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Laser technology for synthesis of AlN films: influence of the incident laser fluence on the films microstructure

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Abstract. Thin AlN films were synthesized by pulsed laser deposition at 800 °C by a KrF* excimer laser source ($\lambda = 248$ nm, $\tau = 25$ ns, 3 Hz) at fluences of 4.8, 8.6 and 10 J/cm² in nitrogen ambient at a dynamic pressure varying from 0.1 to 10 Pa. The film microstructure was studied by X-ray diffractometry, while the film surface morphology of AlN films was examined by AFM imaging. At the low laser fluence (4.8 J/cm²), a stable hexagonal AlN phase was formed, while at the intermediate laser fluence (8.6 J/cm²), both hexagonal and metastable cubic crystallites were observed. At the high laser fluence (10 J/cm²), the films were “XRD amorphous”. The surface roughness shows a tendency to increase with the increase in the nitrogen pressure. The root-mean-square roughness values are 0.2 - 5 nm, depending on N₂ pressure.

1. Introduction

Thin films of AlN with definite microstructure are essential for many applications in surface acoustic wave and UV optical devices. Moreover, AlN films deposited on Si are promising as microelectronic components thanks to their high dielectric constant and thermal conductivity, thermal and chemical stability, and good lattice match with SiC and GaN. However, obtaining AlN films with definite structure and crystalline quality still remains a challenge for most deposition techniques, such as metal organic chemical vapor deposition [1], reactive magnetron sputtering [2,3] and pulsed laser deposition (PLD) [4-7]. Extensive research is still necessary to find out exactly how the crystallographic structure and degree of crystallinity of AlN films depend on the film preparation method and conditions. Vispute *et al.* reported the synthesis of h-AlN with (0001) orientation [6], while Zhu *et al.* obtained cubic AlN films [8]. Metastable c-AlN was found in films synthesized by nitrogen-ion-assisted pulsed laser deposition [7]. Our recent investigations of AlN films prepared by pulsed laser deposition revealed that films deposited at low incident laser fluence on the target surface (3.7 J/cm²) possess cubic polycrystalline structure [9,10] and the amount of nitrogen in the gas ambient during the

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deposition plays a decisive role in the crystallization process [10]. Using a higher laser fluence (10 J/cm^2) and varying the N_2 pressure, amorphous films with AlN nanocrystallites could be grown [11].

Extending the findings of our previous investigations, the present work is concerned with synthesis of AlN thin films by pulsed laser deposition in nitrogen varying the incident laser fluence on the target ($4.8 - 10 \text{ J/cm}^2$) and the N_2 pressure ($0.1 - 10 \text{ Pa}$) in the vacuum chamber. The films microstructure was analyzed by X-ray diffractometry (XRD) and the film surface morphology was examined by atomic force microscopy (AFM). The results obtained are discussed in relation to the deposition conditions.

2. Experimental details

AlN films were PLD synthesized on single crystalline Si(100) substrates. The target was bulk polycrystalline stoichiometric AlN with 99% purity and the distance to the substrate was set at 5 cm. After loading the substrate and target, the deposition chamber was pumped down to a residual pressure $\sim 10^{-5} \text{ Pa}$ and the substrate was heated up to 800°C for 1 hour, aiming at decomposing the native SiO_2 on the Si surface. The substrate temperature was kept at 800°C during deposition, providing conditions for crystalline growth of AlN. The ablation was performed by a COMPEXPro 205 pulsed KrF* excimer laser source (248 nm, 25 ns) operating at an intensity of $\sim 4 \times 10^8 \text{ W/cm}^2$ and repetition rate of 3 Hz. The incident fluence was changed from 4.8, 8.6 up to 10 J/cm^2 . The target was rotated at 0.3 Hz during multi-pulse laser irradiation in order to avoid piercing. For the deposition of one film, 20 000 consecutive laser pulses were applied, which resulted in films thickness ranging from $0.2 - 0.6 \mu\text{m}$ with a tendency to decrease when increasing the nitrogen pressure and/or incident laser fluence.

To characterize the films structure, X-ray diffraction (XRD) measurements were performed on a large-angle ($2\Theta = 0-90^\circ$) Philips X'Pert equipment. The XRD patterns were identified using ASTM files [12]. We should note that due to the film thickness varying with the deposition conditions, the Bragg peaks of the crystalline phases in the thinner films are very weak and the signal-to-noise ratio is greater.

The surface morphology was studied by recording AFM images on an NT-MDT (Russia) Solver type scanning probe microscope. The surface scanning was performed in a semi-contact mode to avoid altering the sample. Cantilevers with typical force constant of 5.5 N/m , curvature radius of 10 nm and resonant frequency of 130 kHz were used.

3. Results and discussion

Typical X-ray diffraction patterns of the PLD AlN films deposited at different conditions are shown in figure 1. Although the XRD data analysis was difficult due to the small amount of film material and the presence of strong Bragg peaks originating from the Si substrate, peaks with low intensity related to the crystalline AlN phase were detected within the broad band of amorphous AlN phase.

In the case of AlN films deposited at an incident laser fluence of 4.8 J/cm^2 (figure 1a), the XRD patterns show the growth of AlN crystallites with a stable hexagonal phase [12]. From the observed broad peaks one can conclude that the size of the crystallites is on a nano-scale. However, precise estimation of the crystallites size is not possible from the given patterns because, on one hand, the number of the detected Bragg peaks is too small and, on the other, the broadening could also be caused by the highly defective crystalline structure. At a pressure of 1 Pa , the corresponding XRD pattern reveals the same AlN phase, but the ratio of the intensities of H(100) and H(002) Bragg peaks changes to the preferable $\langle 001 \rangle$ orientation of AlN crystallites in the films. A further increase of the N_2 pressure up to 10 Pa leads to development of a fully "XRD amorphous" film structure.

Upon increasing the laser fluence to 8.6 J/cm^2 , the shape of the XRD patterns (figure 1b) indicates that crystalline phases are also developed in the growing films. For the AlN films deposited at nitrogen pressure of 0.1 Pa , the Bragg peaks reveal that crystallites are formed in both hexagonal and cubic phases. The co-existence of two crystalline phases is in good agreement with our previous observations on AlN films deposited by PLD using a laser source with different parameters [9]. When increasing the pressure to 10 Pa , only metastable cubic AlN crystallites are formed in the amorphous

matrix. The size of crystallites cannot be determined due to the reasons discussed above, but it is reasonable to assume that the average size is below 100 nm.

When the deposition is performed with a laser fluence of 10 J/cm², the XRD patterns of the AlN films deposited at 0.1 Pa and 10 Pa become featureless (figure 1c), thus revealing that these films are “XRD amorphous”. This means that if there is a crystalline phase, the size of the nanocrystallites is too small or their density is too low to be observed with this XRD technique. Indeed, the TEM observations performed on these AlN films [11] have shown that the films deposited at 0.1 Pa are amorphous, but in the films deposited at 10 Pa the TEM micrographs visualize crystallites inside the amorphous film matrix growing in a metastable cubic AlN phase up to a few nanometers.

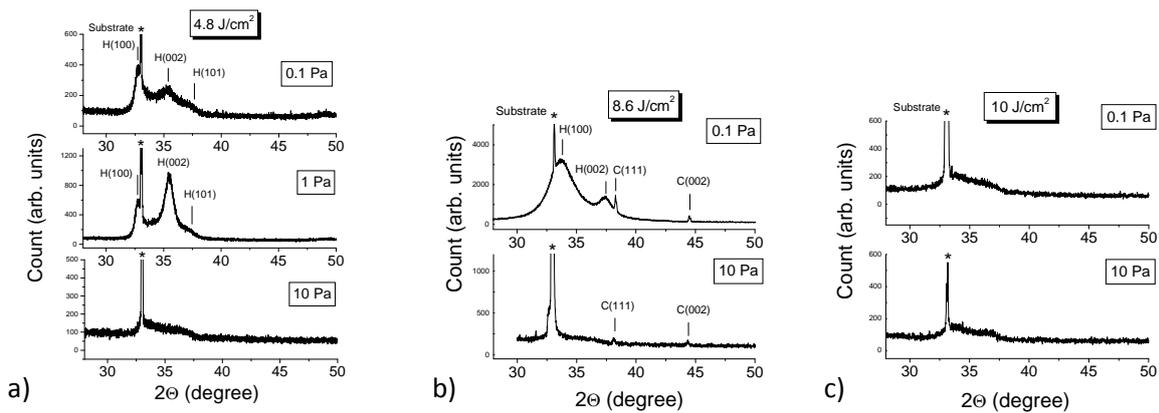


Figure 1. XRD spectra of AlN films deposited in N₂ ambient at different dynamic pressure and at incident laser fluence of 4.8 J/cm² (a), 8.6 J/cm² (b) and 10 J/cm² (c).

At first sight, these XRD results are contradictory to our previous studies on PLD AlN films [9,10] obtained at similar nitrogen pressures but at different laser source parameters. It should be emphasized that in earlier experiments we applied lower laser fluence (3.7 J/cm²), but the most essential difference in the deposition conditions was the pulse duration, which was more than three times as short ($t_{FWHM} = 7$ ns) as that ($t_{FWHM} = 25$ ns) used at the present experiments. Because of these differences, the growing processes and the resulting film microstructures cannot be compared.

The gradual modification of the films structure results also in a change of the surface morphology. Figure 2 represents typical AFM images from 1×1 μm² area surface scans of AlN films deposited at the conditions described. They illustrate well the columnar growth of PLD AlN films. At low and intermediate N₂ pressures, the films have smooth and uniform surface morphology, very low values of the root-mean-square (rms) roughness S_q indicating the amorphous nature of the films studied. At the high pressure of 10 Pa, the films still have a homogeneous surface, but the roughness sharply increases and small droplets of different size appear. In the case when 10 J/cm² laser fluence is used, this effect is stronger and it correlates well with the reduced growth rate at ahigh N₂ pressure.

The present XRD observations reveal that stable hexagonal phase of AlN can be formed in the case when the films are deposited at a considerably low laser fluence and a low nitrogen pressure. A possible explanation for this could be that, as the laser fluence is increased the species evaporated from the target acquire a much higher kinetic energy and, as they reach the surface of the growing film, this excessive energy can be transformed to adatoms obstructing the ordering in a crystalline network. This phenomenon is reflected well in the XRD results (figure 1), in particular for deposition at low nitrogen pressure (0.1 Pa). As the N₂ pressure is raised, the collisions between particles in a gaseous phase most probably reduce considerably the kinetic energy and, hence, the adatoms’ mobility

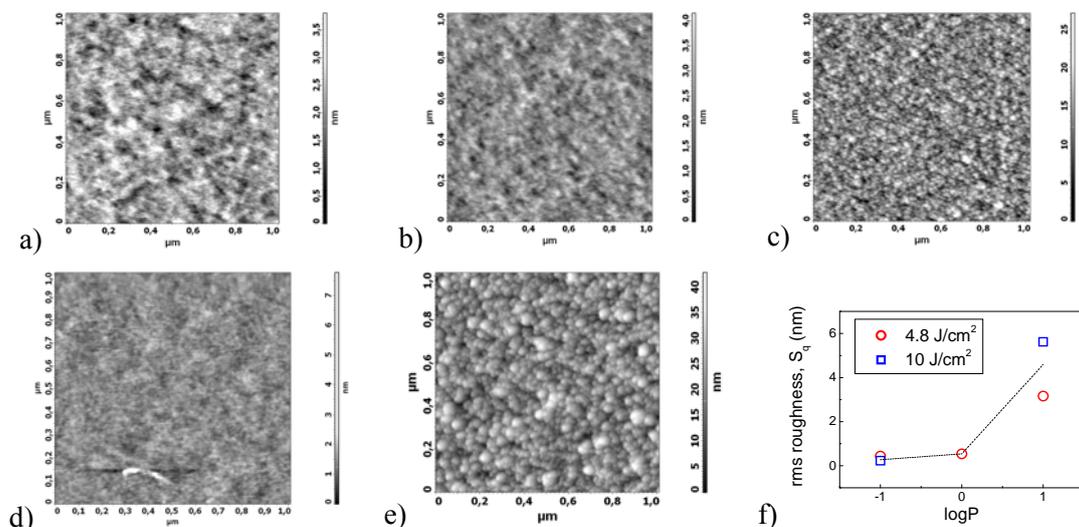


Figure 2. 2D AFM images from a $1 \times 1 \mu\text{m}^2$ surface area of AlN films (a-e) deposited with laser fluence of 4.8 (a,b,c) and 10 J/cm^2 (d,e) at a N_2 pressure of 0.1 Pa (a,d), 1 Pa (b) and 10 Pa (c,e). The rms surface roughness S_q values are also given (f).

on the growth surface. This would facilitate the formation of crystallites. However, increasing the nitrogen pressure further leading to an excess amount of nitrogen atoms adsorbed on the surface of the growing film, larger amounts of nitrogen atoms are build in the growing film volume, thus retarding the crystallization process. According to our observations, this effect becomes dominant for films deposited at 10 Pa, where the film structure is “XRD amorphous” independently from the laser fluence (figure 1). In this case, the excess nitrogen atoms most probably form clusters leading to an irregular growth of film with columnar structure and to an increased surface roughness, as is observed (figure 2f).

Conclusions

The XRD and AFM studies of AlN films synthesized by PLD at the conditions described revealed that by varying the laser fluence incident on the target and the nitrogen pressure during deposition, nanocrystallites with different AlN phases and orientations can be formed in the amorphous AlN matrix. The tendency was found that stable hexagonal AlN phase can be formed only when the film deposition proceeds at low incident laser fluence and low nitrogen pressure. At high laser fluence (10 J/cm^2) the AlN film structure is “XRD amorphous”. The PLD AlN films studied exhibit homogeneous surface morphology and considerably low values of the surface roughness, being dependent only on the N_2 pressure. A possible explanation for the formation of the film microstructure is given.

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