TECHNICAL ARTICLE

Improvement of X-ray reflectivity calculations on a multilayered surface

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X-ray reflectometry is a powerful tool for investigating rough surface and interface structures. Presently, X-ray reflectivity is based on Parratt formalism, accounting for the effect of roughness by the theory of Nevot–Croce. However, the calculated results showed a strange phenomenon in that the amplitude of the oscillation because of interference effects increases in the case of a specific roughness of the surface. We propose that the strange results originated from the currently used equation because of a serious error in which the Fresnel transmission coefficient in the reflectivity equation is increased at a rough interface, and the increase in the transmission coefficient completely overpowers any decrease in the value of the reflection coefficient because of lack of consideration in diffuse scattering. In the present study, we present a new improved formalism that corrects this error, and thereby derives an accurate analysis of X-ray reflectivity from a multilayer surface, taking into account the effect of roughness-induced diffuse scattering. © *2013 International Centre for Diffraction Data*. [doi:10.1017/S0885715613000110]

Key words: X-ray reflectivity, Parratt formalism, theory of Nevot–Croce, surface roughness, diffuse scattering, multilayer surface and interfaces

I. INTRODUCTION

Grazing incidence X-ray scattering is a powerful tool for investigating rough surface and interface structures (Parratt, 1954; Nevot and Croce, 1980; Sinha *et al.*, 1988; Holy *et al.*, 1993; Daillant and Gibaud, 1999; Sakurai, 2009; Fujii *et al.*, 2004, 2005; Fujii, 2010, 2011). In many previous studies of X-ray reflectometry, X-ray reflectivity was based on Parratt formalism (Parratt, 1954), coupled with the use of the theory of Nevot and Croce to include roughness (Nevot and Croce, 1980). However, the calculated results of X-ray reflectivity often showed strange results where the amplitude of the oscillation because of the interference effects would increase for a rougher surface. These strange results disagree with reality and suggest that there is a problem in how the effect of roughness is incorporated into Parratt formalism, and hence the effect of roughness is not accurately represented in the theoretical formula of X-ray reflectivity. In our previous study, we proposed that these strange results originated because of neglect of the effect of a decrease in the intensity of refracted X rays because of diffuse scattering at a rough surface and interface (Fujii, 2010, 2011).

In the present study, we propose more clearly that the strange results originated from a currently used equation because of a serious error in which the Fresnel transmission coefficient in the reflectivity equation is increased at a rough interface. We investigate the calculated result of X-ray reflectivity based on Parratt formalism (Parratt, 1954) with the effect of the roughness incorporated by the theory of Nevot–Croce (Nevot and Croce, 1980), and present a new accurate formalism to correct the error.

II. X-RAY REFLECTIVITY CACULATIONS

The reflectance of an *N*-layer multilayer system can be calculated using the recursive formalism given by Parratt (1954). In the following, we show in detail the process of obtaining Parratt's expression and, further, show that this expression requires conservation of energy at the interface. We go on to show that the dispersion of energy by interface roughness cannot be correctly accounted for Parratt's expression.

A. X-ray reflectivity from a flat surface of a multilayer with flat interfaces

First, we consider the reflection from a flat surface of a multilayer with flat interfaces. We take the vertical direction to the surface as the *z*-axis, with the positive direction pointing towards the bulk. The scattering plane is made the *x*–*z* plane. Following that approach, let n_j be the refractive index of the *j*th layer. The wave vector $\boldsymbol{k}_j$ of the *j*th layer is related to the refractive index n_j of the *j*th layer by

$$\boldsymbol{k}_j \cdot \boldsymbol{k}_j = n_j^2 \boldsymbol{k}_0 \cdot \boldsymbol{k}_0, \quad \boldsymbol{k}_0 \cdot \boldsymbol{k}_0 = k^2 \qquad (1)$$

as this necessitates that the x, y-direction components of the wave vector are constant, then the *z*-direction component of the wave vector of the *j*th layer is

$$k_{j,z} = \sqrt{n_j^2 k^2 - k_{0,x}^2}. \qquad (2)$$

The electric fields $\boldsymbol{E}_j$, $\boldsymbol{E}'_j$ below the interface between the $j - 1$th layer and *j*th layer are expressed as

$$\boldsymbol{E}_j(z_{j-1,j}) = \boldsymbol{A}_j \exp[i(k_{j,x}x + k_{j,y}y - \omega t)],$$
$$\boldsymbol{E}'_j(z_{j-1,j}) = \boldsymbol{A}'_j \exp[i(k_{j,x}x + k_{j,y}y - \omega t)]. \qquad (3)$$

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The amplitudes A_j and A'_j at the jth layer are derived from the equations for the interface between the $j-1$ and j layers and the electric field variation within the jth layer with depth h_j as expressed by the following matrix:

$$\begin{pmatrix} A'_{j-1}\exp(-ik_{j-1,z}h_{j-1}) \\ A_j \end{pmatrix} = \begin{pmatrix} \Psi_{j-1,j} & \Phi_{j,j-1} \\ \Phi_{j-1,j} & \Psi_{j,j-1} \end{pmatrix}$$
$$\times \begin{pmatrix} A_{j-1}\exp(ik_{j-1,z}h_{j-1}) \\ A'_j \end{pmatrix}, \quad (4)$$

where $\Psi_{j-1,j}$ and $\Phi_{j-1,j}$ are the Fresnel coefficient tensors for reflection and refraction at the interface between the $j-1$ and j layers. For s-polarization, the Fresnel coefficients are

$$\Phi_{j-1,j,yy} = \frac{2k_{j-1,z}}{k_{j-1,z}+k_{j,z}},$$
$$\Phi_{j,j-1,yy} = \frac{2k_{j,z}}{k_{j-1,z}+k_{j,z}} \Psi_{j-1,j,yy} = \frac{k_{j-1,z}-k_{j,z}}{k_{j-1,z}+k_{j,z}},$$
$$\Psi_{j,j-1,yy} = \frac{k_{j,z}-k_{j-1,z}}{k_{j-1,z}+k_{j,z}}. \quad (5)$$

The reflection coefficient is defined as the ratio $R_{o,1}$ of the reflected electric field to the incident electric field at the surface of the material and is given by

$$A'_0 = R_{0,1}A_0. \quad (6)$$

The reflection coefficient $R_{j-1,j}$ of the electric field E'_{j-1} to the electric field E_{j-1} at the interface of $j-1$th layer and jth layer is

$$A'_{j-1} = R_{j-1,j}A_{j-1} \quad (7)$$

and the ratio $R_{j-1,j}$ is related to the ratio $R_{j,j+1}$ as follows:

$$R_{j-1,j} = \frac{\Psi_{j-1,j}+(\Phi_{j-1,j}\Phi_{j,j-1}-\Psi_{j-1,j}\Psi_{j,j-1})R_{j,j+1}}{1-\Psi_{j,j-1}R_{j,j+1}}$$
$$\times \exp(2ik_{j-1,z}h_{j-1}). \quad (8)$$

Here, from the relation between the Fresnel coefficient for reflection and the Fresnel coefficient for refraction,

$$\Phi_{j-1,j}\Phi_{j,j-1}-\Psi_{j-1,j}\Psi_{j,j-1} = 1, \quad (9)$$

$$\Psi_{j-1,j} = -\Psi_{j,j-1}, \quad (10)$$

we can formulate the following relationship:

$$R_{j-1,j} = \frac{\Psi_{j-1,j}+R_{j,j+1}}{1+\Psi_{j-1,j}R_{j,j+1}}\exp(2ik_{j-1,z}h_{j-1}). \quad (11)$$

It is reasonable to assume that no wave will be reflected back from the substrate, so that,

$$R_{N,N+1} = 0. \quad (12)$$

Then, the X-ray reflectivity is simply,

$$R = \left|R_{0,1}\right|^2. \quad (13)$$

Thus, we obtain Parratt's expression.

B. Previous calculations of X-ray reflectivity when roughness exists at the surface and interface

When the surface and interface have roughness, the Fresnel coefficient for reflection is reduced (Vidal and Vincent, 1984; Sinha et al., 1988; Holy et al., 1993, 1999; Boer, 1995; Daillant and Gibaud, 1999; Fujii et al., 2004, 2005; Sakurai, 2009; Fujii, 2010, 2011). The effect of roughness was previously put into calculation based on the theory of Nevot and Croce (1980). The effect of such roughness was taken into account only through the effect of changes in density of the medium in a vertical direction to the surface and interface. With the use of relevant roughness parameters such as the root-mean-square (rms) roughness $\sigma_{j-1,j}$ of the jth layer, the reduced Fresnel reflection coefficient Ψ' for s-polarization is transformed as shown below:

$$\Psi'_{j,j-1} = \Psi_{j,j-1}\exp(-2k_{j,z}k_{j-1,z}\sigma^2_{j,j-1}) \quad (14)$$

and the X-ray reflectivity is calculated using the following equation:

$$R_{j-1,j} = \frac{R_{j,j+1}+\Psi'_{j-1,j}}{1+R_{j,j+1}\Psi'_{j-1,j}}\exp(2ik_{j-1,z}h_{j-1}). \quad (15)$$

Figure 1 shows the reflectivity from a tungsten-covered silicon wafer calculated by the theory in use prior to this work.

The ratio of the oscillation amplitude to the value of the reflectivity from a surface with an rms surface roughness of 0.3 nm (dashed line) does not decrease but increases near the angle of incidence of 1.8°. We now consider the above strange result of X-ray reflectivity, which was based on Parratt formalism (Parratt, 1954) with the use of the Nevot and Croce approach to account for roughness (Nevot and Croce, 1980). In that calculation, the X-ray reflectivity is derived using the relation of reflection coefficient $R_{j-1,j}$ and $R_{j,j+1}$ as Eq. (15). However, the relationship between the reflection coefficients $R_{j-1,j}$ and $R_{j,j+1}$ was originally derived
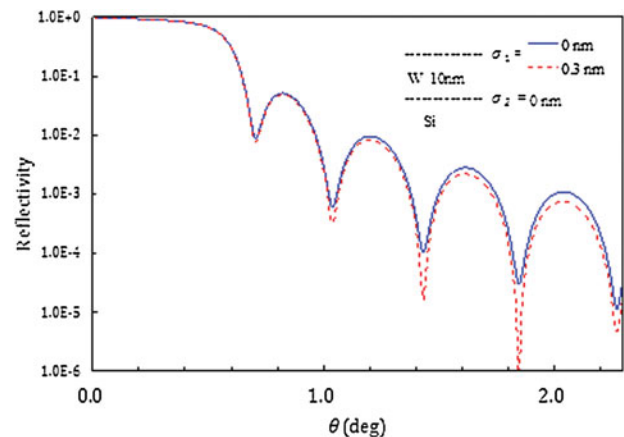

Figure 1. X-ray reflectivity from a silicon wafer covered with a thin (10 nm) tungsten film calculated by the theory in use prior to this work. Solid line shows the case of a flat surface. Dashed line shows the case of a surface with an rms surface roughness of 0.3 nm.

as the following equation:

$$R_{j-1,j} = \frac{\Psi'_{j-1,j} + (\Phi'_{j-1,j}\Phi'_{j,j-1} - \Psi'_{j-1,j}\Psi'_{j,j-1})R_{j,j+1}}{1 - \Psi'_{j,j-1}R_{j,j+1}}$$
$$\times \exp(2ik_{j-1,z}h_{j-1}). \tag{16}$$

Here, the following conditional relations between the Fresnel coefficient for reflection and refraction are relevant to the above equation,

$$\Phi'_{j-1,j}\Phi'_{j,j-1} - \Psi'_{j-1,j}\Psi'_{j,j-1} = 1,$$
$$\Psi'_{j-1,j} = -\Psi'_{j,j-1}, \tag{17}$$

then,

$$\Phi'_{j-1,j}\Phi'_{j,j-1} = 1 - \Psi'^2_{j,j-1}. \tag{18}$$

The Fresnel coefficients for refraction at the rough interface are derived using the Fresnel reflection coefficient Ψ as follows:

$$\Phi'_{j-1,j}\Phi'_{j,j-1} - \Phi_{j-1,j}\Phi_{j,j-1} = \Psi^2_{j,j-1}$$
$$\times \left(1 - \exp(-2k_{j,z}k_{j-1,z}\sigma^2_{j,j-1})\right) > 0 \tag{19}$$

$$\Phi'_{j-1,j}\Phi'_{j,j-1} = \Phi_{j-1,j}\Phi_{j,j-1} + \left(1 - \Phi_{j-1,j}\Phi_{j,j-1}\right)$$
$$\times \left(1 - \exp(-2k_{j,z}k_{j-1,z}\sigma^2_{j,j-1})\right). \tag{20}$$

Therefore, the Fresnel coefficients for refraction at the rough interface are necessarily larger than the Fresnel coefficient for refraction at the flat interface. The resulting increase in transmission coefficient completely overpowers any decrease in the value of reflection coefficient. These coefficients for refraction obviously contain an error as the penetration of X-rays should decrease at a rough interface because of diffuse scattering. We propose that the unnatural results in the previous calculation of X-ray reflectivity originate from the fact that diffuse scattering was not considered. In fact, Eq. (15) contains the X-ray energy conservation rule at the interface as the following identity equation for the Fresnel coefficient:

$$\Phi_{j-1,j}\Phi_{j,j-1} - \Psi_{j-1,j}\Psi_{j,j-1} = \Phi_{j-1,j}\Phi_{j,j-1} + \Psi^2_{j-1,j} = 1. \tag{21}$$

When the Fresnel coefficients at the rough interface obey the following equations:

$$\Phi'_{j-1,j}\Phi'_{j,j-1} - \Psi'_{j-1,j}\Psi'_{j,j-1} = 1, \quad \Psi'_{j-1,j} = -\Psi'_{j,j-1}, \tag{22}$$

these coefficients fulfill X-ray energy flow conservation at the interface, and so diffuse scattering was not considered at the rough interface.

This conservation expression should not apply any longer when the Fresnel reflection coefficient is replaced by the reduced coefficient Ψ' when there is roughening at the interface. Therefore, calculating the reflectivity using this reduced

Fresnel reflection coefficient Ψ' in Eq. (15) will incorrectly increase the Fresnel transmission coefficient Φ', i.e., $\Phi < \Phi'$.

The penetration of X-rays should decrease at the rough interface because of diffuse scattering. Therefore, the identity equations for the Fresnel coefficients become,

$$\Phi'_{j-1,j}\Phi'_{j,j-1} - \Psi'_{j-1,j}\Psi'_{j,j-1} = \Phi'_{j-1,j}\Phi'_{j,j-1}$$
$$+ \Psi'^2_{j-1,j} = 1 - D^2 < 1, \tag{23}$$

where D^2 is a decrease because of diffuse scattering. Then, in the calculation of X-ray reflectivity when there is roughening at the surface or the interface, the Fresnel transmission coefficient Φ' should be used for the reduced coefficient. Several theories exist to describe the influence of roughness on X-ray scattering (Sinha *et al.*, 1988; Holy *et al.*, 1993, 1999; Daillant and Gibaud, 1999). When the surface and interface are both rough, the Fresnel coefficient for refraction has been derived in several theories (Vidal and Vincent, 1984; Sinha *et al.*, 1988; Holy *et al.*, 1993, 1999; Boer, 1995).

C. A new formula for reflectivity of rough multilayer surface

When the z position of the interface of 0th layer and 1st layer $z_{o,1}$ fluctuates vertically as a function of the lateral position because of interface roughness, the relations between the amplitudes A_0, A'_0, A_1, and A'_1 are derived by use of the Fresnel coefficient tensor Φ for refraction and the Fresnel coefficient tensor Ψ for reflection as follows:

$$\begin{pmatrix} A'_0 \\ A_1 \end{pmatrix} =$$
$$\begin{pmatrix} \Psi_{0,1}\dfrac{\exp(i(k_{1,z}+k_{0,z})z_{0,1})}{\exp(i(-k_{0,z}+k_{1,z})z_{0,1})}, & \Phi_{1,0}\exp(i(k_{0,z}-k_{1,z})z_{0,1}) \\ \Phi_{0,1}\exp(i(-k_{1,z}+k_{0,z})z_{0,1}), & \Psi_{1,0}\dfrac{\exp(i(-k_{0,z}-k_{1,z})z_{0,1})}{\exp(i(-k_{0,z}+k_{1,z})z_{0,1})} \end{pmatrix}$$
$$\times \begin{pmatrix} A_0 \\ A'_1 \end{pmatrix}. \tag{24}$$

We therefore consider the derivation of the average value of the following matrix.

We take the average value of this matrix. For Gaussian statistics of standard deviation value σ, the Fresnel reflection coefficient Ψ' is as follows:

$$\Psi'_{0,1} = \Psi_{0,1}\frac{\langle\exp(i(k_{1,z}+k_{0,z})z_{0,1})\rangle}{\langle\exp(i(-k_{0,z}+k_{1,z})z_{0,1})\rangle}$$
$$= \Psi_{0,1}\frac{\exp(-\frac{1}{2}(k_{0,z}+k_{1,z})^2\sigma^2_{0,1})}{\exp(-\frac{1}{2}(k_{0,z}-k_{1,z})^2\sigma^2_{0,1})}$$
$$= \Psi_{0,1}\exp(-2k_{0,z}k_{1,z}\sigma^2_{0,1}). \tag{25}$$

These reduced reflection coefficients accord with the result in prior works (Vidal and Vincent, 1984; Sinha *et al.*, 1988; Holy *et al.*, 1993, 1999; Boer, 1995; Daillant and Gibaud, 1999; Sakurai, 2009). SinceX-rays penetrating an interface reflect from the interface below, and penetrate former

interface again without fail, it is necessary to treat the refraction coefficients $\Phi'_{0,1}$ and $\Phi'_{1,0}$ collectively.

$$\Phi'_{0,1}\Phi'_{1,0}$$

$$= \langle \Phi_{0,1}\exp(i(-k_{1,z}+k_{0,z})z_{0,1})\Phi_{1,0}\exp(i(k_{0,z}-k_{1,z})z_{0,1})\rangle$$

$$= \Phi_{0,1}\Phi_{1,0}\langle\exp(i(-k_{1,z}+k_{0,z})z_{0,1})\exp(i(k_{0,z}-k_{1,z})z_{0,1})\rangle$$

$$= \Phi_{0,1}\Phi_{1,0}\langle\exp(i(2k_{0,z}-2k_{1,z})z_{0,1})\rangle$$

$$= \Phi_{0,1}\Phi_{1,0}\exp(-2(k_{0,z}-k_{1,z})^2\sigma_{0,1}^2). \qquad (26)$$

Then the Fresnel coefficients Ψ' and Φ' are reduced as follows:

$$\begin{pmatrix} A'_0 \\ A_1 \end{pmatrix}$$

$$= \begin{pmatrix} \Psi_{0,1}\exp(-2k_{0,z}k_{1,z}\sigma_{0,1}^2), & \Phi_{1,0}\exp(-(k_{0,z}-k_{1,z})^2\sigma_{0,1}^2) \\ \Phi_{0,1}\exp(-(k_{0,z}-k_{1,z})^2\sigma_{0,1}^2), & \Psi_{1,0}\exp(-2k_{0,z}k_{1,z}\sigma_{0,1}^2) \end{pmatrix}$$

$$\times \begin{pmatrix} A_0 \\ A'_1 \end{pmatrix}. \qquad (27)$$

The Fresnel refraction coefficients Φ' derived by this method are reduced, and can be used to calculate the reflectivity from rough surface and interface. Therefore, we calculate the reflectivity using these newly derived Fresnel coefficients in an accurate reflectivity equation of $R_{j-1,j}$ and $R_{j,j+1}$ as follows:

$$R_{j-1,j} = \frac{\Psi'_{j-1,j}+(\Phi'_{j-1,j}\Phi'_{j,j-1}-\Psi'_{j-1,j}\Psi'_{j,j-1})R_{j,j+1}}{1-\Psi'_{j,j-1}R_{j,j+1}}$$

$$\times \exp(2ik_{j-1,z}h_{j-1}). \qquad (28)$$

Based on the above considerations, we again calculated the X-ray reflectivity of the W/Si system, but now considered the effect of attenuation in the refracted X-rays by diffuse scattering resulting from surface roughness. However, the reduced refraction coefficient in prior work varies (Vidal and Vincent,
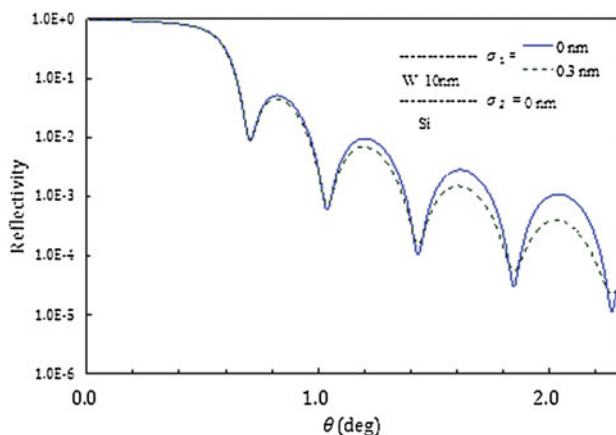

Figure 2. X-ray reflectivity from a silicon wafer covered with a thin (10 nm) tungsten film calculated by the new calculation that considered diffuse scattering. Solid line shows the case of a flat surface. Dashed line shows the case of a surface with an rms surface roughness of 0.3 nm.

1984; Sinha *et al.*, 1988; Holy *et al.*, 1993, 1999; Boer, 1995; Daillant and Gibaud, 1999; Fujii *et al.*, 2004, 2005; Sakurai, 2009; Fujii, 2010, 2011). Then, about the reduced refraction coefient, reduction is the same as the reflection coefficient applied.

In the reflectivity curve from a surface with an rms surface roughness of 0.3 nm (dashed line), the amplitude of oscillation in Figure 1 has reduced in Figure 2. These results are now physically reasonable. The strange results seen in Figure 1 have disappeared in Figure 2. It seems natural that the effect of interference does decrease at a rough surface and interface, because the amount of coherent X-rays should reduce because of diffuse scattering.

III. CONCLUSION

We have investigated the fact that the calculated results of X-ray reflectivity based on Parratt formalism with the effect of the roughness incorporated by the theory of Nevot–Croce show a strange phenomenon in which the amplitude of the oscillation because of the interference effects increase in the case of the rougher surface. We found that the strange results originate in the currently used equation where the Fresnel refraction coefficient in the reflectivity equation is increased at a rough interface. The increase in the transmission coefficient completely overpowers any decrease in the value of the reflection coefficient because of the lack of consideration of diffuse scattering. Then, we have developed a new formalism that corrects this error, producing more accurate estimates of the X-ray reflectivity for systems having surface and interfacial roughness, taking into account the effect of roughness-induced diffuse scattering. The new, accurate formalism is completely described in detail. The X-ray reflectivity R is derived by use of accurate reflectivity equations for $R_{j-1,j}$ and $R_{j,j+1}$ as the following:

$$R = |R_{0,1}|^2, \quad R_{N,N+1} = 0,$$

$$R_{j-1,j} = \frac{\Psi'_{j-1,j}+(\Phi'_{j-1,j}\Phi'_{j,j-1}-\Psi'_{j-1,j}\Psi'_{j,j-1})R_{j,j+1}}{1-\Psi'_{j,j-1}R_{j,j+1}}$$

$$\times \exp(2ik_{j-1,z}h_{j-1}),$$

where the Fresnel coefficients Ψ' for reflection and Φ' for refraction are reduced. The reflectivity calculated with this new, accurate formalism, gives a physically reasonable result. Here, the reduced Fresnel coefficients Ψ' and Φ' in the reflectivity equation need further research and we will continue to refine this theory. The use of this equation resolves the strange numerical results that occurred in the previous calculations that neglected diffuse scattering and is expected that the buried interface structure can now be analyzed more accurately.

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