Experimental studies of very cold neutrons passing through solid deuterium

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The studies of spectral dependence of neutron passing through solid deuterium has been carried out with a vertical beam of very cold neutrons with the wave length of $\lambda \sim 40 - 150$ Å. The work results show the dependence of observed neutron scattering sections on the way of preparation of a solid deuterium sample and on ortho-para composition of deuterium.

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1. Introduction. The ability of ultra-cold neutrons (UCN) to be stored in material and magnetic traps makes them an effective instrument for experimental physics of low energies. The most significant experiments in this field are searches for a static electric dipole moment of neutron (EDM), measurements of the free neutron lifetime and neutron β-decay angular correlation constants. At the present time the most accurate value of neutron lifetime [1] and limits for the value of neutron EDM [2, 3] has been obtained utilizing UCN. The scientific impact of these measurements of the neutron's particle properties is sweeping. For example, the existence of a non-zero neutron EDM would mean a violation of the fundamental CP-symmetry in particle physics. Violation of this symmetry is manifest in the observed asymmetry between matter and antimatter in the universe. In fact, the matter/antimatter asymmetry is believed to be inconsistent with the standard model of particle physics, a belief which has triggered a proliferation of proposed extensions to the standard model which incorporate new phenomenology, such as supersymmetry. One or two orders of magnitude increase in the sensitivity of neutron EDM measurements could serve as a decisive test for many of these proposed models.

However, methodical improvements of these neutron experiments will not result in significantly increased accuracy without a remarkable (by orders of magnitude) increase in the intensity of UCN sources. The first practical step in achieving increased UCN intensities utilizing a solid deuterium (SD_2) UCN source at liquid helium temperature was done in PNPI, Gatchina, Russia [4 – 6]. The advantages of SD_2 can be best utilized under conditions of somewhat smaller heat loads than conventional reactor cold sources, therefore a pulsed mode with a spallation neutron source was proposed [7]. At present, work concerning creation of new high-intensity SD_2 UCN sources is being carried out in several international scientific centers: LANL – USA, PSI – Switzerland, TUM – Germany. For source calculations and UCN yield evaluations, theoretical values of cross-sections for the interaction of very cold neutrons (VCN) and UCN with SD_2 have been used. More recently, detailed calculations of the incoherent, inelastic scattering processes and measurements of UCN lifetime and production in SD_2 as a function of the para-deuterium concentration and temperature have been performed at LANL [8, 9]. However, no direct experimental cross-section data in this neutron energy range are available.

The present work is an attempt to study experimentally the passage of very cold neutrons through SD_2. As a solid deuterium target was utilized the apparatus described thoroughly in the work [10]. The spectral dependence of neutrons passing through the target has been studied by means of time-of-flight techniques.

2. Experimental setup. The experiment scheme is presented in the Fig.1. A bent vacuum-processed mirror neutron guide (*2*) is installed at the exit of the horizontal neutron guide (*1*), which is connected to the SD_2 cold neutron source of the PNPI WWR-M reactor [5]. The bent neutron guide serves to form the vertical beam of VCN. It is made of a sheet polished stainless steel, the neutron guide cross-section is $60 \times 140\,mm^2$, bending radius – 1 m. The neutron guide output window is made of aluminum with 0.5 mm thickness. The cadmium stop (*4*) with a $14 \times 140\,mm^2$ slot is installed before the output window inside the neutron guide vacuum jacket. The mask slot is placed closer to the outer (larger radius) wall of the curved neutron guide and serves to cut out

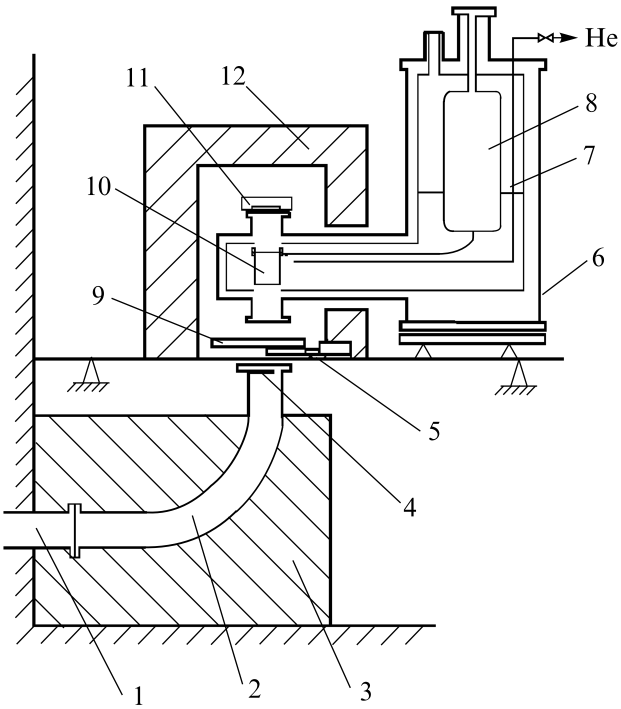

Fig.1. Experimental setup: *1* – horizontal neutron guide; *2* – bent mirror neutron guide; *3* – combined protection; *4* – cadmium mask; *5* – monitor detector; *6* – target-cryostat; *7* – liquid nitrogen tank; *8* – helium tank; *9* – neutron beam chopper; *10* – deuterium vessel; *11* – main neutron detector; *12* – polyethylene shielding

a part of the neutron beam, which runs mainly along this wall. The shielding (*3*) provides for absorption of the direct beam of particles from the horizontal channel and ensures satisfactory background conditions for the scattering measurements.

The neutron beam chopper (*9*) is placed over the neutron guide. The chopper itself is a flat disc with a radial slot, which is made of a neutron-absorbing material. The chopper rotation frequency is $1500\,\mathrm{min}^{-1}$. A detector (*5*) which monitors the flux of neutrons incident on the target is placed between the chopper and the neutron guide. It blocks a part of the neutron guide output cross-section and provides a method to control the intensity of the initial neutron beam.

The target-cryostat (*6*) is located over the chopper. The input and output windows of the cryostat vacuum jacket and the target internal vessel (*10*) with the studied sample are made of zirconium with 0.2 mm thickness. The primary neutron counter (*11*) is placed over the cryostat output window. Proportional ^{3}He-counters serve as the neutron detectors. The distance between the chopper (*9*) and the detector (*11*) is 50 cm.

Deuterium with $D_2 = 99.79\%$ was used in this work, the primary contaminants were HD and H_2, the percentage of the N_2 and O_2 were less then $2 \cdot 10^{-2}$. Deuterium is confined in an internal zirconium vessel (*10*) with a heat exchanger. The target is cooled by flowing helium from the He liquid reservoir (*8*). It is possible to have both liquid and solid deuterium in the hydrogen vessel thanks to regulation of the cold gaseous helium consumption through the target heat exchanger. The SD_2 layer thickness is determined by the volume of gas condensing into target, taking into account the volume of displacers (heat exchanger and lithium-6 shielding).

Neutrons scattered in the target are captured by a 2 mm thick protective shielding made of metallic ^{6}Li. This shielding is placed in the internal vessel of the cryostat and forms the operating volume of the target with a cross-section of $104 \times 40\,\mathrm{mm}^2$ and a height of 196 mm. At that the heat exchanger and the side walls of the zirconium vessel do not irradiate with the scattered neutrons. The high lithium heat conductivity provides for a good leveling of temperature along the entire target volume.

The time-of-flight neutron spectra were measured without a target and with an empty target. They show that the beam of neutrons, which are distributed along the external wall of neutron guide and reach the main detector, is characterized by a wave length of 40–150 Å with a maximum within 60–65 Å interval. It corresponds with the energy range of $E_n = (0.4 - 5) \cdot 10^{-5}\,\mathrm{eV}$. An example of such a spectrum is presented in the figures. Neutrons with a wave length less than 35–40 Å are not able to pass through the neutron guide with our bending radius. There are practically no neutrons with $\lambda > 150$ Å (close to ultra-cold and also UCN itself) in the registered spectrum, firstly, due to their very high angular spread at the end of neutron guide, secondly, due to a high probability of being captured in the air and the material of neutron guide and target "windows".

3. Ortho-deuterium and its preparation. Molecular deuterium can be divided into two states based on their rotational spectra. These states differ by a relative location of spins of deuteron nuclei forming the molecule. If the total nuclear spin of the molecule is denoted by **S**, then these two states are: ortho-deuterium ($\mathbf{S} = 0,\ 2$) and para-deuterium ($\mathbf{S} = \mathbf{1}$). The total wave function of the molecule is symmetric. This means that ortho-deuterium can only have rotating states with even **J**, whereas para-deuterium can only have rotating states with odd **J**. The molecular vibrational states do not get excited under normal conditions.

The ground state energy of the ortho-deuterium rotational band is 7 meV less than that of the para-deuterium molecule. At a given temperature the equilibrium ortho-para composition is determined by the ratio of level populations with even and odd rotational states. At room temperature the ortho-para concentration ratio is 2/1, and such deuterium is called "normal". At low temperatures it is possible to obtain a higher concentration of ortho- deuterium. For instance, at a temperature of 20 K the equilibrium concentration C_{ortho} is $\sim 98\%$. Spontaneous transitions into the equilibrium concentration is a very slow process, however, relaxation into the equilibrium state can be accelerated by paramagnetic catalysts.

As a catalyst a finely-dispersed $Fe(OH)_3$ powder was used in our work. Deuterium targets with different ortho-phase concentration were obtained by passing gas through a cooled container with the catalyst. The container with iron hydroxide was either placed in a bath with liquid nitrogen or was cooled by cold helium down to temperatures of about 20–25 K in a conversion apparatus [10]. Reverse conversion without a catalyst runs very slowly, therefore after conversion the gaseous deuterium was transported into the target by "warm" supplying lines, and then was condensed and frozen there. Analysis of the deuterium ortho-para concentration was carried out using a gas chromatograph LCM-8MD. The measured gas samples were taken directly from the target.

4. Measurement procedure and results. The measurements were carried out in the following way. First, after cooling the cryostat, neutron spectra with an empty target were measured. The total count rate in the primary neutron detector for an empty target was ~ 70 n/s. Neutron spectra were accumulated in a series of 500 second runs. These runs were then summed in the analysis, taking into account the monitor detector readings. Adequate statistics were typically obtained after a few hours of operation in this mode. The target was then filled with a certain volume of deuterium without removing the cryostat from the beam and without disturbing the positioning of the apparatus. The condensation time of $V = 232$ normal liters of gaseous D_2 to liquid deuterium took roughly 2 hours. This volume of gas corresponds to approximately 3 cm of SD_2 along the beam axis. In different experiments we had the layers of SD_2 3 cm and 6 cm. A main uncertainty in the pathlength is connected with an error of calculations of volume of displacers immersed into deuterium. It is about 5%.

Further cooling of the sample could be drive at different speeds by regulating the flow of cold helium through the target heat exchanger. The first cooling of samples from the triple point of deuterium (18.7 K) down to temperatures below 6 K took approximately one hour. Subsequent neutron spectra were then measured over a time interval of a few hours. The state of the deuterium was controlled by regulating the vapour pressure over the liquid or solid deuterium. Additional temperature sensors (carbon Allen-Bradley resistors) were installed on the jacket of the deuterium vessel.

Then deuterium in the target was heated and fully melted at the temperature of $T \sim 19$ K, and after that it was frozen again down to the temperature of $T < 6$ K. The second freezing was done slower than the first one, over an interval of about two hours. The spectra of neutrons were measured anew.

In all measurements the intensity of the initial neutron beam was recorded in the monitoring detector. This permitted us to normalize out changes in the transmitted neutron flux due to reactor power changes and changes in the cold neutron source temperature.

The normalized time-of-flight spectra for neutrons transmitted through the SD_2 samples and the empty target were used to calculate the spectral dependence of the cross-section for neutron interaction with SD_2. As the capture cross-section of deuterium for VCN is small, changes in the transmitted neutron flux were due to scattering processes. The neutron background was taken into account in cross-section calculations. It was evaluated by counts in the first and last channels of the time-base and amounted to an average 0.8 n/s when integrated over the entire spectrum.

Total cross-sections for neutron interactions with the studied sample of SD_2 were calculated using the following formula:

$$\sigma(\lambda) = \frac{\ln\left[N_0(\lambda)/N_L(\lambda)\right]}{n \cdot L} \tag{1}$$

where N_0 and N_L denote the total number of counted neutrons for an empty target and a target with SD_2 respectively; n is the number of deuterium molecules in a volume of 1 cm^3, and L is the sample length along the neutron beam. Errors presented in the figures show the statistical uncertainty of calculation σ only.

The experiments were performed with deuterium having the various ortho-para compositions: 66%, 72% and 93% ortho-phase. The dependencies of "transparency" of the deuterium target on the speed of freezing were observed. For example Fig.2 depicts the cross-section curves for deuterium with a concentration of the ortho-phase $(93\pm3)\%$. For our construction of the cooled volume of the target the best neutron transmissions were obtained at the freezing time about two hours with all samples. The neutron cross-sections $\sigma(\lambda)$ are congruent in the short-wave part of neutron spectrum, but they differ with each other for large λ. The observed difference can be explained only by different sample struc-

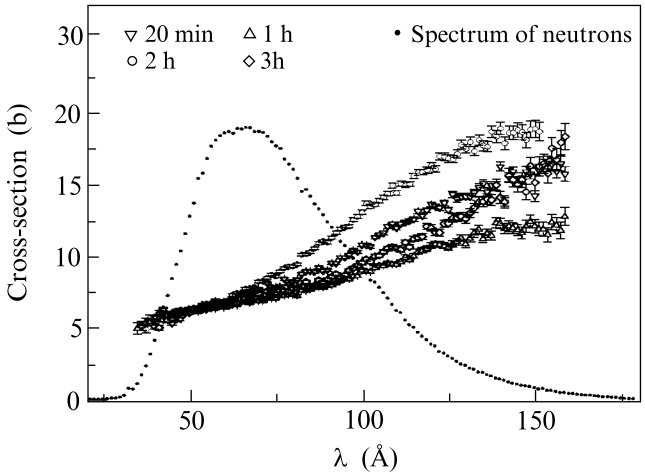

Fig.2. Cross-section as a function of neutron wavelength for deuterium with a concentration of the ortho-phase $(93 \pm 3)\%$ and for different time of freezing

tures obtained at the different manufacturing procedure of a SD_2. It is connected, apparently, with the "ice" quality: variety of crystal sizes, heterogeneities of density in the sample volume, cracks, etc... Because the neutron wavelength sets a rough upper limit on the size scale over which neutron transport is sensitive to structural variations, it also seems reasonable that the sample macrostructure must have the strongest influence in the long-wave part of the spectrum. This trend is also evident in our data.

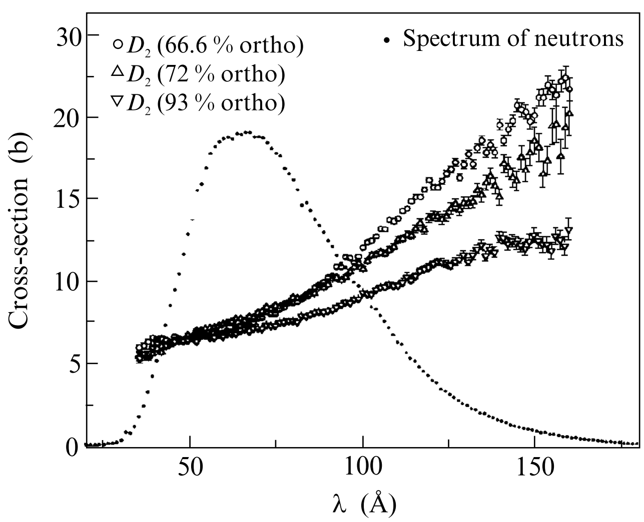

Fig.3. Cross-section as a function of neutron wavelength for different ortho-para ratios and for the same time of freezing – 2 hours

To obtain a dependence of the scattering cross-section on the ortho-para composition of deuterium we compared the data obtained with the like procedures of preparation of samples. Fig.3 depicts cross-section data for different ortho-para ratios and time of freezing 2 h. A dependence of the scattering cross-section on the deuterium ortho-para composition is observed for large λ. For instance, for neutrons with a wavelength of about 150 Å the total cross-section for interaction with normal deuterium molecules (66% of ortho-phase) is approximately 1.5 times greater than those for ortho-deuterium ($\sim$ 93%). The behaviour of cross-sections for neutrons with $\lambda > 150$ Å can not be extracted from these data due to the poor statistics in these parts of spectrum, and due to a relatively large uncertainty in our knowledge of the backgrounds.

The work gives a demonstration of the importance of the SD_2 manufacturing technology for VCN and UCN sources since the yield of these sources depends on the deuterium "transparency" for neutrons. First the experimental dependence for the total cross-section of the VCN scattering on ortho-para composition of the deuterium was observed.

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