Universal behavior of internal friction in glasses below T_g: Anharmonicity versus relaxation

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Comparison of the internal friction Q^{-1} at hypersonic frequencies between a few K and the glass transition temperature T_g for various glasses brings out general features. At low temperature, Q^{-1} is only weakly dependent on the material. At high temperature but still below T_g the internal friction for strong glasses shows a T-independent plateau in a very wide domain of temperature; in contrast, for fragile glass, a nearly linear variation of Q^{-1} with T is observed. Anharmonicity appears dominant over thermally activated relaxational processes at high temperature.

I. INTRODUCTION

Physical properties at low temperature, the mechanism of the glass transition, and the structure and vibrations at the nanometer scale account for many experimental and theoretical contributions in glasses.[1–8] Search for universal behavior or correlations between these properties are the mains fields explored. For this purpose, it has been useful to classify glasses using the concept of fragility[9] depending on how the viscosity (or structural relaxation) variation versus temperature deviates from Arrhenius behavior. One of the most debated questions arises from the nature and the origin of the fast relaxation[3,4] responsible for a broad quasielastic contribution in neutron, x-ray and light scattering, and also for damping in sound waves and dielectric loss.[10] From analysis of susceptibilities, a proportionality between quasielastic neutron or light scattering, infrared absorption, and internal friction or sound attenuation has been established and experimentally verified in a number of glasses.[11–16]

On the one hand, at very low temperature and for a large frequency range, many physical properties (among those the internal friction Q^{-1}) can be quantitatively described within the framework of the "tunneling model"[1,2] assuming a phenomenological potential with two asymmetric wells with a distribution of barriers and asymmetries. To improve the description of data above a few K the so-called "soft potential model"[17] extends and generalizes the previous model including distributed harmonic oscillators. At higher temperature, a phenomenological description of coupling of acoustic waves with unspecified thermally activated defects gives a quantitatively correct description of the acoustical attenuation, or equivalently internal friction, at ultrasonic frequencies assuming distributions of energy barriers.[18,19] Recently[20] this model has been extended to describe data for a large frequency range within the same formalism and to deduce the distribution of the energy barriers. Moreover, some authors claim that inelastic light-scattering results can be accounted for by this description for many orders of magnitude in frequency in silica glass[21] as well as in various fragile glass.[22] In contrast, theoretical calculations explain acoustical damping in amorphous silicon and fused silica by anharmonicity for frequencies in the 10–100-GHz range.[23] Furthermore, analysis and discussion of data obtained using picosecond optical techniques in the same frequency range conclude that

classical relaxation theory cannot explain the frequency and temperature variation observed.[24] Indeed, a description of the sound velocity (or elastic constants) versus temperature, even at low frequencies, cannot be accounted for assuming this relaxational process alone.[25–28]

On the other hand, at hypersonic frequencies available from Brillouin scattering experiments, an attempt to describe experimental results for Q^{-1} in silica glass using the soft potential model[29] did not give satisfactory results when the temperature exceeds 10–20 K. Moreover, comparisons of ultrasonic and hypersonic attenuation in glasses have demonstrated that thermally activated relaxations, dominant for temperatures higher than 10 K at low frequencies, cannot explain quantitatively the values observed at higher frequencies, not only in silica glass (Refs. 30 and 31) but also in other glasses (Refs. 32 and 33). Taking into account that the amplitude of sound attenuation is not very different at high temperature in glasses and in crystals, anharmonicity has been invoked, by analogy with processes well known in crystals, to explain results at hypersonic frequencies. However, other authors[18] explain the same Q^{-1} data in silica glass using the formalism of thermally activated processes. More recently careful analysis in silica glass has quantified the relative parts of different processes responsible for the internal friction[34] and demonstrate that anharmonicity dominates at high temperature. Finally, the frequency dependence of the damping of vibrations up to the THz range yield conflicting interpretations.[35–40] Various frequency dependences at different temperatures are attributed to different origins such as relaxational, anharmonic, or nondynamic processes. Recently from measurements with ultraviolet Brillouin light-scattering experiments[41,42] questions about the structural or dynamic origin of sound scattering were again put forward.

So the description in the literature of the origin of the sound attenuation in glasses presents contradictory interpretations and it is not clear if the frequency and temperature variation of the internal friction can be interpreted in a formalism common to all glasses. Furthermore, most of the Q^{-1} experimental determinations concern temperatures lower than room temperature and most of the data below T_g focus on fragile glasses, and on silica glass as representative of strong glasses. As silica glass, like other tetrahedrally coordinated glasses, shows a number of specific unexplained anomalies,[43–45] the question arises if the behavior observed in silica is similar to that in other glasses, as suggested in
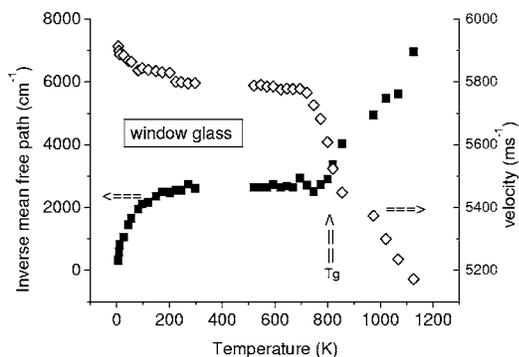
FIG. 1. Left axis: Longitudinal sound velocity vs temperature in a window glass (see composition in Table I) measured at hypersonic frequencies by Brillouin scattering in the backscattering configuration with $\lambda = 514.5$ nm. Right axis: inverse mean free path vs temperature in the same glass.

Ref. 34. To bring some light to these fields, we have made a comparison of results obtained for internal friction at hypersonic frequencies in a number of glasses. This comparison reveals characteristics common to all materials at low temperature and in the temperature range below T_g.

II. EXPERIMENTS AND RESULTS

The general behavior of hypersonic properties of glasses is illustrated in Fig. 1 which gives results for the sound velocity and hypersonic attenuation, shown through the inverse mean free path, in window glass (72% SiO_2, 14% Na_2O, 9% CaO, 3% MgO, 1% Al_2O_3 plus other minor oxides). These data were obtained from Brillouin scattering experiments with experimental conditions common to a series of previously published papers.[30,34,46]

The Brillouin frequency shift $\Delta\nu$ is related to the longitudinal sound-wave velocity V_l, the refractive index n, and the scattering angle θ by

$$\Delta\nu = 2nV_l(\sin\theta/2)/\lambda, \qquad (1)$$

where λ is the wavelenght of the incident light. From the full width of the Brillouin line Γ one can deduce either the mean free path L^{-1},

$$L^{-1} = \Gamma \times 2\pi/V_l, \qquad (2)$$

or the internal friction Q^{-1}, defined as

$$Q^{-1} = \Gamma/\Delta\nu. \qquad (3)$$

The accuracy of the experiments is about 0.1% for the determination of sound velocity and 5–10 % for the mean free path (or the internal friction).[65]

Figure 2 shows results in the full temperature range investigated for the two more standard glass formers, SiO_2 (Refs. 34, 46, and 47) and B_2O_3 (Refs. 48 and 49) together with window glass. At least two characteristics common to these glasses can be pointed out. First, in a large range of temperature, Q^{-1} is temperature independent within the accuracy of the experiments. Similar results have been found for other strong oxide glasses.[15,32,50] Second, at the glass transition

temperature, a strong increase of the internal friction is observed. This feature is associated with the α relaxational processes coupled to acoustic waves. A signature at T_g is also observed for the sound velocity or other related elastic constants (Fig. 1).

Another important feature common to various glasses, not pointed out before, is displayed in Fig. 3 where results for temperatures lower than 150 K were considered. Below this temperature, the amplitudes of the internal friction tend to a common behavior for oxide glasses as different as silica, boron oxide, or window glass. A peak reminiscent of the ultrasonic one, well identified in pure silica, does not appear for any other glass, including the fragile glasses shown in Fig. 4. In this figure, Q^{-1} values are compared for four examples of fragile glass: one electrolytic glass, $LiCl\cdot4H_2O$,[51] one organic polymer [poly (methyl methacrylate) (PMMA)],[52] glycerol,[53] and one inorganic chainlike phosphate glass.[54] The amplitude of the internal friction is similar in the strong window glass and in the fragile electrolyte glass, at low temperature, but significant differences between samples can be observed just below T_g.

When the temperature increases, the internal friction continues to increase in fragile glasses whereas it levels off in
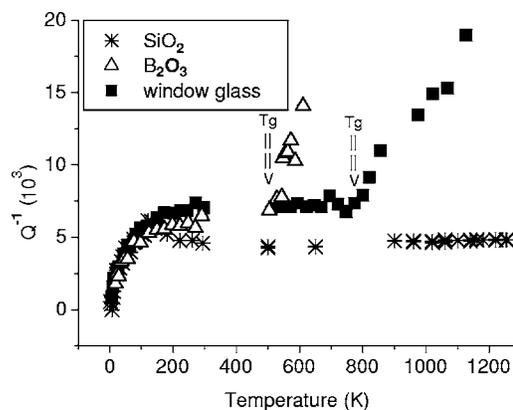

FIG. 2. Comparison of internal friction in strong glasses: silica (Refs. 46 and 34 for the low-temperature part and Ref. 47 for the high-temperature part), boron oxide (Ref. 48 for the low-temperature part and Ref. 49 for the high temperatures), and window glass.
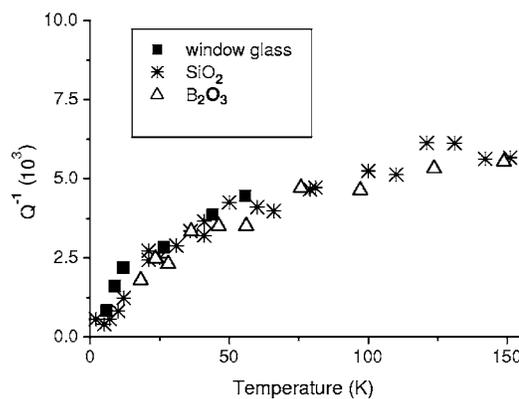

FIG. 3. Zoom on the low-temperature part of internal friction in strong glasses as in Fig. 2.
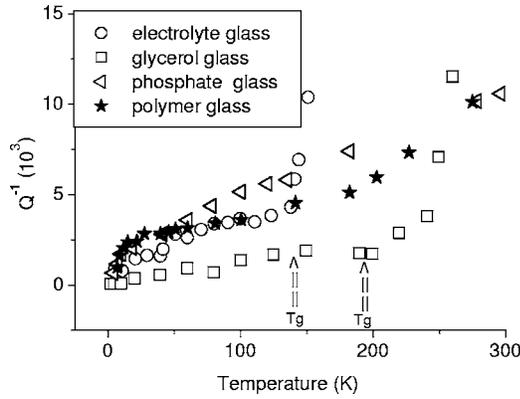
FIG. 4. Comparison of internal friction at low temperature in an electrolytic glass LiCl-4H$_2$O (Ref. [51]), polymer PMMA (Ref. [52]), phosphate glass (Ref. [54]), and glycerol (Ref. [53]).

strong glasses. This behavior has also been demonstrated in other polymeric glasses (polystyrene[58]) or in an optical strong glass[32] above room temperature.

III. DISCUSSION

As pointed in the Introduction, thermally activated relaxational processes have been invoked to explain mechanical damping in glasses below T_g. Those processes are useful to describe peaks observed at ultrasonic frequencies. As the same defects characteristic for the disorder are partly responsible for the very low-temperature properties, the distribution function of barriers and asymmetries of the tunneling model useful to describe internal friction and sound velocity below a few K are the starting point to consider thermally activated processes when the temperature is increased.[1,18] Different hypotheses follow to extend the model and concern the form of the distribution functions. Assuming a flat distribution of asymmetries, Q^{-1} is proportional to the imaginary part of the susceptibility, and can be expressed as a function of $g(V)$ the temperature-independent distribution function of barriers V and $\tau = \tau_0 \exp V/kT$ the relaxation time for hopping between adjacent potential wells.[18,21] At hypersonic frequencies and sufficiently high temperature $\omega \tau_0 \ll 1$ is satisfied, τ_0 being the fastest relaxation time, and $\omega = 2\pi\Delta\nu$. Then

$$Q^{-1} \simeq Tg(V). \tag{4}$$

A maximum in Q^{-1} in ultrasonic experiments implies a cut-off V_{max} in $g(V)$. Different distributions $g(V)$ have been tested in the literature but the general characteristic temperature dependence of Q^{-1} can be discussed without a precise description of $g(V)$.

The rough proportionality to T observed at hypersonic frequencies up to near 100 K for strong glasses or below T_g for other more fragile glasses can be attributed to this process [Eq. (4)]. The similarity of values obtained in different glasses can be related to the parameters of tunneling models deduced from experiments at very low temperature which show close values for strong and fragile glass.[7,59] Q^{-1} does not vary much for different glasses, about a factor of 2, as the parameter characteristic for the contribution of tunneling de-

fects to the internal friction, as pointed out by Pohl.[7] One can note the astoninshingly low value of Q^{-1} for glycerol. For most glasses, in ultrasonic experiments the Q^{-1} peak is broader and appears at higher temperature than in silica glass;[60] a broadened distribution function with higher potential barriers gives a good fit of the data but calculations at hypersonic frequencies of the contribution of these thermally activated processes using the same parameters predict an amplitude much lower than experimental values[30–32,34] or a maximum at a temperature higher than T_g,[33] so that another process, anharmonicity, can be put forward at high temperature.

The second process under consideration is anharmonicity. It can be pointed out that the origin of this anharmonicity can be specific to glasses as developed by the authors of Refs. [55], [61], and [62] but a quantitative test is not possible in the currently available models. Furthermore, these authors have pointed out that fragility is correlated with anharmonicity[55,61,62] and this contribution should occur also in fragile glasses; this has been recently confirmed by molecular-dynamic simulation.[63] The similarity between $Q^{-1}(T)$ curves in glasses and crystals at high temperature suggests that the same formalism can be used in both materials.

In the Akhieser regime[64] using the same formalism of network viscosity as in crystals, the dominant process expected in glasses can be expressed as[23]

$$Q^{-1} = A\omega\tau_{ph}/[1 + (\omega\tau_{ph})^2] \tag{5}$$

with $A = \gamma^2 C_v T V_l / 2\rho V_d^3$ where C_v is the specific heat per unit volume, γ is the Grüneisen constant, V_l is the longitudinal sound velocity, ρ is the density and V_d is the Debye velocity, τ_{ph} is the mean lifetime of thermal phonons. More precise calculations of anharmonicity[34] showed that a plateau is observed at high frequencies.

At high temperature, but below T_g, C_v in glasses presents a slow T variation and the Debye model gives a good quantitative approximation; moreover, in silicate glasses the C_v value is weakly dependent of the chemical composition.[56] Assuming τ is proportional to T^{-1} (verified, for example, in various forms of silica in Ref. [34]) the product $C_v T\tau$ is a constant and can explain the leveling off for the plateau $Q^{-1}(T)$ observed in oxide glasses. Such a contribution also exists in fragile glass and is superimposed on a relaxational one which determine the observed T dependence. In order to quantify the relative part of these two contributions, the anharmonic contribution Q^{-1} deduced from Eq. (5) are calculated and given in Table I for the glasses for which the parameters are known or can be estimated. The Grüneisen parameters γ are taken from the article of Novikov,[55] the specific heat per unit volume from Ref. [56], Debye velocity V_d is obtained from Pohl[7] or from our own measurements. τ_{ph} is taken to be equal to 10^{-13} s, as often in the literature.[34] This lower limit for τ_{ph} provide a lower limit value for Q^{-1}. Due to uncertainties on some parameters (γ and τ_{ph}) only a rough estimate can be made but it appears that an important part of the internal friction can be attributed to anharmonic process in all glasses from room temperature to T_g. Concerning the anomaly of glycerol, for temperatures lower than

TABLE I. Comparison of experimental Q^{-1} and calculated anharmonic contribution of Q^{-1}. The experimental data are given at 300 K. For silica and boron oxide, the experimental values are identical at T_g and room temperature. Data for SiO_2 are from Refs. 46 and 47, data for B_2O_3 from Refs. 48 and 49, data for PMMA from Ref. 52. Anharmonic contribution Q^{-1}, calculated at T_g for SiO_2 and at 300 K for B_2O_3 and PMMA. The calculated values are estimated from Eq. (5). The values of density, Debye and longitudinal sound velocity, specific heat, and Grüneisen parameter used for the calculation are also given in the table. We used $\tau_{ph}=10^{-13}$ s, the Grüneisen parameter γ from Ref. 55 and C_v from Ref. 56 for SiO_2 and PMMA, and from Ref. 57 for B_2O_3.

	Chemical composition	T_g (K)	$\Delta\nu$ (GHz)	Q^{-1} $\times 10^3$	ρ (g cm^{-3})	V_d (km s^{-1})	V_l (km s^{-1})	C_v (J cm^{-3} K^{-1})	γ	Calc. anharm. $10^3 \times Q^{-1}$
Silica glass	SiO_2	1400	33.6	4.8	2.2	4.0	5.9	2.64	1.8	4.9[a]
Boron oxide glass	B_2O_3	526	20.0	6.8	1.8	2.0	3.5	2.16	2.7	6.8
Polymer glass	poly (methyl methacrylate) (PMMA)	370	16.3	10.1	1.2	1.6	3.15	1.75	2.5	10.1

[a]The value of the anharmonic contribution in silica deduced by subtraction of the relaxational part is estimated to be equal to $Q^{-1}=2.8 \times 10^{-3}$ (Ref. 34).

150 K, the contribution of anharmonicity to internal friction is expected to be negligible (the condition $\omega\tau_{ph}\ll 1$ may not be fulfilled). On the other hand, the parameter characteristics for the tunneling amplitude is the same as for silica,[7] so that we do not have an explanation for the astonishingly low values of Q^{-1} in glycerol.

Finally, to verify the consistency of the results, the velocity variation vs temperature can be considered. This is not possible in silicate glasses as structural anomalies dominate the elasticity.[34] Below T_g the variations $V(T)$ in glasses other than silica are linear. From the relative slope $(1/V_l) \times (\delta V_l/\delta T) = \beta\gamma$ where β is the thermal expansion, γ is determined for the mode under study and can be different of γ in Eq. (5) where Q^{-1} is calculated in relation with the thermal bath of phonons. An estimation of the Grüneisen constant γ of the mode can be deduced; we found $\gamma=3$ for both boron oxide and PMMA; these values correlate well with the literature data used for the independent Q^{-1} calculation (2.7 and 2.5, respectively, from Ref. 55).

IV. CONCLUSION

Our contribution demonstrates that, at hypersonic frequencies accessible by Brillouin scattering experiments, the internal friction Q^{-1} in strong oxide glasses is basically T independent above room temperature up to the glass transition. Moreover, the relaxation peak observed in vitreous silica or tetrahedrally coordinated glasses is not observed in any other glass.[66]

In strong and fragile glasses, Q^{-1} differs from that of crystals mainly at low temperature and so the same mechanisms of sound attenuation can be invoked in these two classes of materials at high temperature. Additional contributions from activated relaxational processes specific to glasses are efficient at low temperature and in more fragile glasses and can explain the rough proportionality to T observed. The small variation with chemical composition of the internal friction in the hypersonic regime below 100 K extend the remark about universality of low-temperature properties pointed out in the review paper of Pohl toward high frequencies and toward higher temperatures than expected.[7]

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[65] It can also be noted that our results concern only longitudinal waves for which good accuracy can be obtained for the Brillouin linewidth determination but it has been pointed before (Ref. 30) that the behavior is similar for transversal waves in this frequency range.

[66] In GeO_2, analog to SiO_2 structure, Q^{-1} shows also a very broad Brillouin peak near 300 K; such a peak disappears in chemically multicomponent glasses.