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Aperiodic normal-incidence antimony-based multilayer mirrors in the 8 – 13-nm spectral range

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Abstract**.** **The optical properties of several materials were analysed from the standpoint of fabrication of broadband normal-incidence multilayer mirrors possessing maximal uniform reflectivity in the 8 – 13-nm range. By solving the inverse problem of multilayer optics we show that aperiodic Sb/(B_4C, Sc, Si) multilayer structures optimised for maximum uniform reflectivity in the 8 – 13-nm range are able to afford a normal-incidence reflectivity $R \sim 10\,\%$ throughout this range. The best results are exhibited by the pair Sb/B_4C, for which the average reflection coefficient amounts to about 13 %. The dependence of optimisation result on the programmable limitation on the minimal layer thickness in the multilayer structure was numerically investigated. An empirical rule was established whereby setting the lower bound for a layer thickness at a level $\sim \lambda_{\min}/4$ (in this case, $\lambda_{\min} = 8$ nm) does not result in an appreciable lowering of attainable uniform reflectivity.**

Keywords: *soft X-ray range, antimony-based multilayer mirrors, aperiodic structures, normal incidence of radiation.*

1. Introduction

To solve several optimisation problems which are of importance to soft X-ray (SXR) optics, advantage may be taken of the class of aperiodic multilayer structures (MSs) [1]. Among these problems are, for instance, the calculation and synthesis of MSs that furnish the highest possible uniform reflectivity in a given range of wavelengths or angles of radiation incidence, a high polarisance in a broad wavelength range at a fixed angle of incidence, the highest attainable reflectivity at one or several wavelengths, the highest attainable integral reflection coefficient of one mirror or the highest attainable integral ‘transmittance’ for a system comprising a sequence of several multilayer mirrors (MMs) and filters, etc. The inclusion of the phase of amplitude reflection coefficient (along with its modulus) permits finding the structures suitable for the reflection of attosecond SXR radiation pulses and manipulation of their shape and duration [2, 3].

Broadband aperiodic mirrors are employed in the investigation of elementary processes involving multiply charged ions executed with stigmatic (imaging) spectrographs [4 – 9], for diagnostics of plasmas, including laser-produced microplasma, for recording the high-order harmonic spectra of laser radiation and the SXR pulses generated by free-electron lasers [10] or other sources, for the reflection of attosecond SXR radiation pulses and transformation of their duration, etc. The necessity of maximising the integral ‘transmittance’ for several successive reflections in a system of mirrors arises, in particular, in X-ray lithography. Recently, a Mo/Si MM optimised for maximum uniform reflectivity in the range 12.5 – 25 nm at normal incidence was employed in experiments in the conversion of Ti : sapphire laser radiation ($\lambda \sim 0.8$ μm) to SXR radiation. The frequency was up-converted in the reflection from a relativistic plasma wave driven by a multiterawatt laser in a pulsed helium jet (a relativistic ‘flying mirror’) [11]. In comparison with optical schemes involving grazing-incidence mirrors, schemes with normal-incidence mirrors are particularly valued for their small aberrations and the capacity to form optical images – of course, when it is possible to fabricate aperiodic structures offering a sufficiently high reflection efficiency.

Broadband normal-incidence mirrors based on aperiodic Mo/Si MSs were introduced into practical SXR spectroscopy by Kondratenko et al. [4], Kapralov et al. [5], Ragozin et al. [6], Levashov et al. [7, 8], Beigman et al. [9], and Kando et al. [11], where these mirrors were parts of a stigmatic (imaging) diffraction spectrometer. However, the operating wavelength range of molybdenum-silicon mirrors is limited by the silicon L absorption edge (12.5 nm), and therefore attaining sufficiently high reflectivities at normal incidence at wavelengths shorter than 12.5 nm invites the employment of other material pairs. Artyukov et al. [12] analysed the optical properties of more than 1300 inorganic compounds and elements and found material pairs – components for periodic MMs for the 3 – 30-nm wavelength range. Their results are represented in tabular form; these tables contain information about the composition of multilayer coatings, the theoretically attainable reflection coefficients for periodic mirrors, etc. They determined that good promise is shown, in particular, by periodic structures based on U/C, U/B, U/B_4C, UC/B_4C (naturally, the case in point is depleted uranium, which is composed primarily of ^{238}U), Th/B, La/B, etc. The calculated reflection

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coefficients of these structures amount to 50 % – 80 % in the ranges 4.4 – 11.0, 7.7 – 11.0, 7.7 – 12.3, 6.7 – 7.6, 6.7 – 9.0, and 6.7 – 9.0 nm, respectively.

The requirements imposed on the optical constants of the elements that make up an aperiodic MS depend on the optimisation criterion applied, and they are generally different from those in the case of periodic mirrors. However, the material pairs which yield the best results from the standpoint of the making of periodic structures would also be rather good in the making of aperiodic structures. In particular, the real part of the refractive index of uranium differs from unity much greater than for other elements, while the imaginary part is rather small (see below), which favours the design of efficient uranium-based interference MSs. Calculations of aperiodic MSs in the range 6.7 – 11.1 nm, which rely on the optical constants of pure materials, showed that they are capable of affording a uniform reflectivity of $\sim 7.5\,\%$ (U/B_4C) and $\sim 4.2\,\%$ (La/B_4C) [13].

Several papers (see, for instance, Allred et al. [14], Sandberg et al. [15], Artyukov et al. [16]) are concerned with the optical properties of uranium-based mirrors. At the same time, to our knowledge there are no reports in the literature about the synthesis of stable MMs with the nanolayers of chemically pure uranium. This is hindered by the high chemical activity of uranium (it oxidises and becomes friable, unless the uranium film is under ultrahigh vacuum conditions). In the analysis of the problem of fabrication of stable MMs based on uranium-bearing materials, Artyukov et al. [17] arrived at a conclusion that for the $\lambda > 4.5$-nm range it is expedient to make use of uranium carbides (UC, U_2C_3) and may be of a three-component substance of the type $(UC)_{1-x}(UN)_x$. However, it is evident that the advantage of uranium as the bearer of remarkable optical constants will wane as the fraction of uranium atoms in the uranium-containing layer is made smaller.

The synthesis of periodic $La/B_4C(B_9C)$ MMs for a wavelength of ~ 6.7 nm has been reported by Platonov et al. [18] and Barysheva et al. [19]. However, their reflectivities at normal incidence turn out to be substantially lower than the theoretical limit, which is attributable to the formation of transition layers.

Montcalm et al. [20], Windt et al. [21], and Sae-Lao and Montcalm [22] reported the synthesis of periodic Mo/Y structures for the 8 – 12-nm range. The normal-incidence reflection coefficients of the Mo/Y mirrors amounted to $\sim 45\,\%$, $\sim 35\,\%$, and $\sim 20\,\%$ at wavelengths of 11.5, 9.4, and 8.2 nm, respectively [22]. Wang et al. [23] reported the making of polarisers based on Mo/Y aperiodic structures with approximately constant reflectivities at levels of 5.5 % and 6.1 % in the ranges 8.5 – 10.0 and 9.3 – 11.7 nm, respectively, for an angle of incidence of 45°. In this case, going over to normal incidence would result in a lowering of the MM reflectivity.

This state of affairs has impelled us to resume the quest for material pairs suited for the fabrication of efficient broadband normal-incidence mirrors for the wavelength domain below ~ 13 nm. We therefore set ourselves the goal of elucidating the possibility, in principle, of the fabrication of broadband normal-incidence mirrors in the $\lambda < 13$ nm domain and calculating their highest performance characteristics determined by the optical constants of the corresponding elements. In doing this we operate on the premise that technological problems, should they emerge, in the path of synthesis of these mirrors will be overcome, as were the difficulties encountered by Zuev et al. [24] in the making of stable MMs based on the structure Mg/Si.

2. Selection of materials and numerical techniques

In the selection of material pairs we took into account the values of optical constants on the interval accepted for optimisation, positions of their absorption edges, as well the compatibility of material pairs with consideration for their reactivity.

Attempts to formulate an analytical criterion indicating the optimal material pair for the solution of the problem in hand do not meet with success. The final judgement is reached proceeding from numerical solutions (with the reservation that only a finite number of local extrema of the multiparametric optimisation problem may be found in a calculation time).

The (intensity) reflection coefficient R for an interface between two substances at normal incidence is defined by the formula

$$R = \frac{(\delta_2 - \delta_1)^2 + (\gamma_1 - \gamma_2)^2}{16}, \quad |\delta_i|, \gamma_i \ll 1, \tag{1}$$

where δ_i and γ_i are the real and imaginary additions to unity in the permittivity ε_i: $\varepsilon_i = n_i^2 = 1 - \delta_i + \mathrm{i}\gamma_i$, while the subscript $i = 1, 2$ numbers the substances. The optical constants δ_i and γ_i of the substances are expressed in terms of atomic scattering factors f_1 and f_2:

$$\begin{pmatrix}\delta\\ \gamma\end{pmatrix} = \frac{r_0}{\pi}\lambda^2 N_\mathrm{a} \begin{pmatrix}\sum\limits^{k} \alpha_j f_{1j}\\ \sum\limits_{j=1} \alpha_j f_{2j}\end{pmatrix},$$

where $r_0 = e^2/m_\mathrm{e}c^2 = 2.818 \times 10^{-13}$ cm is the classical electron radius; N_a is the density of atoms; α_j is the fraction of the atoms of sort j ($j = 1, ..., k$); λ is the radiation wavelength in vacuum, with $|\delta|, \gamma \ll 1$; and m_e is the electron mass. Data on the atomic scattering factors are available from the literature for elements with nuclear charges from 1 to 92 in the photon energy range 10 eV – 30 keV [25]. From formula (1) it follows that the reflection from each interface becomes stronger with increasing the difference between δ and γ of the corresponding substances. That is why there is good reason to select for the MMs the pairs of materials with a high reflectivity at their interface and not-too-high an absorption coefficient, so that the effective number of interfering beams is high enough.

Presented in our work are the calculations of aperiodic MMs optimised for maximum uniform reflectivity in a given wavelength interval by minimising the functional $\mathfrak{J}_1 = \int [R(\lambda) - R_0]^2 \mathrm{d}\lambda$ (here, R_0 is an optimisation parameter). Such mirrors, as a rule, possess a substantially higher integral reflection coefficient than any periodic mirror whose resonance peak is located within the same wavelength interval. In this case, the number of optimisation parameters is equal to the number of single layers in an aperiodic structure. In the solution of the optimisation problem, the role of initial structures was played by periodic structures. In doing this, it turned out that different initial structures may lead to solutions of practically equal value (from the standpoint of the optimisation criterion employed), despite the fact that the corresponding (optimised) aperiodic structures are much different.

Furthermore, we present calculations of periodic mirrors optimised to maximise their reflection coefficient at a fixed wavelength. In this case, there are only two optimisation parameters (the layer thicknesses of two materials), with a sole independent parameter of the problem.

The optical constants of 11 substances (Mo, Si, B_4C, C, Ti, Co, W, Ni, Cr, Sb, and Sc) were analysed. Figures 1 and 2 depict the wavelength dependences of δ and γ for those of them which show the greatest promise for making MMs in the 8 – 13-nm range. One can see that the real component of the refractive index of antimony departs from unity much greater than in other elements, while the imaginary is rather small in the domain of interest to us, thus underlying the selection of antimony as the first of MM components. Numerical experiments revealed that Sb and B_4C make up the best pair from the viewpoint of synthesis of MMs for the specified spectral domain.

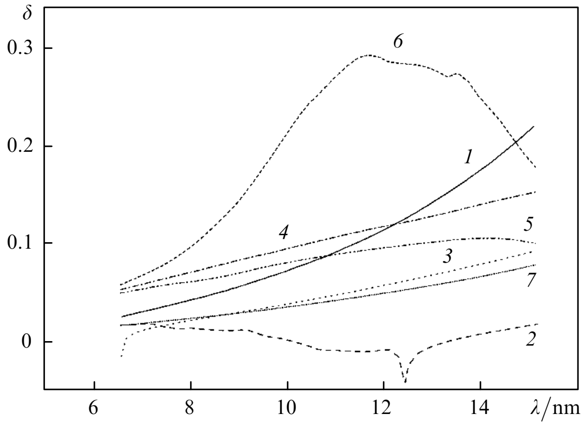

Figure 1. Addition to the real part of the permittivity of Mo (*1*), Si (*2*), B_4C (*3*), Co (*4*), Ni (*5*), Sb (*6*), and Sc (*7*) in the wavelength range 6.5 – 15.2 nm.

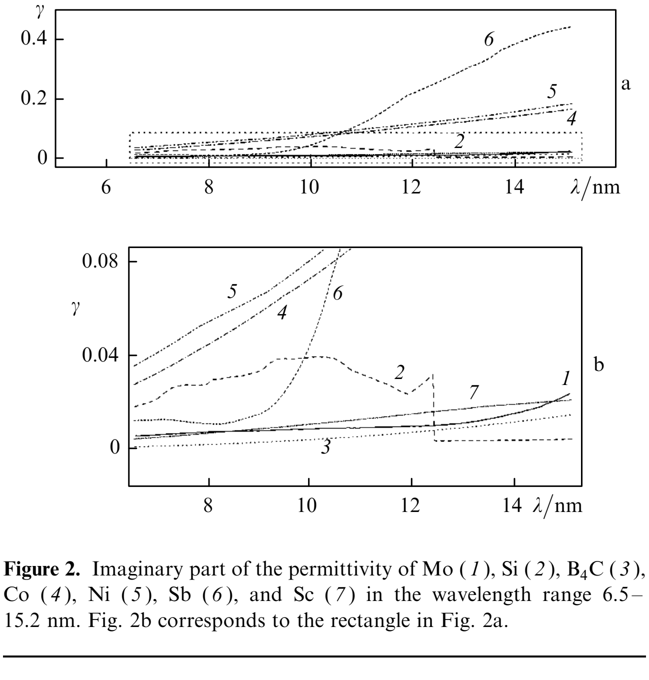

Figure 2. Imaginary part of the permittivity of Mo (*1*), Si (*2*), B_4C (*3*), Co (*4*), Ni (*5*), Sb (*6*), and Sc (*7*) in the wavelength range 6.5 – 15.2 nm. Fig. 2b corresponds to the rectangle in Fig. 2a.

3. Calculation of Sb/B_4C periodic mirrors

We performed calculations of Sb/B_4C-based periodic MSs. They were optimised to attain the highest reflectivity at a fixed wavelength; the layer thicknesses of the two substances served as optimisation parameters. Figure 3 shows the spectral reflection coefficients of the Sb/B_4C(Sc) MSs whose characteristics are collected in Table 1. The number N of layers in each structure was so selected as to attain the effect of saturation, so that a further increase in their number did not lead to a reflectivity increase of any significance. The selected number of layers of the periodic structure set the upper bound for the number of single layers in the optimisation of aperiodic MSs. With increasing wavelength, the number of layers required to attain saturation became smaller, which is attributable to the growth of absorption in antimony. Referring to Fig. 3, shifting to the short-wavelength part of the spectrum relative to the peak at 8.5 nm entails a lowering of the peak reflectivity due to a drastic decrease of δ in antimony, while shifting to longer wavelengths involves a decrease in reflectivity owing to the rapidly growing absorption in antimony (see Fig. 2). Sb/Sc-based MSs rank below the Sb/B_4C structures because of a (two- to three-fold) higher absorption in scandium than in boron carbide.

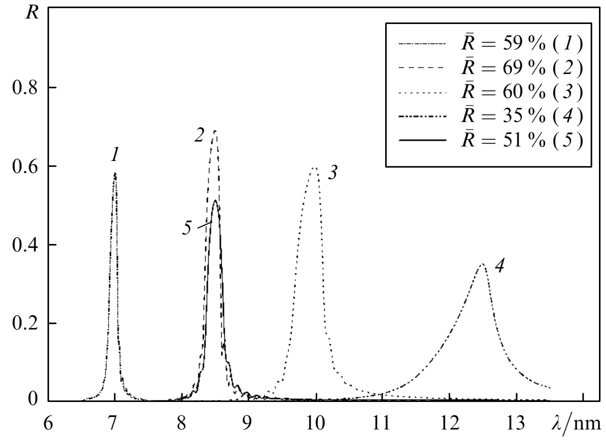

Figure 3. Reflection coefficients of periodic MSs optimised for maximum reflectivity at wavelengths of 7.0 nm [Sb/B_4C, (*1*)], 8.5 nm [Sb/B_4C, (*2*) and Sb/Sc, (*5*)], 10.0 nm [Sb/B_4C, (*3*)], and 12.5 nm [Sb/B_4C, (*4*)].

Table 1. Reflection characteristics of antimony-based periodic structures.

MS	Curve number in Fig. 3	λ/nm	R (%)	$\mathfrak{J}$/nm in the range 6 – 14 nm	N
Sb/B_4C	*1*	7.0	58	0.09	250
Sb/B_4C	*2*	8.5	69	0.21	150
Sb/B_4C	*3*	10.0	60	0.33	100
Sb/B_4C	*4*	12.5	35	0.34	80
Sb/Sc	*5*	8.5	51	0.17	120

4. Broadband normal-incidence mirrors for the 8 – 13-nm range

Aperiodic MSs were optimised to attain a uniform reflection coefficient (all characteristics of the aperiodic MSs of this Section are collected in Table 2). Because of Sb absorption edges (N_1 edge at a wavelength of 8.1 nm, N_2

Table 2. Reflection characteristics of aperiodic MSs.

MS	Figure No. (Curve No.)	Optimisation range/nm	$\bar{R}$ (%)	$\Im$/nm in the optimisation range	$t_{\min}$/nm	R_0 (%)	N
Ni/B_4C	4 (*2*)	6.6 – 8.6	2.2	0.04	0.5	2.2	200
Co/B_4C	4 (*3*)	8.6 – 12.5	2.4	0.08	0.5	2.4	250
Co/B_4C	4 (*1*)	6.6 – 12.5	1.4	0.08	0.5	1.4	200
Sb/Si	5	8.1 – 12.8	8.7	0.42	1.0	10	150
Sb/Sc	5	8.1 – 12.8	10.0	0.47	1.0	11	200
Sb/B_4C	5	8.1 – 12.8	11.9	0.57	1.0	13	200
Sb/B_4C	4, 6, 7	8.1 – 12.8	13.1	0.62	1.5	15	200
Sb/B_4C	6	8.0 – 14.0	11.2	0.67	0.5	13	150
Sb/B_4C	6	8.1 – 15.0	10.1	0.70	1.5	11	200
Sb/B_4C	7, 8	8.1 – 12.8	13	0.61	2.2	15	200
Sb/B_4C	8	8.1 – 12.8	12	0.56	2.2	12.5	200
Sb/B_4C	8	8.1 – 12.8	9	0.42	2.2	9	200

and N_3 edges at a wavelength of 13 nm), the optimisation domain ranged from 8.1 to 12.8 nm. The thicknesses of all single layers were optimisation parameters. It turned out that aperiodic Sb/B_4C MSs ($R = 13.1\,\%$, $N = 200$) are far superior to the corresponding structures based on the substances from the indicated list without antimony ($R \sim 2\,\%$) (Fig. 4). The integral reflectivity $\Im_2 = \int R(\lambda)\mathrm{d}\lambda$ of the antimony-based structure in the range 8 – 14 nm is equal to 0.68 nm, which exceeds the integral Co/B_4C- and Ni/B_4C-based structure reflectivities ($\Im_2 \sim 0.1$ nm) by nearly an order of magnitude.

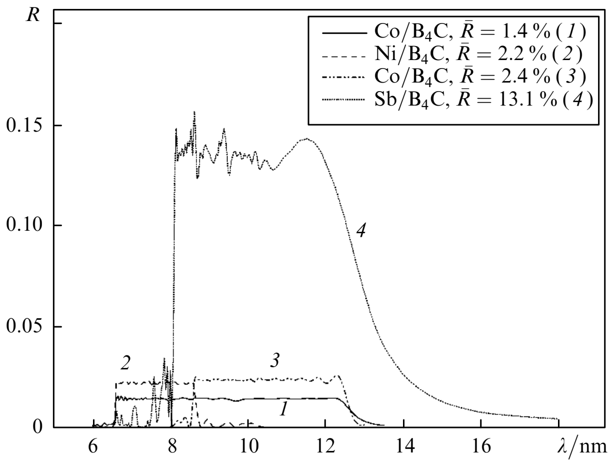

Figure 4. Spectral profiles of the reflection coefficient of aperiodic MSs based on Sb/B_4C (8.1 – 12.8 nm, $N = 200$), Co/B_4C (8.6 – 12.5 nm, $N = 250$), Co/B_4C (6.6 – 12.5 nm, $N = 200$), and Ni/B_4C (6.6 – 8.6 nm, $N = 200$) optimised for maximum uniform reflectivity in the specified spectral ranges; $\bar{R} = \Im_2/\Delta\lambda$ is the average reflection coefficient in the optimisation range $\Delta\lambda$.

Varying the second substance (the first is Sb) showed that Sb/B_4C is the best structure among antimony-based aperiodic MSs optimised for maximum uniform reflectivity. The reflectivities of the Sb/Sc ($N = 200$) and Sb/Si ($N = 150$) pairs are also reasonably high (Fig. 5).

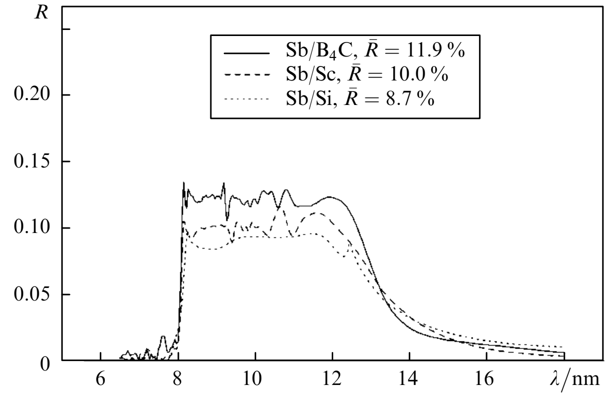

Figure 5. Spectral profiles of the reflection coefficient of aperiodic Sb/(B_4C, Sc, Si) MSs optimised for maximum uniform reflectivity at normal incidence in the 8.1 – 12.8-nm spectral range.

Optimisation of Sb/B_4C-based MSs in a broader range showed that advancement to the long-wavelength part of the spectrum involves a lowering of the average reflectivity and an increase in integral reflectivity (Fig. 6, Table 2). However, an attempt to advance to shorter wavelengths is attended with a substantial lowering of the average reflectivity and an impairment of the uniformity of the reflection coefficient. Near the short-wavelength boundary of the spectral range, the reflectivity becomes lower than at the long-wavelength boundary, and the integral reflectivity becomes lower in this case.

As is well known, the synthesis of MSs may be attended with the formation of transition layers with optical constants different from the constants of pure substances. This results in a lowering of the MS reflectivity. For instance, in

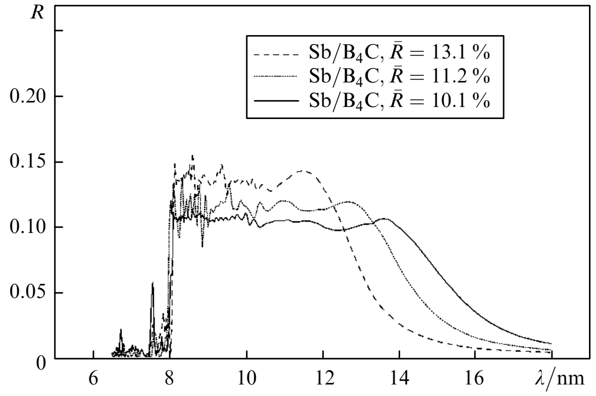

Figure 6. Spectral profiles of the reflection coefficient of Sb/B_4C aperiodic MSs optimised for maximum uniform reflectivity at normal incidence in the spectral ranges 8.1 – 12.8 nm ($N = 200$), 8.0 – 14.0 nm ($N = 150$), and 8.1 – 15.0 nm ($N = 200$).

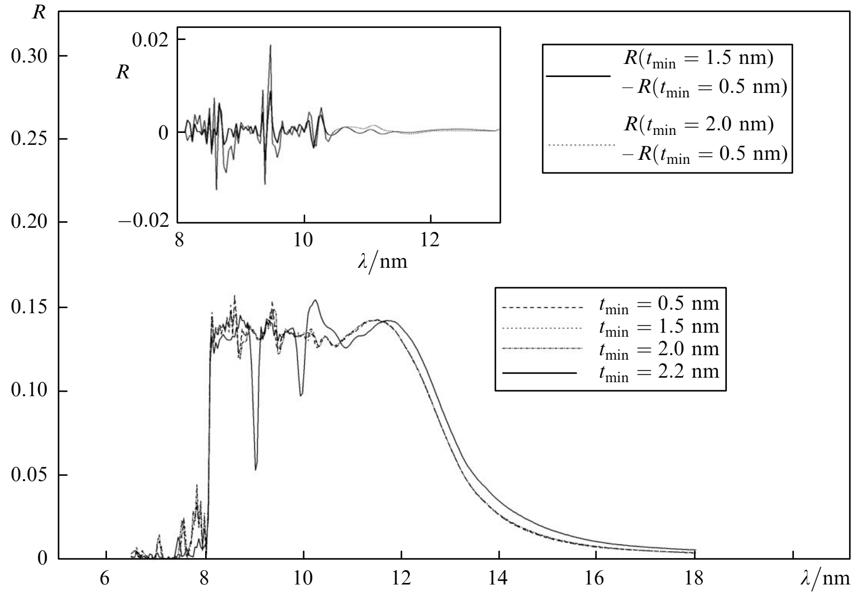

Figure 7. Effect of the programmable minimal layer thickness limit t_{min} on the reflection coefficients of aperiodic Sb/B_4C MSs optimised for maximum uniform reflectivity at normal incidence in the 8.1 – 12.8-nm range. The inset shows the differences between the reflection coefficients R.

the fabrication of Mo/Si mirrors there forms a silicide layer with a composition close to that of $MoSi_2$. The parameters of transition layers in antimony-bearing MSs, to the best of our knowledge, have not been reported.

In the optimisation we introduced a programmed limit on the minimal single-layer thickness. This stems from the necessity to eliminate physically absurd solutions (a layer thickness may not be smaller than the dimension of an atom or a molecule) and improve the stability of the reflectivity of the MS being synthesised relative to the formation of transition layers (ideally, the thicknesses of 'pure' substance layers should far exceed the thicknesses of transition layers). To investigate the effect of such a limitation on the solution of the problem, the MSs were optimised for maximum uniform reflectivity for different values of the minimal possible layer thickness. It turned out that optimisations for $t_{min} = 0.5$, 1.0, 1.5, and 2.0 nm gave rise to different structures, which furnished practically the same spectral profiles of the reflection coefficient (Fig. 7). Structures with thicker layers are preferred for synthesis, because it is not known whether transition layers emerge in the synthesis of Sb/B_4C-based mirrors and, if they do, what is their thickness and composition. The effect of transition layers on the reflectivity will be weaker for thicker 'pure' substance layers. It is pertinent to note that 2 nm is just about $\lambda_{min}/4$, where λ_{min} is the short-wavelength bound of the optimisation range. On introduction of a stronger programmed limitation on the minimal layer thickness ($t_{min} > 2$ nm) without changing the remaining optimisation parameters, the uniformity of the solution was impaired drastically (the solid curve in Fig. 7). A very high uniformity may be obtained at a sacrifice in the average reflection coefficient on the optimisation interval (by lowering the optimisation parameter R_0) (Fig. 8).

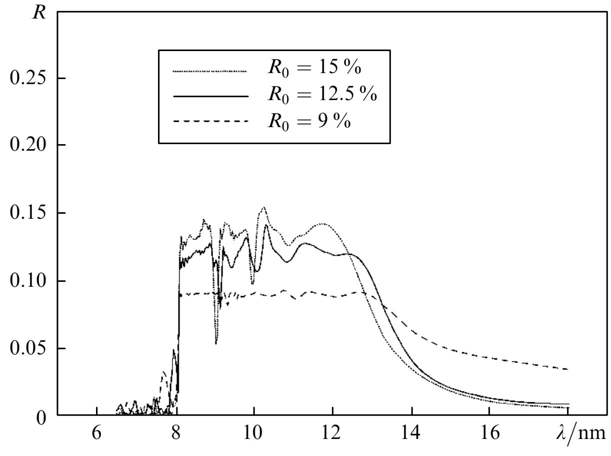

Figure 8. Spectral profiles of the reflection coefficient of aperiodic Sb/B_4C MSs (8.1 – 12.8 nm, $N = 200$, $t_{min} = 2.2$ nm) optimised for maximum uniform reflectivity for different optimisation parameters R_0.

5. Conclusions

With the inclusion of only the optical properties of pure materials, neglecting their chemical activity and the formation of transition layers, Sb/B_4C-, Sb/Sc-, and Sb/Si-based structures optimised for maximum uniform reflectivity in the 8 – 13-nm range were shown to be capable of affording a reflection coefficient $R \sim 10$ % at normal incidence of radiation in this range. The best results are exhibited by the Sb/B_4C pair, whose average reflection coefficient exceeds 13 %. The dependence of optimisation result on the programmed limitation on the minimal layer thickness of the multilayer structure was numerically investigated. An empirical rule was determined, whereby imposing a lower bound for a layer thickness at a level $\sim \lambda_{min}/4$ (in our case, $\lambda_{min} = 8$ nm) does not entail an appreciable lowering of the attainable (uniform) reflectivity.

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