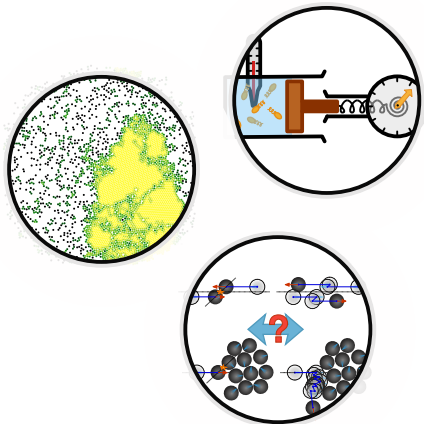


Thermodynamic nature of irreversibility in active matter

Lennart Dabelow



with Ralf Eichhorn (Nordita)

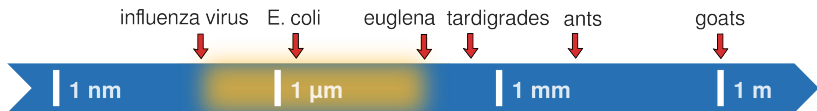


21 October 2024

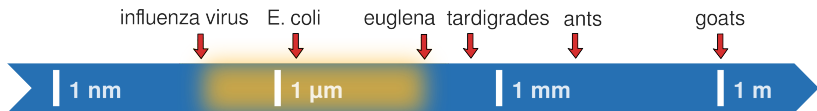
Nordita program "Measuring and manipulating nonequilibrium systems"

- particles/entities that can **consume energy** from their environment and use it to **move persistently and autonomously**

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- emerging collective behavior
(clustering, swarming, communication, ...)

Modeling active matter at mesoscopic scales

- N interacting, spherical Brownian particles in $d = 2$ or 3 which convert energy from surroundings into directed motion

$$\dot{\mathbf{x}}_i(t) = \frac{1}{\gamma} \mathbf{F}_i(\mathbf{x}(t)) + \sqrt{2D} \boldsymbol{\xi}_i(t) + \mathbf{v}_i(t)$$

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- hard-core contact interactions via steep pair potential $U(r)$,

$$\mathbf{F}_i(\mathbf{x}) = - \sum_{j(\neq i)} \mathbf{F}(\mathbf{x}_i - \mathbf{x}_j), \quad \mathbf{F}(\mathbf{r}) = -U'(|\mathbf{r}|) \frac{\mathbf{r}}{|\mathbf{r}|},$$

such that $U(r) \equiv 0$ if $r > 2R$ (R : particle radius)

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- collisions with surrounding molecules lead to
 - dissipation (friction coefficient γ)
 - random forces (“thermal fluctuations”), modeled by Gaussian white-noise processes:

$$\langle \boldsymbol{\xi}_i(t) \rangle = 0$$

$$\langle \xi_i^\mu(t) \xi_j^\nu(t') \rangle = \delta_{ij} \delta_{\mu\nu} \delta(t - t')$$

- fluctuation-dissipation relation: $D = k_B T / \gamma$
- overdamped approximation:
inertia negligible compared to friction/dissipation

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- self-propulsion also erratic, but persistent
- $\mathbf{v}_i(t)$ independent random processes (“active fluctuations”) with correlation function

$$\langle v_i^\mu(t) v_j^\nu(t') \rangle = \delta_{ij} \delta_{\mu\nu} \frac{D_a}{\tau_a} e^{-|t-t'|/\tau_a}$$

- τ_a : correlation time
 - D_a : “active diffusion coefficient” (speed/intensity of self-propulsion)
- effective, phenomenological model, disregarding details of self-propulsion mechanism

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→ entropy introduces a (thermodynamic) “arrow of time”

- **irreversibility**: log-ratio of path probabilities for forward- and backward-in-time trajectories/histories,

$$\Sigma := \ln \frac{P[\{\mathbf{x}(t)\}_{t=0}^{\tau}]}{P[\{\tilde{\mathbf{x}}(t)\}_{t=0}^{\tau}]}, \quad \tilde{\mathbf{x}}(t) := \mathbf{x}(\tau - t)$$

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$$\dot{\mathbf{x}}_i(t) = \frac{1}{\gamma} \mathbf{F}_i(\mathbf{x}(t)) + \sqrt{2D} \xi_i(t) + \cancel{\mathbf{v}_i(t)}$$

→ Σ related to **thermodynamic entropy** S via stochastic energetics and fluctuation theorems [Seifert, PRL 95 (2005)]:

$$k_B \langle \Sigma \rangle = \frac{Q}{T} = S \quad (Q: \text{dissipated heat})$$

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- how does Σ relate to thermodynamics in active matter?

Irreversibility in active matter

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- explicitly known for active Ornstein-Uhlenbeck particles
(when $\mathbf{v}(t)$ is an Ornstein-Uhlenbeck process):

$$\Sigma = \frac{1}{k_B T} \int_0^{\tau} dt \int_0^{\tau} dt' \sum_{i=1}^N \dot{\mathbf{x}}_i(t) \cdot \mathbf{F}_i(t') [\delta(t - t') - \Gamma(t, t')] + \Delta S_{\text{sys}}$$

[LD, Bo, Eichhorn, PRX **9** (2019)]

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- steady-state “irreversibility production rate”:

$$\sigma := \lim_{\tau \rightarrow \infty} \frac{\langle \Sigma \rangle}{\tau}$$

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with $\Gamma(t, t') \sim \frac{\tau_* D_a / D}{2\tau_a^2} e^{-|t-t'|/\tau_*}$ asymptotically ($\tau \rightarrow \infty$),

$$\tau_* = \frac{\tau_a}{\sqrt{1 + D_a/D}}$$

Irreversibility for AOUPs

- explicitly known for active Ornstein-Uhlenbeck particles (when $\mathbf{v}(t)$ is an Ornstein-Uhlenbeck process):

$$\Sigma = \frac{1}{k_B T} \int_0^\tau dt \int_0^\tau dt' \sum_{i=1}^N \dot{\mathbf{x}}_i(t) \cdot \mathbf{F}_i(t') [\delta(t-t') - \Gamma(t, t')] + \Delta S_{\text{sys}}$$

[LD, Bo, Eichhorn, PRX 9 (2019)]

with $\Gamma(t, t') \sim \frac{\tau_* D_a/D}{2\tau_a^2} e^{-|t-t'|/\tau_*}$ asymptotically ($\tau \rightarrow \infty$),

$$\tau_* = \frac{\tau_a}{\sqrt{1+D_a/D}}$$

- partial integration and $\tau \rightarrow \infty$:

$$\sigma = \frac{C}{\gamma} \int_0^\infty ds e^{-s/\tau_*} \sum_{i=1}^N [\langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0+s) \rangle - \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0-s) \rangle],$$

where t_0 is an arbitrary reference time and $C := \frac{D_a}{2D^2\tau_a^2}$

- active systems:

$$\dot{\mathbf{x}}_i(t) = \frac{1}{\gamma} \mathbf{F}_i(\mathbf{x}(t)) + \sqrt{2D} \boldsymbol{\xi}_i(t) + \mathbf{v}_i(t)$$

- **pressure** p : average force per unit area exerted by the particles on the container walls

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- caveat: not always a state function [Solon *et al*, Nat Phys **11** (2015)], but here it is

Pressure in active matter

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- **pressure** p : average force per unit area exerted by the particles on the container walls
- caveat: not always a state function [Solon *et al*, Nat Phys 11 (2015)], but here it is
- three contributions: $p = p_o + p_{\text{int}} + p_a$
 - osmotic/ideal-gas pressure: $p_o = Nk_B T/V$
 - interaction pressure: $p_{\text{int}} = \frac{1}{dV} \sum_i \langle \mathbf{x}_i \cdot \mathbf{F}_i(\mathbf{x}) \rangle$
 - active or **swim pressure**: $p_a = \frac{\gamma}{dV} \sum_i \langle \mathbf{x}_i \cdot \mathbf{v}_i \rangle$

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- low density ($\phi \ll 1$):

$$\sigma = \left(\frac{2^{d-2} V_p}{k_B \tau_a} \right) \frac{N p_a}{T} [1 + O(\phi)]$$

(V_p : particle volume; $\phi = \frac{NV_p}{V}$: packing fraction)

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- high activity (MIPS regime): relation holds approximately
- low density *and* high activity:

$$\sigma = \left(\frac{2^{d-2} V_p}{k_B \tau_a} \right) \frac{N p}{T} [1 + O(\phi, Pe^{-1})]$$

Derivation

$$\sigma = \frac{C}{\gamma} \int_0^\infty ds e^{-s/\tau_*} \sum_{i=1}^N [\langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0+s) \rangle - \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0-s) \rangle]$$

key ingredients and assumptions:

- exactly known expression for σ for AOUPs
- particles are hard spheres (steep interaction potential)
- “most” interactions are pair collisions ($\phi \ll 1$)

main steps:

- replace interaction forces by self-propulsion in correlator:
$$\sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle = -\frac{2^{d-2}\gamma}{d} \phi \sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{v}_i(t_0 \pm s) \rangle [1 + O(\phi)]$$
- evaluate s integral using Novikov's theorem:

$$\int_0^\infty ds e^{-s/\tau_*} \langle \mathbf{x}_i(t_0) \cdot \mathbf{v}_i(t_0 \pm s) \rangle = \frac{\tau_a}{\sqrt{1 + \frac{D_a}{D}} \pm 1} \langle \mathbf{x}_i(t_0) \cdot \mathbf{v}_i(t_0) \rangle$$

Rewriting $\sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle$

- recall: $\mathbf{F}_i = \sum_{j(\neq i)} \mathbf{F}(\mathbf{x}_i - \mathbf{x}_j)$ with $\mathbf{F}(-\mathbf{r}) = -\mathbf{F}(\mathbf{r})$

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$$\dot{\mathbf{r}}_{ij} = \frac{1}{\gamma} [\mathbf{F}_i - \mathbf{F}_j] + \alpha_{ij}$$

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- focus on **pair collisions** (e.g., low particle density):
at any given time, a fixed particle i interacts at most with one other particle j

$$\rightarrow \dot{\mathbf{r}}_{ij} = \frac{2}{\gamma} \mathbf{F}(\mathbf{r}_{ij}) + \alpha_{ij}$$

Rewriting $\sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle$ (2)

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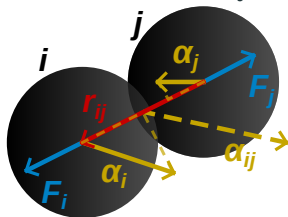
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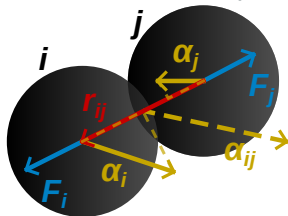
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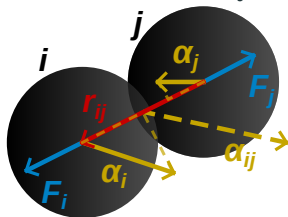
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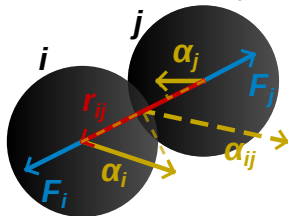
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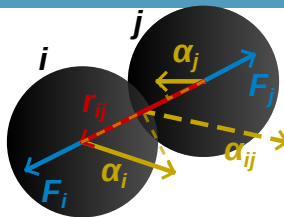
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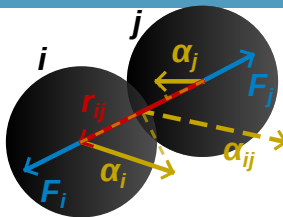


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- inspect

$$\begin{aligned} & \langle \mathbf{r}_{ij}(t_0) \cdot \boldsymbol{\alpha}_{ij}^{\parallel}(t_0 \pm s) \rangle \chi(r_{ij}(t_0 \pm s)) \\ &= \langle [\mathbf{r}_{ij}(t_0) \cdot \mathbf{n}_{ij}(t_0 \pm s)] [\boldsymbol{\alpha}_{ij}(t_0 \pm s) \cdot \mathbf{n}_{ij}(t_0 \pm s)] \chi(r_{ij}(t_0 \pm s)) \rangle \end{aligned}$$



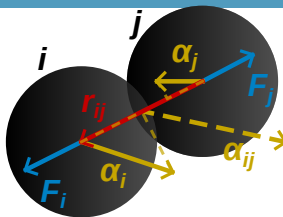
Rewriting $\sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle$ (3)

$$\rightarrow \sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle = -\frac{\gamma}{2} \sum_{i < j} \langle \mathbf{r}_{ij}(t_0) \cdot \boldsymbol{\alpha}_{ij}^{\parallel}(t_0 \pm s) \rangle \chi(r_{ij}(t_0 \pm s))$$

- inspect

$$\begin{aligned} & \langle \mathbf{r}_{ij}(t_0) \cdot \boldsymbol{\alpha}_{ij}^{\parallel}(t_0 \pm s) \rangle \chi(r_{ij}(t_0 \pm s)) \\ & = \langle [\mathbf{r}_{ij}(t_0) \cdot \mathbf{n}_{ij}(t_0 \pm s)] [\boldsymbol{\alpha}_{ij}(t_0 \pm s) \cdot \mathbf{n}_{ij}(t_0 \pm s)] \chi(r_{ij}(t_0 \pm s)) \rangle \end{aligned}$$

- *before* collision, particles move around freely, hence orientations of \mathbf{r}_{ij} and $\boldsymbol{\alpha}_{ij}$ approximately uncorrelated



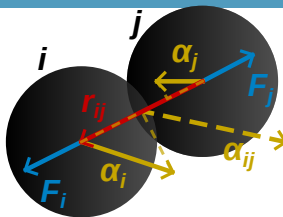
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- *before* collision, particles move around freely, hence orientations of \mathbf{r}_{ij} and $\boldsymbol{\alpha}_{ij}$ approximately uncorrelated
- to *initiate* and *maintain* collision, need $\mathbf{n}_{ij} \cdot \boldsymbol{\alpha}_{ij} < 0$



Rewriting $\sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle$ (3)

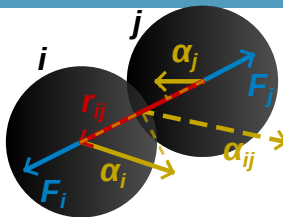
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$$\begin{aligned} \rightarrow & \langle \mathbf{r}_{ij}(t_0) \cdot \boldsymbol{\alpha}_{ij}^{\parallel}(t_0 \pm s) \rangle \chi(r_{ij}(t_0 \pm s)) \\ & \approx \frac{1}{2d} \langle \mathbf{r}_{ij}(t_0) \cdot \boldsymbol{\alpha}_{ij}(t_0 \pm s) \rangle \chi(r_{ij}(t_0 \pm s)) \quad \text{up to errors } O\left(\frac{V_p}{V}\right) \end{aligned}$$



Rewriting $\sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle$ (3)

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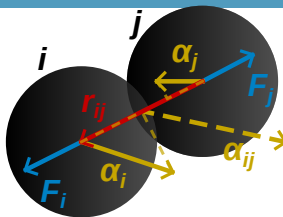
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$$\begin{aligned} \rightarrow \sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle \\ = -\frac{\gamma}{4d} \sum_{i < j} \langle \mathbf{r}_{ij}(t_0) \cdot \boldsymbol{\alpha}_{ij}(t_0 \pm s) \rangle \chi(r_{ij}(t_0 \pm s)) \end{aligned}$$



Rewriting $\sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle$ (4)

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- switch back to absolute positions and fluctuations:

$$\begin{aligned} & \sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle \\ & = -\frac{\gamma}{2d} \sum_{i < j} [\langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_i(t_0 \pm s) \rangle \chi(r_{ij}(t_0 \pm s)) \\ & \quad - \langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_j(t_0 \pm s) \rangle \chi(r_{ij}(t_0 \pm s))] \end{aligned}$$

Rewriting $\sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle$ (4)

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- $\langle \dots \rangle$ is steady-state average over all pair configurations, but $\chi(r_{ij}(t_0 \pm s))$ picks out those where particles overlap

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- $\langle \dots \rangle$ is steady-state average over all pair configurations, but $\chi(r_{ij}(t_0 \pm s))$ picks out those where particles overlap
- to leading order (up to corrections $O(\frac{V_p}{V})$), pair distribution is uniform

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- switch back to absolute positions and fluctuations:

$$\sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle$$

$$= -\frac{\gamma}{2d} \sum_{i < j} [\langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_i(t_0 \pm s) \rangle \chi(r_{ij}(t_0 \pm s)) \\ - \langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_j(t_0 \pm s) \rangle \chi(r_{ij}(t_0 \pm s))]]$$

- $\langle \dots \rangle$ is steady-state average over all pair configurations, but $\chi(r_{ij}(t_0 \pm s))$ picks out those where particles overlap
- to leading order (up to corrections $O(\frac{V_p}{V})$), pair distribution is uniform

$\rightarrow \chi(r_{ij})$ picks out “interaction volume” (sphere of radius $2R$):

$$\langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_{i/j}(t_0 \pm s) \rangle \chi(r_{ij}(t_0 \pm s))$$

$$= \frac{2^d V_p}{V} \langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_{i/j}(t_0 \pm s) \rangle [1 + O(\frac{V_p}{V})]$$

Rewriting $\sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle$ (5)

- $\sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle$
 $= -\frac{\gamma}{2d} \sum_{i < j} [\langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_i(t_0 \pm s) \rangle \chi(r_{ij}(t_0 \pm s))$
 $\quad - \langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_j(t_0 \pm s) \rangle \chi(r_{ij}(t_0 \pm s))]$
- $\langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_{i/j}(t_0 \pm s) \rangle \chi(r_{ij}(t_0 \pm s))$
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- $\sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle$
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- $\langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_{i/j}(t_0 \pm s) \rangle \chi(r_{ij}(t_0 \pm s))$
 $= \frac{2^d V_p}{V} \langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_{i/j}(t_0 \pm s) \rangle [1 + O(\frac{V_p}{V})]$
- $\sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle$
 $= -\frac{2^{d-1} V_p \gamma}{dV} \sum_{i < j} [\langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_i(t_0 \pm s) \rangle$
 $\quad - \langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_j(t_0 \pm s) \rangle] [1 + O(\frac{V_p}{V})]$
- packing fraction: $\phi = \frac{NV_p}{V}$

Rewriting $\sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle$ (5)

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Rewriting $\sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle$ (5)

- $$\sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle$$

$$= -\frac{\gamma}{2d} \sum_{i < j} [\langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_i(t_0 \pm s) \rangle \chi(r_{ij}(t_0 \pm s))$$

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- $$\rightarrow \sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle$$

$$= -\frac{2^{d-1} V_p \gamma}{dV} \sum_{i < j} [\langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_i(t_0 \pm s) \rangle$$

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- $\sum_i \langle \mathbf{x}_i(t_0) \cdot \mathbf{F}_i(t_0 \pm s) \rangle = -\frac{2^{d-2} \gamma}{d} \phi \sum_i \langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_i(t_0 \pm s) \rangle [1 + O(\phi)]$

Integrate over time delay s

- recall: $\sigma = \frac{C}{\gamma} \int_0^\infty ds e^{-s/\tau_*} \sum_i \langle \mathbf{x}_i(t_0) \cdot [\mathbf{F}_i(t_0+s) - \mathbf{F}_i(t_0-s)] \rangle$

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→ to leading order in ϕ ,

$$\sigma = -\frac{2^{d-2}C}{d} \phi \sum_i \int_0^\infty ds e^{-s/\tau_*} [\langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_i(t_0+s) \rangle - \langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_i(t_0-s) \rangle]$$

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- Novikov's theorem [Novikov, Sov. Phys. JETP 20 (1965)]:
 - unbiased Gaussian stochastic process $\alpha(t)$
 - functional $\mathcal{F}[\alpha](t_0)$ of $\alpha(t)$ up to time t_0
 - $\langle \mathcal{F}[\alpha](t_0) \alpha(t) \rangle = \int_0^{t_0} ds \langle \alpha(t) \alpha(s) \rangle \left\langle \frac{\delta \mathcal{F}[\alpha](t_0)}{\delta \alpha(s)} \right\rangle$

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- recall: $\sigma = \frac{C}{\gamma} \int_0^\infty ds e^{-s/\tau_*} \sum_i \langle \mathbf{x}_i(t_0) \cdot [\mathbf{F}_i(t_0+s) - \mathbf{F}_i(t_0-s)] \rangle$

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→ with $\alpha(t) = \alpha_i^\mu(t)$ and $\mathcal{F}[\alpha](t_0) = x_i^\mu(t_0)$:

$$\int_0^\infty ds e^{-s/\tau_*} \langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_i(t_0 \pm s) \rangle = \frac{\tau_a}{\sqrt{1 + \frac{D_a}{D} \pm 1}} \langle \mathbf{x}_i(t_0) \cdot \mathbf{v}_i(t_0) \rangle$$

Integrate over time delay s

- recall: $\sigma = \frac{C}{\gamma} \int_0^\infty ds e^{-s/\tau_*} \sum_i \langle \mathbf{x}_i(t_0) \cdot [\mathbf{F}_i(t_0+s) - \mathbf{F}_i(t_0-s)] \rangle$

→ to leading order in ϕ ,

$$\sigma = -\frac{2^{d-2}C}{d} \phi \sum_i \int_0^\infty ds e^{-s/\tau_*} [\langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_i(t_0+s) \rangle - \langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_i(t_0-s) \rangle]$$

- Novikov's theorem [Novikov, Sov. Phys. JETP 20 (1965)]:

- unbiased Gaussian stochastic process $\alpha(t)$

- functional $\mathcal{F}[\alpha](t_0)$ of $\alpha(t)$ up to time t_0

→ $\langle \mathcal{F}[\alpha](t_0) \alpha(t) \rangle = \int_0^{t_0} ds \langle \alpha(t) \alpha(s) \rangle \left\langle \frac{\delta \mathcal{F}[\alpha](t_0)}{\delta \alpha(s)} \right\rangle$

→ with $\alpha(t) = \alpha_i^\mu(t)$ and $\mathcal{F}[\alpha](t_0) = x_i^\mu(t_0)$:

$$\int_0^\infty ds e^{-s/\tau_*} \langle \mathbf{x}_i(t_0) \cdot \boldsymbol{\alpha}_i(t_0 \pm s) \rangle = \frac{\tau_a}{\sqrt{1 + \frac{D_a}{D} \pm 1}} \langle \mathbf{x}_i(t_0) \cdot \mathbf{v}_i(t_0) \rangle$$

→ $\sigma = \phi \frac{2^{d-2}}{d\tau_a D} \sum_i \langle \mathbf{x}_i \cdot \mathbf{v}_i \rangle = \left(\frac{2^{d-2} V_p}{k_B \tau_a} \right) \frac{N p_a}{T}$

High-activity limit

- main assumption:
irreversibility predominantly produced during pair collisions,
justified for small ϕ

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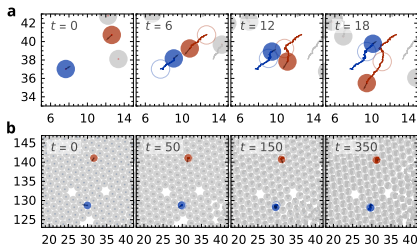
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Numerical simulations

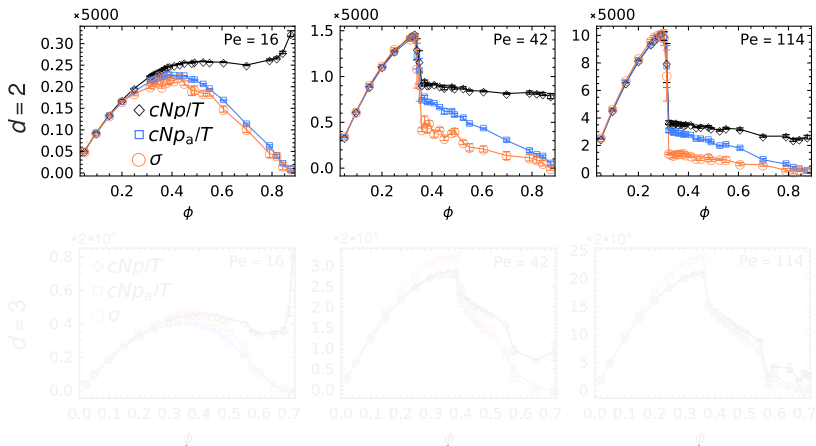
low density ($\phi \ll 1$): high activity ($\text{Pe} \gg 1/\text{MIPS}$):

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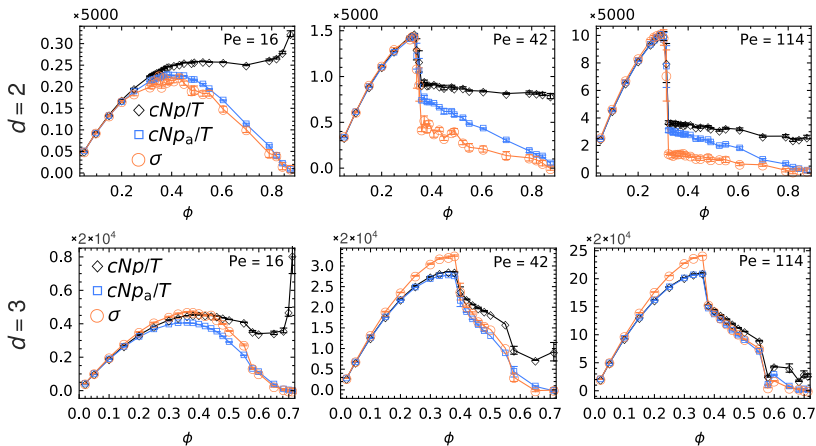
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arXiv:2308.03625

Thank you!

Phase diagram ($d = 2$)

