Decay of Correlations in Classical Fluids with Long-Range Forces

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We show with simple arguments that, as a consequence of the Poisson equation, the correlations of a charged system at equilibrium decay faster than any inverse power, if they are integrable and monotonous at infinity. For all other long-range systems (with potential $\phi(x) \sim b |x|^{-s}$, $|x| \rightarrow \infty$, 0 < s < v, $s \neq v - 2$), the decay is bounded below by an inverse power.

KEY WORDS: Long-range forces; Coulomb systems; BGY hierarchy; correlations.

1. INTRODUCTION

The decay of the correlations in classical fluids has been the subject of a considerable number of investigations from the physical and mathematical viewpoint. The general situation for the high-temperature (low-density) homogeneous phase can be described as follows. If the potential $\phi(x)$ has finite range or is exponential, the correlations cluster exponentially fast. If $\phi(x) \sim b |x|^{-s}$, $|x| \to \infty$, s > 0, is algebraic, the decay of the correlations is also always algebraic, with the only exception of the Coulomb systems (s = v - 2, v = space dimension) where the decay is faster than any inverse power.

More precisely, when $\phi(x) \sim b |x|^{-s}$ with s > v (integrable case) the correlations cluster exactly as $|x|^{-s}$ (see Ref. 1 and the references quoted there). When 0 < s < v (nonintegrable case) and $s \neq v - 2$, the heuristic

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argument that the direct correlation function c(x) behaves as $-\beta\phi(x)$, $|x| \to \infty$ (Ref. 2) leads to a $|x|^{-(2\nu-s)}$ decay. However, in the Coulomb case $(s=\nu-2)$ the Debye-Hückel theory gives an exponential decay, a fact which has been rigorously established in Ref. 3 at sufficiently high temperature or low density. It is also known that in the two-dimensional one-component jellium and for a special value of the temperature, the correlations have a Gaussian decay.⁽⁴⁾ Thus in the class of systems with algebraic potentials, the Coulomb case appears to play an exceptional role since only for it a decay faster than any inverse power can be obtained.

We show in this paper that the basic reason for this situation is the harmonicity of the Coulomb potential. We work in the following setting: we assume that the correlations of an infinite homogeneous phase are given and obey the BGY hierarchy at temperature β^{-1} (the validity of the thermodynamic limit and of the BGY equations are not established here; see Ref. 5). Then we analyze the asymptotic behavior of the terms of the BGY equation in relation with the exponent s of the potential: constraints are imposed on the decay of the correlations, provided that they are integrable and that some natural bounds are verified. When s = v - 2, the relation between the charge density and the field generated by it is local, i.e. given by a differential equation, the Poisson equation. Our main result is that, as a consequence of this fact, the correlations of a charged fluid cannot have a monotonuous algebraic decay. In all other cases, only a power law decay is compatible with the conditions of thermal equilibrium. In the analysis, one must distinguish between the one-component plasma (OCP) and multicomponent systems. The latter case involves two different types of correlations, the charge-charge and the charge-particle correlations, which are linked in the BGY equations, and one must discuss it separately.

It is known that starting from the monotonuous Debye–Hückel regime and lowering the temperature, oscillations will occur.⁽⁶⁾ In fact, the expected analytic properties of the structure factor lead to an exponential oscillating decay.⁽⁷⁾ Such a behavior is not excluded by our arguments, nor do we exclude an inverse power law decay with oscillations (typically like $|x|^{-p} \cos \lambda |x|$). In the non-Coulomb cases, we obtain essentially $|x|^{-(2\nu-s)}$ as lower bound on the decay, in agreement with the usual findings involving assumptions on the direct correlation function. A situation of special interest is $\nu = 2$, s = 1 (the electrons at the surface of the liquid helium) where we have $|x|^{-3}$ as an exact lower bound.

The simplest forms of our arguments are found in Propositions 1 and 4. Some proofs have been relegated to appendixes in order to keep the presentation as simple as possible.

2. GENERAL SETTING

We consider infinitely extended homogeneous equilibrium states of charged particles in \mathbb{R}^{ν} ($\nu =$ space dimension, $\nu \ge 2$).

For an N-component system, we denote by $q = (x, \alpha)$ the position x and thereafter species α of a particle of charge e_{α} , $\alpha = 1, 2, ..., N$. We include also jellium systems with an additional uniform external charge density ρ_{B} .

The particles interact by means of a two-body potential $\phi(q_1q_2) = e_{\alpha_1}e_{\alpha_2}\phi(x_1-x_2)$ with the following properties:

(a)
$$\phi(x) = \frac{b}{|x|^s} + \phi^0(|x|), \quad 0 < s < v, \quad b \neq 0$$

(We include also the two-dimensional Coulomb case, v = 2, s = 0, $\phi(x) = -\ln(|x|/L) + \phi^0(x)$.]

(b) $\phi^0(x)$ is a spherically symmetric potential with compact support, which is differentiable except possibly at x = 0 and such that $(\nabla \phi)(x)$ is integrable at x = 0.

In multicomponent systems, $\phi^0(x)$ includes the local repulsion effects needed for thermodynamic stability. In the Coulomb jellium, we may take $\phi^0(x) = 0$.

Notice that the Fourier transform $\tilde{\phi}(k) = \int dx \,\phi(x) \exp(ikx)$ of $\phi(x)$ behaves as⁽⁸⁾

$$b(k) = C_{\nu} b |k|^{s-\nu} + \phi^{0}(k) \sim C_{\nu} b |k|^{s-\nu}, \qquad |k| \to 0$$

$$C_{\nu} = 2^{\nu-s} \pi^{\nu/2} \frac{\Gamma((\nu-s)/2)}{\Gamma(s/2)}$$
(2.1)

The correlation functions $\rho(q_1 \cdots q_n)$ of the state at temperature β^{-1} $(\rho(q_1), \rho(q_1q_2),...$ being, respectively, the singlet, doublet,... densities) are assumed to satisfy the BGY hierarchy

$$\beta^{-1} \nabla_1 \rho(q_1 \cdots q_n) = \sum_{j=2}^n F(q_1 q_j) \rho(q_1 \cdots q_n) + \int dq F(q_1 q) [\rho(qq_1 \cdots q_n) - \rho(q) \rho(q_1 \cdots q_n)] \quad (2.2)$$

where $F(q_1q_2) = e_{\alpha_1}e_{\alpha_2}F(x_1 - x_2) = -e_{\alpha_1}e_{\alpha_2}(\nabla\phi)(x_1 - x_2)$ is the force.

For all s, 0 < s < v, Eq. (2.2) with the truncation in the integrand of the right-hand side is the appropriate form of the equilibrium equation. When $0 < s \le v - 2$, the system is necessarily overall neutral (Proposition 6 of Ref. 9)

$$\sum_{\alpha=1}^{N} e_{\alpha} \rho_{\alpha} + \rho_{B} = 0 \qquad [\rho_{\alpha} = \rho(q)]$$
(2.3)

and $-\int dq F(q_1q) \rho(q) = e_{\alpha_1} \int dy F(x_1 - y) \rho_B$ is formally the contribution of the background. When v - 2 < s < v there are no constraints on the ρ_{α} , but either $(\nabla F)(x)$ $(v - 2 < s \le v - 1)$ or F(x) (v - 1 < s < v) are integrable at infinity and $\int dq F(q_1q) \rho(q)$ vanishes in a homogeneous state because of the antisymmetry of the force (see the discussion of the BGY equations for long-range forces in Ref. 9).

Throughout the paper we shall only consider fluid phases with decay of the correlations insuring that the integral in the right-hand side of (2.2) is absolutely convergent [see (2.8) below].

It will be convenient to write the hierarchy (2.2) in an alternative equivalent form. We introduce as in Ref. 10 the excess particle density at q when particles are fixed at $q_1 \cdots q_n$:

$$\rho(q \mid q_1 \cdots q_n) = \frac{\rho(qq_1 \cdots q_n)}{\rho(q_1 \cdots q_n)} - \rho(q) + \sum_{j=1}^n \delta_{qq_j}$$
(2.4)
$$\delta_{qq_j} = \delta(x - x_j) \,\delta_{\alpha\alpha_j}$$

The corresponding charge density generates the field $E(x; q_1 \cdots q_n)$ at x

$$E(x; q_1 \cdots q_n) = \int dy F(x - y) \sum_{\alpha = 1}^{N} e_{\alpha} \rho(y \alpha | q_1 \cdots q_n)$$
(2.5)

In terms of these quantities the hierarchy (2.2) becomes (using the fact that the force is antisymmetric)

$$\beta^{-1} \nabla_{x} [\rho(qQ) - \rho(q) \rho(Q)] = e_{\alpha} E(x; Q) \rho(q) \rho(Q) + \sum_{j=1}^{n} F(qq_{j}) [\rho(qQ) - \rho(q) \rho(Q)] + \int dq' F(qq') R(qq'Q)$$
(2.6)

where we have set $Q = (q_1 \cdots q_n)$ and

$$R(qq'Q) = \rho(qq'Q) - \rho(q') \rho(qQ) - \rho(q)[\rho(q'Q) - \rho(q') \rho(Q)] - \rho(Q)[\rho(qq') - \rho(q) \rho(q')]$$
(2.7)

When Q consists of a single point q_1 , $R(qq'q_1)$ is identical with the fully truncated three-point Ursell function $\rho_T(qq'q_1)$ defined in the usual way.

In the following, we shall make the assumption that the truncated correlations have an integrable decay at infinity, i.e.,

$$|\rho_T(q_1\cdots q_n)| \leqslant \frac{M}{r^p} \tag{2.8}$$

 $r = \sup_{ij} |x_i - x_j|$ for some p > v. Other more specific clustering assumptions will be specified later.

3. COULOMB SYSTEMS

In this section, we show under mild clustering assumptions that the charge correlation functions of a Coulomb fluid cannot decay as a monotonous inverse power law. For simplicity we consider first the charge-charge correlations of a homogeneous one-component system in three dimensions with a pure Coulomb force $F(x) = \hat{x}/|x|^2$ (OCP). The same results hold in two dimensions or with an additional finite-range potential. The case of the higher-order correlations in v dimensions will be treated in Section 3.2. General multicomponent systems are discussed at the end of this section.

3.1. The Two-Point Correlation Function of the OCP

Denoting $h(r) = \rho(x0)/\rho^2 - 1$, (r = |x|), the spherically symmetric twopoint Ursell function of the OCP, the second equation of the hierarchy (2.6) reduces in this case to

$$\beta^{-1} \frac{d}{dr} h(r) = eE(r) + \frac{e^2}{r^2} h(r) + \frac{e^2}{\rho^2} \hat{x} \cdot \int dy F(x-y) \rho_T(xy0)$$
(3.1)

where E(r) is the radial component of the field determined by the Poisson equation

$$\frac{1}{4\pi r^2} \frac{d}{dr} \left[r^2 E(r) \right] = e \left[\rho h(r) + \delta(x) \right]$$
(3.2)

The main point of our argument is that, because of the local differential relation (3.2), the decay of the field has to be slower than that of the charge density when the latter is algebraic, a situation which is shown to be incompatible with the equilibrium equation. This is formulated in a simple way in Proposition 1.

Proposition 1. Assume that (1.i) $\lim_{r\to\infty} r^p h(r) = A < \infty$, for some p > 3;

(1.ii) for |x| large enough $|\rho_T(xy0)| \leq M(t)/|x|^p$,

$$t = \min(|x - y|, |y|)$$
 with $\lim_{t \to \infty} M(t) = 0$

then A = 0.

Remark. The condition (1.ii) is slightly stronger than (2.8) in the sense that some joint decay is required in the Ursell function as a second particle is sent to infinity, the third one being fixed at the origin. We show

in Appendix A that (1.ii) is compatible with known exact results and is a weak form of bounds indicated by perturbative expansions.

Proof. Integrating the Poisson equation (3.2) gives for $r \neq 0$ (Gauss theorem)

$$E(r) = \frac{c}{r^2} - \frac{4\pi e\rho}{r^2} \int_r^\infty dr' r'^2 h(r') dr'$$
(3.3)

with $c = e \int dx [\rho h(r) + \delta(x)]$ the total excess charge, and hence, from (1.i)

$$E(r) = \frac{c}{r^2} - \frac{4\pi e\rho}{(p-3)} \frac{A}{r^{p-1}} + o\left(\frac{1}{r^{p-1}}\right)$$
(3.4)

Moreover, the condition (1.ii) implies that (see Lemma 1 in Appendix B)

$$\int dy F(x-y) \rho_T(xy0) = o\left(\frac{1}{r^{p-1}}\right)$$
(3.5)

Inserting (3.4) and (3.5) in (3.1) gives

$$\beta^{-1} \frac{dh(r)}{dr} = e \frac{c}{r^2} - \frac{4\pi e^2 \rho}{(p-3)} \frac{A}{r^{p-1}} + o\left(\frac{1}{r^{p-1}}\right)$$

and therefore

$$\beta^{-1}h(r) = -e\frac{c}{r} + \frac{4\pi e^2 \rho}{(p-3)(p-2)}\frac{A}{r^{p-2}} + o\left(\frac{1}{r^{p-2}}\right)$$

Thus we conclude from (1.i) that c = 0 and A = 0 (c = 0 is the familiar perfect screening rule).

In fact, the *a priori* assumption (1.i) of algebraic decay can be weakened. The stronger Proposition 2 shows that the decay has to be faster than any inverse power whenever h(r) is monotonic at infinity and the three-point function obeys a reasonable bound in terms of h(r). It includes the Proposition 1 as a particular case, but it also excludes nonalgebraic decays, for instance $(\ln r)^q/r^\rho$. The assumed bound on the three-point function is analogous to (1.ii) (see also Appendix A) and the proof of Proposition 2 is given in Appendix C.

Proposition 2. Assume that (2.i) h(r) tends monotonously to zero as $r \to \infty$;

(2.ii) For |x| large enough, $|\rho_T(xy0)| \le |h(x)| M(t)$,

$$t = \min(|x - y|, |y|), \qquad \lim_{t \to \infty} M(t) = 0$$

then $\lim_{r\to\infty} r^p h(r) = 0$ for all p > 0.

In Propositions 1 and 2, the hypothesis of monotonicity played an essential role, and we cannot exclude an oscillatory decay of the type $(\cos \lambda r)/r^p$, because then the field and the charge density are of the same order at infinity. However, certain types of oscillations can also be excluded under slightly stronger conditions. For instance, we show in the next proposition (proof in the Appendix D) that if the dominant asymptotic term of h(r) has oscillations as $r \to \infty$, the latter cannot be "too slow" or "too fast."

Proposition 3. Assume that (3.i) $h(r) = A(\cos \lambda r^{\alpha})/r^{p} + f(r)$ with $\alpha > 0, \ \alpha \neq 1, \ p > 3$, and $f(r) = O(1/r^{p+1}), \ df(r)/dr = O(1/r^{p+1});$ (3.ii) $|x|^{p} \int dy |\rho_{T}(xy0)| \leq M$, then A = 0.

3.2. Higher-Order Correlations

We turn now to the higher-order correlations of the OCP keeping the notation

$$h(x) = \frac{\rho(xQ)}{\rho\rho(Q)} - 1, \qquad Q = (x_1 \cdots x_n)$$

 $\rho(h(x)+1)$ can also be considered as the density in an inhomogeneous state with external charges at $x_1 \cdots x_n$. We allow a short-range potential as in part (b), Section 2, and a general dimension $v \ge 2$.

Proposition 4. Assume that (4.i) $|x|^p |h(x)| \le M$ for some p > 3, and $\lim_{\lambda \to \infty} \lambda^p h(\lambda \hat{x}) = A(\hat{x})$ a.e. $\hat{x} = x/|x|$;

(4.ii) For fixed $x_1 \cdots x_n$ and |x| large enough

$$|x|^{p} |R(xyx_{1}\cdots x_{n})| \leq M(t)$$

$$t = \min(|x-y|, |y|), \qquad \lim_{t \to \infty} M(t) = 0$$

then $A(\hat{x}) = 0$ a.e. \hat{x} .

Remark. In (4.i) h(x) (which is now anisotropic) is assumed to decay radially as the same inverse power law for almost all directions. The condition (4.ii) on the truncated functions (2.7) is the analog of the bound (1.ii) in Proposition 1, and is also discussed in the Appendix A.

The idea of the proof is exactly the same as that of Proposition 1. The technical difference is that here, because of anisotropy, the Poisson equation cannot be integrated in a straightforward manner. It turns out to

be convenient here to study the asymptotics of the BGY hierarchy in the weak sense, i.e., integrating Eq. (2.6) on test functions.

Proof. Let g(x) belong to C_0^{∞} (the set of infinitely differentiable functions with compact support), and g(x) = 0 for $|x| \le a$, $a > \sup_j |x_j|$.

We replace x by λx in Eq. (2.6), multiply it by g(x), and integrate over x.

By the assumption (4.i), the left-hand side of (2.6) is

$$\beta^{-1} \int dx g(x) (\nabla_{\lambda x} h)(\lambda x) = -\frac{\beta^{-1}}{\lambda} \int dx (\nabla g)(x) h(\lambda x) = O\left(\frac{1}{\lambda^{p+1}}\right) (3.6)$$

The second term of the right-hand side is obviously $O(1/\lambda^{p+\nu-1})$, and it is shown in Appendix B that under the condition (4.ii) the last term is

$$\frac{1}{\rho\rho(Q)}\int dxg(x)\int dyF(\lambda x-y)\ R(\lambda xyQ)=o\left(\frac{1}{\lambda^{p-1}}\right)$$
(3.7)

This implies

$$\int dxg(x) E(\lambda x; Q) = o\left(\frac{1}{\lambda^{p-1}}\right)$$

According to parts (a) and (b) of Section 2, we split $E(x; Q) = E^{c}(x) + E^{0}(x)$ into its pure Coulomb part and short-range part, with

$$\nabla \cdot E^{c}(x) = \omega_{v}e\rho(x \mid Q) = \omega_{v}e\left[\rho h(x) + \sum_{j=1}^{n} \delta(x - x_{j})\right]$$
(3.8)

 $(\omega_v = \text{surface of the unit sphere in dimension } v)$ and

$$E^{0}(x) = -e \int dy (\nabla \phi^{0})(x - y) \rho(y | Q)$$

Since $\rho(y|Q)$ is $O(1/|y|^p)$ and $\phi^0(x)$ has finite range, one has also $E^0(x) = O(1/|x|^p)$. This, together with g(x) = 0 for $|x| \le a$, implies

$$\int dxg(x) E^{c}(\lambda x) = o\left(\frac{1}{\lambda^{p-1}}\right)$$
(3.9)

Since (3.9) holds for any $g(x) \in C_0^{\infty}$ with g(x) = 0, $|x| \leq a$, one has also

$$\int dx (\nabla g)(x) \cdot E^{c}(\lambda x) = o\left(\frac{1}{\lambda^{p-1}}\right)$$

for any such g.

But by the Poisson equation (3.8), the fact that g(x) = 0 for $|x| \leq \sup_i |x_i|$ and scaling, we obtain for $\lambda > 1$

$$\int dx (\nabla g)(x) \cdot E(\lambda x) = -\int dx g(x) \nabla_x \cdot E(\lambda x)$$
$$= -\lambda \omega_y e\rho \int dx g(x) h(\lambda x)$$

Hence $\int dxg(x) \lambda^p h(\lambda x) = o(1)$. Taking the limit $\lambda \to \infty$ we get by dominated convergence

$$\int dxg(x) \frac{A(\hat{x})}{|x|^p} = 0$$

Choosing g of the form $g(x) = \varphi(|x|) \psi(\hat{x})$, with $\varphi \in C_0^{\infty}(\mathbb{R})$, $\varphi(|x|) = 0$, $|x| \leq a$ and $\psi(\hat{x})$ any C^{∞} function on the unit sphere in \mathbb{R}^{ν} , we obtain $\int d\hat{x}\psi(\hat{x}) A(\hat{x}) = 0$, and hence $A(\hat{x}) = 0$ a.e. \hat{x} .

We conclude from Proposition 4 that $A(\hat{x})$ vanishes in any open set where it is continuous. Since the correlations of a state extended in the whole space \mathbb{R}^{ν} are continuous, it is natural to have also $A(\hat{x})$ continuous everywhere, and thus h(x) cannot decay algebraically in any direction. The situation will, however, be different for systems confined by hard walls, such as the semi-infinite Coulomb gas. In this case, it is known that even in the high-temperature phase, the decay of correlations parallel to the wall is algebraic ($\simeq |x|^{-3}$ in dimension 3) and faster than any inverse power in all other directions.⁽¹¹⁾ This fact is compatible with the present analysis. The correlations of this semi-infinite system are continuous except at the wall: they satisfy the equilibrium equations in the fluid, but vanish outside of the wall. One can check that Proposition 4 still applies, but now $A(\hat{x})$ can be assumed to be continuous everywhere except at the direction \hat{x} parallel to the wall. We then conclude that the decay is faster than any inverse power in all directions not parallel to the wall. Parallel to the wall, where the decay is indeed algebraic, we cannot draw any conclusion by the present method.

3.3. Multicomponent Systems

To generalize the analysis to multicomponent systems, we consider $\sum_{\alpha_1} e_{\alpha_1} \rho_{\alpha_1} h_{\alpha_1 \alpha_2}(r)$, the charge density at |x| = r when a particle of type α_2 is fixed at the origin, $h_{\alpha_1 \alpha_2}(r) = (1/\rho_{\alpha_1} \rho_{\alpha_2}) \rho(x_1 \alpha_1, x_2 \alpha_2) - 1$ and write the Eq. (2.6) for this quantity,

$$\beta^{-1} \frac{d}{dr} \left[\sum_{\alpha_1} e_{\alpha_1} \rho_{\alpha_1} h_{\alpha_1 \alpha_2}(r) \right] = \left(\sum_{\alpha} e_{\alpha}^2 \rho_{\alpha} \right) E(r) + \frac{e_{\alpha_2}}{r^2} \left[\sum_{\alpha_1} e_{\alpha_1}^2 \rho_{\alpha_1} h_{\alpha_1 \alpha_2}(r) \right] + \frac{\hat{x}}{\rho_{\alpha_2}} \cdot \sum_{\alpha_1 \alpha} e_{\alpha_1}^2 e_{\alpha} \int dy F(x_1 - y) \rho_T(x_1 \alpha_1, 0\alpha_2, y\alpha) \quad (3.10)$$

with

$$\frac{1}{4\pi r^2} \frac{d}{dr} \left[r^2 E(r) \right] = \sum_{\alpha_1} e_{\alpha_1} \rho_{\alpha_1} h_{\alpha_1 \alpha_2}(r) + e_{\alpha_2} \,\delta(x) \tag{3.11}$$

We see that in addition to the charge-particle correlation $\sum_{\alpha_1} e_{\alpha_1} \rho_{\alpha_1} h_{\alpha_1 \alpha_2}(r)$, Eq. (3.10) involves the individual particle-particle correlation $h_{\alpha_1 \alpha_2}(r)$, and these different types of correlations could have different decays. However, in a regime of monotonuous decay, $h_{\alpha_1 \alpha_2}(r)$ has a definite sign for r large enough. For obvious reasons of electrostatic attraction and repulsion, we must have $h_{\alpha_1 \alpha_2}(r)$ positive (respectively negative) when e_{α_1} and e_{α_2} have the opposite (respectively the same) sign, i.e.,

$$e_{\alpha_1}e_{\alpha_2}h_{\alpha_1\alpha_2}(r) < 0$$
 for r large enough (3.12)

With (3.12), the decay of the particle-particle correlations cannot be slower than that of the charge-particle correlations since

$$|\rho_{\alpha_1}e_{\alpha_2}e_{\alpha_2}h_{\alpha_1\alpha_2}(r)| \leqslant \sum_{\alpha_1} |\rho_{\alpha_1}e_{\alpha_1}e_{\alpha_2}h_{\alpha_1\alpha_2}(r)| = |e_{\alpha_2}| \left| \sum_{\alpha_1} e_{\alpha_1}\rho_{\alpha_1}h_{\alpha_1\alpha_2}(r) \right|$$

Then we have the exact analog of Propositions 1 and 2, for instance Proposition 5.

Proposition 5. Assume that (3.12) holds and (5.i) $\lim_{r \to \infty} r^p \left[\sum_{\alpha_1} e_{\alpha_1} \rho_{\alpha_1} h_{\alpha_1 \alpha_2}(r) \right] = A, \ p > 3;$

(5.ii) For $|x_1|$ large enough $|\rho_T(x_1\alpha_1, 0\alpha_2, y\alpha)| \leq M(t)/|x_1|^p$, $t = \min(|x_1 - y|, |y|), \lim_{t \to \infty} M(t) = 0$, then A = 0.

4. NON-COULOMB SYSTEMS

In this section we treat the case of long-range potentials of the type (a), (b) of Section 2 characterized by 0 < s < v, $s \neq v - 2$, and show that in all cases the decay of correlations cannot be faster than any inverse power. We consider a one-component system, with neutralizing background $\rho_B \neq 0$ when 0 < s < v - 2. The results are summarized in the following.

Proposition 6. The truncated correlations of an equilibrium state characterized by Eq. (2.2) and potential (a), (b) $(s \neq v-2)$ cannot decay faster then $|x|^{-(2v-s)}$ when 0 < s < v-1 and $|x|^{-(s+2)}$ when $v-1 \leq s < v$.

This will follow from the next lemma where it is shown that the structure factor $\tilde{S}(k)$

$$\widetilde{S}(k) = e^2 \rho \int dx e^{ikx} \rho(x|0) = e^2 \rho \int dx e^{ikx} [\rho h(x) + \delta(x)]$$
$$= e^2 \rho(\rho \widetilde{h}(k) + 1)$$
(4.1)

has an algebraic singularity at k = 0 when $s \neq v - 2$.

Lemma. Assume that (i) the condition (2.8) holds for p > v + 1 and n = 2, 3, 4 (ii) for |x| large enough $|x|^p \int dy |\rho_T(xy0)| < M$, then

$$\widetilde{S}(k) \sim (\beta b C_{\nu})^{-1} |k|^{\nu - s}, \qquad |k| \to 0$$
(4.2)

Proof. Writing Eq. (2.6) with $Q = \{x_1 = 0\}$, a single point at the origin, gives

$$\beta^{-1}(\nabla h)(x) = e^{2} \int dy F(x - y) \rho(y|0) + \frac{e^{2}}{\rho^{2}} \int dy F(x - y) [\rho_{T}(xy0) + \delta(y) \rho_{T}(x0)] = e^{2} \int dy F(x - y) \rho(y|0) + \frac{e^{2}}{\rho^{2}} \int dy F(y) r(xy)$$
(4.3)

with

$$r(xy) = \rho_T(xy0) + \delta(x - y) \rho_T(x0) + \delta(x) \rho_T(y0)$$
(4.4)

To obtain the last term of (4.3) use has been made of the translation invariance of the correlations and of the antisymmetry of the force. We take the Fourier transform of Eq. (4.3) (notice that by the assumption (ii), r(xy) is jointly integrable in x and y), and with (4.1) we get

$$ik\beta^{-1}\left[\frac{e^2}{\rho}\widetilde{S}(k)-1\right] = -ik\widetilde{\phi}(k)\,\widetilde{S}(k) + \frac{e^2}{\rho}\int dyF(y)\int dxe^{ik\cdot x}r(xy)$$
(4.5)

Multiplying (4.5) by $i(\hat{k}/|k|)$ ($|\hat{k}| = 1$) for $k \neq 0$ leads to

$$\widetilde{\phi}(k)\,\widetilde{S}(k) = \beta^{-1} - \frac{1}{\beta e^2 \rho}\,\widetilde{S}(k) + \frac{f(k)}{|k|} \tag{4.6}$$

where we have set

$$f(k) = -i\frac{e^2}{\rho}\hat{k} \cdot \int dy F(y) \int dx e^{ik \cdot x} r(xy)$$
(4.7)

It is $known^{(10)}$ that under the condition (i) the following sum rules are valid:

$$e \int dx \rho(x \mid x_1 \cdots x_n) = 0 \tag{4.8a}$$

$$e \int dx x \rho(x \mid x_1 \cdots x_n) = 0 \tag{4.8b}$$

for n = 1, 2.

It is easy to see from the definition of the truncated functions that they imply

$$\int dx r(xy) = 0 \tag{4.9a}$$

$$\int dx x r(xy) = 0 \tag{4.9b}$$

We now let $|k| \rightarrow 0$ in (4.6). It follows from (4.8) and (4.9) that $\tilde{S}(k) = o(1)$ and

$$\int dx e^{ik \cdot x} r(xy) = \int dx (e^{ik \cdot x} - 1 - ik \cdot x) r(xy) = o(|k|)$$

as $|k| \rightarrow 0$. Assumption (ii) implies that also f(k) = o(|k|), and hence (4.6) gives

$$\lim_{|k|\to 0} \tilde{\phi}(k) \, \tilde{S}(k) = \beta^{-1}$$

This, with (2.1) gives the result of the lemma.

Then the following considerations give the result of the proposition. Consider first the case where 0 < s < v - 1. If the conditions of the lemma are satisfied, then $\tilde{h}(k)$ has a singularity of order $|k|^{v-s}$ as $|k| \to 0$, and therefore h(x) has a term of order $|x|^{-(2v-s)}$ in its asymptotic development around $|x| = \infty$ [h(x) may however have slower decaying contributions coming from other possible singular points of $\tilde{h}(k)$ located on the real axis]. If the conditions of the lemma do not hold, then some correlations have to decay as or slower than $|x|^{-(v+1)}$, hence not faster than $|x|^{-(2v-s)}$ since 2v - s > v + 1.

When $v-1 \le s < v$, the result of the lemma is still true if the conditions (i) and (ii) hold with p > s + 2. Indeed, in this case, the analysis of Ref. 10 shows that the sum rules (4.8a) and (4.8b) are valid whenever the cluster-

ing is faster than the force $(\sim |x|^{-(s+1)})$ and $|x|^{-(s+2)}$, respectively). Then we must again conclude that h(x) cannot decay faster than $|x|^{-(2\nu-s)}$. Since now $2\nu - s \leq s+2$, there is a contradiction with the hypothesis (i) with p > s+2. Therefore the clustering cannot be faster than $|x|^{-(s+2)}$.

Comments

(1) In the Coulomb case s = v - 2, $\tilde{S}(k)$ is not singular at k = 0 and the result (4.2) of the lemma is the Stillinger-Lovett relation. Its derivation here is essentially identical to that given in Ref. 12.

(2) The behavior (4.2) of $\tilde{S}(k)$ for $k \to 0$ is usually obtained from the heuristic argument that the Fourier transform of the direct correlation function $\tilde{c}(k)$ behaves as $-\beta \tilde{\phi}(k)$ as $k \to 0$.⁽²⁾ Here, under the cluster assumptions of the lemma, Eq. (4.2) is an exact result.

(3) At high temperature and for 0 < s < v - 1 ($s \neq v - 2$), it is believed that the conditions of the lemma are fulfilled, and moreover that $\tilde{S}(k)$ has no other singularity for real k than (4.2). Hence $S(x) \simeq \text{const} \cdot (k_B T/b) |x|^{-(2v-s)}$ would be the correct asymptotic behavior.

In the case of the bidimensional electron film v=2, s=1 (corresponding to the border line s=v-1), we find that S(x) has to decay as or slower than $|x|^{-3}$. At high temperature, it is again believed that $S(x) \sim -4k_BT|x|^{-3}$ is correct.⁽¹⁴⁾

(4) As mentioned in the proof of Proposition 6, h(x) can decay slower than $1/|x|^{2\nu-s}$. This is nicely illustrated in the one-dimensional model of Dyson and Mehta⁽¹⁵⁾ with $-\ln|x|$ interaction. Indeed for $k_BT = 1$ the asymptotic behavior is $-1/\pi^2 \rho^2 |x|^2$, but for $k_BT = 1/4$ one finds $\cos(2\pi\rho |x|)/(4\rho |x|)$.

(5) The extension of the lemma and of Proposition 6 to multicomponent systems is immediate. It is found that the Fourier transform $\tilde{S}(k)$ of the charge-charge correlation

$$S(x) = \sum_{\alpha\gamma} e_{\alpha} e_{\gamma} [\rho(x\alpha, 0\gamma) - \rho_{\alpha} \rho_{\gamma} + \delta_{\alpha\gamma} \delta(x) \rho_{\alpha}]$$

behaves as in (4.2). Hence, the particle correlations (or at least some of them) must have a slow decay.

APPENDIX A

In this Appendix, we give some arguments indicating that the bounds (ii) of Propositions 1, 2, 4 are quite reasonable. We first consider the case of Proposition 1. Since $\rho(xx0) = 0$ in the OCP, we obtain from the definition of the Ursell correlation functions

$$\rho_T(xx0) = \rho_T(x00) = -2\rho^3 h(|x|)$$
(A1)

Furthermore, the perfect screening rule applied to the excess charge density $e\rho(y|x0)$ gives⁽¹⁰⁾

$$\int dy \rho_T(xy0) = -2\rho^2 h(|x|) \tag{A2}$$

It is obvious that the bound (1.ii) is compatible with the exact identities (A1) and (A2).

Starting from the usual Mayer's expansion and using the principles of topological reduction, it can be shown⁽¹³⁾ that

$$\rho_{T}(xy0) = \rho^{3} \{ \exp[\mathcal{T}(xy0)] - 1 \} \{ 1 + h(|x|) + h(|y|) + h(|x - y|) \}$$

+ $\rho^{3} [h(|x|) h(|y|) + h(|x|) h(|x - y|) + h(|y|) h(|x - y|)] \exp[\mathcal{T}(xy0)]$
+ $\rho^{3} h(|x|) h(|y|) h(|x - y|) \exp[\mathcal{T}(xy0)]$ (A3)

where $\mathcal{T}(xy0)$ is formally given by the following diagrammatic expansion:

$$\mathcal{F}(xy0) =$$
 The sum of all simple connected diagrams
with the three root points x, y, and 0,
one or more unlabeled ρ -weighted field
points, three or more h bonds, no
articulation points, no direct bonds
connecting the root points and no articulation
pairs, such that the diagram does not become
disconnected if the root points are removed (A4)

The diagrams appearing in (A4) are highly connected; for instance, a field point is at least connected with three other points. If h decays algebraically, as assumed *a priori* in Proposition 1, there exist some constants b and c such as

$$|h(|x|)| < \frac{b}{c+|x|^p} \tag{A5}$$

which implies

$$\left| \int dy h(|y|) h(|x-y|) \right| < \frac{d}{2^{p}c + |x|^{p}}$$
(A6)

where d is some constant. Using (A6), it can be easily checked that each diagram of (A4) is bounded, for |x| sufficiently large, by $N(t)/|x|^p$, with $t = \min(|y|, |x - y|)$ and N(t) a bounded function which is $O(1/t^p)$ as $t \to \infty$. This indicates that $\mathcal{T}(xy0)$ and consequently $\rho_T(xy0)$ are bounded by similar expressions. Let us emphasize that the bound (1.ii) is much weaker than the previous semiheuristic estimation, since M(t) is only assumed to go to zero at infinity.

The bound (2.ii) is compatible with (A1) and (A2). Assuming a priori a "slow" decay of h [for instance $h(|x|/2) < \text{const} \cdot h(|x|)$ for |x| sufficiently large], we estimate from (A4) that $\rho_T(xy0)$ should be bounded by a constant times |h(|x|)| |h(t)| for |x| and t sufficiently large. The bound (2.ii) is much weaker than this estimation.

In Proposition 4, there are *n* fixed points $(x_1 \cdots x_n) = Q$. Similarly to (A1) and (A2), we have

$$R(xxQ) = -2\rho[\rho(xQ) - \rho\rho(Q)]$$

$$R(xx_jQ) = -\rho[\rho(xQ) - \rho\rho(Q)] - \rho\rho(Q)h(|x - x_j|)$$
(A7)

and

$$\int dy R(xyQ) = -(n+1)[\rho(xQ) - \rho\rho(Q)]$$
(A8)

Taking into account the general clustering property (2.8), we see that (4.ii) is compatible with (A7) and (A8). Similarly to (A4), we may write a diagrammatic expansion of R(xyQ) in terms of h. As before, for the cases of Propositions 1 and 2, the bound (4.ii) appears to be much weaker than the estimations obtained from this expansion.

APPENDIX B

In this Appendix, we prove three lemmas useful in the derivation of Propositions 1, 2, and 4.

Lemma 1. If the condition (1.ii) is fulfilled, then

$$\int dy F(x-y) \rho_T(xy0) = o\left(\frac{1}{|x|^{p-1}}\right)$$
(B1)

Proof. The short-range part of the potential gives a contribution to the left-hand side of (B1) which is bounded by

$$\frac{M}{|x|^p} \int dy \left| (\nabla \phi^0)(x-y) \right| = O\left(\frac{1}{|x|^p}\right)$$
(B2)

where $M = \sup\{M(t); t \ge 0\}$. Thus it is sufficient to consider the Coulomb part of the force for proving (B1). x being given, we have

$$\int dy \cdots = \sum_{i=1}^{3} \int_{\mathscr{D}_{i}} dy \cdots$$
$$\mathscr{D}_{1} = \{ |y| \leq |x|; |y| \leq |y-x| \}$$
$$\mathscr{D}_{2} = \{ |y-x| \leq |x|; |y-x| \leq |y| \}$$
$$\mathscr{D}_{3} = \{ |x| \leq |y|; |x| \leq |y-x| \}$$
(B3)

Using the bound involved in (1.ii), we obtain for |x| large enough

$$\left| \int_{\mathscr{D}_{1}} dy F^{c}(x-y) \rho_{T}(xy0) \right| < \frac{1}{|x|^{p}} \int_{\mathscr{D}_{1}} dy \frac{M(|y|)}{|y-x|^{2}}$$
$$< \frac{1}{|x|^{p}} \int_{\mathscr{D}_{1}} dy \frac{M(|y|)}{|y|^{2}}$$
$$< \frac{1}{|x|^{p}} \int_{|y| \le |x|} dy \frac{M(|y|)}{|y|^{2}}$$
(B4)

Since M(t) goes to zero as $t \to \infty$, we have

$$\int_{|y| \le |x|} dy \frac{M(|y|)}{|y|^2} = o(|x|)$$
(B5)

and thus from (B4)

$$\int_{\mathscr{D}_1} dy F^c(x-y) \,\rho_T(xy0) = o\left(\frac{1}{|x|^{p-1}}\right)$$
(B6)

Similarly, we find

$$\int_{\mathscr{D}_2} dy F^c(x-y) \,\rho_T(xy0) = o\left(\frac{1}{|x|^{p-1}}\right) \tag{B7}$$

Since the points x, y, and 0 play identical roles in $\rho_T(xy0)$ the bound (2.ii) also gives

$$|\rho_T(xy0)| < \frac{M(|x|)}{|y-x|^p}$$
 (B8)

for $y \in \mathcal{D}_3$. Therefore, we have

$$\left| \int_{\mathscr{D}_{3}} dy F^{c}(x-y) \rho_{T}(xy0) \right| < M(|x| \int_{\mathscr{D}_{3}} dy \frac{1}{|y-x|^{p+2}} < M(|x|) \int_{|y-x| \ge |x|} \frac{1}{|y-x|^{p+2}} = \frac{4\pi M(|x|)}{(p-1)|x|^{p-1}} = o\left(\frac{1}{|x|^{p-1}}\right)$$
(B9)

This, with (B6) and (B7) gives the result of the lemma.

Lemma 2. If the conditions of Proposition 2 are fulfilled, then

$$\int dy F(x-y) \rho_T(xy0) = o(|x|h(|x|)) + o\left(\frac{1}{|x|^2} \int_{|x| \le y} dyh(|y|)\right)$$
(B10)

Proof. As in Lemma 1, it is again sufficient to consider the Coulomb part of the force. The integral upon y is split into three parts as in (B3). By identical manipulations to the ones leading to (B6) and (B7), we find

$$\int_{\mathscr{D}_1 \cup \mathscr{D}_2} dy F^c(x - y) \,\rho_T(xy0) = o(|x| \,h(|x|)) \tag{B11}$$

Similarly to (B8), we can write

$$|\rho_{T}(xy0)| < M(|x|)|h(|x-y|)|$$
(B12)

for $y \in \mathcal{D}_3$. Then, we have

$$\left| \int_{\mathscr{D}_{3}} dy F^{c}(x-y) \rho_{T}(xy0) \right| < M(|x|) \int_{\mathscr{D}_{3}} dy \frac{|h(|x-y|)|}{|x-y|^{2}}$$
$$< M(|x|) \int_{|x-y| \ge |x|} dy \frac{|h(|x-y|)|}{|x-y|^{2}}$$
$$< \frac{M(|x|)}{|x|^{2}} \int_{|x| \le |y|} dy |h(|y|)|$$
(B13)

The condition (2.i) implies that h(|y|) has a constant sign for |y| large enough and then

$$\int_{|x| \le |y|} dy \, |h(|y|)| = \left| \int_{|x| \le |y|} dy h(|y|) \right|$$
(B14)

for |x| large enough. Using (B14) in (B13) together with $M(|x|) \rightarrow 0$ as $|x| \rightarrow \infty$, we obtain

$$\int_{\mathscr{D}_3} dy F^c(x-y) \,\rho_T(xy0) = o\left(\frac{1}{|x|^2} \int_{|x| \le |y|} dy h(|y|)\right) \tag{B15}$$

Combining (B11) and (B15), we find (B10).

Lemma 3. If the condition (4.ii) is fulfilled, then

$$\int dxg(x) \int dy F(\lambda x - y) R(\lambda x, y, Q) = o\left(\frac{1}{\lambda^{p-1}}\right)$$
(B16)

Proof. Making the variable change $x' = \lambda x$, we transform the lefthand side of (B16) into

$$\frac{1}{\lambda^3} \int dx' g\left(\frac{x'}{\lambda}\right) \int dy F(x'-y) R(x'yQ)$$
(B17)

Since g(x) vanishes for $|x| \leq a$, the integral upon x' in (B17) is restricted to $|x'| \geq \lambda a$. Using Lemma 1 with R in place of ρ_T , x' in place of x, and Q in place of 0, we then obtain

$$\frac{1}{\lambda^3} \int dx' g\left(\frac{x'}{\lambda}\right) \int dy F(x'-y) \ R(x'yQ) = o\left(\frac{1}{\lambda^3} \int dx' g\left(\frac{x'}{\lambda}\right) \frac{1}{|x'|^{p-1}}\right)$$
(B18)

which leads to (B16) by returning to the variable $x = x'/\lambda$.

APPENDIX C

In this Appendix, we give the proof of Proposition 2.

Using (3.3) and lemma 2 of Appendix B, the BGY equation (3.1) becomes

$$\beta^{-1} \frac{dh(r)}{dr} = e \frac{c}{r^2} - I(r) + o(I(r)) + o(rh(r))$$
(C1)

where

$$I(r) = \frac{4\pi e^2 \rho}{r^2} \int_r^\infty dt t^2 h(t)$$

and c is the total excess charge defined in Section 3.1. Using the general clustering property (2.8) and integrating (C1) from r to ∞ , we obtain

$$\beta^{-1}h(r) = -e\frac{c}{r} + o\left(\frac{1}{r}\right) \tag{C2}$$

which implies c = 0.

Let us integrate now (C1) from r to $r+\delta$ where δ is some fixed parameter, $\delta > 0$. We get

$$\int_{r}^{r+\delta} dt I(t) + o\left(\int_{r}^{r+\delta} dt I(t)\right)$$
$$= \beta^{-1} [h(r) - h(r+\delta)] + o\left(\int_{r}^{r+\delta} dt th(t)\right)$$
(C3)

Using condition (2.i) we have for r sufficiently large

$$|h(r) - h(r+\delta)| < 2|h(r)|$$

$$\left| \int_{r}^{r+\delta} dt th(t) \right| < \frac{\delta}{2} (2r+\delta)|h(r)|$$
(C4)

Thus, for any given $\varepsilon > 0$, there exists R_1 such as for $r > R_1$ the modulus of the right-hand side of (C3) is bounded by

$$2\beta^{-1}|h(r)| + \varepsilon \pi e^2 \rho \delta r |h(r)| \tag{C5}$$

Furthermore, there exists R_2 such as for $r > R_2$, the modulus of the lefthand side of (C3) is bounded below by

$$\frac{1}{2} \left| \int_{r}^{r+\delta} dt I(t) \right| \tag{C6}$$

Denoting $R_3 = \max(R_1, R_2)$, we then obtain for $r > R_3$

$$\left|\int_{r}^{r+\delta} dt I(t)\right| < 4\beta^{-1} |h(r)| + 2\varepsilon \pi \rho e^{2} \delta r |h(r)| \tag{C7}$$

The mean value theorem gives

$$\int_{r}^{r+\delta} dt I(t) = \delta I(r+\xi)$$
(C8)

with $0 \le \xi \le \delta$. The condition (2.i) implies that h(r) has a constant sign for r sufficiently large. We then have

$$|I(r+\xi)| = \frac{4\pi e^2 \rho}{(r+\xi)^2} \int_{r+\xi}^{\infty} dt t^2 |h(t)| > 4\pi e^2 \rho \int_{r+\xi}^{\infty} dt |h(t)|$$
(C9)

which implies using (C8)

$$\left|\int_{r}^{r+\delta} dt I(t)\right| > 4\pi e^{2}\rho\delta\left(\int_{r}^{\infty} dt \left|h(t)\right| - \int_{r}^{r+\xi} dt \left|h(t)\right|\right)$$
(C10)

Combining (C7) and (C10), we obtain

$$\int_{r}^{\infty} dt |h(t)| < \delta \left(1 + \frac{1}{\pi \rho \beta e^2 \delta^2} \right) |h(r)| + \frac{\varepsilon}{2} r |h(r)|$$
(C11)

where we have again used the monotonic decay of |h(r)| and $\xi \leq \delta$. Defining $R = \max[R_3, (2\delta/\epsilon)(1 + 1/\pi\rho\beta e^2\delta^2)]$, (C11) gives for r > R

$$\int_{r}^{\infty} dt |h(t)| < \varepsilon r |h(r)|$$
(C12)

The inequality (C12) can be rewritten as

$$\frac{d}{dr}\ln\int_{r}^{\infty}dt\,|h(t)| < \frac{d}{dr}\ln\left(\frac{1}{r}\right)^{1/\varepsilon}$$
(C13)

Integrating (C13) from R to r and using the monotonicity of the logarithm, we find

$$\int_{r}^{\infty} dt |h(t)| < \left(\frac{R}{r}\right)^{1/\varepsilon} \int_{R}^{\infty} dt |h(t)|$$
(C14)

Since

$$\int_{r}^{\infty} dt |h(t)| > \int_{r}^{2r} dt |h(t)| > r |h(2r)|$$
(C15)

we finally obtain for r > 2R

$$|h(r)| < \left(\frac{2R}{r}\right)^{1+1/\epsilon} \frac{1}{R} \int_{R}^{\infty} dt \, |h(t)|$$
 (C16)

The final step of the proof immediately follows from (C16). For any given p > 0, there exists some R > 0 such as the inequality (C16) holds with $\varepsilon = 1/p$. This implies

$$\lim_{r \to \infty} r^p h(r) = 0 \quad \text{for any} \quad p > 0 \tag{C17}$$

which is the required result.

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APPENDIX D

We prove the Proposition 3.

Working out the asymptotic behavior of the field from (3.3) and (3.i) gives (setting $\lambda = 1$)

$$E(r) = \frac{c}{r^2} - \frac{4\pi e \rho A}{r^2} \int_r^\infty dt \frac{\cos t^{\alpha}}{t^{p-2}} + O\left(\frac{1}{r^p}\right) \\ = \frac{c}{r^2} + \frac{4\pi e \rho A}{\alpha} \left[\frac{\sin r^{\alpha}}{r^{p-1+\alpha}} - \frac{(p+\alpha-3)}{r^2} \int_r^\infty dt \frac{\sin t^{\alpha}}{t^{p+\alpha-2}}\right] + O\left(\frac{1}{r^p}\right)$$
(D1)

and by the Riemann Lebesgue lemma

$$\frac{1}{r^2} \int_r^\infty dt \, \frac{\sin t^\alpha}{t^{p+\alpha-2}} = \frac{1}{r^{p-1+\alpha}} \int_1^\infty dt \, \frac{\sin(tr)^\alpha}{t^{p+\alpha-2}} = o\left(\frac{1}{r^{p-1+\alpha}}\right) \tag{D2}$$

The hypothesis (3.ii) implies that the last term of the righ-hand side of (3.1) is $O(1/r^p)$. Thus with (3.i), (D1) and (D2) we find that the asymptotic behavior of (3.1) is

$$-A\alpha \frac{\sin r^{\alpha}}{r^{p-\alpha+1}} = \frac{4\pi e^2 \rho A}{\alpha} \frac{\sin r^{\alpha}}{r^{p-1+\alpha}} + \frac{ec}{r^2} + \begin{cases} o\left(\frac{1}{r^{p-1+\alpha}}\right), & 0 < \alpha < 1\\ O\left(\frac{1}{r^{p}}\right), & \alpha > 1 \end{cases}$$

This implies c = 0 and A = 0 for all $\alpha > 0$, $\alpha \neq 1$.

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