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by

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Selection of asymptotic states through screening induced fluctuations in Ostwald ripening

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Abstract

In this paper we derive a model for the evolution of the particle radius density in the space of radii for a system of many particles that evolve according to the Mullins-Sekerka problem. The derived model is a correction of the classical LSW theory that takes the effect of the fluctuations of the particle density into account. The main difference between the model derived in this paper and the classical LSW theory is the presence of a second order term yielding a boundary layer effect for large particles. In particular this model provides a possible solution for the so-called "selection problem" in the LSW theory.

Key words: Kinetics of phase transitions, domain coarsening, fluctuations of large particles

1 Introduction

Ostwald ripening denotes the late stage coarsening of heterogeneously nucleated particles within a first order phase transition. If the particle phase is very dilute, one can use the classical theory by Lifshitz, Slyozov and Wagner (LSW) [6, 16] to describe the evolution of the distribution of particle radii by a mean-field equation. The model is based on the assumption that each individual particle interacts with all surrounding particles only by some average mean-field which is the same for all the particles. The LSW model has a scale invariance and a family of self-similar solutions which all predict a rate of growth for the average particle radius of the form $\langle R\rangle \sim Ct^{1/3}$. While it has been predicted in [6, 16] that only one of these self-similar solutions is stable, it is by now well-known [3],[11] that the asymptotic behavior of solutions depends sensitively on the initial data. More precisely it depends on the largest particles, and even non-self-similar asymptotics can appear for certain types of data.

This lack of a selection criterion of self-similar solutions and even more so a significant discrepancy with experiments, which shows larger coarsening rates and broader size distributions than the mean-field theory [15], was the motivation to investigate additional effects which have not been taken into account in the LSW model.

In [9, 14] diffusive effects in the particle sizes due to nucleation of particles are taken into account, which yields via an asymptotic analysis a selection of the LSW solution as the only possible self-similar state. In [2] an asymptotic analysis of the different time regimes in a Becker-Döring model is performed, which predicts a quite narrow size distribution as initial data for the coarsening regime. However, such a theory does not account for the effect of positive volume fraction of particles, which is commonly believed to be responsible for the deviation of the predictions with experimental data.

In [7] a perturbative theory, in the following referred to as Marder's theory, has been developed, which takes the build up of correlations between particles in systems with positive volume fraction

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into account. This theory has also been rederived in a mathematically more rigorous way in [4]. However, as it is pointed out in the review article [10], this theory is not self-consistent since it assumes that correlations between particles are uniformly small. Such an assumption, even if true initially, does not remain satisfied during the evolution for the largest particles in the system.

Thus, the effect of pair correlations between the largest particles has to be studied by different methods, introducing a suitable boundary layer. In this paper we present a method that allows to derive a corresponding model, which will consist of the LSW model plus an additional diffusive type term which is only relevant for the largest particles. The model is self-consistent and it provides a selection criterion for a self-similar solution which is a perturbation of the self-similar solution singled out by LSW.

An effect, which will not be considered in the present paper, but has already been discussed in [6], is the fact that particles can come close to each other, which leads to so called "encounters". Then the particles merge and form a larger particle. In [6] this effect is taken into account in an ad-hoc manner by an additional term in the LSW model which is of the form of a coagulation term with additive kernel. A mathematical analysis of this model will be presented elsewhere, however, it turns out, that in fact this mechanism provides a selection criterion for self-similar states and the induced deviation to the mean-field theory is indeed much larger than the one given by Marder's theory [7] or the model considered in the present paper. However, a self-consistent derivation of a model with encounters from the Mullins-Sekerka evolution does to our knowledge not exist yet.

In the following Section 2 we first present the starting point of our analysis, which is a simplified Mullins-Sekerka evolution for spherical particles. We briefly review the main aspects of the LSW theory and give a brief account of Marder's theory. We also refer to the review article [10] for a more exhaustive summary of the derivation of the theory, its advantages and disadvantages and for further references. Section 3 is the main part of this paper, which contains a derivation of the model which takes fluctuations of largest particles into account. The final result is presented in Section 3.6. In the last Section 4 we show, that the model has a self-similar solution which is a perturbation of the LSW self-similar solution with a Gaussian tail.

2 Basic concepts

2.1 Evolution equations

The starting point of our analysis is the so-called Mullins Sekerka problem

$$\Delta u = 0 \ , \ \ x \in \Omega \setminus \bigcup_i B_{R_i}(x_i) \tag{2.1}$$

$$u = \frac{1}{R_i} \ , \ \ x \in \partial B_{R_i}(x_i) \tag{2.2}$$

$$V_n = \frac{\partial u}{\partial n} \ , \ \ x \in \partial B_{R_i}(x_i) \tag{2.3}$$

where, $\Omega \subset \mathbb{R}^3$, n is the outer normal, x_i is the center of the particle i and R_i is its radius.

Throughout this paper we will consider the case that the volume fraction of the particles, denoted by ϕ, is small, that is $\phi \ll 1$. The evolution under the set of equations (2.1)-(2.3) does not preserve the position of the center of the particles or its sphericity. However, in the case of small volume fraction it has been shown in [1] that these are effects of higher order than considered in this paper (cf. also [7], where an argument is given that the error is of order $\phi^{2/3}$). This justifies to replace (2.3) by

$$\dot{R}_i = \frac{1}{|\partial B_{R_i}(x_i)|} \int_{\partial B_{R_i}(x_i)} \frac{\partial u}{\partial n} dS_x \tag{2.4}$$

For definiteness we assume from now on that Ω in (2.1) is the unit cube enclosing the particles under consideration and that u satisfies periodic boundary conditions.

The model (2.1), (2.2), (2.4) is equivalent to a system of ODEs that we can write as

$$\frac{dx_i}{dt} = 0 \tag{2.5}$$

$$\frac{dR_i}{dt} = -\frac{1}{4\pi (R_i)^2} \sum_{j=1}^{N} \frac{C_{j,i}}{R_j} \tag{2.6}$$

where $C_{j,i}$ are the electrostatic capacity coefficients (see e.g. [5]) defined as

$$C_{j,i} := -\int_{\partial B_{R_j}(x_i)} \frac{\partial v_i}{\partial n} dS_x \tag{2.7}$$

where v_i is the solution of

$$\Delta v_i = 0 \ \ , \ \ x \in \Omega \setminus \bigcup_i B_{R_i}(x_i) \tag{2.8}$$

$$v_i = \delta_{i,j} \ \ , \ \ x \in \partial B_{R_j}(x_i) \tag{2.9}$$

with periodic boundary conditions on $\partial\Omega$.

The capacity coefficients $C_{j,i}$ are functions of the positions and radii of all the particles of the system

$$C_{j,i} = C_{j,i}(x_1, R_1, x_2, R_2, ..., x_N, R_N) \tag{2.10}$$

and satisfy the following properties

$$C_{i,i} > 0, \qquad C_{i,j} < 0 \ \text{ if } \ i \neq j, \qquad C_{i,j} = C_{j,i}\,. \tag{2.11}$$

Moreover, integrating (2.8) over $\Omega \setminus \bigcup_i B_{R_i}(x_i)$, using Green's formula and the periodic boundary conditions, we obtain

$$\sum_{j=1}^{N} C_{i,j} = 0 \qquad \text{for all } i = 1, \ldots, N\,. \tag{2.12}$$

Particles might dissappear in finite time and the evolution of the system after those events must be described in order to completely determine the dynamics of the system. We just eliminate the vanishing particles and continue with the evolution of the remaining ones. Another singular event that can take place is the collision of two or more particles. However, the fraction of particles involved in collisions is small (cf. [12]) and we do not consider this effect in the present paper. As pointed out in the introduction though, the effect on the long-time behavior might still be large.

2.2 Stochastic initial data

We will assume that the initial values for the variables (x_i, R_i) are prescribed by a probability measure of the form

$$d\nu(x_{1,0}, R_{1,0}, x_{2,0}, R_{2,0}, ..., x_{N,0}, R_{N,0}) \equiv \prod_{k=1}^{N} f_{0,N}(R_{k,0})\, dx_{k,0} dR_{k,0} \tag{2.13}$$

where $f_{0,N}$ is a nonnegative probability density with compact support. (For the normalization recall also that $|\Omega| = 1$.)

We assume that all the particles have a similar order of magnitude r_N, where

$$r_N = \langle R_0 \rangle \equiv \int_0^\infty R f_0(R)\, dR \tag{2.14}$$

is the average radius.

We can now formulate the precise problem that we will consider in the rest of the paper. Our goal is to study the solution of the system of ODEs (2.5), (2.6) where $C_{j,i}$ is as in (2.7)-(2.9) and the initial data $x_{1,0}, R_{1,0}, x_{2,0}, R_{2,0}, ..., x_{N,0}, R_{N,0}$ are chosen randomly according to the measure (2.13) with $f_{0,N}$ as in (2.13), where $r_N \to 0$ as $N \to \infty$ and the volume fraction $\phi := N(r_N)^3$ is small but fixed.

2.3 Screening length and approximation of $C_{i,j}$

A crucial length scale in the study of Ostwald ripening is the concept of the screening length that was introduced in the context of this problem in [8] and is similar to the classical Debye-Hückel screening length. It can be understood as follows. Suppose that we release a Brownian particle at a point x_0 in a perforated domain $\mathbb{R}^3 \setminus \bigcup_i B_{R_i}(x_i)$ with trapping boundaries $\partial B_{R_i}(x_i)$. The screening length ξ_N is a characteristic distance that measures how far the Brownian particle diffuses before being trapped in some of the boundaries $\partial B_{R_i}(x_i)$. In the limit of small average radius r_N and for small volume fraction ϕ a convenient measure of the screening length is

$$\xi = \frac{1}{\sqrt{4\pi N \langle R \rangle}} \tag{2.15}$$

Observe, that in Ostwald ripening, the average radius $\langle R \rangle$, the number density N and consequently also the screening length ξ depend on time.

One way of deriving (2.15) heuristically can be taken from electrostatics. Consider a point charge at a point $x_0 = 0$ in a sea of conducting balls $B_{R_i}(x_i)$ of small volume fraction which are homogeneously distributed in space with a number density N. The point charge at 0 creates an electric potential G and induces a negative charge on the boundary of the balls. This induced charge roughly equals $-4\pi R_i G(x_i)$, where $4\pi R_i$ is the capacity of a single particle in $\mathbb{R}^3$. In a dilute system of balls the capacity of the particles is approximately additive whence the total negative charge is approximately $-4\pi N \langle R \rangle G(x)$. Hence, the electric potential satisfies approximately the equation

$$-\Delta G(x) = \delta(x) - 4\pi N \langle R \rangle G(x) \tag{2.16}$$

whose explicit solution is given by

$$G(x) = \frac{e^{-\frac{|x|}{\xi}}}{4\pi |x|} \tag{2.17}$$

Equation (2.16) is the basic screening equation that allows to measure the effect of one particle on the surrounding ones. In [4] it has been shown, that for independently distributed particles, the error between (2.17) and the exact electric potential is of order $\phi^{1/2}$. In principle, the argument is valid only in the whole space. However, we are interested in the case where the screening length is smaller than the domain size, and then the argument is also valid (see also [13]).

If we use the approximation (2.17) for the solution of (2.8)-(2.9), that is $v_i(x) = \frac{R_i e^{-\frac{|x-x_i|}{\xi}}}{|x-x_i|}$, we find

$$C_{j,i} = -\frac{4\pi R_j R_i e^{-\frac{|x_j - x_i|}{\xi}}}{|x_j - x_i|} \quad , \quad j \neq i\,, \tag{2.18}$$

while to leading order we can approximate $C_{i,i}$ by the formula of the electrostatic capacity of an sphere in the whole space, that i

$$C_{i,i} = 4\pi R_i \,. \tag{2.19}$$

2.4 Evolution of statistical distributions

As indicated in Subsection 2.2 the initial distribution of particles is prescribed using the probability measure (2.13). The Liouville equation for the distribution density D_N of particles is given by

$$\frac{\partial D_N}{\partial t} + \sum_{i=1}^{N} \left[\frac{\partial}{\partial x_i} \left(\dot{x}_i D_N \right) + \frac{\partial}{\partial R_i} \left(\dot{R}_i D_N \right) \right] = 0 \ ,$$

or using (2.5) and (2.6) by

$$\frac{\partial D_N}{\partial t} - \frac{1}{4\pi} \sum_{i=1}^{N} \sum_{j=1}^{N} \left[\frac{\partial}{\partial R_i} \left(\frac{C_{j,i}}{(R_i)^2 R_j} D_N \right) \right] = 0 \,. \tag{2.20}$$

The initial data $D_N(\cdot,0) = D_N(x_{1,0}, R_{1,0}, x_{2,0}, R_{2,0}, ..., x_{N,0}, R_{N,0}, 0)$ are given by

$$\begin{aligned} & d\nu\left(x_{1,0}, R_{1,0}, x_{2,0}, R_{2,0}, ..., x_{N,0}, R_{N,0}\right) \\ & \quad = \frac{D_N\left(x_{1,0}, R_{1,0}, x_{2,0}, R_{2,0}, ..., x_{N,0}, R_{N,0}, 0\right)}{(N)!} \prod_{k=1}^{N} \left[dx_{k,0} dR_{k,0} \right] \ , \end{aligned} \tag{2.21}$$

or, equivalently,

$$D_N\left(x_1, R_1, x_2, R_2, ..., x_N, R_N, 0\right) = \frac{N!}{N^N} \prod_{k=1}^{N} \left[f_{0,N}\left(R_k\right) \right] \tag{2.22}$$

Notice that with this choice of $D_N(\cdot,0)$ we have the normalization

$$\int D_N(\eta, t)\, d^N \eta = N! \tag{2.23}$$

where from now on we use the abbreviations

$$\eta_j = (x_j, R_j) \ , \quad \eta = (\eta_1, \eta_2, ..., \eta_N) \ , \qquad d\eta_j = dx_j dR_j \ , \quad d^N \eta = \prod_{j=1}^{N} d\eta_j \,.$$

The motivation for the normalization (2.23) is that we want to compute particle densities instead of probability densities. Therefore D_N is the density in the space of ordered $N-$tuples $\left(\eta_{i(1)}, \eta_{i(2)}, \eta_{i(3)}, ..., \eta_{i(N)}\right)$ where $\{i(1), i(2), ..., i(N)\}$ is a permutation of the integers $\{1, 2, ..., N\}$.

We define the distribution functions for s particles by

$$f_s\left(\eta_1, \eta_2, ..., \eta_s, t\right) = \frac{1}{(N-s)!} \int D_N(\eta, t)\, d\eta_{s+1} d\eta_{s+2} ... d\eta_N \ , \quad s = 1, 2, ... N \tag{2.24}$$

that due to the normalization condition satisfy

$$\int f_s\left(\eta_1, \eta_2, ..., \eta_s, t\right) d\eta_1 d\eta_2 ... d\eta_s = \frac{N!}{(N-s)!} \,, \tag{2.25}$$

such that in particular $\int f_1 d\eta_1 = N$. Integrating (2.20) with respect to the variables $\eta_{s+1}, \eta_{s+2}, ..., \eta_N$ and using (2.24) we obtain

$$\frac{\partial f_s}{\partial t} - \frac{1}{4\pi} \sum_{i=1}^{s} \frac{\partial}{\partial R_i} \left(\frac{1}{(R_i)^2} \left[\left(\frac{1}{(N-s)!} \int \left(\sum_{j=1}^{N} \frac{C_{j,i}}{R_j} \right) D_N d\eta_{s+1} d\eta_{s+2} ... d\eta_N \right) \right] \right) = 0 \qquad (2.26)$$

for $s = 1, 2, ...N$.

2.5 LSW theory

The LSW theory provides a closed equation for the one-particle distribution function f_1. It is based on the assumption that the measure D_N is approximately uncorrelated during the whole evolution of the system, i.e. that it has the form

$$D_N(\eta_1, \eta_2, ..., \eta_s, t) = \prod_{k=1}^{N} [f_1(R_k, t)] \, . \qquad (2.27)$$

That f_1 is independent of x is due to the fact that the system is homogeneous. Now we use (2.26) for $s = 1$ and (2.27) to find

$$\frac{\partial f_1(R_1, t)}{\partial t} - \frac{1}{4\pi} \frac{\partial}{\partial R_1} \left(\frac{1}{(R_1)^2} \frac{C_{1,1}}{R_1} - \frac{1}{R_1^2} \int \frac{C_{1,2}}{R_2} f_1(R_2, t) \, dx_2 \, dR_2 \right) = 0. \qquad (2.28)$$

Employing the approximations (2.18) and (2.19), we find, due to

$$\int \frac{e^{-\frac{|x_1 - x_2|}{\xi}}}{|x_1 - x_2|} dx_2 \sim 4\pi\xi^2 \, ,$$

and the relation $4\pi\xi^2 = \frac{1}{N\langle R \rangle}$ implies that

$$\frac{\partial f_1(R_1, t)}{\partial t} + \frac{\partial}{\partial R_1} \left(\left(-\frac{1}{(R_1)^2} + \frac{1}{\langle R \rangle R_1} \right) f_1(R_1, t) \right) = 0 \, , \qquad (2.29)$$

which is just the classical LSW model. It is well known that it admits a family of self-similar solutions of the form

$$f_1(R_1, t) = \phi \frac{1}{t^{4/3}} \Phi_\gamma \left(\frac{R_1}{t^{1/3}} \right) , \qquad \langle R \rangle = (\gamma t)^{1/3} \qquad (2.30)$$

with $\gamma \in (0, \frac{4}{9}]$. Each of the profiles Φ_γ has compact support, the largest support for $\gamma = \frac{4}{9}$. For each $\gamma \in (0, \frac{4}{9})$ there is a unique $p \in (-1, \infty)$ such that Φ_γ behaves like a polynomial of power p at the end of its support, whereas for $\gamma = \frac{4}{9}$ we obtain

$$\Phi_{LSW} := \Phi_{\frac{4}{9}}(\rho) = C \frac{\rho^2 \exp[\frac{-\rho}{\rho_{LSW} - \rho}]}{\left(1 + \frac{\rho}{2\rho_{LSW}}\right)^{7/3} \left(1 - \frac{\rho}{\rho_{LSW}}\right)^{11/3}}, \qquad \text{for } 0 \leq \rho \leq \rho_{LSW} = \left(\frac{3}{2}\right)^{1/3},$$

where C is a normalization constant such that $\frac{4\pi}{3} \int \rho^3 \Phi_\gamma(\rho) \, d\rho = 1$. We call this solution Φ_{LSW} since it was singled out by LSW as the unique stable self-similar solution. While Wagner rules out – seemingly by some numerical error – the existence of solutions for $\gamma < \frac{4}{9}$, Lifshitz and Slyozov realized their existence, but argued that only Φ_{LSW} would be stable. The argument includes additional regularization terms by accounting for encounters of particles.

After a lively discussion in the applied literature in the end of the eighties, it was predicted in [3] by numerical simulation and shown rigorously in [11] that all self-similar solutions can appear as the large time limit of (2.29). Roughly speaking, a solution converges to the self-similar solution with a certain power law at the end of its support if the initial data have the same polynomial behavior (more precisely, if they are regularly varying with the same power) at the end of their support.

2.6 Marder's theory

In [7] Marder considers the BBGKY-hierarchy for the N-particle distribution function. Under a closure assumption a closed system of equations for f_1 and f_2 is derived, which takes the build up of correlations between particles into account. The same model has been rederived under a natural closure assumption in a mathematically more rigorous way in [4] (see Section 3.2.2). The assumption is that the N-particle distribution is given by a cluster expansion, in which pair correlations are of order $\phi^{1/2}$ and higher order correlations are even smaller. It is easily checked that the model is self-consistent in a regime where $f_1(R)$ is of order 1. However, it was realized later (cf. e.g. [10]), that the model is not self-consistent for the largest particles where $f_1(R)$ is small. For the largest particles correlations become of order 1 during the evolution, even if they are small for the initial data. Thus, a boundary layer appears for the largest particles in the system and it is not enough to study the hierarchy of distribution functions or, equivalently, of the correlation functions. In the next section we will describe how one can correct the LSW model in order to take this effect into account.

3 A correction to the mean-field model for large particles

3.1 Defining a formal evolution for extinct particles

It is more convenient to work with a system containing a fixed number of particles, in order to avoid handling distribution functions with a changing number of variables. To this end we define artificially the evolution of the particles that vanish during the evolution of the system. The evolution of nonextinct particles is given by (2.5), (2.6). We define the evolution of the extinct particles by

$$\frac{dR_i}{dt} = -\frac{1}{(R_i)^2} + \frac{1}{\langle R \rangle R_i} \quad , \quad i = 1, ..., N \tag{3.1}$$

$$\frac{dx_i}{dt} = 0 \quad , \quad i = 1, ..., N \tag{3.2}$$

Notice, that (3.1) implies that if $R_i(t_*) \leq 0$ for some t_*, that then $R_i(t) < 0$ for all $t > t_*$.

We will also assume that a missing particle does not interact with the remaining ones, or equivalently

$$C_{i,j} = 0 \quad , \quad i \neq j \tag{3.3}$$

if $R_i > 0$ and $R_j \leq 0$.

From the physical point of view extinct particles are important because during their life span they contribute to the "noise" that influences the evolution of the surviving particles. Equations (3.1), (3.2) will keep track of this effect. However, there are other methods of introducing this physical effect in the model. The definition of the artificial evolution (3.1), (3.2) is just a convenient mathematical trick.

In the rest of this section we will describe the evolution of the system of particles whose initial distribution

$$R_i(0) = R_{i,0}\,, \qquad\qquad x_i(0) = x_{i,0} \tag{3.4}$$

is determined by means of the density function (2.22) and where the particles evolve by means of the differential equations (2.5), (2.6), (3.1) and (3.2). Notice that all the arguments in Subsection 2.4 might be applied to this problem.

3.2 A closure relation

A key ingredient in our analysis will be a certain closure relation which provides an approximation of the two-particle distribution function f_2 by f_1 evaluated at a suitable shift in R_1 plus an additional term which will turn out to be neglible in the self-similar regime. In this subsection we will derive this closure relation. The main task in the following subsections will be to explicitly compute the shift to leading order in terms of f_1.

We recall that f_1 and f_2 are given by

$$f_1(\eta_1, t) = \frac{1}{(N-1)!}\int D_N(\eta_1, \eta_2, \eta_3, ..., \eta_N, t)\, d\eta_2 d\eta_3 ... d\eta_N\,, \tag{3.5}$$

$$f_2(\eta_1, \eta_2, t) = \frac{1}{(N-2)!}\int D_N(\eta_1, \eta_2, \eta_3, ..., \eta_N, t)\, d\eta_3 d\eta_4 ... d\eta_N \tag{3.6}$$

where $D_N(\eta_1, \eta_2, \eta_3, ..., \eta_N, t)$ solves the Liouville equation (2.20) and its initial data are given by (2.22). For further reference also recall that

$$\langle R\rangle = \langle R\rangle(t) = \frac{\int_0^\infty R_1 f_1(R_1)\, dR_1}{\int_0^\infty f_1(R_1)\, dR_1} \qquad \text{and} \qquad \xi^2(t) = \frac{1}{4\pi\langle R\rangle \int_0^\infty f_1(R_1)\, dR_1}. \tag{3.7}$$

We now introduce two sets of "Eulerian" variables that allow to integrate the Liouville equation (2.20). More precisely we define a new set of variables

$$\eta_{k,0} = \eta_{k,0}(\eta_1, \eta_2, \eta_3, ..., \eta_N, t) \quad , \quad k = 1, ..., N \tag{3.8}$$

that are the initial values for the characteristic equations of the Liouville equation (2.5), (2.6). The solution of the Liouville equation (2.20) can be written in terms of these new variables as

$$\begin{aligned} D_N(\eta_1, \eta_2, \eta_3, ..., \eta_N, t) &= D_N(\eta_{1,0}, \eta_{2,0}, \eta_{3,0}, ..., \eta_{N,0}, 0)\frac{\partial(\eta_{1,0}, \eta_{2,0}, \eta_{3,0}, ..., \eta_{N,0})}{\partial(\eta_1, \eta_2, \eta_3, ..., \eta_N)} \\ &= \frac{(N)!}{N^N}\prod_{k=1}^{N}\left[f_{0,N}(R_{k,0})\right]\frac{\partial(\eta_{1,0}, \eta_{2,0}, \eta_{3,0}, ..., \eta_{N,0})}{\partial(\eta_1, \eta_2, \eta_3, ..., \eta_N)} \end{aligned} \tag{3.9}$$

With the changes of variables

$$(\eta_1, \eta_2, \eta_3, ..., \eta_N) \to (\eta_1, \eta_{2,0}, \eta_{3,0}, ..., \eta_{N,0})$$
$$(\eta_1, \eta_2, \eta_3, ..., \eta_N) \to (\eta_1, \eta_2, \eta_{3,0}, ..., \eta_{N,0})$$

we can rewrite (3.5) and (3.6), using (3.9), in the limit $N \to \infty$ as

$$f_1(\eta_1, t) = \frac{1}{N}\int\prod_{k=1}^{2}\left[f_{0,N}(R_{k,0})\right]\frac{\partial(\eta_{1,0})}{\partial(\eta_1)}d\eta_{2,0}d\nu_N \tag{3.10}$$

$$f_2(\eta_1, \eta_2, t) = \int\prod_{k=1}^{2}\left[f_{0,N}(R_{k,0})\right]\frac{\partial(\eta_{1,0}, \eta_{2,0})}{\partial(\eta_1, \eta_2)}d\nu_N \tag{3.11}$$

where

$$d\nu_N \equiv \frac{1}{N^{N-2}} \prod_{k=3}^{N} \left[f_{0,N}\left(R_{k,0}\right)\right] d\eta_{3,0} d\eta_{4,0}...d\eta_{N,0} \tag{3.12}$$

From now on we will write for simplicity

$$\omega_{0,N} = \left(\eta_{3,0}, \eta_{4,0}..., \eta_{N,0}\right)$$

We define two functions $R_{1,0},\ R_{2,0}$ defined as the values of the initial radii R_1 and R_2 for particles characterized by the values η_1 and η_2 at time t. These functions depend also on the initial positions of the remaining particles $\omega_{0,N}$, so that

$$R_{1,0} = R_{1,0}\left(\eta_1, \eta_2, \omega_{0,N}, t\right)$$
$$R_{2,0} = R_{2,0}\left(\eta_1, \eta_2, \omega_{0,N}, t\right)$$

Using the functions $R_{1,0}$ and $R_{2,0}$ we can rewrite (3.11) as

$$f_2\left(\eta_1, \eta_2, t\right) = \int \prod_{k=1}^{2} \left[f_{0,N}\left(R_{k,0}\left(\eta_1, \eta_2, \omega_{0,N}, t\right)\right)\right] \frac{\partial\left(R_{1,0}, R_{2,0}\right)}{\partial\left(R_1, R_2\right)} d\nu_N \tag{3.13}$$

In the following we denote by $R_{1,0}^{(2)} = R_{1,0}^{(2)}(\eta_1, \omega_{0,N}, t)$ the function $R_{1,0}$ in a system where particle 2 has been removed. Correspondingly we define $R_{2,0}^{(1)}$.

In order to compute $f_2\left(\eta_1, \eta_2, t\right)$ for particles η_1 and η_2 which are placed within the screening radius we introduce two functions $U_i = U_i\left(\eta_1, \eta_2, \omega_{0,N}, t\right)$, $i = 1, 2$ via

$$R_{1,0}\left(\eta_1, \eta_2, \omega_{0,N}, t\right) = R_{1,0}^{(2)}\left(\eta_1 + U_1, \omega_{0,N}, t\right)\ , \tag{3.14}$$

$$R_{2,0}\left(\eta_1, \eta_2, \omega_{0,N}, t\right) = R_{2,0}^{(1)}\left(\eta_2 + U_2, \omega_{0,N}, t\right)\ , \tag{3.15}$$

where we use the notation $\eta_i + U_i = \left(R_i + U_i, x_i\right)\ ,\ i = 1, 2$. Notice that $U_i \to 0$ if $|x_1 - x_2| >> \max_{\{0 \leq s \leq t\}} \xi\left(s\right)$. Combining (3.13), (3.14) and (3.15) we obtain

$$\begin{aligned} f_2\left(\eta_1, \eta_2, t\right) = \int \prod_{k=1}^{2} \left[f_{0,N}\left(R_{k,0}^{(\tau_k)}\left(\eta_k + U_k, \omega_{0,N}, t\right)\right)\right] \\ \cdot \frac{\partial\left(R_{1,0}^{(2)}\left(\eta_1 + U_1, \omega_{0,N}, t\right), R_{2,0}^{(1)}\left(\eta_2 + U_2, \omega_{0,N}, t\right)\right)}{\partial\left(R_1, R_2\right)} d\nu_N \end{aligned} \tag{3.16}$$

where we use the notation $\tau_1 = 2$ and $\tau_2 = 1$.

In Section 3.3 we will show that the terms U_i have a relative size of order $\sqrt{\phi}$. Moreover, it turns out that to leading order the functions U_i depend only on η_1, η_2 and t, but not on $\omega_{0,N}$. Then we can rewrite (3.16), keeping only the terms up to order $\sqrt{\phi}$, as

$$\begin{aligned} & f_2\left(\eta_1, \eta_2, t\right) \\ & = \left[1 + \sum_{j=1}^{2} \frac{\partial U_j}{\partial R_j}\right] \int \prod_{k=1}^{2} \left[f_{0,N}\left(R_{k,0}^{(\tau_k)}\left(\eta_k + U_k, \omega_{0,N}, t\right)\right) \frac{\partial R_{k,0}^{(\tau_k)}\left(\eta_k + U_k, \omega_{0,N}, t\right)}{\partial\left(R_k + U_k\right)}\right] d\nu_N \end{aligned} \tag{3.17}$$

We integrate (3.17) with respect to R_2 and neglect lower order terms to find

$$\int f_2\left(\eta_1, \eta_2, t\right) dR_2 = \int \left(\int \prod_{k=1}^{2} F\left(\eta_k + U_k, \omega_{0,N}, t\right) d\nu_N\right) dR_2 \tag{3.18}$$

where

$$F\left(\eta_k,\omega_{0,N},t\right)=f_{0,N}\left(R_{k,0}^{(\tau_k)}\left(\eta_k,\omega_{0,N},t\right)\right)\frac{\partial R_{k,0}^{(\tau_k)}\left(\eta_k,\omega_{0,N},t\right)}{\partial R_k} \tag{3.19}$$

We are going to derive a second-order evolution equation for f_1 (see (3.71) below), where the second-order term will only play a relevant role in a small boundary layer. Therefore, such boundary layers will give a negligible contribution in the integration with respect to the R_2 variable and it is possible to approximate (3.18) to leading order by

$$\int f_2\left(\eta_1,\eta_2,t\right)dR_2=\int\left(\int F\left(\eta_1+U_1,\omega_{0,N},t\right)F\left(\eta_2,\omega_{0,N},t\right)d\nu_N\right)dR_2$$

Using (3.18) and that $F(\eta_2,\omega_{0,N},t)$ is a stochastic variable with average $f_1(\eta_2,t)$ with respect to the measure $d\nu_N$, we find

$$\begin{aligned}
&\int\left(\int F\left(\eta_1+U_1,\omega_{0,N},t\right)F\left(\eta_2,\omega_{0,N},t\right)d\nu_N\right)dR_2\\
&=\int\left(\int f_1\left(\eta_1+U_1,t\right)f_1\left(\eta_2,t\right)d\nu_N\right)dR_2+\\
&\int\left(\int\left[F\left(\eta_1+U_1,\omega_{0,N},t\right)-f_1\left(\eta_1+U_1,t\right)\right]\left[F\left(\eta_2,\omega_{0,N},t\right)-f_1\left(\eta_2,t\right)\right]d\nu_N\right)dR_2
\end{aligned}$$

We will show in the next section that the relative size of U_1 is of order $\phi^{1/2}$. Hence we have to leading order that

$$\begin{aligned}
&\int\left(\int F\left(\eta_1+U_1,\omega_{0,N},t\right)F\left(\eta_2,\omega_{0,N},t\right)d\nu_N\right)dR_2\\
&=\int f_1\left(\eta_1+U_1,t\right)f_1\left(\eta_2,t\right)dR_2+\\
&\int\left(\int\left[F\left(\eta_1,\omega_{0,N},t\right)-f_1\left(\eta_1,t\right)\right]\left[F\left(\eta_2,\omega_{0,N},t\right)-f_1\left(\eta_2,t\right)\right]d\nu_N\right)dR_2
\end{aligned} \tag{3.20}$$

The right hand side of (3.20) consists of two different types of terms. The first one measures the change of the radius of particle η_1 due to the presence of the particle η_2 and will be computed in the next Subsection 3.3. The second one comes from the fluctuations of F and will be computed in Subsection 3.4.

3.3 Computing $U_1\left(\eta_1,\eta_2,t\right)$.

The goal of this section is to compute to leading order the function U, which was introduced in (3.14) and which measures the effect on the evolution of particle η_1 due to the presence of particle η_2. To that aim we recall that the evolution of the radii R_i is given (cf. (2.6) and (2.19)) by

$$\frac{dR_i}{dt}=-\frac{1}{\left(R_i\right)^2}-\frac{1}{R_i}\sum_{j\neq i}^{N}\frac{C_{i,j}}{4\pi R_iR_j}\quad,\quad i=1,...,N \tag{3.21}$$

$$R_1\left(\bar{t}\right)=\bar{R}_1\quad,\quad R_2\left(\bar{t}\right)=\bar{R}_2\quad,\quad R_{k,0}\left(0\right)=R_{k,0}\,. \tag{3.22}$$

The radii of the particles in the system without particle η_2 are given by

$$\frac{dR_i^{(2)}}{dt}=-\frac{1}{\left(R_i^{(2)}\right)^2}-\frac{1}{R_i^{(2)}}\sum_{j\neq i,2}^{N}\frac{C_{i,j}^{(2)}}{4\pi R_i^{(2)}R_j^{(2)}}\quad,\quad i=1,...,N,\ i\neq 2 \tag{3.23}$$

with the same initial data as in (3.22) except for the fact that the particle η_2 has been removed. The coefficients $C_{i,j}^{(2)}$ are the corresponding capacity coefficients with positions x_i and radii $R_i^{(2)}$.

We write

$$r_i = r_i\left(t,\bar{t},\bar{\eta}_1,\bar{\eta}_2,\omega_{0,N}\right) \equiv R_i - R_i^{(2)} . \tag{3.24}$$

and obtain with (3.21) and (3.23) after a linearization that to leading order r_i satisfies

$$\begin{aligned}\frac{dr_i}{dt} &= \frac{2}{\left(R_i^{(2)}\right)^3} r_i + \frac{r_i}{\left(R_i^{(2)}\right)^2} \sum_{j\neq i,2}^{N} \frac{C_{i,j}^{(2)}}{4\pi R_i^{(2)} R_j^{(2)}} - \\ &- \frac{1}{R_i^{(2)}} \sum_{j\neq i,2}^{N} \left[\frac{C_{i,j}}{4\pi R_i R_j} - \frac{C_{i,j}^{(2)}}{4\pi R_i^{(2)} R_j^{(2)}}\right] - \frac{C_{i,2}}{4\pi \left(R_i\right)^2 R_2} .\end{aligned} \tag{3.25}$$

We approximate the last term in (3.25) by the expression (2.18), that is we use

$$C_{i,2} = -(4\pi R_i)\,(4\pi R_2)\,G(x_i - x_2). \tag{3.26}$$

To compute the second last term in (3.25) we need to compute the difference $\left[\frac{C_{i,j}}{4\pi R_i R_j} - \frac{C_{i,j}^{(2)}}{4\pi R_i^{(2)} R_j^{(2)}}\right]$ for $i \neq j$. In Appendix A we show (cf. (5.6)) that

$$\begin{aligned}\frac{C_{i,j}}{4\pi R_i R_j} - \frac{C_{i,j}^{(2)}}{4\pi R_i^{(2)} R_j^{(2)}} &= (4\pi R_2)\, 4\pi G\left(x_i - x_2\right) G\left(x_2 - x_j\right) + \\ &+ \sum_{k\neq i,j,2} \frac{C_{i,k}^{(2)} C_{k,j}^{(2)}}{4\pi \left(R_k^{(2)}\right)^2} \frac{r_k}{4\pi R_i^{(2)} R_j^{(2)}} , \qquad j \neq i,2\end{aligned} \tag{3.27}$$

Using (3.26) and (3.27) in (3.25) we find

$$\begin{aligned}\frac{dr_i}{dt} &= \frac{2}{\left(R_i^{(2)}\right)^3} r_i + \frac{r_i}{\left(R_i^{(2)}\right)^2} \sum_{j\neq i,2}^{N} \frac{C_{i,j}^{(2)}}{4\pi R_i^{(2)} R_j^{(2)}} - \\ &- \frac{(4\pi R_2)}{R_i^{(2)}} G\left(x_i - x_2\right) \sum_{j\neq i,2}^{N} 4\pi G\left(x_2 - x_j\right) - \frac{1}{R_i^{(2)}} \sum_{k\neq i,j,2} \left[\sum_{j\neq i,2}^{N} \frac{C_{i,k}^{(2)} C_{k,j}^{(2)}}{(4\pi)^2 \left(R_k^{(2)}\right)^2 R_i^{(2)} R_j^{(2)}}\right] r_k \\ &+ \frac{4\pi}{R_i^{(2)}} G\left(x_i - x_2\right) .\end{aligned} \tag{3.28}$$

In the last term we also replaced R_i by $R_i^{(2)}$, which is admissible since we only need the leading order terms. We also recall that $G(x_2 - x_j)$ is to be read as $G\left(x_2 - x_j\right)\chi_{\{R_j>0\}}$, that is we only sum over the particles with $R_j > 0$.

Since $\int G(x)\,dx = \int G(x-y)\,dx = \frac{1}{\xi^2}$ we can approximate

$$\sum_{j\neq i,2}^{N} 4\pi G\left(x_2 - x_j\right) = 4\pi \int_{\{R_1>0\}} f_1\left(R_1,t\right) dR_1 \int G\left(x_2 - y\right) dy = \frac{1}{\langle R\rangle} .$$

On the other hand we use the approximation (2.18) in order to approximate the second and fourth term on the right hand side of (3.28). Therefore, using similar integral approximations as before, we obtain

$$\frac{dr_i}{dt} = a\big(R_i^{(2)}\big) r_i - \frac{1}{R_i^{(2)}\,\langle R\rangle} \sum_{k\neq i,2} 4\pi G\left(x_i - x_k\right) r_k + \frac{4\pi G\left(x_i - x_2\right)}{R_i^{(2)}} \left(1 - \frac{R_2}{\langle R\rangle}\right) \tag{3.29}$$

for $i=1,...,N, \quad i\neq 2$, where

$$a\left(R\right):=\frac{2}{R^{3}}-\frac{1}{\left\langle R\right\rangle R^{2}}$$

These equations must be completed with suitable initial and boundary conditions. Taking into account the initial conditions for R_1, R_2 and $R_k,\ k=3,...,N$, we assume

$$r_1\left(\bar{t}\right)=0 \tag{3.30}$$

$$r_i\left(0\right)=0 \quad , \quad i=3,...,N \tag{3.31}$$

We now make the following key assumption. The term $\sum_{k\neq i,2}4\pi G\left(x_i-x_k\right)r_k$ contains the sum of many small, roughly independent, contributions. This is due to the fact, that correlations between the particles are small except for the largest particles. Those are however few and do not contribute to leading order to the sum. Hence, the above term can be approximated by $I\left(x_i-x_2,t,\bar{t}\right)$ where $I\left(x,t,\bar{t}\right)$ is a smooth function in x.

Second, we approximate $R_i^{(2)}$ by $R_L(t,\bar{t},R_i)$, where R_L is given by

$$\frac{dR_L\left(t,\bar{t},\bar{R}\right)}{dt}=-\frac{1}{\left(R_L\left(t,\bar{t},\bar{R}\right)\right)^2}+\frac{1}{\left\langle R\right\rangle\left(t\right)R_L\left(t,\bar{t},\bar{R}\right)} \tag{3.32}$$

$$R_L\left(\bar{t},\bar{t},\bar{R}\right)=\bar{R}\,. \tag{3.33}$$

Such an approximation is valid to leading order as long as $t\sim O(\bar{t})$. For $t\ll\bar{t}$, however, particles η_1 and η_2 do not interact because for those particles which are still alive at time $\bar{t}$ their distance at time t is much larger than the screening length. Hence, (3.29) can be approximated by

$$\frac{dr_i}{dt}=a\left(R_L\left(t,\bar{t},R_i\right)\right)r_i-\frac{I\left(x_i-x_2,t,\bar{t}\right)}{R_L\left(t,\bar{t},R_i\right)\left\langle R\right\rangle}+\frac{4\pi G\left(x_i-x_2\right)}{R_L\left(t,\bar{t},R_i\right)}\left(1-\frac{R_2}{\left\langle R\right\rangle}\right) \tag{3.34}$$

This equation describes the effect of an additional particle η_2 in the system. The last term measures the direct effect of particle η_2 on the particle η_i, whereas the second term on the right hand side is a mean-field like term, due to the change of the radii of all the other particles.

Taking into account (3.31) we can approximate r_i for $i=3,...,N$ as

$$\begin{aligned}r_i\left(t,\bar{t},\bar{\eta}_1,\bar{\eta}_2,\omega_{0,N}\right)=&\int_0^t\frac{\exp\left(\int_s^t a\left(R_L\left(\lambda,\bar{t},R_i\right)\right)d\lambda\right)}{R_L\left(s,\bar{t},R_i\right)}\cdot\\&\left[4\pi G\left(x_i-x_2\right)\left(1-\frac{R_L\left(s,\bar{t},\bar{R}_2\right)}{\left\langle R\right\rangle\left(s\right)}\right)-\frac{I\left(x_i-x_2,s,\bar{t}\right)}{\left\langle R\right\rangle}\right]ds\end{aligned} \tag{3.35}$$

for $i=3,...,N$. Using the definition of $I\left(x,t\right)$ we obtain

$$\begin{aligned}I\left(x-x_2,t,\bar{t}\right)&=\sum_{k\neq 2}4\pi G\left(x-x_k\right)r_k\\&=4\pi\int_0^t\left(\sum_{k\neq 2}\frac{\exp\left(\int_s^t a\left(R_L\left(\lambda,\bar{t},R_k\right)\right)d\lambda\right)}{R_L\left(s,\bar{t},R_k\right)}G\left(x-y\right)\right)\cdot\\&\left[4\pi G\left(x_k-x_2\right)\left(1-\frac{R_L\left(s,\bar{t},\bar{R}_2\right)}{\left\langle R\right\rangle\left(s\right)}\right)-\frac{I\left(x_k-x_2,s,\bar{t}\right)}{\left\langle R\right\rangle\left(s\right)}\right]ds\end{aligned}$$

and we can now approximate the sum in this formula by an integral. To this end we remark that the distribution of radii R_k at time $\bar{t}$ is $f_1\left(R,\bar{t}\right)$. On the other hand the distribution of particles is homogeneous. Therefore, using also the invariance of the problem under translations, we obtain the following integral equation for $I(x,t)$

$$I(x,t,\bar{t}) = 4\pi \int_0^t \int_{[0,1]^3} \int_{\{R>\underline{R}(t,\bar{t})\}} f_1(R,\bar{t}) \frac{\exp\left(\int_s^t a(R_L(\lambda,\bar{t},R))\,d\lambda\right)}{R_L(s,\bar{t},R)} G(x-y,t) \cdot \tag{3.36}$$
$$\left[4\pi G(y,s)\left(1-\frac{R_L\left(s,\bar{t},\bar{R}_2\right)}{\langle R\rangle(s)}\right) - \frac{I(y,s,\bar{t})}{\langle R\rangle(s)}\right] dRdyds$$

where $\underline{R}(t,\bar{t}) < 0$ is the value of the radius such that $R_L(t,\bar{t},R) > 0$ for $R > \underline{R}(t,\bar{t})$. Notice that in (3.36) we are integrating over a set which includes negative particles. The meaning of this is that extinct particles have generated some noise during their life span.

Taking into account (3.35) it follows that we can approximate r_i as

$$r_i(t,\bar{t},\bar{\eta}_1,\bar{\eta}_2,\omega_{0,N}) = \varphi\left(t,\bar{t},\bar{R}_2,R_i,x_i-x_2\right) \quad , \quad i=3,...,N \tag{3.37}$$

where $\bar{\eta}_i = \left(x_i,\bar{R}_i\right)$ and

$$\varphi\left(t,\bar{t},\bar{R}_2,R,x\right) \equiv \int_0^t \frac{\exp\left(\int_s^t a(R_L(\lambda,\bar{t},R))\,d\lambda\right)}{R_L(s,\bar{t},R)} \cdot$$
$$\left[4\pi G(x,s)\left(1-\frac{R_L\left(s,\bar{t},\bar{R}_2\right)}{\langle R\rangle(s)}\right) - \frac{I(x,s,\bar{t})}{\langle R\rangle(s)}\right] ds \tag{3.38}$$

The set of equations (3.36)-(3.38) yields the procedure to approximate the change of the radii of the particles that are within the screening distance of η_2. Notice that the function $\varphi\left(t,\bar{t},\bar{R}_2,R,x\right)$ yields also the procedure of computing r_1 that is the required change of radius in order to compute U_1. Indeed, (3.30) and (3.34) yield

$$r_1(t,\bar{t},\bar{\eta}_1,\bar{\eta}_2,\omega_{0,N}) = -\int_t^{\bar{t}} \frac{\exp\left(\int_s^t a\left(R_L\left(\lambda,\bar{t},\bar{R}_1\right)\right)d\lambda\right)}{R_L\left(s,\bar{t},\bar{R}_1\right)} \cdot$$
$$\left[4\pi G(x_1-x_2,s)\left(1-\frac{R_L\left(s,\bar{t},\bar{R}_2\right)}{\langle R\rangle(s)}\right) - \frac{I(x_1-x_2,s,\bar{t})}{\langle R\rangle}\right] ds$$

In particular we have due to (3.24) that

$$\begin{aligned} &R_{1,0}(\bar{\eta}_1,\bar{\eta}_2,\omega_{0,N},\bar{t}) - R_{1,0}^{(2)}(\bar{\eta}_1,\omega_{0,N},\bar{t}) \\ &\quad = r_1(0,\bar{t},\bar{\eta}_1,\bar{\eta}_2,\omega_{0,N}) = -\exp\left(-\int_0^{\bar{t}} a\left(R_L\left(\lambda,\bar{t},\bar{R}_1\right)\right)d\lambda\right)\varphi\left(\bar{t},\bar{t},\bar{R}_2,\bar{R}_1,x_1-x_2\right) \end{aligned} \tag{3.39}$$

If we use (3.14) and linearize we obtain

$$\frac{\partial R_{1,0}^{(2)}}{\partial R_1}(\bar{\eta}_1,\omega_{0,N},\bar{t})\,U_1 = r_1(0,\bar{t},\bar{\eta}_1,\bar{\eta}_2,\omega_{0,N})$$

which together with (3.39) implies

$$U_1(\bar{t},\bar{\eta}_1,\bar{\eta}_2) = -\exp\left(-\int_0^{\bar{t}} a\left(R_L\left(\lambda,\bar{t},\bar{R}_1\right)\right)d\lambda\right)\frac{\varphi\left(\bar{t},\bar{t},\bar{R}_2,\bar{R}_1,x_1-x_2\right)}{\frac{\partial R_{1,0}^{(2)}}{\partial R_1}}.$$

The term $\frac{\partial R_{1,0}^{(2)}}{\partial R_1}$ can be approximated to leading order using the approximation $R_{1,0}^{(2)} \approx R_L\left(0, \bar{t}, \bar{R}_1\right)$. Differentiating (3.32), (3.33) it follows that

$$\frac{\partial R_{1,0}^{(2)}}{\partial R_1} \approx \exp\left(-\int_0^{\bar{t}} a\left(R_L\left(\lambda, \bar{t}, \bar{R}_1\right)\right) d\lambda\right),$$

whence

$$U_1\left(\bar{t}, \bar{\eta}_1, \bar{\eta}_2\right) = -\varphi\left(\bar{t}, \bar{t}, \bar{R}_2, \bar{R}_1, x_1 - x_2\right) \equiv -\psi\left(\bar{t}, \bar{R}_2, \bar{R}_1, x_1 - x_2\right) \tag{3.40}$$

Before we proceed we also estimate the order of magnitude of the function U. In equation (3.34) the main source term is the last term on the right hand side. Hence, we can expect, recalling the time scale $\langle R \rangle^3$, that

$$r_i \sim \frac{\langle R \rangle^2}{\xi} e^{-|x-x_2|/\xi}. \tag{3.41}$$

If we use this ansatz in the second term on the right hand side of (3.34) we obtain that the second term has the same order of magnitude as the third one and (3.41) is self-consistent.

3.4 Estimating the fluctuations of F

Our goal is to approximate the term in (3.20) which is due to the fluctuations

$$I \equiv \int \left(\int \left[F\left(\eta_1, \omega_{0,N}, t\right) - f_1\left(\eta_1, t\right)\right]\left[F\left(\eta_2, \omega_{0,N}, t\right) - f\left(\eta_2, t\right)\right] d\nu_N\right) dR_2 \tag{3.42}$$

To this end we recall the definition of F in (3.19). We can approximate the function $R_{k,0}^{(\tau_k)}\left(\eta_k, \omega_{0,N}, t\right)$ using a stochastic differential equation. We can rewrite (3.23) as

$$\frac{dR_k^{(2)}}{dt} = -\frac{1}{\left(R_k^{(2)}\right)^2} + \frac{1}{\langle R \rangle}\frac{1}{R_k^{(2)}} - \frac{1}{R_k^{(2)}}\left[\sum_{j \neq k,2} \frac{C_{k,j}^{(2)}}{4\pi R_k^{(2)} R_j^{(2)}} + \frac{1}{\langle R \rangle}\right]$$

We are interested in computing the fluctuations to the leading order. Thus it suffices to approximate $C_{k,j}^{(2)}$ by (2.18) to obtain

$$\frac{dR_k^{(2)}}{dt} = -\frac{1}{\left(R_k^{(2)}\right)^2} + \frac{1}{\langle R \rangle}\frac{1}{R_k^{(2)}} + \frac{1}{R_k^{(2)}}\left[\sum_{j \neq k,2} \frac{e^{-\frac{|x_k - x_j|}{\xi}}}{|x_k - x_j|}\chi_{\{R_j > 0\}} - \frac{1}{\langle R \rangle}\right] \tag{3.43}$$

As in the last subsection we use again the key assumption that for all times most of the particles are to leading order independently distributed.

With this assumption we can approximate the term between brackets in (3.43) by means of a "noise" term that we will denote as $Z(x,t)$. Then

$$\frac{dR_k^{(2)}}{dt} = -\frac{1}{\left(R_k^{(2)}\right)^2} + \frac{1}{\langle R \rangle}\frac{1}{R_k^{(2)}} + \frac{Z(x,t)}{R_k^{(2)}} \tag{3.44}$$

where

$$\langle Z(x,t) \rangle = 0$$

due to the definition of the screening length.

Using (3.19) and (3.44) we find that F evolves according to

$$\frac{\partial F}{\partial t}+\frac{\partial}{\partial R}\left(\left(-\frac{1}{R^2}+\frac{\left[\frac{1}{\langle R\rangle}+Z\left(x,t\right)\right]}{R}\right)F\right)=0. \tag{3.45}$$

In a strict mathematical sense, we should take the initial data $F\left(\eta,0\right)=f_0\left(R\right)$. However, such an approximation would fail for very long times. In practice we will use (3.45) for self-similar solutions where it is possible to argue as in some previous approximations for the characteristics (cf. (3.32), (3.33)). For a given time $\bar{t}$ we can use the approximation (3.45) for times $t\lessapprox\bar{t}$, and this is the only range of times where we will need to compute the fluctuations because their effect disappears in (3.42) for particles that are separated distances longer than the screening length as it will be seen below.

We conclude this section by deriving some further properties of Z. In the limit $\phi\to 0$ we can also assume that the noise Z is Gaussian and it is possible to compute its correlations in time and space. We have with

$$Z\left(x,t\right)=\left[\sum_{j\neq 2}\frac{e^{-\frac{\left|x-x_j\right|}{\xi}}}{\left|x-x_j\right|}\chi_{\left\{R_j(t)>0\right\}}-\frac{1}{\langle R\rangle\left(t\right)}\right]$$

that

$$\begin{aligned}\langle Z\left(x_1,t_1\right)Z\left(x_2,t_2\right)\rangle&=\sum_{j\neq 2}\sum_{l\neq 2}\left\langle\frac{e^{-\frac{\left|x_1-x_j\right|}{\xi(t_1)}}}{\left|x_1-x_j\right|}\frac{e^{-\frac{\left|x_2-x_l\right|}{\xi(t_2)}}}{\left|x_2-x_l\right|}\chi_{\left\{R_j(t_1)>0\right\}}\chi_{\left\{R_l(t_2)>0\right\}}\right\rangle-\frac{1}{\langle R\rangle\left(t_1\right)}\frac{1}{\langle R\rangle\left(t_2\right)}\\&=\sum_{j\neq 2}\sum_{l\neq 2,l\neq j}\left\langle\frac{e^{-\frac{\left|x_1-x_j\right|}{\xi(t_1)}}}{\left|x_1-x_j\right|}\frac{e^{-\frac{\left|x_2-x_l\right|}{\xi(t_2)}}}{\left|x_2-x_l\right|}\chi_{\left\{R_j(t_1)>0\right\}}\chi_{\left\{R_l(t_2)>0\right\}}\right\rangle\\&\quad-\frac{1}{\langle R\rangle\left(t_1\right)}\frac{1}{\langle R\rangle\left(t_2\right)}+\sum_{j\neq 2}\left\langle\frac{e^{-\frac{\left|x_1-x_j\right|}{\xi(t_1)}}}{\left|x_1-x_j\right|}\frac{e^{-\frac{\left|x_2-x_j\right|}{\xi(t_2)}}}{\left|x_2-x_j\right|}\chi_{\left\{R_j(t_1)>0\right\}}\chi_{\left\{R_j(t_2)>0\right\}}\right\rangle\end{aligned}$$

The measure is $\prod_{j\neq 2}\left[\frac{f_1(R_j,t)}{N}dx_jdR_j\right]$ and includes negative particles. Hence we find in the limit $N\to\infty$ that

$$\langle Z\left(x_1,t_1\right)Z\left(x_2,t_2\right)\rangle=\int\frac{e^{-\frac{\left|x_1-y\right|}{\xi(t_1)}}}{\left|x_1-y\right|}\frac{e^{-\frac{\left|x_2-y\right|}{\xi(t_2)}}}{\left|x_2-y\right|}dy\int_{\left\{R_1(t_1)>0,\ R_1(t_2)>0\right\}}f\left(R_1,t_1\right)dR_1$$

Assuming that $t_1\leq t_2$ and using the definition of $R_L\left(t_1,t_2,0\right)$ in (3.32), (3.33) it follows that

$$\langle Z\left(x_1,t_1\right)Z\left(x_2,t_2\right)\rangle=\Lambda\left(x_2-x_1,t_1,t_2\right)\int_{R_L(t_1,t_2,0)}^{\infty}f\left(R_1,t_1\right)dR_1$$

where

$$\Lambda\left(x_2-x_1,t_1,t_2\right)=\int\frac{e^{-\frac{\left|x_1-y\right|}{\xi(t_1)}}}{\left|x_1-y\right|}\frac{e^{-\frac{\left|x_2-y\right|}{\xi(t_2)}}}{\left|x_2-y\right|}dy$$

If t_1 and t_2 are comparable then $\Lambda\left(x_2-x_1,t_1,t_2\right)$ is of order ξ, and the integral $\int_{R_L(t_1,t_2,0)}^{\infty}f\left(R_1,t_1\right)dR_1$ is of order N. Then $\langle Z\left(x_1,t_1\right)Z\left(x_2,t_2\right)\rangle$ is of order $N\xi=N\xi^2\frac{1}{\xi}=\frac{1}{\langle R\rangle\xi}=\frac{r_N}{\xi}\frac{1}{\langle R\rangle^2}=\frac{\sqrt{\phi}}{\langle R\rangle^2}$, whence $|Z|$ is of order $\phi^{\frac{1}{4}}\frac{1}{\langle R\rangle}$.

3.5 Evolution equation for f_1.

In order to compute the evolution equation for f_1 we start from the Liouville equation (2.20) integrated with respect to the variables $\eta_2, \eta_3, ..., \eta_N$, which gives (2.26) with $s = 1$, that is

$$\frac{\partial f_1}{\partial t} - \frac{1}{4\pi} \frac{\partial}{\partial R_1} \left(\frac{1}{(R_1)^2} \left[\left(\frac{1}{(N-1)!} \int \left(\sum_{j=1}^{N} \frac{C_{j,1}}{R_j} \right) D_N d\eta_2 d\eta_3 ... d\eta_N \right) \right] \right) = 0 \; . \tag{3.46}$$

Using the approximation (2.19) and the symmetry properties of the capacity coefficients we obtain

$$\frac{\partial f_1 (R_1, t)}{\partial t} - \frac{\partial}{\partial R_1} \left(\frac{f_1 (R_1, t)}{(R_1)^2} \right) - \frac{\partial}{\partial R_1} \left(\frac{1}{(R_1)^2} \int \frac{\Phi_2 (\eta_1, \eta_2)}{(N-2)! R_2} d\eta_2 \right) = 0 \tag{3.47}$$

with

$$\Phi_2 (\eta_1, \eta_2) = \int C_{1,2} D_N d\eta_3 ... d\eta_N \; . \tag{3.48}$$

Here we assume, due to the screening property, that N is large and that the quantity is independent of N in the limit $N \to \infty$. This assumption is crucial in order to obtain a closed equation for Φ_2.

We are going to approximate the integral $\Phi_2 (\eta_1, \eta_2)$ for measures with small correlations in most of the space of variables except possibly in some small boundary layer.

Let us denote as $K (x - x_i)$ the solution of the problem

$$-\Delta K (x - x_i) = [\delta (x - x_i) - 1] \quad \text{in} \quad \Omega \tag{3.49}$$

with periodic boundary conditions where Ω is the unit cube. The function $K (\cdot)$ is uniquely determined up to a constant and

$$K (x) = \frac{1}{4\pi |x|} (1 + b |x|) \quad \text{as} \quad |x| \to 0 \tag{3.50}$$

where b is a fixed constant that is independent of N.

In order to compute the coefficients $C_{i,j}$ we will use the monopole approximation that is we assume that dipolar and higher order monopolar terms give a negligible contribution in the approximation of the solution v_j of (2.8), (2.9). More precisely, we make, for $j = 1$ say, the ansatz

$$v_1(x) = \bar{v} + \sum_{i=1}^{N} C_{1,i} K(x - x_i)$$

with a constant $\bar{v}$ which has to be determined such that

$$\sum_{l=1}^{N} C_{1,l} = 0. \tag{3.51}$$

Using the boundary condition $v_1(x) = 0$ for $x \in \partial B_{R_i}(x_i)$, yields

$$\frac{C_{1,2}}{4\pi R_2} + \sum_{l=1, l \neq 2} C_{1,l} K (x_2 - x_l) + \bar{v} = 0. \tag{3.52}$$

If we multiply (3.51) by $\mathcal{D}_N$ and integrating over $\eta_2, \cdots, \eta_N$, we obtain

$$C_{1,1}(N-2)! f_1(R_1) + \int \Phi_2(\eta_1, \eta_2) \, d\eta_2 = 0. \tag{3.53}$$

Similarly we obtain from (3.52) that

$$\frac{\Phi_2(\eta_1,\eta_2)}{4\pi R_2}+(N-2)!(C_{1,1}K(x_1-x_2)+\bar{v})f_2(\eta_1,\eta_2)$$
$$+(N-2)\int C_{1,3}D_N K(x_2-x_3)\,d\eta_3...d\eta_N=0.$$

Let us denote by $C_{1,3}^{(2)}$ the capacity coefficient induced by the particle η_1 on the particle η_3 if the particle η_2 is eliminated from the system. Then

$$\frac{\Phi_2(\eta_1,\eta_2)}{4\pi R_2}+(N-2)!(C_{1,1}K(x_1-x_2)+\bar{v})f_2(\eta_1,\eta_2)$$
$$+(N-2)\left\{\int C_{1,3}^{(2)}D_N K(x_2-x_3)\,d\eta_3...d\eta_N+\int\left[C_{1,3}-C_{1,3}^{(2)}\right]D_N K(x_2-x_3)\,d\eta_3...d\eta_N\right\}=0 \tag{3.54}$$

The coefficient $C_{1,3}^{(2)}$ is independent of η_2. As before we assume that particles whose distance is larger than the screening length are uncorrelated. Then we obtain in the limit $N\to\infty$

$$(N-2)\int C_{1,3}^{(2)}D_N K(x_2-x_3)\,d\eta_3...d\eta_N=f_1(R_2)\int\Phi_2(\eta_1,\eta_3)K(x_2-x_3)\,d\eta_3 \tag{3.55}$$

If we combine (3.54) with (3.55) we obtain the following integral equation for Φ_2

$$\frac{\Phi_2(\eta_1,\eta_2)}{4\pi R_2}+f_1(R_2)\int\Phi_2(\eta_1,\eta_3)K(x_2-x_3)\,d\eta_3+(N-2)!(C_{1,1}K(x_1-x_2)+\bar{v})f_2(\eta_1,\eta_2)$$
$$+(N-2)\int\left[C_{1,3}-C_{1,3}^{(2)}\right]D_N K(x_2-x_3)\,d\eta_3...d\eta_N=0 \tag{3.56}$$

It is not hard to derive an estimate of the order of magnitude of the last term in (3.56). Indeed, (5.3) implies that to leading order

$$C_{1,3}-C_{1,3}^{(2)}=(4\pi R_1)(4\pi R_2)(4\pi R_3)\,G(x_1-x_2)\,G(x_2-x_3) \tag{3.57}$$

In view of (3.67) we are interested in computing $\int\frac{\Phi_2(\eta_1,\eta_2)}{R_2}d\eta_2$. Using the screening approximation (2.18) we can see that the order of magnitude of $\Phi_2(\eta_1,\eta_2)$ for nearly uncorrelated measures is

$$\Phi_2(\eta_1,\eta_2)\approx(N-2)!R_1R_2f_1(R_1)\,f_1(R_2)\,G(x_1-x_2)\ ,$$

whence

$$\int\frac{\Phi_2(\eta_1,\eta_2)}{R_2}d\eta_2\approx(N-2)!\frac{R_1}{\langle R\rangle}f_1(R_1)\approx(N-2)!f_1(R_1) \tag{3.58}$$

Equation (3.57) implies the following order of magnitude

$$(N-2)\int\left[C_{1,3}-C_{1,3}^{(2)}\right]D_N K(x_2-x_3)\,d\eta_2d\eta_3...d\eta_N\approx\sqrt{\phi}\,(N-2)!f_1(R_1)\ . \tag{3.59}$$

We want to derive an approximation for $\int\frac{\Phi_2(\eta_1,\eta_2)}{R_2}d\eta_2$ which contains only the leading order terms for $f_1(R_1)$ and the terms containing derivatives of $f_1(R_1)$ larger than $\sqrt{\phi}$. Therefore the contributions of the terms in (3.59) yield terms containing derivatives of $f_1(R_1)$ which are smaller than $\sqrt{\phi}$ since these derivatives arise from translations in the variable R_1 of order U, i.e. $\sqrt{\phi}$. Thus we can approximate (3.56) by

$$\begin{aligned}\frac{\Phi_2\left(\eta_1,\eta_2\right)}{4\pi R_2} &+ f_1\left(R_2\right)\int \Phi_2\left(\eta_1,\eta_3\right) K\left(x_2-x_3\right) d\eta_3 \\ &+ (N-2)!(C_{1,1}K\left(x_1-x_2\right)+\bar{v}) f_2\left(\eta_1,\eta_2\right) = 0\,.\end{aligned} \tag{3.60}$$

We define

$$\Psi\left(\eta_1,x_2\right) = \int \Phi_2\left(\eta_1,\eta_2\right) dR_2 \tag{3.61}$$

Multiplying (3.60) by $4\pi R_2$ and integrating on R_2 we obtain

$$\begin{aligned}\Psi\left(\eta_1,x_2\right) &+ \frac{1}{\xi^2}\int K\left(x_2-x_3\right)\Psi\left(\eta_1,x_3\right) dx_3 \\ &+ (N-2)!(C_{1,1}K\left(x_1-x_2\right)+\bar{v})\int 4\pi R_2 f_2\left(\eta_1,\eta_2\right) dR_2 = 0\,.\end{aligned} \tag{3.62}$$

Then, eliminating the integral term in (3.60) in Φ_2 with the help of (3.62), integrating the resulting formula with respect to η_2 and using (2.19), we obtain

$$\begin{aligned}\int \frac{\Phi_2\left(\eta_1,\eta_2\right)}{4\pi R_2} d\eta_2 &= \frac{1}{4\pi\left\langle R\right\rangle}\int \Psi\left(\eta_1,x_2\right) dx_2 - (N-2)!\bar{v}\int f_2\left(\eta_1,\eta_2,t\right)\left(1-\frac{R_2}{\langle R\rangle}\right) d\eta_2 \\ &- (N-2)!\left(4\pi R_1\right)\int K\left(x_1-x_2\right) f_2\left(\eta_1,\eta_2,t\right)\left(1-\frac{R_2}{\langle R\rangle}\right) d\eta_2\end{aligned} \tag{3.63}$$

In the following we denote

$$h\left(\eta,t\right) = -F(\eta,t) + f_1\left(R,t\right) \tag{3.64}$$

With (3.64) we can rewrite (3.20) as

$$\begin{aligned}\int f_2\left(\eta_1,\eta_2,t\right) dR_2 &= \int\left(\int f_1\left(\eta_1+U_1,t\right) f\left(\eta_2,t\right) d\nu_N\right) dR_2 \\ &+ \int \left\langle h\left(\eta_1,t\right)\ h\left(\eta_2,t\right)\right\rangle dR_2\,.\end{aligned} \tag{3.65}$$

Similarly we obtain

$$\begin{aligned}\int f_2\left(\eta_1,\eta_2,t\right) R_2 dR_2 &= \int\left(\int f_1\left(\eta_1+U_1,t\right) f\left(\eta_2,t\right) R_2 d\nu_N\right) dR_2 \\ &+ \int \left\langle h\left(\eta_1,t\right)\ h\left(\eta_2,t\right)\right\rangle R_2 dR_2\,.\end{aligned} \tag{3.66}$$

Using (3.65), (3.66) and (3.63) we find

$$\begin{aligned}\frac{1}{(N-2)!}\int \frac{\Phi_2(\eta_1,\eta_2)}{4\pi R_2} d\eta_2 &= -\frac{R_1}{\langle R\rangle} f_1(R_1) - 4\pi R_1\int K(x_1-x_2) f_1(R_1+U) f_1(R_2)\Big(1-\frac{R_2}{\langle R\rangle}\Big) d\eta_2 \\ &- \bar{v}\int f_1(R_1+U)\, f_1(R_2)\Big(1-\frac{R_2}{\langle R\rangle}\Big) d\eta_2 \\ &- 4\pi R_1\int K(x_1-x_2)\langle h(\eta_1,t)\, h(\eta_2,t)\rangle\Big(1-\frac{R_2}{\langle R\rangle}\Big) d\eta_2\,.\end{aligned} \tag{3.67}$$

Taylor expansion in the second and third term on the right hand side yields furthermore

$$
\begin{aligned}
\frac{1}{4\pi R_1}\int \frac{\Phi_2(\eta_1,\eta_2)}{(N-2)!4\pi R_2}\,d\eta_2 = {} & -\frac{f_1(R_1,t)}{4\pi\langle R\rangle} \\
& -\frac{\partial f_1(R_1,t)}{\partial R_1}\Big(\int K(x_1-x_2)\,U(R_1,R_2,x_1-x_2,t)\left(1-\frac{R_2}{\langle R\rangle}\right)f_1(R_2)\,d\eta_2 \\
& -\frac{\bar v}{4\pi R_1}\int U(R_1,R_2,x_1-x_2,t)\left(1-\frac{R_2}{\langle R\rangle}\right)f_1(R_2)\,d\eta_2\Big) \\
& -\int K(x_1-x_2)\langle h(\eta_1,t)\,h(\eta_2,t)\rangle\Big(1-\frac{R_2}{\langle R\rangle}\Big)\,d\eta_2\,.
\end{aligned}
\tag{3.68}
$$

We now argue that the second last term in (3.68) is of higher order than the other terms in the equation.

To this end we integrate (3.62) with respect to x_2 and use (3.53) and (3.66) to obtain

$$
\begin{aligned}
-4\pi R_1 f_1(R_1) - \frac{1}{\xi^2}\int K(x)\,dx + 4\pi R_1\int K(x_1-x_2)\int 4\pi R_2 f_1(R_1+U)f_1(R_2)\,d\eta_2 \\
+\bar v\int dx_2\int 4\pi R_2 f_2(\eta_1,\eta_2)\,dR_2 = 0.
\end{aligned}
\tag{3.69}
$$

Again we use Taylor's expansion and recall that $4\pi\langle R\rangle\xi^2=1$ to find after some cancellations that

$$
\begin{aligned}
-4\pi R_1 f_1(R_1) + 4\pi R_1\int K(x_1-x_2)\,dx_2\int 4\pi R_2\frac{\partial f_1}{\partial R_1}U\,f_1(R_2)\,dR_2 \\
+\bar v\int dx_2\int 4\pi R_2 f_2(\eta_1,\eta_2)\,dR_2 = 0.
\end{aligned}
\tag{3.70}
$$

We compare the order of size of the different terms. The first one is of order $\langle R\rangle f_1$, whereas the second, in view of (3.41)

$$
U\sim\frac{\langle R\rangle^2}{|x|}e^{-|x|/\xi}\,,
$$

is of order $\langle R\rangle^2\frac{f_1}{\xi}=\phi^{1/2}\langle R\rangle f_1$. Finally, the last term is of order $\bar v\frac{L^3}{\xi^2}f_1$ where $L=1$ is the box size. Hence, $\bar v$ is of order $\frac{\langle R\rangle\xi^2}{L^3}=\phi^{1/2}\big(\frac{\xi}{L}\big)^3$ and can be neglected compared to the other terms which are of order $\phi^{1/2}$.

We can now conclude and use (3.67) and (3.68) in (3.47) to find

$$
\begin{aligned}
&\frac{\partial f_1(R_1,t)}{\partial t}-\frac{\partial}{\partial R_1}\left(\frac{f_1(R_1,t)}{(R_1)^2}\right)+\frac{\partial}{\partial R_1}\left(\frac{f_1(R_1,t)}{R_1\langle R\rangle}\right) \\
&+\frac{\partial}{\partial R_1}\left(\frac{4\pi}{R_1}\left(\int K(x_1-x_2)\,U(R_1,R_2,x_1-x_2,t)\left(1-\frac{R_2}{\langle R\rangle}\right)f_1(R_2)\,d\eta_2\right)\frac{\partial f_1(R_1,t)}{\partial R_1}\right) \\
&+\frac{\partial}{\partial R_1}\left(\frac{4\pi}{R_1}\int K(x_1-x_2)\langle h(\eta_1,t)\,h(\eta_2,t)\rangle\Big(1-\frac{R_2}{\langle R\rangle}\Big)\,d\eta_2\right)\;=\;0\,.
\end{aligned}
\tag{3.71}
$$

We remark that equation (3.71) contains a term with second order derivatives that is small but plays a relevant role for the largest particles. The effect of this term will be to introduce boundary layer effects in the region of largest particles.

3.6 The final result

In our last step we use the results from Section 3.3 in (3.71). It turns out that we can slightly simplify the equations which define $\psi\left(\bar{t},\bar{R}_2,\bar{R}_1,x_1-x_2\right)$ and consequently U_1. To that aim it is convenient to define

$$J\left(x,s,\bar{t},\bar{R}_2\right):=4\pi G\left(x,s\right)\left(1-\frac{R_L\left(s,\bar{t},\bar{R}_2\right)}{\langle R\rangle\left(s\right)}\right)-\frac{I\left(x,s,\bar{t}\right)}{\langle R\rangle\left(s\right)} \tag{3.72}$$

$$H\left(t,s,\bar{t}\right):=\int_{\{R>\underline{R}(t,\bar{t})\}}f_1\left(R,\bar{t}\right)\frac{\exp\left(\int_s^t a\left(R_L\left(\lambda,\bar{t},R\right)\right)d\lambda\right)}{R_L\left(s,\bar{t},R\right)}dR \tag{3.73}$$

With these definitions (3.36) can be expressed as

$$\begin{aligned}&J\left(x,t,\bar{t},\bar{R}_2\right)+\frac{4\pi}{\langle R\rangle\left(t\right)}\int_0^t\int_{[0,1]^3}H\left(t,s,\bar{t}\right)G\left(x-y,t\right)J\left(y,s,\bar{t},\bar{R}_2\right)dyds\\&=4\pi G\left(x,t\right)\left(1-\frac{R_L\left(t,\bar{t},\bar{R}_2\right)}{\langle R\rangle\left(t\right)}\right)\end{aligned} \tag{3.74}$$

and (3.38) and (3.40) yield

$$\psi\left(\bar{t},\bar{R}_2,R,x\right)=\varphi\left(\bar{t},\bar{t},\bar{R}_2,R,x\right)=\int_0^{\bar{t}}\frac{\exp\left(\int_s^{\bar{t}}a\left(R_L\left(\lambda,\bar{t},R\right)\right)d\lambda\right)}{R_L\left(s,\bar{t},R\right)}J\left(x,s,\bar{t},\bar{R}_2\right)ds \tag{3.75}$$

The equations (3.40) and (3.73)-(3.75) provide the algorithm to compute $U_1\left(\bar{t},\bar{\eta}_1,\bar{\eta}_2\right)$. In order to simplify the computations we remark that

$$\exp\left(\int_s^{\bar{t}}a\left(R_L\left(\lambda,\bar{t},R\right)\right)d\lambda\right)=\frac{\frac{\partial R_L(\bar{t},\bar{t},R)}{\partial R}}{\frac{\partial R_L(s,\bar{t},R)}{\partial R}}=\frac{1}{\frac{\partial R_L(s,\bar{t},R)}{\partial R}}$$

Therefore (3.73) and (3.75) can be written as

$$H\left(t,s,\bar{t}\right)=\int_{\{R\,:\,R_L(t,\bar{t},R)>0\}}\frac{\frac{\partial R_L(t,\bar{t},R)}{\partial R}}{R_L\left(s,\bar{t},R\right)\frac{\partial R_L(s,\bar{t},R)}{\partial R}}f_1\left(R,\bar{t}\right)dR \tag{3.76}$$

$$\psi\left(\bar{t},\bar{R}_2,R,x\right)=\int_0^{\bar{t}}\frac{J\left(x,s,\bar{t},\bar{R}_2\right)ds}{R_L\left(s,\bar{t},R\right)\frac{\partial R_L(s,\bar{t},R)}{\partial R}} \tag{3.77}$$

The problem can be further simplified taking into account that the relevant quantity that must be computed in (3.71) is the integral

$$\begin{aligned}&\int K\left(x_1-x_2\right)U\left(\eta_1,\eta_2,\bar{t}\right)\left(1-\frac{\bar{R}_2}{\langle R\rangle}\right)f_1\left(\bar{R}_2,\bar{t}\right)d\bar{\eta}_2\\&\qquad=-\int_{\{\bar{R}_2>0\}}\psi\left(\bar{t},\bar{R}_2,R_1,x\right)K\left(x\right)\left(1-\frac{\bar{R}_2}{\langle R\rangle}\right)f_1\left(\bar{R}_2,\bar{t}\right)d\bar{R}_2dx\end{aligned}$$

With (3.77) we find that

$$Z\left(\bar{t},R_1,x\right):=\int_{\{\bar{R}_2>0\}}\psi\left(\bar{t},\bar{R}_2,R_1,x\right)f_1\left(R_2,\bar{t}\right)\left(1-\frac{\bar{R}_2}{\langle R\rangle\left(\bar{t}\right)}\right)d\bar{R}_2=\int_0^{\bar{t}}\frac{W\left(s,\bar{t},x\right)ds}{R_L\left(s,\bar{t},R_1\right)\frac{\partial R_L(s,\bar{t},R_1)}{\partial R}}$$

where

$$W\left(s,\bar{t},x\right)\equiv\int_{\left\{\bar{R}_{2}>0\right\}}J\left(x,s,\bar{t},\bar{R}_{2}\right)f_{1}\left(R_{2},\bar{t}\right)\left(1-\frac{\bar{R}_{2}}{\langle R\rangle\left(\bar{t}\right)}\right)d\bar{R}_{2} \tag{3.78}$$

Hence

$$\begin{aligned}\int K\left(x_{1}-x_{2},\bar{t}\right)U\left(\eta_{1},\eta_{2},\bar{t}\right)f_{1}\left(\bar{R}_{2},\bar{t}\right)d\bar{\eta}_{2}&=-\int Z\left(\bar{t},R_{1},x\right)K\left(x,\bar{t}\right)dx \\ &=-\int_{0}^{\bar{t}}\frac{\int W\left(s,\bar{t},x\right)K\left(x,\bar{t}\right)dx}{R_{L}\left(s,\bar{t},R_{1}\right)\frac{\partial R_{L}\left(s,\bar{t},R_{1}\right)}{\partial R}}ds\end{aligned} \tag{3.79}$$

If we multiply (3.74) by $f_{1}\left(\bar{R}_{2},\bar{t}\right)$ and integrate with respect to $\bar{R}_{2}$ we obtain that the function $W\left(s,\bar{t},x\right)$ defined in (3.78) satisfies

$$\begin{aligned}&W\left(t,\bar{t},x\right)+\frac{4\pi}{\langle R\rangle\left(t\right)}\int_{0}^{t}H\left(t,s,\bar{t}\right)\left(\int_{[0,1]^{3}}G\left(x-y,t\right)W\left(s,\bar{t},y\right)dy\right)ds \\ &=4\pi G\left(x,t\right)\int_{\left\{\bar{R}_{2}>0\right\}}\left(1-\frac{R_{L}\left(t,\bar{t},\bar{R}_{2}\right)}{\langle R\rangle\left(t\right)}\right)\left(1-\frac{\bar{R}_{2}}{\langle R\rangle\left(\bar{t}\right)}\right)f_{1}\left(\bar{R}_{2},\bar{t}\right)d\bar{R}_{2}\end{aligned} \tag{3.80}$$

Let us now formulate the resulting model. Combining (3.71) and (3.79) it follows that the function f_1 solves

$$\begin{aligned}&\frac{\partial f_{1}\left(R,\bar{t}\right)}{\partial\bar{t}}-\frac{\partial}{\partial R}\left(\left(\frac{1}{\left(R\right)^{2}}-\frac{1}{R\left\langle R\right\rangle}\right)f_{1}\left(R,\bar{t}\right)\right) \\ &=\frac{\partial}{\partial R}\Bigg(\left[\frac{4\pi}{R}\int_{0}^{\bar{t}}\frac{\left(\int_{[0,1]^{3}}W\left(s,\bar{t},x\right)K\left(x\right)dx\right)}{R_{L}\left(s,\bar{t},R\right)\frac{\partial R_{L}\left(s,\bar{t},R\right)}{\partial R}}ds\right]\frac{\partial f_{1}\left(R,\bar{t}\right)}{\partial R} \\ &\qquad-\frac{4\pi}{R}\int K(x_{1}-x_{2})\langle h(\eta_{1},t)\,h(\eta_{2},t)\rangle\Big(1-\frac{R_{2}}{\langle R\rangle}\Big)\,d\eta_{2}\Bigg)\end{aligned} \tag{3.81}$$

where the function W satisfies the integral equation (3.80) with the kernel H as in (3.76). Notice that the left-hand side is the classical LSW model. The term on the right yields a corrective effect due to pair interactions between particles.

4 Self-similar solutions

4.1 The equation in self-similar variables

We now look for self-similar solutions of the model (3.76), (3.80), (3.81) in the limit of small volume fraction. Notice that the volume fraction filled by the particles is

$$\int_{[0,1]^{3}}\int_{\{R>0\}}f_{1}\left(R,t\right)R^{3}\,dR\,dx=\phi \tag{4.1}$$

Hence we look for self-similar solutions of the form

$$f_{1}\left(R,t\right)=\phi t^{-4/3}\Phi\left(\rho\right)\quad,\quad\rho=t^{-1/3}R, \tag{4.2}$$

such that

$$\int_{\{\rho>0\}} \rho^3 \Phi(\rho)\, d\rho = 1 \tag{4.3}$$

For such solutions the screening length $\xi(t)$ has the form

$$\xi(t) = \xi_0 t^{1/3} \qquad \text{with} \qquad \frac{1}{\xi_0^2} = \phi\, 4\pi \int_0^\infty \rho\Phi(\rho)\, d\rho =: \ 4\pi\, \phi\, B\,, \tag{4.4}$$

the average radius $\langle R\rangle(t)$ is given by

$$\langle R\rangle(t) = r_0 t^{1/3} \qquad \text{with} \qquad r_0 = \frac{\int_0^\infty \rho\Phi(\rho)\, d\rho}{\int_0^\infty \Phi(\rho)\, d\rho} \tag{4.5}$$

and $R_L(t, \bar{t}, R)$ has the functional form

$$R_L(t, \bar{t}, R) = t^{1/3} r_L(\tau, \rho)\ , \tag{4.6}$$

$$\tau = \ln\left(\frac{t}{\bar{t}}\right), \tag{4.7}$$

$$\rho = (\bar{t})^{-1/3} R\,. \tag{4.8}$$

Taking into account (3.32) and (3.33) it follows that

$$\frac{\partial r_L(\tau,\rho)}{\partial \tau} = -\frac{1}{(r_L(\tau,\rho))^2} + \frac{1}{r_0}\frac{1}{r_L(\tau,\rho)} - \frac{1}{3} r_L(\tau,\rho)\ , \tag{4.9}$$

$$r_L(0,\rho) = \rho\,. \tag{4.10}$$

Notice that this formula is valid both for positive and negative values of $r_L(\tau,\rho)$.
We write also $G(x,t)$ and $K(x)$ in self-similar form via

$$G(x,t) = \frac{1}{\xi_0 t^{1/3}} g\left(\frac{y}{e^{\tau/3}}\right)\ , \tag{4.11}$$

$$K(x) = \frac{1}{\xi_0 t^{1/3}} k\left(\frac{y}{e^{\tau/3}}\right)\ , \tag{4.12}$$

where τ is as in (4.7) and

$$g(z) = \frac{e^{-|z|}}{4\pi|z|} \qquad \text{and} \qquad y = \frac{x}{(\bar{t})^{1/3}\,\xi_0} \tag{4.13}$$

Using (4.2) and (4.6) we obtain with $\chi = \frac{s}{t}$ the following formula for $H(t,s,\bar{t})$

$$\begin{aligned} H(t,s,\bar{t}) &= \phi\, \frac{1}{(\bar{t})^{4/3}} \frac{e^{\tau/3}}{\chi^{2/3}} \int_{\{\rho\,:\, r_L(\tau,\rho)>0\}} \frac{\frac{\partial r_L(\tau,\rho)}{\partial\rho}}{r_L(\chi,\rho)\frac{\partial r_L(\chi,\rho)}{\partial\rho}} \Phi(\rho)\, d\rho \\ &=: \phi\, \frac{B}{(\bar{t})^{4/3}} \kappa(\chi,\tau)\ . \end{aligned} \tag{4.14}$$

It is natural to look for self-similar solutions of equation (3.80) of the form

$$W(t,\bar{t},x) = \frac{\phi^{3/2}}{(\bar{t})^{4/3}} \omega(\tau, y) \tag{4.15}$$

We plug definitions (4.14) and (4.15) into (3.80) and change variables accordingly. Notice that in the limit $\phi \to 0$ the integration in the cube $[0,1]^3$ becomes integration in the whole space. Recalling also (4.4) we obtain

$$\begin{aligned}\omega(\tau, y) + &\frac{1}{r_0 e^{\tau^{2/3}}} \int_0^\tau \kappa(\chi, \tau) \left(\int_{I\!R^3} g\left(\frac{y-\bar{y}}{e^{\tau/3}}\right) \omega(\chi, \bar{y})\, d\bar{y} \right) d\chi \\ &= \frac{4\pi\sqrt{4\pi B}}{e^{\tau/3}} g\left(\frac{y}{e^{\tau/3}}\right) \int_{\{\rho>0\}} \left(1 - \frac{r_L(\tau,\rho)}{r_0}\right)\left(1 - \frac{\rho}{r_0}\right) \Phi(\rho)\, d\rho\,. \end{aligned} \tag{4.16}$$

We also need to introduce self-similar variables for the function F. It is more convenient to work with the integrated function and thus we define

$$\int_R^\infty F(\lambda, t)\, d\lambda = \frac{\phi}{t} S(\rho, \tau) \tag{4.17}$$

such that due to (3.45), (4.7) and (4.8) the function S satisfies

$$\frac{\partial S(\rho, y, \tau)}{\partial \tau} - S(\rho, y, \tau) - \frac{1}{3} y S_y(\rho, y, \tau) + \left(-\frac{1}{\rho^2} + \frac{\left[\frac{1}{\langle\rho\rangle} + \phi^{\frac{1}{4}} \zeta(y, \tau)\right]}{\rho} - \frac{1}{3}\rho \right) \frac{\partial S(\rho, y, \tau)}{\partial \rho} = 0\,, \tag{4.18}$$

where

$$Z(x,t) = \frac{\phi^{1/4}}{t^{1/3}} \zeta(\eta, \tau).$$

In a similar manner we define

$$\Psi(\rho) := \int_\rho^\infty \Phi(\lambda)\, d\lambda\,. \tag{4.19}$$

The characteristics of equation (4.18) are given by

$$\frac{dy}{d\tau} = -\frac{y}{3} \tag{4.20}$$

$$\frac{d\bar{r}_L(\rho, \tau)}{d\tau} = \left(-\frac{1}{\bar{r}_L^2} + \left[\frac{1}{r_0} + \phi^{\frac{1}{4}} \zeta(y, \tau)\right] \frac{1}{\bar{r}_L} - \frac{\bar{r}_L}{3} \right) \tag{4.21}$$

$$\frac{dS}{d\tau} = S \tag{4.22}$$

with initial values for $\bar{r}_L$ given by

$$\bar{r}_L(0, \rho) = \rho. \tag{4.23}$$

We can compute the stochastic properties of $\zeta(y, \tau)$ as follows

$$\langle \zeta(y, \tau) \rangle = 0$$

and

$$\langle \zeta(y_1, \tau_1) \zeta(y_2, \tau_2) \rangle = \sqrt{\phi}\, \xi_0 e^{1/3(\tau_2 - \tau_1)} \lambda\left((y_2 - y_1) e^{\frac{(\tau_2-\tau_1)}{3}}, e^{-\frac{(\tau_2-\tau_1)}{3}} \right) \int_{r_L(0, e^{\tau_2 - \tau_1})}^\infty \Phi(\rho)\, d\rho \tag{4.24}$$

for $\tau_1 \leq \tau_2$, where

$$\lambda\left(y e^{\frac{(\tau_2-\tau_1)}{3}}, e^{-\frac{(\tau_2-\tau_1)}{3}} \right) = e^{\frac{(\tau_2-\tau_1)}{3}} \int \frac{e^{-|z| e^{\frac{\tau_2-\tau_1}{3}}}}{|z|} \frac{e^{-|y-z|}}{|y-z|} dz\,.$$

Finally, due to (4.2), (4.6), (4.11), (4.15) and (4.17) we find that self-similar solutions to equation (3.81) are given by

$$\begin{aligned} &-\frac{4}{3}\Phi(\rho)-\frac{1}{3}\rho\frac{d\Phi(\rho)}{d\rho}-\frac{d}{d\rho}\left(\left(\frac{1}{(\rho)^2}-\frac{1}{r_0\rho}\right)\Phi(\rho)\right)\\ &\quad=\frac{d}{d\rho}\left(\left[\frac{\sqrt{\phi}}{\sqrt{4\pi B}}\frac{1}{\rho}\int_0^1\frac{\int_{\mathbb{R}^3}(\omega(\chi,y)\,k(y))\,dy}{(\chi)^{2/3}\,r_L(\chi,\rho)\,\frac{\partial r_L(\chi,\rho)}{\partial\rho}}d\chi\right]\frac{d\Phi(\rho)}{d\rho}\right)\\ &\quad-\frac{d}{d\rho}\left(\frac{4\pi}{\rho}\int K(y_1-y_2)\frac{\partial^2}{\partial\rho_1\partial\rho_2}C(\rho_1,\rho_2,y_1,y_2)\Big(1-\frac{\rho_2}{r_0}\Big)\,d\rho_2dy_2\right), \end{aligned} \tag{4.25}$$

where

$$C(\rho_1,\rho_2,y_1,y_2):=\langle S(\rho_1,y_1,\tau)\,S(\rho_2,y_2,\tau)-\Psi(\rho_1)\Psi(\rho_2)\rangle$$

is stationary, since S is a stationary stochastic process.

In the rest of this paper we will study solutions of (4.25). This equation is a singular perturbation of the classical LSW equation. We will see later that the main effect of the term on the right hand side of (4.25) is to introduce a boundary layer near the end of the support of the classical LSW solution. As a first step we will show in the next subsection that the last term in (4.25) is negligible.

4.2 Estimating the correlation function $C(\rho_1,\rho_2,y_1,y_2)$

Due to the exponential decay of the correlations the main contribution to the integral

$$I(\rho_1):=\int K(y_1-y_2)\frac{\partial^2}{\partial\rho_1\partial\rho_2}C(\rho_1,\rho_2,y_1,y_2)\Big(1-\frac{\rho_2}{r_0}\Big)\,d\rho_2dy_2$$

comes from points y_1,y_2 whose distance is of the order of the screening length, which is now normalized to 1.

Due to (4.20) the distance between two characteristics $y_1(\tau)$ and $y_2(\tau)$ increases exponentially as $\tau\to-\infty$. As a consequence, the functions $S(\rho_1,y_1(\tau),\tau)$ and $S(\rho_2,y_2(\tau),\tau)$ are independent variables as $\tau\to-\infty$. This fact will be used repeatedly in the following.

Let us begin with the formula

$$\begin{aligned} &\langle S(\rho_1,y_1,0)\,S(\rho_2,y_2,0)\rangle-\Psi(\rho_1)\,\Psi(\rho_2)\\ &=\langle(S(\rho_1,y_1,0)-\Psi(\rho_1))\,(S(\rho_2,y_1,0)-\Psi(\rho_2))\rangle\\ &=\langle(S(r_L(\rho_1,0),y_1,0)-\Psi(r_L(\rho_1,0)))\,(S(r_L(\rho_2,0),y_1,0)-\Psi(r_L(\rho_2,0)))\rangle \end{aligned}$$

The characteristics (in the radius variable) for S are the "stochastic" curves $\bar{r}_L(\rho_1,\tau)$. It will be convenient to define a new function $\tilde{S}$ evolving by means of the characteristics $r_L(\rho_1,\tau)$ that are the characteristics for the equation satisfied by Ψ. By assumption $S(\rho_1,y_1,0)=\tilde{S}(\rho_1,y_1,0)$. Notice that $\tilde{S}$ solves the same equation as Ψ. (There are some additional corrective terms that are very small, of order $\sqrt{\phi}$. Moreover, since they are the same in both equations they would cancel in the next arguments). Using then the evolution by characteristics for the difference $\tilde{S}-\Psi$ we can write

We now use the fact that the functions $S(r_L(\rho_1,0),y_1,0)$ and $\Psi(r_L(\rho_1,0))$ evolve according to the same equation. Notice that we are ignoring the term $\bar{r}_L(\rho_1,0)$ in this argument. Using the evolution by characteristics, and neglecting for the moment the small noise term that would be the same both for $S\left(r_L\left(\rho_1,\tau\right),y_1,\tau\right)$ and $\Psi\left(r_L\left(\rho_1,\tau\right)\right)$ it follows that their effect cancels out and we are left only with the "leading part". Then

$$
\begin{aligned}
&\left\langle\left(S\left(r_L\left(\rho_1,0\right),y_1,0\right)-\Psi\left(r_L\left(\rho_1,0\right)\right)\right)\left(S\left(r_L\left(\rho_2,0\right),y_2,0\right)-\Psi\left(r_L\left(\rho_2,0\right)\right)\right)\right\rangle \\
&=\left\langle\left(\tilde{S}\left(r_L\left(\rho_1,0\right),y_1,0\right)-\Psi\left(r_L\left(\rho_1,0\right)\right)\right)\left(\tilde{S}\left(r_L\left(\rho_2,0\right),y_2,0\right)-\Psi\left(r_L\left(\rho_2,0\right)\right)\right)\right\rangle \\
&=e^{-2\tau^*}\left\langle\left(\tilde{S}\left(r_L\left(\rho_1,\tau^*\right),y_1e^{-\frac{\tau^*}{3}},\tau^*\right)-\Psi\left(r_L\left(\rho_1,\tau^*\right)\right)\right)\right. \\
&\qquad\left.\cdot\left(\tilde{S}\left(r_L\left(\rho_2,\tau^*\right),y_2e^{-\frac{\tau^*}{3}},\tau^*\right)-\Psi\left(r_L\left(\rho_2,\tau^*\right)\right)\right)\right\rangle
\end{aligned}
$$

It is not completely obvious that the variables $\left(\tilde{S}\left(r_L\left(\rho_1,\tau^*\right),y_1e^{-\frac{\tau^*}{3}},\tau^*\right)-\Psi\left(r_L\left(\rho_1,\tau^*\right)\right)\right)$ and $\left(\tilde{S}\left(r_L\left(\rho_2,\tau^*\right),y_2e^{-\frac{\tau^*}{3}},\tau^*\right)-\Psi\left(r_L\left(\rho_2,\tau^*\right)\right)\right)$ are uncorrelated, because although the points $y_1e^{-\frac{\tau^*}{3}}$, $y_2e^{-\frac{\tau^*}{3}}$ are very separated for $\tau^*\to-\infty$ we are using the value of $S\left(\rho_1,y_1,0\right)$ in the definition of $\tilde{S}$, and the difference between $r_L\left(\rho_1,\tau\right)$, $\bar{r}_L\left(\rho_1,\tau\right)$ for τ of order one could give some contribution. Therefore, we need some additional computations. Let us use the notation

$$
\begin{aligned}
&\tilde{S}_1=\tilde{S}\left(r_L\left(\rho_1,\tau^*\right),y_1e^{-\frac{\tau^*}{3}},\tau^*\right)\quad,\quad\tilde{S}_2=\tilde{S}\left(r_L\left(\rho_2,\tau^*\right),y_2e^{-\frac{\tau^*}{3}},\tau^*\right)\\
&S_1=S\left(r_L\left(\rho_1,\tau^*\right),y_1e^{-\frac{\tau^*}{3}},\tau^*\right)\quad,\quad S_2=S\left(r_L\left(\rho_2,\tau^*\right),y_2e^{-\frac{\tau^*}{3}},\tau^*\right)\\
&\Psi_1=\Psi\left(r_L\left(\rho_1,\tau^*\right)\right)\quad,\quad\Psi_2=\Psi\left(r_L\left(\rho_2,\tau^*\right)\right)
\end{aligned}
$$

We then need to compute

$$
\begin{aligned}
\left\langle\left(\tilde{S}_1-\Psi_1\right)\left(\tilde{S}_2-\Psi_2\right)\right\rangle&=\left\langle\left(\left(\tilde{S}_1-S_1\right)-\left(\Psi_1-S_1\right)\right)\left(\left(\tilde{S}_2-S_2\right)-\left(\Psi_2-S_2\right)\right)\right\rangle\\
&=\left\langle\left(\tilde{S}_1-S_1\right)\left(\tilde{S}_2-S_2\right)\right\rangle-\left\langle\left(\tilde{S}_1-S_1\right)\left(\Psi_2-S_2\right)\right\rangle-\left\langle\left(\Psi_1-S_1\right)\left(\tilde{S}_2-S_2\right)\right\rangle\\
&\quad+\left\langle\left(\Psi_1-S_1\right)\left(\Psi_2-S_2\right)\right\rangle
\end{aligned}
$$

The variables Ψ_1-S_1 and Ψ_2-S_2 are uncorrelated, and $\langle\Psi_1-S_1\rangle=\langle\Psi_2-S_2\rangle=0$. Then, the last term dissappears. In order to estimate the remaining terms we need to approximate $\left(\tilde{S}_i-S_i\right)$, $i=1,2$. Integrating by characteristics

$$
\begin{aligned}
S\left(\rho_1,y_1,0\right)&=e^{-\tau^*}S\left(\bar{r}_L\left(\rho_1,\tau^*\right),y_1e^{-\frac{\tau^*}{3}},\tau^*\right)\\
S\left(\rho_1,y_1,0\right)&=e^{-\tau^*}\tilde{S}\left(r_L\left(\rho_1,\tau^*\right),y_1e^{-\frac{\tau^*}{3}},\tau^*\right)
\end{aligned}
$$

whence

$$
S\left(\bar{r}_L\left(\rho_1,\tau^*\right),y_1e^{-\frac{\tau^*}{3}},\tau^*\right)=\tilde{S}\left(r_L\left(\rho_1,\tau^*\right),y_1e^{-\frac{\tau^*}{3}},\tau^*\right)=\tilde{S}_1
$$

and an analogous formula holds true for $\tilde{S}_2$. We introduce

$$
\varepsilon(\rho_i,y_i,\tau^*):=\bar{r}_L(\rho_i,y_i,\tau^*)-r_L(\rho_i,\tau^*),\qquad i=1,2\,.
$$

such that

$$
\begin{aligned}
\tilde{S}_1-S_1&=S\left(\bar{r}_L\left(\rho_1,\tau^*\right),y_1e^{-\frac{\tau^*}{3}},\tau^*\right)-S\left(r_L\left(\rho_1,\tau^*\right),y_1e^{-\frac{\tau^*}{3}},\tau^*\right)\\
&=\frac{\partial\Psi}{\partial\rho_1}\left(r_L\left(\rho_1,\tau^*\right)\right)\varepsilon_1\left(\rho_1,\tau^*\right)\,.
\end{aligned}
$$

Notice that it is enough to obtain the linear approximation, because all the terms above are quadratic. Hence

$$
\begin{aligned}
&\left\langle \left(\tilde{S}_1 - \Psi_1\right)\left(\tilde{S}_2 - \Psi_2\right) \right\rangle \\
&= \left\langle \left(\tilde{S}_1 - S_1\right)\left(\tilde{S}_2 - S_2\right)\right\rangle - \left\langle \left(\tilde{S}_1 - S_1\right)(\Psi_2 - S_2)\right\rangle - \left\langle (\Psi_1 - S_1)\left(\tilde{S}_2 - S_2\right)\right\rangle \\
&= \frac{\partial \Psi}{\partial \rho_1}\left(r_L\left(\rho_1, \tau^*\right)\right)\frac{\partial \Psi}{\partial \rho_2}\left(r_L\left(\rho_2, \tau^*\right)\right)\left\langle \varepsilon(\rho_1, y_1, \tau^*)\,\varepsilon_2(\rho_2, y_2, \tau^*)\right\rangle \\
&- \frac{\partial \Psi}{\partial \rho_1}\left(r_L\left(\rho_1, \tau^*\right)\right)\left\langle \varepsilon(\rho_1, y_1, \tau^*)\,(\Psi_2 - S_2)\right\rangle - \\
&- \frac{\partial \Psi}{\partial \rho_2}\left(r_L\left(\rho_2, \tau^*\right)\right)\left\langle (\Psi_1 - S_1)\,\varepsilon(\rho_2, y_2, \tau^*)\right\rangle
\end{aligned}
$$

Now $\varepsilon(\rho_1, y_1, \tau^*)$ and $\Psi_2 - S_2$ are uncorrelated, and the same is true for $\Psi_1 - S_1$ and $\varepsilon(\rho_2, y_2, \tau^*)$. The we arrive at

$$
\begin{aligned}
&\left\langle S\left(\rho_1, y_1, 0\right) S\left(\rho_2, y_2, 0\right) \right\rangle \\
&\quad = \Psi\left(\rho_1\right)\Psi\left(\rho_2\right) + \lim_{\tau^* \to -\infty} \frac{\partial \Psi}{\partial \rho_1}\left(\rho_1\right)\frac{\partial \Psi}{\partial \rho_2}\left(\rho_2\right)\left\langle \varepsilon\left(\rho_1, y_1, \tau^*\right)\varepsilon\left(\rho_2, y_2, \tau^*\right)\right\rangle \,.
\end{aligned} \tag{4.26}
$$

In the final step we compute $\left\langle \varepsilon\left(\rho_1, y_{1,0}, \tau^*\right)\varepsilon\left(\rho_2, y_{2,0}, \tau^*\right)\right\rangle$. Linearizing (4.21) we obtain

$$
\begin{aligned}
\frac{d\varepsilon\left(\rho_1, y_1, \tau\right)}{d\tau} &= \frac{\partial}{\partial r_L}\left(-\frac{1}{r_L^2} + \frac{1}{r_0}\frac{1}{r_L} - \frac{r_L}{3}\right)\varepsilon\left(\rho_1, \tau\right) + \frac{\phi^{\frac{1}{4}}\zeta\left(y, \tau\right)}{r_L} \\
\varepsilon\left(\rho_1, y_1, 0\right) &= 0
\end{aligned}
$$

whose solution is given by

$$
\begin{aligned}
\varepsilon\left(\rho_1, y, \tau\right) &= -\phi^{\frac{1}{4}}\frac{\partial r_L\left(\rho_1, \tau\right)}{\partial \rho_1}\int_{\tau}^{\bar{\tau}} \frac{\zeta\left(y\left(s\right), s\right)}{r_L\left(\rho_1, s\right)}\frac{ds}{\frac{\partial r_L}{\partial \rho_1}\left(\rho_1, s\right)} \\
y\left(s\right) &= ye^{-s/3}
\end{aligned}
$$

Hence

$$
\begin{aligned}
&\left\langle \varepsilon\left(\rho_1, \tau^*\right)\varepsilon\left(\rho_2, \tau^*\right)\right\rangle \\
&= \sqrt{\phi}\frac{\partial r_L\left(\rho_1, \tau^*\right)}{\partial \rho_1}\frac{\partial r_L\left(\rho_2, \tau^*\right)}{\partial \rho_2}\cdot \\
&\int_{\tau^*}^{\bar{\tau}} \frac{ds_1}{r_L\left(\rho_1, s_1\right)\frac{\partial r_L}{\partial \rho_1}\left(\rho_1, s_1\right)}\int_{\tau^*}^{\bar{\tau}} \frac{ds_2}{r_L\left(\rho_2, s_2\right)\frac{\partial r_L}{\partial \rho_1}\left(\rho_2, s_2\right)}\left\langle \zeta\left(y_1 e^{-s_1/3}, s_1\right)\zeta\left(y_2 e^{-s_2/3}, s_2\right)\right\rangle
\end{aligned} \tag{4.27}
$$

Using (4.24) and the invariance of ζ under translations we find

$$
\begin{aligned}
&\left\langle \zeta\left(y_1 e^{-s_1/3}, s_1\right)\zeta\left(y_2 e^{-s_2/3}, s_2\right) \right\rangle = \left\langle \zeta\left(0, s_1\right)\zeta\left(ye^{-s_2/3}, s_2\right)\right\rangle \\
&\quad = \sqrt{\phi}\xi_0 e^{\frac{\left(\bar{s}_2 - \bar{s}_1\right)}{3}}\lambda\left(y_2 e^{-s_2/3}e^{\frac{\left(\bar{s}_2 - \bar{s}_1\right)}{3}}, e^{-\frac{\left(\bar{s}_2 - \bar{s}_1\right)}{3}}\right)\int_{r_L\left(0, \bar{s}_1 - \bar{s}_2\right)}^{\infty} \Phi\left(\rho\right)d\rho
\end{aligned} \tag{4.28}
$$

where

$$
\bar{s}_1 = \min\left\{s_1, s_2\right\}\ ,\ \bar{s}_2 = \max\left\{s_1, s_2\right\}
$$

We also recall that $\sqrt{\phi}\xi_0 = (4\pi B)^{-1/2} = O(1)$.

We now use (4.27),(4.28) and the identity

$$\int_{r_L(0,\tau)}^{\infty} \Phi(\rho)\, d\rho = Ce^{\tau},$$

for some suitable normalization constant C. For sufficiently large $|\tau^*|$ we arrive after some computations at

$$\begin{aligned}\int dy_2 K(y_2) \Big\langle \varepsilon(\rho_1, y_1, \tau^*)\varepsilon(\rho_2, y_2, \tau^*) \Big\rangle &= C\left[\sqrt{\phi}\xi_0\right]\sqrt{\phi}\frac{\partial r_L(\rho_1,\tau^*)}{\partial\rho_1}\frac{\partial r_L(\rho_2,\tau^*)}{\partial\rho_2}\cdot \\ &\cdot \int_{-\infty}^{\bar{\tau}} \frac{ds_1}{r_L(\rho_1,s_1)\frac{\partial r_L}{\partial\rho_1}(\rho_1,s_1)} \int_{-\infty}^{0} \frac{ds_2 e^{2s_2/3}}{r_L(\rho_2,s_2)\frac{\partial r_L}{\partial\rho_1}(\rho_2,s_2)} e^{\frac{2(\bar{s}_1-\bar{s}_2)}{3}} \\ &\cdot \left[\int\int \frac{e^{-|z|e^{\frac{(\bar{s}_2-\bar{s}_1)}{3}}}}{|z|}\frac{e^{-|\lambda-z|}}{|\lambda-z|}K(\lambda)\, dz d\lambda\right]\end{aligned} \tag{4.29}$$

We can simplify this formula for $\rho_1 \approx \rho_{LSW}$. Indeed, in such a region $r_L(\rho_1, s_1) \approx \rho_{LSW}$ and $\frac{\partial r_L}{\partial\rho_1}(\rho_1, s_1) \approx 1$. Then, combining (4.26) and (4.29), we find

$$I(\rho_1) = \frac{\partial\Phi(\rho_1)}{\partial\rho_1}\int\left[\frac{\partial}{\partial\rho_2}\left[\Phi(\rho_2)Q(\rho_2)\right]\right]\left(1-\frac{\rho_2}{r_0}\right)d\rho_2 \tag{4.30}$$

where

$$\begin{aligned}Q(\rho_2) :=\sim \frac{C}{\rho_{LSW}}\left[\sqrt{\phi}\xi_0\right]\sqrt{\phi}\frac{\partial r_L(\rho_2,\tau^*)}{\partial\rho_2}\cdot \\ \cdot\int_{-\infty}^{\bar{\tau}} ds_1 \int_{-\infty}^{\bar{\tau}} \frac{ds_2 e^{-\frac{2(\bar{\tau}-s_2)}{3}}}{r_L(\rho_2,s_2)\frac{\partial r_L}{\partial\rho_1}(\rho_2,s_2)} e^{\frac{2(\bar{s}_1-\bar{s}_2)}{3}} \left[\int\int \frac{e^{-|z|e^{\frac{(\bar{s}_2-\bar{s}_1)}{3}}}}{|z|}\frac{e^{-|\lambda-z|}}{|\lambda-z|}K(\lambda)\, dz d\lambda\right].\end{aligned}$$

After integrating by parts we find

$$I(\rho_1) = \frac{1}{r_0}\frac{\partial\Phi(\rho_1)}{\partial\rho_1}\left[\int\Phi(\rho_2)Q(\rho_2)\, d\rho_2\right]$$

Since $\xi_0\sqrt{\phi} \sim 1$ it seem that Q is of order $\sqrt{\phi}$. Notice however, that $\frac{\partial r_L(\rho_2,\tau^*)}{\partial\rho_2}$ converges to 0 as $\tau^* \to -\infty$ if $\rho_2 < \rho_{LSW}$. Then $\int\Phi(\rho_2)Q(\rho_2)\, d\rho_2 = o(\sqrt{\phi})$, whence this term is negligible in (4.25).

4.3 Perturbative analysis of self-similar solutions

The results of the previous subsection show that it suffices to study solutions of

$$\begin{aligned}-\frac{4}{3}\Phi(\rho) - \frac{1}{3}\rho\frac{d\Phi(\rho)}{d\rho} - \frac{d}{d\rho}\left(\left(\frac{1}{(\rho)^2}-\frac{1}{r_0\rho}\right)\Phi(\rho)\right) \\ = \frac{d}{d\rho}\left(\left[\frac{\sqrt{\phi}}{\sqrt{4\pi B}}\frac{1}{\rho}\int_0^1 \frac{\int_{I\!R^3}(\omega(\chi,y)k(y))\, dy}{(\chi)^{2/3} r_L(\chi,\rho)\frac{\partial r_L(\chi,\rho)}{\partial\rho}}d\chi\right]\frac{d\Phi(\rho)}{d\rho}\right)\end{aligned} \tag{4.31}$$

We are are now going to construct solutions of (4.31) that are perturbations of the LSW self-similar solutions. In fact, the appearance of the other self-similar solutions to leading order can be ruled out in principle by the argument already given in [6]. In that case the structure of the characteristic curves in self-similar variables implies that a fraction of the particles would remain trapped in some region of the form $\{R \geq at^{1/3}\}$. This is however incompatible with the conservation of volume of particles.

Self-similar solutions satisfy the equation

$$-\frac{4}{3}\Phi(\rho) - \frac{1}{3}\rho\frac{d\Phi(\rho)}{d\rho} - \frac{d}{d\rho}\left(\left(\frac{1}{\rho^2} - \frac{1}{r_0\rho}\right)\Phi(\rho)\right) = 0 \tag{4.32}$$

Let us denote by $\Phi_{LSW}(\rho)$ the solution of (4.32) with maximal support. Therefore

$$\bar{r}_0 = \left(\frac{2}{3}\right)^{\frac{2}{3}}$$

$$\Phi_{LSW}(\rho) = \alpha\frac{\rho^2 \exp\left(-\frac{\rho}{\rho_{LSW}-\rho}\right)}{\left(1+\frac{\rho}{2\rho_{LSW}}\right)^{7/3}\left(1-\frac{\rho}{\rho_{LSW}}\right)^{\frac{11}{3}}} \tag{4.33}$$

where

$$\rho_{LSW} = \left(\frac{3}{2}\right)^{\frac{1}{3}}$$

and where $\alpha > 0$ is a constant such that (4.3) is satisfied. We define

$$D(\rho) := \frac{1}{\sqrt{4\pi B}}\int_0^1 \frac{\int_{\mathbb{R}^3}(\omega(\chi, y)\,k(y))\,dy}{(\chi)^{2/3}\,r_L(\chi,\rho)\,\frac{\partial r_L(\chi,\rho)}{\partial\rho}}d\chi \tag{4.34}$$

In order to be able to apply perturbative arguments it is crucial to determine if the function $D(\rho)$ is positive at least in a neighborhood of $\rho \approx \rho_{LSW}$.

It turns out that the proof of positivity is somewhat tedious. In Appendix B we present a method to reformulate the problem such that it can be solved numerically in an efficient way. Simulations indeed show, that D is positive and has the form as shown in Figure 1.

4.4 Boundary layer structure

In this section we study the solution $\Phi(\rho)$ of (4.25) in the limit $\phi \to 0$ using asymptotic WKB methods. Combining (4.25) and (4.34) we obtain

$$-\frac{4}{3}\Phi(\rho) - \frac{1}{3}\rho\frac{d\Phi(\rho)}{d\rho} - \frac{d}{d\rho}\left(\left(\frac{1}{(\rho)^2} - \frac{1}{r_0\rho}\right)\Phi(\rho)\right) = \sqrt{\phi}\,\frac{d}{d\rho}\left(\left[\frac{D(\rho)}{\rho}\right]\frac{d\Phi(\rho)}{d\rho}\right) \tag{4.35}$$

In the region where $\Phi(\rho)$ is of order one we can approximate the solution of (4.35) by Φ_{LSW} as given in (4.33).

Integrating (4.35) and using (4.19) we obtain

$$-\Psi(\rho) - \left(\frac{1}{3}\rho + \frac{1}{\rho^2} - \frac{1}{r_0\rho}\right)\frac{d\Psi(\rho)}{d\rho} = \sqrt{\phi}\left(\left[\frac{D(\rho)}{\rho}\right]\frac{d^2\Psi(\rho)}{d(\rho)^2}\right) \tag{4.36}$$

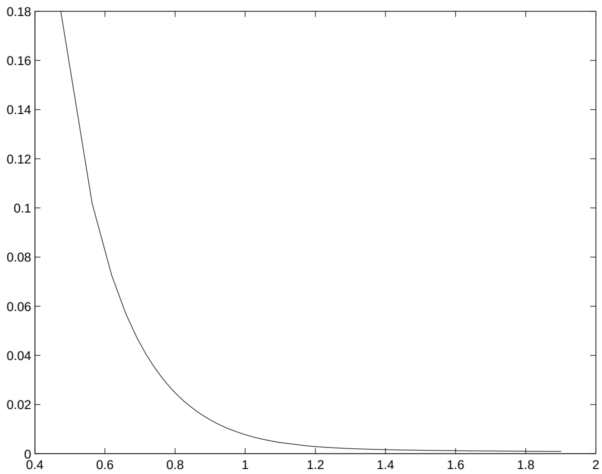

Figure 1: Diffusion coefficient

To leading order r_0 can be approximated as $\left(\frac{2}{3}\right)^{2/3}$. However, the presence of a boundary layer for $\rho_1 \approx \rho_{LSW}$ introduces a small correction in the value of r_0. We write

$$r_0 = \bar{r}_0 + \phi^{1/4}\, r_1 \tag{4.37}$$

where $\bar{r}_0 = \left(\frac{2}{3}\right)^{2/3}$.

In order to study the behaviour of the solutions of (4.36) away from the critical region $\rho \approx \rho_{LSW}$ it is convenient to introduce the WKB change of variables

$$\Psi\left(\rho\right) = \exp\left(\phi^{-1/2} U\left(\rho\right)\right) \tag{4.38}$$

such that

$$-1 - \left(\frac{1}{3}\rho + \frac{1}{\rho^2} - \frac{1}{r_0\rho}\right)\frac{U'(\rho)}{\sqrt{\phi}} = \frac{1}{B}\frac{D(\rho)}{\rho}\left(U''(\rho) + \frac{U'(\rho)^2}{\sqrt{\phi}}\right). \tag{4.39}$$

We see that there are two possibilities for U. Either $U \sim O(\sqrt{\phi})$, then

$$1 + \left(\frac{1}{3}\rho + \frac{1}{\rho^2} - \frac{1}{r_0\rho}\right)\frac{U'(\phi)}{\sqrt{\phi}} = 0\,, \tag{4.40}$$

or $U \sim O(1)$ where

$$-\left(\frac{1}{3}\rho + \frac{1}{\rho^2} - \frac{1}{r_0\rho}\right)U'(\phi) = \sqrt{\phi}\left[\frac{D\left(\rho\right)}{\rho}\right]\left(U'(\phi)\right)^2. \tag{4.41}$$

For $\rho > \rho_{LSW}$ we do not have physically reasonable solutions of (4.40). In fact, it is easily seen that $U(\rho) \sim -\sqrt{\phi}\frac{\ln\rho}{3}$ for $\rho \to \infty$, whence $\Psi(\rho) \sim \frac{1}{\rho^3}$ and thus $\int \rho^2\Psi(\rho)\, d\rho$ is not finite. Therefore the asymptotics of the solutions is given by (4.41) for supercritical particles. Taking into account (4.19) we obtain the following approximation of $\Phi\left(\rho\right)$ for $\rho > \rho_{LSW}$

$$\Phi\left(\rho\right) = \beta\exp\left(-\frac{1}{\sqrt{\phi}}\int_{\rho_{LSW}}^{\rho}\frac{\lambda}{D\left(\lambda\right)}\left[\frac{1}{3}\lambda + \frac{1}{\lambda^2} - \frac{1}{r_0\lambda}\right]d\lambda\right) \tag{4.42}$$

for some suitable constant β. Notice that the resulting solution decays exponentially fast as it could be expected.

We are going to show that there is a unique value of r_1, such that the solution in (4.42) can be matched with Φ_{LSW} as given in (4.33). In the transition region we have $\rho \approx \rho_{LSW}$ and using Taylor's expansion we obtain with (4.37) the following approximation for (4.36)

$$-\Psi(\rho) - \left(\left(\frac{2}{3}\right)^{1/3} (\rho - \rho_{LSW})^2 + \frac{\phi^{1/4} r_1}{\rho_{LSW} (\bar{r}_0)^2} \right) \frac{d\Psi(\rho)}{d\rho} = \sqrt{\phi} \left(\left[\frac{D(\rho_{LSW})}{\rho_{LSW}} \right] \frac{d^2 \Psi(\rho)}{d(\rho_1)^2} \right) \tag{4.43}$$

We now introduce the change of variables

$$\rho - \rho_{LSW} = (\phi)^{1/8} x \qquad\qquad S = \phi^{-3/8} U\,. \tag{4.44}$$

Then, (4.43) becomes

$$A(\phi)^{1/8} S_{xx} + A(S_x)^2 + \left[\left(\frac{2}{3}\right)^{1/3} x^2 + \Gamma_0 \right] S_x + 1 = 0 \tag{4.45}$$

where

$$\Gamma_0 = \frac{r_1}{\rho_{LSW} (\bar{r}_0)^2} \qquad \text{and} \qquad A = \left[\frac{D(\rho_{LSW})}{\rho_{LSW}} \right].$$

This equation can be approximated to leading order, away from boundary layers, by

$$A(S_x)^2 + \left[\left(\frac{2}{3}\right)^{1/3} x^2 + \Gamma_0 \right] S_x + 1 = 0 \tag{4.46}$$

The solution of (4.46) that matches with the solution of (4.40) in the region where $\phi^{1/8} \ll (\rho_{LSW} - \rho) \ll 1$, is

$$\tilde{S}_x = \frac{1}{2A} \left[- \left[\left(\frac{2}{3}\right)^{1/3} x^2 + \Gamma_0 \right] + \sqrt{ \left[\left(\frac{2}{3}\right)^{1/3} x^2 + \Gamma_0 \right]^2 - 4A } \right] \tag{4.47}$$

Notice that $S_x \sim -\left(\frac{3}{2}\right)^{1/3} \frac{1}{x^2}$ as $x \to -\infty$.

We argue now that if follows from (4.47) that $4A \geq (\Gamma_0)^2$. Indeed, otherwise the function S_x in (4.47) is smooth for any $x \in I\!R$ and has the asymptotics $S \sim C + \left(\frac{3}{2}\right)^{1/3} \frac{1}{x}$ as $x \to \infty$. Such a solutions matches in the region $(\rho - \rho_{LSW}) << 1$, $(\rho - \rho_{LSW}) >> (\phi)^{1/8}$ with a nontrivial solution of (4.40) which is not possible as explained before. Therefore, in the limit $\phi \to 0$ we must have $4A \geq (\Gamma_0)^2$. Let us now examine the case in which $4A \sim (\Gamma_0)^2$ as $\phi \to 0$, since a similar argument will rule out the possibility $4A > (\Gamma_0)^2$. To this end we define a new variable δ as

$$\Gamma_0 = (4A)^{\frac{1}{2}} + \delta$$

where $\delta \to 0$ as $\phi \to 0$. We define a new set of variables by

$$x = (A)^{3/8} \left(\frac{3}{2}\right)^{1/8} \phi^{1/16} X \tag{4.48}$$

$$\sqrt{A} S_x + 1 = (A)^{1/8} \left(\frac{2}{3}\right)^{1/8} \phi^{1/16} \psi \tag{4.49}$$

Then (4.45) becomes to leading order

$$\psi_X + (\psi)^2 - X^2 = \sigma := \left(\frac{3}{2}\right)^{1/4} \frac{\delta}{(A)^{3/4}(\phi)^{1/8}} \tag{4.50}$$

with the matching condition, as a consequence of (4.47), which reads

$$\psi \sim |X| \quad \text{as} \quad X \to -\infty \tag{4.51}$$

An analysis of the phase portrait shows for any value of σ there is a unique solution of (4.50) and (4.51). There also exists for any σ a unique solution of (4.50) with the asymptotics

$$\psi \sim -X \quad \text{as} \quad X \to \infty \tag{4.52}$$

It turns out that the only value of σ for which the solution satisfies both, equations (4.51) and (4.52), is $\sigma = -1$. This can be seen with the change of variables $\psi(x) = -x + \phi(x)$. Then (4.50) becomes

$$\phi_x = 2x\phi - \phi^2 + \sigma + 1$$

and we see that the only value for which $\phi(x) \to 0$ as $|x| \to \infty$ is for $\sigma = -1$.

After the transition described above the resulting solution matches with the behaviour

$$S_x = \frac{1}{2A}\left[-\left[\left(\frac{2}{3}\right)^{1/3} x^2 + \Gamma_0\right] - \sqrt{\left[\left(\frac{2}{3}\right)^{1/3} x^2 + \Gamma_0\right]^2 - 4A}\right]$$

and this behaviour yields a exponential decay according to (4.41). To leading order

$$\Psi = \gamma \exp\left(-\frac{1}{3A}\left(\frac{2}{3}\right)^{1/3} \frac{(\rho - \rho_{LSW})^3}{\sqrt{\phi}}\right)$$

as $(\phi)^{1/8} << \rho - \rho_{LSW} << 1$, where γ is a multiplicative constant which can be determined by the higher order terms in the matched asymptotic expansion described above.

5 Appendices

5.1 Appendix A: Change in capacity coefficients

In order to approximately evaluate the second term in (3.25) we compute the difference $\left[\frac{C_{i,j}}{4\pi R_i R_j} - \frac{C_{i,j}^{(2)}}{4\pi R_i^{(2)} R_j^{(2)}}\right]$ for $i \neq j$. This difference in the capacity coefficients is due to two different effects, namely the presence in the computation of the coefficients $C_{i,j}$ of an additional particle η_2, and the difference on the radii of the remaining particles. In order to measure these effects we make the dependence on the radii explicit by writing

$$\begin{aligned} &\frac{C_{i,j}}{4\pi R_i R_j} - \frac{C_{i,j}^{(2)}}{4\pi R_i^{(2)} R_j^{(2)}} \\ &= \frac{1}{4\pi R_i R_j}\left[C_{i,j}(\{R_k\}) - C_{i,j}^{(2)}(\{R_k\})\right] + \left[\frac{C_{i,j}^{(2)}(\{R_k\})}{4\pi R_i R_j} - \frac{C_{i,j}^{(2)}\left(\left\{R_k^{(2)}\right\}\right)}{4\pi R_i^{(2)} R_j^{(2)}}\right] \end{aligned} \tag{5.1}$$

In order to compute the first term on the right-hand side of (5.1) let us denote as v the difference of the potentials associated to the computation of the capacities $C_{i,j}(\{R_k\})$ and $C_{i,j}^{(2)}(\{R_k\})$. This potential vanishes at the boundary of all the particles except the particle η_2. Taking into account (2.16) and (2.17) we find

$$v = -4\pi R_i G(x_i - x_2) \quad \text{at } \partial B_{R_2}(x_2)$$

and thus

$$\begin{aligned} C_{i,j}(\{R_k\}) - C_{i,j}^{(2)}(\{R_k\}) &= -\int_{\partial B_j} \frac{\partial (v_j - v_j^{(2)})}{\partial n} dS \\ &= -\int_{\partial B_j} \frac{\partial (v_j - v_j^{(2)})}{\partial n} v_j \, dS \\ &= \int_{\Omega \setminus \cup B_i} \nabla v \cdot \nabla v_j \\ &= -\int_{\partial B_2} \frac{\partial v_j}{\partial n} v \, dS \\ &\sim C_{2,j} v = 4\pi R_i C_{2,j} G(x_i - x_2) \ . \end{aligned} \tag{5.2}$$

Using the approximation (3.26) we find

$$C_{i,j}(\{R_k\}) - C_{i,j}^{(2)}(\{R_k\}) = (4\pi R_i)(4\pi R_2)(4\pi R_j) G(x_i - x_2) G(x_2 - x_j) \quad , \quad i \neq j \, . \tag{5.3}$$

To treat the last term in (5.1) we need to compute the change in the capacity coefficients $C_{i,j}^{(2)}(\{R_k\})$ due to the change of the radii. Let us suppose that we modify just the radius of a single particle $R_k \to R_k + \delta R_k$ where for the moment $k \neq i, j$. The difference of the potentials associated to the corresponding capacity coefficients, denoted by v, vanishes at all the particles except at the boundary of the particle η_k. Near the particle x_k the potential associated to the capacity coefficient $C_{i,j}^{(2)}(\{R_k\})$ can be approximated by $v = \frac{C_{i,k}^{(2)}}{4\pi}\left(\frac{1}{|x - x_k|} - \frac{1}{R_k^{(2)}}\right)$ such that

$$v = \frac{C_{i,k}^{(2)}}{4\pi \left(R_k^{(2)}\right)^2} \delta R_k \ \text{ at } \ \partial B_{R_k}(x_k) \ ,$$

whence the charge induced at the particle η_j by this change of the radius is

$$C_{k,j}^{(2)} \frac{C_{i,k}^{(2)}}{4\pi (R_k)^2} \delta R_k \quad , \ k \neq i, j, 2 \tag{5.4}$$

Next, we are going to show that the change of the magnitude $\frac{C_{ij}}{4\pi R_i R_j}$ under changes of the radii R_j and R_i are quadratic in ∂R_j and ∂R_i. This can be expected, since in view of (2.18) we see that the quantity $\frac{C_{ij}}{4\pi R_i R_j}$ basically does not depend on R_i and R_j.

Indeed, let $\bar{u}_i$ be the potential for B_{R_i} and u_i be the potential for $B_{R_i + \delta R_i}$, such that $\bar{u}_i \sim \frac{R_i}{|x - X_i|}$ and $u_i \sim \frac{R_i + \delta R_i}{|x - X_i|}$ close to ∂B_{R_i}. Hence, we find that the difference $v = u_i - \bar{u}_i$ approximately satisfies $\frac{\delta R_i}{R_i}$ on ∂B_{R_i} and vanishes in all the other particles. Consequently, arguing similarly as in (5.2) above, we find that the change induced in $C_{i,i}^{(2)}$ is $C_{i,i}^{(2)} \frac{\delta R_i}{R_i}$.

Similarly the difference in potentials corresponding to $C_{i,j}$ when R_j is changed to $R_j + \delta R_j$ satisfies approximately $\frac{C_{i,j}\delta R_j}{4\pi R_j^2}$, whence the change in the capacity coefficient is $C_{j,j}\frac{C_{i,j}\delta R_j}{4\pi R_j^2} \sim \frac{C_{i,j}\delta R_j}{R_j}$. In summary, the change of $\frac{C_{ij}}{4\pi R_i R_j}$ is given by

$$\frac{C_{ij}}{4\pi R_i R_j} - \frac{C_{i,j}\left(1+\frac{\delta R_i}{R_i}+\frac{\delta R_j}{R_j}\right)}{4\pi\left(1+\frac{\delta R_i}{R_i}\right)\left(1+\frac{\delta R_j}{R_j}\right)} \sim \frac{C_{i,j}}{4\pi R_i R_j}\frac{\delta R_i}{R_i}\frac{\delta R_j}{R_j}$$

and is thus quadratic in $\delta R_i, \delta R_j$.

Therefore, in order to compute the last term in (5.1) it is enough to add the contributions due to the changes in the radii δR_k with $k \neq i, j, 2$. Then to leading order

$$\left[\frac{C_{i,j}^{(2)}\left(\{R_k\}\right)}{4\pi R_i R_j} - \frac{C_{i,j}^{(2)}\left(\left\{R_k^{(2)}\right\}\right)}{4\pi R_i^{(2)} R_j^{(2)}}\right] = \sum_{k\neq i,j,2} \frac{C_{i,k}^{(2)} C_{k,j}^{(2)}}{4\pi\left(R_k^{(2)}\right)^2} \frac{r_k}{4\pi R_i^{(2)} R_j^{(2)}} \tag{5.5}$$

and combining (5.1), (5.3) and (5.5) we obtain

$$\frac{C_{i,j}}{4\pi R_i R_j} - \frac{C_{i,j}^{(2)}}{4\pi R_i^{(2)} R_j^{(2)}} = \left(4\pi R_2\right)\left(4\pi G\left(x_i - x_2\right)\right) G\left(x_2 - x_j\right) + \tag{5.6}$$

$$+\sum_{k\neq i,j,2} \frac{C_{i,k}^{(2)} C_{k,j}^{(2)}}{4\pi\left(R_k^{(2)}\right)^2} \frac{r_k}{4\pi R_i^{(2)} R_j^{(2)}} \quad , \quad i \neq i, j$$

5.2 Appendix B: Positivity of the diffusion coefficient

In this appendix we sketch a procedure to transform the original problem (4.16) and (4.25) which determine the coefficient $D(\rho)$ (cf. (4.34)) into an equation which is more convenient to solve numerically.

To that aim it is convenient to introduce

$$J_1(\rho) = \frac{4}{3}\log\left(1+\frac{\rho}{2\rho_{LSW}}\right) + \frac{5}{3}\log\left(1-\frac{\rho}{\rho_{LSW}}\right) + \frac{\rho}{\left(\rho_{LSW}-\rho\right)}\,. \tag{5.7}$$

Using this function the equations for the characteristics in self-similar variables take the simple form

$$J_1\left(r_{LSW}\left(\tau,\rho\right)\right) - J_1\left(\rho\right) = -\tau \tag{5.8}$$

where $J_1(\rho)$ is as in (5.7).

We can now transform (4.16) making the following changes of variables $z = r_{LSW}(\tau,\rho)$, $dz = \frac{\partial r_{LSW}(\tau,\rho)}{\partial\rho}d\rho$. After introducing this change of variables in (4.16) we take the Fourier transform with respect to η. Then we obtain after some lengthy computations we obtain

$$D(\rho) \equiv \frac{1}{2\pi^2}\int_{-\infty}^{0}\left(\frac{l\left(J_1^{-1}\left(J_1(\rho)-s\right)\right)}{J_1'(\rho)} e^{\frac{s}{3}} W(s)\right) ds\,, \tag{5.9}$$

where

$$l(X) \equiv \frac{J_1'(X)}{X} \tag{5.10}$$

$$W(s) \equiv \int_{-\infty}^{s}\left(e^{-\frac{2(s-\tau)}{3}}\left(1-\frac{J_1^{-1}(s-\tau)}{r_0}\right)\int_0^{\infty} f(\tau,r)\,dr\right)d\tau \tag{5.11}$$

and f is the solution of

$$\left(1+r^2 e^{\frac{2\tau}{3}}\right) f(\tau,r)+\int_{-\infty}^{\tau} G(\tau-s)\, f(s,r)\, ds = e^{\frac{2\tau}{3}}\left(1-\frac{J_1^{-1}(-\tau)}{r_0}\right) \tag{5.12}$$

where

$$G(\tau)=\frac{e^{-\frac{\tau}{3}}}{r_0}\int_0^{\infty}\frac{e^{-s}J_1'\left(J_1^{-1}(s+\tau)\right)}{J_1^{-1}(s+\tau)}\frac{ds}{J_1'\left(J_1^{-1}(s)\right)}\,. \tag{5.13}$$

Formula (5.9) is valid for $\rho<\rho_{LSW}$. In the region $\rho>\rho_{LSW}$ the computation is similar with J_1 replaced by J_2 given by

$$J_2(\rho)=\frac{4}{3}\log\left(1+\frac{\rho}{2\rho_{LSW}}\right)+\frac{5}{3}\log\left(\frac{\rho}{\rho_{LSW}}-1\right)+\frac{\rho}{(\rho_{LSW}-\rho)} \tag{5.14}$$

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