FRÉCHET DIFFERENTIABILITY OF MOLECULAR DISTRIBUTION FUNCTIONS II. THE URSELL FUNCTION

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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 thermodynamical limit of the Ursell or pair correlation function. For certain admissible perturbations of the pair potential and sufficiently small activity we rigorously establish Frechet differentiability of the Ursell function in the L^1 norm.

Furthermore, concerning the thermodynamical limit of the pair distribution function we explicitly compute its Fréchet derivative as a sum of a multiplication operator and an integral operator.

Key words. Statistical mechanics, cluster expansion, molecular distribution function, Ursell function, radial distribution function, Fréchet derivative

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1. Introduction. We study a continuous system of identical classical particles in a grand canonical ensemble, where the potential energy is determined by a pair potential which only depends on the distance of the interacting particles. In the first part of this work [3] we have shown that in the thermodynamical limit the corresponding equilibrium molecular distribution functions are differentiable in L^∞ with respect to the pair potential. It is well-known, however, that the correlations between individual observations of particles become small as the distance between the observation points gets large. For example, the so-called *pair correlation function* or (second order) *Ursell function*, which describes the correlations between the occurrence of particles at two different points in space is known to be close to zero (no correlation) for distant points, and the rate of decay is strong enough to guarantee that the thermodynamical limit of the pair correlation function is integrable over the entire space. We mention in passing that the pair correlation function is important for physical chemistry applications (cf., e.g., Rühle et al. [8]) because this is a measurable structural quantity that gives insight into the type of the underlying potential.

One may question whether the Fréchet derivative of the pair correlation function (with respect to the potential) maps also continuously into the space of integrable functions; this does not follow from the L^∞ analysis of the first part of this work and, in fact, it does not seem possible to prove this with the techniques utilized in [3]. We therefore use a different argument in this paper based on cluster expansions of the Ursell functions.

The same approach is subsequently used to derive integral operator representations of the Fréchet derivatives of the thermodynamical limits of the singlet and pair molecular distribution functions, which can easily be reassembled to obtain the derivative of the pair correlation function, when necessary. Among other applications such a representation may open a door to investigate invertibility of these derivatives.

The outline of this paper is as follows. In the following section we state the basic assumptions on the pair potential and its perturbations, and briefly review the main results from [3]. Then, in Section 3 we summarize classical results about cluster

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expansions of grand canonical quantities such as the molecular distribution functions and the Ursell functions; here we also recollect basic properties of the pair correlation function, like its integrability and its asymptotic behavior at infinity. Section 4 is devoted to upper bounds for certain higher order correlation functions; this section can be skipped by readers who are only interested in our main result on differentiability, stated and proved in Section 5 (Theorem 5.3). The estimates from Section 4 are revisited in Section 6, where they are utilized to justify the explicit computation of the thermodynamical limit representation of the Fréchet derivative of the pair distribution function.

2. Background. Let $\Lambda \subset \mathbb{R}^3$ be a bounded cubical box centered at the origin, and $R_i \in \Lambda$, $i = 1, 2, \ldots$, be the coordinates of the individual particles of a grand canonical ensemble in Λ. Repeatedly we use the notation

$$\boldsymbol{R}_N = (R_1, \ldots, R_N) \qquad \text{and} \qquad \boldsymbol{R}_{n,N} = (R_{n+1}, \ldots, R_N)$$

for the coordinates of (some of) the particles of the entire ensemble. When the system is in thermal equilibrium the m particle distribution function given by

$$\rho_\Lambda^{(m)}(\boldsymbol{R}_m) \;=\; \frac{1}{\Xi_\Lambda} \sum_{N=m}^{\infty} \frac{z^N}{(N-m)!} \int_{\Lambda^{N-m}} e^{-\beta U_N(\boldsymbol{R}_N)} \, \mathrm{d}\boldsymbol{R}_{m,N} \tag{2.1}$$

describes – up to proper normalization – the probability density of observing m particles simultaneously at the coordinates $R_1, R_2, \ldots, R_m \in \Lambda$. In (2.1)

$$U_N(R_1, \ldots, R_N) \;=\; \sum_{1 \le i < j \le N} u(|R_i - R_j|) \tag{2.2}$$

is the potential energy of a configuration of an N particle system, assuming that the interactions between the particles can be described by a pair potential and that these interactions only depend on their mutual distances. Furthermore, in (2.1) $\beta > 0$ is the inverse temperature, $z > 0$ is the *activity*, and

$$\Xi_\Lambda \;=\; \sum_{N=0}^{\infty} \frac{z^N}{N!} \int_{\Lambda^N} e^{-\beta U_N(\boldsymbol{R}_N)} \, \mathrm{d}\boldsymbol{R}_N \tag{2.3}$$

is the associated *grand canonical partition function.*

Following [3] we declare the pair potential $u : \mathbb{R}^+ \to \mathbb{R}$ to satisfy the following assumption.

Assumption A. *There exists $s > 0$ and positive decreasing functions $u_*, u^* : \mathbb{R}^+ \to \mathbb{R}$ with*

$$\int_0^s u_*(r)\, r^2 \,\mathrm{d}r \;=\; \infty \qquad \text{and} \qquad \int_s^\infty u^*(r)\, r^2 \,\mathrm{d}r \;<\; \infty\,,$$

such that u satisfies

$$u(r) \;\ge\; u_*(r)\,, \quad r \le s\,, \qquad \text{and} \qquad |u(r)| \;\le\; u^*(r)\,, \quad r \ge s\,.$$

We also introduce the Banach space $\mathscr{V}_u$ of *perturbations* of u as the set of functions v for which the corresponding norm

$$\|v\|_{\mathscr{V}_u} \;=\; \max\{\, \|v/u\|_{(0,s)}, \|v/u^*\|_{(s,\infty)} \,\} \tag{2.4}$$

is finite*. With these prerequisites it has been shown in [3] that for any $0 < t_0 < 1$ the following three properties hold true for all perturbed potentials $\widetilde{u} = u+v$ with $v \in \mathscr{V}_u$, $\|v\|_{\mathscr{V}_u} \leq t_0$, the respective quantities being independent of the particular choice of v:

(i) there exists $B > 0$ such that

$$\sum_{1\leq i<j\leq N} \widetilde{u}(|R_i - R_j|) \,\geq\, -BN \tag{2.5}$$

for every configuration of N particles and every $N \in \mathbb{N}$, i.e., $\widetilde{u}$ is *stable*;

(ii) for every $m \in \mathbb{N}$ and $\boldsymbol{R}_m \in \Lambda^m$ there exists an index $j^*(\boldsymbol{R}_m)$ such that

$$\sum_{\substack{i=1\\ i\neq j^*}}^{m} \widetilde{u}(|R_i - R_{j^*}|) \,\geq\, -2B \tag{2.6}$$

with the same constant B as in (2.5);

(iii) there exists $c_\beta > 0$ with

$$4\pi \int_0^\infty |e^{-\beta\widetilde{u}(r)} - 1|\, r^2 \,\mathrm{d}r \,\leq\, c_\beta\,, \tag{2.7}$$

i.e., $\widetilde{u}$ is *regular*.

As a consequence it follows from classical results (cf., e.g., Ruelle [10]) that for any of these potentials the associated molecular distribution functions $\widetilde{\rho}_\Lambda^{(m)}$, $m \in \mathbb{N}$, converge compactly to a bounded limiting function $\widetilde{\rho}^{(m)}$ as the size of Λ grows to infinity, provided that the activity is sufficiently small, i.e., that

$$0 \,<\, z \,<\, \frac{1}{c_\beta e^{2\beta B+1}}\,. \tag{2.8}$$

This is known as the *thermodynamical limit*.

In [3] it has further been shown that the molecular distribution functions $\rho_\Lambda^{(m)}$ as well as their thermodynamical limits have Fréchet derivatives

$$\partial\rho_\Lambda^{(m)} \in \mathscr{L}(\mathscr{V}_u, \mathscr{L}^\infty(\Lambda^m)) \qquad \text{and} \qquad \partial\rho^{(m)} \in \mathscr{L}(\mathscr{V}_u, \mathscr{L}^\infty((\mathbb{R}^3)^m))$$

with respect to u, and for a given $v \in \mathscr{V}_u$ the directional derivatives $\bigl(\partial\rho_\Lambda^{(m)}\bigr)v$ converge compactly to $\bigl(\partial\rho^{(m)}\bigr)v$ in the thermodynamical limit, uniformly for $\|v\|_{\mathscr{V}_u} \leq 1$.

In this paper we focus on the pair correlation function

$$\omega_\Lambda^{(2)}(R_1,R_2) \,=\, \rho_\Lambda^{(2)}(R_1,R_2) \,-\, \rho_\Lambda^{(1)}(R_1)\rho_\Lambda^{(1)}(R_2)\,. \tag{2.9}$$

In the thermodynamical limit the pair correlation function converges (compactly) to a function $\omega^{(2)}$ that only depends on $|R_2 - R_1|$, and which is related to the so called *radial distribution function* g via

$$g(r) \,=\, 1 \,+\, \frac{1}{\rho_0^2}\,\omega^{(2)}(R,0)\,, \qquad |R| = r\,,$$

where $\rho_0 = \lim_{|\Lambda|\to\infty} \rho_\Lambda^{(1)}$ is the (constant) *counting density* of the system.

*If $\Omega \subset \mathbb{R}^d$ is a domain then $\|\cdot\|_\Omega$ denotes the supremum norm over Ω.

From our aforementioned results it follows that $\omega^{(2)}(R,0)$ as a function of $R \in \mathbb{R}^3$ is Fréchet differentiable in $\mathscr{L}(\mathscr{V}_u, L^\infty(\mathbb{R}^3))$. However, this function is also known to belong to $L^1(\mathbb{R}^3)$, cf. [10], and to converge to zero for $|R| \to \infty$ under mild additional assumptions on u, cf. [6, 1, 2, 7] and Section 3 below. This decay at infinity is not taken into account when studying the distribution functions in L^∞ as has been done in [3]; therefore the purpose of this paper is to extend our results and to prove that $\omega^{(2)}(\,\cdot\,,0)$ has a Fréchet derivative $\partial\omega^{(2)} \in \mathscr{L}(\mathscr{V}_u, L^1(\mathbb{R}^3))$.

Our method of proof utilizes classical graph theoretical cluster expansions that have been developed in the aforementioned papers to derive appropriate bounds for the pair correlation function. We will summarize the corresponding ingredients in the following section.

3. Cluster expansions. A graph $\mathcal{G}$ is a set of (undirected) bonds (i,j) between labeled vertices, where $(i,j) \in \mathcal{G}$ means that there is a bond connecting vertices $\#i$ and $\#j$. In our applications vertex $\#i$ corresponds to the ith particle of the grand canonical ensemble and its coordinates $R_i \in \Lambda$; a bond $(i,j) \in \mathcal{G}$ is associated with a certain interaction of the corresponding two particles, either given by

$$f_{ij} \,=\, f(R_i - R_j)\,, \qquad R_i, R_j \in \mathbb{R}^3\,, \tag{3.1}$$

where

$$f(R) \,=\, e^{-\beta u(|R|)} - 1 \tag{3.2}$$

is the so-called *Mayer f-function*, or by the absolute values of f_{ij}. We refer to Stell [11] as a general reference and for a detailed exposition of graph theory in statistical mechanics.

For our results three types of graphs are relevant. First comes the set of *connected graphs*: in a connected graph every pair of vertices has a connecting path of bonds between them. Connected graphs can be used to specify the sequence $(\omega_\Lambda^{(m)})_{m\geq 1}$ of Ursell functions, i.e.,

$$\omega_\Lambda^{(m)}(\boldsymbol{R}_m) \,=\, \sum_{N=m}^\infty \frac{z^N}{(N-m)!} \sum_{\mathcal{C}_N} \int_{\Lambda^{N-m}} \Bigl(\prod_{(i,j)\in\mathcal{C}_N} f_{ij} \Bigr) \,\mathrm{d}\boldsymbol{R}_{m,N}\,, \tag{3.3}$$

where the sum varies over all connected graphs $\mathcal{C}_N$ with N vertices labeled $\#1$ through $\#N$. The second Ursell function has already been introduced in (2.9) and the first one can be shown to coincide with $\rho_\Lambda^{(1)}$, compare (3.6) below; further examples that we need later on are

$$\begin{aligned} \omega_\Lambda^{(3)}(R_1,R_2,R_3) \,=\, &\rho_\Lambda^{(3)}(R_1,R_2,R_3) \,-\, \rho_\Lambda^{(2)}(R_2,R_3)\rho_\Lambda^{(1)}(R_1) \\ &- \omega_\Lambda^{(2)}(R_1,R_3)\rho_\Lambda^{(1)}(R_2) \,-\, \omega_\Lambda^{(2)}(R_1,R_2)\rho_\Lambda^{(1)}(R_3) \end{aligned} \tag{3.4}$$

and

$$\begin{aligned} \omega_\Lambda^{(4)}(R_1,R_2,R_3,R_4) \,=\, &\rho_\Lambda^{(4)}(R_1,R_2,R_3,R_4) \,-\, \rho_\Lambda^{(2)}(R_1,R_2)\rho_\Lambda^{(2)}(R_3,R_4) \\ &- \omega_\Lambda^{(3)}(R_1,R_2,R_3)\rho_\Lambda^{(1)}(R_4) \,-\, \omega_\Lambda^{(2)}(R_1,R_4)\omega_\Lambda^{(2)}(R_2,R_3) \\ &- \omega_\Lambda^{(3)}(R_1,R_2,R_4)\rho_\Lambda^{(1)}(R_3) \,-\, \omega_\Lambda^{(2)}(R_1,R_3)\omega_\Lambda^{(2)}(R_2,R_4) \\ &- \rho_\Lambda^{(3)}(R_1,R_3,R_4)\rho_\Lambda^{(1)}(R_2) \,-\, \rho_\Lambda^{(3)}(R_2,R_3,R_4)\rho_\Lambda^{(1)}(R_1) \\ &+ 2\rho_\Lambda^{(2)}(R_3,R_4)\rho_\Lambda^{(1)}(R_1)\rho_\Lambda^{(1)}(R_2)\,. \end{aligned} \tag{3.5}$$

This last representation may not be the simplest one, but it is the one that we will exploit below.

To introduce a second set of graphs let $\mathcal{I}, \mathcal{J} \subset \mathbb{N}$ be two disjoint finite sets of vertex labels with cardinalities $|\mathcal{I}| \geq 1$ and $|\mathcal{J}| \geq 0$. We define $\mathfrak{Z}_{\mathcal{I},\mathcal{J}}$ as the set of graphs with vertices given by $\mathcal{I} \cup \mathcal{J}$, out of which those in $\mathcal{I}$ are "highlighted" – being white as opposed to black, say – and where each black vertex has a connecting path of bonds to one of the white vertices. These graphs occur in the expansion

$$\rho_\Lambda^{(m)}(\boldsymbol{R}_m) \,=\, \sum_{N=m}^{\infty} \frac{z^N}{(N-m)!} \sum_{\mathcal{Z}_{N,m}} \int_{\Lambda^{N-m}} \Bigl(\prod_{(i,j)\in \mathcal{Z}_{N,m}} f_{ij} \Bigr) \,\mathrm{d}\boldsymbol{R}_{m,N} \tag{3.6}$$

of the molecular distribution functions, where $\mathcal{Z}_{N,m}$ varies over all graphs in $\mathfrak{Z}_{\mathcal{I}_m,\mathcal{J}_{m,N}}$ with $\mathcal{I}_m = \{1,\dots,m\}$ and $\mathcal{J}_{m,N} = \{m+1,\dots,N\}$.

A special case of the latter graphs are *trees* and *forests*. A tree is a connected graph with a single white vertex, its *root*, such that between each pair of vertices there is one and only one connecting path. A union of trees is a forest; the set of forests whose constituent trees have the same roots $\mathcal{I}$ and the same black vertices $\mathcal{J}$ is denoted by $\mathfrak{F}_{\mathcal{I},\mathcal{J}} \subset \mathfrak{Z}_{\mathcal{I},\mathcal{J}}$.

Concerning trees we quote the following well-known result (for a proof, cf., e.g., Penrose [6]), which will be referred to later on:

LEMMA 3.1. *Let u be a stable and regular pair potential and denote by $\mathfrak{T}_N$ the set of trees with N vertices labeled #1 through #N. If z satisfies (2.8) then the series*

$$\tau_\Lambda^{(1)}(R_1) \,=\, \sum_{N=1}^{\infty} \frac{(ze^{2\beta B})^N}{(N-1)!} \sum_{\mathcal{T}_N \in \mathfrak{T}_N} \int_{\Lambda^{N-1}} \Bigl(\prod_{(i,j)\in \mathcal{T}_N} |f_{ij}| \Bigr) \,\mathrm{d}\boldsymbol{R}_{1,N} \tag{3.7}$$

converges uniformly in Λ, and there holds

$$\|\tau_\Lambda^{(1)}\|_\Lambda \,\leq\, w \,:=\, -\frac{1}{c_\beta}\, W(-zc_\beta e^{2\beta B}) \,<\, \frac{1}{c_\beta}\,, \tag{3.8}$$

where W is the Lambert W-function, cf., e.g., [5]. Moreover,

$$\tau_\Lambda^{(2)}(R_1,R_2) \,=\, \sum_{N=2}^{\infty} \frac{(ze^{2\beta B})^N}{(N-2)!} \sum_{\mathcal{T}_N \in \mathfrak{T}_N} \int_{\Lambda^{N-2}} \Bigl(\prod_{(i,j)\in \mathcal{T}_N} |f_{ij}| \Bigr) \,\mathrm{d}\boldsymbol{R}_{2,N} \tag{3.9}$$

converges uniformly in Λ^2, and there holds

$$\tau_\Lambda^{(2)}(R_1,R_2) \,\leq\, G(R_1-R_2)\,, \qquad R_1,R_2 \in \Lambda\,,$$

where the nonnegative function $G : \mathbb{R}^3 \to \mathbb{R}$ is given by

$$G(R) \,=\, \int_{\mathbb{R}^3} e^{2\pi \mathrm{i}\xi\cdot R}\, \frac{w^2\,\widehat{|f|}(\xi)}{1 - w\,\widehat{|f|}(\xi)} \,\mathrm{d}\xi \tag{3.10}$$

and $\widehat{|f|}$ is the Fourier transform

$$\widehat{|f|}(\xi) \,=\, \int_{\mathbb{R}^3} e^{-2\pi \mathrm{i}\xi\cdot R}\, \bigl| e^{-\beta u(|R|)} - 1 \bigr| \,\mathrm{d}R$$

of the absolute values of the Mayer f-function (3.2).

For later convenience we list a few properties of the function G.

PROPOSITION 3.2. *Let the assumptions of Lemma 3.1 be satisfied. Then the function G of (3.10) is even, bounded, and integrable. Moreover, if $u(r) \to 0$ as $r \to \infty$ (e.,g., when u satisfies Assumption A) then $G(R) \to 0$ for $|R| \to \infty$.*

Proof. According to (3.10) there holds $G(-R) = \overline{G(R)}$, and since this is a nonnegative real number, G must be an even function.

Since $|f|$ is bounded by $e^{2\beta B}$ and belongs to $L^1(\mathbb{R})$, cf. (2.5) and (2.7), respectively, it follows that $f \in L^2(\mathbb{R}^3)$, and then $\widehat{|f|} \in L^2(\mathbb{R}^3)$, too. Therefore, rewriting (3.10) as

$$\widehat{G} \,=\, w^2\,\widehat{|f|} \,+\, \frac{w^3}{1 - w\,\widehat{|f|}}\,\widehat{|f|}^{\,2}$$

we conclude that the second term on the right-hand side belongs to $L^1(\mathbb{R}^3)$ because its numerator is bounded away from zero according to (3.8), and taking the inverse Fourier transform we obtain

$$G - w^2|f| \in C_0(\mathbb{R}^3)\,.$$

From this it follows that G is bounded, and if u vanishes at infinity then so does G.

Finally, it follows from (3.10) and the nonnegativity of G that

$$\int_{\mathbb{R}^3} \bigl|G(R)\bigr|\,\mathrm{d}R \,=\, \widehat{G}(0) \,=\, \frac{w^2\,\widehat{|f|}(0)}{1 - w\,\widehat{|f|}(0)} \,<\, \infty\,,$$

i.e., $G \in L^1(\mathbb{R}^3)$. □

Let $\mathcal{I}, \mathcal{J} \subset \mathbb{N}$ with $\mathcal{I} \cap \mathcal{J} = \emptyset$ be given; furthermore, denote by $\boldsymbol{R}_\mathcal{I}$ and $\boldsymbol{R}_\mathcal{J}$ the coordinates of the particles with labels in $\mathcal{I}$ and $\mathcal{J}$, respectively. Ruelle [9, 10] considered the functions

$$\varphi_{\mathcal{I},\mathcal{J}}(\boldsymbol{R}_\mathcal{I};\boldsymbol{R}_\mathcal{J}) \,=\, \sum_{\mathcal{Z}_{\mathcal{I},\mathcal{J}}} \Bigl(\,\prod_{(i,j)\in\mathcal{Z}_{\mathcal{I},\mathcal{J}}} f_{ij}\Bigr)\,, \tag{3.11}$$

where $\mathcal{Z}_{\mathcal{I},\mathcal{J}}$ varies over all graphs in $\mathfrak{Z}_{\mathcal{I},\mathcal{J}}$; we take $\mathfrak{Z}_{\emptyset,\mathcal{J}}$ to be the empty set and $\varphi_{\emptyset,\mathcal{J}}$ to be zero. Take note that the order of the particles in $\mathcal{I}$ and $\mathcal{J}$ does not affect the value of the right-hand side of (3.11).

Given $i^* = i^*(\mathcal{I},\boldsymbol{R}_\mathcal{I}) \in \mathcal{I}$, and eliminating vertex $\#i^*$ from all graphs in $\mathcal{Z}_{\mathcal{I},\mathcal{J}}$, Ruelle derived the recursion

$$\varphi_{\mathcal{I},\mathcal{J}}(\boldsymbol{R}_\mathcal{I};\boldsymbol{R}_\mathcal{J}) \,=\, d_{\mathcal{I},i^*}(\boldsymbol{R}_\mathcal{I}) \sum_{\mathcal{K}\subset\mathcal{J}} k_\mathcal{K}(R_{i^*};\boldsymbol{R}_\mathcal{K})\varphi_{\mathcal{I}\cup\mathcal{K}\setminus\{i^*\},\mathcal{J}\setminus\mathcal{K}}(\boldsymbol{R}_{\mathcal{I}\cup\mathcal{K}\setminus\{i^*\}};\boldsymbol{R}_{\mathcal{J}\setminus\mathcal{K}})\,,$$

where

$$d_{\mathcal{I},i^*}(\boldsymbol{R}_\mathcal{I}) \,=\, \prod_{i\in\mathcal{I}\setminus\{i^*\}} e^{-\beta u(|R_i - R_{i^*}|)} \tag{3.12}$$

and

$$k_\mathcal{K}(R;\boldsymbol{R}_\mathcal{K}) \,=\, \prod_{j\in\mathcal{K}} f(R_j - R)\,. \tag{3.13}$$

We emphasize that the particular elements of the family of φ-functions that enter into this recursion depend on the actual values of the input coordinates $\boldsymbol{R}_{\mathcal{I}}$ because we explicitly allow i^* to depend on $\boldsymbol{R}_{\mathcal{I}}$; aside of that the listing of the variables $\boldsymbol{R}_{\mathcal{I}}$ and $\boldsymbol{R}_{\mathcal{J}}$ is redundant here and below, because it's always the coordinates of the particles associated with the two indices of φ that are used as corresponding arguments. We therefore simplify our notation and write $\varphi(\mathcal{I};\mathcal{J})$ instead of $\varphi_{\mathcal{I},\mathcal{J}}(\boldsymbol{R}_{\mathcal{I}};\boldsymbol{R}_{\mathcal{J}})$ in the remainder of this work; similarly we will write $d_{\mathcal{I},i^*}$ and $k_{\mathcal{K}}(R)$ for the left-hand sides of (3.12) and (3.13), respectively. The above recursion thus takes the form

$$\varphi(\mathcal{I};\mathcal{J}) \,=\, d_{\mathcal{I},i^*} \sum_{\mathcal{K}\subset\mathcal{J}} k_{\mathcal{K}}(R_{i^*})\varphi(\mathcal{I}\cup\mathcal{K}\setminus\{i^*\};\mathcal{J}\setminus\mathcal{K})\,. \tag{3.14}$$

If we select $i^* = j^*(\mathcal{I},\boldsymbol{R}_{\mathcal{I}}) \in \mathcal{I}$ in accordance with (2.6) in such a way that

$$\sum_{i\in\mathcal{I}\setminus\{i^*\}} \widetilde{u}(|R_i - R_{j^*}|) \,\geq\, -2B \tag{3.15}$$

for every $\widetilde{u} = u + v$ with $\|v\|_{\mathscr{V}_u} \leq t_0$ then it follows from (3.14) that

$$\bigl|\varphi(\mathcal{I};\mathcal{J})\bigr| \,\leq\, e^{2\beta B} \sum_{\mathcal{K}\subset\mathcal{J}} \bigl|k_{\mathcal{K}}(R_{j^*})\bigr|\bigl|\varphi(\mathcal{I}\cup\mathcal{K}\setminus\{j^*\};\mathcal{J}\setminus\mathcal{K})\bigr|\,, \tag{3.16}$$

and by induction Ruelle concluded that

$$\int_{\Lambda^{|\mathcal{J}|}} \bigl|\varphi(\mathcal{I};\mathcal{J})\bigr| \,\mathrm{d}\boldsymbol{R}_{\mathcal{J}} \,\leq\, (|\mathcal{J}|)!\, c_\beta^{|\mathcal{J}|} \bigl(e^{2\beta B+1}\bigr)^{|\mathcal{I}\cup\mathcal{J}|-1}\,, \tag{3.17}$$

with the left-hand side to be interpreted as $\bigl|\varphi(\mathcal{I};\emptyset)\bigr|$ when $\mathcal{J}=\emptyset$. We also note that the functions

$$\psi(\mathcal{I};\mathcal{J}) \,=\, e^{2(N-1)\beta B} \sum_{\mathcal{F}\in\mathfrak{F}_{\mathcal{I},\mathcal{J}}} \Bigl(\prod_{(i,j)\in\mathcal{F}} |f_{ij}|\Bigr)\,, \qquad N = |\mathcal{I}\cup\mathcal{J}|\,, \tag{3.18}$$

satisfy the recursion (3.16) with *equality*, i.e.,

$$\psi(\mathcal{I};\mathcal{J}) \,=\, e^{2\beta B} \sum_{\mathcal{K}\subset\mathcal{J}} \bigl|k_{\mathcal{K}}(R_{j^*})\bigr|\psi(\mathcal{I}\cup\mathcal{K}\setminus\{j^*\};\mathcal{J}\setminus\mathcal{K})\,. \tag{3.19}$$

Following Poghosyan and Ueltschi [7] one can use (3.19) to prove a so-called *tree-graph inequality*†, namely

$$\bigl|\varphi(\mathcal{I};\mathcal{J})\bigr| \,\leq\, \psi(\mathcal{I};\mathcal{J})\,, \qquad\qquad |\mathcal{I}| \geq 2\,, \tag{3.20a}$$

and

$$\bigl|\varphi(\mathcal{I};\mathcal{J})\bigr| \,\leq\, e^{-2\beta B}\psi(\mathcal{I};\mathcal{J})\,, \qquad |\mathcal{I}| = 1\,. \tag{3.20b}$$

The set $\mathfrak{C}_N$ of connected graphs with vertices labeled #1 to #N and the set $\mathfrak{T}_N$ of trees with the same vertices agree – up to the color of their vertices – with the two

†In fact, the respective bounds (3.20) in [7] are off by $e^{2\beta B}$ for $|\mathcal{I}| \geq 2$ and $e^{4\beta B}$ for $|\mathcal{I}| = 1$; this can be fixed by a more careful initialization of the corresponding inductive argument.

sets $\mathfrak{Z}_{\{1\},\mathcal{J}_{1,N}}$ and $\mathfrak{F}_{\{1\},\mathcal{J}_{1,N}}$, respectively, with $\mathcal{J}_{1,N} = \{2,\dots,N\}$. It therefore follows from (3.11), (3.20b), and (3.18) that

$$\begin{aligned}\Bigl|\sum_{\mathcal{C}_N\in\mathfrak{C}_N}\int_{\Lambda^{N-2}}\Bigl(\prod_{(i,j)\in\mathcal{C}_N} f_{ij}\Bigr)\,\mathrm{d}\boldsymbol{R}_{2,N}\Bigr| &= \Bigl|\int_{\Lambda^{N-2}}\varphi(\{1\};\{2,\dots,N\})\,\mathrm{d}\boldsymbol{R}_{2,N}\Bigr|\\ &\le e^{2(N-2)\beta B}\sum_{\mathcal{T}_N\in\mathfrak{T}_N}\int_{\Lambda^{N-2}}\Bigl(\prod_{(i,j)\in\mathcal{T}_N}|f_{ij}|\Bigr)\,\mathrm{d}\boldsymbol{R}_{2,N}\,,\end{aligned}$$

and hence, cf. (3.3) and Lemma 3.1,

$$\begin{aligned}\bigl|\omega^{(2)}_\Lambda(R_1,R_2)\bigr| &\le e^{-4\beta B}\sum_{N=2}^\infty\frac{(ze^{2\beta B})^N}{(N-2)!}\sum_{\mathcal{T}_N\in\mathfrak{T}_N}\int_{\Lambda^{N-2}}\Bigl(\prod_{(i,j)\in\mathcal{T}_N}|f_{ij}|\Bigr)\,\mathrm{d}\boldsymbol{R}_{2,N}\\ &\le e^{-4\beta B}G(R_1-R_2)\,.\end{aligned}\tag{3.21}$$

Since the final upper bound in (3.21) does not depend on Λ we can turn to the thermodynamical limit to conclude that

$$\bigl|\omega^{(2)}(R,0)\bigr| \le e^{-4\beta B}G(R)\,.$$

Accordingly, it follows from Proposition 3.2 that $\omega^{(2)}(\,\cdot\,,0)\in L^1(\mathbb{R}^3)$, and that $\omega^{(2)}(R_1,R_2)\to 0$ as $|R_1-R_2|\to\infty$ if u satisfies Assumption A.

4. Higher order correlation functions. In this section we provide similar estimates for the higher order correlation functions

$$\chi^{(3)}_\Lambda(R_1,R_2,R_3) = \rho^{(3)}_\Lambda(R_1,R_2,R_3) - \rho^{(1)}_\Lambda(R_1)\rho^{(2)}_\Lambda(R_2,R_3)\tag{4.1}$$

and

$$\chi^{(4)}_\Lambda(R_1,R_2,R_3,R_4) = \rho^{(4)}_\Lambda(R_1,R_2,R_3,R_4) - \rho^{(2)}_\Lambda(R_1,R_2)\rho^{(2)}_\Lambda(R_3,R_4)\,,\tag{4.2}$$

which we will need in Section 6. To this end we introduce another set of graphs: For $N\ge 2$ let $\mathfrak{Z}^\times_N$ be the set of graphs in $\mathfrak{Z}_{\mathcal{I}_2,\mathcal{J}_{2,N}}$, where $\mathcal{I}_2=\{1,2\}$ and $\mathcal{J}_{2,N} = \{3,\dots,N\}$, that have no connecting edge between vertices #1 and #2. Moreover, let

$$\varphi^\times_N(\boldsymbol{R}_N) = \sum_{\mathcal{Z}^\times_N\in\mathfrak{Z}^\times_N}\Bigl(\prod_{(i,j)\in\mathcal{Z}^\times_N} f_{ij}\Bigr)\tag{4.3}$$

and

$$\zeta^{(m)}(\boldsymbol{R}_m) = \sum_{N=m}^\infty\frac{z^N}{(N-m)!}\int_{\Lambda^{N-m}}\varphi^\times_N(\boldsymbol{R}_N)\,\mathrm{d}\boldsymbol{R}_{m,N}\,.\tag{4.4}$$

Lemma 4.1. *Let u be stable and regular, and let z satisfy (2.8). Then there holds*

$$\bigl|\zeta^{(3)}(R_1,R_2,R_3)\bigr| \le we^{-4\beta B}\bigl(G(R_1-R_3)+G(R_2-R_3)\bigr)\,,$$

where the constant w and the function G are defined in Lemma 3.1.

Proof. For a given value of $N \geq 2$ we adopt Ruelle's method mentioned in the previous section and eliminate vertex #1 from each of the graphs in $\mathfrak{Z}_N^\times$ to obtain the identity

$$\varphi_N^\times(\boldsymbol{R}_N) \,=\, \sum_{\mathcal{K}\subset\mathcal{J}_{2,N}} k_{\mathcal{K}}(R_1)\varphi(\mathcal{K}\cup\{2\};\mathcal{J}_{2,N}\setminus\mathcal{K})\,.$$

Note that the corresponding term $d_{\mathcal{I}_2,1}$ of (3.14) is missing here because all graphs in $\mathfrak{Z}_N^\times$ lack a connecting edge between vertices #1 and #2. Therefore, (3.20a) and (3.19) yield

$$\bigl|\varphi_N^\times(\boldsymbol{R}_N)\bigr| \,\leq\, \sum_{\mathcal{K}\subset\mathcal{J}_{2,N}} \bigl|k_{\mathcal{K}}(R_1)\bigr|\psi(\mathcal{K}\cup\{2\};\mathcal{J}_{2,N}\setminus\mathcal{K}) \,=\, e^{-2\beta B}\psi(\mathcal{I}_2;\mathcal{J}_{2,N})\,, \qquad (4.5)$$

and inserting this inequality into (4.4) we arrive at

$$\bigl|\zeta^{(3)}(R_1,R_2,R_3)\bigr| \,\leq\, e^{-2\beta B}\sum_{N=3}^\infty \frac{z^N}{(N-3)!}\int_{\Lambda^{N-3}}\psi(\mathcal{I}_2;\mathcal{J}_{2,N})\,\mathrm{d}\boldsymbol{R}_{3,N}\,.$$

From the definition (3.18) of $\psi(\mathcal{I}_2;\mathcal{J}_{2,N})$ we therefore conclude that

$$\bigl|\zeta^{(3)}(R_1,R_2,R_3)\bigr| \,\leq\, e^{-4\beta B}(S_1^{(3)}+S_2^{(3)}) \qquad (4.6)$$

with

$$S_l^{(3)} \,=\, \sum_{N=3}^\infty \frac{(ze^{2\beta B})^N}{(N-3)!}\sum_{\mathcal{F}_l}\int_{\Lambda^{N-3}}\Bigl(\prod_{(i,j)\in\mathcal{F}_l}|f_{ij}|\Bigr)\,\mathrm{d}\boldsymbol{R}_{3,N}\,, \qquad l=1,2\,,$$

where the inner sum varies over all those forests $\mathcal{F}_l \in \mathfrak{F}_{\mathcal{I}_2,\mathcal{J}_{2,N}}$, for which the vertices #3 and #l belong to the same tree.

The forests that occur in $S_1^{(3)}$ consist of all possible combinations of one tree involving vertices #1 and #3 and another tree rooted in vertex #2; hence, we can use Lemma 3.1 and classical graph integral calculus, cf. [4, 11] to estimate

$$S_1^{(3)} \,\leq\, w\,G(R_1-R_3)\,.$$

Likewise we obtain

$$S_2^{(3)} \,\leq\, w\,G(R_2-R_3)\,.$$

Inserting this into (4.6) we thus obtain the assertion. ☐

Next we recall from (3.4) that

$$\chi_\Lambda^{(3)}(R_1,R_2,R_3) \,=\, \omega_\Lambda^{(3)}(R_1,R_2,R_3)+\omega_\Lambda^{(2)}(R_1,R_3)\rho_\Lambda^{(1)}(R_2)+\omega_\Lambda^{(2)}(R_1,R_2)\rho_\Lambda^{(1)}(R_3)\,.$$

From this and (3.3) we conclude that $\chi_\Lambda^{(3)}$ is a sum over all graphs with $N\geq 3$ vertices and the associated graph integrals over $\boldsymbol{R}_{3,N}\in\Lambda^{N-3}$, where the graphs are of either one of the following three types:

(i) a connected graph;

(ii) a graph with two connected components, one of which containing vertex #2 and the other one containing vertices #1 and #3;

(iii) a graph with two connected components, one of which containing vertex #3 and the other one containing vertices #1 and #2.

We use this observation to establish the following result.

Proposition 4.2. *If u is a stable and regular pair potential and the activity z satisfies (2.8) then*

$$\bigl|\chi_\Lambda^{(3)}(R_1,R_2,R_3)\bigr| \,\leq\, we^{-4\beta B}e^{-\beta u(|R_2-R_3|)}\bigl(G(R_2-R_1)+G(R_3-R_1)\bigr)$$

for all $R_1,R_2,R_3\in\Lambda$.

Proof. By its definition $\zeta^{(3)}(R_3,R_2,R_1)$ – note the different ordering of the arguments – is the sum over all graphs with $N\geq 3$ vertices and the associated graph integrals over $\boldsymbol{R}_{3,N}$, where in each graph every vertex is connected to vertex #2 or to vertex #3, but the latter two vertices have no connecting edge. Adding a bond between vertices #2 and #3 to any of these graphs therefore results in a connected graph. Accordingly, any graph occuring in the definition of $\zeta^{(3)}(R_3,R_2,R_1)$ belongs to the list (i)-(iii) above, and so does its counterpart with the additional bond.

Likewise, if $\mathcal{C}\in\mathfrak{C}_N$ with $N\geq 3$, and if one eliminates the edge between vertices #2 and #3 when present, then, still, every vertex has a connecting path to vertex #2 or to vertex #3. We therefore conclude that the graphs appearing in (4.1) consist of all those taken care of in $\zeta^{(3)}(R_3,R_2,R_1)$ and their counterparts with an additional bond between vertices #2 and #3. Thus it follows from the definition of the graph integrals and the distributive law that

$$\chi_\Lambda^{(3)}(R_1,R_2,R_3) \,=\, e^{-\beta u(|R_2-R_3|)}\zeta^{(3)}(R_3,R_2,R_1)\,.$$

Now the assertion follows from Lemma 4.1. □

Finally we turn to $\chi_\Lambda^{(4)}$ of (4.2). A straightforward computation based on (3.5) and (4.1) reveals that

$$\begin{aligned}\chi_\Lambda^{(4)}(R_1,R_2,R_3,R_4) \,&=\, \eta(R_1,R_2,R_3,R_4)\\ &\quad+\,\chi_\Lambda^{(3)}(R_1,R_3,R_4)\rho_\Lambda^{(1)}(R_2)\,+\,\chi_\Lambda^{(3)}(R_2,R_3,R_4)\rho_\Lambda^{(1)}(R_1)\,,\end{aligned}\tag{4.7}$$

where

$$\begin{aligned}\eta(R_1,R_2,R_3,R_4) \,&=\, \omega_\Lambda^{(4)}(R_1,R_2,R_3,R_4)\,+\,\omega_\Lambda^{(3)}(R_1,R_2,R_3)\rho_\Lambda^{(1)}(R_4)\\ &\qquad+\,\omega_\Lambda^{(2)}(R_1,R_4)\omega_\Lambda^{(2)}(R_2,R_3)\,+\,\omega_\Lambda^{(2)}(R_1,R_3)\omega_\Lambda^{(2)}(R_2,R_4)\\ &\qquad+\,\omega_\Lambda^{(3)}(R_1,R_2,R_4)\rho_\Lambda^{(1)}(R_3)\,.\end{aligned}$$

As above we observe that η is the sum of all graph integrals that correspond to connected graphs with $N\geq 4$ vertices having a connecting edge between vertices #3 and #4, and their counterparts which are obtained when deleting this very edge. The latter ones are the graphs from $\cup_{N\geq 4}\mathfrak{Z}_N^\times$ – up to the labeling of the two white vertices; therefore it follows as above that

$$\eta(R_1,R_2,R_3,R_4) \,=\, e^{-\beta u(|R_3-R_4|)}\zeta^{(4)}(R_3,R_4,R_1,R_2)\,.\tag{4.8}$$

Proposition 4.3. *If u is a stable and regular pair potential and the activity z satisfies (2.8) then there exists $C>0$ independent of the size of Λ, such that*

$$\bigl|\chi_\Lambda^{(4)}(R_1,R_2,R_3,R_4)\bigr| \,\leq\, Ce^{-\beta u(|R_3-R_4|)}\sum_{i=1}^{2}\sum_{j=3}^{4}G(R_i-R_j)\tag{4.9}$$

for all $R_1, R_2, R_3, R_4 \in \Lambda$.

Proof. From (4.4), (4.5), and the definition (3.18) of $\psi(\mathcal{I}_2; \mathcal{J}_{2,N})$ we obtain as in the proof of Lemma 4.1 that

$$\bigl|\zeta^{(4)}(R_1, R_2, R_3, R_4)\bigr| \le e^{-4\beta B}(S_1^{(4)} + S_2^{(4)} + S_3^{(4)} + S_4^{(4)}), \tag{4.10}$$

where

$$S_l^{(4)} = \sum_{N=4}^{\infty} \frac{(ze^{2\beta B})^N}{(N-4)!} \sum_{\mathcal{F}_l} \int_{\Lambda^{N-4}} \Bigl(\prod_{(i,j)\in\mathcal{F}_l} |f_{ij}|\Bigr) \,\mathrm{d}\boldsymbol{R}_{4,N}, \qquad l = 1, \dots, 4,$$

where the inner sum varies over all those forests $\mathcal{F}_l \in \mathfrak{F}_{\mathcal{I}_2, \mathcal{J}_{2,N}}$, for which in case of
$l = 1$: vertex #3 belongs to the tree rooted in vertex #1, and vertex #4 does not;
$l = 2$: vertex #4 belongs to the tree rooted in vertex #1, and vertex #3 does not;
$l = 3$: vertex #3 and #4 belong to the same tree rooted in vertex #1;
$l = 4$: vertex #3 and #4 belong to the same tree rooted in vertex #2.
Standard graph analysis and Lemma 3.1 immediately lead to bounds for the first two cases, namely

$$S_1^{(4)} \le G(R_1 - R_3)G(R_2 - R_4), \qquad S_2^{(4)} \le G(R_1 - R_4)G(R_2 - R_3).$$

Concerning $S_3^{(4)}$ we first consider those forests (in $S_{31}^{(4)}$, say) where the connecting path between vertices #1 and #4 passes through vertex #3, and the remaining forests (in $S_{32}^{(4)}$) where the path between vertices #1 to #3 passes through vertex #4. In the first case the trees rooted in vertex #1 can be constructed by glueing together a tree rooted in vertex #1 and containing vertex #3 and a second tree rooted in vertex #3 and containing vertex #4; this yields the bound

$$S_{31}^{(4)} \le \frac{w}{ze^{2\beta B}}\, G(R_1 - R_3)G(R_3 - R_4),$$

where the numerator is due to the fact that vertex #3 is a joint vertex of the two trees that are glued together, and the extra factor w stems from the tree rooted in vertex #2.

Likewise, we obtain corresponding bounds for $S_{32}^{(4)}$ and $S_4^{(4)}$, namely

$$S_{32}^{(4)} \le \frac{w}{ze^{2\beta B}}\, G(R_1 - R_4)G(R_3 - R_4)$$

and

$$S_4^{(4)} \le \frac{w}{ze^{2\beta B}}\, \bigl(G(R_2 - R_3) + G(R_2 - R_4)\bigr)G(R_3 - R_4).$$

Since G is bounded, cf. Proposition 3.2, we finally obtain by inserting all these bounds into (4.10) that

$$\bigl|\zeta^{(4)}(R_1, R_2, R_3, R_4)\bigr| \le C\bigl(G(R_1 - R_3) + G(R_2 - R_3) + G(R_1 - R_4) + G(R_2 - R_4)\bigr).$$

Together with (4.8) this yields

$$\bigl|\eta(R_1, R_2, R_3, R_4)\bigr| \le Ce^{-\beta u(|R_3 - R_4|)} \sum_{i=1}^{2} \sum_{j=3}^{4} G(R_i - R_j).$$

From Proposition 4.2 it follows that a similar inequality (with a different constant) holds true for the last two terms of (4.7) either, hence the proof is done. ∎

5. Differentiability of the pair correlation function in $L^1(\mathbb{R}^3)$. Throughout this section we consider perturbations $\widetilde{u} = u + v$ of a given potential u that satisfies Assumption A, where $v \in \mathscr{V}_u$ with $\|v\|_{\mathscr{V}_u} \leq t_0/2$ is kept fixed. Associated with u and two finite index sets $\mathcal{I}$ and $\mathcal{J}$ with $\mathcal{I} \cap \mathcal{J} = \emptyset$ are the Ruelle functions $\varphi(\mathcal{I};\mathcal{J})$ of (3.11), and we will associate corresponding Ruelle functions $\widetilde{\varphi}(\mathcal{I};\mathcal{J})$ with the perturbed potential $\widetilde{u}$. Later on we also resort to the pair correlation function $\widetilde{\omega}_\Lambda^{(2)}$ corresponding to the grand canonical ensemble with interaction potential $\widetilde{u}$.

We need a few auxiliary estimates from [3]. The first one, compare Lemma 3.2 in [3], concerns the functions $d_{\mathcal{I},j^*}$ of (3.12) with $j^* = j^*(\mathcal{I}, \boldsymbol{R}_\mathcal{I})$ selected as in (3.15): If $\widetilde{d}_{\mathcal{I},j^*}$ is the corresponding function associated with $\widetilde{u}$ and if $\|v\|_{\mathscr{V}_u} \leq t_0/2$ then there holds

$$\|\widetilde{d}_{\mathcal{I},j^*} - d_{\mathcal{I},j^*}\|_{(\mathbb{R}^3)^{|\mathcal{I}|}} \;\leq\; \frac{2e^{2\beta B}}{t_0}\,\|v\|_{\mathscr{V}_u}\,, \tag{5.1a}$$

$$\|\widetilde{d}_{\mathcal{I},j^*} - d_{\mathcal{I},j^*} - (\partial d_{\mathcal{I},j^*})v\|_{(\mathbb{R}^3)^{|\mathcal{I}|}} \;\leq\; \frac{4e^{2\beta B}}{t_0^2}\,\|v\|_{\mathscr{V}_u}^2\,, \tag{5.1b}$$

where $\partial d_{\mathcal{I},j^*}$ is the Fréchet derivative of $d_{\mathcal{I},j^*}$ whose specific form is given in [3] but is not relevant for our purposes below. Take note that the estimates (5.1) make use of the fact that the index $j^*(\mathcal{I}, \boldsymbol{R}_\mathcal{I})$ does not depend on v because of our smallness assumption, cf. (3.15). Second, for $\mathcal{K} \subset \mathbb{N}$ let $k_\mathcal{K}$ be given by (3.13) and $\widetilde{k}_\mathcal{K}$ be the corresponding function associated with $\widetilde{u}$. Then, since $\|v\|_{\mathscr{V}_u} \leq t_0$, there exists a constant $C_\beta > 0$ such that

$$\sup_{R\in\mathbb{R}^3} \bigl\|\widetilde{k}_\mathcal{K}(R) - k_\mathcal{K}(R)\bigr\|_{L^1((\mathbb{R}^3)^n)} \;\leq\; nC_\beta c_\beta^{n-1}\,\|v\|_{\mathscr{V}_u}\,, \tag{5.2a}$$

$$\sup_{R\in\mathbb{R}^3} \bigl\|\widetilde{k}_\mathcal{K}(R) - k_\mathcal{K}(R) - \bigl((\partial k_\mathcal{K})v\bigr)(R)\bigr\|_{L^1((\mathbb{R}^3)^n)} \;\leq\; n^2 C_\beta c_\beta^{n-1}\|v\|_{\mathscr{V}_u}^2\,, \tag{5.2b}$$

where $n = |\mathcal{K}|$; see the proof of Proposition 3.3 in [3]. Again, the specific form of the Fréchet derivative $\partial k_\mathcal{K}$ does not matter.

Now we can estimate the difference between the Ruelle functions associated with u and $\widetilde{u} = u + v$.

Lemma 5.1. *Under the assumptions of this section let $\mathcal{I},\mathcal{J} \subset \mathbb{N}$ be two finite index sets with $\mathcal{I} \neq \emptyset$ and $\mathcal{I} \cap \mathcal{J} = \emptyset$. Then there holds*

$$\begin{aligned}\int_{\Lambda^{|\mathcal{J}|}} &\Bigl|\widetilde{\varphi}(\mathcal{I};\mathcal{J}) \,-\, \varphi(\mathcal{I};\mathcal{J})\Bigr|\,\mathrm{d}\boldsymbol{R}_\mathcal{J}\\ &\leq\; \bigl(|\mathcal{I}\cup\mathcal{J}| - 1\bigr)\,\frac{2c_\beta + t_0C_\beta}{t_0c_\beta}\,(|\mathcal{J}|!)c_\beta^{|\mathcal{J}|}\bigl(e^{2\beta B+1}\bigr)^{|\mathcal{I}\cup\mathcal{J}|-1}\|v\|_{\mathscr{V}_u}\,.\end{aligned} \tag{5.3}$$

Proof. Before we start we define

$$\Delta_0\varphi(\mathcal{I};\mathcal{J}) \,=\, \widetilde{\varphi}(\mathcal{I};\mathcal{J}) \,-\, \varphi(\mathcal{I};\mathcal{J})\,. \tag{5.4}$$

Now the proof proceeds by induction on $|\mathcal{I}\cup\mathcal{J}|$. When $|\mathcal{I}\cup\mathcal{J}| = 1$, i.e., when $\mathcal{I}$ consists of a single element and $\mathcal{J} = \emptyset$, then

$$\varphi_{\mathcal{I},\mathcal{J}} \,=\, \widetilde{\varphi}_{\mathcal{I},\mathcal{J}} \,=\, 1$$

by virtue of (3.11), and hence the assertion is obviously correct. Note that (5.3) is also true when $\mathcal{I}=\emptyset$ and $\mathcal{J}\neq\emptyset$ is arbitrary; this will be used in the induction step.

Concerning the induction, we use (3.14) with $i^*=j^*(\mathcal{I},\boldsymbol{R}_\mathcal{I})$ of (3.15) for $|\mathcal{I}\cup\mathcal{J}|\geq 2$, $\mathcal{I}\neq\emptyset$, to derive the recursion

$$\begin{aligned}\Delta_0\varphi(\mathcal{I};\mathcal{J}) \;=\; &(\widetilde{d}_{\mathcal{I},j^*}-d_{\mathcal{I},j^*})\sum_{\mathcal{K}\subset\mathcal{J}}\widetilde{k}_\mathcal{K}(R_{j^*})\widetilde{\varphi}(\mathcal{I}\cup\mathcal{K}\setminus\{j^*\};\mathcal{J}\setminus\mathcal{K})\\ &+d_{\mathcal{I},j^*}\sum_{\mathcal{K}\subset\mathcal{J}}\bigl(\widetilde{k}_\mathcal{K}(R_{j^*})-k_\mathcal{K}(R_{j^*})\bigr)\widetilde{\varphi}(\mathcal{I}\cup\mathcal{K}\setminus\{j^*\};\mathcal{J}\setminus\mathcal{K})\\ &+d_{\mathcal{I},j^*}\sum_{\mathcal{K}\subset\mathcal{J}}k_\mathcal{K}(R_{j^*})\Delta_0\varphi(\mathcal{I}\cup\mathcal{K}\setminus\{j^*\};\mathcal{J}\setminus\mathcal{K})\,.\end{aligned}$$

Integrating over $\boldsymbol{R}_\mathcal{J}$ and utilizing (5.1a), (5.2a), Ruelle's estimate (3.17), and the induction hypothesis (5.3) we thus obtain

$$\begin{aligned}\int_{\Lambda^{|\mathcal{J}|}}\bigl|\Delta_0\varphi(\mathcal{I},\mathcal{J})\bigr|\,\mathrm{d}\boldsymbol{R}_\mathcal{J}\;\leq\;&\frac{1}{e}(e^{2\beta B+1})^{|\mathcal{I}|+|\mathcal{J}|-1}c_\beta^{|\mathcal{J}|}\,\cdot\\ &\sum_{\mathcal{K}\subset\mathcal{J}}(|\mathcal{J}|-|\mathcal{K}|)!\,\Bigl(\frac{2}{t_0}\,+\,|\mathcal{K}|\,\frac{C_\beta}{c_\beta}\,+\,\bigl(|\mathcal{I}\cup\mathcal{J}|-2\bigr)\frac{2c_\beta+t_0C_\beta}{t_0c_\beta}\Bigr)\|v\|_{\mathscr{V}_u}\,.\end{aligned}$$

Since the right-hand side only depends on the number p of elements in $\mathcal{K}$ we can sum over p instead which gives the upper bound

$$\begin{aligned}\int_{\Lambda^{|\mathcal{J}|}}\bigl|\Delta_0\varphi(\mathcal{I};\mathcal{J})\bigr|\,\mathrm{d}\boldsymbol{R}_\mathcal{J}\;\leq\;&\frac{1}{e}(e^{2\beta B+1})^{|\mathcal{I}|+|\mathcal{J}|-1}c_\beta^{|\mathcal{J}|}(|\mathcal{J}|!)\|v\|_{\mathscr{V}_u}\,\cdot\\ &\sum_{p=0}^{\infty}\frac{1}{p!}\Bigl(\frac{2}{t_0}\,+\,p\,\frac{C_\beta}{c_\beta}\,+\,\bigl(|\mathcal{I}\cup\mathcal{J}|-2\bigr)\frac{2c_\beta+t_0C_\beta}{t_0c_\beta}\Bigr)\end{aligned}$$

which coincides with (5.3). ◻

Under the assumptions of Lemma 5.1 and with the same notation as before we let

$$\varphi'(\mathcal{I};\mathcal{J})\;=\;0\qquad\text{for}\quad|\mathcal{I}|=0\quad\text{or}\quad|\mathcal{I}|=1,\,|\mathcal{J}|=0\,,$$

and for $\mathcal{I}\cap\mathcal{J}=\emptyset$, $|\mathcal{I}\cup\mathcal{J}|\geq 2$, $|\mathcal{I}|\neq 0$, and with $j^*=j^*(\mathcal{I},\boldsymbol{R}_\mathcal{I})$ we define recursively

$$\begin{aligned}\varphi'(\mathcal{I};\mathcal{J})\;=\;&(\partial d_{\mathcal{I},j^*})v\sum_{\mathcal{K}\subset\mathcal{J}}k_\mathcal{K}(R_{j^*})\varphi(\mathcal{I}\cup\mathcal{K}\setminus\{j^*\};\mathcal{J}\setminus\mathcal{K})\\ &+d_{\mathcal{I},j^*}\sum_{\mathcal{K}\subset\mathcal{J}}\bigl((\partial k_\mathcal{K})v\bigr)(R_{j^*})\varphi(\mathcal{I}\cup\mathcal{K}\setminus\{j^*\};\mathcal{J}\setminus\mathcal{K})\\ &+d_{\mathcal{I},j^*}\sum_{\mathcal{K}\subset\mathcal{J}}k_\mathcal{K}(R_{j^*})\varphi'(\mathcal{I}\cup\mathcal{K}\setminus\{j^*\};\mathcal{J}\setminus\mathcal{K})\,.\end{aligned}\tag{5.5}$$

Take note that φ' depends linearly on v.

LEMMA 5.2. *Under the same assumptions as in Lemma 5.1 there exists a constant C such that*

$$\begin{aligned}&\int_{\Lambda^{|\mathcal{J}|}}\Bigl|\widetilde{\varphi}(\mathcal{I};\mathcal{J})\,-\,\varphi(\mathcal{I};\mathcal{J})\,-\,\varphi'(\mathcal{I};\mathcal{J})\Bigr|\,\mathrm{d}\boldsymbol{R}_\mathcal{J}\\ &\qquad\leq\;C(|\mathcal{I}|+|\mathcal{J}|-1)^2(|\mathcal{J}|!)\,c_\beta^{|\mathcal{J}|}(e^{2\beta B+1})^{|\mathcal{I}|+|\mathcal{J}|-1}\|v\|_{\mathscr{V}_u}^2\,.\end{aligned}$$

The constant C only depends on u and on t_0, cf. (5.7), but neither on the size of Λ nor on $\boldsymbol{R}_{\mathcal{I}} \in \Lambda^{|\mathcal{I}|}$.

Proof. Again the proof proceeds by induction on $|\mathcal{I}|+|\mathcal{J}|$, where for $|\mathcal{I}|+|\mathcal{J}|=1$ there is nothing to prove. Utilizing the notations (5.4) and

$$\Delta_1\varphi(\mathcal{I};\mathcal{J}) \,=\, \widetilde{\varphi}(\mathcal{I};\mathcal{J}) - \varphi(\mathcal{I};\mathcal{J}) - \varphi'(\mathcal{I};\mathcal{J}) \tag{5.6}$$

for the zeroth and first order Taylor remainders of $\varphi(\mathcal{I};\mathcal{J})$ we can use the recursions (3.14) and (5.5) for $N := |\mathcal{I}|+|\mathcal{J}|-2 \geq 0$, $|\mathcal{I}| \neq 0$, to obtain

$$\begin{aligned}
\Delta_1\varphi(\mathcal{I};\mathcal{J}) \,=\, &\big(\widetilde{d}_{\mathcal{I},j^*} - d_{\mathcal{I},j^*} - (\partial d_{\mathcal{I},j^*})v\big) \sum_{\mathcal{K}\subset\mathcal{J}} k_{\mathcal{K}}(R_{j^*})\varphi(\mathcal{I}\cup\mathcal{K}\setminus\{j^*\};\mathcal{J}\setminus\mathcal{K})\\
&+ (\widetilde{d}_{\mathcal{I},j^*} - d_{\mathcal{I},j^*}) \sum_{\mathcal{K}\subset\mathcal{J}} \big(\widetilde{k}_{\mathcal{K}}(R_{j^*}) - k_{\mathcal{K}}(R_{j^*})\big)\varphi(\mathcal{I}\cup\mathcal{K}\setminus\{j^*\};\mathcal{J}\setminus\mathcal{K})\\
&+ (\widetilde{d}_{\mathcal{I},j^*} - d_{\mathcal{I},j^*}) \sum_{\mathcal{K}\subset\mathcal{J}} \widetilde{k}_{\mathcal{K}}(R_{j^*})\Delta_0\varphi(\mathcal{I}\cup\mathcal{K}\setminus\{j^*\};\mathcal{J}\setminus\mathcal{K})\\
&+ d_{\mathcal{I},j^*} \sum_{\mathcal{K}\subset\mathcal{J}} \big(\widetilde{k}_{\mathcal{K}}(R_{j^*}) - k_{\mathcal{K}}(R_{j^*}) - ((\partial k_{\mathcal{K}})v)(R_{j^*})\big)\varphi(\mathcal{I}\cup\mathcal{K}\setminus\{j^*\};\mathcal{J}\setminus\mathcal{K})\\
&+ d_{\mathcal{I},j^*} \sum_{\mathcal{K}\subset\mathcal{J}} \big(\widetilde{k}_{\mathcal{K}}(R_{j^*}) - k_{\mathcal{K}}(R_{j^*})\big)\Delta_0\varphi(\mathcal{I}\cup\mathcal{K}\setminus\{j^*\};\mathcal{J}\setminus\mathcal{K})\\
&+ d_{\mathcal{I},j^*} \sum_{\mathcal{K}\subset\mathcal{J}} k_{\mathcal{K}}(R_{j^*})\Delta_1\varphi(\mathcal{I}\cup\mathcal{K}\setminus\{j^*\};\mathcal{J}\setminus\mathcal{K})\,.
\end{aligned}$$

Integrating over $\boldsymbol{R}_{\mathcal{J}} \in \Lambda^{|\mathcal{J}|}$ and using the inequalities (5.1b), (3.17), (5.1a), (5.2a), (5.3), (5.2b), and the induction hypothesis then we obtain in the same way as in the proof of Lemma 5.1 that

$$\int_{\Lambda^{|\mathcal{J}|}} \big|\Delta_1\varphi(\mathcal{I};\mathcal{J})\big| \,\mathrm{d}\boldsymbol{R}_{\mathcal{J}} \,\leq\, \big(C \,+\, 2CN + CN^2\big)(|\mathcal{J}|!)\, c_\beta^{|\mathcal{J}|} e^{(2\beta B+1)(N+1)} \|v\|_{\mathscr{V}_u}^2\,,$$

provided that we let

$$C \,=\, \max\Big\{\, \frac{4c_\beta + 2C_\beta t_0 + 2C_\beta t_0^2}{t_0^2 c_\beta}\,,\, \frac{1}{2}\Big(\frac{2c_\beta + t_0 C_\beta}{t_0 c_\beta}\Big)^2 \,\Big\}\,. \tag{5.7}$$

Since $N+1 = |\mathcal{I}|+|\mathcal{J}|-1$ the induction step is complete. ☐

Now we come to the main result of this section.

THEOREM 5.3. *Let u satisfy Assumption A and z be constrained by (2.8). Then the thermodynamical limit $\omega^{(2)}(\,\cdot\,,0)$ of the corresponding pair correlation function (2.9) is Fréchet differentiable in $\mathscr{L}\big(\mathscr{V}_u, L^1(\mathbb{R}^3)\big)$.*

Proof. From [3] we know that the molecular distribution functions $\rho_\Lambda^{(1)}$ and $\rho_\Lambda^{(2)}$ are Fréchet differentiable with respect to $L^\infty(\Lambda)$, respectively $L^\infty(\Lambda^2)$. Hence, the function

$$\omega_\Lambda^{(2)}(R_1,R_2) \,=\, \rho_\Lambda^{(2)}(R_1,R_2) \,-\, \rho_\Lambda^{(1)}(R_1)\rho_\Lambda^{(1)}(R_2)\,, \qquad R_1,R_2\in\Lambda\,,$$

has a Fréchet derivative $\partial\omega_\Lambda^{(2)}$ (with respect to u) in the same topology, and this implies Fréchet differentiability with the same derivative also in $L^1(\Lambda^2)$. For a given $v \in \mathscr{V}_u$ define

$$\omega'_\Lambda(R_1,R_2) \,=\, \sum_{N=2}^{\infty} \frac{z^N}{(N-2)!} \int_{\Lambda^{N-2}} \varphi'(\{1\};\{2,\dots,N\}) \,\mathrm{d}\boldsymbol{R}_{N-2}$$

with φ' of (5.5). Using the notation from (5.6) it follows from Lemma 5.2 that

$$\begin{aligned}
&\int_\Lambda \Big| \widetilde{\omega}_\Lambda^{(2)}(R_1,R_2) - \omega_\Lambda^{(2)}(R_1,R_2) \,-\, \omega'_\Lambda(R_1,R_2) \Big| \, \mathrm{d}R_2 \\
&\qquad \le \int_\Lambda \sum_{N=2}^\infty \frac{z^N}{(N-2)!} \int_{\Lambda^{N-2}} \Big| \Delta_1 \varphi\big(\{1\}\,;\{2,\dots,N\}\big)\Big| \, \mathrm{d}\boldsymbol{R}_{2,N}\, \mathrm{d}R_2 \\
&\qquad \le \sum_{N=2}^\infty C z^N (N-1)^3 \big(c_\beta e^{2\beta B+1}\big)^{N-1} \|v\|_{\mathscr{V}_u}^2 \,=\, C' \|v\|_{\mathscr{V}_u}^2 \,,
\end{aligned} \tag{5.8}$$

where the constant C' is finite because of (2.8) and independent of the size of Λ and independent of the choice of $R_1 \in \Lambda$. Since ω'_Λ depends linearly on v this inequality reveals that

$$(\partial \omega_\Lambda^{(2)})v \,=\, \omega'_\Lambda \,.$$

From [3] we know that $(\partial\rho_\Lambda^{(2)})v \to (\partial\rho^{(2)})v$ and $(\partial\rho_\Lambda^{(1)})v \to (\partial\rho^{(1)})v$ compactly as $|\Lambda| \to \infty$. The latter is necessarily a constant denoted by $(\partial\rho_0)v$ in the sequel for brevity. We also know that $\rho_\Lambda^{(1)} \to \rho_0$ compactly as $|\Lambda| \to \infty$. It thus follows that

$$\begin{aligned}
\big((\partial\omega_\Lambda^{(2)})v\big)(R_1,R_2) \,&\to\, \big((\partial\rho^{(2)})v\big)(R_1,R_2) \,-\, 2\rho_0(\partial\rho_0)v \\
&= \big((\partial\omega^{(2)})v\big)(R_1,R_2)\,, \qquad |\Lambda| \to \infty\,,
\end{aligned}$$

uniformly on bounded subsets of $(\mathbb{R}^3)^2$, where $\partial\omega^{(2)}$ is the Fréchet derivative of $\omega^{(2)}$ in $\mathscr{L}(\mathscr{V}_u, L^\infty((\mathbb{R}^3)^2)$. Choosing any fixed box $\Lambda' \subset \Lambda$ we obtain from (5.8) that

$$\int_{\Lambda'} \Big| \widetilde{\omega}_\Lambda^{(2)}(R,0) - \omega_\Lambda^{(2)}(R,0) \,-\, \big((\partial\omega_\Lambda^{(2)})v\big)(R,0)\Big| \, \mathrm{d}R \,\le\, C'\|v\|_{\mathscr{V}_u}^2\,,$$

and by letting $|\Lambda| \to \infty$ this implies that

$$\int_{\Lambda'} \Big| \widetilde{\omega}^{(2)}(R,0) - \omega^{(2)}(R,0) \,-\, \big((\partial\omega^{(2)})v\big)(R,0)\Big| \, \mathrm{d}R \,\le\, C'\|v\|_{\mathscr{V}_u}^2\,.$$

Since the box $\Lambda' \subset \mathbb{R}^3$ can be arbitrarily large we thus have proved that $\partial\omega^{(2)}(\,\cdot\,,0)$ is also the Fréchet derivative of $\omega^{(2)}(\,\cdot\,,0)$ in $\mathscr{L}\big(\mathscr{V}_u, L^1(\mathbb{R}^3)\big)$. □

6. Integral operator representations of $\partial\rho^{(1)}$ and $\partial\rho^{(2)}$. In a finite size box $\Lambda \subset \mathbb{R}^3$, and with u satisfying Assumption A, the derivatives $\partial\rho_\Lambda^{(m)}$ can be represented as integral operators acting on $\mathscr{V}_u$. For $m = 1, 2$ these operators have been presented in [3]: There holds

$$\begin{aligned}
\big((\partial\rho_\Lambda^{(1)})v\big)(R_1) \,=\, &-\beta \int_\Lambda v(|R_1 - R'|)\rho_\Lambda^{(2)}(R_1,R')\,\mathrm{d}R' \\
&- \frac{\beta}{2}\int_\Lambda\int_\Lambda v(|R'_1 - R'_2|)\chi_\Lambda^{(3)}(R_1,R'_1,R'_2)\,\mathrm{d}R'_1\,\mathrm{d}R'_2
\end{aligned} \tag{6.1}$$

with $\chi_\Lambda^{(3)}$ of (4.1), and

$$\begin{aligned}\big((\partial\rho_\Lambda^{(2)})v\big)(R_1,R_2) \;=\; &-\beta\, v(|R_1-R_2|)\rho_\Lambda^{(2)}(R_1,R_2)\\ &-\beta\int_\Lambda v(|R_1-R'|)\rho_\Lambda^{(3)}(R_1,R_2,R')\,\mathrm{d}R'\\ &-\beta\int_\Lambda v(|R_2-R'|)\rho_\Lambda^{(3)}(R_1,R_2,R')\,\mathrm{d}R'\\ &-\frac{\beta}{2}\int_\Lambda\int_\Lambda v(|R_1'-R_2'|)\chi_\Lambda^{(4)}(R_1,R_2,R_1',R_2')\,\mathrm{d}R_1'\,\mathrm{d}R_2' \end{aligned}\tag{6.2}$$

with $\chi_\Lambda^{(4)}$ of (4.2). We refer to [3] for physical interpretations of these representations.

The goal of this section is to show that the corresponding formulae for $\partial\rho^{(m)}$, $m=1,2$, are obtained by integrating over $\mathbb{R}^3$ instead, and by dropping all subscripts Λ, where

$$\chi^{(3)}(R_1,R_2,R_3) \;=\; \rho^{(3)}(R_1,R_2,R_3) \;-\; \rho^{(1)}(R_1)\rho^{(2)}(R_2,R_3)$$

and

$$\chi^{(4)}(R_1,R_2,R_3,R_4) \;=\; \rho^{(4)}(R_1,R_2,R_3,R_4) \;-\; \rho^{(2)}(R_1,R_2)\rho^{(2)}(R_3,R_4)\,,$$

Concerning the verification of this assertion for the single integrals appearing in (6.1) and (6.2) we utilize the following auxiliary result.

Lemma 6.1. *Let u satisfy Assumption A, and for some $R_0\in\mathbb{R}^3$ and $C>0$ let $\xi_\Lambda:\Lambda\to\mathbb{R}$ be a family of functions with*

$$|\xi_\Lambda(R)| \;\le\; Ce^{-\beta u(|R-R_0|)}\,, \qquad R\in\Lambda\,, \tag{6.3}$$

independent of Λ. Moreover, let ξ_Λ converge compactly to $\xi:\mathbb{R}^3\to\mathbb{R}$ as $|\Lambda|\to\infty$. Then for every $v\in\mathscr{V}_u$ there holds

$$\int_\Lambda v(|R-R_0|)\xi_\Lambda(R)\,\mathrm{d}R \;\to\; \int_{\mathbb{R}^3} v(|R-R_0|)\xi(R)\,\mathrm{d}R$$

as $|\Lambda|\to\infty$.

Proof. We extend ξ_Λ by zero to $\mathbb{R}^3\setminus\Lambda$ and rewrite

$$\ell_\Lambda(v) := \int_\Lambda v(|R-R_0|)\,\xi_\Lambda(R)\,\mathrm{d}R = \int_{\mathbb{R}^3} v(|R-R_0|)e^{-\beta u(|R-R_0|)}\big(\xi_\Lambda(R)e^{\beta u(|R-R_0|)}\big)\,\mathrm{d}R\,.$$

By virtue of [3, Lemma 3.1] $\mathscr{V}_u$ is continuously embedded into the space $\mathscr{Y}_u$ of functions $v:\mathbb{R}^+\to\mathbb{R}$, for which the corresponding norm

$$\|v\|_{\mathscr{Y}_u} \;:=\; \int_{\mathbb{R}^3} v(|R|)e^{-\beta u(|R|)}\;\mathrm{d}R \tag{6.4}$$

is finite. In view of (6.3) ℓ_Λ is a linear functional in $\mathscr{Y}_u'$, and $\{\ell_\Lambda\}_\Lambda\subset\mathscr{Y}_u'$ is uniformly bounded. Furthermore, for $v\in\mathscr{Y}_u$ with compact support the compact convergence of $\xi_\Lambda\to\xi$ as $|\Lambda|\to\infty$ implies that

$$\ell_\Lambda(v) \;\to\; \ell(v) \;=\; \int_{\mathbb{R}^3} v(|R-R_0|)\xi(R)\,\mathrm{d}R\,, \qquad |\Lambda|\to\infty\,,$$

hence, the assertion of the lemma follows from the Banach-Steinhaus theorem for every $v \in \mathscr{Y}_u$, and hence, every $v \in \mathscr{V}_u$. ☐

To apply this result to (6.1) and (6.2) we need to estimate the molecular distribution functions when their arguments get close.

Proposition 6.2. *Let u be a stable and regular pair potential and let z satisfy (2.8). Then there exists $C > 0$, independent of the size of Λ, such that*

$$\rho_\Lambda^{(m)}(\boldsymbol{R}_m) \,\leq\, C(ze^{2\beta B+1})^m \prod_{i=1}^{m-1} e^{-\beta u(|R_i - R_m|)} \tag{6.5}$$

for all $\boldsymbol{R}_m \in \Lambda^m$ and all $m \geq 2$.

Proof. With $\mathcal{I}_m = \{1, \dots, m\}$, $\mathcal{J}_{m,N} = \{m+1, \dots, N\}$, and $i^* = m$ we conclude from (3.6), (3.11), (3.14), and (3.17) that

$$\begin{aligned}
\big|\rho_\Lambda^{(m)}(\boldsymbol{R}_m)\big| \,&\leq\, \sum_{N=m}^\infty \frac{z^N}{(N-m)!}(e^{2\beta B+1})^{N-2}\, d_{\mathcal{I}_m,m}(\boldsymbol{R}_m)\,\cdot \\
&\qquad\qquad \sum_{\mathcal{K}\subset\mathcal{J}_{m,N}} (N-m-|\mathcal{K}|)!\, c_\beta^{N-m-|\mathcal{K}|} \int_{\Lambda^{|\mathcal{K}|}} |k_\mathcal{K}(R_m;\boldsymbol{R}_\mathcal{K})|\,\mathrm{d}\boldsymbol{R}_\mathcal{K} \\
&\leq\, \sum_{N=m}^\infty \frac{z^N}{(N-m)!}(e^{2\beta B+1})^{N-2}\, d_{\mathcal{I}_m,m}(\boldsymbol{R}_m) c_\beta^{N-m} \sum_{\mathcal{K}\subset\mathcal{J}_{m,N}} (N-m-|\mathcal{K}|)!\,.
\end{aligned}$$

The inner sum only depends on the number p of elements in $\mathcal{K}$, $0 \leq p \leq N-m$, hence

$$\begin{aligned}
\big|\rho_\Lambda^{(m)}(\boldsymbol{R}_m)\big| \,&\leq\, \sum_{N=m}^\infty z^N (e^{2\beta B+1})^{N-2}\, d_{\mathcal{I}_m,m}(\boldsymbol{R}_m) c_\beta^{N-m} \sum_{p=0}^{N-m} \frac{1}{p!} \\
&\leq\, \frac{ez^2}{1 - zc_\beta e^{2\beta B+1}}\,(ze^{2\beta B+1})^{m-2} d_{\mathcal{I}_m,m}(\boldsymbol{R}_m)\,.
\end{aligned} \tag{6.6}$$

The assertion now follows by inserting the definition (3.12) of $d_{\mathcal{I}_m,m}$. ☐

Combining Lemma 6.1 and Proposition 6.2 we readily obtain the thermodynamical limits of the three single integrals occurring in (6.1) and (6.2). The thermodynamical limits of the remaining two double integrals involving $\chi_\Lambda^{(m)}$, $m = 3, 4$ in (6.1) and (6.2), respectively, are more subtle and will be considered next.

Again, we start with an auxiliary result.

Lemma 6.3. *Let u satisfy Assumption A, and $\chi_\Lambda : \Lambda^2 \to \mathbb{R}$ be a family of functions with*

$$|\chi_\Lambda(R_1, R_2)| \,\leq\, e^{-\beta u(|R_1 - R_2|)}\big(X(R_1) + X(R_2)\big) \tag{6.7}$$

for all $R_1, R_2 \in \Lambda$, where $X \in L^1(\mathbb{R}^3)$ is nonnegative and bounded and does not depend on Λ. Furthermore, assume that χ_Λ converges compactly to $\chi : (\mathbb{R}^3)^2 \to \mathbb{R}$ as $|\Lambda| \to \infty$. Then for every $v \in \mathscr{V}_u$ there holds

$$\int_\Lambda \int_\Lambda v(|R_1 - R_2|)\chi_\Lambda(R_1, R_2)\;\mathrm{d}R_1\,\mathrm{d}R_2 \;\to\; \int_{\mathbb{R}^3}\int_{\mathbb{R}^3} v(|R_1 - R_2|)\chi(R_1, R_2)\;\mathrm{d}R_1\,\mathrm{d}R_2$$

as $|\Lambda| \to \infty$.

Proof. Throughout, we extend χ_Λ by zero to $(\mathbb{R}^3)^2 \setminus \Lambda^2$, and this extension, of course, satisfies (6.7) for all $R_1, R_2 \in \mathbb{R}^3$. Because of the compact convergence $\chi_\Lambda \to \chi$ as $|\Lambda| \to \infty$ this inequality also extends to χ, i.e.,

$$|\chi(R_1, R_2)| \,\leq\, e^{-\beta u(|R_1 - R_2|)}\bigl(X(R_1) + X(R_2)\bigr)\,, \qquad R_1, R_2 \in \mathbb{R}^3\,. \tag{6.8}$$

Substituting $R_1' = R_1 - R_2$ and $R_2' = R_1 + R_2$ we obtain

$$\begin{aligned} &\int_\Lambda \int_\Lambda v(|R_1 - R_2|)\chi_\Lambda(R_1, R_2)\,\mathrm{d}R_1\,\mathrm{d}R_2 \\ &\qquad = \frac{1}{8}\int_{\mathbb{R}^3} v(|R_1'|)e^{-\beta u(|R_1'|)}\int_{\mathbb{R}^3} e^{\beta u(|R_1'|)}\chi_\Lambda\bigl(\tfrac{R_1'+R_2'}{2}, \tfrac{R_2'-R_1'}{2}\bigr)\,\mathrm{d}R_2'\,\mathrm{d}R_1'\,, \end{aligned}$$

and hence, since $v \in \mathscr{V}_u \subset \mathscr{Y}_u$, compare (6.4), it only remains to show that

$$J(R_1') \,=\, \int_{\mathbb{R}^3} e^{\beta u(|R_1'|)}\Bigl(\chi_\Lambda\bigl(\tfrac{R_1'+R_2'}{2}, \tfrac{R_2'-R_1'}{2}\bigr) \,-\, \chi\bigl(\tfrac{R_1'+R_2'}{2}, \tfrac{R_2'-R_1'}{2}\bigr)\Bigr)\,\mathrm{d}R_2'$$

is uniformly bounded for $R_1' \in \mathbb{R}^3$, and converges compactly to zero. The uniform boundedness follows readily from (6.7) and (6.8), since

$$|J(R_1')| \,\leq\, 2\int_{\mathbb{R}^3} X\bigl(\tfrac{R_1'+R_2'}{2}\bigr)\,\mathrm{d}R_2' \,+\, 2\int_{\mathbb{R}^3} X\bigl(\tfrac{R_2'-R_1'}{2}\bigr)\,\mathrm{d}R_2' \,=\, 32\,\|X\|_{L^1(\mathbb{R}^3)}\,.$$

To prove the compact convergence $J \to 0$ we introduce for $r' > 0$ the spherical shell $\mathcal{A}_{r'} = \{1/r' \leq |R_2'| \leq r'\}$, and estimate

$$\begin{aligned} |J(R_1')| \,\leq\, &\int_{\mathcal{A}_{r'}} e^{\beta u(|R_1'|)}\Bigl|\chi_\Lambda\bigl(\tfrac{R_1'+R_2'}{2}, \tfrac{R_2'-R_1'}{2}\bigr) \,-\, \chi\bigl(\tfrac{R_1'+R_2'}{2}, \tfrac{R_2'-R_1'}{2}\bigr)\Bigr|\,\mathrm{d}R_2' \\ &\quad + \int_{\mathbb{R}^3\setminus\mathcal{A}_{r'}} e^{\beta u(|R_1'|)}\Bigl(\bigl|\chi_\Lambda\bigl(\tfrac{R_1'+R_2'}{2}, \tfrac{R_2'-R_1'}{2}\bigr)\bigr| \,+\, \bigl|\chi\bigl(\tfrac{R_1'+R_2'}{2}, \tfrac{R_2'-R_1'}{2}\bigr)\bigr|\Bigr)\,\mathrm{d}R_2' \\ \leq\, &\int_{\mathcal{A}_{r'}} e^{\beta u(|R_1'|)}\Bigl|\chi_\Lambda\bigl(\tfrac{R_1'+R_2'}{2}, \tfrac{R_2'-R_1'}{2}\bigr) \,-\, \chi\bigl(\tfrac{R_1'+R_2'}{2}, \tfrac{R_2'-R_1'}{2}\bigr)\Bigr|\,\mathrm{d}R_2' \\ &\quad + 2\int_{\mathbb{R}^3\setminus\mathcal{A}_{r'}}\Bigl(X\bigl(\tfrac{R_1'+R_2'}{2}\bigr) + X\bigl(\tfrac{R_2'-R_1'}{2}\bigr)\Bigr)\,\mathrm{d}R_2' \\ \leq\, &\int_{\mathcal{A}_{r'}} e^{\beta u(|R_1'|)}\Bigl|\chi_\Lambda\bigl(\tfrac{R_1'+R_2'}{2}, \tfrac{R_2'-R_1'}{2}\bigr) \,-\, \chi\bigl(\tfrac{R_1'+R_2'}{2}, \tfrac{R_2'-R_1'}{2}\bigr)\Bigr|\,\mathrm{d}R_2' \\ &\quad + 32\int_{|R|>r} X(R)\,\mathrm{d}R \,+\, 16\int_{\mathcal{B}_+} X(R)\,\mathrm{d}R \,+\, 16\int_{\mathcal{B}_-} X(R)\,\mathrm{d}R\,, \end{aligned}$$

where $r = (r' - |R_1'|)/2$ and $\mathcal{B}_\pm = \{R : |R \pm R_1'/2| < 1/(2r')\}$. Given a compact set $\Omega \subset \mathbb{R}^3$ and any $\varepsilon > 0$ we can fix r' so large that the sum of the latter three integrals is bounded by $\varepsilon/2$ for every $R_1' \in \Omega$. Moreover, for $R_1' \in \Omega$ and $R_2' \in \mathcal{A}_{r'}$ we can use the compact convergence of χ_Λ to also bound the former integral by $\varepsilon/2$ by choosing $|\Lambda|$ sufficiently large. Thus we have shown that

$$|J(R_1')| \,\leq\, \varepsilon \qquad \text{for all } R_1' \in \Omega\,,$$

provided that $|\Lambda|$ is sufficiently large. In other words, there holds $J \to 0$ as $|\Lambda| \to \infty$, uniformly in Ω, which was to be shown. $\square$

This result, together with the estimates of $\chi_\Lambda^{(3)}$ and $\chi_\Lambda^{(4)}$ in Propositions 4.2 and 4.3, respectively, shows that the double integrals (6.1) and (6.2) have a well-defined thermodynamical limit. In particular, taking into account that the thermodynamical limits of $\rho^{(m)}$ are even and translation invariant functions, we find that

$$\begin{aligned}\big((\partial\rho^{(2)})v\big)(R,0) \,=\, &-\beta\, v(|R|)\rho^{(2)}(R,0) \,-\, 2\beta\int_{\mathbb{R}^3} v(|R'|)\rho^{(3)}(R,0,R')\,\mathrm{d}R' \\ &-\frac{\beta}{2}\int_{\mathbb{R}^3} v(|R'|)\int_{\mathbb{R}^3}\chi^{(4)}(R,0,R'',R''+R')\,\mathrm{d}R''\,\mathrm{d}R'\,,\end{aligned} \tag{6.9}$$

with all integrals converging absolutely.

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