Influence of sputtering pressure on polarity distribution of aluminum nitride thin films

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The authors have investigated the influence of sputtering pressure on the polarity distribution of aluminum nitride (AIN) films. They have found that sputtering pressure strongly influences the polarity distribution of AIN films prepared on molybdenum electrodes. The polarity distribution of the AIN films was observed by piezoresponse force microscopy. The polarity orientation is decided with respect to each fine grain constituting the AIN films, and polarity conversion from Al polarity to N polarity is observed with increasing sputtering pressure. The piezoelectric response of the films changes from +3.7 to −4.4 pC/N with increasing sputtering pressure from 0.36 to 4.0 Pa. © 2006 American Institute of Physics. [DOI: 10.1063/1.2405849]

Aluminum nitride (AIN) films have been investigated as an excellent piezoelectric material for bulk and surface acoustic wave applications or for microelectromechanical systems applications. This interest is justified by the excellent chemical and electromechanical properties of AIN films and their compatibility with standard silicon technology. It is commonly accepted that the piezoelectric properties of AIN films are related to the crystal orientation, which is often evaluated by the full width at half maximum (FWHM) of the x-ray rocking curve of (0002) AIN reflection.1–2 However, it was reported that the other factors such as polarity orientation, oxygen content, substrate materials, and grain size, also influence the piezoelectric response of AIN films.3–6 Thus we have investigated the polarity distribution as a significant factor, because the macroscopic piezoelectric response of AIN films is the result of the polarity distribution. Furthermore, the influence of sputtering conditions on polarity distribution has not been reported, although there are reports on the polarity orientation control of III-nitride films using nitridation, substrate polarity, growth temperature, or buffer layers.7–10 Piezoresponse force microscopy (PFM) has been used to image domain structure in ferroelectric thin films and polarity distribution in III-nitride films with nanometer scale spatial resolution.11–14 However, there are no reports that the polarity distribution of AIN films consisting of fine grains was observed by PFM. In this study, we prepared c-axis oriented AIN thin films on molybdenum (Mo) bottom electrodes by rf magnetron sputtering and investigated the influence of sputtering pressure on the polarity distribution and piezoelectric response of the AIN films. The polarity distribution was observed by PFM.

AIN films and Mo bottom electrodes were prepared on (100) Si substrates in a rf magnetron sputtering system (CFS-4ES, Tokuda). The thickness of the Mo electrodes was 200 nm. The optimized sputtering conditions of the AIN films were reported in our previous studies, and the conditions are shown in Table 1.15 The sputtering targets were 75 mm diameter 99.999% pure aluminum and 99.9% pure molybdenum, respectively. The sputtering chamber was evacuated to a pressure below 2×10−4 Pa, and then high-purity argon 99.999% and nitrogen 99.999% were introduced. Before deposition, the aluminum and molybdenum targets were cleaned under the same deposition conditions for 5 min with the shutter closed. The substrates were not heated and self-heating of plasma gave a substrate temperature of approximately 80 °C. The crystal structure and orientation of the AIN films were investigated by x-ray diffraction (XRD; M03X-HF, Mac Science) with Cu Kα radiation. The piezoelectric response of the AIN films was measured with a piezometer system (PM100 Piezotest). The film thicknesses were measured by field emission scanning electron microscopy (S-4300, Hitachi). In this study, the positive sign means that the polarization of the AIN films is predominantly oriented pointing from the film surface towards the substrate interface. The negative sign means the opposite. The PFM observations were performed using a scanning probe microscope (SPI-3800N, Seiko Instr. Inc.) and a lock-in amplifier equipped with an ac signal source at a modulation frequency of 10 kHz with a driving voltage of 50 V applied to the tip.

<table>
<thead>
<tr>
<th>Material</th>
<th>AIN</th>
<th>Mo</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sputtering pressure (Pa)</td>
<td>0.36–4.0</td>
<td>0.3</td>
</tr>
<tr>
<td>Substrate temperature</td>
<td>Self-heating</td>
<td>Self-heating</td>
</tr>
<tr>
<td>Gas composition</td>
<td>Ar:N2=1:1</td>
<td>Ar</td>
</tr>
<tr>
<td>rf power (W)</td>
<td>400</td>
<td>100</td>
</tr>
<tr>
<td>Sputtering time</td>
<td>1–14 h</td>
<td>30 min</td>
</tr>
<tr>
<td>Target substrate spacing (cm)</td>
<td>&gt;8.3</td>
<td>&gt;8.3</td>
</tr>
</tbody>
</table>

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The contrasts of the PFM images were obtained only from the phase difference between the ac signal and piezoelectric response. In the wurtzite III-nitrides, the spontaneous polarization points from the N atom to the nearest neighbor III-metal atom (anion to cation) along the crystallographic c axis, therefore, upward (N polarity) and downward (Al polarity) spontaneous polarizations were represented by dark (out of phase) and bright (in phase) contrasts, respectively.

We prepared AlN thin films on Mo bottom electrodes at different sputtering pressures ranging from 0.36 to 4.0 Pa. We investigated the influence of the sputtering pressure on the crystal structures of the AlN films. Figure 1 shows the XRD patterns of the films. The crystal structures of the AlN films are wurtzite and the crystal structures of the Mo bottom electrodes are body-centered cubic (bcc). The AlN films and the Mo electrodes exhibit AlN (0002) and Mo (110) preferred orientations in all the samples. The peak intensity of the AlN (0002) phase decreases with increasing sputtering pressure. Figure 2 shows the dependence of the AlN film thickness and the FWHM of the (0002) AlN rocking curves on the sputtering pressure. The AlN film thickness decreases with increasing sputtering pressure, because the aluminum particles sputtered from the aluminum target have higher probability to collide with argon or nitrogen gas on its way to the substrate, and the growth rate of the films decreases. Therefore, the decrease of the AlN peak intensity is due to the decrease of the AlN film thickness. On the other hand, the FWHM values slightly increases with increasing sputtering pressure, and are from 6.9° to 10.0°, as shown in Fig. 2. In comparison with the reported values of highly c-axis oriented AlN films, the values are larger. Thus the crystal orientation of the AlN films is comparatively lower. It is thought that the crystal orientation of the AlN films is decreased by the crystal orientation of the Mo bottom electrodes which exhibit large FWHM values from 11.3° to 12.6°, because the surface roughness (Ra) of the Mo electrodes is 0.5 nm and the surface roughness is small. Furthermore, it is known that the migration energy of the adatoms decreases with increasing sputtering pressure, because the adatoms have more collisions with gas atoms under higher pressure. Thus it is considered that they will not be able to reach the stable sites, and the crystal orientation of the AlN films is lower.

We measured the piezoelectric response of the AlN films. Figure 3 shows the dependence of the piezoelectric response on the sputtering pressure. The piezoelectric response changes from +3.7 to −4.4 pC/N, indicating polarity conversion from Al polarity to N polarity with increasing sputtering pressure. It is thought that the polarity conversion results from a reversal of the predominant polarity orientation in the AlN films. It is interesting that the AlN films exhibit relatively high piezoelectric response, although the crystal orientation of the AlN films is lower, as shown in Fig. 3. Furthermore, we prepared AlN films at 0.36 Pa for 1 h (film thickness: 250 nm) and at 4.0 Pa for 14 h (film thickness: 1270 nm) in order to confirm the influence of the AlN film thickness on the piezoelectric response. The piezoelectric responses of the AlN films were +4.1 and −2.9 pC/N, respectively. The polarity orientation hardly depends on the film thickness. Thus we found that sputtering pressure is one of the significant control factors for the polarity conversion of AlN films.

We observed the AlN film surfaces by PFM in order to investigate the polarity distribution in detail. Figure 4 shows the atomic force microscopy (AFM) and PFM images of the AlN films deposited at different sputtering pressures of 0.36, 1.5, and 4.0 Pa. The piezoelectric responses of the films were 3.7, 0.2, and −4.4 pC/N, respectively. The sputtering pressure influences the surface morphology as shown in Figs. 4(a)–4(c). The grain size of the AlN films becomes smaller with increasing sputtering pressure. According to the PFM images, the polarity orientation is decided with respect to each fine grain constituting the AlN films. The PFM image of the AlN film deposited at 0.36 Pa indicates that the bright contrast regions are large, as shown in Fig. 4(d), indicating that the Al-polarity grains are predominant. The PFM image of the film prepared at 1.5 Pa means that the bright and dark contrast regions are randomly mixed, as shown in Fig. 4(e).
The piezoelectric response of the film was 0.2 pC/N and was very weak. The PFM image of the film deposited at 4.0 Pa shows the dark contrast regions over the entire surface, as shown in Fig. 4(f), suggesting that the polarity is well aligned in N polarity. The PFM observation results are consistent with the piezoelectric response. It is clear that piezoelectric response strongly relates to polarity distribution in AlN films. Moreover, it is interesting that both Al- and N-polarity regions are randomly mixed and can coexist, because there is no clear evidence of the mixing state. It is reported that Al-polarity AlN growth is expected under Al-rich conditions by first-principles calculations, because the energy difference between Al polarity and N polarity of AlN films is very small.

Therefore, we think that the decrease of the sputtering pressure increases the number of the sputtered aluminum particles arriving at the substrate surfaces and makes Al-rich conditions. Consequently, the AlN films indicate Al polarity with decreasing sputtering pressure. However, further investigations are needed to fully understand the polarity conversion mechanism.

In summary, we have found that the sputtering pressure significantly influences the polarity distribution and piezoelectric response of the AlN films prepared on the Mo bottom electrodes. The piezoelectric response changes from 3.7 to −4.4 pC/N, increasing the sputtering pressure from 0.36 to 4.0 Pa. It indicates that the predominant polarity of the AlN films changes from Al polarity to N polarity with increasing sputtering pressure. Furthermore, we observed the polarity distribution in the AlN films by PFM and declared that the polarity orientation is decided with respect to each fine grain.

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FIG. 4. (Color online) AFM and PFM images of surfaces of AlN films deposited at different pressures: 0.36, 1.5, and 4.0 Pa. (a) and (d), (b) and (e), and (c) and (f) are identical places, respectively. The white line means grain boundary in (d), (e), and (f).