# Optical properties of $Al_xGa_{1-x}N$/GaN heterostructures on sapphire by spectroscopic ellipsometry

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A method of analysis of spectroscopic ellipsometry (SE) measurement data is proposed for $Al_xGa_{1-x}N$/GaN heterostructures grown on sapphire substrates. The SE data measured at three angles of incidence, 40°, 50°, and 60°, are simultaneously fitted assuming the dielectric function to consist of a Sellmeir dispersion equation and a free-exciton absorption term. The refractive index $n$ and the extinction coefficient $k$ of undoped $Al_xGa_{1-x}N$ films are determined in the spectral range of 1.5–4.13 eV of photon energy. The transition energy of the free exciton, which is in excellent agreement with the reported results for GaN in a previous paper, is found to vary from 3.44 to 3.95 eV when the composition $x$ varies from 0 to 0.151. The refractive index $n$ of $Al_xGa_{1-x}N$ has also been compared with those reported results. © *1998 American Institute of Physics.* [S0003-6951(98)01018-3]

Recently, light-emitting diodes based on InGaN/AlGaN heterostructures grown on sapphire substrates have achieved practical intensity levels[1] and electrically pumped III–V nitride lasers have also been reported.[2] But, these materials and their related optoelectrical devices are still in the preliminary stages, and many physical parameters related to the design of new optoelectronic devices are not understood in detail.

Spectroscopic ellipsometry (SE) is a nondestructive and powerful technique to investigate the optical response of materials, and in particular, to measure simultaneously the thickness and the dielectric function of a multilayer system.[3] Recently, we have determined the optical constants of GaN by using the simple Sellmeir dispersion relationship to fit the SE measured data and have found an evident free exciton at room temperature.[4] Further, we have found that the difference in the refractive indices for $n_\perp(\mathbf{E}\perp c)$ and $n_\parallel(\mathbf{E}\parallel c)$ is smaller than 3% over the wavelength range of 400–830 nm,[5] and thus ignoring this difference would not affect results much when SE measurements are taken at small incident angles. However, a heterostructure of $Al_xGa_{1-x}N$/GaN contains more unknown parameters. For this, more accurate modeling analysis of the dielectric function $\epsilon(\omega)$ and more measured data are required.

In this letter, we first use the sum of a Sellmeir dispersion relationship and a free-exciton absorption term to describe the dielectric function of $Al_xGa_{1-x}N$ below the fundamental band edge and assume a two-layer model for samples of $Al_xGa_{1-x}N$/GaN/sapphire. An excellent simultaneous fit of the model to measured $\Psi$ data of SE at three angles of incidence, 40°, 50°, and 60°, has been obtained.

$Al_xGa_{1-x}N$/GaN single heterostructures were grown on 2-in.-sapphire (0001) substrates using the horizontal atmospheric pressure metal–organic chemical-vapor deposition method. Trimethylgallium (TMG), trimethylaluminum (TMA), and ammonia ($NH_3$) were used as source materials. The sapphire substrate was first heated at 1100 °C for 10 min in a stream of hydrogen followed by the deposition of 30-nm-thick GaN as the buffer layer at 530 °C. Then, a GaN layer with a thickness of about 1.5 $\mu$m and an $Al_xGa_{1-x}N$ layer with a thickness in the range of 0.2–0.5 $\mu$m were grown at a fixed temperature of 1050 °C. The molar fractions of AlN were determined by electron-probe microanalysis and the thicknesses of the films were determined by both high-resolution scanning electron microscopy (SEM) and spectroscopic ellipsometry. The automatic ellipsometry used was of the rotating analyzer type. A 75 W xenon lamp was used as the light source, and the SE measurements were carried out in air over a 300–830 nm wavelength range with a step of 2 nm at three angles of incidence, 40°, 50°, and 60°. Details concerning the structure of three samples studied are listed in Table I.

In our previous work[5] we found that the refractive index of GaN below the fundamental band-gap energy can be adequately described by a Sellmeir dispersion equation and also we observed a clear free-exciton absorption in high-

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TABLE I. Best-fit parameters of $Al_xGa_{1-x}N$/GaN on sapphire substrates determined by SE measurements and SEM thicknesses measurements.

| Sample No. | Structure | $d_1$ ($Al_xGa_{1-x}N$) ($\mu$m) | | $d_2$ (GaN) ($\mu$m) | | The parameters of a Sellmeir dispersion equation | | | The parameters of a free-exciton absorption term | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | SE | SEM | SE | SEM | A | B | $\omega_a$ (eV) | $\alpha_e$ | $\omega_e$ (eV) | $\Gamma_e$ (meV) | $\sigma$ |
| 1 | GaN/sapphire | 0 | / | 1.396 | / | 3.70 | 29.85 | 4.65 | 0.00253 | 3.44 | 35 | 0.167 |
| 2 | $Al_{0.087}Ga_{0.913}N$/GaN /sapphire | 0.225 | 0.2 | 1.532 | 1.5 | 3.00 | 59.64 | 5.57 | 0.00254 | 3.73 | 22 | 0.159 |
| 3 | $AlGa_{0.849}N$/GaN /sapphire | 0.310 | 0.3 | 1.552 | 1.5 | 2.96 | 58.64 | 5.59 | 0.00321 | 3.95 | 78 | 0.232 |

quality GaN at room temperature. So, in order to describe the dielectric function of $Al_xGa_{1-x}N$ more accurately, the sum of the Sellmeir dispersion equation and the exciton expression has been used to fit the SE data, which can be expressed by the following equation:

$$\epsilon(\omega)=A+\frac{B\omega^2}{\omega_a^2-\omega^2}+\frac{4\pi\alpha_e\omega_e^2}{\omega_e^2-(\hbar^2k^2/m^*)\omega_e-\omega^2-i\Gamma_e\omega}. \tag{1}$$

Here, the first two terms together are called the first-order Sellmeir dispersion equation[6] and the third term is the contribution from the exciton, which has been used to analyze the exciton of GaN in reflection spectra by Tchounkeu *et al.*[7] $\omega_e$ is the transverse frequency related to the exciton with an effective mass $m^*$, $4\pi\alpha_e$ is the polarizability of the exciton resonance at $\omega=0$ and $k=0$, and $\Gamma_e$ is the damping parameter used to account for the interactions of the exciton with the phonons and extrinsic defects. It can be mentioned here that the SE parameter $\Psi$ is not very sensitive to the presence of a few thin overlayers on the surface, such as an oxide or roughness layer, and therefore, the comparison between the model and the experiment is made on the $\Psi$ curve only in order to ignore the effect of such overlayers in the fitting.[8] In addition, a simultaneous fit to the spectra of $\Psi$, measured at three angles of incidence (40°, 50°, and 60°) was carried out to further increase the reliability of the parameters obtained by fitting. The root-mean-square fractional error $\sigma$, defined by

$$\sigma^2=\frac{1}{3}\left[\frac{\Sigma_j^n\{(\Psi_j^{\exp}-\Psi_j^{\mathrm{cal}})_{iang=40}\}^2}{\Sigma_j^n\{(\Psi_j^{\exp})_{iang=40}\}^2}+\frac{\Sigma_j^n\{(\Psi_j^{\exp}-\Psi_j^{\mathrm{cal}})_{iang=50}\}^2}{\Sigma_j^n\{(\Psi_j^{\exp})_{iang=50}\}^2}+\frac{\Sigma_j^n\{(\Psi_j^{\exp}-\Psi_j^{\mathrm{cal}})_{iang=60}\}^2}{\Sigma_j^n\{(\Psi_j^{\exp})_{iang=60}\}^2}\right], \tag{2}$$

has been used as judgment of the quality of the fit between the measured and calculated data, which has also been used by Kim *et al.*[9] in simultaneous fits to the dielectric function $\epsilon(\omega)$ and its derivatives with respect to the energy for GaAs.

We first attempt to fit the data of sample 1 using the above-mentioned method, assuming a three-phase (ambient/GaN/sapphire) model. The refractive index function of sapphire was taken from Ref. 10. The parameters obtained by calculating the $\Psi$ spectrum for sample 1 are shown in Table I. An excellent agreement between the measured and calculated spectra has been obtained. The refractive index function of sample 1 in the wavelength range of 300–830 nm, along with the refractive index of GaN taken from the previous report, is shown in Fig. 1. It is obvious that there is no appreciable difference between the two results over the whole wavelength range measured. It can be noted here that for all samples the refractive index $n$ and extinction coefficient $k$ in the wavelength region above the band gap were obtained by assuming a two-phase (such as ambient/GaN) model, and then fitting for $n$ and $k$ at each wavelength. This assumption of a two-phase model is justified by two reasons. First, the penetration depth of light from the surface is about 100 nm in the wavelength range above the band gap. Since, the top layers for all the samples are thicker than 100 nm, the effect of the bottom layer and/or substrate is negligible in the above wavelength region. Second, not considering the overlayers on the surface does not deteriorate the fitting of SE spectra $\Delta$ below the fundamental band edge (see Ref. 4). Therefore, it is reasonable to estimate the refractive index $n$ and the extinction coefficient $k$ of the films at above the band gap in order to compare the results.

The other two fits to the data of samples 2 and 3 are also

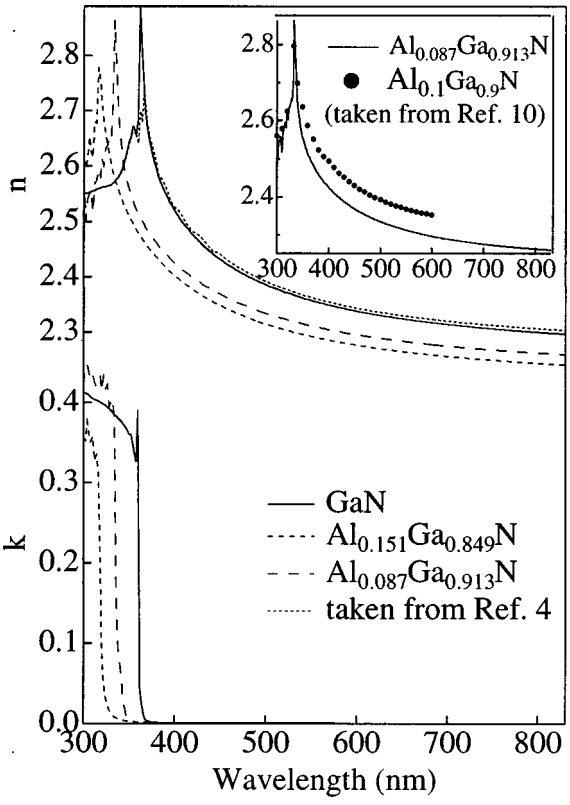


FIG. 1. The optical constants of $Al_xGa_{1-x}N$ vs wavelength. (a) The refractive indices determined by SE. (b) The extinction coefficient $k$. The refractive indices of sample 2 and those from Ref. 10 are shown in the inset.

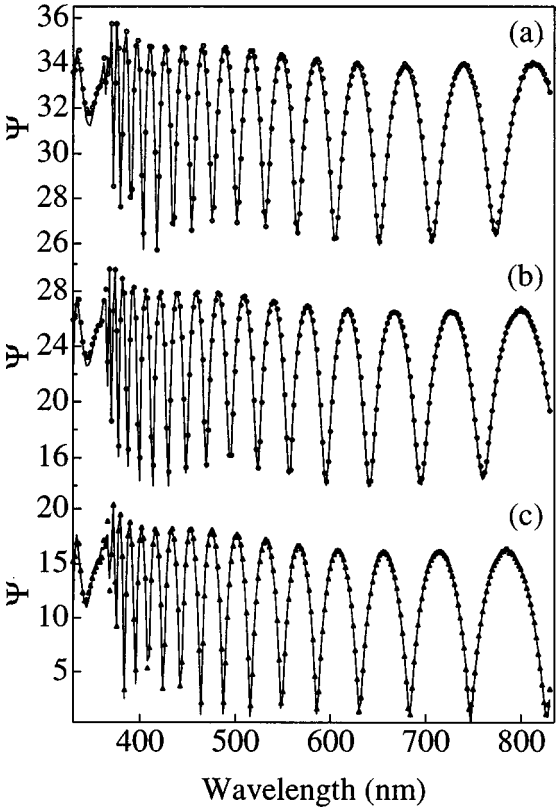


FIG. 2. Measured (dotted line) and calculated (solid line) $\Psi$ spectra of $Al_{0.087}Ga_{0.913}N/GaN$ on sapphire substrates, for incident angles of (a) 40°, (b) 50°, and (c) 60°, respectively.

performed assuming the four-phase (ambient/$Al_xGa_{1-x}N$/GaN/sapphire) model. For GaN, the refractive index function $(n+ik)$ obtained by the fitting analysis for sample 1, which is also shown in Fig. 1, has been used. In Fig. 2 the comparison between the measured and calculated data is shown for sample 2, and the parameters of the two samples, 2 and 3, are summarized in Table I. The refractive indices $n$ and extinction coefficient $k$ as a function of photoenergy are also calculated for both the samples, 2 and 3, and the data are plotted in Figs. 2(a) and 2(b) as dashed lines; in the inset of the same figure the refractive index function of sample 2 ($Al_{0.087}Ga_{0.913}N$) and that of $Al_{0.1}Ga_{0.9}N$ taken from Ref. 11 are shown for comparison. Our refractive index is found to be 2% smaller than that from Ref. 11, this deviation perhaps arises from the two distinct experimental methods. We have further calculated SE data $\Delta$ using the refractive index $(n+ik)$ and thicknesses of each layer. It is observed that the difference between the measured and calculated data is very small. This indicates that our samples have good surface morphology and thin oxide films.

As shown in Table I, the thicknesses obtained by SE and SEM are very close. Therefore, it is proved that our model adequately describes the measured data. The exciton energies have been obtained from Eq. (1), where the term $(\hbar^2k^2/m^*)\omega_e$ has been neglected because of its extremely small value compared to term $\omega^2$. The energy value of sample 1 obtained is in agreement with our previous result for GaN. The exciton energy is also found to increase with composition $x$.

In summary, we have shown that the optical constants of $Al_xGa_{1-x}N$ can be accurately expressed as the sum of a Sellmeir dispersion equation and a free-exciton absorption term below the fundamental band edge. The free-exciton energy of $Al_xGa_{1-x}N$ has been obtained by simultaneously fitting to SE data $\Psi$ measured at three angles of incidence, 40°, 50°, and 60°. The values obtained vary from 3.44 to 3.95 eV when the composition $x$ varies from 0 to 0.151. The results reveal the prospect of determining parameters of the model as a function of composition $x$ for $Al_xGa_{1-x}N$ and the work is in progress in our group.

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