ESRF	**Experiment title:** Dynamics in high temperature levitated liquid oxides	**Experiment number**: HS-2675
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Shifts: 18	**Local contact(s)**: Francesco ALBERGAMO	*Received at ESRF:*

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Report:

In the experiment reported here the microscopic dynamics of the liquid $MgAl_2O_4$ and $MgAl_4O_7$ oxides was studied by inelastic x-ray (IXS) scattering using the containerless environment.

These materials are of technological interest, especially for its refractory properties. Its behaviour under irradiation, thermal stability and rigidity are interesting properties in confining nuclear waste. In addition, MgO containig oxides are a component of the Earth's mantle. Therefore, the transport properties of high-temperature oxide melts are of considerable interest for a variety of applications. In particular, information on melt viscosities and the speed of sound through liquid oxides is essential for testing the validity of theories used in predicting the geophysical behaviour of the Earth's interior.

In order to avoid any problem with containers, the measurements have been performed using a containerless environment. In the past few years, the CRMHT in Orléans has developed apparatus to combine levitation and laser heating techniques with X-ray and neutron diffraction at synchrotron [1,2] and neutron sources [3] and we have built a special setup specially designed for ID16. We used this device for the first time in April 2005.

Spherical samples with diameters between 2.5 and 3mm were aerodynamically levitated using an oxygen/helium gas jet and heated with a 125W CO_2 laser beam up to 2000°C. The gas flow was regulated using a mass flow controler which maintained the sample at a stable position. The temperature was measured by an optical pyrometer focused on the sample. A large part of the sample was out of the levitator so that the beam was transmitted through it. All spectra were taken at 2423 K.

The IXS experiments were carried out using the horizontal high energy-resolution IXS spectrometer installed at ID16. The incident energy was 21.747keV giving a spectrometer resolution around 1.5meV. Five analyzers were available on the 2θ arm. Data were collected over the energy transfer range -50 to 50 meV at 3 positions of the first analyzer (1, 2 and 3 nm^{-1}) covering the Q range 1 to 15 nm^{-1}. The total scan duration for one position of Q was around 6 hours.

The dynamic structural factors S(Q, ω) show a triplet peak structure at wavevectors up to 7 nm^{-1}. The data analysis was performed using generalized hydrodynamics approach. From generalized Langevin equation, it is possible to derive the classical dynamic structure factor in terms of the real and imaginary parts of the Fourier-Laplace transform of a complex memory function M (Q, t) that contains the transport properties information. To determine the sound speed, the oversimplified memory function was used, and the S(Q, ω) behavior is described by hydrodynamics. The linear Q dependence of the excitation frequencies, renormalized by the damping term $\Omega_S = \sqrt{\omega_S^2 + \Gamma_S^2}$, corresponds to a longitudinal sound velocity $v_L=\Omega_s/Q$ (Fig. 1). The longitudinal sound speed is 6380 ± 150 m/s for $MgAl_2O_4$ and 6820 ± 135 m/s for $MgAl_4O_7$.

To determine the viscosities, we used the viscoelastic memory function : $M(Q,t) = \Delta_\alpha^2(Q)e^{-t/\tau_\alpha(Q)} + 2\Gamma_S(Q)\delta(t)$, where τ_α- the structural relaxation time, Δ_α- the structural relaxation strength, and Γ_s -the Brillouin linewidth.

The generalized longitudinal dynamic viscosity determined by : $\eta_l(Q) = \frac{\rho\left[\Delta_\alpha^2\tau_\alpha + \Gamma_S(Q)\right]}{Q^2}$ is reported in Fig. 2 together with the Q-independent viscosity value from Ref. 4.

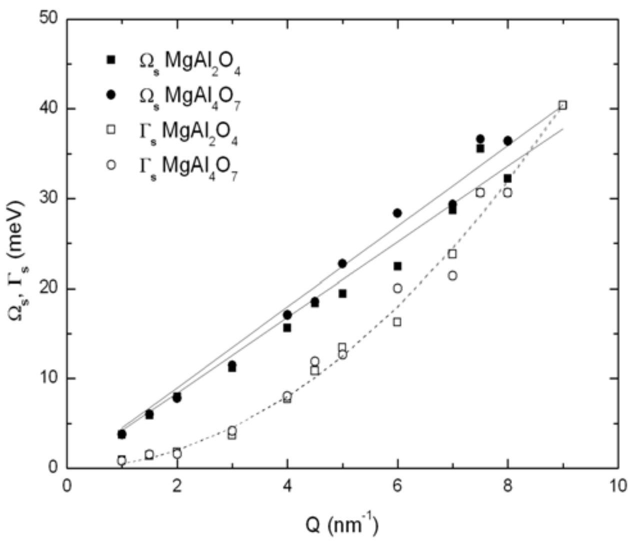

FIG 1. Dependence on the wave vector Q of the frequency Ω_s and half-width Γ_s of the Brillouin peaks for $MgAl_2O_4$ and $MgAl_4O_7$. The solid lines are linear fits with $v_L= \Omega_s/Q$ = 6380±150 m/s and 6820±135 m/s, and the dashed lines are parabolic fits $\Gamma_s = \alpha Q^2$ with α = 0.756±0.012 and 0.761±0.021 mm^2s^{-1}.

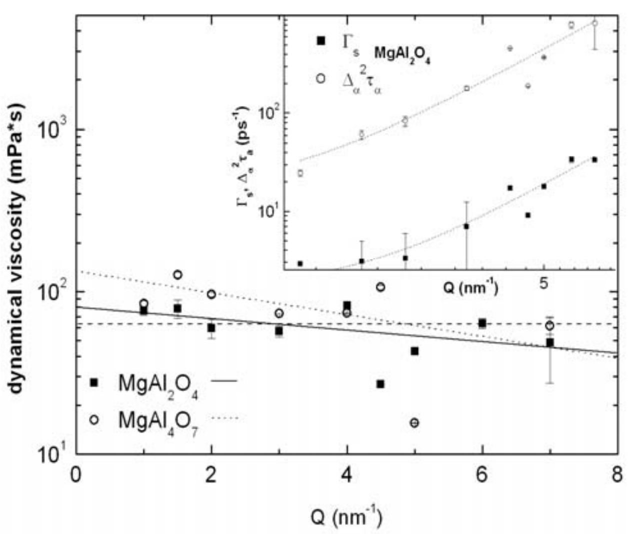

FIG. 2. Values of $\eta_L(Q)$ as determined by the memory function parameters for $MgAl_2O_4$ and $MgAl_4O_7$ samples. Solid and dotted lines are linear fits to the data, dashed line is Q-independent viscosity for liquid Al_2O_3 (from [4]). Inset: partial contributions due to the α- and s-process, respectively for $MgAl_2O_4$. The dashed lines emphasize a Q^2 behavior for two contributions

From the experiments performed, for the first time, the longitudinal sound velocities and viscosities of two refractory oxide melts with extremely high melting points were determined. The viscosity relaxation can be described satisfactory with one relaxation time model. The experimental data is compared with MD simulations. Both experiment and simulation show a viscosity maximum around the $MgAl_4O_7$ composition.

Références

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