# Sound attenuation in a unexplored frequency region: Brillouin ultraviolet light scattering measurements in $v$-$SiO_2$

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(Received 3 February 2005; published 6 May 2005)

We report ultraviolet Brillouin light-scattering experimental data on $v$-$SiO_2$ in an unexplored frequency region, performed with a newly available spectrometer, up to exchanged wave vector $q$ values of 0.075 $nm^{-1}$, at different temperatures. The measured attenuation follows a $q^2$ law and is temperature dependent. Such temperature dependence is found to be in good agreement with that measured at lower $q$, suggesting that the broadening of the Brillouin peak is mainly due to a dynamic attenuation mechanism. The comparison of the present data with those obtained by inelastic x-ray and visible scattering indicates the existence of a crossover among different attenuation mechanisms, whose nature is briefly discussed.

DOI: 10.1103/PhysRevB.71.172201 PACS number(s): 78.35.+c, 62.65.+k, 63.50.+x

Our present understanding of the sound attenuation mechanisms in vitreous systems is poor as compared to that of crystalline materials, although this topic has attracted the interest of several researchers from both the experimental and the theoretical point of view.[1] In particular, the nature of vibrational dynamics of disordered systems, as derived from the study of the low-frequency excitations (in the hypersonic range), has been a highly debated subject in recent years.[2–6] Vitreous silica ($v$-$SiO_2$, or amorphous quartz), considered as the prototype strong glass,[7] has been extensively studied, but the experimental results reported in the literature have had controversial interpretations.[8–12] It is known that a plane-wave excitation can propagate in a disordered structure only when the wavelength is much longer than the scale spanned by microscopic inhomogeneities; as the wavelength shortens, the wave is increasingly distorted and scattered. The question as to the causes of attenuation far from the long wavelength limit, is unlikely to have a single answer,[13–15] and different mechanisms have been suggested: attenuation induced by topological disorder, thermally activated processes, anharmonic effects, two-level systems.[16] In general, anharmonic effects are particularly relevant at relatively high temperatures, while two-level systems are expected to be effective at very low temperatures ($T < 1-10$ K). In any case, plane waves are believed to be a reasonable description of the vibrational excitations up to relatively large wave vectors ($q_0 \approx 5$ $nm^{-1}$), i.e., in the range where one observes a linear relationship between frequency ($\omega$) and exchanged wave vector ($q$).[11,17,18] For higher $q$ values, topological disorder causes (i) a mixing of the polarization of the acoustic modes, which is observed for different glasses both in experiments[17,19,20] and in molecular dynamics (MD) simulations[18,21] and (ii) the possible presence of positive dispersion in the longitudinal acoustic branch,[17] as theoretically predicted[22] and found in MD.[18] Below $q_0$, $v$-$SiO_2$ exhibits a substantially constant sound velocity.

The acoustic attenuation can be measured by the energy width $\Gamma$, at fixed $q$, of the Brillouin doublet characterizing the dynamical structure factor $S(q,\omega)$. In the hypersonic region [i.e., in the GHz region investigated by Brillouin light scattering (BLS)], $\Gamma$ exhibits a strong temperature dependence, and this indicates that the attenuation at these frequencies is to be ascribed to dynamical processes.[23,24] On the contrary, in the mesoscopic range [i.e., in the THz region investigated by the inelastic x-ray scattering technique (IXS)] $\Gamma$ has a negligible temperature dependence, supporting a nondynamical origin of the acoustic attenuation.[3] In vitreous silica two main attenuation mechanisms have been hypothesized in the GHz and THz regions. In the first scenario[23,25,26] the attenuation is characterized by a crossover from a mechanism dominated by dynamical relaxation processes with a frequency dependence of $\Gamma \propto q^2$ in the GHz range, to a mechanism dominated by strong phonon scattering, due to presence of topological disorder in the THz region. It is assumed that the latter process should exhibit a $\Gamma \propto q^4$ dependence (very similar to the Rayleigh light-scattering regime by independent particles). The second scenario[3] also suggests the existence of a crossover between a dynamical, low frequency, attenuation mechanism, and a high frequency, nearly temperature-independent one, due to topological disorder. However, a $\Gamma \propto q^2$ dependence is assigned to both mechanisms.[3,8,18,19,27] Such dependence, valid up to high $q$, has also been observed in MD simulations performed on realistic $v$-$SiO_2$ models,[28] in Lennard-Jones glasses in the harmonic approximation,[29] as well as in hard-sphere systems[22] and disordered linear chains;[30] it is also supported by recent theoretical free-energy landscape studies.[31,32]

BLS and IXS experiments do not cover the whole frequency (and $q$) range from GHz to THz, and investigations within the frequency gap which separates these techniques could be useful to discriminate between the different hypotheses. In this range, the sound attenuation has already been measured by the picosecond optical technique (POT),[33] and

 

no dependence of $\Gamma$ on temperature has been found in the range 80–300 K. However, in the region where there is overlap with the BLS attenuation data, the POT values of $\Gamma$ are always more than a factor 2 larger.

In order to shed light on the underlying attenuation mechanisms, an accurate investigation of the attenuation in the intermediate $q$ region is required. This is now possible thanks to the development of a new spectroscopic apparatus operating in the ultraviolet region.[34] Here we report the results of Brillouin ultraviolet light scattering (BUVS) on $v$ -$SiO_2$ (spectrosil), performed at different temperatures. We find that, in the BUVS regime, the measured linewidth $\Gamma$ is temperature dependent and, at fixed $T$, it varies as $q^2$.

The experimental apparatus used for the measurements with ultraviolet excitation, consists of a newly built spectrometer with high resolution, contrast and luminosity, called HIRESUV.[34] The instrument is based on a double-grating monochromator with a focal length of 4 m, specifically designed for Brillouin spectroscopy under visible (532 nm) and ultraviolet (266 nm) excitation. High luminosity is provided by two large echelle gratings ($400\times208$ mm with 31.6 grooves/mm); the sizes of the gratings and mirrors yield an instrumental $F$-number of 1:40 in the vertical plane (where dispersion occurs), and of 1:20 in the horizontal plane. In the UV range, HIRESUV works at the 230th order and reaches a resolution of about 0.6 GHz. To overcome air turbulence effects and small changes of refractive index, and in order to ensure thermal homogeneity, the whole apparatus, except the sample compartment, is located inside a chamber filled with helium at atmospheric pressure. All optical components (mirror, slits, and light collecting lenses) are positioned and aligned by means of computer-controlled microstep translation stages. The UV radiation is generated by a commercial system based on the second-harmonic generation of a visible laser source. Other details of HIRESUV will be published elsewhere.[34]

A typical spectrum at room temperature is reported in Fig. 1 (the measured resolution in this case was about 0.8 GHz), where the high contrast and resolution reached by the experimental apparatus can be appreciated. The BUVS signal (open circles), which is proportional to the dynamic structure factor $S(q,\omega)$ convoluted with the instrumental resolution function $R(\omega)$, is reported in a log scale together with a fit function (continuous line). To get quantitative information on the peak frequency and on the width $\Gamma$ of the excitations, the data have been fitted by the convolution of the experimentally determined $R(\omega)$, with the sum of an elastic and an inelastic contribution; the former is represented by a $\delta$ function, while the latter has been described by a damped harmonic oscillator model. As an example of the temperature behavior of the Brillouin peak, the anti-Stokes parts of the spectrum at two selected temperatures (open circles), together with their resolution curves (full circles) and the best fit (full lines), are reported in Figs. 2(a) ($T=270$ K) and (b) ($T=150$ K). Directly from the spectra, one notices that the width increases with temperature, indicating that, in this frequency range, at least part of the sound attenuation has a dynamical origin. In the inset of Fig. 2(b), we report the FWHM of the Brillouin peak, divided by $q^2$, as a function of

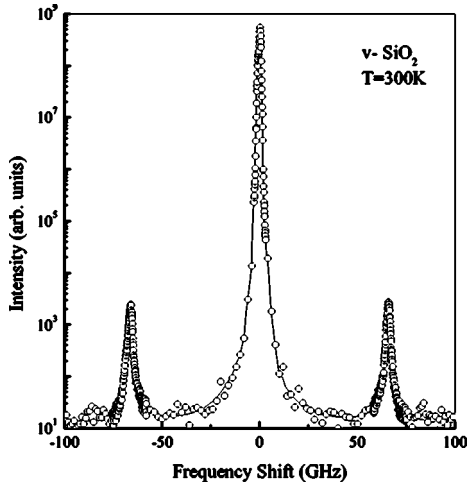


FIG. 1. Typical Brillouin ultraviolet light scattering spectrum on $v$-$SiO_2$ at $T=300$ K (180° scattering geometry, excitation wavelength 266 nm, power $\approx$100 mW). The experimental data (open circles) are reported together with the best fit (continuous line), see text.

temperature; the scaled data are practically coincident with those obtained by BLS.[23] The values of $\Gamma(q)$ measured at room temperature, resulting from the fits of BUVS (full circles) and BLS (full diamond and open circle,[23] open triangle,[35] bold star[36]) data, are reported in Fig. 3. The spectra, collected at 90°-scattering angle, were measured with a horizontal collection angle $\delta\phi_H=1.7$ mrad. In this configura-

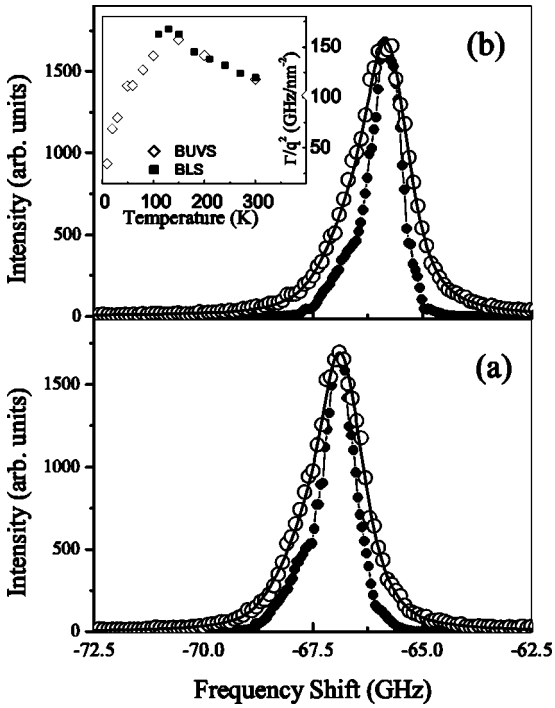


FIG. 2. Anti-Stokes part of BUVS spectra (open circles) at $T=270$ K (a) and $T=150$ K (b). The resolution (full circles) and the best fit (continuous line) are also reported. In the inset is shown the temperature behavior of $\Gamma/q^2$ for BLS (open diamonds) and BUVS (full squares).

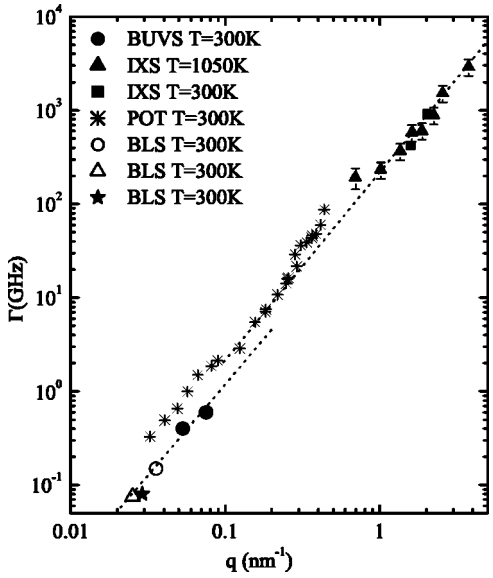


FIG. 3. Log-log plot of Brillouin widths as a function of the exchanged wave vector $q$, obtained at room temperature by BLS [open triangle (Ref. 35), open circle (Ref. 23), bold star (Ref. 36)], BUVS (full circles) in 90° and 180° scattering configurations, and IXS (triangles $T=1050$ K (Ref. 8), squares $T=300$ K, (Ref. [37])). Asterisks represent the results obtained by the POT technique (Ref. 33). The dashed lines, indicating the $q^2$ law, are guides for the eye.

tion the exchanged-wave-vector dispersion arising from the finite acceptance angle $\delta q/q=\delta\phi_H/2$ gives a negligible contribution to the the mode linewidth. The dashed lines represent the $q^2$ behavior, and are guides for the eye.

In the same figure we have reported the POT (asterisks[33]) and IXS data at $T=1050$ K (full triangles[8]) and at $T=300$ K (full squares[37]). As mentioned, the POT data are always about a factor 2 higher than the BLS and BUVS ones at similar $q$ values, as obtained by different groups, which hints at the existence of a systematic overestimate of the width. At high $q$, within the experimental error, $\Gamma(q)$ as obtained by IXS does not show any noticeable temperature dependence in the 300 to 1500 K range,[3] and the measured values are in good agreement with the ones calculated by MD at $T=0$.[28] $\Gamma(q)$ from IXS and MD is well fitted to a $q^2$ law in the 1 to 5 $nm^{-1}$ range. As mentioned, the temperature independence of $\Gamma$ in this range indicates that most of the attenuation has a nondynamical origin and is ascribable to topological disorder.[3] As for the BUVS and BLS data, between $\approx 0.01$ and 0.1 $nm^{-1}$ at 300 K, both consistently follow, again, a $q^2$ law: however, from Fig. 3 it is evident that it is *not* the same straight line as for IXS and MD. As shown in the inset of Fig. 2(b), this $\Gamma(q)=A(T)q^2$ dependence is obeyed at all temperatures in the range 100–300 K, though with $T$-dependent values of the constant $A$. Therefore, the picture that emerges from the present data is the following: for $q>1$ $nm^{-1}$, configurational disorder yields $\Gamma(q)\propto q^2$, independent of $T$; for $q<0.1$ $nm^{-1}$, dynamical disorder yields $\Gamma(q)=A(T)q^2$; the value of $A(T=300)$ is such that the extrapolation of the low-$q$ straight line (in log-log coordinates) lies lower than the high-$q$ one. This necessarily requires that somewhere in the intermediate range (not accessible to the present experimental apparatus), $\Gamma(q)$ grows faster than quadratically with $q$.[38] The precise mechanism for such regime is presently unknown, though some indication is provided by existing MD simulations at $T=0$, concerning the role played by internal stress and frustration.[39] In fact, it was observed that in topologically or elastically disordered systems not affected by internal stress, the nondynamical variation of the width is of the type

$$\Gamma(q)\propto q^{d+1}, \tag{1}$$

where $d$ is the appropriate *spectral* dimension of the system;[40,41] in particular, $d=3$ for homogeneous materials, $d=4/3$ for percolators at threshold. On the contrary, in the presence of internal stress Eq. (1) is not followed by any of the systems examined in Ref. 39, for which it is found that the exponent is in any case smaller than $d+1$, and close to 2 for Lennard-Jones systems and realistic models of $SiO_2$.[28] It might therefore be argued that, for reasons not clear at present, in real $SiO_2$ internal stress becomes ineffective for some value of $q<\approx 1$ $nm^{-1}$, so that Eq. (1) holds again yielding a slope of 4 in this region. This regime holds until the temperature-dependent, dynamical mechanism ($\propto q^2$) takes over at lower $q$. In conclusion in this work, we have reported measurements of the Brillouin spectra performed employing UV excitation on $v$-$SiO_2$, using the new HIRE-SUV spectrometer. The experimental data obtained in the up-to-now unexplored frequency region, hint at the existence, in vitreous silica, of different sound-scattering processes in different regions of exchanged wave vector, in addition to the dynamical one that prevails in the range covered by BLS and BUVS. The mechanism that produces the crossover between the two distinct $q^2$ ranges is not known at present, in particular it is not clear whether it is a matter of a structural or a dynamical one; existing simulations at $T=0$ (Ref. 39) might hint at an effect of structural disorder, but further experimental study is needed in this rather out-of-reach range of exchanged wave vectors.

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