ESRF	**Experiment title:** Liquid-liquid transition in yttrium aluminates. The IXS point of view	**Experiment number**: HD-395
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Report:

Introduction

The aim of the proposal was to study the evolution of the dynamics of Al_2O_3-Y_2O_3 compounds in the supercooled state in order to evidence the liquid-liquid transition observed by Greaves et al using small angle x-ray scattering [1]. This experiment was challenging and not simple due to the strong absorption of the sample at the working energy.

After a long period of alignment and optimization, it appeared that that it was difficult to measure the inelastic signal from the sample ant that the experiment was not feasible using our experimental conditions.

For using the remaining beamtime, we decided to use the glass forming composition $CaAl_2O_4$ that presents much less absorption.

Experiment

The IXS experiments were performed using the high resolution spectrometer at the ID16 beamline. We worked with an incident energy of 23.725 keV using the reflection Si (12,12,12). The global resolution was about 1.3 meV. The high temperature setup and the data treatment are described in details in ref [2].The configuration of the spectrometer makes it possible to measurement the scattered beam at 9 equidistant *Q*-values (*Q*-set). Different *Q*-sets are obtained by rotating the detection arm.

Since our samples required a relatively long acquisition time, we performed the measurements at only one *Q*-set (Q_1 = 1, 2.6, 4.3 and 5.8 nm^{-1}) that was sufficient to provide a good interpretation of the data. It can be noted that for *Q* values above 5.8 nm^{-1}, the signal was too damped and Brillouin peaks were not visible.

The IXS spectra are shown in Fig. 1a. They were fitted by a Damped Harmonic Oscillator (DHO) model but the fit didn't show any agreement with the experimental data.

Considering the results obtained earlier for liquid alumina [3], a model involving two relaxation times as described in ref [2] was used to model the experimental data and we obtained a good agreement between the experimental and calculated spectra (Fig1a).

In this model, thermal fluctuations are neglected and the density fluctuations are described by a memory function given by the sum of two contributions: a slow component following a Debye law (exponential decay) and a fast component that is effectively instantaneous:

$$M(Q,t) = \Delta_\alpha{}^2(Q) e^{-t/\tau_\alpha(Q)} + 2\Gamma_S(Q)\delta(t) \qquad (1)$$

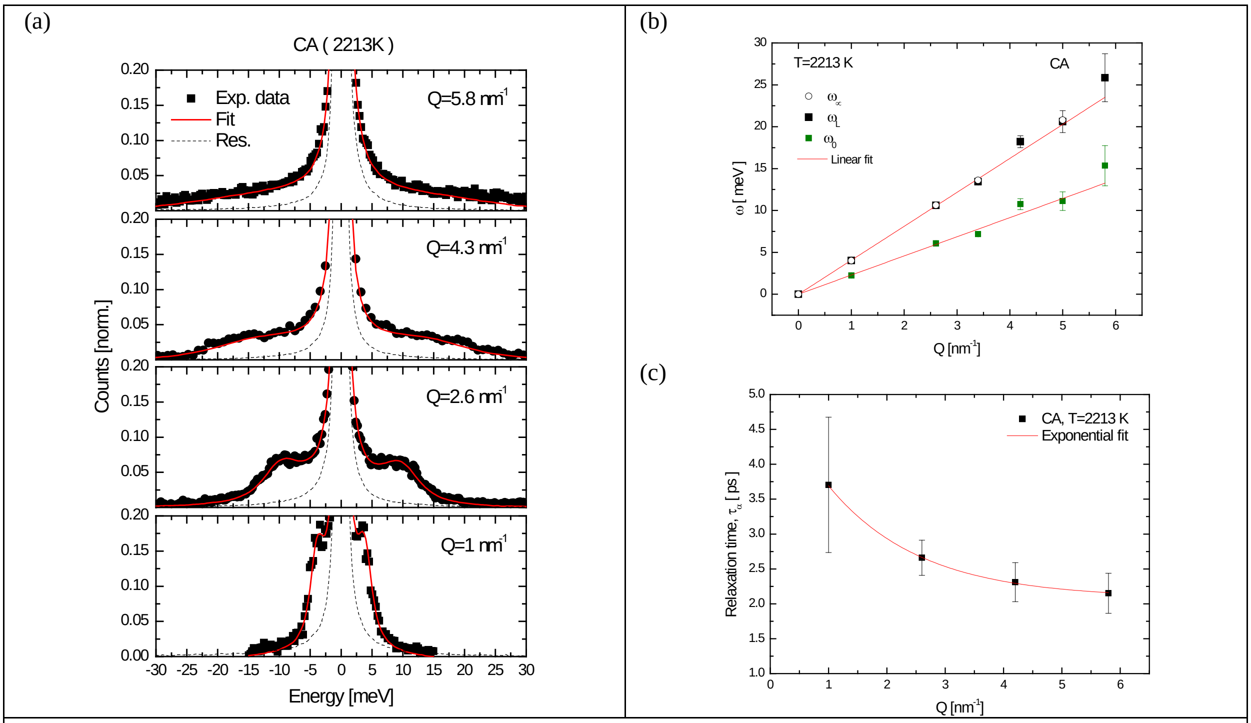

Fig. 1. (a) IXS spectra of liquid CA at 2213K. (b) Apparent, infinite frequency and isothermal sound velocities. (c) Evolution of the relaxation time as a function of Q.

The acoustic dispersion relation $\omega_L(Q)$ is shown in Fig. 1b. The excitation frequency ω_L is taken as the position of the maximum of the longitudinal current correlation spectrum $J_L(Q,\omega) = \omega^2 / Q^2 S(Q,\omega)$. In the same figure we report the $\omega_0(Q)$ obtained from the fit using Eq. (1). From the linear dependence of the excitation frequencies, we can determine the isothermal sound velocity $c_t(Q) = \omega_0(Q)/Q = 3474 \pm 37$ m/s and the apparent sound velocity $c_l(Q) = \omega_l(Q)/Q = 6235 \pm 53$ m/s.

As it can be seen from Fig. 1b, the apparent sound velocity is equal to the high-frequency sound velocity $c_\infty(Q) = \sqrt{\omega_0^2(Q) + \Delta_\alpha^2(Q)}/Q$ over the entire Q range. This implies that the high-temperature liquid responds as a solid to a high-frequency probe

Fig. 1c shows the Q-dependence of the structural relaxation time $\tau_\alpha(Q)$. The red line is an exponential decay fit and we see that it describes well the data. If we extrapolate the fit to $Q = 0$, we obtain a value $\tau_\alpha = 5.1 \pm 0.23$ ps.

Using the eqution $\eta_l(Q) = \rho(\Delta_\alpha^2 \tau_\alpha + \Gamma_S)/Q^2$, we can alxo calulate a longitudinal viscosity of 83 mPa.s.

Further work

Liquid Ca is very fragile and further works will be devoted to the study of the influence of SiO_2 (strong liquid) on the dynamics.

Reference

[1] N. G. Greaves et al, Science, 322, 566-570 (2008)

[2] I. Pozdnyakova, L. Hennet, J. Brun, D. Zanghi, S. Brassamin, V. Cristiglio, D. Price, F. Albergamo, A. Bytchkov, S. Jahn, and M. Saboungi, J. Chem. Phys. 126, 114505 (2007).

[3] H. Sinn, B. Glorieux, L. Hennet, A. Alatas, M. Hu, E. E. Alp, F. J. Bermejo, D. L. Price, and M.-L. Saboungi, Science 299, 2047 (2003)