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Experimental Study of a Solid-deuterium Source of Ultracold Neutrons

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The results of experimental studies of the yield of ultracold neutrons (UCN) from solid deuterium, which were performed on a model source in the WWR-M reactor at the St. Petersburg Institute of Nuclear Physics, are reported. The temperature dependent gain factor in the UCN yield from solid deuterium at 13-14K relative to the UCN yield from a gaseous state at room temperature is equal to 1230 and it is 550 at solid-deuterium temperature of 18.7K (triple point).

KEYWORDS: ultracold neutrons, neutron sources

§.1. Introduction

The possibility of increasing the density of ultracold neutrons (UCN)by using as a solid deuterium source at low temperatures was analyzed in.[1] In the present letter we report the results of an experimental study of UCN yield from solid deuterium, performed on a model UCN source in the WWR-M reactor at the St. Petersburg Institute of Nuclear Physics.

§.2. Experimental procedure

Fig.1 show the arrangement of the source in the reactor.

The 6-liter zirconium chamber of the source (cylinder D = 150 mm, l = 350 mm with elliptic bottoms) has a double wall (2×0.5 mm) where helium flows into the gap from a 150-W cryogenic refrigerator at a temperature of 4.5K. As the chamber is cooled, the deuterium from a 6-m^3 receiver flows into the chamber, condenses, and solidifies.

To obtain deuterium temperatures below 10-12K, a special construction of the source chamber is required. The problem is that as the temperature decreases, the thermal contact between the cooled wall of the chamber and deuterium breaks down. Chilling is possible as long as the saturated-vapor pressure does not fall below several Torr. For example, the saturated-vapor pressure is equal to only 0.75 Torr at a temperature of 12K and 5×10^{-2} Torr at 10K. A possible technical solution is to place an additional cooled spiral tube on the inner wall of the chamber. On cooling, the tube will be squeezed by the deuterium, which should solve the problem of thermal contact. Before making the design of the chamber more complicated, however, we decided at the first stage of the investigation to use a simple design, and then in the next stage to use the same design but with deuterium containing a small quantity of helium in order to ensure heat transfer, and finally to use a more sophisticated design of the source if necessary. In the present letter we report the result of the first stage of the study at temperatures above 13K.

One of the most complex problems in this investigation is to determine the temperature profile of the solid-deuterium source. However, the temperature of the source could be easily determined with low accuracy -according to the residual deuterium pressure in the receiver, since the source, which is connected to the receiver by a pipeline, is a vapor-pressure thermometer in which the saturated vapor pressure is determined by the source temperature. Unfortunately, because of the large volume of the receiver, the relaxation time of the process which establishes an equilibrium pressure is long, which results in a pronounced hysteresis of the experimental dependence of the UCN yield on the pressure in the receiver.

§.3. Experimental results and analyses

The UCN gain factor as a function of the pressure in the receiver during the cooling and heating of the source is shown in Fig.2.

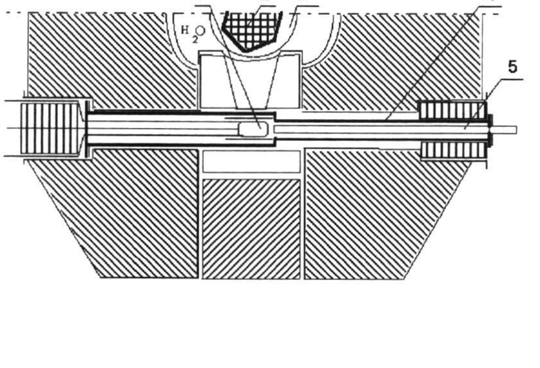

Fig.1. Arrangement of the solid-deuterium source in the reactor. 1 — chamber with solid deuterium; 2 — reactor core; 3 —beryllium reflector; 4 — vacuum container; 5 — UCN guide

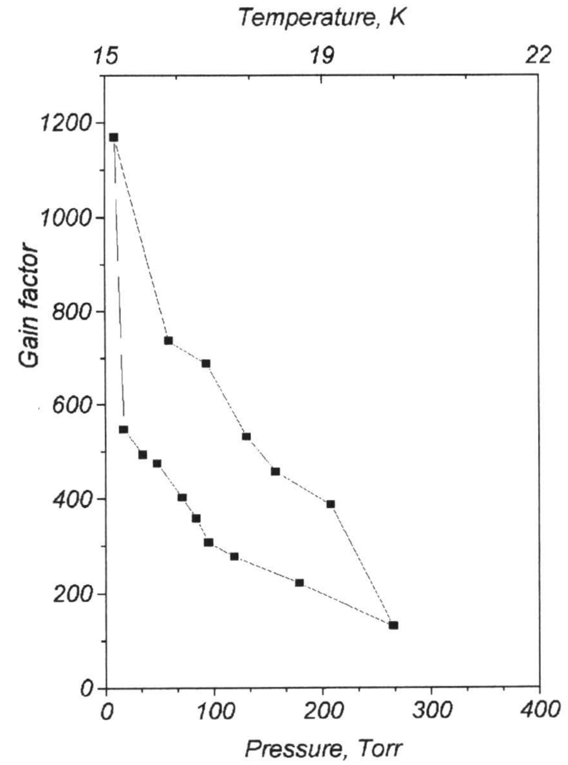

Fig.2. Measurement of the UCN yield upon cooling down of the source (upper curve) and the heating of the source (lower curve)

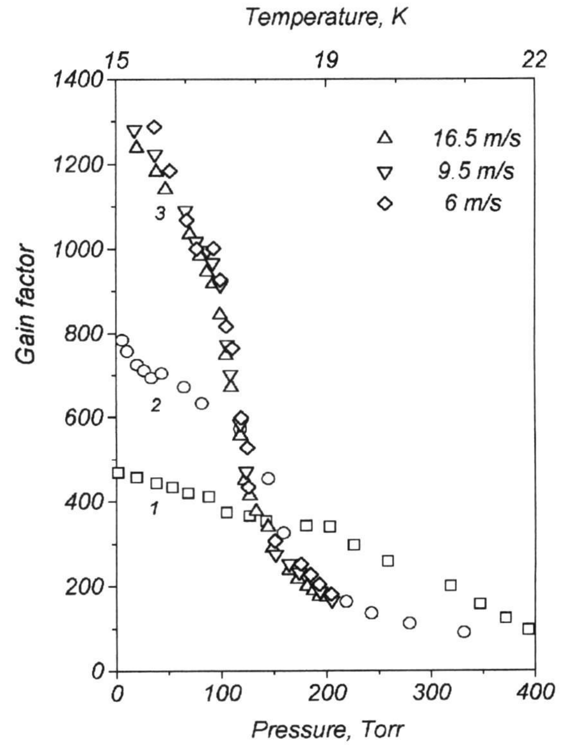

Fig.3. Measurement of the UCN yieldwith the first chillings of the source: curves *1, 2, 3* (the measurements at the curve *3* have been carried out with UCN (6 m/s) and with VCN (9.5 m/s; 16.5 m/s))

Another, but not so trivial, uncertainty in the UCN yield that was observed in the experiment is apparently associated with a change in the ortho-para composition of deuterium under the influence of low temperatures and the radiation from the reactor. Curve *1* in Fig.3 was obtained in the first experiment on cooling of the source.

This experiment was performed at a rapid rate and low reactor power (2 MW). The gain factor referred to room temperature, named here the temperature gain factor was equal to only 450 in this experiment. Subsequent cooling of the source(curve *2* in Fig.3) was performed at a slow rate and with a reactor power of 14 MW; the temperature gain factor was equal to 790. After the source was warm, virtually all of the deuterium in the receiver was in the ortho phase (95 ± 5 %) and remained in this phase without any visible changes in the composition. Concentration of para phase was measured by means of chromatography method. In subsequent cooling of the source, the temperature gain factor reached 1230 and stayed at this level. The curve *3* in Fig.3 corresponds to the fourth cooling. The dependence of the UCN yield on the ortho-para composition of deuterium is the most probable explanation for this phenomenon.

One obvious problem in obtaining a high UCN yield from a solid-deuterium source is that the source must be transparent to UCN. Cracking of solid deuterium as a result of the large temperature stresses could cause the UCN to be scattered by nonuniformities (cracks). When the effective path between nonuniformities is less than the path determined by the inelastic scattering and capture cross sections for UCN, the gain factor no longer increases as the temperature decreases. To investigate this question, we measured the temperature factor of the gain simultaneously for UCN with an average velocity of 6 m/s and for very cold neutrons in two velocity ranges, so that the average velocities were equal to 9.5 m/s and 16.5 m/s. It was done by using neutron guide system, which consist of branches with the different radius of curvature. The neutron guide curvature determine the average velocity of extracted neutrons(6 m/s, 9.5 m/s, 16.5 m/s). This was arranged using UCN storage trap and deflecting neutron guides with different radius of curvature. Since the refraction index depends strongly on the neutron velocity, the effective turbidity of the solid deuterium due to cracking should be manifested primarily for UCN (the refractive indices for 6 m/s, 9.5 m/s, and 16.5 m/s are, respectively, 0.68, 0.886, and 0.964). Curve *3* in Fig.3 is represented by measurements for all neutron velocities indicated above. No appreciable difference in the temperature dependence of the neutron yield for different velocities is observed. This shows that the turbidity of the medium is not yet manifested. The free path at a deuterium temperature of 13K is, according to the calculations, 4 cm, 8 cm, and 16 cm for velocities of 6 m/s, 9.5 m/s, and 16.5 m/s, respectively. Since there is no dependence in the gain factor, it can be concluded that the depth of transparency of solid deuterium in any case is not lower than the indicated values. A more detailed study of this question requires lowering the temperature of the source to 5-6K.

In addition to measuring the UCN yield during cooling heating of the source with a refrigerator (Fig.2), we measured the dependence of the UCN yield with the source heated by radiation with the refrigerator switched off. These experiments made it possible to determine more accurately the gain factor at the triple-point temperature for solid and liquid deuterium, and also the power of the radiation load. Figure 4shows the UCN yield and the pressure in the receiver during heating at a constant thermal load (radiative heating). During the first 28-30minutes (t_0 - t_2) the pressure in the receiver increases linearly and is determined mainly by the heat of sublimation. Linear increase continues until an equilibrium sublimation pressure of 128 Torr at the triple point is established. During the next 25-30 minutes (t_2 - t_3) the solid deuterium melts and the liquid evaporates at the same time by the heating. At this time, the flow of gas into the receiver slows down, since some power is expended on melting. The melting process is completed by the time t_3, after which the flow of gas into the receiver again becomes constant and is determined by the heat of evaporation of deuterium. The heating power, which is equal to 36 W when the reactor is running and 6 W when the reactor is shut down, is determined from the rate of influx of the gas.

A plateum is present in the time dependence of the UCN yield between the times t_1 and t_2. This plateum corresponds to the triple-point temperature.

Initially, at time t_0, the temperature gain factor is equal to 1230 at a temperature of 13-14K. At the time t_1 the deuterium is heated to 18.7K and because of sublimation, this temperature remains constant up to the time t_2, when the pressure in the receiver reaches the equilibrium sublimation pressure. The temperature gain factor for solid deuterium near the triple point is equal to 500-550. At the time t_3, when the deuterium is completely liquid and its temperature is 19.5-20K, the gain factor is equal to 120. Further decrease of the .gain factor is attributable to heating of the liquid.

Appreciable increase in the yield during the crystallization is due to the improved thermalization of the neutron flux in the solid deuterium as compared to liquid deuterium, because of the increase in the total interaction cross section due to Bragg scattering. Though the Bragg scattering is elastic one, it increase the time of wandering in the source and influence on the thermalization. This effect was observed directly in the measurements of the total spectrum of neutrons from the source. Figure 5 shows the time-of-flight spectrum before condensation of deuterium (curve *1*), after condensation (deuterium in the liquid phase (curve *2*), and after crystallization (deuterium in the solid phase, curve *3*). Apparently, the most probable explanation of gain factor at the solidification is related with decreasing of upscattering cross section for UCN, when the phase transition is happened.

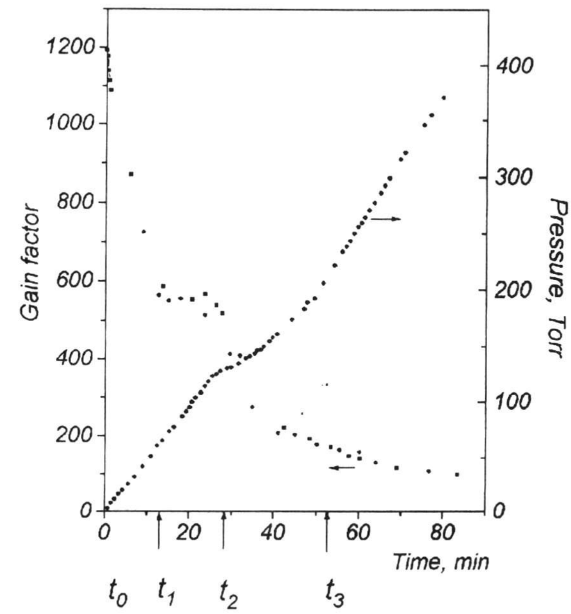

Fig.4. Time diagrams of UCN gain factor and deuterium pressure in the tank at the heating of the source by radiation from reactor.

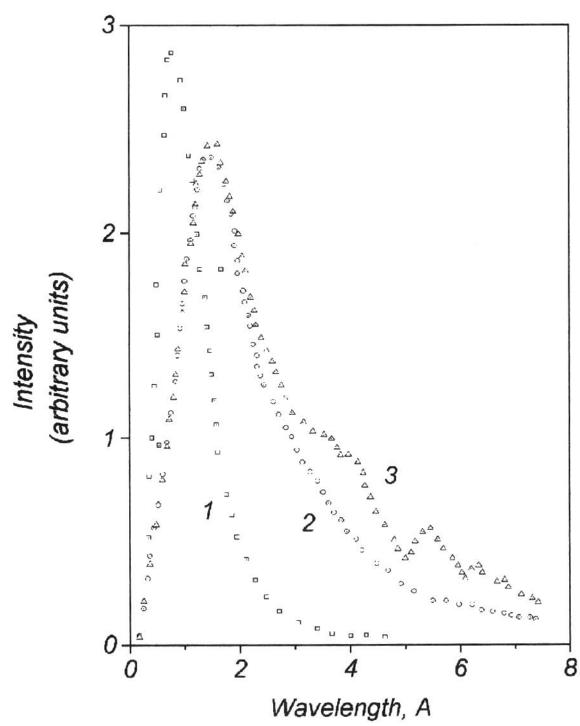

Fig.5. Time–of–flight spectra of the source for different phase states of deuterium.

§.4. Considerations on source neutron spectrum

The effective temperature of the spectrum of the neutrons incident on the source is equal to 600K. Thermalization in the deuterium substantially softens the spectrum, and a difference in the thermalization effect for the liquid and solid phase is observed in the region 4-6 Å. For the solid phase we observed in the neutron spectrum characteristic irregularities associated with Bragg reflections. It is interesting to note that the structure of the reflections was found to be different for the first and second coolings, which could be attributed to the dependence on the ortho-para composition and on the cooling rate. The neutron spectrum from a solid-deuterium source (curve 3 in Fig.5) can be represented as a sum of two spectra: 56% of the intensity with an effective temperature of 180K and 44% of the intensity with effective temperature of 30K. The calculation of the UCN yield with the indicated shape of the spectrum was found to be very close to the calculation with an effective temperature of 100K. Neutrons with wavelength greater than 3 Å can become UCN through a one-phonon process. An increase in the fraction of long-wavelength neutrons therefore increase the UCN yield upon crystallization of the deuterium. As one can see, the solid-deuterium source is more efficient for production of cold and very cold neutrons.

Another reason for UCN gain factor increasing at the crystalization can be related that upscattering cross section for solid state is smaller than for liquid. It can increase the depth of UCN yield. Both effects - improvement thermalization and supression upscattering cross section - gave together factor 4-4.5 in the increasing of UCN intensity at the crystalization of deuterium.

The final analysis of the experimental results is illustrated in Fig. 6, where the experimental temperature dependence (curve 1) and the results of previous investigation for a 1-liter liquid-deuterium source[2] (curve 2) and for a 0.15-liter solid-deuterium[3] (curve 3), are shown with the computation results for absolutely pure deuterium and for effective neutron flux temperatures of 40, 100, 300 and 500K. The curve 4 was computed for an effective temperature of 100K and absorbtion cross sections in deuterium taking into account the hydrogen impurity (0.2 vol.%) and nitrogen impurity (4.6×10^{-3} vol.%).

The UCN yield is increased by decreasing the temperature of the source and by decreasing the effective temperature of the neutron flux as a result of increasing the volume of the source. It should be mentioned that calculated gain factors are normalized at the room temperature (T_{D_2} = 300K) of the source and the room temperature of the neutron flux (T_n = 300K). Experimental normalization (curve 3) was done at the room temperature of the source, but at the temperature of reactor neutron flux 600K. Therefore discrepancy between curve 3 and curve 4 at the source temperature (13 ÷ 19)K is not dramatic, it arised because of perfect thermalization of neutron flux is not happened inside the reactor moderator.

§.5. Concluding remarks

The next stage of the investigations presupposes that the temperature of the source is lowered to 6-7 K for the purpose of studying the possibility of increasing the UCN yield. It should be noted that the thermal conductivity of solid ortho deuterium is an order of magnitude higher at 6-7 K. This should improve the properties of the source with respect to the thermal load.

The experimental results can be used to develop designs for a solid-deuterium source of cold and

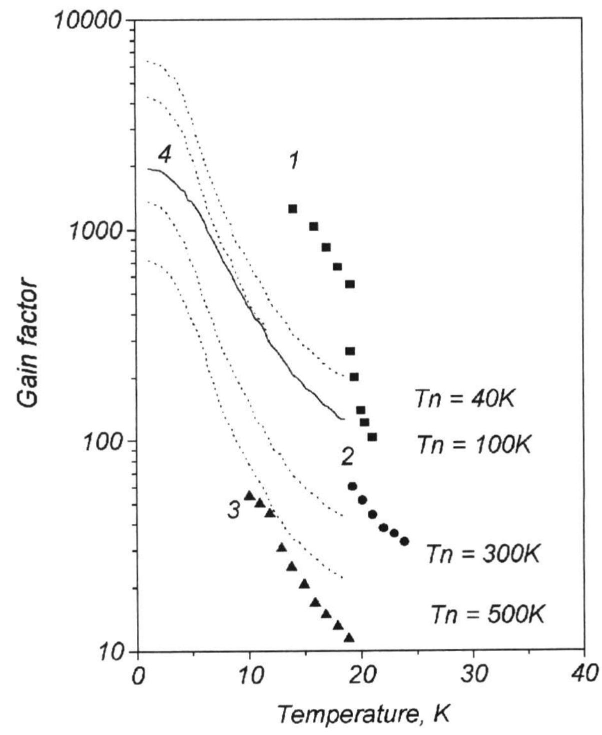

Fig.6. Analysis of the UCN gain factor for deuterium sources with different volumes.

ultracold neutrons in high-flux reactors with a heavy-water reflector, since the heavy water provides effective shielding from fast neutrons and γ-rays and gives a low level of heat load while preserving a high flux of thermal neutrons (the PIK reactor under construction in Gatchina, Russia and the ILL reactor in Grenoble, France, new project of heavy water reactor at Garching, Germany). The other possibility is to use a solid-deuterium source based on neutron spallation sources, where the ratio of the heat load and the neutron flux is appreciably better than for reactors. For example, a solid-deuterium UCN source based on a 1-MWspallation source is being designed at Los Alamos (USA).This type of source could be proposed for spallation neutron source at the Japan Hadron Project (KEK), also for European Spallation Source (ESS).

There is a big interest to develop project of the special source of cold neutrons (CN) very cold neutrons (VCN) and ultra cold neutrons (UCN) in accordance with scheme shown on the Fig.7, where the spallation neutron target is surrounded by the volume with solid deuterium in the bath with liquid helium to provide maximal thermalization of neutron flux.

At last, the solid deuterium neutron source can be successfully used at reactors with moderate power. For example, the placement of solid deuterium neutron source in the thermal column of the reactor KUR(Japan) or the reactor WWR-M (Russia) could significantly increase experimental facilities.

It should be mentioned that solid deuterium source can give almost the same gain factor for very cold neutrons (30-50 m/s) therefore it can be used in the scheme of UCN extraction by means of turbine.

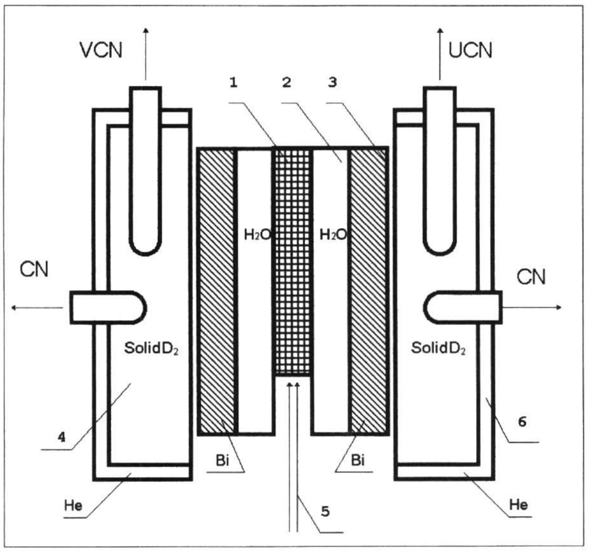

Fig.7. 1 - spallation target, 2 - H_2O moderator, 3 - Bi - shielding, 4 - solid D_2 moderator, 5 - proton beam, 6 - He 4K bath .

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