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Optical properties of GaN epitaxial layers in mid- and far-infrared ranges

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Abstract. In this work, the transmission of microstructures based on gallium nitride with different doping levels in the mid- and far-infrared spectral ranges at T = 300 K was experimentally studied. The transmission of the studied structures in these spectral ranges was modeled using the transfer matrix method. It is shown that the contribution of the lattice, according to the single-phonon resonance model, and the contribution of free electrons, according to the Drude model, to the dielectric constant allows one to satisfactorily describe the optical properties of the studied microstructures up to a quantum energy of 300 meV. The absorption coefficient for CO_2 laser radiation (photon energy 117 meV) has been calculated. It has been shown that in gallium nitride absorption on free electrons at a given photon energy can be observed experimentally at an electron concentration exceeding $6{\cdot}10^{16}$ cm^{-3}. The optimal thickness of GaN layers for experimental observation of the absorption modulation of CO_2 laser radiation in electric field for different doping levels was determined.

Keywords: gallium nitride, free electrons, light transmission, absorption, mid-infrared spectral range, terahertz spectral range

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Материалы конференции
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Оптические свойства эпитаксиальных слоев GaN в среднем и дальнем инфракрасных диапазонах

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Аннотация. В работе экспериментально исследовано пропускание микроструктур на основе нитрида галлия с разным уровнем легирования в среднем и дальнем инфракрасном диапазонах при T = 300 K. Проведено моделирование пропускания исследуемых структур в данных спектральных диапазонах с помощью метода матриц переноса. Показано, что вклад решетки, согласно модели однофононного резонанса, и вклад свободных

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электронов, согласно модели Друде, в диэлектрическую проницаемость позволяет удовлетворительно описывать оптические свойства исследованных микроструктур вплоть до энергии кванта 300 мэВ. Рассчитан коэффициент поглощения для излучения CO_2-лазера (энергия кванта 117 мэВ). Показано, что в нитриде галлия поглощение на свободных электронах при данной энергии кванта экспериментально можно наблюдать при концентрации электронов, превышающей $6 \cdot 10^{16}$ см$^{-3}$. Определена оптимальная толщина пленок GaN для экспериментального наблюдения модуляции поглощения излучения CO_2-лазера в электрическом поле для разных уровней легирования.

Ключевые слова: нитрид галлия, свободные электроны, пропускание, поглощение, средний инфракрасный диапазон, терагерцовый диапазон

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Introduction

Modern opto- and nanoelectronics devices use various III-V group semiconductors. Gallium nitride stands out as a promising material for developing optical devices in the visible and infrared spectral ranges due to its unique combination of physical properties. The operation of many optoelectronic devices is accompanied by heating of charge carriers in an electric field; their functioning is also significantly affected by the absorption of radiation on nonequilibrium charge carriers. To analyze the absorption of radiation in an electric field, information about absorption under equilibrium conditions is required. The goal of this work is experimental and theoretical study of the equilibrium absorption of radiation by free electrons in epitaxial layers of gallium nitride.

Materials and Methods

In this work, we studied films of hexagonal *n*-type gallium nitride grown by metalorganic vapor phase epitaxy on a sapphire substrate with a buffer layer of specially undoped GaN. The thickness of the epitaxial films was 4.3 µm, the thickness of the buffer layer was 2 µm, and the thickness of the sapphire substrate was 440 µm. Films with different doping levels were studied. Based on the study of the Hall effect and electrical conductivity at room temperature, the following values of concentration (mobility) of free electrons were obtained: $3.1 \cdot 10^{17}$ cm^{-3} (246 cm^2/V·s), $1.4 \cdot 10^{18}$ cm^{-3} (231 cm^2/V·s) and $3.4 \cdot 10^{18}$ cm^{-3} (205 cm^2/V·s) for samples No. 1, 2 and 3, respectively. The optical transmission spectra of the microstructures were studied at room temperature in the mid- and far-infrared spectral ranges. The experiments were carried out using a Bruker Vertex 80v vacuum Fourier spectrometer operating in fast scanning mode. The radiation source was a globar. For studies in the mid-infrared (MIR) spectral range, a DLaTGS pyroelectric photodetector and a KBr beam splitter were used, and for studies in the far-infrared (terahertz) spectral range, a DLaTGS FIR pyroelectric photodetector and a Mylar beam splitter were used. The sample was placed in a Fourier transform spectrometer, which was evacuated to a pressure of 5 hPa; radiation from the globar, incident normal to the surface, passed through the sample and was measured by a photodetector.

Results and Discussion

Our samples are multilayer structures: GaN film / GaN buffer layer / sapphire substrate. It is convenient to model the transmission and reflection of radiation by such structures using the transfer matrix method [1, 2]. In this method, the reflection coefficients *R* and transmission

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coefficients T are calculated using a matrix M of size 2×2. To obtain the matrix M, matrices describing the passage of an electromagnetic wave through each j-th layer P_j, as well as describing the passage of an electromagnetic wave through the boundary between the j-th and (j+1) layer $D_{j/(j+1)}$. In the case of normal incidence of light, the matrix P_j is calculated as follows:

$$P_j = \begin{pmatrix} \exp(i\phi_j) & 0 \\ 0 & \exp(-i\phi_j) \end{pmatrix}, \tag{1}$$

where $\varphi_j = \frac{\omega}{c}\tilde{n}_j d_j$ is the phase change of the electric field of the wave when passing through layer j, ω is the angular frequency of the electromagnetic wave, c is the speed of light, $\tilde{n}_j = n_j + ik_j$ is the complex refractive index of layer j, n_j, k_j and d_j are the real refractive index, the extinction coefficient and the thickness for the layer j, respectively. In the case of normal incidence of light, the matrix $D_{j/(j+1)}$ is calculated as follows:

$$D_{j+1/j} = \frac{1}{2\tilde{n}_{j+1}} \begin{pmatrix} \tilde{n}_{j+1} + \tilde{n}_j & \tilde{n}_{j+1} - \tilde{n}_j \\ \tilde{n}_{j+1} - \tilde{n}_j & \tilde{n}_{j+1} + \tilde{n}_j \end{pmatrix}. \tag{2}$$

The transfer matrix M for our structure is written as follows:

$$M = D_{4/3} P_3 D_{3/2} P_2 D_{2/1} P_1 D_{1/0}, \tag{3}$$

where index 0 corresponds to vacuum, index 1 to the doped GaN epitaxial layer, index 2 to the GaN buffer layer, index 3 to the sapphire substrate, and index 4 to vacuum. Knowing the elements of the matrix M, we can calculate the coefficients of reflection:

$$R = \left| \frac{M_{21}}{M_{22}} \right|^2, \tag{4}$$

and transmission:

$$T = \left| M_{11} - \frac{M_{21} M_{12}}{M_{22}} \right|^2. \tag{5}$$

The spectra of reflection and transmission coefficients calculated using (4) and (5) contain oscillations with a small period caused by interference in the “thick” substrate. However, we did not observe them experimentally due to insufficient resolution. Therefore, according to [2], it is possible to exclude these oscillations by considering the substrate as an “incoherent layer.” Then for the substrate, the matrix P is transformed to the following form:

$$P_3(\theta) = \begin{pmatrix} \exp\left(i\left[\phi_3 + \frac{\theta}{2}\right]\right) & 0 \\ 0 & \exp\left(-i\left[\phi_3 + \frac{\theta}{2}\right]\right) \end{pmatrix}, \tag{6}$$

and the transfer matrix is written as follows:

$$M(\theta) = D_{4/3} P_3(\theta) D_{3/2} P_2 D_{2/1} P_1 D_{1/0}. \tag{7}$$

To obtain R and T, it is necessary to average (4) and (5) over θ:

$$R = \frac{1}{2\pi} \int_{-\pi}^{\pi} \left| \frac{M_{21}(\theta)}{M_{22}(\theta)} \right|^2 d\theta, \tag{8}$$

$$T = \frac{1}{2\pi} \int_{-\pi}^{\pi} \left| M_{11}(\theta) - \frac{M_{21}(\theta) M_{12}(\theta)}{M_{22}(\theta)} \right|^2 d\theta. \tag{9}$$

For GaN layers $\tilde{n}$ can be calculated using the Fresnel equation:

$$\tilde{n}^2 = \varepsilon, \tag{10}$$

where ε is the dielectric constant. The dielectric constant contains contributions from the lattice (according to the single-phonon resonance model) and free electrons (according to the Drude model):

$$\varepsilon(\omega) = \varepsilon_\infty \left(1 + \frac{\omega_{LO}^2 - \omega_{TO}^2}{\omega_{TO}^2 - \omega(\omega + i\gamma_q)} - \frac{\omega_{pl}^2}{\omega(\omega + i\gamma_e)}\right), \tag{11}$$

where ε_∞ is the high-frequency dielectric constant, ω_{LO} is the frequency of the longitudinal optical phonon, ω_{TO} is the frequency of the transverse optical phonon, γ_q is the damping constant of the optical phonon, $\omega_{pl} = \sqrt{\frac{4\pi N_e e^2}{\varepsilon_\infty m_e}}$ is the plasma frequency, N_e is the concentration of free electrons, m_e is the effective mass of the electron, $\gamma_e = 1/\tau_e$ is the plasmon damping constant, τ_e is the electron relaxation time determined from expressions for mobility $\mu_e = \frac{e\tau_e}{m_e}$.

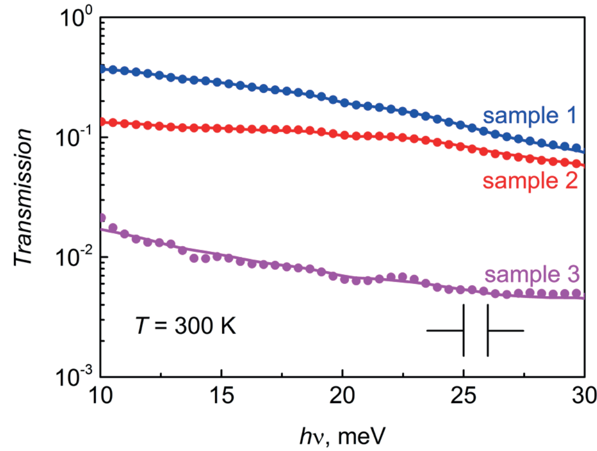

Fig. 1. Experimental (circles) and simulated (lines) transmission spectra in the THz spectral range for sample 1 (blue line and circles), 2 (red line and circles) and 3 (magenta line and circles). The experiment was carried out at room temperature

Using (9), the transmission spectra of the samples under study were simulated. For sapphire, the values n and k from [3, 4] were used, and for GaN layers they were calculated according to (10) and (11). For the buffer layer, characteristic concentration and mobility values for intentionally undoped GaN films were used: $N_e = 4{\cdot}10^{16}$ cm^{-3}, $\mu_e = 200$ cm^2/V·s [5]. The ω_{pl} and γ_e values of the doped GaN layers were used as fitting parameters for the model. For simplicity, the effective electron mass is assumed to be constant for all doping levels and equal to $0.2m_0$. Other necessary parameters of gallium nitride were taken from [6–8]. In Figure 1, solid lines show the results of modeling the transmission of the structures under study at the best-fit parameters. It can be seen that the model calculation well describes the transmission spectrum in the THz spectral range. Table presents the parameters of the doped GaN layers obtained from the study of the Hall effect and electrical conductivity, and the parameters obtained from the simulation of the transmission in the terahertz spectral range. As the doping level increases, the concentration and mobility values obtained from Hall measurements and from transmittance spectrum simulations differ less and less. The concentration values for the least doped GaN film differ the most. Thus, it can be said that the single-phonon resonance model and the Drude model adequately describe the contribution of the lattice and electrons to the dielectric constant of GaN in the terahertz spectral range.

Fig. 2, *a* shows the experimental transmission spectra of the samples in the mid-infrared spectral range. These spectra have several features. Firstly, the spectra exhibit oscillations, the

Table

Parameters of samples obtained by various methods

Sample	Hall and conductivity experiments		Terahertz transmission experiment			
	N_e, cm^{-3}	μ_e, cm^2/V·s	ω_{pl}, meV	γ_e, meV	N_e, cm^{-3}	μ_e, cm^2/V·s
1	$3.1\cdot10^{17}$	246	9	12.1	$6.3\cdot10^{16}$	478
2	$1.4\cdot10^{18}$	231	25.3	18.3	$4.9\cdot10^{17}$	316
3	$3.4\cdot10^{18}$	205	58.6	27.9	$2.7\cdot10^{18}$	207

period of which is almost the same for all the samples under study. These oscillations are caused by the interference of light in the buffer layer and the doped GaN film. At a frequency ω greater than $4.56\cdot10^{14}$ rad/s (photon energy greater than 300 meV), the real refractive index n is practically independent of the doping level, and absorption is low (Fig. 3), i.e., n is determined by the lattice and can be estimated as $\sqrt{\varepsilon_\infty}$. Knowing the period of oscillations and the refractive index, it is easy to obtain the thickness of the layer in which interference occurs. This thickness was 6.3–6.4 μm, which is consistent with the technologists' data. Secondly, at photon energies less than 170 meV there is no transmission. This is due to absorption in the sapphire substrate [9]

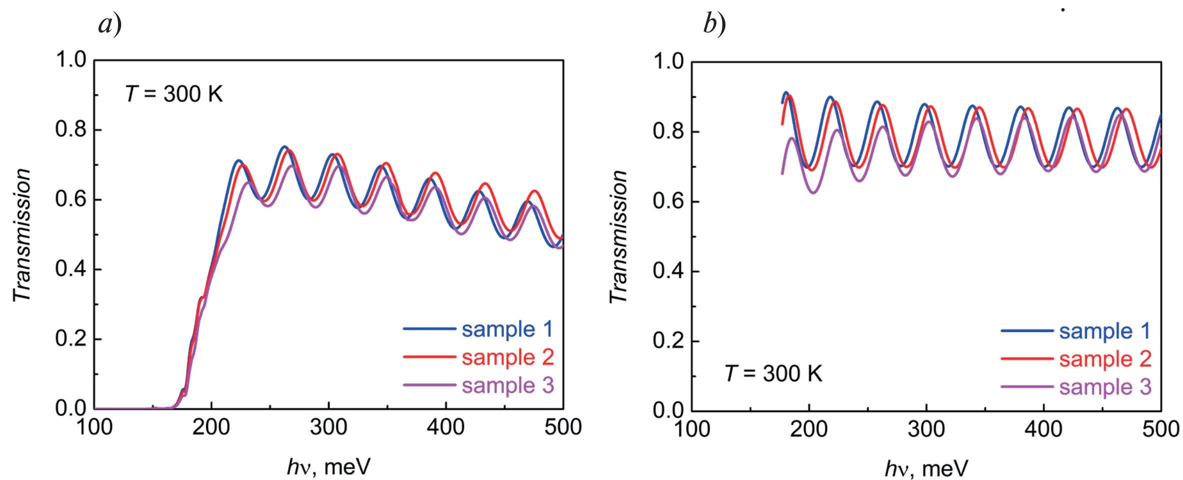

Fig. 2. Experimental (a) and simulated (b) transmission spectra in the mid-infrared spectral range for sample 1 (blue line), 2 (red line) and 3 (magenta line).
The experiment was carried out at room temperature; resolution is 1 meV

Fig. 2, b shows the simulated transmission spectra of the structures under study in the mid-infrared spectral range, calculated using (9). The values of n and k for sapphire were taken from [9], and for GaN layers were calculated using (10) and (11). In this case, the parameters of the buffer layer remained the same, and for the parameters of the doped GaN films, the values obtained as a result of modeling transmission in the THz region were taken (Table). Fig. 2 demonstrates that the period and amplitude of the calculated transmission oscillations coincide well with the experimental ones. It is also clear that at photon energies less than 300 meV, the transmission of samples decreases with increasing doping level, which is due to absorption by free electrons. At the same time, the experimental and theoretical transmission spectra differ. Firstly, the simulated transmission spectra do not demonstrate sharp monotone increase in range from 170 to 220 meV. Secondly, the theoretical transmittance curves are located slightly higher on the OY axis. Thirdly, a decrease in transmission is observed, starting with a photon energy of 300 meV. These differences are explained by ambiguous data for n and k for sapphire in the mid-infrared spectral range, as well as extra absorption mechanisms in GaN, which are not taken into account in model (11). It can be concluded that the single-phonon resonance model and the

Drude model satisfactorily describe the contribution of the lattice and electrons to the dielectric constant in the mid-infrared range up to photon energies of 300 meV.

Fig. 3 shows the spectra of the real refractive index and extinction coefficient of gallium nitride, calculated on the basis of (10) and (11) for the samples under study. These dependences were also calculated for pure GaN ($N_e = 0$), which makes it possible to identify the contribution of free electrons to n and k. The feature in the region of 100 meV, characteristic of all samples, is due to the Reststrahlen band of GaN. From a comparison with n and k for pure GaN, it is clear that the presence of free electrons greatly modifies the spectra. In this way, in the spectra of the real refractive index in the terahertz region, a minimum appears, which shifts to the high-frequency region with increasing concentration of free electrons. The value of extinction coefficient increases monotonically with N_e growth over the entire range.

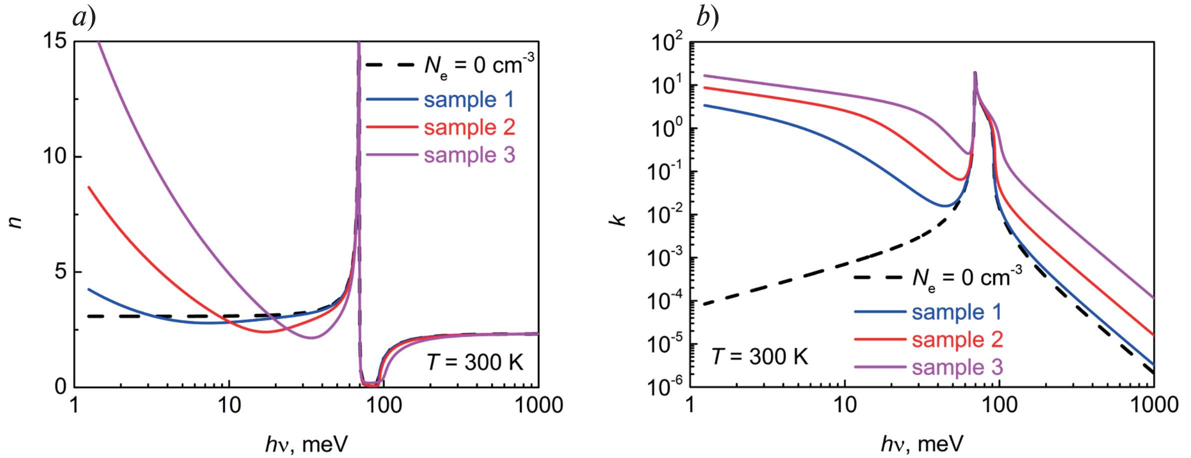

Fig. 3. Spectra of the real refractive index (a) and extinction coefficient (b) for pure GaN (dashed line), sample 1 (blue line), 2 (red line) and 3 (magenta line)

For experimental studies of absorption, including studies under conditions of carrier heating by an external electric field, in the mid-infrared spectral range it is convenient to use a CO_2 laser (radiation wavelength 10.6 μm, photon energy 117 meV). Fig. 4 shows the absorption spectra of the samples under study and pure GaN, calculated in a wide range of photon energies from the relation

$$\alpha = 2\frac{\omega}{c}k. \quad (12)$$

For free electron concentrations of 0, $6.3 \cdot 10^{16}$, $4.9 \cdot 10^{17}$ and $2.7 \cdot 10^{18}$ cm^{-3}, the absorption

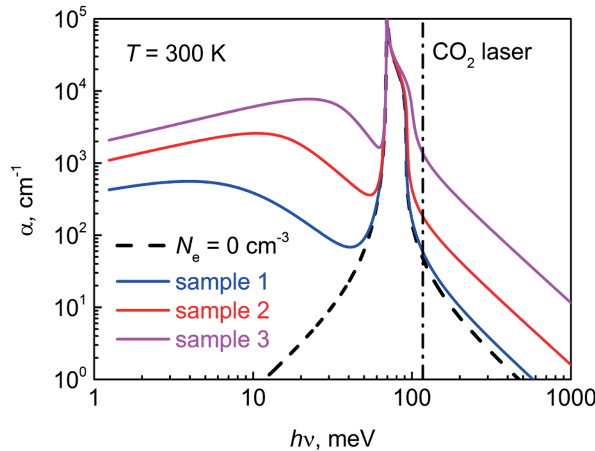

Fig. 4. Absorption coefficient spectra for pure GaN (dashed line), sample 1 (blue line), 2 (red line) and 3 (magenta line). The vertical dash-dotted line corresponds to the energy of the CO_2 laser photon (117 meV)

coefficient at a photon energy of 117 meV is 48, 59, 183 and 1370 cm^{-1}, respectively. Thus, for sample No. 1, the contribution of free electrons to absorption is quite small, therefore, for experimental studies it is necessary to use samples with a concentration noticeably higher than $6 \cdot 10^{16}$ cm^{-3}. To study the effects of electroabsorption in *n*-GaN epitaxial films at the wavelength of a CO_2 laser radiation, the optimal film thickness decreases with increasing concentration of free electrons. To determine it, equality $\alpha d = 1$ can be used. Thus, for electron concentrations of $4.9 \cdot 10^{17}$ and $2.7 \cdot 10^{18}$ cm^{-3}, the optimal film thickness is 55 and 7 μm, respectively. It is advisable to use silicon as a substrate.

Conclusion

In this work, the transmission of GaN-based microstructures in the terahertz and mid-infrared spectral ranges at room temperature was experimentally studied. The transmission of the studied structures was simulated using the transfer matrix method. The dielectric constant of GaN was the sum of the contributions from the lattice (according to the single-phonon resonance model) and free electrons (according to the Drude model). Transmission modeling in the THz range made it possible to obtain refined values of the parameters of doped gallium nitride films, and modeling in the mid-infrared spectral range demonstrated that the single-phonon resonance model and the Drude model in the structures we studied provide adequate results up to photon energies of 300 meV. The absorption coefficient of GaN was also calculated in a wide range of photon energies for different doping levels and the free electrons contribution to the absorption was revealed. In particular, absorption at the photon energy of CO_2 laser radiation is considered. It has been shown that in gallium nitride, absorption on free electrons at a given photon energy can be experimentally observed at an electron concentration exceeding $6 \cdot 10^{16}$ cm^3. Thus, for free electron concentrations of $4.9 \cdot 10^{17}$ and $2.7 \cdot 10^{18}$ cm^{-3}, the optimal thickness for observing the absorption modulation of CO_2 laser radiation in electric field is 55 and 7 μm, respectively.

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