated GaN:Si is rather independent from one another. The center also has the g value of 1.952, independent of the temperature. The measured g value is reasonably consistent with the previous report⁸. The defect was introduced after the neutron irradiation because it was recorded very weak before the neutron irradiation. The signal heights of the signals decrease as the temperature increases. The peak-to-peak linewidths of the signals in the second derivative shape broaden from 3×10^{-4} T at 8 K to 10×10^{-4} T at 36 K. Fig. 4 shows the linewidths and the double integrated values of the center according to the measured temperatures. In Fig. 4, the signal intensity of the center, calculated by the double integration, obeys the well-known Curie-Weiss law, which means the defect is a localized spin at least below 36 K. From these results the defect generated by the neutron irradiation in GaN:Si may be originated from a Si-related complex among residual donors.^{8, 12-14}

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