Band gap bowing and refractive index spectra of polycrystalline AlxIn1−xN films deposited by sputtering

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Citation: Appl. Phys. Lett. **71**, 2439 (1997); doi: 10.1063/1.120112
View online: http://dx.doi.org/10.1063/1.120112
View Table of Contents: http://apl.aip.org/resource/1/APPLAB/v71/i17
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Band gap bowing and refractive index spectra of polycrystalline $Al_xIn_{1-x}N$ films deposited by sputtering

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(Received 2 June 1997; accepted for publication 28 August 1997)

The AlGaInN semiconductor system is currently of high interest for applications in blue light emitting devices. AlInN is a prospective material for lattice matched confinement layers. We measure the refractive index as well as the band gap across the entire compositional range of high-quality polycrystalline AlInN samples. Strong band gap bowing is observed. © *1997 American Institute of Physics.* [S0003-6951(97)01543-X]

(Al, Ga, In) nitrides are under intense investigation for applications in semiconductor blue light emitting devices. Current achievements in this area are mainly based on binary GaN, ternary AlGaN, and ternary GaInN.[1] The third ternary nitride alloy, AlInN, is less investigated. This alloy exhibits the largest variation in band gap and it is a candidate for lattice matched confinement layers in optical devices. However, band gap measurements on AlInN alloys indicate a significant bowing.[2] Refractive index calculations based on these measurements strongly deviate from the linear estimation.[3] Further exploration of AlInN is expected to help predict properties of quaternary AlGaInN alloys. We investigate optical properties of high-quality AlInN thin films obtained by low temperature sputter deposition. Refractive index spectra are reported and the AlInN band gap is measured across the whole compositional range.

The AlInN films were prepared by a Discovery-18 magnetron reactive sputter system.[4] The aluminum plate and the indium plate were separately mounted onto the direct-current powered and radio-frequency powered target, respectively. The advantage of this scheme over the single composite target[2] is that we obtain a better composition control of the plasma reaction and a larger composition variation. The pressure of the chamber was pumped down to 10^{-8}–10^{-9} Torr before deposition. Substrate temperature during deposition was kept at 200 °C. The AlInN alloy was produced by the plasma reaction among Al, In, and N. Silicon, quartz, and glass were employed as substrates.

Rutherford backscattering spectroscopy (RBS) is used[4] to determine the $Al_xIn_{1-x}N$ film composition within an uncertainty of ± 0.05 in x value. Oxygen signals are not detected. A film thickness of about 1000 nm is deduced from the RBS analysis. X-ray diffraction (XRD) is employed to investigate the structure of the deposited films. The XRD spectrum of $Al_{0.64}In_{0.36}N$ indicates a high-quality polycrystalline structure (Fig. 1). The shift of the diffraction peaks with composition (Table I) proves the solubility between AlN and InN. In other words, the sputtered AlInN alloys are stoichiometric materials. Phase separation is not observed. Also, evidenced by the indices of wurtzite AlN and InN, the sputtered AlInN alloys share the wurtzite structure.

Optical transmission from 100 to 800 nm wavelength is measured. The square of the absorption coefficient α is plotted versus photon energy in Fig. 2. The linear high-energy part of these curves confirms direct band gap transitions. By extending this linear part down to the energy axis, the direct band gap E_g is determined at the interception point. Deviations of the absorption curves in Fig. 2 from a straight line

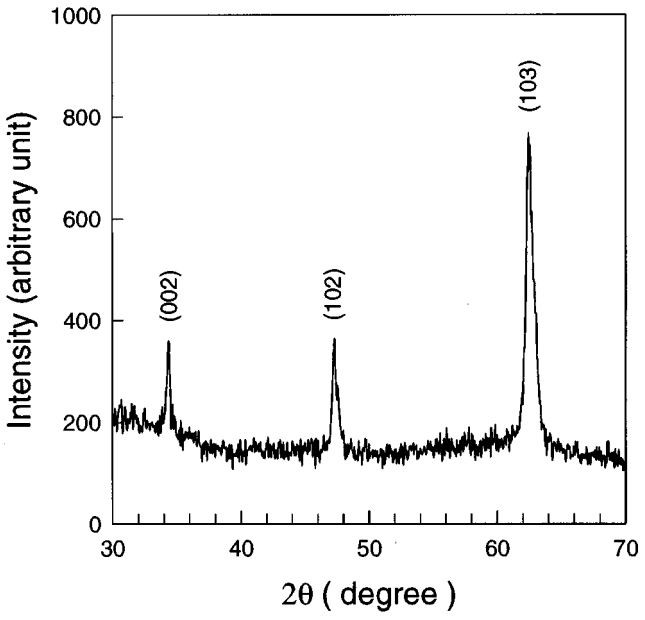

FIG. 1. XRD spectrum of the $Al_{0.64}In_{0.36}N$ sample.

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TABLE I. XRD peaks of $Al_xIn_{1-x}N$ films.

Composition	2θ (102) [degree]	2θ (103) [degree]	Lattice constant a (Å)
InN	43.50	57.00	3.54
$Al_{0.18}In_{0.82}N$	44.15	58.15	3.49
$Al_{0.25}In_{0.75}N$	44.60	58.75	3.46
$Al_{0.50}In_{0.50}N$	46.10	60.75	3.35
$Al_{0.64}In_{0.36}N$	47.40	62.60	3.26
$Al_{0.85}In_{0.15}N$	49.05	65.00	3.16
AlN	49.60	66.20	3.12

are attributed to grain boundary effects (Urbach tail). Those are excluded in our band gap analysis. The band gap is given as a function of the lattice constant a in Fig. 3, together with the results presented by other groups. The typical linear interpolation of binary data is shown for comparison (triangle).[3] In perfect agreement with Ref. 2 a strong energy gap bowing is found for $Al_xIn_{1-x}N$ alloys, which might be attributed to the atomic size difference between Al and In. This result is confirmed by the close match of our binary data with typical values for AlN and InN (triangle corner points in Fig. 3). The measured $Al_xIn_{1-x}N$ energy gap bowing can be approximated by

$$E_g[\mathrm{eV}] = 1.75 + 2.2x - 6.9x^2 + 9.1x^3. \quad (1)$$

It is expected to have a strong impact on device applications of AlInN or AlGaInN alloys. Those materials do not seem to provide good confinement layers on AlGaN or GaInN (lower solid lines in Fig. 3).

The refractive index of the AlInN films is determined from their reflectivity spectra. For AlInN film deposited on silicon substrate, the reflectivity spectrum provides an interference pattern with intensity minima occurring at $m\lambda = 2nd$, where m is the interference order, $n(\lambda)$ denotes the refractive index at wavelength λ, and d represents the film thickness. Based on this scheme, the refractive index spectra $n(\lambda)$ of the sputtered AlInN films are obtained within the transparency region. The dependence of $n(\lambda)$ on alloy composition is shown in Fig. 4. Our measurements of sputtered binary AlN are in good agreement with results from chemical vapor deposited samples.[6] The measured refractive index might be slightly affected by grain boundary effects. The short wavelength end of our curves is given by the band gap. With higher In content, the band gap shrinks and the spectrum $n(\lambda)$ shows the expected shift towards longer wavelengths.

In summary, we have investigated optical properties of polycrystalline AlInN as a function of alloy composition. Strong bowing of the direct energy gap is found. The refractive index spectrum within the transparent wavelength region shows the expected shift to higher values with lower band gap.

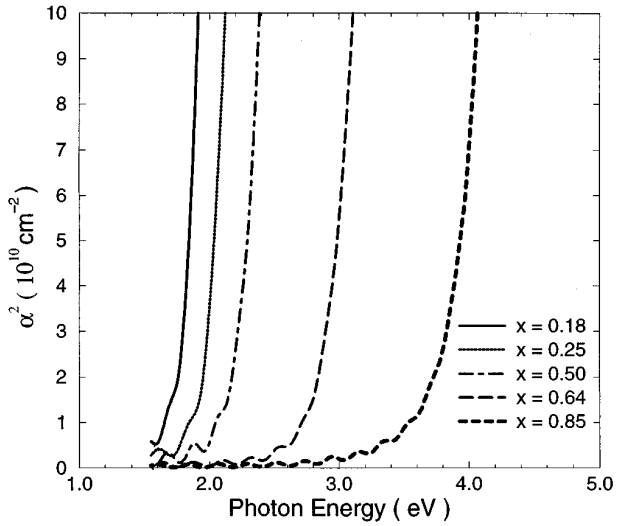

FIG. 2. Absorption spectra of sputtered $Al_xIn_{1-x}N$ films.

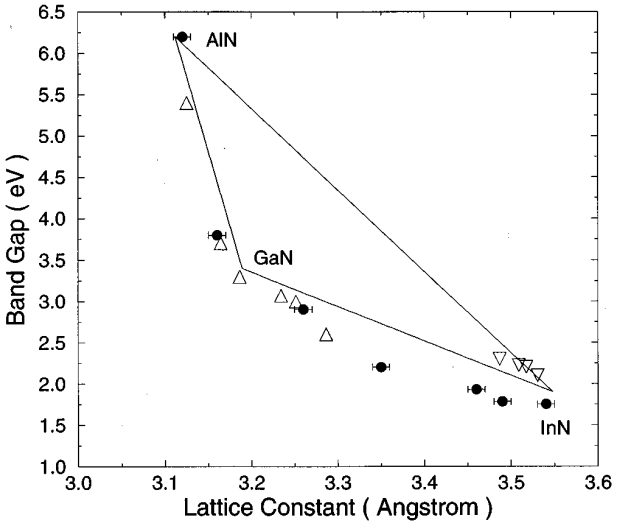

FIG. 3. Band gap vs lattice constant of $Al_xIn_{1-x}N$ alloys. Filled dots give our measurement (●), which is compared to other results [△ (see Ref. 2), ▽ (see Ref. 5)]. The error bars indicate the composition uncertainty and the solid lines show the typical linear interpolation of binary data.

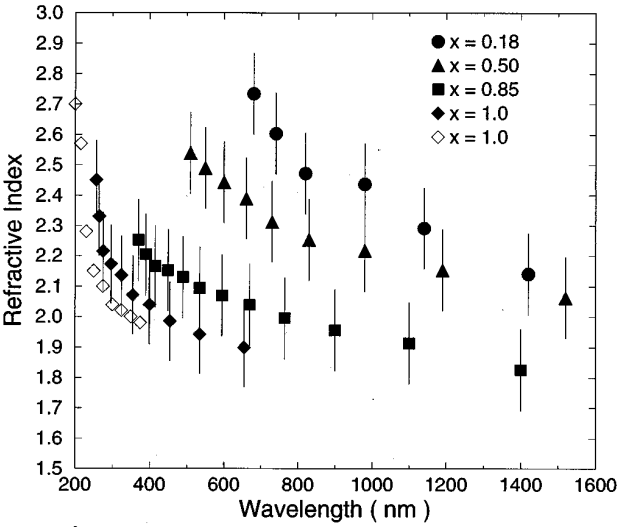

FIG. 4. Refractive index spectra of $Al_xIn_{1-x}N$ films. Filled dots are our measurements, open dots are reported by Bauer *et al.* (see Ref. 6) The error bars are mainly due to the uncertainty in sample thickness.

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