Inhomogeneous elastic response of amorphous solids

Jean-Louis Barrat
Université de Lyon
Institut Universitaire de France

Acknowledgements: Anne Tanguy, Fabien Chay
Goldenberg, Léonforte, Michel Tsamados

Outline

- Global elastic constants
- Non affine deformations
- Local elastic constants
- Response to a point force
- Vibrational modes
- Density of states
- Jammed systems

Elastic constants for a system of particles interacting through a pair potential ϕ(r)

$$C_{\alpha\beta\gamma\delta} = \frac{\partial t_{\alpha\beta}}{\partial \epsilon_{\gamma\delta}} = 2nk_BT(\delta_{\alpha\gamma}\delta_{\beta\delta} + \delta_{\alpha\delta}\delta_{\beta\gamma})$$ **Kinetic term**

$$-\frac{V_0}{k_BT}\left[\langle \widehat{T}_{\alpha\beta}\widehat{T}_{\gamma\delta}\rangle - \langle \widehat{T}_{\alpha\beta}\rangle\langle \widehat{T}_{\gamma\delta}\rangle\right]$$ **Fluctuation term**

$$+C^{Born}_{\alpha\beta\gamma\delta}$$ **Born term**

$\hat{T}_{\alpha\beta}$ microscopic stress tensor (Irving-Kirkwood)

$$C^{Born}_{\alpha\beta\gamma\delta} = \frac{1}{V_0}\langle \sum_{ij} R_{ij,\alpha}R_{ij,\beta}R_{ij,\gamma}R_{ij,\delta}\left(\frac{\phi''(R_{ij})}{R_{ij}^2} - \frac{\phi'(R_{ij})}{R_{ij}^3}\right)\rangle$$

Born term corresponds to the change in energy <u>under a purely affine deformation</u> (see e.g. Aschcroft and Mermin. Solid state physics)

Fluctuation term ?

Derivation: take second derivative with respect to strain of the free energy in a deformed configuration (Squire, Holt, Hoover, Physica 1968)

$$\mathbf{R} = \underline{h}\mathbf{X}$$

$$\exp(-\beta F) = V^N \int d\mathbf{X}_1 d\mathbf{X}_2 .. d\mathbf{X}_N \exp(-\beta H(\{\underline{h}\mathbf{X}_i\}))$$

$$\underline{\epsilon} = \frac{1}{2}\left[(\underline{h}^T{}_0)^{-1}\underline{h}^T\underline{h}(\underline{h}_0)^{-1} - \underline{1}\right]$$

J.F. Lutsko, 1989 Generalized expressions for the calculation of elastic-constants by computer-simulation , *J. Appl. Phys.* **65**, 2991-2997

J-L. Barrat,J-N. Roux, J-P. Hansen, M.L. Klein, 1988, Elastic response of a simple amorphous binary alloy near the glass-transition *Europhysics Letters*, **7**, 707-713

for application to glasses

Microscopic elasticity of complex systems, J-L. Barrat
http://arxiv.org/abs/cond-mat/0601653

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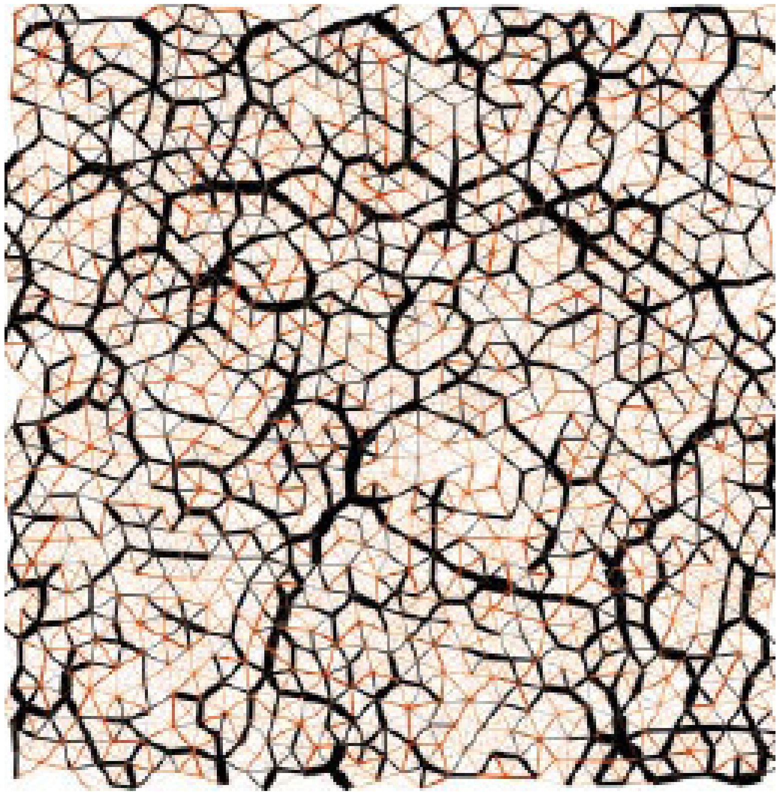
To understand the fluctuation term take a system at zero temperature, deform it and compute stress tensor (here 2d LJ system)

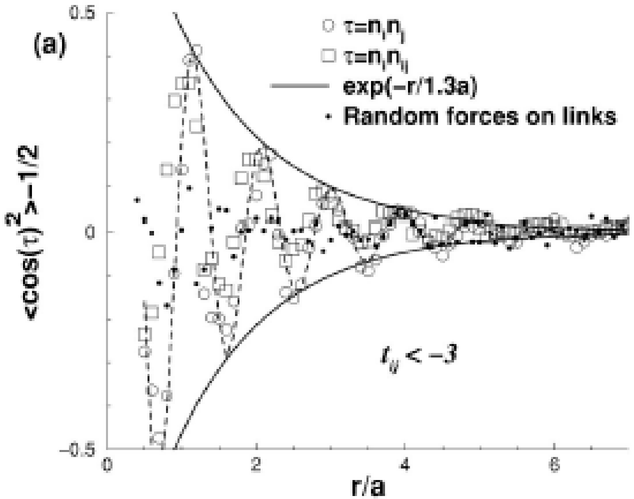


Equilibrium configuration at zero temperature – zero pressure

Vertices: particles

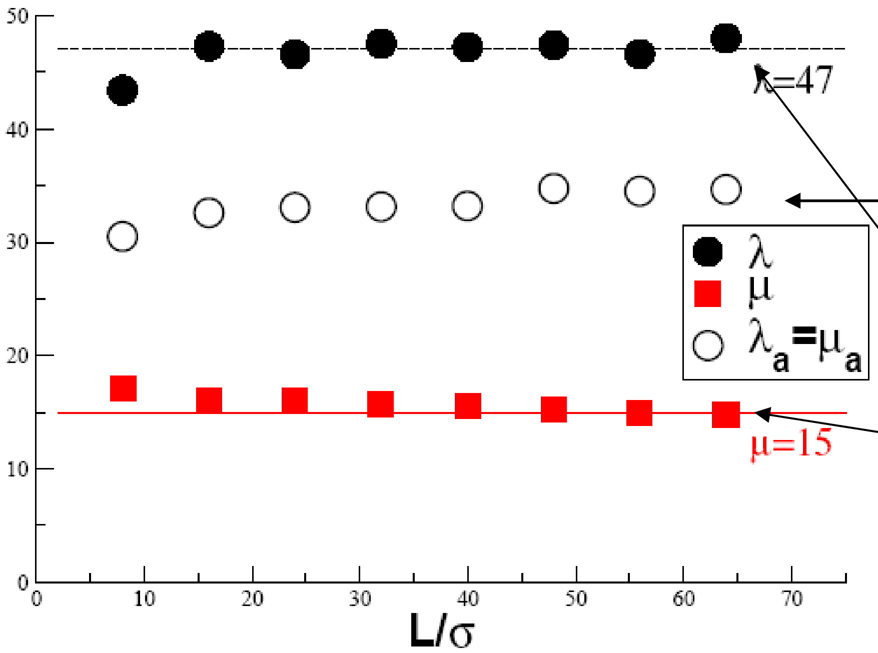
Black lines: repulsive force

Red lines: attractive forces



Force chains ? Nothing obvious in correlation functions

Elastic constants of a model (Lennard-Jones polydisperse mixture) amorphous system at low temperature, vs system size



Born term only (<u>affine deformation at all scales</u>)

Actual result with non affine deformation (<u>fluctuations or relaxation term</u>)

λ, μ Lamé coeffients

$\mu = G$ shear modulus

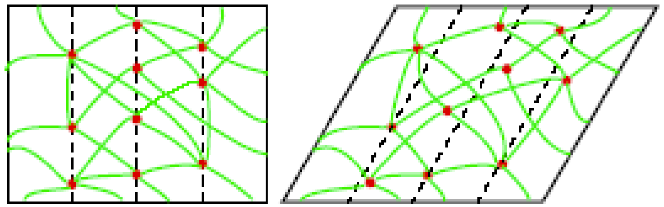
$\lambda + \mu = B$ bulk modulus

Born term innacurate $\Longrightarrow$

-Large fluctuation term (finite T)

-Large fraction of the elastic energy stored in a <u>nonaffine deformation field</u> (zero T)

Deform the sample (rescale all coordinates $X \to X(1+\epsilon)$)
Minimize energy $X(1+\epsilon) \to X'$
Substract affine deformation
$\to$ non affine deformation field $\delta X = X' - X(1+\epsilon)$



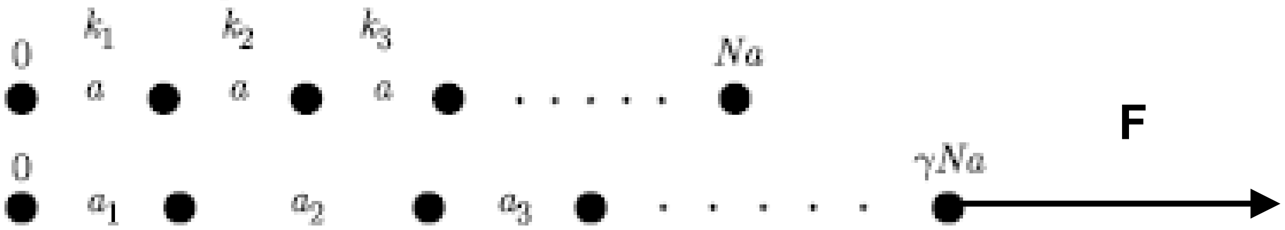
(a) (b)

Contribution from nonaffine field (relaxation) at zero temperature is equivalent to fluctuation term in elastic constants

Lutsko 1994: take derivatives w.r.t. strain at mechanical equilibrium (zero force on all particles)

Nonaffine deformation is due to elastic inhomogeneity:

Example in one dimension



$$\delta_p = a_p - a = F/k_p$$

$$u_p = F \times \sum_{i=1}^{p} k_i^{-1}$$

$$u_p^{NA} = F \times \sum_{i=1}^{p} (k_i^{-1} - < k^{-1} >)$$

$$< (u_p^{NA})^2 > \sim p < (\delta k^{-1})^2 >$$

DiDonna and Lubensky, PRE 2006

Non affine correlations : Response to randomness in local elastic constants

$$\boxed{\begin{aligned} u &= u_a + u_{na} \\ C &= C_0 + \delta C \end{aligned}}$$

$$\nabla \underline{\mathrm{C}} \nabla u = 0$$

$$C_0 \Delta u_{na} = \nabla(\delta C) \nabla u_a$$

$$G(x) = < u_{na}(x) \cdot u_{na}(0) >$$

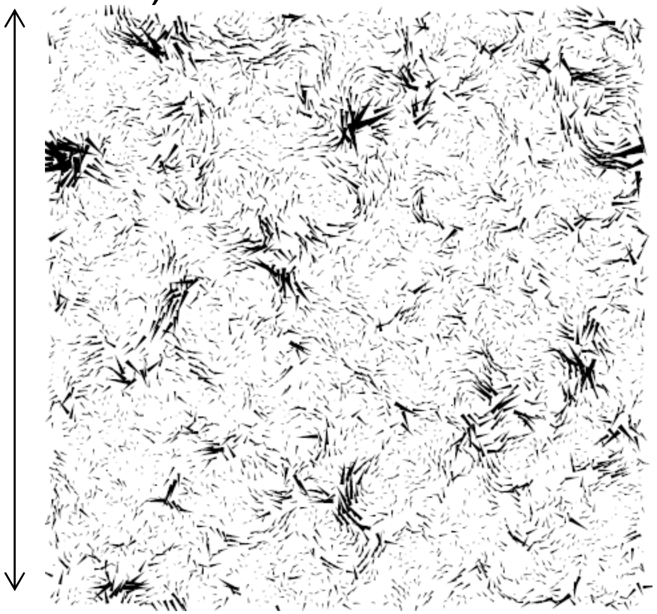
$$G(q) \sim (\delta C)^2 / q^2$$

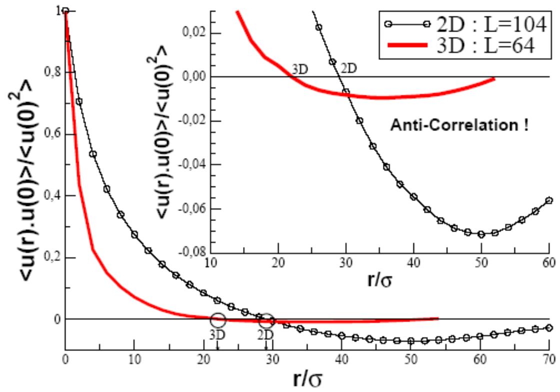
$$G(x) \sim A \ln(x/L) \text{ in 2d} \quad G(x) \sim B/x \text{ in 3d}$$

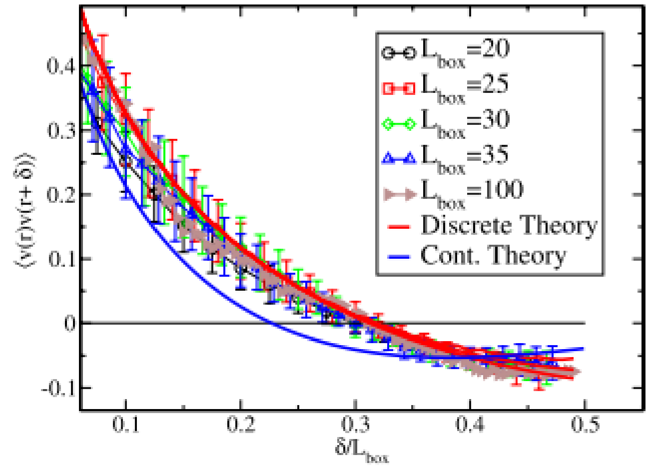
2d, 3d situation (from simulation)

Snapshot of nonaffine displacement (uniaxial extension)

100σ







Maloney, PRL 2006

Correlations of the non affine field reveal the existence of elastic heterogeneities

→ **Continuum, homogeneous elasticity not applicable at small scales**

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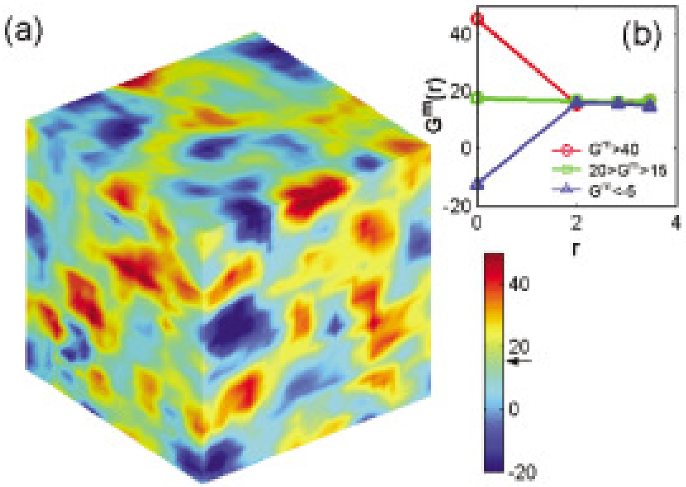
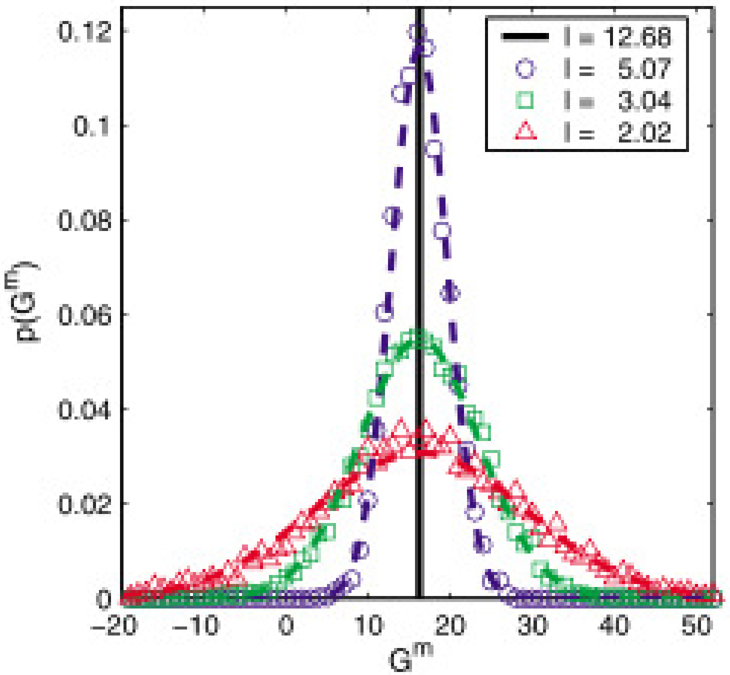
Several possible definitions of local elastic constants

- Use fluctuation formula within a cell of finite size (de Pablo)
- Impose affine deformation except in a small cell, local stress/strain relation (Sollich)
- Use coarse grained displacement and stress fields (Goldenberg and Goldhirsch, Tsamados *et al*)

Example of the first approach for a polymer glass (Yoshimoto, Jain, de Pablo, PRL 2004)





•Note the negative elastic constants (unstable)

Coarse grained displacement, stress and strain fields
(Goldhirsch Goldenberg)

$\phi(r)$ coarse graining function with width w
(gaussian, triangle)

$$\rho(\mathbf{r},t) = \sum_i m_i \phi(\mathbf{r} - \mathbf{r}_i(t)) \ ,$$

$$p_\alpha(\mathbf{r},t) = \sum_i m_i v_{i\alpha} \phi(\mathbf{r} - \mathbf{r}_i) \ ,$$

$$\mathbf{V}(\mathbf{r},t) = \mathbf{p}(\mathbf{r},t)/\rho(\mathbf{r},t) \ .$$

$$\sigma_{\alpha\beta}(\mathbf{r},t) = -\frac{1}{2}\sum_{i,j} f_{ij\alpha}(t) \mathrm{R}_{ij\beta}(t) \int_0^1 ds \phi(\mathbf{r} - \mathbf{r}_i + s\mathbf{r}_{ij}) \ ,$$

$$\mathbf{u}(\mathbf{R},t) = \frac{\sum_i m_i \mathbf{u}_i(t) \phi\left[\mathbf{r} - \mathbf{r}_i(t)\right]}{\sum_j m_j \phi\left[\mathbf{r} - \mathbf{r}_j(t)\right]}$$

$$\varepsilon^{lin}_{\alpha\beta}(\mathbf{r},t) = \frac{1}{2}\left[\frac{\partial u^{lin}_\alpha(\mathbf{r},t)}{\partial r_\beta} + \frac{\partial u^{lin}_\beta(\mathbf{r},t)}{\partial r_\alpha}\right] \ .$$

Procedure:

•Perform 3 independant deformations

•Compute stress and strain at each point in space

•Obtain 9 equations for the 6 unknowns of the elastic tensor

$$\begin{pmatrix} \sigma_{xx} \\ \sigma_{yy} \\ \sqrt{2}\sigma_{xy} \end{pmatrix} = \begin{pmatrix} \mathrm{C}_{xxxx} & \mathrm{C}_{xxyy} & \mathrm{C}_{xxxy} \\ \mathrm{C}_{xxyy} & \mathrm{C}_{yyyy} & \mathrm{C}_{yyxy} \\ \mathrm{C}_{xxxy} & \mathrm{C}_{yyxy} & \mathrm{C}_{xyxy} \end{pmatrix} \begin{pmatrix} \varepsilon_{xx} \\ \varepsilon_{yy} \\ \sqrt{2}\varepsilon_{xy} \end{pmatrix}$$

•Use 6 to get the elastic constants, 3 to check error

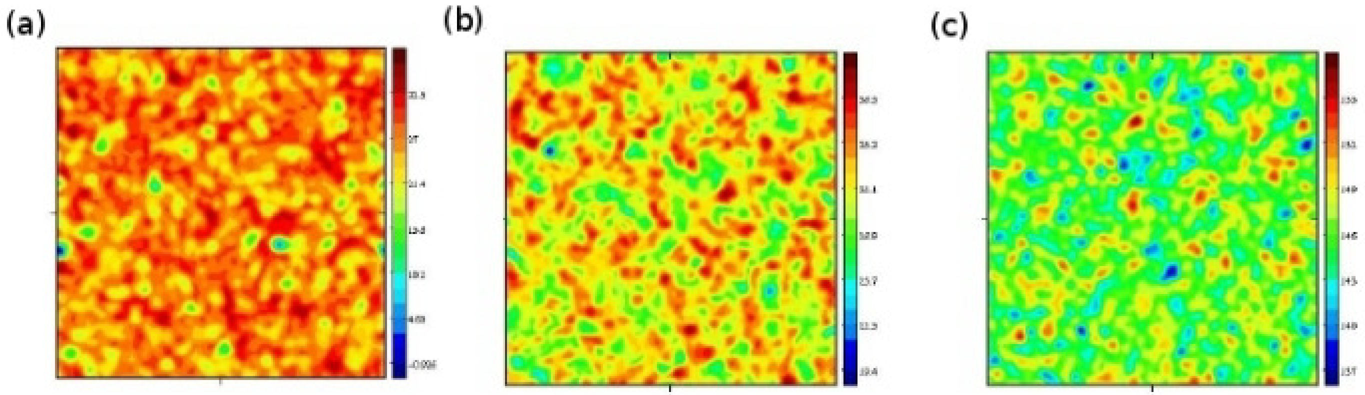
•Diagonalize to get three local elastic constants $C_1 < C_2 < C_3$

Note: for homogeneous isotropic media the stress strain relation is:

$$\begin{pmatrix} \sigma_{xx} \\ \sigma_{yy} \\ \sqrt{2}\sigma_{xy} \end{pmatrix} = \begin{pmatrix} \lambda+2\mu & \lambda & 0 \\ \lambda & \lambda+2\mu & 0 \\ 0 & 0 & 2\mu \end{pmatrix} \begin{pmatrix} \varepsilon_{xx} \\ \varepsilon_{yy} \\ \sqrt{2}\varepsilon_{xy} \end{pmatrix}$$

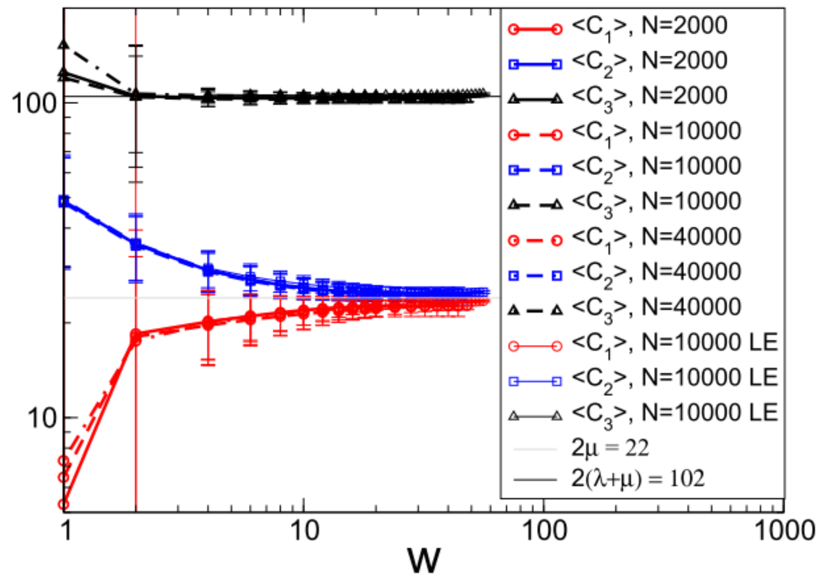
$$C_3 = \lambda + \mu > \mu = C_1 = C_2$$

Map of the three elastic constants for a coarse graining length w=10



Convergence to bulk limit with increasing w

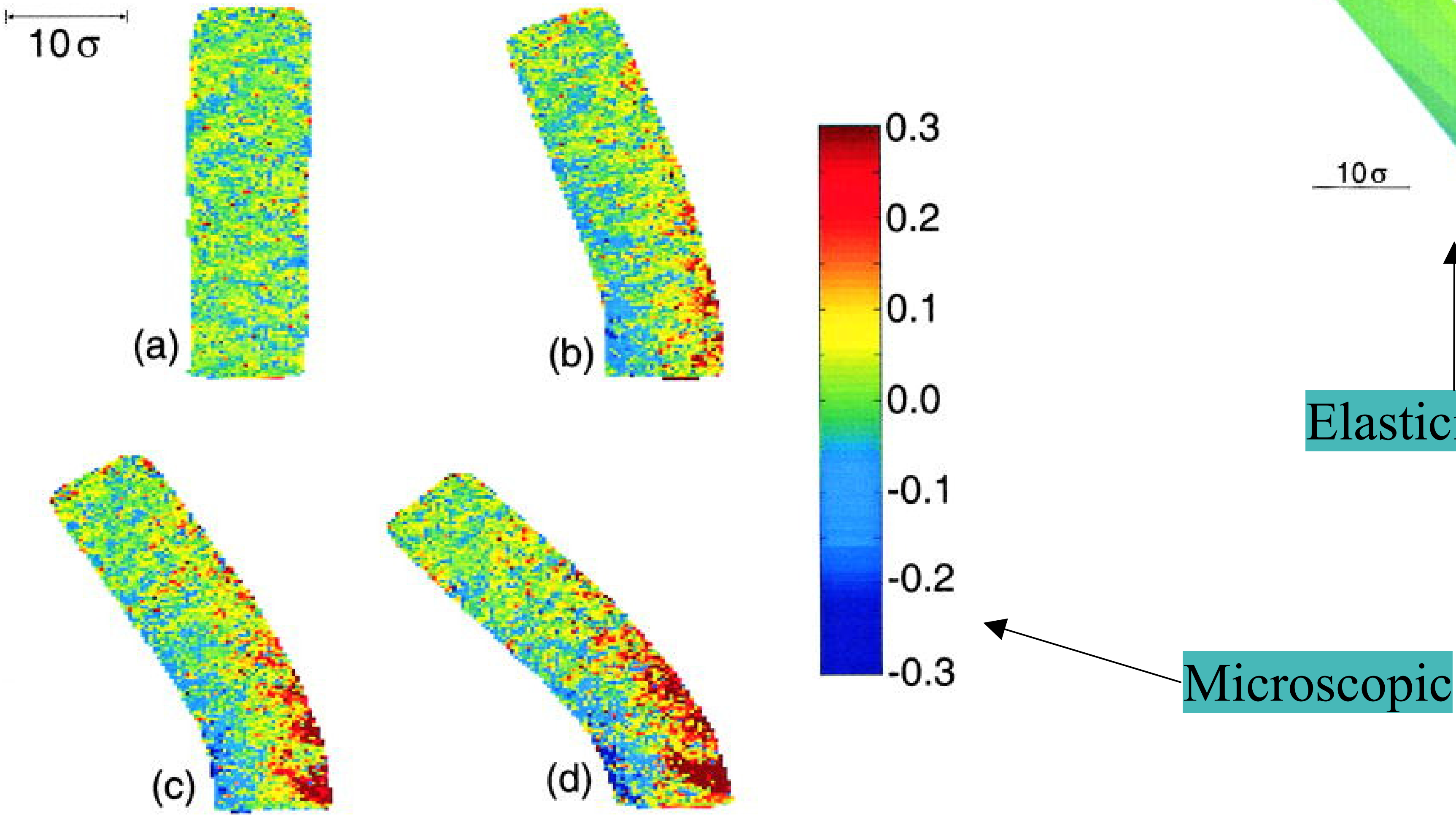
Power law convergence, no characteristic length

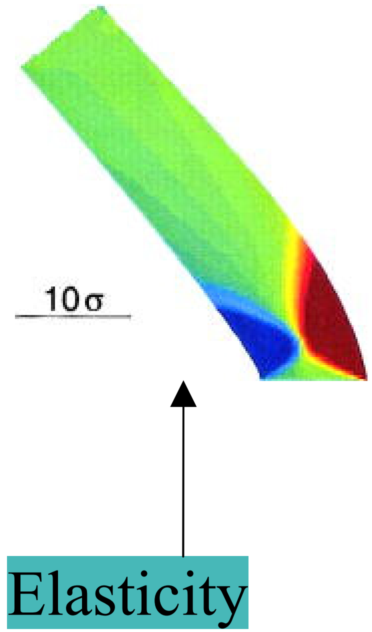


W	0	5	10	15	20
Hooke's law	NO	YES	YES	YES	YES
Homogeneity					
$\frac{\langle\bar{c}\rangle-2\mu}{2\mu}< 10\%$	NO	NO	YES	YES	YES
$\frac{\Delta c}{\langle\bar{c}\rangle}< 10\%$	NO	NO	NO	YES	YES
Isotropy					
$\frac{c_2-c_1}{2\mu}< 10\%$	NO	NO	NO	NO	YES

See Tsamados et al, PRE 2009

Other evidence of elastic inhomogeneity from simulation





Nanometric cantilever bending (amorphous polymer)
Juan de Pablo et al , JCP2001

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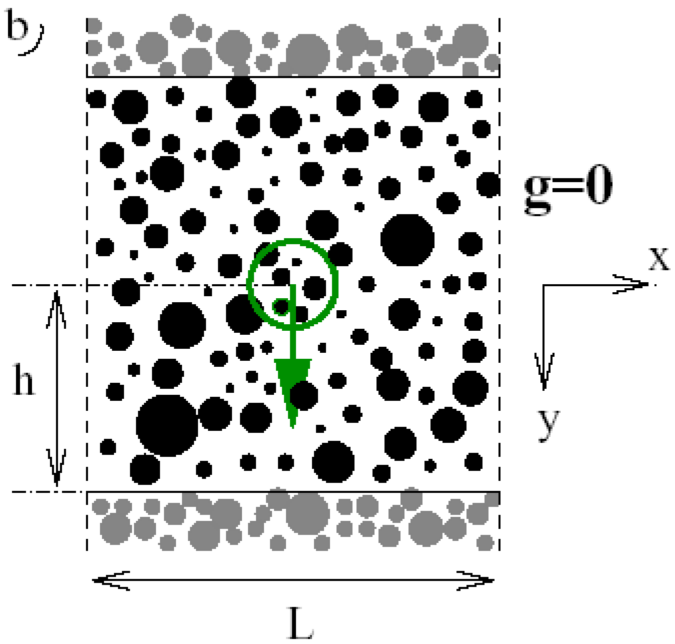
Elastic response to a point force:

- Granular systems
- Nanoindentation

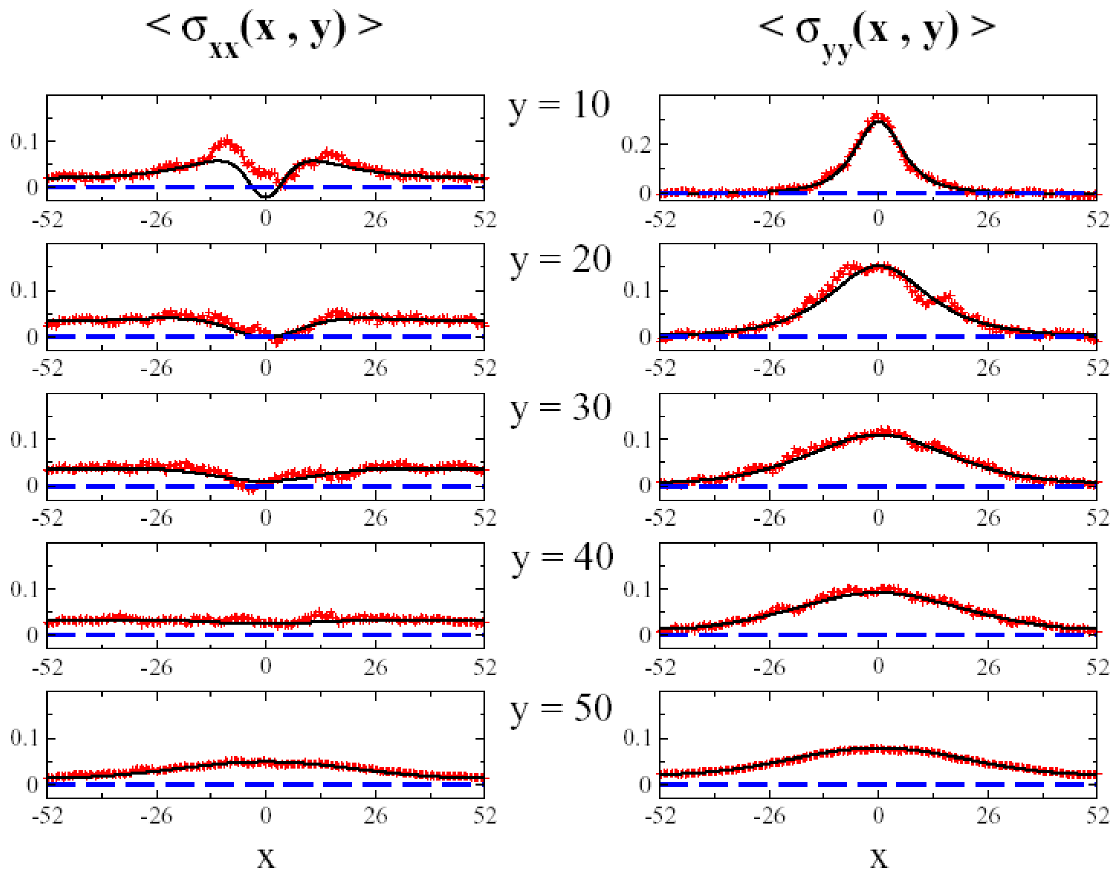


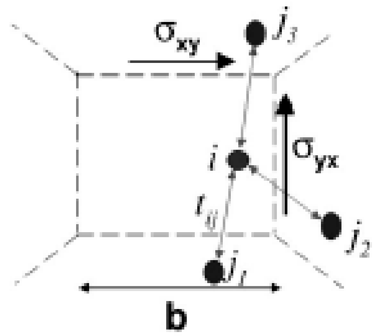
Small force applied to a few particles (source region, diameter 4a).

Fixed walls or compensation force on all particles.

Displacements, forces and incremental stresses computed in the elastic limit.

<u>Average response</u> obeys classical elasticity



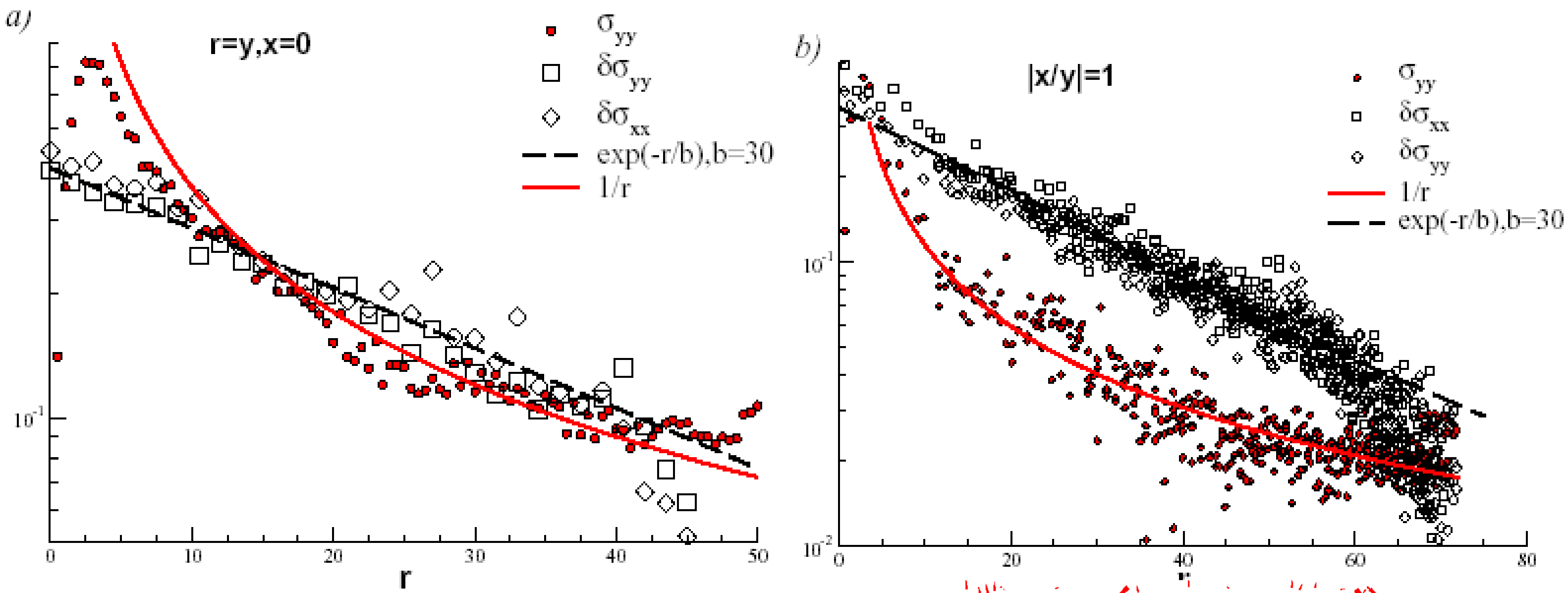


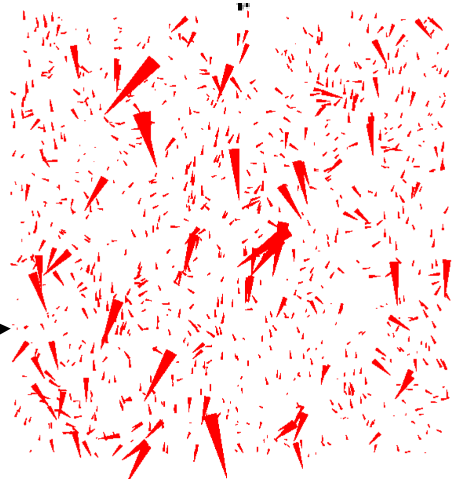
Local stress calculation for a cell of size b

simulation

Continuum prediction

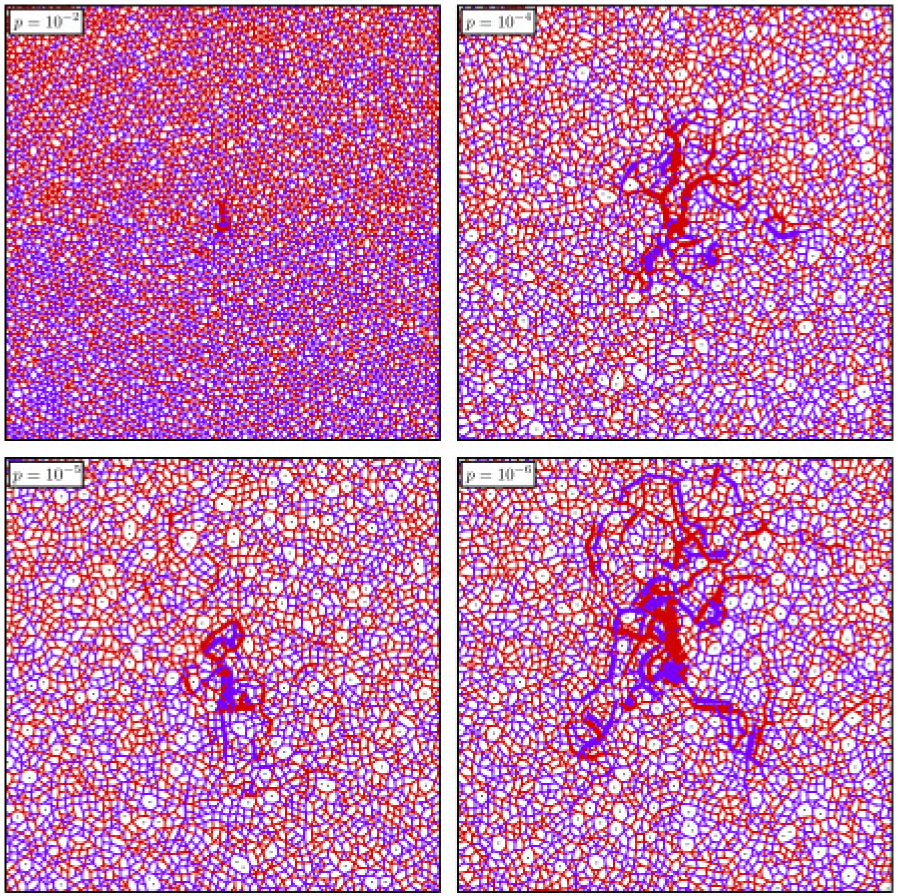
Large fluctuations between different realizations for distances smaller than 50 particle sizes





Source displacement

Note that in a granular system near jamming a corresponding analysis (Ellenbroek, van Hecke, van Saarloos, Phys Rev E 2009, Arxiv 0911.0944) a corresponding analysis reveals the isostatic length scale, l* that diverges at jamming (see below)



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Characteristic length associated with vibrations

- Exact diagonalisation of the dynamical matrix, low frequency spectrum).

OR

- Use continuum elasticity to obtain eigenfrequencies.

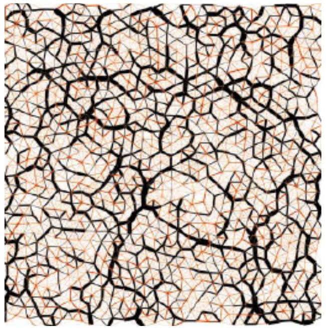
$\Rightarrow$ simple scaling of eigenfrequencies with size

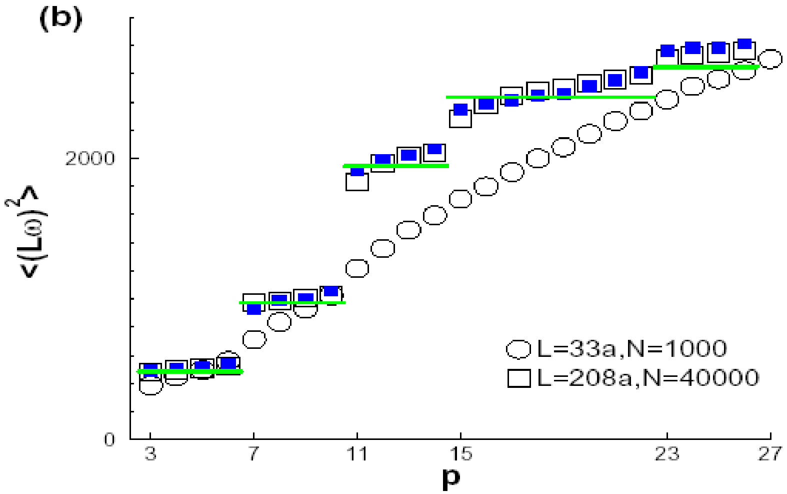
$$\omega_j = \alpha_j \frac{c}{R}$$

($c =$ sound velocity ; R=size; α_j=number)

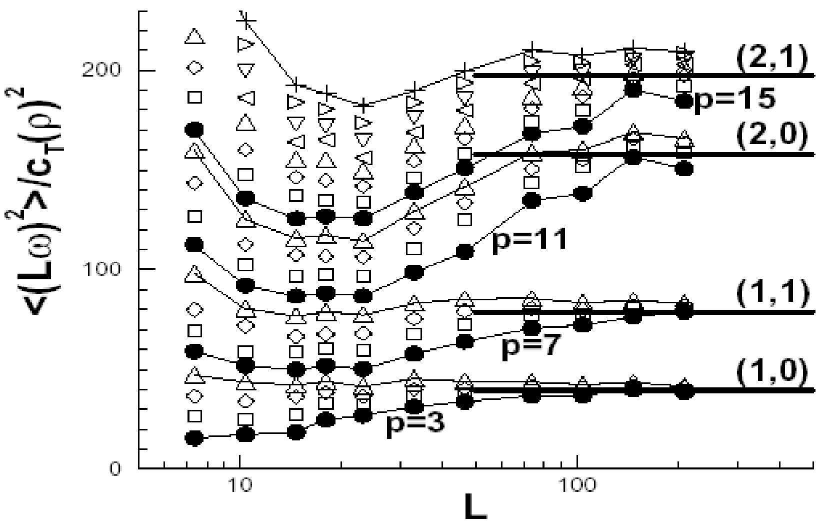
- specific degeneracies associated with 2 quantum numbers of each mode

Results for periodic boundary conditions



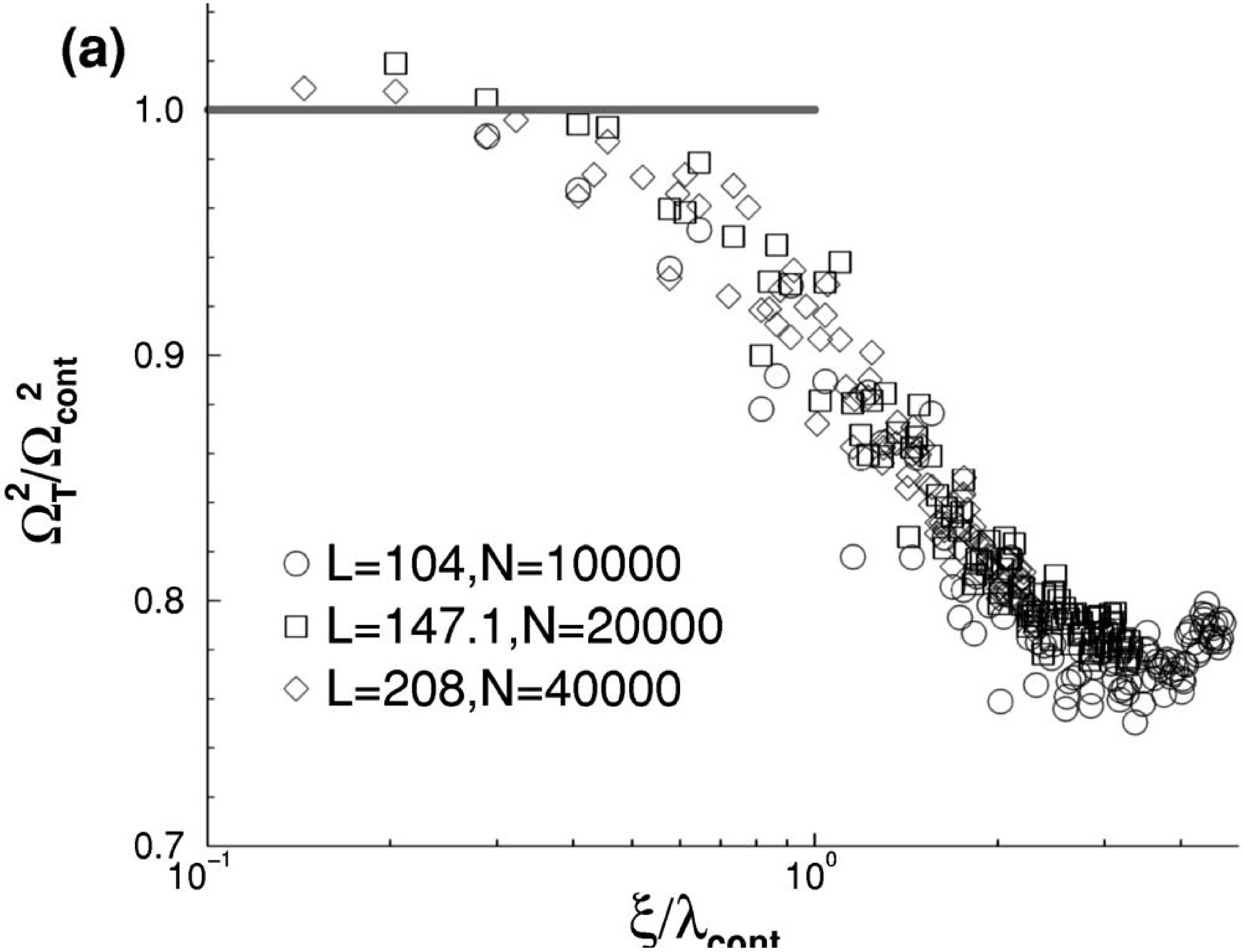


Rescaled frequency vs mode number p



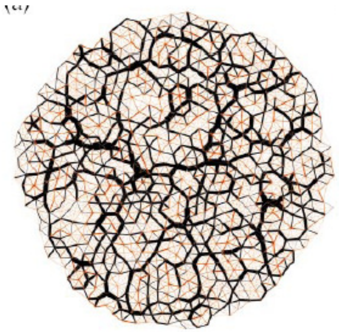
Rescaled frequencies (Lω/c) vs cluster size

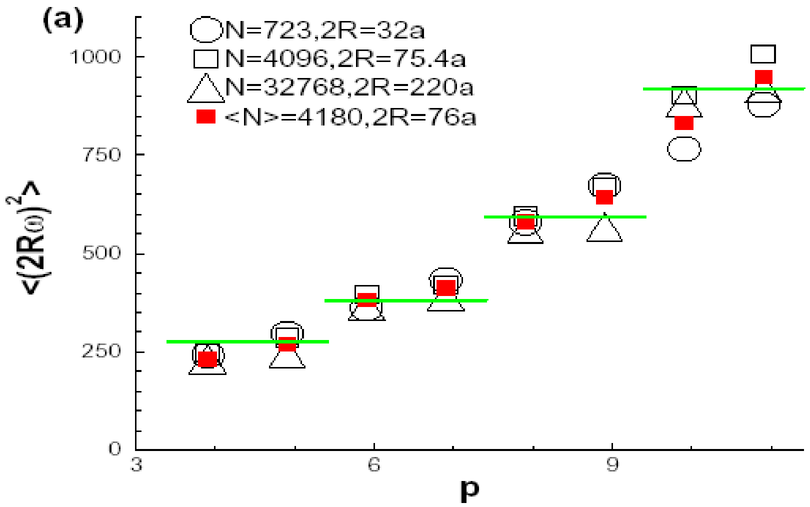
- Horizontal lines are elasticity theory
- Velocity of sound obtained from elastic constants in a large sample



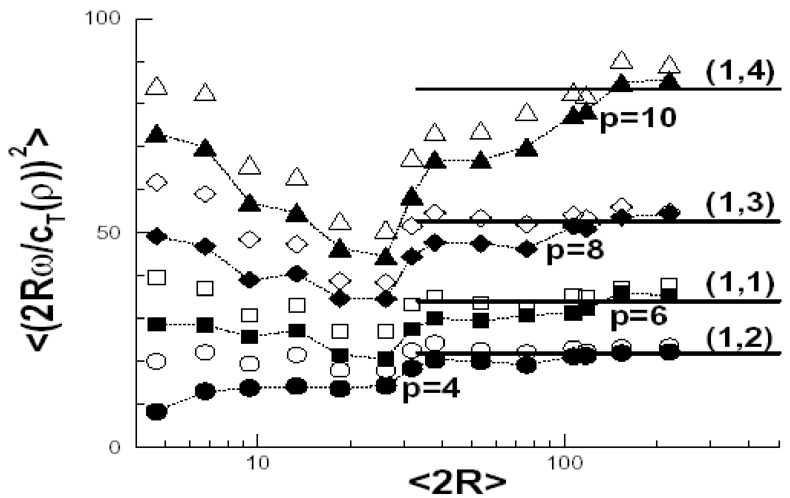
(a)
1.0
0.9
0.8
0.7
10^{-1}
10^{0}
$\Omega_T^2/\Omega_{cont}^2$
ξ/λ_{cont}
L=104,N=10000
L=147.1,N=20000
L=208,N=40000

Results for disk-shaped clusters





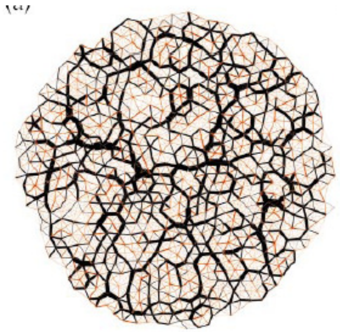
Rescaled frequency vs mode number p

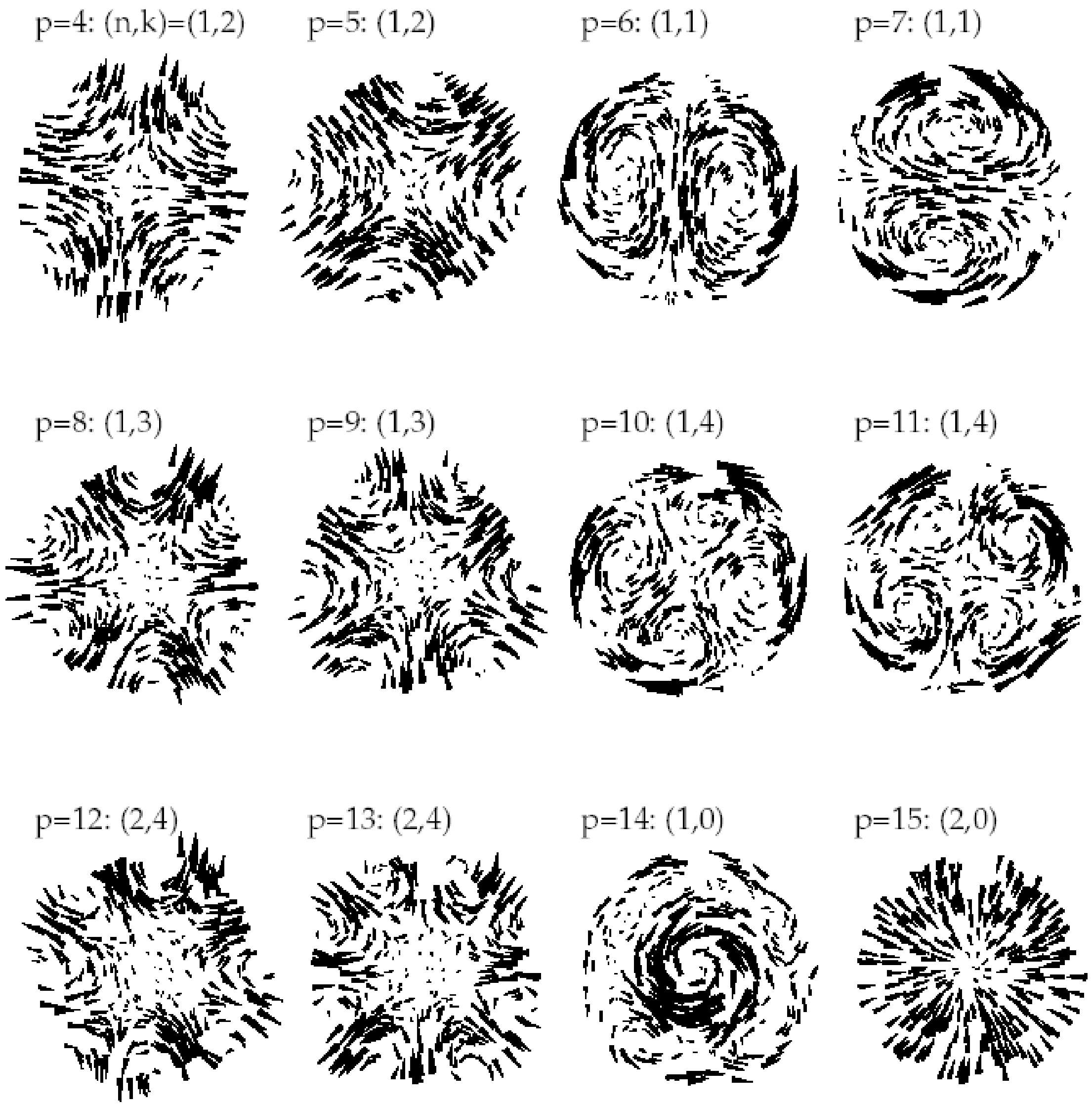


Rescaled frequencies (Rω/c) vs cluster size

Eigenmodes in large disk shape clusters

(continuum prediction: Lamb ca 1900)





- Amorphous systems are inhomogeneous in terms of their linear elastic response
- « Macroscopic » elastic isotropic behaviour observed for large wavelengths (> 50 atomic sizes)
- Consequences for the density of vibrational states ?

<u>Vibrational density of states</u>

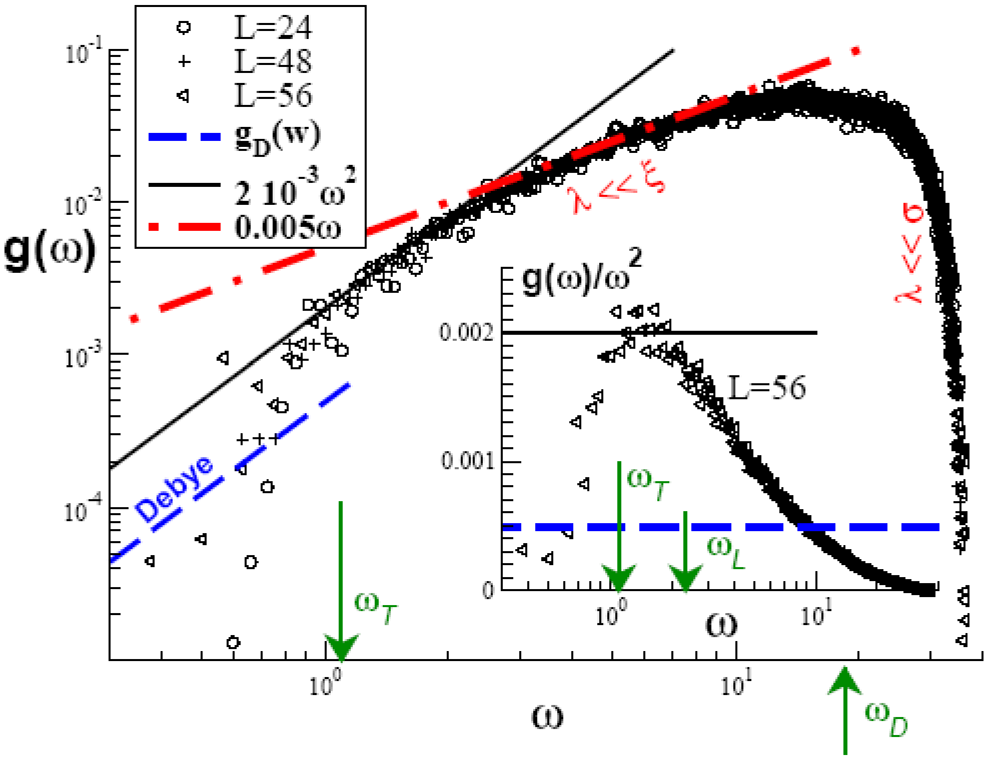
Calculated by exact diagonalization of the dynamical matrix (costly: 3Nx3N) or by Fourier transforming the velocity autocorrelation function

$$Z_\alpha(t) = \frac{1}{N_\alpha}\sum_{i=1}^{N_\alpha}\langle \vec{v}_i(t)\cdot\vec{v}_i(0)\rangle \text{ for species } \alpha$$

Z computed at low temperature (harmonic approximation) using MD:

$$g(\omega) = \sum_\alpha \frac{m_\alpha}{k_B T}\int_{-\infty}^{\infty} dt\, \exp(i\omega t) Z_\alpha(t)$$

VDOS in a 3d Lennard-Jones mixture



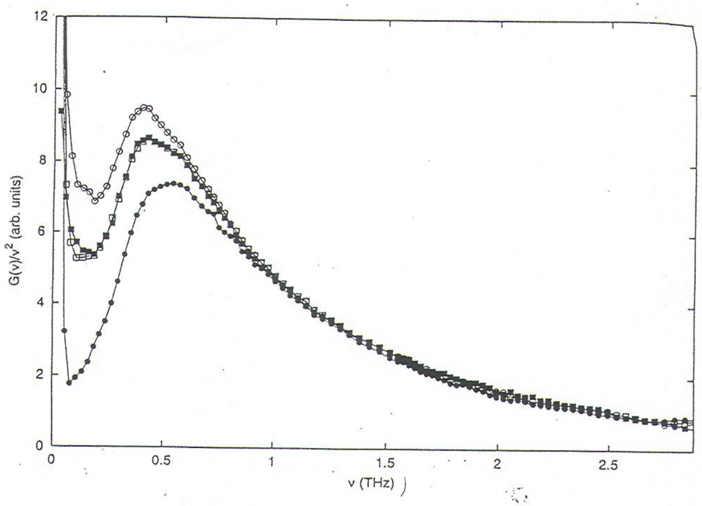
$$\omega_T = 2\pi c_T/\xi$$
$$\omega_L = 2\pi c_L/\xi$$

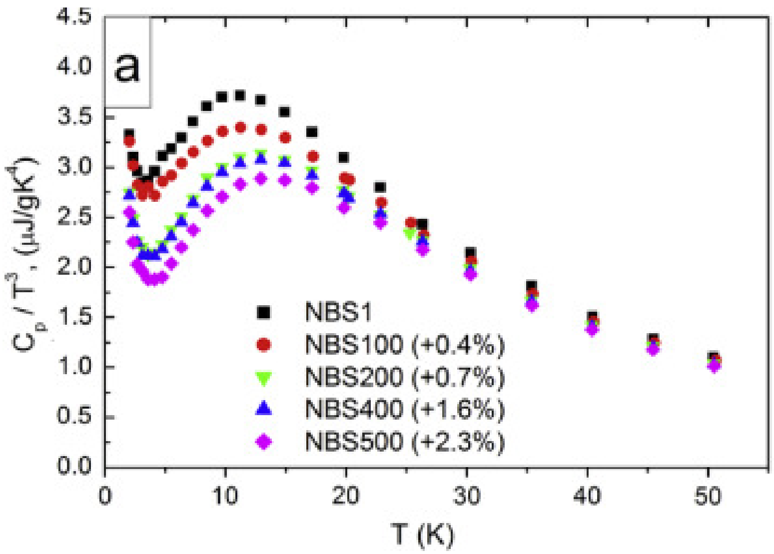
F. Leonforte, A. Tanguy, J. Wittmer, JLB
Phys Rev B 2004, 2005

Debye prediction: $g_D(\omega) \sim \omega^{d-1}$ in d dimensions

Peak in $g(\omega)/\omega^{d-1}$: "Boson peak"

Boson peak observed in many glassy systems, origin controversial





Excess density of states, as compared to Debye prediction, in the Thz region (Neutron scattering PMMA, Duval et al 2001 ; heat capacity SiO2)

Sound waves scattered by soft elastic inhomogeneities

PHYSICAL REVIEW B VOLUME 58, NUMBER 13 1 OCTOBER 1998-I

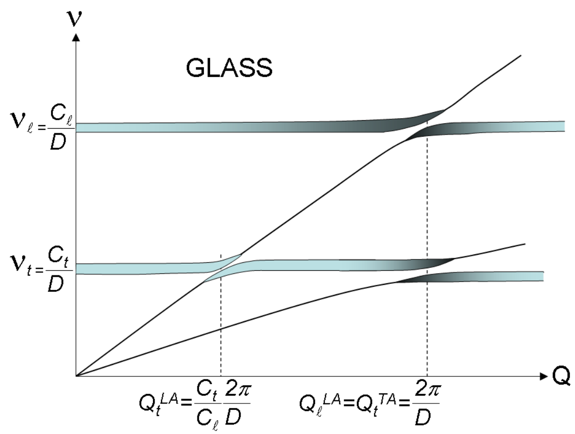
Inelastic x-ray scattering from nonpropagating vibrational modes in glasses

Eugène Duval

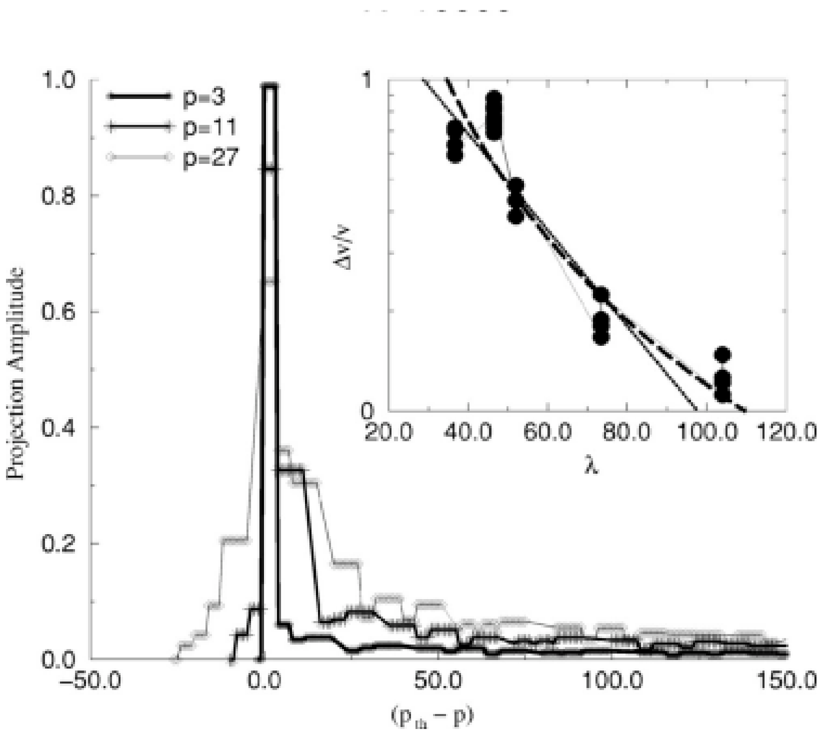
Laboratoire de Physico-Chimie des Matériaux Luminescents, Université Lyon I–UMR-CNRS 5620, 43 Boulevard du 11 Novembre 1918, Bâtiment 205, 69622 Villeurbanne Cedex, France

Alain Mermet

European Synchrotron Radiation Facility, B.P. 220, F-38043 Grenoble Cedex, France



Every eignemodes can be projected onto the plane waves (eignemodes of homogeneous system)

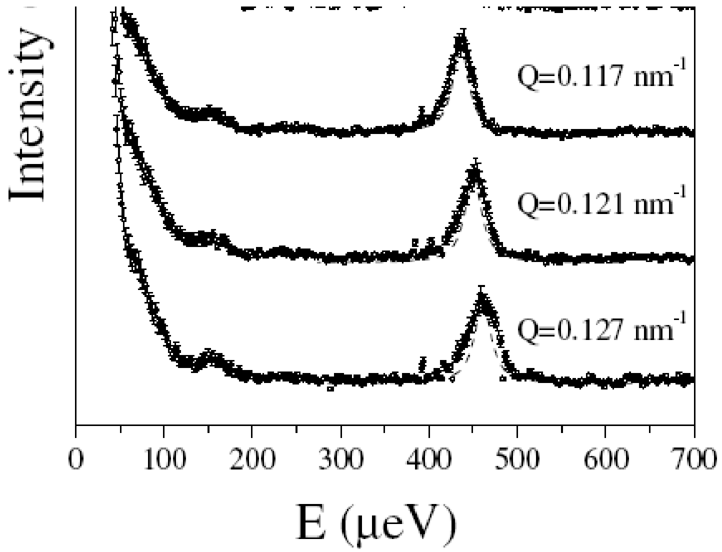


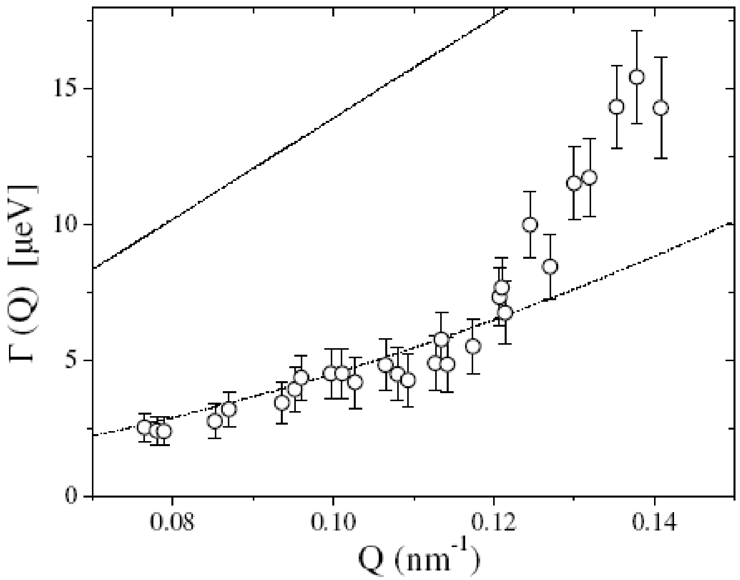
Projection of eigenmodes on plane waves (Tanguy et al, PRB 2005) and magnitude of the « noise » part.

UV Brillouin scattering (Elletra synchrotron, Trieste)

Evidence for a new nano-scale stress correlation in silica glass

C. Masciovecchio[1], G. Baldi[2], S. Caponi[2], L. Comez[3,4], S. Di Fonzo[1], D. Fioretto[3,4], A. Fontana[2,4], A. Gessini[1], S. C. Santucci[1,3], F. Sette[5], G. Viliani[2,4], G. Ruocco[6,4]



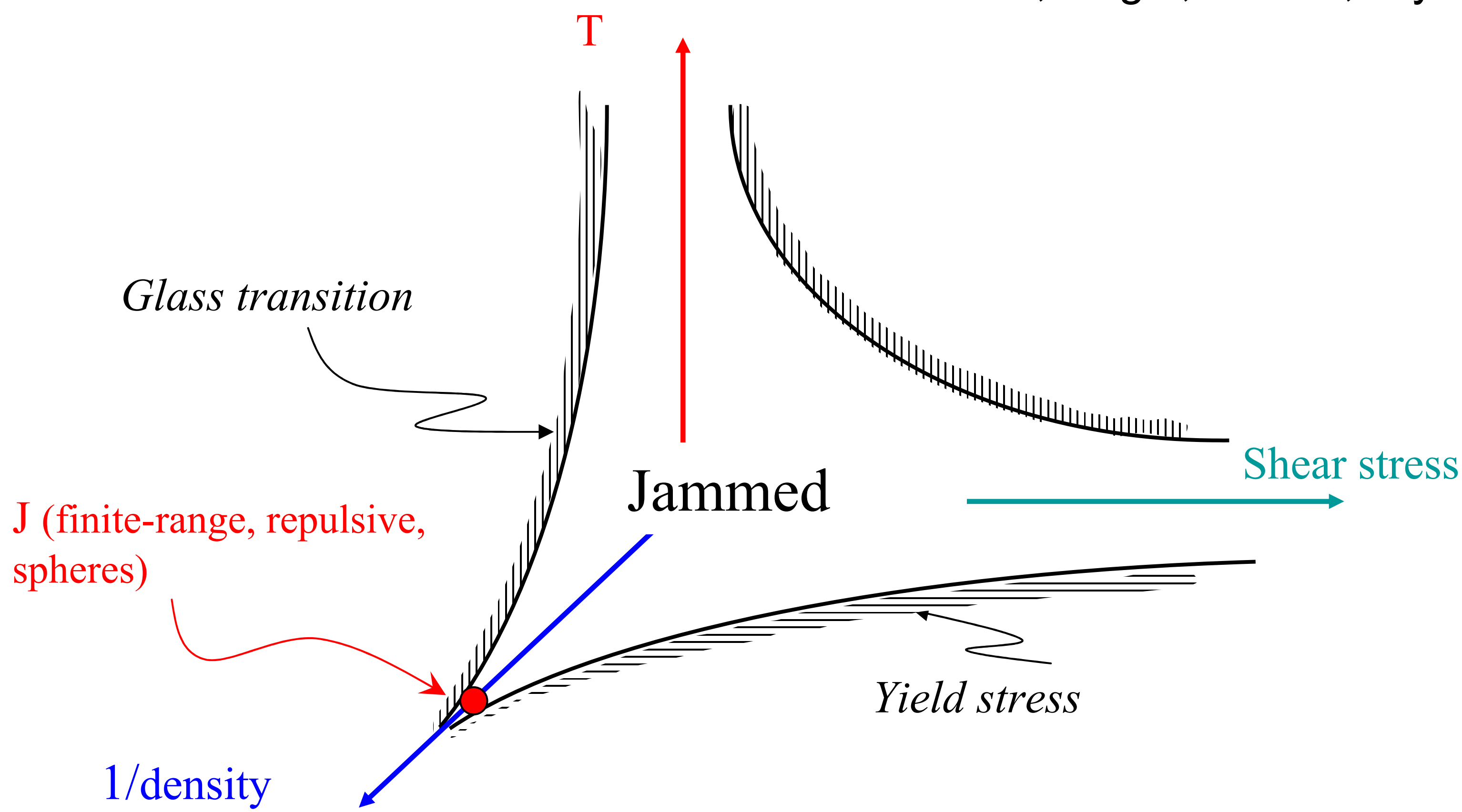


Sound attenuation change for wavelength around 40 nm

Open questions….

- Realistic materials (SiO_2, etc) – Quantitative analysis of Boson peak
- Plasticity ?
- Dynamical heterogeneities (work by Peter Harrowell)?
- Thermal conductivity ?
- Relation to « Jamming » point in granular media ?

Liu, Nagel, O'hern, Wyart



J point reached by progressively increasing the density of a hard sphere packing until overlap removal becomes impossible

Can be studied for any contact potential (e.g. Hertzian contact)

The J point corresponds to an isostatic solid

Minimum number of contacts needed for mechanical stability
Match unknowns (number of interparticle normal forces) to equations

Frictionless spheres in d dimensions:

Number of unknowns per particle = Z/2

Number of equations per sphere = d

$\Rightarrow$ **Z_c = 2d**

Maxwell criterion for rigidity: *global* condition - not *local*.

Friction changes Z_c

Isostatic length scale $\ell^* = 1/|Z - Z_c|$ **diverges at the jamming transition**

$$\ell^{*d}(Z - Z_c) \sim \ell^{*(d-1)}$$

Nonaffine deformation dominates close to point J (see recent review by M. Van Hecke, "Jamming of Soft Particles: Geometry, Mechanics, Scaling and Isostaticity « http://arxiv.org/abs/0911.1384

Shear modulus << Bulk modulus close to jamming, critical behavior…

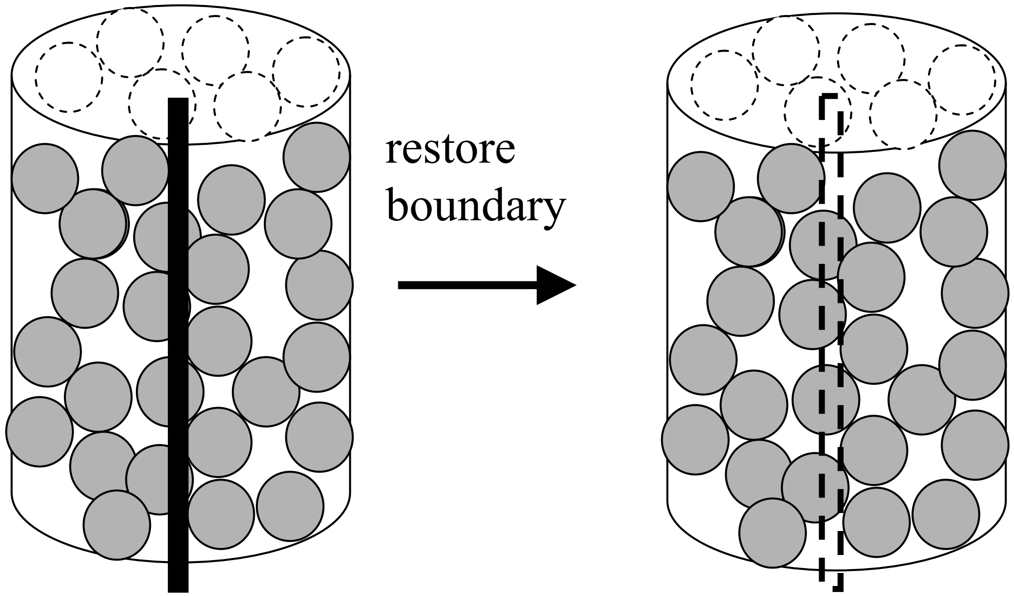
Isostatic solids have an anomalous density of vibrational states at small frequencies

Isostatic packing: excess density of states, $g(\omega) \sim \omega^0$

Construct low-ω modes from soft modes (Matthieu Wyart, Tom Witten, Sid Nagel)

n modes with

$\omega = 0$

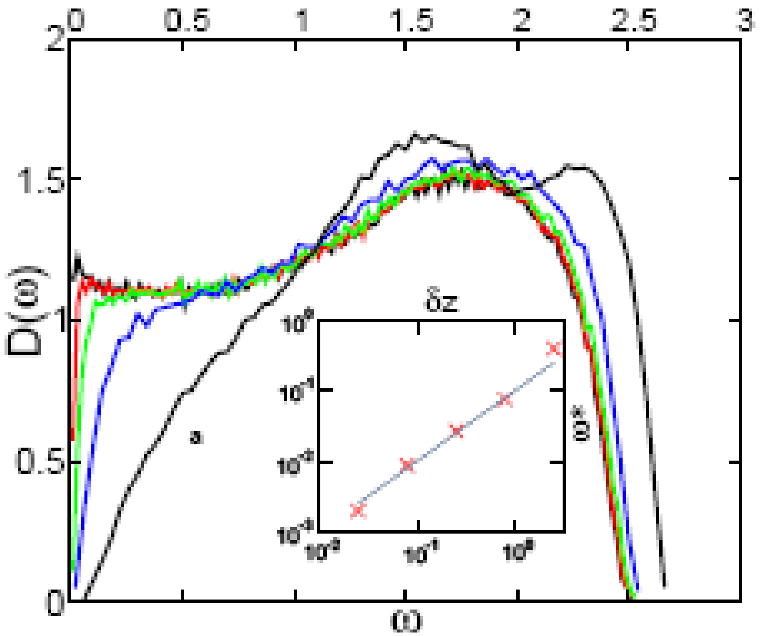


Where are the modes with ω=0 ?

$N(L) \sim L^{d-1}$ floppy modes (from cutting boundaries). These modes are found in an interval $\omega \sim 1/L$

$g(\omega) \sim N / (\omega L^d) \sim L^0$

g(ω)



Wyart, O'Hern, Liu, Nagel, Silbert

Questions

- Relevance of point J to the glassy state ?
- Relevance of anomalous d.o.s. at point J to the Boson peak and anomalous vibrations in glasses ?