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Study of multiple layers coatings for X–ray mirrors

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Abstract. This paper focuses on a theoretical background that motivated the experimental campaign of multiple layer coatings of X-ray mirrors, and the first results of the testing of the prepared samples. Simulations of the use of different overcoats were performed in order to improve the reflectivity of thin iridium coatings designed for X-ray optics effective in the energy up to 10 keV. Samples based on these simulations were prepared and are being tested for the properties that influence the X-ray optical performance, such as layer homogeneity, density and surface micro-roughness. Further the topic of the coating stress was addressed, as it is an issue in case of thin, lightweight X-ray mirrors and affects the time stability of layers. The discussion and preliminary results conclude our contribution.

Key words: X-ray optics – multiple coating layers – X-ray reflectivity

1. Introduction

Coating layers used for producing X-ray reflective surfaces are usually made of heavy metals like gold and iridium and the thickness of these layers varies from tens to hundreds of nanometres (Kolodziejczak et al., 2000). These layers grant the reflectivity of X-ray mirrors in desired energy range which the mirror is designed for and their quality directly affects the effectivity of the optics (Stehlikova et al., 2017).

The quality of the coating can be evaluated from two different points of view. The first is the qualitative rating of the coating material. Here can be assessed the reflectivity of the selected material, the effectivity of reflection in dependence on the incident angle or the reactivity with the environment during the process of layer deposition. The second examines the properties of the prepared layer. Here are evaluated features like adhesivity between the reflective layer and carrying substrate, microroughness achievable using actual deposition methods and stress in the layers, which can affect the time stability of coatings and others (Döhring et al., 2017).

Next chapters describe theoretical study of coatings dedicated for a Lobster-eye type of X-ray optics (Schmidt, 1975; Hudec et al., 2015), with modified design which was already presented (Stehlikova et al., 2016). The main idea was to prepare a reflective layer suitable for relatively soft X-rays, up to 8 keV, 10 keV maximum. There were two real possibilities of layers production, to prepare a golden layer or to prepare an iridium layer. The iridium layers advantage is about 5 % better reflectivity in the energy range from 2.2 keV to 9 keV, the positive difference is ever rising at the higher energies. A disadvantage which is seen in this design was an energetic gap around 2 040 keV, corresponding to the electron binding energy of Fermi level M5. Here, the reflectivity of the material at the considered incident angle drops from about 90 % to about 10 % and then returns to 80 %, with not so prominent decreases at other Fermi levels (M4, M3, M2, M1; 2040.4 eV, 2116.1 eV, 2550.7 eV, 2908.7 eV, 3173.7 eV respectively).

2. Simulations

Following figures are the result of simulations, prepared as a template for an integrated testing set of multiple layers coatings on silicon. Materials of the layers were chosen according to the previous tests and are reflecting the already acquired experiences. The thicknesses of each layer are chosen according to two main parameters. The first is minimal thickness, which is needed to get a full-reflective layer avoiding energetic transmission into substrate. The second is the influence of thickness on microroughness and energy losses in the layers. Another restriction lies in the used equipment.

The apparatus which has been used to produce samples for real tests is able to work with two targets at once (Probst et al., 2017b). Thus, during changing between two materials there is no need to open the chamber and flood it with air. Layers prepared by this procedure are not threatened by oxidation, which could cause material degradation and generation of indefinable interlayers. Disadvantage is that the chamber has only two targets, so the combinations of layers are strongly limited. The simulations work with the following parameters:

- materials of layers and substrates
- roughness of the surface
- density and thickness of the layer
- energy of incident rays and the angle of incident

2.1. Materials

The materials for planned experiments are chromium and iridium, the substrate for the mirrors is a silicon wafer. Other assessed materials were gold and aluminium. The reasons of the final choice are described below.

According to previous studies, an iridium layer sputtered on super-polished silicon does not have adequate adhesivity. Because of internal stress in the layer and insufficient adhesivity between the layer and the substrate causes cracks and peeling off of the layers (Probst et al., 2017a; Broadway et al., 2015; Stehlikova et al., 2017).

To improve the adhesivity, an interlayer prepared of suitable element can be introduced between the substrate and the reflective layer. Previously, we have tested the interlayers based on aluminium and chromium. These elements have similar crystalline structure and adhesivity tests performed according to ISO2409 standard showed that both aluminium and chromium eliminate the peeling-off issue. Chromium was chosen as the inserted element because of its environmental stability which is generally better than of aluminium. As was mentioned above, when wanting to avoid the oxidation influence in between the coatings, only two targets can be mounted, so the chromium has to be used as the overlayer as well. At this moment, the better stability of chromium is beneficial.

2.2. Surface roughness

The parameter of surface microroughness has strong impact on the final reflectivity, because the wavelength of incident rays is similar to the mean value of microroughness and therefore the scattering effect causes strong energy losses. To get relevant results, RMS values measured on the samples of proper materials and thicknesses were used as a simulation input. Mean values got from these measurements are listed in Tab. 1. All the measurements were performed using atomic force microscope.

The samples which were used are listed bellow:

- a pure silicon wafer with crystallographic structure (1,0,0)
- the same grade silicon wafers with a 100 nm thick layer of chromium
- a 30 nm thick layer of iridium
- a 30 nm thick layer of gold

Traced general trend is rising microroughness with thickness, therefore closest values of tested thicknesses were chosen for test.

2.3. Layer thickness

Thickness of the layers depends on the factor of ideally zero transmission into the substrate for the reflective iridium layer and of stress compensation in the chromium-iridium bilayer. The stresses in iridium and chromium layers are inverse and to achieve the lowest possible stress level, the thickness ratio Cr:Ir is 3:1. (Ames et al., 2015).

Table 1. Table of different sputtered layers microroughnesses.

Surface type	RMS (nm)
Silicon substrate	0.17
Chromium	0.35
Iridium	0.50
Gold	0.38

There is the question of the chromium overlayer thickness as well. The iridium layer is the main reflective surface, and should be opaque for the X-rays in the whole judged range. Contrary to that, the overlayer, which has a better reflectivity at lower energies, should be transparent for the higher, to not limit the reflection at the main layer. According to the sputtering abilities of the machine, the thinnest layer with sufficient homogeneity in all the points is 4 nm.

2.4. Optical parameters of optics

All the material and layer parameters are closely associated with the optical design. X-ray mirrors are usually working with very small, flat incident angle of incoming rays. Rising angle causes, for example, deeper transmission of radiation into the surface layers. Although it has justification in a number of applications, in case of designed lobster eye optics the grazing angle has to be small - less than 0.5 degrees. The energy range of optics is designed for ranges up to 10 keV. Construction parameters are briefly listed in Tab. 2. One segment of planned multi-module optics can be seen in Fig. 1.

As relevant reflectivity limit was chosen 50 %. Even with improved optical design, which allows to focus more rays into the focal point, the ray still undergoes two reflections and that doubles the energy loss. Due to the general weakness of potential observation targets, less reflectivity is not worth.

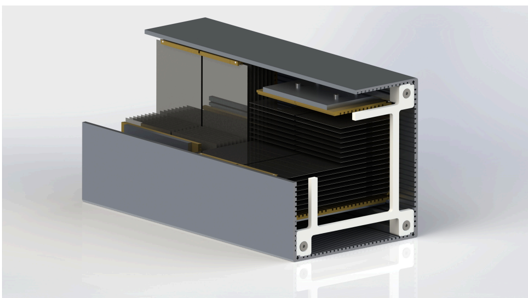


Figure 1. A visualization of lobster eye optical module.

Table 2. Table of basic optics parameters.

Number of mirrors	16
Number of sets	4
Mirror dimension	75 mm to 150 mm
Focal length	2.5 m
Gap between mirrors	6 mm
Thickness of mirror	0.75 mm

2.5. Simulations results and discussion

The simulations were performed for a series of combinations of thicknesses of adhesive layer, main reflective layer and the overlayer. The first problem was to compare the influence of different thicknesses on the absolute reflectivity of main reflecting layer. The comparison of 20 nm, 30 nm, 40 nm and 50 nm of iridium showed only marginal changes (<1%) in reflectivity at energy over 9 keV. The thickness of the adhesive layer was changed as well to ensure the reflectivity will not be changed by using chromium-iridium bilayer. As was expected if the simulation shows that the iridium layer behaves like the opaque one, the underlayer thickness does not have any effects. The risk of changing thickness of adhesive layer is based more on possible rising microroughness with thicker layers, which can then show up in the main layer.

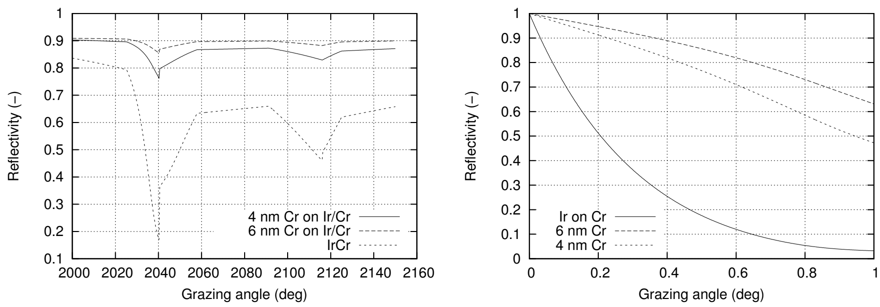


Figure 2. Detail of the energy gap at 2040 eV, where is clearly visible the improvement caused by usage of chromium overlayer (left figure); The dependence of reflectivity on the incident angle for the critical M5 energy. Angle value ranges from 0 to 1 deg, the layer is 4 or 6 nm thick chromium on 30 nm of iridium (right figure)

As was already mentioned, there was a possibility to choose between golden and iridium layer for the main reflective surface. The comparison of reflectivity is in Fig 3, where are shown iridium and golden layers of the same thickness (30 nm)

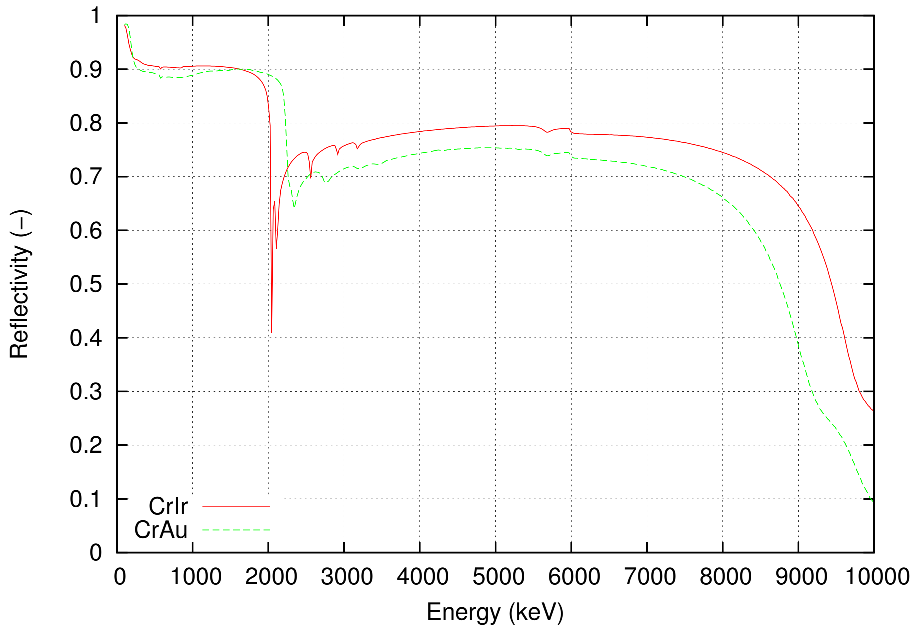


Figure 3. Difference between golden and iridium coating of the same thickness for incident angle 0.5 deg.

and on the same chromium underlayer (90 nm). The reflectivity of iridium is from 3 to 25 percent better in almost the whole scale with an exception of the energy gap at 2 040 eV. The region with reflectivity better than 50 % goes actually even 1 keV further into high energies.

The application of chromium overlayer is illustrated in Fig. 2. There is a detail of the energy gap around M5 for two thicknesses of overlayer and for two conditions; energy change in close surroundings (left figure) and influence of overlayer when changing incidence angle of incoming ray exactly at the binding energy (right figure). The effect of levelling the gaps is clearly visible, thicknesses 4 and 6 nm are accompanied by reference iridium-only surface.

The thickness of the overlayer was scaled from 2 nm to 8 nm. The effect on the reflectivity is antagonistic at low and at high energies. The border between these areas creates at 5 989 eV the K1 line, where the reflectivity of pure chrome layer drops under 10 %. Looking at the figure 4, the thickness of overlayer degrades the reflectivity of iridium equally, but the improvement in the softer rays is more exponential. Preparing of layer thicker than 6 nm seems meaningless, only degrading the high energy part; as ideal thickness for real experiments were chosen 5 nm and 6 nm.

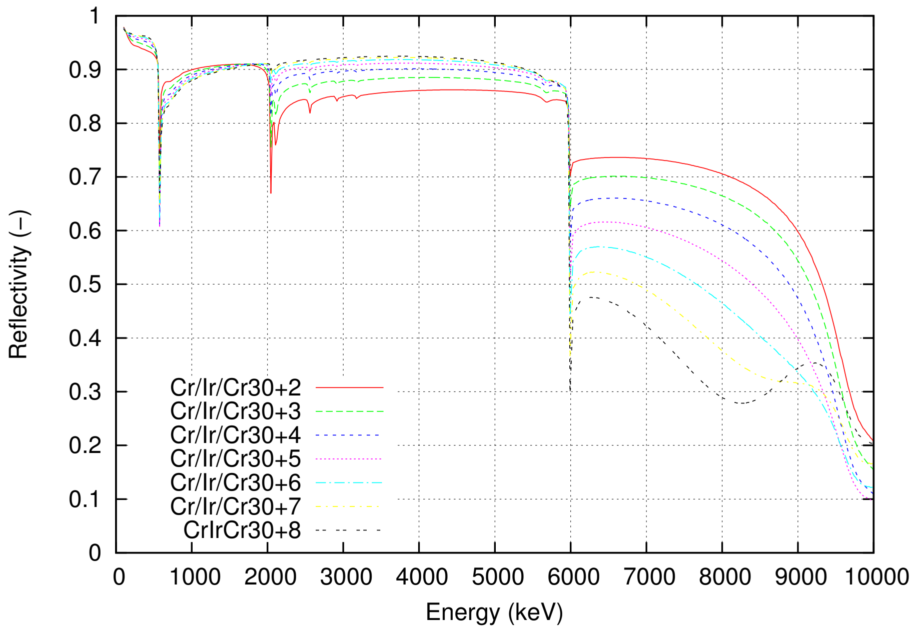
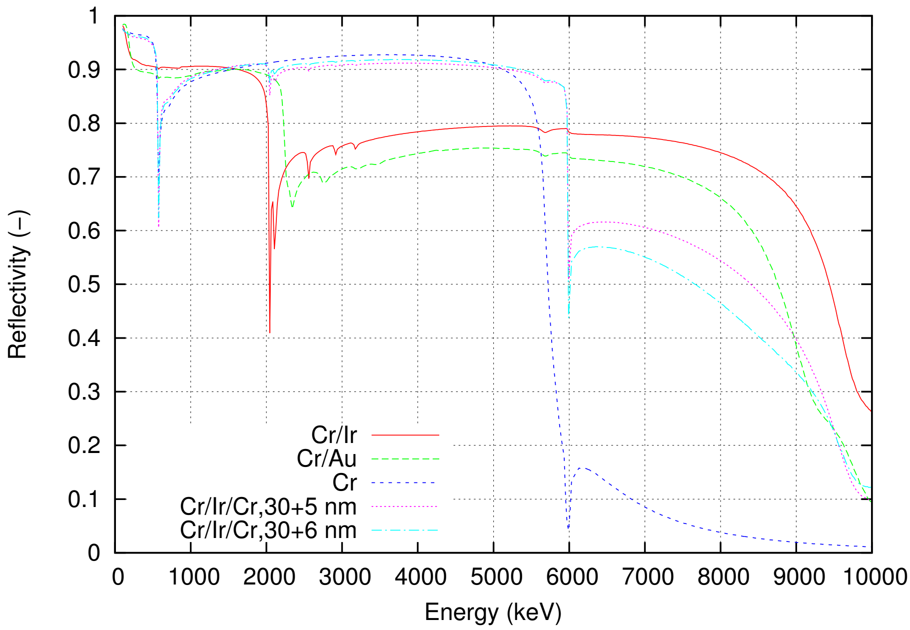


Figure 4. The 30 nm thick reflective iridium surface on 90 nm adhesive chromium interlayer, covered by different thickness (2 nm - 8 nm) chromium overcoating which improves the reflectivity of mirrors at lower energies

For better comparison between the chosen possibilities, there is Fig.2.5. The best compromise seems to be the combination of main iridium reflective layer and 5 nm thick chromium overlayer. It levels the gaps caused by iridium electron binding energies and concurrently does not cause too critical energy losses in the rest of studied energy range. It is necessary to count with not perfect homogeneity of the layers, which can cause fluctuations of layer thickness ±1 nm. Although this value is small, in the context of Fig. 4 is obvious that the effect is not negligible.

3. Real samples production

Based on the theoretical expectations, a series of samples with different layer ratios was produced for testing. To check the layer thickness of thicker layers (tests of 90 nm adhesive layers or main reflective layer) was used tactile stylus profiler Bruker DektakXT. For checking super-thin overlayers, an atomic force microscope (AFM) was used. The AFM was also used to check the microroughness of the surfaces.



From the first tests ensues that with concurrent apparatus set-up it is possible to produce layers of expected homogeneity and thickness. Using variously shaped masks to prepare a measurable step, the fluctuation of the acquired thin chromium overlayers was ± 0.11 nm. The AFM measurement of surface microroughness showed expected rising tendency in dependence on thickness. The difference between the worst and the best case for several tens of samples was 0.236 nm. Still, the most rough surface had RMS 0.408 nm, which value is under chosen 0.5 nm worst-case variant.

4. Conclusion

The paper presents a part of simulations performed to determine the best combination of two materials for future X-ray telescope dedicated for energy range between 1 and 8 keV. The simulation worked with the data and experiences acquired during last years activities. Based on them, input parameters of simulations were chosen to get relevant expectation of a multiple layer coatings behaviour.

According to the simulation results, the most promising coating variant for silicon X-ray mirrors is a configuration of layers silicone - adhesive chromium layer - main reflective iridium layer - levelling and protective chromium overlayer with thicknesses 1.75 mm - 90 nm - 30 nm - 5 nm.

So far were prepared several sets of samples with different combinations of the layers to prove the ability of preparing them with required accuracy. The preliminary results show good quality of surface microroughness which can significantly affect the resultant reflectivity of a mirror, as well as the capability of the sputtering apparatus to prepare very thin homogeneous layers.

Following tests in a vacuum tunnel should examine the X-ray reflectivity of prepared multiple layers to acquire results comparable with the theoretical simulation. If they will be corresponding, a full set of mirrors for the first lobster eye module is going to be produced.

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