Work Package 3:
Material Characterization with Neutrons

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HighNESS General Meeting 2023

Outline

I. Why Very Cold Neutrons (VCN) ?

II. Moderation to the VCN range

III. Clathrate Hydrates as a VCN moderator

IV. WP3 Tasks: Current Status

V. Manufacturing of THF Hydrates & Structure Analysis

VI. Determination of the neutron scattering function S(**q**, ω)

VII. Transmission Experiments

VIII. Outlook

I. A Case for Very Cold Neutrons

Condensed Matter Research

- Gains in spatial & and energy resolution
- Small angle scattering
- ToF Spectroscopy
- Neutron Reflectometry

Particle Physics

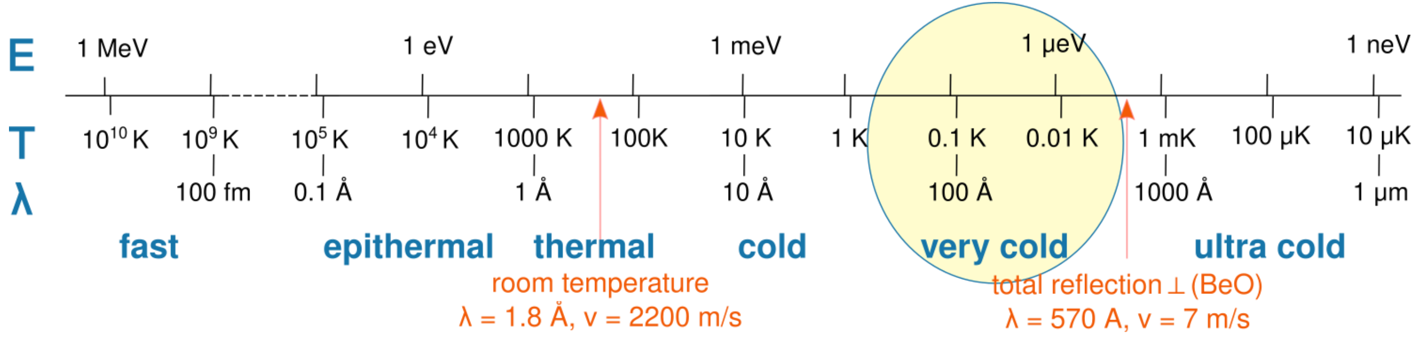
- Increased FOM & counting statistics
- nnbar experiments, neutron charge ($\propto \lambda^2$)
- Neutron beam EDM experiment ($\propto \lambda$)
- In-beam UCN sources

Further Reading:

- Particle Physics at the ESS (arXiv:2211.10396)
- Workshop on very cold and ultra cold neutrons
  https://content.iospress.com/journals/journal-of-neutron-research/24/2

II. How to slow down neutrons?

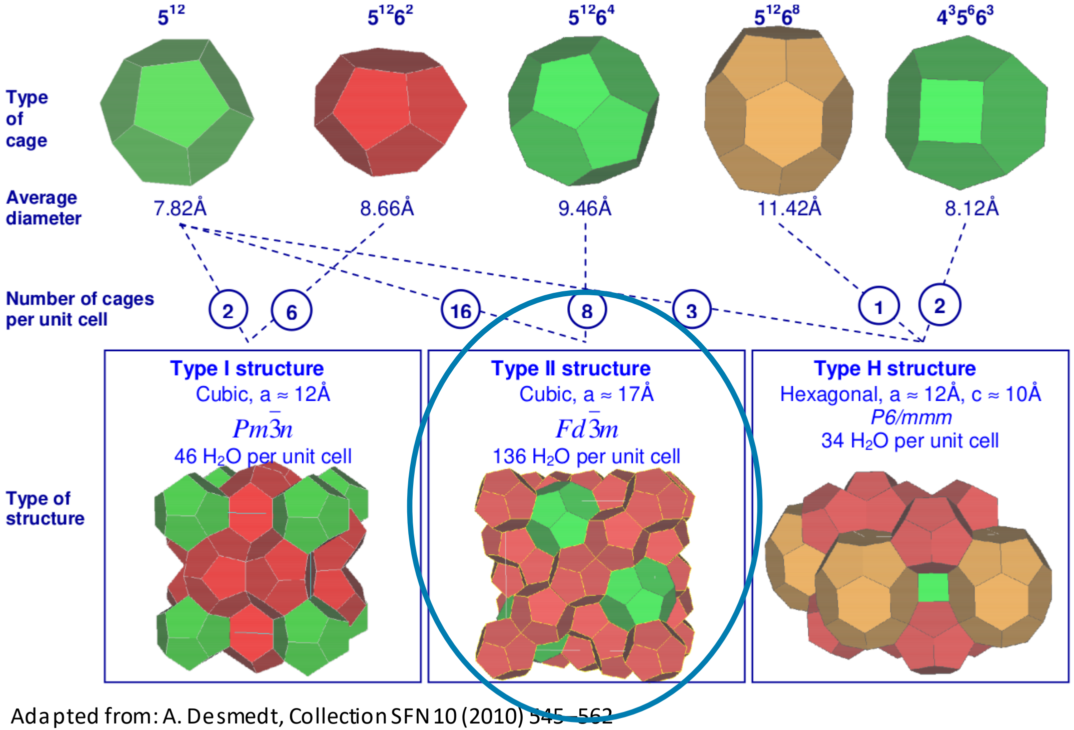
- Many inelastic interactions ⇒ thermal equilibrium with cold medium
- Requirements: weakly absorbing, cold, suitable σ_{scat}



- From CN to VCN: incoherent scattering by local modes:

- Rattling modes
- Rotation
- Libration
- Paramagnetic Species

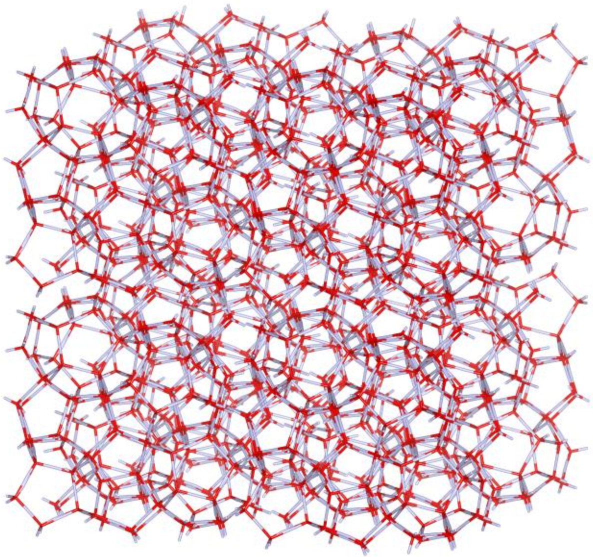
III. Clathrate Hydrates

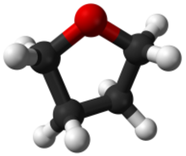
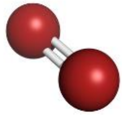


Adapted from: A. Desmedt, Collection SFN 10 (2010) 545–562

- Inclusion compounds: **Network** of hydrogen-bond water molecules that host small **guest molecules**
- Stable up to relatively high temperatures
- High scientific interest (energy storage, seabed sediments, climate tipping points)

III. Clathrate Hydrates



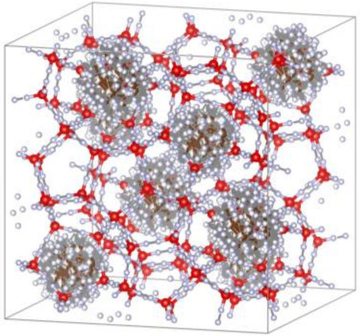
- Tetrahydrofuran (THF, C_4H_8O) & O_2,  & 
- Guest molecules allow for dispersion-free low energy excitations
- Weakly absorbing when fully deuterated
- Unusually-large crystallographic unit cell constitutes a large albedo over the cold neutron range
- Favorable manufacturing conditions

IV. WP3 Tasks

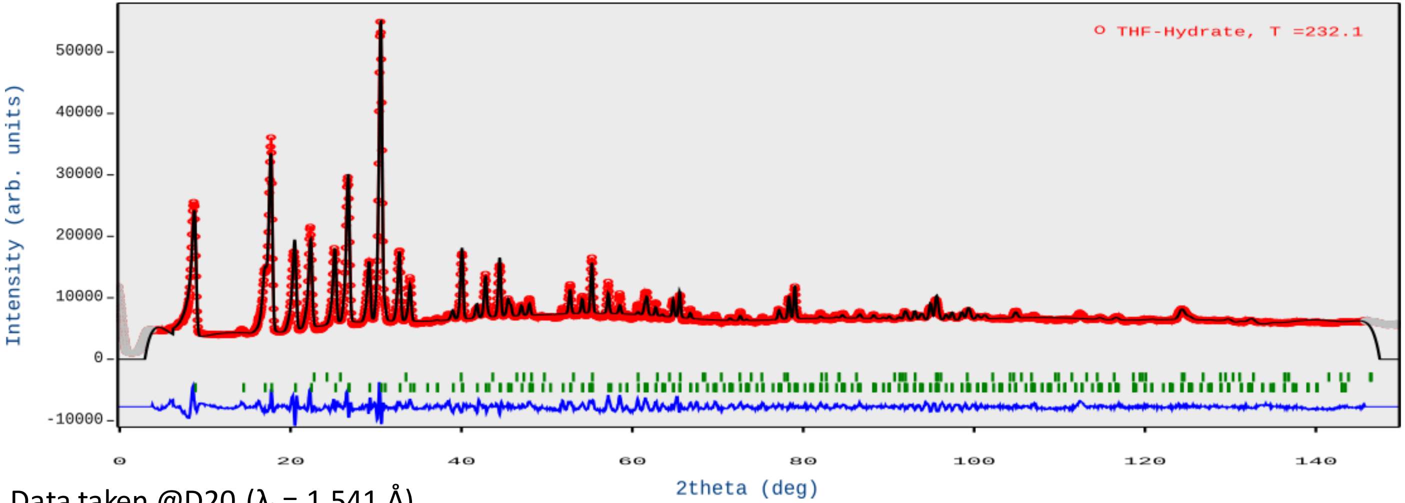
- Task 3.1: Preparation of various experimental tools ☒
- Task 3.2: Analysis of data obtained for O2-hydrate clathrate ☒ → Task 3.5 ?
- Task 3.3: Measurements of neutron transmission and diffuse reflectivity on substances for WP4 and WP6 within allocated beam time after submission of proposals for the public ILL instruments PF1B, PF2/VCN and D17 or SuperADAM ☐→Experiments @BOA; Allocated beam time in May @PF1B
- Task 3.4: Measurements of S(q, ω) 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) ☐

  → Ongoing work in collaboration with WP2

V. Manufacturing of THF-Hydrates



- Easy manufacturing technique from stoichiometric mixture
- Contrast variation: $17 \cdot H_2O:C_4H_8O$, $17 \cdot D_2O:C_4H_8O$, $17 \cdot H_2O:C_4D_8O$, $17 \cdot D_2O:C_4D_8O$

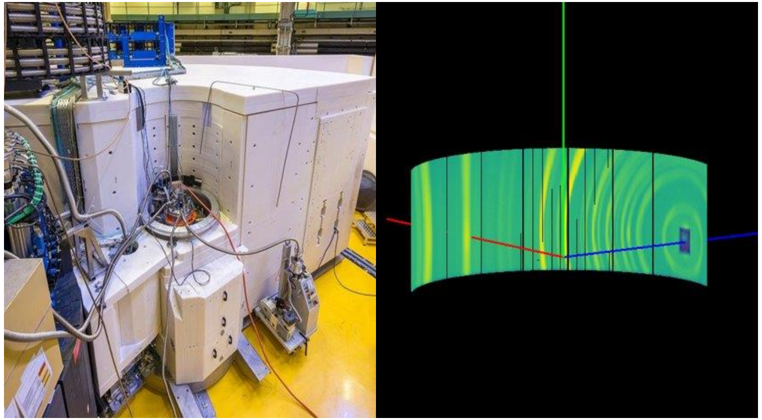


Data taken @D20 (λ_i = 1.541 Å)

VI. Determination of S(**q**, ω) in Absolute Units

- Ultimate goal: A novel moderator with strong enhancement of VCN fluxes ⇒ HighNESS
- Intermediate step: Inelastic neutron scattering study

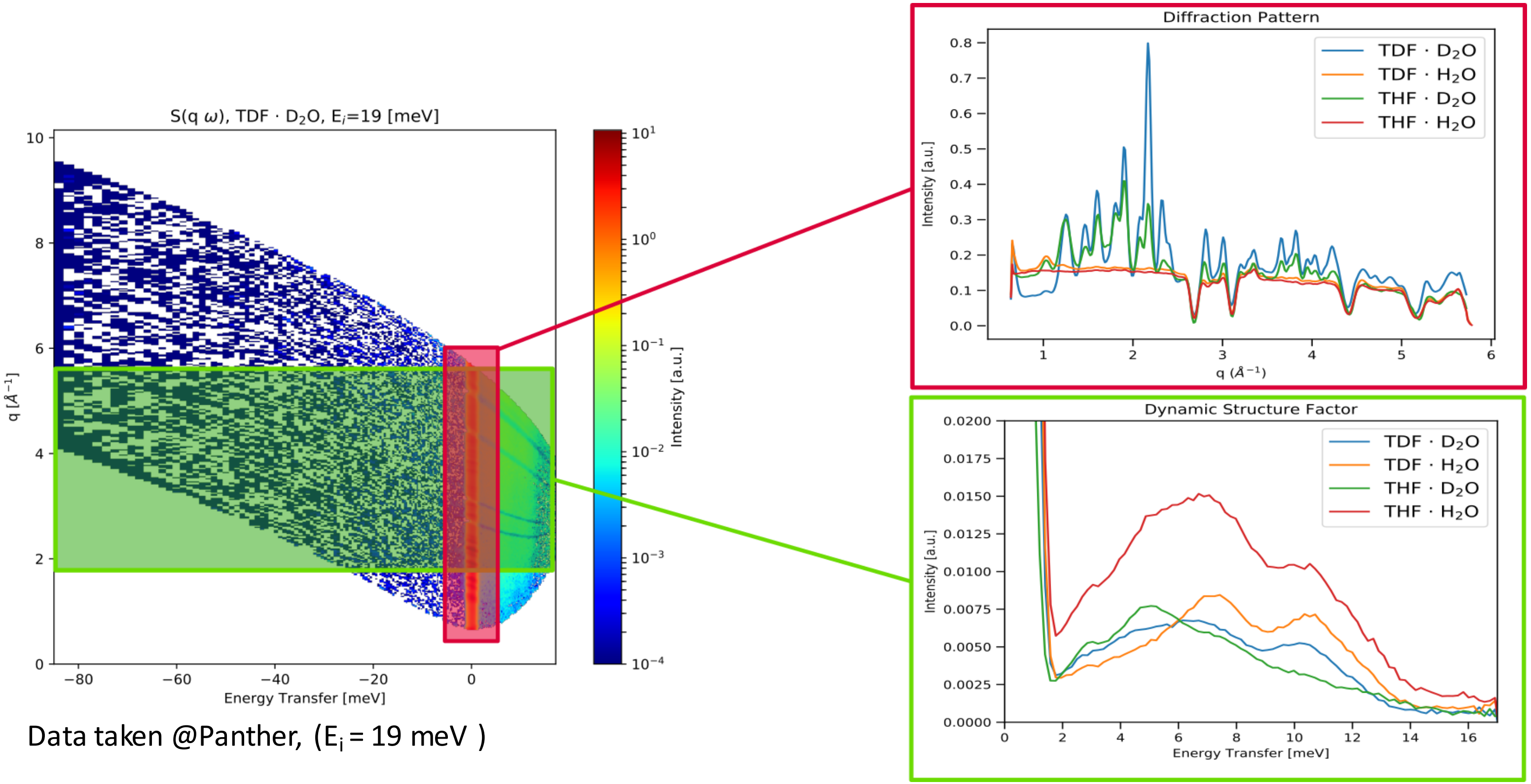
Spectroscopy at ILL' s **Panther** (λ_i = 1 Å, 2 Å) & **IN5** (λ_i = 2 Å, 3 Å)



- Vanadium standard (calibration to absolute units)
- Results serve as a benchmark for new scattering kernels in NCrystal

→WP2 Talk

VI. Preliminary Results I

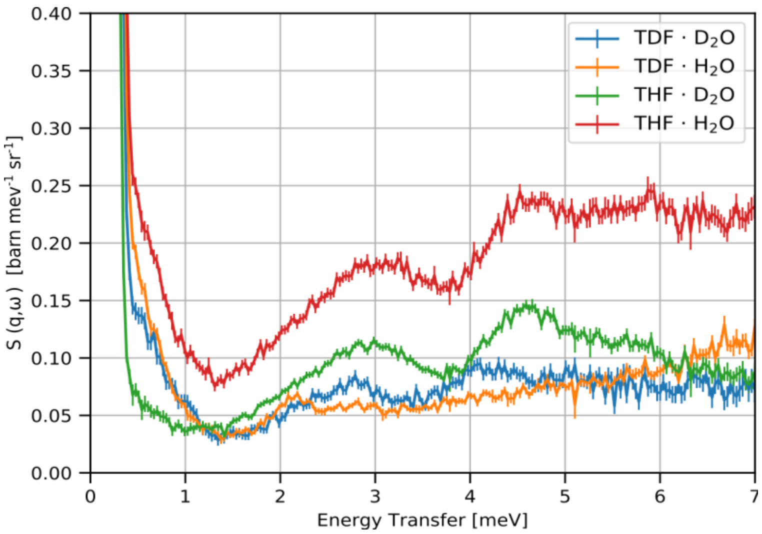


Data taken @Panther, (E_i = 19 meV)

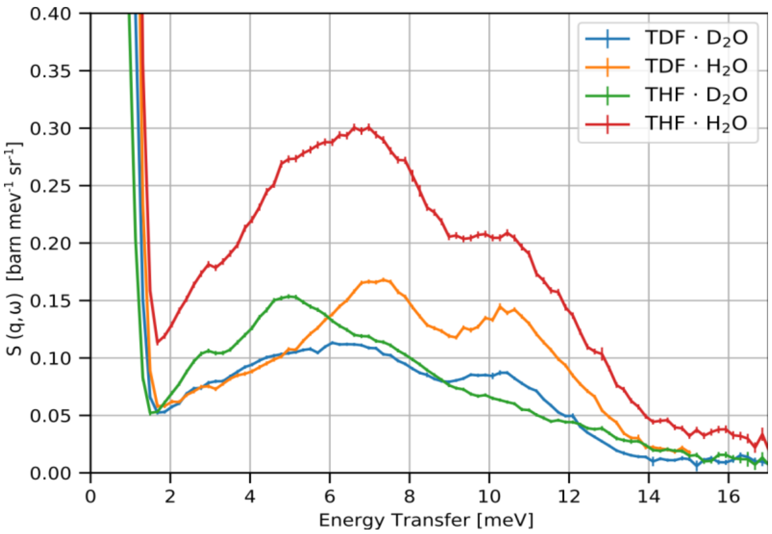
VI. Preliminary Results II

- Contrast variation allows to differentiate between host & guest contribution
- Host lattice modes are in accordance with reported data

(e.g. B. Chazallon et al. ***Phys. Chem. Chem. Phys.***, 2002,**4**, 4809-4816)



E_i = 9 meV, T = 1.5 K, **IN5**

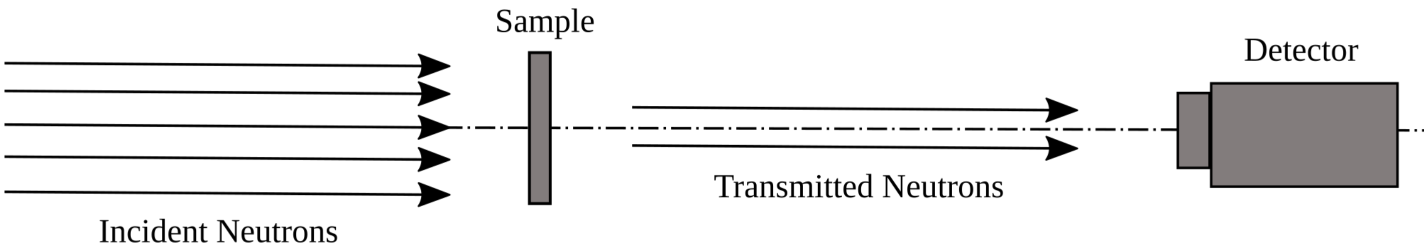


E_i = 19 meV, T = 1.5 K, **Panther**

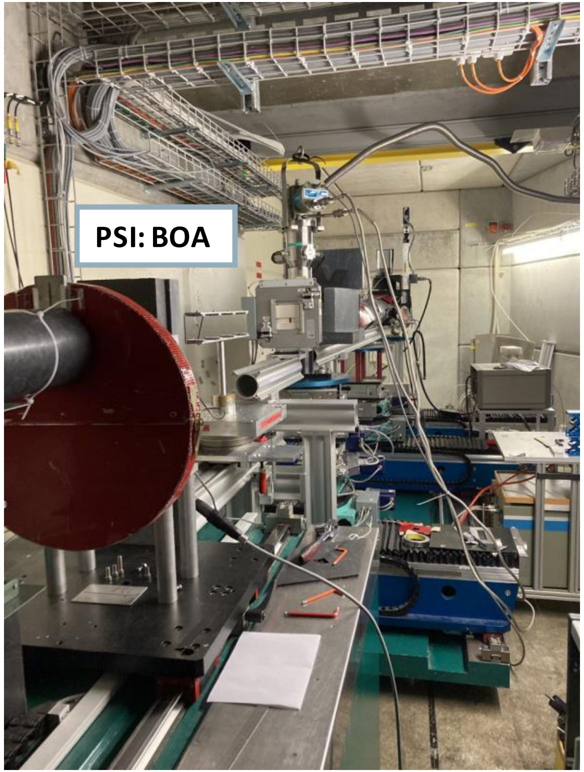
VI. Transmission Experiments

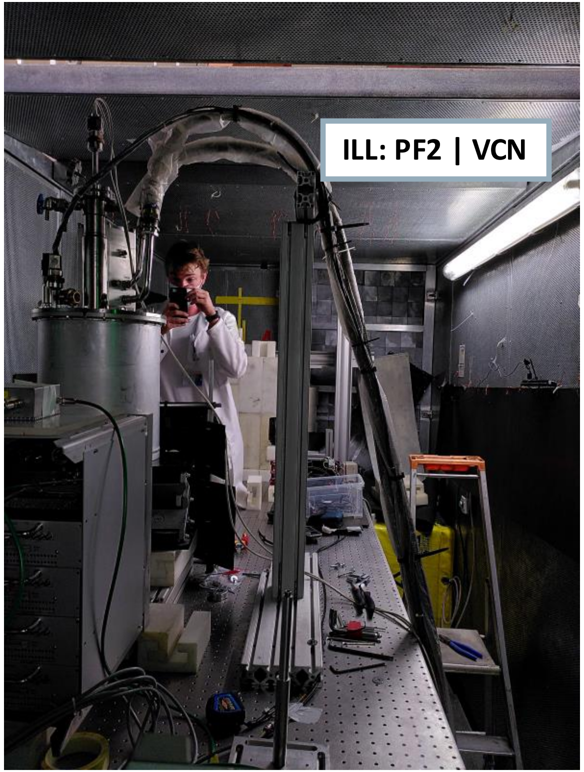
- Determination of the total cross section throughout the CN & VCN range
- Transmission experiments at **ILL** (PF1B, PF2|VCN) & **PSI** (BOA)

$$T = \frac{Z_{sample}}{Z_{empty}} = \exp\left(N_V\, d\, \sigma_{tot}\right), \quad \sigma_{tot} = \frac{1}{N_V d} \cdot \ln\left(\frac{1}{T}\right)$$



VII. Setups at PF2 | VCN & BOA



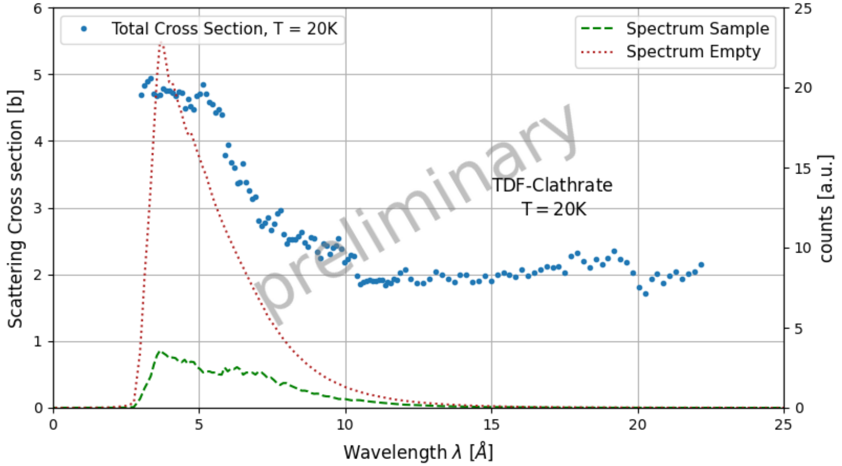


- VCN at ILL & BOA at PSI; multipurpose beam line
- Advanced Shielding & Collimation allowed to measure up to 20 Å at BOA
- Sample in beam: 4 cm
- Temperatures 5 & 20 K

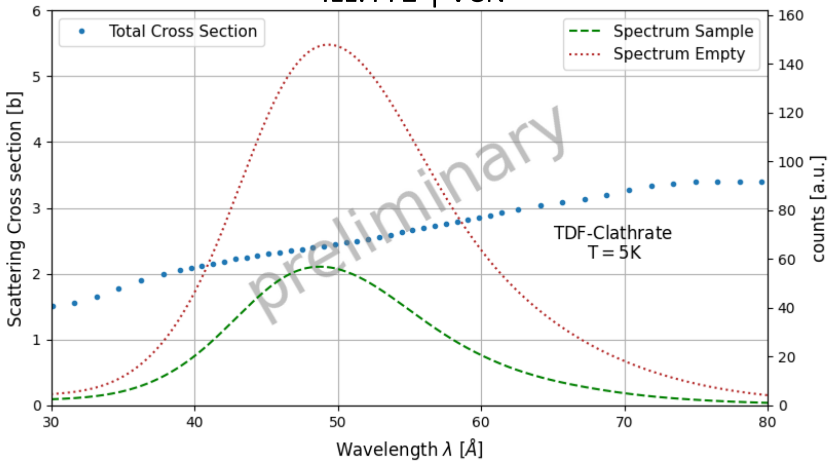
VII. Results PF2 | VCN & BOA

- Observation of the Bragg edges of Type-II Clathrate Hydrates in cold range (left)
- For VCN the cross section is dominated by σ_{inc} of Deuterons (right)
- Additional allocated beamtime in 2023 at PF1B and VCN (ILL)

PSI: BOA

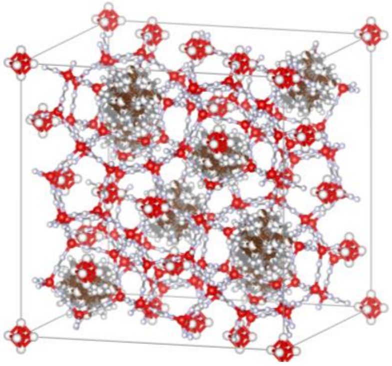


ILL: PF2 | VCN

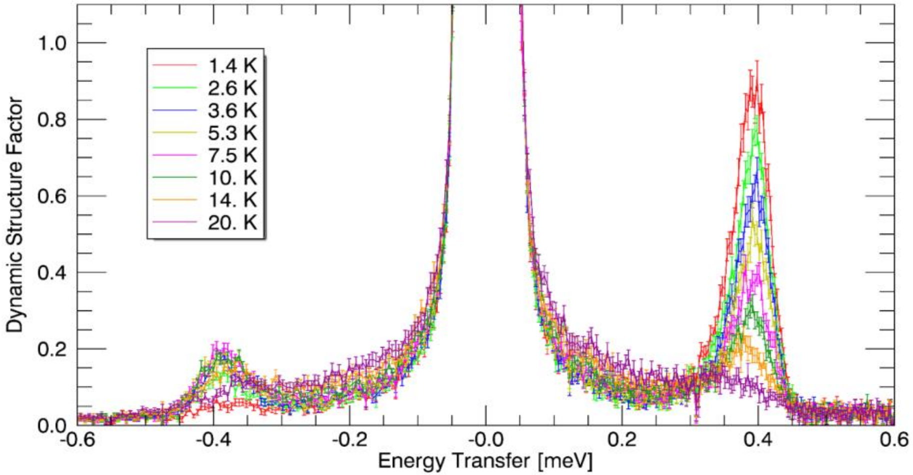


Magnetic Scattering from Encaged O_2

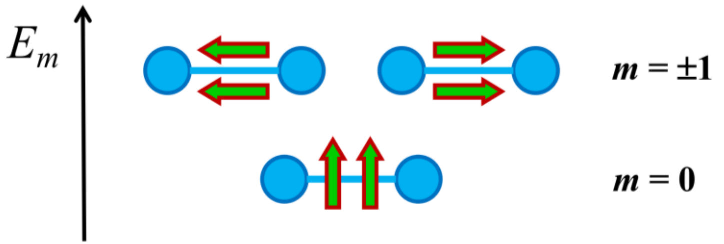
- O_2 is paramagnetic & has a triplet zero-field splitting (~ 0.4 meV)
- Inelastic scattering: E-transfer determined by that zero-field splitting
- Dispersion free ⇒ allows for "cascade cooling"



Talk Shuqi Workshop



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

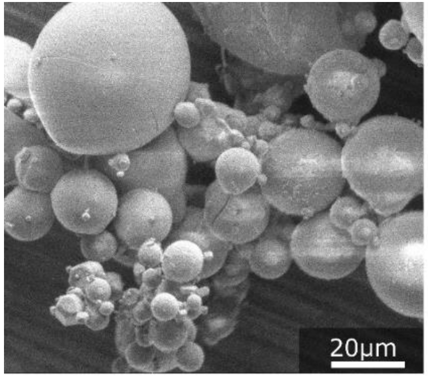


Further Reading: Zimmer, O. (2016): *Phys. Rev. C* **93**, 035503

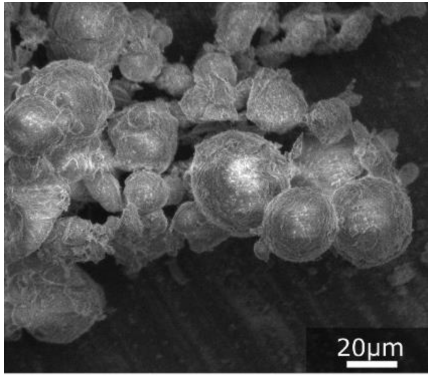
VIII. Looking Ahead

Manufacturing of binary clathrates

- Binary Clathrate Hydrates: Containing two different guest molecules (THF and O_2)
- Ice technique: Starting with tiny grains of THF hydrates ⇒ exposing them to high pressure (> 250 bar) O_2 atmosphere
- Working on a high pressure autoclave



- How do we get uniform grains < 5 µm? Will the structure be preserved in the process?
- ⇒ Spraying, Grinding, Milling and subsequent structure analysis



Kuhs, Werner F. (2018): J. Phys. Chem. Lett. 2018, 9, 12

Experimental Schedule for 2023

Instrument	PF2 | VCN	PF1B	IN5	Panther	Lagrange	D20	IN16
Experiment	Transmissi on	Transmissi on	TOF	TOF	Vibrational Spec.	Diffraction	Backscattering
Sample	THF Hydrates	THF Hydrates	Binary Hydrates	Binary Hydrates	Binary & THF - Hydrates	Binary & THF - Hydrates	Binary Hydrates
Status	allocated	allocated	proposed	proposed	proposed	proposed	proposed
Duration	20 days	10 days	3 days	3 days	5 days	2 days	2 days
When?	May 25 – June 11	June 12 – June 22	3rd - 4th cycle	3rd - 4th cycle	3rd - 4th cycle	3rd - 4th cycle	3rd - 4th cycle

Acknowledgements

PhD-Advisors:

- Oliver Zimmer
- Richard Wagner

Collaborators:

- HighNess Collaboration
- Arnaud Desmedt (ism Bordeaux)

Instrument Scientists (ILL):

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

Instrument Scientists (PSI):

- Matteo Busi (BOA)
- Uwe Filges (BOA)

Questions ?

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Thank you for your attention !

HighNESS is funded by the European Framework for Research and Innovation Horizon 2020, under grant agreement 951782