Jamming Transition in Amorphous Solid Composites

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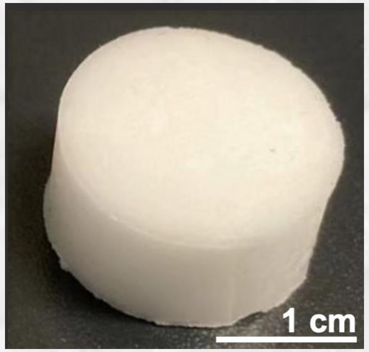

In collaboration with

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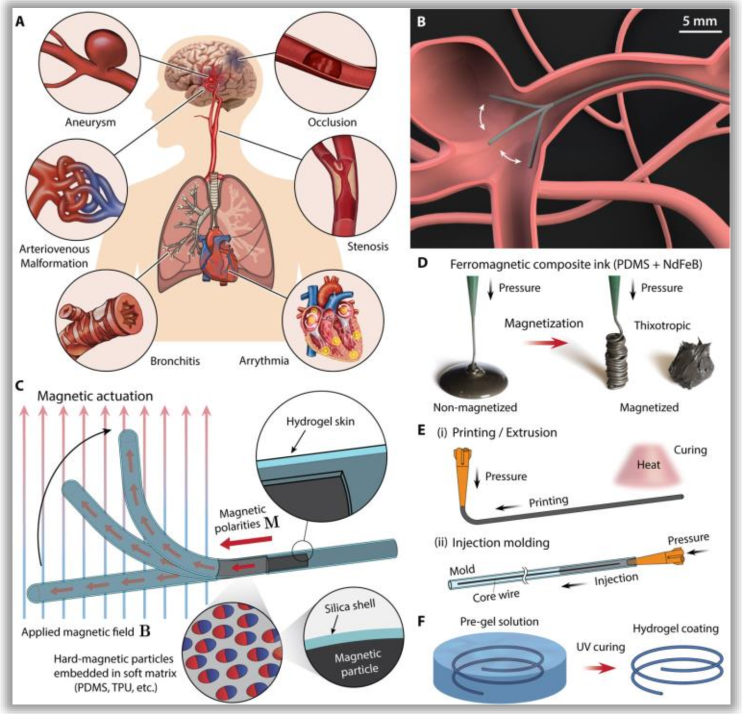
ASPIRE League

Video Source: Behringer Lab @ Duke

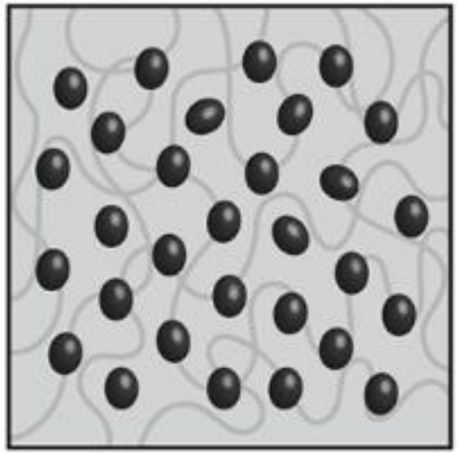
Amorphous Solid Composites = non-Brownian stiff (micron-sized) grains (randomly) embedded in a soft gel matrix

Field-Responsive and Adaptive Soft Robots

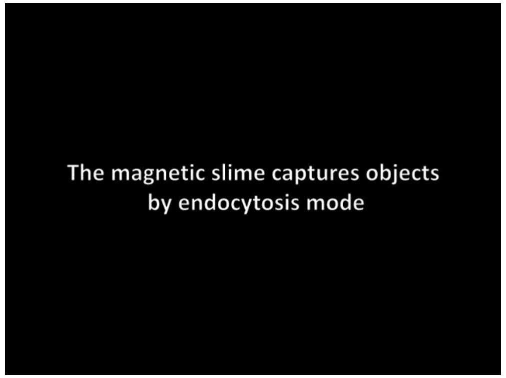
Magnetic Particles in Soft Polymer Matrix



Kim, et al. Science Robotics (2020)

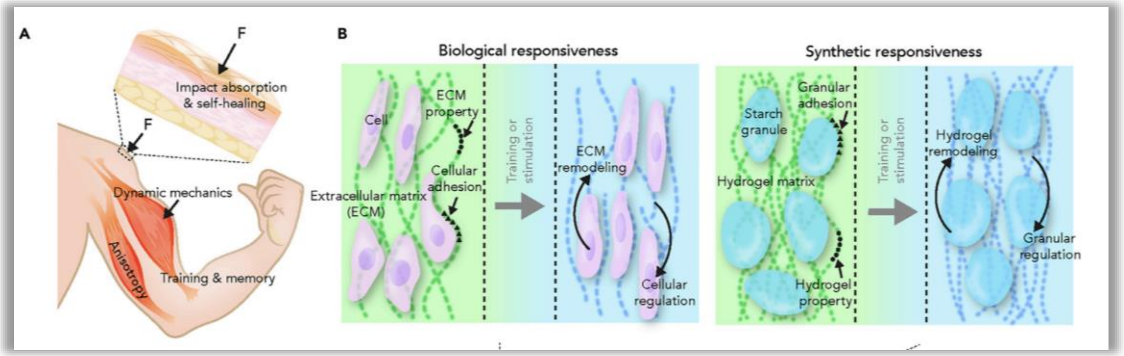


Kim and Zhao, Chem. Rev. (2022)

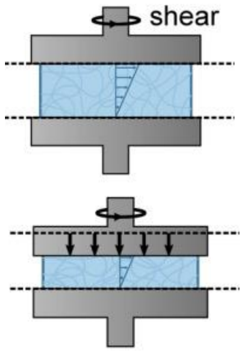


Sun, et al. Adv. Funct. Mater. (2022)

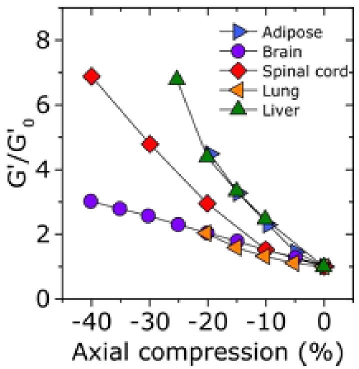
Tissue-like bio-mimetic materials



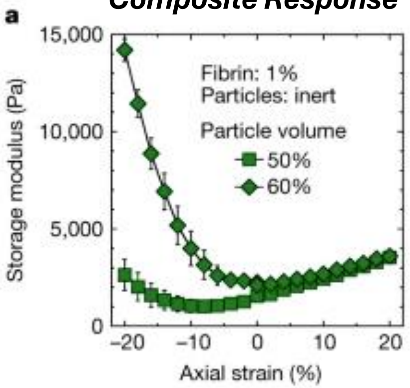
Fang, et al., Matter (2020)



Tissue response



Composite Response



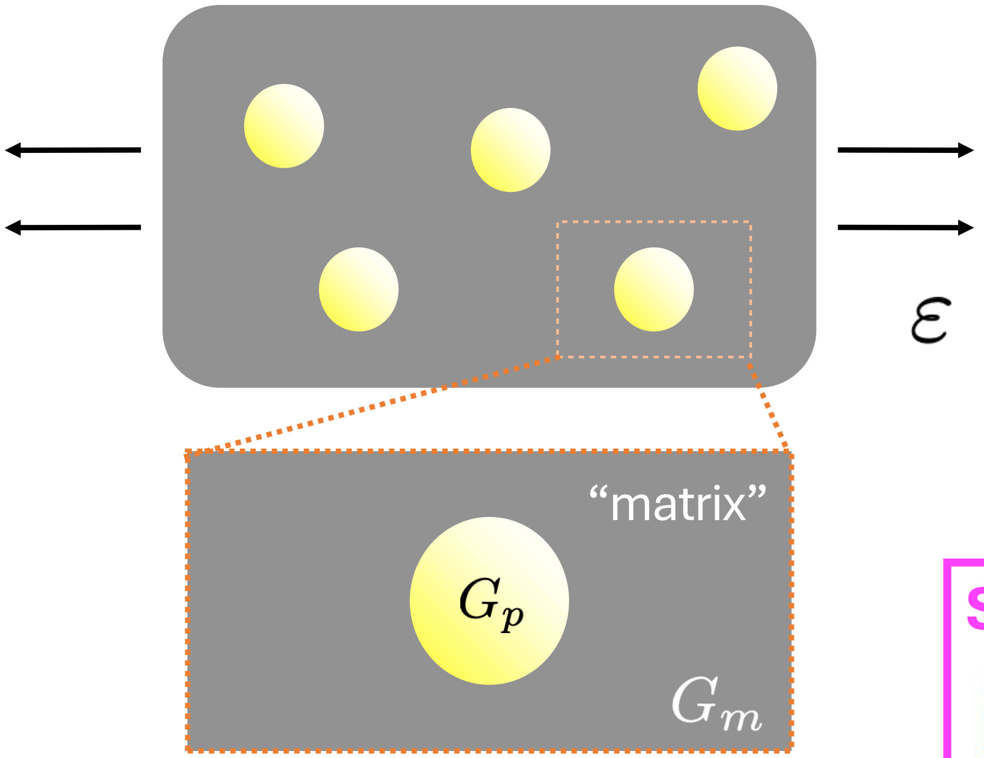
van Oosten et al., Nature 573, 96-101 (2019)
Song et al., J. Appl. Phys. 129, 140901 (2021)
Song et al., arXiv:2307.11687

Adaptive **→ large deformation (soft)**
Responsive **→ more embedded particles (dense)**

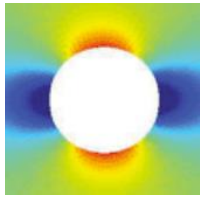
Nonlinear Mechanical Responses

Modelling composites in the dense and soft limits is challenging.

Dilute Composites

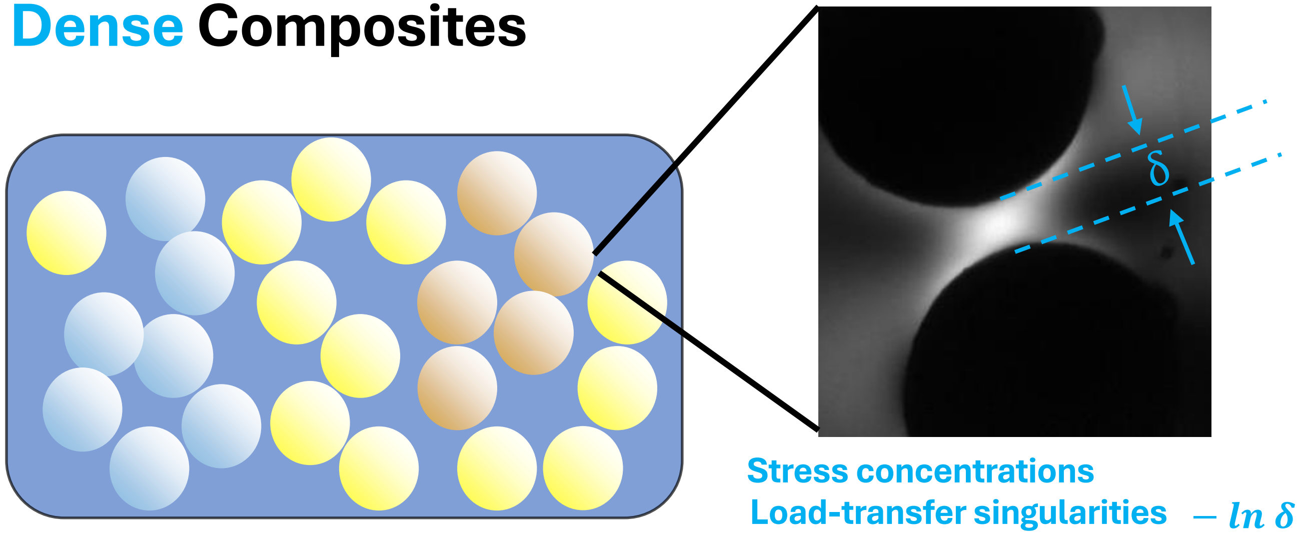


Isolated inclusions



Einstein-Smallwood (1944)
Eshelby (1957)

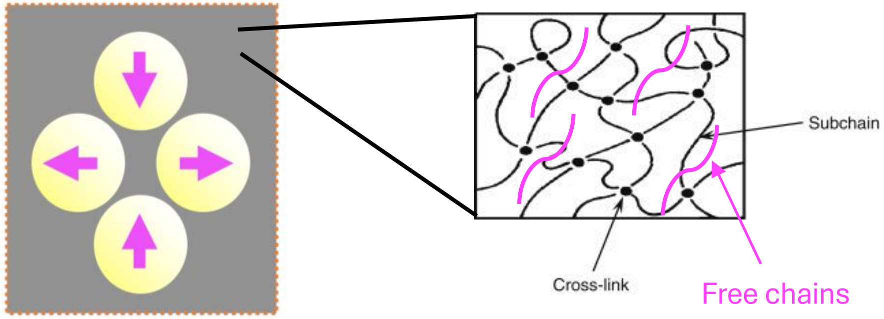
Dense Composites



Stress concentrations
Load-transfer singularities $-\ln\delta$

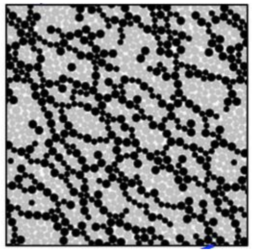
Phan-Thien et al. (1994)

Soft composites

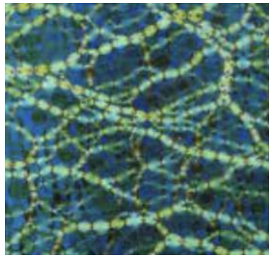


None affine shear transformations

Multi-phase nature of the matrix



Seto, et al. Granular Matter (2019)



Bi et al., Nature (2011)

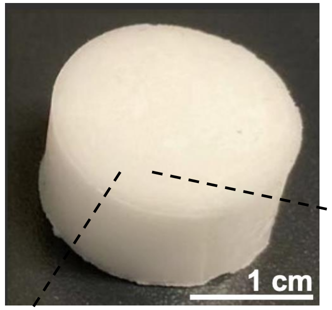
Soft and dense limit:
The role of jamming?

Key Question

What governs the mechanics of soft composites in the dense limit?

Does granular jamming matter at all?

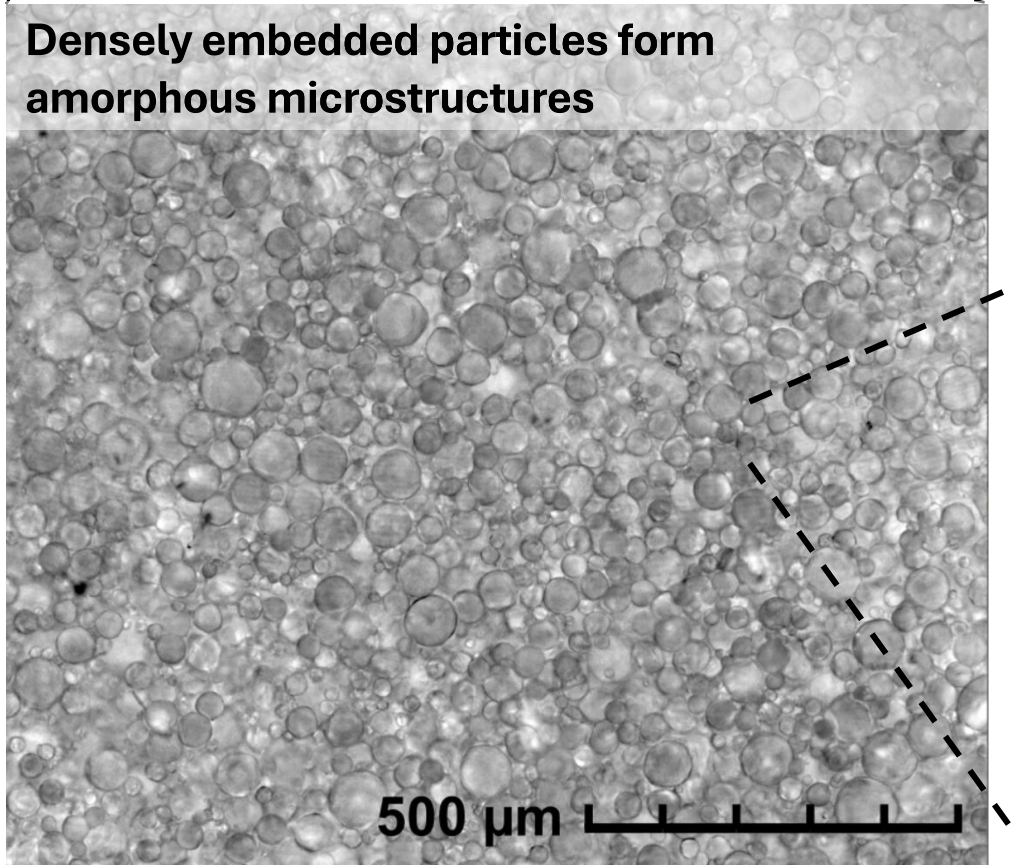
Model soft composite : *athermal* stiff PS micro-spheres in soft PDMS matrix

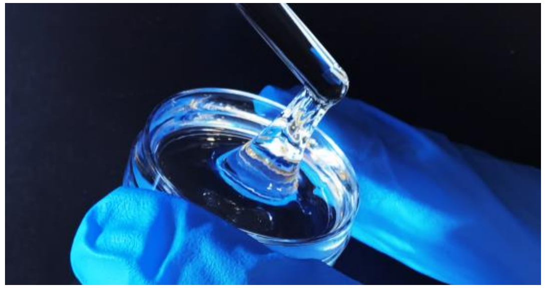


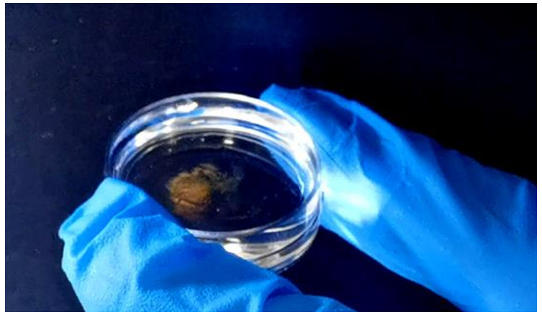
Composite Sample (ϕ, G_m)

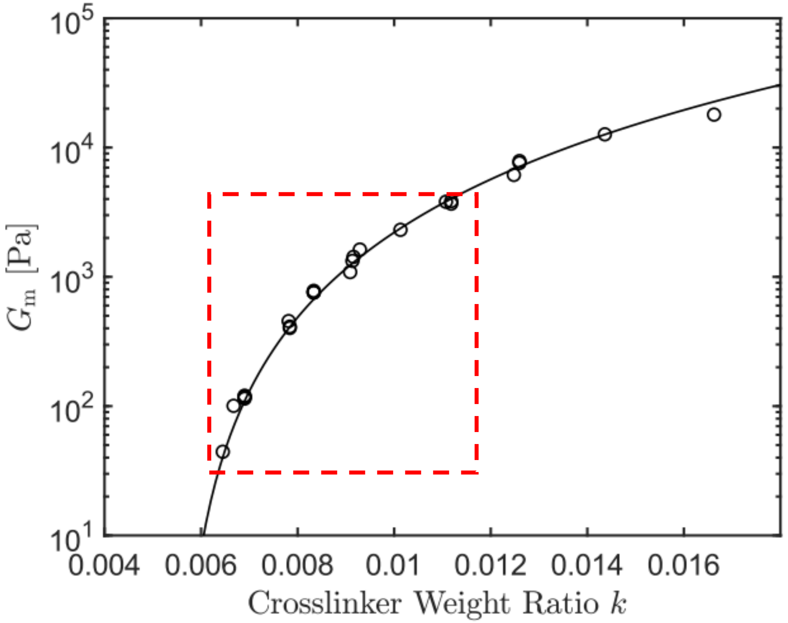
Particle volume fraction
(0.40 to 0.67)

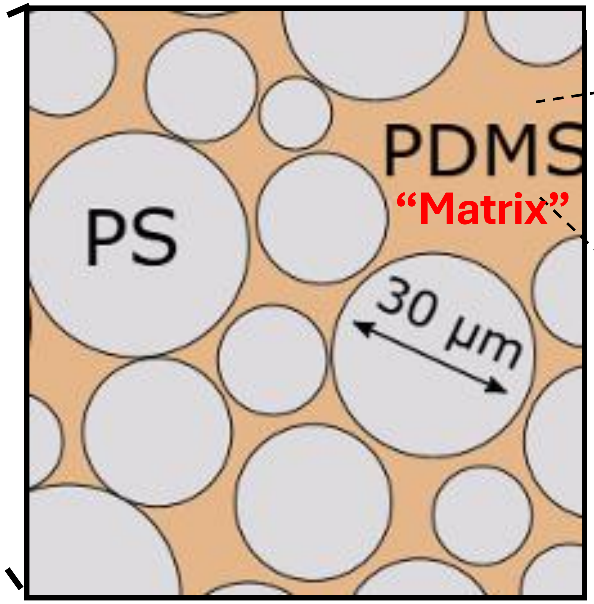
Matrix shear modulus
(0.04 to 4 kPa)

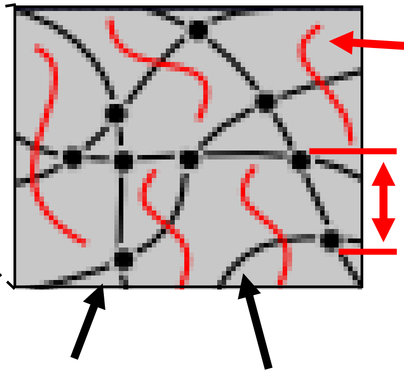












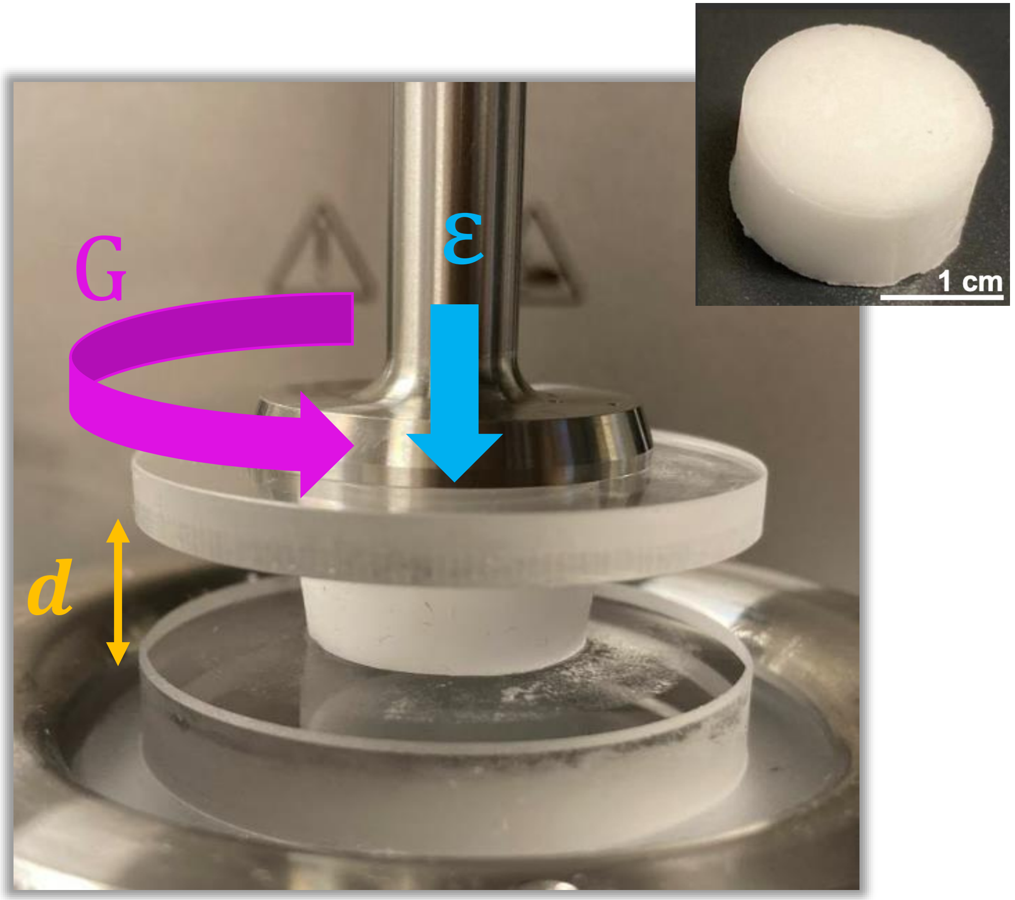
""free chains"

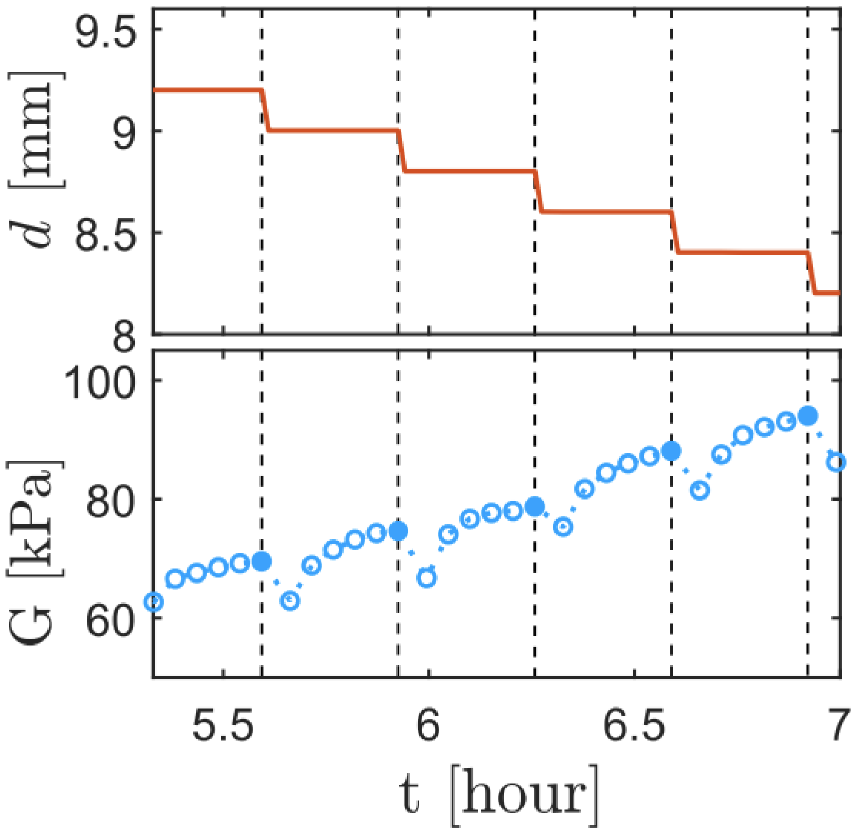
mesh size
~ 10 nm
<< particle size

Crosslink
molecules

crosslinked polymer
network

Experiment: measuring shear modulus (G) under axial pre-strain (ε)



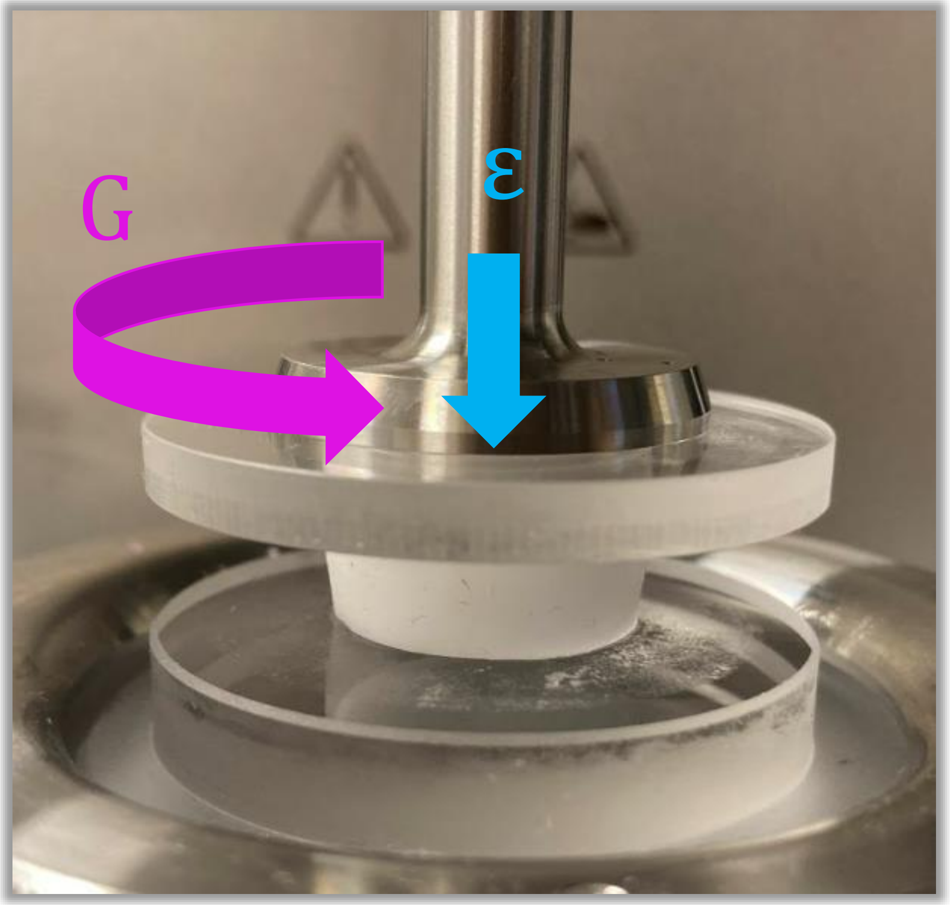


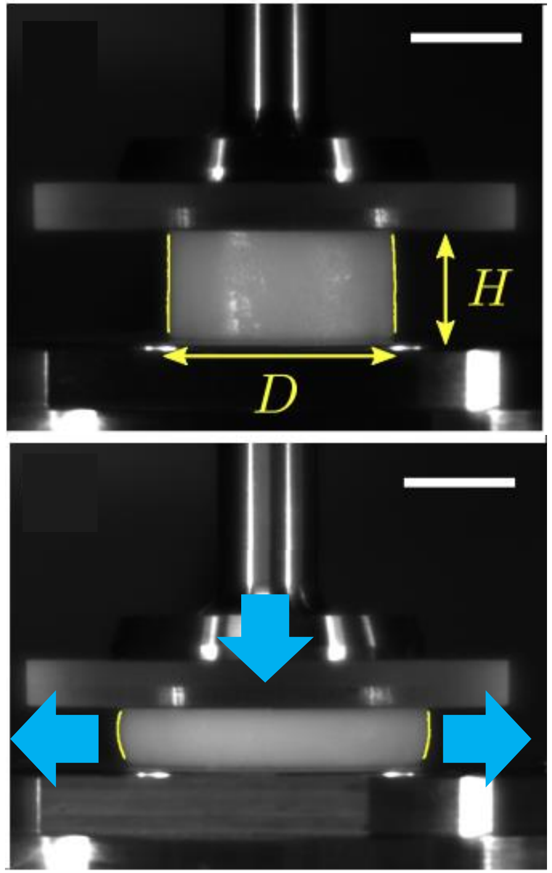
[1] **Axial compressive strain *ε*** applied step-by-step and quasi-statically (which may change particle network)

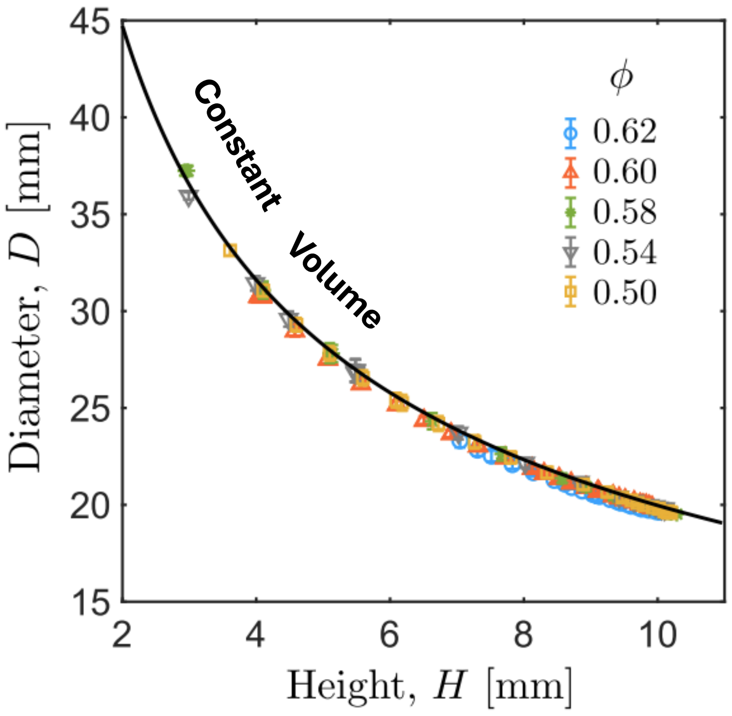
[2] After each compression step, measuring the **small-amplitude shear modulus G** (hopefully do not change network)

[= storage modulus G' under $\omega = 0.1$ rad/s and $\delta\gamma = 0.01\%$]

Experiment: measuring shear modulus (G) under axial pre-strain (ε)



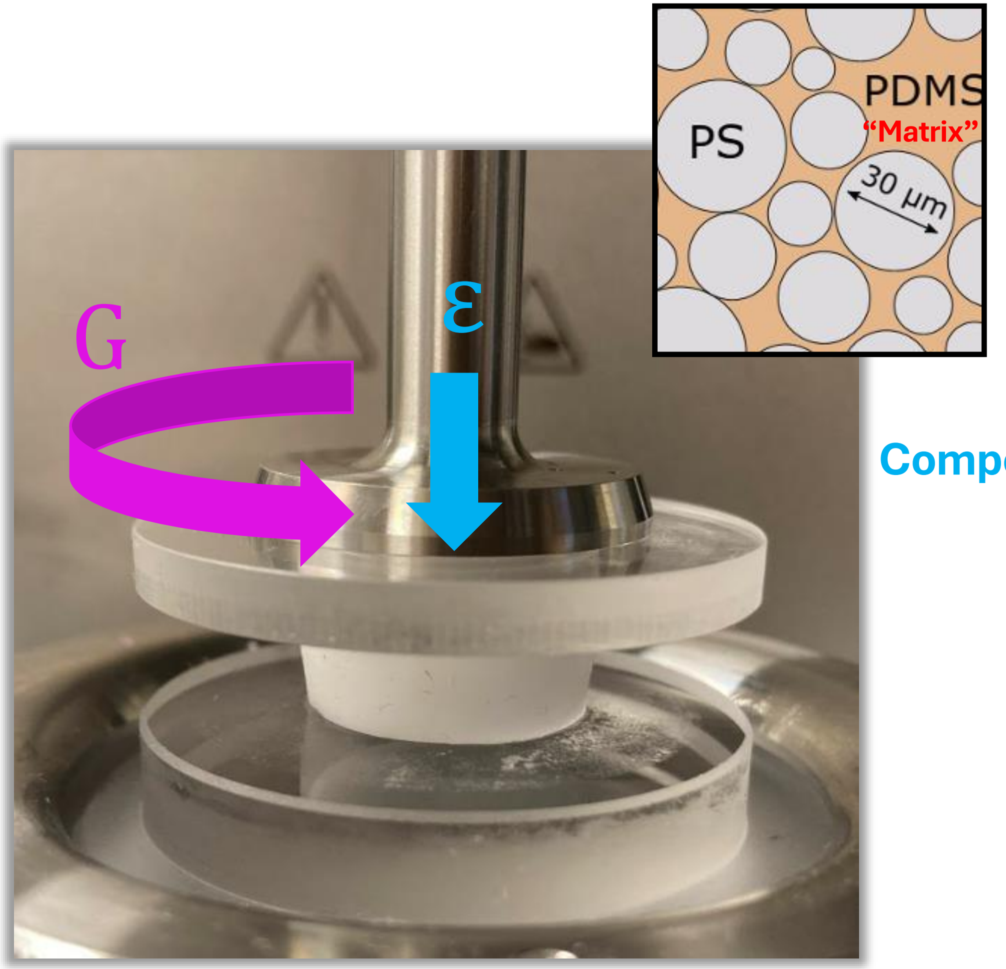


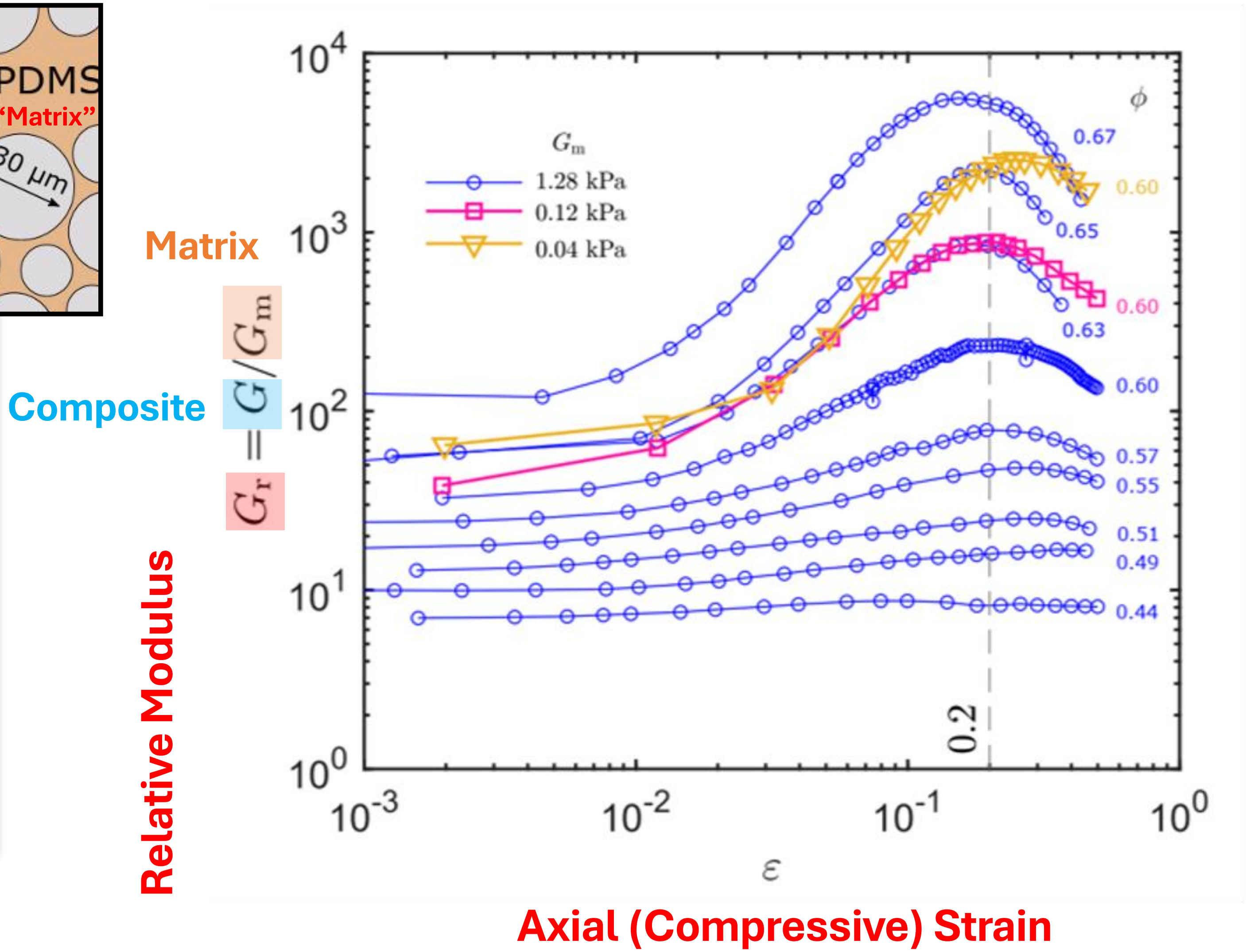


The axial strain ε induces a **pure shear** deformation that **preserves the volume of the sample** (and thus **preserves the volume fraction ϕ of the particles**).

This point will be important later when we consider shear jamming…

Experiment: measuring shear modulus (G) under axial pre-strain (ε)





- ❑ Strain stiffening is stronger for denser and softer samples.

What governs the mechanics of soft composites in the dense limit?

Composite Elasticity

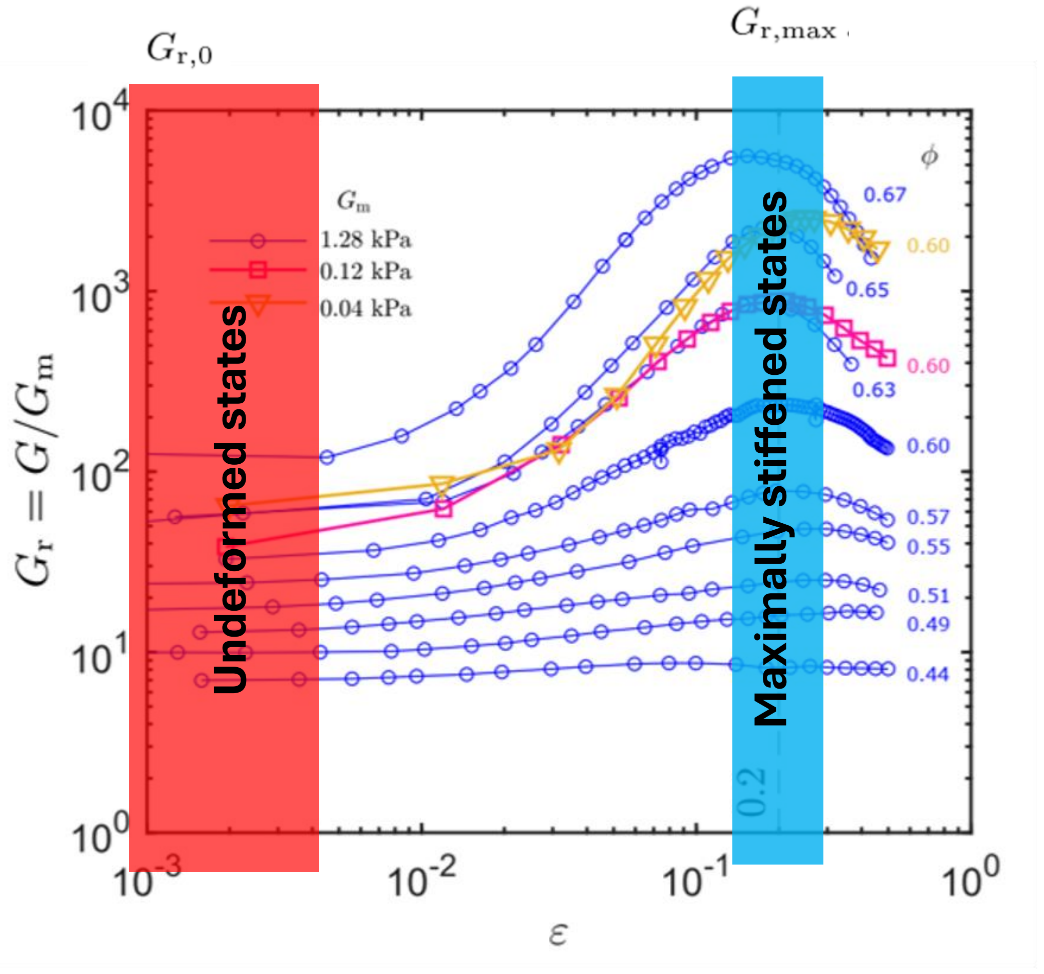
$$G = G(\phi, G_m, \varepsilon)$$

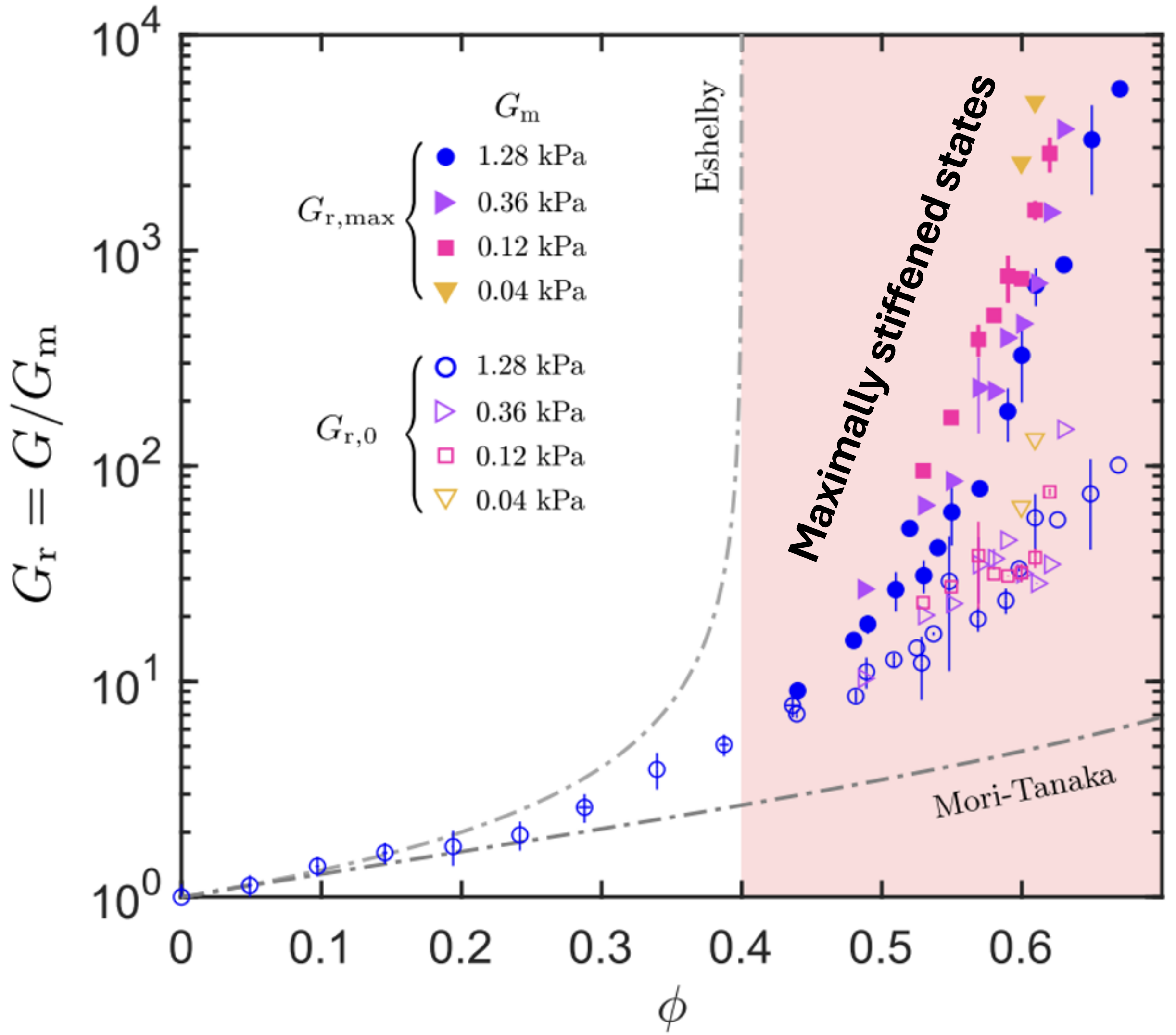
Particle Volume Fraction (ϕ)

Matrix Elasticity (G_m)

Shear Deformation (ε)

The ϕ dependence of the limiting states deviates from classical composite model predictions



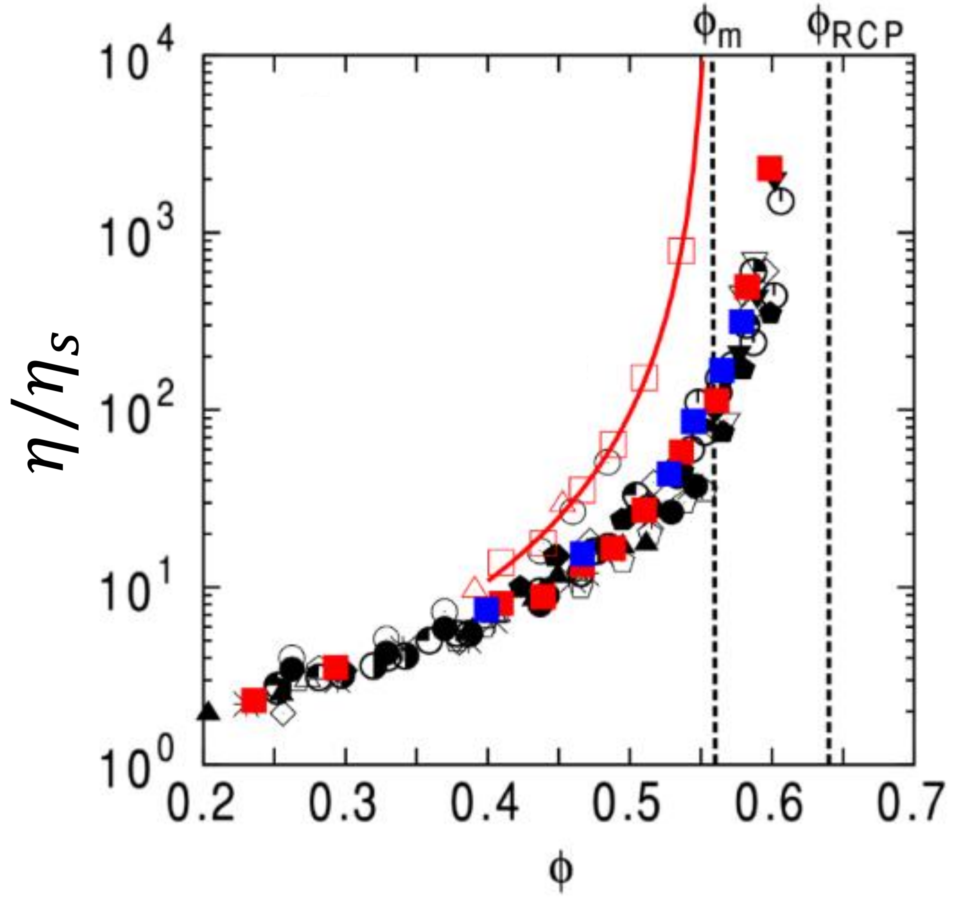


- ❑ Classical composite models do not capture the stiffening regime.

The ϕ dependence of the limiting states appear similar to the jamming-controlled rheology in suspensions

Shear-Thickening Suspensions

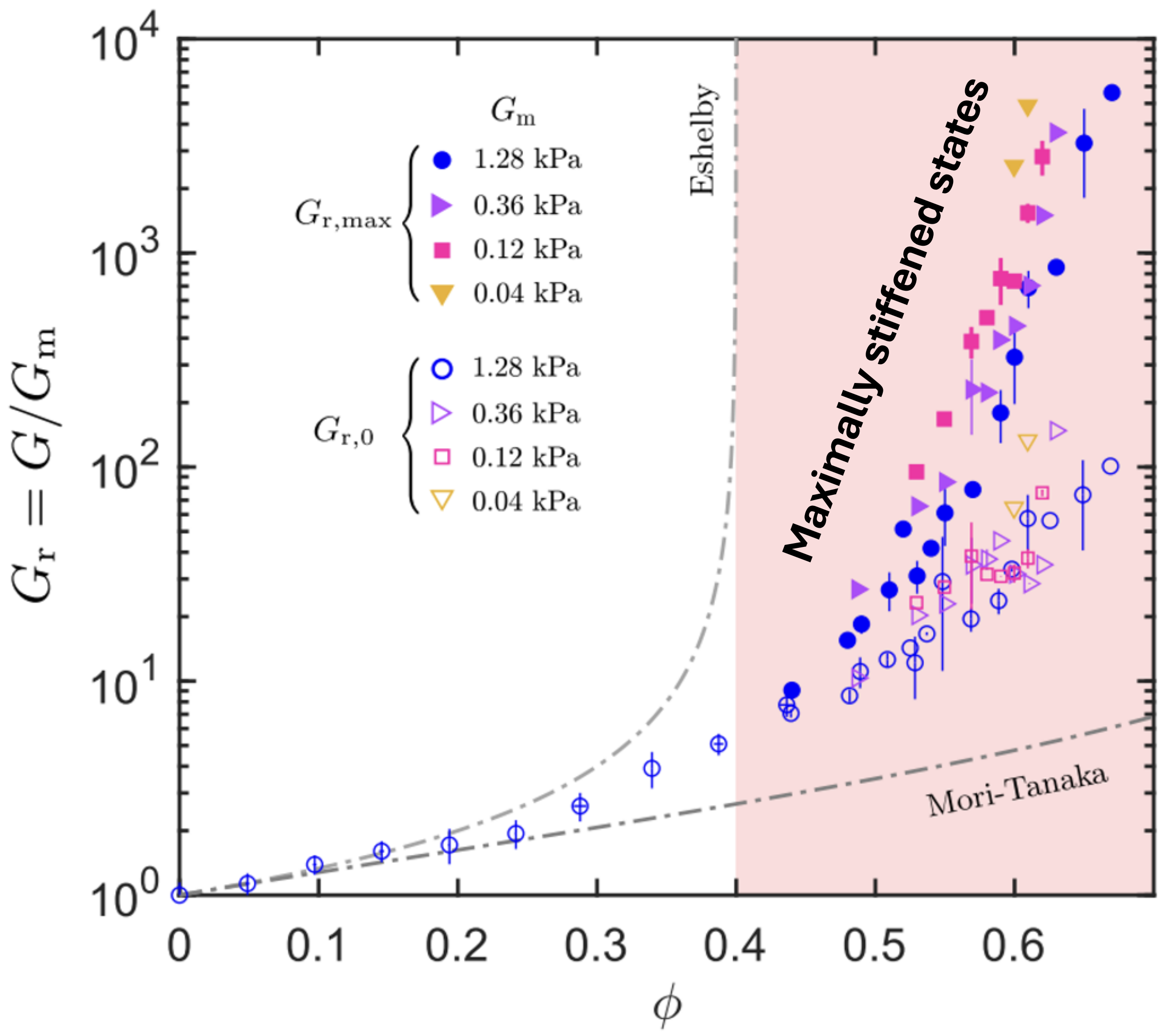
(Particles in viscous liquid)



Guy, Hermes, and Poon, Phys. Rev. Lett. 115, 088304 (2015)

Shear-Stiffening Composites (Our work)

(Particles in elastic solid)

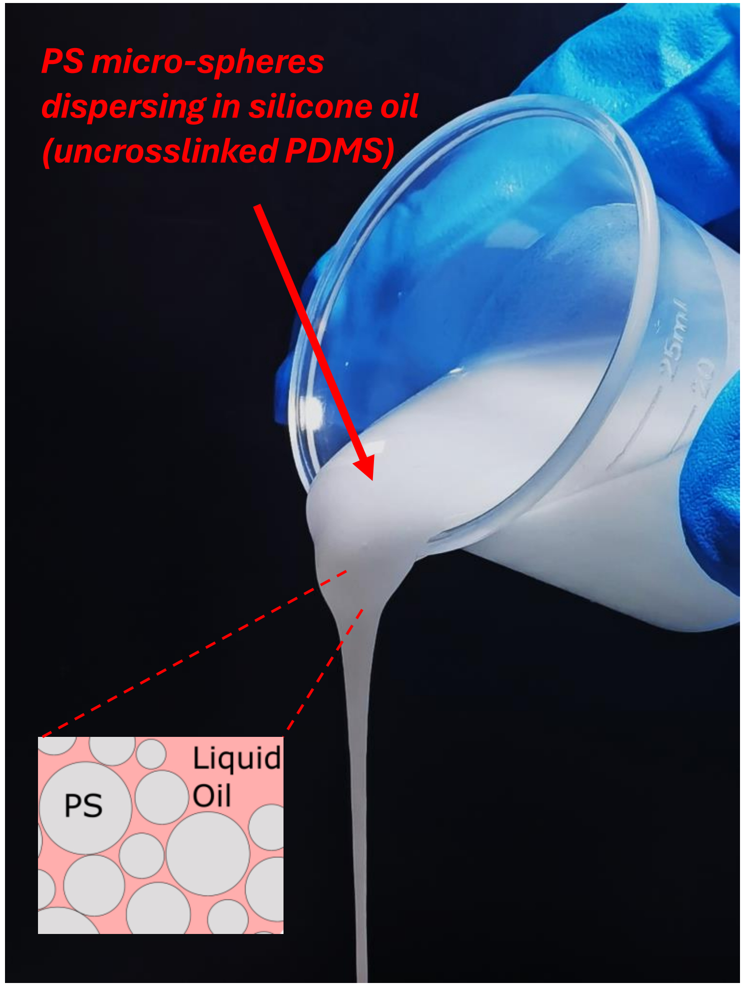


Does the jamming points also control the composite mechanics?

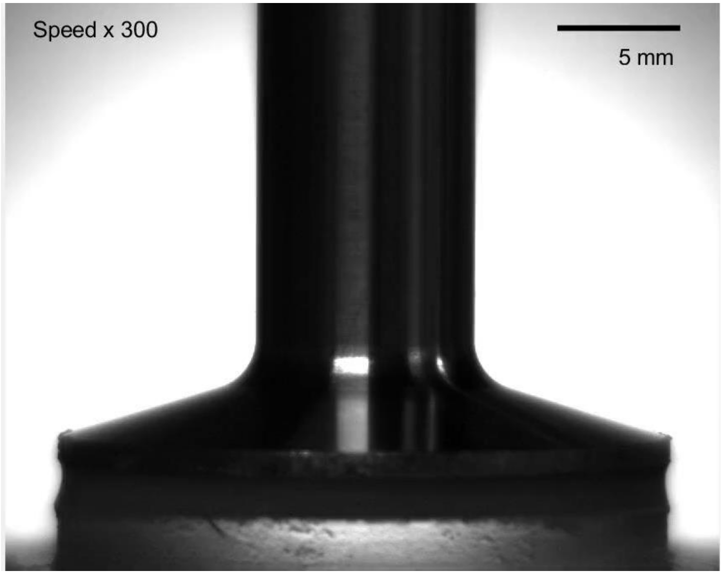
Signatures of jamming transition in the "precursor" suspension

*Experiments: PS-in-oil suspension rheology (**NO elastic matrix** here, particles are dispersing in a liquid)*

same polymer molecules as composite matrix, just not crosslinked

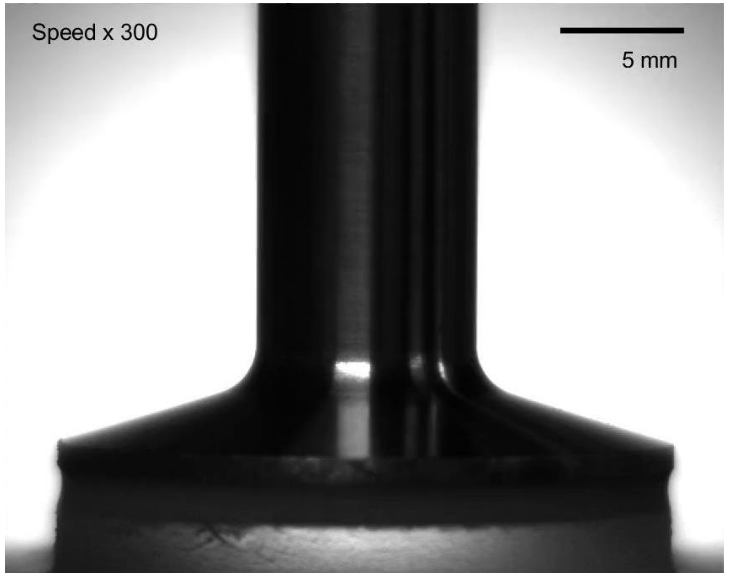


Liquid-like pinch-off



$\phi = 0.57$

Solid-like fracturing



$\phi = 0.61$

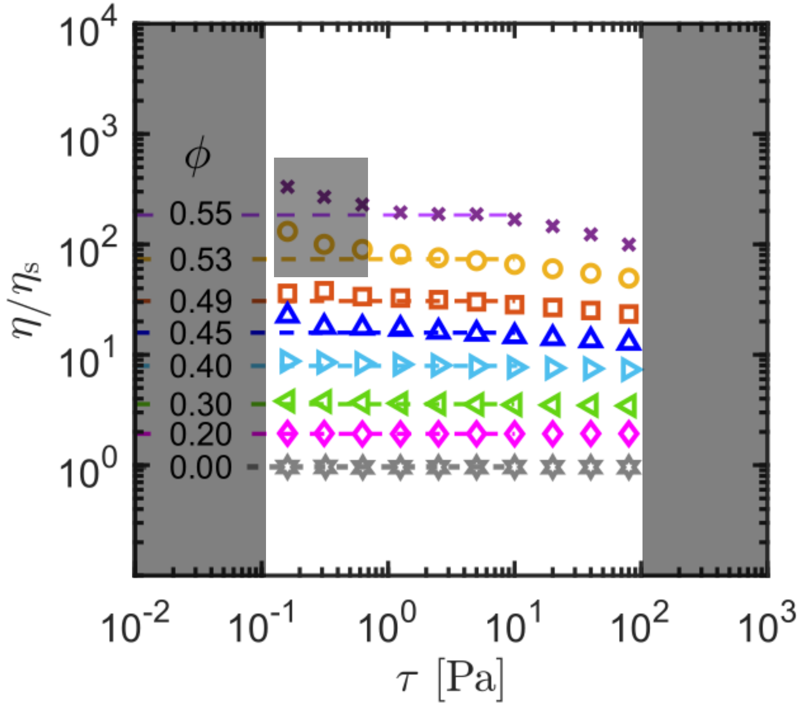
ϕ

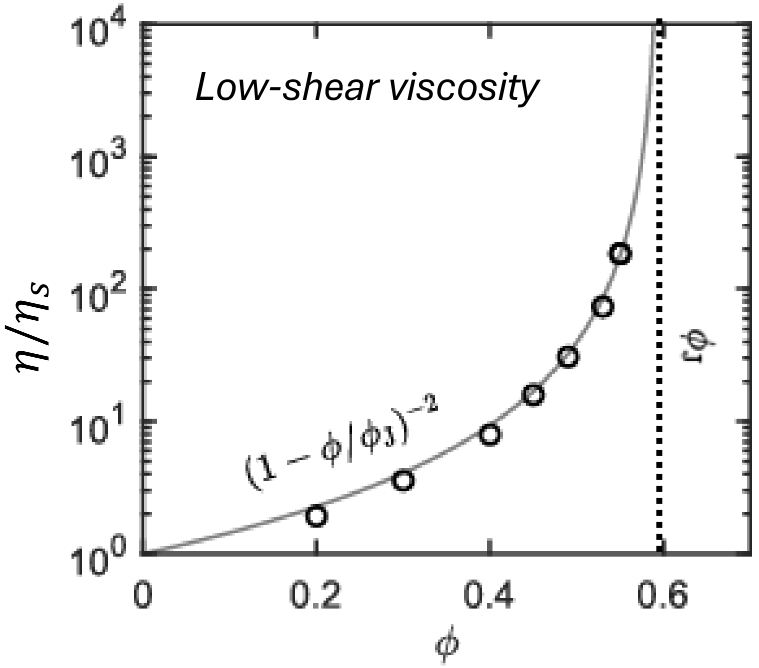
$\phi_J \approx 0.59$

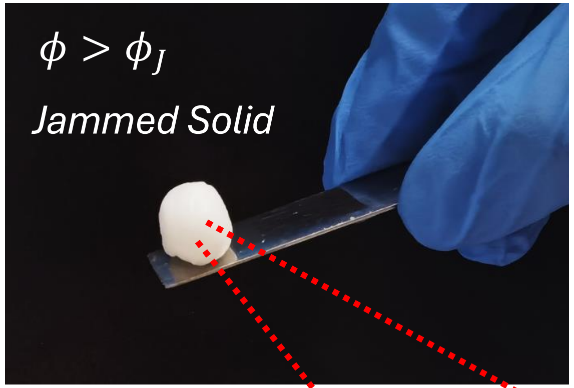
Signatures of jamming transition in the "precursor" suspension

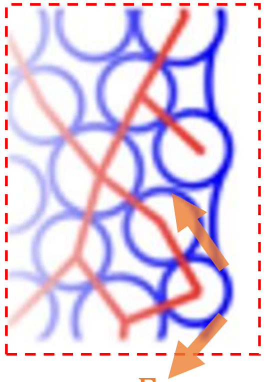
*Experiments: PS-in-oil suspension rheology (**NO elastic matrix** here, particles are dispersing in a liquid)*

same polymer molecule as composite matrix, just not crosslinked









$\phi_J \Rightarrow$ ***Jamming** Transition of the **steadily sheared states***

Γ

Liquid-air interface tension

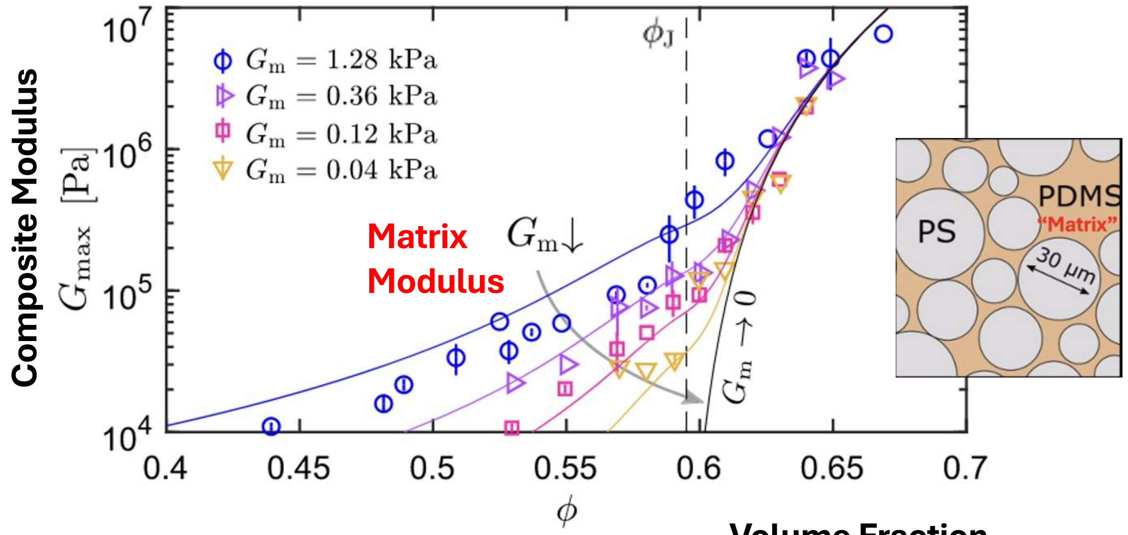
Confining pressure up to $\sim \frac{\Gamma}{D} \sim 1\ kPa$

How does this ϕ_J may affect composites?

- ❑ *the maximally-stiffened states*
- ❑ *they are sheared states and presumably share similar packing structures*
- ❑ *thus may be controlled by the same ϕ_J .*

How does ϕ_J affect composite elasticity?

This work: Composites (MSS) with non-zero matrix modulus G_m



Volume Fraction

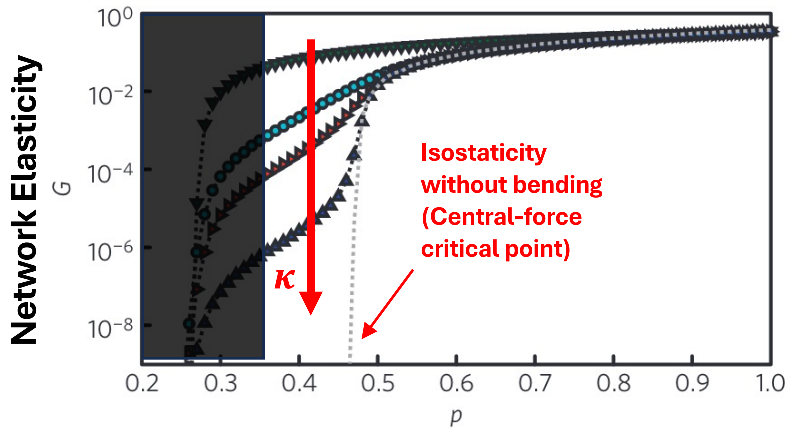
Particle network connectivity

Scaling ansatz

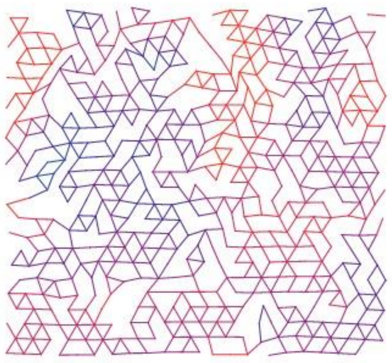
$$G_{max} = |1-\phi/\phi_J|^{\beta} f_{\pm}\left(\frac{G_m}{|1-\phi/\phi_J|^{\Delta}}\right)$$

Is this true for our composites?

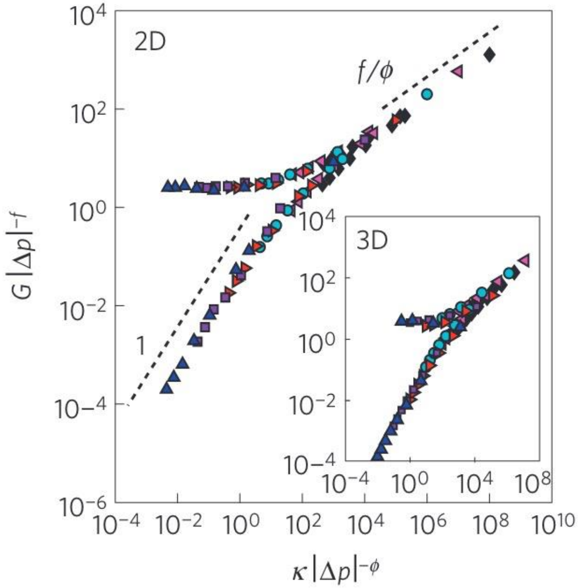
Motivation: Elastic network with non-zero bending rigidity κ



Network Connectivity



Broedersz et al., Nat. Phys. 7, 983-988 (2011)



Scaling ansatz for the bending-induced crossover

$$G \sim |1-p/p_c|^{f} f_{\pm}\left(\frac{\kappa}{|1-p/p_c|^{\phi}}\right)$$

Order parameter

Distance to transition

Field-like variable

Ising model

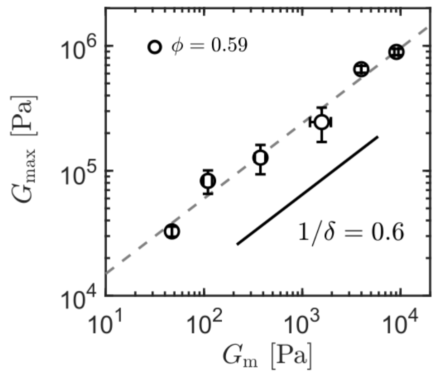
$$M \sim |1-T/T_c|^{\beta} f_{\pm}\left(\frac{H}{|1-T/T_c|^{\Delta}}\right)$$

Scaling collapse for the maximally stiffened states of composites

Scaling ansatz

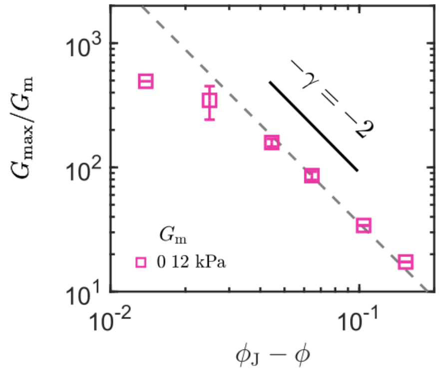
$$G_{max} = |1-\phi/\phi_J|^{\beta} f_{\pm}\left(\frac{G_m}{|1-\phi/\phi_J|^{\Delta}}\right)$$

$\phi = \phi_J$



$$G_{max} \sim G_m^{1/\delta}$$

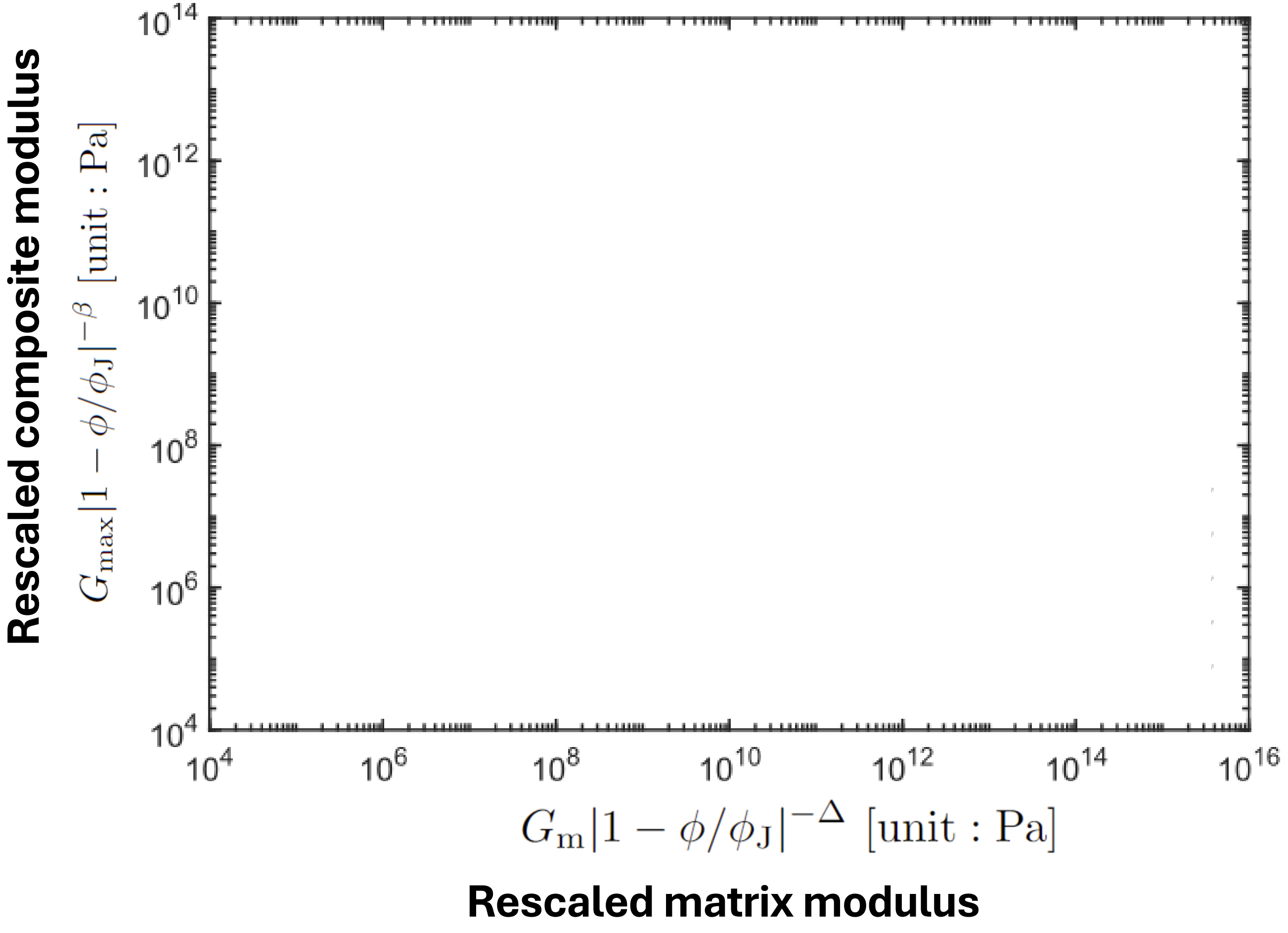
$\phi < \phi_J, G_m \approx 0$



$$\frac{G_{max}}{G_m} \sim (1-\phi/\phi_J)^{-\gamma}$$

- ❑ The exponents are assumed to obey the rules expected for ordinary critical points

$$\beta = \gamma/(\delta-1) = 3 \qquad \Delta = \delta\beta = 5$$



- ❑ **Jamming point controls composite elasticity in a way that resembles critical phenomenon.**

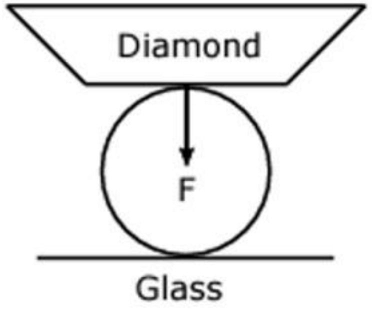
Scaling collapse for the maximally stiffened states of composites

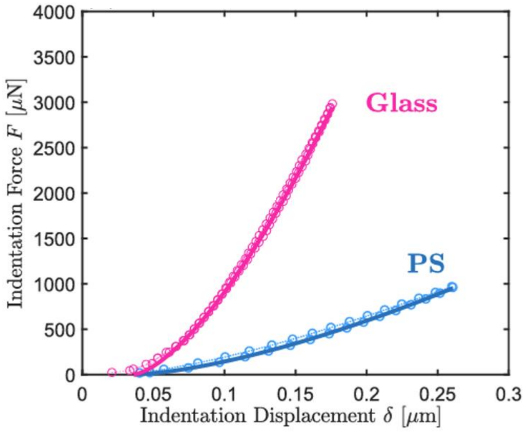
Scaling ansatz

$$G_{max} = |1 - \phi/\phi_J|^{\beta} f_{\pm}\left(\frac{G_m}{|1 - \phi/\phi_J|^{\Delta}}\right)$$

$$\beta = \gamma/(\delta - 1) = 3 \qquad \Delta = \delta\beta = 5$$

Nano-Indentation For Particle Material Stiffness

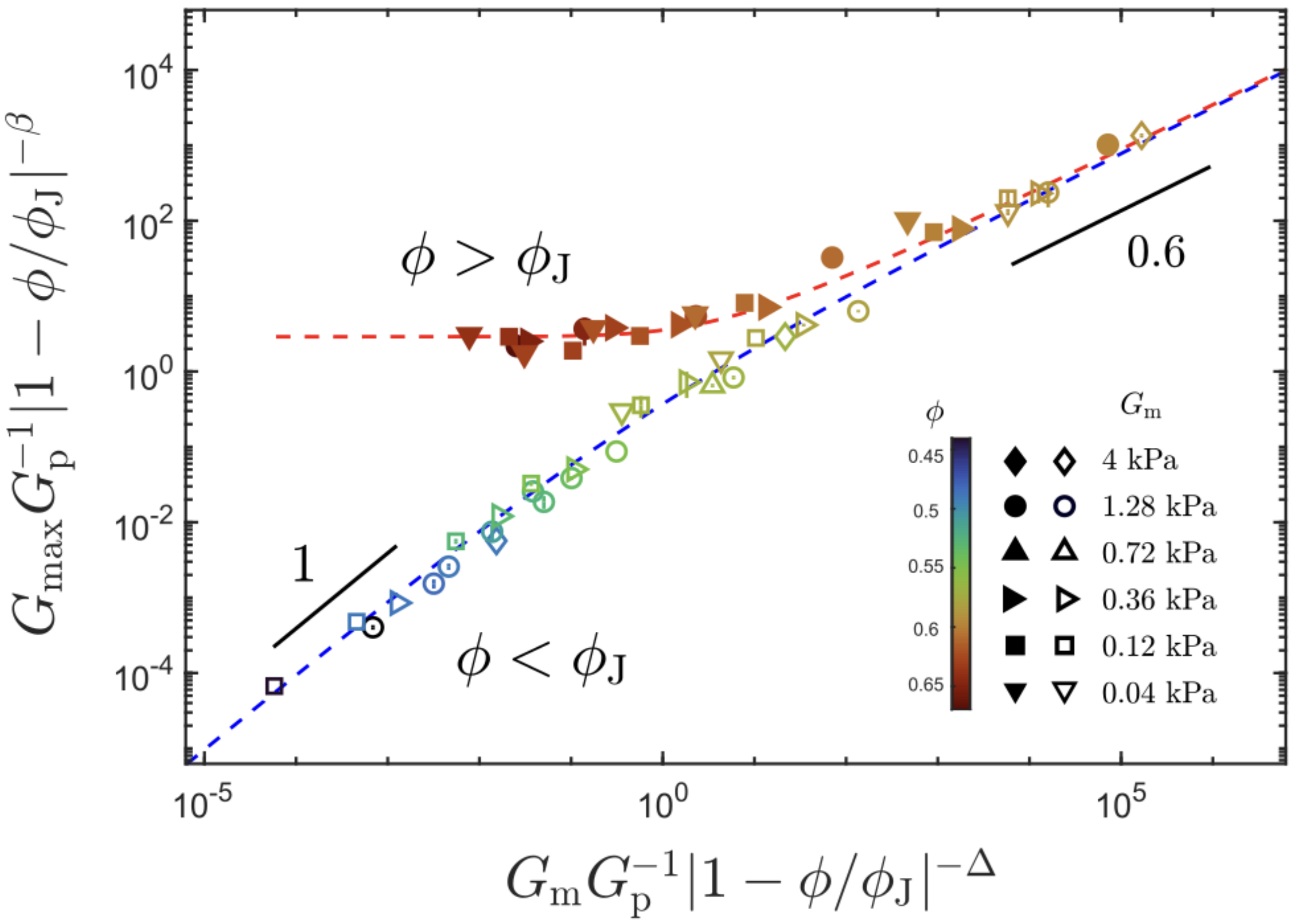




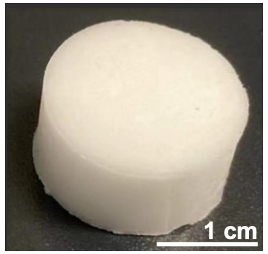
Hertzian contact

$$G_p = 1.6 \pm 0.5 \text{ GPa}$$

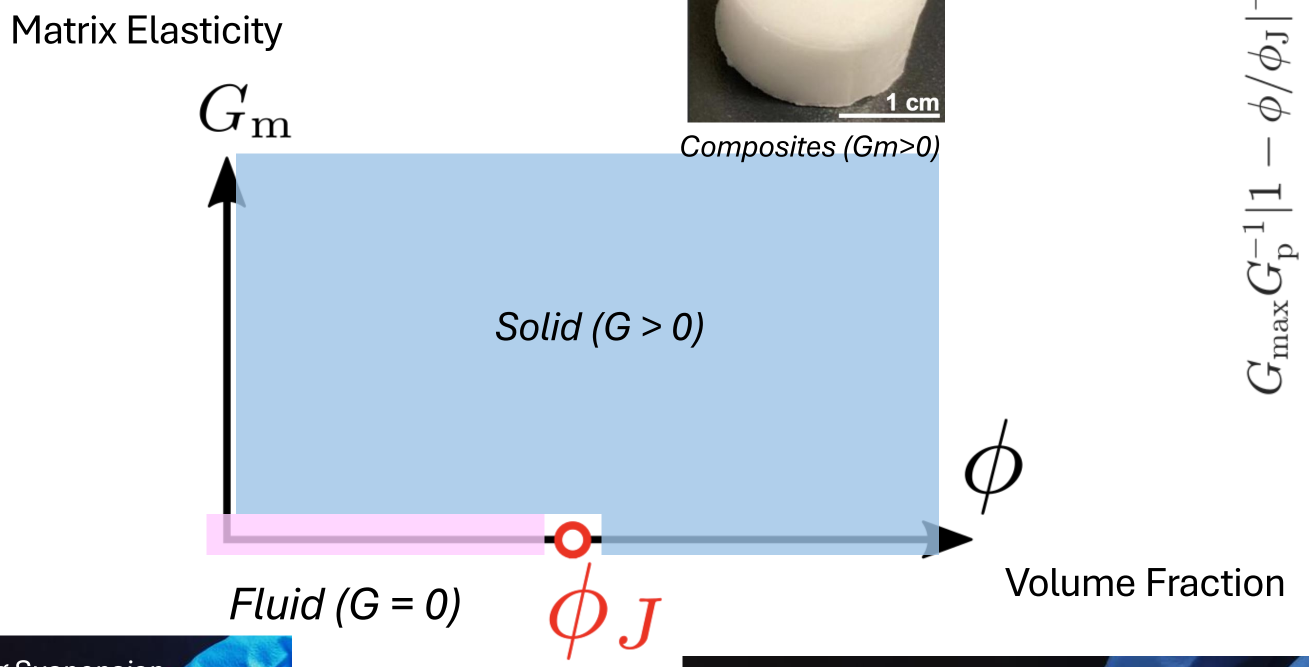
particle material shear modulus

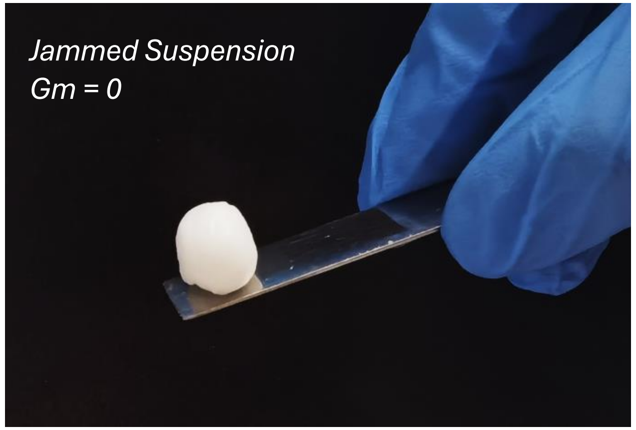


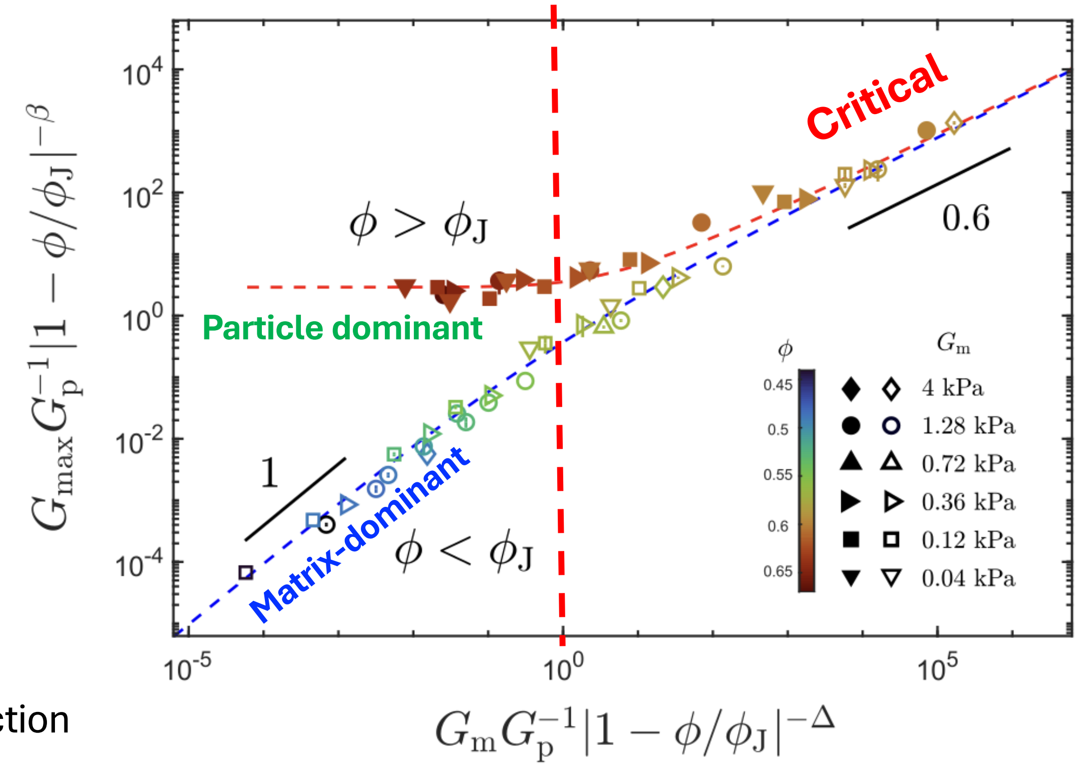
The qualitative picture: how do composites “feel” the jamming point



Composites (Gm>0)

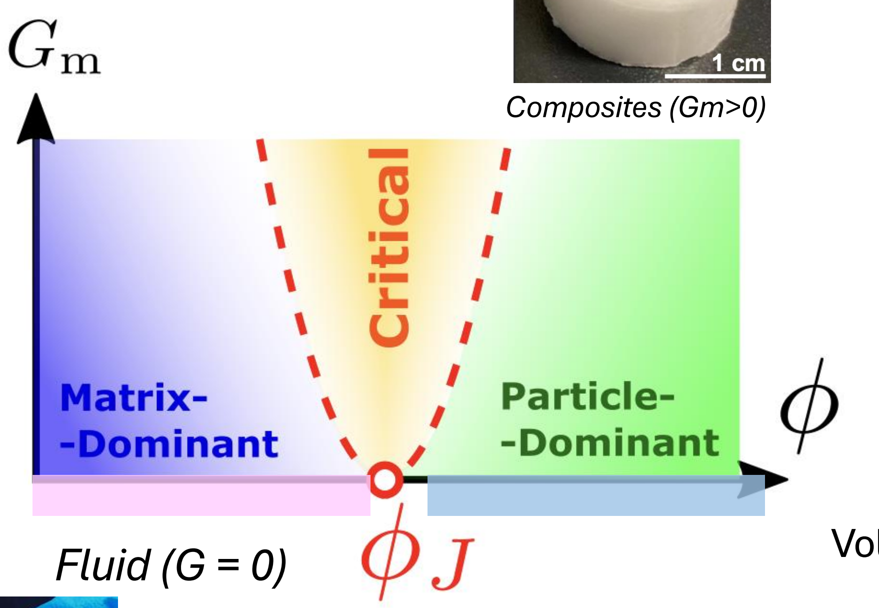


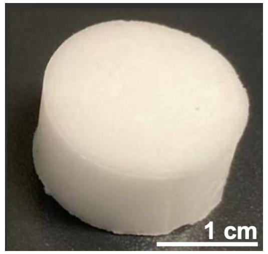




The qualitative picture: how do composites “feel” the jamming point

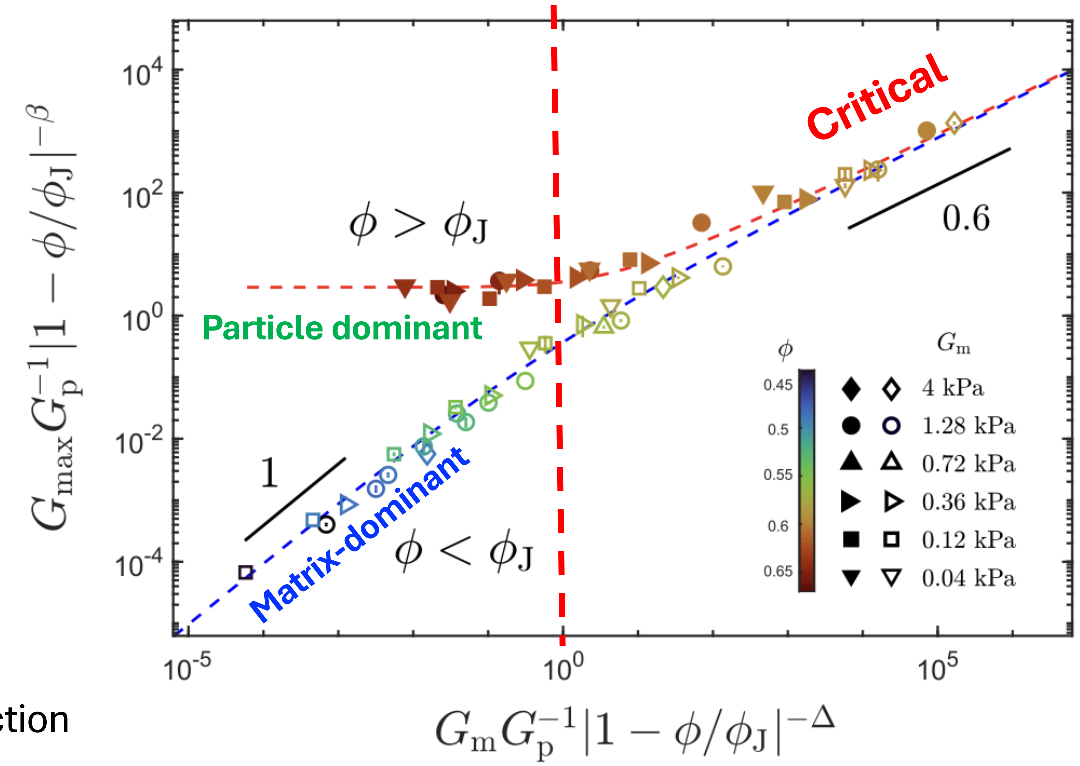
Matrix Elasticity

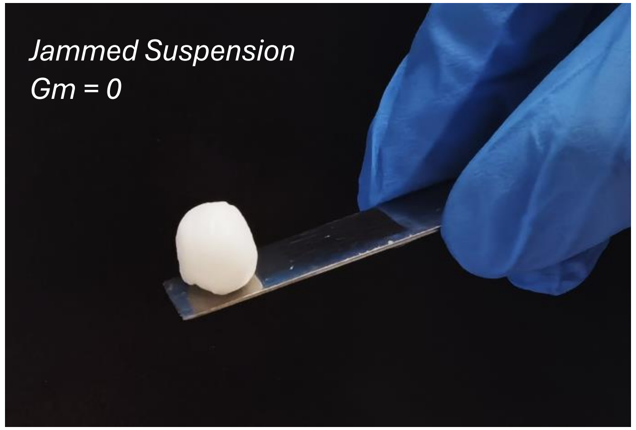




Composites (Gm>0)

Volume Fraction





For a quantitative model: a form of the scaling functions

An empirical fit would give a useful quantitative model.
... and we can choose one that is consistent with a scale-invariant phenomenological free energy.

$$G_{max} = |1-\phi/\phi_J|^{\beta} f_{\pm}\left(\frac{G_m}{|1-\phi/\phi_J|^{\Delta}}\right)$$

[1] Hypothesis: The system sits at the minimum of a Landau-type phenomenological free energy

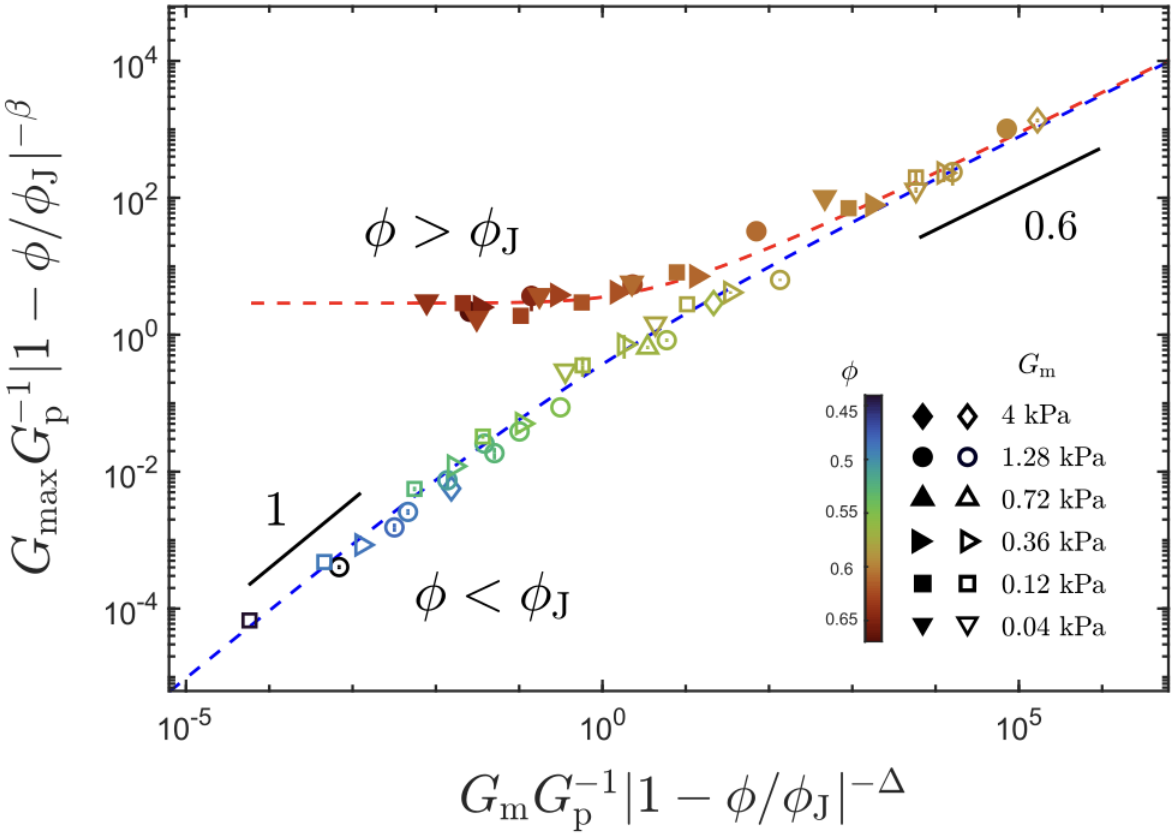
$$L(\Phi, G_{\mathrm{m}}) = F(\Phi, G_{max}) - G_{max} G_{\mathrm{m}}$$

$\Phi \equiv 1 - \phi/\phi_J$ $\qquad l^{-d} F(l^{y_\Phi}\Phi, l^{y_G} G_{max})$

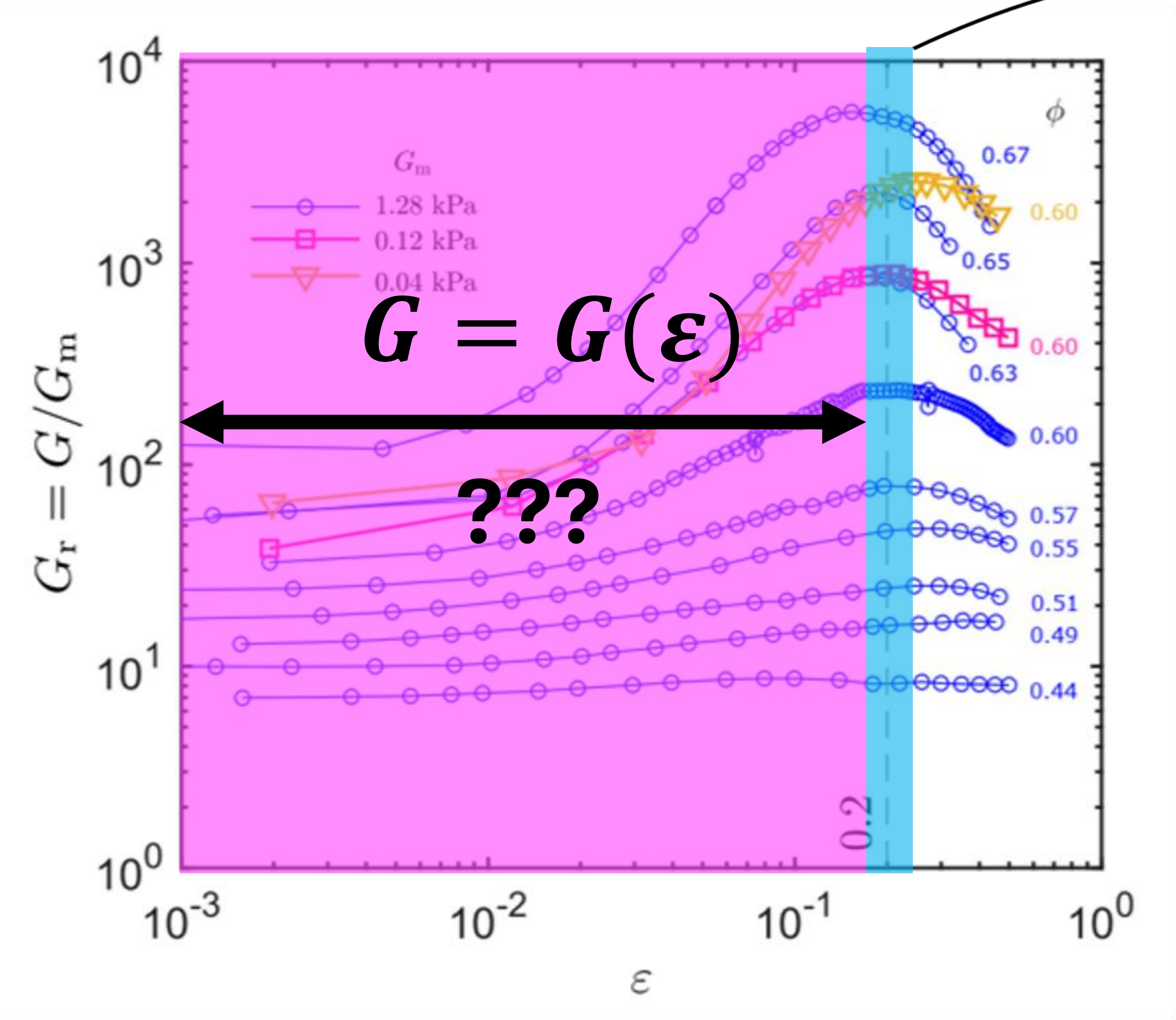
[2] $\partial L/\partial G_{max} = 0 \Rightarrow$ inverse function of $f_{\pm}(x)$:

$$g_{\pm}(x) = c_1 x^{\Delta/\beta} \mp c_2 x^{(\Delta-1)/\beta} \pm x$$

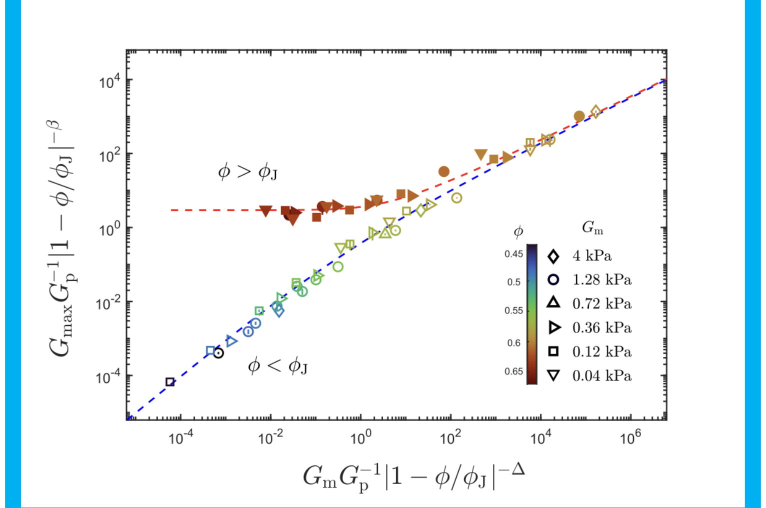
-> a useful quantitative model.



What controls the states in the stiffening regime (under different applied strain)?



$G = G_{max}$

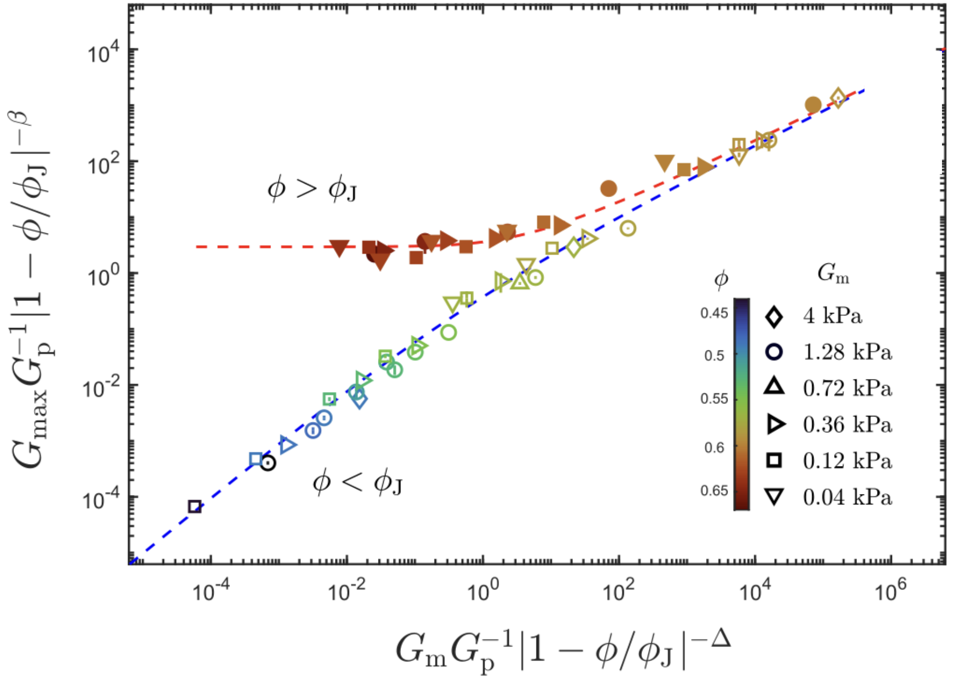


Maximally stiffened states

Collapsing G(ε) using strain-dependent jamming point $\phi_J = \phi_J(\varepsilon)$

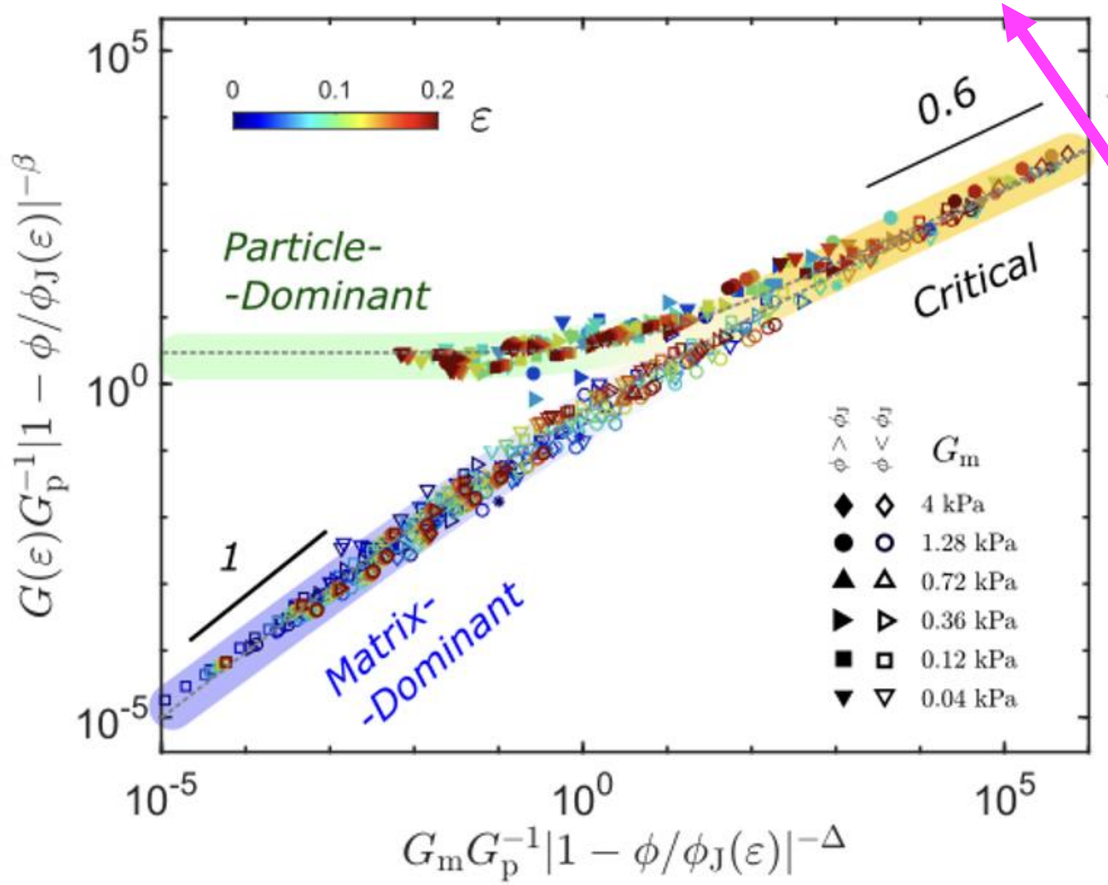
Maximally stiffened states

$$G_{max} = \left|1 - \phi/\phi_J\right|^{\beta} f_{\pm}\left(\frac{G_m}{\left|1 - \phi/\phi_J\right|^{\Delta}}\right)$$



All strained states in the stiffening regime

$$G(\varepsilon) = \left|1 - \phi/\phi_J(\varepsilon)\right|^{\beta} f_{\pm}\left(\frac{G_m}{\left|1 - \phi/\phi_J(\varepsilon)\right|^{\Delta}}\right)$$



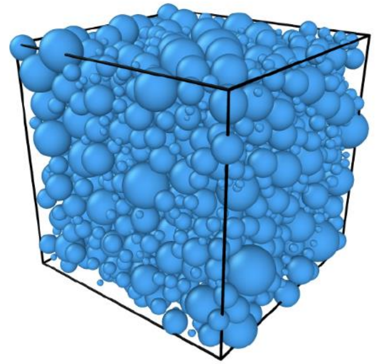
???

Universal scaling functions $f_{\pm}(x)$ and exponents $\beta = 3, \Delta = 5$

Gp: particle material shear modulus

How to understand the $\phi_J(\varepsilon)$ relation that collapse composite data

Frictionless isotropic jamming point (random close packing) (0.676 from simulation)



Yulu Huang @ HKUST

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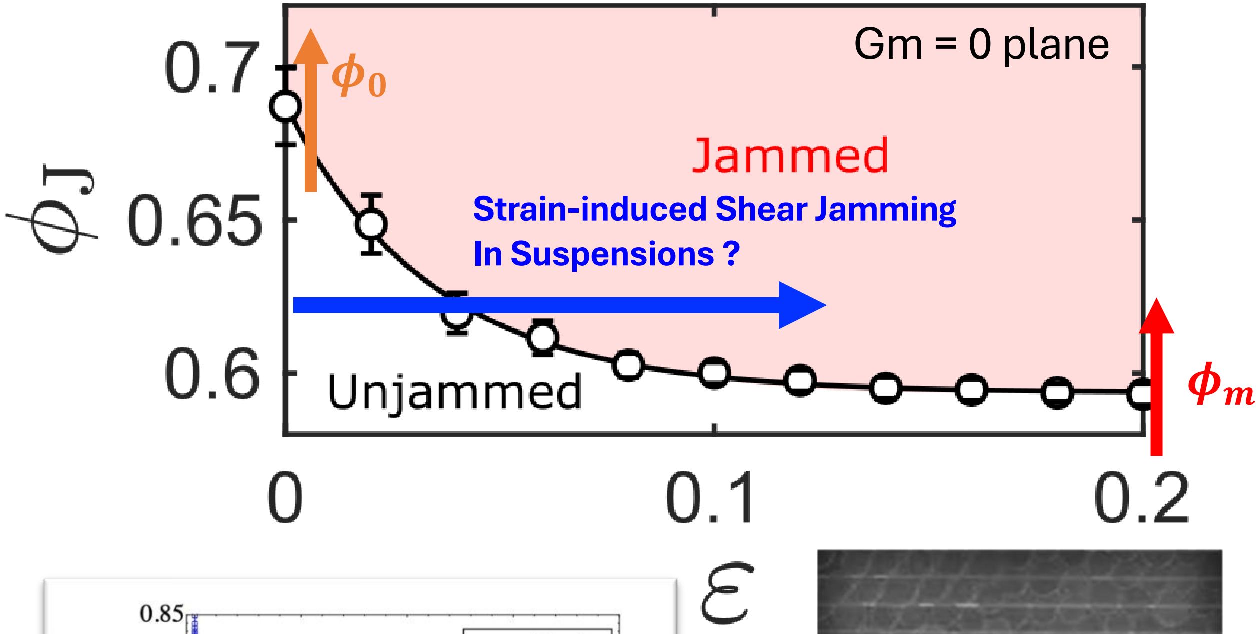
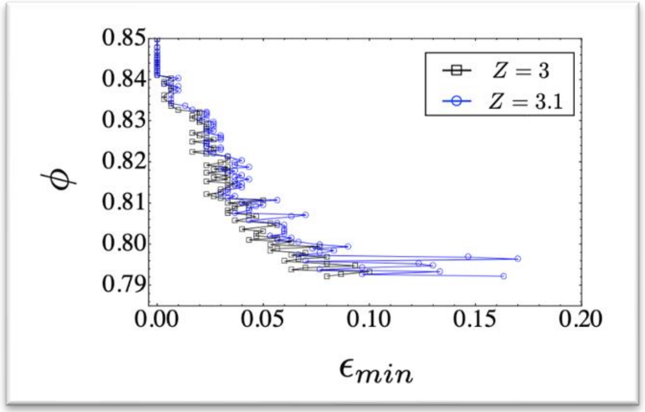
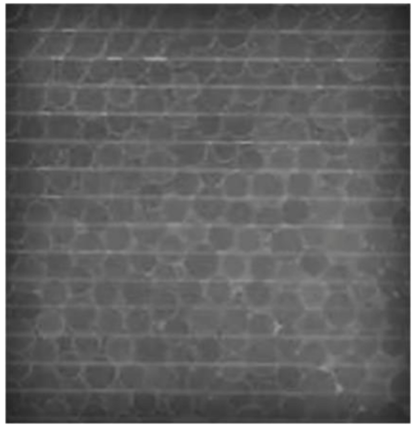


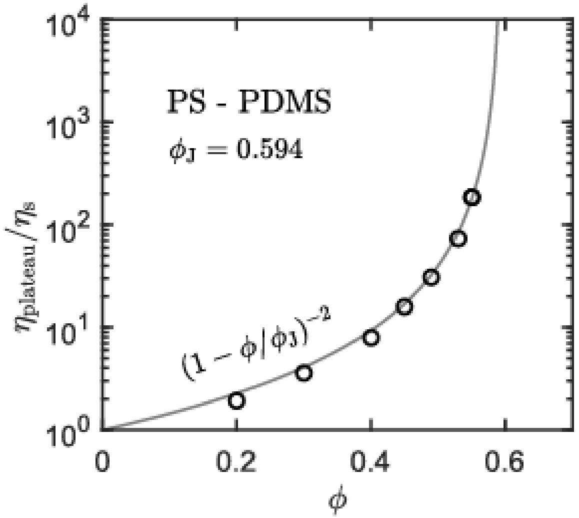


Bi et al., Nature 480, 355-358 (2011)



Source: Behringer Lab @ Duke

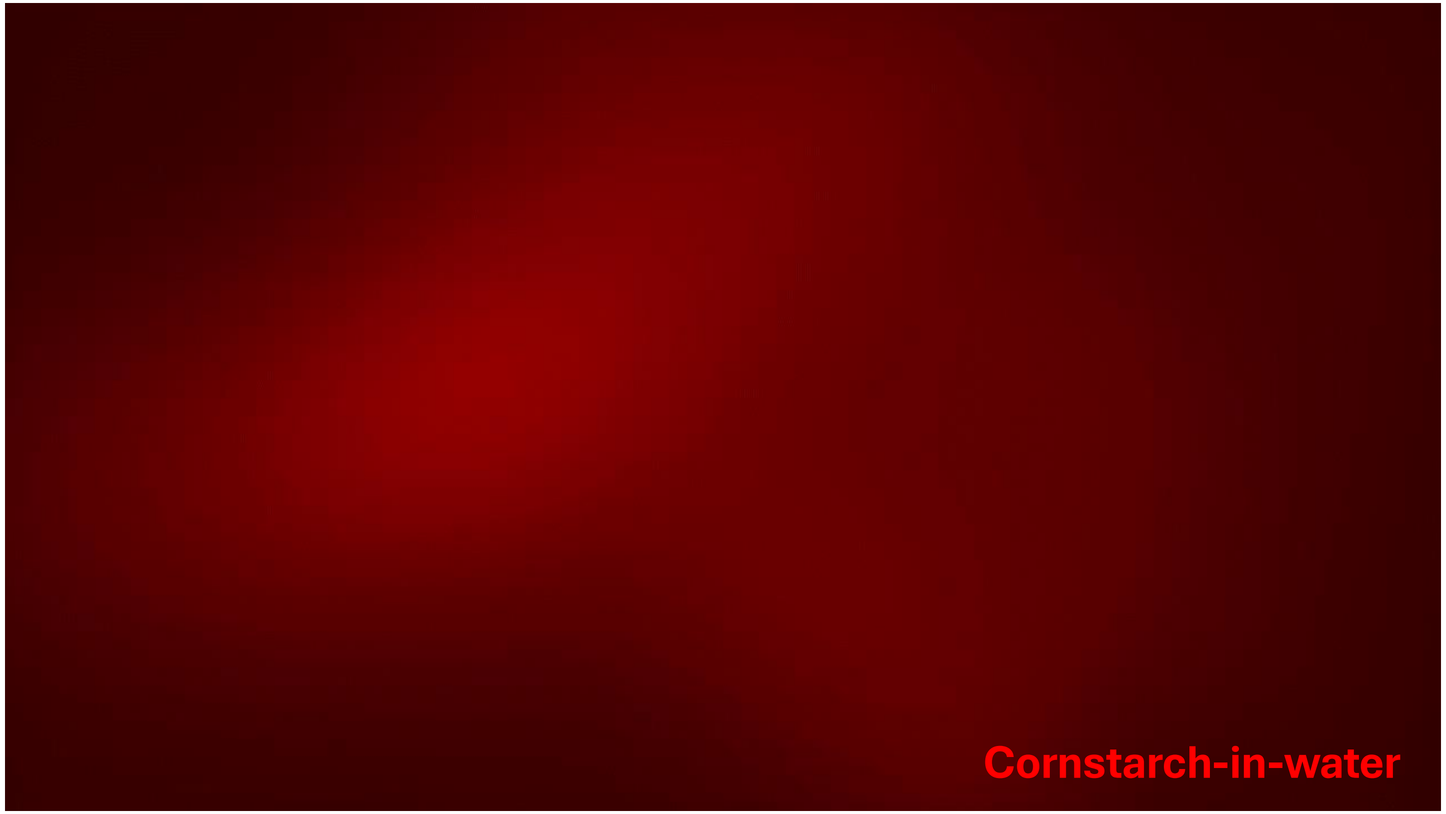
Steady flow jamming point (large shear strain)



PS in silicone oil (uncrosslinked PDMS base polymer)

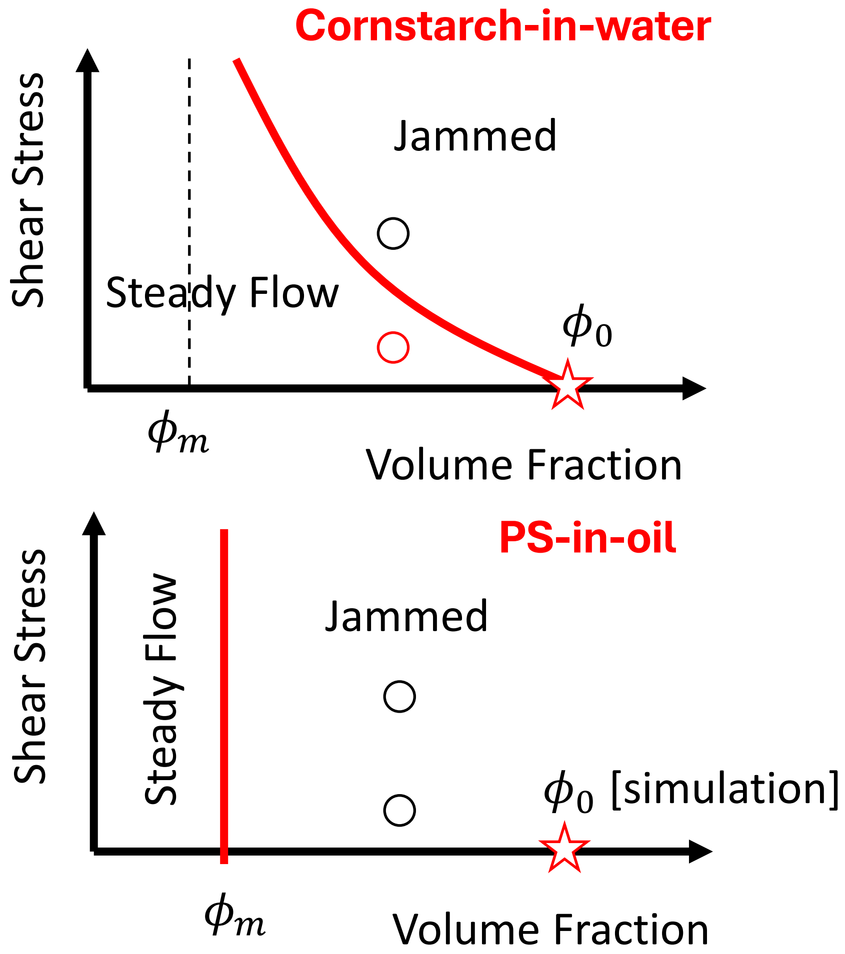
Does a granular suspension shear jam under strain?

- In the suspension literature, shear jamming is usually studied in shear-thickening systems, and is stress-controlled.
- PS-in-oil suspension does not shear thicken, and we did not observe stress-controlled shear jamming.
- Can they shear jam under quasi-static strain like dry granular materials? -> How to prepare the initial state?



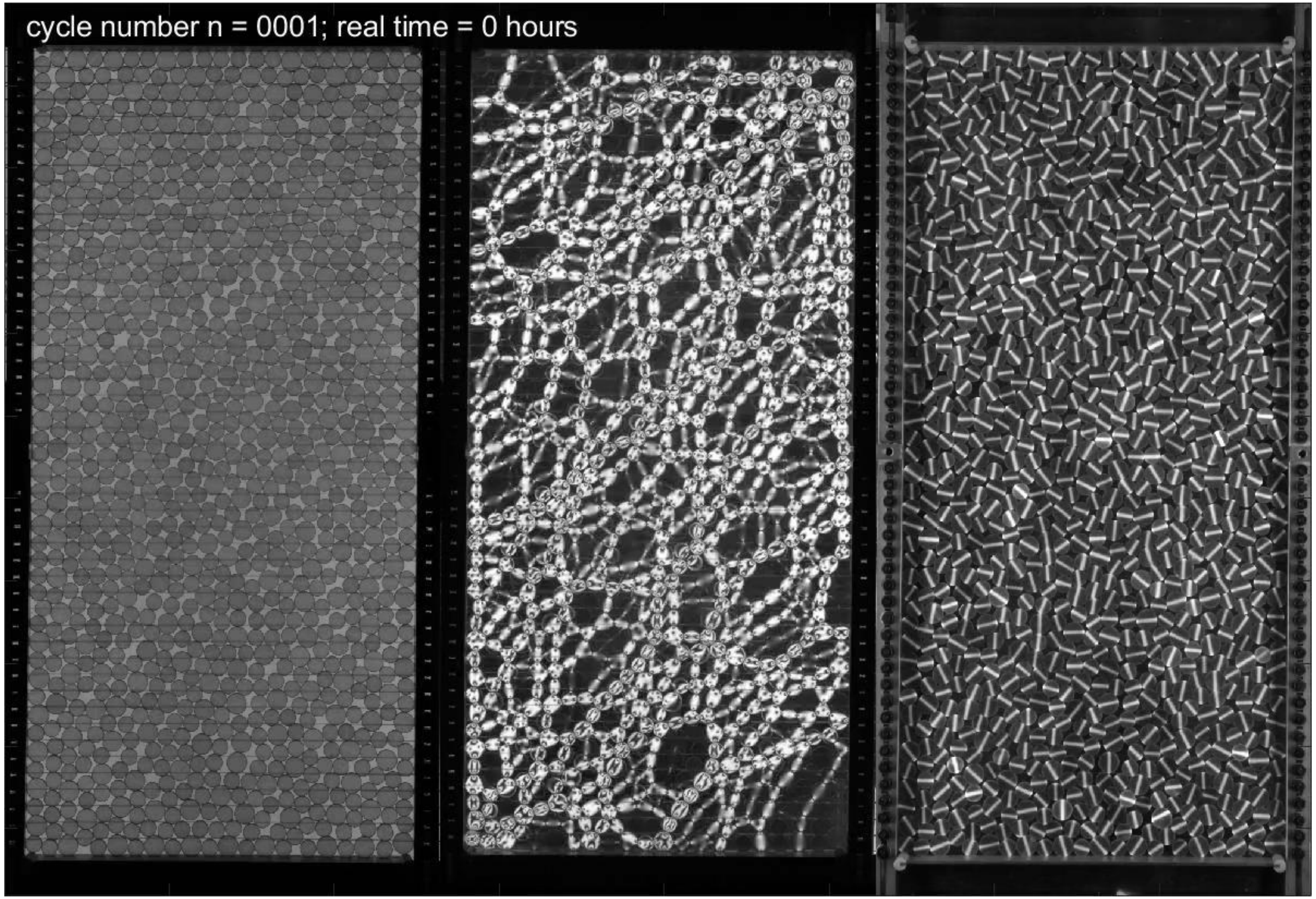
Source: Jaeger Lab @ Chicago

Peters, Majumdar, Jaeger, Nature 532, 214-217 (2016)



Does a granular suspension shear jam under strain?

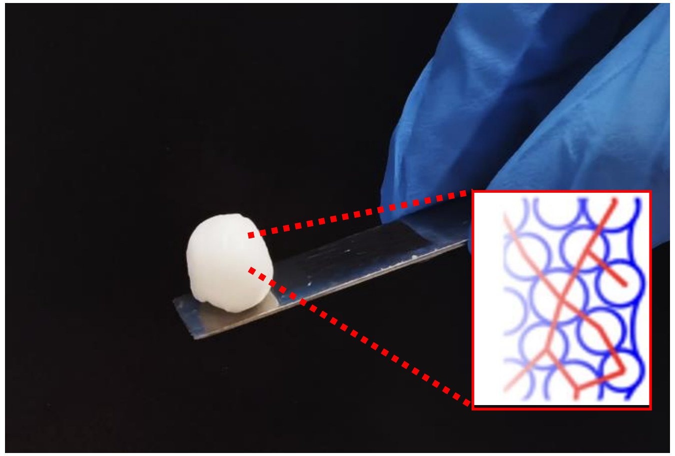
Motivation: Small-amplitude oscillatory can "melt" a jammed solid with $\phi > \phi_{m/SJ}$



Robert P. Behringer @ Duke

Joshua E. S. Socolar @Duke

Bulbul Chakraborty @ Brandeis



Y. Zhao, Y. Zhao, D. Wang, H. Zheng, B. Chakraborty, J. E. S. Socolar, Phys. Rev. X 12, 031021(2022)

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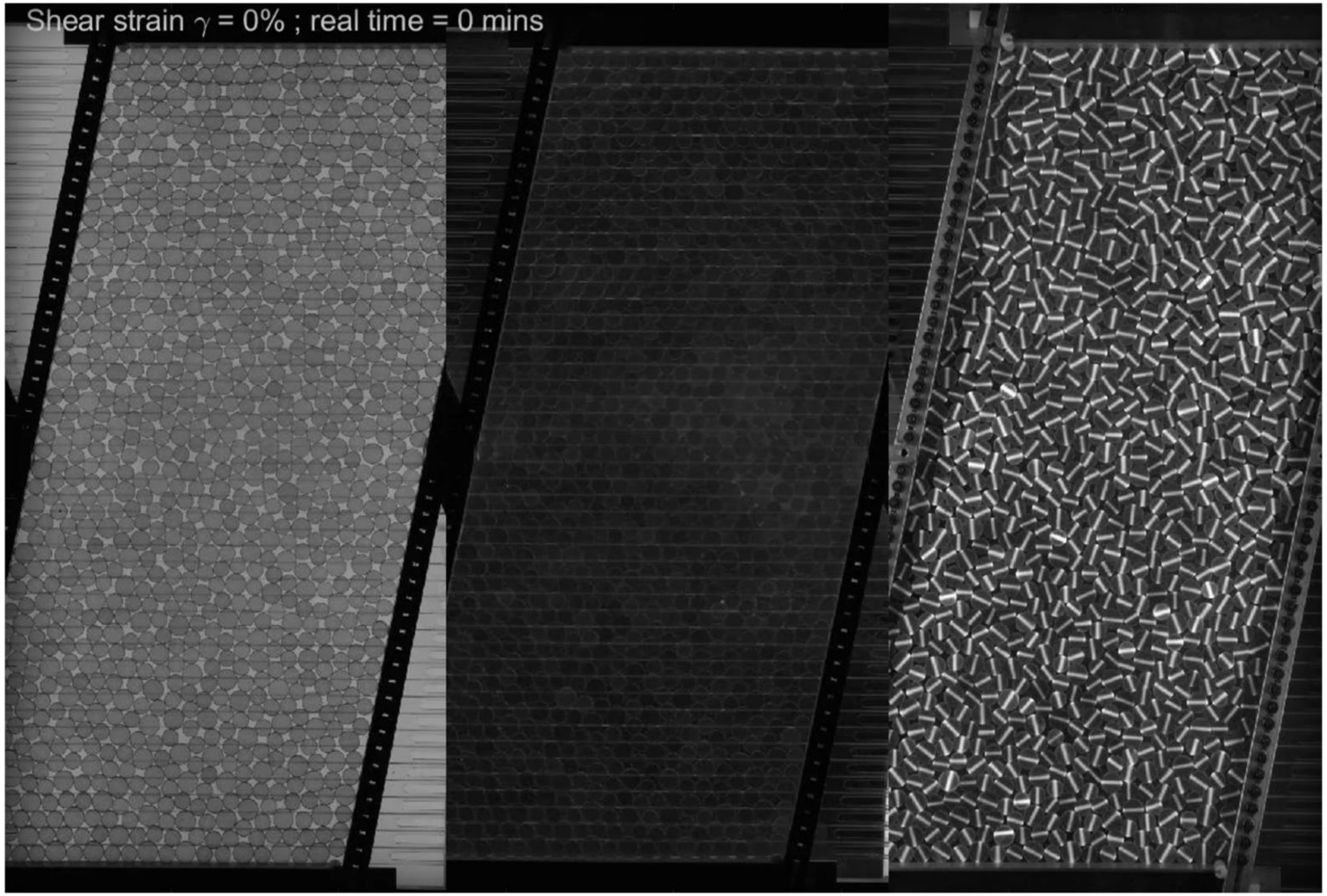

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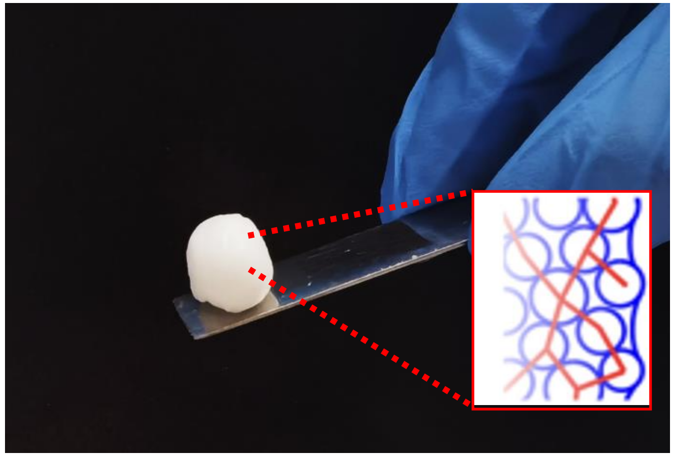

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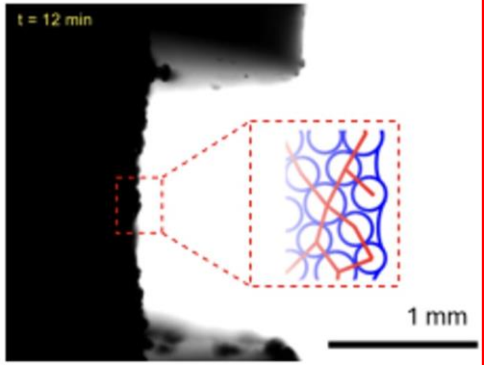
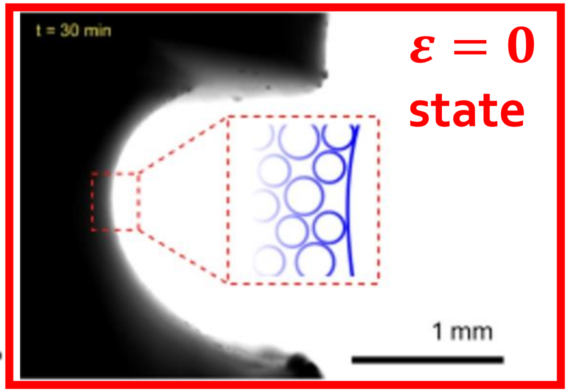
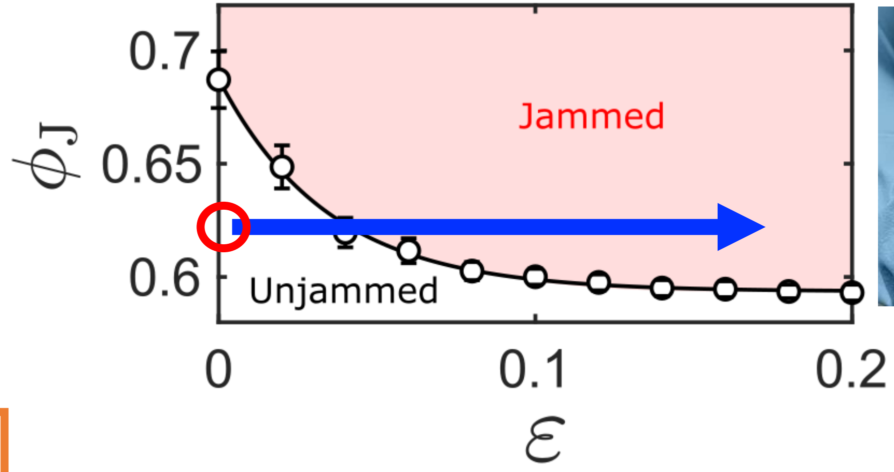
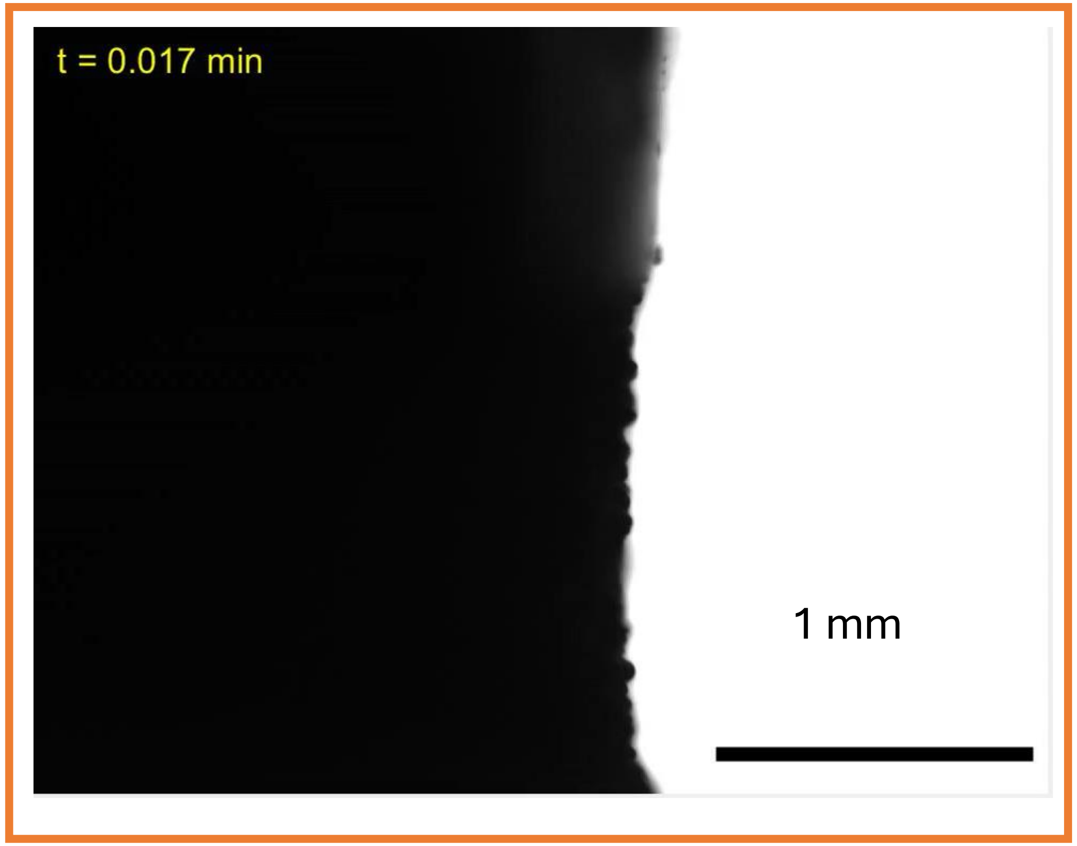
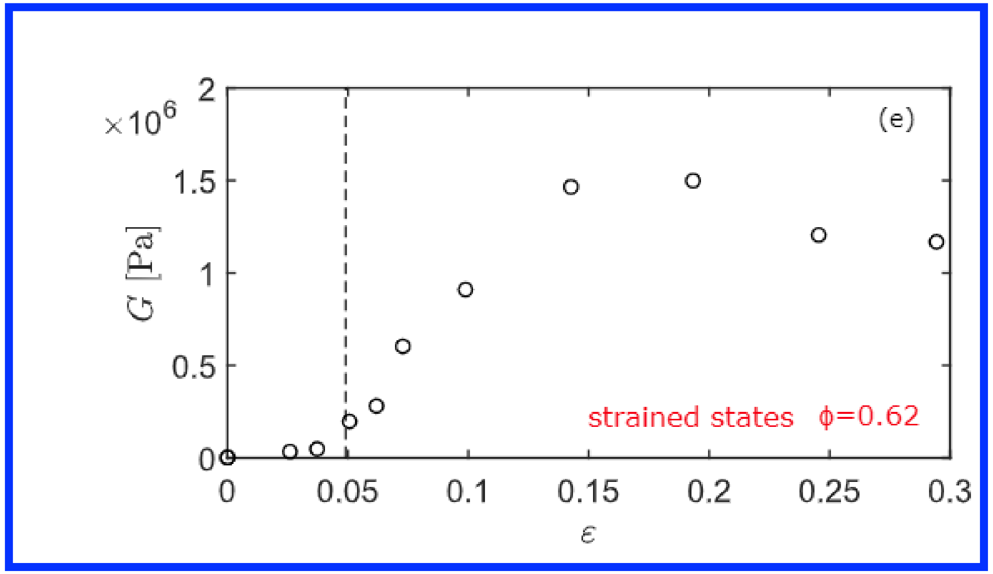






PS in silicone oil (experiment)





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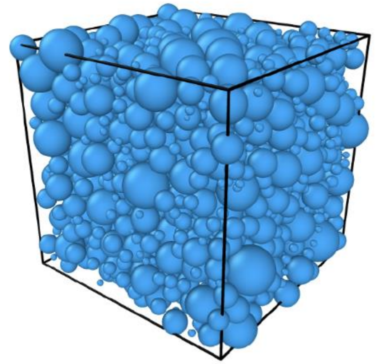

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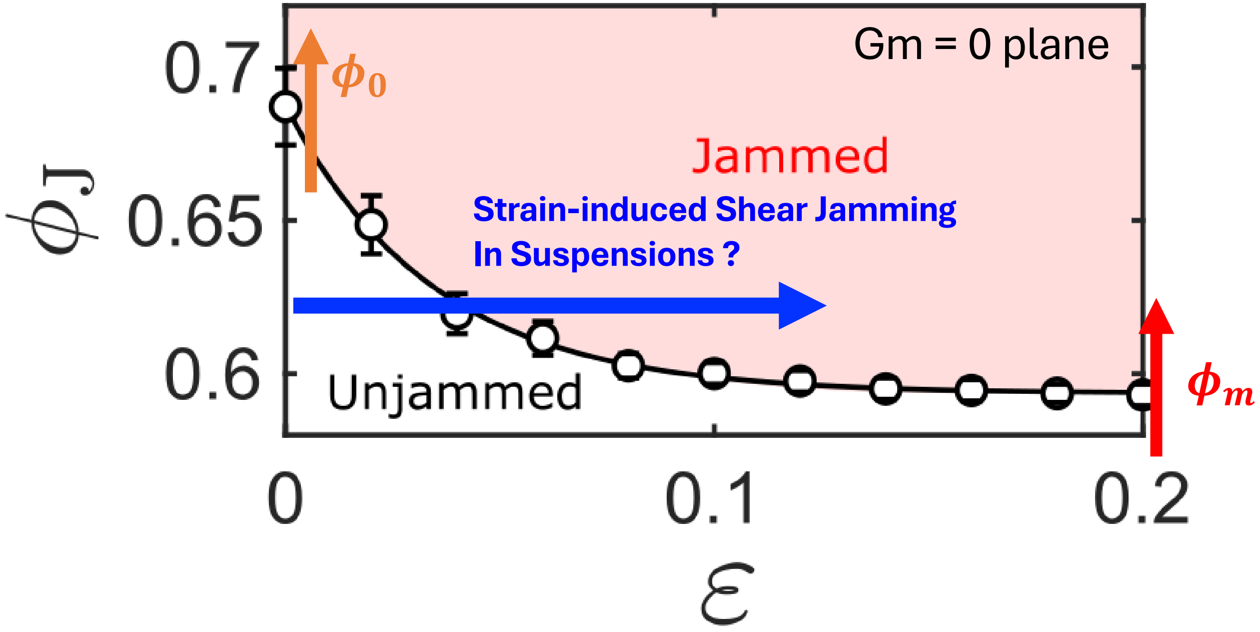

$$\phi_J(\varepsilon) = \phi_m + (\phi_0 - \phi_m)e^{-\varepsilon/\varepsilon^*}$$

Kumar and Luding, Granular Matter 18, 58 (2016)
Han et al., Phys. Rev. Fluids 3 (7), 073301 (2018)
Zhao et al., Phys. Rev. Lett. 123, 158001 (2019)

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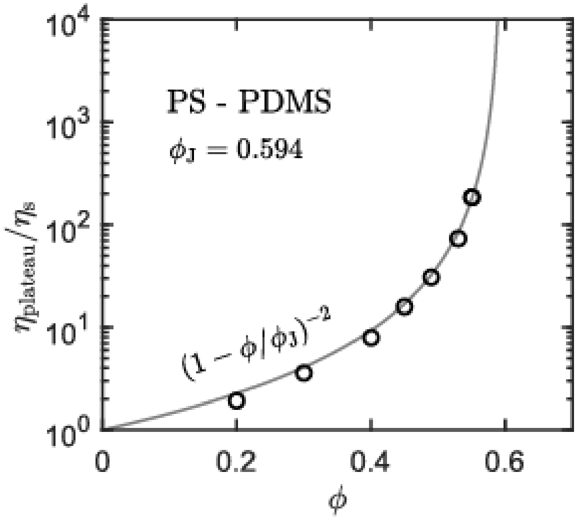


PS in silicone oil (uncrosslinked PDMS base polymer)

What governs the mechanics of soft composites in the dense limit?

Composite Elasticity

$$G = G(\phi, G_m, \varepsilon)$$

Particle Volume Fraction

Matrix Elasticity

Shear Deformation

Quantitative model for composite strain-stiffening in the dense and soft limits

$$\boldsymbol{G = G(\varepsilon, \phi, G_{\mathrm{m}})}$$

[1] The scaling ansatz

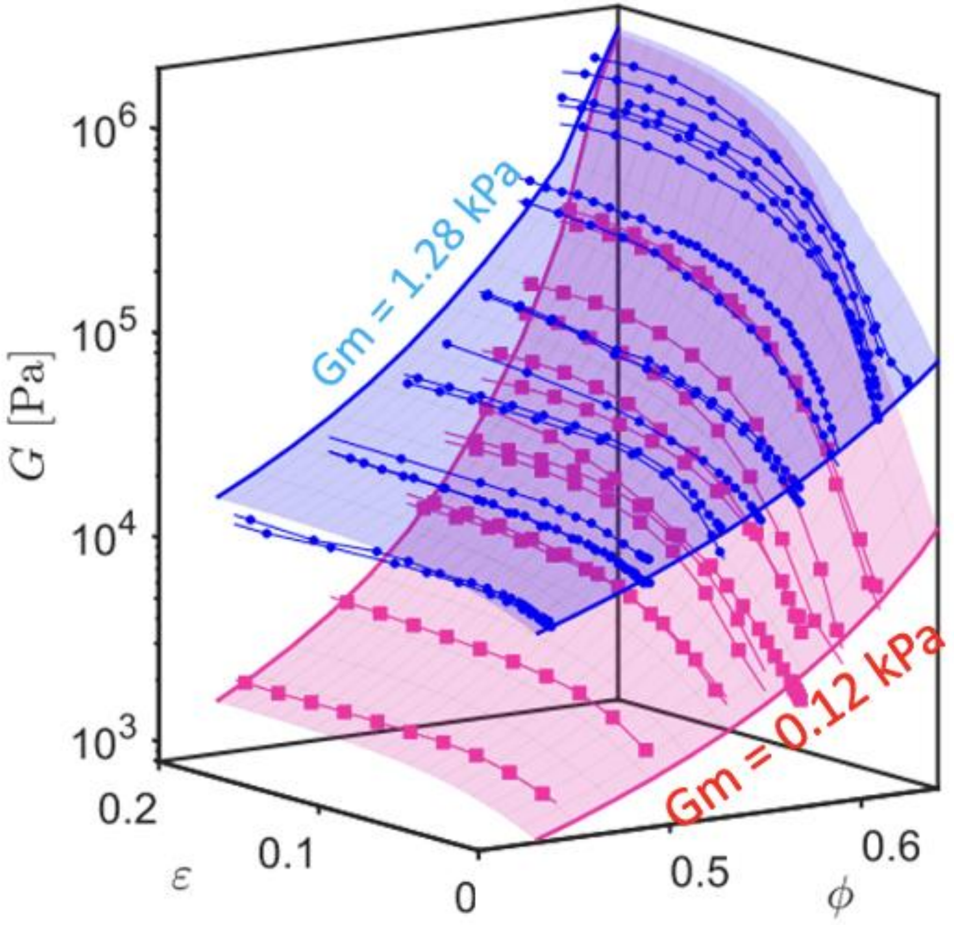
$$G(\varepsilon, \phi, G_{\mathrm{m}}) = |1 - \phi/\phi_{\mathrm{J}}(\varepsilon)|^{\beta} f_{\pm}\left(\frac{G_{\mathrm{m}}}{|1 - \phi/\phi_{\mathrm{J}}(\varepsilon)|^{\Delta}}\right)$$

[2] An explicit form of the (inversed) scaling functions

$$g_{\pm}(x) = c_1 x^{\Delta/\beta} \mp c_2 x^{(\Delta-1)/\beta} \pm x$$

[2] Strain-dependence: Granular shear jamming boundary

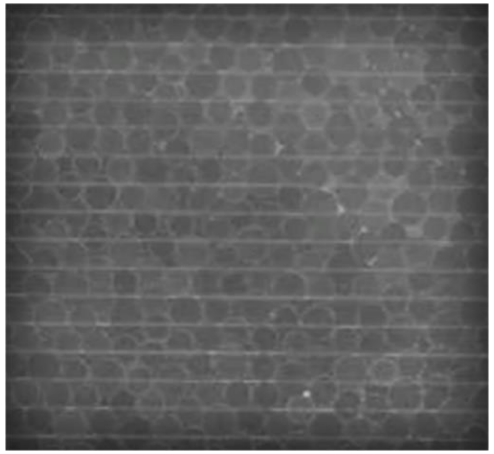
$$\phi_{\mathrm{J}}(\varepsilon) = \phi_{\mathrm{m}} + (\phi_0 - \phi_{\mathrm{m}}) e^{-\varepsilon/\varepsilon^*}$$



Nat. Commun. 15, 1691 (2024)

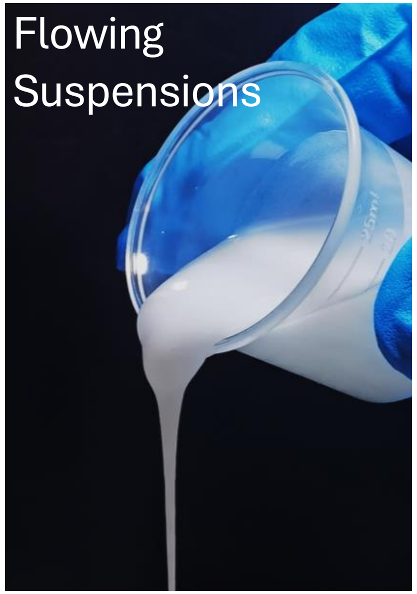
Strain-stiffening in the dense limit as cross-over phenomenon

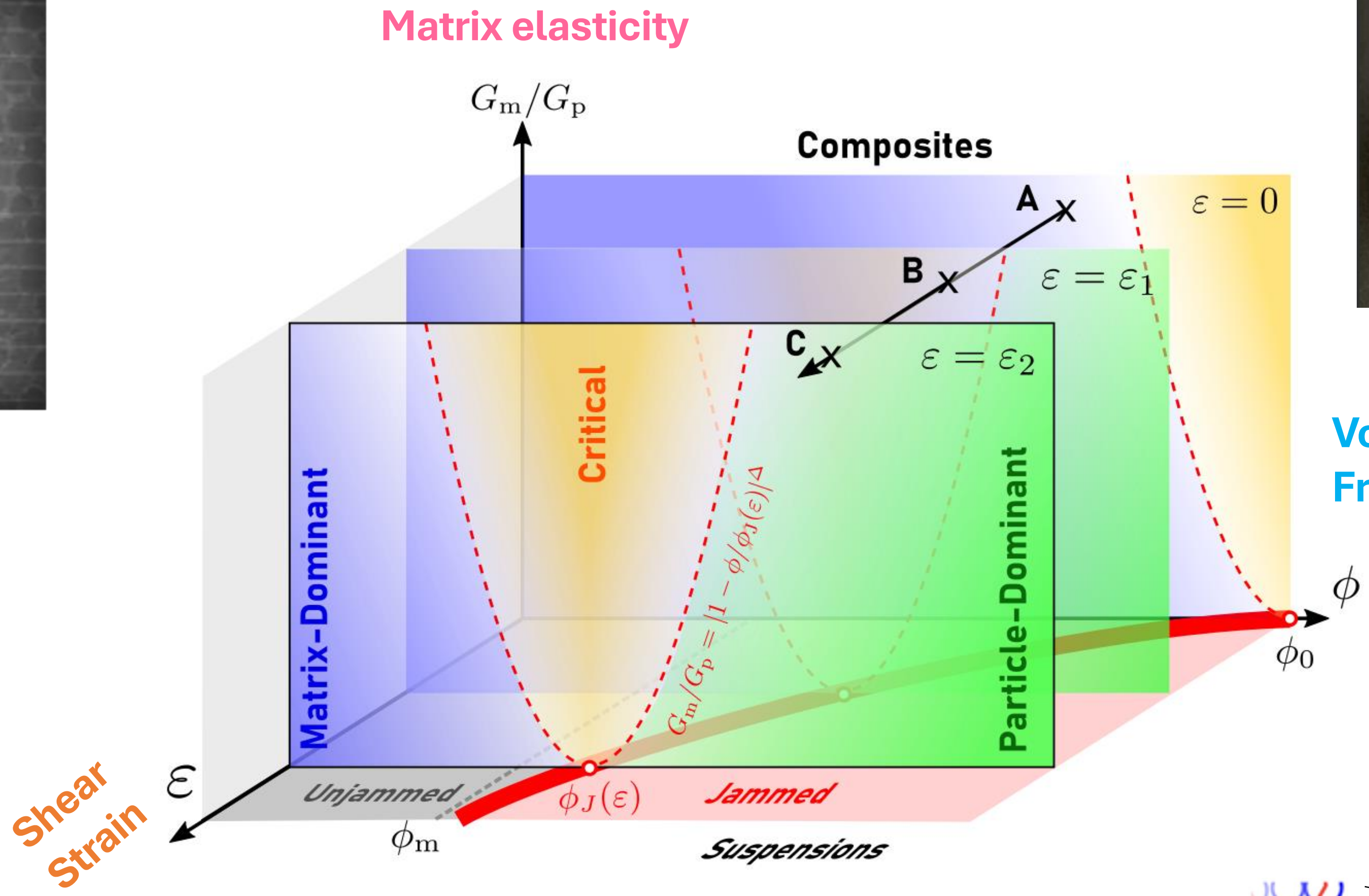
Gm = 0 Shear Jamming Transition



Source: Behringer Lab @ Duke

Gm = 0 Fluids





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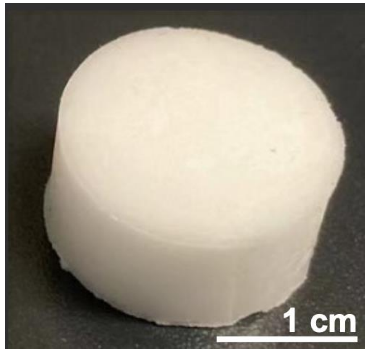
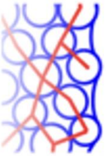
Gm > 0 Solid Composites



Gm = 0 Solids
(Jammed Suspensions)



Source: Jaeger Lab @ Chicago

Conclusion: Jamming in (Dense and Soft) Amorphous Solid Composites

- ✓ **Granular shear jamming affects composite mechanics in a way resembling critical phenomenon**
- ✓ **New design ideas for functional soft materials**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Yifan Wang
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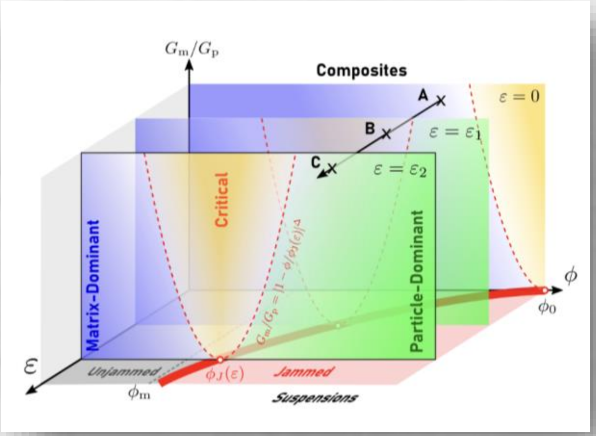

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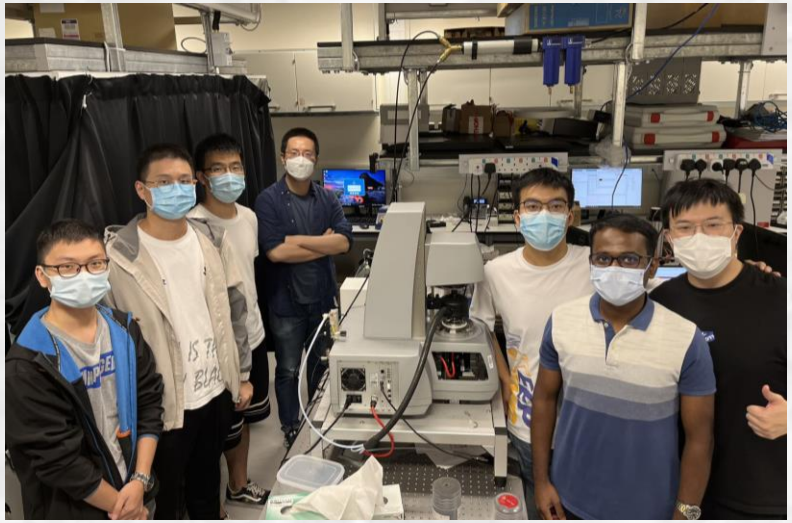

Complex Fluids and Soft Matter @ HKUST

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