Weierstrass Institute for
Applied Analysis and Stochastics

Coagulation equations and particle systems

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Plan of the talk

1. Introduction
   - Smoluchowski's coagulation equation
   - systems of Brownian particles
2. Spatially homogeneous particle models
   - direct simulation model
   - general interactions
   - asymptotic behavior
   - mass flow model
3. Phase transition
   - gelation
   - explosion

Smoluchowski's coagulation equation

$$\frac{d}{dt}\,c(t,x) = \frac{1}{2}\sum_{y=1}^{x-1} K(x-y,y)\,c(t,x-y)\,c(t,y) - \sum_{y=1}^{\infty} K(x,y)\,c(t,x)\,c(t,y)$$

- average number concentration

$$c(t,x) \qquad t \geq 0 \qquad x = 1,2,\ldots$$

- coagulation kernel

$$\begin{aligned} K(x,y) &= \Big(D(x)+D(y)\Big)\Big(R(x)+R(y)\Big) \\ &\sim \Big(x^{-1/3}+y^{-1/3}\Big)\Big(x^{1/3}+y^{1/3}\Big) \end{aligned}$$

[M. v. Smoluchowski. Drei Vorträge über Diffusion, Brownsche Molekularbewegung und Koagulation von Kolloidteilchen. Phys. Z., 17:557–571, 585–599, 1916.]

$$\frac{\partial}{\partial t}\, c(t,r,x) = D(x)\,\Delta_r\, c(t,r,x) + \frac{1}{2}\sum_{y=1}^{x-1} K(x-y,y)\, c(t,r,x-y)\, c(t,r,y) - \sum_{y=1}^{\infty} K(x,y)\, c(t,r,x)\, c(t,r,y)$$

- n particles move in $\mathbb{R}^3$ according to Brownian motion
- at distance ε (sphere of influence) they stick together
- consider $N(t,B,x)\,,\quad t \geq 0\,,\quad B \subset \mathbb{R}^3\,,\quad x = 1,2,\ldots$
- let $n \to \infty$ so that $n\,\varepsilon = \mathrm{const}$
- if $\frac{1}{n}\, N(0,B,x) \;\to\; \int_B c(0,r,x)\, dr \qquad \forall\, B \subset \mathbb{R}^3\,,\quad x = 1,2,\ldots$ then

$$\frac{1}{n}\, N(t,B,x) \;\to\; \int_B c(t,r,x)\, dr \qquad \forall\, t > 0\,,\quad B \subset \mathbb{R}^3\,,\quad x = 1,2,\ldots$$

Convergence results - 2

$$\frac{\partial}{\partial t}\, c(t,r,x) = D(x)\,\Delta_r\, c(t,r,x) +$$
$$\frac{1}{2}\sum_{y=1}^{x-1} K(x-y,y)\, c(t,r,x-y)\, c(t,r,y) - \sum_{y=1}^{\infty} K(x,y)\, c(t,r,x)\, c(t,r,y)$$

- [R. Lang and X. Nguyen. Smoluchowski's theory of coagulation in colloids holds rigorously in the Boltzmann–Grad limit. Z. Wahrsch. Verw. Gebiete, 54:227–280, 1980.]
  - $D(x) = 1 \qquad R(x) = 1 \qquad K(x,y) = 1$
  - special case (unlabeled particles), $m_0(t,r) = \sum_{x=1}^{\infty} c(t,r,x)$

$$\frac{\partial}{\partial t}\, m_0(t,r) = \Delta_r\, m_0(t,r) - \frac{1}{2}\, m_0(t,r)^2$$

Convergence results - 3

$$\frac{\partial}{\partial t} c(t,r,x) = D(x)\,\Delta_r\, c(t,r,x) + \frac{1}{2}\sum_{y=1}^{x-1} K(x-y,y)\, c(t,r,x-y)\, c(t,r,y) - \sum_{y=1}^{\infty} K(x,y)\, c(t,r,x)\, c(t,r,y)$$

$$K(x,y) = \Big(D(x)+D(y)\Big)\Big(R(x)+R(y)\Big)$$

- [J. R. Norris. Brownian coagulation. *Commun. Math. Sci.*, 2(1):93–101, 2004.]

$$D(x) = x^{-1/3} \qquad R(x) = x^{1/3}$$

- [F. Rezakhanlou. The coagulating Brownian particles and Smoluchowski's equation. *Markov Proc. Rel. Fields*, 12:425–445, 2006.]

$R(x) = x^{\alpha} \qquad \alpha \in [0,1) \qquad$ + spatial smoothing

$$\Big(X_1(t), \ldots, X_{N(t)}(t)\Big) \qquad t \geq 0 \qquad N(0) = n$$

time evolution

1. distribution of the waiting time

$$\text{Prob}(\tau \geq t) = \exp(-\lambda(z)\, t)$$

   where

$$\lambda(z) = \frac{1}{n} \sum_{1 \leq i < j \leq N} K(x_i, x_j) \qquad z = (x_1, \ldots, x_N)$$

2. distribution of the indices

$$i, j \quad \sim \quad K(x_i, x_j)$$

3. replace x_i, x_j by $x_i + x_j$ and go to 1.

[Marcus (1968), Gillespie (1972), Lushnikov (1978)]

$$\Big(X_1(t),\ldots,X_{N(t)}(t)\Big) \qquad t \ge 0 \qquad N(0)=n$$

- state space of a single particle $\mathcal{X}$
- state space of the system

$$\mathcal{Z} = \Big\{(x_1,\ldots,x_N) \in \mathcal{X}^N \,:\, N \ge 0\Big\}$$

- interactions

$$x_{i_1},\ldots,x_{i_l} \quad \Rightarrow \quad \big(z_1,\ldots,z_k\big) = z \in \mathcal{Z}$$

- intensities

$$q_l(x_1,\ldots,x_l,dz) \qquad l=0,1,\ldots,L$$

Asymptotic behavior

- empirical measures

$$\mu^{(n)}(t,dx) = \frac{1}{n}\sum_{i=1}^{N(t)} \delta_{X_i(t)}(dx) \qquad t \ge 0$$

- scaling of intensities $q_0, q_1, \ldots, q_L$
- probability distributions $P^{(n)} \in \mathcal{P}(\mathcal{D}([0,\infty), \mathcal{M}_b^+(\mathcal{X})))$

Theorem

Assumptions on $q_0, \ldots, q_L$ and

$$\mu^{(n)}(0,dx) \;\to\; f_0(dx) \qquad \text{for some} \quad f_0 \in \mathcal{M}_b^+(\mathcal{X}))\,.$$

Then

- $(P^{(n)})$ is relatively compact
- accumulation points are concentrated on solutions of $(*)$

Limiting equation

$$\int_{\mathcal{X}} \varphi(x)\, f(t,dx) = \int_{\mathcal{X}} \varphi(x)\, f_0(dx) + \int_0^t \mathcal{G}(\varphi, f(s))\, ds \qquad t \geq 0 \qquad (*)$$

- test functions $\varphi \in \mathcal{C}_c(\mathcal{X})$
- nonlinear operator

$$\mathcal{G}(\varphi,\nu) = \int_{\mathcal{Z}} \Big[\varphi(z_1) + \ldots + \varphi(z_k)\Big] q_0(dz) + \sum_{l=1}^{L} \int_{\mathcal{X}} \nu(dx_1) \ldots \int_{\mathcal{X}} \nu(dx_l) \times$$
$$\int_{\mathcal{Z}} \Big[\varphi(z_1) + \ldots + \varphi(z_k) - \varphi(x_1) - \ldots - \varphi(x_l)\Big] q_l(x_1, \ldots, x_l, dz)$$

Corollary 1 (existence)

For each $P = \lim_{k\to\infty} P^{(n_k)}$,

$$P(\{f \,:\, (*) \text{ holds}\}) = 1\,.$$

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Corollary 2 (convergence)

Assume uniqueness.

$$\Rightarrow \quad P(\{f\}) = 1 \qquad \forall\, P = \lim_{k\to\infty} P^{(n_k)}$$

$$\Rightarrow \quad \lim_{n\to\infty} P^{(n)} = \delta_f$$

$$\sim \quad \lim_{n\to\infty} \mu^{(n)}(t,dx) = f(t,dx) \qquad \forall\, t \ge 0$$

$$\int_{\mathcal{X}} \varphi(x)\, f(t,dx) = \int_{\mathcal{X}} \varphi(x)\, f_0(dx) + \int_0^t \mathcal{G}(\varphi, f(s))\, ds \qquad (*)$$

$$\mathcal{G}(\varphi,\nu) =$$
$$\int_{\mathcal{X}} \nu(dx_1) \int_{\mathcal{X}} \nu(dx_2) \int_{\mathcal{Z}} \Big[\varphi(z_1) + \ldots + \varphi(z_k) - \varphi(x_1) - \varphi(x_2)\Big] q_2(x_1,x_2,dz)$$

- state space $\mathcal{X} = \{1\}$ $\qquad q_2 = 1$
- $1,1 \;\Rightarrow\; 1$ ("unlabeled" coagulation)

$$\frac{d}{dt}\, c(t) = -c(t)^2 \qquad\qquad c(t) = \frac{1}{1+t}$$

- $1,1 \;\Rightarrow\; \emptyset$ ("annihilating" coagulation)

$$\frac{d}{dt}\, c(t) = -2\, c(t)^2$$

Example 2 - direct simulation process

- state space

$$\mathcal{X} = \{1, 2, \dots\} \quad \text{or} \quad \mathcal{X} = (0, \infty)$$

- coagulation jumps

$$x_{i_1}, x_{i_2} \quad \Rightarrow \quad z_1$$

- intensity

$$q_2(x_1, x_2, dz) = \frac{1}{2} K(x_1, x_2)\, \delta_{x_1+x_2}(dz)$$

- nonlinear operator

$$\mathcal{G}(\varphi, \nu) = \frac{1}{2} \int_{\mathcal{X}} \nu(dx_1) \int_{\mathcal{X}} \nu(dx_2) \Big[\varphi(x_1 + x_2) - \varphi(x_1) - \varphi(x_2)\Big] K(x_1, x_2)$$

Smoluchowski's coagulation equation (weak form)

$$\frac{d}{dt} \int_{\mathcal{X}} \varphi(x)\, f(t, dx) = \mathcal{G}(\varphi, f(t))$$

Another stochastic coagulation model

$$\left(\tilde{X}_1(t), \ldots, \tilde{X}_n(t)\right) \qquad t \geq 0 \qquad \tilde{N}(t) = \tilde{N}(0) = n$$

time evolution

1. distribution of the waiting time

$$\mathsf{Prob}(\tilde{\tau} \geq t) = \exp(-\tilde{\lambda}(z)\, t)$$

where

$$\tilde{\lambda}(z) = \frac{1}{n} \sum_{1 \leq i,\, j \leq n} \frac{K(x_i, x_j)}{x_j} \qquad z = (x_1, \ldots, x_n)$$

2. distribution of the indices

$$i, j \quad \sim \quad \frac{K(x_i, x_j)}{x_j}$$

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Another stochastic coagulation model - 2

- state space

$$\mathcal{X} = \{1, 2, \ldots\} \quad \text{or} \quad \mathcal{X} = (0, \infty)$$

- coagulation jumps

$$x_{i_1}, x_{i_2} \quad \Rightarrow \quad z_1, z_2$$

- intensity

$$q_2(x_1, x_2, dz) = \frac{K(x_1, x_2)}{x_2}\, \delta_{x_1+x_2}(dz_1)\, \delta_{x_2}(dz_2)$$

- nonlinear operator

$$\tilde{\mathcal{G}}(\varphi, \nu) = \int_{\mathcal{X}} \nu(dx_1) \int_{\mathcal{X}} \nu(dx_2) \Big[\varphi(x_1 + x_2) - \varphi(x_1)\Big] \frac{K(x_1, x_2)}{x_2}$$

limiting equation

$$\frac{d}{dt} \int_{\mathcal{X}} \varphi(x)\, \tilde{f}(t, dx) = \tilde{\mathcal{G}}(\varphi, \tilde{f}(t))$$

Equivalence of limiting equations

first model

$\frac{d}{dt} \int_{\mathcal{X}} \varphi(x)\, f(t,dx) = \mathcal{G}(\varphi, f(t))$

second model

$\frac{d}{dt} \int_{\mathcal{X}} \psi(x)\, \tilde{f}(t,dx) = \tilde{\mathcal{G}}(\psi, \tilde{f}(t))$

$$
\begin{aligned}
\tilde{\mathcal{G}}(\psi,\nu) \quad &= \int_{\mathcal{X}} \nu(dx_1) \int_{\mathcal{X}} \nu(dx_2) \Big[\psi(x_1+x_2) - \psi(x_1)\Big] \frac{K(x_1,x_2)}{x_2} \\
&= \int_{\mathcal{X}} \frac{\nu(dx_1)}{x_1} \int_{\mathcal{X}} \frac{\nu(dx_2)}{x_2} \Big[\psi(x_1+x_2) - \psi(x_1)\Big] x_1\, K(x_1,x_2) \\
&= \int_{\mathcal{X}} \frac{\nu(dx_1)}{x_1} \int_{\mathcal{X}} \frac{\nu(dx_2)}{x_2} \Big[\psi(x_1+x_2) - \psi(x_2)\Big] x_2\, K(x_1,x_2) \\
&= \frac{1}{2} \int_{\mathcal{X}} \frac{\nu(dx_1)}{x_1} \int_{\mathcal{X}} \frac{\nu(dx_2)}{x_2} K(x_1,x_2) \times \\
&\quad \Big[(x_1+x_2)\,\psi(x_1+x_2) - x_1\,\psi(x_1) - x_2\,\psi(x_2)\Big] = \mathcal{G}(x\,\psi, \nu/x)
\end{aligned}
$$

Equivalence of limiting equations

first model	second model
$\frac{d}{dt}\int_{\mathcal{X}} \varphi(x)\, f(t,dx) = \mathcal{G}(\varphi, f(t))$	$\frac{d}{dt}\int_{\mathcal{X}} \psi(x)\, \tilde{f}(t,dx) = \tilde{\mathcal{G}}(\psi, \tilde{f}(t))$

$$
\begin{aligned}
\tilde{\mathcal{G}}(\psi,\nu) &= \int_{\mathcal{X}} \nu(dx_1) \int_{\mathcal{X}} \nu(dx_2) \Big[\psi(x_1+x_2) - \psi(x_1)\Big] \frac{K(x_1,x_2)}{x_2} \\
&= \int_{\mathcal{X}} \frac{\nu(dx_1)}{x_1} \int_{\mathcal{X}} \frac{\nu(dx_2)}{x_2} \Big[\psi(x_1+x_2) - \psi(x_1)\Big] x_1\, K(x_1,x_2) \\
&= \int_{\mathcal{X}} \frac{\nu(dx_1)}{x_1} \int_{\mathcal{X}} \frac{\nu(dx_2)}{x_2} \Big[\psi(x_1+x_2) - \psi(x_2)\Big] x_2\, K(x_1,x_2) \\
&= \frac{1}{2} \int_{\mathcal{X}} \frac{\nu(dx_1)}{x_1} \int_{\mathcal{X}} \frac{\nu(dx_2)}{x_2} K(x_1,x_2) \times \\
&\quad \Big[(x_1+x_2)\,\psi(x_1+x_2) - x_1\,\psi(x_1) - x_2\,\psi(x_2)\Big] = \mathcal{G}(x\,\psi, \nu/x)
\end{aligned}
$$

so that $\varphi(x) = x\,\psi(x)$ and

$$\tilde{f}(t,dx) = x\, f(t,dx) \quad \Rightarrow \quad \text{mass flow model}$$

Smoluchowski's coagulation equation

$$\frac{d}{dt}\, c(t,x) = \frac{1}{2}\sum_{y=1}^{x-1} K(x-y,y)\, c(t,x-y)\, c(t,y) - \sum_{y=1}^{\infty} K(x,y)\, c(t,x)\, c(t,y)$$

$K(x,y)$	$m_0(t)$	$m_1(t)$	$m_2(t)$
1	$\frac{2}{2+t}$	1	$1+t$
$x+y$	$\exp(-t)$	1	$\exp(2\,t)$

$$m_k(t) = \sum_{x=1}^{\infty} x^k\, c(t,x)$$

$$t \geq 0 \qquad k = 0,1,2,\ldots$$

$$c(0,x) = \delta_{1,x}$$

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$$t \geq 0 \qquad k = 0,1,2,\ldots$$

$$c(0,x) = \delta_{1,x}$$

$K(x,y) \leq \mathrm{const}\,(x+y) \quad \Rightarrow \quad$ unique solution + mass conservation

$$K(x,y) = x\,y$$

$t \in$	$[0,1)$	$[1,\infty)$
$m_0(t)$	$1-\frac{t}{2}$	$\frac{1}{2t}$
$m_1(t)$	1	$\frac{1}{t}$
$m_2(t)$	$\frac{1}{1-t}$	∞

$$m_k(t) = \sum_{x=1}^{\infty} x^k\, c(t,x)$$

$$c(0,x) = \delta_{1,x}$$

Gelation

$$K(x,y) = x\,y$$

$t \in$	$[0,1)$	$[1,\infty)$
$m_0(t)$	$1-\frac{t}{2}$	$\frac{1}{2t}$
$m_1(t)$	1	$\frac{1}{t}$
$m_2(t)$	$\frac{1}{1-t}$	∞

$$m_k(t) = \sum_{x=1}^{\infty} x^k\, c(t,x)$$

$$c(0,x) = \delta_{1,x}$$

gelation time

$$t_{\text{gel}} = \inf\left\{ t \geq 0 \;:\; m_1(t) < m_1(0) \right\}$$

Gelling coagulation kernels

- heuristics

- a homogeneous kernel

$$K(\alpha\, x, \alpha\, y) = \alpha^{\varepsilon}\, K(x,y) \qquad \forall\, \alpha > 0$$

  is gelling, if $\varepsilon > 1$ (homogeneity exponent)

- rigorous results

- if

$$K(x,y) \geq C\,(x\,y)^{a} \qquad \text{for some} \quad a > 0.5\,, \quad C > 0\,, \quad \text{and} \quad K(x,y) = o(x\,y)\,,$$

  then there exist gelling solutions [Jeon (1998)]

- if

$$K(x,y) = C\left(x^{a}\, y^{b} + x^{b}\, y^{a}\right) \qquad \text{for some} \quad a, b \in [0,1]\,, \; a + b > 1\,, \; C > 0\,,$$

  then every weak solution is gelling [Escobedo/Mischler/Perthame (2002)]

- $\mathcal{Y}$ - locally compact separable metric
- $q(y, d\tilde{y})$ - compactly bounded kernel
- $Y_0, Y_1, Y_2, \ldots$ - Markov chain $\sim \frac{1}{\lambda(y)}\, q(y, d\tilde{y})\,, \qquad \lambda(y) = q(y, \mathcal{Y})$
- $\lambda(Y_0), \lambda(Y_1), \lambda(Y_2), \ldots$ - waiting time parameters
- $\tau_1 < \tau_2 < \tau_3 < \ldots$ - jump times

explosion time

$\tau_\infty = \lim_{l\to\infty} \tau_l$

minimal jump process

process on $\mathcal{Y} \cup \{\Delta\}$ (one-point compactification)

$$Y^\Delta(t) = \begin{cases} Y_l & : \ \tau_l \le t < \tau_{l+1} \\ \Delta & : \ t \ge \tau_\infty \end{cases}$$

- recall

kernel $q(y, d\tilde{y})$ $\lambda(y) = q(y, \mathcal{Y})$ $\tau_\infty = \lim_{l \to \infty} \tau_l$ Markov chain (Y_k)

- regular: $\text{Prob}(\tau_\infty = \infty) = 1$ explosive: $\text{Prob}(\tau_\infty < \infty) > 0$

criterion

$$\text{regularity} \quad \Leftrightarrow \quad \sum_{k=0}^{\infty} \frac{1}{\lambda(Y_k)} = \infty \quad \text{a.s.}$$

- λ bounded $\Rightarrow$ regularity
- explosion $\sim$ λ unbounded & $Y_k \to \Delta$ with appropriate speed

Direct simulation process

$$\Big(X_1(t), \ldots, X_{N(t)}(t)\Big) \qquad t \geq 0 \qquad N(0) = n$$

- $\mathcal{Y} = \bigcup_{N=1}^{n} \mathcal{X}^N \qquad \mathcal{X} = \{1, 2, \ldots\}$ or $(0, \infty) \qquad y = (x_1, \ldots, x_N)$
- $\lambda(y) = \frac{1}{2n} \sum_{1 \leq i \neq j \leq N} K(x_i, x_j) \qquad x_i, x_j \quad \Rightarrow \quad x_i + x_j$
- $N(t) \searrow$ and $X_i(t) \nearrow$ bounded $\Rightarrow$ no explosion

$$\big(X_1(t), \dots, X_{N(t)}(t)\big) \qquad t \ge 0 \qquad N(0) = n$$

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largest component for $K(x, y) = x\, y$

$$M_1^{(n)}(t) \sim \begin{cases} \log n & , \text{ if } t < 1 \\ n^{\frac{2}{3}} & , \text{ if } t = 1 \\ n & , \text{ if } t > 1 \end{cases}$$

$\Rightarrow$ random graph theory [Erdös/Rényi (1960), Bollobás (1985, 2001)]

Mass flow process

$$\Big(\tilde{X}_1(t),\ldots,\tilde{X}_n(t)\Big) \qquad t \geq 0 \qquad \tilde{N}(t) = \tilde{N}(0) = n$$

- $\mathcal{Y} = \mathcal{X}^n \qquad \mathcal{X} = \{1,2,\ldots\}$ or $(0,\infty) \qquad y = (x_1,\ldots,x_n)$
- $\tilde{\lambda}(y) = \frac{1}{n}\sum_{i,j=1}^{n} \frac{K(x_i,x_j)}{x_j} \qquad x_i, x_j \quad \Rightarrow \quad x_i + x_j, x_j$
- $\tilde{N}(t) \rightarrow$ but $\tilde{X}_i(t) \nearrow$ unbounded

$$\left(\tilde{X}_1(t), \ldots, \tilde{X}_n(t)\right) \qquad t \geq 0 \qquad \tilde{N}(t) = \tilde{N}(0) = n$$

- $\mathcal{Y} = \mathcal{X}^n \qquad \mathcal{X} = \{1, 2, \ldots\}$ or $(0, \infty) \qquad y = (x_1, \ldots, x_n)$
- $\tilde{\lambda}(y) = \frac{1}{n} \sum_{i,j=1}^n \frac{K(x_i, x_j)}{x_j} \qquad x_i, x_j \quad \Rightarrow \quad x_i + x_j, x_j$
- $\tilde{N}(t) \rightarrow$ but $\tilde{X}_i(t) \nearrow$ unbounded

Theorem

Assume

$$K(x, y) \geq \bar{K}(x, y)$$

where $\bar{K}(1, 1) > 0$ and

$$\bar{K}(\alpha\, x, \alpha\, y) = \alpha^{\varepsilon}\, \bar{K}(x, y) \qquad \forall\, \alpha > 0$$

for some $\varepsilon > 1$.

Then the mass flow process explodes almost surely, for any initial distribution.

Comments

- analytical aspects

  general tool for proving existence

- numerical aspects

  original motivation for MF

  applications in chemical engineering

- stochastic aspects

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$$t_{\mathrm{gel}} = \lim_{n\to\infty} \tau_\infty^{(n)}$$

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conjecture

$$t_{\text{gel}} = \lim_{n\to\infty} \tau_\infty^{(n)}$$

- challenges
  - include transport terms (boundary conditions) $\Rightarrow$ talk Patterson
  - gelation in the spatial setting
  - rigorous derivation of coagulation kernels