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# Dynamic sound attenuation at hypersonic frequencies in silica glass

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In order to clarify the origin of the dominant processes responsible for the acoustic attenuation of phonons, which is a much debated topic, we present Brillouin scattering experiments in various silica glasses of different OH impurities content. A large temperature range, from 5 to 1500 K is investigated, up to the glass transition temperature. Comparison of the hypersonic wave attenuation in various samples allows to identify two different processes. The first one induces a low temperature peak related to relaxational processes; it is strongly sensitive to the extrinsic defects. The second, dominant in the high temperature range, is weakly dependent on the impurities and can be ascribed to anharmonic interactions.

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The origin of sound attenuation in glasses has been a highly debated topic in recent years. Several models have been proposed but no consensus about the interpretation of sound attenuation has been reached.<sup>1,2,3</sup>

Several mechanisms responsible for sound attenuation have been identified for over the year:

1. interactions of acoustic waves with defects characteristic of the disorder. Those defects are often described as tunneling systems responsible for many anomalous properties of glasses in the low temperature regime.<sup>4</sup>
2. thermally activated relaxational processes, as clearly demonstrated by ultrasonic measurements.<sup>5</sup> The microscopic structural origin of the thermally activated processes is often related to tunneling defects or soft modes responsible for the interaction with acoustic waves.<sup>6</sup>
3. Rayleigh-like scattering by static inhomogeneities, independent of temperature. This has been observed for example in porous glasses (xerogels, aerogels).<sup>7,8</sup> This mechanism yield an attenuation varying like  $q^4$ , where  $q$  is the momentum transfer of the interaction in scattering experiment. Rayleigh-like scattering has been also put forward to explain the change of regime for the attenuation in the THz range.<sup>3,9</sup> An alternative explanation of the frequency power dependence in  $\omega^4$  (where  $\omega = 2\pi/\nu$  is the pulsation of the excitation) in terms of fractons<sup>10</sup> has been also proposed.
4. anharmonicity responsible for the interactions of acoustic waves with thermal phonons, as in crystals.<sup>11,12</sup>

Each mechanism dominates in different temperature and frequency ranges. It is expected that the mechanisms 1 and 2 are dominant at low temperatures, 3 in very inhomogeneous media or at wavelengths comparable to a few atomic length and the mechanism 4 at high temperature and high frequencies. Moreover, the influence of several parameters have been studied in order to distinguish between universal behavior of the sound attenuation in glasses and more specific properties, related for example to composition effects, or to the method of preparation (impurity concentration or thermal history).<sup>13</sup> The influence of pressure,<sup>14</sup> irradiation by neutrons<sup>15,16</sup> or permanent densification<sup>9</sup> has also been investigated.

In this paper we present results about Brillouin scattering measurements of sound attenuation at hypersonic frequencies in silica. This work provide new data useful for shedding new light on the origin of processes responsible for the sound wave attenuation. By comparing results for silica with different impurity content, we will demonstrate that in the hypersonic regime:

- the thermally activated relaxational processes which dominate at low temperature are clearly related to specific, clearly identified extrinsic structural defects;
- at high temperature, anharmonic processes are less dependent on these local changes of composition.

Two samples of vitreous silica of different origins were investigated. One of them is a commercial fused quartz “puropsil”) provided by Quartz et Silice, Nemours, France. This sample contains a few ppm (less than 20) of OH impurities. Another sample was prepared by densification of a silica aerogel. The sample of initial density  $0.3 \text{ g cm}^{-3}$ , was heat-treated at about  $1100 \text{ }^\circ\text{C}$  for several hours till its density reaches the same value,  $2.20 \text{ g cm}^{-3}$ , as amorphous silica. This sample has both a different “fictive temperature” (lower than  $1100 \text{ }^\circ\text{C}$  compared to the commercial samples (between  $1200$  and  $1300 \text{ }^\circ\text{C}$ ) and a different OH content (around 3000 ppm, determined with

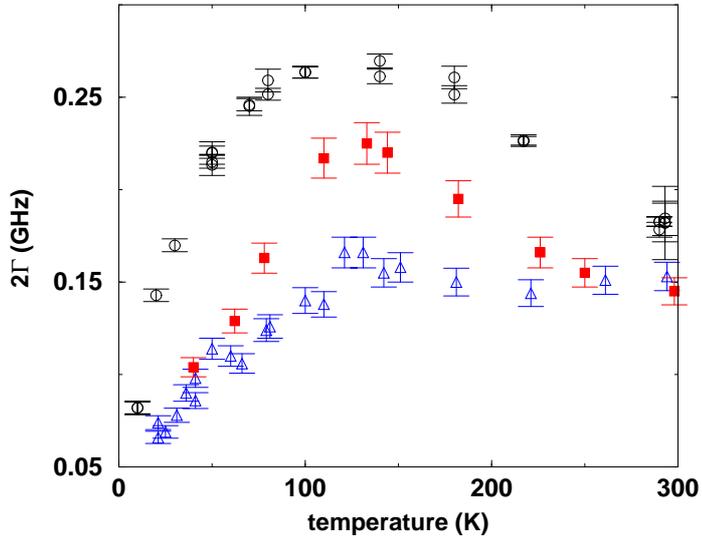


FIG. 1: Full width at half maximum of the Brillouin peak in a three silica samples plotted as functions of temperature from 5 to 300 K for the sol-gel sample ( $\circ$ ) ( $[\text{OH}]=3000$  ppm), for suprasil<sup>18</sup> ( $\square$ ) ( $[\text{OH}]=1200$  ppm) and for puropasil<sup>19</sup> ( $\triangle$ ) ( $[\text{OH}]=20$  ppm).

infrared spectroscopy). In the following, it will be referred to as the sol-gel sample. We used Brillouin scattering measurements performed with a high resolution Fabry-Pérot (FP) spectrometer.<sup>17</sup> The incident light was the 514.5 nm line of a single-mode argon-ion laser (Spectraphysics 2020) with an incident power on the sample of about 1W. This set-up comprises a double-pass plane FP, whose free spectral range is equal to 75 GHz and finesse equal to 40, used as a filter (monochromator), followed by a spherical FP interferometer with a free spectral range of 1.48 GHz and a finesse of 50 as the resolving unit. The measurements of the longitudinal sound wave velocity and attenuation are performed in backscattering geometry. The accuracy of the experimental data for the sound velocity, deduced from the Brillouin shift, and the sound attenuation, related to Brillouin linewidth, are respectively of about 0.1% and 5%. Low temperature measurements were performed using a He cryostat. High temperature measurements were carried out using a Hermann-Moritz oven with optical windows, which allows measurements up to 1500 °C. The data are analyzed using a non-linear fitting procedure. The Brillouin profiles were adjusted with a Lorentzian function of half width at half maximum  $\Gamma$  convoluted with an apparatus function measured using the elastic line of an experimental spectrum. The internal friction parameter  $Q^{-1}$  is deduced from the the full width at half maximum  $2\Gamma$  and the position  $\delta\nu$  of the longitudinal Brillouin line, using:  $Q^{-1} = 2\Gamma/\delta\nu$ .

The linewidth in the region of the minimum for sound velocity, from 5 to 300K, is plotted in Fig. 1 for the sol-gel silica, as well as for two commercial silica samples previously investigated (suprasil and puropasil)<sup>18,19</sup>. The internal friction is shown on Fig. 2a. The results for  $Q^{-1}$  in puropasil are very similar to results in another commercial sample (suprasil W characterized by a very small amount of impurities), measured using a different wavelength ( $\lambda = 488$  nm).<sup>14</sup> The attenuation versus temperature curves exhibit a large peak at low temperatures, centered around 150 K, as first observed at hypersonic frequencies long ago<sup>20</sup> and then a nearly constant value up to the glass transition temperature range. The Brillouin frequency shift for sol-gel silica and puropasil are presented from 5 to 1550 K in Fig. 2b. The frequency shift is needed to calculate the dimensionless internal friction parameter, but also to determine sound velocity. The data of Fig. 2b are close to previous determination of the literature in several commercial silica samples,<sup>21,22,23</sup> and the high temperature part of those curves will be also discussed, in comparison with previous determinations, in another article (Ref. 24).

Two main features can be deduced from the figures. First, the attenuation peak around 150K is more prominent in silica samples containing more OH impurities and having lower fictive temperature. Taking the observed high temperature plateau as a reference level, the peak height for the linewidth in sol-gel silica is about 0.12 GHz vs 0.07 GHz in suprasil, whereas it is very weak (around 0.015 GHz) in puropasil sample which contains very few OH impurities. A similar increase of the attenuation peak can also be observed in Fig. 2 of Ref. 8 where the authors report measurements of the attenuation over a limited temperature range in a densified aerogel, of density  $2.19 \text{ g cm}^{-3}$  and whose OH content is unknown. Second, within the accuracy of the experiments the values of internal friction above room temperature are almost identical in the low and high OH content silica, in fact they are slightly higher for the sol-gel silica compared to commercial samples. Variations of the sound velocity with the OH content can also

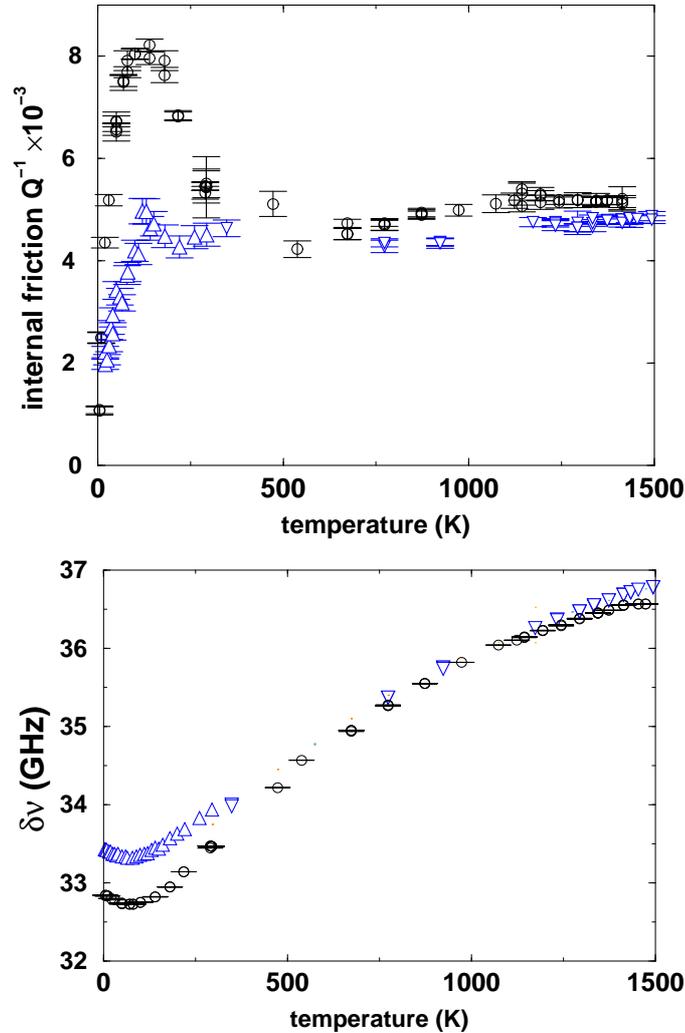


FIG. 2: a) Internal friction in a two silica samples (sol gel ( $\circ$ ) and puropzil ( $\Delta$  ( $\Delta$  for low temperature data<sup>19</sup>,  $\nabla$  for high temperature new data), plotted as a function of temperature, from 5 to 1500K. The results for the entire temperature range for the sol-gel silica and above room temperature for the puropzil are new results. The low temperature results for puropzil were published previously.<sup>19</sup> b) Brillouin frequency shift in sol-gel silica samples ( $\circ$ ), plotted as a function of temperature, from 5 to 1500 K, compared to data for a low OH content silica glass, puropzil ( $\Delta, \nabla$ ). The low temperature measurements are taken from Ref. 19, the high temperature ones are new results. The error bars account for the error in the fitting procedure, an aperture of  $\pm 1^\circ$  for the collected scattering, an error in the determination of the position of the elastic lines and of the thickness of the Fabry-Pérot.

be observed on Fig. 2b, mainly in the range of the attenuation peak and of the glass transition.

It has been proved previously that the low temperature peak has the same origin as the peak observed at lower temperature using lower frequencies, such as ultrasonic measurements. Several authors<sup>14,19,20</sup> have checked that the temperature variation of the peak position is in agreement with thermally activated processes having a distribution of activation energy and a mean activation energy in the 300-600 K range depending on the form of the distribution of energy barriers.<sup>25</sup> Using ultrasonic investigation, many studies have been devoted to the influence of the preparation method<sup>26,27</sup> and of the impurity content (for example OH impurities) on the amplitude of this peak. It can be pointed out that such a peak is observed in many glasses. As examples, the ultrasonic attenuation at 20 MHz exhibit a peak around 170 K in  $\text{GeO}_2$  and around 30 K in  $\text{BeF}_2$ <sup>26,28</sup>. In  $\text{B}_2\text{O}_3$  the amplitude and the temperature position of the peak have been strongly related to OH content.<sup>29</sup> The main difference between the three silica samples whose attenuations are presented in Fig. 1 is the OH content (20 for puropzil, 1200 ppm for suprasil and 3000 ppm for sol-gel silica). Therefore, this result demonstrates, for the first time at hypersonic frequencies, that the impurities are involved in this feature of the acoustic attenuation. This result does not mean necessarily that OH are really involved in the thermally activated jumping. OH defects can induce local structural modifications<sup>30</sup> and create entities participating in jumping.

Indirect structural effects can also be due to a decrease of fictive temperature induced by increasing the OH content. Nevertheless, such a damping peak at hypersonic frequencies related directly to the presence of OH impurities has also been observed in crystals such as KCl (Ref. 31). Observed also by infrared absorption, Raman scattering and thermal conductivity measurements, this relaxational process has been attributed to librational motions of the center of mass of the hydroxyl molecule. This observation in crystals suggest that a direct implication of OH cannot be excluded in glasses also.

If the attenuation peak is related to thermally activated processes, an associated change on the temperature dependence of velocity should be also expected. Such variation is observed on Fig. 2b). It has been pointed out, using ultrasonic measurements,<sup>27</sup> that the fictive temperature does not modify significantly the temperature dependence of velocity in the region of the attenuation peak (around 80K). The temperature variation of sound velocity in this range depend only on the OH content. Thus we could expect that the attenuation in this range depend more strongly on the OH content than on the fictive temperature. Figure 1 of Ref. 27 shows that the influence of fictive temperature on the height of the damping peak at hypersonic frequency decreases with increasing OH content. We expect the same tendency at hypersonic frequency and then the amplitude of the peak in sol-gel silica should be in very large part due to high OH content. Moreover, we have measured the fictive temperature influence at room temperature on the linewidth of a very low OH content silica glass, and we found that a variation of fictive temperature by 400 K induces only a decrease of about 10% in the linewidth (from 0.144 GHz for  $T_f = 1373\text{K}$  to 0.159 GHz for a sample with  $T_f = 1773\text{K}$ ) or in the internal friction ( $4.23 \times 10^{-3}$  to  $4.7 \times 10^{-3}$ ).

In contrast, the presence of impurities seems to have little effect on the friction coefficient at temperatures higher than 300 K. Above room temperature, the attenuation exhibits a plateau over a wide temperature range (up to more than 1500 K for the OH free sample). In contrast to the attenuation peak region, in the high temperature range, the hypersonic attenuation is comparable to that measured in crystals.<sup>32</sup> Moreover it has been pointed out that the attenuation in permanently densified silica is also very close to that of crystalline quartz.<sup>33</sup> As the relaxational process can not explain the amplitude or the frequency dependence in  $\omega^2$  of the attenuation in the high temperature range,<sup>34</sup> anharmonicity has been invoked previously as the dominant process involved in the hypersonic attenuation at high temperature in silica glass.<sup>11,35</sup> Recently new theoretical approaches have been proposed to describe some aspects of anharmonicity in glasses<sup>36,37</sup> but up to now the applicability of these models to quantitative analysis of hypersonic attenuation is lacking. Using the framework of the same formalism as for crystalline materials<sup>38</sup> the anharmonic damping can be understood as a coupling of hypersonic waves with the bath of thermal phonons. Indeed, simulations<sup>12</sup> have shown that anharmonicity process can induce an anomalously large Grüneisen parameter and sound attenuation due to larger anharmonicity in glasses than in crystals. Quantitative analysis in terms of anharmonicity has been recently successful<sup>35</sup> in describing hypersonic attenuation in OH free silica glass up to 300K. To follow the same analysis in a OH rich sample, the influence of local impurities on the phonon vibration spectra should be examined. It is expected and verified by experiments that local intrinsic disorder due to impurities does not contribute to the large variation of the vibrational spectra<sup>39</sup>. So, at high temperature when the population of high energy phonons is significant the mean thermal relaxation of phonons is little modified by the impurities. Therefore, in the framework of the interpretation of an attenuation due to the interaction of hypersonic waves with the whole thermal phonon bath, it follows that the attenuation in this frequency and temperature range is only weakly affected by impurity content as shown in our experiments.

The main contribution to acoustic attenuation in silica glass, measured at hypersonic frequencies by Brillouin scattering for temperature lower than 300K, is a large relaxation peak due to an interaction with structural defects strongly related to extrinsic impurities such as OH impurities. Comparison of acoustic attenuation at various frequencies are often made in the literature. However, the large variations of the damping amplitude which could occur due to the relaxational process related to impurities, require that the origin and impurity content of the samples under comparison should be taken into account. The relaxational contribution, due to thermally activated processes, dominates only at low temperatures and does not account for the high temperature regime of the attenuation where anharmonicity is likely to be the dominant process.

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