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Rapid Note

Quasielastic scattering of ultracold neutrons as possible reason for their energy spreading during long storage in closed traps

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Abstract. It is shown that quasielastic scattering of ultracold neutrons due to diffusive motion of scatterers at the surface of liquid polymer (fomblin) or adsorbed hydrogenous contaminations of the surface of neutron traps may be possible reason of their energy spreading during long storage time in closed traps, which was observed in the recent experiments.

PACS. 61.12.-q Neutron diffraction and scattering – 61.12.Ex Neutron scattering techniques (including small-angle scattering) – 61.25.Hq Macromolecular and polymer solutions; polymer melts; swelling – 66.10.Cb Diffusion and thermal diffusion

1 Introduction

Ultracold neutrons (UCN) can be stored in a material trap if they have energies less than the boundary energy for this material [1]. The latter is usually about $(1–3)\times10^2$ neV which corresponds to neutron velocities $\sim$(4–7) m/s. There is a widespread opinion that UCN bounce from the walls perfectly elastically provided they survive a wall encounter. UCN loss probability per reflection is usually $\sim 10^{-5}$–10^{-3} depending on the material, its temperature and, what is the most important in the majority of experiments, the presence of hydrogenous contaminations on the surface of the wall. The main reasons for UCN losses in material traps are inelastic scattering with acquiring the energy of the order of the wall temperature (10^{-3}–10^{-1} eV) and subsequent escaping from the trap, and neutron capture by the nuclei of the wall.

Recently two experimental groups observed small energy change of ultracold neutrons (UCN) during long storage in closed traps. Very small heating about 5×10^{-2}–10^{-1} neV of UCN with energy $\sim$ 10 neV was observed in [2] after storage during a hundred seconds, which corresponds to $\sim 10^3$ encounters with walls of the trap, covered with the fomblin grease and oil [3]. The subsequent experiments of the same group [4] did not confirm this observation. Authors [4] did not find any measurable overall energy shift of the spectrum, but it seems that their data may be explained by the small spreading of the neutron spectra during UCN keeping in the trap.

In both sets of experiments (and in the experiments discussed below) the high resolution gravitational spectrometry of UCN was used, based on the fact, that 1 neV change of neutron energy corresponds to 9.8 mm change of height of free flight in gravitational field. This gravitational spectrometry consists in measuring the height distribution of UCN density in a trap after different storage times.

Authors [2] explain their results in the spirit of very unconventional ideas outlined in [5,6].

Strong evidence of UCN cooling and heating during long dwelling times in traps with the bottom covered with fomblin has been found in the experiments [7,8]. Maximum energy of stored UCN in [7,8] was about 14 neV, the mean number of encounters with the walls during keeping UCN in the trap reached as large value as $\simeq 2.5 \times 10^4$ (storage times up to 1200 s). UCN energy change during the storage time was, according to [7], consistent with the rare (the probability $\sim 10^{-6}$ per reflection) neutron energy transfer about 3 neV. It is stated in [7] that mechanical vibrations of the wall could give much lower changes of the UCN spectrum.

On the other hand, according [8] UCN cooling and heating was observed with the UCN energy transfer $\sim$ 15 neV and with probability per UCN reflection in the interval 3×10^{-4}–10^{-3} for several investigated materials: Ni, Cu, C, brass, and Be.

Larger UCN energy increase was found [9,10] in stainless steel chamber after shorter (in comparison with measurements [7]) storage time. For the primary energy of stored UCN in the range (0– $\simeq$100 neV) the observed results have been described in [10] as approximate doubling of the UCN energy during storage times $\simeq$ 200 s. Virtually the inexplicable abnormal subbarrier UCN

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transmission through thick (56 μm) beryllium foil, many orders of magnitude exceeding the quantum mechanical tunnel propagation, was found in [9]. This effect of UCN anomalous propagation through foils was confirmed in [11] for 10 μm copper foils with reference, that most probably this transmission is attributed to non-perfect cleaning of the incident UCN spectrum from neutrons with higher energies; but it was not observed for the thicker beryllium and 12 μm stainless steel foils. This anomalous transmission was confirmed in the subsequent experiments [10] for the aluminium foils, and it was demonstrated in this work that the reason for this transmission is increasing of UCN energy during storage time. No explanation of the observed effects was given in [7–10].

It is shown in this paper that results described in the publications [2,4,7,8] may in principle be explained by quasielastic UCN scattering due to diffusive motion of the polymer chains at the fomblin surface in the experiments [2,4,7] and diffusive motion of hydrogen atoms of significant hydrogenous contaminations in the experiments [7–10].

For example the result [7], for the fomblin surface, and interpreted as rare, with probability 10^{-6} UCN energy transfer 3 neV, may be described as diffusion-like energy spreading $\Delta E \simeq \delta E \times n^{1/2}$, where δE is mean energy transfer per UCN collision with the wall, n is the number of collisions during UCN storage in the trap, and ΔE is the total UCN energy spreading. The results [7] may be fitted if to take $\delta E \approx 3 \times 10^{-3}$ neV.

It must be mentioned, that the way by which the quantitative conclusions are obtained in all the cited publications, is approximate. Therefore the scenario proposed in the present work can not be an exact interpretation of these experiments, but may only serve as an indication of the physical processes, leading to the observed phenomena and the order of magnitude estimations of the observed effects.

2 UCN quasielastic scattering on polymer surface

Quasielastic neutron scattering in melt polymers may be described, for example, by the Zimm model [12] in which hydrodynamic effects of long range interaction between subunits of the polymer chain are taken into account. Fomblins are liquid perfluoropolyethers (PFPE) with wide range of molecular weight, having the formula: $CF_3{-}(OC_2F_4CF_2)_p{-}(OCF_2)_q{-}O{-}CF_3$. According to [3] the Fomblin with molecular weight about $M \approx 2650$ is used in the UCN storage experiments.

The region of scattering vector covered in UCN interaction with the surface was $\kappa \leq (1-2) \times 10^{-3}$ Å^{-1} in the experiments [2,4,7,8], and $\simeq$(1-5)$\times 10^{-3}$ Å^{-1} in the experiments [9,10]. This range is much lower than the ordinary in the thermal neutron quasielastic scattering experiments κ between $\sim 10^{-2}$ and several Å^{-1}, and is reaching the κ range of the photon correlation spectroscopy. The intermediate scattering function in the limit of small κ is

$$F_s(\kappa, t) = \exp\bigl[-\kappa^2 \Gamma(t)/2\bigr], \tag{1}$$

with the spreading $\Gamma(t) = 2Dt$, where D is diffusion coefficient relevant to simple diffusion regime. In the Zimm model the spreading of the scattering function $S(\kappa, \omega)$ (half width at half maximum) is:

$$\delta E(\kappa) = \widetilde{W} \hbar (\kappa\sigma)^3, \tag{2}$$

where $\widetilde{W} = \sqrt{2\pi} z W$, $z = \sqrt{6/\pi} b/\sigma$, b is the size of subunit in the polymer chain, σ is the length characteristic of the local structure of the chain and is of the order of a few monomer units, W follows from Einstein-Stokes relation: $W = k_B T/(2\pi\eta b\sigma^2)$, where η is the viscosity.

From the above we are able to estimate roughly the energy spreading δE for UCN reflecting from the fomblin surface. Taking for example $b = 5$ Å, $\sigma = 25$ Å, $z = 0.28$, $\eta = 1.5$ CGSE unit [15,16], $\kappa \approx 2.5 \times 10^{-3}$ Å^{-1} [7], we obtain $\delta E \simeq 0.4 \times 10^{-3}$ neV. The mean energy transfer per UCN collision is much larger (see below) which is in rough agreement with results [7]. It must be noted that there is no information on viscosity and diffusion coefficients for the very thin $\simeq$ (50−100) Å layers of liquid polymers; it may be supposed that viscosity is lower, and diffusion coefficient is larger for a surface than for a bulk liquid. In the latter case the neutron energy spreading in the quasielastic scattering has to be larger.

The probability of the quasielastic scattering at the reflection from the liquid surface is determined by quasielastic scattering cross-section for the essentially coherent scatterer such as PFPE. It follows from the standard consideration that cross-section for coherent scattering with wave vector change κ is determined by the sum of the atomic coherent scattering lengths $\sum_i b_i$ over the sample region $\simeq \kappa^{-1}$ (volume $\sim \kappa^{-3}$) if there is coordinate correlations of atom motion in this sample region:

$$\left(\frac{d^2\sigma}{d\Omega d\epsilon}\right)_{coh} = \frac{b_{coh}^2 k}{2\pi\hbar k_0 N} \sum_{\nu}^{N} \sum_{\nu'}^{N} \int dt e^{-i\epsilon t/\hbar} \times \langle e^{i\kappa \mathbf{r}_\nu(t)} \cdot e^{-i\kappa \mathbf{r}_{\nu'}(0)} \rangle_T. \tag{3}$$

In case of these correlations (slow diffusive motion of large fragments – several monomer units of the polymer chains in melt) integral in (3) is of the order of unity and summation gives N_0^2, where N_0 is number of atoms in the volume of coherent motion, which may give very large cross-section of quasielastic UCN scattering for the experiments [2,4,7,8]. This is not the case for incoherent scatterer and for monoatomic or small molecule liquids, where there is no such correlations of atom motion.

According to the accepted formalism for inelastic neutron losses at UCN reflection from the walls, the probability of quasielastic reflection is

$$\eta = \text{ Im } U/\text{ Re } U,$$
$$U = (2\pi\hbar^2/m) \sum_i N_i b_i,$$
$$\text{and Im } b = k\sigma_{qel}/4\pi. \tag{4}$$

In this expression U is the wall potential for neutrons, N_i and b_i are atomic density and coherent scattering lengths of nuclei in the wall. With very large σ_{qel} the probability of quasielastic UCN reflection approaches to unity.

3 UCN quasielastic scattering on adsorbed hydrogenous contaminations

Now let's discuss the experiments [9,10] in which "large" energy increase – of the order of the primary UCN energy – was observed during storage in the trap. The ordinary problem of UCN traps is significant hydrogenous contaminations of the inner surface of the traps. The experiments [9,10] demonstrate very short experimental UCN life-times in their stainless steel chamber in comparison both with the results, obtained for cleaned and outgassed at high temperature in vacuum traps, and with calculations for clean surface.

It is possible to estimate with high confidence the UCN loss coefficient per one collision with the walls of the stainless steel chamber from the measured storage times, size of the chamber, and the UCN spectrum [9,10]. Monte-Carlo simulation of the UCN density evolution in the stainless steel chamber of the geometry [10] gives for the value of the loss coefficient $\eta \approx (4{-}5) \times 10^{-3}$. The calculated loss coefficient according to equation (4) for the stainless steel gives $\eta \simeq 10^{-4}$ which means that experimental loss coefficient is ≈ 50 times larger than it must be for the clean stainless steel surface. According to [9,10] the chamber was not outgassed at high temperature in vacuum, in this case so large difference may me attributed to the surface hydrogenous contaminations, most probably to the adsorbed water.

Calculation for the quantum mechanical potential, consisting of the stainless steel barrier and the water layer at the surface, shows that large UCN loss coefficient in [9, 10] may be well explained by the adsorbed water layer ≥ 100 Å thick.

Hydrogen diffusion in this thick surface layer may be not very different from diffusion in bulk water at room temperature with the diffusion coefficient $D \simeq 1.8 \times 10^{-5}$ cm^2/s. Assumption that diffusion in the thick physically adsorbed water layer is not so large, but is rather similar to diffusion in frozen water does not change essentially the proposed picture, because it is known from macroscopic measurements (confirmed by the neutron experiments [18]) that diffusion coefficient in water changes only approximately three times in the interval $[-20, 20]$ C. The quasielastic scattering for the hydrogen atom is $\sigma_{qel} = 4\pi b_{inc}^2 \times (\mathrm{E/E_0})^{1/2} \simeq 80b$, where b_{inc} is the hydrogen incoherent scattering length. The inelastic neutron upscattering in the room temperature water (and in many different hydrogen containing compounds [19]) behaves as $\sigma_{inel} \simeq (3-7)b \times 2.2 \times 10^5/v_{ucn}$ (cm/s). For the UCN energy $\simeq 50$ neV the ratio $\sigma_{qel}/\sigma_{inel} \approx 1.6 \times 10^{-2}$ and is decreasing with decreasing UCN energy. It follows that for this particular stainless steel barrel the probability of quasielastic scattering due to diffisive motion of surface hydrogen is less than $\sim 10^{-4}$, *e.g.* inelastic UCN upscattering dominates over quasielastic scattering and is the main mechanism of UCN losses. The observed UCN heating probability with "doubling energy" in the case of stainless steel chamber (10^{-5}–10^{-6}) is orders of magnitude lower than the measured total loss probabilty ($\sim 5 \times 10^{-3}$) in this experiment. So it seems the small UCN heating and cooling, being interesting itself, hardly has relation to the anomalous UCN losses in material traps.

It is not clear yet whether the diffusion scattering may be dominating at lower temperatures where the "Gatchina anomaly" [13] takes place: it may happen in case of abnormally high hydrogen diffusion at low temperatures.

At small changes of the neutron wave vector κ the spreading of scattering function (h.w.h.m) is [14]:

$$\delta E = \hbar \kappa^2 D. \tag{5}$$

Rough estimation gives that with the primary UCN energy $\simeq 50$ neV, $\delta E \simeq 3$ neV, and $\Delta E = \delta E \cdot n^{1/2}$, where the quantity of collisions $n \simeq 250$, we have $\Delta E \simeq 50$ neV. The increasing of the energy gain during storage with the energy of primary neutrons [10] confirms this scenario.

It is possible to calculate the spectrum of quasielastically scattered neutrons using for simplicity the model of classical diffusion, which works well at the conditions: $\kappa^2 \langle R^2 \rangle / 6 \ll 1$, and $\kappa^2 D \tau_0 \ll 1$, where $\langle R^2 \rangle$ is mean squared radius of hydrogen atoms vibrations, τ_0 is the mean time of vibrations before jumping to other site in the diffusion process [14]. These conditions are satisfied very well even at the UCN energy after upscattering as large as 1 μeV, which is far outside the measurement conditions of the experiments [9,10]. Contrary to typical quasielastic neutron scattering experiments in which the energy distribution of the scattered neutrons at the fixed κ, or the probability of the elastic scattering as a function of κ or temperature are studied, in the cited experiments with UCN the angle of scattering is not determined, and the energy change is accumulated as a result of many act of scattering. Integration over solid angle of the expression for quasielastic differential scattering cross-section in the classical limit [14]:

$$\frac{d^2\sigma_{qel}}{d\Omega d\epsilon} = \frac{b_{inc} k}{\pi \hbar k_0} \frac{\kappa^2 D}{(\epsilon/\hbar)^2 + (\kappa^2 D)^2} \tag{6}$$

yields differential quasielastic scattering cross-section as a function of transferred energy ϵ:

$$\frac{d\sigma_{qel}}{d\epsilon} = 4\pi b_{inc}^2 \frac{a}{E_0} \ln \left[\frac{d^2 + b^2 \Big((1+d)^{1/2} + 1 \Big)^4}{d^2 + b^2 \Big((1+d)^{1/2} - 1 \Big)^4} \right], \tag{7}$$

where $a = \hbar/(16\pi MD)$, M is neutron mass, $b = 2MD/\hbar$, and $d = \epsilon/E_0$, E_0 is the incident UCN energy.

This cross-section is asymmetric function in respect to $\epsilon = 0$ for UCN, with dominating upscattering cross-section. Computations give that the mean energy transfer $\langle \epsilon \rangle \gg \delta E$ determined by (5) in the energy range of applicability of the model of classical diffusion. This is also true

for the UCN quasielastic scattering in the melt polymer. The mean energy transfer $\langle\epsilon\rangle$ is much larger than δE of the equations (2, 5) both in upscattering and downscattering quasielastic process. For example for the spreading (5) of the quasielastic Lorentzian $\delta E = 4 \times 10^{-4}$ neV, (which corresponds to the hydrogen diffusion coefficient $D \approx 1.2 \times 10^{-8}$ cm^2/s), and the primary UCN energy $E_0 = 10$ neV, the mean downscattering energy change $\langle\epsilon_{down}\rangle \simeq -3.4 \times 10^{-3}$ neV, and the mean upscattering energy change $\langle\epsilon_{up}\rangle \simeq 2.5 \times 10^{-2}$ neV, the latter being obtained after the integration over the cross-section of the equation (7) up to $\epsilon = 100$ neV. For the case of the adsorbed hydrogen with the diffusion coefficient relevant to water, probability for the UCN with energy $E_0 = 50$ neV to acquire the energy $\epsilon\rangle E_0$ in the act of quasielastic scattering is about 5%, which in combination with total quasielastic scattering probability $\sim 10^{-4}$ fits well the results [10].

According to our hypothesis the neutron spectrum after UCN collision with the wall with large hydrogen contaminations is not the result of the "doubling" of the incident UCN energy but is the broad distribution with long tail at large energies, described by the equation (7). On the other hand the conclusion about doubling UCN energy upon reflection from the wall [10] does not seem convincing. The presented experimental data may be explained by diffusion-like process of energy exchange between UCN and the walls with smooth transferred energy distribution function, if to account for UCN losses in the trap due to prevailing loss UCN mechanism – upscattering to the thermal energy range.

For the clean surfaces or at low temperatures the observed effect of UCN heating [10] according to our hypothesis must be decreased or disappear.

It is worth noting that the experiments of the type [2,3,7–10] have extraordinary high sensitivity of the order 10^{-12} eV in the measuring energy changes of neutrons during collisions with walls due to accumulation of the energy changes in result of many acts of interaction, but have very moderate momentum and energy resolution in view of not determined geometry of reflection. Nevertheless some applications are possible for measuring very slow coherent motion in matter.

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