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Improvement of X-Ray Reflectivity Analysis on Surface and Interface Roughness Estimation

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Abstract: X-ray reflectivity (XRR) is usefull tool to estimate surface and interface roughness. In the conventional XRR analysis, the reflectivity is calculated based on the Parratt formalism, accounting for the effect of roughness by the theory of Nevot-Croce. However, the calculated results have shown often strange behavior due to the fact that the diffuse scattering at the rough interface was not taken into account in the equation. Then we developed new improved formalism to correct this mistake. For deriving more accurate formalism of XRR, we tried to compare the measurements of the surface roughness of the same sample by atomic force microscopy (AFM) and XRR. The results of analysis show that the effective roughness measured by xrrmay depend on the angle of incidence. In this paper, it shows that new improved XRR formalism which derives more accurate surface and interface roughness with depending on the size of the probing area of coherent X-rays.

Keywords: X-Ray Reflectivity, Surface and Interface Roughness, Multilayer Surfaces, Buried Interface

1. Introduction

X-ray reflectivity (XRR) is usefull tool to estimate surface and interface roughness, whichis of prime importance in many applications, such as microelectronics.[1-15] In the XRR analysis, the x-ray reflectivity is usually calculated based on the Parratt formalism,[1] coupled with the use of the theory of Nevot and Croce to include the effect of surface and interface roughness.[2]

However, the calculated results have shown often strange behavior due to the fact that the diffuse scattering at the rough interface was not taken into account in the equation. Then we developed new improved formalism to correct this mistake.[12-14] For deriving more accurate formalism of XRR, we tried to compare the measurements of the surface roughness of the same sample by atomic force microscopy (AFM) and XRR.[15]

In the study, we examined two samples of silicon wafers having a thin sio_2 layer were prepared by the following methods. The sample A was prepared by thermal oxidizing of a Si(001) wafer. The thickness of the prepared sio_2 layer is about 5 nm. The other sample B was prepared by vacuum deposition of an additional sio_2 layer of about 2 nm on the sample A at room temperature. The roughness of the sio_2/Si interface was expected to be the same as the sample A although the surface roughness should be increased after the deposition.

The surfaces of these samples were measured by XRR and AFM.By the AFM observations, we found that the r.m.s. Roughness σ_s at the area of $1 \times 1\ \mu m^2$ of the sio_2 surfaces of sample A and sample B were about 0.17 nm both, and those at the area of $10 \times 10\ \mu m^2$ were about 0.24 nm both. The surface roughness by AFM observation depended on the observation area.[15]

X-ray reflectivity measurements were performed using a Cu-KαX-ray beam from a 3 kw rotating-anode source. The beam size of the x-ray was about 2 mm (perpendicular to the reflection plane) × 0.05 mm (parallel to the reflection plane). The results of the x-ray reflectivity measured for the sample A and B were analysed, and the surface roughness, interface roughness and the thickness of the sio_2 layer were estimated.In comparison with AFM and XRR for the sample A and B, the results of surface roughness by XRR were different with the AFM observations for the both samples. The surface roughness estimated from AFM observation showed small value with those of x-ray reflectivity and smaller at the area of $1 \times 1\ \mu m^2$ than at the area of $10 \times 10\ \mu m^2$.

This suggested that the value of roughness measured by the measurement range might be different in the x-ray reflectivity measurements. And in the x-ray reflectivity measurement, the measurement range changed by an incidence angle, and changed very much at small glancing incidence angle. It

suggested that the effective roughness depending on an incidence angle in XRR calculation should be assume.

In this paper, we show the new improved XRR formalism which derives more accurate surface and interface roughness with depending on the size of the probing area of coherent X-rays.

2. X-Ray Reflectivity Analysis

X-ray reflectivity measurements were performed using a Cu-Kα x-ray beam from a 3 kw rotating-anode source. The beam size of the x-ray was about 2 mm (perpendicular to the reflection plane) × 0.05 mm (parallel to the reflection plane). The experimental detail was shown in previous study [15], then in this paper, only the result of the x-ray reflectivity measured for the sample B and its analytical results are shown in Fig. 1.

In the conventional x-ray reflectivity analysis, the reflectivity is calculated based on the Parratt formalism [1], incorporating the effect of the interface roughness according to the theory of Nevot-Croce[2] as ,

$$R = \left|R_{0,1}\right|^2 ,$$
$$R_{j-1,j} = \frac{\Psi_{j-1,j} + R_{j,j+1}}{1 - \Psi_{j,j-1} R_{j,j+1}} \exp(2ik_{j-1,z} h_{j-1}),\ R_{N,N+1} = 0, \tag{1}$$

Recently, we have found that the conventional formula gives strange results when the interface roughness increases, i.e. the amplitude of the oscillation becomes larger with increasing interface roughness[12-14]. These results were attributed to the fact that the diffuse scattering at the rough interface was not correctly taken into account in the conventional formula by Nevot and Croce. Then we have developed a new formula in which the effects of the surface and interface roughness are correctly treated[12-14]. The x-ray reflectivity R of a multilayer sample consisting of N layers is given by

$$R = \left|R_{0,1}\right|^2 ,$$
$$R_{j-1,j} = \frac{\Psi_{j-1,j} + (\Phi_{j-1,j}\Phi_{j,j-1} - \Psi_{j-1,j}\Psi_{j,j-1}) R_{j,j+1}}{1 - \Psi_{j,j-1} R_{j,j+1}} \exp(2ik_{j-1,z} h_{j-1}), \tag{2}$$
$$R_{N,N+1} = 0,$$

where $R_{\text{j-1,j}}$ is the reflection coefficient at the interface of j-1-th layer and j-th layer, h_j is the thickness of the j-th layer, $h_0 = 0$, $k_{\text{j, z}}$ is the z component of the wave vector in the j-th layer, and $\Psi_{\text{j-1, j}}$ and $\Phi_{\text{j-1, j}}$ are the Fresnel coefficients for reflection and refraction, respectively, at the interface between the (j-1)th layer and the j-th layer. Although the formula for $\Psi_{\text{j-1, j}}$ is well known

$$\Psi_{j-1,j} = \frac{k_{j\text{-}1,z} - k_{j,z}}{k_{j\text{-}1,z} + k_{j,z}} \exp\left(-2k_{j-1,z}k_{j,z}\sigma_{j-1,j}{}^2\right),$$
$$\Psi_{j,j-1} = -\Psi_{j-1,j}, \tag{3}$$

where $\sigma_{j\text{-}1,\,j}$ is the interface roughness between (j-1)-th and j-th layers, an accurate analytical formula for $\Phi_{\text{j-1, j}}$ including the effect of the interface roughness is not available. There are several approximations proposed for $\Phi_{\text{j-1, j}}$ and all these results can be written as

$$\Phi_{j-1,j} = \frac{2k_{j\text{-}1,z}}{k_{j\text{-}1,z} + k_{j,z}} \exp\{-[C_1(k_{j-1,z} - k_{j,z})^2 + C_2 k_{j-1,z}k_{j,z}]\sigma_{0,1}{}^2\},$$
$$\Phi_{j,j-1} = \Phi_{j-1,j}\frac{k_{j,z}}{k_{j\text{-}1,z}} \tag{4}$$

where parameters C_1, C_2 depend on the approximation [3-14]. In the previous work, we choose $C_1 = 2$ and $C_2 = 0$ as the most appropriate approximation.[12-14]

After analyzing the XRR results, the sio_2 layer profiles of sample B were derived. From the period of the oscillation, the thickness of the sio_2 layer was determined to be 7.8 nm. Because the deposition of the additional sio_2 layer of 2 nm does not change the interface roughness we used the interface roughness determined for the sample A (σ_i = 0.42 nm) in the estimation of the surface roughness of the sample B. Using these values (σ_i = 0.42 nm and the thickness 7.8 nm) the reflectivity was calculated with various values of σ_s.

Figure 1 shows the comparison between the calculated and experimental results. None of the calculated results can reproduce the experimental one. At $\theta_i > 1.0°$ the calculated result for σ_s = 0.54 nm agrees with the experimental one while the calculated result deviates from the experimental one at smaller θ_i. On the other hand, the calculated result for σ_s = 1.08 nm agrees with the experimental one at smaller θ_i but it deviates seriously with increasing θ_i. [15]

A possible explanation of the present discrepancy may be that the effective surface roughness measured by XRR depends on the size of the effective probing area on the surface, which is proportional to $1/\sin\theta_i$. In general, the surface roughness increases with increasing size of the probing area.

As a result, the effective roughness observed at smaller θ_i is larger than that at larger θ_i in accordance with the present result. Such a θ_i-dependence of the effective roughness in XRR has been usually neglected. The present result, however, indicates that it should be taken into account of which the effective roughness depends on the size of the probing area of coherent X-ray.

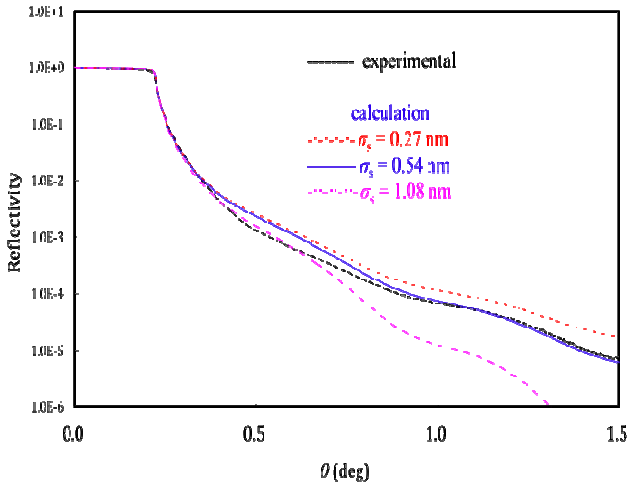

Figure 1. *X-ray reflectivity from sio₂/Si. The experimental result (thick dashed curve) is compared with the calculated ones for σ_i = 0.42 nm and various σ_s (thin curves).*

3. Discussion

We show again the Fresnel coefficient $\Psi_{j-1,\,j}$ for reflection and the Fresnel coefficient $\Phi_{j-1,\,j}$ for refractionas,

$$\Psi_{j-1,j}=\frac{k_{j-1,z}-k_{j,z}}{k_{j-1,z}+k_{j,z}}Q_{j-1,j}\,,\ \Psi_{j,j-1}=-\Psi_{j-1,j} \qquad (5)$$

$$\Phi_{j-1,j}=\frac{2k_{j-1,z}}{k_{j-1,z}+k_{j,z}}P_{j-1,j}\,,\ \Phi_{j,j-1}=\Phi_{j-1,j}\frac{k_{j,z}}{k_{j-1,z}} \qquad (6)$$

where $Q_{j-1,\,j}$ and $P_{j-1,\,j}$ are the reduce factor due to the roughness, and used the following approximations formula as,

$$Q_{j-1,j}=\exp\left(-2k_{j-1,z}k_{j,z}{\sigma_{j-1,j}}^2\right), \qquad (7)$$

$$P_{j-1,j}=\exp\{-[C_1(k_{j-1,z}-k_{j,z})^2+C_2k_{j-1,z}k_{j,z}]{\sigma_{j-1,j}}^2\}, \qquad (8)$$

where parameters C_1, C_2 depend on the proposed approximation as above.

In the previous work(Sinha et al., 1988; Boer, 1995), X-ray scattering from rough surface isstudied, and the effect of the roughness is explained as,

$$Q_{j-1,j}=\frac{\iint_{S_0}e^{-k_{j-1,z}k_{j,z}g(x,y)}e^{-i(q_xx+q_yy)}dxdy}{\iint_{S_0}e^{-i(q_xx+q_yy)}dxdy},\ q_x=k_{j,x}-k_{j,x}\ ,\ q_y=k_{j,y}-k_{j,y}\,, \qquad (9)$$

$$P_{j-1,j}=\frac{\iint_{S_0}e^{-1/4(k_{j-1,z}-k_{j,z})^2g(x,y)}e^{-i(q_xx+q_yy)}dxdy}{\iint_{S_0}e^{-i(q_xx+q_yy)}dxdy},\ q_x=k_{j-1,x}-k_{j,x}\ ,\ q_y=k_{j-1,y}-k_{j,y}\,, \qquad (10)$$

where $g(x,y)=\left\langle\left\{z(x'+x,y'+y)-z(x',y')\right\}^2\right\rangle$ is the square average of the height of the interface at (x'+x,y'+y) separated by (x,y) from (x',y').In the reflected X-ray and the refracted X-ray, q_x=q_y=0.The scattering plane, x-z plane, is considered in the analysis on X-ray reflectivity. Then the reduce factor $Q_{j-1,\,j}$and $P_{j-1,\,j}$are shown as,

$$Q_{j-1,j}=\frac{1}{L_x}\int_{L_x}e^{-k_{j-1,z}k_{j,z}g(x)}dx\,, \qquad (11)$$

$$P_{j-1,j}=\frac{1}{L_x}\int_{L_x}e^{-1/4(k_{j-1,z}-k_{j,z})^2g(x)}dx\,, \qquad (12)$$

where l_x is the length of the probing area of coherent X-ray. The square average $g(x)$ of the height of the interfaceis related to the roughness correlation function $C(x)$ as,

$$g(x)=2o^2-2C(x) \qquad (13)$$

Following Shinha et al. (Sinha et al., 1988), the roughness correlation function $C(x)$ of a fractal surfacehas the form as,

$$g(x)=2\sigma^2\left[1-\exp\left\{-\left(\frac{|x|}{\xi}\right)^{2H}\right\}\right], \qquad (14)$$

where Hurst parameter H $(0<H\le1)$ is connected to its fractal dimension, and the lateral correlation length ξ acts as a cutoff length for the fractal behavior of the surface. Note that we implicitly assumed that ξ is smaller than the coherence length L_x of the radiation parallel to the surface.

In eqs. (9) and (10), L_x depends on the angle θ_i of incident X-ray as

$$\frac{1}{L_x^2}=\frac{\sin^2\theta_i}{L_t^2}+\frac{\cos^2\theta_i}{L_l^2}\,, \qquad (15)$$

where l_t is transverse coherence length and L_l is longitudinal coherence length. Then we can define the effective roughness σ*at the angle θ_i of incident X-ray as the followin geqs.;

$$Q_{j-1,j} = \exp\left(-2k_{j-1,z}k_{j,z}{\sigma^*_{j-1,j}}^2\right)$$
$$= \frac{1}{L_x}\int_{L_x} \exp\left(-2k_{j-1,z}k_{j,z}{\sigma_{j-1,j}}^2\left[1-\exp\left\{-\left(\frac{|x|}{\xi_{j-1,j}}\right)^{2H}\right\}\right]\right)dx \quad (16)$$

$$P_{j-1,j} = \exp\left(-\frac{1}{2}(k_{j-1,z}-k_{j,z})^2{\sigma^*_{j-1,j}}^2\right)$$
$$= \frac{1}{L_x}\int_{L_x} \exp\left(-\frac{1}{2}(k_{j-1,z}-k_{j,z})^2{\sigma_{j-1,j}}^2\left[1-\exp\left\{-\left(\frac{|x|}{\xi_{j-1,j}}\right)^{2H}\right\}\right]\right)dx \quad (17)$$

$\xi << L_x$, g(x) becomes $2\sigma^2$, and the reduce factor $Q_{j\text{-}1,\,j}$ becomes Eq.(5).Based on the above considerations, we again calculated the X-ray reflectivity for the sio_2/Sisurface of sample B, but now consideredthe effective roughness σ^* at the X-ray incident angle θ_i. Figure 2 shows the calculated XRR with using the values (ξ_s = 2μm, L_t = 10nm, and L_l= 2μm). The calculated reflectivity shows good agreement with the experimental one in all range of measured θ_i.

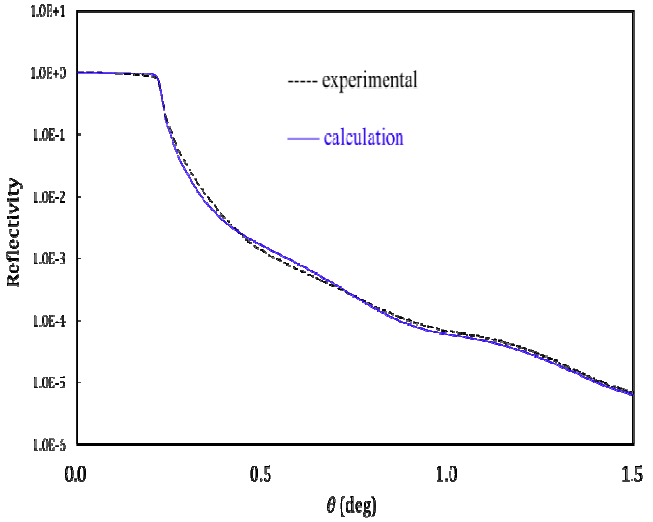

Figure 2. *X-ray reflectivity from sio_2/Si. The experimental result (thick dashed curve) is compared with the calculated ones usingthe effective roughness depending onthe X-ray incident angle θ_i. XRR is calculated with using the values (ξ_s = 2μm, L_t = 10nm, and L_l = 2μm).*

4. Conclusion

In concerned with the calculation of XRR, we consideredthe effective roughness with dependingon the incident angle of X-ray. At the result, it is showed the new improved XRR formalism which derives more accurate surface and interface roughness with depending on the size of the probing area of coherent X-rays.

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