



Control of surface mobility for conformal deposition of Mo–Si multilayers on saw-tooth substrates



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ABSTRACT

Multilayer-coated blazed gratings (MBG) are the most promising solution for ultra-high resolution soft X-ray spectroscopy, since they can have very high groove density and provide high-order operation and very high diffraction efficiency. The performance of MBGs however depends critically on the conformal deposition of the multilayer (ML) stack on a saw-tooth substrate and the minimization of roughness. We present an analysis of the roughening and smoothing processes during growth of Mo/Si multilayers deposited over a range of pressures of Ar sputtering gas on flat and saw-tooth substrates. A Linear Continuum Model (LCM) of the film growth was used to understand the interplay between smoothing and roughening of the ML films and to predict the optimum conditions for deposition. The MBG coated under the optimal deposition conditions demonstrated high diffraction efficiency in the EUV and soft X-ray wavelength ranges

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1. Introduction

Fabrication of multilayer mirrors for EUV, soft and hard X-rays is currently a well established process. Key applications of multilayer mirrors such as EUV lithography, X-ray astronomy and spectroscopy, and synchrotron beamline optics require flat or slightly curved surfaces with a very low figure error ($\leq 1 \mu\text{rad}$), and with high reflectivity, and therefore demands interfacial roughness less than typically 0.3 nm. A number of deposition methods such as dc-magnetron sputtering, e-beam evaporation, ion-beam sputtering, combined with sophisticated technological adaptations such as ion-assisted deposition, deposition followed by a partial sputtering of layers, and application of diffusion barriers are aimed at providing high quality interfaces of the multilayer stack, a prerequisite for high reflectivity. Whatever deposition technique is used, it follows a common strategy. A deposition process should provide an effective relaxation of high spatial frequency roughness of interfaces generated during the deposition due to stochastic time-space variations of the deposition flux, polycrystalline structure of a film, shadowing effects etc., and at the same time prevent or reduce intermixing of the materials at the interfaces. Bulk diffusion across the interfaces should be suppressed while surface

diffusion along the interfaces should be promoted as much as possible. This approach was successfully implemented by a number of groups for flat or slightly curved substrates. As a result of these efforts, high quality multilayers have been developed with performance approaching theoretical limits [1,2].

However, the approach does not always work well for substrates which have a complicated surface relief. Deposition of the MLs on saw-tooth substrates which are of great importance for dense Multilayer-coated blazed grating (MBG) [3,4] applications is extremely challenging. A Fourier spectrum of a saw-tooth surface contains high order harmonics which are of the same frequency as the interfacial roughness. Since traditional deposition techniques aim to suppress high frequency imperfections of a surface, the high Fourier harmonics of the saw-tooth profile are affected by the smoothing as well. As a result, the highly corrugated surface of a saw-tooth substrate undergoes smoothing by multilayers deposited on it [5–7]. This dramatically affects the blazing performance of a MBG resulting in a significant reduction of diffraction efficiency.

To avoid groove profile degradation, a new deposition methodology needs to be developed specifically for MBG applications. We reported recently [8] on improvement of the diffraction efficiency of a MBG by deposition of an optimized Mo/Si multilayer on a saw-tooth substrate with a groove density of 5250 lines/mm. In this paper, we present our ML deposition optimization procedure. A detailed study of surface morphology and internal structure

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of the MLs has been performed and optimal deposition conditions have been found. As a result of this optimization, a record diffraction efficiency in the EUV and soft X-ray spectral ranges was achieved.

2. Experiment

2.1. ML deposition and characterization

Mo/Si multilayers composed of 30–40 pairs of Mo and Si layers and target d -spacing of 6.63 nm were deposited by dc-magnetron sputtering in an Ar gas pressure varied within a range of 1–5 mTorr in the multilayer coating facility at the Center for X-ray Optics at LBNL. The deposition of the layers was performed by continuous motion of the substrates over the magnetron sources at a source/substrate distance of 75 mm. Specially shaped apertures placed in between the sources and the substrates provided high uniformity of the layer thickness over the substrate area. The velocity of the motion and the power of the sources were adjusted to obtain the desired thickness of the layers.

The MLs were deposited on prime quality Si wafers which served as flat reference substrates and saw-tooth gratings which were fabricated with an anisotropic etching technique as described in Si [9–11]. The triangular groove gratings had a period of 190 nm with working facets tilted at an angle of 2° with respect to the grating plane. The height of 6.63 nm of the saw-tooth grooves corresponded to the d -spacing of the multilayers providing a blazed condition for the 1st diffraction order of the MBG.

The surface of the substrates was extensively characterized before and after deposition with a Veeco Dimension 3100 atomic force microscope (AFM). Silicon wafers used as substrates for the deposition were taken from the same batch in order to make sure they have very similar surface morphology and PSD spectra. Based on the AFM data, an analysis of the growth kinetics was performed using a Linear Continuum Model (see the Section 2.2) in order to find an optimal growth regime for saw-tooth substrates.

Measurements of the reflectivity of the multilayers and diffraction efficiency of the MBGs were performed at beamline 6.3.2 of the Advanced Light Source [12]. The internal structure of the multilayer stacks was investigated with cross-sectional TEM.

2.2. Growth analysis with the Linear Continuum Model

To investigate processes of roughening and smoothing of the ML interfaces during the growth, a surface kinetics analysis has been performed. Since the multilayers considered in this paper had a very low surface roughness, the Linear Continuum Model (LCM) [13–15] can be used for the analysis. According to this model, the two-dimensional isotropic power spectral density (PSD) function of the top surface of the coating (PSD_{top}) can be expressed as:

$$\text{PSD}_{\text{top}} = \text{PSD}_{\text{film}} + a\text{PSD}_{\text{sub}} \quad (1)$$

$$\text{PSD}_{\text{film}} = \Omega \frac{1-a}{\sum 2\nu_n f^n}, \quad (2)$$

$$a = \exp \left(2d \sum_n \nu_n f^n \right), \quad (3)$$

where PSD_{sub} is the power spectral density of the substrate. The replication factor, a , characterizes smoothing of a substrate relief by the film, and PSD_{film} characterizes the intrinsic roughening/smoothing dynamics of the film surface during the growth process. The replication factor is a spatial frequency (f) dependent function, and a characteristic of the dependence is defined by a

relaxation exponent, n , which can range from 1 to 4 for different relaxation mechanisms [13,16,17]. The contribution of a particular relaxation mechanism is characterized by a strength parameter, ν_n , and depends on the film thickness, d .

Due to the random nature of a deposition process, stochastic roughening occurs during the growth. This depends on the thickness of a coating and the volume of deposited particles, Ω , which in the simplest case of an amorphous film growth corresponds to the atomic volume of the deposited material [14,15]. Non-stochastic roughening caused by 3D island growth of the film might also occur. This roughening is also frequency dependent and can be taken into account by adding respective terms to the sum in the formula (2) but with an opposite sign as compared to the smoothing parameters [13]. For our simulations we assumed that surface diffusion and desorption, re-sputtering, or ballistic mechanisms characterized with the exponents $n=4$ and $n=2$, and negative strength parameters ν_4 and ν_2 respectively are relevant for the room temperature growth conditions while contributions of viscous flow and bulk diffusion can be neglected ($\nu_1 = \nu_3 = 0$). Also, following earlier work [13] we took into account a non-stochastic roughening caused by island growth at the early stages of deposition of the layers. Such a process is characterized with an exponent $n=1$ and a strength parameter, ν_1 , having a positive sign.

3. Results and discussion

3.1. Impact of the sputtering gas pressure on Mo/Si structure and growth dynamics

Dependence of the surface roughness of the Mo/Si multilayers on the sputtering gas pressure was investigated by AFM. AFM images of the top surface of the multilayers deposited at Ar pressure of 1–5 mTorr (Fig. 1) show that the pressure dramatically affects the surface roughness which progressively increases with the pressure. Surface morphology remains structureless in the range of 1–4 mTorr, but at a pressure of around 5 mTorr, a transition to well-shaped grain morphology occurs which is accompanied by a sharp rise in the surface roughness (Fig. 1f).

Cross-sectional TEM images of the ML deposited at 1, 3.5, and 5 mTorr reveals an internal structure of the multilayer stacks and shows evolution of the interface roughness in the course of the deposition (Fig. 2). No roughness buildup is observed for the deposition at a pressure of 1 mTorr. Non-correlated roughness of the interfaces of the multilayer is fairly low and stays at the same level through the entire thickness of the ML stack. A moderate progressive increase of interface roughness from the substrate toward the top surface of the ML stack is observed for the 3.5 mTorr multilayer, and some roughness correlation for adjacent interfaces is also clearly visible. A strong interface roughness correlation is observed for the 5 mTorr deposition resulting in the formation of well-known column-like structure of the stack [18–21] and a dramatic roughness increase.

Such evolution of structure is the result of the dependence of the dynamics of a growing surface on deposition conditions. Analysis of the PSD spectra of the top surface of the multilayers gives us an insight into the interplay of roughening and relaxation processes taking place during ML growth. The PSD spectra of the top surface of the Mo/Si MLs deposited at Ar pressures between 1 and 5 mTorr are shown in Fig. 3 (symbols). The best fits obtained with the LCM (formulas (1–3)) are shown with solid lines. Values of the fitting parameters for the ML PSDs are listed in Table 1. To take into account the contribution of substrate roughness to the PSDs, one of the silicon substrates was inspected with AFM before the deposition and an empirical PSD_{sub} function was generated (not shown in Fig. 3) and used in the simulation.

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