Experimental Report

19/06/2015

Proposal: **6-02-536** **Council:** 4/2014

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

This proposal is continuation of: 6-02-519

Researh Area: Chemistry

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Local Contact: STUNAULT Anne

Samples: 60%D2O 40%H2O
35.92% D2O 64.08% H2O
H2O

Instrument	Req. Days	All. Days	From	To
D3 CPA	8	8	12/12/2014	20/12/2014

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 and construct structural models that would be consistent with measured data. The outcome would be an unquestionable set of partial pair correlation functions for the most important liquid - water. The present proposal aims at concluding a series of experiments that originally started in 2006.

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

6-02-536

Abstract

Polarized neutrons over a wide Q-range allowed the separation of coherent and incoherent scattering contributions from ambient water. During the present experiment, statistically more accurate datasets were obtained in comparison with the previous measurement (proposal no. 6-02-519). The following H_2O/D_2O mixtures have been measured currently: 100%, 60%, 35.92% D_2O and pure H_2O.

Understanding the structure of water has a great importance for science, 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 ^{1}H. 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 measurements 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 geometry of the sample holder was a custom made 57.4 mm long hollow cylinder, with 8 mm internal and 10.7 mm outer diameter (wall thickness: 0.15 mm), sealed at the bottom end by indium. At the top of the sample holder, a cylindrical shaped single crystal was mounted to monitor the ^{3}He spin filter analysing power. This auxiliary measurement could be performed by elevating the sample holder along the 'z' axis. Filter efficiencies between 60% and 75% have been recorded (the relaxation time of the spin filter is of the order of 80-90 hours). A Helmholtz-coil provided the necessary weak magnetic field for the preservation of the polarisation of the neutron beam before and after the sample. The measurements were performed by scanning the entire scattering angle (and therefore, Q-)range by the spin-filter - detector assembly.

Last time (proposal number 6-02-519), achievements similar to the ones of this project have been attempted; unfortunately, finding the ideal setup took much more time than expected. Thus, only limited time remained for the samples and sufficiently good statistics for coherent scattering parts could not be reached. On the other hand, the incoherent scattering part could be measured perfectly well: it was revealed that it can be approximated empirically by a Gaussian term (plus a constant) [arXiv:1410.0141v1, accepted in PRB].

During the allocated beamtime, we performed measurements on different isotopic mixtures of D_2O and H_2O: 100%, 60%, 35.92% D_2O, and pure H_2O (plus one run on the empty cell). The measurement times have been optimized to the coherent scattering contributions. The raw measured datasets were corrected for the time-dependent analyser efficiency and for the absorption of the empty analyser, and were transformed into coherent and incoherent intensities. The obtained datasets were corrected by sample holder intensities.

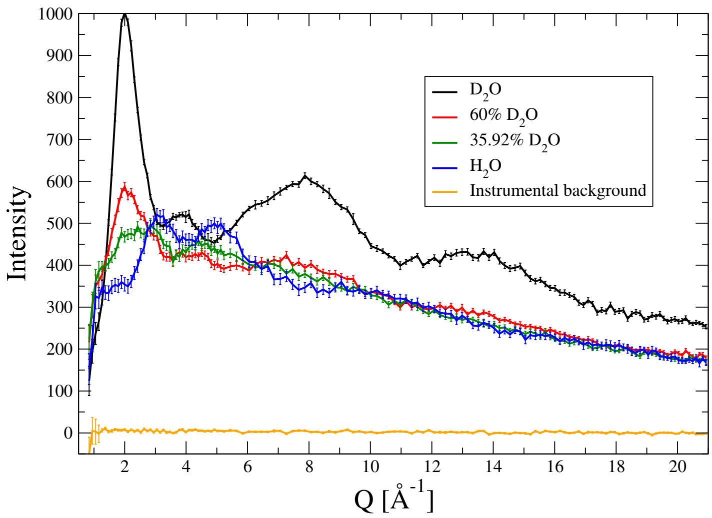

Figure 1: Experimental coherent intensities of ambient liquid water.