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QUASIELASTIC NEUTRON SCATTERING BY DIFFUSIVE ADSORBED HYDROGEN AS A POSSIBLE REASON FOR ULTRACOLD NEUTRONS ENERGY SPREAD DURING LONG STORAGE IN TRAPS

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Small ultracold neutron cooling and heating during long storage in traps has been observed in recent experiments. It is shown that neutron quasielastic scattering due to the diffusive motion of hydrogen at the surface of adsorbed hydrogenous contaminations of the surface may be a possible reason for the spread in the energy of ultracold neutrons during long storage in traps.

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1. 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 $(1\text{–}3)\cdot 10^2$ neV, which corresponds to neutron velocities of $\sim$(4–7) m/s. There is a widespread opinion that UCN bounce perfectly elastically from the walls of the trap, provided they survive a wall encounter. The 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 the acquisition of energy of the order of the wall temperature ($10^{-3} - 10^{-1}$ eV), and the subsequent escape from the trap, and neutron capture by the nuclei of the wall.

Recently, two experimental groups observed a small energy change in UCN during long storage in closed traps.

The UCN energy increase was observed [2, 3] in a stainless steel chamber for the primary energy of stored UCN in the range of 0– $\simeq$100 neV. The results have been described in [3] as an approximate doubling of the UCN energy with probability $\sim 10^{-5}$

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per the trap's wall encounter during the storage time 200 s. Virtually, an inexplicable and abnormal sub-barrier UCN transmission through a thick (56 μm) beryllium foil, exceeding by many orders of magnitude the quantum mechanical tunneling propagation, was found in [2]. This effect of UCN anomalous propagation through foils was confirmed in [4] for 10 μm copper foils, with a comment that, most probably, this transmission should be attributed to a non-perfect cleaning of the incident UCN spectrum from neutrons with higher energies. This effect was not observed [4], however, for thicker beryllium and 12 μm stainless steel foils. This anomalous transmission was then confirmed in the subsequent experiment [3] with aluminium foils, and, what is the most important, it was demonstrated that the reason for this transmission is the increase in UCN energy during the storage time. No explanation of the observed effects was given up to now.

On the other hand, according to the experiments of the second experimental group (Ref. [5]), performed in somewhat different way, UCN cooling and heating was observed with the UCN energy transfer $\sim$ 15 neV and with probability per UCN reflection in the range of $3 \cdot 10^{-4} - 10^{-3}$ for several investigated materials: Ni, Cu, C, brass, and Be.

Suspicions about the possibility of small energy changes in UCN at wall reflections in the traps were voiced many times long ago but without indicating any physical mechanism (see, for example, [6]). The effect of possible undesirable wall sound vibrations was estimated in [7]. The possible effect of low frequency part of the phonon spectrum of solids and the very questionable existence of low frequency vibrating clusters in disordered solids were considered in [8].

Some special experiments were previously undertaken to search for small UCN energy changes during long storage. The authors [9] reported that for UCN in the energy range of (6–28) neV, in the copper traps with a measured loss coefficient of $\sim 10^{-3}$, they observed an overall negative shift of the UCN spectrum $\simeq$(2-3) neV after 140 s of UCN storage in the trap. But bearing in mind that hardly there may be any reason for the directional negative UCN energy change, they reported the result that the neutron energy change per reflection did not exceed $7 \cdot 10^{-2}$ neV.

It is shown in this paper that the results obtained in the experiments ([2, 3, 5]) may be explained by the diffusive motion of hydrogen atoms in significant hydrogenous contaminations of the surface of the traps.

It must be mentioned that the way by which the quantitative conclusions were obtained in all the cited publications is approximate. Therefore, the scenario proposed in the present work cannot 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. The ordinary problem of UCN traps is significant hydrogenous contamination of the inner surface of the traps. The experiments [2, 3] demonstrate very short experimental life-times for UCN in their stainless steel chamber in comparison with the results obtained for traps that were cleaned and outgassed at high temperature in a vacuum, and with calculations for a clean surface. It is possible to estimate, with high certainty, the UCN loss coefficient η per collision with the walls of the chamber from the measured storage time, size of the chamber, and the UCN spectrum [2, 3]. Simple estimation yields $\eta \approx$ $\approx (3-4) \cdot 10^{-3}$, Monte Carlo simulation of the UCN density evolution in the chamber of the geometry in [3] confirms this estimation. The calculated loss coefficient for stainless steel yields $\eta \simeq 10^{-4}$, which means that the experimental loss coefficient is $30-40$ times

larger than it must be for the clean stainless steel surface. According to [2, 3], the chamber was not outgassed at high temperature in a vacuum. In this case, such a large difference can be attributed to surface hydrogenous contaminations, most probably adsorbed water, which was confirmed in numerous experiments [1].

Calculation for the quantum mechanical potential, consisting of the stainless steel barrier and the water layer at the surface, shows that the large UCN loss coefficient in [2, 3] may be explained by an adsorbed or dissolved near surface hydrogenous layer ≥ 100 Å thick.

Hydrogen diffusion in this thick surface water layer may not differ very much from bulk water at room temperature, where the diffusion coefficient is $D \simeq 1.8 \cdot 10^{-5}\,\mathrm{cm}^2/\mathrm{s}$. The assumption that diffusion in a thick, physically adsorbed water layer is not as large, but is rather similar to diffusion in frozen water, does not basically change the proposed picture because it is known from macroscopic measurements (confirmed by the neutron experiments [10]) that the diffusion coefficient in water changes only approximately three times in the range of (–20, 20) °C. Measured by quasielastic neutron scattering hydrogen diffusion coefficients in water adsorbed on silica surfaces was found to be in the range $(2 - 8.5) \cdot 10^{-6}$ cm^2/s, depending on the degree of hydration [11].

On the other hand, hydrogen dissolved in metals has in some cases large diffusion constant. For example, diffusion coefficients of atomic hydrogen in α-Fe at room temperature is as large as $D \approx 1.4 \cdot 10^{-5}$ cm^2/s or even larger depending on particular experiment [12].

In many cases hydrogen absorbed from the atmosphere or from the low vacuum absorbs dissociatively. In the real experiments with UCN, hydrogen on the surface may be present in many different forms with a variety of diffusion coefficients.

Generally the metal surface is covered with oxide layer. There is very low information on hydrogen diffusion in oxides. The studies show that micro-structure and microchemistry of the underlying metal or alloy can affect the characteristics of the oxide and in turn the diffusion of hydrogen through the oxide. In certain cases the oxide layer may be a homogeneous medium for hydrogen diffusion, but in most cases it is heterogeneous and may contain extremely fine interconnected cracks and pores undetected by conventional microanalytical techniques. These cracks and pores are the good cites for the adsorbed hydrogenous contaminations of the near surface layer which is important in UCN experiments.

3. The total neutron quasielastic scattering cross-section for the hydrogen atom is

$$\sigma_{qel} = 4\pi b_{inc}^2 (E/E_0)^{1/2} \simeq 80b,$$

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 [13]) behaves as $\sigma_{inel} \simeq (3 - 7)b \cdot 2.2 \cdot 10^5 / v_{ucn}\,(\mathrm{cm/s})$. For an UCN energy $\simeq 50$ neV, the ratio $\sigma_{qel}/\sigma_{inel} \gtrsim 1.6 \cdot 10^{-2}$, decreasing with decreasing UCN energy, e.g., inelastic UCN upscattering dominates over quasielastic scattering and is the main mechanism of the UCN losses. For this particular stainless steel barrel, the probability per one wall encounter of quasielastic scattering due to diffusive motion of surface hydrogen is less than $\sim 10^{-4}$. Indeed, the reported in [3] UCN heating probability with a doubling energy of $\approx 10^{-5}$ in the case of stainless steel chamber, is more than two orders of magnitude lower than the measured total loss probability ($\sim 4 \cdot 10^{-3}$) in this experiment. More accurate

recent processing of the experimental data of [3] yielded much lower value for the probability of doubling the UCN energy at reflection: between[2] $\sim 5 \cdot 10^{-7}$ and $\sim 10^{-6}$. Thus, it seems that the small UCN heating and cooling due to diffusive motion of hydrogen, being interesting in itself, is not the main reason of anomalous UCN losses in material traps.

It is not yet clear whether diffusion scattering may dominate at lower temperatures, where the "Gatchina anomaly" [14] takes place: it may happen in the case of abnormally high hydrogen diffusion at low temperatures.

At small changes in the neutron wave vector κ, the spreading of the scattering function (h.w.h.m) is [15]

$$\delta E = \hbar \kappa^2 D. \tag{1}$$

It yields that with the primary UCN energy $\simeq 50\,\mathrm{neV}$, and D relevant to water, $\delta E \simeq 3\,\mathrm{neV}$, and $\Delta E = \delta E \cdot n^{1/2}$, where the quantity of collisions is $n \simeq 250$, we have $\Delta E \simeq 50\,\mathrm{neV}$. The increase in the energy gain during storage, with the energy of the primary neutrons, observed in [3] confirms this scenario.

It is possible to calculate the spectrum of quasielastically scattered neutrons using the model of classical diffusion for simplicity, which works well at the conditions $\kappa^2 < R^2 > /6 << 1$ and $\kappa^2 D \tau_0 << 1$, where $< R^2 >$ is the mean squared radius of hydrogen atoms vibrations and τ_0 is the mean time of vibrations before jumping to other sites in the diffusion process [15]. These conditions are satisfied very well even at UCN energies after upscattering as large (in comparison with the incident UCN energies) as $10\,\mu\mathrm{eV}$, which is far outside the measurement conditions of the experiments [2, 3]. In contrast to typical quasielastic neutron scattering experiments, where 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, the angle of scattering in the cited experiments with UCN is not determined, and the energy change is accumulated as a result of many scattering acts. Integration over the solid angle of the expression for the cross-section for quasielastic differential scattering in the classical limit [15]

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

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

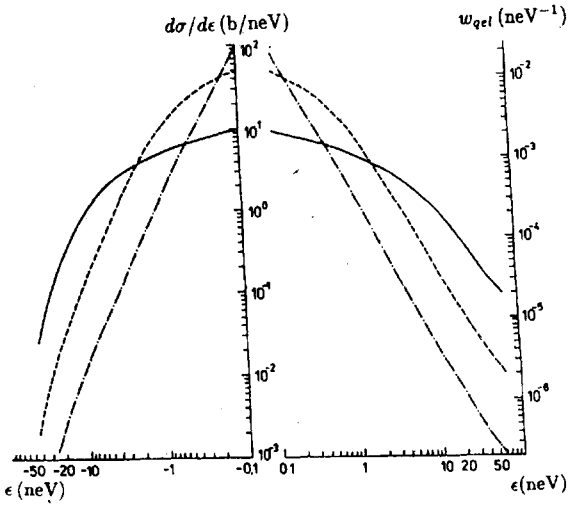
$$\frac{d\sigma_{qel}}{d\epsilon} = 4\pi b_{inc}^2 \frac{a}{E_0} \ln\left[\frac{d^2 + b^2\Big((1+d)^{1/2}+1\Big)^4}{d^2 + b^2\Big((1+d)^{1/2}-1\Big)^4}\right], \tag{3}$$

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

This cross-section is an asymmetric function with respect to $\epsilon = 0$, with the upscattering cross-section dominating. Results of calculations of differential cross-section and probability of UCN quasielastic scattering due to diffusive motion of hydrogen atoms (ratio of the quasielastic scattering to the total UCN loss probability at a wall encounter) are shown in Figure for different values of the diffusion coefficient. Computations yield that the mean energy transfer $< \epsilon > \; >> \delta E$ determined by Eq.(1) in the energy range

[2] E.V.Lychagin, private communication.

for applicability of the model of classical diffusion. For the case of adsorbed hydrogen with a diffusion coefficient relevant to water, the probability for UCN with the energy $E_0 = 50\,\text{neV}$ to acquire the energy $\epsilon > E_0$ in the act of quasielastic scattering is about 5%, which, in combination with the value of quasielastic scattering probability relative to inelastic one of $\approx 1.6 \cdot 10^{-2}$, and reflection probability $4 \cdot 10^{-3}$ fits the results of [3] quite well. The corrected [2)] value of the probability of doubling the UCN energy at reflection of $\sim 5 \cdot 10^{-7} - 10^{-6}$ needs significantly lower adsorbed hydrogen diffusion coefficient than in liquid water. It is even more appropriate for the proposed hypothesis of UCN quasielastic scattering at the diffusive adsorbed hydrogen as possible reason for small UCN heating and cooling during storage in traps.



Differential cross-section $d\sigma/d\epsilon$ (b/neV) and relative (to the total loss) probability w_{qel} (neV^{-1}) of UCN quasielastic scattering due to diffusive motion of hydrogen atoms for different values of the diffusion coefficient D: solid line – $D = 1.85 \cdot 10^{-5}\text{cm}^2/\text{s}$; dashed line – $D = 1.85 \cdot 10^{-6}\text{cm}^2/\text{s}$; dashed-dotted line – $D = 1.85 \cdot 10^{-7}\text{cm}^2/\text{s}$. Incident neutron energy 50 neV

According to proposed scenario, the neutron spectrum after UCN collision with a wall with hydrogen contaminations is not the result of the "doubling" of the incident UCN energy, but is a broad smooth distribution with a long tail at large energies described by Eq. (3).

Additional important confirmation of this scenario is observation of proportionality or at least strong correlation between probabilities of weak heating and the inelastic scattering of UCN to the thermal energy range [5]. Both effects are proportional to the extent of hydrogen contamination of the surface.

For clean solid surfaces or at low temperatures, the observed effect of UCN heating [3, 5] according to our hypothesis must be reduced or disappear.

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