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Thermal behavior of irradiation-induced-deep level in bulk GaN used for fabricating blue light emitting diodes

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Abstract. *Working in an irradiative environment can give rise to defects in GaN-based device, defects induced by electron irradiation in thick free-standing GaN layers grown by halide vapor phase epitaxy were studied by deep level transient spectroscopy. The study will focus on monitoring defects and finding out the nature of these defects. Bulk GaN was irradiated by a 2 MeV electron beam at a fluence of* 5×10^{16} *cm*^{2} *and studied by deep-level transient spectroscopy (DLTS). After irradiation, a broad peak, including at least two traps, was detected. The trap D1 (EC* -0.16 *eV) observed from a broad peak is induced during the annealing process below 550K and completely annealed out at 550K after 10 hours. The annealing process at 550K also forms a new trap D2 (EC ˘0.25 eV). From the isothermal study, the activation energy of the trap D2 in the annihilation process is obtained and has a value of 1.3 ± 0.3 eV. The pre-factor of the annihilation process suggested this process to be related to the free-carrier capture by multi-phonon emission. From the thermal behavior, trap D2 was suggested to be related to gallium vacancy.*

Keywords: GaN; DLTS; defect; irradiation.
Classification numbers: 61.80.Ba; 79.10.na.

1. Introduction

Recent developments within the field of optoelectronics and high-frequency devices have heightened the need for wide bandgap semiconductors such as SiC, GaN, AlN and InN [1–4]. Among them, GaN has been known as the most suitable material for unique applications in light-emitting diodes (LEDs), and lasers in the blue and ultraviolet range due to its wide direct bandgap of 3.4 eV [5–7]. However, a major problem with this kind of application is the efficiency of GaN-based devices, which is strongly dependent upon the quality of the material. Due to a lack of native substrate, GaN is often grown on a foreign substrate having a large lattice mismatch, such as sapphire or SiC. This gives rise to a high concentration of defects, point defects and extend defects, in GaN. The deep levels related to these defects are formed in the bandgap and can behave as traps of charge carriers. Another source of defects is a working environment in which devices suffer

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an irradiation of high-energy particles (electrons, protons or ions). Therefore, understanding the nature and behavior of defects enables us an ability to control defects and thereby improve the GaN-based devices' efficiency.

Regarding irradiation-induced defects, there have been many studies with several important conclusions about the nature and behavior of irradiation-induced defects [8–12]. The most popular technique that has been used in these studies is deep-level transient spectroscopy (DLTS) [13] which gives significant information on defects such as the concentration, capture-cross-section and position in the bandgap. Previous studies reported most of the irradiation-induced deep levels located $0.06 - 0.2$ eV below the conduction band [8–11, 14–17]. These deep levels are suggested to be related to the N-vacancy which is formed when high-energy particles irradiate on GaN.

The annealing process shows that these deep levels can be removed after annealing at a certain temperature. However, there has been no detailed study on the thermal behavior of these deep levels. For this reason, a detailed study of thermal behavior is needed to improve knowledge of electron-irradiation-induced deep levels.

2. Experiment

In this paper, a sample of bulk GaN which was grown on sapphire by Halide Vapor Phase Epitaxy (HVPE) was studied. The horizontal HVPE system which was used for preparing samples is located at Department of Physics, Chemistry and Biology, Linköping University. The samples were grown under an excess ammonia condition at a temperature of 1000˚C and a total pressure of 1 atm. A mixture of H_2 and N_2 with a ratio of 5:2 was used as carrier gas for the precursors NH_3 and GaCl. By using a mixture of H_2 and N_2, we obtain laminar flow and growth conditions that prevent parasitic deposition of GaN in the inlet. The substrate of sapphire was removed to obtain free-standing GaN with a thickness of 300 μm. The Schottky contact of gold with a diameter of 0.6 mm and a thickness of 50 nm was fabricated on the front side by using a thermal evaporator. Silver paint was used for the backside ohmic contact. The sample was then irradiated by the 2 MeV electron beam with a fluence of 5 x 1016 cm-2 at room temperature. This electron beam is expected to damage both of N and Ga sublattices and create intrinsic defects or their complexes with other defects [18]. From the current voltage measurement (IV), the diodes showed a high rectifying characteristic with a leakage current of 3 nA at a reverse bias of 10 V at room temperature. By capacitance voltage measurements (CV), the net donor concentration at room temperature was determined to be approximately $1.7 \times 10^{16}\text{cm}^3$. DLTS measurement was mostly performed within the temperature range between 100 and 200K under the reverse bias of -3V. The filling pulse width of 10 ms and the filling pulse height of 3V were used. The system which was used this report is a homemade setup DLTS located at Department of Physics, Chemistry and Biology, Linköping University..

Isochronal and isothermal annealing methods were used to study the thermal behaviour of irradiation-induced traps. In isochronal annealing, the annealing time is kept constant at increasing temperatures. In isothermal annealing, the temperature is fixed at a certain value, and the annealing time varies. By observing the change of the DLTS peak of each trap, one can get information about the thermal properties of a trap such as the activation energy in the annihilation process and the annihilation rate [19]. In our thermal study, the temperature range of $350 - 650$K was used. The samples were annealed without applying a reverse bias.

3. Result and discussion

Figure 1 shows DLTS spectra of as-grown and irradiated samples, in which a very broad peak named D1 was observed after irradiation. Trap E1, which was detected in the as-grown sample, has a lower amplitude compared to the peak D1. Trap E1 ($Ec - 0.24$ eV) normally appears in HVPE-grown GaN, being reported in previous studies [20, 21]. This trap was suggested to be ascribed to the O_N-related complex near the dislocation sites [22] which shows a dislocation-related electron capture behavior [23]. The broad peak D1 ($E_C - 0.16$ eV) induced by irradiation contains at least two peaks and has been previously reported in Refs. 8,11. In these reports, this trap was probably a nitrogen-vacancy (V_N)-related defect complex such as V_N–N_i, V_N–N_{Ga}–Ga_N–N_i.

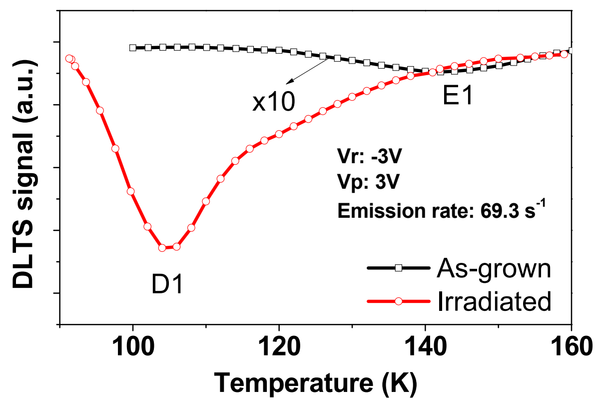

Fig. 1. DLTS spectra of as-grown and irradiated samples. The following DLTS parameters were used: the reverse bias of -3V, the forward bias of 3V and the emission rate of 69.3s^{-1}. The as-grown DLTS spectrum is multiplied by 10 for clarity.

In Fig. 2, we present a series of DLTS spectra after performing the isochronal process in which the sample was annealed in a constant time of 60 min increasing annealing temperature from 350K to 600K. As can be seen, the broad peak amplitude increases when the annealing process was performed at the temperature below 550K. This means that trap D1 can be induced by the annealing process in a temperature range of 350-500K. At the higher temperature (above 550K), annihilation process of the trap D1 happens, possibly due to the replacement of N interstitial (N_i) [24]. After annealing at 600K for 400 min, the trap D1 is annealed out completely and the new trap D2 ($E_c - 0.25$ eV), corresponding to the DLTS peak at ~ 143K, is formed. The trap D2 can be slowly annealed out at a higher temperature.

Figure 3 shows the isothermal study of DLTS spectrum at a fixed temperature of 550K. After annealing the broad peak decreases its amplitude and shifts to the higher temperature. From this figure, one can observe the annihilation process of the trap D1 and the formation of the trap D2. It can be seen that during the first 1 hour the peak decreases rapidly and then the decrease rate is slower. This mean that the annihilation rate of D1 is not constant and depends on its concentration. The rate decreases when the left concentration is small.

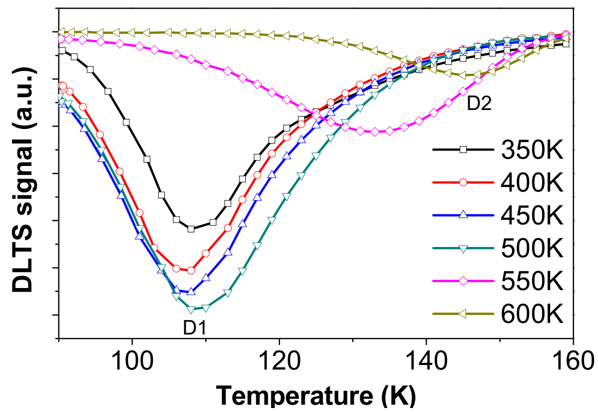

Fig. 2. DLTS spectra collected in isochronal annealing process. The annealing time is fixed at 60 min and the temperature is increased from 350K to 600K with a step of 50K. The following DLTS parameters: $Vr = -3$ V, $Vp = 3$ V, emission rate: 138.6 s^{-1}.

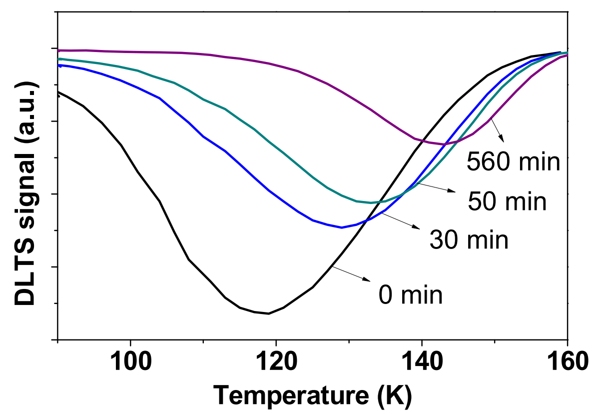

Fig. 3. DLTS spectra in isothermal study at a fixed temperature of 550K.

One important parameter when studying annealing behavior is the activation energy in the annihilation process which can be described as the following equation [19]:

$$R = R_0 \exp\left(-\frac{E_A}{\mathrm{kT}}\right), \tag{1}$$

where R is the annihilation rate, R_o is the pre-factor containing information of annealing process [19], k is the Boltzmann's constant and E_A is the activation energy. The annihilation rate can be determined from observing the reduction of trap concentration during the annealing time, see Fig. 4. Due to strongly overlapping of the trap D1 and trap D2, it is not possible to monitor the change of the peak D1 during annealing process. However, it is possible to study the thermal behavior of the trap D2 by annealing out most of the trap D1 at the temperature of 550K. Fig. 3 shows the change of the trap D2 in logarithmic scale as a function of annealing time at three different temperature: 560, 600 and 650K. The slope of these plots will give information of the annihilation rate corresponding to each temperature. From the determined annihilation rate, one can draw the plot of ln(R) versus $1/kT$ and calculate the slope, which contains the value of the activation energy. From the insert in Fig. 4, the activation energy of the trap D2 in the annealing process is obtained and has a value of (1.3 ± 0.3) eV. The pre-factor R_o of $8.3 \times 10^7 s^{-1}$ is also calculated from the intercept of this plot with a vertical axis. The pre-factor suggests that the annihilation process of the trap D2 is associated with a free-carrier capture by multiphonon emission ($R_o \sim 10^7$ s^{-1}) [19]. By using positron annihilation spectroscopy (PAS), Tuomisto *et al.* found that half of Ga vacancies (V_{Ga}) induced by 2 MeV electron irradiation was annealed out at 600K and a migration barrier of V_{Ga} is about 1.8 ± 0.1 eV [24]. Another report from Saarinen et. al. also shows that the estimated migration barrier of V_{Ga} is 1.5(2) eV and the temperature range for annealing out V_{Ga} is 500-600K [25]. These observations are in good agreement with our results for the trap D2. Therefore, we suggest that trap D2 is possibly related to V_{Ga}.

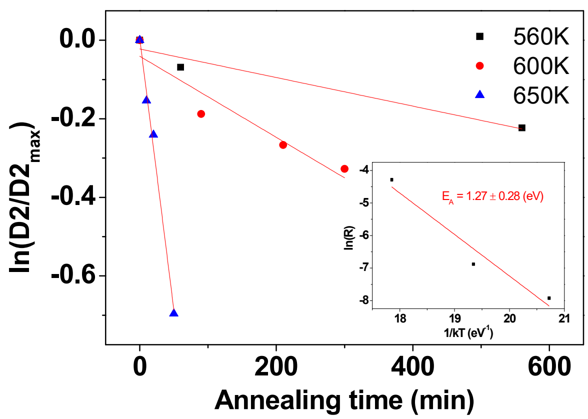

Fig. 4. Normalized peak D2 as a function of the annealing time. The insert shows the Arrhenius plot of the annihilation rate of trap D2.

4. Summary

In summary, we have studied the thermal behavior of the electron-irradiation-induced trap. After irradiation with a 2 MeV electron beam at a fluence of 5×10^{16} cm^2, a broad peak covering from 90 to 160K was found, in which at least two traps exist. The strong peak D1 ($E_C - 0.16$ eV) was dominant after irradiation. The isochronal study showed that the trap D1 is induced during annealing process at low temperature (<550K) and completely annealed out at the temperatures above 550K. The isothermal study of the trap D2 ($E_C - 0.25$ eV) was carried out after removing most of the trap D1, in which the activation energy of the annihilation process is 1.3 ± 0.3 eV. From the pre-factor $R_0 \sim 10^7$ s^{-1}, it is suggested that the annihilation process is related to the free-carrier capture by multi-phonon emission. The trap D2 may be related to V_{Ga}.

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