Multilayer X-Ray Optics, Past and Future

Eberhard Spiller

Thin films with thicknesses in the nanometer range have been produced since thousands of years. B. Franklin (l757) described how a film of water could quiet the waves and it was trivial to calculate the film thickness. Langmuir and Blodgett used similar films to produce multilayers by dipping a slide multiple times into the fluids with a known film on top. They produced multilayers with periods Λ from 3-8 nm, and these were used as x-ray reflectors (Nobel 1932). Natural crystals served as the first x-ray reflectors or deflectors for x-rays (Laue, Ewald, Bragg, Darwin, 1912...) and a full 3-D theoretical treatment was developed. Enhanced transmission of x-rays was observed by Borrmann 1941 for the case that atoms in a crystal were located in the nodes of the standing wave field in the crystal. The theoretical description of that effect was probably known to Ewald around 1917.

Thin films for visible optics (Enhanced reflection or AR coatings) were only developed after 1930 and the Quarter Wave Stack was the standard design for good reflectors. Several papers stated that nothing could be achieved in the UV due to absorption. Utilizing the standing wave pattern in a structure to enhance or reduce the interaction with light was rediscovered by the author in 1970 and allowed to design useful coatings for the UV and x-ray region. These coatings are now used in astronomy, lithography, microscopy, spectroscopy covering the energy range from 20eV to 100keV and have been discussed at many conferences.

Dreams for the future: Enhance other desired properties of solids. Electron, phonons and other excitations have wave properties that might be modified by an optimized standing wave field. Even without a theoretical prediction we might discover the effect by monitoring the desired property during the growth of a film looking for oscillations. Some such oscillation have been observed for superconductors.

Expand our 2-D thin films to 3-D. The size of the atoms is the most severe limitations for further developments of ML films. If we could learn to control the position of each atom in a structure we could open up many new applications and create new materials. Transfer surface effects in a 3-D structure! Enhance performance of capacitors, batteries, memories.

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Multi-parameter Characterization of sub-nanometer Cr/Sc Multilayers based on Complementary Measurements

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Abstract

Cr/Sc multilayer systems can be used as near-normal incidence mirrors for the water window spectral range. We show that a detailed characterization of these multilayer systems with 400 bilayers of Cr and Sc each with individual layer thicknesses below < 1 nm is attainable by the combination of several analytic techniques. We used EUV- and X-ray reflectance measurements, resonant EUV reflectance across the Sc L-edge as well as X-ray standing wave fluorescence measurements. The parameters of our multilayer model were determined based on a particle swarm optimizer and validated using a Markov-chain Monte Carlo maximum likelihood approach. For the determination of the interface roughness diffuse scattering measurements were conducted.

Reconstruction of interfaces of periodic multilayer structures using model independent GIXR and XSW techniques.

PXRNMS , University of Twente, Enschede, 10-11 November 2016

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Development of the state-of-the-art periodic multilayer structures requires advanced interface engineering. Grazing Incidence X-ray Reflectivity (GIXR) is a widely used analysis method sensitive to the structure of interfaces in multilayers. However, conventional so-called model-based approaches of structural reconstruction from GIXRR data are lacking analytical power when dealing with multilayers where interface thicknesses are comparable to total thickness of multilayer period . We have developed the free-form approach that allows to analyze the GIXR data without the need for a priori assumptions on layer or interface parameters. The application of this approach will be demonstrated on the example of analysis of La/B multilayer structures[1].

To study the multilayers with inter-diffusion barriers with a higher precision an atomic-sensitive technique is often required. The ideal candidate is the X-Ray Standing Wave technique (XSW). The X-ray standing wave formed at the Bragg reflection condition in a periodic multilayer structure modulates atom-specific X-ray fluorescence. Previously[2] we have shown that atomic profiles can be recalculated directly from measured X-ray fluorescence yields modulated by XSW. This calculations require the knowledge of the optical constant profile obtained from GIXR analysis. We have demonstrated that, obtained by free-form analysis of GIXR, optical constant profile can be used as an input to calculation of atomic profiles from XSW data, what significantly simplifies the advanced characterization of the structure of periodic multilayers.

1. A. Zameshin, I. A. Makhotkin, et. al., Journal of Applied Crystallography **49** (4), 1300-1307 (2016).
2. S. N. Yakunin, I. A. Makhotkin, et. al., J. Appl. Phys. **115** (13), - (2014).

X-ray at-wavelength metrology of multi-layered surfaces

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Multilayer-coated mirrors are widely used as optical elements to handle the X-ray beam produced by sources such as synchrotrons, notably for spectroscopy and imaging applications. Their assets are a wide and tuneable spectral bandwidth transmission, an optical behaviour free from chromatic aberration, a high reflectivity efficiency and the possibility to use them at incidence angles larger than for mirrors operating at the total reflection angle, thus increasing the optical aperture and lowering the diffraction limit. On the other hand, their performances are limited by the surface shape errors and roughness.

While well-known methods such as reflectivity and diffuse scattering permit to assess statistical characteristics of the multi-layered material along the growth direction [1], the full topological description of the surface is only accessible at the cost of greater experimental efforts [2]. For X-ray imaging applications using coherent light such metrology information is of significant importance since the height errors in the mirror deposition process are responsible for unwanted large intensity modulation of the X-ray beam through an interference process.

The challenge in characterizing these defects lies in the fact that such error amplitudes can be as small as the light wavelength, i.e. in the order of a few nanometers for spatial periods in the millimeter range and above.

This presentation will introduce the methods available for measuring and analysing small spatial frequency aberrations in a beam upon reflection on a multilayer-coated mirror surface. The interest in measuring the wavefront of the X-ray reflected beam is to infer the surface optical defects and to map out the layer deposition errors.
Theoretical and experimental implementation of these characterization methods using hard X-ray light produced by a synchrotron source will be given, especially the ones based on interferometry and speckle.

This metrology will eventually permit to better understand the origin of the wavefront intensity modulation generated by multilayer mirrors and enable the development of multilayer manufacturing process strategies capable of minimizing surface defects and preserve a highly uniform X-ray wavefront.

[1] Kozhevnikov I. V., Peverini L., Ziegler E., “Development of a self-consistent free-form approach for studying the three-dimensional morphology of a thin film”, *Physical Review B*, **85**, p. 125439, (2012).
[2] Berujon S., Wang H., Alcock S., Sawhney K., “At-wavelength metrology of hard X-ray mirror using near field speckle”, *Optics Express*, **22**, pp. 6438-46, (2014).

Self-consistent optical-constant of materials for EUV multilayer coatings

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The preparation of multilayer coatings relies on the availability of precise optical constants of thin-film materials. When these are available, often they are not self-consistent, because they are obtained either on a narrow spectral range or by aggregation of data from different sources. Self-consistency requires that n and k be connected with the Kramers-Kronig (KK) relations.

At GOLD we have been obtaining self-consistent optical constants of several thin-film materials, mostly following two procedures:

a) Transmittance measurements versus energy in a broad spectral range for various film thicknesses. This provides k at each photon energy, and n is calculated with KK analysis.

b) Combinations of measurements including transmittance, reflectance, and ellipsometry in spectral intervals jointly covering a broad spectral range. Starting with these measurements, an iterative double Kramers-Kronig analysis procedure is followed: a first one on reflectance and its phase and a second one on k and n.

Measurements from the near infrared down to a wavelength of 30 nm (up to 41 eV) in the EUV can be performed at GOLD, as well as ellipsometry in the 190-950 nm range. To cover shorter wavelengths (larger energies), we have used BEAR-Elettra and 6.3.2-ALS synchrotron beamlines in collaboration with various groups in Italy and USA.

Self-consistency of n-k data generated with either procedure is tested with the use of two sum rules. Typically, the self-consistency of optical-constants is globally evaluated over the whole spectrum with these sum rules. We have generalized this evaluation to obtain also the local consistency of the optical constants at each photon energy range through a novel sum-rule method which includes window functions.

With the above procedures, various sets of materials have been characterized, including a long series of lanthanide and alkaline-earth metals, along with various carbides, fluorides, oxides, and boron. We will display examples of optical constants and self-consistency tests of interesting materials for multilayers in the EUV and adjacent ranges.

DFT simulations of surfaces, interfaces and multilayers

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I'll give a brief introduction into the underlying concepts of solid state modeling using periodic boundary conditions and then describe the strong and weak points of Density Functional Theory (DFT), and some routes to overcome its limitations. Then I'll discuss various spectroscopies which can be simulated using such quantum mechanical calculations and in particular using our WIEN2k code (www.wien2k.at) like XPS, XANES and EELS or UV/Vis spectroscopies using some specific examples. Finally I'll describe the reconstruction of a new surface model of the Fe_3O_4 (001) surface and a $SrTiO_3$/$LaVO_3$ multilayer system, which we propose as a possible photovoltaic material.

Beryllium-based multilayer mirrors for EUV spectral range

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The development of reflective optics is subject to continuous improvement of the optical properties of the multilayer mirrors MLM. To date, almost all of the MLMs for EUV range (10-40 nm) reached or almost reached the limit values of the reflection coefficients. Of course, this statement refers to the well-known and used in practice combinations of materials.

Improve the performance of structures through the use of some additional methods (barrier layers, ion polishing, ion assisting) is possible, but it can hardly give a substantial effect. It is, in particular, the increment of reflection coefficient for a few percent. Of course, even such a gain is very important for many applications.

Typically, the transition to a qualitatively new level occurs when using new materials. In this work as a basis for design effective MLMs are encouraged to apply beryllium.

There are about 10 published papers in the literature, which are devoted to Mo/Be, Rh/Be and Ti/Be MLM and where Be is considered only as spacer. Our theoretical calculations show that while the wavelength lies in range 11-17 nm Be is only a spacer, but in the 17-40 nm region, it may be considered as a scattering material with the little, “spacer-like” absorption. The combination of these physical properties results in unique X-ray optical properties; particularly theoretical Be/Al, Be/Mg and Be/Si MIS have a maximum reflectance of more than 70%, and a high spectral selectivity at the same time. Equally high reflection coefficients are achievable near the wavelength of 11.2 nm (MLMs Mo/Be, Ru/Be).

In this work we focus primarily on the performance of EUV multilayers based on beryllium (Be/Al and Be/Si/Al) for use in solar physics, specifically in high-spectral-resolution instruments employing normal-incidence mirrors.

Film deposition was carried out by DC magnetron sputtering. Deposition was carried out in an argon environment. The mirror reflectivity in the EUV range was measured using a reflectometer with grazing incidence spectrometer with a spherical grating RSM-500. Structure parameters (viz. period, individual layer thickness, density, transitional layer width) were determined from measurements on a diffractometer Philips X’Pert Pro at a wavelength $\lambda = 0.154$ nm.

As results, we note the following. Firstly, it is shown that, judging by the behavior of the optical constants in the wavelength range 17-40 nm, Be can be used as the scattering material rather than as a spacer. This conclusion was confirmed experimentally. We received a record reflectance (up to 60%) with spectral selectivity $\Delta\lambda_{mirr} = 0.4$ nm. Secondly, the efficiency of application Si interlayers on the boundary of Be-Si-Al (in the structure of Be/Si/Al), which have increased reflectance in the 17 nm to 43 to 60%, is shown. Taking into account the long-term stability of Be/Al multilayer structures, there weren’t changes in reflectance at a wavelength of 17.14 nm within 1 year after deposition, we can confidently expect that the structure of Be/Al can be competitive in the longer wavelength region.

This work was supported by grants from the Russian Foundation for Basic Research: 15-02-07753, in terms of the synthesis and study of samples at a wavelength of 30.4 nm RSF-DFG 16-42-01034.

Tungsten growth on silicon oxide and boron carbide and additional role of spacer in the ultrashort period multilayer X-ray mirrors

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Ultrashort period multilayer X-ray mirrors with the layer thickness down to 1 nm or less represent a technological challenge. Here, the layer thickness becomes comparable to the interface roughness inherited from the substrate which along with clustering effects may result in discontinuous layers producing strong diffuse scattering. Hence, the growth of refractory metal layers on various substrates is an important issue. We present here a study of W growth on SiO_2 and B_4C underlayers performed in real time by the in-situ grazing-incidence small-angle X-ray scattering (GISAXS). A microfocus X-ray source IµS equipped with focusing Montel optics (Incoatec) and 2D X-ray detector Pilatus 200K (Dectris) were mounted on a custom-designed dual-ion beam sputtering apparatus (Bestec). A tungsten 5 nm thick monolayer as well as a 15 period W/B_4C multilayer mirror with 1.8 nm period and 0.33 gamma ratio were sputter deposited at the base argon pressure of 2×10^{-8} mbar and working pressure of 4×10^{-4} mbar on a Si wafer covered with a $\approx$2 nm thick native oxide layer. Simultaneously, the GISAXS frames were collected repeatedly with the integration time of 8 s at the incidence angle of 0.25 degree close to the critical value to get a real-time movie of the process in the reciprocal space. The GISAXS measured close to the critical angle profits from a resonant enhancement of scattered intensity (Yoneda effect). The temporal study of the growing multilayer was performed by analysis of the lateral profiles (along the surface) of consecutive GISAXS frames extracted at the critical angle that provide the power spectral density (PSD) function. This follows from the fact that the lateral component of the scattering vector in the non-coplanar GISAXS geometry is not affected by refraction, hence, the kinematical theory of X-ray scattering is applicable. Two types of the PSD function were considered, namely those for the self-affine and mounded surfaces. The former PSD function typical for randomly rough surfaces decreases monotonously above a cut-off frequency while below it comes to saturation as the roughness cannot grow to infinity. The latter PSD is characteristic by a ridge controled by periodicity of "mounds" and is typical for cluster formation. Hence, by linear fitting in a proper interval of the PSD functions derived from GISAXS frames we were able to distinguish between the 2D (negative slope) and 3D (positive slope) growth. In particular, the growth of W/B_4C multilayer starting with W exhibits a gradual transition from the initial cluster formation to the 2D growth mode. The B_4C layers obviously favor the 2D growth and support such a regime also for W which finally prevails over the initial 3D growth of the first W layers. Contrarily, the GISAXS frames of the growing W monolayer show satellite maxima suggesting the mounded surface and cluster nucleation followed by cluster coalescence. These results indicate different growth modes of W on SiO_2 and B_4C underlayers but also demonstrate additional spacer role which is vitally important for ultrashort period multilayer X-ray mirrors, namely to allow formation of thin continuous refractory layers.

Normal- and grazing incidence mirrors for 6.x nm wavelength

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For future projection lithography applications, the wavelength of 6.x nm may offer improved imaging resolution. For normal incidence, high-reflectance multilayer reflective mirrors based on LaN/B have been successfully synthesized. The record reflectivity of 64% at 6.65 nm at near normal incidence was achieved [1].

For the off-normal (grazing) incidence, 30 nm LaN single films covered with B cap were synthesized, however, those mirrors demonstrated quick complete oxidation accompanied by complete destruction of the surface. Notably, single films based on the elemental La showed good performance and no oxidation. Qualitatively similar behavior has been observed for grazing-incidence LaN/B and La/B multilayers. The surface of the multilayers based on LaN again showed strong oxidation. As a solution we successfully applied a surface La nitridation instead of a full passivation of La layer. Both single films and multilayered structures showed high resistance of the structures against oxidation.

1. D. S. Kuznetsov, A. E. Yakshin, J. M. Sturm, R. W. E. van de Kruijs, E. Louis and F. Bijkerk, “High-reflectance La/B-based multilayer mirror for 6.x nm wavelength”, Optics Letters, Vol. 40, No. 60 (2015)

Neutron Multilayers

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Various types of optical devices based on multilayers have been developed in order to transport or change the properties of neutron beams: mirrors, monochromators, filters, guides, spin polarising mirrors or benders, focusing devices, antireflection/absorbing coatings... All of them are based on the tailored reflection or transmission properties of a surface coated with a multilayer. The most well-known and widespread application is the use of glass guides coated with supermirrors, which is now used extensively to transport neutron beams across large distances from neutron sources to instruments. Besides this, the design of optical devices has to be optimised for each neutron technique, depending on its requirements and instrumental constraints. The performance of the final device generally strongly depends on the precise characteristics of the multilayer, including exact layers thicknesses, roughness of interfaces and their correlations, magnetic structure and properties in the polarised case. This strong connection between multilayer exact properties and final device performance will be illustrated through selected examples, with particular focus on polarising multilayers.

The Hydrogenation Kinetics of a Magnesium Thin Film:

An in-situ Neutron-Reflection and Optical-Transmission Study of a Two-Phase System

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Nanostructuring is widely applied in both battery and hydrogen materials to improve the performance of these materials as energy carriers [1]. It changes the diffusion length as well as the thermodynamics of materials. We study the impact of nanostructuring on the (de-)hydrogenation in a model system consisting of a thin film of magnesium sandwiched between two titanium layers and capped with palladium. We verified optically the coexistence of the metallic α–MgD_x and the insulating β–MgD_{2-y} phase [2], and simultaneously studied the kinetics of the (de-)hydrogenation with (off)-specular neutron reflectometry. When the β–MgD_{2-y} domains exceed the neutron's coherence length, we are able to separately determine the volumetric expansion and the deuterium content of both phases. Our results show that there are significant deviations from the thermodynamic solubility limits in bulk magnesium during the phase transformation. This suggests that the kinetics of the phase transformation in nanostructured battery and hydrogen storage systems is enhanced not only as a result of the reduced length scale but also due to the increased solubility in the parent phases [3].

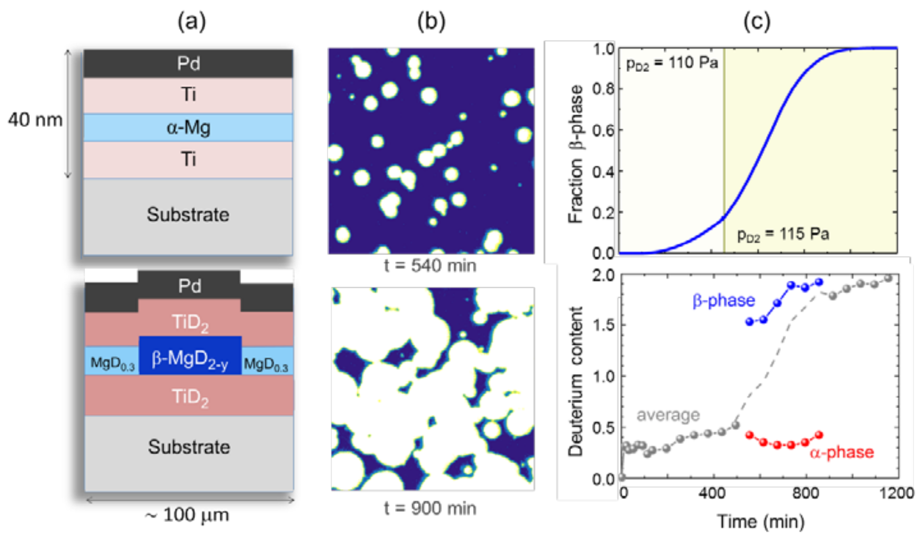

Figure 1 (a) The magnesium-based thin film system under investigation (top) in the virgin state and (bottom) during hydrogenation. (b) Top view optical transmission images at two moments in time. The β–MgD_{2-y} domains show up as bright spots that grow radially outwards. (c, top) Fraction of β– MgD_{2-y} as a function of time as determined from the optical transmission images. (c, bottom) deuterium content of the Mg layer, as determined by neutron reflection.

References

[1] Arico, A.S., Bruce, P, Scrosati, B, Tarascone, J.M., & Van Schalkwijk, W, *Nanostructured materials for advanced energy conversion and storage devices*, Nature Materials **4** (2005) 366-377

[2] Mooij, L. & Dam, B. *Nucleation and growth mechanisms of nano magnesium hydride from the hydrogen sorption kinetics*, PCCP **15** (2013) 11501–11510

[3] Bannenberg, L.J., Schreuders, H., van Eijck, L., Heringa, J.R., Steinke N.J., Dalgliesh R., Dam, B, Mulder, F.M., van Well, A.A., *Impact of nanostructuring on the phase behavior of insertion materials: The hydrogenation kinetics of a magnesium thin film*, J. Phys. Chem. C **120** (2016) 10185-10191

Study of the in-plane magnetic structure of neutron polarizing multilayer mirrors

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Neutron polarizing supermirror is one of the most important optical devices for spin-polarization of a neutron beam. Polarizing supermirror needs to display high polarization efficiency at low external fields to meet a variety of research demands. The magnetic properties of these multilayers with a polycrystalline grain size less than the ferromagnetic exchange length can be explained by the random anisotropy model, i.e. competition between the exchange interaction between neighboring spins and the local magnetocrystalline anisotropy [1]. This study is aimed to verify whether this model is valid in our system by observing the in-plane magnetic structure in the magnetization process. Off-specular scattering (OSS) and grazing-incidence small-angle scattering (GISAS) measurements with polarized neutrons are unique and powerful techniques to observe correlations of small magnetic objects in a layered system. The polarized neutron OSS and GISAS give access to the in-plane magnetic structure because the lateral correlation length obtained by the measurements corresponds to the size of an area with a uniform orientation of the magnetization. These complementary measurements, each of them covering different length scales, together with the data analysis based on the distorted wave Born approximation, revealed lateral correlation in the fluctuating orientation of the magnetization in the layer on a sub-μm length scale [2]. The obtained in-plane magnetic structure was consistent with the random anisotropy model.

[1] G. Herzer, J. Magn. Magn. Mater., 112, 258 (1992).
[2] R. Maruyama, T. Bigault, A.R. Wildes, C.D. Dewhurst, K. Soyama, and P. Courtois, Nucl. Instrum. Methods Phys. Res. A, 819, 37 (2016).

Kossel X-ray standing-waves within a Cr/B_4C/Sc multilayer excited by protons

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A characteristic X-ray line emitted from an atom within a periodic structure can be diffracted by the (emitting) structure itself according to the Bragg law. The subsequent Kossel [1] interferences lead to a modulation of the x-ray line intensity as a function of the detection angle in the vicinity of the Bragg angle value [2]. Standing-wave mechanism and Kossel diffraction can be viewed as space reversed processes by virtue of the reciprocity theorem. Kossel interferences have been yet observed using incident X-ray radiation [3-4], electrons [1-2, 5] and ions [6], in crystals [2,5-6] and in periodic multilayers [2-4].

In the present work, we have studied the characteristic Cr and Sc Kα emissions produced by a periodic Cr/B_4C/S multilayer exposed to a beam of 2 MeV-protons. The period of the multilayer is close to 2 nm. The intensity of these two emission lines is measured as function of the grazing exit angle, *i.e.* the angle between the direction of the detector and that of the surface of the multilayer. In the case of the Sc Kα emission, in Figure 1 we compare the experimental results to those calculated combining a classical recursive approach to the reciprocity theorem. To our knowledge, it is the first time that ions are used to induce Kossel diffraction in a multilayer. Refined details about the structure of the stack could be obtained, especially the profile and nature of the interfaces.

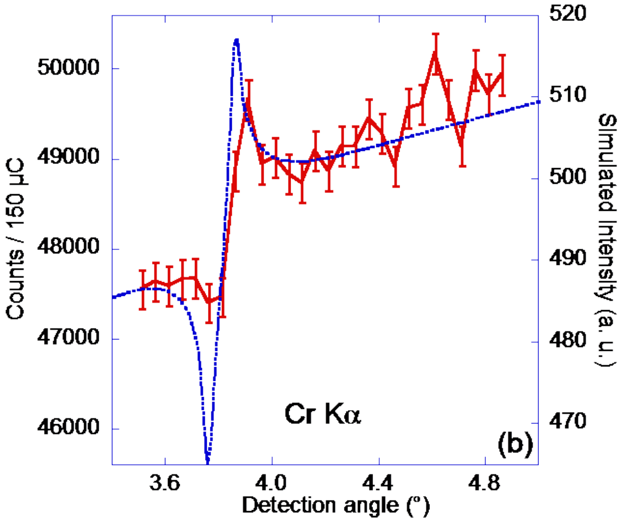

Figure 1: Measured (red) and calculated (blue) intensity of the Sc Kα emission as a function of the detection angle.

[1] W. Kossel, V. Loeck and H. Voges, Z. Für Phys. 94, 139 (1935).
[2] P. Jonnard P., J.-M. André, C. Bonnelle, F. Bridou and B. Pardo, Appl. Phys. Lett. 81 (8), 1524 (2002).
[3] J.-P. Chauvineau and F. Bridou, J. Phys. IV 6, C7 (1996).
[4] Y. Tu, Y. Yuan, K. Le Guen, J.-M. André, J. Zhu, Z. Wang, F. Bridou, A. Giglia and P. Jonnard, J. Synchrotron Radiat. 22, 1419 (2015).
[5] V.V. Lider, Crystallogr. Rep. 56, 169 (2011).
[6] V. Geist and R. Flagmeyer, Phys. Status Solidi A 26, K1 (1974).

High reflective water window collector optics

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The soft x-ray (SXR) region between the K-absorption edges of oxygen (λ_{O2} = 2.3 nm; E_{O2} = 539 eV) and carbon (λ_C = 4.4 nm; E_C = 283 eV) offers a good natural contrast between materials containing carbon and water. Furthermore, SXR allow extremely high resolutions of less than 30 nm by using water window microscopes [1].
Laser produced plasma (LPP) emission is an efficient way to produce SXR. This method allows a more cost-effective and flexible "table-top microscope" experimental set-up compared to synchrotron beamlines, which are very large and have a limited capacity. In order to collect as much SXR radiation as possible, near normal incidence collector mirrors with high reflective multilayer coatings are indispensable.

This paper shows the latest results of the multilayer optimization and coating of a LPP collector mirror with a diameter of 58 mm for the required wavelength of 2.478 nm. Different aspects of the optimization of the Cr/V based multilayer, e.g. use of barrier layers, coating parameters or its improved homogeneity over the whole collector surface will be discussed.
A detailed reflectance mapping measurement of the collector surface has been performed at the BESSY II beamline at PTB Berlin and will be shown.

Topics: water window microscopy, LPP collector, soft X-ray multilayer

Keywords: water window, soft X-ray collector optics, graded multilayer mirror

Preferred presentation: oral

[1] See, e.g., A. Sakdinawat and D. Attwood, Nature Phot. **4**, 840-848 (2010).

Current achievements in thin-film fabrication at HZG

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Advanced research light sources, such as free-electron lasers (FELs) and synchrotron sources, require ultra-precise optical elements such as X-ray mirrors consisting of single, double, triple and multiple layers. Current achievements in X-ray mirror fabrication will be shown to demonstrate possibilities and restrictions of thin-film preparation using magnetron sputtering. Currently, the most important coating materials are boron carbide, carbon, ruthenium and tungsten at HZG. [1-4] Thin-film specimens were investigated by X-ray reflectometry (XRR), X-ray diffraction (XRD), transmission electron microscopy (TEM), X-ray fluorescence spectrometry (XRF), atomic force microscopy (AFM) and optical interferometry in order to optimize the physical layer properties such as layer thickness, interface roughness, slope error, film morphology, microstructure, internal stress, thermal and radiation stability for distinct thin-film applications.

References

[1] M. Störmer, F. Siewert, and H. Sinn, Preparation and characterization of B4C coatings for advanced research light sources, J. Synchrotron Rad. 23, 50-58 (2016). DOI: http://dx.doi.org/10.1107/S1600577515020901

[2] I.V. Kozhevnikov, A.V. Buzmakov, F. Siewert, K. Tiedtke, M. Störmer, L. Samoylova, H. Sinn, Growth of nano-dots on the grazing-incidence mirror surface under FEL irradiation, J. Synchrotron Rad. 23, 78-90 (2016).

[3] A. Aquila, R. Sobierajski, C. Ozkan, V. Hájková, T. Burian, J. Chalupský, L. Juha, M. Störmer, S. Bajt, M. T. Klepka, P. Dluzewski, K. Morawiec, H. Ohashi, T. Koyama, K. Tono, Y. Inubushi, M. Yabashi, H. Sinn, T. Tschentscher, A. P. Mancuso, and J. Gaudin, Fluence thresholds for grazing incidence hard x-ray mirrors, Appl. Phys. Lett. 106, 241905 (2015). DOI: http://dx.doi.org/10.1063/1.4922380

[4] M. Störmer, H. Gabrisch, C. Horstmann, U. Heidorn, F. Hertlein, J. Wiesmann , F. Siewert A. Rack, Ultra-precision fabrication of 500 mm long and laterally graded Ru/C multilayer mirrors for X-ray light sources, Review of Scientific Instruments, 87, 051804 (2016). DOI: http://dx.doi.org/10.1063/1.4950748

Development of efficient and stable Al-based multilayer reflecting coatings for EUV range

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A further progress in scientific and industrial applications of multilayer optics in the extreme ultra-violet range (EUV) requires a high efficiency, specific spectral shape and good stability of multilayer coating as well as some new optical functions such as a control of phase and polarization of reflected EUV radiation.

Laboratoire Charles Fabry (LCF) has been involved in development of multilayer optics since more than 35 years (both theoretical and experimental studies). For instance, in the field of space applications, the EUV multilayer coatings for imaging telescopes of several solar missions were designed, fabricated and characterized. There were the telescopes of SOHO, STEREO and Solar Orbiter (to be launched in 2018 [1]). The latter is realized with Al/Mo/SiC multilayer coatings.

In the last PXRMS conference in 2010 we presented the first results of studies of multilayers made with the use of aluminum as a low absorbing material beyond 17 nm. A significant increase of the measured reflectance of periodic multilayers was obtained by applying a three-material design for the multilayer structure [2]. Here we will report on the progress achieved since then in the development and fabrication of Al-based multilayer coatings for EUV applications in the spectral range between 17 and 40 nm. We will discuss the design of various multilayer structures which is performed by using the IMD simulation software [3] and/or a home-made Matlab code with optimization algorithms. We will describe how complex multilayer systems can be designed and optimized for specific applications in the EUV range.

We will present and discuss the structural parameters and optical performance of periodic and aperiodic Al-based multilayer systems that have been designed and produced in our laboratory. Most of the multilayer mirrors were deposited by magnetron sputtering and characterized by grazing incidence x-ray (GIXR) and EUV reflectivity. The EUV reflectivity measurements were performed at the BEAR beamline of ELETTRA Sincrotrone Trieste and at the metrology beamline of Soleil synchrotron.

References

[1] F. Delmotte et al. *Proc. SPIE* **8862**, 88620A (2013)
[2] E. Meltchakov et al. *Appl. Phys. A* **98**, 111 (2010)
[3] D. Windt. Comput. Phys. **12**, 360 (1998)

EUV optics lifetime – radiation damage, contamination, and oxidation

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Creating high-reflectivity EUV (13.5 nm) multilayer mirrors is a significant challenge, requiring everything from optical design to atomic-level growth- and interface optimization. For application in EUV lithography an as-grown mirrors should not only have a high, ~70% initial reflectivity, but it should stay reflective over many years for economic viability. In this period the mirror is exposed to ionizing radiation and an ambient containing amongst others water, oxygen, and hydrocarbons. In this talk several lifetime-limiting effects are addressed.

First of all, direct radiation damage to the substrate and mirror materials is discussed. Upon EUV irradiation electronic bonds are broken by the absorption of high-energy photons. This can lead to photo-induced desorption of atoms or molecular groups, or to changes in material properties when the bonds 'heal' in a different configuration.

Practical EUV tools operate in imperfect vacuum conditions and the mirror surface thus is exposed to a flux of amongst others hydrocarbons, water, and oxygen. EUV irradiation causes cross-linking and dehydrogenation of adsorbed C_xH_y molecules, resulting in the possible build-up of a carbon contamination layer over time. When C_xH_y partial pressures are sufficiently low compared to O_2 and H_2O partial pressures, oxidation can occur. In this situation reactions with oxygen species produced by EUV-induced dissociation of e.g. H_2O are dominant. Oxidation, carbon growth and their mitigation will be discussed in the second half of the talk.

Design, fabrication, and test of extreme ultraviolet microscope with 30-nm spatial resolution

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Full-field microscopy in extreme ultraviolet (EUV) region has been investigated for various research fields, i.e., materials science and inspection of a lithography mask, where wide field of view would be essentially required for rapid observation. Recently, we proposed an innovative objective enhancing a magnification of 1500 [1-2], which is suitable for using a CCD camera as a detector. Besides, this novel design corrected for off-axis aberrations can be configured to have a large field of view with a diffraction-limited resolution, which allows us a rapid observation within a practical observation time.

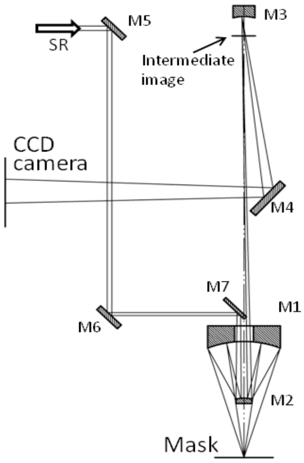

Fig. 1. A schematic of the EUV microscope: Three mirrors (M1-M3) act as a two-stage imaging system.

To demonstrate high spatial resolution of the objective, we are developing a full-field EUV microscope for at-wavelength observation of a lithography mask. As shown in Fig, 1, the optics was made of seven Mo/Si multilayer mirrors, which were optimized at a wavelength of 13.5 nm. The objective was configured as a two-stage imaging system. The Schwarzschild mirror having a numerical aperture of 0.25 projects a bright field image of the mask with a magnification of 30. The intermediate image is magnified again by the concave mirror M3, to have a high magnification of 1480 on the CCD camera. Spatial resolution was evaluated by observing fine line and space patterns with a half pitch between 30 to 80 nm, as shown in Fig. 2. We confirmed that 30-nm half pitch patterns were clearly resolved [3]. This result also indicates that the microscope has high spatial resolution near diffraction limit. In the presentation, we also describe wavefront sensing and control technique for the high magnification objective, which is essential for diffraction-limited spatial resolution.

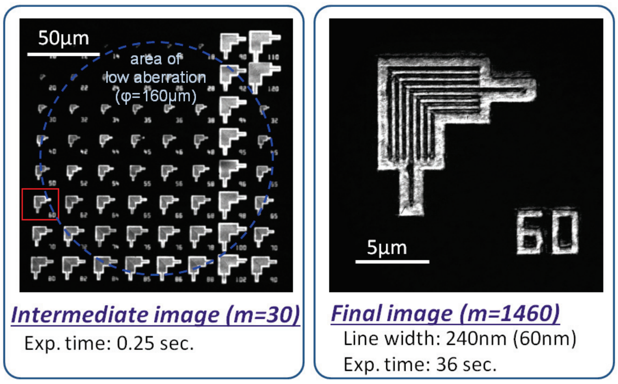

Fig. 2. At-wavelength images of EUV lithography mask.

References

[1] M. Toyoda et. al., Appl. Phys. Express **5**, 112501 (2012).

[2] M. Toyoda, Adv. Opt. Tech. **4**, 339 (2015).

[3] M. Toyoda et. al., Appl. Phys. Express **7**, 102502 (2014).

Hard X-ray multilayers with increased radiation resistance

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Because of its low interfacial roughness W/Si-based multilayers are often used as reflective coatings for hard X-ray mirrors. Adding carbon in W and Si layers makes the interfaces smoother and high reflectivity can be achieved even with ultra-short period WC/SiC multilayers. However, there is a lack of information on their thermal and radiation stability. We investigated the thermal stability of WC/SiC over a wide temperature range for different multilayer periods and compared it with W/Si and W/SiC multilayers. Our conclusions, which are based on measured changes in material microstructure, surface and interface roughness, multilayer period and stress show that WC/SiC system has a very high thermal stability. This has been demonstrated to be a prerequisite for high radiation resistance and makes this material pair an attractive candidate for applications in extreme condition environments.

Stability issues in Pd/B_4C multilayers

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ABSTRACT

Low d-spacing sputtered Pd/B_4C multilayers are attractive candidates for x-ray optical devices operating near photon energies of 20 keV. They form smooth interfaces and provide high reflectance. However, they turn out to be unstable under ambient conditions. They grow with excessive compressive stress and form defects. When exposed to oxygen, boron depletion and oxidation rapidly lead to a significant reduction of the d-spacing and to considerable damage of the layered structure. Several series of multilayers were studied using analytical tools such as x-ray reflectivity, in-situ stress monitoring, scanning electron microscopy, transmission electron microscopy, and energy dispersive x-ray spectroscopy. The sample properties were investigated over periods of time of up to two years. Various types of protection layers were added, which can slow down the degradation process by orders of magnitude.

Optimization and application of attosecond multilayers

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Abstract: The emerging field of attosecond physics aims for the temporal investigation and eventually the control of electron dynamics in matter (atoms, molecules or solids) on its genuine time scale, the temporal range of attoseconds (10^{-18} seconds). Recent experimental examples are the characterization of the photoionization time delays of core electrons in neon atoms [1] or the photoemission time delay between valence and core states of magnesium. Such experiments are based on the generation of attosecond pulses in the extreme ultraviolet to soft x-ray range by high harmonic generation in gases [2], which is nowadays a well established method and is constantly pushed toward ever higher photon energies by improved laser systems. It will eventually reach the important water window spectral range, between the 1s states of carbon and oxygen (284 eV–543 eV), with adequate photon numbers in the foreseen future. This will extend attosecond physics to biological specimens and may facilitate groundbreaking results in the field of biophysics. Besides the source development, there is a strong demand for corresponding optics development to steer and control attosecond pulses. Multilayer mirrors are key components in attosecond physics with a high degree of freedom, facilitating e.g. the tailoring of attosecond pulse parameters [3]. The designs, the accurate nano-fabrication, the characterization and application of periodic as well as aperiodic multilayer mirrors will be presented. The extremely precise control of the ion beam deposition process enabled for the first time the experimental control of the so-called attochirp [4] as well as the realization of the first chirped multilayer mirrors for the water window with a theoretically precise phase control, which can be used for a pulse compression down to 69 attoseconds above 300 eV [5]. The reflectivity performance of the multilayer mirrors was additionally improved by assisted ion beam polishing of the interfaces [6], resulting e.g. in a peak normal incidence reflectivity (21% at the scandium L3-edge) which is among the highest measured reflectivities of a pure chromium/scandium multilayer mirror. Besides the application of multilayer systems as reflective element a transmission multilayer quarter wave retarder has been designed, fabricated and characterized to transform linearly polarized attosecond pulses to elliptically polarized pulses [7], which could allow for the investigation of ferromagnetic materials or chiral molecules with a high temporal resolution. Various material systems have been investigated for multilayer systems [8,9] to allow for (attosecond) optics covering the spectral photon range from the lower extreme ultraviolet with few electronvolts up to the soft X-ray range with kiloelectronvolts. This allowed for example the realization of different multilayer mirrors and their application to spectrally select and shape radiation from a laser-plasma driven undulator table-top source, which resulted e.g. in the successful isolation and characterization of femtosecond pulses in the water window close to 300 eV, the highest measured photon energy from such a table-top source so far. Optimized multilayer mirrors have been applied to high harmonic radiation to access the attosecond time scale in the soft x-ray range by the generation and characterization of isolated attosecond pulses at 145 eV [10], up to now the highest central energy for characterized (by attosecond electron streaking) table-top attosecond pulses. The results demonstrate that the control of attosecond pulses by multilayer optics has been extended to higher photon energies (into the water window), to a better pulse dispersion control (for shorter pulses) and polarization control. This will pave the way for entirely new experiments in attosecond physics in the near future.

References:

[1] M. Schultze et al., Science **328**, 1658–1662 (2010).
[2] E. Goulielmakis et al., Science **320**, 1614 (2008).
[3] M. Hentschel et al., Nature **414**, 509–513 (2001).
[4] M. Hofstetter et al., Optics Express **19**, 1767–1776 (2011).
[5] A. Guggenmos et al., Optics Express **21**, 21728–21740 (2013).
[6] A. Guggenmos et al., Optics Express **22**, 26526–26536 (2014).
[7] J. Schmidt et al., Optics Express **23**, 33564–33578 (2015).
[8] M. Hofstetter et al., New Journal of Physics **13**, 063038 (2011).
[9] A. Guggenmos et al., Optics Letters **41**, 3714–3717 (2016).
[10] A. Guggenmos et al., Optics Letters **40**, 2846–2849 (2015).

Periodic multilayers and FEL radiation

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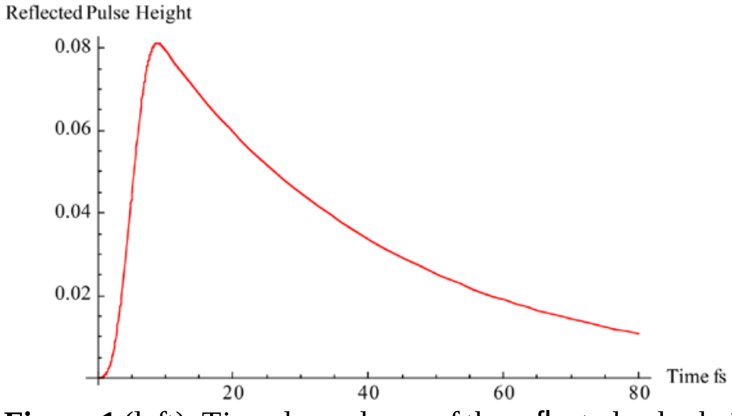
A few x-ray free electron laser (FEL) facilities are now in operation around the world. They produce very intense and very short pulses in the extreme ultra-violet, soft and hard x-ray ranges. However, FEL are not widely used to study the solid state and interaction with a periodic multilayer is generally only considered regarding the damaging effect. This is the aim of this communication to present simulations for periodic multilayers, designed as x-ray components, dealing with the short time and of the high intensity of a FEL pulse.

The time-dependence of the Bragg diffraction by a multilayer of nanometer period and its influence on the short pulse reflection are calculated in the framework of the coupled-wave theory. It appears that the indicial response presents a time-delay effect with a transient time conditioned by the extinction length. A numerical simulation is presented for a Bragg mirror in the x-ray range and a pulse of a sine-squared shape. In the presented example of a Fe/C multilayer illuminated by a 10 fs FEL pulse, Figure 1, the reflectance reaches the maximum value of 0.08 while in the steady state (continuous irradiation) it is 0.64.

We also propose to take advantage of the high intensity of the FEL pulses to make a distributed feedback laser. The idea is to implement a periodic multilayer acting as Bragg resonator acting both as the cavity necessary for the feedback and the active laser medium as one piece. The stimulated emission will be seeded by the spontaneous characteristic emission from one of the element of the periodic stack. We propose to use a Bragg resonator formed by a stack of Mg/SiC bilayers. The stimulated intensity of the Mg $L_{2,3}$ characteristic emission at 49 eV should be observed in a direction given by the Bragg angle, the angle satisfying the Bragg law for the Mg L emission and the considered period of the multilayer. We show, figure 2, based on an extended coupled-wave theory, that it would be possible to obtain a threshold gain compatible with the pumping provided by available X-FEL facilities.



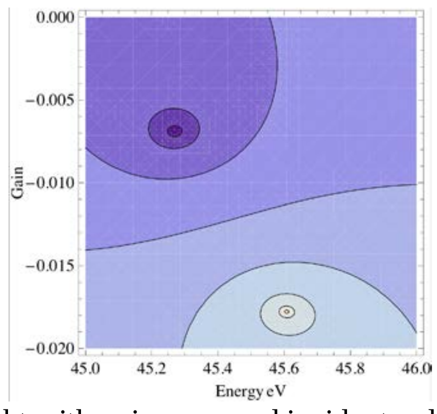


Figure 1 (left): Time dependence of the reflected pulse height with a sine-squared incident pulse of 8.05 keV energy and 10 fs width for the [Fe(2.5nm)/C(2.5nm)]x100 multilayer.

Figure 2 (right): Reflectance of a Mg/SiC multilayer as a function of the photon energy and the net gain in the Mg layers. A lasing condition is found near the first Bragg region at E = 45.61 eV with a net gain of 17.8×10^{-3}, that is a gain value of 78600 cm^{-1}.

Stress optimization of multilayer Laue lens coatings

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Abstract

The application of thin film coating processes for the fabrication of diffractive X-ray optical elements like sputtered-sliced zone plates or multilayer Laue lenses (MLL) is a very promising approach for X-ray focusing down to spot sizes of < 10 nm. However, for practical useful focal length in the order of millimeters or decimeters, multilayer thicknesses of several 10 µm up to a few 100 µm are necessary in order to have large enough numerical apertures of the optical systems.

Currently one of the main challenges is to coat low-stress multilayers with large total thickness in the order of 100 µm. Usually sputter deposition results in thin films with significant compressive stress [1,2]. With increasing thickness of the coatings the risk for delamination, micro cracks and geometric deformation steeply rises. In order to avoid these problems, a new silicon-based multilayer system has been developed that shows low stress with absolute values < 50 MPa. This has been obtained in a broad bilayer thickness range between 5 and 50 nm necessary for multilayer Laue lens designs with focal length of about 10 mm. Additionally, the thermal stability, the change of the internal stress with temperature and the microroughness have been characterized for these multilayers.

[1] C. Liu et al.: „Film stress studies and the multilayer Laue lens project“, Proc. SPIE 63170J-1 (2006)
[2] K. MacArthur et al.: „Periodic variation of stress in sputter deposited Si/WSi_2 multilayers“, Appl. Phys. Lett. 99, 081905 (2011)

Preferred type of presentation: Oral

X-ray nanometer focusing at the SSRF basing on multi-layer Laue lens

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Abstract:

There are many types of hard X-ray focusing devices utilizing reflection, refraction and diffraction optics. Zone plates have realized the highest resolution of 10-12 nm in the soft X-ray range [1]. Capability of focusing to 47 nm with a compound refractive lens has been experimentally demonstrated at wavelength of 0.06 nm [2]. A line focus with a width of 26 nm was achieved with a planar waveguide at photon energy of 13.3 keV [3]. Reflective mirrors have shown great progress recently with the advance of precision manufacturing techniques, and it breaks the 10 nm barrier in hard-X-ray focusing [4]. Diffractive optics is intrinsically well suited to achieve a high spatial resolution because a large numerical aperture (NA) can be achieved by diffraction. A novel approach to making diffractive optics with high NA is Multilayer-Laue-Lens (MLL) [5].

The MLL can be considered as a special type of zone plate and used in Laue geometry. It is fabricated by depositing the depth-graded multilayer inversely on a flat substrate then slicing and thinning the multilayer sample to an ideal cross-section depth. Compared with zone plates, MLL can reach a much larger aspect-ratio (cross section depth to the outermost layer thickness), which makes it capable of focusing hard X-rays with higher efficiency [6]. We designed and fabricated a multi-layer Laue lens (MLL) as a hard x-ray focusing device. WSi_2/Si and WSi_2/AlSi material combinations were chosen owing to their excellent optical properties and relatively sharp interface. The depth-graded multilayers were fabricated by using direct current (DC) magnetron sputtering technology. The stress was measured during the deposition, and stress comparison was made before and after Nitrogen reactive sputtering. After deposition, the thickness of each layer was determined by scanning electron microscopy (SEM) image analysis with the marking layer. Then, slicing, thinning and polishing processed were performed to make multilayer into MLL. The focusing property of the MLL was measured at the Shanghai Synchrotron Facility (SSRF). One-dimensional (1D) focusing resolutions of 92 nm are obtained at photon energy of 14 keV.

Keywords: multilayer Laue lens (MLL), hard X-ray, nano-focusing, synchrotron radiation, stress

Reference

1 C. G. Schroer, O. Kurapova, J. Patommel et al. Appl.Phys.Lett., 2005, 87: 124103

2 M. J. Zwanenburg, J. H. H. Bongaerts et.al. Physica B, 2000, 283: 285

3 H. Mimura, S. Handa, T. Kimura et al. Nat.Phys., 2010, 6: 122

4 H.F. Yan, H. C. Kang, R. Conley et al. X-ray Optics and Instrumentation, 2010, 2010: 401854

5 H. C. Kang, J. Maser, G. B. Stephenson et al. Physical Review Letters, 2006, 96: 127401

6 H. C. Kang, H.F Yan, P Winarski Robert et al. Appl.Phys.Lett., 2008, 92: 1

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Structural and reflective characteristics of Mo/Be multilayer with barrier layers

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Mo/Be multilayer structure (MLS) was actively studied in the late 90s, early 2000s in connection with work on the lithography in the wavelength range of 11.2-11.4 nm. In particular, [1] reported on achieved reflectance at a wavelength of 11.3 nm about 70.1% at normal incidence. The theoretical limit of the reflection coefficient for this pair of materials reaches about 76%. In succeeding years these studies almost not conducted perhaps because of the fact that the wavelength of 13.5 nm and a Mo/Si MLS have been chosen as the most promising for the lithography.

However, active researches in recent years to find a shorter wavelength for next generation lithography [2,3], as well as increased demands to the multilayer mirrors for solar studies in the spectral range of 11-13 nm [4] forced to turn again to this promising pair of materials .

Present work has the following objectives. The first goal is to find out the reasons why the experimentally obtained reflectance of Mo/Be MLSs is relatively low as compared with the theoretical limit. To solve this problem using methods of small-angle scattering of X-rays with a wavelength of 0.154 nm, precision laboratory and synchrotron radiation reflectometry in the spectral range of 11-13 nm, and atomic force microscopy we studied in detail the internal structure of Mo/Be MLSs. As the structural parameters we considered: the profile of the dielectric constant within the unit cell (period) of the MLS, the interfacial surface roughness (component of the transition layer), and the profile of the dielectric constant in the upper (outer) period. When reconstructing the dielectric constant profiles using the X-ray reflection data we used both a model and a model-free approach.

The second goal is the search for optimal materials and thicknesses of the barrier layers to reduce the interlayer transition areas. In this work, as the materials of the barrier layers Si and B_4C were studied.

This work was supported by the Russian and the German Science Foundations, grant RSF-DFG 16-42-01034.

1. C. Montcalm, S. Bajt, P. Mirkarimi, E. Spiller, F. Weber, J. Folta, SPIE 3331, 42-51 (1998).
2. Ch. Wagner, N. Harned, Nature Photon 4, 24 (2010).
3. N.I. Chkhalo, N.N. Salashchenko, AIP Advances, 3(8), 082130 (2013).
4. S. A. Bogachev, et al., Applied Optics, 55 (9), 2126-2135 (2016).

Complete characterisation of a multilayer coated reflection grating by atomic force microscopy (AFM), X-ray diffraction (XRD) and grazing incidence X-ray fluorescence analysis (GIXRF)

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In order to characterise the profile used in diffraction gratings none-destructively one usually refers to scans being made by use of atomic force microscopes (AFM). By this means, which can register only the surface topography, one will not obtain information about the buried interfaces and the substrate. In order to overcome this shortcomings, we thus subjected a reflection grating none-destructively to an X-ray beam in grazing incidence conditions in order to learn, which additional information can be obtained by combining angularly resolved Grazing Incidence X-ray fluorescence spectroscopy (GIXRF), X-ray reflectivity (XRR) and Grazing incidence X-ray diffraction. The layering in a thin film system can give rise to an X-ray standing wave perpendicular to the surface, by which a depth profiling of the constituents of the film system is possible.

A grating with laminar profile was coated with a single gold layer, which was partly overcoated with a 10 period W/C multilayer. The experiment was run at the X-ray fluorescence beamline at Elettra, where the manipulator in the vacuum diffractometer permits to vary the angle of grazing incidence very precisely, while the reflected signal and the energy-dispersed fluorescence from the layer system can be registered. In addition the grating could also be rotated around its pole, so that the plane of incidence can be oriented freely with respect to the grooves. In the most commonly used configuration this plane of incidence cuts through the grooves, while it can also be oriented parallel to the grooves. In the latter case a laminar grating becomes of system of tiny plane mirrors. In this case, when the trajectory of the probing beam is parallel to the grooves with rectangular profile, nevertheless diffraction will be observed as the structure is periodically disturbing the incident wavefield. Then the diffracted intensity will be found on a cone symmetrically oriented around the plane of incidence. And as one would intuitively expect the totally diffracted intensity should be identical to the reflectivity of a plane mirror with the same coating. And likewise one should not be able to distinguish the angularly resolved GIXRF from that of the plane mirror. This is infact observed and indicates thus the successful realization of the groove profile with the indicated shape.

When now the grating is rotated around its pole such that the beam trajectory is inclined with respect to the grooves, then the fluorescence intensity from buried layers should exhibit significantly different angularly dependent behaviour. This is observed and it will be shown that the now more structured angular dependence is in agreement with predictions employing geometrical ray-tracing calculations. X-ray diffraction data confirm the layering of the system as deduced from the previously described experiments.

Accurate computation of the X-ray diffraction efficiency of a multilayer coated grating based on a non-conformal deposition model

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In the 1-5 keV energy range where penetration depth can be significantly large, a multilayer stack deposited on a shallow modulated grating behaves as a double periodic medium and can provide enhanced Bragg reflectivity in proper tuning conditions[1,2]. For several years now, SOLEIL has undertaken to include multilayer coated grating in some of the beamline monochromators to extend the range of energies on soft X-ray or hard X-ray beamlines.
The latest realization was a Cr/B_4C multilayer grating for SOLEIL Sirius beamline. The 6.3 nm period multiyear stack was deposited at Laboratoire Charles Fabry on a 2400 l/mm grating with a 3.3 nm high lamellar profile ion-etched in Si by Horiba Company. The diffraction efficiency of this grating has been extensively measured with the goniometers available at SOLEIL, namely from 500 eV to 1500eV on the Metrology low-energy beamline branch and from 2000 eV to 4400 eV both on Metrology high energy branch and Sirius beamline itself with its crystal monochromator.
These measurements have shown a qualitative agreement with simulations produced with the CARPEM diffraction code[3] and a simple model assuming the conformal replication of a perfect rectangular profile throughout the layer stack and 3 discrete layers per period, the 3rd layer being introduced to account for the diffusion of B_4C in a an underlying Cr layer[4]. An arbitrary 0.8 reduction factor had to be applied to the calculated efficiencies to compensate the model imperfection but some features still could not be explained. The conformal model was also contradicted by AFM measurements of the grating surface which showed that the lamellar profile before coating was turned into an almost sinusoidal one after coating.
To overcome these issues a new grating model was developed and the CARPEM code was substantially modified to handle it. This new model is continuous and accounts for interfacial roughness and interdiffusion. It is also non-conformal. We consider that the local deposition rate is affected by surface tension and increased or reduced by a term proportional to the local curvature of the surface $\frac{\partial Z(x,t)}{\partial t} = v_0\left(1 + k\ \frac{\partial Z}{\partial x^2}\right)$. When the growth interface is under tension, k is positive and the high Fourier components of the surface modulation are damped proportionally to the square of their rank, turning the surface grating to sinusoidal. We show that this model explains all features of the measured data, the refraction shift of diffraction angles, the 55% maximum efficiency at 4100 eV, the reduced efficiency at low energies and even a glitch at 4400 eV which is not predicted by the conformal model. Moreover only the damping k parameter had to be fitted, the multilayer parameters, material densities, interfacial roughness and diffusion length being deduced from Cu K_α reflectivity measurements of a witness mirror coated with the same multilayer.

[1] F. Choueikani et al., Optics Letters **39**(7), 2141-2144 (2014)
[2] D. Voronov et al., Optics Express **24**(11) 11334-11344 (2016) and Optics Express **23**(4) 4771-4790 (2015)
[3] A. Mirone, et al., Appl. Optics **37**(25), 5816-5822 (1998)
[4] C. Burcklen et al., Journal of Applied Physics 119, 125307 (2016)

High Efficiency Multilayer coated Blazed Grating for tender X-rays

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The tender x-ray range between 1.5 keV and 2.5 keV has potential interest for X-ray microscopy and spectroscopy techniques. But till today it is a grey zone for both grating and crystal monochromator systems. In particular plane grating monochromators have low transmission and large amount of stray light in this range, while crystal monochromators suffer from high heat load in normal incidence operation. A multilayer coating optimized for a high line density grating significantly improves its efficiency and makes it feasible for use at high energies.

A blazed grating with 2000 lines/mm has been manufactured at HZB using a mechanical ruling engine in combination with ion etching technique to reduce the blaze angle to a value below 1°. For the resulting blaze angle of 0.84° a multilayer period of 7.3 nm was calculated, deposited and analyzed at the University of Twente (NL) in collaboration with Tongji University (Shanghai, China) [1]. In order to cover the specified large energy range chromium and carbon have been chosen since none of them possesses absorption edges in the energy range between 600 eV and 5900 eV. All measurements reported here were carried out between 600 eV and 1900 eV using the reflectometer at the Optics Beamline [2] and above 2 keV using the compact reflectometer attached to the X-ray beamline KMC-1 at BESSY-II.

An extraordinary efficiency of 35 % was measured at 2 keV while a maximum efficiency of 55 % was achieved at 4 keV. In addition, the multilayer blazed grating shows a very strong suppression of higher orders. This feature can become of significant importance for future beamlines. The reason is understood and will be explained in this contribution in detail.

Acknowledgments

The authors thank the MESA-Institute of the University of Twente (Netherlands) and the MOE-Institute of the Tongji University (China) for the calculation, deposition, and characterization of the multilayer.

References

1. F. Senf, F. Bijkerk, F. Eggenstein, G. Gwalt, Q. Huang, R. Kruijs, O. Kutz, S. Lemke, E. Louis, M. Mertin, I. Packe, I. Rudolph, F. Schäfers, F. Siewert, A. Sokolov, J. M. Sturm, Ch. Waberski, Z. Wang, J. Wolf, T. Zeschke, and A. Erko, Optics Express Vol. 24(12), , 13220-13230 (2016)).
2. F. Schäfers, P. Bischoff, F. Eggenstein, A. Erko, A. Gaupp, S. Künstner, M. Mast, J.-S. Schmidt, F. Senf, F. Siewert, A. Sokolov, Th. Zeschke, Journal of Synchrotron Radiation, Proc. of PhotonDiag workshop Trieste 23(1), 67-77 (2016).