ESRF	**Experiment title:** Relaxational dynamics of supercritical water	**Experiment number**: HS-2606
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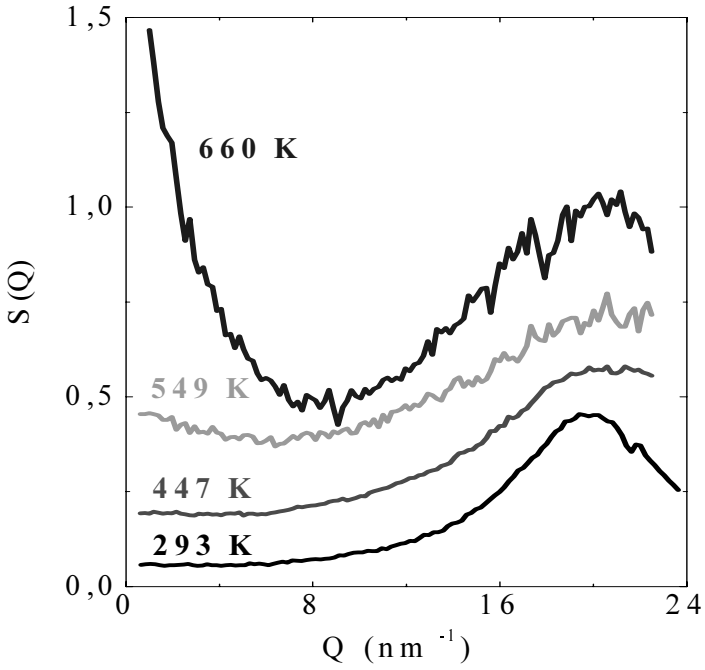
Report:

The high-frequency dynamics of liquid and supercritical water has been studied by inelastic x-ray scattering (IXS). The experiment was performed at an incident photon energy of 21.747 KeV, using the silicon (11,11,11) set-up, providing an overall energy resolution of 1.5 meV (FWHM). The sample has been embedded in a specially designed large volume (~0.7 cm^3) high-pressure, high-temperature cell made out of Inconel. The experiment was performed in isobaric (400 bar) conditions varying the temperature of the sample from 293 K up to 706 K, above the critical point of water (T_c=647 K and P_c=221 bar). The investigated temperatures were: 293, 337, 423, 447, 549, 660 and 706 K. For each thermodynamic point, we acquired one diffraction pattern and 10 IXS spectra in a momentum transfer range between 2 and 15.5 nm^{-1}. 2 or 3 spectra with an integration time of 4h each were accumulated in order to have sufficient statistics. The contribution of the empty cell was measured as well and yielded a negligible contribution to the signal.

In figure 1 we report some selected diffraction patterns, S(Q). The intensity of the first sharp diffraction peak decreases while approaching the critical temperature (647 K) while at low-Q a large increase of the scattering due to the insurgence of critical fluctuations is observed [1]. Figure 2 shows representative IXS spectra, together with their best fit result. This fit was obtained using a model based on a viscoelastic function taking into account one structural relaxation process, the thermal diffusion and a "microscopic" term (for further details see [2-4]). This model function has been convoluted with the experimentally determined resolution function.

Figures 3 and 4 report the main results concerning the structural relaxation: its characteristic timescale τ and strength, Δ. While τ shows an almost linear Q-dependence, Δ is constant throughout the explored Q range. Both quantities decrease with increasing temperature. From the viscoelastic analysis, we can also derive the value of the isothermal and infinite frequency sound dispersion, $\Omega_0(Q)$ and $\Omega_{inf}(Q)$. Alternatively, $\Omega_0(Q)$ can also be derived from the S(Q) measurements, and the values derived by the two approaches are in excellent agreement. Finally, from the maximum of the longitudinal current spectrum, $\omega^2 S(Q,\omega)$, we can derive the apparent sound dispersion, $\Omega_L(Q)$, that represents the characteristic frequency of sound propagation [2-4].



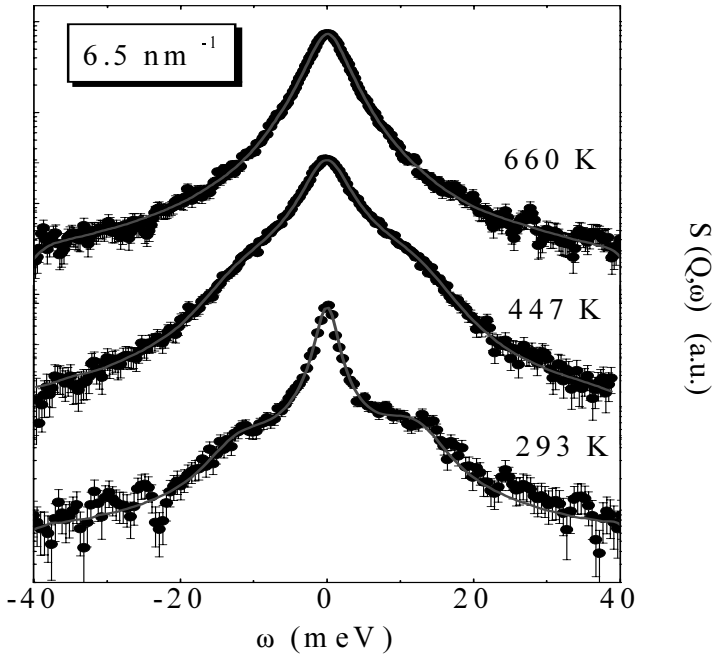
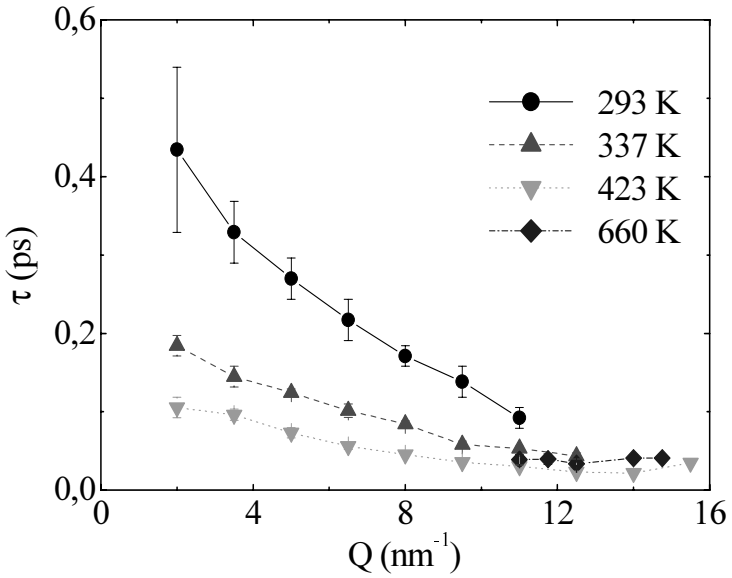


Fig. 1 (left): Diffraction patterns of liquid water at P = 400bar and the indicated temperatures. See text for more details.
Fig. 2 (right): IXS spectra of liquid water at Q=6.5 nm^{-1} at 400bar and the indicated temperatures. The experimental data (circles) and their error bars are shown together with the best fit (red full line). See text for further details.



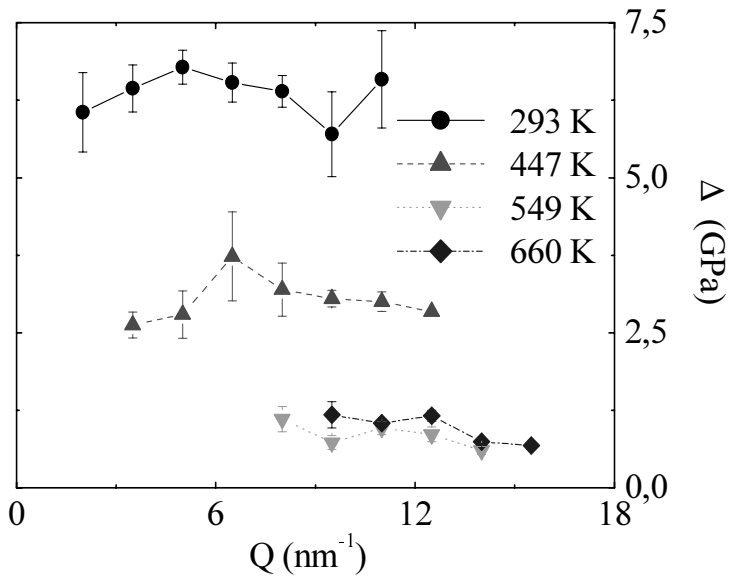


Fig. 3 (left): Characteristic time τ of the structural relaxation.
Fig. 4 (right): Strength, Δ, of the structural relaxation.

References

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