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Exact Decay of Correlations for Mixtures and Rotators

J.-R. Fontaine,[1] Ch. Gruber,[1] and A. Sbaï[1]

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We give the exact asymptotic form, at low activity, of the correlations of a classical fluid consisting of several species of particles interacting by means of integrable two-body potentials. Our results also extend to classical dipoles with r^{-3} potential in two dimensions.

KEY WORDS: Continuous systems; mixtures; rotators; long-range forces; clustering.

1. INTRODUCTION

The exact asymptotic form of the correlation functions has been recently obtained for a classical fluid of identical particles interacting by means of a two-body potential with integrable power law decay.[1] In particular it was proved that all truncated correlation functions decay like the potential either for small activities, or for any values (z, β) of activity and temperature such that the state is unique and has some power law decay. For references related to this old problem, we refer to Ref. 1.

In this paper we extend this analysis to the case of a fluid consisting of *several species of particles* interacting by means of integrable two-body potentials $\phi_{\alpha_1\alpha_2}(x_1 - x_2)$, where $\alpha \in \{1,..., N\}$ denotes the species. Assuming that at least one of the potentials has a power law decay—the others could be finite range or have some power law decay—we obtain the exact asymptotic form of all correlation functions. In particular assuming that the slowest decaying potential behaves like $d\,|x|^{-\gamma}$ as $x \to \infty (\gamma > \nu)$ we prove that

$$\lim_{\lambda \to \infty} \lambda^{\gamma} \rho^{T}_{\alpha_1\alpha_2}(\lambda\hat{x}) = -\beta \sum_{\bar{\alpha}_1,\bar{\alpha}_2} K_{\alpha_1\bar{\alpha}_1} d_{\bar{\alpha}_1\bar{\alpha}_2}(\hat{x})\, K_{\bar{\alpha}_2\alpha_2}$$

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[1] Institut de Physique Théorique, École Polytechnique Fédérale de Lausanne, PHB—Ecublens, CH-1015 Lausanne, Switzerland.

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where $\hat{x}$ is a unit vector,

$$d_{\alpha_1\alpha_2}(\hat{x}) = \lim_{\lambda \to \infty} \lambda^\gamma \phi_{\alpha_1\alpha_2}(\lambda\hat{x})$$

and $K_{\alpha\bar{\alpha}}$ is related to the compressibility tensor:

$$K_{\alpha\bar{\alpha}} \equiv z_\alpha \frac{d}{dz_\alpha} \rho_{\bar{\alpha}} = \rho_\alpha \delta_{\alpha\bar{\alpha}} + \int dy\, \rho^T_{\alpha\bar{\alpha}}(y)$$

Note that $d_{\alpha_1\alpha_2}$ could be zero for some pair $(\alpha_1\alpha_2)$. This result implies that the effective potential between any two particles will always behave like the slowest decaying potential, with factors associated to the partial compressibilities.

We shall also consider the case of a fluid consisting of "*rotators*" interacting by means of "dipole-type" interactions, i.e., $\phi_{\omega_1\omega_2}(x) \sim d_{\omega_1\omega_2}(\hat{x})\, |x|^{-\gamma}$ as $|x| \to \infty$, where ω denotes the orientation of the rotator and $d_{\omega_1\omega_2}(\hat{x}) = (\omega_1, A(\hat{x})\, \omega_2)$ with $A(\hat{x})$ a matrix covariant under $SO(d)$ transformations. For example, for dipolar systems and $v = d = 3$, $A_{ij}(\hat{x}) = m^2(3x_i x_j - \delta_{ij})$. In the case of rotators we prove that

$$\lim_{\lambda \to \infty} \lambda^\gamma \rho^T_{\omega_1\omega_2}(\lambda\hat{x}) = -\beta \left[\rho \frac{\varepsilon - 1}{3y}\right]^2 d_{\omega_1\omega_2}(\hat{x})$$

where ρ is the density of particles, y is the usual factor in the theory of dielectrics [$y = (4\pi/9)\, \beta m^2 \rho$ in three dimensions] and

$$\rho \frac{\varepsilon - 1}{3y} = \rho + \int dy \int d\bar{\omega}\, \rho^T_{\omega\bar{\omega}}(y) \cdot (\omega \cdot \bar{\omega})$$

This result is applicable to a system of dipoles with r^{-3} potential in two dimensions.

Let us recall that on the other hand the expected result for three-dimensional dipoles is

$$\lim \lambda^3 \rho^T_{\omega_1\omega_2}(\lambda\hat{x}) = -\beta \left[\rho \frac{\varepsilon - 1}{3y}\right]^2 \frac{1}{\varepsilon} d_{\omega_1\omega_2}(\hat{x})$$

where in the definition of the dielectric constant only the "short-range part of ρ^T" is included.[2]

2. DEFINITIONS AND RESULTS

The system consists of particles in $\mathbb{R}^v$ having internal degrees of freedom. These internal degrees of freedom are labeled by the points of some measurable space Ω with finite measure. To simplify the discussion we restrict ourselves to the case where Ω is discrete, i.e., $\Omega=\{1,..., N\}$ and $\alpha \in \Omega$ labels the different species, and to the case where Ω is the unit sphere in $\mathbb{R}^d$ with the usual invariant normalized measure, i.e., $\Omega = S^{(d)}$ and $\omega \in S^{(d)}$ represents the axis of the rotator. Whenever there is no confusion we also use the notation ω to label the particles. The extension to general Ω [2] and to general domains $\mathscr{D} \subset \mathbb{R}^\nu$ [1] is straightforward.

We denote a one-particle configuration by $q=(x, \omega)$, $x \in \mathbb{R}^\nu$, $\omega \in \Omega$ and write $\int dq = \int_{\mathbb{R}^\nu} dx \int_{S^{(d)}} d\omega$, respectively, $\int dq = \int dx \sum_{\alpha \in \Omega}$. For n-particle configuration we write

$$Q = Q^{(n)} = (q_1,..., q_n) \qquad \text{and} \qquad \int dQ = \int dq_1,..., \int dq_n$$

The particle interacts by means of a two-body potential $\phi(q_1, q_2)$ such that

$$\phi(q_1, q_2) = \phi(q_2, q_1) = \phi_{\omega_1\omega_2}(x_1 - x_2)$$

and having the following properties:

$$\sum_{1 \leqslant i < j \leqslant n} \phi(q_i, q_j) \geqslant -nB \qquad \text{for some } B \geqslant 0 \text{ and all } (q_1,..., q_n) \tag{1a}$$

$$\lim_{\lambda \to \infty} \lambda^\gamma \phi_{\omega_1\omega_2}(\lambda \hat{x}) = d_{\omega_1\omega_2}(\hat{x}) \qquad \text{uniformly with respect to } \hat{x} \tag{1b}$$

where the functions $d_{\omega_1\omega_2}(\hat{x})$ are continuous on the unit sphere $|\hat{x}|=1$ and are not all identically zero.

Notice that (1a) and (1b) imply that γ is the power of the slowest decaying potential and that

$$|e^{-\beta\phi(q_1 q_2)} - 1| \leqslant c(\beta)[|x_1 - x_2|^\gamma + 1]^{-1} \tag{2}$$

$$\int dq_1\, |e^{-\beta\phi(q_1 q_2)} - 1| \leqslant b(\beta) < \infty \tag{3}$$

The equilibrium states are parametrized by the temperature $\beta = 1/KT$ and by the activity $z(q) = z_\alpha$ in the discrete case, $z(q) = z$ for the rotator.

The small activity expansion of the truncated correlation functions is given by the series

$$\rho^T(Q) = \sum_{n=0}^{\infty} \frac{1}{n!} I_n(Q) \tag{4}$$

$$I_n(Q) = \sum_{g \in G_Q^n} \int d\bar{Q}^{(n)} F_g(Q\bar{Q}^{(n)})\, z(Q\bar{Q}) \tag{5}$$

where G_Q^n is the set of fully connected graphs with $|Q|+n$ vertices $Q\bar{Q}^{(n)}$, Q fixed, and $\bar{Q}^{(n)}$ arbitrary; $F_g(Q\bar{Q}^{(n)})=\prod_{l\in g}(e^{-\beta\phi(l)}-1)$, l being a line of the graph g; and $z(\tilde{Q})=\prod_{q\in\tilde{Q}} z(q)$.

We recall that the series converges absolutely for $z(q)<z_0$. We denote by Q^λ the translate of Q by $d\hat{u}$, where $\hat{u}$ is some fixed unit vector in $\mathbb{R}^\nu$:

$$Q^\lambda=((x_1+\lambda\hat{u},\omega_1),\dots,(x_n+\lambda\hat{u},\omega_n))$$

Our main results are given by the following propositions:

Proposition 1. If the potentials satisfy the conditions (1a, 1b) with $\gamma>\nu$ then for any $Q_1\neq\phi$, $Q_2\neq\phi$, and $0\leqslant z(q)\leqslant z_0$

$$\lim_{\lambda\to\infty}\lambda^\gamma\rho^T(Q_1,Q_2^\lambda)=-\beta\int_\Omega d\bar{\omega}_1\int_\Omega d\bar{\omega}_2 K_{\bar{\omega}_1}(Q_1)\,d_{\bar{\omega}_1\bar{\omega}_2}(\hat{u})\,K_{\bar{\omega}_2}(Q_2)$$

where $K_{\bar{\omega}}(Q)=N_{\bar{\omega}}(Q)\,\rho^T(Q)+\int d\bar{x}\,\rho^T(Q,(\bar{x},\bar{\omega}))$

$$N_{\bar{\omega}}(Q)=\sum_{q_i\in Q}\delta_{\bar{\omega}\omega_i}\tag{6}$$

Furthermore, in the case of N-component systems

$$K_\alpha(Q)=z_\alpha\frac{d}{dz_\alpha}\rho^T(Q)\qquad\text{and}\qquad\int d\bar{\omega}=\sum_{\bar{\alpha}=1}^{N}$$

Proposition 2. For rotator systems interacting by means of two-body potential satisfying (1a, 1b), and such that

$$d_{\omega_1\omega_2}(\hat{x})=(\omega_1,A(\hat{x})\,\omega_2)$$

$$\lim_{\lambda\to\infty}\lambda^\gamma\rho^T(q_1,q_2^\lambda)=-\beta K^2\,d_{\omega_1\omega_2}(\hat{u})$$

$$K=\left[\rho+\int d\bar{y}\int d\bar{\omega}\,\rho^T_{\omega\bar{\omega}}(0,y)\,\omega\cdot\bar{\omega}\right]$$

Remark. The asymptotic behavior of $[\rho(Q_1Q_2^\lambda)-\rho(Q_1)\,\rho(Q_2^\lambda)]$ is similar to Eq. (6) with $K_\omega(Q)$ replaced by

$$K'_\omega(Q)=N_\omega(Q)\,\rho(Q)+\int d\bar{x}[\rho(Q,(\bar{x},\bar{\omega}))-\rho(Q)\,\rho(\bar{x},\omega)]$$

3. PROOFS OF PROPOSITIONS 1 AND 2

To establish the above propositions we follow the proof of Ref. 1: we first compute explicitly the limit $\lambda\to\infty$ of $\lambda^\gamma I_n(Q_1,Q_2^\lambda)$ (Lemma 1) and then we give bounds which justify the permutation of the limit $\lambda \to \infty$ with the summation over n (Lemma 2).

Lemma 1:

$$\lim_{\lambda \to \infty} \lambda^\gamma I_n(Q_1, Q_2^\lambda)$$

$$= -\beta \int d\bar{\omega}_1 \int d\bar{\omega}_2 \sum_{\substack{k=0 \\ k+l=n}}^{n} \frac{n!}{k!\, l!} I_k^{(\bar{\omega}_1)}(Q_1)\, d_{\bar{\omega}_1\bar{\omega}_2}(\hat{u})\, I_l^{(\bar{\omega}_2)}(Q_2)$$

where

$$I_n^{(\bar{\omega})}(Q) = \sum_{g \in G_Q^{(n)}} \int d\bar{Q}^{(n)} F_g(Q, \bar{Q})\, N_{\bar{\omega}}(Q\bar{Q})\, z(Q\bar{Q})$$

$$= N_{\bar{\omega}}(Q)\, I_n(Q) + n \int d\bar{x}\, I_{n-1}(Q, (\bar{x}, \bar{\omega})) \tag{7}$$

Lemma 2. There exists $c_n > 0$ independent of λ such that

$$(n!)^{-1} |\lambda^\gamma I_n(Q_1, Q_2^\lambda)| \leqslant c_n \qquad \text{and} \qquad \sum_{n \geqslant 0} c_n < \infty$$

It then follows from Lemmas 1 and 2, together with Eqs. (5)–(7) that

$$\lim_{\lambda \to \infty} \lambda^\gamma \rho^T(Q_1, Q_2^\lambda) = -\beta \int d\bar{\omega}_1 \int d\bar{\omega}_2\, K_{\bar{\omega}_1}(Q_1)\, d_{\bar{\omega}_1\bar{\omega}_2}(\hat{u})\, K_{\bar{\omega}_2}(Q_2)$$

with

$$K_{\bar{\omega}}(Q) = \sum_{n=0}^{\infty} (n!)^{-1} I_n^{(\bar{\omega})}(Q) = N_{\bar{\omega}}\rho^T(Q) + \int d\bar{x}\, \rho^T(Q, (\bar{x}, \bar{\omega}))$$

$$= z_{\bar{\omega}} \frac{d}{dz_{\bar{\omega}}} \rho^T(Q)$$

Proof of Lemma 1. Following the proof of Ref. 1 we divide the domain of integration into

$$\mathbb{R}^\nu = \mathscr{D}_1^{(\lambda)} \cup \mathscr{D}_2^{(\lambda)} \cup \mathscr{D}_3^{(\lambda)}$$

where $\mathscr{D}_1^{(\lambda)}$ is around the origin, $\mathscr{D}_1^{(\lambda)} = \{x \mid |x| < \lambda/4\}$, and $\mathscr{D}_2^{(\lambda)}$ around $\lambda\hat{u}$, $\mathscr{D}_2^{(\lambda)} = \{|x| \mid |x - \lambda\hat{u}| < \lambda/4\}$.

It is rather intuitive that the only contribution to the $\lim_{\lambda \to \infty} \lambda^\gamma I_n(Q_1 Q_2^\lambda)$ will come from graphs with vertices in $\mathscr{D}_1^\lambda$ *or* $\mathscr{D}_2^\lambda$ and with only *one line* l connecting $\mathscr{D}_1^\lambda$ to $\mathscr{D}_2^\lambda$. For a proof of this fact we refer to Ref. 1. In mathematical terms this means

$$\lim_{\lambda \to \infty} \lambda^\gamma I_n = -\beta \sum_{I \subset \{1,\dots,n\}} \sum_{g_1 \in G_{Q_1,|I|}} \sum_{g_2 \in G_{Q_2,|J|}} \sum_{l \in \mathscr{L}_{I,J}}$$

$$\int d\bar{Q}_I \int d\bar{Q}_J \, F_{g_1}(Q_1\bar{Q}_I)\, F_{g_2}(Q_2\bar{Q}_J)\, z(Q_1\bar{Q}_I)\, z(Q_2\bar{Q}_J)\, d_l(\hat{u})$$

The set of vertices $\{1,\dots, n\}$ is decomposed into two parts: $\{1,\dots, n\} = I \cup J$. The vertices in I are in $\mathscr{D}_1^\lambda$, the vertices in J are in $\mathscr{D}_2^\lambda$. $\mathscr{L}_{I,J}$ denotes the set of lines connecting $Q, \bar{Q}_I$ tot $Q_2^\lambda \bar{Q}_J$. But

$$\sum_{l \in \mathscr{L}_{I,J}} d_l(\hat{u}) = \int d\bar{\omega}_1 \int d\bar{\omega}_2 N_{\bar{\omega}_1}(Q_1\bar{Q}_I)\, N_{\bar{\omega}_2}(Q_2 Q_J)\, d_{\bar{\omega}_1\bar{\omega}_2}(\hat{u})$$

Thus

$$\lim \lambda^\gamma I_n = -\beta \int d\omega_1 \int d\omega_2 \, d_{\omega_1\omega_2}(\hat{u}) \sum_{\substack{k=0 \\ k+l=n}}^{n} \frac{n!}{k!\, l!}$$

$$\left[\sum_{g_1 \in G_{Q_1,k}} \int d\bar{Q}^k \, F_{g_1}(Q_1\bar{Q}^k)\, N_{\bar{\omega}_1}(Q_1\bar{Q}^k)\, z(Q_1\bar{Q}^k)\right]$$

$$\left[\sum_{g_2 \in G_{Q_2,l}} \int d\bar{Q}^l \, F_{g_2}(Q_2\bar{Q})\, N_{\bar{\omega}_2}(Q_2\bar{Q}^l)\, z(Q_2\bar{Q}^l)\right]$$

which concludes the proof of Lemma 1.

Using the estimates Eqs. (2) and (3), the proof of Lemma 2 is identical to the proof given in Ref. 1. ∎

Proof of Proposition 2. Proposition 1 implies

$$\lim_{\lambda \to \infty} \lambda^\gamma \rho^T(q_1, q_2^\lambda) = -\beta \int_\Omega d\bar{\omega}_1 \int_\Omega d\bar{\omega}_2$$

$$\left[\rho(q_1) + \int d\bar{x}_1\, \rho^T(q_1, (\bar{x}_1, \bar{\omega}_1))\right] d_{\bar{\omega}_1\bar{\omega}_2}(\hat{u})$$

$$\left[\rho(q_2) + \int d\bar{x}_2\, \rho^T(q_2, (\bar{x}_2, \bar{\omega}_2))\right]$$

From the convergence of the Mayer expansion it is easy to prove the following Euclidean invariance of the two-point function

$$\rho(q_1, q_2) = \rho_{\omega_1\omega_2}(x_1 - x_2) = \rho_{\mathscr{R}\omega_1, \mathscr{R}\omega_2}(\mathscr{R}(x_1 - x_2))$$

where $\mathscr{R} \in SO(d)$. In particular if $\mathscr{R}(\alpha\hat{\omega}_1)$ denotes a rotation of axis $\hat{\omega}_1$ and angle α, we have

$$\rho((x_1, \omega_1), (x_2, \omega_2)) = \rho_{\omega_1, \mathscr{R}(\alpha\hat{\omega}_1)\omega_2}(\mathscr{R}(\alpha\hat{\omega}_1)(x_1 - x_2)) \tag{8}$$

Now $d_{\bar{\omega}_1\bar{\omega}_2}(\hat{u}) = \bar{\omega}_1 A(\hat{u})\, \bar{\omega}_2$, where $A(\hat{u})$ is a tensor field on the unit sphere.

Let us compute

$$\int d\bar{q}_1 \rho(q_1 \bar{q}_1)\, \bar{\omega}_1 A \bar{\omega}_2$$

We use the decomposition

$$\bar{\omega}_1 = (\bar{\omega}_1 \cdot \omega_1)\, \omega_1 + \bar{\omega}_1^{\perp}$$

The symmetry property (8) together with the rotation invariance of dq implies

$$\int d\bar{q}_1\, \rho(q_1 \bar{q}_1)\, \bar{\omega}_1^{\perp} A \bar{\omega}_2$$

$$= \int d\bar{q}_1\, \rho(q_1 \bar{q}_1) (\mathscr{R}(\alpha\hat{\omega}_1)\, \omega_1^{\perp})\, A \bar{\omega}_2 \tag{9}$$

Choosing α such that $\mathscr{R}(\alpha\hat{\omega}_1)\, \bar{\omega}_1^{\perp} = -\bar{\omega}_1^{\perp}$

$$(9) = -\int d\bar{q}_1\, \rho(q_1 \bar{q}_1)\, \bar{\omega}_1^{\perp} A \bar{\omega}_2$$

Therefore

$$\int d\bar{q}_1\, \rho(q_1 \bar{q}_1)\, \bar{\omega}_1 A \bar{\omega}_2$$

$$= \int d\bar{q}_1 (\omega_1 \cdot \bar{\omega}_1)\, \rho(q_1 \bar{q}_1)\, \omega_1 A \bar{\omega}_2 \tag{10}$$

Similarly,

$$\int d\bar{q}_2\, \rho(q_2 \bar{q}_2)\, \omega_1 A \bar{\omega}_2$$

$$= \int d\bar{q}_2 \bar{\omega}_2 \cdot \omega_2 \rho(q_2 \bar{q}_2)\, \omega_1 A \omega_2 \tag{11}$$

(10) and (11) yields the result. ∎

4. CONCLUDING REMARKS AND CONJECTURE

In the domain of convergence of the activity expansion the state is invariant under translation:

$$\rho(q)=\rho_{\alpha}, \qquad \rho(q,q')=\rho_{\alpha\alpha'}(x-x')$$

$$K_{\alpha}(q')=K_{\alpha\alpha'}=z_{\alpha}\frac{d}{dz_{\alpha}}\rho_{\alpha'}=\rho_{\alpha}\delta_{\alpha\alpha'}+\int dy\,\rho^{T}_{\alpha\alpha'}(y)$$

Since $\rho_{\alpha}=\partial p/\partial\mu_{\alpha}$, with p the pressure, we obtain the interpretation of the coefficients $K_{\alpha\alpha'}$, in terms of the *compressibility tensor* $\chi_{\alpha\alpha'}$; indeed

$$\chi_{\alpha\alpha'}=\frac{\beta}{\rho^{2}}z_{\alpha}\frac{\partial}{\partial z_{\alpha}}\rho_{\alpha'}=\frac{1}{\rho^{2}}\frac{\partial\rho_{\alpha'}}{\partial\mu_{\alpha}}=\frac{1}{\rho^{2}}\frac{\partial^{2}p}{\partial\mu_{\alpha}\partial\mu_{\alpha'}}$$

implies

$$K_{\alpha\alpha'}=\rho^{2}\beta^{-1}\chi_{\alpha\alpha'} \qquad \text{and} \qquad \frac{\partial p}{\partial\rho_{\alpha}}=\beta^{-1}\sum_{\alpha'}K^{-1}_{\alpha\alpha'}\rho_{\alpha'}$$

We have thus obtained as special case of Proposition 1 (with $|Q_{1}|=|Q_{2}|=1$)

$$|x|^{\gamma}\,\rho^{T}_{\alpha_{1}\alpha_{2}}(x)\underset{|x|\to\infty}{\sim}\sum_{\bar{\alpha}_{1},\bar{\alpha}_{2}}K_{\bar{\alpha}_{1}\alpha_{1}}\,d_{\bar{\alpha}_{1}\bar{\alpha}_{2}}(\hat{x})\,K_{\bar{\alpha}_{2}\alpha_{2}}$$

From the stability condition it is expected, and sometime can be proved, that the compressibility tensor is definite positive. Thus Proposition 1 implies that $\lambda^{\gamma}\rho^{T}_{\alpha_{1}\alpha_{2}}(\lambda\hat{x})$ tends to a nonzero limit as $\lambda\to\infty$ for all (α_{1},α_{2}), i.e., all correlation functions decay exactly like the slowest decaying potential whenever $\gamma>\nu$.

On the other hand for very-long-range potential $\gamma\leqslant\nu-1$ ($\gamma=\nu-2$, corresponding to Coulomb potential) it is possible to show that any $\mathscr{L}_{1}$-clustering equilibrium state must obey the following sum rule (4, 5):

$$\sum_{\bar{\alpha}}d_{\alpha\bar{\alpha}}(\hat{u})\,K_{\bar{\alpha}}(Q)=0$$

In particular for $|Q|=1$ it yields

$$d_{\alpha\alpha_{1}}\rho_{\alpha_{1}}+\int d\bar{x}\sum_{\bar{\alpha}}d_{\alpha\bar{\alpha}}(\hat{u})\,\rho^{T}_{\bar{\alpha}\alpha_{1}}(\bar{x})=0$$

These sum rules show that the result expressed by Proposition 1 remain valid for $\gamma\leqslant\nu-1$; the sum rules which express the screening condition for very-long-range potential are exactly the necessary conditions for the correlation to decay faster than the potential.

It is then tempting to conjecture that Proposition 1 will hold for any potentials with power law decay, except that the coefficient $K_{\alpha\bar{\alpha}}$ might be slightly different for $\nu - 1 < \gamma \leqslant \nu$, as it is expected for dipoles systems ($\gamma = \nu$).

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