Experimental Report

17/02/2015

Proposal: **9-13-522** **Council:** 4/2014

Title: Pressure dependence of MSD in supercooled confined water: a new way of testing the Liquid-Liquid Phase Transition hypothesis

This proposal is a new proposal

Researh Area: Soft condensed matter

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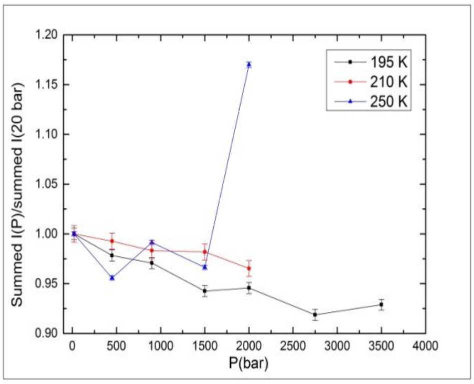
Samples: H_2O in SiO_2

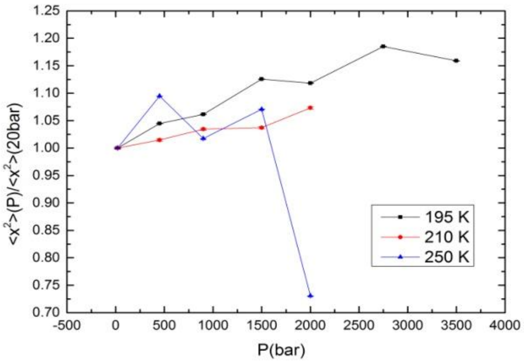
Instrument	Req. Days	All. Days	From	To
IN13	8	8	08/12/2014	16/12/2014

Abstract:

Aim of this proposal is to verify the Liquid-Liquid Phase Transition (LLPT) hypothesis in supercooled confined water. The LLPT hypothesis has been recently proposed in the literature and has given rise to passionate debate; it has been suggested to be present in the hydration water of proteins and to trigger the so called protein dynamical transition. The novelty of our approach is in the use of Elastic Incoherent Neutron Scattering (EINS) to measure the pressure dependence of mean square displacements (MSD) at constant temperature, across the putative LLPT. Moreover, we investigate water confined in the pores of a hydrophilic disordered silica matrix obtained from the alcoxide precursor TMOS via the sol-gel method. In this new matrix the confinement conditions experienced by water in biological systems are closely mimicked and the hydration level can be easily varied.

Proposal 9-13-522 received 8 days beamtime on IN13, the experiments have been performed in December 2014, and a preliminary analysis of the data gave very interesting and promising results reported in the figures below.





The left panel reports the elastic intensities summed over the Q values in the range 0.17÷1.37 A^{-1} (SUM I) as a function of pressure at 250, 210 and 195K; SUM I values have been normalized to their atmospheric pressure values. To proceed, we exploit the well known relation, valid within the Gaussian approximation: SUM I = K·(MSD)$^{-1/2}$, where K is a constant related to the Q interval involved in the summation. For our analysis we used the 0.17÷1.37 A^{-1} interval where the Gaussian approximation holds. The advantage of this procedure over the more commonly used [MSD≈-6dlnS(Q,ω=0)/dQ^2] is that it reduces substantially the error bars. MSD obtained with this procedure are reported in the right panel as a function of the pressure; normalization to the atmospheric pressure values eliminates the constant K. On the other hand, given the relation MSD(t)=6Dt, where t = t$_{res}$ ≈ 100 ps for IN13, the data reported in the right panel give the pressure dependence of the diffusion coefficient (for a more in depth discussion, see Cupane et al., PRL, 2014). The results are, in our opinion, quite interesting. At low pressures, an increase of the diffusion coefficient is observed, the effect being more relevant at 250K (this is a manifestation of the so called “pressure anomaly” of supercooled water and is related to the breaking by pressure of the hydrogen bonds network); however, as the pressure is increased, the diffusion coefficients tend to saturate and, at 250K, a much lower value is observed when the pressure is increased up to 2000 bar. This is an interesting result since it shows that at sufficiently high pressures supercooled water (which at 250K is predominantly in the high density state) recovers a “normal” behavior. Is this true also at lower temperatures, where supercooled water is predominantly in the high density state? Although a saturating behavior seems to be present, the actual data do not allow to give an answer to this question, since the pressure range investigated is not ample enough.