Gelation in cluster coagulation

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Motivation: Coagulation Processes

- Model processes where small particles 'coagulate' or 'come together'.

  'giant' particle

- For example, the formation of stars, the formation of polymers, the formation of monopolies.
- How does the system of particle concentrations evolve? (Concentration of trajectories, WLLN)
- Does one see 'large' particles?

  (Does 'gelation' occur?)

  $\sim$ condensation
  loss of energy

Cluster Coagulation Model

(Norris, 99')

- Consider configurations of 'clusters' in a (metric) measure space $(E, \mathcal{B})$ ← sigma algebra.
- Ingredients: a kernel $K: E \times E \times \mathcal{B} \longrightarrow [0, \infty]$

  a mass function $m: E \longrightarrow [0, \infty)$

- For each $x, y \in E$ $K(x,y,A)$ is rate at which x & y merge to form $z \in A$.

  $K(x, y, dz)$ is a measure describing the rate x & y merge to form z.

  $m(x)$ represents the 'mass' of each cluster.

Axioms of the Model

1. **Symmetric**: for all $x, y \in E$

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

"We only care about the pair of particles involved in a coagulation"

2. **finite**: for all $x, y \in E$

$$\bar{K}(x, y) = K(x, y, E) < \infty$$

"No instantaneous coagulations"

3. **Preserves mass**: for all $x, y \in E$

$m(z) = m(x) + m(y)$ for $K(x, y, dz)$-a.a z.

"coagulations preserves mass"

Dynamics

Suppose we begin with a (finite, labelled) configuration of clusters in E.

- To each pair of clusters x, y, we associate an exponential random variable with parameter $\bar{K}(x,y) = K(x,y,E)$
- When the next 'exponential clock rings', corresponding to x & y, say, remove x & y, and replace them with a cluster z sampled from $\dfrac{K(x,y,dz)}{\bar{K}(x,y)}$ $\Leftarrow$ Prob measure on E.

Example 1: Marcus – Lushnikov Process

(Marcus, '68), (Gillespie, '72), (Lushnikov, '78)

$E = [0,\infty)$, $m(x) = x$

& $K(x,y,dz) = \bar{K}(x,y)\,\delta_{x+y}$

for some symmetric $\bar{K} : [0,\infty)\times[0,\infty) \to [0,$

In this case, we only consider particle masses, and two particles merge at a rate corresponding to their masses.

Eg.

mass 1 particle & mass 3 $\overset{\bar{K}(1,3)}{\longmapsto}$ mass 4.

Examples of Marcus-Lushnikov Process

- The case $\bar{K}(x,y) \equiv 1$ corresponds to Kingman's coalescent (Kingman, '82)

- The case $\bar{K}(x,y) = x+y$ corresponds to the 'standard additive coalescent' (eg. Aldous, Pitman, '98) (Review from (Bertoin, 2003))

More examples of Marcus-Lushikov Process

- The case $\bar{K}(x,y) = xy$ closely connected to the sparse Erdős-Rényi random graph.

- The case $\bar{K}(x,y) = (x^{1/3} + y^{1/3})(x^{-1/3} + y^{-1/3})$ "mean-field" for coagulating Brownian particles, (Smoluchowski, 1916) where particles of radius r (proportional to the cube root of the mass) perform independent Brownian motions with variance $1/r$.

Cluster Coagulation Model

(Norris, 99')

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Example 2: Bilinear coagulation processes

(Patterson, Heydecker, '19)

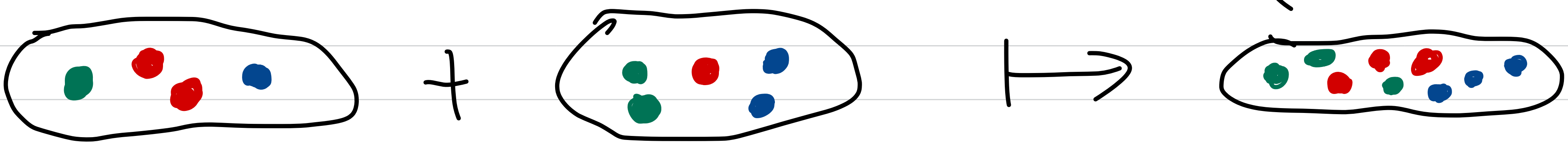
"d-dimensional Marcus-Lushnikov process"

$E = [0,\infty)^d$, $\quad K(x,y,dz) = (x^T A y)\,\delta_{x+y}$

for some symmetric matrix $A \in [0,\infty)^{d\times d}$

and $\quad m(x) = \mathbb{1}^T x \quad$ (sum of entries of vector)

Eg. $\begin{pmatrix}1\\2\\1\end{pmatrix}$ and $\begin{pmatrix}2\\1\\2\end{pmatrix}$ merge to form $\begin{pmatrix}3\\3\\3\end{pmatrix}$



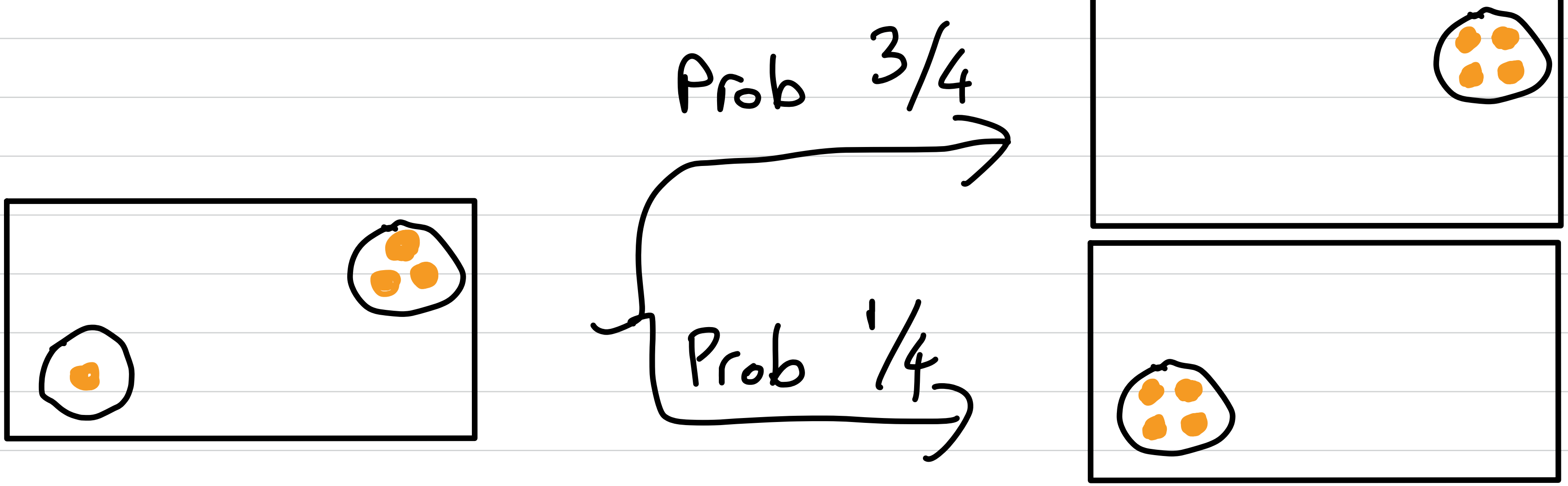
Example 3: Particles in Space

$E = \underset{\text{location}}{S} \times \underset{\text{mass}}{[0,\infty)}$ for some metric space S

$K((x,n),(y,o),dz) = \bar{K}((x,n),(y,o))\left(\frac{n}{n+o}\delta_{(x,n+o)} + \frac{o}{n+o}\delta_{(y,n+o)}\right)$

for some symmetric $\bar{K}: E \times E \to [0,\infty)$

$m(x,n) = n$



Motivation: Limiting Trajectories

- We encode configurations of particles as measures $(L_t^{(N)})_{t \geq 0}$, representing concentrations of particles over time.

$$\underbrace{\sum \delta_{x_i}}_{\text{measure encoding configuration}} \overset{\text{coagulation involving } x \,\&\, y}{\longmapsto} \sum \delta_{x_i} \underbrace{- \delta_x - \delta_y}_{\text{Remove } x \,\&\, y} \underbrace{+ \delta_z}_{\text{New particle.}}$$

- Assume that for some limiting probability measure μ_0, we have $\frac{1}{N} L_0^{(N)} \xrightarrow[\text{Prob}]{N\to\infty} \mu_0$ (weakly, in prob.)

and $\frac{1}{N}\int m(x)\, L_0^{(N)}(dx) \xrightarrow[\text{Prob}]{N\to\infty} \int m(x)\, \mu_0(dx)$

initial mass converges

with $0 < \int m(x)\, \mu_0(dx) < \infty$

- Generally assume that for each N

$$|\bar{L}_0^{(N)}| < \infty$$

Is there concentration or convergence of

$$\underbrace{\left(\frac{1}{N}\, L^{(N)}_{\underbrace{t/N}_{\text{Re-scale time}}}\right)_{t\in[0,\infty)}}$$

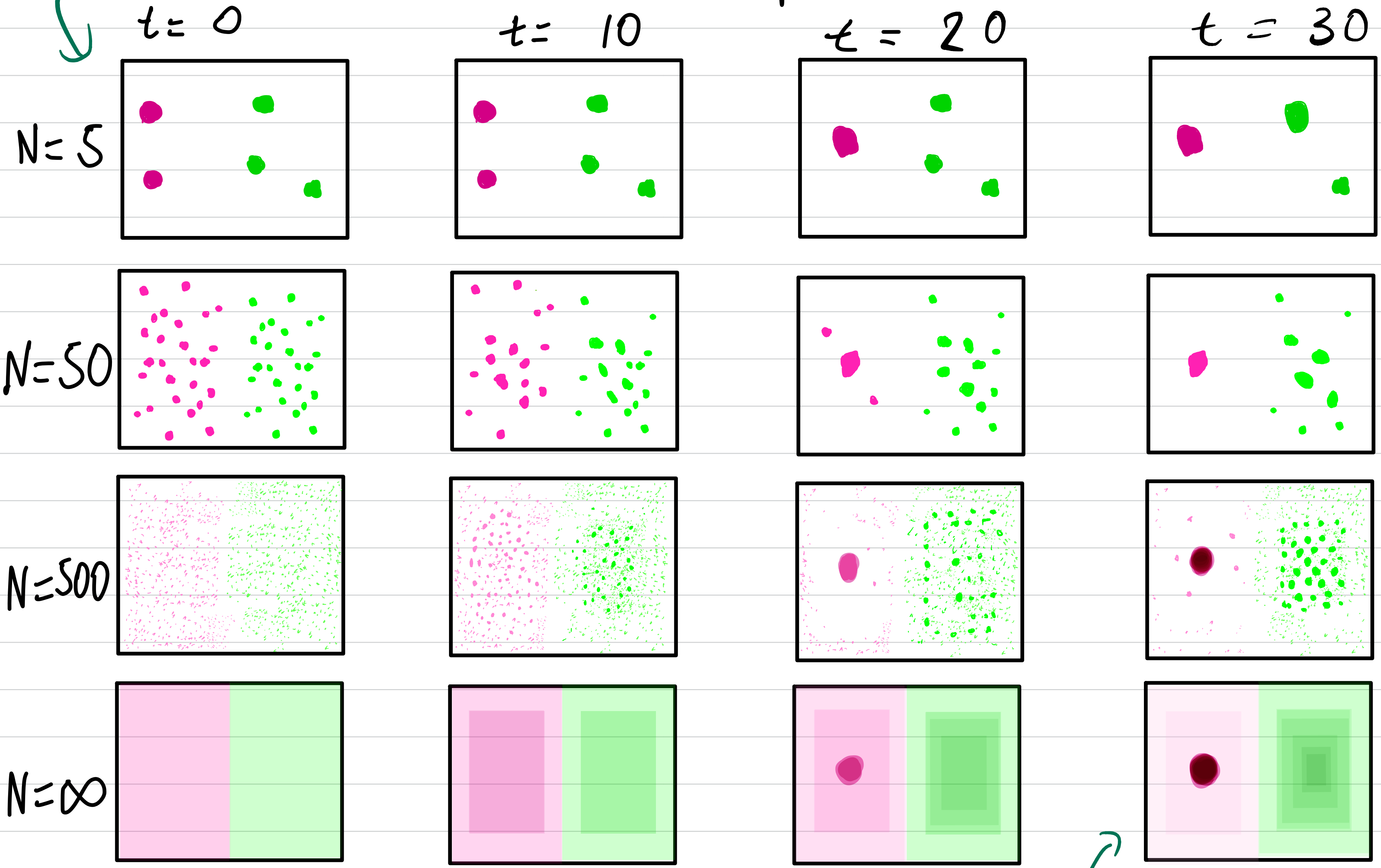
to $(M_t)_{t\in[0,\infty)}$ for some "trajectory" of measures?

Re-scale time

Re-scale total size of the system.

Intuition for measure trajectories: Limits of pixelated animations

measure on E



measure changes over time

mass leaving to ∞

<u>Short-hand:</u> $\bar{L}_t^{(N)} := \frac{1}{N} L_{t/N}^{(N)}$

- Think of each $(\bar{L}_t^{(N)})_{t\geq 0}$ as a random trajectory with Probability measure $\mathbb{P}_N(\cdot)$

- Two notions of convergence: (measure over trajectories of measures)

1) $(\bar{L}_t^{(N)})_{t\geq 0} \xrightarrow{\text{Prob}} (\mu_t)_{t\geq 0}$ for some $(\mu_t)_{t\geq 0}$

2) $\{\mathbb{P}_N(\cdot) : N\in\mathbb{N}\}$ is relatively compact.
converges on a sub-sequence

$\{\mathbb{P}_N(\cdot) : N \in \mathbb{N}\}$ is relatively compact.

Accumulation points $\mathbb{P}^*(\cdot)$

concentrated on solutions of

"measure valued differential equations"

known as "Smoluchowski" or "Flory" equations.

(analogues of Boltzmann)

(Smoluchowski, 1916),

(Flory, 1942)

Some Well-Known Results

When $E = [0,\infty)$, $\bar{K}(x,y) \le x+y$

$$\left(\bar{L}^{(N)}_t\right)_{t\ge 0} \xrightarrow{\text{Prob.}} (\mu_t)_{t\ge 0}$$ satisfying "Smoluchowski" equation.

(Norris, '98)

For arbitrary E, when $\bar{K}(x,y)$ "eventually multiplicative"

$$\left(\bar{L}^{(N)}_t\right)_{t\ge 0} \xrightarrow{\text{Prob}} (\mu_t)_{t\ge 0}$$ satisfying "Flory" equation

(Norris, '99)

$E = [0, \infty)$

When
$$\lim_{x \to \infty} \frac{\bar{F}(x,y)}{x} = \ell(y)$$
(and $\{\mathbb{P}_N(\cdot) : N \in \mathbb{N}\}$ is relatively compact)

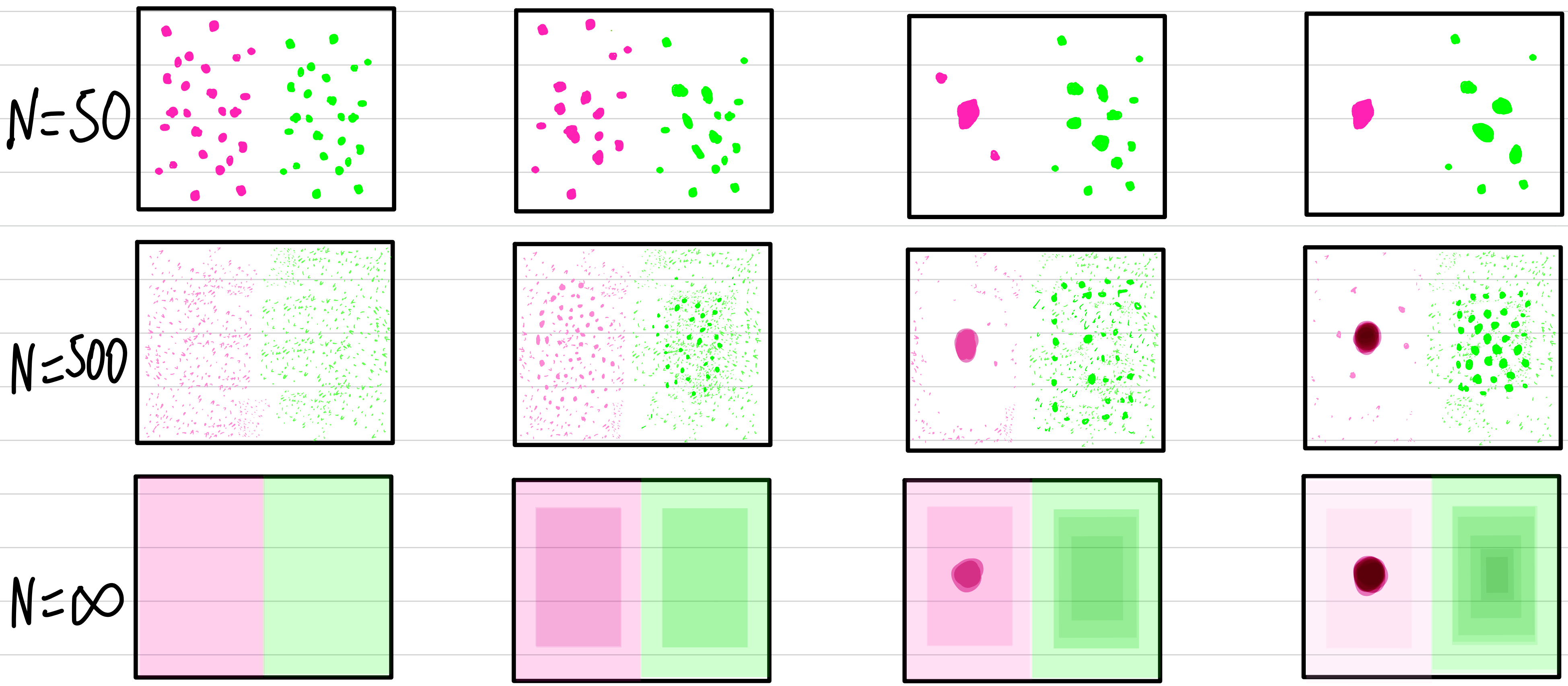
accumulation points of

$\{\mathbb{P}_N(\cdot) : N \in \mathbb{N}\}$ satisfy

"Flory" equation

(Fournier & Giet, 2003)

Motivation: Limiting Trajectories



Will be helpful to find 'invariants' associated with the process (like total mass).

Conserved Quantities

Definition: A function $\phi : E \times E \to [0,\infty)$ is said to be *conservative* if

$\forall x, y, q \in E$ $\quad \phi(z,q) = \phi(x,q) + \phi(y,q)$

for $K(x,y,dz)$-a.a. z.

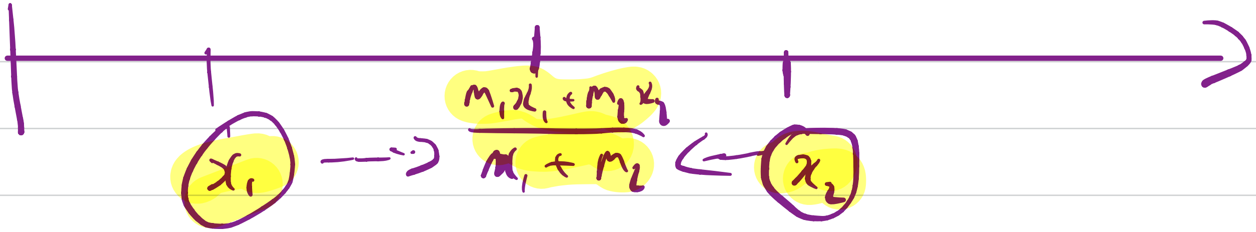
Example: The function $\phi(u,v) = m(u)\,\ell(v)$ is *conservative* since

$\forall x, y, q \in E$ $\quad m(z)\,\ell(q) = \big(m(x) + m(y)\big)\,\ell(q)$.

for $K(x,y,dz)$-a.a z.

<u>Example:</u> $E = [0,\infty) \times [0,\infty)$

consisting of particles with 'location' and 'mass'



$$K((x_1, m_1), (x_2, m_2), dz) =$$

$$\bar{K}((x_1,m_1),(x_2,m_2))\,\delta_{\left(\frac{m_1x_1+m_2x_2}{m_1+m_2},\; m_1+m_2\right)}$$

Then $\phi((x_1,m_1),(x, m_2)) = m_1x_1 + m_2x_2$

is conservative.

Eventually conservative kernels

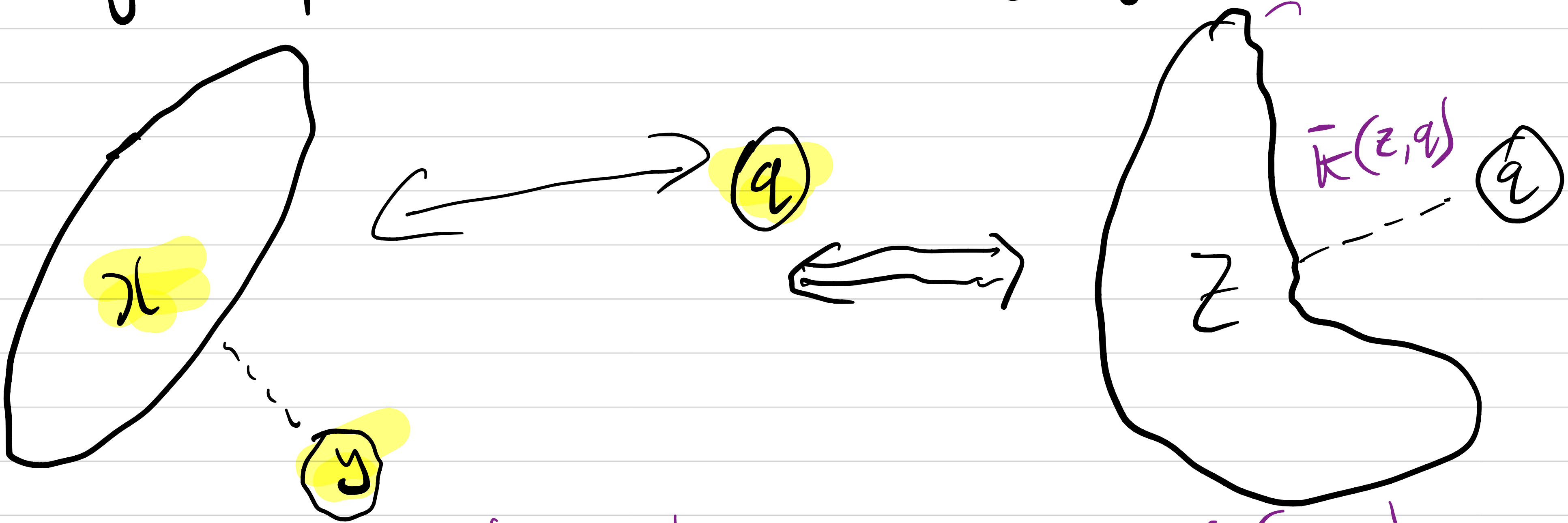
Definition: A kernel $K: E\times E\times B \to [0,\infty)$ is said to be *eventually conservative* if there exists a compact set $C \subseteq E\times E$ such that, for a conservative function $\phi: E\times E \to [0,\infty)$

$$\bar{K} = \phi \quad \text{on} \quad C^c.$$

i.e. $\forall (x,y)\in (E\times E)\setminus C \quad K(x,y,E) = \phi(x,y)$

Motivation: Eventually Conservative

When kernels are "eventually conservative" we can "predict" the behaviour of large particles as they grow.



$$\bar{K}(z,q) = \phi(z,q) = \phi(x,q) + \phi(y,q)$$

Assumptions 1:

1) $\bar{K}: E \times E \to [0,\infty)$ is continuous and for some $C > 0$ $\bar{K}(x,y) \leq C\, m(x)\, m(y)$

$= K(x,y,E)$

(This assumption can be weakened)

2) K is eventually conservative.

or $\bar{K}(x,y) \leq m(x) + m(y)$

3) The measures $\{ L_t^{(N)} : t \geq 0,\ N \in \mathbb{N} \}$ are tight.

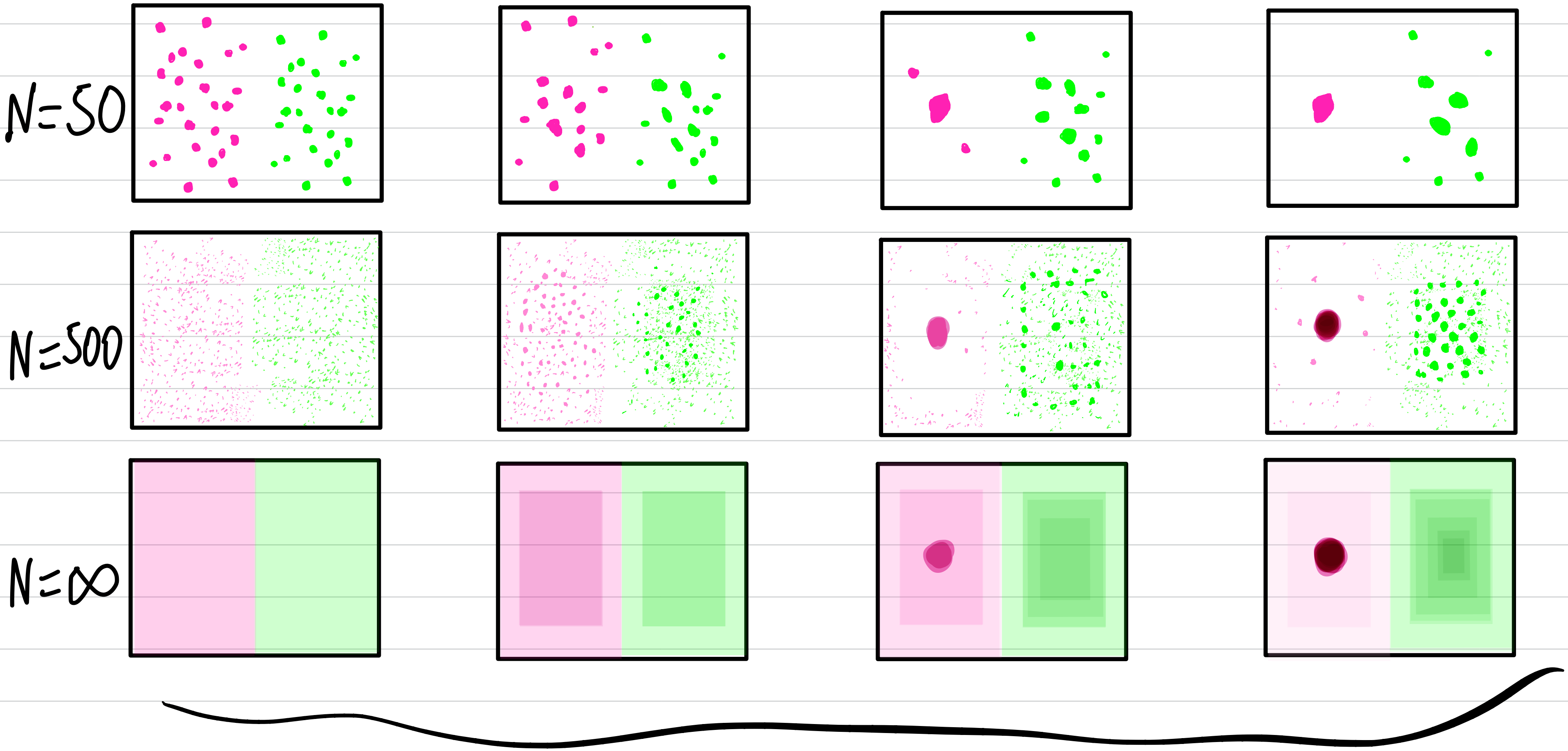
(True under various assumptions, difficult to capture globally)

Theorem: (Andreis, I., Magnanini, 23+)

Assume E is a metric space, and Assumption 1 is satisfied. Then, there exists a trajectory $(\mu_t)_{t\geq 0}$ of measures on E such that

$$\left(L_t^{(N)}\right)_{t\geq 0} \xrightarrow[\text{Prob.}]{N\to\infty} (\mu_t)_{t\geq 0}$$

(wrt Skorokhod J_1 metric, induced by Prokhorov metric on finite measures on E (which metrises weak topology))



Can characterise limiting object!

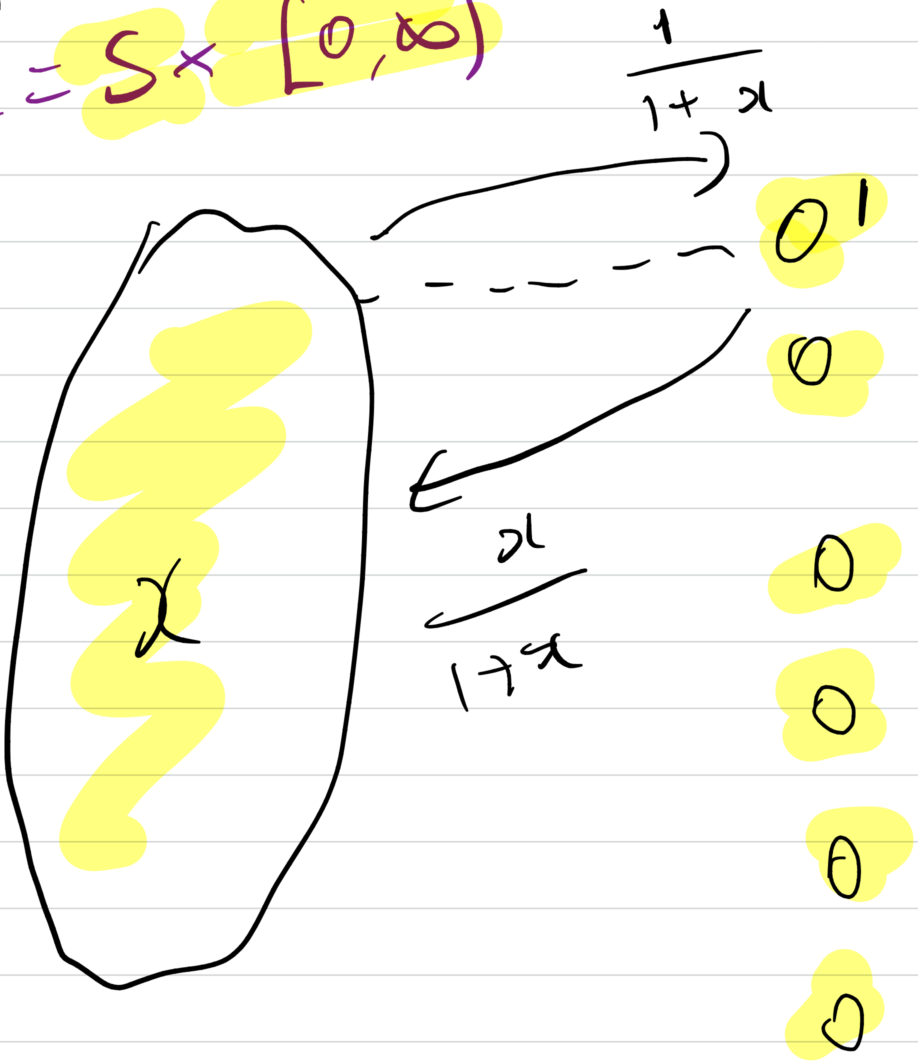
Remark: Have more general criteria for $\{\mathbb{P}_N(\cdot) : N \in \mathbb{N}\}$ to be relatively compact, with accumulation points $\mathbb{P}^*(\cdot)$ concentrating on solutions to "Smoluchowski" and "Flory" equations.

(WLLN if there exists a unique solution)

Remark: Not sure we expect more generality in WLCN.

"Spatial model"

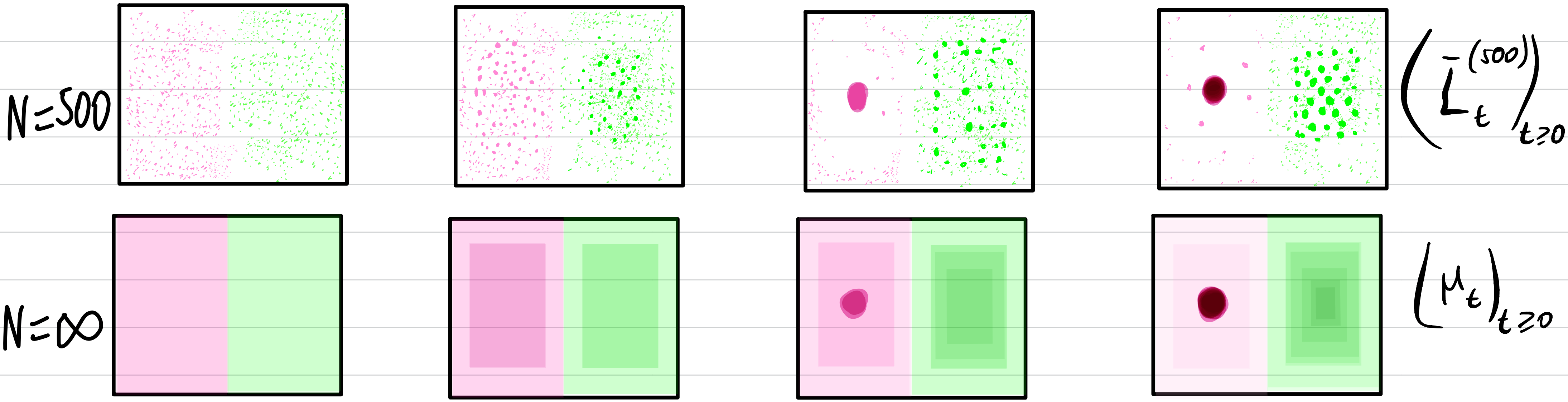
$E = S \times [0, \infty)$



Single 'giant' particles, which influence the system may fluctuate randomly

Gelation: When do we see "large" particles??

- In applications, one is often interested in whether these large particles form.



Is there loss of mass?

Is there mass lost 'to infinity' in $(\mu_t)_{t\geq 0}$?

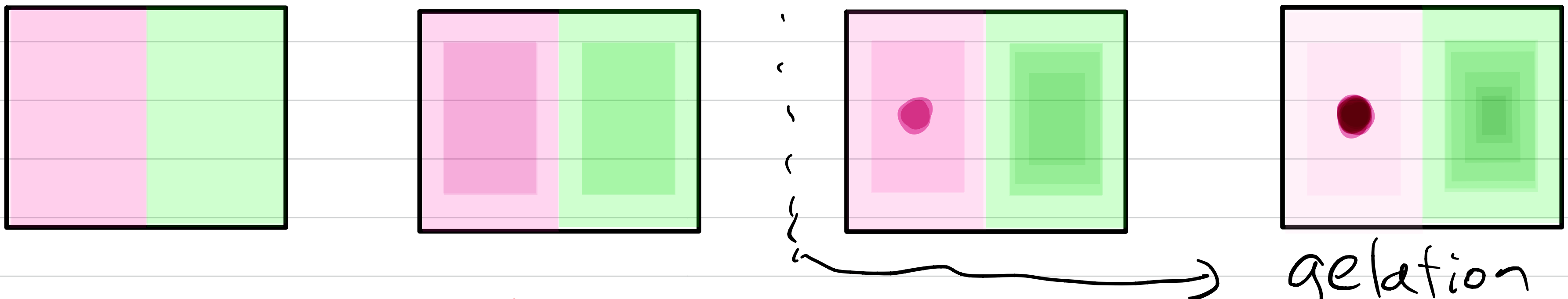
Gelation: When do we see "large" particles??

Definition: Given a trajectory of measures $(\mu_t)_{t\geq 0}$ we say "gelation" occurs if, for some $t^* > 0$

$$\int_E m(x)\,\mu_{t^*}(dx) < \int_E m(x)\,\mu_0(dx)$$

mass "lost" in giant particles.

$N=\infty$

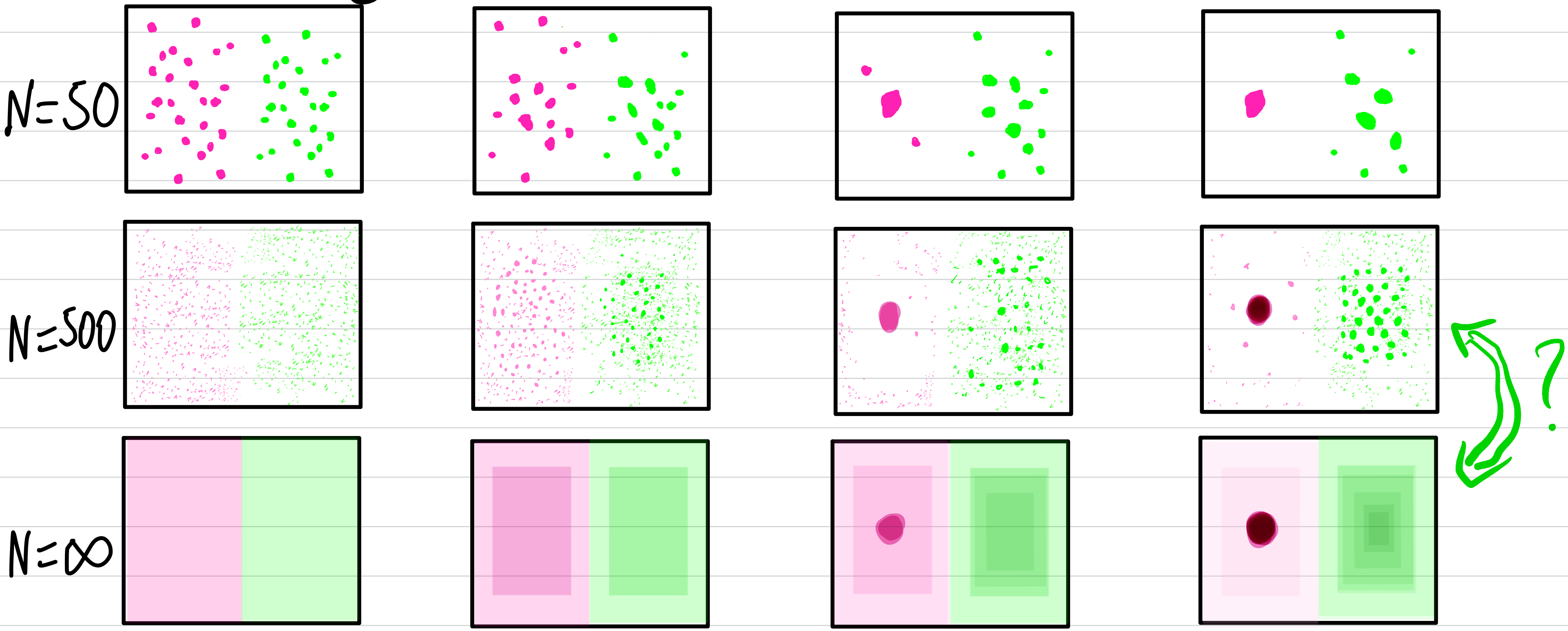


$(\mu_t)_{t\geq 0}$

gelation in pink.

"Analytic" use differential equations

Can we find a notion of gelation using $\left(\bar{L}_t^{(N)}\right)_{t\geq 0,\, N\in\mathbb{N}}$? "Probabalistic"



Stochastic gelation $\Longleftrightarrow$ with positive probability

$$\int m(x)\,\mu_t(dx) < \int m(x)\,\mu_0(dx)$$

Gelation: What fraction of mass is contained in "large" particles?

$$\int_{\{x : m(x) > \psi(N)\}} m(x) \, \bar{L}_t^{(N)}(dx) > c \ ?$$

$$\lim_{N \to \infty} \psi(N) = \infty$$

"Proportion of total mass in particles of mass $\geq \psi(N)$"

Gelation: When are there large particles with positive probability?

Definition: Given a sequence of trajectories $(\bar{L}_t^{(N)})_{t \geq 0}$ and a function $\psi : \mathbb{N} \to \mathbb{N}$ with $\lim_{N \to \infty} \psi(N) = \infty$, and $\delta > 0$,

large particle size.

Set $T^{\psi,\delta} :=$

$$\inf \left\{ t > 0 : \liminf_{N \to \infty} \mathbb{P}\left(\int_{\{x : m(x) > \psi(N)\}} m(x) \, \bar{L}_t^{(N)}(dx) > \delta \right) > 0 \right\}$$

First time δ-prop of mass in particles of size $\geq \psi(N)$ with positive probability uniformly in N.

Definition: We say "stochastic gelation" occurs, if, for some ψ, δ,

$$T^{\psi,\delta} < \infty$$

"There exists some $t_0 > 0$ after which, with positive probability, a positive fraction of the mass of the system is in particles of mass $\geq \psi(N)$"

Equivalence of Criterion

Theorem (Andreis, I., Magnanini, 23+ / Pretty much Jeon '98)

Suppose that $m : E \to [0,\infty)$ is continuous, on a subsequence $\left(\bar{L}_t^{(N_k)}\right)_{t\geq 0} \xrightarrow[k\to\infty]{\text{weak}} \left(\bar{L}^*_t\right)_{t\geq 0}$; where $\left(\bar{L}^*_t\right)_{t\geq 0}$ is continuous, and $\int_E m(x)\, \mu_0(dx) < \infty$. The following are equivalent:

i) Stochastic gelation in $\left(\bar{L}_t^{(N_k)}\right)_{t\geq 0,\, k\in\mathbb{N}}$

ii) Gelation with positive probability in $\left(\bar{L}^*_t\right)_{t\geq 0}$

$\mathbb{P}^*(\text{gelation}) > 0$

$$\int \mu_t(dx)\, m(x) < \int \mu_0(dx)\, m(x)$$

Expect $(\bar{L}^{*}_{t})_{t \geq 0}$ to satisfy Smoluchowski or Flory equations

Probabalistic

Analytic

bridge

(Jeon, 1998)

Stochastic gelation

Existence of gelling solutions to Smoluchowski or Flory equations

Examples:

In the case $E = [0, \infty)$, $m(x) = x$

& $K(x, y, dz) = \bar{K}(x, y)\,\delta_{x+y}$

If $\bar{K}(x,y) = xy$ stochastic gelation occurs around time $t=1$.

('giant component' in Erdős - Rényi random graph).

$\left(L_t^{(N)}\right)_{t \geq 0} \longrightarrow \left(M_t\right)_{t \geq 0}$ satisfying 'Flory' equation

- Gelation occurs in $\left(M_t\right)_{t \geq 0}$.

$\left(\text{i.e} \quad \int m(x)\, M_1(dx) < \int m(x)\, M_0(dx)\right)$

More generally:

If $\bar{K}(x,y) \geq (m(x)\,m(y))^{1/2+\varepsilon}$ stochastic gelation occurs. [Jeon '98, Rezakhanlou 2013]

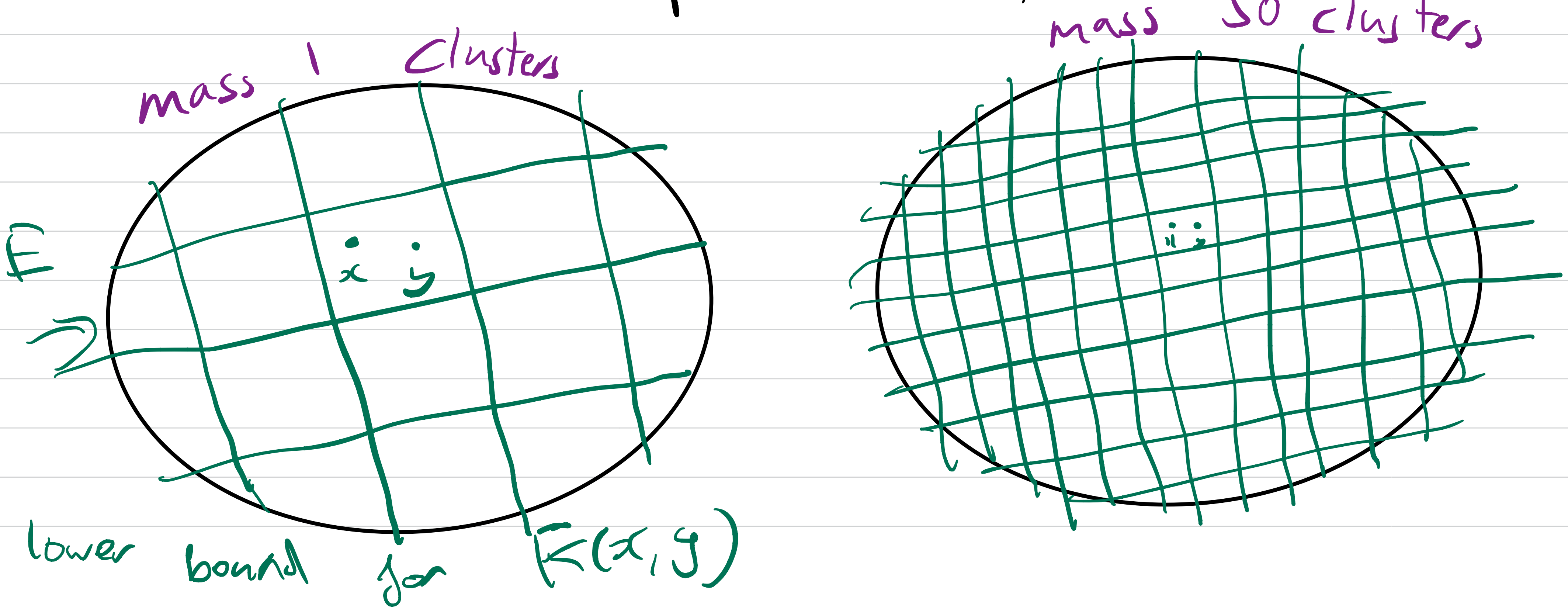
If $\bar{K}(x,y) \leq m(x) + m(y)$ stochastic gelation does not occur [Jeon '98, Norris '98]

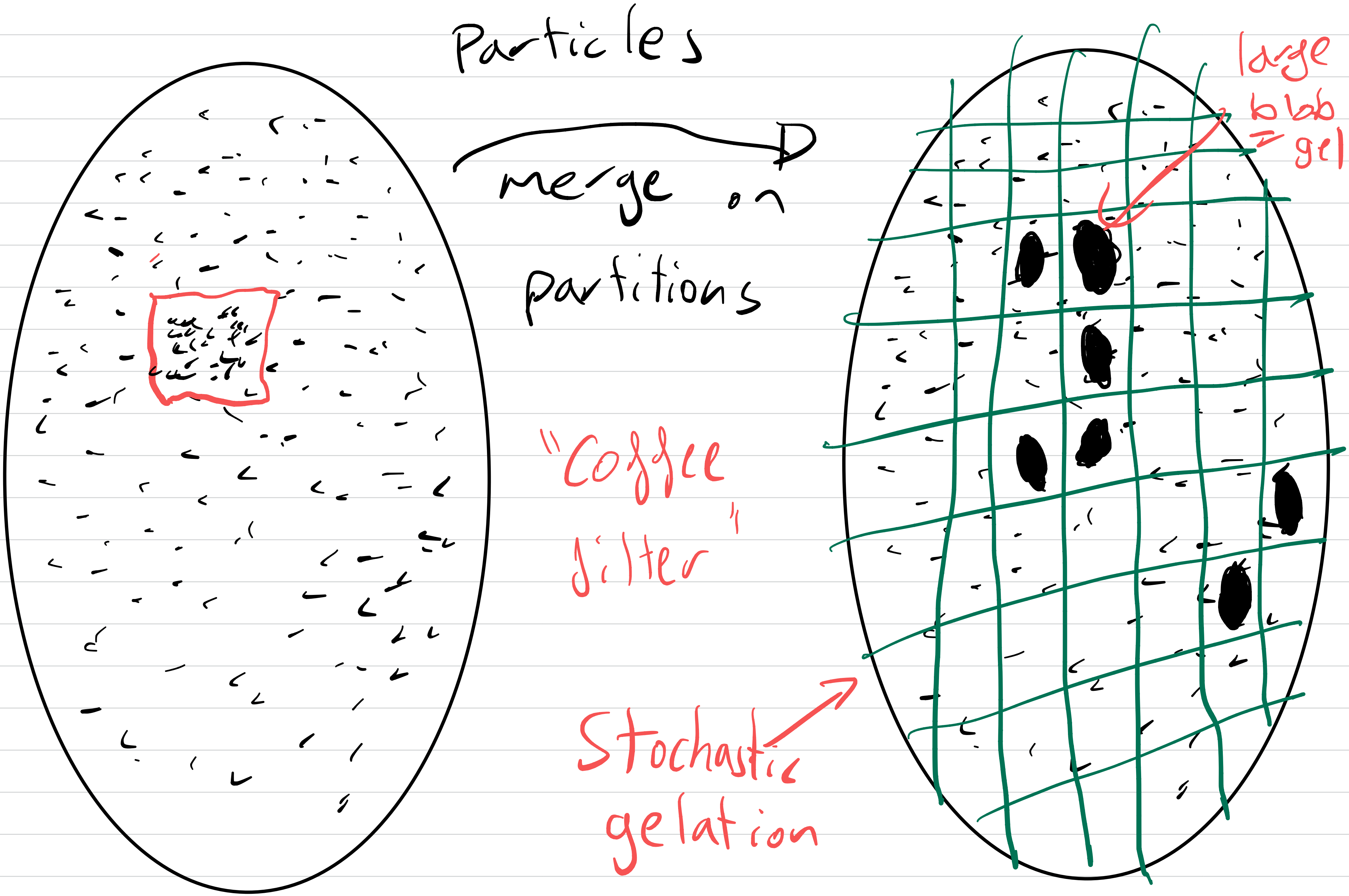
Goal: General criteria for stochastic gelation incorporating information about E

More technical

Assumptions for gelation:

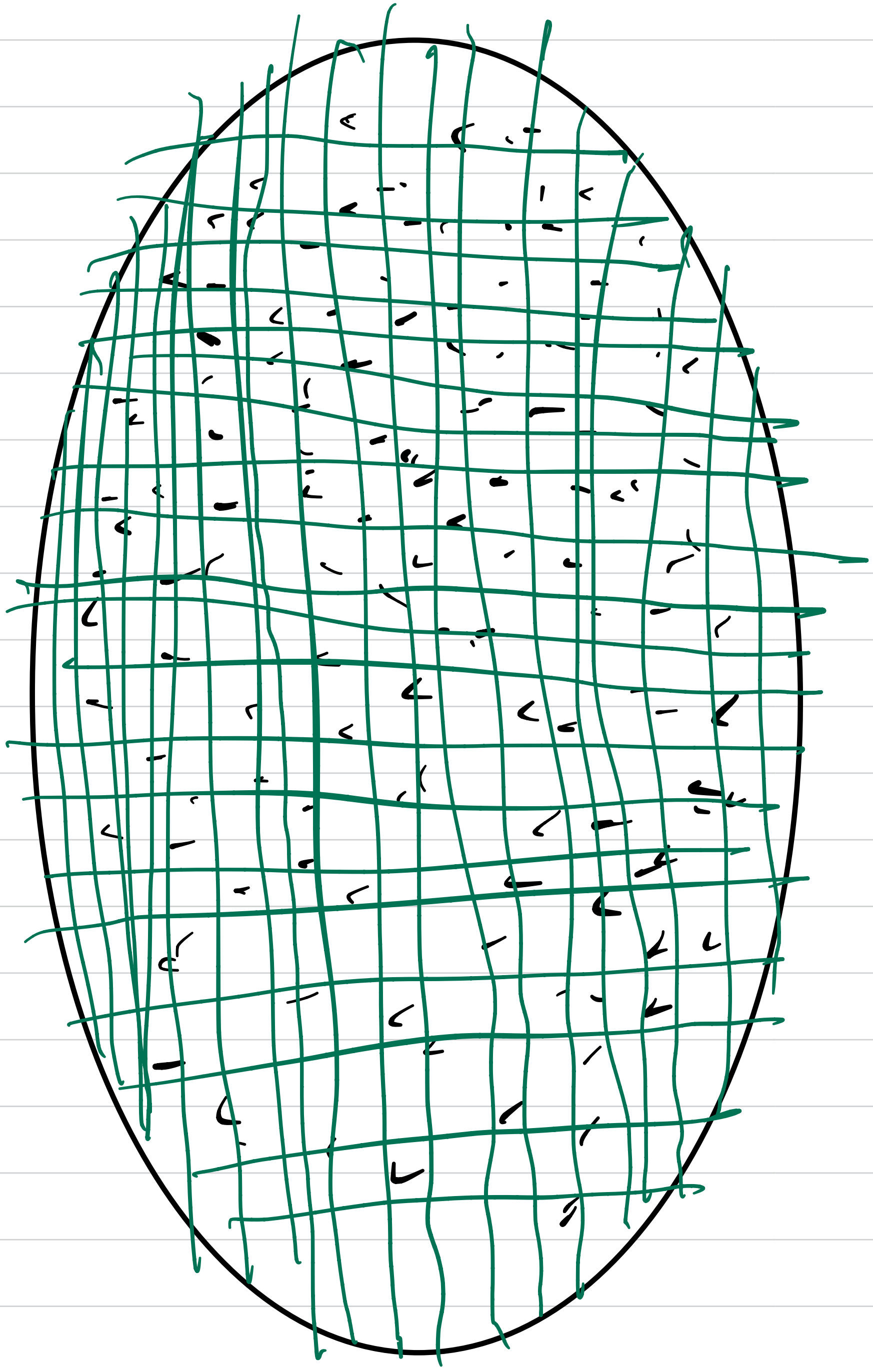
1) We can partition E, to find lower bounds on the rate at which particles merge when in the same set in the partition, and having similar mass.




Particles
merge on partitions
"coffee filter"
Stochastic gelation
large blob = gel

But the partitions can't be too fine!

Particles can't grow from initial condition.



Assumptions for gelation:

Fix a strictly decreasing sequence

$(\delta_i) \downarrow \delta$, $\quad f : \mathbb{N} \to \mathbb{N}$ and

measurement of 'partition size'

a non-decreasing function

$\psi : \mathbb{N} \to \mathbb{N}$, with $\lim_{N \to \infty} \psi(N) = \infty$

proportion of mass in 'gel'

'large particles'

1 There exists a family of partitions $\{\mathcal{P}^{(j)}\}_{j \in \mathbb{N}}$ of E, where

$\forall j \leq \log_2 \psi(N) \quad |\mathcal{P}^{(j)}| \leq \xi(N)$

(Bound on partition size)

and for $P \in \mathcal{P}^{(j)}$ $\exists\ C(P, j) > 0$ such that

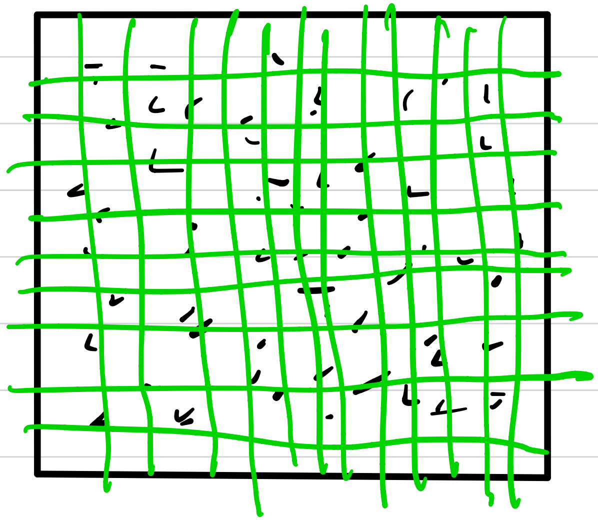
$\bar{K}(x, y) \geq C(P, j)$ when $x, y \in P$ and $2^j \leq m(x), m(y) < 2^{j+1}$

(Bound on the rate at which clusters merge when taking values in certain sets)

② $$\limsup_{N\to\infty} \sum_{j=1}^{\log_2(\psi(N))} \frac{1}{(\delta_j - \delta_{j+1})^2} \left(\sum_{P\in\mathcal{D}^{(j)}} \frac{1}{C(P,j)} \right) 2^j < \infty$$

(Summability condition: uniform bound on waiting time for "large" clusters to form.)

③ $$\lim_{N\to\infty} \frac{\psi(N)\,\xi(N)}{N} = 0$$



(Partition not too "fine")

(4) For each $N \in \mathbb{N}$, there exists $g(N)$ such that $\forall s \in [0,\infty)$

$$\int_{E\times E} \bar{L}_s^{(N)}(dx)\, \bar{L}_s^{(N)}(dy)\, \bar{K}(x,y)\, m(x) < g(N).$$

(Annoying technical condition particles don't move in a way that causes instantaneous things to happen)

Theorem: (Andreis, I., Magnanini, 2023+)

Under the above assumptions stochastic gelation occurs for

$$(\bar{L}_t^{(N)})_{t\geq 0}.$$

Remark: Works for arbitrary measure space $(E,\mathcal{B})$.

Simpler Examples: (Case $E=[0,\infty)$)

Corollary: (Andreis, I., Magnanini)

Stochastic gelation occurs if

1) $\inf_{i\in[1,2]} \bar{K}(1,i) > 0$ & for all x, y sufficiently large

$$\bar{K}(cx, cy) \geq c^{\gamma}\, \bar{K}(x,y)$$

with $\gamma > 1$

Generally accepted principle in scientific modelling literature (see, e.g., (Aldous '99))

<u>Corollary</u>: (Andreis, I., Magnanini)

Stochastic gelation occurs if

2) There exists $\varepsilon > 0$ such that for x, y sufficiently large

$$\bar{K}(x,y) > \big(m(x) \wedge m(y)\big) \log\big(m(x) \wedge m(y)\big)^{3+\varepsilon}$$

<u>Remark</u>:

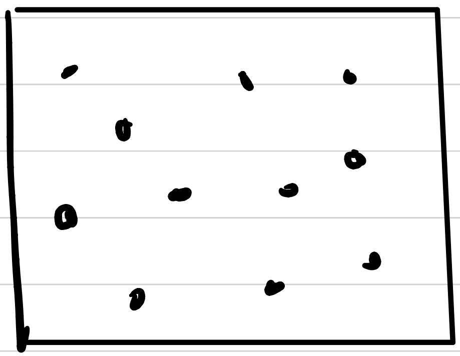
(But we know gelation does not occur if $\bar{K}(x,y) \leq m(x) + m(y)$)

Example

Suppose $E = [0,1]^d \times [0,\infty)$

and we begin with N clusters of mass 1, sampled iid,

$$K((p,n),(s,o)) = \bar{K}((p,n),(s,o))\,\delta\left(\frac{pn+so}{n+o},\, n+o\right)$$



$$\bar{K}((p,n),(s,o)) = \begin{cases} 1/\|p-s\|_\infty^\alpha & \text{if } p \neq s \\ 0 & \text{otherwise} \end{cases}$$

stochastic gelation if $\alpha/d > 1$

Comments / Future Directions

- Exploring criteria for the absence of gelation.

$$\overline{K}(x,y) \leq m(x) + m(y)$$

- Adding fragmentation?

- Spatial models with independent movement?

Thank You!