ESRF	**Experiment title:** Acoustic phonons in ice IX and ice XII	**Experiment number:** HS–1303
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Report:

The simple chemical compound water exhibits the phenomenon of amorphous polymorphism, i.e., it can be prepared in two different amorphous states at temperatures below 100 K. These states are labeled low–density amorphous (LDA) and high–density amorphous (HDA) phase which emphasises their strongly different densities of $\rho \approx 0.93$ g/cm^3 and $\rho \approx 1.17$ g/cm^3. Different theoretical concepts have been developed to account for the origin of HDA and LDA including the idea of two distinct glassy phases.

It has been proved by us in preceding experiments (ID16) that both HDA and in particular LDA exhibit crystal–like dynamic properties. For example, they do not show strong damping behaviour of acoustic excitations with $Q \sim$ nm^{-1} and $\hbar\omega \sim$ meV. This dynamic feature is in clear contrast to dynamic properties found in supercooled liquids so far. In the case of LDA we have shown that the behaviour of its acoustic modes corresponds to the behaviour of longitudinal acoustic phonons in the cubic crystalline water phase I$_c$. This inelastic resamblance is reminiscent of water tetrahedra which are the basic microscopic structure units in both, LDA and cubic ice I$_c$.

The inelastic response of HDA, despite its distinctness from any glass forming system, has not been successfully accounted for by other crystalline water phases, so far. For this reason we have performed two sets of measurements on two distinct high–density crystalline water phases, namely ice IX ($\rho \approx 1.17$ g/cm^3) and ice XII ($\rho \approx 1.30$ g/cm^3). Both crystalline phases can be prepared at similar conditions at which HDA is produced and show comparable stability limits as HDA, emphasising their applicability as crystalline reference systems.

The experiment was performed with Si(11 11 11) (E = 21.75 keV) monochromator reflection, giving a resolution of $\Delta E \approx 1$ meV. Constant Q scans were taken at Q= 2.0, 3.5, 5.0, 6.5, 8.0, 9.5, 11.0, 12.5, 14.0 and 15.5 nm^{-1} covering the range of two Brillouin–zones in the crystalline samples. The energy range -17 to +40 meV was chosen to access all molecular vibration modes in the water phases. All measurements were carried out at $T \approx 70$ K.

A selected set of data is shown in the figure below. The spectra demonstrate the sensitivity of the inelastic x–ray technique to structural correlations which are reflected in the phonon selection rules and demonstrated in the figure by the onset of transverse modes as additional intensity in the vicinity of the Brillouin–zone edge (ice IX at $Q = 5.0 - 6.5$ nm^{-1}). A comparison of the data with results obtained on HDA in preceding experiments indicate a strong resemblance of ice IX and HDA. In particular the extent of the first Brillouin–zone of ice IX agrees very well with the one found in HDA. This implies that the static structure factor of HDA does not reflect the correct structural correlations present in the sample.

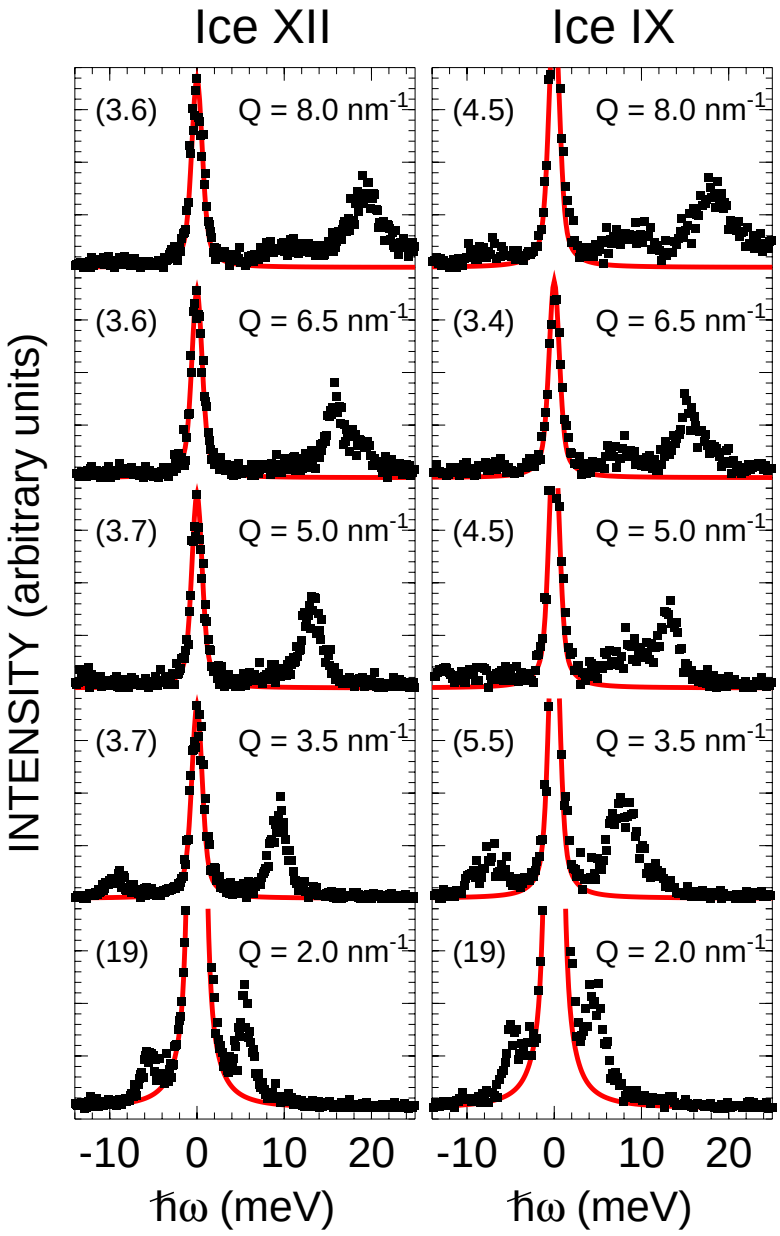

Figure: Inelastic x–ray spectra of ice IX and ice XII measured on ID28. The solid line is the resolution function of the spectrometer. Q numbers are given in the figures, numbers in brackets indicate the maximum elastic intensity.