

	Experiment title: Acoustic modes in molten KCl	Experiment number: HS1988
Beamline: ID28	Date of experiment: from: 29/8/02 to: 5/9/02	Date of report: 1/9/03 <i>Received at ESRF:</i>
Shifts: 18	Local contact(s): M. Lorenzen/R. Verbeni	
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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”). But no hint for acoustic modes could be found in the simulation spectra. Our experiment was intended to search for acoustic like excitations in molten KCl. The chosen KCl has special scattering properties for synchrotron radiation. The scattered intensity of a binary charged liquid can be written generally in the following form:

$$\frac{\partial^2}{\partial Q^2 \partial \omega^2} \left(f_A + f_B \right)^2 S_{NN}(Q, \omega) + \left(f_A^2 - f_B^2 \right) S_{NZ}(Q, \omega) + \left(f_A - f_B \right)^2 S_{ZZ}(Q, \omega)$$

$S_{NN}(Q, \omega)$ represents the scattering law due to density-density correlations and $S_{ZZ}(Q, \omega)$ stands for the charge-charge correlations. $S_{NZ}(Q, \omega)$ is the cross correlation scattering function. The prefactors allow to vary the scattering contrast by the use of different alkali halides similar to isotopic substitution in neutron scattering. For instance due to nearly identical form factors of K^+ and Cl^- only the density fluctuations will be visible in liquid KCl and the acoustic phonons can be determined without deterioration due to optical contributions.

The experiment was performed at the inelastic beamline ID28 using the Si(11 11 11) configuration with an energy resolution of about 1.5 meV (FWHM). The KCl 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 (1070 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 1070 K we have recorded 9 different momentum transfer vectors Q from $2.6 \text{ nm}^{-1} - 14 \text{ nm}^{-1}$. Due to problems with the tightness of the sample container which could be solved during the experiment and a temporary misalignment of the focusing mirror only a part of the Q range could be measured with a poor statistics. Nevertheless we could extract some results about the acoustic dynamics of molten KCl.

Three resulting spectra are shown in Fig. 1 for the lower Q -range. In the spectrum of the lowest Q vector also the acoustic modes of the sapphire are visible. 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.

The inelastic shoulders have been fitted by the DHO model. The fit results are shown in Fig. 2 with the width for the damped harmonic oscillator Γ . The collective modes are damped but not overdamped in a quite large range of momentum transfer vectors. A large positive dispersion of about 60 % can be deduced from the data.

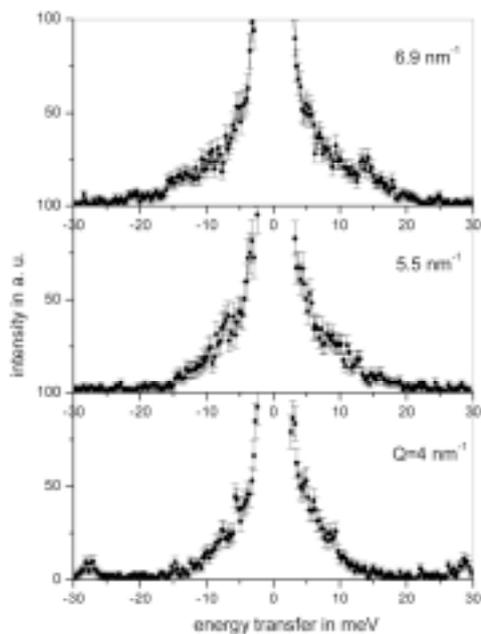


Fig. 1

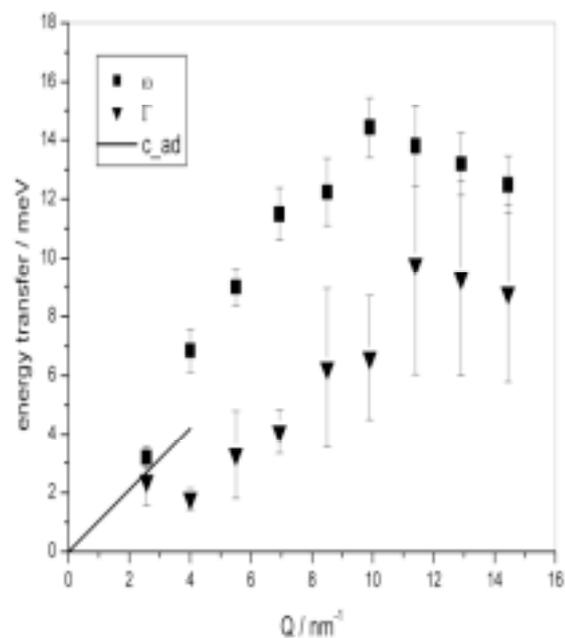


Fig. 2