C-axis tilted AlN films for vibration energy harvesters

H. H. Nguyen, M. Hara, H. Oguchi, H. Kuwano
Department of Nanomechanics, Tohoku University, Japan
E-mail: hung@nanosys.mech.tohoku.ac.jp

Abstract. In this study, we reported on the sputtering growth of the c-axis tilted AlN films, which may have higher electromechanical coupling coefficient $k_{31}$ than [0001]-oriented AlN, on the Si (100) substrates in the aim of high output power vibrational energy harvesters (VEHs). We implemented the incident angle deposition in a wide range of temperature from room temperature (RT) to 650°C. At 420°C the c-axis tilted 22°, which is the largest tilt angle reported for the AlN films grown by incident angle deposition. In addition, by changing growth temperature, we succeeded in preparing c-axis tilted AlN films with different tilt angle. Scanning electron spectroscopy (SEM) revealed that the c-axis tilted films have tilted columnar structures which prevents cracks and pinholes going straight through the films. This feature may help avoiding electrical short between electrodes fabricated on top and bottom of the AlN films in VEHs.

1. Introduction

In recent years much attention has been paid on an AlN thin film as an alternative piezoelectric material to Lead Zirconate Titanate (PZT) in vibrational energy harvesters due to more concern to the environment and human health that asks society to restrict the use of lead in industry. Accordingly researchers have searched for lead-free piezoelectric materials for vibrational energy harvesters (VEHs) [1-3], among lead-free piezoelectric materials developed so far, an AlN (wurtzite structure) film has been considered as one of the most promising materials for vibration energy harvesters because of its high figure of merit (FOM) of output power density [4, 5]. Furthermore AlN, a semiconductor process compatible materials, can be easily integrated into silicon devices [4, 6].

In general people use [0001]-oriented AlN films which have c-axis oriented normal to the substrate surface because electromechanical coupling coefficient ($k_{31}$) is large along this crystal orientation [10] and output power of the VEHs can be increased. However, $k_{31}$ of the [0001]-oriented AlN is still smaller than that of PZT films [4, 6-9]. Thus it is important to further increase $k_{31}$ of the AlN thin films, and tilting c-axis of the [0001]-oriented can be one effective approach [11, 12].

There have been several reports of c-axis-tilted AlN thin films aiming for optoelectronics applications. For example, Rodriguez-Navarro et al. have deposited AlN on a glass substrate using RF reactive sputtering by incident angle deposition where they arranged a substrate and a target so as to set incident angle of the deposition flux to be 45°, and obtained films with ~15° tilted c-axis [13]. In addition, Dellas and Harper obtained films with 8° tilted c-axis [14] using the similar method. However, to further increase $k_{31}$ to catch up with PZT, more tilt angle is desired. Moreover no one investigated $k_{31}$ of the c-axis-tilted AlN thin films.

In this study we aim to grow AlN films with the c-axis tilt angle of more than 15° by RF sputtering. In addition we prepare AlN films with different c-axis tilt angle for the future investigation of $k_{31}$ of the c-axis tilted AlN films and fabrication of high output power VEHs.
2. Experiment

Figure 1 shows a schematic of the sputtering set up for incident angle deposition of the AlN films. A substrate holder and an Al target were arranged so as to set incident angle of deposition flux to be 45° from substrate normal. During film growth Ar and N$_2$ are used as reactive gases. N$_2$ was introduced into the chamber through the ion gun to make nitrogen plasma to facilitate AlN formation [15]. N$_2$/Ar gas flow rate of 3 was fixed to obtain AlN stoichiometry according to the previous investigation shown in Figure 2. Other sputtering parameters were summarized in Table 1. Prior to each AlN film deposition, silicon substrates were cleaned by standard cleaning processes of Radio Corporation of America (RCA1 and RCA2). The Al target was pre-sputtered for 10 minutes in Ar and for 5 minutes in the mixture of Ar and N$_2$. Thicknesses of the films were measured by a surface profiler. Field emission scanning electron microscopy (FE-SEM, Hitachi SU-70) was used to perform morphological analysis. The composition was confirmed by using the EDX spectroscopy. An atomic force microscopy (AFM, Nikon TE2000-U) was used to investigate the surface roughness. X-ray diffractometer equipped with a two dimensional detector (2D-XRD) was employed to study crystal quality and c-axis tile-angle. Tilt angles were calculated from 2D-XRD diffraction images using software named General Area Detector Diffraction System (GADDS).

![Figure 1. Schematic of deposition](image1)

Table 1. Deposition conditions

| Parameter             | Value               |
|-----------------------|---------------------|
| Substrate temperature | RT – 650°C          |
| Back pressure         | 1.5x10$^{-7}$ Torr  |
| Depositing pressure   | 1.2x10$^{-3}$ Torr  |
| RF power              | 140 W               |
| Deposition time       | 1 h                 |
| Film thickness        | 600 nm              |
| Ar gas flow rate      | 2.5 sccm            |
| N$_2$ gas flow rate   | 7.5 sccm            |
| Accelerated voltage   | 1200 V              |
| Beam current          | 43 mA               |

![Figure 2. Dependence of Al and N ratio in the thin film deposited at RT as a function of Ar and N$_2$ flow rate.](image2)

3. Results and discussions

XRD measurements verified that we have succeeded in growing c-axis tilted AlN thin films. Figure 3a shows a 2D-XRD diffraction image of the AlN film grown at 650°C. Except a diffraction peak from Si, only peaks from (0002) and (0004) plane of AlN were observed almost symmetrically along the chi axis, indicating that film has [0001] orientation. With decreasing growth temperature, the (0002) peak gradually moves down along the chi axis. This clearly indicates that the c-axis tilted from the substrate normal. Tilt-angles were determined as Chi angle of the (0002) peak centre (figure 3), and found to be 6° for 560°C (Figure 3b) and 22° for 420°C (Figure 3c). For the films grown at lower than 300°C (figure 3d and 3e) the tilt angle could not be determined due to poor crystallinity, though the c-axis likely tilt more from the substrate normal at lower growth temperature.
To understand the mechanism determining the tilt-angle of the c-axis, we conducted FE-SEM. A cross-sectional FE-SEM image of the film grown at 420°C (tilt-angle: 22°) shows a columnar structure with tilt-angle of 43° (Figure 4b), which is close to the incident angle of the deposition flux. In contrast, the columnar structure for 650°C (tilt-angle: ~ 0°) does not tilt (Figure 4a). These facts indicate that tilting of the c-axis can be obtained by growing crystal along the tilted direction using the incident angle deposition if growth temperature is low enough to suppress atomic migration.

SEM and AFM measurements for the film surface further confirmed our hypothesis. Figure 5 shows SEM micrographs of the thin films grown at different temperatures. The size of the islands decreases with increasing growth temperature from ~50 nm for 300°C to ~10 nm for 620°C. The root mean square roughness \( R_{\text{RMS}} \) of the films deposited at 300°C, 420°C and 650°C determined by AFM measurements are 3.89 nm, 1.15 nm and 1.09 nm, respectively. These observation consistently indicates that more atomic rearrangement occurs at higher growth temperature.

Finally we point out that incident angle deposition can be an appropriate growth method to suppress electrical short between top and bottom electrodes that is often a problem reported for AlN films in micro devices [15, 18, 19]. Because the tilted columnar structures are stacked, films do not allow cracks and pin-holes to go straight through the film. This unique geometry will prevent connection between top and bottom electrodes, which are usually fabricated using physical vapour deposition methods with line-of-sight deposition.
4. Conclusion

In summary, we have succeeded in growing the AlN films with most tilted c-axis ever among the films grown by incident angle deposition. The results of this study revealed that one key parameter controlling the tilt angle of the c-axis is the growth temperature. For the incident angle deposition with incident flux angle of 45°, the tilt angle was successfully changed from 0° for 420°C to 22° for 650°C. This study also indicated that incident angle deposition may be an appropriate method to avoid electrical short through the AlN films by avoiding formation of cracks and pin-holes straight through the films.

References
[1] Le VM, Hara M, Kuwano H. 2013. J. J. Appl. Phys. 52 07HD8
[2] Springer Science 2008 Piezoelectric MEMS, ed Bassiri-Gharb N, chapter 20: Materials and Devices, pp. 413
[3] Hara M. and Kuwano H. 2012 Jpn. J. Appl. Phys. 51 07GC11

[4] Matloub R., Hadad M., Mazzalai A., Chidambaram N., Moulard G., Sandu C. S., Metzger Th., and Muralt P. 2013, Appl. Phys. Let. 102 (15) 152903

[5] Sakurai T, Yamada O, Miyamoto Y. 2006. Mat. Sc. Eng.: A 415 40

[6] Nathan J, Lynette K, Alan M. 2013. Smart Mater. Struc. 22 115033

[7] Tasnádi F, Alling B, Höglund C, Wingqvist G, Birch J, et al. 2010. Phys. Rev. Let. 104 137601

[8] Akira N, Mari A, Tsuyoshi Y, Satoru M, Masanori U, et al. 2015. J. J. Appl. Phys. 54 07HD1

[9] Zhang J, Cao Z, and Kuwano H 2011 Jpn. J. Appl. Phys. 50 09ND18.

[10] Trolier-McKinstry S, Muralt P. 2004. J. Electr. 12 7

[11] Wang Q, Cao Z. and Kuwano H. 2012 Proc. PowerMEMS2012 327

[12] Wang Q, Oguchi H, Hara M, Kuwano H. Micro Electro Mechanical Systems (MEMS), 2014 IEEE 27th International Conference, 636

[13] Rodriguez-Navarro A, Otaño-Rivera W, García-Ruiz JM, Messier R, Pilione L.J. 1997. J. Mat. Res. 12 1689

[14] Dellas NS, Harper JME. 2006. Thin Sol. Fil. 515 1647

[15] Nguyen HH, Oguchi H, Kuwano H. 2014, Journal of Physics: Conference Series 557:012047

[16] Deng R, Muralt P, Gall D. 2012. J. Vac. Sc. Tech. A 30 051501

[17] Jochen MS, Suzanne R, William DS, Allan M. 2000. Journal of Physics D: Applied Physics 33 R173

[18] Kar JP, Bose G, Tuli S. 2005. Mat. Sc. Sem. Proc. 8 646

[19] Kumada T, Ohtsuka M, Fukuyama H. 2015. AIP Advances 5 017136