

FRÉCHET DIFFERENTIABILITY OF MOLECULAR DISTRIBUTION FUNCTIONS I. L^∞ ANALYSIS

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Abstract. For a grand canonical ensemble of classical point-like particles at equilibrium in continuous space we investigate the functional relationship between a stable and regular pair potential describing the interaction of the particles and the corresponding molecular distribution functions. For certain admissible perturbations of the pair potential and sufficiently small activity we rigorously establish Frechet differentiability with respect to the supremum norm in the image space – both for bounded domains and in the thermodynamical limit.

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1. Introduction. We consider the grand canonical description of a continuous system of identical classical particles in thermodynamical equilibrium; cf., e.g., Hansen and McDonald [3]. It is assumed that the potential energy of the system is determined by a pair potential which only depends on the distance of the interacting particles. Under certain additional assumptions on the potential (known as *stability* and *regularity*) it has been rigorously proved in the 60's of the previous century (see the monograph by Ruelle [9]) that the corresponding molecular distribution functions of the particles have a well-defined thermodynamical limit for a small enough activity coefficient. For example, the limiting singlet distribution function determines the (constant) number density of the particles; the corresponding pair distribution function which provides the probability density of observing two particles in prescribed coordinates at the same time only depends on the distance between the given coordinates and gives rise to a so-called *radial distribution function*. The *inverse problem* whether a given radial distribution function can be obtained as the thermodynamical limit of an equilibrium distribution for a certain pair potential is an open problem; there are only few partial results, for example by Henderson [4] on uniqueness and by Koralov [5] on existence of corresponding solutions.

This inverse problem is fundamental for the development of efficient multiscale algorithms for the numerical simulation of complex soft matter phenomena, compare, for example, Rühle et al [7]. Many of these algorithms employ methods for coarse-graining complex molecules and need to derive effective potentials for the coarse-grained *beads* from measured data such as the radial distribution function.

One of the algorithms for solving this inverse problem is the so-called *Inverse Monte-Carlo* method by Lyubartsev and Laaksonen [6], which utilizes the well-known Newton method for the numerical solution of nonlinear equations. As such, this method requires the derivative of the radial distribution function with respect to the pair potential.

While the analyticity of the molecular distribution functions with respect to the inverse temperature is classical (cf., e.g., Ruelle [8]), a rigorous analysis of their differentiability with respect to the potential appears to be lacking. This is the purpose

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of this present work, which is the first part of two consecutive papers. A crucial ingredient is the choice of a proper (and natural) topology for suitable perturbations of a given potential (see Proposition 2.1 below). The topology that we suggest allows to exploit the well-known *Kirkwood-Salsburg system* of equations and the corresponding theory in [9] to rigorously determine the derivative of all molecular distribution functions (and their thermodynamical limit) with respect to the underlying potential in the L^∞ norm of the corresponding distribution functions. We refer to Section 2 for a precise statement of our results.

As far as the radial distribution function is concerned it is also natural to investigate the differentiability of the so-called Ursell function (sometimes also referred to as *pair correlation function* [3]). In the high temperature regime the thermodynamical limit of the Ursell function is known to belong to $L^1(\mathbb{R}^3)$ as a function of the distance between its two input particle positions, cf. [9] again. We can prove that the differentiability of the Ursell function extends to this topology, either. Since the proof requires a completely different set of tools we postpone this and related results to the follow-up paper [2].

The outline of this first part is as follows. In the following section the setting and basic assumptions of this work will be specified. There we also formulate the two main results, Theorem 1 on the differentiability of the molecular distribution functions, and Theorem 2 on the thermodynamical limit of these derivatives. Section 3 is devoted to a proof of Theorem 1 and Section 4 provides the proof of Theorem 2. An explicit formula for the derivative of the pair distribution function and the singlet distribution function in a bounded domain is presented and discussed in Section 5.

2. Problem setting. In the sequel we present our basic assumptions on the system under consideration and review some basic facts; most of them are well known, and unless stated otherwise, we refer to [9] as a standard reference.

We consider a grand canonical ensemble of identical classical point-like particles in a box $\Lambda \subset \mathbb{R}^3$ in thermodynamical equilibrium with defined positive inverse temperature β and activity z . Throughout this work we assume that the box Λ is a cube centered at the origin. The interaction of the particles is given by a pair potential $u : \mathbb{R}^+ \rightarrow \mathbb{R}$, which only depends on the distance between the corresponding particles. Such a pair potential is called *stable*, if there exists a constant $B > 0$ such that

$$\sum_{1 \leq i < j \leq N} u(|R_i - R_j|) \geq -BN$$

for all configurations of N labeled particles (and all $N \in \mathbb{N}$), where $\mathbf{R}_N = (R_1, \dots, R_N) \in (\mathbb{R}^3)^N$ is the N -tupel with the particle coordinates. A stable pair potential u is called *regular*, if the associated *Mayer function*

$$f(R) = e^{-\beta u(|R|)} - 1 \tag{2.1}$$

belongs to $L^1(\mathbb{R}^3)$, i.e., if

$$\int_0^\infty |e^{-\beta u(r)} - 1| r^2 dr < \infty.$$

In order to investigate differentiability with respect to u we need to allow some variability of the potential without disturbing the above two properties. Therefore we will stipulate a slightly more restrictive but much more handy assumption on u .

ASSUMPTION A. *There exists $s > 0$ and positive decreasing functions u_*, u^* :*

$\mathbb{R}^+ \rightarrow \mathbb{R}$ with

$$\int_0^s u_*(r) r^2 dr = \infty \quad \text{and} \quad \int_s^\infty u^*(r) r^2 dr < \infty,$$

such that u satisfies

$$u(r) \geq u_*(r), \quad r \leq s, \quad \text{and} \quad |u(r)| \leq u^*(r), \quad r \geq s.$$

An example of a pair potential that satisfies Assumption A is the familiar *Lennard-Jones potential*

$$u_{\text{LJ}}(r) = 4\varepsilon \left(\left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^6 \right)$$

with parameters $\varepsilon, \sigma > 0$.

Given that the pair potential u satisfies Assumption A we introduce the space \mathcal{V}_u of *perturbations* $v : \mathbb{R}^+ \rightarrow \mathbb{R}$ for which $|v|/u$ is bounded in $(0, s)$ and $|v|/u^*$ is bounded in (s, ∞) ; \mathcal{V}_u is a Banach space when equipped with the norm

$$\|v\|_{\mathcal{V}_u} = \max\{ \|v/u\|_{(0,s)}, \|v/u^*\|_{(s,\infty)} \}. \quad (2.2)$$

Here, and throughout, the notation $\|\cdot\|_\Omega$ refers to the supremum norm of a function acting from some interval $\Omega \subset \mathbb{R}^d$ into a given Banach space. As we see next, the topology of \mathcal{V}_u defines an open neighborhood of stable and regular pair potentials around the given u .

PROPOSITION 2.1. *Let u satisfy Assumption A, \mathcal{V}_u be the Banach space with norm (2.2), and $0 < t_0 < 1$ be given. Then there are constants $B \geq 0$ and $c_\beta > 0$ such that the potential $\tilde{u} = u + v$ satisfies*

$$\sum_{1 \leq i < j \leq N} \tilde{u}(|R_i - R_j|) \geq -BN \quad (2.3)$$

for every $\mathbf{R}_N \in (\mathbb{R}^3)^N$ and

$$4\pi \int_0^\infty |e^{-\beta \tilde{u}(r)} - 1| r^2 dr \leq c_\beta \quad (2.4)$$

for every $v \in \mathcal{V}_u$ with $\|v\|_{\mathcal{V}_u} \leq t_0$, i.e., \tilde{u} is stable and regular. Moreover, for all $N \geq 2$ and all $\mathbf{R}_N \in (\mathbb{R}^3)^N$ there exists $j^* = j^*(\mathbf{R}_N)$ such that

$$\sum_{\substack{i=1 \\ i \neq j^*}}^N \tilde{u}(|R_i - R_{j^*}|) \geq -2B \quad (2.5)$$

for every $\tilde{u} = u + v$ with $\|v\|_{\mathcal{V}_u} \leq t_0$; in particular, \tilde{u} is bounded by $-2B$ from below.

Proof. The crucial observation is, that for a given $t_0 \in (0, 1)$, every $0 < r \leq s$, and every $v \in \mathcal{V}_u$ with $\|v\|_{\mathcal{V}_u} \leq t_0$ there holds

$$\tilde{u}(r) \geq u(r) - |v(r)| \geq (1 - t_0)u(r) \geq qu_*(r)$$

with $q = 1 - t_0 > 0$; at the same time we have

$$|\tilde{u}(r)| \leq |u(r)| + |v(r)| \leq (1 + t_0)u^*(r) \leq (2 - q)u^*(r)$$

for every $r \geq s$. From this we conclude that under the given assumptions on u and v the regularity condition (2.4) does hold true for some constant $c_\beta > 0$ and all v with $\|v\|_{\mathcal{V}_u} \leq t_0$. Concerning the stability of the pair potential \tilde{u} we refer to the argument utilized by Fisher and Ruelle in [1]: Their construction provides a universal even minorant $\underline{u} : \mathbb{R} \rightarrow \mathbb{R}$ of positive type such that

$$\tilde{u}(r) \geq \underline{u}(r), \quad r > 0,$$

for all $v \in \mathcal{V}_u$ with $\|v\|_{\mathcal{V}_u} \leq t_0$. Inequalities (2.3) and (2.5) then follow with $B = \underline{u}(0)/2$. \square

REMARK 2.2. Take note that the index j^* in (2.5) is not unique, in general. In this case it does not matter which one is being used, but we will assume in the sequel that some choice $\mathbf{R}_N \mapsto j^*(\mathbf{R}_N)$ has been assigned for every $\mathbf{R}_N \in (R^3)^N$. \diamond

Writing

$$U_N(R_1, \dots, R_N) = \sum_{1 \leq i < j \leq N} u(|R_i - R_j|) \quad (2.6)$$

for the configurational Hamiltonian of the system, the *molecular distribution function* $\rho_\Lambda^{(m)}$ for m particles, $m \in \mathbb{N}$, being distributed in Λ is defined to be

$$\rho_\Lambda^{(m)}(\mathbf{R}_m) = \frac{1}{\Xi_\Lambda} \sum_{N=m}^{\infty} \frac{z^N}{(N-m)!} \int_{\Lambda^{N-m}} e^{-\beta U_N(\mathbf{R}_N)} d\mathbf{R}_{m,N}, \quad (2.7)$$

where $\mathbf{R}_m \in \Lambda^m$, $\mathbf{R}_{m,N}$ denotes the $(N-m)$ -tupel $(R_{m+1}, \dots, R_N) \subset \Lambda^{N-m}$ with the coordinates of additional $N-m$ particles, and

$$\Xi_\Lambda = \sum_{N=0}^{\infty} \frac{z^N}{N!} \int_{\Lambda^N} e^{-\beta U_N(\mathbf{R}_N)} d\mathbf{R}_N \quad (2.8)$$

is the *grand canonical partition function* for a given inverse temperature β and activity z . The formulations in (2.7) and (2.8) obey the usual convention that the integral of the constant one over Λ^0 is considered to be one.

It is shown in [9] that under the given assumptions on u and for an inverse temperature β and activity z satisfying

$$0 < z < \frac{1}{c_\beta e^{2\beta B+1}} \quad (2.9)$$

the distribution function $\rho_\Lambda^{(m)}$ is bounded in Λ^m , its bound being independent of the size of Λ , and in the *thermodynamical limit* $|\Lambda| \rightarrow \infty$ the distribution functions converge compactly, i.e., uniformly on every compact subset of $(\mathbb{R}^3)^m$; we denote by $\rho^{(m)} : (\mathbb{R}^3)^m \rightarrow \mathbb{R}_0^+$ the corresponding limit function. When $m = 1$ the resulting limit is the constant *counting density* ρ_0 of the system; when $m = 2$ the limit $\rho^{(2)}$ is invariant under translations and rotations and one can define the *radial distribution function* $g : \mathbb{R}^+ \rightarrow \mathbb{R}_0^+$ by

$$g(r) = \frac{1}{\rho_0^2} \rho^{(2)}(R_1, R_2), \quad r = |R_1 - R_2|.$$

In this work we investigate the dependence of the molecular distribution functions on u and we are going to prove the following two results.

THEOREM 1. *Let u satisfy Assumption A and z and β be constrained by (2.9). Then for every $m \in \mathbb{N}$ and every box $\Lambda \subset \mathbb{R}^3$ the molecular distribution function $\rho_\Lambda^{(m)}$*

has a well-defined Fréchet derivative $\partial \rho_\Lambda^{(m)} \in \mathcal{L}(\mathcal{V}_u, L^\infty(\Lambda^m))$ with respect to u , and $\rho^{(m)}$ has a Fréchet derivative $\partial \rho^{(m)} \in \mathcal{L}(\mathcal{V}_u, L^\infty((\mathbb{R}^3)^m))$.

THEOREM 2. *Under the assumptions of Theorem 1 the derivative operator $\partial \rho_\Lambda^{(m)}$ converges to $\partial \rho^{(m)}$ in the thermodynamical limit $|\Lambda| \rightarrow \infty$ in the following sense: For any fixed bounded box $\Lambda' \subset \mathbb{R}^3$ and any $m \in \mathbb{N}$ there holds*

$$\|(\partial \rho_\Lambda^{(m)})v - (\partial \rho^{(m)})v\|_{\Lambda'^m} \rightarrow 0,$$

uniformly for $v \in \mathcal{V}_u$ with $\|v\|_{\mathcal{V}_u} \leq 1$.

The proofs of these two theorems utilize the Kirkwood-Salsburg equations. As such, the Fréchet derivatives are only given implicitly as the solution of a semi-infinite linear system. For a bounded box $\Lambda \subset \mathbb{R}^3$ and associated molecular distribution functions $\rho_\Lambda^{(m)}$ more explicit formulae can be derived directly from (2.7), and we will do so for $m = 1$ and $m = 2$ in Section 5; we mention, though, that it is much more difficult to investigate the thermodynamical limit of the latter and to prove differentiability of $\rho^{(m)}$ by this approach.

3. Proof of Theorem 1. In the sequel we consider a generic box $\Lambda \subset \mathbb{R}^3$ centered at the origin, and, by some abuse of notation, we will even allow Λ to be the entire space.

To begin with we recall the definition (2.5) of the index $j^* = j^*(\mathbf{R}_m)$ for a given $\mathbf{R}_m \in \Lambda^m$; see also Remark 2.2. In the sequel we will omit the argument \mathbf{R}_m for the ease of simplicity, as long as there is no danger of confusion. Given this definition we introduce the (nonlinear) projection $\Pi_m : (\mathbb{R}^3)^m \rightarrow (\mathbb{R}^3)^{m-1}$ via

$$\Pi_m : \mathbf{R}_m \mapsto (\mathbf{R}_{j^*-1}, \mathbf{R}_{j^*,m}). \quad (3.1)$$

We also need to define the following two function sequences $d_m : (\mathbb{R}^3)^m \rightarrow \mathbb{R}$, $m \in \mathbb{N}$, and $k_n : \mathbb{R}^3 \times (\mathbb{R}^3)^n \rightarrow \mathbb{R}$, $n \in \mathbb{N}$, given by

$$d_m(\mathbf{R}_m) = \prod_{\substack{i=1 \\ i \neq j^*}}^m e^{-\beta u(|R_i - R_{j^*}|)}, \quad m \in \mathbb{N}, \quad (3.2)$$

and

$$k_n(R; \mathbf{R}'_n) = \prod_{i=1}^n f(R'_i - R), \quad (3.3)$$

where f is the Mayer function (2.1). The latter are utilized as kernel functions for the integral operators

$$(K_{mn,\Lambda} \varphi_{m+n-1})(\mathbf{R}_m) = \frac{1}{n!} \int_{\Lambda^n} k_n(R_{j^*}; \mathbf{R}'_n) \varphi_{m+n-1}(\Pi_m(\mathbf{R}_m), \mathbf{R}'_n) d\mathbf{R}'_n \quad (3.4)$$

for $m, n \in \mathbb{N}$, and $\varphi_{m+n-1} : \Lambda^{m+n-1} \rightarrow \mathbb{R}$. Finally, we need the extension operators

$$(I_{m,\Lambda} \varphi_{m-1})(\mathbf{R}_m) = \varphi_{m-1}(\Pi_m(\mathbf{R}_m)) \quad (3.5)$$

for $m \in \mathbb{N} \setminus \{1\}$ and $\varphi_{m-1} : \Lambda^{m-1} \rightarrow \mathbb{R}$.

Following Ruelle [9] we introduce the Banach space \mathcal{X}_Λ of sequences $\varphi = (\varphi_m)_m$ of bounded functions $\varphi_m : \Lambda^m \rightarrow \mathbb{R}$, for which the norm

$$\|\varphi\|_{\mathcal{X}_\Lambda} = \sup_{m \in \mathbb{N}} c_\beta^m \|\varphi_m\|_{\Lambda^m}$$

with c_β of (2.4) is finite. On \mathcal{X}_Λ a diagonal multiplication operator

$$D_\Lambda = \begin{bmatrix} d_1 & & & \\ & d_2 & & \\ & & \ddots & \\ & & & \ddots \end{bmatrix} \quad (3.6)$$

and a semi-infinite block integral operator

$$K_\Lambda = \begin{bmatrix} K_{11,\Lambda} & K_{12,\Lambda} & K_{13,\Lambda} & \dots \\ I_{2,\Lambda} & K_{21,\Lambda} & K_{22,\Lambda} & \\ 0 & I_{3,\Lambda} & K_{31,\Lambda} & \ddots \\ \vdots & \ddots & \ddots & \ddots \end{bmatrix} \quad (3.7)$$

are defined by utilizing the functions d_m of (3.2) and the operators $K_{mn,\Lambda}$ and $I_{m,\Lambda}$ of (3.4) and (3.5), respectively. It is shown in [9] that D_Λ and K_Λ belong to $\mathcal{L}(\mathcal{X}_\Lambda)$ with

$$\|D_\Lambda\|_{\mathcal{L}(\mathcal{X}_\Lambda)} \leq e^{2\beta B}, \quad \|K_\Lambda\|_{\mathcal{L}(\mathcal{X}_\Lambda)} \leq c_\beta e. \quad (3.8)$$

These operators can now be used to formulate the celebrated Kirkwood-Salsburg equations:

$$(I - zA_\Lambda)\boldsymbol{\rho}_\Lambda = z\mathbf{e}_1, \quad A_\Lambda = D_\Lambda K_\Lambda, \quad (3.9)$$

where

$$\boldsymbol{\rho}_\Lambda = (\rho_\Lambda^{(m)})_m \quad \text{and} \quad \mathbf{e}_1 = (\delta_{1m})_m,$$

with δ_{1m} being the Kronecker symbol and z being the activity. Since

$$\|A_\Lambda\|_{\mathcal{L}(\mathcal{X}_\Lambda)} \leq c_\beta e^{2\beta B+1} \quad (3.10)$$

according to (3.8) it follows from (2.9) that the linear system (3.9) is uniquely solvable for $\boldsymbol{\rho}_\Lambda \in \mathcal{X}_\Lambda$.

To compute the derivative of the molecular distribution functions we need to determine the derivatives of the functions d_m , f , and k_n in appropriate topologies. This is the purpose of the following three auxiliary results.

LEMMA 3.1. *Let u satisfy Assumption A. Then the Mayer f -function has a Fréchet derivative $\partial f \in \mathcal{L}(\mathcal{V}_u, L^1(\mathbb{R}^3))$ with respect to u given by*

$$(\partial f)v = -\beta e^{-\beta u} v, \quad v \in \mathcal{V}_u,$$

i.e., there exists $C_\beta > 0$ such that

$$\|(\partial f)v\|_{L^1(\mathbb{R}^3)} \leq C_\beta \|v\|_{\mathcal{V}_u}, \quad (3.11a)$$

and for $\|v\|_{\mathcal{V}_u} \leq t_0$ and \tilde{f} the Mayer function associated with $\tilde{u} = u + v$ there holds

$$\|\tilde{f} - f\|_{L^1(\mathbb{R}^3)} \leq C_\beta \|v\|_{\mathcal{V}_u}, \quad (3.11b)$$

$$\|\tilde{f} - f - (\partial f)v\|_{L^1(\mathbb{R}^3)} \leq C_\beta \|v\|_{\mathcal{V}_u}^2. \quad (3.11c)$$

Proof. For $v \in \mathcal{V}_u$ Taylor's theorem yields

$$\begin{aligned} \|\tilde{f} - f - (\partial f)v\|_{L^1(\mathbb{R}^3)} &= 4\pi \int_0^\infty |e^{-\beta\tilde{u}(r)} - e^{-\beta u(r)} + \beta e^{-\beta u(r)} v(r)| r^2 dr \\ &= 4\pi \int_0^\infty e^{-\beta u(r)} |e^{-\beta v(r)} - 1 + \beta v(r)| r^2 dr \\ &\leq 2\pi \int_0^\infty e^{-\beta(u(r)-|v(r)|)} |\beta v(r)|^2 r^2 dr. \end{aligned} \quad (3.12)$$

When $\|v\|_{\mathcal{V}_u} \leq t_0$ then it follows from Proposition 2.1 that

$$e^{-\beta(u(r)-|v(r)|)} |\beta v(r)|^2 r^2 \leq \beta^2 e^{2\beta B} \|v\|_{\mathcal{V}_u}^2 u^*(s) u^*(r) r^2$$

for $r > s$, while for $0 < r \leq s$ we have

$$\begin{aligned} e^{-\beta(u(r)-|v(r)|)} |\beta v(r)|^2 r^2 &\leq \beta^2 \|v\|_{\mathcal{V}_u}^2 e^{-\beta(1-t_0)u(r)} u^2(r) s^2 \\ &\leq \frac{4s^2}{e^2(1-t_0)^2} \|v\|_{\mathcal{V}_u}^2. \end{aligned}$$

Inserting these estimates into (3.12) we readily arrive at (3.11c).

In much the same way we can estimate

$$e^{-\beta u(r)} |\beta v(r)| r^2 \leq \begin{cases} \beta e^{2\beta B} \|v\|_{\mathcal{V}_u} u^*(r) r^2, & r \geq s, \\ (s^2/e) \|v\|_{\mathcal{V}_u}, & 0 < r \leq r_0, \end{cases} \quad (3.13)$$

to deduce (3.11a). Take note that (3.13) holds true for every $v \in \mathcal{V}_u$.

When u is replaced by $u - |v|$ with $\|v\|_{\mathcal{V}_u} \leq t_0$ in (3.13) then the upper bound on the right-hand side increases by at most $1/(1-t_0)$, and hence, (3.13) also provides a convergent majorant to estimate

$$\|\tilde{f} - f\|_{L^1(\mathbb{R}^3)} = 4\pi \int_0^\infty e^{-\beta u(r)} |e^{-\beta v(r)} - 1| r^2 dr \leq 4\pi \int_0^\infty e^{-\beta(u(r)-|v(r)|)} |\beta v(r)| r^2 dr$$

for $\|v\|_{\mathcal{V}_u} \leq t_0$, cf. (3.11b). \square

LEMMA 3.2. *Let u satisfy Assumption A. Then the functions d_m of (3.2) are Fréchet differentiable with respect to the pair potential with derivative $\partial d_m \in \mathcal{L}(\mathcal{V}_u, L^\infty(\mathbb{R}^3))$ given by*

$$((\partial d_m)v)(\mathbf{R}_m) = -\beta d_m(\mathbf{R}_m) \sum_{\substack{i=1 \\ i \neq j^*}}^m v(|R_i - R_{j^*}|)$$

for $v \in \mathcal{V}_u$ and $\mathbf{R}_m \in (\mathbb{R}^3)^m$. There holds

$$\|(\partial d_m)v\|_{(\mathbb{R}^3)^m} \leq \frac{e^{2\beta B}}{t_0} \|v\|_{\mathcal{V}_u} \quad (3.14a)$$

and, if $\|v\|_{\mathcal{V}_u} \leq t_0/2$ then

$$\|\tilde{d}_m - d_m\|_{(\mathbb{R}^3)^m} \leq \frac{2e^{2\beta B}}{t_0} \|v\|_{\mathcal{V}_u}, \quad (3.14b)$$

$$\|\tilde{d}_m - d_m - (\partial d_m)v\|_{(\mathbb{R}^3)^m} \leq \frac{4e^{2\beta B}}{t_0^2} \|v\|_{\mathcal{V}_u}^2, \quad (3.14c)$$

where \tilde{d}_m denotes the function d_m associated with the pair potential $u + v$.

Proof. For the proof of this result it is essential that the chosen index $j^* = j^*(\mathbf{R}_m)$ of (2.5) is independent of the particular pair potential $\tilde{u} = u + v$ as long as $\|v\|_{\mathcal{V}_u} \leq t_0$. For $v \in \mathcal{V}_u$ and fixed $\mathbf{R}_m \in (\mathbb{R}^3)^m$ we use Taylor's theorem to estimate

$$|\tilde{d}_m - d_m - (\partial d_m)v| = |d_m(e^{-\beta \sum v} - 1 + \beta \sum v)| \leq \frac{1}{2} |e^{-\beta \sum u} (\beta \sum v)^2 e^{\beta \sum |v|}|,$$

where all sums extend over every $i = 1, \dots, m$ with $i \neq j^*$, the respective arguments being given by $|R_i - R_{j^*}|$. For every $\gamma > 0$ there holds

$$\frac{1}{2} (\beta \sum v)^2 \leq \gamma^2 \frac{1}{2} \left(\frac{\beta}{\gamma} \sum |v| \right)^2 \leq \gamma^2 e^{\beta \sum |v|/\gamma},$$

and hence

$$|\tilde{d}_m - d_m - (\partial d_m)v| \leq \gamma^2 e^{-\beta \sum (u - |v| - |v|/\gamma)}.$$

For $\gamma = 2\|v\|_{\mathcal{V}_u}/t_0$ the assertion (3.14c) now follows from (2.5) with $\tilde{u} = u - |v| - |v|/\gamma$ provided that $\|v\|_{\mathcal{V}_u} \leq t_0/2$. This proves the differentiability of d_m . The proof of the two estimates (3.14a) and (3.14b) follows along the same lines and is left to the reader. \square

To formulate the following result we introduce the formal derivative of k_n of (3.3) with respect to u in direction v , i.e.,

$$k'_n(R; \mathbf{R}'_n) = \sum_{i=1}^n ((\partial f)v)(|R'_i - R|) \prod_{\substack{j=1 \\ j \neq i}}^n f(R'_j - R) \quad (3.15)$$

for $R \in \mathbb{R}^3$ and $\mathbf{R}'_n \in (\mathbb{R}^3)^n$.

PROPOSITION 3.3. *Under the assumptions of Theorem 1 the operator K_Λ is Fréchet differentiable with respect to u in $\mathcal{L}(\mathcal{V}_u, \mathcal{L}(\mathcal{X}_\Lambda))$, and its derivative ∂K_Λ is given by*

$$(\partial K_\Lambda)v = \begin{bmatrix} K'_{11,\Lambda} & K'_{12,\Lambda} & K'_{13,\Lambda} & \cdots \\ 0 & K'_{21,\Lambda} & K'_{22,\Lambda} & \\ \vdots & 0 & K'_{31,\Lambda} & \ddots \\ \vdots & & \ddots & \ddots \end{bmatrix}, \quad (3.16)$$

where

$$(K'_{mn,\Lambda} \varphi_{m+n-1})(\mathbf{R}_m) = \frac{1}{n!} \int_{\Lambda^n} k'_n(R_{j^*}; \mathbf{R}'_n) \varphi_{m+n-1}(\Pi_m(\mathbf{R}_m), \mathbf{R}'_n) d\mathbf{R}'_n$$

for $m \in \mathbb{N}$, with $\varphi_{m+n-1} : \Lambda^{m+n-1} \rightarrow \mathbb{R}$, Π_m of (3.1), and k'_n of (3.15).

Proof. Let \mathcal{Y}_n be the Banach space of functions $k : \mathbb{R}^3 \times (\mathbb{R}^3)^n \rightarrow \mathbb{R}$ with norm

$$\|k\|_{\mathcal{Y}_n} = \sup_{R \in \mathbb{R}^3} \|k(R; \cdot)\|_{L^1((\mathbb{R}^3)^n)}.$$

We prove that k_n of (3.3) is differentiable with respect to u in $\mathcal{L}(\mathcal{V}_u, \mathcal{Y}_n)$, and that its derivative $(\partial k_n)v$ in direction v is given by k'_n of (3.15). From Lemma 3.1 we readily conclude that $k'_n \in \mathcal{Y}_n$ with

$$\|k'_n\|_{\mathcal{Y}_n} \leq n C_\beta c_\beta^{n-1} \|v\|_{\mathcal{V}_u}. \quad (3.17)$$

We now assume that $\|v\|_{\mathcal{V}_u} \leq t_0$ and introduce short-hand notations \tilde{k}_n for the kernel k_n associated with the pair potential $\tilde{u} = u + v$, and let

$$f_i(R) = e^{-\beta u(|R'_i - R|)} - 1 \quad \text{and} \quad \tilde{f}_i(R) = e^{-\beta \tilde{u}(|R'_i - R|)} - 1$$

for every appropriate value of i ; in the sequel we also omit the obvious arguments R'_i and R , respectively. Then we prove by induction on $n \in \mathbb{N}$ that

$$\|\tilde{k}_n - k_n\|_{\mathcal{V}_n} \leq n C_\beta c_\beta^{n-1} \|v\|_{\mathcal{V}_u}, \quad n \in \mathbb{N}, \quad (3.18)$$

which is obviously true because of (3.11b) when $n = 1$, because $\tilde{k}_1 - k_1 = \tilde{f}_1 - f_1$. The inductive step is then based on

$$\begin{aligned} \tilde{k}_{n+1} - k_{n+1} &= \tilde{k}_n \tilde{f}_{n+1} - k_n f_{n+1} = \tilde{k}_n (\tilde{f}_{n+1} - f_{n+1}) + (\tilde{k}_n - k_n) f_{n+1} \\ &= \tilde{f}_1 \cdots \tilde{f}_n (\tilde{f}_{n+1} - f_{n+1}) + (\tilde{k}_n - k_n) f_{n+1}, \end{aligned}$$

the induction hypothesis, and on the estimates (3.11b) and (2.4).

Still assuming that $\|v\|_{\mathcal{V}_u} \leq t_0$ we now proceed to establish the inequality

$$\|\tilde{k}_n - k_n - k'_n\|_{\mathcal{V}_n} \leq n^2 C'_\beta c_\beta^{n-1} \|v\|_{\mathcal{V}_u}^2, \quad n \in \mathbb{N}, \quad (3.19)$$

for $C'_\beta = \max\{C_\beta, C_\beta^2/(2c_\beta)\}$, which proves the asserted differentiability of k_n . For $n = 1$ inequality (3.19) has been established in the proof of Lemma 3.1, cf. (3.11c). For the induction step we write

$$\begin{aligned} \tilde{k}_{n+1} - k_{n+1} - k'_{n+1} &= \tilde{k}_n \tilde{f}_{n+1} - k_n f_{n+1} - k_n (\partial f_{n+1}) v_{n+1} - k'_n f_{n+1} \\ &= (\tilde{k}_n - k_n - k'_n) f_{n+1} + (\tilde{k}_n - k_n) (\tilde{f}_{n+1} - f_{n+1}) \\ &\quad + f_1 \cdots f_n (\tilde{f}_{n+1} - f_{n+1} - (\partial f_{n+1}) v_{n+1}), \end{aligned}$$

where we have set

$$v_{n+1} = v(|R'_{n+1} - R|).$$

Then it follows from the induction hypothesis (3.19), and from (2.4), (3.18), (3.11b), and (3.11c) that

$$\|\tilde{k}_{n+1} - k_{n+1} - k'_{n+1}\|_{\mathcal{V}_{n+1}} \leq (n^2 C'_\beta c_\beta^n + n C_\beta^2 c_\beta^{n-1} + c_\beta^n C_\beta) \|v\|_{\mathcal{V}_u}^2.$$

Inserting the definition of C'_β we further conclude that the above right-hand side satisfies

$$(n^2 C'_\beta + 2n \frac{C_\beta^2}{2c_\beta} + C_\beta) c_\beta^n \|v\|_{\mathcal{V}_u}^2 \leq (n+1)^2 C'_\beta c_\beta^n \|v\|_{\mathcal{V}_u}^2,$$

hence the induction step is complete.

Having established (3.19) we can now argue as in [9] to show that

$$\|\tilde{K}_\Lambda - K_\Lambda - (\partial K_\Lambda) v\|_{\mathcal{L}(\mathcal{X}_\Lambda)} \leq 2e C'_\beta \|v\|_{\mathcal{V}_u}^2,$$

where \tilde{K}_Λ denotes the block integral operator (3.7) associated with \tilde{u} , $(\partial K_\Lambda) v$ is defined in (3.16), and $\|v\|_{\mathcal{V}_u} \leq t_0$. This shows that ∂K_Λ of (3.16) is the Fréchet derivative of K_Λ when considered a function of the pair potential. \square

Now we can establish Theorem 1.

Proof of Theorem 1. From Lemma 3.2 it follows readily that D_Λ is also Fréchet differentiable as a function of u , and so is $A_\Lambda = D_\Lambda K_\Lambda$, and the derivative ∂A_Λ of A_Λ is given by

$$(\partial A_\Lambda)v = ((\partial D_\Lambda)v)K_\Lambda + D_\Lambda(\partial K_\Lambda)v$$

for $v \in \mathcal{V}_u$. Denote by \tilde{D}_Λ and \tilde{K}_Λ the operators (3.6) and (3.7) associated with the pair potential $\tilde{u} = u + v$, and set $\tilde{A}_\Lambda = \tilde{D}_\Lambda \tilde{K}_\Lambda$. Moreover, let $\tilde{\rho}_\Lambda$ be the respective sequence of molecular distribution functions. Then, for $\|v\|_{\mathcal{V}_u} \leq t_0$ there holds

$$\begin{aligned} \tilde{\rho}_\Lambda - \rho_\Lambda &= ((I - z\tilde{A}_\Lambda)^{-1} - (I - zA_\Lambda)^{-1})ze_1 \\ &= z(I - z\tilde{A}_\Lambda)^{-1}(\tilde{A}_\Lambda - A_\Lambda)(I - zA_\Lambda)^{-1}ze_1 \\ &= z(I - zA_\Lambda)^{-1}((\partial A_\Lambda)v)(I - zA_\Lambda)^{-1}ze_1 + O(\|v\|_{\mathcal{V}_u}^2) \end{aligned}$$

in \mathcal{X}_Λ . Accordingly, ρ_Λ is Fréchet differentiable, and its Fréchet derivative $\partial \rho_\Lambda \in \mathcal{L}(\mathcal{V}_u, \mathcal{X}_\Lambda)$ satisfies

$$(\partial \rho_\Lambda)v = z(I - zA_\Lambda)^{-1}((\partial A_\Lambda)v)(I - zA_\Lambda)^{-1}ze_1 = z(I - zA_\Lambda)^{-1}((\partial A_\Lambda)v)\rho_\Lambda.$$

This implies the statement of Theorem 1. \square

REMARK 3.4. One can also consider the linear system

$$(I - zB_\Lambda)\sigma_\Lambda = ze_1, \quad B_\Lambda = K_\Lambda D_\Lambda \in \mathcal{L}(\mathcal{X}_\Lambda), \quad (3.20)$$

for $\sigma \in \mathcal{X}_\Lambda$, and use the same arguments as before to establish unique solvability and differentiability of σ_Λ with $\partial \sigma_\Lambda \in \mathcal{L}(\mathcal{V}_u, \mathcal{X}_\Lambda)$. Obviously, from (3.9), there holds

$$\rho_\Lambda = D_\Lambda \sigma_\Lambda,$$

and as a corollary one obtains, for example, that

$$\|\rho_\Lambda^{(m)}\|_{\Lambda^m} \leq Cc_\beta^{-m}, \quad m = 1, 2, \dots,$$

with a uniform constant $C > 0$. While this latter statement also follows readily from [9], the differentiability of σ_Λ provides additional information about the molecular distribution functions. \diamond

4. Proof of Theorem 2. In this section we investigate the thermodynamical limit of the sequence $\rho_\Lambda = (\rho_\Lambda^{(m)})_m \in \mathcal{X}_\Lambda$ of molecular distribution functions; throughout this section $\Lambda \subset \mathbb{R}^3$ always denotes a finite size box. Take note that when $\Lambda' \subset \Lambda$ then any sequence $\varphi_\Lambda = (\varphi_\Lambda^{(m)})_m \in \mathcal{X}_\Lambda$ also belongs to $\mathcal{X}_{\Lambda'}$. We will make repeated use of this property without further mentioning by taking the corresponding norm $\|\varphi_\Lambda\|_{\mathcal{X}_{\Lambda'}}$ of φ_Λ . Often, however, it will be convenient to make this embedding more explicit. To this end we introduce the corresponding embedding operator

$$P_{\Lambda'} : \mathcal{X}_\Lambda \rightarrow \mathcal{X}_{\Lambda'}, \quad P_{\Lambda'}\varphi_\Lambda = (\varphi_\Lambda^{(m)}|_{(\Lambda')^m})_m. \quad (4.1)$$

To simplify matters, though, we will not specify explicitly the domain \mathcal{X}_Λ of $P_{\Lambda'}$ in this notation; this domain may differ depending on the context.

In the previous section we have shown that $\boldsymbol{\rho}_\Lambda$ as well as its thermodynamical limit $\boldsymbol{\rho}$ are differentiable with respect to u , their derivative(s) being given by

$$\begin{aligned} (\partial \boldsymbol{\rho}_\Lambda)v &= z(I - zA_\Lambda)^{-1}A'_\Lambda \boldsymbol{\rho}_\Lambda, \\ (\partial \boldsymbol{\rho})v &= z(I - zA)^{-1}A' \boldsymbol{\rho}, \end{aligned} \quad (4.2)$$

respectively. In (4.2) we have used short-hand notations A' and A'_Λ for $(\partial A)v$ and $(\partial A_\Lambda)v$, respectively; we will continue to do so in the remainder of this section as long as $v \in \mathcal{V}_u$ is fixed.

The proof of Theorem 2 will proceed in three steps: First, in Lemma 4.1, we will show that for two boxes $\Lambda'' \subset \Lambda$ with Λ'' being kept fixed we have convergence

$$\|A'_\Lambda \boldsymbol{\rho}_\Lambda - A' \boldsymbol{\rho}\|_{\mathcal{X}_{\Lambda''}} \rightarrow 0 \quad \text{as } |\Lambda| \rightarrow \infty. \quad (4.3)$$

Then, in a second step we fix another box $\Lambda' \subset \Lambda$, and we argue that

$$\|(I - zA_\Lambda)^{-1}P_\Lambda A' \boldsymbol{\rho} - (I - zA)^{-1}A' \boldsymbol{\rho}\|_{\mathcal{X}_{\Lambda'}} \rightarrow 0 \quad \text{as } |\Lambda| \rightarrow \infty. \quad (4.4)$$

As the third part of the proof we use (4.3) in a setting with three boxes $\Lambda' \subset \Lambda'' \subset \Lambda$ to show that

$$\|(I - zA_\Lambda)^{-1}P_\Lambda(A' \boldsymbol{\rho} - A'_\Lambda \boldsymbol{\rho}_\Lambda)\|_{\mathcal{X}_{\Lambda'}} \rightarrow 0 \quad \text{as } |\Lambda| \rightarrow \infty, \quad (4.5)$$

provided that Λ' is kept fixed. A combination of (4.4) and (4.5) then readily yields the desired convergence

$$\|(\partial \boldsymbol{\rho}_\Lambda)v - (\partial \boldsymbol{\rho})v\|_{\mathcal{X}_{\Lambda'}} \rightarrow 0$$

as $|\Lambda| \rightarrow \infty$, which completes the proof of Theorem 2.

LEMMA 4.1. *Let u satisfy the assumptions of Theorem 1 and let A'_Λ and A' be defined as above for a given $v \in \mathcal{V}_u$. Then, for a fixed box $\Lambda'' \subset \mathbb{R}^3$ and for $\Lambda'' \subset \Lambda \subset \mathbb{R}^3$ the convergence (4.3) holds true uniformly for all $v \in \mathcal{V}_u$ with $\|v\|_{\mathcal{V}_u} \leq 1$.*

Proof. Let $\varepsilon > 0$, $d > s$, and a certain box $\Lambda'' \subset \mathbb{R}^3$ be given. We choose a second box $\Lambda^* \supset \Lambda''$ in such a way that $|R'' - R| > d$ for every $R'' \in \Lambda''$ and every $R \in \mathbb{R}^3 \setminus \Lambda^*$. According to [9] there holds

$$\|\boldsymbol{\rho}_\Lambda - \boldsymbol{\rho}\|_{\mathcal{X}_{\Lambda^*}} \leq \varepsilon,$$

provided that $\Lambda \supset \Lambda^*$ is sufficiently large; moreover,

$$\|\boldsymbol{\rho}\|_{\mathcal{X}_{\mathbb{R}^3}}, \|\boldsymbol{\rho}_\Lambda\|_{\mathcal{X}_\Lambda} \leq C_*$$

for some $C_* > 0$, independent of the size of Λ . Then, for any $\mathbf{R}_m \in (\Lambda'')^m$ and any

$n \in \mathbb{N}$ we can estimate

$$\begin{aligned}
& |(K_{mn,\Lambda} \rho_\Lambda^{(m+n-1)})(\mathbf{R}_m) - (K_{mn} \rho^{m+n-1})(\mathbf{R}_m)| \\
&= \frac{1}{n!} \left| \int_{\Lambda^n} k_n(R_{j^*}; \mathbf{R}'_n) \rho_\Lambda^{(m+n-1)}(\Pi_m(\mathbf{R}_m), \mathbf{R}'_n) \, d\mathbf{R}'_n \right. \\
&\quad \left. - \int_{(\mathbb{R}^3)^n} k_n(R_{j^*}; \mathbf{R}'_n) \rho^{(m+n-1)}(\Pi_m(\mathbf{R}_m), \mathbf{R}'_n) \, d\mathbf{R}'_n \right| \\
&= \frac{1}{n!} \left| \int_{(\Lambda^*)^n} k_n(R_{j^*}; \mathbf{R}'_n) (\rho_\Lambda^{(m+n-1)}(\Pi_m(\mathbf{R}_m), \mathbf{R}'_n) - \rho^{(m+n-1)}(\Pi_m(\mathbf{R}_m), \mathbf{R}'_n)) \, d\mathbf{R}'_n \right. \\
&\quad + \int_{\Lambda^n \setminus (\Lambda^*)^n} k_n(R_{j^*}; \mathbf{R}'_n) \rho_\Lambda^{(m+n-1)}(\Pi_m(\mathbf{R}_m), \mathbf{R}'_n) \, d\mathbf{R}'_n \\
&\quad \left. - \int_{(\mathbb{R}^3)^n \setminus (\Lambda^*)^n} k_n(R_{j^*}; \mathbf{R}'_n) \rho^{(m+n-1)}(\Pi_m(\mathbf{R}_m), \mathbf{R}'_n) \, d\mathbf{R}'_n \right| \\
&\leq \frac{c_\beta^{1-m-n}}{n!} \left(\varepsilon \int_{(\mathbb{R}^3)^n} |k_n(R_{j^*}; \mathbf{R}'_n)| \, d\mathbf{R}'_n + 2C_* \int_{(\mathbb{R}^3)^n \setminus (\Lambda^*)^n} |k_n(R_{j^*}; \mathbf{R}'_n)| \, d\mathbf{R}'_n \right). \quad (4.6)
\end{aligned}$$

The first integral in (4.6) is bounded by c_β^n , the second integral can be estimated by $nc_\beta^{n-1}c_{\beta,d}$, where

$$c_{\beta,d} = 4\pi \int_d^\infty |e^{-\beta u(r)} - 1| r^2 \, dr, \quad (4.7)$$

compare the proof of [9, Theorem 4.2.3]. We thus have

$$c_\beta^m \|K_{mn,\Lambda} \rho_\Lambda^{(m+n-1)} - K_{mn} \rho^{(m+n-1)}\|_{(\Lambda'')^m} \leq \frac{c_\beta}{n!} \varepsilon + \frac{2C_*}{(n-1)!} c_{\beta,d},$$

which readily yields

$$\|K_\Lambda \rho_\Lambda - K \rho\|_{\mathcal{X}_{\Lambda''}} \leq e(c_\beta \varepsilon + 2C_* c_{\beta,d}).$$

Since we can let d be arbitrarily large and $\varepsilon > 0$ be arbitrarily small by choosing $|\Lambda^*|$ and $|\Lambda|$ sufficiently big, it follows that

$$\|K_\Lambda \rho_\Lambda - K \rho\|_{\mathcal{X}_{\Lambda''}} \rightarrow 0 \quad \text{as } |\Lambda| \rightarrow \infty. \quad (4.8)$$

We now write D'_Λ for $(\partial D_\Lambda)v$ and K'_Λ for $(\partial K_\Lambda)v$ for some fixed $v \in \mathcal{V}_u$ and any box $\Lambda \subset \mathbb{R}^3$. Then we conclude from (4.8) with the help of (3.14a) that

$$\begin{aligned}
\|D'_\Lambda K_\Lambda \rho_\Lambda - D'K \rho\|_{\mathcal{X}_{\Lambda''}} &= \|D'_{\Lambda''} P_{\Lambda''}(K_\Lambda \rho_\Lambda - K \rho)\|_{\mathcal{X}_{\Lambda''}} \\
&\leq \|D'_{\Lambda''}\|_{\mathcal{L}(\mathcal{X}_{\Lambda''})} \|K_\Lambda \rho_\Lambda - K \rho\|_{\mathcal{X}_{\Lambda''}} \leq \frac{e^{2\beta B}}{t_0} \|K_\Lambda \rho_\Lambda - K \rho\|_{\mathcal{X}_{\Lambda''}} \|v\|_{\mathcal{V}_u} \rightarrow 0
\end{aligned}$$

as $|\Lambda| \rightarrow \infty$, uniformly for all $v \in \mathcal{V}_u$ with $\|v\|_{\mathcal{V}_u} \leq 1$.

In a similar way we can estimate

$$\|D_\Lambda K'_\Lambda \rho_\Lambda - DK' \rho\|_{\mathcal{X}_{\Lambda''}},$$

the main difference for this estimate being that k_n in (4.6) has to be replaced by k'_n of (3.15), i.e.,

$$\begin{aligned} & |(K'_{mn,\Lambda}\rho_\Lambda^{(m+n-1)})(\mathbf{R}_m) - (K'_{mn}\rho^{m+n-1})(\mathbf{R}_m)| \\ & \leq \frac{c_\beta^{1-m-n}}{n!} \left(\varepsilon \int_{(\mathbb{R}^3)^n} |k'_n(R_{j^*}; \mathbf{R}'_n)| d\mathbf{R}'_n + 2C_* \int_{(\mathbb{R}^3)^n \setminus (\Lambda^*)^n} |k'_n(R_{j^*}; \mathbf{R}'_n)| d\mathbf{R}'_n \right) \end{aligned} \quad (4.9)$$

for $\mathbf{R}_m \in (\Lambda'')^m$. Here we can use the bound (3.17) for the first integral, but the second integral requires a different estimate. Following the line of argument employed previously, we obtain from (3.15) the inequality

$$\begin{aligned} & \int_{(\mathbb{R}^3)^n \setminus (\Lambda^*)^n} |k'_n(R_{j^*}; \mathbf{R}'_n)| d\mathbf{R}'_n \\ & \leq \sum_{i=1}^n \int_{(\mathbb{R}^3)^n \setminus (\Lambda^*)^n} |((\partial f)v)(|R'_i - R_{j^*}|)| \prod_{j \neq i} |f(|R'_j - R_{j^*}|)| d\mathbf{R}'_n \\ & \leq \sum_{i=1}^n \left((n-1) \int_{\mathbb{R}^3} |((\partial f)v)(|R|)| dR \left(\int_{\mathbb{R}^3} |f(|R|)| dR \right)^{n-2} \int_{\mathbb{R}^3 \setminus \Lambda^*} |f(|R - R_{j^*}|)| dR \right. \\ & \quad \left. + \int_{\mathbb{R}^3 \setminus \Lambda^*} |((\partial f)v)(|R - R_{j^*}|)| dR \left(\int_{\mathbb{R}^3} |f(|R|)| dR \right)^{n-1} \right). \end{aligned}$$

Using (2.4), (4.7), and (3.11a) this yields

$$\begin{aligned} & \int_{(\mathbb{R}^3)^n \setminus (\Lambda^*)^n} |k'_n(R_{j^*}; \mathbf{R}'_n)| d\mathbf{R}'_n \\ & \leq n(n-1)c_\beta^{n-2}c_{\beta,d}C_\beta \|v\|_{\mathcal{V}_u} + nc_\beta^{n-1} \int_{|R|>d} |((\partial f)v)(|R|)| dR. \end{aligned}$$

Since $d > s$ the remaining integral can be estimated by means of (3.13), and hence,

$$\begin{aligned} \int_{|R|>d} |((\partial f)v)(|R|)| dR &= 4\pi \int_d^\infty \beta e^{-\beta u(r)} |v(r)| r^2 dr \\ &\leq 4\pi \beta e^{2\beta B} \|v\|_{\mathcal{V}_u} \int_d^\infty u^*(r) r^2 dr =: c_{\beta,d}^* \|v\|_{\mathcal{V}_u}. \end{aligned}$$

Take note that $c_{\beta,d}^* \rightarrow 0$ for $d \rightarrow \infty$. We thus conclude from (4.9) that

$$\begin{aligned} & c_\beta^m \|K'_{mn,\Lambda}\rho_\Lambda^{(m+n-1)} - K'_{mn}\rho^{m+n-1}\|_{(\Lambda'')^m} \\ & \leq \left(\frac{C_\beta}{(n-1)!} \varepsilon + \frac{2C_*C_\beta}{c_\beta} \frac{n-1}{(n-1)!} c_{\beta,d} + \frac{2C_*}{(n-1)!} c_{\beta,d}^* \right) \|v\|_{\mathcal{V}_u}. \end{aligned}$$

In view of (3.8) it follows that

$$\begin{aligned} \|D_\Lambda K'_\Lambda \rho_\Lambda - DK' \rho\|_{\mathcal{X}_{\Lambda''}} &= \|D_{\Lambda''} P_{\Lambda''}(K'_\Lambda \rho_\Lambda - K' \rho)\|_{\mathcal{X}_{\Lambda''}} \\ &\leq \|D_{\Lambda''}\|_{\mathcal{L}(\mathcal{X}_{\Lambda''})} \|K'_\Lambda \rho_\Lambda - K' \rho\|_{\mathcal{X}_{\Lambda''}} \\ &\leq e^{2\beta B+1} \left(C_\beta \varepsilon + \frac{2C_*C_\beta}{c_\beta} c_{\beta,d} + 2C_*c_{\beta,d}^* \right) \|v\|_{\mathcal{V}_u}, \end{aligned}$$

which can be made arbitrarily small by choosing $|\Lambda^*|$ and $|\Lambda|$ sufficiently big, uniformly for all $v \in \mathcal{V}_u$ with $\|v\|_{\mathcal{V}_u} \leq 1$.

Since

$$A'_\Lambda \rho_\Lambda - A' \rho = (D'_\Lambda K_\Lambda \rho_\Lambda - D' K \rho) + (D_\Lambda K'_\Lambda \rho_\Lambda - D K' \rho)$$

we thus have established the assertion. \square

As mentioned before another ingredient to the proof of Theorem 2 is the statement (4.4), which we reformulate here in a slightly more precise way.

LEMMA 4.2. *Let u satisfy the assumptions of Theorem 1 and let A' be defined as above for a given $v \in \mathcal{V}_u$. Then, for a fixed box $\Lambda' \subset \mathbb{R}^3$ and for $\Lambda' \subset \Lambda \subset \mathbb{R}^3$ the convergence*

$$\|(I - zA_\Lambda)^{-1} P_\Lambda A' \rho - (I - zA)^{-1} A' \rho\|_{\mathcal{X}_{\Lambda'}} \rightarrow 0 \quad \text{as } |\Lambda| \rightarrow \infty$$

holds true uniformly for all $v \in \mathcal{V}_u$ with $\|v\|_{\mathcal{V}_u} \leq 1$.

Note that if one replaces $A' \rho$ by e_1 in (4.4) then the resulting assertion is that ρ_Λ converges uniformly to ρ in the given box Λ' as $|\Lambda| \rightarrow \infty$, cf. (3.9). In fact, one can reuse the corresponding proof of Ruelle [9, Theorem 4.2.3] with $\alpha = A' \rho$ instead of $\alpha = e_1$ throughout to verify (4.4). The proof of Lemma 4.2 is then an easy consequence because the rate of convergence only depends on the norm of α , and

$$\|A' \rho\|_{\mathcal{X}_{\mathbb{R}^3}} \leq \|\partial A\|_{\mathcal{L}(\mathcal{V}_u, \mathcal{L}(\mathcal{X}_{\mathbb{R}^3}))} \|v\|_{\mathcal{V}_u} \|\rho\|_{\mathcal{X}_{\mathbb{R}^3}} \quad (4.10)$$

is uniformly bounded for $\|v\|_{\mathcal{V}_u} \leq 1$; take note that an upper bound for (4.10) can be chosen in a way to be also an appropriate bound for $\|A'_\Lambda \rho_\Lambda\|_{\mathcal{X}_\Lambda}$.

Proof of Theorem 2. We now turn to a proof of (4.5). To this end we consider a finite number of nested boxes

$$\Lambda' = \Lambda_{k_0} \subset \Lambda_{k_0-1} \subset \dots \subset \Lambda_1 \subset \Lambda, \quad (4.11)$$

each of them centered at the origin, and Λ_k having sides of length

$$\ell_k = \ell_0 + 2(k_0 - k)d, \quad k = 1, \dots, k_0,$$

where ℓ_0 is the length of the sides of the given box Λ' . The particular number k_0 will be chosen later; see (4.13) below. With each box Λ_k we associate the Banach space $\mathcal{X}_k = \mathcal{X}_{\Lambda_k}$, the corresponding projector $P_k = P_{\Lambda_k}$ of (4.1), and the operator $A_k = A_{\Lambda_k} \in \mathcal{L}(\mathcal{X}_k)$. As before we refer to $A = A_{\mathbb{R}^3}$ for the operator corresponding to the full space.

At this stage we quote another auxiliary result, cf. [9, (2.41) in Chapter 4], namely that

$$\begin{aligned} \|P_{k+1} A_1^k P_1 - P_{k+1} A_\Lambda^k\|_{\mathcal{L}(\mathcal{X}_\Lambda, \mathcal{X}_{k+1})} &\leq 2k e^{2\beta B+1} \|A_\Lambda\|_{\mathcal{L}(\mathcal{X}_\Lambda)}^{k-1} c_{\beta,d} \\ &\leq 2k c_\beta^{k-1} e^{k(2\beta B+1)} c_{\beta,d} \end{aligned} \quad (4.12)$$

for $k = 0, 1, \dots, k_0 - 1$. In [9] this result was proved with A_Λ replaced by A (and in $\mathcal{L}(\mathcal{X}_{\mathbb{R}^3}, \mathcal{X}_{k+1})$), however, the argument given there does not need any modification to establish (4.12) as it stands. From (4.12) and (3.10) it follows that

$$\begin{aligned} &\|A_\Lambda^k P_\Lambda (A' \rho - A'_\Lambda \rho_\Lambda)\|_{\mathcal{X}_{k+1}} \\ &\leq \|(A_1^k P_1 - A_\Lambda^k) P_\Lambda (A' \rho - A'_\Lambda \rho_\Lambda)\|_{\mathcal{X}_{k+1}} + \|A_1^k P_1 (A' \rho - A'_\Lambda \rho_\Lambda)\|_{\mathcal{X}_{k+1}} \\ &\leq 2k c_\beta^{k-1} e^{k(2\beta B+1)} c_{\beta,d} \|A' \rho - A'_\Lambda \rho_\Lambda\|_{\mathcal{X}_\Lambda} + c_\beta^k e^{k(2\beta B+1)} \|A' \rho - A'_\Lambda \rho_\Lambda\|_{\mathcal{X}_1}, \end{aligned}$$

and hence, the representation

$$\begin{aligned} & (I - zA_\Lambda)^{-1}P_\Lambda(A'\rho - A'_\Lambda\rho_\Lambda) \\ &= \sum_{k=0}^{k_0-1} z^k A_\Lambda^k P_\Lambda(A'\rho - A'_\Lambda\rho_\Lambda) + \sum_{k=k_0}^{\infty} z^k A_\Lambda^k P_\Lambda(A'\rho - A'_\Lambda\rho_\Lambda) \end{aligned}$$

leads to the upper bound

$$\begin{aligned} & \|(I - zA_\Lambda)^{-1}P_\Lambda(A'\rho - A'_\Lambda\rho_\Lambda)\|_{\mathcal{X}_{\Lambda'}} \\ & \leq \sum_{k=0}^{k_0-1} z^k \|A_\Lambda^k P_\Lambda(A'\rho - A'_\Lambda\rho_\Lambda)\|_{\mathcal{X}_{k+1}} + \sum_{k=k_0}^{\infty} z^k \|A_\Lambda\|_{\mathcal{X}_\Lambda}^k \|A'\rho - A'_\Lambda\rho_\Lambda\|_{\mathcal{X}_\Lambda} \\ & \leq \sum_{k=0}^{k_0-1} 2kc_\beta^{k-1} (ze^{2\beta B+1})^k c_{\beta,d} \|A'\rho - A'_\Lambda\rho_\Lambda\|_{\mathcal{X}_\Lambda} \\ & \quad + \sum_{k=0}^{k_0-1} (zc_\beta e^{2\beta B+1})^k \|A'\rho - A'_\Lambda\rho_\Lambda\|_{\mathcal{X}_1} + \sum_{k=k_0}^{\infty} (zc_\beta e^{2\beta B+1})^k \|A'\rho - A'_\Lambda\rho_\Lambda\|_{\mathcal{X}_\Lambda}. \end{aligned}$$

By virtue of (4.10) $\|A'\rho\|_{\mathcal{X}_{\mathbb{R}^3}}$ and $\|A'_\Lambda\rho_\Lambda\|_{\mathcal{X}_\Lambda}$ are uniformly bounded, hence there exists a constant $c > 0$ (depending only on z and β) with

$$\begin{aligned} & \|(I - zA_\Lambda)^{-1}P_\Lambda(A'\rho - A'_\Lambda\rho_\Lambda)\|_{\mathcal{X}_{\Lambda'}} \\ & \leq c(c_{\beta,d} + \|A'\rho - A'_\Lambda\rho_\Lambda\|_{\mathcal{X}_1} + (zc_\beta e^{2\beta B+1})^{k_0}), \end{aligned} \tag{4.13}$$

provided that $\|v\|_{\mathcal{Y}_u} \leq 1$ and z and β satisfy (2.9).

Now let $\varepsilon > 0$ be given. By virtue of (2.9) and (4.7) we can choose d and k_0 so large that the first and third summand on the right-hand side of (4.13) are both smaller than $\varepsilon/(3c)$. A corresponding choice of nested boxes Λ_k of (4.11) is possible provided that Λ is sufficiently big. In fact, making $|\Lambda|$ even larger the second term on the right-hand side of (4.13) will also become less than $\varepsilon/(3c)$ by virtue of Lemma 4.1, uniformly for $v \in \mathcal{V}_u$ with $\|v\|_{\mathcal{Y}_u} \leq 1$. Accordingly, there holds

$$\|(I - zA_\Lambda)^{-1}P_\Lambda(A'\rho - A'_\Lambda\rho_\Lambda)\|_{\mathcal{X}_{\Lambda'}} \leq \varepsilon$$

for $|\Lambda|$ sufficiently large, which yields (4.5). Again we emphasize that the size of $|\Lambda|$ to achieve a given bound $\varepsilon > 0$ does not depend on v as long as $\|v\|_{\mathcal{Y}_u} \leq 1$.

As mentioned before, a combination of (4.5) and (4.4), cf. Lemma 4.2, implies that

$$\|(\partial\rho_\Lambda)v - (\partial\rho)v\|_{\mathcal{X}_{\Lambda'}} \rightarrow 0 \quad \text{as } |\Lambda| \rightarrow \infty,$$

uniformly for all $v \in \mathcal{V}_u$ with $\|v\|_{\mathcal{Y}_u} \leq 1$. \square

5. Explicit computation of the derivatives of the singlet and pair distribution functions. In the sequel we are going to determine more suitable and implementable formulae for the derivatives of the first two molecular distribution functions in a finite size box $\Lambda \subset \mathbb{R}^3$.

For a given $v \in \mathcal{V}_u$ with $\|v\|_{\mathcal{Y}_u} \leq t_0$ we write

$$V_N(\mathbf{R}_N) = \sum_{1 \leq i < j \leq N} v(|R_i - R_j|) \tag{5.1}$$

in analogy to (2.6). Then, to begin with, a straightforward formal computation provides the derivative $\partial \Xi_\Lambda$ of the grand canonical partition function (2.8):

$$(\partial \Xi_\Lambda)v = -\beta \sum_{N=2}^{\infty} \frac{z^N}{N!} \int_{\Lambda^N} V_N(\mathbf{R}_N) e^{-\beta U_N(\mathbf{R}_N)} d\mathbf{R}_N.$$

That $\partial \Xi_\Lambda$ is a Fréchet derivative in $\mathcal{L}(\mathcal{V}_u, \mathbb{R})$ can readily be checked by following the line of argument of the proof of Lemma 3.2. Since V_N is defined by a pairwise interaction of identical particles we can use the definition of the pair distribution function to rewrite this derivative in the simpler form

$$(\partial \Xi_\Lambda)v = -\frac{\beta}{2} \int_{\Lambda} \int_{\Lambda} v(|R - R'|) \rho_{\Lambda}^{(2)}(R, R') dR dR',$$

which is amenable to numerical computations.

The argument of Lemma 3.2 can also be used to determine the Fréchet derivatives $\partial \Xi_{\Lambda}^{(m)} \in \mathcal{L}(\mathcal{V}_u, L^\infty(\Lambda^m))$ of the numerators

$$\Xi_{\Lambda}^{(m)}(\mathbf{R}_m) = \sum_{N=m}^{\infty} \frac{z^N}{(N-m)!} \int_{\Lambda^{N-m}} e^{-\beta U_N(\mathbf{R}_N)} d\mathbf{R}_{m,N}$$

of the molecular distribution functions $\rho_{\Lambda}^{(m)}$, namely

$$((\partial \Xi_{\Lambda}^{(m)})v)(\mathbf{R}_m) = -\beta \sum_{N=m}^{\infty} \frac{z^N}{(N-m)!} \int_{\Lambda^{N-m}} V_N(\mathbf{R}_N) e^{-\beta U_N(\mathbf{R}_N)} d\mathbf{R}_{m,N}.$$

For $m = 1$ we can utilize (5.1) and the fact that individual particles are indistinguishable to reformulate

$$\begin{aligned} ((\partial \Xi_{\Lambda}^{(1)})v)(R_1) &= -\beta \sum_{N=2}^{\infty} \frac{z^N}{(N-1)!} \int_{\Lambda^{N-1}} \sum_{1 \leq i < j \leq N} v(|R_i - R_j|) e^{-\beta U_N(\mathbf{R}_N)} d\mathbf{R}_{1,N} \\ &= -\beta \sum_{N=2}^{\infty} \frac{z^N}{(N-1)!} \left((N-1) \int_{\Lambda} v(|R_1 - R_2|) \int_{\Lambda^{N-2}} e^{-\beta U_N(\mathbf{R}_N)} d\mathbf{R}_{2,N} dR_2 \right. \\ &\quad \left. + \sum_{2 \leq i < j \leq N} \int_{\Lambda^{N-1}} v(|R_i - R_j|) e^{-\beta U_N(\mathbf{R}_N)} d\mathbf{R}_{1,N} \right) \\ &= -\beta \sum_{N=2}^{\infty} \frac{z^N}{(N-2)!} \int_{\Lambda} v(|R_1 - R_2|) \int_{\Lambda^{N-2}} e^{-\beta U_N(\mathbf{R}_N)} d\mathbf{R}_{2,N} dR_2 \\ &\quad - \frac{\beta}{2} \sum_{N=3}^{\infty} \frac{z^N}{(N-3)!} \int_{\Lambda} \int_{\Lambda} v(|R_2 - R_3|) \int_{\Lambda^{N-3}} e^{-\beta U_N(\mathbf{R}_N)} d\mathbf{R}_{3,N} dR_3 dR_2 \\ &= -\beta \int_{\Lambda} v(|R_1 - R_2|) \Xi_{\Lambda}^{(2)}(\mathbf{R}_2) dR_2 - \frac{\beta}{2} \int_{\Lambda} \int_{\Lambda} v(|R_2 - R_3|) \Xi_{\Lambda}^{(3)}(\mathbf{R}_3) dR_3 dR_2. \end{aligned}$$

Likewise we obtain

$$\begin{aligned}
 ((\partial \Xi_\Lambda^{(2)})v)(R_1, R_2) &= -\beta \sum_{N=2}^{\infty} \frac{z^N}{(N-2)!} \int_{\Lambda^{N-2}} V_N(\mathbf{R}_N) e^{-\beta U_N(\mathbf{R}_N)} d\mathbf{R}_{2,N} \\
 &= -\beta \sum_{N=2}^{\infty} \frac{z^N}{(N-2)!} \left(v(|R_1 - R_2|) \int_{\Lambda^{N-2}} e^{-\beta U_N(\mathbf{R}_N)} d\mathbf{R}_{2,N} \right. \\
 &\quad + (N-2) \int_{\Lambda} v(|R_1 - R_3|) \int_{\Lambda^{N-3}} e^{-\beta U_N(\mathbf{R}_N)} d\mathbf{R}_{3,N} dR_3 \\
 &\quad + (N-2) \int_{\Lambda} v(|R_2 - R_3|) \int_{\Lambda^{N-3}} e^{-\beta U_N(\mathbf{R}_N)} d\mathbf{R}_{3,N} dR_3 \\
 &\quad \left. + \frac{(N-2)(N-3)}{2} \int_{\Lambda} \int_{\Lambda} v(|R_3 - R_4|) \int_{\Lambda^{N-4}} e^{-\beta U_N(\mathbf{R}_N)} d\mathbf{R}_{4,N} dR_4 dR_3 \right),
 \end{aligned}$$

which can be rewritten as

$$\begin{aligned}
 ((\partial \Xi_\Lambda^{(2)})v)(R_1, R_2) &= -\beta v(|R_1 - R_2|) \Xi_\Lambda^{(2)}(\mathbf{R}_2) \\
 &\quad - \beta \int_{\Lambda} v(|R_1 - R_3|) \Xi_\Lambda^{(3)}(\mathbf{R}_3) dR_3 - \beta \int_{\Lambda} v(|R_2 - R_3|) \Xi_\Lambda^{(3)}(\mathbf{R}_3) dR_3 \\
 &\quad - \frac{\beta}{2} \int_{\Lambda} \int_{\Lambda} v(|R_3 - R_4|) \Xi_\Lambda^{(4)}(\mathbf{R}_4) dR_4 dR_3.
 \end{aligned}$$

After these preparations we can employ the quotient rule to obtain

$$\begin{aligned}
 ((\partial \rho_\Lambda^{(1)})v)(R_1) &= \frac{1}{\Xi_\Lambda} ((\partial \Xi_\Lambda^{(1)})v)(R_1) - \rho_\Lambda^{(1)}(R_1) \frac{(\partial \Xi_\Lambda)v}{\Xi_\Lambda} \\
 &= -\beta \int_{\Lambda} v(|R_1 - R'|) \rho_\Lambda^{(2)}(R_1, R') dR' \\
 &\quad - \frac{\beta}{2} \int_{\Lambda} \int_{\Lambda} v(|R'_1 - R'_2|) \rho_\Lambda^{(3)}(R_1, R'_1, R'_2) dR'_2 dR'_1 \\
 &\quad + \frac{\beta}{2} \int_{\Lambda} \int_{\Lambda} v(|R'_1 - R'_2|) \rho_\Lambda^{(1)}(R_1) \rho_\Lambda^{(2)}(R'_1, R'_2) dR'_2 dR'_1
 \end{aligned} \tag{5.2}$$

and

$$\begin{aligned}
((\partial\rho_\Lambda^{(2)})v)(R_1, R_2) &= \frac{1}{\Xi_\Lambda}((\partial\Xi_\Lambda^{(2)})v)(R_1, R_2) - \rho_\Lambda^{(2)}(R_1, R_2) \frac{(\partial\Xi_\Lambda)v}{\Xi_\Lambda} \\
&= -\beta v(|R_1 - R_2|)\rho_\Lambda^{(2)}(R_1, R_2) \\
&\quad - \beta \int_\Lambda v(|R_1 - R'|)\rho_\Lambda^{(3)}(R_1, R_2, R') \, dR' \tag{5.3a}
\end{aligned}$$

$$- \beta \int_\Lambda v(|R_2 - R'|)\rho_\Lambda^{(3)}(R_1, R_2, R') \, dR' \tag{5.3b}$$

$$- \beta \int_\Lambda \int_\Lambda \frac{1}{2} v(|R'_1 - R'_2|)\rho_\Lambda^{(4)}(R_1, R_2, R'_1, R'_2) \, dR'_1 dR'_2 \tag{5.3c}$$

$$+ \frac{\beta}{2} \int_\Lambda \int_\Lambda v(|R'_1 - R'_2|)\rho_\Lambda^{(2)}(R_1, R_2)\rho_\Lambda^{(2)}(R'_1, R'_2) \, dR'_1 dR'_2.$$

REMARK 5.1. To illustrate (5.3) imagine the situation of a fixed pair of coordinates $R_1, R_2 \in \Lambda$ with $R_1 \neq R_2$, when $v = v(r)$ is a delta distribution located in $r' > 0$. Then the three integrals in (5.3a)–(5.3c) provide the expected number of events – up to a factor $4\pi r'^2$ due to the polar coordinate transformation – of encountering at the same time a pair of particles at R_1 and R_2 and a different pair of particles with distance r' . The three individual terms account for events where

(5.3a): R_1 is one of the two members of the second pair of particles,

(5.3b): R_2 is one of the two members of the second pair of particles,

(5.3c): all four particles involved are different; during integration every second pair of particles is counted twice, hence the extra factor $1/2$. \diamond

It is obvious that neither of the two representations of $(\partial\rho_\Lambda^{(1)})v$ and $(\partial\rho_\Lambda^{(2)})v$ has a straightforward extension to the thermodynamical limit because the corresponding double integrals over the entire space will not converge. To extend these formulae to the thermodynamical limit these integrals need to be recombined. This will be reconsidered in part II of this work [2].

6. Conclusions. We have shown that for a grand canonical ensemble of identical point-like particles in thermodynamical equilibrium the molecular distribution functions $\rho_\Lambda^{(m)}$ and their thermodynamical limits $\rho^{(m)}$ are differentiable with respect to the underlying pair potential and the L^∞ norm of the distribution functions. To do so we have considered pair potentials that satisfy Assumption A which is slightly stronger than just being stable and regular; for these potentials we can treat the entire regime of small activities $z > 0$ for which the thermodynamical limit of the molecular distribution functions is known to be well-defined. In physical terms these activities correspond to a region without phase transitions, i.e., the *gas phase* of this molecular fluid, cf. [9].

Assumption A comes with a natural topology to study perturbations v of u : On the one hand perturbations must not be too strong to violate the repulsive nature of the potential when particles get close, hence, $|v|$ must be bounded by u near the origin; on the other hand, perturbations must be sufficiently small for distant particles to maintain regularity. We mention, though, that the choice of u^* allows rather general decay rates near infinity that may be different for the underlying potential u and for a perturbation v .

We have also proved that the derivative of $\rho_\Lambda^{(m)}$ converges compactly to the derivative of $\rho^{(m)}$ as $|\Lambda| \rightarrow \infty$. This justifies the approximate computation of, say, the derivative of the radial distribution function (an often-used structural quantity in chemical physics) by numerical particle simulations in a finite size box, as suggested in [6].

In a subsequent paper [2] we will reconsider the pair distribution function $\rho_\Lambda^{(2)}$ and the corresponding radial distribution function $g = g(r)$ because they call for the investigation of differentiability in another function space that reflects the property that $g(r) \rightarrow 1$ as $r \rightarrow \infty$ for potentials u that satisfy Assumption A. The methods that come to use in [2] employ cluster expansions and are of completely different nature than the ones that have been utilized here.

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