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High frequency acoustic attenuation of vitreous silica: New insight from inelastic x-ray scattering

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ABSTRACT

We present new experimental results on the propagation and damping of the high frequency acoustic-like modes in vitreous silica. The new data are measured by means of the inelastic x-ray scattering technique down to an exchanged wavevector $Q \sim 0.9\ \mathrm{nm}^{-1}$, at the limit of the instrument capabilities. Thanks to the continuous development of the technique, the new spectra are characterized by a very high signal to noise ratio when compared to previous experiments. The higher data quality finally allows for a reliable determination of the position and width of the inelastic excitations. The new data show that the sound damping Γ is marked by a frequency dependence compatible with the Rayleigh law, $\Gamma \sim \nu^4$, for frequencies below the position of the excess vibrational modes at the boson peak. We show that the new data are in good agreement with estimates of the acoustic mean free path from the thermal conductivity, which take into account the peculiar plateau at a few Kelvin. The connection between the boson peak and the Rayleigh law is further confirmed by a comparison of the present data with literature data for the sound attenuation in a permanently densified silica sample.

1. Introduction

The thermal conductivity of amorphous materials is many orders of magnitude smaller than that of the corresponding crystalline structures and presents a peculiar plateau at temperatures of a few Kelvin [1], whose nature is still debated in the literature [2–5]. In crystals heat is carried by phonons, which are quantized vibrational excitations. Phonon-like excitations also exist in glasses up to the terahertz frequency regime, but they are strongly damped [6]. The existence of a plateau in the thermal conductivity of glasses requires the mean free path of the propagating phonon-like excitations to decrease at least with the fourth power of frequency [1]. This strong scattering regime is predicted by substantially different theoretical models for the vibrations in disordered solids [7–10]. Discriminating among them, however, is difficult since the experimental access to this regime is very challenging. Also in the prototypical case of the silica glass the overall picture is not settled [11,12].

The presence of localized impurities in an elastic medium gives rise to a sound attenuation proportional to the fourth power of frequency, known as Rayleigh scattering [13,14]. This strong increase of the sound attenuation with frequency is characteristic of the amorphous state as suggested by various models for the vibrations in disordered systems. It is found, for instance, in models where the atoms vibrate either on a lattice with random elastic constants [8] or off-lattice around a topologically disordered configuration [9]. The same frequency dependence is also predicted by the soft potential model as a resonant interaction between vibrational soft modes and sound waves [7]. In all of these models the acoustic wave damping follows this Rayleigh-like law at frequencies below the boson peak (BP) position [15].

In addition to Rayleigh scattering, the sound waves in glasses can be attenuated by a variety of other attenuation mechanisms, all associated with some degree of anharmonicity [16,17]. These mechanisms give rise to a complex frequency and temperature dependence of the sound attenuation, which is a matter of active research. The most difficult frequency range to explore is the one between tens of GHz and a few THz, above the range probed by Brillouin light scattering (BLS). Controversial results have been reported on the frequency dependence of the sound attenuation at frequencies of some hundreds of GHz. A Brillouin ultraviolet scattering experiment performed on a bulk sample, showed evidence of the onset of the Rayleigh scattering regime at 150 GHz [11]. The observation of this strong scattering regime at such low frequency would exclude any connection with the BP, which is found at much higher frequencies of ~1 THz [15]. A more recent work reported a contrasting result, showing a much smoother frequency dependence of the sound attenuation up to 300 GHz [12]. In this case, however, the measurements were performed on thin films, leaving some doubts on whether they can be considered valid for the bulk sample as well.

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Exploiting the intense development of the inelastic x-ray scattering (IXS) technique during the last decade, we have obtained clear evidences that in vitreous silica this strong scattering regime is located in the few terahertz frequency range [18]. Here we further discuss these results and we present a comparison between the measured damping and that estimated from thermal conductivity data. Furthermore we compare the present results with literature data on a densified vitreous silica sample [19,20]. In both systems the Rayleigh scattering regime persists up to a crossover frequency which coincides with the position of the boson peak, confirming a clear connection between the damping of the acoustic-like excitations and the excess of vibrational modes.

2. Experiment and data analysis

The experiment has been performed at the IXS beamline ID16 of the European Synchrotron Radiation Facility in Grenoble, France. A high energy resolution of 1.4 meV is reached at an incident energy of 23.7 keV. The Q-resolution is fixed to 0.3 nm^{-1}. In the relevant wave-vector range ($Q<4\ \text{nm}^{-1}$) two spectra are collected simultaneously, using two of the nine available analyzer crystals. The sample is a suprasil fused-quartz cylinder[1] with a thickness of 3.5 mm to match the photoelectric absorption length of SiO_2 at the chosen incident energy. The experiment is performed at a temperature $T=1620$ K, i.e. in the supercooled liquid phase, in order to enhance the inelastic signal over the tails of the elastic line. Furthermore at this high temperature the BP position is 50% higher in frequency with respect to room temperature. This allows us to measure the Brillouin peaks at frequencies below the BP position, a region almost inaccessible at lower temperatures. The absence of crystallization has been checked regularly during the experiment by measuring the static structure factor.

The dynamic structure factor is modeled as the sum of a delta function to describe the elastic line plus a damped harmonic oscillator (DHO) for the inelastic contribution [21]:

$$\frac{S(Q,\omega)}{S(Q)} = f(Q)\delta(\omega) + (1-f(Q))\frac{1}{\pi}\frac{\Omega^2(Q)\Gamma(Q)}{\left[\omega^2-\Omega^2(Q)\right]^2+\omega^2\Gamma^2(Q)}. \qquad (1)$$

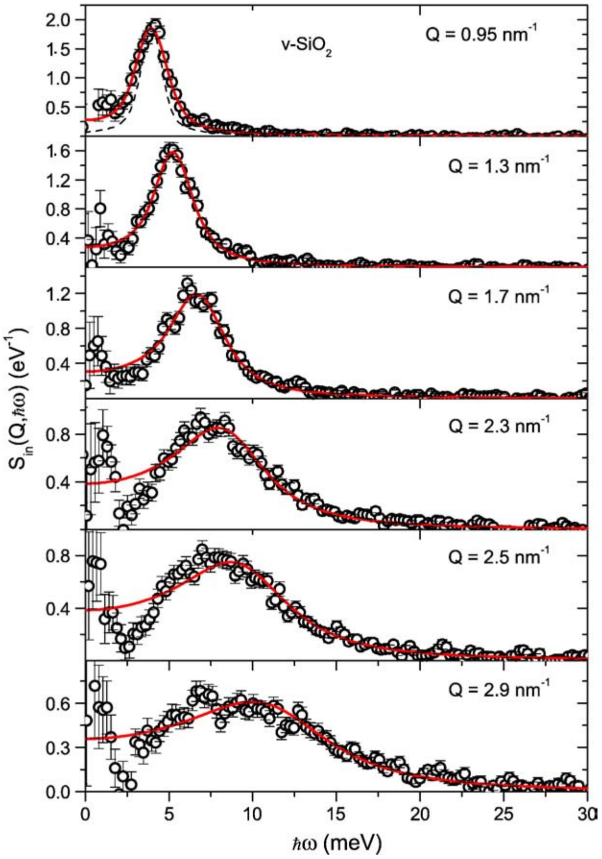

Fig. 1. Inelastic part of the dynamic structure factor of normal vitreous silica at temperature $T=1620$ K, measured with IXS at the indicated wave-vector Q. The continuous line (red) is the best fitting function, a damped harmonic oscillator convoluted with the instrument resolution. The instrument response function is plotted in the upper panel as a dashed line, to show the broadening of the peak with respect to the resolution.

The parameters $\Omega(Q)$ and $\Gamma(Q)$ represent the position and width (F.W.H.M.) of the inelastic peaks in the measured spectra, $S(Q)$ is the static structure factor and $f(Q)$ is the non ergodicity parameter. The measured intensity is fitted to the convolution of the model function defined by Eq. (1) with the instrument response function times an intensity parameter. A baseline is also added to account for the background noise. The fit takes into account also the effect of the finite Q-resolution of the experiment which induces an additional broadening of the peaks, more relevant at small wave-vectors. The fitting procedure is performed by minimizing the χ^2 function in the parameter space. The Stokes side of the inelastic part of the dynamic structure factor, $S_{in}(Q,\omega)$ is plotted in Fig. 1 for a selection of exchanged wave-vectors Q. $S_{in}(Q,\omega)$ is obtained by subtracting the elastic line obtained in the fitting procedure from the measured intensity. The normalization to absolute units is obtained by comparing the measured intensity with literature data for the static structure factor of vitreous silica [22]. Each spectrum has been integrated for 12 h to reach the final signal to noise ratio. The broadening of the peak over the instrument resolution width is measurable in the whole explored Q range, and strongly increases with increasing Q. The instrument resolution is plotted in the upper panel of Fig. 1 (dashed line), for reference. Increasing Q above 2 nm^{-1}, the DHO fails to properly describe the spectrum, in particular around $\omega=0$ [23]. Nevertheless the width of the peak is still well described by the DHO model.

3. Results

Fig. 2 shows the sound attenuation coefficient as a function of frequency in log–log scale. Below a crossover frequency $\nu_c\sim1.5$ THz the sound attenuation is compatible with the Rayleigh law, $\Gamma\sim\nu^4$. Above ν_c the attenuation follows a quadratic frequency dependence. The figure also shows the comparison with a previous IXS experiment on silica, performed more than a decade ago [24]. The old experiment is affected by error bars which are more than a factor ten higher than that of the new data. This improvement is due to a slight increase in the energy resolution of the IXS spectrometer and mainly to the increase in the flux and stability of the beamline. The two data sets are in good agreement only above the crossover frequency ν_c. In the low frequency range the old data set turns out not to be reliable enough, likely because of the low signal to noise ratio. These experimental limitations prevented the observation of the Rayleigh law in that early IXS experiment.

From Fig. 2 we can also estimate the Ioffe–Regel (IR) frequency ν_{IR}, defined by the condition that the acoustic excitation decays in half of the oscillation period, i.e. by the relation $\Gamma=\omega/\pi$ [23,25]. The IR frequency is determined by the intersection of the data with the dashed line in the figure and is found to be close to the crossover frequency ν_c. This frequency also coincides with the position of the Boson Peak, ν_{BP}, at this temperature, as determined by neutron

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[1] Sample purchased from Goodfellow.