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大型 X 射线光学仪器:单层镜和多层镜的制造与表征

Michael Störmer[1], Dietrich Häußler[2], Wolfgang Jäger[2], Rüdiger Bormann[1]

(1. Institute of Materials Research, GKSS-Research Centre Geesthacht GmbH, Max-Planck-Strasse 1, 21502 Geesthacht, Germany; 2. Microanalysis of Materials, Faculty of Engineering, University Kiel, Kaiserstrasse 2, 24143 Kiel, Germany)

摘要:150～500 mm 长度的各种 X 射线光学元件可用于光束导引,光束调整,以及单色化。本文介绍了两种不同的大型 X 射线反射镜。第一种为单层反射镜,这种反射镜以 2°掠入射角在软 X 射线区(50～200 eV)起全反射镜作用,可用于自由电子激光器,如德国汉堡的 FLASH。第二种是多层镜,由于它的布喇格反射特性,适于作为反射镜以 0.4～1°的入射角用于硬 X 射线区(20～50 keV),如层析光束线的同步辐射存储环中。两种反射镜都用最新物理汽相淀积法制备,并用磁控溅射来实现 X 射线光学应用所需要的优良光学品质。这一淀积工艺使不同批次的镀膜稳定性良好,有利于实际反射镜在优质衬底上的最后淀积。单层镜和多层镜在它们的相关能量范围内都有很高的反射率,表面粗糙度也很低,且在整个光学波长区这些特性表现均匀。文中所叙相关研究都是借助 X 射线反射计量(XRR)法,透射电子显微镜(TEM)、光学轮廓仪(OP),以及原子力显微镜(AFM)完成的。

关　键　词:X 射线光学;多层膜反射镜;全反射镜;X 射线反射测量;磁控溅射;同步辐射;自由电子激光器

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Large X-ray optics: fabrication and characterization of single and multilayer mirrors

Michael Störmer[1], Dietrich Häußler[2], Wolfgang Jäger[2], Rüdiger Bormann[1]

(1. *Institute of Materials Research, GKSS-Research Centre Geesthacht GmbH*, *Max-Planck-Strasse* 1, 21502 *Geesthacht*, *Germany*; 2. *Microanalysis of Materials*, *Faculty of Engineering*, *University Kiel*, *Kaiserstrasse* 2, 24143 *Kiel*, *Germany*)

Abstract: Various X-ray optics with an optical length of 150～500 mm are employed for beam guidance, beam alignment and monochromatization. This paper focuses on two different types of large X-ray mirrors. The first optical element is a single-layer mirror which works as a total-reflection mirror in the soft X-ray range (50～200 eV) and at a grazing incidence angle of 2°. Such a mirror is used in free-electron lasers, *e.g.* FLASH in Hamburg, Germany. The second mirror is a multilayer mirror, which is employed as a reflector due to its Bragg reflection for the hard X-ray range (20～50 keV) and at incidence angles of 0.4～1°. Such a mirror could be used at a synchrotron storage ring, for instance in a tomography beamline. In both cases, the mirrors are fabricated by means of state-of-the-art physical vapour deposition techniques, using magnetron sputtering to achieve a good optical quality for

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their X-ray optical application. This deposition process allows good run-to-run stability, which is crucial for the final deposition of the actual mirror on a high-quality substrate. Both the single and the multilayer mirrors have a high reflectivity for their relevant energy range, a low roughness on their surfaces and a good uniformity of these properties over the whole optical wavelength. The investigations described here are performed by means of X-ray reflectometry (XRR), transmission electron microscopy (TEM), optical profilometry (OP) and atomic force microscopy (AFM).

Key words: X-ray optics; multilayer mirror; total-reflection mirror; X-ray Reflectometry (XRR); magnetron sputtering; synchrotron radiation; free-electron lasers

1 Introduction

X-ray optics are employed at beamlines of a synchrotron storage ring or of a free-electron laser for different purposes, such as beam guidance, beam alignment and monochromatization. The requirements of the X-ray optics are therefore defined by the X-ray source and the desired application at the beamline[1]. Here, the wide range of applications leads to many different optics, for instance crystals, Fresnel zone plates, single-layer mirrors and multilayer mirrors[2]. Currently, interest in the thin-film preparation of single-layer and multilayer mirrors is growing[3], mainly because they offer two advantages. Firstly, the substrate material can be different to the top-layer material, which is deposited later. The most widely used substrate materials are silicon or zerodur, which can be given the precise and accurate geometrical shape that is required. After manufacturing the substrates, they exhibit very low slope errors and roughnesses. These properties are essential for their X-ray optical performance. Secondly, the substrate is coated with the material that is necessary to provide the perfect solution for an X-ray optical application. This paper presents two thin-film X-ray optical device as examples: the single surface mirror and the multilayer mirror.

Total-reflection mirrors for the XUV range are used at the free-electron laser FLASH in Hamburg, Germany. More details can be found on the X-FEL web site[4]. These optical elements have to meet several requirements such as high reflectivity and very good radiation stability in the relevant energy range[5-7]. Additionally, the mirrors have to be about 500 mm long, since they need to work under grazing incidence. Therefore, the specifications of the shape and the surface of the Si substrates have to be very precise. Carbon was selected as a suitable coating material because of its absorption edge at 284 eV. These single-layer mirrors have been used under a grazing incidence angle of 2° and in the range of 50～200 eV for several years. No radiation damage has been found on the grazing-incidence mirrors used at FLASH; the mirror properties are under permanent investigation[8-9].

Multilayer mirrors in various dimensions are used at synchrotron beamlines worldwide. The beamline application therefore defines the requirements on the multilayer. The properties of a multilayer depend on its material combination, double-layer thickness and number of bilayers. For various reasons, the performance of a multilayer mirror can be lower than predicted by theory, for instance errors in thickness, contamination and poor interface quality decrease the optical performance. However, multilayer mirrors combine the important advantages of single-crystals and total-reflection mirrors, because they provide high spectral purity and high reflectivity.

2 Experiments

The single and multilayer mirrors were pro-

duced on well-polished silicon substrates by magnetron sputtering in an ultra-high vacuum chamber. Two sputtering sources were installed in the chamber, which could be equipped with DC, MF and RF generators. The base pressure was about 10^{-6} Pa before deposition. Pure argon gas (99.999 99%) was used as the sputtering gas. The gas pressure was between 0.05～0.4 Pa. Two tubes were fixed to opposite flanges of the chamber to move long substrates. Thus, a long deposition distance was achieved so that the movement of the 160～500 mm long substrate could begin and end outside the deposition area of the chamber. The steering of the substrate was computer-controlled, which ensured homogeneous deposition of a single layer or a multilayer. Initially, the calibration of the process was performed with a long aluminium bar, which is bigger in size than the final substrate that is to be used as an X-ray optic device. The bar was covered with several 60 mm long Si-wafer substrates. After the final calibration, the large single-crystal Si-substrate with a distinct surface geometry (plane, toroidal or cylindrical) was coated with the desired single or multilayer stack.

The coatings were investigated with X-ray reflectometry (XRR) using Cu-Kα radiation with a photon energy of about 8 048 eV (λ = 0.154 nm). A diffractometer (D8 from Bruker AXS) was equipped with a Göbel mirror, a reflectometry stage and a knife edge. The reflectivity measurements were analysed with the REFSIM simulation software (Bruker AXS) to determine the film thickness, roughness and density of the layer materials. Some XRR measurements were simulated by the IMD software[10]. The optical constants were taken from the Henke tables[11]. The IMD simulations allowed for variation in the thickness, roughness and density of each layer. Further investigations of roughness, morphology, microstructure and interfaces were performed by means of optical profilometry (OP; Micromap model 560), scanning force microscopy (SFM; Dimension 3 000, Nanoscope IIIa) and transmission electron microscopy (TEM; Philips CM30).

3 Results and discussion

Large total-reflection mirrors coated with amorphous carbon layers are needed for free-electron lasers. Fig. 1 shows four typical reflectivity measurements with Cu radiation to check the quality of the coating at different positions: 15 mm, 165 mm, 310 mm and 440 mm from the edge. Typically for single layer systems, Kiessig-oscillations are visible in each reflectometry scan. Between the total-reflection edge (at a critical angle of θ_{crit} = 0.22°) and an incidence angle of 2°, 20 maxima and minima are visible that are more or less located at the same angles. This demonstrates the good uniformity of the carbon coating and the high precision of the coating process over the whole deposition length. The coating thickness, roughness and density of a

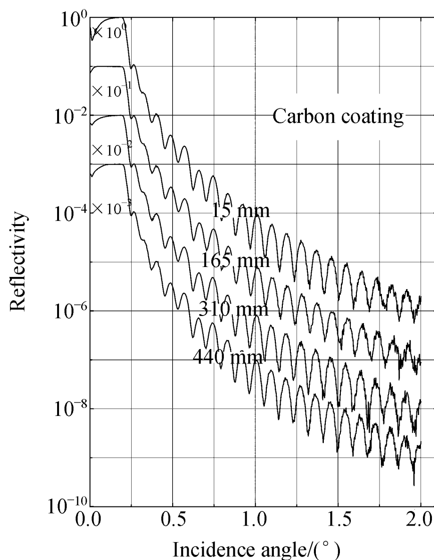


Fig. 1　Reflectivity measurements with Cu radiation of amorphous carbon coatings at various positions from the edge. (Results from IMD simulations: carbon coating: 47 nm thickness, 0.6 nm roughness and 2.2 g/cm^3 density.)

typical amorphous carbon layer can be determined by IMD simulations from each measurement. In this case, the mean thickness was approximately 47 nm, the roughness was 0.6 nm and the density was about 2.2 g/cm^3[5,7].

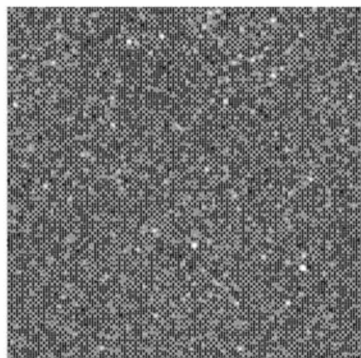
To investigate the changes in film thickness along the whole deposition length, the film thickness of another 500 mm long carbon coating was measured at 24 positions and intervals of about 20 mm (Fig. 2). A mean thickness of 45.4 nm was obtained with a standard deviation of about 0.3 nm and a peak-to-valley value of around 0.9 nm. Therefore, the variation in film thickness amounted to less than 2 % in relation to the mean thickness, which is an excellent result. Consequently, it can be stated that the deposition process itself does not have a significant detrimental effect on the slope error of the mirror. The latter is therefore still determined by the current technological limits of substrate manufacturing.

Fig. 2 Thicknesses of carbon films measured at different substrate distances from the edge. The mean thickness is about 45.4 nm. The peak-to-valley value is 0.9 nm.

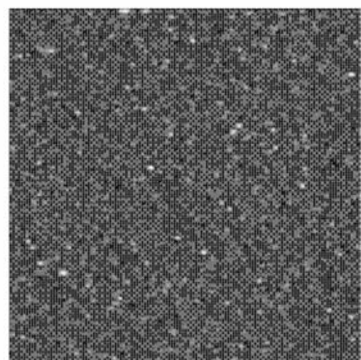
The reflectivity of a total-reflection mirror should be very high in the relevant X-ray range, so that, the selection of a suitable coating material is important; the roughness of the coating also plays an important role. To investigate the influence of the deposition process and to check the film quality, several surfaces were studied by optical phase-shifting interferometry before and after coating. Fig. 3 shows a typical measurement of a 45 nm thick carbon coating on a 300 mm long cylindrical silicon substrate. Here the measured area was 514 μm×520 μm and the micro-roughness was determined to be 0.27 nm. By means of scanning force microscopy, smaller areas (1 μm×1 μm) on top of the coating were examined to estimate the film roughness (Fig. 4). Along the whole deposition length, the mean roughness was virtually unchanged (0.4 nm rms). At each position on the 300 mm long X-ray optic, the roughness of the carbon coating was below 0.5 nm, confirmed with various measuring methods. This indicates that our deposition process for X-ray optics is very precise and the coatings are uniform in roughness over the whole deposition length of about 500 mm.

Fig. 3 Micro-roughness of a 300 mm long cylindrical substrate measured close to its centre, after deposition of an amorphous carbon film, area 514 μm×520 μm: The micro-roughness was below 0.5 nm at each abserved position.

Fig. 5 shows a 510 mm long silicon single-crystal substrate for the free-electron laser FLASH in Hamburg. The insert shows the same mirror after deposition. In the middle of the substrate surface, a strip of 45 nm thick carbon is visible. Such an FEL mirror provides an



(a) In the centre



(b) Close to the edge

Fig. 4　Roughness analysis of amorphous carbon films (on 60 mm long silicon substrates) employing the deposition process for 510 mm long substrates: (a) in the centre and (b) close to the edge with a microroughness of 0.37 nm rms and 0.43 nm rms respectively. The rms-roughness was below 0.5 nm at each observed position.

optical area of more than 490 mm×30 mm. Its shape is plane in the tangential direction with a slope error of below 1″ rms. This total-reflection mirror was fabricated to achieve a reflectivity as high as about 95% under a grazing incidence angle of approximately 2° in the relevant X-ray energy range of 50 to 200 eV. The measured reflectivity was 93%～95%, which is close to the theoretical limit of 95.6%[5].

The next challenge was to check whether it is possible to fabricate large multilayer mirrors for synchrotron applications. A prominent example is the material combination of tungsten

Fig. 5　Typical silicon single-crystal substrate for FEL application: plane mirror, T-mount, 500 mm long. The insert shows the same mirror after depostion. A strip of the 45 nm thick carbon coating is visible in the middle of the surface.

and silicon, which has often been used in the range of higher photon energies (*e. g.* 22～45 keV). In Fig. 6, four XRR measurements of a long W/Si multilayer mirror with 100 pairs and a *d*-spacing of about 3 nm are shown, which were investigated at different positions on the mirror (at 15, 135, 245 and 365 mm from the edge of a 400 mm long substrate). Each measurement shows three orders of Bragg-peaks, with the second order being smaller than the third order due to the equal single layer thickness of W and Si ($\Gamma \approx 0.5$). The patterns of the reflectivity curves are very similar, since the Bragg-peaks occur at almost the same incidence angle. A small deviation is detectable in the position of the third Bragg-peak which corresponds to a very small variation in thickness of the long multilayer mirror. The mean reflectivity was about 66% measured at Cu energy (8 048 eV), demonstrating the good uniformity and high precision of the multilayer deposition process over a 400 mm long deposition length. However, the theoretical reflectivity amounts to 77%, which can be attained with perfect interfaces, equal single-layer thickness, bulk values for density and no roughness. Therefore, the multilayer mirrors were investigated in more detail, to determine

which factors cause the decrease in reflectivity.

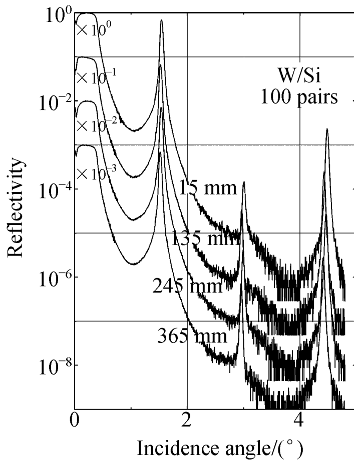


Fig. 6 Reflectivity measurements with Cu radiation of W/Si multilayer mirror at various substrate positions from the edge.

The double-layer spacing of a W/Si multilayer mirror was measured at 6 substrate positions (Fig. 7). The mean d-spacing was about 3 nm and the variation along the mirror length resulted in a peak-to-valley value of 0.06 nm. The percentage variation in multilayer period is therefore approximately 2% over a mirror length of 400 mm.

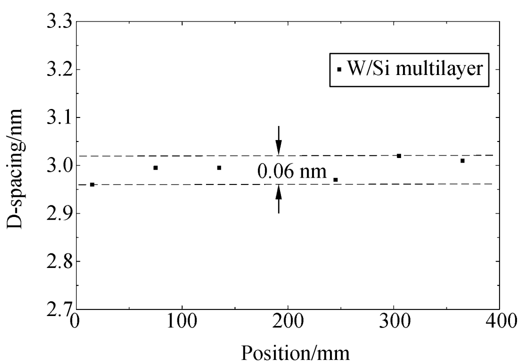


Fig. 7 Double-layer spacing of a W/Si multilayer mirror measured at various substrate distances from the edge. The mean d-spacing is about 2.99 nm. The peak-to-valley value is about 0.06 nm.

The roughness of the multilayer mirror was also investigated at various positions by means of AFM. Fig. 8 shows a typical topographic image of the surface of a multilayer mirror. The mean roughness of the coating was determined to be approximately 0.3 nm and the substrate roughness was nearly equal. Thus, it can be stated that the deposition process replicated the substrate roughness precisely. Such a low roughness is essential, since roughness generates non-specular X-ray scattering, that could reduce the X-ray reflectivity of the multilayer mirror.

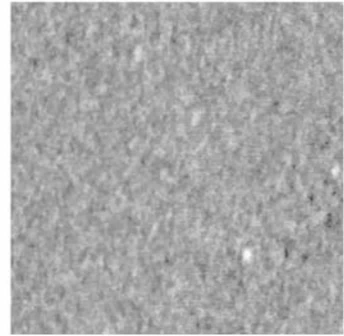


Fig. 8 Roughness measurement of a magnetron-sputtered multilayer mirror by means of AFM. The multilayer roughness was measured at various positions of the mirror. A mean roughness of less than 0.3 nm was measured at each position.

The performance of a multilayer mirror is also determined by the interface quality, since the X-ray reflectivity could be reduced by interdiffusion of the layer materials. Fig. 9 shows a bright field image of a magnetron-sputtered W/Si multilayer. A native silicon oxide layer is visible on top of the Si substrate. The dark layers are tungsten, and the bright layers are silicon. Between the layers, sharp interfaces are visible. The inserted diffraction pattern shows a series of smaller spots indicating the high quality of the multilayer. Even with higher resolution, no evidence was found that indicated formation of a compound at the interface.

Fig. 10 shows a synchrotron mirror, which was coated over a substrate length of 150 mm with two different multilayers. The upper multilayer contains 100 pairs of W/Si and the lower one 100 pairs of Ru/C with a d-spacing of 3 nm

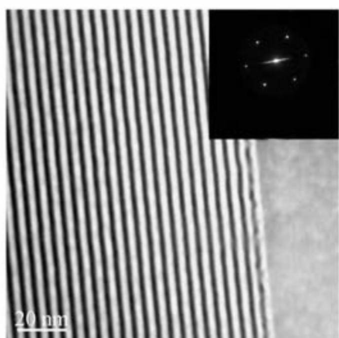


Fig. 9 TEM bright field image of a W/Si multilayer with a magnification of 160 k× (100 μm condenser aperture and 50 μm aperture). The tungsten layers appear dark, while the silicon layers appear bright, due to their different structure factors. The first layer on the silicon substrate is a native oxide. The inserted diffraction pattern demonstrates the high quality of the multilayer stack, as a series of smaller spots are visible representing the Fourier transform of the multilayer.

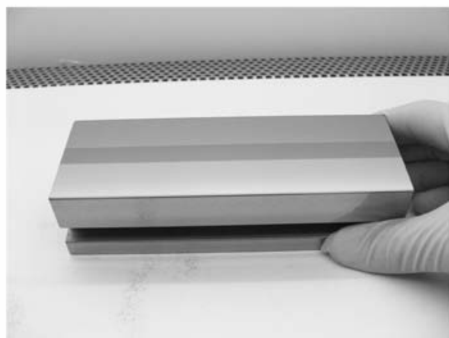


Fig. 10 Image of a synchrotron mirror with 2 multilayer strips: the upper multilayer contains 100 pairs of W/Si and the lower one 100 pairs of Ru/C with different *d*-spacings. With such a device it is possible to switch very quickly and efficiently between two distinct X-ray photon energy ranges. The first one is for higher energies between 22～45 keV and the other one is for lower energies between 10～22 keV.

and 4 nm, respectively. Such a synchrotron optic was fabricated for two different ranges: a high-energy range of 22～45 keV and a low-energy range of 10～22 keV.

4 Conclusions

Large X-ray optics have been fabricated for the soft and hard X-ray energy range. The film properties were investigated at several widely spaced positions on the mirror by XRR, SFM, optical profilometry and TEM.

Single-layer coatings can be used as total-reflection mirrors for beam alignment, beam guidance and monochromatization. In the beamlines of the free-electron laser FLASH at DESY in Hamburg, amorphous carbon films with a deposition length of 500 mm are installed due to the suitable absorption edge of carbon. At a grazing incidence angle of 2° and in the photon energy range of 50～200 eV, these single-layer mirrors provide a high reflectivity of about 94%, which is close to the theoretical limit. The uniformity in thickness over the whole length of about 500 mm is better than 2%, which means that the peak-to-valley value is 0.9 nm at a coating thickness of 45 nm. After coating, the carbon films exhibited micro-roughnesses below 0.5 nm over the whole length of the substrate.

Multilayer coatings can be used as reflective optics over a wide photon energy range. The multilayer mirror with a deposition length of 160 mm presented here is used in an X-ray tomographic microscopy beamline in the range of 22～45 keV. The multilayer stack consists of 100 pairs of tungsten and silicon layers with atomically sharp interfaces and without any compound formation at the interface. The mean *d*-spacing of the multilayer is approximately 2.99 nm with a peak-to-valley value of 0.06 nm over a deposition length of 400 mm. The mean reflectivity was 66% at Cu wavelength. The roughness of the coating was about 0.3 nm rms.

The investigation of the single- and multi-layered mirrors demonstrates that sputtering is a very successful technique to achieve large X-ray optics with an excellent uniformity in thickness, reflectivity and roughness over a deposition length of 500 mm. Additionally, the sputtering technique allows us to fabricate mirrors with high reflectivity and low roughness. Moreover, it is possible to control the thickness of the multilayer. In the near future, the variation of the double-layer thickness will be investigated for the multilayer mirrors and for the single layer mirrors, an increase of the deposition length will be a challenge.

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Authors' biographies: Michael Störmer received the diploma degree in physics and PhD degree in natural sciences from the University of Göttingen, Germany, in 1995 and 1998, respectively. At present he is a senior scientist at the GKSS Research Centre in Geesthacht, a member of the Helmholtz-Association (HGF). He is working at thin film and multilayer technology for the development of X-ray optics and of cor-

rosion protective coating. E-mail: michael. störmer@gkss. de

Dietrich Häußler received the diploma degree in physics from the University of Technology, Dresden, Germany, and the PhD degree in natural sciences from the University of Halle/Saale, Germany, in 1973 and 1982, respectively. At present, he is a senior scientist at the Faculty of Engineering of the University Kiel. He is working at microanalysis of materials especially by means of transmission electron microscopy. E-mail: dih@tf. uni-kiel. de

Wolfgang Jäger studied physics, performed his thesis research at the Max-Planck-Institute for Metal Research and received his PhD degree (Dr. rer. nat.) from the University of Stuttgart, Germany, in 1976, following his positions at the Argonne National Laboratory USA and at the Solid State Research Department of the Research Centre Jülich, Germany. He is at present professor and head of the Microanalysis of Materials Group at the Christian-Albrechts-University of Kiel, Germany. He is working at microstructure research of functional materials and at electron microscopy for materials science. E-mail: wj@tf. uni-kiel. de

Rüdiger Bormann received the diploma degree in physics and PhD degree in natural sciences from the University of Göttingen, Germany, in 1977 and 1979, respectively. At present he is professor of materials science at the Hamburg University of Technology and director at the Institute of Materials Research of the GKSS Research Centre in Geesthacht, a member of the Helmholtz-Association (HGF). He is working at light-weight structural materials, materials for hydrogen storage, nanotechnology and other advanced material technologies. E-mail: ruediger. bormann@gkss. de