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ORIGINAL PAPER

Growth of highly *c*-axis oriented aluminum nitride thin films on *β*-tantalum bottom electrodes

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Abstract We have investigated the influence of tantalum (Ta) bottom electrodes on the crystallinity and crystal orientation of aluminum nitride (AlN) thin films. AlN thin films and Ta electrodes were prepared by using rf magnetron sputtering method. The crystal structure of the Ta electrodes was tetragonal (*β*-Ta, a metastable phase) at room temperature. The crystallinity and orientation of the AlN thin films and Ta electrodes strongly depended on sputtering conditions. Especially, the crystallinity and crystal orientation of the Ta electrodes were influenced by their film thickness and the substrate temperature. When the thickness of the Ta bottom electrodes was 200 nm and the substrate temperature was 100 °C, the AlN thin films indicated high *c*-axis orientation (the full width at half maximum of rocking curve of 3.9°). The crystal orientation of the AlN film was comparable to that of AlN thin films deposited on face centered cubic (fcc) lattice structure metal, such as Au, Pt and Al, bottom electrodes.

Introduction

Quickly developed mobile telecommunication needs film bulk acoustic resonators (FBAR) for higher carrier frequencies [1]. The schematic view of a FBAR is shown in Fig. 1 [2]. The resonance frequency of a FBAR is mainly determined by the thickness of a piezoelectric layer and electrodes. The piezoelectric layer of 2.5 down to 0.5 μm would cover the very high frequency range from 2 GHz to 10 GHz. Zinc oxide (ZnO) was taken as the piezoelectric material due to its high effective coupling coefficient of 0.30 in the early 1980s [1]. However, ZnO has some drawbacks, such as low electrical resistance, low breakdown voltage, and high dielectric losses, because ZnO is usually an n-type semiconductor.

Aluminum nitride (AlN) would be an excellent piezoelectric material for FBARs. With an achievable effective coupling coefficient of 0.25 [3], filter with a sufficiently large bandwidth could be produced for mobile telecommunication. Since the band gap of AlN is large (Eg = 6.2 eV) [4], a high electrical resistance (10^{11}–10^{13} Ω cm) [5], a high breakdown voltage (640 MV/m) [6] and low dielectric loss can be expected. AlN could achieve small thermal drifts, because the thermal expansion coefficient of AlN is low (*c*-axis parallel: 5.3×10^{-6}/K, *a*-axis parallel: 4.2×10^{-6}/K) [7]. Furthermore, AlN films can be expected to be competitive in sensor [8–10] and ultrasound applications [11] where low loss, low thermal drift and high signal-to-noise ratios are demanded.

For thin film resonators, a polycrystalline *c*-axis oriented piezoelectric film is sufficient. However, it has to be grown on a metal electrode [1, 12]. We have investigated the influence of various bottom metal electrodes on the crystallinity and crystal orientation of AlN films. We reported that AlN films deposited on metal electrodes with the face centered cubic (fcc) lattice structure, such as Au, Pt and Al, showed high *c*-axis orientation [13]. The high orientation would be due to the fact that the crystallinity of the bottom electrodes was high, the surface roughness was low, and

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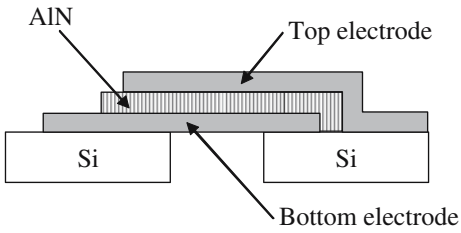

Fig. 1 Schematic view of film bulk acoustic resonator (FBAR) using AlN thin film

Au, Pt and Al (111) planes match well with hexagonal AlN crystal structure. However, we find that AlN deposited on tetragonal tantalum (β-Ta, a metastable phase) bottom electrodes indicated high crystal orientation, although the crystal structure of β-Ta is tetragonal. Furthermore, there is no report on the relationship between the crystal orientation of AlN films and β-Ta bottom electrodes to our knowledge. In this paper, we show the influence of β-Ta bottom electrodes on the crystallinity and crystal orientation of AlN films, and research into the preparation conditions.

Experimental procedure

AlN thin films were prepared by the rf planar magnetron sputtering system. The reactive sputtering conditions of *c*-axis oriented AlN films were as follows [14]. A target was 76.2 mm in diameter, 99.999% pure aluminum. The thickness of AlN films was 1 μm. The substrates were (100) silicon wafers with oxide layer (thickness:1 μm). The detailed sputtering conditions are shown in Table 1. The pressure in the vacuum chamber was maintained below 4×10^{-4} Pa before sputtering deposition, and then high-purity argon (99.999% purity) and nitrogen (99.999% purity) gases were introduced. Before the deposition process, the target was cleaned under the deposition conditions for 5 min. Ta bottom electrodes were prepared by dc sputtering system using only argon gas (99.999% purity) at room temperature. The crystal structure and crystallinity of AlN films and Ta electrodes were confirmed by X-ray diffraction (XRD) using CuK_α radiation. Their crystal orientations were evaluated by the full width at half-maximum (FWHM) of the X-ray rocking curves of the (0002) peaks of AlN films and (002) peaks of β-Ta bottom electrodes, respectively. The cross-sectional structures of AlN films were observed by transmission electron microscopy (TEM).

Table 1 Sputtering conditions of AlN thin films

Sputtering conditions of AIN films	
RF power	325 W
Sputtering pressure	0.5 Pa
Atmosphere	Ar:N_2 = 1:1
Sputtering time	4 h
Sputtering rate	5.2 nm/min
rf frequency	13.56 MHz
Target substrate spacing	>8.3 cm

Results and discussion

We prepared an AlN thin film on a Ta bottom electrode deposited on (100) Si substrate under the optimized sputtering conditions shown in Table 1 [14]. The AlN film was smooth and transparent. The XRD pattern and X-ray rocking curve of the AlN film are shown in Fig. 2. The XRD pattern indicates only two peaks of 33.68° and 36.04°, except for the silicon substrate peaks. The peak at 33.68° is especially high and sharp, and these two peaks are consistent with the tetragonal (002) β-Ta (a metastable phase) [15] and hexagonal (0002) AlN peak positions, respectively. There is no change on the peak positions, and the uniform stress in the film and electrode is not observed. This result means that the crystallinity of the β-Ta electrode is high, and the Ta electrode and AlN film both orient with the *c*-axis perpendicular to the substrate surface. The FWHM of the rocking curve (rotation of sample Θ-axis) of the (0002) AlN peak was 3.9°. It was reported that the FWHMs of AlN films prepared on Au, Pt and Al were 3.4°, 3.5° and 4.1°, respectively [13]. Thus the crystal orientation of the AlN thin film deposited on the β-Ta bottom electrode is comparable to that of AlN thin films deposited on fcc metal bottom electrodes.

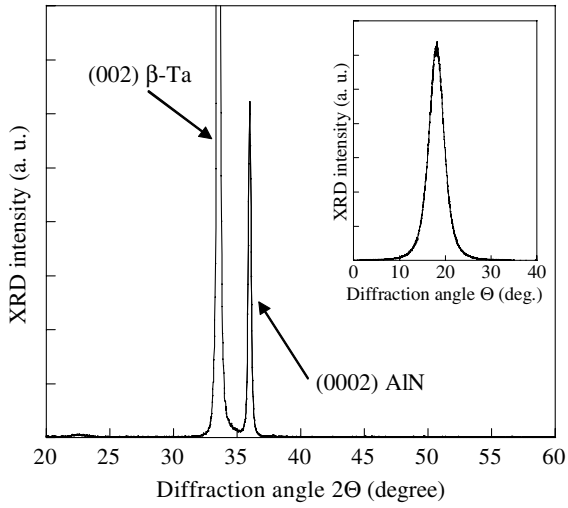

Fig. 2 XRD pattern of AlN film deposited on Ta electrode. Inset shows X-ray rocking curve of (0002) AlN peak. The substrate temperature was 100 °C, and the thickness of the Ta electrode was 200 nm

To further characterize the AlN film structure, we calculated the crystallite size of the film from the Scherrer's equation as follows [16]

$$D = K\lambda/(B\cos\Theta)$$

where D is the crystallite size, K is a fixed number of 0.9, λ is the X-ray wavelength of 0.1541 nm, and B is the full width at half maximum of the 36.04° peak. The crystallite height size of the AlN film was 59.9 nm, in agreement with the average size of AlN films deposited on other substrates [17]. The film thickness of 1 μm is larger than the crystallite size of 59.9 nm. Consequently, it is thought that the highly oriented AlN thin film is composed by many *c*-axis oriented small crystals.

We observed the crystal growth of the AlN film in order to clarify the influence of the β-Ta electrode. Figure 3 shows a TEM image of the cross sectional structure. The microstructure of the film is dense and shows no opening between the grains. This film consists of an array of poorly defined fibrous grains and indicates a high degree of alignment of these fibrous grains [18]. This result is consistent with the XRD data that the AlN film is highly oriented. The formation of the texture can be explained with the model proposed by van der Drift [19] that the crystals with higher growth rate in the direction perpendicular to the substrate surface will possess a higher probability of survival and all other crystals will be overshadowed after competitive growth.

We investigated the dependence of the crystallinity and crystal orientation of β-Ta electrodes on their film thickness in order to optimize β-Ta bottom electrodes. Figure 4 shows the result. The (002) β-Ta peak intensity increased and the FWHM of the X-ray rocking curve decreased with increasing the film thickness. These results indicate that the thicker the thickness of the Ta electrode, the higher the crystallinity and crystal orientation of the Ta electrode.

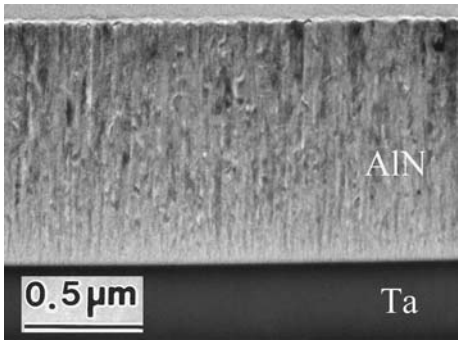

Fig. 3 Cross-sectional TEM photograph of AlN deposited on Ta electrode. The substrate temperature was 100 °C, and the thickness of the Ta electrode was 200 nm

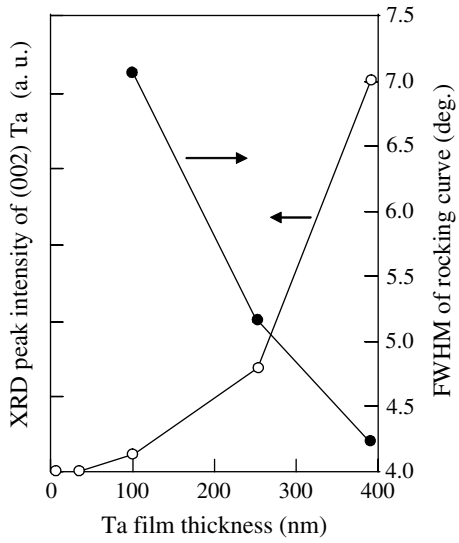

Fig. 4 Dependence of crystallinity and orientation of Ta electrode on film thickness. The substrate temperature was room temperature

Thus we think that thick Ta bottom electrodes are suitable for obtaining oriented AlN thin films.

Since β-Ta layers are metastable phases, they can be transformed into the equilibrium α-phase by annealing [20, 21]. Therefore, we investigated the influence of the substrate temperature on the crystal orientation of the AlN films in order to optimize the sputtering conditions. The dependence of the FWHM of rocking curve of the AlN film on the substrate temperature is shown in Fig. 5. The FWHM increased with increasing the substrate temperature,

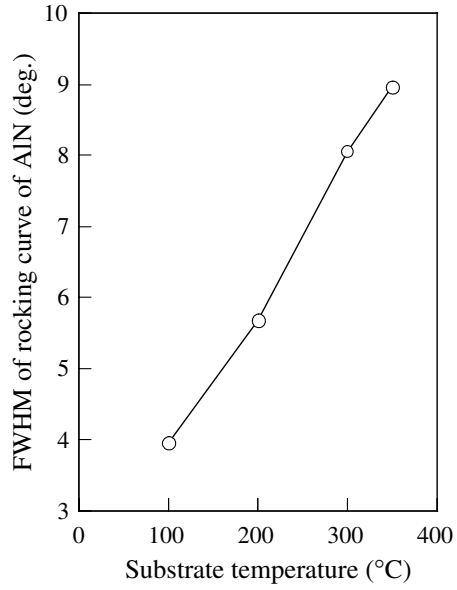

Fig. 5 Dependence of orientation of AlN film deposited on Ta electrode on substrate temperature. The thickness of the Ta electrodes was 200 nm

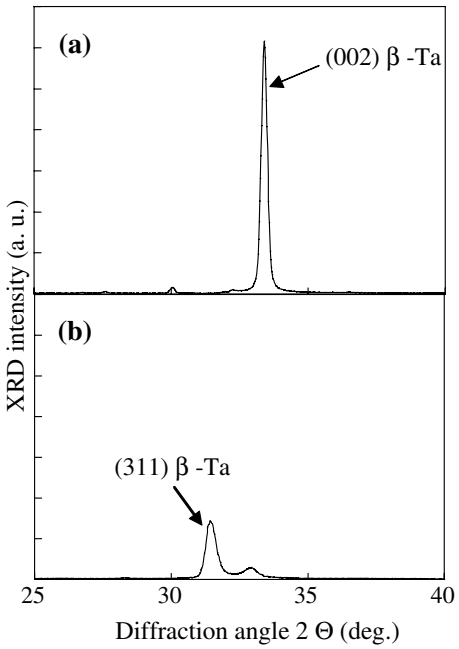

Fig. 6 XRD patterns of Ta electrode at (**a**) room temperature and (**b**) 400 °C. The thickness of the Ta electrode was 200 nm. The time of heat treatment was 1 h

suggesting that AlN films should be prepared at low substrate temperature in order to obtain highly oriented AlN thin films on β-Ta bottom electrodes. The XRD patterns of β-Ta electrodes were measured at various temperatures. The XRD patterns are shown in Fig. 6. A Ta electrode indicated high (002) peak intensity at room temperature. However, the peak intensity drastically decreased around 300 °C, and the Ta electrode indicated (311) β-Ta plane peak around 400 °C. This behavior of the Ta electrode is in agreement with Hoogeveen's data [21]. Moreover, it is reported that the crystallinity and crystal orientation of a bottom electrode strongly influence on the crystal orientation of AlN thin films [22]. Therefore, we think that the crystal orientation of the AlN films is influenced by the substrate temperature owing to the change on the crystallinity and crystal structure of the Ta bottom electrodes.

Conclusions

We investigated the influence of Ta bottom electrodes on the crystallinity and crystal orientation of AlN thin films. The crystal structure of Ta electrodes was tetragonal (β-Ta, a metastable phase), and indicated high crystallinity and *c*-axis orientation at room temperature. The crystallinity and orientation of β-Ta electrodes strongly depended on their film thickness and the substrate temperature. The crystallinity and crystal orientation of AlN films also depended on the Ta electrode thickness and substrate temperature. Consequently, AlN thin films should be prepared at low substrate temperature in order to obtain highly oriented AlN thin films on β-Ta bottom electrodes. The AlN films prepared on the β-Ta electrodes of 250 nm at 100 °C indicated high crystal orientation of the FWHM of 3.9°.

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