DEUTSCHES ELEKTRONEN-SYNCHROTRON DESY

DESY SR-78/21
November 1978

SMOOTH MULTILAYER FILMS SUITABLE FOR X-RAY MIRRORS

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Multilayer coatings consisting of very smooth ReW and carbon films used as near normal incidence reflectors show theoretical performance in the 150-200Å wavelength region and should allow the fabrication of useful normal incidence mirrors for wavelengths as short as 50Å.

Multilayer coatings are widely used in the optical region to fabricate mirrors with enhanced reflectivity. In a similar way, natural crystals are good reflectors for x rays in the 1Å wavelength region if all lattice planes add in phase to the reflected wave (Bragg reflection). For this case, transmission is also drastically enhanced when the atoms of the crystal are located in the nodes of the standing waves produced by the superposition of the incident and the reflected wave (Borrmann effect).[1] Periodic multilayers of organic films produced by the Langmuir-Blodgett method can have larger lattice spacings than natural crystals and have been used for soft x rays.[2] Evaporation techniques give more flexibility in the choice of film thickness and periodicities. Multilayers made of Au and C have been used for the first experimental realization of such coatings for ultrasoft x rays and a reflectivity of 3% has been obtained with a 9-layer coating at $\lambda=190$Å for near normal incidence.[3] We believe that the previously reported performance was limited by the roughness of the gold films. Therefore, we searched for other suitable materials which give smoother films and better performance in a multilayer coating.

Thin films of the highest absorption index are alternated with very weak absorbers for an optimized multilayer reflector.[4-6] The strongly absorbing films have thicknesses between 30 and 50Å for a mirror with $\lambda_{max}=200$Å and normal incidence. Fig. 1a is an electron micrograph of a 50Å thick gold film, evaporated by an electron gun on a carbon foil. The well known network of voids[7] (width of voids around 50Å) between islands of gold is clearly visible. We cannot expect the reflectivity values calculated for smooth films from discontinuous island films like that in Fig. 1a. Attempts to produce smoother films led us to an ReW alloy as a best choice.[8] Both Re and W have a high Debye temperature, and therefore, little mobility of the atoms at room temperature is expected. The alloy grows as an amorphous film, therefore, crystallization of the film which might produce microcrystals does not occur. We found films of ReW to be smoother than pure Re or pure W films with the exact composition being uncritical within the 30-70% Re range. Fig. 1b shows a network structure on a much smaller scale than Fig. 1a; the width of the voids is in the 5-10Å range. The columnar growth

of the films which can be deduced from Fig. 1b is in qualitative agreement with a computer simulation of the growth of thin amorphous films of hard spheres.[9] Our films were evaporated in an electron gun evaporator at pressures around 10^{-6} Torr with the substrate cooled to 77°K.

Figure 2 shows reflectivity curves for our best ReW-C multilayer coating together with the calculated reflectivity of an optimized reflector using the previously measured optical constants of Re.[10] The fact that our experimental reflectivity for the multilayer is above the theoretical values indicates some difference between the optical constants of our films and those given in Ref. 10. The agreement between theory and experiment for the single film around $\lambda=200$Å indicates a better interface quality within the multilayer than at the surface of the single metal film that has been exposed to air.

Not all of our coatings have a performance equal to that shown in Fig. 2. The reasons are thickness errors which accumulated during the deposition process. For theoretical performance all periods of a multilayer have to contribute with equal phase to the reflected wave and the ReW films should be centered in the node of the standing wave formed by the superposition of the incident and reflected beam. We monitored the thickness of each film during deposition with an oscillating quartz crystal (at IBM) and tested the mirror later in a reflectometer[3,11] with synchrotron radiation (at DESY). Errors in the deposition of each film add at random with our present monitoring system and limit us to multilayer coatings of less than 10 layers for reasonable yield in the fabrication. The reflectivity of coatings with a fixed number of layers decreases for shorter design wavelength and more layers are required at shorter wavelength for the same reflectivity as that obtained at longer wavelengths.[5,6] Therefore, the random errors in the monitoring system present also a short wavelength limit at which practically useful reflectors can be made.

An improved monitoring system that measures the reflectivity during the deposition could eliminate the accumulation of random errors. The ultimate limit on the performance at shorter wavelength is determined by the roughness of the films. Figure 3 allows us to predict this limit for our presently used materials. The reflectivity of an 11-layer coating as measured on a computer controlled x-ray diffractometer[12,13] is plotted for a wavelength $\lambda=1.54Å$ versus the glancing angle $\theta=90°-\alpha$. For oblique angles of incidence, the reflectivity curves of coatings shift towards shorter wavelengths, and we observe the maximum reflectivity for $\lambda=1.54Å$ at glancing angles around $\theta=0.5°$. In addition, higher order interference maxima appear at larger glancing angles. We can compare the relative peak heights of these higher order maxima and find that for all our coatings the measured peak intensities for higher order maxima decrease faster than calculated for the multilayer with perfect boundaries. At $\theta=2°$ the measured reflectivity is about a factor two smaller than the calculated value. Under the assumption that the reflectivity R of a real coating is related to that of a perfectly smooth coating R_o by

$$R = R_o \exp - \left(\frac{4\pi\sigma \sin\theta}{\lambda} \right)^2, \quad (1)$$

we can determine an effective surface roughness σ from a fit, obtained by multiplying the theoretical curve with Eq. 1. The dashed curve in Fig. 3 shows such a fit with a value of $\sigma=3Å$. From this value and Eq. 1, we can predict the deterioration of the performance of a coating at any wavelength and angle of incidence. In Fig. 3 we have plotted at the top scale the wavelength $\lambda_{normal}=\lambda/\sin\theta$ (with $\lambda=1.54Å$) for which we can observe the same reduction in reflectivity for normal incidence. Using this scale our measurements show that the performance of multilayer coatings for normal incidence will start to deviate from the theoretical predictions for wavelengths below 100Å, but that useful reflectivities can still be expected around $\lambda=50Å$ (about half of the value calculated for perfectly smooth films). Our conclusions remain valid even for the case that the influence of the surface roughness has to be described by a more complex theory than Eq. 1, as long as wavelength and angle of incidence appear only as the parameter $\lambda/\sin\theta$ in the theory. From curves similar to those in Fig. 3 for 3, 5, 7, 9 and 11 layers, we observe no increase of the roughness for an increasing number of layers; however, it has to be noted that over 100 layers will be required for a mirror with good normal incidence reflectivity around $\lambda=50Å$.

Most of our results were obtained on substrates of commercially available, chemically polished 111-silicon which represents the best surface quality we could obtain. A comparison of 2 coatings evaporated simultaneously on a silicon wafer and on a super-polished quartz plate[14,15] gave $\sigma=9Å$ for the quartz plate. The reflectivity of our coatings was usually measured between 1 week and 1 month after the evaporation. No change in reflectivity for a sample stored in a desiccator was observed within 6 months. Samples stored in humid air, however, showed a discoloration for layers terminated with ReW, but not for those with C.

We conclude that multilayer films made of ReW and C are of sufficient quality to use them for the fabrication of reflecting elements in the soft x-ray region. An improved (in situ) monitoring system is required to produce such coatings for shorter wavelengths (more layers required) in practice. We see potential applications of such reflectors as high throughput, low resolution monochromators, beam deflectors, polarizers and for the fabrication of normal incidence reflecting microscopes and telescopes.

We thank I. Lovas and J. Wilczynski for polishing of the quartz substrats, M. Rumsey and F. Cardone for composition analysis, and P. Chaudhari and C. Kunz for discussions.

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Figure Captions

Fig. 1 Electron micrographs of about 50Å thick films of Au (a) and $Re_{70}W_{30}$ (b) evaporated on a 200Å thick carbon foil and overcoated with a 50Å thick carbon film. Evaporation by electron gun, temperature of substrate holder 77°K. Micrographs taken by S. Herd, IBM Research Center.

Fig. 2 Measured and calculated reflectivity of a single 400Å thick ReW film and of a 3, 5 and 7 layer coating of ReW and C. The 3 and 5 layer coatings were obtained by shadowing part of the wafer by a shutter during the evaporation. The optical constants of Re (Ref. 10) are used for the calculation, thickness of each layer starting at the Si substrate with ReW are: 56.9, 60, 49.9, 62.8, 47.3, 64.6, 49.4Å.

Fig. 3 Measured (full curve) and calculated (dashed curve) reflectivity versus glancing angle at $\lambda=1.54$Å for an 11 layer coating of ReW and C. Thickness values for the calculated curve, starting at substrate: 22, 70.4, 17.7, 69.8, 15.6, 76.1, 13.4, 87, 12.5, 80.9, 9.8Å; refractive index of ReW and C: $1-4.4\times10^{-5}-i\cdot6.8\times10^{-6}$ and $1-6.8\times10^{-6}-i\cdot0.11\times10^{-6}$. Eq. 1 with $\sigma=3$Å has been used to fit the calculated reflectivity to the measured curve. All ReW films are made thinner than in an optimized mirror in order to obtain high reflectivity in the higher order maxima.

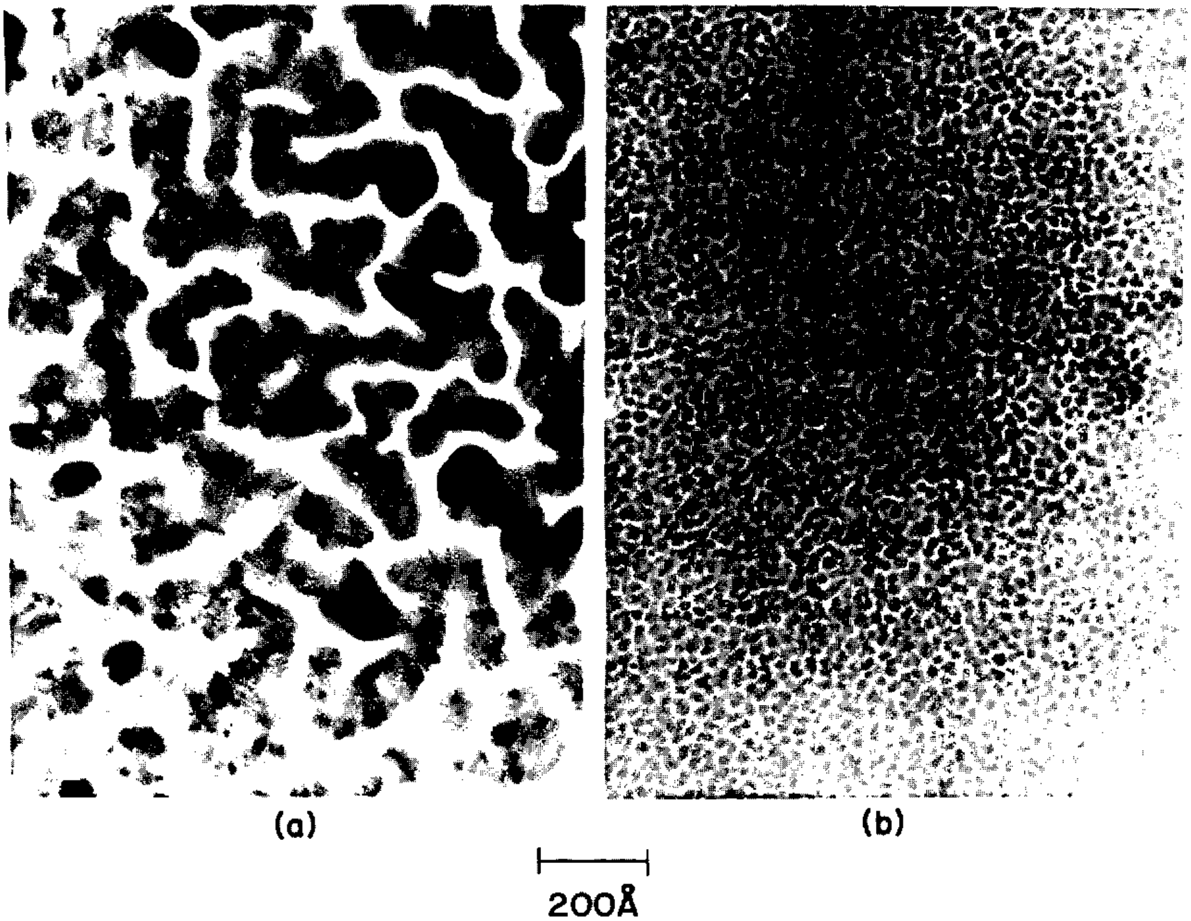
(a)
(b)
200Å

Fig. 1

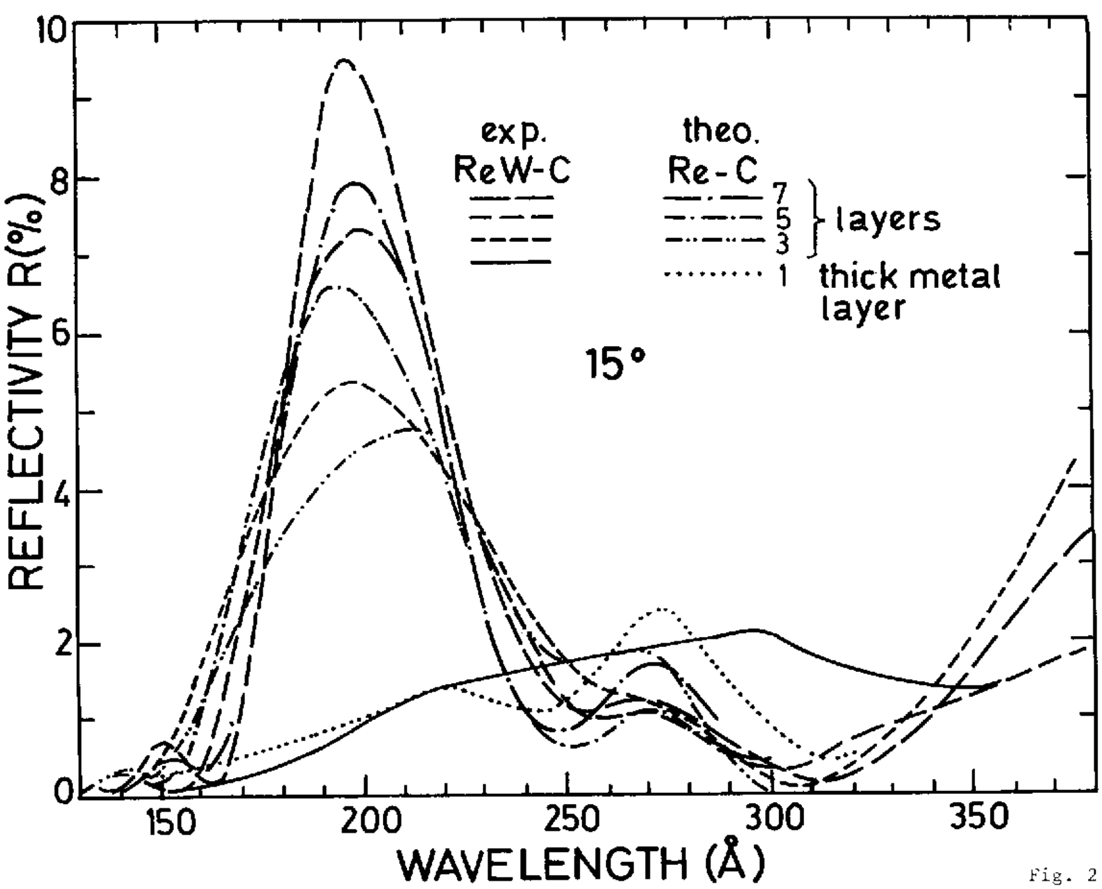
REFLECTIVITY R(%)
WAVELENGTH (Å)
exp.
ReW-C
theo.
Re-C
7
5
3
layers
1
thick metal
layer
15°
0
2
4
6
8
10
150
200
250
300
350

Fig. 2

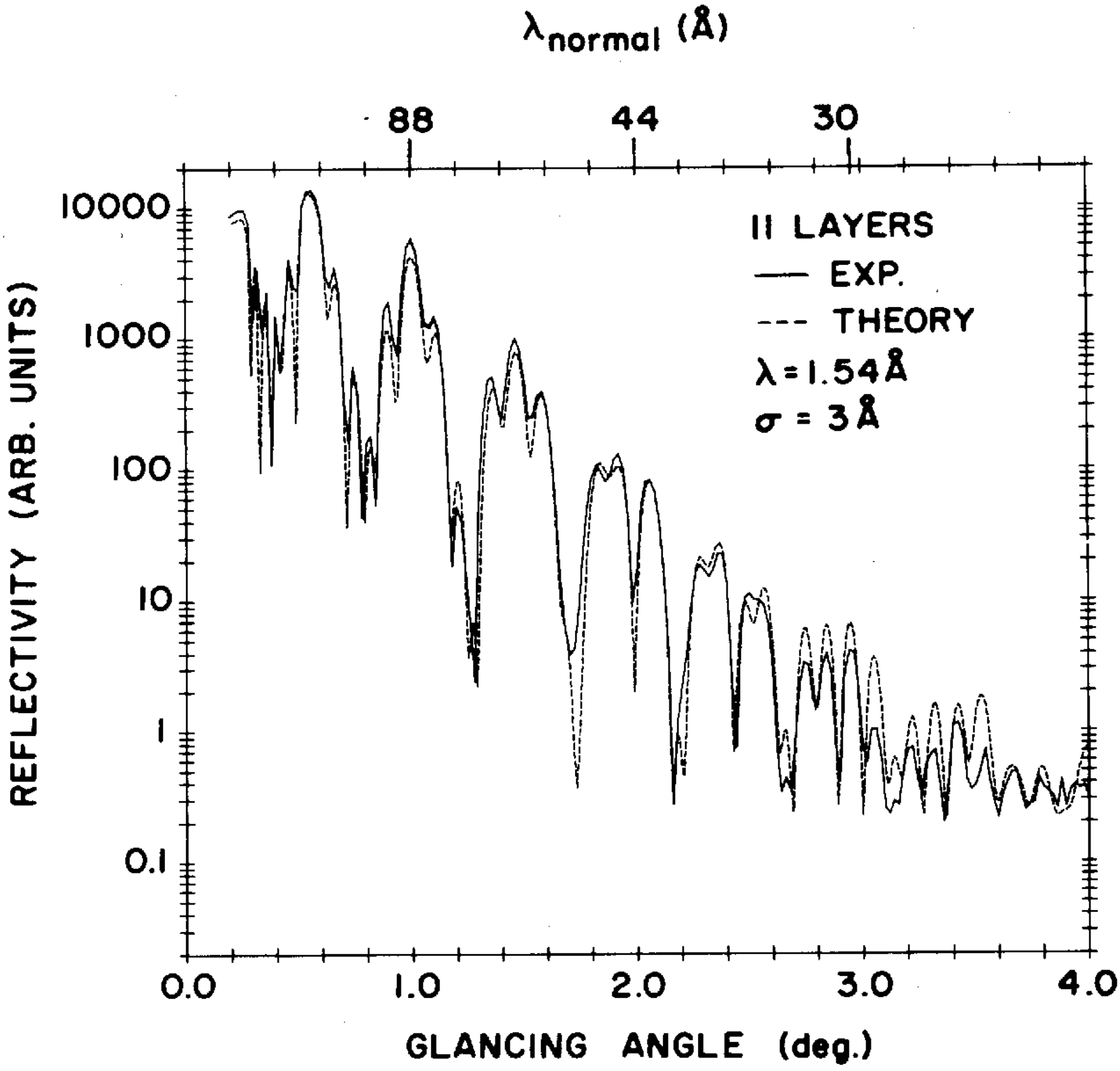
λnormal (Å)
88
44
30
10000
1000
100
10
1
0.1
REFLECTIVITY (ARB. UNITS)
11 LAYERS
EXP.
THEORY
λ=1.54Å
σ = 3Å
0.0
1.0
2.0
3.0
4.0
GLANCING ANGLE (deg.)

Fig. 3