Merging of the acoustic branch with the boson peak in densified silica glass

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Both high-frequency acoustic modes and the boson peak related to SiO_4 librations are observed in a single inelastic x-ray scattering experiment. The experimental data are consistent with a picture where the acoustic modes experience a crossover at a frequency Ω_{co} beyond which plane waves cease to exist. The spectra evolve with the scattering vector to merge into a broad boson peak at $\Omega_{BP} \approx \Omega_{co}$. The latter, although essentially optic in nature, might hybridize with the resonant acousticlike modes, which can be crucial to their strong scattering.

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I. INTRODUCTION

The existence in glasses of an anomalous density of vibrational states (DOS) at low frequencies was recognized long ago.[1] Near 10 K, where modes in the terahertz range dominate thermal properties, the experimental specific heat can exceed by a large amount the one calculated from an extrapolation of the acoustic branches. The corresponding excess of modes produces a broad component in the DOS, as measured by inelastic neutron scattering (INS) at large *scattering* vectors **Q**.[2] This feature is called the ''boson peak'' (BP). It is also seen at optical wavelengths, e.g., in the Raman spectrum,[3] indicating that the excitations are quite dispersionless and suggesting that they might be ''molecularlike.'' The magnitude of this excess depends on the nature of the glass.[4] It is often large for glasses that are ''strong'' in the sense of Angell and Sichina,[5] such as covalent oxides. In contrast, only small excesses are observed in many fragile glasses, such as molecular or ionic ones. The general nature of these excess vibrations—acoustic vs optic—is still quite debated.[6–10] For vitreous silica, one of the strongest glasses that are known, it was recently confirmed by optical spectroscopy that the BP must relate to rigid SiO_4 librational motions,[9] as proposed long ago,[2] and as found in simulations.[11–13] In the permanently densified variety, d-SiO_2, the frequency of the BP maximum, Ω_{BP}, nearly coincides with what can reasonably be called ''the end of acoustic branches.''[14,9] There remains the important issue of the interactions of BP excitations with acoustic modes of the same frequency, as discussed, e.g., in Refs. 15 and 16. An experimental study of this relation is the main topic of the present paper.

For acoustic modes, *plane waves* of definite *wave vector* **q** should be excellent approximate solutions to the equations of motion as long as the glass behaves as a continuum at the scale of the acoustic wavelength. Experiments over a broad range of acoustic frequencies Ω, from ~ 1 MHz to ~ 400 GHz for silica, indicate the existence of such weakly damped waves (see, e.g., the discussion in Ref. 17). Propagation experiments directly demonstate that these waves transport energy.[18] However, upon further increase of Ω, the acoustic excitations must eventually lose this property as shown by the existence of a *plateau* in the temperature (T) dependence of the thermal conductivity.[1] This plateau cannot occur if *acoustic plane waves* would continue to propagate at high frequencies (see, e.g., Ref. 19). In our opinion, these waves must become strongly scattered from some crossover frequency Ω_{co} beyond which the excitations cease to *propagate* to become at best *diffusive*.[20,21] In this case, the corresponding crossover wave vector q_{co} should mark the ''end of the branch'' since there is no physical significance to wave vectors $q > q_{co}$. We have shown previously, using inelastic x-ray scattering (IXS) spectroscopy, that such a crossover can be observed on the longitudinal acoustic (LA) branch of d-SiO_2.[14] This material is more homogeneous than normal vitreous silica, v-SiO_2, and this pushes the crossover to higher values, $\Omega_{co} \simeq 9$ meV (or 2.2 THz) and $q_{co} \simeq 2.2$ nm^{-1}, as opposed to the estimates ~ 4 meV and ~ 1 nm^{-1} for v-SiO_2.[22] Hence, the experimental investigation of d-SiO_2 is possible below the crossover using the present capabilities of IXS, whereas for v-SiO_2 only the regions near and above the crossover are accessible. We believe that it is the latter fact which led the authors of Ref. 23 to conclude to the absence of an Ω_{co} in silica, a claim that seems incompatible with the T position of the plateau.[1] The same authors criticized the analysis in Ref. 14 on the basis of the poor S/N ratio of the spectra.

New IXS data on d-SiO_2, with much improved S/N ratio, are presented here. Not only do they confirm in many points our previous conclusions,[14] including the probable occurrence of an Ω_{co} at ~ 9 meV, but now the data quality allows us to obtain the linewidth Γ of some Brillouin peaks for $Q \leqslant q_{co}$, where our results are consistent with a $\Gamma \propto q^{\alpha}$ relation, with α up to at least 4. This seems at variance with a phenomenology predicting $\Gamma \propto Q^2$, as recently proposed.[24] We also examine the limit of the acoustic signal at high Q. We find a clear emergence of the BP signal at $Q \sim 4q_{co}$, and show its growth at much larger Q values.

II. DATA ACQUISITION AND HANDLING

The experiment was performed on the high-resolution IXS spectrometer at the European Synchrotron Radiation Facility (ESRF) in Grenoble, France. The energy of the monochromatic incident x rays is 21.7 keV. Scattering angles down to 1.5° are used, fixing the lower limit of Q at

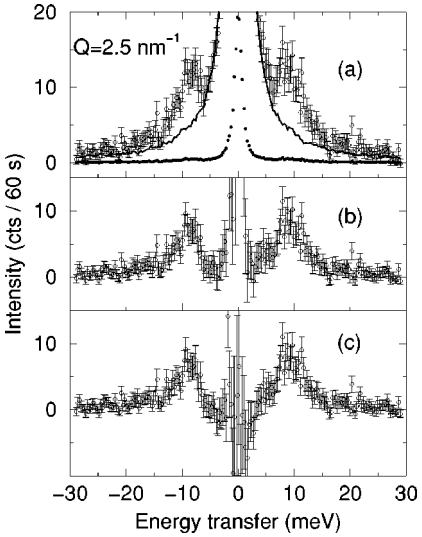

FIG. 1. X-ray Brillouin spectrum of d-SiO_2 at $Q=2.5$ nm^{-1}: (a) The full spectrum $\times 1/25$ (full dots), the wings (open dots with error bars), and the instrumental function matched to the peak at $\omega=0$ (continuous line); (b) the spectrum after subtraction of 90% of the central peak; (c) the same after full subtraction of the central peak.

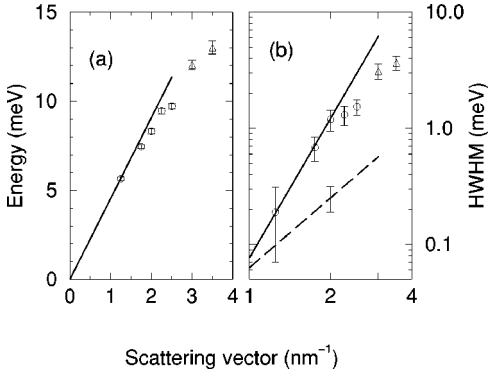

FIG. 2. The parameters of DHO fits to x-ray Brillouin spectra at small Q: (a) $\hbar\omega_0$, where the line extrapolates the Brillouin light scattering result; (b) $\hbar\Gamma$, the solid line is in Q^4, and the dashed line with error bar is $\hbar\Gamma_{hom}$ extrapolated from Brillouin light scattering results. The data points are shown with circles in the region of validity of this approximative line shape and with triangles above it. One should note that the error bars on the energy are smaller than the size of the circles.

1 nm^{-1}. The measured energy resolution is $\sim$1.5 meV. Five detectors are available allowing for simultaneous data collection at several Q values. The sample was the same as in Ref. 14, with a density $\rho=2.62$ g/cm^3 and a thickness of $\sim$2 mm. To optimize the inelastic signal, the data were taken at an elevated temperature, as in Ref. 14, namely, 565 K which is a value where the densified structure does not yet relax over the duration of the experiment. Improvements in the instrument luminosity and to some extent in its resolution result in an appreciable increase of the S/N ratio. The spectrum in Fig. 1 should be compared to that in Fig. 1 of Ref. 14. Clearly resolved Brillouin-like peaks are now well seen on the tails of the elastic line.

For reasons explained below, it is significant to be able to extract the pure inelastic contribution from the measured spectra without relying on models for the form of the inelastic structure factor $S(Q,\omega)$. The wings of the instrumental function do modify the spectral shape, as illustrated in Fig. 1.[25] Figure 1(a) shows a full spectrum and its enlarged inelastic region. The solid line shows the tails of an instrumental profile matched to the height of the strong central peak. With $I_{\rm tot}$ the integrated total intensity, and $I_{\rm ine}$ the integrated inelastic part, we find at all Q's up to 3.5 nm^{-1} that $I_{\rm ine}/I_{\rm tot}\simeq 0.1$. It is known that in glasses the static structure factor $S(Q)$ at small Q, and thus $I_{\rm tot}$, is much larger than would be predicted from the compressibility.[26] There is however no reliable theoretical prediction for $I_{\rm tot}$, and to remove the central peak one must adopt an empirical approach in such cases. It is safe to assume that this peak is essentially elastic as it should result from the frozen-in disorder. Subtracting the solid line (instrumental profile) from the spectrum in (a), one obtains the result (c). The latter indicates that the inelastic intensity tends to zero for ω tending to zero. This is an important observation, as it is a general property of line shapes resulting from the strong scattering of plane waves, whether the mechanism for it is Rayleigh scattering[27] or resonant scattering as in Refs. 19 and 28. Hence, one should check that this important result does not depend on the precise amount of elastic subtraction. To this effect, we also show curve (b) for which only 90% of the elastic peak has been subtracted. This reduced subtraction also indicates a strong decrease of the inelastic line towards $\omega=0$, similar to (c). Owing to the sufficiently narrow instrumental width, above a few meV the difference between (b) and (c) is much smaller than the error bars. As a consequence, the exact amount which is subtracted does not significantly affect the results of numerical adjustments when only data points with $|\omega|>2.7$ meV are considered, as done below. For this reason the ''full'' subtraction, just as in (c), was used to obtain the experimental inelastic spectra such as these shown in Fig. 3. It should be noted that the question of the central peak, and the issue of the evolution of the inelastic spectral shape for ω tending towards 0, is a recurring problem in inelastic x-ray scattering. The problem is obviously more severe for stronger central peaks and also the search for a possible crossover is enormously more difficult when $\Omega_{\rm co}$ is too low. On both counts, the situation is considerably more favorable in d-SiO_2 (Ref. 14) than in v-SiO_2.[7,22,23]

III. THE ACOUSTIC SIGNAL BELOW ITS CROSSOVER

Starting with spectra at the lowest Q, it does make sense *below* $q_{\rm co}$ to adjust them with a damped harmonic oscillator (DHO) response function of frequency ω_0 and half-width Γ. Indeed, below $q_{\rm co}$ the data can be interpreted in terms of phonons of wave vector $q=Q$, acoustic frequency $\Omega=\omega_0$, and damping Γ. In this way one can extract values for Ω and Γ, quite independently from specific models. As usual in doing this, the DHO spectral function is convoluted with the instrumental response and account is taken of the finite acceptance angle of the instrument which gives a spread in Q.

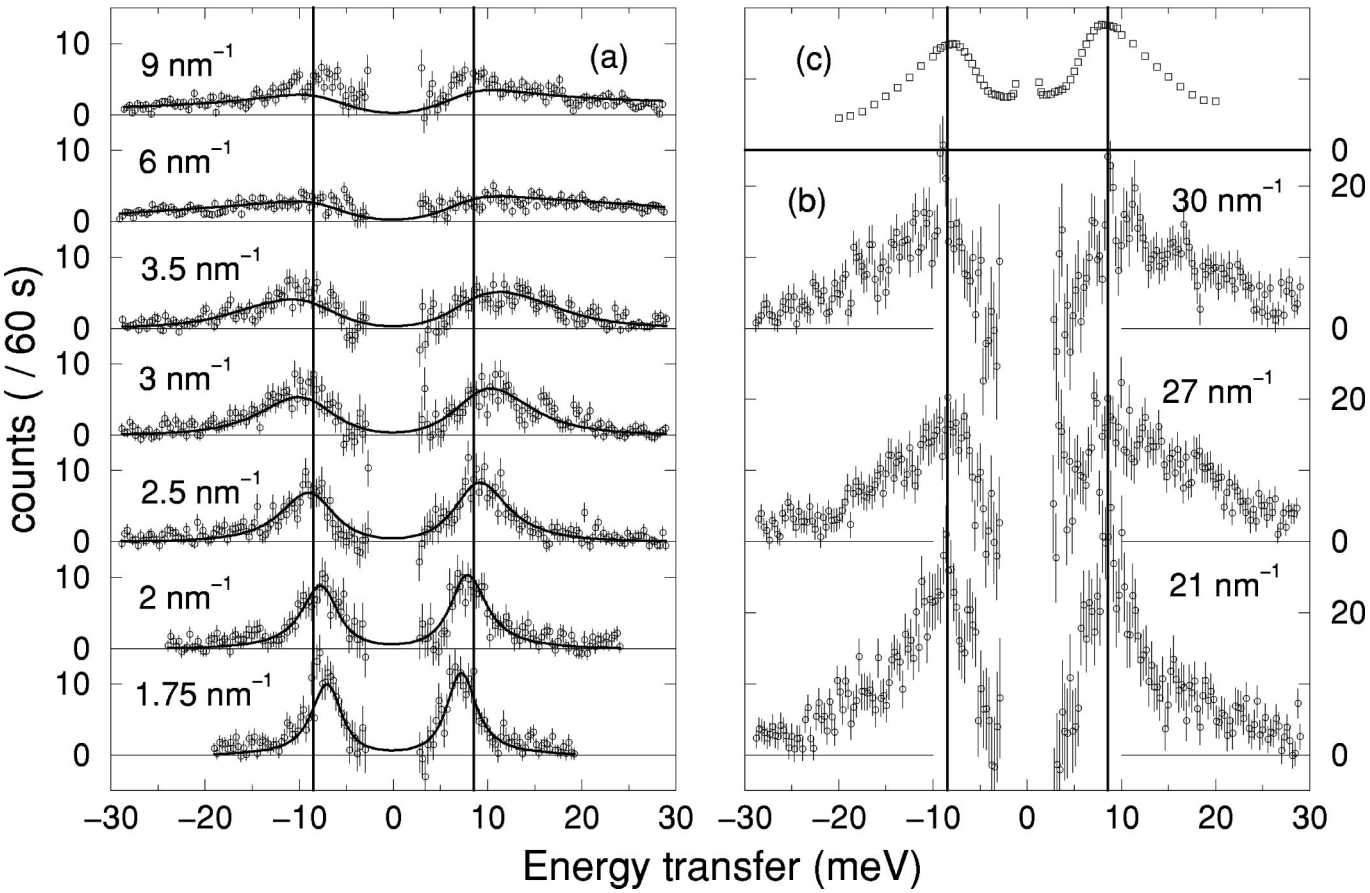

FIG. 3. Examples of inelastic spectra over a broad range of Q: (a) spectra in the Brillouin region. The solid lines represent an EMA prediction as explained in the text; (b) spectra in the boson peak region; (c) the boson peak from INS, averaged over a broad range of Q, on an arbitrary ordinate scale. Vertical lines are drawn at $\pm\Omega_{\mathrm{BP}}$.

The results are shown in Fig. 2. The fits to the DHO were performed up to $Q=3.5$ nm^{-1} which we know is above q_{co}.[14] As seen on Fig. 2(a), ω_0 merges into $c\,Q$ at the smallest Q, where c is the velocity found in Brillouin light scattering. There is a bend in $\omega_0(Q)$ near $Q\simeq 2.25$ nm^{-1}, and then a $\sim$20% jump in ω_0 near 2.5 nm^{-1}. The bend might evoke what is expected for acoustic branches, also in crystals, as one moves up in Q. The jump is very unusual. A remarkably similar jump was already found in Ref. 14. These being independent measurements, we believe that this apparent jump is reproducible. A similar anomaly (a bend followed by a jump) is in fact found in the value of Γ [Fig. 2(b)]. In that case, the jump near $Q\simeq 2.5$ nm^{-1} might be as much as a factor 2. These anomalies are presumably ''artifacts'' that result from using the DHO spectral function beyond q_{co}, i.e., beyond its domain of validity. An example for such effects was already demonstrated in Ref. 27. In that case, Brillouin light-scattering spectra of very high S/N ratio were obtained on silica aerogels at different Q values spanning a crossover to strong scattering. These spectra were adjusted both to a DHO and to a crossover spectral function. Above q_{co}, the peak of the best DHO moves to frequencies above the peak of the spectrum. Simultaneously, the linewidth of the DHO increases to better match the high-frequency wing of the spectrum. We feel that a similar effect, namely, an instability in the DHO fit parameters, might be at work here. If so, 2.5 nm^{-1} is beyond q_{co}, and the values of the DHO parameters for $Q\geqslant q_{\mathrm{co}}$ are not physically meaningful. For this reason they are shown with different symbols in Fig. 2. This artifact gives thus some sort of signature of the crossover on both ω_0 and Γ, and it exhibits the approximative end of the LA branch. Interestingly, the artifact occurs at the value of Q where the acoustic peak crosses the boson peak frequency, as will be seen in Fig. 3.

Another feature shown in Fig. 2(b) is that Γ rapidly grows with Q at small Q values. If the main contribution to the width in the region from $\sim$1 and $\sim$2 nm^{-1} came from Rayleigh scattering of plane waves, one would have $\Gamma\propto q^4$ in that region.[29] Such a dependence is illustrated by the solid line in Fig. 2(b). A similar Ω^4 dependence would result from the resonant scattering of plane waves in the low-frequency wing of a local mode.[19] It is also obtained in the soft-potential model.[15,28] In addition, there should be a homogeneous contribution to the width, $\Gamma_{\mathrm{hom}}\propto q^2$.[30] The latter has been measured on the same sample and at the same temperature using Brillouin light scattering with $Q=3.66\times10^{-2}$ nm^{-1}.[31] Its extrapolation in Q^2 is shown by the dashed line in Fig. 2(b). The error bar on this line represents the accuracy of the Brillouin measurement. We note that near q_{co} the observed Γ is significantly larger than the extrapolated Γ_{hom}.

Hence, there must be a region of q where Γ increases faster than in q^2, for example, with a power q^α and our observations are consistent with $\alpha=4$. It should be remarked that $\alpha=4$ was clearly observed below a crossover to strong scattering in a lithium borate glass.[10] To close the discussion of Fig. 2, we note that the precise value of Ω_{co} will depend on its specific definition. Thus, it can only be discussed on the basis of a model as done further below. On the other hand, the value of q_{co} seems to be quite close to 2.25 nm^{-1}. Finally, one might note that Ω_{co} is already reached when $\Gamma \simeq \Omega/6$ in the DHO fits.

IV. THE MERGING WITH THE BOSON PEAK

Figure 3 illustrates the evolution of the strength and shape of the inelastic contributions over a large range of Q, from 1.75 to 30 nm^{-1}. Care was taken to achieve a single relative scale for the intensities at all Q's. Indeed, the collected intensity could change with Q owing to a number of experimental artifacts, the main ones being either a vignetting of the scattered field by the oven window, or a slight change in the focusing of the analyzers onto the small collection holes placed 6 m away in front of the detectors. To correct for such artifacts, the measured scattering signal was scaled with a normalization factor obtained by comparing the integrated signal $I_{tot}(Q)$ to an independent measurement of the static structure factor of the same sample at the same T. This measurement was obtained under conditions where the above effects are inoperative. The correction is smaller than 10% of the measured signal for all spectra shown in Fig. 3.

Figure 3(a) shows at small Q some of the spectra that disperse like sound, as just discussed. Above $Q=2.25$ nm^{-1} the behavior changes. The position of the maximum in the spectra hardly moves for large increase in Q up to 6 nm^{-1}, while the spectra broaden. The signal becomes extremely broad at 6 nm^{-1} but it seems to narrow again at 9 nm^{-1}. In this region the integrated inelastic contribution increases with the increase of Q, but the S/N ratio degrades owing to the approach of the first sharp diffraction peak, located at 17 nm^{-1}. In Fig. 3(b), the stronger inelastic signal becomes obvious and persists up to our highest measured Q. This signal is, of course, associated with the excess modes. Roughly, its profile resembles the $S(Q,\omega)$ obtained on d-SiO_2 with INS and averaged from 10 to 70 nm^{-1}.[32] The latter, adjusted to the temperature of our measurements, is shown in Fig. 3(c). The maximum of the boson peak is at $\Omega_{BP}\simeq 8.5$ meV. As a guide to the eye, vertical lines at $\pm\Omega_{BP}$ are traced through all the spectra in Fig. 3. It is important to realize that the modes observed are of two kinds. There are the LA modes that scatter coherently, and the BP modes whose structure factor tends to zero at small Q since they are local excitations relating to rigid SiO_4 librations.[2,9] As discussed in Ref. 2, the main trend for the latter is to grow in Q^2, strongly modulated, however, by an inelastic structure factor $I^{(1)}(Q)$. In IXS, there is, in addition, the effect of the atomic form factors. For the LA modes, it is reasonable to use the average form factor of SiO_2, $\overline{f}(Q)$. For the BP modes, the issue of the appropriate form factor is more complicated. The rigid tetrahedra librations are accompanied with translations,[13] so that the correct average of $f_O(Q)$ and $f_{Si}(Q)$ that should be used might even depend on Q. Fortunately, this delicate point is of no consequence for what follows.

The curves in Fig. 3(a) correspond to the ''crossover to strong scattering'' (XSS) model described in Ref. 14. This is a model for the LA-modes that essentially results from an effective medium approximation (EMA).[29] All the curves are drawn with *the same* parameters,[33] in particular, with $\Omega_{co}=9$ meV, and with a *constant amplitude* coefficient A. Account has been taken of the form factor $\overline{f}(Q)$ in that the curves are drawn with an amplitude $A[\overline{f}(Q)/30]^2$. This leads to a 14% reduction at $Q=9$ nm^{-1}. Although the XSS model is to some extent phenomenological, it remarkably describes the six spectra from $Q=1.75$ to $Q=6$ nm^{-1}, just changing the value of Q.[34] One of the points of the model is that it predicts zero spectral weight at $\omega=0$. Owing to the convolution, a slight spectral weight at $\omega=0$ is seen in Fig. 3(a), but it is considerably smaller than the one that would result from DHO fits.[14] Looking now at the spectrum at 9 nm^{-1}, one sees a definite *excess* above the XSS curve. Its shape is quite similar to the spectrum observed at 21 nm^{-1}. We feel that the most probable origin for this excess is just the BP. This agrees with observations at small Q of a BP-like signal in INS from v-SiO_2.[2] There appears in Fig. 3 quite a strong Q dependence in the BP spectral intensity. It would be premature to venture into a more detailed analysis based on these limited data. We just point out that an extensive measurement on the BP spectrum of d-SiO_2 with IXS, and this over a broad range of Q, would be very worthwhile.

V. SUMMARY AND DISCUSSION

To summarize, our experimental results are consistent with the following: (i) the existence of a crossover to strong scattering of plane acoustic waves at $\Omega_{co}\simeq 9$ meV in d-SiO_2; (ii) the presence of a region below q_{co} where the linewidth of these waves increases rapidly with q, possibly with a law $\Gamma \propto q^4$; (iii) the remarkable success of the EMA model in describing the acoustic spectrum from below to well above q_{co}; (iv) the merging of the acoustic signal with the BP at the same frequency, the latter giving an additional signal for Q above a few times q_{co}; and (v) a nonuniform evolution of the BP intensity up to large values of Q.

The issue of the crossover is important as it immediately relates to the structure of glasses at extended length scales, which are the scales *relevant to the glass transition itself*. It is, in principle, difficult to picture how strong scattering of plane acoustic waves can occur in a medium as homogeneous as a dense glass. The value found here for q_{co} corresponds to a wavelength equal to about 20 Si-O distances, which seems quite large. Therefore some authors invoked fluctuations in internal pressure.[35] Large fluctuations in local elasticity were indeed found in simulations.[36] The most probable scenario for d-SiO_2 is that these fluctuations produce considerable local variations in the frequencies of the rigid SiO_4 librations associated with the boson peak. Resonant elastic waves are then likely to hybridize with these nonuni-

formly distributed oscillating entities, and it becomes easier to picture how this can lead to their strong scattering. This would directly explain why $\Omega_{co} \simeq \Omega_{BP}$. At intermediate Q values the waves observed in IXS are probably hybrids that scatter the x rays coherently, giving the acoustic signal, and quasi-incoherently, giving the BP signal.

Finally, one might ask whether the observation $\Omega_{co} \simeq \Omega_{BP}$ is just a fortuitous coincidence for d-SiO_2. There exist now sufficient evidence on other network glasses, which rather suggests that this could well be a universal property of these materials. In B_2O_3 there is a strong Boson peak at $\Omega_{BP} \simeq 3$ meV.[37] Very broad acousticlike spectra were observed with IXS and adjusted to the DHO model.[38] The authors find that $\Omega \simeq 2\Gamma$ at and beyond 5 meV. We use here Γ for the DHO *half-width* as in the discussion of Fig. 2 above. We found above that the Ω_{co} approximately equals the DHO Ω at the frequency where $\Gamma \simeq \Omega/6$. Using the same criterium for the data presented in Ref. 38, one indeed finds that $\Omega_{co} \simeq \Omega_{BP}$. However, B_2O_3 is not a favorable case for IXS measurements given the current resolution of the spectrometers. A much better case is that of the $B_2O_3(Li_2O)_x$ glasses for which Ω_{BP} moves to much higher values,[39] reaching 10 meV for $x=0.5$. Using the same criterium as above, namely, $\Omega_{co} \simeq \Omega \simeq 6\Gamma$, one finds from the published results of DHO fits[10] that Ω_{co} is indeed very close to Ω_{BP} both for $x=0.25$ and 0.5. Thus, there exist four cases where IXS has established that $\Omega_{co} \simeq \Omega_{BP}$.

Interestingly, the only case that remains in discussion is that of normal vitreous silica, v-SiO_2. The authors of Ref. 23 claim that there is no Ω_{co} in the region of the BP. Their evidence is based on constant energy scans obtained at energies near and above Ω_{BP}, which they compare to DHO fits on the one hand and to EMA curves on the other hand (see Fig. 3 of Ref. 23). Their EMA curves were calculated with fixed parameters taken from a 300 K determination that lacked much precision,[22] and, furthermore, the measurements in Ref. 23 are at 1200 K. One knows that the properties of v-SiO_2 change considerably with T (see, e.g., Ref. 40). Taking EMA parameters that are more consistent with the experimental conditions, one can find excellent agreement with the same data, as shown in Fig. 7 of Ref. 17. In that case, $\Omega_{co} \simeq \Omega_{BP}$. It seems that IXS with its current capabilities is still not able, in the special case of v-SiO_2, to establish the existence of Ω_{co}. Hence, the IXS data on v-SiO_2 are not in contradiction with $\Omega_{co} \simeq \Omega_{BP}$, while other measurements on v-SiO_2 agree with Ω_{co} around 4 meV.[30] In fact, we do not know of a single example of oxide glasses for which one would have solidly established that there is no crossover near the BP energy. For the above reasons, it seems, so far, that $\Omega_{co} \simeq \Omega_{BP}$ might be a common property of these materials.

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