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Microstructural evolution of the abnormal crystallite grains in sputtered ScAlN film for piezo-MEMS applications

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***Abstract*—Scandium aluminum nitride (ScAlN) thin film has received wide attention for high performance piezo-MEMS device development. This work studies the large-size abnormal grains in the reactive sputtered ScAlN films. Our experimental results prove the abnormal grain nucleates in the normal film matrix and grows larger along the film thickness direction. The local in-plane strain induced by the lattice deformation results in atomic stacking fault, which is suggested to drive the abnormal nucleation.**

***Keywords*—Scandium aluminum nitride, reactive sputtering, abnormal grains, stacking fault, nucleation and growth**

I. Introduction

Scandium aluminum nitride (ScAlN) with Wurtzite crystallographic structure has been demonstrated as an excellent piezoelectric material for GHz electronic device development [1, 2]. The films comprising high concentrations of Sc exhibit enhanced electromechanical coupling coefficients, which makes them more attractive [3]. Highly textured film with *c*-axis orientation is desired for strong piezoelectric response.

When the Physical-Vapor-Deposition (PVD) technologies are employed to deposit ScAlN films, very large-size (usually several hundred nm in diameter) grains are found on film surfaces, especially in those comprising high concentration of Sc. These abnormal crystallite grains protrude out of the film surface, leading to poor film flatness and degrading the piezoelectric response.

The abnormal grains have been confirmed being Wurtzite-type crystallites but with their out-of-plane orientation different from the normal *c*-axis direction [4, 5]. Their presence highly relates to the deposition process. It is clear that relative low sputtering power and relative high pressure would reduce the *c*-axis stability [6]. The solutions to suppress these abnormal grains include modification of magnetic field in sputter apparatus [7], choosing proper substrate [8], and increasing in the distance between the target and the substrate [4].

To further develop the ScAlN film with higher Sc concentration and better film quality, physical understanding of the nucleation and microstructural evolution processes is necessary. In this paper, we conduct in-depth material research to characterize the crystallographic features of the abnormal grains and uncover their microstructure evolution during the film deposition.

II. Experimental

A. ScAlN film deposition

ScAlN film was deposited using pulse DC reactive sputtering method [9] from a ScAl alloy target containing 15 at. % of Sc. Silicon wafers with 200 mm in diameter were used as the substrates. Firstly, 200 nm molybdenum (Mo) was sputtered, followed by the ScAlN film deposition. The thickness of the ScAlN film varies from 200 to 1000 nm. In some cases, 50 nm aluminum nitride (AlN) is deposited before the ScAlN film. Therefore, two kinds of substrates, Mo/Si and AlN/Mo/Si were provided on which the ScAlN films were prepared.

B. Material characterization

The film surface morphology and the cross-section were characterized using Defect Review Scanning Electron Microscopy (DRSEM) and Transmission Electron Microscopy (TEM), respectively. Focused Ion Beam (FIB) milling was used to prepare the TEM lamella. Energy-dispersive X-ray Spectroscopy (EDX) equipped with the TEM was used for the stoichiometric composition conformation and the composition uniformity assessment. For the crystalline phase determination and the texture structure evaluation, X-ray Diffraction (XRD) is employed.

III. Results and discussion

EDX confirmed the atomic Sc/Al ratio in the ScAlN films is 15/85, the same as that in the target. The nitrogen concentration varies in the range of 50.6 – 51.9 %. Taking the measurement accuracy into consideration, it is concluded that the stoichiometric nitride films were achieved. In addition, the oxygen was hardly to detected in all ScAlN films, suggesting that its concentration is less than 2 %.

XRD θ - 2θ scan patterns show pronounced (0002) diffraction peaks at 2θ = 35.9º. The Full-Width-at-Half-Maximum (FWHM) of ≤ 1.6º was obtained on the (0002) rocking curve of the 1000 nm ScAlN films, demonstrating the highly *c*-axis textured structure of the films. It is also noticed that both the (0002) diffraction peaks and the (0002) rocking curves become narrower as the film thickness increasing, indicating larger grain size and better textured structure in thicker films.

The bright-field TEM image as in Fig. 1(a) shows the cross-section of the 1000 nm ScAlN film deposited on Mo/Si substrate. Columnar growth along [0001] direction is confirmed by the Selected Area Electron Diffraction (SAED) pattern as the inset. The cone-shape abnormal grain is clearly observed in the zoomed-in image (Fig. 1b). It starts from the normal *c*-axis textured matrix near the bottom interface and grows to be larger along the thickness direction. On the film surface the grain becomes 300 nm in lateral size and is more than 100 nm protruding out of the surrounding matrix. The box marks the region where the high-resolution TEM micrograph was recorded and shown in Fig. 2(a).

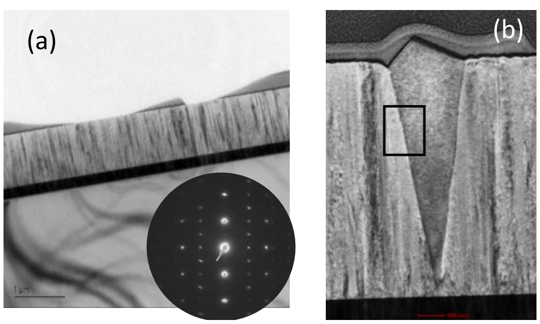

Fig. 1. The bright-field TEM images of the cross-section of 1000 nm ScAlN film sputtered on Mo/Si substrate. The low-resolution image (a) and the SAED pattern as the inset confirms an overall *c*-axis textured Wurtzite structure. The zoomed-in image (b) highlights the cone-shape abnormal cryatllite grain which nucleates in the normal film matrix near the bottom interface and grows through the entire film thickness. The box marks the region where the high-resolution TEM micrograph was taken.

In the high-resolution TEM image as in Fig. 2(a), a grain boundary between the abnormal and the normal columnar grains is highlighted by the orange color dash lines. The zig-zag shape grain boundary demonstrates the abnormal grain grows wider and wider along the film thickness direction, at expense of the normal grains beside it. Fig. 2 (b), (c) and (d) show the Fast-Fourier-Transformation (FTT) patterns at the regions of the red boxes 1, 2, and 3 marked in Fig. 2 (a), which suggest that both the abnormal and the normal grains are in Wurtzite structure. Atomic planes in both the normal and the abnormal grains are marked with the yellow color solid lines. While the normal grains grow along the *c*-axis direction with their [0001] orientation out-of-plane, the abnormal grain tilts by 65° with respect to the substrate normal. As a result, its [0001] orientation becomes in-plane. Our experimental results coincide with the previous reports [4, 5].

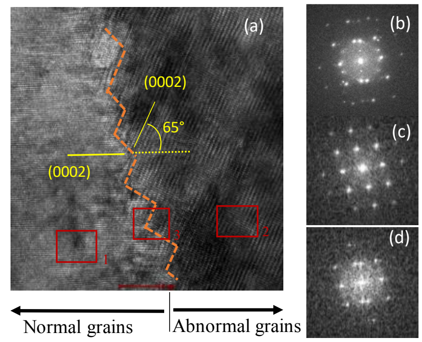

Fig. 2. High-resolution TEM image (a) of the normal and the abnormal grains and the grain boundary marked with the orange color dash line. The atomic planes are clearly dsitinguished and marked with the yellow color solid lines. The FFT patterns in (b), (c) and (d) corespond to the positions marked with the red boxes 1, 2, and 3 in (a).

It is known [8] that the presence of the abnormal grain depends on the substrate material on which the ScAlN film is deposited. To fix the starting positions of the abnormal grains, the dark-field TEM images were recorded as in Fig. 3. On the Mo surface (Fig. 3(a)), after about 10 nm *c*-axis textured film deposition, the abnormal grain appears. But on the AlN surface (Fig. 3(b)), the abnormal grain would not to be seen until more than 100 nm normal film is deposited. It suggests that the abnormal grain nucleates in the normal *c*-axis textured film matrix. Moreover, the substrate affects the driving force of the abnormal nucleation.

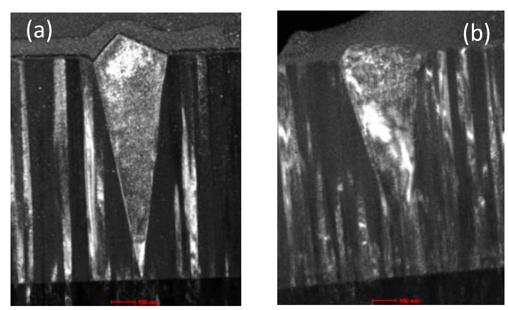

Fig. 3. Dark-field TEM images showing the cross-section of the cone-shape abnormal grains grown in the ScAlN films deposited on (a) Mo/Si and (b) AlN/Mo/Si substrates. The distances from the substrate surface to the bottom of the abnormal grains are about 10 nm and more than 100 nm on the Mo/Si and AlN/Mo/Si, respectively.

The microstructural evolution of the abnormal grain is studied on ScAlN films in various thicknesses on the AlN/Mo/Si substrates. Film surface morphology was characterized using DRSEM as shown in Fig. 4. All micrographs were recorded in the 8-inch wafer center regions. On the 200 nm film, the abnormal grain is hardly found, so the grain size was determined with help of TEM cross-section viewing. As the film thickness increasing, the abnormal grains becomes larger on the film surface and more protruding out of the film matrix. Their sizes are thus easily determined. Fig. 5 plots the abnormal grain sizes on the film surfaces as a function of the film thickness. The mean value of the grain size increases rapidly as the film thickness increasing. Moreover, the spread of the grain size values becomes wider, indicating that the multiple independent abnormal nucleation and growth happened in the film matrix and lasted for the entire film sputtering process.

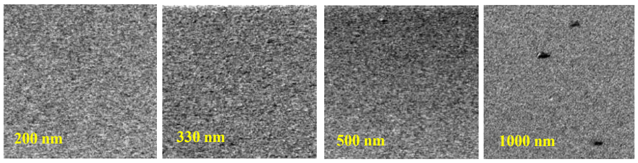

Fig. 4. DRSEM images showing the top surface morphology of the ScAlN films in various thicknesses on AlN/Mo/Si substrates. The side length of each micrograph is 5.0 µm. The abnormal grains become more obvious as the film thickness increasing.

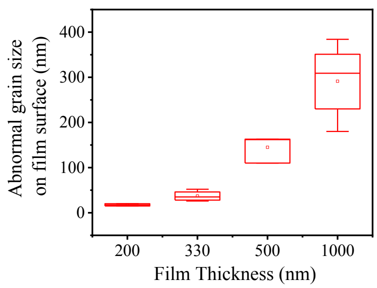

Fig. 5. Plot of the abnormal grain size on the film surface as a function of the film thickness. The abnormal grains grow larger and their sizes spread wider in thicker films.

The presence of the abnormal grain can be attributed to the fault atomic stacking in the normal *c*-axis textured film matrix. When a Sc atom is incorporated into Wurtzite crystal lattice and occupies the Al site, local lattice deformation is induced by the difference of the atomic radius (r_{Sc} = 160 pm, r_{Al} = 125 pm). This deformation generates in-plane compressive strains and causes atomic stacking fault. Both {0001} and $\{11\bar{2}0\}$ facets start to develop rather than stacking along *c*-axis only, which forms the nucleus for the abnormal growth. The abnormal nuclei may generate anywhere in the film matrix when the strain level becomes high enough to trig the atomic stacking fault. When more Sc atoms are doped, more serious lattice deformation is expected and responsible for the presence of more abnormal grains. On the other hand, proper substrate would provide better tolerance to the in-plane strains, and thus retard the abnormal nucleation.

During the ScAlN film sputtering, lower adatom mobility on {0001} facets has been identified [5]. Since the atoms always transfer from the high mobility surface to the low mobility surface, more atoms migrate to {0001} surfaces. The fast growth along [0001] direction promotes the $\{11\bar{2}0\}$ facets evolution. Due to the shadowing effect in sputtering, the abnormal grains capture more particles, allowing them overgrow the surrounding matrix and lateral sizes increasing with the film thickness. One more experimental observation is that less compressive residual stress is found in the region where the abnormal grains present. We believe the abnormal grain nucleation and growth are able to release the local compressive stress through compensating the local lattice deformation and annihilation of grain boundaries during the vary large-size abnormal grain growth.

IV. Conclusions

The large-size and protruding-out abnormal grains are found in sputtered ScAlN films, leading to poor crystal quality. This work investigates the nucleation and growth processes of the abnormal grains in sputtered ScAlN film. We found the abnormal grains nucleate in the normal *c*-axis textured film matrix. As the film thickness increasing, the size of the abnormal grains not only increases to bigger but also spreads wider. Atomic stacking fault induced by the local lattice deformation is identified as the cause of the abnormal nucleation. Although this intrinsic lattice deformation cannot be avoided in ScAlN films, proper substrate can provide better tolerance to the deformation-induced strain and therefore prevent the abnormal nucleation. Sputtering optimization can also contribute to high quality ScAlN film development.

References

[1] Y. Zhu, N. Wang, G. L. Chua, C. Sun, N. Singh, and Y. Gu, "ScAlN-based LCAT mode resonators above 2 GHz with high FOM and reduced fabrication complexity," IEEE Electron Device Lett., vol. 38, pp. 1481–1484, October 2017.

[2] N. Wang, Y. Zhu, G. L. Chua, B, Chen, S. Merugu, N. Singh, and Y. Gu, "Over 10% of k_{eff}^2 demonstrated by 2-GHz spurious mode-free $Sc_{0.12}Al_{0.88}N$ laterally coupled alternating thickness mode resonators," IEEE Electron Device Lett., vol. 40, pp. 957-960, Junne 2019.

[3] M. Akiyama, T. Kamohara, K. Kano, A. Teshigahara, Y. Takeuchi and N. Kawahara, "Enhancement of piezoelectric response in scandium aluminum nitride alloy thin films prepared by dual reactive cosputtering," Adv. Mater., vol. 21, pp. 593–396, February 2009.

[4] S. Fichtner, N.Wolff, G. Krishnamurthy, A. Petraru, S. Bohse, F. Lofink, S. Chemnitz, H. Kohlstedt, L. Kienle, and B. Wagner, "Identifying and overcoming the interface originating c-axis instability in highly Sc enhanced AlN for piezoelectric micro-electromechanical systems," J. Appl. Phys., vol. 122, pp. 035301, July 2017.

[5] C. S. Sandu, F. Parsapour, S. Mertin, V. Pashchenko, R. Matloub, T. LaGrange, B. Heinz, and P. Muralt, "Abnormal grain growth in AlScN thin films induced by complexion formation at crystallite interfaces," Phys. Status Solidi A, vol. 216, pp. 1800569, January 2019.

[6] A. Iqbal, and F. Mohd-Yasin, "Reactive sputtering of aluminum nitrides (002) thin films for piezoelectric applications: a review," Sensors, vol. 18, pp. 1797–1817, June 2018.

[7] T. Yeh, P. Nikkel, K. J. Grannen, C. Feng, and J. Choy, "Fabricating low-defect rare-earth doped piezoelectric layer," US Patent 2015/0311046 A1, October 2015.

[8] B. Heinz, "The growth of piezoelectric $Al_{1-x}Sc_xN$ thin films on Si (100) and Pt (111) using co-sputtering and compound target sputtering," The 13th Annual IEEE Int. Conf. Nano/Micro Eng. Molecular Systems (IEEE-NEMS), Singapore, April 2018.

[9] J. L. Xie, and S. Wickramanayaka, "Effect of process pressure on PVD AlN thin film," IEEE 16th Electronics Packaging Technology Conf. (EPTC), Singapore, Devember, 2014.