Weierstrass Institute for
Applied Analysis and Stochastics

Leibniz
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Spatial particle processes with coagulation: Gibbs-measure approach, limits, and gelation

Wolfgang König
TU Berlin and WIAS

based on joint works with Luisa Andreis, Heide Langhammer and Robert Patterson

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Random Geometric Systems

Luisa Andreis
(Milano)

Heide Langhammer
(formerly WIAS)

Robert Patterson
(formerly WIAS)

- We consider a spatial particle system with pair-wise coagulation after independent exponential random times.
- We find the large-system limit at a given fixed time T.
- We identify criteria for gelation, i.e., the formation of giant particles.
- Our approach identifies autonomously the distribution of the system by a decomposition of the configuration into the particle groups that have coagulated by time T.
- This necessitates a large-deviation approach and a variational characterisation. The minimizer is the limiting distribution of the system.
- The formula considers only microscopic particles.
- In the simpler situation of a spatial Erdős–Rényi graph, we solved the gelation phase transition ($\Longrightarrow$ LUISA's talk).

- What is the joint distribution of the statistics of all the particles at time T?
- Can we prove a large-deviation principle (LDP) for their statistics as $N \to \infty$ at fixed time T? Is there an explicit rate function?
- Is there a law of large numbers towards the minimizers of the rate function?
- Under what circumstances do we have a gelation phase transition (i.e., emergence of large particles that carry macroscopically much total mass) after some gelation time $t_c \in (0, \infty)$?

Remarks:

- We are in the hydrodynamic regime or in a mean-field setting, where all the N particles interact with each other on the same scale, $\frac{1}{N}$. Most particles feel $\asymp N$ other particles and have $\asymp 1$ coagulations per unit time.
- We decided to work with a Poissonnized initial configuration, $\mathbb{P}_{\mathrm{Poi}_{N\mu}} = \int \mathrm{Poi}_{N\mu}(\mathrm{d}k)\, \mathbb{P}_k$, for some probability measure μ on $\mathcal{S}$. Then the number of atoms is Poi_N-distributed. This renders limiting formulas much simpler.

- We took the freedom to extend the MARCUS-LUSHNIKOV model [MARCUS 1968], [GILLESPIE 1972], [LUSHNIKOV 1978] to a spatial mean-field version. It is very closely related to the cluster coagulation model investigated by [NORRIS 1999, 2000].
- FOURNIER/LAURENÇOT (2005-09) derive these equations for a strongly gelling kernel $K(m,\widetilde{m}) = m^\alpha \widetilde{m} + \widetilde{m}^\alpha m$ with $\alpha \in (0,1]$.
- [JEON 1998] and [REZANKHANLOU 2013] give gelation criteria on the kernel: $K(m,\widetilde{m}) = (m\widetilde{m})^a$ with $a > \frac{1}{2}$ and $K(m,\widetilde{m}) = m^q + \widetilde{m}^q$ with $q \in (1,2)$ are gelling.

We are in the situation of a system that consists of many microscopic random entities (atoms) with pair-interactions that are put independently. They imply the formation of a substructure, a decomposition of the system into microscopic groups.

Examples:

1. percolation
2. interacting Bose gas
3. Erdős–Rényi graph
4. random connection model
5. Marcus–Lushnikov model
6. collision models (gas dynamics)

In all these models, there should be a representation of the system in terms of a Poisson point process (PPP) possible that is defined directly on the set of the emerging substructures: the non-interaction between them is incorporated as a pair-interaction between the Poisson points. This has been used for (2) and (5), implicitly for (3), and is on the way for (4), but has a great potential for future exploration.

From now on, fix $T \in (0,\infty)$. Let Γ_T be the set of trajectories $[0,T] \to \mathcal{M}_{\mathbb{N}_0}(\mathcal{S}\times\mathbb{N})$ and

$$\Gamma_T^{(1)} = \{\xi \in \Gamma_T \colon \xi_T(\mathcal{S}\times\mathbb{N}) = 1\}$$

the set of trees on the time interval $[0,T]$, i.e., of trajectories that coagulate into one particle. Decompose $\Xi|_{[0,T]}$ into the subtrees $\Xi^{(C)}$, and consider the empirical measure of the trees,

$$\mathcal{V}_N^{(T)} = \frac{1}{N}\sum_C \delta_{\Xi^{(C)}} \in \mathcal{M}(\Gamma_T^{(1)}).$$

We need the non-coagulation probability as an interaction between trees $\xi,\xi' \in \Gamma_T^{(1)}$:

$$R^{(T)}(\xi,\xi') = -\log \mathbb{P}_{\xi_0\cup\xi_0'}\big(\Xi_1 \not\leftrightarrow \Xi_2 \,\big|\, \Xi_1 = \xi, \Xi_2 = \xi'\big) = \int_0^T \mathrm{d}s\,\langle \xi_s, K\xi_s'\rangle.$$

For our PPP, we take the intensity measure

$$M_{\mu,N}^{(T)}(\mathrm{d}\xi) = N^{|\xi_0|-1}\mathrm{ePoi}_\mu(\Xi\in\mathrm{d}\xi), \qquad \xi \in \Gamma_T^{(1)}.$$

Tree decomposition (with kernel K)

$$\mathbb{P}_{\mathrm{Poi}_{N\mu}}\big(\mathcal{V}_N^{(T)} \in \mathrm{d}\nu\big) = \mathtt{E}\Big[\mathrm{e}^{-\frac{1}{2}\sum_{i\neq j} R^{(T)}(\Xi_i,\Xi_j)}\mathbb{1}\{\tfrac{1}{N}Y \in \mathrm{d}\nu\}\Big]\,\mathrm{e}^{N(|M_{\mu,N}^{(T)}|-1)},$$

where $Y = \sum_i \delta_{\Xi_i} \sim \mathrm{Poi}_{NM_{\mu,N}^{(T)}}$ is a Poisson point process on $\Gamma_T^{(1)}$.

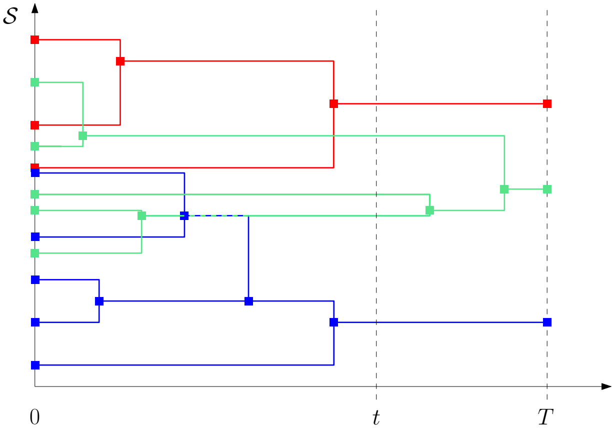

Figure: Decomposition of $(\Xi_t)_{t\in[0,T]}$ into three subprocesses $(\Xi_t^{(T,C_i)})_{t\in[0,T]}$, $i = 1, \dots, 3$, that are distinguished by their colour.

- Our PPP presentation with exponential necessitates a large-deviations principle for $\mathcal{V}_N^{(T)}$.
- From the LDP theory (not so well-known): If Y is a PPP with intensity measure $N\nu_N$ and ν_N converges towards ν, then $\frac{1}{N}Y$ satisfies an LDP with rate function $H(\cdot \mid \nu)$ (the Kulback–Leibler entropy with respect to ν).

Convergence of $M_{\mu,N}^{(T,N)}$ (with kernel $\frac{1}{N}K$)

The following limit exists in the weak sense:

$$M_\mu^{(T)}(\mathrm{d}\xi) = \lim_{N\to\infty} M_{\mu,N}^{(T,N)}(\mathrm{d}\xi)$$
$$= \exp\Big\{\frac{1}{2}\int_0^T \Big[\langle \xi_t, K\xi_t\rangle + \langle \xi_t, K^{(\mathrm{diag})}\rangle\Big]\,\mathrm{d}t\Big\}\,\mathbb{P}_{\mathrm{Poi}_\mu}(\mathrm{d}\xi), \qquad \xi \in \Gamma_T^{(1)}.$$

where $K^{(\mathrm{diag})}(y) = K(y,y)$.

The following assumption implies that gelation takes place not too early:

Upper bound on the kernel

There is an $H > 0$ such that $\quad K((x,m),(\widetilde{x},\widetilde{m})) \leq Hm\widetilde{m} \qquad$ for $x,\widetilde{x} \in \mathcal{S}, m,\widetilde{m} \in \mathbb{N}$.

Here is a formula for the exponential asymptotics under explicit preclusion of gelation. Gelation does not occur if $\mathcal{V}_N^{(T)}$ lies, for some $A > 0$, in

$$\mathcal{A}_{f,A} = \Big\{\nu \in \mathcal{M}(\Gamma_T^{(1)}) \colon \int_{\mathcal{M}_{\mathbb{N}_0}(\mathcal{S})} \nu_0(\mathrm{d}k)\, f(|k|) \leq A\Big\}, \qquad \lim_{r\to\infty} \frac{f(r)}{r\log r} = \infty.$$

The LDP (with kernel $\frac{1}{N}K$)

Pick $T \in (0,\infty)$ and $\mu \in \mathcal{M}_1(\mathcal{S})$. Pick the initial configuration $(\{x_1\},\dots,\{x_N\})$ with $\mu_{\mathbf{x}} = \frac{1}{N}\sum_{i=1}^N \delta_{x_i} \Longrightarrow \mu$.
Then, for any $A > 0$, the distribution of $\mathcal{V}_N^{(T)}$ under $\mathbb{P}^{(N)}_{\mathrm{Poi}_{N\mu}}(\,\cdot\,|\mathcal{V}_N^{(T)} \in \mathcal{A}_{f,A})$ satisfies the LDP on $\mathcal{A}_{f,A}$ with rate function $I_\mu^{(T)} - \chi_\beta$, where

$$I_\mu^{(T)}(\nu) = \Big\langle \nu, \log\frac{\nu}{M_\mu^{(T)}}\Big\rangle + \frac{1}{2}\langle \nu\otimes\nu, R^{(T)}\rangle + 1 - |\nu|,$$

and $\chi_\beta = \inf_{\mathcal{A}_{f,A}} I_\mu^{(T)}$.

- The assumption on K says that gelation occurs in our spatial model after a giant component emerges in a coupled Erdős–Rényi graph.
- Conditioning on $\mathcal{A}_{f,A}$ gives a full LDP without need of thinking about macroscopic particles. ($\Longrightarrow$ future work.)
- Likewise, we could also condition on having all particle sizes $\leq L$ for some $L \in \mathbb{N}$.
- The Euler–Lagrange equations for a possible minimizer $\nu^{(*)}$ of $I_\mu^{(T)}$ read

$$\nu^{(*)}(\mathrm{d}\xi) = M_\mu^{(T)}(\mathrm{d}\xi)\,\mathrm{e}^{-\mathfrak{R}^{(T)}(\nu^{(*)})(\xi)}, \qquad \xi \in \Gamma_T^{(1)},$$

  ($\mathfrak{R}^{(T)}$ is the convolution operator with kernel $R^{(T)}$.)
- $I_\mu^{(T)}$ is not necessarily lower semi-continuous, and it is not necessarily convex. The last thing can be settled by assuming that K is nonnegative definite. The first thing is fishy and has much to do with the gelation phase transition.
- Gelation should occur precisely if and only if $I_\mu^{(T)}$ does have a minimizer.

The coagulation process is a function of $\mathcal{V}_N^{(T)}$, since

$$\frac{1}{N}\Xi_t = \frac{1}{N}\sum_{\widetilde{C}\in P_t} \delta_{(X_{\widetilde{C}}^{(t)},|\widetilde{C}|)} = \frac{1}{N}\sum_{C\in P_T}\sum_{\widetilde{C}\in P_t:\,\widetilde{C}\subset C} \delta_{(X_{\widetilde{C}}^{(t)},|\widetilde{C}|)}$$
$$= \frac{1}{N}\sum_{C\in P_T} \Xi_t^{(T,C)} = \int \mathcal{V}_N^{(T)}(\mathrm{d}\xi)\,\xi_t =: \rho_t(\mathcal{V}_N^{(T)}).$$

$\rho = (\rho_t)_{t\in[0,T]}$ is continuous

Let $(\nu_n)_{n\in\mathbb{N}}$ be a sequence in $\mathcal{A}_{f,A}$ that converges towards some ν with $I_\mu^{(T)}(\nu) < \infty$. Then $\rho(\nu_n) \to \rho(\nu)$ as $n \to \infty$.

The contraction principle yields then:

LDP for Ξ

In the situation of the LDP above, the distribution of $(\frac{1}{N}\Xi_t)_{t\in[0,T]}$ satisfies an LDP on $\mathbb{D}_T(\mathcal{M}(\mathcal{S}\times\mathbb{N}))$ with rate function

$$\rho \mapsto \inf\left\{I_\mu^{(T)}(\nu) - \chi_\beta \colon \nu \in \mathcal{A}_{f,A}, \rho(\nu) = \rho\right\}.$$

Hence, for sufficiently small T (under the upper bound on the kernel K), the process $(\frac{1}{N}\Xi_t)_{t\in[0,T]}$ converges towards the solution $\rho(\nu) = (\int \nu(\mathrm{d}\xi)\, \xi_t)_{t\in[0,T]}$, where ν solves the EL equation

$$\nu(\mathrm{d}\xi) = M_\mu^{(T)}(\mathrm{d}\xi)\, \mathrm{e}^{-\mathfrak{R}^{(T)}(\nu)(\xi)}, \qquad \xi \in \Gamma_T^{(1)}.$$

On the other hand, the limiting dynamics of the process $\frac{1}{N}(\Xi_t)_{t\in[0,\infty)}$ should satisfy the SMOLUCHOWSKI equation, which reads here

$$\partial_t \rho_t(\mathrm{d}x^*, m^*) = \frac{1}{2} \sum_{\substack{m,m'\in\mathbb{N}:\\ m+m'=m^*}} \int_{\mathcal{S}} \int_{\mathcal{S}} \rho_t(\mathrm{d}x, m)\rho_t(\mathrm{d}x', m')\mathbf{K}\big((x,m),(x',m'),\mathrm{d}x^*\big)$$
$$- \rho_t(\mathrm{d}x^*, m^*)\, K\rho_t(x^*, m^*), \qquad x^* \in \mathcal{S}, m^* \in \mathbb{N},$$

The limit satisfies the Smoluchowski equation

Under the upper bound on K, for any $T < \frac{1}{H}\frac{1}{\mathrm{e}^2}\frac{\pi}{1+\pi}$, the solution ν to the EL equation is unique, and $\rho(\nu)$ solves the Smoluchowski equation.

Hence, this is in our approach a nice addition, but not part of the proof for the limiting behaviour.

The quantity

$$\mathrm{NG}_T^{(\mu)} = \lim_{L\to\infty} \limsup_{N\to\infty} \mathbb{E}^{(N)}_{\mathrm{Poi}_{N\mu}} \Big[\| \tfrac{1}{N} \Xi_T \|_{1,\leq L} \Big]$$

is the limiting expected non-gel mass, i.e., the mass outside the gel. The map $T \mapsto \mathrm{NG}_T^{(\mu)}$ is non-increasing with initial value $\mathrm{NG}_0^{(\mu)} = 1$. If $\mathrm{NG}_T^{(\mu)} < 1$, then we say that there is a gel at time T, and we define the gelation time by

$$T_{\mathrm{gel}}^{(\mu)} = \inf \Big\{ T \in (0,\infty) \colon \mathrm{NG}_T^{(\mu)} < 1 \Big\} \in [0,\infty].$$

(Among all possible definitions, this is the “earliest” gelation time one can think of.)

Introduce

$$q_\mu^{(T)} = \limsup_{n\to\infty} \Big(M_\mu^{(T)}\big(\{\xi \in \Gamma_T^{(1)} : |\xi_0| = n\}\big)\Big)^{1/n} \in (0,\infty).$$

$I_\mu^{(T)}$-dependent criteria for non-gelation and for gelation

1. If $q_\mu^{(T)} < 1$, then $I_\mu^{(T)}$ has compact sublevel sets and hence possesses minimisers, and $\mathrm{NG}_T^{(\mu)} = 1$, i.e., there is no gelation at time T.
2. Assume additionally that

   there is a $h > 0$ such that $\quad K((x,m),(\widetilde{x},\widetilde{m})) \geq hm\widetilde{m} \quad$ for $x,\widetilde{x} \in \mathcal{S}, m,\widetilde{m} \in \mathbb{N}$,

   and that $\inf I_\mu^{(T)} > 0$. Then $\mathrm{NG}_T^{(\mu)} < 1$.

T-dependent criteria for non-gelation and for gelation

Assume the two bounds for K. If T is large enough (depending only on H), then $q_\mu^{(T)} < 1$, and the EL equations have precisely one solution. Furthermore,

$$T_{\mathrm{gel}}^{(\mu)} \leq \inf\Big\{T\colon \frac{1}{2T}\Big(\frac{\mathrm{e}}{\pi H} + \frac{(\log(2TH\mathrm{e}^2))^2}{h}\Big) < 1\Big\} < \infty.$$

- We wrote the distribution autonomously (i.e., not by deriving an equation that it satisfies, e.g., the Smoluchowski equation) as a PPP-expectation. To analyse it, we had to use an LDP-framework. To carry this out without bigger problems, we needed to restrict to bounded sizes. The limiting process is characterized as the minimizers in a crucial variational formula, which does not describe macroscopic particles, but admits criteria for their existence.
- We are not happy with our gelation criteria, as they do not reflect any spatial property of the kernel, and rely too strongly on the product form.
- Description of the gel still widely open.
- An adequate framework that also characterises large particles would require an extension of the state space of the PPP; a "macroscopic" space must be added. Such an analysis has been yet rarely done in comparable models, e.g., in [ANDREIS, K., PATTERSON 2021] (non-spatial ER-graph), [ANDREIS, K., LANGHAMMER, PATTERSON 2023] (spatial ER-graph), COLLIN, JAHNEL, K., VOGEL, ZASS,... (variants of the Bose gas).