	Experiment title: Optical phonons in molten salts	**Experiment number**: HS1644
Beamline: ID 28	**Date of experiment**: from: 26/9/01 to:2/10/01	**Date of report**: 28/2/02
Shifts: 18	**Local contact(s)**: M. Lorenzen/H. Requardt	*Received at ESRF:*
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Report:

About 25 years ago, Hansen and McDonald carried out the pioneering MD-work on liquid alkali halides, the prototypes of ionic liquids[1]. These simulations revealed the existence of propagating short wavelength charge fluctuations (“optical phonons”). The aim of our experiment was to obtain a direct experimental confirmation for these excitations in molten NaCl.

The experiment was performed at the inelastic beamline ID28 using the Si(999) configuration with an energy resolution of about 3 meV (FWHH). The NaCl sample was contained in a single crystal sapphire cell with a wall thickness of 0.25 mm. High temperatures could be achieved by an internally heated vessel equipped with Be windows, which can cover scattering angles up to 25°. For a suitable subtraction of the inelastic scattering from the sapphire cells empty cell measurements have been performed at the same temperature (1170 K) and with the same cell. The used setup has the possibility to measure the empty sample cell at high temperature and then fill in the molten salt from a reservoir. At 1170 K we have recorded 20 different momentum transfer vectors Q from 1 nm^{-1} – 29 nm^{-1}. For the interesting low Q region up to 10 scans have been measured for a good statistics.

For the data evaluation a normalisation, a detailed balance correction and an empty cell subtraction with an appropriate absorption factor has been performed. Three resulting spectra are shown in Fig. 1 in a logarithmic scale for $S(Q,\omega)/S(Q)$ as an example. For the fit of the data a model with one Lorentzian line and a damped harmonic oscillator (DHO) function was used. The fit procedure included a convolution with the resolution function, represented by a Voigt function.

Clearly visible in the spectra are strong inelastic shoulders, which have been fitted by the DHO. The results are shown in Fig. 2 with the width for the damped harmonic oscillator Γ. These excitations show a dispersion and are the acoustic modes. Surprisingly these acoustic modes are quite well defined (not overdamped for a wide range of momentum transfer vectors), which was not recognised so clear in former measurements with neutrons or MD simulations. The second remarkable result is a large positive dispersion in this ionic liquid, which gives a phase velocity about 60 % higher than the adiabatic sound velocity. The interpretation of this phenomenon is under way.

The search for optic modes is much more difficult. In Fig. 1 some arrows are given where small intensity peaks appear far away from the acoustic ones and also away from the sapphire phonons. The frequency positions of these arrows fit very well to the predictions of MD simulations. For larger Q- values the optic modes interfere with the acoustic ones. However, a reliable fit is difficult, therefore we are proceeding the data analysis with current correlation functions. The difficulty lies in the fact that we are looking for a weak signal on a high wing of the Lorentzian like resolution function.

[1] J. P. Hansen, I. R. McDonald, Phys. Rev. A **11**, 2111 (1975)

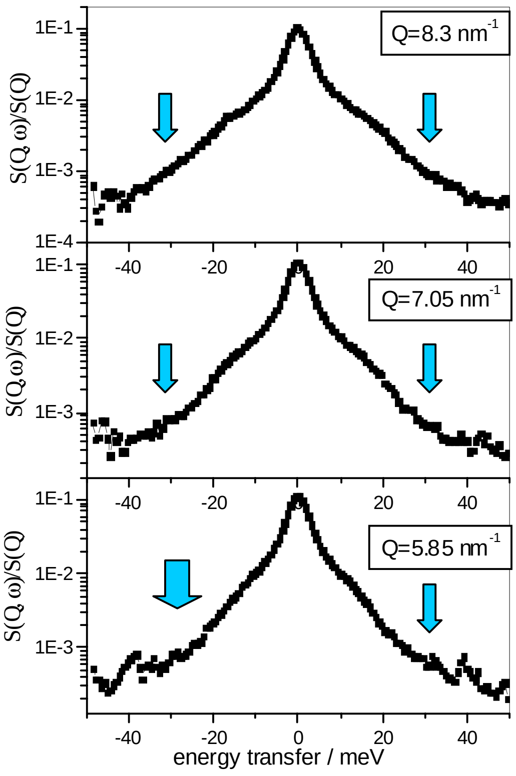

Fig. 1

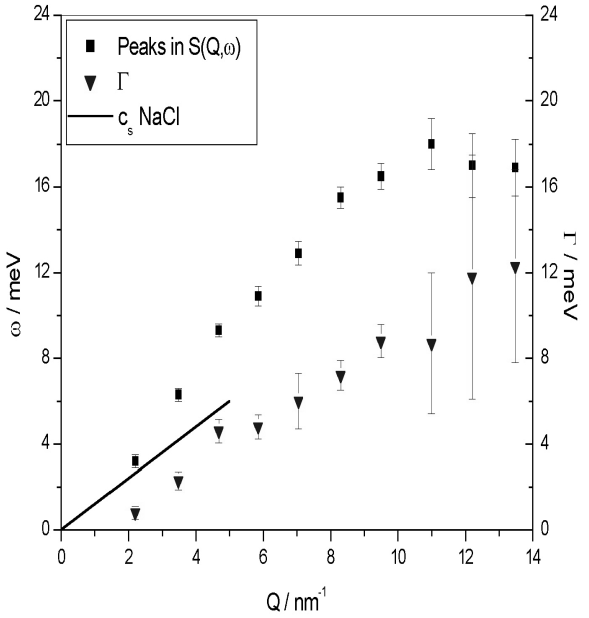

Fig. 2