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A large-deviation approach to coagulation

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based on joint work in progress with Luisa Andreis, Heide Langhammer and Robert Patterson

SPP2265 Random Geometric Systems

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- We consider a spatial particle system with pair-wise coagulation after independent exponential random times.
- We are interested in the large-system limit at a given fixed time T.
- Prospectively, we want to identify criteria for gelation, i.e., the formation of giant particles.
- We decompose the configuration into the particle groups that have coagulated by time T.
- This necessitates a large-deviation approach and a variational characterisation.
- In the simpler situation of a spatial Erdős–Rényi graph, we completely solved the gelation phase transition in recent work.

A Markovian particle model with coagulation on $\mathcal{S} \times \mathbb{N}$ (with $\mathcal{S}$ a compact convex metric space):

- Configuration at time t:

$$\big((X_1(t), M_1(t)), \ldots, (X_{n(t)}(t), M_{n(t)}(t))\big)$$

  with $$M_1(t) \geq M_2(t) \geq \cdots \geq M_{n(t)}(t) \geq 1 \quad \text{and} \quad \sum_{i=1}^{n(t)} M_i(t) = N.$$

- monodispersed initial configuration $M_1(0) = \cdots = M_N(0) = 1$.
- Dynamics: Particles (x, m) and (y, n) are replaced by $(\frac{xm+yn}{m+n}, m+n)$ at rate $\frac{1}{N} K((x,m),(y,n))$ fixing the center of mass.
- All (non-)coagulations occur independently.
- Hence, $(n(t))_{t \in [0,\infty)}$ is a decreasing stochastic process in $\mathbb{N}$.
- $\mathbf{x} = (X_1(0), \ldots, X_N(0))$ fixed, such that $\mu_{\mathbf{x}} = \frac{1}{N} \sum_{i=1}^{N} \delta_{x_i} \Longrightarrow \mu$.

- Joint distribution of the statistics of all the particles (according to the initial configuration that coagulated into them) at time T?
- Large-deviation principle (LDP) for their statistics as $N \to \infty$ at fixed time T? Explicit rate function?
- Law of large numbers at fixed time T towards the minimizers of the rate function?
- Gelation phase transition, i.e., appearance of giant particle $M_1(t) \asymp N$ after some gelation time $t_c \in (0, \infty)$?

Remarks:

- We are in the hydrodynamic regime, where N particles are in a compact space $\mathcal{S}$, not depending on N. Most particles feel $\asymp N$ other particles and have $\asymp 1$ coagulations per time interval
- The system simplifies, since we are only interested in statsitics of the particles present at time T, and hence only into those initial sub-configurations that coagulate into them. However, we would like to keep control on the structure of these initial sub-configurations.

Simplification of the model:

coagulation $\Longrightarrow$ putting an edge.

That is, random growing inhomogeneous graph with vertices in $\mathcal{S}$ instead of particle process with coagulation. These models coincide in one special case:

Fact

For the product kernel : $$K_N(m,\widetilde{m}) = \frac{m\,\widetilde{m}}{N},$$

the model is a time-dependent version of the well-known ERDŐS-RÉNYI random graph model.

Indeed, the vector $(M_i^{(N)}(t))_{i=1}^{n(t)}$ is in distribution equal to the collection of sizes of all the connected components of the graph $\mathcal{G}(N, 1-\mathrm{e}^{-t/N})$.

Explanation:

Equip each $\{i,j\}$ independently with an exponentially distributed random time $\mathrm{e}_{i,j}$ with expected value N. After the elapsure of $\mathrm{e}_{i,j}$, there is a bond created between i and j. At time t, the probability for a bond between i and j is equal to $1-\mathrm{e}^{-t/N}$.

The rate of connecting two components of size m and $\widetilde{m}$ is equal to $\frac{1}{N}m\widetilde{m}$, since $m\widetilde{m}$ is the number of active bonds that can connect these components.

- The Marcus-Lushnikov model is a non-spatial mean-field version [Marcus 1968], [Gillespie 1972], [Lushnikov 1978]
- [Smoluchowski 1916] introduces an ODE system for the evolution of particle sizes:

$$\frac{\mathrm{d}}{\mathrm{d}t}\lambda_k(t) = \frac{1}{2}\sum_{m,\widetilde{m}\in\mathbb{N}\colon m+\widetilde{m}=k}\lambda_m(t)\lambda_{\widetilde{m}}(t)K(m,\widetilde{m}) - \lambda_k(t)\sum_{m\in\mathbb{N}}\lambda_m(t)K(k,m),$$

where $\lambda_m(t) = \lim_{N\to\infty}\frac{1}{N}\#\{\text{particles at time } t \text{ of size } k\}$.

- Convergence of stochastic coagulation processes towards these ODEs was expected for long time, but the first rigorous proof was given only in [Lang, Nguyen 1980].
- A variant, also including the gel, is called Flory's equation.
- 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.
- In progress (Andreis, Iyer, Magnanini): comparison of spatial coagulation particle models to non-spatial ones, using generators, coupling and limiting equations.

Recall the coagulation process

$$Z=(Z_t)_{t\in[0,\infty)}, \qquad \text{with } Z_t=(X_i(t),M_i(t))_{i=1,\dots,n(t)},$$

with mechanism

$$\big((X,m),(Y,\widetilde{m})\big)\mapsto\Big(\frac{Xm+Y\widetilde{m}}{m+\widetilde{m}},m+\widetilde{m}\Big) \qquad \text{with rate } \frac{1}{N}K\big((X,m),(Y,\widetilde{m})\big).$$

Empirical process $\Xi_t(A,m)=\#\{\text{particles in } A \text{ with size } m\}$,

$$\Xi=(\Xi(t))_{t\in[0,\infty)}, \qquad \text{with } \Xi_t=\sum_{i=1}^{n(t)}\delta_{(X_i(t),M_i(t))}\in\mathcal{M}_{\mathbb{N}_0}(\mathcal{S}\times\mathbb{N}),$$

with mechanism

$$\phi\mapsto\phi-\delta_{(x,m)}-\delta_{(x',m')}+\delta_{(\frac{xm+x'm'}{m+m'},m+m')}$$

with rate

$$M_\phi\big((x,m),(x',m')\big)=\frac{1}{N}K(\dots)\times\begin{cases}\phi(\{x\},m)\phi(\{x'\},m'), & \text{if } (x,m)\neq(x',m'),\\ \phi(\{x\},m)(\phi(\{x\},m)-1) & \text{otherwise.}\end{cases}$$

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

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

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,

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

Non-coagulation probability as an interaction between trees:

$$R^{(T)}(\xi,\xi') = -\log \mathbb{P}_{\xi_0\cup\xi_0'}\big(\Xi_1 \not\leftrightarrow \Xi_2 \,\big|\, \Xi_1 = \xi, \Xi_2 = \xi'\big), \qquad \xi,\xi' \in \Gamma_T^{(1)},$$

Tree decomposition

$$\mathbb{P}_{\mathbf{x}}\big(\mathcal{V}_N^{(T)} \in \mathrm{d}\nu\big) = \mathbf{E}\Big[\mathrm{e}^{-\frac{1}{2}\sum_{i\neq j} R^{(T)}(\Xi_i,\Xi_j)} \mathbb{1}\{\tfrac{1}{N}Y \in \mathrm{d}\nu\} \,\Big|\, \tfrac{1}{N}Y_0 \in \mathcal{C}_{\mu_{\mathbf{x}}}\Big] \times \mathrm{e}^{N|\nu_0|}\prod_k \big(\tau_k^{(T,N)}\big)^{N\nu_0(k)},$$

where $Y = \sum_i \delta_{\Xi_i} \sim \mathrm{Poi}_{N\mathrm{Poi}_{\mu_{\mathbf{x}}}\otimes\overline{\mathbb{Q}}^{(T,N)}}$ is a Poisson point process on $\Gamma_T^{(1)}$.

The reference process Y admits a nice formula and is a good starting point for asymptotics.

Introduce the rescaled tree-restriction of the process measure and its total mass (the coagulation probability), when started in the configuration $k \in \mathcal{M}_{\mathbb{N}_0}(\mathcal{S})$:

$$\mathbb{Q}_k^{(T,N)} = N^{|k|-1}\mathbb{P}_k|_{\Gamma_T^{(1)}} \qquad \text{and} \qquad \tau_k^{(T,N)} = \mathbb{Q}_k^{(T,N)}(\Gamma_T^{(1)}).$$

Convergence of $\mathbb{Q}_{\mathbf{k}}^{(\mathbf{T},\mathbf{N})}$ and $\tau_{\mathbf{k}}^{(\mathbf{T},\mathbf{N})}$

$$\mathbb{Q}_k^{(T)} = \lim_{N\to\infty} \mathbb{Q}_k^{(T,N)} \quad \text{and} \quad \tau_k^{(T)} = \lim_{N\to\infty} \tau_k^{(T,N)} = \mathbb{Q}_k^{(T)}(\Gamma_T^{(1)}) \in (0,\infty).$$

(We have explicit formulas in terms of the kernel M.)

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

Assumption on the kernel

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

We have $\tau_k^{(T)} \approx |k|\log(TH|k|)$ as $|k| \to \infty$ under this assumption.

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

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

The LDP

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 $\mathbb{P}_{N\mu_{\mathbf{x}}}(\,\cdot\,|\mathcal{V}_N^{(T)} \in \mathcal{A}_{f,A})$ satisfies the LDP on $\mathcal{A}_{f,A}$ with rate function

$$I_\mu(\nu) = H(\nu|\mathrm{Poi}_\mu \otimes \overline{\mathbb{Q}}^{(T)}) - \langle \nu, \log \tau^{(T)}\rangle + \frac{1}{2}\langle \nu\otimes\nu, R^{(T)}\rangle - |\nu_0|,$$

- We have explicit formulas for $\mathbb{Q}^{(T)}$ and $R^{(T)}$.
- The assumption on K says that gelation occurs in our spatial model after a giant component emerges in some Erdős–Rényi graph.
- The characterisation of the distribution of $\mathcal{V}_N^{(T)}$ in terms of a tree decomposition necessitates the use of large deviations, since the non-coagulation probability terms are exponential in N.
- Conditioning on $\mathcal{A}_{f,A}$ gives a full LDP without need of thinking about macroscopic particles. ($\Longrightarrow$ future work.) Interesting is only $A \to \infty$.
- The Euler–Lagrange equations for a possible minimizer $\nu^{(*)}$ of I_μ read

$$\nu^{(*)}(\mathrm{d}\xi) = (\mathrm{Poi}_\mu \otimes \mathbb{Q}^{(T)})(\mathrm{d}\xi)\,\mathrm{e}^{-\mathfrak{R}^{(T)}(\nu^{(*)})(\xi)+1}\,\mathrm{e}^{\int_{\mathcal{S}} a(x)\,\xi_0(\mathrm{d}x)}, \qquad \xi \in \Gamma_T^{(1)},$$

  with some Euler–Lagrange function $a\colon \mathcal{S} \to \mathbb{R}$. ($\mathfrak{R}^{(T)}$ is the convolution operator with kernel $R^{(T)}$.) This needs to be further analysed in future. $\nu^{(*)}$ should satisfy the Smoluchovski equations.
- Gelation should occur precisely if and only if I_μ does have a minimizer. Understanding this criterion deeper is subject to future work.