Experimental report

13/09/2023

Proposal: 3-14-431 **Council:** 10/2022

Title: Neutron Transmission through Clathrate Hydrate Samples for New Very-Cold-Neutron Moderators

Research area: Physics

This proposal is a new proposal

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Samples: Tetrahydrofuran
Water

Instrument	Requested days	Allocated days	From	To
PF1B	10	10	12/06/2023	22/06/2023
PF2 VCN	20	20	25/05/2023	12/06/2023

Abstract:

The interest for Very Cold Neutrons (VCN) is increasing within the last years in multiple fields of research, as they do not only provide an innovative probe for condensed matter physics, but can also contribute in pushing the limits of fundamental physics experiments employing beams of slow neutrons. This goes along with an increasing interest in new sources of VCN.
Clathrate hydrates seem to be particularly well suited for a moderation medium, as they possess low-energy modes with sufficiently large inelastic neutron scattering cross sections. This is provided by their composition ¿ a network of water molecules, that forms cages of different sizes, which are able to host guest molecules.
Besides its capability of moderation an important parameter for a VCN source
is the mean free path of the produced VCN. This quantity is directly accessibly with neutron transmission experiments. Measurements in the cold neutron range, using time-of-flight analysis of the transmitted direct beam of PF1B, should show the Bragg edges of the polycrystalline structure of our samples. Measurements in the VCN range should complement data previously taken.

Experimental Report 3-14-431: Neutron total cross-section measurements of cold reflector and moderator materials

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1 Introduction

Clathrate hydrates have been identified as a very promising candidate for a moderation material for very cold neutrons (VCN), as they possess low-energy modes that allow pre-moderated neutrons to transfer small quantities of energy to the medium at sufficiently large scattering cross sections. To some extent, the low-energy incoherent excitations of methane and tetrahydrofuran (THF, chemical formula: C_4H_8O) clathrates have already been studied [1]. To develop a prototype of a VCN source using clathrate hydrates as a moderator material not only requires data of $S(q,\omega)$ in absolute units (as measured in different experiments conducted by some of the authors), but also measurements of the mean free path of the VCN produced within the moderator. This quantity is directly accessibly with neutron transmission experiments. If Z^0 is the count rate at a detector with no sample, then the count rate after insertion of a sample of the thickness d is:

$$Z^+ = Z^0 \cdot \exp\left(-N\sigma_t d\right) . \tag{1}$$

The transmission of the sample can be defined as $T = \frac{Z^+}{Z^0}$, which allows the total cross section σ_t to be expressed as:

$$\sigma_t = \frac{1}{N_v d} ln(\frac{1}{T}) = \frac{1}{N_v d} ln(\frac{Z^+}{Z^0}) . \tag{2}$$

The mean free path ℓ is simply given by:

$$\ell = (\sigma_t N)^{-1} . \tag{3}$$

A time of flight setup allows to measure these quantities as a function of the neutron's wavelength λ.

1.1 Samples

The samples with stoichiometric composition $17D_2O \cdot C_4D_8O$ were prepared by mixing the two liquid components. Samples of the mixture were then filled into their respective sample containers in which they were frozen in situ. The containers define the slab geometry of the sample with dimensions of 1.5 cm x 1 cm, which limits multiple scattering, while still exposing sufficient material to the beam.

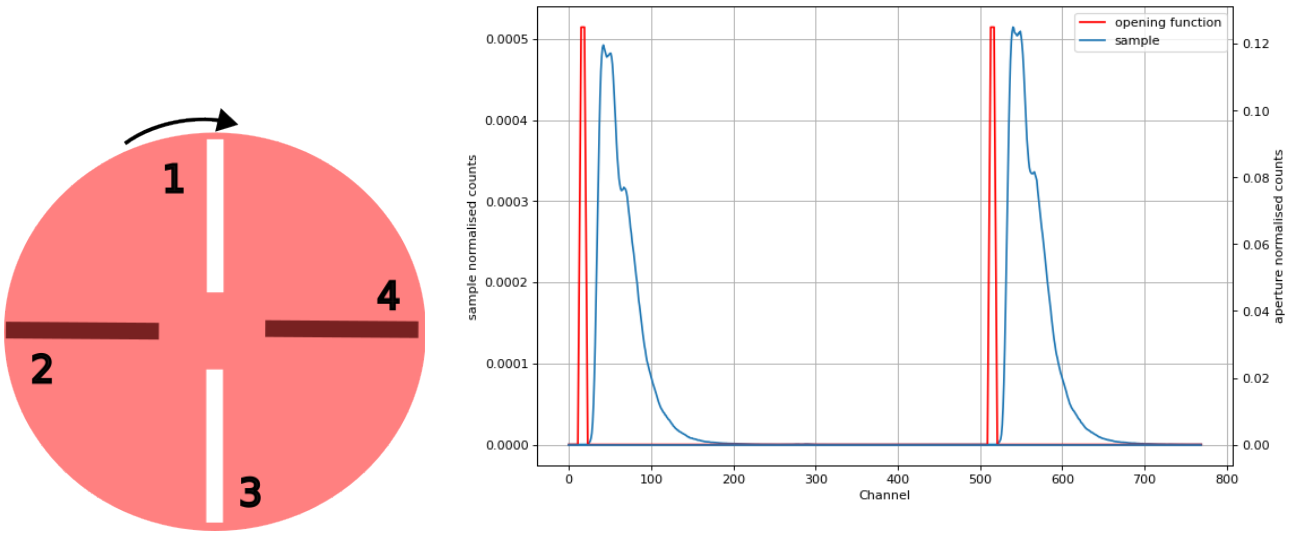
2 Experiment at PF1B

2.1 Data Analysis

A diagram of the chopper used at PF1B is shown in figure 1a. This is a double slit chopper, with a single pickup located before opening 1. The opening time of each slit is non-negligible when compared to the experimental timescale. The red line in figure 1b shows a representative aperture function. Start points of the opening were determined by tuning the detector to detect gamma photons, with the opening width and shape determined from geometric considerations [2]. The measured spectra, blue in figure 1b, is then a convolution of the real spectra and the opening function.
This can be represented as

$$y(t) = (h * x)(t) + n(t) \tag{4}$$

where $y(t)$ is the measured signal, $h(t)$ the chopper opening function, $x(t)$ the real spectra and $n(t)$ the associated noise. To obtain the real spectra a technique called Wiener deconvolution can be used. To obtain the true spectra



(a) Diagram of the chopper.

(b) Raw TOF spectra through sample with chopper opening.

Figure 1: (a) Diagram of the chopper used at PF1B. Openings 1 and 3 are fully open, allowing neutrons to pass freely. Openings 2 and 4 are covered with a neutron absorbing material. The pickup for the detector is located before opening 1. (b) Blue shows the raw TOF spectra of deuterated tetrahydrofuran clathrate hydrate in an aluminium container, y scale is the counts per bin divided by the total number of counts. Red represents the chopper opening function, amplitude scaled so the area under each opening is equal to 1.

the frequency domain, $X(f)$, Wiener deconvolution has the form

$$X(f) = \frac{H^*(f)Y(f)}{|H(f)|^2 Y(f) + \frac{1}{SNR(f)}} = \frac{H^*(f) \cdot Y(f)}{\mid H(f) \mid^2 + c}\,, \tag{5}$$

where $Y(f)$ is the Fourier transform of the measured spectra, $H(f)$ is the Fourier transform of the chopper opening function, $SNR(f)$ is the signal to noise ratio given by the ratio of the mean power spectral density of the original signal and the mean power spectral density of the noise in the frequency domain, and c is the Wiener coefficient [3]. However this assumes signal independent Gaussian noise [4], whilst the measurements are affected by signal dependent Poisson noise. To circumvent this, the signal can first undergo an Anscombe transformation so that the noise is represented by unity variance [5]. Wiener deconvolution can be applied in this domain, and finally the deconvolved data undergoes an inverse Anscombe transformation to obtain the real spectra.

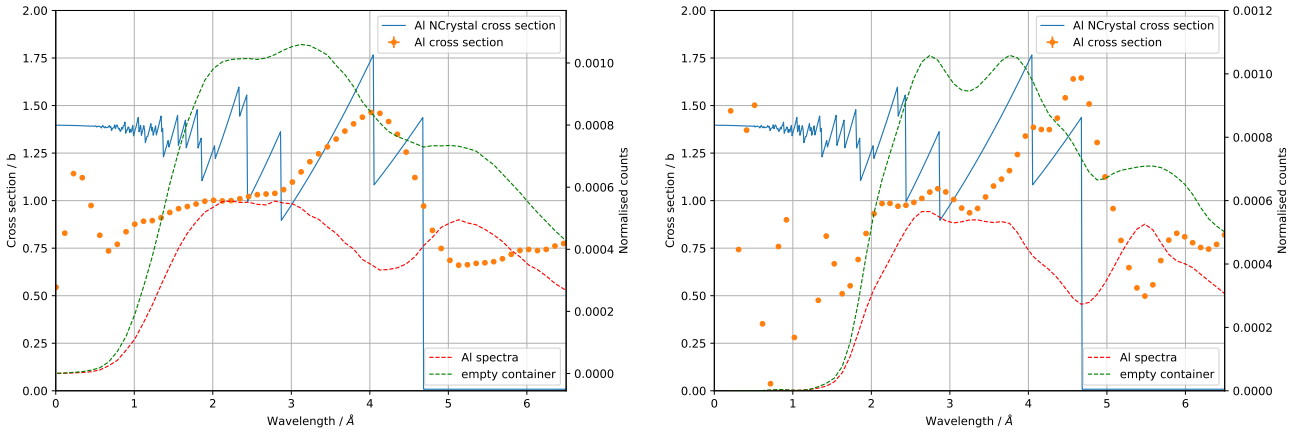
2.2 Preliminary Results

2.2.1 Aluminium

The process was first performed on an aluminium sample to verify the validity of the method using a well documented sample. Comparing the total cross section from the raw data in figure 2a to the data processed with Anscombe transformations and Wiener deconvolution in 2b, it is seen that the main peak at around 4 Å in the raw data is split into two after deconvolution, an additional peak at 2.9 Å introduced and the overall spectra more featured. The peak positions appear to follow the NCrystal simulated data more closely. To determine whether the introduced features are features of the true spectra or artifacts from deconvolution, a linear least squares regression was performed. This used the three peaks mentioned, and a point at 0 Å, determined from the arrival of photons. The corresponding wavelengths were determined from the NCrystal simulated data. This returned a Pearson correlation coefficient of 0.99998, strongly suggesting the introduced features are from the true spectra.

2.2.2 Deuterated Tetrahydrofuran Clathrate Hydrate

Wiener deconvolution was used on data from fully deuterated tetrahydrofuran clathrate hydrate, as shown in figure 3. After deconvolution the spectra is significantly more featured, with prominent peaks at 4.60 Å, 5.96 Å, 7.81 Å and 9.73 Å that before processing were difficult to resolve. The resolution of the Bragg edge at 20 Åis limited by statistics.



(a) Total cross section of Al (raw).

(b) Total cross section of Al (processed).

Figure 2: Aluminium total calculated from raw data (a) and devoncolved (b) data (orange) as well as computed from NCrystal (blue). The corresponding spectra of the Al sample and the empty beam are shown in red and green, normalized by the monitor counts.

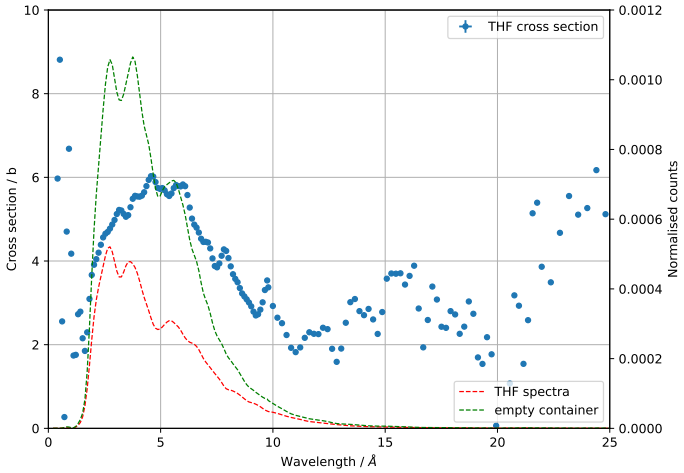


Figure 3: Total cross section and TOF from fully deuterated tetrahydrofuran clathrate hydrate at 4K. Blue is the cross section calculated from deconvoluted data. Red and green are the deconvoluted spectra of the sample and empty container respectively with counts per bin divided by the total number of monitor counts.

Data analysis is ongoing. The report will be updated accordingly.

References

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