Towards tender X-rays by means of multi-layer coated gratings as monochromator optics

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Abstract. Modern soft X-ray beamlines use collimated plane-grating monochromators (cPGM) [1] as monochromatization devices. Multilayer (ML)-coated plane gratings and mirrors allow to extend the available photon energy range of cPGMs towards the so-called tender X-ray photon energy range (up to 5keV), thereby achieving a significantly higher photon flux [2]. This X-ray energy range covers the L- and M-absorption edges of most transition and rare earth metals as well as the K-edges of lighter elements such as silicon, sulfur and phosphorus. Recently, such an ML-based monochromator setup was put into operation at the beamline U41-PGM1-XM at the storage ring BESSY-II in Berlin [3]. This beamline upgrade enables high-resolution spectromicroscopic applications with photon energies of up to 3keV and it is expanding its capabilities to support research in areas such as life sciences, semiconductor development and battery research. We report on the construction, commissioning and performance of this beamline and discuss possible options for new developments in the area of beamlines and end-stations in the tender X-ray energy range (up to 5keV) at existing and future new accelerator-based photon sources.

1. Introduction

Grating monochromators at synchrotron- or FEL-beamlines allow to cover the energy range from a few ten eV up to about 1800eV if reflection gratings under grazing incidence condition are applied [4, 5]. Such gratings are periodical patterned with a laminar or blazed groove profile. The aperture is usually covered by single material coatings like Au, Pt e.g. or amorphous carbon (a-C) the latter for lower energies [6]. To allow investigations at higher photon energies crystal-monochromators are the choice. However crystal monochromators lack sufficient physical stability at energies in the so-called tender-X-ray range between 1.5-5.0keV as their crystals are irradiated under normal incidence condition which causes significant heat load problems. Since long time it is under discussion to overcome such problems by means of multilayer coated (ML) gratings [7, 8, 9]. Related investigations have been started at several laboratories world-wide for laminar [10, 11] as well as for blazed gratings [12, 13, 14]. In our case we have chosen to demonstrate the potential of this concept at the existing U41-plane-grating-monochromator (PGM) beamline at the BESSY-II-storage ring in Berlin.

2. Beamline Design and Concept for the Tender X-Ray Energy Range

The U41-PGM-beamline at BESSY-II is a typical soft X-ray beamline following the concept of a collimated plane grating monochromator covering the energy range between 170 up to 1800eV. Since decades such beamlines are in use with great success at BESSY-II and other mid-energy storage rings [5]. The U41-beamline located at the low-beta section L06 of BESSY-II serves to a full field transmission soft-X-ray microscope (TXM) [3] (see Fig.1 for the beamline layout). The beamline allows both tomographic imaging of biological samples [15, 16] as well as NEXAFS studies for material science [17, 18]. The collimating toroidal M1-mirror and the intermediate

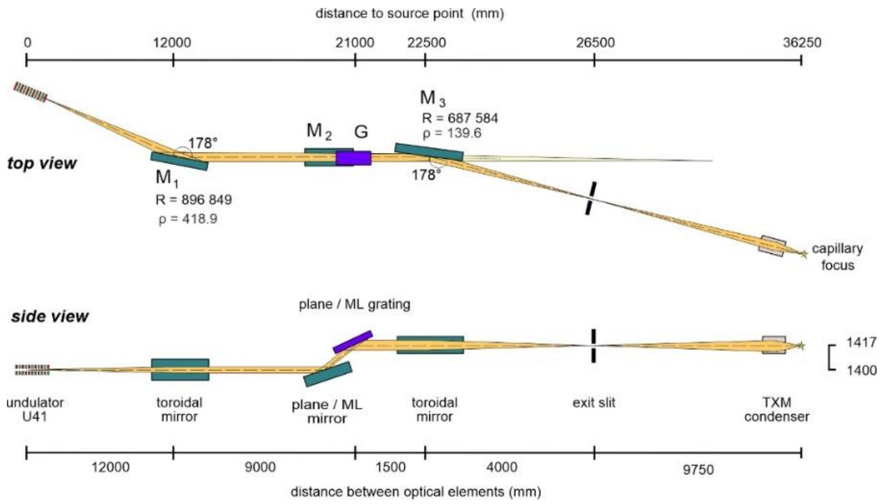

Figure 1. Schema of the optical design of the soft-X-ray transmission microscope at U41-PGM1-beamline at BESSY-II

focussing toroidal M3-mirror are in use under a shallow gracing angle of 1°. The optical parameters of the beamline are chosen to provide a round shaped illumination at the position of the rotational ellipsoidal re-focussing capillary condenser mirror of the TXM. Both toroidal mirrors have a coating of 30nm of Rh. Thus, a high photon-flux up to 3000eV can be provided if a high-efficient grating designed for higher energies is available. Initially the U41-monochromator had one blazed grating of 800l/mm (blaze angel: 0.93°) with a coating of Au manufactured in-house at HZB [19]. A free alignment slot at the monochromator was chosen to place a second, multilayer coated blazed grating at the monochromator. This allows to extend the achievable photon energy range beyond 1800eV with high efficiency. Furthermore, it was necessary to substitute the old plane mirror by a new one with two apertures. One with an Au-coating to enable the beamline serving for the energy range between 170-1800eV like before and a second aperture with a ML-coating of identical composition to that on the new grating. Table 1 gives an overview

Table 1. Optical elements at beamline U41, their parameter and quality.

Optical element	Aperture size	Radius[a]	Slope error[a]	Micro-roughness[a]	Coating	Line density	Blaze angle
Toroidal mirror M1	320x20mm^2	905274mm	890nrad	0.24nm	Rh		
Plane mirror M2	300x20mm^2	>100km	115nrad	0.18nm	Au and Cr/C		
Plane grating	92x12mm^2	>100km	200nrad	0.15nm	Au	800l/mm	0.93°
ML-grating	92x12mm^2	>100km	185nrad	0.16nm	Cr/C	2000l/mm	0.59°
Toroidal Mirror M3	190x20mm^2	691814mm	950nrad	0.7nm	Rh		

[a] all values have been measured at the BESSY-II Optics Lab by means of NOM [20], Fizeau-, White Light - Interferometer and AFM. All values for slope error and micro-roughness are rms values.

on the beamline optical elements, their parameter and quality.

3. Multilayer Coated Grating and Plane Mirror

Previous investigations have shown ML-coated gratings allow to provide high photon-flux up to 5keV and beyond [2, 9, 14, 21]. Our investigations at HZB have been mainly focused on ML-coatings based on the Cr/C-material combination showing excellent results [14] with Cr as absorber and C as spacer. Experimentally it was shown this material combination allows to reach an efficiency of up to 60% in the 1st diffraction order for high line density gratings [14]. Thus we`ve decided to develop a plane pre-mirror/grating set-up for the monochromator based on this material combination. Like the first 800l/mm grating the new grating was manufactured in house by the HZB department of Precision Gratings by means of mechanical ruling [19, 22]. A blazed grating of 2000l/mm (blaze angle: 0.59°, anti-blaze: 1.57°) was manufactured. After ruling a blazed grating profile of 2000l/mm with a blaze angle of about 6° into a layer of Au the final blaze-profile was achieved by ion beam etching which allows a transfer of the grating profile into the silicon bulk of a super polished grating blank. This process enables to achieve small blaze angles as well as a very low value for the micro-roughness of ≤0.2nm rms on the blaze facet. The initial ruling-layer of Au is completely lost after this process step and the final blaze patterning is completely transferred into the super-polished silicon substrate which gives a high mechanical stability. Next the ML-coating was deposited on top of the blazed grating pattern. The knowledge of the final blaze- and anti-blaze angle after ion beam etching is essential for the final ML-parameter design. Note: there is always an uncertainty for the blaze angle achieved after ion beam etching as this process is of limited repetitious accuracy. Both blaze angle and micro-roughness have been inspected by means of scanning force microscopy (AFM) measurements. The ML-coating parameters were defined based on the AFM measurements for the blaze angle. Following the grating equation for reflective gratings and the Bragg condition and taking additional refraction effects at boundaries into account than this is covered by the following equation [14] for the d-spacing d_{ML} of the ML:

$$d_{ML} = \frac{n}{m} d_{GR} \sin\gamma \frac{1}{\sqrt{1-\frac{8\delta}{\lambda^2 m^2}(d_{GR}\sin\gamma)^2}} \qquad (1)$$

with wavelength λ, grating period d_{GR}, blaze angle γ, the design diffraction order of the grating m and the multilayer n and the average refraction coefficient decrement of the ML-material combination δ. This gives in our case an optimum value of 5.5nm for the d-spacing. However further studies showed that a d-spacing of 5.9nm would give a higher transmission of the ML [21], see also [23]. Thus, the ML-coating design parameters were set as follows: d_{ML} 5.9nm±0.1nm with a gamma-ratio (Cr/total) of 0.4. The number of bi-layers is of 30 which is still below efficiency saturation but allows to minimize the risk of grating profile smearing and loosing efficiency due to this. A binding-layer of 7nm Cr between substrate and ML was chosen and a top-layer of 2nm Carbon to avoid Cr-layer oxidation. The ML-coating was processed by magnetron sputtering at the Tongji University (Shanghai) [24]. Both the grating and the plane mirror have been processed simultaneously to enable an identical composition of the coating which is essential to meet the Bragg condition. Figure 2 shows both plane mirror and grating after finishing of the coating process.

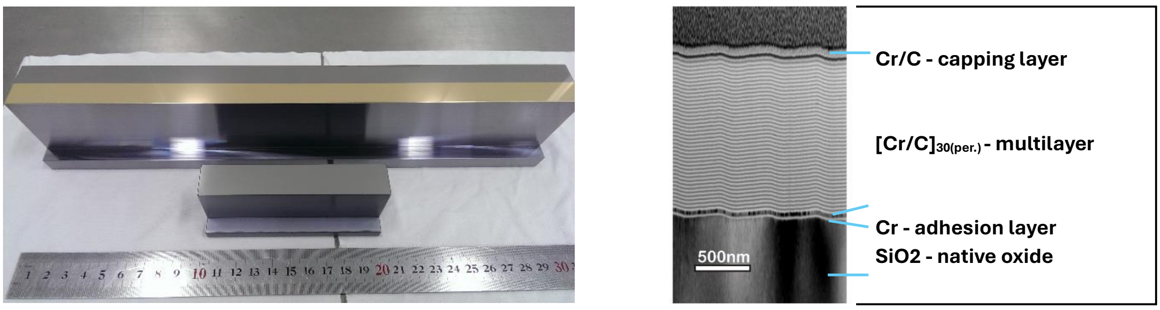

Figure 2. Left: At top the plane mirror with Cr/C-ML-coating and Au-coating, bottom the plane grating with the Cr/C-ML-coating. Right: SEM image of the cross section of a Cr/C-multilayer taken from a prototype grating sample identical to the final grating except for the capping layer.

4. Commissioning and Beamline Performance

In a final step we have conducted a beamline commissioning at the U41-beamline to verify the feasibility of the here discussed idea. We have checked the photon-flux over a large energy range between about 300eV up to 3500eV in comparison to the installed 800l/mm Au-coated grating as well as to simulations for the expected flux of the beamline. Figure 3 shows the result. We`ve found a significant higher photon flux compared to the 800l/mm Au-coated grating by up to 2 orders of magnitude. Also, at photon energies beyond 1800eV a high photon flux is available. At about 3keV we have a significant loss in the flux related to the L_3-absorption edge of Rh at the M1 and M3 toroidal mirrors.

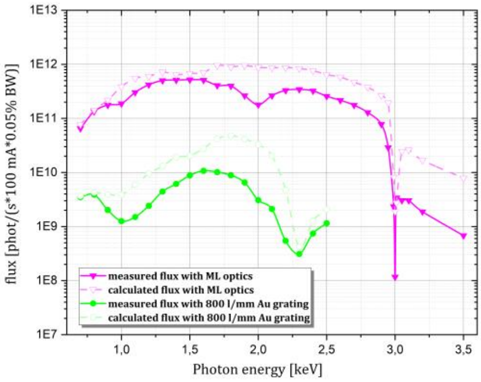

Figure 3. Comparison of the photon flux achieved by Au- and ML-coated gratings as simulated and measured at the beamline by means of a photo-diode. Taken from: [Ref. 23]

The new monochromator permits X-ray imaging in a relatively large photon energy range. In the following, we investigate the influence of the optical setup of the TXM on the imaging formation of a sample at different photon energies from the soft to the tender X-ray range. The interaction of the X-rays with the sample is essentially determined by the distribution of the complex refractive index $n(x,y,z,E) = 1 - \delta(x,y,z,E) - i\beta(x,y,z,E)$ in the object. In general, the δ/β ratio increases with increasing photon energy, i.e. the phase shift in the object is becoming increasingly important. Another significant influence on imaging results from the transmission of the spatial frequencies present in the object by the zone plate objective. Figure 4 shows the analysis of the spatial frequencies present in the images from the IC lamella sample in the photon energy range from 1000 to 2400 eV.

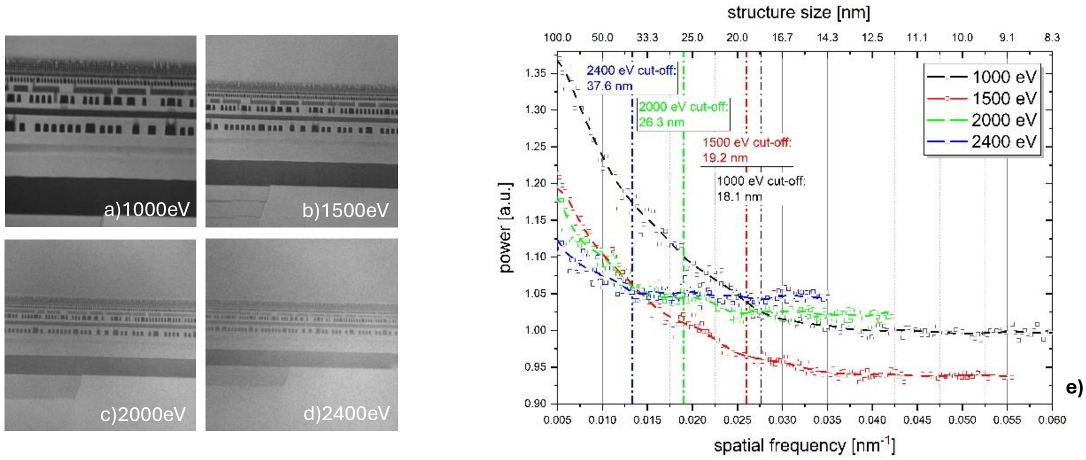

Figure 4. Power spectra (e) extracted from images acquired at different photon energy taken from micro-chip IC structures using the ML-based monochromator setup. The corresponding minimum feature sizes (half-pitch) determined from the TXM images are 18.1 nm, 19.2 nm, 26.3 nm and 37.6 nm, respectively.

The extracted power spectra are the result of several effects. The decrease in spectral power as a function of photon energy is directly related to the weaker interaction of the X-rays with the sample caused by the decreasing δ and β values. To put it simply, the absorption contrast decreases as the photon energy increases. Theoretically the cut-off frequency of the TXM can be determined by the numerical aperture (NA) of the condenser and the NA of the objective lens (i.e. zone plate objective) [25]:

$$f_{cut-off} = \frac{NA_{condenser}+NA_{objective}}{\lambda}.$$

For a reflection optic, the condenser NA is independent of the photon energy. On the other hand, the zone plate NA is a function of E, namely $NA_{ZP}(E) = \lambda/2dr_N = 1239.86/2dr_N E$. Table 2 shows the theoretical cut-off frequencies and the corresponding detectable feature size limits for four different photon energies based on the NA values. As the focal length of a zone plate increases linearly with the photon energy the image magnification according to the lens law for a given distance between objective and detector. Therefore, as the image magnification drops, the corresponding pixel size increases with photon energy.

Table 2. Numerical aperture of the condenser ($NA_{condenser}$) and the objective (NA_{ZP}) as well as the theoretical cut-off frequency, the corresponding theoretical minimum structure size (half-pitch), image magnification, image pixel size and measured minimum structure size of the TXM depending on the photon energy used for imaging.

Photon energy [eV]	**$NA_{condenser}$**	**NA_{ZP}**	**Theoretical Cut-off frequency [nm^{-1}]**	**Theoretical minimum structure size (half-pitch) [nm]**	**Image magnification**	**Image pixel size [nm]**	**Measured minimum structure size**
1000	0.0104	0.0248	0.0284	**17.6**	1683x	**11.8**	**18.1**
1500	0.0104	0.0165	0.0326	**15.4**	1123x	**17.8**	**19.2**
2000	0.0104	0.0124	0.0368	**13.6**	842x	**23.8**	**26.3**
2400	0.0104	0.0103	0.0401	**12.5**	701x	**28.5**	**37.6**

From the power spectra we derived the experimental cut-off frequencies leading to the minimum structure sizes detected in the X-ray images. At 1000 eV photon energy the measured minimum structure size of 18.1 nm is close to the theoretical value of 17.6 nm due to the high magnification. At 1500 eV and higher photon energies the measured minimum structure size deviates increasingly from the predicted theoretical values. It is dominated by the image pixel size restricting the maximum resolving power of the TXM. In future setups the pixel size needs to be adapted to the theoretical resolving power of the TXM. In addition, to compensate for the decreasing absorption contrast with increasing photon energies, the TXM setup has to be extended to phase contrast imaging to exploit the full potential in the tender X-ray regime.

5. Conclusions and Outlook

We have shown ML-coated-gratings in combination with plane mirrors are an efficient option to extend the accessible energy range of grating monochromators at synchrotron and FEL-sources. We also assume this approach to be of interest for many applications at laboratory sources. The upgrade of such monochromators has been demonstrated to be uncomplicated as no change of the existing mechanics, motor or software is required. The verified high contrast and spatial resolution at photon energies up to 2.5keV show an excellent result and verifies the TXM at U41-beamline as a state-of-the-art instrument for spectro-microscopy in the tender X-ray energy range. This might be of interest for biological investigations as well as material science applications related to key elements such as Si, P, S, Y, Zr, Nb or Mo as well as the lanthanides. Further improvement of the beamline is expected by substituting the existing U41 undulator by an elliptical UE30 in-vacuum undulator in the near future. With the emphasis to enable new science in the tender X-ray energy range another three beamline projects at BESSY-II will be equipped by ML-coated blazed gratings allowing to extend the accessible energy range up to 5keV. Alternative ML-combinations like Ni/C- or W/C as well as three-material-coating options at the beam shaping mirrors may offer optimized conditions for dedicated applications in the tender X-ray energy range. This should be a subject of future discussions on beamline innovations.

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