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Deposition Characteristics of AlN Thin Film Prepared by the Dual Ion Beam Sputtering System

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In this experiment, a radio frequency dual ion beam sputtering (DIBS) system was used to prepare aluminum nitride (AlN) films with a bottom Al electrode on a Si (100) substrate. After systematic testing of the processing variables, a high-quality film with preferred *c*-axis orientation was grown successfully on the Si (100) substrate with an Al target under 700 eV energy flux, $N_2/(N_2 + Ar)$ ratio of 55%, and 4×10^{-4} torr in vacuum. The characteristics of the deposited AlN thin films were studied by x-ray diffraction (XRD), scanning electron microscope (SEM), transmission electron microscope (TEM), secondary ion mass spectrometry (SIMS), and electronic spectroscopy for chemical analysis (ESCA). The surface roughness was also measured. It was found that AlN films prepared by DIBS at room temperature are better than those prepared at 300°C, and those prepared with an Al target are better than those prepared with an AlN target. The inferiority of AlN films prepared with AlN targets is due to the AlN bond being broken down by the ion beam source.

Key words: Dual ion beam sputtering (DIBS), aluminum nitride (AlN) film, preferred orientation

INTRODUCTION

Aluminum nitride (AlN) is a III-V nitride-based semiconductor material with a hexagonal wurtzite structure. Its characteristics are a wide direct band gap (~6.3 eV), high thermal conductivity, good chemical and thermal stability, good dielectric properties, very fast Rayleigh velocity,[1] and a high refractive index (>2.1) in the visible light range.[2]

The AlN film has a wide application in microelectronic devices because of its high thermal conductivity (320 w/m·k), high electrical resistance, and attractive mechanical properties.[3] Because its thermal expansion coefficient is similar to that of Si, AlN film has potential applications for metal-insulator-semiconductor structures.[4] In addition, AlN film is a candidate material for ultraviolet light emitters due to its wide band gap energy,[5,6] and its fast acoustic velocity (5,600 m/s) is especially attractive in high-frequency wireless communications such as GHz-grade surface acoustic wave devices.[7]

A variety of deposition methods have been used to prepare AlN films, including metal-organic chemical vapor deposition (MOCVD)[8] and physical vapor deposition.[9–13] Ion beam sputtering deposition[14] and ion beam–assisted deposition[15,16] have also been used.

Chemical vapor deposition, which has been employed to obtain polycrystalline or single crystalline AlN films, is likely a feasible method for continuous production. However, the high processing temperatures (higher than 1,000°C) induce large internal stresses in the films, which restrict the choice of substrate. In addition, a smooth surface can only be formed with difficulty by CVD due to the high grain growth rate at high deposition temperatures.[17,18] Previous reports also show that the stoichiometric AlN is difficult to obtain by CVD.[19]

The preparation of AlN films at low temperatures over a large variety of substrate can be done with the

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reactive sputtering technique, which allows the growth of highly oriented polycrystalline films with piezoelectric properties very similar to those of single crystalline films. Ion beam sputtering systems, such as ion beam–assisted deposition[20] and ion beam sputtering deposition,[21] are a relatively new method of preparing thin films. With the low temperature and pressure,[22] the mean free path of atoms is more important than the distance between the substrate and target, as the mean free path results in an increased energy transfer to the sputtered particles, and a consequently smoother surface and more easily controlled film composition.

In this work, the AlN films were prepared by a dual ion beam sputtering (DIBS) deposition system and the deposition parameters were systematically studied. The properties of the prepared AlN films were studied in detail using various analytical instruments, and the formation mechanism of (002) orientation AlN film was also investigated.

EXPERIMENTAL

The AlN with bottom aluminum electrode was prepared by the radio frequency DIBS system under varied parameters, such as variations in the temperature of deposition, in the concentration of nitrogen (N_2%), in the ion source voltage, and in the substrate temperature. The targets of 99.999% aluminum and AlN were used and an *n*-type Si (100) wafer was used as substrate.

Before the AlN films were deposited, the Si substrates were cleaned ultrasonically with acetone, alcohol, and deionized water. During deposition, the target was presputtered for at least 30 min to remove any surface contaminants. Then the shutter was opened to deposit an aluminum bottom electrode layer about 30-nm thick on the Si substrate under 600 eV. Nitrogen gas was then introduced to grow the AlN film with a growth time of 180 min.

The crystallinity and the preferred orientation of the AlN films were examined by x-ray diffraction (XRD). The microstructure was studied by a scanning electron microscope (SEM) and a transmission electron microscope (TEM). The compositions of the films and the chemical states of the aluminum and nitrogen were analyzed by secondary ion mass spectrometry (SIMS) and by electron spectroscopy for chemical analysis (ESCA), respectively. The surface roughness was measured by an atomic force microscope of Form Talysurf (Taylor Hobson, United Kingdom).

RESULTS AND DISCUSSIONS

The x-ray diffraction results for AlN film deposits prepared by DIBS under the various conditions are shown in Figs. 1–6. Because an AlN film with a strong (002) texture is desired, these figures reveal that AlN films prepared by an aluminum target

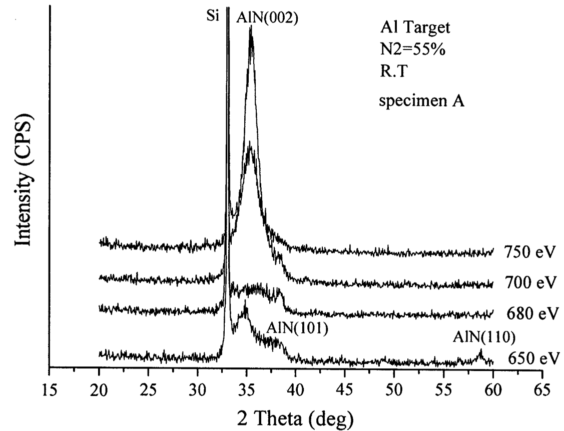

Fig. 1. XRD patterns of AlN films synthesized as a function of flux levels and Al target, using a fixed nitrogen concentration of 55% and no substrate heating. The specimen prepared at 700 eV is designated as specimen A.

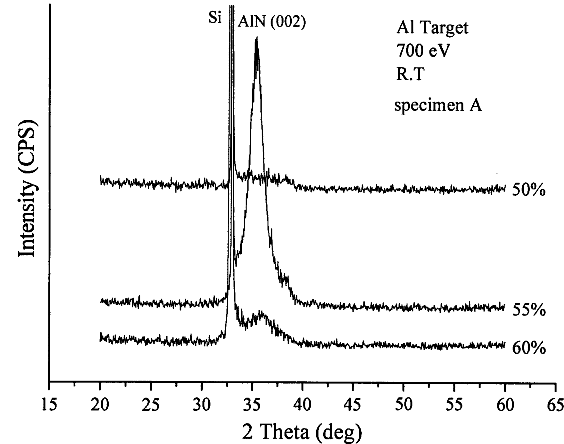

Fig. 2. XRD patterns of AlN films synthesized as a function of a N_2% concentration Al target used with a fixed ion source energy of 700 eV and no substrate heating.

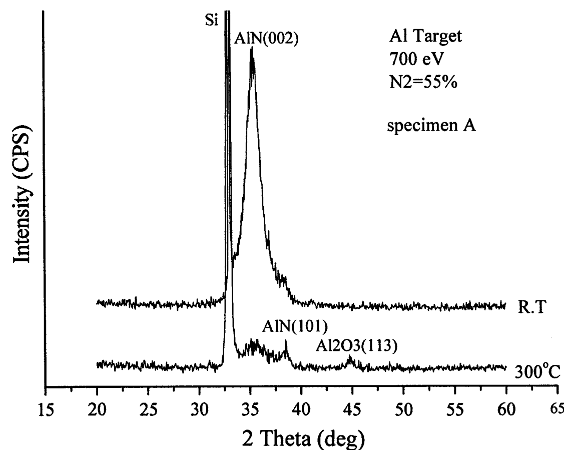

Fig. 3. XRD patterns of AlN films synthesized with and without substrate heating, using an Al target under fixed ion source energy at 700 eV and 55% nitrogen concentration.

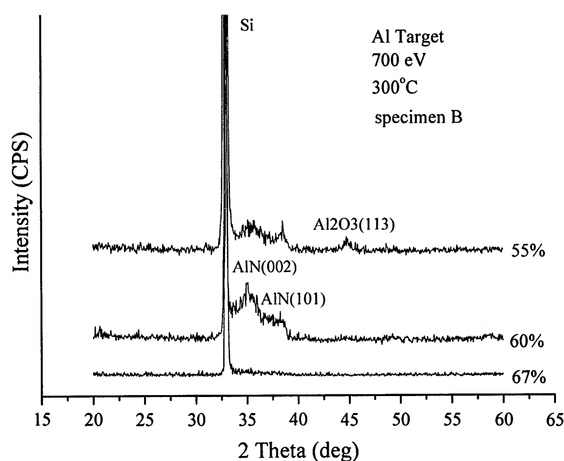

Fig. 4. XRD patterns of AlN films synthesized using a function of the N_2% concentration Al target with a fixed ion source energy of 700 eV and substrate heating to 300°C. The specimen prepared at 55% nitrogen concentration is designated as B.

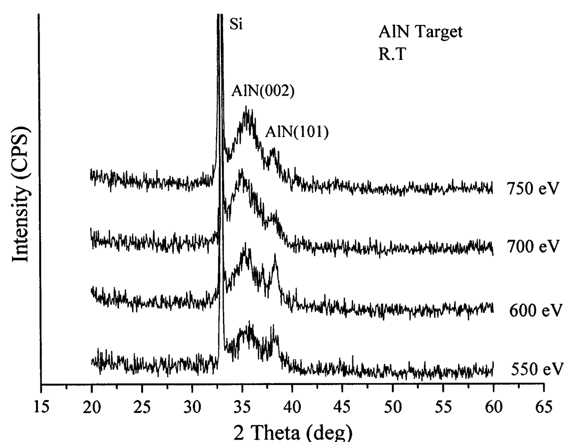

Fig. 5. XRD patterns of AlN films synthesized as a function of fluxes. An AlN target was used with unheated substrate.

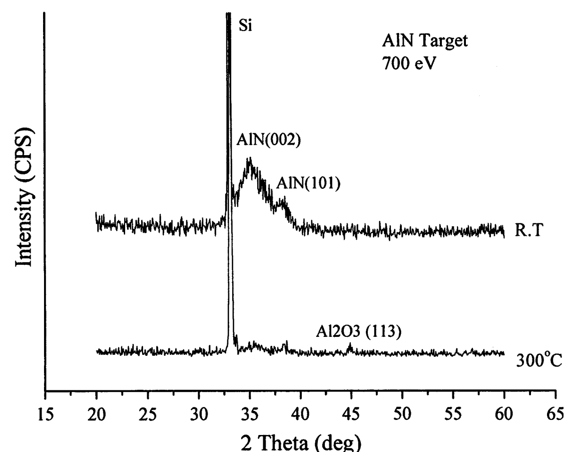

Fig. 6. XRD patterns of AlN films synthesized with and without substrate heating, An AlN target was used.

with added nitrogen gas are generally better (with a stronger (002) texture) than those prepared with an AlN target.

Figure 1 shows that the AlN film deposited at 700 eV beam energy flux using an Al target at 55% N_2 concentration without substrate heats, designated as specimen A, possessed the strongest (002) texture and is therefore the best AlN thin film. Figure 1 also reveals that the deposition rate at a high energy flux level of 750 eV can be so rapid that the atoms may have enough time to migrate to suitable positions, whereas at lower energy flux levels of 650 and 680 eV, there may not be enough kinetic energy for the atoms to move to suitable positions.[23]

Figure 2 shows the crystallinity of AlN films deposited at 700 eV energy flux as a function of nitrogen concentration. This figure reveals an AlN film with strong (002) texture when the N_2 concentration is 55%. However, the strong (002) texture is nearly completely lost, while the N_2 concentration is increased to 60% or decreased to 50%. These results demonstrate that the crystallinity and the strong texture of AlN film are very sensitive to the nitrogen concentration. Because argon is easier to ionize than nitrogen and the bombarding effect of argon ions is better than nitrogen, this sensitivity is due to a proper match between the bombarded aluminum atoms and bombarding nitrogen atoms. Based on the above findings, the AlN film deposited has the best result at 55% N_2 concentration.

Figures 3 and 4 show the effects of the substrate temperature and the N_2 concentration on the prepared AlN films. The results reveal that the (002) texture of AlN films deposited at high temperatures were inferior. This phenomenon is due to the fact that the increased kinetic energy from substrate heating may be excessive, and thus, the texture of the AlN film is decreased. Figures 5 and 6 show that the results are the same regardless of the substrate temperature effect when the AlN target is used.

The SEM micrographs of the surfaces of AlN films are shown in Fig. 7a and b. For these two specimens, both surfaces appear to be smooth and the only observable difference is the grain size. The surface roughnesses of several AlN films are shown in Table 1. These results show that, regardless of the substrate temperature or the targets (Al or AlN), the surfaces of the AlN films are always quite smooth, with nearly the same degree of roughness.

The microstructure of the specimens with the heated AlN film, prepared with 700 eV energy flux and a 55% N_2 concentration without substrate heating, was studied in detail by TEM. In the cross-sectional view in the bright-field image (BFI) in Fig. 8a and in the center dark-field image (DFI) in Fig. 8b, four distinct layers, labeled layers A–D, are identified. Layer A is an aluminum layer about 36-nm thick. The actual AlN film has a thickness of about 500 nm and consists of the three layers, B–D. The randomly oriented crystallites in layer B become aligned into

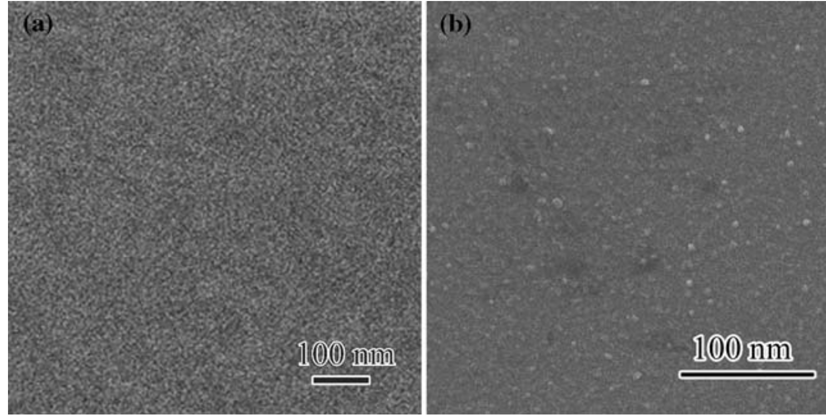

Fig. 7. (a) SEM micrograph of AlN films of specimen A prepared with an Al target at 55% N_2, 700 eV, and nonsubstrate heating. (b) SEM micrograph of AlN films prepared with an AlN target at 700 eV and substrate temperature of 300°C.

columnar structures in layer C, and the size of these column structures increases in layer D toward the film surface. The preferred orientation of the *c*-axis of the hexagonal structure is nearly perpendicular to the substrate, as determined by the XRD results (Fig. 1). Figures 8c and d are the selected area diffraction (SAD) patterns of layers B and D, respectively. The arclike rings in the SAD patterns suggest that the AlN is highly textured, especially in layer D.

Figure 9a shows the diffraction pattern of plane view in layer D of specimen A in Fig. 8. The arclike patterns indicate the strong texture development by confirming the epitaxic growth result shown in Fig. 8d. The rings of the diffraction pattern of the AlN are indexed in sequence as (100), (101), (110), (103), and (112), while the (002) ring is absent. Figures 9 and 8d strongly suggest that the film is highly textured, with the *c*-axis oriented perpendicular to the substrate surface. The diffraction pattern in layer B (Fig. 9b) shows that the pattern is of uniform rings and that the (002) ring is also present. This indicates that the crystals are randomly orientated. The particle size of AlN in layer B, estimated by the DFI in Fig. 9c, is about 6–7 nm.

Based on the plane view (Fig. 9) and cross-sectional view (Fig. 8) microstructures, in the first deposition stage, the AlN forms small crystallites (6–7 nm) with random orientation. This is probably due to the large lattice mismatch between the Al layer and AlN layer and the fact that the unheated substrate does not provide any more kinetic energy to facilitate the diffusion of atoms or clusters to assist in suitable growth and coalescence. With increased deposit time and film growth, the small particles coalesce along the growth axis aligned toward the preferred (002) orientation. However, no amorphous layer exists between the Al layer and the AlN layer in contrast to AlN films deposited by magnetron sputtering.[10]

Figure 10 shows the plane-view TEM SAD patterns of AlN films grown on heated substrates of 300°C, with a 700 eV energy flux, and 55% N_2 concentrations (specimen B). This observation is consistent with Fig. 6, because the indexes in the figure indicate that oxidization to the AlN, Al_2O_3

Table I. Measured Surface Roughnesses of Four Specimens: Specimens A and B, Prepared Using an Al Target at 700 eV Energy Flux, 55% N_2 Concentration, and Substrate Temperature at 25°C and 300°C; and Specimens C and D, Prepared Using an AlN Target at 650 eV Energy Flux, 55% N_2 Concentration, and Substrate Temperature at 25°C and 300°C.

		Roughness				
Specimen Number	**Measurement Items**	**1**	**2**	**3**	**4**	**5**
A5	R_a (nm)	10.8	10.8	11.3	12.1	10.6
	R_q (nm)	13.6	13.3	14.1	15.0	13.1
B	R_a (nm)	10.5	11.2	10.4	11.9	10.2
	R_q (nm)	13.1	13.9	12.9	14.8	12.6
C	R_a (nm)	11.4	11.7	11.5	11.9	11.0
	R_q (nm)	14.2	14.6	11.4	14.9	13.7
D	R_a (nm)	10.5	11.0	11.6	11.6	11.5
	R_q (nm)	13.0	13.9	14.4	14.5	14.2

Remarks:1 Test instrument: Form Talysurf (Taylor-Hobson).2. Data length: 1.040 mm; out off length: 0.08 mm.

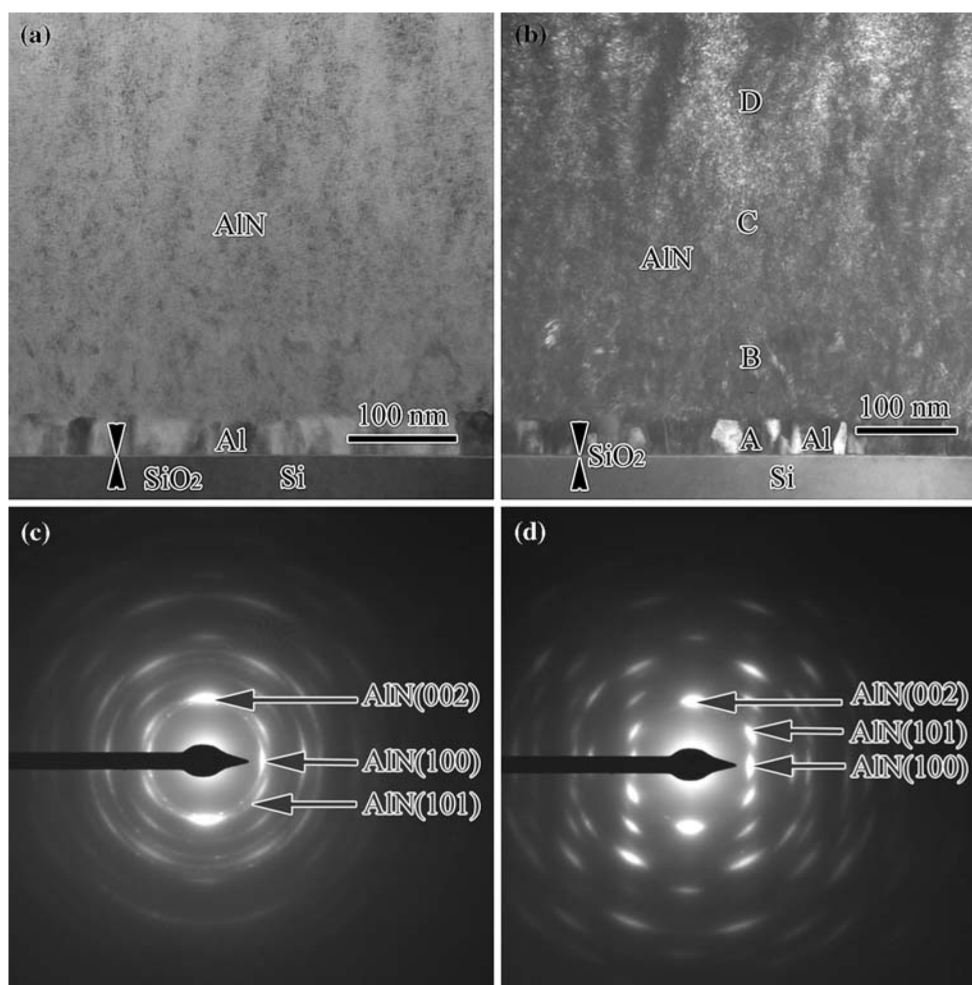

Fig. 8. Cross-sectional TEM micrograph of AlN films of specimen A synthesized at 55% N_2, 700 eV, and room temperature using an Al target showing the following: (a) BFI, (b) DFI, (c) diffraction pattern and index layer B, and (d) diffraction pattern and index of layer D.

particles is present. It is therefore clear that heating the substrate promotes the formation of Al_2O_3 grains, probably due to the release of O_2 previously absorbed by the chamber wall or from the cracking of residual water.

Figure 11 shows the SIMS depth profile of the AlN film prepared by an Al target at 700 eV, 55% N_2, and no substrate heated (specimen A). The results demonstrate that AlN films contain same amount of oxygen even without heating. The oxygen may also come from the oxygen previously absorbed by the chamber wall or from the cracking of residual H_2O. It is known that AlN can be oxidized into Al_2O_3 when heated in an oxygen-containing atmosphere, as has been shown in this experiment with heated substrate.

The ESCA method was used to study the differences between AlN films prepared with an Al target and an AlN target, with Fig. 12 showing the corresponding results. The Al-Al bonding in the AlN target can be clearly observed in Fig. 12c, in addition to the Al-N and Al-O bonding; this figure shows that the bonding occurs regardless of whether the substrates are heated, but cannot be observed in the Al target in Fig. 12a and b. This demonstrates that the AlN bond can be broken by ion source and will be removed by the vacuum system without extra nitrogen being supplied in this process, after which the aluminum atoms will then bond with each other. Because N_2 is supplied in the Al target of Fig. 11a, no Al-Al bond is present, and better AlN film can be prepared. It should be noted that this experiment demonstrates that the AlN target is unsuitable for use with the DIBS system.

CONCLUSIONS

Aluminum nitride (AlN) films of nanometer crystallites were deposited on Si (100) substrates using the DIBS deposition system. The highly textured, *c*-axis oriented AlN films can be synthesized by using Al target at 700 eV energy flux and 55% N_2 concentration, without substrate heating.

The main results can be summarized as follows.

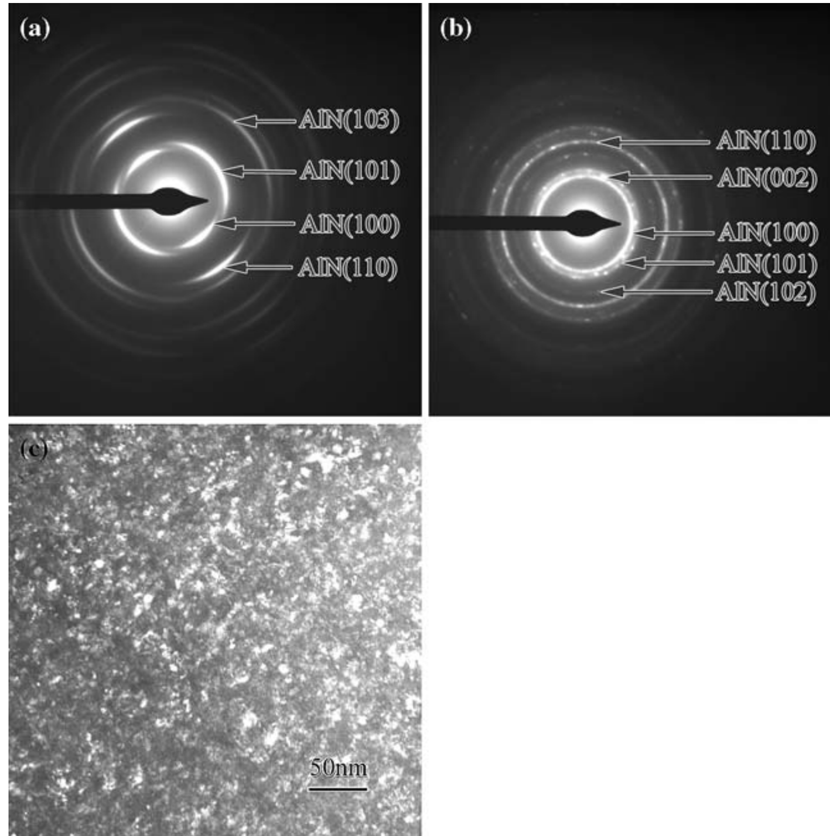

Fig. 9. Plan view TEM micrograph of AlN films of specimen A synthesized at 55% N_2, 700 eV, and RT using an Al target showing the following: (a) diffraction pattern and index of layer D, (b) diffraction pattern and index of layer B, and (c) DFI formed with part of the fist ring of (b).

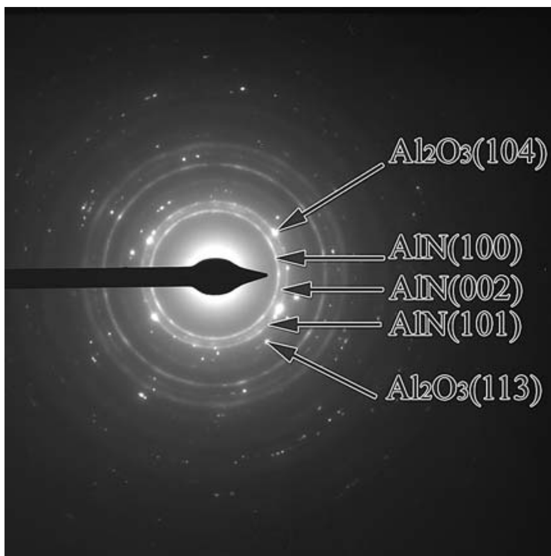

Fig. 10. TEM micrographs of AlN films synthesized at 700 eV and room temperature using an AlN target.

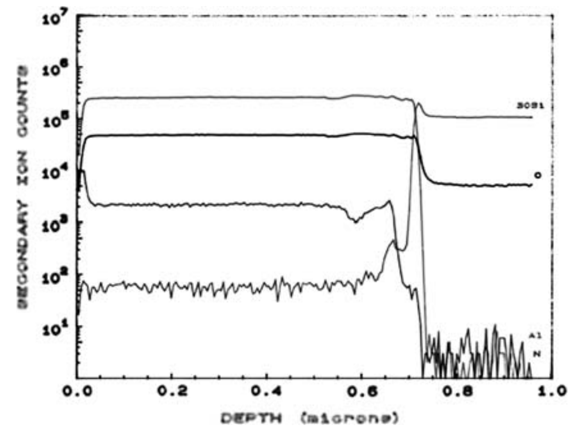

Fig. 11. SIMS depth profile of AlN films of specimen A formed with an Al target at 55% N_2 and 700 eV, with no substrate heating.

1. The AlN film prepared by the DIBS system using an aluminum target with added nitrogen gas is better in quality than that prepared using an AlN target directly. This is due to the fact that the AlN bonding can be broken by a high energy ion source.
2. Regardless of whether an aluminum target or AlN target is used, the quality of AlN films prepared at room temperature is better than those prepared with substrate heating under 300°C.

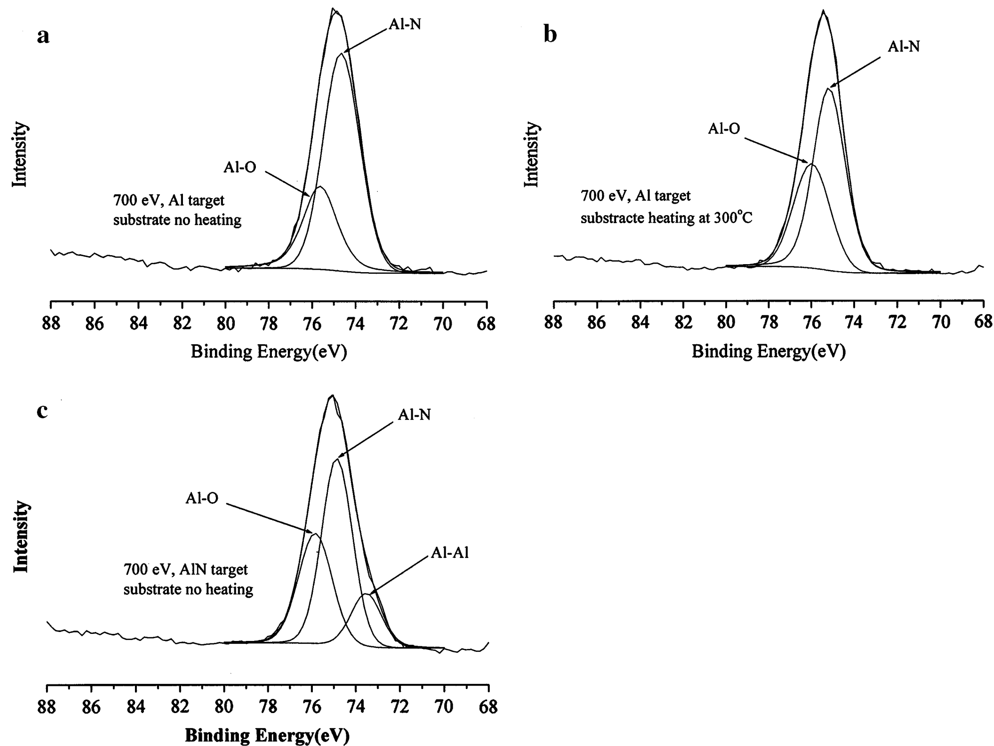

Fig. 12. ESCA results and the simulations of the bonding energy of AlN films formed by (a) an Al target (specimen A); (b) an Al target prepared at 700 eV, with substrate heating to 300°C; and (c) an AlN target prepared at 700 eV, with no substrate heating.

3. The surface roughness of the AlN film deposited at room temperature is better than that deposited at 300°C.
4. It was found that AlN can be divided into three layers: the bottom one, of about 100 nm, consisting of small crystallites with random orientation; the middle layer, where the crystallites start to align with a preferred (002) orientation; and the upper layer, characterized by a very strong (002) texture and increased grain size.

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