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Carrier transport in GaN single crystals and radiation detectors investigated by thermally stimulated spectroscopy

V. KAŽUKAUSKAS* and J.-V. VAITKUS

Semiconductor Physics Department and Institute of Materials Science and Applied Research
Vilnius University, 9 Saulėtekio Ave., LT-2040 Vilnius, Lithuania

We investigated single crystals of GaN and thin film GaN radiation detectors by thermally stimulated currents (TSCs) and thermally stimulated depolarization (TSD) methods in order to characterize carrier transport properties as influenced by material defect structure. In thick GaN, no expressed structure of the TSC spectra was observed in the temperature range from 100 K up to 350 K that could be characteristic for thermal carrier generation from trap levels. The experimental facts imply that TSC spectra might be caused not by carrier generation from traps, but it could be due to thermal mobility changes. Therefore we had applied the numerical analysis by taking into account carrier scattering by ionized impurities and by phonons. It was found that mobility limited by ionized impurities varies as ~ $T^{2.8}$ and lattice scattering causes the dependence ~ $T^{-3.5}$. The highest mobility values were up to 1550 cm^2/Vs at 148–153 K. Such high values indicate relatively good quality of the single GaN thick crystals. In high resistivity GaN detectors irradiated by high doses of high-energy neutrons and X-rays current, the instabilities were observed that could be caused by the change of carrier drift paths in a highly disordered mater. A model of carrier percolation transport is presented.

Keywords: GaN, carrier transport and capture, mobility, carrier scattering.

1. Introduction

Over the past decade, GaN and related nitrides have been in the focus of intense research [1–3]. Being direct, large band gap materials, they lend themselves to a variety of electronic and optoelectronic applications. The advantages associated with a large band gap include higher band breakdown voltages, ability to sustain large electric fields, lower noise generation and high temperature operation. Therefore gallium nitride is one of the most promising materials for high-temperature, high frequency, and radiation hard applications. GaN-based ultraviolet visible-blind photodetectors, blue, violet and green light emitting diodes, laser diodes and transistors of different types have been demonstrated. Nitrides are ideal for applications in displays and high density data storage.

On the other hand, compared with their technological applications, partly due to poor material quality, fundamental research in nitrides, particularly in electronic transport, appears to be far behind. Technologically, a defect density is still very high even in the best samples. Therefore, apart from technological advances in material production, further progress in the development, design and optimization of GaN-based devices necessarily requires a detailed investigation of transport and defect properties by different methods. GaN-based materials have large piezoelectric constants; therefore large piezoelectric fields can be induced in strained layer structures. Spontaneous polarization is present in these wurtzite semiconductors and thus large built in electrostatic fields are induced in layered nitride structures, even in the absence of any strain. Therefore it was our task to investigate single GaN crystals and thin film GaN radiation detectors by thermally stimulated currents and thermally stimulated depolarization methods in order to characterize carrier transport properties as influenced by material defect structure.

2. Samples and experiment

We investigated thick single GaN crystals and thin semi-insulating (SI) GaN structures by means of thermally stimulated currents (TSC), thermally stimulated depolarization (TSD), and current-voltage (IV) techniques. Single high resistivity GaN platelets with thickness of up to ~ 500 µm were from the Lumilog (France). They were fabricated by HVPE technique on sapphire and separated from it. Their carrier concentration ranged from 5×10^{14} cm^{-3} up to 5.5×10^{15} cm^{-3} and mobility was 380–920 cm^2/Vs. Thin epitaxial SI-GaN detector structures were grown by MOCVD technology on a sapphire substrate. They consisted of a 2.5-µm thick SI-GaN upper layer on an *n*-GaN buffer layer. Detectors were fabricated using 1.5-mm diameter evaporated gold Schottky contacts. They were irradiated with 100 keV reactor neutrons (5×10^{14} cm^{-2}) and 10 keV X-rays (600 Mrad).

*e-mail: vaidotas.kazukauskas@ff.vu.lt

Principles of the classical TSC technique can be found in, e.g. Refs. 4 and 5. For the TSC measurements, the samples were cooled down to liquid nitrogen temperature in the dark without applied voltage. Later, they were excited for about 5 min. Two different excitation types were used. One of them was a white light excitation from a 100 W halogen lamp without applied voltage. We also used excitation by an applied voltage in both forward and reverse directions in the dark. In that case, by heating thermally stimulated depolarization is measured. After the excitation the samples were left to relax to the equilibrium state in the dark, and then they were heated at a constant rate of 10 K/min. To reveal the possible influence of the overlapping maxima that could be caused by several competitive processes, we applied the fractional heating technique. This advanced modification is a powerful tool for the discrimination of the overlapping thermally stimulated processes in materials with many levels in the band gap (e.g. Ref. 3). The multiple heating enables the sequential emptying of the initially filled shallower levels thus giving the information about the deeper ones in the repetitive temperature scans. The polarization effects were investigated by TSD.

3. Results and discussion

We investigated carrier transport as influenced by charge trapping and defect structure screening after sample excitation by light or applied voltage. In Fig. 1, TSC curves of the single crystal GaN platelets are presented. It is noticeable that no expressed structure of the TSC spectra was observed in the temperature range from 100 K up to 350 K that could be characteristic for thermal carrier generation from trap levels. Instead only one wide and plane maximum used to appear in single GaN crystals. Moreover, TSCs in log $I \sim 1/T$ scaling were not linear, as it could be expected by taking into account a classical thermal generation model from trap levels. In order to reveal the possible structure of this maximum we applied the fractional heating technique that is a powerful tool for the discrimination of overlapping maxima. Nevertheless, by repetitive thermal scans the shape of the TSC maximum did not change, as it is demonstrated in Fig. 2. Furthermore, application of the TSD method resulted in a similar dependence of the depolarization current on the temperature as it is demonstrated in Fig. 3.

These facts imply that most probably measured dependencies were caused not by thermal carrier generation, but rather by their mobility variation. A similar conclusion that illumination even at 77 K has very little effect on the electron density in GaN but can lead to a noticeable persistent increase in the Hall mobility was made in Ref. 5. The induced persistent photoconductivity (PPC) effect was, there-

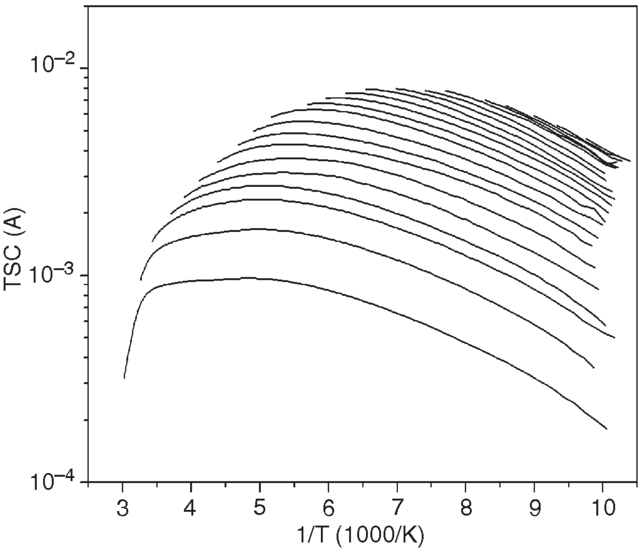

Fig. 2. Thermally stimulated current spectra obtained by repetitive heating of the single GaN crystal.

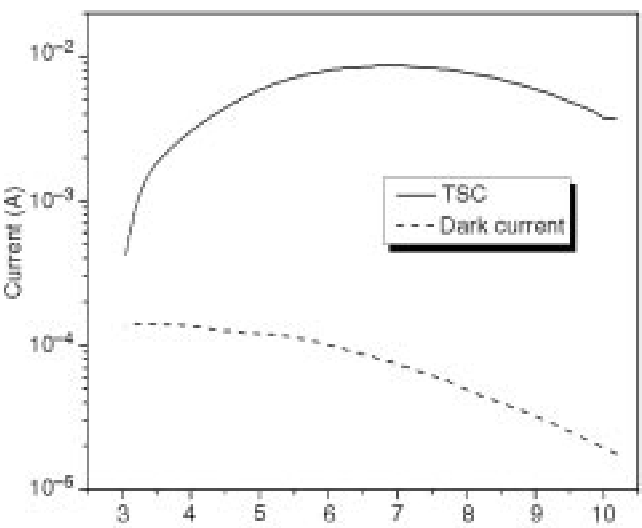

Fig. 1. Thermally stimulated and dark current spectra in a single GaN crystal.

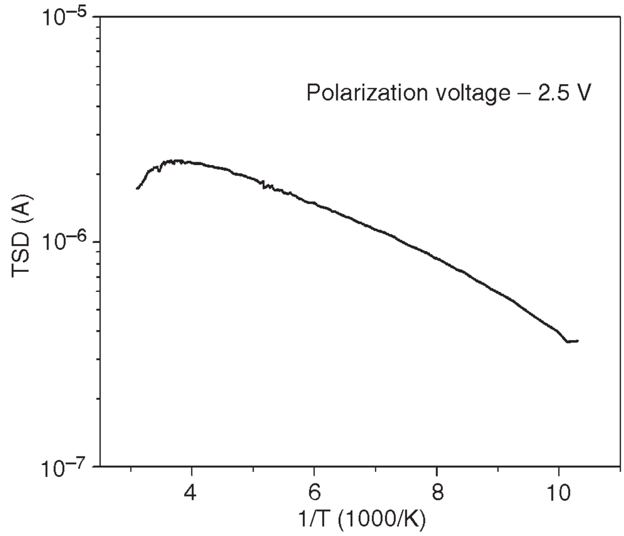

Fig. 3. Thermally stimulated depolarization curve. The sample was polarized at low temperature with 2.5 V voltage under the white light illumination.

fore, attributed to the Hall mobility through intrinsic electrically active defects. The authors also report the very strong ionized impurity scattering that limits the Hall mobility of GaN. Very similar mobility variation with temperature was obtained in Ref. 6. In this paper, the authors empirically fit experimental mobility data presented in Ref. 7. The presented fitting takes into account mobility changes caused by ionized impurity scattering at low temperatures and phonon scattering at higher temperatures.

The first step of the modelling technique consists in an adequate approximation of the doping level dependence of the mobility at the given temperature [7] on the basis of the well known Caughey–Thomas approximation [8]

$$\mu_i(N) = \mu_{\min,i} + \frac{\mu_{\max,i} - \mu_{\min,i}}{1 + \left(\frac{N}{N_{g,i}}\right)^{\gamma_i}}, \tag{1}$$

where $i = n, p$ for electrons and holes, respectively. The model parameters $\mu_{\max,i}$, $\mu_{\min,i}$, $N_{g,i}$, and γ_i depend on the type of semiconductor material, and N is the doping concentration.

On the other hand, at low doping level and room temperature phonon scattering is the main mechanism of scattering. In this case $\mu_{\max,i} = \mu_{I,i}$. The temperature dependencies of mobilities limited respectively by phonon and ionized impurity scattering were supposed to follow the known rules [9]

$$\mu_L = \mu_L(T_0)(T/T_0)^{-\alpha}, \tag{2}$$

$$\mu_I = \mu_I(T_0)(T/T_0)^{-\beta}. \tag{3}$$

Here the coefficient α describes the characteristic power-law mobility temperature variation caused by the lattice scattering, meanwhile the coefficient β stands for ionized impurity scattering. Nevertheless the values of α and β in real semiconductors usually are not equal to that evaluated theoretically, and therefore they have to be found experimentally.

The following empirical expression was derived in Ref. 7 to model mobility, depending on the concentration of scattering centres and temperature

$$\mu_i = (N,T) = \mu_{\max,i}(T_0)\frac{B_i(N)\left(\frac{T}{T_0}\right)^{\beta_i}}{1 + B_i(N)\left(\frac{T}{T_0}\right)^{\alpha_i+\beta_i}}, \tag{4}$$

where

$$B_i(N) = \left[\frac{\mu_{\min,i} + \mu_{\max,i}\left(\frac{N_{g,i}}{N}\right)^{\gamma_i}}{\mu_{\max,i} - \mu_{\min,i}}\right]_{T=T_0}. \tag{5}$$

We had applied the above approximation given by Eqs. (4) and (5), describing the mobility dependence on temperature, and an excellent fit was obtained as it is demonstrated in Fig. 4. By fitting the experimental curves we supposed that carrier concentration remains unchanged and equals 1×10^{15} cm^{-3} as it was indicated by the producer. Analyzing the set of experimental data we were able to find the values of parameters $\mu_{\max;i}$, $\mu_{\min;i}$, $N_{g;i}$, and γ_i providing the best approximation of the data reported for electron and hole mobility in wurtzite GaN. These values are as follows: $\mu_{\max,i}$ = 1600 cm^2/Vs, $\mu_{\min,i}$ = 160 cm^2/Vs, $N_{g,i}$ = 2.70×10^{17} cm^{-3}, γ_i = 1.3, a_i = 3.5, b_i = 2.85, N = 1.20×10^{17} cm^{-3}, T_0 = 185 K. This means that in our single crystal GaN sample mobility limited by ionized impurities varies as ~ $T^{2.8}$ and lattice scattering causes the dependence ~ $T^{-3.5}$. The highest mobility values were up to 1550 cm^2/Vs at 148–153 K. Such high values indicate relatively good quality of the thick crystals. Nevertheless, as it could be seen from Figs. 1 and 2, they were reached only in pre-excited samples. After thermal cycling, the experimental maximum used to decrease and to move towards higher temperatures.

Qualitatively similar conductivity dependencies were obtained also in the thin film radiation detectors before irradiation. Usually such behaviour can be observed if ionised and/or neutral defect scattering is intensifying, e.g. Ref. 10. This can be realised if in pre-excited crystal the effective number of impurities acting as scatterers is reduced. On the other hand, namely in excited semiconductor the number of ionised impurities, from which electrons are generated, should be higher. This seeming contradiction is removed if one takes into account the real structure of many semiconductors in which not only point scattering centres appear, but more complex defects are formed. Usually, in many cases due to doping inhomogeneities, which might be due to the minor temperature variation during the growth process, defects become distributed inhomogeneously too. This is a known problem in emerging materials, technology of which is not developed well enough. Some time ago this was a well known problem in semiinsulating GaAs. The material inhomogeneity problem in GaAs was solved only after more than one decade of intense investigations. In GaN layers inhomogeneities might be also formed by the known grained structure with different grain size and density. Usually, appearance of such spatial inhomogeneities leads to the modulation of the band gap edges and results in the potential relief of the band gap, that plays effective role in charge transport [e.g., Refs. 11, 12, and 13]. In GaN, slow conductivity relaxation effects were also observed many times, that directly evidence effect of inhomogeneities in transport phenomena. Different carrier transport mechanisms were analysed in GaN in, e.g., Refs. 14–16. In Ref. 15, a deep-centre hopping conduction was analysed. The authors of Ref. 16 took into account dislocation scattering in n-type GaN with relatively high dislocation density. Impact of grain boundaries resulting in an exponential temperature dependence of carrier

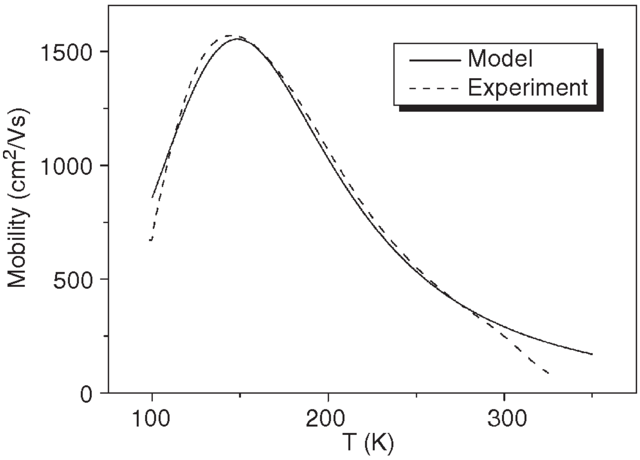

Fig. 4. Numerical fitting (solid curve) of the experimental mobility dependence (dashed curve).

mobility and concentration was revealed in Ref. 17. In our samples carrier and defect concentration was much lower than in previous analyses, so grain boundaries should appear as inaccessible regions for the carriers. Therefore we propose a model in which effect of potential fluctuations due to crystal inhomogeneities is involved, causing percolation transport in GaN. At low temperatures, the carriers generated by light fill potential wells, screening impurities and smoothing the potential relief. By heating, thermally released carriers recombine causing defect inhomogeneities to act as scatterers again. Similar effect of the recapture of excited electrons into illumination-neutralized defects responsible for the persistent mobility in undoped GaN was observed [6]. The model also explains why nearly no mobility dependence on temperature was observed in GaN samples irradiated by high doses of high-energy neutrons (5×10^{14} cm^{-2}) and by 600 Mrad of X-Rays, though their excitation resulted in current increase by up to one order of magnitude (see Figs. 4 and 5). This is because of the big number of defects, i.e., inhomogeneities, introduced by

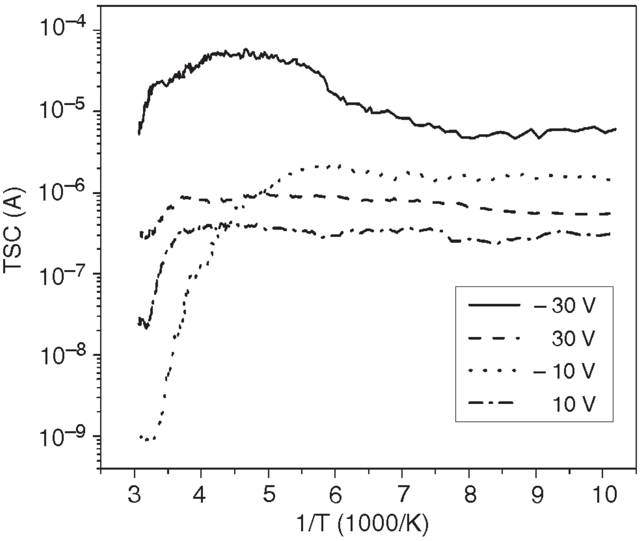

Fig. 5. TSC spectra in GaN detector, irradiated by neutrons.

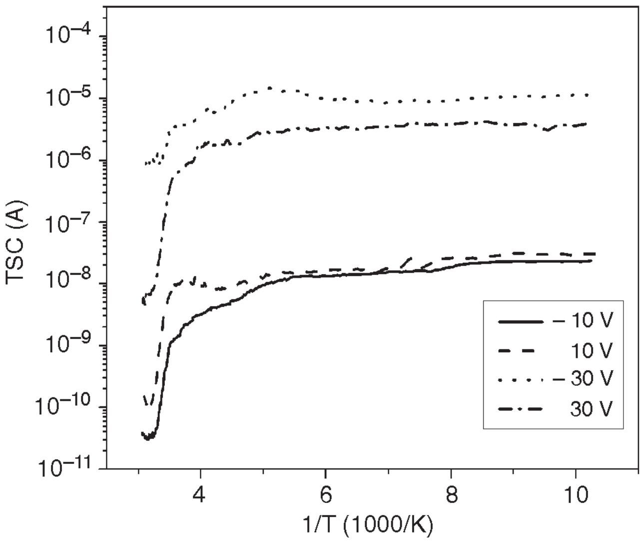

Fig. 6. TSC spectra in GaN detector, irradiated by X-rays.

high-energy irradiation. Furthermore, in these detectors current instabilities were observed that could be caused by the random change of carrier drift paths in a highly disordered mater.

4. Summary and conclusions

Single crystals of GaN and thin film GaN radiation detectors were investigated by means of the thermally stimulated current and thermally stimulated depolarization techniques in order to identify carrier transport peculiarities resulting from prevailing defects. Experimentally, in single high resistivity GaN platelets with thickness of up to ~ 500 µm, no expressed structure of the TSC spectra was observed in the temperature range from 100 K up to 350 K, that could be characteristic for thermal carrier generation from trap levels. Instead, only one wide and plane maximum used to appear in single GaN crystals. Moreover, TSCs in log $I \sim 1/T$ scaling were not linear. In order to reveal the possible structure of this maximum we applied the fractional heating technique as a powerful tool for the discrimination of overlapping maxima. Nevertheless by repetitive thermal scans the shape of the TSC maximum did not change. These facts imply that most probably TSCs were caused not by thermal carrier generation, but rather by their mobility variation. Therefore the numerical analysis was applied by taking into account carrier scattering by ionized impurities and by phonons. It was found that mobility limited by ionized impurities varies as ~ $T^{2.8}$ and lattice scattering causes the dependence ~ $T^{-3.5}$. The highest mobility values were up to 1550 cm^2/Vs at 148–153 K, indicating relatively good quality of the single GaN thick crystals. Nevertheless they were reached only in pre-excited samples. After thermal cycling, the experimental maximum decreased and moved towards higher temperatures. Usually, such behaviour can be observed if impurity scattering is intensifying. In high resistivity GaN detectors irradiated by high doses of high-energy neutrons (5×10^{14} cm^{-2}) and by 600 Mrad of X-Rays nearly no mobility dependence on temperature was observed, though their excitation resulted in current increase by up to one order of magnitude. Furthermore, in these samples current instabilities took place that could be caused by the change of carrier drift paths in a highly disordered material. A model of carrier percolation transport is presented, which foresees a significant influence of spatially extended defect complexes and/or associations, leading to the potential inhomogeneities of the band gap.

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Press release
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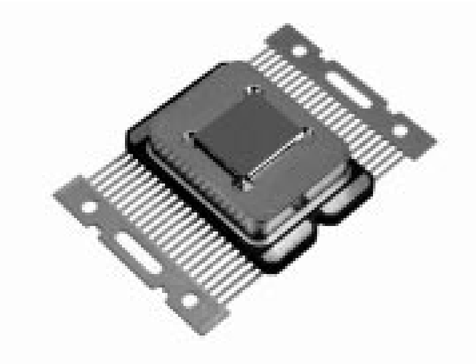

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ULIS S.A.S
Z.I les Iles Cordées, BP 27
38113 Veurey-Voroize
E-mail : ulis@ulis-ir.com
Tel: + 33 (0)4 76 53 74 70
Fax: + 33 (0)4 76 53 74 80
Web: www.ulis-ir.com

