Neutron Optics: Towards Applications for Hot Neutrons

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Abstract. Supermirrors with large critical angles of reflection, i.e. large index $m$ are an essential ingredient to transport, focus and polarise neutrons over a wide range of energy. Here we summarise the recent developments of supermirror with very large critical angles of reflection and high reflectivity that were conducted at SwissNeutronics as well as their implementation in devices. Approaching critical angles $m = 8$ times the critical angle of natural nickel makes new applications possible and extends the use of reflection optics towards the regime of hot and epithermal neutrons. Based on comparisons of simulations with experiment we demonstrate future possibilities of applications of large-$m$ supermirrors towards devices for neutrons with short wavelength.

1. Introduction

The introduction of the concept of supermirrors in 1967 [1] and its realisation [2,3] laid the foundation to increase the efficiency for the transport of neutrons by internal reflection in guides and the manufacturing of polarising mirrors by accepting reflection angles larger than the total reflection angle of Ni. In 1989, Hayter and Mook published a formalism to compute an adequate layer sequence for supermirror [4]. Using this algorithm, the supermirror technology was applied exclusively for the first time for the complete guide system at the Swiss Spallation Source Sinq at the Paul Scherrer Institute (Switzerland) using mirrors with $m = 2$ [5]. In passing we note that the critical angle for supermirror is given by $\theta_c = 0.099m\lambda$. Here the maximum angle of reflection $\theta_r$ is given in degrees and the wavelength $\lambda$ in Å. The following years witnessed enormous developments for neutron guides combining new guide concepts, e.g. ballistic guides [6] and non-linearly tapered parabolic and elliptic guides [7] with supermirrors whose performance has been improved continuously [8]. Moreover supermirrors with high reflectivity on metallic substrates [9] have enabled new applications namely extending neutron guides close to the moderator and for beam focusing based on Montel optics thus improving the performance of beam lines for neutron scattering and imaging further. As a result of the continuous developments of the deposition techniques for supermirrors and the super-polishing of substrates on large areas, non-magnetic and polarising supermirrors are available with large $m$-values, excellent reflectivity and high polarization. Here we report about the state-of-the-art of these supermirrors and their possible use towards applications for hot neutrons.

2. Performance of non-magnetic supermirrors

2.1. Preparation of supermirrors

Atomically smooth substrates and layers are required to obtain a high reflectivity of neutron supermirrors. Dedicated processes are developed for the super-polishing and coating of glass, silicon,
and metallic substrates on large areas of the order of $350 \times 500 \text{ mm}^2$. We probe the substrate surface by atomic force microscopy (AFM) using a desktop instrument easyScan 2 AFM from Nanosurf. Figure 1 shows the AFM image of a typical surface scanned on an area of $10 \mu\text{m} \times 10 \mu\text{m}$. The RMS roughness is $\sigma < 1 \text{ Å}$. This quality is regularly obtained on glass, silicon and metallic substrates, and is required for the excellent reflectivities presented below.

The supermirrors are coated using DC magnetron sputtering. The process is highly sophisticated in order to obtain layers with extremely low roughness and low internal stress. The thicknesses of the single layers are computed using the formalism by Hayter and Mook [4]. Non-magnetic supermirrors comprise a combination of materials of nickel-molybdenum alloy and titanium.

2.2. Reflectivity of supermirrors
The reflectivity of all supermirrors is measured at the reflectometer NARZISS at SINQ (PSI), which is operated in an angular dispersive mode at a wavelength of $\lambda = 5 \text{ Å}$ [10]. Figure 2 shows a compilation of measurements of supermirrors with $2 \leq m \leq 8$. The slope of the reflectivity is similar for all $m$-values, e.g. supermirrors with $m = 2$ and $m = 8$ have essentially the same reflectivity at $m = 2$. The observation of an almost constant slope is remarkable because when fabricating a supermirror with $m = 8$, more than 16500 thin layers for $2 < m \leq 8$ are deposited first before the layers for the reflection at $m = 2$ are coated. The high reflectivity at $m = 2$ demonstrates that the growth mode of the layers does not change, i.e. the interface roughness remains essentially constant. As a result, supermirrors with large $m$-values do not compromise the performance of the supermirror for intermediate and small $m$-values. Therefore, the increase of the $m$-values leads to a true gain of intensity due to the increased divergence of the neutron beam.

Figure 1. Surface image of a super-polished substrate measured with AFM. The RMS roughness over the scan area of $10 \times 10 \mu\text{m}^2$ is $\sigma < 1 \text{ Å}$.

Figure 2. Reflectivity profiles of supermirrors with $m$-values $2 \leq m \leq 8$. The slope of $R$ is largely independent of the $m$-value indicating a stable interface roughness, which is essentially constant for all layers.
The supermirror with \( m = 8 \) shows a pronounced decrease of the reflectivity in the range \( 7 \leq m \leq 8 \). For a proper understanding we have conducted computations of the reflectivity. The good agreement between the model calculation of the supermirror sequence with \( m = 8 \) and the reflectivity data (Figure 3) indicates that the physics of the deposition process is well understood. As we do not want to disclose the details of the coating process, we simply summarise the essential features of the model as follows: The reflectivity was calculated using the matrix formalism from [11]. The real and imaginary (absorption, incoherent scattering) parts of the scattering length densities for Ni and Ti were taken from the literature [12]. The roughness at the interfaces was assumed to attain the value as measured at the surface of the substrate by x-ray reflectivity and was taken into account in the simulations by a static Debye-Waller factor. In addition, the non-constant yield of the sputter targets due to the consumption of the materials was taken into account.

The results in Figure 3 show that absorption (compare black-dotted and red lines) causes a significant reduction of the reflectivity of about 10% at large \( m \)-values. The deviation of the sputter yield of the target from its nominal value during the very long sputter runs leads to significant deviations of the thickness of the layers from the sequence calculated by Hayter and Mook [4]. The change in deposition rate is clearly visible at very large \( m \) leading to the observed drop in reflectivity. In future runs this effect can be compensated by adjusting the parameters of the sputter process. Note also the small dip in reflectivity around \( m = 7.9 \). It is caused by an incomplete single layer due to a software error during the run. The dip is reproduced in the simulation by eliminating layer #15701 from the layer sequence.

2.3. Long-term stability of supermirror

As shown above, the performance of present-day supermirror [13] is excellent up to \( m = 8 \). This raises the question about the long-term stability of the supermirrors. The comparison of the reflectivity of the very first supermirror with \( m = 6 \) as produced by SwissNeutronics in 2006 with measurements conducted in the following years as shown in Figure 4 demonstrates the perfect stability of the mirror sequence. Obviously, the measured reflectivity does not change. Visual inspections show neither a degradation nor a delamination of the supermirror.
2.4. Applications of supermirror with large \( m \)-value

Supermirrors with large \( m \) allow reflecting neutrons at large momentum transfers \( Q \), i.e. at large angles of reflection even for beams with short wavelength. The large-\( m \) supermirrors are typically required for the focusing of neutrons and for their efficient transport by means of non-linearly tapered guides. Using various optical concepts such as parabolic or elliptic focusing guides or even Montel geometries [14,15], significant gains in intensity can be realised because the brilliance transfer [16,17] can be improved. Indeed, the installation of an elliptic neutron guide for the powder diffractometer HRPD at ISIS has witnessed an intensity gain of up to two orders of magnitude [18] when compared with the previous conventional guide system. Increasing the \( m \)-value of its coating from \( m = 3 \) to \( m = 8 \) would extend the wavelength range towards the hot regime, i.e. \( \lambda \approx 0.3 \text{ Å} \).

In addition, the use of focusing mirrors allows concentrating neutron beams at the sample position [19]. Therefore, small samples can be investigated even under extreme conditions because the scattering from the sample environment can be strongly reduced leading to an improved signal-to-noise ratio. Various experiments with cold neutrons demonstrate the benefit of focusing optics [20,21,22]. Supermirrors with large \( m \) will be essential for an efficient operation of the planned beam lines at the new European Spallation Source ESS as they allow an efficient extraction of the neutrons from the moderator and the possibility to focus the neutrons while maintaining a large brilliance transfer [17]. In all applications the large \( m \)-values enable also the handling of neutrons with higher energies, e.g. hot and even epithermal neutrons.

3. Performance of polarising Fe/Si supermirrors

3.1. Reflectivity, transmission and polarisation of magnetised Fe/Si supermirrors

Polarising Fe/Si supermirrors are prepared using the same sputtering facility as for the non-magnetic supermirrors (section 2.1). Spin-dependent neutron reflectivity and transmission measurements are performed at the reflectometer NARZISS at SINQ (PSI). Figure 5 shows the spin-dependent reflectivity \( R \) and the polarisation \( P \) of a polarising supermirror Fe/Si with \( m = 5.5 \) coated on borosilicate glass with a thickness \( t = 0.3 \text{ mm} \). The reflectivity for the spin-up state is \( R > 70\% \) at the critical edge and \( P \approx 1 \) across the entire supermirror regime. Spin-down neutrons are reflected below \( m < \approx 0.7 \) because of the mismatch between the nuclear and magnetic scattering length density of Fe.
Figure 5. Spin-dependent reflectivity $R$ and polarisation $P$ of a supermirror Fe/Si with $m = 5.5$. $R > 70\%$ at the critical edge and $P \approx 1$ for the entire regime of the supermirror. For $m < 0.5$ the sample is over-illuminated.

Figure 6 shows the spin-dependent transmission and the polarisation from a Si wafer that is coated on both sides with polarising supermirror Fe/Si with $m = 5$. The thickness of the Si wafer is 0.3 mm. Below $m \approx 0.5$ the effects of over-illumination at grazing incidence are visible. For $m < 0.7$ both spin-states are totally reflected leading to $P = 0$. Within the supermirror regime the transmission for the spin-down neutrons increases while it is essentially suppressed for the spin-up neutrons. The polarisation is close to unity for $1 < m < 2.5$ and decreases to 0.91 at $m = 5$. The decrease of $P$ is a result of the finite reflectivity of the supermirror, which leads to the slight increase of the transmission for the spin-up neutrons near $m = 5$. The average polarisation within the supermirror regime is $P \approx 0.96$.

Figure 6. Spin-dependent transmission and polarisation of a polarising supermirror Fe/Si with $m = 5$ coated on both sides of a Si wafer with a thickness $t = 0.3$ mm. The average polarisation is $P \approx 0.96$.

3.2. Applications of polarising supermirror with large $m$-value

Polarising supermirrors are an essential ingredient for a large number of polarising devices such as V-cavities, single and S-shape curved benders, reflectors, and wide angle analysers. In particular V-cavities became a very convenient concept for polarising neutron beams. V-cavities can be arranged parallel and/or in series to shorten the device and/or increase the polarisation, respectively [23]. Figure 7 shows spin-dependent neutron transmission measurements of a V-cavity comprising 11 channels.
Each channel is equipped with 2 Vs in series coated with polarizing supermirror \( m = 5 \) allowing to polarise neutrons with a lower critical wavelength \( \lambda_c = 2.1 \) Å. The transmission is measured as a function of the lateral position across the entrance of the cavity (\( W = 74 \) mm), i.e. the cavity is translated across a well-collimated neutron beam with a wavelength of \( \lambda = 5 \) Å. Pronounced features of the dividing walls and the overlaps of the Si wafers at the tips of the Vs are observed because the beam is blocked there. The average transmission is 26% (100 unpolarised neutrons in – 26 polarised neutrons out). The average polarisation is \( P = 0.99 \). To simulate the performance of the V-cavity we have written a dedicated component for the Monte-Carlo simulation tool McStas [24,25] that takes into account various features of the cavity such as the measured spin-dependent reflectivity of the double-sided coated Si wafers, absorption by the coating and the Si wafers, and the precise arrangement of the Si wafers namely the overlap at the V-tip and the intrusions at the sides of the channels. Figure 8 shows the transmission \( T \) of the spin-down neutrons. All features are precisely reproduced. The simulated value \( T = 29\% \) is slightly higher than measured. The reason for the difference is because the model does not yet consider refraction of the neutron beam, which increases the path length of the neutrons in the material and thus the absorption. An analytic estimation of the effect of refraction increases the absorption by approximately 2.5\%. The excellent agreement between measurement and simulation proves that polarising devices can be easily designed for any anticipated application.

![Figure 7. Spin-dependent transmission and polarisation of an 11-channel double-V-cavity as measured across the width of the cavity.](image1.png)

![Figure 8. Simulated transmission of an 11-channel double-V-cavity as function of the lateral position across the width of the cavity.](image2.png)

The presently available polarising mirrors with \( m = 5.5 \) lead to large critical angles of reflection even for neutrons with short wavelengths. These mirrors are ideally suited for analysing the polarisation of thermal neutrons over large angular ranges. Recently, such polarising analysers were realised for the time-of-flight spectrometers HYSPEC at SNS and POLANO at J-PARC using remanent supermirror FeCoV/TiN \(_x\) [26] and Fe/Si, respectively. They allow analysing neutrons with energies as large as approximately 30 meV. The optimisation was performed using McStas.

4. Conclusions
We have shown that with modern sputtering technologies it is possible to control roughness and interdiffusion of multilayers on an atomic scale for heterostructures with the number of layers exceeding \( N = 16000 \). The thinnest layers for supermirror with \( m = 8 \) have a thickness \( t \approx 18 \) Å which is comparable with the lattice constant of novel materials in condensed matter physics. Moreover, the mechanical properties, i.e. the stress can be essentially reduced to zero thus preventing the coatings from delamination. Last but not least the coatings are radiation resistant. As the critical angle of
reflection $\theta_c$ for Ni/Ti supermirror is directly proportional to $m$ and the number of required layers is approximately given by $N = 4m^4$ [27]. $N$ increases dramatically with increasing $\theta_c$. Our model calculations indicate that it is the absorption that essentially limits the number of layers. As the physical properties of Ni and Ti are given, a reasonable upper limit may be around $m = 10$. Clearly, it depends on the specified reflectivity to be obtained. Using a combination of $^{58}$Ni and $^{62}$Ni [1] will lead to a significant decrease of $N$ due to the very large scattering contrast between the two isotopes thus allowing the production of supermirror with $m$ exceeding 10. The upper limit of $m$ for polarising Fe/Si supermirror may also be significantly increased because monocrystalline Fe develops the full moment already for a layer thickness $t < 10$ Å. It has to be evaluated at what minimal $t$ the Fe-layers develop monocrystalline grains in a sputtering process. Anyway, the limit will be above $m = 6$.

Obviously, the availability of supermirror $m \approx 8$ facilitates the transport and focusing of hot neutrons because reasonably divergent beams can be handled. For example, for $\lambda \approx 0.3$ Å (900 meV) neutrons with a divergence $\alpha \approx 0.5^\circ$ can be processed. Using non-linear tapering, $\alpha$ can be increased further. It is indeed the availability of large-$m$ supermirror in combination with elliptic geometries [7] and metallic substrates [9] that will make the efficient use of hot and epithermal neutrons at spallation sources feasible.

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