



Experiment title: Investigation of the amorphous polymorphism of H₂O ice

Experiment number:

HS-358

Beamline:

ID16

Date of Experiment:

from: 10-Oct-97

to: 18-Oct-97

Date of Report:

25-Feb-98

Shifts:

21

Local contact(s) :

F. Sette

Received at ESRF:

02 MAR. 1998

Names and affiliations of applicants (*indicates experimentahsts):

*H. Schober, ILL

*M. Koza, *A. Tölle, University of Dortmund

*F. Sette, *C. Masciovecchio, *A. Mermet, ESRF

*F. Sciortino, University of Rome

Report:

Using the very high resolution inelastic x-ray beamline ID16 we have investigated the dynamical response of high- (hda) and low-density (lda) amorphous water modifications as well as cubic crystalline ice.

Hda was produced by compression of hexagonal ice in a piston-cylinder apparatus at 77 K. The metastable hda was retrieved from the cell and transferred to a steel sample container mounted magnetically to the cold finger of a precooled He-refrigerator. Special care was taken to prevent heating of the sample beyond 80 K during the manipulations.

It is known that when producing hda in the above described way there is an appreciable chance for high-density crystalline (hdc) contaminations. We checked the quality of all our samples by careful measurements of the static structure factor $S(Q)$ and indeed found Bragg scattering in a large number of samples. All the inelastic results presented were obtained on non-contaminated samples.

Unfortunately the temperature of the sample turned out appreciably higher than indicated by the sensor mounted on the cold finger as indicated by the slow conversion of hda towards lda during the measurement at nominally 80 K. The state of the hda sample is, therefore, rather ill-defined.

Figure 1 shows the spectra of Ida and I, as measured at 80 K. The excitations of the first spectrum at 3 nm^{-1} lie within the boundary of the first Brillouin (pseudo-Brillouin zone) of I_c and Ida, while the other spectra sample the region close to and outside the zone boundary. The peaks originating from the longitudinal sound mode are clearly visible in all three cases. The LA-peaks are broadened and shifted to somewhat smaller center positions in Ida as compared to cubic ice. Averaging over all Q -values below 5 nm^{-1} the shift in peak position translates into a shift in the speed of sound from about 3750 m/s in cubic ice to 3450 m/s in Ida. These values compare favorably with the density of states determined by inelastic neutron scattering. As expected for a crystalline system, there is only longitudinal scattering in I, within the first zone. Weak additional scattering between the elastic and LA peaks sets in when crossing the zone boundary. In Ida signs of this additional scattering are observed also within the first pseudo Brillouin zone, in accordance with the amorphous nature of the sample. Within the experimental difficulties outlined above the speed of sound in hda is determined as approximately 3400 m/s , i.e. not appreciably different from Ida.

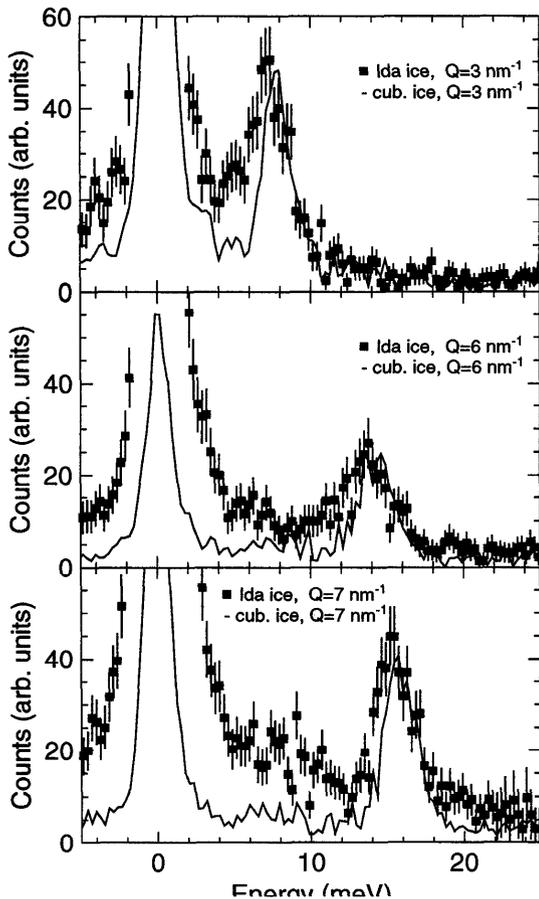


Figure 1:
Measured inelastic X-ray spectra (approx. 80 K) of cubic and Ida ice at several Q -values.