ИНСТИТУТ ЯДЕРНОЙ ФИЗИКИ
им. Г.И. Будкера СО РАН

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MULTILAYER MIRROR ON COPPER-NICKEL SUBSTRATES FOR INTENSE X-RAY BEAMS

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Abstract

The micro probe for X-ray fluorescence measurements in the photon energy range 6-15 keV is developing at the Institute of Nuclear Physics, Novosibirsk. Multilayers deposited on the spherical copper-nickel substrates will be used as monochromators and focusing elements. The micro probe will be designed according to the Kirkpatrick-Baez optical scheme. The metallic substrates will be used because of high heat loads of the "white" SR beam from the VEPP-4M storage ring.

The possibility of manufacture the multilayers with high performance on the metallic substrates is discussed. The multilayers were deposited by means of pulsed laser evaporation method. The influence of substrate roughness on interfacial roughness of the multilayer was studied. The procedure of manufacture of copper-nickel substrates is described. The reflection characteristics of the W/C multilayers deposited on the copper-nickel substrates are presented.

1. INTRODUCTION

Multilayer interference coatings, initially intended for soft x-ray radiation (the wavelength is 1-30 nm) can be successfully used in the of hard X-radiation range (λ~0.1 nm) as coatings for grazing incidence mirrors. Unlike the mirrors of total external reflection, these coatings, first, increase the working angles by a factor of 2-5. Second, they, reduce the radiation load at the next optical element by several times, according to calculations in [1], and, third, make it possible to selectively decrease the reflection in undesirable orders by choosing the fraction of a highly-absorbing substance in the period.

In work with intense SR beams (the power density is 10 W/cm^2 and higher) the problem of mirror heating arises [2]. To reduce the heating, it is necessary to use efficient cooling systems and materials with high thermal conductivity. The choice of such materials is very limited because they must be well polished too. That is why silicon carbide and copper, electrolytically coated by nickel, are practically major candidates [3,4].

From the viewpoint of polishability and thermal properties, silicon carbide is an ideal material. Its disadvantages are that it is difficult and expensive in fabrication. More accessible is the manufacture of copper-nickel mirrors. However, as is shown in [5] and as our experience in polishing such mirrors shows it is not possible to reach the surface roughness better than 1 nm. Acceptable at the critical angle, this magnitude drastically increases when increasing the working angles.

As shown in [6], the interfacial roughness of multilayers prepared by means of pulsed laser evaporation (PLE) method is substantially less than that of the substrate. The present work is focused on the possibility to manufacture highquality multilayer coatings by the PLE method on copper-nickel substrates. The first part of the paper deals with the technique for measuring the roughness of substrates. The roughness values for 7 samples are presented. In the second part, the dependence of the interfacial roughness of the multilayers on the substrate roughness, on quartz multilayers as an example, is studied. The technique for defining the roughness is presented in detail. The third part describes at great length the technique for manufacturing copper-nickel substrates and gives the reflection characteristics of W/C multilayers on these substrates. The fourth part reports the results of exposure of the multilayers by the "white" SR beam at a power density of 1 W/cm^2.

2. MOTIVATION

X-ray fluorescent trace element microprobe with the using synchrotron radiation (SR-XRF microprobe) is very popular now, due to environmental application. The most promising fields of such type research are next:

- single particle analysis in environmental aerosol samples, which makes easier the task of identification of the source of the atmospheric pollution;
- analysis of the distribution of elements along the human hair, which shows the dynamic of accumulation of the harmful pollutions in organism.

For these applications SR-XRF microprobe has same advantages in minimal detection limit and range of the elements to be analyzed in comparison with electron and proton microprobe.

For analysis are mentioned above it is necessary to have a spatial resolution ~ 5 µm. In doing so, for obtaining detection limit better than 1 ppm, it is requisite that photons flux in the region of analysis become more than $5 \cdot 10^{10}$ photons/s (if count rate of fluorescence photons about 10 Hz).

The ability of the producing of multilayer spherical mirrors for energy of photons 16-17 keV and incidence angle about 0.5° makes possible to achieve the flux of a photons that required. The Kirkpatrick-Baez scheme is the simplest one for such type applications. However the large radiation dose to the fist mirror (0.6 W/mm^2 in SR beam cross-section in case of VEPP-4 storage ring: I=10 mA, E_e =4 GeV, 5 poles wiggler 1.4 T, without preliminary monochromatization, at a distance of 40 m from source) makes difficult to use the conventional multilayer mirrors on quartz or silicon substrates. So it is very promising way to produce the multilayer mirrors on metallic substrate, which have a good thermal conductivity, resistance to radiation damage and availability.

The project of SR-XRF microprobe for experiments described above is in the designing now. The fist mirror on copper substrate has the radius 145 m for focusing SR beam in vertical direction. Because of the radiation power to second mirror is very smaller (approximately to 10^4 time) it is possible to produce the later on quartz substrate. The radius of second mirror is 45 m for the focusing in horizontal direction. The distance between second mirror and focus point is equal 200 mm, it is enough for the placement of the collimation system.

3. ROUGHNESS MEASUREMENTS

The substrate roughness was measured by the method of studying the angular dependences of the reflection coefficient $R_m(\Theta)$ at radiation wavelength λ = 0.154 nm. The influence of the roughness on the dependence R (Θ) was taken into consideration by introducing the Debye-Waller factor:

$$R_m(\Theta) = R_i(\Theta) \cdot \exp(-4\pi \cdot \sigma \cdot \sin\Theta/\lambda)^2, \qquad (1)$$

where R (Θ) is the reflection coefficient for an ideal surface and Θ is the grazing angle.

Fig. 1 shows the angular dependences R(Θ) for a copper-nickel mirror (sample 7): the dots are for experiment and solid lines are for the theoretical dependences corresponding to the following roughness values: 0; 0.8; 1.0, 1.2 and 1.4 nm. The best coincidence between experiment and theory is observed at σ=1

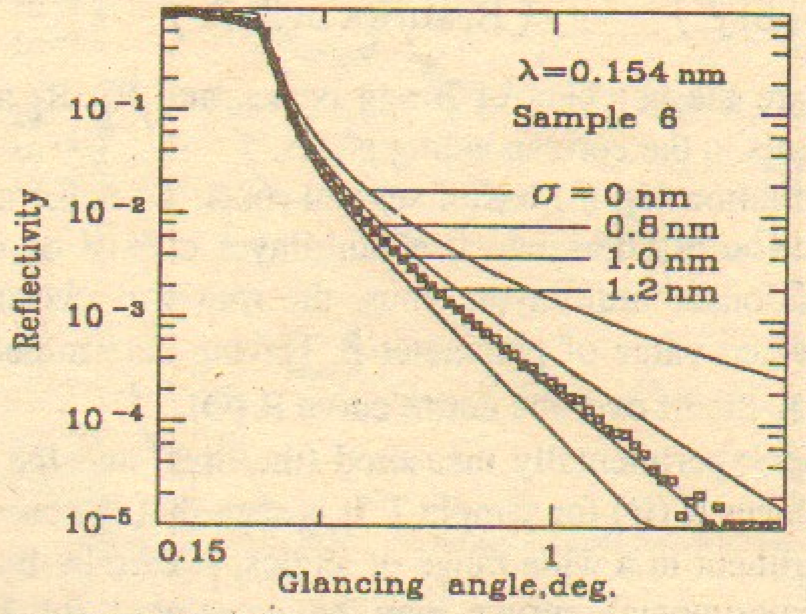

Figure 1. The theoretical for different roughnesses σ and experimental reflectivity curves for the copper-nickel substrate (sample 6). The best agreement between experiment and theory is at σ=1.0 nm, and the density of nickel 8.1 g/cm^3.

nm. In the calculation, the weight density of nickel was taken equal to 8.1 g/cm^3.

The table lists the measured values of roughness and the information for seven examined samples which served as substrates for the multilayers.

4. THE DEPENDENCE OF THE INTERFACIAL ROUGHNESS OF MULTILAYERS ON THE ROUGHNESS OF SUBSTRATES

The value of interfacial roughness σ was obtained by using formula 1. The parameters of the multilayers: period d and the portion of a highly-absorbing substance in the period β, which are necessary to define the reflection coefficient for an ideal structure, were determined from analysis of the angular dependences of the reflection coefficients R (Θ) (the radiation wavelength λ=0.154 nm, a Si (111) monochromator, the angular divergence of radiation is less than 30") for our samples. To find the period, we used the Bragg condition, with the refractivity in a multilayer substance taken in to account:

$$d = m\lambda/2\sin\Theta_m \cdot (1 - (2\bar{\delta} - \bar{\delta}^2)/\sin^2\Theta_m)^{1/2}, \qquad (2)$$

where Θ is the Bragg angle of m-th order and $\bar{\delta}$ is the dispersive addition to the refractive index, which is averaged over the structure. The parameter β was determined from the solution of the transcendent equation taken from [8]:

$$\left(\frac{R_l \cdot \sin^2(\pi \cdot j \cdot \beta) \cdot l^6}{R_j \cdot \sin^2(\pi \cdot l \cdot \beta) \cdot j^6}\right)^{j^2-m^2} - \left(\frac{R_m \cdot \sin^2(\pi \cdot j \cdot \beta) \cdot m^6}{R_j \cdot \sin^2(\pi \cdot m \cdot \beta) \cdot j^6}\right)^{j^2-l^2} = 0, \qquad (3)$$

where j, l and m are the numbers of Bragg peaks, and R_j, R_l and R_m are the reflection coefficients in the corresponding peaks.

As a rule, the relationship (3) has of several roots. To make a proper choice, the rates of deposition of films which a multilayer consist of were measured, prior to the deposition of multilayer. Thus, the root was chosen which is the nearest to the expected value of parameter β. Having determined the quantities d, β and σ, we made the fit over the entire curve R (Θ).

Fig.4 shows the experimentally measured (the dots) and the calculated (the solid line) dependences R (Θ) for sample 7. It is seen that the curve satisfactorily describes the experiment in a wide range of angles. A certain discrepancy in the theoretical and experimental curves may be accounted for by that, in the calculation, the transient layer between tungsten and carbon films and the fluctuations in films thickness were neglected. The following parameters were used in the calculation: W density 15 g/cm^3 , C density 2.6 g/cm^3 , d=5.7 nm, β=0.29, σ=0.4 nm.

Fig.2 presents the angular dependences of the reflection coefficients at radiation wavelength 0.154 nm on five samples deposited on quartz substrates of different roughness. The parameters of the mirrors are given in the Table 1.

Table 1. Parameters of samples.

Sample number	Substrate material	Substrate roughness σ_{sub}, nm	Multilayer structure	Number of periods	Period d,nm	β	Interfacial roughness, σ_{int}, nm
1	SiO_2	0.5	W/C	20	5.5	0.29	0.25
2	SiO_2	0.85	W/C	20	5.1	0.29	0.30
3	SiO_2	1.2	W/C	20	5.0	0.28	0.45
4	SiO_2	1.5	W/C	20	5.6	0.31	0.65
5	SiO_2	2.0	W/C	20	5.4	0.26	0.90
6	Cu-Ni	1.0	W/C	25	6.8	0.20	0.50
7	Cu-Ni	1.0	W/C	25	5.7	0.29	0.40

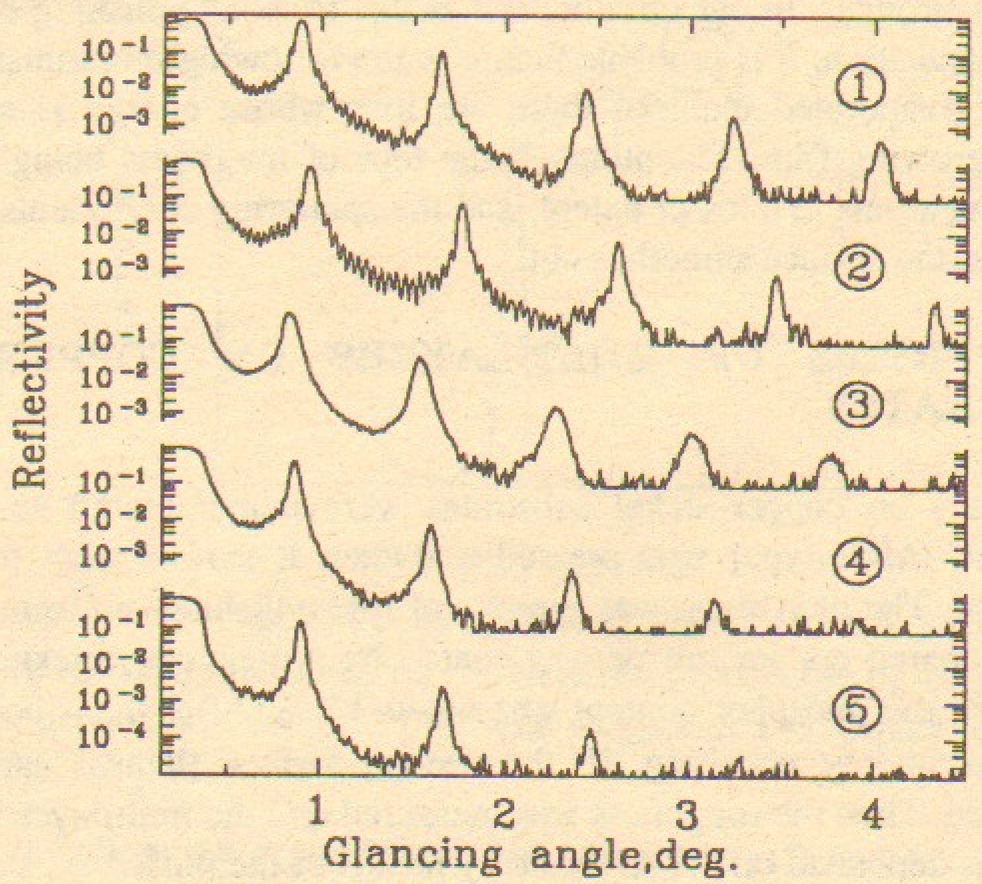

Figure 2 The reflectivity curves for samples 1, 2, 3, 4 and 5 deposited by pulsed laser evaporation method on the quartz substrates with roughness 0.5 nm, 0.85 nm, 1.2 nm, 1.5 nm and 2.0 nm, respectively.

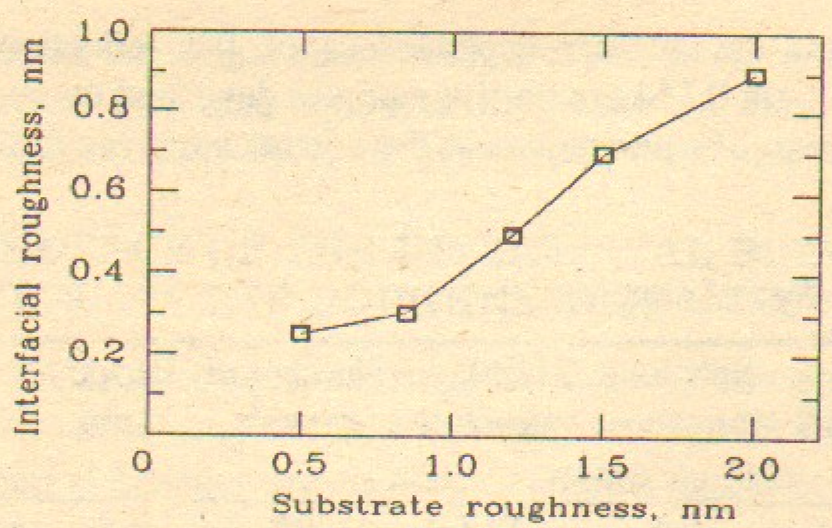

Figure 3. The interfacial roughness of multilayers versus the substrate roughness. This picture shows the "smoothing" effect provided by pulsed laser evaporation technology.

The interfacial roughness σ_{int} versus the substrate roughness σ_{sub} is shown in Fig.3. As is seen, before σ_{sub} = 0.9 nm this dependence is weak, but after this value it gets stronger. In our opinion, this is due to "ion" polishing of the layers during the deposition. It is probable that here the following mechanism works: in the flow of evaporated material there are ions whose energy is sufficient to sputter the growing film. The atoms at the tops of roughness being bounded to the remaining atoms to a lesser extent, and the sputtering coefficients for them is higher. Thus, the surface smoothes out.

5. PROPERTIES OF MULTILAYERS ON COPPER-NICKEL SUBSTRATES

Multilayers on copper-nickel substrates were manufactured as follows. A copper blank (MOb-type) was pressed and then it surface was treated by a diamond tool. The next stage was rough and fine polishing, up to mirror finish. The thus prepared copper mirror was coated by a nickel of thickness 100-150 µm, in which the phosphor content was about 12 %. After that, the mirror was subject to secondary polishing till the needed surface flatness and roughness were obtained. Then the roughness was measured and the multilayer interference structure was deposited on the substrate by means of the pulsed laser evaporation method.

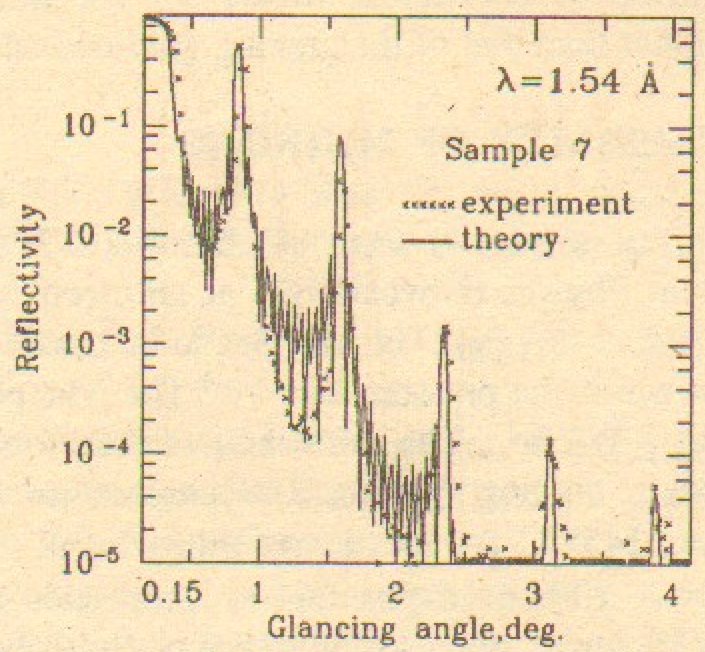

Figure 4. Reflectivity curves for sample 7. The fitting parameters are given in Table 1. The reflection coefficient in the first Bragg peak is 52 % at an angle 0.88°.

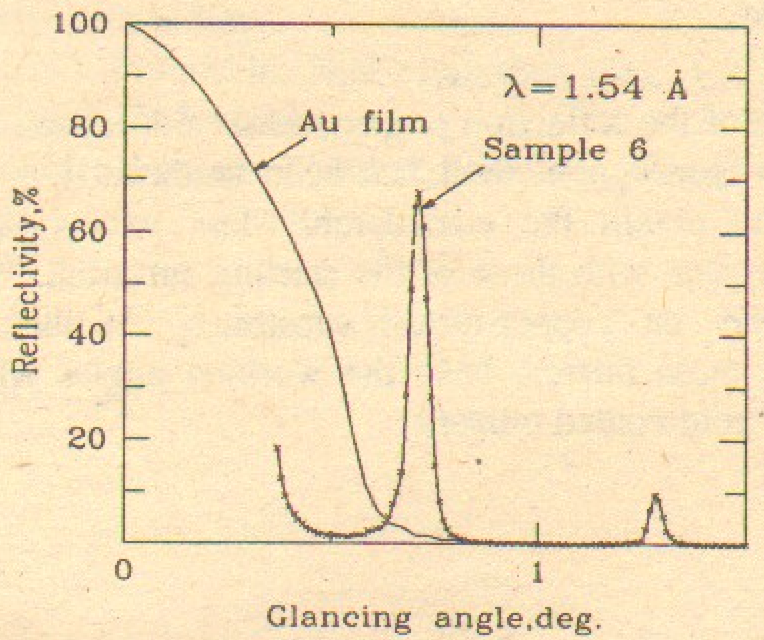

Figure 5. Reflectivity curves of X-ray with λ=0.154 nm: dots - experimental dependence for W/C multilayer on Cu-Ni substrate (sample 6), solid line the theoretical dependence for gold film with roughness 1 nm, thickness 50 nm and density 18.9 g/cm^3.

Figs. 4 and 5 show the dependences R(Θ) for samples 7 and 6, respectively. For sample 7 the reflection coefficient in the first Bragg peak is 52 % at angle Θ=0.88° and for sample 6, R=67% at Θ=0.71°. For comparison, the solid line in Fig.5 is for the dependence of the reflection coefficient from a gold film whose thickness is 50 nm, roughness is 1 nm and wight density is 18.9 g/cm^3. As is seen, at the same reflection coefficient (R=67 %) the working angle of the multilayer is twice higher than that of the grazing gold-coated mirror.

6 RADIATION STABILITY OF MIRRORS

The radiation stability of mirrors was studied on the VEPP-3 storage ring of the Institute of Nuclear Physics (Novosibirsk) at an electron energy of 2 GeV and a beam current 70-120 mA [9]. The samples to be examined were placed in a vacuum chamber in which the pressure was 10^{-3} Tor. The power density of the "white" SR beam was 1 W/cm^2 . The mirrors were fixed on the chamber wall, without the use of special cooling systems. The temperature was controlled by a thermocouple and was 145° C for the quartz mirrors and 75° for the copper-nickel mirrors. After a 10-hour exposure, the properties of the multilayers remained unchanged. However, one can see traces of photochemical dissociation of the substrate material on the quartz substrates. This is likely to bring about the destroy of mirror if the irradiation dose will be further increased.

7. CONCLUSION

The dependence of the reflection properties of multilayers, manufactured by the pulsed laser evaporation method has been studied. It is shown that this method enables the obtain the considerably less values of the interfacial roughness in comparison with those of the starting substrate. We manufactured the W/C multilayers on copper-nickel substrates. At the same reflection coefficient (67 %), these mirrors have the working angles which are 2 times larger than those of gold-coated mirrors.

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