Development of grazing incidence optics for neutron imaging and scattering

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I. Program scope

Because of their wave nature, thermal and cold neutrons can be reflected from smooth surfaces at grazing incidence angles, be reflected by multilayer coatings or be refracted at boundaries of different materials. The optical properties of materials are characterized by their refractive indices which are slightly less than unity for most elements and their isotopes in the case of cold and thermal neutrons as well as for x-rays. The motivation for the optics use for neutrons as well as for x-rays is to increase the signal rate and, by virtue of the optic's angular resolution, to improve the signal-to-noise level by reducing the background so the efficiency of the existing neutron sources use can be significantly enhanced.

Both refractive and reflective optical techniques developed for x-ray applications can be applied to focus neutron beams. Typically neutron sources have lower brilliance compared to conventional x-ray sources so in order to increase the beam throughput the neutron optics has to be capable of capturing large solid angles. Because of this, the replicated optics techniques developed for x-ray astronomy applications would be a perfect match for neutron applications, so the electroformed nickel optics under development at the Marshall Space Flight Center (MSFC) can be applied to focus neutron beams. In this technique, nickel mirror shells are electroformed onto a figured and superpolished nickel-plated aluminum cylindrical mandrel from which they are later released by differential thermal contraction. Cylindrical mirrors with different diameters, but the same focal length, can be nested together to increase the system throughput. The throughput can be increased further with the use of the multilayer coatings deposited on the reflectivr surface of the mirror shells. While the electroformed nickel replication technique needs to be adopted for neutron focusing, the technology to coat the inside of cylindrical mirrors with neutron multilayers has to be developed. The availability of these technologies would bring new capabilities to neutron instrumentation and, hence, lead to new scientific breakthroughs. We have established a program to adopt the electroformed nickel replication optics

Fig. 1: *Pure nickel mirrors electroformed using pulse plating technique*

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technique for neutron applications and to develop the neutron multilayer replication technology.

II. Recent progress

a. Pure nickel replication technique development

Modern electroformed nickel mirrors for astrophysical applications are made from a high-strength nickel-cobalt alloy. This is done because the pure nickel tends to crystallize during conventional electroform plating leading to an optical surface with higher micro-roughness, and because pure nickel has higher stresses that can potentially distort the axial figure profile of a mirror. Both the potential figure degradation and the higher surface micro-roughness can lead to a lower optical performance of neutron mirrors. On other hand, the large amount of cobalt in the mirror material is not acceptable for neutron applications. Therefore, the pure nickel plating process needs to be enhanced in order to control the material stress and to suppress the crystallization of the nickel on the mirror surface.

To address these issues we have applied an innovative pulse plating technique to mirror electroforming. In this, an alternating current, instead of the usual direct current, is utilized so that the plating of the nickel is followed by an etching of the material which removes misplaced nickel atoms. The technique results in a layered nickel structure which makes the material more amorphous-like and suppresses the nickel crystallization. To optimize this, the pulse plating parameters were studied using flat replica samples with the goal to lower the stress and to obtain a good surface roughness. The flat nickel replicas have demonstrated a surface roughness below 6A, the acceptable level for the pure nickel neutron mirrors. Then the process was transferred to cylindrical mandrels. Three nickel mirrors fabricated using the pulse plating technique were nested and assembled into microscope system with magnification factor of four. Figure 1 shows one of the shell mirrors fabricated using the pulse plating technique. The microscope system was tested using the microfocus x-ray source. For comparison we used the similar microscope system fabricated previously using the conventional (non-pulsed) electroformed nickel plating technique. The results are shown in table 1. Please note, the improved optical performance is a result of several factors: improved axial figure of the mandrel, reduced scattering due to surface microroughness and less deformation of the mirrors in the microscope housing due to reduced azimuthal mirror deformations. The microscope system assembled from the pulse-plated pure nickel mirrors is intended for imaging experiments planned to be performed at the National Institute of Standards and Technology’s Center for Neutron Research in July 2012.

Table 1. Focal spot size for the four fold magnified image of microfocus x-ray source

Focal spot size	Half Power Diameter, mm	Full width on Half Maximum, mm
Conventional technique	1.48	0.39
Pulse plating	0.89	0.18

b. Neutron multilayer replication technique development

We are developing neutron supermirror multilayers suitable for direct replication from nickel masters to produce axisymmetric neutron supermirrors. This work is performed in collaboration with the Massachusetts Institute of Technology (MIT) and the Smithsonian Astrophysical Observatory (SAO). The research is focused on two tasks. First, the development of a separation layer applicable for direct replication of multilayers from a master, second, the development of a multilayer coating suitable for nickel replicated mirrors as the substrates.

Fig. 2: *Two conical mandrels used for replication tests (shown in background); three un-coated conical shells, replicated from these (foreground).*

Experiments have been carried out with flat mandrels using thin TiN layers vacuum deposited on to the mandrel followed by the multilayer and the subsequent nickel shell. TiN provides just enough adhesion to support multilayer growth but not enough to inhibit removal. Extensive testing has shown that an 80 A layer of TiN is optimal; the release layer preserves the micro-roughness of the substrate surface, while multilayers replicated from this release layer exhibit no loss of performance, as quantified by x-ray reflectivity measurements. To verify the release mechanism on cylindrical shells, and to tune the plating bath parameters for optimal coatings, replication tests were performed using straight cone mandrels. In these tests, the TiN layer was coated onto cone mandrels and nickel replicas were fabricated from these. Figure 2 shows a selection of mandrels and replicas produced from them. Surface figure measurements were used to compare the figure of the replicated cone with the figure of the mandrel. The samples show clean separation of the replica and mandrel. We are preparing to perform next step in the development program, the direct replication of the neutron supermirror from a mandrel fabricated in accordance with two reflections optical prescription.

A NiC/Ti graded-d-spacing deposition process suitable for direct replication was also developed. First, the experiments were carried with perfectly flat and exceptionally smooth super-polished fused silica substrates with the goal to set the deposition parameters. X-ray reflectivity data were obtained for all samples to assess the quality of the coatings. Figure 3 is an example of the X-ray reflectivity data collected for a fused silica substrate sample coated with m=1.9 multilayers and the model fit. X-ray reflectivity measurements are an excellent means of providing snapshots of the multilayer quality in preparation for more detailed neutron reflectivity measurements that cover larger sample areas and probe the multilayer more deeply.

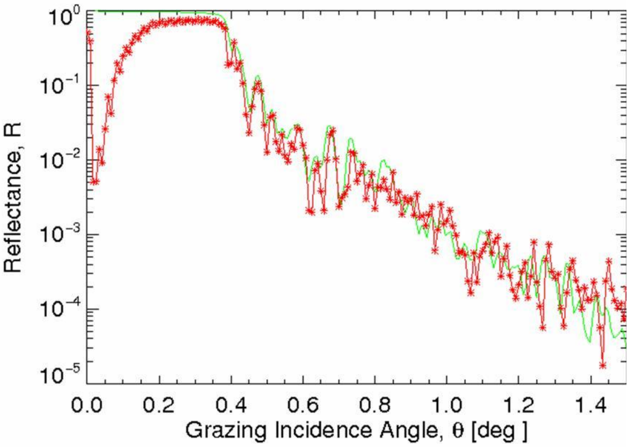

Fig. 3: *Cu K-α (1.54Å) X-ray reflectivity of a NiC/Ti supermirror deposited on superpolished fused silica substrate (red) with model fit (green).*

Using the x-ray measurements, the deposition parameters were tuned so that the microroughness of the interface layers obtained from the fit data was found to be from 7 to 8 A for the fused silica substrate samples, meeting the requirements for the neutron supermirrors. Then, the tuned deposition process was used to deposit films on the flat nickel mandrels which had been polished to have the surface microroughness of 3-4A the level that we expect for the nickel cylindrical mandrels to be used to replicate the neutron supermirrors. X-ray data from these confirmed that the interface microroughness was in the same range, this time between 7.5 to 8.5 A. The multilayer films were also deposited on flat mandrels coated with TiN release layer. The films were over-plated with nickel and released from the mandrels. After x-ray testing confirmed the quality of the multilayer coating, several samples were tested in neutron beams at the MAGICS Reflectometer at SNS. The goal of this test was to verify the quality of the multilayer coatings for neutron reflection and to fine tune the deposition rate parameters. Reflectivity data along with model fit for one of the flat nickel master samples, with 19 layer pairs, m=1.9 and theoretical reflectivity of 97%, are shown in Figure 4 which is a plot of specular reflectivity vs. Qz. The microroughness of the interfaces obtained from the neutron reflectivity data is in good agreement with those obtained from the x-ray reflectivity data for these samples, confirming the value of the quick-look x-ray data. Below the critical angle the measured neutron reflectivity matches the theoretical value very well making the developed process applicable to neutron optics.

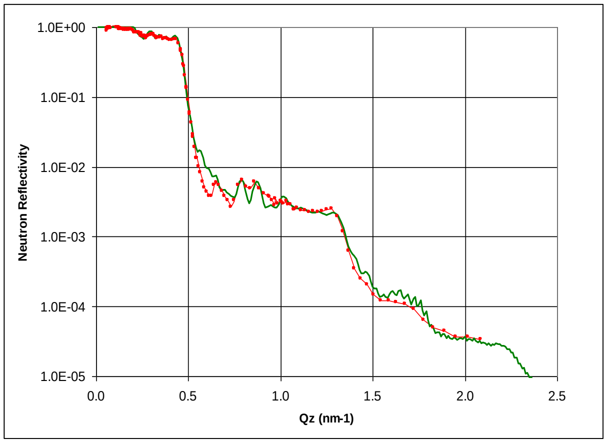

Fig. 4: *Neutron specular reflectivity of a NiC/Ti supermirror (flat nickel mandrel #G09) on Ni substrate (red) and the model fit (green).*

III. Future plans

Pure nickel multilayer-coated mirrors replicated from mandrels fabricated in accordance with two reflections optical prescription will be demonstrated in the near future. A three-mirror pure nickel optic fabricated using a pulse plating technique will be soon tested at the HFIR (ORNL) to demonstrate SANS applications for the developed technology.

Research supported by the U.S. Department of Energy, Office of Basic Energy Sciences, under Awards # DE-FG02-09ER46556 and DE-FG02-09ER46557.

IV. Publication resulting from work supported by the DOE project over the last two years

[1] B. Khaykovich, M. V. Gubarev, Y. Bagdasarova, B. D. Ramsey, and D. E. Moncton, “From x-ray telescopes to neutron scattering: Using axisymmetric mirrors to focus a neutron beam.” Nucl. Instrum. Methods Phys. Res. Sect. A: Accel. Spectrom. Det. Ass. Equip. 631 (2011), 98.

[2] D. Liu, M. V. Gubarev, G. Resta, B. D. Ramsey, D. E. Moncton, and B. Khaykovich, “Axisymmetric Grazing-Incidence Focusing Optics for Small-Angle Neutron Scattering”, Nucl. Instrum. Methods Phys. Res. Sect. A: Accel. Spectrom. Det. Ass. Equip. To be published (2012); doi:10.1016/j.nima.2012.05.056; arxiv:1205.0524v1

[3] M. V. Gubarev, B. Khaykovich, B. D. Ramsey*, et al*, “From x-ray telescopes to neutron focusing”, Proc. SPIE 8147 (2011), 81470B-1.

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Motivation.

Because of their wave nature, thermal and cold neutrons can be reflected from smooth surfaces at grazing incidence angles, be reflected by multilayer coatings or be refracted at boundaries of different materials. The optical properties of materials are characterized by their refractive indices which are slightly less than unity for most elements and their isotopes in the case of cold and thermal neutrons as well as for x-rays. The motivation for the optics use for neutrons as well as for x-rays is to increase the signal rate and, by virtue of the optic's angular resolution, to improve the signal-to-noise level by reducing the background so the efficiency of the existing neutron sources use can be significantly enhanced.

Birth of X-Ray Astronomy

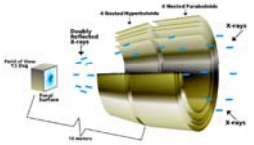
- In 1962, Riccardo Giacconi and colleagues at AS&E flew sounding rocket to look at x-ray fluorescence from the moon
- Lunar signal was overshadowed by very strong emission from the Scorpious region
- Discovered the first extra-solar x-ray source, Sco X-1, and pervasive x-ray background
- This was the effective birth of x-ray astronomy

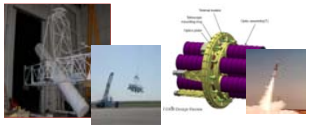
Early observations

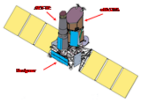
•From these early observations a picture emerged of a typical x-ray source:
•A compact object (neutron star, black hole, white dwarf) orbiting around a normal star
•Matter streams down on to the compact object forming an accretion disk
•As the matter spirals down and is compressed it gets very hot and emits x rays

• X-ray optics are very challenging to fabricate. Because of very short wavelength of x-rays the mirror surface must be smooth to ~ 0.5 nm rms.

• Also, for good angular resolution, the figure must be accurate to < 1 micron.







Different types of X-ray shells fabricated at MSFC

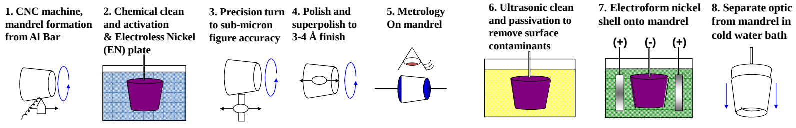

60 cm length – 5 cm diameter - 250 µm thick


Down to 50 µm thick


Up to 50 cm diameter


Down to 2.5 cm diameter

Process



1. CNC machine, mandrel formation from Al Bar

2. Chemical clean and activation & Electroless Nickel (EN) plate

3. Precision turn to sub-micron figure accuracy

4. Polish and superpolish to 3-4 Å finish

5. Metrology On mandrel

6. Ultrasonic clean and passivation to remove surface contaminants

7. Electroform nickel shell onto mandrel

8. Separate optic from mandrel in cold water bath

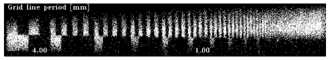
Deposit neutron multilayers

Change Ni-Co alloy to pure Ni.

Challenge – neutron sources have low brilliance and existing beamlines have tight space to place optics
Need for direct multilayer replication technique

Pure nickel replication technique development

Conventional technique



The microscope is capable to resolve the period of 0.290 mm (0.145 mm spatial resolution)

Pulse Plating

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Neutron multilayer replication technique development

Typical release layers for nickel electroforming:
Gold (ebeam)
Nickel oxide

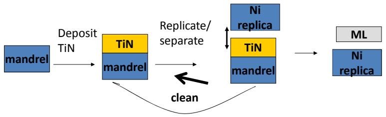
Ideal Properties for release layer

- Smooth (< 4 A microroughness for ML)
- 'Non-sticky'
- High Vickers hardness
- Low stress

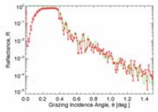
Titanium Nitride: conductor, extreme hardness

Material	Vickers Hardness	Density (g/cm^3)
Ni	~700	8.9
TiN	~2200	5.3
SiC	~2500	3.2

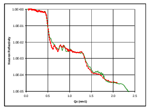
Process




SAO DC magnetron sputtering chamber has 22 inch diameter x 14 inch height.



Cu K-α (1.54Å) X-ray reflectivity of a NiC/Ti supermirror deposited on superpolished fused silica substrate (red) with model fit (green).



Neutron specular reflectivity of a NiC/Ti supermirror (flat nickel mandrel #G09) on Ni substrate (red) and the model fit (green).

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