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Jpn. J. Appl. Phys. Vol. 35 (1996) pp. L 810–L 812
Part 2, No. 7A, 1 July 1996

Analysis of Influence of Alkyl Sources on Deep Levels in GaN by Transient Capacitance Method

Jenn-Fang CHEN, Nie-Chuan CHEN, Wen-Yen HUANG, Wei-I LEE and Ming-Shiann FENG[1]
Department of Electrophysics, National Chiao Tung University, Hsinchu, Taiwan, R.O.C.
[1]*Institude of Materials Science and Engineering, National Chiao Tung University, Hsinchu, Taiwan, R.O.C.*

(Received March 7, 1996; accepted for publication May 21, 1996)

The transient capacitance method was used to analyze GaN samples grown by low-pressure organometallic vapor phase epitaxy (OMVPE) with triethylgallium (TEGa) or trimethylgallium (TMGa) as the alkyl source. Two deep levels at 1.10 and 1.27 eV were observed in the TMGa sample, while a deep level at 0.60 eV was observed in the TEGa sample. Using light illumination, levels deeper than those above were investigated in TEGa and TMGa samples.

KEYWORDS: GaN, deep levels, transient capacitance, OMVPE

The recent development of blue and blue-green light-emitting diodes (LED's) based on GaN and its alloys has attracted much attention.[1–4] However, further advance in III-V nitride technology has been hindered by poor crystalline quality and high n-type background carrier concentration, resulting from native defects believed to be mainly nitrogen vacancies.[3,5] In previous work,[6] two deep levels with activation energies of 0.14 and 0.49 eV were observed by deep-level transient capacitance spectroscopy (DLTS) in GaN grown by organometallic vapor phase epitaxy (OMVPE). Another level appeared at temperatures higher than 450 K. However, due to the limited range of operating temperature of the DLTS stage, the parameters of this level could not be obtained accurately.

In the present work, we continue this study by using a transient capacitance method to characterize the very deep levels in samples grown by a conventional, horizontally configured, low-pressure OMVPE method with triethylgallium (TEGa) or trimethylgallium (TMGa) as the Ga precursors. NH_3 was used as the nitrogen source. The TEGa sample was undoped, while the TMGa sample was doped with Si using SiH_4. GaN films were grown on sapphire at 1050°C affer the growth of a 500-Å-thick GaN buffer layer at 525°C. The GaN films used in this study were about 2 μm thick. Schottky and ohmic contacts were fabricated by evaporating Au and Al, respectively. The reverse saturation current of the Schottky diodes was about 3.8×10^{-9} A/cm^2. Room-temperature capacitance-voltage measurements yielded essentially uniform distributions of free electrons with concentrations of 2.4×10^{17} cm^{-3} and 9.0×10^{16} cm^{-3} for the TEGa and TMGa samples, respectively. Transient capacitance measurements were carried out for five wafers; three TEGa and two TMGa samples. About 5 diodes were measured at different locations in each wafer (about 7×7 mm^2). A deep level was observed in some diodes but not in others. This deep level was attributed to a surface or an unknown effect and data related to this level were not used. Only the deep levels which were always observed in all diodes are discussed in this work.

The transient capacitance was measured using a HP4194 impedance analyzer. A reverse bias of 2 V was applied to remove electrons from the deep levels in the depletion region. The frequency of the capacitance meter was set at 50 kHz with a rms oscillation level of 0.2 V. When the reverse bias was applied, the capacitance dropped immediately and then increased exponentially to a constant value in the case of the TEGa sample, indicating the existence of a single electron level, as shown in Fig. 1. The leakage current was less than 1 μA at the highest temperature. The time constants are plotted as solid circles in the Arrhenius plot in Fig. 2. When these points were fitted, it was found that the fitting line intersected the two points observed by DLTS,[6] indicating that they are related to the same deep level. The activation energy of this level was accurately determined to be 0.60 eV. A concentration of 8.8×10^{15} cm^{-3} and an electron capture cross section of 2.4×10^{-20} cm^2 were obtained for this level. Hacke *et al.*[5] reported a deep level at 0.58 eV in a GaN sample grown by hydride vapor phase epitaxy (HVPE). Although this activation energy is similar to that obtained in our experiment, the deep levels are not the same judging from the large difference in the capture cross sections. The exact origin of this level is not yet clear and is the subject of further investigation.

Similar transient capacitance spectra, but with time constants much longer than those for the level at 0.60 eV,

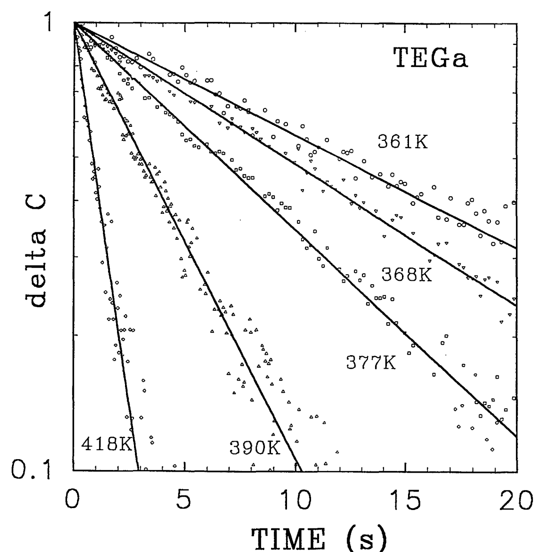

Fig. 1. The transient capacitance ΔC versus time at different temperatures for a TEGa sample.

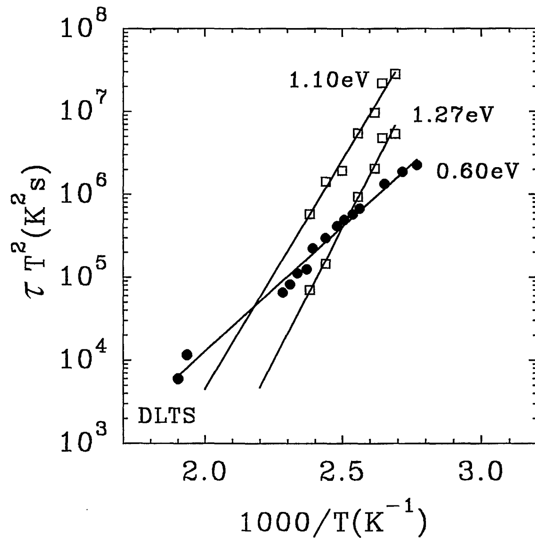

Fig. 2. Arrehenius plot for TEGa and TMGa samples. Two deep levels at 1.10 and 1.27 eV were observed for the TMGa sample, while a deep level at 0.60 eV was observed for the TEGa sample. The two solid circles on the left were obtained previously by deep-level transient spectroscopy with an emission rate window of $23.26\,s^{-1}$ and a pulse width of 100 ms.

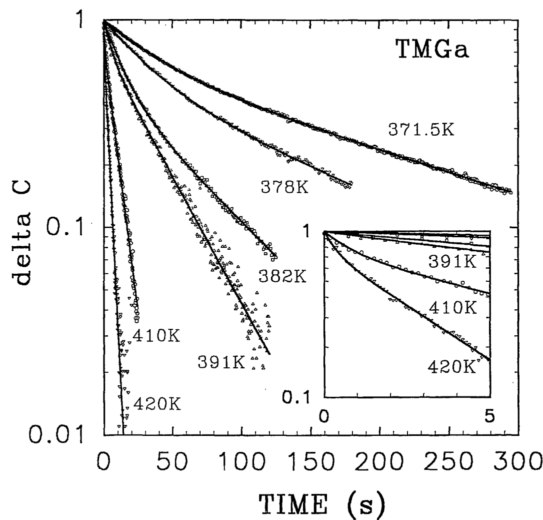

Fig. 3. The transient capacitance ΔC versus time at different temperatures for a TMGa sample. These capacitance spectra can be fitted by combining two time constants, as shown by the solid lines. Spectra for 420 and 410 K from 0 to 5 s are shown in the inset to demonstrate the effects of two time constants.

were observed for the TMGa samples, as shown in Fig. 3. Detailed examination revealed that these spectra could not be fitted using a single time constant, but could be fitted very well by combining two time constants. For example, the capacitance spectrum for 371.5 K can be fitted very well by combining two exponential functions with time constants of 203.9 s and 38.8 s. The capacitance spectra for all other temperatures were fitted in this way and two sets of time constants were obtained as shown by the open rectangles in Fig. 2. Two deep levels with activation energies of 1.10 eV ($n_t = 5.9 \times 10^{14}\,cm^{-3}$, $\sigma_n = 7.4 \times 10^{-15}\,cm^2$) and 1.27 eV ($n_t = 3.5 \times 10^{14}\,cm^{-3}$, $\sigma_n = 8.4 \times 10^{-12}\,cm^2$) were obtained for the TMGa samples. These results are summarized in Table I.

Table I. Summary of GaN deep-level data.

Sample No.	E_t (eV)	σ (cm^2)	N_t (cm^{-3})
TMGa	1.10	7.4×10^{-15}	5.9×10^{14}
	1.27	8.4×10^{-12}	3.5×10^{14}
TEGa	0.60	2.4×10^{-20}	8.8×10^{15}

Oxygen in GaN is thought to be responsible for the broad photoluminescence emission band near 560 nm[7] and the broad cathodoluminescence band near 520 nm.[8] These wavelengths are very similar to those of the deep levels at 1.10 and 1.27 eV observed in the TMGa samples. In addition, the deep level associated with oxygen impurities substituting on Ga sites was calculated by Jenkins and Dow[9] to be 1.27 eV. Therefore, the deep levels at 1.10 and 1.27 eV observed in the TMGa samples may be due to oxygen impurities and the broad optical band observed at room temperature may be an effect due to the combination of these two levels. Since the capture cross sections for these two levels are different, further investigation of the optical spectra obtained at low temperatures is necessary to determine whether the two peaks can be resolved.

One possible reason for the difference in the deep levels of the TMGa and TEGa samples in the present study may be the difference in growth conditions, such as the growth temperature and the doping. However, the difference could also be due to impurities, such as oxygen or carbon in the reaction sources. It is known that during OMVPE growth, TMGa pyrolyzes to produce highly reactive CH_3 radicals, which leads to a high degree of carbon contamination. We doped Si to the TMGa samples because a conductive n-type layer cannot always be obtained. Sometimes the TMGa samples showed higher resistivity than the TEGa samples. This indicates that there may be carbon contamination in the TMGa samples. However, carbon usually acts as a shallow dopant in III-V compounds and should affect the properties of the shallow levels rather than those of the deep levels. However, at the present, it is not clear whether carbon could affect the deep levels in GaN. On the other hand, oxygen is thought to introduce deep levels in most III-V compounds. Correlation between the degree of oxygen contamination and the broad optical emission near 560 nm has been illustrated.[7] Correlation between the oxygen and the deep levels is under investigation using the transient capacitance method.

Materials with large bandgaps such as GaN are thought to have levels deeper than those previously found. To determine whether this is true, samples were illuminated using a tungstern light during the transient capacitance measurements. The concentration of the deep levels involved in the photoexcitation processes was indicated by the difference between the capacitances before and after the illumination.

Figure 4 shows the capacitance spectrum at 362 K for a TEGa sample obtained using a dark-light-dark sequence. First, a voltage of -2 V was applied at $t = 0$ s, and the capacitance increased exponentially to a constant value

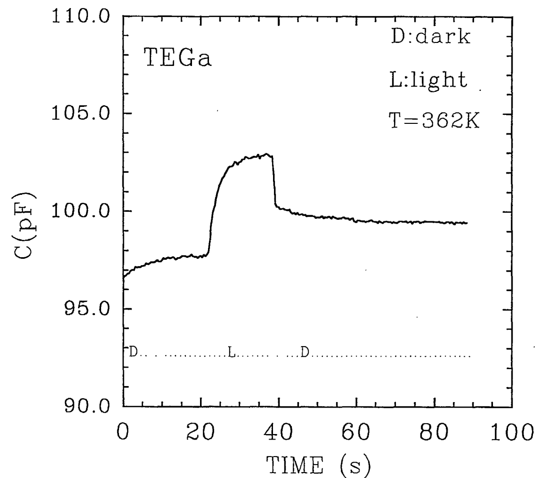

Fig. 4. Variation of the capacitance of a TEGa-grown GaN Schottky diode with a dark-light-dark illumination sequence.

of 97.8 pF with a time constant of 15 s, corresponding to the emission rate of the level at 0.60 eV determined previously. At this point, the electrons captured in levels with time constants less than or equal to 15 s had already been emitted to the conduction band. Then, light was illuminated at $t = 22$ s and the capacitance increased to about 103 pF. When the light was switched off (at $t = 38$ s), the capacitance did not return to the constant value of 97.8 pF obtained before illumination, but reached a constant value of 98.8 pF. A difference of 1 pF ($\Delta C/C = 1\%$) was observed, which corresponded to a carrier concentration of 4.8×10^{15} cm^{-3}. There are several complex processes which might occur due to the illumination of light. However, once the light was switched off the major processes likely to occur in the depletion region were the emission of electrons and holes from deep levels. If the time constants for the emission of holes are much larger than the measuring time, the capacitance would not be observed to return to its value before illumination. Another process which might contribute to the difference in the capacitance is the existance of very deep levels with time constants longer than 15 s, some of the electrons captured by which were excited to the conduction band upon illumination. Both processes involve deep levels with time constants much longer than 15 s. Therefore, the concentration obtained in this way indicates that there are at least that concentration of deep levels in GaN which have not yet been observed.

A similar experiment was performed on the TMGa sample. When a steady state for the emission of electrons from the levels at 1.10 and 1.27 eV was reached at $T = 371$ K, light of the same intensity was illuminated and a difference of 3 pF ($\Delta C/C = 12\%$) was observed in the capacitance of the TMGa sample before and after illumination. This corresponds to a concentration of deep levels of at least 2.2×10^{16} cm^{-3} in the TMGa sample, which has not yet been observed. A stage which can be operated at higher temperatures is under construction so that these very deep levels can be fully characterized.

This work was supported in part by the National Science Council of the R.O.C.

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