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X-ray reflectivity and scanning-tunneling-microscopy study of surface roughness scaling of molybdenum films

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Abstract. – An X-ray reflectivity (XR) study of the dynamic evolution of the film surface was carried out for molybdenum (Mo) sputter-deposited onto silicon substrates. The Mo-air interface width grows with time, and exhibits a power law behavior. The growth exponent β is found to be 0.42. The time-invariant self-affine behavior on the short-range scale has also been observed, and is consistent with the dynamic scaling theory. The roughness exponent α is found to be 0.89 ± 0.05. Scanning tunneling microscopy (STM) was also used to characterize the surface and showed good agreement with the XR measurements.

Thin films have been applied in a wide variety of areas. The quality of thin films is determined by the interfacial roughness. Characterization and understanding of the microstructures of interface is then of central importance, since the quality of the interface influences considerably the properties and device performance of the thin films.

Many researches have been performed on film growth process. The nonequilibrium dynamics of interface growth has attracted much attention recently [1]. A lot of theoretical effort has been devoted to it, and the generally accepted result is that the surface morphology and dynamics of a growing interface exhibit simple dynamics scaling behavior [1]. In the scaling regime, the growth front is proposed to have a self-affine fractal surface. According to the dynamic scaling approach, the interface width W at time t for a measured surface size L is expected to be

$$W(L,t) \approx L^{\alpha} f(t/L^{\alpha/\beta}) \tag{1}$$

with $f(x) \approx x^{\beta}$ for $x \ll 1$ and $f(x) = \text{const}$ for $x \gg 1$; α is referred to as the roughness

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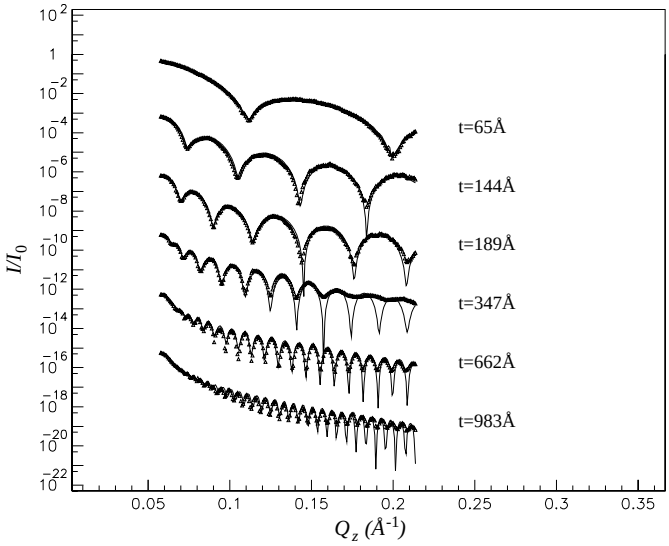

Fig. 1. – Specular X-ray reflectivity at 0.6 Pa Ar pressure and room temperature for Mo films. The solid lines are the fit of the experimental data (triangle).

exponent describing the surface roughness of the self-affine fractal surface and β is the growth exponent related to the growth process.

According to KPZ [2], the exponents $\alpha = 1/2$ and $\beta = 1/3$ in $d = 1 + 1$ dimensions. This conclusion is consistent with the results by computational studies. For $2 + 1$ dimensions, the theoretical values of the exponents varied in the different models [1]-[3]. Although many theoretical efforts have been carried out, there are few experimental studies of the growth dynamics which cover both scaling laws related with the long-range (β) and the short-range (α) regimes. So it is practical and valuable to make sufficient experimental investigations of the exponents to determine if they are really universal [4].

In the previous experimental studies of the scaling power law, He *et al.* [5] obtained $\beta = 0.22$, $\alpha = 0.79$ for Fe on Fe(001) by MBE; Ernst *et al.* [6] observed $\alpha = 1$ for deposition at 200 K and 160 K, while β amounts to 0.56 at 200 K and 0.26 at 160 K for Cu on Cu(100) by MBE; You *et al.* [7] obtained $\alpha = 0.42$ and $\beta = 0.4$ at 300 K, 0.42 at 220 K for Au films sputter-deposited onto Si(111). Thompson *et al.* [8] observed $\alpha = 0.7$ and $\beta = 0.26$ for vapor-deposited Ag films onto Si substrates. Gollins *et al.* [9] found $1 > \alpha > 0.9$ and $1 > \beta > 0.6$ for the growth of plasma polymer films at different deposition rates. The diversity of these measured values indicates that the thin-film dynamic growth is quite complicated for different material systems or with different surface diffusion mechanisms [10] and it also shows there is no coherent result reached.

The purpose of this study is to determine the exponents experimentally and to compare them with theoretical models. To our knowledge, there have been no experimental studies of the scaling power law in Mo, a material that is often used as the component of optical multilayer. We report measurements of α and β for Mo sputter-deposited thin films by means of X-ray reflectivity (XR) and scanning tunneling microscopy (STM), and discuss the factors that have a large effect on the two exponents.

Mo films with different thickness ranging from 50 to 1000 Å were deposited in a dc magnetron sputter chamber with a base pressure 10^{-4} Pa, and the Ar pressure was 0.6 Pa

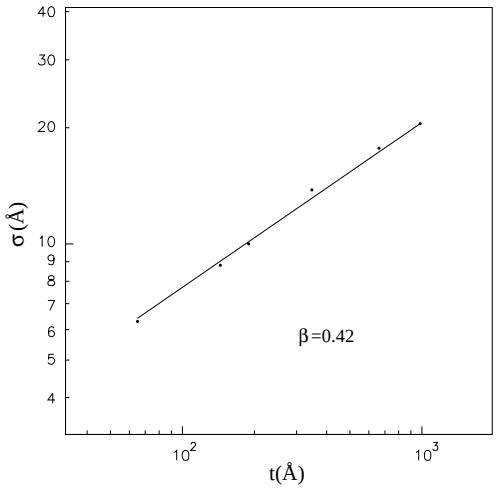

Fig. 2

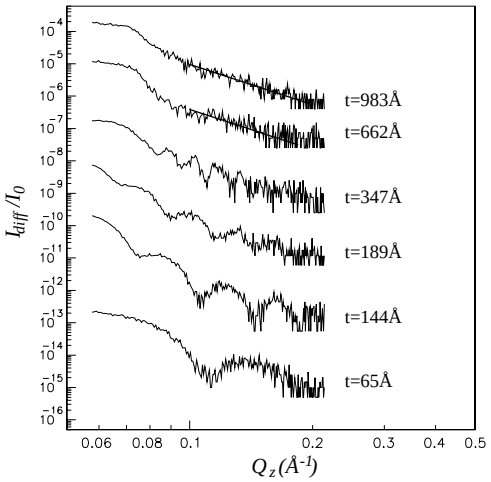

Fig. 3

Fig. 2. – The surface width *vs.* film thickness is plotted on a log-log scale. The solid line is the power law fit with $\beta = 0.42$.

Fig. 3. – The off-specular diffuse reflectivity recorded at a 0.1° offset from the specular condition for the six samples. The solid lines are the linear fits based on the asymptotic form.

during deposition at room temperature. The substrates were silicon(111) single-crystal wafers offered by Universität München with roughness of $\sim$ 3 Å. The X-ray measurements were performed at a conventional two-circle X-ray diffractometer, the source is a 18 kW Rigaku Cu X-ray generator. STM images of the Mo thin film surfaces were obtained with a grid density of 128×128, operating at 1 V bias voltage and 300 pA tunneling current.

Specular and off-specular X-ray reflection measurements were made. The true specular signals were obtained by subtracting the measured off-specular scattering contributions from the raw specular reflection data, they are shown in fig. 1. The oscillations shown in fig. 1 are caused by the interference between the X-ray reflection from the surface (air-Mo) and the interface (Mo-substrate). The oscillation frequency increases with the thickness of the sample, and the interference fringes decay with increasing thickness due to the increasing surface roughness of films. The theoretical fit was calculated by the optical X-ray dynamical method [11] where the Debye-Waller–like factor was considered for roughness interfaces. The surface roughness as one of the fitting parameters was obtained and shown in fig. 2 as a function of layer thickness; here, we assumed that the deposition time is directly proportional to the thickness. The exponent β is obtained from fig. 2 with the power law scaling equation (1), the value of β is 0.42. STM images with different scan regions of 500 Å × 500 Å, 1000 Å × 1000 Å, 2000 Å × 2000 Å, 4000 Å × 4000 Å were recorded on 347 Å, 662 Å, 983 Å three samples. The obtained surface roughness of the three samples is 13.92 Å, 18.06 Å and 23.02 Å, respectively. They are in good agreement with the results of 13.8 Å, 17.7 Å, and 20.5 Å obtained by XR.

According to Sinha *et al.* [12], in Distorted-Wave Born Approximation (DWBA), the differential cross-section for diffuse scattering can be expressed as

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = L_x L_y \frac{|k_0^2(1-n^2)|^2}{16\pi^2} |T(K_1)|^2 |T(K_2)|^2 S(q_t)\,,$$

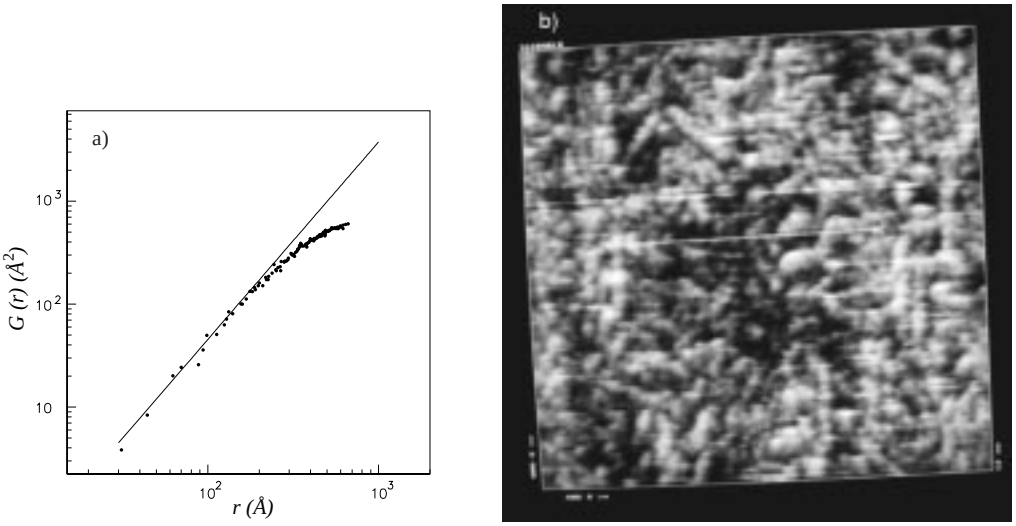

Fig. 4. – (a) $G(r)$ calculated from the STM image of a 662 Å Mo film. The solid line is a fit according to the power law equation (3). (b) The STM image recorded on the 662 Å Mo film.

$$S(q_t) = \frac{\exp[-[(q_z^t)^2 + (q_z^{t*})^2]W^2/2]}{|q_z^t|^2} \int_0^\infty (e^{|q_z^t|^2 C(X)} - 1)\cos(q_x X)\mathrm{d}X\,. \tag{2}$$

Based on eq. (2), Thompson *et al.* [8] gave an asymptotic form about the diffuse cross-section of a self-affine surface with no cut-off ($\xi \to \infty, q_x = 0$) as $I(q_z) \propto W^{-2/\alpha} q_z^{-(3+1/\alpha)}$. Figure 3 is a log-log scale plot of 0.1° offset diffuse reflectivity for the six samples. α is obtained as 0.84 from the fit of the linear portions of the data by the asymptotic form for 662 Å and 983 Å two samples.

To compare with the result of the XR measurement, the mean-square surface fluctuation function $G(r)$ was calculated from STM images. $G(r)$, which is generally used to describe a growing interface assumed as an isotropic Gaussian distribution, has the form for a self-affine surface as [1],

$$G(r) \sim r^{2\alpha}\,, \tag{3}$$

when $r \ll L$, where $r = (x^2 + y^2)^{1/2}$. Figure 4(a) displays the function $G(r)$ *vs.* r for a 662 Å thick sample from the STM image shown in fig. 4(b). The solid line is the fit based on eq. (3) at small r. α was obtained as 0.94. It is a little larger than that obtained from the X-ray data. The α value from the X-ray data is based on the model of single-interface analysis, it is the lower limit to the range of permissible value [8]. In addition, there is an appropriate range of q_z values for the best fit of the linear portion of the X-ray data; the range is within as high as possible q_z values, while the intensity is still real diffuse scattering. This is the reason why our α value for XR is lower than that from STM. A detailed discussion will be published later.

On the condition of sputtering-deposited Mo thin films with Ar pressure 0.6 Pa and at room temperature, we obtained α as 0.89 ± 0.05 and β as 0.42 by means of XR and STM. This result is in the range of $0.2 \leq \alpha \leq 1$, $0.2 \leq \beta \leq 0.56$[13,3–10] given by different theortical models and experimental results. The α value coincides with ref. [5] for the Fe film, with ref. [6] for the Cu film and with ref. [8] for the Ag film. The β value is in agreement with ref. [7] that uses

sputtering-deposition technique, but is larger than the one in ref. [5], [6] and [8], where MBE and vapor deposition are the growth process.

The value $\alpha = 0.89$ is larger than the KPZ value but more consistent with the conservative growth models [14]. Since the substrate temperature in our experiment is high and the metal thin film lacks local chemical bonds, the horizontal mobility of deposited particles is increased, and the surface diffusion is the dominant relaxation process. So the α value we measured is more close to the conservative growth model. Another exponent β is 0.42, which is higher than 0.25 which was predicted by KPZ equation for the $d = 2 + 1$ system, but it coincides with the result of ref. [7] for the sputter deposition process. β is related to the time-dependent long-range feature, *i.e.* it is related to the growth process, so compared with α, β is more dependent on film deposition techniques. It might reflect some inner relations between the β value and deposition techniques during the nonequilibrium dynamics of interface growth.

Most recent experiments on metal thin films deposited with different techniques [5]-[8] gave the α value 0.7–1. In our recent experiments for studying the relation between Ar pressure and α value, we obtained that the Ar pressure in sputtering deposition had a great influence on the α value. The Ar pressure affects the momenta of the deposited particles, but the latter plays an important role in the competition between the horizontal mobility and the deposition rate of the particles. The competition between the two factors might play a main role in the surface morphology. In our recent experimental result [15], higher Ar pressure (1 Pa) gives rise to a lower α value which deviates from the range 0.7–1 for the metal thin films. The detailed discussion will be presented in a later publication.

In conclusion, we observed both time-invariant and time-dependent scaling power laws of the dynamic growth of Mo thin films in a sputtering-deposition (2+1)-dimensional system, we obtained the related exponents α and β, and the factors which have a strong effect on α and β were discussed.

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