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Dynamic scaling in sputter grown tungsten thin films

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Abstract

The evolution of a tungsten thin film grown by magnetron sputtering was studied using a dynamic scaling approach. Film growth was followed in-situ and in real-time by monitoring both the specular and the diffuse X-ray scattered intensities as a function of the time of deposition. The analysis of the scattering data allowed us to determine the two Power Spectral Density (PSD) functions, which describe the thin film topography. The time-dependent PSD-function, which describes the dynamic of the external film surface, is found to obey a universal scaling form, which characterizes the thin film growth. The data collapse of these PSDs into a single master curve was achieved using scaling exponents $\alpha = 0.18 \pm 0.02$ and $\beta = 0.06 \pm 0.01$. In addition, by analyzing the temporal variation of the roughness conformity, it has been demonstrated that the replication factor decreases exponentially with increasing film thickness and spatial frequency. Hence, for a 25 nm thick film the vertical correlation disappears for spatial frequencies p greater than 3.6 μm^{-1}.

Keywords: X-ray scattering; Surface roughness; Dynamic scaling

1. Introduction

The surface of a film growing under non-equilibrium conditions often develops in agreement with the concept of dynamical scaling [1,2]. In this concept, several attributes, known as scaling exponents, can be used as the signatures in space and time of highly complex film growth processes. By comparing the experimental scaling exponents with the theoretical predictions one should be able to recognize which type of differential equation best describes the film growth and, hence, classify various growth processes within universality classes [2]. Nowadays various experimental techniques are used to determine the scaling exponents (see, e.g. in Ref. [3] and references therein). Among them the X-ray scattering (XRS) technique is a unique method to study the evolution of roughness of a growing film, in-situ, in real-time, and in vacuum environment. Moreover, it is a way of investigating buried interfaces and the correlation between the film and the substrate relief.

This paper describes results from in-situ real-time investigations of the roughness evolution during the growth of tungsten films using grazing incidence X-ray scattering and a novel experimental apparatus available on the BM5 beamline at the ESRF. We show that, by recording in-situ a single scattering diagram for a grazing angle of a probe beam exceeding the critical angle of total external reflection, it is possible to uniquely determine the two PSD-functions describing the micro-topography of the external surface and the film-substrate roughness conformity. Using this approach, the temporal evolution of the roughness of a sputter-deposited tungsten film has been studied and the thickness-dependent power spectral density (PSD) has been determined to obey a universal scaling form [1]. In accordance with the scaling model [1,2] the 1D PSD-function characterizing the external film topography, may be collapsed into a single master curve provided the scaling exponents are properly chosen.

2. Film growth, scattering measurements and determination of the PSD-functions

A super-polished silicon substrate flattened to $\lambda/10$ ($\lambda = 632.8$ nm) has been processed and a W layer has been progressively grown onto it to reach a final thickness of about 25 nm. The experiment was performed at room temperature and the sputter time was adjusted to observe a significant

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variation of the surface roughness while increasing the processing time. Before process the background pressure in the sputter chamber was below 3.6×10^{-7} hPa. Then, magnetron sputtering was conducted in pure (99.999%) Ar gas at a working pressure of 13.3 10^{-3} hPa. The film growth was realized using a bias voltage of 370 V and a total target current of 30 mA. The pressure set point was achieved by fixing the gas flow rate at a value of 3 sccm.

XRS measurements were performed by setting the X-ray energy at 17.5 keV with a double-crystal Si (1 1 1) monochromator and fixing the sample in stationary position. The divergence in the vertical direction was $3 \cdot 10^{-6}$ rad and the spectral purity $\Delta E/E$ of the order of 10^{-4}. The angle of the probe beam with the sample was set to $\theta_0 = 0.5°$, i.e., out of the total external reflection. For comparison, the critical angle is 0.25° for bulk tungsten at this wavelength. The X-ray detection was performed using a combination of two detectors operating simultaneously: the first one, consisting of an ionization chamber, monitored the total intensity of the reflected beam and the second one, composed of a cryogenically cooled CCD camera (1024 × 256 pixels, equivalent pixel size: 150 μm), collected the scattered beam. Here the X-ray photons are converted into visible light by a phosphor screen coupled with the entrance window of an image intensifier. Finally, the intensifier output is imaged onto the CCD detector by a lens system. The reflectivity variation as a function of the deposition time, which was recorded at the same time as the XRS diagrams, was analyzed to infer the tungsten deposition rate (12.3 pm/s) and the film density (18 g/cm^3). The scattering diagrams were measured while growing the film and at 5 s time intervals.

The bi-directional scattering diagram $\Phi(\theta,\varphi) = (1/W_{\mathrm{inc}})(\mathrm{d}W_{\mathrm{scatt}}/\mathrm{d}\Omega)$, which describes the power of radiation $\mathrm{d}W_{\mathrm{scat}}$ scattered from a rough surface within a small solid angle $\mathrm{d}\Omega$ and normalized to the power of the incident beam W_{inc}, is a function of two scattering angles: the angle θ referencing in the scattering plane (vertical in our case) and the azimuth angle φ referencing in the plane of the sample surface. During our experiment the vertical beam size was set to 0.2 mm and the horizontal size to 5 mm, to favor a high intensity onto the detector. As the scattering diagram is very narrow in the azimuth (horizontal) plane, the scattered intensity was integrated in this direction as $\Pi(\theta) = \int \Phi(\theta,\varphi)\mathrm{d}\varphi$. An example of scattering measurements processed in this way is shown in Fig. 1 (solid line). The sharp minimum observed on this curve, at the position of the specular peak, is due to the presence of the beamstop placed in front of the CCD detector to prevent it from saturation.

In the frame of the first-order scalar perturbation theory the integrated scattering diagram from a rough isotropic film is written in the following form:

$$\Pi(\theta,h) = \frac{1}{W_{\mathrm{inc}}}\frac{\mathrm{d}W_{\mathrm{scat}}}{\mathrm{d}\theta} = \frac{k^3}{16\pi\sin(\theta_0)}\left[A_{\mathrm{f}}(\theta_0,\theta,h)\cdot \mathrm{PSD}_{\mathrm{ff}}(p,h) + A_{\mathrm{s}}(\theta_0,\theta,h)\cdot \mathrm{PSD}_{\mathrm{ss}}(p) + A_{\mathrm{sf}}(\theta_0,\theta,h)\cdot \mathrm{PSD}_{\mathrm{sf}}(p,h)\right]; p = \frac{1}{\lambda}|\cos\theta_0 - \cos\theta|; k = \frac{2\pi}{\lambda} \quad (1)$$

where W_{inc} and $\mathrm{d}W_{\mathrm{scat}}$ are, respectively, the radiation power of the incoming beam impinging under an angle θ_0 and the radiation power of the beam scattered within an angular interval $\mathrm{d}\theta$ under an angle θ with respect to the sample surface.

The electrodynamic factors A_{f}, A_{s}, and A_{sf} have the following expressions:

$$\begin{aligned} A_{\mathrm{f}} &= |(1-\varepsilon_{\mathrm{f}})[1 + r(\theta_0,h)][1 + r(\theta,h)]|^2 \\ A_{\mathrm{s}} &= |(\varepsilon_{\mathrm{f}}-\varepsilon_{\mathrm{s}})t(\theta_0,h)t(\theta,h)|^2 \\ A_{\mathrm{sf}} &= 2\mathrm{Re}\{(1-\varepsilon_{\mathrm{f}})(\varepsilon_{\mathrm{f}}-\varepsilon_{\mathrm{s}})^*[1 + r(\theta_0,h)][1 + r(\theta,h)]t^*(\theta_0,h)t^*(\theta,h)\} \end{aligned} \quad (2)$$

The dielectric constants of the substrate and the film are noted as ε_{s} and ε_{f}. The amplitude reflectance and transmittance of a perfectly smooth film of thickness h are noted $r(\theta,h)$ and $t(\theta,h)$, respectively. The scattering properties of a rough film are thus characterized entirely by three 1D PSD-functions, $\mathrm{PSD}_{\mathrm{ss}}(p)$ and $\mathrm{PSD}_{\mathrm{ff}}(p,h)$, which describe the roughness of the substrate and the roughness of the external film surface, and $\mathrm{PSD}_{\mathrm{sf}}(p,h)$, which determine the statistical correlation (conformity) between film and substrate roughnesses: $\mathrm{PSD}_{ij}(p) = 4\int\langle\zeta_i(\vec{r})\zeta_j(0)\rangle\cos(p\rho)\mathrm{d}\rho$ with $i,j = \{s,f\}$. The stochastic functions $\zeta_s(\vec{r})$ and $\zeta_f(\vec{r})$ describe the substrate and the film relief, respectively, and $\vec{r}$ is the vector lying in the plane of the surface. The angular brackets denote an ensemble averaging and p is the spatial frequency.

In a recent work [4], a new approach for the characterization of growing thin films based on the analysis of two independent sets of scattering data acquired at different grazing angles of incidence has been proposed. Such analysis, using the same physical model of a film, provides convincing argument about the accuracy of the procedure. However, it requires a particularly demanding experimental protocol as two "identical experiments" differing only by the angle of the probe beam must be performed. Practically, all causes of instabilities that may occur during a temporal scan (e.g. X-ray beam and/or sputtering stability) have to be reduced as much as possible. Moreover, it requires a set of samples with statistically identical properties.

A careful analysis of formula (1) shows some interesting features which allows us to improve the experimental method

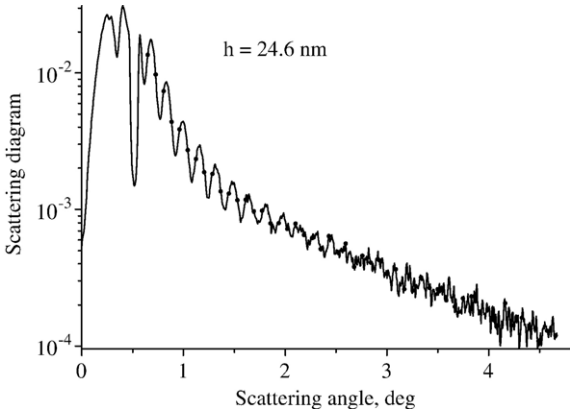

Fig. 1. XRS diagram (circles) measured in-situ and in real-time on a tungsten thin film 24.6 nm thick (solid curve). The red dots denote the points where the electrodynamics factor A_{sf} appearing in formula (1) equal zero.