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Report:

Vitreous silica is the prototype of strong glasses, being characterized by a low value of the fragility parameter, of the order of 20. This low fragility value is associated to very high glass transition and melting temperatures, around 1450 and 2000 K respectively.
The determination of its vibrational dynamics by means of inelastic X-rays scattering (IXS) in the liquid phase thus represents a challenging experimental task.

In the glassy phase, vitreous silica has been the subject of many IXS investigations, and the main results in the low-Q region (exchanged wave vector $Q < 4$ nm^{-1}), are the following: *i)* the IXS Brillouin spectra are described by a single acoustic mode with energy position Ω linearly dispersing with Q [1] *ii)* in this low Q region the width of the inelastic peaks, Γ, follows a Q^2 law and doesn't seem to be temperature dependent [2].
A recent study of silica at frequencies around 100 GHz [3] by inelastic UV scattering has shown the presence of a characteristic step-like behaviour of $\Gamma(Q)$ for Q_c values around 0.15 – 0.2 nm^{-1}. A similar crossover value has been found in a densified silica sample at Q values ten times bigger, $Q_c \sim 2$ nm^{-1}, by IXS [4].

The physical mechanism underlying the observed Q^2 dependence of the sound attenuation and the change in slope towards an higher Q dependence below Q_c have been the subject of numerous theoretical interpretations but none of these appears to give a satisfactory explanation [5,6]. Between these theories we may recall the mode coupling theory that Gotze and Mayr [6] applied to the vibrational dynamics of an hard spheres system. These authors predict a deviation from the Q^2 law for wave-vectors between 0.8 and 2 nm^{-1}.

To investigate the THz frequency dynamics of vitreous silica in the liquid phase we have developed a new oven for high temperature IXS studies. The oven operates under vacuum and employs a graphite cylinder as heating element. The graphite is enclosed in a boron nitride screen to reduce heat irradiation and is heated by

a DC power supply. This solution allows to reach very high temperatures, being limited only by the vacuum evaporation rate of graphite which becomes important only above 2500 K.

The confinement of the sample at temperatures around 2000 K is a quite difficult task, for two main reasons: silica is characterized by a very high vapour pressure and the difficulty to find a material transparent to x-rays and solid at such temperatures. To this aim we have used a sapphire cell inside the graphite heating element. However this choice has revealed to be unstable at temperatures above melting (T_m ~ 2000 K). The reason for this instability is probably due to the direct contact between sapphire and graphite and to skin effects associated to the high current (250 Amps) in the graphite.

We are now testing a new graphite heater in which the direct contact between sapphire and graphite is avoided. This solution appears to be stable and to allow for future IXS measurements above the melting point of silica.

We have performed a set of constant Q-scans measurements of the dynamic structure factor S(Q,ω) at three temperatures T=1800, 1970 and 2070 K.

As an example of the quality of the measured spectra, the S(Q,ω) at two temperatures and at Q=2 nm^{-1} is plotted in figure 1 together with the best fitted function (an elastic line plus a damped harmonic oscillator convoluted with the resolution).

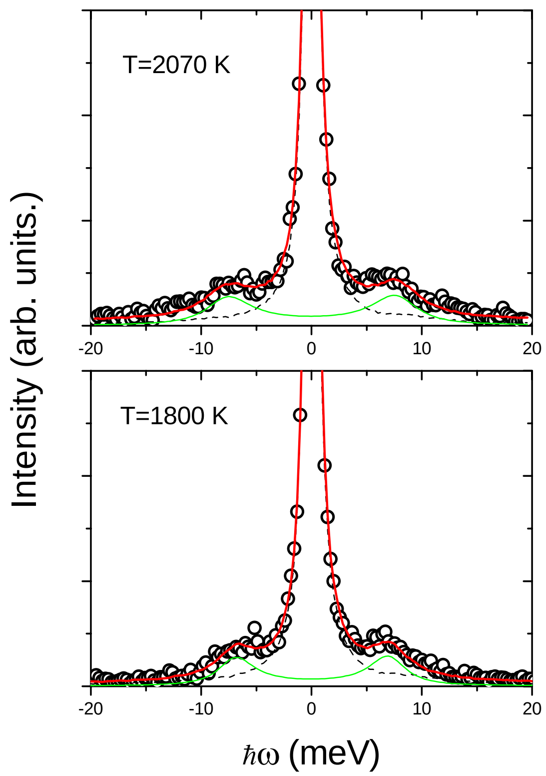

Figure 1. IXS spectra of SiO2 in the supercooled liquid phase (T=1800 K) and in the liquid phase (T=2070 K) at an exchanged wave-vector Q = 2 nm^{-1}.

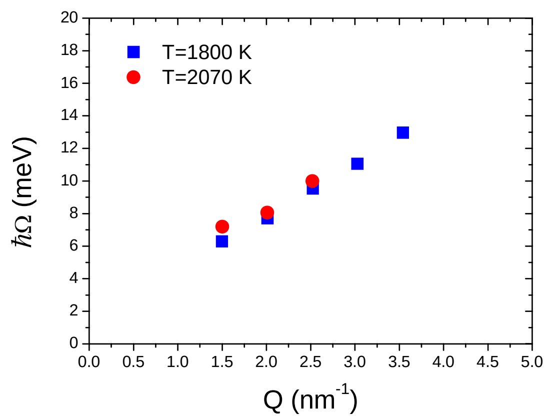

Figure 2. Dispersion curve at two of the three measured temperatures

The oven instability at temperature above T_m has made it very difficult to measure the IXS signal. For this reason it has been possible to measure only a limited portion of the dispersion curve with only a few points, as shown in figure 2.

In order to complete the proposed experiment we will submit a continuation proposal to probe the vibrational dynamics in the liquid phase using the new heating element and sapphire container.

References

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