STOCHASTIC ANALYSIS AND APPLICATIONS Vol. 22, No. 1, pp. 113–141, 2004

A Rigorous Derivation of Smoluchowski's Equation in the Moderate Limit

S. Großkinsky,^{1,*} C. Klingenberg,² and K. Oelschläger³

 ¹Zentrum Mathematik M5, Technische Universität München, München, Germany
 ²Institut für Angewandte Mathematik, Universität Würzburg, Würzburg, Germany
 ³Institut für Angewandte Mathematik, Universität Heidelberg, Heidelberg, Germany

ABSTRACT

Smoluchowski's equation is a macroscopic description of a many particle system with coagulation and shattering interactions. We give a microscopic model of the system from which we derive this equation rigorously. Provided the existence of a unique and sufficiently regular solution of Smoluchowski's equation, we prove the law of large numbers for the empirical processes. In contrast to previous derivations we assume a moderate scaling of the particle interaction, enabling us to estimate the critical fluctuation terms by using martingale inequalities. This approach can be justified in

113

DOI: 10.1081/SAP-120028026 Copyright © 2004 by Marcel Dekker, Inc. 0736-2994 (Print); 1532-9356 (Online) www.dekker.com

^{*}Correspondence: S. Großkinsky, Zentrum Mathematik M5, Technische Universität München, D-85748 Garching bei München, Germany; E-mail: stefang@ma.tum.de.

ORDER		REPRINTS
-------	--	----------

the regime of high temperatures and particle densities, which is of special interest in astrophysical studies and where previous derivations do not apply.

Key Words: Smoluchowski's equation; Moderate limit; Many particle system; Martingale.

I. INTRODUCTION

We consider a system of dust particles of $R \in \mathbb{N}$ different masses m_1, \ldots, m_R , embeded in a *d*-dimensional hot gas. Particles of size $r \in \{1, \ldots, R\}$ are drifting according to the velocity field $\vec{v}_r : \mathbb{R}^d \times \mathbb{R}_0^+ \to \mathbb{R}^d$ with a superimposed Brownian motion with diffusion constant $\sigma_r \in \mathbb{R}^+$. Two particles of size r and q collide with rate $\hat{a}_{rq} : \mathbb{R}^d \times \mathbb{R}_0^+ \to \mathbb{R}_0^+$. The material coefficients $\hat{e}_{rql} \in \mathbb{N}_0$ determine the number of particles of size $l = 1, \ldots, R$ produced by that collision event, deciding for coagulation or shattering events. In this first model we take a macroscopic viewpoint, where two colliding particles occupy the same position in spacetime (\vec{x}, t) , where the function \hat{a}_{rq} is evaluated. A complete description of the above model is given by Smoluchowski's equation,^[15] in our case a system of reaction diffusion equations for the particle densities $s_r : \mathbb{R}^d \times \mathbb{R}_0^+ \to \mathbb{R}_0^+, r = 1, \ldots, R$ (cf.,^[14] Sec. 2) with initial conditions s_r^0 : $\mathbb{R}^d \to \mathbb{R}_0^+$:

$$\begin{aligned} \partial_t s_r(\vec{x}, t) \\ &= -\vec{\nabla} \cdot (\vec{v}_r(\vec{x}, t) s_r(\vec{x}, t)) + \frac{1}{2} \sigma_r^2 \Delta s_r(\vec{x}, t) - s_r(\vec{x}, t) \sum_{q=1}^R \hat{a}_{rq}(\vec{x}, t) s_q(\vec{x}, t) \\ &+ \frac{1}{2} \sum_{q,t=1}^R \hat{a}_{qt}(\vec{x}, t) \hat{e}_{qlr} s_q(\vec{x}, t) s_l(\vec{x}, t) \\ &s_r(\vec{x}, t) = s_r^0(\vec{x}) \quad \text{for all } r = 1, \dots, R \end{aligned}$$

We suppose that for any size r the particles consist of several atoms of size 1 and we set $m_1 = 1$. Moreover, the masses of the particles are ordered as $m_1 < m_2 < \cdots < m_R$. The conservation of the total mass $M(t) = \sum_{r=1}^R \int_{\mathbb{R}^d} m_r s_r(\vec{x}, t) d^d x$ of the system under the above dynamics, i.e. $M(t) = M(0), t \ge 0$, is assured by

$$\sum_{l=1}^{R} m_l \hat{e}_{rql} = m_r + m_q \quad \text{and} \quad \hat{a}(\vec{x}, t) = \hat{a}_{qr}(\vec{x}, t).$$
(1)

ORDER		REPRINTS
-------	--	----------

for all $\vec{x} \in \mathbb{R}^d$, $t \in \mathbb{R}_0^+$, $r, q \in \{1, \dots, R\}$. We also assume that \hat{e}_{rql} is symmetric in r and q, i.e., $\hat{e}_{ral} = \hat{e}_{arl}$.

The above model is commonly used to describe reaction diffusion systems and there have been rigorous approaches to identify equation (MA) as the limit dynamics of a suitable many particle system. These derivations are restricted to one-dimensional systems,^[11] the spatially homogeneous case^[10] or a spatially discretized microscopic model.^[2,4] In Ref.^[8] there is a derivation accounting for the full space dependence of the problem, using the Boltzmann–Grad limit which is applicable for very small particle densities. In Refs.^[4,10] existence and uniqueness of a solution of Smoluchowski's equation are also studied.

In this article we give a microscopic particle model (MI) in Sec. II, from which we rigorously derive (MA) in the spatially inhomogeneous (general) case without space discretization or restrictions on space dimension. Our many particle system properly describes an astrophysical system recently studied in Refs.^[6,14], which is explained in Sec. V.A. It corresponds to a situation of high gas temperatures and particle densities, which is not covered by the derivation in the Boltzmann–Grad limit.^[8]

In this regime the dominating particle interactions are shattering collisions, so it is justified to neglect coagulation events. That means that the mass of each of the two interaction partners may not increase by the collision, but they are shattered into fragments of smaller or equal mass. This constitutes a constraint on the material coefficients \hat{e}_{rql} given in (9), which is important to ensure compatibility with the microscopic particle model. Our main theorem in Sec. III states the convergence of the empirical processes (4) to a solution of (MA) and is proved in Sec. IV. Before giving a short conclusion in the last section we also discuss two apparent generalizations of the microscopic model (MI).

The most important feature of our approach is the moderate scaling of the collision interaction, which is introduced in Sec. II.B (M3) and discussed on a physical level in Sec. V.A. It enables us to use a technique developed by Oelschläger,^[11] which was previously applied to derive the porous medium equation,^[12] or in the description of aggregation phenomena in the biological populations.^[9,13] With this technique we are able to derive Smoluchowski's equation in the spatially inhomogeneous form (MA), in a regime where the previous approaches cannot be applied.

II. MICROSCOPIC PARTICLE MODEL

Given the macroscopic model of Sec. I we present a corresponding microscopic many particle system. The most important modeling



ORDER		REPRINTS
-------	--	----------

assumptions are marked by (M1) to (M4) and are discussed in Secs. V.A and V.B.

A. Dynamics Without Interaction

Let $N_r(t)$ be the number of particles of species $r \in \{1, ..., R\}$ and $N(t) = \sum_{r=1}^{R} N_r(t)$ the number of all particles at time $t \in \mathbb{R}_0^+$. The system size N is characterized by the number of atoms of mass $m_1 = 1$ at time t = 0:

$$N = \left(\sum_{r=1}^{R} N_r(0)m_r\right) / m_1 = \sum_{r=1}^{R} N_r(0)m_r$$
(2)

Let $M(N, t) \subset \mathbb{N}$ be the set of all particles and $M(N, r, t) \subset M(N, t)$, r = 1, ..., R, the subsets of particles of species r at time t, where each particle is identified with a unique integer number.

- (M1) The particles are considered to be point masses with positions $\vec{X}_N^k(t) \in \mathbb{R}^d$, $k \in M(N, t)$, at time t in a system of size N. Each particle of species $r \in \{1, ..., R\}$ is given the rescaled mass $m_{N,r} = m_r/N$, which keeps the initial mass $M_N(0) = \sum_{r=1}^R m_{N,r} N_r(0) = 1$ independent of the system size according to (2).
- (M2) Neglecting the hydrodynamic drag interaction between gas and particles, we consider the latter to move according to the given velocity fields \vec{v}_r and Brownian motion with diffusion constants σ_r , r = 1, ..., R, introduced in the macroscopic equation (MA).

Between two subsequent collision events the system at time t is then described by N(t) uncoupled stochastic differential equations:

$$d\bar{X}_{N}^{k}(t) = \vec{v}_{r}(\bar{X}_{N}^{k}(t), t)dt + \sigma_{r}d\bar{B}^{k}(t), \quad k \in M(N, r, t), \quad r = 1, \dots, R$$
(3)

The $(\overline{B}^k(t))_{t \in \mathbb{R}^+_p}$, $k \in \mathbb{N}$, are independent Wiener processes modelling the Brownian motion of the particles. We always assume the existence of a filtration $(\mathcal{F}_t)_{t \geq 0}$, with respect to which the stochastic processes under consideration are adapted (cf.^[11], Sec. 2.B) and which fulfills the usual conditions.^[7]

The particle interaction is described by suitable changes of the sets M(N, r, t) and is explained in the next subsection. A microscopic

ORDER		REPRINTS
-------	--	----------

quantity comparable to the particle density in (MA) is given by the measure-valued, empirical processes:

$$S_{N,r}: \mathbb{R}_0^+ \to \mathcal{M}(\mathbb{R}^d), \ S_{N,r}(t) := \frac{1}{N} \sum_{k \in \mathcal{M}(N,r,t)} \delta_{\bar{X}_N^k(t)}, \ r = 1, \dots, R,$$
 (4)

where $\mathcal{M}(\mathbb{R}^d)$ denotes the space of positive, finite measures on \mathbb{R}^d and $\delta_{\vec{x}}$ is the Dirac measure concentrated in $\vec{x} \in \mathbb{R}^d$. $S_{N,r}$ describes the timeevolution of the spatial distribution of particles within the subpopulation of species *r*. It is known by the law of large numbers that the empirical distribution of *N* independent, identically distributed random variables converges to their probability distribution in the limit $N \to \infty$. In this paper we prove the convergence for stochastic processes which are not independent for times t > 0, due to the particle interaction.

B. Description of the Particle Interaction

Due to (M1) we have to specify a model for the "collision" interaction of two point particles.

(M3) We take a stochastic model determined by a rate depending on the distance of the interaction partners k and l. The scaling of this rate is given by

$$\frac{1}{N}W_N(\vec{X}_N^k(t) - \vec{X}_N^l(t)), \text{ where } W_N(\vec{x}) = \alpha_N^d W_1(\alpha_N \vec{x}), \tag{5}$$

with $\alpha_N = N^{\beta/d}$ and a moderate scaling parameter $0 < \beta < 1$. We assume that W_1 is symmetric and positive with $||W_1||_1 = 1$. It follows that $||W_N||_1 = 1$ for all $N \in \mathbb{N}$ and $\lim_{N \to \infty} W_N(.-\vec{x}) = \delta_{\vec{x}}$ for all $\vec{x} \in \mathbb{R}^d$ in the sense of distributions.

In contrast to the usual hydrodynamic scaling with $\beta = 1$ this leads to a microscopically large interaction volume. This assumption is motivated and justified in a physical context in Sec. V.A.

(M4) Instead of considering pair interactions (see Sec. V.B) we assume that every particle $k \in M(N, r, t)$ interacts with an effective field of all other particles of species q with rate

$$a_{N,rq}(\vec{X}_{N}^{k}(t), t) := \min \{C_{a}, \hat{a}_{rq}(\vec{X}_{N}^{k}(t), t) \\ \times \{(S_{N,q}(t) * W_{N})(\vec{X}_{N}^{k}(t)) - \delta_{r,q}W_{N}(\vec{0})/N\}\},$$
(6)

where \hat{a}_{rq} is the macroscopic collision rate given in Sec. I.

Copyright © Marcel Dekker, Inc. All rights reserved.

ORDER		REPRINTS
-------	--	----------

In (6) we used the generalized convolution product

$$S_{N,q}(t) * W_N := \int_{\mathbb{R}^d} W_N(. - \vec{x}) S_{N,q}(t) (d^d x)$$

= $\frac{1}{N} \sum_{l \in \mathcal{M}(N,q,t)} W_N(. - \vec{X}_N^l(t)).$ (7)

By substraction of the term including Kronecker's delta in (6) self-interaction is excluded. The rate is bounded uniformly in N by a suitable constant C_a , which is specified in condition (C5) in Sec. III.B. This cut-off prevents diverging interaction rates due to high particle concentrations in the limit $N \rightarrow \infty$. Each possible interaction event is described by a jump process

$$a_{N,rq}^{*,k}(t) := \beta_{N,rq}^{k} \left(\int_{0}^{t} \chi_{M(N,r,s)}(k) a_{N,rq}(\vec{X}_{N}^{k}(s), s) ds \right) \in \{0, 1\},$$
(8)

where $\beta_{N,rq}^k : \mathbb{R}_0^+ \to \mathbb{N}_0$ are independent standard Poisson processes with a transformed time argument in the brackets (cf.^[11]) and $\chi_A \in \{0, 1\}$ is the indicator function of the set *A*.

The process $a_{N,rq}^{*,k}$ jumps from 0 to 1 at some time $t \ge 0$ if particle k exists in t-, belongs to species r and interacts with a particle of species q at time t. After the interaction the number k is removed from the sets M(N, r, t) and M(N, t). The mass of particle k is distributed on the interaction products according to the microscopic material coefficient $e_{rql} \in \mathbb{N}_0$. The latter fulfills conservation of mass and is related to its macroscopic counterpart \hat{e}_{rql} in the following way:

$$\sum_{l=1}^{K} m_l e_{rql} = m_r, \ \hat{e}_{rql} = \hat{e}_{qrl} = e_{rql} + e_{qrl} \text{ for all } r, q, l = 1, \dots, R.$$
(9)

We note that this constitutes a condition on \hat{e}_{rql} , corresponding to the absence of coagulation mentioned in Sec. I. The particles resulting from the interaction are located at $\vec{X}_N^k(t)$ and obtain new numbers starting with max{ $p \in M(N, s) : s \leq t$ } + 1, which were previously not assigned to any particle. These numbers are added to M(N, t) and the subsets corresponding to the various species. We note that any process $a_{N,rq}^{*,k}$ only jumps once, since after that jump the respective particle k disappears, i.e., $\chi_{M(N,r,t+)}(k) \equiv 0$.

C. Complete Description of the Model

Using a generalized L^2 -scalar product we can formulate the time evolution of the empirical processes in a weak sense. For all

Copyright © Marcel Dekker, Inc. All rights reserved



ORDER		REPRINTS
-------	--	----------

 $f \in C_b^2(\mathbb{R}^d, \mathbb{R})$ and $r = 1, \ldots, R$ we have

$$\langle S_{N,r}(t), f \rangle := \int_{\mathbb{R}^d} f(\vec{x}) S_{N,r}(t) (d^d x) = \frac{1}{N} \sum_{k \in \mathcal{M}(N,r,t)} f(\vec{X}_N^k(t)) = \frac{1}{N} \sum_{k \in \mathcal{M}(N,r,0)} f(\vec{X}_N^k(0)) + \int_0^t d\left(\frac{1}{N} \sum_{k \in \mathcal{M}(N,r,s)} f(\vec{X}_N^k(s))\right).$$
(10)

Inserting the expression for $d\vec{X}_N^k(s)$ from Eq. (3) and using Itô's formula^[7] we get:

$$\begin{split} \langle S_{N,r}(t), f \rangle \\ &= \frac{1}{N} \sum_{k \in M(N,r,0)} f(\vec{X}_{N}^{k}(0)) + \frac{\sigma_{r}}{N} \int_{0}^{t} \sum_{k \in M(N,r,s)} \vec{\nabla} f(\vec{X}_{N}^{k}(s)) \cdot d\vec{B}^{k}(s) \\ &+ \frac{1}{N} \int_{0}^{t} \sum_{k \in M(N,r,s)} \left(\vec{\nabla} f(\vec{X}_{N}^{k}(s)) \cdot \vec{v}_{r}(\vec{X}_{N}^{k}(s), s) + \frac{\sigma_{r}^{2}}{2} \Delta f(\vec{X}_{N}^{k}(s)) \right) ds \\ &- \frac{1}{N} \sum_{q=1}^{R} \int_{0}^{t} \sum_{k \in M(N,r,s)} f(\vec{X}_{N}^{k}(s)) a_{N,rq}^{*,k}(ds) \\ &+ \frac{1}{N} \sum_{q,l=1}^{R} \int_{0}^{t} \sum_{k \in M(N,q,s)} f(\vec{X}_{N}^{k}(s)) e_{qlr} a_{N,ql}^{*,k}(ds) \end{split}$$
(11)

The first integral term describes the stochastic fluctuations of the particle positions and the second one particle transport and diffusion, resulting from the interaction free description (3). The next two terms consider the change of the sets M(N, r, s) in (10) due to the loss of particles of species r after interactions with others, and the gain of such particles from products of other interactions. We separate the fluctuation terms due to stochasticity in the free particle dynamics and the interaction in stochastic integrals. So we get for all $f \in C_b^2(\mathbb{R}^d, \mathbb{R})$ and $r = 1, \ldots, R$ the complete description of our microscopic model:

$$\begin{split} \langle S_{N,r}(t), f \rangle &= \langle S_{N,r}(0), f \rangle + \int_0^t \left\langle S_{N,r}(s), \vec{\nabla} f \cdot \vec{v}_r(.,s) + \frac{\sigma_r^2}{2} \Delta f \right\rangle ds \\ &- \sum_{q=1}^R \int_0^t \langle S_{N,r}(s), f a_{N,rq}(.,s) \rangle ds \end{split}$$

Copyright @ Marcel Dekker, Inc. All rights reserved.



ORDER		REPRINTS
-------	--	----------

$$+ \sum_{q,l=1}^{R} \int_{0}^{t} \langle S_{N,q}(s), fe_{qlr} a_{N,ql}(.,s) \rangle ds + M_{N,r}^{1}(f,t) + M_{N,r}^{2a}(f,t) + M_{N,r}^{2b}(f,t), \text{ with} M_{N,r}^{1}(f,t) = \frac{\sigma_{r}}{N} \int_{0}^{t} \sum_{k \in \mathcal{M}(N,r,s)} \vec{\nabla} f(\vec{X}_{N}^{k}(s)) \cdot d\vec{B}^{k}(s) M_{N,r}^{2a}(f,t) = -\frac{1}{N} \sum_{q=1}^{R} \int_{0}^{t} \sum_{k \in \mathcal{M}(N,r,s)} f(\vec{X}_{N}^{k}(s)) \times (a_{N,rq}^{*,k}(ds) - a_{N,rq}(\vec{X}_{N}^{k}(s), s) ds) M_{N,r}^{2b}(f,t) = \frac{1}{N} \sum_{q,l=1}^{R} \int_{0}^{t} \sum_{k \in \mathcal{M}(N,q,s)} f(\vec{X}_{N}^{k}(s)) e_{qlr} \times (a_{N,ql}^{*,k}(ds) - a_{N,ql}(\vec{X}_{N}^{k}(s), s) ds)$$

and initial conditions $S_{N,r}(0) = N^{-1} \sum_{k \in \mathcal{M}(N,r,0)} \delta_{\tilde{X}_{N}^{k}(0)}.$ (MI)

This set of equations combines all features mentioned in the preceding two subsections and is used to derive the macroscopic model (MA), shown in the next section.

III. DERIVATION OF SMOLUCHOWSKI'S EQUATION

We show how to obtain (MA) heuristically from our microscopic particle model (MI), leading us to a proper formulation of the main theorem.

A. Heuristic Derivation of the Macroscopic Equation

The empirical processes $S_{N,r}$ are defined as solutions of (MI). For this subsection we assume that for every r = 1, ..., R they converge to limit processes $S_r : [0, T] \to \mathcal{M}(\mathbb{R}^d)$ on a compact time interval [0, T] in a yet unspecified sense. The limit processes are assumed to be absolutely continuous with respect of Lebesque measure on \mathbb{R}^d and therefore have densities, s_r , which should be in $C_b^2(\mathbb{R}^d \times \mathbb{R}_0^+, \mathbb{R}_0^+)$. With the generalized scalar product defined in (10) we therefore have $\langle S_r(t), g(., t) \rangle = \langle s_r(., t), g(., t) \rangle$ for all $g \in C_b(\mathbb{R}^d \times [0, T], \mathbb{R})$. We also assume the validity of conditions (C1) to (C7) given in the next subsection.



Marcel Dekker, Inc. 270 Madison Avenue, New York, New York 10016

ORDER		REPRINTS
-------	--	----------

In Sec. IV.D we get the following for the stochastic integrals in (MI) for any T > 0:

$$\lim_{N \to \infty} E \left[\sup_{t \in [0,T]} |M_{N,r}^{i}(f,t)| \right] = 0 \quad \text{for } i = 1, 2a, 2b,$$
(12)

so the fluctuation terms asymptotically vanish in any compact time interval and the limit equation is supposed to be deterministic (see (14)). The convergence of the $S_{N,r}$ should be sufficiently strong to assure the following:

$$\lim_{N \to \infty} E[\langle S_{N,r}(t), g(., t) \rangle] = \langle s_r(., t), g(., t) \rangle$$
$$\lim_{N \to \infty} E[\langle S_{N,r}(t), (S_{N,q}(t) * W_N)g(., t) \rangle] = \langle s_r(., t), s_q(., t)g(., t) \rangle, \quad (13)$$

for all $r, q = 1, ..., R, t \in [0, T]$ and $g \in C_b(\mathbb{R}^d \times [0, T], \mathbb{R}^d)$. The first condition assures the convergence of the drift and diffusion term in (MI) and the second one is needed for the interaction terms. We formally substitute the above limits into (MI) and notice that the self interaction term in (6) vanishes for $N \to \infty$. Therefore we get the following deterministic integral equation for all test functions $f \in C_b^2(\mathbb{R}^d, \mathbb{R}), t \in [0, T]$ and r = 1, ..., R:

$$\langle s_{r}(.,t), f \rangle = (s_{r}(.,0), f) + \int_{0}^{t} ds \left\langle s_{r}(.,s), \vec{\nabla}f \cdot \vec{v}_{r}(.,s) + \frac{1}{2}\sigma_{r}^{2}\Delta f \right\rangle$$

$$- \sum_{q=1}^{R} \int_{0}^{t} ds \left\langle s_{r}(.,s), f \hat{a}_{rq}(.,s) s_{q}(.,s) \right\rangle$$

$$+ \sum_{q,l=1}^{R} \int_{0}^{t} ds \left\langle s_{q}(.,s), f e_{qlr} \hat{a}_{ql}(.,s) s_{l}(.,s) \right\rangle$$

$$(14)$$

After partial integration in the transport and diffusion terms one immediately recognizes this as a weak version of Smoluchowski's equation. Using (1) and (9) it is easy to get the last line in the form (MA).

Therefore we showed that, assuming the empirical processes converge, their limit densities fulfill a weak form of Smoluchowski's equation. In the next subsection we explain how to prove this convergence in an appropriate rigorous limit sense, which can be seen from (13) to be of L^2 -type.



ORDER		REPRINTS
-------	--	----------

B. Convergence Theorem

To formulate the convergence theorem we use the following distance function between the empirical processes (MI) and the solution of Smoluchowski's equation (MA) specified in (C3) below:

$$d_{N,r}(.,t) := h_{N,r}(.,t) - s_r(.,t), \text{ where } h_{N,r}(.,t) := S_{N,r}(t) * \widehat{W}_N,$$
(15)

for all r = 1, ..., R and $t \in \mathbb{R}_0^+$. The convolution kernel \widehat{W}_N smooths out the empirical processes and obeys the following regularity conditions:

(C1) \widehat{W}_N is a different scaling of the interaction function W_N and both have to fulfill:

$$\widehat{W}_N(\vec{x}) = \widehat{\alpha}_N^d W_1(\widehat{\alpha}_N \vec{x})$$
 and $W_N(\vec{x}) = \alpha_N^d W_1(\alpha_N \vec{x})$,

where

$$\hat{\alpha}_N = N^{\hat{\beta}/d}, 0 < \hat{\beta} < \frac{d}{d+2} \text{ and } \alpha_N = N^{\beta/d}, 0 < \beta < \frac{\hat{\beta}}{d+1}$$

The scaling parameter $\hat{\beta}$ plays no role in the dynamics of the manyparticle system. However, by the above assumptions some restrictions on the parameter β determining the moderate interaction are introduced.

(C2) The unscaled function $W_1 \in L^1 \cap C_b^2(\mathbb{R}^d, \mathbb{R})$ is symmetric, positive and standardized, i.e., $||W_1||_1 = 1$. We also need $\int_{\mathbb{R}^d} |\vec{x}| W_1(\vec{x}) d^d x < 0$ ∞ and the Fourier transform \widehat{W}_1 has to fulfill:

- (a) $\widehat{W}_1 \in C_b^2(\mathbb{R}^d)$. (b) $|\widehat{W}_1(\vec{\tau})| \le C \exp(-C'|\vec{\tau}|)$. (c) $|\Delta \widehat{W}_1(\vec{\tau})| \le C(1+|\vec{\tau}|^2) |\widehat{W}_1(\vec{\tau})|$. (d) $v \mapsto |\widehat{W}_1(v\vec{\tau})|, v \ge 0$, monotonicly decreasing for all fixed $\vec{\tau} \in C_{\tau}$. \mathbb{R}^{d} .

A Gaussian probability density is an example for W_1 which obeys these conditions. To the knowledge of the authors there is no proof of the existence of a sufficiently smooth solution of the macroscopic equations,



ORDER		REPRINTS
-------	--	----------

therefore we have to assume the following:

(C3) There exists a positive, unique C_b^2 -solution (s_1, \ldots, s_R) of Smoluchowski's equation (MA) in the time interval $[0, T^*]$ for some positive T^* . The functions $s_r(., t)$ and their partial derivatives are $L^2(\mathbb{R}^d, \mathbb{R}^d_{+})$ -bounded uniformly in $t \in [0, T^*]$.

(C4) The macroscopic collision rate $\hat{a}_{rq}(., t)$ given in Sec. I should be differentiable bounded and fulfill the conditions (1) for all $t \in [0, T^*]$. The macroscopic material coefficient \hat{e}_{rql} should obey condition (1) and together with its counterpart e_{rql} given in Sec. II.B, it should fulfill (9) and be symmetric in r and q.

(C5) The upper bound C_a for the microscopic interaction rates (6) is given so that the limit equation is not affected, $C_a > \max_{r,q \in \{1,...,R\}} \sup_{t \in [0,T^*]} \|\hat{a}_{rq}(.,t)s_q(.,t)\|_{\infty}$.

(C6) The velocity fields of the different particle species have to fulfill

$$\vec{v}_r \in C_b^1(\mathbb{R}^d \times [0, T^*], \mathbb{R}^d)$$
 for all $r = 1, \dots, R$.

(C7) The diffusion constants of all particle species have to be positive, i.e., $\sigma_r > 0$ for all r = 1, ..., R.

We note that our proof only applies if all particles are Brownian. Now we are ready to formulate our main convergence result.

Theorem. With conditions (C1) to (C7) and $\lim_{N\to\infty} E\left[\sum_{r=1}^{R} \|d_{N,r}(.,0)\|_{2}^{2}\right] = 0$ it is

$$\lim_{N \to \infty} E \left[\sum_{r=1}^{R} \sup_{t \in [0, T^*]} \| d_{N, r}(., t) \|_2^2 \right] = 0.$$
(16)

Convergence at time t = 0 is given if the initial conditions of (MA) and (MI) are compatible. One possibility is to take the particle positions $\vec{X}_N^k(0), k \in M(N, r, 0)$ as independent, identically distributed random variables with suitably normalized densities $s_r^0 / \sum_{r=1}^R \langle s_r^0, 1 \rangle$ for all r = 1, ..., R. For discussion of this point see Ref.^[12] (Sec. 4B).



ORDER		REPRINTS
-------	--	----------

To formulate the result without the smoothing convolution kernel \widehat{W}_N we introduce a metric on $\mathscr{M}(\mathbb{R}^d)$ by

$$D(\mu, \nu) := \sup\{|\langle \mu - \nu, f \rangle| : f \in \Omega_D\} \text{ for all } \mu, \nu \in \mathcal{M}(\mathbb{R}^d) \text{ and}$$
$$\Omega_D := \{f \in C_b^1 \cap L^2(\mathbb{R}^d, \mathbb{R}) : \|f\|_{\infty} + \|\vec{\nabla}f\|_{\infty} + \|f\|_2 \le 1\}.$$
(17)

This quantifies a distance between the empirical processes defined in (MI) and the processes $S_r(t), t \in [0, T^*]$, with densities $s_r(., t)$ given by the solution (C3) of the macroscopic equation. As the theorem states convergence in an L^2 -sense the convergence in the weak sense (17) is easy to conclude.

Corollary. With the conditions of the theorem we have

$$\lim_{N \to \infty} E \left[\sum_{r=1}^{R} \left(\sup_{t \in [0, T^*]} D(S_{N, r}(t), S_r(t)) \right) \right] = 0.$$
(18)

IV. PROOF OF THE CONVERGENCE RESULT

A. Preliminaries

The following lemma is useful in central estimates of Sec. IV.

Lemma. With $f \in L^2 \cap C_b^1(\mathbb{R}^d, \mathbb{R}), \, \vec{\nabla} f \in L^2(\mathbb{R}^d, \mathbb{R}^d)$ we have

$$\|f - f * W_N\|_2^2 \le C\alpha_N^{-2} \|\bar{\nabla}f\|_2^2 \quad and \quad \|f - f * W_N\|_{\infty} \le C\alpha_N^{-1} \|\bar{\nabla}f\|_{\infty}.$$
(19)

An analogous estimate is true, if W_N and α_N are replaced by \widehat{W}_N and $\hat{\alpha}_N$. For any finite, positive measure μ on \mathbb{R}^d and with $U_N(\vec{x}) := |\vec{x}| \widehat{W}_N(\vec{x})$ there is

$$\|\mu * U_N\|_2^2 \le C \hat{\alpha}_N^{2\epsilon-2} \|\mu * \widehat{W}_N\|_2^2 + \langle \mu, 1 \rangle^2 \exp(-C' \hat{\alpha}_N^{\epsilon}) \quad for \ all \ \epsilon > 0.$$
(20)

For any finite, signed measure μ on \mathbb{R}^d it is

$$\|\mu * W_N\|_2^2 \le \|\mu * \widehat{W}_N\|_2^2. \tag{21}$$

Proof. see Ref.^[11] Sec. 4A, B and Ref.^[12] Sec. 5B, or Ref.^[3], Sec. 4.3

Marcel Dekker, Inc.

270 Madison Avenue, New York, New York 10016



Ŵ

ORDER		REPRINTS
-------	--	----------

In the proof of the the lemma there is essentially made use of the conditions (C1) and (C2) on the interaction function W_N and the kernel \widehat{W}_{N} . Due to the conservation of mass in the microscopic system (9) and with (2) we get the following bound on the empirical processes,

$$\sum_{r=1}^{R} \langle S_{N,r}(t), 1 \rangle = \frac{1}{N} \sum_{k \in M(N,t)} 1 \le \frac{N}{N} = 1.$$
(22)

We also use the following property without explicitly noting it for all suitable f and g, such that the expressions are well defined:

$$\langle f, g * W_N \rangle = \langle f * W_{N,g} \rangle$$
 and $\langle f, g * \widehat{W}_N \rangle = \langle f * \widehat{W}_{N,g} \rangle$, (23)

because W_1 is symmetric according to (C2). Throughout this chapter C, C' etc., denote suitably chosen constants, whose value can vary from line to line.

B. Proof of the Theorem

To prove statement (16) we first look at the time evolution of the quantity

$$\|d_{N,r}(.,t)\|_{2}^{2} = \|h_{N,r}(.,t)\|_{2}^{2} - 2\langle h_{N,r}(.,t), s_{r}(.,t)\rangle + \|s_{r}(.,t)\|_{2}^{2}.$$
 (24)

The dynamics of the first two terms is obtained analogous to (11) using (3), (8), (23) and Itô's formula:

$$\|h_{N,r}(.,t)\|_{2}^{2} = \frac{1}{N^{2}} \sum_{k,l \in \mathcal{M}(N,r,t)} (\widehat{W}_{N} * \widehat{W}_{N}) (\vec{X}_{N}^{k}(t) - \vec{X}_{N}^{l}(t))$$
$$\langle h_{N,r}(.,t), s_{r}(.,t) \rangle = \frac{1}{N} \sum_{k \in \mathcal{M}(N,r,t)} (s_{r}(.,t) * \widehat{W}_{N}) (\vec{X}_{N}^{k}(t)).$$

We just have to replace the test function f in (11) by $S_{N,r}(t) * (\widehat{W}_N * \widehat{W}_N)$ resp. $s_r(., t) * \widehat{W}_N$.

The expansion of the third term in (24) follows from the macroscopic equation (MA):

$$\|s_r(.,t)\|_2^2 = \|s_r(.,0)\|_2^2 + \int_0^t ds \langle s_r(.,s), -2\vec{\nabla} \cdot (\vec{v}_r(.,s)s_r(.,s)) + \sigma_r^2 \Delta s_r(.,s) \rangle$$

Copyright @ Marcel Dekker, Inc. All rights reserved.

Marcel Dekker, Inc. 270 Madison Avenue, New York, New York 10016

ORDER		REPRINTS
-------	--	----------

$$+\sum_{q=1}^{R}\int_{0}^{t} ds \Big\langle s_{r}(.,s), -2s_{r}(.,s)\hat{a}_{rq}(.,s)s_{q}(.,s) + s_{q}(.,s)\sum_{l=1}^{R}\hat{e}_{qlr}\hat{a}_{ql}(.,s)s_{l}(.,s)\Big\rangle$$

Combining the parts suitably by using (9) to express \hat{e}_{qlr} in terms of e_{qlr} we get:

$$\begin{split} \|d_{N,r}(.,t)\|_{2}^{2} &= \|d_{N,r}(.,0)\|_{2}^{2} + \sigma_{r}^{2} \int_{0}^{t} ds \langle d_{N,r}(.,s), \Delta d_{N,r}(.,s) \rangle \\ &+ 2 \int_{0}^{t} ds (\langle S_{N,r}(s), \bar{\nabla}(d_{N,r}(.,s) * \hat{W}_{N}) \cdot \vec{v}_{r}(.,s) \rangle \\ &- \langle s_{r}(.,s), \bar{\nabla}d_{N,r}(.,s) \cdot \vec{v}_{r}(.,s) \rangle) \\ &- 2 \sum_{q=1}^{R} \int_{0}^{t} ds (\langle S_{N,r}(s), (d_{N,r}(.,s) * \hat{W}_{N})a_{N,rq}(.,s) \rangle \\ &- \langle s_{r}(.,s), d_{N,r}(.,s) \hat{a}_{rq}(.,s)s_{q}(.,s) \rangle) \\ &+ 2 \sum_{q,l=1}^{R} \int_{0}^{t} ds \, e_{qlr}(\langle S_{N,q}(s), (d_{N,r}(.,s) * \hat{W}_{N})a_{N,ql}(.,s) \rangle \\ &- \langle s_{q}(.,s), d_{N,r}(.,s) \hat{a}_{ql}(.,s)s_{q}(.,s) \rangle) \\ &+ 2 \int_{0}^{t} \frac{\sigma_{r}}{N} \sum_{k \in \mathcal{M}(N,r,s)} \bar{\nabla}(d_{N,r}(.,s) * \hat{W}_{N})(\vec{X}_{N}^{k}(s)) \cdot d\vec{B}^{k}(s) \\ &- 2 \sum_{q=1}^{R} \int_{0}^{t} \frac{1}{N} \sum_{k \in \mathcal{M}(N,r,s)} (d_{N,r}(.,s) * \hat{W}_{N})(\vec{X}_{N}^{k}(s)) \\ &\times (a_{N,rq}^{*,k}(ds) - a_{N,rq}(\vec{X}_{N}^{k}(s), s) ds) \\ &+ 2 \sum_{q,l=1}^{R} \int_{0}^{t} \frac{1}{N} \sum_{k \in \mathcal{M}(N,q,s)} (d_{N,r}(.,s) * \hat{W}_{N})(\vec{X}_{N}^{k}(s)) \\ &\times e_{qlr}(a_{N,ql}^{*,k}(ds) - a_{N,ql}(\vec{X}_{N}^{k}(s), s) ds) \\ &- \sigma_{r}^{2} \frac{\Delta(\widehat{W}_{N} * \widehat{W}_{N})(\vec{0})}{N} \int_{0}^{t} ds \langle S_{N,r}(s), a_{N,rq}(.,s) \rangle \end{split}$$

Copyright @ Marcel Dekker, Inc. All rights reserved.



ORDER		REPRINTS
-------	--	----------

$$+ \frac{(\widehat{W}_{N} * \widehat{W}_{N})(\vec{0})}{N} \sum_{q,l=1}^{R} \int_{0}^{t} ds \, e_{qlr} \langle S_{N,q}(s), a_{N,ql}(.,s) \rangle$$

$$+ \frac{(\widehat{W}_{N} * \widehat{W}_{N})(\vec{0})}{N^{2}} \sum_{q=1}^{R} \int_{0}^{t} \sum_{k \in \mathcal{M}(N,r,s)} \left(a_{N,rq}^{*,k}(ds) - a_{N,rq}(\vec{X}_{N}^{k}(s),s)ds\right)$$

$$+ \frac{(\widehat{W}_{N} * \widehat{W}_{N})(\vec{0})}{N^{2}} \sum_{q,l=1}^{R} \int_{0}^{t} \sum_{k \in \mathcal{M}(N,q,s)} e_{qlr}\left(a_{N,ql}^{*,k}(ds) - a_{N,ql}(\vec{X}_{N}^{k}(s),s)ds\right)$$

$$= \|d_{N,r}(.,0)\|_{2}^{2}$$

$$+ \int_{0}^{t} \left(T_{N,r}^{1}(s) + T_{N,r}^{2}(s) + T_{N,r}^{3}(s) + T_{N,r}^{4}(s))\right) ds$$

$$+ 2\left(\widehat{M}_{N,r}^{1}(t) + \widehat{M}_{N,r}^{2a}(t) + \widehat{M}_{N,r}^{2b}(t)\right)$$

$$+ T_{N,r}^{0}(t) + T_{N,r}^{0a}(t) + T_{N,r}^{0b}(t) + \widehat{M}_{N,r}^{0a}(t) + \widehat{M}_{N,r}^{0b}(t)$$

$$(25)$$

The terms in the above sum are labeled line by line. T_N^1 derives from the diffusion due to Brownian motion, T_N^2 from the particle transport, T_N^3 from the loss and T_N^4 from the gain of particles due to interactions. The stochastic integrals $\widehat{M}_{N,r}^1, \widehat{M}_{N,r}^{2a}$ and $\widehat{M}_{N,r}^{2b}$ represent the fluctuations due to stochasticity in the free particle dynamics and the interaction. The remaining terms are corrections resulting from the expansion of $\|h_{N,r}(.,t)\|_2^2$. With Itô's formula and (3) we get for $T_{N,r}^0$:

$$\begin{split} \frac{1}{N^2} \sum_{\substack{k,l \in \mathcal{M}(N,r,t)}} d(\widehat{W}_N \ast \widehat{W}_N)(\vec{X}_N^k(t) - \vec{X}_N^l(t)) \\ &= \frac{1}{N^2} \sum_{\substack{k,l \in \mathcal{M}(N,r,t) \\ k \neq 1}} \left(\vec{\nabla}(\widehat{W}_N \ast \widehat{W}_N)(\vec{X}_N^k(t) - \vec{X}_N^l(t))(d\vec{X}_N^k(t) - d\vec{X}_N^l(t)) \\ &\quad + \frac{\sigma_r^2}{2} \Delta(\widehat{W}_N \ast \widehat{W}_N)(\vec{X}_N^k(t) - \vec{X}_N^l(t))(dt + dt) \right) \\ &= 2 \langle S_{N,r}(t), \vec{\nabla}(h_{N,r}(.,t) \ast \widehat{W}_N) \cdot \vec{v}_r(.,t) \rangle dt \\ &\quad + \frac{2\sigma_r}{N} \sum_{\substack{k \in \mathcal{M}(N,r,t)}} \vec{\nabla}(h_{N,r}(.,t) \ast \widehat{W}_N) \cdot d\vec{B}^k(t) \\ &\quad + \sigma_r^2 \langle h_{N,r}(.,t), \Delta h_{N,r}(.,t) \rangle dt - \frac{\sigma_r^2}{N^2} \sum_{\substack{k \in \mathcal{M}(N,r,t)}} \Delta(\widehat{W}_N \ast \widehat{W}_N)(\vec{0}) dt \end{split}$$

Copyright @ Marcel Dekker, Inc. All rights reserved.



ORDER		REPRINTS
-------	--	----------

 $T_{N,r}^{0a}$ and $T_{N,r}^{0b}$ can be derived analogously by considering the change of the sets M(N, q, t), q = 1, ..., R. The fluctuations of these corrections are separated in stochastic integrals $\widehat{M}_{N,r}^{0a}$ and $\widehat{M}_{N,r}^{0b}$.

Estimate of the correction terms.

128

With (C1), (C2) and (22) we get:

$$\begin{aligned} |T_{N,r}^{0}(t)| &\leq \frac{t\sigma_{r}^{2}}{N} \bigg(\int_{\mathbb{R}^{d}} |\widehat{W}_{N}(\vec{x})\Delta\widehat{W}_{N}(\vec{0}-\vec{x})|d^{d}x \bigg) \sup_{s\leq t} \langle S_{N,r}(s), 1 \rangle \\ &\leq \frac{C}{N} t \int_{\mathbb{R}^{d}} N^{\hat{\beta}(1+2/d)} \|\Delta W_{1}\|_{\infty} |\widehat{W}_{N}(\vec{x})|d^{d}x \leq C t N^{\hat{\beta}(1+2/d)-1} \end{aligned}$$

The term vanishes in the limit $N \to \infty$, because with (C1) it is $\hat{\beta} < d/(d+2)$. Using also (6) and (9) we get completely analogous:

 $|T_{N,r}^{0a}(t)| + |T_{N,r}^{0b}(t)| \le CtN^{\hat{\beta}-1}$

The stochastic integrals are estimated in Sec. IV.D.

Estimate of $T_{N,r}^1(s) = \sigma_r^2 \langle d_{N,r}(., s), \Delta d_{N,r}(., s) \rangle$. After partial integration we get with condition (C7): $T_{N,r}^1(s) =$ $-\sigma_r^2 \|\vec{\nabla} d_{N,r}(.,s)\|_2^2 < 0$. This term is negative and can be used to cancel positive contributions of the same kind arising in the estimates of $T_{N,r}^2$ and $\widehat{M}^1_{N,r}$.

Estimate of $T_{N,r}^2(s) = 2(\langle S_{N,r}(s), \vec{\nabla}(d_{N,r}(.,s) * \widehat{W}_N) \cdot \vec{v}_r(.,s) \rangle - \langle s_r(.,s), v \rangle$ $\nabla d_{N,r}(.,s) \cdot \vec{v}_r(.,s) \rangle$).

To contract the two brackets we make the following replacement:

$$\begin{split} \langle S_{N,r}(s), (\vec{\nabla}d_{N,r}(.,s) * \widehat{W}_N) \cdot \vec{v}_r(.,s) \rangle \\ &= \langle h_{N,r}(.,s), \vec{\nabla}d_{N,r}(.,s) \cdot \vec{v}_r(.,s) \rangle + R_{N,r}^2(s), \end{split}$$

where the correction term is estimated using (C6):

$$\begin{split} &|R_{N,r}^{2}(s)| \\ &= \left| \left\langle S_{N,r}(s), \int_{\mathbb{R}^{d}} d^{d} u \widehat{W}_{N}(\vec{u}) \vec{\nabla} d_{N,r}(.-\vec{u},s) \cdot (\vec{v}_{r}(.,s) - \vec{v}_{r}(.-\vec{u},s)) \right\rangle \right| \\ &\leq \left\langle S_{N,r}(s), \int_{\mathbb{R}^{d}} d^{d} u \widehat{W}_{N}(\vec{u}) |\vec{u}| |\vec{\nabla} d_{N,r}(.-\vec{u},s)| \right\rangle \|\vec{\nabla} \vec{v}_{r}(.,s)\|_{\infty} \\ &\leq C \langle S_{N,r}(s) * U_{N}, |\vec{\nabla} d_{N,r}(.,s)| \rangle \\ &\leq C \left(\widetilde{C} \|S_{N,r}(s) * U_{N}\|_{2}^{2} + \frac{1}{\widetilde{C}} \|\vec{\nabla} d_{N,r}(.,s)\|_{2}^{2} \right), \end{split}$$

Copyright © Marcel Dekker, Inc. All rights reserved



ORDER		REPRINTS
-------	--	----------

with $U_N(\vec{x}) := |\vec{x}| \widehat{W}_N(\vec{x})$. This is true for all $\widetilde{C} > 0$ using Cauchy's inequality. With the second statement of the lemma (20) and (22) we have for all $\epsilon > 0$

$$\begin{split} &|R_{N,r}^{2}(s)| \\ &\leq C\widetilde{C}\left(\hat{\alpha}_{N}^{2\epsilon-2}\|h_{N,r}(.,s)\|_{2}^{2}+\langle S_{N,r}(s),1\rangle^{2}e^{-C'\hat{\alpha}_{N}^{\epsilon}}\right)+\frac{C''}{\widetilde{C}}\|\vec{\nabla}d_{N,r}(.,s)\|_{2}^{2} \\ &\leq C\widetilde{C}\left(\hat{\alpha}_{N}^{2\epsilon-2}(\|d_{N,r}(.,s)\|_{2}^{2}+C')+e^{-C''\hat{\alpha}_{N}^{\epsilon}}\right)+\frac{C'''}{\widetilde{C}}\|\vec{\nabla}d_{N,r}(.,s)\|_{2}^{2}, \end{split}$$

Using (C3), because with the triangle inequality it is

$$\begin{split} \|h_{N,r}(.,s)\|_{2}^{2} &\leq (\|d_{N,r}(.,s)\|_{2} + \|s_{r}(.,s)\|_{2})^{2} \\ &\leq 2\|d_{N,r}(.,s)\|_{2}^{2} + 2\|s_{r}(.,s)\|_{2}^{2}. \end{split}$$

 \widetilde{C} is chosen after the estimate of $\widehat{M}_{N,r}^1$, so that the term arising there and $C'''/\widetilde{C} \|\vec{\nabla}d_{N,r}(.,s)\|_2^2$ cancels with the negative contribution from the estimate of $T_{N,r}^1$. Choosing $\epsilon = \frac{1}{2}$ the constant terms in the above estimate of $|R_{N,r}^2(s)|$ and the prefactor of $\|d_{N,r}(.,s)\|_2^2$ vanish in the limit $N \to \infty$. Now we can write

$$T_{N,r}^{2}(s) = 2\langle d_{N,r}(.,s), (\vec{\nabla}d_{N,r}(.,s)) \cdot \vec{v}_{r}(.,s) \rangle + 2R_{N,r}^{2}(s).$$

With (C6) and the estimate

$$\begin{split} \left| \langle d_{N,r}(.,s), (\vec{\nabla}d_{N,r}(.,s)) \cdot \vec{v}_{r}(.,s) \rangle \right| &= \frac{1}{2} \left| \langle \vec{\nabla}d_{N,r}^{2}(.,s), \vec{v}_{r}(.,s) \rangle \right| \\ &= \left| -\frac{1}{2} \langle d_{N,r}^{2}(.,s), \vec{\nabla} \cdot \vec{v}_{r}(.,s) \rangle \right| \\ &\leq \frac{1}{2} \| \vec{\nabla} \cdot \vec{v}_{r}(.,s) \|_{\infty} \| d_{N,r}(.,s) \|_{2}^{2} \end{split}$$

we get after a suitable arrangement of constants and terms:

$$|T_{N,r}^{2}(s)| \leq C(1+\widetilde{C}) \|d_{N,r}(.,s)\|_{2}^{2} + \frac{C'}{\widetilde{C}} \|\vec{\nabla}d_{N,r}(.,s)\|_{2}^{2} + \widetilde{C}O(N^{-\hat{\beta}/d}),$$

because the *N*-dependent prefactors of $||d_{N,r}(., s)||_2^2$ vanish monotonically with $N \to \infty$ and $N^{-\hat{\beta}/d}$ is the leading order in *N* of all constant terms.

Estimate of
$$T_{N,r}^3(s) = -2\sum_{q=1}^R \left(\langle S_{N,r}(s), (d_{N,r}(.,s) * \widehat{W}_N) a_{N,rq}(.,s) \rangle - \langle s_r(.,s), d_{N,r}(.,s) \hat{a}_{rq}(.,s) s_q(.,s) \rangle \right).$$

2

ORDER		REPRINTS
-------	--	----------

First we make the same substitution as before:

$$\begin{aligned} \langle S_{N,r}(s), (d_{N,r}(.,s) * \widehat{W}_N) a_{N,rq}(.,s) \rangle \\ &= \langle h_{N,r}(.,s), d_{N,r}(.,s) a_{N,rq}(.,s) \rangle + R_{N,rq}^{3a}(s), \end{aligned}$$

and we can estimate the correction term analogously to $R_{N,r}^2$ using (6), (C1), (C2), (C4), (20) and (22):

$$\begin{aligned} |R_{N,rq}^{3a}(s)| &= \left| \left\langle S_{N,r}(s), \int_{\mathbb{R}^{d}} d^{d} u \widehat{W}_{N}(\vec{u}) d_{N,r}(.-\vec{u},s) \right. \\ &\times \left(a_{N,rq}(.,s) - a_{N,rq}(.-\vec{u},s) \right) \right\rangle \right| \\ &\leq \left\langle S_{N,r}(s), \int_{\mathbb{R}^{d}} d^{d} u \widehat{W}_{N}(\vec{u}) |\vec{u}| |d_{N,r}(.-\vec{u},s)| \right\rangle \\ &\times \left(\| \hat{a}_{rq}(.,s) \|_{\infty} \| \vec{\nabla} (S_{N,q}(s) * W_{N}) \|_{\infty} \\ &+ \| \vec{\nabla} \hat{a}_{rq}(.,s) \|_{\infty} \| S_{N,q}(s) * W_{N} \|_{\infty} \right) \\ &\leq \left\langle S_{N,r}(s) * U_{N}, |d_{N,r}(.,s)| \right\rangle \\ &\times C(\alpha_{N}^{d+1} + \alpha_{N}^{d}) (\| W_{1} \|_{\infty} + \| \vec{\nabla} W_{1} \|_{\infty}) \\ &\leq C\alpha_{N}^{2d+2} (\hat{\alpha}_{N}^{2\epsilon-2} (\| d_{N,r}(.,s) \|_{2}^{2} + C') + e^{-C'' \hat{\alpha}_{N}^{\epsilon}}) + \| d_{N,r}(.,s) \|_{2}^{2} \end{aligned}$$

For the first term in the above estimate to vanish in the limit $N \to \infty$, we choose $\epsilon > 0$ so that $\gamma := (\beta(2d+2) + \hat{\beta}(2\epsilon-2))/d < 0$. That means $0 < \epsilon < \frac{1}{\hat{\beta}}(\hat{\beta} - \beta(d+1))$, which is possible due to condition (C1) on the scaling parameters β and $\hat{\beta}$. Now we look at the remaining term in $T_{N,r}^3$:

$$-2\sum_{q=1}^{R} \left(\langle h_{N,r}(.,s), d_{N,r}(.,s)a_{N,rq}(.,s) \rangle - \langle s_{r}(.,s), d_{N,r}(.,s)\hat{a}_{rq}(.,s)s_{q}(.,s) \rangle \right)$$

To contract the two brackets we have to compare the microscopic and macroscopic interaction rates:

$$\langle h_{N,r}(.,s), d_{N,r}(.,s)a_{N,rq}(.,s) \rangle - \langle s_r(.,s), d_{N,r}(.,s)\hat{a}_{rq}(.,s)s_q(.,s) \rangle = \langle d_{N,r}(.,s), d_{N,r}(.,s)a_{N,rq}(.,s) \rangle + R^{3b}_{N,rq}(s),$$

ORDER		REPRINTS
-------	--	----------

with the correction term

$$R_{N,rq}^{3b}(s) = \langle s_r(.,s), d_{N,r}(.,s)(a_{N,rq}(.,s) - \hat{a}_{rq}(.,s)s_q(.,s)) \rangle.$$

With the definition of $a_{N,rq}(., s)$ in (6) and (C5) we get the following estimate:

$$\begin{aligned} |R_{N,rq}^{3b}(s)| \\ &\leq \left\langle s_r(.,s), |d_{N,r}(.,s)||\hat{a}_{rq}(.,s)| \right. \\ &\left. \times \left(|s_q(.,s) - S_{N,q}(s) * W_N| + \delta_{r,q} \frac{1}{N} W_N(\vec{0}) \right) \right\rangle \\ &\leq \|s_r(.,s)\hat{a}_{rq}(.,s)\|_{\infty} \|d_{N,r}(.,s)\|_2 \|s_q(.,s) - S_{N,q}(s) * W_N\|_2 \\ &\left. + \delta_{r,q} N^{\beta-1} W_1(\vec{0})(\|\hat{a}_{rq}(.,s)\|_{\infty}^2 \|s_r(.,s)\|_2^2 + \|d_{N,r}(.,s)\|_2^2) \end{aligned}$$

It is $||s_q(., s) - S_{N,q}(s) * W_N||_2 \le ||(s_q(., s) - S_{N,q}(s)) * W_N||_2 + ||s_q(., s) - s_q(., s) * W_N||_2$ and with the third statement (21) of the lemma

$$\begin{aligned} \|(s_q(.,s) - S_{N,q}(s)) * W_N\|_2 &\leq \|(s_q(.,s) - S_{N,q}(s)) * \widehat{W}_N\|_2 \\ &\leq \|d_{N,q}(.,s)\|_2 + \|s_q(.,s) - s_q(.,s) * \widehat{W}_N\|_2. \end{aligned}$$

Therefore we get with the first statement (19) and (C3)

$$\begin{aligned} |R^{3b}_{N,rq}(s)| &\leq C \left(\|d_{N,r}(.,s)\|_{2}^{2} + \|d_{N,q}(.,s)\|_{2}^{2} + C'(\alpha_{N}^{-2} + \hat{\alpha}_{N}^{-2}) \|\bar{\nabla}s_{q}(.,s)\|_{2}^{2} \right) \\ &+ \delta_{r,q} C'' N^{\beta-1} (1 + \|d_{N,r}(.,s)\|_{2}^{2}). \end{aligned}$$

After treating the correction terms we get for the main contribution using (6):

$$|\langle d_{N,r}(.,s)d_{N,r}(.,s)a_{N,rq}(.,s)\rangle| \le C_a ||d_{N,r}(.,s)||_2^2$$

Arranging all terms analogously to $T_{N,r}^2$ we finally have the estimate

$$|T_{N,r}^{3}(s)| \leq C \sum_{q=1}^{R} ||d_{N,q}(.,s)||_{2}^{2} + O(N^{\gamma}), \text{ with } \gamma < 0.$$

Estimate of $T_{N,r}^4(s) = 2 \sum_{q,l=1}^{R} e_{qlr}(\langle S_{N,q}(s), (d_{N,r}(.,s) * \widehat{W}_N) a_{N,ql}(.,s) \rangle - \langle s_q(.,s), d_{N,r}(.,s) \hat{a}_{ql}(.,s) s_l(.,s) \rangle).$

Copyright @ Marcel Dekker, Inc. All rights reserved.



ORDER		REPRINTS
-------	--	----------

Obviously, the estimate of this term is completely analogous to the one of T_N^3 with the same result except for different constants,

$$|T_{N,r}^4(s)| \le C \sum_{q=1}^R \|d_{N,q}(.,s)\|_2^2 + O(N^{\gamma}), \text{ with } \gamma < 0.$$

Putting all the estimates together, taking the absolute value, the supremum over all $t \in [0, T]$ for some $T < T^*$ (see (C3)) and the sum $\sum_{r=1}^{R}$ on both sides of Eq. (25) we arrive at:

$$\sum_{r=1}^{R} \left(\sup_{t \in [0,T]} \|d_{N,r}(.,t)\|_{2}^{2} + \left(\sigma_{r}^{2} - \frac{C}{\widetilde{C}}\right) \int_{0}^{T} ds \|\vec{\nabla}d_{N,r}(.,s)\|_{2}^{2} \right)$$

$$\leq \sum_{r=1}^{R} \left(\|d_{N,r}(.,0)\|_{2}^{2} + TC' \sup_{t \in [0,T]} \|d_{N,r}(.,t)\|_{2}^{2} \right) + (1 + \widetilde{C} + T)O(N^{\gamma})$$

$$+ \sum_{r=1}^{R} \sup_{t \in [0,T]} \left(2|\widehat{M}_{N,r}^{1}(t)| + 2|\widehat{M}_{N,r}^{2a}(t)| + 2|\widehat{M}_{N,r}^{2b}(t)| + |\widehat{M}_{N,r}^{0b}(t)| + |\widehat{M}_{N,r}^{0b}(t)| \right)$$
(26)

The choice of \tilde{C} below ensures the positivity of all occuring terms. The leading order of all constant terms, including the estimated correction terms, is characterized by $\gamma < 0$ defined in the estimate of $T_{N,r}^3$. Taking the expectation on both sides of (26) we can use the estimates (35) and (36) of the stochastic integrals in Sec. IV.D and get:

$$E\left[\sum_{r=1}^{R} \left(\sup_{t\in[0,T]} \|d_{N,r}(.,t)\|_{2}^{2} + \left(\sigma_{r}^{2} - CN^{(\hat{\beta}-1)/2} - \frac{C'}{\widetilde{C}}\right) \times \int_{0}^{T} ds \|\vec{\nabla}d_{N,r}(.,s)\|_{2}^{2}\right)\right]$$

$$\leq E\left[\sum_{r=1}^{R} \left(\|d_{N,r}(.,0)\|_{2}^{2} + TC'' \sup_{t\in[0,T]} (\|d_{N,r}(.,t)\|_{2}^{2})\right)\right]$$

$$+ (1 + \widetilde{C} + T)O(N^{\gamma}), \qquad (27)$$

where the leading order of constant terms remains unchanged with (C1). Now we can choose \tilde{C} , occuring in the estimate of $T_{N,r}^2$, and $N_0 \in N$ large enough, so that the prefactor of $\int_0^T ds \|\vec{\nabla} d_{N,r}(.,s)\|_2^2$ on the lefthand side of (27) is positive for all $N > N_0$ (see (C7)). Consequently this term can



ORDER		REPRINTS
-------	--	----------

be neglected and after a short rearrangement we have for all $N > N_0$ and 0 < T < 1/C'', where C'' is taken from (27):

$$E\left[\sum_{r=1}^{R} \sup_{t \in [0,T]} \|d_{N,r}(\cdot,t)\|_{2}^{2}\right] \leq \frac{1}{1 - TC''} E\left[\sum_{r=1}^{R} \|d_{N,r}(\cdot,0)\|_{2}^{2}\right] + (1+T)O(N^{\gamma})$$
(28)

Taking the limit $N \to \infty$ on both sides, the constant terms vanish as $\gamma < 0$ and convergence at t = 0 is given in the theorem. This finally proves statement (16) for all $t \in [0, T]$, but the above constraint on the time interval is not essential. At t = T all the conditions for the theorem are fulfilled as long as $T < T^*$, which enables us to apply the proof again with the same constants arising. The length of the next time interval is subject to the same constraint and after a finite number of repetitions this proves the theorem.

C. Proof of the Corollary

We have the following estimate:

$$\begin{split} \left| \left\langle S_{N,r}(t) - s_{r}(\cdot, t), f \right\rangle \right| &\leq \left| \left\langle S_{N,r}(t) - h_{N,r}(\cdot, t), f \right\rangle \right| + \left| \left\langle d_{N,r}(\cdot, t), f \right\rangle \right| \\ &\leq \left| \left\langle S_{N,r}(t), f - f \ast \widehat{W}_{N} \right\rangle \right| + \|f\|_{2} \|d_{N,r}(\cdot, t)\|_{2} \\ &\leq \left\langle S_{N,r}(t), 1 \right\rangle \|f - f \ast \widehat{W}_{N}\|_{\infty} + \|f\|_{2} \|d_{N,r}(\cdot, t)\|_{2} \end{split}$$

Therefore we get for all $f \in \Omega_D$ (see 17), using statement (19) of the lemma and (22): $|\langle S_{N,r}(t) - s_r(\cdot, t), f \rangle| \le C(\|\vec{\nabla}f\|_{\infty}\hat{\alpha}_N^{-1} + \|d_{N,r}(\cdot, t)\|_2)$. Hence we have for the metric D

$$|D(S_{N,r}(t), S_r(t))| \le C(\hat{\alpha}_N^{-1} + ||d_{N,r}(\cdot, t)||_2) \text{ for all } r = 1, \dots, R.$$
(29)

It is straightforward to see that the statement (16) of the theorem implies

$$\lim_{N \to \infty} E \left[\sum_{r=1}^{R} \sup_{t \in [0, T^*]} \| d_{N, r}(., t) \|_2 \right] = 0.$$



ORDER		REPRINTS
-------	--	----------

Using this, Eq. (29) immediately gives

$$\lim_{N \to \infty} E \left[\sum_{r=1}^{R} \left(\sup_{t \in [0, T^*]} D(S_{N,r}(t), S_r(t)) \right) \right]$$

$$\leq C \lim_{N \to \infty} E \left[\sum_{r=1}^{R} \sup_{t \in [0, T^*]} \|d_{N,r}(., t)\|_2 \right] = 0,$$

which proves the statement (18) of the corollary.

D. Estimate of the Stochastic Integrals

In the following we estimate the stochastic integrals occuring in (MI) and (25). First we show for every fixed system size $N \in \mathbb{N}$ that the integrands and integrators fulfill the necessary regularity conditions so that standard techniques of stochastic integration taken from Refs.^[5,7] can be applied. This part is kept short and can be read in more detail in Ref.^[3] (Sec. 3.1.1, appendices B and C), following the work in Ref.^[11]. According to those results we find an estimate uniformly in N using Doob's inequality.^[7] First we consider the terms

$$M_{N,r}^{1}(f,t) = \frac{\sigma_{r}}{N} \sum_{k=1}^{\infty} \sum_{i=1}^{d} \int_{0}^{t} I_{N,r,k,i}^{1}(f,s) dB_{i}^{k}(s)$$

where $I^{1}_{N,r,k,i}(f, t) := \chi_{M(N,r,t)}(k) \partial_{x_i} f(\vec{X}^{k}_{N}(t)),$

$$M_{N,r}^{2a}(f,t) = -\frac{1}{N} \sum_{q=1}^{R} \sum_{k=1}^{\infty} \int_{0}^{t} I_{N,r,k}^{2a}(f,s) dP_{N,rq}^{k}(s)$$

where $I_{N,r,k}^{2a}(f, t) := \chi_{M(N,r,t)}(k)f(\vec{X}_{N}^{k}(t))$ and

$$P_{N,rq}^{k}(t) := a_{N,rq}^{*,k}(t) - \int_{0}^{t} \chi_{M(N,r,s)}(k) a_{N,rq}(\vec{X}_{N}^{k}(s), s) ds$$
(30)

occuring in (MI). $M_{N,r}^{2b}$ can be handled analogously to $M_{N,r}^{2a}$. The integrators B_i^k and $P_{N,rq}^k$ in (30) are Brownian motions and time-inhomogeneous compensated Poisson processes (8) with bounded rates (6). Therefore they are square integrable martingales with respect to the filtration



ORDER		REPRINTS
-------	--	----------

 $(\mathcal{F}_t)_{t\geq 0}$, mentioned after Eq. (3). The integrands $I^1_{N,r,k,i}$ and $I^{2a}_{N,r,k}$ can be replaced by their left continuous versions as we have

$$E\left[\int_{0}^{t} |I_{N,r,k,i}^{1}(f,s) - I_{N,r,k,i}^{1}(f,s-)|^{2}d\langle B_{i}^{k}\rangle(s)\right]$$

= $E\left[\int_{0}^{t} |I_{N,r,k}^{2a}(f,s) - I_{N,r,k}^{2a}(f,s-)|^{2}d\langle P_{N,rq}^{k}\rangle(s)\right] = 0,$

because the quadratic variational processes $\langle B_i^k \rangle(s) = s$ and $\langle P_{N,rq}^k \rangle(s) =$ $\int_0^s \chi_{M(N,r,u)}(k) a_{N,rq}(\vec{X}_N^k(u), u) du \text{ are continuous in } s. \text{ On Poisson processes}$ with time dependent rates see e.g. Ref.^[5] Theorem II.3.1 on page 60. Therefore we can assume the integrands to be predictable processes with respect to $(\mathcal{F}_t)_{t\geq 0}$. For every fixed system size $N \in \mathbb{N}$ it is

$$E\left[\int_{0}^{t}|I_{N,r,k,i}^{1}(f,s)|^{2}d\langle B_{i}^{k}\rangle(s)\right], E\left[\int_{0}^{t}|I_{N,r,k}^{2a}(f,s)|^{2}d\langle P_{N,rq}^{k}\rangle(s)\right] < \infty,$$
(31)

using $f \in C_b^2(\mathbb{R}^d, \mathbb{R})$ and estimates analogous to (32) and (33) below. Hence $M_{N,r}^1(f, t)$ and $M_{N,r}^{2a}(f, t)$ are martingales with respect to $(\mathcal{F}_t)_{t\geq 0}$. On the other hand we have for all $r = 1, \ldots, R, t \geq 0$ and $N \in \mathbb{N}$:

$$\begin{split} E\left[|M_{N,r}^{1}(f,t)|^{2}\right] \\ &= E\left[\frac{\sigma_{r}^{2}}{N^{2}}\left(\int_{0}^{t}\sum_{k\in M(N,r,s)}\vec{\nabla}f(\vec{X}_{N}^{k}(s)).d\vec{B}^{k}(s)\right)\right. \\ &\times \left(\int_{0}^{t}\sum_{t\in M(N,r,s)}\vec{\nabla}f(\vec{X}_{N}^{t}(s)).d\vec{B}^{l}(s)\right)\right] \\ &= \frac{\sigma_{r}^{2}}{N^{2}}E\left[\int_{0}^{t}\sum_{k,t\in M(N,r,s)}\sum_{i,j=1}^{d}\partial_{x_{i}}f(\vec{X}_{N}^{k}(s))\partial_{x_{j}}f(\vec{X}_{N}^{t}(s))d\langle B_{i}^{k},B_{j}^{t}\rangle(s)\right] \\ &= \frac{\sigma_{r}^{2}}{N^{2}}E\left[\int_{0}^{t}\sum_{k\in M(N,r,s)}|\vec{\nabla}f(\vec{X}_{N}^{k}(s))|^{2}ds\right] = \frac{\sigma_{r}^{2}}{N}\int_{0}^{t}E\left[\langle S_{N,r}(s),|\vec{\nabla}f|^{2}\rangle\right]ds \\ &\leq \frac{\sigma_{r}^{2}}{N}\|\vec{\nabla}f\|_{\infty}^{2}\int_{0}^{t}E\left[\langle S_{N,r}(s),1\rangle\right]ds \leq C\frac{\sigma_{r}^{2}}{N}\|\vec{\nabla}f\|_{\infty}^{2}t < \infty \end{split}$$
(32)

ORDER		REPRINTS
-------	--	----------

$$E[|M_{N,r}^{2a}(f,t)|^{2}]$$

$$= \frac{1}{N^{2}} \sum_{p,q=1}^{R} E\left[\int_{0}^{t} \sum_{k,l \in \mathcal{M}(N,r,s)} f(\vec{X}_{N}^{k}(s))f(\vec{X}_{N}^{l}(s))d\langle P_{N,rp}^{k}, P_{N,rq}^{l}\rangle\right]$$

$$= \frac{1}{N^{2}} \sum_{q=1}^{R} E\left[\int_{0}^{t} \sum_{k \in \mathcal{M}(N,r,s)} f^{2}(\vec{X}_{N}^{k}(s))a_{N,rq}(\vec{X}_{N}^{k}(s), s)ds\right]$$

$$\leq \frac{1}{N} \|f\|_{\infty}^{2} \left(\sup_{s \in [0,t]} \sum_{q=1}^{R} \|a_{N,rq}(.,s)\|_{\infty}\right) \int_{0}^{t} E[\langle S_{N,r}(s), 1\rangle]ds$$

$$\leq \frac{RC_{a}}{N} \|f\|_{\infty}^{2} t < \infty$$
(33)

This is true with (22), $f \in C_b^2(\mathbb{R}^d, \mathbb{R})$, and (M4). After an estimate of $M_{N,r}^{2b}$ analogous to $M_{N,r}^{2a}$ we can apply Doob's inequality and get for i = 1, 2a, 2b, r = 1, ..., R and all T > 0:

$$E\left[\sup_{t\in[0,T]}|M_{N,r}^{i}(f,t)|\right]^{2} \leq E\left[\sup_{t\in[0,T]}|M_{N,r}^{i}(f,t)|^{2}\right] \leq 4E\left[M_{N,r}^{i}(f,T)|^{2}\right],$$
(34)

so the result (12) follows from (32) to (34).

For the terms occuring in (25) we have $\widehat{M}_{N,r}^{i}(t) = M_{N,r}^{i}(d_{N,r}(.,t) * \widehat{W}_{N}, t)$ for i = 1, 2a, 2b and therefore a similar reasoning applies in this case. To get an estimate in the limit $N \to \infty$ we note that with (C1) and (C2) $\widehat{W}_{N}(\vec{x} - .)$ is a probability density on \mathbb{R}^{d} for all $\vec{x} \in \mathbb{R}^{d}$. Hence we have for all $f : \mathbb{R}^{d} \to \mathbb{R}$, where $E_{\widehat{W}_{N}}$ denotes the corresponding expectation value:

$$|(f * \widehat{W}_N)(\vec{x})|^2 = \left| \int_{\mathbb{R}^d} \widehat{W}_N(\vec{x} - \vec{y}) f(\vec{y}) d^d y \right|^2 = E_{\widehat{W}_N}[f]^2$$
$$\leq E_{\widehat{W}_N}[f^2] = (|f|^2 * \widehat{W}_N)(\vec{x})$$

We also have with (C1), (C2) and (22) for all $t \in [0, T^*]$ and $r = 1, \dots, R$:

$$\begin{split} \|h_{N,r}(.,t)\|_{\infty} &\leq \hat{\alpha}_{N}^{d} \frac{1}{N} \sum_{k \in \mathcal{M}(N,r,t)} \|W_{1}(\hat{\alpha}_{N}(.-\vec{X}_{N}^{k}(t)))\|_{\infty} \\ &\leq CN^{\hat{\beta}} \langle S_{N,r}(t), 1 \rangle \leq CN^{\hat{\beta}} \end{split}$$

Copyright @ Marcel Dekker, Inc. All rights reserved.



ORDER		REPRINTS
-------	--	----------

With the last two considerations we get for all $T \in [0, T^*]$, starting with the third line of (32) and second line of (33) and using (23),

$$\begin{split} E\left[|\widehat{M}_{N,r}^{1}(T)|^{2}\right] &\leq \frac{\sigma_{r}^{2}}{N} E\left[\int_{0}^{T} \left\langle S_{N,r}(s), |\vec{\nabla}(d_{N,r}(.,s) \ast \widehat{W}_{N})|^{2} \right\rangle ds\right] \\ &\leq \frac{\sigma_{r}^{2}}{N} E\left[\int_{0}^{T} \left\langle h_{N,r}(.,s), |\vec{\nabla}d_{N,r}(.,s)|^{2} \right\rangle ds\right] \\ &\leq CN^{\hat{\beta}-1} \sigma_{r}^{2} E\left[\int_{0}^{T} ||\vec{\nabla}d_{N,r}(.,s)||_{2}^{2} ds\right], \\ E\left[|\widehat{M}_{N,r}^{2a}(T)|^{2}\right] &\leq \frac{1}{N} \sum_{q=1}^{R} E\left[\int_{0}^{T} \left\langle S_{N,r}(s), |d_{N,r}(.,s) \ast \widehat{W}_{N}|^{2} a_{N,rq}(.,s) \right\rangle ds\right] \\ &\leq \frac{RC_{a}}{N} E\left[\int_{0}^{T} \left\langle h_{N,r}(.,s), |d_{N,r}(.,s)|^{2} \right\rangle ds\right] \\ &\leq CN^{\hat{\beta}-1} E\left[\int_{0}^{T} ||d_{N,r}(.,s)||_{2}^{2} ds\right]. \end{split}$$

Using Doob's inequality in the form (34) we get for all $T \in [0, T^*]$:

$$E\left[\sum_{r=1}^{R}\sup_{t\in[0,T]}|\widehat{M}_{N,r}^{1}(t)|\right] \leq CN^{(\hat{\beta}-1)/2}\left(1+\sum_{r=1}^{R}E\left[\int_{0}^{T}\|\vec{\nabla}d_{N,r}(.,s)\|_{2}^{2}ds\right]\right)$$
$$E\left[\sum_{r=1}^{R}\sup_{t\in[0,T]}|\widehat{M}_{N,r}^{2a}(t)|\right] \leq C\left(N^{\hat{\beta}-1}+T\sum_{r=1}^{R}E\left[\sup_{t\in[0,T]}\|d_{N,r}(.,t)\|_{2}^{2}\right]\right).$$
(35)

With (9) the analogous estimate of \widehat{M}_N^{2b} yields the same result as for \widehat{M}_N^{2a} except for the constant *C*. The correction terms $\widehat{M}_{N,r}^{0a}$ and $\widehat{M}_{N,r}^{0b}$ are much easier to handle and with (C1), (C2), (33) and (9) we get

$$E\left[\sum_{r=1}^{R} \left(\sup_{t \in [0,T]} |\widehat{M}_{N,r}^{0a}(t)| + \sup_{t \in [0,T]} |\widehat{M}_{N,r}^{0b}(t)|\right)\right] \le CTN^{\hat{\beta}-3/2}$$
(36)

V. DISCUSSION

A. Connection to Astrophysics

In the astrophysical context studied in Refs.^[6,14] the particles are dust grains in a star forming cloud of hydrogen gas. Depending on the



ORDER		REPRINTS
-------	--	----------

grain size distribution these particles determine the opacity of the gas cloud and influence thermodynamic properties by emission and absorption of heat radiation. They also interact with the gas via hydrodynamic drag and influence chemical reactions via catalysis. A realistic expression for the collision rate of two ball shaped grains with radii l_r and l_q is $\hat{a}_{rq}(\vec{x}, t) \sim (l_r + l_q)^{d-1}g(|\vec{v}_r(\vec{x}, t) - \vec{v}_q(\vec{x}, t)|)$, proportional to the cross section and depending on the relative velocity of the two particles (cf.^[14]). The material coefficient \hat{e}_{rql} should also depend on the relative velocity of the two collision partners. If the latter is high, shattering collisions with several outgoing particles of masses $m_1 < m_r, m_q$ are prefered. If it is low, the colliding particles are more likely to coagulate, so that there is one outgoing particle of mass $m_i = m_r + m_q$. For a precise form of this function from empirical data for different grain materials we refer to Ref.^[6] (Table 1) and references therein.

Of special interest in Refs.^[6,14] is the situation after a shock with very high gas temperatures and particle densities. In this regime shattering is the dominant process, justifying (C4), and it seems natural to assume that all particles are Brownian (C7). The stochasticity in the interaction coming from regularity conditions on W_1 in (M3) also appears to be reasonable. The effective field interaction introduced in (M4) is a simplification we have to make in order to include the space dependence in \hat{a}_{ra} , which cannot be included in a model with pair interactions (see Sec. V.B). The cut-off of the interaction rate in (M4) prevents a divergence due to high particle concentrations and seems natural, as for real grains the density is limited due to the positive particle volume. As the focus of this paper is on the interaction and not on transport terms in (MA), we left out the complicated hydrodynamic drag interaction between particles and gas in (M2). We note that all realistic features are covered by our derivation, except for the velocity dependence of \hat{e}_{ral} , which can be included in a direct generalization explained in the next subsection.

The moderate interaction scaling is technically important, as seen in the proof, and can also be interpreted on a physical level. In the limit $N \to \infty$ the scaling of the mean distance between particles is given by $\Lambda_N \sim N^{-1/d}$ and due to (5) the interaction radius scales like $r_N \sim N^{-\beta/d}$ with $\beta < 1$. It vanishes more slowly with N than Λ_N does and the number of interaction partners of a particle diverges in the limit $N \to \infty$. In contrast to the hydrodynamic scaling with $\Lambda_N \sim r_N$ this introduces a self-averaging effect and the influence of the interaction partners is determined by the local particle density. In the astrophysical gas cloud after a shock the cross section for a collision interaction can be thought of being effectively enlarged by strong Brownian motion of the particles. Together with high particle densities this leads to a large number of

Marcel Dekker, Inc.

270 Madison Avenue, New York, New York 10016

ORDER		REPRINTS
-------	--	----------

interaction partners and justifies the moderate scaling in our microscopic model (M3), whereas the scaling in the Boltzmann–Grad limit^[8] is not appropriate in this regime.

B. Modification of the Microscopic Model

As explained above a modified macroscopic model (MA'), where the material coefficients \hat{e}_{rql} depend on the relative velocity of the collision partners is more realistic. As the microscopic material coefficients $e_{rql} \in \mathbb{N}_0$ are integer numbers they cannot depend on (\vec{x}, t) , because they have to be Lipschitz continuous for our proof (see (C4)). So we define analogous to (6) and (8) a proper set of *m* different, possible collision events.

$$(a_{N,rq}^{*,k})^{i} := \beta_{N,rq}^{k,i} \left(\int_{0}^{t} \chi_{M(N,r,s)}(k) a_{N,rq}^{i}(\vec{X}_{N}^{k}(s), s) ds \right), \quad i = 1, \ldots, m,$$

with rates

$$\begin{aligned} a_{N,rq}^{i}(X_{N}^{k}(t),t) &:= \min \left\{ C_{a}, \hat{a}_{rq}^{i}(X_{N}^{k}(t),t) \right. \\ & \left. \times \left((S_{N,q}(t) * W_{N})(\vec{X}_{N}^{k}(t)) - \delta_{r,q}W_{N}(\vec{0})/N \right) \right\} \end{aligned}$$

and corresponding outcomes e_{rql}^i . So the process with the most probable outcome for the relative velocity of the collision partners can be given the highest rate, whereas the others are small. With this microscopic model (MI') we introduce a dependence of the collision outcome on the relative velocity. To obtain compatibility with the macroscopic model (MA') certain conditions on \hat{a}_{rq}^i and e_{rql}^i have to be satisfied, and our proof of convergence applies with some minor changes.

Another modification of the microscopic model is to include pair interactions. Here we have to define a process for every pair of particles with $k \neq j$:

$$\begin{aligned} a_{N,rq}^{*,kj}(t) &:= \beta_{N,rq}^{kj} \left(\int_0^t a_{N,rq}^{kj}(s) ds \right) & \text{for } k < j \\ a_{N,rq}^{*,kj}(t) &:= a_{N,qr}^{*,jk}(t) & \text{for } k > j, \end{aligned}$$

with rates

$$a_{N,rq}^{kj}(t) := (1 - \delta_{kj})\chi_{M(N,r,t)}(k)\chi_{M(N,q,t)}(j)\frac{1}{N}W_N(\vec{X}_N^k(t) - \vec{X}_N^j(t))\hat{a}_{rq}(t).$$

Our proof of convergence can be applied, but works only for spatially independent macroscopic collision rates $\hat{a}_{rq}(t)$. It assures convergence only up to a stopping time, as long as the summed rates



ORDER		REPRINTS
-------	--	----------

140

Großkinsky, Klingenberg, and Oelschläger

 $\sum_{j \in M(N,r,t)} a_{N,rq}^{kj}(t)$ are bounded uniformly in N by some predefined constant (cf. (M4)). For this reason the first modification seems to be more attractive, as it covers all realistic features explained in Sec. V.A. Nevertheless pair interactions are more realistic descriptions of collision events and coagulation could be included in such a model.

C. Conclusion

In this article we specified a microscopic particle model (MI) from which we rigorously derived Smoluchowski's equation in the space dependent form (MA). Using the technique of the moderate limit developed in Ref.^[11], we could estimate the critical fluctuation terms with martingale inequalities. This scaling assumption is a good approximation of real systems in the regime of high temperatures and particle densities, which has been of interest in the study of interstellar gas clouds after shocks.

In the framework of the moderate limit, a further interesting question is the asymptotic behaviour of fluctuations for large system sizes and the formulation of a central limit theorem for this problem. One can as well try to eliminate some of the technical conditions for the proof, such as the restrictions to the scaling parameter in (C1) or the finite number of particle sizes. There is also hope to proof a convergence result for a microscopic model with pair interactions without the constraints mentioned in V.B, by using a suitable Sobolev-norm.

It would be certainly of most interest to derive Smoluchowski's equation in the hydrodynamic limit, but this task cannot be achieved with the methods used here. Nevertheless we could prove the validity of the spatially inhomogeneous equation in a regime, which is of great interest in astrophysics, and where previous derivations do not supply.

REFERENCES

- Arnold, L.; Theodosopulu, M. Deterministic limit of the stochastic model of chemical reactions with diffusion. Adv. Appl. Prob. 1980, 12, 367–379.
- De Masi, A.; Ferrari, P.A.; Lebowitz, J.L. Reaction-Diffusion equations for interacting particle systems. J. Stat. Phys. 1986, 44, 589–644.
- Großkinsky, S. Herleitung der Smoluchowski-Fragmentations-Gleichung aus einem Vielteilchenmodell im moderaten Limes. Diploma thesis, Institut f
 ür Angewandte Mathematik and Statistik,





ORDER		REPRINTS
-------	--	----------

Julius-Maximilians-Universität Würzburg, 2000. http://www-m5.ma. tum.de/pers/stefang/diplom.ps.gz

- 4. Guias, F. Coagulation-fragmentation processes: Relations between Finite Particle Models and Differential Equations, Preprint 98-41, SFB 359, Ruprechts-Karls-Universität Heidelberg (1998) and Convergence Properties of a Stochastic Model for Coagulation-Fragmentation Processes with Diffusion. Stoch. Anal. Appl. **2001**, *19*, 245–278.
- 5. Ikeda, N.; Watanabe, S. *Stochastic Differential Equations and Diffusion Processes*, 2nd Ed.; North-Holland Publishing Company, Kodansha Ltd.: Tokyo, 1989.
- 6. Jones, A.P.; Tielens, A.G.G.M.; Hollenbach, D.J. Grain shattering in shocks: The interstaller grain size distribution. ApJ **1996**, *469*, 740–764.
- Karatzas, I.; Shreve, S.E. Brownian Motion and Stochastic Calculus, Ist Ed.; Springer: New York, 1988. Itô's formula: Chapter 3, Theorem 3.3 on page 150, Doob's inequality: Chapter 1, Theorem 3.8 on page 14, Usual conditions: Chapter 1, definition 2.25.
- Lang, R.; Xanh, N.X. Smoulchowski's theory of coagulation in colloids holds rigorously in the Boltzmann-Grad-Limit. Z. Wahrscheinlichkeitstheorie verw. Gebiete 1980, 54, 227–280.
- Morale, D.; Capasso, V.; Oelschläger, K. A rigorous derivation of a nonlinear integro-differential equation from a system of stochastic differential equations for an aggregation model. Preprint 98-38, SFB 359, Ruprechts-Karls-Universität Heidelberg (1998).
- Norris, J.R. Smoluchowski's coagulation equation: Uniqueness, non-uniqueness and a hydrodynamic limit for the stochastic coalescent. Ann. Appl. Prob. 1999, 9, 78–109.
- Oelschläger, K. On the derivation of reaction-diffusion equations as limit dynamics of systems of moderately interacting stochastic processes. Probab. Theory Related Fields 1989, 82, 565–586.
- 12. Oelschläger, K. Large systems of interacting particles and the porous medium equation. J. Diff. Equ. **1990**, 88, 294–346.
- Stevens, A. Derivation of chemotaxis-equations as limit dynamics of moderately interacting stochastic many particle systems. SIAM J. Appl. Math. 2000, 61, 183–212.
- Suttner, G.; Yorke, H.W.; Lin, D. Dust coagulation in protostellar envelopes, I. compact grains. ApJ 1999, 524, 857–866.
- van Smoluchowski, M.; Drei Vorträge über Diffusion, Brownsche Bewegung und Koagulation von Kolloidteilchen. Physik. Z. 1916, 17, 557–585.



Request Permission or Order Reprints Instantly!

Interested in copying and sharing this article? In most cases, U.S. Copyright Law requires that you get permission from the article's rightsholder before using copyrighted content.

All information and materials found in this article, including but not limited to text, trademarks, patents, logos, graphics and images (the "Materials"), are the copyrighted works and other forms of intellectual property of Marcel Dekker, Inc., or its licensors. All rights not expressly granted are reserved.

Get permission to lawfully reproduce and distribute the Materials or order reprints quickly and painlessly. Simply click on the "Request Permission/ Order Reprints" link below and follow the instructions. Visit the U.S. Copyright Office for information on Fair Use limitations of U.S. copyright law. Please refer to The Association of American Publishers' (AAP) website for guidelines on Fair Use in the Classroom.

The Materials are for your personal use only and cannot be reformatted, reposted, resold or distributed by electronic means or otherwise without permission from Marcel Dekker, Inc. Marcel Dekker, Inc. grants you the limited right to display the Materials only on your personal computer or personal wireless device, and to copy and download single copies of such Materials provided that any copyright, trademark or other notice appearing on such Materials is also retained by, displayed, copied or downloaded as part of the Materials and is not removed or obscured, and provided you do not edit, modify, alter or enhance the Materials. Please refer to our <u>Website</u> User Agreement for more details.

Request Permission/Order Reprints

Reprints of this article can also be ordered at http://www.dekker.com/servlet/product/DOI/101081SAP120028026