Experimental report

15/09/2016

Proposal: 6-02-543 **Council:** 4/2014

Title: Separation of coherent and incoherent differential cross section of two isomers of propanol using polarized hot neutrons

Research area: Physics

This proposal is a new proposal

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Samples: C3D8O

Instrument	Requested days	Allocated days	From	To
D3	6	5	05/12/2014	10/12/2014

Abstract:

The aim of this experiment is the separation of the coherent and incoherent parts of the differential cross section of propanol (two isomers: 1- and 2-propanol) in the liquid state. The experiment will use the unique characteristics provided by polarised neutrons on D3 (hot neutrons) just to observe the small differences in the structure factor due to isotopic effects. So far, all experiments have been done with deuterated samples and the isotopic effect could not be observed. The experiment requires 6 days of beamtime on D3.

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Structure factor determination of deuterated 1- and 2-propanol using diffraction experiments with polarization analysis

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We present the experimental structure factors of deuterated 1- and 2-propanol as determined at the Spin Polarized Hot Neutron Beam Facility (D3) of the Institut Laue Langevin. Polarized neutron scattering with polarization analysis has the advantage of experimentally separating the coherent and incoherent scattering intensities. Using a linear combination of non-spin-flip and spin-flip diffractograms, one can determine the coherent intensity, related to the structure factor. The corrections of experimental data for multiple scattering, attenuation and inelasticity are carried out using a Monte Carlo (MC) simulation code developed for this kind of experiments. This (hybrid) MC method is based on the combination of a modelled energy exchange and the experimental angular distribution. The good agreement observed between our simulations and the experimental results, confirms the goodness of this model. We also compare our results with experimental data from other authors and we stress the need of more experimental data in the low-Q region.

Keywords: liquids; Monte Carlo; neutron diffraction; polarization; structure

1. Introduction

In hydrogen-containing molecules, hydrogen often plays a key role, since many properties are derived directly from the inter-molecular hydrogen bond. For such molecules, neutron scattering, with high scattering contrast is ideally suited to study the structural and dynamical properties. However, due to the additional complexity of the enormous incoherent signal from hydrogen, most studies have, up to now, been performed on deuterated samples.

Using polarized neutron diffraction with polarization analysis, one can almost directly measure the incoherent cross section [1]: since the neutron has a spin 1/2, it is sensitive to the nuclear spin, giving different scattering intensities, whether the nuclear spins are parallel or anti-parallel to the neutron polarization. This is the source of the (spin) incoherent scattering observed for hydrogen, but also the basis of the method proposed here. Experimentally, using a set of polarizers/analysers, one can measure separately the scattered intensities with polarization parallel (non-spin-flip, NSF) or anti-parallel (spin-flip, SF) to the primary beam polarization (see Section 2 for details). Simple linear relationships then lead from

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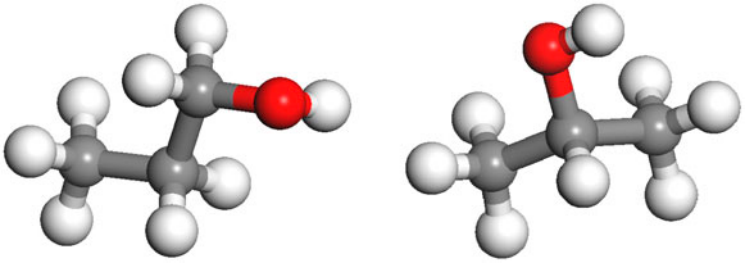

1-propanol **2-propanol**

Figure 1. (colour online) Molecular structure of the 1- and 2-propanol, $CD_3CD_2CD_2OD$ and $CD_3CD(OD)CD_3$, respectively. Red (dark grey in b/w version), light grey and grey spheres are O, D and C, respectively.

the measured NSF/SF intensities to the incoherent and coherent scattering cross sections (Section 5).

The traditional [2–4] or new empirical [1] methods for correcting for background, attenuation, inelasticity and multiple scattering treat all these contributions independently and not as a whole. In Sections 3 and 4, we propose a consistent treatment of all corrections, based on a Monte Carlo (MC) simulation of the neutron–sample interaction (Section 4). Compared to traditional [2–4] or new empirical [1] methods, this one should represent a step forward, towards more accurate data corrections.

In the past, the structures of the liquid phases of the lowest members in the alcohol series (methanol, ethanol and iso-propanol) have been determined by neutron diffraction [5–11], mostly from deuterated samples. As a first test, we study 1- and 2-propanol, which are rather simple molecules with a high proportion of hydrogen and ideally suited to develop and test the method. In this work, we use deuterated 1- and 2-propanol, $CD_3CD_2CD_2OD$ and $CD_3CD(OD)CD_3$ (Figure 1). The corrections are expected to be small, due to much reduced incoherent contribution, and our results can be easily compared to previous, unpolarized work (Section 5).

Finally, in Section 6, we make a careful analysis of the obtained structure factors, to gain some insight into the structures of both molecules. In particular, in the case of 2-propanol, we compare our results with those from other authors. This comparison leaves some open questions about the structure factor at low-Q, that should be resolved with new experimental evidence.

2. Experimental

The experiment was performed at the Spin Polarized Hot Neutron Beam Facility (D3) of the ILL (Institut Laue Langevin, Grenoble, France). A detailed description of the diffractometer can be found in Refs. [13,14]. The unpolarized neutron beam is monochromated and polarized using a Heusler crystal Cu_2MnAl ((1 1 1) Bragg reflection). Guide field along the beam path preserve the polarization, while nutators before and after the sample can reverse (flip) it. The neutrons scattered by the sample are analysed using a polarized ^{3}He spin filter [15,16] and the transmitted neutrons are collected in a single ^{3}He detector scanned over an

angular range $2\theta = 4° - 120°$. With a wavelength of 0.52 Å, the NSF and SF diffractograms are recorded for a momentum transfer of 0.8–21 Å^{-1} ($Q = \frac{4\pi}{\lambda} \sin\theta$).

The samples of fully deuterated 1-propanol (98%at. D) and 2-propanol (99%at. D) were bought from Isotec Inc. and Aldrich Chemical Co, respectively, and the experiments were performed in the liquid state at room temperature. The macroscopic densities (ρ) and isothermal compressibilities (χ_T) are: 0.912 g/cm^3 (or 0.00806 at./Å^3) [17] and 1.21×10^{-9} Pa^{-1} [11] for deuterated 1-propanol, and 0.890 g/cm^3 (or 0.00787 at./Å^3) [17] and 1.12×10^{-9} Pa^{-1} [11] for deuterated 2-propanol. The densities will be used in the normalization process to obtain the structure factor on an absolute scale. The samples were loaded into a cylindrical double-walled vanadium cell (hollow cylinder) of 10.7 mm external diameter, 7.7 mm internal diameter, 0.15 mm walls thickness and 58 mm height, giving a sample thickness of 1.2 mm and a volume of 0.829 cm^3 in the beam. The mass was checked before and after the experiment, confirming that the container had no leakage.

Measurements of NSF and SF diffractograms were carried out for a period of 24 h for each sample. The signal from the empty instrument, the empty vanadium container and a vanadium rod (10 mm diameter by 60 mm height) were measured in shorter times. Short measurements were periodically performed on a Ge single crystal placed at the sample position [14] to monitor the analysing power of the ^{3}He spin filter. All diffractograms were corrected for the absorption by the empty ^{3}He filter and normalized to the monitor counts. Figure 2 shows the NSF/SF diffractograms for 1-propanol after correcting for the spin-filter efficiency: empty instrument background $I^{\mathrm{B}}_{\mathrm{exp}}(Q)^{\mathrm{NSF,SF}}$, sample container (+background) $I^{\mathrm{CB}}_{\mathrm{exp}}(Q)^{\mathrm{NSF,SF}}$ and sample (+container+background) $I^{\mathrm{SCB}}_{\mathrm{exp}}(Q)^{\mathrm{NSF,SF}}$. In the low-$Q$ range, the background subtraction is a delicate issue because the empty instrument signal becomes comparable to those of the sample and the container.

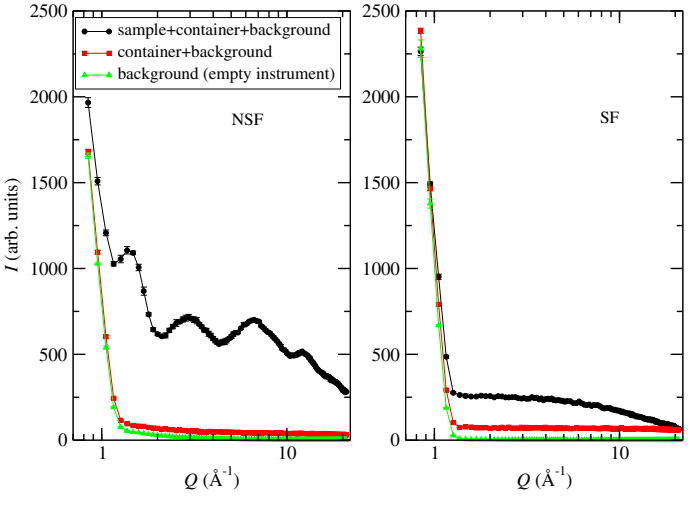

Figure 2. (colour online) Experimental diffractograms (NSF/SF) for deuterated 1-propanol, empty container and background. The logarithmic scale in the abscissa emphasizes the low-Q region.