Technical Physics, 2022, Vol. 67, No. 8

09

Prospective wavelengths for projection lithography using synchrotron radiation

© N. I. Chkhalo, V. N. Polkovnikov, N.N. Salashchenko, R.A. Shaposhnikov

Institute of Physics of Microstructures, Russian Academy of Sciences,
607680 Nizhny Novgorod, Russia
e-mail: chkhalo@ipmras.ru

Received April 26, 2022
Revised April 26, 2022
Accepted April 26, 2022

Promising wavelengths for next-generation lithography with a wavelength shorter than 13.5 nm based on a synchrotron X-ray source are discussed. Theoretical and experimental values of the reflection coefficients of multilayer X-ray mirrors providing the maximum reflectivity in the range of 11.4−3.1 nm are presented. The theoretical efficiency of multilayer optics is compared for different wavelengths.

Keywords: X-ray lithography, multilayer X-ray optics, multilayer X-ray mirrors.

DOI: 10.21883/TP.2022.08.54567.102-22

Introduction

The successes achieved so far by extreme ultraviolet (EUV) lithography with a working wavelength of 13.5 nm, allow us to achieve a resolution of 13 nm [1]. This means that in a single process of illumination / manifestation without the use of multiple exposure, the minimum period of the created structures in the resistor is from 26 nm or more. Moving towards increasing the resolution of the projection scheme of the EUV lithograph requires an increase in the numerical aperture NA of the projection lens. A projection lens with $\mathrm{NA} = 0.5$ is currently being discussed. However, as shown in [2], developers are facing serious technological problems that have repeatedly shifted the timing of the appearance of such a lens. In addition to optical problems, fundamental ones have also arisen. The problem lies in the strong shielding of the reflective elements of the mask at increased irradiation angles. To solve this problem, the authors in [2] suggested using a lens with different magnifications in different directions.

An alternative to increasing the numerical aperture to increase the resolution of lithography is to reduce the wavelength, in proportion to which the resolution will increase. In the study [3], it was proposed for the first time to use a wavelength in the vicinity of 6.7 nm for next-generation lithography. This wavelength was chosen because, firstly, it immediately doubles the resolution, and secondly, in this La/B range, multilayer mirrors theoretically have a reflection coefficient of more than 80%. It should also be noted that at that time La/B_4C multilayer X-ray mirrors (MXRM) already reflected about 40% [4].

However, subsequently, due to insufficiently high experimental reflection coefficients of multilayer mirrors, about 60% [5,6], narrow, in comparison with the emission band of the source, the bandwidth of the multi-mirror system, low conversion efficiency of laser-plasma sources based on ions Tb and Gd [7,8], and also, the mismatch of the reflection maxima of the MXRM and the emission band of the laser-plasma source, in [9], a conclusion was made about the futility of this wavelength for lithography.

In general, one of the main factors that reduce the performance of promising schemes of lithographic installations is the discrepancy between the maxima of reflection of the MXRM and the emission of plasma sources. In fact, only Sn and Li ions emit in the region where the maximum reflection coefficient Mo/Si of multilayer mirrors is observed.

This problem is automatically eliminated in the case of using synchrotron radiation sources, since the wavelength and spectral width of undulatory radiation can be adjusted within wide limits. Therefore, when choosing the working wavelength of the lithograph, you need to focus only on the maxima of the reflection coefficients of the MXRM and the spatial resolution that can be obtained at the selected wavelength.

In this paper, a corresponding analysis is made and the theoretical efficiency of 11 mirror systems at different wavelengths is compared. The latest results on the achieved experimental values of reflection coefficients MXRM are also presented.

1. Justification of the choice of the type of mirror optics for lithography with soft X-ray radiation

The features of the interaction of EUV and soft X-ray (SXR) radiation with a substance are its weak polarizability, the refractive index is almost equal to one, and absorption is observed for all substances. According to [10], the refractive index can be written as

$$n = 1 - \delta - i\gamma, \qquad (1)$$

$$\begin{pmatrix}\delta\\ \gamma\end{pmatrix} = \frac{r_0}{2\cdot\pi}\cdot\lambda^2\cdot N\cdot\begin{pmatrix}f_1\\ f_2\end{pmatrix}, \qquad (2)$$

where δ — dispersion additive to the refractive index, γ — imaginary part responsible for absorption, r_0 — classical electron radius, λ — wavelength, N — the concentration of atoms per unit volume, f_1 and f_2 — the real and imaginary parts of the atomic scattering factor. Values of atomic scattering factors in the photon energy range 50–30 000 eV can be found in [11], and the parameter γ is quite significant. The radiation transmission length in a substance μ can be expressed as

$$\mu = 4\pi\gamma/\lambda. \qquad (3)$$

In the wavelength range 3–13 nm μ is the value from fractions to units of micrometers. Therefore, in this wavelength range, optics can only be of the reflective type and must be used in a vacuum.

Since the refractive index is slightly less than one, the phenomenon of total internal reflection is observed. In X-ray optics, it is called a complete external. The angle of total external reflection, or critical, is determined by the parameter δ:

$$\theta_c \approx \delta^{1/2} \qquad (4)$$

and can be approximately written as [12]:

$$\theta_c = \begin{cases}(1-2)\lambda, & 1.5\,\text{nm} \le \lambda \le 20\,\text{nm},\\ (1-3)\lambda, & \lambda \le 1.5\,\text{nm},\end{cases} \qquad (5)$$

where θ_c is expressed in degrees, and λ — in nanometers. For example, for $\lambda = 6$ nm and for a rhodium mirror, the critical angle will be about 6° or sin 0.1 rad. In other words, in the SXR range, the sliding angle of total external reflection does not exceed several degrees.

On the other hand, it is known from the Rayleigh criterion that the diffraction-limited spatial resolution is related to the wavelength and numerical aperture of the mirror constructing the image by the ratio

$$\delta x = k\cdot(\lambda/NA), \qquad (6)$$

where for unpolarized radiation $k = 0.61$. For the case of sliding fall optics, the expression (6) can be rewritten as

$$\delta x \approx 0.61\cdot(\lambda/\theta_c). \qquad (7)$$

Substituting in (7) the value of the critical angle, it can be shown that for the optics of the grazing incidence, the maximum resolution will be about 10 wavelengths. For example, for a wavelength of 6 nm, the resolution will be 60 nm. Thus, it is obvious that even though the wavelength is shorter than in EUV lithography, grazing incidence optics cannot be used for projection lenses of lithographic systems. Therefore, it is necessary to use multilayer X-ray optics in this range as well.

2. Calculation of the reflective characteristics of the MXRM and comparison of the effectiveness of various systems

Multilayer X-ray mirrors are a system of periodically arranged pairs of layers of various materials, of which one is — weakly absorbing (anti-scattering), the second — strongly absorbing (scattering). The number of periods in the MXRM varies from tens to several hundred. The principle of operation of the MXRM is based on the interference of waves reflected from different boundaries. When the Bragg condition is met

$$2d\cdot\sin\theta_{\text{Br}} = m\cdot\lambda, \qquad (8)$$

where d — period, θ_{Br} — Bragg angle measured from the surface, m — reflection order and λ — wavelength, waves interfere constructively. This leads to the fact that, despite the reflection coefficients from individual borders being low in fractions of percent, the total reflection coefficient reaches tens of percent.

A large number of works have been devoted to the basic principles and the search for optimal pairs of materials to achieve the maximum possible reflection coefficients of multilayer mirrors at specific wavelengths [13,14]. We will not dwell on this problem in detail, we will summarize only the fact that everything has come down to the fact that it is advantageous to use a material with K-, L- as a weakly absorbing (anti-scattering) material, and sometimes M-the absorption edge is as close as possible and slightly shorter in wavelength than the working one. If there are several candidates, it is better to choose the lightest material.

The choice of a highly absorbing (scattering) material is not so obvious. Initially, it was thought that it should be as heavy a material as possible, most often tungsten, since it provides maximum optical contrast and, accordingly, the reflection coefficient at the boundary of a multilayer mirror. However, it quickly became clear that, in addition to high optical contrast, this material should also have minimal absorption. In particular, in the range of 4.3–6.6 nm, W/C MXRM was replaced by systems based on transition metals: Ni/C, Co/C and Cr/C.

Since the wavelength range of 3–13 nm (the wavelength limit will be discussed below) has already been well studied in the literature, the most promising materials were known, which allowed us to limit ourselves to several pairs of materials.

Since not only the peak values of the reflection coefficients are important for constructing an X-ray optical scheme, but also the spectral reflection band, since it forms a requirement for the spectral radiation band of the undulator, the peak values of the reflection coefficient and the spectral bandwidth (at half the height) of the MXRM were calculated. The maximum reflection coefficients of multilayer X-ray mirrors were analyzed in a wide wavelength range of 3–13 nm. In calculations, the best, or

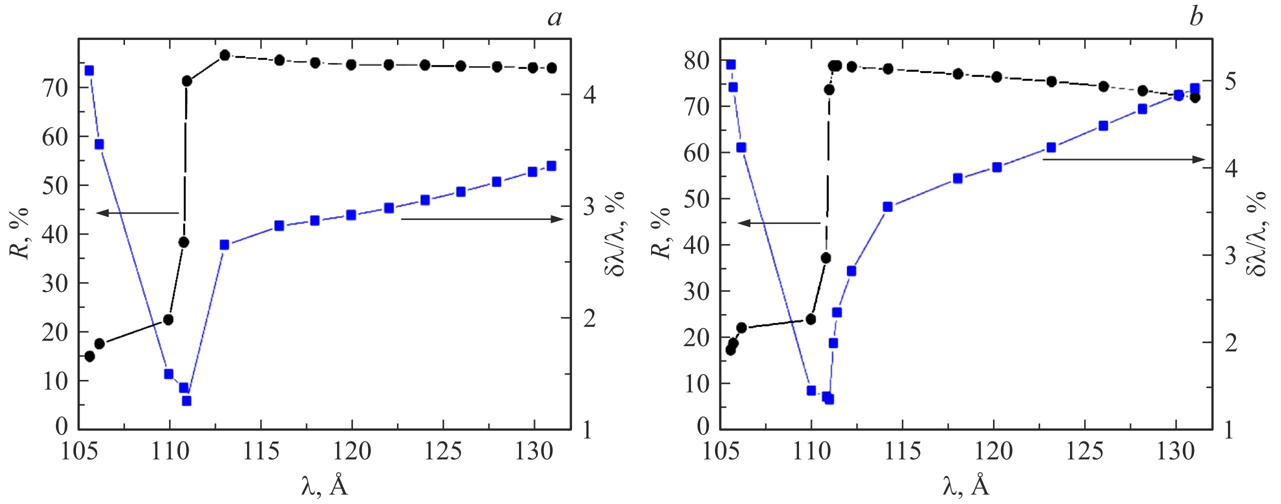

Figure 1. Calculated graphs of the dependence of the reflection coefficient (round symbols) and spectral resolution (square symbols) on multilayer systems Mo/Be (*a*) and Ru/Be (*b*) in the wavelength range 10.7–13.1 nm.

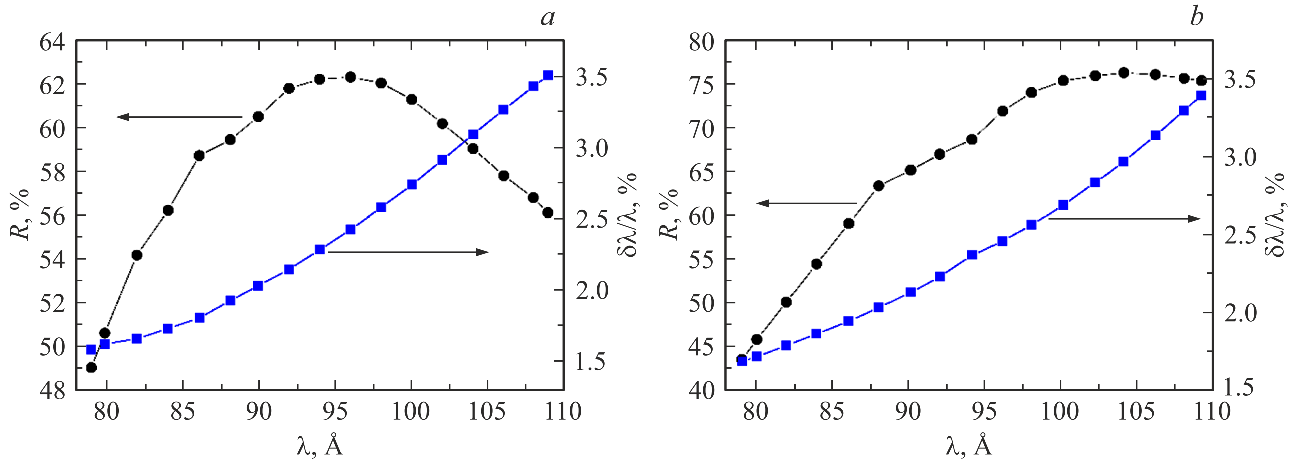

Figure 2. Calculated graphs of the dependence of the reflection coefficient (round symbols) and spectral resolution (square symbols) on multilayer systems Pd/Y (*a*) and Sr/Rh (*b*) in the wavelength range 7.9–10.9 nm.

with an alternative, MXRM are presented. The calculation does not take into account the defects of the multilayer mirror, such as interlayer roughness and the difference in the densities of the film materials from the tabular values.

For the wavelength range 11.1–13.0 nm, the most promising materials are Mo/Be and Ru/Be MXRM. Fig. 1 shows the calculated graphs of the dependence of the reflectivity and spectral resolution on multilayer systems Mo/Be (Fig. 1, *a*) and Ru/Be (Fig. 1, *b*) in the wavelength range 10.7–13.1 nm. The number of periods in multilayer systems is $N = 100$. The ratio of the thickness of the strongly absorbing layer to the period $\chi = 0.35$. Here and further on the left is a scale with reflection coefficients expressed as a percentage, on the right — spectral selectivity as a percentage.

As can be seen from Fig. 1, Ru/Be has large reflection coefficients and the width of the Bragg peak. The reflection coefficient reaches a maximum value of about 79% at a wavelength of 11.1 nm. Reflection coefficients drop sharply at wavelengths shorter than 11 nm from - to *K*-absorption edges Be.

In the wavelength range of 8.0–10.8 nm, Pd/Y and Sr/Rh systems are the most promising. Fig. 2 shows calculated graphs of the dependence of the reflection coefficient and spectral resolution on multilayer Pd/Y systems (Fig. 2, *a*) and Sr/Rh (Fig. 2, *b*) in the length range waves 7.9–10.9 nm. The number of periods in multilayer systems is $N = 130$. The ratio of the thickness of the strongly absorbing layer to the period in the system Pd/Y $\chi = 0.43$, in the system Sr/Rh $\chi = 0.35$. As can be seen from the figure, the Sr/Rh system exceeds the Pd/Y system in terms of reflection coefficient in almost the entire specified range, and in the vicinity of the wavelength 10.3 nm, the reflection coefficient reaches 76%. The spectral bandwidth is also quite wide, 3%.

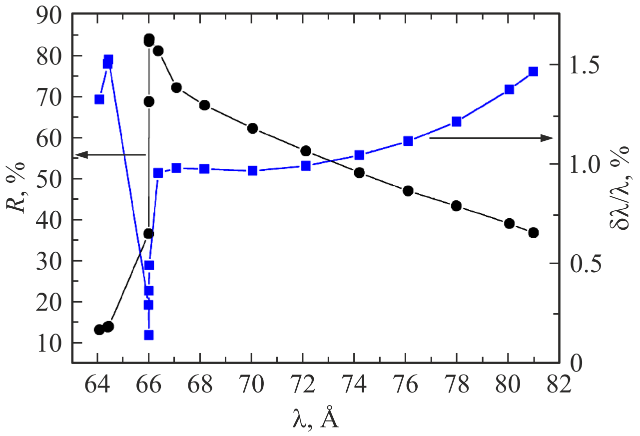

Figure 3. Calculated graphs of the dependence of the reflection coefficient (round symbols) and spectral resolution (square symbols) on the multilayer La/B system in the wavelength range 6.4–8.1 nm.

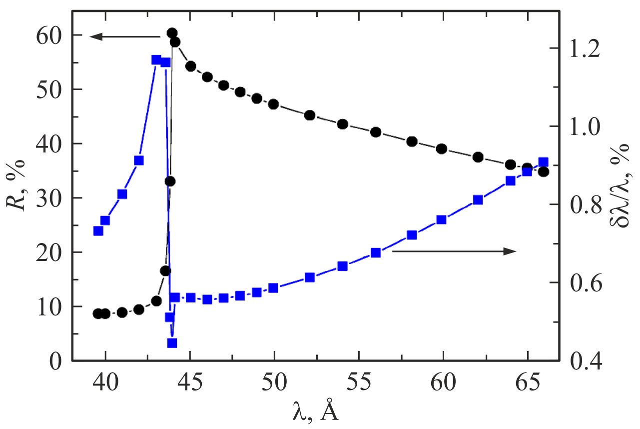

Figure 4. Calculated graphs of the dependence of the reflection coefficient (round symbols) and spectral resolution (square symbols) on a multilayer Co/C system in the wavelength range 3.8–6.6 nm.

In the wavelength range 6.6–8.0 nm, the La/B system has the highest reflection coefficient. Fig. 3 shows the calculated graphs of the dependence of the reflection coefficient and spectral resolution on the multilayer La/B system in the wavelength range 6.4–8.31 nm. The number of periods of the multilayer system $N = 200$, the ratio of the thickness of the strongly absorbing layer to the period $\chi = 0.46$. The maximum reflection coefficient is reached at a wavelength of 6.62 nm and is 84%. The sharp decline in the reflection coefficient after 6 nm is due to the *K*-edge of boron absorption.

In the wavelength range 4.4–6.5 nm, Co/Cr-based systems have the highest reflection coefficients. Fig. 4 shows the calculated graphs of the dependence of the reflection coefficient and spectral resolution on the multilayer Co/C system in the wavelength range 3.8–6.6 nm. The number of periods of the multilayer system $N = 300$, the ratio of the thickness of the strongly absorbing layer to the period $\chi = 0.28$. As in the previous case, there is a decrease in the reflection coefficient when moving away from the *K*-edge of the absorption of a weakly absorbing material, in this case carbon. The maximum reflection coefficient is reached at a wavelength of 4.4 nm and is 60%.

Fig. 5 shows the calculated graphs of the dependence of the reflection coefficient and spectral resolution on the multilayer Cr/Sc system in the wavelength range 2.8–4.1 nm. The number of periods of the multilayer system $N = 400$, the ratio of the thickness of the strongly absorbing layer to the period $\chi = 0.41$.

The table shows the name of the structure, the wavelength in nanometers corresponding to the maximum reflection, the reflection coefficient and spectral selectivity of the MXRM, as well as the reflection coefficients of 11 mirror X-ray optical systems, expressed as a percentage. As can be seen from the table, theoretically the maximum efficiency (reflectivity) is observed for La/B optics at wavelengths of 6.6 nm. Beryllium-based optics are in second place. The last column shows the spectral bandwidth of 11 mirror systems. This column indicates the optimal emission band of the undulator for the selected wavelength. If the bandwidth is narrower than the bandwidth of the undulator, then this means, accordingly, a decrease in the efficiency of such a system.

3. Experimental reflection coefficients

Despite the rather high theoretical reflection coefficients of multilayer mirrors in the considered wavelength range, the reflection coefficients obtained at the moment are often inferior to them. Interlayer roughness has the greatest negative effect on reflection coefficients. For example, at a wavelength of 3.12 nm, the theoretical reflection coefficient Cr/Sc MXRM is 63%, while in practice the record reflection coefficient is only 21–23% [15,16] and, due to the fact that

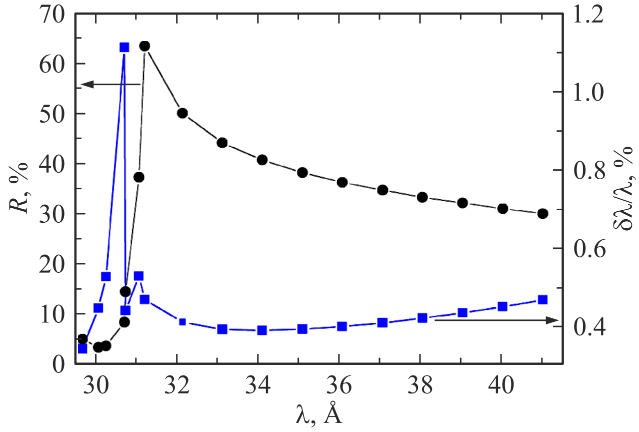

Figure 5. Calculated graphs of the dependence of the reflection coefficient (round symbols) and special resolution (square symbols) on a multilayer Co/C system in the wavelength range 2.8–4.1 nm.

X-ray optical characteristics of the most promising wavelengths and materials for next-generation lithography

MXRM	λ, nm	R, %	$\Delta\lambda$, nm	R^{11}, %	$\Delta\lambda^{11}$, nm
Mo/Be	11.31	76.14	0.29	4.99	0.1210
Ru/Be	11.43	78.66	0.27	7.13	0.2040
Pd/Y	9.60	62.28	0.23	0.55	0.1070
Sr/Rh	10.40	76.16	0.31	5.00	0.1501
La/B	6.60	83.92	0.03	14.54	0.0212
Co/C	4.40	60.48	0.02	0.40	0.0065
Cr/Sc	3.12	63.21	0.01	0.64	0.0060

the level of roughness already achieved is at the atomic level and is about 0.3 nm, there is no need to wait for drastic improvements.

Even worse is the real situation in the area of 4−6 nm. With theoretical reflection coefficients of Co/C MXRM about 60%, in practice about 17% [17] was obtained. In these mirrors, the interlayer roughness is about 0.4 nm. Even if it is possible to reduce the roughness to a record 0.3 nm, this will lead to an increase in the reflection coefficient to less than 30%.

The situation is much better at a wavelength of 6.6 nm. On mirrors La/B$_4$With and La/B with antidiffusion layers, reflection coefficients of 59−64% [5,6] were obtained. Since the interlayer roughness in these structures is at the level of 0.4−0.6 nm, there are prospects for further increase in reflection coefficients.

At a wavelength of 9 nm, theoretically, the reflection coefficient exceeds 60%, but in practice, the record reflection coefficient was 43% on the Rd/Y pair and in a fairly short time degraded to 34% [18]. A larger reflection coefficient, 56% (in neighborhood 11 nm), was obtained on the Ru/Y/B$_4$C structure in [19].

In the neighborhood of 11 nm beyond the absorption edge of Be, the Rh/Sr structure theoretically has the highest reflection coefficient, but experimentally on a close Mo/Sr system, the reflection coefficient was about 45% and degraded to almost zero within a few days [20]. Therefore, the reflection coefficient at the level of 56% obtained on the Ru/Y/B$_4$C system was a record for this area.

In recent work [21], the structure of Ru/Sr/B$_4$C was first reported. At a wavelength of 10.3nm, a reflection coefficient of 61% was obtained. The stability of the reflection coefficient during three months of observation was confirmed. Taking into account that this is the first result, and the optimization of layer thicknesses has not been carried out, higher reflection coefficients can be expected.

In front of the Be absorption edge at a wavelength of 11.2 nm, multilayer mirrors Mo/Be [22,23] have record reflection coefficients of 70.1−70.3%. Currently, a study of Ru/Be MXRM has been initiated, which theoretically have large reflection coefficients.

Conclusion

In this paper, the analysis of the capabilities of multilayer X-ray optics in terms of providing high reflection coefficients in the wavelength range of 3.1−11.4 nm is carried out. Both theoretical and experimental values of reflection coefficients are considered. On the short-wave side, the wavelength range is limited to a wavelength of 3.1 nm, which is due to the complete lack of prospects for obtaining reflection coefficients of the MXRM of normal line incidence greater than 20% at wavelengths shorter than 3.1 nm.

Theoretically, all the considered types of MXRM have a sufficiently high reflection efficiency — from 60 to 80%. As can be seen from the table, MXRM La/B, wavelength 6.6 nm have the greatest efficiency. They are followed by Ru/Be MXRM, $\lambda = 11.2$ nm. In our opinion, the range of 9−10.3 nm is promising for lithography, where reflection coefficients greater than 60% have been experimentally obtained on the Ru/Sr/B$_4$C structure. There are clear prospects for increasing the reflection coefficient.

Wavelengths shorter than 6.6 nm, in our opinion, are not of real interest for lithographic applications for two main reasons. First-of all, there is no prospect of achieving high reflection coefficients, even above 30%. Secondly,-the spectral reflection band 11- of the mirror system is very narrow, significantly narrower than the emission band of the undulator, which further reduces the efficiency of such systems.

The conducted research also made it possible to formulate requirements for the width of the emission band of the undulator for each of the considered wavelengths.

Funding

The study was carried out with the financial support of the Russian Federation represented by the Ministry of Science and Higher Education, agreement № 075-15-2021-1350 dated 5 October 2021. (internal number 15.SYN.21.0004).

Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] [Electronic source] Available at: https://www.asml.com/en/products/euv-lithography-systems/twinscan-nxe3400b

[2] A. Pirati, J. van Schoot, K. Troost, R. van Ballegoij, P. Krabbendam, J. Stoeldraijer, E. Loopstra, J. Benschop, J. Finders, H. Meiling, E. van Setten, N. Mika, J. Dredonx, U. Stamm, B. Kneer, B. Thuering, W. Kaiser, T. Heil, S. Migura. Proc. SPIE, **10143**, 101430G (2017). DOI: 10.1117/12.2261079

[3] N.N. Salashchenko, N.I. Chkhalo. Vestnik RAN, **78** (5), 450 (2008). (in Russian).

[4] S.S. Andreev, M.M. Barysheva, N.I. Chkhalo, S.A. Gusev, A.E. Pestov, V.N. Polkovnikov, N.N. Salashchenko, L.A. Shmaenok, Yu.A. Vainer, S.Yu. Zuev. Nucl. Instrum. Methods in Phys. Res. A, **603** (1−2), 80 (2009). DOI: 10.1016/j.nima.2008.12.165

[5] N.I. Chkhalo, S. Kunstner, V.N. Polkovnikov, N.N. Salashchenko, F. Schäfers, S.D. Starikov. Appl. Phys. Lett., **102**, 011602 (2013). DOI: 10.1063/1.4774298

[6] D.S. Kuznetsov, A.E. Yakshin, J.M. Sturm, R.W.E. van de Kruijs, E. Louis, F. Bijkerk. Opt. Lett., **40** (16), 3776 (2015). DOI: 10.1364/OL.40.003778

[7] S.S. Churilov, R.R. Kildiyarova, A.N. Ryabtsev, S.V. Sadovsky. Phys. Scr., **80** (4), 045303 (2009). DOI: 10.1088/0031-8949/80/04/045303

[8] T. Otsuka, D. Kilbane, J. White, T. Higashiguchi, N. Yugami, T. Yatagai, W. Jiang, A. Endo, P. Dunne, G. O’Sullivan. Appl. Phys. Lett., **97**, 111503 (2010). DOI: 10.1063/1.3490704

[9] N.I. Chkhalo, N.N. Salashchenko. AIP Adv., **3** (8), 082130 (2013). DOI: 10.1063/1.4820354

[10] B.L. Henke, P. Lee, T.J. Tanaka, R.L. Shimabukuro, B.K. Fujkawa. Atom. Nucl. Data Table, 27 (1982). DOI: 10.1016/0092-640X(82)90002-X

[11] B.L. Henke, E.M. Gullikson, J.C. Davis. Atom. Data and Nuclear Data Tables, **54** (2), 181−342 (1993). DOI: 10.1006/ADND.1993.1013

[12] A.V. Vinogradov, I.A. Brytov, F.Ya. Grudsky, M.T. Kogan, I.V. Kozhevnikov, V.A. Slemzin. *Zerkalnaya rentgenovskaya optika*, under the general ed. A.V. Vinogradov (Mashinostroyenie. Leningradskoye otd., L., 1989). (in Russian)

[13] A.V. Vinogradov, B.Ya. Zeldovich. Appl. Opt., **16** (1), 89 (1977). DOI: 10.1364/AO.16.000089

[14] V.N. Polkovnikov, N.N. Salashchenko, M.V. Svechnikov, N.I. Chkhalo. UFN, **190**, 92 (2020) (in Russian). DOI: 10.3367/UFNr.2019.05.038623

[15] C. Burcklen, S. de Rossi, E. Meltchakov, D. Dennetière, B. Capitanio, F. Polack, F. Delmotte. Opt. Lett., **42** (10), 1927 (2017). DOI: 10.1364/OL.42.001927

[16] V.N. Polkovnikov, S.A. Garakhin, D.S. Kvashennikov, I.V. Malyshev, N.N. Salashchenko, M.V. Svechnikov, R.M. Smertin, N.I. Chkhalo. ZhTF, **90** (11), 1893−1897 (2020). (in Russian). DOI: 10.21883/JTF.2020.11.49980.143-20

[17] I.A. Artyukov, Y. Bugayev, O.Yu. Devizenko, R.M. Feschenko, Y.S. Kasyanov, V.V. Kondratenko, S.A. Romanova, S.V. Saveliev, F. Schäfers, T. Feigl, Y.A. Uspenski, A.V. Vinogradov. Proc. SPIE, **5919**, 59190E (2005). DOI: 10.1117/12.620037

[18] D.L. Windt, E.M. Gullikson. Appl. Opt., **54** (18), 5850 (2015). DOI: 10.1364/AO.54.005850

[19] V.N. Polkovnikov, R.A. Shaposhnikov, N.I. Chkhalo, N.N. Salashchenko, N.A. Djuzhev, F.A. Pudonin, G.D. Demin. Bull. Lebedev Phys. Institute, **48** (12), 406 (2021). DOI: 10.3103/S1068335621120101

[20] B. Sae-Lao, C. Montcalm. Opt. Lett., **26** (7), 468 (2001). DOI: 10.1364/OL.26.000468

[21] V.N. Polkovnikov, R.A. Shaposhnikov, C.Yu. Zuev, M.V. Svechnikov, M.G. Sertsu, A. Sokolov, F. Schafers, N.I. Chkhalo. Materials of the XXIV Mezhdunar. simpozium „Nanofizika i nanoelektronika“, **1**, 585 (2022). (in Russian).

[22] C. Montcalm, S. Bajt, P. Mirkarimi, E. Spiller, F. Weber, J. Folta. SPIE, **3331**, 42 (1998). DOI: 10.1117/12.309600

[23] M.V. Svechnikov, N.I.Chkhalo, S.A. Gusev, A.N. Nechay, D.E. Pariev, A.E. Pestov, V.N. Polkovnikov, D.A. Tatarskiy, N.N. Salashchenko, Y.A. Vainer, M.V. Zorina, F. Schafers, M.G. Sertsu, A. Sokolov. Opt. Express, **26** (26), 33718 (2018). DOI: 10.1364/OE.26.033718