Long-term Workshop on
Frontiers in Non-equilibrium Physics 2024
Yukawa Institute for Theoretical Physics,
Kyoto University, Japan

August 1st (Thu.), 2024
10:20－11:10 (35+15mins)

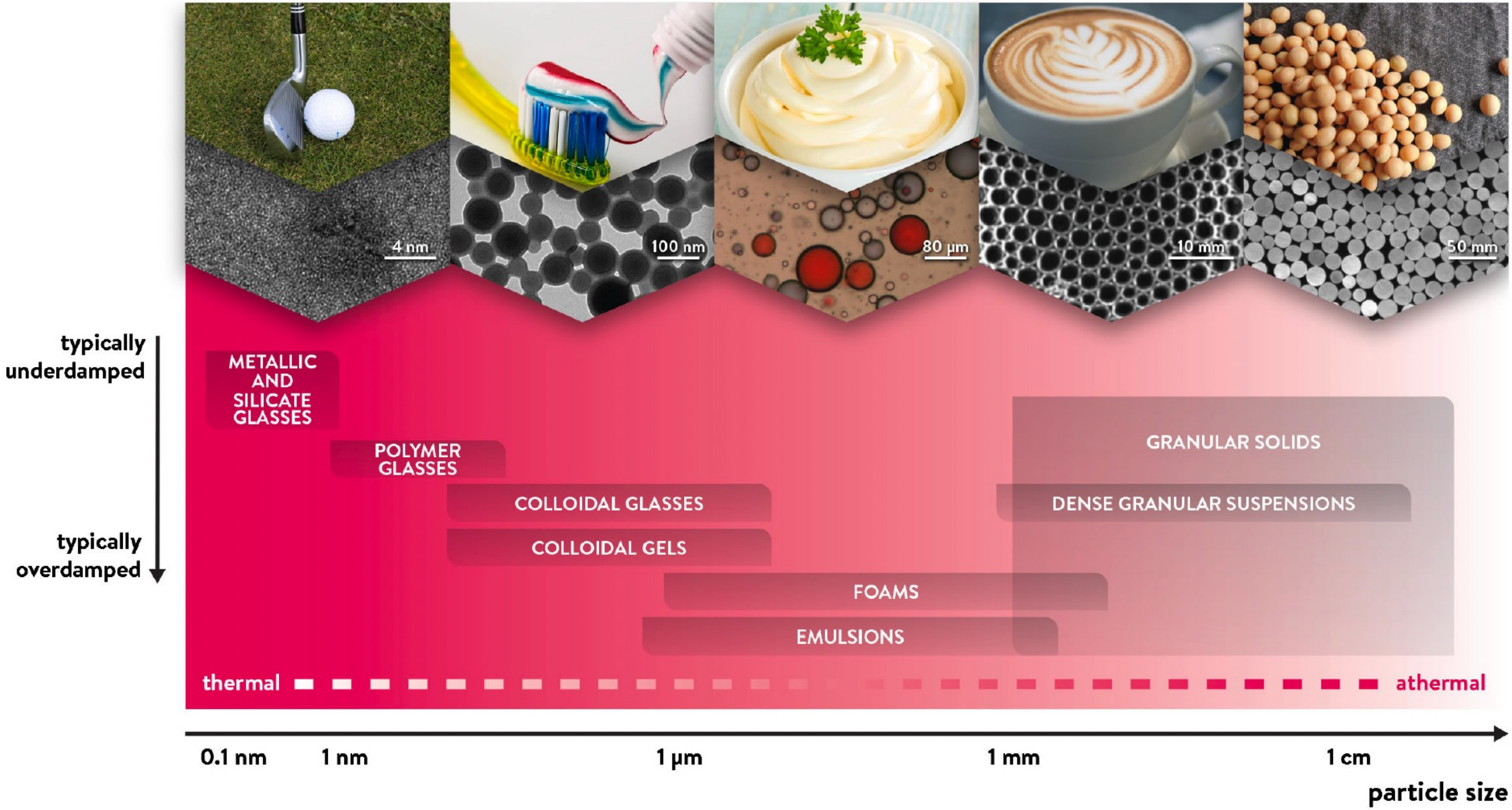
Effective medium theory for viscoelasticity of soft jammed solids

Hideyuki Mizuno

The University of Tokyo

Mizuno and Ikeda, arXiv:2407.15323

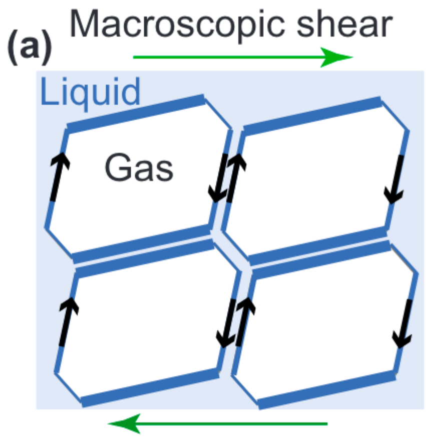
Soft jammed solids: foams, emulsions, soft colloids



✓ There are various amorphous solids at various scales in our world

Nicolas et al., Rev. Mod. Phys. (2018)

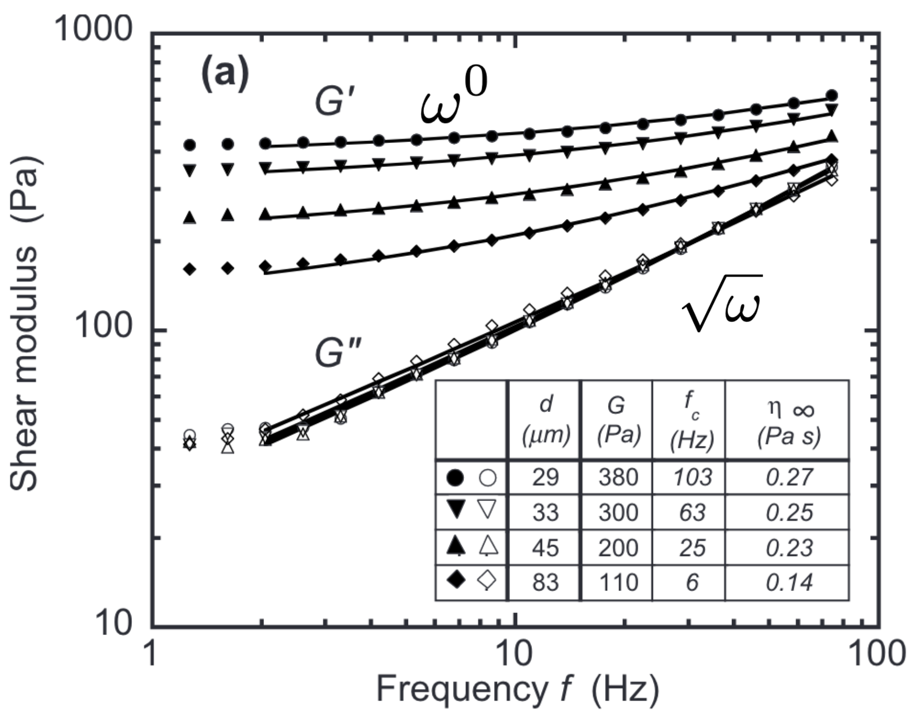
Experimental measurements (1/2)



Macrorheology experiment

Aqueous foams
subjected to an oscillatory shear

→ Measure complex modulus



✓ Storage modulus (real part)

$$G' \propto \omega^0$$

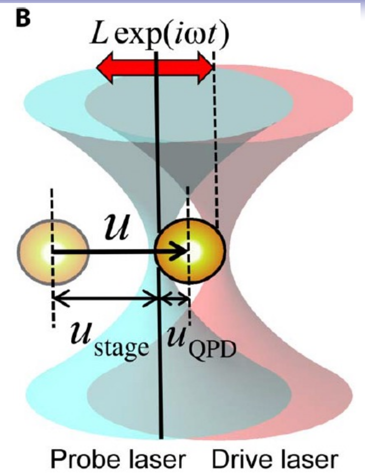
✓ Loss modulus (imaginary part)

$$G'' \propto \sqrt{\omega}$$

Anomalous viscous loss has been observed in many experiments

Krishan, et al., PRE 2010

Experimental measurements (2/2)

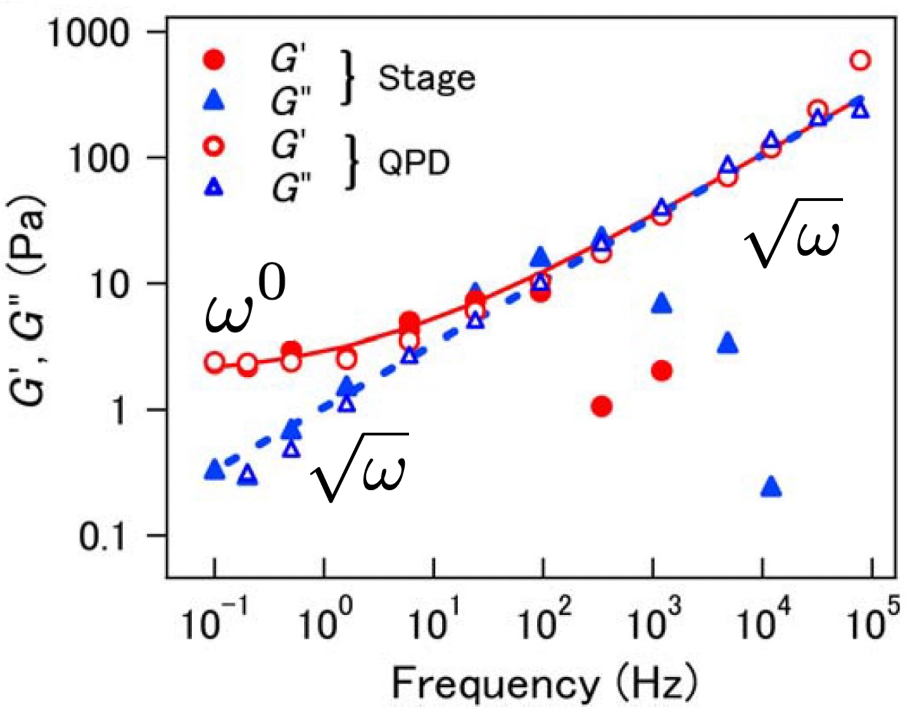


Microrheology experiment

Measure microscopic displacements of a probe particle $\alpha(\omega)$

→ Transform to complex modulus using generalized Stokes relation

$$G(\omega) = \frac{1}{6\pi a \alpha(\omega)}$$



- Lower frequencies

$$G' \propto \omega^0 \quad G'' \propto \sqrt{\omega}$$

Anomalous viscous loss

- higher frequencies

$$G' \approx G'' \propto \sqrt{\omega}$$

Nishizawa, et al., Sci. Adv. 2017

Theoretical understanding (1/4)

✓ Contact force (harmonic potential):

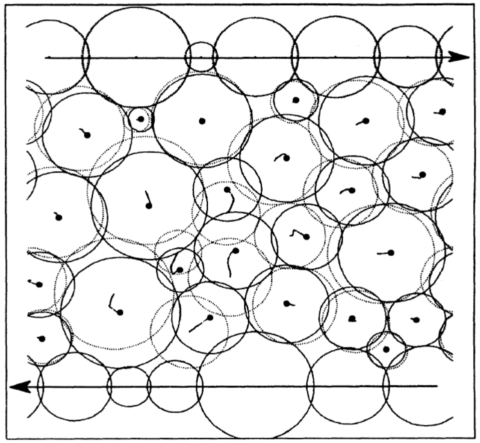
$$\phi(r) = \frac{k}{2}(\sigma - r)^2 H(\sigma - r)$$

✓ Contact damping:

$$\vec{f}^{\mathrm{visc}} = b\Delta\vec{v}$$

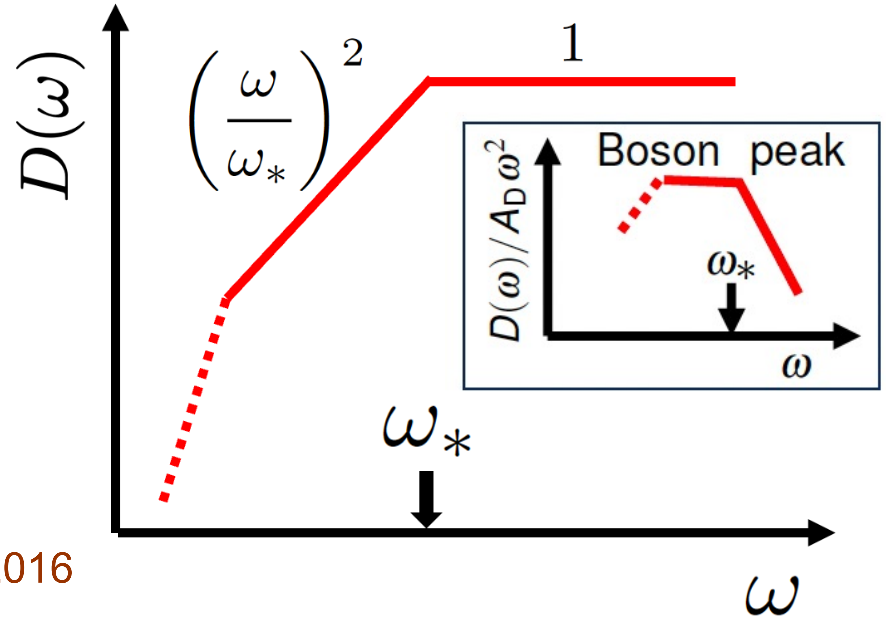
Strong damping due to viscous forces

Durian, PRL 1995



Vibrational density of states (vDOS):

$$D(\omega) \propto \begin{cases} \left(\frac{\omega}{\omega_*}\right)^2 & (\omega < \omega_*), \\ 1 & (\omega_* < \omega). \end{cases}$$



Silbert et al., PRL 2005; Charbonneau et al., PRL 2016

Theoretical understanding (2/4)

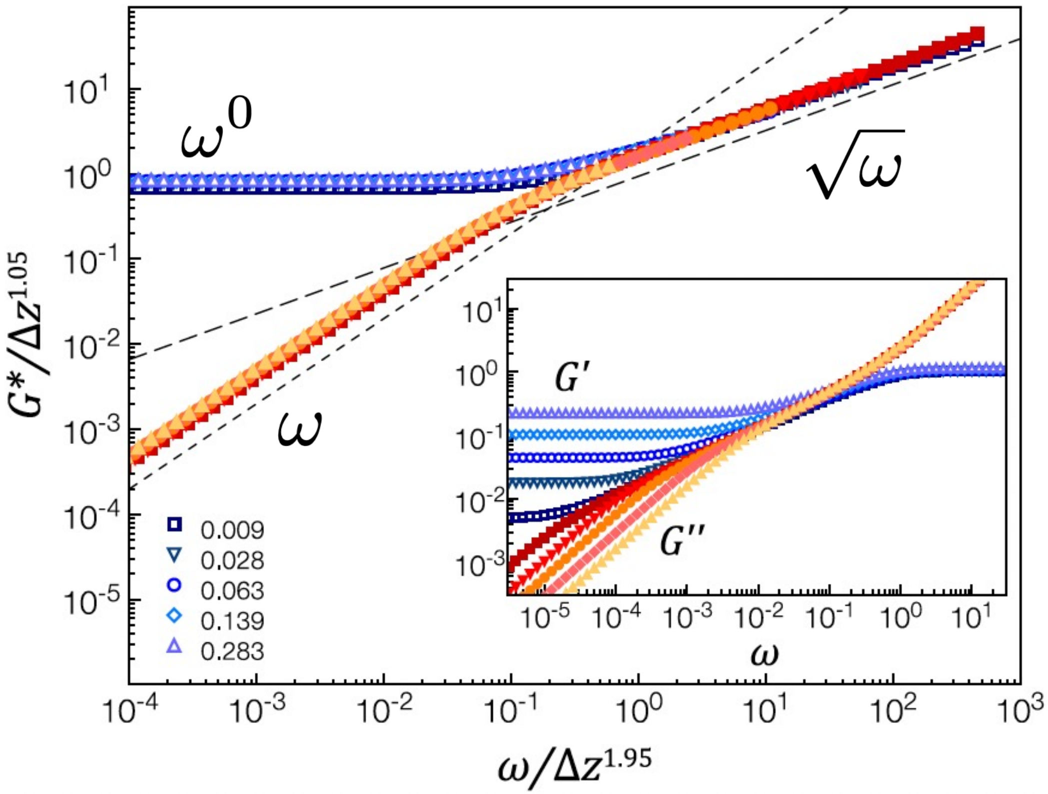
Macrorheology

Tighe, PRL 2011

Formulate complex modulus based on vDOS

$$\frac{1}{G^*(\omega)} \gtrsim \int_{s^*}^{1} \frac{s^{-1/2} ds}{s + \imath\omega}$$

$s = \omega'^2$: eigenvalues



✓ Scaling laws

$$G' \sim \begin{cases} \Delta z \\ \omega^{1/2} \\ 1 \end{cases}$$

$$G'' \sim \begin{cases} \omega/\Delta z, & \omega \lesssim s^*, \\ \omega^{1/2}, & s^* \lesssim \omega \lesssim 1, \\ \omega, & 1 \lesssim \omega. \end{cases}$$

- higher frequencies

$$G' \approx G'' \propto \sqrt{\omega}$$

Theoretical understanding (3/4)

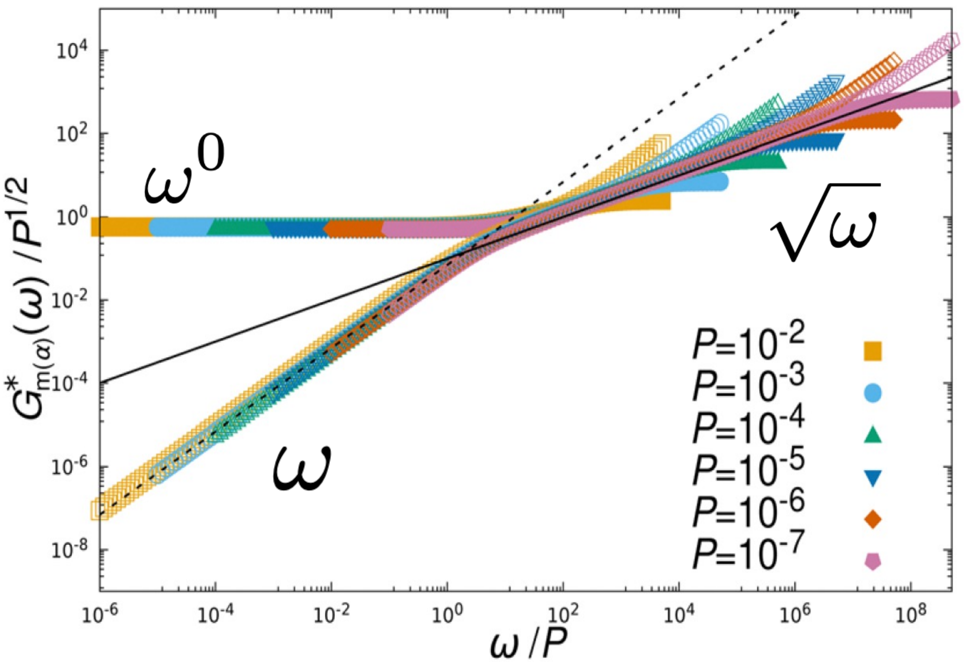
Microrheology

Hara, et al., Soft Matter 2023

$$\frac{1}{G^*(\omega)} = \int d\omega' \frac{D(\omega')}{(\omega')^2 + i\omega}$$

Formulate complex modulus based on vDOS

ω' : eigenfrequency



✓ Scaling laws

$$G^*_{m(\alpha)}(\omega) \approx \begin{cases} \sqrt{\omega} + i\sqrt{\omega} & ({\omega_*}^2 \ll \omega \ll 1) \\ \omega_* + i\dfrac{\omega}{\omega_*} & ({\omega_*}^3 \ll \omega \ll {\omega_*}^2) \\ \omega_* + i\sqrt{\omega_* \omega} & (\omega \ll {\omega_*}^3) \end{cases}$$

- higher frequencies

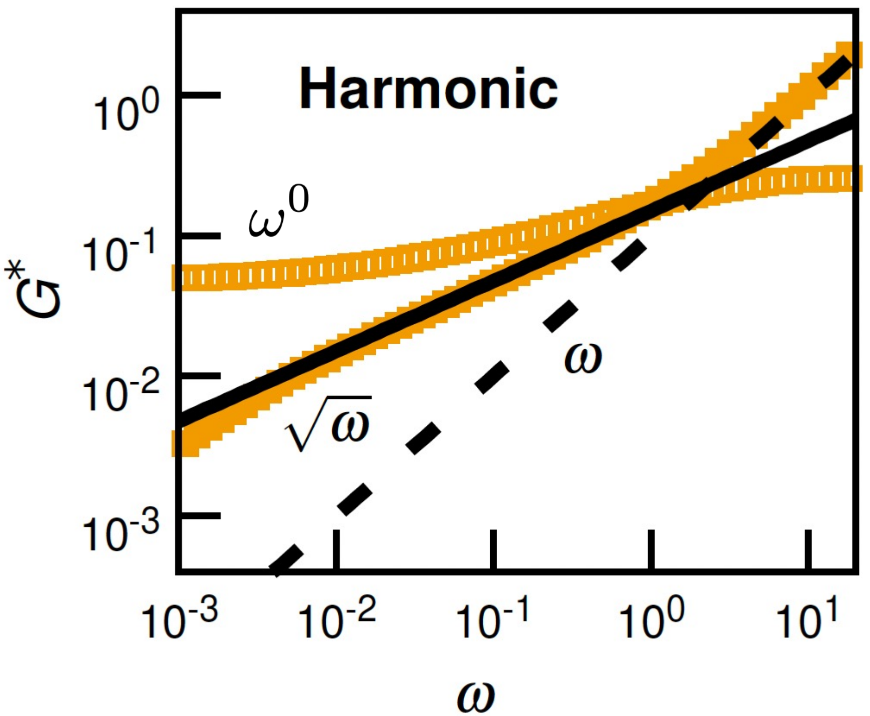
$$G' \approx G'' \propto \sqrt{\omega}$$

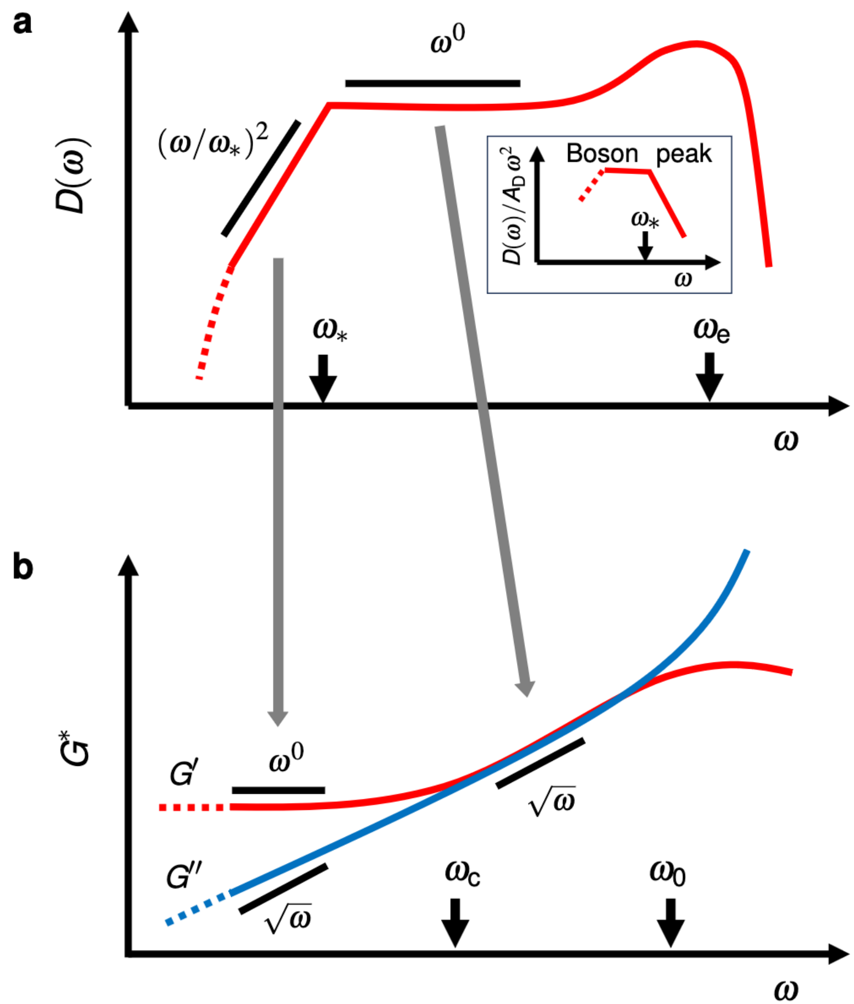
Theoretical understanding (4/4)

Microrheology

Hara, et al., arXiv 2024

$$\frac{1}{G^*(\omega)} = \int d\omega' \frac{D(\omega')}{(\omega')^2 + i\omega}$$





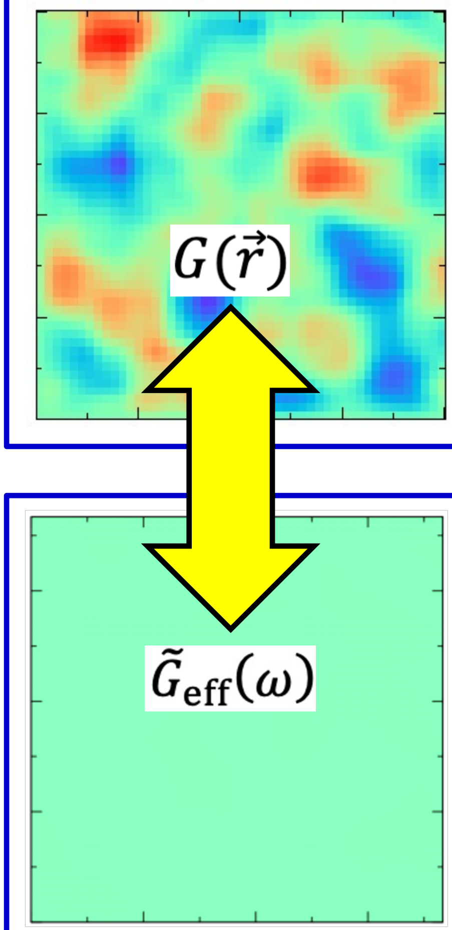
Anomalous viscous loss is linked to non-Debye scaling law (boson peak)

Microscopic theory for soft jammed solids

Effective medium theory (EMT)
[or coherent potential approximation (CPA) theory]

- ✓ Electronic energy levels in disordered metallic alloys
  Yonezawa and Morigaki, Progress of Theoretical Physics Supplement 1973
- ✓ Conductance in electrical resistor networks
  Kirkpatrick, Rev. Mod. Phys. 1973
- ✓ Rigidity percolation problem in spring networks
  Feng et al., PRB 1985; He and Thorpe, PRL 1985
- ✓ Heterogeneous elasticity theory for glasses
  Schirmacher et al., EPL 2006, PRL 2006

Effective medium theory: Heterogeneous elasticity theory for glasses



✓ Equation of motion with fluctuating shear modulus

$$-\omega^2\vec{u}(\vec{r},\omega) = \frac{1}{\rho}\left(K + \frac{G(\vec{r})}{3}\right)\vec{\nabla}\left(\vec{\nabla}\cdot\vec{u}(\vec{r},\omega)\right) + \frac{G(\vec{r})}{\rho}\vec{\nabla}^2\vec{u}(\vec{r},\omega)$$

✓ Green function

$$\mathcal{G}(\vec{r},\omega) = \left[-\omega^2 - \left(\tilde{K} + \frac{\tilde{G}(\vec{r})}{3}\right)\vec{\nabla}(\vec{\nabla}\cdot) - \tilde{G}(\vec{r})\vec{\nabla}^2\right]^{-1} I\delta(\vec{r})$$

✓ Effective Green function

$$\mathcal{G}_{\mathrm{eff}}(\vec{r},\omega) = \mathcal{M}_{\mathrm{eff}}^{-1}(\vec{r},\omega) I\delta(\vec{r}),$$

$$\mathcal{M}_{\mathrm{eff}}(\vec{r},\omega) = -\omega^2 - \left(\tilde{K} + \frac{\tilde{G}_{\mathrm{eff}}(\omega)}{3}\right)\vec{\nabla}(\vec{\nabla}\cdot) - \tilde{G}_{\mathrm{eff}}(\omega)\vec{\nabla}^2,$$

Theory explains characteristic properties of glasses
: nonaffine deformation, boson peak, scattering of sound waves, low thermal conductivity

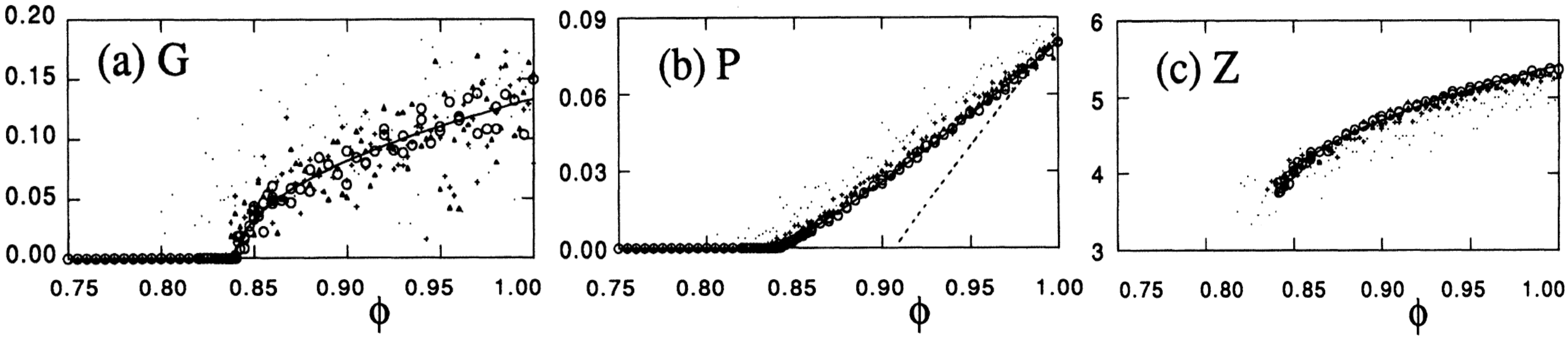
Schirmacher et al., EPL 2006, PRL 2006

Effective medium theory: Jammed amorphous solids (1/4)

✓ Contact force (harmonic potential): $\phi(r) = \frac{k}{2}(\sigma - r)^2 H(\sigma - r)$

Durian, PRL 1995

Shear modulus — Pressure — Contact number



$G \propto (\phi - \phi_J)^{1/2}$ $\quad P \propto \phi - \phi_J$ $\quad z - z_c \propto (\phi - \phi_J)^{1/2}$

✓ Jamming transition occurs at $\phi = \phi_J$

✓ At the transition, the system becomes isostatic with contact number $z_c = 2d$

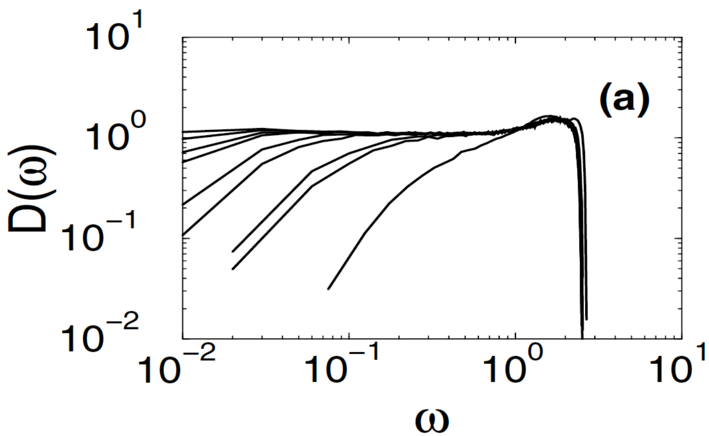
✓ Above the jamming, excess contact number controls the rigidity

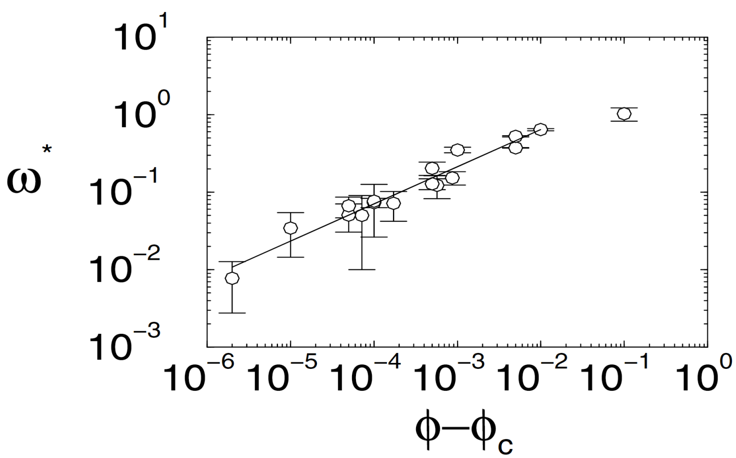
$$G \propto z - z_c = \delta z$$

Effective medium theory: Jammed amorphous solids (2/4)

✓ Contact force (harmonic potential): $\phi(r) = \dfrac{k}{2}(\sigma - r)^2 H(\sigma - r)$

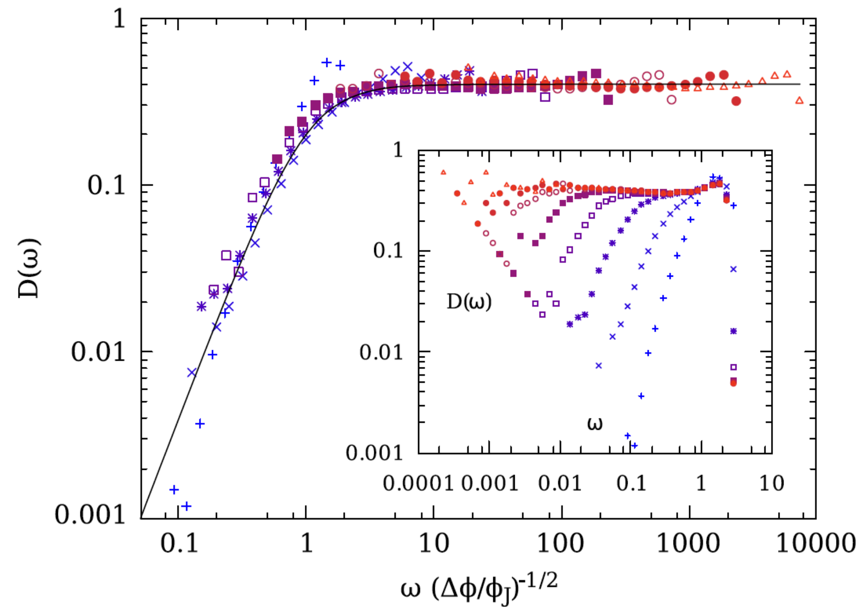
Silbert et al., PRL 2005





$$\omega_* \propto (\phi - \phi_J)^{1/2} \propto \delta z$$

Charbonneau et al., PRL 2016



$$D(\omega) \propto \begin{cases} \left(\dfrac{\omega}{\omega_*}\right)^2 & (\omega < \omega_*), \\ 1 & (\omega_* < \omega). \end{cases}$$

Effective medium theory: Jammed amorphous solids (3/4)

✓ Random spring network

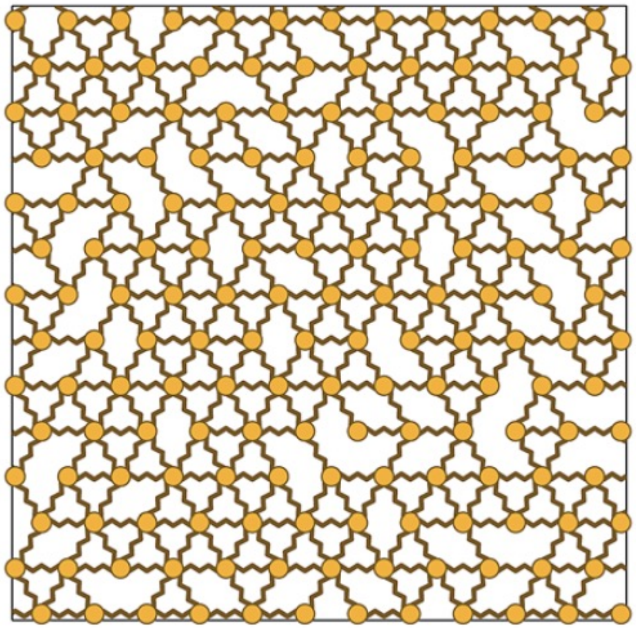
Feng et al., PRB 1985

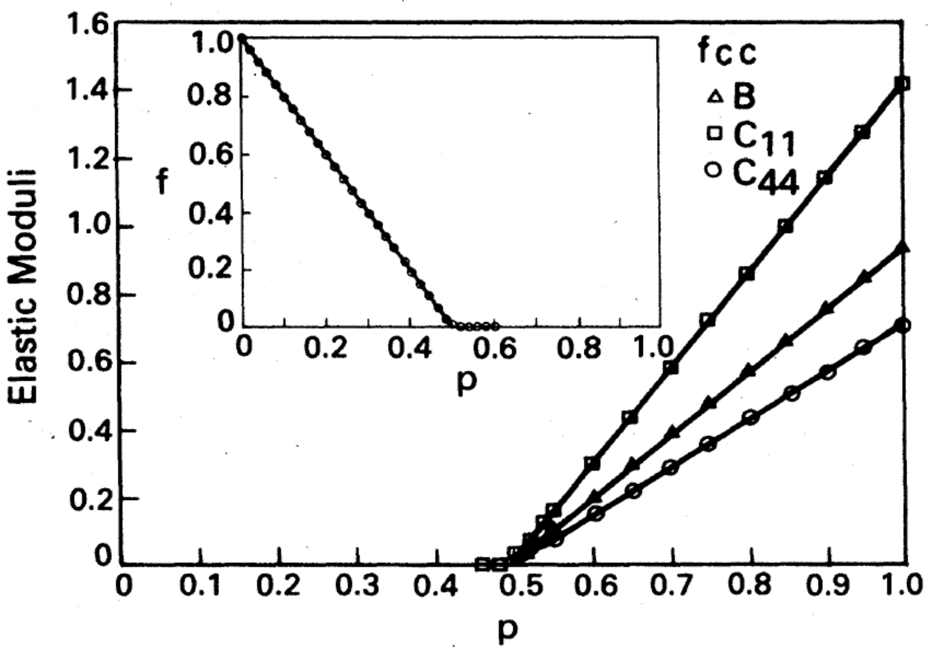
$$V = \sum_{\langle ij \rangle} \frac{k_{ij}}{2} \left[(\vec{u}_i - \vec{u}_j) \cdot \vec{n}_{ij} \right]^2$$

$Nz/2$ pairs of particles are connected

$$P(k_{ij}) = \frac{z}{z_0} \delta(k_{ij} - 1) + \left(1 - \frac{z}{z_0}\right) \delta(k_{ij})$$

$$p = \frac{z}{z_0}$$





$$G \propto z - z_c = \delta z$$

Effective medium theory: Jammed amorphous solids (4/4)

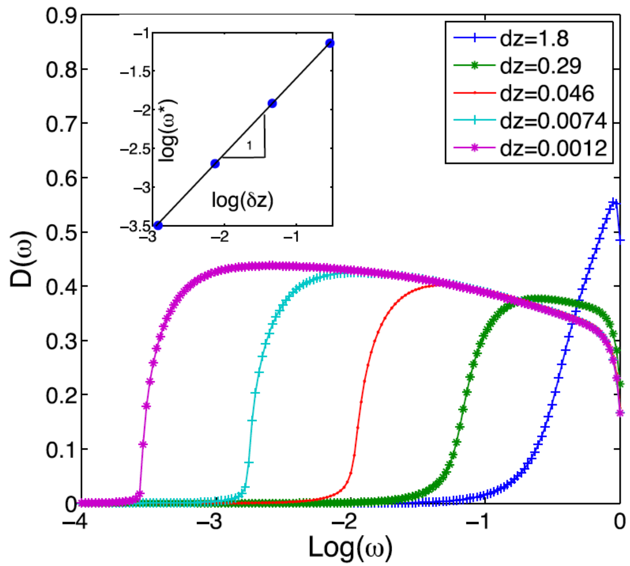
✓ Random spring network

Wyart, EPL 2010; DeGiuli et al., Soft Matter 2014

$$V = \sum_{\langle ij \rangle} \left\{ \frac{k_{ij}}{2} \left[(\vec{u}_i - \vec{u}_j) \cdot \vec{n}_{ij} \right]^2 - \frac{f_{ij}}{2\sigma_0} \left[(\vec{u}_i - \vec{u}_j) \cdot \vec{n}_{ij}^{\perp} \right]^2 \right\}$$

Energy reduction due to repulsive forces

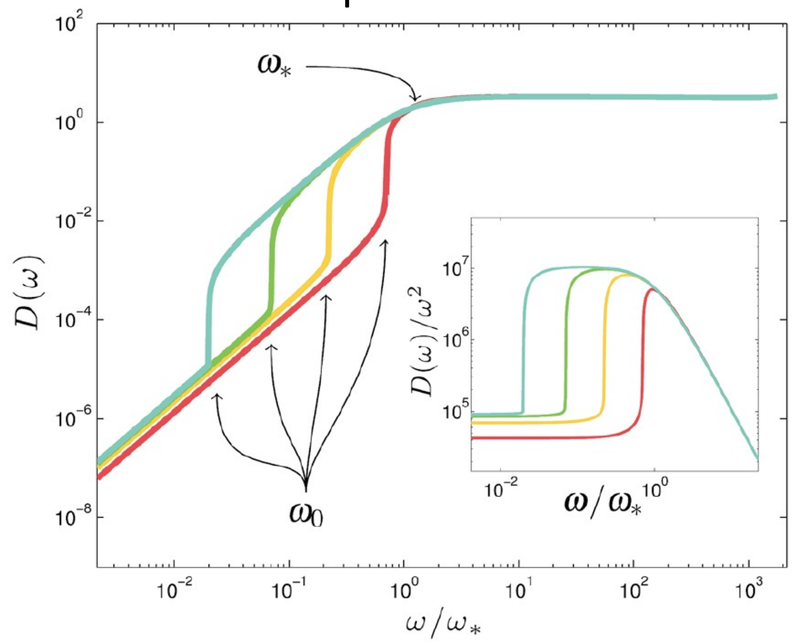
Without repulsive forces



$$\omega_* \propto z - z_c = \delta z$$

$D(\omega) \propto 1$ from connectivity

With repulsive forces

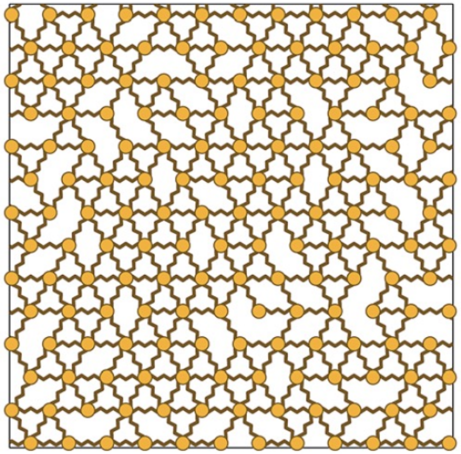


$D(\omega) \propto \left(\frac{\omega}{\omega_*}\right)^2$ from repulsive forces

Present work

Aim

- ✓ Develop EMT for viscoelasticity
- ✓ Macrorheology and microrheology
- ✓ Random spring network model
- ✓ Integrate contact damping effect



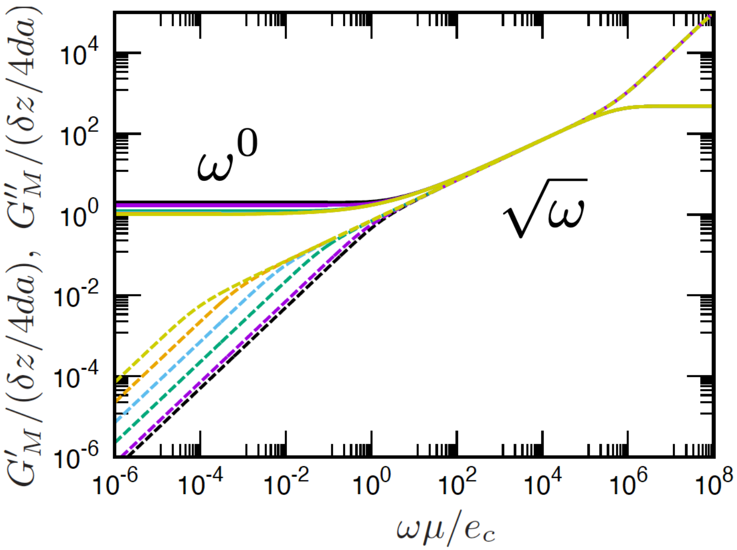
Results

- ✓ Explain experimental results
  - higher frequencies

    $G' \approx G'' \propto \sqrt{\omega}$
  - Lower frequencies

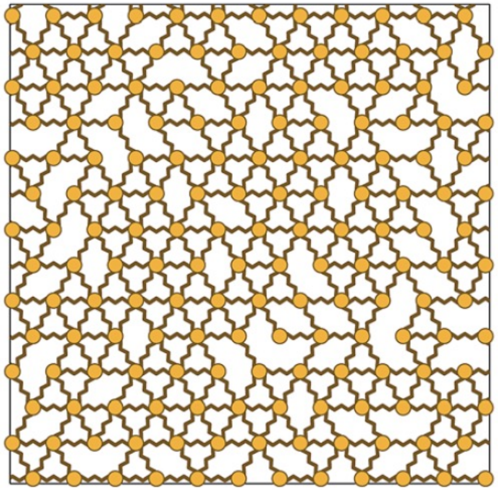
    $G' \propto \omega^0 \qquad G'' \propto \sqrt{\omega}$

    Anomalous viscous loss



Random spring network model (1/3)

Feng et al., PRB 1985; Wyart, EPL 2010; DeGiuli et al., Soft Matter 2014



✓ Point particles placed at lattice sites with contact number z_0

✓ Connect nearest neighbors' particles by springs

✓ Repulsive forces between connected pairs of particles

$$e_{ij} = \frac{f_{ij}}{k_{ij}\sigma_0} \equiv e \ (> 0)$$: Prestress

✓ Randomly cut springs -> control contact number $z \ (< z_0)$

$$V = \sum_{\langle ij \rangle} \left\{ \frac{k_{ij}}{2} \left[(\vec{u}_i - \vec{u}_j) \cdot \vec{n}_{ij} \right]^2 - \frac{f_{ij}}{2\sigma_0} \left[(\vec{u}_i - \vec{u}_j) \cdot \vec{n}_{ij}^{\perp} \right]^2 \right\}$$

$$P(k_{ij}) = \frac{z}{z_0} \delta(k_{ij} - 1) + \left(1 - \frac{z}{z_0} \right) \delta(k_{ij})$$

✓ Contact damping occurs between connected pairs of particles

$$\vec{f}_i^{\mathrm{damp}} = -\mu(\vec{v}_i - \vec{v}_j)$$ μ : Viscosity

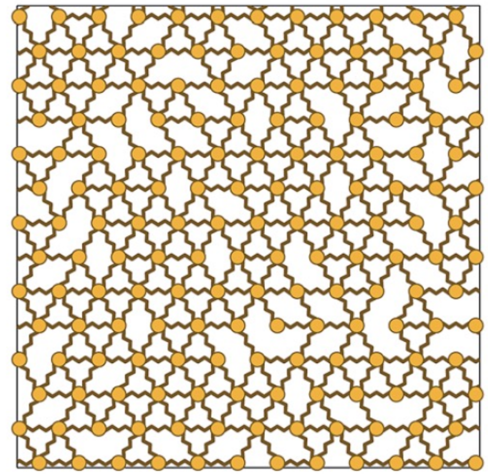
Random spring network model (2/3)

✓ Equation of motion (overdamped dynamics)

$$C\frac{d}{dt}|u\rangle = -M|u\rangle + |F\rangle$$

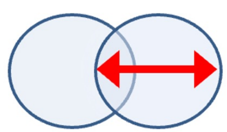
$$|u\rangle = [\vec{u}_1, \cdots, \vec{u}_N]$$: Displacement

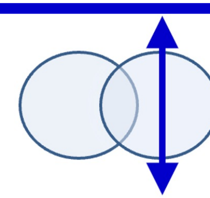
$$|F\rangle = \left[\vec{F}_1, \cdots, \vec{F}_N\right]$$: External force



$$M = \frac{\partial^2 V}{\partial\vec{r}\partial\vec{r}} = \sum_{\langle ij\rangle} |ij\rangle k_{ij}\left[\vec{n}_{ij}\otimes\vec{n}_{ij} - e\left(I_d - \vec{n}_{ij}\otimes\vec{n}_{ij}\right)\right]\langle ij|$$

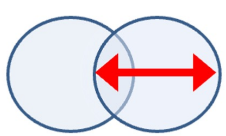
: Hessian matrix (contact force)

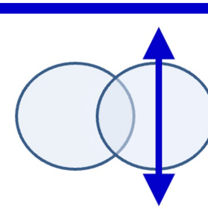




$$C = \sum_{\langle ij\rangle} |ij\rangle \mu k_{ij}\left[\vec{n}_{ij}\otimes\vec{n}_{ij} + \left(I_d - \vec{n}_{ij}\otimes\vec{n}_{ij}\right)\right]\langle ij|$$

: Damping matrix (contact damping)



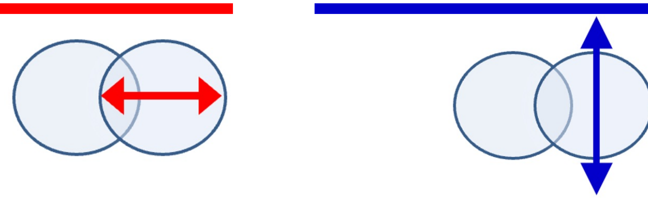


Random spring network model (3/3)

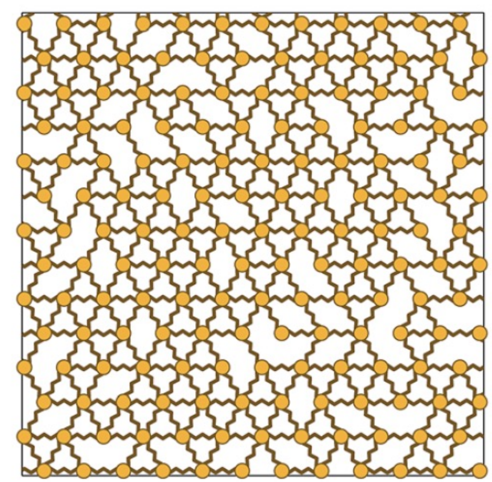
$$G(\omega\mu) = (M - i\omega C)^{-1} = \widetilde{M}^{-1}$$: Green function

$$\widetilde{M} = M - i\omega C$$: Complex hessian matrix

$$= \sum_{\langle ij \rangle} |ij\rangle \widetilde{k}_{ij} \left[\vec{n}_{ij} \otimes \vec{n}_{ij} - \widetilde{e}\left(I_d - \vec{n}_{ij} \otimes \vec{n}_{ij}\right)\right] \langle ij|$$



$$P\left(\widetilde{k}_{ij}\right) = \frac{z}{z_0}\delta\left(\widetilde{k}_{ij} - \widetilde{k}\right) + \left(1 - \frac{z}{z_0}\right)\delta\left(\widetilde{k}_{ij}\right)$$



$$\widetilde{k} = 1 - i\omega\mu$$: Complex stiffness

$$\widetilde{e} = \frac{e + i\omega\mu}{1 - i\omega\mu}$$: Complex prestress

✓ Effects of contact damping are integrated into complex stiffness and prestress

-> We can apply the effective medium theory

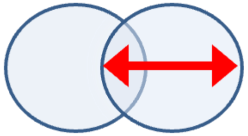
Effective medium theory (1/2)

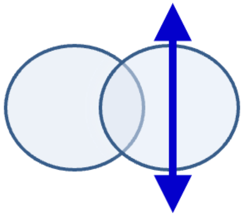
DeGiuli et al., Soft Matter 2014

✓ Introduce effective green function

$$G_{\text{eff}}(\omega\mu) = M_{\text{eff}}^{-1}$$

$$M_{\text{eff}} = \sum_{\langle ij \rangle} |ij\rangle \left[\underline{k^{\|} \vec{n}_{ij} \otimes \vec{n}_{ij}} - \underline{\tilde{e} k^{\perp} \left(I_d - \vec{n}_{ij} \otimes \vec{n}_{ij} \right)} \right]$$





$k_{\text{eff}} = k^{\|} - (d-1)\tilde{e} k^{\perp}$: Effective stiffness

$G^{\|} = \vec{n}_{ij} \langle ij | G_{\text{eff}} | ij \rangle \vec{n}_{ij}$: Longitudinal component

$G^{\perp} = \frac{1}{d-1} \left[\mathrm{Tr} \langle ij | G_{\text{eff}} | ij \rangle - G^{\|} \right]$: Transverse component

✓ Effective medium theory maps Green function G to effective G_{eff}

Effective medium theory (2/2)

✓ Green function $G = G_{\mathrm{eff}} + G_{\mathrm{eff}} T G_{\mathrm{eff}}$ DeGiuli et al., Soft Matter 2014

Transfer matrix

⇩ $\langle\rangle = \int d\widetilde{k}_{ij} P(\widetilde{k}_{ij})$

✓ EMT equation $\langle G \rangle = G_{\mathrm{eff}} \Longleftrightarrow \langle T \rangle = 0$

$$\Longleftrightarrow \quad G^{\parallel} = \frac{k^{\parallel} - \widetilde{k}(z/z_0)}{k^{\parallel}\left(k^{\parallel} - \widetilde{k}\right)} \qquad G^{\perp} = -\left[\frac{k^{\perp} - \widetilde{k}(z/z_0)}{\tilde{e}k^{\perp}\left(k^{\perp} - \widetilde{k}\right)}\right]$$

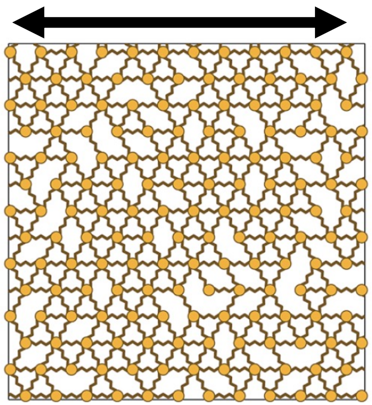
✓ Assume isotropy of green function $G^{\parallel} = G^{\perp}$

$$G_{\mathrm{eff}}(\omega\mu) = M_{\mathrm{eff}}^{-1} \Longleftrightarrow G^{\parallel} = G^{\perp} = \frac{2d}{z_0}\left[\frac{1}{k^{\parallel} - (d-1)\tilde{e}k^{\perp}}\right]$$

✓ We obtain closed equations for $k^{\parallel}, k^{\perp}, G^{\parallel}, G^{\perp}$

Complex modulus

✓ Macrorheology experiment

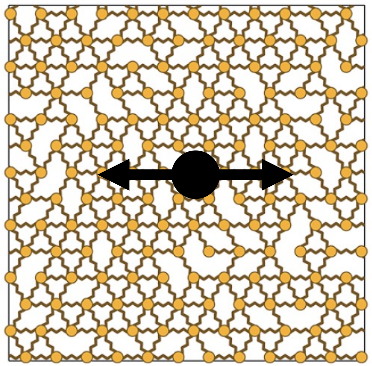


Measures global modulus when applying an oscillatory strain

$$G'_M - iG''_M = k_{\rm eff}$$

$$k_{\rm eff} = k^{\parallel} - (d-1)\tilde{e}k^{\perp}$$: Effective stiffness

✓ Microrheology experiment



Measures microscopic displacements of a probe particle when applying an oscillatory external force to it

$$g = \frac{1}{d}{\rm Tr}\langle i|G_{\rm eff}|i\rangle = \frac{2d}{z_0}\frac{1}{k_{\rm eff}}$$: Response function

→ Transform to complex modulus using generalized Stokes relation

$$G'_m - iG''_m = \frac{1}{3\pi\sigma_{pr}g} = \frac{1}{3\pi\sigma_{pr}}\frac{z_0}{2d}k_{\rm eff}$$

✓ Macrorheology and microrheology output same complex moduli

Set up

✓ EMT Equations

$$G^{\parallel} = \frac{k^{\parallel} - \widetilde{k}(z/z_0)}{k^{\parallel}\left(k^{\parallel} - \widetilde{k}\right)}$$

$$G^{\perp} = -\left[\frac{k^{\perp} - \widetilde{k}(z/z_0)}{\widetilde{e}k^{\perp}\left(k^{\perp} - \widetilde{k}\right)}\right]$$

$$G^{\parallel} = G^{\perp} = \frac{2d}{z_0}\left[\frac{1}{k^{\parallel} - (d-1)\widetilde{e}k^{\perp}}\right]$$

$$\widetilde{k} = 1 - i\omega\mu$$

$$\widetilde{e} = \frac{e + i\omega\mu}{1 - i\omega\mu}$$

$$k_{\text{eff}} = k^{\parallel} - (d-1)\widetilde{e}k^{\perp}$$

$$G'_M - iG''_M = k_{\text{eff}}$$

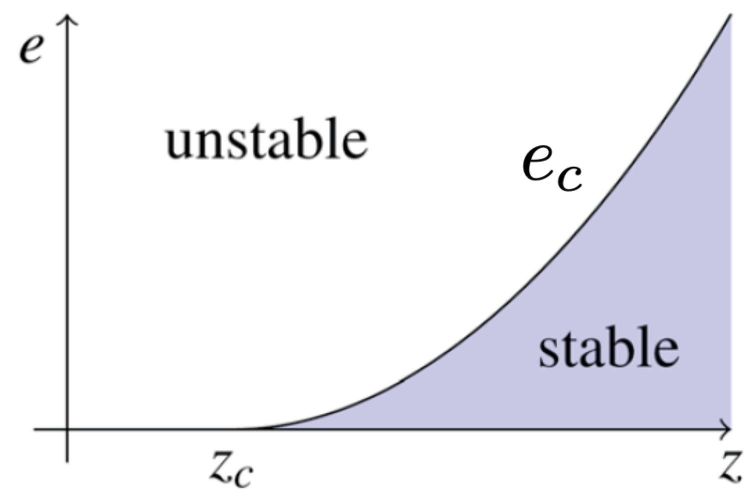
✓ Three dimensional space $d = 3$

✓ FCC lattice sites $z_0 = 12$

✓ Control parameters

$$\delta z = z - z_c \geq 0$$

$$0 \leq e \leq e_c \ (\propto \delta z^2)$$



DeGiuli et al., Soft Matter 2014

Results (1/5): Analytical solution

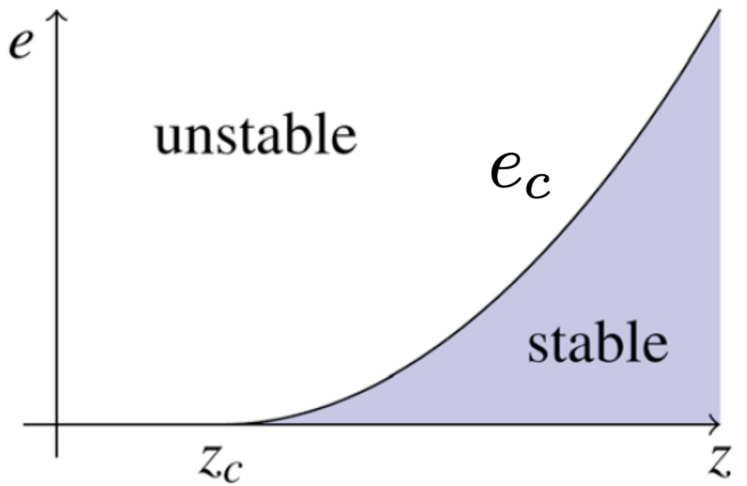
✓ Analytical solution

$$G'_M - iG''_M = \left(1 + \sqrt{\frac{e_c - e - i\omega\mu}{e_c}}\right)\frac{\delta z}{4da} + o\left(\delta z\right)$$

$$e_c = \left(\frac{1}{32d^2a}\frac{z_0}{2d}\right)\delta z^2 \sim \omega_*^2 \propto \delta z^2$$

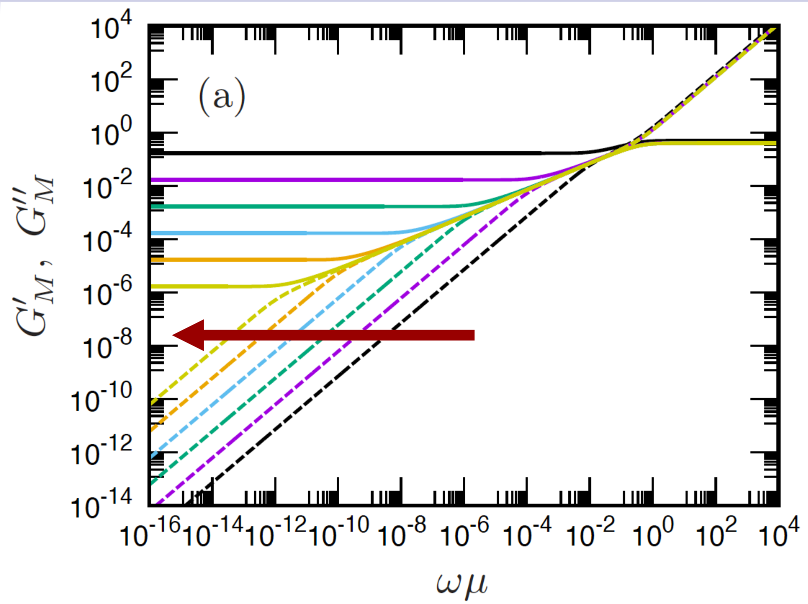
✓ Static modulus (zero frequency)

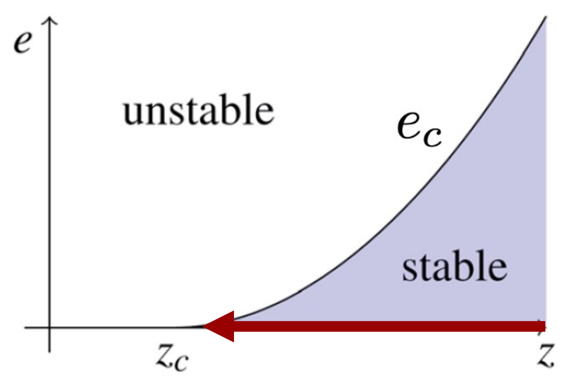
$$G'_{M0} = \left(1 + \sqrt{\frac{e_c - e}{e_c}}\right)\frac{\delta z}{4da} \propto \delta z$$

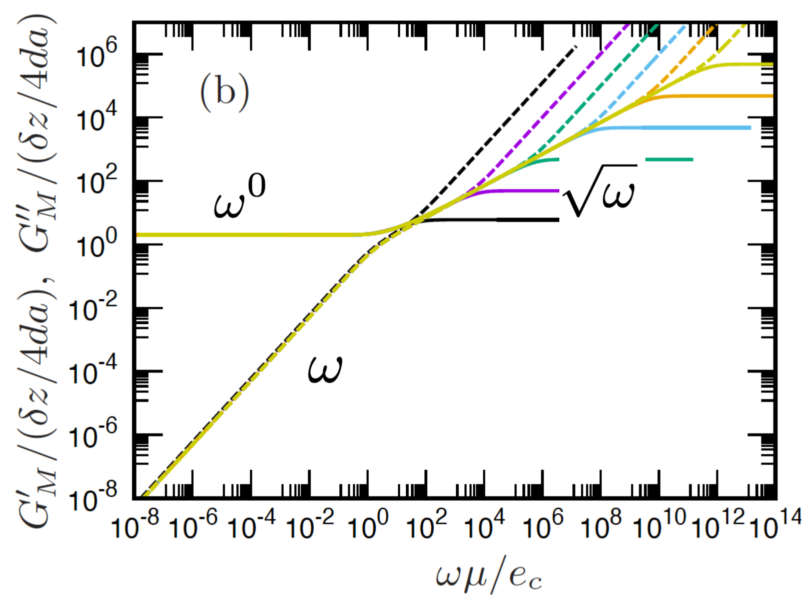


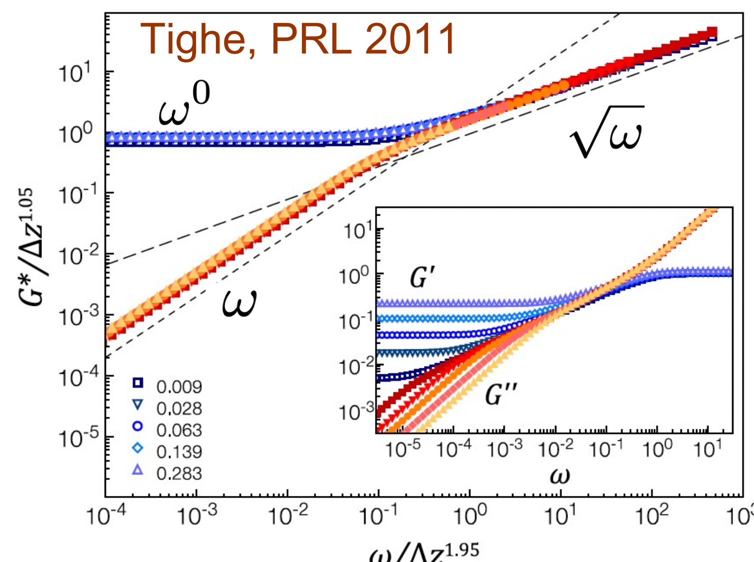
✓ Static modulus becomes real number in a stable region at $e \leq e_c$

Results (2/5): Zero prestress case $e = 0$



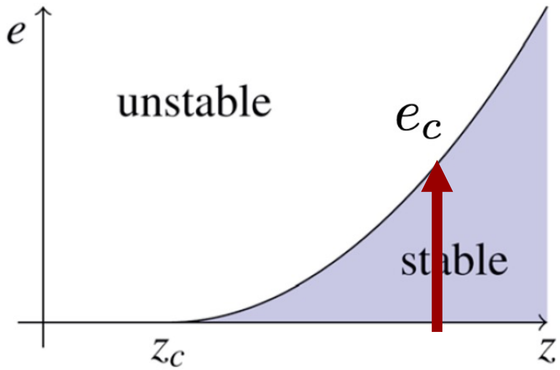


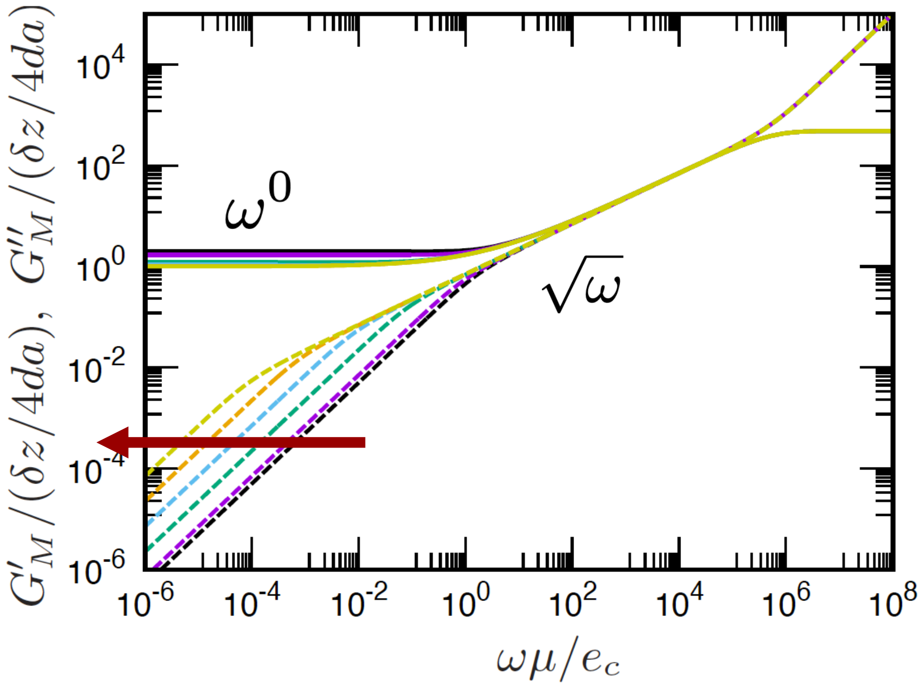


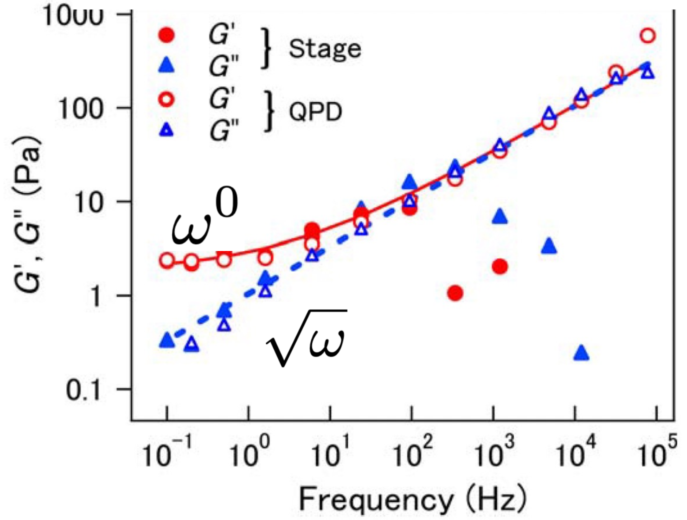


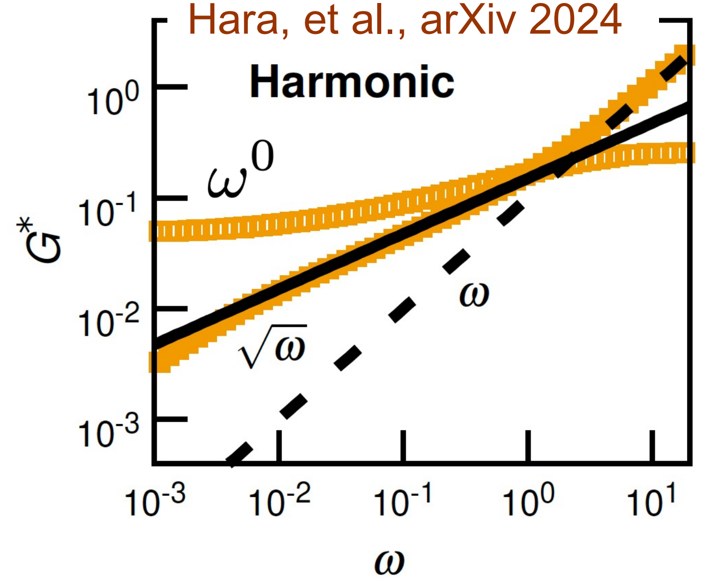
✓ EMT explains $G' \approx G'' \propto \sqrt{\omega}$

Results (3/5): Finite prestress $0 < e \le e_c$





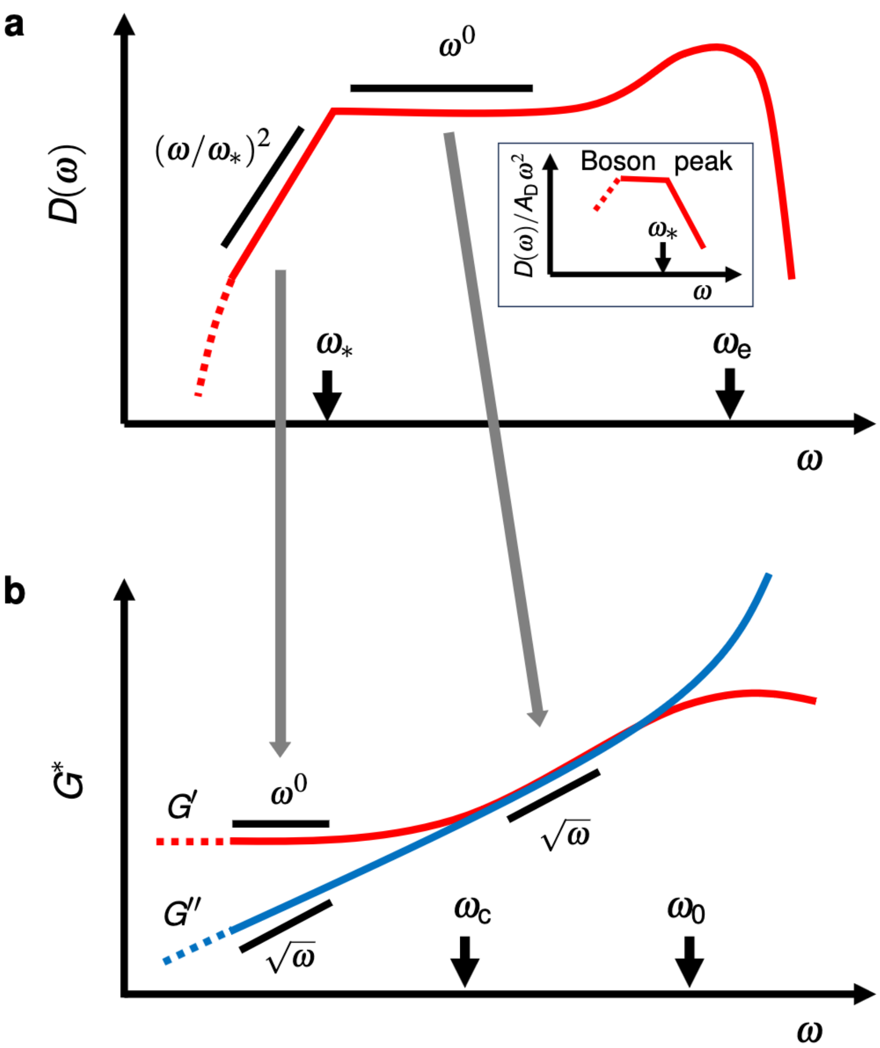


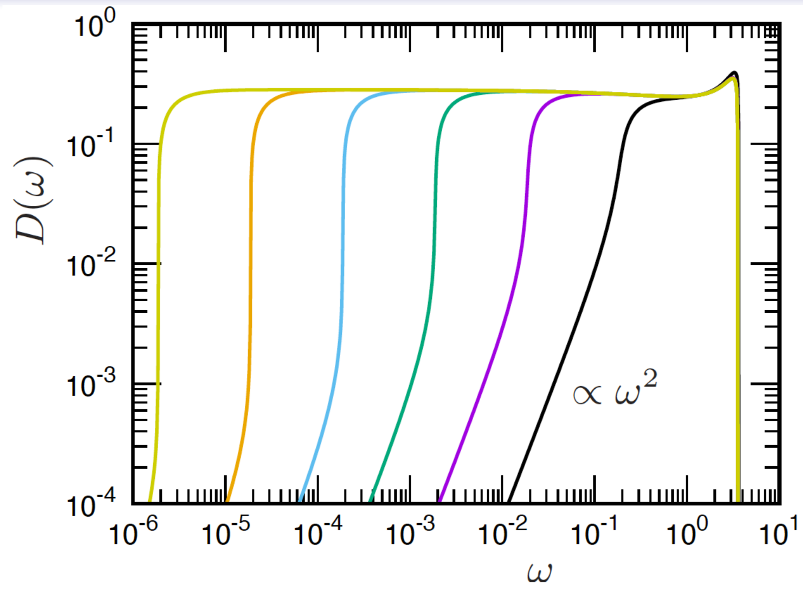


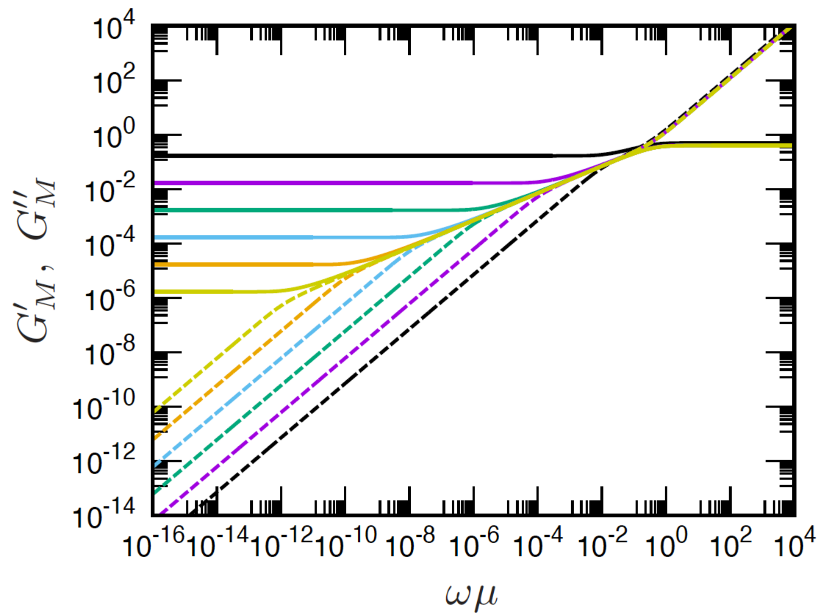
✓ EMT explains anomalous viscous loss

Results (4/5): vDOS and complex modulus at $e = 0$

Hara, et al., arXiv 2024

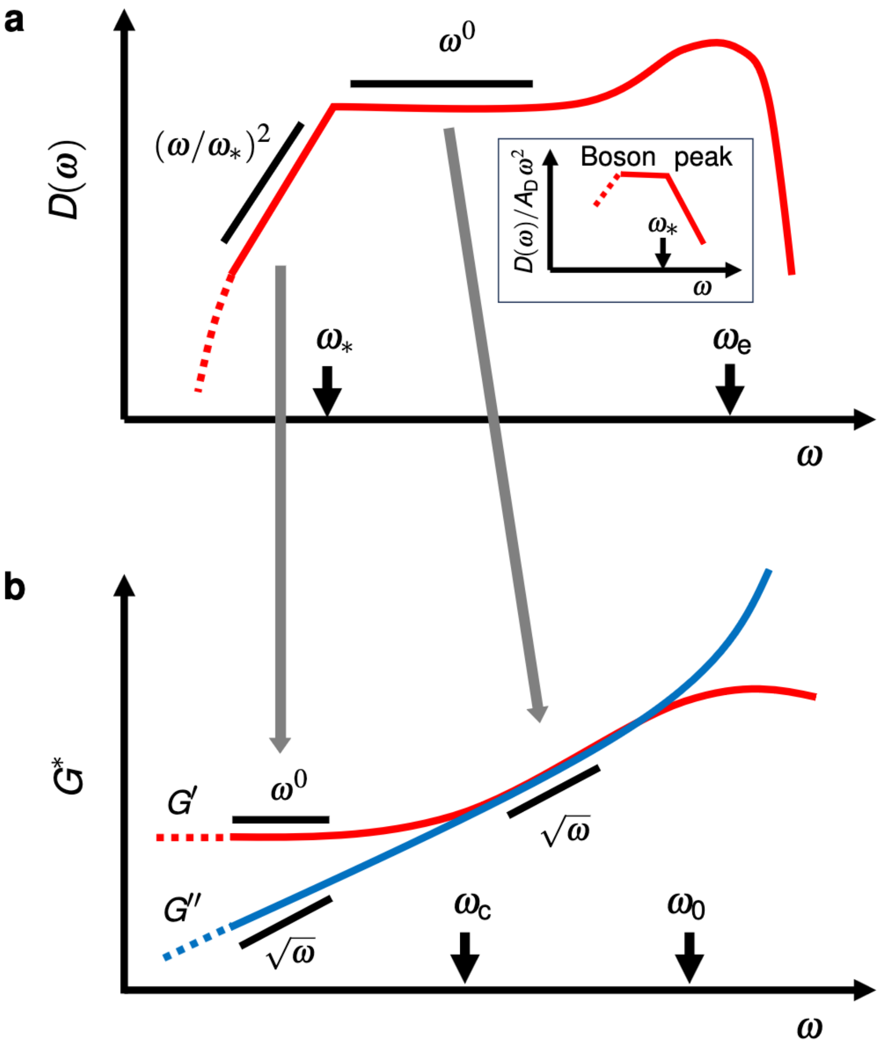


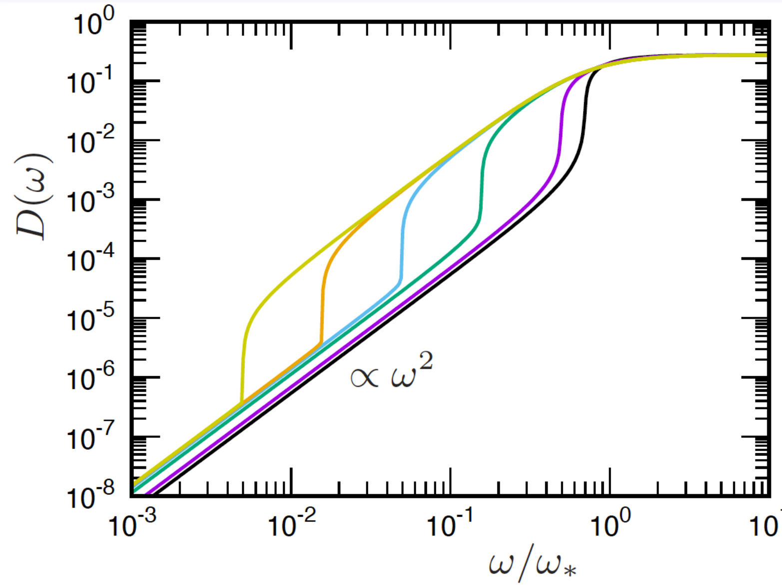


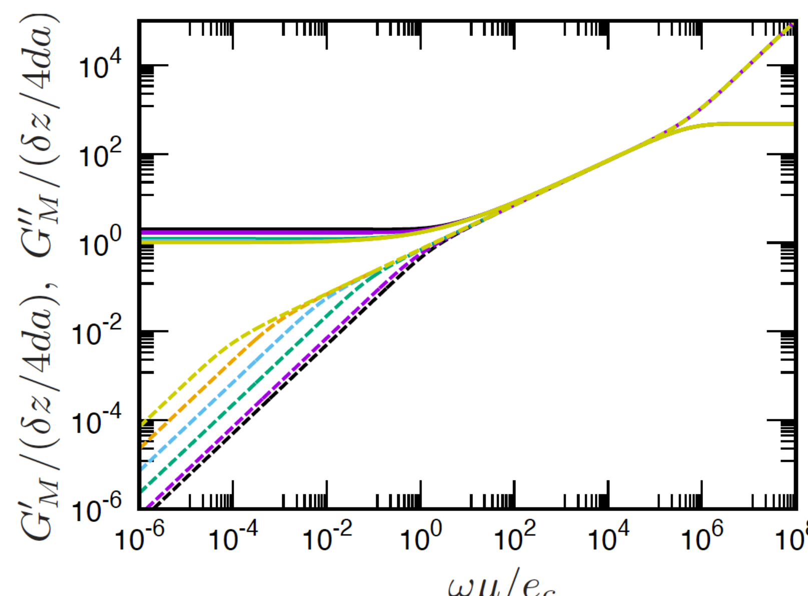


Results (5/5): vDOS and complex modulus at $0 < e \leq e_c$

Hara, et al., arXiv 2024

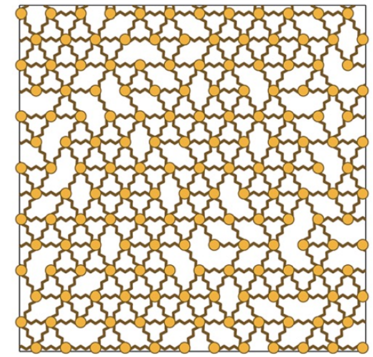






Conclusions

Mizuno and Ikeda, arXiv:2407.15323



- ✓ Develop EMT for viscoelasticity of soft jammed solids
- ✓ Random spring network model
- ✓ Integrate contact damping effect
- ✓ Theory explains experimentally observed viscoelasticity
  - Macrorheology and microrheology show the same complex moduli
  - higher frequencies $G' \approx G'' \propto \sqrt{\omega}$
    - ➢ Linked to plateau in vDOS
    - ➢ Controlled by contact number δz
  - Lower frequencies $G' \propto \omega^0 \quad G'' \propto \sqrt{\omega}$

    Anomalous viscous loss
    - ➢ Linked to non-Debye law (boson peak) in vDOS
    - ➢ Controlled by prestress e