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Effect of roughness, deterministic and random errors in film thickness on the reflecting properties of aperiodic mirrors for the EUV range

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Abstract. **By the example of three aperiodic multilayer Mo/Si mirrors (AMM) for the wavelength ranges 17–21 nm, 24–29 nm, and 28–33 nm we have studied numerically the effect of the linearly determinisctic and random fluctuations of the film thickness and the interlayer roughness on the spectral dependences of the reflection coefficient. The simulation results are used to solve the inverse problem of reconstructing the interlayer roughness and the thickness of individual films from the measured dependences of the extreme UV radiation reflection coefficients. It is shown that the 'asymmetry' of the boundaries affects the magnitude and slope of the reflection coefficient plateau. Random fluctuations of the film thickness with the variance of 1 %–2 % weakly influence the reflection characteristics of AMMs and allow reliable reconstruction of the thickness of individual films. The fluctuations with the variance 8 %–10 % allow the estimation of individual thicknesses, but the reflection curve in this case strongly differs from the desirable one. Larger fluctuations do not allow the reconstruction of the AMM structure. The basic criteria for high-quality AMM synthesis are formulated.**

Keywords: *aperiodic multilayer mirrors, extreme ultraviolet radiation, thin films.*

1. Introduction

Aperiodic multilayer mirrors (AMMs) with the spectral reflection band (angular, at a fixed wavelength) broadened as compared to that of the conventional periodic structures are widely used in scientific research. They are applied in astronomy to increase the integral reflection coefficient in grazing incidence telescopes, operating in the hard X-ray range [1], and in the extreme UV (EUV) range as components of 'stigmatic' spectrometers with diffraction gratings for solar studies [2]. The application of AMMs in X-ray microscopy in the water transparency window (the wavelengths 2.3–4 nm) increases the recorded signal by two–three times [3], which is of particular importance when operating with the laboratory radiation sources. The instrument comprises at least two multilayer mirrors for the projection objective and the collector mirror illuminating the studied sample. For the latter it is also preferable to use a multilayer mirror from the point of view of the signal magnitude [4]. In the case of periodic multilayer mirrors having the reflection spectral band in this range $\Delta\lambda/\lambda = 1/200-1/300$, the efficient operation of the scheme requires the distribution of periods over the surface of each mirror to be performed with better accuracy (~0.1 %), and the Bragg condition should be fulfilled for each ray passing through the system of mirrors. In this sense, the mirrors should be 'identical' with the accuracy of about 0.1 %. In practice, it is almost impossible. The use of AMMs essentially reduces the requirements to the accuracy of film fabrication, which makes it real to develop high-resolution and efficient microscopy based on normal-incidence multilayer mirrors.

Among multiple applications of AMMs one should note their use in one- and two-dimensional focusing of hard X-ray radiation in the Kirkpatrick–Baez system [5] and in controlling the spatial, temporal and spectral characteristics of femto- and sub-femtosecond pulses of electromagnetic radiation in the EUV region [6]. In recent time, in relation with the studies of the extreme states of matter, the field of interest has moved towards atto- and sub-attosecond pulses with the spectrum in the X-ray or EUV range [7].

Practically, to fabricate a particular optimal AMM three main problems are to be solved. The first problem is to calculate the optimal AMM structure (thickness of layers) using the objective function of the spectral (angular) dependence of the reflection coefficient (direct problem). The second problem is to reconstruct the 'true' thickness of layers in the synthesised AMM using the X-ray reflectometry (inverse problem) for correcting the technological process. The third problem is to characterise the fabricated mirror.

At present different calculation algorithms exist for the solution of the direct problem, from the probabilistic Monte Carlo methods to the solution of the global minimisation problem, which proved their capability in different ways, including the experimental ones [8–11]. However, even for this problem there are issues still to be studied. First, the presence of film defects (the difference of material densities from their bulk values, the roughness or gradient interlayer interfaces related to the diffusion and chemical interaction of materials) not only reduces the actual reflection coefficient, but also essentially changes the shape of the spectral reflection curve. Moreover, if these factors are taken into account at the very beginning of the mirror optimisation process, then the resulting layer thicknesses in the AMM may appear quite different as compared to the case, when the thicknesses are optimised for an 'ideal' AMM. In particular, this effect has been observed in the region of photon energies 10 keV [12]. In the EUV range due to the strong absorption, one can expect

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even a greater effect of the film defects on the optimisation of the film thicknesses in AMMs.

We could not find in the literature any examples of solving the inverse problem of determining the real film thicknesses in AMMs from the reflection data in the X-ray range. Note only, that as compared to the inverse problem for periodic mirrors, where the parameters are the film material density, the periodic film thickness, and the form of the interlayer functions (actually, the permittivity profile within a single period of the multilayer mirror) [13, 14], in the AMM the number of parameters to be determined increases with increasing number of layers in the multilayer structure.

To specify the approaches and to clarify the problems of solving the inverse problem for AMMs, in the present paper we numerically study the effect of linearly deterministic and random fluctuations of film thicknesses and interlayer roughness on the spectral dependences of reflection coefficients in the Mo/Si AMM, intended for the Cortes spectroheliograph [15]. Based on the result of the study, we draw conclusions about the influence of roughness on the reflection coefficients and the necessity to take it into account in the calculation of optimal thicknesses of the AMM films, as well as on the possibility to separate the effect of random and deterministic variations of the film thickness on the reflection coefficients of the AMM by the character of their influence on the shape and spectral shift of the reflection curve. It is shown that this approach allows the optimisation of a particular layer thickness in the AMM and then the reconstruction of the film thicknesses in real AMMs at small (maximal deviation from the nominal value no greater than 10%) thickness fluctuations. For larger fluctuations, the problem has no satisfactory solution. The results of these studies will be taken into account in the AMM synthesis for practical applications.

2. Direct problem. The calculation method

We numerically studied three Mo/Si AMMs having spectrally uniform reflection coefficients with a maximal possible magnitude in the wavelength ranges 17–21 nm, 24–29 nm, and 28–33 nm. The first two structures were also studied experimentally. The calculation of the layer thicknesses was carried out for ideal structures (with a tabulated material density and zero interlayer roughness). The minimal layer thickness was chosen to be 2 nm, and the number of layers did not exceed 40. The acceptable variations of the reflection coefficients within the spectral dependence plateau of the reflection coefficient amounted to 2% for the range 17–21 nm, 0.5% for the range 24–29 nm, and 0.5% for the range 28–33 nm.

The problem of determining the thicknesses of AMM layers consists in the minimisation of the objective function, which in our case had the form

$$F(\{d\},\theta_0) = \int \{-w_1 R_i(d,\lambda) + w_2[(R_0 - R_i(d,\lambda)]^2\} \mathrm{d}\lambda,$$

where $\{d\} = \{d_1, d_2, \ldots, d_N\}$ are the thicknesses of the layers; N is the number of layers; θ_0 is the grazing reflection angle of the X-ray radiation; $R_0(\lambda)$ is the desired profile of the reflection coefficient; $R_i(\lambda)$ is the current calculated profile of the reflection coefficient, varying in the process of the solution; and $w_{1,2}$ determine the weight of integral reflection and root-mean-square deviation of the real reflection coefficient from the desired one, respectively. The first term provides the maximal integral reflection, and the second one provides the minimal deviation from the specified profile. For the direct problem, the profile R_0 has a plateau with a certain given constant value within a definite wavelength interval. For the inverse problem, it is the reflection profile measured for real samples.

The reflection coefficients of multilayer mirrors were calculated using the classical Parratt iteration formulae [16]. The interlayer roughness was taken into account by multiplying the appropriate complex Fresnel reflection coefficients by the Debye–Waller factors at each interlayer boundary (this operation is valid in the case of correlated roughness [17]). By the interlayer roughness we mean both the geometric roughness as such and the width of the interlayer transition zones that arise due to the diffusion and chemical interaction between the materials of adjacent layers.

The objective function extremum was found using the alternating-variable descent method, in which the objective function was minimised sequentially for each layer. The formula for the dependence of the reflection coefficient R_i on the thickness of any layer was expressed in the explicit form [3], which in the case of a one-time use of the algorithm requires only a single run of the recurrent procedure, thus reducing the total calculation time.

The numerical modelling was performed for the effect of both the random error in the thickness of the structure layers and their deterministic deviations. The random errors were simulated by the generator of random numbers with the normal distribution, zero mean value and unit variance [18]. The effect of deterministic variation of the layer thickness was studied for the deviation Δd within ± 0.15 nm.

The AMM optimisation programme that solved the problem of obtaining the maximal uniform integral reflection coefficient within the specified wavelength band was implemented in the Fortran language. We elaborated a special graphic interface AMuLet (aperiodic multilayer engineering tools) that significantly simplified the work with the programme code. In the algorithm of the AMuLet programme, the possibility of solving the inverse problem in the presence of interlayer roughness is available from the very beginning of the iteration process.

2.1. Effect of interlayer roughness on the reflection coefficient

Figure 1 presents the calculated spectral dependences for the reflection coefficients of three AMMs, optimised for different spectral regions and a few values of the interlayer roughness. With the growth of roughness the reflection coefficient of mirrors predictably decreases, and the initial degree of uniformity in the plateau region is integrally preserved. However, for further AMM diagnostics the fact of interest is that for different roughness magnitude of different boundaries (multilayer structure with ‘asymmetric’ roughness) the spectral plateau possesses a slope with the sign depending on which of the boundaries has greater roughness. The effect is clear in the case of strong inequality of roughness, namely, when the roughness of the Mo on Si boundary is much greater than that of the Si on Mo boundary, which is actually observed in the experiments. The plateau slope grows with increasing wavelength. Thus, the reflection coefficient, smaller than calculated theoretically, and the presence of an apparent slope of the plateau of the spectral dependence are indirect signs of the asymmetry of interlayer boundaries that can be used to optimise the algorithm of the inverse problem solution.

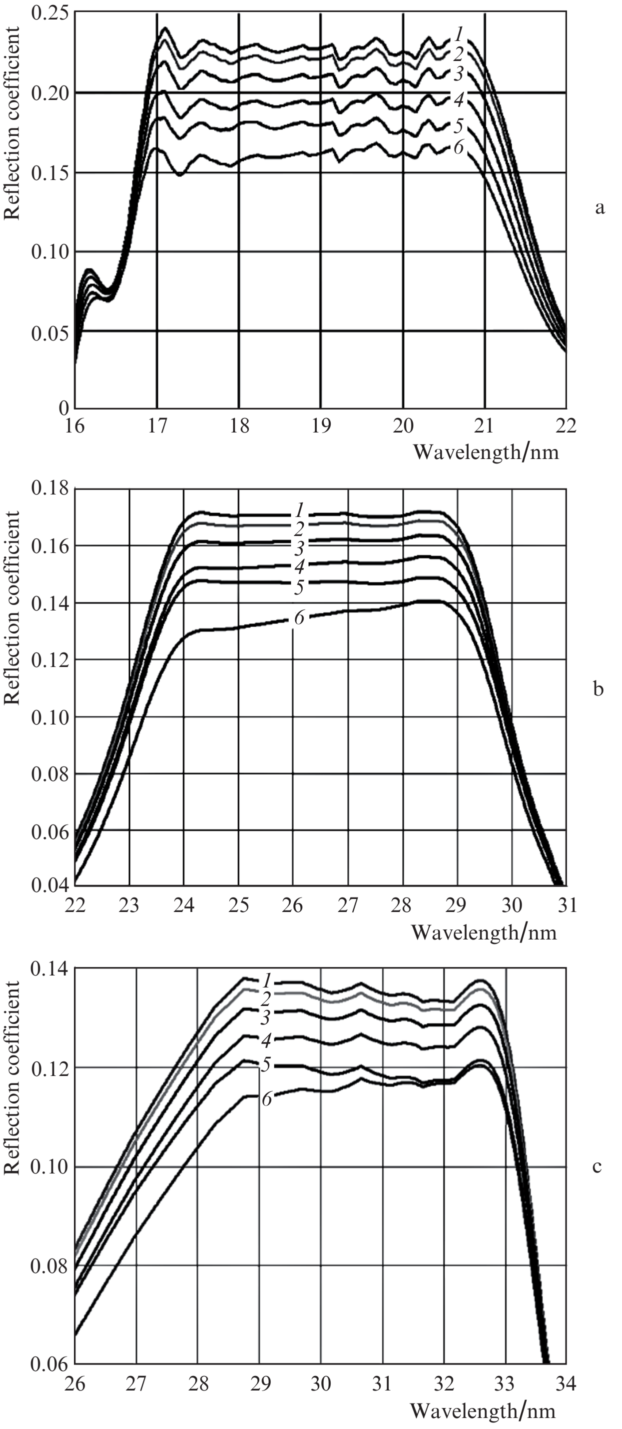

Figure 1. Calculated spectral dependences of Mo/Si AMMs refection coefficients in the ranges (a) 17–21 nm, (b) 24–29 nm, and (c) 28–33 nm for different values of roughness at the Mo on Si and Si on Mo boundaries: (*1*) 0 and 0, (*2*) 0.3 nm and 0.3 nm, (*3*) 0.5 nm and 0.5 nm, (*4*) 0.7 nm and 0.7 nm, (*5*) 1.2 nm and 0.6 nm, and (*6*) 0.6 nm and 1.2 nm.

Earlier [12] by the example of the AMM with the reflection coefficient at the wavelength 0.154 nm it was shown that by taking the interlayer roughness into account at the very beginning of the optimisation algorithm execution, one can achieve considerable improvement of the reflection characteristics as compared to the AMM, in which the interlayer boundaries were initially considered ideal, and the roughness was taken into account only after the calculation was carried out. To study this effect in the EUV range, we optimised Mo/Si AMMs for the following values of roughness: 1.2 nm for the Mo on Si boundary and 0.6 nm for the Si on Mo boundary. The chosen roughness parameters were determined using the results of the study of periodical multilayer Mo/Si structures with the periods about 7 nm. The present values of roughness were used in the initial approximation in the solution of the problem of optimising the mirror parameters. The values of thin film densities were also found experimentally from the data of studying the corresponding periodic structure. In the calculations, we used the density of molybdenum equal to 0.95 of the tabulated value. The density of silicon layers was assumed to have the tabulated value.

Note that the *a priori* allowance for the presence of interlayer roughness did not lead to a large increase in the mirror reflectivity, which can be seen, e.g., in Figs 2a and 2b. The only positive effect, obtained due to the *a priori* account for the roughness asymmetry, was the reduction of the plateau slope. Figure 2b presents an example of this improvement.

Thus, the performed numerical study allows a number of conclusions, important for optimising the composition of the AMM films, as well as for reconstructing the layer parameters from the data of reflectometry measurements. First, the roughness most strongly affects the magnitude of the reflection coefficient at the plateau. Here it is important to take the possible asymmetry of interlayer boundaries into account. Second, the roughness does not lead to additional oscillations of the reflection coefficient at the plateau, but is able to affect its slope, particularly in the long-wavelength region. Third, when designing an AMM for a particular application in the EUV range, it is necessary (or at least extremely desirable) to know the values of the interlayer roughness and, in principle, the real densities of the layer material, which, in general, depend on the layer thickness. These values can be obtained from the study of the appropriate periodic multilayer mirrors with a close thickness of layers. All these data should be taken into account either from the very beginning of the optimisation algorithm execution, or in the repeated solution of the optimisation problem. Such an approach allows the preservation of the reflection coefficient uniformity in the desired range without the loss of integral reflection.

2.2. Influence of deterministic and random variations of film thickness on the AMM reflection coefficient

One more factor affecting the shape of spectral dependence of the reflection coefficient and its magnitude are the errors in the thickness of the AMM films. The errors can be caused by both random and systematic processes. For example, in the case of magnetron deposition the random errors are caused by the fluctuations of voltage at the magnetrons and the working gas pressure, as well as by microbreakdowns. Systematic errors are associated with the permanent consumption of the target erosion zone in the process of synthesis and poorly determined correspondence between the velocity of passing the sample above the target and the rate of film deposition. This problem is particularly urgent in the synthesis of AMMs, since for periodic mirrors the passing velocity varies within wide limits depending on the thickness of the layers. The analysis of the effect of deterministic and random variations of the film thicknesses in AMMs is important for two reasons. First, it allows one to estimate their influence on the reflection spectral curves and thus to elaborate the criteria for admissible errors that obviously depend on each particu-

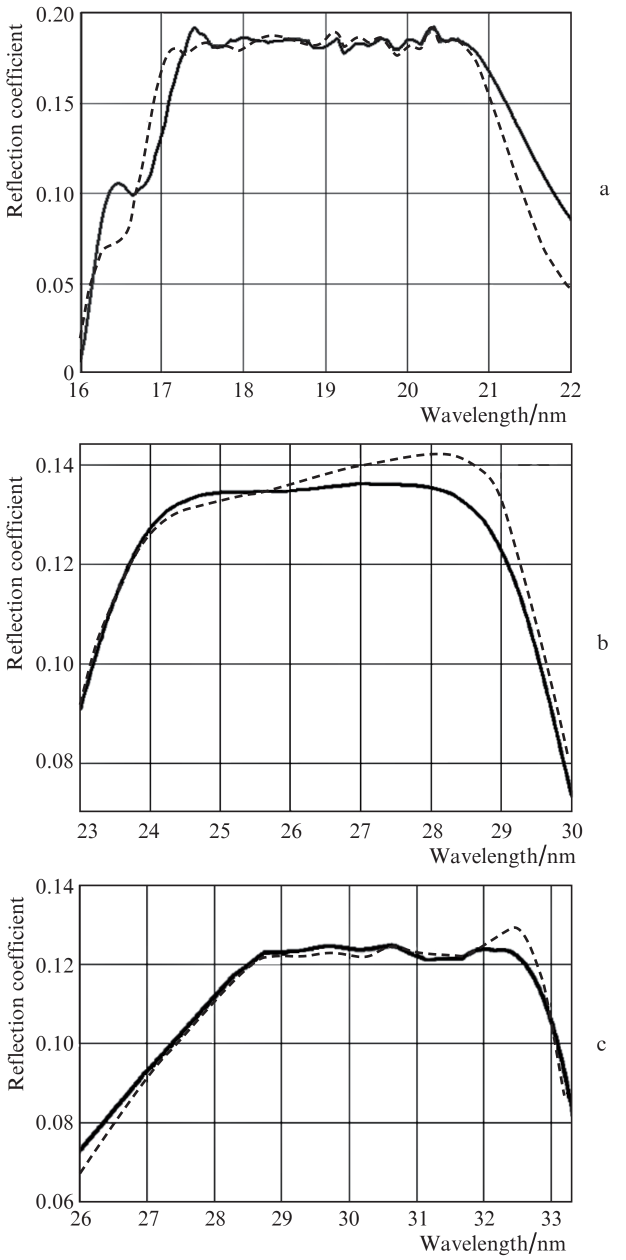

Figure 2. Calculated spectral dependences of Mo/Si AMMs refection coefficients with the roughness 1.2 nm for Mo on Si and 0.6 nm for Si on Mo in the ranges (a) 17–21 nm, (b) 24–29 nm, and (c) 28–33 nm. The dashed curves correspond to the optimisation of the AMM film thicknesses followed by the allowance for the roughness at the boundaries. The solid curves correspond to the optimisation with the roughness taken into account from the very beginning of the calculation.

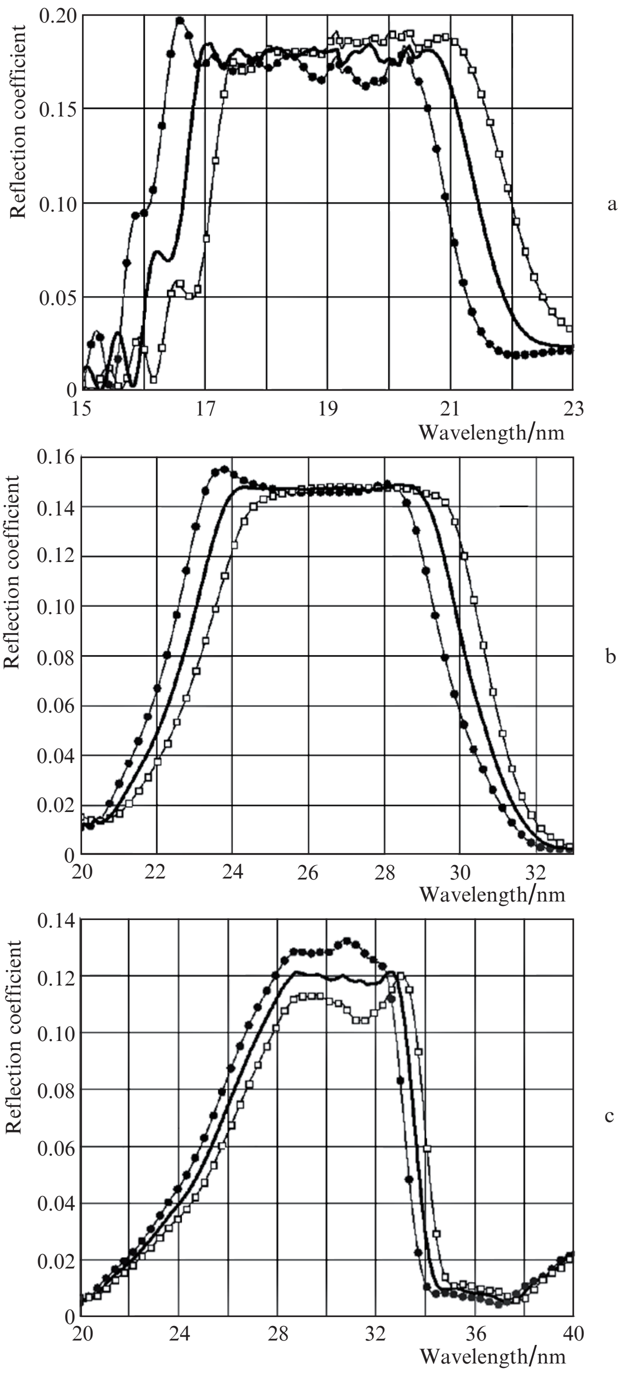

Figure 3. Spectral dependences of the reflection coefficients for three AMMs with the calculated (solid curves) and deterministic variations of thicknesses in each period Δd = (●) −0.15 and (□) 0.15 nm.

lar problem. Second, this analysis helps one to solve the inverse problem of reconstructing real AMM parameters from the measured reflection coefficients.

Figure 3 presents the spectral dependences of the reflection coefficients for three AMMs with the calculated film thicknesses and deterministic variation of the thickness in each period by the same value within ±0.15 nm that corresponds to the period change by nearly 1 %. It is seen that these changes mostly affect the position of the reflection coefficient plateau, shifting it to the long wavelength or short wavelength region, depending on the sign of Δd. The height and spectral width of the reflection coefficient plateau, as well as its oscillations, change insignificantly. For the third AMM a greater decrease in the reflection coefficient is observed. One can also see that the greater the period change, the stronger the shift and the deformation of the initial profile.

The influence of random fluctuations of the films thickness with different variances (0.3 %, 0.5 %, 1 %, 2 %, and 5 %) on the reflection coefficient in the middle of each spectral range for three AMMs is illustrated in Fig. 4, presenting the envelopes of the histograms of the reflection coefficient distributions in the middle of each spectral range for 3000 realisations. The analysis of the presented curves, as well as thousands of random realisations, shows that these errors weakly affect the width and position of the plateau of the reflection coefficient, slightly changing only the shape. The fluctuation most strongly affects the oscillations and the absolute value of the reflection coefficient. In particular, in the case of fluctuations of the layer thickness with the variance 1%, the half-width of the reflection coefficient distribution for the first AMM amounts to 0.71%, for the second one – to 0.32%, and for the third one – to 0.66%. For the fluctuations with the variance 5%, the half-width of the reflection coefficient distribution exceeds 25% of the nominal value and it is practically unreal to obtain the desirable uniform reflection coefficient. The effect increases under the shift towards the long wavelength region. From the presented data, one can conclude that in spite of the wide reflection bandwidth of the AMM (in the present case $\Delta\lambda/\lambda \approx 25\%$), the variance of admissible fluctuations lies within the range of 1%–2%. This condition is essentially harder than in the case of multilayer periodic mirrors, where the admissible fluctuations can be comparable with the relative transmission bandwidth of the mirrors.

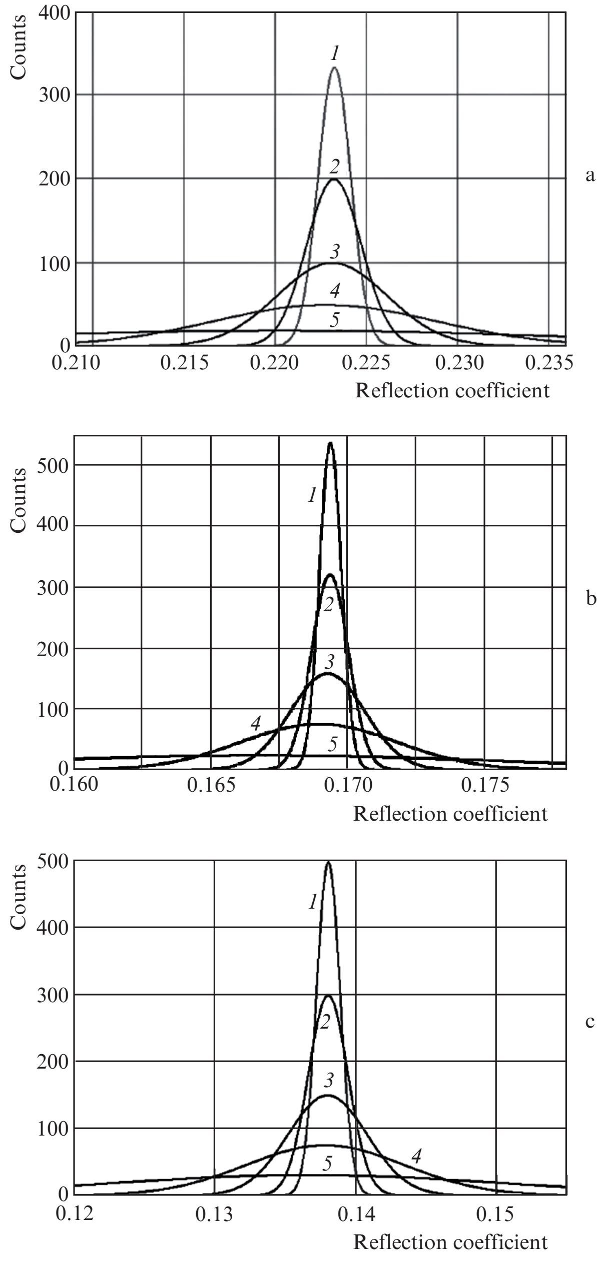

Figure 4. Histogram envelopes for the distribution of AMM reflection coefficients, calculated for the variances of film thickness fluctuations (*1*) 0.3%, (*2*) 0.5%, (*3*) 1%, (*4*) 2%, and (*5*) 5% in the ranges (a) 17–21 nm, (b) 24–29 nm, and (c) 28–33 nm.

3. Experiment. Inverse problem

The samples were synthesised at the Institute for Physics of Microstructures, Russian Academy of Sciences, using the magnetron deposition method in the argon atmosphere at the pressure 8×10^{-4} Torr. The films were deposited onto silicon substrates with the root-mean-square roughness of the surface 0.2–0.3 nm. Since the real roughness in the multilayer structure amounts to 0.6–1.2 nm, this value could not affect the reflection coefficient of the multilayer mirror. The preliminary measurements of reflection characteristics were carried out using the laboratory reflectometer [19] at the Institute for Physics of Microstructures and then thoroughly examined at the BESSY-2 synchrotron [20–23]. Figure 5a presents the spectral dependences of the reflection coefficient for the first AMM. For simplicity we did not allow for the deterministic variation of the layer thicknesses, and the approximation was performed using only two values of the roughness (for the Mo on Si and Si on Mo boundaries) and the random deviations of the film thickness from the nominal values. To accelerate the procedure of searching for the best realisation, the maximal deviation of the film thickness from the nominal values was assumed to be ±10% (the root-mean-square amounts to 2%–3%, which is quite sufficient for practical applications). The solid curve in Fig. 5a, providing the best fit with the experimental results, was obtained for the roughness 1.15 nm for the Mo on Si boundary and 0.55 nm for the Si on Mo boundary. These values are in good agreement with the data for periodic multilayer mirrors.

Figures 5b and 5c present the thicknesses of the films calculated and reconstructed from the AMM reflection curve, as well as the ratios of the reconstructed film thicknesses to the nominal values, respectively. From the analysis of Fig. 5 one can see that the reconstructed AMM parameters seem physically reasonable, and the reflection curve provides a good description of the experimental data.

The results of a similar study for the second and third AMMs are presented in Figs 6 and 7. For the second AMM optimised to the range 24–29 nm, the results of measurements and approximation could be made close to each other for the value of roughness of 1.15 nm for the Mo on Si boundary and 0.55 nm for the Si on Mo one. However, the calculated fluctuations of thicknesses for the second AMM appeared to be essentially larger, and for some layers achieved ±40%. Probably, such large fluctuations affected the accuracy of determining the roughness of the interlayer boundaries, too.

For the AMM optimised to the range 28–33 nm the best coincidence of results of calculations and experiments is observed for a zero interlayer roughness, which is a physically inconsistent result. From Fig. 7b one can see that the AMM structure has thicker films and greater spread of nominal thicknesses and, possibly, due to the greater spread of real film thicknesses it becomes necessary to analyse a much greater number of realisations in order to find the suitable one.

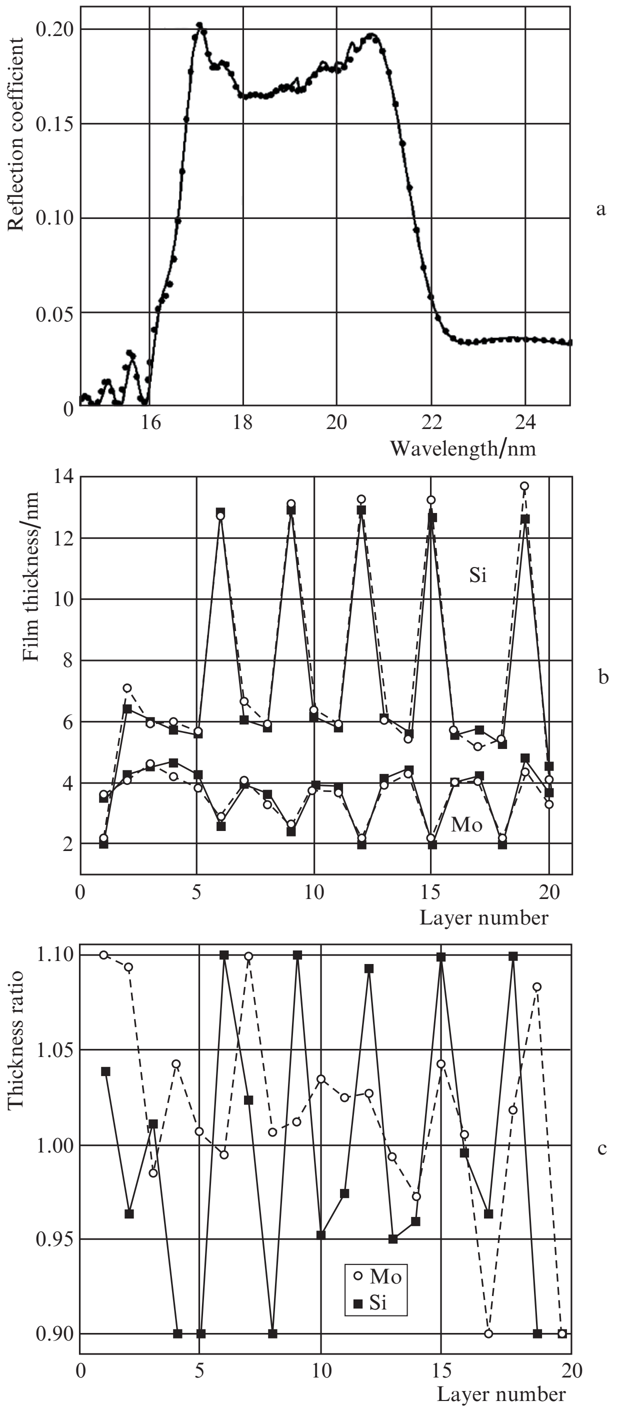

Figure 5. (a) Measured (points) spectral dependence of the reflection coefficient for the first AMM and its best approximation (solid curve), as well as (b) the calculated (■) and reconstructed from the reflection curve (○) film thicknesses in the AMM, and (c) the ratios of the reconstructed thicknesses to the calculated ones as functions of the layer number.

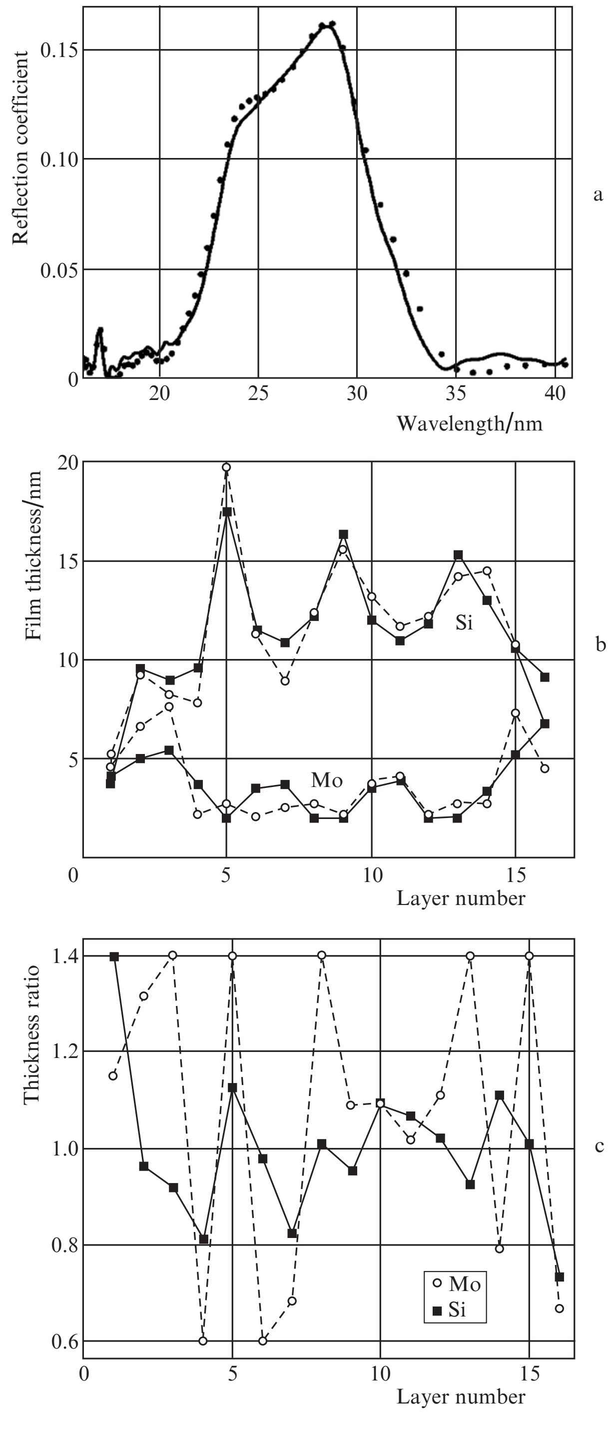

Figure 6. (a) Measured (points) spectral dependence of the reflection coefficient for the second AMM and its best approximation (solid curve), as well as (b) calculated (■) and reconstructed from the reflection curve (○) film thicknesses in the AMM, and (c) ratios of the reconstructed thicknesses to the calculated ones as functions of the layer number.

4. Discussion of the results and basic conclusions

The numerical modelling and experimental study carried out in the present paper by the example of three Mo/Si AMMs allow a number of conclusions, important both for optimising the composition of AMMs and for reconstructing the layer parameters from the data of reflectometry measurements. It is shown that the roughness of interlayer boundaries, as well as the random and deterministic variations of the film thicknesses, considerably affects the shape of the spectral dependence of the reflection coefficient and its magnitude. The possibility of reconstructing the composition of AMMs and the interlayer roughness from the data of reflectometry measurements in the EUV range is demonstrated.

It was found that the presence of interlayer roughness does not lead to additional oscillations in the plateau of the

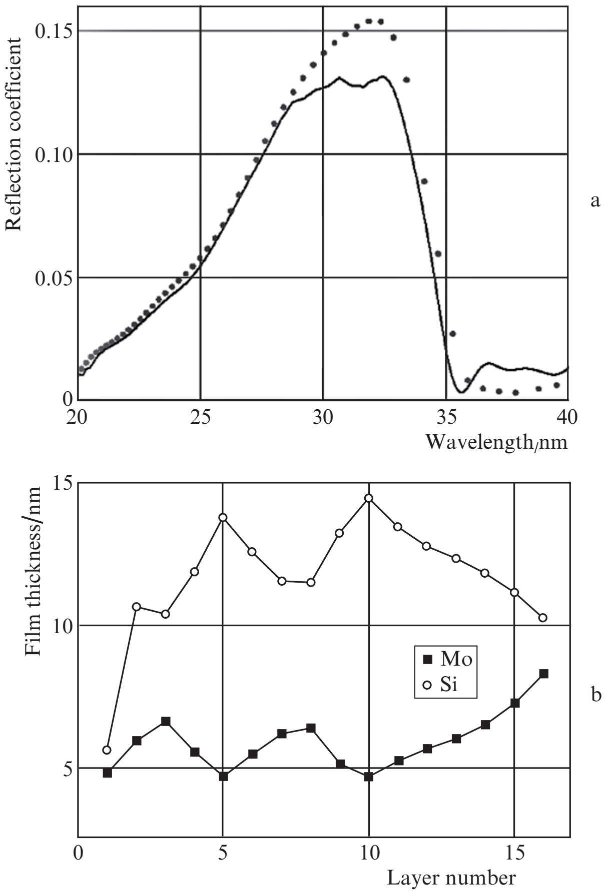

Figure 7. (a) Measured (points) spectral dependence of the reflection coefficients for the third AMM and its best approximation (solid curve) and (b) calculated thickness of films in AMM as functions of the layer number.

spectral dependence of the reflection coefficient. The plateau is only lowered; however, its slope can be affected by the roughness, particularly, in the long wavelength region. The reflection curve is sensitive to the asymmetry of boundaries, and the appropriate change in the reflection coefficient can attain 5%–7% (see Fig. 1). The allowance for the roughness at the very beginning of the optimisation algorithm execution, or the repeated calculation with the solution obtained for the ideal structure taken as the initial approximation, allows at least the preservation of uniformity of the reflection coefficient, avoiding the loss of the integral and peak AMM reflection.

Random fluctuations of layer thicknesses affect the oscillations of the reflection coefficient in the plateau and its absolute value. In particular, for the fluctuations with the variance 5% the distribution half-width of the reflection coefficient exceeds 25% of the nominal value, and it is practically impossible to obtain the desired uniform reflection coefficient. The effect increases under the shift towards the long wavelength region. From the presented data, one can draw the conclusion that in spite of the wide reflection band of the AMM (in our case $\Delta\lambda/\lambda \approx 25\%$), the admissible thickness fluctuations do not exceed 1%–3%. This condition is essentially harder than in the case of periodic multilayer mirrors.

Systematic errors in the layer thicknesses cause the shift of the position of the reflection coefficient plateau towards the long wavelength or short wavelength region depending on the sign of Δd in the process of the AMM growth. In this case, the oscillations in the plateau of the reflection coefficients change insignificantly. The spectral width of the reflection band is practically unchanged at small systematic variations of the thicknesses (with the variance ~1%).

The comparison of the experimental data with the results of numerical modelling has shown that in the presence of random fluctuations of the film thicknesses with the variance 1%–2% it is possible to obtain AMMs with the reflection characteristics, close to the expected ones, and the individual thicknesses can be reconstructed from the data of measurements in the EUV range. The fluctuations with the variance 7%–10% allow the estimation of individual thicknesses, but the reflection curve will be essentially different from the desired one. Larger fluctuations do not allow even the reconstruction of the AMM structure.

Note that the best results were obtained for the AMM, optimised to the wavelength range 17–21 nm. This mirror consists of layers having the minimal thickness, and the individual layer thicknesses are grouped around the values of 2, 4, 6, and 13 nm. At the initial stage of fabricating this AMM a thorough calibration of the film growth rate (i.e., the current of magnetron sputterers and the velocity of substrate passing above the magnetrons) was undertaken within the above thickness ranges. Two other AMMs had thicker layers and the thickness values were distributed almost over the entire range from the minimal value of 2 nm to 17 nm. They were synthesised using the growth rates taken from the approximation of experimental data for the first AMM. From the measurement results, it is seen that this approach does not provide the required accuracy of the layer fabrication.

To summarise, we can formulate the basic criteria to be satisfied in the synthesis of high-quality AMMs. First, at the stage of AMM structure design it is desirable to choose implementations with a minimal number of layer thicknesses. If it appears impossible, then using periodic mirrors as test ones, it is necessary to perform the calibration of the growth parameters for a maximal number of film thicknesses, ideally for each thickness value. Second, it is necessary to keep the stability of the technological process, so that the variance of fluctuations and the deterministic variation of the film thicknesses did not exceed 1%–2%. Third, in the calculation of the AMM film thickness either the roughness should be taken into account from the very beginning of the optimisation algorithm execution, or the repeated calculation should be performed with the solution obtained for an ideal structure taken as the initial approximation. It is necessary to know the roughness of both boundaries and allow for the fact that they can vary depending on the layer thickness. Practically these data can be obtained in the course of analysing the reflection characteristics of periodic mirrors.

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