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MORPHOLOGY OF GLASS SURFACES: INFLUENCE ON THE PERFORMANCE OF SUPERMIRRORS

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ABSTRACT

Multilayer structures are often used for guiding, filtering, shaping, and monochromatizing electromagnetic radiation or particle beams. In order to assess the importance of the properties of the substrate on the performance of multilayers we have investigated glass substrates by means of atomic force microscopy, profilometry, and neutron- and x-ray reflectometry. Whereas the waviness of polished glass is excellent, when compared to float glass, the surface roughness of polished glass is significantly larger than for float glass. We find that the performance of supermirror coatings depends as essentially on the surface morphology as on the loss mechanisms that occur in the multilayer itself.

1. Introduction

Artificial multilayers are widely used for optical applications in neutron and x-ray scattering. In order to obtain a high performance of the devices it is necessary to reduce interface roughness and interdiffusion to the lowest possible values. During the production of supermirror coatings for the neutron guide system of the spallation source SINQ [1] we realized that the surface morphology of the glass substrates has also a strong influence on the reflectivity of supermirror coatings.

In visible light optics, the individual layers of the coatings have a thickness d of the order of the wavelength of light, i.e. $\lambda \simeq 5000$ Å. Hence a roughness [2] of the substrate $r_s \simeq 10$ Å does not impede the performance significantly. In contrast, for neutrons and x-rays, d is typically of the order of 10 to 700 Å, hence r_s must be very small in order to obtain useful coatings. For most applications the waviness of the substrates must also be small.

Before going into more detail, we should like to explain the term supermirror. Neutrons can be guided to the neutron spectrometers in hollow glass tubes by total internal reflection, similarly as light in a wave guide. The most suitable material is Ni, because it has the largest angle of total reflection, θ_c, of all naturally occuring materials. It is given by $\theta_c^{\mathrm{Ni}} = c\lambda$. Here $c = 0.099°/\text{Å} = 1.73$ mrad/Å. The glass plates must have a small waviness w and their alignment must be excellent because θ_c is so small. Typical neutron guides are more than 50 m long and have cross sections of the order of 120 mm by 50 mm.

Keywords: Supermirror, Glass, Roughness, Neutron Guide

In order to increase the neutron flux at the instruments, it has been suggested [3] to extend θ_c to $m\theta_c^{\mathrm{Ni}}$ ($m > 1$) by using interference coatings consisting of thin layers of Ni and Ti that reflect neutrons according to Bragg's law for $\theta_c > \theta_c^{\mathrm{Ni}}$. Such coatings are called supermirrors and have been developed at various places [4]. For $m = 2$ mirrors we shall achieve at SINQ flux gains of more than a factor of three for neutrons with $\lambda \simeq 4$Å [5].

As an example, we show in Fig. 1 the reflectivity R (squares) and the critical angle in units of θ_c^{Ni}, m (triangles), for a sequence of 8 successive production runs of supermirror coatings for the spallation source SINQ. They consist of 82 layers $\mathrm{NiN}_x\mathrm{O}_y/\mathrm{TiV}_z$ [1, 6]. The two pairs of polished (boron containing) glasses 50 × 500 mm^2 (open symbols) and the pair of float glass 50 × 500 mm^2 (filled symbols) were coated at the same time. The measurements have been done by determining the transmission of neutrons (λ = 4.08 Å) through a microguide that was assembled from a pair of plates [1, 6]. It can be seen that the sputtering process is very reproducible and that the reflectivity of supermirrors on polished glass is significantly ($\simeq$ 5%) lower than of supermirrors on float glass. Obviously, the performance of supermirrors is strongly affected by the substrate.

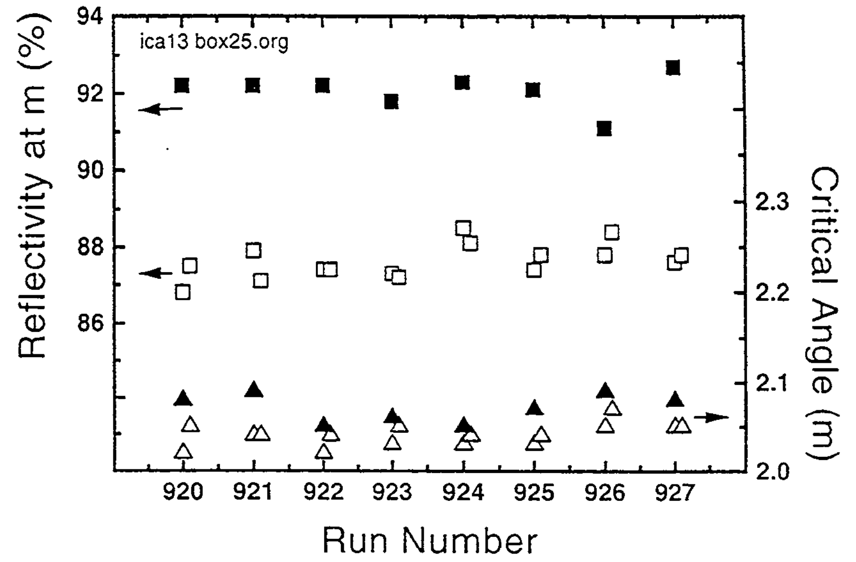

Figure 1: *Reflectivity (squares) of supermirrors $m \simeq 2$ at the angle of total reflection (triangles). θ_c is given in units of θ_c^{Ni}. The substrates are regular float glass ($d = 2$ mm, filled symbols) and polished glass (open symbols).*

In order to assess the influence of the substrate on the reflection properties of $\mathrm{NiN}_x\mathrm{O}_y/\mathrm{TiV}_z$ supermirrors we have investigated the surface of different glasses and coatings by means of x-ray diffraction, atomic force microscopy, and profilometry. In addition, we have measured the reflectivity of supermirror coatings by means of neutron reflection. Reflectivity losses due to imperfections of the multilayer structure itself have already been discussed in the literature [4]. Our results indicate clearly that the reflection losses due to non-perfect substrates can be as large as the losses caused by the imperfections of the coating.

2. Experimental

Atomic force microscopy (AFM) measurements on glass plates (500 × 150 mm^2) for the neutron guides of the spallation source SINQ have been performed on a Digital Instruments NanoScope III Large Sample Scanning Probe Microscope. Small samples have been investigated on a Park Scientific Instruments Autoprobe CP microscope. The microfabricated Si_3N_4 stylus tips have a diameter of $\simeq$ 100 Å, yielding a lateral resolution $l_r < 100$ Å. The AFM's were used in the tapping mode in order to overcome the electrostatic forces.

The grazing angle x-ray diffraction measurements were performed on a Philips X'Pert MPD diffractometer at PSI and on a reflectometer at HMI. From a comparison of the Q-dependence of the reflected intensity, $R(Q)$, with the reflectivity R_0 of an ideal surface we extracted the root mean square roughness r_s under the assumption of a Debye-Waller type behavior

$$W(r_s) = \frac{R(Q)}{R_0} = \exp\left(\frac{-(Qr_s)^2}{2}\right). \tag{1}$$

Such measurements yield an average of the surface roughness over the illuminated area of the sample.

The waviness w of the substrates was determined by means of a Wenzel 3-d coordinate measuring machine. In a first step the surface profile $z(x, y)$ was measured along four different rows with an accuracy of ± 1 μm yielding 188 data points per glass side. In a second step the surface normal in every point was determined by calculating the derivative of z with respect to x and y. We defined w to be the standard deviation of the surface normals from their mean value.

The surface properties of the float glass [7] and the extruded and fire-polished glass [8] were not specified by the supplier, whereas the roughness of the polished glass [9] was specified on the basis of interferometric measurements performed by means of a WYKO profilometer. We designate this number by r_w. Clearly r_w is expected to be smaller than r_s as determined by x-ray reflection or AFM, because of the coarse lateral resolution of the optical method.

3. Results

3.1 Roughness

The AFM patterns in Fig. 2 show that the surface profiles of float glass (thickness d = 10 mm) and polished glass are very different. Float glass consists of large smooth areas with $r_s < 3.7$ Å [10] and spots with crater-like damages having an amplitude of $\simeq$ 150 Å. They are caused by outgassing during solidification of the glass. The roughness over the measured area 25 × 25 $\mu\mathrm{m}^2$ is $r_s = 16.4$ Å and is mostly determined by the localized damages (Table 1). Therefore the Debye-Waller approach for $R(Q)$ (Eq. 1) cannot be applied. We find that the reflectivity R of supermirror coatings decreases with an increasing number of damages, leading us to qualify float glass as being "good", if $R > 90\%$ and "bad", if $R < 88\%$.

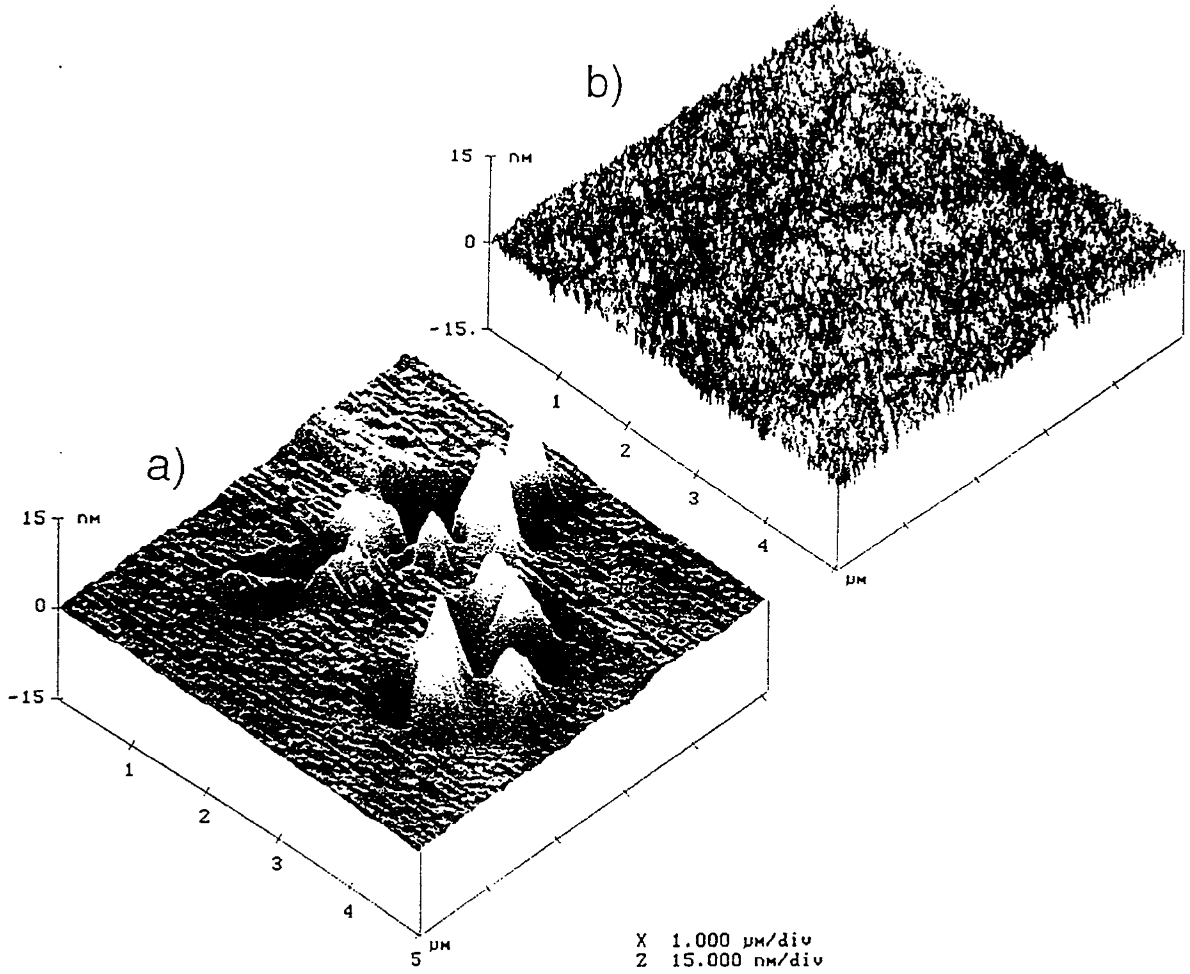

Figure 2: a) *AFM profile of a ”damaged“ part of float glass (d = 10 mm). The average distance between the damages is typically $\simeq$ 10 µm. b) AFM profile of polished, boron containing glass (d = 15 mm).*

In contrast, the polishing technique gives rise to a homogenous, rough surface. It contains scratches with an amplitude of about 25 Å from the polishing procedure. The roughness $r_s \simeq 8.4$ Å is about twice as large as the interferometric values, $2.5 \leq r_w \leq 5.0$ Å. Often, but not always, we find a clear correlation between r_w, as quoted by the supplier, and the reflectivity of supermirror coatings at $m = 2$ [1] (Fig. 3).

The roughness of various glass surfaces has been characterized also by means of x-ray reflection. ”Good“ float glass yielded $r_s \simeq 4$ Å for the side that floated on the tin bath during production and $r_s \simeq 9$ Å for the non-floated side [11]. Similar measurements performed at PSI on polished glass yield $r_s \simeq 9$ Å. These values compare well with the AFM results, i.e. $r_s < 3.7$ Å for the smooth areas of float glass and $r_s \simeq 8.4$ Å for polished glass. Therefore, x-ray reflection is mostly sensitive to the smooth areas of float glass, whereas the damaged areas may be considered as non-reflecting spots.

We have also found that the surface morphology of float glass depends on the thickness of the glass. All the investigated samples with $d \geq 8$ mm had crater-like

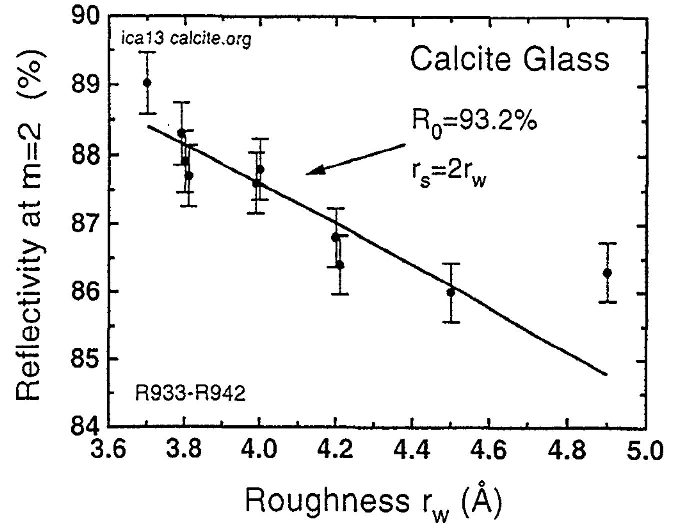

Figure 3: *Dependence of the neutron reflectivity on the surface roughness of polished calcite glass coated with supermirror $m = 2$. The solid line is a calculation as explained in the text.*

damages, whereas float glass and extruded fire-polished glass with $d \leq 2$ mm was essentially smooth (Fig. 4). The highest reflectivities have always been obtained with coatings on thin float glass.

The above results indicate that the different type of roughness between float glass and polished glass is responsible for the different reflectivity of the supermirror coatings. If we assume that our coatings have a reflectivity $R_0 \simeq 93\%$ on an ideal substrate, then we can estimate the influence of the roughness of the substrate on R on the basis of Eq. 1. If we set $r_s = 2r_w$, as discussed above, then we obtain the solid line in Fig. 3 that roughly follows the trend of the reflectivity data for supermirrors on polished glass.

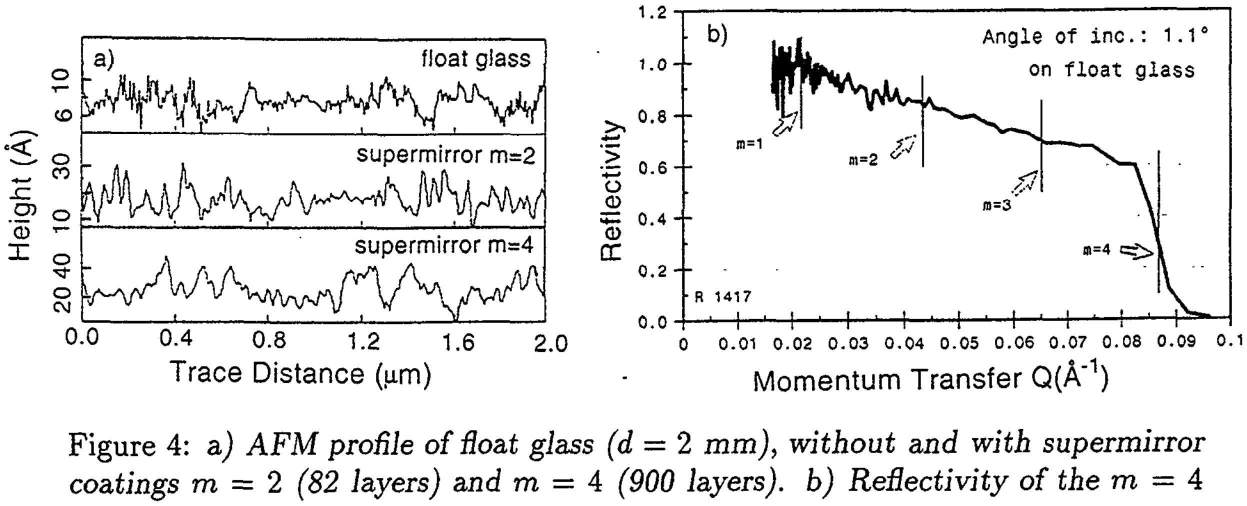

Figure 4: a) *AFM profile of float glass (d = 2 mm), without and with supermirror coatings m = 2 (82 layers) and m = 4 (900 layers). b) Reflectivity of the m = 4 supermirror.*

Finally, we measured the surface profile of supermirrors deposited on float glass (Fig. 4a). r_s increases slowly with an increasing number of layers, reaching $r_s = 7.8$ Å for $N = 900$ layers and a thickness of the coating $d_c = 4.5$ μm (Table 1). The reflectivity of the $m = 4$ mirror is shown in Fig. 4b. Above the critical angle of reflection of

Table 1: *Roughness and reflectivity measurements.* $W(r_s)$ *is given for* Q *at* m *times the critical angle of Ni* [12]. R_c *is the calculated reflectivity, including roughness, absorption and incoherent scattering,* R_{exp} *is the measured reflectivity.*

sample	d(mm)	N	$d_c(\mu\text{m})$	r_s(Å) (from AFM)	$W(r_s)$	R_c	R_{exp}
extruded glass	0.21	0	0	1.5 ± 0.5			
float glass	2	0	0	1.6 ± 0.5			
with $m = 2$	2	82	0.9	4.4 ± 1	0.98	0.96	0.92
with $m = 3$	2	450	2.8	not measured			0.79
with $m = 4$	2	900	4.5	7.8 ± 1.5	0.80	0.72	0.60
float glass	10	0		$3.7 \pm 1^{\spadesuit}/16.4 \pm 3^{\heartsuit}$			
with $m = 2$	10	82	0.9	$5.2 \pm 1^{\spadesuit}$	0.97	0.95	0.90
polished glass	15	0		$8.4 \pm 2^{\diamondsuit}$			
with $m = 2$	15	82	0.8	8.6 ± 2	0.93	0.91	0.87
with $m = 3.3$	15	450	2.8	not measured			0.64

$^{\spadesuit}$ smooth area, 7.5×7.0 μm^2 (R957)
$^{\heartsuit}$ whole surface, 25×25 μm^2
$^{\diamondsuit}$ area 5×5 μm^2

bulk Ni ($m = 1$) R decreases to $\simeq 85\%$ at $m = 2$, finally reaching $R \simeq 60\%$ at $m \simeq 4$. At $m = 2$, R is only 2% lower than R of an $m = 2$ supermirror on polished glass (Fig. 1), indicating again that it is also the roughness of the substrate and not only the coating itself, that limits the reflectivity of mirrors. However, the low value $R_{\text{exp}} = 0.60 < R_c$ can only be understood if interdiffusion and roughness between the layers is taken into account. Interdiffusion is particularly important for the thinnest layers that have a thickness of only $\simeq 35$ Å.

3.2 Waviness

In real neutron guides losses occur not only due to imperfections of the coatings and the substrates. Losses occur also due to a possible misalignment of the guide sections and due to the waviness of the coated glass plates. Polished glass is usually specified to have $w < 0.1$ mrad. To put this number into perspective, a neutron guide with a supermirror coating $m = 2$ has $\theta_c = 6.9$ mrad $= 0.4°$ for neutrons with $\lambda = 2$ Å. Therefore the maximum loss per reflection due to waviness is 1.5%. For a Ni-coating it is 3%.

In order to assess the usefulness of float glass for the fabrication of neutron guides we have used profilometric measurements to determine the waviness of an arbitrary batch of 60 glass plates having a thickness $d = 10$ mm and a surface area 150 mm by 520 mm. Typical surface profiles are shown in Fig. 5a. The undulations on the front and the back side are correlated and their periode is $\simeq 250$ mm. Fig. 5b shows that the distribution of w for the same glass plate can be well described by a Gaussian distribution having a standard deviation $\sigma = 0.13$ mrad, i.e. a Gaussian with a HWHM of 0.16 ± 0.02 mrad.

A summary of the results from all 60 plates is given in Fig. 5c, and it shows that one half of the glass plates have a waviness $w < 0.2$ mrad. This corresponds to $\simeq 3\%$ of θ_c for $m = 2$ and $\lambda = 2$ Å, i.e. to a loss of less than 3% per neutron

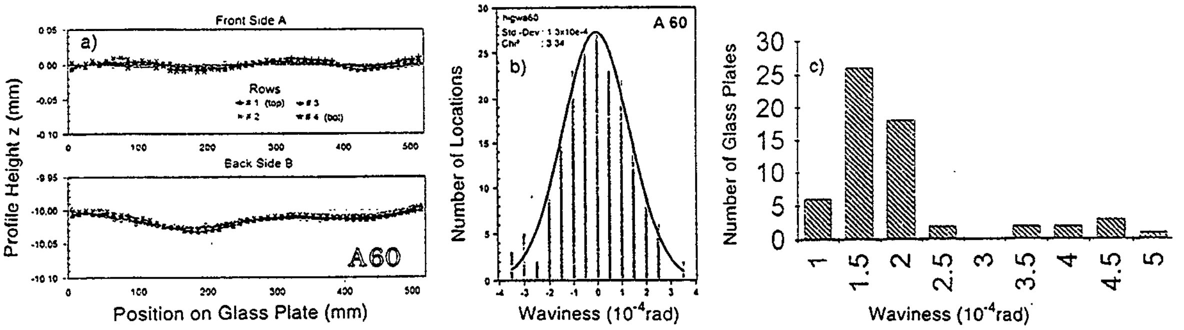

Figure 5: *a) Profile of glass surface. b) Distribution of waviness. c) Number of occurencies of glass plates with a certain waviness.*

reflection. These losses are partly compensated by those parts of the surface that have an opposite slope. The losses can be reduced simply by increasing θ_c by $2w$, i.e. by 6% to $m = 2.12$.

4. Conclusions

The very different surface morphology of polished glass and float glass has important implications for their use as substrates for neutron optical devices. Polished glass may be useful for applications that require high geometrical precision and moderate m, for example thermal neutron guides. For applications with large m one should avoid polished glass because of the exponential decrease of $R(Q)$ (Eq. 1) with increasing m. Hence, for focussing devices, float glass seems more favorable due to the small roughness and because the waviness is of minor concern for large reflection angles. At present the maximum angles of reflection for the coatings from our laboratoy are $m \simeq 4$. The reason being that the mechanical stress in the supermirror coatings is rather large, i.e. $\sigma \simeq 3 \cdot 10^9$ dyn/cm^2. For $d_c \simeq 4.5$ μm the force of the coating exceeds the mechanical strength of the glass and it fractures. This effect is more pronounced in boron containing glass [13].

Finally we should like to compare the reflectivity losses caused by substrate imperfections with the losses caused by imperfections of the coating. The maximum reflectivity, $R_{\rm exp}^{\rm max}$, for an $m = 2$ supermirror we have ever achieved until now on float glass, is $94 \pm 0.5\%$ at $\theta_c = 2\theta_c^{\rm Ni}$ [6]. The design reflectivity R_d was 99.3% [14]. Losses occur (see Table 2) due to absorption, $\Delta_a = 0.7\%$, incoherent scattering, $\Delta_i = 0.7\%$, and due to roughness of the substrate, $\Delta_s^f \simeq 1.3\%$ ($r_s \simeq 3.7$ Å). The losses due to waviness can be neglected because the mirrors were mechanically constrained to be flat. Hence we obtain a theoretical reflectivity $R_c = 96.6\%$. We have neglected the losses due to Bragg scattering. Therefore, the losses due to interdiffusion and roughness at the interfaces of the coating account for $\Delta_{\rm sm} = R_c - R_{\rm exp}^{\rm max} \simeq 2.6 \pm 0.5\%$. Obviously, $\Delta_{\rm sm}$ is comparable or even smaller than the losses Δ_s and Δ_w caused by the substrate itself.

In conclusion, we point out that with today's sputtering techniques it is possible to grow multilayers that are so excellent, that the properties of the substrate become

Table 2: *Loss mechanisms of an $m = 2$ supermirror with a design reflectivity $R_d =$ 99.3%. The losses due to waviness decrease with increasing λ and m.*

mechanism		R / loss	comments
absorption	Δ_a	0.7%	Ni and Ti
incoherent scattering	Δ_i	0.7%	Ni and Ti
geometrically perfect superm.	R_c	96.6%	$R_c = R_d - \Delta_a - \Delta_i - \Delta_s^f$
maximum measured R	$R_{\text{exp}}^{\text{max}}$	$94.0 \pm 0.5\%$	Ref. [6]
interdiff., interface roughness	Δ_{sm}	$2.6 \pm 0.5\%$	$\Delta_{\text{sm}} = R_c - R_{\text{exp}}^{\text{max}}$
roughness of polished glass	Δ_s^p	$\simeq 6.4\%$	$r_s = 8.4$ Å
waviness of polished glass	Δ_w^p	$\simeq 1.5\%$	$\lambda = 2$ Å, $w = 0.1$ mrad
roughness of float glass (10 mm)	Δ_s^f	$\simeq 1.3\%$	$r_s = 3.7$ Å
waviness of float glass (10 mm)	Δ_w^f	$\simeq 3\%$	$\lambda = 2$ Å, $w = 0.2$ mrad

a major concern. The ideal substrate should i) have a low roughness, ii) be flat, iii) be mechanically strong, and iv) be affordable. For very large m the layer thicknesses become rather small and interdiffusion and stress must be minimized. Here, new sputtering techniques combined with smoothing and stress-releasing layers may improve the performance of multilayers by another few percent.

5. Acknowledgements

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[2] Roughness designates surface undulations with a length scale of the order of a few thousand Å or less. Waviness is a measure for surface undulations with a length scale of a few micrometers up to meters or more. Usually it is characterized by reflecting a laser beam from the surface.

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[9] CILAS, Compagnie Industrielle des Lasers, F-91460 Marcoussis, France.

[10] 3.7 Å is an upper limit because the large glass plate was very susceptible to the vibrations of the building. The measurements were performed on the tin bath side.

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[12] One may argue that the roughness r_s of the glass substrate should be used for calculating the Debye-Waller factor at $m\theta_c^{\mathrm{Ni}}$. However, we believe, that it is more appropriate to use r_s of the coating: The upper layers become progressively more rough, thus scattering neutrons in non-specular directions. Therefore, some neutrons that are supposed to be reflected by the layers close to the substrate do not arrive there.

[13] On float glass ($d = 2$ mm), $m = 4$ coatings are stable between 2 days and several months and $m = 3.6$ coatings last for half a year or more; $m = 4$ coatings on boron containing glass started to peal off immediately after coating.

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