ядерных исследований

Дубна

E17-98-288

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INTERACTION OF ULTRACOLD NEUTRONS WITH LIQUID SURFACE MODES AS A POSSIBLE REASON FOR NEUTRON ENERGY SPREAD DURING LONG STORAGE IN FLUID WALL TRAPS

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1998

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)$\cdot 10^2$ 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 reason for UCN losses in material traps is 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. Another possible reason for UCN losses is 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. Very small heating, about $5 \cdot 10^{-2} - 10^{-1}$ neV, of UCN with an energy of $\sim$ 10 neV was observed in the experiments of Ref. 2 after a storage of a hundred seconds, which corresponds to $\sim 10^3$ encounters with the walls of the trap covered with Fomblin grease and oil [3]. Subsequent experiments of the same group [4] may be interpreted as small UCN cooling. The authors of Ref. 4 did not find any measurable overall energy shift of the spectrum, but it seems that their data may be explained by a small spreading of the neutron spectra during UCN storage in the trap.

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

The authors of Ref. 2 and 4 explain their results as the temporary adhesion of some UCN to the bottom of the trap. This is explained as being a result of some neutron wave localization on the surface, under the effect of gravity, and in the spirit of the very interesting but unconventional idea outlined in Refs. 5 and 6. Briefly, this idea was motivated by the problem of the nonexponentiality of the decay of an unstable particle and consists in the introduction of a novel universal degree of freedom for any kind of interaction — in this particular case — the gravitational one. The quantum motion in the gravitational field is a sequence of statistical changes in quantum states with different parameters and their life-times $\tau = (\hbar/2gp_z)^{1/2}$, where g is the gravitational acceleration and p_z is the vertical momentum component of the neutron. The flight parabola is the sequence of straight segments in this picture. The energy scale follows from uncertainty relations: $b = \hbar/2\tau$, and for UCN, is $\simeq 10^{-2}$ neV. The quantity b is introduced into the postulated nonlinear Schrödinger equation with the logarithmic term. The wave function in this equation depends on variables attributed to the novel degree of freedom. The stationary solutions of the Schrödinger equation form the energy spectrum. It is further suggested that transitions to different states of internal degree of freedom take place during wall encounters, with corresponding small changes in the neutron kinetic energy.

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

It must be mentioned that some evidence for very small ($\sim 10^{-2}$ neV) energy changes of UCN with energies close to the boundary energy in traps covered with Fomblin oil was found several years ago in [9]. It was pointed out, however, that the effect could be caused by mechanical vibrations excited by the vacuum pump.

It is shown in this paper that the results described in the publications [2, 4, 7, 8] may, in principle, be explained by UCN interaction with surface vibrational modes of viscoelastic liquid.

For example, the result [7] for the Fomblin surface, interpreted as rare, with a probability of 10^{-6} UCN energy transfer of 3 neV, may be described as a diffusion-like energy spread $\Delta E \simeq \delta E \cdot n^{1/2}$, where δE is the 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 spread. The results [7] may be fitted if we set $\delta E \approx 3 \cdot 10^{-3}$ neV.

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 process leading to the observed phenomena and the order of magnitude estimations of the observed effect.

2 Neutron interaction with liquid surface modes

We solve first the problem of the neutron interaction with a surface wave. In what follows we reproduce shortly the standard method for solving the problem by the perturbation method (see for example the V. K. Ignatovich's book in [1]. The Schrödinger equation describing the UCN interaction with the wall $z = 0$ is:

$$\Big[i\hbar\frac{\partial}{\partial t} + \frac{\hbar^2}{2m}\Delta - V_0 \cdot \theta(z) - \delta V(\vec{r}, t)\Big]\psi(\vec{r}, t) = 0, \qquad (1)$$

where $V_0 = \frac{\hbar^2}{2m}4\pi\sum_i N_i b_i^{coh} = \frac{\hbar^2}{2m}v_0$ is unperturbed wall potential, N_i is the number of nuclei in a unit volume, b_i is the neutron coherent scattering length, m is the neutron mass, $\theta(z)$ is the step function, which is equal to unity at $z > 0$, and is zero at $z < 0$. $\delta V(\vec{r}, t) = \frac{\hbar^2}{2m}\delta v(\vec{r}, t)$ is the perturbation potential due to surface fluctuations.

The solution of Eq. (1) is:

$$\begin{aligned}\psi(\vec{r}, t) &= \psi_0(\vec{r}, t) + \int G(\vec{r}, t; \vec{r}', t')\delta v(\vec{r}', t')\psi_0(\vec{r}', t')d^3r'dt' = \\ &\psi_0(\vec{r}, t) + \delta\psi(\vec{r}, t), \end{aligned} \qquad (2)$$

where the unperturbed solution with the incident neutron wave $e^{i(\vec{k}_0\vec{r} - \omega_0 t)}$, is

$$\psi_0(\vec{r}, t) = e^{i(\vec{\kappa}_0\vec{\rho} -- \omega_0 t)}Y_{k_0}(z). \qquad (3)$$

In Eq. 3 $\vec{k}_0 = (\vec{\kappa}_0, k_{0\perp})$.

$Y_k(z)$ is one of two linearly independent solutions of one-dimentional equation

$$[Y_k''(z) + k^2 - v_0 \cdot \theta(z)]Y_k(z) = 0 \qquad (4)$$

$$Y_k(z) = \Big(e^{ik_\perp z} + \frac{k_\perp - iK}{k_\perp + iK}e^{-ik_\perp z}\Big)\theta(-z) + \frac{2k_\perp}{k_\perp + iK}e^{-Kz}\theta(z), \quad (5)$$

where $K = (V_0 - k_\perp^2)^{1/2}$.

The second solution of Eq. (4) is

$$Z_k(z) = e^{-ik_\perp z}\theta(-z) + \Big(\frac{K - ik_\perp}{2K}e^{Kz} + \frac{K + ik_\perp}{2K}e^{-Kz}\Big)\theta(z). \quad (6)$$

With these two solutions it is possible to construct Green function for Eq. (1):

$$G(\vec{r},t;\vec{r}',t') = 1/(2\pi)^3 \int (1/2ik_\perp) e^{i\vec{\kappa}(\vec{\rho}-\vec{\rho}')-i\omega(t-t')} \cdot$$
$$[\theta(z-z')Y_k(z)Z_k(z') + \theta(z'-z)Y_k(z')Z_k(z)]\, d^2\kappa d\omega. \qquad (7)$$

The perturbation term

$$\delta v(\vec{r},t) = v_0 \left[\theta(z - \xi(\vec{r},t)) - \theta(z)\right] \qquad (8)$$

can be simplified in the case of very small deformations of the surface to

$$\delta v(\vec{r},t) = -v_0 \xi(\vec{r},t)\delta(z). \qquad (9)$$

Spatially periodic surface wave with wave vector $\vec{q}$ and frequency $\omega_{\vec{q}}$ is

$$\xi(\vec{r},t) = \xi(\vec{\rho},t) = \xi_{\vec{q},\omega_{\vec{q}}} e^{i(\vec{\kappa}\vec{\rho}-\omega t)}. \qquad (10)$$

After caculations it is possible to obtain

$$\delta\psi = 2e^{i(\vec{k}\vec{r}-\omega t)} \xi_{\vec{q},\omega_{\vec{q}}} k_{0\perp} \frac{k_{0\perp} - iK_0}{k_\perp + iK} \qquad (11)$$

The probability of neutron scattering with wave vector change $\vec{k}_0 \rightarrow \vec{k}$ $(\vec{k} = (\vec{\kappa}, k_\perp))$ is

$$w_{\vec{q},\omega_{\vec{q}}} = 8k_{0\perp}k_\perp < \xi^2_{\vec{q},\omega_{\vec{q}}} > \left| \frac{k_{0\perp} - iK_0}{k_\perp + iK} \right|^2 \delta(\vec{k} - \vec{k}_\pm), \qquad (12)$$

where

$$\vec{k}_\pm = (\vec{\kappa}_\pm, k_{\perp\pm}), \quad \vec{\kappa}_\pm = \vec{\kappa}_0 \pm \vec{q}, \quad k^2_\pm = k^2_0 \pm 2m\omega/\hbar,$$
$$k^2_{\perp\pm} = k^2 - \kappa^2_\pm = k^2 - (\kappa^2_0 + q^2 \pm 2\kappa_0 q \cdot cos\phi), \qquad (13)$$

where ϕ is the angle between $\vec{\kappa}_0$ and $\vec{q}$. The "plus" and "mines" signs correspons to the neutron upscattering and downscattering cases.

We are interested in very small energy changes when the scattered neutron remains to be the sub-barrier one. In this case K is real, and $|(k_{0\perp} - iK_0)/(k_\perp + iK)|^2 = 1$.

Our goal is to calculate the probability of neutron scattering as a function of the final neutron energy $w(\vec{k}_0 \to E)$. To do it we first use the relations

$$w_{\vec{q},\omega_{\vec{q}}}(\vec{k} \to k) = \int k^2 \cdot w_{\vec{q},\omega_{\vec{q}}}(\vec{k} \to \vec{k}) d\vec{\Omega}_{\vec{k}},$$

$$\int \delta(\vec{k}_0 - \vec{k}) d\Omega_{\vec{k}} = 2/k \cdot \delta(k_0^2 - k^2) \tag{14}$$

Thus we have

$$w_{\vec{q},\omega_{\vec{q}}}(\vec{k} \to k) = 16 \cdot \delta(k^2 - k_\pm^2) k k_{0\perp} k_\perp < \xi^2_{\vec{q},\omega_{\vec{q}}} > \tag{15}$$

Then we must perform integration over all possible surface waves:

$$w(\vec{k}_0 \to k) = \int w_{\vec{q},\omega_{\vec{q}}}(\vec{k} \to k) d^2\vec{q} d\omega \tag{16}$$

using the surface mode dynamic structure factor $S(q, \omega)$:

$$S(\vec{q}, \omega_{\vec{q}}) = (2\pi)^3 < \xi^2_{\vec{q},\omega_{\vec{q}}} > \tag{17}$$

The dynamic structure factor for isotropic viscoelastic medium has the form [10]:

$$S(q,\omega) = \frac{8k_B T \cdot Re[\eta(\omega)] q^3}{|D(q,\omega)|^2} \left\{ 1 + \frac{1}{2Re[\alpha(q,\omega)]} - 2Re\left(1 + \frac{1}{1 + \alpha(q,\omega)}\right) \right\}, \tag{18}$$

where the surface mode dispersion relation is $D(q, \omega) = 0$, with

$$D(q,\omega) = (i\omega + 2\nu(\omega) q^2)^2 - 4\nu(\omega)^2 q^4 \left(1 + \frac{i\omega}{\nu(\omega) q^2}\right)^{1/2} + \gamma q^3/\rho. \tag{19}$$

In these expressions $\nu(\omega) = \eta(\omega)/\rho$ is the kinematic viscosity, and the complex, frequency dependent viscosity, which crosses from viscous behaviour at low frequences to elastic behaviour at high frequences is

$$\eta(\omega) = G\tau/(1 + i\omega\tau), \tag{20}$$

$$\alpha(q,\omega) = \left(1 + \frac{i\omega}{\nu(\omega)q^2}\right)^{1/2}, \tag{21}$$

k_B is the Boltzmann constant, T is the temperature, γ is the surface tension, ρ is the density, τ is the liquid polymer reptation time and G is the frequency independent shear modulus of the polymer network.

In this simple Maxwell model the surface dynamics of liquid for any $(\vec{q},\omega)$, as well as the interaction of neutrons with liquid surface is determined by four parameters T, ρ, G and τ.

The probability

$$w(\vec{k}_0 \rightarrow E) = \frac{16k_{0\perp}}{(2\pi)^3}\frac{1}{2\hbar}\int S\left(q, \frac{\Delta E}{\hbar}\right)\vec{k}_\perp \cdot q \cdot dq \cdot d\phi \tag{22}$$

Figure 1 shows the results of computations of $w(\vec{k}_0 \rightarrow E)$ (neV^{-1}) according to Eq. 22 as a function of UCN energy change ΔE for monoenergetic neutrons with an incident energy $E_0 = 10\,\mathrm{neV}$, having an angle $\pi/4$ with a normal to the liquid surface. Different combinations of G and τ at the condition $\eta(\omega = 0) = 1.5\,\mathrm{p}$ [11, 12] were used in these calculations. Integration over surface wave vector q was performed within interval ($1 - 10^7 cm^{-1}$) relevant to our energy change ΔE range of interest. At small $\Delta E < 1\,\mathrm{neV}$ the probability of scattering is symmetric function around $\Delta E = 0$.

It is possible to calculate also the mean-square displacement of the fluctuating liquid surface:

$$< \xi^2 >= \frac{1}{(2\pi)^3}\int S(q,\omega)d^2\vec{q} \cdot d\omega \tag{23}$$

As is well known [13] it diverges logarithmically in the low q limit. Our calculation in the limits for q: $q_{min} = 10^{-2} cm^{-1}$, $q_{max} = 10^{8} cm^{-1}$, and for ω: $\omega_{min} = 1 s^{-1}$, $\omega_{max} = 10^{10} s^{-1}$, yields $< \xi^2 >^{1/2} \sim 6$ Å. The calculation of $< \xi^2 >^{1/2}$ in the limits for q: $q_{min} = 3 \cdot 10^{4} cm^{-1}$, $q_{max} = 10^{8} cm^{-1}$, and for ω: $\omega_{min} = 1 s^{-1}$, $\omega_{max} = 10^{10} s^{-1}$, yields $< \xi^2 >^{1/2} \sim 2.2$ Å which does not contradict significantly to experimental data for water [14] $< \xi^2 >^{1/2} =$ 3.24 Å, obtained in the appropriate experimental conditions for the low q limit.

It is seen from Fig. 1 that in result of many ($10^2 - 10^4$) reflections from the surface, UCN energy spread of the order $10^{-1} - 1$ neV is experimentally measurable effect.

The calculations for low viscosity liquid, where the very low frequency capillary waves due to surface tension dominate, yield much lower UCN energy spread. Analysis of the ω dependence of the dynamic structure factor of Eq. 18 [10] shows that the main contribution to UCN energy spread of interest ($10^2 \leq \omega \leq 10^5$) comes from (visco)elastic surface modes when the surface tension is negligible. In the latter case the dispersion relation is $\omega \simeq (E/\rho)^{1/2} q$ with wave velocity $v = (E/\rho)^{1/2} \simeq 10 - 10^3$cm/s, which is very low in comparison with the solids due to very low liquid state shear modulus E. Contrary to solids the major part of the energy of thermal liquid surface fluctuations is concentrated at low frequences. Therefore the amplitudes of these fluctuations $< \xi^2 > \sim k_B T/\omega^2$ are large, which leads to large (in comparison with Rayleigh elastic waves at the surface of solids) probability of neutron interaction with surface waves, but with much lower neutron energy change.

3 Acknowledgments

I am grateful to Dr. V. K. Ignatovich for his valuable consultation about his previous work on UCN interaction with matter.

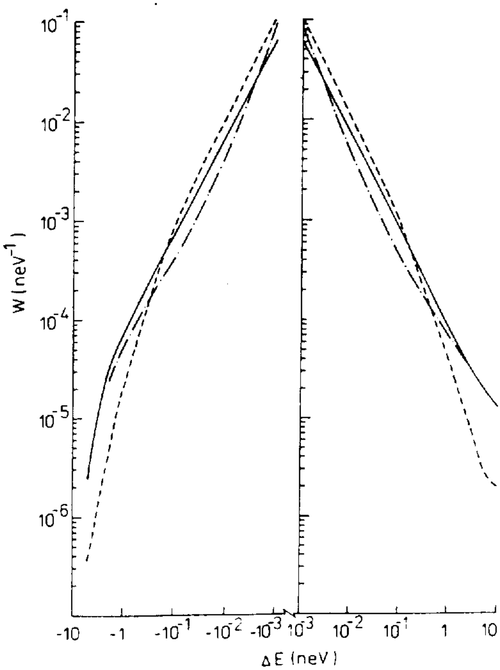

Fig. 1 Results of computations of $w(\vec{k}_0 \rightarrow E)$ (neV^{-1}) according to Eq. 22 as a function of UCN energy change ΔE for monoenergetic neutrons with an incident energy $E_0 = 10\,\mathrm{neV}$, having an angle $\pi/4$ with a normal to the liquid surface. Different combinations of G and τ at the condition $\eta(\omega = 0) = 1.5\,\mathrm{p}$ [11, 12] were used in this calculations: solid line - $G = 1.5 \cdot 10^2$, $\tau = 1. \cdot 10^{-2}$; dashed line - $G = 1.5 \cdot 10^5$, $\tau = 1. \cdot 10^{-5}$; dotted line - $G = 1.5 \cdot 10^8$, $\tau = 1. \cdot 10^{-8}$.

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Received by Publishing Department
on October 13, 1998.

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Покотиловский Ю.Н. E17-98-288
Взаимодействие ультрахолодных нейтронов
с поверхностными колебаниями жидкости
как возможная причина энергетического расплывания нейтронов
при их хранении в ловушках с жидкими стенками

В недавних экспериментах наблюдались слабые нагрев и охлаждение ультрахолодных нейтронов при их хранении в ловушках. Показано, что в ловушках, стенки которых покрыты жидким полимером (фомблином), причиной энергетического расплывания нейтронов может быть взаимодействие нейтронов с поверхностными колебаниями жидкости.

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

Сообщение Объединенного института ядерных исследований. Дубна, 1998

Pokotilovski Yu.N. E17-98-288
Interaction of Ultracold Neutrons
with Liquid Surface Modes as a Possible Reason
for Neutron Energy Spread during Long Storage in Fluid Wall Traps

Small ultracold neutron cooling and heating during long storage in closed traps has been observed in recent experiments. It is shown that interaction of ultracold neutrons with surface modes of liquid polymer (fomblin) may be a possible reason for the spread in the energy of the neutrons during long storage in closed traps.

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

Communication of the Joint Institute for Nuclear Research. Dubna, 1998

Макет Т.Е.Попеко

Подписано в печать 26.10.98
Формат 60 × 90/16. Офсетная печать. Уч.-изд. листов 1,13
Тираж 310. Заказ 50970. Цена 1 р. 35 к.

Издательский отдел Объединенного института ядерных исследований
Дубна Московской области