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Measurement of the Diffusion Length and the Lifetime of Free Excitons in Gallium Nitride Using Cathodoluminescence under Different Conditions of Luminescence Excitation

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Abstract—Using single-crystal *n*-GaN as an example, some possibilities of a procedure for quantitative cathodoluminescence of the direct-gap material are considered. This procedure is based on using the dependence of the cathodoluminescence intensity on the beam-electron energy at a constant level of electron–hole pair generation. Estimates of the diffusion length and the lifetime of free excitons and of the spectral dependence of the coefficient of recombination-radiation self-absorption are obtained using this procedure.

Keywords: gallium nitride, cathodoluminescence, diffusion length of free excitons, lifetime of free excitons, coefficient of recombination-radiation self-absorption

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INTRODUCTION

The diagnostics of electrophysical parameters is an important direction in cathodoluminescence (CL) studies of semiconductor materials making it possible to effectively determine the possibilities of their use in opto- and microelectronics in practice. The authors of [1–4] showed that some electrophysical parameters of direct-gap semiconductors can be found from the dependence of the CL intensity on the energy of the primary electrons of a scanning electron microscope (SEM); in this case, a constant level of minority charge-carrier (MCC) generation was realized.

In the present paper, we consider the possibilities and some problems of the application of our developed procedure for calculating the diffusion length and the lifetime of free excitons in GaN using the dependence of the CL intensity on the energy of a broad electron beam at a constant level of exciton generation. Previously, this procedure has been successfully used for a series of direct-gap semiconductor III–V and II–VI compounds in the study of CL radiation, including that related to the impurity recombination of nonequilibrium charge carriers, and was described in detail in [4–12]. The importance of this study is determined by the small amount of data on determination of the electrophysical parameters of GaN from analysis of the dependence of the CL intensity on the beam-electron energy in spite of a large number of papers dedicated to studying the properties of this material.

APPLIED EQUIPMENT AND THE SAMPLES UNDER STUDY

We studied undoped single-crystal GaN of *n*-type conductivity with a background-impurity concentration on the order of 10^{16} cm^{-3}. It was grown by means of the method of hydride vapor-phase epitaxy (HVPE). The CL spectra were recorded using a modified installation of a commercial electron microscope that made it possible to record the integrated CL signals or the spectral composition of CL signals with a high resolution in a broad range of sample temperatures as well as to control the current and the accelerating voltage of the electron beam [13]. As an alternative source, we used an electron gun (RHEED-gun), which makes it possible to reach much higher currents (up to 50 nA), but with a much lower excitation density [14].

Series of measurements were carried out using a modified scanning electron microscope at a temperature of 10 K in the range of primary-electron beam energies of E_0 = 3–13 keV, and series of measurements were performed at room temperature in the range of energies of E_0 = 3–11 keV using reflection high-energy electron diffraction (RHEED). Because the scanning electron microscope used in the experiment made it impossible to maintain a constant level of exciton generation for all values of the accelerating voltage, the electron-beam current in each measurement was corrected so that the power G of the free exciton source (the number of MCCs generated per time unit)

$$G = iE_0\left[1 - \eta\left(1 - Z^{-1/3}\right)\right]\Big/ q\varepsilon,$$

remains constant. Here, i is the beam current, E_0 is the beam-electron energy, η is the coefficient of primary electron backscattering, Z is the average atomic number of the sample, q is the elementary charge, and ε is the energy of the formation of one electron–hole pair.

The choice of relatively low electron energies was due to the desire to decrease the influence of radiation self-absorption on the recorded CL signal (the self-absorption coefficient was $\alpha \approx (3–6) \times 10^5$ cm^{-1} [15]) and to decrease the possible contribution of the impurity recombination in the surface region under study depleted of majority charge carriers [16], as well as due to the absence of the CL signal below 3 keV for the SEM and due to the impossibility of maintaining a constant level of the source power G above 11 keV for CL measurements performed using RHEED-gun.

PROCEDURE FOR DETERMINING THE ELECTROPHYSICAL PARAMETERS OF SEMICONDUCTOR MATERIALS

To process the experimental data, we used a procedure based on studying the dependence of the CL radiation intensity on the beam electron energy. Previously, the authors of [3, 9] obtained a solution to the direct problem concerning description of the dependence of the CL intensity in a semiconductor with known electrophysical parameters on the SEM electron-beam energy for the procedure proposed in [1] for the first time. The authors of [6–8, 10, 11] used this approach to measure the important parameters of a series of semiconductor materials and optoelectronic structures: MCC diffusion lengths, lifetimes τ, and surface recombination rate S, as well as the spectral dependence of the absorption coefficient $\alpha(\lambda)$ of the edge radiation in direct-gap semiconductors.

We consider the main points of application of this procedure. They include the typical example of solution of the inverse problem in the case of which the sought electrophysical parameters of semiconductors are determined from the results of comparing the calculated and experimental dependences of the CL intensity $I_{CL}(E_0, L, S, \alpha)$ on the probe electron energies for specific values of a series of parameters: the diffusion length, the reduced surface recombination rate, and the radiation self-absorption coefficient. The procedure has two distinctive features. The first feature is determination of the diffusion length for MCCs that participate in the formation of the CL of a specific radiation recombination channel in a semiconductor. In our case, this is the recombination of free excitons, unlike the method of induced current in a short-circuited diode [17], in which this parameter is determined by the simultaneous action of all recombination channels, but, in fact, only of the most short-lived one. The second feature of this procedure is the inclusion of the influence of the surface on the generated MCCs via the generalized parameter of the generation region, namely, its center of gravity z_c, the introduction of which makes it possible to describe I_{CL} under conditions where the linear sizes of the MCC-generation region exceed L with increasing E_0, and the standard model of independent sources becomes inapplicable [9]:

$$z_c = \int_0^\infty z\rho(z)dz \Big/ \int_0^\infty \rho(z)dz, \tag{1}$$

where $\rho(z)$ is the sample-depth distribution of the generated-MCC density.

If relation (1) is taken into account, the expression for the CL intensity in the one-dimensional approximation (a broad electron beam) has the form

$$I_{CL}(E_0) \sim \left(1 - \frac{S}{S-1}\exp\left(-\frac{z_c}{L}\right)\right)\exp(-\alpha z_c). \tag{2}$$

Here, $S = \upsilon_s L/D$ is the reduced rate of MCC surface recombination (υ_s is the surface recombination rate for MCCs diffusing to the region depleted of majority carriers, and D is the diffusion coefficient) [18], z_c is the center of gravity of the generation region, L is the MCC diffusion length, and α is the radiation-absorption coefficient.

RESULTS AND DISCUSSION

Analysis of the obtained CL radiation spectra showed that CL radiation related to free-exciton recombination dominated at a low He temperature and at room temperature [19]. For lightly doped n-GaN with a donor concentration on the order of 8×10^{15} cm^{-3}, the maximum electric-field strength near the GaN surface is 5×10^3 V/cm, which is insufficient for exciton decay even at room temperature. Figure 1a shows the CL spectrum obtained in the study of the GaN sample at room temperature and for a beam-electron energy of 7 keV. For comparison, Fig. 1b shows the photoluminescence spectrum of the GaN sample given in [19].

Based on the data obtained at a temperature of 10 K, to obtain z_c, we calculated the dependence of the function $\rho(z)$ of the density of the distribution of free excitons generated in the GaN sample over the depth z at normal incidence of the beam with energies ranging from 3 to 13 keV. The distribution density function

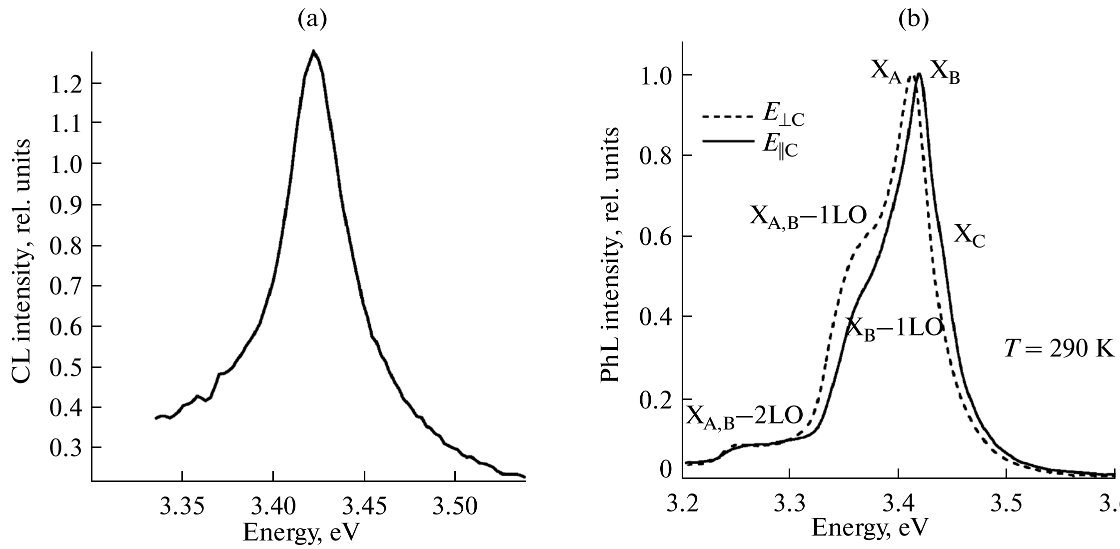

Fig. 1. Spectra of GaN samples: (a) the CL spectrum obtained in the study of the GaN sample at room temperature and an accelerating voltage of 7 keV and (b) the photoluminescence spectrum of the GaN sample given in [19].

for free excitons was calculated within the framework of the model in [20] in accordance with the formula

$$\rho(z)=\begin{cases}\dfrac{A_N(1-\eta)}{\pi^{0.5}z_p}\exp\left(-\dfrac{(z-z_p)^2}{z_p(z_p+k(2z_p-z))}\right)\\ +\dfrac{1.085\eta}{\pi^{0.5}z_{tr}}\exp\left(-\dfrac{(z-z_{ss})^2}{z_{ss}^2}\right), \quad z_p<z,\\ \dfrac{A_N(1-\eta)}{\pi^{0.5}z_p}\exp\left(-\dfrac{(z-z_p)^2}{z_p(z_p+kz)}\right)\\ +\dfrac{1.085\eta}{\pi^{0.5}z_{tr}}\exp\left(-\dfrac{(z-z_{ss})^2}{z_{ss}^2}\right), \quad z\ge z_p.\end{cases} \qquad (3)$$

Here, A_N is the normalization factor, η is the coefficient of beam-electron backscattering, $k = 0.6$ [20] is a coefficient taking into account the influence of inelastic scattering on the spatial distribution of electron energy losses during multiple beam-electron scattering, z_{tr} is the beam-electron transport length in the sample, $z_p = 0.77z_{tr}$ is the most probable path of absorbed electrons that experienced multiple scattering in the sample, $z_{ss} = Z^{-1/3}z_{tr}$ is the depth of the maximum energy losses of backscattered beam electrons, and Z is the average atomic number of the sample material.

Function (3) takes into account the contributions of absorbed and backscattered electrons correctly and, as shown in [20], agrees well with experimental data in a broad range of primary beam-electron energies.

The sample-depth-distribution density functions (calculated in accordance with formula (3)) for free excitons generated in GaN are shown in Fig. 2. The corresponding values of z_c for the chosen range of energies E_0 in GaN are given in the table.

The diffusion length L for free excitons was determined from comparison of the experimental data with the corresponding calculated curves $I_{CL}(E_0)$. In the range of obtained experimental data, this value was 0.25 ± 0.05 μm for room temperature and 0.1 ± 0.05 μm for a temperature of 10 K. The obtained values of L correspond, on the whole, to the results of measuring this parameter by means of other methods [16, 17]. Their important feature is that, in our case, they are related to and characterize precisely the free exciton transport in epitaxial GaN.

Starting from these values of L and the relation $\tau = L^2/D$, we estimated the free-exciton lifetime τ.

Values of the center-of-gravity coordinate z_c of the generation region calculated using formula (1)

E_0, keV	3	4	5	6	7	8	9	10	11	12	13
z_c, μm	0.0241	0.0392	0.0578	0.0786	0.1029	0.1283	0.1568	0.1864	0.2191	0.2529	0.2897

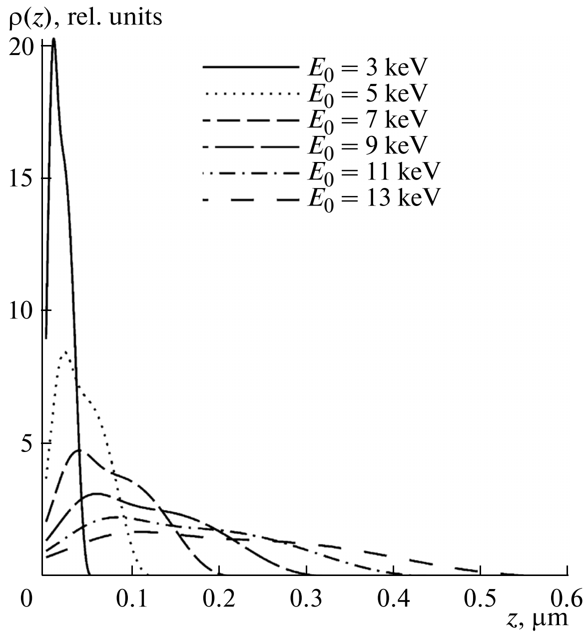

Fig. 2. Calculated dependences of the function $\rho(z)$ of the density of the distribution of free excitons generated in the GaN sample over depth z at normal incidence of the beam with energies ranging from 3 to 13 keV. The dependences were calculated using formula (3) on the basis of experimental data obtained in studying the GaN sample at a temperature of 10 K.

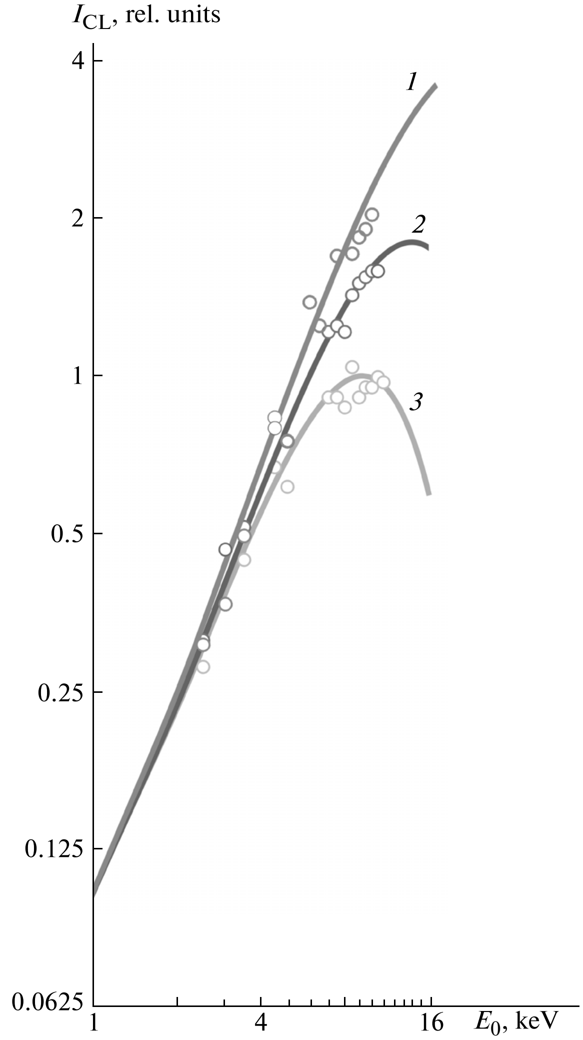

Fig. 3 Calculated dependences of the CL radiation intensity (I_{CL}) on the beam-electron energy (E_0) at room temperature and the diffusion length of free excitons $L = 0.25 \pm 0.05$ μm for different portions of the GaN spectrum with energies E = (*1*) 3.4070, (*2*) 3.4160, (*3*) 3.4340 eV and for the absorption coefficients α = (*1*) 3×10^3, (*2*) 1.4×10^4, and (*3*) 5×10^4 cm^{-1}. The experimental data obtained in studying the GaN sample at room temperature are denoted by points.

The main problem of determination of the lifetime τ from the measurement of L involves correct choice of the diffusion coefficient D, because data on the diffusion coefficient obtained by different authors differ greatly. If it is assumed that the diffusion coefficient depends slightly on temperatures and is 1–2 cm^2/s, then the free-exciton lifetime ranges from 310–630 ps at room temperature to 50–100 ps at liquid-helium temperature.

In addition to estimates of the free-exciton diffusion length L, those of the edge radiation-absorption coefficient α were given for the experimental data obtained at room temperature.

Figure 3 shows the calculated dependences of the CL radiation intensity I_{CL} on the beam-electron energy E_0 at room temperature and the free-exciton diffusion length $L = 0.25 \pm 0.05$ μm for different portions of the GaN spectrum with the energies E = 3.4070, 3.4160, and 3.4340 eV and for the absorption coefficients $\alpha = 3 \times 10^3$, 1.4×10^4, and 5×10^4 cm^{-1}, respectively. The points denote the experimental data obtained at room temperature, and the solid lines correspond to the calculated dependences $I_{CL}(E_0)$ calculated using formula (2).

In Fig. 4, the solid line corresponds to the spectral dependence of the edge radiation-absorption coefficient α at room temperature obtained using the approach illustrated in Fig. 3. For comparison, the spectral dependence of the radiation-absorption coefficient given in [15] is shown by a dashed line. It can be seen from Fig. 4 that the calculated values of the absorption coefficient obtained using the given procedure correspond to the results in [15].

CONCLUSIONS

We have considered some problems of CL diagnostics of the electrophysical parameters of GaN, which was based on studying the dependence of the CL radiation intensity on the beam-electron energy. We

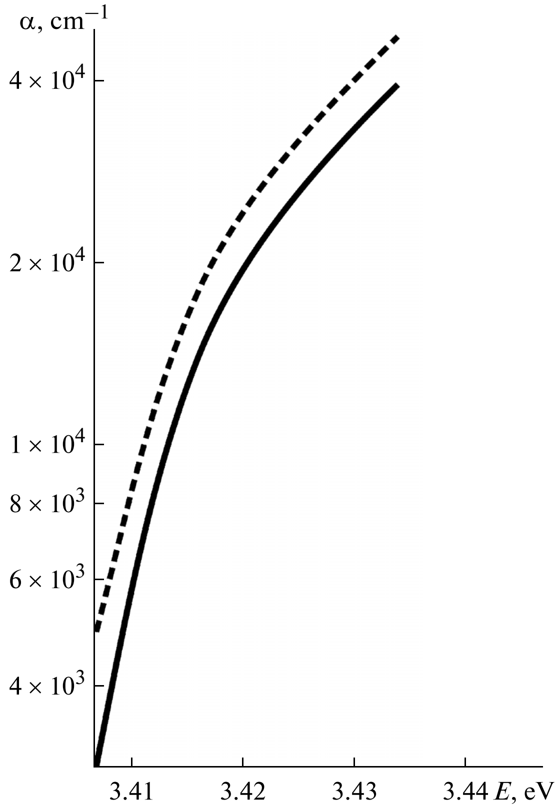

Fig. 4. Spectral dependences of the edge radiation-absorption coefficient α at room temperature. The solid line corresponds to the calculated dependence obtained using the data of the CL study of the GaN sample at room temperature, and the dashed line, to the results in [15].

obtained estimates of the diffusion length and lifetime of free excitons in GaN and showed the possibilities of determining the edge radiation-absorption coefficient. We found that, on the whole, the obtained values of the electrophysical parameters correspond to the results of measuring these parameters by means of other methods.

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