Supplementary information:

Understanding Surface Morphology Evolution in Magnetron Sputtered AlN Templates: Mitigating Tensile Stress and Enhancing Crystal Quality

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The crystal orientation of AlN films with different thicknesses:

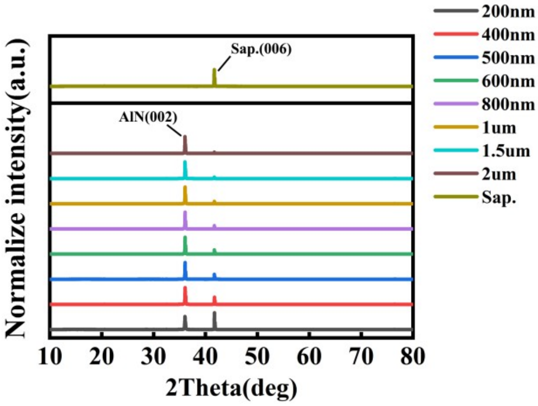

Fig. S1 XRD 2Theta-Omega test of a wide range (10°–80°) on samples with different thicknesses and sapphire substrate.

To investigate whether the "flower-like" morphology corresponds to a special crystal orientation. The wide range (10°–80°) XRD 2Theta-Omega tests were conducted on samples with different thicknesses. The results show that there is no other AlN crystal orientation in the structure, and the XRD results of the thicker samples (the surface is all covered by the "flower-like" morphology) are also dominated by the AlN (0002) plane.

The AFM results of samples with different thicknesses:

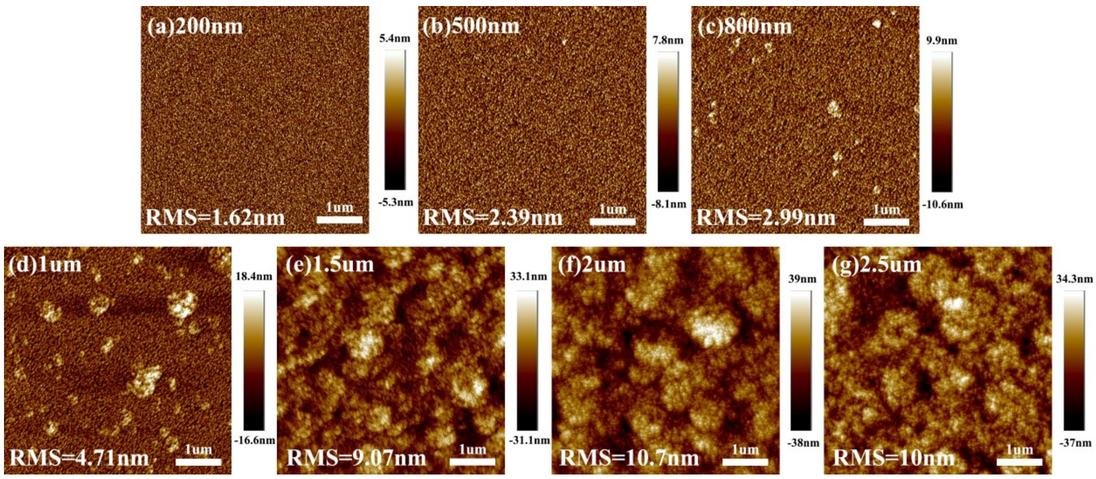

Fig. S2 AFM results of samples with different thicknesses. (a-g) Films with thicknesses of 200nm, 500nm, 800nm, 1 μm, 1.5 μm, 2 μm, and 2.5 μm, respectively.

The AFM images of Fig. S2 samples with thicknesses of 200 nm, 500 nm, 800 nm, 1 μm, 1.5 μm, 2 μm, and 2.5 μm were analyzed. The columnar structure of the 200 nm AlN film has a small grain size, lead to a small R_a and R_q. The R_a and R_q of 500 nm and 800 nm AlN films increase slightly due to the increase of grain size of columnar structure. The surface roughness of 800 nm AlN film is not significantly different from that of 500 nm AlN film because it is in the early stage of "flower-like" morphology formation (complete "flower-like" morphology is not formed). From 1 μm, the sample has basically formed a complete "flower-like" morphology. As the thickness increases (≥1.5 μm), the "flower-like" morphology gradually forms a multi-layer structure. Therefore, the surface roughness is greatly improved. For the 1 μm AlN sample, the height of the "flower-like" structures is approximately 20 nm higher than that of the columnar structures. For the 1.5 μm, 2 μm, and 2.5 μm samples, a complete multi-layer "flower-like" morphology has developed, with the "flower-like" structures rising approximately 40 nm above the columnar structures.

The roughness results of samples with different thicknesses are shown in the Table S1.

Table S1 The surface roughness of samples with different thickness in the range of 5um×5um

thickness(nm)	average roughness R_a(nm)	root-mean-squared roughness R_q(nm)	maximal roughness R_z(nm)
200	1.32	1.62	11.7
500	1.94	2,39	23.6
800	2.38	2.99	37.2
1000	3.66	4.71	46.6
1500	7.19	9.07	72.5
2000	8.45	10.7	86.7
2500	7.96	10	87.7

The raman mapping results of samples:

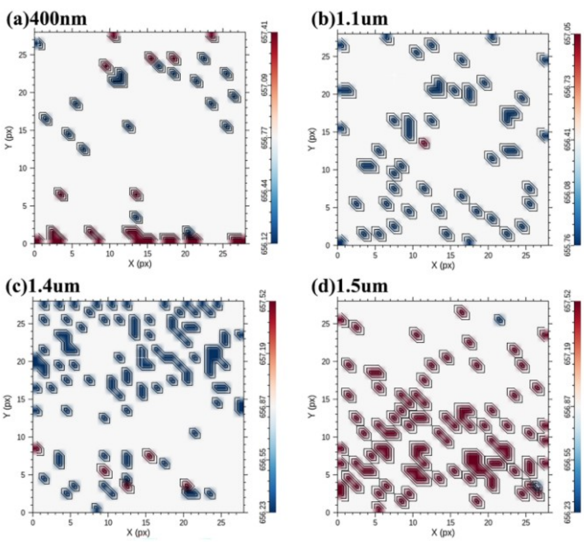

Fig. S3 Raman mapping of samples with different thicknesses. (a-d)Films with thicknesses of 400 nm, 1.1 μm, 1.4 μm, and 1.5 μm, respectively

Raman mapping tests were conducted on AlN films of varying thicknesses, with a scanning area of 4 um×4 um and a step size of 0.15 um, shown in Fig. S3. The results indicate that, regardless of the film thickness, the stress distribution across the entire AlN film is uniform, which confirms the reliability of our stress characterization results. Additionally, the test results suggest that the "flower-like" morphology is a macroscopic manifestation of the overall stress release within the film.

The SEM results of samples with varying thickness across different batches:

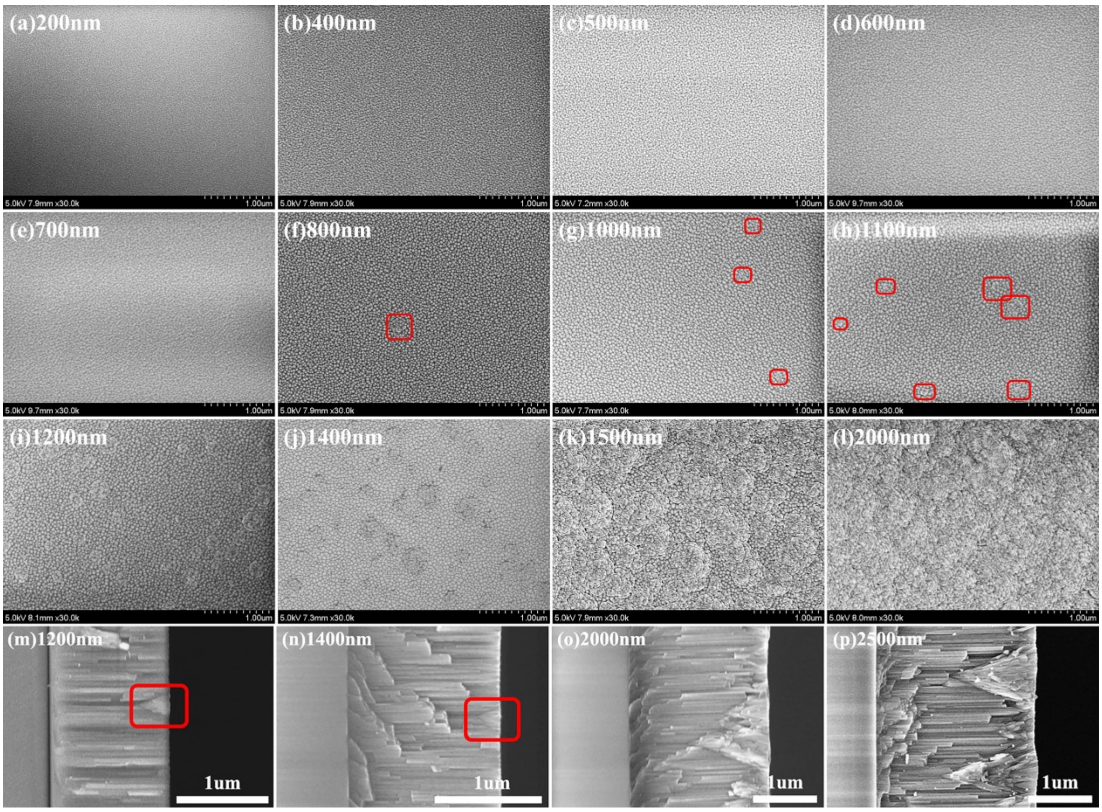

Fig. S4 SEM results of samples with varying thickness across different batches. (a–l) Surface morphology of the films with thicknesses of 200 nm, 400 nm, 500 nm, 600 nm, 800 nm, 1 μm, 1.1 μm, 1.2 μm, 1.4 μm, 1.5μm and 2 μm, respectively. (m–p) Cross-sectional of the films with thicknesses of 1.2 μm, 1.4 μm, 2 μm and 2.5 μm, respectively.

To ensure the accuracy and reliability of the phenomenon, multiple sets of repeated experiments were conducted under different gradients and ranges: ①200 nm to 800 nm, with a gradient of 200 nm; ②500 nm to 2000 nm, with a gradient of 500 nm;

③200 nm to 1400 nm, with a gradient of 300 nm; ④500 nm to 1200 nm, with a gradient of 100 nm. The results from all these experiments consistently show that when the AlN film thickness reaches approximately 800 nm to 1000 nm, the “flower-like” morphology begins to randomly appear on the surface. Moreover, the proportion of this morphology increases as the thickness grows, leading to the formation of a multi-layer structure. The surface morphology of the samples with different thicknesses are shown in Fig. S4(a)–(l). Due to the low density and incomplete structure during the initial formation of “flower-like” morphology, the cross-section testing may not be able to detect it. Therefore, the cross-section only show partial thicknesses of the samples in Fig. S4(m)–(p).

The relationship between the quality of AlN films with and without annealing:

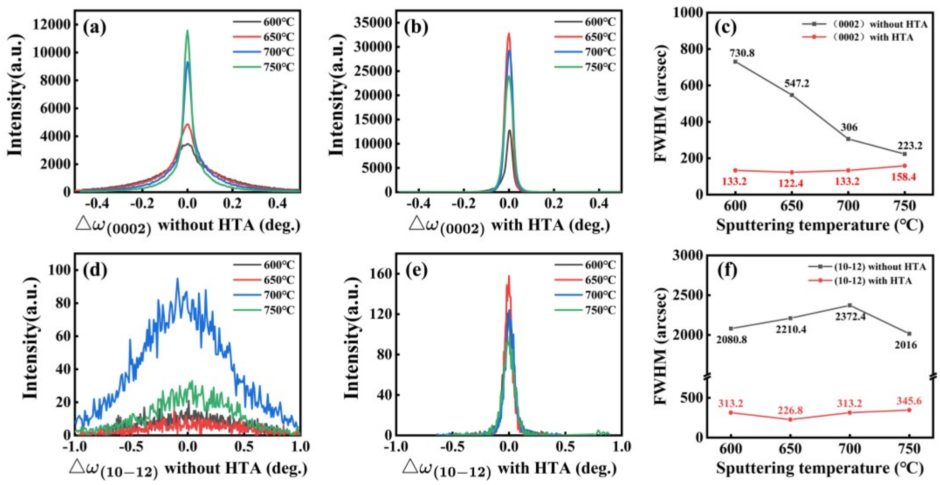

Fig. S5 XRD results of 200 nm AlN films sputtered at different temperatures (600℃, 650℃, 700℃, 750℃).

Since the 2 μm AlN without “flower-like” morphology exhibited superior crystallographic quality compared to that with “flower-like” morphology before HTA. To investigate whether AlN with better quality before annealing leads to better quality of AlN films after annealing. The XRD test on the 200 nm AlN films (there is no “flower-like” morphology at such thin thickness) are sputtered at different temperatures (600°C, 650°C, 700°C, 750°C).

Before HTA, the (0002) and (10-12) plane XRC FWHM of AlN sputtered at 750°C is the narrowest among the samples. But after HTA, the quality of AlN sputtered at 650°C is the highest among those samples. The result indicates that it is not correct that the better quality AlN films before HTA lead to better quality AlN films after HTA, which supporting the conclusion that the “flower-like” morphology is harmful to obtain high quality HTA-AlN films.