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by

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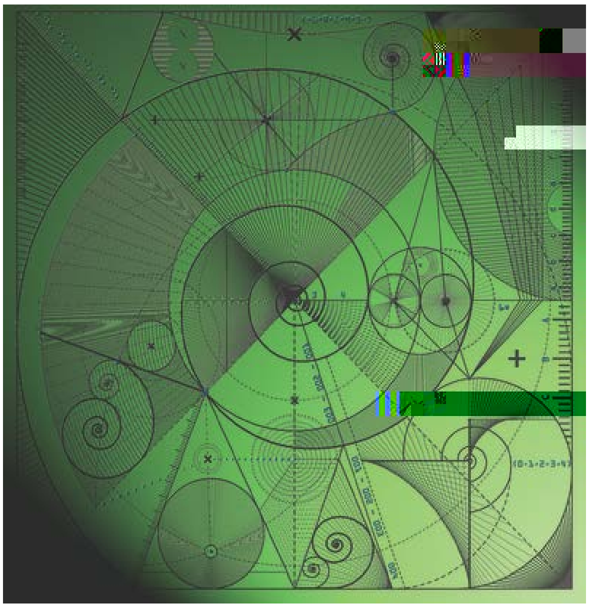

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CONVERGENCE OF DENSITY EXPANSIONS OF CORRELATION FUNCTIONS AND THE ORNSTEIN-ZERNIKE EQUATION

TOBIAS KUNA AND DIMITRIOS TSAGKAROGIANNIS

Abstract. We prove absolute convergence of the multi-body correlation functions as a power series in the density uniformly in their arguments. This is done by working in the context of the cluster expansion in the canonical ensemble and by expressing the correlation functions as the derivative of the logarithm of an appropriately extended partition function. In the thermodynamic limit, due to combinatorial cancellations, we show that the coefficients of the above series are expressed by sums over some class of two-connected graphs. Furthermore, we prove the convergence of the density expansion of the "direct correlation function" which is based on a completely different approach and it is valid only for some integral norm. Precisely, this integral norm is suitable to derive the Ornstein-Zernike equation. As a further outcome, we obtain a rigorous quantification of the error in the Percus-Yevick approximation.

1. Introduction

Correlation functions of interacting particle systems provide important information of the macroscopic as well as the microscopic properties of the system. This was well captured already in the literature in the 30's, see [21]. Around the same period, with the development of power series expansions by Mayer and his collaborators, [29], a direct perturbative representation of correlation functions in terms of integrals over configurations associated to a graphical expansion has been suggested in [30], where the density expansion of the n-body correlation function has been derived. However, being perturbative expansions around the ideal gas, the density expansions of the correlation functions are not expected to be valid at the densities of the liquid regime. So, one tries to "develop a theory of classical fluids without using the density expansion formulas" [34].

A candidate for deriving such relations is the original Ornstein-Zernike (OZ) equation, [37], which, however, cannot be solved as an equation as it contains two unknown quantities, namely the correlation function and the direct correlation function. Hence one has to postulate a relation between them, that is what one calls a closure scheme. A lot of effort has been made in this direction and various suggestions have appeared. In [49], G. Stell systematically relates the most popular closure schemes (such as the Born-Green-Yvon (BGY) hierarchy, [6, 52], the Hyper-Netted Chain (HNC) and the Percus-Yevick (PY) equation [40]) to graphical expansions and tries to quantify them in this way. Ever since an enormous body

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of works was created, which by now is a standard tool in liquid state theory, see e.g. [17].

Let us discuss the points raised above in more detail. The first mathematically rigorous construction of the correlation functions in the thermodynamic limit was obtained in the high temperature and low density regime in [4] based on a fixed point argument for the Kirkwood-Salsburg (KS) equations. Then, further progress has been made in the 60's starting with the works of Groeneveld, Penrose and Ruelle. Ruelle used in [47] a fixed-point argument, while Penrose uses an iteration of the (KS) equations in [38]. Closely related, in [39] Penrose introduced the so-called tree-graph estimate, further developed in [7], cf. also [43] and references therein for recent progress. After the 60's, the technique of cluster expansion has been further developed and its validity has been established for a large class of different systems, for example with the introduction of the abstract polymer model [16, 23]. We refer to [13] for a review of the different sufficient conditions for convergence. For the case of the classical gas, all results are based on the grand canonical ensemble as the techniques that have been used exploit the infinite sum over the number of particles. However, in this paper we are considering expansion in the density. The coefficients of this expansion were identified as sums over 2-connected graphs already in the 40's, cf. [29].

In order to derive from the expansion in the activity an expansion in the density, two further steps are required, as in any resumming: first, some \inversion" theorem from analytic function theory in order to show the convergence of the density expansion and second a combinatorial relation between graphs, e.g. a \topological reduction" in the language of Stell, to identify the coefficients in the density expansions. Part of the latter is to check the admissibility of the rearrangement of terms in the series necessary to realize the combinatorial relations. In general, this is an issue because the series in the graphs is only conditionally convergent. For Mayer's combinatorial identities [29] this is not an issue due to the iterative structure leading in each order to finite many identities. For details as well as for a lower bound on the radius of convergence, see [25]. See also [42] for a recent improvement mainly for potentials with negative part. For the multi-species case see [51] as well as [20]. In [27] this relation between graphs is put in the systematic context of operations between combinatorial \species".

In [25], it was also pointed out that one can derive the convergence of the correlation functions following similar arguments as for the free energy. The coefficients $a_k^{(n)}$ of the density expansion of the truncated correlation functions $u^{(n)}(q_1, \dots, q_n) = {}^{n} \sum_{k=0}^{\infty} {}^{k} a_k^{(n)}(q_1, \dots, q_n)$ are themselves functions of the position. A straightforward application of the arguments from [25] gives only convergence for fixed $q_1, \dots, q_n$. However, in order to work with the expansion, e.g. in order to show that it satisfies the Ornstein-Zernike equation, one needs that the series $_k a$ $(q_1$7::01.4Td [()]-2P is absolutely convergent with respect to the uniform norm in the arguments or the L^1-norm, that isP the Fourier transform of the series in density with the series of the Fourier transform of its summands.

In this paper, we follow a direct and natural approach to obtain the density expansion starting from the canonical ensemble. In [44] the validity of the cluster expansion in the canonical ensemble has been established for the free energy combining the cluster expansion techniques for abstract polymer model and tree-graph estimates for particle systems. Because of the latter, no significant improvement for the radius of convergence for the virial expansion over the activity expansion was achieved. Extending these techniques, in this paper, we prove the convergence of the density expansions for both the correlation and the direct correlation function working directly in the canonical ensemble. Here, it is worthwhile to note that the direct correlation functions is quite different from the truncated correlation functions. First, there exists no natural graphical expansion for the direct correlation function in the activity. Second, even in the density, we are not aware of a natural expansion of the direct correlation function in infinite volume, but one can define an expansion which is at least correct in the thermodynamical limit. Third, we prove directly the convergence of this expansion which is represented by two connected graphs without using any thermodynamical relations. On(functions)-2i39 regime it is a priori clear that these calculations can be made rigorous. At least in this regime, as a by-product of the validity of the convergence for the expansions proved here, we can evaluate the error

$V(q) = V(\ q)$. A potential V is called stable, whenever there exists $B \ 0$ such that:

$$\sum_{1 \ i<j \ N} V(q_i \ q_j) \ BN; \tag{2.1}$$

for all N and all $q_1, \dots, q_N$. In particular, bounded below. A potential V is called regular, whenever

$$C(\ ) := \int_{\mathbb{R}^d} |e^{\ V(q)} \ 1| dq < \infty \ : \tag{2.2}$$

The latter condition holds for a potential bounded below if and only if $\int_{\mathbb{R}^d} |V(q) \wedge 1| dq < \infty$. The hard-core potential fulfills all these assumptions with $C(\ ; R) = |B_R(0)|$, the volume of the ball with radius the interaction range R.
The energy of the system H is defined as

$$H(q) := \sum_{1 \ i<j \ N} V(q_{i,j} \ i<j$$

This is the generating functional of the correlation functions associated to the canonical ensemble. In fact, by expanding the product in (2.7) we obtain

$$L_B(\phi) = \sum_{n=0}^{N} \frac{1}{n!} \int_{\Lambda^n} \phi(q_1)\cdots\phi(q_n)\, \rho^{(n)}_{\Lambda;N}(q_1,\dots,q_n)\, dq_1\dots dq_n, \tag{2.8}$$

where for $n \leq N$ and the points $q_1,\dots,q_n \in \Lambda$ we have defined the n-point correlation function in the canonical ensemble $\rho^{(n)}_{\Lambda;N}(q_1,\dots,q_n)$ as:

$$\rho^{(n)}_{\Lambda;N}(q_1,\dots,q_n) := \frac{1}{(N-n)!} \int_{\Lambda^{N-n}} dq_{n+1}\dots dq_N \frac{1}{Z_{\beta,\Lambda,N}} e^{-\beta H_\Lambda(q)}. \tag{2.9}$$

Note that $\rho^{(0)}_{\Lambda;N} = 1$ and $\rho^{(1)}_{\Lambda;N} = \frac{N}{|\Lambda|}$. Thus, in the thermodynamic limit we obtain $\rho^{(1)} = \rho$. The existence of the thermodynamic limit $\rho^{(n)}$ for $n \geq 2$, that is the limit when $|\Lambda| \uparrow \infty$ with $N = \lfloor \rho|\Lambda| \rfloor$, is more subtle than for thermodynamic quantities like pressure and free energy which are on a logarithmic scale. Analogous results in the grand-canonical ensemble are well-established [46, 47]. Furthermore, for small values of the activity, the correlation functions can be represented as power series in the activity. A by-product of our analysis below is that we also establish the convergence of the thermodynamic limit in the high-temperature-low-density regime in the canonical ensemble. The only related previous result we are aware of is [5].

The logarithm of the Bogoliubov function

$$\log L_B(\phi) =: \sum_{n\geq 1} \frac{1}{n!} \int_{\Lambda^n} \phi(q_1)\cdots\phi(q_n)\, u^{(n)}_{\Lambda;N}(q_1,\dots,q_n)\, dq_1\dots dq_n, \tag{2.10}$$

is the generating function for $u^{(n)}_{\Lambda;N}(q_1,\dots,q_n)$, the sequence of truncated correlation functions or Ursell functions. Relation (2.10) can be understood as the definition of $u^{(n)}_{\Lambda;N}(q_1,\dots,q_n)$. These are the analogues of the cumulants for the sequence of correlation functions. The correlation functions and the Ursell functions can be

holds also in L^1. In order to obtain an explicit description of the limiting $h^{(n)}$, we need an explicit asymptotic expression for $B_{;}$ $(n; m; k)$ in terms of a graphical representation. The resulting expression for $h^{(n)}$ was already shown in [30], [33] and [48], and a proof for pointwise convergence was sketched in [25]. First, we introduce some concepts from combinatorics and graph theory. We also denote by $f_{i;j} := e^{V(q_i \quad q_j)} \quad 1$ Mayer's f-function. Partially following [27] we define:

Definition 2.3. A <u>(simple) graph</u> is a pair $g := (V(g); E(g))$, where $V(g)$ is the set of <u>vertices</u> and $E(g)$ is the set of <u>edges</u>, with $E(g) \quad f\,U \quad V(g) : jUj = 2g$, $j \quad j$ denoting the <u>cardinality</u> of a set. A graph $g = (V(g); E(g))$ is said to be <u>connected</u>, if for every pair $A; B \quad V(g)$ such that $A \,[\, B = V(g)$ and $A \setminus B = ?$, there is an edge $e \; 2 \; E(g)$ such that $e \setminus A \; 6= \; ?$ and $e \setminus B \; 6= \; ?$. Singletons are considered to be connected. We use C_V to denote the set of connected graphs on the set of vertices V $[N]$, where we use the notation $[N] := f\,1; \dots; N\,g$.

Definition 2.4. A <u>cutpoint</u> of a connected graph g is a vertex of g whose removal (with the attached edges) yields a disconnected graph. A connected graph is called <u>2-connected</u> if it has no cutpoint. A <u>block</u> in a simple graph is a maximal 2-connected subgraph. The block-graph of a graph g is a new graph whose vertices are the blocks of g and whose edges correspond to a pair of blocks having a common cutpoint.

Cutpoints are frequently also called <u>articulation points</u>. In this article, we reserve the latter notion for the following slightly more general concept. We use this terminology in order to stay close to Stell's seminal presentation [48] of these graphical constructions.

Definition 2.5. Let $k \; 2 \; N$, $n \; 2 \; N_0$. We consider graphs with $n + k$ vertices, of which the first n vertices are singled out and for simplicity we call them \white". All other vertices are considered to be \black". The set of all such graphs is denoted by $G_{n;n+k}$. Single vertices are not considered as graphs. Similarly, we denote by $C_{n;n+k}$ the set of all connected graphs on $n + k$ vertices with n white vertices.
A vertex is called <u>articulation</u> vertex if upon its removal the component of which it is part separates into two or more connected pieces in such a way that at least one piece contains no white vertices.
We denote by $B^{AF}_{n;n+k}$ the subset of $C_{n;n+k}$ free of articulation vertices.

The easiest example to distinguish cutpoint from articulation point is the graph: 1 (white) - 2 (black) - 3 (white), which is an articulation free graph, but it is not a 2-connected one, as the vertex 2 is a cutpoint (but not an articulation point).
This concept of articulation vertices free graph is also crucial for the definition of the so-called direct correlation function, see below in (2.27) and (2.28). Motivated by the distinction between an articulationonent of which7F0(as9t(an)-326(articulation)-32ed)-23mponent of which7F0(as9t

We denote by $\mathcal{B}_{n;n+k}$ the set of all connected graphs over n white and k black vertices free of articulation and of nodal vertices. The latter coincides with the collection of all two-connected graphs on $n+k$ vertices with n white vertices.

The nodal points are exactly the cutpoints of a graph that are not articulation points. For a graph $g \in \mathcal{G}_{n;n+k}$ we define the activity

$$\tilde{w}_{\Lambda}(g;\{1,\dots,n\}) := \int \prod_{i=1}^{n+k} \frac{dq_i}{|\Lambda|} \prod_{\{i,j\}\in E(g)} f_{i,j} \prod_{i\in\{1,\dots,n\}} \varphi(q_i); \tag{2.22}$$

as well as its version without the test function φ, but with dependence on a fixed configuration $q_1,\dots,q_n$:

$$\tilde{w}_{\Lambda}(g;q_1,\dots,q_n) := \int_{\Lambda^k} \prod_{j=n+1}^{n+k} dq_j \prod_{\{i,j\}\in E(g)} f_{i,j}; \tag{2.23}$$

where $f_{i,j} := e^{-\beta V(q_i-q_j)} - 1$. If φ is compactly supported around some point in Λ, then $\tilde{w}_{\Lambda}$ scales as $|\Lambda|^{-n-k}$ while $\tilde{w}_{\Lambda}$ is of order one. Note also that in this paper we tend to denote with a ~ all quantities that depend on the positions $q_1,\dots,q_n$. Now we are ready to state the theorem about the existence of the infinite volume limit of (2.13):

Theorem 2.7. There exists a constant $c_0 > 0$ such that for all $C(\beta) < c_0$ we have:

$$h^{(n)}(q_1,\dots,q_n) := \lim_{\substack{\Lambda\uparrow\mathbb{R}^d; N\to\infty;\\ N=\lfloor\rho|\Lambda|\rfloor}} h^{(n)}_{\Lambda;N}(q_1,\dots,q_n) = \sum_{k\geq 0} \rho^k \frac{1}{n!k!} \sum_{g\in\mathcal{B}^{AF}_{n;n+k}} \tilde{w}(g;q_1,\dots,q_n); \tag{2.24}$$

where

$$\tilde{w}(g;q_1,\dots,q_n) := \lim_{\Lambda\uparrow\mathbb{R}^d} \tilde{w}_{\Lambda}(g;q_1,\dots,q_n) = \int_{\mathbb{R}^{dk}} \prod_{j=n+1}^{n+k} dq_j \prod_{\{i,j\}\in E(g)} f_{i,j}. \tag{2.25}$$

Moreover, at infinite volume, we have the following bound:

$$\sup_{q_1} |$$

for fixed volume and number of particles $N+2$:

$$c^{(2)}_{;N+2}(q_1;q_2) := \sum_{k=0}^{N} \frac{{}^{k}}{k!} \sum_{g\in B_{2;2+k}} \tilde{\ }(g;q_1;q_2): \tag{2.27}$$

Then we have the following theorem:

Theorem 2.9. There exists a constant $c_0 > 0$ such that for all $C(\ ) < c_0$, the direct correlation function $c^{(2)}_{;N+2}$ in (2.27) converges in the thermodynamic limit, to

$$c^{(2)}(q_1;q_2) := \sum_{k=0}^{\infty} \frac{{}^{k}}{k!} \sum_{g\in B_{2;2+k}} \tilde{\ }(g;q_1;q_2); \tag{2.28}$$

which is an analytic function in , for $C(\ ) < c_0$. Furthermore, the series (2.28) converges in the following sense:

$$\sup_{q_1\in} \int dq_2 \frac{{}^{k}}{k!} \sum_{g\in B_{2;2+k}} \tilde{\ }(g;q_1;q_2) \quad C e^{ck}; \tag{2.29}$$

uniformly in .

Furthermore, the direct correlation function $c^{(2)}_{;N+2}$ in (2.27) fulfils the Ornstein-Zernike equation (2.16) up to the order $O(1/|\ |)$ and the limit function fulfils the Ornstein-Zernike equation (2.16).

This follows from the fact that

$$\mathcal{L}_B(\psi) = \frac{Z_{\beta,\Lambda,N}(\psi)}{Z_{\beta,\Lambda,N}(0)}, \qquad Z_{\beta,\Lambda,N}(0) \equiv Z_{\beta,\Lambda,N}.$$

We define the space $\mathcal{V}_N$ whose elements are all ordered pairs (V, A) where $V \subset \{1, \dots, N\}$ and $A \subset V$. We say that two elements (V_1, A_1) and (V_2, A_2) are compatible, and denote it by $(V_1, A_1) \sim (V_2, A_2)$, if and only if $V_1 \sim V_2$, where two sets V_1, V_2 are called compatible (denoted by $V_1 \sim V_2$) if $V_1 \cap V_2 = \emptyset$; otherwise we call them incompatible ($\nsim$).
Then we split (3.2) as

$$Z_{\beta,\Lambda,N}(\psi) = \frac{|\Lambda|^N}{N!} Z^{\mathrm{int}}_{\beta,\Lambda,N}(\psi) \tag{3.3}$$

and write

$$Z^{\mathrm{int}}_{\beta,\Lambda,N}(\psi) = \sum_{\{(V_1,A_1),\dots,(V_k,A_k)\}} \prod_{i=1}^{k} \zeta((V_i, A_i)), \tag{3.4}$$

where

$$\zeta((V, A)) := |\Lambda|^{|A|} \sum_{g \in \mathcal{C}_V} \tilde{\varphi}(g; A), \qquad \tilde{\varphi}(g; A) := \int \frac{dq_V}{|\Lambda|^{|V|}} \prod_{\{i,j\} \in E(g)} f_{i,j} \prod_{i \in A} \psi(q_i), \tag{3.5}$$

with the latter as already defined in (2.22), and dq_V is a shorthand for the product measure $\prod_{i \in V} dq_i$. In the literature, see [2], this is called subset gas and it is a special case of the general Abstract Polymer Model, which consists of (i) a set of polymers $\mathcal{V}_N$, (ii) a binary symmetric relation $\sim$ of compatibility between the polymers (i.e., on $\mathcal{V}_N \times \mathcal{V}_N$) and (iii) a weight function $\zeta : \mathcal{V}_N \to \mathbb{C}$. We also define the compatibility graph $\mathcal{G}_{\mathcal{V}_N}$ to be the graph with vertex set $\mathcal{V}_N$ and with an edge between two polymers (V_i, A_i) and (V_j, A_j) if and only if they are an incompatible pair. In this framework we have the following formal relation for the logarithm, which will be justified rigorously in Theorem 3.1 below (see [23]):

$$\log Z^{\mathrm{int}}_{\beta,\Lambda,N}(\psi) = \log\left(\sum_{\{(V_1,A_1),\dots,(V_k,A_k)\}} \prod_{i=1}^{k} \zeta((V_i, A_i)) \right) = \sum_{I \in \mathcal{I}(\mathcal{V}_N)} c_I \, \zeta^I, \tag{3.6}$$

where

$$c_I = \frac{1}{I!} \sum_{G \subset \mathcal{G}_I} (-1)^{|E(G)|}. \tag{3.7}$$

The sum in (3.6) is over the set $\mathcal{I}(\mathcal{V}_N)$ of all multi-indices $I : \mathcal{V}_N \to \{0, 1, \dots\}$. We use the shortcut $\zeta^I := \prod_{(V,A)} \zeta((V, A))^{I((V,A))}$, but for notational simplicity in stating the main theorem of cluster expansion, we use the notation $\gamma := (V, A)$ for the generic polymer consisting of the ordered pair $(V, A) \in \mathcal{V}_N$. Then, defining $\operatorname{supp} I := \{\gamma \in \mathcal{V}_N : I(\gamma) > 0\}$, we denote by $\mathcal{G}_I$ the graph with $\sum_{\gamma \in \operatorname{supp} I} I(\gamma)$ vertices induced from the restricted $\mathcal{G}_{\mathcal{V}_N}$ in $\operatorname{supp} I$, by replacing each vertex γ by the complete graph on $I(\gamma)$ vertices. Furthermore, the sum in (3.7) is over all connected subgraphs G of $\mathcal{G}_I$ spanning the whole set of vertices of

G_I and $I! := \prod_{\in \mathrm{supp}\, I} I(\ )!$. Note that if I is such that G_I is not connected (i.e., I is not a cluster) then $c_I = 0$.

We state the general theorem for the special case of the subset gas, following [23] to which we refer for the proof.

Theorem 3.1 (Cluster Expansion). Assume that there are two non-negative numbers a, c 0 such that

$$\sum_{(V;A):\, V \ni 1} |\ ((V;A))|e^{a|V|+c|V|} \quad a: \tag{3.8}$$

Then, for every polymer $(V^0;A^0) \in \mathcal{V}_N$, we obtain that

$$\sum_{I:\, I((V^0;A^0)) \quad 1} |c_I \ ^I| e^{\sum_{(V;A)\in \mathrm{supp}\, I} I((V;A))\, c|V|} \quad |\ ((V^0;A^0))|e^{a|V^0|+c|V^0|}; \tag{3.9}$$

where the coefficients c_I are given in (3.7).

Proof of Theorem 2.1: From (3.1), (3.2) and by representing the partition function by the subset gas, we first check the validity of the convergence condition (3.8) of Theorem 3.1. In order to bound the activity $((V;A))$ we use the tree-graph inequality (see the original references [28], [39], [7]; here we use the particular form given in [41], Proposition 6.1 (a)):

]TJ/F29 11.9552 Tf 6.7233 Td [(2)e003

[(1]TJ/F46 11.9146 65 025.1.52(+)]TJkF30 5.9(A)]TJ/F15 11 T97 0 Td [(V)]36F32 7.TJ/F31 11.9552 T92 0 Td [(Vk)

corresponding term in $B_{\ ;\ }$ is of the order of:

$$| \ |^{|V_1|+|V_2|+|V| \quad 2}\frac{1}{| \ |^{|V_1|}}\frac{1}{| \ |^{|V_2|}}\frac{1}{| \ |^{|V| \quad 1}} = \frac{1}{| \ |}:$$

Hence, the structure of the leading term at the level of the multi-indices is quite simple: only one polymer, call it $(V_0;A_0)$ has $A_0 \neq \emptyset$. Then, for all other polymers with $A = \emptyset$ we can have a further structure as explained below (and as in [44]). Since it is always true that the total number of labels $(m+k)$ should satisfy $m + k \quad \sum_{V\in \operatorname{supp} I}(|V| \quad 1) + 1$ (due to the fact that each $(V;A)$ should be incompatible with at least one of the other polymers, i.e., have at least one common label and $V_0 \cup \bigcup_{(V;A)\in \operatorname{supp} I;\, V\neq V_0} V = [m+k]$), overall we have:

$$I((V;A)) = 1;\ \forall (V;A) \in \operatorname{supp} I; \quad \text{and} \tag{4.1}$$

$$m + k = |V_0| + \sum_{(V;A)\in \operatorname{supp} I;\, V\neq V_0} (|V| \quad 1): \tag{4.2}$$

Hence, we restrict the summation over multi-indices in this subclass satisfying properties (4.1), (4.2) and containing only one polymer $(V_0;A_0)$ with $A_0 \neq \emptyset$. We denote this fact by adding a superscript at the sum as e.g. in (4.4) below. The polymers $(V;\emptyset)$ can be attached to the polymer with $A \neq \emptyset$ either on a vertex not in A (a black circle in the terminology of Stell) or in a vertex in A (a white circle in the terminology of Stell). In order to visualize the last case, we give the following example: consider the following multi-index I: I is equal to one on the two polymers $(\{1;2\};\{1;2\})$ and $(\{1;3\};\emptyset)$, zero otherwise. The two polymers intersect in the label 1. We have:

$$\begin{aligned}
| \ |^{3 \quad I} &= | \ |^{3 \quad 2}\int (q_1)\ (q_2) f_{1;2}(q_1 \quad q_2)\frac{dq_1}{| \ |}\frac{dq_2}{| \ |}\int f_{1;3}(q_1 \quad q_3)\frac{dq_1}{| \ |}\frac{dq_3}{| \ |}\\
&= \int_{\ ^2} (q_1)\ (q_2) f_{1;2}(q_1 \quad q_2)dq_1dq_2\ \frac{1}{| \ |}\int f_{1;3}(q_1 \quad q_3)dq_1dq_3:\\
&= \int_{\ ^2} (q_1)\ (q_2) f_{1;2}(q_1 \quad q_2)dq_1dq_2 \int f_{1;3}(q_3)dq_3:
\end{aligned}$$

As we will explain later, this term will be canceled by one summand from the term $| \ |^{3 \quad I'}$, with I' being the multi-index which is one only on the polymer $(\{1;2;3\};\{1;2\})$ and in particular with the summand in $| \ |^{3 \quad I'}$ which is associated with the graph on $\{1;2;3\}$ with exactly two edges $\{3;1\}$ and $\{1;2\}$. Let us start with the formal proof for these cancelations.

Proof of Theorem 2.7: Following the discussion above, we split $B_{\ ;\ }$ from (3.14) as follows:

$$B_{\ ;\ }(n;m;k) = B_{\ ;\ }(n;k)\ _{n;m} + R_{\ ;\ }(n;m;k); \tag{4.3}$$

where

$$B_{\ ;\ }(n;k) := \frac{| \ |^{(n+k)}}{n!k!}\sum_{I:A(I)=[n+k]} c_I \ ^{I} \tag{4.4}$$

and

$$A(I) := \cup_{V\in \operatorname{supp} I} V: \tag{4.5}$$

Recall that the superscript indicates that the sum is over all multi-indices that satisfy properties (4.1), (4.2) and that contain only one polymer with $A \neq \emptyset$, for which we have already chosen its labels and we call it $A_0 := \{1, \dots, n\}$. For this reason, we can now consider multi-indices in $I(V_{n,k})$, where the class $V_{n,k}$ consists of all subsets of the labels corresponding to the white vertices $\{1, \dots, n\}$ and the black vertices $\{n+1, \dots, n+k\}$. The new polymers either they contain A_0 or they intersect it at most one point. Therefore, in the new set-up with $I \in I(V_{n,k})$ the conditions (4.1) and (4.2) can be rewritten as

$$I(V) = 1, \ \forall V \in \operatorname{supp} I, \quad \text{and} \tag{4.6}$$

$$n + k = |V_0| + \sum_{V \in \operatorname{supp} I,\, V \neq V_0} (|V| - 1), \tag{4.7}$$

where7a47ere774nV

hence we need to prove convergence of the cluster expansion with activities being functions of $q_1; : : : ; q_n$ in an appropriate norm. From (2.19) using the splitting (4.3) we have:

$$F_{;\ ;N}(n;k) = P_{N;j\ j}(n+k)B_{;}\ (n;k) + \sum_{m=1}^{n\ 1} P_{N;j\ j}(m+k)R_{;}\ (n;m;k); \qquad (4.9)$$

where the second term is vanishing in the limit " R^d. Substituting in (2.18) we obtain:

$$\int_{n} \prod_{i=1}^{n} (dq\ \ (q_i))\ {}^{n}h^{(n)}_{;N}(q_1; : : : ; q_n) =$$

$$\sum_{k\ \ 0} P_{N;j\ j}(n+k)\frac{1}{n!k!} \sum_{g2B^{AF}_{n;n\ +k}} \int_{n+k} \prod_{j=1}^{n+k} dq \prod_{f\,i;j\ g2E(g)} f_{i;j} \prod_{i=1}^{n} (q_i)$$

$$+ \sum_{k\ \ 0} \sum_{m=1}^{n\ 1} P_{N;j\ j}(m+k)R_{;}\ (n;m;k): \qquad (4.10)$$

Then, having the bounds (2.20) and (4.8) we can take the thermodynamic limit on the right hand side of (4.10) and obtain:

$$\sum_{k\ \ 0}\ {}^{n}\ {}^{k}B^{k}_{q}$$

Notice that one of them, b_0 (without loss of generality), contains all white vertices with labels in A_0. We denote by $F(g)$ the collection of all $F \subset B(g)$ such that $\bigcup_{b\in F} b$ is a connected graph, where we use the notation $\bigcup_{b\in F} b := (\bigcup_{b\in F} V(b); \bigcup_{b\in F} E(b))$ for the union of graphs. We also define $H(g)$ to be the collection of all such graphs:

$$H(g) := \{ g' : g' = \bigcup_{b\in F} b; F \in F(g)\}. \tag{4.13}$$

Similarly,

$$A(g) := \{ V(g'); g'$$

even easier since we win a power of j j because of non translation invariance, therefore we refrain from repeating the proof here and we refer the reader to [44].

Thus, since we know that in (4.15) the component b_0 has to appear in each summand and since by Lemma 4.3 there should be only one component, then the only non-zero contribution comes from the articulation vertex free component, i.e., $g\ 2\ B^{AF}_{n;n+k}$. In other words, we have that for every $g\ 2\ C_{n;n+k} \backslash\ (B^{AF}_{n;n+k})^c$,

$$\sum_{\substack{l\,:\,\mathrm{supp}\, l\ \ A\ (g);\ A(l)=V \\ jV \backslash\ V^0 j=1\ ;8V;V^0 2\ \mathrm{supp}\, l}} c_l = 0 \tag{4.17}$$

and $= 1$, otherwise. Notice the difference with respect to [44]: here, the element $b_0\ 2\ B(g)$ as it appears in $A(g)$ (via $H(g)$, defined above) is special and consists of articulation free graphs in their new definition within the presence of \white" vertices. This concludes the proof of Lemma 4.1.

Proof of Lemma 4.2. Recall the use of the shortcut l^0

as each of the polymers $V_1; \ldots ; V_r$ intersects with V_0 at most at one label. Alluding to the constraints (4.6) and (4.7) we split the integral as follows:

$$\int_{\Lambda^k}\prod_{j=1}^{k} dq_{n+j}\,\frac{1}{|\Lambda|^{n+k}}\sum_{\substack{g\in\mathcal{C}_{n;n+k}:\\ g\ \ l}}\ \prod_{\{i,j\}\in E(g)} f_{i,j}\qquad\frac{1}{|\Lambda|^{n}}\prod_{j=0}^{r}\ \{V\ \wedge\ (V$$

some correlation function due to the absolute value in (4.20). Thus, from (4.21), using (3.9), we obtain that

$$\sum_{\substack{l\,2I\,(V_{n;N}\quad n)\\ jA(l)nA_0j=k}} jc_l\,jj^{\wedge}\,j^{l}\quad e^{\;ck}\qquad \sum_{\substack{l\,2I\,(V_{n;N}\quad n)\\ jA(l)nA_0j=k}} jc_l\,jj^{\wedge}\,j^{l}\,e^{ck}\quad Ce^{\;ck}; \tag{4.25}$$

for some $C > 0$ as in (3.15), depending on n.

5. Direct correlation function, proof of Theorem 2.9

Using (2.18), Theorem 2.7 and definition (2.23) the leading order of the second Ursell function can be expressed as follows:

$$\int_{2} dq_1dq_2\ \ (q_1)\ \ (q_2)u^{(2)}_{;N}(q_1;q_2) =$$

$$\int_{2} dq_1dq_2\ \ (q_1)\ \ (q_2)\sum_{k\ \ 0} P_{N;j\ \ j}(2+k)\frac{1}{2!k!}\sum_{g2B^{AF}_{2;2+k}}\tilde{\ }\ (g;q_1;q_2) + O\left(\frac{1}{j\ \ j}\right)\ : \tag{5.1}$$

In order to derive the Ornstein-Zernike equation in the canonical ensemble, we split the graphs in the right hand side of (5.1) at the nodal points (recall Definition 2.6). These are the points through which pass all paths joining q_1 to q_2, hence we can order them. Given $g\ 2\ B^{AF}_{2;2+k}$, we choose the first nodal point starting from q_1 and call its label j. Note that by the definition of articulation points, $j \not= 1;2$. Upon the removal of this point the graph g splits into two connected components: g_1 with $l+2$ vertices and g_2 with $k\quad l+1$ vertices with the only common vertex being the one with label j. Note that g_1 contains q_1 and g_2 contains q_2. Since q_j is the location of a nodal point, we can write

$$\tilde{\ }\ (g;q_1;q_2) = \int dq\ \ \tilde{\ }\ (g_1;q_1;q_j)\ \tilde{\ }\ (g_2;q_j;q_2):$$

Then, the leading term in (5.1) yields

$$\int_{2} dq_1dq_2\ \ (q_1)\ \ (q_2)\sum_{k=0}^{N\ \ 2} P_{N;j\ \ j}(2+k)\frac{1}{2!k!}\left[\ \sum_{g2B}\ \int\ \ (g\ \ q \ldots\ \ j\ \ k=\ \ \int\ {}^{k}(g_2;q\ \ q_j + \ \ j=\ \ (g_2;q\ ;q_2\right.$$

(2 Tf 10.821 0 Td [(k)]2]01TJ/F35 7.9701 Tf 4.621 0 Td

We rewrite this in such a way that direct two-point correlation function (uniquely defined up to leading order) as given in (2.27) appear. By choosing the label $|\ | = 3$ in (5.2) we obtain

$$\int_{\ ^2} dq_1 dq_2 \frac{1}{2}\ (q_1)\ (q_2) \left[\sum_{k=0}^{N-2} P_{N;|\ |}(2+k)\frac{1}{k!}\sum_{g\in\mathcal{B}_{2;k+2}} \tilde{\ }(g;q_1;q_2) + \right.$$

$$\left. + \sum_{k=0}^{N-2} P_{N;|\ |}(2+k)\sum_{l=0}^{k-1}\int dq_3 \frac{1}{l!}\sum_{g_1\in\mathcal{B}_{2;l+2}} \tilde{\ }(g_1;q_1;q_3)\frac{1}{(k-1-l)!}\sum_{g_2\in\mathcal{B}^{AF}_{2;k-l+1}} \tilde{\ }(g_2;q_3;q_2)\right]:$$

By using new labels $l_1 := l$ and $l_2 := k-1-l$, the last summand can be rewritten as follows

$$\sum_{l_1=0}^{N-3}\int dq_3 \frac{1}{l_1!}\sum_{g_1\in\mathcal{B}_{2;l_1+2}} \tilde{\ }(g_1;q_1;q_3)\sum_{l_2=0}^{N-3-l_1} P_{N;|\ |}(l_1+l_2+3)\frac{1}{l_2!}\sum_{g_2\in\mathcal{B}^{AF}_{2;l_2+2}} \tilde{\ }(g_2;q_3;q_2): \qquad (5.3)$$

Let us introduce the following shorthands

$$C\ (2;l_1+2;q_1;q_3) := \frac{1}{l_1!}\sum_{g_1\in\mathcal{B}_{2;l_1+2}} \tilde{\ }(g_1;q_1;q_3) \qquad (5.4)$$

and

$$B\ (2;l_2+2;q_3;q_2) := \frac{1}{l_2!}\sum_{g_2\in\mathcal{B}^{AF}_{2;l_2+2}} \tilde{\ }(g_2;q_3;q_2): \qquad (5.5)$$

Then we can rewrite (5.3) as

$$\int dq_3 \sum_{l_1=0}^{N-3} P_{N;|\ |}(l_1+1)\,C\ (2;l_1+2;q_1;q_3)$$

$$\sum_{l_2=0}^{N-3-l_1} \frac{P_{N;|\ |}(l_1+l_2+3)}{P_{N;|\ |}(l_1+1)\,P_{N;|\ |}(l_2+2)} P_{N;|\ |}(l_2+2)\,B\ (2;l_2+2;q_3;q_2); \qquad (5.6)$$

Lemma 5.1. Under the hypothesis of the previous theorems, the function $c^{(2)}_{;N}$ defined in (2.27) fulfills the Ornstein-Zernike equation to leading order in the following sense:

$$\begin{aligned}\int_{2} (q_1)\,(q_2)u^{(2)}_{;N}(q_1;q_2)\,dq_1dq_2 &= {}^{2}\int_{2} (q_1)\,(q_2)c^{(2)}_{;N}(q_1;q_2)\,dq_1dq_2\\ &+\int_{2} (q_1)\,(q_2)\int c^{(2)}_{;N}(q_1;q_3)u^{(2)}_{;N}(q_3;q_2)dq_3\;\;dq_1dq_2\\ &+O\;\frac{1}{j\;\;j}\;:\end{aligned}\tag{5.7}$$

Proof.

In order to show that the above bound is of order $O(1=j \; j)$, one notes that both summands contain the following two factors which are tails of the corresponding convergent series:

$$\sup_{q_2;q_3} \sum_{l=N+1}^{1} P_{N;j \; j}(l+2) \; B \; (2; l+2; q_3; q_2) \quad Ce^{\; cn} \tag{5.10}$$

and

$$\sup_{q_1} \sum_{l=N+1}^{1} P_{N;j \; j}(l+1) \int dq_3 \; C \; (2; l+2; q_1; q_3) \quad Ce^{\; cn}; \tag{5.11}$$

for some constants $C; c > 0$. The rst follows from the bound in (4.12), while the second is claimed in (2.29) and proved in the next lemma.

The second result is about the convergence and integrability of $c_N^{(2)}(q_1; q_2)$ as $N \; ! \; 1$. In order to take the limit in (5.7) and get the in nite volume version of the OZ equation, we need to prove (2.29) which is given in the following lemma:

Lemma 5.2. For some positive constants C and c independent of N and and for every $l_1 \; 2 \; N$ and $q_1 \; 2$ we have that

$$P_{N;j \; j}(l_1+1) \int dq_2 \; C \; (2; l_1+2; q_1; q_2) \quad C \; e^{\; cl_1}; \tag{5.12}$$

for large enough.

Remark 5.3. As it will be clear in the proof, for the above estimate to hold it is important that we have an integral in q_2, that is an integral over the variable corresponding to the second white vertex. For short we call it the integrated white vertex.

Proof. The proof follows the line of calculation in Lemma 4.2. The main di erence is that here we do not require that there exists a special polymer V_0 containing both white vertices. Hence we restrict to the class

$$I(V) = 1; \; 8V \; 2 \; \mathrm{supp} I; \quad \text{and} \tag{5.13}$$

$$m + k = \sum_{V \, 2 \, \mathrm{supp} \, I} (jVj \quad 1) + 1 \tag{5.14}$$

and we denote it by using the superscript over the sum, in order to distinguish it from the previous case. Recalling the shortcut g for the class of multi-indic2 Tf 31 11.the11..051 + 1 in (4.15), we have:

$$P_{N;|\ |}(l_1+1)\int dq_2\ C\ (2;l_1+2;q_1;q_2)\ =$$

$$=\ \frac{N(N-1)\ldots(N-(l_1+1)+1)}{|\ |^{l_1+1}}\int dq_2\ \frac{1}{l_1!}\sum_{g\in C_{2;2+l_1}}\tilde{\ }\ (g;q_1;q_2)\sum_{l\ g}c_l$$

$$=\ \frac{N}{|\ |}\binom{N-1}{l_1}\int dq_2\sum_{\substack{l\in I\ (V_{2;l_1})\\ A(l)=[l_1+2]}}c_l\ \frac{1}{|\ |^{l_1}}\sum_{\substack{g\in C_{2;2+l_1}:\\ g\ \ l}}\tilde{\ }\ (g;q_1;q_2)\ : \qquad (5.15)$$

The class $V_{2;l_1}$ consists of all subsets of the labels corresponding to the white vertices $\{1;2\}$ and the black vertices $\{3;\ldots;l_1+2\}$. The class $g\ \ l$ is as before in (4.18). The compatibility graph of the polymers is a connected graph whose blocks are complete graphs (usually called Husimi graphs, see [27, 19]). Within this structure we denote by $V_1;\ldots;V_r$ the chain of pairwise incompatible polymers such that the label $1\in V_1$ and the label $2\in V_r$. Note that r could be equal to 1, but in this case the structure would be exactly as in the previous theorem. We denote by i_j the common label of V_j and V_{j+1}, $j=1;\ldots;r-1$ and by V_s^0, for s from an index set S, the remaining polymers attached to the rest of the structure by the label i_s. Note that by translation invariance the activity associated to V_s^0 does not depend on the label that connects it to F15 11.955;l

in [27], this is just the following combinatorial identity interpreted as formal power series:

$$C_n = (C\ )^n\, B_n^{AF}(C\ ); \tag{6.1}$$

where C_n, B_n^{AF} respectively, denotes the set of connected, articulation point free respectively, graphs with n special vertices. C denotes the set of graphs with one special vertex, but multiplied with the activity.
The case n = 1 has a more difficult structure. Let us derive it in more detail; we have

$$\frac{^{(1)}(q_1)}{z} = \frac{1}{(z)}\sum_{n\ 1}\frac{z^{n\ 1}}{(n\ \ 1)!}\int_{n\ 1} dq_2 \dots dq_n\, e^{\ H\ (q)}; \tag{6.2}$$

where z is the activity and (z) the grand-canonical partition function. Writing $^{\ H\ (q)} = \sum_{g\in G_n}\prod_{\{i,j\}\in E(g)} f_{i,j}$, we split the graph and the integral over the connected components of each graph. Recalling the definition of the activity $\tilde{\ }(g;q_1;\dots;q_n)$ given in (2.23), we get that (6.2) equals to

$$\frac{1}{(z)}\sum_{n\ 1}\frac{1}{(n\ \ 1)!}\sum_{k\ 1}\frac{1}{k!}\sum_{(P_0,\dots,P_k)\in\ (2\ ,\dots,n)}\left(z^{|P_0|}\sum_{g\in C(P_0\cup\{1\})}\tilde{\ }(g;q_1)\right)\prod_{j=1}^{k}\left(z^{|P_j|}\sum_{g\in C(P_j)}\tilde{\ }(g;?)\right)$$

$$= \sum_{n\ 1} n z^n$$

recalling that

$$ {}_{;m} := \frac{1}{m!} \sum_{g \in B_{1;m+1}} \int_{m} \prod_{\{i;j\} \in E(g)} (e^{\ V(q_i \ \ q_j)} \ \ 1)\, dq_2 \dots dq_{m+1}; \quad q_1 \text{ fixed}; \tag{6.6}$$

is the virial coefficient and $F(\ ) := \sum_{m \ 1} \frac{1}{m+1} \ {}_{;m} \ ^{m+1}$.
This is exactly the combinatorial identity given in [27], Theorem 1:1. The above calculation is also one of the motivations to define (following [48]):

$$h^{(1)}(q_1) := \log(\ (q_1)) \quad \log(z) = \sum_{m \ 1} \ {}_m(\ (q_1))^m; \tag{6.7}$$

in the thermodynamic limit. Note that because of translation invariance both $h^{(1)}(q_1)$ and (q_1) are constant. This is also closely related to the Legendre transform giving the equivalence of ensembles between pressure and free energy at the thermodynamic limit:

$$p(z) = \sup \{ \ \log z \quad f(\ )\}; \qquad f(\ ) = \sup_z \{ \ \log z \quad p(z)\}:$$

In the first case the sup is attained at $\log z = f'(\ )$ and hence

$$h^{(1)} = \log \qquad f'(\ ) = F'(\ ); \tag{6.8}$$

where $F(\ ) = \ (\log \quad 1) \quad f(\ )$ is the free energy corresponding to the "interaction" between the particles.

We conclude this section by noting that the OZ equation corresponds to the following easy combinatorial fact. For the second correlation functions the expansion in the density is given by the sum over all graphs free of articulation vertices. Hence the block graph associated to such a graph is actually a chain connecting the two white vertices. The OZ equation is nothing more than an iterative representation of this fact.

7. Application to liquid state theory in the gas regime

The rigorous expansions that we present in this paper can serve as a tool for quantifying the error in existing theories which are extensively used in the liquid state, as well as for suggesting systematic error-improving schemes. However, this is only possible in the gas regime where all these expansions are valid. Extending these results to the liquid state regime is a highly nontrivial problem, if even possible. We give here a first glimpse of this. To start, we recall that the Ornstein-Zernike equation (2.16) is not a closed equation as it involves both correlation functions $h^{(2)}(q_1; q_2)$ and $c(q_1; q_2)$. One suggestion for a closure is the Percus-Yevick (PY) equation [40] that we describe below. Starting from the OZ equation for $h^{(2)}(r)$ and $c(r)$, following [49], one first introduces a new function t as follows:

$$t(r) := c \ \ h^{(2)}(r); \tag{7.1}$$

where we use the convolution: $c \ \ h^{(2)}(r) := \int c(r')h^{(2)}(r \quad r')dr'$. Then the OZ equation taregime(()]TJ/o5h h h h h h

Note that all involved functions ($h^{(2)}$, c and t) are analytic functions in . Furthermore, $c(r)$ can be written as

$$c(r) = f(r)(1 + t(r)) + m(r); \qquad (7.3)$$

where $f(r) := e^{\quad V(r)} \quad 1$ is a known function of the potential $V(r)$. The relation (7.3) is essentially the definition of $m(r)$ which is an analytic function of as well. Following [49] the function m can be expressed as a sum over two connected graphs which upon removal of the direct link f connecting the white vertices (if it is present) it is two-connected (no articulation and no nodal points). For example, the first term of $m(r)$ is the graph 1 3 2 4 1. However, in [49], \the manipulations involved in obtaining these infinite sums ... have been carried out in a purely formal way and we have not examined the important but difficult questions of convergence and the legitimacy of the rearrangement of terms". The present paper establishes this convergence with respect to f-bonds. The convergence allows to quantify the error after truncating these terms. For example, m is of order ^{2}. Furthermore, a future plan is to investigate whether another suggestion could be made, relating some of the terms in $m(r)$ with respect to $t(r)$, or by introducing another function (instead of $t(r)$) as a candidate for a good choice for \closing" OZ equation. Combining (7.1) with (7.2) and (7.3) we obtain:

$$t = [f(1 + t) + m] \quad [f(1 + t) + m] + [f(1 + t) + m] \quad t: \qquad (7.4)$$

One version of PY equation is setting $m(r) \quad 0$ and obtaining a closed equation for $t(r)$. Alternatively, using (7.2) and (7.3) one can introduce the functions $y(r)$ and $d(r)$ by

$$g^{(2)}(r) = e^{\quad V(r)}(1 + t(r)) + m(r) =: e^{\quad V(r)}y(r); \qquad y(r) =: 1 + t(r) + d(r); \qquad (7.5)$$

and hence $m(r) = e^{\quad V} d(r)$. Thus, we can rewrite (7.4) as

$$y \quad = \quad 1 + d + [f\, y + d] \quad [e^{\quad V} y \quad 1]: \qquad (7.6)$$

Again, setting $d(r) \quad 0$ we obtain another version of PY equation. All involved functions are analytic in and our results imply that the formal order in of d coincides with the actual order. Now, one can investigate a method of systematically improving the PY equation, by adding some terms from d (or from m for hard-core potentials). For example, in [49] it was suggested to set d equal to the first order term in its expansion, since this gives a \PY approximation that it leads to an approximateg that is exact through terms of order ^{2} in its virial expansion". A partial goal of the analysis in the present paper is to provide a framework in which one can further investigate such closure schemes and estimate the relevant error.

Other closures include the Hypernetted Chain(HNC) equation, the Born-Green-Yvon (BGY) hierarchy and many others for which we could investigate the validity of the corresponding graphical expansions. We conclude by mentioning that another direction that has attracted considerable interest is the construction of exact solutions of the PY equation, which however usually cannot be expressed as truncations of convergent series. But still, several suggestions have been made for models of rigid spheres; see [10] and the references therein for a comparison of the different methods.

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