Brillouin light-scattering linewidth measurements from silica glass

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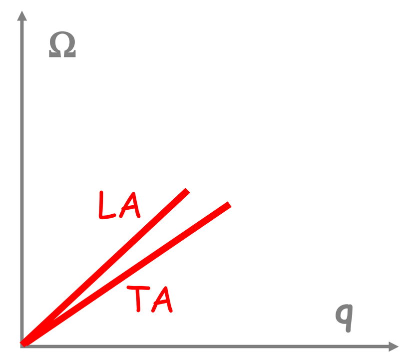
1. Motivations

Acoustic waves (Ω,q) in glasses



Linear dispersion $\Omega = vq$

Energy mean free path l^{-1} (spatial attenuation α) or internal friction Q^{-1}

$$l^{-1} = \alpha = \frac{2\Gamma}{v} \qquad Q^{-1} = \frac{2\Gamma}{\Omega} = \frac{l^{-1}v}{\Omega}$$

In crystals mechanisms for Q^{-1} are phonon relaxations via anharmonic interactions (Akhiezer), $\Gamma \propto \Omega^2$

In glasses several mechanisms lead to sound attenuation and dispersion :

- coupling with two-level systems (TLS) dominant below 3K
- coupling with thermally activated relaxations (TAR) of structural « defects » : dominant at sonic and ultrasonic frequencies
- anharmonicity or « network viscosity »
- Rayleigh scattering of the sound waves by static density or elastic constant fluctuations
- coupling with modes in excess (Boson peak) → end of acoustic branches (?)

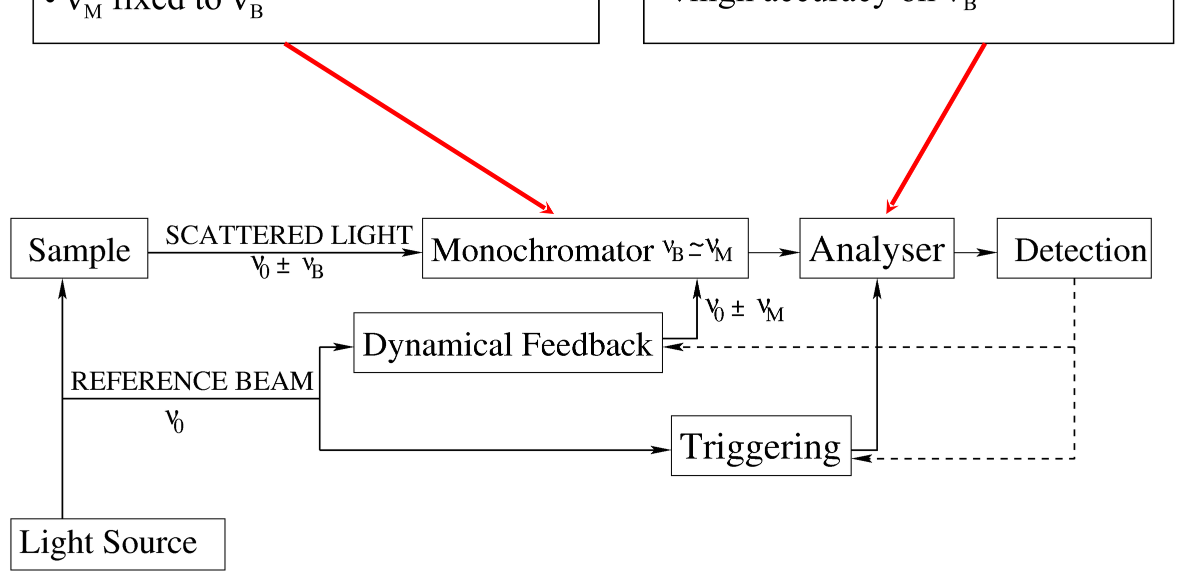
Damping of sound and velocity dispersion

Relative variation of sound velocity : $\frac{\delta v}{v} = \frac{v(\Omega,T) - v_0}{v_0}, \quad v_0 = v(\Omega, T \to 0)$

Q^{-1} and $2\delta v/v$ are Kramers-Krönig transforms of each other : $-\frac{2\delta v(\Omega,T)}{v} = \frac{1}{\pi} P\int_{-\infty}^{+\infty} \frac{Q^{-1}(x,T)}{x-\Omega}dx$

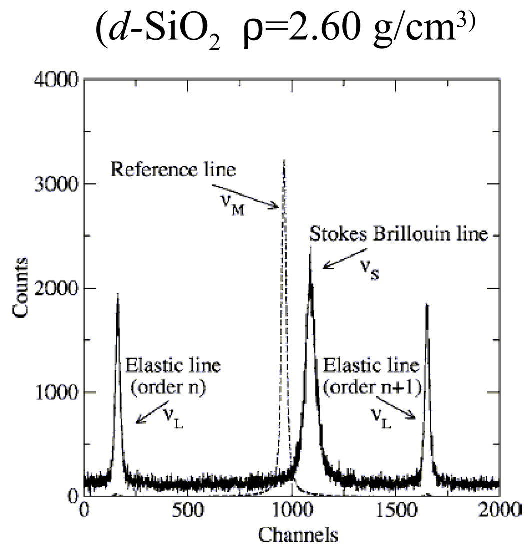
2. High resolution Brillouin light spectroscopy

Plane Fabry-Perot (PFP)	Spherical Fabry-Perot (SFP)
Free Spectral Range (FSR)~100 GHz	FSR ~1.5 GHz
4 passes ⇒ high contrast C ~ 10^{10}	instrumental Half-Width ~15 MHz
• stabilized with a reference signal generated by electro-optic modulation of the laser light ($\nu_0 \pm \nu_M$)	calibration with the reference signal ν_0
• ν_M fixed to ν_B	⇒high accuracy on ν_B



Typical spectrum in densified silica

(d-SiO_2 ρ=2.60 g/cm³)



High resolution and accuracy

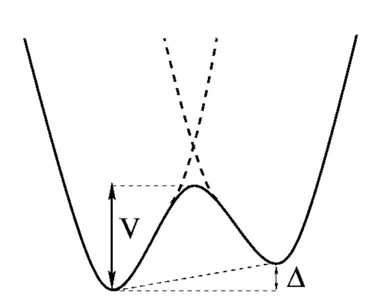
$\nu_B = 42.320 \pm 0.003$ GHz

$\Gamma = 25 \pm 3$ MHz

[H. Sussner, R. Vacher, Appl. Opt. **18** 3815 (1979) ● R. Vacher, H. Sussner, M. Schickfus, Rev. Sci. Instrum. **51** 288 (1980) R. Vialla, Opt. Instrum., Coll. de la société française d 'optique, ed. Bouchareine (1996) ● E. Rat et al, PRB, **72** 214204 (2005)]

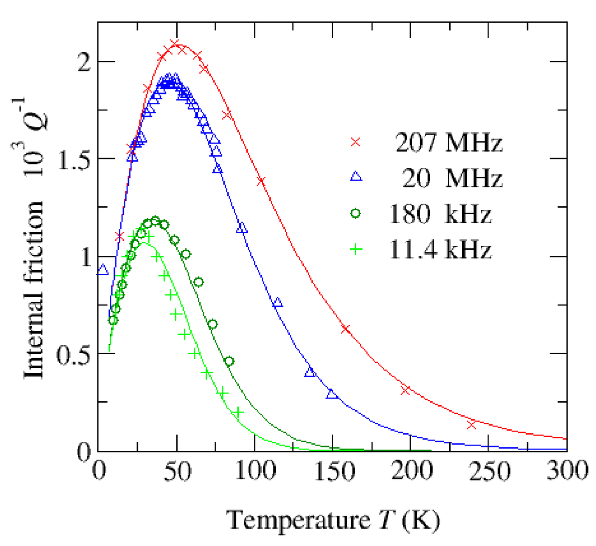
3. Sonic and ultrasonic data

Thermally activated relaxations (TAR) : atoms or groups of atoms move in a double-well potentiel → when the temperature is sufficient, the atoms jump above the barrier V with thermic activation



[W.A.Phillips,J.Low.Temp.Phys. **7** 351 (1972) ● P.W.Anderson, B.I.Halperin and C.M.Varma, Philos.mag **25** (1972)]

Ultrasonic data for v-SiO_2



× R.Vacher *et al* JNCS **45** 397 (1981)

Δ O.L.Anderson *et al* J.Am.Ceram.Soc. **38** 125 (1955)

O R.Keil *et al* JNCS **164** 1183 (1993)

+ D.Tielbürger *et al* PRB **45** 2750 (1992)c

Random defects distribution

$$P(\Delta, V) = f(\Delta)g(V) \quad \begin{cases} g(V) \propto V^{-\xi}\exp\left(-\frac{V^2}{2V_0^2}\right) \\ f(\Delta) \propto \exp\left(-\frac{\Delta^2}{\Delta_c^2}\right) \\ \tau = \tau_0 \exp\left(\frac{V}{T}\right)\operatorname{sech}\left(\frac{\Delta}{2T}\right) \end{cases}$$

Parameters of the TAR model

V_0=659±19 K (high value of V_0)

$\log_{10}\tau_0$=-12.2 ±0.18

V_0/Δ_c=8.2 ±0.6 (small cutoff value Δ_c)

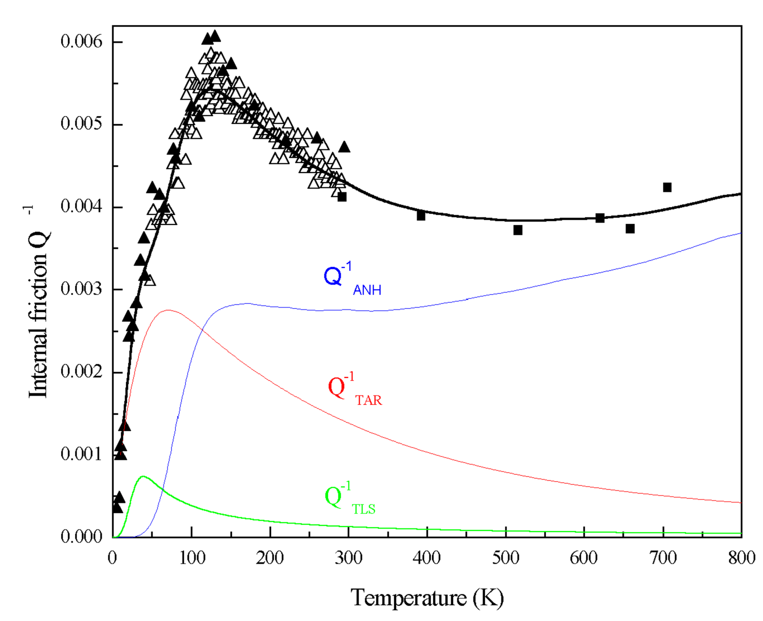
ξ=1/4

C=1,4.10⁻³ (constant value over 4 decades)

[R.Vacher, E.Courtens and M.Foret, PRB **72** 214205 (2005)]

4. Brillouin light scattering data

v-SiO_2, 35 GHz



▲ R.Vacher J.Pelous PRB **14** 823 (1976)

■ J.Pelous R.Vacher, Sol.Stat.Com. **18** 657 (1976)

△ D.Tielbürger et al, PRB **45** 2750 (1992)

Anharmonic processes :

interaction of the acoustic wave with the thermal phonons. The return to the equilibrium is characterized by τ_{th} (mean thermal lifetime).

$$\Omega\tau_{th} << 1 : \begin{cases} Q^{-1}_{anh} = A\Omega\tau_{th} \\ \delta v/v = -A/2 \end{cases} \qquad A = \frac{\gamma_{th}^2 C_v T v}{2\rho v_D^3}$$

Otherwise : $Q^{-1}_{anh} = A\Omega\tau_{th}/1 + \Omega^2\tau_{th}^2$

[H.J.Maris *in Physical Acoustics* Vol. VIII (1971)]

Mathiessen rule : $Q^{-1} = Q^{-1}_{TAR} + Q^{-1}_{anh} + (Q^{-1}_{TLS})$

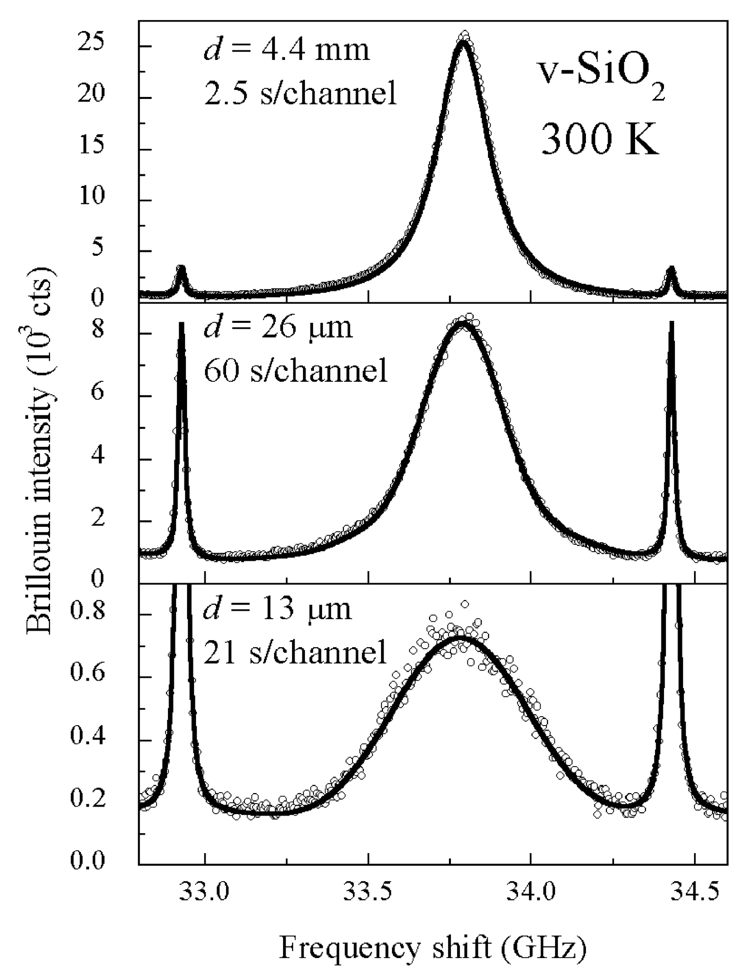
5. Broadening due to finite size effects

[R.Vacher, S.Ayrinhac *et al* (2006)]

Restricted scattering volumes ⇒ additional broadening resulting from the spread in Q

• **Brillouin scattering in thin samples**

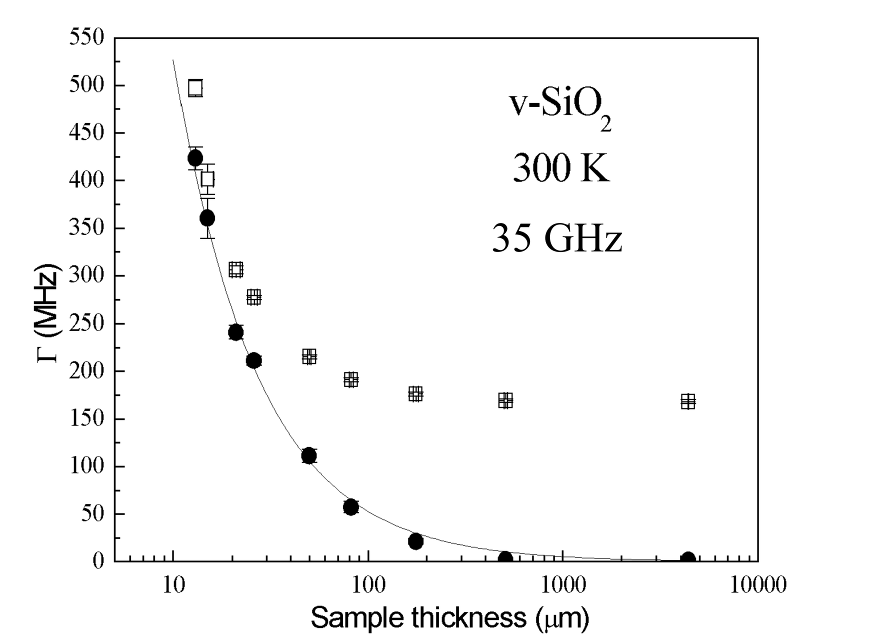
Brillouin Stokes spectra in backscattering geometry (λ_0 = 514.5 nm)



Broadening is characterized by :

$$f(\omega) \sim \left[\frac{\sin[(\omega-\Omega)d/2v]}{[(\omega-\Omega)d/2v]}\right]^2$$

[A.Dervisch and R.Loudon, J.Phys.C : Solid State Phys. **9** L669 (1976) J.R.Sandercock, PRL **29** 1735 (1972)]



● thickness dependence of the width of the function f(ω) derived from fits of the measured spectra to a DHO convoluted with both the instrumental function **and** f(ω).

— calculated width of f(ω) using the known values of thickness.

❑ apparent width resulting from fits of measured spectra to a DHO convoluted with instrumental function.

• **Brillouin scattering in strongly absorbing medium** for the exciting radiations

Additionnal contribution to the width in case of light absorption [J.R. Sandercock, PRL **28** 237 (1972)]

$$\frac{\Delta\Omega}{\Omega} = \frac{2n_2}{n_1} = \frac{\alpha_0\lambda_0}{2\pi n_1} \qquad \alpha_0 \text{ absorption coefficient}$$

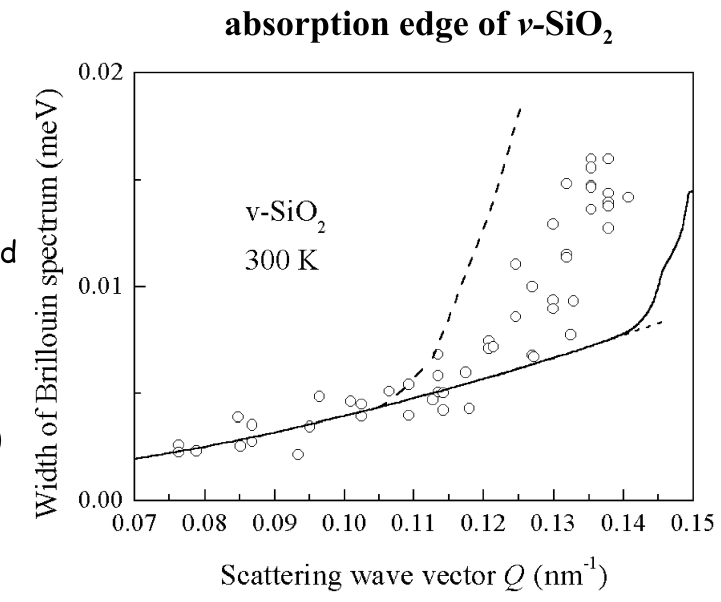
❍ **experimental data** from C.Masciovecchio, G.Ruocco *et al* presented at 5th IDRMC (Lille-2005)

calculated width ($\Delta\Omega+\Gamma_{anh}$) using Γ_{anh}= AQ² and (n_1,n_2) data from :

— A.Appleton *et al* in *The physics of SiO_2 and its interfaces* (1978)

--- G.L.Tan *et al*, PRB **12** 205117 (2005)

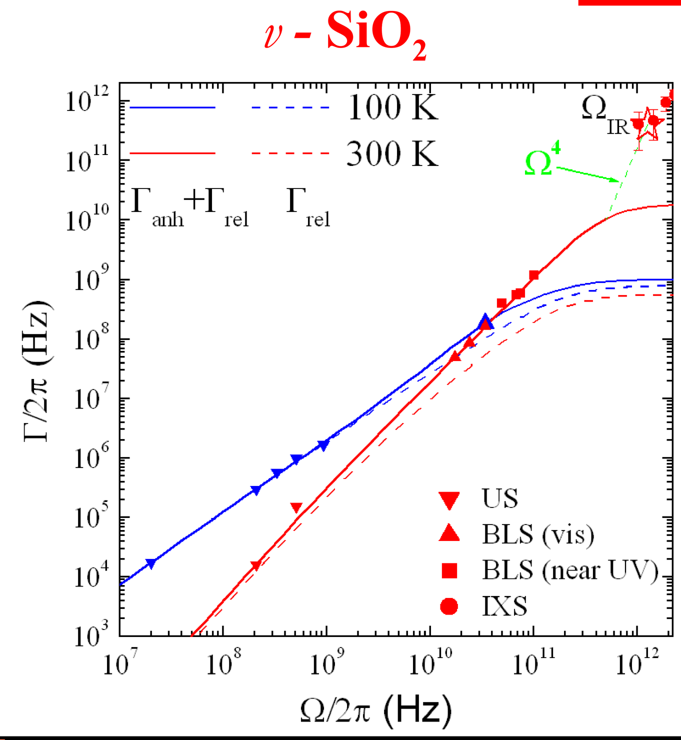
Brillouin width near the UV absorption edge of v-SiO_2



⇒ The apparent rapid increase in the sound damping near 0.12 nm⁻¹ might simply result from light absorption.

6. Overall frequency dependence of damping

v - SiO_2



[E. Courtens, B. Rufflé, R. Vacher, Journal of Neutron Research (2006)]

⇒ Ioffe-Regel crossover (Ω_{IR}) at resonance with the boson peak modes

→ end of acoustic branches

R.Vacher *et al* JNCS **45** 397 (1981)

O.L.Anderson *et al* J.Am.Ceram.Soc. **38** 125 (1955)

R.Keil *et al* JNCS **164** 1183 (1993)

D.Tielbürger *et al* PRB **45** 2750 (1992)c

■ C.Masciovecchio *et al* PRL **92** 247401 (2004)

P.Benassi *et al* PRB **71** 172201 (2005)

● C.Masciovecchio *et al*, PRL **76** 3356 (1996)

To summarize, there exist several sound damping mechanisms glasses whose strength generally depends on the material and on T. Several crossovers may be present in Γ(Ω) and a single law Γα Ω² is generally not meaningful. The analysis of sound damping requires high quality measurements over a broad range of Ω.

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