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Fabrication and Characterization of Nickel Titanium Multilayer Neutron Supermirrors

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We have designed and fabricated two Nickel Titanium based multilayer neutron supermirrors following a design formalism proposed by Hatyer and Mook. One mirror has 10 bilayers and the other one has 30 bilayers. The neutron reflectivity profiles of these supermirrors have been measured. The critical angles for total external reflections are 36 and 41 minutes of an arc for the mirrors with 10 and 30 bilayers respectively for neutrons of wavelength 4.06 Å. The profiles match the expected calculated reflectivity reasonably well.

KEYWORDS: Neutron, Supermirror, Reflectivity

§.1. Introduction

Neutrons undergo total external reflection from most of the material surfaces at near grazing incidence. The refractive index of the medium depends on the optical potential experienced by the neutron inside the material. One can calculate the refractive index of a medium for neutrons from optical theory,[1)] where the medium is considered as a homogeneous dispersive media. Refractive index 'n' of a media for neutrons of wavelength λ is given by the expression,

$$n = 1 - \lambda^2 \rho b_{coh} / 2\pi, \qquad (1.1)$$

where ρ is the number density of the medium and b_{coh} is the coherent scattering length of the material. Most of the materials have positive b_{coh} and have refractive index lower than unity with respect to vacuum. Neutrons undergo total external reflection from surfaces of such materials. The critical θ_c for total external reflection is given by,

$$\theta_c = \sqrt{2(1-n)} \qquad (1.2)$$

which depends on the wavelength λ, number density ρ, and coherent scattering length b_{coh} of the medium. Materials with larger ρ and b_{coh} will have larger critical angles. Utilizing total external reflection of neutrons from smooth metal surfaces, neutrons are transported from their source to tens of metres of distance away in neutron guide tubes. The guide walls are usually made from nickel coated on float glass substrate. Nickel has a large coherent scattering cross-section of 1.03×10^{-12} cm and low absorption cross-section among the metals. Ni has a critical angle of about 6 minutes of an arc per Å. Float glass substrate has low surface roughness and reduces diffuse scattering of the beam. Usually about 1000 Å to 1500 Å thickness of Ni coating is sufficient for total external reflection of neutrons in a guide.[2)] To increase the critical angle for total external reflection, instead of a single layer, multiple bilayer stacks of varying thicknesses with materials of widely different coherent scattering length have been developed.[3)] In his pioneering work, Mezei had suggested a formalism to design such stacks, aptly called supermirors, based on continuous variation of the layer thicknesses.[4)] If these bilayers are made from magnetic and non-magnetic alternating layers, which is usually the case, under a magnetic field the critical angle for neutrons with spins parallel to the field will have widely different critical angle of reflection with respect to neutrons with spin antiparallel to the field.[3)] The origin of the development of supermirrors lies in attempts to produce polarized neutron beam by mirror reflection.[3-4)]

In the present work we report fabrication and characterization of two supermirrors using nickel and titanium following an algorithm, given by Hayter and Mook,[5)] in which they account for the discrete nature of the bilayer stacks and also derive the correct continuum limit for thicknesses of individual layers in a particular bilayer of the stack. Titanium has a negative coherent scattering length of -0.33×10^{-12} cm and has very good contrast with respect to nickel ($b_{coh} = 1.03 \times 10^{-12}$cm) for neutrons. It is understood that to increase the critical angle of a supermirror, the number of bilayers should be large. But increase in the number of bilayers will also result in large cumulative deviation from the design thicknesses, after deposition, with respect to a mirror with smaller number of bilayers. We have designed two mirrors, one with 10 bilayers of nickel and titanium and another with 30 bilayers of nickel and titanium, in an attempt to study the effect of increase in number of bilayers on the critical angle and on the reflectivity profile.

The supermirrors were deposited in an electron beam heated vacuum deposition system in Nagoya University, Japan. The supermirrors have been characterized using a neutron reflectivity measurement set up in DHRUVA reactor, India.[6)] In section 2 we discuss the design of the supermirrors, following the formalism of Hayter and Mook. Section 3 contains the

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details of the vacuum deposition system. In section 4 the neutron reflectometer set up is described, followed by the results of the experiments, discussions, and the conclusions in section 5.

§.2. Design

Hayter and Mook have observed that the correct continuum limit for the thicknesses of layers in the k-th bilayer of the supermirror is given by equations,[5]

$$d_{k1} = d_c / \xi_k,$$
$$d_{k2} = (d_{k1}^{-2} + d_c^{-2})^{-1/2}, \qquad (2.1)$$

where 1 denotes the optically less dense material. d_c is given by,

$$d_c = \lambda / 2\theta_c. \qquad (2.2)$$

ξ_k is a root of the equation

$$\xi_k^4 - 2k^{1/4}\xi_0\xi_k^3 + \xi_k^2 - 2k^{1/4}\xi_0\xi_k + k^{1/2}\xi_0^2 = 0 \qquad (2.3)$$

and the constant

$$\xi_0 = 0.5657864. \qquad (2.4)$$

One can calculate the thicknesses of the layers, given the materials used in the bilayers from eq. (2.1). We have designed two supermirrors using Ni/Ti combination, one with 10 bilayers and the other with 30 bilayers in order to study the increase in the critical angle with the increase in the number of bilayers and other related changes in the reflectivity profile. This also helped to find out the robustness of the design formalism. It is known that quality of the film and roughness increases with increase in the number of layers which tends to modify the measured critical edge and the diffuse scattering. The first layer at the air-mirror interface is nickel in case of both the mirrors fabricated by us. According to the calculation this layer thickness should be 252.12 Å. We have doubled it to 504.2 Å in our design to inhibit any drop in reflectivity at the critical edge of nickel due to incomplete reflection in the first layer. It was intuitively felt that by doubling the layer thickness we do not introduce drastic difference in the phase relationship between the incident and reflected waves.

§.3. Deposition of the Supermirrors

The supermirrors were deposited in clean ultra-high vacuum system (10^{-11} torr or better) by electron beam heating of the target materials nickel and titanium. The details of the deposition system has been discussed elsewhere.[7] A schematic of the system is given in Fig. 1. The substrates used for deposition of the multilayers were float glass of very low surface roughness. During the deposition the substrate was kept cooled to liquid nitrogen temperature. This was done to inhibit the inter layer diffusion at the Ni/Ti interfaces, so that the deposited films are as close to the ideal design thicknesses and morphology as possible. The deposition rate is normally 0.1 Å /sec as monitored in a quartz oscillator. The thickness can be controlled to about 0.2 Å. After deposition the multilayers were slowly brought back to room temperature, being

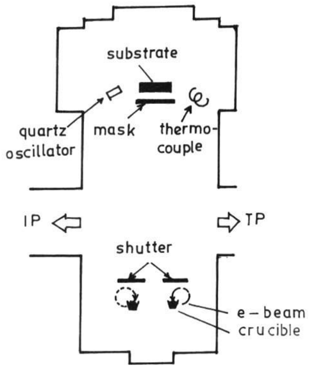

Fig. 1. The schematic of the vacuum deposition unit. TP denotes turbomolecular pump and IP denotes ion pump.

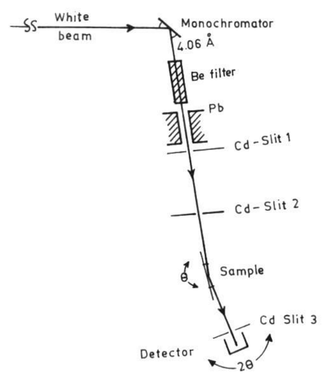

Fig.2. The neutron reflectometer setup at Dhruva. The monochromator used is pyrolytic graphite, giving 4.06 Å neutrons. The lead block with a slit of about 5mm × 35mm is used to reduce the gamma background. Beryllium filter is used to remove 2 Å neutrons from the beam. Cd slit 1 and Cd slit 2 are the cadmium slits which form the collimator, giving a beam of 6 minutes of arc divergence.

maintained under vacuum during the heating. The size of the mirrors deposited are about 30mm by 70mm. The mirror with 10 bilayers and the other with 30 bilayers were deposited in the same run. The substrate of the mirror with ten bilayers was masked while the first twenty bilayers from the substrate upwards were deposited on the other mirror, following which both the mirrors were opened and the rest ten bilayers were deposited. So the top twenty layers of both the mirrors are nearly identical.

§.4. Characterization of the Supermirrors

The neutron reflectivity profiles of the supermirrors were measured using a reflectivity measuring attachment on an existing Filter Detector Spectrometer in DHRUVA reactor which can measure reflectivity profile of vertical samples.[6] The schematic is shown in Fig.2. This consists of a collimator with two cadmium slits of widths 0.8mm and 0.4mm respectively and height about 35mm, separated by 600mm distance. Neutrons of wavelength 4.06 Å from a graphite monochromator after passing through a Be filter (for removing $\lambda/2$ contamination from 2 Å neutrons in the beam) pass through this

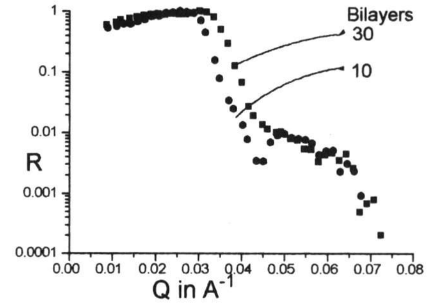

Fig.3. Reflectivity (R) plotted against Q for both the supermirrors. The mirror with 10 bilayers has a critical angle of 36 minutes of an arc and the mirror with 30 bilayers has a critical angle of 41 minutes of an arc.

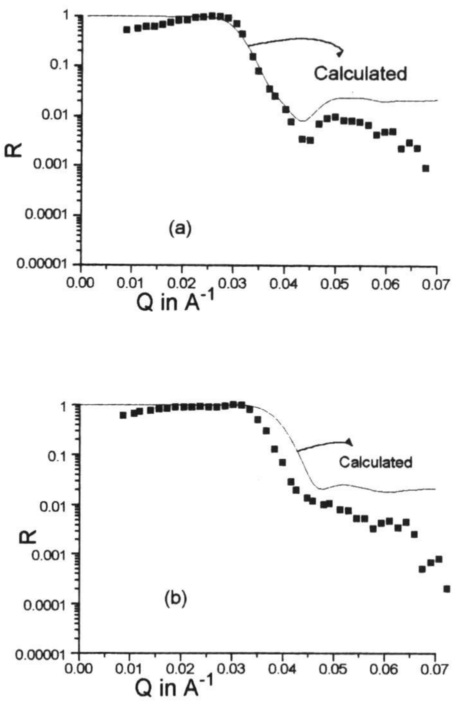

Fig.4. (a) The measured reflectivity, *R* (circles) vs. *Q* for the mirror with 10 bilayers and the ideal calculated profile (continuous line); (b) Measured *R* (circles) and ideal calculated (continuous line) for the mirror with 30 bilayers.

collimator and impinge on the sample. The collimator reduces the beam divergence to about 6 minutes of an arc. This amounts to a Q (momentum) resolution of about 0.006 $Å^{-1}$. The collimator is positioned on a *x*-displacement table with an accuracy of movement better than 10 microns, so that the neutron beam can be positioned at the centre of the sample table very accurately. The slit before the sample (slit 2, 0.4mm) is also placed on a similar displacement table, which is used to align the slits before the sample. There is another slit of 1mm width (slit 3) in front of the detector to reduce the background at the detector. This also restricts the overlap of the direct beam with the reflected beam during reflectivity measurement at the lowest possible angle. The sample table and the detector are moved in θ -2θ mode to scan the specular reflection profile. This attachment can measure wave vector transfer down to 0.008 $Å^{-1}$.

§.5. Results and discussion

The reflectivity profiles of both the supermirrors, measured in the above setup are shown in Fig.3. The mirror with 10 bilayers has a critical angle of 36 minutes of an arc and the mirror with 30 bilayers has a critical angle of 41 minutes of an arc. The expected ideal reflectivity profile as per the design thicknesses along with the measured reflectivities are also shown in Fig.4.

It is clear that increase in the number of bilayers has resulted in increase in the critical angle of total external reflection. The measured profiles of the mirror with ten bilayers are in good agreement with the calculated profile. The mirror with thirty bilayers has a critical angle lower than what is expected. This may happen due to several reasons. To point out a few: cumulative effect of deviation from the design thicknesses is larger in case of mirror with larger number of layers, interfacial roughness is also supposed to be more in the case of mirror with larger number of layers. The rapid fall in reflectivity beyond the total reflectivity region can be attributed to the surface and interface roughnesses of the deposited multilayers. The reflectivity of the mirrors below their critical angles are quite flat and we did not observe any sharp drops in reflectivity in this region for both the mirrors. The slow fall towards lower Q is known as "foot-print effect" and is understood to happen because of the fact that the sample area becomes smaller than the beam divergence at near zero incidence. We have not attempted any geometric correction of the data for this effect. The calculated ideal curves are the reflectivity profiles of the mirrors, computed using the design thicknesses and assuming ideally flat substrate. We have not tried any fit to the reflectivity profile to find out the bilayer thicknesses after deposition.

In conclusion the supermirrors fabricated using the said formalism have reflectivity profiles in good agreement with the calculated profiles. The mirror with thirty bilayers has a larger critical angle than the mirror with ten bilayers. The larger deviation from ideal profile in case of the mirror with larger number of bilayers may be due to cumulative deviations from calculated layer thicknesses and larger interface roughnesses.

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