Deuterated polyethylene coatings for ultra-cold neutron applications

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Abstract

We report on the fabrication and use of deuterated polyethylene (dPE) as a coating material for ultra-cold neutron (UCN) storage and transport. The Fermi potential has been determined to be 214 neV and the wall loss coefficient $\eta$ is $2 \cdot 10^4$ per wall collision. The coating technique allows for a wide range of applications and new possibilities in this field of physics. In particular, flexible and quasi-massless UCN guides with slit-less shutters and slit-less UCN storage volumes become possible. These properties enable the use in next-generation measurements of the electric dipole moment of the neutron.

Keywords:

1. INTRODUCTION

Ultra-cold neutrons (UCN) have velocities below $\sim 7$ m/s or few 100 neV kinetic energy. Their properties are rather specific, making them a unique tool in fundamental physics. Gravity causes a change in potential energy of $1.02 \cdot 10^{-7}$ eV per meter height, the magnetic moment is $|\mu_n| \sim 6 \cdot 10^{-8}$ eV/T and the interaction with the strong interaction causes repulsion from many material surfaces [1]. Due to the large wavelength of UCN, $\lambda > 50$ nm, the wave function experiences an average coherent interaction with materials, resulting in a wave-like reflection on a potential well with only little probability for absorption. The so-called Fermi pseudo potential [2, 3] consists of a real and an imaginary part $V_F = V - iW$:

$$V_F = \frac{2\pi\hbar^2}{m_n} Nb$$  \hspace{1cm} (1)

$$W = \frac{\hbar^2}{2} N\sigma v.$$  \hspace{1cm} (2)

Here, $m$ and $v$ are the neutron’s mass and velocity, respectively, $N$ is the atomic number density of the wall material, $b$ is its bound coherent scattering length and $\sigma$ is the loss cross-section. The real part of $V_F$ determines the height of the wall potential and the ratio $\eta = W/V$ the loss coefficient, which is related to the angle-averaged loss probability per wall collision

$$\mu(E) = 2\eta\left[\frac{V_F}{E} \sin^2\left(\frac{1}{2V_F} - \left(\frac{V_F}{E} - 1\right)^{1/2}\right)\right],$$  \hspace{1cm} (3)

with $E$ the kinetic energy. For $E < V_F$, UCN are reflected from a surface under any angle of incidence. $V_F$ reaches values up to 250 neV for beryllium or diamond-like carbon surfaces [5].

One of the flagship experiments with UCN is the search for the electric dipole moment of the neutron [4] and in most approaches in the past [6] as well in next-generation experiments (e.g. [7]), UCN are confined in a trap at room temperature. The statistical sensitivity of the EDM experiment is

$$\sigma_d = \frac{\hbar}{2\alpha ET \sqrt{N}},$$  \hspace{1cm} (4)

with a quality parameter $\alpha$, an applied electric field $E \sim 20$ kV/cm, the storage time $T \sim 250$ s and $N$ the number of detected neutrons at the end of the experiment. Here, $V_F$ is important due to the typical spectra
of UCN from the source, which is comparable to the low-energy tail of a Maxwellian distribution. Thus, a larger initial number of neutrons in the camber can be trapped. A low loss coefficient $\eta$ and low outgassing rates enables long $T$ and thus also a large $N$.

In the simultaneous presence of $E$, highly electrically insulating behavior and resilience against electrostatic field breakdowns is required. Previously, the coating for the insulating ring of the UCN storage chamber has been realized by bulk quartz with $V_F = 95$ neV [6, 8] or deuterated polystyrene [5] with $V_F \approx 160$ neV. In addition to a relatively low $V_F$, the materials suffer from problems with water contamination of the surfaces or large outgassing in vacuum without prior baking processes. Both of these contaminations lead to UCN losses due to upscattering on hydrogen or interactions with the residual gas. Further, to obtain a large quality parameter $\alpha$, the spin-flip probability per wall collision must be small ($\sim 10^{-6}$, for typical behaviors of materials see e.g. [9, 10, 11]).

Being a plastic insulator, fully deuterated polyethylene with a calculated scattering length density (SLD) $N \cdot b$ of $8.323 \cdot 10^{-6}$ Å$^{-2}$ (216.7 neV) seems a viable option for such a material, despite its rather unpleasant processing properties.

Our work describes the development of a technique to produce deuterated polyethylene (dPE) coatings or large surfaces and experimental tests of this material.

2. COATING PROCEDURE

The general idea of the coating procedure is to dissolve dPE powder in a deuterated solvent (d-xylene), wet the surface to be coated with the solution and let the solvent evaporate. Polyethylene in general only dissolves at temperatures around 145°C, which is also very close to its melting point. Hence, the procedure has to take place inside an oven. A schematic view of the coating apparatus for cylindrical glass tubes is shown in Fig. 1. It allows us to coat the walls of glass tubes with diameters of 80-125 mm and lengths of up to 500 mm. The tubes are cleaned thoroughly by hand with isopropanol and acetone before the coating procedure to remove any dirt on the surface. Afterwards, the tubes are closed on both ends with Teflon caps, since Teflon is chemically inert when exposed to xylene. The tubes are then baked at 100°C for about an hour to remove any remnants of the cleaning agents. The required amount of deuterated polyethylene powder (Polyethylene-d4 98 Atom%D, SigmaAldrich) and deuterated xylene (xylene-d10, ARMAR Chemicals) is filled into the cylinder through a hole in one of the end caps. The entire assembly is then mounted inside the oven in such a way that it is freely rotatable around the cylinder axis. On one side, it is connected to the axle of a stepping motor, on the other side there is an opening that allows us to flush the setup during the coating procedure and recover the evaporated solvent. The oven is closed and heated to approximately 145°C. After the dPE powder has dissolved completely, the tube starts rotating with a speed of a few rotations per minute. Typically, a few large scale structures of melted dPE appear on the walls, these get washed out and disappear as they repeatedly pass through the solvent puddle that is in the bottom of the tube. The rotation is continued until all large scale structures have disappeared and it is apparent that the wall is wetted homogeneously with the solution. The volume is then flushed with nitrogen to remove the evaporated xylene from the system. The xylene re-condenses in a tube and is recovered for later use. Once all of the solvent is gone, the heating is turned off and the system can cool down. After the system has cooled down to room temperature, the tube is removed from the system. Fig. 2 shows a tube with 500 mm length and an inner diameter of 115 mm that has been coated with a layer of about 10 µm of dPE, forming a homogeneous opaque layer. Next to samples of the presented size, also rings for EDM-type UCN storage
chamber have successfully been coated with this robust procedure, including e.g. coating around the edges of the ring.

Depending on the substrate and the coating thickness, the dPE coatings show a quite different behavior: although all coatings tend to stick well to the surfaces, it was possible to remove whole coatings from smooth glass tube substrates without damage. For rough (optically opaque) quartz substrates and thinner (< 5 µm) coatings, removal of the coatings was only possible using the inverse coating procedure with hot solvents.

3. SAMPLE CHARACTERIZATION

3.1. Vacuum performance

The coated tubes were placed into a 0.5 m³ vacuum chamber and pumped down into the 10⁻⁷ mbar range. Analysis of the outgassing of an 80 mm tube gave rates on the order of 10⁻⁸ mbar l/s cm⁻² for the entire vacuum system. There was no marked difference when measuring with a 120 mm tube despite the increased dPE coated surface, indicating that the outgassing was dominated by the vacuum chamber and the rate due to dPE-coated walls is lower than 10⁻⁸ mbar l/s cm⁻².

3.2. Surface roughness

To obtain a characteristic dPE sample, the coating was removed from one tube, resulting in a 10 µm thick foil. An atomic force microscope measurement was performed on both sides of the foil. It showed roughnesses of a few 100 nm on the inner side (exposed to air during the coating) and a few 10 nm on the outer side (attached to the glass wall). These values are both suitable for storage experiments and can be improved significantly by applying a more thorough cleaning technique, comparable to the preparation of NiMo coated glass tubes [12]. Additionally, moving the oven into a clean-room area will reduce the amount of dust particles and improve surface roughness on the outer side.

3.3. UCN storage measurements

The sample from Fig. 2 with ID = 115 mm and length = 500 mm has been tested using trapped UCN at the PF2 instrument at ILL [14]. For these measurements, the tube was closed on both sides with NiMo coated end caps. A schematic of the setup can be seen in Fig. 3. The resulting UCN storage curve is shown in Fig. 4. No cleaning procedures other than simply wiping the surface have been applied to the coating. Also, no baking in vacuum or other treatment has been applied. The vacuum during the measurements was in the 10⁻⁶ mbar range. It should be noted that the dPE layers are hydrophobic. At t = 0, the interpolated density of stored UCN is about 1 cm⁻³, which is comparable to results obtained with a fully NiMo coated volume. Such a volume was tested in the same reactor cycle. The comparable densities already indicate that \( V_{\text{F,NiMo}} \sim V_{\text{F,dPE}} \). The spectrum of the UCN coming from the source includes neutrons with kinetic energies which are greater than \( V_{\text{F,dPE}} \) and are thus only marginally trapped and will be lost over the first ~ 30 s of the storage time. To account for this “cleaning” time, the data are fitted with a sum of two exponentials, which is a sufficient approximation for this purpose:

\[
N(t) = A_1 e^{-t/\tau_1} + A_2 e^{-t/\tau_1} \tag{5}
\]
Here, the number of neutrons $N(t)$ is dominated by the storage time constant $\tau_1$ for the marginally trapped neutrons and $\tau_2$ for the UCN with $E < V_F$. Here, $\tau_2 = 60 \pm 2$ s. The surface ratios inside the volume are 180550 mm$^2$ dPE coated surface, 20760 mm$^2$ NiMo 85-15 coated end caps (0.1 fraction of the total surface) and 36 mm$^2$ (2.10$^{-4}$ of the total surface) fully absorbing slits at the connections of the end-caps. Therefore, the storage time is composed of

$$\frac{1}{\tau_{\text{meas}}} = \frac{1}{\tau_n} + \frac{1}{\tau_{\text{slit}}} + \frac{1}{\tau_{\text{NiMo}}} + \frac{1}{\tau_{\text{dPE}}} \quad (6)$$

Here, $\tau_n$ causes 7% of the neutrons to escape from the bottle after 60 s. The storage life-times caused by the material surfaces are $\tau_{\text{mat}} = 1/\nu$, $\nu$ is the wall collision frequency and $\mu(E)$ the energy dependent loss per wall collision. With $V_F = 214$ neV (see Sec. 3.4), we obtain $E_{\text{dPE}} = 150$ neV and from kinetic gas theory we obtain for $\nu = \lambda/v = 46$ Hz, using $\lambda = 115$ mm and $v = 5.36$ m/s. The loss coefficient $\eta$ is $\sim \mu/1.2$ for our spectrum and the assumed $V_F$. To estimate the loss on the end-caps, we conservatively determine $\mu(E)_{\text{NiMo}} = 3.5 \times 10^{-4}$ from Ref. [15]. Then, the value for $\eta = 1.2 \pm 0.3 \times 10^{-4}$. Here, the uncertainty is dominated by the unknown exact size of the slits, at the end-caps, which has been mechanically determined to about 20% precision.

3.4. Determination of the Fermi potential

The critical angle of the wall coating was measured by cold-neutron reflectometry at the instrument MIRA at the research reactor FRM-II in Garching at a wavelength of 4 Å. The cold-neutron beam was scattered directly off the inner surface of the coated tubes used for UCN storage measurements. A reflectivity curve is shown in Fig. 5. Due to the curved surface and the high roughness, the curve is broadened significantly. However, a fit with a very simple model consisting only of an air layer and a layer of dPE gives a value for the SLD of $(8.25 \pm 0.21) \times 10^4 \text{ Å}^{-2}$, resulting in a $V_F$ of $(214.8 \pm 5.2)$ neV. The uncertainties are those returned from the fit procedure and do not take into account systematic effects such as curvature of the guides, surface roughness or surface contaminants and are shown for the sake of consistency. Also, very good agreement of $V_F$ deduced from the UCN density at $t = 0$ and with $V_F$ determined with flat samples measured at PSI by Ref. [16] of $214 \pm 10$ neV is obtained.

![Figure 5: Reflectivity vs. momentum transfer of cold neutrons scattered off dPE coated guides. The solid line is the result a very simple two-layer fit, the uncertainty is the one returned by the fit routine.](image)

4. DISCUSSION AND CONCLUSION

We have for the first time demonstrated large surfaces with dPE in a way that makes the container useful for UCN storage. The material is superior to other deuterated plasic coatings (e.g. dPS), due to its high Fermi potential, transparence for UV light, low outgassing, mechanical strength, hydrophobic behavior and simple useability, electric isolation and resilience against electrostatic breakdowns. Whereas in previous approaches only comparably small samples of dPE coated surfaces were realized (e.g. using the spin-coating technique for window coatings in the EDM experiment by Ref. [6]), here a versatile, comparably simple, safe and reliable technique to provide coatings for large surfaces is available. A variety of applications can be envisaged:

- **Coating of UCN storage volumes without slits:** Since the coatings can be removed from smooth surfaces if produced with larger than about 5 μm thickness, guides with flexible sections can be realized. An application for such a configu-
ration is a guide with an integrated shutter without any joints of components and thus no slits.

- **dPE replica tubing**: After removing the coatings from the smooth surface, the smooth side can be everted to obtain a smooth inside of the volume (e.g. a seam-less tube).

- **Radiation-hard quasi-massless guides**: dPE is comparably radiation hard and - as a foil - in addition has very low mass. This can be used to form a UCN guide to transport UCN from an in-pile UCN source to outside of the biological shield.

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