

**Experiment title:**

Temperature dependence of the positive dispersion in vitreous silica

**Experiment number:**

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**Report:****Microscopic Relaxation in vitreous silica****Abstract**

The dynamic structure factor of vitreous silica is measured by means of inelastic X-rays scattering at exchanged wavevectors between 4 and 15 nm<sup>-1</sup> and at temperatures above and below the glass transition temperature  $T_g = 1450$  K.

The results of a detailed analysis confirm the presence of two excitations in the spectra in the entire explored wavevector region. A positive dispersion of the higher frequency mode is detected and it is found to be temperature independent from deeply below to above  $T_g$ .

This result confirms the harmonic nature of the vibrational dynamics in the silica glass in the THz frequency region and sustains the hypothesis of a microscopic relaxation process to be at the basis of the observed positive dispersion. The nature of the modes is then further investigated by looking at the frequency dependence of their damping and integrated intensity.

A breakdown of the ubiquitous  $\Omega^2$  dependence of the line width is found to take place at frequencies slightly above that of the Boson peak.

## Experimental details and results

The sample used for the experiment is a disk shaped Suprasil fused quartz sample purchased by Goodfellow, with a 2 mm diameter and a thickness of 1.4 mm, chosen to match the photoelectric absorption length of vitreous silica at the used incident energy of 17.8 KeV.

The sample is placed in a cylindrical graphite holder of 5 mm diameter which presents two circular openings of 1 mm diameter along the beam trajectory. The graphite holder is enclosed in an high temperature chamber specifically designed for IXS experiments. The oven is a vacuum chamber, maintained at a pressure around  $10^{-6}$  mbar, closed with Be windows and externally water cooled. The heat dispersion is reduced by means of three concentric tantalum screens located around the sample inside the chamber.

The sample is placed at the center of the vacuum chamber and heated by a graphite foil by means of radiation heating.

The sample temperature is measured by means of two different thermocouples (type B and S) placed near the sample and near the heating element. The sample was polished with hydrofluoric acid before the experiment to reduce the risk of devitrification as suggested in the literature.

The dynamic structure factor is measured at the two temperatures  $T=1570$  K and  $T=920$  K.

These values are chosen in order to span a wide temperature range from deeply below to above the glass transition temperature,  $T_g \sim 1450$  K.

The spectra are collected on the Stokes side in view of the necessity to cover a wide dynamical range, up to 100 meV, to properly observe the dispersion of the longitudinal excitations up to the border of the first pseudo-Brillouin zone.

Only a small portion of the anti-Stokes part is measured to properly describe the elastic line. To achieve a good signal to noise ratio each spectrum is collected with a long integration time of around 15 minutes per point. In particular the spectra are collected over an overall time of around 45 and 30 hours at 920 K and 1570 K respectively.

This long integration time is necessary because vitreous silica is characterized by a low inelastic to elastic intensity ratio due to its low fragility. Moreover the inelastic intensity on the longitudinal modes tends to lower at increasing  $Q$  values because of the broadening of the excitations, requiring an increased statistical accuracy.

The sample didn't present any presence of crystallization during the experiment. The absence of devitrification as been checked by measuring the static structure factor  $S(Q)$  at regular intervals during the experiment.

A selection of spectra at the two measured temperatures and at different wave vectors is shown in figure 1 in logarithmic scale in the y axis.

The good signal to noise ratio can be appreciated by the smoothness of the points in the tail of the spectrum, where the count rate is of the order of a few counts per minute.

The spectra in figure 1 are presented together with the instrumental response function and the best fitting function.

The inelastic part of the spectrum is characterized by the presence of two excitations. The first excitation is almost non dispersing and situated near the elastic line, while the second mode presents an evident dispersion and is well detectable over the entire explored wave vector range.

The two excitations are described by two damped harmonic oscillators convoluted with the instrumental resolution function, which has been measured on a plexiglass sample.

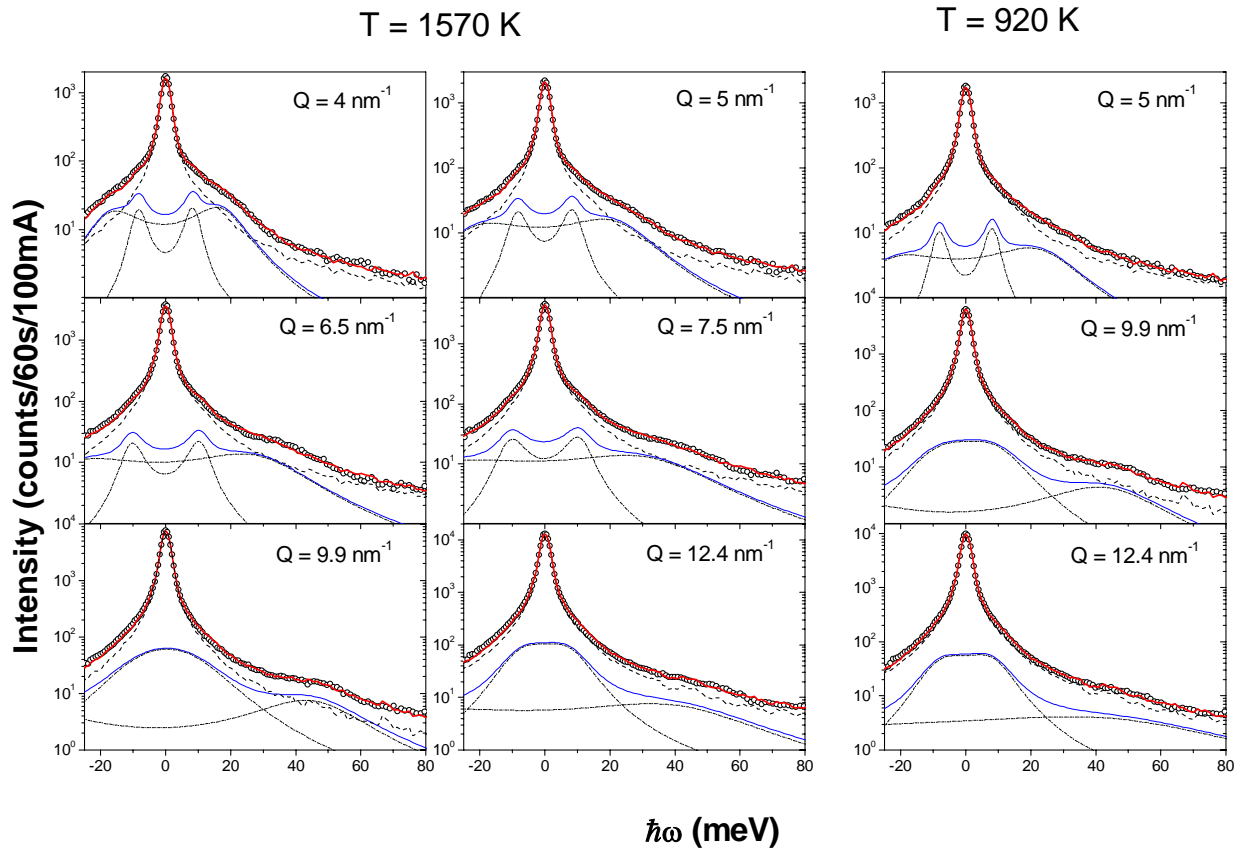


Figure 1: Selection of IXS spectra at the indicated exchanged wave vectors at the two measured temperatures. The spectra are plotted together with the best fitting function (red) with two excitations (tick continuous line) and its main components: the elastic line (dashed), the inelastic contribution (blue, continuous) and the two inelastic excitations (dash-dotted). The baseline  $y_0$  lies below the plotted window.