WP3: Material characterization with neutrons

Valentin Czamler

HighNESS General Meeting

Lund, June 21, 2022

Outline

1. Why Very Cold Neutrons (VCN)?
2. Moderation to the VCN range
3. Clathrate hydrates as a VCN moderator
4. Results: Time-of-Flight spectroscopy
5. Results: VCN-Transmission
6. Outlook

A Case for Very Cold Neutrons

Neutron scattering:

- From an instrumentation point of view:

	resolution at fixed geometry	intensity at fixed resolution
SANS	λ^{-1}	λ^{0}
Reflectometry	λ^{-1}	λ^{-2}
TOF-INS	λ^{-3}	λ^{2}
NSE	λ^{-3}	$\lambda^{2} - \lambda^{4}$

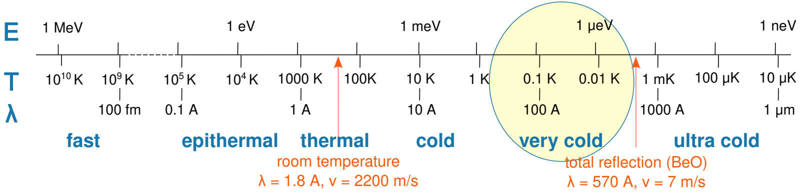
from: proceedings of workshop on application of a VCN source at Argonne, 2005

Particle Physics (counting statistics):

- Neutron-antineutron oscillation experiment
- Neutron beam EDM experiment
- In-beam UCN sources

How to slow down neutrons?

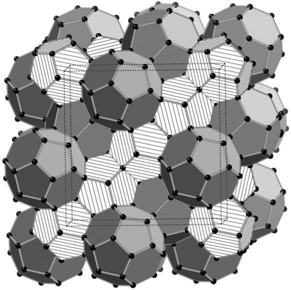
- Many inelastic interactions: → thermal equilibrium with cold medium
- Requirement: weakly absorbing cold medium with suitable scattering cross sections



- From cold (CN) to very cold (VCN): ⇒ incoherent scattering by local modes (neutrons deposit their energy):

1. Molecular rotations
2. Librational modes of confined molecules
3. Weakly absorbing paramagnetic species (O_2)

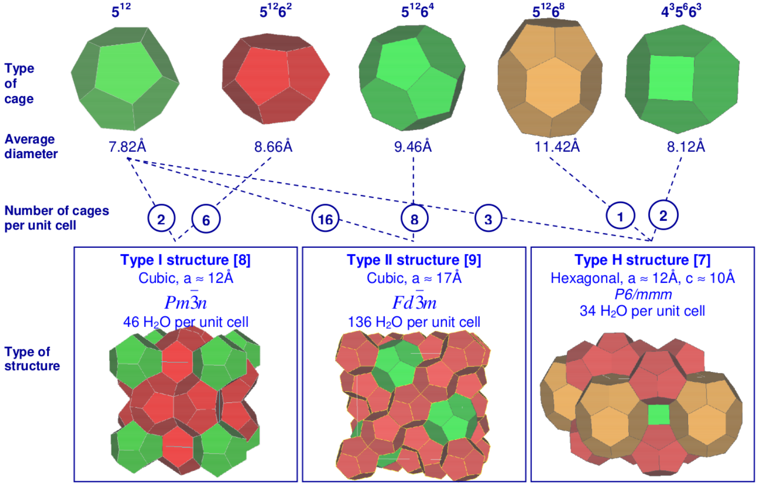
Clathrate Hydrates I



J. Am. Chem. Soc. 2013, **135**, 5, 1696-1699

- Network of water molecules that host small guest molecules
- Introducing THF (C_4H_8O) and O_2 as guest molecules
- Large unit cells $\Rightarrow$ large albedo for the cold neutrons
- Fully deuterated: weakly absorbing
- Consits only of non-dangerous materials

Clathrate Hydrates II



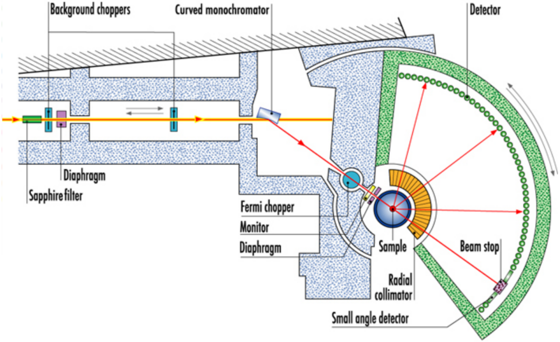
Adapted from: A. Desmedt https://doi.org/10.1051/sfn/2010013

WP3 Tasks

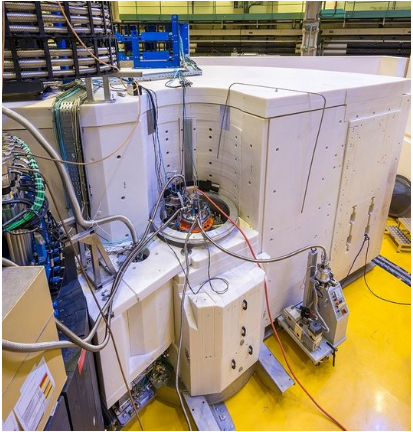
- **Task 3.1**: Preparation of various experimental tools
- **Task 3.2**: Analysis of data obtained for O2-hydrate clathrate
- **Task 3.3**: Measurements of neutron transmission and diffuse reflectivity on substances for WP4 and WP6 within allocated beam time after submission of proposals for the public ILL instruments PF1B, PF2/VCN and D17 or SuperADAM
- **Task 3.4**: Measurements of $S(q,\omega)$ and neutron diffraction for characterization of samples of clathrate hydrates at ILL instruments IN5, Panther, D20, D7, PF1B and PF2/VCN
- **Task 3.5**: Publication of results (utilizing tools developed in WP2)

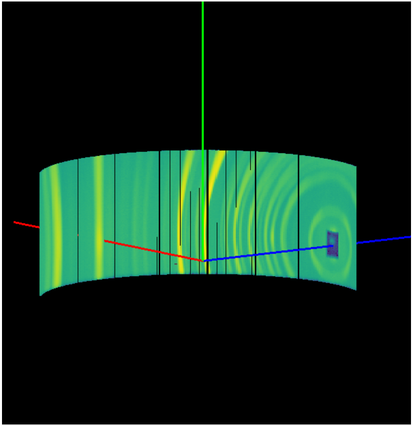
Time-of-flight (ToF) spectroscopy

- Incident wavelength: 1 Å, 2 Å (Panther); 2 Å, 3 Å (IN5)
- Sample temperature: 1.5 K
- Protonated and deuterated components for contrast variation
- Vanadium sample for normalisation



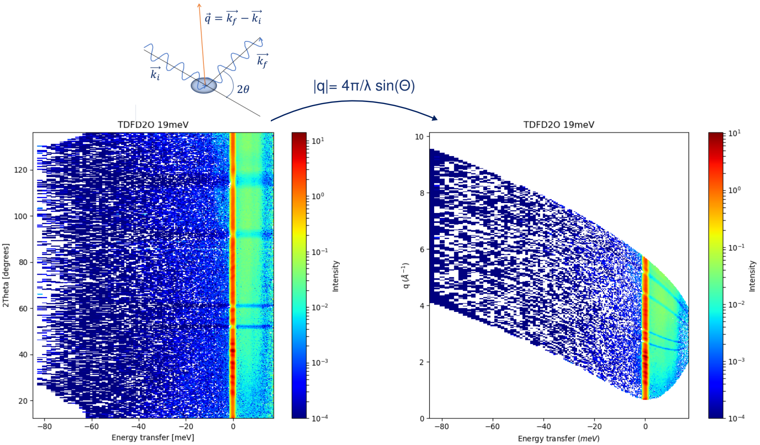
ILL — Panther





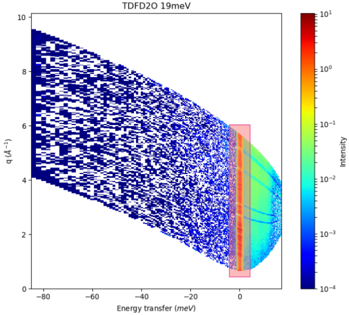
Results I

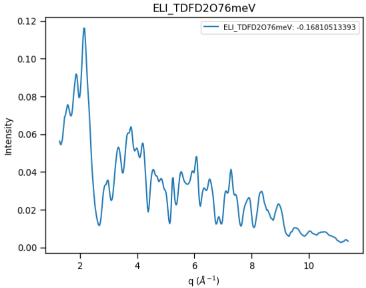
$S(q,\omega)$ for a fully deuterated $17D_2O$: TDF, $E_i = 19$ meV ~ 2 Å



Results II

$S(q,\omega)$ for a fully deuterated $17D_2O$: TDF, $E_i = 19$ meV ~ 2 Å

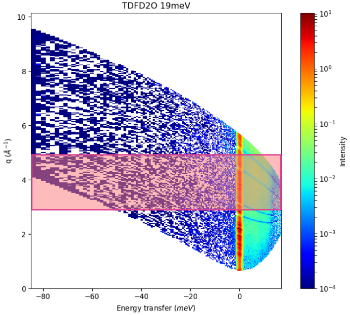


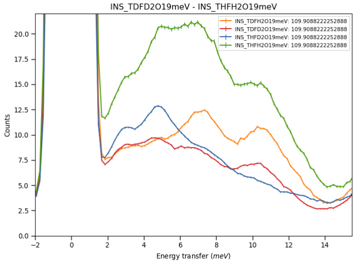


Constant energy-slice at the elastic peak

Results III

$S(q,\omega)$ for a fully deuterated $17D_2O$: TDF, $E_i = 19$ meV ~ 2 Å





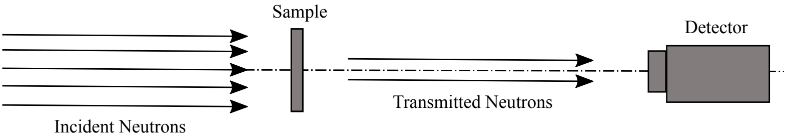
Constant q-slice

VCN - Transmission

- The transparency of the moderator for VCN is an important measure
- Transmission measurements at VCN beam

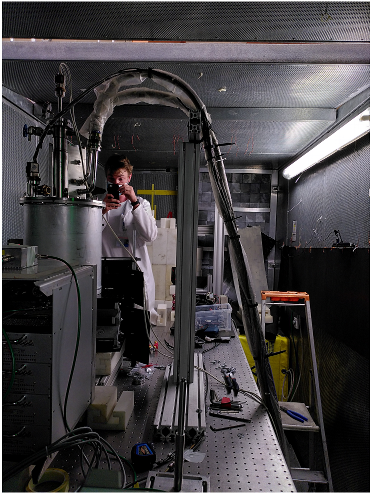
$$T = \frac{Z_{sample}}{Z_{empty}} = \exp(-\Sigma_{tot} d) = \exp(-\sigma_{tot} N_v d)$$

$$\sigma_{tot} = \frac{1}{N_v d} ln(\frac{Z_{sample}}{Z_{empty}})$$

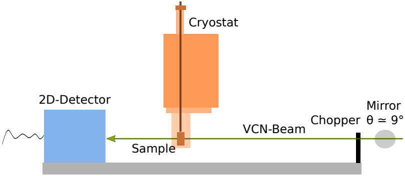


Outline of a Transmission experiment. Adapted from: Beckurts & Wirtz (1964)

Transmission Experiment at PF2 – VCN

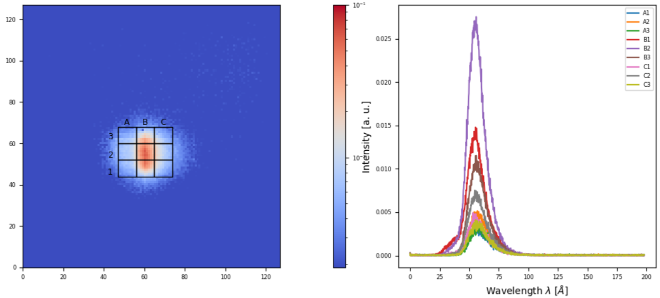


- Time-of-Flight (ToF) set-up
- Sample temperature: 5 K
- Fully deuterated sample:
  $17D_2O : THF - d$
- Sample length in beam: 4 cm



Layout of the VCN-Transmission Measurement.

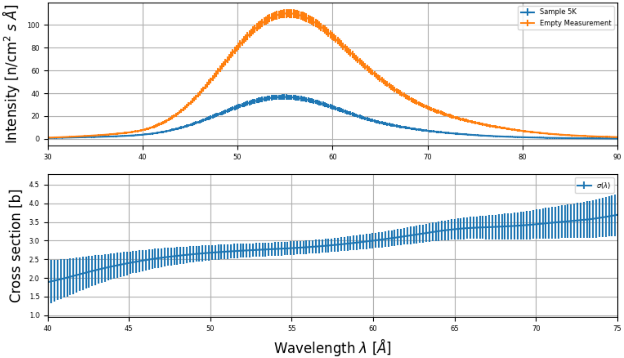
Results (preliminary) I



Transmission through sample 2D (left), ToF spectrum for each segment (right).

Results (preliminary) II

Total cross section computed from an empty measurement (orange) and a transmission through the sample (blue):



Looking Ahead

- Comprehensive analysis of the presented data
- Measurement of the **Bragg-cutoff** (ILL-PF1B, PSI-BOA?)
- Providing the data to the collaborators of WP2
- Manufacturing procedure of binary clathrates:
  $17D_2O : THF - d - 2O_2$
- Requirement of O_2 under high pressure
- + determination of cage filling
- Comparison O_2-filling with already taken data (D20)

Acknowledgements

Supervisors:

- Oliver Zimmer
- Richard Wagner

Collaborators:

- HighNESS Collaboration
- Arnaud Desmedt (ism Bordeaux)

Instrument Scientists:

- Thomas Hansen (D20)
- Tobias Jenke (PF2)
- Michael Koza (Panther)
- Jaques Olivier (IN5)
- Stephanie Roccia (PF2)

Université Grenoble Alpes

EUROPEAN SPALLATION SOURCE

HighNess

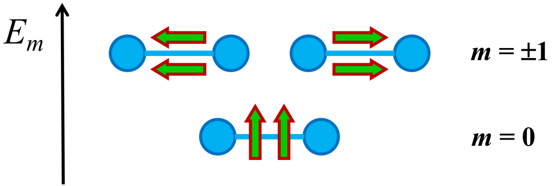
Questions?

INSTITUT LAUE LANGEVIN

NEUTRONS FOR SOCIETY

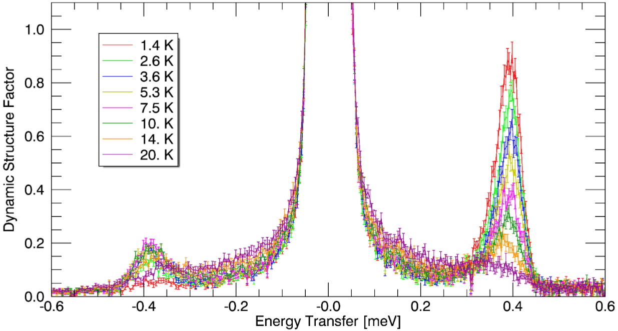
The oxygen's contribution

- O_2 is paramagnetic and has a triplet zero-field splitting ~ 0.4 meV
- allows the neutron to deposit energy
- no dispersion relation $\rightarrow$ allows "cascade cooling"



Zero field splitting of oxygen for cascade cooling. See: Zimmer (20016), DOI: 10.1103/PhysRevC.93.035503

Experiments on O_2-Hydrates

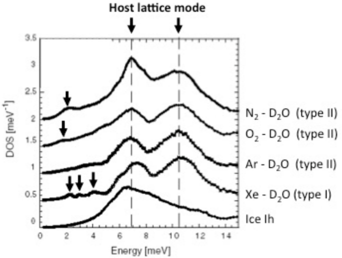


exp. Team: A. Falenty, T. Hansen, M. Koza, W. Kuhs, O. Zimmer

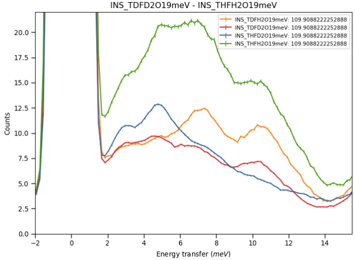
→ **Goal**: Measure the magnetic contribution in the binary clathrate.

Comparison with former Measurements

- Location of the excitation around 7 and 10 meV arise in most type II clathrate hydrates
- not dependend on the guest molecule
  $\rightarrow$ host lattice modes



Schober et. al. (2001), DOI: 10.1039/b202464k



Panther $E_i = 19$ meV