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Energy band bowing parameter in $Al_xGa_{1-x}N$ alloys

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Molecular-beam epitaxy grown $Al_xGa_{1-x}N$ alloys covering the entire range of alloy compositions, $0 \leqslant x \leqslant 1$, have been used to determine the alloy band gap dependence on its composition. The Al chemical composition was deduced from secondary ion mass spectroscopy and Rutherford backscattering. The composition was also inferred from x-ray diffraction. The band gap of the alloy was extracted from low temperature optical reflectance measurements which are relatively more accurate than photoluminescence. Fitting of the band gap data resulted in a bowing parameter of $b = 1.0$ eV over the entire composition range. The improved accuracy of the composition and band gap determination and the largest range of the Al composition over which our study has been conducted increase our confidence in this bowing parameter. *© 2002 American Institute of Physics.*
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Nitride semiconductors are currently under intensive research, driven by the wide application potential in electronic and optical devices.[1–5] Key to all of these devices is $Al_xGa_{1-x}N$, which is an indispensable component of both optical and electronic devices based on wide band gap nitrides. When used in single or multiple quantum well AlGaN/(In,Ga)N structures for enhanced performance, knowledge of the energy band gap in relation to the Al composition, x, is crucial for carrier and optical confinement for emitters and the cutoff wavelength for detectors. When used in FETs, accurate knowledge of the band gap discontinuity and band-offset ratio are pivotal in predicting the behavior of two-dimensional electron gas, which is also affected by piezoelectric effects, which in turn is related to the composition.[6,7] In both groups of devices, precise knowledge of a band gap as a function of x is a prerequisite for band gap engineering in order to fulfill device applications. So far, there have been scores of papers in the literature discussing the bowing parameter of $Al_xGa_{1-x}N$ alloys.[8–19] A close examination of previous works[20–32] on the band gap dependence of $Al_xGa_{1-x}N$ on composition indicates a wide scatter in the data, as shown in Fig. 1. The dispersion of bowing parameters reported by various researchers extends from -0.8 eV (upward bowing) to $+2.6$ eV (downward bowing), most likely emanating from $Al_xGa_{1-x}N$ alloys prepared by different techniques with various quality and, in some cases, the range of alloy compositions explored being narrow. In this letter, we report a systematic study of the dependence of $Al_xGa_{1-x}N$ band gap on Al mole fraction over the entire range of aluminum composition, $0 \leqslant x \leqslant 1$. The band gap of our AlN films has been determined in a separate collaborative effort.[33]

The $Al_xGa_{1-x}N$ samples were grown in an MBE system using N_2 rf plasma source on c-plane sapphire with highly purified nitrogen. An initial AlN buffer layer (~40 nm) was first grown on nitridated sapphire substrates followed by the growth of AlGaN layers of interest. The AlGaN layers were grown at what we consider to be a medium temperature range (600–670 °C) for nitride MBE.[34] Unlike conventional methods where the ratio of Ga/Al flux was used to control the Al mole fraction, we accomplished the same in an unconventional manner.[35] This method consistently leads to much higher quantum efficiencies when compared to samples grown using a conventional MBE growth of AlGaN. Both thin (~0.15 μm) and thick (~0.8 μm) AlGaN layers were grown for different Al compositions to gain confidence in the control of Al composition as a function of thickness. From an x-ray rocking curve (ω-scan), the best sample with Al composition up to $x = 0.26$ exhibited a remarkably narrow full width at half maximum (FWHM) of 1.9 arcmin for (0002) diffraction, while the higher Al sample ($x = 0.71$) still exhibits a FWHM of ~5.5 arcmin. Even for $x = 0.92$, the FWHM of (0002) is 8.8 arcmin. We attribute a good deal of this variation in x-ray diffraction (XRD) linewidth to the nitridation process which is affected by the rf source used for this purpose.

Al mole fraction x was obtained from the out-of-plane lattice constant (c) measured by a set of high resolution x-ray rocking curves using a measurement technique described by Fatemi.[36] A possible deviation from Vegard's law and distortion by deformed c/a ratio for different Al mole fraction layers with different thickness have been modeled in previous

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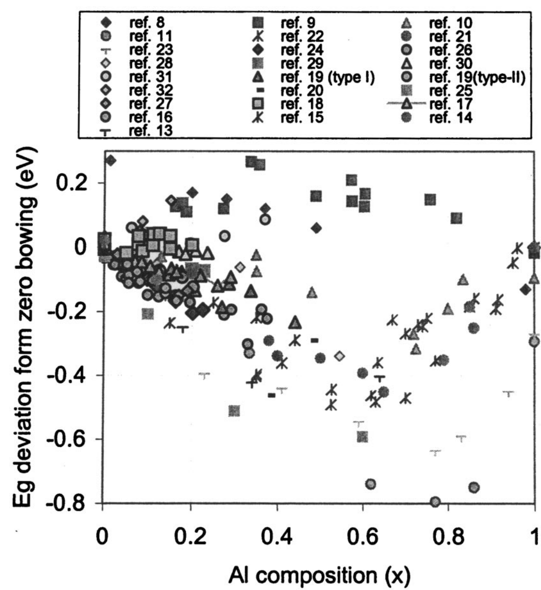

FIG. 1. Experimental data from published works of Al composition in AlGaN versus energy band gap plotted as the deviation from zero bowing.

TABLE I. Energy band gap data measured by reflectance spectra on $Al_xGa_{1-x}N$ alloys. The aluminum mole fraction was measured by XRD, and corroborated by SIMS, and RBS. A correction was made based on second-order calibration factors obtained from SIMS and RBS.

Measured Al mole fraction, x			Second-order corrections, x			Band gap (eV) from reflectance
XRD	SIMS	RBS	Δx_1	Δx_2	calibrated x	
0.13	0.13	⋯	0.003	0.014	0.136	3.78±0.05
0.26	0.26	0.29	0.010	0.023	0.276	4.09±0.05
0.36	0.37	0.38	0.014	0.027	0.374	4.32±0.05
0.38	0.40	⋯	0.014	0.028	0.389	4.30±0.05
0.46	0.49	⋯	0.016	0.029	0.480	4.55±0.05
0.51	⋯	⋯	0.017	0.030	0.525	4.65±0.05
0.63	0.64	0.65	0.016	0.028	0.645	5.00±0.05
0.67	⋯	⋯	0.016	0.026	0.679	5.06±0.10
0.69	⋯	⋯	0.015	0.025	0.703	5.15±0.10
0.71	⋯	⋯	0.015	0.024	0.723	5.18±0.10
0.92	⋯	⋯	0.007	0.009	0.920	5.87±0.10

works.[12,21] Experimental correction of strain-related effects requires precise measurement of in-plane lattice constant (a) of each $Al_xGa_{1-x}N$ layer. We found this to be hard to obtain with confidence due to the somewhat broad asymmetric diffraction peaks which become insensitive to tilt correction, especially for high Al composition samples. To be certain about the composition, we relied on secondary ion mass (SIMS) and Rutherford backscattering spectroscopy (RBS) measurements for composition verification and calibration which turned out to confirm the figures extracted from x-ray data. Both techniques are capable of depth profiling of Al variation, if any, and former is strain independent.

SIMS analyses were carried out using a Physical Electronics, Inc. quadrupole system. The Al composition was obtained by comparing the atomic fractions of Ga and Al, using one of the samples (x=0.26) as a standard to calibrate the signal intensity. RBS was operated with 2.275 MeV He^{++} as incident beam, and backscattering detector at 160° azimuth angle. For the three samples characterized by RBS, uniform depth profiles were acquired for two samples, whereas the third one (x~0.29) showed a detectable depth variation of Al mole fraction caused by a change in N flow during growth, which underscores the importance of controlling and maintaining the reactive N flux. This effect was taken into account when reporting the Al mole fraction.

The results of Al mole fraction determined by XRD, SIMS, and RBS methods are summarized in Table I. While the Al mole fractions obtained from SIMS and RBS are, in general, close to those from XRD, they tend to be a little higher than those from XRD measurements, with a maximum deviation of ~3%. Assuming a parabolic distribution of deviation, a second-order approximation was used.

Reflectance and PL spectra were measured at 15 K. For reflectance measurements, a 30 W deuterium lamp was used as a light source. For PL measurements, a 60 mW He–Cd laser (325 nm) was used. The reflected and emitted light was dispersed by a 0.5 m focal length spectrometer with 1200 grooves/mm grating blazed at 250 nm. Signals were detected by a photomultiplier using photon counting. An $Al_xGa_{1-x}N$ band gap was determined from the low-temperature reflectance spectra as the critical photon energy where reflectance oscillations fade due to above-band absorption. The results are included in Table I. Special care was taken to ensure characterization on the same spot for each sample for all measurements to rule out any possible spatial distribution across the wafer since we did not rotate the substrate during growth. Despite these efforts, we believe that the band gap determination is the main source of any notable inaccuracy in the process. We also add that samples with Al mole fractions near 50% are the ones that affect the bowing parameter the most. Photoreflectance measurements are more appropriate as a redshift in PL could occur from carrier localization caused by potential fluctuations as depicted in the inset of Fig. 2. In addition, it is not always possible to determine whether the observed transitions in the emission measurements, such as in PL, are band to band and what their bind-

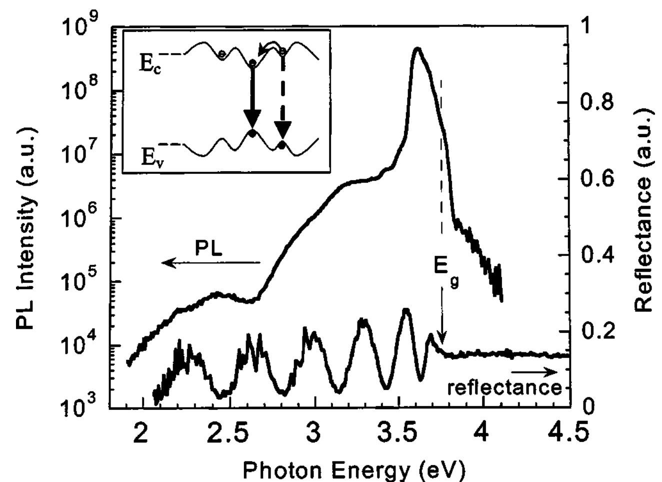

FIG. 2. Low-temperature (15 K) reflectance and PL spectra of AlGaN (x~0.13) sample. The inset is a schematic energy band diagram showing the effects of any local AlGaN fluctuation.

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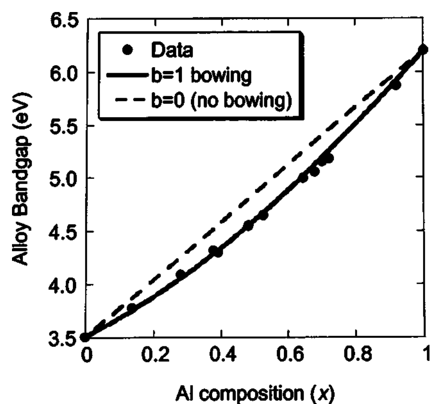

FIG. 3. Experimental data of energy band gap of AlGaN ($0 \leq x \leq 1$) plotted as a function of Al composition (solid circle), and the least squares fit (solid line) giving a bowing parameter of $b = 1.0$ eV. The dashed line shows the case of zero bowing.

ing energies are. This, at least in part, accounts why $Al_xGa_{1-x}N$ band gap obtained from PL always tends to be lower than the real figure, as exemplified clearly in Fig. 2 for an $Al_xGa_{1-x}N$ ($x = 0.13$) sample measured at 15 K.

Bowing parameter b is defined as the coefficient of the parabolic term in the phenomenological expression of alloy energy band gap: $E_g(x) = E_g^1 x + E_g^0(1-x) - bx(1-x)$, where E_g^0 and E_g^1 are the energy band gaps of GaN and AlN binary end points, respectively. Note that a positive value of b represents a downward bowing, and a negative b represents an upward bowing. We took the band gaps of GaN as $E_g^0 = 3.505$ eV (at 4 K)[37] and AlN as $E_g^1 = 6.20$ eV (at 2 K),[33] and plotted the energy band gaps of $Al_xGa_{1-x}N$ alloys versus the calibrated Al mole fractions as depicted in Fig. 3. A least-squares fit to the data, solid circles, yields a bowing parameter of $b = 1.0$ eV for the entire range of alloy compositions.

In conclusion, we investigated the band gap energy dependence of $Al_xGa_{1-x}N$ alloys on Al mole fraction for $0 \leq x \leq 1$ grown by MBE. We employed three different characterization techniques (XRD, SIMS, and RBS) for determining the Al composition. Low temperature reflectance measurements were used to extract the energy band gap of each AlGaN alloy. By fitting the experimental data to the phenomenological band gap–composition relationship, a bowing parameter of $b = 1.0$ eV, downward, was obtained. The entire range of Al mole fractions studied and the well-established analyses techniques employed increase our confidence in the bowing parameter attained.

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