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On First-Order Corrections to the LSW Theory II: Finite Systems

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We consider first–order corrections to the classical theory by Lifshitz, Slyozov and Wagner (LSW) for systems with a finite number of particles. Numerical simulations in V. E Fradkov *et al.* [*Phys. Rev. E* **53**:3925–3932 (1996)] show a cross–over in the scaling of the correction term from $\phi^{1/3}$ to $\phi^{1/2}$ (ϕ is the volume fraction of particles), when the system size becomes larger than the screening length. We rigorously derive this cross–over for the rate of change of the energy, starting from the monopole approximation. The proof exploits the fact that the rate of change of energy has a variational characterization.

KEY WORDS: Ostwald ripening; monopole approximation; stochastic homogenization.

1. INTRODUCTION

Ostwald ripening denotes the last stage of a phase separation in an off–critical binary mixture, where precipitate particles undergo competitive growth to reduce their total surface energy. In the regime of small volume fraction of particles the classical theory of Lifshitz, Slyozov and Wagner[(1,2)] provides an evolution law for the radii of the particles. It is based on the assumption that particles only interact via a common mean field.

However, the quantitative predictions of the LSW theory deviate from standard experiments.[(3)] It is generally conjectured that this deviation is due to the fact that the volume fraction ϕ of the particles is small but

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finite. Hence in order to extend the range of validity of the LSW theory, it is of large interest to identify a first–order correction in ϕ.

In our companion paper,[4] to which we refer in the following as Part I, we describe in detail the scenario of Ostwald ripening, we examine several aspects of the LSW theory and give an overview of the physics literature on this topic. The main aim of Part I is to present a novel approach, to identify the first-order correction to the LSW theory in statistically homogeneous infinite systems. In this case the first-order correction is of order $\phi^{1/2}$.

In the present paper, Part II, we present a mathematically rigorous analysis for systems with a finite number of particles, where the argument in Part I for the $\phi^{1/2}$–scaling does not apply. Indeed, Fradkov *et al.*[5,6] have numerically observed a cross–over in the scaling of the first-order correction term for finite systems. It changes from $\phi^{1/3}$ to $\phi^{1/2}$ when the system size becomes larger than the screening length. The screening length is the important intrinsic length scale in the system and describes the effective range of particle interactions. It is proportional to the typical interparticle distance times $\phi^{-1/6}$ (see Part I for more details). We will rigorously establish this numerically observed cross-over under the assumptions used in the numerical simulations.[5] That is we use the monopole approximation and the snapshot perspective, varying just the number n of particles in the system at given volume fraction ϕ.

2. THE SETTING OF THE PROBLEM AND THE MAIN RESULT

2.1. The Monopole Approximation and the LSW Solution

Late–stage coarsening is well–described by the Mullins–Sekerka problem, which in dimensionless variables is given by

$$V = [\nabla u \cdot \vec{n}] \qquad \text{on the particle–matrix interface,} \tag{2.1}$$

$$-\Delta u = 0 \qquad \text{in the bulk,} \tag{2.2}$$

$$u = \kappa \qquad \text{on the interface.} \tag{2.3}$$

Here V denotes the normal velocity of the interface, $\vec{n}$ its normal, pointing into the majority phase, u is a dimensionless diffusion potential $[\nabla u \cdot \vec{n}]$ denotes the jump of the normal component of the gradient across the interface and κ denotes the mean curvature of the interface, defined to be positive if the minority phase forms a ball. Equation (2.1) is the kinematic Stefan condition, (2.2) means that the potential is in quasistatic equilibrium and (2.3) is the well–known Gibbs–Thomson law which accounts for surface tension. This evolution decreases the total surface area while it keeps the volume fraction of each phase conserved. It also has an interpretation as a gradient flow: it is the gradient flow of the (isotropic) surface energy with respect to the H^{-1}-norm in the bulk.

For small volume fraction ϕ of $n \gg 1$ particles, the particles are approximately balls with radius R_i and fixed center X_i, $i = 1, \ldots, n$. This can be observed in experiments and has been worked out in a rigorous manner for solutions of the Mullins–Sekerka problem in refs. 7, 8. Thus, a natural Ansatz for u is

$$u(x) = u_\infty + \sum_j \frac{B_j}{|x - X_j|}, \tag{2.4}$$

where $\{4\pi B_i\}_i$ are the growth rates of the particle volumes, that is

$$-B_i := \frac{d}{dt}\left[\frac{1}{3} R_i^3\right] = R_i^2 \frac{dR_i}{dt}. \tag{2.5}$$

The constant u_∞ is called the "mean field" and is determined by the constraint that the volume fraction of particles is conserved which enforces

$$\sum_i B_i = 0. \tag{2.6}$$

From the Gibbs–Thomson law (2.3) we obtain

$$\frac{1}{R_i} \approx u_\infty + \frac{B_i}{R_i} + \sum_{j \neq i} \frac{B_j}{d_{ij}},$$

where $d_{ij} := |X_i - X_j|$ denotes the distance between particle centers. Thus, we can view the mean field u_∞ and the growth rates $\{B_i\}_i$ as the solution of the linear system of equations

$$\frac{1}{R_i} = u_\infty + \frac{B_i}{R_i} + \sum_{j \neq i} \frac{B_j}{d_{ij}} \tag{2.7}$$

under the constraint (2.6). Equations (2.5), (2.6) and (2.7) are the monopole approximation of the Mullins–Sekerka problem. It has been widely used in the applied literature and will also be the starting point of our analysis. We refer to Part I for a more detailed description and some remarks on its limitations.

The classical LSW solution, which we denote by $\{B_i^{\mathrm{LSW}}\}_i$, is now given by the truncation of (2.7)

$$\frac{1}{R_i} = u_\infty^{\mathrm{LSW}} + \frac{B_i^{\mathrm{LSW}}}{R_i}, \tag{2.8}$$

which together with (2.6) yields

$$B_i^{\mathrm{LSW}} = 1 - R_i\, u_\infty^{\mathrm{LSW}} \quad \text{and} \quad u_\infty^{\mathrm{LSW}} = \frac{\sum_i 1}{\sum_i R_i}. \tag{2.9}$$

In particular, the LSW mean field is given by the inverse of the mean radius of particles.

2.2. Numerical Simulations

Fradkov *et al.*[5,6] were the first who systematically investigated the cross-over in the scaling from small to large systems. Their starting point was the monopole approximation and the "snapshot" perspective.

The "snapshot" perspective means the following. One considers a finite system $\{(X_i, R_i)\}_{1\leqslant i\leqslant n}$ with $n \gg 1$, where the R_i are independently and identically distributed. The distribution of the R_i is usually chosen to be the self-similar distribution of radii from the LSW theory. The distribution of positions X_i is chosen to be uniform within a sphere except that overlap is excluded. Then the growth rates $\{B_i\}_{1\leqslant i\leqslant n}$ are determined numerically according to (2.7) for many realizations of the radii keeping the positions fixed. Finally, it is analyzed how the $\{B_i\}_{1\leqslant i\leqslant n}$ deviate from the LSW growth rates $\{B_i^{\mathrm{LSW}}\}_{1\leqslant i\leqslant n}$ given by (2.9). No time integration is performed – therefore the name "snapshot".

Motivated by the analysis of Marqusee and Ross[9] Fradkov *et al.* measured the deviation of $\{B_i\}_{1\leqslant i\leqslant n}$ from $\{B_i^{\mathrm{LSW}}\}_{1\leqslant i\leqslant n}$ by the expected relative deviation in the rate of change of the mean radius

$$\Delta = \frac{\langle \frac{1}{n} \sum_i \frac{1}{R_i^2} (B_i - B_i^{\mathrm{LSW}}) \rangle}{\langle \frac{1}{n} \sum_i \frac{1}{R_i^2} B_i^{\mathrm{LSW}} \rangle}. \tag{2.10}$$

In order to calculate the expectation value Fradkov *et al.* averaged the deviation Δ of the coarsening rate over many realizations of the radii. For example, the number of realizations used in ref. 6 was such that an ensemble of least of a million particles was involved in the calculation at a studied volume fraction.

Fradkov *et al.* found a cross-over between a $\phi^{1/3}$ scaling and a $\phi^{1/2}$ scaling at a cross-over volume fraction $\phi^* \approx 1/3n^2$, which corresponds to the point when the system size becomes of the order of the screening length. An investigation of their numerical results (graphically, see Fig. 4 in ref. 6) shows that the deviation of the coarsening rate is independent of the number of particles when the system size is larger the screening length. Furthermore they found that the numerical results were relatively insensitive to the distribution of radii employed for volume fractions smaller than 0.01.

2.3. The Formulation of the Problem

To our knowledge, this numerically observed cross-over has not yet been reproduced by any type of analysis, which is the goal of our paper. We will reproduce this cross-over under assumptions used in the numerical simulation,[5] that is, the monopole approximation and the snapshot perspective.

Let us discuss what is to be expected in terms of our *two* nondimensional parameters ϕ and n. As argued in Section 2.1 of Part I, the cross-over should occur when the system size is of the order of the screening length. Since

$$\frac{\text{system size}}{\text{typical particle distance}} \sim n^{1/3},$$
$$\frac{\text{screening length}}{\text{typical particle distance}} \sim \phi^{-1/6},$$

we expect the cross-over in the deviation term at

$$n \sim \phi^{-1/2}. \tag{2.11}$$

The typical particle distance is here defined as

$$\left(\frac{3}{4\pi}\,\frac{\text{volume of the system}}{\text{number of particles}}\right)^{1/3}.$$

We denote in the following particle systems as “subcritical systems” or “supercritical systems” if they are smaller or larger than the screening length respectively.

The heuristic arguments in Section 2.5 of Part I and the numerical simulations in ref. 5, 6 suggest that the deviation in a supercritical system should scale as

$$\Delta \sim \phi^{1/2} \quad \text{for} \quad n \gg \phi^{-1/2}. \tag{2.12}$$

Furthermore, in Section 2.2. of Part I we gave some heuristic arguments that the deviation term for subcritical systems should scale as $\phi^{1/3}$. Therefore, both scalings coincide at the cross-over $n \sim \phi^{-1/2}$ when the deviation term in a subcritical system scales as

$$\Delta \sim n^{-1/3} \phi^{1/3} \quad \text{for} \quad n \ll \phi^{-1/2}. \tag{2.13}$$

Instead of considering the expected relative deviation in the rate of change of the mean radius (2.10) we will investigate the relative deviation in the rate of change of energy:

$$\frac{\dot{E}^{\mathrm{LSW}} - \dot{E}}{|\langle \dot{E}^{\mathrm{LSW}} \rangle|}. \tag{2.14}$$

We recall that the average (interfacial) energy is given by

$$E = \frac{1}{2n} \sum_i R_i^2$$

and its rate of change is

$$\dot{E} = -\frac{1}{n} \sum_i \frac{B_i}{R_i},$$

while

$$\dot{E}^{\mathrm{LSW}} = -\frac{1}{n} \sum_i \frac{B_i^{\mathrm{LSW}}}{R_i},$$

with

$$B_i^{\mathrm{LSW}} = 1 - R_i u_\infty^{\mathrm{LSW}}, \qquad u_\infty^{\mathrm{LSW}} = \frac{\sum_i 1}{\sum_i R_i} := \frac{1}{\bar{R}}.$$

Since the energy is decreasing $\dot{E}$ is always negative. $\dot{E}^{\mathrm{LSW}}$ is also always negative, but we expect the difference in (2.14) to be positive for most realizations, since the LSW theory should underestimate the coarsening rate. Notice that we have normalized the energy by the particle number, which is more convenient for the subsequent analysis and of course irrelevant for the ratio (2.14). The reason for measuring the deviation in terms of (2.14) is that because of the gradient flow structure of the evolution, the quantity (2.14) can be expressed variationally (see (2.21)).

In Theorems 2.2 and 2.1 we will show that with high probability

$$\frac{\dot{E}^{\mathrm{LSW}} - \dot{E}}{|\langle \dot{E}^{\mathrm{LSW}} \rangle|} = \frac{\frac{1}{n}\sum_i \frac{B_i^{\mathrm{LSW}}}{R_i} - \frac{1}{n}\sum_i \frac{B_i}{R_i}}{|\langle \frac{1}{n}\sum_i \frac{B_i^{\mathrm{LSW}}}{R_i} \rangle|} \sim \left\{ \begin{matrix} n^{-1/3}\phi^{1/3} & \text{for} \quad n \ll \phi^{-1/2} \\ \phi^{1/2} & \text{for} \quad n \gg \phi^{-1/2} \end{matrix} \right\}. \tag{2.15}$$

Observe that this result is somewhat stronger than (2.12) and (2.13) in the sense that we make a (qualitative) statement about the entire distribution, not just its expected value.

2.4. Assumptions on the Particle Arrangement

In the following we consider a system of $n \gg 1$ particles in a sphere $B_{n^{1/3}}(0)$ of radius $n^{1/3}$ with volume fraction $\phi \ll 1$ centered at the origin. Therefore the typical interparticle distance is 1, the typical radius is $\phi^{1/3}$. We recall that the screening length in this setting scales as $\phi^{-1/6}$.

We take a fixed distribution of centers $\{X_i\}_i$ for which we assume the following regularity properties. There exists a constant $C_0 > 0$ such that

(H1)

$$\inf(\{d_{ij} \,|\, i \neq j\}) \geqslant \frac{1}{C_0}, \tag{2.16}$$

i.e. the particles have minimum distance of the order of the mean interparticle distance 1.

(H2) If $n \gg \phi^{-1/2}$, i.e. if the system size is larger than the screening length, the following holds: For all cubes $Q \subset B_{n^{1/3}}(0)$ of size $\phi^{-1/6}$ the number of particles in the cube, denoted by $n(Q)$, satisfies

$$n(Q) \geqslant \frac{1}{C_0}\phi^{-1/2}. \tag{2.17}$$

We remark that hypothesis (H1) ensures that the number of particles in a cube Q of size L is also bounded from above via

$$n(Q) \leqslant \frac{3}{4\pi} C_0^3 L^3. \tag{2.18}$$

A few comments on assumptions (H1) and (H2) are in order. In the following we will always consider a fixed distribution of particle centers such that (H1) and (H2) are valid. In ref. 10 it is shown that (H2) is satisfied with probability converging to one as $n \to \infty$ (at least if $\phi \leqslant 1/(\log n)^5$) if the particle centers are distributed independently. However, (H1) is not satisfied with probability close to one, but it is shown in ref. 10 that only few particles can violate (2.16). Thus, one would expect that the inclusion of those particles would only contribute through small error terms and would not change the results in this paper. However, a rigorous proof of this fact is beyond the scope of this paper.

Notice, however, that since the typical distance is increasing in time and the volume fraction is preserved, if (H1) and (H2) are valid for the initial data they remain to be valid during the Mullins–Sekerka evolution.

We denote in the following by $\{R_i\}_i$ the radii which are rescaled with respect to the typical particle radius $\phi^{1/3}$. We assume (without loss of generality with the same constant C_0 as before):

(H3) $\{R_i\}_i$ are positive random variables which are independent for $i \neq j$ and identically distributed with respect to a bounded probability density ν with $\|\nu\|_\infty \leqslant C_0$. We assume that ν has compact support, such that the supremum of the random variable is bounded, that means:

$$\sup(R_1) \leqslant C_0. \tag{2.19}$$

We denote in the following

$$\langle R^k \rangle := \int R_1^k \nu(R_1)\, dR_1, \quad \text{for } k = 1, 2, \ldots.$$

2.5. The Result

After rescaling the radii as described in Section 2.4 the monopole approximation reads

$$\frac{1}{R_i} = u_\infty + \frac{B_i}{R_i} + \phi^{1/3} \sum_{j \neq i} \frac{B_j}{d_{ij}}, \quad \sum_i B_i = 0, \tag{2.20}$$

with suitably rescaled u_∞. Our aim is to estimate the deviation from B_i^{LSW}. As described in Section 2.3 we consider the relative rate of change of energy (2.14). We know that with our sign convention $\dot{E}$ and $\dot{E}^{\mathrm{LSW}}$ are negative, and we anticipate that $\dot{E} - \dot{E}^{\mathrm{LSW}}$ is negative for most realizations since the LSW theory should underestimate the coarsening rate.

We now formulate our main results. With $\langle\cdot\rangle$ we will always denote the expected value w.r.t. the joint probability measure P of the variables $\{R_i\}_i$.

Theorem 2.1. *(The super–critical regime).* There are constants $N_0 = N_0(C_0) \gg 1$ and $\phi_0 = \phi(C_0) \ll 1$ such that the following holds. If $n \geqslant N_0\,\phi^{-1/2}$ and if $\phi \leqslant \phi_0$ we have with high probability that

$$-C\,\phi^{1/2} \leqslant \frac{\dot{E} - \dot{E}^{\mathrm{LSW}}}{|\langle \dot{E}^{\mathrm{LSW}}\rangle|} \leqslant -\frac{1}{C}\,\phi^{1/2}.$$

More precisely: For all $\varepsilon > 0$ there exists a constant $C = C(\varepsilon, C_0)$ such that

$$P\left(\left\{-C\,\phi^{1/2} \leqslant \frac{\dot{E} - \dot{E}^{\mathrm{LSW}}}{|\langle \dot{E}^{\mathrm{LSW}}\rangle|} \leqslant -\frac{1}{C}\,\phi^{1/2}\right\}^c\right) \leqslant \varepsilon.$$

The proof of Theorem 2.1 is the main contribution of this paper and is the content of Section 3.

Theorem 2.2. *(The sub–critical regime).* If $n \ll \phi^{-1/2}$ we have with high probability that

$$\frac{\dot{E} - \dot{E}^{\mathrm{LSW}}}{|\langle \dot{E}^{\mathrm{LSW}}\rangle|} \geqslant -C\,n^{-1/3}\,\phi^{1/3}.$$

More precisely: For all $\varepsilon > 0$ there exists a constant $C = C(\varepsilon, C_0)$ such that

$$P\left(\left\{-C\,n^{-1/3}\phi^{1/3} \leqslant \frac{\dot{E} - \dot{E}^{\mathrm{LSW}}}{|\langle \dot{E}^{\mathrm{LSW}}\rangle|}\right\}^c\right) \leqslant \varepsilon.$$

Furthermore

$$\frac{\langle \dot{E} - \dot{E}^{\mathrm{LSW}}\rangle}{|\langle \dot{E}^{\mathrm{LSW}}\rangle|} \leqslant -C\,n^{-1/3}\phi^{1/3}.$$

The proof of Theorem 2.2 will be the content of Section 4.

Remark. Unfortunately, we were not able to prove in the sub-critical regime the existence of a strong upper bound as in the super-critical case, but we can only control the expected value. Indeed, we have some numerical evidence, that for all $M>0$, $P(\{(\dot{E}-\dot{E}^{\mathrm{LSW}})/|\langle\dot{E}^{\mathrm{LSW}}\rangle| \leqslant M n^{-1/3}\phi^{1/3}\}^c)$ is bounded below by a positive constant for all n. In fact, in a periodic setting it is possible to prove this statement in a rigorous way. This will be shown in a forthcoming article.(11)

The proof of our result relies on the fact that the underlying evolution has the structure of a gradient flow and thus (2.14) has a variational formulation. To see that note that a solution of (2.20) can be characterized as a solution of

$$\min_{\{\tilde{B}_i\}_i;\sum_i \tilde{B}_i=0} \left\{ \frac{1}{n}\sum_i \frac{1}{2R_i}\tilde{B}_i^2 + \frac{\phi^{1/3}}{n}\sum_i\sum_{j\neq i}\frac{\tilde{B}_i\tilde{B}_j}{2d_{ij}} - \frac{1}{n}\sum_i \frac{\tilde{B}_i}{R_i} \right\}.$$

For the solution B_i we have

$$\frac{1}{n}\sum_i \frac{1}{2R_i}B_i^2 + \frac{\phi^{1/3}}{n}\sum_i\sum_{j\neq i}\frac{B_iB_j}{2d_{ij}} - \frac{1}{n}\sum_i \frac{B_i}{R_i} = -\frac{1}{n}\sum_i \frac{B_i}{2R_i} = \frac{1}{2}\dot{E}.$$

Therefore we can write the deviation in the rate of change of energy from the LSW result in the form:

$$\begin{aligned}\dot{E}-\dot{E}^{LSW} &= \min_{\{\tilde{B}_i\}_i,\sum_i \tilde{B}_i=0} \left\{ \frac{1}{n}\sum_i \frac{1}{R_i}\tilde{B}_i^2 \right.\\ &\quad \left. + \frac{\phi^{1/3}}{n}\sum_i\sum_{j\neq i}\frac{\tilde{B}_i\tilde{B}_j}{d_{ij}} - \frac{1}{n}\sum_i \frac{2\tilde{B}_i}{R_i} + \frac{1}{n}\sum \frac{B_i^{LSW}}{R_i} \right\},\end{aligned}$$

provided the quadratic form is positive semidefinite. This will be an implicit consequence of the lower bound which will be proved in Proposition 3.3. We recall that $B_i^{\mathrm{LSW}} = 1 - \frac{R_i}{\overline{R}}$, where $\overline{R} = \frac{1}{n}\sum_i R_i$, and use $\sum_i \tilde{B}_i = 0$ to find

$$\sum_i \frac{1}{R_i}\left(\tilde{B}_i - \left(1 - \frac{R_i}{\bar{R}}\right)\right)^2 = \sum_i \frac{1}{R_i}\tilde{B}_i^2 - \sum_i \frac{2}{R_i}\tilde{B}_i\left(1 - \frac{R_i}{\bar{R}}\right) + \sum_i \frac{1}{R_i}\left(1 - \frac{R_i}{\bar{R}}\right)^2$$
$$= \sum_i \frac{1}{R_i}\tilde{B}_i^2 - \sum_i \frac{2\tilde{B}_i^2}{R_i} + \sum_i \left(\frac{1}{R_i} - \frac{1}{\bar{R}}\right).$$

Thus we can express the deviation of the rate of change of energy in the compact form:

$$\dot{E} - \dot{E}^{\mathrm{LSW}} = \min_{\{\tilde{B}_i\}_i; \sum_i \tilde{B}_i} \left\{ \frac{1}{n}\sum_i \frac{1}{R_i}\left(\tilde{B}_i - B_i^{\mathrm{LSW}}\right)^2 + \frac{\phi^{1/3}}{n}\sum_i \sum_{j \neq i} \frac{\tilde{B}_i \tilde{B}_j}{d_{ij}} \right\}. \tag{2.21}$$

The variational formulation has the advantage that one can obtain an upper bound by finding a suitable trial field $\tilde{B}_i$. The construction of a proper trial field in the super-critical case (cf. Section 3.3) is guided by the intuition that due to the screening effect the system separates into independent subsystems of the size of the screening length. Indeed, the LSW construction in subsystems of the size of order $\phi^{-1/6}$ will do the job.

3. THE SUPER-CRITICAL REGIME

Our aim in this section is to prove Theorem 2.1, that is we are going to show that if $n\phi^{1/2}$ is sufficiently large, then we have (cf. (2.21)) with high probability

$$T := \min_{\{\tilde{B}_i\}_i; \sum_i \tilde{B}_i = 0} \left\{ \phi^{-1/2}\frac{1}{n}\sum_i \frac{1}{R_i}\left(\tilde{B}_i - B_i^{\mathrm{LSW}}\right)^2 + \phi^{-1/6}\frac{1}{n}\sum_i \sum_{j \neq i} \frac{\tilde{B}_i \tilde{B}_j}{d_{ij}} \right\} \stackrel{=}{\sim} -1. \tag{3.1}$$

Here and in the following we use for simplicity the notation

$$A \stackrel{<}{\sim} B, \; A \stackrel{>}{\sim} B \;\text{ or }\; A \stackrel{=}{\sim} B,$$

which means that there exists a positive constant C, such that

$$A \leqslant CB, \; A \geqslant CB \quad \text{or} \quad C^{-1}B \leqslant A \leqslant CB$$

respectively.

We notice, that (3.1) implies the statement of Theorem 2.1, since $|\langle \dot{E}^{\mathrm{LSW}} \rangle| \sim 1$.

3.1. Moments of B_i^{LSW} and Large Deviation Estimates

Here, we give some estimates for $\langle (1/\overline{R})^m \rangle$ for $m \in \{1, \ldots, 4\}$ and for moments of B_i^{LSW}.

In order to estimate $\langle (1/\overline{R})^m \rangle$ we first state some results from *large deviation analysis* (see for example ref. 12, Ch. 2). For the sequence of i.i.d. random variables $\{R_i\}_{i=1,\ldots,n}$:

$$P(\overline{R} \geqslant r) \leqslant e^{-n\Delta^*(r)} \qquad \text{for} \qquad r > \langle R \rangle,$$

and

$$P(\overline{R} \leqslant r) \leqslant e^{-n\Delta^*(r)} \qquad \text{for} \qquad r < \langle R \rangle.$$

Here, Δ^* is the Fenchel–Legendre transform of the cumulant generating function Δ; that means,

$$\Delta^*(r) := \sup_{\lambda \in \mathbb{R}} (\lambda r - \Delta(\lambda)), \;\; \Delta(\lambda) := \ln M(\lambda) := \ln \langle e^{\lambda R} \rangle.$$

It follows from the definition that $\Delta(\lambda)$ and $\Delta^*(r)$ are convex functions.

Lemma 3.1. Assume $\{R_i\}_i$ are i.i.d with bounded probability density ν. Then we have for any $m \in \mathbb{N}$

$$\left\langle \left(\frac{1}{\overline{R}} \right)^m \right\rangle \lesssim 1 \tag{3.2}$$

uniformly in n.

Proof. We estimate for $\lambda < 0$

$$M(\lambda) = \int_0^\infty e^{\lambda s} \nu(s)\, ds \leqslant C \int_0^\infty e^{\lambda s}\ \ ds = \frac{C}{|\lambda|}.$$

This implies

$$\begin{aligned} \Delta^*(r) &= \sup_\lambda (\lambda r - \Delta(\lambda)) \\ &\geqslant \sup_{\lambda<0} (\lambda r - \Delta(\lambda)) \\ &\geqslant \sup_{\lambda<0} \left(\lambda r - \ln\left(\frac{-C}{\lambda}\right)\right). \end{aligned}$$

The maximum of the last function is achieved for $\lambda = -1/r$. Therefore we have,

$$\Delta^*(r) \geqslant -1 - \ln(Cr),$$

which implies

$$e^{-n\Delta^*(r)} \leqslant C^n r^n e^n.$$

Hence we have for $r < \langle R \rangle$ that $P(\overline{R} \leqslant r) \leqslant C^n r^n$. Now we denote by $\bar{\nu}(s) = dP(\overline{R} \leqslant s)/ds$ the probability density of the average radius and choose an arbitrary number η such that $0 < \eta < \langle R \rangle$; then we have

$$\int \frac{1}{\overline{R}^m} dP = \int_0^\infty \frac{1}{s^m} \bar{\nu}(s)\, ds = \int_0^\eta \frac{1}{s^m} \bar{\nu}(s)\, ds + \int_\eta^\infty \frac{1}{s^m} \bar{\nu}(s)\, ds.$$

For the first integral on the right side of the last equation we write:

$$\begin{aligned} \int_0^\eta \frac{1}{s^m} \bar{\nu}(s)\, ds &= \int_0^\eta \frac{1}{s^m} \frac{d}{ds} P(\overline{R} \leqslant s)\, ds \\ &= \left[\frac{1}{s^m} P(\overline{R} \leqslant s)\right]_0^\eta + m \int_0^\eta \frac{1}{s^{m+1}} P(\overline{R} \leqslant s)\, ds. \end{aligned}$$

Now it holds for $n > m+1$ that

$$\begin{aligned} m \int_0^\eta \frac{1}{s^{m+1}} P(\overline{R} \leqslant s)\, ds &\leqslant m \int_0^\eta \frac{1}{s^{m+1}} e^n C^n s^n\, ds \\ &= m(Ce)^n \int_0^\eta s^{n-m-1}\, ds \end{aligned}$$

$$= \frac{m(Ce)^n}{n-m} \left[s^{n-m}\right]_0^\eta$$
$$= \frac{m(Ce)^m}{n-m} (\eta Ce)^{n-m}.$$

If one chooses

$$\eta := \frac{1}{2Ce}$$

it follows for sufficiently large n

$$m \int_0^\eta \frac{1}{s^{m+1}} P(\overline{R} \leqslant s)\, ds \leqslant \frac{m(Ce)^m}{n-m} \left(\frac{1}{2}\right)^{n-m} \leqslant \frac{1}{n}.$$

Further we estimate for sufficiently n large that

$$\left[\frac{1}{s^m} P(\overline{R} \leqslant s)\right]_0^\eta \leqslant \frac{1}{\eta^m} (eC\eta)^n = e^m C^m \left(\frac{1}{2}\right)^{n-m} \leqslant \frac{1}{n}$$

and

$$\int_\eta^\infty \frac{1}{s^m} \bar{\nu}(s) ds \leqslant \frac{1}{\eta^m} \int_0^\infty \bar{\nu}(s) ds = (2Ce)^m \lesssim 1\,.$$

This proves the lemma. ∎

Lemma 3.2. From the definition of the random variables $\{B_i^{\mathrm{LSW}}\}_i$ it follows that:

$$\langle B_i^{\mathrm{LSW}} B_j^{\mathrm{LSW}} \rangle = -\frac{1}{n-1} \langle (B_1^{\mathrm{LSW}})^2 \rangle \quad \text{for } i \neq j, \tag{3.3}$$

$$\langle (B_i^{\mathrm{LSW}})^2 B_j^{\mathrm{LSW}} B_k^{\mathrm{LSW}} \rangle = -\frac{1}{n-2} K_1 \quad \text{for} \quad i \neq j, \quad i \neq k, \quad j \neq k \tag{3.4}$$

and

$$\langle B_i^{\mathrm{LSW}} B_j^{\mathrm{LSW}} B_k^{\mathrm{LSW}} B_l^{\mathrm{LSW}} \rangle = \frac{3}{(n-3)(n-2)} K_1 \quad i,\ j,\ k,\ l \quad \text{all different,} \tag{3.5}$$

where

$$K_1 := \langle (B_1^{\text{LSW}})^2 (B_2^{\text{LSW}})^2 \rangle - \frac{1}{n-1} \langle (B_1^{\text{LSW}})^4 \rangle.$$

Further it holds that

$$\langle (B_1^{\text{LSW}})^2 \rangle, \ \langle (B_1^{\text{LSW}})^2 (B_2^{\text{LSW}})^2 \rangle, \quad \langle (B_1^{\text{LSW}})^4 \rangle \quad \text{and } |K_1| \lesssim 1.$$

Proof. Since the random variables $\{B_i^{\text{LSW}}\}_i$ are identically distributed we have by definition

$$\begin{aligned} \langle B_i^{\text{LSW}} B_j^{\text{LSW}} \rangle &= \langle B_1^{\text{LSW}} B_2^{\text{LSW}} \rangle, \\ \langle (B_i^{\text{LSW}})^2 B_j^{\text{LSW}} B_k^{\text{LSW}} \rangle &= \langle (B_1^{\text{LSW}})^2 B_2^{\text{LSW}} B_3^{\text{LSW}} \rangle, \\ \langle B_i^{\text{LSW}} B_j^{\text{LSW}} B_k^{\text{LSW}} B_l^{\text{LSW}} \rangle &= \langle B_1^{\text{LSW}} B_2^{\text{LSW}} B_3^{\text{LSW}} B_4^{\text{LSW}} \rangle. \end{aligned}$$

Further since $\sum_i B_i^{\text{LSW}} = 0$ we can write:

$$\begin{aligned} 0 &= \left\langle B_1^{\text{LSW}} \left(\sum_i B_i^{\text{LSW}} \right) \right\rangle = \langle (B_1^{\text{LSW}})^2 \rangle + \sum_{i \geqslant 2} \langle B_1^{\text{LSW}} B_i^{\text{LSW}} \rangle \\ &= \langle (B_1^{\text{LSW}})^2 \rangle + (n-1) \langle B_1^{\text{LSW}} B_2^{\text{LSW}} \rangle \end{aligned}$$

and therefore

$$\langle B_1^{\text{LSW}} B_2^{\text{LSW}} \rangle = -\frac{1}{n-1} \langle (B_1^{\text{LSW}})^2 \rangle$$

and similarly

$$\langle (B_1^{\text{LSW}})^3 B_2^{\text{LSW}} \rangle = -\frac{1}{n-1} \langle (B_1^{\text{LSW}})^4 \rangle. \tag{3.6}$$

In the same way the identities

$$\langle (B_1^{\text{LSW}})^2 B_2^{\text{LSW}} (\sum_i B_i^{\text{LSW}}) \rangle = 0$$

and

$$\langle B_1^{\text{LSW}} B_2^{\text{LSW}} B_3^{\text{LSW}} (\sum_i B_i^{\text{LSW}}) \rangle = 0$$

show that

$$\langle (B_1^{\mathrm{LSW}})^2 B_2^{\mathrm{LSW}} B_3^{\mathrm{LSW}}\rangle = -\frac{1}{n-2}\Big(\langle (B_1^{\mathrm{LSW}})^2 (B_2^{\mathrm{LSW}})^2\rangle + \langle (B_1^{\mathrm{LSW}})^3 B_2^{\mathrm{LSW}}\rangle\Big), \tag{3.7}$$

and

$$\langle B_1^{\mathrm{LSW}} B_2^{\mathrm{LSW}} B_3^{\mathrm{LSW}} B_4^{\mathrm{LSW}}\rangle = -\frac{3}{n-3}\langle (B_1^{\mathrm{LSW}})^2 B_2^{\mathrm{LSW}} B_3^{\mathrm{LSW}}\rangle. \tag{3.8}$$

Substituting (3.6) in (3.7) gives (3.4) and finally (3.4) in (3.8) implies (3.5).

In order to estimate the moments $\langle (B_1^{\mathrm{LSW}})^2\rangle$, $\langle (B_1^{\mathrm{LSW}})^2 (B_2^{\mathrm{LSW}})^2\rangle$ and $\langle (B_1^{\mathrm{LSW}})^4\rangle$, we write

$$|B_1^{\mathrm{LSW}}| \leqslant 1 + \frac{R_1}{\overline{R}} \leqslant 1 + \frac{C_0}{\overline{R}}.$$

Hence, it follows from the preceding lemma that

$$\begin{aligned} |\langle (B_1^{\mathrm{LSW}})^2\rangle| &\leqslant \langle \Big(1+\tfrac{C_0}{\overline{R}}\Big)^2\rangle \lesssim 1,\\ |\langle (B_1^{\mathrm{LSW}})^2 (B_2^{\mathrm{LSW}})^2\rangle| &\leqslant \langle \Big(1+\tfrac{C_0}{\overline{R}}\Big)^4\rangle \lesssim 1,\\ |\langle (B_1^{\mathrm{LSW}})^4\rangle| &\leqslant \langle \Big(1+\tfrac{C_0}{\overline{R}}\Big)^4\rangle \lesssim 1, \end{aligned}$$

which proves the last part of the lemma. ∎

3.2. Lower Bound

The aim of this section is to prove a lower bound on T as defined in (3.1). From the mathematical point of view this is the most interesting part of the proof of Theorem 2.1.

Proposition 3.3. Assume $n \geqslant \phi^{-1/2}$ and that $\phi \leqslant \phi_0$ where $\phi_0 = \phi_0(C_0)$ is sufficiently small. Then there exists for all $\varepsilon > 0$ a constant $C = C(\varepsilon, C_0)$ such that

$$P\,(T \leqslant -C) \leqslant \varepsilon.$$

With $\{B_i\}_i$ we will denote as before the minimizer which realizes T. It will be useful to approximate B_i^{LSW} by

$$L_i := 1 - \frac{R_i}{\langle R\rangle} \quad \text{for } \; i = 1, \ldots, n.$$

This has the advantage that the random variables L_i are independent. Then (3.1) can be written in the form,

$$\begin{aligned} T = & \phi^{-1/2}\frac{1}{n}\sum_i \frac{1}{R_i}(B_i - L_i)^2 + \phi^{-1/6}\frac{1}{n}\sum_i\sum_{j\neq i}\frac{B_i B_j}{d_{ij}} \\ & + \phi^{-1/2}\frac{1}{n}\sum_i \frac{1}{R_i}\left(L_i - B_i^{\mathrm{LSW}}\right)^2 \\ & + \phi^{-1/2}\frac{2}{n}\sum_i \frac{1}{R_i}(B_i - L_i)\left(L_i - B_i^{\mathrm{LSW}}\right). \end{aligned}$$

Since

$$-\frac{1}{4}(B_i - L_i)^2 - (L_i - B_i^{\mathrm{LSW}})^2 \leqslant (B_i - L_i)(L_i - B_i^{\mathrm{LSW}}),$$

we have

$$\begin{aligned} T \; \geqslant & \phi^{-1/2}\frac{1}{2n}\sum_i \frac{1}{R_i}(B_i - L_i)^2 + \phi^{-1/6}\frac{1}{n}\sum_i\sum_{j\neq i}\frac{B_i B_j}{d_{ij}} \\ & - \phi^{-1/2}\frac{1}{n}\sum_i \frac{1}{R_i}\left(L_i - B_i^{\mathrm{LSW}}\right)^2. \end{aligned} \tag{3.9}$$

At first we address the term in $(L_i - B_i^{\mathrm{LSW}})^2$. The following lemma proves that it is in most cases smaller than $O(1)$.

Lemma 3.4. It holds for all $M > 0$

$$\begin{aligned} P & \left(\phi^{-1/2}\frac{1}{n}\sum_i \frac{1}{R_i}\left(L_i - B_i^{LSW}\right)^2 \geqslant M\right) \\ & \leqslant \frac{1}{\sqrt{M}}\left(\frac{\mathrm{Var}[R]}{\langle R\rangle^2}\langle\frac{1}{R}\rangle\right)^{1/2}\left(\frac{1}{\phi^{1/2}n}\right)^{1/2} \lesssim \frac{1}{\sqrt{M}}. \end{aligned}$$

Note that $\langle\frac{1}{R}\rangle$ is bounded as we have seen in Lemma 3.1.

Proof. Since

$$\frac{1}{n}\sum_i \frac{1}{R_i}\left(L_i - B_i^{\mathrm{LSW}}\right)^2 = \overline{R}\left(\frac{1}{\langle R\rangle} - \frac{1}{\overline{\overline{R}}}\right)^2,$$

we estimate using first Chebyshev–Markov and then Cauchy–Schwarz inequality:

$$\begin{aligned} P\left(\phi^{-1/2}\overline{R}\left(\frac{1}{\langle R\rangle} - \frac{1}{\overline{\overline{R}}}\right)^2 \geqslant M\right) &\leqslant \frac{1}{\sqrt{M}}\langle\frac{|\langle R\rangle - \overline{R}|}{\sqrt{\overline{R}}\langle R\rangle}\rangle\frac{1}{\phi^{1/4}} \\ &\leqslant \frac{1}{\sqrt{M}}\left(\langle(\langle R\rangle - \overline{R})^2\rangle\left\langle\frac{1}{\overline{R}\langle R\rangle^2}\right\rangle\right)^{1/2}\frac{1}{\phi^{1/4}} \\ &\leqslant \frac{1}{\sqrt{M}}\left(\frac{\mathrm{Var}[R]}{\langle R\rangle^2}\left\langle\frac{1}{\overline{\overline{R}}}\right\rangle\right)^{1/2}\left(\frac{1}{\phi^{1/2}n}\right)^{1/2}, \end{aligned}$$

where we used that the random variables R_i are independent and identically distributed to obtain

$$\langle(\overline{R} - \langle R\rangle)^2\rangle = \frac{1}{n}\mathrm{Var}[R]. \quad \blacksquare \tag{3.10}$$

Defining

$$\Delta B_i := B_i - L_i,$$

we write the first term in (3.9) which is a local term in the form,

$$T_0 := \phi^{-1/2}\frac{1}{n}\sum_i \frac{1}{R_i}(\Delta B_i)^2.$$

Since this term is positive, we now address the second, nonlocal term and show that its modulus is in most cases smaller than $O(1) + \eta T_0$, where $\eta < 1$. In order to show this, we introduce a length ξ. Later ξ will be chosen to be smaller than the screening length $\phi^{-1/6}$. We use ξ to split the kernel,

$$\frac{1}{d_{ij}} = \frac{e^{-d_{ij}/\xi}}{d_{ij}} + \frac{1 - e^{-d_{ij}/\xi}}{d_{ij}}$$

and accordingly the second term of (3.9) splits as

$$\begin{aligned} T_1 &:= \phi^{-1/6}\frac{1}{n}\sum_i\sum_{j\neq i}\frac{B_iB_j}{d_{ij}} \\ &= \phi^{-1/6}\frac{1}{n}\sum_i\sum_{j\neq i}\frac{e^{-d_{ij}/\xi}}{d_{ij}}B_iB_j + \phi^{-1/6}\frac{1}{n}\sum_i\sum_{j\neq i}\frac{1-e^{-d_{ij}/\xi}}{d_{ij}}B_iB_j. \end{aligned}$$

This splitting is motivated by the method of Ewald summation (see e.g. ref. 13).

Remark. A lower bound for the term T_1 is given by a classical bound from the theory of charged particle systems (see refs. 14–16). In order to employ this bound, we enforce the constraint of hypothesis (H1) and define the hard core potential

$$\varphi_{ij}(d) := \begin{cases} B_iB_j/d & \text{if } d \geqslant C_0^{-1} \\ +\infty & \text{if } d < C_0^{-1}. \end{cases}$$

Therefore it is

$$T_1 = \phi^{-1/6}\frac{1}{n}\sum_i\sum_{j\neq i}\varphi_{ij}(d_{ij}).$$

Since $\varphi_{ij}(d)$ is bounded from below by the interaction energy of two spheres of radii $C_0^{-1}/2$ with constant surface charge density of $B_iC_0^2/\pi$, $B_jC_0^2/\pi$, respectively and with distance d of the centers, a lower bound for T_1 is given by the negative self energy of the spheres.[15] More precisely we have

$$T_1 \geqslant -\frac{1}{C_0}\phi^{-1/6}\left(\frac{1}{n}\sum_i B_i^2\right).$$

However, this bound is too coarse and we take in the following advantage of some cancellation effects between the "charges" B_i (see Lemmas 3.8, 3.9). Lemma 3.5 can be proved using the method used by Fisher and Ruelle,[15] but we give here a short proof for completeness.

In the following we denote

$$T_{11} := \phi^{-1/6}\frac{1}{n}\sum_i\sum_{j\neq i}\frac{e^{-d_{ij}/\xi}}{d_{ij}}B_iB_j$$

as the near-field component and

$$T_{12} := \phi^{-1/6} \frac{1}{n} \sum_i \sum_{j \neq i} \frac{1 - e^{-d_{ij}/\xi}}{d_{ij}} B_i B_j$$

as the far–field component of T_1. We first estimate the far–field component which turns out to be easy. Since $lim_{d\to 0}(1 - e^{-d/\xi})/d = 1/\xi$, we can write T_{12} as

$$\begin{aligned} T_{12} &= \phi^{-1/6} \frac{1}{n} \sum_i \sum_j \frac{1 - e^{-d_{ij}/\xi}}{d_{ij}} B_i B_j - \phi^{-1/6} \frac{1}{\xi} \frac{1}{n} \sum_i B_i^2 \\ &=: T_{121} - T_{122}. \end{aligned}$$

Lemma 3.5. It holds that $T_{121} \geqslant 0$.

Proof. We note that the Fourier transform of the kernel $f(y) = \frac{1 - e^{-|y|/\xi}}{|y|}$ is positive, since

$$\mathcal{F}\left[\frac{1 - e^{-|y|/\xi}}{|y|}\right](k) = \frac{1}{|k|^2} - \frac{1}{|k|^2 + \frac{1}{|\xi|^2}} \geqslant 0.$$

Since f is even, we have $\mathcal{F}(\mathcal{F}(f(y))) = f(-y) = f(y)$. Thus, f is the Fourier transform of a positive measure and therefore a function of positive type (this is the easy part of Bochner's theorem; see e.g. ref. 17, Theorem IX.9). ∎

Lemma 3.6. It holds that

$$0 \leqslant T_{122} \lesssim \phi^{-1/6} \frac{1}{\xi} (\phi^{1/2} T_0 + 1).$$

Proof. Using (2.19) we find

$$\begin{aligned} T_{122} &= \phi^{-1/6} \frac{1}{n\xi} \sum_i (\Delta B_i + L_i)^2 \lesssim \phi^{-1/6} \frac{1}{\xi} \left(\frac{1}{n} \sum_i \frac{1}{R_i} \Delta B_i^2 + 1 \right) \\ &\lesssim \phi^{-1/6} \frac{1}{\xi} (\phi^{1/2} T_0 + 1). \quad ∎ \end{aligned}$$

In order to proceed, the near field term T_{11} has to be rewritten in terms of ΔB_i and L_i,

$$\begin{aligned} T_{11} &= \phi^{-1/6}\frac{1}{n}\sum_i\sum_{j\neq i}\frac{e^{-d_{ij}/\xi}}{d_{ij}}\Delta B_i\Delta B_j + 2\phi^{-1/6}\frac{1}{n}\sum_i\sum_{j\neq i}\frac{e^{-d_{ij}/\xi}}{d_{ij}}L_j\Delta B_i \\ &\quad +\phi^{-1/6}\frac{1}{n}\sum_i\sum_{j\neq i}\frac{e^{-d_{ij}/\xi}}{d_{ij}}L_iL_j \\ &=: T_{111}+T_{112}+T_{113}. \end{aligned}$$

The term T_{111} can be estimated by the following convolution argument.

Lemma 3.7. We have,

$$|T_{111}| = \left|\phi^{-1/6}\frac{1}{n}\sum_i\sum_{j\neq i}\frac{e^{-d_{ij}/\xi}}{d_{ij}}\Delta B_i\Delta B_j\right| \lesssim \phi^{1/3}\xi^2 T_0.$$

Proof. First we use Cauchy–Schwarz inequality

$$\begin{aligned} |T_{111}| &= \left|\phi^{-1/6}\frac{1}{n}\sum_i\left[\sum_{j\neq i}\frac{e^{-d_{ij}/\xi}}{d_{ij}}\Delta B_j\right]\Delta B_i\right| \\ &\leqslant \phi^{-1/6}\frac{1}{n}\left(\left(\sum_i\left[\sum_{j\neq i}\frac{e^{-d_{ij}/\xi}}{d_{ij}}\Delta B_j\right]^2\right)\sum_i(\Delta B_i)^2\right)^{1/2}. \end{aligned}$$

It follows from Appendix 4.2, that the sums over spatial particle coordinates can be bounded by means of the corresponding Riemann integrals; for example we have:

$$\sum_{j\neq i}\frac{e^{-d_{ij}/\xi}}{d_{ij}} \lesssim \int_{B_{n^{1/3}}(0)}\frac{e^{-|x|/\xi}}{|x|}d^3x \leqslant \int_{\mathfrak{R}^3}\frac{e^{-|x|/\xi}}{|x|}d^3x \lesssim \xi^2.$$

This gives

$$\sum_i \left[\sum_{j\neq i} \frac{e^{-d_{ij}/\xi}}{d_{ij}} \Delta B_j\right]^2 \leqslant \sum_i \left[\sum_{j\neq i} \frac{e^{-d_{ij}/\xi}}{d_{ij}}\right]\left[\sum_{j\neq i} \frac{e^{-d_{ij}/\xi}}{d_{ij}} (\Delta B_j)^2\right]$$
$$\lesssim \xi^2 \sum_i \sum_{j\neq i} \frac{e^{-d_{ij}/\xi}}{d_{ij}} (\Delta B_j)^2$$
$$= \xi^2 \sum_j \left(\sum_{i\neq j} \frac{e^{-d_{ij}/\xi}}{d_{ij}}\right) (\Delta B_j)^2 = \xi^4 \sum_j (\Delta B_j)^2.$$

Hence we obtain

$$|T_{111}| \lesssim \phi^{-1/6} \xi^2 \frac{1}{n} \sum_j (\Delta B_j)^2 \lesssim \phi^{1/3} \xi^2 T_0. \quad \blacksquare$$

In the following we address the cross-terms T_{112} and T_{113}. Here we need a stochastic argument, that means the bounds we establish for these terms are valid except for a set with a small joint probability measure of the random variables $\{R_i\}_i$.

Lemma 3.8. We estimate

$$|T_{112}| \lesssim \phi^{1/12} \left(\frac{1}{n} \sum_i w_i\right)^{1/2} (T_0 + 1), \tag{3.11}$$

where

$$w_i = \left(\sum_{j\neq i} \frac{e^{-d_{ij}/\xi}}{d_{ij}} L_j\right)^2$$

and

$$P\left(\left|\frac{1}{n} \sum_i w_i\right| \geqslant M\xi\right) \lesssim \frac{1}{M}. \tag{3.12}$$

Proof. Inequality (3.11) follows immediately by Cauchy–Schwarz. Since we have for $i \neq j$,

$$\langle L_i L_j \rangle = \langle L_i \rangle \langle L_j \rangle = 0, \quad i \neq j, \quad \langle L_i^2 \rangle = O(1), \tag{3.13}$$

we obtain for the expectation value of $\{w_i\}_i$

$$\langle w_i \rangle = \sum_{j \neq i} \frac{e^{-2d_{ij}/\xi}}{d_{ij}^2} \langle L_1^2 \rangle \lesssim \int_{\Re^3} \left(\frac{e^{-|x|/\xi}}{|x|} \right)^2 d^3x \sim \xi.$$

Now we are in the position to prove the lemma. From the positivity of w_i it follows,

$$P\left(|\frac{1}{n} \sum_i w_i| \geqslant M\xi \right) \leqslant \frac{1}{M\xi} \left\langle |\frac{1}{n} \sum_i w_i| \right\rangle = \frac{1}{M\xi} \left\langle \frac{1}{n} \sum_i w_i \right\rangle \lesssim \frac{1}{M\xi} \frac{1}{n} n\xi = \frac{1}{M}.$$

∎

Finally we treat the term T_{113}.

Lemma 3.9. It holds that

$$P\left(|T_{113}| \geqslant \frac{M\xi^{1/2}}{\phi^{1/6} n^{1/2}} \right) \lesssim \frac{1}{M}.$$

Proof. We write

$$T_{113} = \phi^{-1/6} \frac{1}{n} \sum_i v_i,$$

where

$$v_i := \sum_{j \neq i} \frac{e^{-d_{ij}/\xi}}{d_{ij}} L_i L_j.$$

Using (3.13) we have $\langle v_i \rangle = 0$ and

$$\langle v_i^2 \rangle = \sum_{j \neq i} \sum_{k \neq i} \frac{e^{-d_{ij}/\xi}}{d_{ij}} \frac{e^{-d_{ik}/\xi}}{d_{ik}} \langle L_i^2 L_j L_k \rangle = \sum_{j \neq i} \left(\frac{e^{-d_{ij}/\xi}}{d_{ij}} \right)^2 \langle L_1^2 \rangle^2 \lesssim \xi.$$

Furthermore for $i \neq i'$

$$\begin{aligned}\langle v_i v_{i'} \rangle &= \sum_{j\neq i}\sum_{k\neq i'} \frac{e^{-d_{ij}/\xi}}{d_{ij}} \frac{e^{-d_{i'k}/\xi}}{d_{i'k}} \langle L_j L_{i'} \rangle \langle L_k L_i \rangle \\ &= \left(\frac{e^{-d_{ii'}/\xi}}{d_{ii'}} \right)^2 \langle L_1^2 \rangle^2 \lesssim \left(\frac{e^{-d_{ii'}/\xi}}{d_{ii'}} \right)^2\end{aligned}$$

and we observe

$$\left\langle \left(\frac{1}{n} \sum_i v_i \right)^2 \right\rangle \lesssim \frac{\xi}{n} + \frac{1}{n^2} \sum_i \sum_{j\neq i} \frac{e^{-2d_{ij}/\xi}}{d_{ij}^2} \lesssim \frac{\xi}{n} + \frac{1}{n^2} n\xi \sim \frac{\xi}{n}.$$

Therefore, it follows,

$$P\left(|T_{113}| \geqslant \frac{M\xi^{1/2}}{\phi^{1/6} n^{1/2}} \right) \leqslant \frac{n^{1/2}}{M\xi^{1/2}} \langle \left(\frac{1}{n} \sum_i v_i \right)^2 \rangle^{1/2} \lesssim \frac{n^{1/2}}{M\xi^{1/2}} \frac{\xi^{1/2}}{n^{1/2}} = \frac{1}{M}. \quad \blacksquare$$

Proof. *(of Proposition 3.3)*. For all realisations of the random variables $\{R_i\}_i$, except for a set with a small joint probability it follows from Lemmas 3.4, 3.5, 3.6, 3.7, 3.8, and 3.9 that

$$\begin{aligned} T &\geqslant \frac{1}{2} T_0 + T_{121} - |T_{122}| - |T_{111}| - |T_{112}| - |T_{113}| \\ &\quad - \phi^{-1/2} \frac{1}{n} \sum_i \frac{1}{R_i} (L_i - B_i^{\mathrm{LSW}})^2 \\ &\geqslant \frac{1}{2} T_0 - \frac{C}{2} \left[\phi^{-1/6} \frac{1}{\xi} (\phi^{1/2} T_0 + 1) + \phi^{1/3} \xi^2 T_0 + \phi^{1/12} \xi^{1/2} (T_0 + 1) \right. \\ &\qquad \left. + \frac{\xi^{1/2}}{\phi^{1/6} n^{1/2}} + 1 \right] \end{aligned}$$

for a generic universal constant C. We introduce the ratio of ξ to $\phi^{-1/6}$, which is the scaling of the screening length (see Part I):

$$\delta := \xi \phi^{1/6}$$

and obtain

$$T \geqslant \frac{1}{2}T_0\left(1-C\left[\frac{1}{\delta}\phi^{1/2}+\delta^2+\delta^{1/2}\right]\right)$$
$$-\frac{C}{2}\left(\frac{1}{\delta}+\delta^{1/2}+\delta^{1/2}\left(\frac{1}{\phi^{1/2}n}\right)^{1/2}+1\right).$$

Since $n\phi^{1/2}\geqslant 1$ we can first choose δ and then ϕ small enough to prove Proposition 3.3. ▮

3.3. Upper Bound

Proposition 3.10. There exists $N_0 = N_0(C_0) \gg 1$ such that if $n \geqslant N_0\phi^{-1/2}$ the following holds. There exists for all $\varepsilon > 0$ a constant $C = C(\varepsilon, C_0)$ such that

$$P\left(T \geqslant -\frac{1}{C}\right) \leqslant \varepsilon.$$

To compute the scaling of an upper bound in the super critical regime we cover the system $B_{n^{1/3}}(0)$ by disjoint cubes Q_α, $\alpha = 1,\ldots,m$ in the following way. In the interior of $B_{n^{1/3}}(0)$ we choose Q_α, in the following referred to as clusters or subclusters, to be cubes of side length $L\phi^{-1/6}$ whereas near the boundary we choose cubes of side length up to $2L\phi^{-1/6}$ with L satisfying $L \gg 1$. The constant N_0 will then be chosen such that $N_0 \gg L$. It is clear that we can in this way cover $B_{n^{1/3}}(0)$ and with (2.17) and (2.18) we have for the number of particles in Q_α, denoted by n_α, that

$$Cn' \geqslant n_\alpha \geqslant \frac{1}{C}n', \qquad \text{with} \qquad n' := \frac{L^3}{\phi^{1/2}}, \tag{3.14}$$

and by definition

$$\sum_{\alpha=1}^{m} n_\alpha = n.$$

Furthermore it holds for the number m of cubes that

$$m \sim \frac{n\phi^{1/2}}{L^3} \geqslant \frac{N_0}{L^3} \gg 1.$$

We introduce the cluster index $[i]$:

$$x_i \in Q_\alpha \text{ iff } [i]=\alpha.$$

Now, we define the trial field $B_i^S, i=1,\dots,n$,

$$B_i^S := 1-\frac{R_i}{\overline{R}_{[i]}}, \qquad \text{where} \qquad \overline{R}_{[i]} := \frac{1}{n_{[i]}}\sum_{j,[j]=[i]} R_j$$

which is obviously admissible and we obtain

$$\begin{aligned} T &\leqslant \phi^{-1/2}\frac{1}{n}\sum_i \frac{1}{R_i}\left(B_i^S - B_i^{\mathrm{LSW}}\right)^2 + \phi^{-1/6}\frac{1}{n}\sum_i\sum_{j\neq i}\frac{B_i^S B_j^S}{d_{ij}} \\ &=: F_1 + F_2. \end{aligned} \tag{3.15}$$

In the rest of this section we estimate the right-hand side, which is a straightforward but somewhat tedious computation.

The term F_1 can be written in a more appropriate form,

$$\begin{aligned} F_1 &:= \phi^{-1/2}\frac{1}{n}\sum_i \frac{1}{R_i}\left(B_i^S - B_i^{\mathrm{LSW}}\right)^2 \\ &= \phi^{-1/2}\frac{1}{n}\sum_i\left(\frac{R_i}{\overline{R}_{[i]}^2} - \frac{2R_i}{\overline{R}_{[i]}\overline{\overline{R}}} + \frac{R_i}{\overline{\overline{R}}^2}\right) \\ &= \phi^{-1/2}\frac{1}{n}\sum_{\alpha=1}^m \sum_{j,[j]=\alpha}\left(\frac{R_j}{\overline{R}_{[j]}^2} - \frac{2R_j}{\overline{R}_{[j]}\overline{\overline{R}}}\right) + \phi^{-1/2}\frac{1}{\overline{\overline{R}}} \\ &= \phi^{-1/2}\frac{1}{n}\sum_{\alpha=1}^m n_\alpha\left(\frac{1}{\overline{R}_\alpha} - \frac{1}{\overline{\overline{R}}}\right). \end{aligned}$$

Here we employed the notation

$$\overline{R}_\alpha := \frac{1}{n_\alpha}\sum_{j,[j]=\alpha} R_j. \tag{3.16}$$

First, we estimate the term F_1

Lemma 3.11. We have for all $M>0$

$$P\left(F_1 \geqslant \frac{1}{\phi^{1/2}n'}M\right) \lesssim \frac{1}{M^2}. \tag{3.17}$$

Proof. We start with the following identity

$$\begin{aligned}\frac{1}{n}\sum_{\alpha=1}^{m} n_\alpha\left(\frac{1}{\overline{R}_\alpha}-\frac{1}{\overline{\overline{R}}}\right) &= \frac{1}{n}\sum_{\alpha=1}^{m} n_\alpha\left(\left[\frac{1}{\overline{R}_\alpha}-\left(\frac{1}{\langle R\rangle}-\frac{1}{\langle R\rangle^2}(\overline{R}_\alpha-\langle R\rangle)\right)\right]\right.\\ &\quad\left.-\left[\frac{1}{\overline{\overline{R}}}-\left(\frac{1}{\langle R\rangle}-\frac{1}{\langle R\rangle^2}(\overline{R}-\langle R\rangle)\right)\right]\right)\\ &= \frac{1}{n}\sum_{\alpha=1}^{m} n_\alpha\left(\frac{(\langle R\rangle-\overline{R}_\alpha)^2}{\overline{R}_\alpha\langle R\rangle^2}-\frac{(\langle R\rangle-\overline{R})^2}{\overline{R}\langle R\rangle^2}\right).\end{aligned}$$

Hence, we have

$$\begin{aligned}\frac{1}{n}\sum_{\alpha=1}^{m} n_\alpha\left(\frac{1}{\overline{R}_\alpha}-\frac{1}{\overline{\overline{R}}}\right) &\leqslant \frac{1}{n}\sum_{\alpha=1}^{m} n_\alpha\left(\frac{(\langle R\rangle-\overline{R}_\alpha)^2}{\overline{R}_\alpha\langle R\rangle^2}\right)\\ &= \frac{1}{n}\sum_{\alpha=1}^{m} n_\alpha\left[\frac{(\overline{R}_\alpha-\langle R\rangle)^2}{\langle R\rangle^3}-\frac{(\overline{R}_\alpha-\langle R\rangle)^3}{\overline{R}_\alpha\langle R\rangle^3}\right]\\ &=: \frac{1}{n}\sum_{\alpha=1}^{m} n_\alpha\left(X_\alpha-Y_\alpha\right),\end{aligned}$$

where we introduce the i.i.d. random variables X_α and Y_α, $\alpha=1,\ldots,m$. In order to proceed, we calculate $\langle X_\alpha\rangle$, $\langle (X_\alpha)^2\rangle$ and $\langle (Y_\alpha)^2\rangle$. Since $\{R_i\}_i$ are i.i.d., we find

$$\begin{aligned}\langle X_\alpha\rangle &= \frac{1}{(n_\alpha)^2\langle R\rangle^3}\sum_{i,[i]=\alpha}\ \sum_{j,[j]=\alpha}\langle (R_i-\langle R\rangle)(R_j-\langle R\rangle)\rangle\\ &= \frac{1}{n_\alpha}\left(\frac{\langle (R-\langle R\rangle)^2\rangle}{\langle R\rangle^3}\right).\end{aligned} \tag{3.18}$$

Similarly, we compute

$$
\begin{aligned}
\langle (X_\alpha)^2 \rangle &= \frac{1}{(n_\alpha)^4 \langle R \rangle^6} \sum_{i,[i]=\alpha} \sum_{j,[j]=\alpha} \sum_{k,[k]=\alpha} \sum_{l,[l]=\alpha} \\
&\quad \langle (R_i - \langle R \rangle)(R_j - \langle R \rangle)(R_k - \langle R \rangle)(R_l - \langle R \rangle) \rangle \\
&= \frac{1}{\langle R \rangle^6} \left(\frac{1}{(n_\alpha)^3} \langle (R - \langle R \rangle)^4 \rangle + \frac{3n_\alpha (n_\alpha - 1)}{(n_\alpha)^4} \langle (R - \langle R \rangle)^2 \rangle^2 \right) \\
&\lesssim \frac{1}{(n_\alpha)^2}. \qquad (3.19)
\end{aligned}
$$

The estimate for the second moment of Y_α relies on the large deviation estimates. Following the last part of the proof to Lemma 3.1 we find, that there exists for n_α sufficient large, a generic constant ξ_0 such that

$$
\int_0^{\xi_0} \frac{1}{r^3} P(\overline{R}_\alpha \leqslant r)\, dr \lesssim \frac{1}{(n_\alpha)^3}
$$

and

$$
\frac{1}{\xi_0^2} P(\overline{R}_\alpha \leqslant \xi_0) + \lim_{\xi \to 0^+} \frac{1}{\xi^2} P(\overline{R}_\alpha \leqslant \xi) \lesssim \frac{1}{(n_\alpha)^3}.
$$

Since

$$
\begin{aligned}
&Y_\alpha^2 \lesssim \frac{1}{(\overline{R}_\alpha)^2} \quad \text{always}, \\
&Y_\alpha^2 \lesssim (\overline{R}_\alpha - \langle R \rangle)^6 \quad \text{provided} \quad \overline{R}_\alpha \geqslant \xi_0,
\end{aligned}
$$

we can estimate accordingly

$$
\begin{aligned}
\langle (Y_\alpha)^2 \rangle &= \int_0^{\xi_0} Y_\alpha^2 \frac{d}{dr} P(\overline{R}_\alpha \leqslant r)\, dr + \int_{\xi_0}^{\infty} Y_\alpha^2 \frac{d}{dr} P(\overline{R}_\alpha \leqslant r)\, dr \\
&\lesssim \int_0^{\xi_0} \frac{1}{r^2} \frac{d}{dr} P(\overline{R}_\alpha \leqslant r)\, dr + \langle (\overline{R}_\alpha - \langle R \rangle)^6 \rangle.
\end{aligned}
$$

Similarly to (3.19), we compute and estimate

$$
\langle (\overline{R}_\alpha - \langle R \rangle)^6 \rangle \lesssim \frac{1}{(n_\alpha)^3}.
$$

Further, we estimate for sufficient large n_α

$$\int_0^{\xi_0} \frac{1}{r^2}\frac{d}{dr}P(\overline{R}_\alpha \leqslant r)\,dr = \left[\frac{1}{r^2}P(\overline{R}_\alpha \leqslant r)\right]_0^{\xi_0} + 2\int_0^{\xi_0}\frac{1}{r^3}P(\overline{R}_\alpha \leqslant r)\,dr \lesssim \frac{1}{(n_\alpha)^3}.$$

Hence, it is

$$\langle (Y_\alpha)^2\rangle \lesssim \frac{1}{(n_\alpha)^3}. \tag{3.20}$$

Using these results we finally prove the lemma. It follows from (3.18), (3.19) and (3.14) that

$$\begin{aligned} P\left(\left|\frac{1}{n}\sum_\alpha n_\alpha X_\alpha\right| \geqslant \frac{1}{n'}M\right) &\leqslant \frac{(n')^2}{M^2}\left\langle\left(\frac{1}{n}\sum_\alpha n_\alpha X_\alpha\right)^2\right\rangle \\ &= \frac{(n')^2}{M^2n^2}\left(\sum_\alpha\sum_{\beta,\,\beta\neq\alpha} n_\alpha n_\beta\langle X_\alpha\rangle\langle X_\beta\rangle + \sum_\alpha n_\alpha^2\langle X_\alpha^2\rangle\right) \\ &\lesssim \frac{(n'm)^2}{M^2n^2} \lesssim \frac{1}{M^2}. \end{aligned}$$

The last part can be estimated in the same line using (3.20):

$$\begin{aligned} P\left(\left|\frac{1}{n}\sum_\alpha n_\alpha Y_\alpha\right| \geqslant \frac{1}{n'}M\right) &\leqslant \frac{(n')^2}{M^2}\left\langle\left(\frac{1}{n}\sum_\alpha n_\alpha Y_\alpha\right)^2\right\rangle, \\ &\leqslant \frac{(n')^2}{M^2}\frac{1}{n}\sum_\alpha n_\alpha^2\langle Y_\alpha^2\rangle \\ &\lesssim \frac{1}{M^2}\frac{n'm}{n} \lesssim \frac{1}{M^2}. \qquad \blacksquare \end{aligned}$$

To treat the second term F_2 we write

$$\begin{aligned} F_2 := &\left\langle \phi^{-1/6}\frac{1}{n}\sum_i\sum_{j\neq i,[j]=[i]}\frac{B_i^S B_j^S}{d_{ij}}\right\rangle \\ &+\phi^{-1/6}\frac{1}{n}\sum_i\sum_{j\neq i,[j]=[i]}\frac{1}{d_{ij}}\left(B_i^S B_j^S - \langle B_i^S B_j^S\rangle\right) \end{aligned}$$

$$+\phi^{-1/6}\frac{1}{n}\sum_i \sum_{j\neq i,[j]\neq[i]} \frac{B_i^S B_j^S}{d_{ij}}$$
$$= F_{21} + F_{22} + F_{23}.$$

Hence, the terms F_{21} and F_{22} include interactions within a single subcluster while F_{23} describes interactions between different subclusters.

We first remark that the statements of Lemma 3.2, that is (3.3), (3.4) and (3.5) are valid on every subcluster, with n_α instead of n. We denote

$$K_1^{[\alpha]} := \langle (B_1^{[\alpha]})^2 (B_2^{[\alpha]})^2 \rangle - \frac{1}{n_\alpha - 1}\langle (B_1^{[\alpha]})^4 \rangle,$$
$$K_2^{[\alpha]} := \langle (B_1^{[\alpha]})^2 (B_2^{[\alpha]})^2 \rangle,$$

where

$$B_1^{[\alpha]} = 1 - \frac{R_{i1}}{\overline{R}_\alpha}, \qquad B_2^{[\alpha]} = 1 - \frac{R_{i2}}{\overline{R}_\alpha} \qquad \text{and} \qquad B_3^{[\alpha]} = 1 - \frac{R_{i3}}{\overline{R}_\alpha},$$

where $i1$ is the number of one fixed particle in cluster Q_α and $i2$, $i3$ are numbers of particles in the cluster different from $i1$.

Lemma 3.12. It holds that

$$F_{21} \lesssim -\frac{1}{(\phi^{1/2}n')^{1/3}}.$$

Proof. Using the results of Lemma 3.2 we compute

$$F_{21} = \phi^{-1/6}\frac{1}{n}\sum_i \sum_{j\neq i,[j]=[i]} \frac{1}{d_{ij}}\langle B_i^S B_j^S \rangle$$
$$= -\phi^{-1/6}\frac{1}{n}\sum_i \frac{1}{n_{[i]}-1} \sum_{j\neq i,[j]=[i]} \frac{1}{d_{ij}}\langle (B_1^{[i]})^2 \rangle.$$

The factor $\langle (B_1^{[i]})^2 \rangle$ can be bounded from below

$$\langle (B_1^{[i]})^2 \rangle = \left\langle \left(1 - \frac{R_{i1}}{\overline{R}_{[i]}}\right)^2 \right\rangle = \left\langle \frac{(\overline{R}_{[i]} - R_{i1})^2}{\overline{R}_{[i]}^2} \right\rangle$$
$$\gtrsim \frac{1}{C_0^2}\left(1 - \frac{1}{n_{[i]}}\right)\left(\langle R^2 \rangle - \langle R \rangle^2\right) \gtrsim 1.$$

Now we use (3.14) and the fact that in each cube Q_α it holds $d_{ij} \leqslant 2(n')^{-1/3}$ which gives for all $i = 1, \dots, n$,

$$\sum_{j\neq i,[j]=[i]} \frac{1}{d_{ij}} \gtrsim n_{[i]}(n')^{-1/3} \gtrsim (n')^{2/3}.$$

This implies the statement of the lemma. ∎

Lemma 3.13. We have

$$P\left(|F_{22}| \geqslant \left(\frac{n'}{\phi}\right)^{1/6} \frac{1}{n^{1/2}} M\right) \lesssim \frac{1}{M^2}.$$

Proof. First we define the random variables

$$f_i := \sum_{j\neq i,[j]=[i]} \frac{1}{d_{ij}} B_i^S B_j^S, \quad i = 1, \dots, n$$

and calculate the variance, using (3.3) and (3.7)

$$\begin{aligned}
\langle (f_i - \langle f_i\rangle)^2\rangle &= \sum_{j\neq i,[j]=[i]} \sum_{k\neq i,[k]=[i]} \frac{1}{d_{ij}} \frac{1}{d_{ik}} \Big(\langle (B_i^S)^2 B_j^S B_k^S\rangle - \langle B_i^S B_j^S\rangle\langle B_i^S B_k^S\rangle\Big)\\
&= \sum_{j\neq i,[j]=[i]} \sum_{k\neq i,j,[k]=[i]} \frac{1}{d_{ij}} \frac{1}{d_{ik}} \left\langle (B_1^{[i]})^2 B_2^{[i]} B_3^{[i]}\right\rangle\\
&\quad + \sum_{j\neq i,[j]=[i]} \frac{1}{d_{ij}^2} \left\langle (B_1^{[i]})^2 (B_2^{[i]})^2\right\rangle\\
&\quad - \sum_{j\neq i,[j]=[i]} \sum_{k\neq i,[k]=[i]} \frac{1}{d_{ij}} \frac{1}{d_{ik}} \left\langle B_1^{[i]} B_2^{[i]}\right\rangle^2\\
&= -\frac{K_1^{[i]}}{n_{[i]}-2} \sum_{j\neq i,[j]=[i]} \sum_{k\neq i,j,[k]=[i]} \frac{1}{d_{ij}} \frac{1}{d_{ik}} + K_2^{[i]} \sum_{j\neq i,[j]=[i]} \frac{1}{d_{ij}^2}\\
&\quad - \frac{1}{(n_{[i]}-1)^2} \langle (B_1^{[i]})^2\rangle^2 \sum_{j\neq i,[j]=[i]} \sum_{k\neq i,[k]=[i]} \frac{1}{d_{ij}} \frac{1}{d_{ik}}.
\end{aligned}$$

In order to compute the covariance $\langle (f_i - \langle f_i\rangle)(f_j - \langle f_j\rangle)\rangle$, we discriminate between $[i] \neq [j]$ and $[i] = [j]$. Since the random variables f_i and f_j are independent for $[i] \neq [j]$ it holds in this case,

$$\langle (f_i - \langle f_i\rangle)(f_j - \langle f_j\rangle)\rangle = 0 \quad \text{for } [i] \neq [j].$$

For $[i]=[j]$ and $i\neq j$ we derive,

$$\begin{aligned}
\langle (f_i - \langle f_i\rangle)(f_j-\langle f_j\rangle)\rangle &= \langle f_i f_j\rangle - \langle f_i\rangle\langle f_j\rangle\\
&= K_2^{[i]}\frac{1}{d_{ij}^2} - \frac{K_1^{[i]}}{n_{[i]}-2}\sum_{k\neq i,j,[k]=[i]}\frac{1}{d_{ik}}\frac{1}{d_{jk}} - \frac{2K_1^{[i]}}{n_{[i]}-2}\frac{1}{d_{ij}}\sum_{k\neq i,j,[k]=[i]}\frac{1}{d_{ik}}\\
&\quad + \frac{3K_1^{[i]}}{(n_{[i]}-2)(n_{[i]}-3)}\sum_{k\neq i,j,[k]=[i]}\sum_{l\neq i,j,k,[l]=[i]}\frac{1}{d_{ik}}\frac{1}{d_{jl}}\\
&\quad - \frac{1}{(n_{[i]}-1)^2}\langle (B_1^{[i]})^2\rangle^2\sum_{k\neq i,[k]=[i]}\sum_{l\neq j,[l]=[i]}\frac{1}{d_{ik}}\frac{1}{d_{jl}}.
\end{aligned}$$

We remark, that the derivation is similar to the one in Lemma 4.4, where the steps are worked out in more detail. Now

$$\begin{aligned}
P\left(|F_{22}| \geqslant \left(\frac{n'}{\phi}\right)^{1/6}\frac{M}{n^{1/2}}\right) &\leqslant \frac{n\phi^{1/3}}{M^2 n'^{1/3} n^2\phi^{1/3}}\left\langle \sum_i\sum_j (f_i-\langle f_i\rangle)(f_j-\langle f_j\rangle)\right\rangle\\
&= \frac{1}{M^2 n n'^{1/3}}\left(\sum_i 2K_2^{[i]}\sum_{j\neq i,[j]=[i]}\frac{1}{d_{ij}^2}\right.\\
&\quad -\sum_i\sum_{j\neq i,[j]=[i]}\frac{4K_1^{[i]}}{n_{[i]}-2}\sum_{k\neq i,j,[k]=[i]}\frac{1}{d_{ij}}\frac{1}{d_{ik}}\\
&\quad +\sum_i\sum_{j\neq i,[j]=[i]}\frac{3K_1^{[i]}}{(n_{[i]}-2)(n_{[i]}-3)}\sum_{k\neq i,j,[k]=[i]}\sum_{l\neq i,j,k,[l]=[i]}\frac{1}{d_{ik}}\frac{1}{d_{jl}}\\
&\quad \left.-\sum_i\sum_{j\neq i,[j]=[i]}\langle (B_1^{[i]})^2\rangle^2\frac{1}{(n_{[i]}-1)^2}\sum_{k\neq i,[k]=[i]}\sum_{l\neq j,[l]=[i]}\frac{1}{d_{ik}}\frac{1}{d_{jl}}\right).
\end{aligned}$$

The first term on the left side of the last inequality can be estimated via

$$\left|\sum_i\sum_{j\neq i,[j]=[i]}\frac{1}{d_{ij}^2}\right| \lesssim n\int_{B_{Cn'^{1/3}}(0)}\frac{1}{|x|^2}d^3x \lesssim nn'^{1/3}$$

and similarly the other terms are bounded by a term of order $nn'^{1/3}$ using again (3.14). Together with (3.21) this proves the lemma. ∎

Finally, we treat the term F_{23}.

Lemma 3.14. It holds that

$$P\left(|F_{23}| \geqslant \frac{1}{(\phi^{1/2}n)^{1/3}} M\right) \lesssim \frac{1}{M^2}.$$

Proof. Similarly as before, we define the random variables

$$t_i := \sum_{j\neq i,[j]\neq[i]} \frac{1}{d_{ij}} B_i^S B_j^S, \quad i=1,\ldots,n,$$

which have zero expectation values. For the variance we derive

$$\begin{aligned}
\langle (t_i)^2\rangle &= \sum_{j\neq i,[j]\neq[i]} \sum_{k\neq i,[k]\neq[i]} \frac{1}{d_{ij}} \frac{1}{d_{ik}} \langle (B_i^S)^2 B_j^S B_k^S\rangle \\
&= \sum_{j\neq i,[j]\neq[i]} \frac{1}{d_{ij}^2} \langle (B_1^{[i]})^2\rangle \langle (B_1^{[j]})^2\rangle \\
&\quad - \sum_{j\neq i,[j]\neq[i]} \sum_{k\neq i,j,[k]=[j]} \frac{1}{(n_{[j]}-1)} \frac{1}{d_{ij}} \frac{1}{d_{ik}} \langle (B_1^{[i]})^2\rangle \langle (B_1^{[j]})^2\rangle.
\end{aligned}$$

The covariance $\langle t_i t_j\rangle$, $i\neq j$, follows in the case $[i]=[j]$ from

$$\begin{aligned}
\langle t_i t_j\rangle &= \sum_{k\neq i,[k]\neq[i]} \sum_{l\neq j,[l]\neq[i]} \frac{1}{d_{ik}} \frac{1}{d_{jl}} \langle B_i^S B_j^S B_k^S B_l^S\rangle \\
&= \sum_{k\neq i,[k]\neq[i]} \sum_{l\neq j,[l]=[k]} \frac{1}{d_{ik}} \frac{1}{d_{jl}} \langle B_i^S B_j^S\rangle \langle B_k^S B_l^S\rangle \\
&= -\frac{\langle (B_1^{[i]})^2\rangle}{(n_{[i]}-1)} \sum_{k\neq i,j,[k]\neq[i]} \frac{1}{d_{ik}} \frac{1}{d_{jk}} \langle (B_1^{[k]})^2\rangle \\
&\quad + \frac{\langle (B_1^{[i]})^2\rangle}{(n_{[i]}-1)(n_{[j]}-1)} \sum_{k\neq i,[k]\neq[i]} \sum_{l\neq j,k,[l]=[k]} \frac{1}{d_{ik}} \frac{1}{d_{jl}} \langle (B_1^{[k]})^2\rangle.
\end{aligned}$$

and in the case $[i]\neq[j]$ from

$$\begin{aligned}\langle t_i t_j\rangle &= \sum_{k\neq i,[k]=[j]}\ \sum_{l\neq j,[l]=[i]} \frac{1}{d_{ik}}\frac{1}{d_{jl}}\langle B_i^S B_l^S\rangle\langle B_j^S B_k^S\rangle\\
&= \left(\frac{1}{d_{ij}^2} - \frac{1}{(n_{[j]}-1)}\sum_{k\neq i,j,[k]=[j]}\frac{1}{d_{ik}}\frac{1}{d_{ji}} - \frac{1}{(n_{[i]}-1)}\sum_{l\neq i,j,[l]=[i]}\frac{1}{d_{ij}}\frac{1}{d_{jl}}\right.\\
&\quad \left.+\frac{1}{(n_{[i]}-1)(n_{[j]}-1)}\sum_{k\neq i,j,[k]=[j]}\ \sum_{l\neq i,j,[l]=[i]}\frac{1}{d_{ik}}\frac{1}{d_{jl}}\right)\langle (B_1^{[i]})^2\rangle\langle (B_1^{[j]})^2\rangle.\end{aligned}$$

Thus we have

$$\begin{aligned}\sum_i\sum_j\langle t_i t_j\rangle &= \sum_i\langle t_i^2\rangle + \sum_i\sum_{j\neq i,[j]=[i]}\langle t_i t_j\rangle + \sum_i\sum_{j\neq i,[j]\neq[i]}\langle t_i t_j\rangle\\
&\lesssim \left(2\sum_i\sum_{j\neq i,[j]\neq[i]}\frac{1}{d_{ij}^2} + \frac{1}{n'}\sum_i\sum_{j\neq i,[j]\neq[i]}\ \sum_{k\neq i,j,[k]=[j]}\frac{1}{d_{ik}}\frac{1}{d_{ij}}\right.\\
&\quad\left.+\frac{1}{(n')^2}\sum_i\sum_{j\neq i,[j]=[i]}\ \sum_{k\neq i,[k]\neq[i]}\ \sum_{l\neq j,k,[l]=[k]}\frac{1}{d_{ik}}\frac{1}{d_{jl}}\right). \qquad (3.21)\end{aligned}$$

The first term of (3.21) can be estimated as

$$\left|\sum_i\sum_{j\neq i,[j]\neq[i]}\frac{1}{d_{ij}^2}\right| \lesssim n^{4/3},$$

and similarly the other terms are bounded by a term of order $n^{4/3}$. Therefore we have

$$P\left(|F_{23}|\geqslant \frac{1}{(\phi^{1/2}n)^{1/3}}M\right)\leqslant \frac{(\phi^{1/2}n)^{2/3}}{M^2}\frac{1}{\phi^{1/3}n^2}\sum_i\sum_j\langle t_i t_j\rangle \lesssim \frac{1}{M^2}.$$

This finishes the proof of the lemma. ▮

Proof. *(of Prop. 3.10).* From Lemmas 3.11, 3.12, 3.13 and 3.14 we obtain that there exist constants C_1 and C_2 such that with high probability

$$\begin{aligned}T &\leqslant C_1\left(\frac{1}{\phi^{1/2}n'} + \left(\frac{n'}{\phi n^3}\right)^{1/6} + \frac{1}{(\phi^{1/2}n)^{1/3}}\right) - \frac{C_2}{(\phi^{1/2}n')^{1/3}}\\
&\leqslant C_1\left(\frac{1}{L^3} + \frac{L^{1/2}}{N_0^{1/2}} + \frac{1}{(N_0)^{1/3}}\right) - \frac{C_2}{L}.\end{aligned}$$

Now we choose N_0 sufficiently large, which depends on the constants C_1, C_2 which again depend only on C_0, and $1 \ll L \ll N_0^{1/3}$ to obtain $T \lesssim -1$ which finishes the proof of Proposition 3.10. ∎

4. THE SUB–CRITICAL REGIME

In this section we will prove Theorem 2.2, i.e. we are going to show that if $n\phi^{1/2}$ is sufficiently small, then

$$S := \min_{\{\tilde{B}_i\}_i; \sum_i \tilde{B}_i = 0} \left\{ \left(\frac{n}{\phi}\right)^{1/3} \frac{1}{n} \sum_i \frac{1}{R_i} \left(\tilde{B}_i - B_i^{\mathrm{LSW}}\right)^2 + \frac{1}{n^{2/3}} \sum_i \sum_{j \neq i} \frac{\tilde{B}_i \tilde{B}_j}{d_{ij}} \right\} \tag{4.1}$$

is of order one in n and ϕ.

4.1. Lower Bound

The aim of this section is to prove a lower bound for S as defined in (4.1).

Proposition 4.1. Assume $n \ll \phi^{-1/2}$. Then there exists for all $\varepsilon > 0$ a constant $C = C(\varepsilon, C_0)$ such that

$$P(S \leqslant -C) \leqslant \varepsilon.$$

Again we will denote by $\{B_i\}_i$ the minimizer and define

$$\Delta B_i := B_i - B_i^{\mathrm{LSW}}.$$

We will follow a similar strategy as in the super-critical regime and write

$$S = \left(\frac{n}{\phi}\right)^{1/3} \frac{1}{n} \sum_i \frac{1}{R_i} \left(B_i - B_i^{\mathrm{LSW}}\right)^2 + \frac{1}{n^{2/3}} \sum_i \sum_{j \neq i} \frac{B_i B_j}{d_{ij}} =: S_0 + S_1,$$

where

$$
\begin{aligned}
S_1 &= \frac{1}{n^{2/3}} \sum_i \sum_{j \neq i} \frac{1}{d_{ij}} (\Delta B_i + B_i^{\mathrm{LSW}})(\Delta B_j + B_j^{\mathrm{LSW}}) \\
&= \frac{1}{n^{2/3}} \sum_i \sum_{j \neq i} \frac{1}{d_{ij}} \Delta B_i \Delta B_j + \frac{2}{n^{2/3}} \sum_i \sum_{j \neq i} \frac{\Delta B_j B_i^{\mathrm{LSW}}}{d_{ij}} \\
&\quad + \frac{1}{n^{2/3}} \sum_i \sum_{j \neq i} \frac{1}{d_{ij}} B_i^{\mathrm{LSW}} B_j^{\mathrm{LSW}} \\
&=: S_{11} + S_{12} + S_{13}.
\end{aligned}
$$

Lemma 4.2. We have that

$$
|S_{11}| \lesssim \left(n\phi^{1/2}\right)^{2/3} S_0.
$$

Proof. Using Cauchy–Schwarz inequality we get,

$$
S_{11} \leqslant \frac{1}{n^{2/3}} \left(\sum_i \left[\sum_{j \neq i} \frac{1}{d_{ij}} \Delta B_j \right]^2 \right)^{1/2} \left(\sum_i [\Delta B_i]^2 \right)^{1/2}.
$$

Further, it follows similarly

$$
\begin{aligned}
\sum_i \left(\sum_{j \neq i} \frac{1}{d_{ij}} \Delta B_j \right)^2 &\leqslant \sum_i \left(\sum_{j \neq i} \frac{1}{d_{ij}} \right) \left(\sum_{j \neq i} \frac{1}{d_{ij}} (\Delta B_j)^2 \right) \\
&\lesssim \left(\int_{B_{n^{1/3}}(0)} \frac{1}{|x|} d^3x \right) \left(\sum_i \sum_{j \neq i} \frac{1}{d_{ij}} (\Delta B_j)^2 \right) \\
&\lesssim n^{2/3} \sum_i \sum_{j \neq i} \frac{1}{d_{ij}} (\Delta B_j)^2 \\
&= n^{2/3} \sum_j \left(\sum_{i \neq j} \frac{1}{d_{ij}} \right) (\Delta B_j)^2 \\
&\lesssim n^{4/3} \sum_j (\Delta B_j)^2.
\end{aligned}
$$

This implies,

$$
\begin{aligned}
|S_{11}| &\lesssim \frac{1}{n^{2/3}} \left(n^{4/3} \sum_i (\Delta B_i)^2 \right)^{1/2} \left(\sum_i (\Delta B_i)^2 \right)^{1/2} \\
&\leqslant \sum_i (\Delta B_i)^2 \\
&\leqslant \left(n\phi^{1/2} \right)^{2/3} \sup_i (R_i) S_0 \\
&\sim \left(n\phi^{1/2} \right)^{2/3} S_0. \qquad (4.2)
\end{aligned}
$$

Hence the term $|S_{11}|$ is much smaller than S_0 since in the sub-critical regime we assume $n\phi^{1/2} \ll 1$. ▮

Lemma 4.3. We have

$$
|S_{12}| \lesssim n^{1/6} \phi^{1/6} \Big(\frac{1}{n} \sum_i w_i \Big)^{1/2} (S_0 + 1) \qquad (4.3)
$$

with

$$
w_i := \Big(\sum_{j \neq i} \frac{B_j^{\mathrm{LSW}}}{d_{ij}} \Big)^2
$$

and

$$
P \left(|\frac{1}{n} \sum_i w_i| \geqslant M n^{1/3} \right) \lesssim \frac{1}{M}. \qquad (4.4)
$$

Proof. To prove the lemma we first apply Cauchy–Schwarz inequality,

$$
\begin{aligned}
|S_{12}| &= \frac{2}{n^{2/3}} \left| \sum_i \Delta B_i \sum_{j \neq i} \frac{B_j^{\mathrm{LSW}}}{d_{ij}} \right| \\
&\leqslant \frac{2}{n^{2/3}} \left(\sum_i (\Delta B_i)^2 \right)^{1/2} \left(\sum_i \left[\sum_{j \neq i} \frac{B_j^{\mathrm{LSW}}}{d_{ij}} \right]^2 \right)^{1/2}
\end{aligned}
$$

$$\leqslant \sum_i (\Delta B_i)^2 + \frac{1}{n^{4/3}} \sum_i \left(\sum_{j \neq i} \frac{B_j^{\mathrm{LSW}}}{d_{ij}} \right)^2$$
$$\lesssim \left(n\phi^{1/2} \right)^{1/3} S_0 + \sum_i \eta_i .$$

The expectation values of the random variables $\{\eta_i\}_i$ are

$$\langle \eta_i \rangle = \frac{\langle (B_1^{\mathrm{LSW}})^2 \rangle}{n^{4/3}} \sum_{j \neq i} \frac{1}{d_{ij}^2} - \frac{\langle (B_1^{\mathrm{LSW}})^2 \rangle}{n^{4/3}(n-1)} \sum_{j \neq i} \sum_{k \neq i,j} \frac{1}{d_{ij} d_{ik}} .$$

Therefore we estimate

$$P\left(|\sum_i \eta_i| \geqslant M \right) \leqslant \frac{1}{M} \sum_i \langle \eta_i \rangle$$
$$= \frac{\langle (B_1^{\mathrm{LSW}})^2 \rangle}{M n^{4/3}} \sum_i \left(\sum_{j \neq i} \frac{1}{d_{ij}^2} - \frac{1}{(n-1)} \sum_{j \neq i} \sum_{k \neq i,j} \frac{1}{d_{ij} d_{ik}} \right)$$
$$\lesssim \frac{1}{M} . \quad \blacksquare$$

Lemma 4.4. We estimate

$$P\left(|S_{13}| \geqslant M \right) \lesssim \frac{1}{M^2} . \tag{4.5}$$

Proof. We proceed as in the corresponding Lemma 3.9. Since here $\{B_i^{\mathrm{LSW}}\}_i$ are not independent, the computations are slightly more involved. We introduce

$$v_i := \sum_{j \neq i} \frac{1}{d_{ij}} B_i^{\mathrm{LSW}} B_j^{\mathrm{LSW}} ,$$

and compute expectation values, variances and covariances of the random variables $\{v_i\}_i$ using the results from Lemma 3.2:

$$\langle v_i \rangle = \sum_{j \neq i} \frac{1}{d_{ij}} \langle B_i^{\mathrm{LSW}} B_j^{\mathrm{LSW}} \rangle = -\frac{1}{n-1} \langle (B_1^{\mathrm{LSW}})^2 \rangle \sum_{j \neq i} \frac{1}{d_{ij}} , \tag{4.6}$$

$$
\begin{aligned}
\langle v_i^2\rangle &= \sum_{j\neq i}\sum_{k\neq i}\frac{1}{d_{ij}}\frac{1}{d_{ik}}\langle (B_i^{\mathrm{LSW}})^2 B_j^{\mathrm{LSW}} B_k^{\mathrm{LSW}}\rangle \\
&= \langle (B_1^{\mathrm{LSW}})^2 (B_2^{\mathrm{LSW}})^2\rangle \sum_{j\neq i}\frac{1}{d_{ij}^2} + \langle (B_1^{\mathrm{LSW}})^2 B_2^{\mathrm{LSW}} B_3^{\mathrm{LSW}}\rangle \sum_{j\neq i}\sum_{k\neq i,j}\frac{1}{d_{ij}}\frac{1}{d_{ik}} \\
&= K_2\sum_{j\neq i}\frac{1}{d_{ij}^2} - \frac{K_1}{n-2}\sum_{j\neq i}\sum_{k\neq i,j}\frac{1}{d_{ij}}\frac{1}{d_{ik}},
\end{aligned}
$$

with K_1 as in Lemma 3.2 and $K_2 := \langle (B_1^{\mathrm{LSW}})^2 (B_2^{\mathrm{LSW}})^2\rangle$. For $i\neq j$ we find after some computation

$$
\begin{aligned}
\langle v_i v_j\rangle = K_2\left(\frac{1}{d_{ij}}\right)^2 - \frac{K_1}{n-2}\sum_{k\neq i,j}\frac{1}{d_{ik}}\frac{1}{d_{jk}} \\
-\frac{2K_1}{(n-2)}\frac{1}{d_{ij}}\sum_{k\neq i,j}\frac{1}{d_{ik}} + \frac{3K_1}{(n-3)(n-2)}\sum_{k\neq i,j}\sum_{l\neq i,j,k}\frac{1}{d_{ik}}\frac{1}{d_{jl}}
\end{aligned}
$$

and obtain

$$
\begin{aligned}
P\left(|S_{13}|\geqslant \frac{1}{M}\right) &\leqslant \frac{n^{2/3}}{M^2}\left\langle\left(\frac{1}{n}\sum_i v_i\right)^2\right\rangle \\
&= \frac{n^{2/3}}{M^2}\left(\frac{1}{n^2}\sum_i\langle v_i^2\rangle + \frac{1}{n^2}\sum_i\sum_{j\neq i}\langle v_i v_j\rangle\right) \\
&= \frac{1}{M^2 n^{4/3}}\left(2K_2\sum_i\sum_{j\neq i}\frac{1}{d_{ij}^2} - \frac{4K_1}{n-2}\sum_i\sum_{j\neq i}\sum_{k\neq i,j}\frac{1}{d_{ij}}\frac{1}{d_{ik}}\right. \\
&\quad \left. + \frac{3K_1}{(n-3)(n-2)}\sum_i\sum_{j\neq i}\sum_{k\neq i,j}\sum_{l\neq i,j,k}\frac{1}{d_{ik}}\frac{1}{d_{jl}}\right).
\end{aligned}
$$

It is easily seen that each term in the brackets is of order $n^{4/3}$ which finishes the proof. ∎

Since in the sub-critical regime we have $n \ll \phi^{-1/2}$ Lemmas 4.2, 4.3 and 4.3 finish the proof of Proposition 4.1.

4.2. Upper Bound

Proposition 4.5. It holds that

$$\langle S \rangle \lesssim -1. \tag{4.7}$$

Proof. We take as a trial field in the variational principle $\{B_i^{\mathrm{LSW}}\}_i$. Thus

$$S \leqslant \frac{1}{n^{2/3}} \sum_i \sum_{j \neq i} \frac{B_i^{\mathrm{LSW}} B_j^{\mathrm{LSW}}}{d_{ij}} = \frac{1}{n^{2/3}} \sum_i v_i,$$

where v_i is as in Lemma 4.4. The assertion of the lemma follows immediately using (4.6). ∎

APPENDIX A: UPPER BOUNDS FOR SOME SPATIAL SUMS

In this appendix we bound some sums, as e.g.

$$\sum_{j=1,\, j\neq i}^{n} \frac{1}{d_{ij}}, \quad \sum_{j=1,\, j\neq i}^{n} \frac{1}{d_{ij}^2} \quad \text{or} \quad \sum_{j=1,\, j\neq i}^{n} \frac{e^{-d_{ij}/\hat{\xi}}}{d_{ij}},$$

by integral expressions. The general structure of these sums is

$$\sum_{j=1,\, j\neq i}^{n} f(d_{ij})$$

where the function

$$f : d \longrightarrow f(d)$$

is a monotone decreasing, smooth function of the particle distance d. From assumption (2.16) we conclude, that there exists a radius

$$d_{\min} := \frac{1}{2} \inf\{d_{ij} \,|\, i \neq j\} = O(1)$$

such that all spheres $\{B_{d_{\min}}(X_i)\}_i$ are disjoint. Defining the points

$$X_j^{(i)} = (X_j - X_i) - \frac{d_{\min}}{2d_{ji}}(X_j - X_i) \in B_{d_{\min}}(X_j) \quad \text{for all } j \neq i,$$

we have

$$B_{\frac{d_{\min}}{2}}(X_j^{(i)}) \subset B_{d_{\min}}(X_j) \quad \text{for all } j \neq i$$

and

$$|x - X_i| \leqslant d_{ij} \quad \text{for all } x \in B_{\frac{d_{\min}}{2}}(X_j^{(i)}).$$

Therefore we estimate

$$\begin{aligned}
\sum_{j=1,\, j\neq i}^{n} f(d_{ij}) &= \sum_{j=1,\, j\neq i}^{n} \frac{1}{\frac{4\pi}{3}\left(\frac{d_{\min}}{2}\right)^3} \int_{B_{\frac{d_{\min}}{2}}(X_j^{(i)})} f(d_{ij}) d^3 y \\
&\leqslant \sum_{j=1,\, j\neq i}^{n} \frac{1}{\frac{4\pi}{3}\left(\frac{d_{\min}}{2}\right)^3} \int_{B_{\frac{d_{\min}}{2}}(X_j^{(i)})} f(|y - X_i|) d^3 y \\
&\leqslant \frac{1}{\frac{4\pi}{3}\left(\frac{d_{\min}}{2}\right)^3} \int_{B_{2n^{1/3}}(0)} f(|y|) d^3 y \\
&\lesssim \int_{B_{n^{1/3}}(0)} f(|y|) d^3 y.
\end{aligned}$$

This establishes an upper bound in form of an integral expression.

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