A.

ESRF	**Experiment title:** Search for phonon-Rayleigh scattering in x-ray Brillouin scattering from high-density silica.	**Experiment number:** HS-660
Beamline: BL21-ID16	**Date of experiment:** from: 16/09/98 at 15:00 to: 22/09/98 at 07:00	**Date of report:** February 24, 1999
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The nature of the vibrations in glasses at frequencies ω near the end of acoustic branches is a question of considerable interest and actuality, both theoretically [1] and experimentally, owing in part to new measuring possibilities at large instruments. A central prediction is that at sufficiently high ω, phonons should crossover to a regime where they become strongly scattered by intrinsic inhomogeneities, so that they cease to propagate as plane waves.[1] The universal low temperature (T) plateau observed in the thermal conductivity κ of glasses is generally attributed to such an effect.[2] However, until now, it had not been possible to clearly identify the existence of a frequency crossover, ω_{co}, in the inelastic x-ray spectra of acoustic phonons in glasses. On ID16, a large quantity of data has been accumulated which has been mostly analyzed as if this crossover did not exist.[3] We are convinced that the reason for the difficulty is that in most glasses the crossover occurs in a region of ω, and/or scattering vector, Q, that lies either outside or nearly outside the accessible range of spectrometers. It is to circumvent this problem that we undertook an investigation of permanently densified silica glass, d-SiO_2. At a density ρ = 2.62 g/cm^3, this material ought to be much more homogeneous at the relevant length scale than usual silica glass, v-SiO_2, for which ρ = 2.20 g/cm^3. An indication for this is that the plateau in κ is located at much higher T in d-SiO_2 than in v-SiO_2.[4] Thus, we predicted that ω_{co} should be sufficiently high in d-SiO_2 to become well accessible to measurements on ID16. The results described below and in [5] vindicate this forecast.

Fig. 1 shows a typical spectrum obtained after ~20 hours of accumulation time in d-SiO_2 at 575 K, and for Q = 2.5 nm^{-1}. The instrument background is subtracted from all spectra displayed in this

report. In Fig.1, the continuous line is a spline fit through the instrumental function matched to the spectrum peak. In spite of the smallness of the inelastic signal, whose strength corresponds to the prediction on the basis of the known elastic properties, one clearly sees an inelastic contribution in the wings. In Fig. 2, the spectra have been folded to improve the visibility, and the elastic central peak has been subtracted using a fitting procedure described in [5]. For spectra (a) to (c) one can adjust the inelastic part to several models, in particular to a lorentzian or to a damped harmonic oscillator (DHO). Up to $Q = 2$ nm^{-1} the spectral width is too small to be extracted. The width suddenly increases at $Q = 2.5$ nm^{-1}, and at 4 nm^{-1} the spectrum becomes very broad. Interestingly, it cannot be adjusted to a DHO, as shown by the best fit to a DHO displayed in Fig. 2(f). The reason is that there is no spectral intensity near ω=0. The DHO is a model implying *lifetime broadening* and so, for broad lines, it gives a large intensity at ω=0. The solid lines in Fig. 2 (a-e) result from a model implying *disorder broadening*, which starts from 0 at ω=0, and with $\omega_{co} \approx 9$ meV. The latter value agrees remarkably with a plateau in κ near 20 K, as observed.

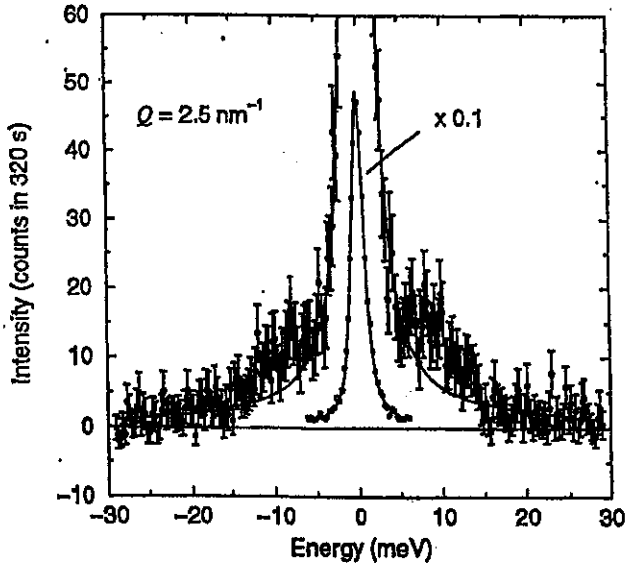

Figure 1. A typical x-ray Brillouin spectrum of *d*-SiO_2 (Stokes on the right).

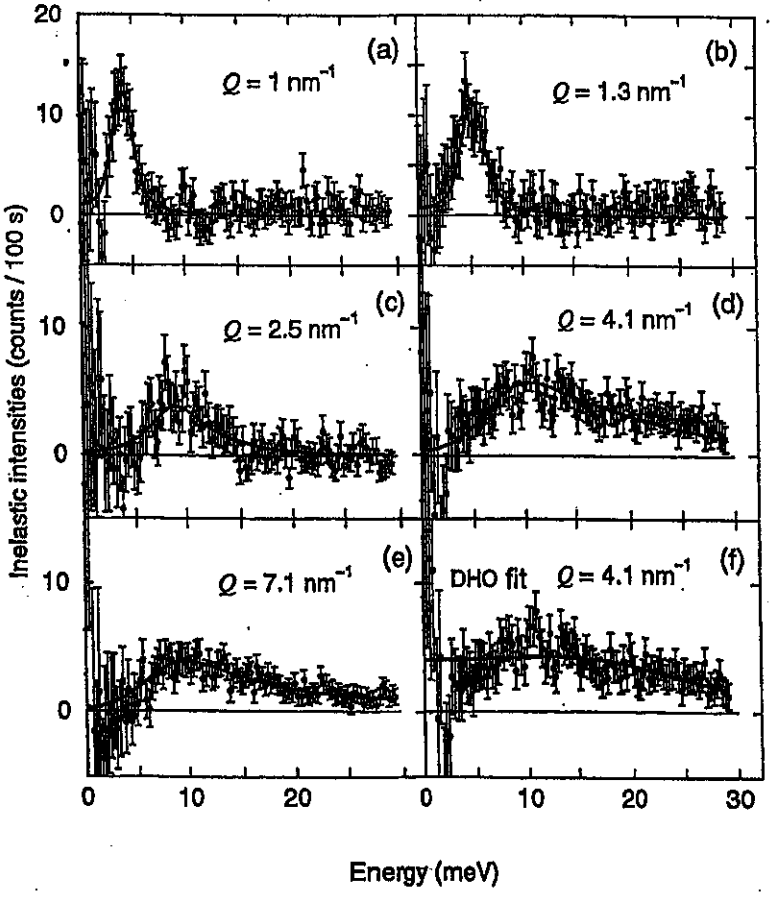

Figure 2. Examples of a few inelastic contributions to folded spectra. The lines in (a-e) are from fits with a crossover function as explained in [5]. For comparison, (f) is for the same spectrum as in (d), but fitting to the DHO model.

References

[1] see, *e.g.*, W. Schirmacher *et al.*, Phys. Rev. Lett. **81**, 136 (1998), and references therein.
[2] R.C. Zeller and R.O. Pohl, Phys. Rev. B **4**, 2029 (1971).
[3] see, *e.g.*, F. Sette et al., Science **280**, 1550 (1998), and references therein.
[4] Da-Ming Zhu, Phys. Rev. B **50**, 6053 (1994).
[5] E. Rat, M. Foret, E. Courtens, R. Vacher, M. Arai, and C. Masciovecchio, submitted to Phys. Rev. Lett..