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ScienceDirect
Acta Materialia 58 (2010) 5150–5159
Acta MATERIALIA
www.elsevier.com/locate/actamat

The role of self-shadowing on growth and scaling laws of faceted polycrystalline thin films

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Received 27 November 2009; received in revised form 21 April 2010; accepted 27 May 2010
Available online 25 June 2010

Abstract

We investigate, via both experiment and simulation, the effects of self-shadowing on the growth of faceted polycrystalline thin films. Faceted aluminum thin films were sputtered and the anomalous scaling behaviour of their surfaces was characterized. To understand the causes of this anomalous behavior, growth of faceted thin films was simulated by coupling a level set construction to a ballistic deposition model. The angular distribution function of deposition flux was varied to control the degree of self-shadowing. We show how differing degrees of self-shadowing strongly modify film surface morphologies and compare these results with experimental findings.

Keywords: Thin film growth; Computer simulations; Grain morphology; Vapor deposition

1. Introduction

Polycrystalline thin films deposited from the vapor phase are important for many technological and scientific applications. Unlike single-crystal or amorphous materials, polycrystalline materials possess structure on scales much larger than atomic lengths. This type of microstructure can significantly modify the mechanical [1,2], electrical [3,4] and thermal [5,6] properties of the film relative to those of its single-crystalline counterpart. Film morphology strongly depends on the deposition technique and parameters used. Because of the large number of possible deposition parameters, many different models have been proposed to explain various aspects of polycrystalline thin film growth [7].

Like many other types of thin film growth, polycrystalline film growth is characterized by self-affine kinetic roughening of the surface [8]. The typical model used to analyze this type of growth is the Family–Vicsek equation. This equation relates root-mean-square (RMS) surface roughness σ (sometimes called interface width) to thickness t and the in-plane system size L [9]:

$$\sigma(L,t) \propto \begin{cases} L^{\alpha} & L^{\alpha} \ll t^{\beta} \\ t^{\beta} & L^{\alpha} \gg t^{\beta} \end{cases} \quad (1)$$

where α is the roughness exponent and β the scaling exponent [9]. With this model, different types of film growth can be classified into universality classes with different values of the exponents (α, β). An example is the growth model described by the well-known KPZ equation [10]; various authors have attempted to find the values of α and β for the KPZ equation in both analytical [11,12] and numerical studies [13].

In many growth systems, however, the scaling laws depend on the local length scale l of the measurement, a scenario described as anomalous roughening [14]. Various authors have introduced an additional universality class exponent to incorporate the anomalous scaling. Examples are a separate local exponent α_{loc} [15], or a new exponent for spectral roughness [16]. Even more recently, several authors have reported a measured experimental dependence of the β exponent on the local length scale l [17,18].

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1359-6454/$36.00
doi:10.1016/j.actamat.2010.05.051

Many studies have been performed to analyze the effects of differing materials, temperature and deposition rate on polycrystalline film growth [19]. However, self-shadowing has received comparatively little attention. In this work, we deposited thin films where both faceting and self-shadowing are expected to occur and measured their surface statistics. We then used simulation to understand the growth of faceted polycrystalline thin films under realistic growth conditions; specifically we incorporate self-shadowing by using a ballistic deposition model. This allows us to model various physical vapor deposition techniques such as sputtering, evaporation or low-pressure chemical vapor deposition (LPCVD). Finally, we analyze the anomalous scaling behaviors of faceted polycrystalline film growth under these different conditions.

2. Faceted polycrystalline thin films

The dominant surface features of many polycrystalline thin films are low-index crystalline facets. The growth rate of these facets determines the equilibrium grain shape (referred to as the kinetic Wulff shape [20]) if the diffusion rate along the low-index facets is much higher than the rate of other diffusion events [21]. The regime of interest is where the deposition temperature, and therefore the adatom diffusion lengths, are high enough to form facets but not high enough for kinetic roughening to occur [22,23]. For some materials this regime occurs for room temperature deposition [24] and for others at elevated temperatures [25,26].

To demonstrate the surface structure of this class of surfaces, we have prepared aluminum thin films of different thicknesses by magnetron sputtering at 200 W (1.7 Å/s) onto cleaned silicon wafers with a thin natural oxide. We have used atomic force microscopy to map the Al film surfaces. Fig. 1 shows the slopemaps of some of the films. The surface slope color channels are linear combinations of the formulae $\theta_x = \tan^{-1}(\partial z/\partial x)$ and $\theta_y = \tan^{-1}(\partial z/\partial y)$.

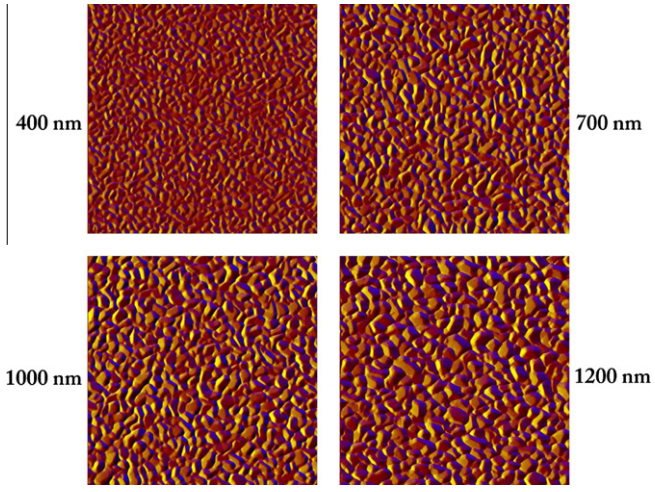

Fig. 1. AFM slopemaps of sputtered Al thin films at four different thicknesses. Each slopemap has side length 2 μm.

From our Al films, we have measured the dependence of the scaling exponent β on local length l by calculating the RMS surface roughness from square windows with side length l. These results are plotted in Fig. 2. A large difference between the local and global scaling exponents is clearly visible. The global value appears to be asymptoting towards $\simeq 0.82$ and the local exponent to $\simeq 0.41$, around half of the global value. Our measured disparity between the local and global exponents is even stronger than those measured by Yanguas-Gil et al. [17]. Our scaling exponents also appear to be asymptoting for small lengths as well as large lengths.

Finally, we have measured the local roughness exponent α_{loc} to be very close to 1 for all films. Since the global roughness exponent α is certainly higher than 1 for our films, their growth can be classified as a super-rough process [16].

3. Simulation methodology

3.1. van der Drift model

Previous simulations of polycrystalline grain growth have typically assumed that the growth rate of each facet is determined only by its crystallographic orientation, and is constant for all times. This simplified model was initially proposed by Kolmogorov [27] and van der Drift [28] and is usually referred to as the van der Drift model. Growth of this type is typical in interface-limited growth such as CVD. Thijssen et al. derived scaling laws for this model and were among the first to simulate competitive grain growth in three dimensions [29]. Smereka et al. used a level set methodology to produce the first true 3-D simulations of this model [30]. In a previous study we used a similar methodology to calculate detailed growth statistics of the van der Drift model in $2+1$ dimensions for CVD [31].

In this study, rather than assuming that the growth of each facet depends on crystallographic orientation, we

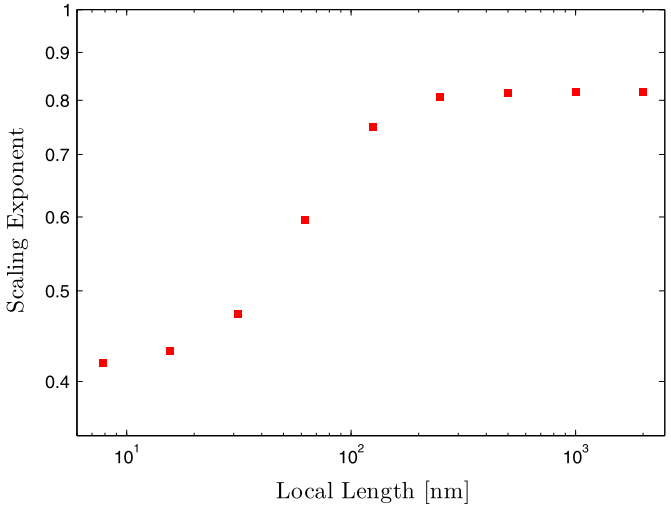

Fig. 2. Scaling exponents of sputtered Al films measured over different length scales.