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Room temperature photoreflectance investigations have been performed on a series of AlGaN layers grown both by metalorganic vapor phase epitaxy and molecular beam epitaxy on *c*-plane sapphire substrates. The aluminum composition was ranging between 0% and 20%, and was determined independently in the different growth laboratories, by various methods. It is found that within the experimental uncertainty, there is no detectable bowing parameter in these alloys. This contradicts some previous experimental investigations and confirms other ones. © *1999 American Institute of Physics.* [S0003-6951(99)01122-5]

There are currently a lot of industrial challenges for devices based on group III nitride semiconductors. Tremendous activity concerns the GaInN–GaN combination in straightforward relation with the huge market which exists for light emitters which can operate in the visible part of the electromagnetic spectrum.[1] There is also a need for the ultraviolet region. High performance transistors are expected, which could operate in hostile conditions, and with high power and high frequencies. GaN and its heterostructures with AlGaN are the best candidates for these applications.[2] It is important to optimize, the sheet electron density at hetero-interface to realize high quality modulation-doped field effect transistors. This depends on several parameters among which is the conduction band offset. It is also of crucial importance to determine this band offset ratio in order to elucidate the mechanisms which rule the carrier confinements in GaN-based quantum wells and superlattices. There are many ingredients to include in models. At this stage we can predict without any risk that the first issue is to address the dependence of the AlGaN band gap with alloy composition. There are a few papers in the literature about this topic,[3–10] theoretical investigations and experimental ones. From the experimental point of view there are two main difficulties, namely:

- The determination of the aluminum composition. In the present work we bypass this issue by studying a series of samples of different origins, grown by different methods. The aluminum composition was determined by various methods such as reflection high-energy electron diffraction (RHEED) oscillations,[11] or x-ray diffraction (XRD). Our approach permits us to restrict the uncertainty in the alu-

minum composition to spatial fluctuation of the composition in the crystals. We emphasize the fact that samples were grown in chambers where material science aspects are sufficiently well controlled that it is possible to obtain GaN–AlGaN quantum wells with fairly sharp interfaces.[1,11–13]

- The measurement of the band gap in alloys. We utilize photoreflectance which is more appropriate than photoluminescence to determine the band gap of alloys. This method is, we believe it also, more straightforward than transmission spectroscopy: it is always very difficult, sometimes even impossible to resolve the excitonic absorption peak in alloys.

In this letter, we report on photoreflectance studies of AlGaN alloy grown either by metalorganic chemical vapor epitaxy (MOVPE) or by molecular beam epitaxy (MBE). *The AlGaN band gap exhibits a linear variation with the Al mole fraction in the 0%–20% range.* Thus, we find the bowing parameter of the band gap (namely the half of the second derivative of the band gap energy versus alloy composition) to be very close to zero in our samples. This value ranges in between the two extrema of the previously determined values: $b = -1$ (Ref. 3) and $b = 1.3$ (Ref. 10) and agrees with two previous works (Refs. 4 and 7).

Photoreflectance spectra were collected at room temperature. The sample was shined by the monochromatically dispersed light of a quartz–tungsten–halogen lamp. The 325 nm radiation of a HeCd laser was used to change the dielectric constant of the semiconductors. The frequency of the chopper was low and adapted such as that the response of our photodetector—a Si photodiode biased in reversed polarization—was optimized. We also had to sometimes use filters. The data for the 20% aluminum was restricted to re-
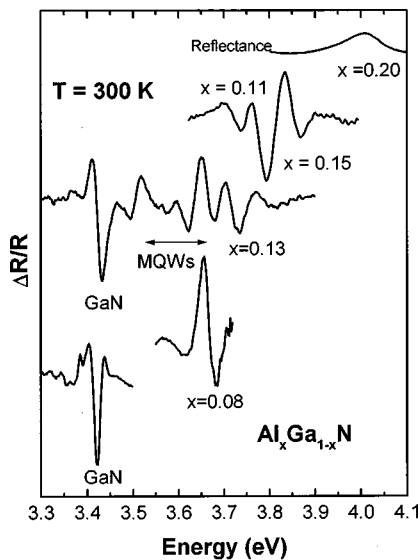
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FIG. 1. Room temperature photoreflectance of AlGaN alloys for aluminum compositions ranging up to 15%. At the right-hand top corner is the reflectance data of a $Al_{0.2}Ga_{0.8}N$ layer. We also inserted in the middle of the figure the photoreflectance feature obtained for a $GaN–Al_{0.13}Ga_{0.87}N$ multiple quantum well structure with wells having 4, 8, 12, and 16 monolayer widths. Photoreflectance structures of these wells are in the energy region of the horizontal arrow.

FIG. 3. Plot of the transition energies versus aluminum composition. The geographical origins of the samples are distinguished using different symbols. Black symbols correspond to the position of the C line, open symbols indicate the energy position of the A–B lines. Their splitting is not resolved in the alloys.

flectance investigation: we did not manage to collect a nice enough photoreflectance signal to publish it.

Figure 1 illustrates some selected photoreflectance spectra. At the right-hand bottom corner of the figure is inserted the reflectance spectrum for the $Al_{0.2}Ga_{0.8}N$ layer. To illustrate the strength of photoreflectance spectroscopy, we have selected a sample with four quantum wells of respective thickness 10, 20, 30, and 40 Å and separated by 50 Å $Al_{0.13}Ga_{0.87}N$ barrier layers. Obviously derivative structures corresponding to the GaN buffer, the AlGaN alloy layers and the four quantum wells can be resolved.

The line-shape fitting was made in the context of Aspne's third derivative line shape method.[14,15] The agreement between experiment and theory is shown in Fig. 2 for GaN [Fig. 2(a), sample No. N192] and for a multilayered structure with two aluminum compositions: 11% and 15% [Fig. 2(b), sample No. 178783].

Figure 3 reports the fitted values of the transition energies as a function of the aluminum composition. For each
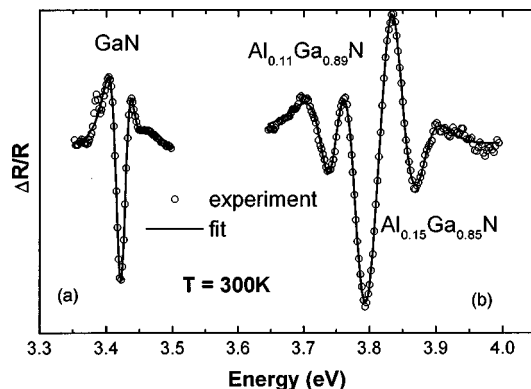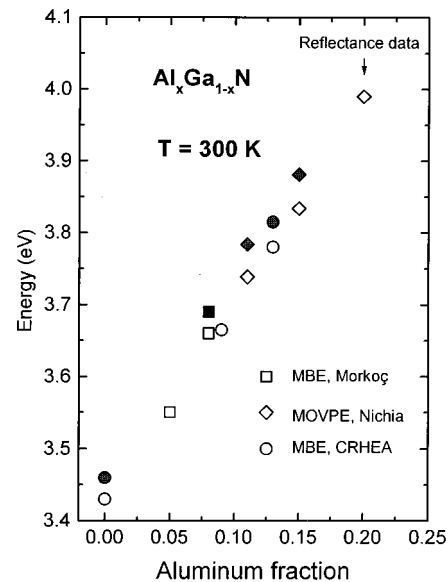
composition, open symbols correspond to the unresolved pair of A–B lines while full symbols indicated the position of the C line, when resolved. Circles correspond to samples grown at Centre de Recherche sur Hetero-Epitaxie et Applications by MBE using ammonia as nitrogen precursor. The aluminum composition was determined from RHEED intensity oscillations.[11] Diamonds correspond to samples grown by Shuji Nakamura, at Nichia, by MOVPE. The aluminum composition is obtained from XRD. Last, squares correspond to samples grown by Hadis Morkoç by MBE. Again, XRD was used to deduce the aluminum composition. Nitrides were grown with strain on sapphire. This study was restricted to samples grown on GaN buffer, and for which the signatures of GaN were almost at the same energy at 2 K

FIG. 2. Experimental data (open circles) and line-shape fitting (straight line) for two samples.
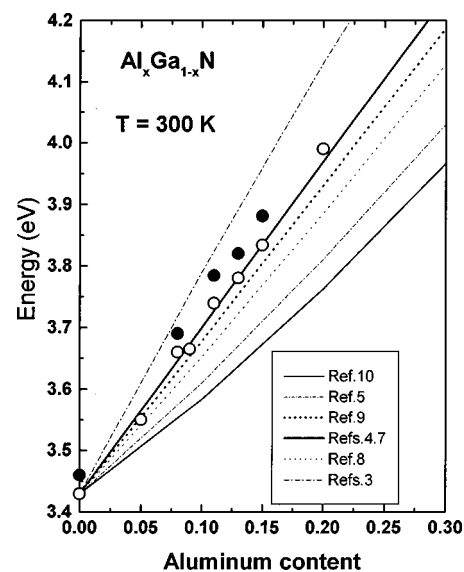

FIG. 4. Plot of the transition energies vs aluminum composition together with band gap composition dependencies previously reported in the literature.

(3.48 eV roughly for the A line). This limits the scattering of the energies due to difference in strain. In addition, we wish to emphasize the fact that the thickness of the AlGaN layer were of the order of few tens of angstroms, and that some of the growths were terminated by the growth of a thin GaN capping layer. This is not found to drastically influence the energies of the AlGaN while there is an impact at the scale of the phase of the photoreflectance signal.

In Fig. 4 is plotted the experimental data together with results of preceding investigations. Obviously, in contrast to GaInN,[16] there is no bowing in AlGaN alloys lattice-matched to GaN.

In conclusion, we have reported the nonobservation of band gap bowing in wurtzite AlGaN alloys lattice-matched to GaN, for aluminum compositions ranging up to 0.2. Our measurement was made on a series of samples of various origins, grown by various methods.

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