Glasses and Gels

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Boulder summer school 7/5/06

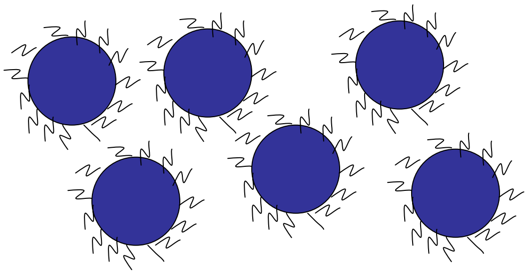
- •General introduction
- •Experimental techniques
- •Colloidal glasses
- •Colloid Aggregation
- •Colloidal gels

http://www.deas.harvard.edu/projects/weitzlab

Colloidal particles

- Good model system
  - Control size, shape
  - Control strength, range of interactions
- Statistical properties of phase behavior
- Model for complex fluid systems
- Equilibrium properties
- Non-equilibrium properties (often dominate)

Colloidal Particles



Stability:

Short range repulsion

Sometimes a slight charge

Colloid Particles are:

•Big

•~ a ~ 1 micron

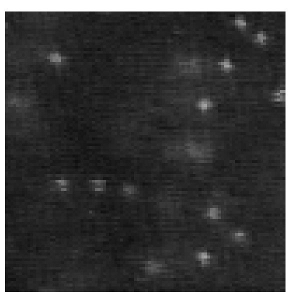
•Can "see" them

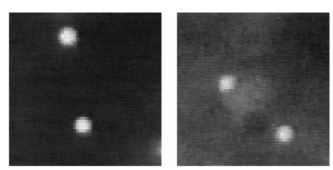
Slow

• $\tau \sim a^2/D$ ~ ms to sec

•Follow individual particle dynamics

Colloidal particles undergoing Brownian motion





Thermal motion ensures particles are always equilibrated with the fluid

They can explore phase space

Soft Materials

Easily deformable → Low Elastic Constant:

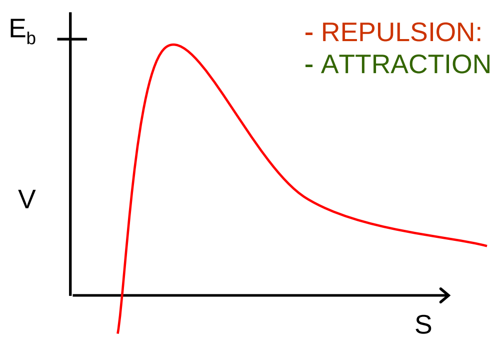
Elastic Constant: → Pressure $\frac{\text{Force}}{\text{Area}}$ $\frac{\text{Energy}}{\text{Volume}}$

Atoms: $\frac{eV}{\dot{A}^3}$ ~GPa

Colloids: $\frac{k_B T}{\mu m^3}$ ~Pa

Colloidal interactions – stabilizing

- REPULSION: Coulomb; Steric
- ATTRACTION: van der Waals; Chemical



- Short range attraction
- Long range repulsion
- Sum: Stabilizing barrier

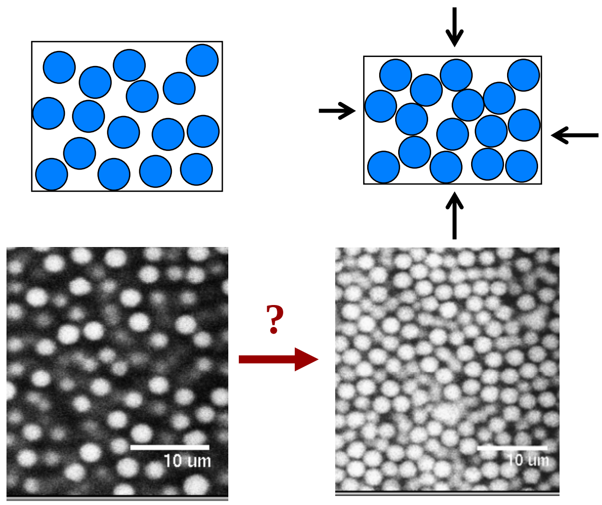
$E_b > k_B T$ → Colloid stable against aggregation

OTHER POSSIBLE REPULSIVE INTERACTIONS

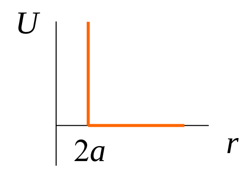
Hard Spheres: Only Volume Exclusion

Volume Fraction Controls Phase Behavior

Increase ϕ => Decrease Tempreature

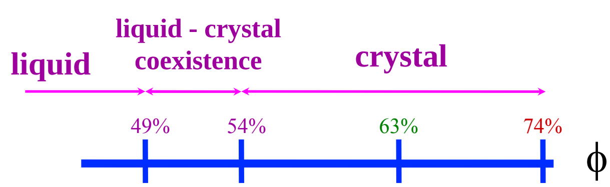


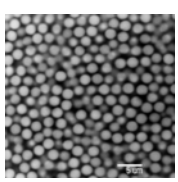
$F = \not{U}^{\,0} - TS$

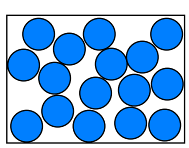


Hard Sphere

Hard Sphere Phase Diagram

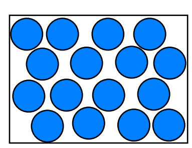






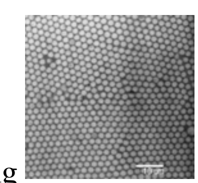
maximum packing

$\phi_{RCP} \approx 0.63$



maximum packing

$\phi_{HCP} = 0.74$

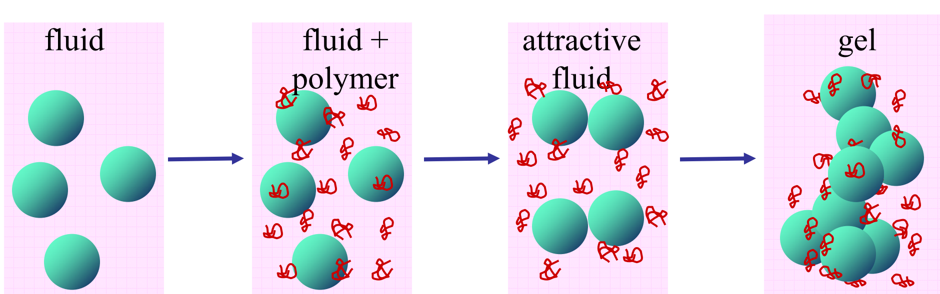


Colloidal interactions – destabilizing

- Add electrolyte → screen repulsion
- Reduce steric repulsion
- Always have van der Waals attraction
  - Colloids are inherently unstable
- Can also induce controlled attraction

Controlled Attraction of Colloidal Particles

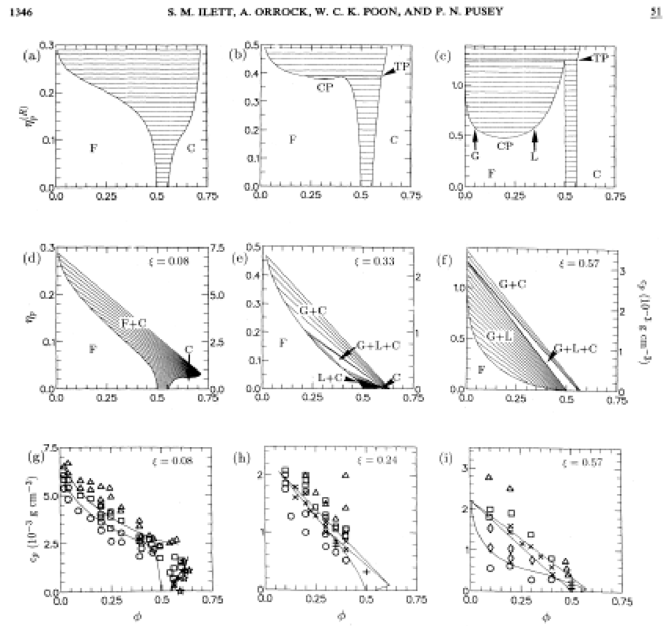
Depletion attraction



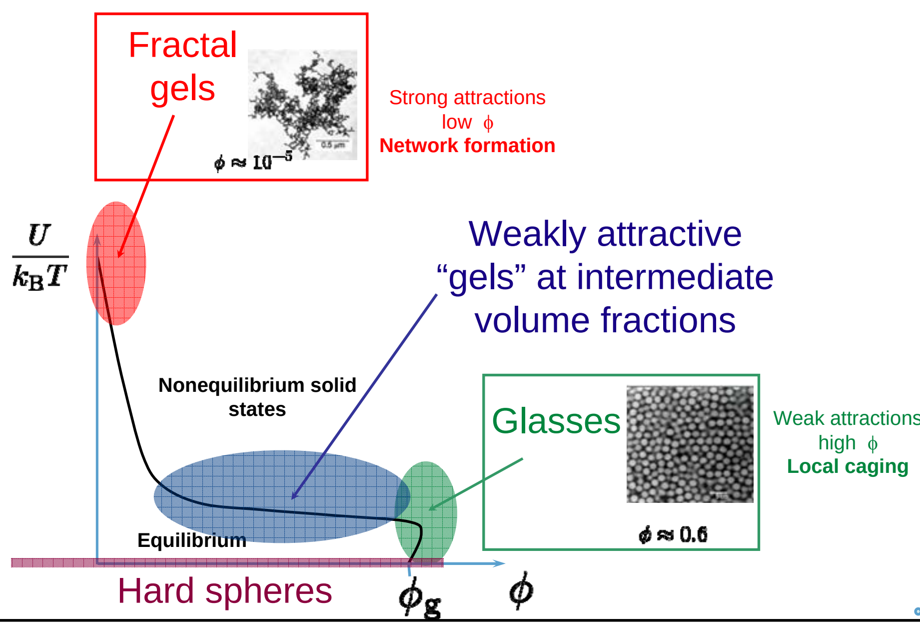
Polystyrene polymer, R_g=37 nm + PMMA spheres, r_c=350 nm

T. Dinsmore

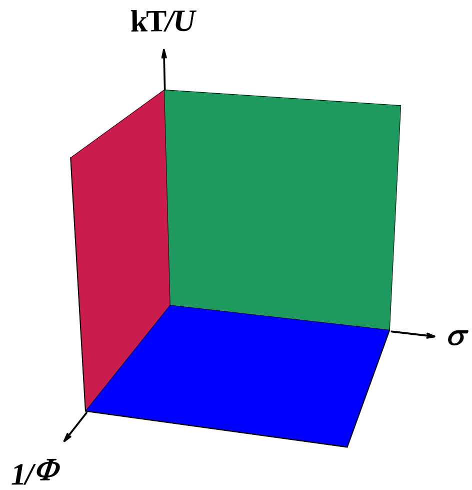
Phase diagram for attractive colloids



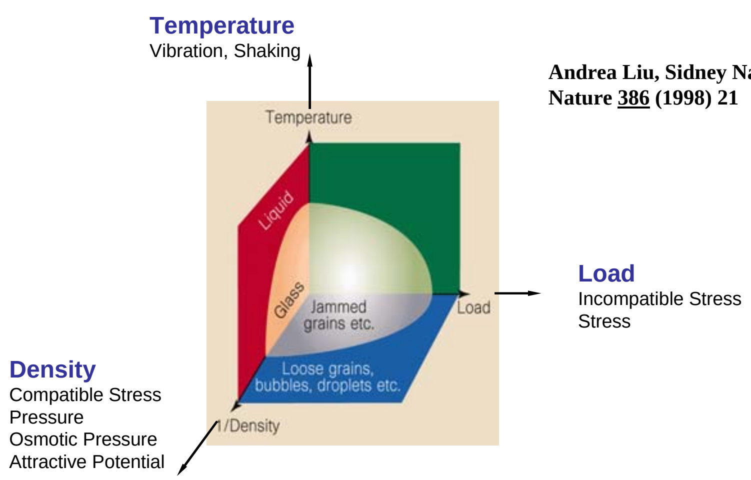
State diagram for colloidal particles



Jamming Transitions for Colloidal Systems with Attractive Interactions

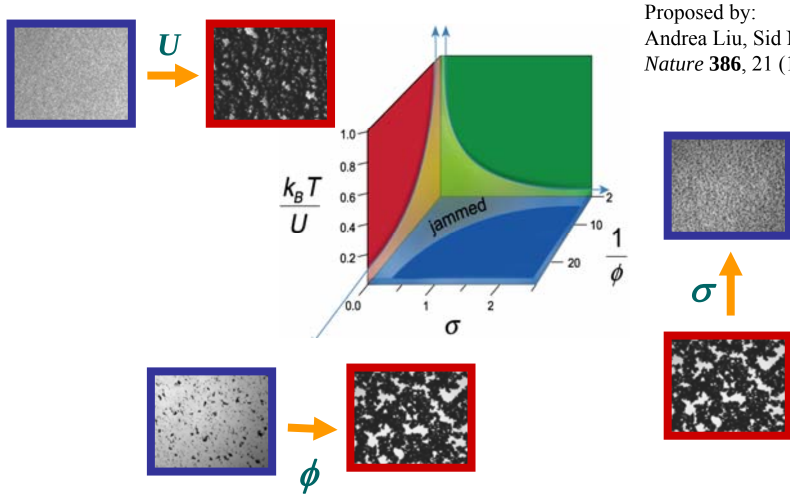


Jamming Transition – Arrest of Motion



Andrea Liu, Sidney Nagel
Nature 386 (1998) 21

Jamming Phase Diagram for Attractive Systems



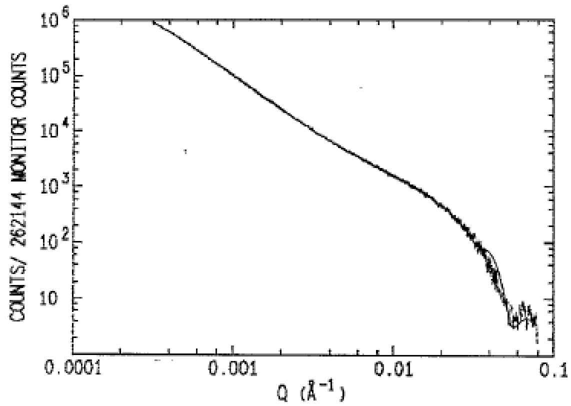
Proposed by:
Andrea Liu, Sid Nagel
Nature **386**, 21 (1998)

Trappe *et al*, *Nature* **411**, 772 (2001).

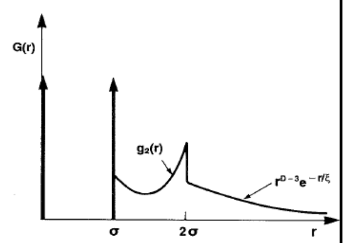
Experimental Techniques

- Light scattering
  - Static light scattering
  - Dynamic light scattering
  - Ultra-small angle dynamic light scattering
  - Diffusing-wave spectroscopy
- Microscopy
- Rheology

Static Light Scattering



X-ray scattering from gold aggregates



Model g(r)

P. Dimon, SofK Sinha

Static Scattering: Structure

Scattered field from single particle

$$E_m(q) = A_m e^{i\vec{q}\cdot\vec{r}_m} e^{-i\omega t}$$

Scattered intensity per particle — Phase factor — Frequency of light

Measure the scattered intensity from collection of particles

$$I(q) = \sum_{m,n} E_m E_n^* = \sum_{m,n} A_m A_n e^{i\vec{q}\cdot(\vec{r}_m - \vec{r}_n)} = P(q)S(q)$$

Form factor — Structure factor

- Measure q dependence of scattering
- Probes spatial Fourier Transform of density correlations

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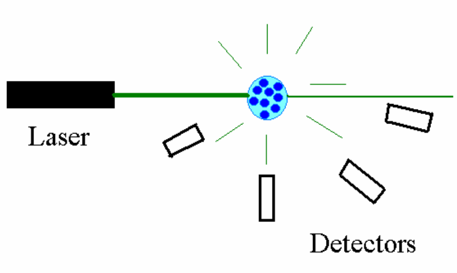
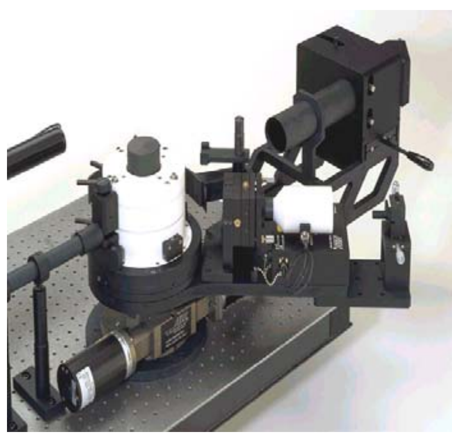
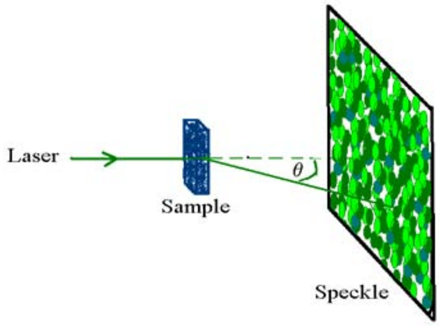
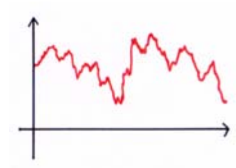
Dynamic Light Scattering





Structure and Dyanmics: Light Scattering



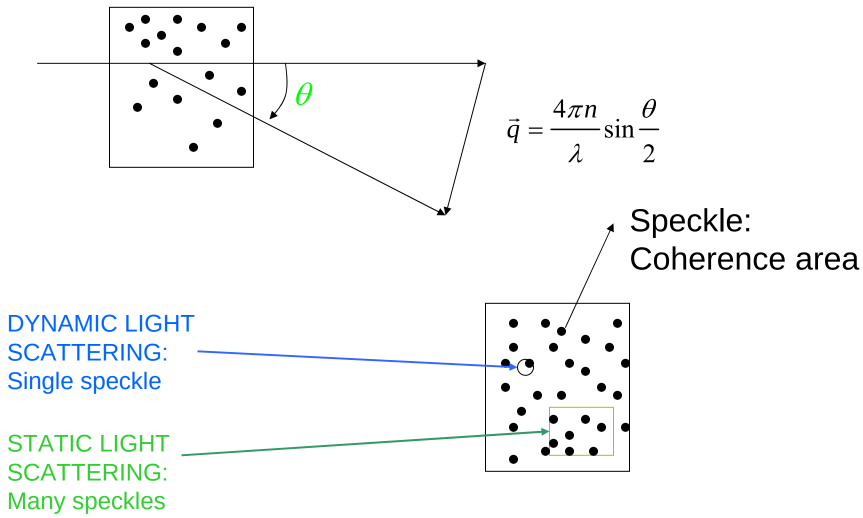


$\xi = q^{-1}$ $q = 4\pi n/\lambda \sin\theta/2$

- SLS: $< I >$ vs. q ⟶ probe structure
- DLS: $< I(q,t)I(q,t+\tau) >$ ⟶ probe dynamics $f(q,\tau)$

Light Scattering

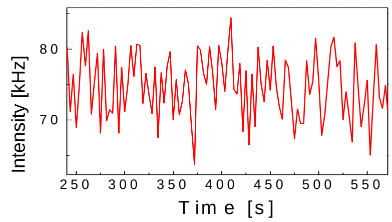
Probes characteristic sizes of colloidal particles



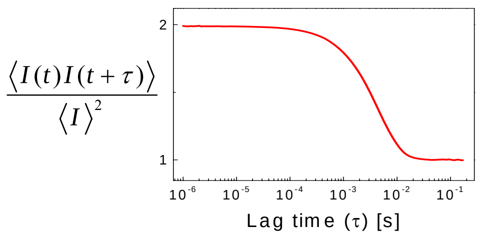
$$\vec{q} = \frac{4\pi n}{\lambda}\sin\frac{\theta}{2}$$

Dynamic Light Scattering

Measure temporal intensity fluctuations



Obtain an intensity autocorrelation function



Dynamic Light Scattering

Measure temporal correlation function of scattered light:
Intermediate structure factor

$$f(q,t) \sim \langle E(0)E(t)\rangle$$

$$\langle E(0)E(t)\rangle = \left\langle A^2 \sum_{m,n} e^{i\vec{q}\cdot\{\vec{r}_m(0)-\vec{r}_n(t)\}} \right\rangle$$

Time average over all particles

$$\sim e^{-q^2\langle \Delta r^2(t)\rangle}$$

Correlations only between the same particles
Cumulant expansion: $\Delta r^2(t) \sim Dt$

$$\sim e^{-q^2 Dt}$$

Physics: How to change the phase of the field by π
Each particle must move by $\sim\lambda$

Experimental Techniques

- Light scattering
  - Static light scattering
  - Dynamic light scattering
  - Ultra-small angle dynamic light scattering
  - Diffusing-wave spectroscopy
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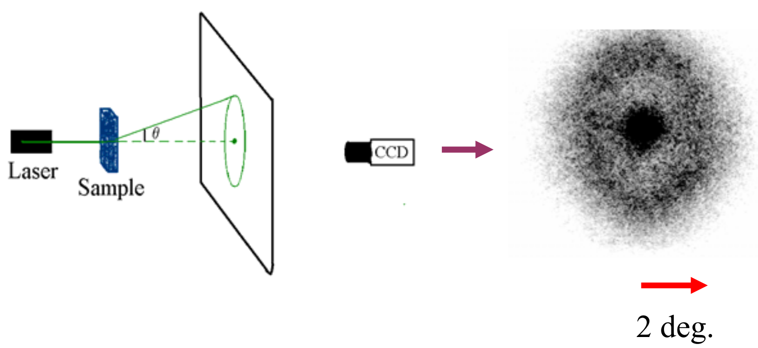
Ultra Small Angle Light Scattering

Probe Structure

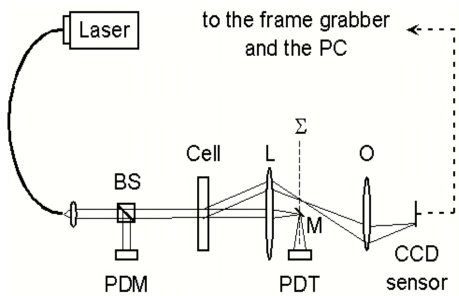


L. Cipelletti

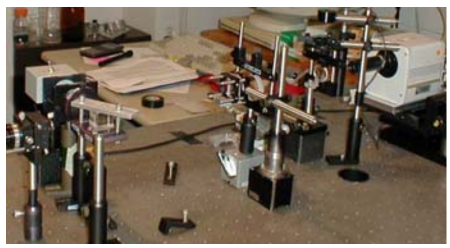
Experimental setup

Luca Cipelletti
Montpellier

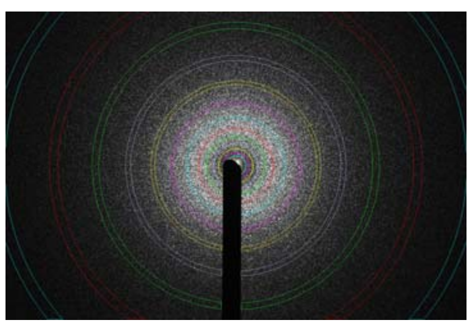
Suliana Manley
Harvard



0.07 deg to 5.0 deg



Multispeckle Detection



0.07 deg to 5.0 deg
$100\ cm^{-1} < q < 7000\ cm^{-1}$

Average over constant *q*:

- non-ergodic samples
- avoid excessive time averaging

Experimental Techniques

- Light scattering
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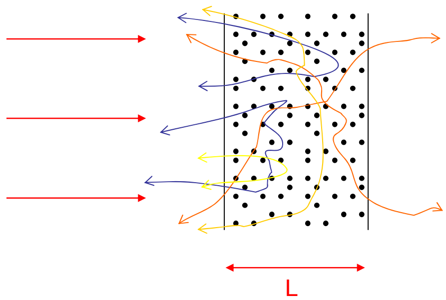
  - Diffusing-wave spectroscopy
- Microscopy
- Rheology

Diffusing Wave Spectroscopy: Very strong scattering

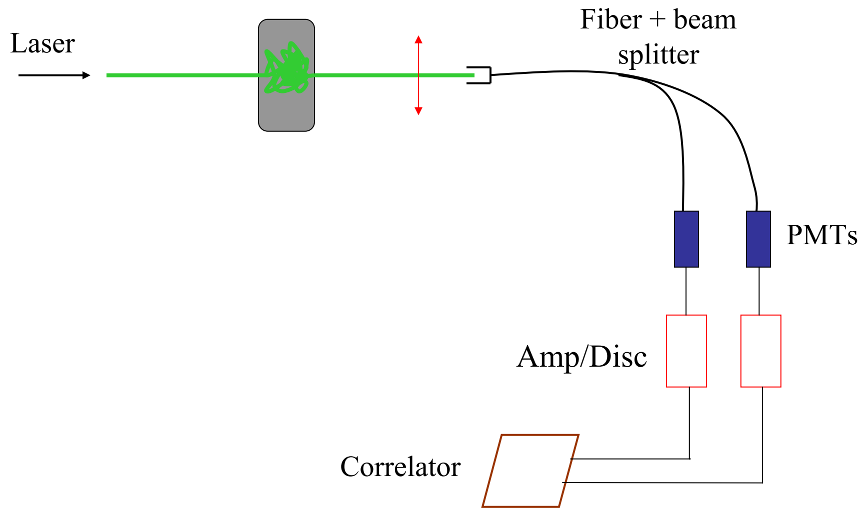
TRANSMISSION



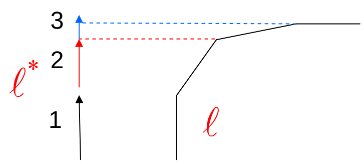
MOST LIGHT IS SCATTERED BACK
→ MILK IS WHITE!!

D. Pine, P. Chaikin, E. Herbolzheimer

Diffusing Wave Spectroscopy



TRANSPORT MEAN FREE PATH



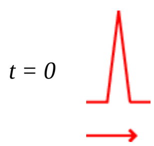
PROJECT LENGTHS ONTO INITIAL DIRECTION

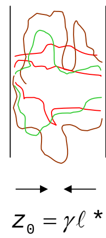
$$\ell^* = \sum_i \ell \langle \cos\theta \rangle^i$$

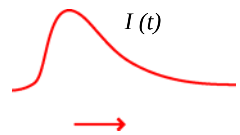
$$\ell^* = \frac{\ell}{1 - \langle \cos\theta \rangle} > \ell$$

ANALOGY – PERSISTENCE LENGTH FOR SEMI-RIGID POLYMER

P(s): DIFFUSION EQUATION







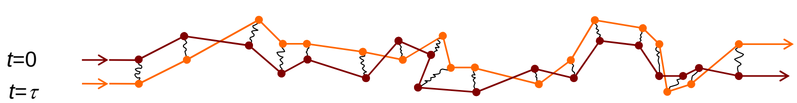
PRL **60**, 1134 (88)

I(t) ~ # PATHS OF LENGTH $s = ct$

⟶ **P(s)**

SINGLE PATH

[MARET & WOLFE]



n **SCATTERING EVENTS**

$s = n\ell$ → PATH LENGTH

$$g_1^n(\tau) = \left\langle e^{i\sum_{j=1}^{n} \vec{q}\cdot\Delta\vec{r}(\tau)} \right\rangle$$

PHASE CHANGE DUE TO **TOTAL** PATH LENGTH

$$= \left\langle e^{i\vec{q}\cdot\Delta\vec{r}(\tau)} \right\rangle_q^n$$

STATISTICAL APPROACH

n LARGE → REPLACE BY AVERAGE
DON'T CONSERVE q AT EACH SCATTERING

$$= e^{-\langle q^2\rangle_q \left\langle \frac{\Delta r^2(\tau)}{6} \right\rangle n}$$

→ APPROX. AT SHORT τ

$$= e^{-2k_0^2 \frac{\ell}{\ell^*} \frac{\langle \Delta r^2(\tau)\rangle}{6} \frac{s}{\ell}}$$

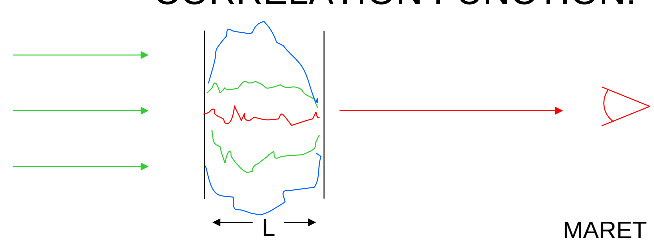
$$\langle q^2 \rangle = 2k_0^2 \frac{\ell}{\ell^*}$$

$$s = n/\ell$$

$$g_1^n(\tau) = e^{-2\left(\tau/\tau_0\right)\left(s/\ell^*\right)}$$

$$\tau_0 = \frac{1}{k_0^2 D}$$

CORRELATION FUNCTION:



MARET & WOLF

SUM OVER DISTRIBUTION OF LIGHT PATHS.

$$G_1(\tau) = \int_0^\infty P(s)\, e^{-2(\tau/\tau_0)(s/\ell^*)} ds$$

$P(s)$ DISTRIBUTION OF PATHS OF LENGTH S
→ DEPENDS ON GEOMETRY
→ DIFFUSION OF EQUATION FOR LIGHT

SINGLE SCATTERING DECAY

NUMBER OF (RANDOMIZING) SCATTERING EVENTS

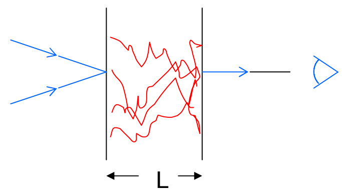
TRANSMISSION THROUGH A SLAB

FUNCTIONAL FORMS ARE KNOWN

-- USE MORE EXACT BOUNDARY CONDITIONS

-- J. de Phys. **51**, 2101 (1990)

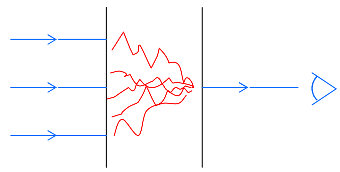
POINT SOURCE:



$$g_1(t) = \int_{\frac{L}{\ell^*}\sqrt{\frac{6t}{\tau_0}}}^{\infty} \left[A(s)\sinh s + e^{-s\left(1-\frac{4}{3}\,\ell^*/L\right)} \right] ds$$

with $$A(s) = \frac{\left(\frac{2}{3}\frac{\ell^*}{L}s - 1\right)\left[\frac{2}{3}\frac{\ell^*}{L}e^{-\frac{4}{3}\frac{s\ell^*}{L}} + \left(\sinh s + \frac{2}{3}\frac{\ell^* s}{L}\cosh s\right)e^{-s\left(1-\frac{4}{3}\frac{\ell^*}{L}\right)}\right]}{\left(\sinh s + \frac{2}{3}\frac{\ell^*}{L}s\cosh s\right)^2 - \left(\frac{2}{3}\frac{\ell^*}{L}s\right)^2}$$

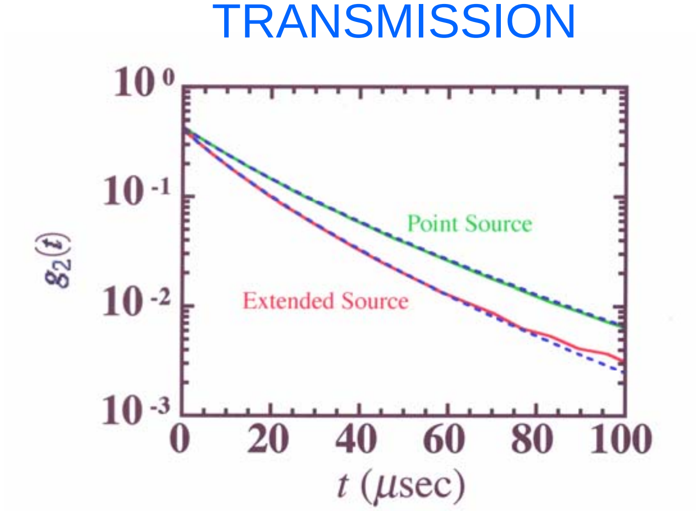
EXTENDED SOURCE



$$g_1(t) \approx \frac{\left(\frac{L}{\ell^*}+\frac{4}{3}\right)\sqrt{\frac{6t}{\tau_0}}}{\left(1+\frac{8t}{3\tau_0}\right)\sinh\left[\frac{L}{\ell^*}\sqrt{\frac{6t}{\tau_0}}+\frac{4}{3}\sqrt{\frac{6t}{\tau_0}}\cosh\left[\frac{L}{\ell^*}\sqrt{\frac{6t}{\tau_0}}\right]\right]}$$

CHARACTERISTIC TIME SCALE: $\tau_0\left(\frac{\ell^*}{L}\right)^2$

TRANSMISSION



$d = 0.6\mu\text{m}$

$\phi = 2\%$

PHYSICS

-- TOTAL PATH LENGTH CHANGES BY λ

-- CONTRIBUTIONS FROM *MANY* SCATTERING EVENTS

SMALL τ → LONG PATH → MANY SCATTERERS

→ SMALL MOTION → FAST DECAY

LARGE τ → SHORT PATH → FEW SCATTERERS

→ LARGER MOTION → SLOWER DECAY

DWS PROBES MOTION ON *SHORT* LENGTH SCALES

PHASE OF PATH CHANGES WHEN PATH LENGTH CHANGES BY ~1 WAVELENGTH

$$\lambda \sim 5000 \text{ Å}$$

BUT: LIGHT IS SCATTERED FROM MANY PARTICLES

$$\left(\text{Estimate:} \quad \left(\frac{L}{\ell^*}\right)^2 \approx \left(\frac{10^3}{10}\right)^2 \approx 10^4\right)$$

∴ MOTION OF EACH **INDIVIDUAL** PARTICLE CAN BE **MUCH** LESS

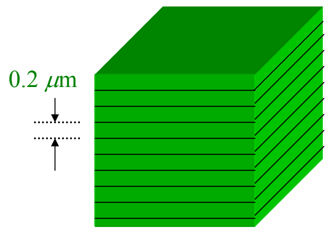
→ CAN MEASURE PARTICLE MOTION ON SCALE OF ~ 5 Å

Experimental Techniques

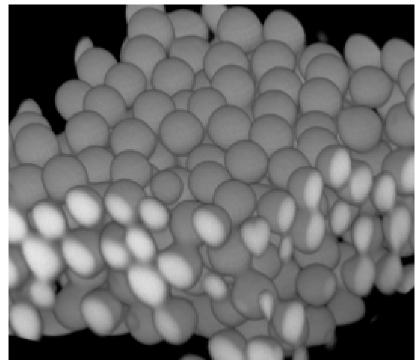
- Light scattering
  - Static light scattering
  - Dynamic light scattering
  - Ultra-small angle dynamic light scattering
  - Diffusing-wave spectroscopy
- Microscopy
- Rheology

The "PINK Monster"
Keep that door closed!!!!

Confocal microscopy for 3D pictures

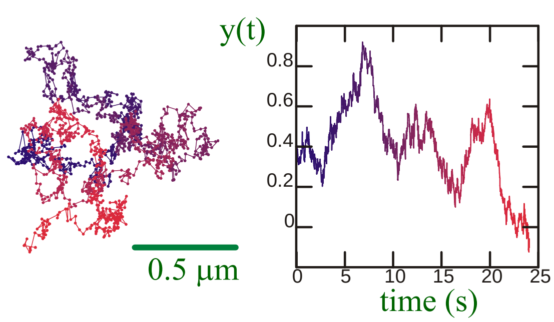


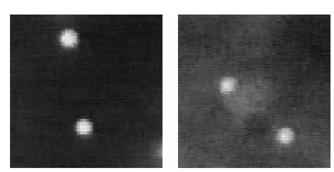
Scan many slices, reconstruct 3D image



Brownian Motion

(2 μm particles, **dilute** sample)





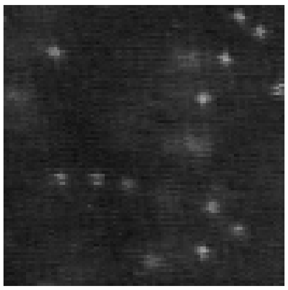
Leads to normal diffusion: $\langle \Delta x^2 \rangle = 2Dt$

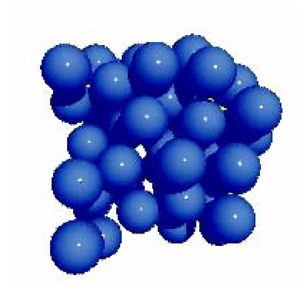
$$D = \frac{k_B T}{6\pi\eta a}$$

Particle size a

viscosity η

Brownian Motion in Real Time





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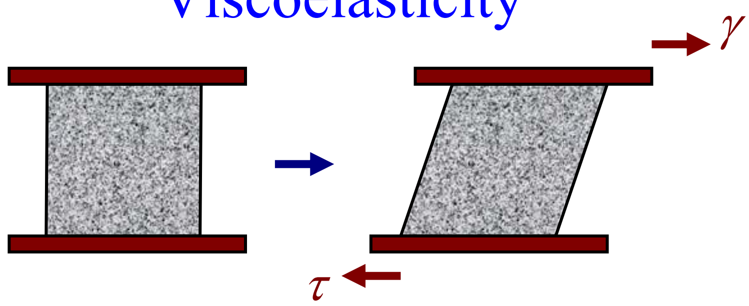
  - Static light scattering
  - Dynamic light scattering
  - Ultra-small angle dynamic light scattering
  - Diffusing-wave spectroscopy
- Microscopy
- Rheology

Mechanical Properties of Soft Materials: Viscoelasticity



Solid: $\tau = G\gamma$

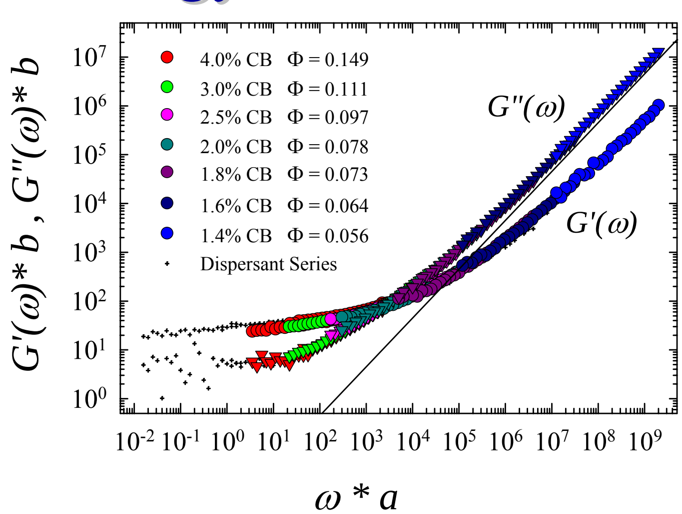
Fluid: $\tau = \eta\dot{\gamma}$

$$\gamma = \gamma_0 e^{i\omega t}$$

$$\tau = \left[G'(\omega) + iG''(\omega)\right]\gamma$$

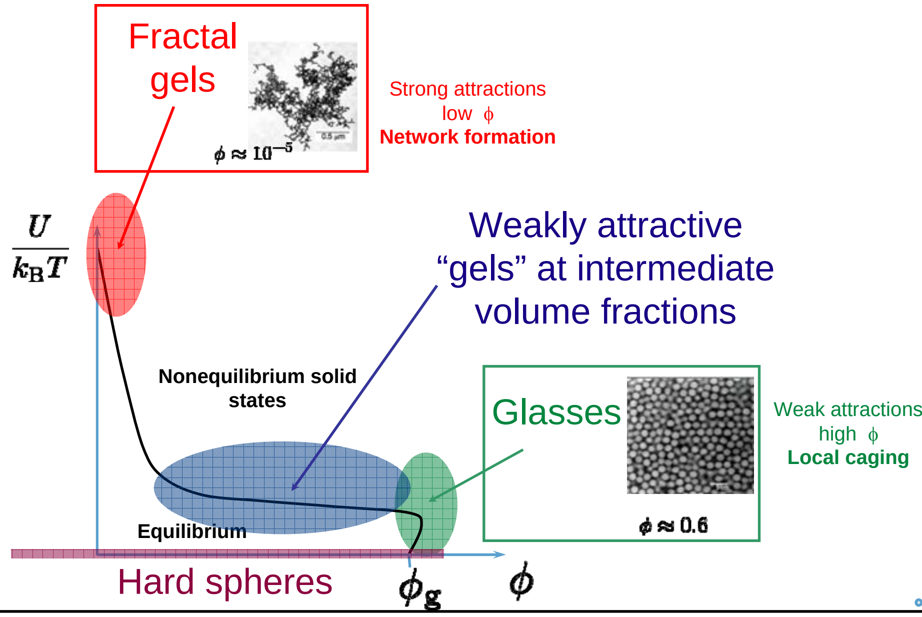
Elastic: $G'(\omega)$ — Viscous: $G''(\omega)$

Rheology of soft materials



Scaling plot

State diagram for colloidal particles



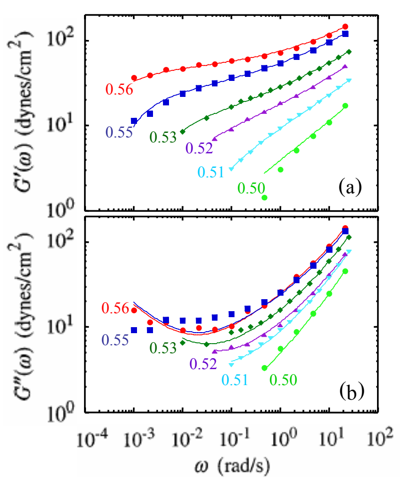
Storage and Loss Modulii of Hard Spheres

Storage

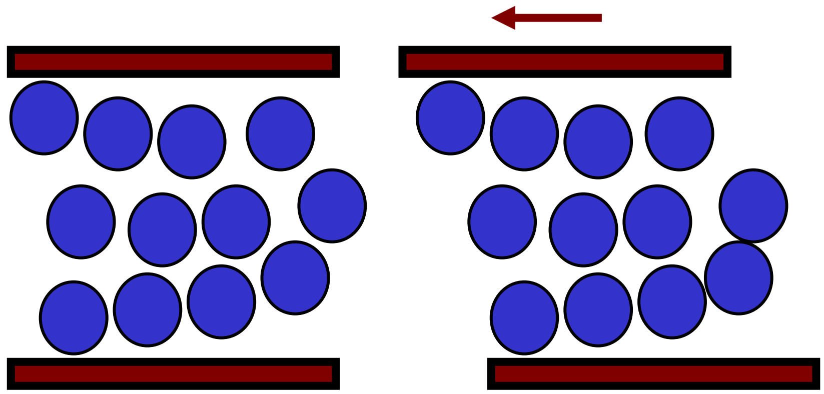
$G'(\omega)$

Loss

$G''(\omega)$

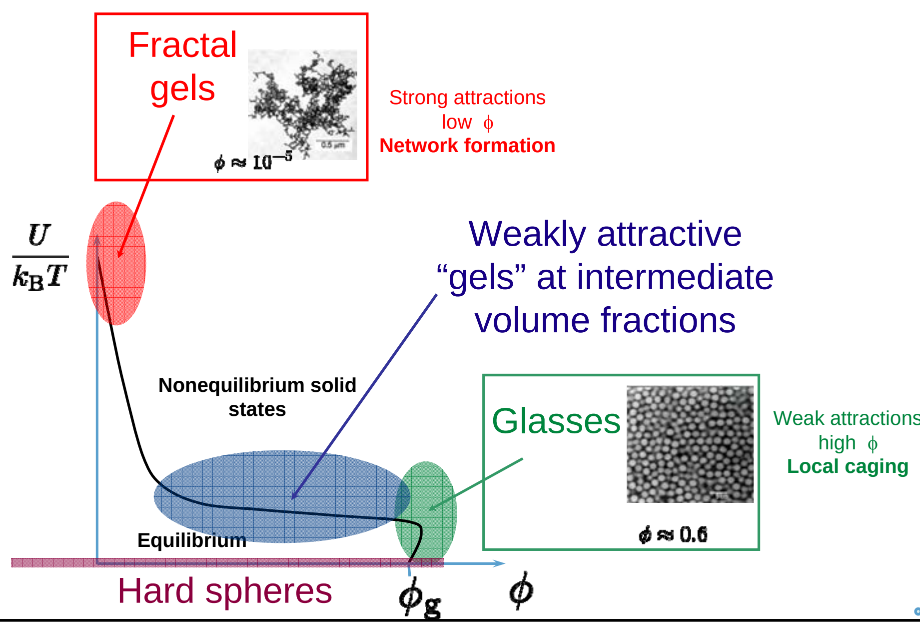


Origin of Elasticity

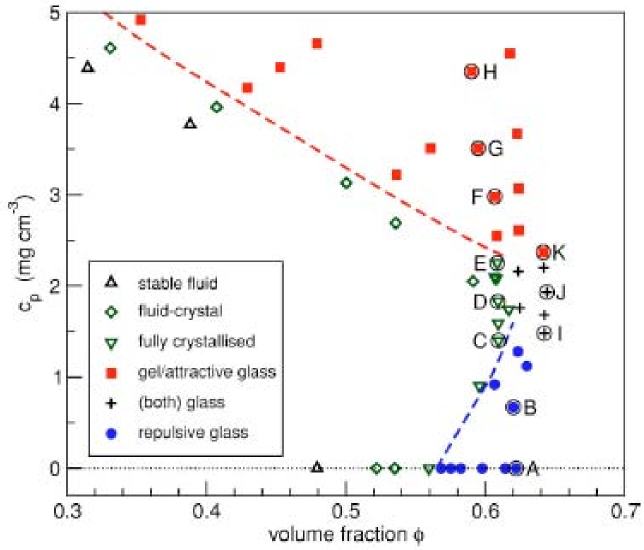


Distort equilibrium positions

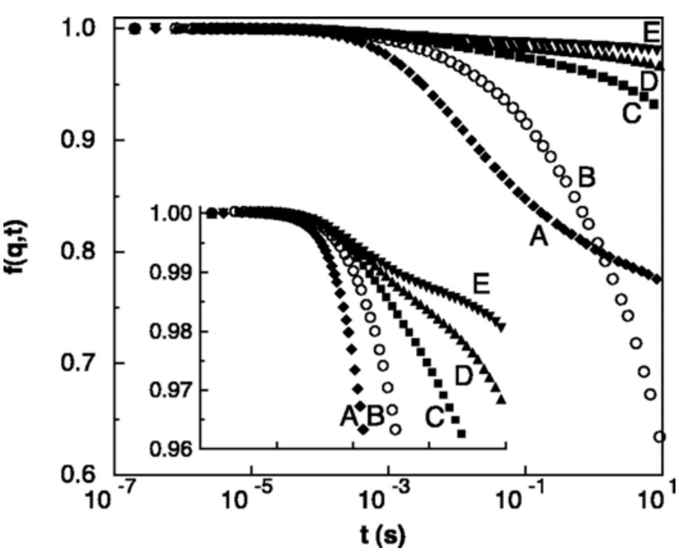
State diagram for colloidal particles



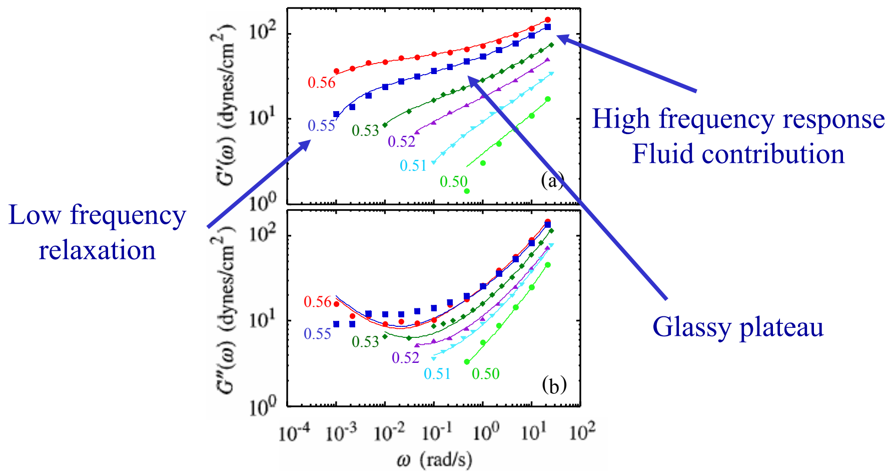
Attractive Glasses – Phase Behavior



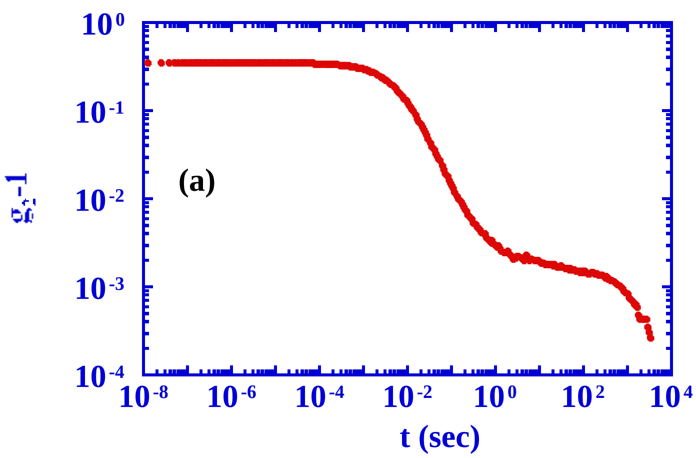
Attractive Glasses – Light Scattering



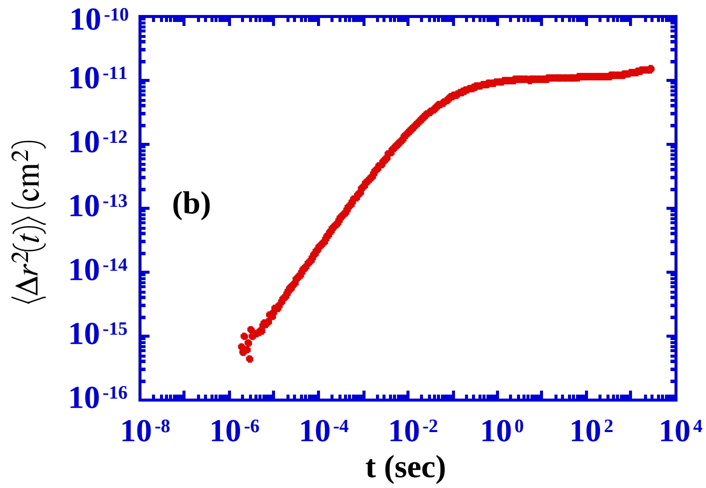
Viscoelasticity of Hard Spheres



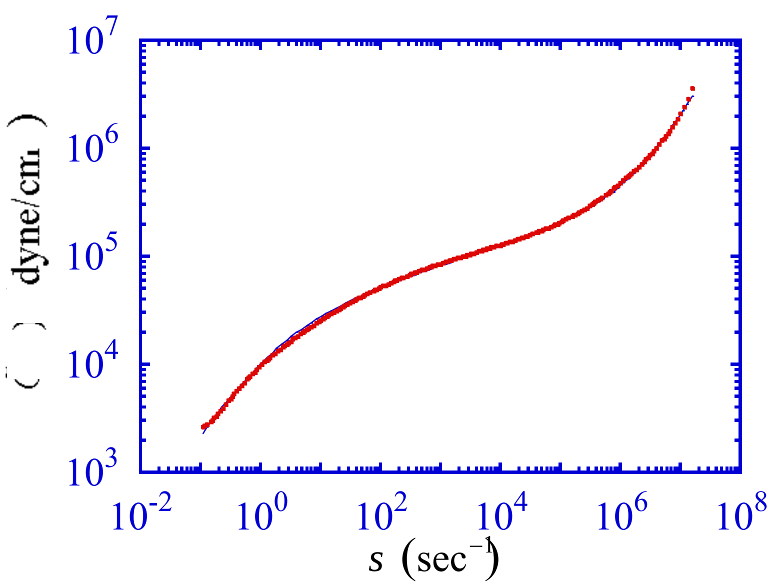
DWS correlation function of Hard Spheres



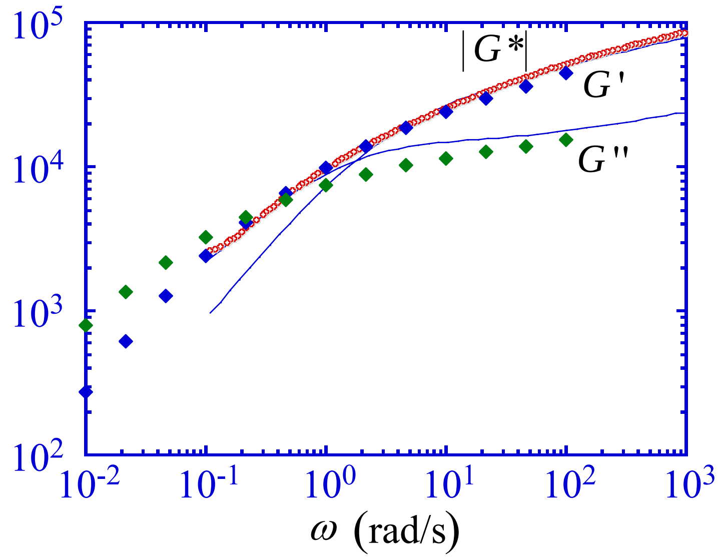
Mean Squared Displacement for Hard Spheres



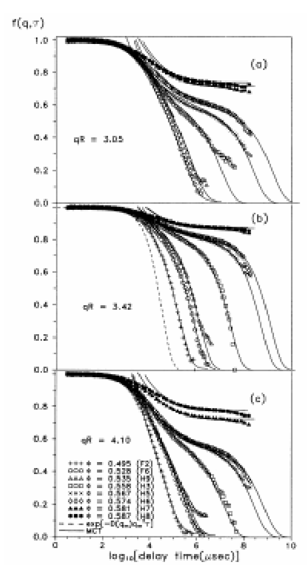
Frequency-Dependent Modulus for Hard Spheres



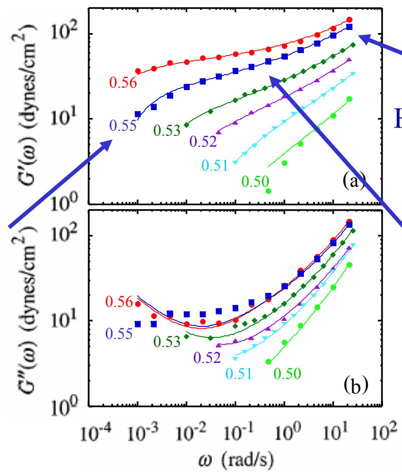
Frequency-Dependent Viscoelasticity for Hard Spheres



Correlation function of Hard Spheres



Viscoelasticity of Hard Spheres



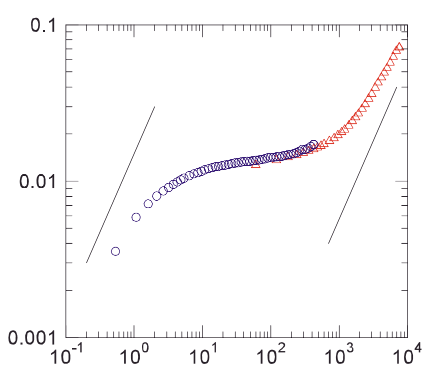
Low frequency relaxation

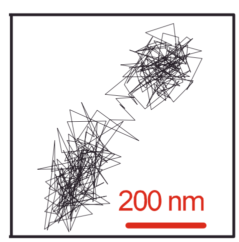
High frequency response
Fluid contribution

Glassy plateau

Mean-squared displacement

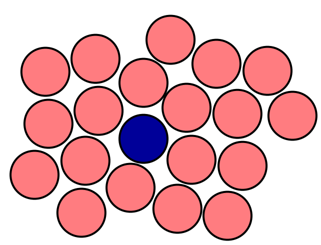
ϕ=0.53 -- "supercooled fluid"





ϕ=0.56, 100 min
(supercooled fluid)

Cage trapping:

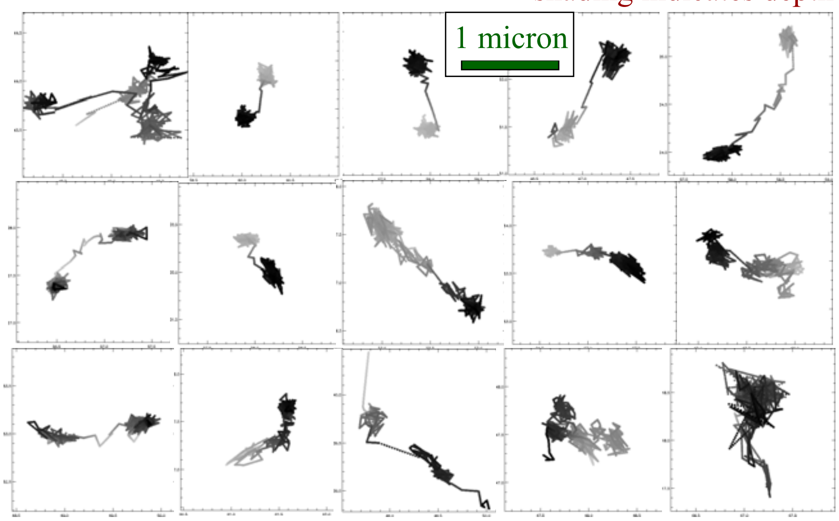


- Short times: particles stuck in "cages"
- Long times: cages rearrange

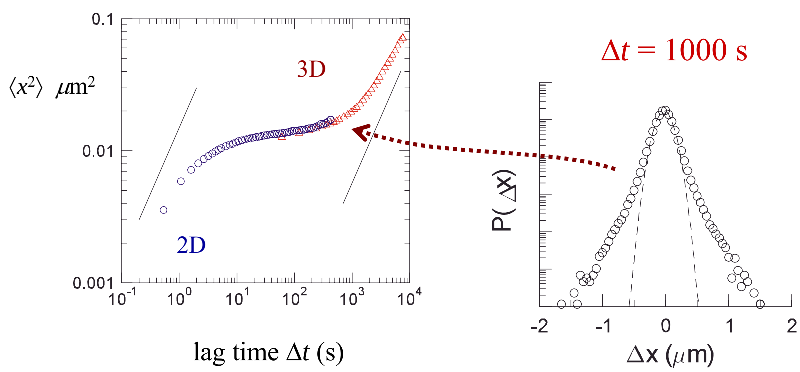
E. Weeks, J. Crocker

Trajectories of "fast" particles, ϕ=0.56

shading indicates depth



Displacement distribution function

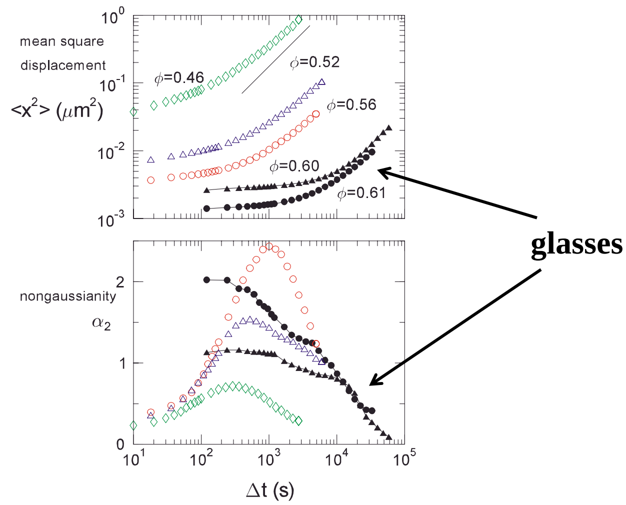


ϕ = 0.53: "supercooled fluid"

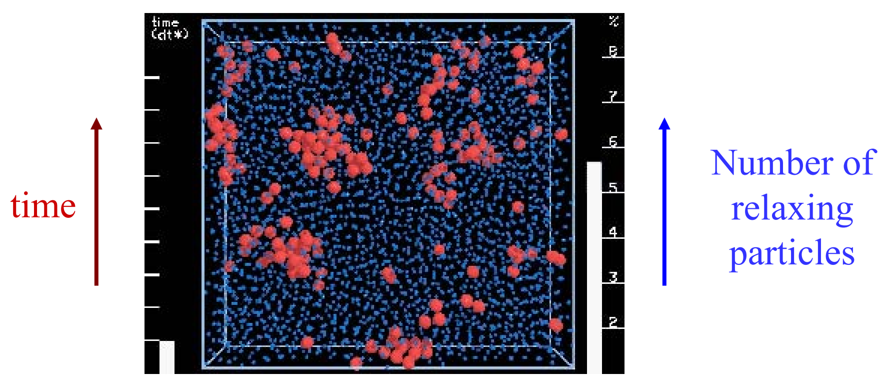
Nongaussian Parameter

$$\alpha_2 = \frac{\langle x^4 \rangle}{3\langle x^2 \rangle^2} - 1$$

How to pick Δ*t** for glasses?



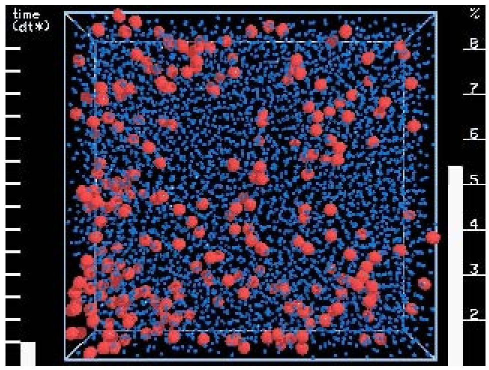
Structural Relaxations in a Supercooled Fluid



Relaxing particles are highly correlated spatially

Structural Relaxations in a Glass

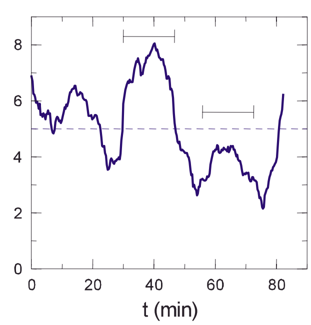
time

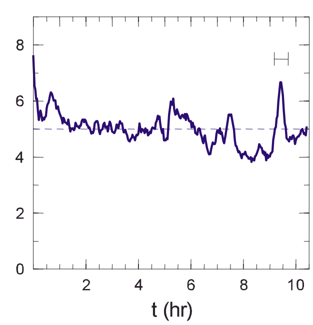


Number of relaxing particles

Relaxing particles are NOT correlated spatially

Fluctuations of fast particles

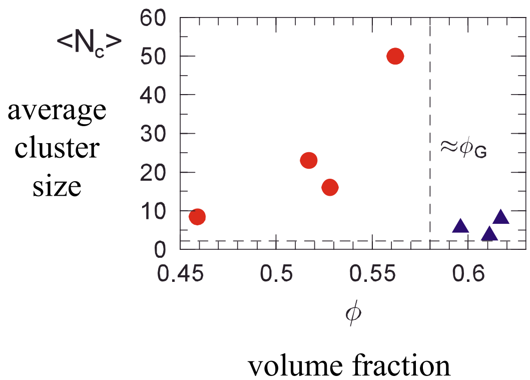




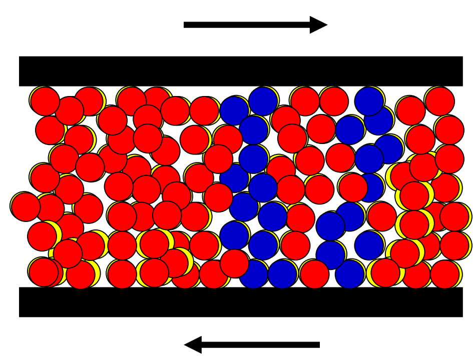
Supercooled fluid $\phi = 0.56$

Glass $\phi = 0.61$

Cluster size grows as glass transition is approached



What is a Glass?



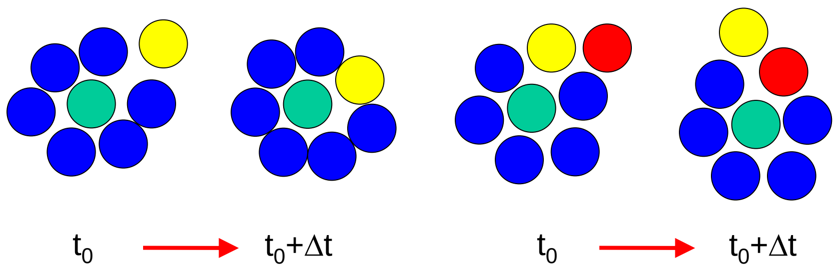
•Glass must have a low frequency shear modulus

•Must have force chains to transmit stress

J. Conrad, P Dhillon, D. Reichman

Topological Change: $\Delta nn\ (\Delta t)$

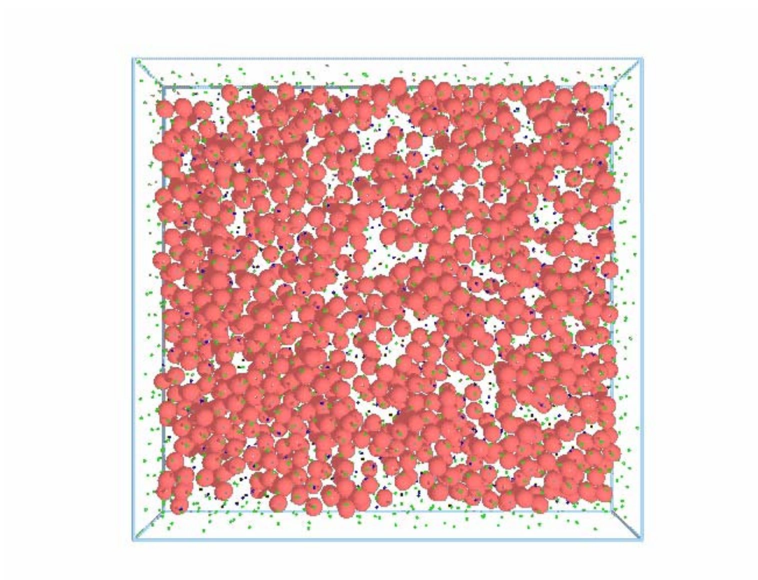
Identify nearest neighbors, calculate $\Delta nn(\Delta t)$



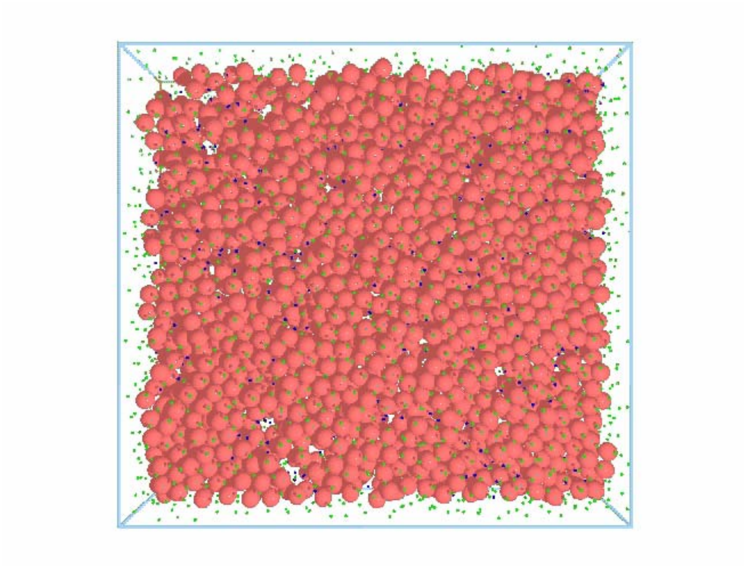
$\Delta nn = 1$ $\Delta nn = 2$

B. Doliwa and A. Heuer, J. Non-Cryst. Solids **307**, 32 (2002).

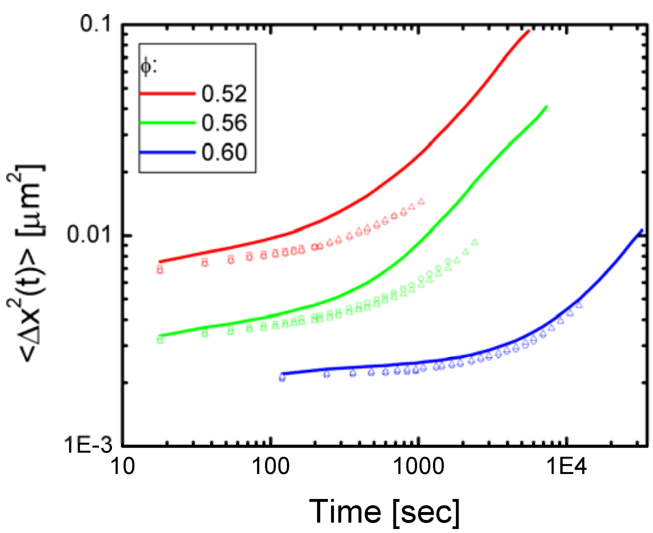
Static Particles ($\Delta nn = 0$) – $\phi = 0.52$



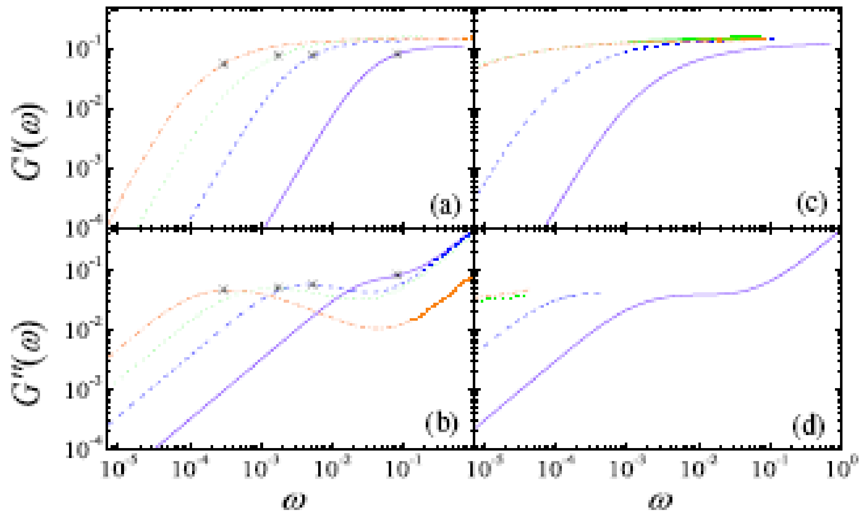
Static Particles (Δnn = 0) – ϕ = 0.60



Static Particles Move Much Less

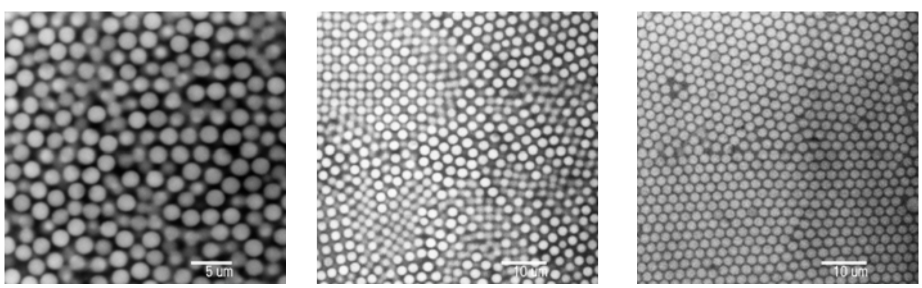


Rheology of slow particles



All particles Slow particles

Intermezzo – nucleation of HS crystals

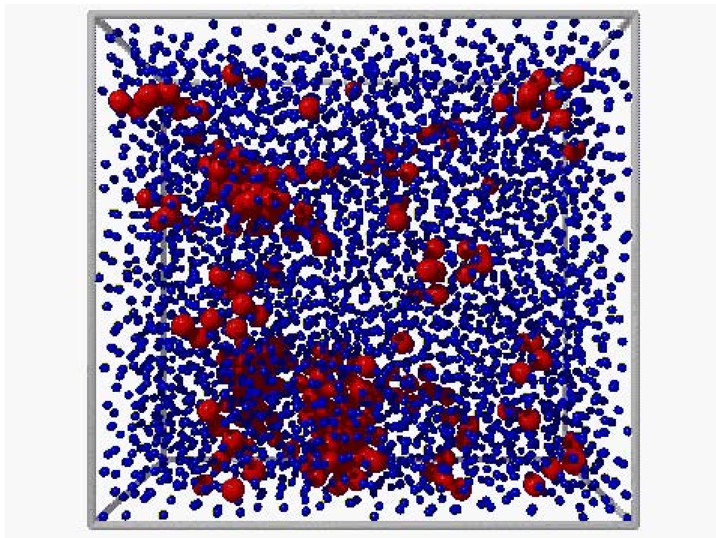


2.3 μm diameter PMMA spheres

Must identify incipient crystal nuclei

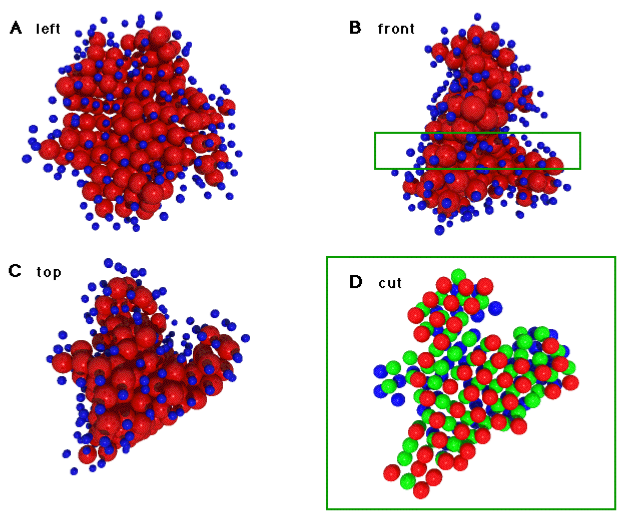
U. Gasser, E. Weeks, J. Crocker

Colloidal Crystallization



Crystal Nucleus Structure

$R \sim R_c \quad \phi = 0.47$



Finding Surface Tension

$$\Delta G = \gamma\left(4\pi r^2\right) - \Delta\mu\left(\tfrac{4}{3}\pi r^3\right)$$

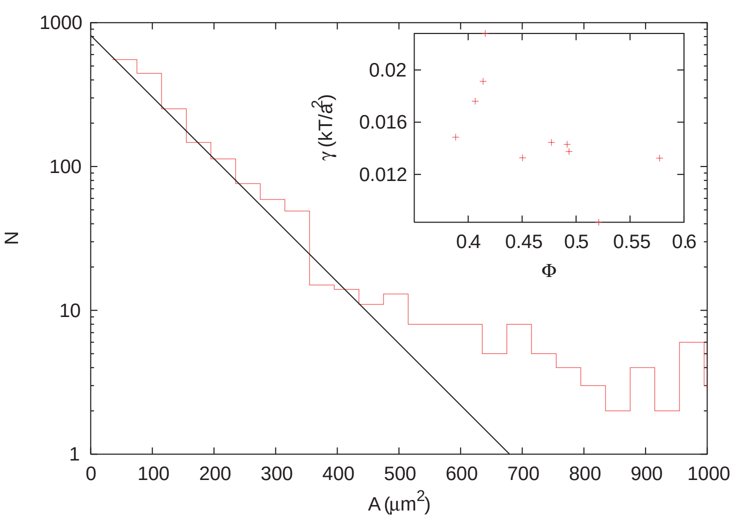
Surface energy

Chemical potential

$$P(r) \approx \exp\left(\frac{-\Delta G}{k_B T}\right) \approx \exp\left(-\gamma r^2\right)$$

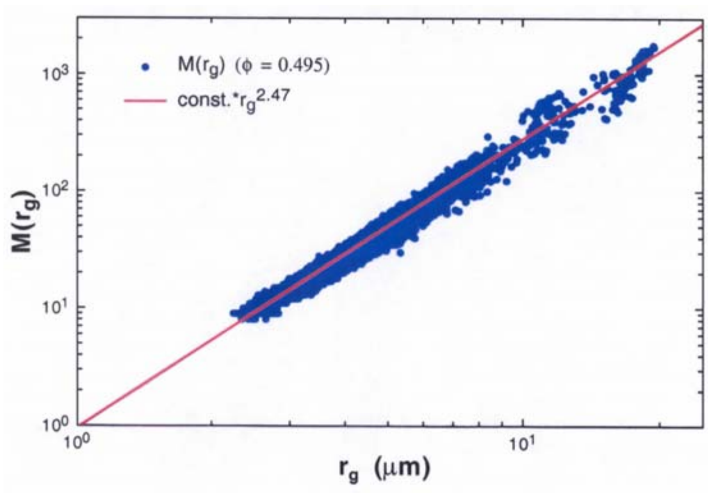
(for small r)

Measurement of Surface Tension

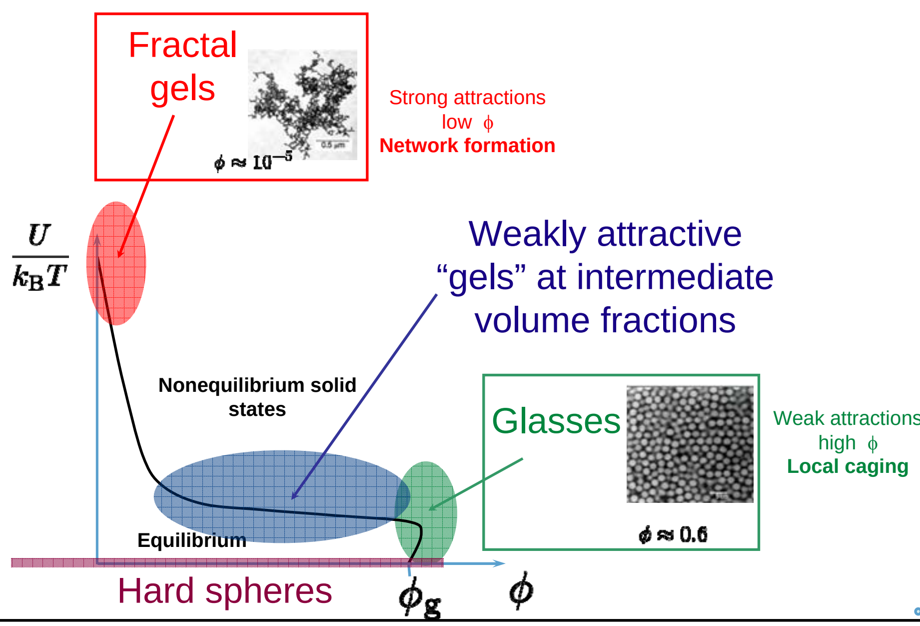


Surface tension is very low

Fractal Dimension of the Nuclei

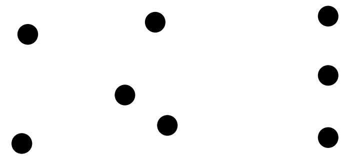


State diagram for colloidal particles



COLLIOD AGGREGATION

DILUTE, STABLE SUSPENSION

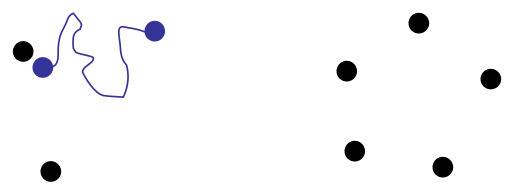


R

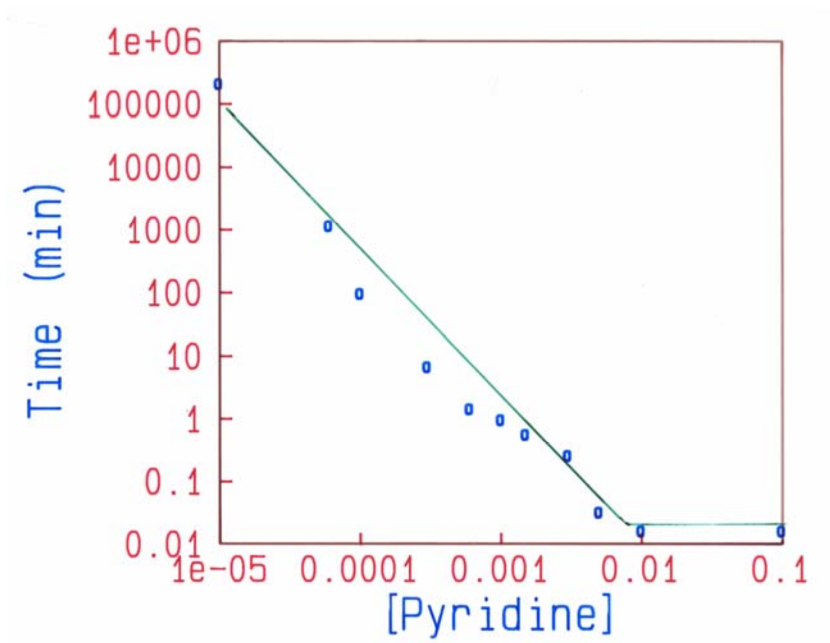
$$D = \frac{kT}{6\pi\eta R}$$

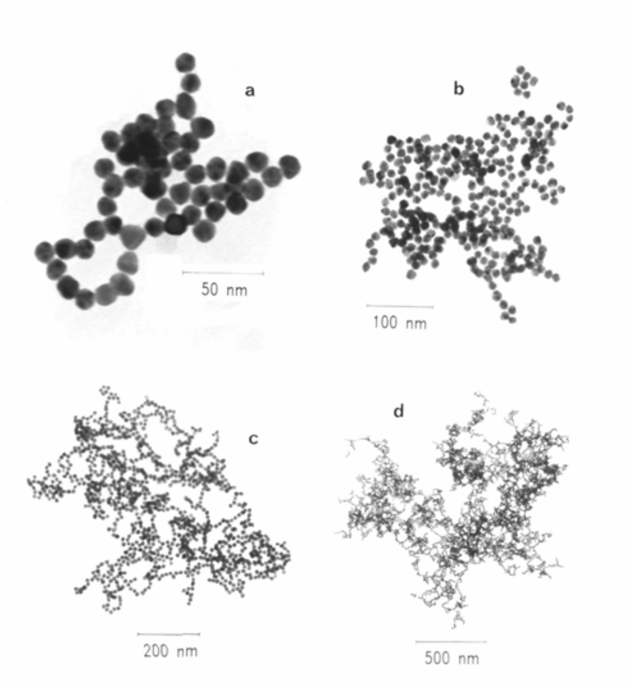
ϕ

DESTABILIZE

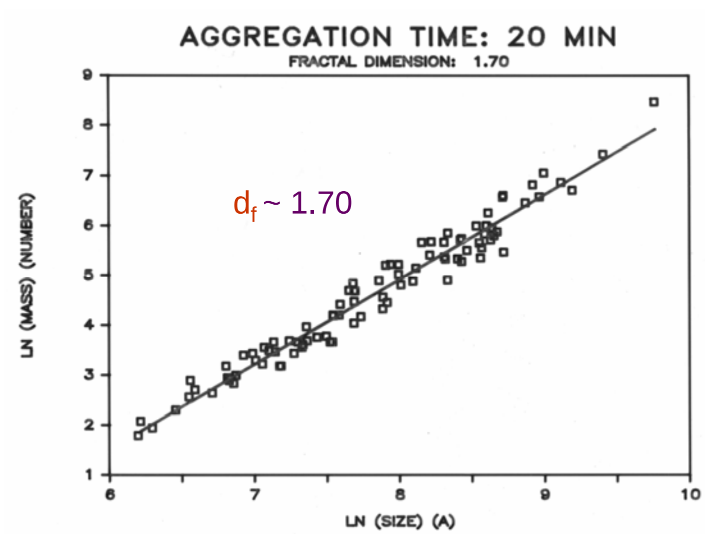


Aggregation Time





a
b
50 nm
100 nm
c
d
200 nm
500 nm



AGGREGATION TIME: 20 MIN
FRACTAL DIMENSION: 1.70
LN (MASS) (NUMBER)
LN (SIZE) (A)
d_f ~ 1.70

M. Oliveria

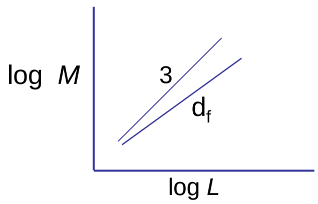
FRACTAL:

- SELF-SIMILAR
  NO CHARACTERISTIC LENGTH SCALE

$$M \sim R^{d_f}$$

d_f: FRACTAL DIMENSION

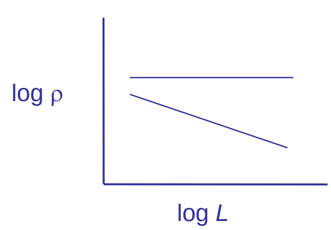
NON-INTEGRAL



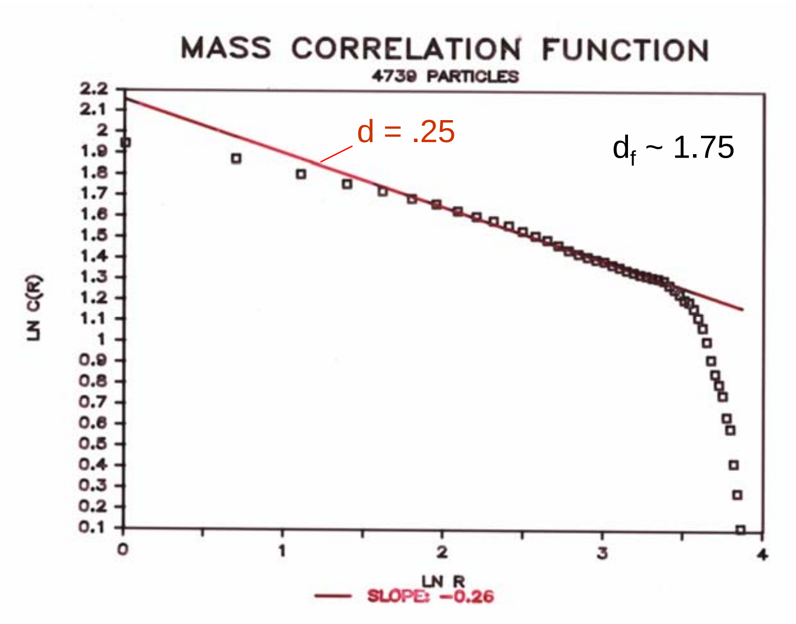
$d_f < d$

DENSITY: DECREASES WITH SIZE

$$\rho = \frac{M}{V} = \frac{L^{d_f}}{L^d} = L^{d_f - d}$$

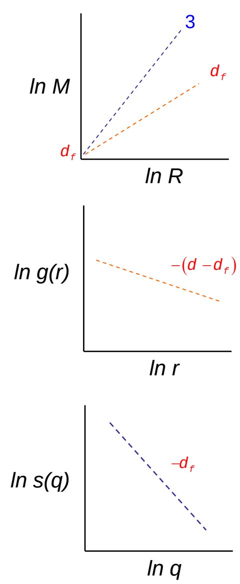


← FRACTAL



STRUCTURE OF AGGREGATES

-SCALE INVARIANT - FRACTAL



$$M \sim R^{d_f}$$

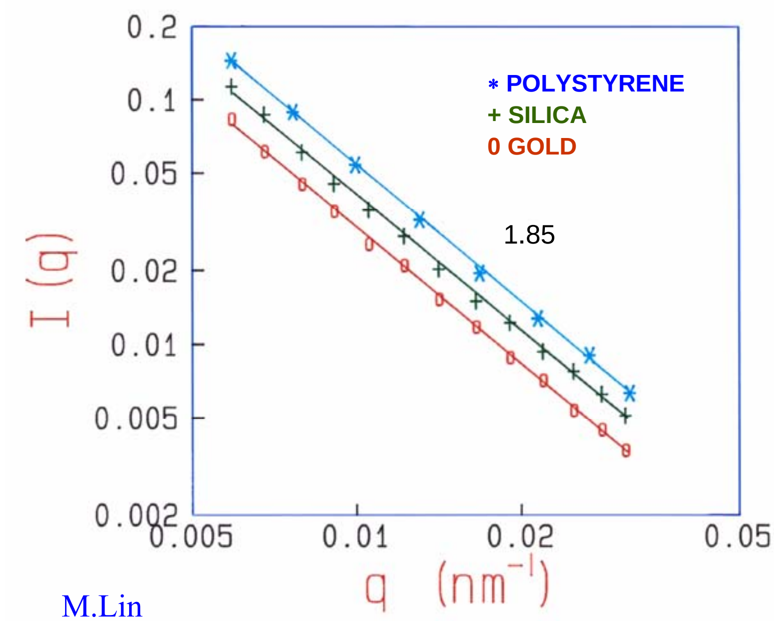
$$d_f < d$$

$$g(r) \sim \frac{1}{r^{d-d_f}}$$

$$S(q) \sim \frac{1}{q^{d_f}}$$

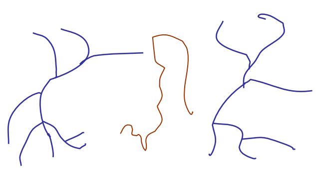
$$S(q) \sim N_q M_q^2 \sim \frac{M_T}{M_q} M_q^2 \sim R^{d_f} \sim q^{-d_f}$$

DIFFUSION-LIMITED COLLOID AGGREGATION

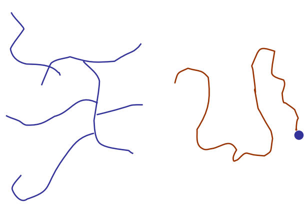


M.Lin

DIFFUSION-LIMITED CLUSTER AGGREGATION



$$d_f^1 + d_f^2 + d_t = 1.75 + 1.75 + 2 > 3.$$

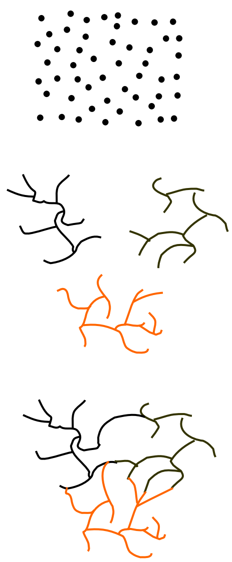


$$d_f^1 + d_f^2 + d_t = 1.75 + 0 + 2 > 3.$$

NO INTERPENETRATION
BUT CLUSTERS STICK WITH OTHER CLUSTERS

$\therefore d_f \sim 1.8$ in 3-d.

DIFFUSION-LIMITED GELATION



MASS	NUMBER	VOLUME FRACTION
M_0	N_0	Φ_0

$$M_c \simeq \left(\frac{R_c}{a}\right)^{d_f}$$

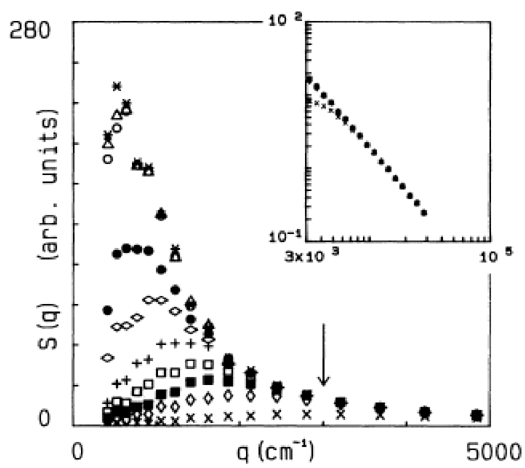
$$N_c = \frac{N_o}{M_c} = N_0 \left(\frac{R_c}{a}\right)^{-d_f}$$

$$\phi_c = \frac{V_c}{V} N_c = \frac{R_c^3}{V} N_0 \left(\frac{R_c}{a}\right)^{-d_f} = \phi_0 \left(\frac{R_c}{a}\right)^{3-d_f}$$

$$\phi_g = \phi_c = 1$$

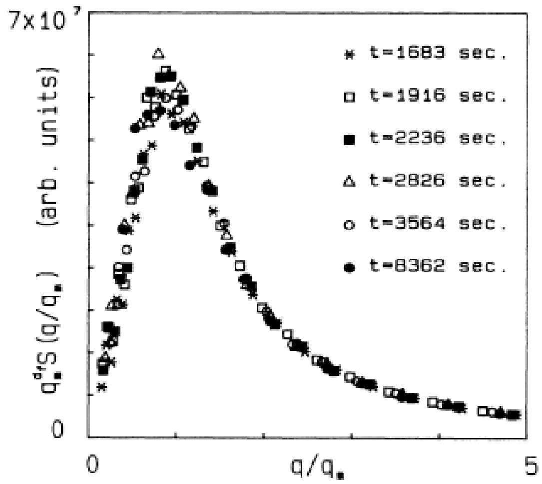
$$R_c = a\phi_0^{1/3-d_f}.$$

Static scattering from gelling colloid



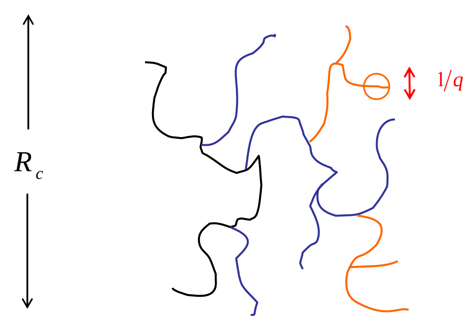
Carpineti and Giglio, PRL (1992)

Scaling of static scattering from gelling colloid



Carpineti and Giglio, PRL (1992)

DYNAMICS OF A FRACTAL COLLOIDAL GEL



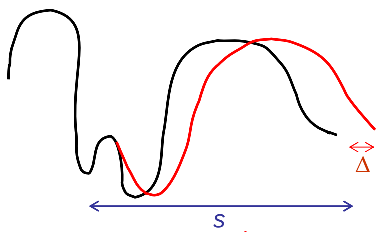
Krall

- CONSIDER BLOB OF SIZE $1/q = R_q$
- MOTION DUE TO FLUCTUATIONS OF ALL OTHER CONNECTED BLOBS
    - INDEPENDENT MODES
    - OVERDAMPED

LARGER BLOBS: LARGER DISPLACEMENT
SLOWER TIMESCALES

DYNAMICS "INSIDE" CLUSTER

KANTOR, WEBMAN; BALL



$$\Delta \simeq S^2 \left(\frac{s}{a}\right)^{d_B} a \frac{1}{k_0 a^3} f$$

SIZE-DEPENDENT SPRING CONSTANT

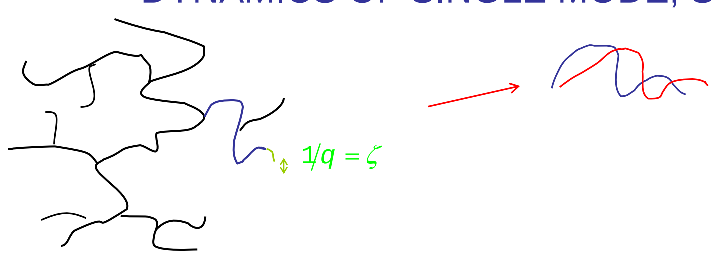
$$k(s) \simeq k_0 \left(\frac{s}{a}\right)^{-\beta} \qquad \beta = 2 + d_\beta \simeq 3.1$$

$$k(s) = k_c \left(\frac{s}{R_c}\right)^{-\beta}$$

↑

AVERAGE CLUSTER SPRING CONSTANT

DYNAMICS OF SINGLE MODE, S



CONSTRAINED BROWNIAN MOTION $\left\langle \Delta r_\zeta^2(t) \right\rangle_s = \frac{kT}{k(s)n(s)}\left[1 - e^{-t/\tau(s)}\right]$

1. SPRING CONSTANT:

$$k(s) = k_0 \left(\frac{s}{a}\right)^{-\beta}$$

SINGLE PARTICLE SPRING CONSTANT

SIZE DEPENDENT

KANTOR-WEBMAN

$$\beta = 2 + d_B$$

→ "BOND" DIMENSION

2. TIME CONSTANT:

$$\tau(s) = \frac{6\pi\eta s}{k(s)}$$

VISCOUS DAMPING

3. MAXIMUM AMPLITUDE:

$$\delta_m^2 = \frac{kT}{k(s)n(s)}$$

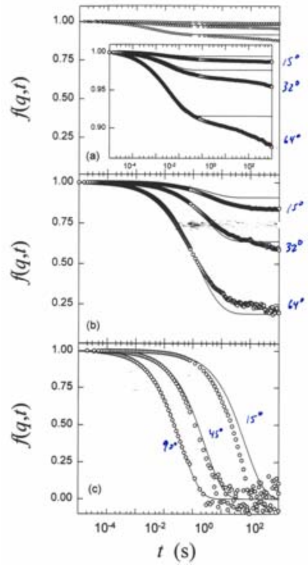
kT / NORMAL MODE

n(s): DENSITY OF LOCAL MODES

$$n(s) = N_c \left(\frac{a}{s}\right)^{d_f}$$

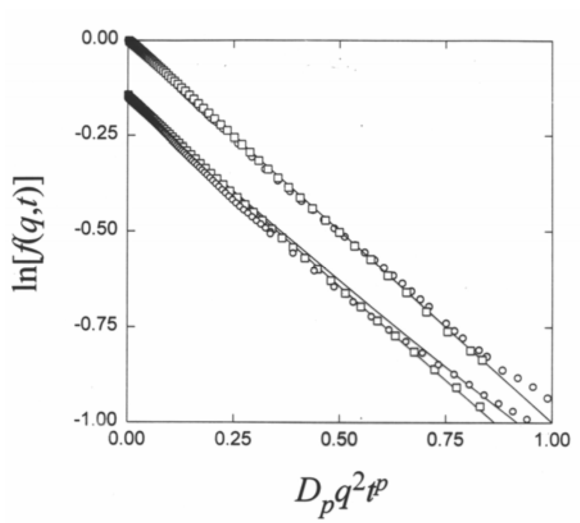
DYNAMIC LIGHT SCATTERING FROM COLLOIDAL GEL

BOUYANCY-MATCHED POLYSTYRENE ***a = 9.5 nm***



ϕ_0
5.0×10^{-3}
1.5×10^{-3}
1.7×10^{-4}

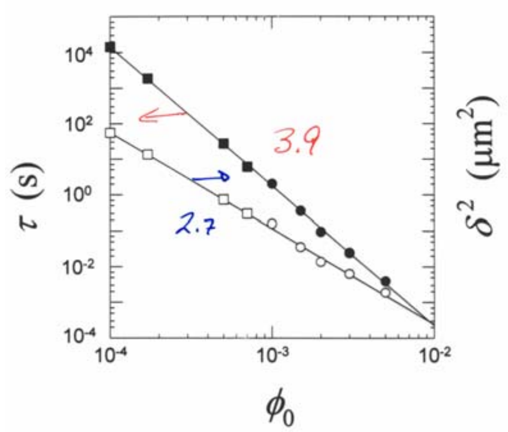
SCALING STRETCHED EXPONENTIAL



$P = 0.66$

VOLUME FRACTION DEPENDENCE

LIGHT SCATTERING PROBE OF MODULUS

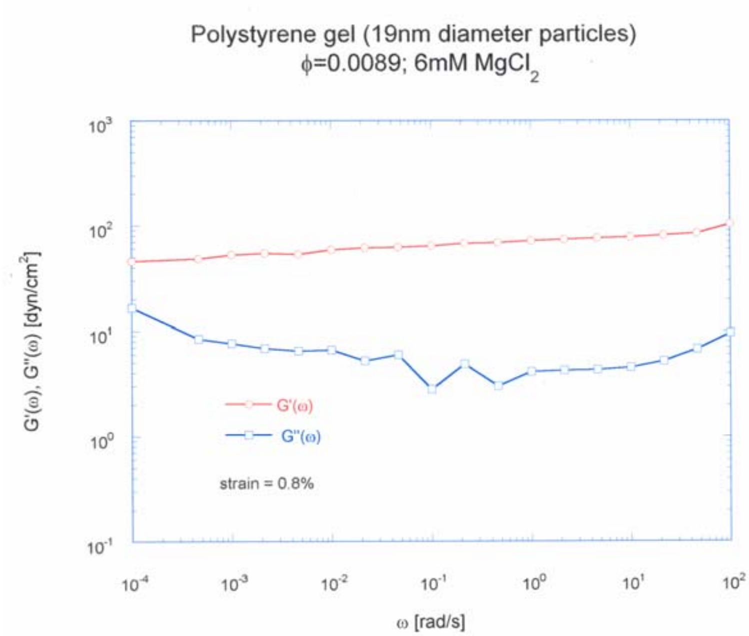


$$G \simeq \frac{k_c}{R_c}$$

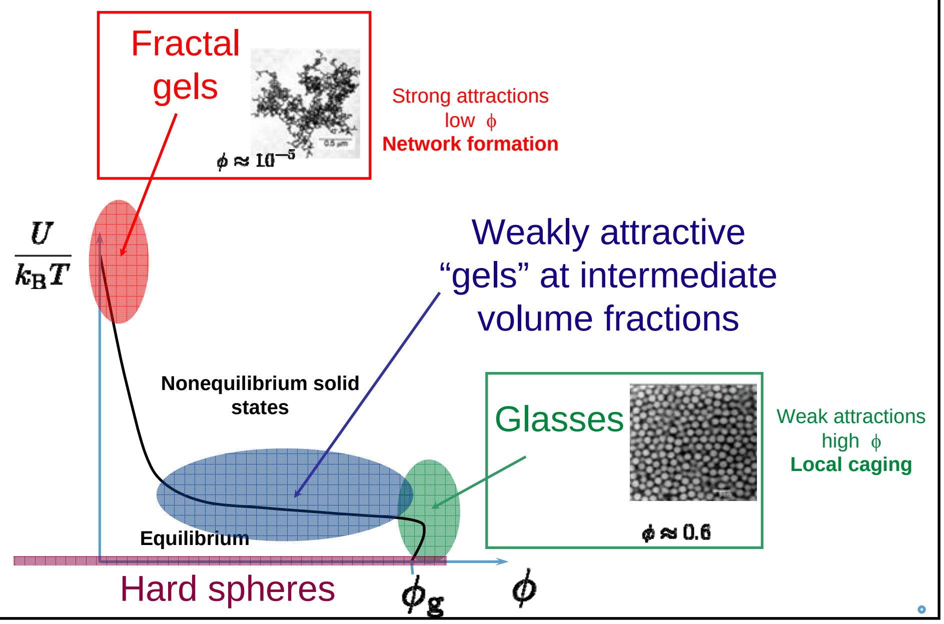
$$\text{BUT } \tau_c \sim \frac{R_c}{k_c}$$

$$G \sim \phi^{3.9}$$

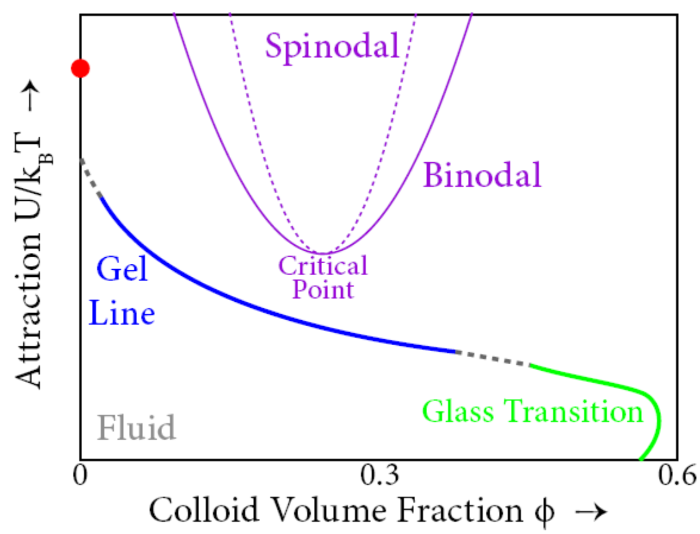
R_c ONLY LENGTH IN PROBLEM



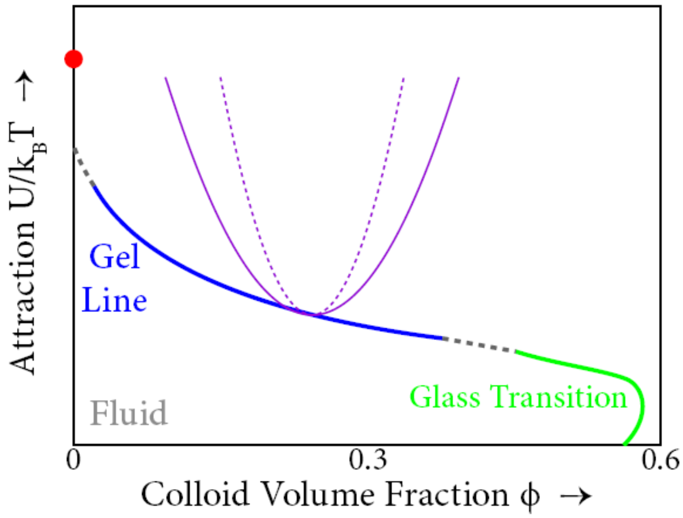
State diagram for colloidal particles



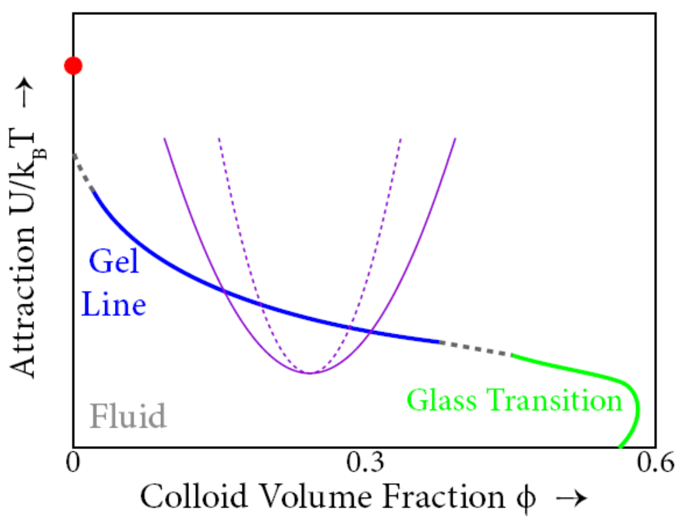
Attractive colloidal particles



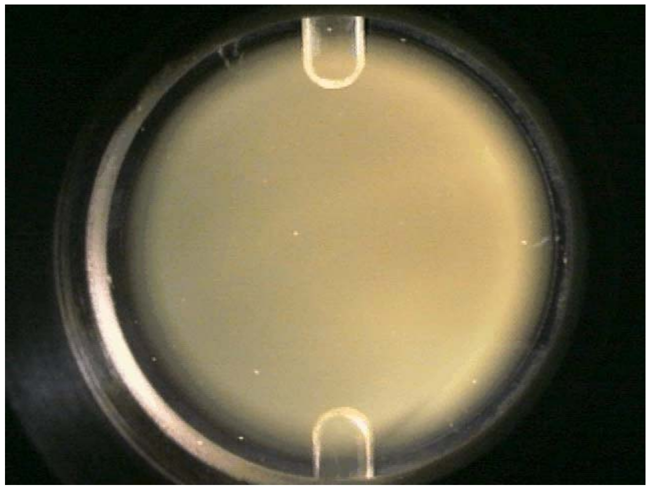
Attractive colloidal particles



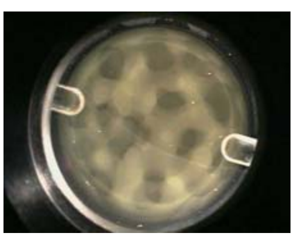
Attractive colloidal particles



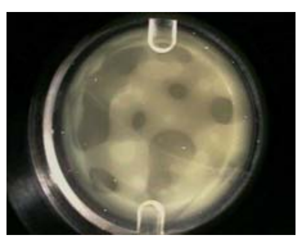
Spinodal Decomposition of Colloid Polymer



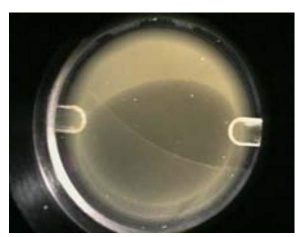
Col-Pol Critical Point: Sample Evolution



2. 3 hrs 50 min after mix



3. 13 hrs 24 min after mix

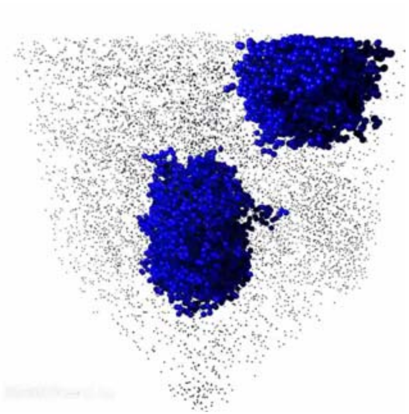


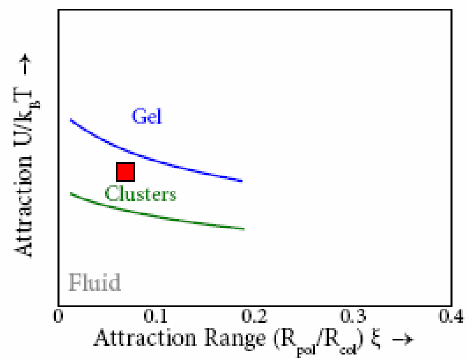
4. 35 Days, 2 hrs, 54 min after mix

Large scale phase separation - Totally unprecedented
Structure is 10,000 times larger

Stable clusters: Binodal decomposition

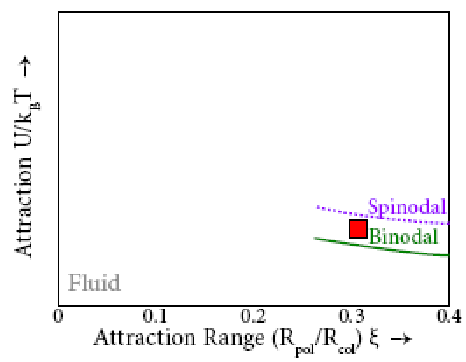
Shorter-range interaction



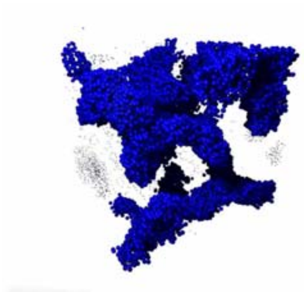


Stable clusters: Binodal decomposition

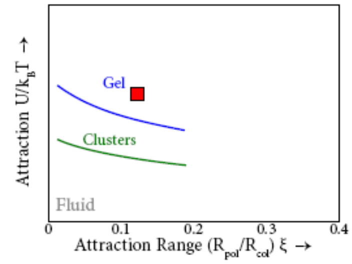
Long-range interaction

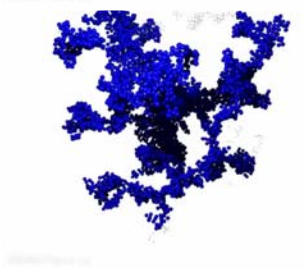


Structure depends on range

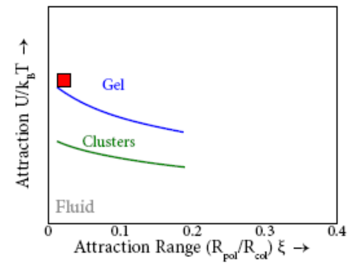


Long-range attraction

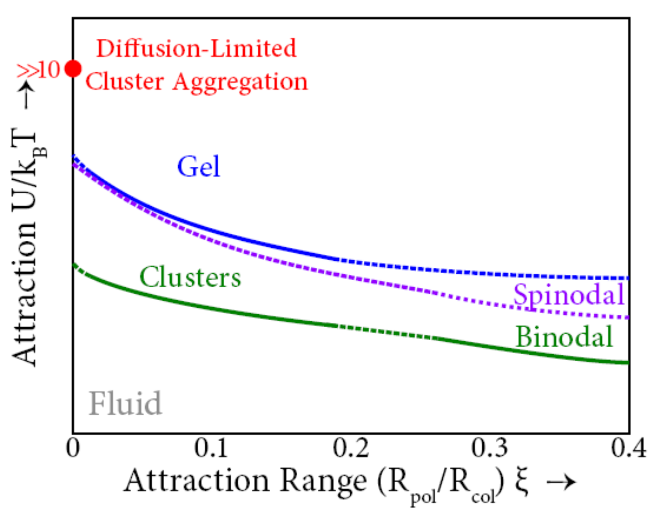




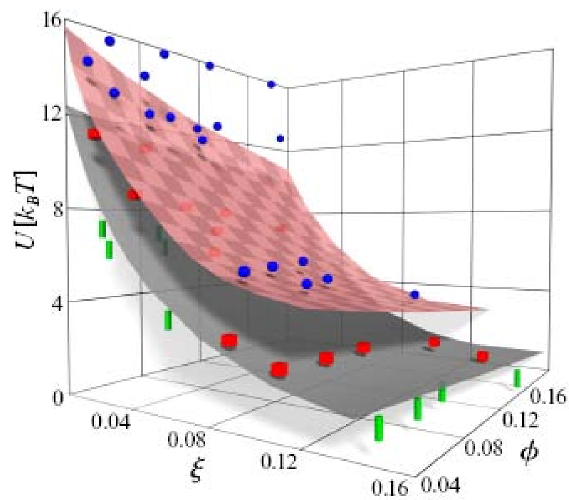
Short-range attraction



Unified Picture



Gel structure – long-range attraction



State diagram for colloidal particles