Optical and structural properties of Fe-doped AlN films prepared by reactive DC magnetron sputtering

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***Abstract*—Fe-doped AlN films were deposited on n-type Si (100) and quartz substrates by a reactive direct current magnetron sputtering system in an atmosphere of Ar and N_2 at room temperature. The target was a mixture of Al and Fe with a weight ratio 10:1. In order to study the effect of N_2 flow rate (F_{N2}) on the structure and optical properties of Fe-doped AlN film, F_{N2} changed from 0 to 50 sccm. The microstructure, surface morphology, and optical properties of the films were investigated by X-ray diffraction, atomic force microscopy, and spectrophotometer, respectively. Experimental results show that the films are amorphous when F_{N2} is less than 3 sccm. AlN (100) diffraction peak can be seen in all the samples as F_{N2} is larger than 6 sccm. Fe_3N (202) and AlN (103) peaks are found in the samples at F_{N2} of 20 and 30 sccm. AlN (110) peak appears at 40 sccm sample. Surface roughness and grain diameter of the samples on both Si and quartz substrates sharply decrease as F_{N2} increases from 0 to 10 sccm. When F_{N2} is larger than 10 sccm, the change in surface roughness and grain diameter is small. Optical transmittance of the film on quartz substrates increases with increasing F_{N2}. The maximum transmittance is higher than 90% when F_{N2} is higher than 20 sccm. When F_{N2} increases from 10 to 40 sccm, the optical energy gap increases from 5.483 to 5.601 eV.**

Keywords-component; Aluminium nitride; Fe-doped; Films; Sputtering; Nitrogen flow rate

I. INTRODUCTION

Aluminium nitride (AlN) has two crystal structures, a stable wurtzite structure with hexagonal symmetry and a metastable zinc-blends structure with cubic symmetry. AlN has been widely investigated due to its excellent properties, such as high electrical resistivity, high surface acoustic velocity, high thermal stability, high elastic modulus, a wide direct band gap of 6.2 eV, and good piezoelectric properties. AlN films, therefore, are used for surface passivation of thin films, insulating layers, optical sensors in the ultra violet spectral range, and surface acoustic wave devices [1-6]. AlN has also been used as an outer layer in solar thermal collectors [7, 8].

Many deposition methods have been used to prepare AlN films, such as chemical vapor deposition [9], reactive sputtering [3-5,10-14], filtered arc deposition [6], molecular beam epitaxy growth [15,16], and pulsed laser deposition [1,2,17]. Among these methods, reactive sputtering is the most common used method because the advantages of sputtering are stability, good adhesion of film on the substrate, reproducibility, and high-deposition rate.

In previous research, it has been found that the structure and property of AlN films strongly depend on N_2 flow rate (F_{N2}). For example, Venkataraj et al [5] had deposited AlN film in an Ar-N_2 atmosphere on Si (100) and glass substrates. Their results show that the stoichiometric AlN films can be obtained as F_{N2} is larger than 5 sccm. The influence of sputter deposition parameters on piezoelectric and mechanical properties of AlN films had been investigated by Ababneh et al [18]. It is found that the degree of c-axis orientation increases with increasing nitrogen concentration. Jejurikar et al [2] had studied the effect of substrate temperature and ambient nitrogen pressure on the structural and optical properties of pulsed laser deposited AlN film on sapphire substrate. They found the crystallographic orientations of the film depend on both the substrate and the ambient nitrogen pressure. However, few researches about the effect of F_{N2} on the structure and optical properties of Fe-doped AlN films had been reported [22].

In this paper, we focus on the effect of nitrogen flow rate on the structure, morphology, and optical property of Fe-doped AlN films. Fe-doped AlN films were deposited by reactive direct current (DC) magnetron sputtering on n-type Si (100) and quartz substrates with F_{N2} changing from 0 to 50 sccm.

II. EXPERIMENT

Fe-doped AlN films were deposited in a DC reactive magnetron sputter system. The target (60 mm diameter and 5 mm thick) was a mixture of Al and Fe (purity > 99.0%) with a weight ratio 10:1. The vacuum chamber was evacuated down to a pressure of 4.0×10^{-4} Pa before sputtering. Sputtering was done in an atmosphere of Ar and N_2 at room temperature with a target substrate distance of 50 mm and a pressure of 2.0 Pa. The substrates were n-type Si (100) and quartz.

A standard two step process was used to clean the n-type Si (100) substrates, firstly Si substrate was ultrasonically cleaned in an acetone and ethanol solution about 15 min for removing grease and stuff, then was washed by distilled water. Secondly, the Si substrate was etched in a 40% (mass fraction) hydrofluoric acid for 1 min immediately after first step to remove oxide impurities and produce a smooth surface,

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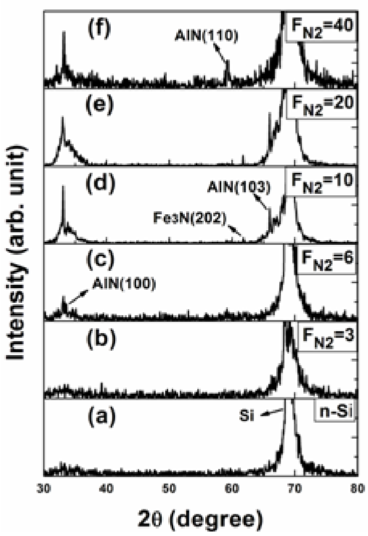

Figure 1. XRD patterns of etched Si substrate (a) and Fe-doped AlN films deposited on Si substrates at various F_{N2} (b)-(f).

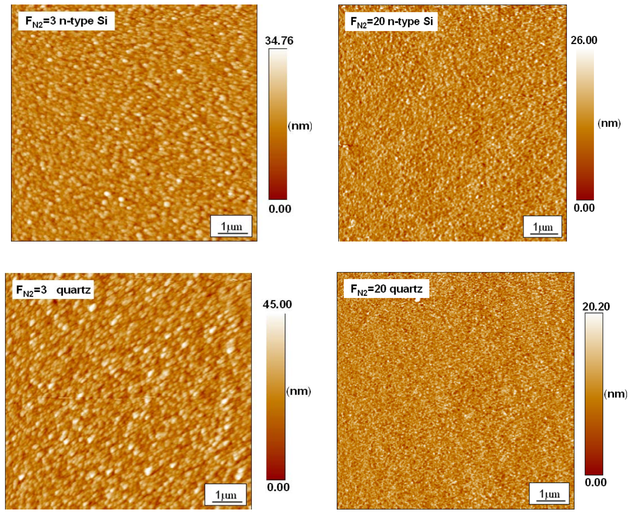

Figure 2. AFM images of Fe-doped AlN films deposited at different F_{N2} on Si and quartz substrates.

then was washed by distilled water, and went on natural withering.

Prior to depositing Fe-doped AlN films, the targets were presputtered for 5 min to remove their surface contaminator. Deposition power was 150 W and deposition time was typically 30 min for all films. Ar and N_2 flow rates were controlled by two mass flow meters respectively. Ar flow rate was maintained at 30 sccm. N_2 flow rate (F_{N2}) was changed from 0 to 50 sccm.

The thickness of the films was determined using a stylus profilometer (ZYGO Newview 5000). The structural properties of the films were investigated by X-ray diffraction (XRD) technique. XRD study was carried out on an X-ray diffractometer (Y-2000) with high intensity Cu Kα radiation (λ=1.54056 Å). The surface morphology and root mean square (RMS) surface roughness of films were investigated by a CSPM 4000 atomic force microscopy (AFM) in contact mode with 10 μm × 10 μm scanning areas. The normal incidence transmittance and near normal incidence reflectance (5 degree incidence angle) were measured by a double light beam spectrophotometer (UV-2450) in the wavelength range 200-900 nm.

III. Results and Discussions

A. X-ray analysis

Fig. 1 shows the XRD patterns of the n-type Si substrate and Fe-doped AlN films with different F_{N2}. The Fe-doped AlN films are amorphous when F_{N2} is lower than 3 sccm because there is no AlN diffraction peak in Fig. 1(b). When F_{N2} is 6 sccm, a weak AlN (100) peak can be observed, which indicates that c-axis AlN crystallite is parallel to the substrate [2]. As F_{N2} increases to 10 sccm, the AlN (100) peak becomes stronger, and two weak peaks appear, corresponding to the Fe_3N (202) and AlN (103) [19] peaks, respectively. The AlN (100) peak is weaker, and the peaks of both Fe_3N (202) and AlN (103) become stronger as F_{N2} increases to 20 sccm. When F_{N2} is 30 sccm, the peaks of both Fe_3N (202) and AlN (103) disappear, but a new AlN (110) peak appears (not shown in Fig. 1). The AlN (100) peak is weakest, but the peak of AlN (110) is stronger when F_{N2} is 40 sccm as shown in Fig. 1(f).

It has been reported that a longer distance between target and substrate, and a higher sputtering pressure are advantageous for the growth of AlN (100) films [11]. In this research, we believe that best condition for AlN (100) film on n-type Si (100) is sputtering at 2.0 Pa pressure and F_{N2} is 10 sccm. When F_{N2} increases from 10 to 40 sccm, the intensity of AlN (100) peak is weaker because the increase of F_{N2} will decrease kinetic energy of the deposition atoms on substrate [4].

B. Surface morphology

AFM images of Fe-doped AlN films deposited at F_{N2} with 3 and 20 sccm on Si and quartz substrates are shown in Fig. 2. The grain size and height of the grain on film surface are different with different F_{N2} and substrate. The RMS surface roughness and grain diameter (D) of the samples were calculated by AFM software from AFM image data. The calculated results are indicated in Fig. 3. Comparing Fig. 3(a) with Fig. 3(b), variation tendency of the RMS roughness and D for Fe-doped AlN films on Si and quartz substrates is similar. Both RMS roughness and D sharply decrease when F_{N2} increases from 0 to 10 sccm. The change of RMS roughness and D is relatively small as F_{N2} is larger than 10 sccm. RMS roughness of the film on Si substrate is smaller than that on quartz substrate because Si substrate has a smoother surface than quartz substrate. The film deposited at 0 sccm has the largest RMS and D, which is the result of the films are metal Al and Fe [5].

It has been reported that a higher N_2 pressure leads to a reduction in adatom mobility on substrate, which results in more distinct columnar structure and decrease of density of the film. Therefore, the variation of RMS roughness and D of Fe-doped AlN films is consistent with the structure zone model proposed by Thornton [12].

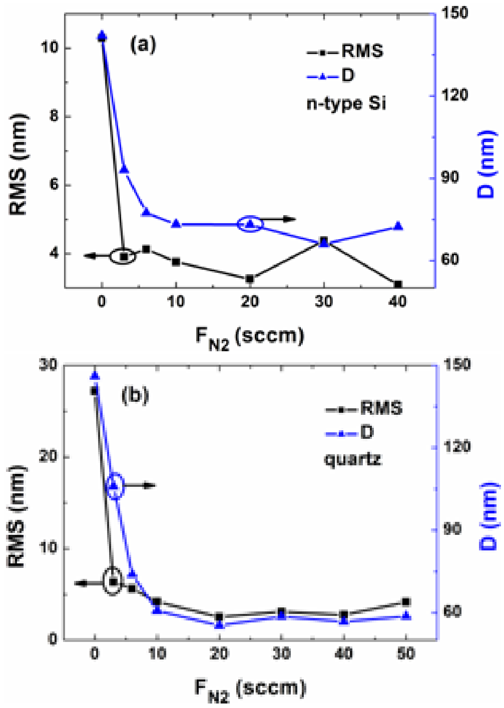

Figure 3. RMS surface roughness and grain diameter (D) of Fe-doped AlN films deposited at different F_{N2} on Si (a) and quartz (b) substrates.

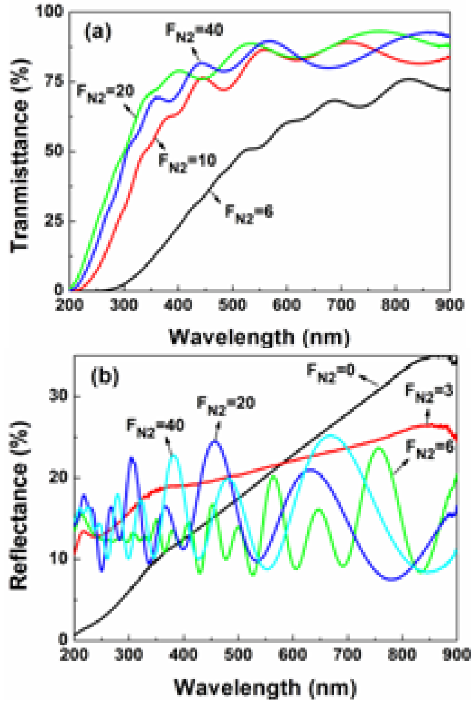

Figure 4. Transmittance (a) and reflectance (b) of Fe-doped AlN films on quartz substrates.

C. Optical properties

Transmittance and reflectance of the films deposited on quartz substrates are shown in Fig. 4. The transmittance is zero when $F_{N2} \leq 3$ sccm because the most of film is metal Al and Fe. As seen in Fig. 4(a), the transmittance increases with the increase of F_{N2}. The average transmittance (wavelength between 500 and 900 nm) is about 85% as F_{N2} is larger than 6 sccm. The maximum transmittance is higher than 90% when F_{N2} is larger than 20 sccm. There is not interference ring in reflectance when $F_{N2} \leq 3$ sccm. The interference rings can be seen obviously in reflectance as $F_{N2} \geq 6$ sccm because the films contain AlN which is confirmed by XRD results in Fig. 1.

The absorption coefficient (α) of a film can be determined by [20]

$$T = \frac{(1-R)^2 e^{-\alpha d}}{1-R^2 e^{-2\alpha d}}, \qquad (1)$$

where T and R are transmittance and reflectance, and d is film thickness. (1) can also be written as

$$\alpha = \frac{1}{d}\ln\left\{\frac{(1-R)^2}{2T} + \left\{\left[\frac{(1-R)^2}{2T}\right]^2 + R^2\right\}^{-\frac{1}{2}}\right\}. \qquad (2)$$

The energy band gap (E_g) can be derived from the Tuac's relationship between α and the photon energy ($h\nu$). For our films, the relationship is follow for a direct band gap material [21]

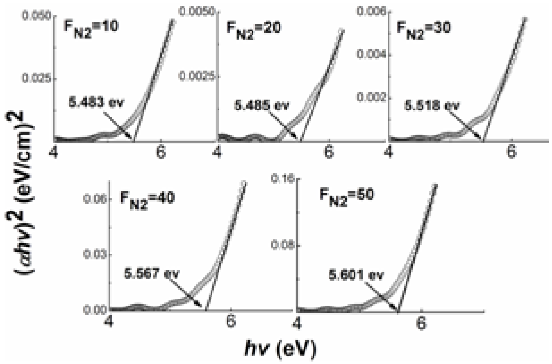

Figure 5. Plots of $(\alpha h\upsilon)^2$ versus $h\upsilon$ of the films on quartz substrates.

$$(\alpha h\nu)^2 = A(h\nu - E_g) \qquad (3)$$

where A is a constant. With the data in Fig. 4, α of Fe-doped AlN films can be calculated by (2). Then $(\alpha h\upsilon)^2$ versus $h\upsilon$ can be obtained and shown in Fig. 5. The extrapolation of the linear part of the curve to zero yields E_g. E_g increases from 5.483 to 5.601 eV when F_{N2} increases from 10 to 50 sccm, which may be attributed to the change in crystalline size [21],different stoichiometric Fe_XN_Y and Al_XN_Y, and the different of relative content between iron nitride and aluminum nitride.

IV. Conclusions

Fe-doped AlN films were deposited by DC reactive magnetron sputtering on n-type Si (100) and quartz substrates. N_2 flow rate changed from 0 to 50 sccm and Ar flow rate was 30 sccm during deposition. XRD results of the films on Si substrates show that the films are amorphous when F_{N2} is lower than 3 sccm. All the films have an AlN (100) peak when F_{N2} is larger than 6 sccm. Fe_3N (202) and AlN (103) peaks are found in the films deposited at 20 and 30 sccm F_{N2}. AlN (110) peak is found at 40 sccm sample. RMS surface roughness and grain diameter for the films on both Si and quartz substrates decrease sharply as F_{N2} increases from 0 to 10 sccm. The change in RMS and grain diameter is relatively small when F_{N2} is larger than 10 sccm. Transmittance of the film on quartz substrate increases with the increase of F_{N2}. The maximum transmittance is higher than 90% when F_{N2} is larger than 20 sccm. The average transmittance (wavelength between 500 and 900 nm) is about 85% and the interference rings in reflectance can be seen obviously as F_{N2} is larger than 10 sccm. The energy band gap increases from 5.483 to 5.601 eV when F_{N2} increases from 10 to 50 sccm.

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