Understanding of ultra-cold–neutron production in solid deuterium

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Abstract – Our recent neutron scattering measurements of phonons and other quasi-particle excitations in solid deuterium (sD2) and the extraction of the density of states for phonons and rotational transitions in sD2 have led us to a new understanding of the production of ultra-cold neutrons (UCN) in sD2. The UCN production rate reaches a maximum at an equivalent neutron temperature of $T_n = 40$ K for a neutron flux with Maxwellian energy distribution. The cross-section for UCN production in sD2 has been determined by using the density of states $G_1(E)$ in combination with the incoherent approximation as well as by a direct calibration of our measured neutron cross-sections with the known cross-section of the $J = 1 \rightarrow 0$ rotational transition in deuterium. Using this cross-section we deduced the production rate of UCN in sD2 which agrees quite well with direct measurements of this energy averaged UCN production cross-section.

Introduction. – Ultra-cold neutrons (UCN) are slow enough to be confined [1] in traps, which can be formed by materials with a high Fermi potential (up to about 300 neV) or by a magnetic field (60 neV/T). UCN can be observed for several tens of minutes in these traps, and are excellent tools for high-precision measurements, as the life time of the neutron itself [2,3], and the search for a possible electric-dipole moment of the neutron [4] (current upper limit $2.9 \times 10^{-26} e \cdot cm$).

For further improvement of these experiments, powerful UCN sources are needed. Different groups [5–10] are working on the development of strong sources, based on sD2 as a converter for down-scattering of thermal or sub-thermal neutrons into the UCN energy region (typical $E < 300$ neV). A converter based on sD2 should be operated at temperatures below $T < 10$ K in order to avoid subsequent upscattering of UCN by phonons within the solid deuterium [11]. The understanding of the down-scattering of neutrons into the UCN region crucially relies on a detailed knowledge of the energy loss of thermal or sub-thermal neutrons in the sD2 converter material. One major reaction channel is based on the excitation of phonons in the solid-deuterium crystal by the neutrons.

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Experimental results. – We have recently investigated the phonon system in sD2 by neutron time-of-flight measurements at the IN4 (ILL Grenoble) and at the TOFTOF (FRM II, Munich). These measurements are described in detail in [12].

The measured inelastic cross-section $S(Q, E)$ contains all necessary information which is needed to determine the density-of-states (DOS) [12] or calculate directly the cross-section for UCN production with the values of $S(Q, E)$ lying on the free-neutron dispersion parabola. Figure 1 shows two examples of our neutron scattering data (angular averaged) for two different concentrations $c_o$ of ortho-D2 molecules. On the energy gain side of the neutrons (negative $E$) we can identify the rotational transition $J = 1 \rightarrow 0$. The intensity of this peak decreases with reduced para-D2 molecule concentration $c_p = 1 - c_o$. A similar rotational transition (the $J = 0 \rightarrow 1$ transition) can also be observed on the neutron energy loss side (positive $E$). We find that the elastic cross-section ($E = 0$ meV) of sD2 is quite large. Our data indicates that the elastic cross-section makes up approximately 50% of the total cross-section at thermal neutron energy. Another interesting feature is the neutron scattering intensity on the neutron energy gain side close to the elastic peak ($E = 0$ meV). An increase of the concentration of the...
Fig. 1: An example of a dynamical neutron cross-section of solid D₂ at T = 7 K. Comparison of two ortho-concentrations c₀ = 66.7% (□) and c₀ = 98% (○). Data from TOF-TOF measurements at the FRM II. Initial energy of the thermal neutrons is E₀ = 20.4 meV.

Fig. 2: S(Q, E) of sD₂ for c₀ = 95.2% at T = 4 K for different fixed values of Q. Data from IN4 measurements.

para-D₂ molecules lead to a larger neutron up-scattering close to the elastic line. The origin of this cross-section is very likely induced by phonons, which are interlinked with rotational transitions J = 1 → 0 of the para-molecules. This up-scattering has serious implications to the up-scattering of UCN in the sD₂ converter material, and reduces the achievable density of UCN in sD₂. This up-scattering was studied the first time by measuring the UCN life time in sD₂ (see [7]). A detailed analysis of these effects will be presented in a forthcoming paper.

In addition the scattering function S(Q, E) is shown for fixed values of momentum transfer Q in fig. 2. The dynamical scattering function S(Q, E) comprises the phonon branches of the hcp-sD₂ [13] crystals and the J = 0 → 1 rotational transition. The acoustical phonons (transversal and longitudinal) are clearly visible in S(Q, E) while the optical phonon branches are not. The rotational transition J = 0 → 1 shows a significant Q-dependence and it is purely incoherent. The position of the J = 0 → 1 transition peak (E₀₁ = 7.4 meV) does not change much at different Q-values. The cross-section dσ_J=0→1/dΩ for this transition is shown in fig. 3. This cross-section was extracted from our data by an integration of S(Q, E) at E₀₁ = 7.4 meV with a width of ΔE = 1 meV at different Q-values. The Q-dependence of this cross-section was fitted using the incoherent scattering length of the J = 0 → 1 transition [12]

\[
d\sigma_{J=0\rightarrow1}/d\Omega \sim j_1^2(Qa_s/2) \cdot e^{-\frac{1}{2}q^2\langle u^2 \rangle}.
\]

The parameter a_s = 0.74 Å is the distance of the deuterons within the D₂ molecule, while \langle u^2 \rangle is the mean square displacement of the D₂ molecule in the lattice. The form factor j₁ for the J = 0 → 1 transition is described by a spherical Bessel function of first order. The result for \langle u^2 \rangle ≈ 0.245 ± 0.02 Å² is in good agreement with earlier published results [13]. The peak-like deviations in fig. 3 at certain Q-values are the result of coherent neutron scattering on phonons.

For example the peak at Q ≈ 1.5 Å⁻¹ is very likely a signature of [100] longitudinal acoustic phonons, while the peak at Q ≈ 2.5 Å⁻¹ belongs to the [110] longitudinal acoustic phonon branch. At Q ≈ 2.8 Å⁻¹ the longitudinal optical branch of phonons of the [110] orientation is, maybe, seen. In principle the TOF method sees in our case (fast frozen deuterium — see [12]) only an average over all possible crystal orientations (powder average), and cannot resolve the individual phonon dispersions with high accuracy. Therefore, the assignment of the peaks in fig. 3 to different crystal orientations has to be looked at in a rather hypothetical way.

With the aid of our neutron data we are able to determine the UCN production cross-section by two ways. One way is the determination of the quasi-particle (phonons

Fig. 3: Cross-section dσ_J=0→1/dΩ of sD₂ for c₀ = 95.2% o-D₂. ■: data from IN4 measurement. Black line: fit result using eq. (1).
prepared by fast freezing (several minutes) of liquid D$_2$ to a temperature of T < 10 K. $\int_0^{E_{\text{max}}} G_1(E) \cdot dE = 1$

and rotational excitations of the D$_2$ molecules) density of states $G_1(E)$. The other way is the direct integration of the dynamical neutron cross-section in the kinematical region along the free-neutron dispersion parabola.

**UCN production cross-section — incoherent approximation.** — With the knowledge of the quasi-particle density of states $G_1(E)$ it is possible to calculate the dynamical neutron cross-section $\frac{d\sigma}{dE}(E)$ (averaged over the scattering angle, thus $Q$). Vice versa it is also possible to extract $G_1(E)$ from a measured $\frac{d\sigma}{dE}$ data using Turchin’s theory [14] for this cross-section applying the incoherent approximation. The absolute normalization of $\frac{d\sigma}{dE}$ is not needed, because the extracted $G_1(E)$ has to be normalized to unity anyway. This approach is only valid within the Born-approximation.

The method for the determination of $G_1(E)$ from our data is described in detail in [12]. Contributions of higher-order multi-phonons to $\frac{d\sigma}{dE}$ are incorporated. The result of our analysis [12] concerning $G_1(E)$ and a comparison with a Debye model is shown in fig. 4. The characteristics of $G_1(E)$ was already discussed in detail in [12] but it is worth to summarize these results also here, because they have an important impact on the UCN production cross-section. One major feature is the occurrence of these excitations above $E \sim 10 \text{ meV}$ in the one-quasi-particle density of states $G_1(E)$. These excitations above $E \sim 10 \text{ meV}$ depend on the concentration of ortho-D$_2$. The optical phonons in the region ($8 \text{ meV} < E < 10 \text{ meV}$) are not clearly visible in $G_1(E)$.

In the case of UCN production the energy transfer of the down-scattered neutron $E = E_0 - E_f$ is approximately equal to the initial neutron energy $E_0$ ($E_f = E_U \ll E_0, E_U$: UCN energy). The total cross-section for UCN production can be calculated by

$$\sigma_{\text{UCN}}(E_0) = \int_0^{E_{\text{max}}} \frac{d\sigma(E_0)}{dE} dE_U. \quad (2)$$

It is shown in fig. 5 and can be compared with recent published data of the UCN group at the PSI [15] which has studied UCN production at a cold-neutron beam ($E_0 \sim 1.4 \text{ meV}$ to $20 \text{ meV}$). The agreement between data and our calculation, using our data for $G_1(E)$ and Turchin’s incoherent approximation is very good. The calculated cross-section comprises the contribution of one-quasi-particle and two-quasi-particle excitations. Three-quasi-particle excitations do not appear below $E \sim 14 \text{ meV}$ (see fig. 9 in [12]) and are rather small in their contribution. The authors of [15] compared their results to a calculated cross-section based on a multi-phonon Debye model. They also reported, that using a “more realistic model” for the density of states (Yu et al. [13,16]) leads to a considerably worse fit of their data. Using a Debye model with a cut-off at $k_B \cdot \Theta_D = 9.5 \text{ meV}$ (Debye temperature $\Theta_D = 110 \text{ K}$) only the contribution of two-phonon excitation have to be included in order to fit the measured cross-section of $\sigma_{\text{UCN}} = 1.55 \cdot 10^{-7}$ barns at $E = 14.7 \text{ meV}$ [15]. This also applies for the DOS model of Yu et al. Our own calculated cross-section shows a combination of one-quasi-particle and two-quasi-particle excitations contributing at these energies. In detail our data indicate (see fig. 5) that the UCN production cross-section is mainly determined by one-quasi-particle excitation for energies below $E = 15 \text{ meV}$ (see fig. 4). However, the two-quasi-particle contribution cannot be neglected in the region of $E = 5–25 \text{ meV}$. 

![Fig. 4: Comparison of the one-quasi-particle density of states of solid D$_2$ for $c_o = 66.7\%$ (○) and $c_o = 98\%$ (□) at $T = 7 \text{ K}$ with a Debye model (line). Data from TOFTOF measurements (energy resolution: $\Delta E \sim 1.24 \text{ meV}$). The sD$_2$ polycrystal is prepared by fast freezing (several minutes) of liquid D$_2$ down to a temperature of $T < 10 \text{ K}$. $\int_0^{E_{\text{max}}} G_1(E) \cdot dE = 1$](image1)

![Fig. 5: UCN production cross-section of $c_o = 98\%$ solid D$_2$. UCN energy range 0–150 meV inside the solid D$_2$. Solid line: cross-section calculated in incoherent approximation, using $G_1(E)$ from [12]. Dashed line: one-quasi-particle contribution. Dotted line: two-quasi-particle contribution. □: data from measurements at the PSI [15].](image2)
The application of the incoherent approximation in the
case of sD$_2$ has certainly to be questioned. The sD$_2$ crystal
scatters neutrons more coherently than incoherently. This
fact leads to the question: Is it possible to get the UCN production cross-section directly from the neutron
scattering data?

UCN production cross-section — direct determination. — The easiest way of determining the cross-
section for UCN production is the use of the dynamical
scattering function $S(Q, E = \frac{E^2}{2m}Q^2)$ in the $(Q, E)$-phase
space along the free-neutron parabola (dispersion curve
with $E \approx E_0 = \frac{E^2}{2m}k_0^2$ and $Q \approx k_0$). This approach relies on the Born approximation. This method allows the incorpora-
tion of all coherent and incoherent contributions to the
UCN production cross-section. Possible coherent contribu-
tions, which cannot be treated exactly with the inco-
herent approximation, appear directly in the deduced
cross-section. Therefore this method is superior to the
result obtained by incoherent approximation. The basic
condition for this method is an appropriate calibration of
measured values of $S(Q, E)_{\text{data}}$.

The UCN production cross-section can be determined by

$$
\sigma_{\text{UCN}}(E_0) = \frac{\sigma_0}{k_0} S \left( k_0, \frac{h^2}{2m}k_0^2 \right) \frac{2}{5} k_{U}^{\text{max}} E_{U}^{\text{max}}.
$$

(3)

Here the term $E \approx E_0 = \frac{E^2}{2m}k_0^2$ is the energy of the incoming
neutron in the down-scattering process, while $k_{U}^{\text{max}}$ and $E_{U}^{\text{max}}$ are the upper limits for the UCN momentum
and energy. In order to obtain absolute cross-sections the dynamical scattering function of solid deuterium
extracted from our neutron scattering data has to be cali-
brated to absolute values. This can be done by using the
rotational transition $J = 1 \rightarrow 0$ as reference for calibration.
The cross-section for the $J = 1 \rightarrow 0$ transition was calculated
by Hamermesh and Schwinger [17] for ortho- and
para-deuterium molecules (gas).

These calculations include also thermal movements
(Maxwellian distribution of velocities) of the deuterium
molecules in the gas. The application of these calculations to
the rotational transitions of the D$_2$ molecules, which are
pinned in a crystal at fixed positions may of course be
questionable. However the rotational transitions in sD$_2$
are not very much hindered by the crystal binding of the
crystal [18]. Indeed the molecules are of course not
at rest, they perform motions around their equilibrium
positions. These movements are caused by the zero-point
motions and also by existing phonon excitations in the
crystal. A similar problem was considered by Lamb [19] in
the past. He calculated the capture of neutrons by atoms
in crystals. His calculations for crystals with weak lattice
binding delivered the finding that atoms in such crystals
can be treated as a gas with an effective temperature,
which depends on the Debye temperature of the crystal.
This effective temperature can be higher than the real
temperature of the solid and is just a result of the
average energy of the vibrational degrees of freedom plus
the zero-point motion. We used this ansatz to calculate
the effective temperature ($T_{\text{eff}} \approx 40$ K at a deuterium
temperature of 4 K) of the D$_2$ molecules and used it for
the calculation of the rotational transition ($\sigma_{J = 1 \rightarrow 0}$) of our
quasi-free molecules, following the theory of Hamermesh
and Schwinger.

The cross-section $\sigma_{J = 1 \rightarrow 0}$ in pure para-deuterium has a value of $\sigma_{J = 1 \rightarrow 0} = 0.61$ barns for $E_0 = 17.2$ meV (corre-
sponding to the energy of the incoming neutrons at the
IN4 experiment) at a deuterium temperature of $T = 4$ K.
The comparison (see fig.1) of our neutron scattering
data of natural deuterium ($c_o = 66.7\%$) and ortho
enriched deuterium indicated an ortho-concentration of
$c_o = (95.2 \pm 0.3)\%$.

Scaling with the para-concentration of our sample ($c_p = 1 - c_o = 4.8\%$) thus leads to a value of $\sigma_{J = 1 \rightarrow 0} = 0.029$
barns for our ortho enriched solid-D$_2$ sample. This value
for $\sigma_{J = 1 \rightarrow 0}$ can be used to calculate a scaling factor for
$S(Q, E) = \kappa \cdot S_{\text{data}}(Q, E)$.

By means of this value it is possible to calculate the
total neutron scattering cross-section for $E_0 = 17.2$ meV
neutrons: $\sigma_{\text{tot}}(E_0 = 17.2$ meV$) = 23.4$ barns. The assumption
is of course only valid, if the kinematical area of our
neutron scattering experiment covers most of the possible
scattering processes. A good cross-check of this value is
the calculated value for the total cross-section for solid
polycrystalline deuterium, using the incoherent approxi-
mation of Turchin [14].

The result of this calculation leads to a value of
$\sigma_{\text{tot}}(E_0 = 17.2$ meV$) = 23.8$ barns, which is close to the
value of our calibration. The dynamic response func-
tion $S(Q, E)$, resolved from our data analysis, was not
corrected for multiple scattering effects. Therefore the esti-
mated error of this calibration is approximately 12% (see
footnote 1). The result of this calibration and determina-
tion of the UCN production cross-section as a function of
the energy of the incoming neutrons, and a comparison
with measurements of this cross-section [15] is shown in
fig. 6. This plot contains also data, which were obtained
with higher incoming-neutron energy ($E_0 = 67$ meV) scat-
tering. The intrinsic resolution at this energy is quite
large ($\Delta E \approx 3.5$ meV) and smears out the coherent phonon
contributions. The calibration of $S(Q, E)$ at that energy
was done by using the calculated value of the total neutron
scattering cross-section at $E \approx 67$ meV within the incoherent approxi-
imation. This procedure is certainly valid, because the
kinematical area at that high energy is large, and all possi-
ble excitations of sD$_2$ are incorporated in the data.

The agreement of the absolute values of the measured
cross-section with the resolved values form our $S(Q, E)$
data is reasonable. The comparison of the calculated UCN
production cross-sections, extracted from the incoherent

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1The treatment of multiple scattering effects in our solid-
deuterium samples is discussed in detail in ref. [12].
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A UCN energy range of 0–150 neV inside the solid D₂ is assumed. Cross-section determined by an integration of \( S(Q,E) \) along the free dispersion of the neutron (sample: fast frozen solid deuterium (\( T = 4 \) K)); data from IN4 measurements. Blue □: \( E_0 = 17.2 \) meV. Red filled ◦: \( E_0 = 67 \) meV. ■: direct UCN production data from measurements at the PSI [15].

approximation and parabola method, shows (see fig. 5 and fig. 6) a discrepancy in the region of \( E \sim 6 \) meV. The cross-section, determined by the parabola method exhibits a pronounced maximum in the region of \( E \sim 6 \) meV as compared to the incoherent approximation result. The direct measurements of the UCN production cross-section were performed with a broad energy distribution of the incoming cold neutrons (see fig. 4 of [15]). It is obvious that the double-peak structure in our cross-section determined by incoherent approximation, using \( G_1(E) \), cannot be reproduced by the data presented in [15] (cold-neutron energy resolution). A new experiment at a more intense cold-neutron beam with a better energy resolution would be desirable. Furthermore it should be emphasized that the coherent phonon contribution to the UCN production cross-section at \( E \approx 5 \) meV, which is clearly seen in fig. 6, is a major down-scattering channel for UCN production.

In fig. 7 the dynamical scattering function \( S(Q,E) \) (neutron energy loss side) of fast frozen solid ortho-deuterium is shown. The black line corresponds to the dispersion of the free neutron.

The parabola of the free neutron crosses the acoustical phonon dispersion curve at \( E \sim 6 \) meV (see fig. 7). At this point, the UCN production cross-section is predominantly determined by coherent scattering. This can explain a deviation from the production cross-section in incoherent approximation. Nevertheless the general agreement of the incoherent approximation with the PSI data is remarkable (see fig. 5).

The effective UCN production rate \( P \) (UCN cm⁻³ s⁻¹) from a neutron beam is determined by integrating the product of the UCN production cross-section \( d\sigma/dE \) and the spectral flux \( d\Phi/dE \) (Maxwell spectrum with effective neutron temperature \( T_n \)). UCN energy range: 0–150 neV inside the solid D₂. Neutron capture flux \( \Phi_C = 1 \cdot 10^{14} \) cm⁻² s⁻¹. Solid line: total production rate (one- and two-particle excitation). Dashed line: one-particle production rate. Dotted line: two-particle production rate.

The main conclusion from these results is the new understanding of possible higher energetic loss channels (one-quasi-particle and two-quasi-particle excitations) in solid deuterium for the down-scattering of thermal or cold neutrons.

\[
P(T_n) = N_{D_2} \int_0^{E_{U,\text{max}}} \int_0^{E_{U,\text{max}}} \frac{d\Phi(T_n)}{dE_0} \frac{d\sigma}{dE_0} dE_U dE_0.
\]

(4)

In fig. 8 the result for the UCN production rate in solid ortho-deuterium, exposed to Maxwellian shaped neutron flux for different effective neutron temperatures is shown, using eq. (4).

The main conclusion from these results is the new understanding of possible higher energetic loss channels (one-quasi-particle and two-quasi-particle excitations) in solid deuterium for the down-scattering of thermal or cold neutrons.
neutrons in the conversion process to UCN. The best value for the effective neutron temperature is in the region of $T_n \sim 40$ K, which is larger than the value ($T_n \sim 30$ K) reported by Yu et al. [16] in an earlier publication. A similar result was obtained by Serebrov et al. [20] using a Debye model for sD$_2$ in a theoretical calculation, based on the incoherent approximation. The agreement of our result with the results of the calculation applying a Debye model is not a surprise, because Serebrov et al. include multi-phonon excitations in their calculations, while Yu et al. considered only one-phonon excitations. The Debye model and our $G_1(E)$ leads to a similar $T_n$, but the underlying physics for both applied DOS is different. The Debye model assumes only one-quasi-particle excitations up to $E \simeq 10$ meV. Contributions to the UCN production cross-section at energies $E \geq 10$ meV are only caused in this model by multi-quasi-particle excitations. Our DOS $G_1(E)$ (see fig. 5) leads to a more subtle picture. The major contribution to UCN production at energies $10$ meV $\leq E \leq 15$ meV is in this case induced by one-quasi-particle excitation.

**Conclusion.** – In summary, new neutron scattering data of solid deuterium lead to a better understanding of UCN production in this converter material. The new results for the density of states in sD$_2$ and the results for the UCN production cross-section, extracted directly from the dynamical scattering function $S(Q, E)$ predict a significant UCN production cross-section for incoming neutrons with energies higher than $E_0 > 10$ meV. This observation was confirmed [15] by direct measurements of the UCN production cross-section. An optimized sD$_2$ UCN source should be exposed to a cold-neutron flux with an effective neutron temperature of $T_n \simeq 40$ K.

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