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Influence of Ion Beam Irradiation on Crystallographic Structure and Surface Morphology of Aluminium Nitride Thin Films

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Abstract: Aluminum nitride (AlN) thin films have been synthesised by ion beam assisted deposition method, and the influence of ion beam irradiation on microstructure and surface morphology has been studied by thin film X-ray diffraction (TFXRD) and by atomic force microscopy (AFM). In order to elucidate the influence of ion beam irradiation on the crystallographic structure, 10 nm thick *c*-axis oriented films synthesised with the ion beam energy of 0.05 keV are irradiated with nitrogen ions of 0.5 keV (no aluminum evaporation) after deposition for the same period as the deposition time and this process is repeated until the film grows to 200 nm in thickness. The TFXRD results reveal that the "piled up" films do not show the oriented structure, but are disordered to the multi-oriented state. AFM observations reveal that (1) the films synthesised with the nitrogen ion beam of 0.05 keV show the extreme smooth surface and the surface roughness increases with increasing the nitrogen ion beam energy and (2) the film surface synthesised with the ion beam energy of 0.05 keV does not change drastically by 2-keV ion beam irradiation after deposition. These results show that synthesis with high energy ion beam makes AlN films disordered structure and rough surface. © 1998 Elsevier Science Limited and Techna S.r.l. All rights reserved

1 INTRODUCTION

Aluminum nitride (AlN) is one of the most promising materials for microelectronics and optoelectronics devices due to its unique properties, such as high electrical resistivity, high thermal conductivity and high hardness. With the development of various deposition techniques, it has become possible to synthesize AlN thin films on many kinds of substrates under various conditions.

Among deposition techniques, the deposition methods using ion beams, e.g. ion beam assisted deposition (IBAD) and ion beam sputtering deposition (IBSD), have the advantage of an independent and well defined control of the ion bombardment parameters.[1] Okano *et al.* developed a deposition method using electron cyclotron resonance dual-ion-beam sputtering[2–5] and studied the orientation control[2] and the dependence of the crystallinity and surface smoothness of AlN films,[5] and tried to apply AlN films to surface acoustic wave (SAW) devices.[3,4] Ogata *et al.* prepared AlN thin films with an IBAD method and studied the relationship between the crystalline growth of the films and the ion beam energy.[6] Wang *et al.* formed AlN thin films with IBSD of aluminium using pure nitrogen or a nitrogen (75%) and hydrogen (25%) gas mixture.[7,8] They studied the role of substrate temperature on film composition and optical properties,[7] and also investigated the surface morphology, composition and structure.[8]

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In a previous paper, the present authors synthesised AlN thin films at low temperature (around room temperature and at 200°C) with IBAD method, and studied the microstructure and nanometer-scale surface morphology of AlN films.[9–12] We reported that AlN films with uniform and smooth surfaces on the nanometer scale can be synthesised on silicon single crystal and soda-lime glass substrates,[10] and the changes in the surface morphology due to the deposition rate by atomic force microscope (AFM) observations.[12] In addition, it was reported that multi-oriented polycrystalline films composed of hexagonal AlN (10·0), (00·2) and (10·1) planes are synthesised when the ion beam energy is higher than 0.1 keV, while the *c*-axis oriented films are synthesised with the ion beam energy of 0.05 keV.[11] Although the change in the orientation state of AlN films from *c*-axis oriented to multi-oriented with increasing ion beam energy is already reported in the literature,[2,6] the mechanism of the change is not understood completely.

Recently, in addition, J. H. Edgar *et al.* studied the effects of ion beam energy on the properties of AlN films prepared by an IBAD method.[13] Although they have shown that the structure, stress, energy band gap and hardness of AlN films are changed with changing the ion beam energy, the ion beam energy was limited to below 200 eV. Thus, it is an exciting objective to study the ion beam irradiation effect in much higher energy region.

On the other hand, in applying AlN films to practical usage, e.g. insulating layers or coating materials in microelectronics fields, surface smoothness is of importance and thus it is worthy to study controllability of surface morphology by regulating deposition parameters in an IBAD method.

Therefore, in this paper, we attempt to synthesise AlN films by the IBAD method and study the influence of ion beam irradiation on the crystallographic structure and surface morphology of films in the ion beam energy region of below 2 keV.

2 EXPERIMENTAL

AlN thin films were synthesised with the IBAD method. Nitrogen gas (99.999% pure) and aluminum (99.99% pure) were used as an ion source and a target. Film synthesis was carried out on a silicon single crystal substrate, Si (100), placed into the load lock in a high vacuum chamber.

After evacuating the vacuum chamber to around 2.7×10^{-4} Pa, pure nitrogen gas was introduced to the ionisation chamber and nitrogen ions were generated by an arc discharge. Then a nitrogen ion beam was obtained with electric-field lenses for focusing and accelerating. The accelerating voltage was varied from 0.05 to 2.0 keV and the current density of the ion beam was approximately 70 μA cm^{-2}. Aluminum was evaporated by electron bombardment and the evaporation rate was monitored by a quartz sensor. The deposition rate was kept at 0.07 nm s^{-1}. Substrate temperature was kept at around room temperature.

Microstructure of each film was determined by thin film X-ray diffraction (TFXRD) with copper Kα radiation (RINT2500, Rigaku Co.). The sample was rotated at the speed of 50 rpm.

In order to study the influence of nitrogen ion-beam irradiation on change in the orientation state, the "piled-up" film was prepared as follows: (1) first, 10 nm thick *c*-axis oriented film is synthesised with the ion beam energy of 0.05 keV, (2) second the synthesised film is irradiated with nitrogen ion beam of 0.5 keV (no aluminum evaporation) after the deposition for the same period as the deposition time in the process (1) (approximately 2 min), and (3) the processes (1) and (2) are repeated until the film grows to 200 nm in thickness. This procedure was performed based on the assumption that the microstructure of each 10 nm thick film is maintained during the piling up process. The schematic chart of time sequence is shown in Fig. 1.

The surface of the films was observed by AFM. Observations by AFM were performed by the tapping mode in air with a NanoScope III (Digital Instruments) equipped with commercial silicon tips. The AFM images were acquired at 256×256 points per frame and were corrected by subtraction of the background slope.

3 RESULTS AND DISCUSSION

3.1 Microstructure

Figure 2(a) and (b) show a part of TFXRD patterns of the films, of 300 nm in thickness, synthesised with the ion bean energy of 0.05 keV and 0.5 keV, respectively. This 2θ scanning region is covered over the first three strong lines of hexagonal AlN, (10·0), (00·2) and (10·1) planes. As we reported in a previous paper,[11] the film synthesised with an ion beam energy of 0.05 keV is highly oriented along the *c*-axis and, in contrast, the film synthesised with the ion beam energy of 0.5 keV shows a multi-oriented state and the main crystalline growth is the (10·0) plane.

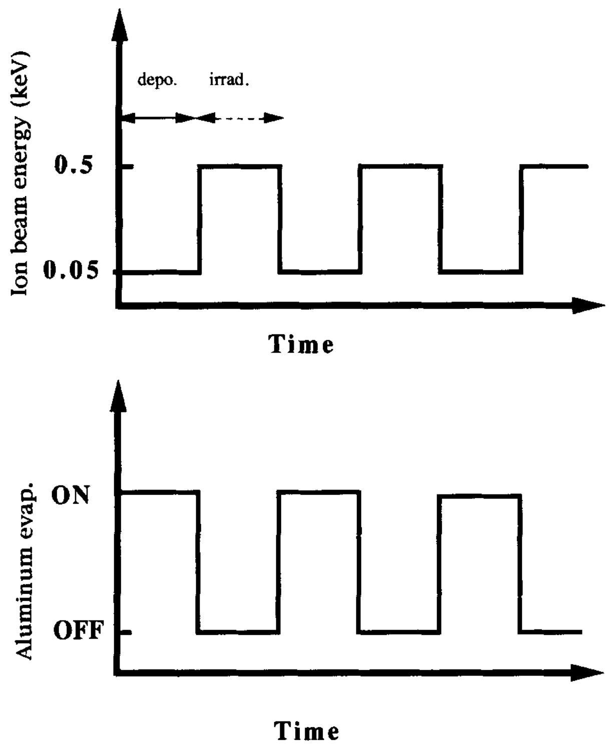

Fig. 1. Time sequence for synthesis of piled up films. As the first step, AlN film of 10 nm thickness is synthesised with the nitrogen ion beam of 0.05 keV and then the film is irradiated with the nitrogen ion beam of 0.5 keV (no aluminium evaporation) as the second step. These processes are repeated until the film thickness becomes approximately 200 nm.

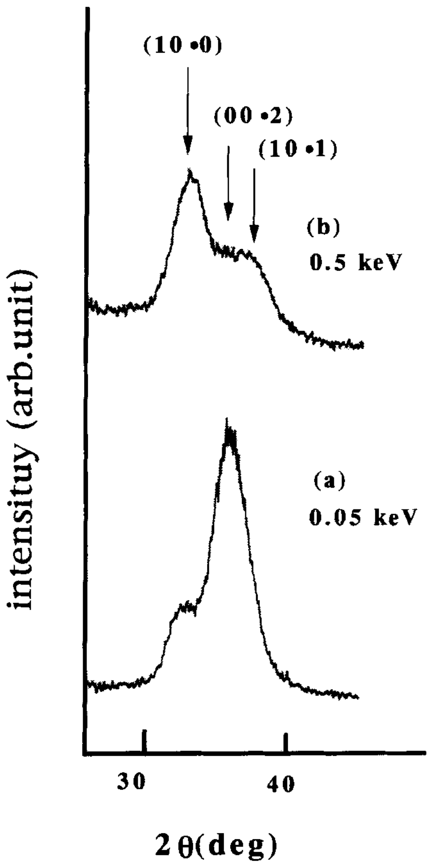

Fig. 2. XRD patterns of AlN thin films synthesised on the Si (100) substrate with the nitrogen ion beam of (a) 0.05 keV and (b) 0.5 keV.

Okano *et al.* reported that the AlN (00·2) peak intensity decreases with increasing nitrogen ion beam energy with a nitrogen ion beam of more than 250 eV.[2] Similarly, Ogata *et al.* observed a decrease in the AlN (00·2) intensity with increasing nitrogen ion beam energy of 3 to 20 keV.[6] Although the deposition conditions are different among the previous reports[2,6] and the present study, the tendency of decreasing the (00·2) intensity with increasing nitrogen ion beam energy agrees well with the previous results. Ogata *et al.* proposed that regions of quasi-high temperature and quasi-high pressure are produced at the point of collision and these local conditions affect the crystalline growth of the films.[6] However, they did not discuss a mechanism of decreasing the (00·2) intensity.

Figure 3 displays a part of TFXRD result of the "piled-up" film of 200 nm in thickness. It can be seen that the film does not show the *c*-axis oriented structure, although the ion beam energy was kept at 0.05 keV during deposition, and this pattern is similar to Fig. 2(b), which shows the film structure synthesised with the ion beam energy of 0.5 keV.

For explaining this change in the orientation state of films due to 0.5 keV nitrogen ion beam irradiation, two models are proposed; (1) the *c*-axis oriented film synthesised with the ion beam energy of 0.05 keV was changed to a multi-oriented state by some interaction between AlN crystallites and nitrogen ions during 0.5 keV ion irradiation, or (2) AlN crystallites with the (00·2) plane was etched selectively by 0.5 keV ion irradiation, resulting in the multi-orientation state.

With only the present experimental results, it is difficult to establish a mechanism of changing the orientation state due to nitrogen ion beam

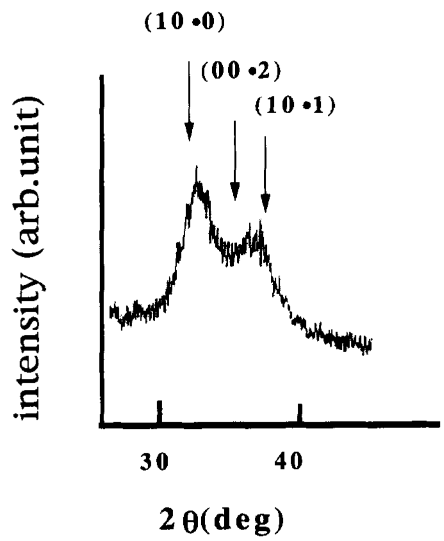

Fig. 3. XRD pattern of the piled up AlN films synthesised by the procedure shown in Fig. 1. Substrate is the Si (100) plate.

irradiation. However, it can be said that the *c*-axis oriented films are easily disordered by high energy ion beam irradiation.

The (00·2) plane of hexagonal AlN is the closest packing plane and a low-energy configuration.[14] In other words, the multi-oriented state is a high-energy configuration. When 0.5 keV nitrogen ion beam irradiates the *c*-axis oriented film, energetic ions impinge on the film surface and transfer their momentum to AlN crystallites. Receiving ions' momentum, some crystallites with the (00·2) plane may change their growth direction and some crystallites may be rearranged and/or be converted, resulting in the multi-oriented state. In addition, some nitrogen ions etch out AlN particles near the surface and some ions are implanted into AlN films.

Moreover, according to Ogata *et al.* regions of quasi-high temperature are locally produced at the point of collision between ions and film.[6] Suppose that this model is applicable to the present experimental conditions preparing the *c*-axis oriented AlN thin films, recrystallization occurs in the quasi-high temperature regions during cooling down, resulting in the multi-oriented film.

As a summary, the present experiments proved that the *c*-axis oriented film is easily changed to the multi-oriented state by irradiation with 0.5 keV nitrogen ions and more detailed study will be necessary to understand the mechanism of change in the orientation state.

3.2 Surface morphology

Figure 4(a) and (b) show the typical AFM images of the surface of AlN films synthesised with the nitrogen ion beam energy of 0.05 keV and 2.0 keV, respectively. From these images, it can be concluded that (1) the film synthesised with 0.05 keV nitrogen ion beam is covered with uniform particle-like features and the surface is extremely smooth and (2) the film surface synthesised with the nitrogen ion beam energy of 2.0 keV becomes rough and particle-like features seem to be aggregated.

The surface average roughness, R_a, of the films is plotted as a function of the ion beam energy and shown in Fig. 5. From this figure, it is clear that the surface roughness increases monotonously with increasing the nitrogen ion beam energy. This result indicates that the surface roughness of AlN films synthesised by the IBAD method can be controlled by regulating the nitrogen ion beam energy and it will be useful for practical applications of this film.

As a mechanism of the roughening of the surface with increasing the ion beam energy during

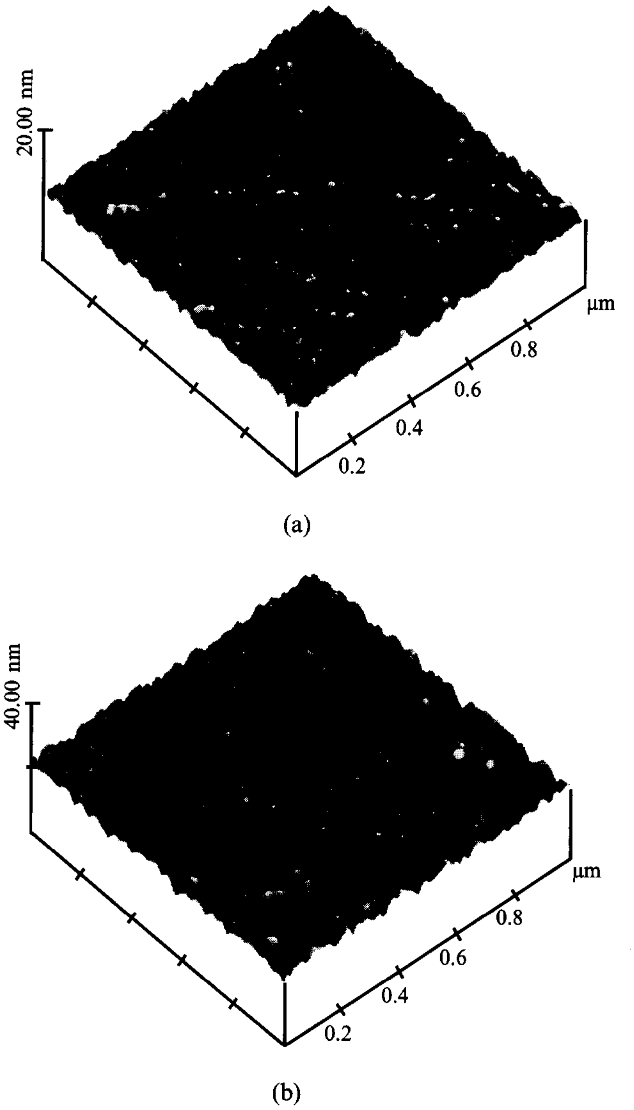

Fig. 4. AFM images of the surface of AlN films synthesised on the Si (100) substrate with the nitrogen ion beam of (a) 0.05 keV and(b) 2.0 keV. Substrate is kept at around room temperature.

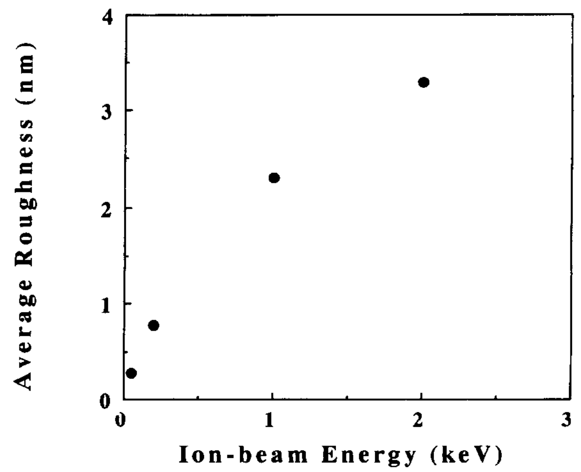

Fig. 5. The dependence of the average roughness of the films on the nitrogen ion beam energy. The values of roughness are calculated from the AFM data obtained from the scan of 1 micron square.

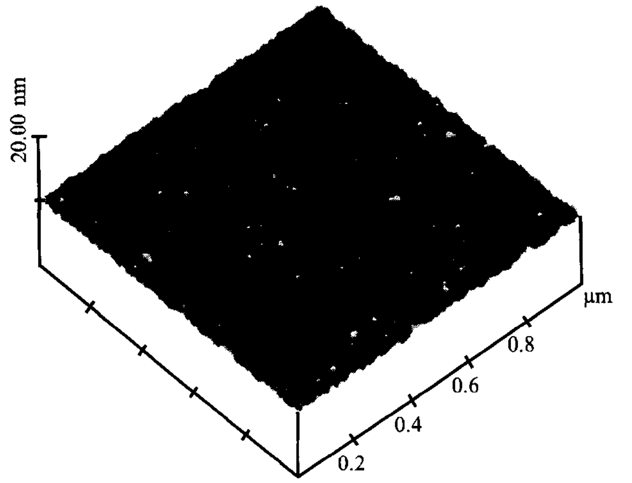

Fig. 6. AFM image of the surface of AlN films synthesised on the Si (100) substrate with the nitrogen ion beam of 0.05 keV and irradiated by the nitrogen ion beam of (b)2.0 keV after synthesis.

synthesis, the possible model is as follows: (1) disordering of the arrangement of AlN crystallites by receiving nitrogen ion energy; or (2) increase of etching of surface by ion beam irradiation during synthesis.

In order to study the etching effect by ion beam irradiation, the film synthesised with the ion beam energy of 0.05 keV was irradiated by nitrogen ion beam of 2.0 keV after the deposition during the same period as the deposition time. A typical AFM image of the surface of the films after irradiation with the 2.0 keV nitrogen ion beam is shown in Fig. 6. This AFM image reveals that the surface of the film after ion beam irradiation is not changed drastically from the surface before irradiation as shown in Fig. 4(a), and seems to be smoother rather than that before irradiation. This result suggests that the ion beam irradiation after deposition is not an important effect on the surface roughening.

It is summarised that (1) surface roughening occurs during deposition with high energy ion beam and (2) the rough surface is closely related to the disordered structure.

4 CONCLUSIONS

Aluminum nitride thin films have been synthesised by the IBAD method, and the effect of nitrogen ion irradiation on the microstructure and surface morphology of the film has been studied. The TFXRD studies reveal that the film synthesised with the ion beam energy of 0.05 keV shows the *c*-axis oriented state, but the oriented state is easily disordered by irradiation with the ion beam energy of 0.5 keV after deposition. The AFM observations show that the surface roughness increases monotonously of the AlN films with increasing the nitrogen ion beam energy and ion beam irradiation after deposition does not affect the surface morphology drastically.

Although the mechanisms of changing in the crystal orientation and surface roughness are not well established, the effect of ion beam irradiation on the orientation state and surface morphology becomes clear and this study will give some clue to full understanding of deposition process with use of energetic ion beam.

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