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X-ray reflection and diffuse scattering from sputtered gold films

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Abstract

Gold films were prepared by DC sputtering on quartz glass substrates under two different conditions, once in argon at 3×10^{-3} mbar (Au1-films), and once in residual air at 0.3 mbar (Au2-films). Specular X-ray reflection showed that the surface of the Au1-films was as smooth as that of the substrate, whereas the Au2-films were distinctly rougher. The diffuse scattering could be measured with a laboratory equipment by recording rocking curves and detector scans. The application of existing theories to the diffuse scattering data showed that slight modifications of the given equations were necessary. With these modifications the experimental data could be fitted very well, and the height–height correlation functions of the surfaces of the films and of the substrate were determined. In the Au1-films the surfaces of the film and the substrate are perfectly correlated, whereas in the Au2-films no cross correlation is found.

Keywords: Gold films; X-ray reflection; Diffuse scattering

1. Introduction

Specular reflection from thin films provides information about their thickness, density and mean-square roughness, more generally spoken, about the structure perpendicular to the surface (z-direction) [1]. The diffuse scattering is measured under the nonspecular condition where the angle between the diffracted wave and the sample surface, β, is different from the angle of incidence, α. It yields information about structural features along the surface (x, y), such as the height–height correlation function $c(x,y)$ of a rough surface. In the present work we study the feasibility of diffuse diffraction work by means of a laboratory setup with a sealed X-ray tube. The application of the diffuse scattering theories by Sinha et al. [2], Sinha [3] and Pynn [4] to sputtered Au films is demonstrated. Two representative examples were selected from Ref. [5], where the experimental methods and the data reduction procedures are described in detail.

2. Experimental

The films were deposited on quartz glass substrates by DC sputtering, applying two different sputtering conditions. Au1-films were prepared in a magnetron DC sputtering device under a high-purity argon atmosphere at 3×10^{-3} mbar. Au2-films were DC sputtered in residual air at 0.3 mbar.

The measurements of the specular reflectivity and the diffuse scattering were performed with a Siemens D 500 diffractometer, equipped with a

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cutting edge above the sample surface, a detector slit of 67 μm, and a secondary monochromator to select Cu K_α radiation.

3. Results and discussion

3.1. Specular reflection

Fig. 1 shows the experimental reflectivities of the Au-films (dots). From fitting theoretical expressions (lines) using the well established optical matrix method (see e.g. [1]) the parameters in Table 1 were derived.

The Au1-film appears rather perfect. Its reflectivitiy curve exhibits pronounced oscillations. Its roughness, $\sigma_F = 7.7$ Å, is about the same as that of the quartz glass substrate, $\sigma_S = 7.0$ Å, and its density, $\rho_F = 19.0\,\mathrm{g/cm^3}$ is only slightly lower than the density of bulk gold, $\rho_{Au} = 19.25\,\mathrm{g/cm^3}$.

In contrast to this, the Au2-film is obviously much less perfect. The rather large roughness, $\sigma_F = 23.0$ Å, causes a rapid damping of the oscillations. The low apparent density, $\rho_F = 15.5\,\mathrm{g/cm^3}$, indicates that the film is not homogeneous. The introduction of nitrogen and oxygen atoms during the sputtering process reduces the lateral mobility of the Au atoms in the film plane, and thus enhances the tendency for island formation.

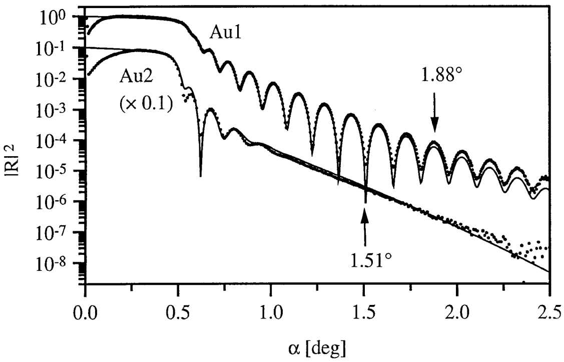

Fig. 1. Specular reflection with gold films Au1 and Au2: (●) experimental, (—) theoretical fit. The curves for Au2 are shifted in the plot.

Table 1
Parameters derived from specular reflectivity

	d (Å)	σ (Å)	ρ (g/cm^3)
Substrate	—	7.0	2.20
Au1-film	282	7.7	19.0
Au2-film	235	23.0	15.5

3.2. Diffuse scattering

Fig. 2 (dots) shows a detector scan of the Au1-film at $\alpha = 1.88°$, where the specular reflection in Fig. 1 exhibits a peak. The lines show an attempt to fit the experimental data, using theories from literature: the dashed line according to Sinha (Eqs. (13) and (14) in Ref. [3]) and the solid line according to Pynn (Eqs. (25) and (29) in Ref. [4]). The slit geometry of the measurements was taken into account by integrating the equations over the component q_y of the scattering vector $\boldsymbol{q}$ perpendicular to the scattering plane x–z. This corresponds to a transition from the two-dimensional correlation function $c(x, y)$, to the one-dimensional case $c(x)$ in the equations. Obviously, it is not possible to obtain good agreement between fit and experimental data, in particular with respect to the phase of the oscillations.

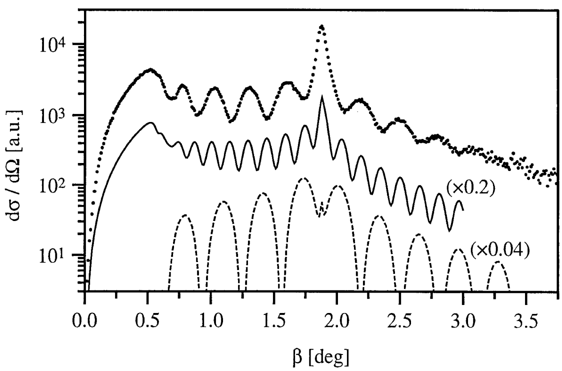

Fig. 2. Detector scan with Au1-film at $\alpha = 1.88°$: (●) experimental, (—) theoretical fit according to Pynn, (— — —) according to Sinha. The fit curves are shifted in the plot.

Therefore the equations from Refs. [3] and [4] were modified in the present work. With the applied modifications the contributions of the free surface of the film (F), the substrate-film interface (S), and the cross-correlation between the two surfaces (C), respectively, to the total scattering cross-section $(\mathrm{d}\sigma/\mathrm{d}\Omega)$ are written as follows

According to Sinha:

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)^{\mathrm{S}}_{\mathrm{S}} = k\,B^{\mathrm{S}}_{\mathrm{S}}\frac{\exp(-\mathrm{Re}[q^2_{z,\mathrm{S}}]\sigma^2_{\mathrm{S}})}{|q_{z,\mathrm{S}}|^2} \times \int \mathrm{d}x\cos(q_x x)\{\exp[|q_{z,\mathrm{S}}|^2 c_{\mathrm{S}}(x)]-1\}, \tag{1a}$$

where $B^{\mathrm{S}}_{\mathrm{S}} = \Delta\rho^2_{\mathrm{S}}|T_\alpha|^2|T_\beta|^2$.

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)^{\mathrm{S}}_{\mathrm{F}} = k\,B^{\mathrm{S}}_{\mathrm{F}}\frac{\exp(-\mathrm{Re}[q^2_{z,\mathrm{F}}]\sigma^2_{\mathrm{F}})}{|q_{z,\mathrm{F}}|^2} \times \int \mathrm{d}x\cos(q_x x)\{\exp[|q_{z,\mathrm{F}}|^2 c_{\mathrm{F}}(x)]-1\}, \tag{1b}$$

where $B^{\mathrm{S}}_{\mathrm{F}} = \Delta\rho^2_{\mathrm{F}}|t_{\alpha,\mathrm{F}}|^2|t_{\beta,\mathrm{F}}|^2$.

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)^{\mathrm{S}}_{\mathrm{C}} = k\,B^{\mathrm{S}}_{\mathrm{C}}\frac{\exp(-\mathrm{Re}[q^2_{z,\mathrm{F}}][\sigma^2_{\mathrm{F}}+\sigma^2_{\mathrm{S}}]/2)}{|q_{z,\mathrm{F}}|^2} \times \int \mathrm{d}x\cos(q_x x)\{\exp[|q_{z,\mathrm{F}}|^2 c_{\mathrm{C}}(x)]-1\}, \tag{1c}$$

where $B^{\mathrm{S}}_{\mathrm{C}} = 2\,\Delta\rho_{\mathrm{F}}\,\Delta\rho_{\mathrm{S}}|T_\alpha|^2|T_\beta|^2\cos(\mathrm{Re}[q_{z,\mathrm{F}}]d)$.

The calibration constant k contains all q-independent quantities. $\Delta\rho$ is the electron density difference, $\boldsymbol{q}$ is the wave vector transfer, $t_{\alpha,\mathrm{F}}$ and $t_{\beta,\mathrm{F}}$ are the transmission coefficients at the film surface at the angle α of the incoming and the angle β of the reflected wave, T_α and T_β are the transmission coefficients of the entire film–substrate system, and d is the thickness of the film. The transmission coefficients were calculated for rough surfaces.

According to Pynn:

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)^{\mathrm{P}}_{\mathrm{S}} = \left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)^{\mathrm{S}}_{\mathrm{S}}, \tag{2a}$$

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)^{\mathrm{P}}_{\mathrm{F}} = k\,\Delta\rho^2_{\mathrm{F}}\sum_{n=1}^{6}(-1)^{n+1}F_n \times \frac{\exp[-(f^2_n+g^2_n)\sigma_{\mathrm{F}}/2]}{f_n\;g_n}\int \mathrm{d}x\cos(q_x x) \times\{\exp[(-1)^{n+1}f_n g_n c_{\mathrm{F}}(x)]-1\}. \tag{2b}$$

The F_n, f_n and g_n are the same as defined in Ref. [4] (Eqs. (26) and (27)).

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)^{\mathrm{P}}_{\mathrm{C}} = k\,B^{\mathrm{P}}_{\mathrm{C}}\frac{\exp[-(q^2_z\sigma^2_{\mathrm{F}}+\mathrm{Re}[q^2_{z,\mathrm{S}}]\sigma^2_{\mathrm{S}})/2]}{q_z\,|q_{z,\mathrm{S}}|} \times \int \mathrm{d}x\cos(q_x x)\{\exp[q_z|q_{z,\mathrm{S}}|\,c_{\mathrm{C}}(x)]-1\}, \tag{2c}$$

where $B^{\mathrm{P}}_{\mathrm{C}} = 2\,\Delta\rho_{\mathrm{F}}\,\Delta\rho_{\mathrm{S}}|T_\alpha||T_\beta|\,\mathrm{Re}[\exp(\mathrm{i}\,q_{z,\mathrm{F}}\,d)]$.

The modifications to the equations given in Refs. [3,4] are: *Substrate*, *Eq.* (*1a*): the component q_z of the wave vector transfer is substituted by the corresponding value in the substrate, $q_{z,\mathrm{S}}$. Then $(\mathrm{d}\sigma/\mathrm{d}\Omega)^{\mathrm{S}}_{\mathrm{S}} = (\mathrm{d}\sigma/\mathrm{d}\Omega)^{\mathrm{P}}_{\mathrm{S}}$. *Film*, *Eq.* (*1b*): $q_{z,\mathrm{F}}$ instead of q_z and $t_{\alpha,\mathrm{F}}, t_{\beta,\mathrm{F}}$ instead of T_α, T_β. *Cross correlation*, *Eq.* (*1c*): $q_{z,\mathrm{F}}$ instead of q_z; Eq. (2c), $q_{z,\mathrm{F}}$ instead of $q_{z,\mathrm{S}}$ and $2|T_\alpha||T_\beta|$ instead of $T_\alpha\cdot T_\beta$.

For the height–height correlation functions $c(x)$ an expression for self-affine rough surfaces (see e.g. Ref. [3]) was applied:

$$c(x) = \sigma^2\exp[-(x/\xi)^{2H}], \tag{3}$$

where ξ is the lateral correlation length of the height fluctuations, and H is the roughness exponent.

Fig. 3a shows the fits to the experimental data of Fig. 2 using the modified equations. With the modifications, the detector scan and the rocking curve in Fig. 3b can be fitted very well, both with the Sinha approach, Eqs. (1a), (1b) and (1c), and with the Pynn approach, Eqs. (2a), (2b) and (2c). The results for $\alpha = 1.51\,^\circ$, where the specular reflectivity in Fig. 1 has a minimum, are plotted in Fig. 4a and b. All four scans in Figs. 3 and 4 could be fitted with a consistent set of parameters, which are listed in Table 2. The contribution of the cross-correlation $(\mathrm{d}\sigma/\mathrm{d}\Omega)^{\mathrm{P}}_{\mathrm{C}}$ is additionally plotted on a linear scale in Fig. 3a and Fig. 4a, and we note that the pronounced oscillations of the detector scans are caused by a strong cross correlation between the free surface of the film and that of the substrate. The high quality of the Au1-film, already observed with the specular reflectivity, is confirmed by the fitting parameters of the cross-correlation function: The roughness, $\sigma_{\mathrm{C}} = 6.8$ Å, is comparable to those of the quartz glass substrate and the film, and the

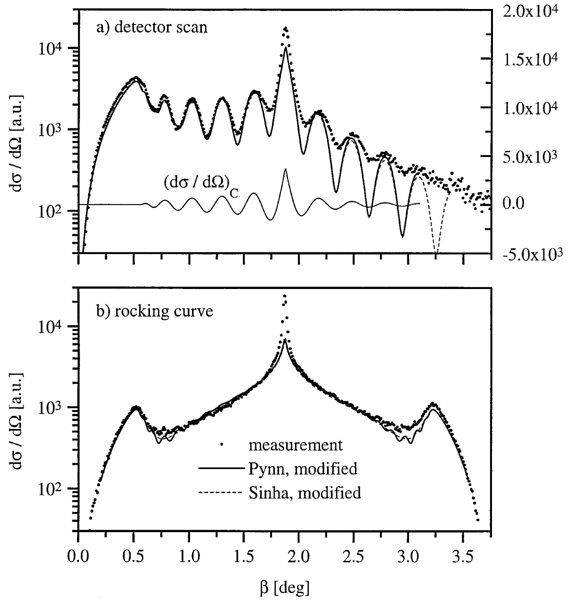

Fig. 3. Detector scan (a) and rocking curve (b) with Au1-film at $\alpha = 1.88\,°$: (●) experimental, (— — —) fit with Eqs. (1a), (1b) and (1c), (—) fit with Eqs. (2a), (2b) and (2c). The right-hand scale in (a) refers to the cross-correlation $(d\sigma/d\Omega)^P_C$ (Eq. (2c)).

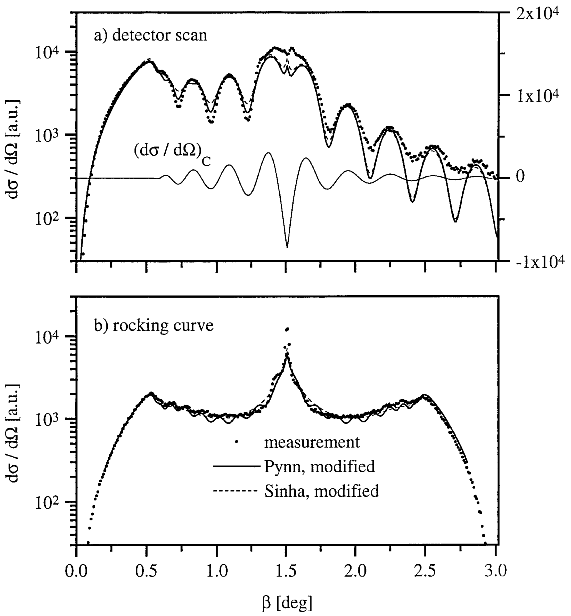

Fig. 4. Detector scan (a) and rocking curve (b) with Au1-film at $\alpha = 1.51\,°$: (●) experimental, (— — —) fit with Eqs. (1a), (1b) and (1c), (—) fit with Eqs. (2a), (2b) and (2c). The right-hand scale in (a) refers to the cross-correlation $(d\sigma/d\Omega)^P_C$ (Eq. (2c)).

Table 2
Parameters from diffuse scattering (according to Pynn, modified). c_F: free surface of the film; c_C: cross-correlation. The errors refer to the variation of the single scan fits

	σ (Å)	ξ (Å)	H
Substrate	7.0 ± 2.0	2000 ± 200	0.25 ± 0.05
Au1-film, c_F	7.8 ± 0.7	1300 ± 200	0.25 ± 0.04
Au1-film, c_C	6.8 ± 0.2	1300 ± 200	0.25 ± 0.04
Au2-film	15.0 ± 1.0	350 ± 50	0.75 ± 0.05

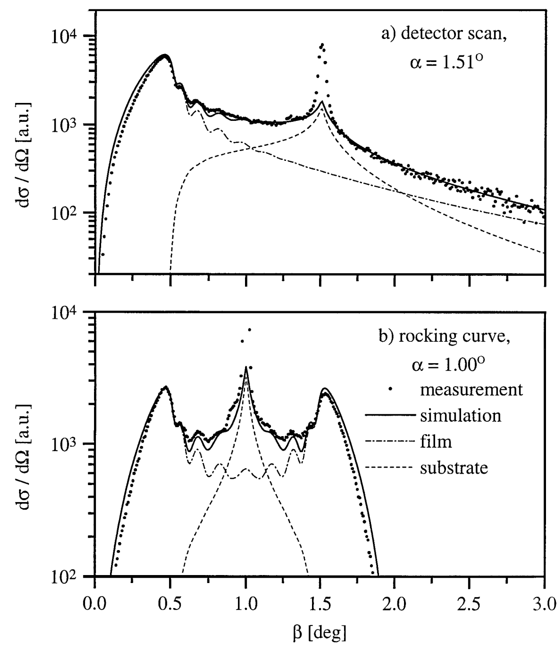

Fig. 5. Detector scan (a) and rocking curve (b) with Au2-film: (●) experimental, (—) fit with Eqs. (2a), (2b) and (2c), (— — —) substrate contribution $(d\sigma/d\Omega)^P_S$ (Eq. (2a)), (— · —) film contribution $(d\sigma/d\Omega)^P_F$ (Eq. (2b)).

correlation length, $\xi_C = 1300$ Å, is as large as that of the film surface. In addition, the values of the roughness exponent H are the same for the three contributions. This indicates that the surface of the gold film is a replication of the substrate surface.

A detector scan and a rocking curve of the Au2-sample are plotted in Fig. 5a and b together with the fits according to Eqs. (2a), (2b) and (2c). These fits show that no cross-correlation at all between the two surfaces exists, i.e. $c_C(x) = 0$.

The contributions of c_S and c_F, shown separately in Fig. 5a and b, are sufficient to describe the diffuse scattering. The correlation length of the surface of the Au2-film, $\xi_F = 350\,\text{Å}$, is distinctly smaller than that of the Au1-film. The larger roughness exponent, $H_F = 0.75$, indicates a less jagged surface.

4. Conclusions

X-ray reflectivity and diffuse scattering by means of a laboratory equipment can be sucessfully employed to characterize the quality of gold films sputtered on quartz glass substrates under different ambient conditions. The diffuse scattering shows that smooth Au films of high quality, as proved by their specular reflectivity, exhibit a perfect cross-correlation between the free surface of the film and the surface of the substrate. Slight modifications of theoretical expressions from literature for the diffuse scattering allow a quantitative description of the experimental data.

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