Correlation between threading dislocation density and the refractive index of AlN grown by molecular-beam epitaxy on Si(111)

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We report on the influence of the structural properties on the refractive index of AlN films grown on Si(111) substrates by molecular-beam epitaxy using ammonia. The structural properties are assessed by reflection high-energy electron diffraction, atomic force microscopy, transmission electron microscopy, and x-ray diffraction. Refractive index values are deduced from room-temperature spectroscopic ellipsometry. Optical data analysis is performed using the Kramers-Krönig relation in the transparent spectral region, from 1.6 to 3.2 eV. Evidence is presented showing the influence of strain and dislocation density on the AlN layer refractive index. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1558217]

The application of GaN, AlN, and their alloys for the fabrication of short-wavelength optoelectronics devices[1] encourages the development of vertical-cavity structures consisting of such distributed Bragg reflectors (DBRs) as resonant microcavity light emitters, detectors, and Fabry–Perot modulators.[2,3] The first difficulty encountered in the epitaxial growth of high-quality nitride DBRs is the lack of knowledge of refractive indices, which play a key role in the design of such devices. Although nitrides have been actively studied for more than a decade, there are still significant inconsistencies in the published data for the refractive index as function of the photon energy and alloy composition.[4–6] This can be due to the different measurement methods used (prism coupling, ellipsometry, and interferometry), but also to the various growth procedures used to grow single-crystal nitride thin films. The strain state, surface roughness, and dislocation density of these thin films are believed to be very dependent on the growth methodology. Shokhovets *et al.*[7] have, for example, shown the strong influence on the optical data of the surface roughness and the nature of the interface layer between the nitride film and the substrate. In addition, Ozgür *et al.*[5] have suggested that the difference in the bowing parameter of AlGaN alloy between molecular-beam epitaxy (MBE)-grown samples and metalorganic chemical vapor deposition (MOCVD)-grown samples, explains the discrepancy in refractive index values. A key point which is specific to group-III nitride epitaxial layers is that nitrides layers are generally grown on highly lattice-mismatched substrates and therefore experience strain, which in turn leads to change in band-gap-energy-dependent features.[8,9] As a result, the optical properties will be slightly different in the transparent spectral region. This is true particularly when epitaxial growth is performed on a silicon substrate. Indeed, although the use of this substrate is of considerable potential interest for the development of low-cost devices, Si presents a large mismatch with GaN and related materials in terms of both lattice parameter and thermal expansion coefficient. Actually, group-III epitaxial growth on Si substrates requires the growth of a specific strain-balanced buffer layer.[10,11]

In this letter, the influence of the structural quality on the refractive index of AlN thin films grown on Si substrate is investigated in the transparent spectral region, through combined measurements of reflection high-energy electron diffraction (RHEED), transmission electron microscope (TEM), atomic force microscopy (AFM), x-ray diffraction (XRD), and spectroscopic ellipsometry.

The three nitride samples investigated in detail here were grown on Si(111) by MBE, using ammonia as the N precursor and standard solid sources for the group-III elements. Details of the growth procedure have been reported elsewhere.[10] Samples A and B were grown directly on Si substrate and consist of a 250-nm- and 1.055-μm-thick AlN layer, respectively. Sample C was also grown on Si, but consists of the following layer sequence, starting from the Si substrate: AlN(40 nm)/GaN(250 nm)/AlN(250 nm). The thickness of each layer has been determined during growth by *in situ* laser reflectivity and verified *a posteriori* by scanning electron microscopy.

The main results of the structural property investigation of the samples are first summarized. Both RHEED and AFM were used to assess the surface morphology of the AlN layers. For samples A and B, the AlN RHEED pattern evolution during the direct growth on Si indicates that the growth first follows a three-dimensional mode and progressively becomes two-dimensional (2D) after some tenths of nanometers. Conversely, the RHEED pattern of the final AlN layer of sample C reveals a 2D growth mode from the very first monolayers deposited on GaN.[12] These observations are confirmed by AFM images of the surface. Measurements of the

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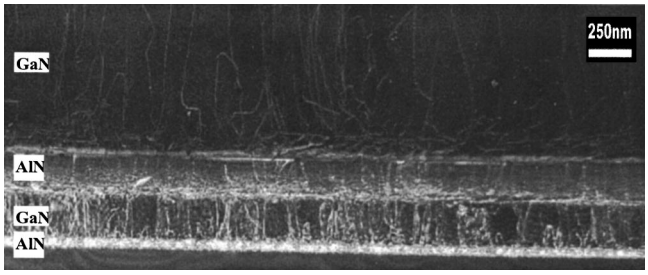

FIG. 1. A cross-sectional TEM image of a layer sequence of AlN(40 nm)/GaN(250 nm)/AlN(250 nm)/GaN.

rms roughness, typically 0.4 nm, from a 3×3 μm^2 scan, for both samples A and B, do not show a dependence on the thickness of the AlN grown directly on Si layers. The surface layer of sample C is also very flat, having rms roughness, calculated from a 3×3 μm^2 scan, of less than 0.9 nm. A cross-sectional TEM image of a sample corresponding to the same layer sequence as sample C, but upon which a thick GaN layer has been subsequently grown, is reported in Fig. 1. The epitaxial sequence, starting from the Si(111) substrate, is the following: AlN(40 nm)/GaN(250 nm)/AlN(250 nm)/GaN. One can first note that the three interfaces clearly contribute to the reduction of the dislocation density in the final GaN layer. One can also remark that in the AlN layers, dislocations, tilting, or bending are not observed, contrary to the case of GaN layers where interactions between dislocations leads to their reduction.[12] A point of particular interest here is the strong difference between the dislocation density in the AlN layer directly grown on the Si substrate and the second one grown on the GaN intermediate layer. Even if the interface with Si is sharp and no amorphous interfacial layer can be observed[12] for the first grown AlN layer (which is the case for samples A, B, and C), a highly defective region with a very high threading and misfit dislocations density is present due to the large lattice mismatch between AlN and Si. The dislocation density in the AlN layer directly grown on Si was estimated from plane-view TEM images for samples A and B [Fig. 2(a)], and is about 5×10^{11} cm^{-2}. On top of such layers, the GaN growth allows a reduction in dislocation density in the subsequent AlN layer by a factor 10 to 20, as shown in Fig. 2(b) corresponding to sample C. Thus, one would expect a significant difference between the refractive indexes of samples A, B, and C if dislocations play a role.

Figure 3 reports the index of refraction measured at 300 K as a function of photon energy in the range of 1.6 to 3.2 eV for samples A, B, and C. It can be seen that there is a noticeable difference in refractive index values among the three samples, and especially between sample C and the two others. Since the index of refraction is fundamentally linked to the band-gap energy,[13] one can postulate that observed differences in refractive index values is due to different band gap energies for the three AlN layers examined. Let us first consider the difference between samples A and B. During the cooling of the sample after the growth, an extensive stress appears due to the large thermal mismatch between Si and the nitride layers ($\approx$100%). The extensive stress is correlated to the thickness and the strain state of the layer at the end of the growth before the cooling procedure. Optical microscopy reveals that, unlike sample B, sample A, which is thicker

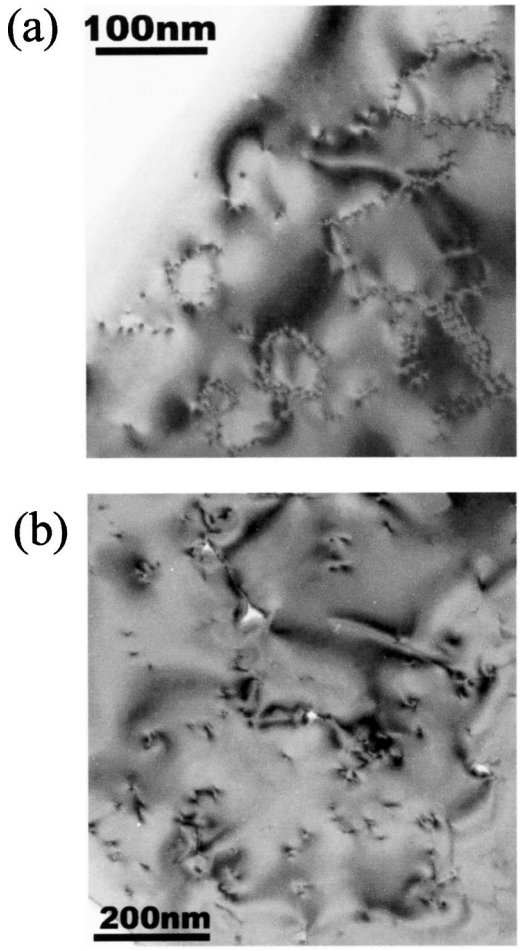

FIG. 2. Plane-view TEM images of (a) sample A and (b) sample C.

(1.055 μm), is partially cracked. We can therefore assume that sample A is more relaxed than sample B. This assumption is confirmed by XRD measurements, which show that samples A and B have out-of-plane ϵ_{zz} deformations of -1.7% and -2.2%, respectively. When the strain becomes more and more tensile, the upper valence band gap becomes Γ_7, and at the same time the band gap decreases.[8,14] This implies that the band gap of sample A is slightly larger than the one of sample B, and explains the observed difference in

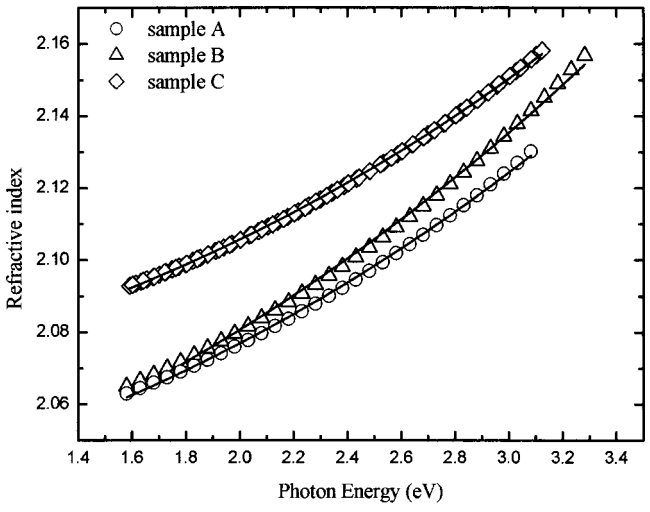

FIG. 3. Index of refraction of the AlN films vs photon energy at room temperature [solid lines are a fit using Eq. (1)].

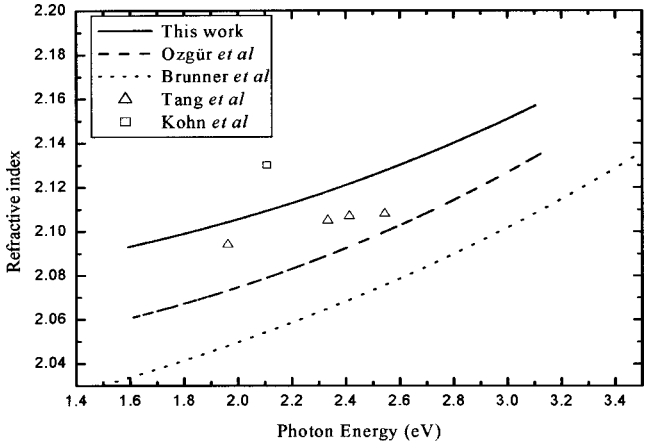

FIG. 4. Index of refraction of AlN film vs photon energy at room temperature determined from our ellipsometry experiments and in comparison with published data.

refractive index values. However, such an extensive stress effect cannot explain the large shift observed when comparing samples A and B with C. Indeed, sample C has an out-of-plane ϵ_{zz} deformation of -2%, and therefore the band gaps of the three samples are similar. Also, the rms roughness is similar and thus is not expected to appreciably influence the optical data.[9] One can therefore assume that the difference in the refractive index, comparing samples A and B with sample C, is mainly due to the difference in the dislocation density. The experimental data reported in Fig. 3 can be fitted using the Kramers–Krönig dispersion relation:[13]

$$n^2-1=\frac{K}{E^2}[2E_g{}^{1/2}-(E_g+E)^{1/2}-(E_g-E)^{1/2}] + \frac{G}{E_p^2-E^2}, \quad (1)$$

where E_g is the band gap, E_p the energy transition above the band gap, K and G are adjusting factors. Here, the first and the second terms represent contributions from the fundamental absorption edge and from high-energy bands, respectively. Due to strain effects, we take $E_g=6.1$ eV for samples A and C, and $E_g=6.08$ eV for sample B.[15] We assume E_p is independent of the strain and its value is taken at 11 eV.[16] The parameters K and G found from the fit are $K=2.791$, $G=376.42$ for sample A, $K=4.505$, $G=371.415$ for sample B, and $K=2.2029$, $G=393.633$ for sample C. The satisfactory data fit indicates that Eq. (1) provides an accurate formula for the refractive index dispersion values of the AlN layers. Figure 4 shows the refractive index fit of sample C, with published values[4,5,17,18] for comparison. The different strain state and dislocation density of the AlN epitaxial layers can presumably explain, at least partly, the divergence between the overall published data. Moreover, most of samples studied to date have been grown directly on a highly mismatched substrate (Al_2O_3), without strain-balanced structures, and present a dislocation density larger than in sample C. The optical properties, that is, the refractive index and optical propagation loss, are related to the density of defects as reported by Dogheche *et al.*[19] for AlGaN materials. These results similarly show the optical properties of the AlN layers are dependent on the dislocation density. Thus, in more complex structures corresponding to optoelectronic devices, the structural properties of a given layer are very much dependent on its place in the epitaxial layer stacking. This means that, in order to properly design and optimize vertical-cavity-based optoelectronic devices, it is necessary to measure the refractive index of the different layers used in a configuration that is very close or identical to that of the final device stacking.

In summary, a correlation between structural and optical properties is reported for AlN films grown on Si(111) by MBE. The structural quality of the films is shown to strongly influence the index of refraction as measured at 300 K by spectroscopic ellipsometry in the spectral range of 1.6 to 3.2 eV. Taking into account the different strain state and using a formalism based on the Kramers–Krönig dispersion relation, a formula is used to describe the refractive index of AlN for photon energies below the band gap. A comparison with other published data reveals significant differences in the refractive index values that are most likely related to variations in strain and dislocation density, which are a consequence of the various growth conditions used.

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