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## Nucleation mechanism for epitaxial growth of aluminum films on sapphire substrates by molecular beam epitaxy



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### ABSTRACT

Aluminum (Al) epitaxial films with various thicknesses are grown on sapphire substrates by molecular beam epitaxy (MBE). The nucleation evolution of surface morphology and structural property during the growth of Al epitaxial films on sapphire substrates are investigated in detail. It is found that the 10 nm-thick Al epitaxial films grown on the sapphire substrates show the full-width at half-maximum (FWHM) for Al(111) of 0.35° and the root-mean square (RMS) surface roughness of 2.4 nm. When the thickness increases, the surface initially starts to roughen and then becomes smoother. At the same time, the crystal quality of the Al epitaxial films reaches 800 nm, the FWHM for Al(111) is 0.04° and the RMS surface roughness is 0.14 nm, indicating the high crystal quality and flat surface morphology of Al epitaxial films. The corresponding nucleation mechanism of Al epitaxial films grown on sapphire substrates is hence proposed. This work is of great significance for the fabrication of Al-based devices.

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#### 1. Introduction

Nowadays, aluminum (Al) epitaxial film has attracted considerable attentions due to its superior properties [1,2]. So far, Al epitaxial films have been widely used in superconductors, microelectronics devices, temperature sensors, surface acoustic wave devices [3-6]. Therefore, enormous effort has been devoted to epitaxial growth of Al epitaxial films on sapphire and Si substrates [7–10]. Actually, sapphire seems to be one of the most suitable substrates to grow Al epitaxial films due to the following aspects. On the one hand, the lattice mismatch between sapphire substrates and Al epitaxial films is as small as 3.9% [11,12], which is beneficial to the nucleation of Al atoms and achieving high-quality Al epitaxial films. On the other hand, sapphire substrates are of good chemical and physical stability even at high temperature, and are advantageous to the growth of Al epitaxial films which usually take place at high temperature [13,14]. To date, many researchers have devoted considerable effort to the growth of Al epitaxial films on sapphire substrates. For example, Medlin et al. investigated the orientation and microstructure of Al epitaxial films grown on sapphire (0001) substrates [15], and Li's group achieved highquality Al epitaxial films on sapphire substrates by molecular beam epitaxy (MBE) [11,12]. In fact, most researches are focusing on optimizing the growth conditions to obtain high-quality Al epitaxial films. However, it lacks an insightful study on the growth evolution for epitaxial growth of Al films on sapphire substrates.

In this work, Al epitaxial films with various thicknesses have been grown, and the surface morphology and structural property of as-grown Al epitaxial films on sapphire by MBE are studied by various methods, such as *in-situ* reflection high-energy electron diffraction (RHEED), field-emission scanning electron microscope (FESEM), atomic force microscopy (AFM), high-resolution X-ray diffraction (HRXRD), and high-resolution transmission electron microscopy (HRTEM). Based on the results, the corresponding nucleation mechanism of Al films grown on sapphire substrates by MBE is hence proposed.

#### 2. Experimentals

The as-received sapphire (0001) substrates were given a degassing treatment for 30 min in an ultra-high vacuum (UHV) loadlock chamber with a background pressure of  $1.0 \times 10^{-8}$  Torr.

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Afterwards, they were transferred into the UHV MBE growth chamber with a background pressure of  $2.0 \times 10^{-10}\,\text{Torr.}$  The asdegassed sapphire substrates were annealed at 850 °C for 30 min to obtain an atomically flat substrate surfaces. Solid-state highpurity (99.9999%) Al was used as the precursor of Al and the Al comcell was set at 1100 °C. The growth temperature was set as the 750 °C. During the growth, high-purity N<sub>2</sub> (99.99999%) was supplied throughout an inert gas purifier with a flow rate of 1 sccm. The rotation rate of 5 revolutions-per-min for the substrates was set as constant to guarantee the homogeneous thickness of Al epitaxial films. The thickness of Al epitaxial films was grown from 10 to 800 nm to study the nucleation mechanism for Al epitaxial films grown on sapphire substrates. The as-grown Al epitaxial films were characterized by in-situ RHEED, FESEM (Nova Nano SEM 430Holland), AFM (Bruker Dimension Edge, American), HRXRD (Bruker D8 X-ray diffractometer with Cu Ka1 X-ray source  $\lambda$ =1.5406 Å), and HRTEM (JEOL 3000 F, field emission gun TEM working at a voltage of 200 kV with a point to point resolution of 0.23 nm) for surface morphology and structural property, respectively.

#### 3. Results and discussion

*In-situ* RHEED measurement is adopted to monitor the epitaxial behavior of Al/sapphire throughout the whole growth process.

Fig. 1(a) illustrates the RHEED patterns for sapphire substrates after 30 min annealing at 850 °C. As observed, the RHEED patterns are streaky with nearly lined patterns. This result reveals that residual surface contaminants on sapphire surface have been removed, and a very smooth surface has been obtained for the subsequent growth [11,16,17]. Fig. 1(b) displays sharp and streaky streaks for 10 nm-thick Al epitaxial films, which demonstrates the very smooth surface of the Al epitaxial films grown on sapphire substrates in this case. Fig. 1(c) exhibits the RHEED pattern of 50 nm-thick Al epitaxial films. The spotty pattern reveals the rough surface of the as-grown Al epitaxial films in this stage. Fig. 1 (d) shows the streaky and clear RHEED patterns for the 800 nmthick Al epitaxial films, which indicates that Al epitaxial films with the very flat surface have been obtained [18]. Based on the RHEED measurement, an in-plane epitaxial relationship of Al[1-10]//sapphire[1-100] between as-grown Al epitaxial films and sapphire substrates can be obtained [11,19].

The morphology evolution of the Al epitaxial films surface at different stages is studied by AFM and SEM measurements, respectively. Figs. 2(a) and 3(a) indicate the AFM and SEM images of sapphire substrates after annealing process at 850 °C for 30 min. The surface of sapphire substrates is very flat with a root-mean square (RMS) surface roughness of 0.18 nm, which means that the contaminations on the sapphire substrates are removed completely, and is beneficial to the subsequent growth of Al epitaxial films. Figs. 2(b) and 3(b) show the AFM and SEM images for



Fig. 1. RHEED patterns for (a) sapphire substrates after annealed at 850 °C for 30 min. Al epitaxial films with the thickness of (b) 10, (c) 50, and (d) 800 nm.

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