Technical Physics, 2022, Vol. 92, No. 13

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Manufacturing and research of mirrors with a wide bandwidth for synchrotron applications

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Received April 28, 2021
Revised April 28, 2021
Accepted April 28, 2021

The work is devoted to the development, fabrication and analysis of broadband W / Si multilayer mirrors for a broadband monochromator, calculated for the spectral range of 7-10 keV. The possibility of using the stacking approach to obtain multilayer mirrors with a reflection coefficient of about 30% and a spectral bandwidth $\Delta E/E$ of about 20% is shown. The results of measurements of the angular and spectral reflection curves of the mirror obtained on a laboratory diffractometer and on a synchrotron in Novosibirsk are presented.

Keywords: hard X-ray range, monochromator, synchrotron radiation, broadband mirrors, stack structures, multilayer X-ray mirrors.

DOI: 10.21883/TP.2022.13.52227.128-21

Introduction

Over the course of 40 years the periodic multi-layer mirrors (PMM) serve as a universal tool for building the optical schemes, intended for operation in the whole X-ray and extreme ultraviolet (EUV) ranges of wavelengths (0.01–60 nm). PMM uniqueness compared with other X-ray elements is related to high (up to 70%) reflection coefficients at normal incidence angles in EUV and soft X-ray (SX) ranges, thus providing the high numerical aperture, minimum aberrations and effective use of the source radiation. Development of the manufacturing technology and methods of validation of such structures at this point has allowed to come just to the edge of the theoretical limit of the reflection coefficient values [1–5]. PMM spectral selectivity (ratio between the resonance wavelength and spectral width at half-height of the reflection coefficient) is 20–500, that allows to tune away from the adjacent lines and minimize the background noise. However, PMM are not intended for tasks, where wide bandwidth dE/E is required. Therefore, almost from the beginning [6,7] there was a problem of manufacturing the mirrors with period, that changes with depth, with high (compared to the periodic mirrors) integral reflection coefficient, wider spectral and angular reflectance band. Thus, the multi-layer W/Si mirrors, operating at wavelengths of up to 0.18 Å at glancing incidence angles of 3 mrad, were examined for a research telescope, built as per Kirkpatrick–Baez scheme [8]. The mirror period monotonically decreased with structure depth as per function of $d_i = a(b+i) - c$, where d_i — value of i-th period from surface, $c \approx 0.26$, a and b are optimization parameters. This approach is called ABC-model or supermirror. In studies [9,10] the numerical and analytical methods of calculation of such mirrors parameters in accordance with the required profile of the reflection coefficient were proposed. This approach was used at building the mirrors for synchrotron radiation channels and flow increase from X-ray tube on a sample [11].

The monography [12] is dedicated to development and application of aperiodic mirrors in the area of soft X-ray and EUV radiation.

Despite the success achieved in the area of synthesis of aperiodic mirrors [13–15], on a practical level this task remains very complicated. The main difficulties during manufacturing of X-ray mirror, consisting of a large number (up to 100 and more) of layers with individual thickness and the required reflection coefficient, are mainly related to complexity and duration of the calibration process. Process errors in layer thickness of about 1–2%, not significantly affecting the reflection coefficient of the periodic multi-layer structure, in a broadband mirror can result in substantial distortions [16]. On a practical level, for spattering of N various thicknesses with the required accuracy the N various calibrations are required. At the same time, the materials should be sufficiently studied at the stage of periodic mirrors manufacturing, i.e. film density and interlayer roughness should be known. Such a priori knowledge is required, since

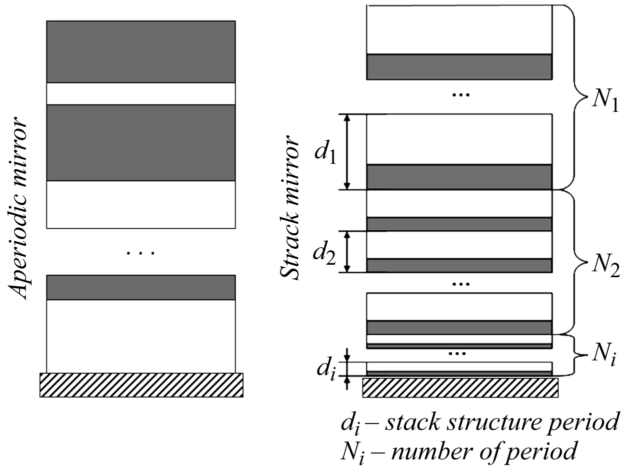

Figure 1. Aperiodic and stack structures.

the result of optimization task solving will be essentially defined by roughness, i.e. the values of optimal thickness of aperiodic multi-layer mirror will be different depending on limits characteristics. After mirror manufacturing its certification is performed. In case of deviations from the design parameters, it is necessary to solve the task of the mirror internal structure reconstruction for the following correction of the process. Inverse problem solving in aperiodic structure class is very complicated, usually any significant information on individual films structure as per reflectometric measurements data is hard to get.

For some task types the approach, proposed in [17] for X-ray telescope of „hard“ X-ray range of (20–40 keV, radiation incidence angle of 0.3°), can be applied. In study [17] the broadband reflecting coating consisted of 8 sputtered on each other periodic multi-layer Pt/C mirrors with periods of 60–30 Å, declining towards substrate. The structure consisted of 168 layers overall, while Pt thickness was fixed at the level of 15 Å, except for the top layer, in which Pt and C were selected equal to suppress the second Bragg peak. Due to its simplicity this method was successfully applied during development of grazing incidence X-ray mirrors for X-ray telescopes InFOCμS and ASTRO-H, as well as for other applications [18–20].

Aperiodic mirrors of such type — stack, consist of several periodic mirrors with dislocated resonance reflection peaks (Fig. 1). In [21], using the example of W/Si mirror, the formula for calculation of stacks number, layers thickness and their sequence, that can be used for determination of the start structure during the mirror structure optimization, is given.

In this study, using the example of multi-layer W/Si mirrors for a broadband monochromator, intended for spectral range of 7–10 keV, the methods, that we developed for optimization, manufacturing and reconstruction of the internal structure of samples as per X-ray reflection data, are described in detail. The results of measurements of angular and spectral curves of the mirror reflection, observed at laboratory diffractometer and experimental station for „X-ray fluorescence elemental analysis“ of VEPP-3 storage of the Institute of Nuclear Physics of the Russian Academy of Sciences, are presented.

Such broadband mirrors can be used in X-ray absorption spectroscopy to suppress glitches, caused by crystals diffraction high-order contributions.

1. Stack structure parameters optimization

Mathematically the optimization task is a minimization of functional [14]

$$F = \int [R(\lambda) - R^{target}]^{2m} d\lambda, \qquad (1)$$

where R^{target} — the target function for the reflection coefficient, defines the „plateau“height. Integral is calculated in the area of determination of R^{target} and examined as a function of layer thickness. The procedure includes several integrations; in the beginning the plateau is selected on the level, corresponding to the periodic mirror, and gradually reduced until reaching the accepted smoothness.

Optimization was performed using the differential evolution algorithm under Multifitting program developed in [22]. As opposed to widely used analogue IMD [23], Multifitting allows to examine the number of period in Ni stacks as a parameter, thus making possible to solve the optimization task in stack structure class. Besides, the transition layers are presented as linear combination of the simplest functions, including error function, that in the best possible way describes the roughness and step-function, describing stoichiometrical layers in the transition area [24].

In case of stack multi-layer mirror, the number of reconstructed parameters is significantly less, the reverse problem can be solved with high level of accuracy, and the corresponding process correction can be performed. The presented observations explain the advantage of stack design use during broadband X-ray mirrors manufacturing, where it is possible.

To calculate the wideband mirror, providing the uniform reflection with coefficient of $R \geq 30\%$, spectral reflectance band $\Delta E/E \sim 20\%$ and suppression of the 3rd harmonic of at least 10^3 in a range of 7–10 keV, the Mo/Si and W/Si

Table 1. Design thickness of Mo/Si layers of stack multi-layer mirrors for range of 7–10 keV

PMM number from of surface	Number of periods, Ni	Material	Thickness of PMM layers, nm
1	13	Si Mo	3.89 1.92
2	32	Si Mo	2.66 2.82

Table 2. Design thickness of W/Si layers of stack multi-layer mirrors for range of 7–10 keV

PMM number from of surface	Number of periods, N_i	Material	Thickness of PMM layers, nm
1	6	Si W	2.78 2.23
2	9	Si W	1.40 3.05
3	21	Si W	1.43 2.97

structures were examined. During calculations the Mo-to-Si transition areas width of 1.2 nm and Si-to-Mo — of 0.6 nm, characteristic for multi-layer Mo/Si mirrors [25–27], were used; film material density values were taken from the table. For W/Si structure the transition area width was 0.3 nm at both limits [28]. Composition of the optimized structures is presented in Tables 1 and 2.

Fig. 2 and 3 show that Mo/Si structure is better in terms of higher harmonics suppression, but worse in terms of reflection coefficient and smoothness, and has more narrow reflectance band at the specified reflection coefficient. Also the operating angles of Mo/Si mirrors are less than of Wi/Si. Therefore, the W/Si structure was selected for synthesis and study.

2. Experimental procedure

The samples were prepared using magnetron sputtering method under an argon atmosphere at pressure of $1 \cdot 10^{-3}$ Torr; polished quartz substrates with effective roughness $\sigma = 0.2{-}0.3$ nm in a spatial frequency range of 0.024–65 μm^{-1} were used as the substrates. Multilayer Cr/Y structure, that allows chemical etching of multilayer mirror in case of failed sputtering process, was used as a sub-layer. The detailed description of the process and unit is presented in [29]. Thickness of sputtered film of the material is defined by a time of substrate passing above target and value of discharge current, therefore the

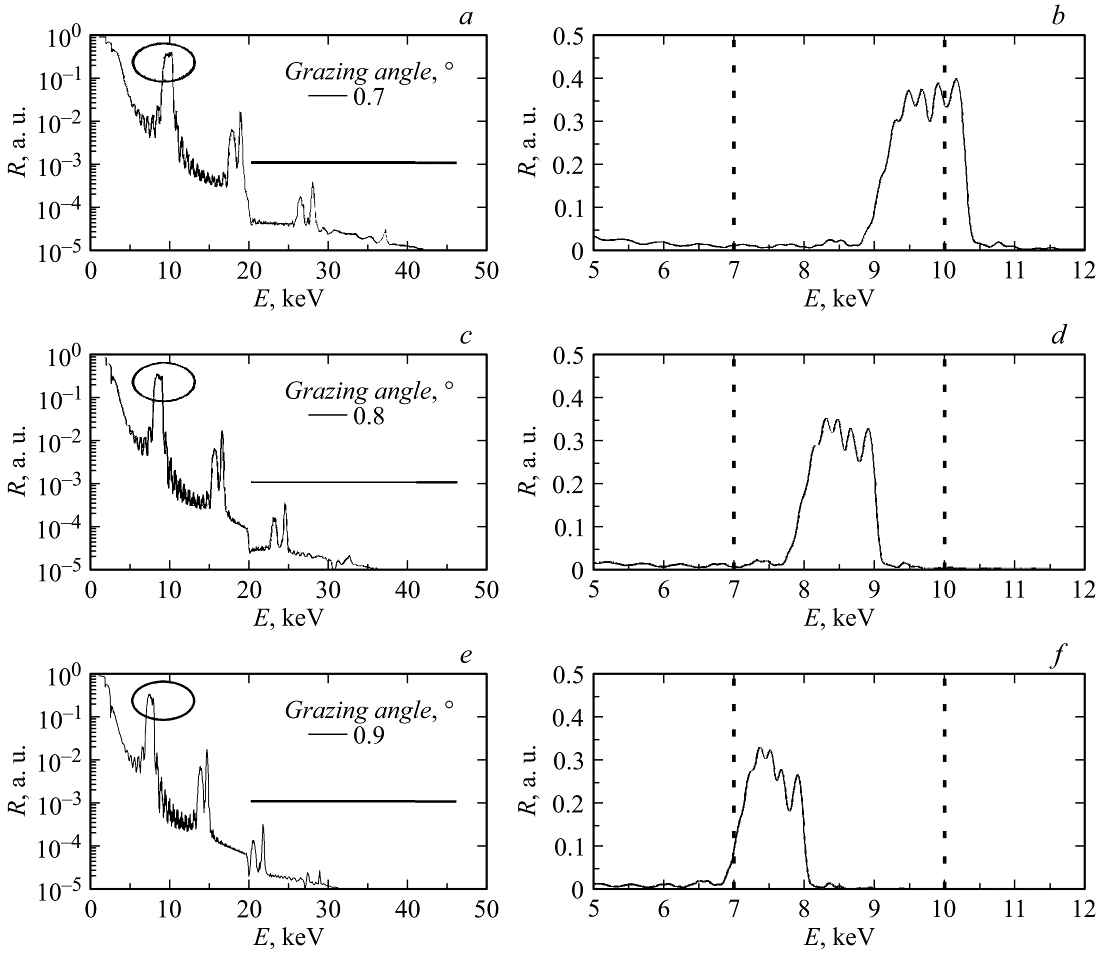

Figure 2. $R(E)$ dependencies for Mo/Si structure at the angle of 0.7° (a, b), 0.8° (c, d) and 0.9° (e, f) with notification of the high-order reflections (left pictures). On the right (on scale) is the reflection coefficient in energies operating range of 7–10 keV for the corresponding angles.

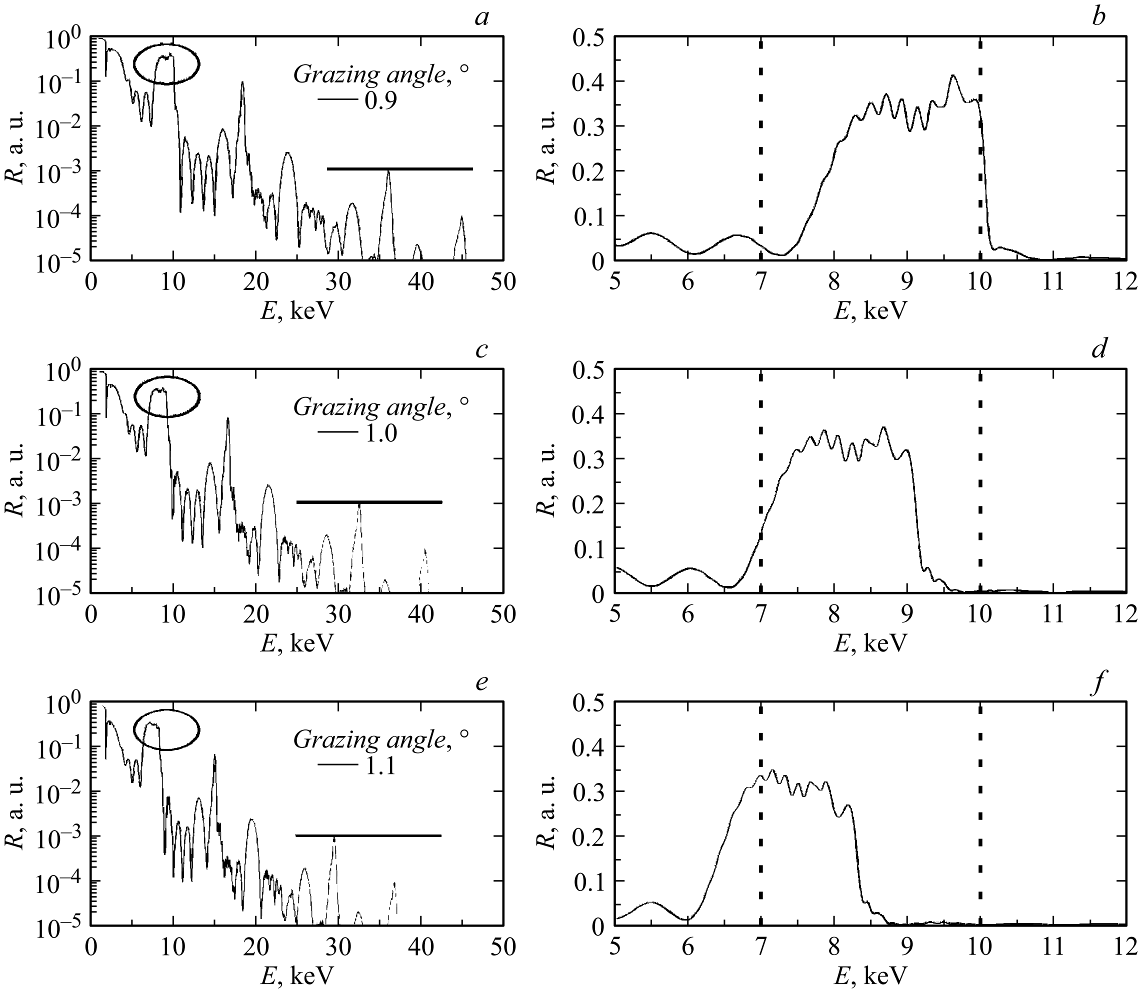

Figure 3. Calculated $R(E)$ dependencies for stack W/Si structure at the angle of 0.7° (*a, b*), 0.8° (*c, d*) and 0.9° (*e, f*) with notification of the high-order reflections (left pictures). On the right (on scale) is the reflection coefficient in energies operating range of 7–10 keV for the corresponding angles.

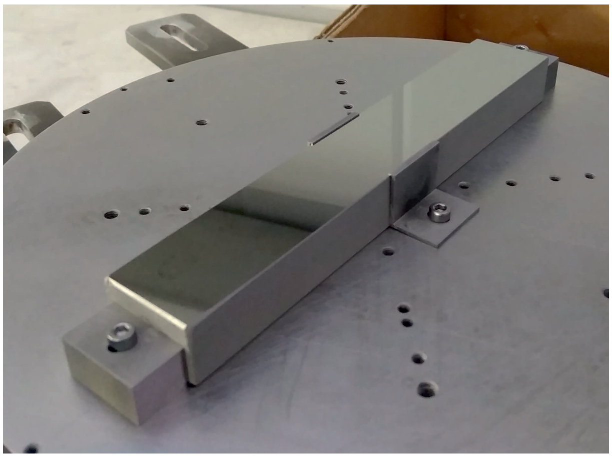

Figure 4. Picture of the synthesized mirror fixed on sputtering substrate holder.

mirror manufacturing process is preceded by calibration procedure, consisting of synthesis of several preliminary periodic mirrors with layers thickness, corresponding to the ones in design broadband structure.

As per the low-angle X-ray diffraction data, observed with use of four-crystal high-resolution diffractometer PANalitycal X'Pert Pro (wavelength is 0.154 nm), the inverse problem is solved, resulting in determination of synthesis parameters of thickness of layers, containing in the required broadband mirror, with high accuracy. The technique of reconstruction of stack parameters as per X-ray reflection data, as well as its application during practical tasks solving are described in detail in [30,31]. However, the inevitable drift of discharge electrical parameters, microbreakdowns, fluctuations and systematic variations of working gas pressure can result in films thickness deviation from the specified values. The procedure of multi-layer mirror manufacturing is iterative and requires determination of mirror parameters and the corresponding correction of the synthesis process. Characteristics of stacks, constituting

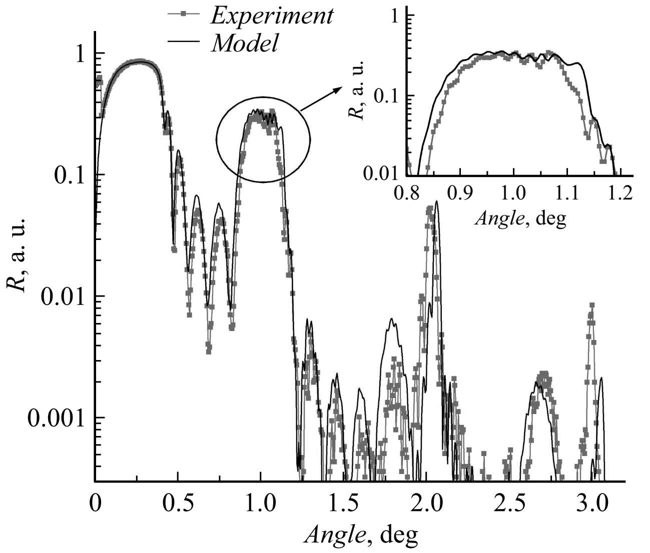

Figure 5. Measured and design angular dependencies of reflection coefficients of W/Si stack structure at $\lambda = 0.154$ nm. Dots — experiment data, solid line — structure model.

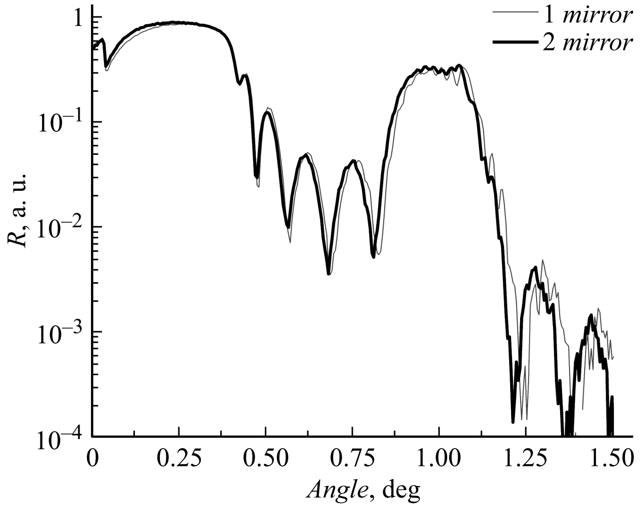

Figure 6. Comparison of experimental curves of $R(\theta)$ at wavelength of 0.154 nm for two identical synthesized stack mirrors.

the broadband mirror, are used as optimization parameters: thickness and density of materials, roughness films/transition areas scales. In case of observing the parameters of reconstructed structure, different from the specified, the sputtering process correction is performed at the following structure synthesis.

Two identical mirrors were made under this study. Picture of one of them on a substrate holder after application of the reflecting coating is presented in Fig. 4. Fig. 5 shows the measured and design angular dependencies of reflection coefficients, taken in angles broad range, up to the 3rd reflection order, for one of the samples. In the upper right corner the curve parts, corresponding to the first reflection order, are presented in detail. In general, it can be said that the experiment and theory are in good agreement.

Good reproducibility of reflecting characteristics of multilayer stack structures should also be mentioned, illustrated in Fig. 6, where the measured reflection curves at wavelength of 0.154 nm for two sequentially synthesized mirrors are presented. The figure shows the good match of the curves.

Due to lack of possibility to perform measurements in the target spectral range, re-calculation was performed using parameters of stack, observed from angular dependence, taken at wavelength of 0.154 nm, to spectral at incidence angle of 1°. Fig. 7 shows the results of re-calculation. As shown on the picture, the expected reflection curve is in good agreement with the target one. At the same time, the actual profile width is a bit more narrow than the target one, which is probably caused by some errors of film thickness during synthesis.

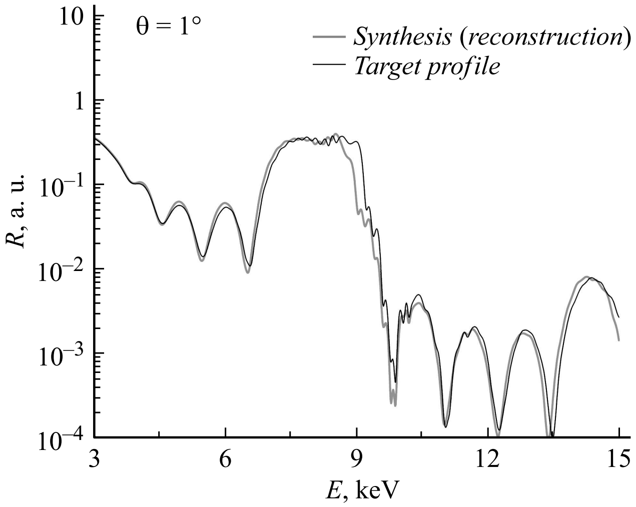

Figure 7. Result of comparison of the expected (based on recalculation data from angular dependence) spectral curve of reflection with target profile at radiation incidence angle of 1°.

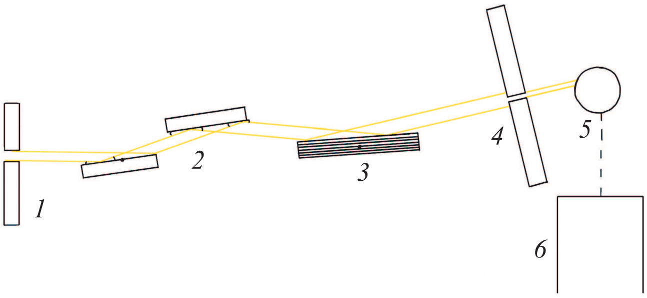

Figure 8. Experiment scheme at the station of „X-ray fluorescence elemental analysis“ of VEPP-3 storage of the Institute of Nuclear Physics of the Russian Academy of Sciences: *1* — entrance slits of monochromator 11 × 3 mm (W × H), *2* — „butterfly-“ monochromator Si(111), *3* — multi-layer mirror, *4* — exit slits of monochromator 6 × 1 mm, for signal not to change due to „butterfly“ shift with energy variation, *5* — diffuser (Bil standard sample), *6* — Si(Li) detector.

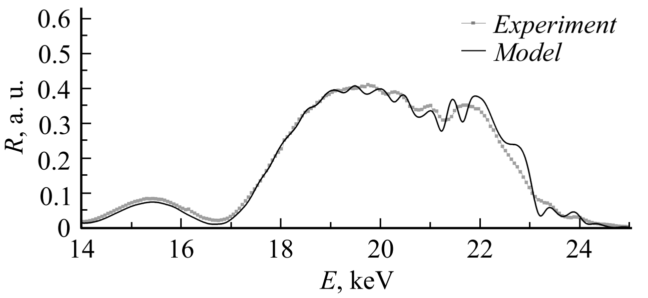

Figure 9. Measured, using synchrotron (symbols), and calculated (solid line), using the reconstructed stack parameters from reflection curve at wavelength of 0.154 nm, spectral dependencies of reflection coefficients.

3. Results of synchrotron measurements

Broadband mirrors samples are intended for X-ray absorption spectroscopy in photons energy range of 7–10 keV. At this stage the measurements of the spectral reflection curve were performed at the experimental station „X-ray fluorescence elemental analysis“ of VEPP-3 storage of the Institute of Nuclear Physics of the Russian Academy of Sciences; the scheme of the experiment is presented in Fig. 8.

Measurements were performed in energy range of 14–25 keV, that is significantly different from the design range. But these measurements are very important not only for synchrotron applications, but also for the technology and manufacturing of the broadbands stack mirrors. The point is that under laboratory conditions the measurements of only angular dependencies of reflection coefficients at narrow characteristic lines of X-ray tube anode material are possible. In our case it is the line of Cu$K_{\alpha 1}$. Synchrotrons usage at this stage of mirrors manufacturing will significantly increase the production time and result in significant increase of such mirrors cost. Therefore, it was extremely important to check the validity of the method, based on reconstruction of stack parameters from angular dependencies of reflection coefficients with the following re-calculation of spectral curves using these data.

Fig. 9 shows the measured, using synchrotron (symbols), and calculated (solid line), using the reconstructed stack parameters from reflection curve at wavelength of 0.154 nm, spectral dependencies of reflection coefficients. As shown on the picture, there is a good match between experimental and design curves, indicating the validity of the approach used at multi-layer stack mirrors manufacturing.

Conclusion

The study is dedicated to development, manufacturing and analysis of multi-layer W/Si mirrors for the broadband monochromator, designed for the spectral range of 7–10 keV and intended for experiments on X-ray absorption spectroscopy. Effectiveness of the stack approach use in manufacturing and characterization of the broadband multi-layer mirrors for synchrotron applications is shown. The main results of this study were the following.

First of all, the effectiveness of the stack approach, when the reflecting coating consists of the set (stack) of periodic multi-layer mirrors with various resonance energies, for broadband mirrors manufacturing for synchrotron applications is shown. The pair of almost identical mirrors, with reflection coefficient of about 30% and spectral bandpass $\Delta E/E$ of about 20% in photons energy range of 7–10 keV, was manufactured.

Secondly, despite the structure optimization for a certain spectral range, it can be used in other ranges as well without strong degradation of the reflection coefficient profile.

Thirdly, it was shown, that samples study using laboratory diffractometer, allowing to study only angular dependencies of reflection coefficients on characteristic lines, is sufficient for forecasting the spectral reflection curves in a broad range of photons energy. The latter is extremely important for the technology of manufacturing the broadband multi-layer mirrors with specified spectral characteristics.

Funding

The study was performed under the grant of the Russian Foundation for Basic Research №20-02-00708 with use of equipment of the Common Research Center „Physics and technology of micro- and nanostructures“ at the Institute for Physics of Microstructures of the Russian Academy of Sciences.

Conflict of interest

The authors declare that they have no conflict of interest.

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