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SEARCH FOR LOW-ENERGY UPSCATTERING OF ULTRACOLD NEUTRONS FROM A BERYLLIUM SURFACE

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## 1 Introduction

There is the well-known and long-standing puzzle of ultracold neutron (UCN) storage times in closed volumes or, equivalently, of anomalous losses of UCN upon reflection from the inner surfaces of UCN traps. The most surprisingly large discrepancy in the experimental and predicted loss coefficients was observed for the most promising materials for high UCN storage times: cold beryllium [1, 2] and solid oxygen [3]. The anomaly consists in an almost temperature independent (in the temperature interval 10-300 K) wall loss coefficient ( $\sim 3 \cdot 10^{-5}$ ), corresponding to the extrapolated inelastic thermal neutron cross section  $\sigma^* \sim 0,9b$ . This experimental figure for Be is two orders of magnitude higher than the theoretical one, the latter being completely determined at low temperatures by the neutron capture in Be (0.008b). The experiment/theory ratio for a very cold oxygen surface achieves three orders of magnitude [3]. Approximate universality of the loss coefficient for beryllium and oxygen, and the independence of the Be figures from temperature, forces one to suspect a universal reason for this anomaly. A series of experiments to find the channel by which UCN leave the trap are described in [2]. None of the suspected reasons has been confirmed: surface contaminations by dangerous elements with large absorption cross-sections, penetration of UCN through possible micro-cracks in the surface layers of Be, the hypothetical process of milliheating of UCN due to collisions with a low frequency vibrating surface, the upscattering of UCN due to thermal vibrations of the wall nuclei. This last item deserves special and more careful consideration.

According to the description of the experiment [4] (the conclusion[2] about the absence of UCN upscattering from the beryllium surface at liquid nitrogen temperature is entirely based on the experiment[4]), upscattered neutrons prior to entering the neutron detector passed through 1.5 mm of copper, 1.1 mm of stainless steel and 2 mm of Al. For the isotropic distribution of upscattered neutrons this means that the efficiency of the registration of upscattered neutrons with energies of 0.5meV was less than 0.2 and decreasing at lower neutron energies. The reported [2] cross- section of the upscattering of UCN from the Be surface at a surface temperature of 80 K was equal to

0.14b with an uncertainty of 30%, therefore, it is quite possible that the UCN upscattering takes place to the energy range below 1meV. This hypothesis does not contradict the observed[2] temperature independence of anomalous losses of UCN, if the vibrations causing this upscattering are not thermal in nature. The frequency of these vibrations (possibly, surface waves) is in the range of  $10^8 - 10^{12} Hz$ . From the purely experimental point of view (without going into any hypotheses about the reasons for the UCN anomalous losses) this lowenergy upscattering channel is almost the only one that has not yet been investigated with the conclusive results.

Additional, qualitative considerations in favor of the possible high frequency surface sound wave UCN upscattering comes from a rough coincidence of the typical surface roughness correlation length  $T \sim$ 300 - 500 Å, UCN wavelength  $\lambda$  is close to these values, and possible surface sound wavelength. This coincidence may, in principle, to increase the UCN upscattering probability due to some kind of "resonance": If the surface sound velocity  $c = 10^5 cm/s$ , then the upscattered neutron energy  $E = hc/\lambda \simeq 10^{-4} eV$ , which is just outside of the investigated[2] energy range.

Recently the new results have been published of the experiments on searching for the UCN upscattering from the beryllium foil surface [5, 6] with the use of gas counters for the upscattered neutron detection. According to[5], the total (to the energy range  $10^{-7} - 10^{-2}eV$ ) reduced upscattering cross-section was equal  $0 \pm 0.2b$  at liquid nitrogen temperature, and  $0 \pm 0.3b$  at room temperature. The first result does not contradict the early data[2], but the second one is in serious contradiction with the previous results. The authors[5] point out that these figures are not final, and "these values were obtained after the subtraction of the large background from the trap walls and separating foil and it is necessary to increase the accuracy of measurements to establish these values".

On the other hand, the publications[6, 7] give quite different figures for the upscattering loss factor at the UCN reflection from the Be surface:  $1.47\pm0.15\cdot10^{-4}$  and  $1.39\pm0.18i0^{-4}$  for two different beryllium samples at room temperature, and  $2.6\pm0.3\cdot10^{-5}$ , and  $1.7\pm0.2\cdot10^{-5}$ at liquid nitrogen temperature for the same two beryllium samples. According to the usual formalism with which UCN interaction with surface is considered, the UCN reflection probability as a function of the normal component of the neutron moment  $p_{\perp}$  is:

$$w = 2\eta \cdot x/\sqrt{1-x^2}, \qquad x = p_\perp/p_{bound},$$
  
the loss factor  $\eta = Imb/Reb, \qquad Imb = \sigma_{inel}/2\lambda,$  (1)

where b is the coherent scattering length of the wall substance,  $p_{bound} = 2\hbar (\pi Nb)^{1/2}$  is the boundary moment of UCN, characterizing the reflecting wall,  $\sigma_{inel}$  is the sum cross-section of all inelastic processes of UCN interaction with the wall surface.

Using the above relations, it is easy to obtain from [6, 7] the reduced to the thermal point inelastic scattering cross sections for the two beryllium samples  $\sigma_{in} = 2\lambda \cdot Reb \cdot \eta$ :  $4.1 \pm 0.4b$  and  $3.9 \pm 0.5b$  for the room temperature samples, and  $0.73 \pm 0.08b$  and  $0.48 \pm 0.06b$  for the liquid nitrogen samples.

It is seen from the above that the results[2, 5, 6, 7] are in serious contradiction with each other.

For the sake of completeness it is necessary to mention also the results of the measurements[8] of the UCN upscattering probability to the thermal energy range from the surface of the room temperature beryllium foil after different high temperature procedures:  $18.5 \cdot 10^{-4}$  before any heating of the sample,  $10.9 \cdot 10^{-4}$  after heating at the 450C,  $11.5 \cdot 10^{-4}$  after heating at 700C and subsequent 5 min exposure to the atmospheric air, and  $6.2 \cdot 10^{-4}$  after heating at 700C in vacuum.

Taking into account that according to[8] the mean velocity of the stored UCN was measured to be  $\simeq 3.2m/s$  it is possible to transform these figures into the reduced to the thermal point UCN upscattering cross- section, if to assume that the imaginary part of the wall potential is attributed to this cross-section according eq.(1). This procedure gives the following values for the reduced upscattering cross-sections for the cases mentioned above: 76, 16, 17, and9b respectively per one atom of the wall. These and above[2, 5, 6, 7] figures of the reduced upscattering cross-section lead to unrealistically large concentrations of hydrogen in the surface layer of wall if to take into account that the reduced UCN upscattering cross section at the hydrogen atom at room temperature is  $\simeq (7-8)b$ . It means that this usual procedure to relate

by means eq.(1) the upscattering cross-section and imaginary part of the wall potential is incorrect and may serve only for the unification of results of different experiments.

# 2 Experimental method

The measurements were performed on the test channel of the UCN turbine source at ILL[9]. The scheme of irradiation is shown in Fig.1.



Fig.1 The scheme of the experiment for the search of the low energy upscattering of UCN from Be surface. 1. Vacuum stainless steel chamber  $\phi 60x0.5mm$ . 2. Vertical UCN guide  $\phi 60x0.5mm$ , height 120cm. 3. Rouleau of aluminium foils with beryllium deposition. 4. Cylindrical stack of In foils. 5. Detector of UCN ( $He^3$  propotional counter). 6. Vertical UCN guide  $\phi 60x0.5mm$ , height 60cm. 7. Shielding (borated polyethylene).

UCN enter the stainless steel cylindrical chamber 1 ( $\phi = 60mm$ , wall thickness 0.5mm) through the vertical (height 120cm) stainless steel neutron guide 2, and bounce from the surface of the specimen 3 made of aluminium foils covered with a beryllium layer (thickness of the deposition is  $(2-3) \cdot 10^3$  Å). The specimens had the form of a corrugated ribbon rolled into a spiral with an overall (two sides) area of  $\sim 0.5m^2$  or  $\sim 0.25m^2$ . The upscattered neutrons leaving the trap penetrate the cylindrical stack of In foils surrounding the tube and activate them with the activation cross-section according to the inverse velocity law. The In foils were  $5-50\mu m$  thick and were manufactured by means of electrolytic deposition on the surface of  $10\mu m$  copper foil. The homogeneity of the In thickness was verified thoroughly by the method of cutting the test foils into numerous small specimens and weighting these small specimens, and was found to be better than 5%. The density of the UCN flux in the trap was calibrated by means of the activation measurement of the flux of upscattered UCN from small polyethylene samples located in the center of the irradiation chamber and monitored with the <sup>3</sup>He proportional counter 5 located after the UCN trap and connected with the trap via a vertical neutron guide 6 through a small  $(0.5cm^2)$  hole. The measured in this way UCN flux at the beryllium sample was  $\simeq 40cm^{-2}s^{-1}$ .



Fig.2. Results of Monte Carlo simulations of UCN detector (<sup>3</sup>He counter with aluminium membrane) efficiency for two different positions of the detector in respect to the irradiation chamber: dashed line - horizontal one, and solid line - at the level 60cm lower, and for two different probabilities  $\eta$  of UCN diffuse reflection during the transport from the foil to detector through the stainless steel neutron guide with diameter 60mm: a -  $\eta = 0.1$ ; b -  $\eta = 0.5$ . The chamber is connected with the neutron guide through a small  $0.5cm^2$  hole.

The efficiency of the UCN detector in the geometry of the experiment was simulated with the Monte Carlo method in different assumptions about the probability of diffuse UCN reflection from the neutron guide walls between the small hole and the membrane of the detector. Figure 2 shows the results of this simulation.



Fig.3. Activation of the stack of indium foils as a result of the UCN upscattering from the polyethylene scatterers with the surface area:  $1 - 8.8cm^2$ ,  $2 - 5.3cm^2$ , 3 - the empty stainless steel chamber.



Fig.4. The measured effective polyethylene sample area obtained from indium foil activation measurements as a function of the polyethylene scatterer surface area.

With the known efficiency of the UCN detector it is possible to determine from these measurements the effective areas of the polyethylene samples and to compare them with the real ones. Figures 3 and 4 show the results of the measurements of activation of indium stacks for two polyethylene samples with the surface areas of 5.3 and  $8.8cm^2$ .

The response function of the activation of the stack of indium foils was calculated by the Monte Carlo method. The upscattered neutrons were assumed to fly out of the Be scatterer isotropically, having their starting points on the surface of the Be spiral. The reflection and absorption of the upscattered neutrons along their trajectories were rigorously taken into account.

The detailed results of these simulations will be published elsewhere [10].

The activity of the In foils was measured with the high efficiency  $(\sim 70\%) 4\pi$  scintillation  $\beta$ -counter with the active  $(4\pi$  plastic anticoincident counter) and passive (lead) shielding. The area of the In foils whose activity is measured simultaneously is  $\sim 200 cm^2$ . The background of the counter was about  $1.05s^{-1}$  in these measurements. The efficiency of the counter was carefully measured for different thicknesses of irradiated In and Cu foils. The description of the counter and results of the calibration will be published in[10].

This method of measuring the slow neutron spectra with the help of the activation of a stack of In foils was calibrated by means of the irradiation of such stacks in the beam of monochromatic thermal neutrons and in the precisely measured (with the time-of-flight method) quasi-Maxwellian spectrum of cold and thermal neutrons. The measured and calculated distributions of the foil activity along the stacks were in good agreement.

In case of the absence of any low energy anomalous upscattering UCN acquire energy from the thermal vibrations of beryllium lattice, and with greater probability from the vibrations of the surface contaminations - mostly hydrogenous. The spectrum of the upscattered neutrons has "thermal" character but it is not known. The information about the spectrum of possible "anomalous" upscattering is even more obscure. Therefore, the measured In foil activity as a function of position in the stack (thickness coordinate) was approximated under very general assumption that the spectrum of upscattered neutrons consists of two Maxwellian flux components: one with  $v_{th} = 2.2 \cdot 10^5 cm/s$ ("normal" upscattering from room temperature Be) and the second one with low  $v_0$ , the latter being chosen in the interval 10 - 300m/s(anomalous upscattering).



Fig.5. The results of computer indium foil stack "activation experiments" and subsequent restoration of the incoming upscattered neutrons in the assumption that the spectrum consists of two Maxwellian flux components: one with  $v_0 = 2.2 \cdot 10^5 cm/s$  and the second one with  $v_0 = 10^4 cm/s$ , the latter had the weight 1/20 of the first one. The results of computer "activation" of indium foil stack before the restoration procedure were statistically Gaussian-distributed with a standard deviation 5% for each foil.

The overall thickness of indium stacks did not exceed in our mea-

surements  $250\mu m$ ,  $(n\sigma \sim 0.3 \text{ for the isotropic thermal neutron flux})$ , therefore, the sensitivity of establishing the upscattered thermal neutron spectrum is not good. But it was demonstrated by rigorous Monte Carlo simulation [10] that it is possible not only to distinguish the low energy component of the upscattering from the large thermal background, but also to perform rough spectrometry of this low energy part of the spectrum.

Figure 5 demonstrates some results of the computer indium stack "activation experiment" and restoration of the incoming spectrum of upscattered UCN in the assumption that the spectrum consists of two Maxwellian flux components: one with  $v_0 = 2.2 \cdot 10^5 cm/s$  and the second one with  $7v_0 = 10^4 cm/s$ , the latter had the weight 1/20 of the first one.

The results of computer "activation" of indium foil stack before restoration procedure were statistically Gaussian-distributed with a standard deviation 5% for each foil. It is seen that the method is able to restore with high reliability the small low energy admixture to the intensive thermal background, but it is not dependable in extracting from the indium activation data the thermal component of the spectrum.

## 3 Results and Discussion

Figure 6 shows the measured activity of irradiated In foils as a function of the thickness coordinate for two beryllium samples with different areas.

In our measurements we used the compact surface sample with the increased area so that the mean gap between the adjacent turns of our spiral-like ribbon sample with the area  $\sim 0.5m^2$  was about 1mm. The question arises about the effective using in the UCN upscattering the full surface area of the sample with such narrow channels for UCN diffusion between the adjacent turns. Therefore, the additional activation measurements were performed with the sample of the area  $\sim 0.2m^2$  with  $\sim 2.5$  times larger gaps bitween the adjacent turns. Figure 7 representing the measured indium stack activity as a function of the sample surface area shows good proportionality between

the area and activity. This is the evidence of the uniform and effective UCN upscattering over the full sample area.



Fig.6. Measured In foil activation points as a function of indium thickness coordinate for two different beryllium samples areas: full points -  $0.5cm^2$ , empty points -  $0.2cm^2$ 



Fig.7. Total UCN upscattering probability from the beryllium sample as a function of the sample area.

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The total measured flux of upscattered UCN from the beryllium sample with area  $\simeq 0.5m^2$  was  $\simeq 50s^{-1}$ .

As was mentioned, the method has low reproducibility in extracting the spectrum of the thermal component of upscattered UCN. Therefore, the experimental data were processed in different reasonable assumption about the temperature of the thermal component. Figure 8 represents the 90% exclusion contours for the cross-sections  $\sigma_{anom}^*$  and  $\sigma_{th}^*$  deduced from the In foil activation measurement of upscattered UCN fluxes from the normal temperature Be surface assuming that the thermal component of upscattering represents the Maxwellian neutron flux with room temperature.



Fig.8. The 90% exclusion contours for the cross sections  $\sigma_{anom}^*$  and  $\sigma_{th}^*$  deduced from the In foil activation measurement of upscattered UCN fluxes from the normal temperature Be surface in the assumption that thermal component of upscattering represents the Maxwellian neutron flux with room temperature. The contours are presented for three different characteristic velocities  $v_0$  of anomalously (low-energy) upscattered UCN assumed to have a Maxwellian flux form:  $1 - v_0 = 15m/s$ ;  $2 - v_0 = 50m/s$ ;  $3 - v_0 = 200m/s$ .

The contours are presented for three different characteristic velocities  $v_0$  of anomalously (low-energy) upscattered UCN assumed to have a Maxwellian flux form:  $v_0 = 15m/s$ ;  $v_0 = 50m/s$ ;  $v_0 = 200m/s$ .

As is seen from Fig.8 the reduced UCN upscattering cross-section to the thermal energy range is very high for not outgassed beryllium  $\sim 22b$ , which does not contradict to the result of[5] and some of the results[8]. We attribute so large upscattering cross-section partly to the presence in the incoming UCN spectrum of neutrons with the energies larger than the boundary energy of beryllium, but mostly to upscattering from the aluminium cuts of sample ribbons, not covered with beryllium layer. In both these cases this upscattering takes place not on the sample surface but in the bulk of aluminium. This increased thermal upscattering was not very essential for our search for the the low energy anomalous component in the upscattered neutron spectrum, but it increased the thermal background.

Figure 9 shows the 90% CL restriction curves for the reduced crosssection as a function of the characteristic velocity  $v_0$  of the Maxwellian flux of anomalous low energy UCN upscattering from the room temperature beryllium sample in different assumptions about the characteristic velocity  $v_{th}$  of the thermal flux.



Fig.9. The 90% CL restriction curves for the reduced cross-section as a function of the characteristic velocity  $v_0$  of the Maxwellian flux of anomalous low energy UCN upscattering from the room temperature beryllium sample in different suppositions about the characteristic velocity  $v_{th}$  of the thermal flux:  $1 - v_{th} = 1600m/s$ ;  $2 - v_{th} = 2200m/s$ ;  $3 - v_{th} = 2800m/s$ .

In addition to (1) the following formulae were used in the data processing. Averaged over isotropic angular distribution loss probability of UCN with the velocity v upon reflection from the surface with the boundary velocity  $v_b$ :  $\bar{\mu}_{loss} = 2\eta(\arcsin(y) - y\sqrt{1-y^2})/y^2$ , where  $y = v/v_b$ .  $\tilde{\mu} = \int_0^{v_{lim}} \bar{\mu}f(v)dv$  is the UCN loss coefficient averaged over the normalized UCN flux spectrum  $f(v) = 4v^3/v_{lim}^4$ , which is the lowenergy tail of the Maxwellian spectrum. In our case,  $v_{lim} = 3.9m/s$ .

As may be seen from Fig.9 we were not able to completely exclude in this experiment low energy UCN upscattering in all energy interval of interest:  $0.1 - 10^{3} \mu eV$ , but significant part of this energy range:  $\simeq (1 - 200) \mu eV$  is excluded in our measurements. The main reason for that was comparatively high  $\beta$ -counter background (higher than  $1s^{-1}$ ), comparatively low UCN intensity (the flux at the PF2-UCN test channel is about order of the magnitude lower than all the rest UCN channels of ILL UCN source), and short measurement time.

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