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Hard X-ray mirrors for Nuclear Security

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Hard X-ray mirrors for Nuclear Security

LDRD 13-ERD-0046 M.-A. Descalle (PI)

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Abstract

Research performed under this LDRD aimed to demonstrate the ability to detect and measure hard X-ray emissions using multilayer X-ray reflective optics above 400 keV, to enable the development of inexpensive and high-accuracy mirror substrates, and to investigate applications of hard X-ray mirrors of interest to the nuclear security community. Experiments conducted at the European Synchrotron Radiation Facility demonstrated hard X-ray mirror reflectivity up to 650 keV for the first time. Hard X-ray optics substrates must have surface roughness under 3 to 4 Angstrom rms, and three materials were evaluated as potential substrates: polycarbonates, thin Schott glass and a new type of flexible glass called Willow Glass®. Chemical smoothing and thermal heating of the surface of polycarbonate samples, which are inexpensive but have poor intrinsic surface characteristics, did not yield acceptable surface roughness. D263 Schott glass was used for the focusing optics of the NASA NuSTAR telescope. The required specialized hardware and process were costly and motivated experiments with a modified non-contact slumping technique. The surface roughness of the glass was preserved and the process yielded cylindrical shells with good net shape pointing to the potential advantage of this technique. Finally, measured surface roughness of 200 and 130 μm thick Willow Glass sheets was between 2 and 2.5 A rms. Additional results of flexibility tests and multilayer deposition campaigns indicated it is a promising substrate for hard X-ray optics.

The detection of U and Pu characteristics X-ray lines and gamma emission lines in a high background environment was identified as an area for which X-ray mirrors could have an impact and where focusing optics could help reduce signal to noise ratio by focusing signal onto a smaller detector. Hence the first one twelvetant of a Wolter I focusing optics for the 90 to 140 keV energy range based on aperiodic multilayer coating was designed.

Finally, we conducted the first demonstration that reflective multilayer mirrors could be used as diagnostic for HED experiment with an order of magnitude improvement in signal-to-noise ratio for the multilayer optic compared a transmission crystal spectrometer.

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Background and Research Objectives

The last two decades have seen the fast development of hard X-ray focusing optics driven by the astrophysics community needs, and the development of multilayer coatings for new applications related to synchrotron research, medical imaging and non-proliferation. The first multilayer-coated hard X-ray focusing optics for energies above 10 keV was launched in 2012 on board the NASA NuSTAR satellite. [Hayley2010] [Harrison2011] While the NuSTAR telescopes were designed for energies below 80 keV, multilayer coated hard X-ray mirror specular reflectance was recently demonstrated at unprecedented energies up to 384 keV opening new possibilities for applications. [Fernandez-Perea2013]

The goal of the LDRD was to demonstrate the ability to detect and measure hard X-ray emissions using multilayer X-ray reflective optics in a range of energy up to 650 keV – almost a factor of two increase in operational range that could enable new applications related to the detection of 511 keV positron annihilation line for example-; to enable the development of inexpensive and high-accuracy mirror substrates building on the experience gained with the design of the NuSTAR focusing optics, and to investigate applications of hard X-ray mirrors of interest to the nuclear security community. Results obtained in the course of the LDRD are summarized in the following sections.

Scientific Approach and Accomplishments

First demonstration of hard X-rays mirror reflectance above 400 keV

Experiments conducted at the European Synchrotron Radiation Facility (ESRF) demonstrated that 508 keV - close to the 511 keV positron annihilation line and at 644 keV X-rays are reflected by multilayer mirrors at the predicted Bragg angle. Results and analysis are published in [Brejnholt2014].

Hard X-ray mirror substrates

Research was conducted to make inexpensive, x-ray optics substrates (plastics or glass) with predictable, reliable performance. The production of smooth slumped glass substrates is expensive and current techniques to obtain segmented cylindrical shells rely on direct slumping of thin glass sheets on ultrasmooth diamond-turned cylindrical quartz mandrels to maintain smooth surface characteristics. As an example, the NuSTAR optics was optimized for the 10 to 79 keV range and is made of over 2376 mirror segments organized in 133 concentric layers where each segment is made of 210 microns thick D263 Schott glass with surface roughness under 3 to 4 A. Over 40 mandrels of increasing radii were required for the slumping process representing a significant part of the cost of a nested focusing optics. For the range of energy considered here, three types of substrates were considered polycarbonates, Schott glass and a new type of flexible glass. Substrate figure of order 2 to 3 A at most is required and Atomic Force Microscopy (AFM) was conducted on each sample to evaluate surface roughness post-treatment.

Polycarbonate

A substrate made of lightweight thin Polyethylene Terephthalate (PET) plastic foil for X-ray telescopes were first investigated by Schnopper (2002) (2004) and Barbera(2007) however the plastic used in their experiments had unique surface properties not found in common polycarbonates. Polycarbonates are a type of plastic readily available in sheet, that can be easily bent but have poor surface finish [Taibi2007]. Chemical surface smoothing of 375 mm thick polycarbonate sheets with methylene chloride under various conditions of exposure time and temperature and proximal thermal heating as a function of distance, time and temperature did not yield the surface roughness required for hard X-ray mirrors substrates.

Glass Slumping

Combining non-contact mirror slumping technology and electrostatic imprinting, we investigated slumping of 210 microns D263 Schott glass into concave graphite mold.[Al Husseini2011][Liang2003][Youn2012] Graphite is a factor of 20 times cheaper than quartz mandrel, can be turned into smooth surface and has a coefficient of thermal expansions (CTE) well matched to that of glass. The glass was heated in a dedicated furnace to temperature between 600 and 800 degree Celsius. Argon gas was injected through the mold to provide a thin gas-bearing film between the glass and the mold to avoid any imprinting of the mold surface imperfections through the glass. In addition, electrostatic force assist was applied to compensate for a ‘bubble effect’ at the surface of the glass due to gas film pressure differential. AFM confirmed that the glass surface roughness was preserved and remained <3A. Furthermore, the net shape of the slumped cylindrical shell was measured at the NEVIS laboratories at Columbia University and was of order 25 +/-9 arcsec, a result well within the desired range.

Flexible glass substrate

During the last year of the project, a new type of thin flexible glass called Willow Glass® (Dow Corning, MI) became available. Flexible substrates could open the doors to significantly cheaper focusing optics assuming the substrate could be coated flat and bent into the desired shape thus by-passing the glass slumping phase and simplifying the geometry of the multilayer deposition process.

Several parameters were studied in order to determine Willow glass® (WG) potential as substrate for hard X-ray multilayer coatings, namely surface roughness, minimum bend radius, multilayer adhesion and reflectivity. The surface roughness measured with AFM was about 2 to 2.5 A rms, suitable for a high energy X-rays optics substrate. 200 and 130 microns willow glass sheets were bent around cylinders with radii ranging from 10 to 3.8 cm. The 130 microns willow glass could be bend around 4.4 cm matching the smallest radii of the NuSTAR optics.

WC/SiC multilayer coatings with d-spacing of order 1.5 nm were deposited on both glass thickness with N=150 and 300 layers respectively. Coating showed good adhesion to the glass. XRR measurement of reflectivity at 8 keV on N=150 layers deposited on 200 microns willow glass (Figure 1). Predicted and measured reflectivities were 32% and 22% respectively, the difference potentially being due to thermal effects between the glass and the fixture. Results are very promising and the next step would be to study the newly available 100 microns WG and optimize the deposition process as a function of glass thickness. A manuscript summarizing these results is in preparation.

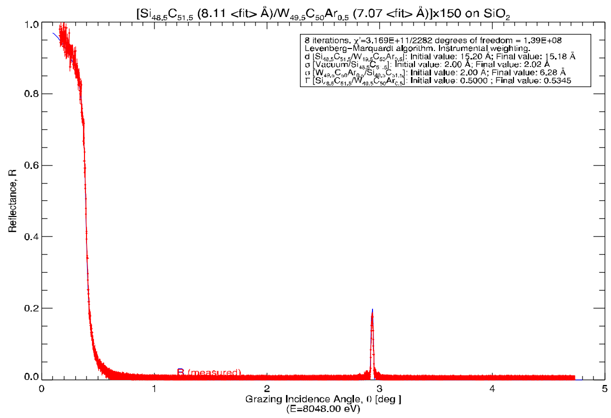

Figure 1. XRR and simulated reflectivity for an 8 keV narrow beam configuration (spot-size was 2.5mm x150 microns at 3 degree). The multilayer consist of N=150 layers of WC/SiC bilayers with d-spacing of 1.5 nm and □ of 0.45

First focusing optics design for the 90-140 keV range

Among the range of applications of interest for national security, detection of weak lines, such as U and Pu characteristics X-ray lines, in a high background environment was identified as a an area for which X-ray mirrors could have an impact. One aspect investigated in this LDRD was the ability to develop nested focusing optics rather than flat filtering optics in the range of energy from 90 to 140 keV since focusing optics could help reduce signal to noise ratio by focusing signal onto a smaller detector placed at the focal plane.
Several issues were identified and investigated from the ML coating recipe to the quality of axial multilayer uniformity deposited on cylindrical glass segments. In collaboration with RXO, LLC, a company with a strong track record for periodic, depth graded and aperiodic multilayer deposition and the capability to coat cylindrical glass segments – used for some of NuSTAR optics-, we investigated the performance of W/SiC and WC/SiC multilayer coatings with d-spacing ranging from 40 to 10 A for hard X rays, more specifically in the energy range from 90 to 140 keV. [Windt2003] Hard X-ray reflectance vs. energy (HXRR) measurements were made at various incidence angles, using RXO's Hard X-Ray Reflectometer that operates up to 160 keV.[Windt2015] Measured reflectivity matched the predicted response very well for coatings with d-spacing greater than 20 A, and the SiC/W films deposited in RXO's coating chamber provide

higher reflectance than SiC/WC in (almost) all cases, but especially for films having periods smaller than d~2 nm.
Secondly, the band pass properties in the 90-140 keV range was evaluated for three depth-graded multilayer coatings. Reflectivities were of order 5 to 20% and differed from simulations. It is anticipated that aperiodic multilayers would deliver a flatter response between 15 and 20%, thus improving the ML performance across that range of energy, and this solution was chosen for the final ML recipe of the focusing optics..
Finally, normal-incidence EUV reflectance measurements were used to determine the coating uniformity of W/SiC ML deposited on NuSTAR cylindrical shells in the RXO coating chamber: although the coating thickness was reduced by about 2% towards the edge of the shell, the axial uniformity was excellent, below a fraction of a percent (Figure 2).

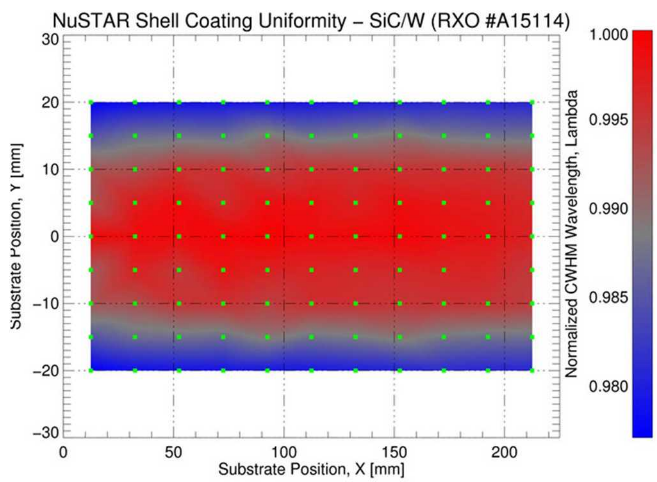

Figure 2. Coating thickness uniformity map for a cylindrical NuSTAR shell.

Based on these results and on the LLNL team expertise designing highly nested focusing optics, a demonstration optic corresponding to one twelvetant of a Wolter I focusing optics for the 90 to 140 keV energy range was designed. It is comprised of four nested layers of 30 degree segments of NuSTAR cylindrical glass shells within a range of angles between 1.904 and 1.962 degree.

First demonstration of use of hard X-rays mirrors in laser-plasma experiment
Experiments conducted at the Titan/Jupiter Laser Facility demonstrated that reflective multilayer mirrors could be used as diagnostic for high energy density physics experiment on four high Z targets. The Kα line complexes of Ta, W, Au, and U were preferentially reflected and successfully detected. Comparisons to data acquired with a transmission crystal spectrometer showed an order of magnitude improvement in signal-to-noise ratio for the multilayer optic. Results and analysis are presented in [Brejnholt2015].

Impact on Mission

The post doctoral fellow hired for this project authored three publications under this LDRD, and made significant contributions on several other laboratory mission-relevant projects related to adaptive optics and NIF diagnostics.
The results obtained in the course of the LDRD positioned LLNL as the leader and expert in hard X-ray reflective optics for safeguards applications. Furthermore, this project helped establish an end-to-end set of expertise, skills and capabilities unique among National laboratories, to model, design, fabricate and assemble nested hard X-ray multilayer focusing optics: this has proven of great interest to several governmental agencies.

Conclusion

Prior to the completion of this LDRD, it was believed hard X-ray multi-layer mirrors range of usability was limited to energy below a few hundreds keV, a range above which Laue Lenses were considered the only option.

The results of this LDRD are rather encouraging and several records or first were established:

- First demonstration of reflectivity at 508 and 644 keV at ESRF
- Development of multilayer models demonstrating that mirror reflectivity can be extrapolated at high energy from lower energy measurements
- First demonstration of usability of willow glass as potential substrate for hard X-ray multilayer coatings
- Design of a >100 keV Walter I nested focusing optics for safeguards applications
- First demonstration that ML coated mirror can be used as diagnostics in laser-plasma experiment at LLNL Titan/JLF.

Two records of invention for polycarbonate chemical polishing and the electro-assist slumping process and three peer-reviewed publications resulted from this LDRD. [Brejnholt2014 and 2014*][Brejnholt2015]

These results were also instrumental in obtaining work for others funding namely a three-year NA22 award to study hard X-ray mirrors for NDA of nuclear spent fuel and a two-year NASA ARPA award to improve Pt and W measured optical properties around L edges.

Through this project, a strong collaboration with RXO, LLC was established to continue research on material systems and aperiodic coatings which is resulting in a DOE SBIR proposal.

The next obvious steps for this research would be to optimize the slumping process, to further investigate the Willow glass properties as a hard X-ray mirror substrate as it could drastically cut the cost of focusing optics., to build the designed optic, test it at wavelength in a synchrotron facility and finally field demonstration experiments.

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