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High resolution transmission electron microscopy study on the microstructures of aluminum nitride and hydrogenated aluminum nitride films prepared by radio frequency reactive sputtering

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Aluminum nitride (AlN) and hydrogenated aluminum nitride (AlN:H) films on Si substrate have been deposited by the radio frequency reactive magnetron sputtering. The microstructures of the two films have been examined and compared by transmission electron microscopy (TEM) and high-resolution TEM. In the growth of the AlN:H film, it has been observed that the amorphous phase is formed at the initial stage of deposition, and c-axis oriented crystallite nucleates at the amorphous layer. The lattice mismatch between the film and substrate and the stress in the film are reduced, and the film surface is smooth due to this amorphous phase. A schematic model explaining the growth of AlN and AlN:H films is proposed, and the reason for the easy formation of the amorphous phase in AlN:H film is discussed. © *1997 American Institute of Physics.*
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Recently, the frequency band used by telecommunication systems such as mobile cellular phones and broadcasting satellites is required to be high due to the increasing volume of information and communication media. Since the aluminum nitride (AlN) film with a wurtzite hexagonal structure has a piezoelectric property with high velocity, it has drawn attention as a piezoelectric material for high frequency surface acoustic wave (SAW) devices.[1]

AlN films prepared by radio frequency (rf) reactive sputtering show c-axis preferred orientation normal to the substrate under certain deposition conditions[2] and good SAW filter characteristics.[3] Most work on AlN film growth to date has been focused on obtaining high crystallinity and preferred orientation.[2–5] Nowadays, several notable results have been reported from experiments with hydrogen gas added to reactive gas during the deposition of AlN film. Wang *et al.*[6] have reported that the hydrogenated aluminum nitride (AlN:H) films are nanocrystalline with preferred c-axis orientation and an extremely smooth surface. Yong *et al.* have observed that the c-axis preferred orientation is maintained up to 10% of H_2 addition to reactive gas, the surface becomes smooth and the stress is relieved as the content of H_2 increases.[7] Therefore, it has been found that the electromechanical coupling coefficient of SAW filter using AlN:H film (10% H_2 addition) (1.35%) is larger than that using AlN film (0.83%) because of the improvement of film properties by H_2 addition.[7]

Although AlN:H film has some special properties and a lot of advantages, the reason for the differences in properties between AlN and AlN:H films mentioned above is not known. In order to elucidate such observations, the atomic scale characterization on the growth of AlN and AlN:H films should be performed. This microstructural information may offer clues regarding the causes of the difference in macroscopic properties between AlN and AlN:H films.

In this article, the observation of microstructures of AlN and AlN:H films, especially that corresponding to the initial stage of the growth, were carried out using transmission electron microscopy (TEM) and high resolution TEM (HRTEM). The reasons for the difference in the property between two films and the advantages of AlN:H film were examined and a schematic model explaining these phenomena was proposed. Moreover, the underlying mechanism for the easy formation of amorphous phase in AlN:H film was also discussed.

The AlN film was fabricated by the rf magnetron sputtering of an aluminum target in the gas mixture of argon and N_2 gas, and the AlN:H film was deposited by adding H_2 gas (10 vol %) to the gas mixture. The purity of all the gases was 99.9999%. The aluminum target was 5 cm in diameter and its purity was 99.999%. The substrates used in this experiment were Si (100) wafers (2.0×2.0 cm^2). Degreasing of substrates was carried out in the ultrasonic baths of acetone, ethanol, and de-ionized water, successively. Native oxide on the Si wafer was removed by etching in the diluted HF solution. The substrates were dried using N_2 gas and immediately inserted into the vacuum chamber. The base pressure was lower than 6.7×10^{-5} Pa (5×10^{-7} Torr). The sputtering conditions for AlN films are listed in Table I, which were selected to obtain c-axis oriented AlN film with a reasonable deposition rate.

A JEOL JEM 2000EX microscope was used in TEM observation. The filament of this apparatus was LaB_6 with an accelerating voltage of 200 kV.

Figure 1 shows a cross-sectional view of AlN:H/AlN bilayered films deposited on a Si substrate. In order to com-

TABLE I. Sputtering condition of AlN and AlN:H films.

Target	Aluminum (99.999%, 5 cm diameter)
Substrate	Si (100) wafer
Base pressure (Pa)	$<6.7\times10^{-5}$ (5×10^{-7} Torr)
Target-substrate distance (cm)	8.0
rf power (W)	250
Sputtering pressure (Pa)	1.1 (8 mTorr)
Ar gas flow rate (sccm)	6.0
N_2 gas flow rate (sccm)	6.0
Amount of H_2 addition (%)	0 or 10

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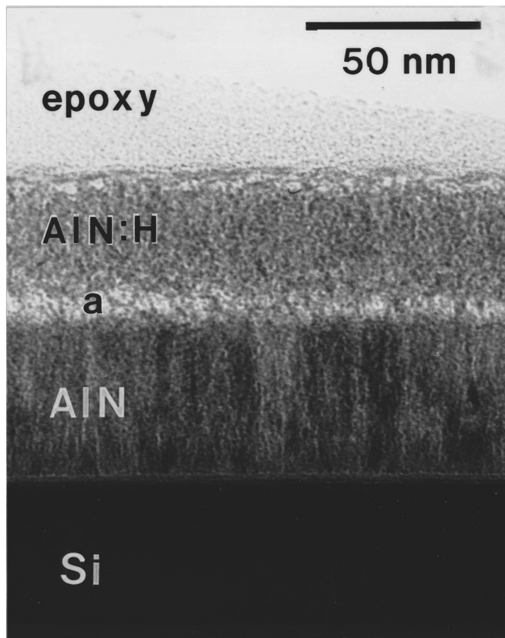

FIG. 1. Cross-sectional TEM micrograph of AlN:H/AlN bilayered films deposited on Si substrate.

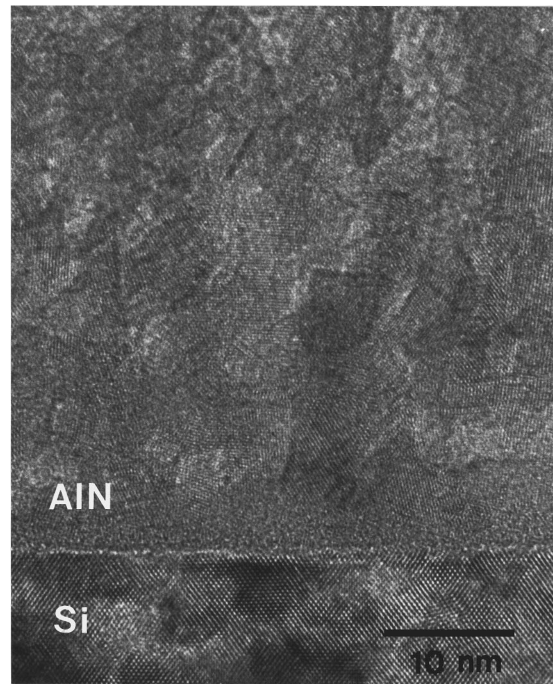

FIG. 2. Cross-sectional HRTEM image of the AlN films deposited on Si substrate.

pare the two films in which other parameters except the H_2 addition are excluded, the two films were deposited one after another without breaking the vacuum. While development of a columnar structure is observed in the AlN layer, no column is found in the AlN:H layer. Also, the surface of the AlN layer (interface between AlN and AlN:H layers) is rough, in contrast to the smooth AlN:H layer. One noticeable point is that the amorphous phase of about 5 nm is formed uniformly at the initial stage of deposition of the AlN:H layer. To observe each film at a greater magnification, HRTEM observations on AlN and AlN:H films have been performed separately.

The cross-sectional HRTEM image of the AlN film on Si substrate is shown in Fig. 2. Randomly oriented crystallites with a size of about 10 nm are confirmed in the AlN film. This randomness in orientation arises from the difference in the lattice mismatch and crystal structure between AlN and Si. The growth of columnar grains on structures of random orientation results in the roughness and stress of the film.

Figure 3 shows the cross-sectional HRTEM image of the AlN:H film on the Si substrate. It is found that the amorphous phase is uniformly formed at the initial stage of growth, which is consistent with Fig. 1. The c-axis oriented crystallite normal to the substrate nucleates at this amorphous layer, while the amorphous region is present around its surrounding. The amorphous phase is a disordered, open structure, so the mismatch with the Si substrate is small. Thus, the stress in the AlN:H film is lower than that in the AlN film. Because a short range ordering exists in the amorphous AlN:H layer, it can offer the nucleation sites for the growth of c-axis oriented crystallite, in which the (0002) plane is parallel with the substrate. Some studies show that the production of oriented thin films on amorphous substrates is related to surface energy of the deposited films.[8,9] According to authors, for a system without any epitaxial relationship between the film and the substrate (e.g., amorphous substrate), islands corresponding to the lowest surface energy can nucleate and grow in an orientation parallel to the substrate to reduce the free energy. In the current system close packed planes, such as the (0002) plane in the hexagonal structure, would have the lowest surface energy. Thus, films of (0002) plane can be easily obtained. The surface of the AlN:H film is smooth because there are only c-axis oriented crystallite and amorphous phases in it.

The schematic model explaining the growth of AlN and

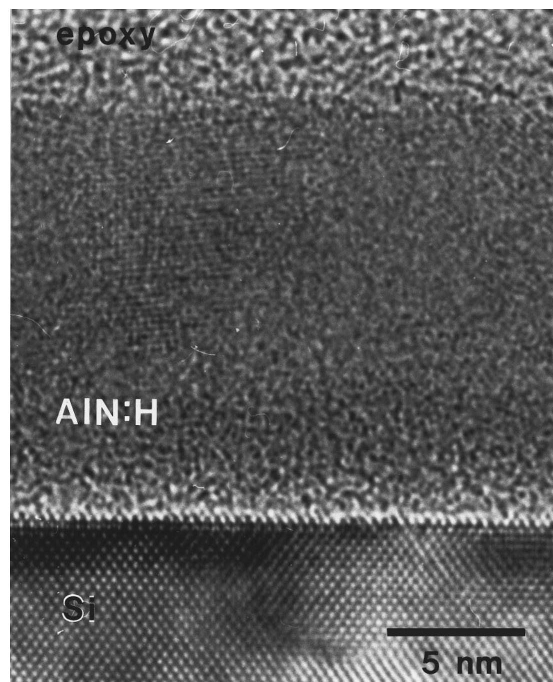

FIG. 3. Cross-sectional HRTEM image of the AlN:H films deposited on Si substrate.

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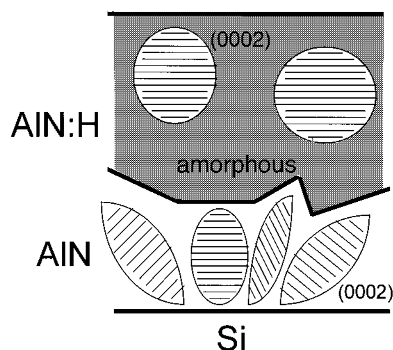

FIG. 4. Schematic model explaining the growth of AlN and AlN:H films.

AlN:H films is shown in Fig. 4. Because lattice mismatch exists between AlN and Si during the growth of AlN film, a large stress is formed in the film and c-axis oriented crystallites are inclined. Thus, the surface of the AlN film composed of these crystallites is rough. However, when the AlN:H film is deposited, an initially formed uniform amorphous layer reduces the lattice mismatch and offers the nucleation sites of c-axis oriented crystallites normal to the substrate. In addition, by surrounding the growing crystallites, the amorphous phase acts as a ''stress-damper.'' Therefore, the resulting surface of AlN:H film is smooth, and the stress is reduced.

Finally, we explain the reason why the amorphous phase is easily formed in AlN:H film. The free energy of formation at room temperature of AlH_3 is 46.516 kJ/mol and that of NH_3 is -16.409 kJ/mol.[10] This means that the formation of NH_3 is spontaneous, but that of AlH_3 is not possible. This difference of hydrogen affinity with the Al and N atom causes hydrogen atoms to preferentially seek interstitial sites surrounded by N atoms. Without a sufficient thermal energy to move Al and N atoms, only an Al atom which is smaller than a N atom can move in short distance, and this induces a lattice distortion, resulting in the loss of long range ordering in the AlN lattice. Thus, similar to the hydrogen induced amorphization behavior in intermetallic compounds,[11] the formation of amorphous phase occurs in this system.

In conclusion, we have discussed some of the advantages of AlN:H film and the difference in properties between AlN and AlN:H films through microstructural study. The amorphous phase plays an important role in the properties such as surface smoothness and stress of AlN:H film. Finally, a possible mechanism for the easy formation of amorphous phase in AlN:H film is proposed.

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[1] T. Shiosaki, T. Yamamoto, T. Oda, and A. Kawabata, Appl. Phys. Lett. **36**, 643 (1980).
[2] H.-C. Lee, G.-H. Kim, S.-K. Hong, K.-Y. Lee, Y.-J. Yong, C.-H. Chun, and J.-Y. Lee, Thin Solid Films **261**, 148 (1995).
[3] H. Okano, Y. Takahashi, T. Tanaka, K. Shibata, and S. Nakano, Jpn. J. Appl. Phys., Part 1 **31**, 3446 (1992).
[4] H. Okano, T. Tanaka, K. Shibata, and S. Nakano, Jpn. J. Appl. Phys., Part 1 **31**, 3017 (1992).
[5] R. D. Vispute, H. Wu, and J. Narayan, Appl. Phys. Lett. **67**, 1549 (1995).
[6] X. D. Wang, W. Jiang, M. G. Norton, and K. W. Hipps, Thin Solid Films **251**, 121 (1994).
[7] Y.-J. Yong and J.-Y. Lee, J. Vac. Sci. Technol. A **15**, 390 (1997).
[8] H. Yoshiyama, S. Tanaka, Y. Mikami, S. Ohshio, J. Nishiura, H. Kawakami, and H. Kobayashi, J. Cryst. Growth **86**, 56 (1988).
[9] N. Fujimura, T. Nishihara, S. Goto, J. Xu, and T. Ito, J. Cryst. Growth **130**, 269 (1993).
[10] Ihsan Barin, *Thermochemical Data of Pure Substances* (VCH, Weinheim, 1989), pp. 38, 942.
[11] Y.-J. Yong and J.-Y. Lee, J. Non-Cryst. Solids **169**, 96 (1994).

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