Characterization of Mo/Si Multilayer
by X-Rays

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Mo/Si multilayer is synthesized and its structure is evaluated on atomic scale using various soft X-rays (Mg-Kα, F-Kα, O-Kα, and C-Kα). Two types of models based on the optical and diffraction methods were considered. It is found that the latter theory is more effective in extracting multilayer structure.

1. Introduction

Recently, the synthetic multilayer for X-ray optical element has been increasingly used light element analysis in the PIXE, XRF, X-ray microscopy, and X-ray lithography.
Material combinations of the multilayer as X-ray reflectors are selected to get a high peak reflectivity in the Bragg reflection. Since interstitial roughness and errors in periodicity of the multilayer may drastically reduce their quality, decrease the peak reflectivity and increase the inherent bandwidth, it is generally considered that the quality of the multilayer requires investigation before such applications can confidently be pursued.

Petford-Long et al. [1] showed in their electron micrograph for the cross section of a Mo/Si multilayer that there was a mixing region, δ of 10-17Å at the interface. On the other hand, Yamashita et al. [2] concluded that values of 6-7Å for the Interfacial roughness, σ, in Fresnel equations of Mo/Si multilayers were obtained from the peak reflectivity for CuKα, however with Fresnel equations, σ becomes larger as the X-ray energy decreases. Although sharp boundaries of dielecctric constant are assumed in the optical method, a diffraction pattern can be obtained for an arbitrary distribution of the dielectric constant in the crystal lattice with the diffraction method. Ito et al. [3] estimated the interfacial mixing, δ, at 17Å on the Mo/Si multilayer from the diffraction method using the Bragg reflection.

Mo/Si multilayer was prepared and invesitigated in order to elucidate the structure on atomic scale by the diffraction method using various characteristic soft X-rays and the result obtained from the diffraction model was compared with that of the optical model.

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Y. ITO, T. MUKOYAMA, K. OMOTE, K. SHIMIZU, A. NISAWA, T. SHOJI, H. TERAUCHI and S. KUEHNER

2. Experimental

The Mo/Si multilayer was fabricated by the dual electron beam evaporation method in an UHV chamber equipped with an ion pump and a Ti sublimation pump. Initial vacuum in the deposition chamber was in a range of 10^{-8} Pa and increased to 10^{-6} Pa during evaporation. A silicon wafer (625 μm in thick) was used as a substrate. The silicon wafer was placed on a molybdenum block and desorb gas trapped on the substrate. In order to reduce the interface mixing, the substrate was cooled and kept at room temperature during the deposition. The purity of the source materials used was 99.9999% and 99.95% for Si and Mo, respectively. The thickness of each layer was monitored with two water-cooled quartz crystal oscillators. The evaporation rates were 0.2Å/sec for Si and 0.5Å/sec for Mo. The ratio of Mo sublayer to Si sublayer is 7 to 8 in thickness. The multilayer has a bilayer thickness of 98.8Å with 8 layers. The thickness of the multilayer, d was obtained from the 6th order Bragg reflection for Cu-K α radiation (RIGAKU RU-300, for thin film x-ray diffractometer).

A schematic diagram of the reflectivity measurement system is shown in Fig.1.
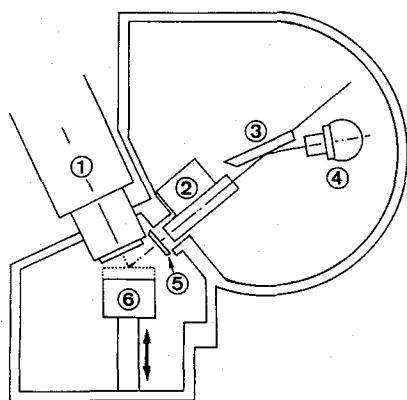

.Fig.1a. Schematic diagram of the spectro-
meter; 1.X-ray tube (Rh tube was
used in this experiment) , 2.soller
slit, 3.multilayer, 4.proportional
counter, 5.attenuator, 6.sample hol-
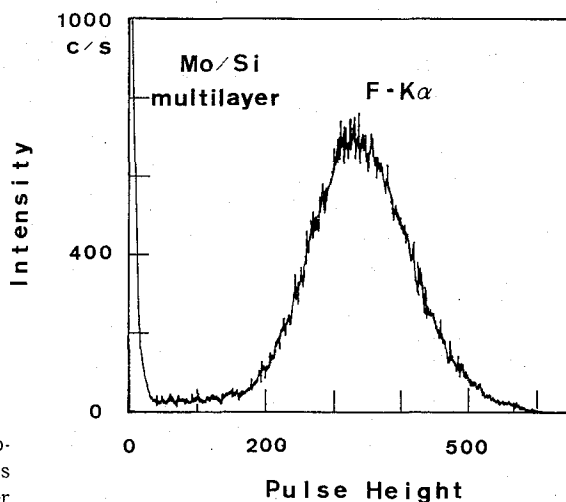der (for the fluorescence x-ray).

Fig.1b. Pulse height distribution curve of
F-K α measurement.

A thin-window gas flow proportional counter (FPC) is mounted on the rotation arm in the vacuum chamber. The incident beam is collimated by a soller slit with an angular divergence of 4.5×10^{-3} rad.. Characteristic X-rays produced from the secondary targets (Fig.1a) are distinguished by the pulse height distribution of the proportional counter (Fig.1b). The intensity of the first and higher order Bragg reflections and their FWHM (Full Width at Half Maxima) were measured and the reflectivities of the multilayer obtained for each characteristic X-ray wavelength.

3. Results and Discussion

The two diffraction curve (F-K α and C-K α) of four characteristic X-rays for Mo/Si (d=98.8Å, N=8, number of layer pairs) are shown in Fig.2. The intensity of the diffraction

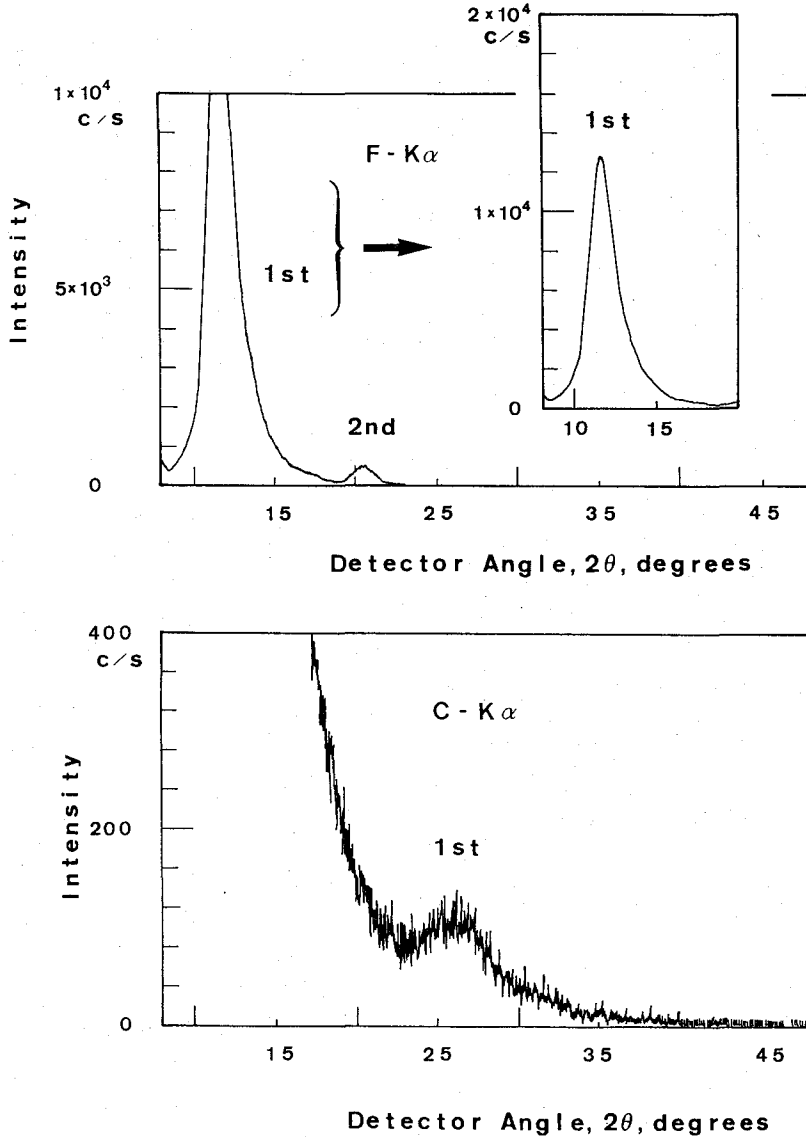Fig.2. Diffraction curves of Mo/Si (d=98.8A, N=8) tor F-K α and C-K α .

decreases as the order in reflection is higher in this figure.

The observed peak reflectivity and FWHM of the first and higher order Bragg reflections are shown in Table 1 and compared with calculated ones based on optical and diffraction

Y. Ito, T. Mukoyama, K. Omote, K. Shimizu, A. Nisawa, T. Shoji, H. Terauchi and S. Kuehner

Table 1. The observasion and calculation of reflectivity (Rp) and FWHM in Mo/Si multilayer with diffraction and optical method for various characteristic X-rays.

		Diffraction method			Optical method	
	obs.	I	II	III	δ =0	δ =17Å
Mg-K α (λ [#]=9.89Å)						
n[*]=2 Rp (%)	1.0	0.25	0.2	0.2	1.6	0.01
FWHM (2 θ) (°)	1.05	0.72	0.66	0.74	0.55	0.50
n=3 Rp	0.02	2.0	0.01	0.1	3.1	-
FWHM	1.40	0.72	0.72	0.66	0.60	-
F-K α (λ [#]=18.37Å)						
n=1 Rp	5.4	4.0	3.3	3.3	19	4.0
FWHM	2.15	3.24	3.64	3.68	1.60	1.40
n=2 Rp	0.2	0.05	0.04	0.03	0.6	0.004
FWHM	1.60	1.22	1.66	1.42	1.10	1.10
n=3 Rp	0.007	0.35	0.002	0.04	1.40	-
FWHM	-	1.40	1.41	1.56	1.3	-
O-K α (λ [#]=23.62Å)						
n=1 Rp	0.4	1.4	1.2	1.2	12	2.5
FWHM	2.85	5.86	5.82	5.60	2.05	1.95
n=2 Rp	0.02	0.02	0.02	0.01	0.4	0.002
FWHM	2.40	2.66	3.20	2.96	2.00	1.60
n=3 Rp	0.001	0.1	0.001	0.02	0.9	-
FWHM	-	1.60	-	2.44	1.80	-
C-K α (λ [#]=44.7Å)						
n=1 Rp	0.1	0.3	0.2	0.1	1.5	0.4
FWHM	4.00	5.76	5.98	5.48	3.70	3.40

[#]:after J.A.Bearden[10]

[*]:2dsin θ =n λ

methods. Dynamical effects based on the absorption as well as on the interaction between incident and scattered radiations are considered to yield significant contribution to the reflectivity. We have the two types of the models: optical and diffraction-based. Optical constants of f' and f" in Fresnel equations in the optical method were taken from Henke et al. [4] and atomic scattering factors in the diffraction method were given by the International Tables for X-ray Crystallography, Vol. III [5] , Energies and cross sections for each orbital used in the dispersion calculations were obtained from Los Alamos Scientific Laboratory Report [6] .

For an actual multilayer, the boundary between the layers is not expected to be discrete since mixing as well as interfacial roughness take place between the layers during evaporation. The former has been confirmed by analyzing the composition of Mo/Si multilayer through high-resolution electron microscopy [1]: The interfacial mixing, δ , in Mo/Si multilayer was 17Å for Si sublayer and 10Å for Mo sublayer.

First, we calculated the reflectivity and it's bandwidth of Mo/Si multilayer by optical method based on Fresnel equations [3,7,8] . The value of the interfacial roughness, σ can be evaluated by comparing the theoretical reflectivity for a given sample with the experimentally observed reflectivity. The result is shown in Table 1. In calculating the reflectivity with this method, we used the value of the density of ρ (Mo) =9.46 g/cm^3 and ρ (Si) =2.14 g/cm^3. From the observed reflectivity for various X-ray energies in Table 1, the in-

terfacial roughness in the multilayer is estimated at $\sigma \sim 17\text{Å}$. Intensities of observed higher order reflections do not match the calculated ones except for those of the first Bragg reflections. While the optical-based model can predict the reflected intensity at a selected Bragg peak, that is, the order reflection, this type model gives poor fits for the rest of the spectrum or the higher order spectrum. As shown in Fig.3a, the concept of the atomic mixing at the interface between each layer is not included in the optical method. Only a variation in the interface of each layer is considered in this treatment.

Second, we used another approach including interfacial mixing, based on the diffraciton method [3,9] , to extract structural information of the multilayer. When multilayer is periodic, we can consider the structure as an artificial crystal and calculate the diffraction
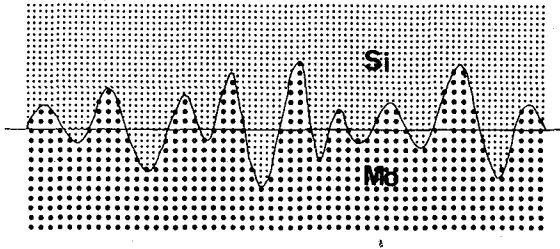

Fig.3a. The interfacial roughness, σ at presents the rms of the fluctuation between Si and Mo layers in the optical method.
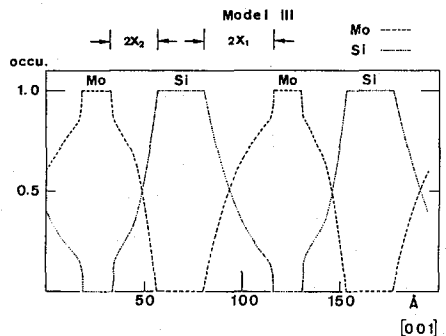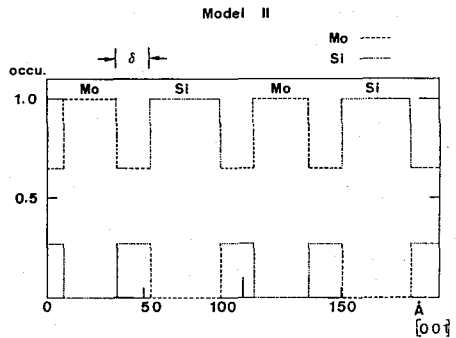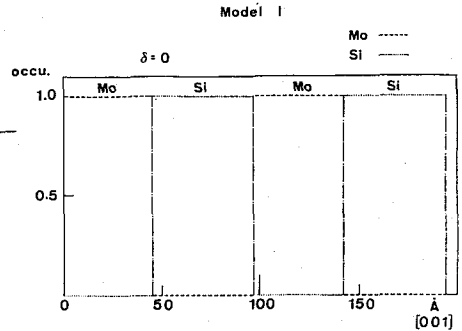
Fig.3b. Diffraction models for the Mo/Si multilayer. The models in the interface between Mo and Si layers are as follows; I is discrete type (δ =0) , II is a statistical type ($\delta \sim 17\text{Å}$) and the concentration is constant, and III has a concentration gradient for each atom represented by $\exp(-x/\delta)$, where x is the mixing length and δ is the rms interfacial mixing. δ_1 is the mixing length along the growth direction [001] between the preceding Si layer and Mo layer ($\delta_1 \sim 17\text{Å}$) , δ_2 is the mixing length along the growth direction [001] between the preceding Mo and Si layer ($\delta_2 \sim 10\text{Å}$) .

Y. ITO, T. MUKOYAMA, K. OMOTE, K. SHIMIZU, A. NISAWA, T. SHOJI, H. TERAUCHI and S. KUEHNER

profile with formalism of the diffraction theory, even if the multilayer shows that the Mo sublayers are crystalline and the Si sublayers considered to be amorphous. Three types of structure models have been proposed for the multilayer by comparing the theoretical reflectivity of a given sample with the experimentally observed one (Fig.2b and Table 1). In the calculation, we used the same value for the density of each layer as in the optical method. As shown in Fig.3b, in Model I, interfaces are discrete between Mo and Si sublayers. Model II is a statistical type without the gradient concentration of the atom and Model III has the gradiation of the atomic concentration expressed by $\exp(-x/\delta)$, where x is the mixing length, $x=2\delta$ in this case, and msr interfacial mixing, $\delta=10\text{Å}$ for Si sublayers and $\delta=17\text{Å}$ for Mo sublayers from the result reported by Petford-Long at al. [1]. As seen in Table 1, the diffraction method can more correctly, to a certain extent, predict the reflected intensity at a first Bragg peak than the optical method. Moreover, Model II can exactly match the heigher order, especially the 3rd reflected intensities over a range of various characteristic energies. The inference may be made regarding multilayer structure from this fact: The interfacial mixing exists between Mo and Si sublayers and the distribution of the atom in the interfacial mixing region may be considered to be not exponential as in Model III, but statistical as in Model II. The dominant factor in determining the width of a low angle superlattice peak is the number of repeat layers comprising the multilayer. As seen in Table 1, the calculated peak width in the diffraction method of larger than the observed one and decreases rapidly as the order of Bragg reflection becomes higher, to second order in this case, but the observed peak width is not reduced so fast to the higher order reflection. The difference may be due to bilayer thickness fluctuations.

The result that Model II is suitable for the Mo/Si multilayer, is comparable with the ones of the electron microscopy study [1] that each atom can penetrate into each layer not by atomic diffusion but by momentum and a mixing occurs at the interface of the Mo/Si multilayer structure.

4. Conclusion

With various characteristic soft X-rays, we distinguished two types of models: optical and diffraction-based. The latter was more fruitful in acquiring the structural information from the Mo/Si multilayer. The dominant factor in determining the reflectivity of higher order reflection is not the interfacial roughness but the interfacial mixing in the multilayer. Therefore, the diffraction method can more correctly predict the reflected intensity at Bragg peak over a range of q than the optical method.

We intended to extend this study to a number of other multilayer structures, since we feel that the diffraction method can be valuable in characterizing such materials.

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