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Fourier Analysis of Interference Structure in X-Ray Specular Reflection from Thin Films

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Interference oscillation observed in X-ray total external reflection from thin films was analyzed by the Fourier transform algorithm. The peak position in Fourier space was in good agreement with the layer thickness, and was determined independently from the surface/interface roughness. The principle of the present technique and its application to SiO_2/Si thin films are shown. The advantages of the experiments using tunable synchrotron X-rays are also discussed.

KEYWORDS: X-ray total reflection, interference oscillation, multilayered thin films, Fourier analysis, X-ray reflectivity, grazing incidence, film thickness determination, synchrotron radiation, SiO_2 thickness

The oscillation of X-ray specular reflectivity often observed from multilayered thin films has been known as the Kiessig structure since the early days of X-ray physics.[1-5] Due to interference caused by multiple reflections of X-rays at each interface, the X-ray electric field in the matter is strongly modulated. Consequently, the reflectivity and the signal proportional to the amplitude of the electric field (for example, fluorescent X-rays, electron yield) show a complicated angular/energy dependence.

From the perspective of analysis of thin films, it is convenient to use this interference oscillation for determining each layer thickness, density and the interface roughness. When thin films do not have periodic layered structures, it is often difficult to analyze them by X-ray diffractometric procedures,[6,7] which are poweful tools in the analysis of semiconductor super-lattices and other multilayers. Interference oscillation in X-ray specular total reflection is feasible for such cases.

In the analysis of the oscillating structure in X-ray specular reflection, the least-squares curve fitting procedures using theoretical models have usually been employed.[8,9] 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 order to reduce the number of parameters, it is important to use conspicuous graphical features such as periods of the oscillation, which directly relate to layer thicknesses. In fact, in pioneering works in the 1950s,[4,5] the period of the interference fringe observed in the reflectivity was examined to determine film thickness or density, though the practical application was extremely limited.

In the present work, Fourier analysis of the interference structure is proposed to determine each layer thickness of multilayered thin films, independent of surface/interface roughness. The present technique permits frequency analysis to be done numerically, and therefore, much more accurate determination is expected than in conventional analysis such as direct reading of the positions of maxima.[4,5] In this letter, first, the background of the present method is discussed, and then, its application to SiO_2/Si thin films is shown. Finally, the advantages of energy tunability of synchrotron radiation (SR) are discussed.

The principle of the present technique is that the interference oscillation observed in the angular dependence of the reflectivity is essentially expressed as the sum of cosine function. The reflectivity from a multilayered thin film is usually calculated using a recursive equation.[3,10] For the sake of simplicity, a three-layered model (i.e., a single film layer besides air and the substrate) without absorption is considered here. Reflectivity R is written as

$$\begin{aligned} R &= |R_{1,2}|^2 \\ &= |(R_{2,3}+F_{1,2})/(R_{2,3}F_{1,2}+1)|^2 \\ &= (\exp(-i\gamma)F_{2,3}+F_{1,2})^2/(\exp(i\gamma)F_{2,3}F_{1,2}+1)^2 \qquad (1) \\ \gamma &= 4\pi d(\sqrt{\theta^2-\theta_c^2}/\lambda), \end{aligned}$$

where $R_{j-1,j}$ 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 and θ_c are the glancing angle and critical angle, respectively; λ is the wavelength of the X-rays; and d is the layer thickness. This equation is rewritten, when R is small:

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

That is, R has the form of $(B+C\cos\gamma)/A$, where A, B and C are the non-oscillating terms. It is important that γ is written as the product of $\sqrt{\theta^2-\theta_c^2}/\lambda$ and d. This indicates that the data plotted as a function of $\sqrt{\theta^2-\theta_c^2}/\lambda$ are converted to the distribution of d by Fourier transform, after the extraction of cosine oscillating parts. Accordingly, a single peak, whose position gives the layer thickness, is obtained in Fourier space. When considering the absorption effect in eq. (1), the correction term should be included in γ. However, it is negligible for small R, and therefore eq. (2) is used for the case with absorption. When the layer number is increased, the reflectivity contains the frequency component corresponding

to each layer thickness. It can be analyzed in a similar manner using the critical angle of each layer, with some exceptions owing to the fact that the terms corresponding to the sum of the thicknesses of neighboring layers are included in the equation.

The experiment was performed using SR on beam line 4A at the Photon Factory, in order to use tunable monochromatic X-rays with sufficiently high flux density. The apparatus for X-ray reflectivity measurements is shown in the inset of Fig. 1, and is essentially the same that described in our previous work.[11-13] SR beams were monochromatized by a Si(111) double-crystal sagittal focusing monochromator. The beam size was adjusted so as to obtain appropriate incident intensity, and was typically 2.5 mm wide and 0.07 mm high. 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/rotational motion of the sample stage. The measurement was made in air.

The samples used were SiO_2/Si thin films prepared by the conventional thermal oxidization process. The thickness of the SiO_2 layer was determined by ellipsometry. Figure 1 shows the experimental results of reflectivity of a SiO_2[501 Å]/Si sample at 8.0 keV. The critical angle θ_c at this energy is 3.81 mrad. The amplitude of oscillation observed above the critical angle is weak owing to the rather small $F_{1,2}$, i.e., Fresnel coefficient of SiO_2 and Si media, but is clearly visible.

The reflectivity measured is then expressed as a function of $\sqrt{\theta^2-\theta_c^2}/\lambda$, and is shown in Fig. 2(a). In our study, θ_c in the experimental data was determined from the calculated reflectivity curve for SiO_2. θ_c was determined within 0.1 mrad, and this accuracy is sufficient in this case. To compensate for the attenuation of the amplitude in the higher angle region, the data are normalized by the average curve (see the broken line in Fig. 2(a)) obtained from polynomial equation fitting in the logarithmic plot. After that, the non-oscillating background is removed. Thus, the oscillation is extracted from the experimental reflectivity and is Fourier transformed.

Figure 2(b) shows the magnitude of Fourier transform. A single sharp peak was clearly obtained, which gives the

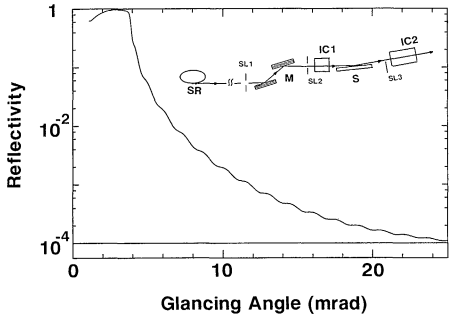

Fig. 1. Interference structure of SiO_2[501 Å]/Si in the 8 keV X-ray specular reflection. Schematic drawings of the experimental arrangement are shown in the inset; M: monochromator; IC1, IC2: ionization chambers for incident and reflected X-rays, respectively; S: sample; SL1, SL2, SL3: slits.

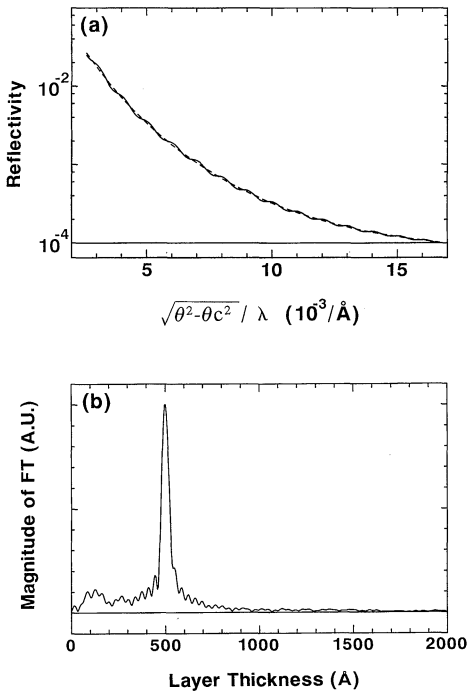

Fig. 2. (a) Reflectivity from SiO_2[501 Å]/Si expressed as the function of $\sqrt{\theta^2-\theta_c^2}/\lambda$. The broken line shows the average curve in the logarithmic plot of the data. (b) Magnitude of the Fourier transform of the oscillation extracted from Fig. 2(a). The peak indicates the frequency component of the oscillation and agrees with the film thickness.

frequency of the oscillation. It is significantly important that the peak position agrees well with the thicknesses of the SiO_2 layer. Thin films with various SiO_2 thickness were also measured and analyzed. The results are summarized in Table I. They are in good agreement with the thickness determined by ellipsometry, and the differences are around 1%, in spite of the rather weak interference oscillation of SiO_2/Si film. It was estimated that this technique can determine SiO_2 thickness of about one hundred to several thousand Å.

The dependence of interference oscillation on incident X-ray energy was investigated using the Cr_2O_3/Si sample, and is shown in Fig. 3. The film thickness is determined to be 1665 Å by the present technique. The results are essentially not dependent on the X-ray energy, since the period of oscillation is the same in terms of $\sqrt{\theta^2-\theta_c^2}/\lambda$. However, the amplitude of oscillation is sensitive to the energy and is significantly enhanced when measured at 5.5 keV, i.e., below the chromium absorption edge. This demonstrates the advantage of tuning X-ray energy. Small absorption is desirable, since absorp-

Table I. Comparison of thickness data obtained using ellipsometry and the present technique.

Ellipsometry d_1 [Å]	Present method 8 keV X-rays d_2 [Å]	$(d_2-d_1)/d_1$ [%]
195	192.5	0.30
501	499.5	0.30
988	972.5	1.57

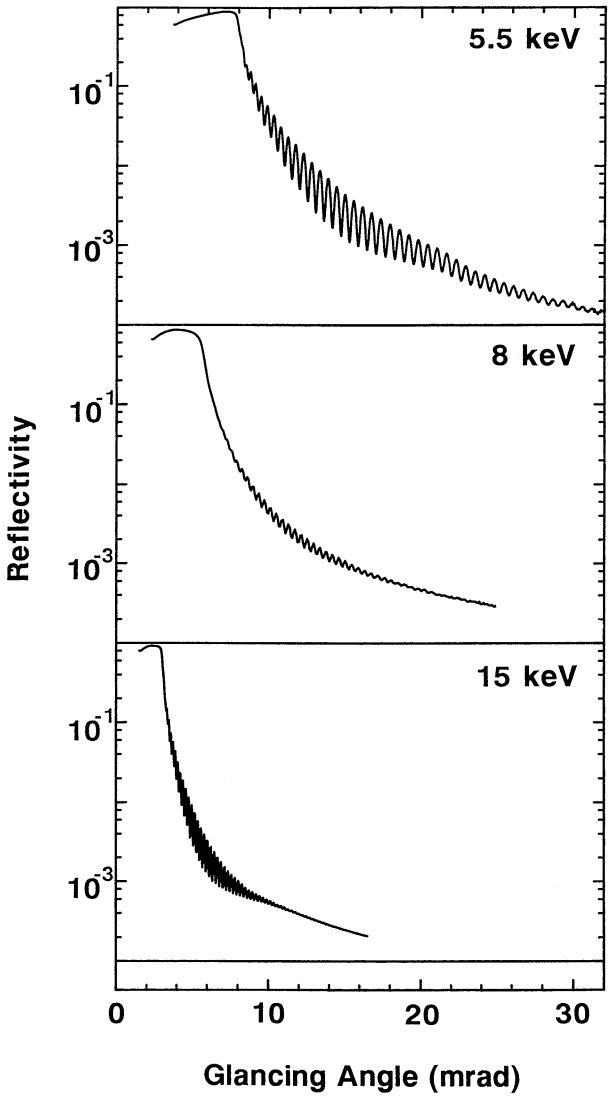

Fig. 3. The incident X-ray energy dependence of the reflectivity data from Cr_2O_3/Si.

tion smears the interference oscillation. However, for experiments using higher energy X-rays, careful positioning of the sample and higher angular resolution of the system are required because of the extremely small critical angle and the short oscillation period in terms of the glancing angle. The present results demonstrate that tuning the X-ray energy below the absorption edges of elements of film is favorable. The choice of X-ray energy is important in the analysis of the practical complex reflectivity curve. This is furnished by the continuum spectrum with sufficient X-ray intensity of SR.

The present results show that Fourier transform is effective in the analysis of the interference oscillation observed in the X-ray specular reflectivity. The advantages of this technique will become more apparent when applied to cases where the number of layers is increased. For such cases, since several frequency components are included in the reflectivity oscillation, the curve often seems complicated. Fourier analysis is advantageous to separate the frequency components, and consequently the thickness of each layer can be determined. Furthermore, combination with the least-squares curve fitting procedure is useful for attaining interface information; i.e., interface roughness or sharpness. Besides the layer thickness determination, this technique might be convenient for detecting the change of the density at surface/interface, for example, formation of a new layer by diffusion or any reaction. Frequency analysis employed in the present study is also useful for such determinations.

Acknowledgments

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