

Convergence of the Marcus-Lushnikov process

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Abstract

The Smoluchowski coagulation equation describes the concentration $c(t, x)$ of particles of mass $x \in]0, \infty[$ at the instant $t \geq 0$, in an infinite system of coalescing particles. It is well-known that in some cases, *gelation* occurs: a particle with infinite mass appears. But this infinite particle is inert, in the sense that it does not interact with finite particles.

We consider the so-called Marcus-Lushnikov process, which is a stochastic finite system of coalescing particles. This process is expected to converge, as the number of particles tends to infinity, to a solution of the Smoluchowski coagulation equation. We show that it actually converges, for $t \in [0, \infty[$, to a modified Smoluchowski equation, which takes into account a possible interaction between finite and infinite particles.

Key words : Marcus-Lushnikov process, Smoluchowski coagulation equations, gelation.

MSC 2000 : 45K05, 60H30.

1 Introduction

The Marcus-Lushnikov process [12], [11] describes the stochastic Markov evolution of a finite system of coalescing particles. The (deterministic) Smoluchowski coagulation equation describes the evolution of the concentration $c(t, x)$ of particles of mass $x \in]0, \infty[$ at the instant $t \geq 0$, in an infinite system of coalescing particles. Both models depend on a “coagulation kernel” $K(x, y)$ which stands for the rate of coalescence between particles of masses x and y .

We are interested in the convergence of the Marcus-Lushnikov process to the Smoluchowski equation as the number of particles tends to infinity (Open Problem 10 of Aldous, [1]). First, this problem is interesting from the numerical point of view. Indeed, the Marcus-Lushnikov process can be simulated exactly. One thus would like to use it in order to approximate the solution of the Smoluchowski equation. Next, it has a physical issue: the Smoluchowski equation is

often derived by passing to the limit in the Marcus-Lushnikov process. Rigorous justifications seem to be needed.

When $K(x, y) \geq c(x^\alpha y^\beta + x^\beta y^\alpha)$, with $c > 0$, $\alpha, \beta \in [0, 1]$, it is proved in Escobedo-Mishler-Perthame [7] that if $\alpha + \beta > 1$, then gelation occurs in the Smoluchowski equation: there exists an instant $T_{gel} < \infty$ such that the mass consisting in infinite particles becomes positive.

Extending some results of Jeon [9], Norris shows in [13] the convergence of the Marcus-Lushnikov process to the Smoluchowski equation when (essentially) $K(x, y)/y$ tends to 0 as y tends to infinity, for any fixed x .

In [14], he shows the convergence of the Marcus-Lushnikov process to a modified Smoluchowski equation when (essentially) $K(x, y) = xy$ as soon as $x > A$ and $y > A$ for some constant A . The “modified” Smoluchowski equation takes into account a possible interaction between finite and infinite particles, after the gelation time. This modified Smoluchowski equation had already been introduced by Flory [8], see also Ernst-Ziff-Hendriks [5] (when $K(x, y) = xy$), Escobedo-Laurençot-Mishler [6] (when $K(x, y) = (xy^\beta + x^\beta y)$, for some $\beta \in]0, 1[$).

The aim of the present paper is to extend the results of Norris to (essentially) any coagulation kernel satisfying: for all $x > 0$, $\lim_{y \rightarrow \infty} K(x, y)/y = l(x)$ exists and is finite.

The quantity $l(x)$ will quantify the disappearance rate of particles of mass x because of coalescence with the infinite particle.

Our result will in particular cover the case where $K(x, y) = (xy^\beta + x^\beta y)$, for some $\beta \in [0, 1]$, which was recently studied in [6], and the case where $K(x, y) = (x + c)(y + c)$, for some $c > 0$, which is known as a Condensation/Branched Chain Polymerisation kernel, see Aldous [1].

The paper is organized as follows: in Section 2, we state our main result, while the proof is handled in Section 3. We finally present numerical simulations in Section 4.

Notation 1.1 A measurable map $K :]0, \infty[\times]0, \infty[\mapsto \mathbb{R}_+$ is called a “coagulation kernel” as soon as it is symmetric ($K(x, y) = K(y, x)$).

For a measurable space E , we denote by $M_f^+(E)$ the set of nonnegative finite measures on E , and by $\mathcal{P}(E)$ the set of probability measures on E .

For a measure ν and a function f , we set $\langle \nu, f \rangle = \langle \nu(dx), f(x) \rangle = \int f(x)\nu(dx)$.

For an interval I of $[0, \infty[$, we denote by $C_b(I)$, (resp. $C_b^1(I)$) the set of bounded continuous functions (resp. of class C^1 , bounded with their derivative); and by $C_c(I)$ (resp. $C_c^1(I)$) the set of continuous (resp. of class C^1) functions on I with compact support (in I).

For an interval I of $[0, \infty[$, a sequence ν_n of finite measures on I is said to converge weakly (resp. vaguely) to ν if for all functions $f \in C_b(I)$ (resp. $f \in C_c(I)$), $\lim_n \langle \nu_n, f \rangle = \langle \nu, f \rangle$.

2 Main result

In this section, we recall the dynamics of the Marcus-Lushnikov process, and then we define a modified Smoluchowski equation. We finally state and comment on our main result.

The Marcus-Lushnikov process

We consider a coagulation kernel K (see Notation 1.1) and a finite particle system initially consisting in $n \geq 2$ particles, of masses $x_1 > 0, \dots, x_n > 0$. We denote by $m_n = x_1 + \dots + x_n$ the total mass of the system. Then we assume that the system evolves according to the following dynamics: each pair of particles (of masses x and y) may coalesce (i.e. disappear and form a new particle of mass $x + y$) with an exponential rate $K(x, y)/m_n$. The Marcus-Lushnikov process describes the evolution of the empirical concentration $\mu_t^n = m_n^{-1} \sum_{i=1}^{n(t)} \delta_{X_t^i}$, where $n(t)$ stands for the number of particles at the instant t , and $X_t^1, \dots, X_t^{n(t)}$ denote their masses. This simply means that for any $t > 0$, any $z > 0$, $\mu_t^n(\{z\})$ is the concentration (number per unit of mass) of particles of mass z at instant t in the system. We now define it rigorously.

Definition 2.1 Consider a coagulation kernel K , and an initial state $\mu_0^n = m_n^{-1} \sum_{i=1}^n \delta_{x_i}$, with $x_1 > 0, \dots, x_n > 0$, and $m_n = x_1 + \dots + x_n$.

The Marcus-Lushnikov process $(\mu_t^n)_{t \geq 0}$ associated with the pair (K, μ_0^n) is a Markov $M_f^+(\mathbb{R}, \mathcal{B}(\mathbb{R}))$ -valued càdlàg process satisfying:

- (i) $(\mu_t^n)_{t \geq 0}$ takes its values in $\{m_n^{-1} \sum_{i=1}^k \delta_{y_i}, k \leq n, y_i > 0\}$.
- ii) its generator is given, for all measurable functions ψ from $M_f^+(\mathbb{R}, \mathcal{B}(\mathbb{R}))$ into \mathbb{R} , all states $\mu = m_n^{-1} \sum_{i=1}^k \delta_{y_i}$, by

$$L^{K, \mu_0^n} \psi(\mu) = \sum_{i \neq j} \left\{ \psi \left[\mu + m_n^{-1} (\delta_{y_i + y_j} - \delta_{y_i} - \delta_{y_j}) \right] - \psi[\mu] \right\} \frac{K(y_i, y_j)}{2m_n} \quad (2.1)$$

This process is well-known to be well-defined and unique, see e.g. Norris [13]. This is anyway immediate, since conditionally to μ_0^n , the process $(\mu_t^n)_{t \geq 0}$ is a continuous-time Markov chain with finite state space.

Furthermore, it clearly satisfies that for any $t \geq 0$, $\langle \mu_t^n(dx), x \rangle = 1$ a.s.

A modified Smoluchowski equation

We now consider an infinite particle system corresponding to the previously described finite particle system. We give our assumptions following the ideas of Norris, [13] which are particularly well-adapted to the study of coagulation phenomena.

Let us recall that a map $\phi :]0, \infty[\rightarrow]0, \infty[$ is *subadditive* if for all $x > 0, y > 0$, $\phi(x + y) \leq \phi(x) + \phi(y)$. For example, any function $\phi(x) = x^{-\alpha} + x^\beta$, for $\beta \leq 1$ and $\alpha \geq 0$ is subadditive.

Assumptions (L): The coagulation kernel K is a symmetric nonnegative map on $]0, \infty[^2$. There exists a continuous subadditive function

ϕ on $]0, \infty[$ such that for all $x > 0$, all $y > 0$,

$$\phi(x) \geq 1, \quad K(x, y) \leq \phi(x)\phi(y), \quad \text{and } l(x) = \lim_{y \rightarrow \infty} K(x, y)/y \quad (2.2)$$

exists and is finite. Finally, the map $C(x, y)$, defined for $x > 0$ and $y \geq 0$ by

$$C(x, y) = yK(1/x, 1/y) \text{ if } y > 0 \quad \text{and} \quad C(x, 0) = l(1/x) \quad (2.3)$$

is continuous on $]0, \infty[\times]0, \infty[$.

We note that under (L), there exists a constant A such that for all $x > 0$,

$$1 + l(x) \leq A\phi(x) \quad (2.4)$$

Definition 2.2 Assume (L). Let $\mu_0 \in M_f^+(]0, \infty[)$ satisfy $\langle \mu_0(dx), x \rangle = 1$ and $\langle \mu_0, \phi \rangle < \infty$. A family $(\mu_t)_{t \geq 0}$ of measures of $M_f^+(]0, \infty[)$ is said to solve the modified Smoluchowski equation (MS) if

- (i) for all $T < \infty$, $\sup_{[0, T]} \langle \mu_t(dx), \phi(x) \rangle < \infty$ and $\sup_{[0, T]} \langle \mu_t(dx), x \rangle \leq 1$.
- (ii) for all test functions $g \in C_c^1([0, \infty[)$, all $t \geq 0$,

$$\begin{aligned} \langle \mu_t, g \rangle = \langle \mu_0, g \rangle + \frac{1}{2} \int_0^t \langle \mu_s(dx) \otimes \mu_s(dy), [g(x+y) - g(x) - g(y)] K(x, y) \rangle ds \\ - \int_0^t \langle \mu_s(dx), g(x)l(x) \rangle [1 - \langle \mu_s(dx), x \rangle] ds \end{aligned} \quad (2.5)$$

We note that under (L), condition (i), (2.4), and the fact that g is bounded imply that every term in (2.5) is well-defined.

Equation (MS) describes the evolution of the concentration measure $\mu_t(dx)$ of particles of mass x in an infinite system of particles in which particles of mass x and y coagulate with rate $K(x, y)$, and in which the mass consisting in infinite particles (given by $1 - \langle \mu_s(dx), x \rangle$) interacts with finite particles of mass x , by “absorbing” them with rate $l(x)$.

We remark that if μ_0 is discrete (i.e. $\mu_0(\mathbb{N}^*) = 1$), then this corresponds to the usually called “discrete coagulation equations”, while when μ_0 is continuous (i.e. when $\mu_0(dx) = c_0(x)dx$), this corresponds to the so-called “continuous coagulation equations”.

We point out that the Smoluchowski equation is the same as (2.5) without the last term. The examples below are taken from Escobedo et al. [7]. For non-gelling kernels (e.g. $K(x, y) \leq A(1 + x + y)$) or gelling kernels such that $l \equiv 0$ (e.g. $K(x, y) = x^\alpha y^\beta + y^\alpha x^\beta$, with $\alpha + \beta > 1$, $\alpha, \beta \in [0, 1[$), the two equations are equivalent for all times. For gelling kernels such that $l(x) > 0$ (e.g. $K(x, y) = xy^\alpha + yx^\alpha$, with $\alpha \in]0, 1[$), the two equations are equivalent for $t \in [0, T_{gel}]$, but differ on $]T_{gel}, \infty[$.

The convergence result

We can now state our result. For definitions of tightness and Skorokhod spaces, we refer to Ethier-Kurtz [4].

Theorem 2.3 Consider a coagulation kernel K satisfying assumptions (L), and an initial condition $\mu_0 \in M_f^+(]0, \infty[)$ satisfying

$$\langle \mu_0(dx), x \rangle = 1 \quad ; \quad \langle \mu_0(dx), \phi(x) \rangle < \infty \quad (2.6)$$

Consider, for each $n \geq 2$, a Marcus-Lushnikov process $(\mu_t^n)_{t \geq 0}$ associated with K and with an initial condition $\mu_0^n = m_n^{-1} \sum_{i=1}^n \delta_{x_i}$. Assume that

$$\begin{aligned} \sup_{n \geq 2} \langle \mu_0^n, \phi \rangle < \infty \quad ; \quad \lim_{\varepsilon \rightarrow 0} \sup_{n \geq 2} \langle \mu_0^n, \phi \mathbb{1}_{]0, \varepsilon[} \rangle = 0 \\ \forall f \in C_c(]0, \infty[), \lim_n \langle \mu_0^n, f \rangle = \langle \mu_0, f \rangle \end{aligned} \quad (2.7)$$

(i) Then the sequence of processes $(\mu_t^n)_{t \geq 0}$ is tight in $\mathbb{D}(]0, \infty[, M_f^+(]0, \infty[))$. The space $\mathbb{D}(]0, \infty[, M_f^+(]0, \infty[))$ is endowed with the Skorokhod topology associated with the vague topology on $M_f^+(]0, \infty[)$.

(ii) Consider a limit point (as n tends to infinity) $(\mu_t)_{t \geq 0}$ of a subsequence of $(\mu_t^n)_{t \geq 0}$. Then $(\mu_t)_{t \geq 0}$ satisfies a.s. equation (MS).

While the first and third conditions in (2.7) are fundamental, the second one is probably technical. We need it to show point (ii) of the theorem, since it ensures uniform integrability near 0 of the sequence $\{\mu_t^n\}_n$ for any $t \geq 0$. We believe this technical condition is not restrictive for possible applications.

If one knows about uniqueness for equation (MS) (see Norris, [14] for a specific almost multiplicative case), then one deduces that the sequence $(\mu_t^n)_{t \geq 0}$ converges in probability to the unique solution $(\mu_t)_{t \geq 0}$ to equation (MS). We point out that Norris, extending the results of Jeon [9], proves Theorem 2.3 when $l \equiv 0$ in [13], and when $K(x, y) = xy$ as soon as $x > A, y > A$ for some constant A , in [14].

Note that two cases remain open: what happens when $l(x)$ does not exist or is infinite. When $l(x)$ is infinite, a conjecture (see Aldous [1]) says that $T_{gel} = 0$. In equation (MS), it seems clear that if so, a solution $(\mu_t)_{t \geq 0}$ will satisfy $\mu_t = 0$ for all $t > 0$. Hence one might conjecture that the Marcus Lushnikov process converges, as n tends to infinity, to the trivial measure 0.

As a final remark, we note that Theorem 2.3 might suggest that the relevant equation after gelation is (MS) and not the Smoluchowski equation. However, Laurençot [10], obtains (in particular) global existence to the Smoluchowski equation with multiplicative kernel $K(x, y) = xy$ by using the approximating non-gelling kernels $K^n(x, y) = xy \mathbb{1}_{\{x \leq n, y \leq n\}}$. This result thus suggests that the Smoluchowski equation still has some sense after gelation, even when $l(x) > 0$.

3 Proof

We now prove Theorem 2.3. First, we will introduce some notations, transfer the gelation problem from infinity to zero, and recall some martingale properties of the Marcus-Lushnikov process. Next, we will explain the tightness result, and finally, the convergence result.

We begin by a classical property of the Marcus Lushnikov processes.

Lemma 3.1 Consider a Marcus-Lushnikov process $\{\mu_t^n\}$, and a subadditive function f on $[0, \infty[$. Then a.s., $\langle \mu_t^n, f \rangle$ is non increasing.

The proof is immediate, since the jumps of $\langle \mu_t^n, f \rangle$ have the form $m_n^{-1}[f(x+y) - f(x) - f(y)] \leq 0$.

Now, we substitute the Marcus-Lushnikov process $(\mu_t^n)_{t \geq 0}$ in order to obtain a new process $(R_t^n)_{t \geq 0}$ which has the following advantages: (a) it is a probability measure on $[0, \infty[$, (b) the gelation problem, which concerns the possible limits of μ_t^n by their values at infinity, will concern the possible limits of R_t^n by their values at 0.

Notation 3.2 Assume (L), (2.6) and (2.7). For each $n \geq 2$, consider the Marcus-Lushnikov process $\mu_t^n = m_n^{-1} \sum_{i=1}^{n(t)} \delta_{X_i}$. We define $R_t^n = m_n^{-1} \sum_{i=1}^{n(t)} X_i \delta_{1/X_i}$. Then $(R_t^n)_{t \geq 0}$ is a càdlàg process with values in $\mathcal{P}([0, \infty[)$. Note that for all functions f on $]0, \infty[$, all $t \geq 0$, $\langle R_t^n, f \rangle = \langle \mu_t^n(dx), xf(1/x) \rangle$ and $\langle \mu_t^n, f \rangle = \langle R_t^n(dx), xf(1/x) \rangle$.

We now introduce an integral operator.

Notation 3.3 Assume (L), and recall the definition of map C (Assumption (L)). We define the operator L , for any measurable function f on $[0, \infty[$, any $x \geq 0$, $y \geq 0$, by

$$Lf(x, y) = \mathbb{1}_{\{x>0\}} \left[f\left(x - \frac{x^2}{x+y}\right) - f(x) \right] C(x, y) \quad (3.8)$$

We then consider the following deterministic equation, which is obtained from (MS) by replacing $f(x)$ by $xf(1/x)$.

Definition 3.4 Assume (L) and (2.6). Consider the probability measure R_0 on $[0, \infty[$ defined by $\langle R_0, f \rangle = \langle \mu_0(dx), xf(1/x) \rangle$. A family of probability measures $(R_t)_{t \geq 0}$ on $[0, \infty[$ is said to solve equation (IM) if
(a) for all $T < \infty$, $\sup_{[0, T]} \langle R_s(dx), x\phi(1/x)\mathbb{1}_{\{x>0\}} \rangle < \infty$,
(b) for all functions $f \in C_c^1(]0, \infty[)$, all $t \geq 0$,

$$\langle R_t, f \rangle = \langle R_0, f \rangle + \int_0^t \langle R_s(dx) \otimes R_s(dy), Lf(x, y) \rangle ds \quad (3.9)$$

Note that every term in (IM) is well-defined thanks to the fact that f is $C_c^1(]0, \infty[)$, assumptions (L), (2.4), and condition (a).

In the next lemma, we show that a solution to (IM) leads to a solution to (MS).

Lemma 3.5 Assume (L) and (2.6). Consider a solution $(R_t)_{t \geq 0}$ to (IM). Define, for each $t > 0$, the measure μ_t on $]0, \infty[$ by: for all bounded functions f on $]0, \infty[$, $\langle \mu_t, f \rangle = \langle R_t(dx), xf(1/x)\mathbb{1}_{\{x>0\}} \rangle$. Then the family $(\mu_t)_{t \geq 0}$ satisfies equation (MS) with initial condition μ_0 and coagulation kernel K .

Proof First, note that for each t , $\langle \mu_t, \phi \rangle = \langle R_t(dx), x\phi(1/x)\mathbb{1}_{\{x>0\}} \rangle$. We deduce from (a) that that $(\mu_t)_{t \geq 0}$ is a family of finite measures and that for all $T < \infty$, $\sup_{[0, T]} \langle \mu_t, \phi \rangle < \infty$. Furthermore, since R_t is a probability measure for each t , we deduce that $\langle \mu_t(dx), x \rangle = \langle R_t(dx), \mathbb{1}_{\{x>0\}} \rangle \leq 1$. Thus conditions (i) of Definition 2.2 holds.

Next, note that for each $t \geq 0$, $R_t(\{0\}) = 1 - \langle R_t(dx), \mathbb{1}_{\{x>0\}} \rangle = 1 - \langle \mu_t(dx), x \rangle$. Consider now a function $g \in C_c^1([0, \infty[)$. A computation, using (3.9) with $f(x) = xg(1/x)$ (which belongs to $C_c^1([0, \infty[)$), splitting $\langle R_s \otimes R_s, Lf \rangle$ into $\langle R_s(dx) \otimes R_s(dy), Lf(x, y)\mathbb{1}_{\{y>0\}} \rangle$ and $\langle R_s(dx), Lf(x, 0) \rangle R_t(\{0\})$ leads to (2.5). We still have to extend (2.5) to any function $g \in C_c^1([0, \infty[)$. We consider a sequence g_n of $C_c^1([0, \infty[)$ -functions such that $\sup_n \sup_x |g_n(x)| \leq A$, and such that $g_n(x) = g(x)$ for all $x \geq 1/n$. Then (2.5) holds for each g_n , and one can make n tend to infinity, using the dominated convergence Theorem, thanks to the fact that $\langle \mu_t(dx), (1 + x + \phi(x) + l(x)) \rangle < \infty$. \square

In view of the previous notations and results, we will in fact only have to check that the following two propositions hold.

Proposition 3.6 *Assume (L), (2.6) and (2.7). Then the family of stochastic processes $(R_t^n)_{t \geq 0}$ is tight in $\mathbb{D}([0, \infty[, \mathcal{P}([0, \infty[))$. Here $\mathbb{D}([0, \infty[, \mathcal{P}([0, \infty[))$ is endowed with the Skorokhod topology associated with the weak convergence on $\mathcal{P}([0, \infty[)$.*

Note that such a weak (and not vague) tightness result can not hold for $\{\mu^n\}$, since we expect that $\lim_n \langle \mu_t^n, 1 \rangle \neq \langle \mu_t, 1 \rangle$, for $t > T_{gel}$.

Proposition 3.7 *Assume (L), (2.6) and (2.7). Consider a weak limit $(R_t)_{t \geq 0}$ of $(R_t^n)_{t \geq 0}$. Then $(R_t)_{t \geq 0}$ satisfies almost surely (IM).*

To prove these results, we will use the following martingale properties.

Lemma 3.8 *For each $n \geq 2$, consider the process $(R_t^n)_{t \geq 0}$ defined in Notation 3.2. Then for all functions $f \in C_c([0, \infty[)$, the process*

$$\begin{aligned} O_t^{n,f} &= \langle R_t^n, f \rangle - \langle R_0^n, f \rangle - \int_0^t \langle R_s^n \otimes R_s^n, Lf \rangle ds \\ &+ \frac{1}{m_n} \int_0^t \langle R_s^n(dx), [f(x/2) - f(x)] K(1/x, 1/x) \rangle ds \end{aligned} \quad (3.10)$$

is a martingale with (predictable) quadratic variation

$$\begin{aligned} \langle O^{n,f} \rangle_t &= \frac{1}{2m_n} \int_0^t \left\langle R_s^n(dx) \otimes R_s^n(dy), \left[(x+y)f\left(\frac{xy}{x+y}\right) \right. \right. \\ &\quad \left. \left. - xf(y) - yf(x) \right]^2 \frac{K(1/x, 1/y)}{xy} \right\rangle ds \\ &- \frac{1}{2m_n^2} \int_0^t \left\langle R_s^n(dx), [2f(x/2) - 2f(x)]^2 \frac{K(1/x, 1/x)}{x} \right\rangle ds \end{aligned} \quad (3.11)$$

Proof Noting that $\langle R_t^n, f \rangle = \psi(\mu_t^n)$, with $\psi(\mu) = \langle \mu(dx), xf(1/x) \rangle$, we obtain, using Definition 2.1, that

$$\psi(\mu_t^n) - \psi(\mu_0^n) - \int_0^t L^{K, \mu_0^n} \psi(\mu_s^n) ds \quad (3.12)$$

is a martingale. Rewriting this formula in terms of R_t^n , using (2.1), leads to (3.10). To compute the bracket of $O^{n,f}$, we use the fact that

$$\psi^2(\mu_t^n) - \psi^2(\mu_0^n) - \int_0^t L^{K, \mu_0^n} [\psi^2](\mu_s^n) ds \quad (3.13)$$

is a martingale. Rewriting this expression in terms of R_t^n , and comparing the obtained formula to (3.10) leads to (3.11). \square

We now can give the

Proof of Proposition 3.6 First of all, we introduce a distance on $\mathcal{P}([0, \infty[)$. It is easy to check that there exists a sequence of $C_b^1([0, \infty[)$ functions $(g_p)_{p \geq 1}$ satisfying $\|g_p\|_\infty + \|g'_p\|_\infty \leq 1$ such that the distance d defined by

$$P, Q \in \mathcal{P}([0, \infty[), \quad d(P, Q) = \sum_{p \geq 1} 2^{-p} |\langle g_p, P \rangle - \langle g_p, Q \rangle| \quad (3.14)$$

gives rise to the topology of weak convergence on $\mathcal{P}([0, \infty[)$. Indeed, one may set $\hat{f}_{a,b,c} = \mathbb{1}_{[a,b]} \star G_c$, for $a < b$ in \mathbb{Q}_+ and c in $\mathbb{Q}_+/\{0\}$. Here G_c stands for the density of the Gaussian law with mean 0 and variance c , while \star stands for the convolution product. Of course, $\hat{f}_{a,b,c}$ is C_b^1 for all a, b, c . Then, one may set $f_{a,b,c} = \hat{f}_{a,b,c} / (\|\hat{f}_{a,b,c}\|_\infty + \|\hat{f}'_{a,b,c}\|_\infty)$, and consider a sequence $\{g_p\}_{p \geq 1}$ such that $\{g_p\}_{p \geq 1} = \{f_{a,b,c}\}_{a < b \in \mathbb{Q}_+, c \in \mathbb{Q}_+/\{0\}}$. See Billingsley [2] for the proof of similar results.

It now suffices to check that $\{R^n\}_{n \geq 2}$ satisfies the Aldous criterion for tightness, see Ethier-Kurtz [4], Theorem 8.6 p 137. It suffices to check the two following points:

- (i) There exists a compact subset Γ of $\mathcal{P}([0, +\infty[)$ such that for all $t \geq 0$, all $n \geq 2$, $R_t^n \in \Gamma$ a.s.
- (ii) For all $T < \infty$, if $\mathcal{A}(T)$ stands for the set of stopping times bounded by T ,

$$\lim_{\delta \rightarrow 0} \sup_{n \geq 2} \sup_{S \in \mathcal{A}(T)} \sup_{0 \leq u \leq \delta} \mathbb{E}(d(R_S^n, R_{S+u}^n)) = 0. \quad (3.15)$$

Point (i) is immediate. Indeed, thanks to condition (2.7), $A = \sup_n \langle \mu_0^n, 1 \rangle < \infty$. Hence we deduce, denoting by $\mu_t^n = m_n^{-1} \sum_{i=1}^{n(t)} \delta_{X_t^i}$, recalling that $n(t)$ is non-increasing (and thus smaller than n for all t), and using Notation 3.2, that for any $a < \infty$, any $t \geq 0$, any $n \geq 2$, a.s.,

$$R_t^n([a, \infty[) = m_n^{-1} \sum_{i=1}^{n(t)} X_t^i \mathbb{1}_{\{1/X_t^i \geq a\}} \leq m_n^{-1} \sum_{i=1}^{n(t)} a^{-1} \leq a^{-1} \frac{n}{m_n} \quad (3.16)$$

But $nm_n^{-1} = \langle \mu_0^n, 1 \rangle$ and thus is smaller than A . Hence, for all t , all n , R_t^n belongs a.s. to

$$\Gamma = \{r \in \mathcal{P}([0, \infty[); \forall a, r([a, \infty[) \leq a^{-1}A\} \quad (3.17)$$

which is a compact subset of $\mathcal{P}([0, \infty[)$ for the weak topology.

We now check point (ii). We fix $T > 0$ and $0 < u < \delta$, and we consider a stopping time $S \in \mathcal{A}(T)$. Then we use Lemma 3.8, and (3.14) to obtain

$$\mathbb{E}(d(R_S^n, R_{S+u}^n)) \leq \sum_{p \geq 1} 2^{-p} [A_p^1 + A_p^2 + A_p^3] \quad (3.18)$$

where

$$\begin{aligned} A_p^1 &= \mathbb{E} \left[\int_S^{S+\delta} |\langle R_s^n(dx) \otimes R_s^n(dy), \left[g_p \left(x - \frac{x^2}{x+y} \right) - g_p(x) \right. \right. \\ &\quad \left. \left. yK(1/x, 1/y) \right\rangle ds \right] \\ A_p^2 &= \mathbb{E} \left[\frac{1}{m_n} \int_S^{S+\delta} |\langle R_s^n, (g_p(x/2) - g_p(x))K(1/x, 1/x) \rangle ds \right] \\ A_p^3 &= (\mathbb{E} [|\langle O^{n, g_p} \rangle_{S+\delta} - \langle O^{n, g_p} \rangle_S|])^{\frac{1}{2}} \end{aligned} \quad (3.19)$$

Using (L), the fact that g'_p is bounded uniformly by 1, the inequality $x^2/(x+y) \leq x$, and then the relation between R_t^n and μ_t^n , we obtain that

$$A_p^1 \leq \mathbb{E} \left[\int_S^{S+\delta} |\langle R_s^n \otimes R_s^n, xy\phi(1/x)\phi(1/y) \rangle ds \right] = \mathbb{E} \left[\int_S^{S+\delta} \langle \mu_s^n, \phi \rangle^2 ds \right] \quad (3.20)$$

But ϕ is subadditive, thus by Lemma 3.1, we deduce using (2.7) that for some constant A , $A_p^1 \leq \delta \langle \mu_0^n, \phi \rangle^2 \leq A\delta$.

Next, we obtain with similar arguments

$$A_p^2 \leq \frac{1}{m_n} \mathbb{E} \left[\int_S^{S+\delta} \langle R_s^n(dx), x\phi^2(1/x) \rangle ds \right] = \frac{1}{m_n} \mathbb{E} \left[\int_S^{S+\delta} \langle \mu_s^n, \phi^2 \rangle ds \right] \quad (3.21)$$

But $m_n^{-1} \langle \mu_s^n, \phi^2 \rangle = m_n^{-2} \sum_{i=1}^{n(t)} \phi^2(X_t^i) \leq \left[m_n^{-1} \sum_{i=1}^{n(t)} \phi(X_t^i) \right]^2 = \langle \mu_s^n, \phi \rangle^2 \leq \langle \mu_0^n, \phi \rangle^2$. Thus we also obtain $A_p^2 \leq A\delta$.

Writting $\frac{xy}{x+y}$ as $x - \frac{x^2}{x+y}$ or as $y - \frac{y^2}{x+y}$, and using the fact that g'_p is smaller than 1, we obtain:

$$\left[(x+y)g_p \left(\frac{xy}{x+y} \right) - xg_p(y) - yg_p(x) \right]^2 \leq 4(xy)^2 \quad (3.22)$$

By (3.22), (3.11), and assumption (L), we deduce:

$$\begin{aligned}
(A_p^3)^2 &\leq \frac{2}{m_n} \mathbb{E} \left[\int_S^{S+\delta} \langle R_s^n(dx), x\phi(1/x) \rangle^2 ds \right] \\
&\quad + \frac{1}{2m_n^2} \mathbb{E} \left[\int_S^{S+\delta} \langle R_s^n(dx), x\phi^2(1/x) \rangle ds \right] \\
&= \frac{2}{m_n} \mathbb{E} \left[\int_S^{S+\delta} \langle \mu_s^n(dx), \phi(x) \rangle^2 ds \right] + \frac{1}{2m_n^2} \mathbb{E} \left[\int_S^{S+\delta} \langle \mu_s^n(dx), \phi^2(x) \rangle ds \right] \quad (3.23)
\end{aligned}$$

We obtain, exactly as in the cases of A_p^1 and A_p^2 , that $(A_p^3)^2 \leq A\delta$. This concludes the proof of (ii). \square

In order to prove Proposition 3.7, we state the following Lemma.

Lemma 3.9 *Assume (L), (2.7). Consider a weak limit $(R_t)_{t \geq 0}$ of $(R_t^n)_{t \geq 0}$. Then for any $f \in C_b^1([0, \infty[)$, the process $(\langle R_t, f \rangle)_{t \geq 0}$ is a.s. continuous.*

Proof The jumps of $\langle R_t^n, f \rangle$ are of the form

$$m_n^{-1} \left[(X_{s-}^i + X_{s-}^j) f \left(\frac{1}{X_{s-}^i + X_{s-}^j} \right) - X_{s-}^i f \left(\frac{1}{X_{s-}^i} \right) - X_{s-}^j f \left(\frac{1}{X_{s-}^j} \right) \right] \quad (3.24)$$

But for all $x > 0, y > 0$, $(x+y)f(1/(x+y)) - xf(1/x) - yf(1/y) \leq 2\|f'\|_\infty$. We deduce that $\lim_n \sup_{t \geq 0} |\Delta \langle R_t^n, f \rangle| = 0$. This concludes the proof, since for all $T > 0$, the map $x \in \mathbb{D}([0, \infty[, \mathbb{R}) \mapsto \sup_{s \leq T} |\Delta x(s)|$ is continuous. \square

We carry on with a lemma concerning the martingale $(O_t^n)_{t \geq 0}$.

Lemma 3.10 *Assume (L) and (2.7). Then for all $T < \infty$, all $f \in C_b^1([0, \infty[)$,*

$$\lim_n \mathbb{E} [\langle O^{n,f} \rangle_T] = 0 \quad (3.25)$$

Proof Exactly as in the proof of Proposition 3.6, we obtain

$$\begin{aligned}
\mathbb{E} [\langle O^{n,f} \rangle_T] &\leq \mathbb{E} \left[\int_0^T \left\{ \frac{2}{m_n} \langle \mu_s^n, \phi \rangle^2 + \frac{1}{2m_n^2} \langle \mu_s^n, \phi^2 \rangle \right\} ds \right] \\
&\leq \frac{3}{m_n} \mathbb{E} \left[\int_0^T \langle \mu_0^n, \phi \rangle^2 ds \right] \leq AT/m_n \quad (3.26)
\end{aligned}$$

where $A = 3 \sup_n \langle \mu_0^n, \phi \rangle^2 < \infty$ thanks to (2.7). \square

Lemma 3.11 *Assume (L), (2.7). Then for all $T < \infty$, all $f \in C_c^1([0, \infty[)$, $\sup_{[0, T]} |D_t^{n,f}|$ tends to 0 a.s. and in L^1 as n tends to infinity, where*

$$D_t^{n,f} = \frac{1}{m_n} \int_0^t \langle R_s^n(dx), K(1/x, 1/x) [f(x/2) - f(x)] \rangle ds \quad (3.27)$$

Proof Since $f \in C_c^1(]0, \infty[)$, we deduce thanks to (L) that for all $x \in]0, \infty[$, $K(1/x, 1/x)[f(x/2) - f(x)] \leq \phi(1/x)x\mathbb{1}_{\{x \in [a, A]\}}$, where $0 < a < A < \infty$. Hence, $K(1/x, 1/x)[f(x/2) - f(x)] \leq C_f$ for some constant C_f depending only on f . Since R_s^n is a probability measure for each n , each s , the conclusion is straightforward. \square

Proof of Proposition 3.7 We consider a weak limit $(R_t)_{t \geq 0}$ of a subsequence of $(R_t^n)_{t \geq 0}$ that we still denote by $(R_t^n)_{t \geq 0}$.

Step 1 Since for each t , R_t is the weak limit of probability measures on $]0, \infty[$, we deduce that R_t is a probability measure on $]0, \infty[$. We now check that R satisfies condition (a) of Definition 3.4. But for every n , every t , $\langle R_t^n(dx), x\phi(1/x)\mathbb{1}_{\{x > 0\}} \rangle = \langle \mu_t^n(dx), \phi(x)\mathbb{1}_{\{x < \infty\}} \rangle = \langle \mu_t^n, \phi \rangle$. Since ϕ is subadditive, Lemma 3.1 yields $\langle R_t^n(dx), x\phi(1/x)\mathbb{1}_{\{x > 0\}} \rangle \leq \langle \mu_0^n, \phi \rangle$ which is uniformly bounded by some A thanks to (2.7). Hence for all t , $\langle R_t(dx), x\phi(1/x)\mathbb{1}_{\{x > 0\}} \rangle \leq A$.

Step 2 For some fixed $t \geq 0$ and $f \in C_c^1(]0, \infty[)$, we define a map $\Gamma = \Gamma_1 - \Gamma_2$ from $\mathbb{D}(]0, \infty[, \mathcal{P}(]0, \infty[))$ into \mathbb{R} by

$$\Gamma_1(\nu) = \langle \nu_t, f \rangle - \langle \nu_0, f \rangle \quad ; \quad \Gamma_2(\nu) = \int_0^t \langle \nu_s \otimes \nu_s, Lf \rangle ds \quad (3.28)$$

We have to show that a.s., $\Gamma(R) = 0$. First note that thanks to (3.10), Lemmas 3.10 and 3.11,

$$\lim_n \mathbb{E} [|\Gamma(R^n)|] = 0 \quad (3.29)$$

Next, we deduce from Lemma 3.9 that Γ_1 is a.s. continuous at R . One can check that Lf is continuous on $]0, \infty[^2$, using (L) and the fact that $f \in C_c^1(]0, \infty[)$. Furthermore, $Lf(x, y) \leq \|f'\|_\infty \mathbb{1}_{\{x > 0\}} [\mathbb{1}_{\{y > 0\}} xy\phi(1/x)\phi(1/y) + xl(1/x)]$. Since ϕ is continuous and subadditive, for all $\eta > 0$, there exists C_η such that for all $x \geq \eta$, $\phi(x) \leq C_\eta x$. Hence, using (2.4), for all $\varepsilon > 0$, the map $L^\varepsilon f(x, y) = Lf(x \wedge (1/\varepsilon), y \wedge (1/\varepsilon))$ is continuous and bounded on $]0, \infty[^2$. We thus deduce that the function $\Gamma_2^\varepsilon(\nu) = \int_0^t \langle \nu_s \otimes \nu_s, L^\varepsilon f \rangle ds$ is continuous on $\mathbb{D}(]0, \infty[, \mathcal{P}(]0, \infty[))$. Setting $\Gamma^\varepsilon = \Gamma_1 - \Gamma_2^\varepsilon$, we obtain, for each $\varepsilon > 0$,

$$\Gamma^\varepsilon(R) = \lim_n \Gamma^\varepsilon(R_n) \quad \text{in law} \quad (3.30)$$

Finally, a computation using (2.7) shows that a.s.,

$$\begin{aligned} \limsup_{\varepsilon \rightarrow 0} \sup_{n \geq 2} |\Gamma(R_n) - \Gamma^\varepsilon(R_n)| &\leq A \limsup_{\varepsilon \rightarrow 0} \sup_{n \geq 2} \int_0^t \langle \mu_s^n(dx), \phi(x)\mathbb{1}_{\{x < \varepsilon\}} \rangle^2 ds \\ &\leq At \limsup_{\varepsilon \rightarrow 0} \sup_{n \geq 2} \langle \mu_0^n(dx), \phi(x)\mathbb{1}_{\{x < \varepsilon\}} \rangle^2 = 0 \end{aligned} \quad (3.31)$$

where the last inequality comes from Lemma 3.1, since $\phi\mathbb{1}_{]0, \varepsilon[}$ is subadditive. One can show in the same way that a.s.,

$$\lim_{\varepsilon \rightarrow 0} |\Gamma(R) - \Gamma^\varepsilon(R)| = 0 \quad (3.32)$$

We finally may conclude: using (3.29), (3.30) and (3.31), we deduce that a.s., $\lim_{\varepsilon \rightarrow 0} \Gamma^\varepsilon(R) = 0$. Using (3.32) concludes the proof. \square

We finally give the

Proof of Theorem 2.3 We have to check that for any subsequence $\{\mu^{n_k}\}_{k \geq 1}$ of $\{\mu^n\}_{n \geq 1}$, one can find a subsequence $\{\mu^{n_{k_l}}\}_{l \geq 1}$ converging weakly to some μ , a.s. solution to (MS), for the Skorokhod topology on $\mathbb{D}([0, \infty[, M_f^+([0, \infty[))$, the set $M_f^+([0, \infty[)$ being endowed with the vague topology. But thanks to Propositions 3.6 and 3.7, this property holds for the sequence $\{R^n\}_n$, replacing equation (MS) by equation (IM). Lemma 3.5 allows us to conclude. \square

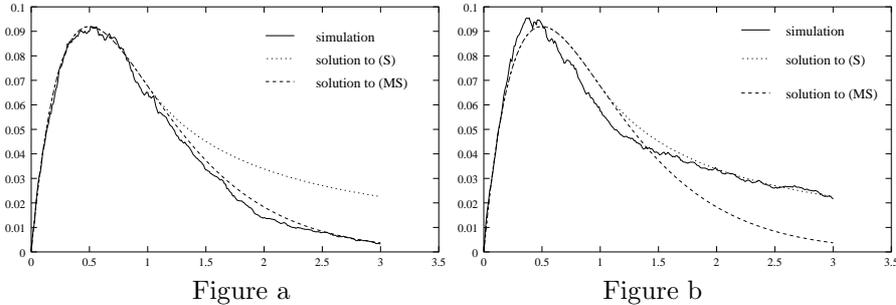
4 Numerical simulations

We now would like to illustrate the convergence result we have proved by numerical simulations. We consider the *monodisperse* initial condition $\mu_0 = \delta_1$, and the *multiplicative kernel* $K(x, y) = xy$. Under these conditions, there exists an explicit solution to the Smoluchowski equation (S) (that is, equation (MS) with $l \equiv 0$), see [3]. This solution is given by $\hat{\mu}_t(dx) = \sum_{k \geq 1} \hat{c}(t, k) \delta_k(dx)$, with

$$\hat{c}(t, k) = \frac{k^{k-2}}{k!} t^{k-1} e^{-kt} \text{ if } t \in [0, 1] \text{ and } \hat{c}(t, k) = \frac{k^{k-2}}{k!} t^{-1} e^{-k} \text{ if } t \geq 1 \quad (4.1)$$

We also have an explicit solution to (MS), see [3]. It can be written as $\mu_t(dx) = \sum_{k \geq 1} c(t, k) \delta_k(dx)$, with

$$c(t, k) = \frac{k^{k-2}}{k!} t^{k-1} e^{-kt} \text{ for } t \geq 0 \quad (4.2)$$



On Figures a and b, the dashed (resp. dotted) line represents the evolution of $c(t, 2)$ (resp. $\hat{c}(t, 2)$) for $t \in [0, 3]$. Figure a shows in full line the evolution of $\mu_t^n(\{2\})$, for $n = 5000$, obtained with one simulation using the kernel $K(x, y) = xy$ and the initial condition $\mu_0^n = \mu_0 = n^{-1} \sum_1^n \delta_1$. We observe that $\mu_t^n(\{2\})$ is close to $c(t, 2)$. Figure b shows in full line the evolution of $\mu_t^n(\{2\})$, for $n = 5000$, obtained with one simulation using the kernel $K^{1000}(x, y) = xy \mathbb{1}_{\{x+y \leq 1000\}}$ and the initial condition $\mu_0^n = \mu_0 = n^{-1} \sum_1^n \delta_1$.

We realize that $\mu_t^n(\{2\})$ is close to $\hat{c}(t, 2)$.

Our result shows that the Marcus-Lushnikov process does not converge to the Smoluchowski equation, but to equation (MS). This is due to the emergence of a *giant* particle, on which other particles coagulate quite quickly. Figure b suggests that if we forbid coalescence between the giant particle and other particles, the Marcus-Lushnikov process does converge to the Smoluchowski equation. This is not surprising, since the term involving l in (MS) represents the coalescence between the giant particle and finite particles.

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