2nd ISIS-CSNS Workshop
Neutron Total Scattering: Methods, Instrumentation and Data Modelling

Prof Robert McGreevy
Director of the ISIS Neutron and Muon Source

Science & Technology Facilities Council
ISIS

Reverse Monte Carlo Simulation: a new technique for the determination of disordered structures

McGreevy R L and Pusztai L, *Molecular Simulation* **1**(1988) 35

We have developed a new technique, based on the standard Monte Carlo simulation method with Markov chain sampling, where a set of three dimensional particle configurations are generated that are consistent with the experimentally measured structure factor, A(Q), and radial distribution function, g(r), of a liquid or other disordered system. Consistency is determined by a standard χ^2 test using the experimental errors. No input potential is required. We present initial results for liquid argon. Since the technique can work directly from the structure factor it promises to be extremely powerful for modelling the structures of glasses or amorphous materials. It also has many other advantages in multicomponent systems and as a tool for experimental data analysis.

Key words: Monte Carlo, structure factor, radial distribution function, liquid, glass.

Total scattering methods can be applied to many problems

Science & Technology Facilities Council
ISIS

And can answer many questions

Science & Technology Facilities Council
ISIS

ISIS team for the PDF and Total Scattering Workshop

Daniel Bowron

Sam Callear

Alex Hannon

Dave Keen

Helen Playford

Alan Soper

7^{th}-9^{th} November 2016

Science & Technology Facilities Council
ISIS

Calls for proposals to use ISIS are made twice a year
Submission deadlines are 16th April and 16th October

http://www.isis.stfc.ac.uk/apply-for-beamtime/apply-for-beamtime2117.html

Newton Funding for Indian, Chinese and South African researchers

ISIS has been awarded funds as part of the UK Government's Newton Fund to support researchers from China, India and South Africa to use ISIS.

ISIS is able to support a limited number of experiments each round from users from these three countries. For supported experiments we can fund up to two researchers to come to ISIS for the experiment, and will pay for economy flights, accommodation and food costs for those researchers. Accommodation arrangements should be made through the ISIS user office, who will also provide a per diem amount for food; claims for flights should also be made through the user office and require supporting receipts.

To apply for Newton Funding for an ISIS experiment, please tick the box on page 3 of the ISIS online proposal system saying that you would like funding when you are creating your beamtime application (this mechanism will be available from ISIS round 16/2 onwards – for experiments approved before then, please contact Philip King to ask about the possibility of funding).

Details of the UK Government's Newton Fund can be found online.

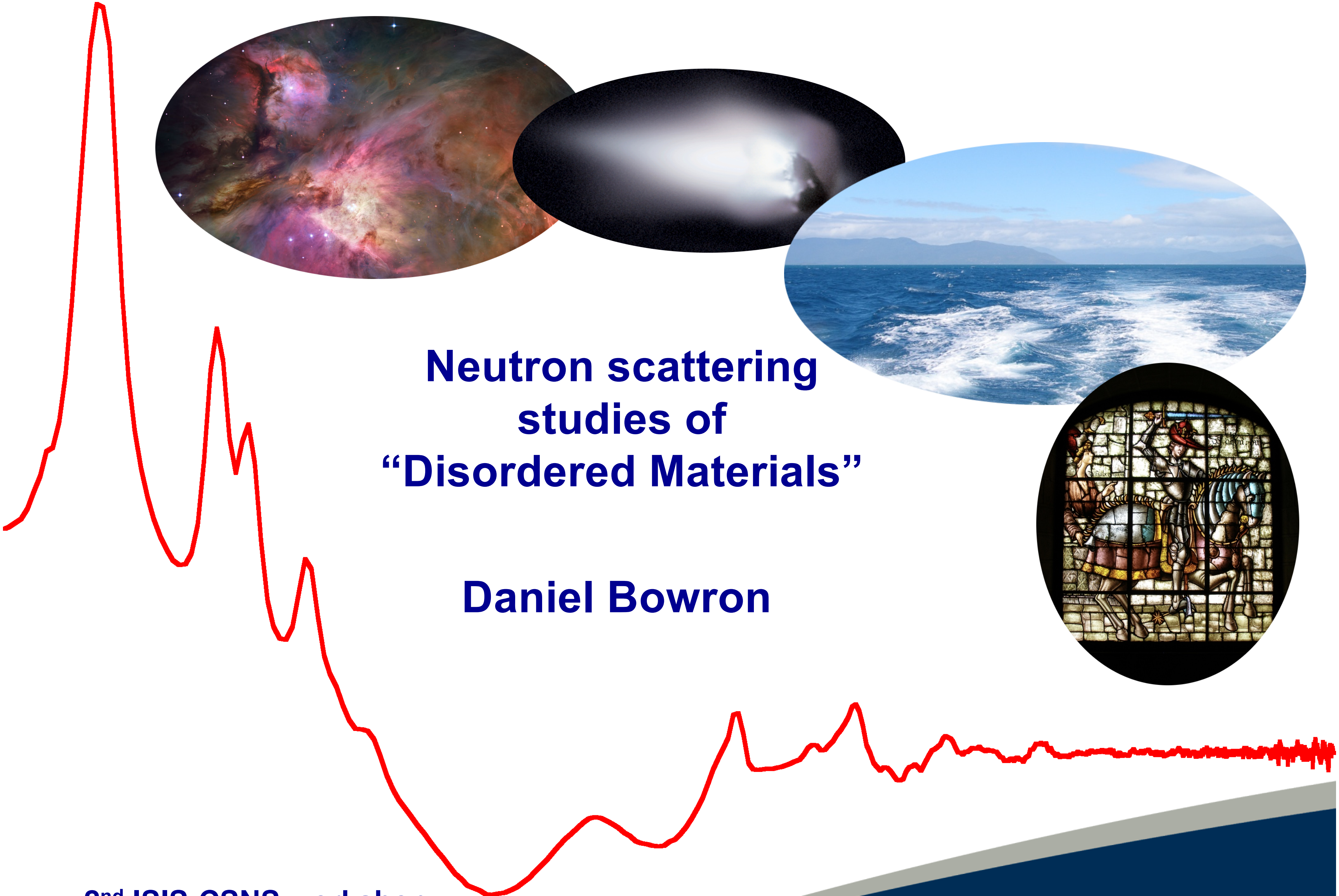
SHARE

Science & Technology Facilities Council
ISIS

Wishing you all a successful and informative workshop and hoping to see you at ISIS one day

Prof Robert McGreevy
Director of the ISIS Neutron and Muon Source

Science & Technology Facilities Council
ISIS



Neutron scattering studies of "Disordered Materials"

Daniel Bowron

2nd ISIS-CSNS workshop
PDF and Total Scattering Methods
7th – 9th November 2016

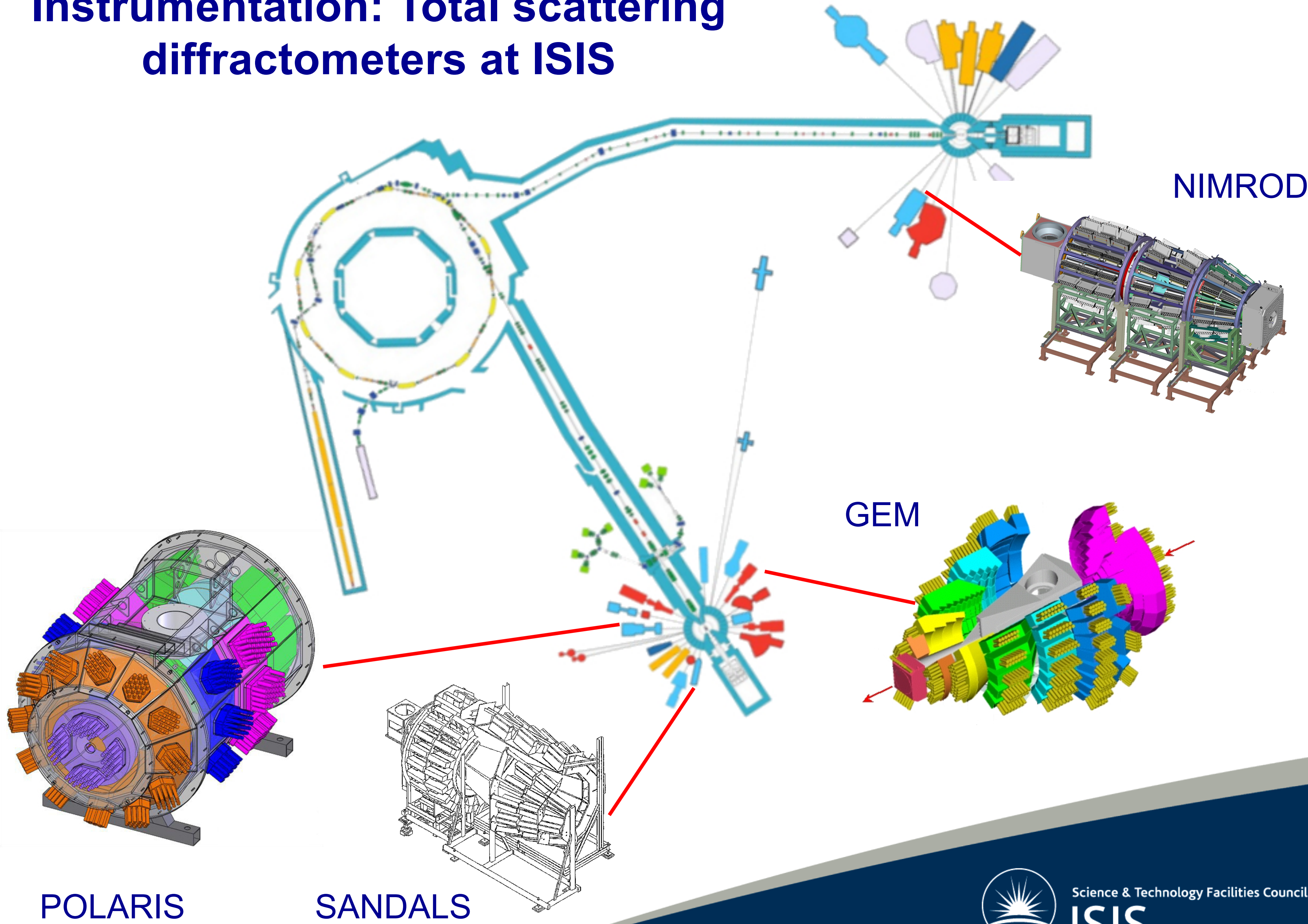
Science & Technology Facilities Council
ISIS

2nd ISIS-CSNS workshop
PDF and Total Scattering Methods
7th – 9th November 2016

Aims of workshop

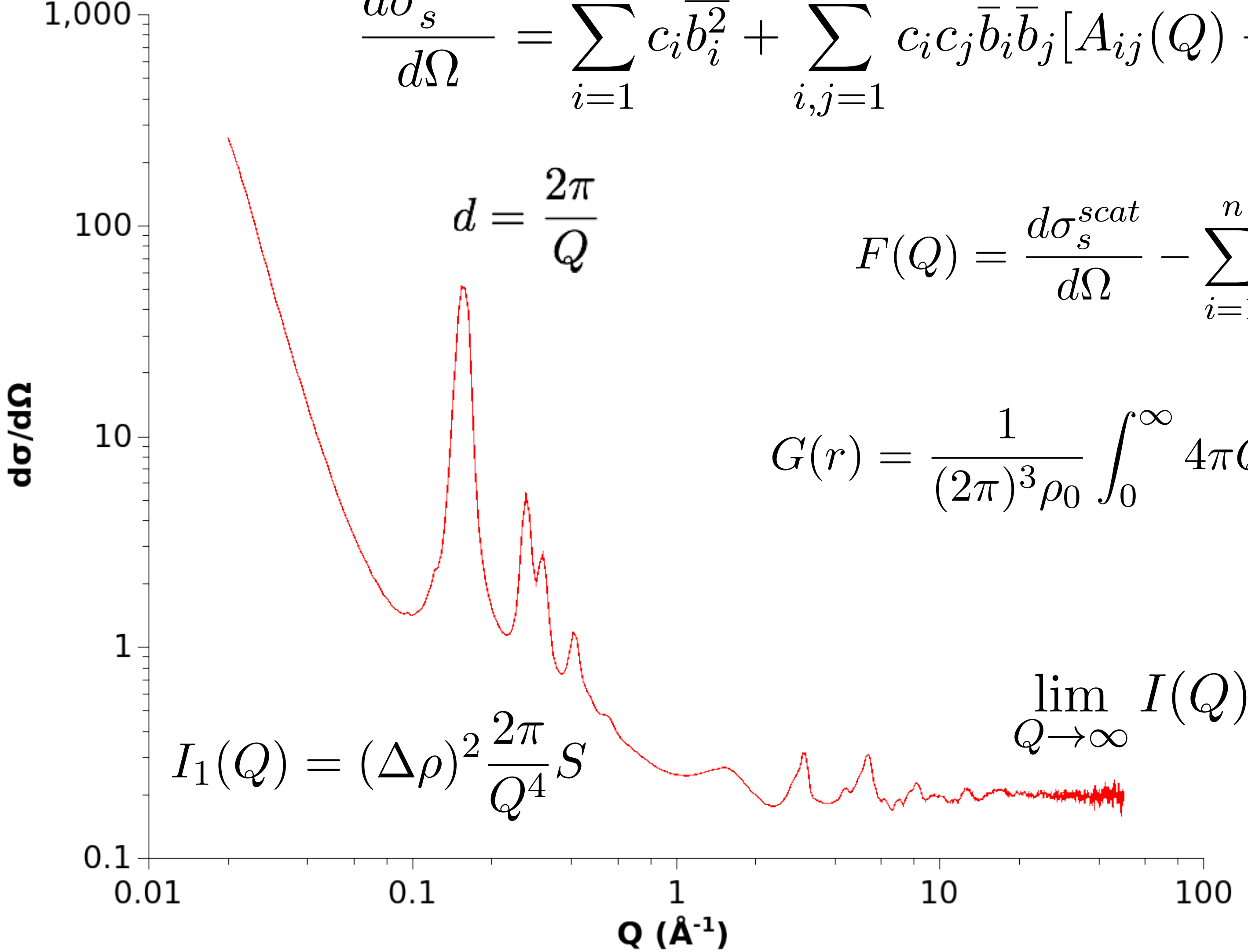
(1) Give some examples of the scientific applications of neutron total scattering
(2) Outline instrumentation requirements
(3) Give an overview of underlying scattering theory
(4) Explain the corrections that need to be applied to measured data
(5) Introduce methods that can be used to bring understanding of scattering data using atomistic models: Empirical Potential Structure Refinement (EPSR) and Reverse Monte Carlo (RMC) modelling

Instrumentation: Total scattering diffractometers at ISIS



Theory: Differential Scattering Cross Section

$$\frac{d\sigma_s^{scat}}{d\Omega} = \sum_{i=1}^{n} c_i \overline{b_i^2} + \sum_{i,j=1}^{n} c_i c_j \bar{b}_i \bar{b}_j [A_{ij}(Q) - 1]$$



$$d = \frac{2\pi}{Q}$$

$$F(Q) = \frac{d\sigma_s^{scat}}{d\Omega} - \sum_{i=1}^{n} c_i \overline{b_i^2}$$

$$G(r) = \frac{1}{(2\pi)^3 \rho_0} \int_0^\infty 4\pi Q^2 F(Q) \frac{\sin(Qr)}{Qr} dQ$$

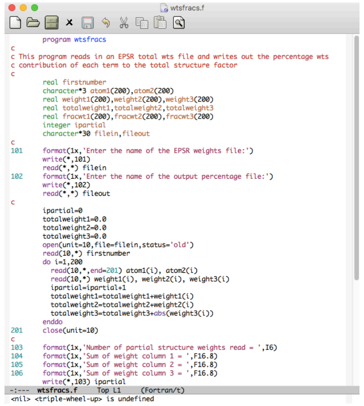
$$\lim_{Q\to\infty} I(Q) = \frac{\rho t f \sum_i c_i \sigma_i^{scat}}{4\pi}$$

$$I_1(Q) = (\Delta\rho)^2 \frac{2\pi}{Q^4} S$$

$$Q = \frac{4\pi}{\lambda} \sin\theta$$

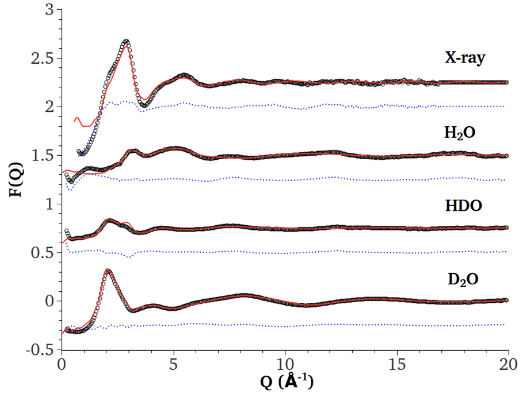
Science & Technology Facilities Council
ISIS

Modelling: Turning total scattering data into scientific impact requires more than just a good diffractometer



Code

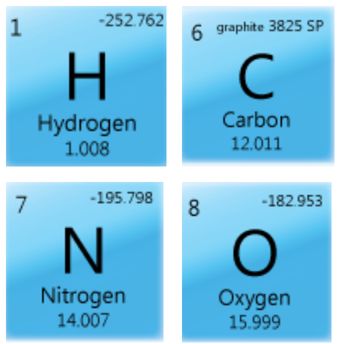
Data



Mathematics

$$U_{\alpha\beta}^{LJ} = 4\epsilon_{\alpha\beta}\left[\left(\frac{\sigma_{\alpha\beta}}{r}\right)^{12} - \left(\frac{\sigma_{\alpha\beta}}{r}\right)^{6}\right]$$

Model

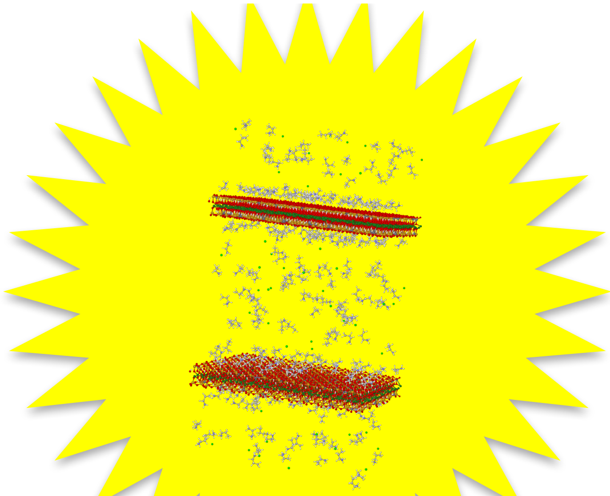


ρ

σ

ε

Constraints



Scientific Discovery

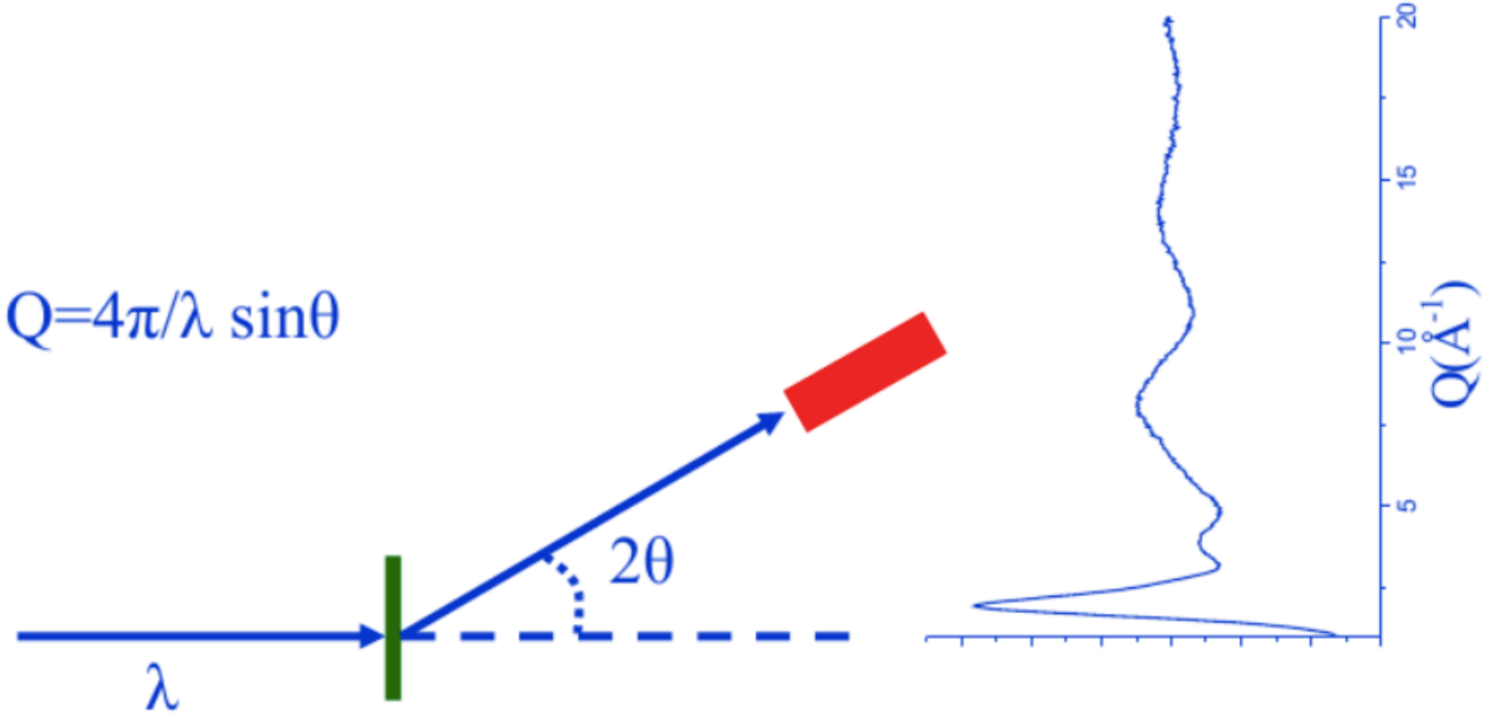
Science & Technology Facilities Council

ISIS

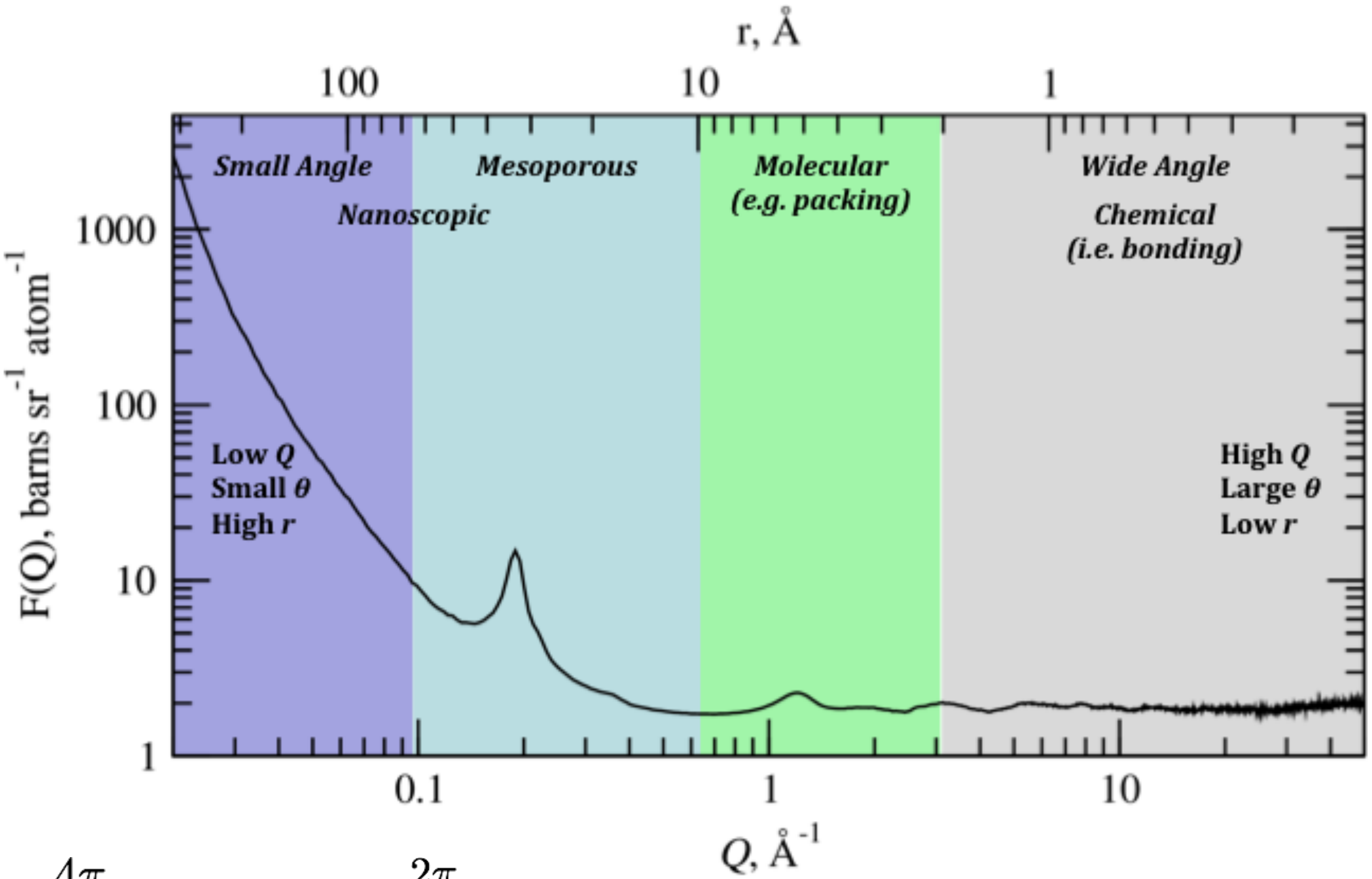
Part 1

The scattering experiment

Schematic of a neutron scattering measurement



Scattering probes a wide range of length scales



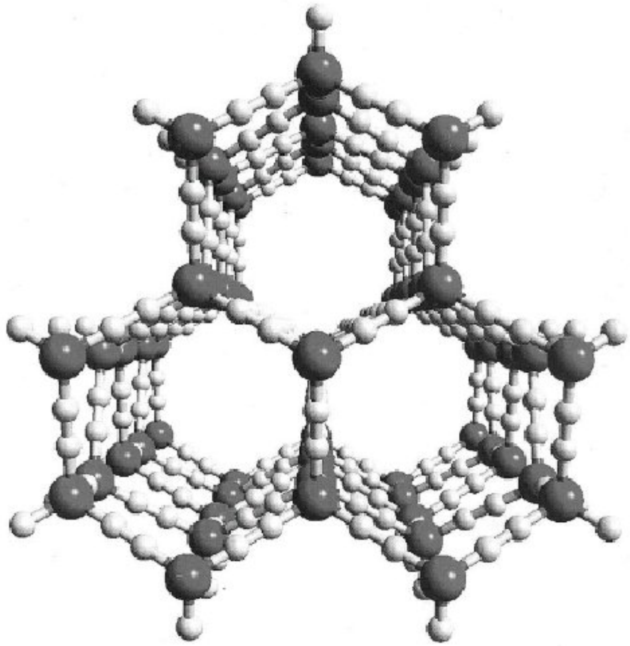
$$Q = \frac{4\pi}{\lambda}\sin\theta$$

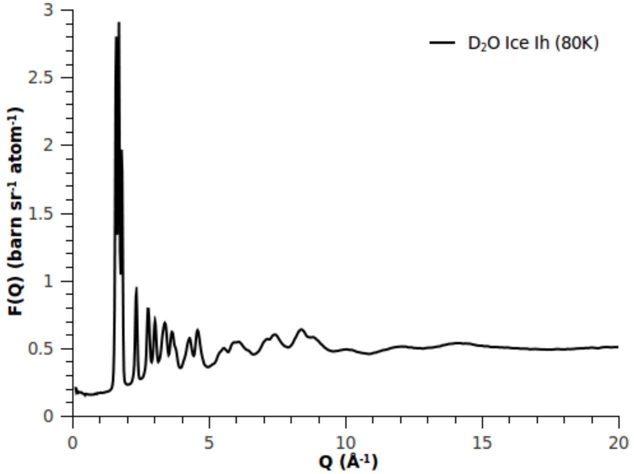
$$d = \frac{2\pi}{Q}$$

$$r = \frac{\pi}{Q}$$

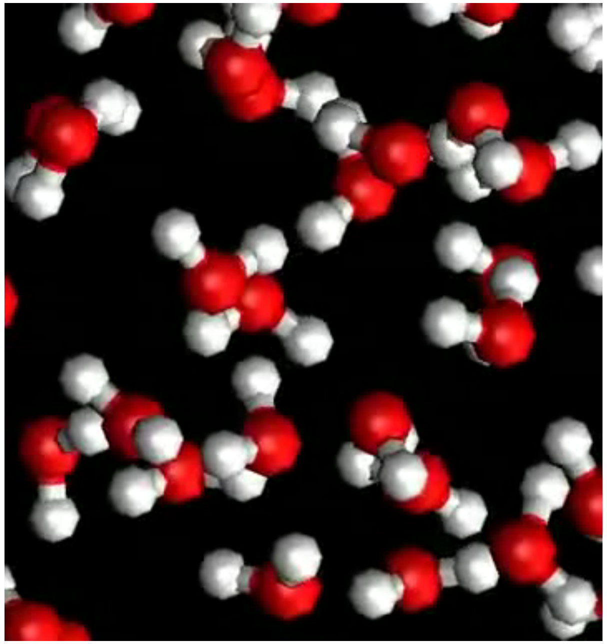
Total scattering methods are widely applicable

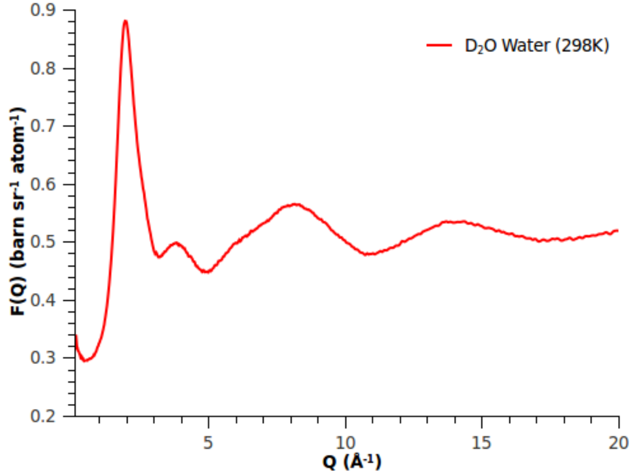
Crystalline solid



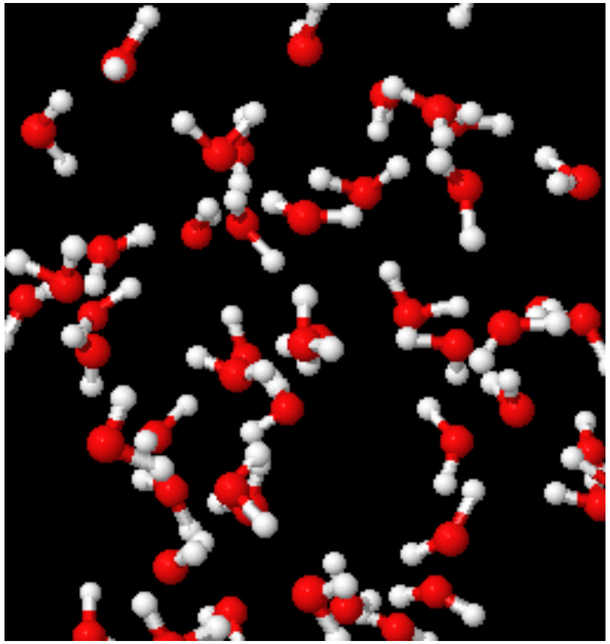


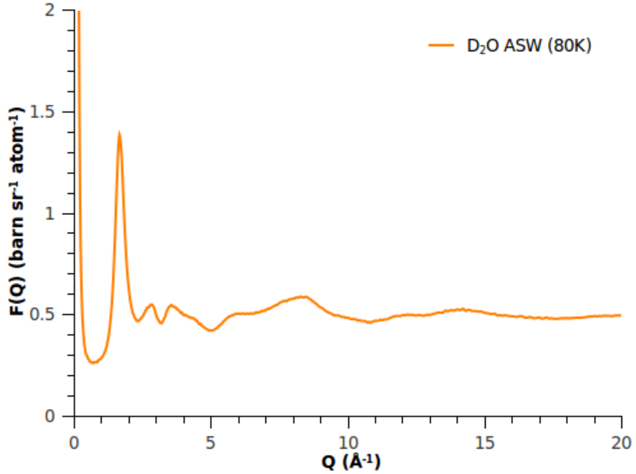
Liquid



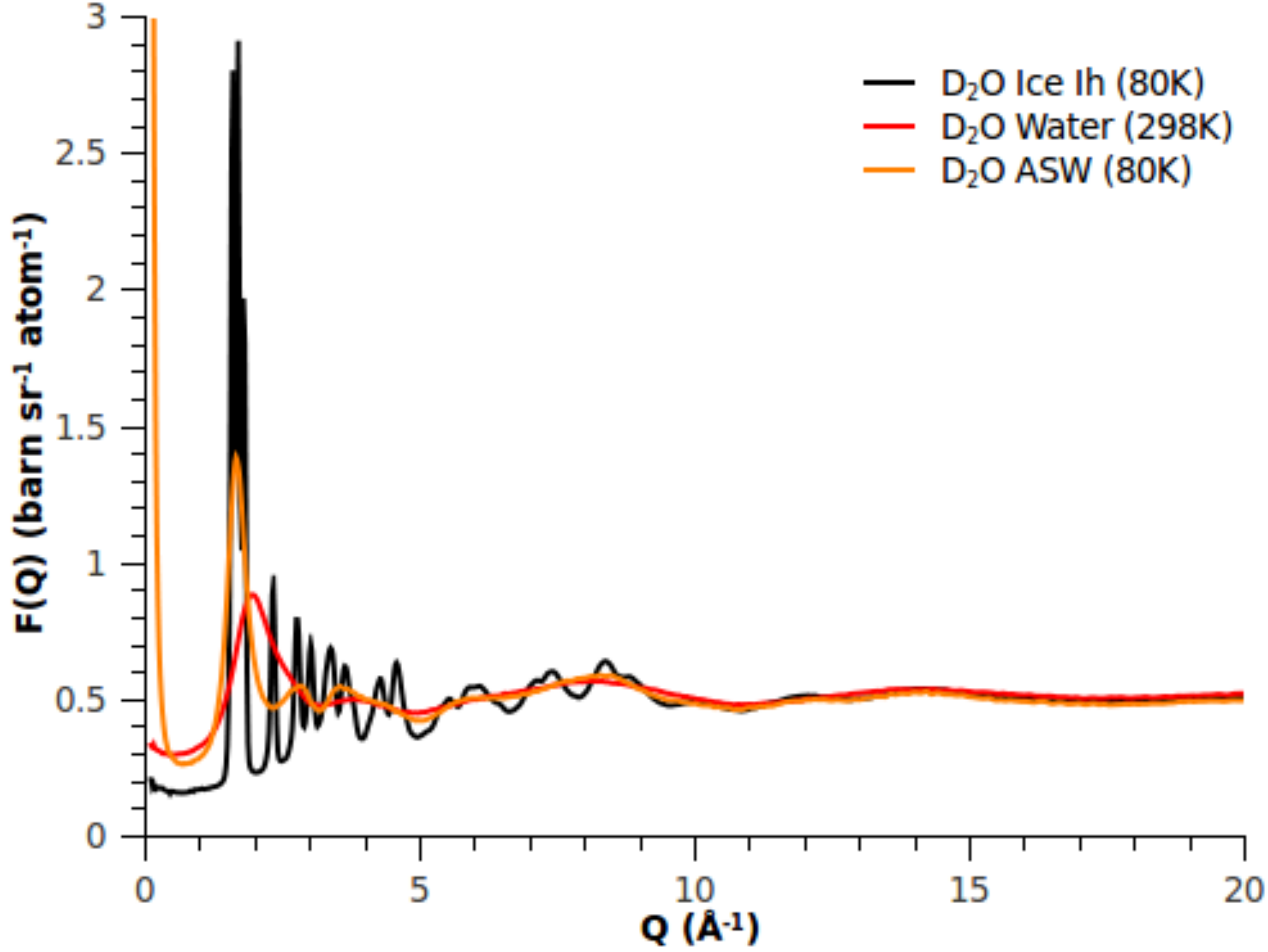


Glass





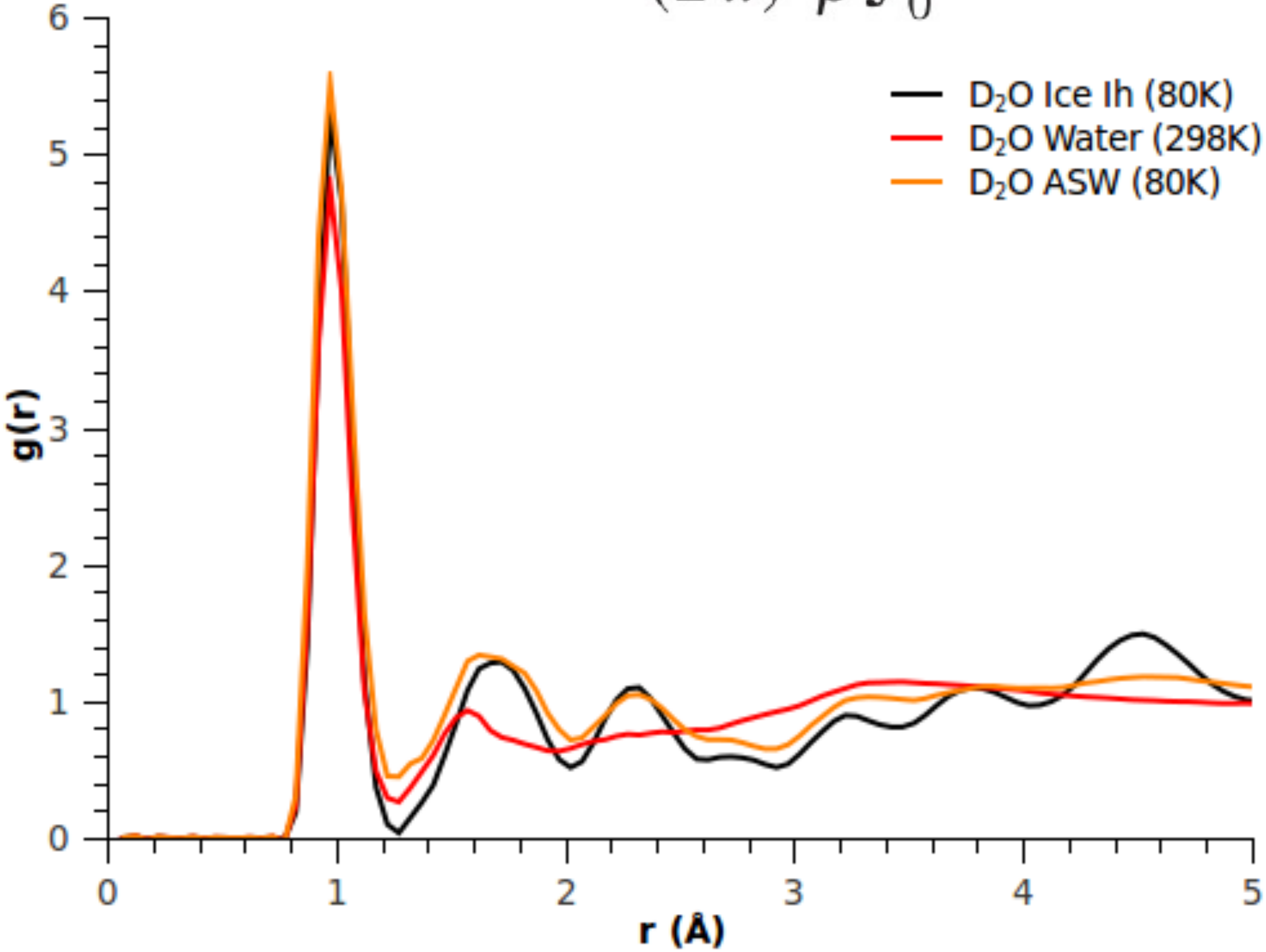
Solid ➔ Liquid ➔ Glass

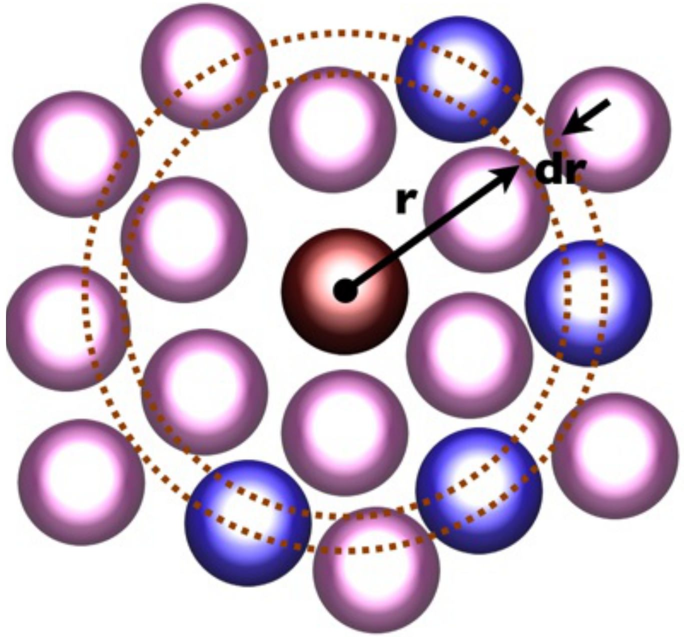


Science & Technology Facilities Council
ISIS

The radial distribution function

$$g(r) - 1 = \frac{1}{(2\pi)^3 \rho} \int_0^\infty 4\pi Q^2 F(Q) \frac{\sin Qr}{Qr} dQ.$$

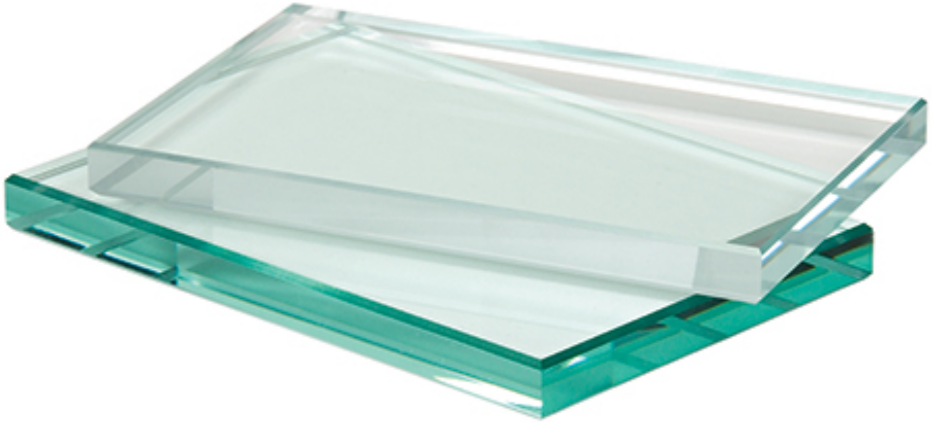




Part 2

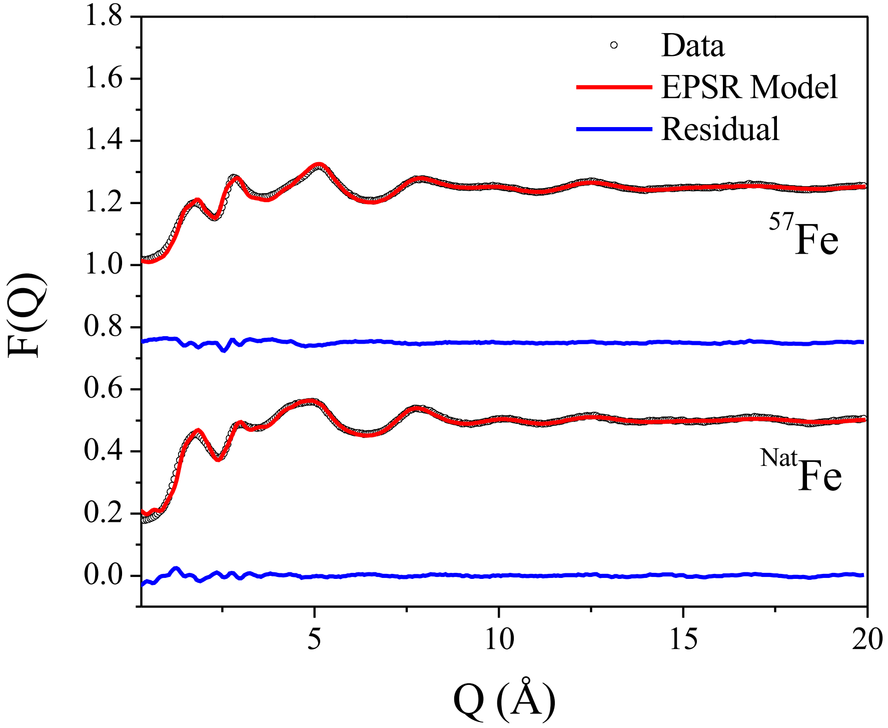
Examples of application

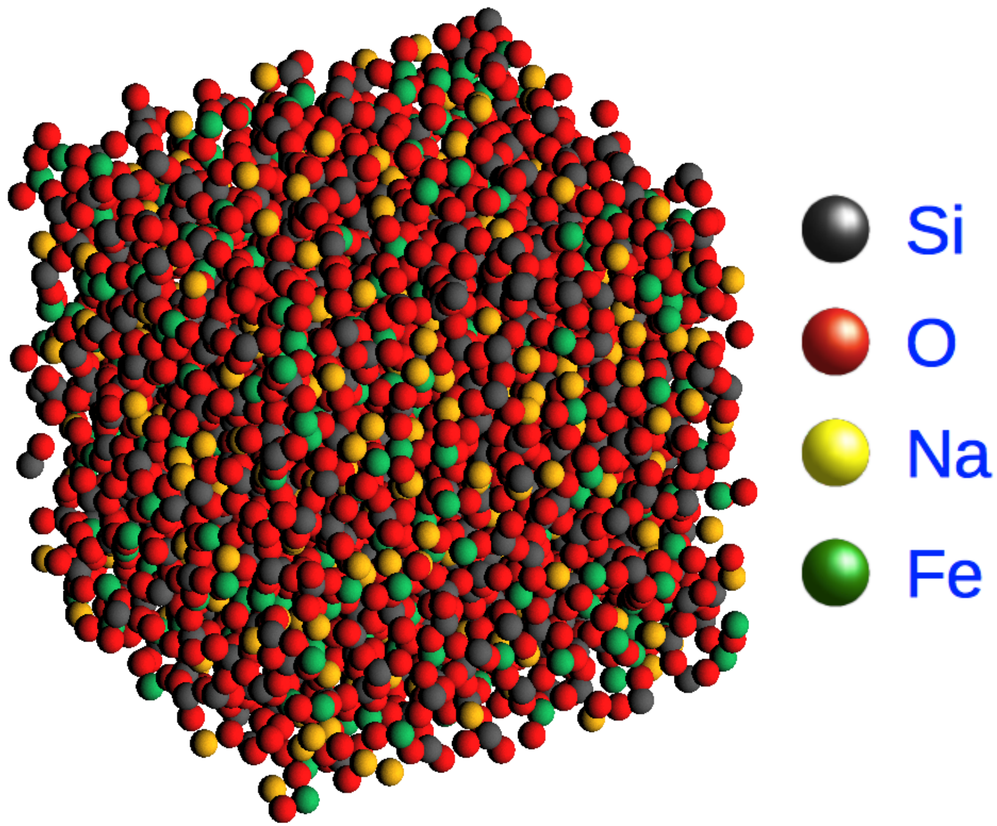
The structure of glass and ceramic glazes



Science & Technology Facilities Council
ISIS

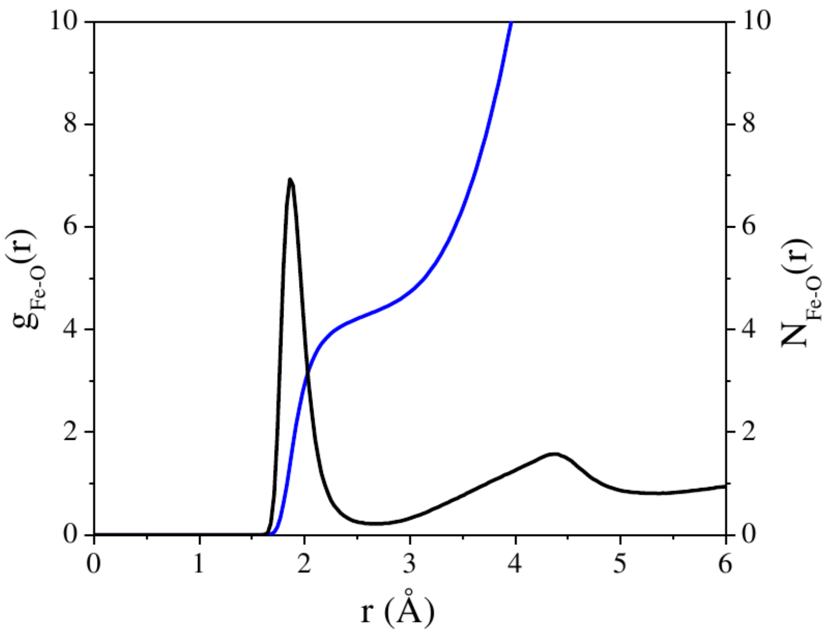
Investigating the incorporation of iron in $FeNaSi_2O_6$ glass

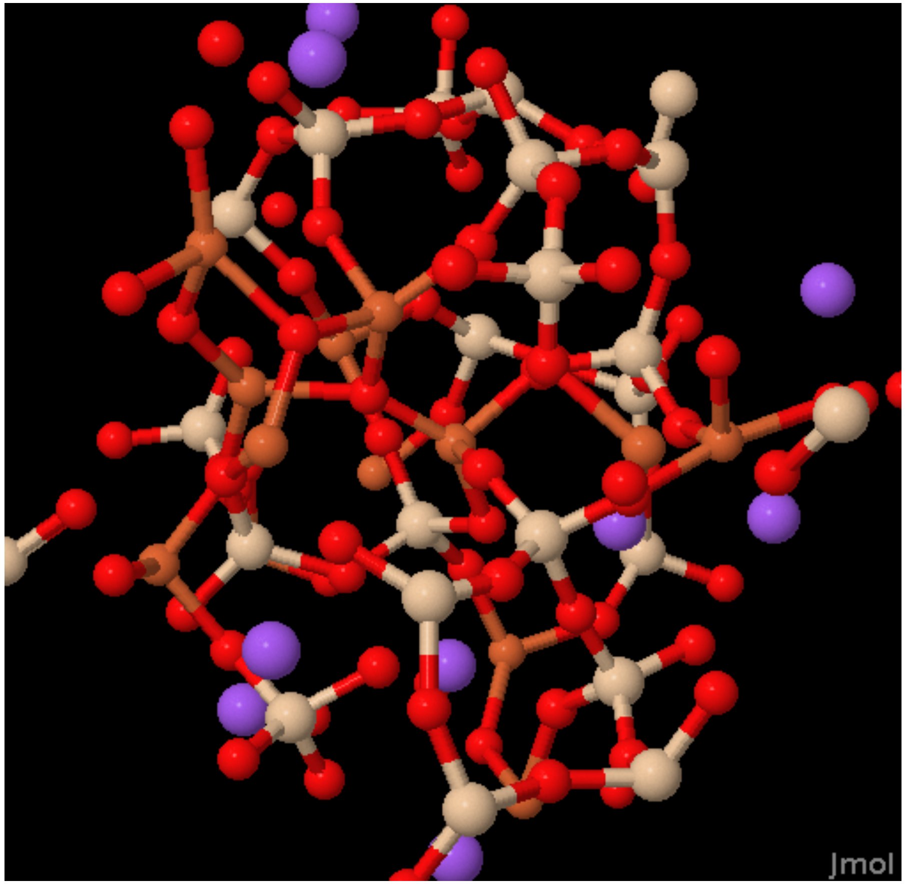


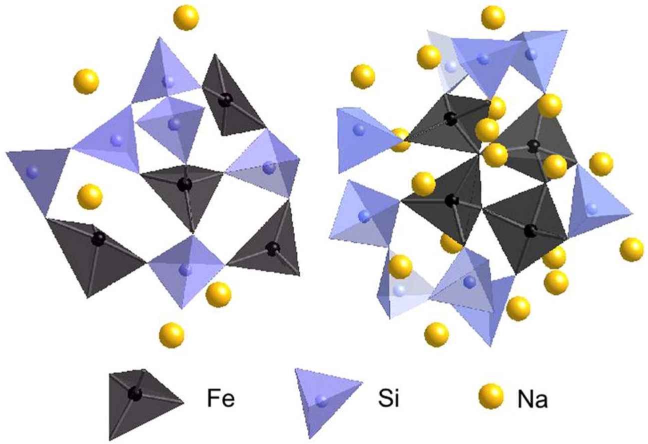


C. Weigel, L. Cormier, L. Galoisy, G. Calas, D. Bowron and B.Beuneu
App. Phys. Lett. 89 (2006) 141911

Investigating the incorporation of iron in $FeNaSi_2O_6$ glass







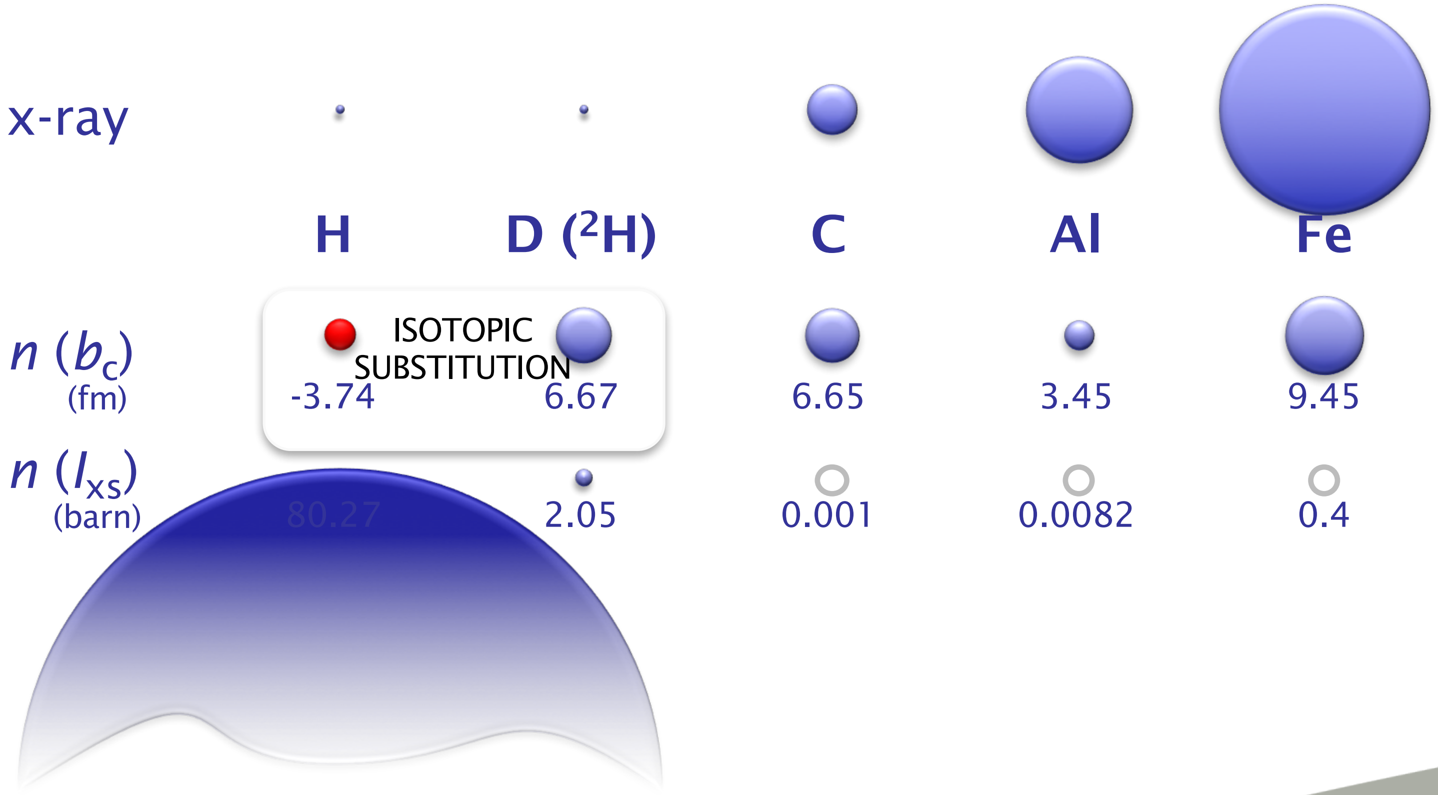
76% of total iron found as Fe^{3+} in tetrahedral sites
Remaining iron found in five-coordinate sites, 4% and 20% respectively for Fe^{3+} and Fe^{2+}

C. Weigel, L. Cormier, L. Galoisy, G. Calas, D. Bowron and B.Beuneu
App. Phys. Lett. 89 (2006) 141911

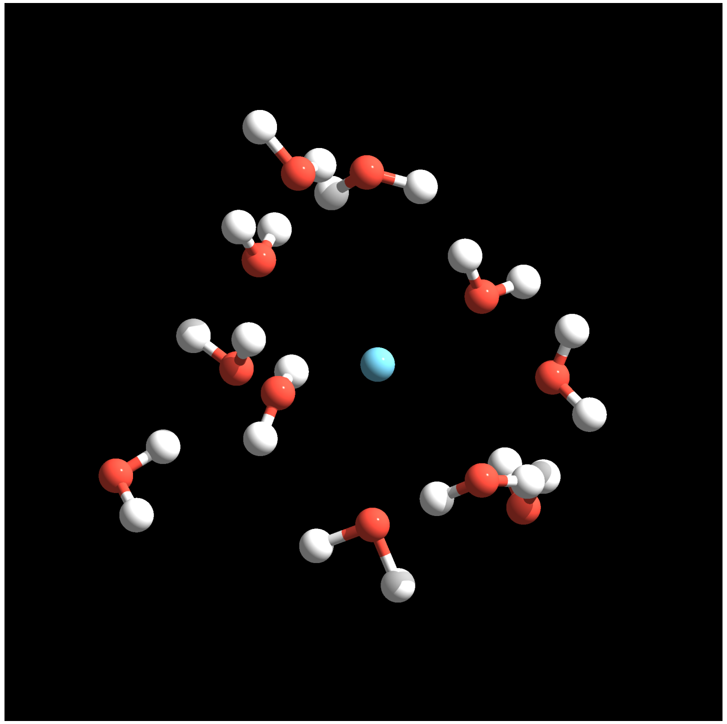
The structure of complex solvent media

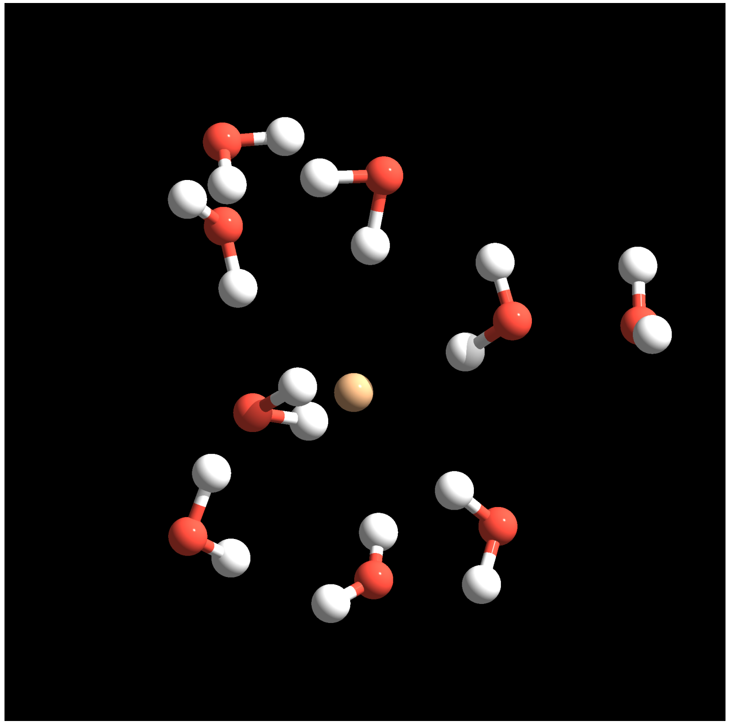
H/D isotopic substitution – a route to enhanced insight

- Neutrons interact weakly with matter, interacting with only the nucleus
- Neutrons are sensitive to **element and isotope**
  - X-ray scattering of elements depends on number of electrons
  - Neutron scattering of elements varies across periodic table



The neutron's view of hydrogen lets us….





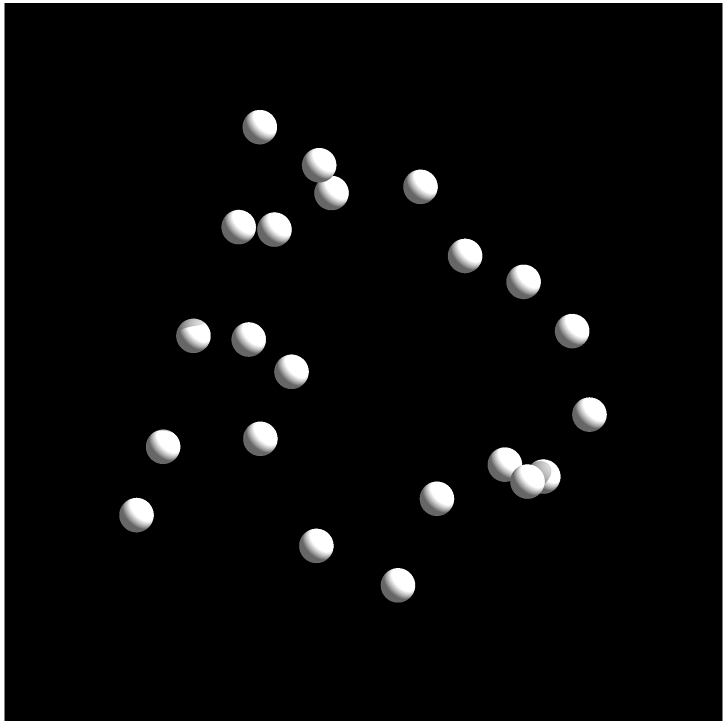
+
v
e

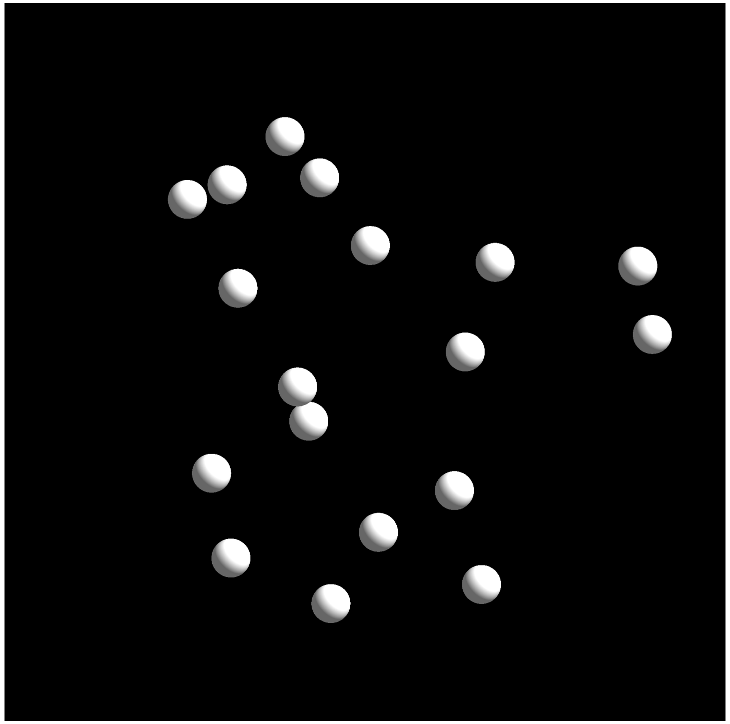
See all the atoms

-ve

D.T.Bowron, J.L.Finney and A.K.Soper
J. Phys. Chem. B **102** 3551-3563 (1998)

The neutron's view of hydrogen lets us….



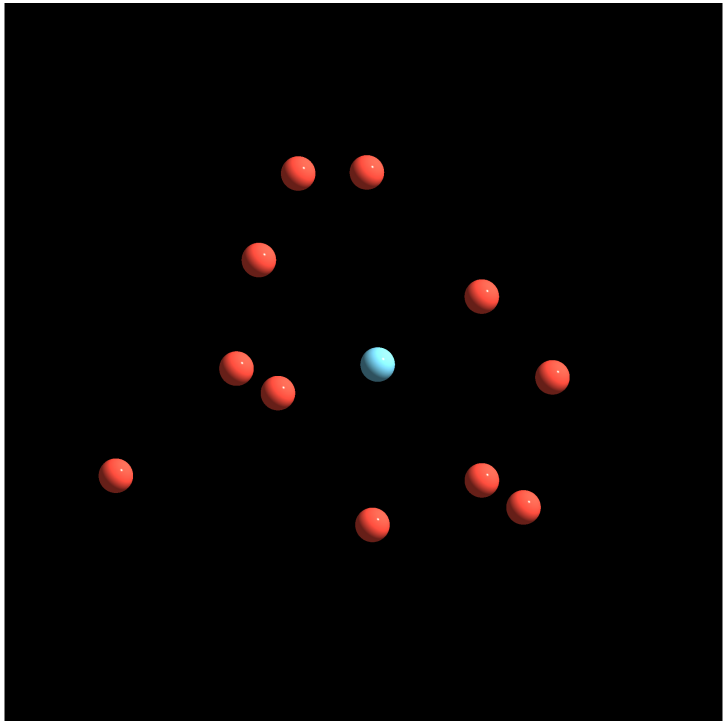


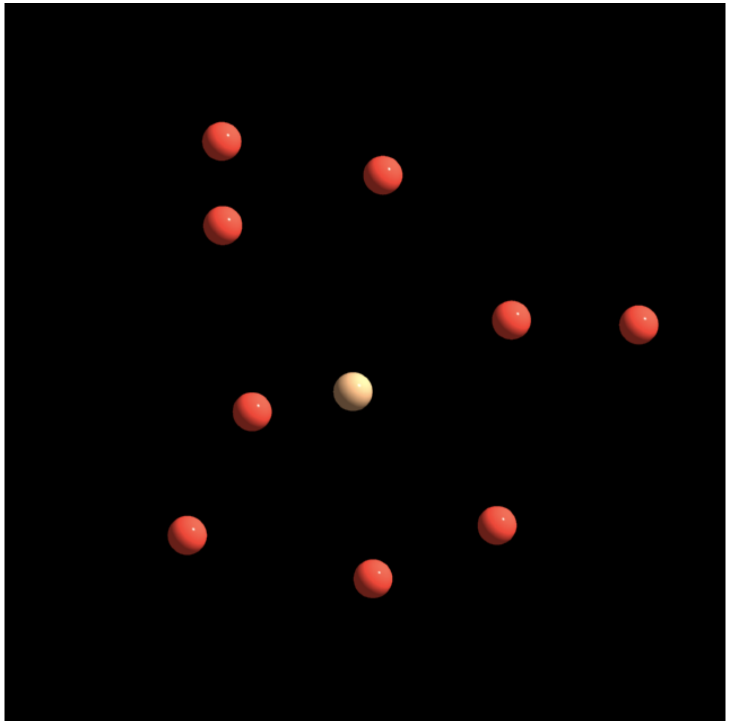
Only see the hydrogen atoms

D.T.Bowron, J.L.Finney and A.K.Soper
J. Phys. Chem. B **102** 3551-3563 (1998)

Science & Technology Facilities Council
ISIS

The neutron's view of hydrogen lets us….

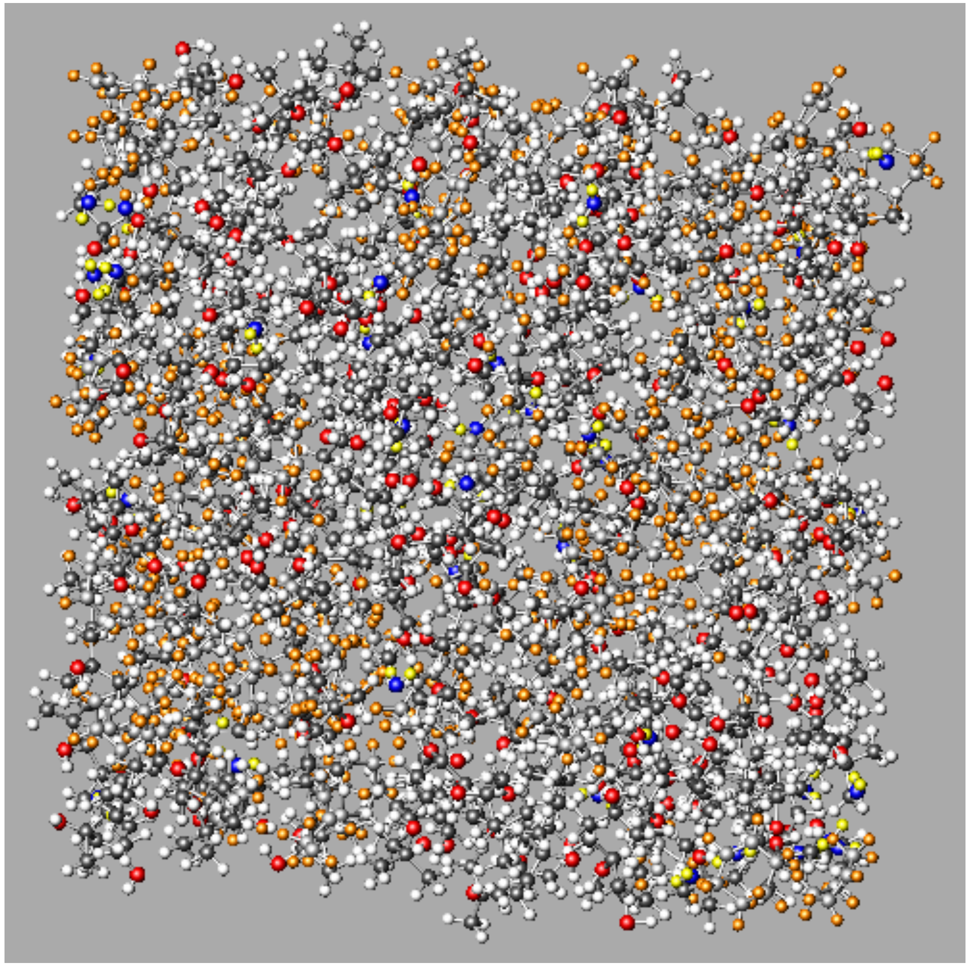




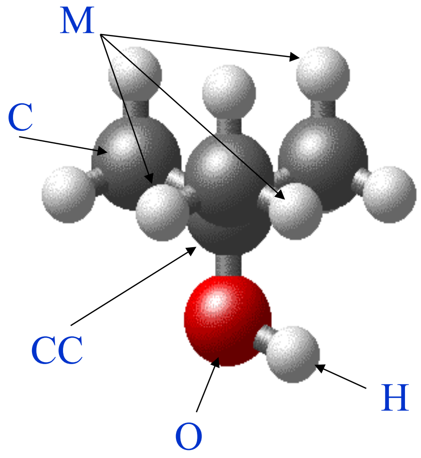
Make the hydrogen invisible

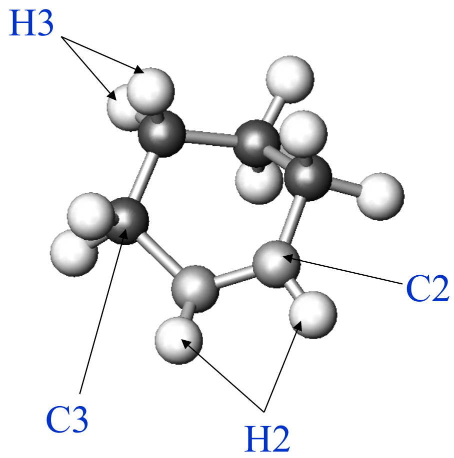
D.T.Bowron, J.L.Finney and A.K.Soper
J. Phys. Chem. B **102** 3551-3563 (1998)

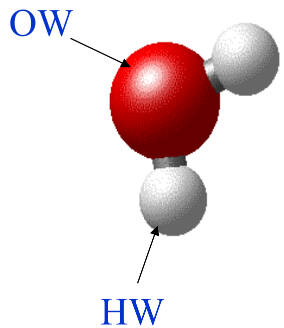
6:1:2 tertiary butanol:water:cyclohexene



240 $(CH_3)_3COH$ molecules
40 H_2O molecules
80 C_6H_{10} molecules



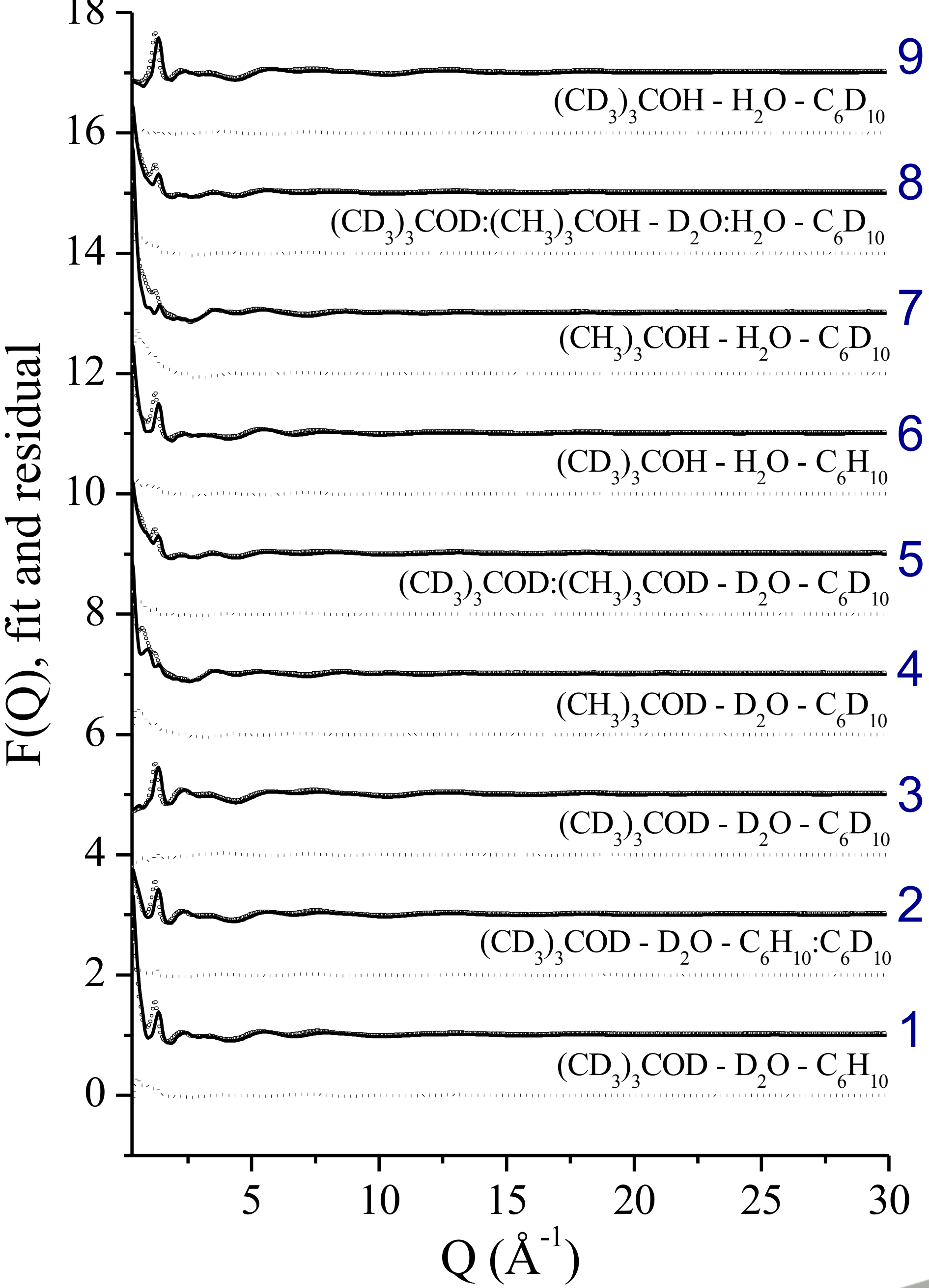




D.T.Bowron and S. Diaz Moreno
JPC-B **109** 16210-16218 (2005)

Science & Technology Facilities Council
ISIS

6:1:2 tertiary butanol:water:cyclohexene



Complex systems require comprehensive experiments

1,2 & 3 give cyclohexene-cyclohexene correlations

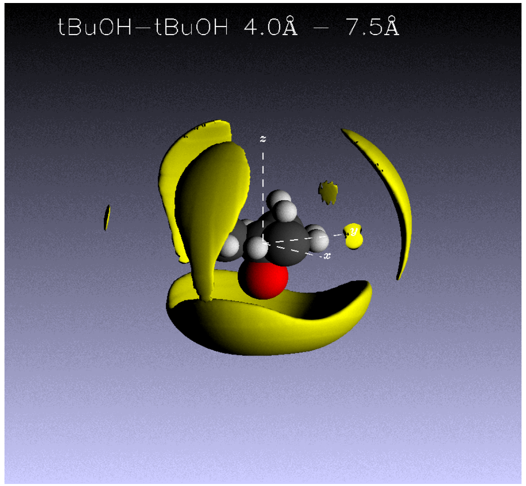
3,4 & 5 give tertiary butanol – tertiary butanol correlations

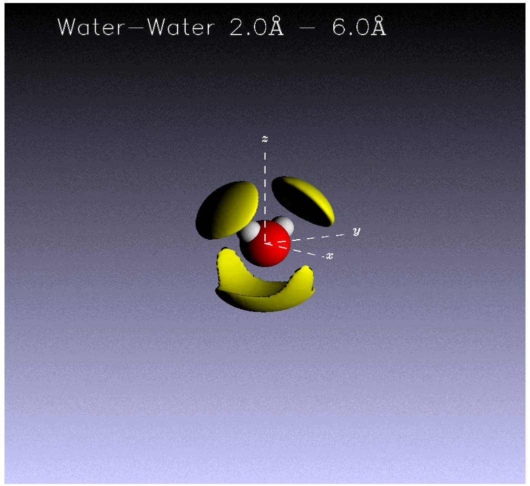
3,7 & 8 give tertiary butanol and water correlations

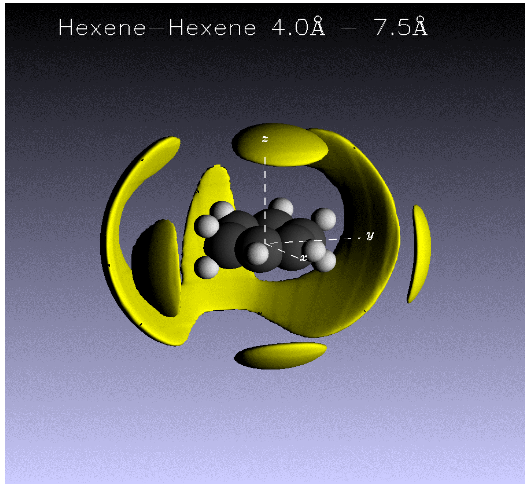
3 & 9 or 1 & 6 give hydroxyl hydrogen and water hydrogen information

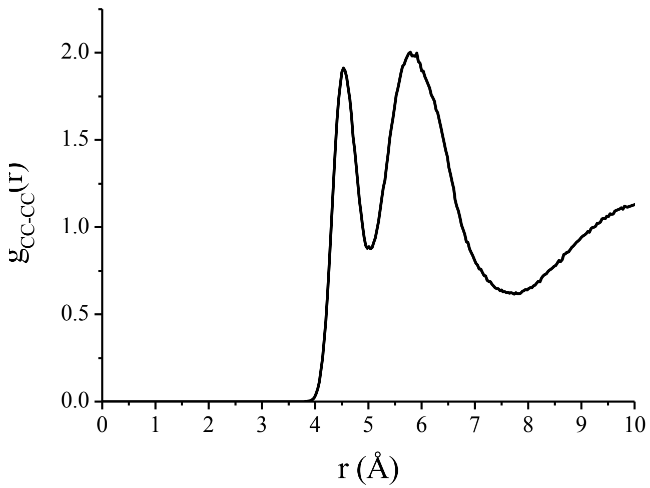
Science & Technology Facilities Council ISIS

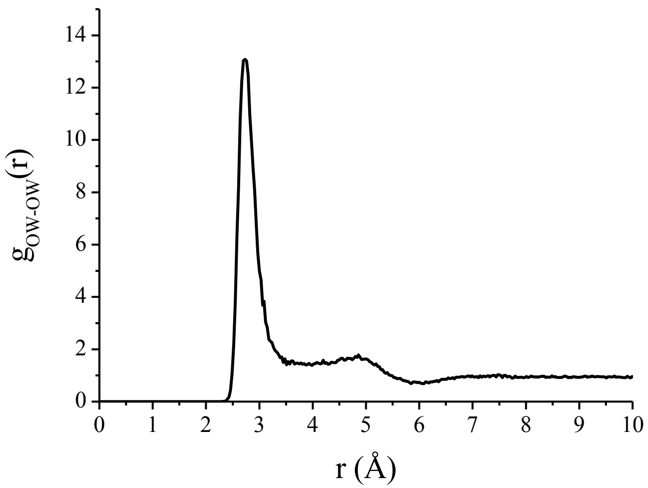
6:1:2 tertiary butanol:water:cyclohexene

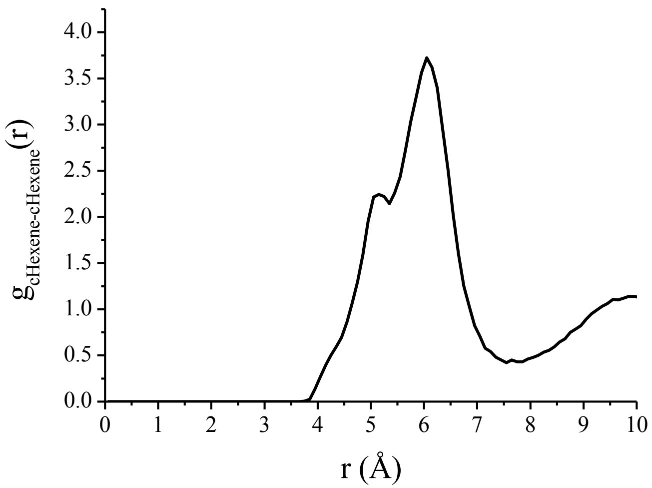












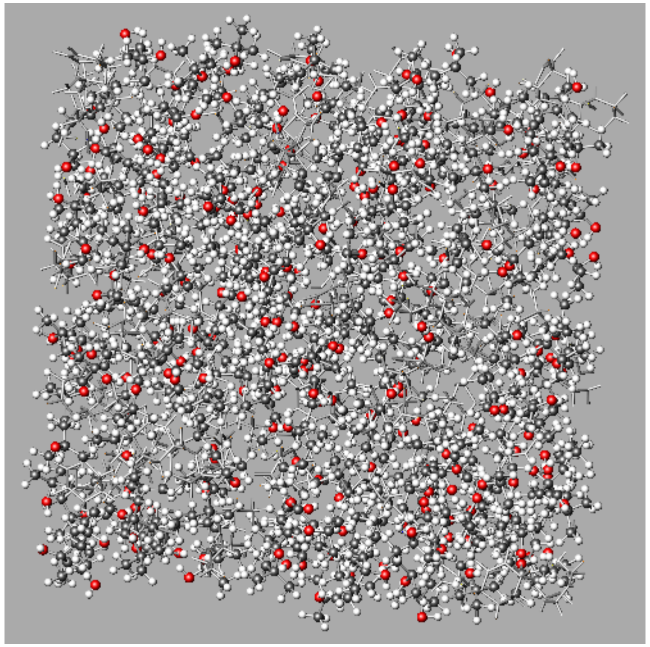
1.4±0.1 between 4.0Å and 5.0Å
7.5±0.3 between 5.0Å and 7.5Å

0.5±0.1 between 2.3Å and 3.8Å

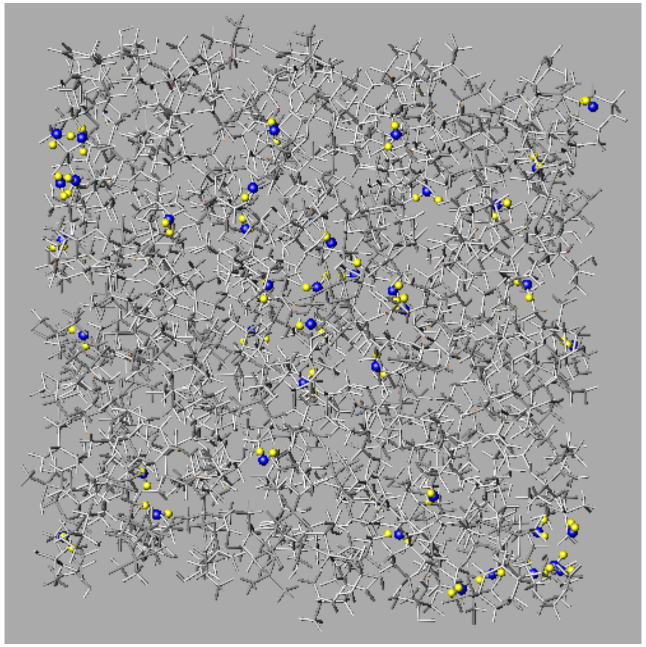
≈4.2 between 3.6Å and 7.5Å

Like - like interactions in the tri-molecular mixture

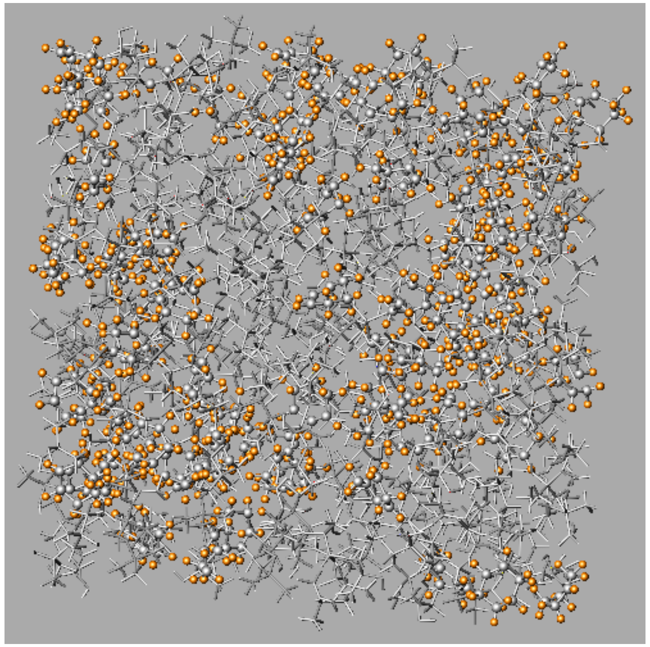
6:1:2 tertiary butanol:water:cyclohexene



Tertiary Butanol



Water



Cyclohexene

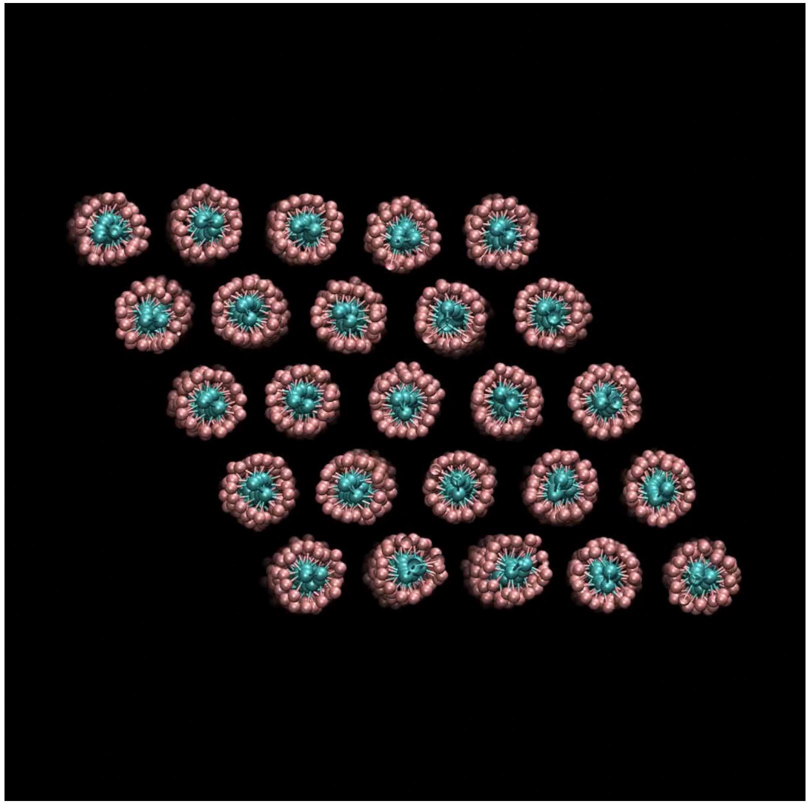
Mixing state of a 6:1:2 tertiary butanol-water-cyclohexene solution

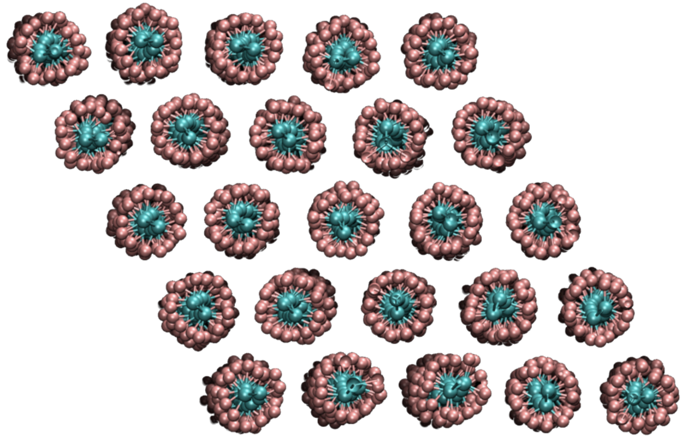
The structure correlations in polymers

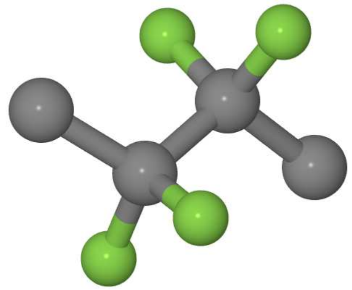
Total scattering – diffuse plus Bragg

Structural disordered in crystalline and partially crystalline systems is a growing area of interest. Examples include the correlations between polymer chains in materials such as PTFE (crystalline) and KelF (PCTFE) (disordered crystalline).

A.K.Soper, K.Page and A.Llobet, J. Phys.: Condens. Matter **25** 454219 (2013)

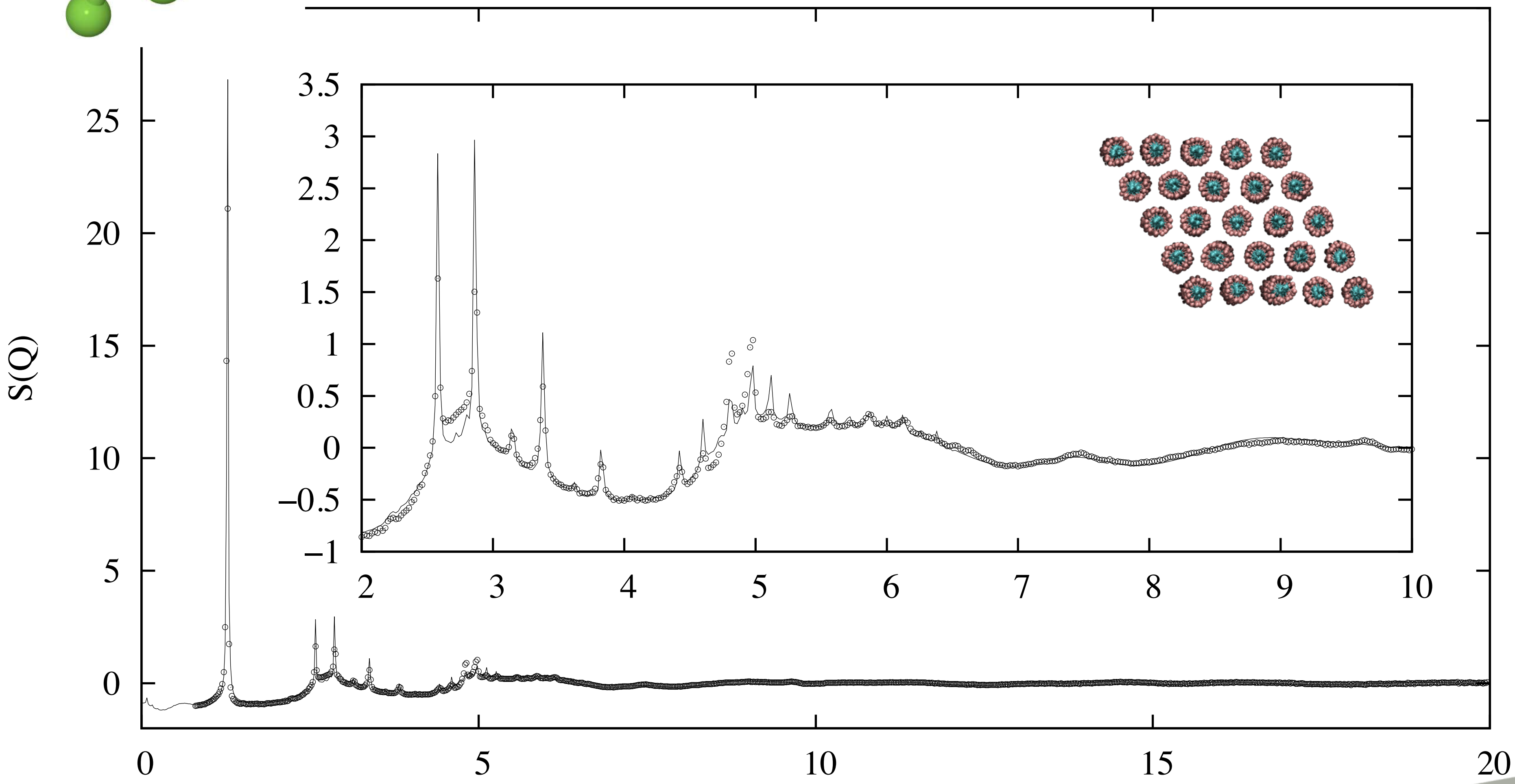






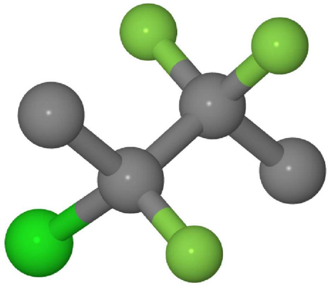
PTFE

Data from LANSCE: HIPD & NPDF



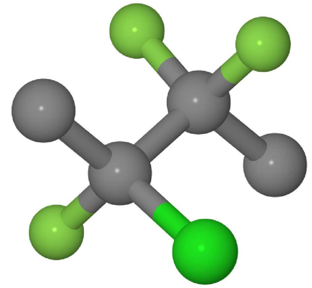
Refined model (EPSR) of crystalline PTFE with both diffuse and Bragg scattering features

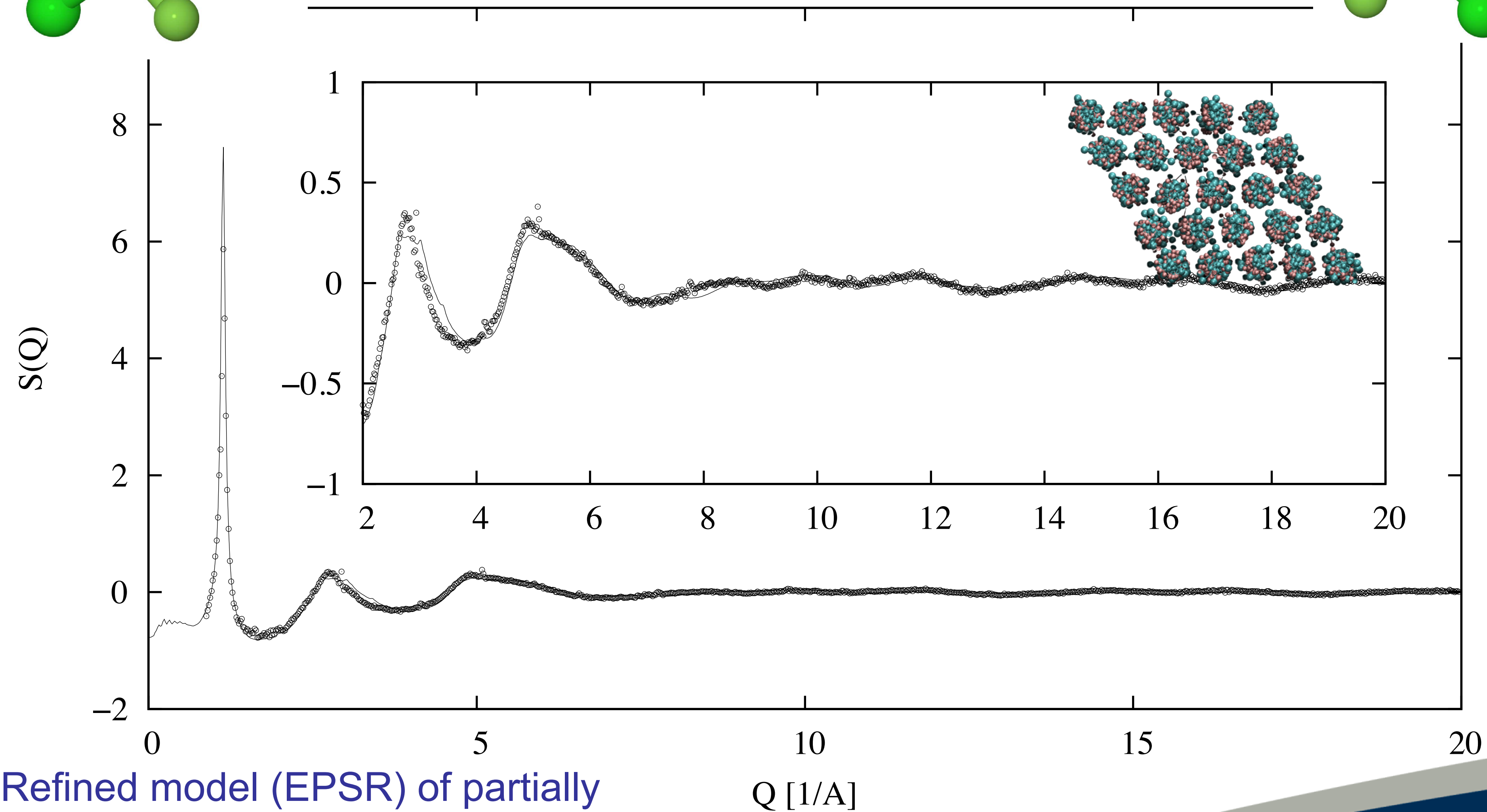
A.K.Soper, K.Page and A.Llobet,
J. Phys.: Condens. Matter **25** *454219* (2013)



KelF (PCTFE)

Data from LANSCE: HIPD & NPDF





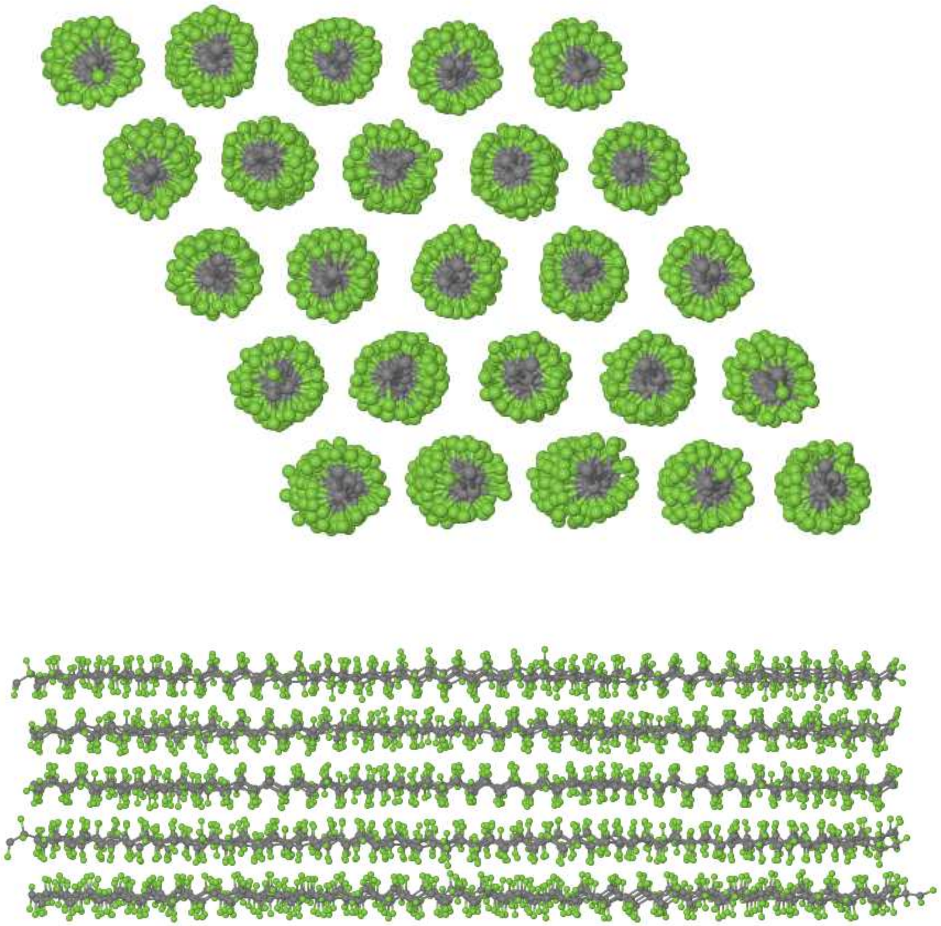
Refined model (EPSR) of partially crystalline PCTFE with both diffuse and Bragg scattering features

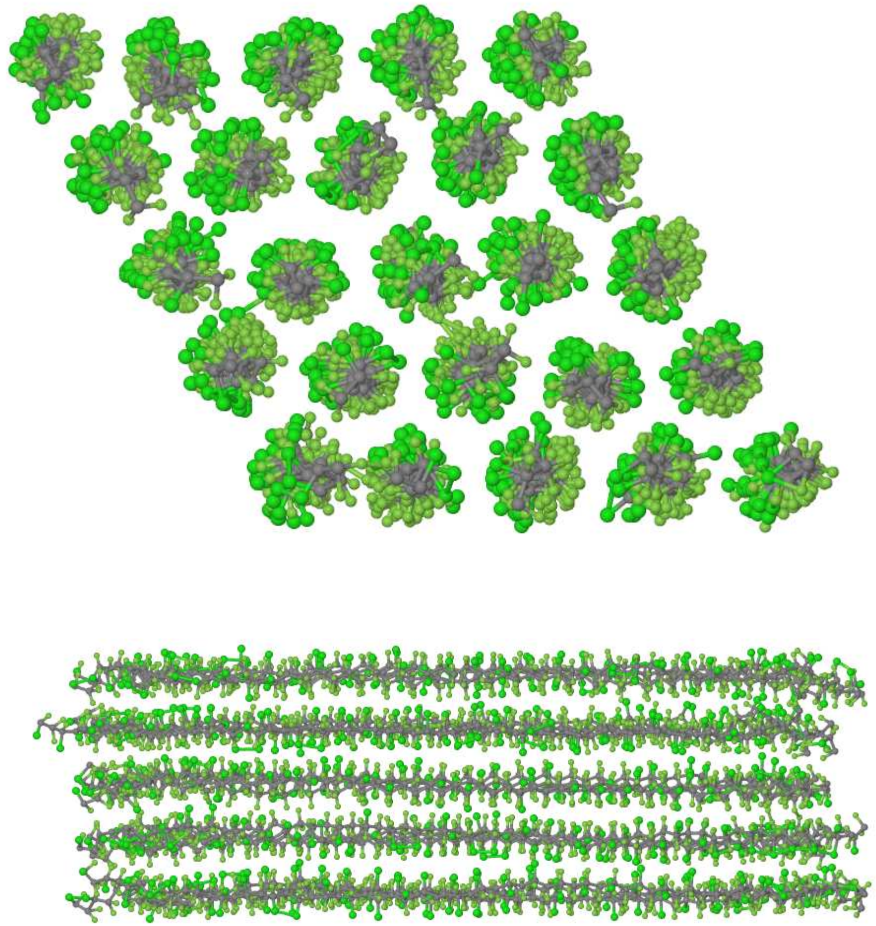
A.K.Soper, K.Page and A.Llobet,
J. Phys.: Condens. Matter **25** *454219* (2013)

Science & Technology Facilities Council
ISIS

PTFE

PCTFE





A.K.Soper, K.Page and A.Llobet,
J. Phys.: Condens. Matter **25** *454219* (2013)

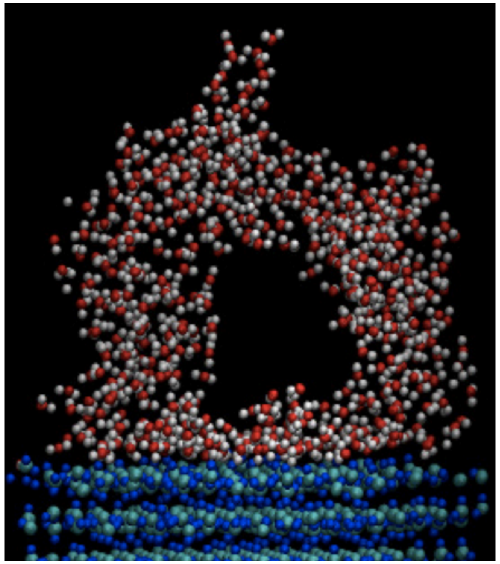
Science & Technology Facilities Council
ISIS


Interstellar Ice

Ice is thought to play an major role in astrophysical processes such as star formation

Credit: NASA

Ice in space forms as a porous amorphous solid and plays a role in promoting chemical reactions whilst also acting as a reservoir for trapping volatile gases

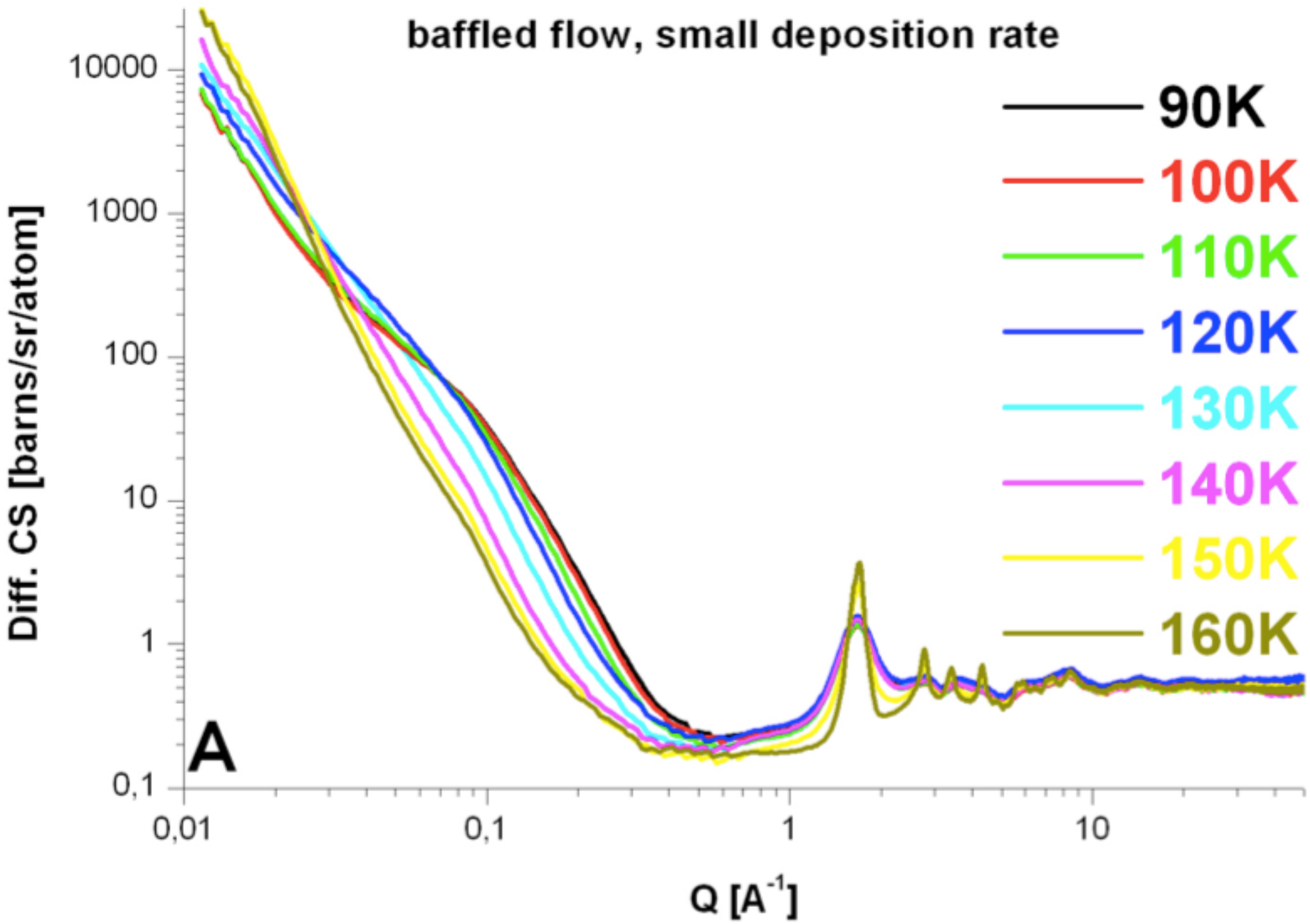


Credit: Helen Fraser

The collapse of interstellar gas clouds to form stars is governed by a balance between gravity and heat. Coolant gases moderate the kinetic effects of heat, via radiation, and act to promote cloud collapse. Porous ice is believed to help keep these gases present for longer periods of time.

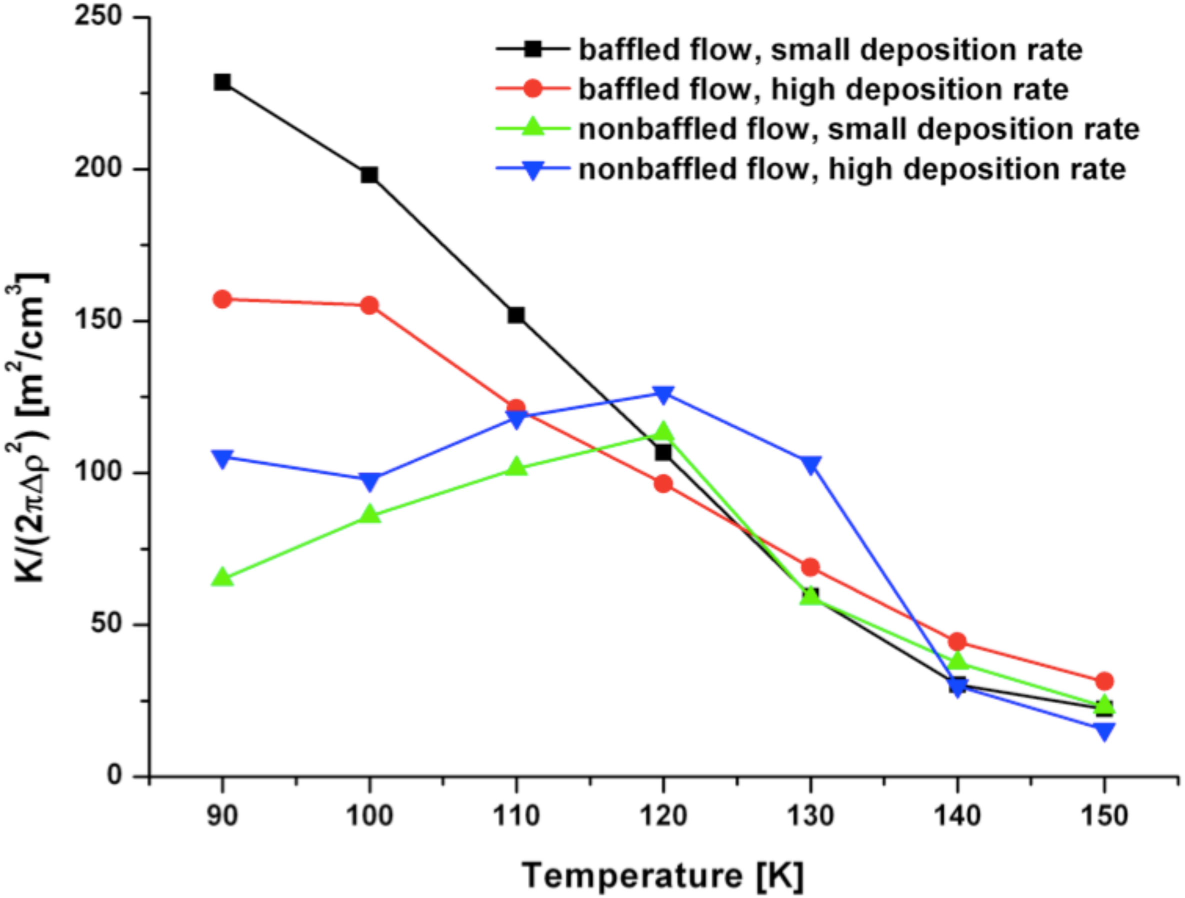
It is important to know how pores in amorphous ice behave as a function of temperature.

Neutron scattering from amorphous ice as it is warmed to 160K



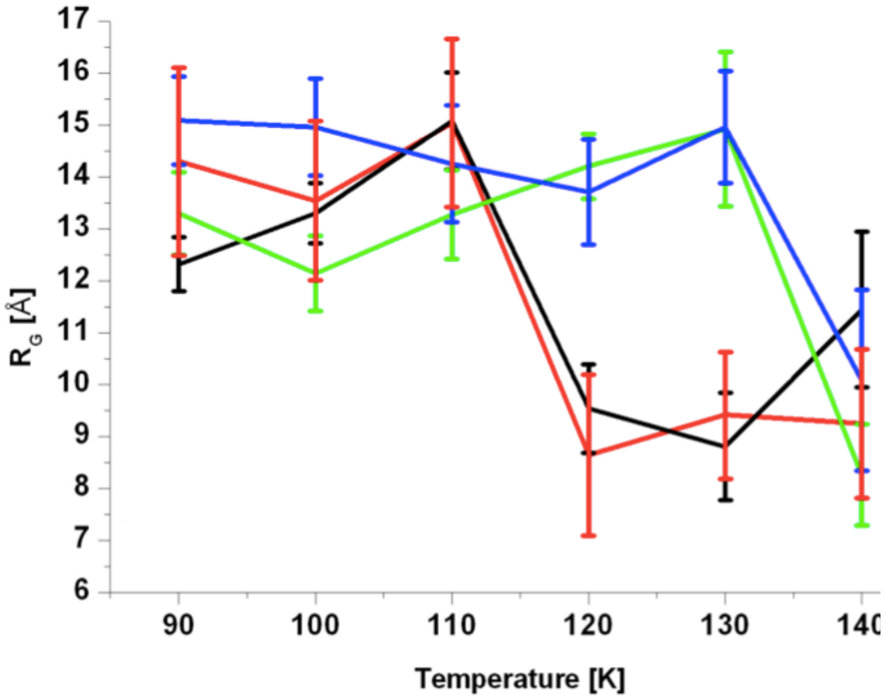
C.Mitterdorfer, M.Bauer, T.G.A.Youngs, D.T. Bowron, H.J.Fraser, J.L.Finney and T.Loerting PCCP **16** 16013 (2014)

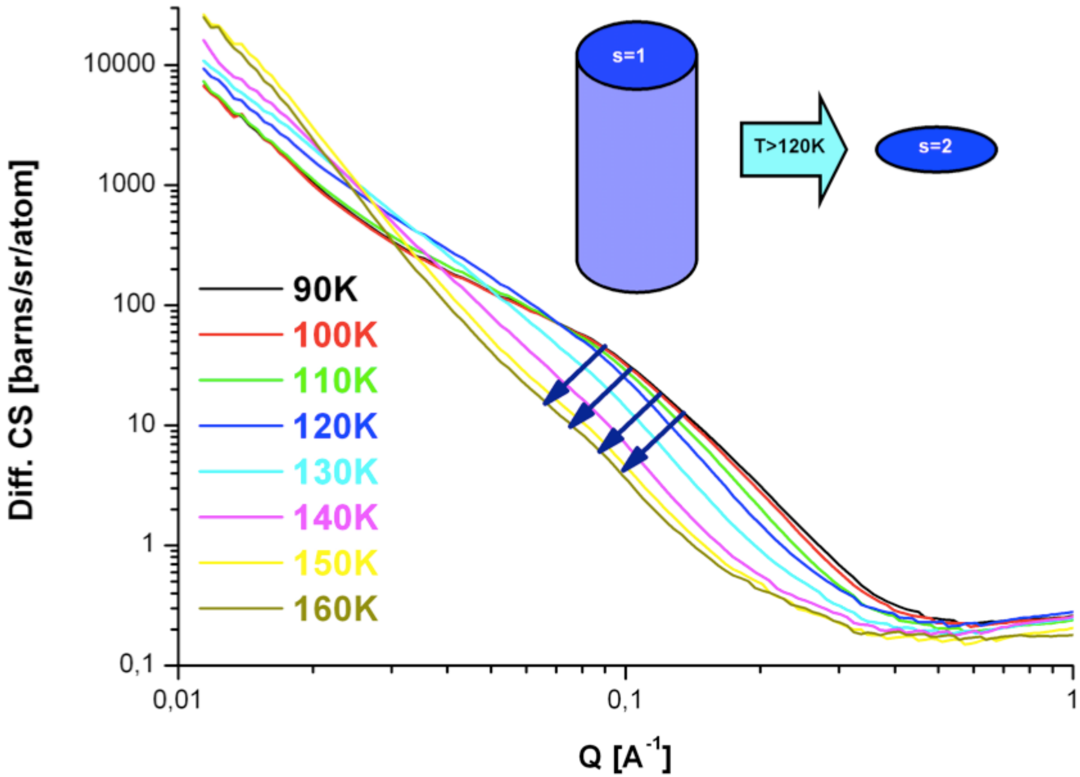
Pore collapse is characterized by a continuous decrease in surface area



C.Mitterdorfer, M.Bauer, T.G.A.Youngs, D.T. Bowron, H.J.Fraser, J.L.Finney and T.Loerting PCCP **16** 16013 (2014)

Pore collapse rapidly transitions from a three dimensional pore to a two dimensional pore of smaller radius of gyration

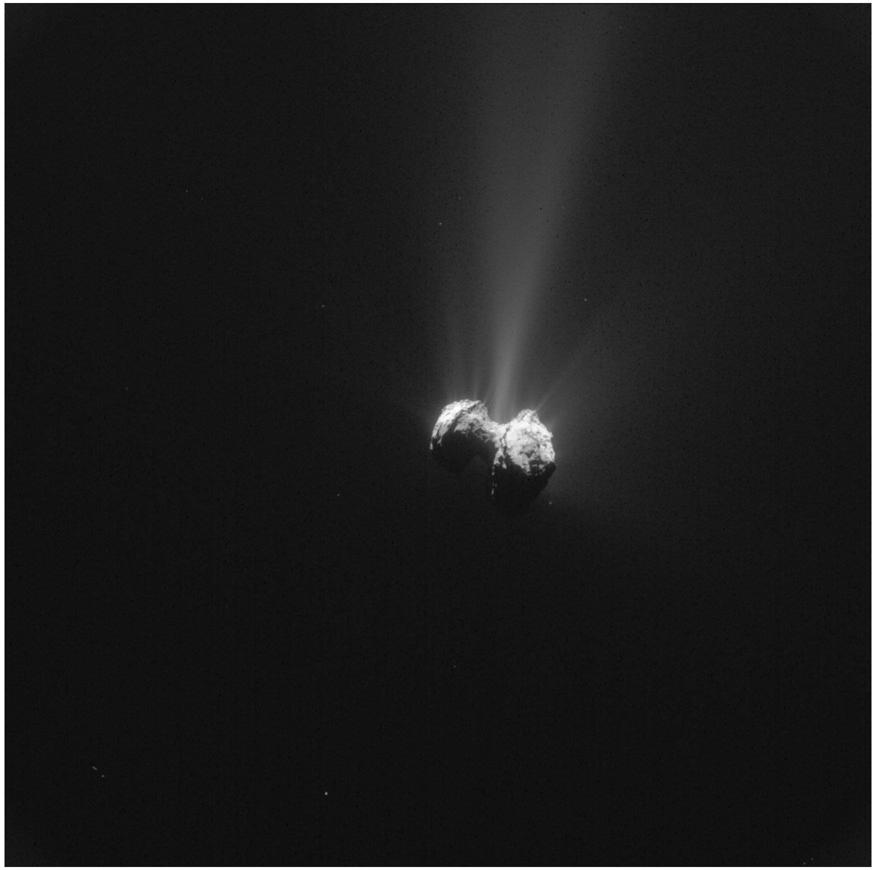


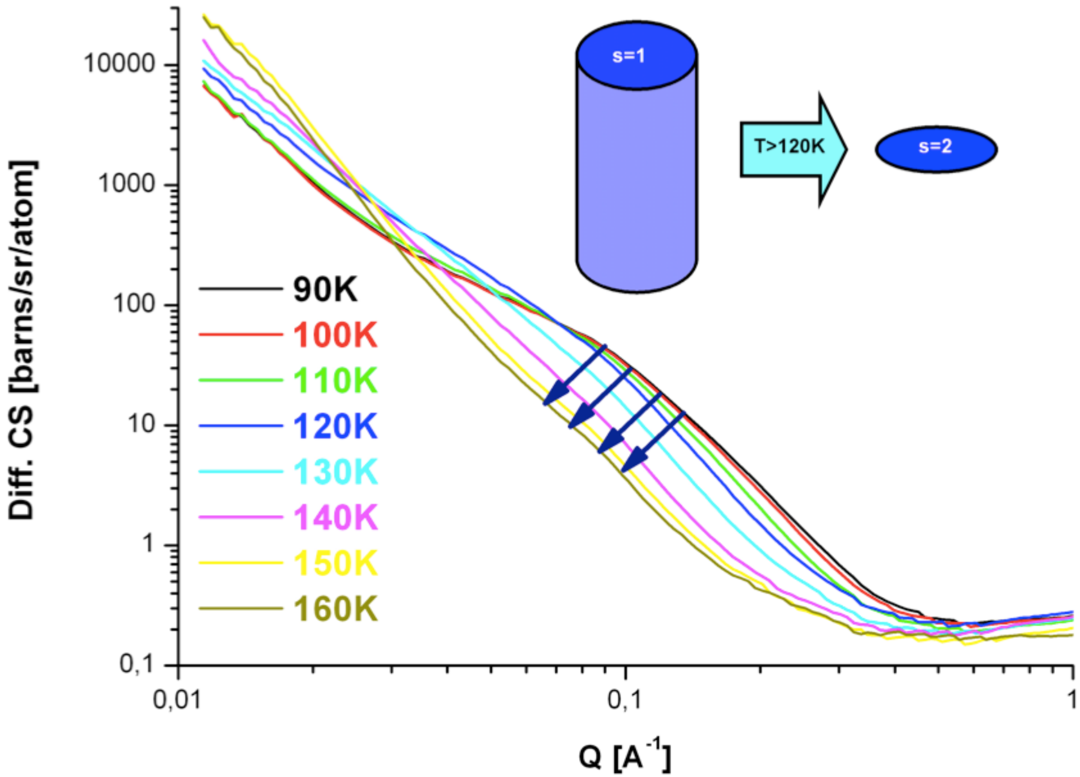


C.Mitterdorfer, M.Bauer, T.G.A.Youngs, D.T. Bowron, H.J.Fraser, J.L.Finney and T.Loerting PCCP **16** 16013 (2014)

Science & Technology Facilities Council
ISIS

Pore collapse rapidly transitions from a three dimensional pore to a two dimensional pore of smaller radius of gyration

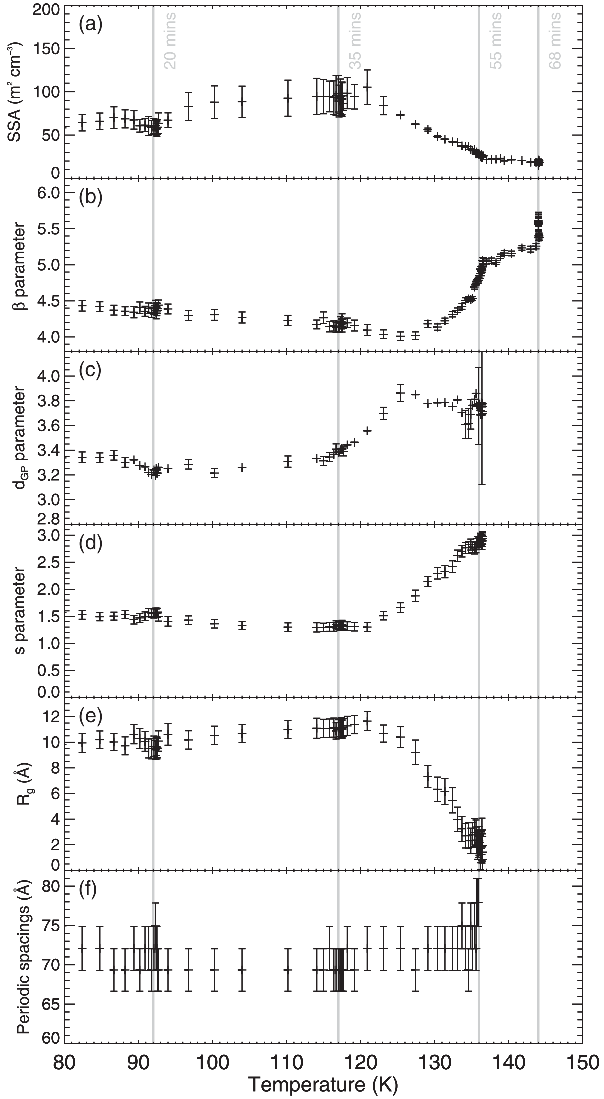




Rosetta: Comet 67P/Churyumov-Gerasimenko
European Space Agency

Science & Technology Facilities Council
ISIS

Kinetic studies show that diffusive motion in porous ASW starts at 121K



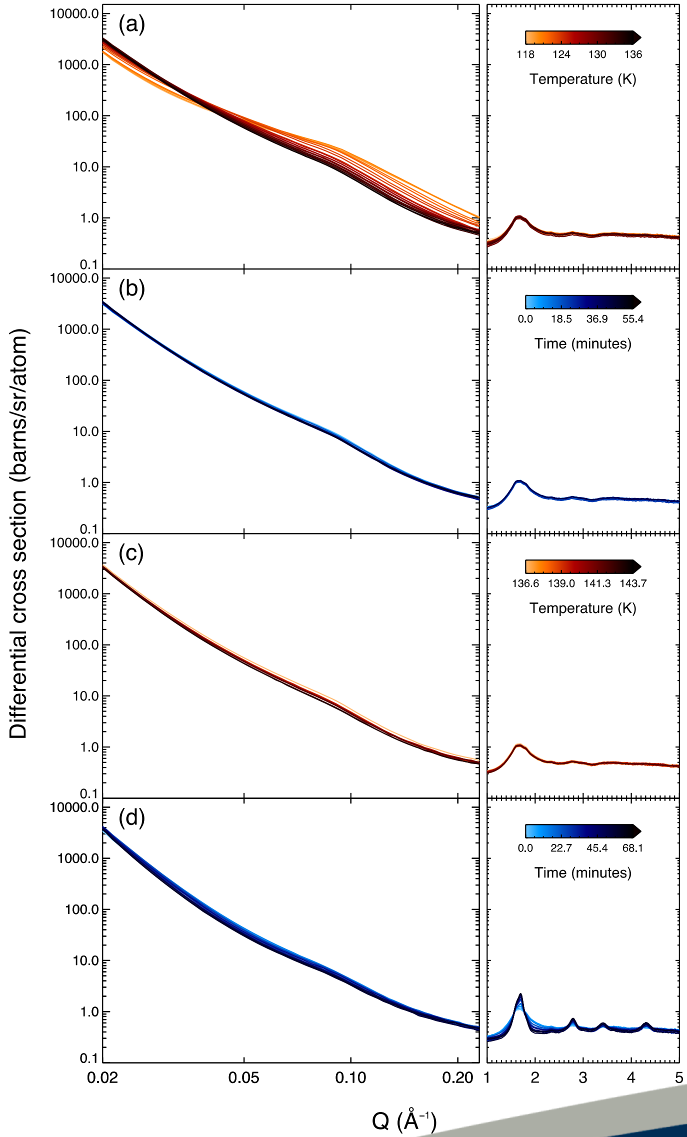
Heating @ 0.4 $Kmin^{-1}$

$T_{non\text{-}trans\ start}$ = 115K

$T_{g,start}$ = 121K

$T_{g,end}$ = 136K

T_X = 141K



C.R.Hill, C.Mitterdorfer, T.G.A.Youngs, D.T.Bowron, H.J.Fraser and T.Loerting
Phys. Rev. Lett. **116** 215501 (2016)

Science & Technology Facilities Council
ISIS

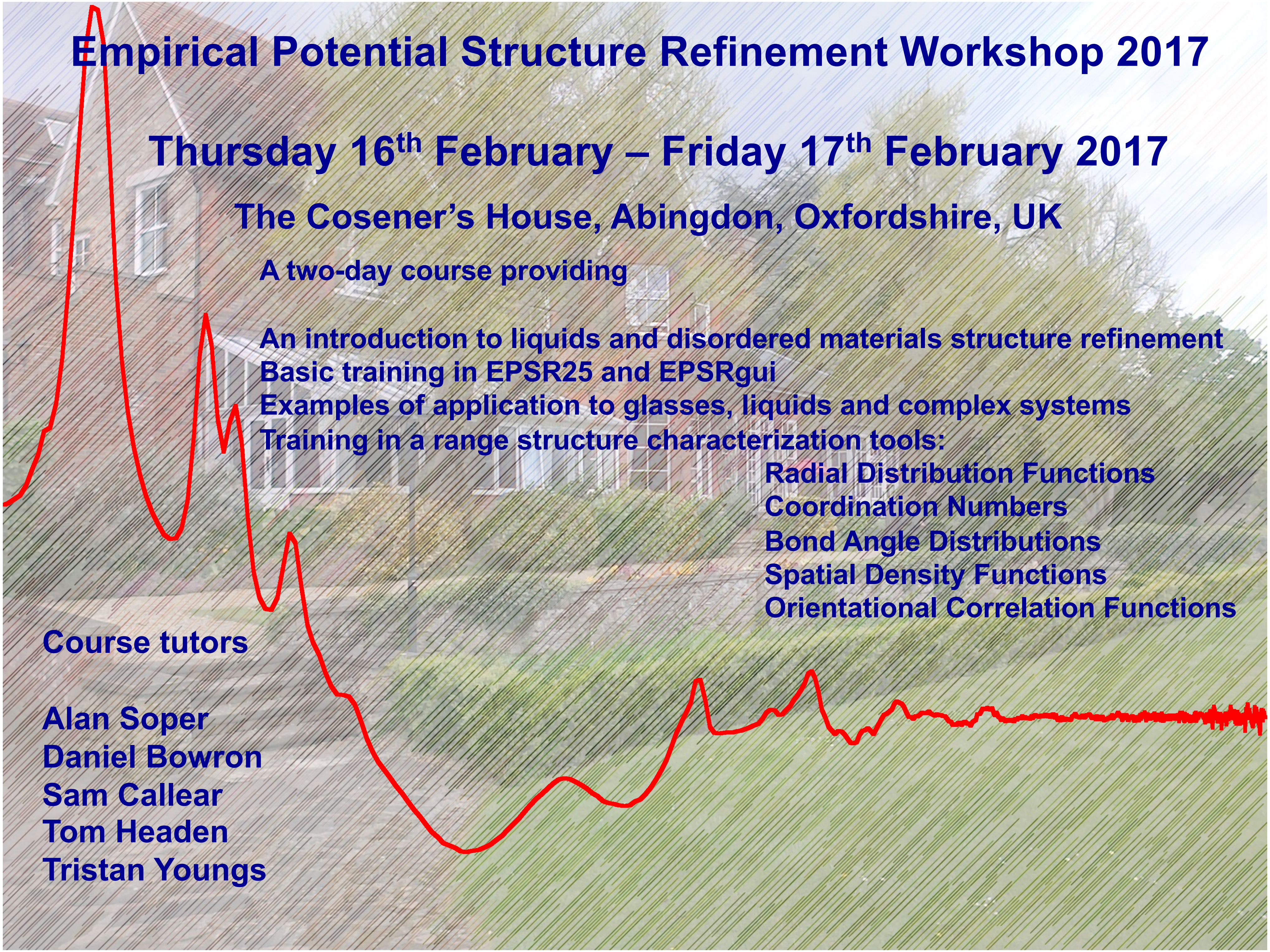
Summary: Issues we will aim to highlight in this workshop

Issue 1: The diversity of science that can be probed using neutron total scattering and PDF methods

Issue 2: Key aspects of the instrumentation that is required to make the measurements

Issue 3: The analysis methodologies that are required to extract the scientific information in the measured data.

Issue 4: Structure refinement of liquid and disordered materials data using atomistic models


Empirical Potential Structure Refinement Workshop 2017
Thursday 16th February – Friday 17th February 2017
The Cosener's House, Abingdon, Oxfordshire, UK
A two-day course providing
An introduction to liquids and disordered materials structure refinement
Basic training in EPSR25 and EPSRgui
Examples of application to glasses, liquids and complex systems
Training in a range structure characterization tools:
Radial Distribution Functions
Coordination Numbers
Bond Angle Distributions
Spatial Density Functions
Orientational Correlation Functions
Course tutors
Alan Soper
Daniel Bowron
Sam Callear
Tom Headen
Tristan Youngs