Quantum Electronics **43** (4) 388 – 391 (2013)

PACS numbers: 07.85.Fv; 52.38.Ph; 78.67.–n; 81.07.–b
DOI: 10.1070/QE2013v043n04ABEH015130

Freestanding film structures for laser plasma experiments

E.B. Kluenkov, A.Ya. Lopatin, V.I. Luchin, N.N. Salashchenko, N.N. Tsybin

Abstract. **The technique is developed for fabricating 5 – 500-nm-thick freestanding films of various materials and multilayer compositions. Apart from the traditional use in spectral filtration of soft X-ray and extreme ultraviolet radiation, the possibility of using the ultrathin films fabricated by this technique as targets in experiments on laser acceleration of ions is considered. A sample of the target in the form of a 5-nm-thick carbon film on a supporting net is fabricated.**

Keywords: *freestanding multilayer structure, thin-film laser target, EUV filter.*

1. Introduction

Thin metal films are widely used as transmitting optical elements in the instruments and systems operating in the extreme ultraviolet (EUV) and soft X-ray wavelength ranges. The choice of the thickness and composition is determined by the specificity of a particular application. Thus, in polarimetry of synchrotron radiation a multilayer thin film can serve as an efficient transmitting phase shifter in the classical ellipsometer scheme [1, 2]. In this case, the key parameter is the phase shift between the s- and p-polarised components of radiation, introduced by the film. This shift must provide reliable assessment of the polarisation characteristics, and the use of multilayer films of submicron thickness, composed of layers of two materials, alternating with rigorous periodicity (up to a few hundreds of periods), appears to be optimal. The composition and thickness of film filters, used in the satellite instrumentation for solar studies in the EUV range, are chosen to satisfy the requirement of blocking the visible radiation by 10^{-12} orders of magnitude [3]. The necessary suppression of the background light is achieved by using a pair of filters, installed in series at the input of the device and in front of the radiation detector. In spectral studies of laboratory sources the requirements to blocking properties of the filter are usually less severe, so that the used film structures have smaller total thickness (50 – 200 nm), providing small absorption in the EUV spectral range. The prospects of using freestanding thin-film elements in the devices of projection nanolithography with the operation wavelength 13 nm are a subject of active discussion [4]. For commercial systems of nanolithography, the pulsed laser-plasma sources of EUV radiation with a high mean power are developed [5], which imposes additional requirements to the size and thermal stability of film filters, suppressing the high-power background radiation of the source. Besides the spectral selection, the filter in the UEV lithography scheme can function as a protector of optical elements from contamination with the products of target erosion. The possibility of individual protection of the photomask from contamination using a closely spaced ultrathin film with the transmission coefficient $T = 85\%–90\%$ at $\lambda =$ 13 nm is presently under consideration [6, 7]. The application of thin-film structures as targets in the experiments on laser acceleration of ions is worth separate mentioning (see reviews [8, 9]). The thickness of the film target is one of the parameters that determine the ion acceleration efficiency. Depending on the irradiation regime (i.e., the intensity and wavelength of the femtosecond laser pulse), the optimal thickness of the target takes the values within the interval 5 – 200 nm [9, 10].

At the Institute for Physics of Microstructures of the Russian Academy of Sciences, a significant experience is accumulated in the field of fabrication and attestation of freestanding thin-film elements for the above applications. The transmitting phase shifters were fabricated on the basis of multilayer structures Cr/C, Cr/Sc, W/B_4C, efficiently operating in various regions of the spectral range 1.5 – 4.5 nm. In particular, it was demonstrated that the Cr/Sc structure provides the phase shift of 90° at the wavelength 3.1 nm, the transmission coefficient for each polarisation component being equal to 0.4%, i.e., this structure is analogous to a quarter-wave plate [11]. The EUV filters were developed using the Zr/Si and Al/Si structures with the thickness 200 nm and 300 nm, respectively, deposited on supporting nets. These filters are installed in the channels of the solar telescopes, operating at λ = 13.2, 17.1, and 30.4 nm (TESIS project) [12]. Considerable work is carried out on manufacturing multilayer film filters and investigating their properties for projection EUV lithography and laboratory plasma diagnostics. The application of the used techniques to fabrication of thin-film targets for laser ion acceleration experiments is in the process of approval.

2. Technology for fabricating freestanding film structures

The thin-film structure was formed using the method of magnetron deposition. Preliminary, a thin metal underlayer was deposited on the polished silicon substrate and then, layer by layer, the materials of the structure itself. The availability of

E.B. Kluenkov, A.Ya. Lopatin, V.I. Luchin, N.N. Salashchenko, N.N. Tsybin Institute for Physics of Microstructures, Russian Academy of Sciences, ul. Akademicheskaya 5, 607680 Afonino, Kstovskii district, Nizhnii Novgorod region, Russia;
e-mail: luchin@ipm.sci-nnov.ru

Received 24 December 2012
Kvantovaya Elektronika **43** (4) 388 – 391 (2013)
Translated by V.L. Derbov

systems having four or six magnetrons allows deposition in one technological cycle of both the underlayer and the multilayer structure that, if necessary, may consist of several different materials. The construction of the magnetron systems offers the possibility to obtain structures on substrates up to 200 mm in diameter with the uniformity of layer thickness up to the accuracy of 1 %. To obtain a freestanding film, the substrate with the deposited structure was placed on the surface of the selective solvent. After the dissolution of the underlayer, the vessel, in which the etching was performed, was filled with water and the film, floating on the water surface, was caught on a frame having a window of necessary size. The frame was oriented vertically, which helped to provide the distribution of forces, acting on the film from the liquid, optimal from the point of view of the final result. The higher mechanical strength of multilayer structures as compared with homogeneous films allowed avoiding thin film damage at the stage of etching and removal from the liquid.

3. Freestanding absorption filters for laser-plasma sources of EUV radiation

In the laser-plasma experiments the thin-film elements are mostly used as absorption filters, operating together with multilayer interference mirrors in high-aperture spectral instruments and EUV radiation recording channels. The necessity to use filters is due to two major reasons. First, the plasma intensely radiates beyond the EUV range and also scatters the incident laser light. Second, the multilayer mirrors usually incorporate metal coatings, the refection coefficients of which are high not only within the narrow interference maximum, but also in a wide spectral region, covering the UV, visible, and IR ranges. In each particular case they usually try to choose the thickness of filters, intended for EUV plasma diagnostics, with a certain reserve, based on *a priori* considerations about the form of the spectrum. Practically, the thickness may be considered as sufficient, if the signal from the radiation detector after installing the additional filter decreases proportionally to the transmission coefficient of the latter at the recorded wavelength. The choice of materials is determined by the required transparency band. In the spectral region near $\lambda = 13$ nm the transparent materials are Si and the metals Y, Zr, Nb, and Mo. We fabricated thin-film structures based on multilayer structures Zr/Si, Nb/Si, and Mo/Si with the transmission coefficients $T = 40\%-50\%$ at the wavelength 13 nm, and $T = 10^{-5}-10^{-6}$ at the wavelength 633 nm. They were used, e.g., in the EUV survey spectrometer of the laboratory laser-plasma source and in the photoresist testing scheme [13], as well as in the EUV plasma diagnostic channels in the experiment on laser proton acceleration [14].

To equip the high-aperture von Hamos spectrometer recording the radiation of laser-induced plasma in the range 3–4 nm, a multilayer Cr/Sc filter was fabricated [15], suppressing the radiation in the visible range by no less than six orders of magnitude. The technique of fabricating freestanding films was also applied to constructing the focusing mirror dispersion element of the spectrometer. The cylindrical mirror was implemented in the form of a multilayer structure, transferred onto the plane surface of a thin mica plate with subsequent bending (the bending radius 20 mm) [16, 17].

The freestanding multilayer filters Mo/C and Zr/Al were designed and fabricated for the experiments on recording the EUV radiation, generated when a laser pulse is reflected from a relativistic plasma mirror [18]. The Mo/C structures are characterised by sufficiently high (40%–50%) transmission coefficients in the band 6–13 nm and efficient suppression of extra-band radiation ($T = 5\times10^{-6}$ at $\lambda = 633$ nm). In the development of the Zr/Al filters the major attention was paid to providing the maximal transmission bandwidth. These filters can be used in any spectral region at $\lambda < 22$ nm, but they possess smaller transmission coefficients in the EUV range and less efficiently suppress the background radiation, e.g., at $\lambda = 633$ nm, $T \approx 10^{-4}$. The spectral characteristics in the EUV region for some multilayer films are presented in Fig. 1.

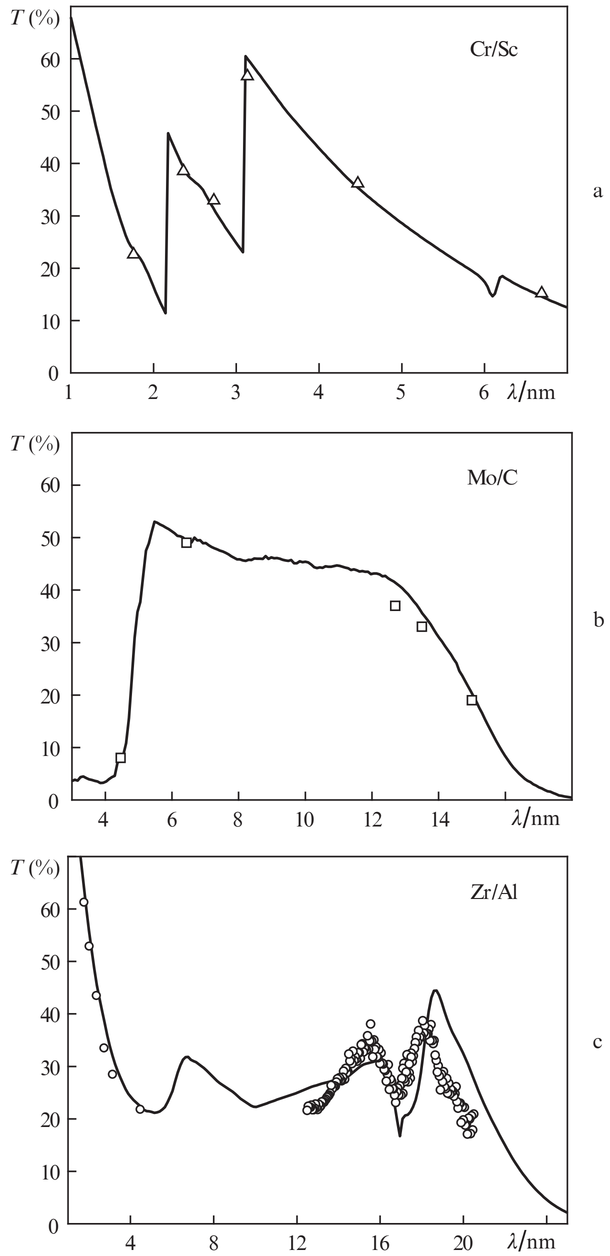

Figure 1. Transmission coefficients of some thin-film filters in the EUV region (solid curve – the results of calculation using optical constants, available at http://henke.lbl.gov/optical_constants, points – the results of measurements): (a) the Cr/Sc structure (the number of periods in the multilayer structure $N = 63$, the layer thickness of the structure materials, forming a period, $d_{Cr} = 1.5$ nm, $d_{Sc} = 1.6$ nm); (b) the Mo/C structure ($N = 60$, $d_{Mo} = 2.1$ nm, $d_C = 0.6$ nm); (c) the Z/Al structure ($N = 11$, $d_{Zr} = 5.1$ nm, $d_{Al} = 3.6$ nm).

For the goals of projection lithography, at the wavelength 13 nm the large-aperture Mo/$ZrSi_2$ filters with enhanced thermal stability and $T > 70\%$ were designed and fabricated [4]. The thickness of multilayer films amounted to 54 nm, their transmission coefficient in the UV, visible and near IR ranges did not exceed 2%. Since it is planned to install the Mo/$ZrSi_2$ filters in the lithography setup with laser-plasma EUV source on the basis of CO_2 lasers, the optical characteristics at the wavelength 10.6 μm are also important. The measured transmission coefficient of the samples at this wavelength amounted to 0.85% and the reflection coefficient 85%. The composition of filters was chosen following the results of comparative testing of a number of film structures made of materials with small absorption under the conditions of long-term vacuum heating [19]. It was shown that the chosen structure is able to stand the heating up to temperatures of 900–950 °C, corresponding to the released power per unit area up to 5 W cm^{-2}, during many hours without essential reduction of the transparency. The possibility of manufacturing a freestanding Mo/$ZrSi_2$ filter with the aperture 160 mm was demonstrated.

4. Prospects of applications of ultrathin film structures

In the most experiments with laser-plasma sources the thickness of the films used for filtering EUV radiation amounts to 100–200 nm. The developed technique allows production of ultrathin, sufficiently strong films with the thickness of a few tens of nanometres. We expect them to find application as elements of the EUV nanolithograph projection scheme (the spectral filter and the film, protecting the photomask from contamination) and as targets in the experiments on laser ion acceleration.

The value $T = 70\%–80\%$ is considered as acceptable for using the transmission element in a commercial nanolithograph. For the protective film, placed in front of the reflecting mask, the transparency requirements are more severe, $T = 85\%–90\%$, because of double pass of radiation (incident and reflected from the mask). In the EUV range such values are attainable with the film thickness of 20–30 nm. The diameter of the protective film has to be nearly 170 mm. As well as the spectral filter, the protective film as a part of nanolithograph must function for long time at a high temperature and under severe conditions of rarefied gas environment, containing hydrogen, residual oxygen, water vapours, radicals and various admixtures. The first results, obtained by means of the developed technique, are as follows: the samples of ultrathin freestanding silicon and multilayer protective films with the thickness 20–25 nm on the basis of the Mo/$ZrSi_2$ and Mo/$NbSi_2$ with $T = 84\%–90\%$ at 13.5 nm were fabricated [7]. The composition of the film structures includes the molybdenum silicide coating, inhibiting the oxidation process under the conditions of heavy thermal loads. The maximal aperture of the fabricated samples was 80 mm (Fig. 2). Despite the small thickness of the protective film, its spectral selectivity is preserved to a substantial degree. According to the results of measurements, the radiation at the wavelength 10.6 μm is suppressed by 10 times (per one pass), the UV radiation being blocked even more efficiently.

The considered technique of producing ultrathin films may appear useful in the search of optimal targets for various regimes of laser acceleration of ions. The necessity to optimise the thickness of the target was demonstrated, e.g., by the authors of [10], who observed the formation of homoenergetic beam of carbon C^{6+} ions with the energy 30 MeV in the process of interaction of the laser pulse (duration 45 fs, intensity 5×10^{19} W cm^{-2}) with ultrathin diamond-like films with the thickness $d = 2.9–40$ nm. The maximal energy of the ions was obtained at $d = 5.3$ nm.

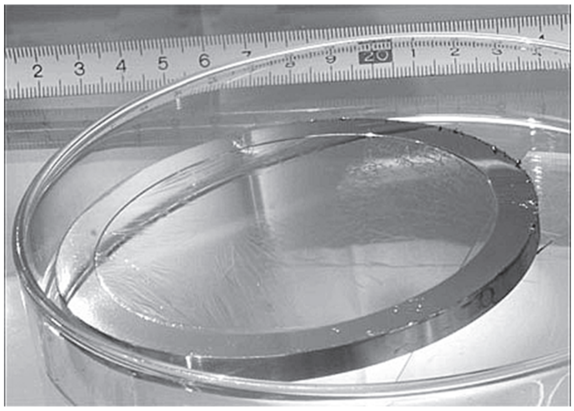

Figure 2. Freestanding thin Mo/$NbSi_2$ film having the diameter 80 mm – the prototype of a protective screen for the photomask in the scheme of projection EUV lithography; $T = 84\%$ at $\lambda = 13$ nm.

To demonstrate the capabilities of the developed technique, we fabricated a sample of carbon target in the form of a 5-nm-thick film deposited on a metal net with the total dimensions 33×38 mm, each mesh being a square with the side 1.7 mm (Fig. 3). The carbon film obtained by magnetron deposition, apparently, is worse in strength than the diamond-like foil. However, the proposed construction possesses the advantage that the area of the film damage under the action of a laser pulse can be limited within a single mesh of the net. We can use various materials and multilayer composition for fabricating the targets. In particular, it is easy to implement a double-layer target, comprising a very thin layer of light atoms and a thicker layer of heavy ones. The thickness of the heavy atoms layer, optimal for efficient acceleration of ions, depends on the intensity and wavelength of radiation. The layer of light atoms must be much thinner to provide the ion beam energy homogeneity [9]. As an example, demonstrating the possibility of formation of a double-layer target of light and heavy atoms, one can consider the freestanding film structure, consisting of 42-nm-thick Mo/Si base and the 20-nm-thick silicon layer. The multilayer structure provides

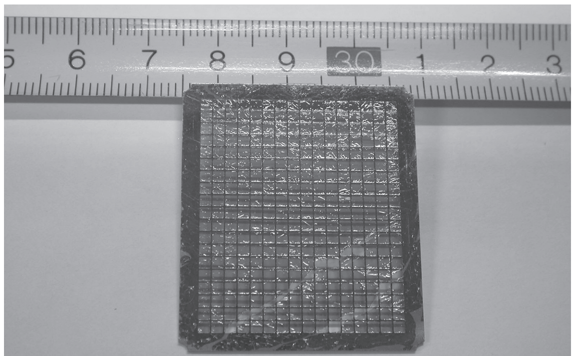

Figure 3. Sample target for laser ion acceleration experiments. The carbon film thickness is 5 nm.

the strength of the freestanding film, necessary for performing technological operation and further exploitation of the target. It is not difficult to form such a structure with a very thin layer of silicon or an element with smaller atomic mass.

5. Conclusions

The work on designing the sources and equipment for efficient instrumentation of projection EUV lithography stimulated the development of the technique for manufacturing freestanding thin films with the characteristics, hardly attainable in the immediate past. This fact not only expands the possibilities of traditional application of thin-film elements as absorption filters in the EUV range, but also forms the basis for new applications. The 20–25-nm-thick ultrathin films with the aperture above 100 mm will be tested as protective screens for photomasks in the projection lithography system. Yet thinner freestanding films, produced using the developed technology, both homogeneous and multilayer, are considered by us as promising targets for laser-plasma experiments, aimed at generating high-energy monochromatic ion beams. The sample of a monolayer thin-film target was demonstrated, implemented as the 5-nm-thick carbon film on the supporting net.

Acknowledgements. The work was supported by the Russian Foundation for Basic Research (Grant No. 10-02-01073-a) and the programme 'Extreme Light Fields and Their Applications' of the Presidium of the Russian Academy of Sciences.

References

1. Kortright J.B., Underword J.H. *Nucl. Instr. Meth. A*, **291**, 272 (1990).
2. Kortright J.B., Kimura H., Nikitin V., Mayama K., Yamamoto M., Yanagihara M. *Appl. Phys. Lett.*, **60**, 2963 (1992).
3. Mitrofanov A.V., Tokarchuk D.N. *Nucl. Instr. Meth. A*, **282**, 546 (1989).
4. Chkhalo N.I., Drozdov M.N., Kluenkov E.B., Lopatin A.Ya., Luchin V.I., Salashchenko N.N., Tsybin N.N., Sjmaenok L.A., Banine V.E., Yakunin A.M. *J. Micro/Nanolith. MEMS MOEMS*, **11**, 021115 (2012).
5. Banine V.Y., Koshelev K.N., Swinkels G. *J. Phys. D–Appl. Phys.*, **44**, 18 (2011).
6. Shroff Y.A., Leeson M., Yan P.Y., Gullikson E., Salmassi F. *J. Vac. Sci. Technol. B*, **28**, C6E36 (2010).
7. Salashchenko N., Luchin V., Lopatin A., Tsybin N., Chkhalo N., Kluenkov E., Drozdov M., Sjmaenok L., Belik V., Banine V., Yakunin A., Scaccabarozzi L., Diago P.R., Nikipelov A. https://www.sematech.org/10258/poster/P-OP-12.pdf.
8. Korzhimanov A.V., Gonoskov A.A., Khazanov E.A., Sergeev A.M. *Usp. Fiz. Nauk*, **181**, 19 (2011) [*Phys. Usp.*, **54**, 9 (2011)].
9. Daido H., Nishiuchi M., Pirozhkov A.S. *Rep. Prog. Phys.*, **75**, 056401 (2012).
10. Henig A., Steinke S., Schnürer M., Sokollik T., Hörlein R., Kiefer D., Jung D., Schreiber J., Hegelich B.M., Yan X.Q., Meyer-ter-Vehn J., Tajima T., Nickles P.V., Sandner W., Habs D. *Phys. Rev. Lett.*, **103**, 245003 (2009).
11. Andreev S.S., Bibishkin M.S., Kimura H., Kluenkov E.B., Lopatin A.Ya., Luchin V.I., Prokhorov K.A., Salashchenko N.N., Hirono T., Tsybin N.N., Chkhalo N.I. *Izv. Ross. Akad. Nauk, Ser. Fiz.*, **69**, 207 (2005) [*Bull. Russ. Acad. Sci., Phys.*, **69**, 226 (2005)].
12. Kuzin S.V., Shestov S.V., Pertsov A.A., Reva A.A., Zuyev S.Yu., Lopatin A.Ya., Luchin V.I., Zhou H., Huo T. *Poverkhvost*, (7), 19 (2008) [*J. Surface Investigation. X-ray, Synchrotron and Neutron Techniq.*, **2** (4), 527 (2008)].
13. Bulgakova S.A., Lopatin A.Ya., Luchin V.I., Mazanova L.M., Molodnyakov A.S., Salashchenko N.N. *Nucl. Instr. Meth. A*, **448**, 487 (2000).
14. Ragozin E.N., Pirozhkov A.S., Yogo A., Ma J., Ogura K., Orimo S., Sagisaka A., Mori M., Li Zh., Nishiuchi M., Daido H. *Rev. Sci. Instrum.*, **77**, 123302 (2006).
15. Borozdin Yu.E., Kazakov E.D., Luchin V.I., Salashchenko N.N., Tolstikhina I.Yu., Chernov V.V., Chkhalo N.I., Shevel'ko A.P., Yakushev O.F. *Pis'ma Zh. Eksp. Teor. Fiz.*, **87**, 33 (2008) [*JETP Lett.*, **87** (1), 27 (2008)].
16. Bibishkin M.S., Kazakov E.D., Luchin V.I., Salashchenko N.N., Chernov V.V., Chkhalo N.I., Shevel'ko A.P. *Kvantovaya Elektron.*, **38**, 169 (2008) [*Quantum Electron.*, **38**, 169 (2008)].
17. Lopatin A.Ya., Luchin V.I., Salashchenko N.N., Chkhalo N.I., Shevel'ko A.P., Yakushev O.F. *Zh. Tekh. Fiz.*, **80**, 105 (2010) [*Tech. Phys.*, **55**, 7 (2010)].
18. Pirozhkov A.S., Esirkepov T.Zh., Kando M., Fukuda Y., Ma J., Chen L-M., Daito I., Ogura K., Homma T., Hayashi Y., Kotaki H., Sagisaka A., Mori M., Koga J.K., Kawachi T., Daido H., Bulanov S.V., Kimura T., Kato Y., Tajima T. *J. Phys.: Conf. Ser.*, **112**, 042050 (2008).
19. Chkhalo N.I., Gusev S.A., Drozdov M.N., Kluenkov E.B., Lopatin A.Ya., Luchin V.I., Pestov A.E., Salashchenko N.N., Shmaenok L.A., Tsybin N.N. *Proc. SPIE Int. Soc. Opt. Eng.*, **7521**, 752105 (2010).