| | **Experiment title:** Study of line shapes near the crossover to strong phonon scattering in densified silica glass | **Experiment number:** HS-2062 |
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| **Beamline:** BL21-ID16 | **Date of experiment:** from: 10 March 2003 to: 19 March 2003 | **Date of report:** September 2, 2003 |
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Report:

The nature of the vibrations in glasses at frequencies in the THz range is a question of current debate. A central issue is whether acoustic phonons experience *strong scattering* above a crossover frequency $\Omega_{\mathrm{co}}$, leading to inhomogeneous phonon lineshapes. We did find in previous ID16 experiments a clear crossover phenomenon on a densified form of silica, $d$-$SiO_2$ [1]. In that case, $\Omega_{\mathrm{co}} \simeq 9$ meV and the corresponding crossover wavevector $q_{\mathrm{co}}$ is around 2 $\mathrm{nm}^{-1}$. It was also observed that above $q_{\mathrm{co}}$ the phonon lineshape starts to deviate from the damped harmonic oscillator (DHO) model. The aim of the experiment reported here was to measure IXS spectra with high S/N ratio just above this $q_{\mathrm{co}}$ to study in detail the inelastic lineshape modifications and its relation with the low-frequency modes forming the boson peak (BP).

In a first step, the IXS spectra have been adjusted to the usual DHO model plus an elastic peak, convoluted with the experimental resolution function. Account is taken of the broadening caused by the finite aperture of the analyzers. Fig. 1-a shows the inelastic spectra, after subtraction of the freely adjusted elastic contribution, at $Q$-values ranging from 2 to 3 $\mathrm{nm}^{-1}$ and accumulated on analyzer 1 for $\simeq$ 24 hours. The bottom spectrum at 5.4 $\mathrm{nm}^{-1}$ is the sum of data accumulated on analyser 2 at 5, 5.2, 5.4, 5.7 and 6 $\mathrm{nm}^{-1}$. This average is justified as the $Q$-dependence is very weak at such large $Q$-values. The solid lines are the DHO fits. The dotted lines that nearly coincide with the x-axis show the measured background. It clearly appears from Fig. 1-a that the standard DHO line shape is unable to describe accurately the measured data above $q_{\mathrm{co}}$: the higher is $Q$, the more the DHO fails to reproduce the strong asymmetric broadening of the spectral lines. This obsviously poses the question of the physical meaning of parameters extracted from such fits.

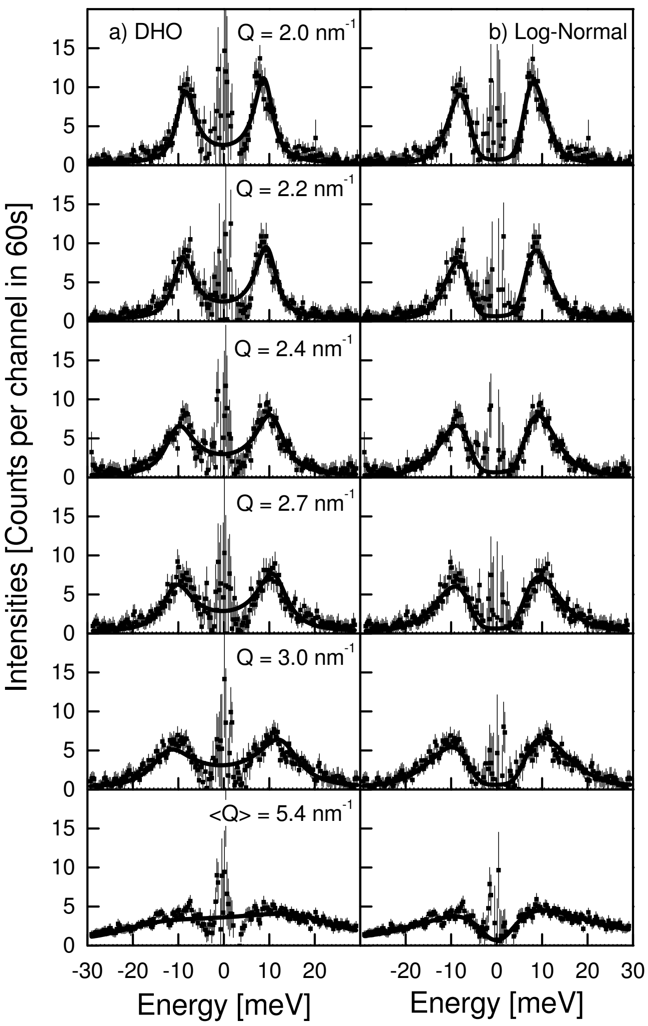
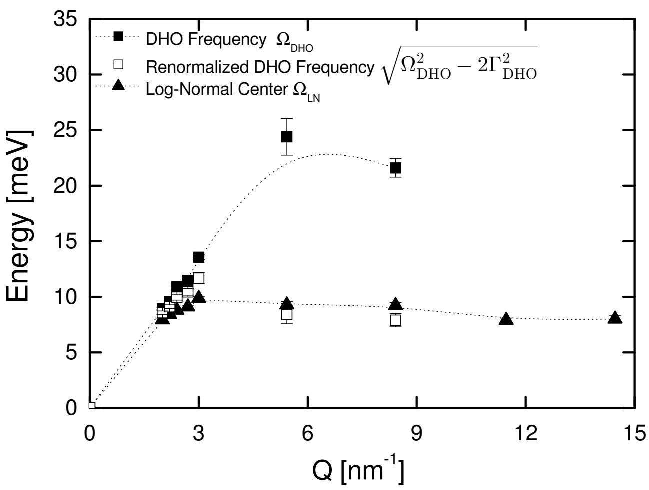



Fig. 1 : fits of the inelastic spectra after subtraction of the freely adjusted elastic contribution at $Q$-values ranging from 2 nm$^{-1}$ to 3 nm$^{-1}$. The bottom spectra at 5.4 nm$^{-1}$ are the sum of data accumulated on analyser 2 at 5, 5.2, 5.4, 5.7 and 6 nm$^{-1}$. (a) DHO fits. (b) Log-Normal fits.

Fig. 2 : DHO frequencies $\Omega_{\rm DHO}$ (solid squares) and Log-Normal peak positions $\Omega_{\rm LN}$ (solid triangles) extracted from the fits of Fig. 1. Also shown are the peak positions of the DHO fits given by $\sqrt{\Omega^2_{\rm DHO} - 2\Gamma^2_{\rm DHO}}$ (empty squares), where $\Gamma_{\rm DHO}$ is the half-width of the DHO function.




It also strongly indicates that inhomogeneous broadening rather than anharmonicty dominates the spectral width of the very high frequency acoustic-like excitations in $d$-$SiO_2$ [1]. To obtain better insight into the phonon-lineshape evolution in the crossover region, we used in a second step the well-known Log-Normal function. Although devoid at present of a firm theoritical basis, this choice takes advantage of a suitable description of the growing asymmetry of the measured spectral lines above the crossover as shown in Fig. 1-b. It is interesting to note that the $\chi^2$ obtained in this case are 15 to 35% better than those obtained with the DHO model. A second feature of the Log-Normal function is that the fitted frequency parameter $\Omega_{\rm LN}$ describes the peak position of the inelastic spectrum. Fig. 2 shows the $\Omega_{\rm LN}$ frequencies resulting from these fits for all the available $Q$'s. One clearly observes that the experimental peak position does not really shift beyond the BP frequency, which is 8.5 meV in $d$-$SiO_2$. For comparison, Fig. 2 also shows the DHO frequencies $\Omega_{\rm DHO}$ as well as the peak positions of the DHO fits given by $\sqrt{\Omega^2_{\rm DHO} - 2\Gamma^2_{\rm DHO}}$.

Our results reveal that around the crossover in $d$-$SiO_2$, the acoustic excitations strongly broaden with an asymmetric spectral lineshape growing with $Q$ and deviating from the DHO. Above the crossover, a much better description of the inelastic signal is achieved with the Log-Normal function. Its peak position is always close to the frequency maximum of the BP. These data call for a theoretical description of the interaction (hybridization) of acoustic excitation with the BP ones. This is currently in progress.

[1] B. Rufflé *et al.*, Phys. Rev. Lett. **90**, 095502 (2003); E. Courtens *et al.*, J. Phys. Condens. Matter **15**, S1279 (2003).