LAYER THICKNESS DETERMINATION OF THIN FILMS BY GRAZING INCIDENCE X-RAY EXPERIMENTS USING INTERFERENCE EFFECT

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ABSTRACT

A novel method using Fourier transform algorithm is proposed to determine each layer thickness of multi-layered thin films from interference oscillation observed in X-ray specular reflection. The peak position in Fourier space gives each layer thickness of the film. The principle of the present technique as well as its applications are described.

INTRODUCTION

Grazing incidence X-ray techniques recently have been extensively used for the near-surface study of materials and the characterization of thin films.[1-9] In those experiments, as is often the case in practical analysis of multi-layered thin films, a complicated oscillating structure is observed in the angular/energy dependence of X-ray reflectivity and the signal proportional to the amplitude of the electric field (for example, fluorescent X-rays, electron yield). This is due to interference caused by multiple reflections of X-rays at each interface,[1,10,11] and therefore, such oscillation includes the information on layer thickness and on interface roughness.

In the earlier works,[12-14] the period of the reflectivity oscillation, which directly relate to layer thickness, were analyzed by direct reading of the position of maxima and minima. This method is effective for a single film layer, but has inherent limitations in the analysis of the complicated curve for increased layer numbers. On the other hand, recently, least-squares curve fitting procedures has been often employed using theoretical models.[4,15] Layer thicknesses as well as surface/interface roughness are determined automatically. However, it is difficult to obtain a best fit for the entire reflectivity curve, when neither every layer thickness nor every interface roughness is known.

In the present paper, a new technique for determining each layer thickness of multi-layered thin films is reported. A frequency analysis of reflectivity oscillation based on Fourier transform is promising.[16] This works well when any interface roughness is not given.

FORMULATION OF OSCILLATING PART OF REFLECTIVITY

The reflectivity from a multi-layered thin film is usually calculated using a recursive equation.[1,17] For the simple three-layered model, i.e., a single film layer (2nd layer) besides air (1st layer) and the substrate (3rd layer), reflectivity R is written as follows, when absorption is negligible :

$$R = R_{1,2}{}^2 = (R_{2,3} + F_{1,2})^2 / (R_{2,3} F_{1,2} + 1)^2$$
$$= (\exp(-i\gamma_2) F_{2,3} + F_{1,2})^2 / (\exp(-i\gamma_2) F_{2,3} F_{1,2} + 1)^2$$

where

$$\gamma_j = 4\pi d_j \{(\theta^2 - \theta c_j{}^2)^{1/2} / \lambda\}$$

$R_{j-1,j}$ and $F_{j-1,j}$ are the reflection coefficient and the Fresnel coefficient at the interface between (j-1)th and jth layers, respectively; θ is the glancing angle; d_j and θc_j are the critical angle and the layer thickness of jth layer, respectively; λ is the wavelength of the X-rays.

When R is small, this equation is simply rewritten:

$$R = (F_{1,2}{}^2 + F_{2,3}{}^2 + 2 F_{1,2} F_{2,3} \cos\gamma_2) / \{(1 - F_{1,2}{}^2)(1 - F_{2,3}{}^2)\} \qquad (1)$$

This equation indicates that the oscillation part of reflectivity is expressed as a simple cosine function. Furthermore, it is important that γ_2 in $\cos\gamma_2$ is the product of d_2 and $(\theta^2-\theta c_2{}^2)^{1/2}/\lambda$. That is, the cosine oscillating part of the data, when plotted as a function of $(\theta^2-\theta c_2{}^2)^{1/2}/\lambda$, are converted by Fourier transform to the distribution of d_2. A single peak, whose position gives the layer thickness d_2, is obtained in the Fourier space.

As the number of layers is increased, the equation becomes rather complicated, but essentially the same analysis is possible. For example, for four-layered model, R is written :

$$R = (A \cos\gamma_2 + B \cos\gamma_3 + C \cos(\gamma_2+\gamma_3) + D \cos(\gamma_2-\gamma_3) + E) / F \qquad (2)$$

$$A = 2 F_{1,2} F_{2,3} (1 + F_{3,4}{}^2);\ B = 2 F_{2,3} F_{3,4} (1 + F_{1,2}{}^2);\ C = 2 F_{1,2} F_{3,4};$$
$$D = 2 F_{1,2} F_{2,3}{}^2 F_{3,4};\ E = F_{1,2}{}^2 + F_{2,3}{}^2 + F_{3,4}{}^2 + F_{1,2}{}^2 F_{2,3}{}^2 F_{3,4}{}^2;$$
$$F = (1 - F_{1,2}{}^2)(1 - F_{2,3}{}^2)(1 - F_{3,4}{}^2)$$

In this equation, reflectivity oscillation is expressed as the sum of four cosine wave. The distribution of d_j is obtained by Fourier transform of the data plotted as a function of $(\theta^2-\theta c_j{}^2)^{1/2}/\lambda$.

Though the correction term for absorption should be included in γ_j, it is negligible when R is small, and therefore, equations (1) and (2) can be used for cases with absorption.

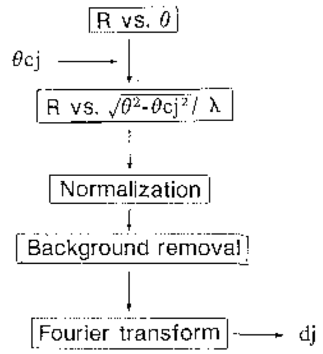

Figure 1
Analytical scheme for determining each layer thickness of multi-layered thin films.

ANALYTICAL SCHEME

The scheme for the analysis of interference oscillation was summarized in Fig.1. First, the experimental angular distribution of reflectivity is measured. Then, the data is re-plotted as a function of $(\theta^2\text{-}\theta c_j^{\,2})^{1/2}/\lambda$. To compensate for the attenuation in the higher angle region, the data are normalized by the average curve, which is calculated in the logarithmic plot. After that, the oscillating part is extracted by subtracting the non-oscillating background. Finally, the data are Fourier transformed.

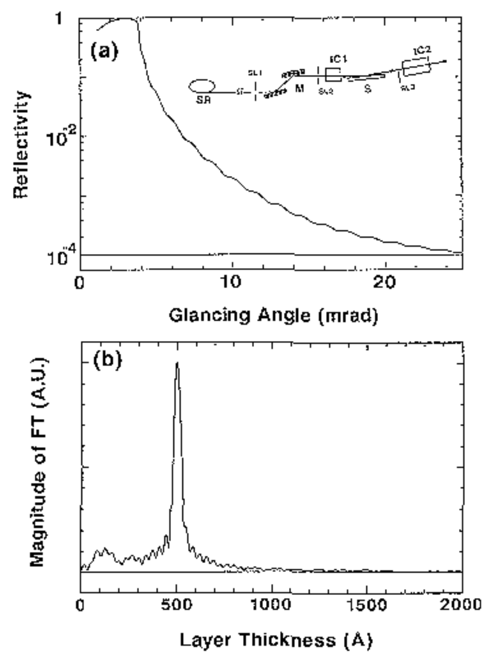

Figure 2
(a) Interference structure of SiO_2[501Å]/Si in the 8 keV X-ray specular reflection. Schematic drawings of the experimental arrangement is shown in an inset; M: a monochromator, IC1, IC2: ionization chambers for incident and reflected X-rays, respectively, S: a sample, SL1, SL2, SL3: slits. (b) Magnitude of the Fourier transform of the oscillation extracted from Fig.2(a), after plotting as a function of $(\theta^2\text{-}\theta c_2^{\,2})^{1/2}/\lambda$. The peak indicates the frequency component of the oscillation, and agrees with the film thickness.

When the sample has several layers, for determining each layer thickness, the critical angle θc_j should be determined for each layer. The data are plotted as a function of each $(\theta^2-\theta {c_j}^2)^{1/2}/\lambda$ abscissa. Fourier analysis is done for each curve to determine each layer thickness d_j.

APPLICATION TO SiO_2/Si THIN FILMS [16]

The experiment was done using synchrotron X-rays on beam line 4A at the Photon Factory, in order to use tunable monochromatic X-rays with sufficiently high photon flux density. The apparatus for X-ray reflectivity measurements is shown in the inset of Fig.2(a), and is essentially the same as that described in our previous work.[7-9] Synchrotron radiation was monochromatized by a Si(111) double-crystal sagittal focusing monochromator. Reflectivity ranging from 1 to $\sim 10^{-5}$ was measured by detecting the intensities of incident and reflected X-rays with two ionization chambers. Optical alignment was optimized by the translational and rotational motion of the sample stage. The measurement was made in air.

Figure 2(a) shows the experimental results of reflectivity of 8 keV X-rays for a SiO_2/Si sample, which were prepared by the conventional thermal oxidization process. The thickness of the SiO_2 layer obtained from ellipsometry was 501Å. The critical angle θc was 3.81 mrad, which was determined within 0.1 mrad by the direct comparison with the calculated reflectivity curve for SiO_2, and this accuracy is sufficient in this case.

The data were analyzed using the scheme shown in Fig.1. Figure 2(b) shows the magnitude of Fourier transform. A single sharp peak was clearly obtained, which gives the frequency of the oscillation observed in Fig.2(a). It is important that the peak position (499.5Å) agrees well with the thickness of the SiO_2 layer (501Å). The differences are less than 1 %.

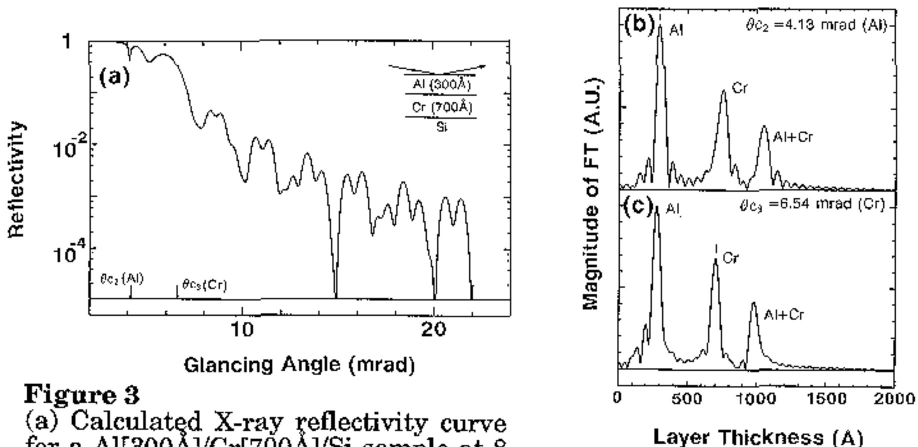

Figure 3
(a) Calculated X-ray reflectivity curve for a Al[300Å]/Cr[700Å]/Si sample at 8 keV. (b), (c) Magnitude of the Fourier transform of the oscillation extracted from Fig.3(a), after plotting as a function of $(\theta^2-\theta {c_j}^2)^{1/2}/\lambda$; (b) for θc_2=4.13 mard, (c) for θc_3=6.54 mrad.

CALCULATED RESULTS FOR FOUR-LAYERED EXAMPLE

The advantages of this technique will become more apparent when applied to cases where the number of layers is increased. Figure 3(a) shows the calculated reflectivity of 8 keV X-rays for a Al[300Å]/Cr[700Å]/Si sample. The curve seems rather complicated owing to the mixture of several frequency components. The present technique is feasible for determining both aluminium and chromium layer thickness.

First, the data are plotted as a function of $(\theta^2-\theta c_2^2)^{1/2}/\lambda$ abscissa (θc_2, critical angle for aluminium layer is 4.13 mrad) and $(\theta^2-\theta c_3^2)^{1/2}/\lambda$ abscissa (θc_3, for chromium layer, is 6.54 mrad). Then, the oscillation part of each curve are extracted and Fourier transformed. Figures 3(b) and (c) show the Fourier transformed results for each curve, respectively. In each figure, three peaks are clearly seen. They qualitatively correspond to the thickness of the aluminium layer, chromium layer and their sum. A peak corresponding to their differences is too weak to be observed, since the coefficient D in eq.(2) is quartic. Accurate peak position corresponding to thickness d_2 (for aluminium layer) and d_3 (for chromium layer) is just given in the data obtained through the scheme using θc_2 and θc_3, respectively. Both d_2 and d_3 determined to be 301.3Å and 696.7Å from Fig.3(b) and (c), respectively, are in good agreement with the assumed thicknesses.

SUMMARY

In conclusion, Fourier transform is effective in the analysis of the interference oscillation of reflectivity observed in grazing incidence X-ray experiments. The peak position in the Fourier space was in good agreement with the layer thickness. The present technique is feasible for the analysis of complicated oscillating structure obtained from multi-layered thin films. Furthermore, combining Fourier analysis with least-squares curve fitting procedure is useful for attaining interface information, i.e., interface roughness or sharpness.

Information on the present computer programs (PASCAL codes) are available from the authors. They are designed to work on a personal computer (NEC PC-9801), but it is not so difficult to translate them for other machines.

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