ABSORPTION COEFFICIENT AND REFRACTIVE INDEX OF GaN, AlN AND AlGaN ALLOYS

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ABSTRACT

The design of optoelectronic devices fabricated from III-nitride materials is aided by knowledge of the refractive index and absorption coefficient of these materials. The optical properties of GaN, AlN and AlGaN grown by MOVPE on sapphire substrates were investigated by means of transmittance and reflectance measurements. Thin (less than 0.5 μm) single crystal films were employed to insure that transmission measurements could be obtained well above the optical band gap. The influence of alloy broadening on the absorption edge was investigated by using a series of AlGaN alloy samples with a range of Al compositions. The optical absorption coefficient above the band gap was obtained for AlGaN having up to 38% Al composition. The refractive index below the band gap was determined for the same series of samples. These properties provide information critical to the optimal design of solar blind detectors or other optoelectronic devices.

INTRODUCTION

The III-nitrides, including GaN, InN, and AlN, have proven to be robust materials for the development of blue/green LEDs and violet lasers lasers [1]. The AlGaN alloys also hold great promise for developing ultraviolet photodetectors [2]. In photodetector applications, the wide band gap of the nitrides aids to minimize the dark current, thereby increasing the detector sensitivity. The transparency to visible light will reduce the need for the extensive external filters that are required when detectors sensitive to the visible portion of the spectrum are employed. Multilayered Bragg mirrors and filters fabricated from these materials also have applications for lasers and photodetectors. Currently there has been only a limited amount of information reported [3,4] about the fundamental optical properties of AlGaN alloys which makes design of these devices difficult. The relative immaturity of this materials system also means that there may be some variance in the properties of the materials produced by different workers. In this paper, we report the refractive index and absorption coefficient for AlGaN thin film compositions of up to 38 % obtained by means of reflectance/transmittance spectroscopy.

For light below the band gap of the semiconductor, the interference within the thin film modulates both the reflection and transmission spectrum. Above the band gap, the high absorption coefficient causes the film to absorb any multiple reflections of the light. For these materials, unless the film is less than ~ 1 μm thick, there is seldom enough signal for commercial spectrophotometers to obtain an accurate ratio measurement. Due to the short penetration depth of the light above the band gap, reflection measurements become more dependent on the surface

condition of the semiconductor, but have been used with great success to gather information about band structure.

In the spectral region around the band gap the situation is complicated, for in addition to going from a transparent to an absorbing Fabry-Perot cavity, the excitonic structure becomes important. This is especially true in the case of nitride materials, where the binding energy of the exciton is strong (> ~20 meV). The three closely spaced valence bands lead to three strong absorption features, the A, B, and C exciton. The selection rules also make the transmission and reflection polarization dependent. The birefringence of the hexagonal material and the scattering from the columnar grain structure found in most nitride materials further complicates matters. In semiconductor alloys, fluctuations in composition broaden the exciton making the absorption features less distinct. However, the continuum absorption of the exciton still influences the absorption spectra, even when the excitonic features are no longer clearly visible. In general, all nitride epitaxial films are also highly strained, with strain being dependent on substrate material and on buffer layer.

In general, the traditional incident plane wave approach to thin film transmission and reflection problems assumes that the films are of uniform thickness and have a flat surface morphology. In practice, this is a challenge especially when the crystal grower would like to have a reasonable growth rate and also has to dope the film for electrical properties.

In the case of thin films that have surface roughness, some of the light is scattered and misses the detector. This lowers the experimental values of transmittance and reflectance with the effect being more pronounced at shorter wavelengths. Even a small error in the experimental values can lead to large errors in the determining the index of refraction [5]. The scattering also disrupts the phase of the plane wave leading to a loss of coherence in the thin film and a dampening of the modulation of the transmission and reflection spectra. Swanepoel [6,7], Szcxyrbowski [8], and Nowak [9] have considered the effects of surface roughness and thickness variations on the optical spectrum of thin films and their models were incorporated into the present model.

Another factor to consider in these thin film models is that the solution contains an index-thickness product (nd). Thus, the index of refraction and the thin film thickness are intimately related. In very thin films such as those used in this study, although the order number of the interference peaks is known, only a few data points can be obtained for the Swanepoel procedure for determining the thickness. With the film thickness known experimentally to ~ 7% from SEM measurements, the index of refraction of the films was determined using the same method as Ambacher [3]. The absorption coefficient was found using the reflectance data and the incoherent multilayer model by Wemple [10].

The exciton theory of Elliott [11] was used to fit the absorption coefficient data to find the optical band gap. Even with the exciton dissociated, it has a strong influence upon the absorption spectra that non-excitonic models can not fit [12,13]. Since the individual excitons could not be observed for alloy compositions above 5 % due to alloy broadening, for practicality the three excitons were grouped into one parameter and assigned a single binding energy.

EXPERIMENTAL DETAILS

The GaN, AlGaN, and AlN epitaxial films measured in this work were prepared in a low pressure, vertical flow, cold walled, high speed substrate rotation MOVPE reactor. This system was designed and built at NCSU and is integrated into a multi-chamber nitride growth cluster tool. The substrates for deposition were 2-inch diameter double-side-polished sapphire wafers. Sample preparation was as previously described [14]. Epitaxial films were deposited in a typical two step process, with a low temperature buffer layer to nucleate growth [15]. The low

temperature buffer layer was GaN for GaN growth, and AlN for AlGaN and AlN growth in order to minimize the effects of the buffer layer optical properties on the measurements of the epitaxial films. The epilayer growths of AlGaN and AlN films were performed at chamber pressures ranging from 20 to 76 Torr with the trimethylaluminum injected separately from the trimethylgallium source using hydrogen carrier gasses in both cases. Separate metallorganic injection and reduced pressures allow for the independent control of composition and velocity of the metallorganic species in order to limit parasitic reaction of the group III precursors [16]. With this technique, precise control of epilayer composition, uniformity, growth rate (0.5 – 1.5 μm/hr), and crystal quality was achieved.

The aluminum composition of the samples was estimated from the cathodoluminescence (CL) spectra. The CL was measured using a JEOL JSM-6400 SEM with an Oxford instruments mono-CL accessory. With this unit it is possible to obtain spectral CL scans from 180 nm to 900 nm, or to obtain monochromatic images within the available wavelength range. For these samples, an accelerating voltage of 5-10 kV was used with a beam current of 10 nA. The CL emission is from an interaction volume which extends through the sample into the substrate, including the lower quality buffer layer material.

The surface quality of the films was evaluated using differential interference contrast optical microscopy in order to anticipate any difficulties with light scattering from the surface.

Optical transmission intensity measurements were made from 200-3300 nm using a Cary 5E spectrophotometer. The reflectance measurements were taken from 250-900 nm with a Perkin-Elmer Lambda 9 spectrophotometer with a normal incidence reflectance accessory and calibrated uv-enhanced mirrors for use as references. The spectrophotometers were used in a double beam mode, where the optical energy passing through the samples is ratioed with the optical energy of a reference beam. No apertures were used in the reference and sample beams. This was very beneficial for making measurements above the band gap since it is experimentally difficult to obtain truly identical apertures and balance the instrument at very high absorbance. The spectrophotometer slit width was kept constant in the visible and UV portion of the spectrum to keep the spectral bandwidth constant. The spectral bandwidth was 0.1 nm to allow for observation of sharp excitonic features or closely spaced interference fringes. The double-side-polished substrates served to minimize scattering of the light.

RESULTS

The properties of the $Al_xGa_{1-x}N$ films analyzed in this study are summarized in Table 1.

Table 1: Thin $Al_xGa_{1-x}N$ films on double-polished sapphire.

Sample	1	2	3	4	5	6
CL Peak (eV)	3.40	3.76	3.96	4.11	4.24	5.88
x	0	0.18	0.27	0.34	0.38	1
Thickness (μm)	0.37	0.39	0.37	0.39	0.42	0.4
Optical Band Gap (eV)	3.43	3.80	4.00	4.18	4.36	6.20

Figure 1 shows the CL of the samples which was used to assign the alloy composition of the material using a bowing parameter of 1 eV [17]. The thin nature of the films and the relatively low accelerating voltage minimized the effect of self absorption upon the true peak position [18] although the peak position was still red shifted in relation to the optical band gap

found by fitting the absorption data. The CL indicated that the material was of high quality, although some deep level luminescence was observed.

The transmission data obtained for the sequence of thin samples is shown in Figure 2. The AlN layer demonstrates the influence of surface morphology upon transmission measurements. Due to scattering, the amplitude of the modulations is suppressed and the transmission steadily decreases with shorter wavelengths. The transmission results for the AlGaN nitride films ranging in aluminum composition from 0.18 to 0.38 (films 2-5) show that little scattering occurs and that the films are of uniform thickness. The thin GaN layer grown on a GaN buffer layer shows some unusual behavior. In addition to the excitonic band edge which produced a distinct room temperature excitonic feature indicative of high material quality, another absorption slope just below the excitonic edge was visible. This was distinct from the Urbach tail which normally occurs from phonon broadening of the exciton. Examination of the film by differential interference contrast optical microscopy revealed the presence non-uniform regions within the film. This increased the light scattering in the film and also influenced the effective refractive index.

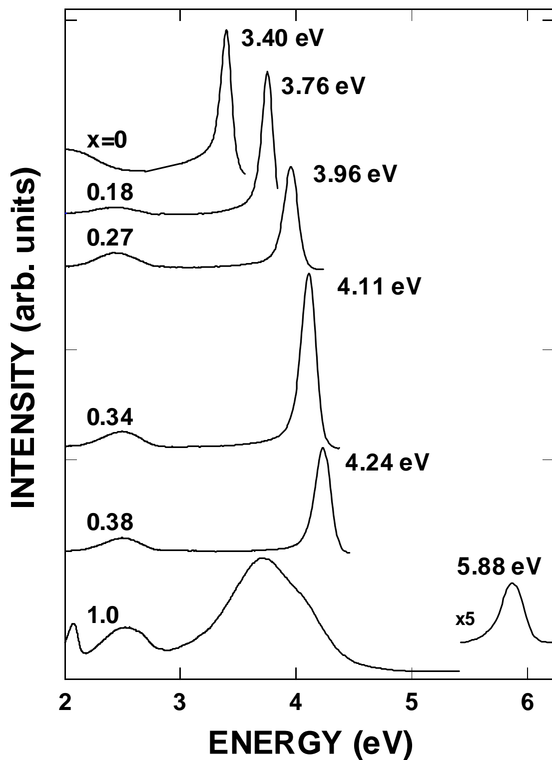

Figure 1. Cathodoluminescence for $Al_xGa_{1-x}N$ Samples

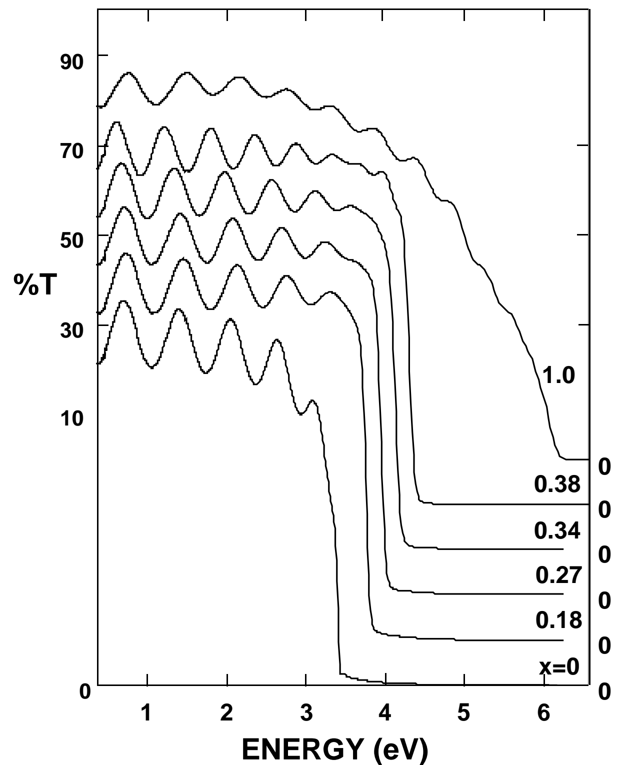

Figure 2. Transmission data for $Al_xGa_{1-x}N$ Sample

The index of refraction, shown in Figure 3, was computed by fitting the transmission data as described in the introduction. The values obtained were about 3 percent lower than those obtained by Ambacher [3]. This is perhaps due to the influence of the buffer layer on the effective index of the film.

The calculated absorption coefficients are plotted in Figure 4. The thin nature of the films and smooth surface morphologies permitted extraction of the absorption coefficient well

above the band gap. In the binary compound, the GaN exciton is clearly visible, and just below the excitonic band edge an absorption tail attributed to the buffer layer is visible. In the alloy films, x=0.27, 0.34, and 0.38, the exciton is suppressed by alloy broadening. The clearly recognizable AlN band edge is shown, but due to the excessive scattering it was not possible to obtain accurate values for the absorption coefficient above 1.5 x 10^5 cm-1.

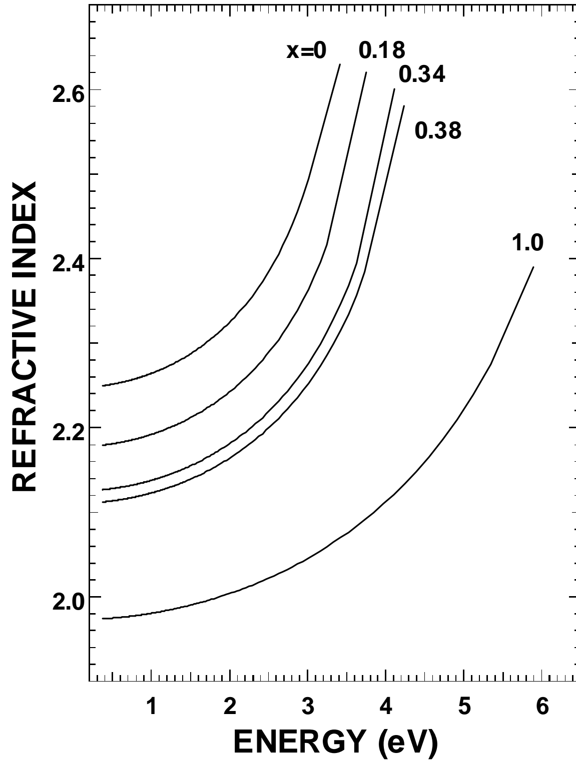

Figure 3. Below band gap refractive index for $Al_xGa_{1-x}N$

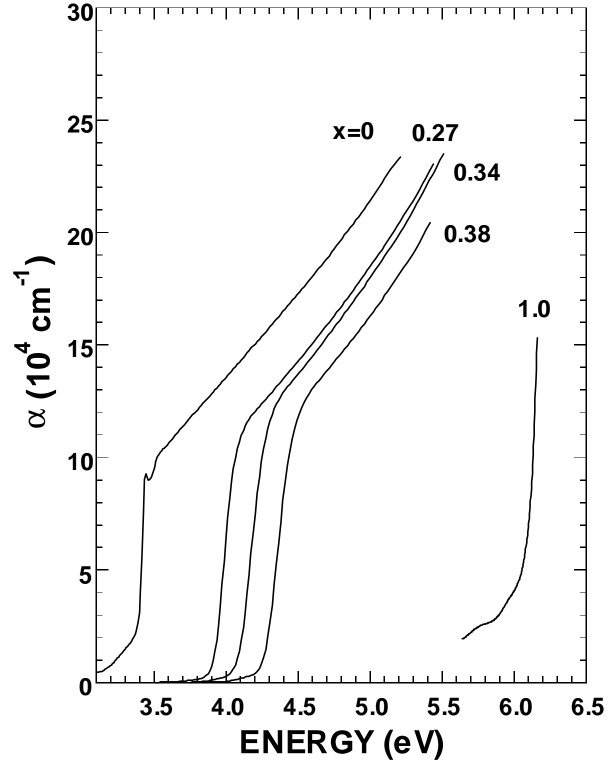

Figure 4. Absorption coefficient determined from transmission measurements

CONCLUSIONS

We have investigated the optical properties of GaN, AlGaN alloys and AlN using scanning electron microscopy cathodoluminescence along with reflectance and transmission spectroscopy. The films used were less than 0.5 μm in thickness which permits direct calculation of the absorption coefficient well above the band gap. The index of refraction below the band gap was obtained by fitting the transmission spectra, and the optical band gap was obtained by fitting the transmission data with Elliott's theory of absorption. The absorption coefficient and index of refraction show reasonable agreement with those determined by other workers. These results provide further information of importance toward the design optimization of optoelectronic devices employing the III-nitrides.

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