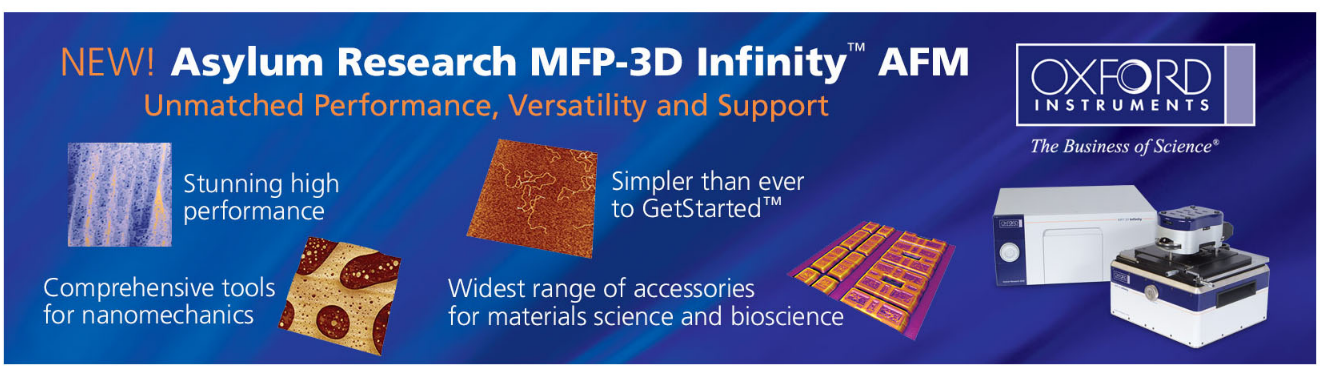
AIP | Applied Physics Letters

Band-edge photoluminescence of AlN epilayers

J. Li, K. B. Nam, M. L. Nakarmi, J. Y. Lin, and H. X. Jiang

Citation: Applied Physics Letters **81**, 3365 (2002); doi: 10.1063/1.1518558
View online: http://dx.doi.org/10.1063/1.1518558
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/81/18?ver=pdfcov
Published by the AIP Publishing



Band-edge photoluminescence of AlN epilayers

J. Li, K. B. Nam, M. L. Nakarmi, J. Y. Lin, and H. X. Jiang[a)]
Department of Physics, Kansas State University, Manhattan, Kansas 66506-2601

(Received 15 April 2002; accepted 9 September 2002)

AlN epilayers with high optical qualities have been grown on sapphire substrates by metalorganic chemical vapor deposition. Deep ultraviolet photoluminescence (PL) spectroscopy has been employed to probe the optical quality as well as optical transitions in the grown epilayers. Band-edge emission lines have been observed both at low and room temperatures and are 6.017 and 6.033 eV at 10 K. It was found that the peak (integrated) emission intensity of the deep impurity related transition is only about 1% (3%) of that of the band-edge transition at room temperature. The PL emission properties of AlN have been compared with those of GaN. It was shown that the optical quality as well as quantum efficiency of AlN epilayers is as good as that of GaN. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1518558]

Although much progress has been made for III-nitrides research and development in terms of both fundamental understanding as well as devices applications, the materials we understood relatively well including only the GaN compound and In (Al) GaN alloys with In (Al) content less than 30% (50%). InGaN alloys with high In contents (>50%), which emit light in the orange to red color spectral range, can be replaced by other semiconductors. However, AlGaN alloys with high Al contents, covering 350–200 nm, cannot be replaced by any other semiconductor system due to the fact that no other semiconductors possess such a large direct band gap (diamond is 5.4 eV with indirect band gap) as well as the ability of band-gap engineering through the use of heterostructures.

Currently, there is much need of solid-state ultraviolet (UV) emitters for many applications, including next generation lighting and chem-bio-agent detections. In such applications based on III-nitride wide band-gap semiconductors, both *n*- and *p*-type high quality AlGaN alloys with high Al contents are indispensable. However, achieving device quality AlGaN with high conductivities and quantum wells with high Al contents remains as one of the foremost challenging tasks for the nitride community. In particular, our knowledge concerning the optical properties of AlN is very scarce, despite its importance for fundamental understanding of wide band-gap semiconductor properties as well as for device applications. Band-edge photoluminescence (PL) emission lines in AlN have not been reported previously due to the lack of high quality materials as well as technical difficulties involved in the deep UV (down to 200 nm) PL measurements. As a consequence, very little is known of all fundamentally important optical transitions in AlN. The reported band gap and optical data were mostly obtained through the optical absorption measurements. Although band-edge emission lines near 6.0 eV have been observed in AlN at room temperature by cathodoluminescence (CL) measurement,[1,2] the broad feature of the emission bands (linewidth larger than 100 meV) due to the presence of high concentration of defects/impurities as well as the use of the high-energy electron beam excitation made the identification of the transition mechanisms in AlN epilayers very difficult. AlN is an end point of the AlGaN alloy system, we would not have a complete image for the AlGaN alloy system, particularly Al rich AlGaN alloys, before we obtain a better understanding of the AlN compound.

With its large direct band gap of about 6.1 eV at room temperature, high thermal conductivity and hardness, and high resistance to chemicals,[3] AlN has many attractive features.[4] For example, AlN has applications for surface acoustic wave (SAW) devices because of its piezoelectric properties.[5] It is needless to say that the future applicability of AlN devices depends on the development of methods for producing high quality materials and device structures as well as on the full understanding of the basic properties of this material. From the materials growth point of view, feedback from different material characterization techniques is essential for achieving further improvements in materials quality. Due to its insulating nature, electrical characterization methods such as Hall effect measurement, which has been extremely effective for optimizing GaN growth, is no longer applicable to AlN. It has also been difficult to characterize the optical properties of AlN due to its wide band gap. Although x-ray diffraction measurement (XRD) can provide information about crystalline quality, it provides very little information about electrical and optical qualities of AlN epilayers. Because of our optical measurement capabilities for working with AlN,[6,7] we are equipped with ''eyes'' for monitoring the optical quality as well as for probing the fundamental optical properties of AlN. In this letter, we report the achievement of high quality AlN epilayers that emit exciton PL emission.

The 1 μm thick AlN epilayers were grown by metalorganic chemical vapor deposition (MOCVD) on sapphire (0001) substrates with low temperature AlN nucleation layers. Trimethylaluminum (TMAl) and NH_3 were used as Al and N sources. (XRD) revealed that the full width at half-maximum (FWHM) of the AlN (0002) rocking curves varied from 150 to 500 arcsec, which are among the best values reported for AlN epilayers.[2,8,9] For PL measurements carried

[a)]Electronic mail: jiang@phys.ksu.edu

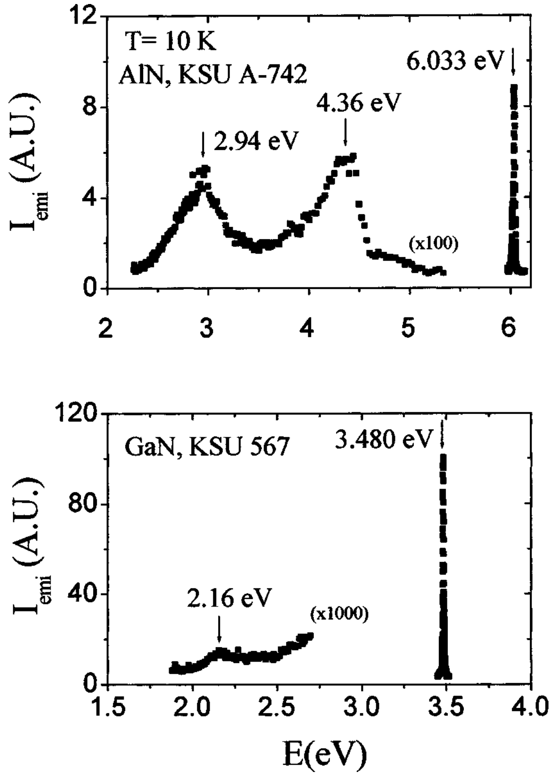


FIG. 1. PL spectra of AlN and GaN epilayers measured at 10 K, which covers a broad spectral range from 2.2 to 6.2 eV for AlN and 1.8–3.6 eV for GaN.

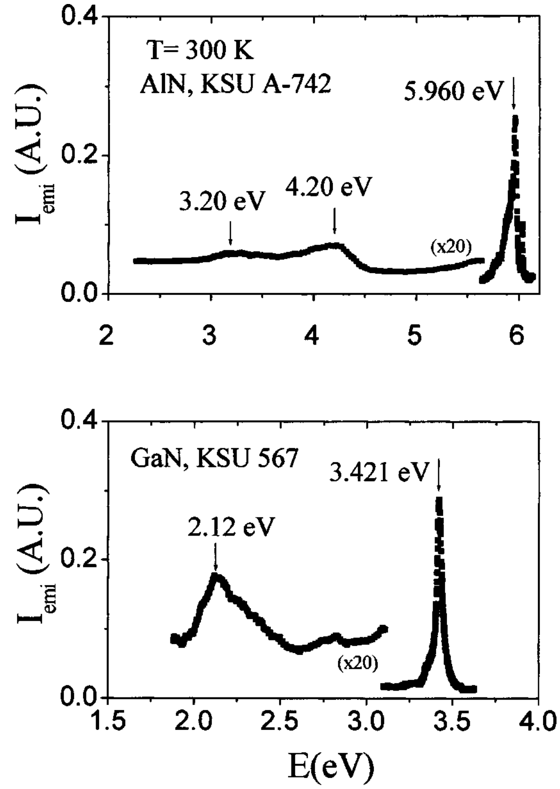


FIG. 2. PL spectra of AlN and GaN, epilayers measured at 300 K, which covers a broad spectral range.

out on AlN, a specially designed deep UV laser spectroscopy system was utilized, which basically consists of a quadrupled Ti:sapphire laser together with a streak camera, providing an excitation power of about 3 mW at 196 nm.[6,7] For GaN, in addition to the 196 nm deep UV laser, a second laser system with excitation wavelength tunable from 285 to 320 nm was also used as an excitation source. This second laser system consisted of a cavity-dumped dye laser with 6G dye solutions, which was pumped by a YAG laser with a frequency doubler, while the output of the dye laser was frequency doubled again to provide a tunability from 285 to 320 nm.[6] The PL results of GaN obtained by the two laser systems of different excitation wavelengths are similar. For example, the ratio of the PL emission intensity at room temperature to that at 10 K in GaN obtained by the two laser systems of different excitation wavelengths was about the same. Hence, we believe that the effect due to the variation in optical absorption depth as a result of the use of different excitation wavelengths for different materials (AlN versus GaN) is negligibly small.

Figure 1 compares the low temperature (10 K) PL spectra of our AlN and GaN epilayers covering a broad spectral range from 2.2 to 6.2 eV for AlN and 1.8–3.6 eV for GaN. One can see that the peak emission intensity of the deep level impurity related transition at 2.16 eV (the yellow line) in our GaN epilayers is about four orders of magnitude lower than that of the band-edge transition at about 3.48 eV, revealing the high optical quality of our GaN. In AlN, there are two broad emission bands related with deep level impurities at about 2.94 and 4.36 eV, however with peak (integrated) emission intensity being only 1% (3%) of that of the band-edge emission line at 6.033 eV, which indicates that the optical quality of our AlN epilayers is also sufficiently high. It was observed that the optical quality or the intensity ratio of the band-edge to the deep level impurity transitions depends strongly on the growth conditions.

Figure 2 compares the room temperature PL spectra of AlN and GaN epilayers, again covering broad spectral ranges. One sees that at room temperature the PL emission intensity of the deep level impurity related transition is also about two orders of magnitude lower than that of the band-edge transition in our AlN epilayers. This points to a much-improved optical quality of our AlN epilayers over those in previous CL studies, in which a comparable peak emission intensity for the deep level impurity related and the band-edge transition lines was observed in AlN grown on sapphire substrates.[1,2] The low temperature PL emission spectrum of AlN is replotted in Fig. 3 in a narrower spectral range to reveal more detailed features of the band-edge emission, in

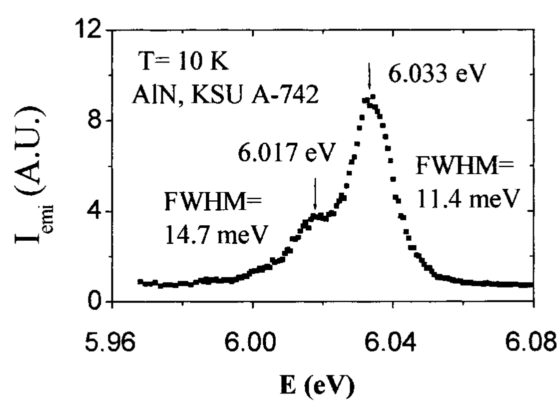


FIG. 3. Edge-emission spectrum of AlN measured at 10 K.

TABLE I. Comparison of PL emission properties of AlN and GaN, including PL peak position (E_p), full width at half maximum (FWHM), peak emission intensity (I_p), and integrated emission intensity (I_{int}).

		Band-edge			Deep impurity		
		AlN		GaN	AlN		GaN
$T=10$ K	E_p (eV)	6.017	6.033	3.480	2.94	4.36	2.16
	FWHM (meV)	14.7	11.4	5.0	700	600	200
	I_p (a.u.)	3.6	8.8	100	0.050	0.052	0.010
	I_{int} (a.u.)	8	19	100	2	2.8	0.3
$T=300$ K	E_p (eV)		5.960	3.420	3.20	4.20	2.12
	FWHM (meV)		31	28	1000	800	550
	I_p (a.u.)		0.27	0.3	0.0026	0.0032	0.0085
	I_{int} (a.u.)		3	3	0.08	0.1	0.4

which two transition lines at 6.017 and 6.033 eV can be clearly distinguished. The emission spectral line shape resembles those of GaN epilayers produced at earlier stage in which both free- and bound-excitons emission lines were present.[10] Our preliminary results from time-resolved PL studies suggest that the 6.033 eV line may result from the free-exciton recombination (FX), while that at 6.017 eV from the donor-bound-exciton recombination (or I_2). However, more careful studies are needed and are in progress to identify the origins of these two transitions.

Table I summarizes the PL emission properties of AlN and GaN epilayers measured at 10 and 300 K, including PL peak position (E_p), FWHM, peak intensity (I_p), and integrated intensity (I_{int}). The peak and integrated intensities for AlN and GaN are measured relative to that of GaN at 10 K (as 100). It is interesting to note from Table I and Figs. 1 and 2 that although the 10 K band-edge emission intensity is about one order of magnitude lower in AlN than in GaN, the room temperature emission intensities are comparable for both compounds. This implies that the thermal quenching of PL emission intensity is greatly reduced in AlN over GaN, which suggests that the detrimental effect of impurities and dislocations or nonradiative recombination channels in AlN is much less severe than in GaN. This points to the great potential of AlN for many device applications, because it is already well known that the detrimental effect of dislocations/impurities in GaN is much smaller than in other III–V and II–VI semiconductors.

In the past, AlN is referred to as a ceramic due to its very large band gap, poor quality, and highly insulating nature and is considered useful as a semiconductor only when alloyed with GaN or used as buffer and spacer layers in nitride structures and devices. Our results show that it is now emerging as an important semiconductor material, namely AlN epilayers of high optical qualities can be achieved by MOCVD. Since it is still at a very early stage for AlN epilayer growth, significant improvements in materials quality are anticipated. For example, during the last a few months, we have enhanced (suppressed) the emission intensity of the band-edge transition (deep level impurity related transition) by three orders of magnitude. However, an important issue for achieving a true semiconducting AlN is how to control its conductivity. Indeed, it was shown recently that the conductivity of AlN can be controlled and *n*-type conduction with a free electron concentration of about 1×10^{17} cm^{-3} has been achieved by Si doping.[11] With the demonstrated abilities of achieving high optical quality here and the *n*-type conductivity control of AlN epilayers, many applications of III-nitrides are conceivable. The most important message we want to deliver in this letter is that AlN can be grown as good as GaN and that the detrimental effect of nonradiative recombination channels in AlN is much less severe than in GaN.

In summary, AlN epilayers with high optical qualities have been grown by MOCVD on sapphire substrates. Very efficient band-edge PL emission lines have been observed with above band-gap deep UV laser excitation. The emission intensities of the deep level impurity transitions are about two orders of magnitude lower than that of the band-edge transition at room temperature. We have shown that the optical quality of AlN can be as good as GaN as demonstrated by quantum efficiency of PL especially at room temperature.

The research is supported by grants from ARO, DARPA, DOE (96ER45604/A000), NSF (DMR-9902431), BMDO, and ONR.

1. F. R. B. Hossain, X. Tang, K. Wongchotigul, and M. G. Spenser, Proc. SPIE **2877**, 42 (1996).
2. C. M. Zettering, M. Ostling, K. Wongchotigul, M. G. Spencer, X. Tang, C. I. Harris, N. Ordell, and S. S. Wong, J. Appl. Phys. **82**, 2990 (1997).
3. *Semiconductors-Basic Data*, edited by O. Madelung (Springer, New York, 1996), p. 69.
4. *Properties of Advanced Semiconductor Materials*, edited by M. E. Levinshtein, S. L. Ramyantsev, and M. S. Shur (Wiley, New York, 2001), p. 31.
5. K. M. Lakin, J. K. Liu, and K. L. Wang, J. Vac. Sci. Technol. **13**, 37 (1976).
6. http://www.phys.ksu.edu/area/GaNgroup
7. K. B. Nam, J. Li, K. H. Kim, J. Y. Lin, and H. X. Jiang, Appl. Phys. Lett. **78**, 3690 (2001).
8. M. A. Khan, J. N. Kuznia, R. A. Skogman, D. T. Olson, M. M. Millan, and W. J. Choyke, Appl. Phys. Lett. **61**, 2539 (1992).
9. K. Dovidenko, S. Oktyabrsky, J. Narayan, and M. Razeghi, J. Appl. Phys. **79**, 2439 (1996).
10. G. D. Chen, M. Smith, J. Y. Lin, H. X. Jiang, M. Asif Khan, and C. J. Sun, Appl. Phys. Lett. **67**, 1653 (1995).
11. Y. Taniyasu, M. Kasu, and N. Kobayashi, Appl. Phys. Lett. **81**, 1255 (2002).