Comparison of specularly reflecting mirrors for GRANIT

V.V. Nesvizhevsky\textsuperscript{a,\,*}, G. Pignol\textsuperscript{b}, K.V. Protasov\textsuperscript{b}, G. Quemener\textsuperscript{b}, D. Forest\textsuperscript{c}, P. Ganau\textsuperscript{c}, J.M. Mackowski\textsuperscript{c}, Ch. Michel\textsuperscript{c}, J.L. Montorio\textsuperscript{c}, N. Morgado\textsuperscript{c}, L. Pinard\textsuperscript{c}, A. Remillieux\textsuperscript{c}

\textsuperscript{a}Institute Laue-Langevin, 6 rue des Martyrs, Grenoble 38042, France
\textsuperscript{b}LPSC, Université Joseph Fourrier Grenoble 1, CNRS/IN2P3, Institut National Polytechnique de Grenoble, France
\textsuperscript{c}LMA, Université Claude Bernard Lyon 1, CNRS/IN2P3, Villeurbanne, France

Received 27 April 2007; received in revised form 11 May 2007; accepted 18 May 2007
Available online 2 June 2007

Abstract

The specularity of ultracold neutron reflection was compared for different “promising” surfaces, including sapphire, silica, silica with carbon (diamond-like), and copper coatings with very small roughness. The probability of total losses of ultracold neutrons (UCN) from a specular trajectory was dominated by diffusive (non-specular) elastic scattering of UCN. In all the cases considered the quality of reflection was sufficiently high for storage of UCN at specular trajectories for the first stage of GRANIT experiment.

\textsuperscript{c} 2007 Elsevier B.V. All rights reserved.

PACS: 03.65.--w; 28.20.--v

Keywords: Ultracold neutrons; Specular reflection; Coatings

1. Introduction

The experimental study described here was prompted by experiments on the gravitationally bound quantum states of neutrons in the Earth’s gravitational field \cite{1}, and, in particular, by the GRANIT project to measure the resonance transitions between gravitationally bound quantum states of neutrons \cite{2,3}. The precision of these experiments is defined mainly by the uncertainty of the energy $\Delta E_n$ of a neutron in a given quantum state (with number n); this is proportional to the reciprocal storage time $t_{\text{st}}^n$ of neutrons in this quantum state, as follows from the uncertainty principle $\Delta E_n = \hbar / t_{\text{st}}^n$. The storage time $t_{\text{st}}^n$ of neutrons in the quantum states is severely limited by the finite probability of specular reflection of UCN at mirrors. This is an important criterion in the selection of mirrors for the GRANIT experiment. The second criterion is the value of the critical velocity of the mirror (or its coating): this should be as high as possible in order to improve the otherwise extremely low statistics in these experiments.

This explains our decision to perform a detailed study of the specularity of UCN reflection at surfaces, which could be used for the GRANIT in the future: sapphire, silica, silica with carbon (diamond-like) coating, and silica with copper coatings. The choice of these mirrors was supported by the following arguments: (1) The critical velocity value is irrelevant for the bottom mirrors. It would in any case be higher than the vertical component of the neutron velocity, which is equal to a few centimeters per second. Silica with no coating has too low a critical velocity and could therefore be used for the bottom mirror only. (2) The critical velocity value is extremely important for the vertical wall mirrors. It defines the total number of trapped neutrons. Diamond-like carbon or copper (isotope) coatings have very high critical velocity values. (3) If the coated mirrors were to fail, a compromise could be achieved by using sapphire, which has an intermediate critical velocity value. The mechanical hardness of sapphire offers important advantages from a practical point of view.

The following requirements had to be met for the preparation and use of the mirrors: the roughness of the silica mirrors (with/without coatings) had to be sufficiently low to rule out any diffusive scattering of neutrons at the
surfaces due to their geometrical roughness; the roughness of the sapphire mirror had to approximate the values achievable with the large sapphire mirrors (30 cm by 30 cm) of the GRANIT experiment; the flatness of the mirrors had to be sufficient to rule out any loss of neutrons from specular trajectories due to surface waviness; the thickness of the coatings (carbon, copper) had to be sufficient to provide a high probability of UCN reflection, but not excessive (in order to minimize deformation of the mirrors by stresses in the coatings); the presence of hydrogen in the coatings had to be avoided as it could increase UCN loss probability (due to inelastic scattering); dust had to be avoided on all surfaces as it could increase UCN scattering from specular trajectories; the standard requirements for experiments with UCN had to be satisfied.

The neutron measurement methodology used was identical to that in Ref. [4]. It allows investigation of the probability of non-specular reflection, even if it is as low as a few times $10^{-4}$ per collision. The procedure for preparing the mirror sample was optimized for experiments with neutrons and their parameters were measured using non-neutron methods.

2. The sample mirrors

The sample mirrors measured $50 \times 50 \times 10 \text{ mm}^3$. The pairs of fused silica mirrors (without any coating, with carbon, and copper coatings) are shown in Fig. 1. All the substrates were produced by General Optics (GSI Group).

The surface quality for all silica mirrors was measured using a defect detection technique developed by LMA. The relative area of the defects ranged between $4 \times 10^{-7}$ and $10^{-6}$ for silica surfaces with/without coatings.

The roughness of their reflecting surfaces was found to be equal to 0.5–0.8 A RMS for all silica mirrors with/without coatings and 10–15 A for sapphire mirrors.

The flatness of the non-coated silica mirrors was equal to 60–80 nm, of the copper coated mirrors equal to 100 nm, and of the carbon coated mirrors equal to 220 nm (the constraints of the carbon coating are estimated to be $\sim 2 \text{ GPa}$); the flatness of sapphire mirrors was equal to 200 nm.

The thickness of the carbon and copper coatings was equal to 0.2 mm. The choice of deposition technique for the carbon coating had to exclude chemical vapour deposition, which, because it uses methane, may introduce hydrogen into the carbon layer. Two different techniques were investigated: RF magnetron sputtering, and ion beam sputtering (IBS). The former technique generated a low deposition rate and weak layer adherence. Ion beam sputtering was therefore chosen. The RF magnetron sputtering technique was used for copper deposition.

The adhesion of the carbon layer was checked using the Grittington test. After 1 h of testing the layer showed no scratches and good adhesion. Longer testing time (8 h) showed scratches in the layer, indicating that the IBS carbon layer is not as hard as the CVD DLC layer. Nevertheless, the adhesion and hardness of the IBS carbon layer seemed sufficient for the GRANIT experiment. A “Scotch test”, only, was used for the copper layer. No decohesion of the layer has been observed.

Once the mirrors had been produced and their surfaces cleaned they were stored in a clean room at LMA. To avoid any oxidation of the copper layer the corresponding

Fig. 1. From left to the right: two silica mirrors without coating, two silica mirrors with carbon coating; two silica mirrors with copper coating.
mirrors were placed in a vacuum. There was nevertheless some degradation observed in the coatings after a few months. This can be considered an integral part of the present test: the conditions of the coatings will be reproduced in the GRANIT experiment. Any degradation in the coating over a period of several months (the typical time span for neutron experiments) is not acceptable; copper coatings should not be chosen. No change was found in the carbon coatings, thus favouring the choice of carbon coating for the GRANIT experiment.

3. Measurement methodology

The measurement was made with the gravitational spectrometer used for studying the gravitationally bound quantum states of neutrons [5]; the method of measurement was identical to that used in Ref. [4]. The UCN beam produced in the gravitational spectrometer had a spread of vertical velocity components of only $\pm 0.1 \text{m/s}$. It was transmitted through a long narrow slit between two identical, vertical, parallel plates as shown in Fig. 2. The angle between the plates and the initial neutron beam axis was equal to $\sim 30^\circ$. The measurements of the total UCN flux through the vertical slit between two polished equivalent plates as a function of slit size revealed very low total loss of neutrons for all the surfaces studied (see Fig. 3). These losses were due to absorption, inelastic scattering of neutrons, and diffusive scattering; the first two loss channels are expected to be negligible compared to diffusive scattering. The theoretical dependence could be described as follows: $F(\Delta x) = z\Delta x(1 - \mu)/\Delta x$ where $F(\Delta x)$ is the UCN flux as a function of slit size $\Delta x$, $\mu$ the total loss probability per collision, $\phi$ the angle between the initial beam direction and the plane of the plates, and $z$ the normalization coefficient.

4. The experimental results

The results are presented in Fig. 3. The measured total loss coefficients are equal to $(0.5 \pm 0.2_{\text{stat}} \pm 0.2_{\text{syst}}) \times 10^{-3}$ for silica; $(4.0 \pm 0.4_{\text{stat}} \pm 0.2_{\text{syst}}) \times 10^{-3}$ for diamond coating
on silica; \((6.1 \pm 0.6_{\text{stat}} \pm 0.2_{\text{syst}}) \times 10^{-3}\) for sapphire; and 
\((15.9 \pm 4.0_{\text{stat}} \pm 0.3_{\text{syst}}) \times 10^{-3}\) for copper coating on silica.

During the measurements all the mirrors were kept in a small portable clean room and installed in the neutron spectrometer with no break in the clean chain. The neutron measurement was free from any significant systematic effects. Thus, the neutron detector background was negligible compared to the characteristic neutron count rate; mirror positioning using precision translation plates was accurate to a few micrometers; the maximum uncertainty in the installation of the “zero” distance between the two mirrors, as well as that in the parallelism of the mirrors was equal to a few micrometers (these positioning uncertainties cause corresponding systematic uncertainties in the values presented for the total loss coefficients). The systematic and the statistical uncertainties in the values presented for the total loss coefficients are approximately equal to each other; the total accuracy of the measurement cannot be significantly improved. The accuracy is sufficient however for measuring the non-zero values of the total loss coefficients for almost all the mirrors (except for silica); it does not constraint the upper limit for these values. The precise value of the total loss probability for the silica mirrors is of minor importance—it cannot be used for the vertical walls anyway, because of the low critical velocity. Thus, all the important parameters of the mirrors to be studied were measured.

5. Conclusion

The quality of the specular reflection of UCN at all mirrors was found to be sufficiently high to store UCN at specular trajectories in the first stage of the GRANIT experiment. The total loss coefficients measured were equal to 
\((0.5 \pm 0.2_{\text{stat}} \pm 0.2_{\text{syst}}) \times 10^{-3}\) for silica; 
\((4.0 \pm 0.4_{\text{stat}} \pm 0.2_{\text{syst}}) \times 10^{-3}\) for diamond coating on silica; 
\((6.1 \pm 0.6_{\text{stat}} \pm 0.2_{\text{syst}}) \times 10^{-3}\) for sapphire; and 
\((15.9 \pm 4.0_{\text{stat}} \pm 0.3_{\text{syst}}) \times 10^{-3}\) for copper coating on silica.

The lowest loss coefficient \((0.5 \pm 0.2_{\text{stat}} \pm 0.2_{\text{syst}}) \times 10^{-3}\) was measured on the silica mirror. Silica would therefore seem to be a good choice for the bottom mirror (the critical velocity value is not important in this case). The UCN incidence angle in the GRANIT experiment will be much smaller than that in the present test; the total loss coefficient will therefore be even smaller than the value presented here. The disadvantage of silica is its poor mechanical hardness compared to sapphire, for instance. In order to avoid damage of the mirror edges, we should avoid any movable parts in the assembly of the mirrors in the GRANIT experiment.

The loss coefficient for sapphire \((6.1 \pm 0.6_{\text{stat}} \pm 0.2_{\text{syst}}) \times 10^{-3}\) was higher than that measured in Ref. [4] due to a surface roughness \((\sim 14\,\text{A})\) higher than that in Ref. [4] \((\sim 7\,\text{A})\); this was as expected. The difficulty of producing and polishing large-size mirrors (> 30 cm) to a roughness level of \(\sim 7\,\text{A}\) or better will probably prevent us from being able to use them for GRANIT. At the roughness level considered, surface roughness is clearly the main reason for UCN loss from a specular trajectory.

The relatively large coefficient of UCN total loss at copper coatings \((15.9 \pm 4.0_{\text{stat}} \pm 0.3_{\text{syst}}) \times 10^{-3}\) does not allow their use for GRANIT. In addition to that, the stability of the coating over time does not appear to be sufficient for long-term experimentation (over several years).

The small probability of UCN loss from a specular trajectory of \((4.0 \pm 0.4_{\text{stat}} \pm 0.2_{\text{syst}}) \times 10^{-3}\) at carbon (diamond-like) coatings makes these attractive for use on the vertical walls of the GRANIT trap, given their high critical velocity. This would allow us to increase considerably the number of UCN trapped. One should mention, however, that the loss coefficient is significantly higher than for silica mirrors with an equivalent surface roughness. This means that UCN loss with carbon coatings is determined (most probably) by the inhomogeneity of the coating density, rather than by the “geometrical” surface roughness. The “effective” carbon coating roughness is of the order of \(\sim 1\,\text{nm}\).

One should note that, although the results obtained are promising, the angular resolution of the present measurement is not sufficient to constrain quasi-specular reflections absolutely. Only the quantum trap of the GRANIT experiment allows such precision.

Acknowledgement

We are grateful to the French Agence Nationale de la Recherche (ANR) for supporting this project.

References