STM STUDY OF THE INITIAL GROWTH STAGES OF AlF_3 ON *Cu*(100)

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The composition, growth mechanism and structure of thin films of insulators deposited on several metal surfaces are topics that have attracted widespread interest in recent years. Work in this field has been motivated by the quality requirements of the thin films needed to develop advanced microelectronic, optical, and magnetic devices, as well as nanometer-scale structures [1]. Aluminium fluoride (AlF_3) films are of particular interest, because of their potential applications in nanometer-scale patterning through electron beam lithography [2], due to that under electron irradiation they show radiolysis, i.e. the desorption of the fluoride with the consequent formation of an aluminium metallic layer [3].
Recently, AlF_3 growth over different substrates has been characterized by means of electron and ion spectroscopies. Sánchez *et al.* reported a layer-by-layer growth of AlF_3 thin films on *Al*(111) surfaces studied by Auger electron spectroscopy (AES) and electron energy loss spectroscopy [4]. Vergara *et al.*, characterized the growth process of AlF_3 films on *GaAs*(110) from sub-monolayer coverages up to several layers, by AES, ion sputter depth profiling and time of flight-direct recoil spectroscopy [5]. In this work, we characterize the AlF_3 film formation in the sub-monolayer regime through scanning tunnelling microscopy (STM). The extreme spatial resolution of STM allows us to gain knowledge about the mechanism of surface diffusion and island formation as a preliminary step of multilayer growth.
In Fig. 1, a series of STM images show the evolution of AlF_3 growth on *Cu*(100). At very low coverages, AlF_3 molecules preferentially nucleate at the substrate step-edges, decorating both sides of them. The nucleation in terraces decreases with their width (Fig 1a), showing even step-flow growth for the narrower ones. For quite low coverages ($\approx$ 0.05 monolayers), the terrace islands display a shape transition from a compact to a fractal-like form (Fig. 1b). Upon further evaporation the fractal-like islands grow in size (Fig 1c). Although, they do not show a complete coalescence, they form a sort of lateral 2D film, which covers the substrate with a single monolayer until 0.80 ML. With further deposition, the covered surface area keeps around 80% but black patches appear over some islands (Fig. 1d). We interpret these dark areas as a bi-layer, or even thicker, islands of AlF_3. This is supported by the fact that the dark areas increase with further depositions. So, the system behaves as at coverages beyond 0.80 ML the 2D growth turns into a 3D islands mode growth.
In order to understand the compact to fractal islands shape transition, we show in Fig. 2 a series of STM images which reveal the evolution of the terrace islands along the growth. In the very beginning stages of the growth, the AlF_3 islands are exclusively compact (Fig. 2a). With increasing coverage the size of the islands evolve until a critical size A_c, of the order of 2.5 nm (island marked with an arrow in Fig. 2b). Then, through the clustering of several critical size islands, bigger islands are formed. As the size of the clustered islands increases, their shapes become more irregular, showing fractal-like (randomly ramified) shapes (Fig. 2b and 2c). So, AlF_3 islands are able to grow in a compact shape only until a critical size (~100 molecules of α-AlF_3). As the growth continues, no new ad-molecules join these compact islands; instead by their aggregation larger islands are formed.
In summary, our STM study shows a lateral 2D AlF_3 film growth until 0.80 ML, changing with further deposition to a 3D islands growth mode. The terrace islands at very low coverages display a compact to fractal-like shape evolution by the clustering of islands of critical size A_c.

References:

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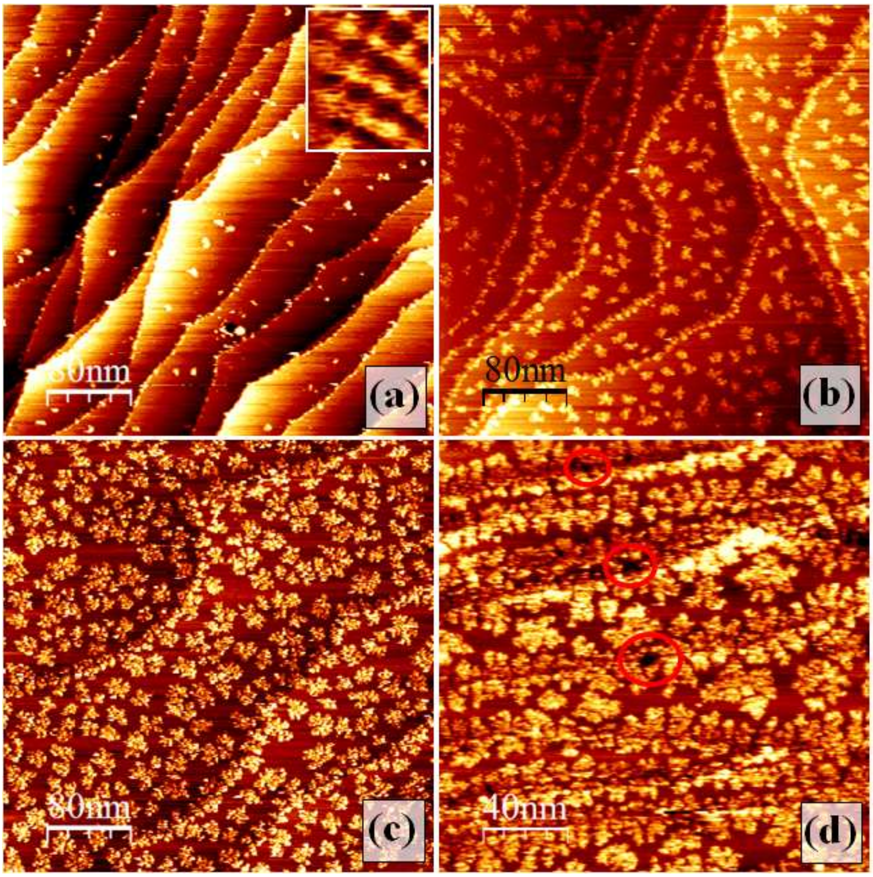

Figure 1: STM images (400 nm × 400 nm) of AlF_3 deposited on Cu(100) at 300 K, (a) 0.05, (b) 0.25 and (c) 0.50 ML. (d) STM image (200 nm × 200 nm) of 0.85 ML of AlF_3, the circles show the black patches mentioned in the text. The images were acquired with a sample bias voltage of V_S= +2.5 V and a tunnel current of I_t= 0.1 nA. The inset in (a) shows a STM image (0.8 nm × 0.9 nm) recorded between the AlF_3 islands displaying atomic resolution on Cu(100). The image was acquired with V_S= +0.2 V and I_t= 1.0 nA.

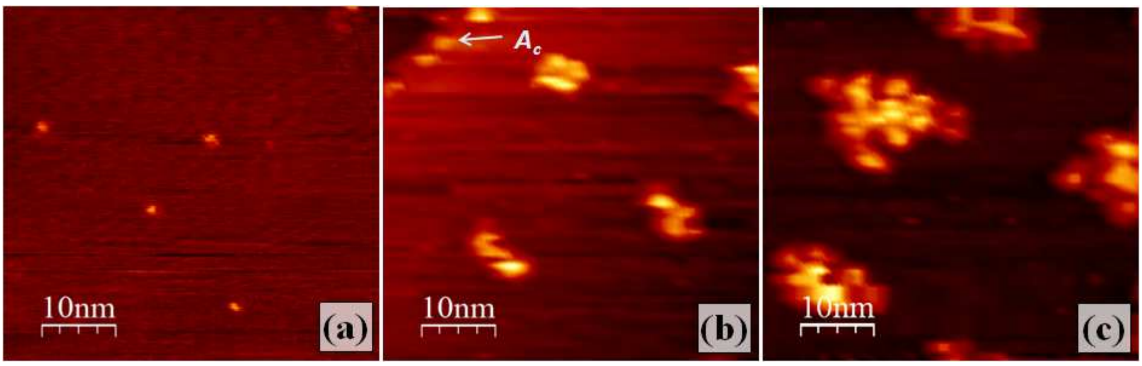

Figure 2: STM images (50 nm × 50 nm) of AlF_3 deposited on Cu(100) at 300 K, (a) 0.02, (b) 0.05 and (c) 0.40 ML. In (b) a critical size (A_c) island is marked. The images were acquired with V_S= +2.5 V and I_t= 0.1 nA.