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Impact of cooling condition on the crystal structure and surface quality of preferred *c*-axis-oriented AlN films for SAW devices*

ZHANG Geng-yu(张庚宇), **YANG Bao-he**(杨保和)**, **ZHAO Jian**(赵健), **LI Cui-ping**(李翠平), and **LI Ming-ji**(李明吉)
Tianjin Key Laboratory of Film Electronic and Communication Devices, Tianjin University of Technology, Tianjin 300384, China

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AlN films with preferred *c*-axis orientation are deposited on Si substrates using the radio frequency (RF) magnetron sputtering method. The post-processing is carried out under the cooling conditions including high vacuum, low vacuum under deposition gas ambient and low vacuum under dynamic N_2 ambient. Structures and morphologies of the films are analyzed by X-ray diffraction (XRD) and atomic force microscopy (AFM). The hardness and Young's modulus are investigated by the nanoindenter. The experimental results indicate that the (100) and (110) peak intensities decrease in the XRD spectra and the root-mean-square of roughness (R_{rms}) of the film decreases gradually with the increase of the cooling rate. The maximum values of the hardness and Young's modulus are obtained by cooling in low vacuum under deposition gas ambient. The reason for orientation variation of the films is explained from the perspective of the Al-N bond formation.

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Aluminium nitride (AlN) is an attractive piezoelectric material applied in surface acoustic wave (SAW) devices due to its high dielectric constant (8.7 F/m), wide band gap (6.2 eV) and outstanding coefficient of thermal conductivity[1-3]. In particular, AlN film with preferred *c*-axis orientation exhibits many advantages: high phase velocity, excellent piezoelectricity and outstanding temperature stability[4,5]. Up to now, some techniques have been proposed to deposit AlN on silicon (Si) substrate, such as RF magnetron sputtering, metalorganic compound chemical vapour deposition (MOCVD)[6], pulsed laser deposition (PLD)[7,8] and molecular beam epitaxy (MBE)[9]. However, the effect of cooling condition on the orientation of the film has been rarely discussed. The cooling rate plays an important role in the orientation variation of the films. To release the internal stress and maintain the preferred orientation, it is necessary to study how the cooling condition affects the preferred orientation of AlN films deeply and comprehensively.

In this work, AlN films are deposited by the RF magnetron sputtering technique and cooled down under the following conditions: high vacuum, low vacuum under deposition gas ambient and low vacuum under dynamic N_2 ambient.

AlN films are deposited using a RF magnetron sputtering system (JGP500DI RF *f*=13.56 MHz), which is evacuated by a turbo molecular pump. The apparatus consists of an injecting room and a deposition chamber. Firstly, three Si (400) slices with the same size (1 cm×1 cm) and the same thickness (380 ± 20 μm) are cleaned following a standard cleaning procedure (30 min in boiling H_2SO_4: H_2O_2 (1:1), 10 s etching in HF: H_2O (1:10) and then washing in demonized water for 30 min). Then the pre-sputtering is performed in an Ar discharge at the power of 50 W for 30 min. The Si slice is introduced into the deposition chamber. Deposition is carried out in N_2 (99.999 %) /Ar (99.999 %) gas mixtures for 1 h. The deposition parameters are listed in Tab.1. After each deposition execution, the growth temperature should decrease to room temperature (25 °C) to eliminate the residual stress in the films. Due to self-cooling, only one cooling rate can be obtained with each cooling condition. The cooling rates are 0.9°C/min, 1.2°C/min and 1.8°C/min corresponding to the specimens A, B and C, respectively. The cooling conditions are shown in Tab.2.

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** E-mail: bhyang207@163.com

Tab.1 Deposition parameters of AlN films by RF magnetron sputtering

Parameter	Value
RF power	350 W
Substrate to target distance	~ 5 cm
Substrate temperature	300 °C
Sputtering gas pressure	1.0 Pa
Component ratio between N_2 and Ar	1:1
Gas flow rate	10 sccm
Base pressure	8×10^{-5} Pa
Al target (pure Al 99.999%)	6-cm diameter; 3-mm thickness
Growth rate	1.4 μm/h

The characterization techniques for AlN films are given as follows:

1. The crystallographic properties of the films are measured by grazing incidence X-ray diffraction (Japanese D/MAX-2500) operating at 40 kV and 150 mA (Cu Kα wavelength at 0.154056 nm, with an incidence angle of 3°).

2. The surface roughness is measured by AFM (Agilent 5500) in contact mode. The surface range of the image used in AFM image is 1 μm×1 μm.

3. The hardness and Young's modulus are analyzed by the nanoindenter (Nano indenter XP system of MTS Cooperation, USA).

Tab.2 Cooling parameters for AlN films cooled down at different gas pressures, cooling rates and N_2 flow rates

Cooling parameter	A (High vacuum)	B (Low vacuum under deposition gas ambient)	C (Low vacuum under dynamic N_2 ambient)
Gas pressure (Pa)	4.0×10^{-4}	6.5	7
N_2 flow rate (sccm)	–	–	15
Cooling rate (°C/min)	~0.9	~1.2	~1.8

Fig.1 shows the XRD spectra of the films under different cooling conditions. The effect of the substrate Si (400) peak can be eliminated by grazing incidence X-ray diffraction.

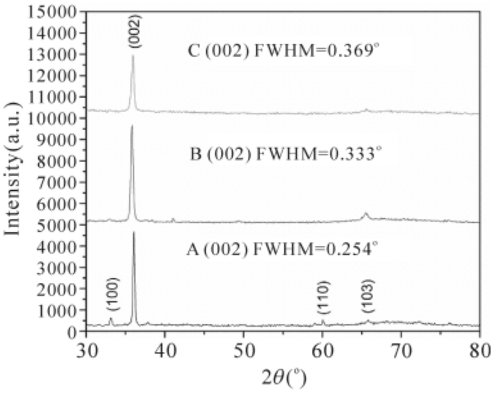
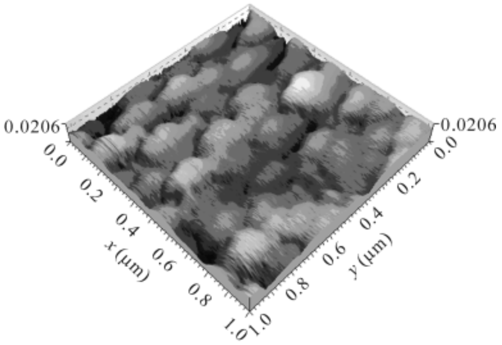


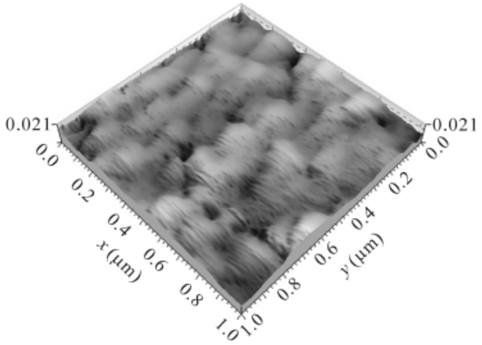
Fig.1 Typical XRD spectra of AlN films under different cooling conditions

The (002) and (103) peaks are observed in three specimens. From A to C, the (002) peak intensity decreases and the (100) peak disappears gradually. (110) and (103) peaks can also be observed in specimen A, which indicates that the cooling effect in high vacuum is the worst case among the three conditions. Moreover, the (002) peak intensity of the specimen C is the lowest while that of the specimen A is the highest and the FWHM is the narrowest in three specimens. Additionally, the (002) peak shift in A and B could be explained by the residual stress of the films.

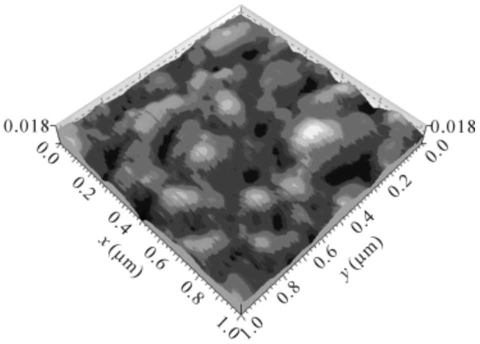
Three-dimensional (3D) AFM images (1 μm×1 μm) of AlN surfaces under different cooling conditions are given by Fig.2. The highest peaks in the *z*-axis direction are 20.5 nm, 21.0 nm and 18.0 nm respectively corresponding to three specimens.



(a) Typical AFM image of specimen A with R_{rms}=2.83 nm



(b) Typical AFM image of specimen B with R_{rms}=2.59 nm



(c) Typical AFM image of specimen C with R_{rms}=2.46 nm

Fig.2 Typical AFM images (1 μm×1 μm) of the film surfaces under different cooling conditions

Generally, the surface roughness of the film is required to be less than 30 nm for SAW devices. Therefore, three specimens follow the requirement of SAW devices. The R_{rms} values estimated from AFM measurement listed in Tab.2 are 2.83 nm, 2.59 nm and 2.46 nm respectively (Tab.3). The surface of the specimen C is the smoothest. Rough surface can induce scattering and attenuation of waves, so the penetration depth could be affected by the surface roughness severely. Moreover, the penetration depth of SAW in the layered structure decreases with an increase of the central frequency. Therefore, the specimen C is the best one among the three samples to minimize the propagation loss of SAWs, particularly at GHz frequency range.

The hardness and Young's modulus of the films are investigated based on the continuous stiffness measurement (CSM) technique using the nanoindenter[10]. The technique allows the continuous measurement of the contact stiffness during loading. The hardness and Young's modulus can be obtained from a single indentation process as a continuous function of the displacement into surface. In this work, the maximum loading value is set to be 200 mN. The target frequency is 45 Hz and the target strain rate is 0.05 s^{-1}.

Figs.3 and 4 show the hardness and Young's modulus varying with the displacement into surface of the films under different cooling conditions. The displacement into surface is obtained in the range of 0–21 % film thickness (0 –300 nm).

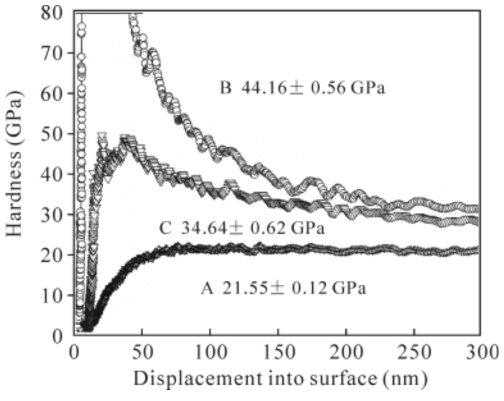


Fig.3 Hardness versus the displacement into surface of AlN films under different cooling conditions

Practically, the credible values of the hardness and Young's modulus usually appear at 10% –15% film thickness[11]. The hardness and Young's modulus of the films are obtained by averaging measurement data. This approach aims at reducing the effects of surface contamination and substrate. The hardness and Young's modulus of the specimen B are the highest as shown in Figs.3 and 4, and the hardness and Young's modulus values are listed in Tab.3. Accordingly, after the cooling process in low vacuum under deposition gas ambient, the AlN film with preferred *c*-axis orientation demonstrates a series of better mechanical properties and shows the substantial potential for the improvement of SAW phase velocity.

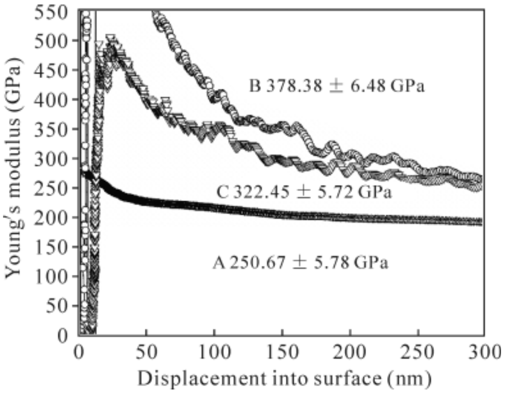


Fig.4 Young's modulus versus the displacement into surface of AlN films under different cooling conditions

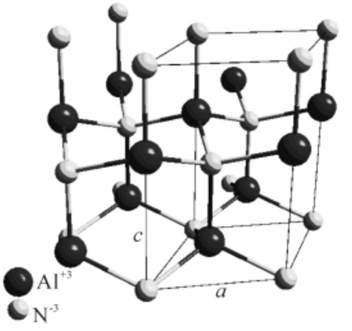
Tab.3 Characteristic parameters for AlN films cooled down under different conditions

Parameter	A	B	C
(002)FWHM(°)	0.254	0.333	0.369
R_{rms} (1μm×1μm) (nm)	2.830	2.590	2.460
Hardness (GPa)	21.55±0.12	44.16±0.56	34.64±0.62
Young's modulus (Gpa)	250.67±5.78	378.38±6.48	322.45±5.72

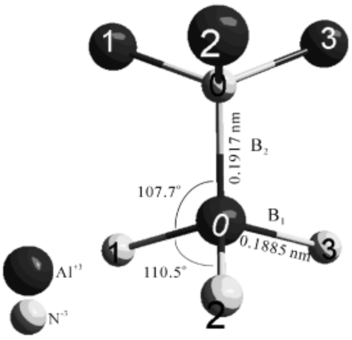
When preparing AlN films using the RF magnetron sputtering method, the grains in a crystal film can grow in a directional or non-directional way. According to the XRD spectra, some differences among these specimens can be found in Fig.1.

AlN has a hexagonal wurtzite structure with a 6-mm point group and P63mc space group. The crystal lattice constants are $a = 0.3110$ nm and $c = 0.4980$ nm (Fig.5(a)). The crystal lattice structure of AlN is shown in Fig.5(b). In the crystal cell, the Al and N atoms form four sp3-hybridized orbits. The Al atom has one empty orbit and three semi-full orbits, while the N atom has one full orbit and three semi-full orbits. Each Al atom is surrounded by four N atoms, forming a distorted tetrahedron. Two tetrahedrons make up a triangular prism. The tetrahedron is composed of three $A1\text{-}N_i$ $(i = 1, 2, 3)$ bonds named as B_1 and one Al - N_0 bond named as B_2. The bond angles of N_0- Al -N_1 and N_1- Al - N_2 are 107.7° and 110.5°, respectively. The bond lengths of B_1 and B_2 are 0.1885 nm and 0.1917 nm, respectively.

The bond B_1 is formed by the coupling between the Al semi-full orbit and the N semi-full orbit. The bond B_2 is formed by the coupling between the Al empty orbit and the N full orbit. Therefore, the ionic characteristic of the bond B_2 is greater. The bond energy for B_2 is smaller than that for other equivalent bonds B_1, which leads to the result that the energy required for the formation of the bond B_2 is greater. Planes (100) and (110) have only bond B_1 while planes (002) and (103) have bonds B_1 and B_2 together.



(a) Crystal lattice constants of AlN



(b) Bond angles and lengths of N-Al-N bond

Fig.5 Crystal lattice structure of AlN

It is concluded that planes (002) and (103) have already been formed during deposition in three specimens. The variation of the planes (100) and (110) in specimens can be explained in terms of the different cooling rates. With the cooling rate decreasing, the specimen can maintain a longer time of temperature decreasing. Desorptions of the Al and N atoms forming bond B_2 might appear due to thermal fluctuations and internal stress. The atoms of Al and N diffuse to some dangling bonds or the bonds needing lower bond energy, forming the bond B_1. As the planes (100) and (110) have only bond B_1 while the planes (002) and (103) have bonds B_1 and B_2, it is advantageous to form the planes (100), (110), (002) and (103). Therefore, from the specimens A to C, the (100) and (110) peak intensities in the XRD spectra decrease gradually even disappear in the specimen C completely.

The preferred *c*-axis-oriented AlN films are prepared by RF magnetron sputtering on Si substrates. The (100) and (110) peak intensities in the XRD spectra are affected by the cooling rate of the specimen. The hardness and Young's modulus of the film are the highest by cooling in low vacuum under deposition gas ambient. Three AlN films conform to the requirement of SAW devices, particularly at GHz frequency range. The reasons for the variation of the intensity and FWHM of the planes (002) and (103) will be investigated in future work.

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