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Estimation of Surface and Interface Roughness Using X-ray Reflectivity and TEM Observation

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Abstract Surface and interface roughness of multilayer surface are often estimated with using X-ray reflectivity (XRR). 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 result showed a strange phenomenon. The strange result had its origin in a currently used an equation due to serious mistake in which the Fresnel transmission coefficient in the reflectivity equation was increased at a rough interface because of a lack of consideration of diffuse scattering. Then we have developed a new improved formalism that corrects this mistake. In this study, we present the applying of the new improved formalism with the use of TEM observation results. The new improved formalism derives a more accurate analysis of the x-ray reflectivity from a multilayer surface of thin film material.

Keywords Surface , Interface Roughness, Multilayered Thin Film Materials, X-Ray Reflectivity

1. Introduction

X-ray reflectivity (XRR) is a powerful tool for investigations on rough surface and interface structures of material surfaces as multilayered thin film[1-11]. In many previous XRR analysis, the X-ray reflectivity was calculated based on the Parratt formalism[1], coupled with the use of the theory of Nevot and Croce to include roughness[2]. However, the calculated results of the X-ray reflectivity done in this way often showed strange results where the amplitude of the oscillation due to the interference effects would increase for a rougher surface. And the estimation results of surface and interfacial roughness by x-ray reflectivity measurements did not correspond to those from transmission electron microscope (TEM) observation[6]. The origin of the strange behavior was attributed to the fact that the diffuse scattering at the rough interface was not correctly taken into account by Nevot and Croce[2]. Then we have developed a new formalism in which the effects of the surface and interface roughness are included correctly. The new improved formalism derives an accurate analysis of the x-ray reflectivity from a multilayer surface of thin film materials, taking into account the effect of roughness-induced diffuse scattering. In the new improved formulae for the x-ray reflectivity, the well known reduced Fresnel coefficients for reflection is applied to the Fresnel coefficients for reflection at rough interface. While an accurate analytical formula for the Fresnel coefficients for refraction at rough interface is not available. There are several approximations proposed so far and all these results can be written by including any parameters depend on the proposed approximations. In the present work, we tried to determine these parameters experimentally by comparing the results of the TEM observation and x-ray reflectivity.

2. Comparison between X-ray Reflectivity Measurement and TEM Observation

The surface sample for examination was prepared as follows; a GaAs layer was grown on Si(110) by molecular beam epitaxy (MBE). From the TEM observations, the thickness of the GaAs layer was 48 nm, the root-mean-square (rms) roughness σ_1 of the GaAs surface was about 4.3 nm, the rms roughness of the interface between GaAs and Si was about 0.7 nm. (We estimated σ_1=2.8nm in the previous study, now we revise the σ_1 in 4.3nm.) Figure 1 shows a cross section image of this GaAs / Si(110) sample observed by TEM.

X-ray reflectivity measurements were performed using a Cu-Kα_1 x-ray beam from an 18 kW rotating-anode source. In Figure 2, the solid line shows the measured x-ray reflectivity from the surface sample of the GaAs layer on the silicon wafer. The oscillations in decrease signal for angles larger than the total reflection critical angle are caused by interference between x-rays that reflect from the surface of GaAs layer and those that reflect from the interface of the GaAs layer and Si substrate. The characteristics of these

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oscillations reflect the surface roughness and the interface roughness.

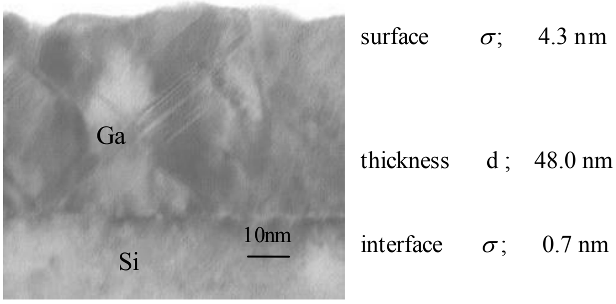

Figure 1. Cross section image of GaAs / Si(110) by TEM observation

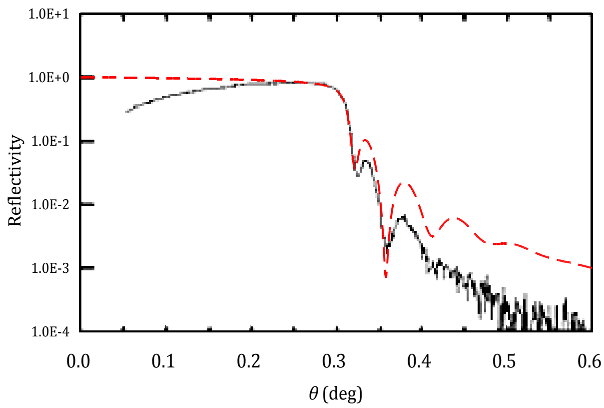

Figure 2. Calculated (dashed line) and measured (solid line) reflectivity from a GaAs layer with a thickness of 48 nm on a Si substrate. The surface roughness σ_1 is 4.3 nm and the interfacial roughness σ_2 is 0.7 nm

In the conventional x-ray reflectivity analysis, the reflectivity R from a multilayer consisting of N layers with a flat surface and flat interfaces is calculated based on the Parratt formalism[1] as the following equations;

$$R = \left| R_{0,1} \right|^2 \ ,$$

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

$$R_{N,N+1} = 0 \tag{1}$$

$$k_{j,z} = k\sqrt{n_j^{\,2} - \cos^2\theta} \tag{2}$$

where the reflection coefficient $R_{0,1}$ is defined as the ratio of the reflected electric field to the incident electric field at the surface of the material, the reflection coefficient $R_{j\text{-}1,j}$ is defined as the ratio of the reflected electric field to the incident electric field at the interface of j-1-th layer and j-th layer of the material, h_j is the thickness of j-th layer, $k_{j,z}$ is the z-direction component of the wave vector in the j-th layer, k=2π/λ , λ; wave length, θ; glancing angle of incidence. Here the refractive index of the j-th layer $n_j = 1 - \delta_j - i\beta_j$, $n_0 = 1$. The real and imaginary parts of the refractive index are related to the atomic scattering factor and electron density of the j-th layer material. For x-rays of wavelength λ, the optical constants of the j-th layer material consisting of N_{ij} atoms per unit volume can be expressed as

$$\delta_j = \frac{\lambda^2 r_e}{2\pi} \sum_i f_{1i} N_{ij} \ , \quad \beta_j = \frac{\lambda^2 r_e}{2\pi} \sum_i f_{2i} N_{ij} \ , \tag{3}$$

where r_e is the classical electron radius and f_{1i} and f_{2i} are the real and imaginary parts of the atomic scattering factor of the i-th element atom, respectively.

$\Psi_{j\text{-}1,\,j}$ is the Fresnel coefficients for reflection at the interface between (j-1)-th and j-th layers as

$$\Psi_{j-1,j} = \frac{k_{j-1,z} - k_{j,z}}{k_{j-1,z} + k_{j,z}} \ , \quad \Psi_{j,j-1} = -\Psi_{j-1,j} \tag{4}$$

When the surface and interface have roughness, the conventional x-ray reflectivity is calculated based on the Parratt formalism[1], incorporating the effect of the interface roughness according to Nevot and Croce[2]. The Fresnel coefficient for reflection from rough surface and rough interfaces is reduced by the roughness[8-14]. The effect of such roughness was taken into account only through the effect of the changes in density of the medium in a vertical direction to the surface and interface. With the use of relevant roughness parameters like the root-mean-square (rms) roughness $\sigma_{j\text{-}1,j}$ between (j-1)-th and j-th layers, the formula for the reduced Fresnel reflection coefficient $\Psi'_{j\text{-}1,\,j}$ is well known as showed

$$\Psi'_{j-1,j} = \frac{k_{j-1,z} - k_{j,z}}{k_{j-1,z} + k_{j,z}} \exp\left(-2k_{j-1,z} k_{j,z} \sigma_{j-1,j}^{\;2}\right),$$

$$\Psi'_{j,j-1} = -\Psi'_{j-1,j} \tag{5}$$

In Figure 2, the dashed line shows the result of a calculation based on these expressions of the reflectivity of x-rays from a GaAs layer with a thickness of 48 nm on Si substrate. The rms roughness of the interface of GaAs and Si was set to 0.7 nm, the value derived from the TEM observations. The rms roughness of the GaAs surface was set to 4.3 nm, the value derived from atomic force microscope (AFM) measurements. The agreement of the calculated and experimental results in Figure 2 is not good. This disagreement is mainly caused by the fact that the diffuse scattering at the rough interface was not correctly taken into account by Nevot and Croce[2].

3. Improved Formalism of X-ray Reflectivity

We have developed a new formula in which the effects of the surface and interface roughness are correctly treated. 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 the energy by interface roughness cannot be correctly accounted for Parratt's expression.

In the 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 electric field of x-ray radiation at a glancing angle of incidence θ is expressed as

$$\mathbf{E}_0(z) = \mathbf{A}_0 \exp[i(\mathbf{k}_0 \cdot \mathbf{r} - \omega t)] \quad (6)$$

The incident radiation is usually decomposed into two geometries to simplify the analysis, one with the incident electric field E parallel to the plane of incidence (p-polarization) and one with E perpendicular to that plane (s-polarization). An arbitrary incident wave can be represented in terms of these two polarizations. Thus, E_{0x} and E_{0z} correspond to p-polarization, and E_{0y} to s-polarization; those components of the amplitude's electric vector are expressed as

$$A_{0x} = -A_{0p}\sin\theta,\ A_{0y} = A_{0s},\ A_{0z} = A_{0p}\cos\theta. \quad (7)$$

The components of the wave vector $\boldsymbol{k}_0$ of the incident x-rays are

$$k_{0x} = \frac{2\pi}{\lambda}\cos\theta \ ,\ k_{0y} = 0 \ ,\ k_{0z} = \frac{2\pi}{\lambda}\sin\theta \ . \quad (8)$$

The electric field of reflected x-ray radiation at an exit angle θ is expressed as

$$\mathbf{E}'_0(z) = \mathbf{A}'_0 \exp[i(\mathbf{k}'_0 \cdot \mathbf{r} - \omega t)] \ . \quad (9)$$

where $k'_{0x} = k_{0x},\ k'_{0y} = k_{0y},\ k'_{0z} = -k_{0z}.$ (10)

We consider the electric field $\boldsymbol{E}_{j-1}$ of x-rays propagating in the j-1-th layer material, and the electric field $\boldsymbol{E}_j$ of x-rays propagating in the j-th layer material, and the electric field $\boldsymbol{E}'_{j-1}$ of x-rays reflected from the j-th layer material at z=$z_{j-1,j}$ of the interface between the j-1-th and j-th layers. The electric fields $\boldsymbol{E}_{j-1}$, $\boldsymbol{E}'_{j-1}$ **at** the interface between the j-1-th layer and j-th layer and 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

$$\mathbf{E}_{j-1}(z_{j-1,j}) = \mathbf{A}_{j-1}\exp[i(k_{j-1,x}x + k_{j-1,y}y + k_{j-1,z}h_{j-1} - \omega t)],$$
$$\mathbf{E}'_{j-1}(z_{j-1,j}) = \mathbf{A}'_{j-1}\exp[i(k_{j-1,x}x + k_{j-1,y}y - k_{j-1,z}h_{j-1} - \omega t)],$$
$$\mathbf{E}_j(z_{j-1,j}) = \mathbf{A}_j \exp[i(k_{j,x}x + k_{j,y}y - \omega t)] \ ,$$
$$\mathbf{E}'_j(z_{j-1,j}) = \mathbf{A}'_j \exp[i(k_{j,x}x + k_{j,y}y - \omega t)] \ . \quad (11)$$

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

$$\boldsymbol{k}_j \cdot \boldsymbol{k}_j = n_j^{\,2}\boldsymbol{k}_0 \cdot \boldsymbol{k}_0 \ ,\ \boldsymbol{k}_0 \cdot \boldsymbol{k}_0 = k^2, \quad (12)$$

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

$$k_{j,z} = \sqrt{n_j^{\,2}k^2 - k_{0,x}^{\,2}} \quad (13)$$

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

$$\begin{pmatrix} \boldsymbol{A}'_{j-1}\exp(-ik_{j-1,z}h_{j-1}) \\ \boldsymbol{A}_j \end{pmatrix} = \begin{pmatrix} \boldsymbol{\Psi}_{j-1,j} & \boldsymbol{\Phi}_{j,j-1} \\ \boldsymbol{\Phi}_{j-1,j} & \boldsymbol{\Psi}_{j,j-1} \end{pmatrix} \begin{pmatrix} \boldsymbol{A}_{j-1}\exp(ik_{j-1,z}h_{j-1}) \\ \boldsymbol{A}'_j \end{pmatrix}, \quad (14)$$

where $\boldsymbol{\Psi}_{j-1j}$ and $\boldsymbol{\Phi}_{j-1,j}$ are the Fresnel coefficient tensor for reflection and refraction at the interface between the j-1 and j layers. For s-polarization, the Fresnel coefficients are,

$$\boldsymbol{\Phi}_{j-1,j,yy} = \frac{2k_{j-1,z}}{k_{j-1,z} + k_{j,z}} \ ,$$
$$\boldsymbol{\Phi}_{j,j-1,yy} = \frac{2k_{j,z}}{k_{j-1,z} + k_{j,z}} \quad (15)$$
$$\Psi_{j-1,j,yy} = \frac{k_{j-1,z} - k_{j,z}}{k_{j-1,z} + k_{j,z}} \ ,$$
$$\Psi_{j,j-1,yy} = \frac{k_{j,z} - k_{j-1,z}}{k_{j-1,z} + k_{j,z}} \quad (16)$$

The reflection coefficient is defined as the ratio $\boldsymbol{R}_{0,1}$ of the reflected electric field to the incident electric field at the surface of the material and is given by,

$$\boldsymbol{A}'_0 = \boldsymbol{R}_{0,1}\boldsymbol{A}_0 \ . \quad (17)$$

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

$$\boldsymbol{A}'_{j-1} = \boldsymbol{R}_{j-1,j}\boldsymbol{A}_{j-1} \ , \quad (18)$$

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

$$\boldsymbol{R}_{j-1,j} = \frac{\boldsymbol{\Psi}_{j-1,j} + (\boldsymbol{\Phi}_{j-1,j}\boldsymbol{\Phi}_{j,j-1} - \boldsymbol{\Psi}_{j-1,j}\boldsymbol{\Psi}_{j,j-1})\boldsymbol{R}_{j,j+1}}{1 - \boldsymbol{\Psi}_{j,j-1}\boldsymbol{R}_{j,j+1}} \cdot \exp(2ik_{j-1,z}h_{j-1}) \ . \quad (19)$$

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

$$\boldsymbol{\Phi}_{j-1,j}\boldsymbol{\Phi}_{j,j-1} - \boldsymbol{\Psi}_{j-1,j}\boldsymbol{\Psi}_{j,j-1} = 1$$
$$\boldsymbol{\Psi}_{j-1,j} = -\boldsymbol{\Psi}_{j,j-1} \quad (20)$$

we can formulate the following relationship

$$\boldsymbol{R}_{j-1,j} = \frac{\boldsymbol{\Psi}_{j-1,j} + \boldsymbol{R}_{j,j+1}}{1 + \boldsymbol{\Psi}_{j-1,j}\boldsymbol{R}_{j,j+1}}\exp(2ik_{j-1,z}h_{j-1}) \ . \quad (21)$$

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

$$\boldsymbol{R}_{N,N+1} = 0 \ . \quad (22)$$

Then, the x-ray reflectivity is simply,

$$R = \left|R_{0,1}\right|^2 \ . \quad (23)$$

Thus, we obtain the precious expression of Parratt's formalism.

When the surface and interface have roughness, the Fresnel coefficient for reflection is reduced by the roughness. The effect of the roughness was previously put into the calculation based on the theory of Nevot and Croce[2]. We now consider the x-ray reflectivity which was calculated based on the Parratt formalism[1] with the use of the Nevot and Croce approach to account for roughness[2].

In that calculation, the x-ray reflectivity is derived using the relation of the reflection coefficient $R_{j-1,j}$ and $R_{j,j+1}$ as equation (1). However, the relationship between the reflection coefficients $R_{j-1,j}$ and $R_{j,j+1}$ was originally derived as the equation (19). Then, the following conditional relations between the reduced Fresnel reflection coefficient $\varPsi'$ and the Fresnel refraction coefficient $\varPhi'$ are included in the above equation (1),

$$\boldsymbol{\varPhi}'_{j-1,j}\,\boldsymbol{\varPhi}'_{j,j-1} - \boldsymbol{\varPsi}'_{j-1,j}\,\boldsymbol{\varPsi}'_{j,j-1} = 1,\qquad \boldsymbol{\varPsi}'_{j-1,j} = -\boldsymbol{\varPsi}'_{j,j-1}\ , \tag{24}$$

From eqs (20) and (24), the Fresnel refraction coefficients at the rough interface are derived using the Fresnel reflection coefficient $\varPsi$ as follows,

$$\begin{aligned}&\mathbf{\Phi}'_{j-1,j}\,\mathbf{\Phi}'_{j,j-1} - \mathbf{\Phi}_{j-1,j}\mathbf{\Phi}_{j,j-1}\\ &= \mathbf{\Psi}^2_{j,j-1}\left(1-\exp(-2k_{j,z}k_{j-1,z}\sigma^2_{j,j-1})\right) > 0\\ &\mathbf{\Phi}'_{j-1,j}\,\mathbf{\Phi}'_{j,j-1}\\ &= \mathbf{\Phi}_{j-1,j}\mathbf{\Phi}_{j,j-1} + \left(1-\mathbf{\Phi}_{j-1,j}\mathbf{\Phi}_{j,j-1}\right)\left(1-\exp(-2k_{j,z}k_{j-1,z}\sigma^2_{j,j-1})\right)\end{aligned} \tag{25}$$

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 the transmission coefficient completely overpowers any decrease in the value of the reflection coefficient. These coefficients for refraction obviously contain a mistake because 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 the x-ray reflectivity originate from the fact that diffuse scattering was not considered. In fact equation (1) contains the x-ray energy conservation rule at the interface as the following identity equation for the Fresnel coefficient,

$$\varPhi_{j-1,j}\varPhi_{j,j-1} - \varPsi_{j-1,j}\varPsi_{j,j-1} = \varPhi_{j-1,j}\varPhi_{j,j-1} + \varPsi_{j-1,j}{}^2 = 1 \tag{26}$$

When the Fresnel coefficients at the rough interface obeys the following equations,

$$\mathbf{\Phi}'_{j-1,j}\,\mathbf{\Phi}'_{j,j-1} - \mathbf{\Psi}'_{j-1,j}\,\mathbf{\Psi}'_{j,j-1} = 1,\ \mathbf{\Psi}'_{j-1,j} = -\mathbf{\Psi}'_{j,j-1}, \tag{27}$$

these coefficients fulfil 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 $\varPsi'$ when there is roughening at the interface. Therefore, calculating the reflectivity using this reduced Fresnel reflection coefficient $\varPsi'$ in equation (1) will incorrectly increase the Fresnel transmission coefficient $\varPhi'$, *i.e.*, $\varPhi < \varPhi'$.

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

$$\varPhi'_{j-1,j}\,\varPhi'_{j,j-1} - \varPsi'_{j-1,j}\,\varPsi'_{j,j-1} = \varPhi'_{j-1,j}\,\varPhi'_{j,j-1} + \varPsi'_{j-1,j}{}^2 < 1 \tag{28}$$

Then, in the calculation of x-ray reflectivity when there is roughening at the surface or the interface, the Fresnel transmission coefficient $\varPhi'$ should be used for the reduced coefficient. Several theories exist to describe the influence of roughness on x-ray scattering[8-10,12]. When the surface and interface are both rough, the Fresnel coefficient for refraction has been derived in several theories[12-14].

We derived the the Fresnel transmission coefficient $\varPhi'$ at rough interfaces as the following.

When the z position of the interface of 0-th layer and 1-th layer $z_{o,1}$ fluctuates vertically as a function of the lateral position because of the interface roughness, the relations matrix between the amplitudes A_0, A'_0, A_1, and A'_1 of the electric field $\boldsymbol{E_0}$ and $\boldsymbol{E'_0}$ of x-rays in the 0-th layer and the electric field $\boldsymbol{E_1}$ and $\boldsymbol{E'_1}$ of x-rays in the 1-th layer material are derived by the use of the Fresnel coefficient tensor $\boldsymbol{\varPhi}$ for refraction and the Fresnel coefficient tensor $\boldsymbol{\varPsi}$ for reflection as follows

$$\begin{pmatrix} A'_0 \\ A_1 \end{pmatrix} = \begin{pmatrix} \mathbf{\Psi}_{0,1}\dfrac{\exp(i(k_{1,z}+k_{0,z})z_{0,1})}{\exp(i(-k_{0,z}+k_{1,z})z_{0,1})} & \mathbf{\Phi}_{1,0}\exp(i(k_{0,z}-k_{1,z})z_{0,1}) \\ \mathbf{\Phi}_{0,1}\exp(i(-k_{1,z}+k_{0,z})z_{0,1}) & \mathbf{\Psi}_{1,0}\dfrac{\exp(i(-k_{0,z}-k_{1,z})z_{0,1})}{\exp(i(-k_{0,z}+k_{1,z})z_{0,1})} \end{pmatrix} \begin{pmatrix} A_0 \\ A'_1 \end{pmatrix} \tag{29}$$

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 $\varPsi'$ is as follows

$$\begin{aligned}\mathbf{\Psi}'_{0,1} &= \mathbf{\Psi}_{0,1}\frac{\left\langle \exp(i(k_{1,z}+k_{0,z})z_{0,1})\right\rangle}{\left\langle \exp(i(-k_{0,z}+k_{1,z})z_{0,1})\right\rangle}\\ &= \mathbf{\Psi}_{0,1}\frac{\exp(-\frac{1}{2}(k_{0,z}+k_{1,z})^2\sigma_{0,1}{}^2)}{\exp(-\frac{1}{2}(k_{0,z}-k_{1,z})^2\sigma_{0,1}{}^2)}\\ &= \mathbf{\Psi}_{0,1}\exp(-2k_{0,z}k_{1,z}\sigma_{0,1}{}^2)\end{aligned} \tag{30}$$

These reduced reflection coefficient accord with the result in prior work[12-14]. Because x-rays that penetrate 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.

$$\begin{aligned}\mathbf{\Phi}'_{0,1}\mathbf{\Phi}'_{1,0} &= \left\langle \mathbf{\Phi}_{0,1}\exp(i(-k_{1,z}+k_{0,z})z_{0,1})\mathbf{\Phi}_{1,0}\exp(i(k_{0,z}-k_{1,z})z_{0,1})\right\rangle\\ &= \mathbf{\Phi}_{0,1}\mathbf{\Phi}_{1,0}\left\langle \exp(i(-k_{1,z}+k_{0,z})z_{0,1})\exp(i(k_{0,z}-k_{1,z})z_{0,1})\right\rangle\\ &= \mathbf{\Phi}_{0,1}\mathbf{\Phi}_{1,0}\left\langle \exp(i(2k_{0,z}-2k_{1,z})z_{0,1})\right\rangle\\ &= \mathbf{\Phi}_{0,1}\mathbf{\Phi}_{1,0}\exp(-2(k_{0,z}-k_{1,z})^2\sigma_{0,1}{}^2)\end{aligned} \tag{31}$$

Then the Fresnel coefficients $\varPsi'$ and $\varPhi'$ are reduced as follows

$$\begin{aligned}\boldsymbol{\varPsi}'_{0,1} &= \boldsymbol{\varPsi}_{0,1}\exp(-2k_{0,z}k_{1,z}\sigma^2_{0,1})\ ,\\ \boldsymbol{\varPsi}'_{1,0} &= \boldsymbol{\varPsi}_{1,0}\exp(-2k_{0,z}k_{1,z}\sigma^2_{0,1})\\ \boldsymbol{\varPhi}'_{0,1} &= \boldsymbol{\varPhi}_{0,1}\exp(-(k_{0,z}-k_{1,z})^2\sigma_{0,1}{}^2)\ ,\\ \boldsymbol{\varPhi}'_{1,0} &= \boldsymbol{\varPhi}_{1,0}\exp(-(k_{0,z}-k_{1,z})^2\sigma_{0,1}{}^2)\end{aligned} \tag{32}$$

The Fresnel refraction coefficients Φ' derived by this method are reduced, and can be used to calculate the reflectivity from rough surface and interface.

The relation between the amplitudes A_j and A'_j at the j-th layer and the amplitudes A_{j-1} and A'_{j-1} at the j-1-th layer is expressed by the following matrix

$$\begin{pmatrix} \mathbf{A}'_{j-1} \exp(-ik_{j-1,z}h_{j-1}) \\ \mathbf{A}_j \end{pmatrix} = \begin{pmatrix} \mathbf{\Psi}'_{j-1,j} & \mathbf{\Phi}'_{j,j-1} \\ \mathbf{\Phi}'_{j-1,j} & \mathbf{\Psi}'_{j,j-1} \end{pmatrix} \begin{pmatrix} \mathbf{A}_{j-1} \exp(ik_{j-1,z}h_{j-1}) \\ \mathbf{A}'_j \end{pmatrix}, \quad (33)$$

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

$$\boldsymbol{A}'_{j-1} = \boldsymbol{R}_{j-1,j} \boldsymbol{A}_{j-1}, \quad (34)$$

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,

$$\mathbf{R}_{j-1,j} = \frac{\mathbf{\Psi}'_{j-1,j} + (\mathbf{\Phi}'_{j-1,j}\mathbf{\Phi}'_{j,j-1} - \mathbf{\Psi}'_{j-1,j}\mathbf{\Psi}'_{j,j-1})\mathbf{R}_{j,j+1}}{1 - \mathbf{\Psi}'_{j,j-1}\mathbf{R}_{j,j+1}} \exp(2ik_{j-1,z}h_{j-1}) \quad (35)$$

Based on the above considerations, we again calculated the x-ray reflectivity for the GaAs/Si system, but now considered the effect of attenuation in the refracted x-rays by diffuse scattering resulting from surface roughness, *i.e.*, in the equation (35), the reduced refraction coefficient $\Phi'_{j-1,j}$ was derived from Eqs. (15), (16), (32) and applied as

$$\Phi_{j-1,j} = \frac{2k_{j-1,z}}{k_{j-1,z} + k_{j,z}} \exp\{-(k_{j-1,z} - k_{j,z})^2 \sigma_{0,1}{}^2\},$$
$$\Phi_{j,j-1} = \Phi_{j-1,j} \frac{k_{j,z}}{k_{j-1,z}} \quad (36)$$

Figure 3, the dashed line shows the calculation result of x-ray reflectivity provided in these improved formula. The calculation results got closer to the experimental results. However, the agreement is not good.

Although formula for $\Psi_{j-1,j}$ is well known[9-11],

$$\Psi_{j-1,j} = \frac{k_{j-1,z} - k_{j,z}}{k_{j-1,z} + k_{j,z}} \exp\left(-2k_{j-1,z}k_{j,z}\sigma_{j-1,j}{}^2\right),$$
$$\Psi_{j,j-1} = -\Psi_{j-1,j} \quad (37)$$

an accurate analytical formula for $\Phi_{j-1,j}$ including the effect of the interface roughness is not available. There are several approximations proposed so far and all these results can be written with the use of parameters C_1, C_2 as

$$\Phi_{j-1,j} = \frac{2k_{j-1,z}}{k_{j-1,z} + k_{j,z}} \exp\{-[C_1(k_{j-1,z} - k_{j,z})^2 + C_2 k_{j-1,z}k_{j,z}]\sigma_{0,1}{}^2\}, \quad (38)$$
$$\Phi_{j,j-1} = \Phi_{j-1,j} \frac{k_{j,z}}{k_{j-1,z}}$$

where parameters C_1, C_2 depend on the proposed approximation[7,12-14]. In the present work, we tried to determine these parameters experimentally by comparing the measurements of TEM observation results and x-ray reflectivity. The TEM observation shows that the thickness of GaAs layer is 48 nm, the surface roughness σ_1 of GaAs is 4.3 nm, and the interfacial roughness σ_2 on Si substrate is 0.7 nm. Using Eqs. (35), (37), (38), the reflectivity was calculated with various values of C_1, C_2. After choosing the parameters C_1, C_2 so that the calculation result of X-ray reflectivity accorded with the experimental result in the TEM observation, $C_1 = 0.5$ and $C_2 = 0.5$ were provided. In Figure 4, the dashed line shows the calculation result of x-ray reflectivity provided in these way. The calculation results reproduce the experimental results almost well. It is thought that the value of the parameter C_1, C_2 depends on the structure of a parallel direction on the surface in the surface roughness and the interface roughness. Therefore, the investigation about many samples will be necessary in future.

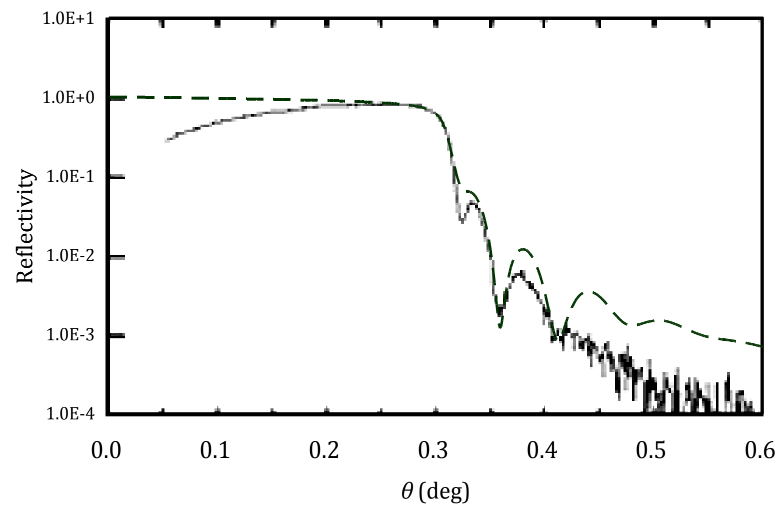

Figure 3. Calculated (doted line) reflectivity by improved formalism and measured (solid line) reflectivity from a GaAs layer with a thickness of 48 nm on a Si substrate. The surface roughness σ_1 is 4.3 nm and the interfacial roughness σ_2 is 0.7 nm

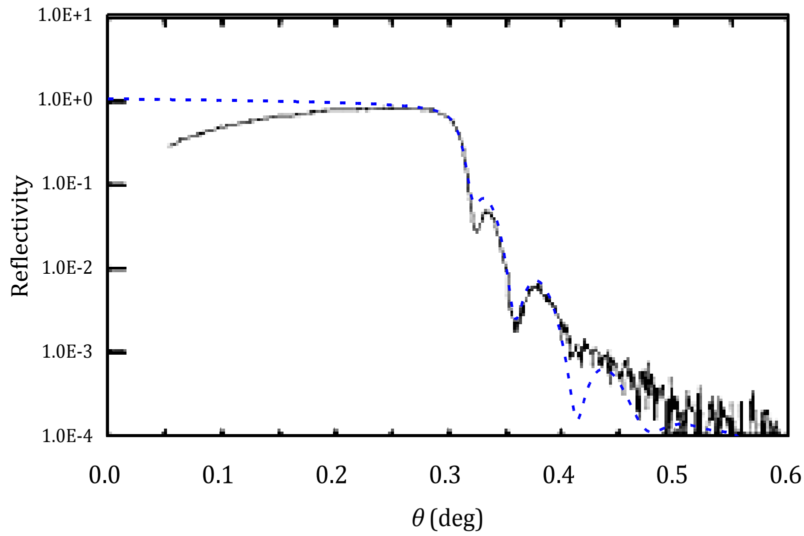

Figure 4. Calculated (doted line) reflectivity by improved formalism with the parameters in the Fresnel transmission coefficient and measured (solid line) reflectivity from a GaAs layer with a thickness of 48 nm on a Si substrate. The surface roughness σ_1 is 4.3 nm and the interfacial roughness σ_2 is 0.7 nm

4. Conclusions

We have developed a new improved formalism of x-ray reflectivity. In this study, we present the applying of the new improved formalism with the use of TEM observation results. In the new improved formulae for the x-ray reflectivity, an accurate analytical formula for the Fresnel coefficients for refraction at rough interface was not available. In this study, we show to be able to determine these parameters in the Fresnel coefficients for refraction experimentally by comparing the measurements of TEM observation results and x-ray reflectivity. It is thought that the value of these parameters depend on the structure of a parallel direction on the surface in the surface roughness and the interface roughness. Therefore, the investigation about many samples will be necessary in future so that we can use these parameters for the calculation of the x-rays reflectivity in the structure analysis of the similar surface layer and enable structure analysis of good precision.

REFERENCES

[1] Parratt, L. G., 1954,. Surface Studies of Solids by Total Reflection of X-Rays, Phys. Rev. 95, 359-369.

[2] Nevot, L. and Croce, P., 1980, Caracterisation des surfaces par reflexion rasante de rayons X. Application a l'etude du polissage de quelques verres silicates, Rev. Phys. Appl. 15, 761-779.

[3] Fujii, Y., Nakayama, T., and Yoshida, K., 2004, Roughness Estimation of Polycrystalline Iron Surface under High Temperature by Small Glancing Angle X-ray Scattering, ISIJ International 44, 1549-1553.

[4] Fujii, Y., Komai, T., and Ikeda, K., 2005, Depth profiling of polycrystalline layers under a surface using x-ray diffraction at small glancing angle of incidence, Surf. Interface Anal. 37, 190-193.

[5] Fujii, Y., 2010, Influence of surface roughness on near-surface depth analysis from X-ray reflectivity measurements, Surf. Interface Anal. 42, 1642-1645.

[6] Fujii, Y., 2011, Comparison of Roughness Estimations by X-ray Reflectivity Measurements and TEM observations, Series: Materials Science and Engineering 24 012008-1-8.

[7] Fujii, Y., 2011, Improved x-ray reflectivity calculations for rough surfaces and interfaces, Series: Materials Science and Engineering 24 012009-1-21.

[8] Holy, V., Pietsch, U., and Baumbach, T. (Eds.), 1999, *High-Resolution X-ray Scattering from Thin Films and Multilayers* (Berlin Springer

[9] Holy, V., Kubena, J., Ohlidal, I., Lischka, K., and Plotz, W., 1993, X-ray reflection from rough layered systems, Phys. Rev. B 47, 15896-15903.

[10] Daillant, J. and Gibaud, A. (Eds.), 1999, *X-ray and Neutron Reflectivity, Principles and Applications* (Berlin Springer).

[11] Sakurai, K. (Ed.), 2009, *Introduction to X-ray Reflectivity* (Kodansha Scientific).

[12] Sinha, S. K., Sirota, E. B., Garoff, S., and Stanley, H. B., 1988, X-ray and neutron scattering from rough surfaces, Phys. Rev. B 38, 2297-2311.

[13] Vidal, B and Vincent, P., 1984,.Metallic multilayers for x rays using classical thin-film theory, Applied Optics 23, 1794-1801.

[14] Boer, D. K. G., 1995, X-ray reflection and transmission by rough surfaces, Phys. Rev. B 51, 5297-5305.