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Atomic force microscopy study, kinetic roughening and multifractal analysis of electrodeposited silver films

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ABSTRACT

We used atomic force microscopy (AFM) to study surface morphology and kinetic roughening of Ag films. X-ray diffraction (XRD) technique is used to verify the films crystalline structure. The influence of film thickness on the kinetic roughening was investigated using AFM data and roughness calculation. It is revealed that the surface roughness increases with increasing the film thickness. The data also consist with a complex behavior which is called as anomalous scaling. Scaling laws analysis for Ag films presents two distinct dynamics including large local and scale roughness and indicates a power law dependency on the thickness of film.

AFM images have been characterized by the multifractal analysis. This analysis shows that the self-similar and multifractal characteristics as well as anomalous scaling exist in the Ag film morphologies. Description of the quantitative growth and surface morphology was done by the multifractal spectra, $f(\alpha)-\alpha$. It is found that the multifractal spectrum shape is left hook-like (that is difference of height interval of the multifractal spectrum, $\Delta f = f(\alpha_{\min}) - f(\alpha_{\max}) > 0$). The results indicate that the surfaces having greater roughness give rise the wider multifractal spectrum width($\Delta\alpha$) and the greater Δf, thus, the nonuniformity of the height probabilities becomes larger. It indicates that the multifractality of the films becomes more pronounced at the higher thickness.

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1. Introduction

Silver is a lustrous white, ductile, malleable metallic element. It has many practical applications in different fields including, electronic [1], optical [2], catalysis [3], sensors [4] superhydrophobicity [5], solar cells [6], medicine [7], surface-enhanced Raman scattering [8] and cloud seeding [9]. The functional properties of silver surface are related to its roughness. For example, in superhydrophobicity and surface-enhanced Raman scattering application contact angle and signal enhancement factor directly depend on the nanoscale morphology [5,10]. Identification of the structures and properties of monolayer and multilayer thin films are very important, because they show some interesting and unique physical properties which is quite different from those made by bulk materials, therefore they have potential applications in different magnetic, electronic and optical devices [6,11–13]. Surface/interface morphology influence on electrical, optical, mechanical, tribological and magnetic properties of thin film forcefully and plays important roles in manufacture of nanostructured materials in a controlled way in order to gain desired properties.

In the last decade, there has been an increasing interest for understanding the film growth mechanisms, and thus its dependency on the parameters ruling on the deposition process, from both practical and theoretical. The investigation of surface evolution of the thin film growth can offer an insight on how basic kinetics which controls the film growth [14,15].

Most techniques that fabricate thin films are usually based on vacuum including molecular beam epitaxy, sputtering and evaporation. Electrochemical deposition as an alternative deposition technique is important because of its affordability, flexibility and simplicity in experimental setup. The mechanism of electrodeposition is complicated, because it is a non-equilibrium growth process and involves a large number of variables that influences the process including cation diffusion, pH and concentration of electrolyte and substrate type. Evolution of thin films under a non-equilibrium condition produces complex surfaces which are not sufficiently specified by conventional methods. The surface growth is expected to develop with structure of self-similar or self-affine [16].

Atomic force microscopy (AFM) technique provides a real space imaging. It has been broadly used to study the surface morphology of thin films and gives the surface morphological characteristics

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with high spatial resolution. AFM can also be employed to perform a quantitative study of the interface width using dynamic scaling method. Several research groups [17–22] have made efforts to understand the surface roughness and growth mechanism.

Fractal and multifractal concepts are useful tools for describing the surface morphology and its complexity. A fractal system is described by only one scaling exponent. However in a multifractal system which is the generalization of the fractal system, just a single exponent (depending on the position in the structure) is not enough to describe its dynamic. Instead, it is necessary to use continuous spectrum of exponents (known as singularity spectrum). Multifractal spectrum is used to identify the surface roughness as well as to determine the shape of valleys and peaks in various rough surfaces [23]. Multifractal analysis of a film surface gives more detailed information of the surface compared with fractal analysis [24] and is very useful in understanding the relationship between structure and properties of material [25]. It has been used in different studies because of high precision, low computation time and easy implementation.

In this work, we studied multifractal analysis and kinetic roughening of the AFM images of Ag films with various thickness prepared by electrodeposition technique. Our analysis indicates that the self-similar and multifractal characteristics as well as anomalous scaling exist in the Ag film morphologies. An attempt has been done to discover the dependency of these features on the films thickness.

1.1. Scaling theory

Correlation of film thickness and surfaces roughness is an effective way to study the dynamic scaling [18]. A quantitative description of surfaces should be performed to understand the effect of thickness on film roughness. Dynamic scaling theory has studied scaling analysis of surface evolution. This model is based on calculation of $W(l,t)$, which is the root mean square (RMS) height of the surface and determined as follow:

$$W(l,t) = \langle \mathrm{h}(t)^2 - \langle \mathrm{h}(t) \rangle^2 \rangle^{1/2} \tag{1}$$

Where t is the film thickness, l is the length-scale over which W is measured, h is the surface height, measured by AFM and $< h >$ is their overall average over length scale, l. The value of W depends on the area of the surface which is studied. For instance, if W is calculated on the length scale which is selected small enough to be at the top of the individual nucleus of growth, its value will be much smaller than which is calculated for a larger surface region. In this analysis, as the length scale, used to calculate W, increases, the value of W also increases up to a certain value in $l = l_c$, where l_c gives the maximum range of lateral correlations. If then, the region is taken larger than l_c, the W_{sat} value of W remains unchanged and is shown as W_{sat}. Understanding of microscopic mechanism of electrodeposition growth can be provided by this analysis.

Normal dynamic scaling which is also known as Family–Vicsek scaling [26] represented by expression as:

$$w(l,t) = l^H f\left(t/l^{H/\beta}\right) \tag{2}$$

where H and β are Hurest and growth exponent, respectively. The Hurest and growth exponents respectively indicate the saturated interface roughness, and the time-dependent dynamics of the rough surface. The scaling function behaves as:

$$f\left(\mathrm{t}/l^{H/\beta}\right) \sim \begin{cases} constant & t/l^{H/\beta} \gg 1 \\ \left(t/l^{H/\beta}\right)^{\beta} & t/l^{H/\beta} \ll 1 \end{cases} \tag{3}$$

Moreover l_c scales as $t^{\beta/H}$

The normal scaling is used to investigate the influence of additives on two dimensional growth of nucleation [27,28]. Also, it has been studied in Eden cell growth model [26,29], unrestricted and restricted ballistic deposition models with one and two dimensional substrates [26,30] and models defined by continuum equations [30–34].

A more complex type of scaling for kinetic roughening is predicted by Schwarzacher et al. [35,36] and called anomalous dynamic scaling. It represents by the following relation:

$$w(l,t) = l^H t^{\beta_{loc}} f\left(t/l^{H/\beta}\right) \tag{4}$$

where β_{loc} is known as the local exponent. It indicates effect of local influence on the growth exponent β. In this type, l_c scales as $t^{\beta/H}$ and relates to W_{sat} by a power law of $W_{sat} \sim l_c^{\alpha^\circ}$. α° is known as the roughness exponent and is given by:

$$\alpha^\circ = H\left(1 + \frac{\beta_{loc}}{\beta}\right) \tag{5}$$

A number of (1 + 1)-dimensional systems including cracks in wood [37] and stone [38], and (2 + 1)-dimensional systems such as insulator, semiconductor and metal films which are grown by vacuum based deposition techniques [39–41], etched surfaces [42] and sputtered films [43,44] show anomalous dynamic scaling. (1 + 1) and (2 + 1)-dimensional electrodeposited films were also studied based on anomalous scaling. It is interested because of the possibility of access to a variety of experimental behavior by changing growth conditions [45].

1.2. Multifractal analysis

In the most of the cases the scaling behavior may changes from point to point. This situation is explained by multifractal formalism. The most popular multifractal analysis is used in the spectrum of singularities $f(\alpha)$ multifractal method. It is briefly described as follow.

In multifractal analysis, the box-counting method is applied. In this method, an AFM image is divided into $N(\varepsilon)$ boxes with size of ε, which $\varepsilon = l/L \leq 1$. l and L are length scale and upper bound on l, respectively. P_{ij} is an average deposition probability which is measured for the film in the box(i,j) and is determined as:

$$P_{ij}(\varepsilon) = \frac{h_{ij}}{\sum h_{ij}} \tag{6}$$

where h_{ij} is the average height of the box (i,j) with size of ε measured from the data planes which have similar depths for the Ag deposited film and substrates, so that $\sum P_{ij} = 1 \langle REFNUMLINK \rangle$[46]. For multifractal measures, for the box (i,j) in scale ε, P_{ij} scales as:

$$P_{ij} \propto \varepsilon^{\alpha} \tag{7}$$

where the exponent α is singularity of the probability subsets. It is also called Lipschitz–Holder exponent. The exponent α, is used for labeling boxes that cover the set supporting a measurement. It represents monofractality, if all boxes have the similar exponent α. However it indicates multifractal if different boxes scale have different exponents of α, which corresponds to various of measured strengths.

The number of boxes with size ofε that have the sameα exponent is given by

$$N_{\alpha}(\varepsilon) \propto \varepsilon^{-f(\alpha)} \tag{8}$$

where $f(\alpha)$ is singularity spectrum and is a continuous function of α. It is also determined the fractal dimension of the subset of the measure that has α exponent. The values of the α and $f(\alpha)$ permit a quantitative assessment of the degree of reaction probability distri-

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