# **Experimental report**

**Proposal:** 3-14-374 **Council:** 10/2016

**Title:** Measurement of the mean free path of UCN in liguid and solid hydrogenand deuterium at various temperatures

Research area: Nuclear and Particle Physics

This proposal is a new proposal

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Samples: hydrogen

deuterium

Instrument	Requested days	Allocated days	From	To
PF2 EDM	35	47	19/01/2017	07/03/2017

#### Abstract:

In recent UCN transmission experiments on liquid and solid o-D2 we were able to successfully determine the UCN scattering cross sections in the low-energy range (3-14-311 and 3-14-351). In a forthcoming experiment (3-14-370) we plan to measure these cross sections for p-H2 (liquid, solid) as well.

The above-mentioned UCN experiments are extremely time-consuming, as good statistics have to be achieved over the whole range of UCN energies (100...2000 neV). Hence, the data collection so far was possible for only 1 temperature per state (liquid, solid, p-H2, o-D2)

We now want to determine the UCN mean free path length  $l_mfp = (n*sigma_tot)^-1$  at a set of 3-4 temperatures in the liquid and the solid state for H2 and D2 each. In order to reduce the data acquisition time, we will take data in the region of the UCN flux maximum (v = 12.6m/s) only.

Through this we will be able to better determine the life time  $tau_0 = l_mfp/v_UCN$  of UCNs in o-D2 and p-H2, which are known with insufficient accuracy at the moment. These data will enable us to better estimate the UCN density that can be achieved in UCN sources working with o-D2 (or p-H2) moderators at low temperatures.

# **Experimental Report**

for Proposal Number: 3-14-374 (09/2016), at the instrument PF2-EDM

# Measurement of the mean free path of ultracold neutrons (UCNs) in liquid and solid hydrogen and deuterium at various temperatures

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Experimental Period: 19 January – 8 March 2017

#### Motivation

Even after decades of neutron scattering work, the precise measurement of UCN cross sections of the cold hydrogens ( $H_2$  and  $D_2$ ) in the solid and liquid state is yet an unfinished story. First results were obtained in the 1960 [1-4] and even later attempts in the 1990 [5-6] only supplied us with imprecise data, which can hardly be used for the calculation and design of cold neutron (CN) and UCN sources. Recent UCN experiments [7-9] put a new emphasis on the physics of UCN moderators and led us to a first series of UCN transmission experiments on liquid and solid *ortho*-deuterium (o- $D_2$ ) [10-11].

### Aim of the Experiment

In continuation of our UCN work on cold hydrogens and considering the unsatisfactory results from a previous measurement campaign (see experimental report no. 3-14-351), we repeated UCN transmission measurements through *ortho*-deuterium, this time using an optically transparent and highly polished sample container [12, and experimental report no. 3-14-370].

#### The Experiment

The time-of-flight (TOF) measurements were carried out using the cryostat and TOF setup as described in [10,12] with slight modifications. The UCN chopper was placed in front of the deuterium sample to reduce cold neutron background, to keep the flight path rather short and to have enough space to install an optical viewport. Deuterium and hydrogen samples for Raman analyses were drawn each time before the sample container was filled and when the respective sample was evacuated. This way it could be ensured that the sample had a high *ortho*-deuterium or *para*-hydrogen content, respectively, and that this concentration remained constant during the measurement run. The Raman equiment was provided by the UCN Group at Paul Scherrer Institut in Villigen, Switzerland. The results showed that the deuterium samples had an *ortho* content of  $c_{\text{ortho}} = 0.982 \pm -0.002$  and the hydrogen samples were  $c_{\text{para}} = 0.998 \pm -0.002$  para-hydrogen.

As could be demonstrated in a previous experiment [12], the highly polished sample container windows made from amorphous silica show virtually no surface scattering. Therefore, all UCNs scattered out of the direct beam can be assumed to have undergone interactions in the sample bulk. It is hence not necessary to measure two samples of different thicknesses to separate surface from bulk scattering. Furthermore, considering the difficult sample preparation, which became apparent during optical observations, it is doubtable whether the exact same sample (same amount of defects) can be reproduced within a small error margin over and over again. Therefore, the two-thicknesses method has to be reserved for homogenous samples or samples with a high degree of reproducibility.

# Preliminary Experimental Results for Liquid and Solid Deuterium

The preliminary total cross sections (scattering plus absorption, corrected for reflection at the vacuum interface) of liquid *ortho*-deuterium ( $c_{\text{ortho}} = 0.982$ ) for UCNs are shown in Fig. 1. The experimental data for the temperatures 19.0 K, 20.6 K and 23.0 K overlap well with the model published in [10], which was calculated for the respective temperatures using the self-diffusion coefficient  $D_{\text{self}}$  of liquid deuterium from O'Reilly et al. [13] and Guarini et al. [14].

The temperature uncertainty for the experimental data was  $\pm 0.2$  K. The new experimental data on liquid deuterium should be seen as an improvement (and replacement) of the *experimental* data published in [10] and [11].

The preliminary total cross sections (scattering plus absorption, corrected for reflection at the vacuum interface) of solid *ortho*-deuterium ( $c_{\text{ortho}} = 0.982$ ) for UCNs are shown in Fig. 2. The black squares represent *normal*-deuterium at 15 K. Below them, the red circles represent solid *ortho*-deuterium at 10 K and the green down triangles stand for the same crystal at 14.5 K. The blue up triangles stand for the same crystal after the temperature cycling 10 K/14.5 K/10 K. It is clear that temperature cycling does not lower the scattering cross section of a crystal. In the best case, it remains the same. Qualitatively similar findings were published in [7,15]. In the particular case shown here, the cross section increased slightly as a result of temperature cycling. The red dots from Fig. 2 (solid *ortho*-deuterium at 10 K) are plotted again in Fig. 3. There, they are decomposed into their constituents: 1-phonon up-scattering, incoherent elastic nuclear scattering and elastic scattering contributions from defects ( $R_d = 88 \text{ Å}$ ,  $c = 8.2 \times 10^{-11} \text{ per D}_2$  molecule) as calculated using the Guinier approximation [16]. This is the first time that an estimation of the size and concentration of defects in deuterium crystals was done for the purposes of UCN scattering.

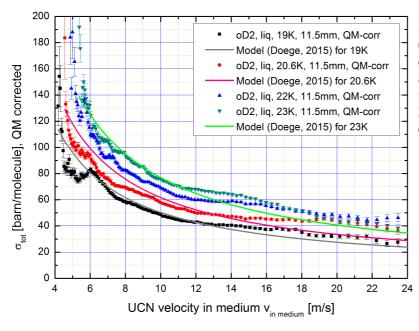


Figure 1. Preliminary experimental data (solid colored symbols) for the total cross section of liquid *ortho*-deuterium at various temperatures. The theoretical model [15] was calculated for T = 19 K/ 20.6 K/ 23.0 K at  $c_{\text{ortho}} = 0.982$  (solid colored lines).

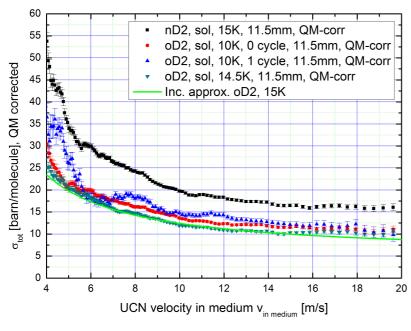


Figure 2. Preliminary experimental data for solid normal- and ortho-deuterium at T = 10 K and 15 K and after no and after one temperature cycling. The theoretical model (incoherent approximation for 1-phonon upsolid green scattering, line) calculated for T = 15 K.

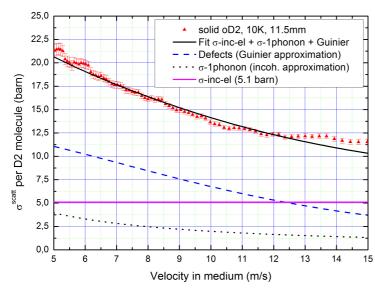


Figure 3. Preliminary scattering cross section of solid *ortho*- $D_2$  (T = 10 K) and its decomposition into 1-phonon up-scattering, incoherent elastic nuclear scattering and the elastic scattering contributions from defects ( $R_d = 88 \text{ Å}$ ,  $c = 8.2 \times 10^{-11} \text{ per } D_2 \text{ molecule}$ ) as calculated using the Guinier approximation [16].

Taking the total cross sections of liquid and solid deuterium at v = 10 m/s from Figs. 1 and 2, it is possible to plot the mean free path for UCNs in *ortho*-D<sub>2</sub> over temperature,

where  $\lambda_{mfp} = (\sigma_{tot} * N_v)^{-1}$ . In Fig. 4 it can be clearly seen that UCNs travel farther in solid deuterium than in its liquid state. For low-energy UCNs, it can clearly be inferred from Fig. 3 that elastic defect scattering is the dominant scattering process in the solid state. Therefore,  $\lambda_{mfp}$  is determined essentially by the crystal defects. Our new value for  $\lambda_{mfp}$  is about a factor of 3 smaller than the value known up to now, 8 cm, from [17].

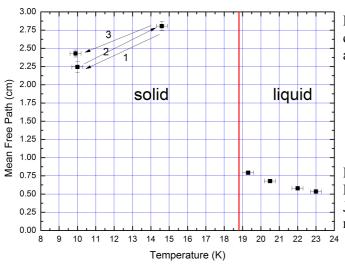


Figure 4. Mean free path length  $\lambda_{mfp}$  of UCNs in condensed *ortho*-D<sub>2</sub> ( $c_{ortho} = 0.982$ ) for v = 10.0 m/s at 6 temperatures.

Large parts of this report will be published as S. Doege et al. ISINN-25 Conference Proceedings, JINR Dubna (Russia) (2018) and in a peer-reviewed journal later on.

#### References

- [1] J. Young, J. Koppel, Phys. Rev. 135, A603 (1964).
- [2] P. Egelstaff, B. Haywood, F. Webb, Proc. Phys. Soc. 90, 681 (1967).
- [3] V. F. Sears, Canad. J. of Phys. 44, 1279 (1966).
- [4] W. Seiffert, Report No. EUR 4455d (1970).
- [5] F. Bermejo et al., Phys. Rev. B 47, 15097 (1993).
- [6] M. Mukherjee et al., Europhys. Lett. 40, 15 (1997).
- [7] F. Atchison et al., Phys. Rev. Lett. 95, 182502 (2005) and Phys. Rev. Lett. 94, 212502 (2005).
- [8] A. Frei et al., Phys. Rev. B 80, 064301 (2009).
- [9] C. Liu et al., Phys. Rev. B 62, R3581 (2000).
- [10] S. Döge et al., Phys. Rev. B 91, 214309 (2015).
- [11] S. Döge (Doege) et al. ISINN-23 Conference Proceedings, JINR Dubna (Russia), pp. 119-130 (2016).
- [12] S. Döge and J. Hingerl, Rev. Sci. Instrum. 89, 033903 (2018).
- [13] D. E. O'Reilly and E. M. Peterson. Selfdiffusion of liquid hydrogen and deuterium. *Journal of Chemical Physics* 66, 934 (1977).
- [14] E. Guarini et al. Velocity autocorrelation by quantum simulations for direct parameter-free computations of the neutron cross sections. II. Liquid deuterium. *Phys. Rev. B* 93, 224302 (2016) and priv. comm.
- [15] T. Brys, PhD thesis, ETH Zurich (2008).
- [16] A. Guinier and G. Fournet, Small Angle Scattering of X-Rays. John Wiley, New York (1955).
- [17] C.L. Morris, J.M. Anaya et al., Phys. Rev. Letters 89, 272501 (2002).