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The virial series for a gas of particles with uniformly repulsive pairwise interaction and its relation with the approach to the mean field

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Abstract. The pressure of a gas of particles with a uniformly repulsive pair interaction in a finite container is shown to satisfy (exactly as a formal object) a "viscous" Hamilton–Jacobi (H–J) equation whose solution in power series is recursively given by the variation of constants formula. We investigate the solution of the H–J and of its Legendre transform equation by the Cauchy-majorant method and provide a lower bound to the radius of convergence on the virial series of the fluid which goes beyond the threshold established by Lagrange's inversion formula. Such results obtained in (On the virial series for a gas of particles with uniformly repulsive pairwise interactions (2014) Preprint) are reviewed and regarded as the first step towards the solution of a problem posed by Kac, Uhlenbeck and Hemmer (*J. Math. Phys.* **4** (1963) 216–228), questioning on the relation of the approach to the mean field theory with Ursell–Mayer expansion.

1 Introduction

We intend in this article to answer and bring to attention some questions regarding the Kamerlingh Onnes virial series of a system of particles interacting through two-body repulsive potentials. We shall restrict ourselves to a two-parameter family of mean field type models ($t, \varepsilon \in \mathbb{R}_+$) for which the equation of state

$$\beta P = \rho + \frac{t}{2}\rho^2 \tag{1.1}$$

is attained in the limit as $\varepsilon \to 0$. As usual, P, ρ and $\beta = 1/kT$ denote the fluid pressure, density and inverse temperature (subsequently fixed to 1) and (1.1) is the equation of state of a system of hard spheres in infinitely many dimensions (Frisch, Rivier and Wyler (1985)), for which the Mayer series is known explicitly as well as other quantities that will be referred to in our discussion about the radius of convergence. All results presented in this article (mainly in Sections 2, 3 and 4) have been obtained in a joint work with D. Brydges whose preliminary version is available in the archive (Brydges and Marchetti (2014)). Sketch and parts of the proofs are reproduced in the present text to make it readable.

Key words and phrases. Virial expansion, uniformly repulsive potential, Hamilton–Jacobi equation, Cauchy-majorant method.
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Context

Kac, Uhlenbeck and Hemmer, in their successful attempt at understanding the approach to the mean field theory (Kac, Uhlenbeck and Hemmer (1963)) have addressed the van der Waals theory of vapor–liquid equilibrium using a one-dimensional system introduced by Kac (1959) for which the calculations can be carried out quite explicitly. For N particles with hard-core diameter δ moving in a linear container of size L and interacting in pairs through an attracting potential $\beta\phi(x) = -\alpha\gamma e^{-\gamma|x|}$, as N, L go to ∞ with $\rho = N/L$ finite, following by $\gamma \to 0$, they show that this system satisfies exactly the van der Waals equation of state

$$\beta P = \frac{\rho}{1-\delta\rho} - \frac{\alpha}{2}\rho^2, \tag{1.2}$$

together with the Maxwell rule. For finite γ^{-1}, that is for finite range of attractive force, the system does not show a phase transition. However, for the weak but very long-range force (as $\gamma \to 0$), a phase transition appears for α large enough, manifested by the presence of a plateau replacing part of the graph of (1.2) at the saturation pressure $P = p_{\text{sat}}$ between the vapor and liquid saturation densities ρ_v and ρ_l.

Together with van Kampen's ideas (van Kampen (1964)), the mean field approach introduced by Kac–Uhlenbeck–Hemmer was subsequently extended by Lebowitz and Penrose (1966) to d-dimensional systems of particles interacting through a pair potential consisting of two parts: $\phi(\mathbf{x}) = \varphi_0(\mathbf{x}) + \gamma^d\varphi(\gamma\mathbf{x})$, where $\varphi_0(\mathbf{x})$ and $\varphi(\mathbf{x})$ are short-range potentials, φ is either a nonpositive or a nonnegative-definite function such that $\pm\alpha = \int_{\mathbb{R}^d}\varphi(\mathbf{x})\,d^dx$ ($\alpha \geq 0$) exists as a Riemann integral. The result of Lebowitz and Penrose (1966) may be summarized as follows (see statement therein for precise conditions). Let $f(\rho)$ (resp. $f_0(\rho)$) denote the Helmholtz free energy density for a system of N particles moving in a finite container Λ interacting in pairs through ϕ (resp. φ_0) and let $N, |\Lambda|$ go to ∞ with $\rho = N/|\Lambda|$ finite. Then, in the so-called van der Waals limit ($\gamma \to 0$), the free energy density is given by the convex envelope[1]

$$f(\rho) = \sup\left\{h(\rho) : h(\rho) \leq f_0(\rho) \pm \frac{\alpha}{2}\rho^2, h \text{ convex}\right\}. \tag{1.3}$$

The equation of state is defined by $P = \partial(vf(1/v))/\partial v$ with $v = 1/\rho$ the specific volume which, for finite γ^{-1}, reads $P(\rho;\gamma) = \rho f'(\rho;\gamma) - f(\rho;\gamma)$. As the right and left derivatives exist for a.e. ρ by (1.3), it follows that $\gamma \to 0$ commutes with

[1] The supremum over convex functions $h(x)$ may be replaced by the supremum over affine functions $A(\rho) = f(\rho_0) + m(\rho - \rho_0)$ satisfying the same inequality.

the derivative operation and

$$\begin{aligned} P(\rho) &= \lim_{\gamma\to 0} P(\rho;\gamma) \\ &= \left(\rho\frac{d}{d\rho}-1\right)\mathrm{Conv}\left(f_0(\rho)\pm\frac{\alpha}{2}\rho^2\right) \\ &= P_0(\rho)\pm\frac{\alpha}{2}\rho^2 \end{aligned} \tag{1.4}$$

holds together with the Maxwell rule. The geometric interpretation of (1.4) is that $-P(\rho)$ is the ordinate of the point for which the tangent at ρ to the graph of $f(\rho)$ intercepts the f-axis and such interpretation is similar to the Legendre transform of $f(\rho)$ for which we shall deal with in our approach (see Proposition A.4 and Presutti (2009)). Note that, if the graph of $f(\rho)$ has a straight segment, $P(\rho)$ would remain constant along it, by the geometric interpretation, and the same behavior is satisfied for the chemical potential $\mu(\rho)=f'(\rho)=(f(\rho)+P(\rho))/\rho$. A first-order phase transition occur in thermodynamics if $P(\rho)=p_{\mathrm{sat}}$ and $\mu(\rho)=\mu_{\mathrm{sat}}$ are both constants in (ρ_v,ρ_l), under which the Maxwell equal area rule $\int_{\rho_v}^{\rho_l}(P_0(\rho)-\alpha\rho^2/2-p_{\mathrm{sat}})/\rho^2\,d\rho=0$ is implied by the Gibbs–Duhem equation.

Equation (1.4) includes the two cases (1.1) and (1.2), whose reference systems satisfy the ideal gas equation $\beta P_0=\rho$ and Tonks (1936) hard-rods equation $\beta P_0=\rho/(1-\delta\rho)$, respectively. The mean field correction to the reference system is negative or positive depending on whether the Kac potential is attractive or repulsive (or, more generally, nonnegative-definite with $\int_{\mathbb{R}}\varphi(\mathbf{x})\,d^dx>0$). The derivation of (1.4) involves several limits, taking according to four characteristic lengths: the range δ of the short-range potential, the size $|\lambda|$ of the coarse-graining cells, the range γ^{-1} of the Kac potential and the size of the container $|\Lambda|$. The order that the limits are taken must respect the relationship $\delta\ll|\lambda|^{1/d}\ll\gamma^{-1}\ll|\Lambda|^{1/d}$ to make their results attained, particularly in the attractive case. The order is, however, less relevant in the repulsive case and since the sum of two convex functions, $f_0(\rho)$ and $\alpha\rho^2/2$, is already convex, first order phase transition through this mechanism does not occur. In view of these considerations, our present investigation of a system of particles interacting through repulsive Kac-like potential shall not respect the order of the limits neither will use as many characteristic lengths.

In and out of scope

The present article does not touch on the perturbation about mean field theory as well as further developments involving the theory of minimizing the free energy functionals of Landau–Ginzburg-type and other hybrid models with Kac potentials as these topics have been explained so well by Prof. Presutti in his enlightening lectures. *Perturbation about*, as opposed to *approach to*, means that $f(\rho;\gamma)$ has a straight line segment characterizing a first order phase transition not only at $\gamma=0$ but for every γ sufficiently small (Lebowitz, Mazel and Presutti (1999)).

For an account on these and other issues related to first order phase transitions, see Presutti (2009) and references therein. We shall turn back, instead, to some problems posed by the distinguished authors Kac, Uhlenbeck and Hemmer (1963) (see pp. 224–225 therein) that have not received satisfactory answer so far: 1. What is the relation of the approach to mean field theory with the Yang–Lee theory (Yang and Lee (1952)) of condensation? 2. What is the relation of the approach to mean field theory with the Ursell–Mayer theory?

Except for some brief observations, we do not have anything to add regarding question 1. The zeros of the grand canonical partition function $\Xi_L(z)$ accumulate in layers on the complex activity $z = e^{\beta\mu}$ plane as the container size L tends to infinity. If they concentrate in a layer which crosses the positive real axis at $z = z_0 = e^{\beta\mu_0}$ then the pressure $\beta p(\mu) = \lim_{L\to\infty}(1/L)\log \Xi_L(e^{\beta\mu})$, for μ real, will consist of two analytic pieces, one for $\mu < \mu_0$ and one for $\mu > \mu_0$, continuous but the right and left derivatives at μ_0 will be discontinuous. This picture is illustrated by an artificial model proposed by Ford (see Chapter 3 of Uhlenbeck and Ford (1963) and Section 5) which has been unexpectedly evoked in our approach (see Remark 4.5). The inverse of $\beta p(\mu)$, the Gibbs free-energy per particle, given by equations (18) and (19) of Kac, Uhlenbeck and Hemmer (1963), $\mu(p) = \beta\alpha\gamma/2 - \log \lambda_0(\beta p)$, where $\lambda_0(s)$ is the largest eigenvalue of the so-called Kac integral equation, is a monotone increasing concave function, whose derivative is discontinuous at the saturation pressure p_{sat} in the limit as $\gamma \to 0$. Despite of the exact description of the thermodynamics, the authors Kac, Uhlenbeck and Hemmer (1963) were unable to substantiate the Yang–Lee picture because the behavior of the largest eigenvalue $\lambda_0(s)$ in the complex $\zeta = s + i\eta$ plane was unknown for $\eta \neq 0$.

Regarding question 2, the authors Kac, Uhlenbeck and Hemmer (1963) were unable of finding a "graphological" interpretation for the coefficients a_n of the Laurent series $e^{-\beta\alpha\gamma/2}\lambda_0(\zeta) = 1/\zeta + \sum_{n\geq 0} a_n\zeta^n$ thereby connecting only few a_n with few coefficients b_n of the Mayer series. The Mayer coefficient b_n is a sum over connected linear graphs (Mayer graphs) of size n, the weight of each graph is the integral of $\prod_{(ij)} f_{ij}$ over the space of configurations Λ^n where $f_{ij} = \exp(-\beta\varphi(x_i - x_j)) - 1$ if the pair (ij) is connected and 1 otherwise. It is possible to introduce Mayer functions f_{ij} for the repulsive and for the attractive part of the potential and the question is which simplifications occur if the attractive force is long range and whether such simplifications lead to an asymptotic formula of the b_n.

Our scope is as follows. We shall not attempt to solve questions 1 and 2, at least for the approach to the mean field theory presenting a vapor–liquid phase transition. Question 2 will be addressed when the mean field theory has equation of state (1.1). Once a Kac-like repulsive potential excludes phase transitions of vapor–liquid or fluid–solid types, the radius of convergence of the Mayer (or virial) series is not expected to be affected by a (physical) singularity in the real positive axis.

The analysis of the Mayer series, however, encounters sooner or later a limitation coming from a (unphysical) singularity of combinatorial origin and the question to be faced is whether it can be circumvented, so that the virial series, independently of any estimate for the Mayer series, would converge beyond the present bounds on its radius of convergence (see, e.g., Theorem 4.3.2 of Ruelle (1969)). The present investigation may be considered the first step toward the resolution of question 2.

The approach

We study a gas of point particles interacting through a uniformly repulsive pair potential $\phi(\mathbf{x};t) = t/|\Lambda|$, $t > 0$, in a finite volume Λ of size $|\Lambda| = 1/(2\varepsilon)$, which may be thought of as a Kac-like potential $\phi(\mathbf{x};t) = t\gamma^d \chi(\gamma|\mathbf{x}|)$ with $\chi(r) = 1$ if $0 \le r < 1$ and 0 otherwise and γ^d proportional to $|\Lambda|^{-1}$, where Λ is a large fraction of the total volume in which the system is in local equilibrium. The system is approached using a method introduced by Brydges and Kennedy (1987) by which Mayer expansions are obtained through an initial value problem for an evolution equation of Hamilton–Jacobi type. As in the renormalization group flow equations originally proposed by Wilson, such an approach allows an iterated (multi-scaling) Mayer expansion but we shall not exploit this possibility here.

Employing a system of equations satisfied by the Ursell functions (Lemma 3.3 of Brydges and Kennedy (1987)), we show that the pressure $p = p(t,\mu)$, as a function of the parameter t that interpolates the ideal to the real gas and μ the chemical potential, satisfies exactly a "viscous" Hamilton–Jacobi equation

$$p_t + \varepsilon(p_{\mu\mu} - p_\mu) + \tfrac{1}{2}(p_\mu)^2 = 0 \tag{1.5}$$

with $p(0,\mu) = e^\mu$. The repulsive interaction, expressed by the "wrong" sign in front of the Laplacian, avoids the collapse of particles into a single point (equilibrium stability).

We are looking for solutions of (1.5) in the form of a power series in the activity $z = e^\mu$:

$$p(t,\mu) = \sum_{n=1}^{\infty} b_n(t) z^n := \wp(t,z) \tag{1.6}$$

which are regular at $\varepsilon = 0$ (infinite $|\Lambda|$ limit). The so-called Mayer solution exists globally in the sense that the solution for (1.5) exists in a domain $\Omega \subset \mathbb{R}_+ \times \mathbb{C}$, defined by

$$e\lambda(t)|z| < 1 \tag{1.7}$$

with $\lambda(t)$ bounded for all $t \ge 0$,[2] as a holomorphic function of z, uniformly in ε. The global existence of (1.5) has to be contrasted with the finite t existence statement in Proposition 2.6 of Brydges and Kennedy (1987) for the theory of Mayer

[2] $\lambda(t)$ will be identified later with twice the second virial coefficient B_2.

expansion (see also Theorem 2.2 of Guidi and Marchetti (2004) for alternate formulation and proof of the same result). Note that our repulsive potential fulfills the conditions for the application of the general theorem and if it is plugged into the existence domain defined in the statement, then $\lambda(t)$ in (1.7) is replaced by an exponentially increasing function $\tau(t) = e^{t/|\Lambda|}\lambda(t)$ and finite t existence would be concluded instead. The general existence theorem uses Cauchy majorant method applied to a system of equations satisfied by the Ursell functions and a majorant PDE equation is deduced afterward. The global existence improvement is due to the alluded simplification of Kac-like potentials by which an exact PDE equation (1.5) is obtained right away, preserving the original sign of the nonlinear term. The introduction of Brydges and Kennedy (1987) is recommended for additional explanations regarding this issue.

To establish the existence domain (1.7), we apply the variation of constants formula to (1.5) (actually, to an equation for $\wp_z$, the derivative of $\wp$ w.r.t. z) which yields an iterated system of integral equations for the b_n to which the Cauchy majorant methods is applied. In this way, being the generating function for labelled enumeration of simply connected Mayer graphs, (1.6) is majorized by the corresponding sum over labelled trees. If $\mathcal{R}_\wp$ denotes the radius of convergence of (1.6) the inequality (1.7) implies $e\lambda\mathcal{R}_\wp \geq 1$ for all $t \geq 0$ and the equality holds in the limit $\varepsilon \to 0$ (see Theorem 3.1).

Stronger results can actually be proven (see Theorem 3.2). It follows from Lieb's inequalities (see, e.g., Ruelle (1969), Section 4.5, and references therein) that the alternating sign property (a.s.p.), $(-1)^{n-1}b_n > 0$ and the upper and lower bounds

$$e^{-1} \leq \lambda\mathcal{R}_\wp \leq 1 \tag{1.8}$$

on the radius of convergence of the Mayer series for the pressure $\wp$ (or density $z\wp_z$), hold for any nonnegative potential with λ equal twice the second virial coefficient; the upper bound is attained for the Ford model. We prove in addition that (i) $\lambda\mathcal{R}_\wp$ is monotone increasing in t; the inequalities (1.8) saturate at both extremities: (ii) $\lim_{t\to 0}\lambda\mathcal{R}_\wp = e^{-1}$ and (iii) $\lim_{t\to\infty}\lambda\mathcal{R}_\wp = 1$, for any $0 < \varepsilon < \infty$.

Motivations

The convergence of Mayer series for the class of stable and tempered potentials (including nonnegative ones) (Ruelle (1969), Chapter 4) is affected by the presence, in its majorant, of a (movable and unphysical) singularity in the negative real line of the complex z-plane which would be inherited by the virial series

$$P(t,\rho) = \sum_{n=1}^{\infty} B_n\rho^n, \tag{1.9}$$

whether the Lagrange inversion formula is applied to the composition $\wp(t, Z(t,\rho))$, where $Z = Z(t,\rho)$ solves $z\wp_z(t,z) = \rho$ for z, to express the pressure as a function of the density ρ, as in the classic paper Lebowitz and Penrose (1964). See equations (5.1) and (5.2) in Section 5.

Alternatively, the virial coefficients $(B_n)_{n\geq 1}$ and the *irreducible cluster integrals* $(\beta_n)_{n\geq 1}$ which satisfy $B_1 = 1$ and

$$B_n = -\frac{n-1}{n}\beta_{n-1}, \qquad n \geq 2, \tag{1.10}$$

may be assessed from the Helmholtz free energy density $f(t,\rho)$ through a Legendre transform of $p(t,\mu)$ (see Proposition A.4). Excluding from the free energy

$$f(t,\rho) = \rho\log\rho - \rho - \beta(t,\rho), \tag{1.11}$$

the ideal gas contribution $f(0,\rho) = \rho\log\rho - \rho \equiv f^{\text{ideal}}$, the derivative $\beta_\rho(t,\rho)$ generates the labelled "enumeration" (total weight of species) of (2-connected) irreducible Mayer graphs

$$\varphi(t,\rho) = \sum_{n=1}^{\infty}\beta_n(t)\rho^n. \tag{1.12}$$

It follows by (1.10), that the radii of convergence $\mathcal{R}_\beta$, $\mathcal{R}_P$ and $\mathcal{R}_\varphi$ of the power series $\beta(t,\rho)$, $P(t,\rho)$ and $\varphi(t,\rho)$ about $\rho = 0$ are all the same. The Helmholtz free energy (extracted the ideal contribution) $f - f^{\text{ideal}} = -\beta$ has been recently addressed by cluster expansion Pulvirenti and Tsagkarogiannis (2012) and Morais and Procacci (2013) have shown, by means of an expression already known by Mayer, that $\mathcal{R}_\beta$ satisfies Lebowitz–Penrose's lower bound on the radius of convergence of the virial series. It thus seems quite opportune to inquire whether the referred singularity on the Mayer series could somehow be prevented.

The present work has been motivated by the following open problem:

> *Is there a system of interacting particles for which β, P and/or φ can be directly assessed, Lagrange inversion formula be avoided and Lebowitz–Penrose's lower bound on $\mathcal{R}_P$ be improved?*

Results

We prove (see Appendix C of Brydges and Marchetti (2014)) that if $p(t,\mu)$ satisfies the initial value problem (1.5) then $\varphi(t,\rho)$ satisfies

$$\varphi_t + \rho + \varepsilon\left(\frac{1+\rho^2\varphi_{\rho\rho}}{(1-\rho\varphi_\rho)^2} - 1\right) = 0 \tag{1.13}$$

with $\varphi(0,\rho) = 0$ and an affirmative answer to the previous question is provided for the system of particles interacting through a uniformly (Kac-like) repulsive pair potential, described by equation (1.13), with ε playing a role of Kac parameter γ.

Using a modified version of the Cauchy-majorant method, we establish the following result (see Theorem 4.1). There exists a function $\Phi(t,\rho) = \sum_{n\geq 1} \Phi_n(t)\rho^n$ that majorizes $\varphi(t,\rho)$ in the sense that $|\beta_n(t)| \leq \Phi_n(t)$ holds for all $n \in \mathbb{N}$ and $t \in \mathbb{R}_+$, uniformly in ε, such that its radius of convergence $\mathcal{R}_\Phi$ satisfies

$$\lambda \mathcal{R}_\varphi \geq \lambda \mathcal{R}_\Phi = \varkappa \tag{1.14}$$

with $\varkappa = \varkappa(\eta)$ given by (4.1). Here $\eta = e^{-2\varepsilon t}$ and $\lambda = (1-\eta)/(2\varepsilon)$ is the function in (1.7), the L_1-norm of the Mayer f-function $f(\mathbf{x};t) = e^{-\phi(\mathbf{x};t)} - 1$ of a uniformly repulsive potential $\phi(\mathbf{x};t) = 2\varepsilon t$, $\mathbf{x} \in \Lambda$. We observe that the factor $\lambda = 2B_2$, B_2 is the second virial coefficient, makes the radius of convergence of different systems comparable with each other. The curve defined by the r.h.s. of (1.14) stays above the Lebowitz–Penrose's lower bound $\lambda \mathcal{R}_P > 0.144767$ (equation (3.11) of Lebowitz and Penrose (1964) with $u = 1$ and $B = \lambda$) for all t while, for $\varepsilon t \leq 0.00538$, the pre-factor $\varkappa$ goes beyond the threshold 0.278465, established for nonnegative potentials $\lambda \mathcal{R}_P > 0.278465$ (see Ruelle (1969), Theorem 4.3.2 et seq.). $\varkappa$ as a function of η is plotted in Figure 2. By (1.14), $\lambda \mathcal{R}_\varphi(t) > 0$ for all $(t,\varepsilon) \in \mathbb{R}_+ \times \mathbb{R}_+$ and the t and $\varepsilon = 1/(2|\Lambda|)$ for which φ converges in the domain below the hyperbole $\varepsilon t = 0.00538$ represent the regime of "high temperatures" and/or large volumes. The proof of Theorem 4.1, one of the main contributions of Brydges and Marchetti (2014), introduces some novelties with respect to the traditional majorant method and involves technical difficulties (see Proposition E.1 of Brydges and Marchetti (2014)).

As $\varepsilon t \to 0$, we prove (see Theorem 4.1 of Brydges and Marchetti (2014)) that

$$\frac{\beta_n}{\lambda^n} = (-1)^{n+1}\varepsilon t\big(1 + O(\varepsilon t)\big), \tag{1.15}$$

for $n \geq 2$ ($\beta_1/\lambda \equiv -1$) and, by continuity, the β_n satisfy both a.s.p. and $\lambda \mathcal{R}_\varphi = 1$ if $t\varepsilon$ is sufficiently small. For $t\varepsilon \ll 1$, $\lambda \mathcal{R}_P = 1$ holds even though, as explained in Section 5, we have $\lambda \mathcal{R}_P \geq W(e^{-1}) = 0.278465\ldots$, by Lagrange inversion formula. For t large, we have

$$\lim_{t\to\infty} n\beta_n/\lambda^n = -1$$

and an explicit computation using Mathematica indicates that $\lambda \mathcal{R}_\varphi = \lambda \mathcal{R}_P = 1$ is expected to hold for all (t,ε).

Related issues

There are other issues regarding the virial coefficients of systems with repulsive potentials that can be addressed by the present (mean field) model. The work by Clisby and McCoy (2006) on the virial coefficients for the hard spheres in d-dimensions, which collected and reviewed a great deal of informations, has provided strong evidence that the leading singularity for the virial series lies away from the positive real line. We compute the β_n $(= -(n+1)B_{n+1}/n)$ from an exact recursion relation satisfied for our model and show that, as function of n, they oscillate about the axis. The calculation indicates that the period increases from 1 (the alternating sign behavior (1.15)) to infinity as t varies from 0 to ∞. Each $n\beta_n/\lambda$, as a function of t, oscillates until it reaches a maximum at t_n^* and from there on it converges rapidly to -1 (see Figure 4 of Brydges and Marchetti (2014)), indicating that the leading singularity of the virial series remains away from the positive real line, at least for $t < t_\infty^* = \lim_{n\to\infty} t_n^*$. Other models whose leading singularities are out of the real line include the Gaussian model (see, e.g., Clisby and McCoy (2006) and references therein) and hard-core lattice gases in two-dimensions, particularly the hard-hexagon model whose radius of convergence of the virial series has been determined exactly by Joyce (1988) (see equation (12.30) therein) and which is less than its critical density ρ_c.

The Mayer and virial series at low temperature have been addressed recently by Jansen (2012) for a class of potentials satisfying a n-particle ground state geometry condition and some of her results extend to nonnegative potentials as well. We should mention that her results on the radii of convergence of the virial series (1.9) and of the series of $\wp \circ Z(\rho)$ in power of ρ go, however, in the opposite direction of ours for the uniformly repulsive potential (see, e.g., Theorem 3.8 of Jansen (2012)). We should warn that, since we are fixing $\beta = 1$ (activity and density are given by $z = e^{\mu}$ and $\rho = p_\mu$) the limit $t \to \infty$ does not really mean low temperature limit, although we sometimes abuse of language. We mention that equation (1.13) has a stationary solution: $\varphi_0(\rho) = \log(1 - \rho/(2\varepsilon))$ (solves (1.13) with $\varphi_t = 0$), from which one obtains the pressure

$$P_0(\rho) = -2\varepsilon \log\big(1 - \rho/(2\varepsilon)\big)$$

of the hard-core lattice gas or Ford model (5.16) in the low density regime.

Outline

In Section 2, we introduce our potential model and establish a relationship between macroscopic functions and their corresponding PDEs. Sections 3 and 4 contain the (global) existence theorems for equations (1.5) and (1.13), respectively, using Cauchy-majorant and asymptotic methods. Section 5 provides an overview on the convergence of virial series. Our conclusions and open problems are stated in Section 6. In the Appendix we review the Mayer theory of imperfect fluids in thermodynamic equilibrium.

2 Thermodynamic functions and PDEs

Two-parameter potential model

We consider an equilibrium system of *point-particles* in a finite container $\Lambda \subset \mathbb{R}^d$ interacting through a uniformly repulsive two-body potential

$$\phi_{ij} \equiv \phi_\Lambda(t; x_i - x_j) = \frac{t}{|\Lambda|} \tag{2.1}$$

for every $x_i, x_j \in \Lambda$, $t > 0$.

The interacting potential $\phi_\Lambda = \phi_\Lambda(t; x) = t/|\Lambda|$ depends on the size $|\Lambda|$ of the "container" and on a parameter t which plays a double role of "time" (or evolution variable) and "inverse temperature" (so, we set $\beta = 1$ in (A.3) and replace β by t in all functions defined in the Appendix). In contrast with the hard-core gas of particles, whose partition function is a polynomial of activity for finite Λ, the system of point particles in a continuum space is already in the "macroscopic limit", despite its volume $|\Lambda|$ is kept finite.

Note that ϕ_Λ is positive ($\phi_\Lambda(t; x) > 0$) and of positive-definite type:

$$\sum_{1 \le i, j \le N} z_i \phi_\Lambda(t; x_i - x_j) \bar{z}_j = \frac{t}{|\Lambda|} \left| \sum_{i=1}^{N} z_i \right|^2 \ge 0,$$

for any collection $(x_i)_{i=1}^N$ in Λ and complex numbers $(z_i)_{i=1}^N$, satisfying thereby *stability*:

$$\sum_{1 \le i < j \le N} \phi_\Lambda(t; x_i - x_j) \ge \frac{-t}{2|\Lambda|} N$$

and "*integrability*" (in the $L_1(\Lambda; d^d x)$-sense):

$$\|\phi_\Lambda(t; \cdot)\|_1 := \int_\Lambda \phi_\Lambda(t; y)\, d^d y = t$$

and

$$\int_\Lambda \left| e^{-\phi_\Lambda(t; y)} - 1 \right| d^d y = |\Lambda| \left(1 - e^{-t/|\Lambda|}\right) \equiv \lambda\left(t, |\Lambda|\right), \tag{2.2}$$

where $\lambda(t, |\Lambda|)$ is monotone increasing function of t and $|\Lambda|$ such that $\lambda(\mathbb{R}_+, |\Lambda|) = [0, |\Lambda|]$ and $\lambda(t, \mathbb{R}_+) = [0, t]$ remain bounded for $|\Lambda|, t > 0$. As claimed in the Introduction, the time for which the Mayer expansion is proven to exist (see Proposition 2.6 of Brydges and Kennedy (1987)), satisfies $e\tau(t)|z| < 1$, where

$$\begin{aligned} \tau(t) &= \int_0^t \|\dot{\phi}_\Lambda(s; \cdot)\|_1 \exp\left(\int_s^t \dot{\phi}_\Lambda(s'; 0)\, ds' \right) ds = \int_0^t e^{(t-s)/|\Lambda|}\, ds \\ &= e^{t/|\Lambda|} \lambda\left(t, |\Lambda|\right), \end{aligned}$$

provides only finite time existence to our system. We shall prove global existence by majorant method which takes into account the specific form of mean field potential ϕ_Λ.

We refer to the Appendix for basic properties and formal expressions of the thermodynamic functions pertaining to this section.

Weight of a connected graph G

The problem of evaluating B_n, the nth coefficient (A.18) of Kamerlingh Onnes virial series (A.16) may be divided into two distinct ones. The combinatorial problem (see the Appendix for a brief account), is independent of the law of interacting forces between any two particles. The second problem is the integration over the configuration space Λ^n. For interacting potential (2.1), the weight $w_\Lambda(G)$ of a irreducible (2-connected) graph G with n vertices and $l = |E(G)|$ edges satisfies exactly

$$\begin{aligned} w_\Lambda(G) &= \frac{1}{|\Lambda|}\int_{\Lambda^n} \prod_{(ij)\in E(G)} \left(e^{\phi_\Lambda(x_i-x_j)} - 1\right) d^d x_1 \cdots d^d x_n \\ &= (-1)^l |\Lambda|^{n-l-1} \lambda^l\big(t, |\Lambda|\big) \end{aligned} \tag{2.3}$$

by (2.2), reducing the two problems to a purely combinatorial one—this should be contrasted with the “soft-core” Gaussian model whose explicit evaluation, $w(G) = (-1)^l (\pi/\alpha)^{3(n-1)/2} \gamma^{-3/2}$ depends on the graph complexity $\gamma = \gamma(G)$ of G in addition to the two other dependences, on the number of vertices and edges, already in (2.3) (see Uhlenbeck and Ford (1963), for definition, evaluation and motivations).

The number of edges $l = |E(G)|$ of a connected graph G with n vertices satisfies $l \geq n-1$, with equality only for tree graphs T. Since for any connected graph G we have

$$w_\infty(G) = \lim_{|\Lambda|\to\infty} w_\Lambda(G) = \begin{cases} (-t)^l, & \text{if } G = T, \\ 0, & \text{otherwise,} \end{cases}$$

in view of (2.3) and $\lambda(t, \infty) = t$, and since the only 2-connected tree is, by definition, the graph with two vertices connected by a single edge T_2, the limit weight $w_\infty(G)$ vanishes for all irreducible (2-connected) graphs G different from T_2. In the limit as $|\Lambda| \to \infty$, the virial series (A.16) thus reads

$$P^\infty(t, \rho) = \rho + \frac{t}{2}\rho^2$$

by dissymmetry theorem (see, e.g., Bergeron, Labelle and Leroux (1998)), which agrees with the pressure of a system of hard spheres in infinitely many dimensions.

Another limit is attained when t tends to ∞ (the “low temperature limit”) with $|\Lambda|$ finite. In this limit, the weight of a 2-connected graph G reads

$$\lim_{t\to\infty} w_\Lambda(G) = (-1)^l |\Lambda|^{n-1}$$

and, by a subtle cancellation (exactly as in the hard-core lattice gas, for which $f_{ij} = -1$ if $x_i = x_j$ and $f_{ij} = 0$ if $x_i \neq x_j$, except that the sum of a n-particle configuration gives $|\Lambda|$ instead $|\Lambda|^n$),

$$P(\infty, \rho) = \frac{-1}{|\Lambda|} \log\big(1 - |\Lambda|\rho\big), \qquad \rho < 1/|\Lambda|$$

agreeing, this time, with the pressure (5.16) of Ford model at the low density regime. Both limit functions will be shown to be attained by our investigation of macroscopic functions (pressure and the Helmholtz free energy) as power series solution of related partial differential equations.

Partial differential equations

As a consequence of (2.3), we have (Proposition 2.1 of Brydges and Marchetti (2014)):

Proposition 2.1.

(a) *The pressure* $p = p_\Lambda(t, \mu)$ *of a uniformly repulsive pairwise interacting system, as a function of* t *and the chemical potential* $\mu = \log z$, *for any finite* Λ, *satisfies a partial differential equation* (*PDE*)

$$p_t + \varepsilon(p_{\mu\mu} - p_\mu) + \tfrac{1}{2}(p_\mu)^2 = 0, \tag{2.4}$$

$\varepsilon = 1/(2|\Lambda|)$, *with initial condition* $p_\Lambda(0, \mu) = e^\mu$.

(b) *The Helmholtz free energy* $f = f_\Lambda(t, \rho)$ *defined by the* (*formal*) *Legendre transform of* $p_\Lambda(t, \mu)$ *w.r.t.* μ (*equation* (A.20)) *satisfies*

$$f_t - \varepsilon\left(\frac{1}{f_{\rho\rho}} - \rho\right) - \frac{1}{2}\rho^2 = 0 \tag{2.5}$$

with $f_\Lambda(0, \rho) = \rho \log \rho - \rho$.

(c) *The function* $\varphi = \varphi(t, \rho)$, *defined by*

$$f_\rho = \log \rho - \varphi, \tag{2.6}$$

satisfies

$$\varphi_t + \rho + \varepsilon\left(\frac{1 + \rho^2 \varphi_{\rho\rho}}{(1 - \rho\varphi_\rho)^2} - 1\right) = 0 \tag{2.7}$$

with $\varphi(0, \rho) = 0$ *and generates*, *by Proposition* A.4, *the irreducible cluster integrals* β_n, $n = 1, 2, \ldots,$ *defined by* (A.10), *that is*, *the* β_n *are the coefficients of a formal power series* (1.12) *in* ρ *of* φ.

We omit the proof of item (a) of Proposition 2.1 which is given in Appendix C of Brydges and Marchetti (2014). Equation (2.4) is deduced straitforwardly from the Brydges–Kennedy equations (see Lemma 3.3 of Brydges and Kennedy (1987)) satisfied by the Ursell functions.

Proof of Proposition 2.1, parts (b) and (c). See Section 6 in Chapter 1 of Courant and Hilbert (1989). The Legendre transform of a formal power series $p(t, \mu)$ in e^μ, with respect to μ, is

$$f(t, \rho) = \rho\mu^* - p\big(t, \mu^*\big), \tag{2.8}$$

where $\mu^* = \mu^*(t, \rho)$ solves

$$\rho = p_\mu(t, \mu) \tag{2.9}$$

for μ; the Legendre transform of $f(t, \rho)$ with respect to ρ is

$$p(t, \mu) = \rho^* \mu - f\big(t, \rho^*\big), \tag{2.10}$$

where $\rho^* = \rho^*(t, \mu)$ solves $\mu = f_\rho(t, \rho)$ for ρ. From these, we deduce

$$\frac{1}{f_{\rho\rho}} = p_{\mu\mu}, \tag{2.11}$$

$$-f_t = p_t. \tag{2.12}$$

Equations (2.12), (2.9) and (2.11) substituted into (2.4) yields (2.5) and concludes the proof of part (b).

Differentiating (2.5) w.r.t. ρ together with (2.6) yields (2.7). We observe that the operations involved in the Legendre transform, together with (2.6) (sum, multiplication, derivatives, inverse, composition, reciprocal, ...), apply over the ring $\mathcal{C}^1[[z]]$ of formal power series in $z = e^\mu$ with $\mathcal{C}^1$ coefficients $b_n(t)$, $n \geq 1$.

The proof of the statement (c) starts from (2.8):

$$f(t, \rho) = \rho\mu^*(t, \rho) - P(t, \rho), \tag{2.13}$$

where $p(t, \mu^*(t, \rho)) = P(t, \rho)$. Differentiating (2.13) w.r.t. ρ, together with $\mu^*(t, \rho) = f_\rho(t, \rho) = \log\rho - \varphi(t, \rho)$, yields $P_\rho(t, \rho) = 1 - \rho\varphi_\rho(t, \rho)$ which can be formally integrated (with $P(t, 0) = 0$). We conclude by

$$P(t, \rho) = \rho - \sum_{n=1}^{\infty} \frac{n}{n+1} \beta_n(t) \rho^{n+1} \tag{2.14}$$

together with (A.17) that the β_n are the irreducible cluster integrals. The proof of Proposition 2.1 is now complete. □

The Kamerlingh Onnes virial series (1.9) is thus given by (2.14). So, the power series (1.12) of $\varphi(t, \rho)$ converges if, and only if, the virial series converges.

3 Global existence of Mayer series

We shall write a majorant series whose radius of convergence attains the best known lower bound. We prove, in addition, that the radii of convergence of both, the majorant and the Mayer series for equation (2.4), agree in the limit as εt tends to 0. By Lieb's inequalities (see Section 4.5 of Ruelle (1969)), the alternating sign property (a.s.p.): $(-1)^{n-1} b_n > 0$ and upper and lower bounds: $e^{-1} \leq \lambda\mathcal{R}_\wp \leq 1$ on the radius of convergence of the Mayer series for the pressure $\wp$ (or density $z\wp_z$) hold, in general, for any nonnegative potential and, in particular, for the uniformly repulsive potential in consideration. For the latter model, we prove that $\lambda\mathcal{R}_\wp$ is monotone increasing in $t \in (0, \infty)$ and the inequalities saturate and become equalities, the left at 0 and the right at ∞ (see Theorem 4.5 of Brydges and Marchetti (2014)).

Majorant method

Theorem 3.1. *Let $\wp(t,z)$ be the pressure of a system of pointwise particles interacting through uniformly repulsive pair potential, represented by the power series (1.6). Let $W(x)$ be the Lambert W-function defined by the principal branch (regular at origin) of the inverse equation $W(x)e^{W(x)} = x$ given by $W_0(x) = \sum_{n\geq 1}(-n)^{n-1}/n!$, by Lagrange's theorem, and let $\lambda(t)$ be given by (2.2). Then, a majorant relation*

$$\wp_z(t,z) \ll \frac{-1}{\lambda(t)z} W_0\bigl(-\lambda(t)z\bigr)$$

is valid for all $t > 0$ in the sense that the $(n-1)$th coefficient of the power series on both sides satisfy

$$\bigl|nb_n(t)\bigr| \leq \frac{n^{n-1}}{n!}\lambda(t)^{n-1} \tag{3.1}$$

and the Mayer solution of equation (2.4) exists globally. Since $W_0(x)$ has a branch point at $-e^{-1}$, the radius of convergence $\mathcal{R}_\wp = 1/\limsup_{n\to\infty}|b_n(t)|^{1/n}$ of the pressure $\wp$ satisfies $e\lambda\mathcal{R}_\wp \geq 1$ and this relation is an equality as $\varepsilon = 1/(2|\Lambda|) \to 0$ for any $t \in (0,\infty)$.

Proof. Writing $q(t,z) = \sum_{k=0}^{\infty} q_k(t)z^k = \wp_z(t,z)$, we have

$$q_k = (k+1)b_{k+1} \tag{3.2}$$

and $\mathcal{R}_\wp = \mathcal{R}_q$, by definition of radius of convergence.

Equation (2.4) can be written in the form of a conservation law

$$q_t + \bigl(\varepsilon z^2 q_z + \tfrac{1}{2}z^2q^2\bigr)_z = 0 \tag{3.3}$$

with $q(0,z) = 1$. Denoting by $\mathbf{q}$ the sequence $(q_k)_{k\geq 0}$ of coefficients of its power series, (3.3) is equivalent to an infinite system of differential equations

$$\begin{aligned} \dot{q}_0 &= 0, \\ \frac{1}{k+1}\dot{q}_k + \varepsilon k q_k &= -\frac{1}{2}(\mathbf{q}*\mathbf{q})_{k-1}, \qquad k\geq 1 \end{aligned} \tag{3.4}$$

with $q_0(0) = 1$ and $q_k(0) = 0$, $k \geq 1$.[3] The convolution product is defined by $(\mathbf{q}*\mathbf{q})_m = \sum_{j=0}^{m} q_j q_{m-j}$. By the variation of constants formula, (3.4) is analogous to

[3] Here, $\dot{q}_k$ denotes the time derivative of q_k. The kth element q_k of the sequence $\mathbf{q}$ should not be confused with the partial derivatives of the function $q(t,z)$ w.r.t. t or z.

a system of integral equations

$$q_0(t) = 1$$

and

$$q_k(t) = \frac{-1}{2}(k+1)\int_0^t e^{-\varepsilon k(k+1)(t-s)} \big(\mathbf{q}(s) * \mathbf{q}(s)\big)_{k-1}\, ds, \qquad k \geq 1. \tag{3.5}$$

For $k = 1$, we have

$$q_1(t) = \frac{-1}{2\varepsilon}\big(1 - e^{-2\varepsilon t}\big) = -\lambda,$$

where $\lambda = \lambda(t, 1/(2\varepsilon))$. By convenience, we write $\lambda = (1-\eta)/(2\varepsilon)$, $\eta(t) = e^{-2\varepsilon t}$, and introduce $\tilde{\mathbf{q}} = (\tilde{q}_k)_{k\geq 0}$ with $\tilde{q}_k = q_k/\eta^k$, so (3.5) can be written as

$$\tilde{q}_k(t) = \frac{-1}{2}(k+1)\int_0^t e^{-\varepsilon k(k-1)(t-s)} \frac{1}{\eta(s)} \big(\tilde{\mathbf{q}}(s) * \tilde{\mathbf{q}}(s)\big)_{k-1}\, ds. \tag{3.6}$$

A sequence $\tilde{\mathbf{Q}} = (\tilde{Q}_k)_{k\geq 0}$ that majorizes $\tilde{\mathbf{q}} = (\tilde{q}_k)_{k\geq 0}$, is obtained as follows:

$$\tilde{q}_0(t) = \tilde{Q}_0(\tau) = 1$$

and

$$\begin{aligned} |\tilde{q}_k(t)| &\leq \frac{1}{2}(k+1)\int_0^t \frac{1}{\eta(s)} \big(|\tilde{\mathbf{q}}(s)| * |\tilde{\mathbf{q}}(s)|\big)_{k-1}\, ds \\ &\leq \frac{1}{2}(k+1)\int_0^\tau \big(\tilde{\mathbf{Q}}(\tau') * \tilde{\mathbf{Q}}(\tau')\big)_{k-1}\, d\tau' := \tilde{Q}_k(\tau), \end{aligned} \tag{3.7}$$

where $\tau = \tau(t) = \int_0^t (1/\eta(s))\, ds = \lambda(t)/\eta(t)$ is an strictly increasing function of t. Writing $\omega = \eta(t)z$, a sequence $\mathbf{Q} = (Q_k)_{k\geq 0}$ that majorizes $\mathbf{q} = (q_k)_{k\geq 0}$ satisfies

$$Q(t,z) := \sum_{k=0}^{\infty} Q_k(t) z^k = \sum_{k=0}^{\infty} \tilde{Q}_k(\tau)\omega^k := \tilde{Q}(\tau, \omega) \tag{3.8}$$

with the system of equations for $(\tilde{Q}_k)_{k\geq 1}$, going backward through the steps (3.4)–(3.5), being equivalent to the following PDE (compare with (3.3))

$$\tilde{Q}_\tau - \tfrac{1}{2}\big(\omega^2 \tilde{Q}^2\big)_\omega = 0 \tag{3.9}$$

with $\tilde{Q}(0,\omega) = 1$, whose solution can be written explicitly in terms of the Lambert W-function (see Section 5.1 of Guidi and Marchetti (2004))

$$Q(t,z) = \tilde{Q}(\tau,\omega) = \frac{-1}{\tau\omega} W_0(-\tau\omega) = \frac{-1}{\lambda z} W_0(-\lambda z). \tag{3.10}$$

See Corless et al. (1996) for the properties of $W_0(x)$ stated in the theorem, including its Taylor series at $x = 0$ given in equation (3.1). As a consequence, (3.8) converges provided

$$e\tau|\omega| = e\lambda|z| < 1 \tag{3.11}$$

which, together with (3.7) and $\mathcal{R}_{\wp} = \mathcal{R}_q$, establishes: $e\lambda\mathcal{R}_{\wp} \geq e\lambda\mathcal{R}_Q \geq 1$. Note that the majorant relation (3.7) (denoted by $q \ll Q$) is preserved by derivation, anti-derivation integration w.r.t. t and composition (see, e.g., van der Hoeven (2003)).

The equality $e\lambda\mathcal{R}_{\wp} = 1$ holds as ε tends to 0 for any $t \in \mathbb{R}_+$, as one can see by taking $\varepsilon = 0$ in (3.3) (or indirectly from (3.5)), whose solution is given by (3.10) with z replaced by $-z$. This concludes the proof of Theorem 3.1. □

Alternating sign property and monotonicity

Theorem 3.2. *The (normalized) radius of convergence $\lambda\mathcal{R}_{\wp}$ of the Mayer series for the pressure (or density) of a system of point-particles with uniformly repulsive pairwise potential is a (strictly) monotone increasing function of $t \in \mathbb{R}_+$ and satisfies*

$$\frac{1}{e} \leq \lambda\mathcal{R}_{\wp} \leq 1 \tag{3.12}$$

with equalities at the two extreme points $t = 0$ and ∞.

Proof. To prove equality, we introduce another sequence $\mathbf{c} = (c_k)_{k\geq 0}$ with $c_k = (-1)^k q_k/\lambda^k$. As one can see from (3.5), the factor $(-1)^n$ compensates the alternating sign of $(q_k)_{k\geq 0}$ (see Theorem 4.5.3 of Ruelle (1969)) so $c_k \geq 0$ holds for every k. It also follows from (3.5) that

$$\begin{aligned} c_k(t) &= \frac{k+1}{2}\frac{1}{\lambda^k(t)} \\ &\quad\times \int_0^t e^{-\varepsilon k(k+1)(t-s)}\lambda^{k-1}(s)\big(\mathbf{c}(s) * \mathbf{c}(s)\big)_{k-1}\, ds, \qquad k \geq 1, \end{aligned} \tag{3.13}$$

with $c_0(t) \equiv 1$. The asymptotic $\lambda(t) \sim t$, as t tends to 0, together with right continuity of $c_k(t)$ at $t = 0$ yield

$$c_k(0) = \frac{k+1}{2k}\big(\mathbf{c}(0) * \mathbf{c}(0)\big)_{k-1}, \qquad k \geq 1, \tag{3.14}$$

with $c_0 = 1$, whose solution is well known (see Lemma 4.2 of Brydges and Kennedy (1987)):

$$c_k(0) = \frac{(k+1)^k}{(k+1)!}. \tag{3.15}$$

Note that, by (3.2), $(-1)^{n-1}c_{n-1}(0) = nb_n/\lambda^{n-1} = (-n)^{n-1}/n!$ coincide with the coefficients of the Mayer series (5.5) for the density of the hard-spheres gas in infinitely many dimensions and also (in absolute value) with the coefficients of the majorant function (3.10), proving the equality $\lim_{t\to 0} e\lambda(t)\mathcal{R}_{\wp}(t) = 1$.

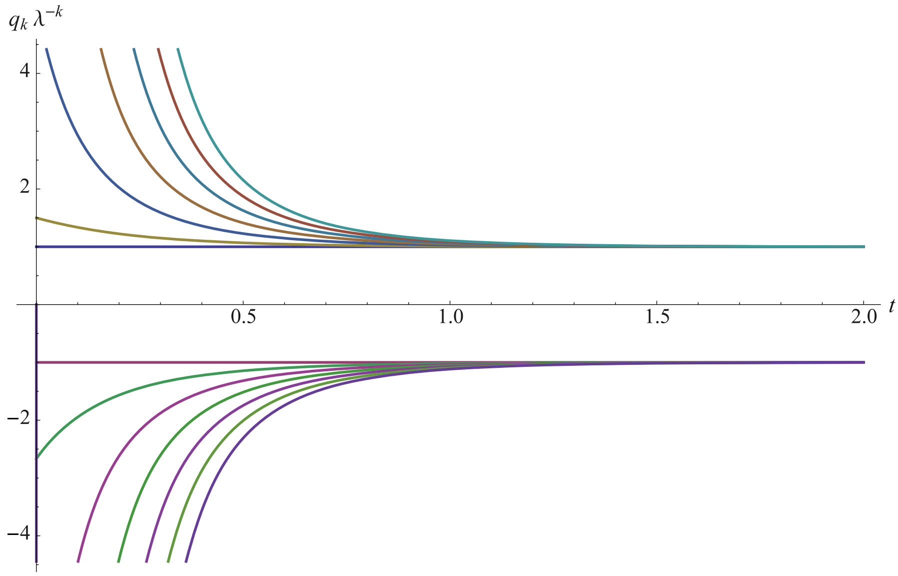

Figure 1 *Mayer's coefficients* q_k/λ^k, $k = 0, \ldots, 13$, *as a function of* t.

To prove Theorem 3.2, we need to show that $\dot{c}_k(t)$ remains negative for all $t \in \mathbb{R}_+$ and $\lim_{t\to\infty} c_k(t) = 1$—see computer evaluation of $q_k(t)/\lambda^k(t)$, $k = 0, \ldots, 13$, in Figure 1. We formulate these statements in the following proposition, whose proof (considerably more technical) we omit (see Appendix E of Brydges and Marchetti (2014)).

Proposition 3.3. *Let* $(c_k(t))_{k\geq 0}$ *be given by* $c_k = (-1)^k q_k/\lambda^k$ *with* $(q_k)_{k\geq 0}$ *the solution of* (3.5) *with* $q_0(t) \equiv 1$. *Then, for every* k, $c_k(t)$ *is monotone decreasing in* t, $c_k(0) = \lim_{t\searrow 0} c_k(t)$ *satisfies* (3.15) *and* $\lim_{t\to\infty} c_k(t) = 1$.

This concludes the proof of Theorem 3.2. □

4 Global existence of virial series

Using the Cauchy-majorant method similar but more elaborated than the one employed in the previous section to equation (2.4), we prove in Brydges and Marchetti (2014) the following theorem.

Theorem 4.1. *The solution* $\varphi(t, \rho)$ *of the initial value problem* (2.7) *exists, globally and uniformly in* ε *as the unique holomorphic function of* ρ, *inside the domain* $\Omega = \{(t, \rho) \in \mathbb{R}_+ \times \mathbb{C} : \lambda|\rho| < \varkappa\}$ *where*

$$\varkappa = \frac{2 + \sqrt{1-\eta} - 2\sqrt{1 - \eta/2 + \sqrt{1-\eta}}}{1+\eta} \tag{4.1}$$

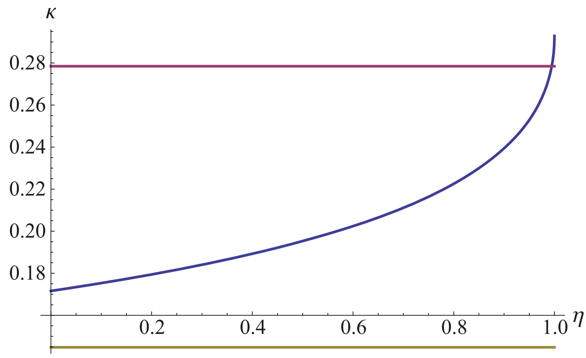

Figure 2 *$\varkappa$ as a function of η.*

$\eta = e^{-2\varepsilon t}$ and $\lambda = (1-\eta)/(2\varepsilon)$ is defined in (2.2).

The radius of convergence $\mathcal{R}_P(t)$ of the virial series (2.14) for a system of point particles interacting through a uniformly repulsive pair potential ϕ_Λ thus satisfies

$$\lambda \mathcal{R}_P \geq \varkappa. \tag{4.2}$$

For finite $\varepsilon = 1/(2\Lambda)$, $\varkappa$ varies from $3 - 2\sqrt{2} = 0.171573$ to $1 - 1/\sqrt{2} = 0.292893$ as η varies from 0 to 1. Figure 2 plots $\varkappa$ as a function of η where the two constants are Lebowitz–Penrose's lower bound ($\varkappa = 0.144767$) and the threshold ($\varkappa = 0.278465$) established for nonnegative potentials by Lagrange inversion formula (see Section 5, equation (5.14)).

The second part of Theorem 4.1 is an immediate consequence of the observation after equation (2.14). After the completion of its proof, we shall comment on the asymptotic behavior of $\varphi(t,\rho)$ as t tends to 0 and as t tends to ∞.

Proof of Theorem 4.1. The proof will be divided into two parts. First, the standard Cauchy-majorant method is applied to (2.7). We then elaborate the method to push it further—not as far as it can go, but enough to go beyond the threshold.

Solution of (2.7) in power series

The nonlinearity of (2.7), being more severe than that of equation (2.4), requires the control of convolutions of arbitrary large order (compare equation (3.4) with (4.5) and (4.6)). In Appendix D of Brydges and Marchetti (2014), we introduce a general majorant method capable of dealing with equation of the form $u_t + \mathcal{A}(t, u_x, u_{xx}) = 0$, $\mathcal{A}(t,a,b) = \sum_{n,m\geq 0} A_{n,m}(t) a^n b^m$ with $A_{n,m}(t)$ satisfying certain conditions so that Mayer solution $u(t,x) = \sum_{n\geq 1} u_n(t) e^{nx}$ of the initial value problem with $u(0,x) = e^x$ exists globally. The method is able of taming convolutions but it does not produce the refined estimates we need on the existence domain. We observe that for a solution of this kind of equations, in power series of $z = e^x$, $\partial/\partial x$ acts over a function of z as $z\partial/\partial z$. In equation (2.7), ρ plays the role of the z variable and we write $\mathcal{A} = \mathcal{A}(\rho, a, b)$ with a and b in the place of $\rho\varphi_\rho$ and $\rho(\rho\varphi_\rho)_\rho$, respectively.[4]

Since $\rho^2\varphi_{\rho\rho} = \rho(\rho\varphi_\rho)_\rho - \rho\varphi_\rho = b - a$, equation (2.7) can be written in the form of a conservation law:

$$\varphi_t + \big(\mathcal{J}(\rho, \rho\varphi_\rho)\big)_\rho = 0 \tag{4.3}$$

with

$$\mathcal{J}(\rho, a) = \frac{\rho^2}{2} + \varepsilon\rho\left(\frac{1}{1-a} - 1\right), \tag{4.4}$$

and initial condition $\varphi(0, \rho) \equiv 0$. Note that $\mathcal{A} = \mathcal{J}_\rho + \mathcal{J}_a \cdot (\rho\varphi_\rho)_\rho = \rho + \varepsilon((1 + b - a)/(1-a)^2 - 1)$.

Integral equation for the $\boldsymbol{\beta_k}$

For convenience, we introduce a sequence $\boldsymbol{\gamma} = (\gamma_k)_{k\geq 1}$, with $\gamma_k = k\beta_k$, and the convolution product $\boldsymbol{\gamma} * \boldsymbol{\delta}$ of two sequences $\boldsymbol{\gamma}$ and $\boldsymbol{\delta}$ is defined by $(\boldsymbol{\gamma} * \boldsymbol{\delta})_k = \sum_{j=1}^{k-1} \gamma_{k-j}\delta_j$.[5] The power series (1.12) of φ substituted into the integral of (4.3) over $[0, \rho]$ yields a system of first order differential equations

$$\frac{1}{k(k+1)}\dot{\gamma}_k + \varepsilon\gamma_k = -h_k(\gamma_1, \ldots, \gamma_{k-1}; t), \qquad k \geq 1, \tag{4.5}$$

with $\gamma_k(0) = 0$, where the h_k are given by $h_1 = 1/2$ and, for $k \geq 2$,

$$h_k(\gamma_1, \ldots, \gamma_{k-1}; t) = \varepsilon \sum_{n=2}^{k} (\underbrace{\boldsymbol{\gamma} * \cdots * \boldsymbol{\gamma}}_{n})_k. \tag{4.6}$$

The solution of (4.5) for $k = 1$:

$$\tfrac{1}{2}\dot{\gamma}_1 + \varepsilon\gamma_1 = -\tfrac{1}{2}$$

with $\gamma_1(0) = 0$ is

$$\gamma_1(t) = \frac{-1}{2\varepsilon}\big(1 - e^{-2\varepsilon t}\big) = -\lambda \tag{4.7}$$

with $\lambda = \lambda(t, |\Lambda|)$ the function defined by (2.2) (recall $\varepsilon = 1/(2|\Lambda|)$).

By the variation of constants formula, (4.5) is equivalent to a system of integral equations

$$\gamma_k(t) = -k(k+1)\int_0^t e^{-\varepsilon k(k+1)(t-s)} h_k(\gamma_1, \ldots, \gamma_{k-1}; s)\, ds, \qquad k \geq 2 \tag{4.8}$$

[4] Equation (2.7) has a linear dependence on ρ and no explicit dependence on t.

[5] Observe that our sequences $\boldsymbol{\gamma} = (\gamma_k)_{k\geq 1}$ start from $k = 1$.

which can be evaluated recursively starting from $\gamma_1(t) = -\lambda$. For $k = 2$, $h_2 = \varepsilon\gamma_1^2(t)$ and

$$\gamma_2(t) = -2\varepsilon\lambda^3. \tag{4.9}$$

The integral in the r.h.s. of (4.8) retaining in (4.6) only the terms with n equal to $k-1$ and k can be performed explicitly (see Proposition E.1 in Appendix E of Brydges and Marchetti (2014)).

An exact equation satisfied by the majorant

To approach equation (2.7) by the Cauchy-majorant method, we need the following (stronger than usual).

Definition 4.2. A function

$$\Phi(t,\rho) = \sum_{n=1}^{\infty} \Phi_n(t)\rho^n \tag{4.10}$$

is a majorant of $\varphi(t,\rho)$, whose power series in ρ is given by (1.12), if:

1. each $\Phi_n(t)$ is positive, continuous and monotone increasing function of t;
2. there exists a family of domains $\Omega_t = [0,t) \times \mathbb{D}_{r(t)}$, in which the series (4.10) converges absolutely in $\mathbb{D}_{r(t)}$ and

$$|\beta_n(s)| \le \Phi_n(s) \tag{4.11}$$

is satisfied for $0 \le s < t$, uniformly in ε.

We write $\varphi \ll \Phi$ for the majorant relation.

We observe that the majorant relation is preserved by derivative w.r.t. ρ, integration in both t and ρ, convex combination, multiplication and composition (van der Hoeven (2003)).

We start with

$$|\gamma_1(t)| = \lambda\bigl(t, (2\varepsilon)^{-1}\bigr) = \Phi_1(t) \tag{4.12}$$

and generate a recursive equation for $(\Phi_k)_{k\ge 1}$ through (4.8). For convenience, we introduce a majorant

$$\Psi = \rho\Phi_\rho = \sum_{k=1}^{\infty} \Psi_k \rho^k, \qquad \Psi_k = k\Phi_k, \tag{4.13}$$

of $\rho\varphi_\rho$ and observe that $\varphi \ll \Phi \iff \rho\varphi_\rho \ll \Psi$. In our first attempt of constructing Ψ, we shall not push the method to its limit.

Assuming (4.11) holds for $1 \le n \le k-1$, (4.8) can be bounded as

$$\begin{aligned} \left|\gamma_k(t)\right| &\le k(k+1)\int_0^t e^{-\varepsilon k(k+1)(t-s)} h_k\big(|\gamma_1|,\ldots,|\gamma_{k-1}|;s\big)\,ds \\ &\le \frac{1}{\varepsilon} h_k(\Psi_1,\ldots,\Psi_{k-1};t) := \Psi_k(t). \end{aligned} \tag{4.14}$$

For $k=2$, by (4.9), we can do better:

$$\left|\gamma_2(t)\right| = 2\varepsilon\lambda^3(t) = \frac{1}{\varepsilon} h_2(\Psi_1;t) - \eta(t)\lambda^2 := \Psi_2(t), \tag{4.15}$$

where we have used $1-2\varepsilon\lambda = e^{-2\varepsilon t} = \eta(t)$.

Summing the above recursive relation for the Ψ_k, multiplied by ρ^k:

$$\sum_{k\ge 2}\Psi_k\rho^k = \frac{1}{\varepsilon}\sum_{k\ge 2} h_k(\Phi_1,\ldots,\Phi_{k-1};t)\rho^k - \eta\lambda^2\rho^2,$$

for t fixed, results

$$\Psi - \Psi_1\rho = \frac{1}{1-\Psi} - 1 - \Psi - \eta\lambda^2\rho^2 \tag{4.16}$$

by (4.13), (4.6) and (4.4). We observe that (4.16) is equivalent to a quadratic polynomial equation: $2\Psi^2-(1+r-\eta r^2)\Psi + r - \eta r^2 = 0$, with $r=\lambda\rho$, whose solution yields

$$\Psi(t,\rho) = H(\lambda\rho) \tag{4.17}$$

with

$$H(r) = \tfrac{1}{4}\Big(1+r-\eta r^2 - \sqrt{\big(1+r-\eta r^2\big)^2 - 8\big(r-\eta r^2\big)}\Big), \tag{4.18}$$

where we have chosen the branch of the square root in (4.18) for which every coefficient c_k of the power series $\sum_{k\ge 1} c_k r^k$ of H in r remains positive for $0 \le \eta < 1$.

The discriminant polynomial $p(r) = \eta^2 r^4 - 2\eta r^3 + (1+6\eta)r^2 - 6r + 1$ of the quadratic equation has four roots:

$$r_{\pm,\pm} = \frac{1 \pm \sqrt{1-(12\pm 8\sqrt{2})\eta}}{2\eta}.$$

The nearest to the origin $r_{-,-} = r_{-,-}(t,\varepsilon)$, determines the radius of convergence of H. So, the power series of H in r converges provided $|r| < r_{-,-}$.

It follows from (4.17) that the radius of convergence of Ψ (consequently, of Φ too) is $r_{-,-}/\lambda$. It is clear from definition (4.14), (4.12) and (4.15) that $\Psi_k(t)$ is monotone increasing function of t, uniformly in $\varepsilon \in \mathbb{R}_+$. The inequality (4.14) thus proves that Ψ is a majorant of $\rho\varphi_\rho$ in the sense of Definition 4.2 with $r(t) = r_{-,-}(t,\varepsilon)/\lambda(t,1/2\varepsilon)$.

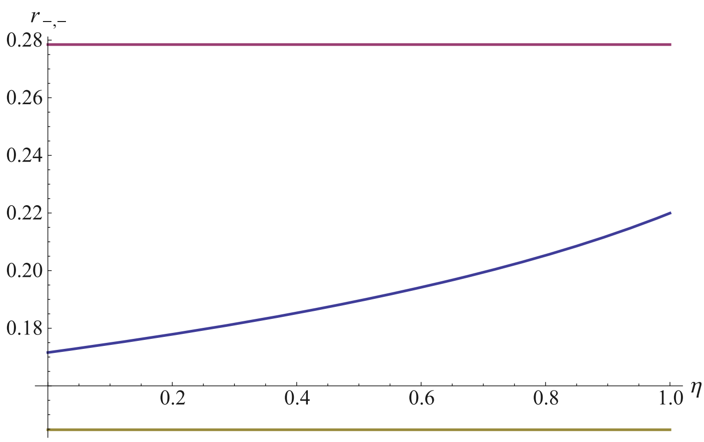

Figure 3 *Plot of $r_{-,-}$ as a function of η.*

As one can see from Figure 3, the improved definition (4.15) of Ψ_2 yields a radius of convergence lying above Lebowitz–Penrose's lower-bound (equation (5.12) with $\kappa = 1$: $\lambda\mathcal{R} \geq 0.144767$). Since $r_{-,-}(t, \varepsilon)$ has not reached the threshold 0.278465 for any $t, \varepsilon \in \mathbb{R}_+$, the improvement, however, is not enough.

Improved majorant equation

We come to the second part of the proof. The kth and $(k-1)$th terms of $h_k(\gamma_1, \dots, \gamma_{k-1}; t)$, $l_k(\gamma_1; t) = \varepsilon\gamma_1^k$ and $m_k(\gamma_1, \gamma_2; t) = \varepsilon(k-1)\gamma_1^{k-2}\gamma_2$ depend only on $\gamma_1 = \beta_1 = -\lambda$ and $\gamma_2 = 2\beta_2 = -2\varepsilon\lambda^3$, by (4.7), (4.9). Their integral

$$\begin{aligned} \mathcal{S}_k(t) &:= k(k+1)\frac{1}{\lambda^k(t)} \\ &\quad\times \int_0^t e^{-\varepsilon k(k+1)(t-s)}\big(l_k(|\gamma_1|; s) + m_k(|\gamma_1|, |\gamma_2|; s)\big)\, ds \end{aligned} \tag{4.19}$$

will be estimated more accurately than the respective integral for the remaining terms

$$\tilde{h}_k(\gamma_1, \dots, \gamma_{k-1}; t) = h_k(\gamma_1, \dots, \gamma_{k-1}; t) - l_k(\gamma_1; t) - m_k(\gamma_1, \gamma_2; t). \tag{4.20}$$

To improve (4.14) for $k \geq 3$, let $\boldsymbol{\delta} = (\delta_k)_{k\geq 1}$ be given by $\gamma_k(t) = \lambda^k(t)\delta_k(t)$, and note that $\sum_{k\geq 1}\delta_k r^k = \rho\varphi_\rho(t, \rho)$, $r = \lambda\rho$. Equation (4.8) can be written as $\delta_1 = -1$, $\delta_2 = -(1-\eta)$,

$$\begin{aligned} \delta_k(t) &= -\big(1 - \eta(t)\big)\frac{k(k+1)}{2}\frac{1}{\lambda^{k+1}(t)} \\ &\quad\times \int_0^t e^{-\varepsilon k(k+1)(t-s)}\lambda^k(s)\frac{1}{\varepsilon}h_k(\delta_1, \dots, \delta_{k-1}; s)\, ds, \end{aligned} \tag{4.21}$$

for $k \geq 3$, and a majorant $\Psi(t,\rho) = \sum_{k\geq 1} C_k r^k$ of $\rho\varphi_\rho(t,\rho)$ can be constructed in the sense of Definition 4.2 for the variable r. Supposing that $|\delta_n(s)| \leq C_n(s)$ holds for $1 \leq n \leq k-1$, equation (4.21) can be majorized by

$$\begin{aligned} \left|\delta_k(t)\right| &\leq \frac{1}{\varepsilon}\tilde{h}_k(C_1,\ldots,C_{k-1};t) + \mathcal{S}_k(t) \\ &\leq \frac{1}{\varepsilon}h_k(C_1,\ldots,C_{k-1};t) - \mathcal{T}_k(t), \end{aligned} \tag{4.22}$$

where, by (4.19), (4.20) and $\lambda(t) = (1-\eta(t))/2\varepsilon$,

$$\mathcal{T}_k(t) = \mathcal{Q}_k(t) + (k-1)\big(1-\eta(t)\big)\mathcal{Q}_{k+1}(t) \tag{4.23}$$

with

$$\mathcal{Q}_k(t) = 1 - \big(1-\eta(t)\big)\frac{k(k+1)}{2}\frac{1}{\lambda^{k+1}(t)}\int_0^t e^{-\varepsilon k(k+1)(t-s)}\lambda^k(s)\,ds \tag{4.24}$$

is such that:

Lemma 4.3. *For every $k \geq 3$, the function defined by (4.23), as a function of η:*

$$\mathcal{T}_k(t) = T_k \circ \eta(t)$$

is a positive (concave) polynomial of degree $k(k+1)/2+1$ which satisfies $T_k(0) = 0$, $T_k(1) = 1$ and

$$T_k(\eta) > \eta, \qquad 0 < \eta < 1. \tag{4.25}$$

The proof of Lemma 4.3 follows immediately from Proposition E.1.i–iv of Brydges and Marchetti (2014), whose statements and proof are omitted in the present work. A positive polynomial is a polynomial with positive coefficients. Proposition E.1 is the most technical result of Brydges and Marchetti (2014), which is also useful in the proof of Theorem 3.2.

Referring to (4.14), if Lemma 4.3 is applied to (4.22), the majorant sequence $(\Psi_k)_{k\geq 1}$ can be redefined for $k \geq 2$ as:

$$\left|\gamma_k(t)\right| \leq \frac{1}{\varepsilon}h_k(\Psi_1,\ldots,\Psi_{k-1};t) - \eta\lambda^k := \Psi_k(t); \tag{4.26}$$

summing (4.26) multiplied by ρ^k, yields an algebraic equation, given by (4.16) with $\eta\lambda^2\rho^2$ replaced by $\eta\lambda^2\rho^2/(1-\lambda\rho)$, equivalent to a quadratic polynomial equation for Ψ, whose solution is given by (4.17) with $H(r)$ replaced by

$$H_1(r) = \frac{1}{4}\left(1 + r - \eta\frac{r^2}{1-r} - \sqrt{\left(1 + r - \eta\frac{r^2}{1-r}\right)^2 - 8\left(r - \eta\frac{r^2}{1-r}\right)}\right).$$

As long as $|r| < 1$, the discriminant polynomial $p_1(r) = (1-r^2-\eta r^2)^2 - 8(r(1-r)^2 - \eta r^2(1-r))$ has four real roots:

$$R_{\sigma,\sigma'}(t,\varepsilon) = \frac{2-\sigma'\sqrt{1-\eta}+2\sigma\sqrt{1-\eta/2-\sigma'\sqrt{1-\eta}}}{1+\eta}, \qquad \sigma,\sigma' \in \{-1,1\}, \tag{4.27}$$

the smallest one, $R_{-,-}$, together with the threshold: $r = 0.28952$, has been depicted in Figure 2 as a function of η.

It follows from (4.26) that $\Psi_k(t)$ is positive and monotone increasing function of t, which proves that Ψ is a majorant of $\rho\varphi_\rho$ in the sense of Definition 4.2. Moreover, as $H_1|_{\eta=1}(r) = r$ for $0 \le r \le 1 - 1/\sqrt{2}$, all coefficients of the power series of $\rho\Phi_\rho$ in $\lambda\rho$, except the first one, vanish in this limit since they are proportional to $(1-\eta)$.

This concludes the proof of Theorem 4.1. □

Summary

We summarize the results obtained so far and present some extensions, conclusions and comparisons.

1. The (normalized) radius of convergence $\lambda\mathcal{R}_\Phi$ for the majorant Φ of φ (r.h.s. of (4.1)) increases monotonously from $3/2-\sqrt{2} = 0.171573$ to $1-1/\sqrt{2} = 0.292893$ as η varies from 0 to 1. We recall that $\lambda = 2B_2$, where B_2 is the second virial coefficient, which is multiplied by the radius of convergence to compare with other models.
2. The radius of convergence for the pressure $P(t,\rho)$, the Helmholtz free energy $f(t,\rho) - f^{\mathrm{ideal}}(\rho) = -\beta(t,\rho)$ and the generating function $\varphi(t,\rho)$ satisfy

$$\lambda\mathcal{R}_P = \lambda\mathcal{R}_\beta = \lambda\mathcal{R}_\varphi \ge \lambda\mathcal{R}_\Phi$$

by (2.14), Proposition 2.1 and (4.11). Inequality (4.2) improves Lebowitz–Penrose lower bound (5.12) for nonnegative potentials (put $\kappa = 1$ and $B = \lambda$ there) and surpasses the threshold 0.278465 for $\eta \ge 0.99463$ or $\varepsilon t \le 0.00538$ (see (5.6) and Theorem 4.3.2 of Ruelle (1969) et seq.).

Asymptotic solution as εt tends to 0

The majorant sequence (4.26) may be modified to improve (4.2) further but we shall not follow this route. Instead, we re-address equation (4.21), satisfied by the coefficients $\delta_k = k\beta_k/\lambda^k$ of the power series of $\rho\varphi_\rho$ in the variable r, in order to obtain their asymptotic limit as εt tends to 0.

Our third conclusion is implied by the following asymptotic behavior of the irreducible cluster integrals (see Theorem 4.1 of Brydges and Marchetti (2014)): $\beta_1 = -\lambda = -t(1+O(\varepsilon t))$,

$$\beta_k(t) = (-1)^{k+1}\lambda^k(t)\varepsilon t\big(1+O(\varepsilon t)\big), \qquad k \ge 2, \tag{4.28}$$

as $\varepsilon t \to 0$, and since the k dependence of the $O(\varepsilon t)$ term is algebraic, we have

$$\varphi(t, \rho) = -t\rho - \frac{t^2\rho^2}{1+t\rho}\varepsilon t\big(1 + O(\varepsilon t)\big)$$

for $t|\rho| < 1$ which, together with (5.4)–(5.6), yields

3. For $t\varepsilon \ll 1$, $\lambda\mathcal{R}_P = 1$ holds even though $\lambda\mathcal{R}_P \geq W(e^{-1}) = 0.278465\ldots$, by the Lagrange inversion theorem.

The stationary solution φ_0 of (2.7)

We compare $\lambda\mathcal{R}_\Phi|_{t=\infty} = 0.171573$ with the radius of convergence $\mathcal{R}_{\psi_0}$ of the power series $\psi_0(\rho) = \sum_{k\geq 1}\tilde{\gamma}_n\rho^k$ whose coefficients $\tilde{\gamma}_k$ solve the system of integral equations (4.8) in the limit as t goes to ∞. Although the (limit) system of integral equations can be solved explicitly, the limit $\rho\varphi_\rho(\infty, \rho) = \lim_{t\to\infty}\sum_{k\geq 1}\gamma_n\rho^k$ is passed inside the sum in the domain $|\rho| < \mathcal{R}_\Phi|_{t=\infty}$, for which (1.12) is known to be uniformly convergent, by Definition 4.2. Consequently,

$$\lambda\mathcal{R}_\Phi|_{t=\infty} \leq \lambda\mathcal{R}_\varphi|_{t=\infty} \leq \lambda\mathcal{R}_{\psi_0}|_{t=\infty} = 1,$$

where the equality results from an explicit solution.

Let $\tilde{\gamma}_k$ be the kth coefficient of the series of $\rho\varphi_0'(\rho)$ in power of ρ, with φ_0 the stationary solution of (2.7). We shall first calculate $\psi_0 = \rho\varphi_0'$. We set $\varphi_t = 0$ in (4.3). $\mathcal{J}(\rho, \rho\varphi_0') = 0$, where $\mathcal{J}$ is given by (4.4), yields

$$\frac{\rho}{2} + \varepsilon\frac{\psi_0}{1-\psi_0} = 0, \tag{4.29}$$

whose solution

$$\psi_0(\rho) = \frac{-\rho}{2\varepsilon}\frac{1}{1-\rho/(2\varepsilon)}, \tag{4.30}$$

implies $\lambda\mathcal{R}_{\psi_0}|_{t=\infty} = 1$. Note that $\lambda(\infty) = 1/(2\varepsilon)$. In view of (4.33), we have:

4.
$$0.171573 \leq \lim_{t\to\infty}\lambda\mathcal{R}_P \leq 1 \tag{4.31}$$

holds in the "low-temperature" limit ($t \to \infty$).

Remark 4.4. Substituting in (4.22) the inequality $T_k(\eta) \geq \frac{67}{45}\eta$, valid for $k \geq 3$ as $\eta \to 0$ (see Figure 3 of Brydges and Marchetti (2014)), the lower bound 0.171573 in (4.31) can be replaced by 0.275451, still far from 1.

Integrating $\varphi_0'(\rho) = \psi_0(\rho)/\rho = (\log(1-\rho/2\varepsilon))'$ with $\varphi_0(0) = 0$, gives

$$\varphi_0(\rho) = \log\Big(1 - \frac{\rho}{2\varepsilon}\Big). \tag{4.32}$$

Replacing $\varphi_0'(\rho)$ into (A.14), taking into account that $\psi_0(\rho) = (\rho + 2\varepsilon \log(1 - \rho/(2\varepsilon)))'$ yields, for $\varepsilon > 0$, the pressure at the stationary (ground) state

$$P_0(\rho) = -2\varepsilon \log\left(1 - \frac{\rho}{2\varepsilon}\right). \tag{4.33}$$

Note that the singularity at $\rho = 2\varepsilon$ of $P_0(\rho)$ is on the (positive) real line, as the one in Ford model (5.16). Summarizing the conclusions of the last two paragraphs, we distinguish three different limits for the pressure of our simple particle system:

$$\begin{aligned}
&\lim_{t\to 0} P(t,\rho) = \rho,\\
&\lim_{\varepsilon\to 0} P(t,\rho) = \rho + \frac{t}{2}\rho^2,\\
&\lim_{t\to \infty} P(t,\rho) = -2\varepsilon \log\left(1 - \frac{\rho}{2\varepsilon}\right)
\end{aligned}$$

attained for ρ on the domain of convergence (see items 3 and 4 above).

Remark 4.5. Equation (4.33) can be rescaled to become independent of ε. As a matter of fact,

$$P_\varepsilon(t,\rho) = \varepsilon P_1\left(\varepsilon t, \frac{\rho}{\varepsilon}\right)$$

holds for any $(t,\rho) \in \mathbb{R}_+ \times \mathbb{C}$ by (2.7) and (2.14). We shall call (4.33) equation of state of Ford's type as it is similar to equation (5.16), for $|\rho| < 1/2$, and to the equation of state of a hard-core lattice gas.

5 Convergence of virial series: Overview

We intend to give an overview on Lebowitz–Penrose's method and to explain with our illustrative example, when it approaches the mean field, how a limitation on the convergence of virial series, due to the unphysical singularity in the majorant of the Mayer series, can be circumvented by a direct approach.

The proof of convergence of virial series by Lebowitz and Penrose (1964) (see also Theorem 4.3.2 et seq. of Ruelle (1969) for nonnegative potentials) remains appealing after 50 years of its publication (Tate (2013)) and has received a great deal of attention Morais and Procacci (2013), despite of the recently new cluster expansion for the canonical partition function Pulvirenti and Tsagkarogiannis (2012) proposed as a direct and natural way of studying the Helmholtz free energy density $f(\rho)$. The straight route has not been used so far and the expansion of Pulvirenti and Tsagkarogiannis (2012), revisited by Morais and Procacci (2013) by means of an expression already known by Mayer, has provided a lower bond for the radius of convergence of $f(\rho)$ identical to Lebowitz–Penrose's lower bound on the radius of convergence of the virial series.

Lebowitz–Penrose's method starts from the Lagrange–Bürmann formula (see, e.g., Theorem 1 of Henrici (1964) and references therein)

$$\wp \circ Z(\rho) = \sum_{n=1}^{\infty} \frac{1}{n} \operatorname{Res}\big(\wp' \rho^{-n}\big)\rho^n, \tag{5.1}$$

where $Z \circ \rho(z) = z$ and $\operatorname{Res}(F)$ means the residue of F at $z = 0$. If $\wp(z)$ and $\rho(z)$ are analytic functions in certain domain D containing the origin, then the residue in (5.1) can be evaluated through an integral around a contour $\mathcal{C}$ inside D (see equations (2.2) and (2.3) of Lebowitz and Penrose (1964)). As a consequence,

$$P(\rho) = \wp \circ Z(\rho) \qquad \text{is holomorphic in } |\rho| < \underline{\rho} := \min_{z\in\mathcal{C}} \big|\rho(z)\big| \tag{5.2}$$

and the equation $\rho(z) = \rho$ has a unique solution for z, denoted by $Z(\rho)$, holomorphic in $|\rho| < \underline{\rho}$ by Rouche's theorem (see Theorem 9.4.1 of Hille (1982)). We observe that a purely algebraic proof of (5.1) exists in the field of Laurent formal series (Henrici (1964)).

Gas of hard spheres in infinite dimensions

We begin studying the implications of the second Mayer theorem (A.13) for the case of a gas of hard-spheres in infinitely many dimensions (the mean field). In this case, the weight $w(G)$ of a n-block G with $n > 2$ vanishes and the generating function (A.12) is simply given by

$$\varphi(\rho) = \rho \tag{5.3}$$

(compare (A.13) and (5.3) with equation (6) of Frisch, Rivier and Wyler (1985)). We look at (A.13) as a map from the complex ρ-plane to the complex z-plane: $\rho \in \mathbb{C} \longmapsto Z(\rho) \in \mathbb{C}$ given by

$$Z(\rho) = \rho e^{\rho}. \tag{5.4}$$

Figure 4 depicts images of various circles $\{Z(\rho) : |\rho| = 0.1n, n = 1, \ldots, 10\}$, in the complex z-plane. Note the formation of a cusp at $-e^{-1}$ (the image $Z(\rho)$ of $\rho = -1$) as a consequence of the fact that $Z'(\rho) = (1+\rho)e^{\rho}$ vanishes at $\rho = -1$ (the derivative $\rho'(z) = 1/Z' \circ \rho(z)$ diverges at $z = -e^{-1}$). Recall that an analytic function $f : D \longrightarrow \mathbb{C}$ is *univalent* in an open domain D if $f(\rho_1) \neq f(\rho_2)$ for all $\rho_1, \rho_2 \in D$ with $\rho_1 \neq \rho_2$. Hence, $Z(\rho)$ is univalent in any open disk $\mathbb{D}_\tau$ centered at origin with radius $\tau \leq 1$.

The inverse $\rho(z)$ of $Z(\rho)$ is the Lambert W-function $W(z)$, a multivalued function whose principal branch $W_0(z)$ is defined in the slit domain $\mathbb{C} \setminus (-\infty, e^{-1}]$ (see the construction of $W_0(z)$ in Figures 5 and 6 of Corless et al. (1996)). We observe that the circles with $|\rho| < 1$ are inside the image $W_0(\mathbb{C} \setminus (-\infty, e^{-1}])$ and $z \longmapsto \rho(z)$ is, in fact, a conformal (bijective) map from $\mathbb{C} \setminus (-\infty, e^{-1}]$ to the latter image domain, whose boundary is described by the curve $\xi = -\eta \cot \eta$, with $\rho = \xi + i\eta$.

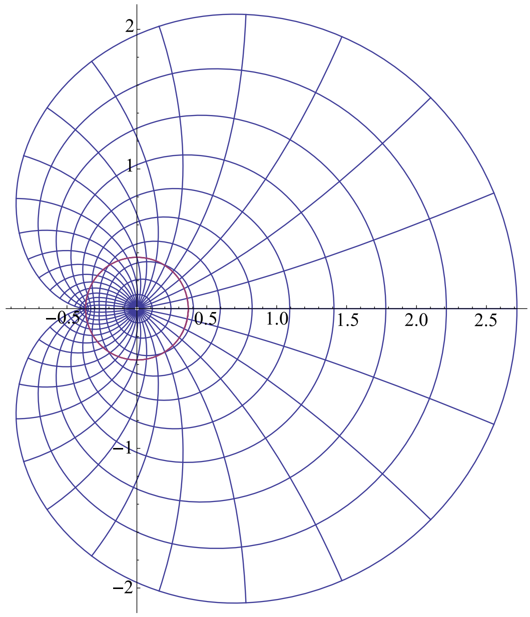

Figure 4 *Image of circles under $\rho \longmapsto Z(\rho)$ together with a circle of radius e^{-1} centered at the origin.*

The Mayer series $\rho(z) = \sum_{n\geq 1} nb_n z^n$, for the density of the hard-sphere gas in the infinite dimension limit, is a sum of free diagrams which may be evaluated from $\rho(z) = W_0(z)$ by the Lagrange inversion formula. Replacing $\wp$ by identity and $Z(\rho)$ by its inverse $\rho(z) = W_0(z)$ in (5.1), we have

$$nb_n = \frac{1}{n}\operatorname{Res}\big(z^{-n}e^{-nz}\big) = \frac{1}{n!}\frac{d^{n-1}}{dz^{n-1}}e^{-nz}\Big|_{z=0} = \frac{(-n)^{n-1}}{n!}, \tag{5.5}$$

by (5.4); its radius of convergence is thus $r = e^{-1}$ (see Section 3 of Corless et al. (1996) for an alternate proof). Now, suppose we want to study the radius of convergence of the virial series taking into account solely that $\wp(z)$ given by (A.6) has the same radius of convergence of $\rho(z)$. Using the bijection, $P(\rho) = \wp \circ Z(\rho)$ is holomorphic in a disk $\mathbb{D}_{r'}$ of radius r' in the ρ-plane such that $|Z(\rho)| < e^{-1}$ holds for every $\rho \in \mathbb{D}_{r'}$, that is, $Z(\mathbb{D}_{r'}) \subset \mathbb{D}_{e^{-1}}$. Since $|Z(\rho)| \leq |\rho|e^{|\rho|}$ holds as equality for $\rho \geq 0$ (with no absolute values), the inverse of Z restricted to the positive real axis yields

$$r' = W\big(e^{-1}\big) = 0.278465 \tag{5.6}$$

(see Figure 4, where the image of two (almost three) circles of radius $i/10$, $i = 1, 2$, are inside the disk $\mathbb{D}_{e^{-1}}$) despite of $Z(\rho)$ having a radius of univalence $\tau = 1$.

The virial series can, however, be derived directly from (5.4). Using (A.14), (5.3) and $P(0) = 0$, we have

$$\beta P(\rho) = \int_0^\rho \left(1 - \tilde{\rho}\varphi'(\tilde{\rho})\right) d\tilde{\rho} = \rho + \frac{1}{2}\rho^2$$

which is the equation of state (1.1). We observe that the image of circles: $P(\tau e^{it})$, $-\pi \le t < \pi$, of radius τ centered at origin, are simple curves for $\tau < 1$ which approaches a cardioid at $\tau = 1$ with a cusp at $\rho = -1$. So, the radius of univalence of $P(\rho)$ is also $\tau = 1$.

It was our inability of evaluating the virial series, as a formal power series through the Lagrange–Bürmann formula (5.1), which led us to an estimate on the radius of convergence (5.6), missing all hidden cancellations manifested in (5.3). It turns out that in this rare case the virial series may be calculated explicitly but in most cases this resource is not available. We note that the estimate (5.6) is determined by the singularity at $z = -e^{-1}$ in the Mayer series and no trace of this singularity is seen in the virial series, which is a polynomial of second degree. We shall come back to those issues in the next paragraph where we shall discuss lower bounds on the radius of convergence of the virial series when we approach to the mean field.

Remark 5.1. If equation (5.3) is replaced by $\varphi(\rho) = \lambda\rho$, then the same conclusions hold with r' in (5.6) replaced by $\lambda r'$. For this, we have $\lambda Z(\rho) = \lambda\rho e^{\lambda\rho}$ whose inverse is $\lambda\rho(z) = W_0(\lambda z)$ and $\beta\lambda P(\rho) = \lambda\rho + (\lambda\rho)^2/2$. In Frisch, Rivier and Wyler (1985) z, ρ and p are multiplied by its proper natural scale υ_0, the volume of a d-dimensional sphere with radius a, the diameter of the hard-sphere, prior taking the dimension to infinity.

Lebowitz–Penrose lower bound

The Lambert W-function plays an important rule as well for a system of particles interacting through a pair potential ϕ satisfying (i) (*stability*)

$$\exists\Phi \ge 0: \qquad \sum_{1\le i<j\le n} \phi(x_i - x_j) \ge -n\Phi \tag{5.7}$$

for every $n \ge 2$ and $(x_1, \dots, x_n) \in \mathbb{R}^{nd}$; and (ii)

$$\left\| e^{-\beta\phi} - 1 \right\|_1 = B(\beta) < \infty, \tag{5.8}$$

with $\|f\|_1 = \int_{\mathbb{R}^d} |f(x)|\, dx$ the L^1-norm in $\mathbb{R}^d$. A ϕ satisfying (i) and (ii) is called regular potential. For systems with regular potentials, Penrose's estimate (Penrose (1967)) yields

$$\left|\rho_\Lambda(z) - z\right| \le \frac{-1}{\kappa^2 B} W\left(-\kappa B|z|\right) - \frac{|z|}{\kappa}, \tag{5.9}$$

where $\kappa = \exp(2\beta\Phi) \geq 1$, uniformly in Λ, provided

$$we^{-w} \equiv \kappa B|z| < e^{-1}. \tag{5.10}$$

Note that $0 \leq w < 1$ preserves this inequality. Equation (5.9), together with $W(-we^{-w}) = -w$, implies

$$\begin{aligned} |\rho_\Lambda(z)| &\geq \left(1 + \frac{1}{\kappa}\right)|z| + \frac{1}{\kappa^2 B} W(-\kappa B|z|) \\ &= \left((1+\kappa)e^{-w} - 1\right)\frac{w}{\kappa^2 B} \end{aligned} \tag{5.11}$$

which, by maximizing in w satisfying (5.10), yields (see Lebowitz and Penrose (1964), equation (3.8) et seq.)

$$|\rho_\Lambda(z)| \geq 0.28952 \frac{1}{(1+\kappa)B} \equiv R_0. \tag{5.12}$$

Applying Lagrange–Bürmann formula (5.1), together with $\beta z \wp'(z) = \rho(z)$, the coefficients of virial series (A.16) reads

$$B_n = \frac{1}{n}\operatorname{Res}\left(\beta\wp'\rho^{-n}\right) = \frac{1}{n}\frac{1}{2\pi i}\oint_{|z|=\delta_0} \rho^{-n+1}(z)\frac{dz}{z} \leq n^{-1}R_0^{-n+1}. \tag{5.13}$$

Equivalently, in view of (A.18),

$$\beta_n \leq n^{-1}R_0^{-n}.$$

By the ratio test, R_0 is thus a lower bound for the radius of convergence $\mathcal{R}_P$ of the virial series. Recently, re-visiting the cluster expansion proposed by Pulvirenti and Tsagkarogiannis (2012), together with Penrose's estimate on the Mayer coefficients, Morais and Procacci (2013) have reached to the same lower bound for the radius of convergence of the Helmholtz free energy in powers series of the density $\rho = N/|\Lambda|$ (see Theorem 1 and Remarks 2 and 4 therein).

Apart from the factor $1+\kappa \geq 2$, which came into (5.11) replacing the inequality $-|\rho_\Lambda(z)| + |z| \leq ||\rho_\Lambda(z)| - |z|| \leq |\rho_\Lambda(z) - z|$ in the l.h.s. of (5.9), the constant 0.28952 appearing in both, (5.12) and Morais and Procacci (2013), is very near to r' defined by (5.6). If Lagrange's theorem is applied to the hard-spheres gas in infinitely dimensions, whose density is exactly given by $\rho(z) = W_0(z)$, the same estimate r' on the radius of convergence of its virial series is obtained (see (5.2) and equation (3.1) of Lebowitz and Penrose (1964)):

$$\mathcal{R}_P^{\text{h.s.}} \geq \underline{\rho} = \min_{0\leq t<2\pi} |\rho(e^{-1+it})| = W_0(e^{-1}), \tag{5.14}$$

where the contour $\mathcal{C}$ has been chosen on the domain $e|z| \leq 1$ such that the minimum value on $\mathcal{C}$ is the largest possible (see Figure 5). However, repeating the same steps (5.9)–(5.12),

$$|\rho_\Lambda(z) - z| \leq -W_0(-|z|) - |z|$$

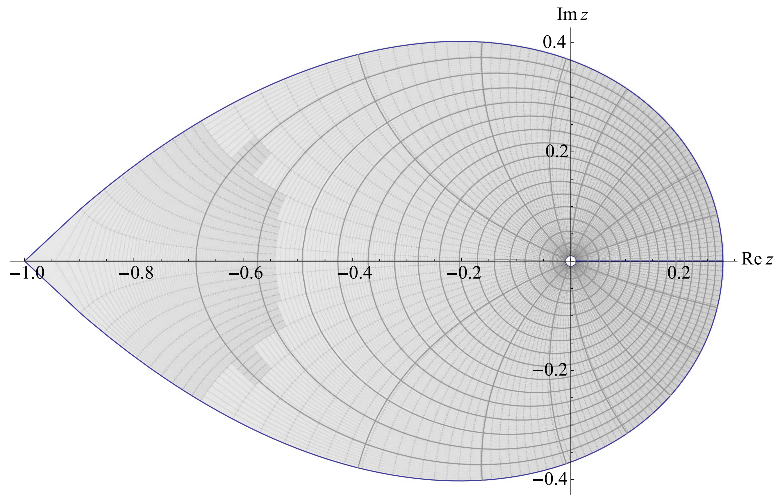

Figure 5 *Image of circles under* $z \longmapsto \rho(z)$.

with $\mathcal{C} = \{z = re^{it}, 0 \le t < 2\pi\}$ and $we^{-w} \equiv r < e^{-1}$, yields an estimate $0.52 \times r'$ (instead of $0.5 \times r'$):

$$\begin{aligned}\left|\rho_\Lambda(z)\right| &\ge \max_{0 \le r < e^{-1}} \big(2r + W_0(-r)\big) \\ &= \max_{w \ge 0} \big(2e^{-w} - 1\big) w \simeq 0.14476 = \frac{0.28952}{2}.\end{aligned}$$

The numerical factor that multiplies r' has increased slightly because the curve $\mathcal{C}$ in the Lagrange's theorem is chosen to minimize the amount lost in replacing the coefficients of $\rho_\Lambda(z) - z$ by their absolute values. We have done something similar in our majorant method in Section 4 when (4.14) is replaced by (4.26). Despite of the loss caused by the method, the lower bound on $\mathcal{R}_P(t)$ is above Lebowitz–Penrose's estimate for nonnegative potentials and exceeds the threshold r' (unsurpassed by Lagrange inversion formula) for all t, provided ε is small enough. This means that the mean field (unphysical) singularity of the Lambert W-function has been circumvented by our method.

Example of an equation of state presenting a plateau

The purpose here is review an explicit example in which the condensation phenomenon is not determined by the singularities present on $P(\rho)$.

The Yang–Lee theory (Yang and Lee (1952)) is capable to explain condensation directly from the partition function (A.1). To illustrate how this phenomenon takes place, Uhlenbeck and Ford (see Section III.4 of Uhlenbeck and Ford (1963)) have devised an artificial example in which the grand-canonical partition function is given by

$$\Xi_\Lambda(z) = (1+z)^{|\Lambda|}\frac{1-z^{|\Lambda|}}{1-z}$$
$$= \exp\left(|\Lambda|\log(1+z) + \sum_{n=0}^{|\Lambda|-1}\log\big(1-ze^{-2\pi in/|\Lambda|}\big) - \log(1-z)\right).$$

One sees that

$$\wp(z) = \lim_{|\Lambda|\to\infty}\frac{1}{|\Lambda|}\log\Xi_\Lambda(z) = \begin{cases}\log(1+z), & \text{if } |z|\le 1,\\ \log(1+z)+\log z, & \text{if } |z|>1\end{cases} \tag{5.15}$$

is the logarithmic potential due to one unit of charge at $z=-1$ and one unit of charge uniformly distributed on the unit circle. The pressure $P(\rho)=\wp\circ(z\wp')^{-1}(\rho)$ in the so-called Ford model does present a plateau

$$P(\rho) = \begin{cases}\log\big(1/(1-\rho)\big), & \text{if } 0\le\rho<1/2,\\ \log 2, & \text{if } 1/2\le\rho<3/2,\\ \log(\rho-1)/(2-\rho)^2, & \text{if } 3/2\le\rho<2,\end{cases} \tag{5.16}$$

although Padé approximation (Aguillera-Navarro et al. (1984), Clisby and McCoy (2006)) is unable to detect the singularity on its virial expansion.

Although the power series of $P_1(\rho)=\log(1/(1-\rho))$ converges for $|\rho|<1$, the image of $\Re\mathrm{e}\,\rho>1/2$ by $Z_1(\rho)=(z\wp_1')^{-1}(\rho)=\rho/(1-\rho)$ is on the complement $\mathbb{C}\setminus\mathbb{D}_1$ of the unit disk $\mathbb{D}_1$, where $\wp_1(z)=\log(1+z)$ is defined ($P(\rho)$ is, indeed, not defined at any point ρ of the forbidden domain $1/2<\Re\mathrm{e}\,\rho<3/2$) (Presutti (2009)). The presence of plateau results from the convex envelop of the Helmholtz free energy, defined by (A.20) in each of the two branches:

$$f(\rho) = \begin{cases}\rho\log\rho+(1-\rho)\log(1-\rho), & \text{if } 0\le\rho<1/2,\\ -\log 2, & \text{if } 1/2\le\rho<3/2,\\ (\rho-1)\log(\rho-1)+(2-\rho)\log(2-\rho), & \text{if } 3/2\le\rho<2\end{cases}$$

with the horizontal line being tangent to both curves $(\rho, f_i(\rho))$, $i=1,2$, the first at $\rho=1/2$ and the second at $\rho=3/2$.

6 Conclusions and open problems

The virial series of a particle system interacting though uniformly (Kac-like) repulsive interaction has been directly addressed by the method of Cauchy together with asymptotic methods and its radius of convergence is shown to exceed the threshold established by the application of Lagrange inversion formula for the case of non-negative potentials.

The approach to the mean field equation (1.1) through the PDEs (1.5) and (1.13) may be considered the first step in the resolution of the problem posed by the authors of Kac, Uhlenbeck and Hemmer (1963): what is the relation of the approach to a mean field theory (presenting vapor–liquid phase transition) with the Ursell–Mayer theory? The ingredient needed for this is the energy of interacting particles through Kac potential in the mean field limit ($\gamma \to 0$), a problem that remains open even for one-dimensional case.

Whether the virial series is capable to detect phase transition of van der Waals vapor–liquid type remains an open challenging problem. The virial series for the Ford model is, however, known to be convergent beyond the condensation point and other thermodynamic functions would be more appropriate than the pressure or the Helmholtz free-energy. The Gibbs free-energy is the Legendre transform of the Helmholtz free-energy w.r.t. the specific volume $v = 1/\rho$ and a relation of the coefficients of the power series in p with the irreducible cluster integrals could in principle be established. This open problem suggested in Kac, Uhlenbeck and Hemmer (1963) is interesting even for particles interacting through uniformly repulsive pair potential for which is possible to write an equation for the Gibbs free-energy.

Appendix: Irreducible cluster integrals and their relation to the virial coefficients

Some of notations, definitions and results referred in our text regarding the virial expansion are briefly touched in the present appendix and we suggest Uhlenbeck and Ford (1963), Leroux (2004), and references therein, for details.

Mayer series

The grand-canonical ensemble of interacting particles in a container Λ, a regular domain in $\mathbb{R}^d$ of volume $V = |\Lambda|$, with activity (fugacity) z at the inverse temperature $\beta = 1/(kT)$ has a partition function

$$\Xi_\Lambda(\beta, z) = \sum_{n=0}^{\infty} z^n Q_{\Lambda,n}(\beta), \tag{A.1}$$

where ($Q_{\Lambda,0} = 1$)

$$Q_{\Lambda,n}(\beta) = \frac{1}{n!}\int_{\Lambda^n} e^{-\beta U(x)}\, d^d x_1 \cdots d^d x_n \tag{A.2}$$

is the canonical partition function and $U(x) = \sum_{1\le i<j\le n} \phi(x_i - x_j)$ is the pairwise interacting energy of a configuration $x = (x_1, \ldots, x_n) \in \Lambda^n$ of n particles.

In the course of the present work, we have used two basic combinatorial tools of the theory of Mayer and virial expansion know as the first and second Mayer theorems. To state them, define $f_{ij} = e^{-\beta\phi_{ij}} - 1$ with $\phi_{ij} \equiv \phi(x_i - x_j)$ and write the Boltzmann factor as a sum over the set $\mathcal{M}$ of Mayer (simple linear) graphs G in the set $\{1, 2, \ldots, n\}$ of labelled vertices:

$$e^{-\beta U(x)} = \prod_{1\le i<j\le n} (1 + f_{ij}) = \sum_{G\in\mathcal{M}} \prod_{(ij)\in E} f_{ij}, \tag{A.3}$$

where the product runs over the (ij) in the set of edges $E = E(G)$ of G. The grand-partition function (A.1) can thus be written as

$$\Xi_\Lambda(z) = \sum_{G \in \mathcal{M}} \frac{z^n}{n!} W(G),$$
$$W(G) = \int_{\Lambda^n} \prod_{(ij) \in E(G)} f_{ij}(x)\, d^\sharp x. \tag{A.4}$$

Here $n = n(G)$ is the number of vertices in G, $W(G)$ is the weight of Mayer graph G and the restriction of $d^\sharp x$ to Λ^n is the Lebesgue measure. The first Mayer theorem reads (see Theorem I of Uhlenbeck and Ford (1963)):

Theorem A.1.

$$\log \Xi_\Lambda(z) = \sum_{G \in \mathcal{M}\colon\, G \text{ connected}} \frac{z^N}{N!} W(G). \tag{A.5}$$

Proof. We observe that the weight function (A.4) is independent of the labelling of the N vertices and, for any Mayer graph G whose connected parts are $G_1, \ldots, G_k$, we have $W(G) = W(G_1) \cdots W(G_k)$. These are the ingredients behind its proof, which we refer to Ruelle (1969), Brydges (1986). □

By Theorem A.1, the pressure can be written as a formal power series

$$\beta \wp_\Lambda(\beta, z) = \sum_{n=1}^{\infty} b_{\Lambda,n} z^n, \tag{A.6}$$

where, for $n \geq 1$,

$$b_{\Lambda,n} = \frac{1}{n!} \sum_{\substack{G \in \mathcal{M}\colon\, G \text{ connected},\\ N(G)=n}} \frac{1}{|\Lambda|} \int_{\Lambda^n} \prod_{(ij) \in E(G)} f_{ij}(x)\, d^\sharp x \tag{A.7}$$

are the Mayer coefficients. We omit from now on the dependence on the inverse temperature β to avoid notational conflict.

Irreducible cluster integrals

Referring to (A.4) with G connected, we define $w(G)$ by holding one vertex, let us say x_1, fixed at origin while $x = (x_1, \ldots, x_N)$ is integrated over Λ^{N-1}

$$w(G) = \int_{\Lambda^{N-1}} \prod_{(ij) \in E(G)} f_{ij}(x)\, d^\sharp x. \tag{A.8}$$

Definition A.2. A vertex i_0 is said to be an *articulation point* of a connected Mayer graph G if G becomes disconnected after its removal. A graph G with no articulation points is called a *block* or a *irreducible graph*.

A weight function w is said to be *block-multiplicative* if for any connected Mayer graph G, whose blocks are $G_1, \ldots, G_k$, we have

$$w(G) = w(G_1) \cdots w(G_k). \tag{A.9}$$

We define the Mayer graph consisting of a single vertex to be not a block. The simplest block (2-block) consists of a single edge together with two end points. The next simplest one (3-block) has three vertices and three edges cyclically connected.

Referring to (A.5), with connected Mayer graphs G replaced by blocks, we define, analogously,

$$\beta_\Lambda(\rho) = \sum_{G \in \mathcal{M}:\ G \text{ is a block}} \frac{\rho^N}{N!} w(G) := \sum_{n=2}^{\infty} \frac{1}{n} \beta_{n-1} \rho^n, \tag{A.10}$$

where (A.10) defines the β_{n-1}, which are called *irreducible cluster integrals* of order n. The second Mayer theorem may be stated as (see Theorem II of Uhlenbeck and Ford (1963) for a proof).

Theorem A.3. *The thermodynamic limits* $\rho(z) = \lim_{m\to\infty} \rho_{\Lambda_m}(z)$ *and* $\beta(\rho) = \lim_{m\to\infty} \beta_{\Lambda_{mn}}(\rho)$, $(\Lambda_m)_{m\geq 1}$ *being an increasing sequence of regular domain, satisfy a functional equation*

$$\rho(z) = z e^{\beta' \circ \rho(z)}. \tag{A.11}$$

A simpler proof which holds for formal power series is provided in Leroux's article on combinatorial species (Leroux (2004), Theorem 1.3). The key ingredient is the block-multiplicative property of the weight function (A.8) in the thermodynamic limit (see Proposition 2.2 of Leroux (2004) and Appendix 2 of Pulvirenti and Tsagkarogiannis (2012) for an extension of Theorem A.3 to large but finite volume).

Kamerlingh Onnes virial series

By definition (A.10),

$$\beta'(\rho) = \sum_{n=1}^{\infty} \beta_n \rho^n := \varphi(\rho), \tag{A.12}$$

$\varphi(\rho)$ is the generating function of irreducible cluster integrals $(\beta_n)_{n\geq 1}$, equation (A.11) can be written as

$$z = \rho(z) e^{-\varphi \circ \rho(z)}. \tag{A.13}$$

Taking the derivative in both sides, yields $z\rho'(z) = \rho(z)/(1 - \rho(z)(\varphi' \circ \rho)(z))$ and, together with $\rho(z) = z\wp'(z)$, $P(\rho) = \wp \circ Z(\rho)$ and the rules of Calculus, which hold in the realm of formal power series,

$$\beta P'(\rho) = \frac{\beta \wp'}{\rho'}$$

$$= 1 - \rho\varphi'(\rho) \tag{A.14}$$

$$= 1 - \sum_{l=1}^{\infty} l\beta_l \rho^l. \tag{A.15}$$

The Kamerlingh Onnes virial series

$$\beta P(\rho) = \sum_{n=1}^{\infty} B_n \rho^n \tag{A.16}$$

is thus obtained by integrating term-by-term (A.15):

$$\beta P(\rho) = \rho - \sum_{l=1}^{\infty} \frac{l}{l+1} \beta_l \rho^{l+1} \tag{A.17}$$

which gives, by (A.10),

$$B_n = -(n-1)\beta_{n-1}/n, \tag{A.18}$$

$$\beta_{n-1} = \frac{1}{(n-1)!} \sum_{\substack{G:\ G \text{ is a block} \\ \text{of order } n}} \frac{1}{|\Lambda|} \int_{\Lambda^N} \prod_{(ij) \in E(G)} f_{ij}(x)\, d^\sharp x. \tag{A.19}$$

The Helmholtz free-energy

Alternatively, we may follow another direction already known by Mayer (see, e.g., Mayer and Mayer (1977), equations (13.47)–(13.50)):

Proposition A.4. *The Helmholtz free energy* $f(\rho) = -\log Q_{\Lambda,N}/|\Lambda|$, *with* $\rho = N/|\Lambda|$, *in the thermodynamic limit, is formally given by the Legendre transform*

$$f(\rho) = \sup_{\mu}\big(\mu\rho - \beta\wp(e^{\mu})\big) = \rho\log\rho - \rho - \beta(\rho), \tag{A.20}$$

where $\beta(\rho)$ *is defined by* (A.10). *Hence, by* (A.12), $\varphi(\rho) = \beta'(\rho) = -f'(\rho) + \log\rho$ *generates the irreducible cluster integrals* β_n, $n = 1, 2, \ldots$.

Proof. As a formal power series, $\mu^* = \mu^*(\rho)$ solves the equation $\rho = \beta\wp'(e^{\mu}) \cdot e^{\mu}$ for μ, and $P(\rho) = \wp \circ e^{\mu^*}(\rho)$ is the pressure (as a function of ρ). The two first terms in the r.h.s. of (A.20) are ideal gas contributions: $f^{\text{ideal}}(\rho) = \sup_{\mu}(\mu\rho - e^{\mu}) = \mu^*\rho - e^{\mu^*} = \rho\log\rho - \rho$ by the ideal gas equation of state $\rho = z = e^{\mu}$. The last term of (A.20) is obtained by solving (A.13) for μ with $z = e^{\mu}$ and φ given by (A.12):

$$\mu^* = \mu^*(\rho) = \log \rho - \sum_{n=1}^{\infty} \beta_n \rho^n. \tag{A.21}$$

Replacing μ^* into $f(\rho) = \mu^* \rho - \beta \wp(e^{\mu^*}) = \mu^* \rho - \beta P(\rho)$, together with (A.17) and (A.10), yields (A.20). $\square$

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