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Impurity breakdown and terahertz luminescence in *n*-GaN epilayers under external electric field

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We report on the observation and experimental studies of impurity breakdown and terahertz luminescence in *n*-GaN epilayers under external electric field. The terahertz electroluminescence is observed in a wide range of doping levels (at noncompensated donor density from 4.5×10^{16} to 3.4×10^{18} cm^{-3}). Spectra of terahertz luminescence and photoconductivity are studied by means of Fourier transform spectrometry. Distinctive features of the spectra can be assigned to intracenter electron transitions between excited and ground states of silicon and oxygen donors and to hot electron transitions to the donor states. © *2009 American Institute of Physics*.
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I. INTRODUCTION

In the past decade, gallium nitride has attracted much attention for applications in the electrically pumped terahertz (THz) emitters due to the unique combination of the high value of the electron drift velocity saturation, large energy of optical phonons, strong interaction of electrons with polar optical phonons, high electrical strength, high temperature stability, and good chemical resistance. Sub-THz and THz emission from the GaN high electron mobility transistors due to current instabilities has been reported.[1,2] The Monte Carlo simulation of the transit-time resonance in bulk zinc blende and wurzite GaN was performed in Ref. 3 and it has been demonstrated that this wide-gap semiconductor is very promising for generation of THz radiation. Stimulated emission of millimeter radiation related to the electron transit-time resonance was experimentally observed in bulk InP,[4] but as far as we know all experimental attempts to realize the transit-time resonance in bulk *n*-GaN has been unsuccessful due to deficient crystal quality of the material.

However, an alterative way to achieve THz emission in bulk *n*-GaN is to use the breakdown of shallow impurity centers in electric field. The emitters of THz radiation based on semiconductors with shallow impurities have been under the extensive study. Promising results were obtained in bulk silicon doped with various donors and acceptors. Terahertz lasing under optical pumping was observed in silicon doped by shallow donors of different types (see Ref. 5 and references therein). Intense spontaneous emission under electrical pumping was observed in bulk silicon doped by both donors and acceptors.[6–8] Remarkable results were obtained in uniaxially stressed *p*-Ge,[9–11] namely THz lasing was observed under electrical pumping. The corresponding optical transitions were attributed to the resonant impurity states which emerge due to the splitting of the valence band and acceptor states under an uniaxial stress. Spontaneous THz emission in strained *p*-doped GaAsN layers under electrical pumping has been observed and studied in Ref. 12. Applications of new materials, such as GaN, provide a possibility for increase in emission intensity and expansion of operating temperature range of the electrically pumped THz emitters based on breakdown of shallow impurities.

This paper reports on the experimental observation of impurity-assisted THz emission from *n*-GaN epitaxial layers under external electric field and presents the results of spectroscopic studies of the emission. The origin of the observed THz emission is also discussed; it can be assigned to the intracenter electron transitions between excited and ground states of Si and O donors and to the hot electron transitions to the donor states.

II. EXPERIMENTAL DETAILS

Gallium nitride epitaxial layers of 4 μm thickness were grown on *c*-plane sapphire substrates covered by 2 μm GaN buffer layer by metal-organic vapor phase epitaxy (MOVPE). Ammonia was used as nitrogen and trimethylgallium as gallium precursor. Silane was used as *n*-type dopant to control the room temperature free electron concentration of the samples from unintentionally doped 4.1×10^{16} to silicon-doped 3.1×10^{18} cm^{-3}. In the MOVPE grown gallium nitride the unintentional *n*-doping originates from residual Si and O donors and N vacancies.[13–15] The electron mobility at room temperature was approximately the same for all the samples, namely 210–270 cm^2/V s. Two Ti/Au electrical contacts of a 3–3.8 mm length with a distance $d = 0.9-3.4$ mm were patterned on the top surface of the samples by a standard photolithography process.

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The current-voltage characteristics were measured with samples immersed in liquid helium or liquid nitrogen and at room temperature as well. To avoid overheating the samples, we used 2 μs voltage pulses with a repetition rate of 1 Hz. The integral THz emission intensity was measured in the same pulse regime using a liquid-helium-cooled Ge:Ga photodetector equipped with a cold black polyethylene filter to prevent unwanted interference of near infrared and visible radiation. The detector sensitivity range (at the level of 20%) extended from 41 to 134 μm (from 9.3 to 30.4 meV).

The THz electroluminescence spectra were measured using a Fourier transform spectrometer (Vertex 80 v Bruker Optics) operating in the step-scan mode. A Mylar beamsplitter was applied. THz radiation was detected using a liquid-helium-cooled silicon bolometer, an amplifier, and a box-car integrator. To eliminate any influence from water vapor absorption, practically whole optical path was filled with gaseous helium. Voltage pulses of 20 μs duration and a repetition rate of 10 Hz were used in these experiments.

Spectral measurements of THz photoconductivity were carried out using the same spectrometer operating in the rapid-scan mode. A mercury lamp was used as a THz radiation source. DC bias was applied to the sample; a low noise current preamplifier (SR 570 Stanford Research Systems) and a low noise voltage preamplifier (SR 560) were used to amplify the signal.

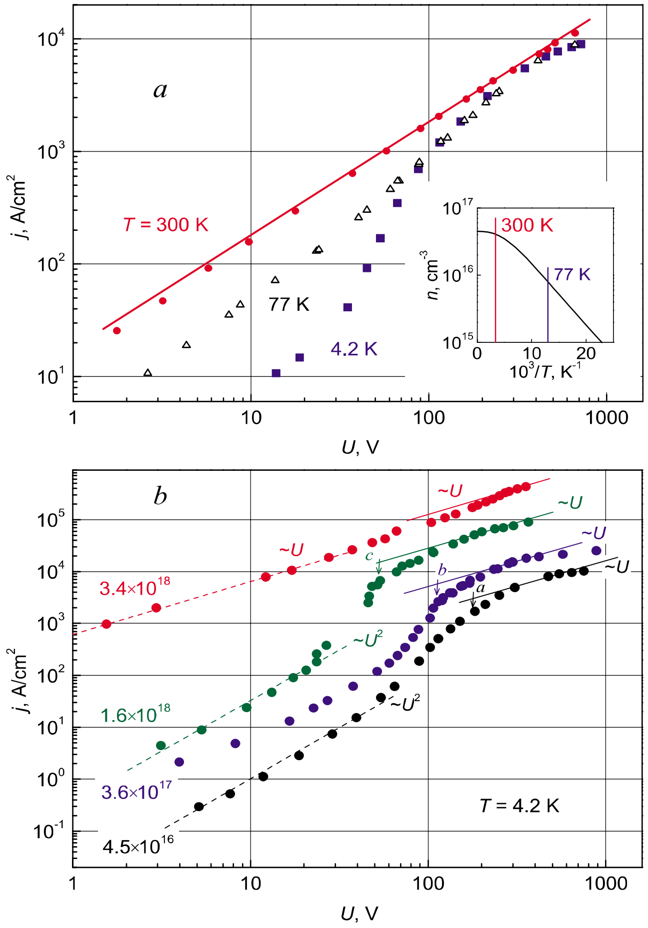

FIG. 1. (Color online) Current-voltage characteristics of n-GaN epitaxial layers. Distance between the contacts $d=0.9$ mm. (a) Measurements at different temperatures for the sample with $N_D-N_A=4.5\times10^{16}$ cm^{-3}. In the inset: calculated dependence of free electron concentration upon inverse temperature for uncompensated n-GaN with $N_D=4.5\times10^{16}$ cm^{-3}. (b) Measurements for different samples at a temperature of 4.2 K. Numbers near the experimental curves denote doping level N_D-N_A in cm^{-3}.

III. RESULTS AND DISCUSSION

Impurity breakdown can be clearly seen at low temperature current-voltage characteristics in semiconductor samples with a relatively low doping level. The doping level needs to be low enough so that an impurity band does not emerge yet. In our experiments we have found that in the case of n-GaN it corresponds to the condition of noncompensated donor density less than 3×10^{18} cm^{-3}.

Experimental dependences of the current density j upon the applied voltage U for one of unintentionally doped GaN samples (noncompensated donor density $N_D-N_A=4.5\times10^{16}$ cm^{-3}) are presented in Fig. 1(a) in the logarithmic scale.

It is interesting to compare the results for various temperatures. At room temperature, when practically all the donors are thermally ionized, the current-voltage characteristic exhibits the linear behavior in the voltage range from 5 to 700 V. At liquid helium temperature one can see a sharp upward rise of conductivity (more than one order of magnitude) at the voltages exceeding 50 V [the threshold voltage is clearly seen in the same graph plotted in the linear scale, similar to Fig. 2(b)]. This feature corresponds to the free electron concentration raise due to processes of impurity breakdown. At the voltage of 300 V all the donors are ionized and further increase in the voltage results in a sublinear behavior of the current-voltage characteristic due to electron mobility decrease caused by free carrier heating.[16–18]

It should be noted that in GaN samples under consideration less than 15% of donors are thermally ionized at liquid nitrogen temperature. Calculated after,[19] the dependence of free electron concentration upon the inverse temperature for uncompensated n-GaN sample with $N_D=4.5\times10^{16}$ cm^{-3} is shown in the inset in Fig. 1(a). For the calculation the ground state donor energy of -30.2 meV was used [this value corresponds to Si donor in GaN (Ref. 20)]. In contrast to Si:P where practically all donors are ionized at 77 K,[6,8] in n-GaN at this temperature it is still possible to realize the impurity breakdown by means of electric field. The experimental curve for $T=77$ K in Fig. 1(a) approves this statement. The impurity breakdown threshold voltage at this temperature is approximately the same as at $T=4.2$ K, namely 50 V.

The low-field conduction of GaN at 300 and 77 K is caused by free electrons thermally released from donor centers. The ratio of low-field conductivities at these temperatures is about 3.5 [see Fig. 1(a)] that is smaller than the ratio for electron concentrations (see inset in Fig. 1) because the electron mobility at 300 K is 1.5 times less than at 77 K. The values of low-field conductivities at these two temperatures obtained using two-probe pulsed method [Fig. 1(a)] are in a good agreement with the independent measurements of conductivity and Hall effect performed in the same structure by means of dc four-probe method. Consequently, one can conclude that the voltage drop at the contacts in the experiments

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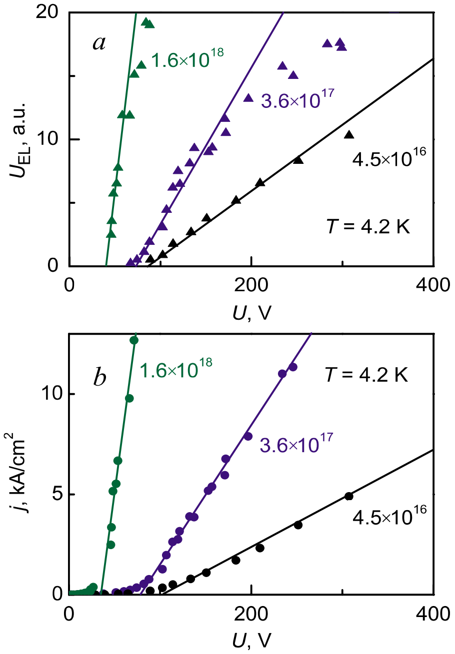

FIG. 2. (Color online) Integrated intensity of THz electroluminescence measured by means of the Ge:Ga photodetector (*a*) and electric current density (b) vs applied voltage. Measurements are carried out at the temperature of 4.2 K. Numbers near experimental curves denote doping level N_D-N_A in cm^{-3}. Distance between the contacts $d=0.9$ mm.

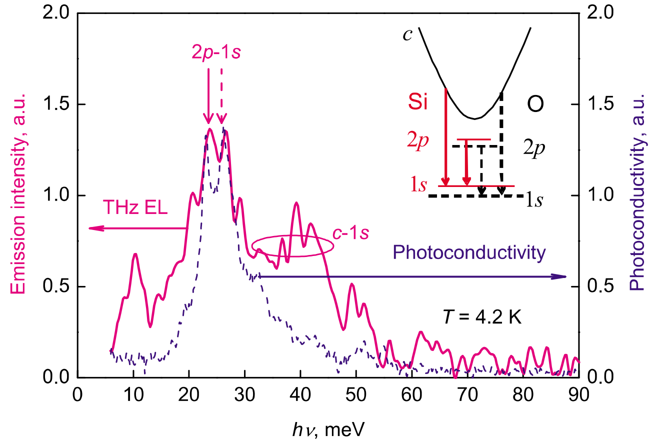

FIG. 3. (Color online) (a) Fourier-transform spectra of THz electroluminescence (the solid curve) and photoconductivity (the dashed curve) at $T=4.2$ K. Distance between the contacts $d=3.4$ mm. The inset shows optical transitions with the participation of the impurity states. Solid arrows correspond to Si donor and dashed arrows correspond to O donor.

considered in Fig. 1(a) is negligible. It allows the determination of a value of the electric field strength inside the sample from the ratio $E=U/d$. In particular, the threshold field of impurity breakdown in unintentionally doped sample is found to be 550 V/cm.

It should be noted that for some samples made from the same wafer, the threshold voltage was higher than 50 V [up to 103 V, see curve for $N_D-N_A=4.5\times10^{16}$ cm^{-3} in Fig. 2(b)]. We believe that the higher threshold voltage is due to the higher contact resistance.

Let us consider current-voltage characteristics for different levels of doping [Fig. 1(b)]. The contact resistance of the samples was checked to ensure a uniform quality of the Ti/Au contacts. All data are taken at liquid helium temperature and plotted in the logarithmic scale. In three samples with lower doping level one can see that at low enough voltages the current density is quadratic in the applied voltage. Such a behavior corresponds to the electron injection through the contact (so-called "space-charge-limited currents"). Under the moderate voltages (35–300 V) the impurity breakdown processes result in sharp increase in the current. At higher voltages the sublinear behavior takes place due to decrease in electron mobility.

With the increase in the doping level (from 4.5×10^{16} to 1.6×10^{18} cm^{-3}), the impurity breakdown threshold shifts to the region of lower voltages (from 103 to 35 V). This is most probably caused by the impurity level broadening. It is well known that under donor concentration increase, the distance between neighbor impurity atoms decreases and the interference of screened Coulomb potential of one donor to another becomes stronger that leads to so-called "classical impurity level broadening." As a result, the ionization energy for the ground impurity state decreases. Such decrease in the ionization energy with increasing impurity concentration was experimentally observed for both donors and acceptors in Ge, Si, GaAs, and other A^3B^5 compounds (see, for example, Ref. 21).

Further increase in the donor concentration leads to a quantum broadening of the impurity levels which results in formation of the impurity band. This is seen in the highly doped sample ($N_D-N_A=3.4\times10^{18}$ cm^{-3}) which demonstrates another type of the current-voltage characteristic. At low voltages in this sample, the current density is linear in the voltage that corresponds to the impurity band conduction. With increase in the voltage, the impurity breakdown takes place and when all the impurities are ionized, the current-voltage characteristic again becomes linear. The increase in the conductivity with respect to low voltages corresponds to the increase in the electron mobility under the carrier transfer from the impurity band to the conduction band.

THz electroluminescence was observed with the Ge:Ga photodetector in all the samples. The integral THz emission intensity at liquid helium temperature was measured as a function of voltage; the experimental results for three samples with different doping levels are presented in Fig. 2(a) in the linear scale. For comparison, current-voltage characteristics of the same samples are shown in Fig. 2(b) (also in the linear scale). One can see from Fig. 2 that the THz electroluminescence has a threshold character and its threshold is close to the threshold of the impurity breakdown for all studied samples. The decrease in the threshold voltage of the impurity breakdown with the increase in doping level results in a corresponding decrease in the threshold voltage of the THz electroluminescence.

To determine the origin of the THz electroluminescence, the emission spectra were studied. The measured spectrum for the unintentionally doped sample with $d=3.4$ mm at $U=325$ V (which is approximately two times larger than the threshold voltage for the sample under consideration) is presented in Fig. 3 (the solid curve). Additionally the spectral dependence of photoconductivity was studied in the same

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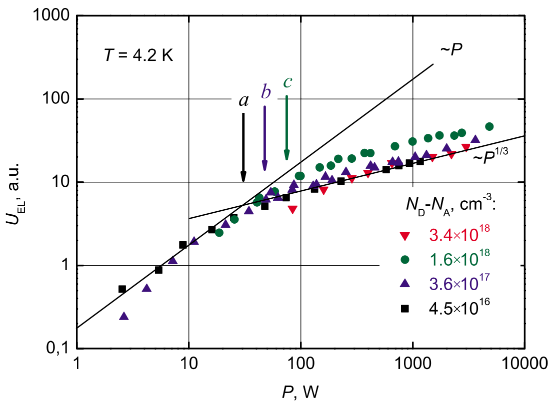

FIG. 4. (Color online) Dependences of the integral intensity of electroluminescence upon the applied electric power for four samples with different doping level. Distance between the contacts $d = 0.9$ mm. Arrows a, b, and c denote the critical electric power related to the switch from the linear behavior to sublinear one (for three samples with lower doping levels). Corresponding critical values of the applied voltage are shown by homonymous arrows in Fig. 1(b).

sample at dc bias of 3 V (the dashed curve in Fig. 3). The both spectra were measured at liquid helium temperature. Two peaks at 23 and 26 meV dominate the photoconductivity spectrum. We attribute these two peaks to 1s-2p intracenter electron transitions for silicon and oxygen donors, respectively. Exactly at the same energies Moore *et al.*[20] observed the optical absorption in n-GaN due to 1s-2p transitions for Si donors (at 23 meV) and for O donors (at 26 meV). The same two peaks dominate the THz electroluminescence spectrum. A wide band with the center at 40 meV exists also in the latter spectrum. We assign this band to the hot electron transitions from the conduction band to the ground states of Si and O donors. The luminescence peaks with energies of 20 meV and lower are most probably caused by the hot electron transitions to the excited donor states.

It should be noted that Ge:Ga detector is very suitable to study the radiative intracenter donor transitions 2p-1s in GaN:Si(O). The detector sensitivity range is from 9.3 to 30.4 meV, so it can be used to measure the *integral intensity* of the intracenter Si and O donor transitions. In Fig. 4 the integral intensity of electroluminescence is plotted as a function of electric power applied to the sample. In the figure the pulse power $P = U \times I$, where U is the amplitude of pulsed voltage applied to n-GaN sample and I is the amplitude of pulsed electric current through the sample. One can see that the power dependencies of the photodetector response U_{EL} for all the samples are very close to each other. All dependences are linear in power for small powers and switch to sublinear dependences at higher powers. For three samples with lower concentration the critical power corresponding to this switch is marked in Fig. 4 by arrows a, b, and c.

What is the reason for such a variation in the power dependence of electroluminescence due to the intracenter 2p-1s transitions? To answer the question one can use the experimental results from the paper[22] where the populations of the ground and excited donor states were studied in n-Ge under conditions of the impurity breakdown. It was found that the population of the ground state 1s monotonically decreases while the population of the excited state 2p reaches the maximum value at the certain electric field. It should be mentioned that at this electric field the population of the ground state amounts approximately one half of its initial value.[22] We suppose that the value of critical power for the power dependence of electroluminescence corresponds to the maximal population of the excited state 2p. This hypothesis is proved by the current-voltage characteristics shown in Fig. 1. The arrows a, b, and c in Fig. 1(b) mark the critical voltages corresponding to the abovementioned critical values of power for different samples. If we analyze the impurity breakdown behavior by means of the current-voltage characteristics [see Fig. 1(b)] we can see that at the arrow positions nearly one half of donors are ionized. Indeed at the abscissa marked by the arrow, the ordinate of the experimental point is approximately two times less than the ordinate of the low-voltage extrapolation of the linear fragment of the current-voltage characteristic related to postbreakdown fields.

Thus, the simultaneous analysis of the electroluminescence spectra, power dependence of electroluminescence and current-voltage characteristics proves that the THz electroluminescence in n-GaN is mostly caused by the intracenter 2p-1s donor transitions.

The main advantage of n-GaN is that it can emit THz radiation under conditions of the shallow impurity breakdown also at liquid nitrogen temperature (and even at higher ones). For unintentionally doped sample the THz electroluminescence was observed at $T = 77$ K. Under the voltage of 400 V ($d = 3.4$ mm) the band of the THz radiation emission belongs to the spectral range from 10 to 35 meV and its spectrum is qualitatively the same as at liquid helium temperature. One can conclude that the same mechanisms are responsible for the THz emission at liquid nitrogen as at liquid helium temperature, namely the intracenter transitions and hot electron transitions from conduction band to the donor states.

IV. SUMMARY

The emission of THz radiation from n-type GaN epitaxial layers was observed under the external electric fields exceeding the impurity breakdown threshold at liquid helium and liquid nitrogen temperatures. Simultaneous analysis of the electroluminescence spectra, power dependence of the electroluminescence intensity and current-voltage characteristics was used to study the emission mechanisms. The observed THz emission can be assigned to the intracenter electron transitions between the excited and ground states of Si and O donors and to the hot electron transitions to the ground donor states. The experimental results on the electroluminescence in GaN are promising for development of the THz emitters operating at 77 K and at higher temperatures.

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