Wideband multilayer mirrors with minimal layer thicknesses variation

I.V. Kozhevnikov,[1,*] A.E. Yakshin,[2] and F. Bijkerk[2]

[1]*Shubnikov Institute of Crystallography, Russian Academy of Sciences, Leninskiy pr. 59, Moscow 119333, Russia*
[2]*MESA Institute for Nanotechnology, University of Twente, P.O. Box 217, 7500 AE Enschede, The Netherlands*
[*]*ivk@crys.ras.ru*

Abstract: Wideband multilayers designed for various applications in hard X-ray to Extreme UV spectral regions are based on a layered system with layer thicknesses varying largely in depth. However, because the internal structure of a thin film depends on its thickness, this will result in multilayers in which material properties such as density, crystallinity, dielectric constant and effective thickness vary from layer to layer. This variation causes the fabricated multilayers to deviate from the model and negatively influences the reflectivity of the multilayers. In this work we solve this problem by developing designs of wideband multilayers with strongly reduced layer thickness variations in depth, without essential degradation of their optical characteristics.

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References and links

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1. Introduction

Multilayer mirrors (MMs) are widely used in X-ray and Extreme UV technologies because of their high reflectivity at any angle including normal incidence. However, being analogous to 1D artificial crystals and based on interference of waves reflected from different interfaces, standard periodic MMs are characterized by a narrow spectral and angular bandwidth that limits the range of their applications. However for many applications it is desirable that either the spectral or angular bandwidth is essentially increased as compared to conventional MMs. The typical examples include X-ray astronomy [1], synchrotron radiation beam steering [2], Göbel optics for increasing the flux from X-ray tubes [3], and EUV lithography [4].

A way generally used to increase the reflectivity bandwidth of MMs is a variation of the period with the depth of a multilayer structure. This way radiation of different wavelengths λ or of different angles φ incident onto the MM, according to the Bragg law, are reflected from stacks of layers placed at different depth. Several theoretical approaches were developed to design MM with any desired reflectivity profile R(λ) or R(φ) (see, e.g., [1, 2, 5–8] and references therein). Notice that all approaches result in non-monotonic oscillating variation of the layer thickness with depth, even though the desired profile of the reflectivity curve is very simple (for example, constant). Actually, as it was theoretically proved in [2], the monotone variation of the period with the depth always results in strongly oscillating reflectivity curve. The inverse statement is also valid: MM providing smooth change of the reflectivity curve is always characterized by nonmonotonic variation of the period, which often oscillates strongly with the depth.

Because the internal structure of a thin film will generally depend on its thickness, variation of the thickness can result in inaccuracies in the deposition process and errors in the layer thicknesses. Factors are a changing fraction of nano-crystalline material, changes in the materials density, a changing amount of interface material interdiffusion, or even different compound material being formed at the interfaces. Therefore, the question arises whether it is possible to strongly reduce the layer thickness variation in depth without essential degradation of its optical characteristics. This paper answers this question positively and demonstrates such designs using a Mo/Si based multilayer stack.

2. Control of layer thicknesses in wideband MMs

Several successful experiments on fabrication and characterization of wideband MMs operating in the hard X-ray to Extreme UV spectral regions were described in literature (see, e.g., [4, 9–11] and references therein). While the reflectivity bandpass typically corresponds to the desired one, the reflectivity curve is usually deformed as compared to the prescribed one. As it was demonstrated in [4, 10,11], small deviations of the layer thickness from normative values by a fraction of an Angstrom can explain deformation of the reflectivity curve observed in experiments. Nowadays the most widely used technique of hard X-ray and EUV MMs fabrication is magnetron sputtering. This is a relatively stable process where the layer thicknesses are controlled with high accuracy by deposition time. Analysis of the layer thickness fluctuations shows that the random fluctuations of the layer thickness can be better

than 10^{-4} nm [12]. However in reality the layer control method results in the control of the deposited material mass rather than the layer thickness as such. These two parameters are practically equivalent for periodic MMs. However, the situation is more complex in the case of depth-graded MMs because the internal structure of a thin film generally depends on its thickness. As an example, the density of a sputtered tungsten film was found in [13] to increase from Si substrate to the film top by 15% within the 2 nm thick layer. Furthermore, with increasing thickness an amorphous film at the initial stage of growth may transform to a polycrystalline structure. The degree of crystallinity will be generally enhanced with the film growth. While a tungsten film transforms to a polycrystalline structure at a thickness exceeding 25-30 nm [14], a molybdenum film is only amorphous up to only 2 nm thickness [15,16]. Crystallization results, first, in a film density change and second, in a modification of the structure of interlayers (thickness, density, chemical composition) forming at the boundary of neighboring materials [15,16]. This simple consideration demonstrates that the precise control of the deposited mass does not guarantee correct control of the layer thickness.

In situ X-ray reflectometry during MM deposition could be used to control layer thicknesses [17]. However first of all, the use of reflectometry is practically impossible, if the substrate moves from one magnetron to another. Furthermore, determination of the layer thickness from the temporal dependence of the reflectivity at a fixed incidence angle relies upon accurate knowledge of the internal structure of the MM. However the accurate reconstruction of the internal MM structure is based on solving the inverse problem of reflectometry [13], i.e. on measurements of the whole reflectivity curve versus the incidence angle in any point of deposition time. This is only possible with the use of high intensity synchrotron radiation.

A more practical way to solve the problem would be to design a wideband MM with the variation in layer thicknesses as small as possible. Then it can be expected that the internal structure of the layers will only differ insignificantly. As a result, the layer thicknesses will deviate less from the design values, even when existing methods of the deposited mass control are used.

3. Design of broad band MM

For our study we selected the Mo/Si based MM system since it is probably the most frequently studied MM system. This is due to its importance as a high reflectance coating for EUV lithography and astronomy. We will perform analysis for a MM consisting of 50.5 periods with the following composition: $SiO_2/[Si/B_4C/Mo/B_4C]_{50}/Si/substrate(SiO_2)$, with the B_4C barrier layer of a fixed 0.3 nm thickness as used between the Mo and Si layers to prevent interdiffusion [18–21]. The native SiO_2 oxide on the MM top is assumed to be 1.5 nm thick. The material densities are supposed to be equal to the bulk ones. Interfacial roughness effect is neglected.

The task is formulated as follows. It is necessary to design a MM providing a constant reflectivity R_0 = 50% at a wavelength λ = 13.5 nm in the [0, 18°] interval of an incidence angle φ (the case of naturally polarized radiation). The range [0, 18°] is taken as an example from [4]. Optimization parameters are Mo and Si layer thicknesses with the imposed limitations on their possible values. An empirically found minimum Mo thickness d_{min} = 2.2 nm is taken to provide that all Mo layers have the same crystallographic structure. It is known that an amorphous to crystalline transition occurs in Mo layers for a thickness around 2nm [15,16]. A maximum Mo thickness of d_{max} = 6 nm is used since the Mo layer can develop roughness with thickness.

The MM optical quality is characterized by the mean value <R>, the relative dispersion RD, and the peak-to-valley value (PVV) of the plateau reflectivity according to the following expressions:

$$< R >= \frac{1}{\varphi_{max} - \varphi_{min}} \int_{\varphi_{min}}^{\varphi_{max}} R(\varphi)\, d\varphi \qquad (1)$$

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$$\mathrm{RD}=\left[\frac{1}{\varphi_{\max}-\varphi_{\min}}\int_{\varphi_{\min}}^{\varphi_{\max}}\left(\frac{<R>-R(\varphi)}{<R>}\right)^{2}d\varphi\right]^{1/2} \quad (2)$$

$$\mathrm{PVV}=\max R(\varphi)-\min R(\varphi)\,,\quad \varphi\in[\varphi_{\min},\varphi_{\max}] \quad (3)$$

where $\varphi_{min} = 0$ and $\varphi_{max} = 18°$. The values of RD and PVV are closely related with each other and characterize the deformation of the reflectivity plateau. Optical characteristics of the MMs designed are listed in Table 1.

Table 1. Optical parameters of MMs designed to provide the constant reflectivity of naturally polarized radiation at λ = 13.5 nm in the [0, 18°] range of the incident angle.

MM	R_0, %	Q	<R>, %	RD, %	PVV, %
A1	50	0	50.0	0.16	0.27
A2	50	0.01	50.0	0.28	0.55
A3	50	1	50.0	0.55	1.10
A4	50	20	49.7	2.76	7.6
B1 = A1	50	0	50.0	0.16	0.27
B2	51	0	51.0	0.17	0.38
B3	52	0	52.0	0.33	0.77
B4	53	0	53.0	0.40	0.83
B5	54	0	54.0	0.56	1.27
B6	55	0	54.9	0.69	1.54
C1 = A3	50	1	50.0	0.55	1.10
C2	51	1	51.0	0.64	1.39
C3	52	1	52.0	0.78	1.64
C4	53	1	52.9	0.85	1.70
C5	54	1	53.8	1.03	2.04
C6	55	1	54.7	1.32	2.46

First of all, we design a MM with the desired optical parameters using the conventional merit function as in our previous paper [4]:

$$\mathrm{MF}_1=\sum_{m=1}^{M}\left[R_0-R(\varphi_m)\right]^2,\quad \varphi_m\in[0,18^{\circ}] \quad (4)$$

where M is typically equal to the number of optimization parameters, i.e. 101 in our analysis. The minimization of the merit function is performed with the use of the Levenberg-Marquardt algorithm [22]. As a starting guess we choose the simplest distribution of the bi-layers thickness in the form $d_j = a + b / j$, where j is the number of bi-layer counted from the top, and the thickness ratio, or metal fraction, is put at a constant $d_{Mo} / (d_{Mo} + d_{Si}) = 0.4$ for all bi-layers. More accurate choice of the starting guess, as, e.g., in [2], proves to be unnecessary, because the reflectivity bandwidth is only twice of that of a periodic MM.

The multilayer structure obtained as a result of the merit function minimization is denoted as A1. The depth-distribution of the layer thickness and the reflectivity versus the incidence angle are shown in Fig. 1. The reflectivity plateau is flat, with the reflectivity variation PVV being only 0.27%. At the same time, the thickness distribution is the complex and quickly oscillating function of the depth, the Mo layer thickness being changed in the maximum allowed interval from d_{min} = 2.2 nm to d_{max} = 6 nm. As we discussed above, such a layer thickness distribution is quite typical for wideband EUV mirrors.

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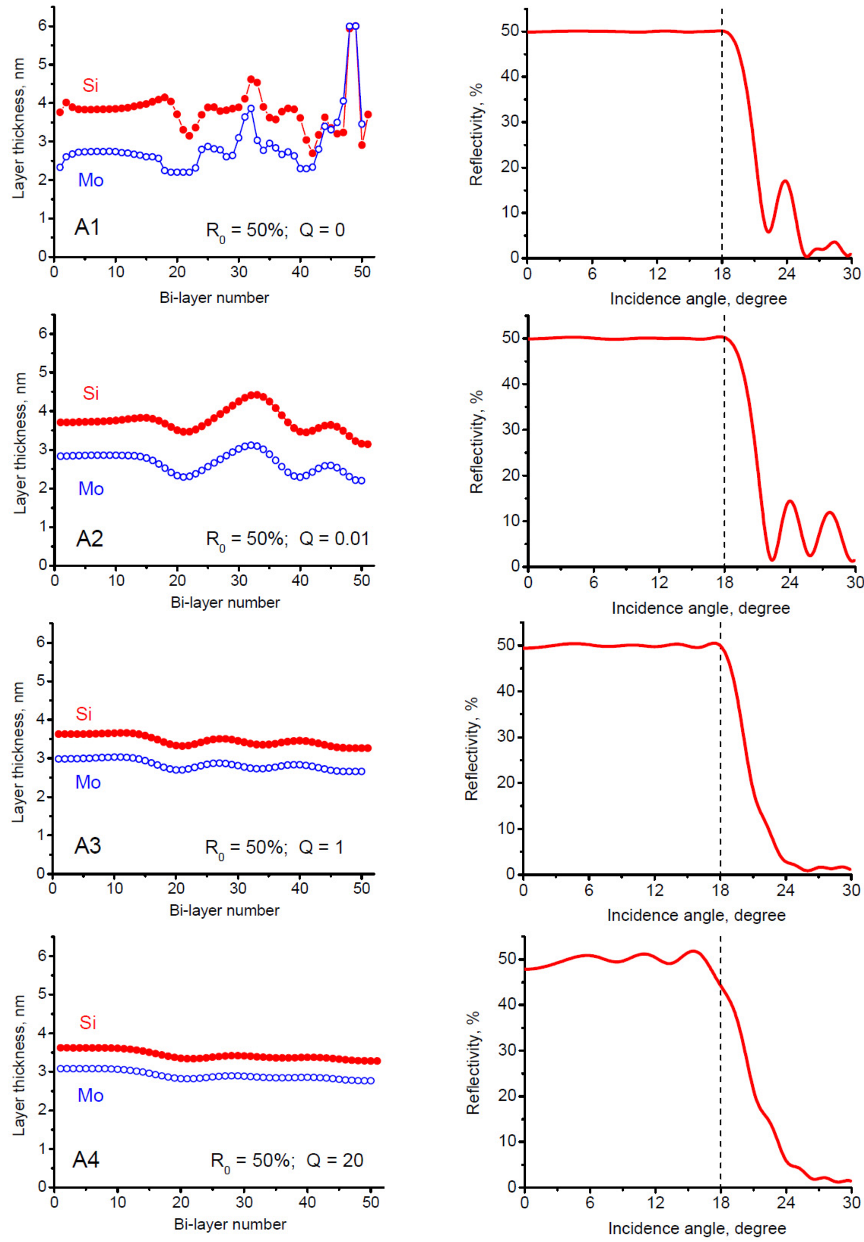

Fig. 1. Depth-distribution of the layer thicknesses (left column) and corresponding reflectivity versus the incidence angle (right column) for MMs A1 – A4 designed to provide a constant reflectivity in the [0, 18°] interval of the incident angle at 13.5 nm wavelength. Naturally polarized radiation is considered. Bi-layer number is counted from the top.

To design MMs with smaller variation in the layer thickness we introduce a new merit function

$$\mathrm{MF}_2 = \int_{\varphi_{\min}}^{\varphi_{\max}} [R_0 - R(\varphi)]^2 \, d\varphi + \frac{Q}{(\Delta d)^2} \sum_j \left[(d_{2j+1} - d_{2j-1})^2 + (d_{2j+2} - d_{2j})^2 \right] \qquad (5)$$

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where Q is additional minimization parameter that controls a summation over all the bi-layers of MM. The purpose of the sum is to minimize the difference in thicknesses of all odd (Si) and even (Mo) layers. The constant $\Delta d = d_{max} - d_{min} = 3.8$ nm is introduced in order to keep the parameter Q dimensionless. If Q = 0, we obtain a conventional merit function (4). If Q tends to infinity, minimization of (5) will result in a periodic MM.

Putting Q = 0.01 in (5), we obtain the MM A2 in Fig. 1 with a smoother layer thickness distribution than in A1 while with still having a quite flat reflectivity profile (PVV = 0.55%). This shows that it is in principle possible to obtain solutions with a smoother layer distribution without sacrificing reflectivity properties of the wideband mirror. However a really interesting solution was obtained when increasing the parameter Q even further. Putting Q = 1 we obtained the MM A3 in Fig. 1. The MM A3 contains significantly smoother layers thickness distribution than in the previous solutions and still a practically flat reflectivity plateau (PVV = 1.1%). The variation in the layer thicknesses over the whole depth of the MM A3 does not exceed 0.39 nm, 10 times smaller compared to the MM A1, with the period variation being less than 0.76 nm. If we write a Bragg condition of reflection for two periodic structures, at $\varphi = 0$ and $\varphi = 18°$ incidence angle (the minimum and maximum working angle of our wideband mirror), we will find that the period of the two periodic structures would differ by about 0.35 nm. This means that the MM A3 is close to the best possible solution that can solve the problem formulated above. Indeed, increasing the parameter Q further results in already a very small difference in the layer thickness variation, while significantly increasing deformation of the reflectivity plateau. An example is the MM A4 designed at Q = 20 and resulting in PVV = 7.6%.

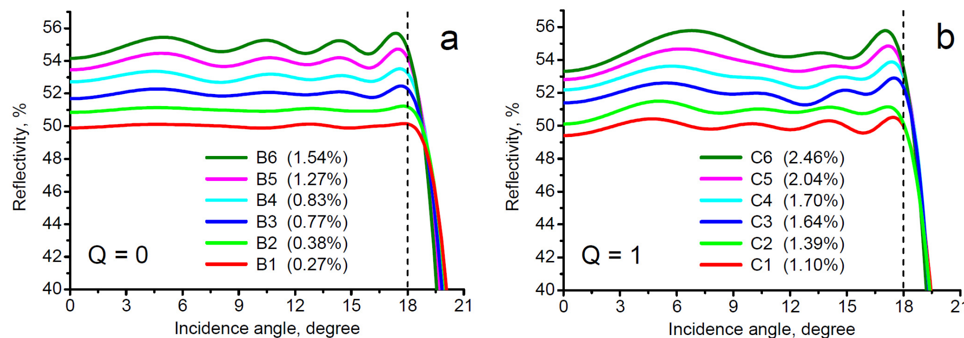

Fig. 2. Reflectivity plateau for MMs of B-type (a) and C-type (b) designed to provide a constant reflectivity in the [0, 18°] interval of the incident angle at 13.5 nm wavelength. The desired value of the reflectivity is increased from 50% for MMs B1 and C1 up to 55% for MMs B6 and C6 with a 1% step. The maximal variation of the plateau reflectivity (PVV) is shown in brackets. Naturally polarized radiation is considered.

The plateau reflectivity $R_0 = 50\%$ used for designing all of the previously described MMs is still far from the maximum possible. To illustrate the improvement possible we considered a set of MMs denoted as B in the Table 1 and designed these with the use of the conventional merit function (4) to obtain an increasingly higher reflectivity plateau in the same [0, 18°] incidence angle interval. The reflectivity plateaus are shown in Fig. 2a, with the desired values of R_0 being increased from 50% for the MM B1 up to 55% for the MM B6. As a result the maximal variation of the plateau reflectivity PVV increased from 0.27% for B1 to 1.54% for B6. Moreover, the mean reflectivity $\langle R \rangle$ of B6 was observed to start to deviate from the desired value of R_0. Evidently, MMs designed to obtain higher R_0 would show sharply increased PVV and deviation in $\langle R \rangle$ [4]. Therefore, they are not considered here. The layer thickness distributions for the MMs of the B-type shown in Fig. 3, left column, represent, as usually, quickly varying functions with high oscillation amplitude.

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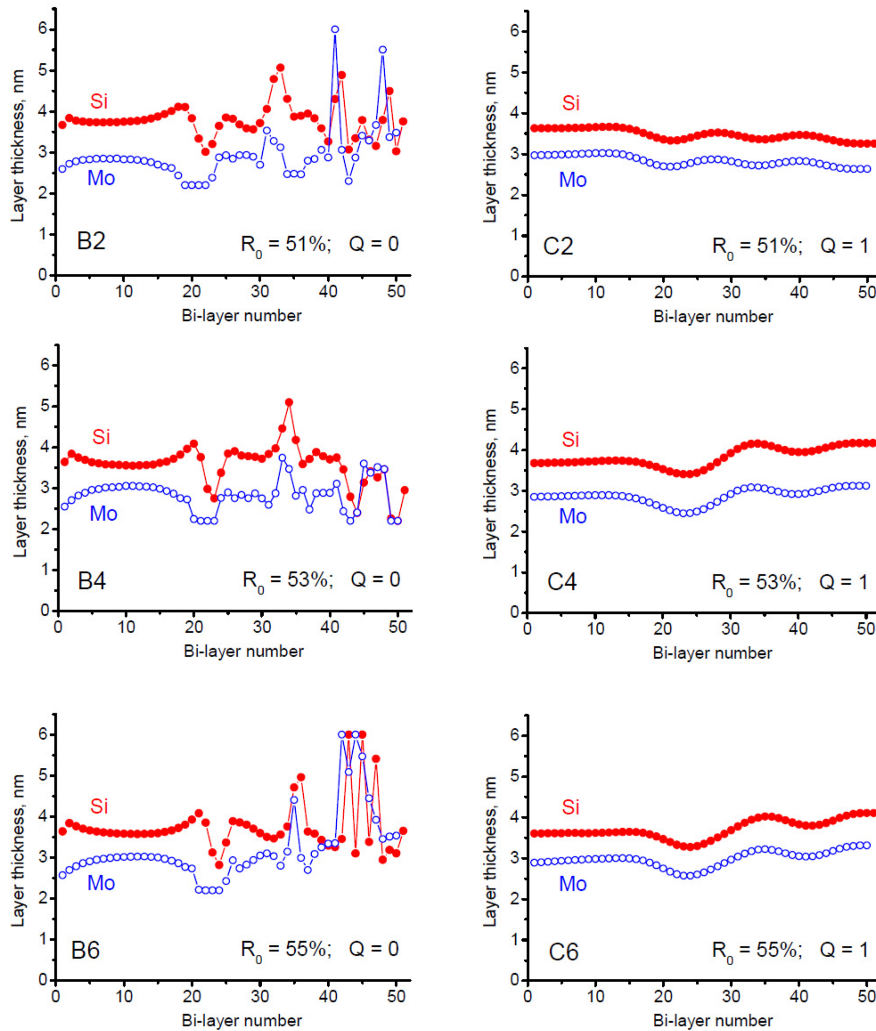

Fig. 3. Comparison of the depth-distribution of the layer thicknesses for MMs of B and C-type designed to provide a constant reflectivity in the [0, 18°] interval of the incidence angle at 13.5 nm wavelength with the use of the merit function (4) (MMs of B-type) or (5) at Q = 1 (MMs of C-type). Naturally polarized radiation is considered. Bi-layer number is counted from the top.

Further, we designed MMs of a C-type with the same desired optical parameters as for the MMs of B-type, but with lesser variation of layer thicknesses with depth. As for MMs of an A-type, we used the merit function (5) and put, for definiteness, the parameter Q = 1. Notice that the MM C1 coincides with the MM A3 considered above. The obtained depth-distributions of the layer thicknesses are shown in Fig. 3, right column, and the corresponding reflectivity plateaus in Fig. 2b. We observe that although the maximal variation of the reflectivity PVV is slightly increased, the layer thickness distributions are considerably smoother as compared to the MMs of the B-type even with the ultimately high reflectivity R_0. It is interesting that the thickness distributions for the MMs C1 and C2 as well as for the MMs C3 – C6 are very similar. The periods of C1 – C2 are slightly decreasing, on average, with depth. In contrast the periods of C3 – C6 are slightly increasing with depth.

Finally, choosing a compromise between the desired value of the reflectivity plateau R_0 and its maximum variation PVV, and using the merit function (5), it is possible to design wideband MMs with very smooth depth-distribution of the layers thickness. Furthermore the

maximal variation of the thickness can be several times smaller as compared to the MMs designed with the use of the conventional merit function (4). The phase of the reflectivity amplitude in all designs is a smooth monotonous function of the incident angle on the reflectivity plateau. It is varied within about 2 rad range for all MMs considered.

4. Conclusion

We have developed designs of wideband-reflecting multilayer XUV mirrors with strongly reduced in-depth layer thickness variations, without essentially degrading their optical characteristics. Using such designs on multilayer systems in which in practice the structure of the film depends on the individual layer thickness, should lead to a reduced complexity of the design procedure, a reduced inaccuracy in the deposition process and a better agreement of the deposited structures with the model.

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