2001

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Systematic measurement of Al$_x$Ga$_{1-x}$N refractive indices

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(Received 13 August 2001; accepted for publication 5 October 2001)

Dispersion of the ordinary and extraordinary indices of refraction have been measured systematically for wurtzitic Al$_x$Ga$_{1-x}$N epitaxial layers with 0.0$\leq x$,$\leq$1.0 throughout the visible wavelength region. The dispersion, measured by a prism coupling waveguide technique, is found to be well described by a Sellmeier relation. Discrepancies among previous measurements of refractive index dispersion, as a consequence of different growth conditions and corresponding band gap bowing parameters, are reconciled when the Sellmeier relation is parameterized not by $x$ but by band gap energy. © 2001 American Institute of Physics. [DOI: 10.1063/1.1426270]

Optoelectronic devices based on group-III nitrides (GaN, In$_x$Ga$_{1-x}$N, and Al$_x$Ga$_{1-x}$N) have been actively developed for short wavelength emitters and detectors in the green through the near ultraviolet spectral regions. The wurtzite group-III nitrides lack cubic symmetry and therefore have anisotropic optical properties. The anisotropy results in uniaxial birefringence, two different refractive indices for polarization parallel ($n_o$) and perpendicular ($n_e$) to the c axis. To date, refractive index dispersion has been measured in a variety of ways (ellipsometry, interferometry, and prism coupling) in samples grown by a variety of methods [metal-organic chemical vapor deposition (MOCVD), molecular-beam epitaxy (MBE), and metalorganic vapor phase epitaxy (MOVPE)] with a variety of results stemming from the uncertainties and idiosyncrasies of the various techniques used. In addition to a few reports on select Al$_x$Ga$_{1-x}$N layers, there have been four somewhat comprehensive studies of the refractive indices of Al$_x$Ga$_{1-x}$N. However, each of these incompletely covered the range of Al content $x$ or polarization, and there is a considerable discrepancy among the refractive indices as a function of $x$. To date, there has been no satisfying reconciliation of the disparate index measurements that would provide a systematic and reliable method for estimating index values to the precision required for optoelectronic applications.

In this letter, $n_o(\lambda)$ and $n_e(\lambda)$ are systematically measured in the visible range of wavelengths 457$<$\lambda$<$800 nm for a variety of Al$_x$Ga$_{1-x}$N samples with 0.0$\leq x$,$\leq$1.0. From the dispersion curves, measured by a highly accurate prism coupling technique, equations are developed that estimate the ordinary and extraordinary refractive index dispersion as functions of $\lambda$ and band gap energy ($E_g$). It is shown that the disparate measured index values are a consequence of less accurate index measurements and differing growth conditions with concomitant variations in strain, composition-induced inhomogeneities, and bowing parameters.

Al$_x$Ga$_{1-x}$N films were grown on (0001) sapphire by MBE using either rf activated nitrogen or ammonia as the nitrogen source. As-received sapphire substrates were subjected to a chemical treatment followed by a high temperature anneal, yielding an atomically smooth surface. The growth experiments followed a sequence of high temperature thermal treatment, high temperature hydrogen treatment, and a 30 nm thick AlN buffer layer growth. This was followed by the deposition of the 0.5 to 2.0 $\mu$m thick AlGaN layers investigated ($x=0.0, 0.15, 0.25, 0.35, 0.44, 0.77, 1.0$). Keeping the Al cell temperature constant and varying the Ga cell temperature controlled the AlN mole fraction in the ternary.

High-resolution x-ray rocking curves were measured by a Philips X’Pert MRD system equipped with four-crystal Ge (220) monochromator. The instrument resolution is verified to be $\approx$10 arcs under this diffraction geometry where the Cu $K\alpha_1$ line of x-ray source is used. Both symmetric (0002) x-ray diffraction and asymmetric (1014) peak diffraction were measured and used to compute the mole fraction.

The Al content measured by x-ray diffraction was compared with optical photoluminescence (PL) and absorption measurements. A tripled mode-locked Ti:Sapphire laser of wavelength $\sim$240 nm was used for PL measurements. Absorption measurements were performed using a 300 W Xe lamp, with wavelengths $>$220 nm. Transmitted light was dispersed by a 0.3 m imaging spectrometer and detected by a liquid nitrogen cooled charged coupled device camera. The absorption data is well characterized by

$$E_g(Al_xGa_{1-x}N) = 6.13x + 3.42(1-x) - bx(1-x),$$

(1)

where 6.13 eV and 3.42 eV are the room temperature band gap values for AlN and GaN, respectively, and $b=1.08$ eV is the measured bowing parameter. The latter is consistent with independent measurements of MBE-grown AlGaN films, but as will be discussed next, much larger than the bowing parameter ($b=-0.39$ eV) which characterized our previous study of MOCVD-grown Al$_x$Ga$_{1-x}$N samples. This technique-dependent variability in bowing parameters, which is a consequence of variations in growth conditions, is common in the literature.

A prism coupling technique, recently used to measure the birefringent indices of refraction of GaN, AlN, and Al$_x$Ga$_{1-x}$N epilayers, was used here. The laser wave-
lengths were derived from an Ar + laser (457.9, 476.5, 488, 496.5, and 514.5 nm), a HeNe laser (632.8 nm), a semiconductor AlInGaP laser (676 nm) and a Ti:Sapphire laser (800 nm). Routing the beams through a periscope changed the polarization.

The accuracy of the method is determined by the accuracy of the prism (n_p) and substrate (n_0) refractive indices and the accuracy of the angle measurements. The uncertainty in our measurement of n is dominated by the uncertainty in n_p. The maximum discrepancy in the reported values of n_p at a given wavelength is \( \pm 0.001 \).\(^{14}\) The uncertainty introduced by the sapphire index n_0 is negligible,\(^{15}\) and the coupling angles were measured to an accuracy of \( \pm 0.01^\circ \), which contributes an uncertainty of only \( \pm 0.0001 \) in n. (Of course, the largest source of relative uncertainty in n between samples is the uncertainty in the accuracy of x, which x-ray data constrain to \( \pm 10% \) in our data.)

The presence of the 30 nm AlN buffer layer introduces additional uncertainty in index, thickness, and indirectly, band gap values. Calculations using a multilayer waveguide program\(^{16}\) suggested that an error of \( \leq 0.0005 \) was introduced into n by ignoring the buffer layer. Thus, the absolute accuracy of n is \( \pm 0.01 \), but the relative uncertainty in n from sample to sample is \( \pm 0.0005 \). By contrast, the absolute uncertainty in n from interferometric techniques, such as that used in Brunner et al., is estimated to be \( \pm 1\% \), the uncertainty in thickness.\(^{4}\) The relative uncertainties are \( \pm 0.01 \), limited by the accuracy of wavelength measurements.

The ordinary and extraordinary indices measured at various laser lines are shown in Fig. 1. The data for each sample was fit to the first-order Sellmeier dispersion formula:

\[
\frac{n(\lambda)^2 - 1}{2} = \frac{A_0 \lambda^2}{\lambda^2 - \lambda_0^2}
\]

where \( A_0 \) and \( \lambda_0 \) are adjustable parameters.\(^{17}\) The resulting values of \( A_0 \) and \( \lambda_0 \) are shown in Fig. 2.

These index values differ markedly with four prior systematic studies of refractive index dispersion.\(^{4-7}\) For example, the discrepancy between the refractive index measurements of the current MBE-grown samples and our previous measurements of MOCVD-grown samples of x \( \approx 0.2 \) grew systematically with increasing Al content.\(^5\) To account for this variation, recall that the bowing parameter for the MOCVD-grown samples was \(-0.39\text{ eV}\), while it is \(1.08\text{ eV}\) for the MBE-grown samples. More generally, our measurements of GaN and AlN agreed with all values previously presented in the literature,\(^{4,5,12,13}\) but the discrepancies among all reported measurements grew as the Al content approached 50%.

Since the indices of refraction are fundamentally linked to the band gap energy and the onset of absorption through a Kramers–Köning relationship,\(^{18}\) we postulate that many of the reported variations in index dispersion are a consequence of varying growth conditions and concomitant band gap energies. To test this hypothesis and generalize these measurements, the Sellmeier relationship was parameterized by \( E_g \) not by x:\(^5\)

\[
A_0(E_g) = B_0 + B_1E_g + B_2E_g^2,
\]

\[
\lambda_0(E_g) = C_0 + C_1E_g + C_2E_g^2.
\]
The fitted curves are shown in Fig. 2, and the coefficients are given in Table I. The curves in Fig. 1, plotted from Eqs. (2) using Eqs. (3) and (4) and the coefficients in Table I, reproduce the data to within $\pm 0.007$.

As shown in Fig. 2, the resulting fit could reproduce both MOCVD and MBE-grown data sets. For further comparison, the Brunner et al. and Tisch et al. data for 457$<\lambda<800$ were fit with a Sellmeier relation parameterized by their reported band gap energies.\(^4\)\(^6\) The resulting coefficients are also plotted in Fig. 2. Given the greater uncertainty associated with their technique, the agreement with our measurements is satisfactory and much better than when parameterized by $x$. Similar results were obtained when this same parameterization was applied to the survey accomplished by Laws et al.,\(^7\) suggesting that Eqs. (2)–(4) provide an accurate estimate of $n_o(\lambda)$ and $n_e(\lambda)$ for all Al$_x$Ga$_{1-x}$N films. Measurement of the band gap energy is not necessary to extract the refractive index if the bowing parameter is known. Note that extrapolations beyond the wavelength region of our measurements are less reliable.\(^7\)

In summary, we have completed a systematic measurement of the ordinary and extraordinary indices of refraction for Al$_x$Ga$_{1-x}$N epitaxial layers. In this study, $n_o(\lambda)$ and $n_e(\lambda)$ have been measured to an accuracy of $\pm 0.01$ for seven Al$_x$Ga$_{1-x}$N (0.0$\leq x \leq 1.0$) MBE-grown layers on sapphire substrates with 457$<\lambda<800$ nm. The data were fit by a simple Sellmeier relationship, reproducing the refractive indices as a function of $\lambda$. A comparison with other measurements of refractive index dispersion reveals discrepancies that are correlated with variations in band gap, which are a consequence of differing growth conditions. When the Sellmeier relationship is parameterized as a function not of $E_g$, a universal method for estimating refractive index dispersion of Al$_x$Ga$_{1-x}$N is revealed.

The authors thank Arup Neogi for help with sample characterization. This work was supported in part by U.S. Army Research Office Grant No. DAAH04-96-0076. The VCU portion of this work was funded by AFOSR (Dr. G. L. Witt), NSF (Dr. L. Hess and Dr. G. Pomrenke), and ONR (Dr. C. E. C. Wood and Dr. Y. S. Park).


### Table I. Coefficients for the adjustable parameters in the Sellmeier dispersion formula.

<table>
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<tr>
<th>Coefficient</th>
<th>$n_o$</th>
<th>$n_e$</th>
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<td>$B_0$</td>
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<td>7.042</td>
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<tr>
<td>$B_1$</td>
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<td>-1.054</td>
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<tr>
<td>$B_2$</td>
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<td>0.0733</td>
</tr>
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<td>$C_0$</td>
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<td>381.2</td>
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