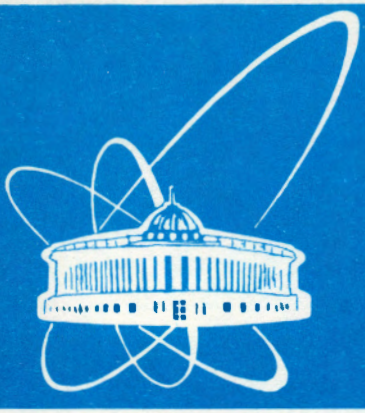

ОБЪЕДИНЕННЫЙ
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Yu.N.Pokotilovski[1]

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

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[1]E-mail: POKOT@nf.jinr.ru

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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)·10^2 neV, which corresponds to neutron velocities of ∼(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 ∼ 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− ≃100 neV. The results have been described in [3] as an approximate doubling of the UCN energy with probability ∼ ·10^{-5} per the trap wall encounter during the storage time ≃200 s. Virtually, an inexplicable and abnormal sub-barrier UCN transmission through a thick (56μm) 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 10μm 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 also confirmed in the subsequent experiments [3] with aluminium foils, work, 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 in [2, 3].

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 ∼ 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 ∼ 10^{-3}, they observed an overall negative shift of the UCN spectrum ≃(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 approx-

imate. 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 Hydrogen contaminations of the surface of the UCN traps

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. The authors of [2, 3] do not directly report the total UCN loss probability for their stainless steel storage chamber, but these values can be easily extracted from the UCN density decay curves shown in [2, 3]: $\tau \simeq 60s$ for the experiment [2] and $\tau \simeq 80s$ for the experiment [3].

It is possible to estimate, with high certainty, the UCN loss coefficient 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 according to $\bar{\mu} \approx d/(\bar{v}\tau)$, where $\bar{\mu}$ is the mean UCN loss coefficient per relection, d is the UCN mean free path inside the trap, $\bar{v}$ is the mean UCN velocity and τ is the measured UCN life-time in the trap yields $\eta \approx (3-4)\cdot 10^{-3}$ for the value of the loss coefficient. Monte Carlo simulation of the UCN density evolution in the chamber of the geometry in [3] confirms this estimation. According to the accepted formalism for neutron losses at UCN reflection from the walls [1], the loss

probability is:

$$\eta = \text{Im } U/\text{ Re } U, \quad \text{where Re } U = (2\pi\hbar^2/m)\sum_i N_i \text{ Re } b_i,$$
$$\text{and Im U } = k\sigma_{qel}/4\pi. \qquad (1)$$

In this expression, U is the wall potential for neutrons, and N_i and b_i are the atomic density and coherent scattering lengths of nuclei in the walls, respectively. The calculated loss coefficient, according to Eq. (1) 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.

For example, [10] presents the results of systematic investigations of the influence of the vacuum outgassing on the UCN loss coefficient and UCN upscattering cross-section for a beryllium surface. The authors [10] reached almost an order of magnitude decrease in the UCN loss coefficient (beginning from the value which was more than two orders of magnitude higher than calculated one for clean beryllium at room temperature) after heating the sample in vacuum at 700°C. They found a very good correlation between the UCN losses due to upscattering and the quantity of adsorbed hydrogen (measured by simultaneous gamma-radiation analysis from UCN capture in hydrogen on the surface of the beryllium samples) in the surface layer of the beryllium samples in their entire procedure of vacuum high temperature cleaning. Thus, it has been directly shown [10] that the UCN losses in their experiments were due to hydrogen contamination of the beryllium surface.

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 water layer $\geq 100\mathring{A}$ thick. Additional argument in favor of this hypothesis is the small difference between the UCN life-times in the same stainless steel barrel for two very different UCN spectra: 0-200 neV in the experiment [2] and 0-50 neV in the experiment [3]. This may happen in the case where the UCN loss coefficient does not depend or weakly depends on the UCN energy for the upper part of the UCN spectrum. The calculations performed for the mentioned quantum mechanical potential show exactly the same behaviour for the UCN loss coefficient as a function of the UCN momentum (see Fig. 1). The calculations also demonstrate the decrease in the UCN loss coefficient for the UCN velocity component normal to the surface that is larger than 5 m/s, in contrast to the clean surface.

Hydrogen diffusion in this thick surface water layer probably does 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 [11]) that the diffusion coefficient in water changes only approximately three times in the range of (-20, 20) K. Measured by quasielastic neutron scattering hydrogen diffusion coefficients in water adsorbed on silica surfaces was found to be in the range $(2-8.5)\cdot 10^{-6}\ \mathrm{cm}^2/\mathrm{s}$, depending on the degree of hydration [12].

On the other hand, hydrogen dissolved in metals has in some cases large diffusion constant. For example, diffusion coefficients of atomic hydrogen in $\alpha - Fe$ at room temperature is as large as $D \approx 1.4 \cdot 10^{-5}\ \mathrm{cm}^2/\mathrm{s}$ or even larger depending on particular experiment [13].

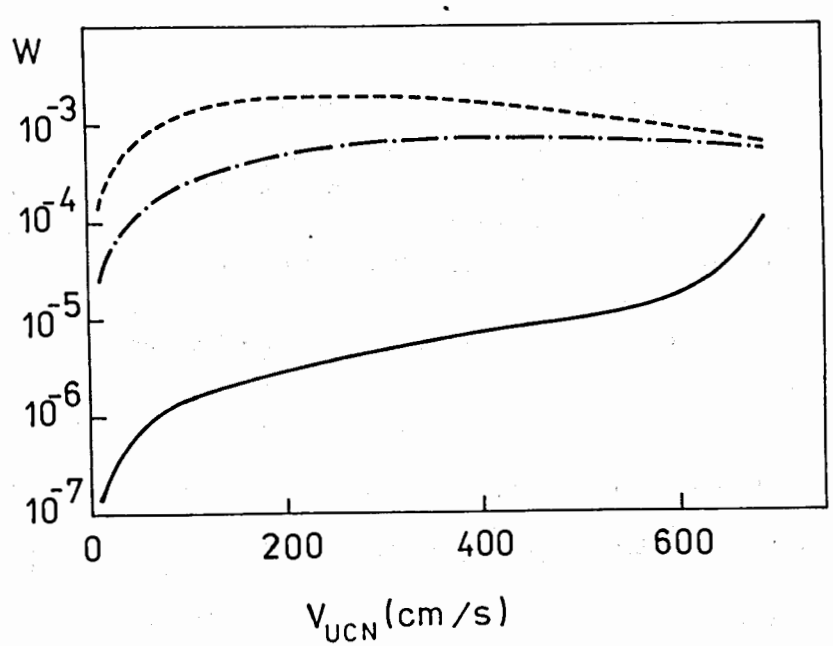

Fig. 1 Calculated UCN loss coefficient as a function of the normal to the surface component of neutron velocity for a beryllium wall covered with water layer of different thickness: solid line - clean beryllium, dashed-dotted line - 100 Å, dashed line - 200 Å.

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 micro-chemistry 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 UCN quasielastic scattering at the adsorbed hydrogen

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

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

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 [14]) behaves as $\sigma_{inel} \simeq 7b \cdot 2.2 \cdot 10^5/v_{ucn}\,(\mathrm{cm/s})$. For an UCN energy $\simeq 50\,\mathrm{neV}$, the ratio $\sigma_{qel}/\sigma_{inel} \approx 1.6{\cdot}10^{-2}$, 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 $\sim \quad 10^{-5}$ in the case of stainless steel chamber, is 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 $\sim 5 \cdot 10^{-7}$ and $\sim 10^{-6}$ [15]. 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" [16] 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 [17]

$$\delta E = \hbar\kappa^2 D. \qquad (2)$$

Rough estimation shows that with the primary UCN energy $\simeq$ 50 neV, $\delta E \simeq 3\,\mathrm{neV}$, 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 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 [17]. These conditions are satisfied very well even at UCN energies after upscattering as large (in comparison with the incident UCN

energies) as $10\mu eV$, 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 [17]

$$\frac{d^2\sigma_{qel}}{d\Omega d\epsilon} = \frac{b_{inc}k}{\pi\hbar k_0}\frac{\kappa^2 D}{(\epsilon/\hbar)^2 + (\kappa^2 D)^2} \tag{3}$$

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

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

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 Fig. 2 for different values of the diffusion coefficient. Computations yield that the mean energy transfer $<\epsilon>$ $>> \delta E$ determined by Eq. 2 in the energy range 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 $E_0 = 50\,\mathrm{neV}$ to acquire the energy $\epsilon > E_0$ in the act of quasielastic scattering is

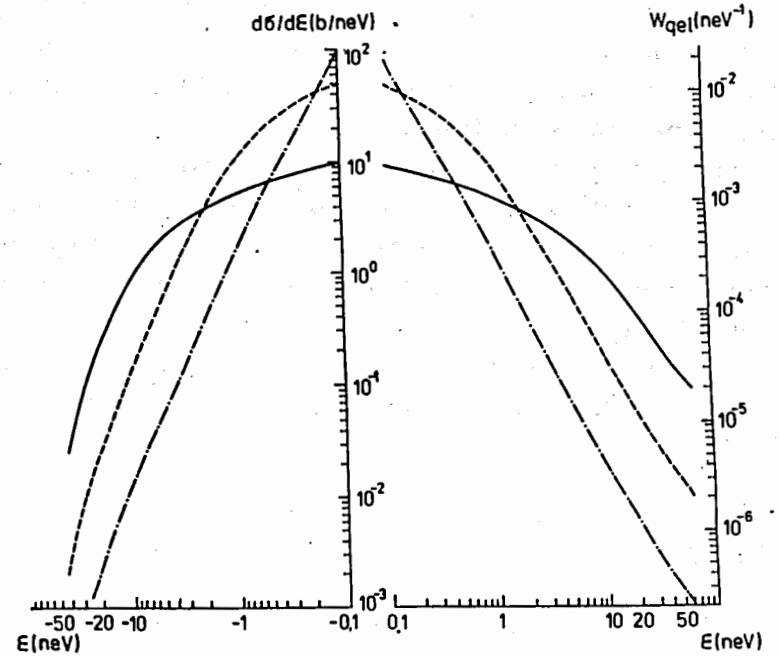

Fig. 2 Differential cross-section $d\sigma/d\epsilon$ (b/neV) and relative probability w_{qel} (neV^{-1}) of UCN quasielastic scattering due to diffusive motion of hydrogen atoms for different values of the diffusion coefficient D: solid line - $D = 1.85 \cdot 10^{-5}\mathrm{cm}^2/\mathrm{s}$; dashed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

about 5%, which, in combination with the value of quasielastic scattering probability relative to inelastic one of $\sim 10^{-2}$, fits the results of [3] quite well. The corrected [15] 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.

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. (4).

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

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Покотиловский Ю.Н. Е3-98-310

Квазиупругое рассеяние нейтронов на диффундирующем адсорбированном водороде как возможная причина энергетического расплывания ультрахолодных нейтронов при их хранении в ловушках

В недавних экспериментах наблюдались слабые нагрев и охлаждение ультрахолодных нейтронов при их хранении в ловушках. Показано, что в ловушках с твердыми стенками причиной энергетического расплывания нейтронов может быть квазиупругое рассеяние на диффундирующем адсорбированном водороде.

Работа выполнена в Лаборатории нейтронной физики им. И.М.Франка ОИЯИ.

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Pokotilovski Yu.N. E3-98-310

Quasielastic Neutron Scattering by Diffusive Adsorbed Hydrogen as a Possible Reason for Ultracold Neutrons Energy Spread during Long Storage in Closed Traps

Small ultracold neutron cooling and heating during long storage in closed 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 closed traps.

The investigation has been performed at the Frank Laboratory of Neutron Physics, JINR.

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