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# Characterization of molybdenum/silicon X-ray multilayers

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## Abstract

Mo/Si multilayers (MLs) with variable Mo thickness were fabricated using electron beam evaporator. Percolation thickness for Mo was determined experimentally. MLs with Mo thickness below percolation show low reflectivity due to discontinuous nature of Mo film. As the number of layer pair increases, the interfacial roughness increases, due to increase in correlated roughness. Extreme ultra violet reflectivity was measured using synchrotron radiation. The fitting result reveals that the graded interface layer exists at each interface. Cross-sectional transmission electron microscopy has been done on some of these MLs.

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## 1. Introduction

Molybdenum/silicon (Mo/Si) multilayers (MLs) have high reflectance in the extreme ultra violet (EUV) region of 130–300 Å [1] and are finding extensive applications as optical elements in EUV lithography, plasma diagnostics and astronomy [2–4]. The combination of Mo and Si forms good quality MLs and high normal incidence reflectivity is obtained. Reflectivity in MLs strongly depends on the imperfection e.g. grain boundaries, faceting, columnar growth and others, at the interface. In addition, inter-diffusion and reaction at the interface can reduce the contrast in optical constants, which can reduce the reflectivity. The stability of these MLs to thermal loading is important. Thus there is a need to study the structure and growth of layers and interfaces. In this paper we report the study of interfaces in Mo/Si MLs using X-ray reflectivity (XRR) and high-resolution electron microscopy.

## 2. Experimental and data analysis techniques

The Mo thin films and Mo/Si MLs were deposited on good quality float glass and silicon (1 1 1) wafers by using an ultra high vacuum electron beam evaporation system [5]. In situ resistance of 200 Å Mo film was measured during deposition by four-probe method. The quartz crystal monitor was used for in situ thickness control. The deposition rate was typically 0.1 Å/min for

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in situ resistance measurement and 1 Å/min for ML fabrication. XRR measurements were carried out at $\lambda = 1.54$ Å using Siemens D5000 diffractometer [6]. EUV/soft XRR measurements were performed on a reflectivity beamline on Indus-1 synchrotron storage ring [7]. The transmission electron microscope (Philips CM 200) was used for micro structural characterizations of some of the Mo/Si MLs.

The measured X-ray and EUV reflectivity data were fitted using the Parratt formalism [8]. Interface roughness effect is taken into account by modifying the Fresnel reflection coefficients at each interface according to the Nevot–Croce model [9].

## 3. Results and discussion

A typical in situ resistance measurement during growth of a Mo film on float glass is shown in Fig. 1. The graph of resistance $R$ (Ω) versus thickness $t$ (nm) shows three distinct regions. An initial "induction" region (nucleating stage) where $R$ changes only slowly with thickness and the resistance is predominantly that of the substrate surface. This is followed by metallic nuclei growth stage where a rapid decrease in $R$ over many orders of magnitude. In the third region $R$ remain almost constant at the value of $R \approx 60$ Ω. The inset shows $Rt^2$ versus $t$ plot of the resistance measurement, which indicates a pronounced minimum at $t = t^{\min}$. As

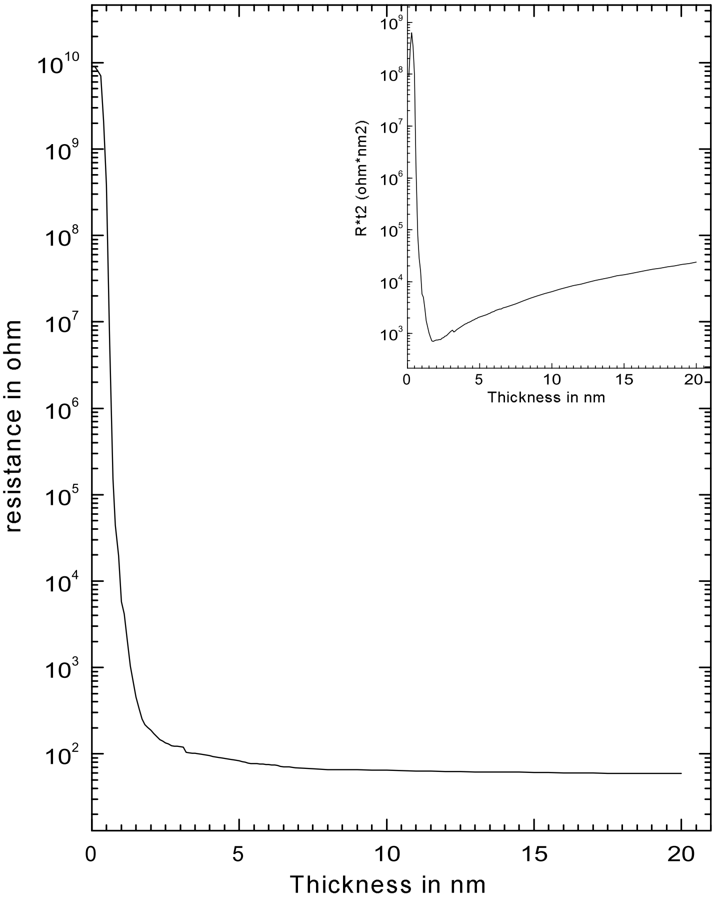

Fig. 1. The variation of resistance $R$ with thickness '$t$' during growth of 200 Å Mo thin film on float glass measured using four-probe method. Inset shows the $Rt^2$ versus thickness $t$ plot of resistance measurement.

the thickness increases over the range $0 \ll t \ll t^{\min}$ the metallic island nuclei grow in size to form a conducting metallic network, which "in fills" to form a continuous film at $t \approx t^{\min}$. The percolation threshold derived was 18 ± 0.5 Å. Veldkamp et al. [10] have determined the percolation threshold of 25 Å for rf sputter deposited Mo films while in our case it is 18 Å for e-beam evaporated films. This difference could be attributed to the different deposition techniques adopted and/or the use of different in situ resistance measurement procedure.

Mo/Si MLs with 50 Å periodicity and $N = 5$ layer pairs were fabricated. To correlate XRR measurements with in situ resistance data, Mo thickness in MLs were varied from 13 to 30 Å. The fitted values of the four Mo/Si MLs with ascending order of Mo thickness are given in Table 1. As Mo thickness increases from below percolation to just above of the percolation, the rms roughness $\sigma$ decreases significantly and the peak reflectivity increases by one order of magnitude. This was due to the discontinuous nature of Mo film for sample 'A' and 'B'. For sample 'C' and 'D', Mo forms a continuous layer leading to lower value of rms roughness and significant increase in peak reflectivity. Mo/Si ML mirrors with $d = 100$ Å keeping Mo thickness just above of the percolation were fabricated with number of layer pair $N$ from 6 to 20. Table 2 shows the fitted result of three MLs with $N = 6$, 10 and 20. A typical XRR spectrum of 27 Å Mo/78 Å Si with $N = 6$ is shown in Fig. 2. It shows Bragg peak up-to fifth order along with the secondary minima, which indicates good control of thickness in the ML structure. The fitted pattern using the Parratt formalism is shown by continuous curve. As $N$ increases the interfacial roughness increases slightly. This is due to increase in correlated roughness for larger number of layer pairs as observed in cross-sectional transmission electron microscopy measurements on these samples.

Table 1
XRR fitted results of Mo/Si MLs with $d = 50$ Å

| Sample no. | No. of layer pairs, $N$ | Thickness, $d_{\mathrm{Mo}}$ (Å) | Roughness, $\sigma_{\mathrm{Mo}}$ (Å) | Roughness, $\sigma_{\mathrm{Si}}$ (Å) | Peak reflectivity (%) |
|---|---|---|---|---|---|
| A | 5 | 13 | 15 | 12 | 0.2 |
| B | 5 | 17 | 9 | 8 | 1 |
| C | 5 | 19 | 7 | 6.5 | 6 |
| D | 5 | 30 | 7 | 6.5 | 4 |

The Mo layer thicknesses were varied from below to above percolation threshold.

Table 2
XRR fitted results of Mo/Si MLs with $d = 100$ Å (22 Å Mo/78 Å Si)

| Sample no. | No. of layer pairs, $N$ | Roughness, $\sigma_{\mathrm{Mo}}$ (Å) | Roughness, $\sigma_{\mathrm{Si}}$ (Å) | Peak reflectivity (%) |
|---|---|---|---|---|
| 1 | 6 | 6 | 5.5 | 48 |
| 2 | 10 | 7 | 7 | 44 |
| 3 | 20 | 8 | 8 | 50 |

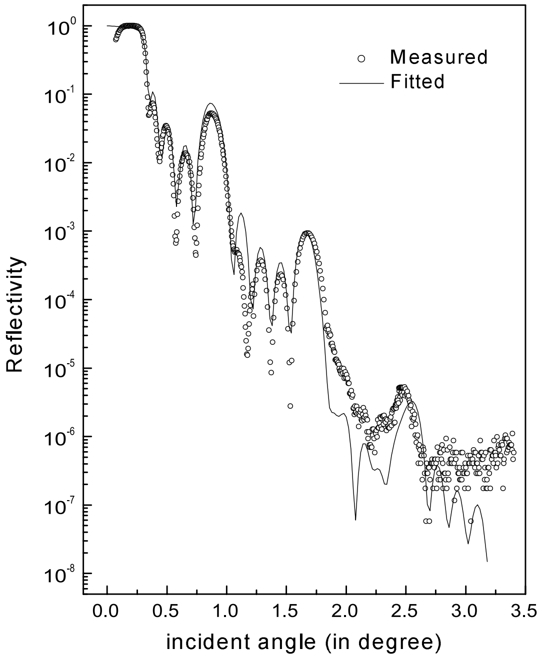

Fig. 2. XRR measured at CuKα (1.54 Å) for Mo/Si ML (27 Å Mo/78 Å Si)$_6$. The measured data is shown as open circles and fitted curve is shown as continuous line.

Angle dependent EUV reflectivity spectrum of Mo/Si MLs deposited on float glass substrate of period 89 Å (30 Å Mo/59 Å Si) with five layer pairs, measured at 80 Å wavelength, is shown in Fig. 3. Measured spectrum is shown by circles whereas dotted line represents calculated spectrum assuming a two-layer model without any interdiffused layer. The disagreement between calcu-

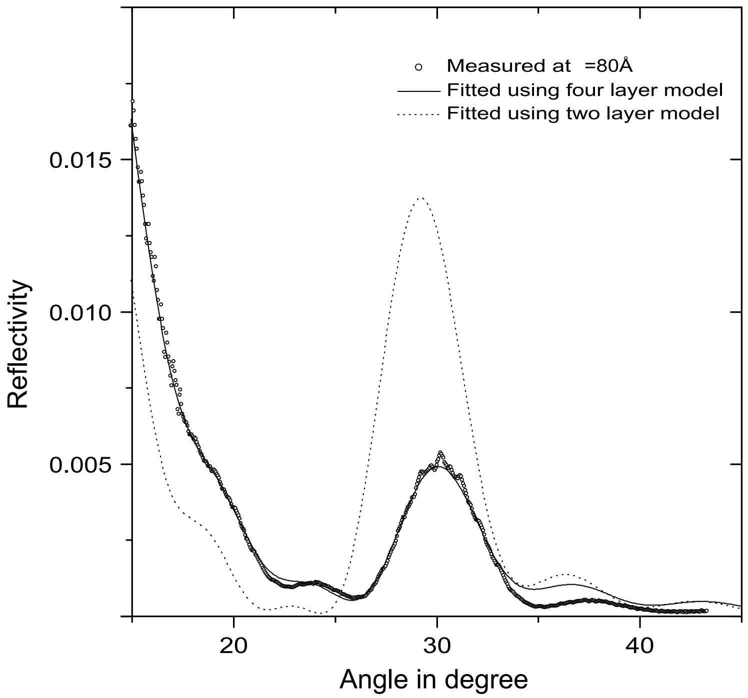

Fig. 3. EUV reflectivity measured at $\lambda = 80$ Å for a Mo/Si ML (30 Å Mo/59 Å Si)$_5$. Open circles show measured data. The dotted line shows calculated spectrum by assuming two-layer model. The best fit represented by continuous curve was obtained by assuming an interdiffused layer in between pure layers of Mo and Si. The average rms roughness of 5.5 Å was considered for all interfaces to get best fit.

lated and measured spectrum is quite distinct. This suggests that the ML structure is not a simple two-layer periodic system but is separated by an inter-layer of Mo–Si. Detailed fitting reveals that an inter-layer of Mo–Si exists in between the pure layer of Mo and Si. The thickness of two inter-layers formed on Si-on-Mo interface and Mo-on-Si interface is 8 and 10 Å, respectively. Morgan and Boercker [11] in their molecular beam dynamics study, explained the thickness asymmetry. The different degree of penetration and inter diffusion of the ad atoms during deposition are basic factor behind the asymmetry. Stearns et al. [12] have suggested that the Mo atoms can get embedded more easily into the relatively open and disordered lattice of amorphous Si, than are the Si into the more compact crystalline Mo lattice. A systematic study to understand the behavior of these inter-layers is of major interest. Hence, ML systems with metal layer thickness near to percolation threshold need to be investigated in detail for the behavior of inter-layers.

Cross-sectional electron microscopy studies have been carried out to study the interfaces of Mo/Si MLs, which govern the reflectivity property in soft X-ray region. Fig. 4 shows the cross-sectional TEM for Mo/Si ML where the Si layers are light and Mo layers are dark. The photograph shows that the ML has distinct layered structure but roughness increases from bottom to top of the stack. This is typical in thin films, which experience columnar growth, typically due to low mobility of the depositing atoms. To study the effect of high temperature on the interface of these MLs, micro structural changes at high temperatures were studied. At 750 °C, all the interface diffuse into one another, forming a $Mo_5Si_3$ crystalline phase [13].

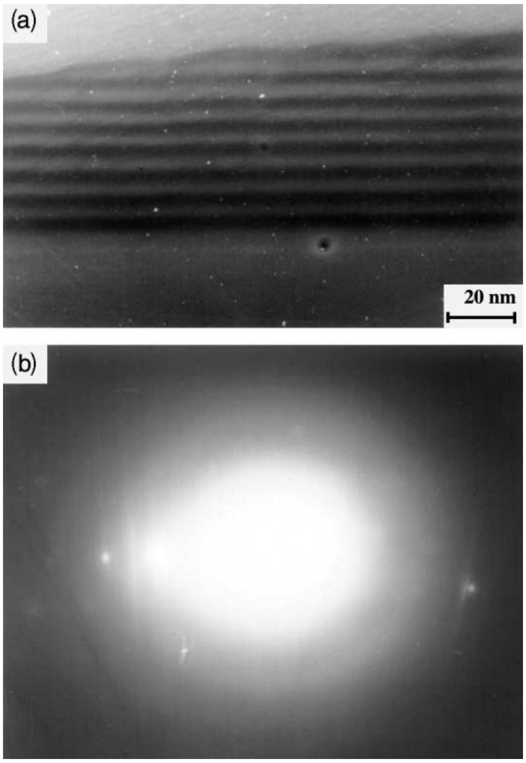

Fig. 4. High-resolution image of Mo/Si ML deposited on Si(1 1 1) substrate.

## 4. Conclusions

To obtain good interfaces in Mo/Si MLs, the Mo thickness should be greater than the percolation threshold of 18 Å. For the electron beam evaporated samples, the interfacial roughness increases with increase in number of layer pairs. This is because of the low energy deposition process. Detailed reflectivity fitting in EUV region shows that intermixed layer of Mo and Si exists at the interface. The thickness of this intermixed layer is different for the two interfaces. Direct structural characterization of interfaces has been done using cross-sectional TEM.

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