5th Edwards Symposium – Future Directions in Soft Matter

Rigidity, Softness and Stress Redistribution in soft particulate gels

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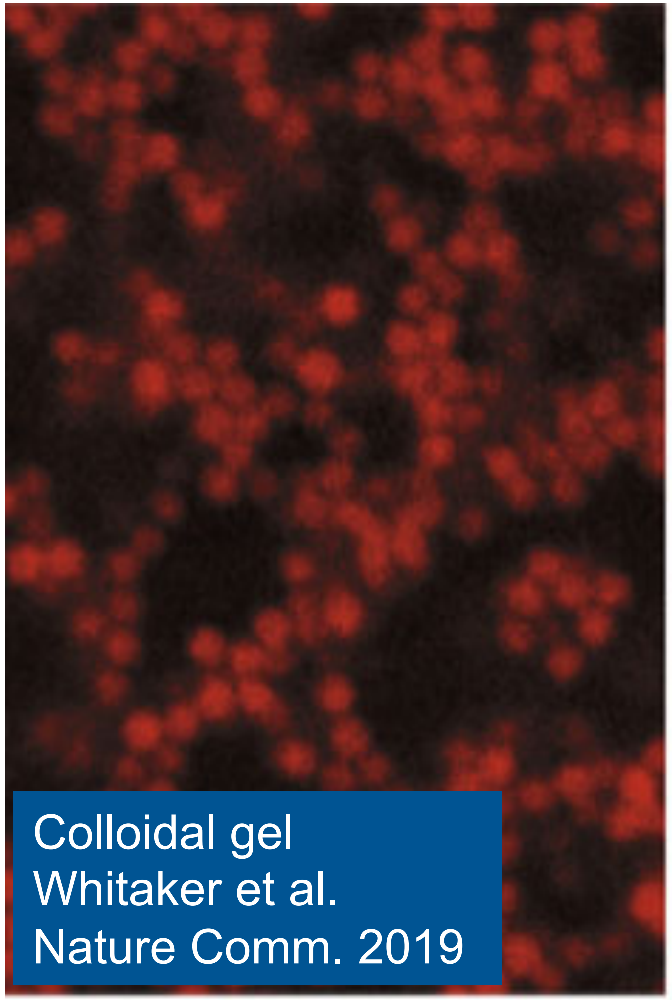
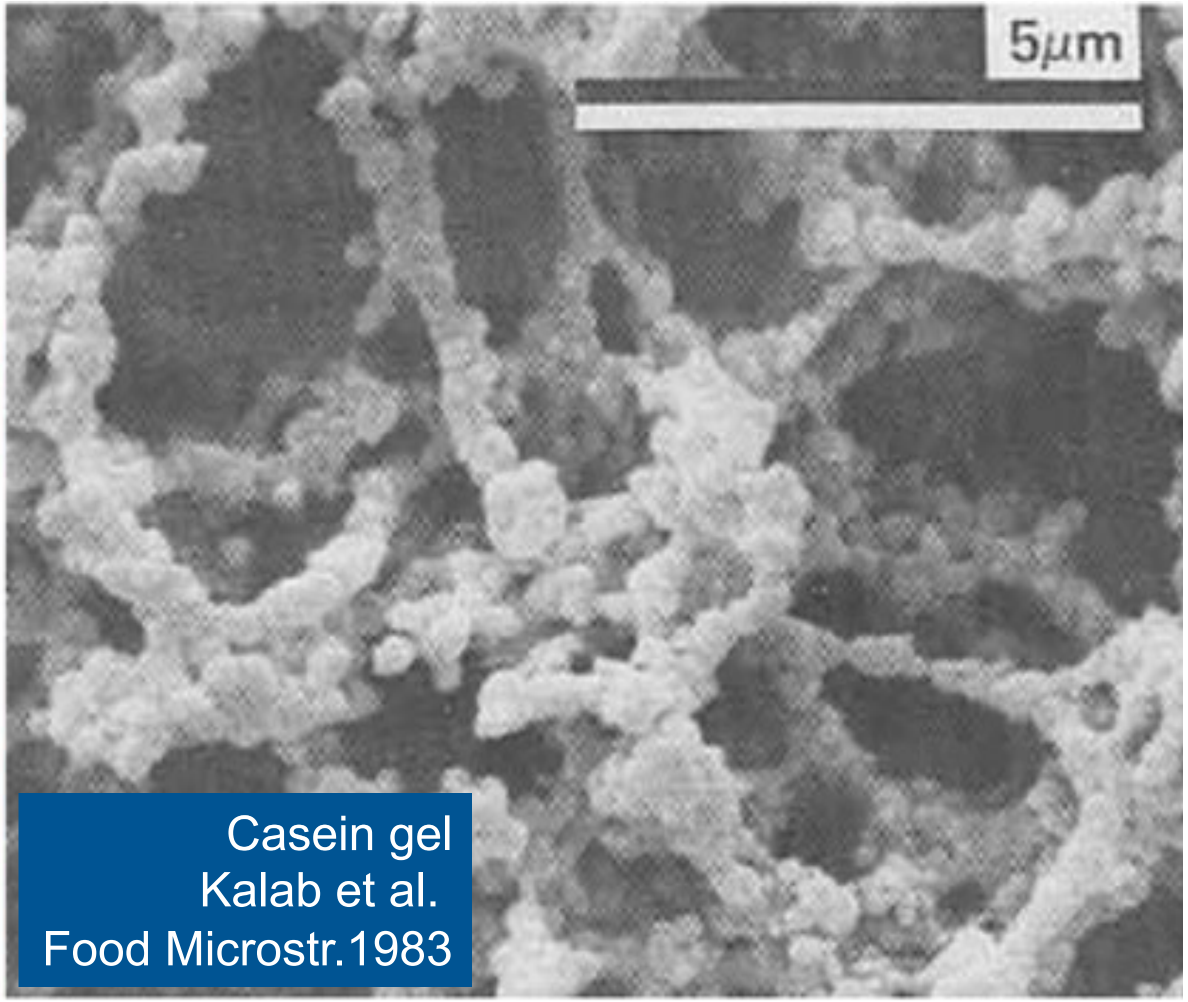
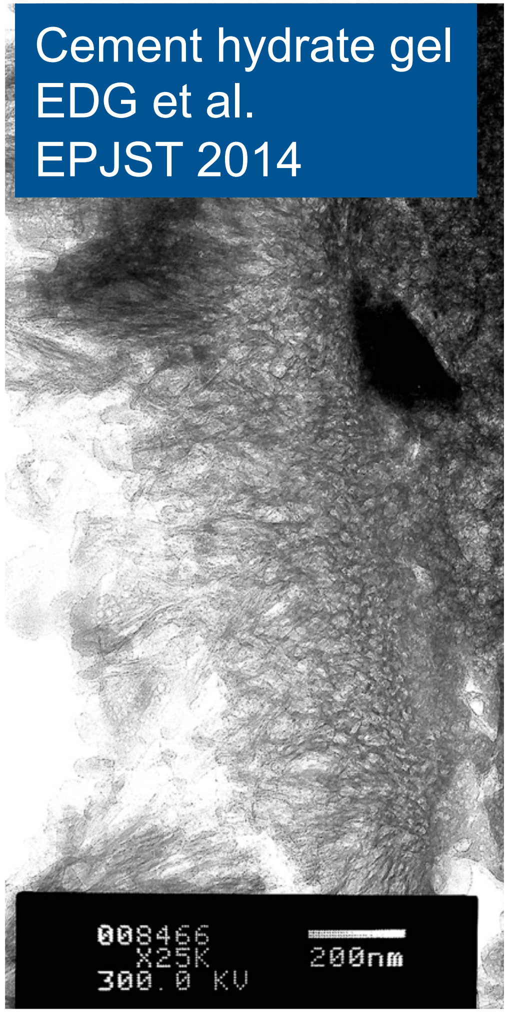
Soft particulate gels



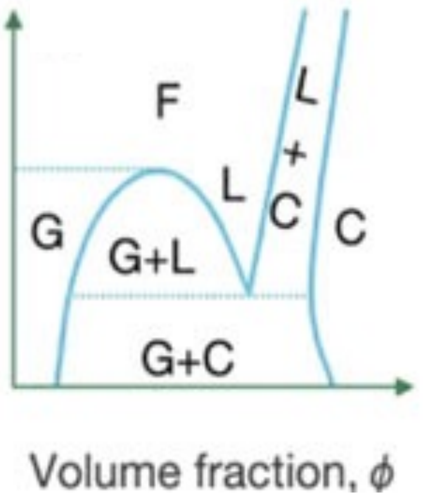




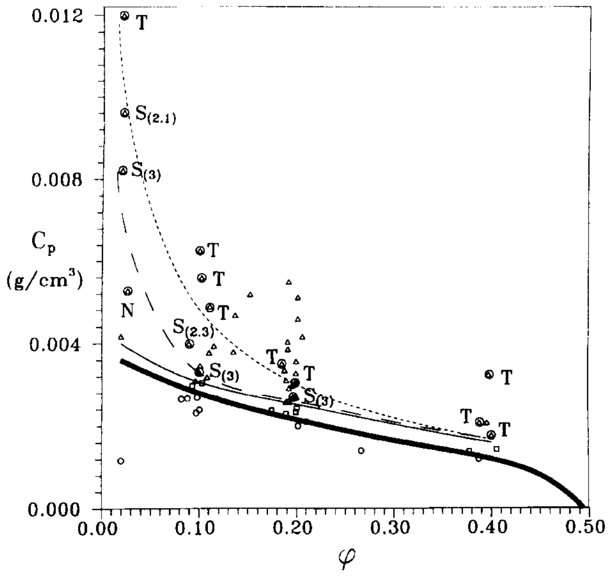
See also batteries materials, biological scaffolds and matrices, clays gels, ceramics…

EDG, Stress Localization in Soft Particulate Gels,
Encyclopedia of Complexity and Systems Science, Springer 2021

Colloidal gels



Anderson & Lekkerkerker, Nature 2002



Poon, Pirie, Haw & Pusey, Physica A 1997

- From physical chemistry to microscopic forces and phase boundaries (ex. DLVO)

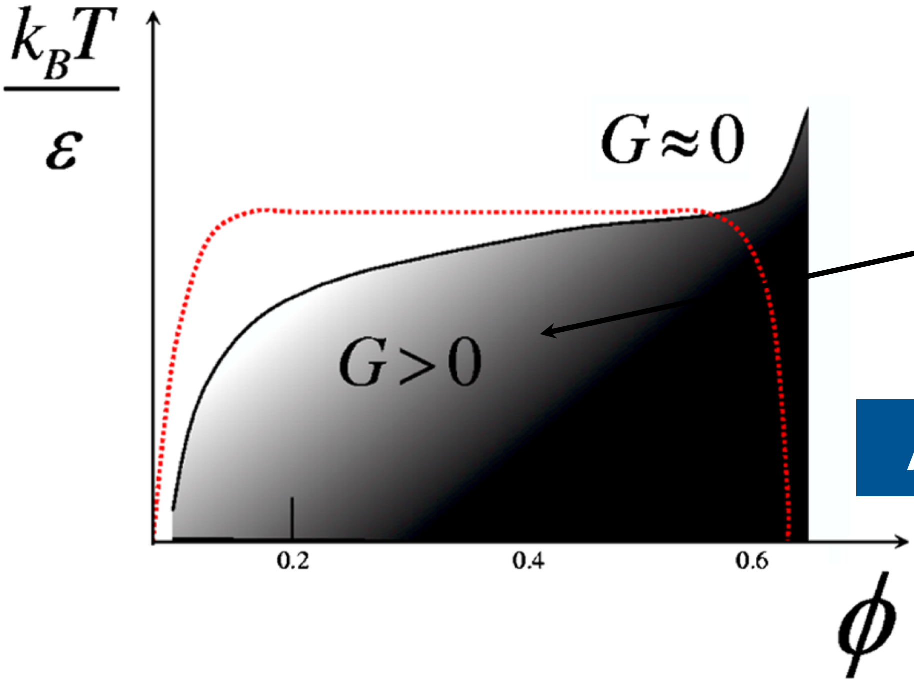
Gelation can be initiated by:

- Equilibrium phenomena: phase separation, microphase separation, hierarchical self-assembly …
- Non-equilibrium aggregation: diffusion limited processes, fractal growth…

Ultimately these gels result from **frustration** in the growth of larger scale structures that emerge from microscopic collective processes -> **localized stress patterns**

Are there general organizing principles for the mechanics of these structures?

Elasticity and rigidity



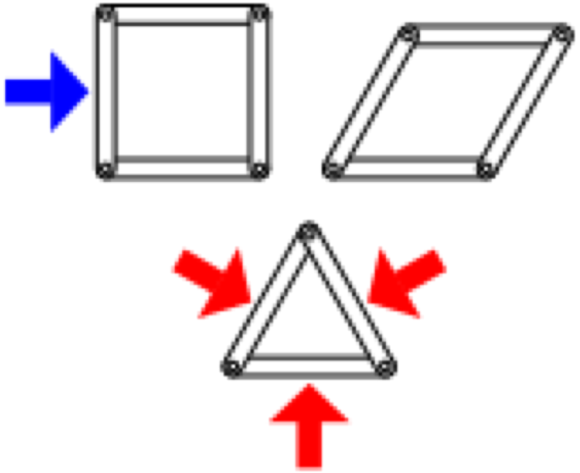
Different structures/lengthscales contributing to and controlling elasticity, depending on microscopic forces, aggregation paths, gelation kinetics, external forces …

A **sub-set** of the elastic connections controls the elasticity

A. Zaccone, H. Wu and EDG, PRL 2009

Elasticity is mainly **enthalpic** and connectivity or local coordination does not guarantee rigidity: role of **rigidity percolation**.

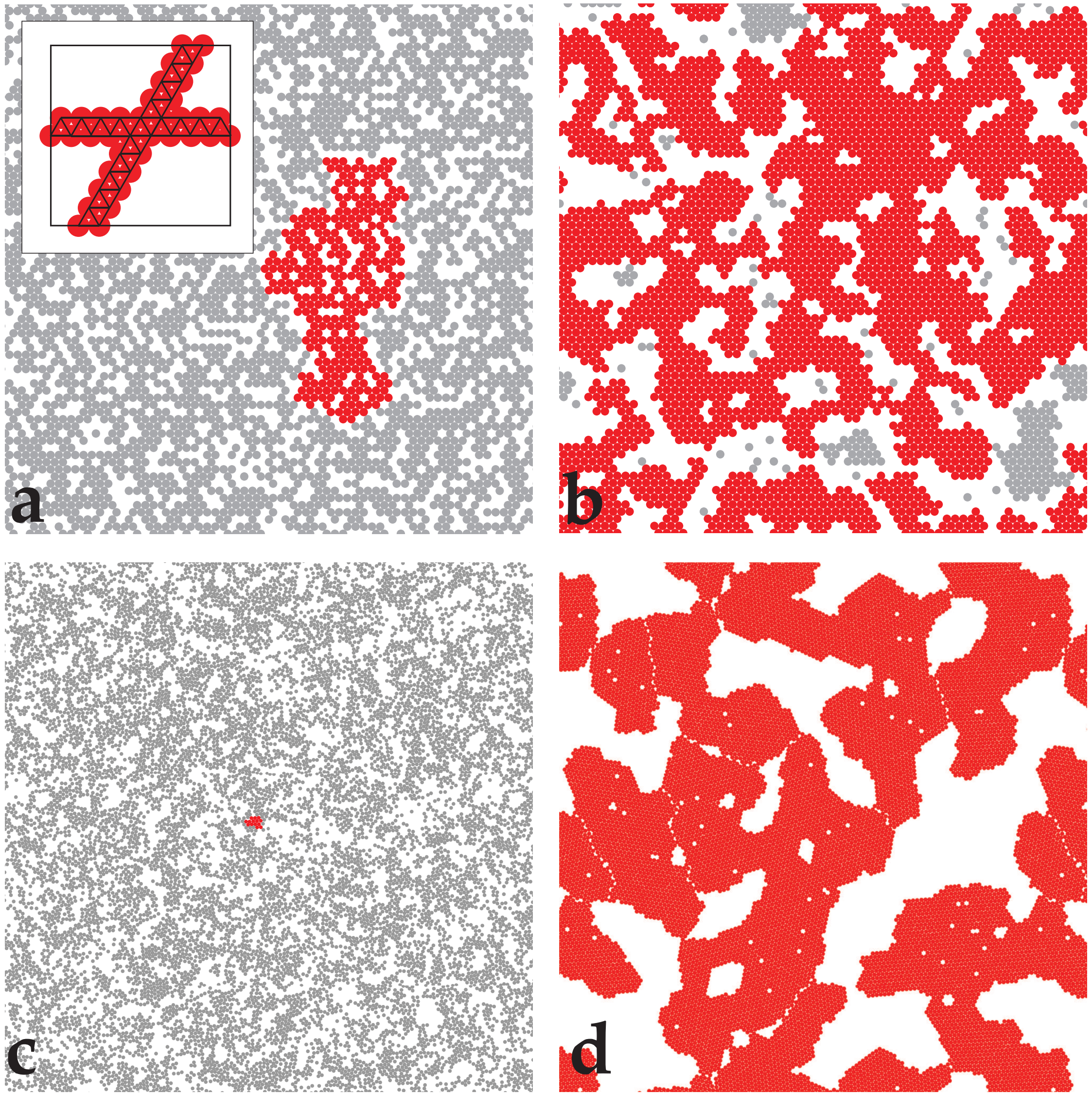
S. Alexander, Phys. Rep. 1998; M. Sahimi, Phys. Rep. 1998; He & Thorpe, PRL 1985; Jacobs & Thorpe, PRL 1995; Broedersz et al., Nat. Phys, 2011



Source: Wikipedia

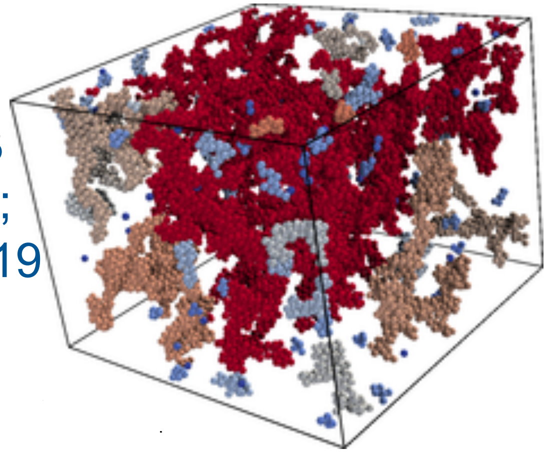
EDG, Stress Localization in Soft Particulate Gels, Encyclopedia of Complexity and Systems Science, Springer 2021

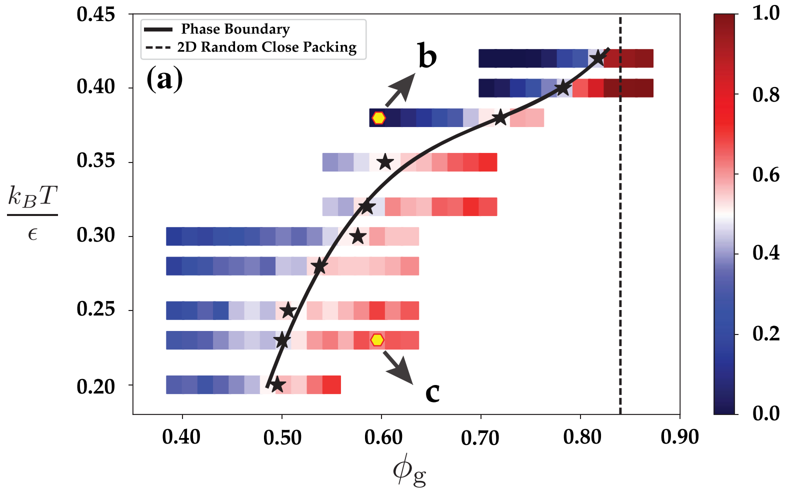
Emergence of rigidity



The correlations due to attractive interactions and associated density fluctuations help develop rigid structures that percolate even at very low volume fractions.

See Valadez-Pérez et al., PRE 2013
Tsurusawa et al., Science Adv. 2019;
K. A. Whitaker et al., Nat. Comm 2019





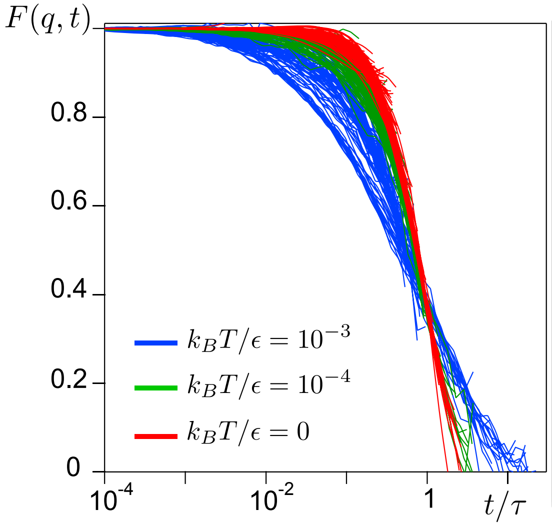
Self-organization of **thin structures** that help **transmit stresses**

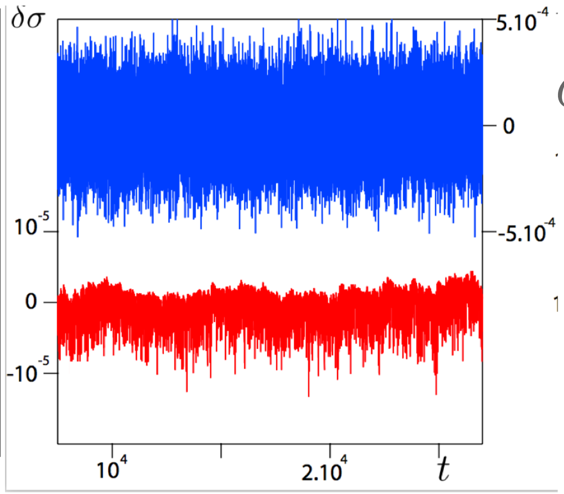
S. Zhang, L. Zhang, M. Bouzid, D. Zeb Rocklin, EDG & X. Mao PRL 2019

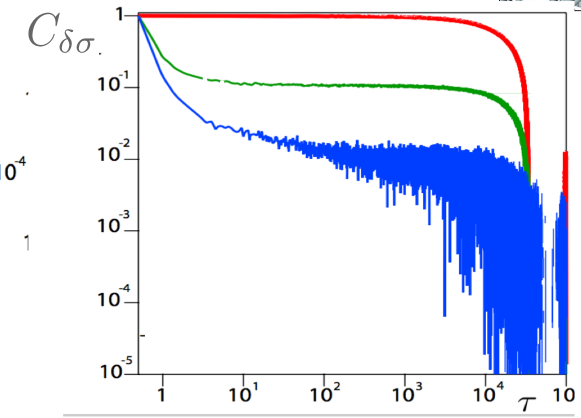
Elastically driven intermittent dynamics

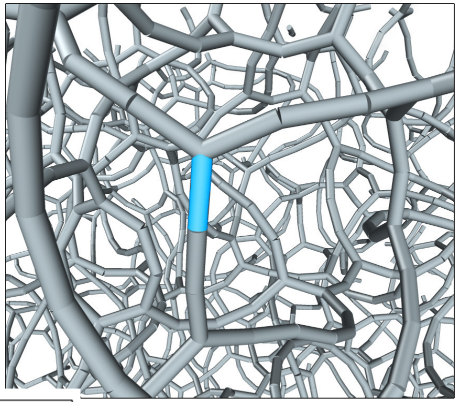
Stress fluctuations controlled by the competition between:

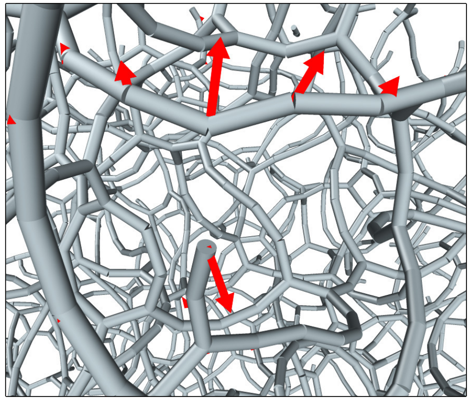
- elastic recovery
- thermal fluctuations









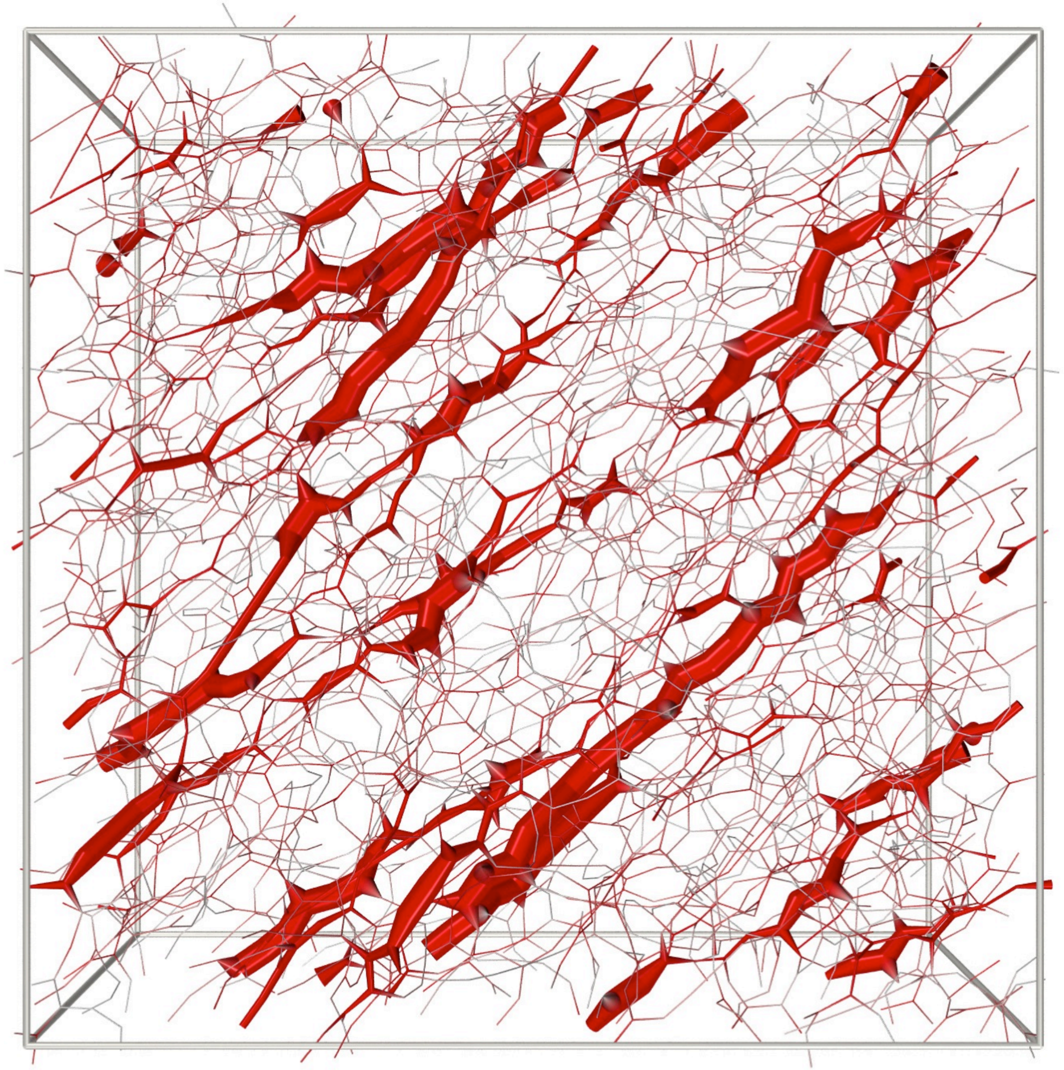


See also Knaebel et al. EPL 2000; Ramos et al. PRL 2001; Bellour et al. PRE 2003; Bandyopadhyay et al. PRL 2004; Duri et al. EPL 2006; Maccarrone et al. Soft Matter 2010; Lieleg et al. Nat. Mat. 2011; Ruta et al. PRL 2012; Angelini et al. Soft Matter 2013 Ruta et al. Soft Matter 2014; Evenson et al. PRL 2015; Gao et al. Soft Matter 2015 ; Helgeson et al. Soft Matter 2015 ...

$k_BT/\varepsilon \approx 0$: stress relaxation tends to occur through elastic recovery. Intermittent dynamics and quasi-ballistic collective motion: **compressed** exponential relaxation.

M. Bouzid, J. Colombo, L. Vieira Barbosa and EDG, Nature Comm. 2017

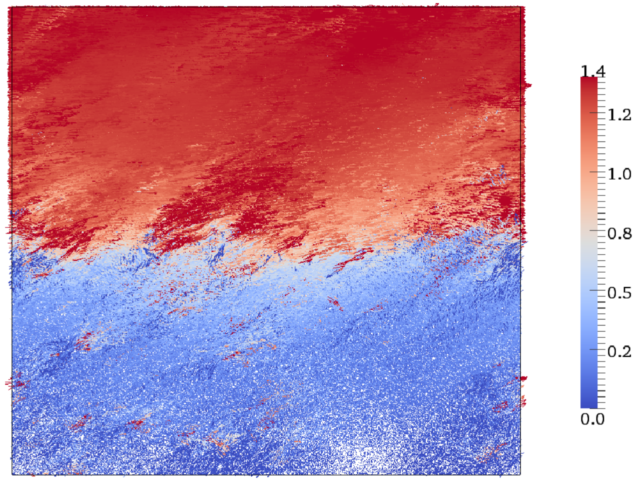
Stress localization, stiffening and damage



Most of the **stress** carried by a small portion of the gel structure. **Local softness** is **topologically controlled** and determines softening, hardening, failure.

See also Burla et al. PNAS 2020

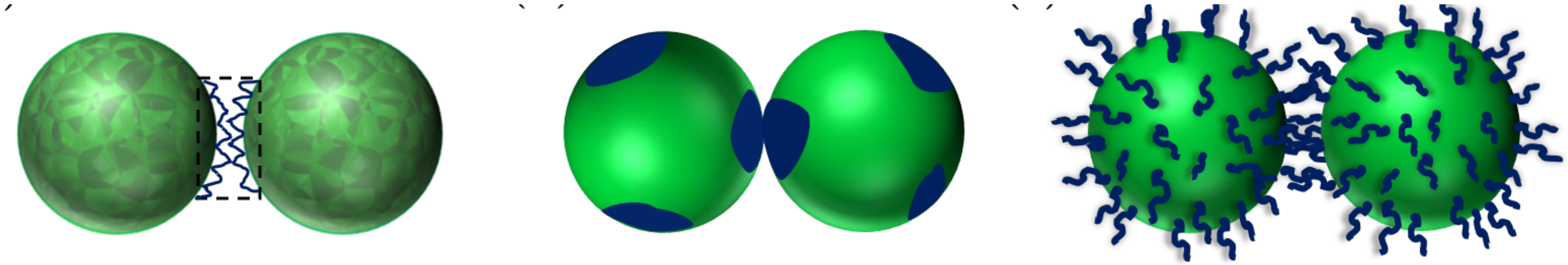
Microscopic precursors and rate-dependent structural reorganization under shear drive switch from ductile to brittle failure.



See also Perge et al. JOR 2014,
Saint-Michel et al. Soft Matter 2017,
Leochmach et al. PRL 2014
Aime, Ramos & Cipelletti JOR 2018, PNAS 2018

J. Colombo and EDG, JOR 2014
M. Bouzid and EDG, Langmuir 2018

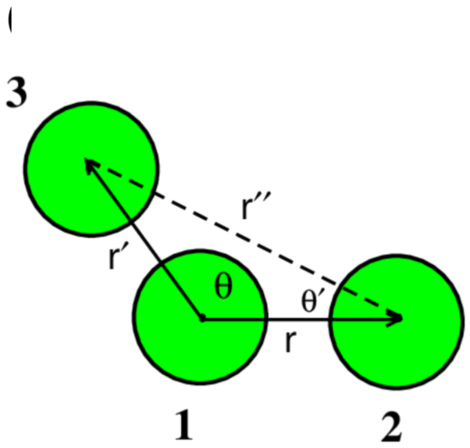
A «fruit fly» model for particulate gels

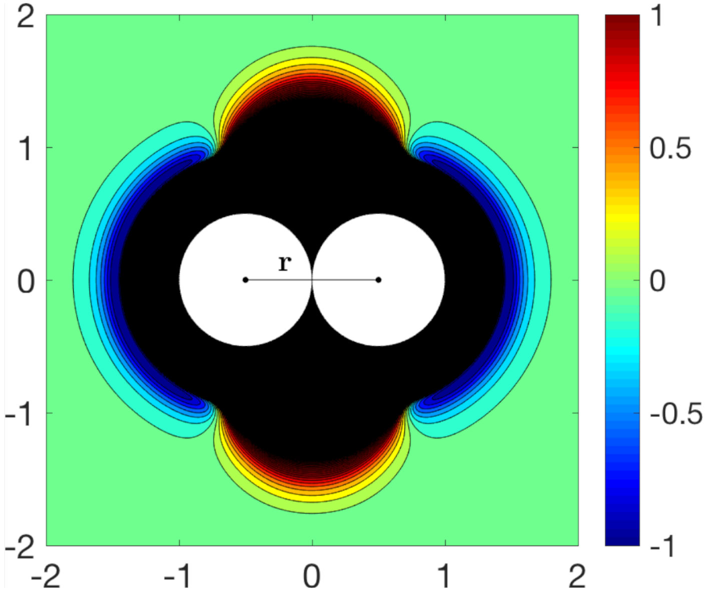


Surface contacts between colloidal units are complex

Pantina & Furst PRL 2004; Dibble et al. PRE 2003; Monti et al. Langmuir 2020; Bonacci et al. Nature Mat 2020; Wu et al. PRL 2020

$$U = \epsilon \left[\sum_{i>j} u_2\left(\frac{\mathbf{r}_{ij}}{d}\right) + \sum_{i} \sum_{j>k}^{j,k\neq i} u_3\left(\frac{\mathbf{r}_{ij}}{d}, \frac{\mathbf{r}_{ik}}{d}\right) \right]$$





Molecular Dynamics
~10^5,10^6 particles
Φ ~ 0.05 - 0.2

$$\sigma_{\alpha\beta} = \frac{1}{V} \sum_{i=1}^{N} \frac{\partial U}{\partial r_i^{\alpha}} r_i^{\beta}$$

J. Colombo, A. Widmer-Cooper and EDG PRL 2013
J. Colombo & EDG, Soft Matter 2014, JOR 2014
M. Bantawa, W. Fontaine-Seiler, P.D. Olmsted and EDG, JPCM 2021

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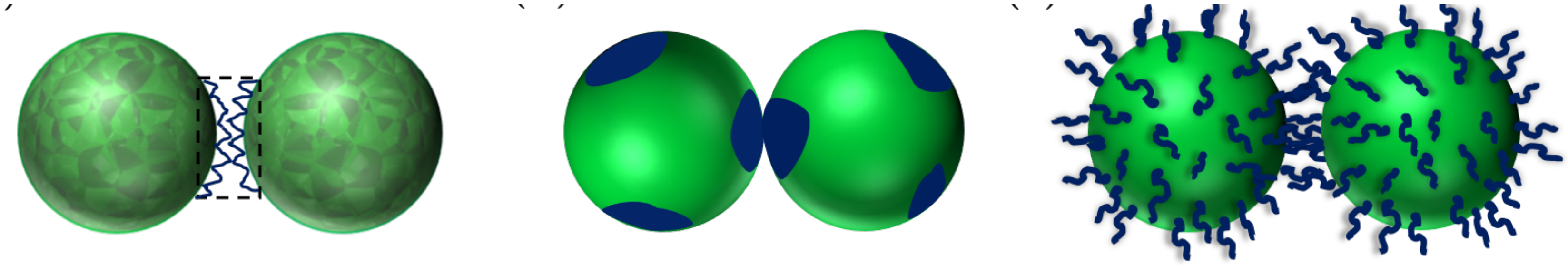

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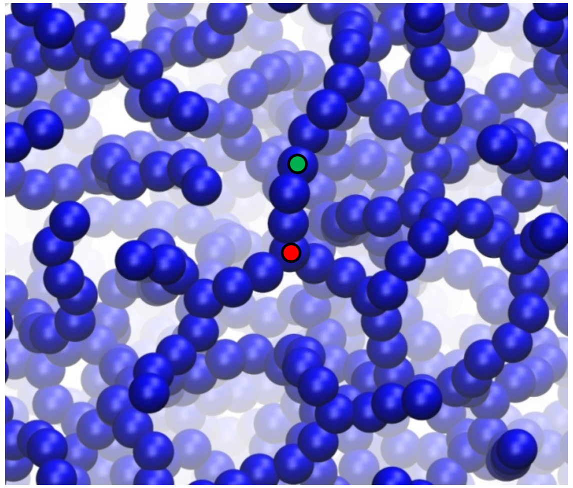
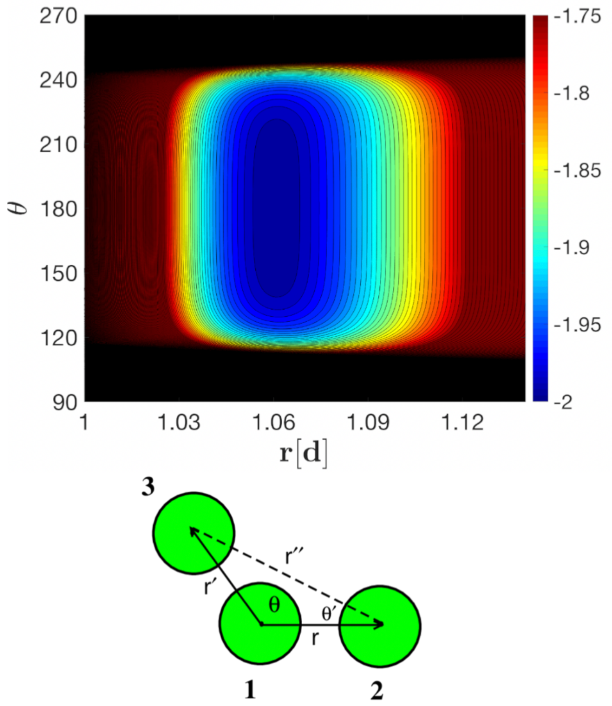
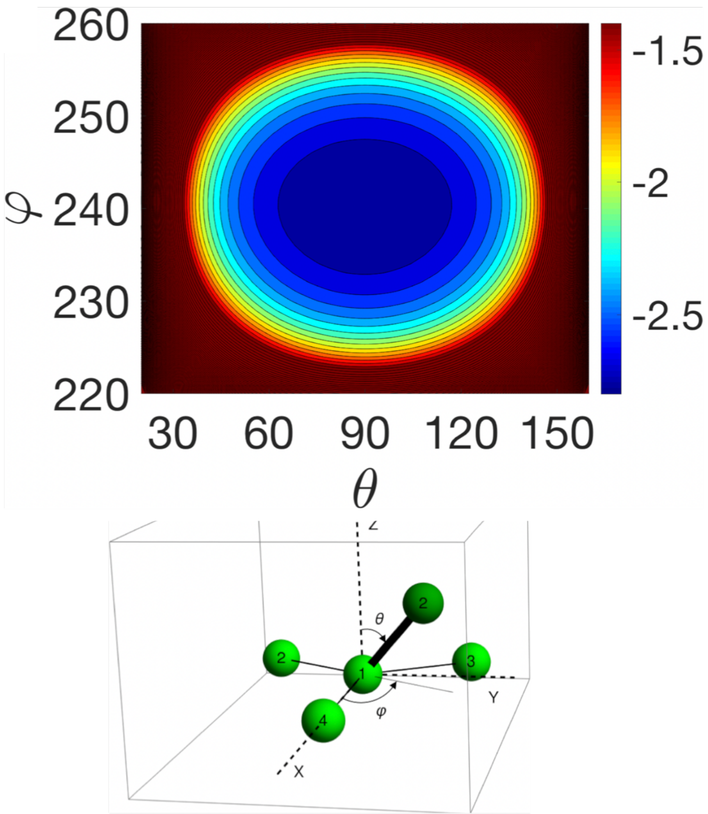


3-coordinated particle (branching point)

2-coordinated particle (strand)





M. Bantawa, W. Fontaine-Seiler, P.D. Olmsted and EDG, JPCM 2021

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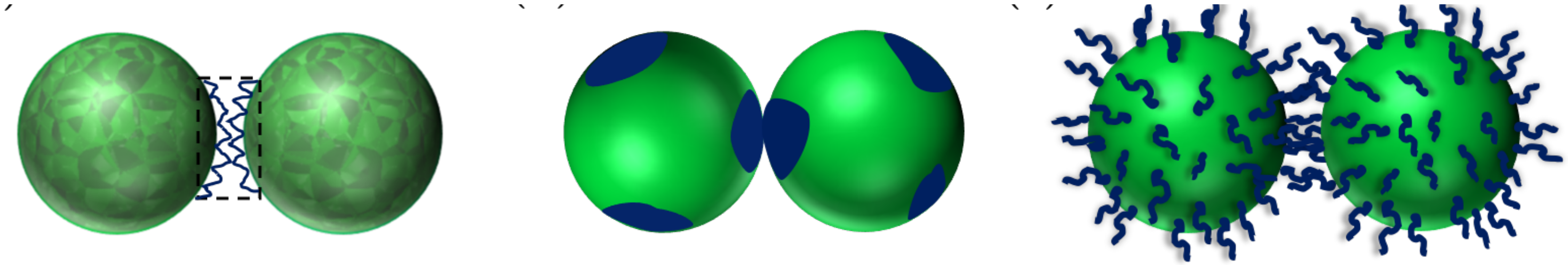

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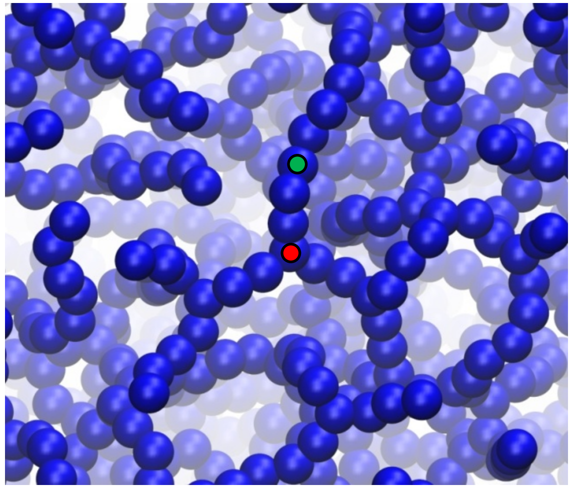
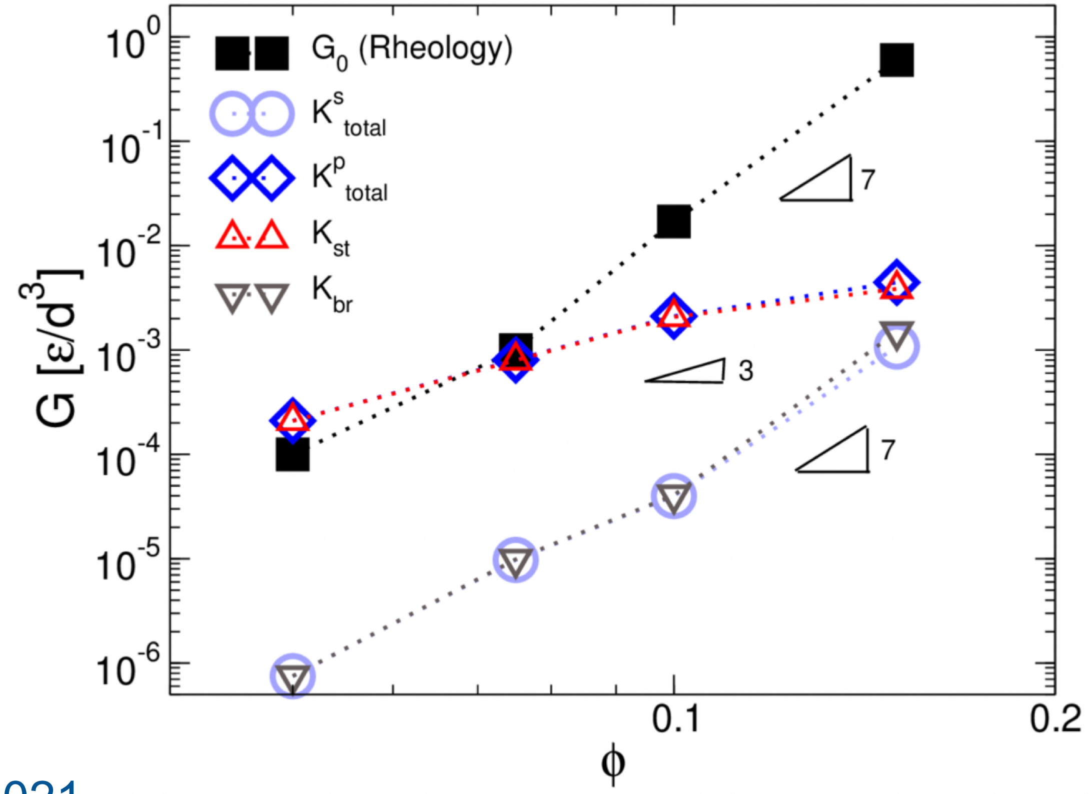


3-coordinated particle (branching point)
2-coordinated particle (strand)

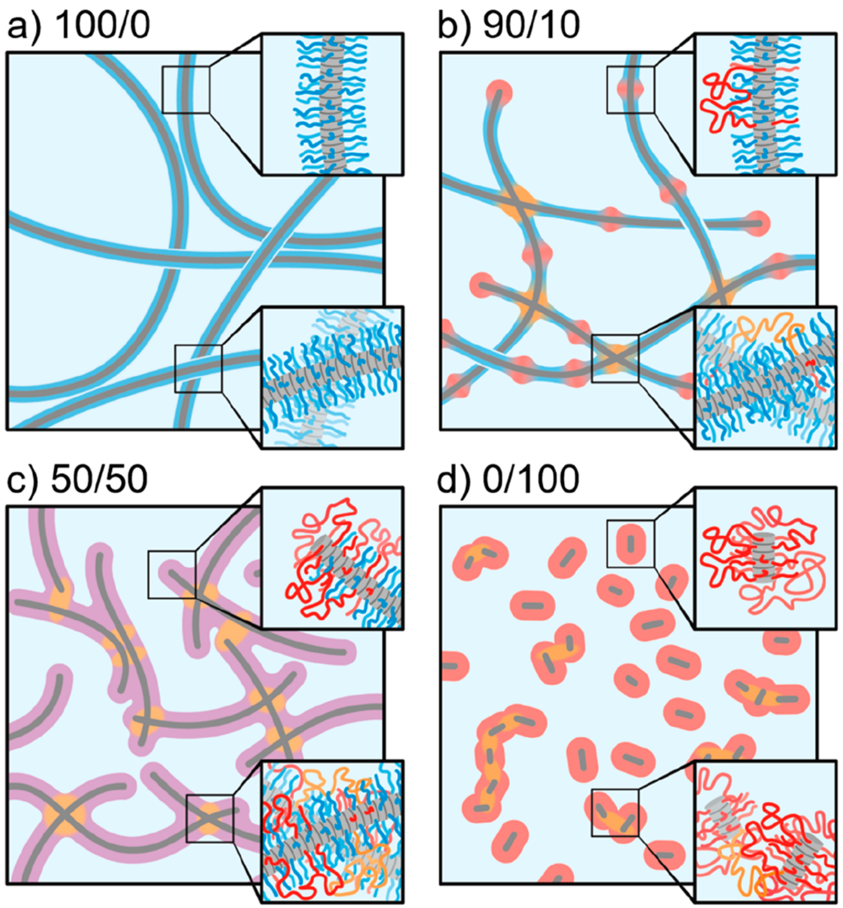
Summing up all contributions of elastic elements as independent (in series or parallel) does not make up for the gel modulus.

Coupling of elastic elements through the network **topology**



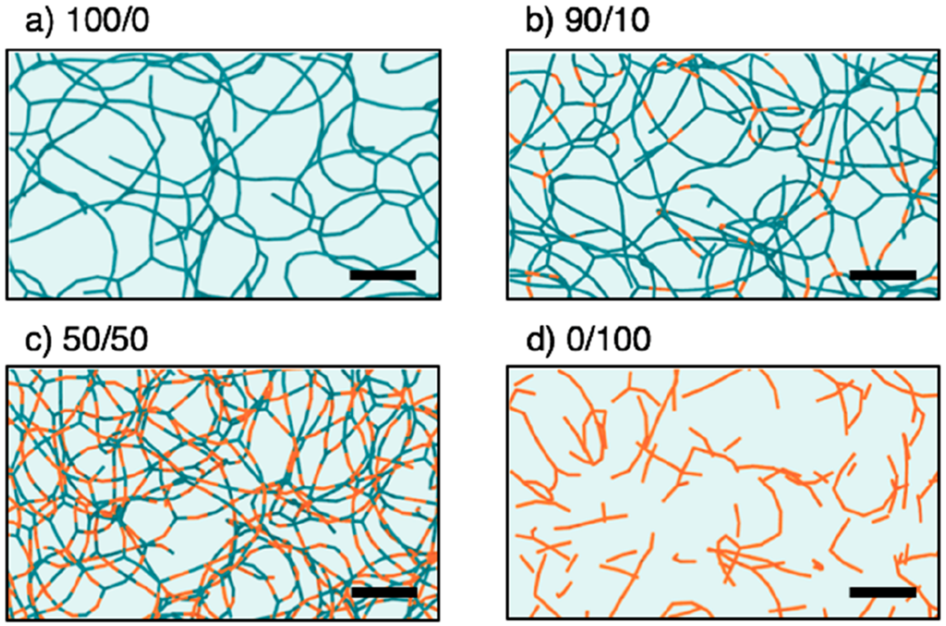
M. Bantawa, W. Fontaine-Seiler, P.D. Olmsted and EDG, JPCM 2021

Topological constraints in binary hydrogels



Space topology set by the network-forming component controls the mechanics of the the composite: non-monotonic dependence of viscoelasticity on composition

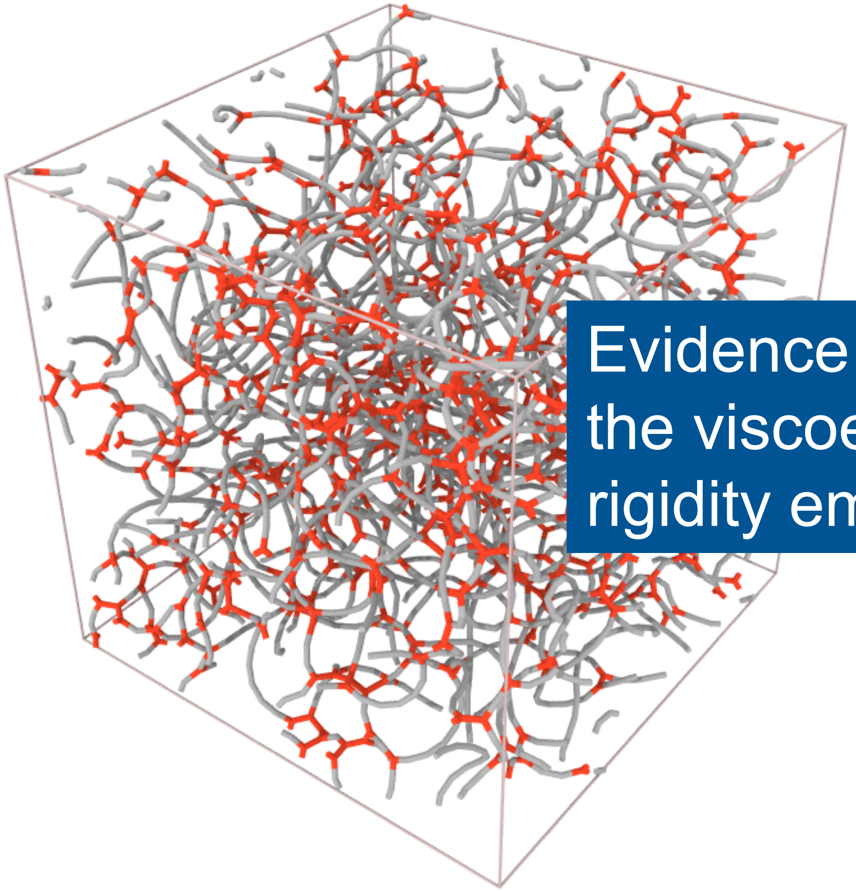
See also Shen et al. PRL 2021

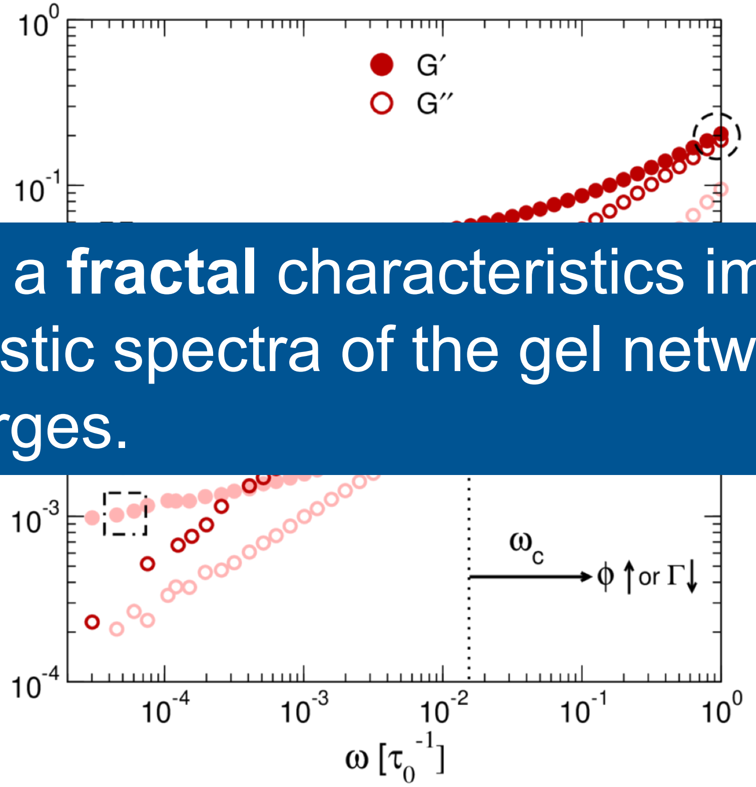


Need to control **topology** through **spatial organization**, not only composition.

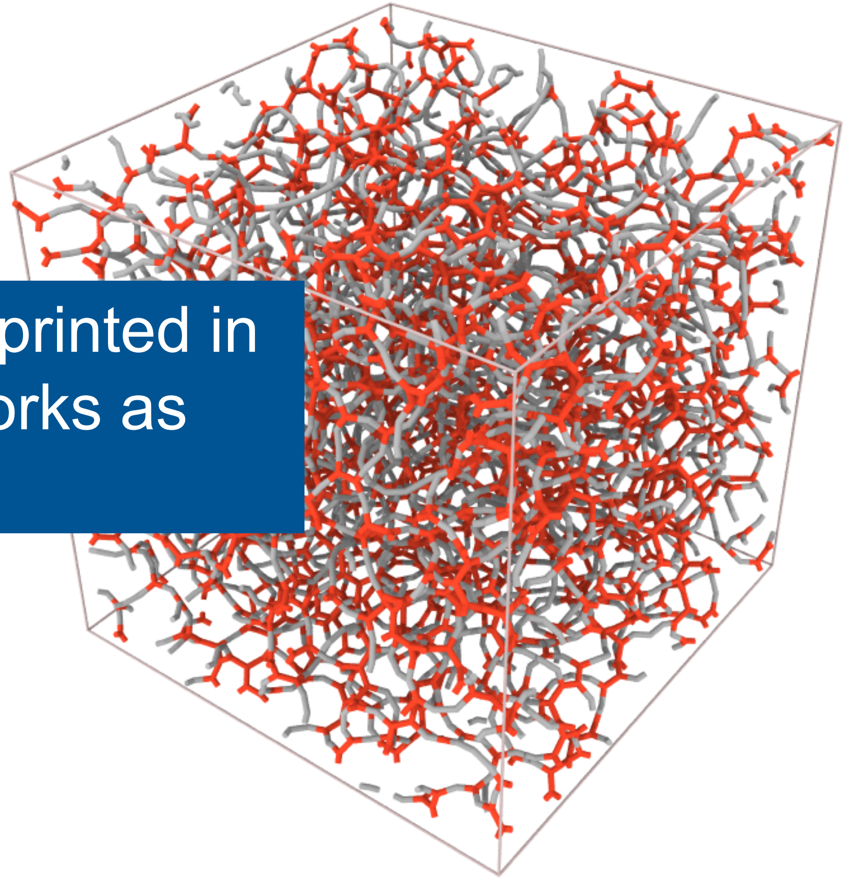
M. Vereroudakis, M. Bantawa, R. Lafleur, D. Parisi, N. Matsumoto, J. Peeters, EDG, EW Meijer and D. Vlassopoulos, ACS central science 2020

Searching for a hidden structural characteristics





Evidence of a **fractal** characteristics imprinted in the viscoelastic spectra of the gel networks as rigidity emerges.



Gel microstructure (pores, connectivity …) depends on the path through which non-ergodicity is attained.

Implications for **viscoelastic response** and stress transmission?

M. Bouzid, B. Keshavarz, M. Geri, T. Divoux, EDG and G. McKinley, JOR 2018
M. Bantawa, B Keshavarz et al., preprint 2021

Conclusions and outlook

- In soft particulate gels, **localized stress patterns** emerge from the mechanical constraints that develop through the network topology as the material solidifies. They control evolution and response of these materials.

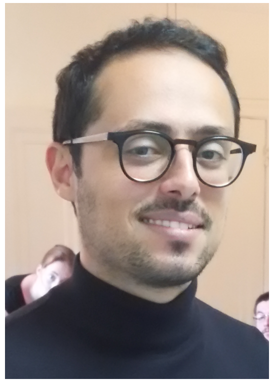
- Understanding the **emergence of rigidity** provides insight into the self-organization of stress transmission through the material and the development of frozen-in stresses. **Connecting localized stress patterns in soft gels to force chains in granular materials* or localized excitations in amorphous solids*.**

- Local morphology, particle volume fractions and single particle properties reveal only part of the story: **viscoelastic spectra** carry information of **fractal characteristics** that govern **stress transmission** and **hierarchy** of time/lenghtscale beyond structural variability.

* See also Nampoothiri et al PRL 2020
Cates et al. PRL 1998

Thanks to

Minaspi Bantawa



Mehdi Bouzid

Bavand Keshavarz

Michela Geri

Gareth McKinley

Thibaut Divoux

Dimitris Vlassopoulos

Shang Zhang
Xiaoming Mao

Zeb Rocklin

Bert Mejier

Peter D. Olmsted

Bulbul Chakraborty