J. Synchrotron Rad. (1998). **5**, 711–713

Fabrication and characterization of multilayer supermirrors for hard X-ray optics

K. Yamashita,* K. Akiyama, K. Haga, H. Kunieda, G. S. Lodha,† N. Nakajo, N. Nakamura, T. Okajima, K. Tamura and Y. Tawara

Department of Physics, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8602, Japan. E-mail: yamasita@satio.phys.nagoya-u.ac.jp

(*Received 4 August 1997; accepted 15 December 1997*)

Multilayer supermirrors stacked with three sets of Pt/C combinations have been fabricated on a flat float-glass and conical replica foil mirror using a magnetron DC sputtering system, and applied to X-ray optical systems in the hard X-ray region. The design of the supermirror is optimized to obtain the highest integrated reflectivity in the energy band and at the grazing angle concerned. X-ray reflectivities of 30% in the 25–35 keV band at an incidence angle of 0.3° were obtained.

Keywords: multilayer supermirrors; hard X-rays; X-ray reflectivity; interfacial roughness.

1. Introduction

The use of total external reflection by single-layer-coating mirrors has been made in grazing-incidence X-ray optical systems, such as X-ray telescopes, X-ray microscopes and synchrotron radiation beamlines in the energy region 0.1–10 keV (Michette, 1986). The critical angle of the mirror defines the highest energy of the reflected X-rays. For example, a gold mirror has a critical angle of 0.55° at 8 keV (1.5 Å). Beyond this angle the X-ray reflectivity rapidly falls, so that for practical use the energy region is limited to below 10 keV. Consequently, in the hard X-ray region, the incidence angle becomes extremely small, as it is linearly proportional to the X-ray wavelength.

Multilayer coatings on the reflecting surface have the advantage of enhancing the reflectivity at incidence angles beyond the critical angle, although the energy band becomes narrow under the constraint of the Bragg condition (Lodha *et al.*, 1994). The application of depth-graded multilayers, known as multilayer supermirrors, makes it possible to extend continuously the energy into the hard X-ray region, since the photoelectric absorption in multilayers becomes less effective above 10 keV (Joensen *et al.*, 1993). Therefore, a hard-X-ray imaging optical system will be available by making use of supermirrors (Christensen, 1997; Yamashita, 1997). We present the design, fabrication and characterization of supermirrors and discuss their properties.

† On leave from Centre for Advanced Technology, Indore, India.

2. Design of supermirrors

The reflectivity of a multilayer *versus* the wavelength (λ) and incidence angle (θ) is calculated by the Fresnel equation as a function of thickness of a layer pair (the periodic length, d), number of layer pairs (N), thickness ratio of the heavy and light elements ($\Gamma = d_H/d$; $d = d_H + d_L$), optical constants and interfacial roughness (σ). Detailed calculation methods can be found in various textbooks (for example, Michette, 1986). The peak reflectivity is obtained at values of λ and θ satisfying the Bragg condition. The total layer thickness (Nd) is estimated by referring to the penetration depth in the multilayer structure. A Pt/C combination was selected to obtain the highest reflectivity in the hard X-ray region with the least number of layer pairs (Lodha *et al.*, 1994). Pt is chemically stable as used for X-ray mirrors. First-order peak reflectivities of Pt/C multilayers were calculated against d and N, with $\Gamma = 0.4$ and $\sigma = 0$ or 3 Å and the incidence angle 0.3°, and are shown in Fig. 1. The rate of reduction of reflectivity caused by σ is expessed by the Debye–Waller factor as $\exp[-(4\pi\sigma\sin\theta/\lambda)^2]$.

The reflectivity of supermirrors is calculated in a similar way by stacking multilayers with different sets of d and N to obtain the highest integrated reflectivity and flat-peak reflectivity in the wide energy band. d decreases from the outermost layer to substrate, whereas N increases, since the higher-energy X-rays have greater penetration depth. Photoelectric absorption becomes less effective in the multilayer structure with increasing X-ray energy. Therefore the Bragg condition is continuously satisfied in the wide energy band without significant reduction of reflectivity by making use of a supermirror (Joensen *et al.*, 1993). Referring to Fig. 1, we have designed a supermirror stacked with three sets of Pt/C multilayers. The three multilayers have d = 46–50, 40 and 36 Å, and N = 5, 8 and 13, respectively. Γ is fixed at 0.4 and the incidence angle is 0.3°. The reflectivity is shown in Fig. 2. It is obvious that higher-energy X-rays are reflected by smaller d-spacing layers and attenuated by upper layers with larger d-spacing. The observed reflectivity would be reduced by an increased interfacial roughness, as shown by the dashed curve in Fig. 2.

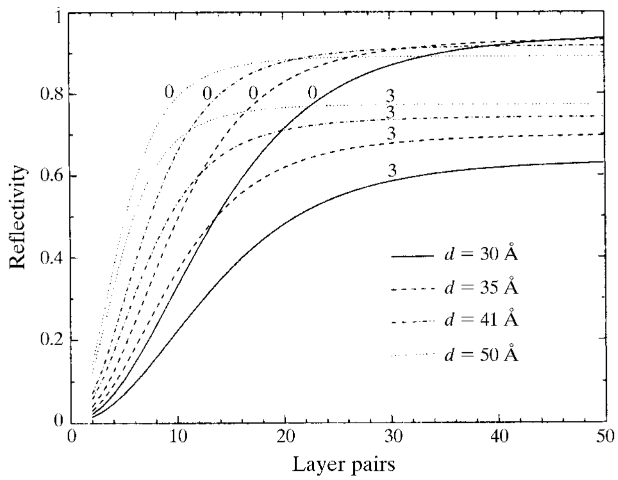

Figure 1
First-order peak reflectivities of Pt/C multilayers *versus* number of layer pairs calculated for four different periodic lengths (d) with σ = 0 and 3 Å at an incidence angle of 0.3°. d values of 30, 35, 41 and 50 Å correspond to X-ray energies of 40, 35, 30 and 25 keV, respectively. σ values are marked on each curve.

Table 1
First-order peak reflectivity of Pt/C multilayers.

Pt layer thickness (Å)	C layer thickness (Å)	Periodic length d (Å)	Number of layer pairs N	Cu $K\alpha$ observed reflectivity (%)	Interfacial roughness σ (Å)
21.6	26.4	48.0	20	68	3.5
17.4	26.1	43.5	20	60	3.0
16.8	26.2	43.0	50	68	3.2
12.0	19.5	31.5	25	38	4.0
13.7	13.7	27.4	60	40	3.0
10.9	16.3	27.2	60	32	4.0
7.5	17.5	25.0	60	18	5.0

3. Fabrication

Multilayers and supermirrors thus designed were fabricated on float glass and conical-shaped replica foil mirrors by a magnetron DC sputtering system, specially designed for deposition on the inner surface of conical or aspheroidal mirrors, with a grazing angle of less than 1° matched to the configuration of an X-ray telescope (Tawara *et al.*, 1996). Two pairs of sputtering targets facing each other are mounted on the upper and lower parts of the vacuum chamber. Ar plasma is produced and maintained at 1 mtorr. The substrates are placed on the rotation table which moves up and down and rotates around the targets. Deposition rates are controlled by the ion current and the rotation speed of the substrate. The layer thickness is monitored by a quartz crystal oscillator. Substrates used in this system are 200–500 mm in diameter and 150 mm in height. A replica foil mirror was composed of 125 µm-thick Al, 15 µm epoxy and 0.1 µm Au replicated from a glass mandrel, as used in an X-ray telescope (Serlemitsos, 1997). The size of the conical replica foil was 100 mm long × 150 mm wide with a radius of curvature of 100 mm and a tilting angle of 0.3°. Deposited layer thicknesses were controlled with an accuracy of within 1 Å. The uniformity of the layer thickness in the vertical direction is adjusted to be less than 2% by using a specially shaped mask mounted in front of a substrate. Multilayers with the same parameters are reproduced with an accuracy of within 1%. The most important factor is the interfacial roughness, which depends on the surface roughness of substrates, the material combinations, the substrate temperature and the deposition rate. An interfacial roughness of at best 3 Å was obtained, which is comparable to the surface roughness of float glass.

4. Characterization

Characterization of multilayers is carried out by measuring the reflectivity against X-ray energy and incidence angle with the characteristic Al $K\alpha$ (1.5 keV), Cu $K\alpha$ (8.04 keV) and Mo $K\alpha$ (17.5 keV) X-rays and continuum X-rays up to 50 keV. The measurement system in our laboratory consists of a rotary-target X-ray generator connected by a 10 m duct to a measurement chamber installed with a θ–2θ rotation system. A thin-window gas-flow proportional counter and a CdZnTe solid-state detector are mounted on the rotation arm. The incident beam is collimated to an angular divergence of 20 arcsec by a 0.2 mm-diameter pinhole and monochromated by an absorption-edge transmission filter and multilayer reflector. The observed reflectivities of Pt/C multilayers deposited on float glass for Cu $K\alpha$ are summarized in Table 1, which shows that the interfacial roughness and the rate of reduction of peak reflectivities become larger when the thickness of the Pt layer is less than 12 Å.

The reflectivity measurements of a supermirror stacked with the three sets of multilayers mentioned above were carried out with Cu $K\alpha$ and continuum X-rays of 15–45 keV at a fixed incidence angle. The observed reflectivity for Cu $K\alpha$ is shown in Fig. 3. Fitting of the second-order peak is improved by changing Γ and σ, but the fitting of the total external reflection in the small-angle region becomes poor. The hard X-ray reflectivity of a supermirror is shown in Fig. 4. This is simply derived from the intensity ratio of the reflected spectrum to the incident spectrum measured with a CdZnTe solid-state detector which has an energy resolution (FWHM) of 3% at 30 keV. The average reflectivity obtained was 30% in the energy band 25–35 keV at 0.3° incidence angle. Calculated reflectivity, shown with the

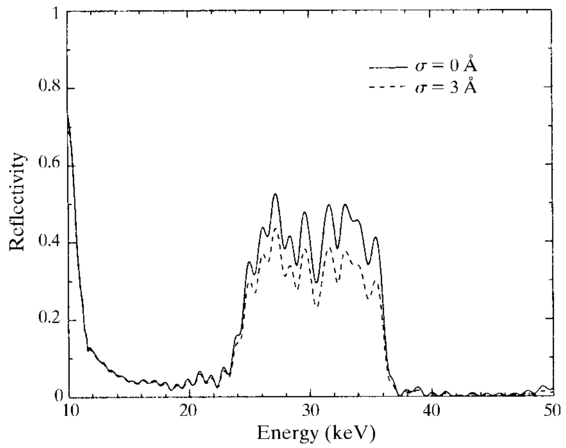

Figure 2
Calculated reflectivity of a supermirror stacked with three sets of Pt/C multilayers in the 10–50 keV region at the incidence angle of 0.3° and Γ = 0.4. The multilayers had d = 46–50, 40 and 36 Å, and N = 5, 8 and 13, respectively. Solid curve, σ = 0 Å; dashed curve, σ = 3 Å.

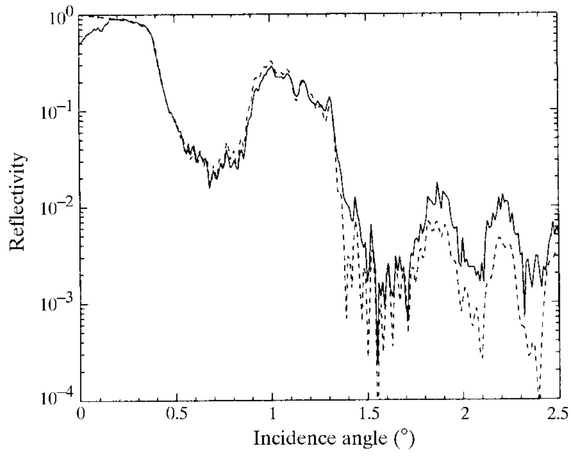

Figure 3
Observed reflectivity of the supermirror shown in Fig. 2 and deposited on float glass for Cu $K\alpha$ (solid curve). Calculated reflectivity with σ = 3.8 Å is shown with a dashed curve.

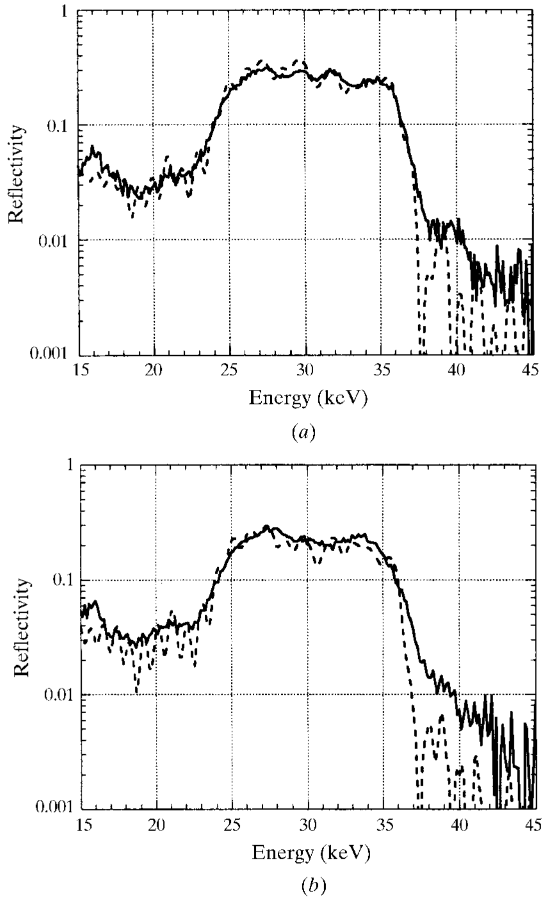

Figure 4
Observed reflectivity of the supermirror shown in Fig. 2 and deposited on (*a*) float glass and (*b*) replica foil in the 15–45 keV region at the incidence angle of 0.3°. The dashed curve shows calculated reflectivities with (*a*) σ = 4.1 Å and (*b*) σ = 5.3 Å.

dashed curve, has to be smeared out with the energy resolution of the detector in order to be compared with the observed reflectivity.

5. Discussion

In order to minimize the interfacial roughness of the multilayers, we have investigated the dependence of reflectivity on the deposition rate, vacuum pressure, substrate temperature and number of layer pairs. It turns out that the interfacial roughness can be understood as a replication of the surface roughness of the substrate σ_s (3 Å for float glass and 4–5 Å for replica foil) and/or the intrinsic property of a material combination. According to our experimental results, the interfacial roughness becomes smaller with increasing numbers of layer pairs and attains the value of σ_s at the saturated peak reflectivity. This effect seems to be caused by the density of the ultrathin Pt layer, which can be shown to be 80% of the bulk density. This means that the lower density of Pt makes the interfacial roughness smaller. The temperature of the replica foil has to be kept below 323 K during the deposition to protect the surface from damage. The interfacial roughnesses obtained were 3 Å for multilayers on float glass, 4 Å for supermirrors on float glass and 5 Å for supermirrors on replica foil. These differences are caused by the properties of the multilayer structure and, partly, the surface roughness of the substrates.

Supermirrors thus designed and fabricated have flat reflectivity in the 25–35 keV band at the incidence angle of 0.3°. Ideal reflectivity (σ = 0 Å) is estimated to be 50%, but in practice is reduced to 30% due to the interfacial roughness. If the energy region extends up to 40 keV, it is necessary to add a further set of multilayers with d = 33 Å and N = 18 layer pairs. Our design method aims at making the energy band as wide as possible while keeping the reflectivity at 30–40%. The minimum periodic length could be d = 25 Å for practical use, which corresponds to 50 keV at 0.3° incidence.

6. Summary

Multilayer supermirrors have been successfully deposited on float glass and replica foil mirrors by a magnetron DC sputtering system. We were able to obtain an almost flat reflectivity of 30% in the energy band 25–35 keV. This result is promising for the construction of a hard X-ray telescope. However, more effort still needs to be made to improve the reflectivity and extend the energy band.

The authors would like to thank Dr P. J. Serlemitsos, NASA Goddard Space Flight Center, who kindly supplied us with replica foil mirrors. This work was supported in part by a Grant-in-Aid for Scientific Research on Specially Promoted Research, contract No. 07102007, from the Ministry of Education, Science, Sports and Culture, Japan.

References

Christensen, F. E. (1997). *Proceedings of the Workshop on the Next Generation of X-ray Observatories*, *Leicester X-ray Astronomy Group Special Report XRA97/02*, edited by M. J. L. Turner & M. G. Watson, pp. 133–145. University of Leicester, UK.

Joensen, K. D., Hoghoj, P., Christensen, F. E., Gorenstein, P., Susini, J., Ziegler, E., Freund, A. K. & Wood, J. L. (1993). *Proc. SPIE*, **2011**, 360–372.

Lodha, G. S., Yamashita, K., Suzuki, T., Hatsukade, I., Tamura, K., Ishigami, T., Takahama, S. & Namba, Y. (1994). *Appl. Opt.* **33**, 5869–5874.

Michette, A. G. (1986). *Optical Systems for Soft X-rays*. New York: Plenum.

Serlemitsos, P. J. (1997). *Proceedings of the Workshop on the Next Generation of X-ray Observatories*, *Leicester X-ray Astronomy Group Special Report XRA97/02*, edited by M. J. L. Turner & M. G. Watson, pp. 123–131. University of Leicester, UK.

Tawara, Y., Yamashita, K., Kunieda, H., Haga, K., Akiyama, K., Furuzawa, A., Terashima, Y. & Serlemitsos, P. J. (1996). *Proc. SPIE*, **2805**, 236–243.

Yamashita, K. (1997). *Proceedings of the Workshop on the Next Generation of X-ray Observatories*, *Leicester X-ray Astronomy Group Special Report XRA97/02*, edited by M. J. L. Turner & M. G. Watson, pp. 115–121. University of Leicester, UK.