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Light dispersion and edge absorption in $Ga_{1-x}Al_xN$ thin films (x = 0; 0.03; 0.07)

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The dispersion of the refractive index and the region of the fundamental absorption edge in $Ga_{1-x}Al_xN$ (x = 0; 0.03; 0.07) thin films obtained by high-frequency ion-plasma sputtering have been investigated. It is shown that the dispersion dependence of the obtained films has a complex form. It was found that the value of the optical band gap increases from 3.28 to 3.58 eV with increasing Al concentration from 0 to 7 mol%. Based on the determined direct-band allowed photoelectron transitions in the region of the fundamental absorption edge, the value of the combined effective mass of free charge carriers and their concentration was estimated. It is shown that the shift of the fundamental absorption edge in $Ga_{1-x}Al_xN$ thin films (x = 0, 0.03, 0.07) is due to the Burstein-Moss effect.

Keywords: gallium nitride, thin films, refractive index dispersion, fundamental absorption edge.

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Introduction

In recent years, thin films of gallium nitrides and oxides have been the subject of intensive research due to the prospects of their use in the creation of efficient sources of visible and near ultraviolet (UV) radiation, electroluminescent displays, and receivers of ionizing and UV radiation [1-7]. In addition, due to its unique parameters – large band gap, high charge carrier drift velocity, high breakdown voltage, and high chemical and thermal stability - gallium nitride GaN is a very promising material for creating new-generation microelectronic and nanoelectronic devices [8-10]. In general, the optical and electrical characteristics of GaN thin films are determined by the manufacturing methods, deposition modes, and the introduction of impurities that can purposefully change the properties of the films. In this work, $Ga_{1-x}Al_xN$ thin films (x = 0, 0.03, 0.07) were studied, in which some Ga^{3+} ions were replaced by Al^{3+} ions, which did not require local compensation of the electric charge. This is because AlN thin films are also quite promising in terms of their use in micro- and nanoelectronics, optoelectronics, and luminescent technology [11-13]. The study of the optical properties of such films seems to be quite relevant. In particular, the value of the refractive index determines the resolution of the films, and the edge absorption provides important information about the energy structure of the films. In this regard, the present work investigates the dispersion properties and the fundamental absorption edge of $Ga_{1-x}Al_xN$ thin films obtained by radio-frequency (RF) ion-plasma sputtering, which is optimal for producing both multicomponent semiconductor and dielectric films [14].

I. Methodology of the experiment

$Ga_{1-x}Al_xN$ (x = 0; 0.03; 0.07) thin films with a thickness of 0.3 -1.0 μm were obtained by RF ion-plasma sputtering on sapphire substrates (Al_2O_3). The RF sputtering was carried out in a nitrogen atmosphere at pressures from $5\cdot10^{-3}$ to $5\cdot10^{-2}$ Torr. The target for sputtering was metallic Ga. The temperature of the substrates during sputtering was 600°C, and the RF discharge power was 100 W.

The structure and phase composition of the obtained films were studied by X-ray diffraction analysis (Shimadzu XDR-600). X-ray diffraction studies showed the presence of a polycrystalline structure with a predominant orientation in the (002) plane.

Diffractograms of all types of the studied films are similar and coincide with the diffractograms of pure GaN films, which are presented and analyzed in [15].

The samples were analyzed using an OXFORD INCA Energy 350 energy dispersive spectrometer. The studies were performed at several points on the surface of the thin films. The calculations confirmed that the percentage of components in the studied films corresponds to the percentage in the formula $Ga_{1-x}Al_xN$ (x = 0, 0.03, 0.07).

The thicknesses and optical constants of the thin films were determined based on the interference pattern in the transmission spectra according to the method [16]. The transmission spectra were measured on a CM 2203 spectrofluorimeter with a Hamamatsu R928 measuring head.

II. Results and discussion

The characteristic transmission spectra for the obtained Ga1-xAlxN thin films are shown in Fig. As can be seen from Fig. 1, a clear interference pattern is observed in the transmission spectra, based on which the thicknesses and optical constants were calculated according to Swanepoel's method [16].

According to this technique, the refractive index can be calculated to the first approximation by Eq:

$$n = \left[N + (N^2 - n_n^2)^{1/2}\right]^{1/2} \quad (1)$$

$$N = \frac{(n_n^2+1)}{2} + 2n_n \frac{(T_{max}+T_{min})}{T_{max}T_{min}} \quad (2)$$

where T_{max} and T_{min} are the maximum and minimum transmittance values at a certain wavelength, where the minimum transmittance is observed, and the maximum for a given wavelength is calculated from the envelope curve, n_n is the refractive index of the substrate.

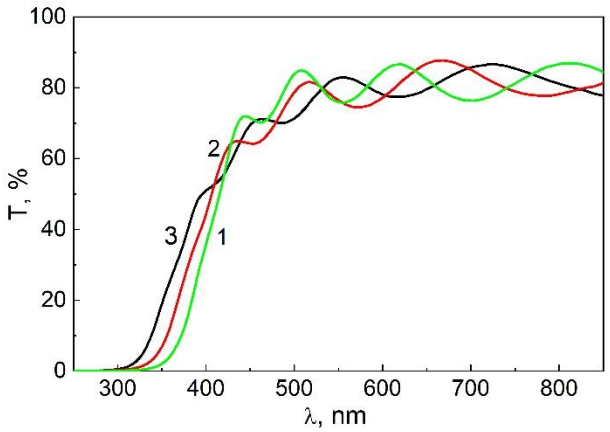

Fig. 1. Transmission spectra of $Ga_{1-x}Al_xN$ thin films at x = 0 (1), x = 0.03 (2), x = 0.07 (3), T = 295 K.

In addition, if n_1 and n_2 are the refractive indices for two adjacent maxima (or minima) at wavelengths λ_1 and λ_2, then the film thickness is determined by the expression:

$$d = \frac{\lambda_1\lambda_2}{2(\lambda_1 n_2 - n_1\lambda_2)} \quad (3)$$

The absorption coefficient is calculated by the ratio:

$$k = \frac{\alpha\lambda}{4\pi} \quad (4)$$

where α is the absorption coefficient.

$$\alpha = \frac{1}{d}\ln\frac{(n-1)(n-n_n)\left[(T_{max}/T_{min})^{1/2}+1\right]}{(n+1)(n+n_n)\left[(T_{max}/T_{min})^{1/2}-1\right]} \quad (5)$$

When using the methodology of [16], according to the results of [17], the error in determining the refractive index *n* does not exceed 2 %, and the error in determining the absorption index *k* does not exceed 1 %.

The dispersion dependences n(λ) for the obtained films are shown in Fig.2. As can be seen from Fig. 2, these dependencies are complex. In particular, for GaN and $Ga_{0.97}Al_{0.03}N$ films, there are areas with normal and anomalous dispersion, the forms of dependence are very similar to each other with close values of the refractive index n. This may indicate that the concentration of Al impurity up to x=3 mol% has little effect on the spectral dependence of n(λ) in GaN thin films and the properties of the substrate itself determine it. At higher concentrations of Al impurity, in particular, at x = 7 mol%, this dependence changes, and in the spectral range of 480-860 nm, we observe an anomalous dispersion of n(λ).

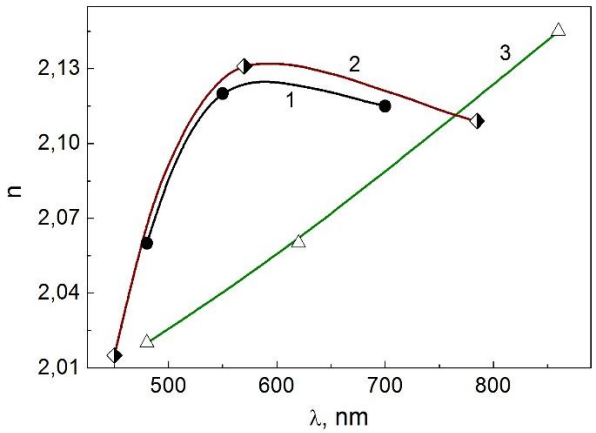

Fig. 2. Refractive index dispersion of $Ga_{1-x}Al_xN$ thin films at x = 0 (1), x = 0.03 (2), x = 0.07 (3), T = 295 K.

Based on the fact that in the region of strong (interband) absorption, interference in the transmission spectra is practically absent, the relation [16] was used to determine the absorption coefficient of thin films α(*hν*):

$$\alpha(h\nu) = -\frac{1}{d}\cdot\ln\left[\frac{(n(h\nu)+1)^3(n(h\nu)+n_n^2)\cdot T}{16n_n\cdot(n(h\nu))^2}\right], \quad (6)$$

where *d* is the value of the film thickness determined by the method [16]; *T* is the transmittance; n and np are the refractive indices of the film and substrate, respectively. The required values of n in the calculation of α(*hν*) were determined based on the obtained dependences of *n*(*hν*) in the transparency and weak absorption region according to the method [16] by extrapolating to this frequency region. The determination of such dependences on the example of β- Ga_2O_3 films was described in detail in [18].

As a result of the studies, it was found that for all types of $Ga_{1-x}Al_xN$ films, the absorption coefficient $\alpha(h\nu)$ in the region of the fundamental absorption edge is described by the expression

$$\alpha(h\nu) = \frac{A(h\nu - E_g)^{1/2}}{h\nu} \qquad (7)$$

which makes it possible to determine the width of the forbidden zone E_g. The obtained dependences $(\alpha \times h\nu)^2 = f(h\nu)$ are shown in Fig. 3.

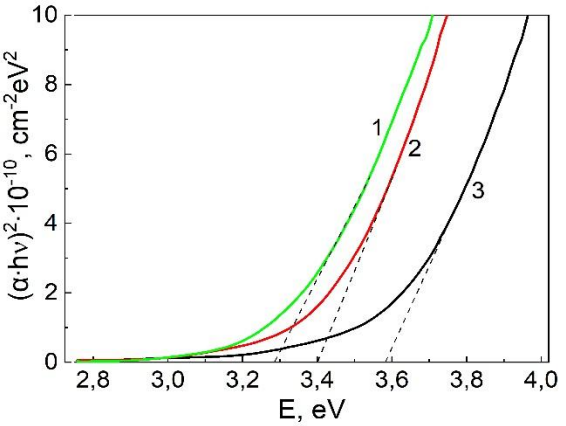

Fig. 3. The fundamental absorption spectrum in the coordinates $(\alpha \times h\nu)^2 = f(h\nu)$ for $Ga_{1-x}Al_xN$ thin films at x = 0 (1), x = 0.03 (2), and x = 0.07 (3).

The obtained shape of the fundamental absorption edge indicates that in $Ga_{1-x}Al_xN$ thin films it is formed by allowed direct phototransitions of electrons [19].

Analyzing the relation (7), which describes the edge of the fundamental absorption, the optical band gap in $Ga_{1-x}Al_xN$ thin films increases with increasing *x*. At the same time, no dependence of the value of the coefficient A in relation (7) on the change in the value of *x* was found. The characteristic values of the determined values of E_g and *A* for the studied films are given in Table 1.

The obtained results show that a change in the stoichiometry of GaN thin films due to the introduction of an AlN impurity leads to an increase in the optical band gap E_g in thin films, and the magnitude of the increase is proportional to the concentration of the impurity. This situation is in good agreement with the known results of studies of Ga and Al nitrides, which found that in $Ga_{1-x}Al_xN$ compounds when x changes from 0 to 1, the E_g value increases from 3.5 to 6.28 eV [20, 21].

The increase in the energy of the bandgap E_g with increasing impurity concentration and, accordingly, an increase in the number of structural defects and the concentration of current carriers can be explained based on the Burstein-Moss effect [22]. According to this effect, the fundamental absorption edge spectrum of heavily doped or highly defected (degenerate) semiconductors is similar to the absorption edge spectrum of non-degenerate semiconductors but shifted to higher energies. As can be seen from Fig. 3, this situation is realized in $Ga_{1-x}Al_xN$ thin films, which, with increasing *x*, can be considered as GaN thin films heavily doped with an AlN impurity. At the same time, an increase in the value of *x* leads to an increase in the defectiveness of the films.

Taking into account, according to (7), the presence of direct allowed electron phototransitions for the parabolic conduction and valence bands, we can write [23]

$$E_g = E_{g0} + \Delta E_g^{\text{Б-М}}, \qquad (8)$$

where $E_{g0}$is the intrinsic band gap of the undegenerate semiconductor, and $\Delta E_g^{Б-М}$ is the Burstein-Moss shift caused by the filling of the lower energy levels in the conduction band. The value of the Burstein-Moss shift is defined as

$$\Delta E_g^{\text{Б-М}} = (h^2/8\pi^2\mu)(3\pi^2 N)^{2/3} \qquad (9)$$

where *N* is the concentration of free charge carriers, and μ is their combined effective mass. It follows from equation (9) that the value of the Burstein-Moss shift $\Delta E_g^{\text{Б-М}}$ is proportional to the concentration of free charge carriers. To estimate the concentration of free charge carriers, we first determine the value of the combined effective mass of free charge carriers based on the spectra of the fundamental absorption edge.

It is known [24] that in direct-band compounds, at the electronic transition between the valence and conduction bands, the spectral course of the absorption coefficient is given by a more detailed expression (7) in the form:

$$\alpha(h\nu) = \frac{2e^2(2\mu)^{3/2}}{m^2 c\hbar^2 n}\left|P_\mu\right|^2 \frac{(h\nu - E_g)^{1/2}}{h\nu}, \qquad (10)$$

where $\left|P_\mu\right|^2$ is the square of the matrix element of the dipole transition; n is the refractive index in the absorption edge region.

Expressing $|P_\mu|^2$ through the oscillator strength f_μ of the interband transition:

$$\left|P_\mu\right|^2 = \frac{mh\nu}{2} f_\mu \qquad (11)$$

Taking into account the presence of allowed transitions, we get $f_\mu \approx 1$. As a result, we get the ratio:

$$\alpha \approx \frac{(2\mu)^{3/2}e^2}{mc\hbar^2 n}\left(h\nu - E_g\right)^{1/2}. \qquad (12)$$

Table 1.

The value of the band gap E_g and the coefficient *A* in relation (7), the summed effective mass μ, and the concentration of free charge carriers *N* in $Ga_{1-x}Al_xN$ thin films

Thin film	E_g, eV	A, $cm^{-1}\times eV^{-1/2}$	μ	N, cm^{-3}
GaN	3.28	3.98×10^7	$0.058m$	
$Ga_{0.97}Al_{0.03}N$	3.40	3.65×10^8	$0.133m$	9.38×10^{17}
$Ga_{0.93}Al_{0.07}N$	3.58	3.74×10^8	$0.198m$	6.75×10^{18}

Substituting the numerical values using the straight-line section of the dependence $(\alpha \times h\nu)^2 = f(h\nu)$ (Fig. 3), we can estimate the total effective mass of free charge carriers in $Ga_{1-x}Al_xN$ thin films. As a result, for GaN films we obtain $\mu \approx 0.058$ m, for $Ga_{0.97}Al_{0.03}N$ films we have $\mu \approx 0.133$ m, and for $Ga_{0.93}Al_{0.07}N$ films $-\mu \approx 0.198$ m. The obtained increase in the value of the combined effective mass after doping GaN films with an AlN impurity can be explained by an increase in the concentration of impurity and defect centers on which free charge carriers can localize. The obtained values of the consolidated effective mass of free charge carriers for $Ga_{1-x}Al_xN$ thin films are shown in Table 1.

It should be noted that the values of μ obtained by us are in good agreement with a number of previous studies. According to [25], in epitaxial InN films, the value of μ is 0.050 m, in epitaxial GaN:Si films [26], μ is 0.200 m, in $Ga_{0.28}Al_{0.72}N$ films [27], μ is 0.336 m. The determined values for β–Ga_2O_3 films [28] are $\mu = 0.281m$, and for β–Ga_2O_3 single crystals [29] $\mu = (0.276\text{-}0.311)m$. The calculated effective masses of free charge carriers in GaN single crystals according to [21] are $m_\perp = 0.18m$, and $m_\parallel = 0.16m$.

By estimating the value of the combined effective mass of free charge carriers in $Ga_{1-x}Al_xN$ thin films and experimentally measuring the value of the band gap shift using Eq. (9), we estimate the value of the concentration of free charge carriers N. Based on the calculations, we obtain for $Ga_{0.97}Al_{0.03}N$ thin films the value of $N \approx 9.38\times10^{17}$ cm^{-3} and for $Ga_{0.93}Al_{0.07}N$ thin films the value of $N \approx 6.75\times10^{18}$ cm^{-3}. The values obtained of N correlate well enough with previous studies. According to [20], in GaN $N = 2.08\times10^{18}$ cm^{-3}. According to [26], in epitaxial GaN films, the value of $N = 1\times10^{17}$ cm^{-3}, in GaN:Si films with a thickness of 1 μm $N = 5\times10^{17}$ cm^{-3}, and with a thickness of 1.5 μm $N = 5\times10^{18}$ cm^{-3}. In thin films of β–Ga_2O_3:Y, depending on the heat treatment conditions, the value of $N = (1.32\text{–}5.20)\times10^{18}$ cm^{-3} [28].

It is known [30] that in heavily doped, degenerate semiconductors, the concentration of charge carriers varies from 10^{16} cm^{-3} and can reach up to 10^{20} cm^{-3}. The values of N obtained by us indicate that the estimated values of the concentration of free charge carriers in the studied $Ga_{1-x}Al_xN$ films are typical for degenerate semiconductors in which the Burstein-Moss effect is observed. This confirms the validity of using this effect for $Ga_{1-x}Al_xN$ thin films when replacing a part of Ga^{3+} ions with Al^{3+} ions in GaN thin films, when the edge of the fundamental absorption shifts to the high-energy region. As we have shown earlier, a similar situation is observed for $(Y_{0.06}Ga_{0.94})_2O_3$ thin films, when a part of Ga^{3+} ions was replaced by Y^{3+} ions in gallium oxide β–Ga_2O_3 [28] and is explained based on the Burstein-Moss effect.

Conclusions

The studies have shown that in $Ga_{1-x}Al_xN$ (x = 0, 0.03, 0.07) thin films obtained by radio-frequency ion-plasma sputtering, the dispersion dependence has a complex appearance, and the edge of the fundamental absorption is formed by direct allowed electron phototransitions. It is found that the optical band gap increases from 0 to 7 mol% with increasing Al concentration from 28 to 3.58 eV. The total effective mass of free charge carriers and their concentration were estimated, and it was shown that the shift of the fundamental absorption edge in $Ga_{1-x}Al_xN$ thin films (x = 0, 0.03, 0.07) is due to the Burstein-Moss effect.

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Дисперсія світла та крайове поглинання в тонких плівках $Ga_{1-x}Al_xN$ (x = 0; 0.03; 0.07)

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Досліджено дисперсію показника заломлення та область краю фундаментального поглинання в тонких плівках $Ga_{1-x}Al_xN$ (x = 0; 0.03; 0.07), отриманих методом високочастотного іонно-плазмового розпилення. Показано, що дисперсійна залежність отриманих плівок має складний вигляд. Встановлено, що величина оптичної ширини забороненої зони при зростанні концентрації Al від 0 до 7 мол.% зростає від 3,28 до 3,58 еВ. На основі встановлених прямо зонних дозволених фотопереходів електронів в області краю фундаментального поглинання проведено оцінку величини зведеної ефективної маси вільних носіїв заряду та їхньої концентрації. Показано, що зсув краю фундаментального поглинання в тонких плівках $Ga_{1-x}Al_xN$ (x = 0; 0,03; 0,07) зумовлений ефектом Бурштейна-Мосса.

Ключові слова: нітрид галію, тонкі плівки, дисперсія показника заломлення, край фундаментального поглинання.