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Determination of Group III Nitride Film Properties by Reflectance and Spectroscopic Ellipsometry Studies

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Reflectance measurements under normal incidence of light and variable angle spectroscopic ellipsometry (SE) studies are applied for characterizing hexagonal GaN films grown by molecular beam epitaxy on both GaAs (111)B and 6H-SiC substrates. A comparison of the reflectance above the band gap with model calculations for a smooth film yields definitely the root mean square surface roughness. By analyzing the envelopes of the reflectance spectra, the influence of a buffer layer and/or a non-abrupt substrate/film interface is verified and qualitative conclusions about the interface properties are deduced which are emphasized by transmission electron microscopy studies. On this basis a suitable model for data fitting is established which contains only a small number of parameters to be adjusted. From the fit, the complex refractive index versus photon energy as well as the thicknesses of the active layer and the interlayers are obtained. A meticulous analysis of the SE data below the band gap shows that from those studies the extraordinary refractive index of hexagonal GaN can be determined for the first time.

1. Introduction

Multilayered epitaxial structures based on the hexagonal (h-) modification of InN, GaN, AlN and their ternary alloys are intensively studied nowadays due to their potential application for various optoelectronic devices in the visible/ultraviolet regions of the electromagnetic spectrum, such as light emitting diodes and injection lasers [1], photodetectors [2], and full color high resolution displays [3]. For their design and optimization, both detailed knowledge on complex refractive index $\tilde{N} = n + ik$ (n refractive index, k extinction coefficient) in the band gap energy range of the materials as well as methods to monitor the thickness and optical properties of these grown films are essential.

Since the first work by Ejder [4], a large number of studies has been devoted to the determination of n below and around the band gap of h-GaN. Due to the lack of large

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bulk crystals, most of the investigations by transmission/reflectance or spectroscopic ellipsometry (SE) measurements refer to layers grown heteroepitaxially on foreign non-lattice-matched substrates, for which the crystal quality depends critically on the used deposition technique. At low energies, the reported refractive indices show deviations up to 6% from each other and the energy dispersion close to the band gap differs up to an order of magnitude (for a review see Ref. [5]). However, these differences are probably less a result of the different h-GaN layer quality but caused by the single layer model (SLM) applied for data analysis in most works. In the SLM framework, the presence of interface layers (e.g. low-temperature buffer layers, amorphous layers, void formation, high density of point defects and threading dislocations or highly strained material) is generally neglected. Using a multi-layer model (MLM) we have discussed recently the influence of those layers with different refractive indices on the optical data [5], while first experimental results were reported in Refs. [6] and [7].

In this paper, we present for the first time results of the MLM analysis of reflectance and SE data for h-GaN layers grown on 6H-SiC substrates. Although for this system high GaN layer quality is expected due to the relatively low lattice mismatch, the necessity of such an analysis is evidenced by comparing the spectra with those found for h-GaN on GaAs(111)B substrates. Finally, the influence of the anisotropy of refractive indices on the SE studies is discussed.

2. Experimental Details

The undoped h-GaN films were grown by molecular beam epitaxy using a rf activated plasma source to provide atomic nitrogen and an elemental source for Ga. Oxide removal for the (111)B-oriented n^+ GaAs substrates was carried out at 620 °C under As flux. Then, for the two samples mg458 and mg459 a low temperature nucleation layer was generated at 610 °C by 10 min. nitridation and initiated growth, respectively, while mg448 was directly heated to the growth temperature of 710 °C. All samples were grown under N-rich conditions with an identical N/Ga ratio [7].

Sample g235 is a single crystalline h-GaN grown on the Si-terminated (0001) surface of an on-axis 6H-SiC substrate. Cleaning of the substrate from the residual oxygen was carried out under optimized silicon flux at 950 °C until Kikuchi lines become visible. It should be noted that this pretreatment does not remove the scratches from the polished substrates [8] which might contribute to the interlayer formation as demonstrated below. Growth was initiated at 500 °C by opening the Ga and N shutters simultaneously. Then, substrate and Ga cell temperature were increased gradually until the final growth temperature of 700 °C was reached. GaN epitaxy at high temperature was interrupted after 30 min until the reflection high energy electron diffraction pattern got sharper.

Double crystal X-ray diffraction (XRD) using CuK_α radiation showed the characteristic patterns of h-GaN with a *c*-axis orientation of the films being normal to the substrate for all samples. Neither XRD nor low temperature photoluminescence measurements yielded any evidence for the additional formation of cubic GaN. The root mean square (rms) roughness δ was determined by atomic force microscopy (AFM). Cross-sectional transmission electron microscopy (TEM) examination was carried out on g235 using JEOL JEM-3010.

Prior to the optical studies at room temperature all samples were treated by methanol which was shown to remove the organic contaminants from nitride surfaces [9]. For the reflectance studies under nearly normal incidence, light of a Xe arc lamp was dispersed through a 0.64 m focal-length monochromator, the spectral resolution was set to 0.3 nm. To achieve high sensitivity, especially in the range of the reflectance minima, the monochromatic radiation was chopped mechanically and finally amplified by a lock-in system. In order to avoid the correlation between the layer thickness and refractive index below the band gap of GaN, the ellipsometric parameters Ψ and Δ were measured in two configurations: spectral scans at fixed incidence angles, and angular scans at different photon energies. Finally, the optical properties of both types of substrates were determined separately by SE and used in the following analysis.

3. Results and Discussion

Reflectance spectra taken below and above the absorption edge of GaN (3.4 eV) exhibit some characteristic features being related to the properties of the boundary epilayer/substrate and surface roughness [5 to 7]. In the following section we discuss how to notice these effects by comparing experimental data with model calculations for the GaN/GaAs system. Then we apply the established model for data analysis to the GaN/SiC system where the effects are much less distinct.

3.1 h-GaN on GaAs(111)B substrate

The solid line in Fig. 1 shows a typical theoretical result of the reflectance for a h-GaN film (1.6 μm thick, $\tilde{N}$ data from Refs. [6] and [10]) on GaAs substrate if no interface is assumed (SLM). The reflectance of the GaAs substrate (dashed line) and the envelope of the interference extrema (dotted lines) are shown for comparison. The following peculiarities should be noticed. First, the envelopes are independent of the layer thickness. Second, below 2.8 eV the envelope of the maxima coincides with the reflectivity

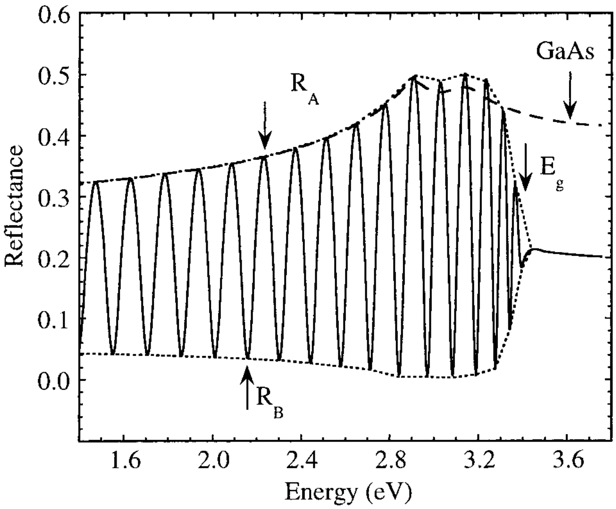

Fig. 1. Calculated reflectance (SLM) for a 1.6 μm thick h-GaN film on GaAs substrate (solid line). The dashed and dotted lines represent the reflectance of the substrate and the envelope of the interference extrema, respectively

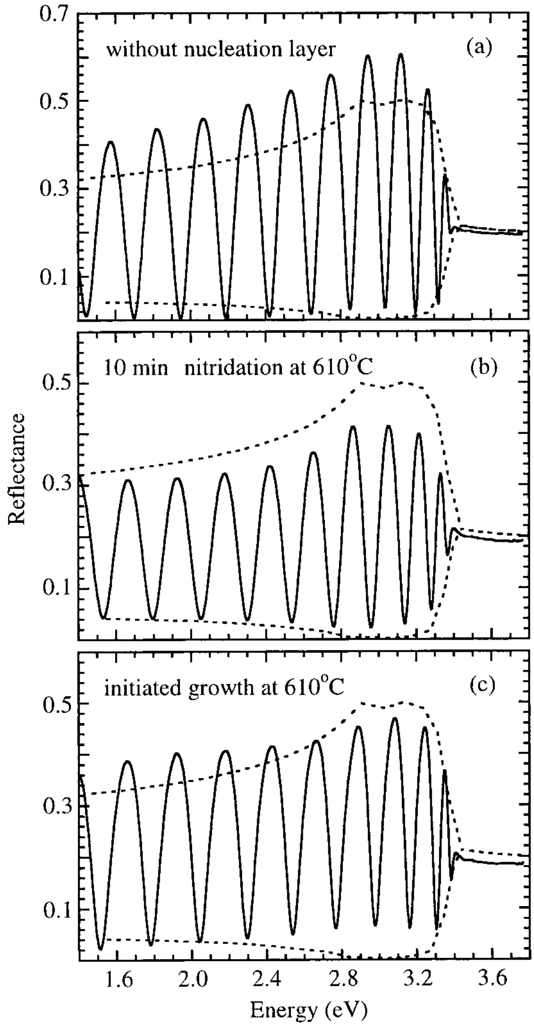

Fig. 2. Experimental reflectance spectra (solid lines) for the three GaN on GaAs samples: a) mg 448, b) mg 458, and c) mg 459. The calculated envelopes of Fig. 1 are shown for comparison

of GaAs (R_A). This can serve as a test for the applicability of the SLM since it does not depend on the GaN refractive indices. Third, the only information regarding n can be obtained by analyzing both the spacing between the oscillations (dispersion) and the height of the minima R_B (absolute value) [5].

Fig. 2 shows the experimental reflectance spectra for the three samples grown on GaAs(111)B substrates. The envelopes of Fig. 1 are shown for comparison by the dotted lines. Characteristic deviations to the SLM are observed over the whole investigated range. Above the band gap the difference between the experimental and theoretical data characterizes the influence of surface roughness which leads to lower reflectance R compared to a smooth film (reflectance R_0). For the data analysis the effect can be taken into account by

$$R = R_0 \exp\left(\frac{-16\pi^2 n_{\mathrm{amb}}^2 \delta^2}{\lambda^2}\right), \qquad (1)$$

where λ and n_{amb} denote the photon wavelength and the refractive index of the ambient medium, respectively [11]. Since all quantities except δ are known it is possible to obtain optically a measure for the surface quality. Fig. 3 emphasizes this possibility by the comparison of the optical data (δ_{Refl}) with the results of AFM studies (δ_{AFM}) for at least thirteen samples (the three samples presented here exhibit δ_{Refl} values between 8 and 10 nm). A linear relation is unambiguously found. The AFM data are always higher which is explained by a deviation of the real roughness distribution from a random one assumed for the derivation of Eq. (1). Second, the investigated areas are very different, $10 \times 10\,\mu m^2$ for AFM and 1 mm diameter for the optical studies.

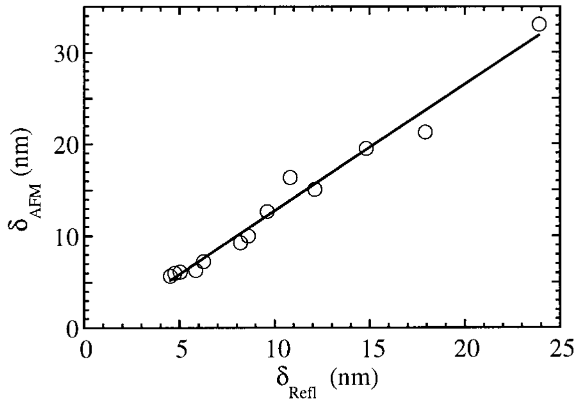

Fig. 3. Comparison of rms roughness determined by reflectance and AFM studies

Next we discuss the peculiarities in the below band gap range in Fig. 2. Neither for the untreated sample nor for the two others the reflectance maxima coincide with envelope. Investigations in media with different refractive indices n_{amb} evidenced that surface roughness influences the reflectance spectra in this range as well but can also be described via Eq. (1). Thus for example, an identical sample as in Fig. 2a, however with a smooth surface, would exhibit an even higher reflectance in contrast to the SLM. Generally, all spectra can be only explained by the formation of an interface layer with optical properties being different to both the substrate and the GaN layer. This makes the application of MLM recommendatory. As demonstrated in Ref. [7] an enhanced reflectance in the maxima (Fig. 2a) is a result of an interface refractive index n_{int} which is lower than for GaN (n), while the relation $n < n_{\mathrm{int}} < n_{\mathrm{GaAs}}$ leads to a reduced reflectance (Fig. 2b).

Under growth conditions used here (N-rich without As flux), the main origin of such interlayers is the formation of pits (voids) at the GaAs substrate surface with a sharp boundary to the GaN layer as TEM studies revealed [6,12]. The refractive index for a

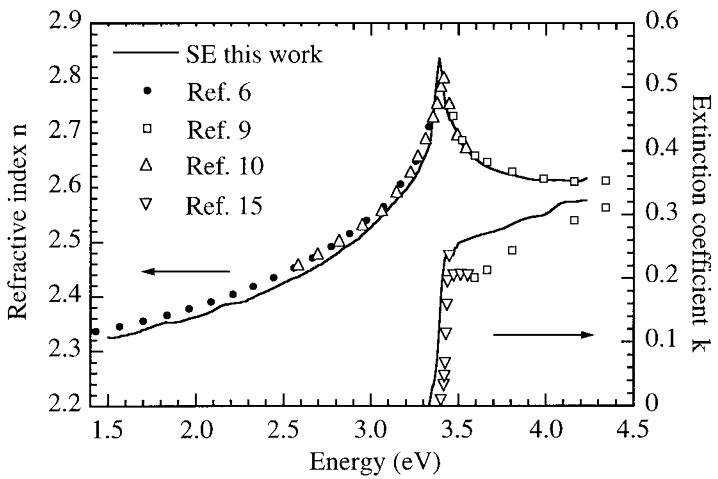

Fig. 4. Real and imaginary part of the complex refractive index for h-GaN. The full linesare obtained from the SE studies of GaN on 6H-SiC using an isotropic model for data analysis

Table 1
Void fraction in the substrate/film interface for samples grown on GaAs

sample	substrate		
	pretreatment	f_{sv}	f_{nv}
mg448	–	0.64	0.26
mg458	nitridation	0.15	0.01
mg459	initiated growth	0.53	0.17

GaAs-with-voids layer can be calculated by Bruggeman effective medium approximation (EMA) [13] considering the void fraction f_{sv} as a parameter dependent on growth conditions. Finally, it should be noted that good agreement between experimental data and fit results can only be achieved if besides a disturbed GaN layer close to the interface is introduced [7, 14]. Its lower refractive index is attributed to the high density of point defects and dislocations and can be modeled again by EMA (GaN-with-voids, void fraction f_{nv}).

Having established a suitable MLM (GaAs substrate, two interface layers, thick GaN, surface roughness) we were able to fit the spectra of all samples using identical $\tilde{N}$ data as shown by the symbols in Fig. 4. The adjusted void fractions are presented in Table 1. Compared to the directly grown sample (mg448), the nitridation process carried out for mg458 reduces strongly the void formation at the GaAs surface and increases the quality of the GaN layer in the interface region.

3.2 h-GaN on 6H-SiC substrate

The experimental reflectance data for the 680 nm thick h-GaN film on 6H-SiC substrate (g235) and the substrate reflectance are represented in Fig. 5 by the circles and dotted line, respectively. From the discussion in the previous section it is obvious that also in this case an extended interface was formed during growth. Qualitative interpretation yields the following results: (i) The envelope of the maxima crosses the reflectance curve of the substrate. This effect can not only be explained by a single EMA interlayer (a mixture of GaN with voids or SiC) since these media possess a weaker

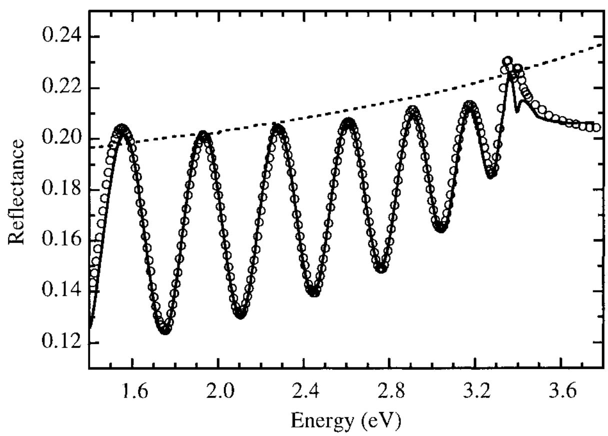

Fig. 5. Experimental reflectance data (circles) and fit result (full line) for sample g235 (h-GaN on 6H-SiC). The dotted line shows the substrate reflectance

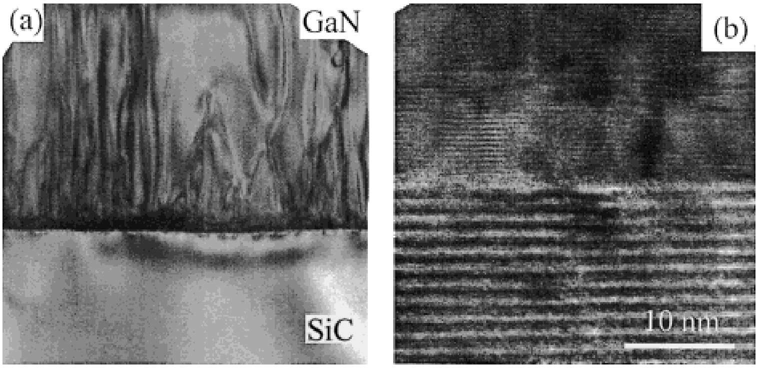

Fig. 6. a) Cross-sectional TEM micrograph for g235. The GaN film is 680 nm thick. b) The GaN/6H-SiC boundary is shown in higher resolution demonstrating the good quality of the epilayer in this region

energy dispersion of n than GaN [5]. (ii) The optical properties of the two interlayers are only slightly different compared to GaN or SiC.

In order to get insight how to model the interface layers cross-sectional TEM studies were performed. As can be seen from the bright-field TEM image in Fig. 6a the sample exhibits flat epilayer surface (δ_{Refl} 1.5 nm) and a well-defined epilayer/substrate boundary, however, with a small lateral modulation across the investigated range. Within 30 to 50 nm away from the boundary there is a clearly visible strain contrast (dark area) in the GaN epilayer and a high density of threading dislocations of mixed character which self-annihilate and decrease in density as the film becomes thicker. No evidence was found for either extended void formation or amorphous layers as evidenced by the higher-resolution image of Fig. 6b. Furthermore, it should be noticed that the GaN lattice planes are parallel aligned to the substrate even in the interface region.

Thus, for the fit shown in Fig. 5 an upper interlayer of disturbed GaN and a lower interlayer of SiC-with-GaN (due to the remaining scratches at the substrate surface which are probably filled with GaN [8]) were assumed. If the $\tilde{N}$ data determined in the previous section are used (symbols in Fig. 4) we get layer thicknesses of 48 nm (18 nm)

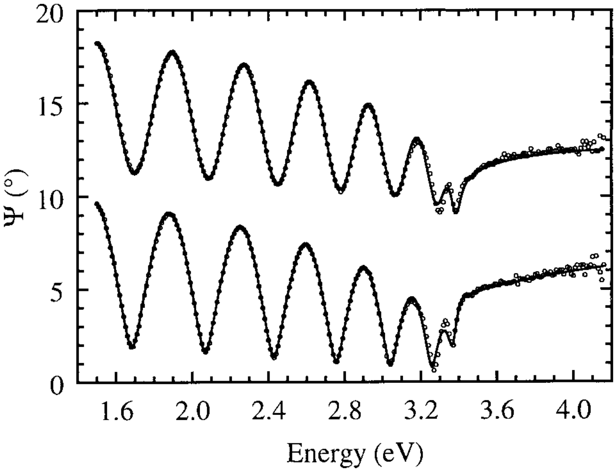

Fig. 7. Examples of the SE fit results for g235 using an isotropic model. The upper and lower curves were obtained at 75° and 70° angle of incidence, respectively

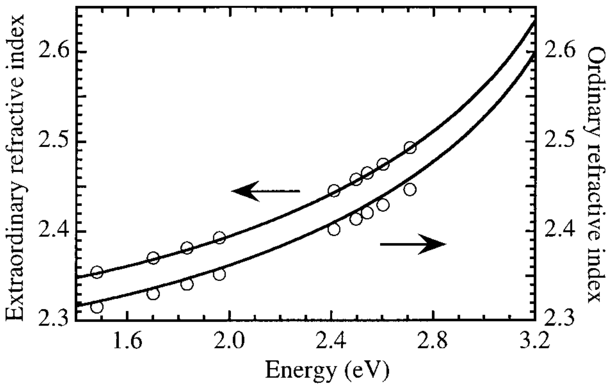

Fig. 8. Ordinary and extraordinary refractive indices for h-GaN determined by SE data analysis for an epilayer grown on 6H-SiC (full lines). The circles were taken from Ref. [18]

and 1.2% void fraction (9% GaN fraction) for the upper (lower) interlayer. The differences observed in the gap energy range of GaN (3.4 eV) are attributed to the $\tilde{N}$ data. Due to the excellent layer quality a higher n value might be expected in this range which is evidenced by the SE studies.

Figure 7 shows the fit results for the SE parameter Ψ for two angles of incidence (of total four) if the same MLM as for the reflectance analysis is used and if the anisotropic behavior of h-GaN is neglected (identical approach as reported in Refs. [10, 14, 16]). Similar good agreement is obtained for the other angles and all Δ data. Indeed, as the comparison in Fig. 4 demonstrates, the n values are found higher than for the samples grown on GaAs, however, in the below gap energy range n is determined appreciably lower. This effect is less a result of the different layer quality but caused by the isotropic model applied so far. If the anisotropy of h-GaN is taken into account the SE data in the energy range from 1.5 to 3.2 eV can be fitted yielding the values for the ordinary ($\mathbf{E} \perp \mathbf{c}$) and extraordinary ($\mathbf{E} \parallel \mathbf{c}$) refractive indices shown by the lines in Fig. 8. The surprising result is that both indices are higher than the isotropic one. This effect is confirmed by model calculations and will be discussed in a forthcoming paper [17]. Our data are strongly emphasized by recently published experimental results of Bergmann et al. who applied a prism-coupled waveguide technique for the determination of both indices [18] and the calculated high frequency dielectric constants $\varepsilon_{\perp}(\infty) = 5.21$ and $\varepsilon_{\parallel}(\infty) = 5.41$ [19]. The extrapolation ($\lambda \to \infty$) of the data presented here yields $\varepsilon_{\perp}(\infty) = 5.19$ and $\varepsilon_{\parallel}(\infty) = 5.33$.

4. Summary

We have demonstrated the high sensitivity of reflectance and spectroscopic ellipsometry studies for the characterization of hexagonal GaN films grown on foreign substrates. Clear evidence was given for the influence of surface roughness and the occurrence of extended interface layers by comparing experimental reflectance data (GaN on GaAs, grown with different substrate pretreatments) with single layer model calculations. The fit of the spectra yields both the complex refractive index for h-GaN as well as parameters characterizing the interface and/or surface region. The investigations of a h-GaN film on 6H-SiC substrate revealed that also for this system a multi-layer data analysis has to be performed. The interface formation was confirmed by transmission electron microscopy studies. By SE data analysis we determined the refractive index for this epilayer which was found higher in the gap energy range than for the films grown on GaAs. This emphasizes the superior crystalline quality of the whole layer. Finally, for the first time results were presented on the determination of the extraordinary refractive index for h-GaN by SE.

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