High-frequency propagating modes in vitreous silica at 295 K

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Collective modes in vitreous silica (v-SiO_2) are found at 295 K and at momentum transfers $Q=1.5$ and 2.0 nm^{-1}. They propagate with a sound velocity of $\approx$ 6000 m/s, a value equivalent to that in the $Q\rightarrow 0$ limit, and to that recently measured at 1050 K in the 1–3.5 nm^{-1} Q range. The data complete the scenario on the acousticlike dynamics in v-SiO_2, showing its temperature independence and propagating rather than localized nature in the whole 295–1050 K and 0.01–2.0 nm^{-1} temperature and Q ranges. [S0163-1829(97)05113-8]

The recent development of inelastic x-ray scattering (IXS) with meV energy resolution has opened the possibility of beginning detailed studies of the high-frequency collective dynamics in disordered systems.[1–4] The determination of the dynamic structure factor $S(Q,E)$ at momentum transfers Q in the 1–20 nm^{-1} region with unconstrained energy transfer E gives access to the atomic density fluctuations spectrum in the microscopic limit, and therefore to the investigation of the solidlike behavior of systems without translational invariance in a time domain that is short compared to any macroscopic structural relaxation process.

Important domains of the application of IXS are the study of glasses,[3,5,6] liquids,[1,2,4] glass formers, and disordered solids.[7] In the specific case of v-SiO_2, the textbook example of a strong network forming glass as well as an extremely important system for its many technological applications,[8] a recent IXS measurement shows the presence of collective modes propagating up to $Q=3.5$ nm^{-1} with a velocity of sound of 5800 m/s.[5] These measurements were performed at high temperature, namely at $T=1050$ K, to enhance the weak inelastically scattered signal with respect to the intense elastic line due to the static structural disorder. Despite the high temperature, although still $\approx$400 K smaller than the glass transition temperature T_g, the peak positions and the energy widths of the modes measured by IXS in the 1–3.5 nm^{-1} Q region were found to match extremely well the extrapolation to large Q of the Brillouin light-scattering[9,5] (BLS) and picosecond optical technique[10] (POT) determinations made in v-SiO_2 at room temperature (RT). Namely, the energy position (energy width) found by IXS at 1050 K quantitatively follows the linear (quadratic) Q dependence found at 295 K by BLS and POT at Q smaller than 0.2 nm^{-1}. The striking temperature independence of the energy widths in such a wide temperature region is not obvious. It seems to indicate a direct relation to the structural disorder, which is also temperature independent. Moreover, the linear Q dependence of the energy positions of the modes implies that in v-SiO_2 the sound velocity does not change in the whole 295–1050 K temperature and 0.01–3.5 nm^{-1} momentum regions: this would demonstrate the absence of relaxation processes in this strong glass. This idea, based on the temperature independence of collective dynamics, is not

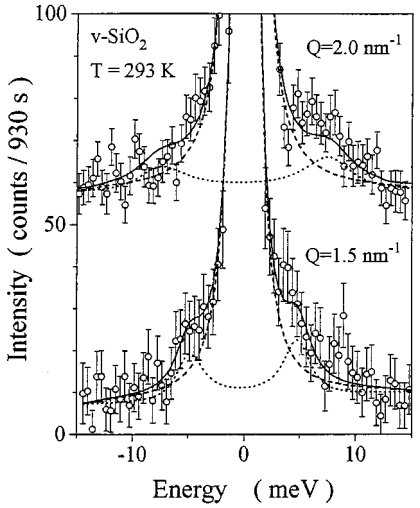

FIG. 1. Inelastic x-ray scattering spectra of v-SiO_2 at $T=295$ K, measured at $Q=1.5$ nm^{-1} (lower panel) and $Q=2$ nm^{-1} (upper panel). To better indicate the presence of the inelastic contribution, the vertical scale has been chosen to show the tails of the central peak. The peak intensity is 630 counts/930 s in both spectra. The dashed line is the experimentally determined instrumental resolution function, which is shown aligned and scaled to the central peak to emphasize the presence of inelastic scattering. The full line is the best fit to the data as discussed in the text. The dotted line represents the inelastic contribution to the fit.

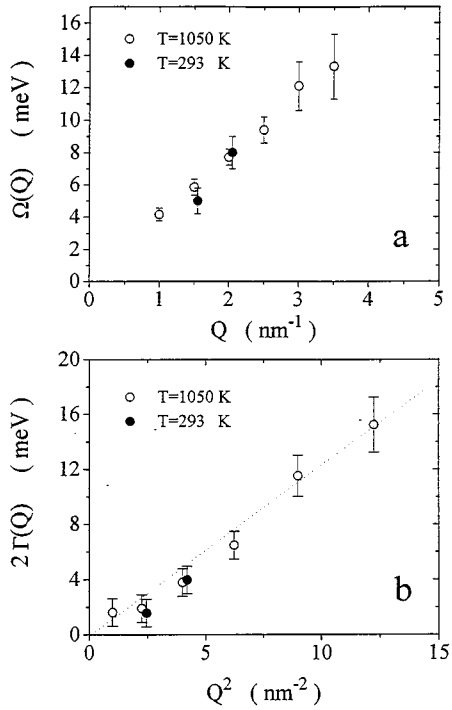

FIG. 2. Excitations energy, $\Omega(Q)$ (a) and full width at half maximum $2\Gamma(Q)$ (b) of the excitations as derived from the fits. The full symbols (●) refer to the 295 K data, and the open symbols (○) to the high-temperature ($T=1050$ K) data. The dashed line corresponds to the extrapolation of the energy widths found by BLS (Ref. 9) and POT (Ref. 10) at low Q values.

in contrast with the inelastic neutron-scattering measurements of Buchenau *et al.*,[11] who showed that the dynamics of v-SiO_2 at room temperature and at energy above 2 meV and Q above 10 nm^{-1} is predominantly harmonic. To settle this issue, however, it is necessary to perform a direct measurement of the collective dynamics of v-SiO_2 at room temperature and in the high-Q region.

In this paper we report the determination of $S(Q,E)$ in v-SiO_2 by IXS at $T=295$ K at the two Q values of 2 and 1.5 nm^{-1}. This was possible thanks to an energy resolution improvement with respect to the previous study,[5] leading to an improved contrast between the inelastic and the elastic signal. We find a *propagating* collective mode which, at the two examined Q values, has energies and widths equivalent to those found at $T=1050$ K, thus confirming the present understanding of the high-frequency dynamics in v-SiO_2. Namely, it shows the quasiharmonic behavior even at RT,[11] the absence of localization of the modes for Q values at least as large as 2 nm^{-1},[5] and, consequently, the absence of relaxation processes in v-SiO_2 in the considered temperature and momentum ranges.

The IXS experiment was carried out at the very high-energy resolution inelastic x-ray-scattering beamline (BL21-ID16) at the European Synchrotron Radiation Facility. This instrument is based on backscattering from high-order reflections in perfect silicon crystals, and the spectra reported here were measured using the Si(11 11 11) reflection at $\approx$ 21.748 keV. The total instrumental resolution function was measured using a plexiglass scatterer at the maximum of its static structure factor ($Q=10$ nm^{-1}), where the scattering is dominated by the elastic component; the energy resolution, full width at half maximum (fwhm), was 1.5 $\pm$ 0.1 meV. The momentum transfer was selected at 1.5 and 2.0 nm^{-1}, and the Q resolution was set to 0.3 nm^{-1} fwhm by a slit in front of the analyzer crystal. Energy scans were performed by varying the relative temperature between the monochromator and analyzer crystals by ± 0.45 K with a step of 0.0075 K. Each scan took about 120 min, and each Q value was obtained by averaging 15 scans (the total integration time was 930 s per point). The data were normalized to the intensity of the incident beam. Further details on the IXS beamline are reported elsewhere.[12–14] The SiO_2 suprasil sample, purchased from Goodfellow, was the same 2 mm diameter rod used in Ref. 5.

The two inelastic x-ray-scattering spectra are reported in Fig. 1 at the indicated Q values, together with the fits to be discussed in the following. The spectra are shown on an enlarged vertical scale to emphasize the weak inelastic scattering signal. The peak intensity of the elastic line at zero energy transfer is 630 counts/930 s. The inelastic scattering signal, with a peak intensity of $\approx$10 counts/930 s, is clearly visible over the wings of the dashed line, which corresponds to the experimental resolution function aligned and scaled to the elastic peak. Already from the raw data, it is possible to see that the inelastic signal is dispersing with Q, and that it moves towards higher energy transfers going from 1.5 to 2 nm^{-1}, thus indicating the propagating nature of these excitations. Similar to Ref. 5, the energy position and width of these excitations is derived by a fitting procedure of the IXS data, obtained by the convolution of the experimental resolution function with the model function $F(Q,E)$ given by

$$F(Q,E)=I_0(Q)\,\delta(E)+[n(E)+1]\times I(Q)\frac{E\Gamma(Q)\Omega(Q)}{[\Omega(Q)^2-E^2]^2+\Gamma(Q)^2E^2}. \quad (1)$$

It consists of a δ function to account for the elastic scattering, and a damped harmonic oscillator model[15] for the two side peaks. $I_0(Q)$ and $I(Q)$ are related to the intensities of the central peak and of the inelastic contributions, respectively, $\Omega(Q)$ and $\Gamma(Q)$ are the excitation energy and width, and $n(E)$ is the Bose factor. A standard χ^2 minimization procedure has been utilized to determine the fitting parameters.

The two values of $\Omega(Q)$ and $\Gamma(Q)$ obtained from the fit are reported in Figs. 2(a) and 2(b), respectively. They are both shown together with the IXS measurements at 1050 K. In Fig. 2(b), the dashed line refers to the extrapolation of the energy widths measured at RT and at low Q using the BLS and POT techniques. The agreement between the two sets of measurements is excellent. The absence of relevant shifts in the energy positions implies that the modes found at 295 K propagate with the same velocity as those found at high temperature. Similarly, the equivalence of the energy widths, and their consistency with the extrapolation of the low-Q measurements, confirm the Q^2 dependence of $\Gamma(Q)$ in the whole 0.01–2 nm^{-1} range.

In conclusion, the equivalence, within the error bars, of $\Omega(Q)$ and $\Gamma(Q)$ in the 295–1050 K range confirms the temperature independence of the collective dynamics in v-SiO_2, and the existence, even at room temperature, of a propagating collective dynamics at least up to the Q value of 2 nm^{-1}. In particular, it shows that up to the energies of the modes reported here, extending to the peak value of 8 meV, these excitations are not localized but quasiharmonic. Moreover, since their energy is comparable to the density of states feature referred to as the *boson* peak,[16] these results show that even at room temperature the boson peak must have a contribution from the propagating collective dynamics reported here.

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