

### Initial growth stages of $AlF_3$ on $Cu(100)$ : an STM study

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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, for their potential applications in nanometer-scale patterning through electron beam lithography [2], due to under electron irradiation it shows radiolysis, i.e. the desorption of the fluoride with 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. Thus, Sánchez *et al.* reported a layer by layer growth of  $AlF_3$  thin films on  $Al(111)$  surfaces studied by means of Auger electron spectroscopy 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 means of AES, ion sputter depth profiling and time of flight-direct recoil spectroscopy [5]. In spite of this, nothing has been reported concerning with the initial stages of  $AlF_3$  growth on metal surfaces by means of scanning tunnelling microscopy (STM). So, in this work we present a STM study of the initial growth stages of  $AlF_3$  on  $Cu(100)$  at room temperature, as well as the response of the insulating film to annealing treatments.

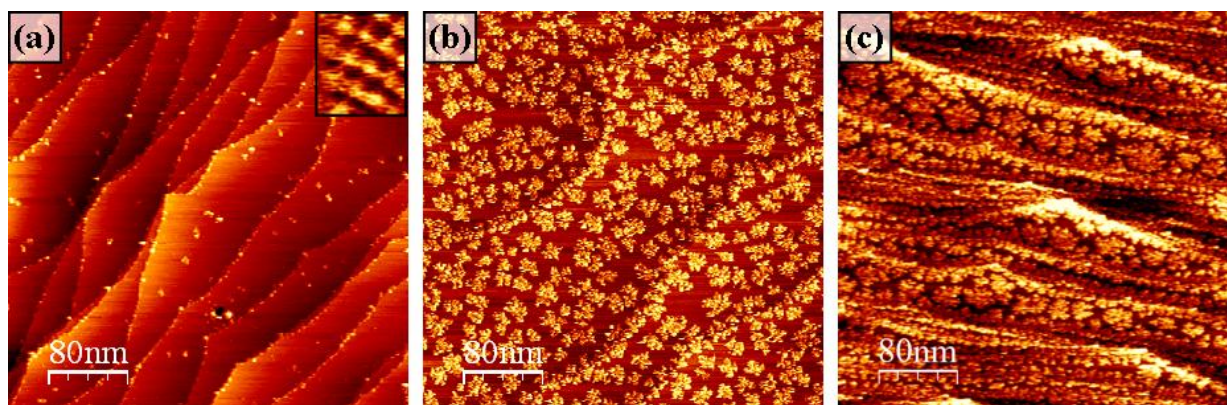
At very low coverages,  $AlF_3$  molecules decorate both sides of the substrate step-edges (Fig. 1a). Once the step-edges are saturated compact islands start to nucleate on terraces (Fig. 1b). Around coverages of 0.1 ML, the terrace islands display a shape evolution from a compact to a fractal-like form (Fig. 1a and 1b). Upon further evaporation the fractal-like islands grow in size. 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 0.8 ML (Fig. 1c). With further deposition the covered surface area is still 80 % but some black patches appear over some islands (Fig. 2a). We interpret these dark areas as bi-layer, or even thicker islands of  $AlF_3$ . So, at coverages beyond 0.8 ML the 2D growth turns into a 3D islands mode growth. This is supported by the fact that the dark areas increase with further depositions of  $AlF_3$ , becoming impossible to acquire STM images at coverages close to 1.5 ML and beyond. On the other hand, a post-annealing treatment of the  $AlF_3/Cu(100)$  surface above 625 K leads to the formation of square islands with a specific azimuthal relation with the high symmetry directions of the  $Cu(100)$  surface, which show a metallic-like behaviour (Fig. 2b and 2c).

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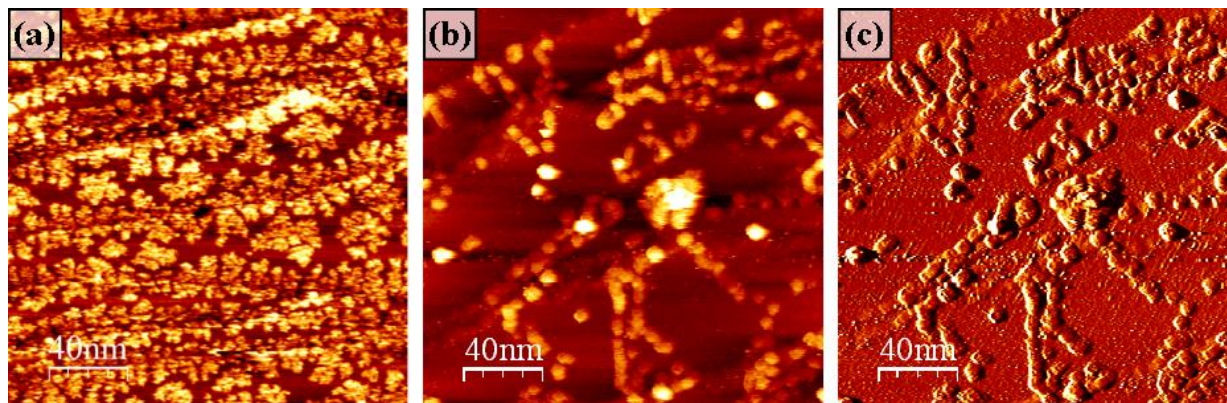
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## Figures:



**Figure 1:** STM images ( $400 \text{ nm} \times 400 \text{ nm}$ ) of  $\text{AlF}_3$  deposited on  $\text{Cu}(100)$  at 300 K, (a) 0.05, (b) 0.5 and (c) 0.8 ML. They were acquired with a sample bias voltage of  $V_s = +2.5 \text{ V}$  and tunnel currents of  $I_t = 0.1\text{-}0.15 \text{ nA}$ . The inset in (a) shows a STM image ( $0.8 \text{ nm} \times 0.9 \text{ nm}$ ) recorded between the  $\text{AlF}_3$  islands displaying atomic resolution on  $\text{Cu}(100)$ . The image was acquired at  $V_s = +0.2 \text{ V}$  and  $I_t = 0.1 \text{ nA}$ .



**Figure 2:** STM images ( $200 \text{ nm} \times 200 \text{ nm}$ ) of 0.85 ML of  $\text{AlF}_3$  deposited on  $\text{Cu}(100)$  at 300 K. Topographic images acquired before (a) and after (b) a post-annealing treatment at 735 K was done. (c) Current STM image acquired simultaneously with the topographic one presented in (b). The images were acquired at  $V_s = +2.5 \text{ V}$  and  $I_t = 0.10\text{-}0.15 \text{ nA}$ .