High-frequency dynamics in liquids and glasses at the mesoscopic scale

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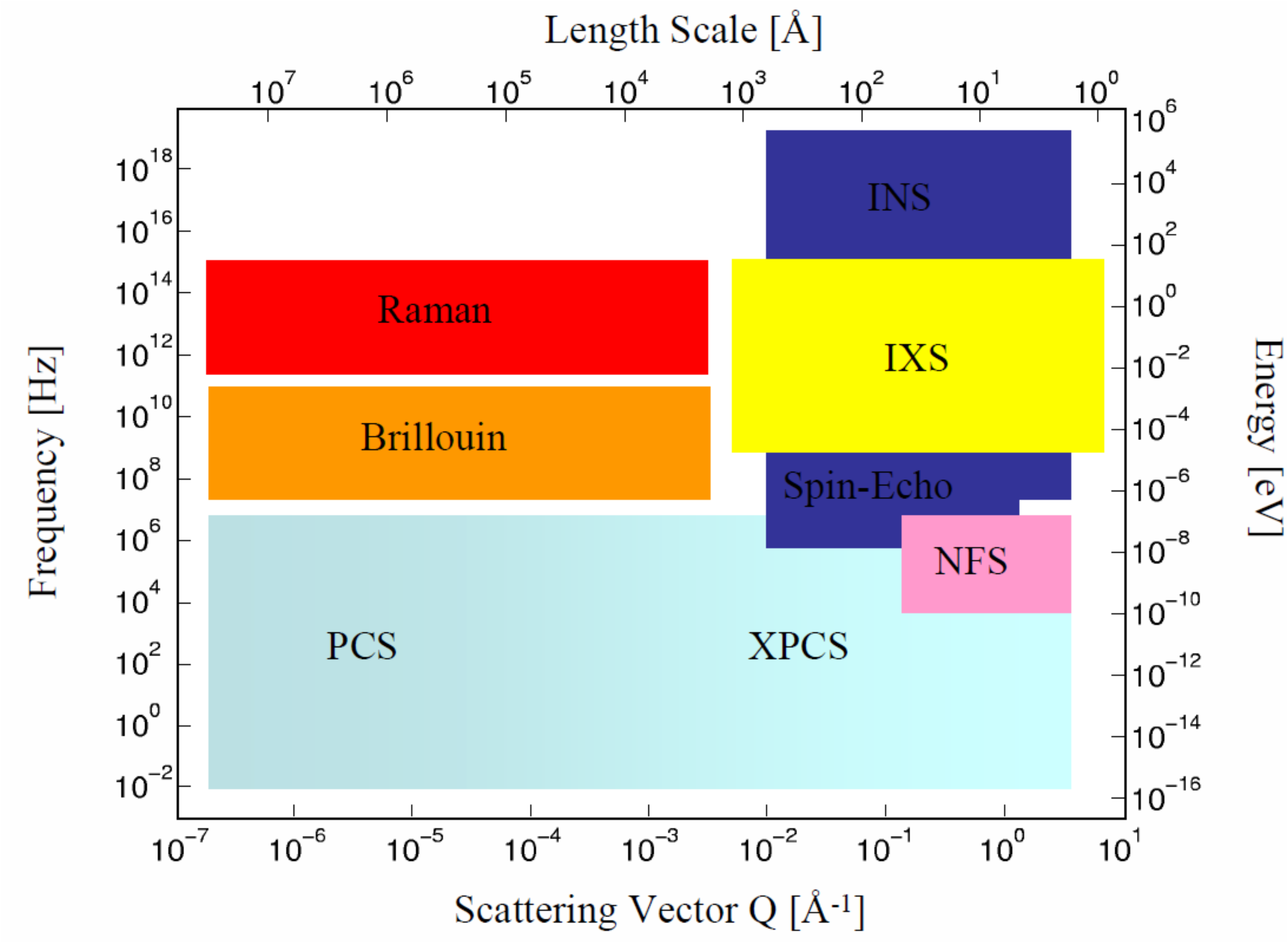
outline:

- IXS: science case for amorphous systems
- IXS applications in liquids
- IXS applications in glasses

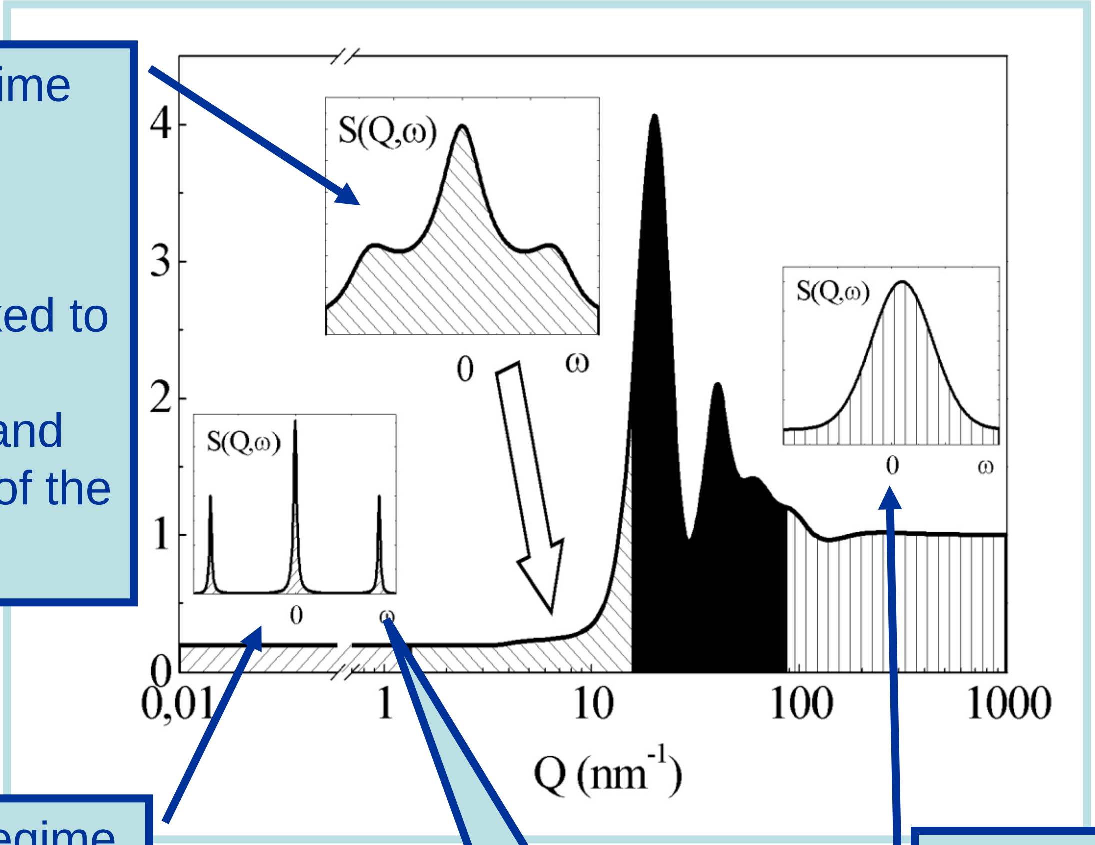
IXS & disordered systems



- Interplay between structure and dynamics
- Vibrations & relaxations at the picosecond time scale

Long standing puzzle of the high-frequency dynamics

Microscopic regime
→ not complete understanding
→relaxation processes invoked to account for the spectral shape and the broadening of the excitations

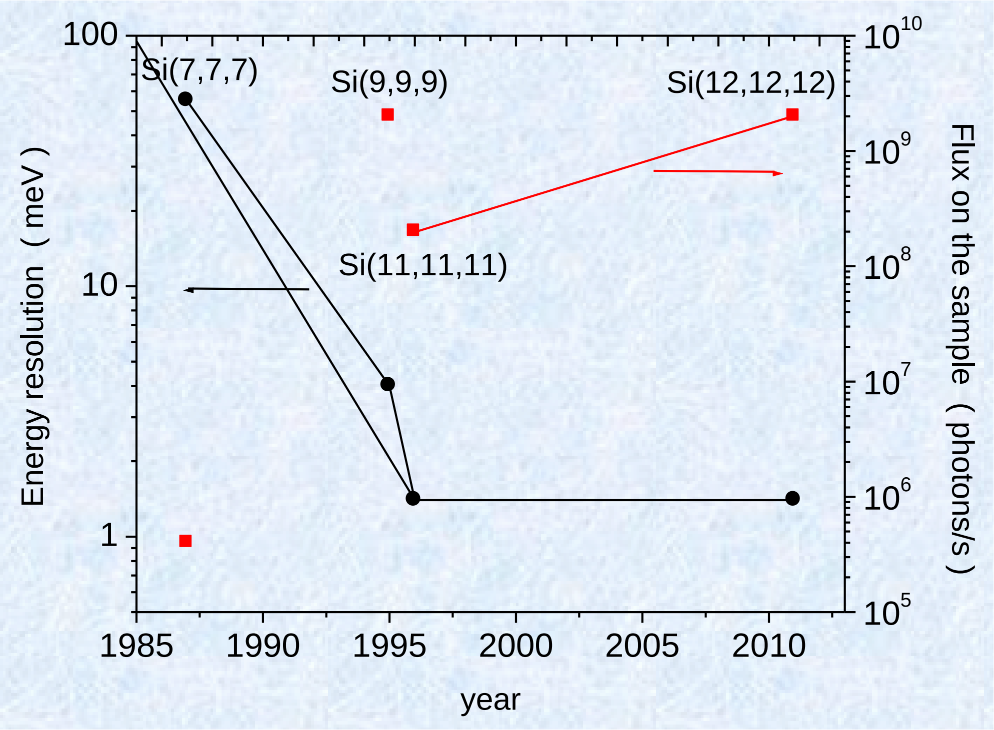


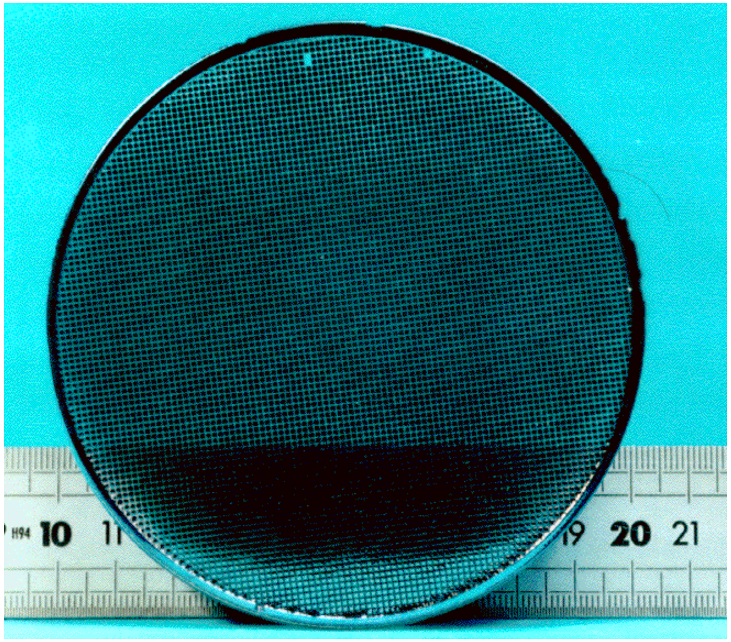
Macroscopic regime
→ hydrodynamics

v=ħω/q

Free particle regime:
impulse approximation

1992-2016 – development of IXS @ ESRF

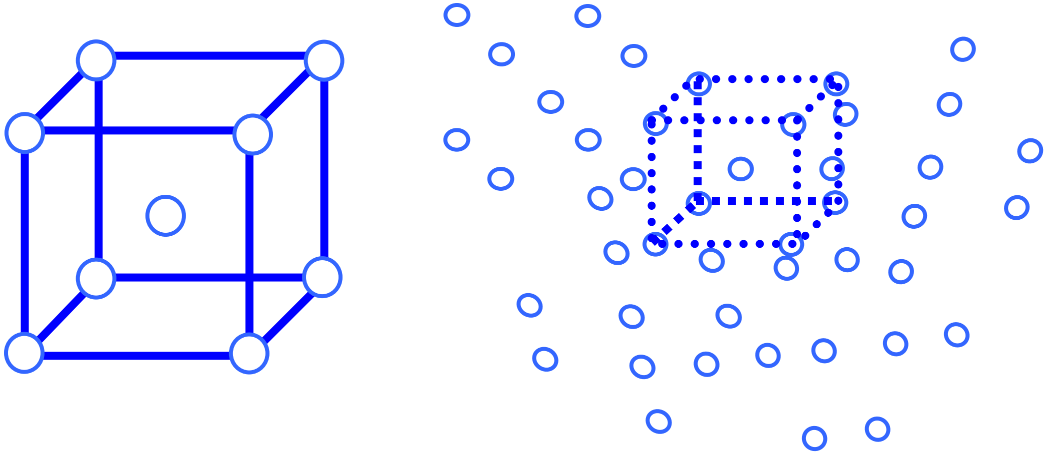


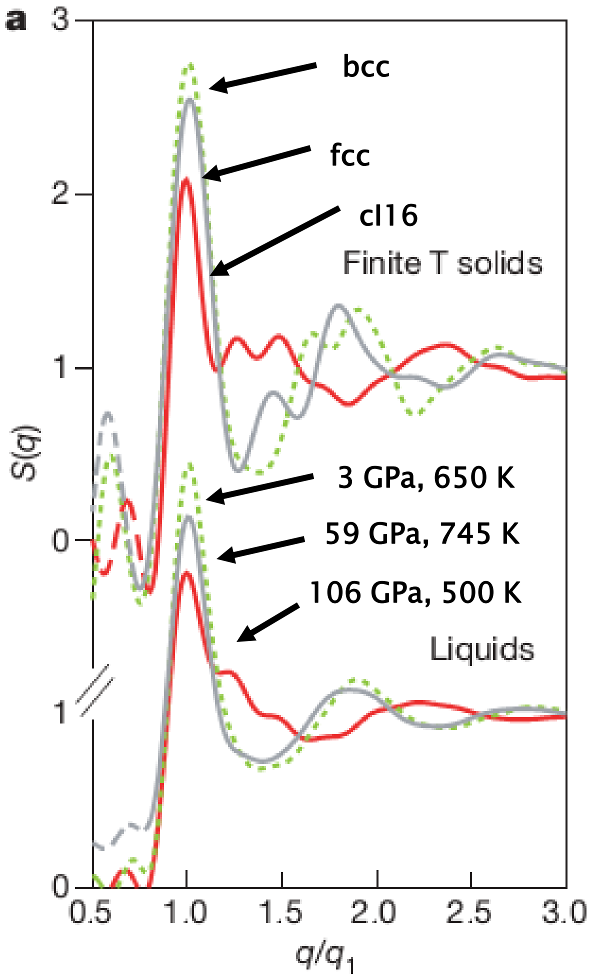


- IXS feasible at large third generation synchrotron radiation sources (ESRF, APS, SPring-8)
- steady development of undulators since the 1990s

-~10.000 cubes of 0.6x0.6x2.3 mm^3
- perfect crystal properties
- collection of sufficient solid angle

Short range order in liquids



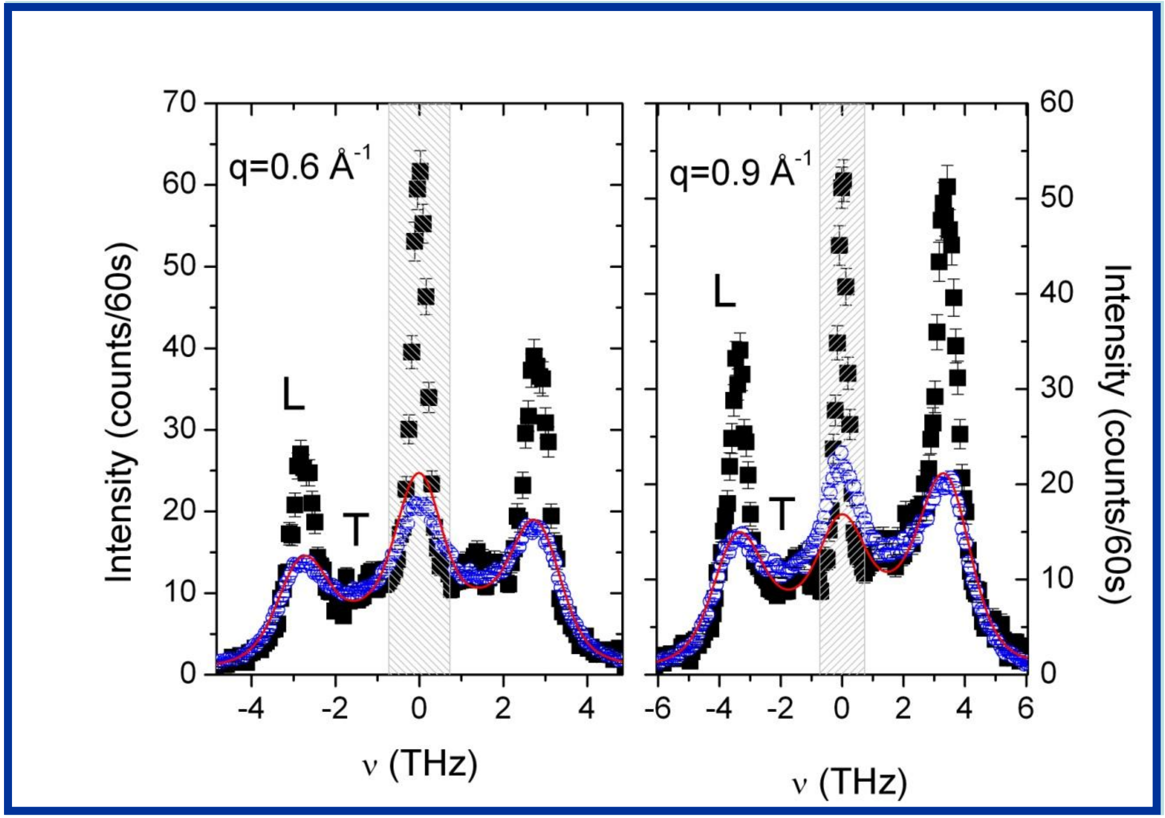


Es. sodium: the short range order of the liquid evolves with P and T following similar transformations as in the solid phase

Raty et al., Nature 449, 448 (07)

The dynamic structure factor of sodium

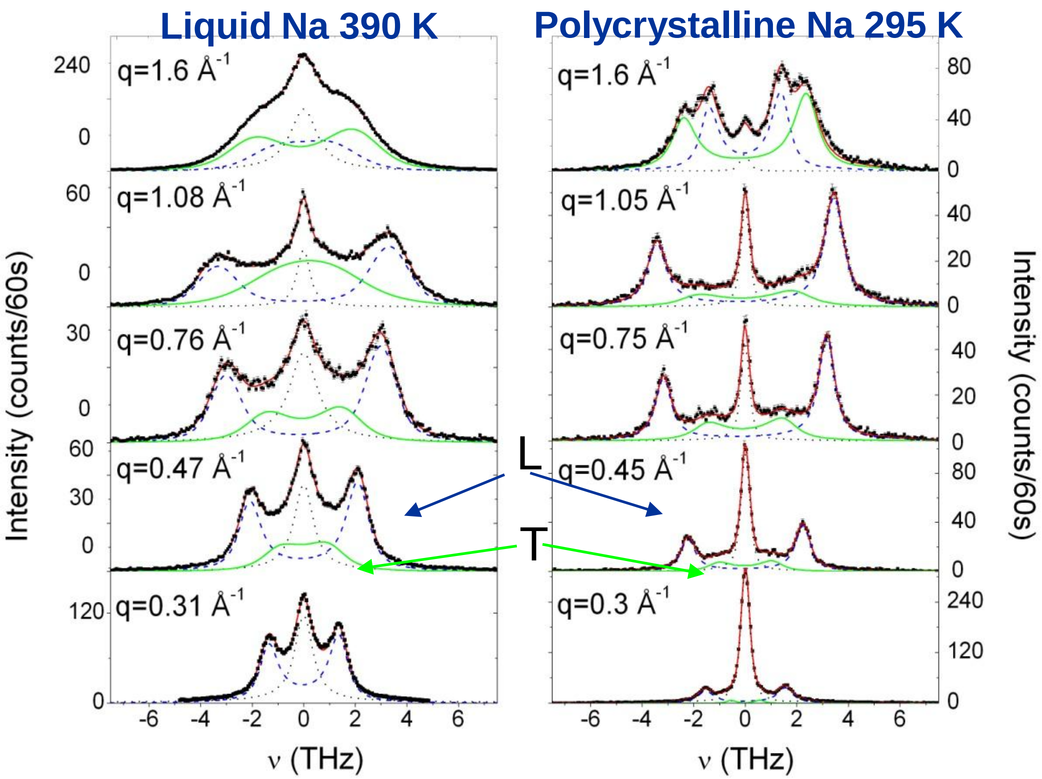
Inelastic x-ray scattering data @ ID16, E=23.724 keV, ΔE=1.4 meV



The spectrum of the liquid is a broadened version of that of the (poly)crystal

Giordano & g.m., PNAS 107, 21985 (10)

A simple crystal-like model

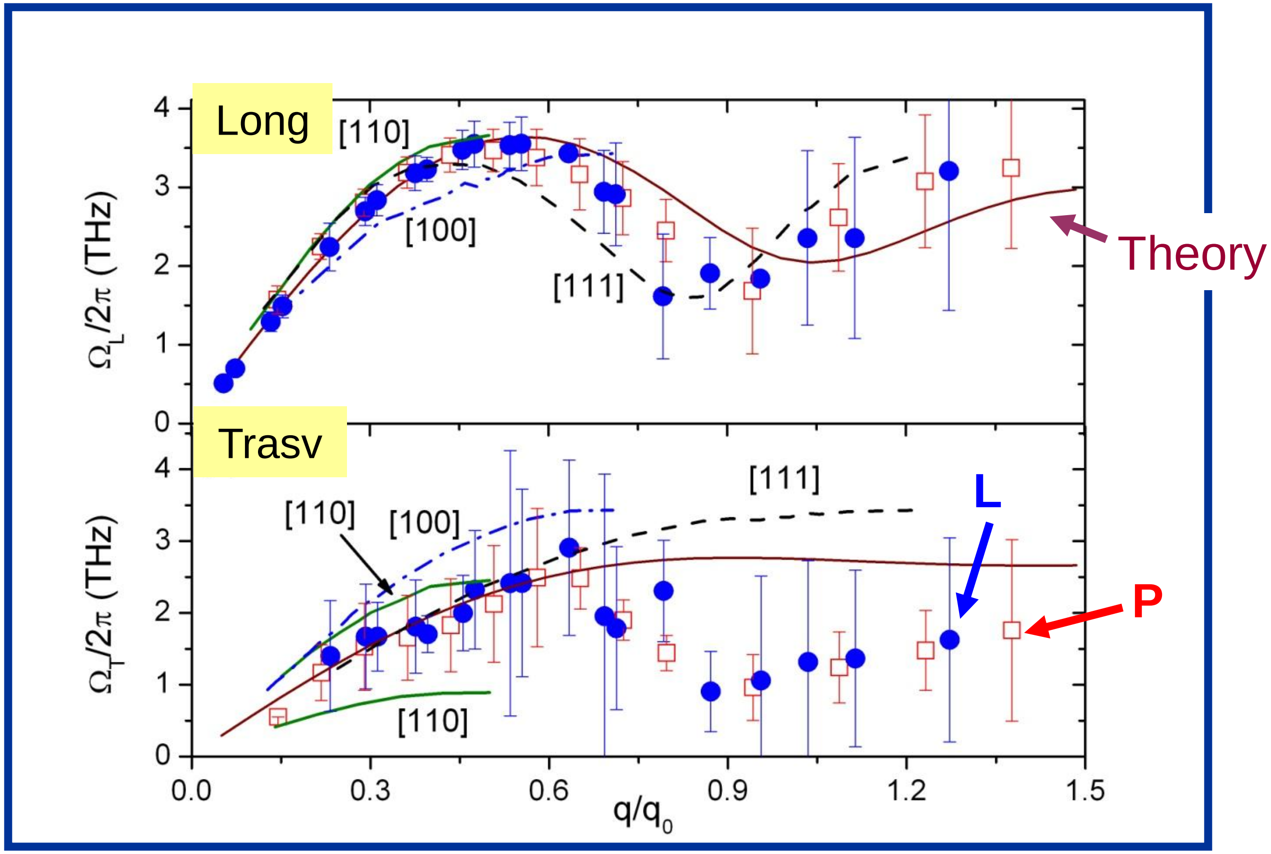


model:

$$S(q,\omega) = S_{el}\frac{\Gamma_0}{\omega^2+\Gamma_0^2} + S_{in}(q,\omega)\otimes G(\omega)$$

$$S_{in}(q,\omega) = I_L\frac{\Omega_L^2\Gamma_L}{(\omega^2-\Omega_L^2)^2+\omega^2\Gamma_L^2} + I_T\frac{\Omega_T^2\Gamma_T}{(\omega^2-\Omega_T^2)^2+\omega^2\Gamma_T^2}$$

The order fingerprint



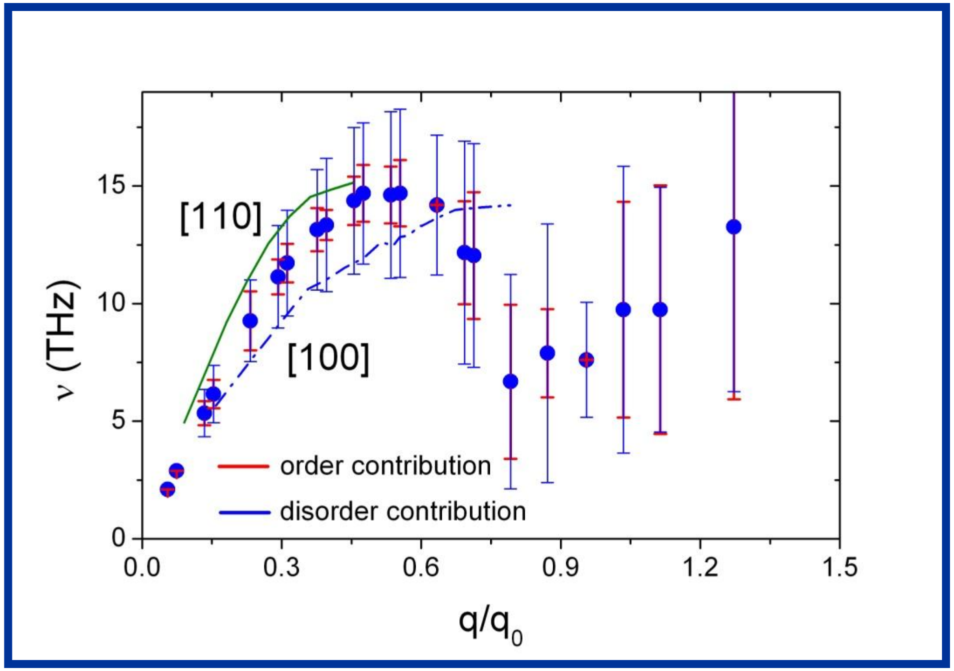
Giordano & g.m., PNAS 107, 21985 (10)

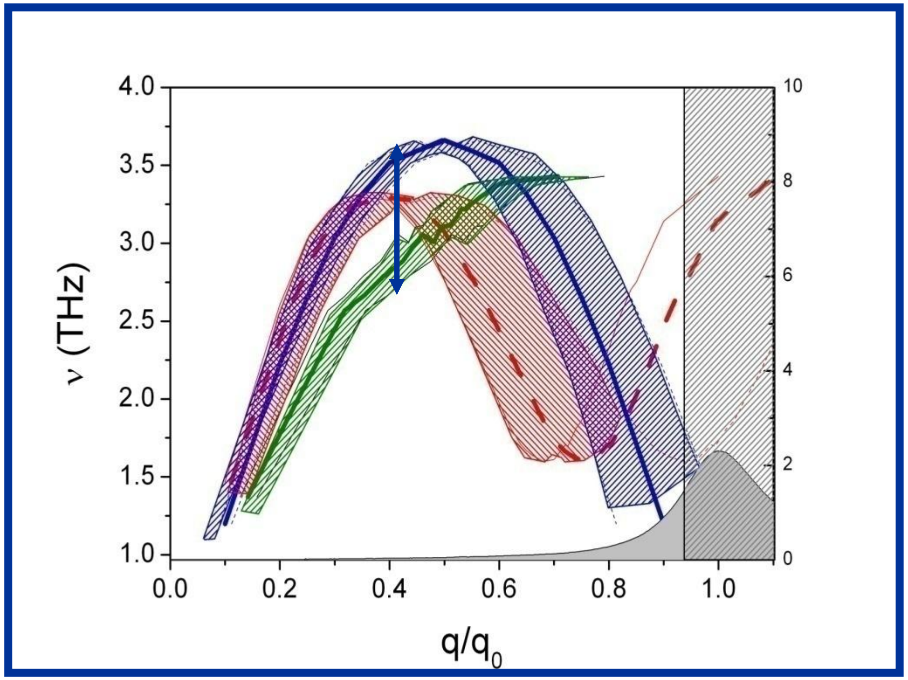
First Brillouin zone:

The longitudinal and transverse dispersion curves in the liquid reflect those for the polycrystal: an orientational average over the high symmetry branches of the single crystal

(density scaling of frequencies for the solid)

The disorder fingerprint





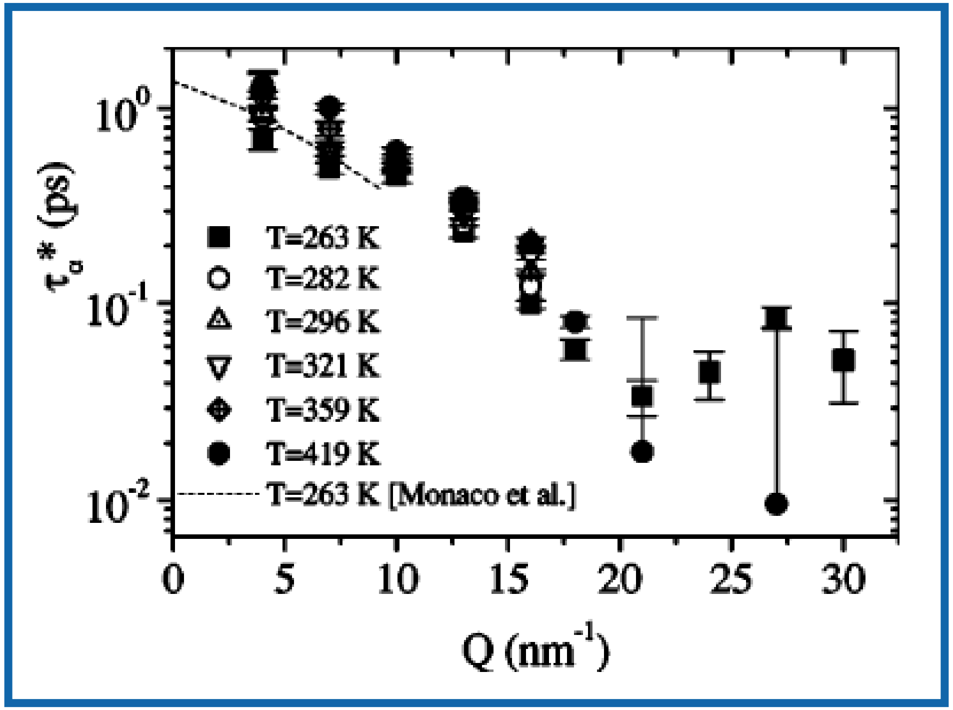
- ➢ **Local order → dispersion**
- ➢ **Average disorder → broadening**

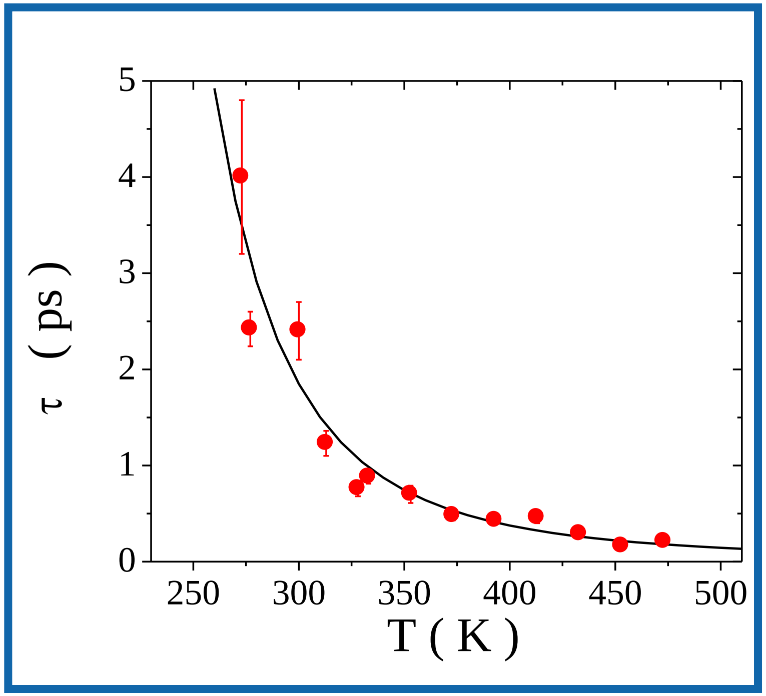
Giordano & g.m., PNAS 107, 21985 (10)

Liquids: reaching the macroscopic limit

At high q (> 1 nm^{-1}) the dynamics is q-dependent

At low q (< 1 nm^{-1}) macroscopic hydrodynamics is recovered

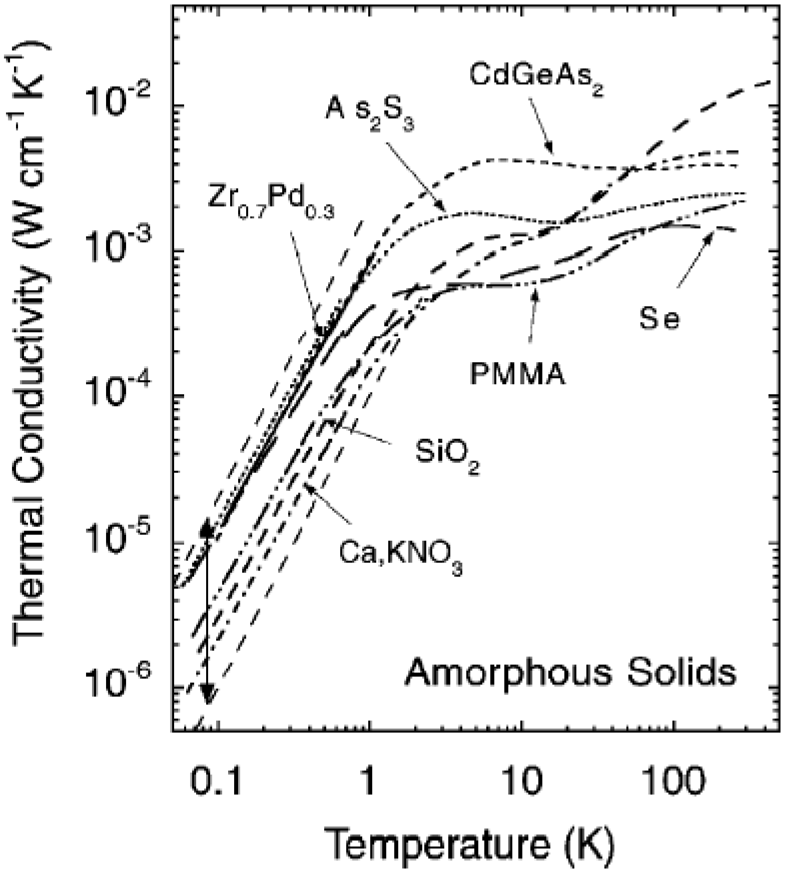




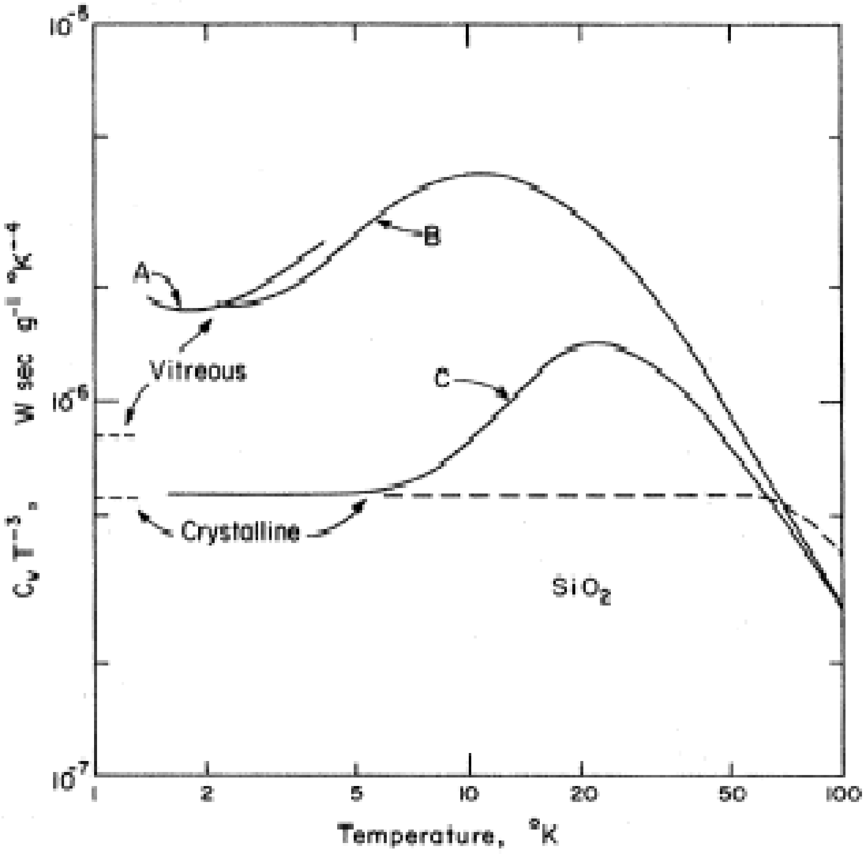
Pontecorvo et al., PRE 71, 011501 (2005)

g.m. et al., PRE 60, 5505 (1999)

The universal anomalies of glasses



Pohl et al., Rev. Mod. Phys. 74, 991 (02)

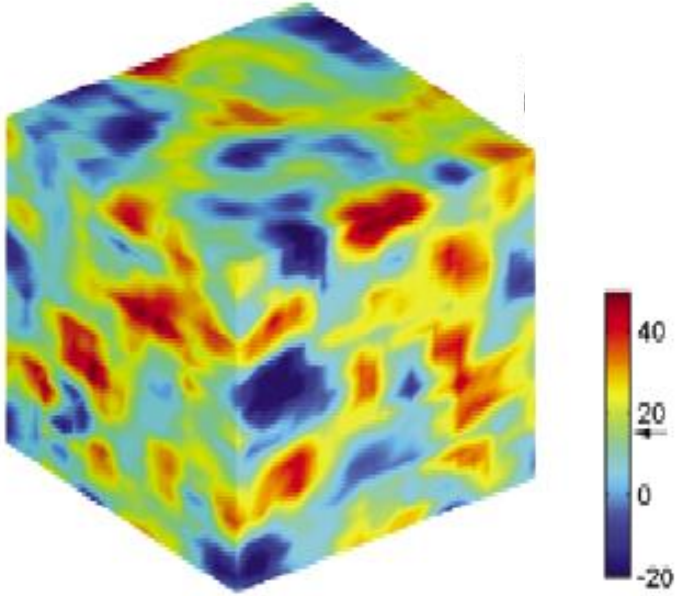


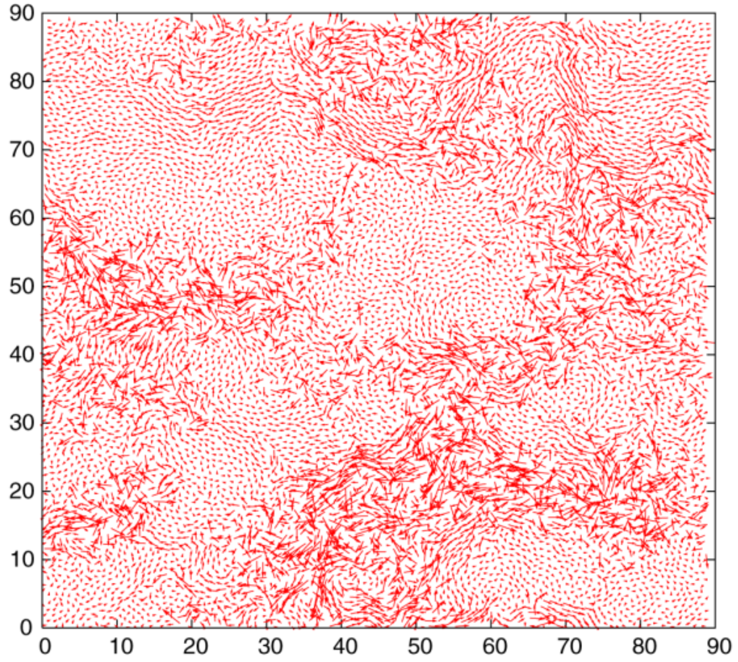
Zeller and Pohl, PRB 4, 2029 (71)

Elastic heterogeneities - I

Spatial heterogeneities of the local shear modulus of a model polymer glass

Non affine displacement field (silica)





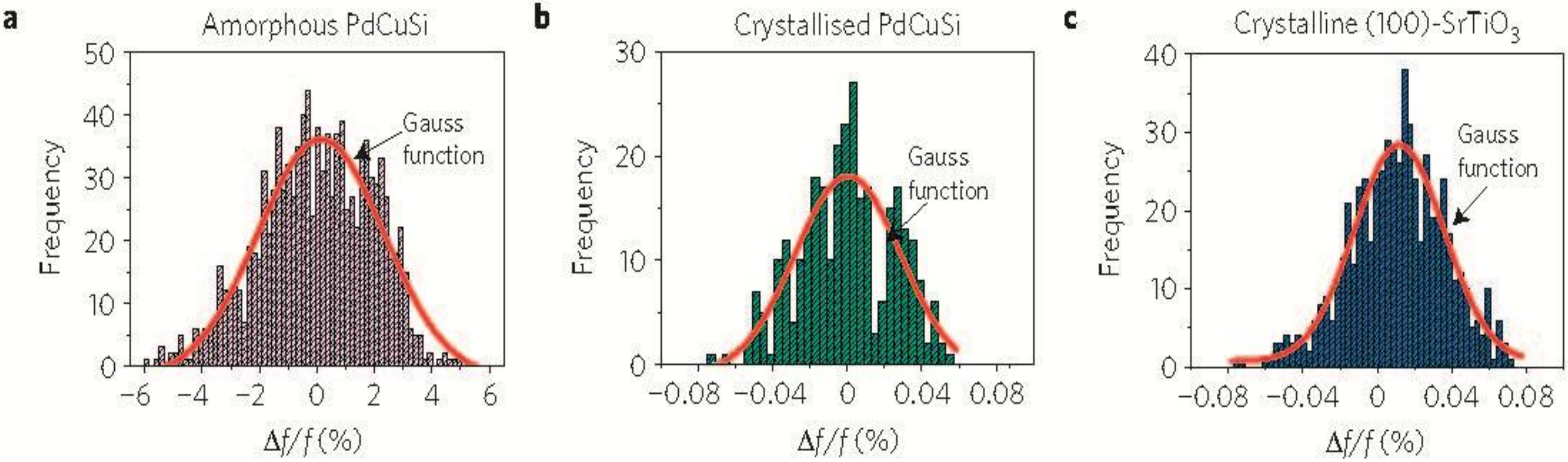
Yoshimoto et al., PRL 93, 175501 (04)

Leonforte et al., PRL 97, 055501 (06)

Elastic heterogeneities - II

Very limited experimental information on the local elasticity of glasses

Atomic force acoustic microscopy:

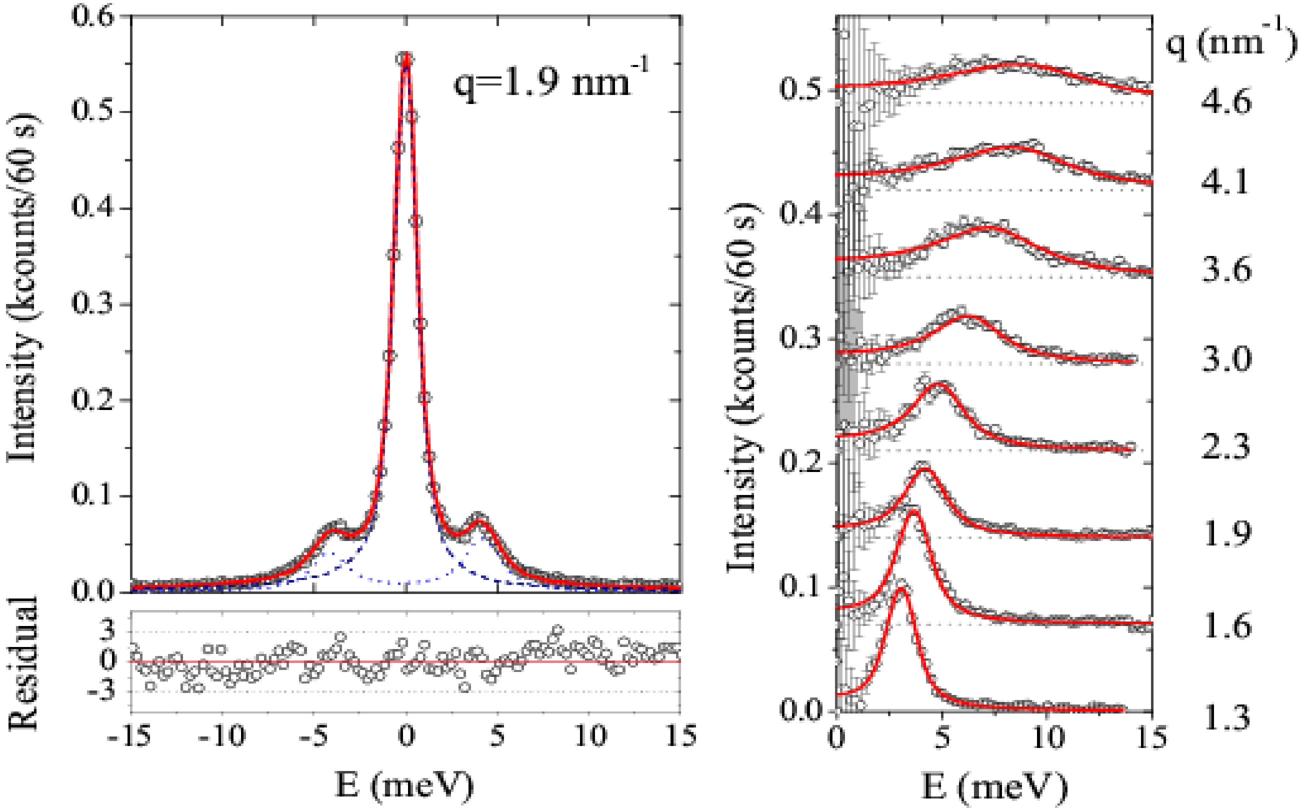


f: contact resonance frequency ⟶ local indentation modulus

Wagner et al., Nat. Mat. 10, 439 (11)

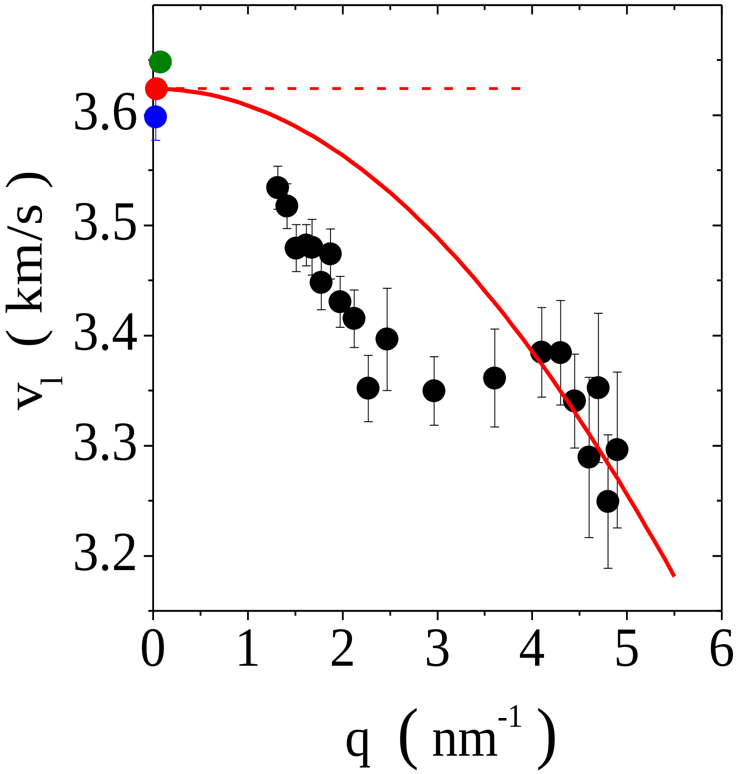
The dynamic structure factor of glycerol

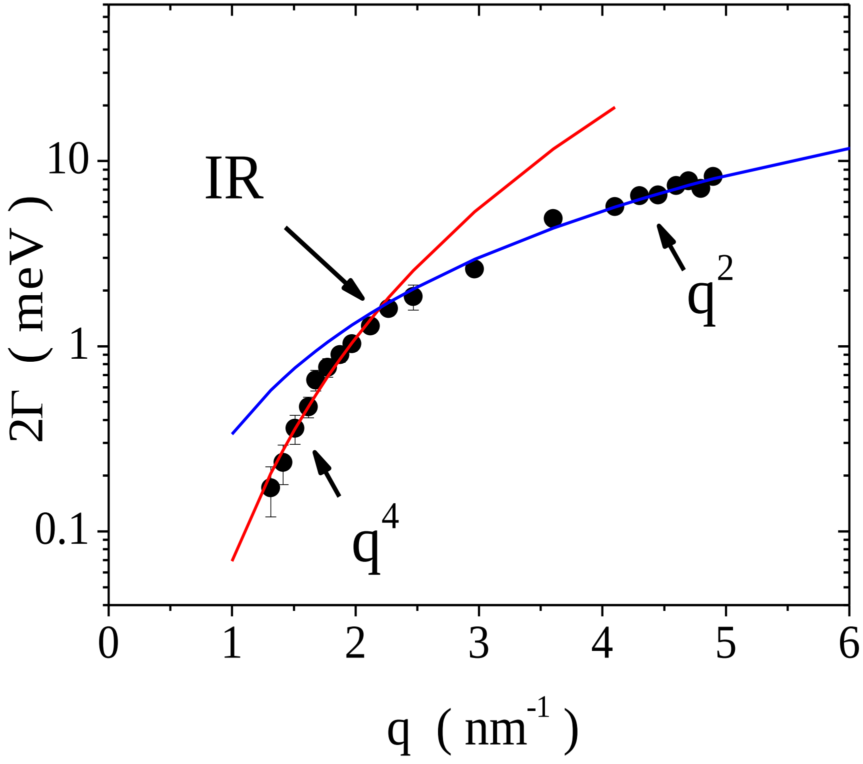
Inelastic x-ray scattering data @ ID16, E=23.724 keV, ΔE=1.4 meV



g.m. & Giordano, PNAS 106, 3659 (09)

Breakdown of the continuum approximation in glasses

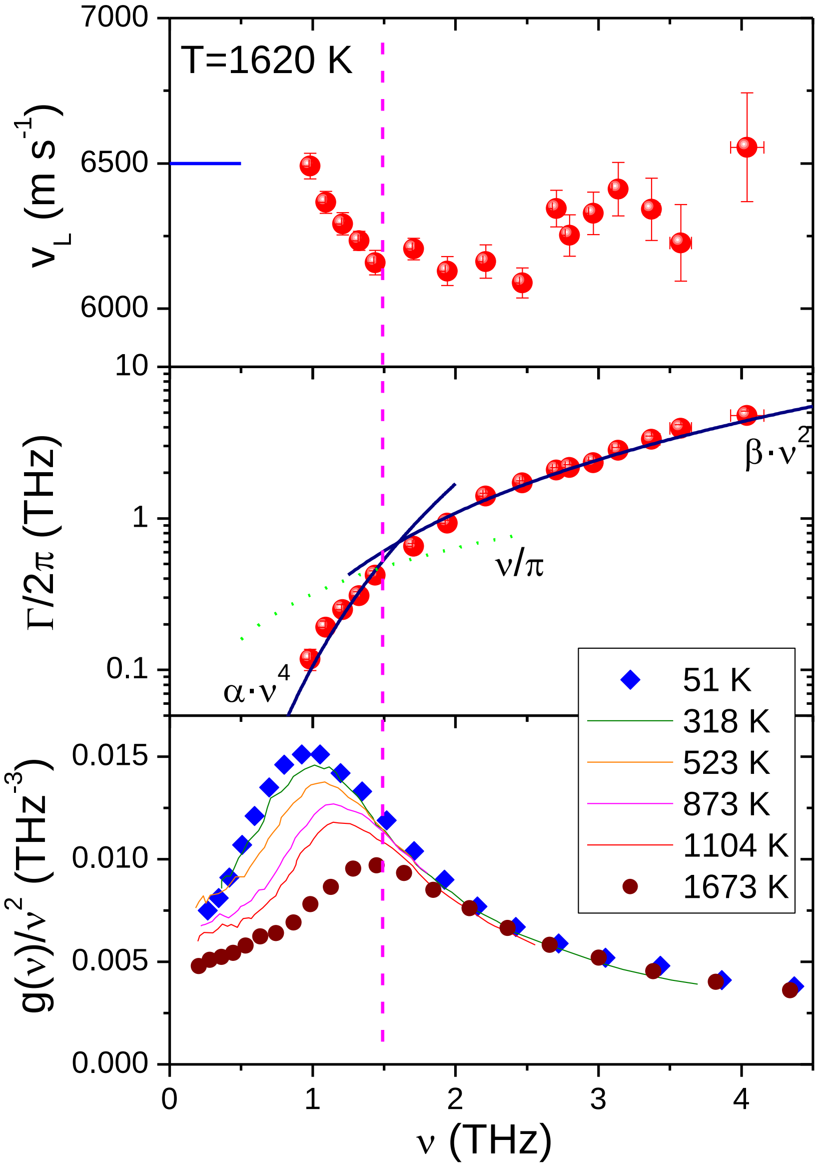




Breakdown of the Debye approximation & appearence of a Reyleigh scattering regime on the mesoscopic lengthscale of glasses

g.m. & Giordano, PNAS 106, 3659 (09)

The case of SiO_2 - I



Below the BP frequency:

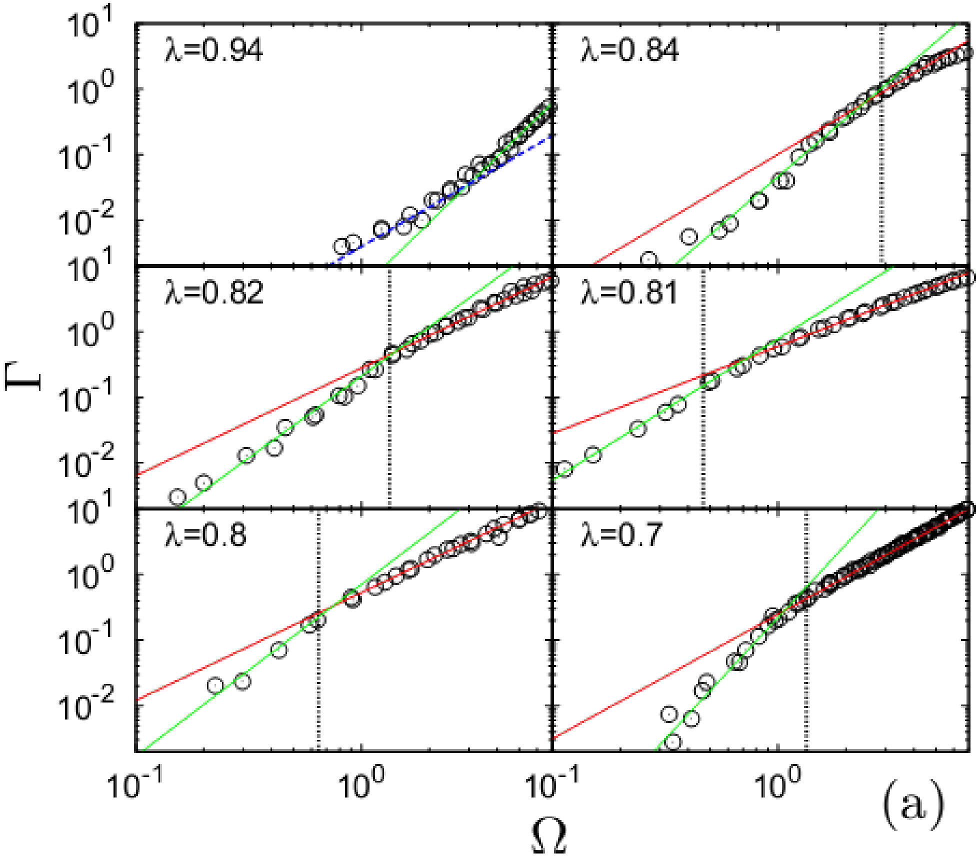
Negative dispersion of the sound velocity

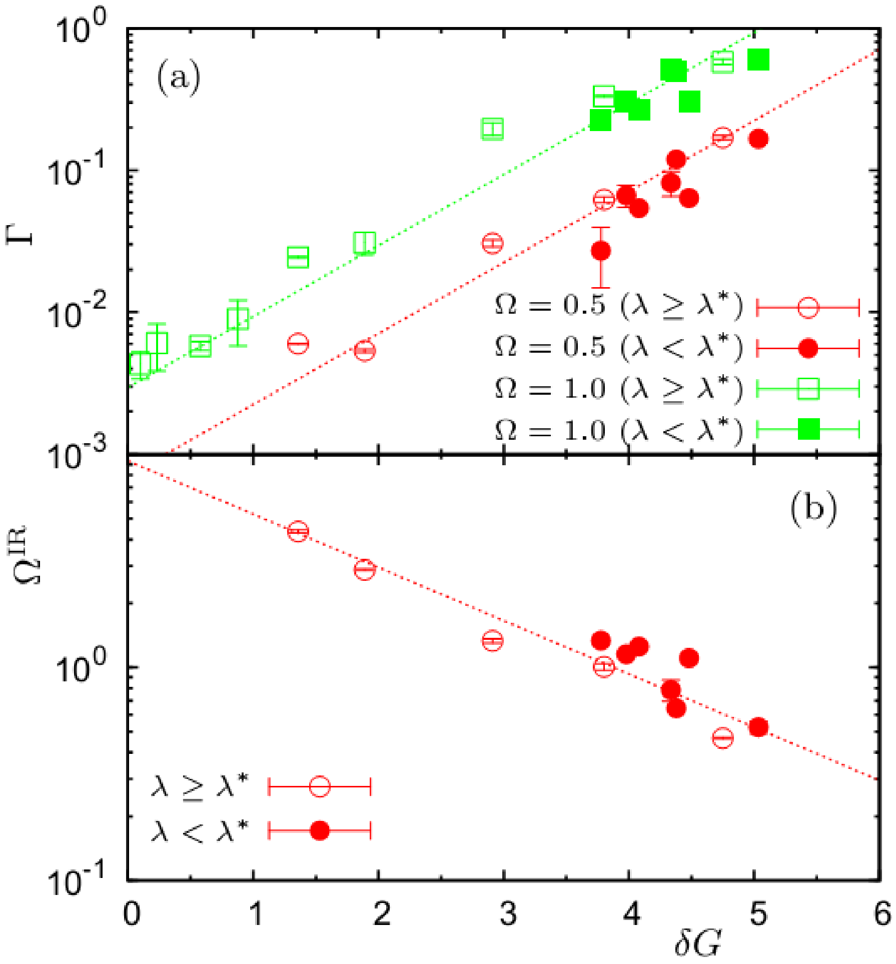
Damping ∝ ν^4

$\nu_c \sim \nu_{BP} \sim \nu_{IR}$

Baldi et al., PRL 104, 195501 (10)

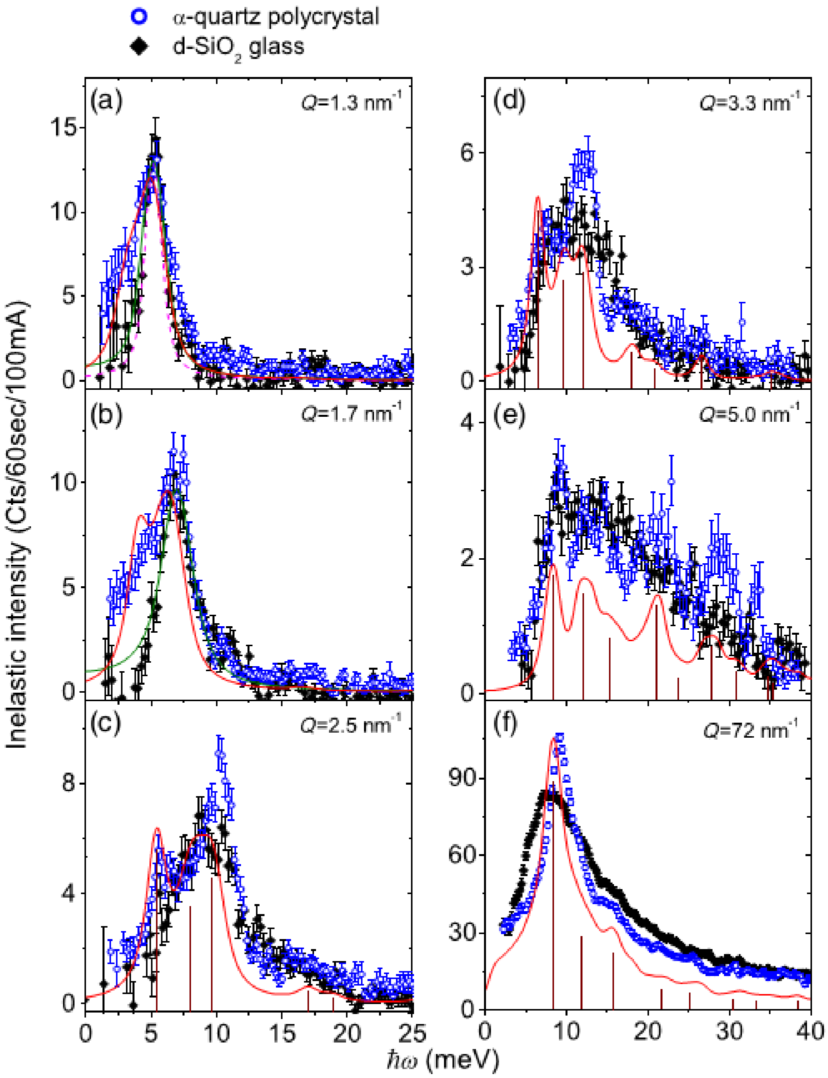
Elastic heterogeneities - III

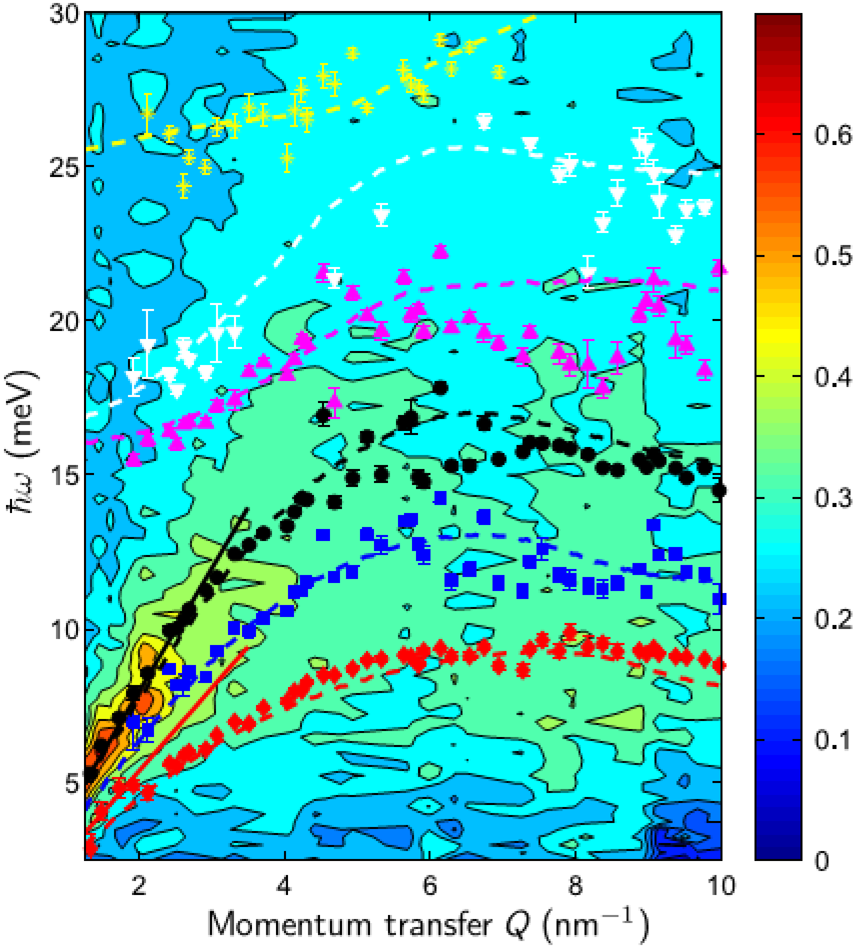




Mizuno et al., PNAS 111, 11949 (2014)

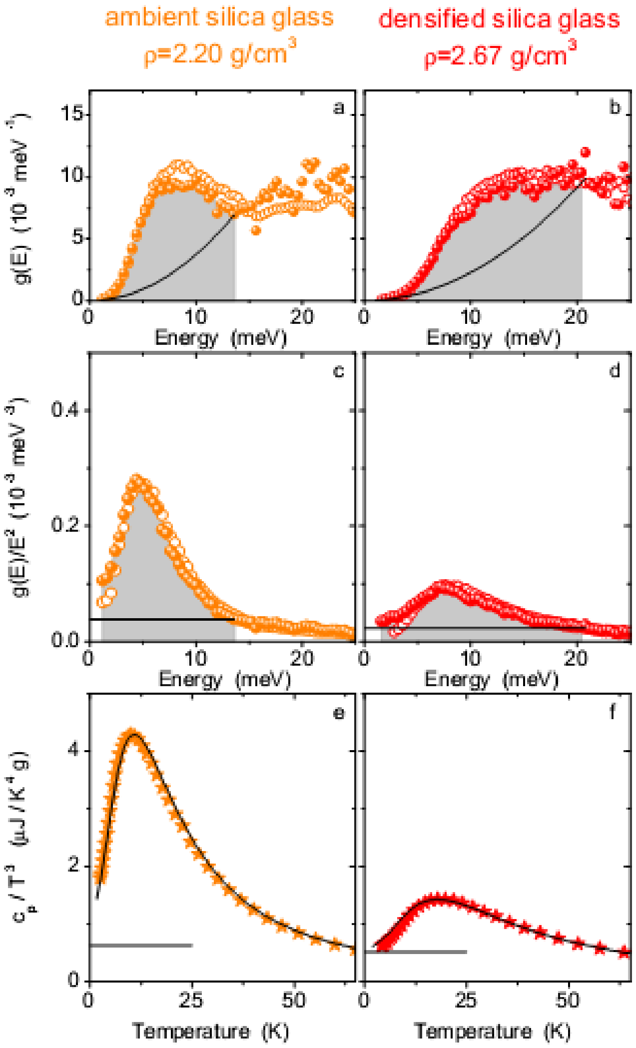
The case of SiO_2 - II

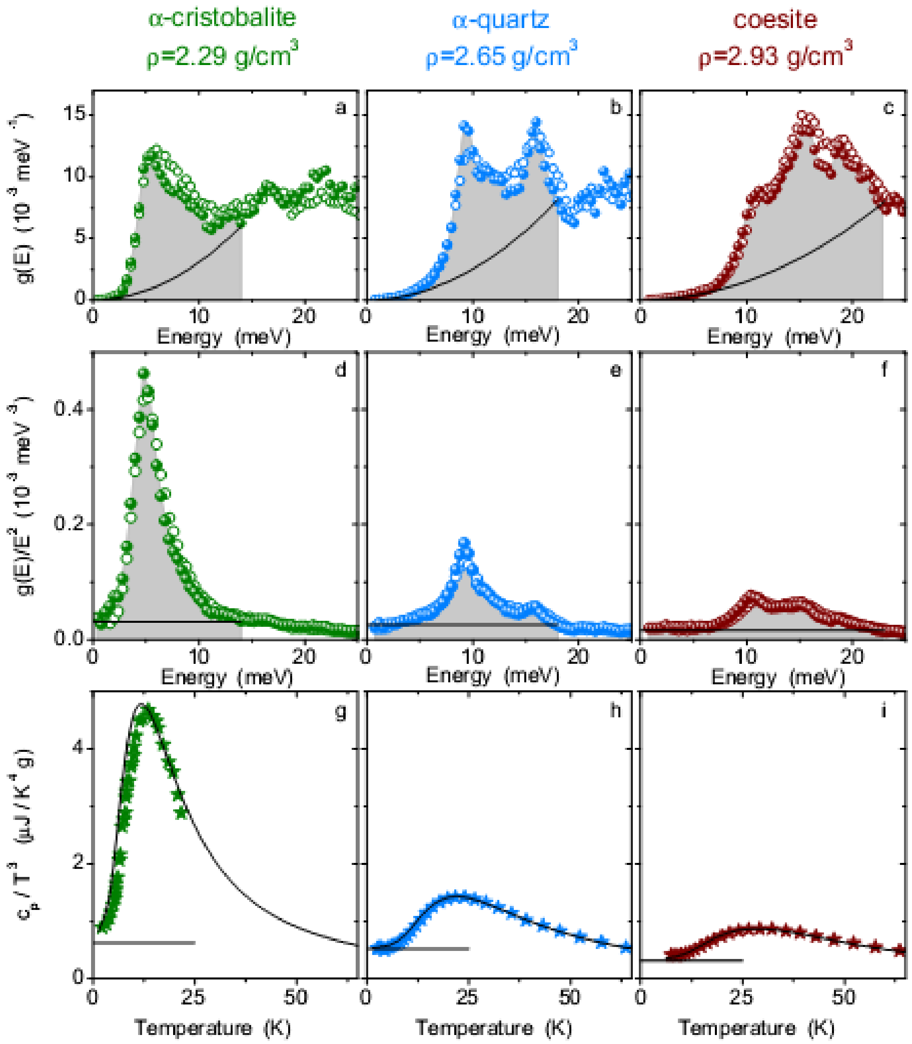




Baldi et al., PRL 110, 185503 (13)

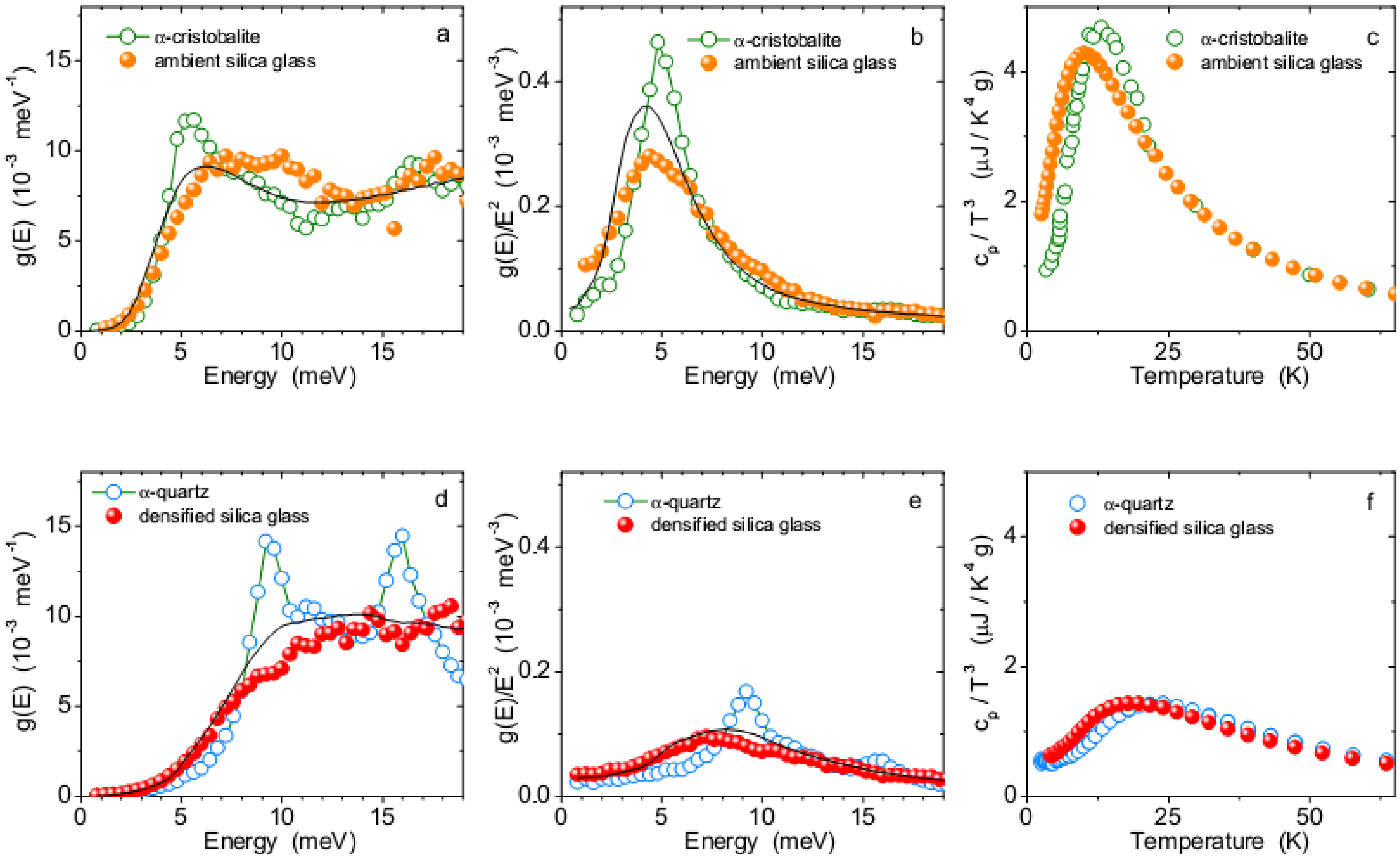
The case of SiO_2 - III





Chumakov et al., 112, 025502 (2014)

The case of SiO_2 - IV



Chumakov et al., 112, 025502 (2014)

Conclusions

1. The X-ray echo scheme promises to offer 10^2-10^3 times higher signal than at current IXS beamlines. Experiments not feasible at storage rings will be feasible, and those already feasible will be much faster…

2. In liquids it will be possible to study the q-dependence of the relaxation dynamics up to a fraction of a ns. This includes, in particular, the structural relaxation time in the critical temperature region for the dynamical arrest.

3. In both glasses and liquids, the mesoscopic range will be accessible with a single technique! This can be crucial for metastable systems…

Thank you !