STRUCTURAL RELAXATION IN SUPERCOOLED SELENIUM

Benoît Ruffl[1] and Stéphane Longeville[2]

[1]Laboratoire des Verres, Université Montpellier II, 34095 Montpellier Cedex 5, France
[2]Laboratoire Léon Brillouin (CEA-CNRS), CEA-Saclay, 91191 Gif-sur-Yvette, France

In the last decade many experiments as well as computer simulations have established that the so-called mode-coupling theory of the glass transition (MCT) [1] is able to give a *qualitatively* correct description of the structural relaxation dynamics of fragile supercooled liquids at least in the weakly supercooled state. In previous works, we have studied the structural relaxation dynamics of a complex oxide glass (Na_2O-Li_2O-$2P_2O_5$) by means of neutron scattering in a broad range of temperature [2]. It has been clearly demonstrated that also intermediate glass formers show features that are in *qualitative* agreement with the predictions of MCT [2,3]. However, starting from the static properties of the system, MCT is able to give a *quantitative* description of the relaxation dynamics of supercooled liquids. In particular it is possible to calculate from the knowledge of the static structure factor *S(q)* the time dependence of the coherent intermediate scattering function *F(q,t)*. These type of calculations have been done only for very few systems (most are fragile and/or monoatomic ***simulated*** systems), since they are quite involved even for binary systems [4].

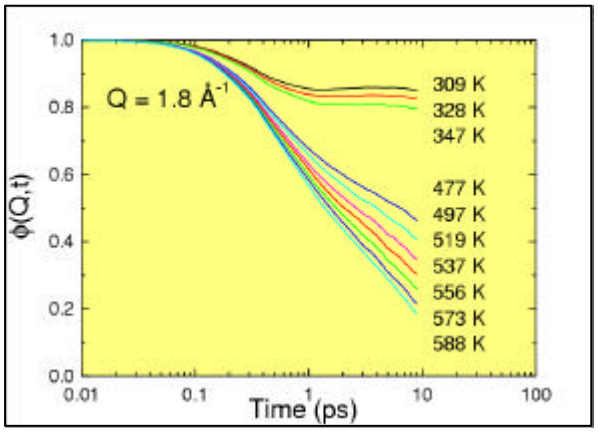

Figure 1. Temperature dependence of the normalized intermediate scattering function of Selenium at Q=1.8 Å^{-1} measured on Mibemol.

The final aim of the reported experiments here is to compare, for the first time, detailed MCT calculations and inelastic neutron scattering experiments on liquid Selenium, a monoatomic system. Selenium is an inorganic polymerlike glass former of intermediate fragility with a low glass-transition temperature T_g=303 K (T_m=494 K). Due to its predominant coherent scattering cross-section Selenium is a particularly good candidate for neutron spin echo experiments and such an analysis. Fig.1 shows the temperature dependence of the normalized intermediate coherent scattering function ϕ(Q,t) measured at the LLB on the time-of-flight spectrometer Mibemol at Q=1.8 Å^{-1}, a value close to the first maximum of the static structure factor. The typical slowing down of the structural relaxation when cooling the liquid from above the melting temperature down to the glass transition temperature is observed. At very short time (~0.1 ps) the correlation functions decay due to the vibrational dynamics. This is followed by a two-step decay: the *b*-regime (~0.1-1 ps) and the structural or *a*-relaxation which is hardly temperature dependent. The gap between 477 K and 347 K corresponds to the temperature range

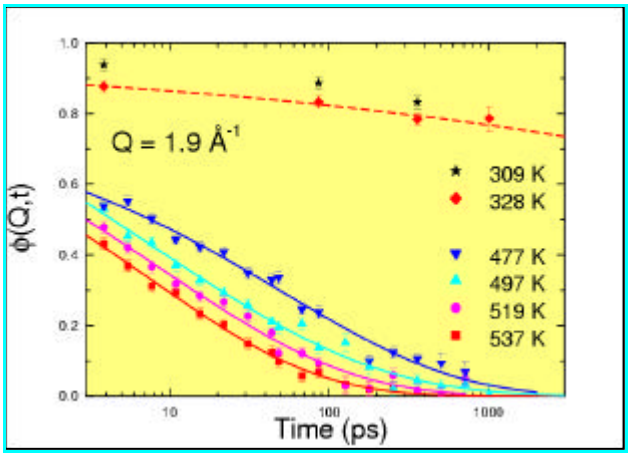

Figure 2. Temperature dependence of the normalized intermediate scattering function of Selenium at Q=1.9 Å^{-1} measured on G1bis.

where crystallization could not be avoided. In order to get in more details the shape of the structural relaxation, its temperature and wavenumber dependence, we have completed the long-time part of some of the TOF spectra with neutron spin-echo data measured on G1Bis at the LLB. Fig. 2 exhibits the temperature dependence of the normalized intermediate coherent scattering

function $\phi(Q,t)$ measured at the LLB on the spin-echo spectrometer G1Bis at a selected Q value, namely Q=1.9 Å^{-1}. The solid lines are fits using the usual simple stretched exponential function with all parameters free. In a first analysis all the high temperature spin-echo data, above the crystallization region, are compatible with a common stretch exponent b=0.5. However, due to the scatter of data and the limited time-scale of the spin-echo machine, a small temperature and/or wavenumber dependence cannot be excluded. Combining the TOF spectra together with the spin-echo data will certainly make it clear. An efficient way to visualize the Q-dependence of the structural relaxation time with a spin-echo machine is to measure the polarization at selected times t only. As shown for example in Fig. 3 for T=500 K, the data clearly reveal a peak around 1.2 Å^{-1}, a value close to the position where a small prepeak is detected in the static structure factor of liquid Selenium at high temperature [5]. Assuming a common stretch exponent parameter b, the Q-dependence of the structural relaxation timescale can be estimated from these data and have been plotted in Fig. 4. The complete time dependence of the solution of the mode-coupling equations in which the full Q dependence is taken into account has been calculated with the experimental static structure factor of liquid Selenium at 531 K [5] as only input (apart particle density). A first result of these mode-coupling calculations is that the shape of the structural relaxation seems to be rather different from the experimental one. The estimated stretch exponent parameter is indeed around b_{MCT}=0.75-0.8 and not 0.5 as found here in these experiments. As recently found for silica [4], the prototype of strong liquids, the mode-coupling equations used could be too simple to describe Selenium. On the other hand the Q-dependence of the calculated structural relaxation timescale, and plotted in Fig.4, clearly reproduces the experimental peak found around 1.2 Å^{-1}. The detail of the Q-dependence is again far from being perfect probably for the same reason. It has also to be noted that the exact values of the relaxation times cannot be compared at this stage of the analysis.

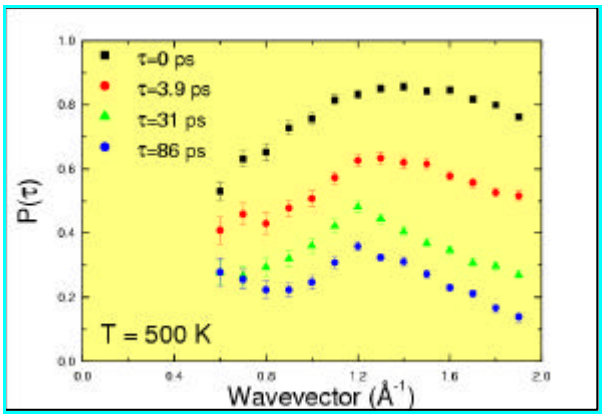

Figure 3. Q-dependence of the polarization measured on G1bis at T=500 K for selected t values.

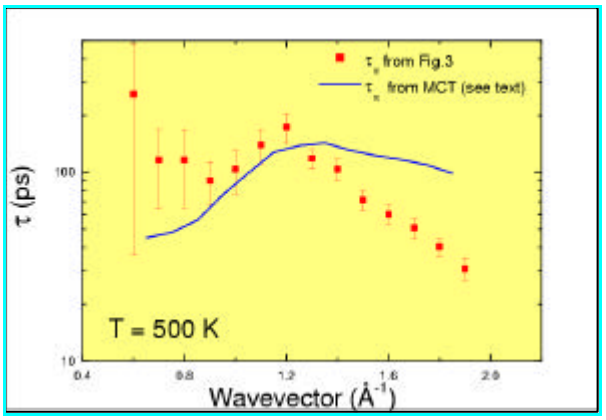

Figure 4. Q-dependence of the a-relaxation time deduced from Fig. 3. The full line is the a-relaxation time Q-dependence deduced from MCT calculations.

References

[1] W. Götze, *J. Phys. Condens. Matter* **10** (1999) A1.
[2] B. Rufflé, C. Ecolivet and B. Toudic, *Europhys. Lett.* **45** (1999) 591.
B. Rufflé and al., *J. of Non-Cryst. Solids* **235-237** (1998) 244.
B. Rufflé and al, *Phys. Rev. B* **56** (1997) 11546.
[3] T. Franosch, W. Götze, M.R. Mayr and A.P. Singh, *Phys. Rev. E* **55** (1997) 3183.
[4] F. Sciortino and W. Kob, *Phys. Rev. Lett.* **86** (2001) 648.
[5] R. Bellissent and G. Touraud, J; *Non-Cryst. Solids* **35&36** (1980) 1221.