Finite size effects in Brillouin scattering from silica glass

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Finite sample sizes increase the measured Brillouin scattering linewidths. This should not be mistaken for a genuine damping of sound. Using high-resolution interferometry, we show that in silica glass under ambient conditions and with visible light excitation, the additional width becomes observable for sample thicknesses below 0.2 mm. Similarly, light absorption reduces the scattering volume, producing a strong contribution to the width for excitation of bulk samples above their absorption edge. The resulting limitations are illustrated for UV Brillouin scattering from silica.

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In Brillouin light scattering (BLS), one usually observes sound waves at frequency shifts $\Omega = vQ$. Here, v is the sound velocity and Q is the scattering vector fixed by the experimental geometry. Q is also the wave vector of the observed sound wave. Essential information often derived from these experiments is contained in the broadening of the spectrum beyond the instrumental resolution. For sufficiently large scattering volumes and homogeneous materials, this broadening is generally produced by sound attenuation. However, it has been known for a long time that in restricted scattering volumes there appears an additional component to the broadening. It results from the spread in Q which, by virtue of the Fourier transformation, directly relates to the restricted range of overlap of the incident and scattered waves along the corresponding spatial direction, say x. One expects that $\Delta Q \Delta x \approx 1$. In quantum terms, this is the uncertainty principle $\Delta p \Delta x \geqslant \hbar/2$, since the observed phonons have a momentum $p = \hbar Q$. In practice, a small Δx can occur in two ways. First, in thin samples, the extent of waves propagating perpendicularly to the sample boundaries are, independently from the measurement technique, restricted in that spatial coordinate. Second, if the medium is strongly absorbing for the exciting radiation, the investigated volume is thereby reduced, also leading to uncertainty broadening. Both cases were first observed long ago,[1,2] and they were subject to extensive analyses, e.g., in Refs. 3–5 and references therein.

There are several reasons to investigate the effect of restricted scattering volumes in glasses. First, BLS experiments are nowadays performed under high pressure. Typically, the exciting beam and the scattered photons traverse then the anvils of a diamond-anvil cell, and the sample might be quite thin. Linewidths must then be corrected for uncertainty broadening. Second, there is considerable interest in amorphous materials that only exist as thin films. The measurements of their acoustic properties by BLS cannot ignore their small thicknesses. Third, many glasses or amorphous materials have a limited transparency range which can restrict appreciably the scattering volume. This is particularly true for amorphous semiconductors or metals observed with visible excitation. In this Brief Report we discuss the simple example of vitreous silica. This is a glass, which under ambient conditions has a relatively narrow sound-damping linewidth.[6] Using high performance interferometric spectroscopy,[7] it is possible to observe uncertainty broadening with visible light excitation in plates that may be as thick as ~0.2 mm. On the other hand, there is also a fundamental interest in investigating sound up to very high frequencies.[8,9] To this effect, one might attempt achieving higher Q values using the excitation of shorter vacuum wavelength λ_0. Indeed, Q is given by

$$Q = 4\pi n_1 \sin(\theta/2)/\lambda_0, \tag{1}$$

where n_1 is the real part of the complex refractive index, $n = n_1 + in_2$, and θ is the internal scattering angle. Hence, Q could in principle be increased using UV rather than visible light excitation.[10,11] However, n varies very sharply in the region of the UV edge. This can produce uncertainty broadening[1] that should not be mistaken for intrinsic sound damping.

Results obtained with plates of finite thickness are described first. Several optically polished plates of decreasing thickness were prepared out of a single large piece of Tetrasil SE silica.[12] A single mode argon-ion laser operating at 514.5 nm is used for excitation, and the scattering is observed in the backscattering geometry ($\theta = 180°$). The samples are at room temperature, and their normal is tilted by about 15° from the incident beam to eliminate back reflections. Thus, the wave vector Q of the investigated phonon is not perpendicular to the faces of the plate. In this condition, there is no standing-wave interference effect on these phonons. As shown in Ref. 2 the uncertainty broadening is then characterized by a $(\sin y/y)^2$ line shape, specifically

$$f(\omega) \propto \frac{\sin^2[(\omega - \Omega)d/2v]}{[(\omega - \Omega)d/2v]^2}. \tag{2}$$

In this expression, ω is the spectral frequency, $\Omega = vQ$ is the mean sound frequency, and d is the geometric length of the light path in the sample.

The spectra are analyzed with a modern version of the instrument described in Ref. 7. It contains two Fabry-Perot (FP) interferometers in series. The first is a planar FP of 1.5 mm spacing, utilized in four passes. It is maintained at a *fixed* spacing which is dynamically adjusted to one of the Brillouin lines with the help of an electro-optically modulated signal at the Brillouin frequency. This FP acts as a prefilter effectively eliminating other spectral components

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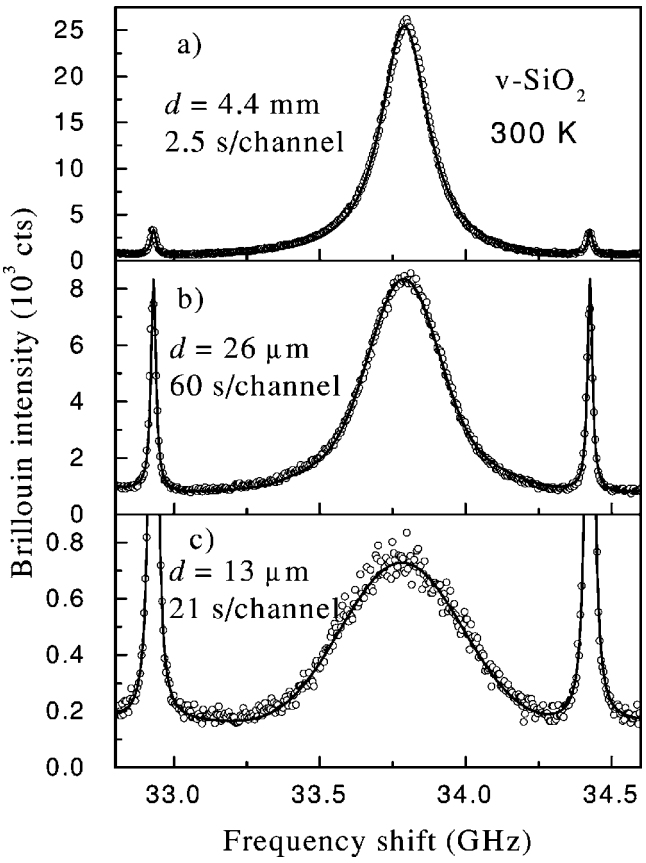

FIG. 1. BLS Stokes spectra observed in backscattering at three thicknesses d. The narrow peaks, spaced by the free spectral range, are the remaining elastic signals. The continuous lines show the adjustment explained in the text. One remarks on the considerable decrease in intensity and the increase in apparent width for decreasing thickness. Spectrum (a) is just broadened by sound absorption. The increase in width for the thinner samples is produced by the uncertainty in Q.

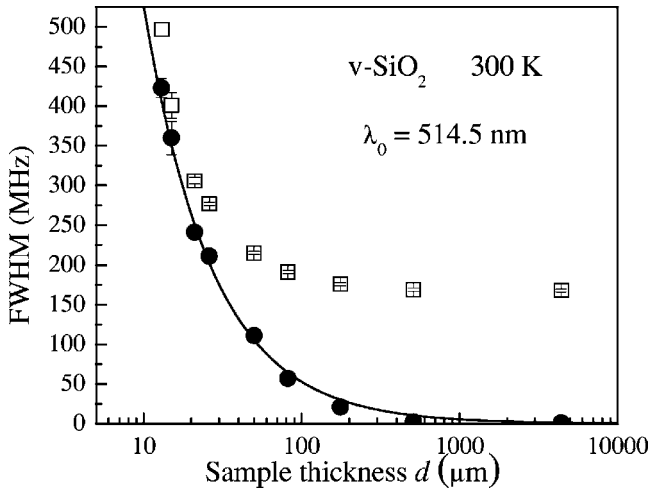

FIG. 2. Linewidths extracted from fits of Brillouin spectra. The open squares are obtained adjusting simply with a DHO convoluted with the instrumental profile. These are "effective" widths. For the two *thickest* samples, this width is due to sound absorption only. The solid dots show the width of the function $f(\omega)$ obtained from the fits using a double convolution. The solid line is the width of $f(\omega)$ calculated with Eq. (2). For the two *thinnest* samples, this uncertainty broadening is the main contribution to the effective width.

and reducing considerably the elastic signal strength. The second FP is a confocal one of 50 mm spacing, which is scanned for the analysis. Its instrumental full width is below 30 MHz so that broadenings down to $\approx$5 MHz can be extracted by numerical treatment of the data. The scan is also calibrated with the modulated signal so that spectra are presented on an absolute frequency scale.

Typical spectra are illustrated in Fig. 1. The free spectral range of 1.5 GHz is much smaller than the Brillouin shift, so that the Stokes line is observed at an interference order very different from that of the elastic lines. The measured spectra are the *double* convolution of $f(\omega)$ with the instrumental response and with a damped harmonic oscillator (DHO) line shape of frequency Ω and width Γ representing the acoustic damping. In addition, for broad spectra, the transmission curve of the prefilter must be taken into account. Finally, in these experiments, since the scattering volumes are rather small, large collection angles of external half-aperture $\Delta\theta = 10°$ have been used. The effect of this was checked for a bulk sample and this additional broadening was taken into account in the overall adjustement of the spectra. In Fig. 1(a), the uncertainty broadening is negligible, in which case the profile is simply the DHO convoluted with the instrumental response. The full width of the DHO is found to be $\Gamma/2\pi = 166$ MHz. The same value was then kept for all spectra. Figure 1(b) shows an intermediate case where the effect of $f(\omega)$ can already be recognized in the profile shape. The full line shows the adjustment with the double convolution explained above. Figure 1(c) corresponds to the smallest d that has been used, in which case a considerable broadening is observed.

Figure 2 shows the thickness dependence of the full width at half maximun of the function $f(\omega)$ derived from fits of the experimental spectra, as explained above. It should be compared to the solid line which shows the theoretical width of $f(\omega)$ calculated from Eq. (2) using the known values of d and v. One notices the excellent agreement. This also suggests that the intrinsic sound damping, characterized by $\Gamma/2\pi = 166$ MHz, is not appreciably modified by the cutting and polishing. The same is true for the frequency Ω which is remarkably constant over the series of samples. Also shown in Fig. 2 are widths obtained by fitting the observed spectra to a single DHO convoluted with the instrumental response. This gives an estimate for the total width which starts increasing measurably when d falls below 0.2 mm. This clearly illustrates that uncertainty broadening is an important contribution for samples thinner than $\sim$30 μm. Such small thicknesses are often employed in BLS experiments under pressure. It must be remarked that the effective width in Fig. 2 is not the sum of Γ plus the width of $f(\omega)$. It is smaller, resulting from the convolution with the non-Lorentzian profile of Eq. (2). This emphasizes that the only proper way to extract Γ from Brillouin spectra on thin samples is to proceed with the double convolution.

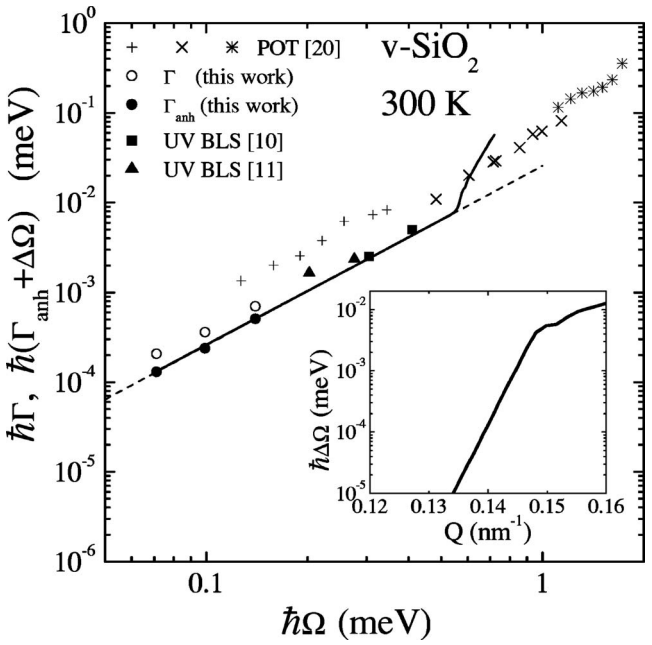

FIG. 3. Full widths $\Gamma(\Omega)$ measured in different spectroscopies, as indicated. Electron volts are used as usual at these energies. The dashed line represents Γ_{anh}. It terminates near 1 meV beyond which Γ_{anh} saturates. The full line shows $(\Gamma_{anh}+\Delta\Omega)$ vs Ω. Inset: the value $\Delta\Omega(Q)$ calculated for silica based on α_0 from Refs. 13 and 14.

We now turn our attention to finite-size effects produced by the absorption of the incident radiation. As shown in, Ref. 1 and as confirmed by more detailed calculations,[3,4] the uncertainty broadening produced by light absorption is given by

$$\frac{\Delta\Omega}{\Omega} = \frac{2n_2}{n_1} = \frac{\alpha_0\lambda_0}{2\pi n_1}. \tag{3}$$

Here $\Delta\Omega$ is the additional contribution to the observed full width, and α_0 is the power absorption coefficient of the *light*. The exponential light attenuation in the sample is expected to lead to an additional Lorentzian-like broadening in backscattering. Approximating the two broadenings as Lorentzians, the observed linewidth will then be $\Delta\Omega+\Gamma$, where Γ is produced by sound attenuation in the material $\Gamma=\alpha v$ where α is the power-absorption coefficient for *sound* at the frequency Ω. To calculate $\Delta\Omega$ for silica glass, we use a standard curve for $\alpha_0(E)$,[13] or equivalently another recent determination.[14] The absorption edge of silica is located at a photon energy $E\approx 8$ eV, corresponding to $\lambda_0\approx 155$ nm.[13] In relating Q to E with Eq. (1), the rapid dispersion of $n_1(E)$ must be taken into account at these energies.[15] The resulting $\Delta\Omega(Q)$ is shown in the inset of Fig. 3. It should be noted that this result is sample dependent. Indeed, the Urbach tail and the optical band gap of silica do depend on the sample quality and conditions. Thermal disorder lowers both the edge and the gap,[14] while lower glass-stabilization temperatures improve the sample transparency.[16] A significantly higher edge is achieved with slight fluorine doping,[16] while a very low edge at $Q\approx 0.11$ nm^{-1} ($E\approx 6.9$ eV) is derived from Fig. 6 of Ref. 17 In the latter case, it is likely that it is irradiation by the strong laser-plasma source[18] that produces this sizable downward shift, as suggested in Ref. 19.

In Fig. 3, $\Delta\Omega$ is compared to Γ. At low frequencies and sufficiently low temperatures, the sound absorption α is controlled by thermally activated relaxation (TAR), as reviewed in Ref. 6. However, as $\hbar\Omega$ increases beyond $\sim$0.1 meV, α at room temperature becomes dominated by anharmonic sound damping[6] which gives $\Gamma_{anh}\propto\Omega^2$. The Ω^2 dependence of Γ in this region was first observed using a picosecond optical technique (POT).[20] The data obtained with POT on three samples[20] are shown with different symbols in Fig. 3. The lowest frequency set used a rather thick silica film, in which case irregularities in the film thickness contributed to the observed width.[20] That there is such an extra width is now obvious from comparing with the other data shown in Fig. 3. At the lowest Ω values, we show three points that are new determinations obtained with the BLS instrument described above. These were obtained at scattering angles θ of 61°, 90°, and 180°, and at a laser wavelength $\lambda_0=514.5$ nm. The experimental widths Γ are shown with open dots in Fig. 3. After subtraction of the TAR contribution following Ref. 6, the full dots are obtained. A line $\hbar\Gamma_{anh}=A\hbar^2\Omega^2$ is traced through the 180° scattering point, with $A\simeq 0.025$ meV^{-1}. For the points obtained with UV Brillouin scattering,[10,11] TAR becomes negligible so that the highest frequency points align very well with the $A\hbar^2\Omega^2$ law passing through the lower BLS points. Thus, this region of the $\Gamma(\Omega)$ curve is rather well understood now.

Oppositely, for sound quanta above 0.5 meV, there are several questions that remain unsettled. Anharmonic damping is expected to saturate at higher energies above $\sim$1 meV.[21] On the other hand, we know from other oxide glasses that a rapid increase of damping, possibly in Ω^4, takes place on the approach from below the boson peak maximum.[9,22] In silica the boson peak is located at $\approx$4 meV, and indeed the POT data suggest a possibly faster increase in Γ. It is in this region, and up to $Q\approx 1$ nm^{-1} (or $\hbar\Omega\approx 4$ meV), that accurate sound absorption data are dearly missing in silica glass. Indeed, this region is not accessible to current inelastic x-ray scattering. Unfortunately, it is also not reachable with deep UV BLS. To see this, one should consider the line $\Delta\Omega+\Gamma_{anh}$ in Fig. 3. We observe a rapid increase beyond 0.5 meV, corresponding to $Q>0.14$ nm^{-1}. This rapid increase of $\Delta\Omega$ marks the end of the useful range of UV Brillouin scattering. In addition, one calculates that for $\Delta\Omega=\Gamma_{anh}$ one has $n_2\approx 0.01$. The corresponding α_0 is then $\approx$8000 cm^{-1}, which attenuates the signal considerably, making the spectroscopy unpractical. The readers should note that reports of deep UV scattering in silica have recently been presented for values of Q below 0.14 nm^{-1}.[23]

In conclusion, we have shown that it is really crucial not to forget the possible influence of uncertainty broadening when performing quantitative Brillouin scattering measurements. Although this was known for a long time, it seemed appropriate to remind the glass community (using as an example the case of silica) of these well-established limitations. These can be important whether one works with thin samples or close to an absorption edge.

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