RECENT PROGRESS IN SUPERMIRRORS AT PSI

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ABSTRACT

Recently, significant progress has been made in increasing the performance of supermirror coatings for neutron guide tubes. Using reactive DC-magnetron sputtering techniques we have developed at PSI a process for the series production of neutron guides with twice the critical angle of reflection of bulk Ni and with an integral reflectivity of more than 90% over a substrate area of 500 × 300 mm^2. For the optimization of the reflectivity, we have used various thin film measurement techniques, like atomic force microscopy, transmission electron microscopy and Auger depth profiling for the characterisation of the interfaces and surfaces, as well as standard X-ray and neutron scattering techniques. We shall also report on our first successful experience with the series production of supermirrors for the spallation source SINQ, which will be the first neutron source that relies almost entirely on the performance of supermirror coated guides.

I. INTRODUCTION

Neutron guide tubes are a means to efficiently transport neutrons from the source to spacious areas of low background [1]. Until now most guide systems rely either on a coating consisting of natural Ni which has an angle of total reflection for neutrons given by $\theta_c/\lambda = 0.00173$ rad/Å. Using ^{58}Ni this angle can be increased by a factor $m = 1.2$ [2], resulting in flux gains at the end of a guide of roughly 40%. Many years ago Turchin [3] and Mezei [4] proposed to use a sequence of thin artificial layers that increase the critical angle even further via Bragg reflection. Recently reflectivities R for such superstructures have been reported that exceed 0.95 at $m = 2$ [5] and $m = 3$ [6]. In fact a group is claiming to be ready for series production of supermirrors $m = 2$ with $R > 0.95$ [7]. Therefore, in addition to flux gains of a factor of four or more that can be achieved with supermirrors, it is now possible to transport even thermal neutrons with a reasonably large divergence (i.e. $\theta \simeq 0.4^0$ at $\lambda = 2$ Å) in an efficient way.

As anounced at the last ICANS meeting in 1990 [8] a commercial DC-magnetron sputtering facility has been installed during summer 1991 at Paul Scherrer Institute. After various acceptance tests and adjustments we started the development of supermirror coatings in January 1992. During this time it became clear that the anticipated configuration of the machine proved to be right, in particular, excellent multilayers can be produced when the layers are deposited with high power and at low process pressure [8].

In Fig. 1 we show the transmission of one of our better supermirror coatings ($m = 2$) sputtered on regular float glass. In a single reflection measurement performed at a TOF reflectometer in Saclay we obtain after deconvolution of the data a reflectivity close to $R = 0.94$ at twice the angle of total reflection of bulk Ni ($q = 4\pi m\theta_c/\lambda = 0.0435$ Å^{-1}). In contrast, when a measurement over the whole surface area is made using a micro-guide set-up (to be explained below), we obtain $R = 0.92$. Note that the neutrons have undergone roughly nine reflections near $m = 2$. The difference between the two results can be explained by small variations of R and m along the mirror surface (in particular at the beginning and at the end of the glass plate) for which the latter method is more sensitive to. In the following sections we describe the research that enables us to produce now such supermirror structures.

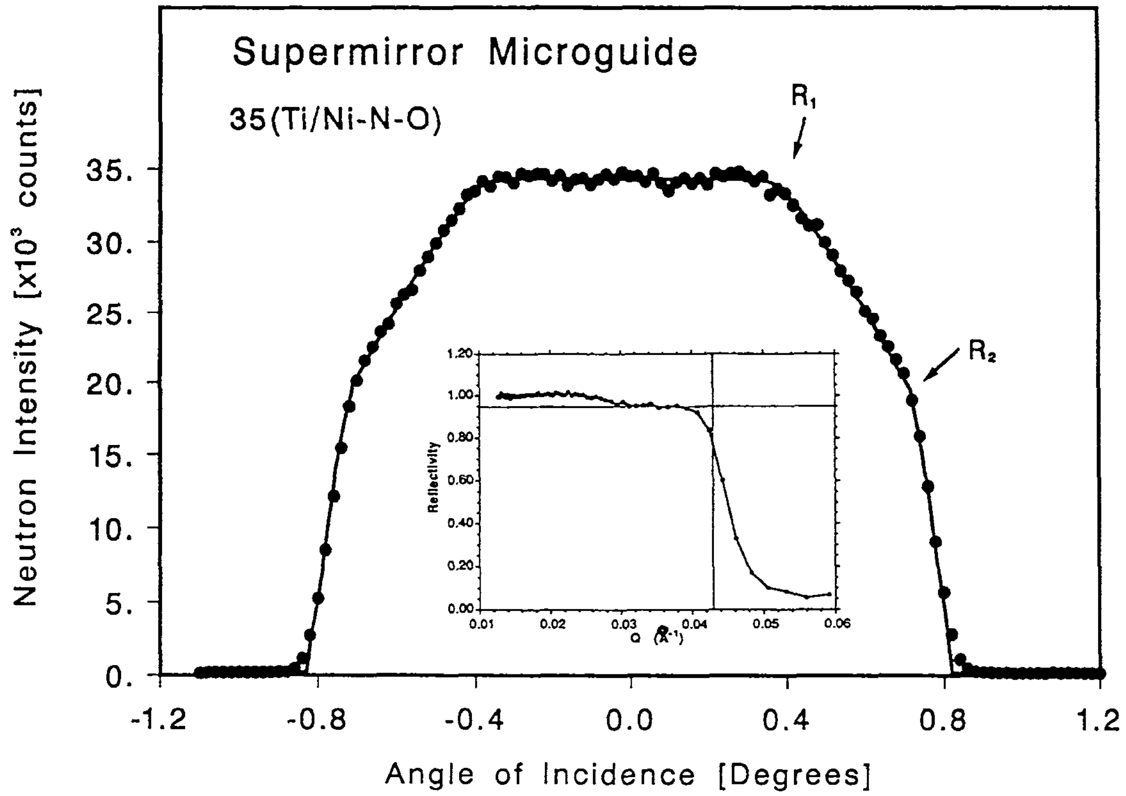

Fig. 1: Transmission profile of a micro-guide with a supermirror coating consisting of 35 Ni(air)/Ti bilayers yielding a reflectivity of 0.92. Inset: single reflection profile for same mirror, $R \simeq 0.94$ ($\lambda = 4.08$ Å).

II. EXPERIMENTAL

For the investigation of the morphology and the interface structure of the Ni/Ti based multilayers we have produced various samples with constant bilayer spacings (usually between 50 Å and 200 Å) as well as single layers with a thickness $1000 < d < 2000$ Å on silicon wafers or regular float glass by means of DC-magnetron sputtering. The stress S and d have been measured directly by means of a Tencor long scan profiler P2. Combining these results with mass density measurements using quartz the density of the layers can be determined.

Conductivity measurements prove to be particularly useful because they provide an integral characterisation of the properties of the layers. In particular, the conductivity does not only depend on the thickness of the layers, it also depends on the grain size and on the amount of impurities within the layers. Fig. 2 shows a comparison between conductivity and optical transmission of a sputtered 100 Å layer of $Ti_{72}V_{28}$ on glass. The two measurements are obviously correlated along the substrate length of 500 mm, indicating that the layers have not only a well defined thickness $d = 100$ Å$\pm$0.8%, they have also a very similar morphology. For comparison a pure Ti layer yields 165 Ω/sq.

Reflection profiles of the samples were measured on neutron reflectometers installed at the pulsed spallation source ISIS and at the Orphée reactor in Saclay using a fixed sample geometry, providing constant illumination during a wavelength scan. Most measurements at fixed wavelength have been performed on the double axis spectrometer P2ax and on the recently installed test spectrometer TOPSI (since Dec. 1992) at reactor Saphir at PSI. Usually two coated glass plates were assembled like a narrow guide with a gap of 0.4 mm (0.8 mm) for $L = 250$ mm (500 mm), where L is the length of the glass plates. This so-called micro-guide was inclined with respect to the incoming neutron beam ($\lambda = 4.08$ Å) and the transmitted intensity was recorded (Fig. 1). Near the critical edge of the supermirror the neutrons undergo up to 9 reflections.

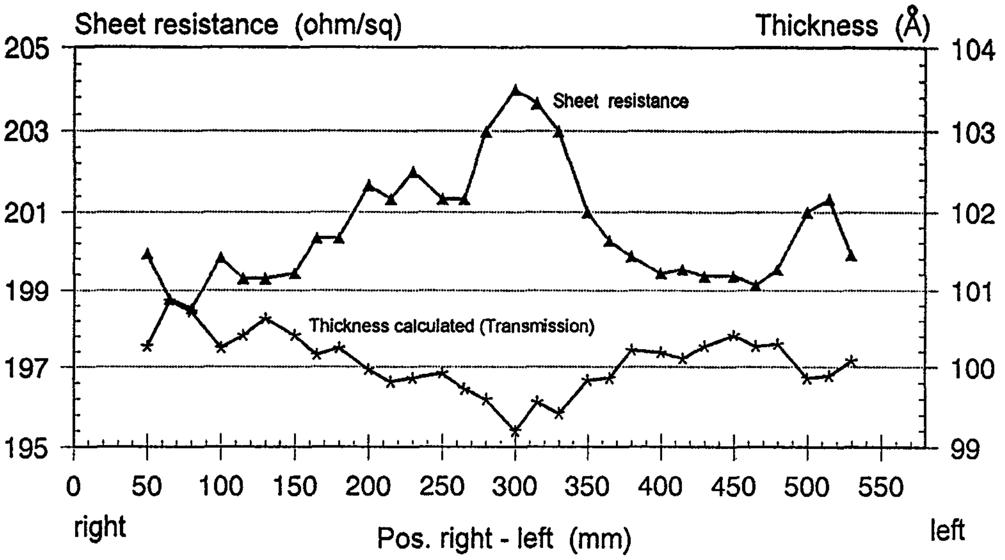

Fig. 2: Sheet resistance and thickness versus position of a sputtered film $Ti_{72}V_{28}$. Note the strong correlation between the two data sets.

III. RESULTS

It is known that multilayers consisting of pure Ti and Ni tend to develop rough interfaces possibly due to the large grains in the individual layers. In additon significant interdiffusion of Ni into Ti takes place in particular at elevated temperatures. In order to reduce these effects it has become common practice to add C to the Ni layers. This procedure does not only lead to an amorphization of the layers, it also increases the scattering density of the Ni layers. Values for bN of $9.6 \cdot 10^{-6}$ Å^{-2} [6] and $11.2 \cdot 10^{-6}$ Å^{-2} [5] have been reported. In order to further increase the contrast between the layers Vidal et al. [5] have hydrogenated the Ti layers using reactive RF magnetron sputtering. They claim that the scattering density for the TiH layers can be increased to $-7.8 \cdot 10^{-6}$ Å^{-2} which is much higher than the value for the stochiometric compound TiH_{2-x} ($Nb = -5.11 \cdot 10^{-6}$ Å^{-2}).

In a first step [9-11] we also produced multilayers consisting of these materials and observed that addition of C up to 12% reduces the size of the Ni grains (Table 1) and leads to amorphization of layers that are only a few nm thick and to reorientation of grains ([111] → [200]) in thicker layers. Further increase of C leads to the formation of Ni_3C grains and to amorphous layers for C contents $> 45\%$.

Hydrogenation of Ti in sufficient quantities leads to the formation of cubic TiH_{2-x} with [111] perpendicular to the surface. Coherent interfaces are formed if the thickness of the TiH_{2-x} layers does not exceed a few nm. The best interfaces are obtained with multilayers that contain either NiC_x/Ti or Ni/TiH_y exemplified by the appearance of superlattice peaks of order 11 at scattering angles $2\theta \simeq 11^0$ for CuKα radiation. The quality of the interfaces of NiC/TiH multilayers was inferior.

Based on our experience with NiC and TiH we have prepared some supermirror coatings NiC/TiH on regular float glass using a sequence of layers calculated with a program from J. B. Hayter and H. A. Mook [12]. After annealing the samples during 16 h at 120^{0}C the performance was $R = 0.900(2)$ at $m = 2.06(5)$ [10].

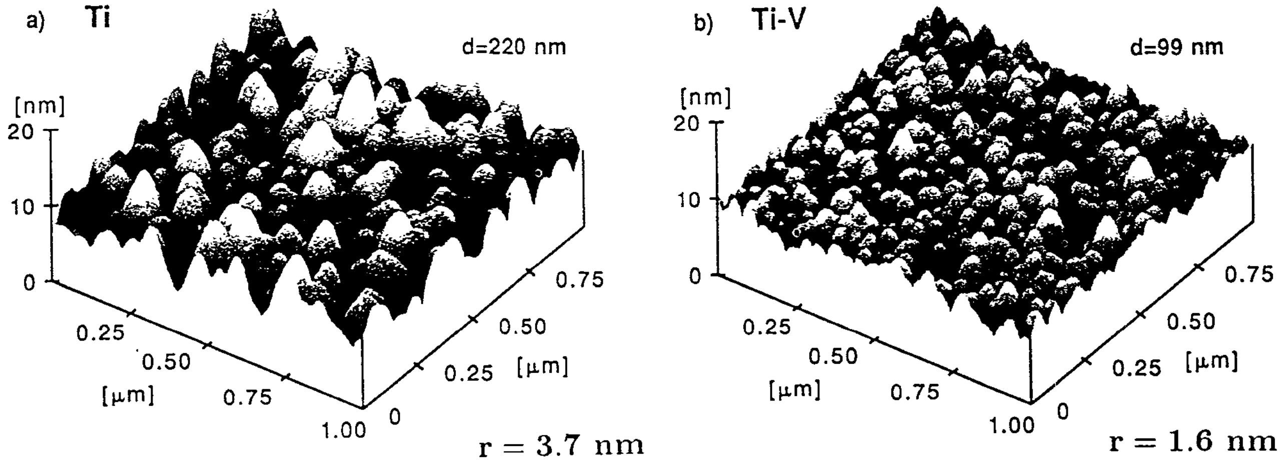

Fig. 3: Atomic force microscopy pattern of monolayers of a) pure Ti and b) $Ti_{72}V_{28}$

Several reasons have led us to give up the further exploration of supermirrors consisting of NiC and TiH:

- the quality of NiC layers depends on the way, how C is incorporated into the Ni target material, raising the question of maintaining the target quality
- NiC targets are very expensive
- hydrogen has a large absorption and incoherent scattering cross section for neutrons
- H_2 is not pumped well with turbo pumps due to the low molecular weight
- supermirrors must be annealed after production

There is not much choice for improving the structure of the Ti layers by incorporating other elements with a negativ scattering length because their number is limited (H, Li, V, Mn). Mn is known to diffuse [6], in addition it becomes activated under neutron irradiation. Li and Mn have a higher absorption than Ti. Therefore we incorporated V into Ti. Atomic force microscopy measurements [13] presented in Fig. 3a and 3b demonstrate clearly that the grain size and the amplitude of the roughness are significantly reduced in $Ti_{72}V_{28}$ when compared with pure Ti. It is interesting to note that Ni layers (RMS = 0.22 nm) are more than a factor of 10 smoother than Ti layers (RMS = 3.72 nm). The results compiled in Table 1 indicate the excellent resolution of the atomic force microscope. The roughness measurements over an area of 100×100 nm^2 yield in general too small values whereas samplings over larger areas are consistent with each other.

In a search for a good replacement of C in the Ni containing layers we decided to use N_2 which has even a higher scattering length than C. X-ray measurements show similar effects on the morphology of the layers on increasing the concentration of N_2 as for C. Again a reorientation Ni [111] $\longrightarrow$ Ni [200] takes place and the formation of NiN_3 is observed. However, an amorphisation and grain size reduction of Ni does not occur [11].

Table 1: Roughness and lateral extension of surface inhomogeneites measured with atomic force microscopy for different sampling areas. d is the thickness of the sputtered layers.

material	d (nm)	RMS (nm)			lat. ext. (nm)
		100 × 100 nm^2	500 × 500 nm^2	6 × 6 μm^2	
Si	0	0.14	0.11		flat
Ni	95	0.16	0.22	0.25	50
$Ni_{90}C_{10}$	210	0.17	0.36	0.34	30
$Ni_{45}C_{55}$	210	0.45	1.07	1.17	40
Ti	224	2.53	3.72	3.84	70
$TiH_{1.1}$	188	3.40	5.13	4.18	60
$Ti_{72}V_{28}$	99		1.6		30

Table 2 shows that NiN_x is an excellent and cheap material for increasing the critical angle of single layer coatings. Unfortunately, multilayers consisting of NiN_x/Ti are not stable at elevated temperatures possibly because N_2 diffuses into the Ti and builds the famous hard coating (or artificial gold) TiN, thereby reducing the contrast between the Ni and Ti containing layers.

Table 2: Critical angle of reflection in units of the critical angle θ_c of reflection for bulk Ni for different materials sputtered on borkron glass 500 × 150 mm^2.

material	Ni	NiN_x	$Ni_{87.1}N_{7.7}O_{2.4}$
m	0.99	1.03	1.03

Based on the observation of Schärpf [14], that venting an evaporation process after completion of individuel layers improves the performance of supermirrors, we replaced N_2 by dry air [15]. We expected that O_2 builds up a diffusion barrier TiO_2 at the interface. Indeed, the performance of the mirrors remained excellent and most importantly, the structures were stable up to $T \simeq 260^0$C (see Table 3). The highest reflectivity we obtained with this recipe on regular float glass was 92.5% at $m = 2$ when measured using the micro-guide set-up. Atomic force microscopy shows that the Ni-N-O surface is actually very smooth [13].

Table 3: Temperature dependence of the performance of a supermirror sputtered on regular float glass 250×50 mm^2. R is the reflectivity at the critical angle of the supermirror $\theta = m\theta_c$. θ_0 is the angle of total reflection of the top layer.

$T(^0)$C	20	180	220	240	260	270
θ_0 (mrad)	1.38	1.36	1.40	1.29	1.28	1.00
m	2.00	2.00	2.04	1.94	2.01	2.07
R	0.90	0.89	0.87	0.86	0.85	0.80

IV. DISCUSSION

The main advantages of supermirrors made of Ni(air)/$Ti_{1-x}V_x$ [15] are their thermal (and therefore expected long term) stability and their cheap and simple production. In April 1993 the series production of supermirror coatings for the SINQ guides began and presently we produce in one machine run two borkron glass plates 500×150 mm^2 in less than six hours. The coatings are characterized immediately after production using the micro-guide set-up at TOPSI. The parameters R and m are measured at three different heights (-40 mm, 0 mm, 40 mm) of the guide.

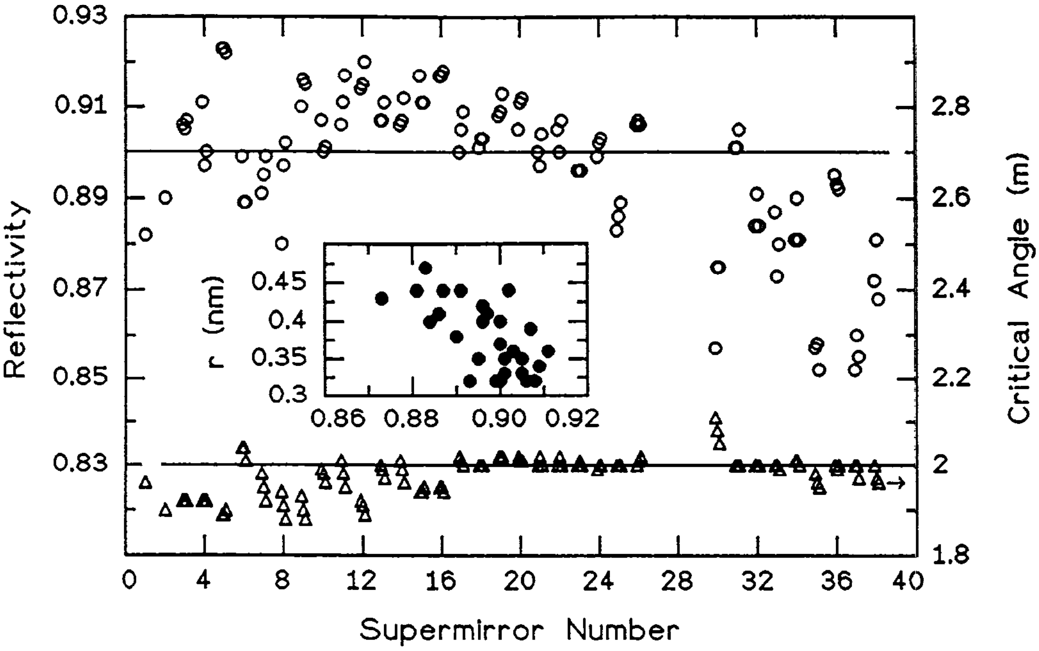

Fig. 4: Reflectivity (circles) and critical angle (triangles) of all supermirror coatings produced so far at PSI. Mirrors 17 to 26 have been produced within 2.5 days using an identical recipe. Note the drop in R of sample 25. The inset shows roughness versus reflectivity for the samples produced with an identical recipe.

Fig. 4 shows R and m for *all* runs we have done in the period April 26 until May 20, 1993. After some adjustments of the sequence at the beginning of production we have now settled at $m = 2$ and $R \geq 0.90$ for substrates with a surface roughness (RMS) $r \simeq 3.5$ Å (runs 17-26). There is a trend that R decreases rather dramatically if $r > 4$ Å(runs 25, 32 - 38). Such dramatic fluctuations in R have never been observed when using commercial float glass. We hope that we can routinely obtain a reflectivity $R > 0.9$ when we are provided with borkron glass with $r < 3.5$ Å, or (intrinsically smooth) float glass, resulting in flux gains at SINQ [16] of a factor of three when compared with a Ni coated guide. An improvement from $R = 0.90$ to $R = 0.95$ would increase the flux only by another 20%.

One of the major problems we still have to resolve is the build-up of waveness in the mirror sequence. We have observed that the reflectivity at the critical edge $m \simeq 2$ increases by $\simeq 0.02$ if the thick bilayers are left out, whereas R decreases only by $\simeq 0.01$ if 230 thinner layers are added yielding $R = 0.83$ at $m = 2.7$. We believe that the wavy parts of the supermirror reflect neutrons out of the incident beam (in wrong directions) that are supposed to be reflected by the thin layers.

Based on the scattering densities of the layers and the known roughness of the borkron glass $r \simeq 0.4 \pm 0.1$ nm [17] we have calculated R for our supermirror sequence [18] and obtain $R \simeq 0.96$ at $m = 2$. Absorption and incoherent scattering has been taken into account. This result indicates, that we loose roughly 0.02 to 0.04 because of imperfections of our mirrors.

Finally we would like to compare the performance of the supermirrors made at PSI with those, made by other groups. Vidal et al. [5] produced supermirrors composed of 49 layers NiC/TiH and obtained $R = 0.95$ near twice the critical angle of Ni. We honestly do not understand how they manage to obtain this result. In Fig. 5 we show the theoretical reflectivity for their sequence with and without taking absorption, incoherent scattering and an overall roughness $r = 4$ Å into account. The theoretical reflectivity at $m = 2$ is only $\simeq 0.92$. Indeed the recently installed supermirror neutron guide at the ORPHEE reactor in Saclay has only a reflectivity $r = 0.77$ at $m = 1.9$ [19]. Ovonic Synthetic Materials Company of Troy, Michigan have made supermirrors with $m = 2.77$ [20] and $R > 0.95$ [6]. Assuming a layer roughness $r = 4$ Å, one obtains for the ideal supermirror (no losses) $R \simeq \exp(-(Qr)^2/2) = 0.97$. Taking into account absorption, incoherent scattering and waveness, that amounts to at least 0.02, it is indeed surprising for us that such high quality mirrors can be produced.

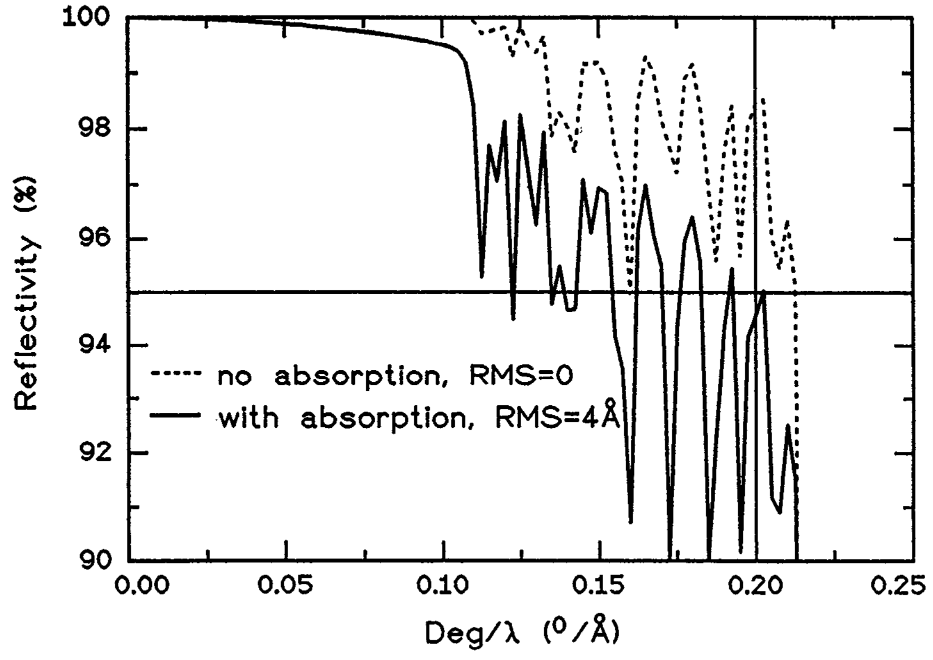

Fig. 5: Calculated reflectivity profiles for supermirrors of Ref. [5,7]. The following scattering densities have been used: $g_{NiC} = (11.2 \cdot 10^{-6} - 2.46 \cdot 10^{-9} i)$ Å^{-2} and $g_{TiH} = (-6.00 \cdot 10^{-6} - 12.4 \cdot 10^{-9} i)$ Å^{-2}.

Concluding we remark that we are able to produce supermirror coatings reproducably on surface areas of at least 500×300 mm^2 on super-polished borkron glass ($r < 3.5$ Å) with $m = 2$ and $R > 0.90$ (see Fig. 4). The process can be upgraded in a straight forward way to much longer guide pieces. From our experience we learn, that absorption and incoherent scattering in the layer materials and the roughness of the substrate are very important parameters which are decisive for the performance of the supermirrors. During series production it is advisable to immediately measure R and m using a micro-guide set-up in order to garantee an optimum performance of the neutron guide to be produced. The production is most easily done on commercially available float glass with a deposition system located close to a neutron scattering center using DC-magnetron sputtering with materials Ni(air)/TiV [15]. We hope that it will be possible to finish the major part of the deposition process for $m = 2$ supermirror guides for SINQ until the end of 1993. Afterwards we are going to concentrate on the development of other neutron optical components like polarizers and focussing units.

Acknowledgment

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