| ESRF | **Experiment title:** Studies on the "boson peak" in **silica** xerogels at different stages of densification by ther-ma1 treatments | **Experiment number:** HS-516 |
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## Report:

The aim of the experiment HS-516 was to assess whether the acoustic-like excitations previously ob served in vitreous silica [l] are present in low-density, $SiO_2$ based materials as the silica xerogel.

In **a first set of** measurements, we determined the static structure factor (S(Q)) of xerogles with **different degrees of** thermal treatment (namely 500, 860 and 900 C) and compare them with the S(Q) of pure silica. The data, obtained pasting different Q-resolution and Q-range scans, are reliable in the $Q$=0.1 ÷ 30 $nm^{-1}$ region. As can be observed in Fig. 1, the static structure factors of xerogels shows the presence of a strong scattering at low Q, whose intensity decreases with increasing the thermal treatment temperature and disappears in pure silica. This scattering is peaked at Q ≈ 0.3 $nm^{-1}$ and can be ascribed to the presence of voids (channels) of rather well defined dimension in these systems. **The fast decay of the scattering** intensity at high Q (between 1 and 10 $nm^{-1}$) follows a $Q^{-4}$ law. This law is in agreement with what it is expected from surface scattering, and shows the absence of any fractal effect.

In vitreous silica the inelastic excitations were observed in the 1÷4 $nm^{-1}$ region. The sound velocity in these xerogels is ≈ 10% ÷ 20 % lower than in silica, and, therefore, we expect that the excitations in the xerogel samples should appear in the same Q region of v – $SiO_2$. However, the elastic-&inelastic **intensity ratio** in pure silica was very high, and only measurements made at high temperature allowed us to detect a weak inelastic signal. As can be seen in Fig. 1, in the Q region of interest (1÷4 $nm^{-1}$), the elastic-to-inelastic intensity ratio in xerogels is at least a factor 10 worse than in the case of pure silica This observation, together with the impossibility to make measurements at high temperature (T cannot in any case exceed the thermal treatment temperature), lead us to conclude on the impossibility to detect an inelastic signal with a good statistic in the available beam time.

Differently from the low Q region, in Fig. 1 one can observe that in the region of the sharp diffraction peak the presence of voids in the xerogels induces only minor modifications on the microscopic structure: the peak position is not modified while its width seems to depend from the specific heat treatment.

Motivated by recent neutron scattering result, we decided to investigate the high Q region (**Q** ≥ 15 $nm^{-1}$). Recently Buchenau et al [2], using inelastic neutron scattering in kinematic conditions, have measured a constant energy (E) cut of the dynamic structure factor ($S(Q,E)$ of vitreous silica as a function of Q. The selected energy corresponded to the energy of the boson peak (E = 5 meV) and the spectrum showed a peak at Q ≈ 30 $nm^{-1}$. A similar structure has been found also in the present experiment. In particular we measured: *i)* Different energy scans at constant Q (=17, 20, 23, 26 and 29 $nm^{-1}$) in pure silica and two different xerogel samples ($T_t$ = 860 and 900 C); and ii) A Q scan at constant energy (E = 5 meV) in the pure silica sample.

In the constant Q scans we were able to observe an inelastic signal at about 5 meV with a tail extending up to ≈ 15 meV. Its shape is nearly Q independent. Its intensity, on the contrary, is found to have a marked Q dependence, as also best shown by the constant E scan reported in Fig. 2.

At the present, preliminary, level of analysis, no differences are found between the inelastic signal detected in pure silica and in the xerogels. More anlaysis and much better statistics are needed to further clarify this point.

In Fig. 2 one can observe a structured $S(Q,E)$, similar to what observed in neutron scattering. However, Important quantitative differences are present between the IXS and INS results. In particular, in our IXS measurements we find: i) a bump around Q=23 $nm^{-1}$ (at variance to INS, where the peak is observed at Q ≈ 28 $nm^{-1}$), and ii) that the peak-to-background contrast is much lower than in INS. These differences can be explained on the basis of the different way in which neutrons and x-rays couple to the silicon and oxygen atoms: while the x-rays mainly scatter from silicon, the neutrons scatter with similar strength from either the silicon or the oxygen nuclei. This result, therefore, provides a very nice example on the complementarity of the two techniques. In the present case, this comparison allows one to identify the specific atoms involved in the observed vibrational modes. More importantly, using its capability to study the acoustic excitations in kinematical condition, the IXS technique can access also the low Q region, allowing the simulaneous detection of the Brillouin and the high Q peaks in a **constant *E*** cut **of the S(Q, *E*). A trace** of the Brillouin peak can be seen in Fig. 2 at low Q, consistently with the previous observations that at ***E*** = 5 meV, the peak in Q is expected at ≈1.5 $nm^{-1}$.


References

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