Determination of fine layer structure in Ni/C multilayer using soft X-ray resonant reflectivity

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A fine layer structure in the Ni/C multilayer (3−4 nm/6−7 nm) is deposited by magnetic sputtering by combining soft X-ray resonant reflectivity curve at 4.48 nm and grazing incidence X-ray reflectivity (GIXR) curve at 0.14 nm. It is found that the thickness of Ni-on-C interface is much rougher than C-on-Ni interface. By analyzing the optical constants, it shows that the interface in the Ni/C multilayer that of system is a mixture of Ni and C atoms; the Ni and C in multilayer system have excellent stability, and no interlayer is formed.

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Advantages in using Ni-like soft X-ray lasers have been mentioned by various materials[1−3]. Ni-like Ta laser at 4.48-nm wavelength is known to provide possible source in the "near water window region"[4] (4.4−6.7 nm), thus making multilayer reflector at this wavelength be a popular research topic since it is an indispensable component of soft X-ray optical systems. Multilayer mirrors are fabricated by the alternating layers of low-Z (spacer) and high-Z (absorber) materials. At wavelength range of 4.3−6.7 nm, C, whose K-edge is 4.3 nm, has been observed as the best spacer when used with heavy metals like U, W, and 3d transition metals (i.e., Fe, Co, Ni, and Cr) from the absorber of the material group. To our knowledge, a normal incidence reflectivity of 13% obtained at 4.5 nm, which is less than a third of the theoretical value, is the most appropriate highest value at this wavelength range[5,6] since both inter-diffusion and formation of compounds between the two materials make it difficult to obtain a smooth interface[7]. Hence, the study on layer structure and interface properties is extremely important at this wavelength. Imperfections resulting from the formation of compound and interlayer have been found in other multilayer systems, such as Mo/Si using transmission electron microscopy (TEM), X-ray photoelectron spectroscopy (XPS), and X-ray emission spectroscopy (XES)[8−10]; however, these methods are all known to be destructive.

In the letter, we analyze the fine structure of Ni/C multilayer system using a non-destructive combined with soft X-ray resonant reflectivity curve at 4.48 nm and grazing incidence hard X-ray (GIXR) curve at 0.14 nm.

Ni/C multilayers were deposited on the quartz substrate by a DMD-450 magnetron sputtering system with Ni in direct current (DC) and C in radio frequency (RF) mode. The sputtering powers and currents were $P_{\rm Ni}$= 460 W, $I_{\rm Ni}$= 1 A, $P_{\rm C}$= 500 W, and $I_{\rm C}$= 0.6 A. A relatively large sputtering power of Ni was used in order to create an interlayer. Multilayers were designed based on the parameters, where period thickness $\Lambda = 10$ nm, thickness ratio of W layer to period $\Gamma = 0.4$, and number of period $N = 10$. Base pressure was 2×10^{-3} Pa, and working gas (Ar) was at a pressure of 0.2 Pa. Soft X-ray resonant reflectivity was measured at a wavelength of 4.48 nm using the 3W1B beam line of Beijing Synchrotron Radiation Facility (BSRF). Grazing incidence reflectivity was measured at 0.14 nm wavelength using the U7B beam line of the National Synchrotron Radiation Laboratory (NSRL) in Hefei, China.

All the reflectivity calculations utilized a recursion method given by Parratt[11]. For the s-polarized incident wave, the Fresnel reflection coefficient of the interface between the jth and j+1th layers can be given by

$$F_{j,j+1} = \frac{E_j^{\rm R}}{E_j} = \frac{k_j - k_{j+1}}{k_j + k_{j+1}} \qquad (1)$$

with

$$k_j = \frac{2\pi}{\lambda}\left(n_j^2 - \cos^2\theta\right)^{1/2},$$

where E_j and $E_j^{\rm R}$ are the amplitudes of electric vector of incident and reflected waves on the interface between jth and j+1th layers, respectively. Herein, n_j is the complex refraction index of the jth layer, and θ is the incident angle. In the X-ray region, complex refractive index for matter is $n = 1-\delta-{\rm i}\beta$, where δ and β are the optical constants. The recursion relation of the Fresnel reflection coefficient of different interfaces can be given by

$$R_{j,j+1} = \alpha_j^2 \frac{R_{j+1,j+2} + F_{j,j+1}}{1 + R_{j+1,j+2}F_{j,j+1}} \qquad (2)$$

with

$$\alpha_j = \exp(-{\rm i}k_j d_j),$$

where d_j is the thickness of the jth layer. The reflectivity of the multilayer system can then be written as

$$|R_{1,2}|^2 = \frac{I_{\rm R}}{I_0} = \left|\frac{E_1^{\rm R}}{E_1}\right|^2. \qquad (3)$$

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The recursion method starts with the bottom layer with $R_{N,N+1}= 0$ since there is no reflection from the thick substrate. In real multilayer systems, an error factor must be introduced since the layer is incompletely flat. The factor, which is to be multiplied with Eq. (2), can be written as

$$EF = \exp(2k_{j,z}k_{j+1,z}\sigma^2), \quad (4)$$

where roughness (σ) is the root mean square (RMS) deviation of the layer with respect to a completely flat layer.

To analyze the multilayer system, the imperfect interfaces in real multilayer systems are usually handled by a statistical method or the incorporation of an interlayer between two-layer systems[7]. In the statistic method, σ was introduced in the calculation of the two-layer model, and it was the only parameter used to describe the interface imperfections in the calculation. Hence, the σ denoted a statistical parameter in this model since it included all the effects of the interface imperfections, such as roughness of layer, interdiffusion of layer, and formation of compounds. In the four-layer model, an interlayer was incorporated at each interface. The thickness and optical constants of the interlayer indicate the occurrence of interdiffusion and formation of a compound. σ was also introduced in the model to describe whether the layer was sharp or not. In this letter, through fabrication, both models were used to describe actual Ni/C multilayer systems. By comparing and analyzing the models' results and measured curves, the fine structure of the multilayer system was obtained.

Figures 1(a) and (b) were observed to be the best fitted results using the four-layer model and two-layer model, respectively. Schemes of the two models are also shown in the figure. We can see that both fitted results agreed well with measured data. Hence, it is inappropriate to conclude which model was better in describing the real multilayer system, thus necessitating further analysis. Tables 1 and 2 show the layer structure and optical constants obtained from the two fittings. Period thickness obtained from the two fittings deviated dramatically from the designed structure due to the limitation of our experimental conditions. Optical constants obtained by both model deviated from the values given by Henke's database[12] (in brackets). For results obtained by the two-layer model, the real part of optical constants (dispersion) of layers changed dramatically compared with the values given by Henke's database, possibly because of its rapid variations resulting from changes in composition near the K-edge of C. This was also similar to the change in the imaginary part of the refraction index (absorption). Values of the Ni layer decreased while C layer increased, which was mainly caused by interdiffusion. For results obtained by the four-layer model, deviation of optical constants exhibited the same trend as the two-layer model. Optical constant of assumed interlayer 1 (Ni on C) was almost equal to that of the Ni layer, indicating that this

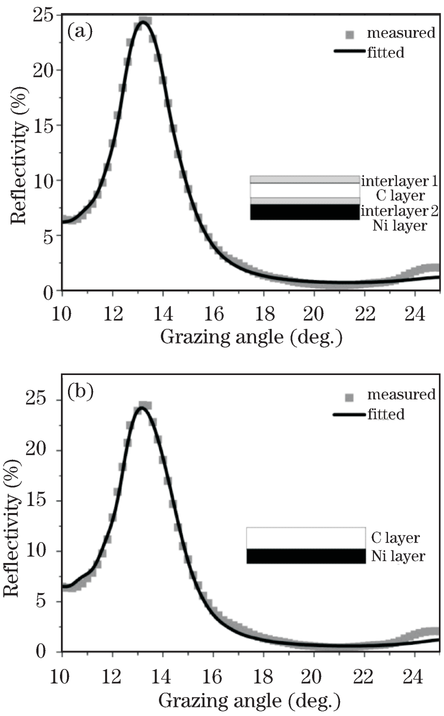

Fig. 1. Best fitted results using (a) the four-layer and (b) two-layer models at 4.48 nm.

Table 1. Fitted Results From the Two-layer Model

Two-layer Model	Thickness (nm)	$\delta(\times10^{-3})$	$\beta(\times10^{-4})$
Ni layer	3.643	19.6 (14.1)	45.5 (68.0)
C layer	7.071	3.9 (1.3)	4.03 (1.54)
		Roughness (nm)	
Interface of C on Ni		0.446	
Interface of Ni on C		0.640	

Table 2. Fitted Results From the Four-layer Model

Four-layer Model	Thickness (nm)	Roughness (nm)	$\delta(\times10^{-3})$	$\beta(\times10^{-4})$
Ni on C	0.799	0.309	19.7	64.3
Ni	2.563	0.478	20.3 (14.1)	50.1
C on Ni	0.659	0.450	8.6	10.1
C	7.488	0.627	3.8	5.5

assumed interlayer was actually a Ni layer. The optical constant of interlayer 2 (C on Ni) varied significantly as compared with the Henke constants, indicating that this assumed interlayer is a mixture of Ni and C. However, numerous variables in a fitting may be caused in unreliable results. Thus, further analysis was necessary.

Figure 2 shows the calculated results of the Ni/C mutilayer system at 4.48 nm with optical constants from the database of Henke *et al*[12]. We used the four-layer model and changed the composition (e.g., NiC and Ni_3C) of the interlayer in the calculation. Model parameters were $N = 10$, $d_{\mathrm{Ni}} = 3$ nm, $d_{\mathrm{C}} = 5$ nm, and $d_{\mathrm{interlayer}} = 1$ nm. The interfaces of each layer were assumed as sharp. From

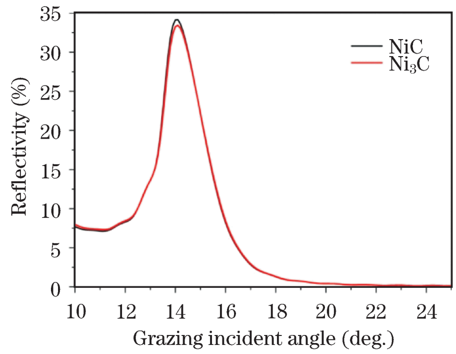

Fig. 2. Comparison of the calculated reflectivities of the Ni/C multilayers with different interlayer composition at 4.48 nm.

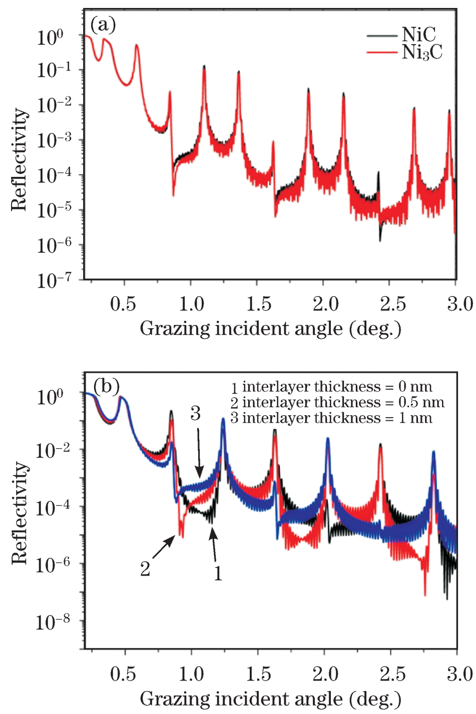

Fig. 3. Comparison of the calculated reflectivities of Ni/C multilayers at 0.14 nm. (a) Multilayers with different interlayer composition and (b) Multilayers with different interlayer thicknesses.

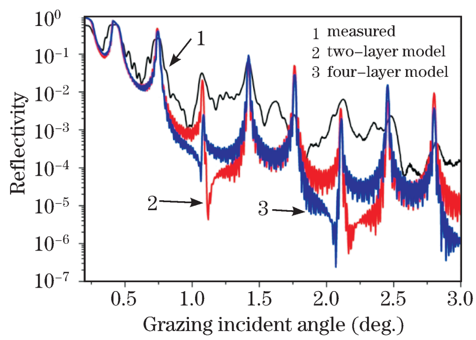

Fig. 4. Comparison of the measured XRR at 0.14 nm and the calculated results using structure parameters obtained from the two fittings.

the results, we could see that the reflectivity obviously changed due to the different composition of interlayer near the K-edge of C. This was because X-ray reflectivity (XRR) showed a resonant behavior in the absorption edge due to the rapid variation of the optical constants. It was reported that the soft X-ray resonant reflectivity achieved subnanometer scale sensitivity to the roughness and interlayer formation at the interface[13]. Thus, by fitting the measured reflectivity to the model reflectivity at the absorption edge (Fig. 1), the layer structure, including information on the interface, can be obtained.

Figure 3 shows the calculated results of the Ni/C mutilayer system at 0.14 nm with optical constants from the database of Henke *et al*[12] The model used in the calculation of Fig. 3 (a) is the same as in Fig. 2. In the calculation of Fig. 3 (b), the reflectivities with different thicknesses of the interlayer were calculated. The parameters of the model with no interlayer and sharp interface were $N = 10$, $d_{\mathrm{Ni}} = 4$ nm, and $d_{\mathrm{C}} = 6$ nm. As thickness of interlayer changes, the corresponding thickness was added to or subtracted from the Ni and C thicknesses to in order to retain the period constant. The calculation results of Fig. 3(a) show that XRR was not so sensitive to the composition change of the interface at wavelength of 0.14 nm. This was probably due to the small difference of optical constants between different compositions at the wavelength far away from the absorption edge. Figure 3(b) shows the grazing reflectivity curves with different thicknesses of the interlayer. The first order diffraction peaks were nearly the same. However, higher order peaks exhibited different features. Some peaks appeared or disappeared with variations in interlayer thickness. This proved that the XRR remained sensitive to the structure's parameters, such as thickness of interlayer in the hard X-ray region.

Structure parameters (i.e., thickness and roughness) obtained from the fittings of two models were used to calculate grazing incidence reflectivity at 0.14 nm. As discussed above, XRR at 0.14 nm was sensitive to the structure parameters and not with the composition of the interlayer. The composition of interlayer was assumed to be Ni_3C. The calculated and measured results are shown in Fig. 4. We could see that most peaks and the reflectivity of the three curves agreed well, except for the third order diffraction peak. This peak in the calculation curve from the four-layer model disappeared, which dramatically differed from the measured results. In comparison, results from the two-layer model agreed well with the measured curve, including the position and value of the third diffraction peak. This phenomenon indicates that the two-layer model was more similar to the real layer structure, and that there is no interlayer formation at the Ni/C interface, which was identical with the results in the optical constants analysis. This results also show that the Ni/C multilayer system had excellent stability since the parameters set in the fabrication were aimed to form an interlayer by interdiffusion.

In conclusion, the layer structure of the Ni/C multilayer system is analyzed using a soft X-ray resonant reflectivity at 4.48 nm, which has subnanometer scale sensitivity. Combined with the XRR at 0.14 nm, analysis on the structure of the multilayer system shows that there is no interlayer formation at the interface. Moreover, the Ni/C multilayer system achieves excellent stability. The optical constants of each layer in the two-layer model at 4.48 nm are obtained; these are useful in the further research of Ni/C mutilayer system at this wavelength.

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