Structure and morphology evolution of ALN films grown by DC sputtering

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Abstract

Aluminium nitride (AlN) films have been deposited on (100) oriented silicon substrates by dc reactive magnetron sputtering for different deposition times (10 ≤ t ≤ 200 min) at a constant growth rate of 5 nm/min. The films have been characterized by tapping mode atomic force microscopy (AFM) and X-ray diffraction (XRD) techniques in order to study the correlation between the film growth and structural properties. The film growth has been characterized through the analysis of the temporal evolution of surface roughness, σ, obtained from the AFM data. The XRD measurements have shown the presence of a polycrystalline hexagonal phase for all film thicknesses. For t < 40 min, the films are composed of (002) textured regions embedded in a less ordered matrix. In this regime, the steep change of σ with the deposition time as σ ∝ t is consistent with an unstable growth mode. In contrast, for 40 ≤ t ≤ 200 min, a homogeneous structure with hexagonal (002) texture is mainly present, and the film roughening considerably decreases to σ ∝ t0.37 due to growth stabilization. Hence, the XRD and AFM data have revealed the existence of two growth regimes, and indicate that there is a strong link between the film structure and crystallinity and the film growth mode.

Keywords: Reactive sputtering; Aluminium nitride; X-Ray diffraction; Atomic force microscopy

1. Introduction

AlN thin films have attracted great interest because of their appealing properties such as high values of hardness and thermal conductivity or high resistance to temperature even in hostile environments. Therefore AlN films have been widely used as protective coatings [1,2]. Besides, AlN is a group III–V semiconductor with a large optical band gap, which renders it a promising candidate for applications in high power/high frequency devices, surface acoustic wave (SAW) filters, optical devices and insulating layers [3,4]. Moreover, it is employed in many applications as an intermediate buffer layer for optical and electronic devices as it improves the quality of the epilayer [5,6]. While the above-mentioned physical properties have been deeply studied, the AlN thin film growth dynamics in terms of the structure and roughness evolution has not been exhaustively addressed yet. In addition to the growth dynamics itself, such a study has important implications also in technological devices, in which the surface morphology or roughness can play an important role on the optical and electrical response [5,6]. The aim of this work is to study the dynamic of these growing surfaces and correlate it to their structural properties.

2. Experimental

AlN films were deposited onto (100) oriented silicon substrates at room temperature in a reactive DC magnetron sputtering system. Silicon substrates were sequentially cleaned in ultrasonic baths of trichloroethylene, acetone and ethanol for 15 min each and dried in dry nitrogen before being loaded into the deposition chamber. A high-purity (99.999%) aluminium target was operated at 100 W DC cathode power in an Ar and N2 gas mixture (29.5 sccm total flow: 20% Ar, 80% N2). The vacuum pumping system, composed of a turbomolecular pump backed by a mechanical pump, provided a base pressure lower than 10−4 Pa. The working
pressure was 0.4 Pa. These deposition conditions provided AlN films with $x$ close to 1 and enabled us to grow AlN films with a controlled stoichiometry at a constant growth rate of 5 nm/min [1]. In order to fully study the film growth dynamics, the films have been deposited for gradually growing times ranging from 10 to 200 min.

The atomic structure inside the layers was examined by X-ray diffraction (XRD). The XRD measurements were performed at CuK$_\alpha$ wavelength on a Siemens D 5000 diffractometer both in Bragg–Brentano (BB) and surface-sensitive grazing incidence (GI) modes at different $\alpha$ incidence angles fixed at 0.5, 1.0 and 1.5° in the latter case.

Surface morphology of the layers was analysed by atomic force microscopy (AFM) with a Nanoscope IIIa equipment (from Digital Instruments, CA) operating in tapping mode under ambient conditions. Silicon cantilevers with a nominal radius of curvature of 10 nm were employed. Finally, the film morphology was also analysed by scanning electron microscopy (SEM) by using a Hitachi S-2700 equipment.

3. Results

3.1. AFM and SEM results

AFM measurements enable us to characterize the film morphology evolution. In particular, we can analyse the film surface roughening and grain coarsening processes. Roughness data were obtained directly from the images by the AFM software whereas the typical surface grain size was obtained by averaging the different values found in the images. It is worth mentioning that the substrate surface was measured also by AFM giving an extremely low surface roughness close to 0.4 nm. Usually, the film surface roughness, $\sigma$, changes with the growth time, $t$, as $\sigma \propto t^\beta$, where $\beta$ is known as the growth exponent and describes the surface roughening process, i.e. how the surface grows in the direction perpendicular to the substrate plane [7]. The grain coarsening process is analysed by studying the change of the average grain size, $d$, of the surface granular structure with $t$. This dependence usually obeys the relationship: $d \propto t^{1/z}$. In this case, $1/z$ is known as the coarsening exponent and describes how the granular structure grows in the substrate plane [7].

In Fig. 1, we show the AlN film surface morphology measured by AFM for three deposition times, namely 24, 72 and 200 min. A granular morphology is observed. Since all images are displayed in the same vertical scale, the surface roughening and grain coarsening are clearly observed. The AFM measurements allow a quantitative analysis of the film growth dynamics by plotting the change of $\sigma$ with the deposition time (Fig. 2). From these data, we can distinguish two different growth regimes. The first one, up to 40 min of deposition, shows a steep surface roughening since $\beta = 1.0 \pm 0.1$. Moreover, the experimental data are consistent with an exponential dependence of the surface roughness with the deposition time, indicating an unstable growth mode [7]. The second growth regime for $t > 40$ min is characterized by a slower surface roughening since $\beta = 0.37 \pm 0.04$. Although two growth regimes are detected in surface roughness behaviour, a continuous coarsening process characterized by $1/z = 0.32 \pm 0.05$ has been found for grain size evolution with the deposition time (Fig. 2). Finally, the SEM cross-section analysis of the
thickest AlN film (Fig. 3) shows a clear columnar morphology. Accordingly, the top parts of the growing columns are measured by AFM, leading to the observed granular surface morphology. The coarsening process of the grains can be followed by AFM, but it cannot be observed by SEM because of its limited resolution.

3.2. XRD results

In order to better understand the difference between the two growth regimes revealed by AFM, we have analysed the same samples by XRD. The polycrystalline hexagonal structure of wurtzite type (file No. 25-1133 of JCPDS-ICDD diffraction database PDF-2) was detected in all films. The 002 diffraction was always the strongest one both in GI and BB diffraction patterns (Fig. 4a and b), indicating a preferred orientation of (001) planes parallel to the substrate with a rather broad angular distribution. Such one axial hexagonal texture with $c$ axis perpendicular to the substrate has been detected in AlN films previously [8,9].

The texture perfection can be judged from the shape of the rocking curve measured on the characteristic texture diffraction. While such a rocking curve for the second growth regime displays a simple symmetric bell-like shape (Fig. 5a), it is more complex for the films belonging to the first growth regime (Fig. 5b). The simple shape indicates a uniformly developed texture over the film volume while the more complex shape suggests the presence of well textured regions (i.e. columns) with aligned grains, producing the sharp ridge around the specular position, which are embedded in a
rather randomly oriented matrix contributing to the broad baseline.

4. Discussion

The results obtained show that there is a correlation between the film surface roughening and the film texturing processes. From our data it is evident that for short deposition times, when the film is formed by well textured as well as by randomly ordered regions, the film roughening is considerably higher than for long deposition times, for which the texture is well developed over the whole film volume. This difference could be due to a different local growth rate at the surface of both textured and randomly oriented regions. For short deposition times, the value of the $\beta$ growth exponent, close to unity, together with the coarsening value, indicate an unstable growth consistent with shadowing effects [10]. In this case, these shadowing effects arise from the fact that well textured regions would be growing faster, at the expense of the randomly oriented ones. As the deposition proceeds, the well textured regions keep growing and spread, covering finally the randomly oriented regions, so the film volume reaches a homogeneous columnar texture (Fig. 3). Under these conditions, the growth rate would be similar at the whole growing interface, leading to the observed reduction in the film roughening. The $\beta$ value, considerably less than 1, indicates that some sort of growth stabilization is operating. This value, together with that of the coarsening for the second growth regime, is not explained by any of the known growth models. In this case, the $\beta$ value is still higher than the values predicted by the models, which consider stabilization mechanisms such as surface diffusion or lateral growth [7]. Our results suggest that the observed behaviour is related to the high texturing degree of the film. Unfortunately, the variations in crystallinity in the polycrystalline growing films have been left out of the theoretical analysis [11], which prevents the identification of the main growth mechanism determining the surface growth morphology evolution.

5. Conclusions

Highly stoichiometric AlN films were deposited by dc magnetron sputtering under the same deposition conditions with deposition times ranging from 10 to 200 min. Two growth regimes have been found for growth times shorter and longer than 40 min. For the initial growth regime, the film is composed of well textured regions embedded in a rather randomly oriented matrix. Coarsening and $\beta$ values agree with an unstable growth, consistent with that observed when shadowing effects are present. A tentative explanation for this behaviour is based on the fact that differently textured regions occurring in the first growth regime would grow with different local growth rates leading to an unstable growth associated to shadowing effects. In the second growth regime, for $t>40$ min, the film volume is homogeneously textured. The smaller $\beta$ value implies that some sort of stabilization effect is operating, which is consistent with the fact that the film surface, at this growth stage, is formed only by well textured columns. However, the obtained $\beta$ and $1/\gamma$ values are not explained by any of the known growth models, which generally do not take into account the crystalline properties of the growing film. Our results do show that the growth behaviour is directly related to the texture development in the film volume and that the local degree of crystallinity is an important property that must be included in the modelling of the growth of polycrystalline films.

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References