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Calculation of Feasibility of Grazing-Incidence X-Ray Fluorescence as a Method for Holographic Analysis

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Grazing incidence X-ray fluorescence (GIXF) was used to obtain information on composition as a function of depth. Glancing-angular dependence of the fluorescence intensity from a layered sample typically shows Kiessing fringe similar to the X-ray reflectivity curves, and it can be explained by theory of the atomic resolution X-ray holography, which is recently developed structural analysis method, where fluorescence intensities are measured as a function of direction of a crystal sample. In the GIXF, X-rays directly reaching at a layer emitting fluorescence are regarded as reference waves in holography, and X-rays singly reflected at interfaces act as object waves. Therefore, the angular dependence of fluorescence intensity is considered to be a hologram. The aim of the present work is the real space reconstruction from the angular dependence of the fluorescence intensity obtained by the calculation. Here, I made a multilayer model whose top layer was monatomic, and calculated the angular dependence of the fluorescence intensity from the top layer. The calculated pattern showed holographic oscillation. In the Fourier transform of the oscillation, there exists peaks corresponding to the depths of the interfaces in the multilayer, revealing that the present technique can provide film structure information without a priori knowledge.
Key words: grazing incidence X-ray fluorescence, X-ray holography, multilayer, Fourier transformation

1. INTRODUCTION

When X-rays impinge on a flat material under a small glancing angle, a large variety of physical phenomena can occur, such as total reflection, interface fringes from a periodic multilayer.[1] Grazing incidence X-ray fluorescence (GIXF) analysis was one of methods using these phenomena, and has been used to obtain information on composition as a function of depth.[2,3] Glancing angle variation of the fluorescence intensity from a layered sample typically shows Kiessing fringe due to the interference effect between direct and reflected X-rays from the interface. Of course, this effect appears in the ordinary X-ray reflectivity curves.[4] Sakurai et al. proposed to apply the Fourier transformation to the oscillations in X-ray reflectivity, and successfully obtained the thickness values of layers in the measured film sample.[5] Taking into account this analogy, it is considered that the oscillation in the fluorescence intensity can also provide information on the film structure.

In this report, I modeled a Fe/Si/Ti/Ag/Au multilayer, whose top Fe layer was monatomic, and calculated the glancing-angular variation of the Fe fluorescence intensity. The oscillation extracted from the fluorescence intensity was Fourier transformed to obtain film structure without a priori knowledge.

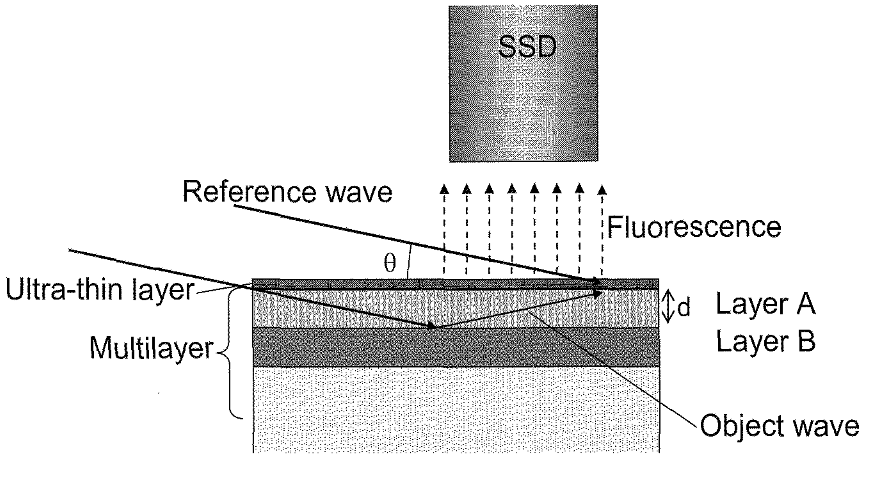

Fig.1 Scheme of generation of holographic signal from layered material under grazing incidence

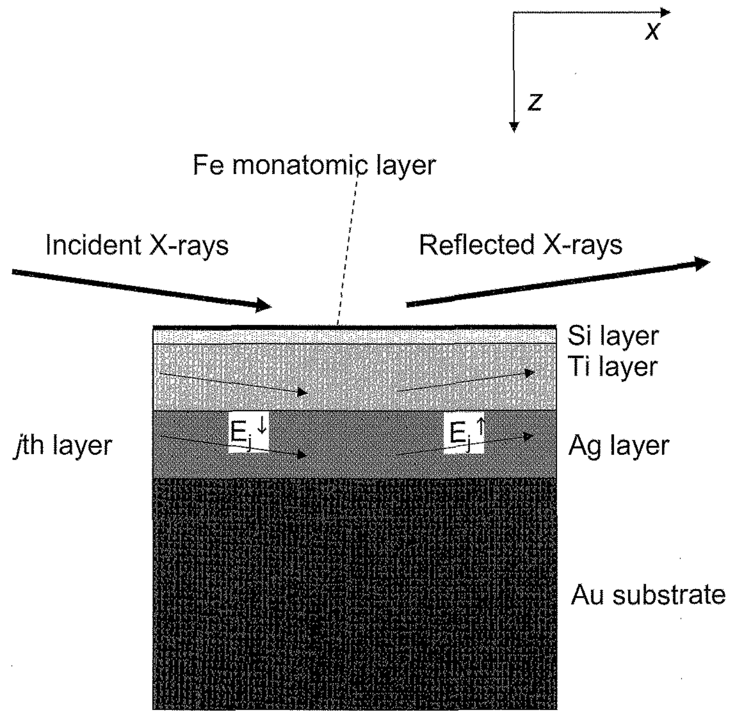

Fig.2 Illustration of the multilayer model employed to calculate X-ray fluorescence yield.

2. THEORY

Atomic resolution X-ray holography is a recently developed structural analysis technique, where fluorescence intensities are measured as a function of direction of a crystal sample.[6] The glancing-angular dependence of the fluorescence intensity can be explained using the holography theory. Figure 1 shows the illustration of the generation of the holographic signal from the layered material under the grazing incidence condition. X-rays directly reaching at a layer (ultra-thin layer in Fig.1) emitting fluorescence are regarded as reference waves in holography, and X-rays singly reflected at interfaces act as object waves. These reference and object waves interfere each other and form X-ray standing wave. X-ray intensity at the ultra-thin layer is modulated with the glancing angle of the incident X-ray beam, and the fluorescence intensity from the ultra-thin layer is proportional to it.

Let's discuss the X-ray intensity due to the X-ray standing wave. The path difference between the reference and object wave is briefly expressed as,

$$\Delta l = 2d \cdot \sin\theta \cong 2d\theta, \quad (1)$$

where the θ is the glancing angle of the incident X-rays above the critical angle for total reflection θ_c, and the d is the depth of the interface. Thus, the fluorescence intensity from the top layer oscillates with the phase $2d\theta/\lambda$, where λ is the wavelength of the incident X-rays. Therefore, the angular dependence of fluorescence intensity is considered to be a hologram.

3. RESULTS AND DISCUSSION

As shown in Fig.2, I made an Fe/Si/Ti/Ag/Au multilayer model whose Fe top layer was monatomic (0.2 nm), where the thicknesses of the Si, Ti and Ag layers were 5 nm, 50 nm and 80 nm, respectively. The densities of the Fe, Si, Ti, Ag and Au layers were 7.86, 2.35, 4.50, 10.5 and 19.3 g/cm^3, respectively, which were same to the bulk crystal ones. The roughness at each surface and interface was assumed to be 0.5 nm. The X-ray energy for the calculation was 10.0 keV (0.124 nm). The X-ray reflectivity curve and the angular dependence of the fluorescence intensity of Fe K lines were calculated using the formalism described in Ref. [3].

I briefly explain the way of the X-ray fluorescence yield calculation using Fig.2. In jth layer of the multilayer sample, the vector of refraction is assumed to be $\mathbf{N}_j$. For all j, the x component of N_j N_{jx} is $\cos\theta$. The z component $\mathbf{N}_j$ is

$$N_{jz} = (\varepsilon_j - \cos^2\theta)^{1/2}, \quad (2)$$

where ε_j is the complex dielectric constant of material j.

Electric fields of the transmitted and reflected X-rays are respectively expressed as

$$E_j^{\downarrow} = E_j^t \exp(-i\frac{2\pi}{\lambda}N_{jz}z)\exp(i(\omega t - \frac{2\pi}{\lambda}N'_{jx}x)) \quad (3)$$

and

$$E_j^{\uparrow} = E_j^r \exp(i\frac{2\pi}{\lambda}N_{jz}z)\exp(i(\omega t - \frac{2\pi}{\lambda}N'_{jx}x)), \quad (4)$$

where E_j^t and E_j^r are the transmitted and reflected fields at the top layer j, and ω is the frequency of the X-rays and t is the time. The values of E_j^t and E_j^r can be calculated using the recursion formula described in Ref [7]. Total electric field in layer j is the sum of the transmitted contribution $E_j^{\downarrow}$ and a reflected contribution $E_j^{\uparrow}$:

$$E_j = E_j^{\downarrow} + E_j^{\uparrow}. \quad (5)$$

Here, P_{jz} is the z component of the Poynting vector. The $-\partial P_{jz}/\partial z$ is proportional to $|E_j^{\downarrow} + E_j^{\uparrow}|^2$, and is

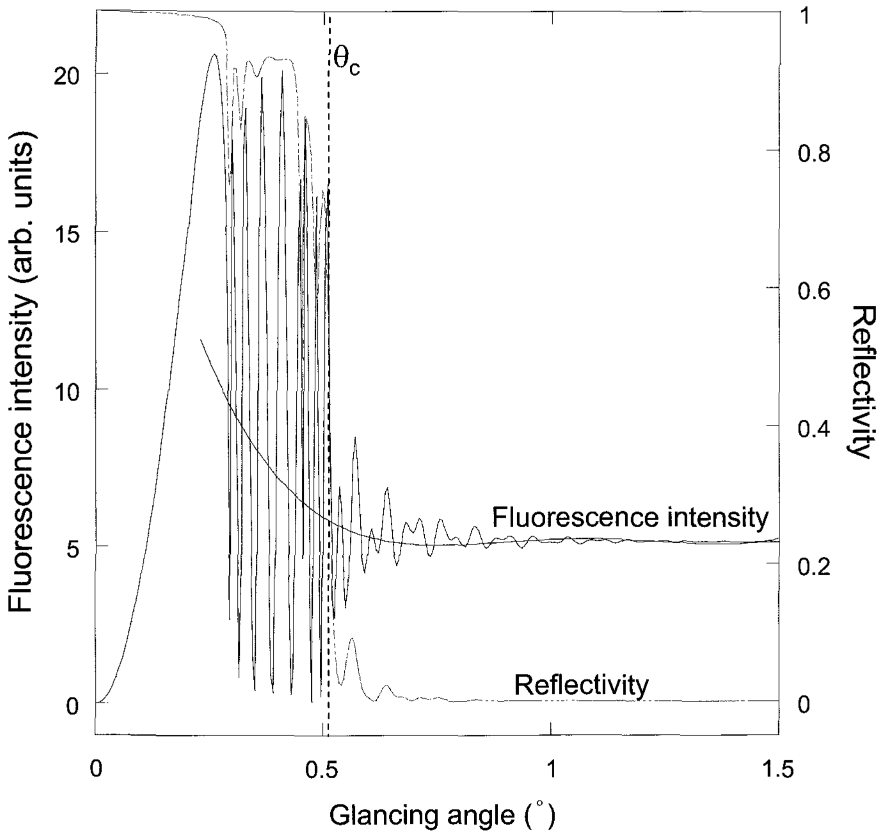

Fig.3 Angular dependence of the Fe K fluoresce intensity and reflectivity curves.

expressed precisely in Ref. [3]. The fluorescence intensity I_f is obtained using the equation of

$$I_f = C \int_{d_{j-1}}^{d_j} dz(-\frac{\partial P_{jz}}{\partial z})\exp(-\mu_f z), \quad (6)$$

where d_j is the depth of the interface between jth and j+1th layers, C is the constant and μ_f is the penetration depth of the fluorescent X-rays. In the present case, μ_f can be replaced by 1, because I assume to detect the fluorescence from 0.2 nm thick top layer.

Using Eq. (6), I calculated the fluorescence intensities from the top Fe layer with the change of the glancing angle of the incident X-rays, and they are plotted in Fig. 3. The fluorescence pattern exhibits clear fringe patterns. The fringe above the critical angle for total reflection θ_c (= 0.515°) is regarded as the holographic oscillation. Thus, this holographic oscillation was obtained by subtracting the baseline intensity from the fluorescence intensity in the range of $\theta > \theta_c$. The baseline, which is depicted in Fig. 3, was determined by polynomial equation fitting method.

This holographic oscillation was Fourier transformed as shown in Fig. 4. We can see markedly three peaks corresponding to the depths of the interface in the multilayer. Positions of these three peaks estimated from the Fourier transformation were 2.8, 51.2 and 83.0 nm, respectively. The depth value of 2.8 nm, which corresponds to Si/Ti interface, is half of the actual thickness of Si layer. While, the depth values at Ti/Ag and Ag/Au interfaces were 1 – 3 nm larger than those of the Si/Ti and Ti/Ag interfaces in the model multilayer. It is considered that these discrepancies originate from the refraction of X-rays at the surface and interfaces. The construction of the theory including the refraction phenomenon is further task.

4. CONCLUSION

The holographic analysis of the grazing incidence X-ray fluorescence data has been discussed through calculations of the angular dependence of the X-ray fluorescence intensity of Fe K lines from the Fe/Si/Ti/Ag/Au multilayer model, and successfully reconstructed depth positions at the interface from the oscillation in its angular dependence using the Fourier transformation. This method has a potential to analyze film structure without a priori knowledge

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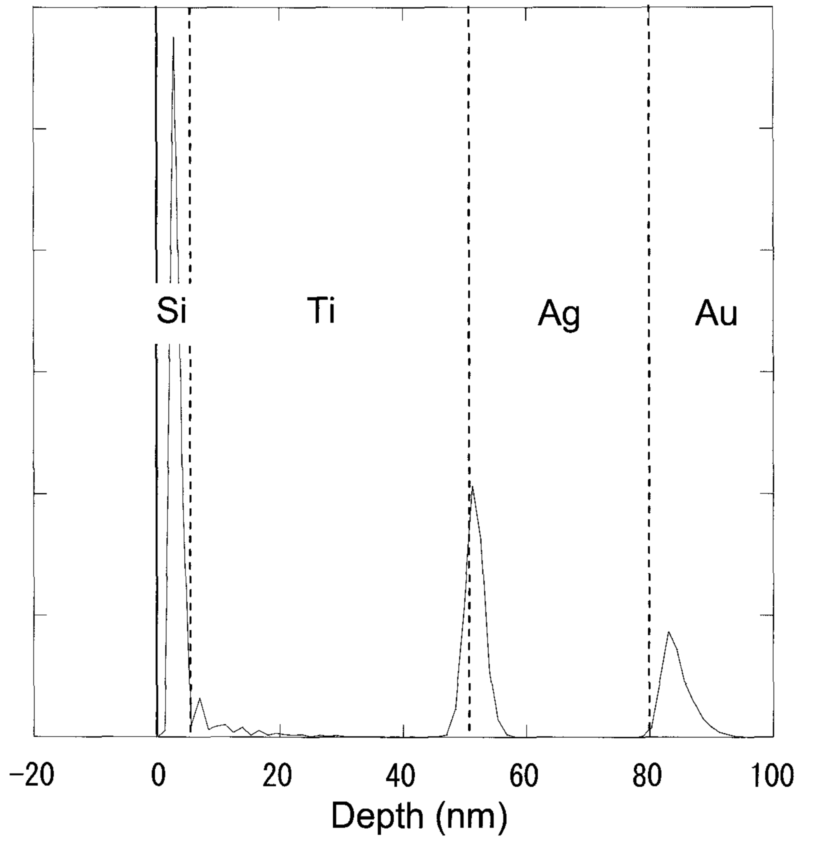

Fig.4 Fourier transform of holographic oscillation in angular variation of the Fe K fluorescence intensity.

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