Development of thin film multilayer Neutron and X-ray optics

Arup Biswas, Piyali Sarkar Roy, Abharana N and Dibyendu Bhattacharyya*
Atomic & Molecular Physics Division, Bhabha Atomic Research Centre, Mumbai – 400 085

Abstract

Over the last few years, a comprehensive facility has been developed at A&MPD, BARC for development of large area thin film multilayer devices for application in synchrotron and neutron based experiments for condensed matter research. This includes a 9 m long in-house developed in-line magnetron sputtering system and a state-of-art dual ion beam sputtering system. The devices being fabricated regularly in these systems include long strip-type synchrotron mirrors, multilayer X-ray mirrors, neutron supermirrors and supermirror polarisers. The performance of these devices are found to be comparable with international standards and these devices are employed in real applications in neutron and synchrotron facilities.

Keywords: DC Magnetron Sputtering, Ion Beam Sputtering, Thin film, Multilayer, X-ray mirror, Neutron Supermirror

Introduction

In condensed mater research neutron and X-ray are being extensively used as probes for understanding various physical properties of materials. Recently these have been boosted worldwide by the availability of high flux neutron and X-ray sources and technological improvement in thin film multilayer devices required for focussing and monochromatisation of these sources. Monochromatic, polarized, collimated and focused beam at different sample environments have been achieved by such thin film multilayer devices[1,2]. Multilayer thin film devices operate on the principle of Bragg reflection. The multilayers can be periodic as shown in Fig 1(a) or non-periodic as shown in Fig 1(b). By creating periodic artificial contrast in neutron scattering length high reflection can be achieved by Bragg principle. In non-periodic multilayers hundreds of layers are arranged in such a fashion that all these Bragg peaks merge and a high reflectivity is achieved continuously over a large angular range. This device is also known as super mirror and it is normally designated by its *m*-value which is defined as the ratio of critical angle of the super mirror compared to that of natural Ni[1]. In some special material combination of the super mirror it can be used for spin polarization of neutron as well as for analysing spin of scattered neutron. Similarly, multilayer with electron density contrast can generate high X-ray reflection. Such multilayers have applications as optical element in lithography, spectroscopy, microscopy and free electron laser experiments[2]. Recently it has shown promise as soft γ-ray mirror also in the non-destructive assay (NDA) of spent nuclear fuel.

Multilayer development facilities

Fabrication of such multilayer structures with layer thickness of the order of nm and sub-nm is a big technological challenge particularly to maintain thickness uniformity over hundreds of layers and to maintain the desired surface and interface morphology. In multilayer devices the surface roughness and

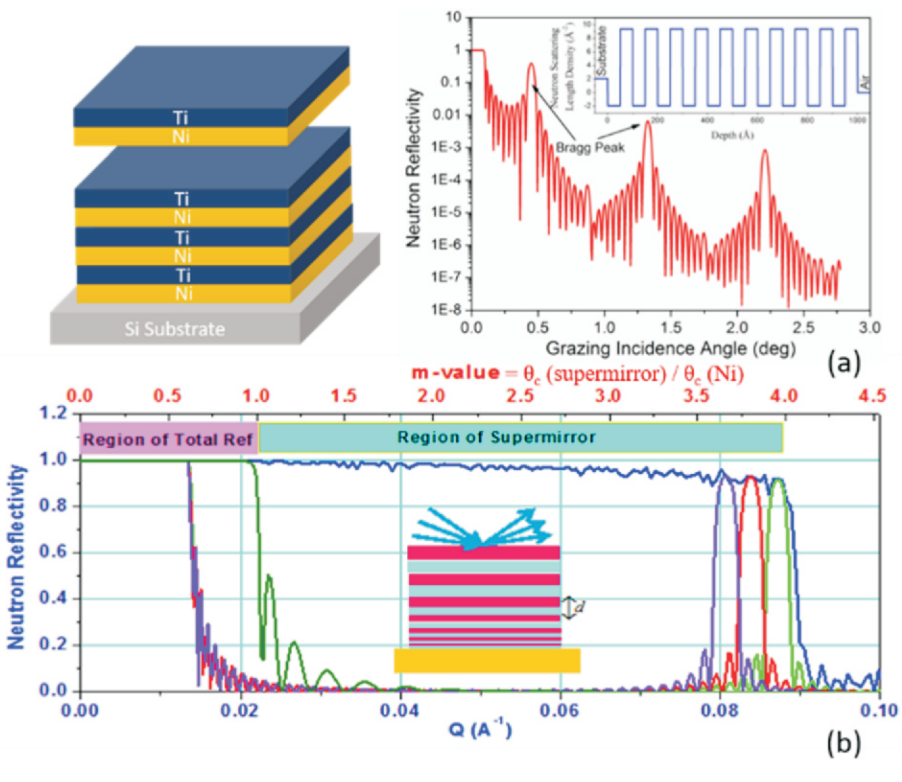

Fig. 1: Structure and neutron reflectivity of (a) periodic multilayer (b) non-periodic multilayer or supermirror

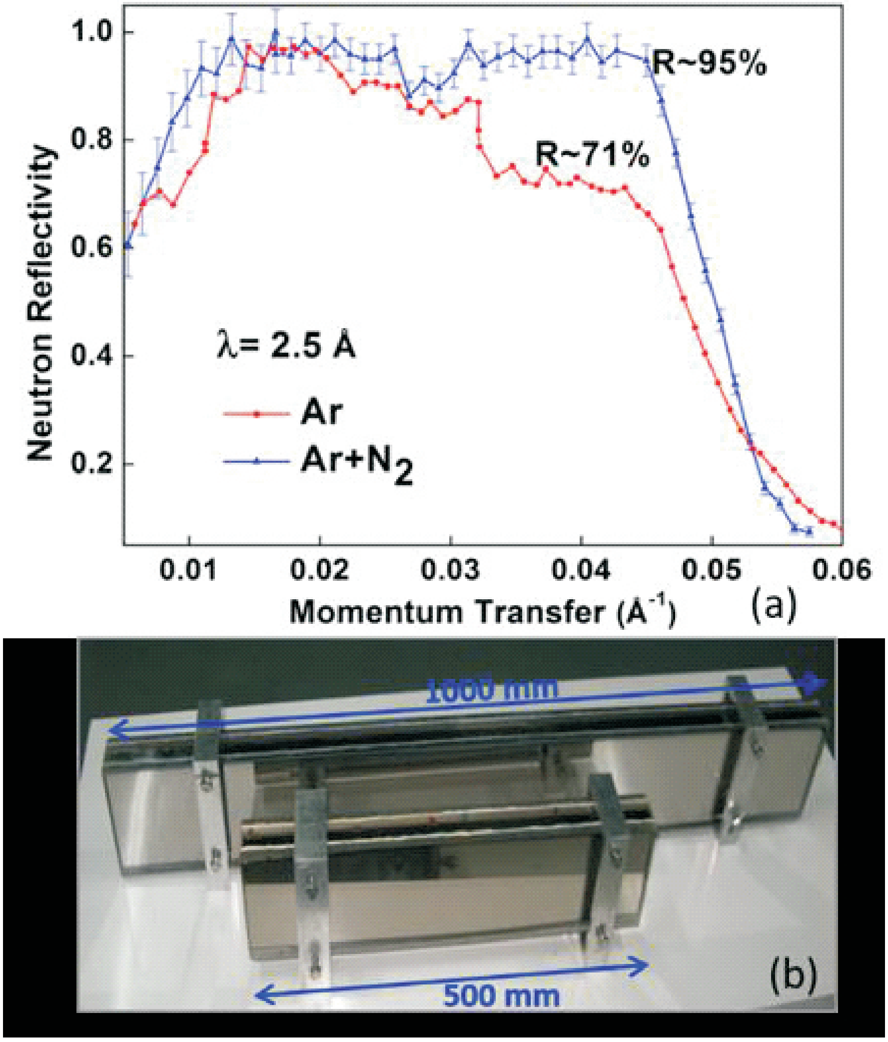

Fig. 2: (a) Measured neutron reflectivity of 98-layer m=2.0 neutron supermirror deposited under pure Ar and Ar+N_2 ambience (b) Prototype 500 mm long supermirror guide tube element for DHRUVA

interface roughness should be maintained below 1 nm throughout the structure to prevent scattering losses. In application of low grazing incidence angle X-ray mirror large size mirrors are essential to cover the whole foot print of the X-ray beam from a synchrotron source. In our laboratory, magnetron sputtering and ion beam sputtering techniques are being used for the development of such multilayers since sputtering techniques yield higher adatom energies at the substrate surface which ultimately results in smoother surface and interfaces in the multilayers. A 9 m long DC/RF magnetron sputtering large area coating system (LACS) has been designed and built indigenously in our laboratory for this purpose[3]. The fully automated computer-controlled system is capable of depositing more than 500-layer multilayer thin films on substrate of maximum dimension 200 mm x 1500 mm following a pre-determined recipe without manual intervention. A state-of-art Dual Ion Beam sputtering (DIBS) system IonSys-800 (Meyer Burger, Germany) has also been installed in our laboratory recently which can deposit layers with sub nm precision and bulk-like density. Using these two systems recently we have developed several neutron supermirrors, neutron supermirror polarizers, Synchrotron X-ray mirrors, soft X-ray mirrors for XUV and water window region which have various applications in condensed matter research. Few examples are given below:

Neutron Supermirror

Neutron supermirrors can be used for fabricating neutron guide tubes which can deliver neutrons from source to the experimental station with higher flux. Also curved supermirrors can be used for focusing neutrons on samples with a spot size ~100 μm[4]. Using the in-house developed 9 m long magnetron sputtering system of our laboratory, 98-layer m=2.0 Ni/Ti neutron supermirrors have been developed following the design carried out by an in-house developed code[5]. The reflectivity of the supermirrors have been improved significantly to 95% by depositing the Ni layers of the multilayers under a mixed ambience of Ar+N_2 instead of under pure Ar ambience. The neutron reflectivity of two supermirrors deposited with under pure Ar and under Ar+ N_2 mixed ambience are shown in Fig 2(a), Fig.2(b) shows the photographs of 1000 mm and 500 mm long Ni/Ti supermirrors produced for applications in guide tubes of DHRUVA.

In order to investigate the reason for increase of neutron reflectivity of Ni/Ti supermirror due to deposition of Ni layer in ambience of Ar+N_2 several periodic multilayers are deposited at similar condition and characterized by grazing incidence X-ray reflectivity (GIXRR), X-Ray Diffraction (XRD), grazing incidence extended X-ray absorption fine structure (GI-EXAFS) and grazing incidence X-ray fluorescence (GIXRF) techniques [6,7]. In Fig 3(a), GIXRR of 10-bilayer Ni/Ti periodic multilayers deposited under pure Ar ambience and under mixed ambience of Ar+ N_2 are shown. By fitting these data it is found the Ni-on-Ti interface width decrease from 0.5 nm to 0.4 nm and Ti-on-Ni interface width decreases from 0.7 nm to 0.2 nm upon deposition under mixed ambience. X-ray diffused scattering measurements on the multilayers show that the reduction of this roughness is more due to reduction of interface diffusion. These two multilayers also characterized by GIEXAFS technique at Ni K-edge using BL-9 beamline Indus-2 as shown in Fig 3(b). This again manifests less interface diffusion in multilayer deposited under mixed ambience with Ar+N_2. XRD patterns of the two multilayers, as shown in Fig 3(c), reveal that the reduction in diffusion is due to formation of amorphous Ni layer when sputtered under mixed ambience of Ar+N_2. Nuclear resonance reaction (NRR) studies reveal that presence of nitrogen in Ni lattice leads to amorphisation of Ni layers which ultimately leads to less interface diffusion. Element specific diffusion of the Ni and Ti have been investigated using GIXRF technique at as shown in Fig 3(d), analysis of which also shows that at interfaces the change in Ni concentration is sharper in the multilayer deposited under Ar+N_2 mixed ambience compared to the multilayer deposited under pure Ar ambience.

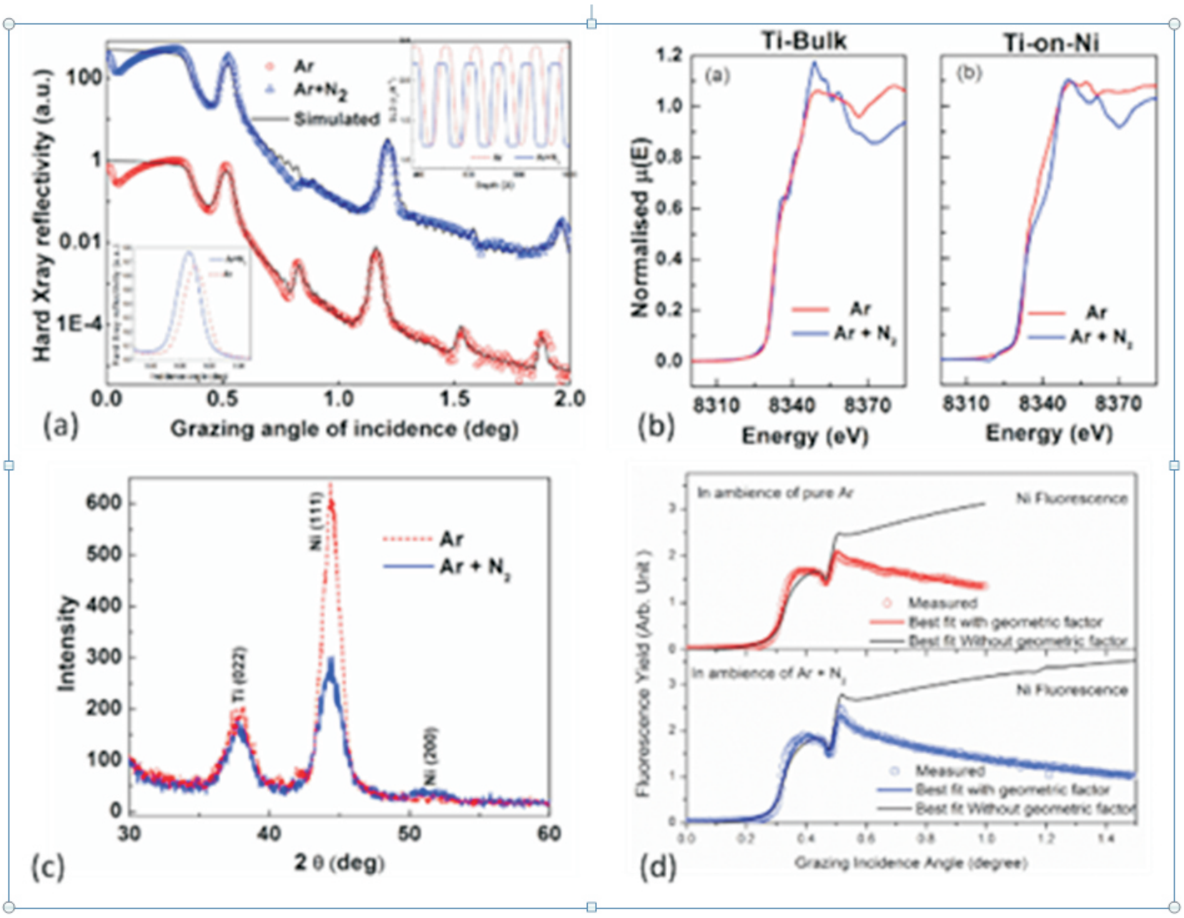

Fig. 3: Characterization of Ni/Ti periodic multilayer deposited under Ar and under mixed ambience of Ar+N_2 (a) GIXRR (b) GIEXAFS (c) XRD (d) GIXRF

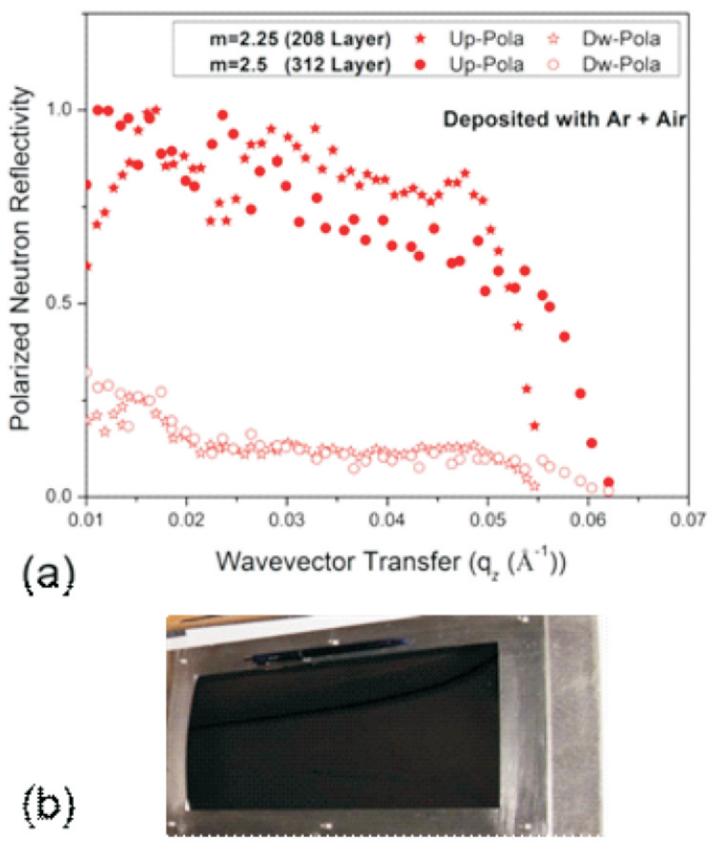

Fig. 4: (a) m=2.25 and m=2.5 Co/Ti supermirror polarizer (b) A Co/Ti supermirror polarizer having dimension of 251 mm x 141 mm element.

Neutron Supermirror polarizer

In a supermirror polarizer alternate layers of magnetic and non-magnetic materials are chosen such that neutrons with spin polarization in the direction of the magnetization get reflected from the multilayer and neutrons with spin polarization in the opposite directions pass through it. These devices are used to polarize the neutron beam in neutron scattering experiment as well as to analyse the spin of scattered neutrons from a sample, like in a spin echo measurement. Using the 9 m long magnetron sputtering system Co/Ti supermirror polarizers of m=2.25 (208 layer) and m=2.5 (312 layer) have been developed[3]. It is found that the polarized reflectivity of the supermirror improves when Co layers are deposited in under mixed ambience of Ar and air. In Fig 4(a) the polarized neutron reflectivity (PNR) spectra of these supermirrors are shown. At this condition Co/Ti supermirror polarizers are developed on both sides of 500 μm thick glass substrates having dimension of 141 mm x 251 mm, a photograph of which is shown in Fig 4(b).The cross sectional TEM micrographs of two m=2.5 supermirrors

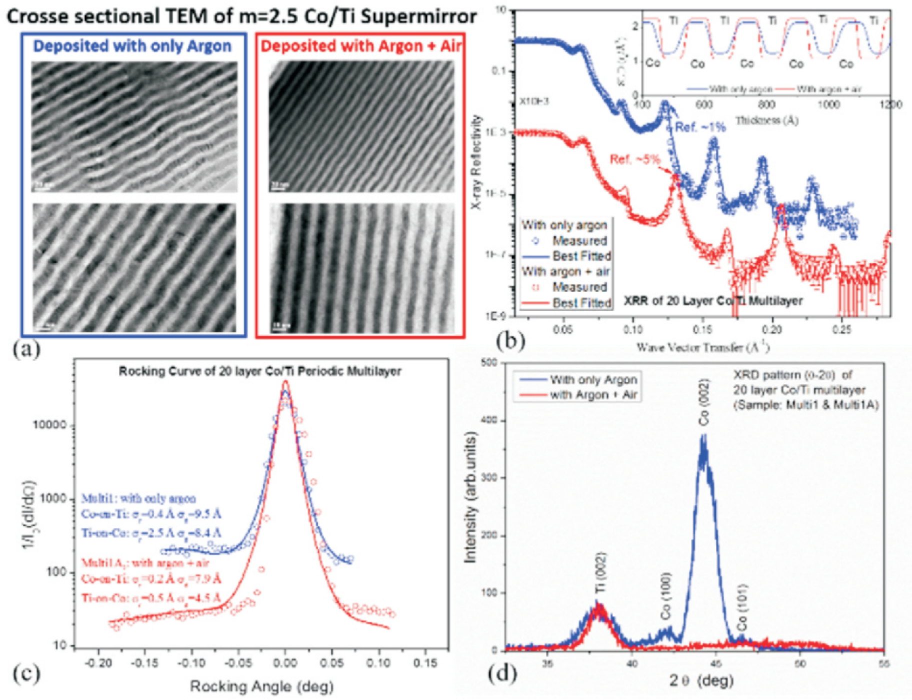

Fig. 5: Characterization of Co/Ti multilayer prepared under pure Ar ambience and under mixed ambience of Ar+air (a) TEM of m=2.5 supermirrors (b) XRR of periodic multilayers (c) rocking scan of periodic multilayers and (d) XRD of periodic multilayers.

are shown in Fig 5(a), it is seen that the layers are wavier in the supermirror deposited under pure Ar ambience compared to the supermirror deposited under mixed ambience of Ar and air, which also has relatively sharper interfaces. Further investigations have been done by preparing 20 layer Co/Ti periodic multilayers and characterizing them by GIXRR, PNR and XRD techniques[8]. In Fig 5(b) it is seen that the Co-on-Ti and Ti-on-Co interface widths decrease when Co layers are sputtered under mixed ambience of Ar and air, also the SLD at the interfaces are found to be sharper with higher contrast. Diffused X-ray scattering measurements at grazing incidence are shown in Fig 5(c), analysis of which reveal that this change of interface width is due to reduction of interface diffusion. XRD patterns of these two multilayers shown in Fig 5(d) reveal that the reduction of interface diffusion or waviness is due to amorphization of Co layer when sputtered under mixed ambience of Ar and air.

Synchrotron X-ray Mirror

In Synchrotron beamlines grazing incidence mirror are very much essential for collimation and focusing of the beam since reflectivity of X-rays is very high at grazing angle of incidence. As these mirrors work at very low grazing angle on the principle of total external reflection, the lengths of these mirrors are generally large (1000mm or above) to cover the footprint of the whole SR beam. Using the in-house developed 9m long LACS system Cr/Ausynchrotron mirrors have been deposited on plane (500 mm x 51 mm x 51 mm) and spheroidal(300 mm x 51 mm x 47 mm) zerodour substrates[9]. In Fig. 6 the hard X-ray (0.154 nm) reflectivity, soft X-ray reflectivity and the photograph of the mirrors are shown. The soft X-ray reflectivities of the mirrors are found to agree well with theoretical simulations. These mirrors have been installed at the PAS beamline of Indus-1 SRS.

EUV and SXR Multilayer Mirror

X-ray multilayer mirrors are also essential for reflecting and monochromatising extreme ultra violet (EUV) and soft X-ray (SXR) (1-60 nm) wavelengths since in this region natural crystals and gratings are not available2. Apart from applications as optical components in beamlines of synchrotron and free electron laser sources, these mirrors have some other specific practical applications. For example, in EUV lithography systems where soft X-ray radiation from 6-14 nm are used and in soft X-ray microscopes operating in the water window region of 2.3–4.4 nm, where water is transmissive but carbon based organic materials shows strong absorption, these multilayer mirrors have acquired great interest. Using the DIBS system of our laboratory, Mo/Si multilayers with bi-layer thickness of 7 nm have been developed which can reflect 13 nm wavelength at higher grazing angle of incidence. In Fig. 7(a) and (b)the hard X-ray and soft X-ray reflectivities of the 25 bi-layer Mo/Si multilayers are respectively shown. It is found that the multilayer deposited with C buffer layer at the Mo/Si interfaces has better reflectivity than that deposited without the buffer layer. The Mo/Si multilayer with C buffer layer has achieved

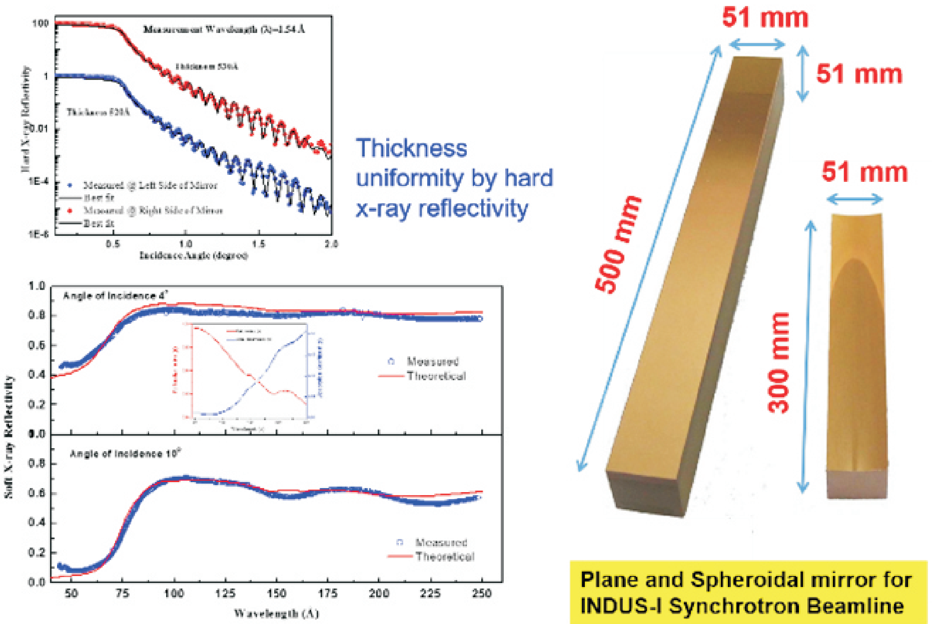

Fig. 6: Hard X-ray and soft X-ray reflectivities of Cr/Au grazing incidence Synchrotron mirror for PAS beam line at Indus-1SRS.

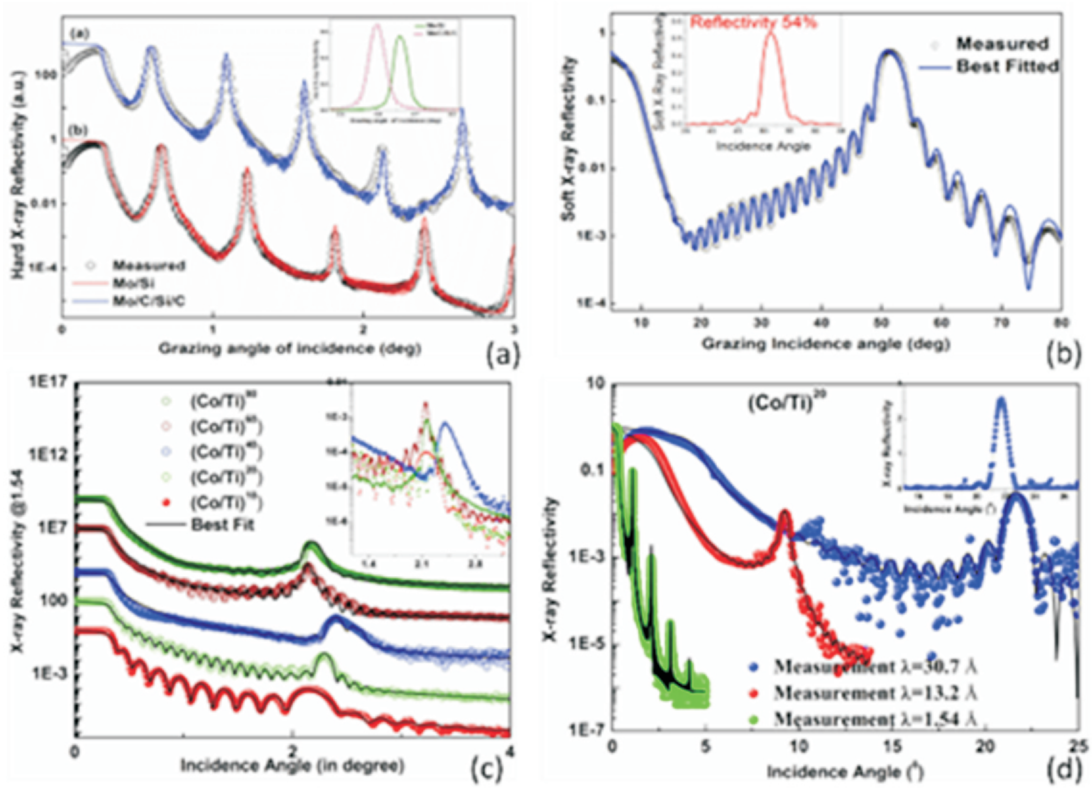

Fig. 7: (a) Hard X-ray (b) Soft X-ray reflectivities of 25 bi-layer pair Mo/Si multilayer for EUV lithography application. (c) Hard X-ray (d) Soft X-ray reflectivities of Co/Ti multilayers for water window application.

54% reflectivity at 51° grazing angle of incidence[10]. Further GIEXAFS measurements with Indus-2 SRS confirm lowering of interface diffusion in this multilayer due to the presence of C buffer layer.

For applications in water window region however, due to lower value of the probing wavelength (2.2-4.4 nm) multilayers with lower bilayer thickness are required. Co/Ti multilayers with low bi-layer thickness of 1.8 nm (with nominally 0.9 nm of Co and 0.9 nm of Ti layers) have been deposited in the LACS system with a specially designed SS mask, placed below the target at a distance of 50 mm which reduces the exposed length of the target along the direction of the substrate trolley movement. The variation of hard X-ray reflectivity of Co/Ti multilayer samples with no. of layers are shown in Fig.7© which shows that maximum reflectivity is obtained for the 60 bi-layer sample[11]. Fig.7(d) shows that reflectivity of 2.5% has been obtained for the Co/Ti multilayer sample at 21.5° grazing angle of incidence and for 3.07nm of soft X-ray wavelength[12].

Using the DIBS system Cr/Ti multilayers with bi-layer thickness ranging from 3.8 nm to 2.1 nm with an optimised Cr to bi-layer thickness ratio of 0.4 have been deposited. Fig. 8(a) and (b) show the variation in the diffused X-ray scattering spectra and Cross-sectional TEM micrographs of Cr/Ti multilayers with different bi-layer thicknesses. From the above measurements it could be concluded that for all the ML samples Ti layer thickness is higher than 1.3 nm and Ti forms continuous layer on Cr for all cases. However, Cr layer thickness varies from 1.5-0.8 nm in the above range of bi-layer thickness and Cr undergoes continuous-to-discontinuous transition as the layer thickness reduces below 1 nm[13].

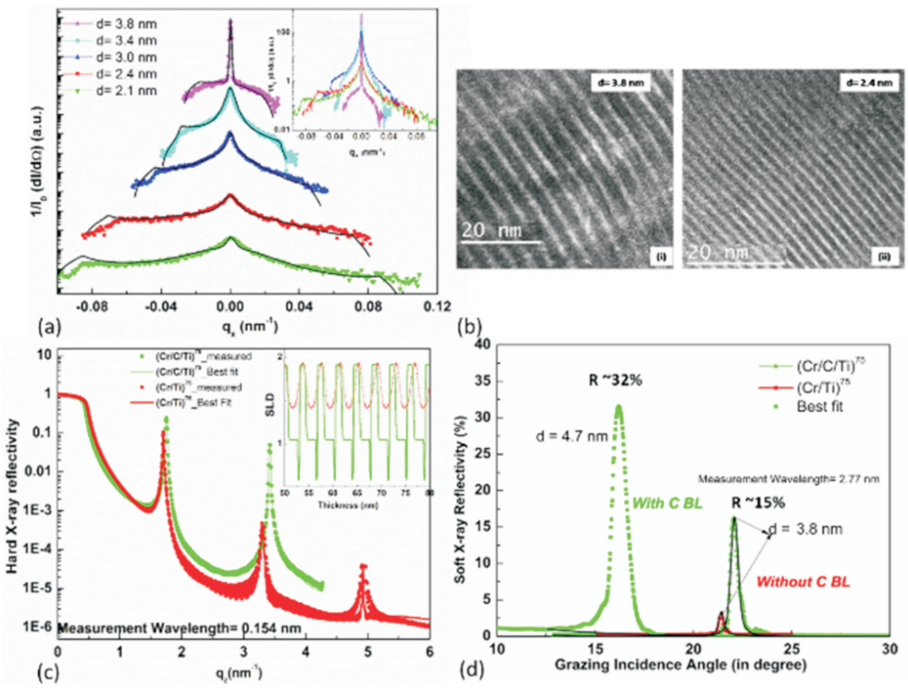

Fig. 8: Characterization of Cr/Ti multilayer (a) Diffused X-ray scattering (b) Cross sectional TEM (c) Hard X-ray reflectivity (d) Soft X-ray reflectivity

The hard X-ray and soft X-ray reflectivities of Cr/Ti periodic multilayers with and without C buffer layers at the interfaces are shown in Fig. 8(c) and 8(d). It can also be seen from the above figure that for a Cr/C/Ti multilayer of slightly higher bilayer thickness of 4.7 nm, very high (~31.6%) reflectivity for 16.2° grazing angle of incidence can be obtained at water window soft X-ray wavelength of 2.77 nm, which is the highest reflectivity reported so far in the literature in this wavelength regime[14].

Summary

Development of multilayer optics is indispensable in the condensed mater research using neutrons and X-rays as probe. Over the years a comprehensive facility has been created at A&MPD, BARC consisting of the in-house developed large area coating system (LACS) and the commercially procured DIBS system, for fabrication of these multilayer devices with sub-nm level control over thickness and interface morphology. Using the above facility non-periodic Ni/Ti neutron supermirrors and Co/Ti supermirror polarizers have been developed for application in neutron optics, while 1.5m long Cr/Au grazing incidence Synchrotron X-ray mirror, Mo/Si multilayer mirrors for soft X-ray EUV applications and Co/Ti and Cr/Ti multilayer mirrors for soft X-ray water-window applications have been successfully developed. In each of the above cases by optimizing the process condition or introducing buffer layer at the interfaces the performance of the multilayer device has been improved to the quality comparable to or better than that reported so far in the literature.

Corresponding Author*
Dibyendu Bhattacharyya
(dibyendu@barc.gov.in)

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