

Scattering cross sections of solid ortho-deuterium for ultracold neutrons: Role of defects, phonons, and temperature

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(Received 9 September 2024; revised 1 November 2025; accepted 11 March 2026; published 17 April 2026)

Solid ortho-deuterium is a widely used converter material in ultracold-neutron (UCN) sources. Although it has been studied with neutrons for more than two decades, some of its scattering properties remain poorly understood until this day. We remeasured the total scattering cross section of solid deuterium for ultracold neutrons and—as an improvement over previous experiments—eliminated side effects by employing low-roughness, transparent sample container windows. In this paper, we show the decomposition of the total scattering cross section into its constituents: incoherent elastic scattering, one-phonon up-scattering (using the corrected incoherent approximation), and coherent scattering from crystal defects. Applying both the Guinier approximation and a model using a spherical form factor to describe the defects, we were able to quantify the size and concentration of defects in the deuterium crystal at two different temperatures. Surprisingly, they account for about half of the total scattering cross section. Our work challenges the current picture of ultracold-neutron scattering in solid ortho-deuterium and explains the inability of state-of-the-art simulations to reproduce earlier experimental results. It represents a crucial step toward reliable scattering kernels for the planning of new UCN sources. Ultimately, our measurements allow to deduce the mean free path of UCNs in solid ortho-deuterium, which is 2.5 to 3.5 cm in the relevant temperature range and for a neutron velocity of 10 m/s. The optimal operating temperature for UCN converters is deduced to be between 10 and 12 K, contrary to the 5 K accepted hitherto.

DOI: [10.1103/5rx2-j8cq](https://doi.org/10.1103/5rx2-j8cq)

I. INTRODUCTION

Ultracold neutrons (UCNs) can be used for a wide range of particle physics and other fundamental experiments [1,2], among which are the determination of the free-neutron lifetime [3], the search for a nonzero electrical dipole moment of the neutron [4], UCNs as a low-energy probe for physics beyond the standard model [5], and neutron-optical and interference phenomena [6].

After the inception of the PF2 facility [7,8] at the Institut Laue–Langevin, which mechanically slows very cold neutrons (VCN) down to UCN velocities and provides UCN beams to the scientific user community, most activities of developing a higher intensity UCN converter (often simply called a “source”) have focused on solid ortho-deuterium (sD₂) [9–16] and liquid helium-based converters [17–19]. Note: Throughout this paper, “D” used in the text denotes the hydrogen isotope deuterium ²H.

Given the attention that sD₂ currently receives as a UCN converter material, it seems appropriate to look at the exper-

imental data on which scattering kernels for simulations are based and whether they still represent the state of the art.

In his Ph.D. thesis from 1970 [20,21], Seiffert published the first experimental scattering cross-section data for thermal and cold neutrons (energy range 80 meV to 1.4 meV) in solid ortho-deuterium at $T = 17$ K. In 2005, Atchison *et al.* [22] published total scattering cross sections for even slower neutrons—VCN and UCN.

The data from Atchison *et al.* [22] are the experimental reference data for the total scattering cross sections of solid deuterium today and have been widely cited; see, for example, Bernnat *et al.* [23], Granada *et al.* [24], and Plompen *et al.* [25]. These data are also the only reference for UCN scattering in sD₂ listed in the EXFOR database maintained by the International Atomic Energy Agency (IAEA) [26,27].

The data of Ref. [22] have three shortcomings that motivated us to measure the scattering of UCNs in solid deuterium again: The experiment was done using a sample container with rough-surface windows that bulged under pressure, which (i) gave rise to an error of 5 to 10% in the total cross section, and (ii) caused a non-negligible amount of UCN scattering on the rough surface of the sample container [28]. (iii) The use of the incoherent approximation (IA) [29,30] for calculating the one-phonon up-scattering cross section of UCNs in solid deuterium (“scattering model”) and thus the conclusion that “the deuterium crystals produced at 18 K would seem to be close to ideal,” i.e., free of defects, was unjustified as sD₂ is a predominantly coherent scatterer.

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For the latter reason, the incoherent approximation yields a cross section for sD₂ at 18 K that is a factor of 2 too high [31]. This has already been conjectured by Lavelle *et al.* [17], who measured a one-phonon incoherent up-scattering cross section of sD₂ that was a factor of 2–4 lower than the value predicted by the incoherent approximation. These findings support our preliminary conclusion that the current reference data for scattering in sD₂ need to be carefully reassessed.

II. THEORY BACKGROUND

Since crystal defects cannot be measured directly in a transmission experiment, such as we describe here, we must know all other UCN loss channels and then attribute the remainder to crystal defects. The loss channels are

- (i) Absorption and up-scattering by impurities: The total molecular spin S of the deuterium molecule gives rise to two species, or collections of rotational states, *ortho* and *para* [32]. Ortho-deuterium has the two molecular spin quantum numbers $S = 0, 2$ and even rotational quantum numbers J , where only the $J = 0$ rotational energy level is occupied at $T \leq 19$ K, the temperature below which deuterium is solid. Para-deuterium, on the other hand, has the molecular spin quantum number $S = 1$ and odd rotational quantum numbers J , where only the $J = 1$ rotational energy level is occupied at $T \leq 19$ K. Both species can be converted into one another. The low-temperature equilibrium is close to 99% ortho-deuterium, which is the material of interest for the construction of UCN converters. Para-deuterium acts as a contaminant in UCN converters because it up-scatters UCNs to higher energies by transferring its rotational energy ($J = 1 \rightarrow 0$, 7.4 meV) to the neutron. The cross section for this incoherent and inelastic up-scattering process is denoted as σ_{inc}^{10} . The *para* fraction of our D₂ sample was taken into account in the data treatment as a loss channel, along with neutron absorption by trace hydrogen and deuterium ($\sigma_{\text{abs}}^{\text{H/D}}$).

The variables I and S , which denote spin in this section, are not to be confused with I_d and I_0 for neutron counts and S^{inc} for the incoherent scattering function elsewhere in this publication.

- (ii) Up-scattering by ortho-D₂: As we explained above, the correct calculation of the one-phonon up-scattering cross section $\sigma_{\text{inc}}^{-1\text{ph}}$ of solid ortho-deuterium is of crucial importance when interpreting the measured data. We applied the incoherent approximation with a correction due to interference effects in coherent scatterers. The theory behind this correction was developed by Placzek and Van Hove [33] and is expounded in detail in Döge *et al.* [31]. Only the key equation shall be repeated here:

$$\sigma_{\text{inc}}^{-1\text{ph}} = \underbrace{\sigma^{\text{sc-mol}} S^{\text{inc}}}_{\sigma^{\text{IA}}} \times \underbrace{\left[1 + \frac{\sigma^{\text{coh}}}{\sigma^{\text{sc-mol}}} \left(\frac{\delta S}{S^{\text{inc}}} \right) \right]}_{\text{correction factor } \chi}, \quad (1)$$

where σ^{IA} is the one-phonon up-scattering cross section calculated using the incoherent approximation, S^{inc} is the incoherent scattering function, and $\delta S/S^{\text{inc}}$ are correction coefficients calculated for ortho-deuterium at the relevant temperatures. The expression in the brackets illustrates that this correction is small for largely incoherent scatterers ($\sigma^{\text{coh}}/\sigma^{\text{sc-mol}} \ll 1$) and large for coherent scatterers ($\sigma^{\text{coh}}/\sigma^{\text{sc-mol}} \approx 1$), such as deuterium.

- (iii) Incoherent elastic scattering by the D₂ molecule: The nuclear spin of a deuteron atom is $I = 1$. The nuclear-spin physics of the deuterium molecule D₂ gives rise to an incoherent elastic scattering cross section $\sigma_{\text{inc}}^{00,\text{el}}$, which is 5.13 b per molecule in the UCN energy region, not 4.1 b as often reported [22,34,35]. This stems from the fact that the incoherent cross section of the *ortho* species needs to be averaged over both *molecular* spin states $S = 0$ and $S = 2$ [36,37] with a multiplicity of $2S + 1$:

$$\sigma_{\text{inc}}^{\text{el}} = 4\pi \frac{\langle S(S+1) \rangle}{2I(I+1)} (2b_{\text{inc}}^2), \quad \text{where} \quad (2)$$

$$\frac{\langle S(S+1) \rangle}{2I(I+1)} = \frac{1}{4} \left(\frac{1}{6} \times \langle 0 \rangle + \frac{5}{6} \times \langle 6 \rangle \right) = \frac{5}{4}. \quad (3)$$

Therefore, the incoherent elastic scattering cross section of a bound deuterium *molecule* is $\sigma_{\text{inc}}^{00,\text{el}} = 5/4 \times 2 \times 2.05$ b, where $4\pi b_{\text{inc}}^2 = 2.05$ b is the incoherent cross section of one deuterium *atom*.

One way to estimate the size and concentration of defects in solid deuterium is to apply the two-phase model by Guinier [38,39]. This model assumes homogeneously distributed, sharp-edged, noncorrelated “phase 1 particles” (defects in our case) inside a matrix of “phase 2” (D₂ crystal in our case). All defects have the same size. Applied to a real deuterium crystal this is, of course, a simplification; one that allows for the first estimate of the defect size in solid deuterium. In addition, all defects are assumed to be of spherical shape with a generalized radius of gyration R_g . This radius of gyration is related to the geometric radius of the spheres R_s by $R_g = \sqrt{(3/5)}R_s$. The Guinier model is valid for $k_0 R_g < 1.2$ [39], which is fulfilled for typical UCN velocities and defect sizes of about 100 Å and less.

The double-differential cross section for the Guinier two-phase model is

$$\frac{d\sigma}{d\Omega}(q) = \underbrace{\frac{N_p}{N_s}}_{c_p} \underbrace{\left([2N_s b_s^{\text{coh}} - N_p \underbrace{b_p^{\text{coh}}}_{=0}]^2 V_p^2 \right)}_{(\Delta\rho)^2} \times \exp\left(-\frac{q^2 R_g^2}{3}\right), \quad (4)$$

where N_p is the number density of defects, N_s the number density of the sample *molecules*, i.e., deuterium (the factor of 2 accounts for the two atoms per D₂ molecule), c_p the defect concentration per sample molecule, b_s^{coh} the coherent scattering length of one deuterium *atom*, $b_p^{\text{coh}} = 0$ the coherent scattering length of the empty (defect) phase, $\Delta\rho$ the difference in scattering length density between the sample bulk and

the defects (vacuum), V_p the volume of the spherical defects, and k_0 the wave vector of the incoming UCN.

When this formula is integrated from 0 to $2k_0$, it gives an expression for the removal cross section due to crystal defects, which is valid for $k_0 R_s \lesssim 1.5$,

$$\sigma^{\text{el,Guinier}}(k_0) = 4\pi \times c_p \times [2N_s b_s^{\text{coh}}]^2 V_p^2 \times \frac{1}{x^2} \times [1 - \exp^{-x^2}], \quad (5)$$

where $x^2 = 4/5(k_0 R_s)^2$.

An extension for neutrons of larger k_0 can be added to the model by using the spherical form factor $F_k(q) = 3 \times j_1(qR_s)/qR_s$, instead of the exponential

$$\frac{d\sigma}{d\Omega}(q) = \underbrace{\frac{N_p}{N_s}}_{c_p} ([2N_s b_s^{\text{coh}}]^2 V_p^2) \times \left[\frac{3j_1(qR_s)}{qR_s} \right]^2, \quad (6)$$

which has a Porod-like behavior ($d\sigma/d\Omega \propto 1/q^4$) for $q \rightarrow \infty$.

After integration, this expression yields

$$\sigma^{\text{el,sph-form}}(k_0) = c_p \times [2N_s b_s^{\text{coh}}]^2 V_p^2 \times \frac{9\pi}{2(k_0 R_s)^2} \times [1 - j_0^2(2k_0 R_s) - j_1^2(2k_0 R_s)], \quad (7)$$

where j_0 and j_1 are spherical Bessel functions.

In the limit of very slow neutrons ($k_0 \rightarrow 0$), this reduces to the same expression as Eq. (5).

Neutron scattering on defects can be explained by the full scattering model using the spherical (or other geometric shapes) form factor. This model correctly replicates the low- q behavior and approaches the Porod behavior for large q . The Guinier model is a simple and useful approximation for low q , which is the case for UCNs. In the low- q limit, both models deliver very similar results when fitting the defect cross section σ_{def} , as is shown in the results section. The defect cross section is obtained from the measured total cross section by subtracting all known loss cross sections: $\sigma_{\text{def}} = \sigma_{\text{tot}} - \sigma_{\text{abs}}^{\text{H/D}} - \sigma_{\text{inc}}^{10} - \sigma_{\text{inc}}^{00,\text{el}} - \sigma_{\text{inc}}^{-1\text{ph}}$. The other two important cross sections considered here are defined as follows: $\sigma^{\text{sc-mol}} = \sigma_{\text{inc}}^{00,\text{el}} + \sigma_{\text{coh}}^{00,\text{el}}$, and $\sigma_{\text{scatt}} = \sigma_{\text{inc}}^{00,\text{el}} + \sigma_{\text{inc}}^{-1\text{ph}} + \sigma_{\text{def}}$.

III. EXPERIMENT SETUP

We took the data [40] described here during the same measuring campaign as the data on liquid deuterium published earlier in Döge *et al.* [41]. The setup was identical to the one described in our previous paper. We shall, therefore, restrict ourselves to reporting here only the main parameters and key improvements over previous experiments.

The “Turbine” [7] beamline PF2-EDM of the Institut Laue–Langevin (ILL) in Grenoble, France, was used as the source of UCN. Before the measurements were carried out, the neutron flux ϕ at that beamline was characterized [8].

The experiment was performed using the neutron time-of-flight (TOF) method with a setup similar to that described earlier [42] and a chopper with titanium shutters, which was placed upstream of the solid ortho-deuterium sample, see

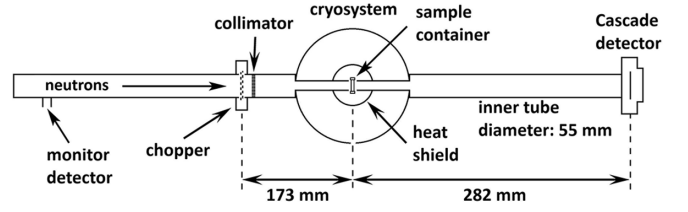


FIG. 1. The TOF geometry of the UCN transmission measurements on solid ortho-deuterium.

Fig. 1. The chopper’s opening function was determined to be $\tau = 13.6$ ms and the total flight path was 455 mm long. A monitor detector enabled the tracking of possible changes of the UCN flux, which were less than 1% over the course of the experiment.

In-depth analysis of the literature [43] and our own trials led us to the conclusion that rough surfaces cause tremendous scattering of UCNs [28] and must be avoided in TOF experiments with UCNs. Furthermore, previous experiments by various groups have shown that aluminum windows tend to bulge under pressure [34] leading to poorly defined sample thicknesses and introducing avoidable errors to the data treatment procedure. To reduce these potential error sources and to improve counting statistics, we developed and used in this experiment a sample container with low-roughness, transparent windows [44].

This sample container made it possible to observe the crystal growth out of the liquid phase. It also revealed a difficulty during sample preparation. When the container was completely filled with a liquid/solid mix, the gas inlet froze over. After the liquid had almost completely solidified to form an optically transparent crystal, the density difference between the liquid and the solid phase caused a bubble with deuterium snow in the middle of the sample. Careful melting and refreezing made it possible to reduce the size of the bubble and convert the snow into deuterium ice [44]. Since the bubble always formed in the same place and had the same dimensions, we were able to blind it out from the UCN beam with a cadmium flap.

While the importance of careful freezing of the sD_2 sample had been pointed out by Seiffert [21], a survey of experiments with sD_2 [45] revealed that, in many cases, experimenters did not report whether their crystals were frozen from the liquid or gas phase. Those that did report their method have not come to an unanimous conclusion as to which freezing method produces the most transparent crystals. Evidence suggests that crystals grown from the liquid phase grow faster and require less complex handling.

We grew our crystals from the liquid phase. Visual inspection showed they were transparent. As UCNs in transmission experiments cannot distinguish between single and polycrystals and are not susceptible to different crystal orientations, we did not determine those parameters of our D_2 crystals. No radiation damage occurred to the crystals as the UCN flux used in the experiment ($\phi < 20$ neutrons/cm²/s) would constitute an ultrahigh vacuum if the neutrons were particles.

The deuterium gas used had a purity grade of N30, meaning at least 99.90% deuterium. The temperature gradient

across the sample container was $\Delta T = \pm 0.25$ K, the same as for our experiment with liquid D₂ [41]. Its ortho-deuterium fraction was measured by Raman spectroscopy, as described by Silvera [32], in the gaseous state during the transfer from the storage vessel to the sample container. We determined an *ortho* fraction of $c_{\text{ortho}} = 97.2 \pm 0.02\%$.

IV. MEASURED TOTAL CROSS SECTIONS AND EXTRACTION OF DEFECT PARAMETERS

A. Data treatment and deconvolution

The recorded experimental data were corrected for background counts and reflection at the sample interface [46], and then normalized and deconvolved. The deconvolution method described below improves upon the data presented in the Ph.D. thesis of Döge [45].

Our previous paper about liquid deuterium [41] served as the blueprint for the deconvolution procedure. We assumed the measured experimental data to be a convolution of the Maxwell–Boltzmann-shaped neutron spectrum [8] $M(v)$ with the chopper opening function $G(v)$. The unperturbed neutron spectrum measured with an empty sample container can thus be described by $\tilde{I}_0(v) = M(v) * G(v)$ and the spectrum measured with the sD₂ sample in the beam is then $\tilde{I}_d(v) = [M(v)T^{\text{expt}}(v)] * G(v)$, with $T^{\text{expt}}(v)$ the measured transmission through the sample and d the sample thickness. We then calculated the transmission from theory, $T^{\text{th}}(v)$, convolved it with the spectra and extracted the convolution factors $C(v)$,

$$\frac{[M(v)T^{\text{th}}(v)] * G(v)}{M(v) * G(v)} \stackrel{\text{def}}{=} C(v) \frac{[M(v)T^{\text{th}}(v)]}{M(v)} = C(v)T^{\text{th}}(v). \quad (8)$$

The deconvolution factors $D(v) = C^{-1}(v)$ were then applied to the experimental data as we did previously [41],

$$D(v) \frac{\tilde{I}_d(v)}{\tilde{I}_0} = T^{\text{deconv}}(v). \quad (9)$$

While this approach is not mathematically rigorous, we validated it qualitatively. The scattering cross sections for low velocities remain virtually unchanged by the deconvolution due to the narrow $G(v)$ while the cross sections for faster UCNs are depressed; the deconvolution does not change the spectra around their respective peaks at about 8 m/s. Applied to the data from liquid deuterium, this approach had an error of less than 5% for the velocity range from 4 to 15 m/s.

The velocity-dependent total cross section $\sigma_{\text{tot}}(v)$ is calculated from the measured and deconvolved transmission, as follows:

$$T^{\text{deconv}}(v) = e^{-N_v \sigma_{\text{tot}}(v)d}, \quad (10)$$

where $T^{\text{deconv}}(v)$ represents the measured and deconvolved absolute transmission through the sample, N_v the particle-number density of the sample, d the sample thickness, and $\sigma_{\text{tot}}(v)$ the *total* UCN loss cross section of the sample bulk for the neutron velocity v in the medium, i.e., taking into account the neutron-optical potential of the sample.

The total cross section is then corrected for the absorption of neutrons [47] in the sample by deuterium

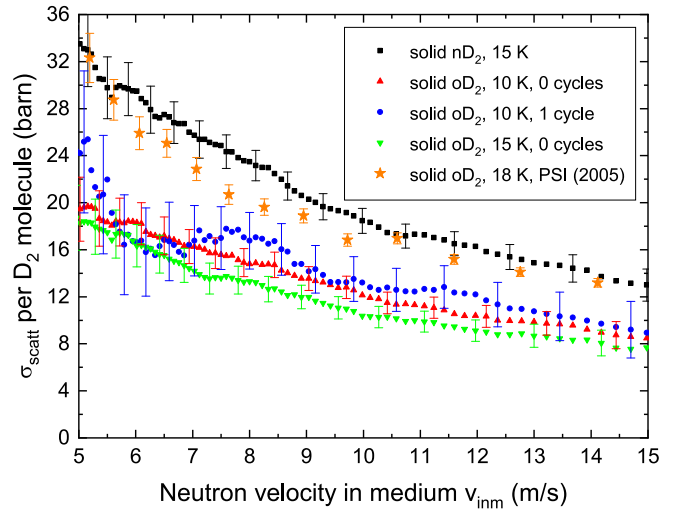


FIG. 2. Experimental data for solid normal- and ortho-deuterium at $T = 10$ K and 15 K with and without temperature cycling. The cross sections of normal-deuterium lie significantly higher than those of ortho-deuterium due to incoherent up-scattering caused by the rotational relaxation $J = 1 \rightarrow 0$ in the former. The cross sections from Atchison *et al.* (PSI 2005) [22] at 18 K were added for comparison. For better legibility, only every fifth error bar is shown.

$[\sigma_{\text{abs,D}} = 2.28 \text{ (b m/s)/v}]$ and hydrogen impurities $[\sigma_{\text{abs,H}} = 1463 \text{ (b m/s)/v}]$ as well as up-scattering from the para-D₂ fraction in the sample $[\sigma_{\text{inc}}^{10} = 271 \text{ (b m/s)/v}]$ [48] to obtain σ_{scatt} of pure ortho-D₂. It should be noted that, in the case of the solid ortho-deuterium used here, the corrections for absorption amounted to less than 1% and those for the para-D₂ fraction for 2...6% over the entire UCN range. Details are described in the Ph.D. thesis of Döge [45].

B. Scattering cross sections of deuterium at 10 and 15 K

The corrected experimental scattering cross sections of solid ortho-deuterium for UCNs are shown in Fig. 2. Normal-deuterium ($c_{\text{ortho}} = 2/3$, $c_{\text{para}} = 1/3$) at 15 K is represented by black squares. Below them, the red up triangles represent solid ortho-deuterium at 10 K and the green down triangles stand for the same crystal at 15.0 K. The blue circles stand for the same crystal after the temperature cycle $10 \text{ K} \rightarrow 15 \text{ K} \rightarrow 10 \text{ K}$. It is easy to see that our temperature cycling did not lower the scattering cross section of a freshly grown crystal. Qualitatively similar findings were published in Refs. [22,49]. In the particular case shown here, the cross section increased slightly as a result of temperature cycling.

The red up triangles from Fig. 2 (solid ortho-deuterium at 10 K) are plotted again in Fig. 3. Calculating the UCN-loss cross sections and applying the Guinier fit model, which are described in Sec. II, the scattering cross section is decomposed into its constituents: one-phonon up-scattering (using the corrected incoherent approximation, described in Döge *et al.* [31]), incoherent elastic scattering from the D₂ molecule, and elastic scattering contributions from defects. The Guinier fit of the defects cross section yields the spherical defect radius $R_d = (114 \pm 2) \text{ Å}$ and the defect concentration

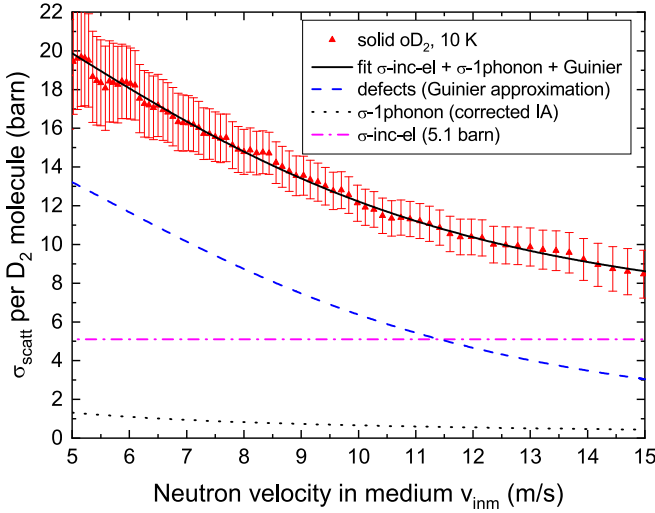


FIG. 3. Experimental scattering cross section of solid ortho- D_2 ($T = 10$ K) and its decomposition into one-phonon up-scattering (corrected IA), incoherent elastic nuclear scattering and the elastic scattering contributions from defects [$R_d = (114 \pm 2)$ Å, $c_p = (2.3 \pm 0.2) \times 10^{-11}$ per D_2 molecule] as calculated using the Guinier approximation.

$c_p = (2.3 \pm 0.2) \times 10^{-11}$ per D_2 molecule. The same fit model yields $R_d = (187 \pm 8)$ Å, $c_p = (1.0 \pm 0.2) \times 10^{-12}$ per D_2 molecule for solid ortho-deuterium at 15 K, see Fig. 4.

This means that a warmer sD_2 crystal has an order of magnitude fewer defects than a colder crystal while the defect sizes are not much different.

An alternative to the Guinier model is a model using the spherical form factor, which is valid not only in the UCN region, but also for higher-energy neutrons. When the data

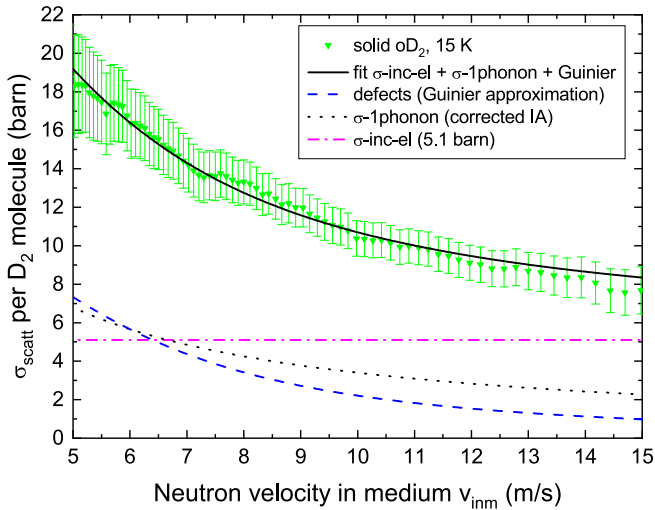


FIG. 4. Experimental scattering cross section of solid ortho- D_2 ($T = 15$ K) and its decomposition into one-phonon up-scattering (corrected IA), incoherent elastic nuclear scattering and the elastic scattering contributions from defects [$R_d = (187 \pm 8)$ Å, $c_p = (1.0 \pm 0.2) \times 10^{-12}$ per D_2 molecule] as calculated using the Guinier approximation.

from Fig. 3 are fitted using Eq. (7), the extracted defect radius is $R_d = (111 \pm 3)$ Å and their concentration is $c_p = (2.9 \pm 0.4) \times 10^{-11}$ per D_2 molecule. They are very similar to the parameters extracted from the same data using the Guinier model.

The quantities c_p and R_d are highly correlated as $\sigma_{\text{def}} \propto c_p R_d^6$, see Eq. (5). This product is the same within the uncertainty interval for the undeconvolved data [45] and the deconvolved data presented here.

V. DISCUSSION

A. Defect scattering and previous results

If instead of the corrected incoherent approximation the “old” IA [31] is used for the fit in Fig. 3, the Guinier model yields $R_d = (126 \pm 5)$ Å and $c_p = (1.2 \pm 0.2) \times 10^{-12}$ per D_2 molecule for 10 K, which means significantly fewer defects. For solid ortho-deuterium at 15 K, the “old” IA leaves no room for defects at all.

The scattering cross sections presented by Atchison *et al.* [22] for 18 K lie 45–60% above those presented here for 15 K. This could be explained by a temperature effect or additional scattering on the rough deuterium sample surface, which very likely formed due to the rough-surface aluminum sample container windows used by these experimenters. If, however, surface scattering was deemed to be negligible and the same fit as in Figs. 3 and 4 (with the corrected IA calculated for 18 K) was applied, the extracted defect size would be $R_d = (97 \pm 10)$ Å and their concentration would be $c_p = (4.4 \pm 2.2) \times 10^{-11}$. This c_p for 18 K is more than one order of magnitude higher than the value reported above for 15 K. As shown here and in Bryś [49] and Atchison *et al.* [22], the magnitude of defect scattering should be *lower* for a *warmer* crystal. It stands therefore to reason that at least part of the UCN scattering reported by Atchison *et al.* is not due to (hypothetical) defects, but uncompensated surface scattering.

B. UCN mean free path in solid ortho-deuterium

Taking the scattering cross sections of solid and liquid deuterium at $v = 10$ m/s from Fig. 2 and Döge *et al.* [41], respectively, and adding the absorption cross section of D_2 and H_2 as well as the rotational relaxation cross section to account for a maximum of 2.8% para- D_2 , $\sigma_{\text{tot}} = \sigma_{\text{scatt}} + \sigma_{\text{abs}}^D + \sigma_{\text{abs}}^H + \sigma_{\text{inc}}^{10}$, it is possible to plot the mean free path λ_{mfp} for UCNs in a real ortho- D_2 crystal over temperature, where $\lambda_{\text{mfp}} = (\sigma_{\text{tot}} \times N_v)^{-1}$ [50]. In Fig. 5 it can be clearly seen that UCNs travel farther in solid deuterium than in its liquid state. For UCNs, it can be inferred from Fig. 3 that elastic defect scattering is the dominant scattering process in the low-temperature solid state. Therefore, the mean free path is essentially dominated by the number and size of crystal defects. The new values of $\lambda_{\text{mfp}} = 2.5 \dots 3.25$ m/s presented here are about a factor of 3 smaller than the experimental value known up to now, 8 cm, from Morris *et al.* [51]. Earlier calculations even estimated the mean free path of UCNs in sD_2 to be about 15 cm at 6 K [52], while UCN scattering on crystal defects was not taken into account. Kahlenberg *et al.* [13] wrote that by using values from Leung *et al.* [53], which were derived for

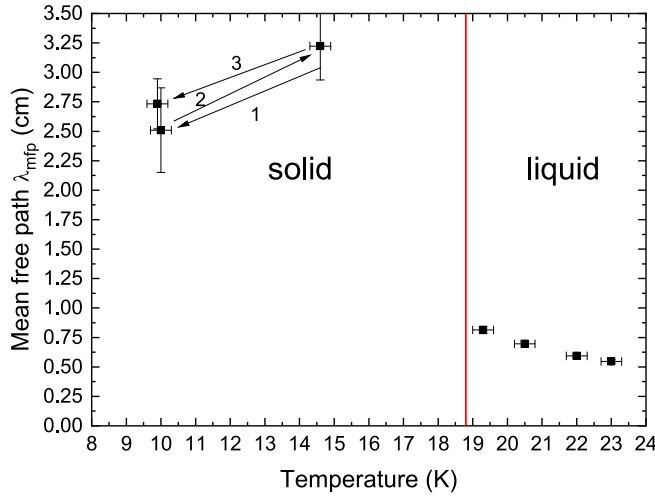


FIG. 5. Mean free path λ_{mfp} of UCNs in condensed ortho- D_2 ($c_{\text{ortho}} = 0.972$) for $v = 10.0$ m/s at 6 different temperatures. The solid deuterium data are the same as in Fig. 2 (here uncorrected for absorption and up-scattering). The arrows 1, 2, 3 represent the temperature cycles of the deuterium crystal.

superfluid helium, they extracted a mean free path of UCNs in their sD_2 source of $4.6 \text{ cm} < \lambda_{\text{mfp}} < 10.0 \text{ cm}$ at $T \approx 5 \text{ K}$ from simulations. Considering Fig. 3, this is likely not the full picture.

These findings reiterate the requirement that UCN converters must be thin-film converters [54,55] instead of bulk converters [10]. In other words, the active conversion zone, from which UCNs can escape the converter crystal, is only a few centimeters thick. Most UCNs produced deeper inside the converter cannot escape and thus not be used in experiments.

C. UCN converter temperature

Comparing Figs. 3 and 4, it is worth noting that the suppression of one-phonon up-scattering at low temperatures is counteracted by more elastic scattering on defects in the deuterium crystal, which leads to the total scattering cross section being almost constant over the temperature range 10–15 K. At temperatures below 10 K, the defect scattering cross section increases further while the up-scattering cross section is less than 10% of the defect scattering cross section at 10 K in magnitude already. At 18 K, the up-scattering cross section alone is higher than up-scattering and defect scattering at 15 K taken together [31]. While elastic scattering is not a UCN loss channel, it increases the dwell time of the UCNs inside the converter medium and increases the chance of UCN loss in the converter.

The results presented in this paper suggest the lowest total cross section to be at a deuterium temperature of 10 to 15 K. This is in line with empirical measurements of UCN yields of converters operating at the moment:

- (i) Frei [56] found that decreasing the temperature of the sD_2 converter below 10–11 K did not increase the UCN output of the source he investigated.

- (ii) Lavelle *et al.* [17] reviewed all yields of UCN sources and their temperature dependence that had been published until 2010. The UCN yield from the source at Los Alamos, which does not have an exit window but releases UCNs into the beam tube directly from the sD_2 crystal layer, showed no temperature dependence for $T \leq 10 \text{ K}$, while all other surveyed sources showed only a negligible temperature dependence of the UCN yield between 5 and 12 K.
- (iii) Atchison *et al.* [57] reported gains in UCN output from sD_2 -based converters of only 9% for cooling the D_2 crystal from 12 K to 8 K.

Only above 12 K does the UCN yield from most converters start to decline significantly with temperature. Earlier calculations using the incoherent approximation with higher one-phonon up-scattering cross sections had estimated the upper saturation temperature to be 4–5 K [54,55,58]. We thus conclude a temperature of 10–12 K to be the optimum operating temperature of UCN converters. This opens up the possibility of saving cooling power without compromising the UCN yield.

VI. CONCLUSION

In this paper, experimental scattering cross sections of solid ortho-deuterium for ultracold neutrons (UCNs) are presented. They were taken using a sample container with highly polished amorphous silica windows, which suppressed parasitic neutron scattering on surfaces as far as possible. Moreover, this container allowed for an optical inspection of the samples before the neutron measurements were started. It was developed specifically for more reliable UCN transmission measurements on cryogenic samples.

For solid ortho-deuterium, it is demonstrated quantitatively how the total scattering cross section can be decomposed into its constituents: one-phonon up-scattering, incoherent elastic scattering, and coherent defect scattering. This decomposition allows for estimating the size and concentration of defects in solid deuterium crystals using the Guinier approximation and the spherical form-factor model known from small-angle neutron scattering (SANS). Both models show excellent agreement with one another in the UCN range.

Applying these models to ortho-deuterium crystals at 10 K, they deliver a spherical defect radius of $R_d = (114 \pm 2) \text{ \AA}$ and a defect concentration of $c_p = (2.3 \pm 0.2) \times 10^{-11}$ per D_2 molecule. For crystals at 15 K, the values are $R_d = (187 \pm 8) \text{ \AA}$ and $c_p = (1.0 \pm 0.2) \times 10^{-12}$, respectively.

Furthermore, we showed that the scattering cross sections reported by Atchison *et al.* [22] contain contributions from either scattering on rough surfaces or scattering from crystal defects, although the authors deemed the crystal to be “close to ideal,” i.e., without any defects. The proportions of either cannot be determined retroactively.

Converting the experimental cross sections from this work into mean free paths for UCNs inside deuterium crystals yields typical values of about 2.5 cm, which is a factor of 3 to 4 smaller than previously reported values [13,51].

The UCN total scattering cross sections of ortho-deuterium have their low at a temperature between 10 and 15 K. A

previous survey of UCN-converter yields at various temperatures supports this result, which led us to conclude that 10–12 K is the optimum operating temperature for UCN converters. Considering that the current operating temperature of UCN sources is 5 K, this opens up the possibility of saving considerable quantities of cooling power over the UCN source's lifetime without compromising the UCN yield. A forthcoming publication [59] will address the consequences of our findings for the design and operation of UCN sources using ortho-deuterium as the converter material.

ACKNOWLEDGMENTS

The authors are grateful to Ekaterina Korobkina and Robert Golub of NCSU in Raleigh, North Carolina, USA, and to Valery Nesvizhevsky of the Institut Laue–Langevin (ILL) for fruitful discussions. Furthermore, we wish to thank the reactor crew, the instrument scientists, and the technicians

of the mechanical workshops at the Institut Laue–Langevin, Grenoble, for their support during the Beamtime No. 3-14-374. We gratefully acknowledge Xavier Tonon and Eddy Lelièvre-Berna of the ILL Cryogenics Service for valuable advice in improving the cryostat, and Brian C. Dye of the University of Louisiana at Lafayette for a critical reading of the manuscript. Bernhard Lauss from the UCN group of the Paul Scherrer Institut (PSI) in Villigen, Switzerland, provided a Raman spectrometer for our experiments and Nicolas Hild (also from the PSI) evaluated the Raman measurements. This work received funding from FRM II/ Heinz Maier-Leibnitz Zentrum (MLZ), Munich, Germany.

DATA AVAILABILITY

The data that support the findings of this article are openly available [40].

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