Cosmology in a petri dish? Simulation of collective dynamics of colloids at fluid interfaces

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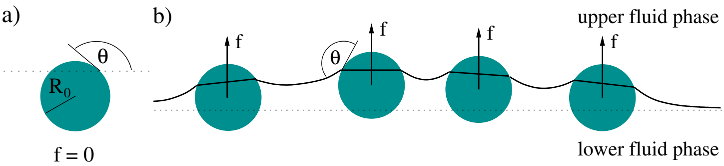
Outline

Cosmology in a petri dish

J. Bleibel, A. Domínguez, M. Oettel, S. Dietrich, EPJ E 34, 125 (2011)
A. Domínguez, M. Oettel, S. Dietrich, Phys. Rev. E 82, 011402 (2010)
M. Oettel, S. Dietrich, Langmuir 24, 1425 (2008)

Micrometer sized colloidal particles ($R_0 \approx 5 - 20\mu m$), trapped at a fluid interface:



- effectively two–dimensional (2D) system with long–ranged capillary attractions
- mean field description for the interfacial deformation:
  - → screened electrostatics in 2D
  - → 2D screened gravitational interaction

Deformation of interface due to trapped colloids

Vertical deformation $u(\vec{r})$ of the Interface due buoyancy:
linearized Young-Laplace Eq.

$$\nabla^2 u - \frac{u}{\lambda^2} = -\frac{1}{\gamma}\Pi \qquad (1)$$

$$\Pi = f\delta(\vec{r}) \qquad (2)$$

λ	screening length
γ	surface tension
Π	ext. Pressure
f	"charge"

Solution: pairwise potential for two capillary monopoles f:

$$V(d) = -\frac{f^2}{2\pi\gamma}K_0(d/\lambda) \qquad (3)$$

K_0	modified Bessel function
d	distance

for small d and in the limit $\lambda \to \infty$ (Gravity)

$$V(d) \sim \ln(\lambda/d)$$

A weak force...

- for $R = 10\mu m$ sized colloids: $U_0 \sim k_B T$
- 2 capillary monopoles of equal sign attract each other

  $\rightarrow$ 2D screened gravitational interaction with f: either positive or negative "masses"

  $\rightarrow$ like gravity in 3D: destabilization of many initial homogeneous configurations: "gravitational" collapse

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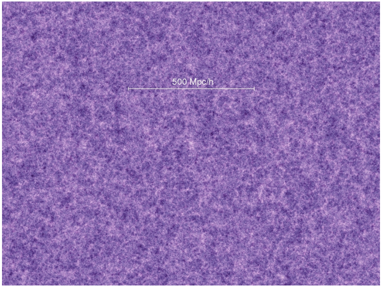

"Millennium" Simulation: Dark Matter Distribution

$\rightarrow$ A self gravitating fluid of 2D hard discs.

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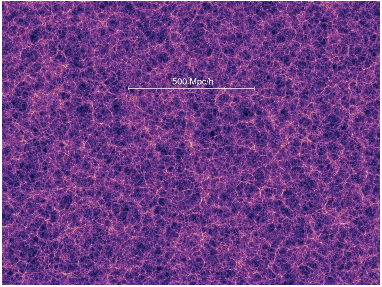

"Millennium" Simulation: Matter lumps together

$\rightarrow$ A self gravitating fluid of 2D hard discs.

A weak force...

- for $R = 10\mu m$ sized colloids: $U_0 \sim k_B T$
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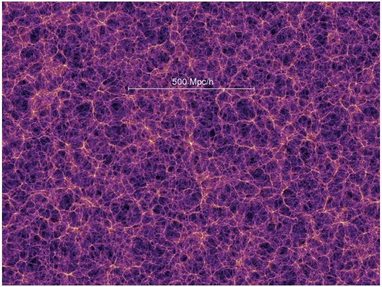

"Millennium" Simulation: Clusters and filaments

→ A self gravitating fluid of 2D hard discs.

A weak force...

- for $R = 10\mu m$ sized colloids: $U_0 \sim k_B T$
- 2 capillary monopoles of equal sign attract each other

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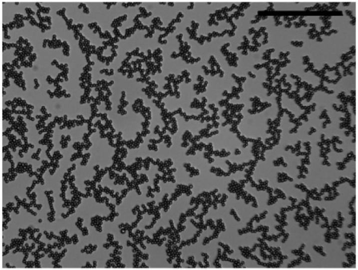
→ like gravity in 3D: destabilization of many initial homogeneous configurations: "gravitational" collapse



Colloids at interface Vermant et al., Langmuir 2006

→ A self gravitating fluid of 2D hard discs.

A highly tunable system:

- wide range of length– and timescales possible:
  Recall equation (1):

$$\nabla^2 u - \frac{u}{\lambda^2} = -\frac{1}{\gamma}\Pi$$

$\longrightarrow$ coarse-graining (averaged interface deformation):

$$\nabla^2 U - \frac{U}{\lambda^2} = -\frac{f}{\gamma}\rho(\vec{r}) \qquad (4)$$

  - R_0, f $\longrightarrow$ size and weight of the colloids
  - λ, γ $\longrightarrow$ surface tension
  - ρ $\longrightarrow$ packing fraction

  are tunable
- experimental setup for the collapse is feasible

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Toolkit

Model Toolkit for the study of 2D long-ranged interactions

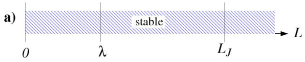
linear instability of a homogeneous configuration

J. H. Jeans, Phil. Transactions of the Royal Society of London Series A, Vol. 199 (1902) pp. 1-53

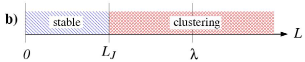
- linear stability analysis: recover Jeans' instability for $\lambda \to \infty$
- stability determined by λK_j:

Jeans' Wavenumber Jeans' Length

$$K_j = f\sqrt{\frac{\rho_h p'(\rho_h)}{\gamma}} \qquad L_j = \frac{1}{K_j}$$



stable for $\lambda K_j < 1$



unstable for $\lambda K_j > 1$

- perturbations: $k < K_c$ exponential growth, $k > K_c$ damped, with $K_c \approx K_j$, the critical wavenumber

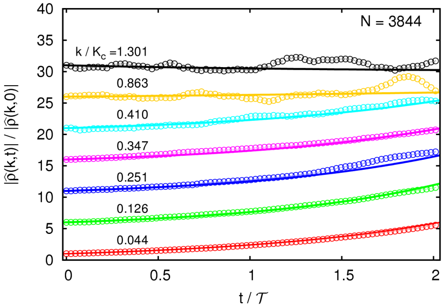
Exponential growth for amplitudes with $k < K_c$

Associated with this instability is a characteristic time

$$\mathcal{T} = \frac{\gamma}{\Gamma f^2 \rho_h}$$

ρ_h	(homogeneous) density
Γ	mobility

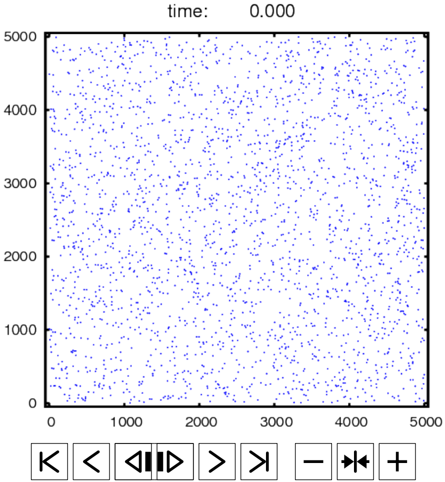
$\mathcal{T}$ sets the timescale for the growth of perturbations.



- good agreement up to $t/\mathcal{T} \sim 1.5$
- damping visible for $k > K_c$.

$\longrightarrow$ What happens for $t/\mathcal{T} > 1.5$?

Colloids at an air water interface ($\lambda = 2.7$ mm)



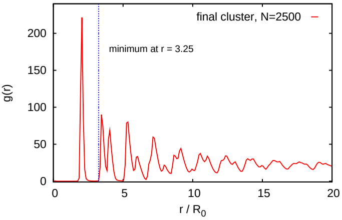
Expectations

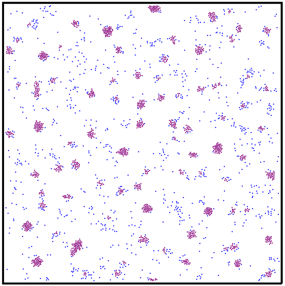
We know: System is unstable, i.e. $\lambda K_j > 1$

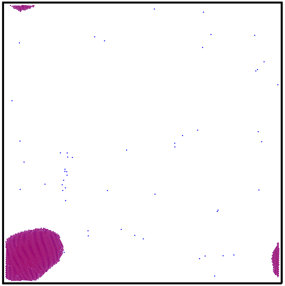
- Colloids will lump together
- Effect of long–ranged nature of the interaction? $\longrightarrow$ vary λ
- short ranged: Van-der-Waals like ($\lambda \sim R_0$) short range attraction and hard core repulsion $\rightarrow$ spinodal decomposition
- long ranged: self gravitating fluid ($\lambda \rightarrow \infty$) of hard discs $\rightarrow$ gravitational collapse?
- Finite size Effects? λ / L matters!
- System is tunable $\rightarrow$ experimentally meaningful

How to quantify clustering phenomenology?

Ansatz: define a Cluster via distances:

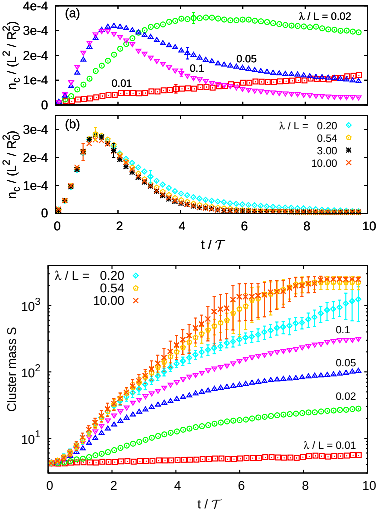






- define cluster if $N > 3$ with $d < 3.25R_0$
- use to mark clusters
- Investigate the mean number of clusters and the mean cluster size

cluster dynamics



- two distinct regimes for times and λ
- large λ: rapid clustering up to $t \simeq 1.5\mathcal{T}$
- rapid domain growth afterwards
- scaling for $\lambda \geq 1mm$ $(\lambda/L = 0.2)$
- for small λ: Minimum shifted, no scaling
- no characteristic time $\rightarrow$ spinodal decomposition
- power law domain growth

Cold and warm collapse

J. Bleibel, A. Domínguez, M. Oettel, S. Dietrich, EPJ E 34, 125 (2011) A. Domínguez, M. Oettel, S. Dietrich, Phys. Rev. E 82, 011402 (2010)

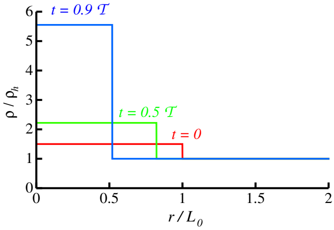
The cold collapse scenario:

- standard scenario in the astrophysical literature (collapse of a self gravitating pressure less fluid)
- can be solved analytically for $\lambda \to \infty$ (top-hat profile, Newtonian limit)
- radial evolution

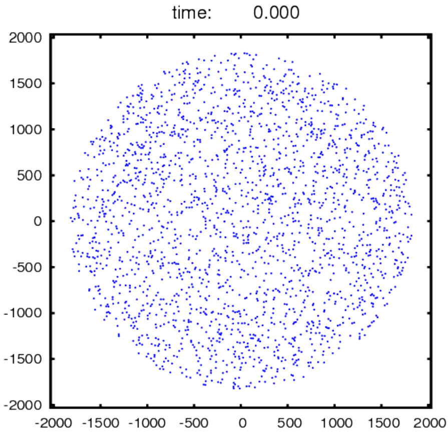
$$R(t, R_0) = R_0 \sqrt{\frac{\widehat{\rho}}{\rho_h} + \left(1 - \frac{\widehat{\rho}}{\rho_h}\right) e^{t/\mathcal{T}}}$$

- time of collapse:

$$\mathcal{T}_{coll} = -\mathcal{T} \ln \left(1 - \frac{\rho_h}{\widehat{\rho}}\right)$$



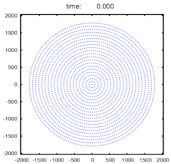
hot gas disc, $\lambda/R_0 = 1.5$



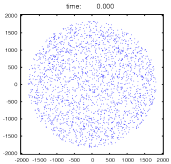
Collapsing colloids, the limit $\rho_h \to 0$

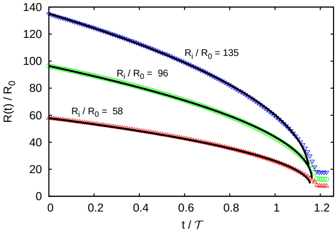
$$\rho_h \to 0 \quad \Longrightarrow R(t, R_0) = R_0 \sqrt{1 - \frac{t}{\mathcal{T}}}, \quad T_{coll} = \mathcal{T}$$

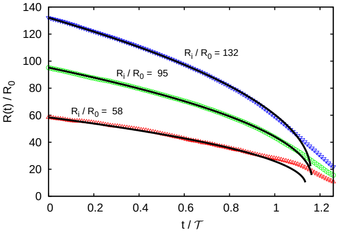
cold rings



hot gas







dense packing reached later: overall deviation $\simeq 15\%$

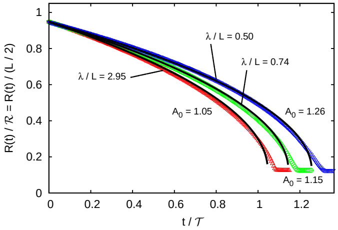
The collapse taken to the limits

What is the reason for the deviation?

- fit radial evolution according to

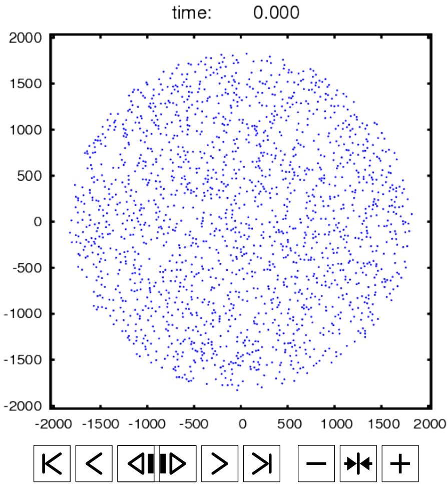
$$R(t) - R_f = [R_i - R_f]\sqrt{1 - \frac{t}{A_0 \mathcal{T}}}$$

- Investigate the radial evolution and stretching of the time of collapse ($\mathcal{T}_{coll} \approx A_0 \mathcal{T}$) for the limits $\lambda \to \infty$ and $\lambda \ll 1$.



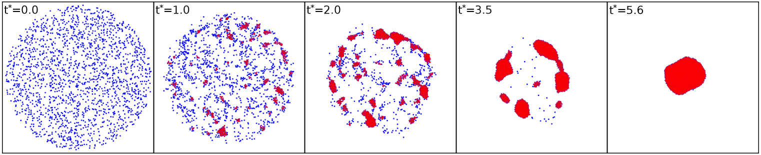
- $A_0 \to 1$ for $\lambda \to \infty$ (gravitational limit)
- A_0 grows (diverges) for $\lambda < 1$

hot gas disc, $\lambda/R_0 = 0.25$



Collapsing colloids, the limit $\lambda/L \ll 1$

J. Bleibel, S. Dietrich, A. Domínguez, M. Oettel, PRL 107, 128302 (2011)



Ringlike meta–structure: inbound shockwave for $\lambda/L = 0.25$?

- Mean field diffusion equation (ensemble averaged):

$$\frac{\partial \rho}{\partial t} = -\nabla(\rho \vec{v}) = -\Gamma \nabla (f \rho \nabla U - \nabla p(\rho)) \qquad (5)$$

- reduced units (L: system size)

$$\hat{r} = \frac{r}{L}, \quad \hat{\rho} = \frac{\rho}{\rho_0}, \quad \hat{p} = \frac{p}{k_\mathrm{B} T \rho_0}, \quad \hat{\lambda} = \frac{\lambda}{L}, \quad \hat{t} = \frac{t}{\mathcal{T}}$$

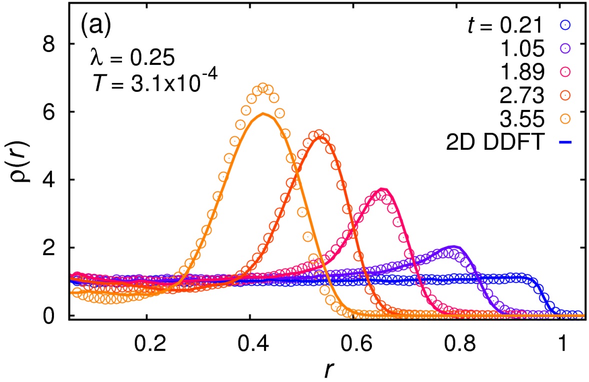
Shock waves – dynamical DFT

introducing the effective Temperature T_{eff}:

$$T_{\text{eff}} = \frac{\gamma k_{\mathrm{B}} T}{f^2 \rho_0 L^2} \tag{6}$$

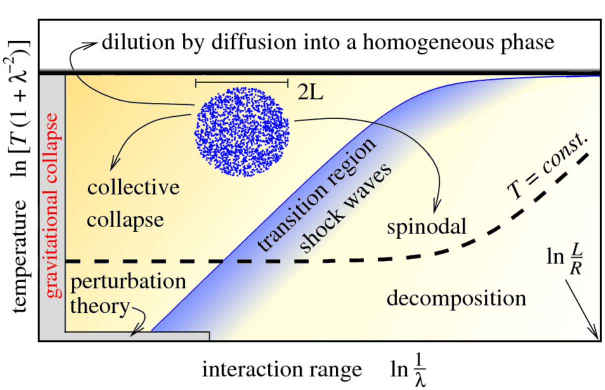
$\Longrightarrow$ evolution equation for the density:

$$\frac{\partial \hat{\rho}}{\partial \hat{t}} = -\nabla(\hat{\rho} \nabla U[\hat{\rho}] - T_{\text{eff}} \nabla \hat{p}(\hat{\rho})) \tag{7}$$



Dynamical phase diagram

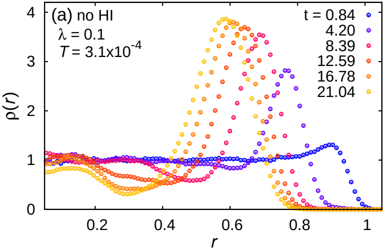
J. Bleibel, S. Dietrich, A. Domínguez, M. Oettel, PRL 107, 128302 (2011)



- $\lambda/L \to \infty$: homogeneous collapse
- $\lambda/L < 1$: inbound traveling shockwave
- $\lambda/L \ll 1$: individual clustering (spinodal decomposition)

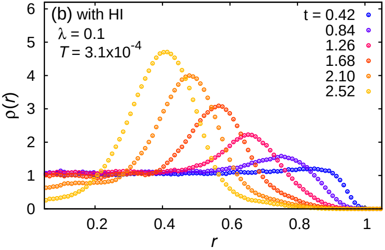
Collapse with HI

- circular patch, $\lambda/L = 0.1$



- characteristic time scale changes (speedup)

- shock wave phenomenology unchanged



Summary & Conclusions I

Brownian Dynamics

dynamics of colloids with long–ranged interactions

- Langevin dynamics with PM Method for capillary forces
- tunable system from short to long–ranged attraction

Gravitational Instability

Jeans' instability also seen in 2D colloids at interfaces

- good agreement with theoretical stability analysis
- phenomenology changes from spinodal decomposition to collapse of a self gravitating fluid
- Analysis of clustering (Minkowski functionals)

Summary & Conclusions II

Shock waves

Collapse of a circular patch

- visible for intermediate interaction ranges
- dynamical phase diagram

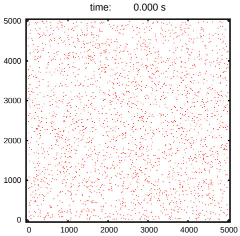
Hydrodynamic interactions

HI incorporated at the two particle level

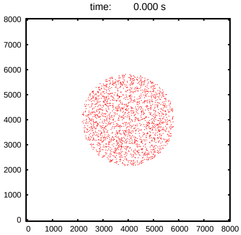
- linear stability analysis predicts speedup due to hidden dimension
- speedup seen in simulations (collapse & bulk system)

Initial conditions

- colloids can be distributed randomly, avoiding overlap $\longrightarrow$ hard disc gas, averaged over many realizations.



- place colloids surrounded by "vacuum" $\longrightarrow$ reduce "tidal" forces by periodic images, radial symmetry



by default: choose the hard disc gas

MD-Simulation with Brownian Dynamics

Aim: a full dynamic simulation of capillary forces between colloids trapped at interfaces

- Use Molecular Dynamics approach
- Interplay of particle and interface $\longrightarrow$ Brownian Dynamics
- Ermak algorithm: integration of the Position–Langevin equation

$$\dot{\vec{r}} = \frac{D}{k_B T}\vec{f}_{tot} + \mathring{\vec{r}} \qquad (8)$$

$\mathring{\vec{r}}$	delta correlated random velocity
D	diffusion const.

Ingredients

- short–ranged interparticle repulsion (WCA-Potential) $\longrightarrow$ no overlapping of particles ("hard" discs).
- neglect hydrodynamic interactions
- periodic boundary conditions: long–ranged forces extend up to periodic images
  $\rightarrow$ exponential tail of K_0 in the potential only for $d > \lambda \sim L$ (system size)
- special treatment of the long-ranged forces needed:

  $\rightarrow$ capillary forces calculated with Particle–Mesh method (PM) ($\rightarrow$ cosmological N-body simulations).

The Particle-Mesh (PM) Method

Idea: solve Helmholtz equation (4) by means of Fourier transformation:

$$
\begin{aligned}
\nabla^2 u(\vec{r}) - \frac{u(\vec{r})}{\lambda^2} &= -\frac{f}{\gamma}\rho(\vec{r}) \\
\mathcal{F}\left[\nabla^2 u(\vec{r}) - \frac{u(\vec{r})}{\lambda^2}\right] &= \mathcal{F}\left[-\frac{f}{\gamma}\rho(\vec{r})\right] \\
\left(-k^2 - \frac{1}{\lambda^2}\right)\widehat{u}(\vec{k}) &= -\frac{f}{\gamma}\widehat{\rho}(\vec{k}) \\
u(\vec{r}) &= \mathcal{F}^{-1}\left[-\frac{f}{\gamma}\,G(\vec{k})\,\widehat{\rho}(\vec{k})\right] \qquad (9)
\end{aligned}
$$

$\mathcal{F}$ and $\mathcal{F}^{-1}$: Fourier transform and inverse Fourier transform
$G(\vec{k}) = (-k^2 - \frac{1}{\lambda^2})^{-1}$: Greens' function

The Particle-Mesh (PM) Method: How to...

... proceed:

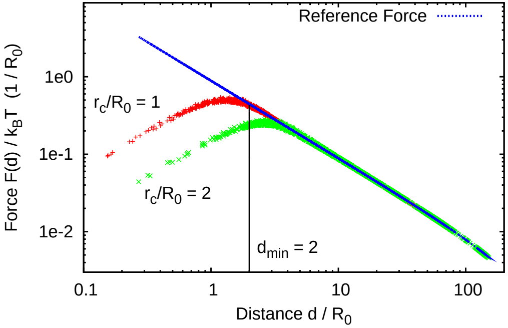
- assign colloids to a grid (or mesh) by means of a mass assignment scheme
- use standard FFT for Fourier transformation of the grid
- multiply with the Greens function of the corresponding Helmholtz equation.
- multiply the resulting potential in Fourier space with ik $\longrightarrow$ differentiation in real space and transform back with FFT^{-1}.
- interpolate the resulting forces back to the particles positions using the inverse mass assignment scheme

A. Knebe, ''PM Codes'' slides available at:
http://popia.ft.uam.es/aknebe/page3/files/ComputationalCosmology/07PMcodes.pdf

PM Method: Details

Benefits:

- full potential, no cutoff
- standard FFT routines can be used (FFTW)
- accuracy down to small distances of the order of the grid spacing r_c. We choose $r_c = R_0 = 10\mu m$, the colloids radius.

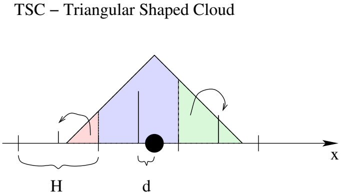


PM Method: Details

discretize density $\vec{r}_i \to \rho(\vec{g}_k)$:

- construct density with "Triangular Shaped Cloud" (TSC) Method (9 neighbor cells) $\longrightarrow$ smoothing of the density

1D mass assignment function:



$$\vec{d} = \vec{r}_i - \vec{g}_k$$

$$\rho(\vec{g}_k) = \frac{mW(d_x)W(d_y)}{H^2}$$

$$W(d) = \begin{cases} \frac{3}{4} - \left(\frac{d}{H}\right)^2 & d \leq \frac{H}{2} \\ \frac{1}{2}\left(\frac{3}{2} - \frac{d}{H}\right)^2 & \frac{H}{2} < d \leq \frac{3H}{2} \\ 0 & otherwise \end{cases}$$

PM Method: Details

discretized version of the greens function:

$$G(k_{lm}) = \frac{1}{-\sin^2(\frac{k_x}{2}) - \sin^2(\frac{k_y}{2}) - \frac{1}{\lambda^2}}$$

with

$$k_x = \frac{2\pi l}{L}, \quad k_y = \frac{2\pi m}{L}$$

and $G(k_{l=0,m=0}) = 0$ (avoids the singularity at the origin for $\lambda \to \infty$).

PM Method: Details

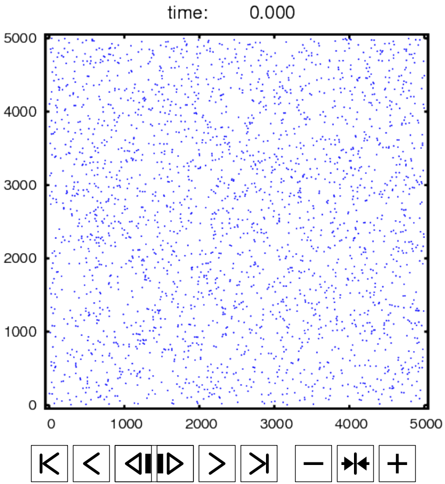
Forces, Interpolation, time step:

- after $\imath k$–differentiation: interpolate forces (inverse CiC or inverse TSC)
- calculate additional (repulsive) short–ranged forces (direct sum)
- calculate maximum possible time step
  - requirement: displacement due to random velocity process small (avoids large repelling forces $\rightarrow$ hard core of WCA potential)
  - correlation of the random process:

$$\langle r_i(t), r_j(t+\Delta t)\rangle = \sqrt{2D\Delta t}\,\delta_{ij}$$

  - choose Δt such that displacement due to forces $\sim 3\mu m$ $\longrightarrow$ random displacement mostly of same order (compromise!)
- then: integration of the equations of motion

$\lambda/L = 0.02$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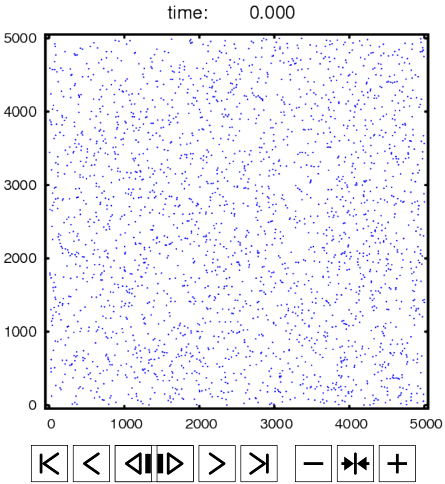
5000
4000
3000
2000
1000
0
0
1000
2000
3000
4000
5000

$\lambda/L = 10$



time: 0.000
5000
4000
3000
2000
1000
0
0
1000
2000
3000
4000
5000

Minkowski functionals

Problem: Definition of clusters is arbitrary $\longrightarrow$ need a more systematic approach!

Minkowski functionals in integral Geometry:

$$M_\nu \sim \frac{\omega_{2-\nu}}{\omega_\nu \omega_2} W_\nu(A), \quad \nu = 0, \cdots, d \tag{10}$$

ω_d: volume of a d-dimensional unit sphere
$W_0(A)$: Volume of A, $W_1(A)$: its surface
$W_\nu(A)$: for $\nu \geq 2$ defined as surface integrals over the boundary of a compact domain A and the ν-th elementary symmetric polynomial of its principal radii of curvature
In 2D: only one principal radius of curvature:

$$M_0 = F, \quad M_1 = U/2\pi, \quad M_2 = \chi/\pi$$

χ: Euler-characteristic (measure of connectivity)

Minkowski functionals

K. R. Mecke, ‘‘Additivity, Convexity, and Beyond: Applications of Minkowski Functionals in Statistical Physics’’ in: Statistical Physics and Spatial Statistics, Springer, Berlin (2000)

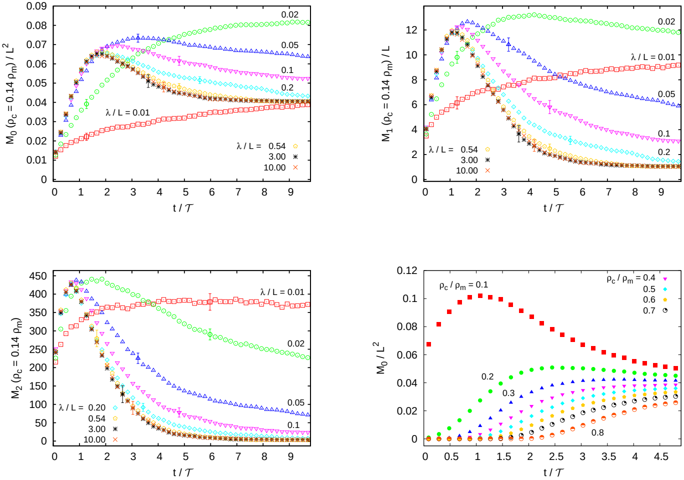
need: compact domain $A \longrightarrow$ use a threshold density:

$$A = \left\{ \bigcup_i A_i \,|\, \varrho(A_i) > \varrho_c \right\}$$

- study the Minkowski functionals as function of ϱ_c
- easy to implement with existing density grid ($\rightarrow$ pixels)
- harder to interpret (?)
- M_2 (average curvature) serves as measure for the number of clusters (clusters without holes)
  $\longrightarrow \chi =$ number of clusters minus number of holes
- no theory for dynamical evolution available

Evolution of M_0, M_1 and M_2

choose $\varrho_c = 0.14$ (in units of maximum packing density ϱ_m)



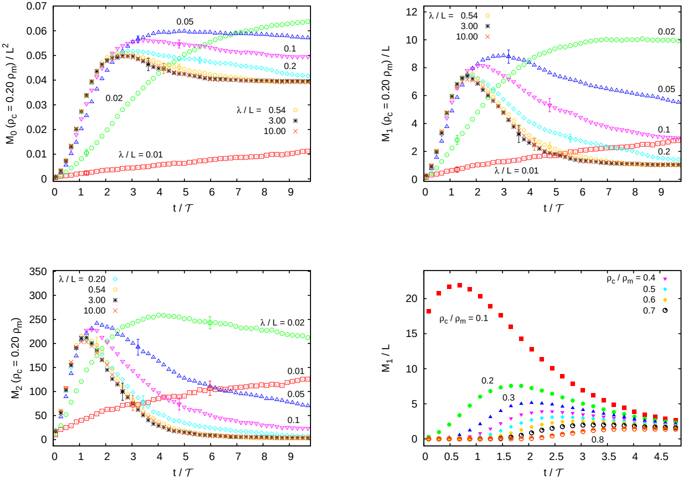
⟶ qualitatively similar to previous findings (scaling holds)

Cosmology in a petri dish? Simulation of collective dynamics of colloids at fluid interfaces

Johannes Bleibel

Evolution of M_0, M_1 and M_2

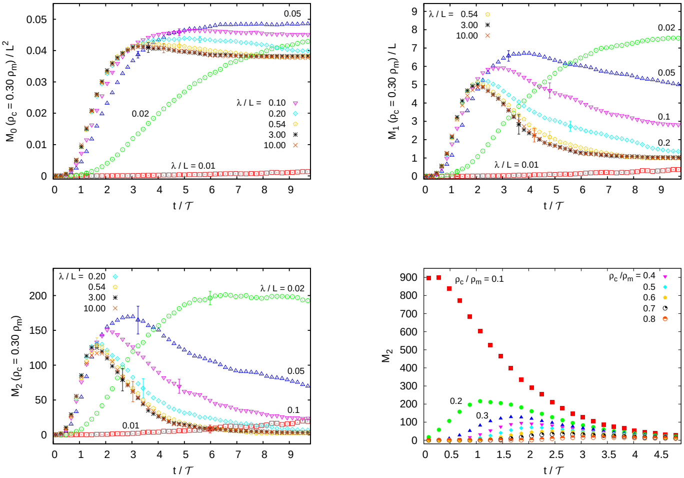
choose $\varrho_c = 0.20$ (in units of maximum packing density ϱ_m)



⟶ qualitatively similar to previous findings (scaling holds)

Evolution of M_0, M_1 and M_2

choose $\varrho_c = 0.30$ (in units of maximum packing density ϱ_m)



⟶ qualitatively similar to previous findings (scaling holds)

Hydrodynamic interactions

- So far: collective Brownian behavior of a dilute patch
- Overdamped dynamics – appropriate for microparticles
- Include hydrodynamic interactions perturbatively on the two-particle level

On the individual particle level (pair-terms only):

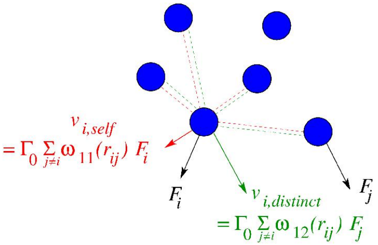
$$\vec{v}_i = \mathbf{D}_{ij}\vec{F}_j^{ext} + noise$$

$$\mathbf{D}_{ij} = \Gamma_0 \mathbb{1}\delta_{ij} + \mathbf{D}^{(2)}(\vec{r}_i - \vec{r}_j), \quad \Gamma_0 = \frac{1}{6\pi\eta a}$$

Self and distinct interaction terms:

$$\mathbf{D}^{(2)}(\vec{r}_{ij}) = \Gamma_0 \left[\delta_{ij}\sum_{i\neq l}\omega_{11}(\vec{r}_{il}) + (1-\delta_{ij})\omega_{12}(\vec{r}_{ij})\right]$$

Hydrodynamic interactions



- neglect self term ($\omega_{11}(\vec{r}) \propto r^4$)
- use bulk Rotne Prager Tensor for distinct part:

$$\omega_{12}(\vec{r}) = \frac{3}{4}\frac{a}{r}(\mathbb{1} + \hat{\vec{r}}\hat{\vec{r}}) + \frac{1}{2}\frac{a^3}{r^3}(\mathbb{1} - 3\hat{\vec{r}}\hat{\vec{r}}) \qquad (11)$$

Stokesian Dynamics:

- Superposition of stokeslets: solution for stokes equation for velocity field $\vec{u}(\vec{r})$ of the 3D fluid:

$$\eta \nabla^2 \vec{u} - \nabla p = -\delta(\vec{r})\vec{F}, \quad \nabla \cdot \vec{u} = 0 \qquad (12)$$

Collective evolution with HI

Mass conservation

$$\frac{\partial \rho}{\partial t} = \nabla(\rho \vec{v})$$

now includes velocities

$$\vec{v}(\vec{r}, t) = \Gamma_0 \vec{F}(\vec{r}, t) + \vec{u}(\vec{r}, t)$$

with the fluid velocity field $\vec{u}(\vec{r}, t)$ determined self–consistently via

$$\eta \nabla^2 \vec{u} - \nabla p = -\rho \vec{F} \qquad (13)$$

Formal solution:

$$\vec{u}(\vec{r}, t) = \frac{1}{8\pi\eta} \int d\vec{r}' \mathbf{G}(\vec{r} - \vec{r}') \rho(\vec{r}') \vec{F}(\vec{r}') \qquad (14)$$

with the corresponding Green's function $\mathbf{G}(\vec{r} - \vec{r}')$.

Stability analysis with HI

Bulk: Kirkwood tensor

$$\mathbf{G}(\vec{r}_{ij}) = \Gamma_0 \left[\mathbb{1}\delta_{ij} + (1-\delta_{ij})\omega_{12}(\vec{r}_{ij}) \right]$$

DDFT by Rex & Löwen (PRL 2008):

$$\frac{\partial \rho}{\partial t} = \Gamma_0 \nabla \left(\rho \vec{F}(\vec{r}) + \int d\vec{r}' \rho^{(2)}(\vec{r},\vec{r}')\omega_{12}(\vec{r}-\vec{r}')\vec{F}(\vec{r}') \right) \tag{15}$$

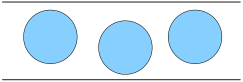
linear stability analysis:

- same result, modified only by a hydrodynamic factor:

$$\frac{\partial \delta\tilde{\rho}(\vec{k},t)}{\partial t} \sim (1 + \rho_0 \hat{\vec{k}} \tilde{\omega}_{12} \hat{\vec{k}}) \delta\tilde{\rho}(\vec{k},t) \tag{16}$$

- fluctuations grow/ decrease exponentially

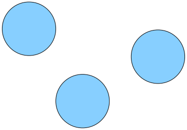
A "hidden" dimension



2D Colloids
2D Fluid

$$\omega_{12}(\vec{r}) \sim \left(\mathbb{1} \log \frac{L}{r} + \frac{\vec{r}\vec{r}}{r^2} \right) + \cdots$$

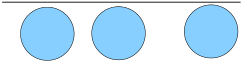
$$\xrightarrow{2D\ FT} \hat{\vec{k}} \tilde{\omega}_{12} \hat{\vec{k}} \sim 0 \cdot \mathcal{O}(k^{-2}) + \cdots$$



3D Colloids
3D Fluid

$$\omega_{12}(\vec{r}) \sim \frac{1}{r} \left(\mathbb{1} + \frac{\vec{r}\vec{r}}{r^2} \right) + \cdots$$

$$\xrightarrow{3D\ FT} \hat{\vec{k}} \tilde{\omega}_{12} \hat{\vec{k}} \sim 0 \cdot \mathcal{O}(k^{-2}) + \cdots$$



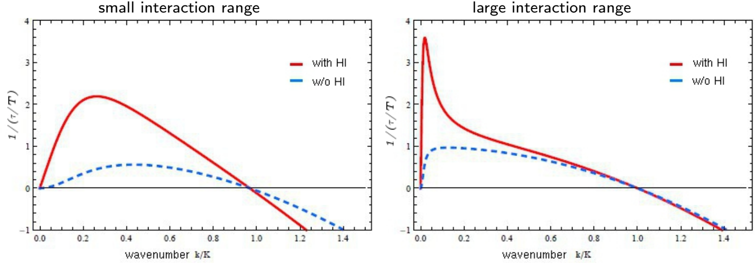
2D Colloids
3D Fluid

$$\omega_{12}(\vec{r}) \sim \frac{1}{r} \left(\mathbb{1} + \frac{\vec{r}\vec{r}}{r^2} \right) + \cdots$$

$$\xrightarrow{2D\ FT} \hat{\vec{k}} \tilde{\omega}_{12} \hat{\vec{k}} \sim \frac{1}{k} + \cdots$$

Hydrodynamic acceleration

exponential growth/ damping: additional k–dependence



- exponent $\mathcal{T}/\tau(k)$ divergent for small k ($\sim 1/k$)
- drastic enhancement for small wavenumbers – large scales

$\Longrightarrow$ Hydrodynamics speed up the collapse

Results from simulations

- Stokesian dynamics at Rotne Prager level
- $R_0 = 10\mu$m, $\lambda = 2.7$mm (large interaction range)
- Bulk system, fastest growing mode: $k/K_j = 0.044$