ESRF	**Experiment title:** Collective dynamics of very high density amorphous water	**Experiment number:** HS2231
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Report:

Very recently the existence of a new, third amorphous ice phase notified as very high–density amorphous (vHDA) ice has been conjectured [1]. This apparently successful preparation of an amorphous ice structure distinct from the well known low–density (LDA) and high–density (HDA) amorphous phase has imposed important questions about the nature of the phenomenon of amorphous polymorphism, which has not been understood, yet [2].

We have shown in previous experiments, that despite the obviously glass–like static structure factor the inelastic response of HDA and LDA is rather crystal–like displaying sharp phonon branches, showing harmonic behaviour in respect of temperature and energy and indicating the applicability of phonon selection rules [3,4]. In other words, judging from our inelastic results HDA and LDA could be nano–crystalline materials suggesting vHDA could be also related to a strongly disordered crystal structure rather than a liquid state.

To test this potential nano–crystalline scenario we have carried out high–resolution inelastic x–ray scattering experiments to sample phonon line shapes, dispersion and selection rules of vHDA. The inelastic beamline ID16 has been utilised for this purpose in an Si(111111) mode giving an energy resolution of 1.5 meV with an incoming energy of 21.7 keV. The ω–Q–phase space has been sampled in constant Q mode between $1.25 \leq Q \leq 16.25$ nm^{-1} with $\Delta Q = 0.75$ nm^{-1} and $-20 \leq \hbar\omega \leq 40$ meV with $\Delta\hbar\omega = 0.25$ meV.

[**1**] T. Loerting et al., *Phys. Chem. Chem. Phys.* 3, 5355, (2001). [**2**] O. Mishima et al., *Nature* 310, 393 (1984). [**3**] H. Schober, M.M. Koza et al., *Phys. Rev. Lett.* 85, 4100, (2000). [**4**] M.M. Koza, H. Schober et al., *Phys. Rev. B* 69, 24204, (2004).

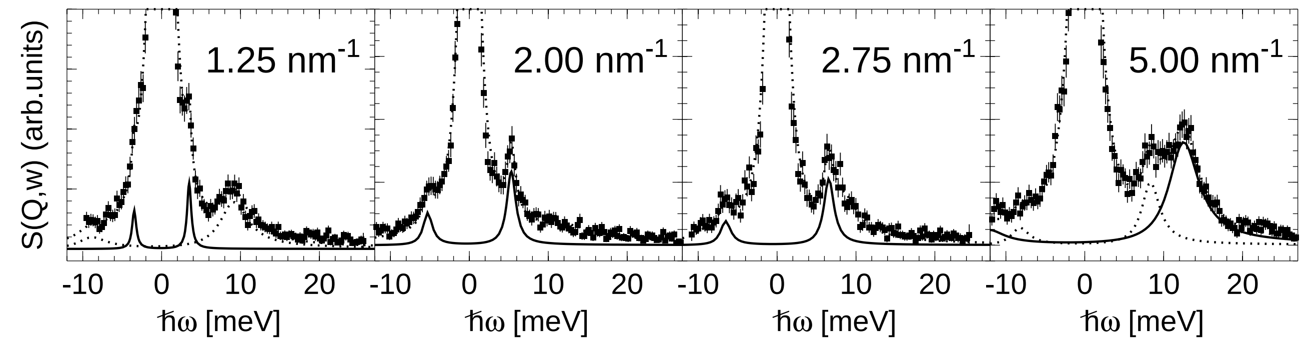

Figure 1: Spectra taken at the indicated Q values. Thick solid line indicates the sharp longitudinal acoustic phonon in the very high density amorphous ice phase.

In full accordance with the inelastic properties of HDA and LDA the vHDA ice has pronounced crystal–like response. A selected set of spectra are reported in Figure 1. The x–ray spectra of vHDA indicate a resemblance of the vHDA response with the dynamics of HDA. The obvious merit of the vHDA structure is the rather low elastic intensity accentuating details in the inelastic signal. This reduced elastic scattering gives evidence of a more homogeneous structure of vHDA in comparison to HDA.
Please note that in preceeding neutron scattering experiments we have proven that the HDA phase is a heterogeneous state. This heterogeneous state can be formed by two ways: first, by compression of crystalline ice or LDA, second, by heating of vHDA. Our finding leaves a lot of open questions concerning the relation between these two HDA forms. A detailed study of the dynamics of these two HDA forms is of highest importance since they can not be discerned by diffraction techniques!
Beyond, this very successful results on vHDA, we have experienced problems with the performance of the spectrometer. We have found out in different test measurements that at some Q setups our data have been obscured by an artificial signal. This signal is detected as a maximum at an energy corresponding with the tranverse optic mode in the sample. Therefore we require additional measurements to be able to estimate the contribution by this artificial signal. Figure 2 reports two spectra, the spectrum at 1.25 nm^{-1} displaying the artificial contribution.

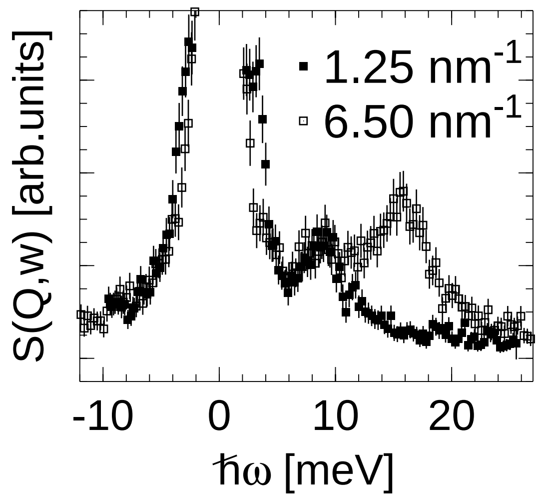

Figure 2: Spectra taken at the indicated Q values. Please note the strong artificial signal in the inelastic response taken at $Q = 1.25\ nm^{-1}$, which can not be discerned form the transverse optic phonon at 9 meV.