Determination of the types of optical transitions and concentrations of donors and acceptors in GaN by the dependence of photoluminescence intensity on the excitation power

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The results of the calculated and experimental depen-dence of the photoluminescence intensity on the excitation power density for silicon-doped GaN layers grown by molecular beam epitaxy are presented. A model was constructed for transitions in a compensated semiconductor upon interband generation of electron-hole pairs. It is shown that the dependence of the photoluminescence intensity on the excitation power density can be used to determine the recombination mechanism and concentrations of donors and acceptors in semiconductor.

Keywords: GaN, ammonia-MBE, photoluminescence, heterostructures, point defects.

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1. Introduction

The nitrides of the III-group metal and the solid solutions thereof are direct-band semiconductors, which are promising for manufacturing the light-emitting instruments for the visible and ultra-violet ranges of the spectrum. Changing the content of aluminium in the AlGaN solid solutions, we can produce a material with the band gap (E_g) from 3.43 to 6.2 eV, thereby overlapping the spectrum range 200−365 nm. That is why these materials have been applied in practice when creating the light-emitting instruments designed to radiated in all the visible and ultraviolet ranges of the spectrum [1,2]. The effective light-emitting semiconductor instruments can not be created without doping the epitaxial layers for obtaining the *n*- and *p*-type of conductivity. The formation of the GaN epitaxial layers of the *n*-type of conductivity using silicone (Si) as a donor does not create significant difficulty, and the concentration of the electrons can be controllably increased to $10^{20}\,\mathrm{cm}^{-3}$. However, the epitaxial growth of the semiconductor structures is associated with formation of point defects, which can be acceptors and compensate for an alloying impurity.

The photoluminescent spectroscopy (PL) is a contactless fast non-destructive method of the characteristics of the optic diagnostics of such heterostructures. Besides, it requires a small quantity of the substance needed for the investigation. The GaN luminescence spectra have been studied to reveal several bands associated with the electron transitions to the centers with energy levels deeply within the band gap [3]. The „yellow band“ of luminescence with the maximum around 2.2 eV is most actively studied among them. Since the yellow band of luminescence within the GaN layers is observed regardless of the growing technology and manifests itself after implantation of GaN with various ions, then it is associated with intrinsic defects of the GaN crystal lattice and with the main background impurities– silicon, oxygen and carbon or the compounds thereof. According to one of the hypotheses, the yellow band of luminescence is associated with two deep centers with activation energies 300 and 800 meV [4]. Another model suggests two stages of the recombination process involving the three states: a nonradiative capture of the electron from a small neutral hydrogen-like donor to a singly charged deep donor and subsequent radiation recombination between the levels of the deep donor and the small hydrogen-like acceptor [5]. Most of all, it has recognized a model relating the yellow luminescence band to the transitions between the shallow donor levels and the deep acceptor [6]. The yellow band is observed in the undoped GaN layers, i.e. the small donor can be presented by the background impurities– silicon or oxygen. The silicon involvement is confirmed by the fact that the intensity of the yellow band increases with silicon doping, but with the concentration of the silicon atoms above $10^{19}\,\mathrm{cm}^{-3}$ the intensity of the yellow band is decreasing [7–11]. The main candidates to act as an acceptor: the complex $V_{Ga}-O_N$ [12–16] the complex C_N-O_N or the isolated impurity CN [17–20].

Thus, despite that the yellow luminescence band in GaN has been known for a long time, the recombination mechanism and the nature of the centers are still uncertain. At the same time, it provides information on crystal defects and is actively used to improve the technology of their growth, since the ratio of the intensities of the edge ultraviolet band and the yellow band in the GaN luminescence spectrum can be a criterion of its crystal perfection.

However, the procedures of quantitative determination of the concentrations of the donors and the acceptors by the intensities of the edge and yellow bands are unknown.

Usually, the intensity of the yellow band linearly depends on the density of the power of excitation to a certain critical value. At the higher density of the power of excitation, the dependence becomes a sublinear one, while the approximation by the exponential function of the kind $I(J) \sim J^{\gamma}$ provides the power coefficient γ within the range from 1/3 to 2/3 [21–23]. The approximation of this dependence by the function of the kind $I(J) \sim \ln(1 + J/J_1)$ in the studies [24,25] describes the linear portion at $J < J_1$ and the sublinear portion at $J > J_1$. It has been shown that the magnitude J_1 depends on the concentration of the acceptors. The calculation of the acceptor concentration from the magnitude J_1 also requires the measurement of the external quantum efficiency of the yellow band, since within such a model this magnitude takes into account the influence of competing channels of the recombination. It is not possible to calculate the concentration of the donors by this model. A stricter account of other channels of the recombination consists in solving the system of the kinetic equations in the stationary case together with the electroneutrality equation. This approach for explanation of the power dependences of the intensity of the edge photoluminescence bands due to the exciton recombination was suggested in the study [26]. The present study suggests to use the solution of the system of the kinetic equations for determination of the mechanism of recombination of the yellow band in the silicon-doped GaN layers and the concentration of the donors and the acceptors therein. The proposed model is a universal one and can be used for any compensated semiconductors at the high temperatures.

2. Procedures of calculations and experiment

The dependence of the intensity of the various PL channels in the compensated semiconductor on the power of excitation was calculated for the interband mechanism of generation of the electron-hole pairs. Fig. 1 shows the diagram of the electron transitions. The semiconductor includes the donors (D), the acceptors (A) and the centers of nonradiative recombination (NR). The model includes the three channels of the radiation recombination — the interband(BB), the band-acceptor one (eA) and the donor-acceptor one (DA) with involvement of one type of the acceptors and one channel of nonradiative recombination (NR). Following the scheme of the transitions, we write the system of the continuity equations for the free electrons n, the free holes p, the neutral donors N_D^0, the neutral acceptors N_A^0 and the neutral nonradiative centers N_T^0 together with the electroneutrality equation. This system of the nonlinear algebraic equations can be used to obtain the dependence of the concentrations of the charge carriers and the neutral centers on the density of the power

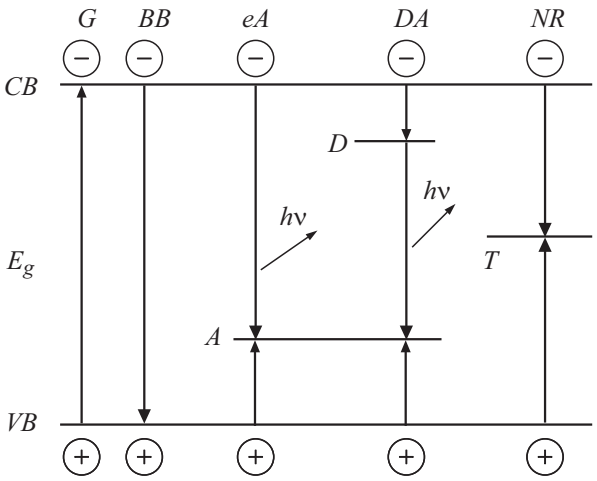

Figure 1. Diagram of the transitions in the compensated semiconductor in interband generation of the charge carriers.

of excitation J. It should be noted that the last but one equation (either of the five) is a linear combination of the previous ones and given here only for the symmetry of the writing:

$$
\begin{aligned}
0 &= \alpha J + W_t \cdot N_D^0 - W_f \cdot n \cdot p - W_{eD} \cdot n \cdot (N_D - N_D^0) \\
&\quad - W_{eA} \cdot n \cdot N_A^0 - W_{eT} \cdot n \cdot N_T^0, \\
0 &= \alpha J - W_f \cdot n \cdot p - W_{hA} \cdot p \cdot (N_A - N_A^0) \\
&\quad - W_{hT} \cdot n \cdot (N_T - N_T^0), \\
0 &= W_{hT} \cdot p \cdot (N_T - N_T^0) - W_{eT} \cdot n \cdot N_T^0, \\
0 &= W_{hA} \cdot p \cdot (N_A - N_A^0) - W_{eA} \cdot n \cdot N_A^0 - W_{DA} \cdot N_D^0 \cdot N_A^0, \\
0 &= W_{eD} \cdot n \cdot (N_D - N_D^0) - W_{DA} \cdot N_D^0 \cdot N_A^0 - W_t \cdot N_D^0, \\
p &+ (N_D - N_D^0) = n + (N_T - N_T^0) + (N_A - N_A^0).
\end{aligned}
\tag{1}
$$

Here α — the coefficient of absorption at the wavelength of laser radiation, W_f — the probability of the radiation recombination of the free charge carriers; W_{eD}, W_{eA} and W_{eT} — the probabilities of the electron capture to the donor, the acceptor and to the nonradiative center; W_t — the probability of thermal emission of the electron from the donor level to the conductivity band, W_{hA} and W_{hT} — the dependences of the capture of the hole to the acceptor and the nonradiative center; W_{DA} — the calculated average probability of the donor-acceptor recombination; N_D, N_A and N_T — the full concentrations of the donors, the acceptors and the nonradiative centers. The probability of the capture of the charge carriers to the centers can be presented as the product of the section of the center capture and the thermal rate of the charge carriers

$$W_{ij} = \sigma_i \cdot v_j. \tag{2}$$

The index i is related to the centers (the donor, the acceptor, the nonradiative one), so is the index j to the carriers (the electron, the hole). The probability of the thermal emission of the electron from the donor level to the conductivity band has been evaluated as

$$W_t = \sigma_e \cdot v_e \cdot N_{\text{eff}} \cdot e^{-\frac{E_d}{kT}}, \tag{3}$$

where N_{eff} — the effective density of the states within the band, $E_d = 26\,\text{eV}$ — the donor depth [27]. Since the recombination probability in the DA-pairs depends on the distance between the donor and the acceptor in the pair (r), it impossible to describe the (D,A)-channel by a simple expression for a rate of the transitions with a fixed value of the transition probability. That is why for solution of the system of the equations, the entire ensemble of the DA pairs can be divided into a countable number of the subassemblies, wherein each of them accepts the pairs in a narrow range of the distances r so as to consider the probability W_{DA} in each subassembly to be constant and solve the system of the equations for each subassembly separately. In order to avoid the complication of the calculation diagram, we have entered the averaged value of the recombination probability, which indicates the most probable value of this magnitude:

$$W_{DA} = \int_0^\infty W(r) f(r) dr, \tag{4}$$

where $W(r) = W_0 e^{-\frac{2r}{a}}$ — the probability of recombination in the donor-acceptor pair the radius r. Here W_0 — the constant indicating the probability of recombination at $r = 0$, a — the Bohr radius of the carrier on the impurity (in this case— the electron on the donor), $f(r) = C_1 r^2 e^{-\frac{e^2}{\varepsilon r k T}} e^{-\frac{4\pi N r^3}{3}}$ — the function of the distribution of the density of the random non-interacting donor-acceptor pairs along the radius r. N — the concentration of the dominant impurity, ε — the permittivity, e — the elementary electrical charge, k — the Boltzmann's constant, T — the temperature, $C_1 = \frac{1}{\int_0^\infty f(r) dr}$ — the normalization factor.

By solving the system (1) of the equations, it is possible to derive the dependences of the rates of the recombination channels, which are written as functions of the density of the photons J:

$$\begin{aligned} I_{BB}(J) &= W_f \cdot n(J) \cdot p(J), \\ I_{eA}(J) &= W_{eA} \cdot n(J) \cdot N_A^0(J), \\ I_{DA}(J) &= W_{DA} \cdot N_D^0(J) \cdot N_A^0(J), \\ I_{NR}(J) &= W_{eT} \cdot n(J) \cdot N_T^0(J). \end{aligned} \tag{5}$$

The studied GaN layers of the thickness of $\sim 1\,\mu\text{m}$ have been grown by the molecular-beam epitaxy (MBE) from ammonia on the Riber unit CBE-32. The substrates were taken to be (0001) oriented sapphire $2''$ plates, one-sided and two-sided polished, having a thickness of $400\,\mu\text{m}$. The layers were doped by silicon (Si) from a gas source of monosilane. The concentration of the silicon atoms in the layers was determined by the secondary ion mass-spectrometry (SIMS) by the procedures described in the article [28], and was $\sim 8 \cdot 10^{18}\,\text{cm}^{-3}$. The concentration of the electrons was determined by the measurements of the Hall effect in the Van der Pauw geometry in the magnetic field of 0.5 T at the room temperature and was $\sim 3 \cdot 10^{17}\,\text{cm}^{-3}$. The PL were measured at the room temperature. The PL was measured in the wide range of the power of excitation by using the 4-th harmonic of the stationary Nd:YAG-laser ($\lambda_{\text{las}} = 266\,\text{nm}$) with the maximum density of the radiation power $12.6\,\text{W/cm}^2$.

3. Results and discussion

Fig. 2 shows the calculated dependences of the rates of the BB, NR, eA and DA channels of recombination and generation G on the density of the power of excitation J. With the laser photon energy $E_{\text{las}} = 4.66\,\text{eV}$, the coefficient of absorption in GaN was $1.8 \cdot 10^5\,\text{cm}^{-1}$ [29]. In the calculation, the following values of the sections of the captures of the charge carriers to the recombination centers have been taken: $\sigma_{eD} = 10^{-21}\,\text{cm}^2$, $\sigma_{eA} = 6 \cdot 10^{-21}\,\text{cm}^2$, $\sigma_{hA} = 6 \cdot 10^{-15}\,\text{cm}^2$, $\sigma_{hT} = \sigma_{eT} = 10^{-16}\,\text{cm}^2$ from the studies [30,31]. The value W_0 was evaluated as $W_0 = W_{\max}/N_A$, since the concentration of the donor-acceptor pairs is specified by the concentration of the non-basic impurity in the material, the value $W_{\max}$ for the donor-acceptor recombination in GaN was selected to be $10^6\,\text{s}^{-1}$ in accordance with the experimental results for measuring the kinetics of the DA recombination, as provided in the study [3]. The concentrations of the donors and the

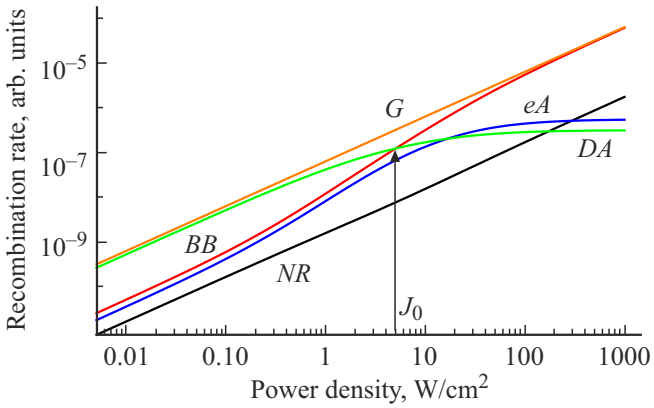

Figure 2. Calculated dependences of the rates of the NR, BB, eA and DA channels of recombination and the rate of generation G on the density of the power of excitation. The concentrations of the donors $N_D = 8 \cdot 10^{18}\,\text{cm}^{-3}$, the concentration of the equilibrium electrons $n_0 = 3 \cdot 10^{17}\,\text{cm}^{-3}$, the concentration of the acceptors $N_A = 2 \cdot 10^{18}\,\text{cm}^{-3}$, the concentration of the nonradiative centers $N_T = 3.5 \cdot 10^{18}\,\text{cm}^{-3}$.

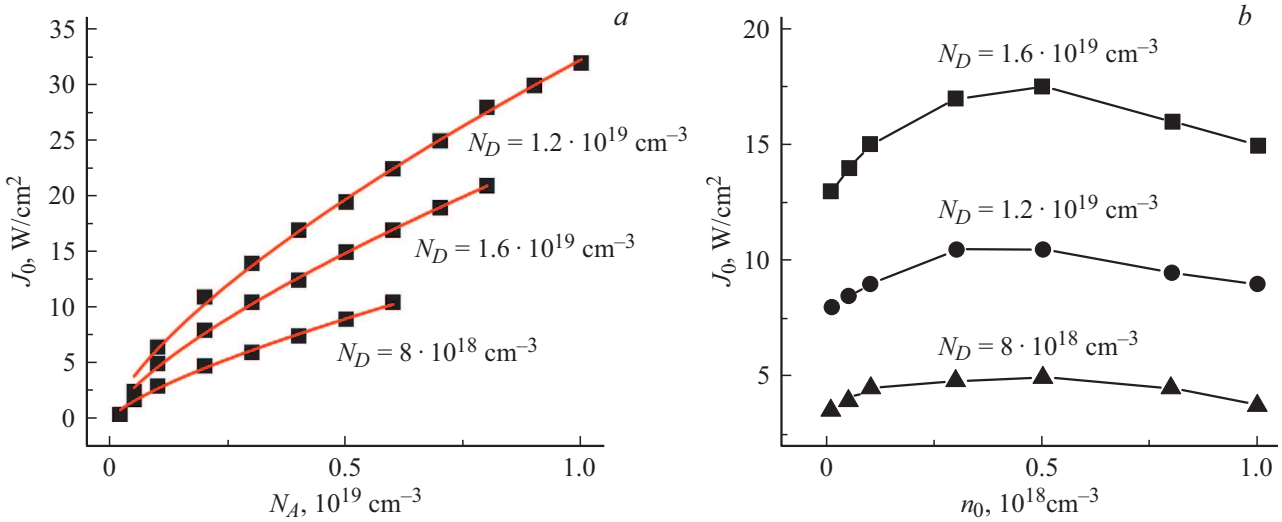

Figure 3. Calculation of the density of the density of the power of excitation J_0, at which the rates of recombination of the BB and DA channels, for $N_D = 8 \cdot 10^{18}$, $1.2 \cdot 10^{19}$ and $1.6 \cdot 10^{19}$ cm^{-3}: a — on the concentration of the acceptors N_A at $n_0 = 3 \cdot 10^{17}$ cm^{-3}, b — on n_0 at $N_A/N_D = 0.25$.

equilibrium electrons were taken to be $N_D = 8 \cdot 10^{18}$ cm^{-3} and $n_0 = 3 \cdot 10^{17}$ cm^{-3}. The concentrations of the acceptors and the nonradiative centers $N_A = 2 \cdot 10^{18}$ cm^{-3} and $N_T = 3.5 \cdot 10^{18}$ cm^{-3}. At $J = 0$, the concentration of the neutral donors $N_D^0 = N_D - N_A - N_T - n_0$, the concentrations of the holes, the neutral acceptors and the neutral nonradiative centers are zero.

At the low values of the density of the power of excitation, the concentrations of the electrons and the neutral donors are approximately equal to the equilibrium ones, and the concentrations of the holes, the neutral acceptors and the neutral nonradiative centers linearly depend on J. In accordance with the system (5) of the equations, the rates of the NR, DA, eA and BB channels linearly depend on J, and the DA channel is dominant. With the increase in the density of the power of excitation above the value $J > 0.1$ W/cm^2, the concentration of the non-equilibrium electrons starts exceeding n_0, and the concentration of the neutral donors starts decreasing, thereby leading to the superlinear dependence of the rates of th eBB and eA channels and the sublinear dependence of the DA channel on J. When $J > 1$ W/cv^2, the acceptors are occupied by the holes due to the big ration of the sections of the capture of the holes and the electrons to the acceptors. As a result, the superlinear dependence of the rate of the eA channel ceases and starts being saturated, so does the rate of the DA channel. At $J_0 = 5$ W/cm^2, the rates of the BB and DA channels are equal, while at the big values of J the BB channel is dominant. The rate of the BB channel increases superlinearly with the increase in J until it is equal to the rate of generation G at $J > 100$ W/cm^2, at doing so, the rates of the eA and DA channels go to the constant level. The rate of the NR channel is not saturated due to an accepted equality of the rates of the sections of the capture of the holes and the electrons to the nonradiative centers. The increase in the concentration of the acceptors at the fixed value of the concentration of the donors and the electrons results in shifting the region (in which the nature of the dependences of the rates of the recombination channels is changing) towards the higher densities of the power of excitation. In the experiment, it is more convenient to record the density of the power J_0, at which the intensities (the recombination rates) of the BB and DA channels are equal.

Fig. 3, a shows the calculation dependences of the magnitude J_0 on the ratio N_A/N_D for $N_D = 8 \cdot 10^{18}$, $1.2 \cdot 10^{19}$ and $1.6 \cdot 10^{19}$ cm^{-3}. The approximation of the dependences by the exponential function ensures the power coefficient $\gamma = 0.7$. Fig. 3, b shows the calculation dependences of the magnitude J_0 on the concentration of the equilibrium ions n_0 at the ratio $N_A/N_D = 0.25$ for $N_D = 8 \cdot 10^{18}$, $1.2 \cdot 10^{19}$ and $1.6 \cdot 10^{19}$ cm^{-3}. In accordance with these dependences, J_0 is insignificantly and non-monotonically changing with the change of n_0. With the known magnitude N_A, it is possible to determine the magnitude N_D in the studied samples GaN by the shift J_0.

Fig. 4, a shows the experimentally measured PL spectra of the typical GaN sample, which are dominated by the bands of the edge PL with the maximum at 3.45 eV and the yellow PL with the maximum at 2.18 eV. The both bands are characterized by the monotonic increase in the integral intensity with the increase in the density of the power of excitation (Fig. 4, b). The intensity of the edge PL band increases superlinearly across the entire density of the power of excitation, and the approximation by the exponential function ensures the power coefficient $\gamma = 1.17$. The intensity of the yellow band increases linearly to $J \sim 1$ W/cm^2, and at the higher values the dependence turns out to be sublinear, while the approximation by the exponential function ensures the power coefficient

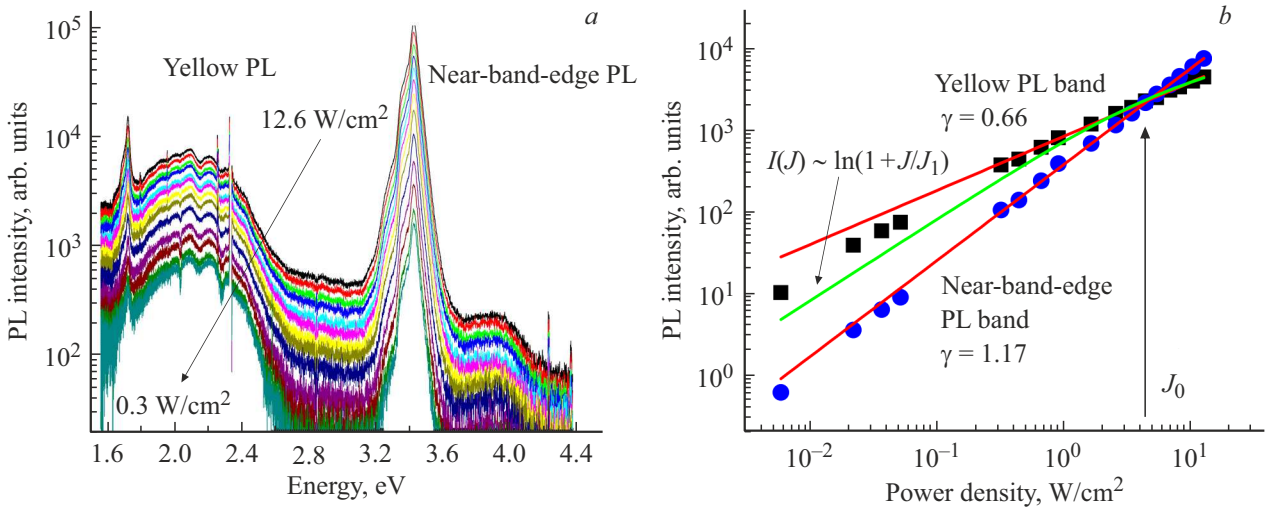

Figure 4. PL GaN spectra (a) and the dependences of the intensity of the edge and yellow PL bands (b) on the power of excitation at 300 K. The solid lines show the approximation by the exponential function of the kind $I(J) \sim J^{\gamma}$ for the both PL bands and by the logarithm of the kind $I(J) \sim \ln(1 + J/J_1)$ for the yellow PL band. (A color version of the figure is provided in the online version of the article).

$\gamma = 0.66$, which is close to the value taken from study [22]. The intensities of the edge and yellow bands are compared at $J_0 \sim 4.5\,\mathrm{W/cm^2}$. These dependences coincide with the calculation dependences of the BB and DA channels of recombination (Fig. 2). Thus, the dependence of the intensity of luminescence on the density of the power of excitation confirms that the yellow band in GaN is caused by the donor-acceptor transitions. The approximation by the function of the kind $I(J) \sim \ln(1 + J/J_1)$ [25] provides the value $J_1 \sim 3.4\,\mathrm{W/cm^2}$, which depends on N_A as follows:

$$\frac{J_1}{E_{\mathrm{las}}} = \frac{N_A}{\alpha\tau\eta}, \qquad (6)$$

where $\tau = 1/(n_0 W_{eA})$ — the lifetime and η — the external quantum efficiency of the yellow band, respectively. Substituting the value $\eta = 0.06$, typical for the yellow band [23–25], in the relationship (6) gives the estimation $N_A = 2 \cdot 10^{18}\,\mathrm{cm^{-3}}$. Thus, as per Fig. 3, a, the values $N_A = 2 \cdot 10^{18}\,\mathrm{cm^{-3}}$ and $J_0 = 4.5\,\mathrm{W/cm^2}$ are correlated to the value $N_D = 8 \cdot 10^{18}\,\mathrm{cm^{-3}}$, which coincides with the value of the full concentration of silicon obtained by the SIMS method. As it is known, silicon in GaN is substantially a small donor not prone to the formation of the DX centers and complexes with intrinsic point defects [19,32,33]. This confirms that this procedure provides the reliable information on the concentrations of the donors and acceptors in GaN.

4. Conclusion

This study has built a model of electron transitions in the compensated GaN, which takes into account the radiation interband, band-acceptor and donor-acceptor transitions and the nonradiative transitions in the interband generation of the electron-hole pairs. The model includes the calculation of the dependences of the rates of the interband, band-acceptor, donor-acceptor and nonradiative recombination on the density of the power of excitation. The dependences of the edge and yellow photoluminescence bands on the density of the power of excitation for the n-doped layers of GaN, grown by the molecular beam epitaxy. It has been shown that the yellow band of photoluminescence in GaN is caused by the donor-acceptor optical transitions. The dependences of the intensity of the interband and donor-acceptor photoluminescence bands on the density of the power of excitation have been analyzed to evaluate the concentration of the donors and acceptors in GaN. The correctness of the performed evaluation of the concentrations shows the coincidence of the concentration of the donors with the full concentration of silicon.

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Conflict of interest

The authors declare that they have no conflict of interest.

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