2001

Systematic measurement of AlxGa1−xN refractive indices

Ü. Özgür
Duke University, uozgur@vcu.edu

Grady Webb-Wood
Duke University

Henry O. Everitt
Duke University

Feng Yun
Virginia Commonwealth University

Hadis Morkoç
Virginia Commonwealth University, hmorkoc@vcu.edu

Özgür, Ü., Webb-Wood, G., Everitt, H.O., et al. Systematic measurement of AlxGa1−xN refractive indices. Applied Physics Letters, 79, 4103 (2001). Copyright © 2001 AIP Publishing LLC.

APPLIED PHYSICS LETTERS VOLUME 79, NUMBER 25 17 DECEMBER 2001

Systematic measurement of $Al_xGa_{1-x}N$ refractive indices

Ümit Özgür, Grady Webb-Wood, and Henry O. Everitt[a]
Department of Physics, Duke University, Durham, North Carolina 27708

Feng Yun and Hadis Morkoç
Department of Electrical Engineering, Virginia Commonwealth University, Richmond, Virginia 23284

(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 $Al_xGa_{1-x}N$ epitaxial layers with $0.0 \leqslant x \leqslant 1.0$ 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 parameter, 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, $In_xGa_{1-x}N$, and $Al_xGa_{1-x}N$) have been actively developed for short wavelength emitters and detectors in the green through the near ultraviolet spectral regions.[1] 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 [metalorganic 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.[2–6] In addition to a few reports on select $Al_xGa_{1-x}N$ layers,[2,3] there have been four somewhat comprehensive studies of the refractive indices of $Al_xGa_{1-x}N$.[4–7] 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 $Al_xGa_{1-x}N$ samples with $0.0 \leqslant x \leqslant 1.0$. 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 λ 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.

$Al_xGa_{1-x}N$ 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.[8] 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 μm 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 $\leqslant 10$ 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 ~ 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

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

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,[4] but as will be discussed next, much larger than the bowing parameter ($b = -0.39$ eV) which characterized our previous study of MOCVD-grown $Al_xGa_{1-x}N$ samples.[5] This technique-dependent variability in bowing parameters, which is a consequence of variations in growth conditions, is common in the literature.[9]

A prism coupling technique,[10,11] recently used to measure the birefringent indices of refraction of GaN,[12] AlN,[13] and $Al_xGa_{1-x}N$ epilayers,[5] was used here. The laser wave-

---

[a] Also U.S. Army Research Office, Research Triangle Park, NC 27709-2211; electronic mail: everitt@arl.aro.army.mil

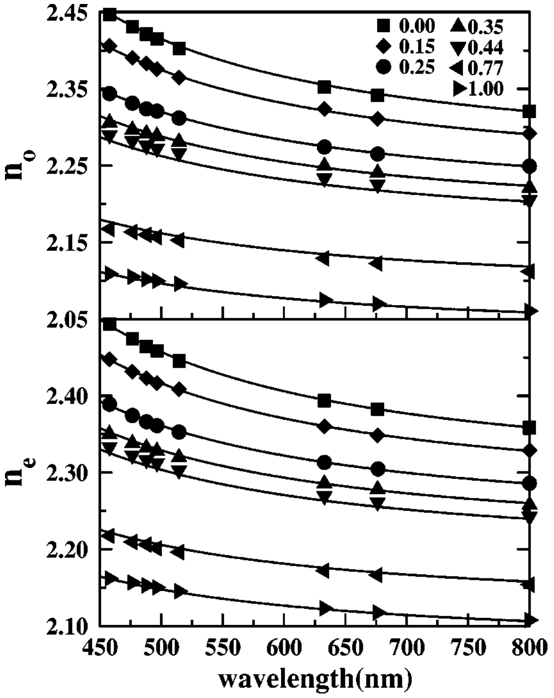

FIG. 1. Measured ordinary and extraordinary index dispersion for the MBE-grown samples with different x values. The lines are a result of the Sellmeier parameterization with band gap energy as described in Fig. 2.

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 $\sim\pm 0.01$.[14] The uncertainty introduced by the sapphire index n_0 is negligible,[15] and the coupling angles were measured to an accuracy of $\pm 0.01°$, which contributes an uncertainty of only ± 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 $\leqslant 0.0005$ was introduced into n by ignoring the buffer layer. Thus, the absolute accuracy of n is $\sim\pm 0.01$, but the relative uncertainty in n from sample to sample is $\leqslant\pm 0.0005$. 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 $\sim\pm 0.01$, 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,

$$n(\lambda)^2 = 1 + \frac{A_0\lambda^2}{\lambda^2 - \lambda_0^2}, \tag{2}$$

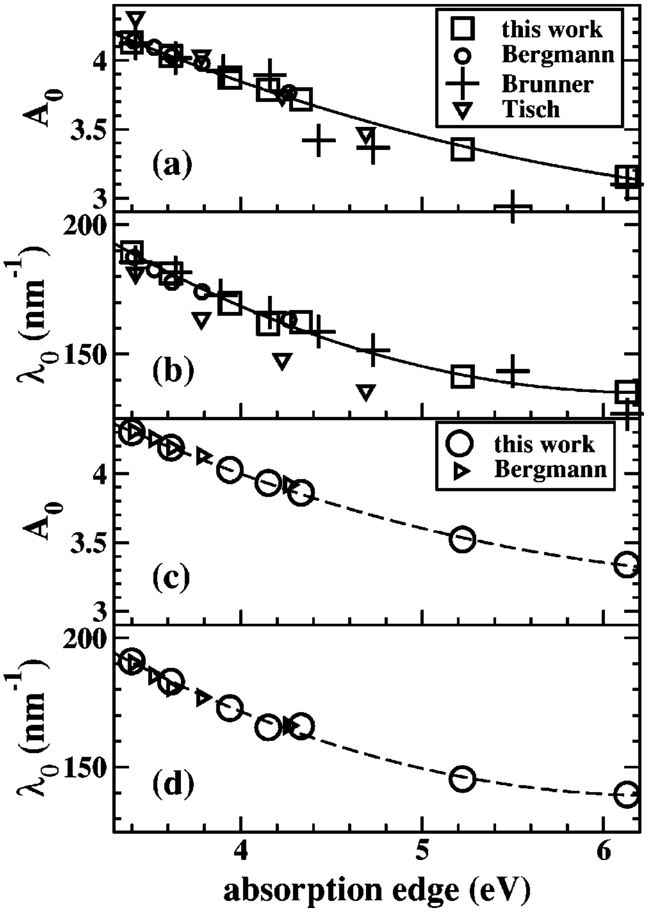

FIG. 2. Comparison of A_0 and λ_0 for various measurements of ordinary [(a) and (b)] and extraordinary [(c) and (d)] refractive indices. The solid lines are the Sellmeier parameterizations by E_g for the ordinary index parameters, and the dashed lines are for the extraordinary index parameters.

where A_0 and λ_0 are adjustable parameters.[17] The resulting values of A_0 and λ_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 $x \leqslant 0.2$ 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 eV, while it is 1.08 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–Kronig 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]

$$A_0(E_g) = B_0 + B_1E_g + B_2E_g^2, \tag{3}$$

$$\lambda_0(E_g) = C_0 + C_1E_g + C_2E_g^2. \tag{4}$$

TABLE I. Coefficients for the adjustable parameters in the Sellmeier dispersion formula.

Coefficient	n_o	n_e
B_0	6.626	7.042
B_1	−0.934	−1.054
B_2	0.0598	0.0733
C_0	396.8	381.2
C_1	−84.12	−76.68
C_2	6.758	6.068

The fitted curves are shown in Fig. 2, and the coefficients are given in Table I. The curves in Fig. 1, plotted from Eq. (2) using Eqs. (3) and (4) and the coefficients in Table I, reproduce the data to within $\sim\pm 0.007$.

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 $Al_xGa_{1-x}N$ 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 $Al_xGa_{1-x}N$ epitaxial layers. In this study, $n_o(\lambda)$ and $n_e(\lambda)$ have been measured to an accuracy of $\sim\pm 0.01$ for seven $Al_xGa_{1-x}N$ $(0.0 \leqslant x \leqslant 1.0)$ 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 λ. 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 x but of E_g, a universal method for estimating refractive index dispersion of $Al_xGa_{1-x}N$ 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).

[1] H. Morkoç, *Nitride Semiconductors and Devices* (Springer, Berlin, 1999).
[2] H. Amano, N. Watanabe, N. Koide, and I. Akasaki, Jpn. J. Appl. Phys., Part 2 **32**, L1000 (1993).
[3] G. Yu, H. Ishikawa, M. Umeno, T. Egawa, J. Watanabe, T. Jimbo, and T. Soga, Appl. Phys. Lett. **72**, 2202 (1998).
[4] D. Brunner, H. Angerer, E. Bustarret, F. Freudenberg, R. Höpler, R. Dimitrov, O. Ambacher, and M. Stutzmann, J. Appl. Phys. **82**, 5090 (1997).
[5] M. J. Bergmann, Ü. Özgür, H. C. Casey, Jr., H. O. Everitt, and J. F. Muth, Appl. Phys. Lett. **75**, 67 (1999).
[6] U. Tisch, B. Meyler, O. Katz, E. Finkman, and J. Salzman, J. Appl. Phys. **89**, 2676 (2001).
[7] G. M. Laws, E. C. Larkins, I. Harrison, C. Molloy, and D. Somerford, J. Appl. Phys. **89**, 1108 (2001).
[8] J. Cui, A. Sun, M. Reshichkov, F. Yun, A. Baski, and H. Morkoç, MRS Internet J. Nitride Semicond. Res. **5**, 7 (2000).
[9] S. F. Lee, A. F. Wright, M. H. Crawford, G. A. Petersen, J. Han, and R. M. Biefeld, Appl. Phys. Lett. **74**, 3344 (1999).
[10] P. K. Tien, R. Ulrich, and R. J. Martin, Appl. Phys. Lett. **14**, 291 (1969).
[11] R. Ulrich and R. Torge, Appl. Opt. **12**, 2901 (1973).
[12] H. Y. Zhang, X. H. He, Y. H. Shih, M. Schurman, Z. C. Feng, and R. A. Stall, Appl. Phys. Lett. **69**, 2953 (1996).
[13] X. Tang, Y. Yuan, K. Wongchotigul, and M. G. Spencer, Appl. Phys. Lett. **70**, 3206 (1997).
[14] We used n_p from J. Rams, A. Tejeda, and J. M. Cabrera, J. Appl. Phys. **82**, 994 (1997).
[15] W. J. Tropf and M. E. Thomas, in *Handbook of Optical Constants of Solids III*, edited by E. D. Palik (Academic, San Diego, 1998), p. 653.
[16] M. J. Bergmann and H. C. Casey, Jr., J. Appl. Phys. **84**, 1196 (1998).
[17] M. Born and E. Wolf, *Principles of Optics*, 6th ed. (Pergamon, Oxford, 1989).
[18] P. Yu and M. Cardona, *Fundamentals of Semiconductors*, (Springer, Berlin, 1996).