Comment on ''Evidence of High Frequency Propagating Modes in Vitreous Silica''

In a recent Letter [1], Benassi *et al.* report an inelastic x-ray scattering (IXS) investigation of vitreous silica (v-SiO_2) at high temperature (T). They assume that the inelastic structure factor, $S(q, \omega)$, is a damped harmonic oscillator (DHO) response. From their fits, they conclude that high frequency acoustic modes *propagate* at frequencies ω well above the boson peak. Another recent Letter [2] reports combined inelastic neutron scattering (INS) and IXS data on v-SiO_2 at room T. These were analyzed with an $S(q, \omega)$ that implies strong acoustic-phonon scattering leading to a Ioffe-Regel crossover [3]. The crossover wave vector $q_{co} \cong 1\ \mathrm{nm}^{-1}$ and frequency $\omega_{co} \cong 4$ meV *are derived from INS spectra*, but agree with the (quite weak) IXS signals. The crossover energy corresponds to the boson peak. Although by themselves measurements of $S(q, \omega)$ cannot demonstrate localization [4], we feel that it is rather meaningful to investigate the applicability of the models.

Hence, we analyzed the data presented in Fig. 2 of [1] with the model and parameters of [2]. We simply allowed ω_{co} and the intensities to vary. Our excellent fits, with a mean $\chi^2 \simeq 1$, are illustrated in Fig. 1. As seen, the crossover model predicts a strong q dependence of the inelastic line shapes even for excitations that no longer propagate, for q up to a few times q_{co}. In view of the small inelastic contribution on top of the wings of the huge elastic peak, this dependence could easily be taken for a shift in a DHO peak frequency.

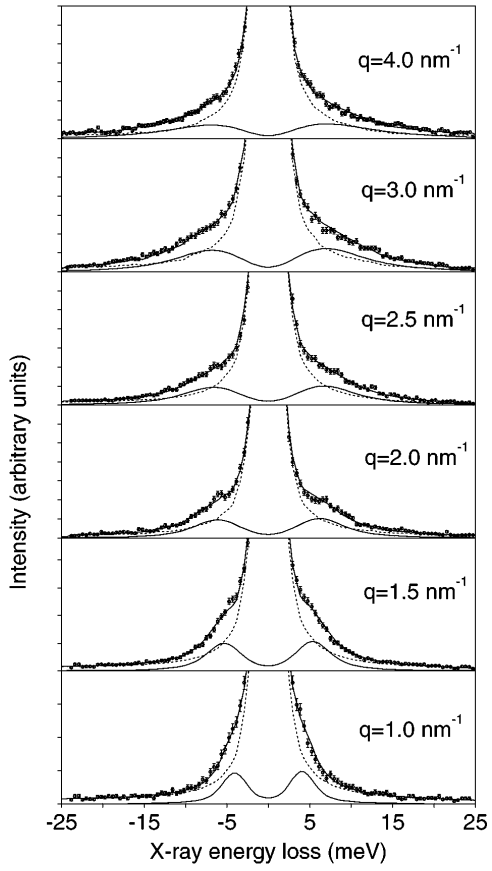

FIG. 1. IXS data taken from [1] (points) and their fits to the model of [2] (line) with $\omega_{co} = 6.1$ meV, or $q_{co} \simeq 1.5\ \mathrm{nm}^{-1}$. The calculated inelastic signal (solid line) and the elastic peak plus background (dashed line) are also shown separately.

The issue addressed here is not the likely significance of an increase of ω_{co} with T [5]. Our main point, illustrated in Fig. 1, is that the propagation of acoustic modes at ω beyond the boson peak of glasses is *not demonstrated* from available IXS data [1,5–7]. A corresponding remark of caution applies to the use of the DHO line shape at high ω, also for liquids [8]. The interpretation of the DHO in terms of *propagating* excitations [1,6–8] is arbitrary at this stage. The latter points rejoin one aspect of an earlier Comment on water [9]. Further progress of IXS, application to well selected test cases, and results from alternate spectroscopies such as INS, will presumably all be needed to clarify this central issue.

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M. Foret,[1] E. Courtens,[1] R. Vacher,[1] and J.-B. Suck[2]
[1]Laboratoire des Verres, UMR 5587 CNRS
Université de Montpellier II
F-34095 Montpellier Cedex 5, France
[2]Institut für Physik
T.U. Chemnitz-Zwickau
D-09107 Chemnitz, Germany

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