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Rapid growth of large aggregates by correlated successive coalescences

joint work with
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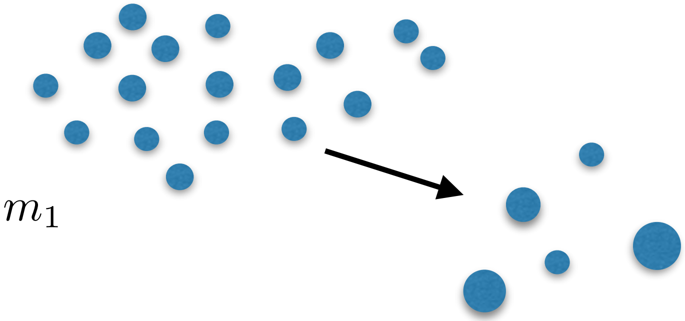
Growth by coalescences

In both cases: very **dilute** solid particles suspended in a **turbulent** gas

Initially: almost mono-disperse size distribution

monomers with mass $\approx m_1$



How fast are large aggregates/drops created?

Time-evolution of the number $N_i(t)$ of particles with mass $i\, m_1$?

Coagulation kinetics

Standard approach: **Smoluchowski** kinetics:

$$i\, m_1 + j\, m_1 \xrightarrow{\mathcal{K}_{i,j}} (i+j)\, m_1$$

$\mathcal{K}_{i,j}$: collision kernel between particles with masses *i* and *j*

$$\boxed{\frac{\mathrm{d}}{\mathrm{d}t} N_i = \frac{1}{2} \sum_{j=1}^{i-1} \mathcal{K}_{j,i-j}\, N_j\, N_{i-j} - \sum_{j=1}^{\infty} \mathcal{K}_{j,i}\, N_j\, N_i}$$

Short times: $N_1(t) \approx N_1(0)$ and creations are dominant

$$\frac{\mathrm{d}}{\mathrm{d}t} N_2 \approx \frac{1}{2} \mathcal{K}_{1,1}\, N_1^2 \ \Rightarrow N_2 \propto t$$

$$\frac{\mathrm{d}}{\mathrm{d}t} N_3 \approx \mathcal{K}_{1,2}\, N_1\, N_2 \propto t \ \Rightarrow N_3 \propto t^2$$

$$\frac{\mathrm{d}}{\mathrm{d}t} N_i \approx \frac{1}{2} \sum_{j} \mathcal{K}_{j,i-j}\, N_j\, N_{i-j} \quad \Rightarrow \quad \boxed{N_i \propto t^{i-1}}$$

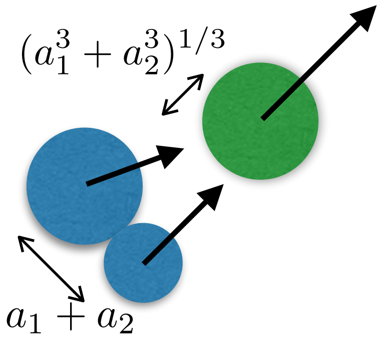
The exponent
does not depend
on the kernel

Direct numerical simulations

Heavy inertial (point) particles with an effective size

$$\ddot{\boldsymbol{X}} = -\frac{1}{\tau_\mathrm{p}}\left[\dot{\boldsymbol{X}} - \boldsymbol{u}(\boldsymbol{X}, t)\right] + \boldsymbol{g}$$

$$\tau_\mathrm{p} = \frac{2\,\rho_\mathrm{p}\,a^2}{9\,\nu\,\rho_\mathrm{f}}$$

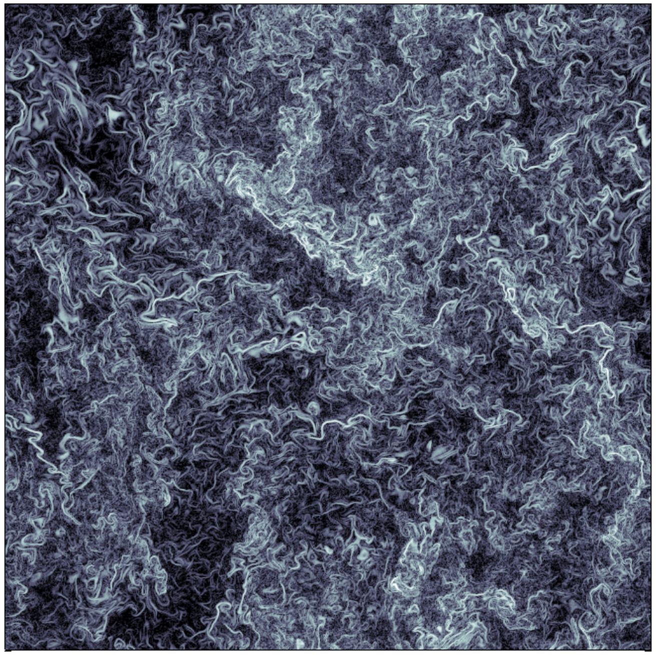


Coalescences upon touching, conserving mass and momentum

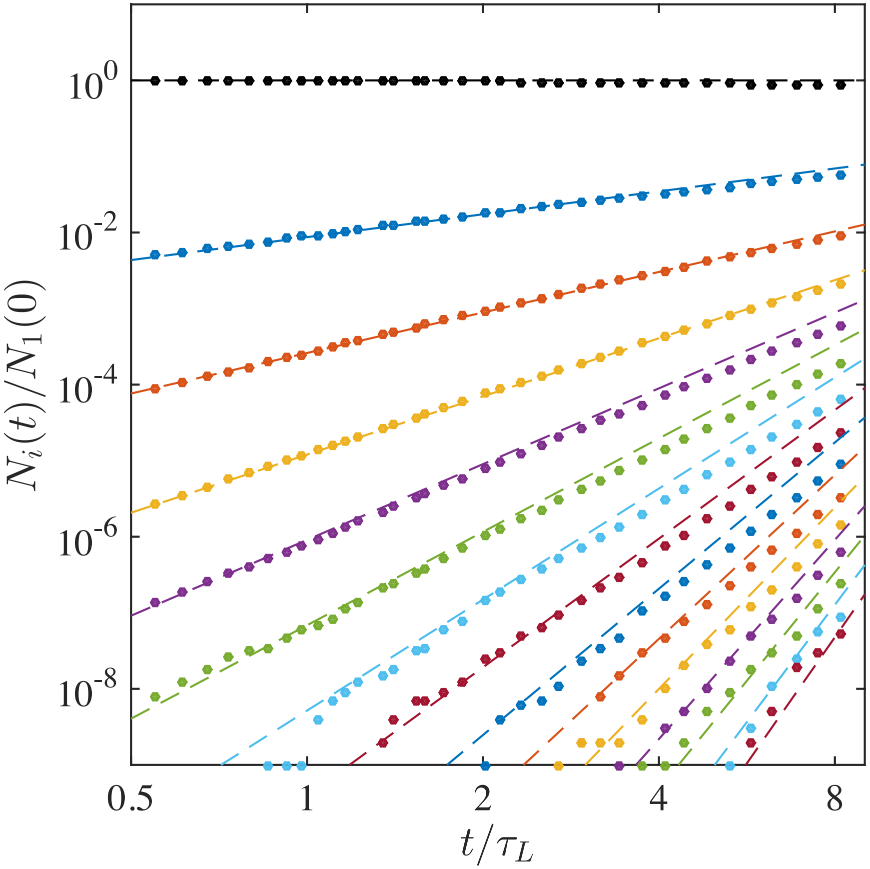
Start from a population of monomers with mass m_1

Numerics: incomprssible Navier–Stokes pseudo-spectral 2048^3 ($R_\lambda \approx 460$) initially 10^9 particles
$a_1 \approx \eta/10$ ($St \approx 0.1$ – weak inertia)

Very dilute: volume fraction $\Phi_v \approx 5 \cdot 10^{-5}$
$\approx$ 1 particle per box of size $10\eta^3$



Short-time growth of large particles



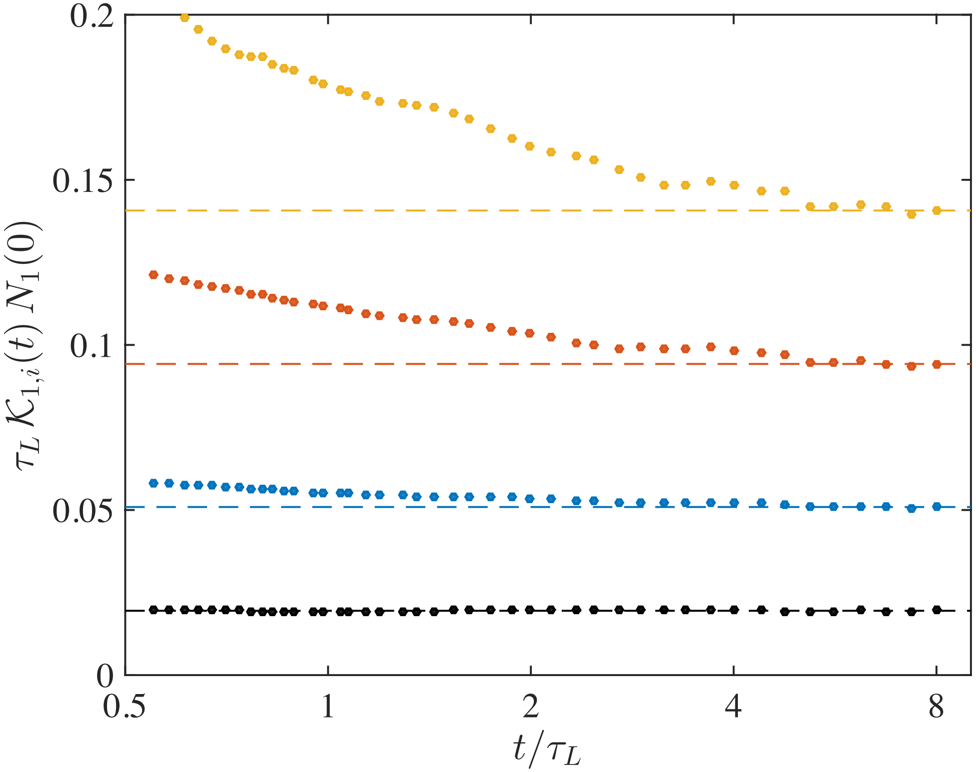
Data suggests: $N_i(t) \propto t^{0.75(i-2)+1}$
at short times

⇒ faster than the expected t^{i-1}

Measured collision rate is not constant

Mean-field kinetics not valid at short times?

⇒ **Correlations between collisions?**



Time evolution of the size distribution

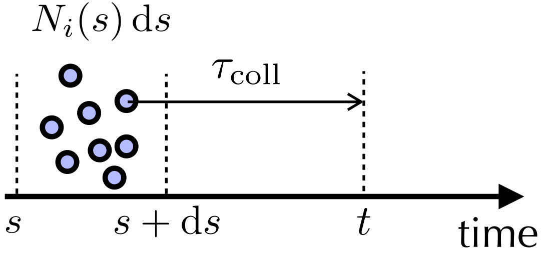
- Back to basics:

Exact equation

$$\boxed{\frac{\mathrm{d}}{\mathrm{d}t}N_i = \frac{1}{2}\sum_{j=1}^{i-1}\phi_{i-j,j}(t) - \sum_{j=1}^{\infty}\phi_{i,j}(t)}$$

$\phi_{i,j}(t)\,\mathrm{d}t$ average number of collisions $i+j$ in $[t, t+\mathrm{d}t]$

- Expression for the collision rate:



$$\phi_{i,j}(t) = -\int_0^t \frac{\mathrm{d}}{\mathrm{d}t}\left[\log\int_t^{\infty} \underset{\uparrow}{p(\tau, j|s,i)}\,\mathrm{d}\tau\right]\dot N_i(s)\,\mathrm{d}s$$

joint density of the collision time τ
and the size j of the collider

- Time to next collision: exponential distribution **with non-constant rate**

$$p(\tau, j|s,i) = \lambda_{i,j}(\tau,s)\,\mathrm{e}^{-\int_s^{\tau}\lambda_{i,j}(\tau',s)\mathrm{d}\tau'} \Rightarrow \boxed{\phi_{i,j}(t) = \int_0^t \lambda_{i,j}(t,s)\,\dot N_i(s)\,\mathrm{d}s}$$

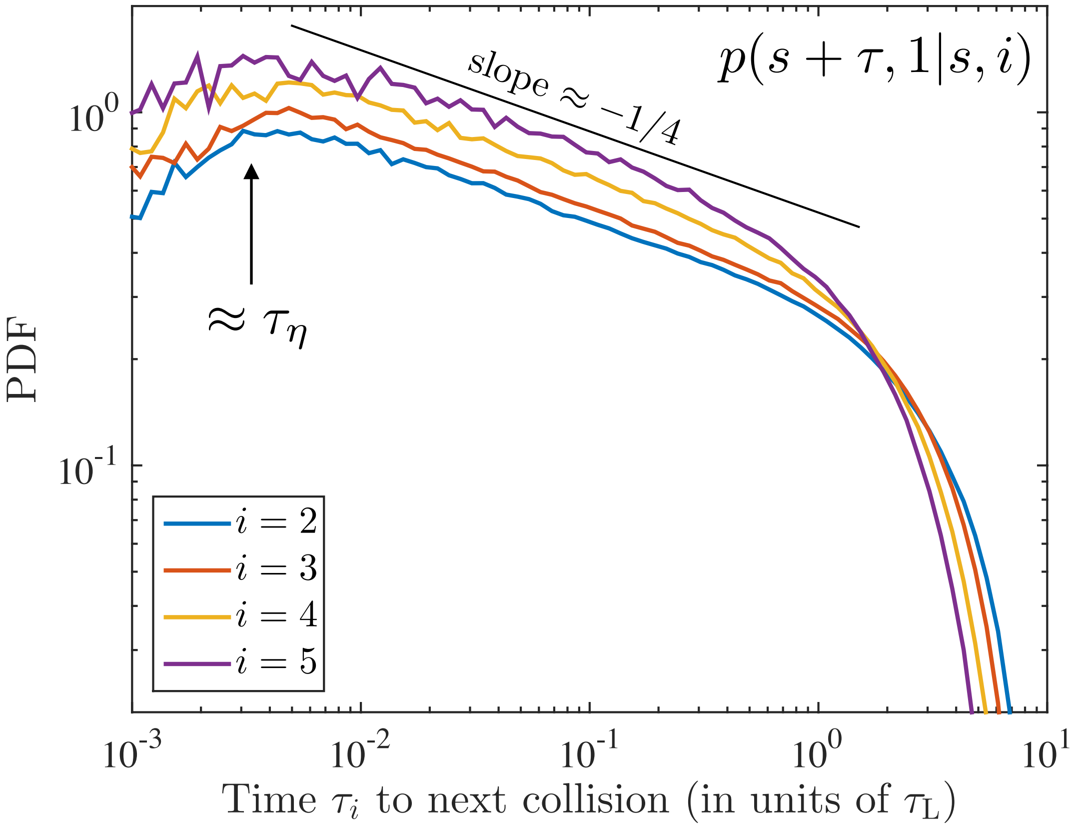
- **“Mean-field rate”:** $\lambda_{i,j}(t,s) = \mathcal{K}_{i,j}\,N_j(t)$

$$\Rightarrow\ \phi_{i,j}(t) = \mathcal{K}_{i,j}\,N_j(t)\int_0^t \dot N_i(s) = \mathcal{K}_{i,j}\,N_j(t)\,N_i(t)$$

Smoluchowski

Long-range correlated collisions

Probability distribution of particles **mean-free times** (inter-collision times)



$$p(s+\tau, j|s, i) \sim \tau^{-\alpha} \mathrm{e}^{-c\,\tau^{1-\alpha}}$$

$$\boxed{\lambda_{i,j}(t,s) \sim |t-s|^{-\alpha}} \qquad \alpha \approx 1/4$$

Consequences on size evolution:

$$\mathcal{K}_{1,1} = \text{const} \qquad N_1(t) \approx \text{const}$$

$$\Rightarrow \quad \dot{N}_2 = \frac{1}{2}\mathcal{K}_{1,1} N_1^2$$

$$\frac{\mathrm{d}N_3}{\mathrm{d}t} \simeq \phi_{1,2} = \int_0^t \dot{N}_2(s)\lambda_{1,2}(t,s)\mathrm{d}s$$

$$\simeq \frac{1}{2}\mathcal{K}_{1,1} N_1^2 \int_0^t |t-s|^{-\alpha}\mathrm{d}s$$

$$\Rightarrow N_3(t) \propto t^{2-\alpha}$$

$$\alpha \approx 0.25 \Rightarrow N_3(t) \propto t^{1.75}$$

$$\frac{\mathrm{d}N_i}{\mathrm{d}t} \approx \int_0^t \frac{\mathrm{d}}{\mathrm{d}s} N_{i-1}(s)\; p(t, 1|s, i-1)\,\mathrm{d}s \quad \Rightarrow \quad \boxed{N_i(t) \propto t^{i-1-\alpha(i-2)}}$$

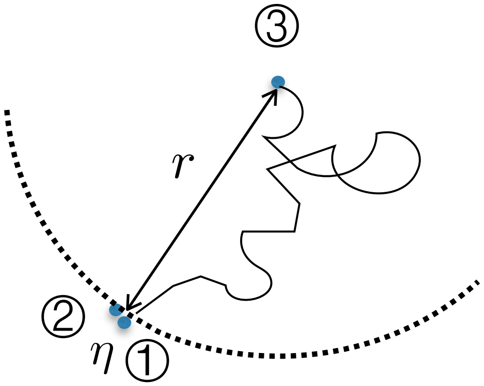
Dimensional estimates

$p(s+\tau, j|s, i) \sim \tau^{-\alpha}$ when $\tau_\eta \ll \tau \ll \tau_L$ ⇒ inertia is negligible

⇒ Purely due to turbulent mixing?

Naive phenomenology for the distribution of inter-collision times:

Assume particles collide with a given probability once they have approached at a distance $\lesssim \eta$



Two contributions:

Number density of particles ③ at distance r:

$$n(r) = r^2 N_1 / L^3$$

Probability that a particle ③ initially at distance r approaches at a distance η from the newly created ①+②:

$$p(\eta, t|r, 0) \sim \underset{\underset{\text{solid angle}}{\uparrow}}{\left(\frac{\eta}{r}\right)^2} \frac{1}{t^{3/2}} \Psi \underset{\underset{\text{Richardson scaling}}{\uparrow}}{\left(\frac{r}{t^{3/2}}\right)}$$

Approaching rate:

$$\lambda(t) \propto \int u_\eta \, p(\eta, t|r, 0)\, n(r)\, \mathrm{d}r \sim \frac{N_1 \eta^2 u_\eta}{L^3} \int \Psi\left(\frac{r}{t^{3/2}}\right) \frac{\mathrm{d}r}{t^{3/2}} = \text{const}$$

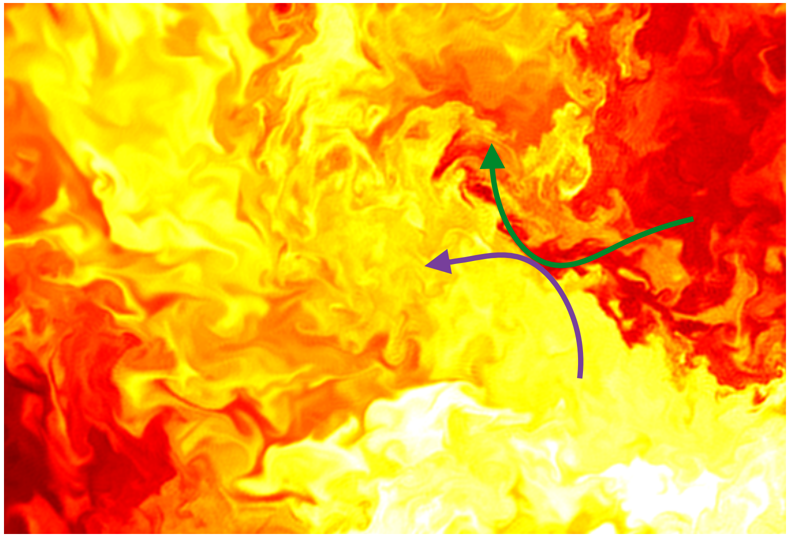
not a power law!

Anomalies in turbulent mixing

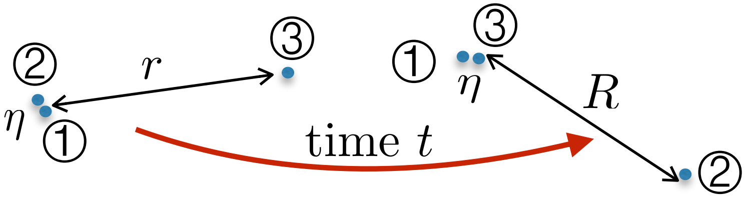
Advection of a passive scalar

$$\partial_t\theta + \boldsymbol{v}\cdot\nabla\theta = \kappa\nabla^2\theta + \Phi$$

$$\Rightarrow \quad \langle(\delta_R\theta)^q\rangle \sim R^{\zeta_q} \quad \rightsquigarrow q\text{-point motion}$$



In our case: three-point motion



Transition probability

$$p_3(R,\eta,t\,|\,\eta,r,0) \sim \left(\frac{\eta}{r}\right)^{3-\zeta_3}_{\uparrow} \frac{1}{t^3}\,\Phi\left(\frac{R}{t^{3/2}},\frac{r}{t^{3/2}}\right)$$

compare with 2 for $\zeta_3 < 1$:
approaching events are less
probable when $r/\eta \to \infty$

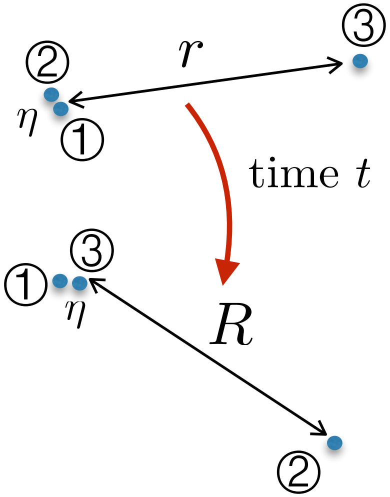
Related to "Lagrangian statistical conservation laws"

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle \underset{\substack{\uparrow\\ \text{size}}}{\mathcal{R}^{\zeta_q}}\, \underset{\substack{\uparrow\\ \text{shape}}}{f_q}(\hat{\boldsymbol{R}}_1,\ldots,\hat{\boldsymbol{R}}_{q-1})\rangle = 0$$

Bernard, Gawedzki, Kupiainen, J. Stat. Phys. (1997)
Shraiman & Siggia, Nature (2000)
Celani & Vergassola, PRL (2001)

Actual rates

Collision rate: $\lambda(t) \propto \int u_\eta \, p_3(R, \eta, t \,|\, \eta, r, 0) \, n(r) \, \mathrm{d}R \, \mathrm{d}r$



Again two contributions:

$$\begin{cases} n(r) = r^2 N_1 / L^3 \\ \text{(unchanged)} \\ p_3(R, \eta, t \,|\, \eta, r, 0) \sim \left(\frac{\eta}{r}\right)^{3-\zeta_3} \frac{1}{t^3} \, \Phi\left(\frac{R}{t^{3/2}}, \frac{r}{t^{3/2}}\right) \\ \text{(enhanced for small } r \text{)} \end{cases}$$

$$\lambda(t) \propto \frac{1}{t^3} \int r^{\zeta_3 - 1} \Phi\left(\frac{R}{t^{3/2}}, \frac{r}{t^{3/2}}\right) \mathrm{d}R \, \mathrm{d}r \propto t^{\frac{3}{2}(\zeta_3 - 1)}$$

$$\zeta_3 \approx 0.82 \;\Rightarrow\; \alpha = \frac{3}{2}(1 - \zeta_3) \approx 0.27 \qquad \Rightarrow \qquad \boxed{N_i(t) \propto t^{0.73(i-2)+1}}$$

Summary

Kinetic approaches for coagulation fails at short times

- As a consequence, the number of large particles grows as $N_i(t) \propto t^{0.75\,i}$ and not t^i

- “Rapid” successive collisions/reactions are correlated (mean-field breaks) when they involve inertial-range physics. This is a purely turbulent-mixing effect

- Can one modify kinetic models (via multiple collisions) to account for that?