Experimental Report

Proposal: **6-02-519** **Council:** 10/2012

Title: A novel approach to the structure of 1H-containing liquids: the case of liquid water

This proposal is continuation of: 6-02-362

Researh Area: Chemistry

Main proposer: **TEMLEITNER Laszlo**

Experimental Team: TEMLEITNER Laszlo

Local Contact: STUNAULT Anne

Samples:
D2O
(D2O)0.6(H2O)0.4
(D2O)0.33(H2O)0.67
H2O
(D2O)0.8(H2O)0.2

Instrument	Req. Days	All. Days	From	To
D3 CPA	8	10	28/06/2013	08/07/2013

Abstract:

The primary difficulty with the determination of the structure of liquid water is the huge incoherent inelastic scattering that arises due to the exceptionally high level of spin-incoherency of 1H. As a result of that, more than 95 % of the measured diffraction (using non-polarized neutron beam) signal from pure H2O is useless ('background') from the structural point of view. Spin-incoherence, however, can be bypassed if the neutron beam is polarized; the structure factor of even pure H2O can then be determined, without handling a 'background' which is 20 times larger than the desired coherent scattering. Using the D3 instrument, it would be possible to measure accurate (coherent) static structure factors of liquid water samples, containing a varying proportion of 1H, over a sufficiently wide Q-range. Using the Reverse Monte Carlo technique would make it possible to combine these data sets

A novel approach, using polarized neutrons, to the structure of ^1H-containing liquids : the case of liquid water

Abstract

Polarized neutrons over a wide Q-range allowed the separation of coherent and incoherent scattering contributions from ambient water. During the allocated beamtime, the following samples were measured: 100%, 80%, 60%, 35.92% D_2O and pure H_2O.

Understanding the structure of water has a great importance for sciences, industry and many aspects of life itself. The primary difficulty with the determination of the structure of liquid water is the huge incoherent scattering that arises due to the exceptionally high level of spin-incoherency of ^1H. As a result of that, more than 95 % of the measured diffraction signal (using non-polarized neutron beam) from pure H_2O is useless ('background') from the structural point of view. Spin-incoherence, however, can be bypassed if the neutron beam is polarized; the structure factor of even pure H_2O can then be determined, without handling a 'background' which is 20 times larger than the desired coherent scattering.

The measurement were performed on the polarised neutron instrument *D3*, at a wavelength of 0.52Å, using a Hf filter to supress higher order harmonics. With this short wavelength we could explore a rather large Q-range, up to 20 $Å^{-1}$. The high instrument background that had hampered previous test measurements was reduced to less than 1 count/s, due to some drastic improvements in the shielding. The geometry of the sample holder was a custom made 57.4 mm long hollow cylinder, with 8 mm internal and 10.7 outer diameter (wall thickness: 0.15 mm), sealed at bottom end by indium. At the top of the sample holder, a cylindrical shaped single crystal has been mounted to monitor the ^3He spin filter analysing power. This measurement could be performed by elevating the sample holder along the 'z' axis and showed filter efficiencies between 60% and 75% (relaxation time of the spin filter of the order of 80-90 hours). A Helmholtz-coil provided a weak magnetic field aimed at to preserve the polarisation of the neutron beam before and after the sample. The measurements were performed by scanning the spin-filter - detector assembly over the whole Q-range.

Great care was taken in adjusting the beam size to ensure simultaneously high flux, high level of beam polarisation and homogeneity of the both accross the sample volume. These checks have been performed by a hollow Si-wafer (homogenity), an empty cell and D_2O sample (background and intensity). Moreover, we prepared identical size sample holders made from vanadium and aluminium. Both materials have advantages and inconveniences: aluminium

provides low background (except the regions of Bragg-peaks) due to its small incoherent scattering cross-section, but the reproducibility of the intensity of the coherent (Bragg) scattering was bad due to its texture. In contrast, vanadium provides higher background for both channels but there are very small Bragg-peaks only. Eventually, the vanadium cell looked more favourable for the actual conditions.

During the remaining beamtime, we performed measurements on different isotopic mixtures of D_2O and H_2O: pure, 80%, 60%, 35.92% D_2O, pure H_2O, and on the empty cell.

The raw measured datasets were corrected for the time-dependent analyser efficiency, the absorption of the empty analyser, and were transformed into coherent and incoherent intensities. The obtained datasets were corrected by sample holder intensities.

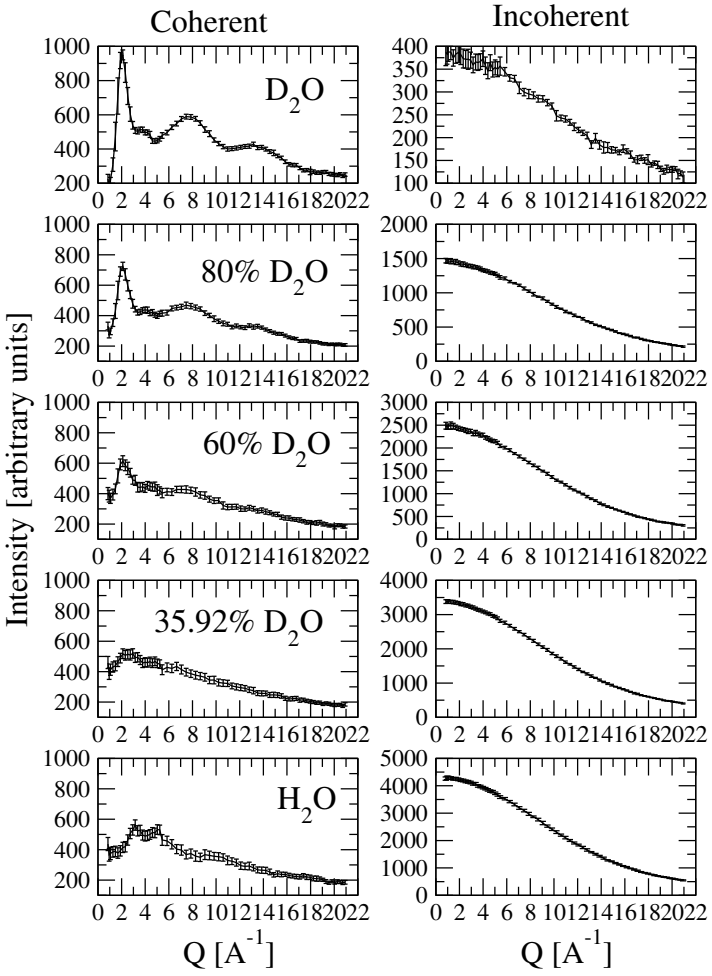

Figure 1: Experimental coherent and incoherent intensities of ambient liquid water.