Gelation of stochastic diffusion-coagulation systems

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Abstract

We investigate aerosol systems diffusing in space and study their gelation properties. In particular we work out the role of fluctuations which turn stable configurations into metastable ones, and the influence of randomly distributed sources and sinks.

Key words: Smoluchowski equation, diffusion, random perturbations, gelation.

MSC classification: 82C99, 60K35

1 Introduction

This paper continues the investigation of the influence of random fluctuations on the gelation process of an aerosol system which has been initiated in [5]. The governing equation for aerosol dynamics is the Smoluchowski equation with the particle interaction operator being an integral operator which describes the growth of aerosol particles due to clustering. *Gelation* is the effect that particles may grow to *macroparticles* (i.e. particles of infinite mass) within finite time.

For some decades, the study of gelation has attracted considerable attention in the

scientific community; it is now a well-investigated phenomenon – for special integral kernels and as far as the space-homogeneous case is considered. (As a survey, look at the paper of Aldous [1].) The case of a coagulating system in combination with some spatial dynamics, however, was treaten very rarely. In [5] it was shown that a typical feature of coagulating systems in the presence of sinks and sources is a certain *fold-bifurcation* structure. It consists of the presence of two steady solutions, one of which is stable, the other unstable. Crossing the unstable solution leads to the effect of gelation. *Fluctuations* eventually lead to crossing the instability bound, thus turning formerly stable states into metastable ones.

The objective of the paper is threefold. First, we consider how gelation can be modelled in a numerical simulation. While the emergence of *physical* macroparticles can happen only after an infinite number of particle interactions, *numerical* methods have to be based on some truncation technique. We use the second moment of the aerosol distribution as control parameter and establish a useful truncation criterion. Second, we illuminate in a simple example the role of fluctuations in destabilazing formerly stable configurations. Fluctuations may enter into the system via the choice of random instead of deterministic sources, or via replacing diffusion with classical random walk. Our method of choice is to use as a numerical method a Monte Carlo scheme which naturally introduces fluctuations. As a third point, we demonstrate how a random distribution of sources and sinks can serve as a nucleus for gelation.

The scope of the paper is as follows. In section 2 we introduce the Smoluchowski equation and a Monte Carlo scheme for its numerical simulation. We show how the second moment system can be used to introduce a threshold for the simulation of gelation. In section 3 we combine the Smoluchowski equation with a diffusion model in space. For the deterministic case we prove the existence of stable configurations which turn out to become metastable once fluctuations are introduced. Section 4 concerns the case of a completely deterministic dynamics in a steady random environment. Here, we consider coagulation as a small perturbation of the classical Parabolic Anderson Model;

we demonstrate how the leading eigenmodes of this model are initiators of the gelation process.

2 The Smoluchowski system

2.1 The equations

The *space homogeneous discrete Smoluchowski equation* is a system of ordinary differential equations on $\mathbb{R}_+^{\mathbb{N}}$. Denoting $\mathbf{f} = (f_i)_{i=1}^{\infty}$, these equations read

$$\partial_t f_i = \frac{1}{2} \sum_{j=1}^{i-1} K(j, i-j) f_j f_{i-j} - f_i \sum_{j=1}^{\infty} K(i,j) f_j. \tag{2.1}$$

Here, $K(x,y)$ is some symmetric nonnegative kernel. Throughout the paper we will use

$$K(i,j) = ij \quad . \tag{2.2}$$

Denote the α-th moment of $\mathbf{f}$ by

$$M_\alpha[\mathbf{f}] := \sum_{i=1}^{\infty} i^\alpha f_i. \tag{2.3}$$

Assuming $M_1[\mathbf{f}] = 1$, $\mathbf{f}$ represents a particle distribution of integer-valued masses with total mass 1. if_i represents the fraction of the whole mass which is given by the particles of mass i. The Smoluchowski system describes the growth of the particles due to two-particle clustering (coagulation).

An equivalent formulation for $\mathbf{g} = (g_i)_{i=1}^{\infty} = (if_i)_{i=1}^{\infty}$ with the kernel (2.2) is given as

$$\partial_t g_i = \sum_{j=1}^{i-1} j g_j g_{i-j} - i g_i \sum_{j=1}^{\infty} g_j =: B_i[\mathbf{g}, \mathbf{g}] \quad . \tag{2.4}$$

A particularly important role is played by the second moment

$$M_2[\mathbf{f}] = \sum_{j=1}^{\infty} j^2 f_j = \sum_{j=1}^{\infty} j g_j. \tag{2.5}$$

One easily finds the evolution equation for $u(t) = M_2[\mathbf{f}(t)]$ as

$$\partial_t u = u^2. \tag{2.6}$$

Obviously the solution of (2.6) blows up to infinity in finite time. As long as u remains finite, all the sums of equation (2.1) are absolutely summable and $M_1[\mathbf{f}(t)]$ is a conserved quantity. After the *gelation time* t^*, defined by

$$t^* = \inf\{t > 0 | u(t) = \infty\}, \tag{2.7}$$

part of the particles grow to particles of mass ∞ and thus are not represented by the distribution $\mathbf{f}$; as a consequence, $M_1[\mathbf{f}]$ decreases in time. This effect is called *gelation*.

Due to the choice of the kernel $K(.,.)$ (see (2.2)) large particles coagulate more frequently with other particles than smaller ones. Suppose that one particle gelates into a macroparticle at some finite time t^*. Then t^* is a cumulation point of all times of interaction with the ensemble. In numerical simulations, such cumulation points are hardly recognized. E.g. when using an explicit discretization with fixed time step Δt, solutions of (2.6) remain finite for all times. On the other hand, decreasing Δt adaptively allows to approach the gelation time – but not to pass it. The numerical passage of the gelation time can only be handled efficiently by using some appropriate truncation technique. We propose two methods for which we use the second moment u as a control parameter.

2.1 Discretization of the gelation phase: (a) One method of simulating the transition to gelation is to introduce a threshold θ for the discretized second moment $u^{(n)} \sim u(n\Delta t)$. As a threshold we choose the smallest value such that the solution u of the non-discretized initial value problem for the second moment explodes within the next time step. In our simple example, we have to solve the initial value problem

$$\partial_t u = u^2, \quad u(n\Delta t) = \theta. \tag{2.8}$$

Since the solution of (2.8) is $u(t) = \theta/(1 - \theta(t - n\Delta t))$, we end up with the threshold

$$\theta = 1/\Delta t. \tag{2.9}$$

Whenever this threshold is passed, enough mass in the highest states of $\mathbf{g}^{(n)}$ is to be removed (i.e. particles are "numerically gelating") to move u below this threshold.
(b) Instead of solving the non-discretized system (which is of course simple in the simple model above but maybe more complicated in other situations) one may estimate by extrapolating the values $1/u^{(n)}, 1/u^{(n-1)}, \ldots$ the time when $1/u$ will pass the line 0, and remove mass from $g^{(n)}$ whenever the estimated passage will be in the time interval $(n\Delta t, (n+1)\Delta t]$. E.g., linear extrapolation leads to gelation whenever

$$u^{(n)} \geq 2u^{(n-1)}. \tag{2.10}$$

2.2 Monte Carlo systems

The mass flow process. In [3] a Monte Carlo scheme was proposed for the numerical simulation of the Smoluchowski system. It is based on the Euler time discretization for $\mathbf{g}^{(n)} = \mathbf{g}(n \cdot \Delta t)$

$$\mathbf{g}^{(n+1)} = \mathbf{g}^{(n)} + \Delta t \cdot \mathbf{B}[\mathbf{g}^{(n)}, \mathbf{g}^{(n)}] \tag{2.11}$$

where $\mathbf{B} = (B_i)_{i=1}^{\infty}$ represents the evolution operator for $\mathbf{g}$, see (2.4). In its weak formulation, this equation reads

$$\sum_{i=1}^{\infty} \phi_i g_i^{(n+1)} = \sum_{i=1}^{\infty} \sum_{j=1}^{\infty} \left[i\Delta t \cdot \phi_{i+j} + (1 - i\Delta t) \cdot \phi_i \right] g_i^{(n)} g_j^{(n)} \tag{2.12}$$

for any bounded test function ϕ on $\mathbb{N}$. This equation indicates a mass flow from state i to state $i+j$ with ratio proportional to $i\Delta t$. It is the basis for the following scheme for stochastic simulation.

Assuming $M_1[\mathbf{f}(0)] = 1$, we start with an N-tupel $\mathbf{z}^{(0)} = (z_1, \ldots, z_N) \in \mathbb{N}^N$ (the entry z_i representing a particle of mass $1/N$ with "label" i) such that

$$\frac{1}{N} \sharp\{j | z_j = i\} \approx g_i^{(0)}. \tag{2.13}$$

The transition from $\mathbf{z}^{(n)} = (z_1, \ldots, z_N)$ to $\mathbf{z}^{(n+1)} = (z'_1, \ldots, z'_N)$ is given by the following random game.

(i) For $i = 1, \ldots, N$ choose independent equidistributed random numbers $\pi_i \in \{1, \ldots, N\}$;

(ii) define

$$z_i' := \begin{cases} z_i & \text{with probability} \quad 1 - \Delta t \cdot z_i \\ z_i + z_{\pi_i} & \text{with probability} \quad \Delta t \cdot z_i \end{cases} .$$

2.2 Remarks: (a) Let ϕ be a test function on $\mathbb{N}$ and denote

$$\overline{\phi(\mathbf{z})} := \frac{1}{N} \sum_{i=1}^{N} \phi(z_i) \tag{2.14}$$

for any given vector $\mathbf{z} = (z_1, \ldots, z_N)$. The conditional expectation after one time step is

$$\mathcal{E}(\overline{\phi(\mathbf{z}^{(n+1)})} | \mathbf{z}^{(n)}) = \frac{1}{N^2} \sum_{i=1}^{N} \sum_{j=1}^{N} \left[z_i \Delta t \cdot \phi(z_i + z_j) + (1 - z_i \Delta t) \phi(z_i) \right]. \tag{2.15}$$

From this and the law of large numbers follows that in the limit $N \to \infty$ the distribution of $\mathbf{z}^{(n+1)}$ converges a.s. to $\mathbf{g}^{(n+1)}$ provided the distribution of $\mathbf{z}^{(n)}$ converges to $\mathbf{g}^{(n)}$. (A proof can be found in [3].) In this sense the Monte Carlo game produces solutions of the time discretized Smoluchowski system.

(b) Choosing $\phi(\mathbf{z}) = \sum z_i$, $\overline{\phi(\mathbf{z}^{(n)})}$ represents the second moment of $\mathbf{f}$, and we find

$$\mathcal{E}(\overline{\phi(\mathbf{z}^{(n+1)})} | \mathbf{z}^{(n)}) = \overline{\phi(\mathbf{z}^{(n)})} + \Delta t \cdot \overline{\phi(\mathbf{z}^{(n)})}^2 \tag{2.16}$$

which corresponds to the discretization of equation (2.6).

2.3 Modification: The time discretization for the mass-flow process only makes sense as long as $\Delta t \cdot \max_j z_j < 1$. This means that the time step has to be permanently reduced while particles progress into the domain of increasing mass. To circumvent this, we split step *(ii)* of the above algorithm into z_{π_i} partial steps by the following sequence written in pseudo code.

put $z_i' := z_i$

for $k = 1$ to z_i do

$z_i' := z_i' + z_{\pi_i}$ with probability Δt

2.4 Remark: For small Δt, the conditional expectation of the modified scheme satisfies

$$\mathcal{E}(\overline{\phi(\mathbf{z}^{(n+1)})}|\mathbf{z}^{(n)}) = \frac{1}{N^2}\sum_{i=1}^{N}\sum_{j=1}^{N}[z_i\Delta t\cdot\phi(z_i+z_j)+(1-z_i\Delta t)\phi(z_i)]+\mathcal{O}(\Delta t^2). \quad (2.17)$$

As for the mass flow scheme the law of large numbers may be applied to prove that for $N\to\infty$ and for $\Delta t\to 0$ the Monte Carlo process provides the solution of the Smoluchowski system in the time interval $[0,T]$ provided T is smaller than the gelation time t^*.

A commonly used way to cope with gelation is to introduce a threshold S and to identify particles with infinite-mass particles once they pass this threshold. If particles are removed of the system once they pass S, then this corresponds to a *passive* gel, while an *active* gel lets these particles still interact with the others. The Smoluchowski system describes a passive gel. The modification for an active gel reads

$$\partial_t g_i = \sum_{j=1}^{i-1} j g_j g_{i-j} - i g_i \quad (2.18)$$

– provided $M_1[\mathbf{f}(0)] = 1$.

2.3 Numerical simulation of the gelation phase

We test the modified algorithm simulating the Smoluchowski system with active and with passive gel for the initial condition $g_i(0) = \delta_{i1}$. From solving the second-moment system $u' = u^2$, $u(0) = 1$ we know that the gelation time is $t^* = 1$. From a heuristic argument which has been proposed and investigated in [4] we may learn the asymptotic evolution of the system in the gelation phase. Denote

$$\Phi(t) := \left(\sum_{i=1}^{\infty} g_i(t)\right)^{-1}. \quad (2.19)$$

Then $\Phi(t)\cdot\mathbf{g}$ is a system with ℓ_1-norm $\|\Phi(t)\cdot\mathbf{g}\|_1 = 1$. The basic assumption is that this system converges for $t\to\infty$ to a constant sequence $\mathbf{r}$. In the case of a *passive* gel, denote

$$\mathbf{h}(\tau) := \Phi(t)\cdot\mathbf{g}(t) \quad \text{with} \quad \frac{\partial\tau}{\partial t} = \Phi(t). \tag{2.20}$$

The evolution equation for $\mathbf{h}$ is

$$\partial_\tau h_i = \partial_t\Phi\cdot h_i + \sum_{j=1}^{i-1} j h_j h_{i-j} - i h_i. \tag{2.21}$$

Necessary for this system to converge to $\mathbf{r}$ is that $\partial_t\Phi\to c = \text{const}$ for $t\to\infty$ indicating that

$$\sum_{i=1}^{\infty} g_i(t) \sim \frac{1}{t} \quad \text{and} \quad \sum_{i=1}^{\infty} f_i(t) \sim \frac{1}{t}. \tag{2.22}$$

In the case of an *active* gel choose

$$\mathbf{h}(t) := \Phi(t)\cdot\mathbf{g}(t). \tag{2.23}$$

In this case, assuming that $\Phi(t)\to\infty$ for $t\to\infty$ we obtain

$$\partial_t h_i = \frac{\partial_t\Phi}{\Phi}\cdot h_i + \frac{1}{\Phi}\sum_{j=1}^{\infty} j h_j h_{i-j} - i h_i \to \frac{\partial_t\Phi}{\Phi}\cdot h_i - i h_i. \tag{2.24}$$

Necessary for the convergence is that $\partial_t\Phi/\Phi\to c = \text{const}$ proving that

$$\sum_{i=1}^{\infty} g_i(t) \sim \exp(-ct) \quad \text{and} \quad \sum_{i=1}^{\infty} f_i(t) \sim \exp(-ct). \tag{2.25}$$

We want to mention that at least for the passive gel these results are backed up by rigorous theory (cf. [1, 6]).

The figures 1 and 2 show the results of Monte Carlo simulations. They exhibit the evolution of the particle numbers (M_0, dotted lines) and of the total mass (M_1, solid lines). They confirm the above heuristic arguments. (Notice that Fig. 1 is plotted on a *log-log*-scale and Fig. 2 on a *log*-scale.)

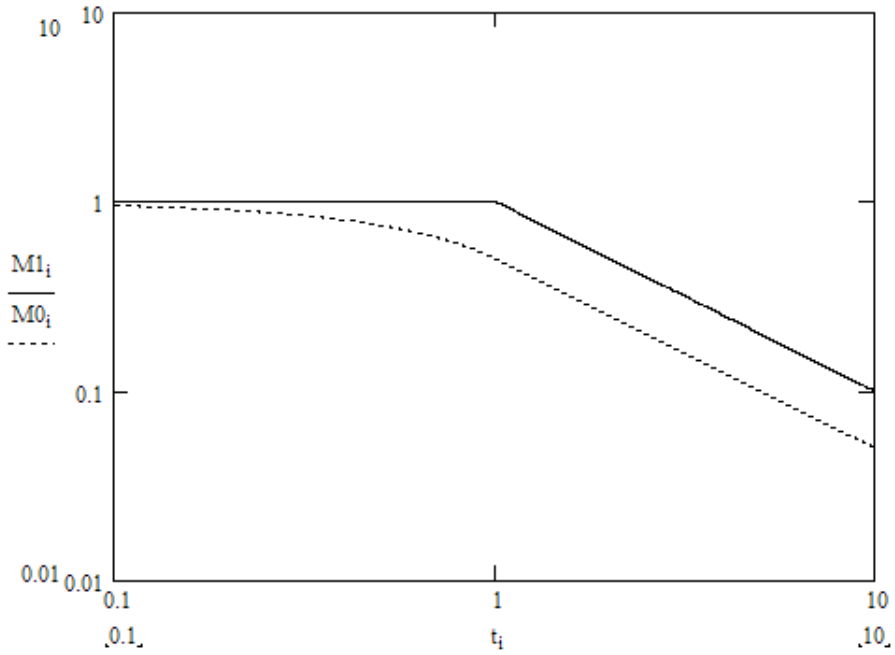

Fig. 1: First moments of a passive gel

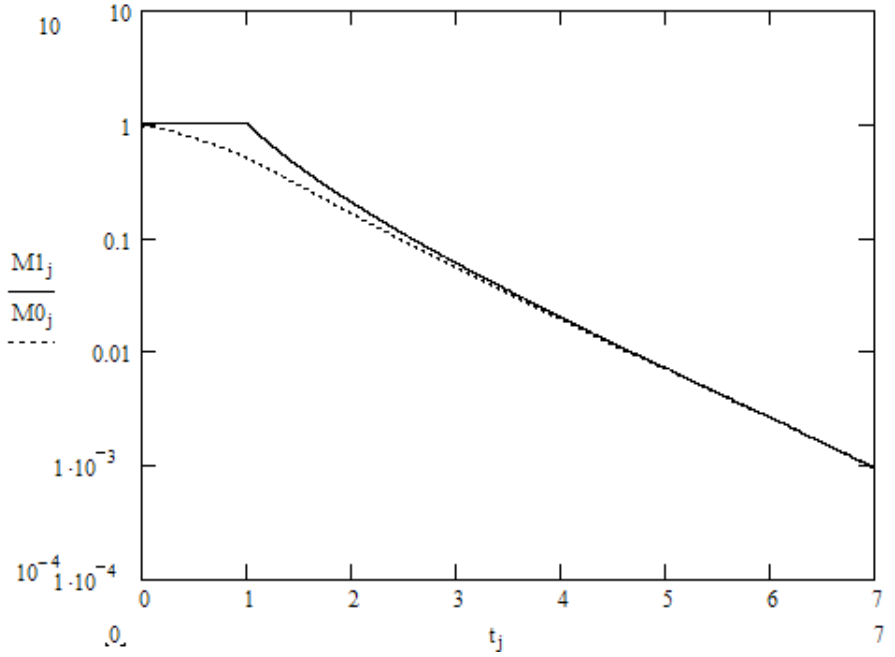

Fig. 2: First moments of an active gel

3 Coagulation with spatial diffusion

3.1 The deterministic – i.e. non-fluctuating – case

We study an aerosol system with diffusion in physical space in the presence of a constant monomer source s and a linear sink σ. The physical space is a discretization of the interval $\Omega = [-1, 1]$ given by $\Omega_h = h \cdot \{-p, -p+1, \ldots, p-1, p\}$, where $p \cdot h = 1$. The motion in Ω_h is given as discrete diffusion with periodic boundary conditions described by the $2p \times 2p$-matrix

$$\Delta_h = \frac{1}{h^2} \begin{pmatrix} -2 & 1 & & & 1 \\ 1 & -2 & 1 & & \\ & \ddots & \ddots & \ddots & \\ & & 1 & -2 & 1 \\ 1 & & & 1 & -2 \end{pmatrix}. \tag{3.1}$$

(Here, the points $-ph$ and $+ph$ have been identified.) The equation for the distribution function $\mathbf{g}(t, x) = (g_i(t, x)_{i=1}^{\infty}$ reads

$$\partial_t g_i(t, x) = \Delta_h g_i(t, x) + B_i[\mathbf{g}(t, x), \mathbf{g}(t, x)] + \delta_{i1} s - \sigma g_i(t, x) \quad . \tag{3.2}$$

The evolution equation for the second moment

$$u(t, x) = \sum_{i=1}^{\infty} i g_i(t, x) \tag{3.3}$$

follows as

$$\partial_t u = \Delta_h u + u^2 - \sigma u + s. \tag{3.4}$$

It is easy to prove that an upper bound $v(t)$ for the maximum $\overline{u}(t) = \max_x(u(t, x))$ is given by the solution of the IVP

$$\partial_t v = v^2 - \sigma v + s, \quad v(0) \geq \overline{u}(0) > 0. \tag{3.5}$$

Solutions to this problem have been analyzed in [5, 3.2 Corollary]. In particular it has been shown that the stationary version exhibits a *fold bifurcation*. This means that up

to a limit s_0 for the strength s of the source, there exist two stationary solutions, while for $s > s_0$ there exists none. In the evolution problem, the lower of these solutions is stable, while the upper one is unstable. The precise result reads

3.1 Lemma: (a) If $s > \sigma^2/4$, then there exists $t^* < \infty$ such that $\lim_{t \nearrow t^*} v(t) = \infty$.
(b) Suppose $s < \sigma^2/4$ and define $v_\pm := \sigma/2 \pm \sqrt{\sigma^2/4 - s}$. If $v(0) < v_+$ then $v(t) \searrow v_-$ as $t \to \infty$. If $v(0) > v_+$ then there exists $t^* < \infty$ such that $\lim_{t \nearrow t^*} v(t) = \infty$.

We come back to the full space dependent system (3.4) with initial condition

$$u(0, x) = u_0(x) \geq 0 \quad \text{for all} \quad x \in \Omega_h. \tag{3.6}$$

The periodic boundary conditions allow for the establishment of an easy to analyze homogenized version. Denote

$$a(t) := \frac{1}{2p} \sum_x u(t, x) \tag{3.7}$$

and

$$a_0 := \frac{1}{2p} \sum_x u_0(x). \tag{3.8}$$

Then a is the solution of the IVP

$$\partial_t a = a^2 + V[u] - \sigma a + s, \quad a(0) = a_0, \tag{3.9}$$

with the variance $V[u]$ defined as

$$V[u(t)] = \frac{1}{2p} \sum_x (u(t, x) - a(t))^2. \tag{3.10}$$

We find

$$\partial_t V[u] \;=\; \frac{2}{2p} \sum_x (u - a) \Delta_h u + \frac{2}{2p} \sum_x (u + a - \sigma)(u - a)^2 \tag{3.11}$$

and

$$\sum_x (u - a) \Delta_h u \;=\; \sum_x u \Delta_h u = -\frac{1}{2h^2} \sum_x (u_{x+h} - u_x)^2 \leq 0. \tag{3.12}$$

Thus if we can guarantee that $u + a - \sigma \leq 0$ then we have a control over $V[u]$ and can formulate conditions under which the coagulation-diffusion system (3.2) will not gelate in finite time. A necessary and a sufficient condition are given in the following.

3.2 Theorem: We assume $s < \sigma^2/4$ and define $v_\pm$ as im Lemma 3.1.
(a) Suppose $a_0 > v_+$. Then the solution of the IBVP (3.4), (3.6) explodes in finite time, i.e. there exists $t^* < \infty$ such that

$$\max_x u(t,x) \nearrow \infty \quad \text{for} \quad t \nearrow t^*. \tag{3.13}$$

(b) Suppose

(i) the source strength s and the initial variance $V[u_0]$ satisfy

$$s + V[u_0] < \sigma^2/4; \tag{3.14}$$

(ii) there exists $v_0 \in (v_-, \sigma/2)$ satisfying

$$\max_x u_0(x) \quad \leq \quad v_0. \tag{3.15}$$

Then the solution $u(t,x)$ remains bounded for all times and satisfies

$$\lim_{t\to\infty} u(t,x) = v_- \quad \text{for all} \quad x. \tag{3.16}$$

Proof *of (a)*: Since $V[u(t)] \geq 0$, a subsolution w for a is given as the solution of the IVP

$$\partial_t w = w^2 - \sigma w + s \quad , w(0) = a_0. \tag{3.17}$$

Corresponding to Lemma 3.1(b), w explodes in finite time.
Proof of (b): $u(t,x)$ (and therefore $a(t)$) is bounded from above by the solution v of the

IVP (3.5) which according to Lemma 3.1(b) is monotonically decreasing and converges to v_-. Thus for $t \geq 0$,

$$u(t) + a(t) \leq v(t) + a(t) \leq 2v(t) \leq 2v_0 \leq \sigma \tag{3.18}$$

or equivalently

$$u(t) + a(t) - \sigma \leq 0. \tag{3.19}$$

Together with (3.11) and (3.12) this proves that $V[u(t)]$ is monotonically decreasing in time and thus convergent. Denote

$$V_\infty := \lim_{t\to\infty} V[u(t)]. \tag{3.20}$$

Define $\tilde{s} := s + V[u(0)] \leq s + V[u(t)]$. Then $a(t)$ is bounded from above by the bounded solution $\tilde{a}$ of the IVP

$$\partial_t \tilde{a} = \tilde{a}^2 - \sigma\tilde{a} + \tilde{s}, \quad \tilde{a}(0) = a_0. \tag{3.21}$$

We conclude that a is bounded and that

$$a_\infty := \lim_{t\to\infty} a(t) \tag{3.22}$$

exists and is the lower solution of the stationary equation

$$a^2 - \sigma a + (s + V_\infty) = 0. \tag{3.23}$$

Furthermore, from the convergence of $V[u(t)]$ and from

$$\partial_t V[u(t)] \leq -\frac{1}{2ph^2} \sum_x (u_{x+h} - u_x)^2 \tag{3.24}$$

follows

$$\sum_x (u_{x+h}(t) - u_x(t))^2 \to 0 \quad \text{for} \quad t \to \infty \tag{3.25}$$

and thus

$$u_x(t) \to a_\infty \quad \text{for all} \quad x. \qquad \bigcirc \tag{3.26}$$

Suppose $(\mathbf{g}^{(n)})_{n=0,1,2,\ldots}$ is the solution of the Euler discretization of (3.2) with the corresponding second moments $(u^{(n)})_{n=0,1,2,\ldots}$. In the spirit of the considerations at the end of section 2.1 we want to derive pointwise thresholds θ_x at which the ensemble at $x \in \Omega_h$ starts to gelate. To be as minute as possible, we would have to solve exactly the system (3.4) with the initial values $u(n\Delta t, x) = u^{(n)}(x)$. This is very expensive. Instead, we derive a more crude method by replacing (3.4) with

$$\partial_t w = -\frac{2}{h^2} w + w^2 - \sigma w + s =: w^2 - \lambda w + s \tag{3.27}$$

(with $\lambda = \sigma + 2/h^2$) which gives a lower solution to the above problem but decouples into local scalar ODE's. Standard ODE techniques yield

3.3 Lemma: Define $\mu := \sqrt{\lambda^2/4 - s}$ and assume $s < \lambda^2/4$ and $w_0 > \lambda/2 + \mu$. Then with $z_0 = (w_0 - \lambda/2 - \mu)^{-1}$ the solution of (3.27) with initial data $w(0) = w_0$ is

$$w(t) = \frac{\lambda}{2} + \mu - \frac{\mu}{1 - (\mu z_0 + 1) \cdot \exp(-\mu t)} \quad . \tag{3.28}$$

In particular, the solution explodes at time

$$t^* = \frac{1}{\mu} \cdot \ln(1 + \mu z_0). \tag{3.29}$$

For w_0 large enough, we have $\mu z_0 \ll 1$ and we get an appropriate threshold θ from the requirement

$$\Delta t = t^* \approx z_0. \tag{3.30}$$

Inserting $w_0 = \theta$ yields

$$\theta = \frac{1}{\Delta t} + \frac{\lambda}{2} + \mu. \tag{3.31}$$

Whenever $u^{(n)}(x)$ reaches this level, then the system $\mathbf{g}^{(n)}(x)$ reaches the gelation point and has to be reduced as described in 2.1.

3.2 Coagulation-diffusion with fluctuations

In contrast to the results above the picture changes as soon as we introduce random fluctuations. The process developed in section 2 serves us as a tool to provide numerical solutions to (3.2) including random perturbations. We will show that *in any case* the corresponding realizations lead to gelation *within finite time*.

We are going to construct a stochastic simulation scheme for the system (3.2) with constants $s, \sigma > 0$ and an initial condition $g_i(0,x) = g_i^{(0)}(x)$. First we choose a fixed time step $\Delta t > 0$ and a sufficiently large integer N. ($1/N$ represents the *particle weight*.) Consider the following random game for a particle ensemble on $\mathbb{N} \times \Omega_h$ which is based on operator splitting of (3.2).

3.4 Random game: (a) *Initializitation:* For each $x \in \Omega_h$ choose an integer random number $N_x^{(0)}$ and an $N_x^{(0)}$-tuple $\mathbf{z}^{(0)}(x) = (z_1^{(0)}(x), \ldots, z_{N_x}^{(0)}(x))$ such that

$$\frac{1}{N} \cdot \sharp\{j | z_j^{(0)}(x) = i\} \approx g_i^{(0)}. \tag{3.32}$$

(b) *Time step:* Denote as $N_x := N_x^{(n)}$ the number of particles at time n in x and by the N_x-tuple $\mathbf{z} := \mathbf{z}^{(n)}(x)$ the collection of all these particles.

(i) coagulation:	from $\mathbf{z}$ construct $\mathbf{z}' \in \mathbb{N}^{N_x}$ as prescribed in Modification 2.3 of the mass flow algorithm.
(ii) sinks and sources:	remove each of the N_x particles independently with probability $\sigma\Delta t$; add to the remaining particles N_s new particles with state 1 (monomers), where N_s is a random variable with finite variance and expectation $\mathbf{E}N_s = s$. Collect the remaining and the new particles in a new tupel $\mathbf{z}''$.
(iii) diffusion:	with probability $2\Delta t/h^2$ move each of the particles of $\mathbf{z}''$ independently either to the left or to the right.

Denote by $(z_1^{(n)}(x), z_2^{(n)}(x), ...)$ the collection of particles in x at time n and define the discrete-time stochastic process on $\mathbb{N} \times \Omega_h$,

$$G^{(n)}(i,x) := \frac{1}{N} \cdot \sharp\{j | z_j^{(n)}(x) = i\}. \tag{3.33}$$

Clearly, this is a Markov process if the above dynamics is ruled by independent random variables (which we assume from now on). For N large enough its realizations are approximations of the time-discretized version of (3.2) which again can be derived in a straightforward manner from the law of large numbers. Gelation is controlled by the reduced process

$$U^{(n)}(x) = \frac{1}{N} \sum_j z_j^{(n)}(x) \tag{3.34}$$

which represents the first moment of $\mathbf{g}^{(n)}(x)$. For the simulation of gelation we use the truncation of $G^{(n)}(x)$ whenever $U^{(n)}(x)$ passes a threshold θ. For the following result we admit *any* positive value θ, not only that given by (3.31). Given θ, we denote by n_x^* the gelation time at x, i.e. the first discrete time when $U^{(n)}(x) \geq \theta$. The following result states that fluctuations a.s. lead to an overall gelation within finite time and thus result in a behaviour qualitatively different from that of the deterministic case.

3.5 Theorem: Given $\theta > 0$, then a.s.

$$\sup_{x \in \Omega_h} n_x^* < \infty \quad . \tag{3.35}$$

Proof: It is easy to prove that there exist constants $p > 0$ and $K \in \mathbb{N}$ such that

$$\text{Prob}\left(U^{(K)}(x) \geq \theta | U^{(0)}(x) < \theta\right) \geq p. \tag{3.36}$$

(The simplest way to see this is: Let no particle diffuse away from x or be absorbed from the sink; furthermore, let at each time step at least $s_0 > 0$ particles be produced from

the source. Then after no more than $[N\theta/s_0]+1$ time steps, the threshold is crossed.) It follows that for all $\ell \in \mathbb{N}$,

$$\begin{aligned} \text{Prob}\left(U^{(n)}(x) < \theta, n = 1, \ldots, K\cdot\ell\right) &\leq \text{Prob}\left(U^{(mK)}(x) < \theta, m = 1.\ldots, \ell\right) \\ &\leq (1-p)^{\ell} \to 0 \quad \text{for} \quad \ell \to \infty. \qquad \bigcirc \end{aligned}$$

3.6 Remark: In the case of small initial condition and source, gelation is generated by large deviations and is an effect to be observed on a *large time scale*. For $N \to \infty$, the gelation time will diverge, and in the limit gelation will not be observed. However, since numerical stochastic schemes and real physical statistical ensembles are represented by *finitely many* particles, the occurrence of gelation is generic. As a consequence, the formerly stable steady states are transformed into *metastable* ones.

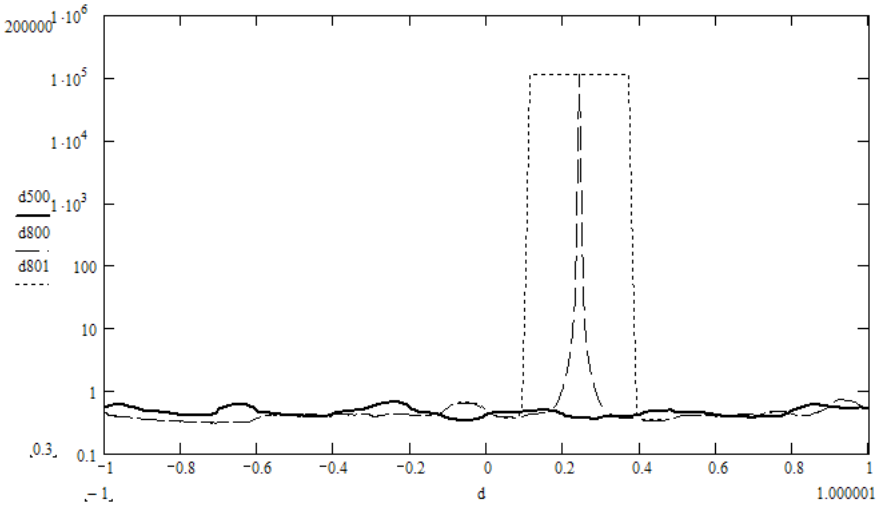

Fig. 3: System with random sources; (a) $t = 50$**, (b)** $t = 75.7$**, (c)** $t = 75.79$

3.3 Numerical illustration

For the numerical example we choose the sink $\sigma = 1$ and the source $s = 0.24$. The corresponding steady solutions are $v_- = 0.4$ (stable) and $v_+ = 0.6$ (unstable). For the

space discretization we choose $p = 100$, i.e. $h = 0.01$.

In the stochastic simulation we choose $N = 256$; the time step is restricted by the choice of h and is taken as $\Delta t = h^2/4 = 2.5 \cdot 10^{-5}$. The initial data are given as slight perturbations of the stable steady solution $v_- = 0.4$. Without fluctuations, the system would converge to the steady state. Fluctuations come into play by the random dynamics and by the choice of the source which is a stochastic perturbation of $s = 0.24$. These source terms are deviced as piecewise constant sources which randomly change after a couple of time steps. More precisely, they are given as

$$S_x^{(n)} = s_{j,k} \quad \text{for} \quad x/h \in -p + 10j + \{1, \dots, 10\}, \quad n \in 3700k + \{0, \dots, 3699\},$$

where the $s_{j,k}$ are independent and identically distributed in $[0, 2s]$.

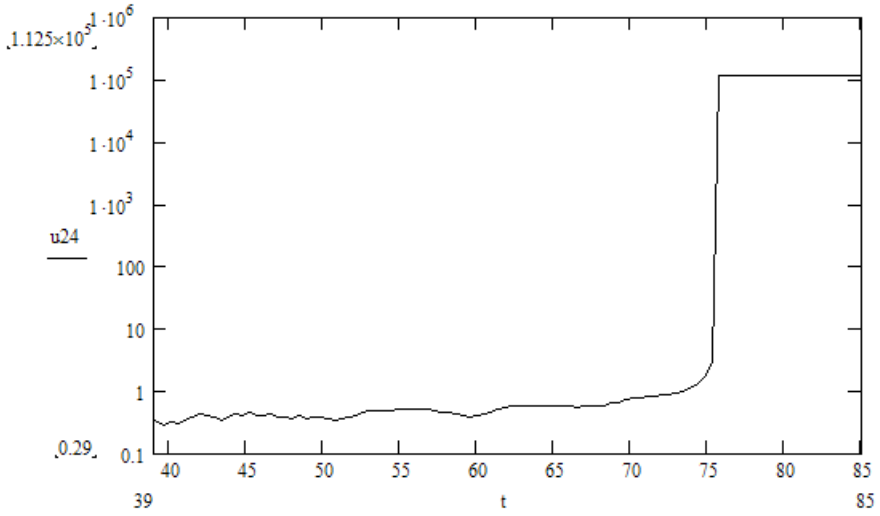

Fig. 4: System with random sources: Evolution of $\|U^{(n)}\|_\infty$

Fig. 3 shows one typical realization of $U^{(n)}$. The thick solid line shows the distribution after a moderate number of time steps ($t \approx 50$); it indicates that the solution floats around the stable solution. The broken line is taken after approximately 3.000.000 time steps (t=75.7); it indicates a strong blow up at one specific point $x_0 \in \Omega_h$. At this time, the local distribution reaches the gelation threshold (here, $\theta = 50000$). Shortly after

this time, the gelation zone quickly spreads over the neighborhood of x_0 (dotted line, $t = 75.79$) and after a short period of time covers all of the domain. Fig. 4 shows the development of $U^{(n)}(x_0)$ around the gelation time. It indicates a sharp phase transition from the metastable state to gelation.

4 Coagulation and the Parabolic Anderson Model

4.1 The model

In this section we study a completely deterministic dynamics in a random environment. Let $\xi : \Omega_h \to [\underline{\xi}, \overline{\xi}]$ be a fixed time-independent random field and consider the system

$$\partial_t \mathbf{g} = \Delta_h \mathbf{g} + \xi \cdot \mathbf{g} + \epsilon \cdot \mathbf{B}[\mathbf{g}, \mathbf{g}] \tag{4.1}$$

with a small parameter $\epsilon > 0$. As before, Δ_h denotes discrete diffusion and $\mathbf{B}[.,.]$ describes the Smoluchowski operator for the first moment system. ξ denotes a linear operator with the negative values representing a random sink while the positive ones give rise to a linear source. The second moment system is given as

$$\partial_t u = \Delta_h u + \xi \cdot u + \epsilon \cdot u^2. \tag{4.2}$$

For the *unbounded* grid $\Omega_h = h \cdot \mathbf{Z}$ and $\epsilon = 0$, equation (4.2) is well-investigated. It is the classical Parabolic Anderson Model (PAM) which has been derived to study conductivity properties in disordered media [2, 8]. For an introduction see [7]. In the following we investigate coagulation as a small perturbation of the PAM on a *finite* grid with periodic boundary conditions as defined in section 3.

In the following denote the linear operator of (4.2) as

$$L = \Delta_h + \xi. \tag{4.3}$$

Since L is symmetric, there exists an orthonormal basis $\mathbf{b}_1, \ldots, \mathbf{b}_{2p}$ of eigenvectors with real eigenvalues

$$\lambda_1 \geq \lambda_2 \geq \cdots \geq \lambda_{2p}. \tag{4.4}$$

Thus the following result is evident.

4.1 Lemma: Solutions of

$$\partial_t u = Lu \tag{4.5}$$

are of the form

$$u(t,x) = \sum_{i=1}^{2p} \alpha_i \exp(\lambda_i t) \cdot \mathbf{b}_i(x). \tag{4.6}$$

Suppose e.g. that ξ_i are independent identically distributed random values in some interval $[a,b]$, $a < b$. Then $\lambda_1 \neq 0$ a.s. Perturbing (4.4) with $\epsilon \cdot u^2$ for ϵ small may either result in a finite gelation time (if $\lambda_1 > 0$) or in exponential decay of the solution ($\lambda_1 < 0$). The precise results are

4.2 Theorem: Consider the system (4.1) with not-vanishing nonnegative initial data $\mathbf{g}(0,x) = \mathbf{g}^{(0)}(x)$. Let the eigenvector expansion of $u^{(0)} = u(0,.)$ be given by

$$u^{(0)} = \sum_{i=1}^{2p} \alpha_i^{(0)} \mathbf{b}_i \tag{4.7}$$

and assume $\alpha_1^{(0)} \neq 0$.

(i) If $\lambda_1 < 0$ then there exist $\epsilon_0, \lambda, C > 0$ such that for all $\epsilon < \epsilon_0$,

$$\|\mathbf{g}(t,.)\|_\infty \leq C \cdot \exp(-\lambda t), \tag{4.8}$$

where the norm is defined as usual as

$$\|\mathbf{g}\|_\infty = \sup\{|g_i(x)|, i \in \mathbb{N}, x \in \Omega_h\}. \tag{4.9}$$

(ii) If $\lambda_1 > 0$ then for all $\epsilon > 0$ the system is gelating at some finite gelation time $t^*(\epsilon)$. There exist constants $A, B > 0$ such that

$$t^*(\epsilon) \leq A - B\left(\ln(\epsilon)\right)^- . \tag{4.10}$$

(For $a \in \mathbb{R}$, we write $a^- = \min\{a, 0\}$.)

Proof: *(i), $\lambda_1 < 0$:* Define r_{ij}^k such that

$$\mathbf{b}_i \cdot \mathbf{b}_j = (b_i(x)b_j(x))_{x\in\Omega_h} = \sum_{k=1}^{2p} r_{ij}^k \mathbf{b}_k. \tag{4.11}$$

The eigenvector expansion of the solution u of (4.2),

$$u(t,.) = \sum_{k=1}^{2p} \alpha_k(t)\mathbf{b}_k \tag{4.12}$$

leads to the system

$$\alpha_k' = \lambda_k\alpha_k + \epsilon \sum_{i,j} r_{ij}^k \alpha_i\alpha_j, \quad \alpha_k(0) = \alpha_k^{(0)}. \tag{4.13}$$

An upper bound is given by

$$\beta' = \lambda_1\beta + \epsilon\rho\beta^2, \quad \beta(0) = \beta^{(0)}, \tag{4.14}$$

where

$$\rho \geq 4p^2 \cdot \max_k \left| \sum_{i,j} r_{ij}^k \right|, \tag{4.15}$$

$$\beta^{(0)} \geq \max_k \left|\alpha_k^{(0)}\right|. \tag{4.16}$$

If ϵ is small enough such that

$$\epsilon\rho\left(\beta^{(0)}\right)^2 \leq \frac{|\lambda_1|}{2}\beta^{(0)} \tag{4.17}$$

then

$$0 < \beta(t) \leq \beta^{(0)} \cdot \exp\left(-\frac{|\lambda_1 t|}{2}\right). \tag{4.18}$$

(ii), $\lambda_1 > 0$: Choose $\mu > 0$ such that

$$\frac{2}{h^2} + |\min \xi_k| \geq \mu. \tag{4.19}$$

Subsolutions for u are on one hand given as the solution v of the linear problem,

$$v(t) = \sum_{k=1}^{2p} \alpha_k \exp(\lambda_k t)\mathbf{b}_k \tag{4.20}$$

and on the other hand as the solution w of

$$\partial_t w = \epsilon w^2 - \mu w; \tag{4.21}$$

since $\mathbf{b}_k$ is defined as an orthonormal system we conclude

$$\sum_{x\in\Omega_h} v^2(t,x) = \sum_{k=1}^{2p} \alpha_k^2 \exp(2\lambda_k t) \tag{4.22}$$

and thus

$$\|u(t,.)\|_1 \geq |\alpha_1| \exp(\lambda_1 t). \tag{4.23}$$

From this follows

$$\|u(t,.)\|_\infty \geq \frac{1}{2p}|\alpha_1| \exp(\lambda_1 t) =: \gamma \exp(\lambda_1 t). \tag{4.24}$$

Define

$$t_0 := \frac{1}{\lambda_1} \cdot \left(\ln \frac{2\mu}{\alpha_1} - \ln(\epsilon) \right) =: A_1 + B \ln(\epsilon). \tag{4.25}$$

Then for $t \geq t_0$ u is bounded from below by

$$\|u(t_0)\|_\infty \geq \frac{2\mu}{\epsilon} \quad \text{and} \quad \partial_t u \geq \frac{\epsilon}{2} u^2, \tag{4.26}$$

proving that $t^*(\epsilon) \leq t_0 + 1/\mu$. $\bigcirc$

4.2 Numerical illustration

We choose $p = 100$; the random numbers $\xi_i = \xi^{(0)} + rd_i \cdot \Delta\xi$ are independent and equidistributed on $[\xi^{(0)} - \Delta\xi, \xi^{(0)} + \Delta\xi]$. For $\xi^{(0)} + \Delta\xi < 0$, L has a purely negative spectrum with a.s. different eigenvalues. The eigenvector corresponding to the leading eigenvalue is shown in Fig. 5. Increasing $\xi^{(0)}$, L gets a mixed and later on a purely positive spectrum. Once there is a positive eigenvalue, introduction of coagulation leads to gelation within finite time. Fig. 6. shows $\|U^{(n)}\|_\infty$ in the case of small ϵ. Here, the perturbed system follows for some time the contours of the linear system and then explodes. Figs. 7.a),b),c) show $U^{(n)}$ for $n = 0$ and for two values of n shortly before

gelation. In these plots, the curves are rescaled such that $\|U^{(n)}\|_\infty = 1$. We realize that the system first approaches the leading eigenmode and afterwards this eigenmode becomes the nucleus for gelation.

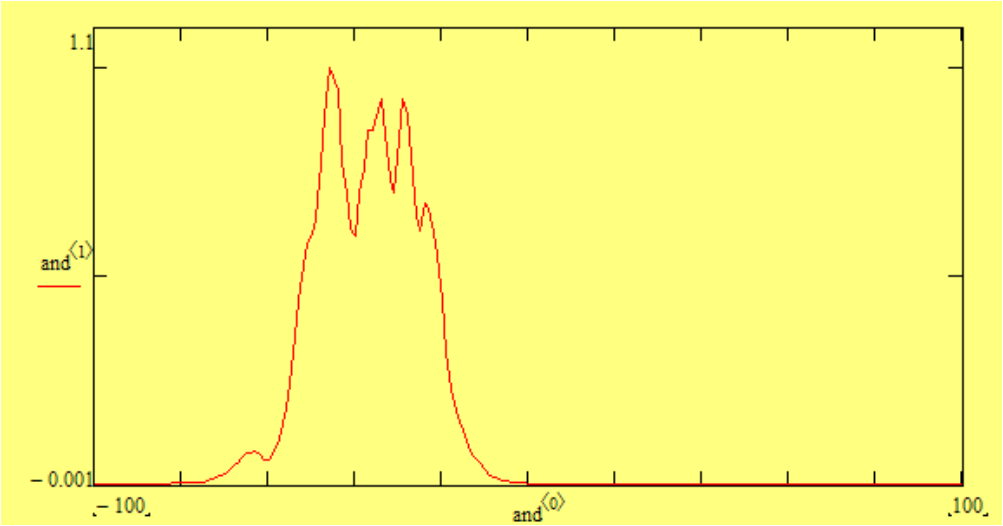

Fig. 5: Leading eigenmode of PAM

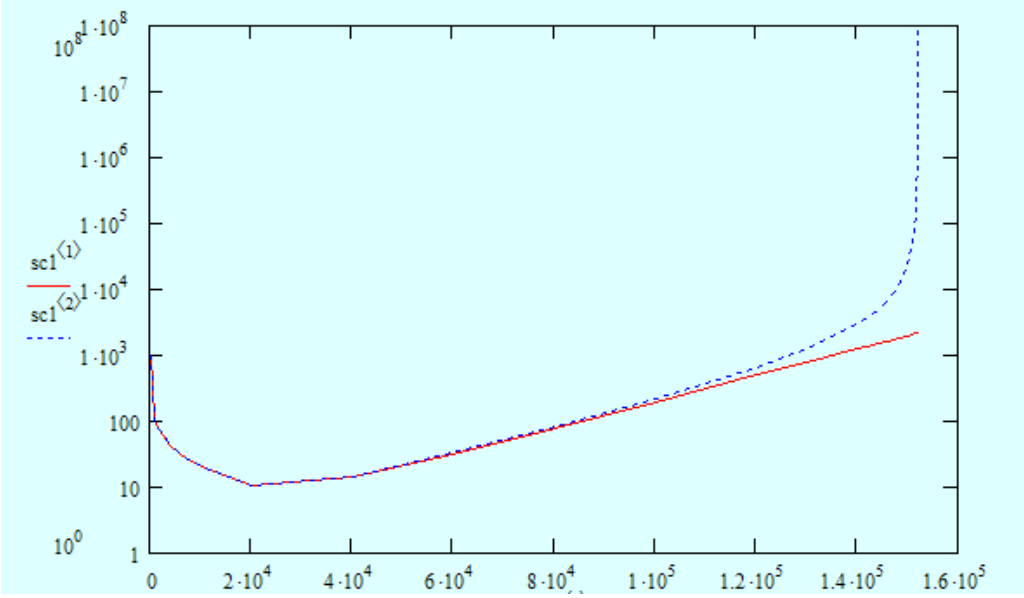

Fig.6: PAM, perturbed by coagulation

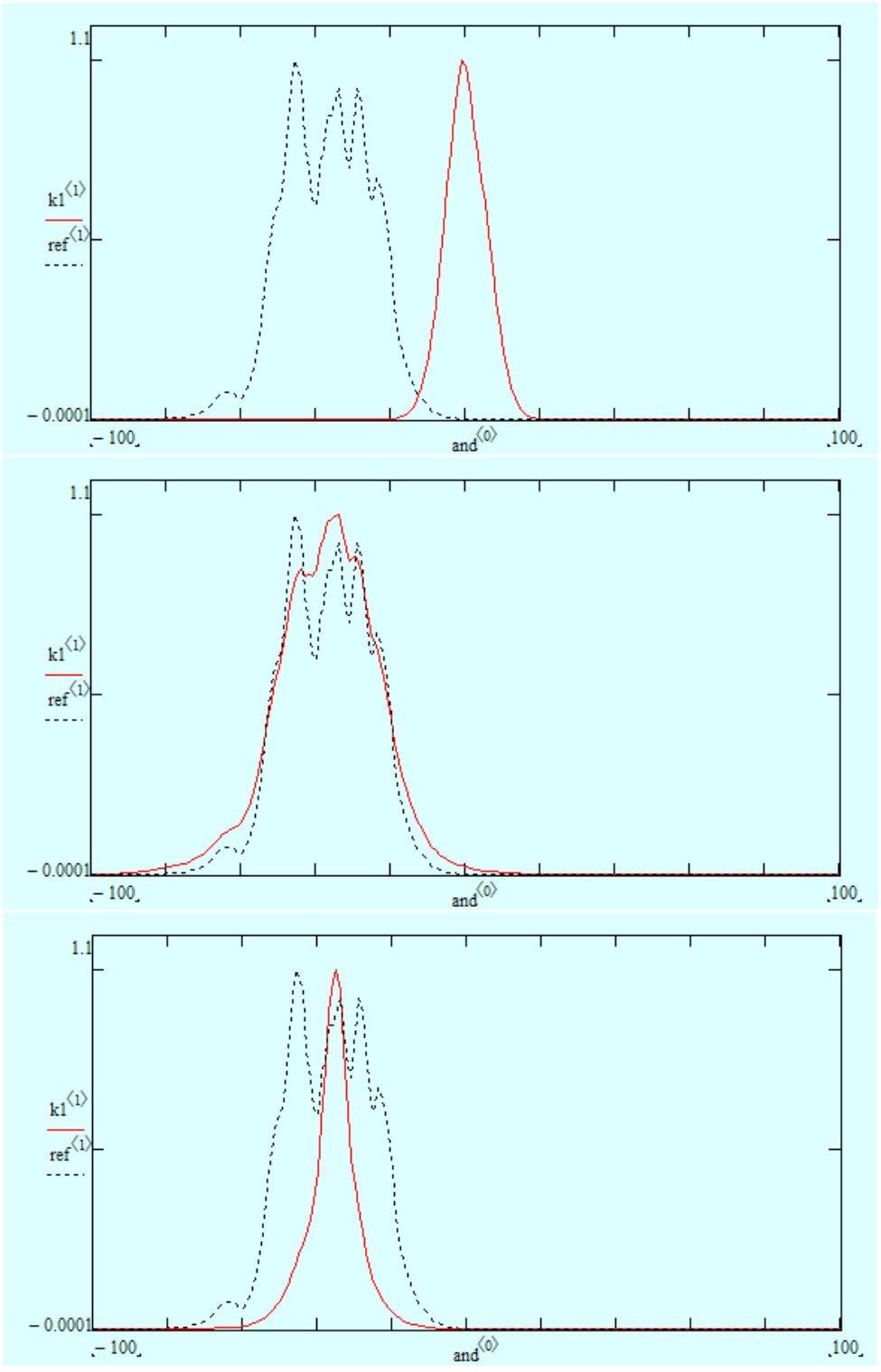

Fig. 7: Approaching the gelation phase

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