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# Algebraic infection of charge correlations of a classical electrolyte at the critical point of the liquid-gas transition

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## Abstract

We consider a classical Two-Component Plasma analog of the Restricted Primitive Model of electrolyte, where the hard-core interaction is replaced by a soft differentiable potential. Within the Born-Green-Yvon hierarchy for the equilibrium distribution functions, we shed light on an infection mechanism where the charge correlations are polluted by the density correlations at the critical point of the liquid-gas transition. This implies an algebraic decay of critical charge correlations. Such breakdown of exponential clustering should provide dielectric rather than conducting properties at the critical point, leading to the violation of certain charge-charge sum rules. This is in agreement with Monte Carlo simulations.

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## I. INTRODUCTION

The liquid-gas transition of a classical electrolyte is a long standing problem, which has been nicely reviewed by Fisher [1]. This has motivated numerous experimental and theoretical works. On the theoretical side, there still remain difficult questions which have not been satisfactorily answered. A first widely debated problem is about the universality class of the behavior of thermodynamic properties near the critical point [2]. It was believed, over many years, that the critical exponents are given by mean-field approaches. However, since the end of the last century, careful numerical and experimental inspections at the immediate vicinity of the critical point strongly suggest that these exponents are of the Ising-type. There exists now a reasonably general consensus that the transition does belong to the Ising universality class [3].

A second controversial question is by concerning the conducting versus dielectric nature of the critical point, in particular for the fully (charge and size) symmetric Restricted Primitive Model (RPM) [1, 4]. Away from the critical point, numerical simulations, see *e.g.* [4] or [5], have convincingly shown that both the liquid and the gas phases display perfect screening properties that are typical of conducting phases. However, a first suspicion about the breakdown of perfect screening properties, close to the critical point, was pointed out by Caillol [6]. More recently, such breakdown was confirmed through sophisticated Monte-Carlo simulations [4, 7]: they show via state-of-the-art finite-size scaling analysis [8] that the second moment Stillinger-Lovett (SL) sum rule [9] for equilibrium charge correlations is violated. This implies that infinitesimal external charges are no longer perfectly screened (see *e.g.* [10]). Here note that the charge-charge structure factor can be written in even powers of the wave number  $k$ . The square root of the coefficient of  $k^2$ , the second moment charge-charge correlation length, equals the Debye screening length [1]. This is a consequence of screening and is referred to as the second SL sum rule. The first SL sum rule dictates that the structure factor is zero at  $k = 0$ . This emerges from the charge neutrality in the system.

The large-distance decay of charge-charge correlations,  $S(r)$ , at the critical point

is a central question, directly related to the system's conducting or dielectric nature. If the density-density correlations decay as a power law [1], namely as  $1/r^{1+\eta}$  with  $\eta > 0$ , various theoretical approaches predict an exponential decay of equilibrium charge correlations for the RPM. In particular, this is observed in a solvable mean-spherical model [11], which is expected to share common properties with the RPM. In this paper, we consider a fully symmetric Two-Component Plasma (TCP), which is the analog of the RPM where the hard core interaction is replaced by a smooth differentiable potential. Our main purpose is to show that the critical charge correlations for this TCP are infected by the slow algebraic decay of the critical density correlations. In other words, the charge correlations also decay in a power law fashion. This is achieved through an inspection of the large-distance behavior of the equilibrium Born-Green-Yvon (BGY) equations. Our analysis sheds light on the infection mechanism, alongside providing the power law decay of  $S(r)$  as  $1/r^{6+\eta}$ . Note that within the solvable mean-spherical model [11], an infection mechanism occurs for its charge-asymmetric version but not for the symmetric version analog to the present model and the RPM. An analogous coupling between the charge-charge and density-density correlations can possibly be realized, even for the RPM, within the generalized Debye-Hückel theory of Lee and Fisher, via consideration of charge fluctuation that complements the density fluctuations.

In Section II, we describe the considered TCP model. Like the RPM, this model is expected to undergo a liquid-vapor transition at low density, general features of which are briefly stated. The BGY hierarchy is introduced in Section III. We argue that the corresponding BGY equations should remain valid at the critical point. The infection mechanism which results from the coupling between charge and density correlations is highlighted in Section IV. We show how this mechanism prevents the exponential decay of  $S(r)$ . A plausible algebraic-decay scenario for two-and three-body particle correlations is shown to be consistent with the BGY equations in Section V A. Accordingly  $S(r)$  is found to decay as  $1/r^{6+\eta}$ . We briefly discuss in Section V B how our results strongly suggest that the critical point is indeed dielectric, in agreement with numerical simulations [7].

## II. CHARGE SYMMETRIC IONIC FLUID

### A. Pairwise interactions

We consider a two-component classical plasma (TCP) made of two species ( $\alpha = +, -$ ) of mobile particles, carrying charges  $\pm q$ , in space dimension  $d = 3$ . The particles interact via a sum of pairwise interactions

$$\begin{aligned} u_{++}(r) &= u_{--}(r) = v_{\text{SR}}(r) + q^2 v_{\text{C}}(r) \\ u_{+-}(r) &= u_{-+}(r) = v_{\text{SR}}(r) - q^2 v_{\text{C}}(r), \end{aligned} \quad (\text{II.1})$$

which include the familiar Coulomb potential  $v_{\text{C}}(r) = 1/r$  and a short-range repulsive interaction  $v_{\text{SR}}(r)$ . The short-range interaction diverges positively faster than  $1/r$  when  $r \rightarrow 0$  in order to avoid the collapse between oppositely charged particles. A possible choice for this is

$$v_{\text{SR}}(r) = V_0 \left[ \frac{\sigma}{r} \right]^{12} \exp(-r/\sigma), \quad (\text{II.2})$$

with  $V_0 > 0$ . In the following, the analysis will be performed for general forms of  $v_{\text{SR}}(r)$  and is not specific to the choice (II.2).

We restate, the present fully symmetric TCP is quite similar to the celebrated RPM, which is fully symmetric with respect to the charges  $\pm q$  and the hard-core diameters  $\sigma$ . Here the hard-core potential is replaced by a soft form which is differentiable everywhere, except at  $r = 0$ . This allows us to introduce the BGY hierarchy as described in the next Section III.

### B. Liquid-vapor phase transition

In this paper, we assume that the thermodynamic limit (TL) of the present model exists for any choice of  $v_{\text{SR}}(r)$ . Note that this has been proven only for short-range regularizations of the Coulomb interaction [12] (see also the review [13]). Overall charge neutrality in the system is imposed. Not at too low temperatures, the system in a fluid phase with a common uniform particle density  $\rho$  for both species, is invariant under translations once the TL has been taken. As suggested by numerical

simulations for similar systems [14], like the RPM for instance [2], the present model is expected to undergo a liquid-vapor phase transition. Similar to what occurs for classical fluids made with neutral particles and Lenard-Jones interactions [15], at a fixed temperature ( $T$ ) lower than some critical value ( $T_c$ ), depending upon the overall density, in equilibrium there will be coexistence of vapor and liquid phases with densities  $\rho_{\text{vap}}(T)$  and  $\rho_{\text{liq}}(T)$ . At the critical point ( $T_c, \rho_c$ ),  $\rho_c$  being the critical value of overall density,  $\rho_{\text{vap}}$  and  $\rho_{\text{liq}}$  become identical, i.e.,  $\rho_c = \rho_{\text{liq}}(T_c) = \rho_{\text{vap}}(T_c)$ .

In the vapor phase, at low densities, particle correlations should decay exponentially fast at large distances. This is strongly suggested by Debye theory, and also by systematic corrections to this mean-field approach derived within the Abe-Meeron diagrammatic expansions [16, 17] (see also the rigorous proof by Brydges and Federbush [18] for similar systems). Perfect screening of external charges is then observed, as encoded in the SL second moment sum rule [9] concerning the charge correlations. When the density increases, the system is expected to remain in a conducting state for both vapor and liquid phases, with perfect screening properties. Note that although particle correlations then might decay slower than an exponential [19], sufficiently fast power-law decays ensure the validity of the SL rule [20] (see also the review [10]). Moreover, the numerical simulations show the persistence of free charges which in turn ensure that screening properties still hold. However, as shown via sophisticated Monte-Carlo simulations for the RPM [7], this picture becomes quite doubtful at the critical point ( $T_c, \rho_c$ ). As a consequence of the coupling between fluctuations in particle and charge correlations, the latter is expected to be infected by the slow power-law decay of critical particle correlations, and ultimately the second SL sum rule is violated. The main purpose of the present paper is to analyze the infection mechanism within the BGY hierarchy.

### III. THE BGY HIERARCHY

The BGY hierarchy should be *a priori* valid for the distribution functions of any infinitely extended equilibrium state, provided that the involved spatial integrals do converge in the infinite space. We first write the second BGY equation for the pair distribution functions  $\rho_{\alpha_1\alpha_2}(\mathbf{r}_1, \mathbf{r}_2)$  of the infinite system in an homogeneous fluid

phase. Then we show that all terms are well-behaved if we assume weak clustering properties for the particle correlations, which are consistent with their expected large-distance decays in the fluid phase, including the critical point. This confirms that the BGY hierarchy can be safely used, even at the critical point.

### A. The second BGY equations

In a fluid phase, the particle distribution functions are invariant under translations, and the second BGY equations for the pair distribution functions  $\rho_{++}(\mathbf{r}_1, \mathbf{r}_2) = \rho_{--}(\mathbf{r}_1, \mathbf{r}_2)$  and  $\rho_{+-}(\mathbf{r}_1, \mathbf{r}_2) = \rho_{-+}(\mathbf{r}_1, \mathbf{r}_2)$ , can be written by fixing one particle at the origin,  $\mathbf{r}_1 = \mathbf{0}$ , and by taking the gradient with respect to the position  $\mathbf{r}_2 = \mathbf{r}$  of the second particle. This provides

$$\begin{aligned} \nabla \rho_{++}(\mathbf{0}, \mathbf{r}) &= \beta \rho_{++}(\mathbf{0}, \mathbf{r}) \mathbf{F}_{++}(\mathbf{0}, \mathbf{r}) \\ &+ \beta \int d\mathbf{r}' [\rho_{+++}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \mathbf{F}_{++}(\mathbf{r}', \mathbf{r}) + \rho_{++-}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \mathbf{F}_{-+}(\mathbf{r}', \mathbf{r})] , \end{aligned} \quad (\text{III.1})$$

and

$$\begin{aligned} \nabla \rho_{+-}(\mathbf{0}, \mathbf{r}) &= \beta \rho_{+-}(\mathbf{0}, \mathbf{r}) \mathbf{F}_{+-}(\mathbf{0}, \mathbf{r}) \\ &+ \beta \int d\mathbf{r}' [\rho_{+--}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \mathbf{F}_{+-}(\mathbf{r}', \mathbf{r}) + \rho_{+-+}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \mathbf{F}_{-+}(\mathbf{r}', \mathbf{r})] . \end{aligned} \quad (\text{III.2})$$

In these equations,  $\mathbf{F}_{\alpha_1 \alpha_2}(\mathbf{r}', \mathbf{r}) = -\nabla_{\mathbf{r}} u_{\alpha_1 \alpha_2}(\mathbf{r}', \mathbf{r})$  is the force exerted on a particle with species  $\alpha_2$  and position  $\mathbf{r}$ , by a particle with species  $\alpha_1$  and position  $\mathbf{r}'$ . This force can be decomposed as

$$\begin{aligned} \mathbf{F}_{++}(\mathbf{r}', \mathbf{r}) &= \mathbf{F}_{\text{SR}}(\mathbf{r} - \mathbf{r}') + q^2 \mathbf{F}_{\text{C}}(\mathbf{r} - \mathbf{r}') \\ \mathbf{F}_{-+}(\mathbf{r}', \mathbf{r}) &= \mathbf{F}_{\text{SR}}(\mathbf{r} - \mathbf{r}') - q^2 \mathbf{F}_{\text{C}}(\mathbf{r} - \mathbf{r}') \end{aligned} \quad (\text{III.3})$$

with the short-range part

$$\mathbf{F}_{\text{SR}}(\mathbf{r} - \mathbf{r}') = -\nabla_{\mathbf{r}} v_{\text{SR}}(\mathbf{r} - \mathbf{r}') \quad (\text{III.4})$$

and the Coulomb part

$$\mathbf{F}_{\text{C}}(\mathbf{r} - \mathbf{r}') = -\nabla_{\mathbf{r}} v_{\text{C}}(\mathbf{r} - \mathbf{r}') = \frac{\mathbf{r} - \mathbf{r}'}{|\mathbf{r} - \mathbf{r}'|^3} . \quad (\text{III.5})$$

For further purposes, it is useful to express the two- and three-body distribution functions in terms of the corresponding particle correlations, whose dimensionless counterparts are the Ursell functions, namely

$$\begin{aligned}\rho_{\alpha_1\alpha_2}(\mathbf{0}, \mathbf{r}) &= \rho^2[1 + h_{\alpha_1\alpha_2}(r)] \\ \rho_{\alpha_1\alpha_2\alpha_3}(\mathbf{0}, \mathbf{r}, \mathbf{r}') &= \rho^3[1 + h_{\alpha_1\alpha_2}(r) + h_{\alpha_1\alpha_3}(r') + h_{\alpha_2\alpha_3}(|\mathbf{r}' - \mathbf{r}|) + h_{\alpha_1\alpha_2\alpha_3}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}')] .\end{aligned}\tag{III.6}$$

Thanks to both the translational and rotational invariance of the fluid phase, the two-body Ursell functions  $h_{\alpha_1\alpha_2}$  only depend on the relative distance between the fixed particles. Similarly, the three-body Ursell function  $h_{\alpha_1\alpha_2\alpha_3}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}')$  only depends on the relative distances  $(r, r', |\mathbf{r}' - \mathbf{r}|)$ , i.e. the three sides of the triangle formed by the particles. Using the decompositions (III.3) and (III.6) of the force and of the distribution functions, we recast the BGY equations (III.1, III.2) as

$$\begin{aligned}\nabla h_{++}(r) &= \beta h_{++}(r) \mathbf{F}_{\text{SR}}(\mathbf{r}) + \frac{\beta}{2\rho} \int d\mathbf{r}' N(\mathbf{r}') \mathbf{F}_{\text{SR}}(\mathbf{r} - \mathbf{r}') \\ &\quad + \beta \rho \int d\mathbf{r}' \left[ h_{++++}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') + h_{+++-}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \right] \mathbf{F}_{\text{SR}}(\mathbf{r} - \mathbf{r}') \\ &\quad + \beta q^2 h_{++}(r) \mathbf{F}_{\text{C}}(\mathbf{r}) + \frac{\beta}{2\rho} \int d\mathbf{r}' S(\mathbf{r}') \mathbf{F}_{\text{C}}(\mathbf{r} - \mathbf{r}') \\ &\quad + \beta q^2 \rho \int d\mathbf{r}' \left[ h_{++++}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') - h_{+++-}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \right] \mathbf{F}_{\text{C}}(\mathbf{r} - \mathbf{r}') ,\end{aligned}\tag{III.7}$$

and

$$\begin{aligned}\nabla h_{+-}(r) &= \beta h_{+-}(r) \mathbf{F}_{\text{SR}}(\mathbf{r}) + \frac{\beta}{2\rho} \int d\mathbf{r}' N(\mathbf{r}') \mathbf{F}_{\text{SR}}(\mathbf{r} - \mathbf{r}') \\ &\quad + \beta \rho \int d\mathbf{r}' \left[ h_{+--+}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') + h_{+-}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \right] \mathbf{F}_{\text{SR}}(\mathbf{r} - \mathbf{r}') \\ &\quad - \beta q^2 h_{+-}(r) \mathbf{F}_{\text{C}}(\mathbf{r}) - \frac{\beta}{2\rho} \int d\mathbf{r}' S(\mathbf{r}') \mathbf{F}_{\text{C}}(\mathbf{r} - \mathbf{r}') \\ &\quad - \beta q^2 \rho \int d\mathbf{r}' \left[ h_{+--+}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') - h_{+-}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \right] \mathbf{F}_{\text{C}}(\mathbf{r} - \mathbf{r}') .\end{aligned}\tag{III.8}$$

In these equations,  $N(\mathbf{r}')$  is the correlation between particle densities at points  $\mathbf{0}$  and  $\mathbf{r}'$ ,

$$N(\mathbf{r}') = 2\rho^2 [h_{++}(r') + h_{+-}(r')] + 2\rho\delta(\mathbf{r}') ,\tag{III.9}$$

while  $S(\mathbf{r}')$  is the correlation between charge densities at points  $\mathbf{0}$  and  $\mathbf{r}'$ ,

$$S(\mathbf{r}') = 2q^2\rho^2 [h_{++}(r') - h_{+-}(r')] + 2q^2\rho\delta(\mathbf{r}') .\tag{III.10}$$



The BGY equations for the density-density and charge-charge correlations are readily obtained by combining equations (III.7) and (III.8), and they become for  $\mathbf{r} \neq \mathbf{0}$

$$\begin{aligned} \nabla N(\mathbf{r}) = & \beta N(\mathbf{r}) \mathbf{F}_{\text{SR}}(\mathbf{r}) + 2\beta\rho \int d\mathbf{r}' N(\mathbf{r}') \mathbf{F}_{\text{SR}}(\mathbf{r}-\mathbf{r}') + 2\beta \int d\mathbf{r}' H_{\text{dd}}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \mathbf{F}_{\text{SR}}(\mathbf{r}-\mathbf{r}') \\ & + \beta S(\mathbf{r}) \mathbf{F}_{\text{C}}(\mathbf{r}) + 2\beta \int d\mathbf{r}' H_{\text{dc}}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \mathbf{F}_{\text{C}}(\mathbf{r}-\mathbf{r}') , \end{aligned} \quad (\text{III.11})$$

and

$$\begin{aligned} \nabla S(\mathbf{r}) = & \beta S(\mathbf{r}) \mathbf{F}_{\text{SR}}(\mathbf{r}) + 2\beta q^2 \int d\mathbf{r}' H_{\text{cd}}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \mathbf{F}_{\text{SR}}(\mathbf{r}-\mathbf{r}') \\ & + \beta q^4 N(\mathbf{r}) \mathbf{F}_{\text{C}}(\mathbf{r}) + 2\beta q^2 \rho \int d\mathbf{r}' S(\mathbf{r}') \mathbf{F}_{\text{C}}(\mathbf{r}-\mathbf{r}') + 2\beta q^4 \int d\mathbf{r}' H_{\text{cc}}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \mathbf{F}_{\text{C}}(\mathbf{r}-\mathbf{r}') . \end{aligned} \quad (\text{III.12})$$

The various three-body correlations  $H^{(3)}$  are defined as linear combinations of the three-body Ursell functions,

$$\begin{aligned} H_{\text{dd}}^{(3)} &= \rho^3 \left[ h_{++++}^{(3)} + h_{++--}^{(3)} + h_{+-+-}^{(3)} + h_{+--+}^{(3)} \right] \\ H_{\text{dc}}^{(3)} &= \rho^3 \left[ h_{++++}^{(3)} + h_{+---}^{(3)} - h_{++--}^{(3)} - h_{+-+-}^{(3)} \right] \\ H_{\text{cd}}^{(3)} &= \rho^3 \left[ h_{++++}^{(3)} + h_{++--}^{(3)} - h_{+-+-}^{(3)} - h_{+--+}^{(3)} \right] \\ H_{\text{cc}}^{(3)} &= \rho^3 \left[ h_{++++}^{(3)} + h_{+--+}^{(3)} - h_{++--}^{(3)} - h_{+-+-}^{(3)} \right] , \end{aligned} \quad (\text{III.13})$$

and they are related to the equilibrium averages of products of three microscopic particle-density or charge-density operators. As shown by the structure of equations (III.11, III.12), the density and charge correlations are coupled together, as it can be *a priori* expected.

## B. Validity of the BGY equations at the critical point

The various integrals over  $\mathbf{r}'$  involved in the BGY equations (III.11) and (III.12) do converge under rather weak clustering assumptions on the decay of two- and three-body particle correlations. Indeed, since the short-range force decays as an exponential at large distances  $|\mathbf{r}' - \mathbf{r}|$ , the integrals upon  $\mathbf{r}'$  of  $\mathbf{F}_{\text{SR}}(\mathbf{r}' - \mathbf{r})$  times particle correlations are always well behaved. This is not the case of the integrals

with the Coulomb force since  $\mathbf{F}_C(\mathbf{r}' - \mathbf{r})$  decays as  $1/|\mathbf{r}' - \mathbf{r}|^2$  when  $\mathbf{r}'$  is separated from a fixed  $\mathbf{r}$  by infinite distance. In order to ensure the (absolute) convergence of the related integrals, the correlations  $S(\mathbf{r}')$ ,  $H_{dc}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}')$  and  $H_{cc}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}')$  have to decay faster than  $1/|\mathbf{r}'|^{1+\epsilon}$  with  $\epsilon > 0$  when  $|\mathbf{r}'| \rightarrow \infty$ . For any equilibrium state where such weak algebraic decays hold, all terms in the BGY equations are finite: this strongly suggests that these equations are indeed satisfied by the corresponding equilibrium particle correlations.

At the critical point, one expects a slow algebraic decay of all  $n$ -body Ursell functions, with  $n = 2, 3, \dots$ , typically as  $1/r^{1+\eta}$  with a strictly positive exponent  $\eta > 0$ . Hence, correlations  $S(\mathbf{r}')$ ,  $H_{dc}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}')$  and  $H_{cc}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}')$  decay at least as  $1/|\mathbf{r}'|^{1+\eta}$  when  $|\mathbf{r}'| \rightarrow \infty$  since they are linear combinations of two- and three-body Ursell functions. According to the previous analysis, this implies that the BGY equations remain valid at the critical point.

#### IV. THE INFECTION MECHANISM

In order to extract constraints from the BGY equations for  $N(r)$  and  $S(r)$ , we first introduce weak assumptions for the respective decays of two- and three-body Ursell. Such assumptions are shown to be consistent with the internal charge sum rules which are expected to hold in any phase and at the critical point (Section IV A). Then, we highlight an infection mechanism which prevents the exponential decay of  $S(\mathbf{r})$  (Section IV B).

##### A. Clustering assumptions and charge sum rules

At the critical point, according to their respective definitions of particle density (III.9) and charge (III.10) correlations,  $N(\mathbf{r})$  and  $S(\mathbf{r})$  decay at least as  $1/r^{1+\eta}$  at large distances  $r$ . Such a decay should hold for the density correlations  $N(\mathbf{r})$ , in agreement with the divergence of its integral over  $\mathbf{r}$ , implied by the compressibility sum rule (note that here the total density is  $2\rho$ )

$$\int d\mathbf{r} N(\mathbf{r}) = 4\rho^2 k_B T \chi_T . \quad (\text{IV.1})$$

Indeed, the isothermal compressibility  $\chi_T = [\rho \partial P / \partial \rho]^{-1}$ , where  $P$  is the pressure of the system, diverges at the critical point. For the charge correlations  $S(\mathbf{r})$ , one expects a decay faster than  $1/r^{1+\eta}$  in order to satisfy the internal perfect screening rule

$$\int d\mathbf{r} S(\mathbf{r}) = 0 , \quad (\text{IV.2})$$

which requires the integrability of  $S(\mathbf{r})$  over the whole space. Discarding oscillatory behaviors, this implies that  $S(\mathbf{r})$  decays at least as  $1/r^{3+\epsilon}$  with  $\epsilon > 0$  when  $r \rightarrow \infty$ . This leads us to infer

$$h_{++}(r) \sim \frac{A}{r^{1+\eta}} \quad , \quad h_{+-}(r) \sim \frac{A}{r^{1+\eta}} \quad \text{when} \quad r \rightarrow \infty , \quad (\text{IV.3})$$

where the common amplitude  $A$  does not depend on the charges carried by the particles. Note that the charge sum rule (IV.2) is crucial for the consistency of the present picture: it guarantees a minimal screening of Coulomb interactions which in turn do not affect the leading critical tails.

The perfect screening rule (IV.2) means that the total charge carried by the polarization cloud surrounding a given fixed particle, exactly cancels its charge. If now two particles with charges  $e_{\alpha_1}$  and  $e_{\alpha_2}$  are fixed at positions  $\mathbf{0}$  and  $\mathbf{r}$ , the total charge carried by the corresponding polarization cloud should exactly reduce to  $-(e_{\alpha_1} + e_{\alpha_2})$ . Hence, the three-body Ursell functions are expected to satisfy the sum rules [10]

$$\int d\mathbf{r}' \rho \left[ h_{+++}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') - h_{++-}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \right] = -2h_{++}(\mathbf{0}, \mathbf{r}) \quad (\text{IV.4})$$

and

$$\int d\mathbf{r}' \rho \left[ h_{+-+}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') - h_{+--}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \right] = 0 . \quad (\text{IV.5})$$

These sum rules imply that the three-body Ursell functions are integrable when two particle positions are fixed while the third one is sent to infinity. Discarding oscillatory behaviors as for  $S(\mathbf{r})$ , this leads to

$$h_{+++}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \sim h_{++-}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \sim \frac{A_{++}(\mathbf{0}, \mathbf{r})}{|\mathbf{r}' - \mathbf{r}/2|^{1+\eta}} \quad \text{when} \quad r' \rightarrow \infty \quad (\text{IV.6})$$

and

$$h_{+-+}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \sim h_{+--}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \sim \frac{A_{+-}(\mathbf{0}, \mathbf{r})}{|\mathbf{r}' - \mathbf{r}/2|^{1+\eta}} \quad \text{when} \quad r' \rightarrow \infty , \quad (\text{IV.7})$$

while the differences  $[h_{+++}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') - h_{++-}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}')] \quad \text{and} \quad [h_{+-+}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') - h_{+--}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}')] \quad \text{decay as least at } (1/r')^{3+\epsilon}$ . These large-distance behaviors follow from the assumption that the leading critical tails are not affected by the charges of particles. This assumption has been used for deriving (IV.3) for two-body correlations. More generally, the correlations between a given group of  $n > 1$  particles with barycenter  $\mathbf{R}$  on the one hand, and a single particle with position  $\mathbf{r}'$  on the other hand, do not depend at leading order when  $r' \rightarrow \infty$  on the charge of this single particle. The corresponding critical tail reduces to  $1/|\mathbf{r}' - \mathbf{R}|^{1+\eta}$  times an amplitude which only depends on the relative distances between the  $n$  particles in the considered group. In (IV.6) and (IV.7),  $n = 2$  and  $\mathbf{R} = \mathbf{r}/2$ , while the functions  $A_{++}(\mathbf{0}, \mathbf{r})$  and  $A_{+-}(\mathbf{0}, \mathbf{r})$  can be reasonable assumed to be invariant under rotations, namely they only depend on  $r$ . Moreover, and similar to the two-body Ursell functions  $h_{++}$  and  $h_{+-}$ , they should behave as

$$A_{++}(r) \sim A_{+-}(r) \sim \frac{A^{(2)}}{r^{1+\eta}} \quad \text{when } r \rightarrow \infty, \quad (\text{IV.8})$$

while the difference  $[A_{++}(r) - A_{+-}(r)]$  decays as least at  $1/r^{3+\epsilon}$ .

For further purposes, it is interesting to notice that, by symmetry, the behaviors (IV.6) and (IV.7) are valid for any triangular configuration where one distance is kept fixed while the two remaining ones diverge, namely

$$\begin{aligned} h_{+++}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') &\sim \frac{A_{++}(\mathbf{0}, \mathbf{r}')}{|\mathbf{r} - \mathbf{r}'/2|^{1+\eta}} \quad \text{when } r \rightarrow \infty, \mathbf{r}' \text{ fixed} \\ h_{++-}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') &\sim \frac{A_{+-}(\mathbf{0}, \mathbf{r}')}{|\mathbf{r} - \mathbf{r}'/2|^{1+\eta}} \quad \text{when } r \rightarrow \infty, \mathbf{r}' \text{ fixed} \\ h_{+++}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') &\sim \frac{A_{++}(\mathbf{r}, \mathbf{r}')}{|(\mathbf{r} + \mathbf{r}')/2|^{1+\eta}} \quad \text{when } r \rightarrow \infty, (\mathbf{r}' - \mathbf{r}) \text{ fixed} \\ h_{++-}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') &\sim \frac{A_{+-}(\mathbf{r}, \mathbf{r}')}{|(\mathbf{r} + \mathbf{r}')/2|^{1+\eta}} \quad \text{when } r \rightarrow \infty, (\mathbf{r}' - \mathbf{r}) \text{ fixed}, \end{aligned} \quad (\text{IV.9})$$

and

$$\begin{aligned}
h_{++-}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') &\sim \frac{A_{++}(\mathbf{0}, \mathbf{r}')}{|\mathbf{r} - \mathbf{r}'/2|^{1+\eta}} \quad \text{when } r \rightarrow \infty, \mathbf{r}' \text{ fixed} \\
h_{+--}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') &\sim \frac{A_{+-}(\mathbf{0}, \mathbf{r}')}{|\mathbf{r} - \mathbf{r}'/2|^{1+\eta}} \quad \text{when } r \rightarrow \infty, \mathbf{r}' \text{ fixed} \\
h_{++-}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') &\sim \frac{A_{-+}(\mathbf{r}, \mathbf{r}')}{|(\mathbf{r} + \mathbf{r}')/2|^{1+\eta}} \quad \text{when } r \rightarrow \infty, (\mathbf{r}' - \mathbf{r}) \text{ fixed} \\
h_{+--}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') &\sim \frac{A_{--}(\mathbf{r}, \mathbf{r}')}{|(\mathbf{r} + \mathbf{r}')/2|^{1+\eta}} \quad \text{when } r \rightarrow \infty, (\mathbf{r}' - \mathbf{r}) \text{ fixed} . \quad (\text{IV.10})
\end{aligned}$$

Moreover, since the present TCP is fully symmetric, the amplitude functions satisfy the symmetry relations  $A_{++} = A_{--}$  and  $A_{+-} = A_{-+}$ .

The sum rule (IV.4) implies another sum rule for the amplitude functions  $A_{++}$  and  $A_{+-}$ . Let us consider the limit  $r \rightarrow \infty$  of both sides of (IV.4). In the integral in the l.h.s., the leading contributions arise from the regions  $\mathbf{r}'$  close to the origin on the one hand, and  $\mathbf{r}'$  close to  $\mathbf{r}$  on the other hand. According to the decays (IV.9), both regions give identical contributions which lead to

$$\int d\mathbf{r}' \rho \left[ h_{+++}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') - h_{++-}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \right] \sim 2\rho \frac{\int d\mathbf{x} [A_{++}(x) - A_{+-}(x)]}{r^{1+\eta}} \quad , \quad r \rightarrow \infty . \quad (\text{IV.11})$$

Notice that  $[A_{++}(x) - A_{+-}(x)]$  decays as  $1/x^{3+\epsilon}$  like  $S(x)$ , so  $\int d\mathbf{x} [A_{++}(x) - A_{+-}(x)]$  does converge. Comparing the behavior (IV.11) with the large- $r$  decay of the r.h.s of (IV.4) inferred from (IV.3), we find

$$\rho \int d\mathbf{x} [A_{++}(x) - A_{+-}(x)] = -A . \quad (\text{IV.12})$$

Analogous manipulations can be repeated for the sum rule (IV.5). The leading contributions of regions  $\mathbf{r}'$  close to the origin, and  $\mathbf{r}'$  close to  $\mathbf{r}$ , then exactly cancel out by virtue of the symmetry relations  $A_{++} = A_{--}$  and  $A_{+-} = A_{-+}$ . Hence, no additional constraints on  $A_{++}$  and  $A_{+-}$  are imposed by sum rule (IV.5).

We stress that the previous clustering assumptions on the three-body Ursell functions turn out to be perfectly consistent with the three-body charge sum rules. Furthermore, they are satisfied by the Kirkwood superposition approximation [21],

$$\rho_{\alpha_1 \alpha_2 \alpha_3}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) = \rho_{\alpha_1 \alpha_2}(\mathbf{r}_1, \mathbf{r}_2) \rho_{\alpha_1 \alpha_3}(\mathbf{r}_1, \mathbf{r}_3) \rho_{\alpha_2 \alpha_3}(\mathbf{r}_2, \mathbf{r}_3) . \quad (\text{IV.13})$$

which provides the three-body Kirkwood Ursell functions

$$h_{\alpha_1\alpha_2\alpha_3}^{(3,K)} = h_{\alpha_1\alpha_2}h_{\alpha_1\alpha_3} + h_{\alpha_1\alpha_2}h_{\alpha_2\alpha_3} + h_{\alpha_1\alpha_3}h_{\alpha_2\alpha_3} + h_{\alpha_1\alpha_2}h_{\alpha_1\alpha_3}h_{\alpha_2\alpha_3} . \quad (\text{IV.14})$$

## B. Breakdown of the exponential decay of charge correlations

Within the previous clustering assumptions, we have seen that some combinations of correlations decay faster than  $1/r^{1+\eta}$  because of cancellations. In particular such mechanism arises for the charge correlations  $S(r)$  which should decay at least as  $1/r^{3+\epsilon}$ . Let us assume *a priori* that  $S(r)$  decays exponentially fast at the critical point. Consistently, we then also assume that the combinations of three-body correlations, similar to  $[h_{++} - h_{+-}]$  in  $S(r)$ , where cancellations of the critical  $1/r^{1+\eta}$ -tails occur, also decay exponentially fast. Using the behaviors (IV.9) and (IV.10) for the three-body Ursell functions, the corresponding exponential-decay scenario (**EDS**) reads

- **EDS1**:  $S(r)$  decays exponentially fast when  $r \rightarrow \infty$
- **EDS2**:  $H_{\text{cd}}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}')$  decays exponentially fast when  $r \rightarrow \infty$  with either  $\mathbf{r}'$  fixed or  $(\mathbf{r}' - \mathbf{r})$  fixed
- **EDS3**:  $H_{\text{cc}}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}')$  decays exponentially fast when  $r \rightarrow \infty$  with  $(\mathbf{r}' - \mathbf{r})$  fixed
- **EDS4**:  $[A_{++}(x) - A_{+-}(x)]$  decays exponentially fast when  $x \rightarrow \infty$

Then, the strategy consists in showing that this exponential-decay scenario is not consistent with the large- $r$  behavior of the BGY equation (III.12).

Let us analyze, within **EDS**, the large-distance behavior of the various terms in (III.12). In the l.h.s.  $\nabla S(\mathbf{r})$  decays exponentially fast by virtue of **EDS1**. In the r.h.s., we first consider the two terms involving the short-range force  $\mathbf{F}_{\text{SR}}$ . The direct short-range term

$$\mathbf{R}_{\text{SR}}(\mathbf{r}) = \beta S(\mathbf{r}) \mathbf{F}_{\text{SR}}(\mathbf{r}) \quad (\text{IV.15})$$

obviously decays exponentially fast. Because of the exponential decay of  $\mathbf{F}_{\text{SR}}$ , the sole contributions in the three-body short-range term

$$\mathbf{R}_{\text{SR}}^{(3)}(\mathbf{r}) = 2\beta q^2 \int d\mathbf{r}' H_{\text{cd}}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \mathbf{F}_{\text{SR}}(\mathbf{r} - \mathbf{r}') \quad (\text{IV.16})$$

which might decay slower than an exponential arising from the region where  $\mathbf{r}'$  is close to  $\mathbf{r}$ . However, because of **EDS2**,  $H_{\text{cd}}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}')$  decays exponentially fast for such configurations. Hence,  $\mathbf{R}_{\text{SR}}^{(3)}(\mathbf{r})$  also decays exponentially fast.

In a second step, we study the three terms which involve the Coulomb force  $\mathbf{F}_C$ . The mean-field term

$$\mathbf{R}_{\text{MF}}(\mathbf{r}) = 2\beta q^2 \rho \int d\mathbf{r}' S(\mathbf{r}') \mathbf{F}_C(\mathbf{r} - \mathbf{r}') \quad (\text{IV.17})$$

decays exponentially fast, by virtue of the charge sum rule (IV.2) and of the rotational invariance of  $S(\mathbf{r}) = S(r)$ . The direct Coulomb term

$$\mathbf{R}_C(\mathbf{r}) = \beta q^4 N(\mathbf{r}) \mathbf{F}_C(\mathbf{r}) \quad (\text{IV.18})$$

decays algebraically, namely

$$\mathbf{R}_C(\mathbf{r}) \sim 4\beta q^4 \rho^2 \frac{A}{r^{3+\eta}} \hat{\mathbf{r}} \quad , \quad r \rightarrow \infty \quad (\text{IV.19})$$

with  $\hat{\mathbf{r}} = \mathbf{r}/r$ , discarding exponentially fast decaying corrections. In the three-body Coulomb term

$$\mathbf{R}_C^{(3)}(\mathbf{r}) = 2\beta q^4 \int d\mathbf{r}' H_{\text{cc}}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \mathbf{F}_C(\mathbf{r} - \mathbf{r}') \quad , \quad (\text{IV.20})$$

there are exponentially decaying contributions from the region  $\mathbf{r}'$  close to  $\mathbf{r}$  as a consequence of **EDS3**. However, there are algebraic contributions from the region  $\mathbf{r}'$  close to the origin  $\mathbf{0}$  which arise from the large-distance behavior

$$H_{\text{cc}}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \sim 2 \frac{[A_{++}(\mathbf{0}, \mathbf{r}') - A_{+-}(\mathbf{0}, \mathbf{r}')] }{|\mathbf{r} - \mathbf{r}'/2|^{1+\eta}} \quad \text{when } r \rightarrow \infty \quad \text{with } \mathbf{r}' \text{ fixed} \quad , \quad (\text{IV.21})$$

discarding exponentially decaying terms. Hence, we find

$$\mathbf{R}_C^{(3)}(\mathbf{r}) \sim 4\beta q^4 \rho^3 \int d\mathbf{r}' \frac{[A_{++}(\mathbf{0}, \mathbf{r}') - A_{+-}(\mathbf{0}, \mathbf{r}')] }{|\mathbf{r} - \mathbf{r}'/2|^{1+\eta}} \mathbf{F}_C(\mathbf{r} - \mathbf{r}') \quad , \quad (\text{IV.22})$$

discarding exponentially decaying terms.

The previous analysis shows that all terms in the BGY equation (III.12) decay exponentially fast, except the sum  $[\mathbf{R}_C(\mathbf{r}) + \mathbf{R}_C^{(3)}(\mathbf{r})]$  which, according to (IV.19) and (IV.22) provides the algebraic contribution

$$4\beta q^4 \rho^2 \int d\mathbf{r}' \frac{[\rho A_{++}(\mathbf{0}, \mathbf{r}') - \rho A_{+-}(\mathbf{0}, \mathbf{r}') + A\delta(\mathbf{r}')] }{|\mathbf{r} - \mathbf{r}'/2|^{1+\eta}} \mathbf{F}_C(\mathbf{r} - \mathbf{r}') \quad . \quad (\text{IV.23})$$

At large distances  $r$ , its asymptotic representation in power series of  $1/r$  is generated by the expansion of  $\mathbf{F}_C(\mathbf{r} - \mathbf{r}')/|\mathbf{r} - \mathbf{r}'/2|^{1+\eta}$  in Taylor series with respect to  $\mathbf{r}'$ . The *a priori* leading term of order  $1/r^{3+\eta}$  has a vanishing amplitude by virtue of the sum rule (IV.12). The amplitude of the next term of order  $1/r^{4+\eta}$  is proportional to the first moment of  $[\rho A_{++}(\mathbf{0}, \mathbf{r}') - \rho A_{+-}(\mathbf{0}, \mathbf{r}') + A\delta(\mathbf{r}')]$  and it also vanishes because of rotational invariance. The first *a priori* non-vanishing term is of order  $1/r^{5+\eta}$  and its amplitude is proportional to the second moment of  $[\rho A_{++}(\mathbf{0}, \mathbf{r}') - \rho A_{+-}(\mathbf{0}, \mathbf{r}') + A\delta(\mathbf{r}')]$ . Hence the sum  $[\mathbf{R}_C(\mathbf{r}) + \mathbf{R}_C^{(3)}(\mathbf{r})]$  decays algebraically, namely

$$\mathbf{R}_C(\mathbf{r}) + \mathbf{R}_C^{(3)}(\mathbf{r}) = \frac{(1+\eta)(8+\eta)\beta q^4 \rho^2 M_2}{6 r^{5+\eta}} \hat{\mathbf{r}} + o(1/r^{5+\eta}) \quad , \quad r \rightarrow \infty \quad (\text{IV.24})$$

with the second moment

$$M_2 = \rho \int d\mathbf{x} x^2 [A_{++}(x) - A_{+-}(x)] \quad . \quad (\text{IV.25})$$

If  $M_2$  vanishes, one has to pursue the asymptotic large- $r$  expansion of (IV.23) to next orders. The amplitude of the term of order  $1/r^{2n+\eta}$  with  $n \geq 2$  is proportional to the  $2n$ -th moment  $M_{2n}$  of  $[A_{++}(x) - A_{+-}(x)]$ . The leading behavior is obtained for the first non-vanishing moment  $M_{2n}$ . Such moment necessarily exists since otherwise  $\rho[A_{++}(x) - A_{+-}(x)]$  would reduce to  $-A\delta(\mathbf{x})$ , in contradiction with the physical expectation that  $\rho[A_{++}(x) - A_{+-}(x)]$  is a smooth function of  $x$ . Hence, in any case,  $[\mathbf{R}_C(\mathbf{r}) + \mathbf{R}_C^{(3)}(\mathbf{r})]$  indeed decays algebraically.

Since all the other terms than  $[\mathbf{R}_C(\mathbf{r}) + \mathbf{R}_C^{(3)}(\mathbf{r})]$  in (III.12) decay exponentially fast, we conclude that the assumed **EDS** is not consistent with the BGY hierarchy. Thus, at the critical point, the charge correlations cannot decay exponentially fast, and they are polluted by the algebraic tails in the density-density correlations. The infection mechanism arises from the contributions of three-body correlations, where the critical  $1/r^{1+\eta}$ -tails are coupled to the Coulomb  $1/r^2$ -force: the resulting effective  $1/r^{3+\eta}$ -force created by a spherically symmetric cloud decays algebraically at large distances as far as this cloud displays a finite spatial extension.

## V. A PLAUSIBLE SCENARIO

The infection mechanism described in the previous Section leads to an algebraic decay of charge correlations. In Section V A, we propose an algebraic-decay scenario



(**ADS**), which is consistent with the BGY hierarchy, contrary to the **EDS**. A few concluding comments are given in Section V B.

### A. Power-law decay of charge correlations

Now we assume that  $S(r)$  decays as  $1/r^s$  with  $s > 3$ . Similarly to the **EDS**, we also assume that the same power-law  $1/r^s$  controls the decay of the combinations of three-body correlations for configurations where the  $1/r^{1+\eta}$ -critical tails cancel out. Accordingly, the corresponding algebraic-decay scenario reads

- **ADS1:**  $S(r)$  decays as  $1/r^s$  when  $r \rightarrow \infty$

- **ADS2:**  $H_{\text{cd}}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}')$  behaves as

$$H_{\text{cd}}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \sim \frac{[B_{++}(\mathbf{0}, \mathbf{r}') + B_{+-}(\mathbf{0}, \mathbf{r}')] }{|\mathbf{r} - \mathbf{r}'/2|^s} \quad \text{when } r \rightarrow \infty \quad \text{with } \mathbf{r}' \text{ fixed ,} \quad (\text{V.1})$$

$$H_{\text{cd}}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \sim \frac{[B_{++}(\mathbf{r}, \mathbf{r}') + B_{+-}(\mathbf{r}, \mathbf{r}')] }{|(\mathbf{r} + \mathbf{r}')/2|^s} \quad \text{when } r \rightarrow \infty \quad \text{with } (\mathbf{r}' - \mathbf{r}) \text{ fixed .} \quad (\text{V.2})$$

- **ADS3:**  $H_{\text{cc}}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}')$  behaves as

$$H_{\text{cc}}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}') \sim \frac{[B_{++}(\mathbf{r}, \mathbf{r}') - B_{+-}(\mathbf{r}, \mathbf{r}')] }{|(\mathbf{r} + \mathbf{r}')/2|^s} \quad \text{when } r \rightarrow \infty \quad \text{with } (\mathbf{r}' - \mathbf{r}) \text{ fixed .} \quad (\text{V.3})$$

- **ADS4:**  $[A_{++}(x) - A_{+-}(x)]$  and  $[B_{++}(x) - B_{+-}(x)]$  decay as  $1/x^s$  when  $x \rightarrow \infty$

We show that such algebraic decays are consistent with the BGY equations. Furthermore the corresponding analysis provides  $s = 6 + \eta$ .

Let us consider the BGY equation (III.12) for  $S(r)$ . In the l.h.s.  $\nabla S(\mathbf{r})$  decays exponentially as  $1/r^{s+1}$  by virtue of **EDS1**. In the r.h.s., the direct short-range term  $\mathbf{R}_{\text{SR}}(\mathbf{r})$  decays exponentially fast because of the exponential decay of  $\mathbf{F}_{\text{SR}}$ . In the three-body short-range term  $\mathbf{R}_{\text{SR}}^{(3)}(\mathbf{r})$ , the contributions of the region  $\mathbf{r}'$  close to the origin also decay exponentially fast for the same reason. According to (V.2), algebraically decaying contributions arise from the region where  $\mathbf{r}'$  is close to  $\mathbf{r}$ . The corresponding power series of  $1/r$  are generated by the expansion of  $\mathbf{F}_{\text{SR}}(\mathbf{r} - \mathbf{r}')/|(\mathbf{r} +$

$\mathbf{r}')/2|^s$  in Taylor series with respect to  $\mathbf{r}'$ . Combining the rotational invariance of the amplitude functions,  $B_{++}(\mathbf{r}, \mathbf{r}') = B_{++}(|\mathbf{r} - \mathbf{r}'|)$  and  $B_{+-}(\mathbf{r}, \mathbf{r}') = B_{+-}(|\mathbf{r} - \mathbf{r}'|)$ , with the antisymmetry of  $\mathbf{F}_{\text{SR}}$ , we see that the first non-vanishing term decays as  $1/r^{s+1}$ , and so does  $\mathbf{R}_{\text{SR}}^{(3)}(\mathbf{r})$ .

Since

$$\nabla \cdot \mathbf{R}_{\text{MF}}(\mathbf{r}) = -8\pi\beta q^2 S(r) \quad (\text{V.4})$$

as a consequence of Poisson's equation, the mean-field term  $\mathbf{R}_{\text{MF}}(\mathbf{r})$  decays as  $1/r^{s-1}$ . The direct Coulomb term  $\mathbf{R}_{\text{C}}(\mathbf{r})$  again decays as (IV.19) discarding terms of order  $1/r^{s+2}$ . In the three-body Coulomb term  $\mathbf{R}_{\text{C}}^{(3)}(\mathbf{r})$ , the contribution of the region  $\mathbf{r}'$  close to the origin again behaves as (IV.22) discarding terms of order  $1/r^{s+2}$ . The contribution of the region  $\mathbf{r}'$  close to  $\mathbf{r}$  can be determined as that arising in  $\mathbf{R}_{\text{SR}}^{(3)}(\mathbf{r})$  by using (V.3) and is found to decay as  $1/r^{s+1}$ .

In conclusion, the two slowest-decaying contributions in (III.12) arise from the mean-field term  $\mathbf{R}_{\text{MF}}(\mathbf{r}) \sim \text{cst}/r^{s-1}$  on the one hand, and from the combination  $[\mathbf{R}_{\text{C}}(\mathbf{r}) + \mathbf{R}_{\text{C}}^{(3)}(\mathbf{r})] \sim \text{cst}/r^{5+\eta}$  on the other hand. Thus, this BGY equation for  $S(r)$  can be satisfied if and only if the two previous powers are identical, *i.e.*  $s = 6 + \eta$ .

It can be checked that **ADS** is also consistent with the large-distance behavior of the BGY equation (III.11) for  $N(r)$ . Indeed, the l.h.s. of (III.11) decays as  $1/r^{2+\eta}$ , and in the r.h.s. there are various terms which also decay as  $1/r^{2+\eta}$ , namely the mean-field term and the two three-body terms. Note that the corresponding  $1/r^{2+\eta}$ -decays are obtained by combining the antisymmetry of the forces  $\mathbf{F}_{\text{SR}}$  and  $\mathbf{F}_{\text{C}}$  with the asymptotic behavior of  $N(r')$ ,  $H_{\text{dd}}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}')$  and  $H_{\text{dc}}^{(3)}(\mathbf{0}, \mathbf{r}, \mathbf{r}')$  when  $r \rightarrow \infty$  with  $(\mathbf{r}' - \mathbf{r})$  fixed. Thus, the algebraic-decay scenario (**ADS**) is fully consistent with the large-distance behavior of the BGY hierarchy, provided that  $s = 6 + \eta$ .

## B. Concluding comments

The key ingredients of our derivations are plausible *a priori* assumptions on the decay of three-body Ursell functions. In fact, such three-body correlations can be represented by diagrammatic series where the bonds are the two-body Ursell

functions (see *e.g.* [22]). The algebraic decays of the  $h^{(3)}$ 's are then related to those of the  $h$ 's. This leads to replace the crude assumptions (IV.6) and (IV.7), by asymptotic expansions in inverse power of  $r$ . However, their structures are identical to those derived from (IV.6) and (IV.7). Hence, the infection mechanism found in Section IV B still holds, with a three-body contribution in (III.12) which again decays as  $1/r^{5+\eta}$ . Including the refined three-body decays in the analysis of Section V A, one finds that  $S(r)$  indeed decays as  $1/r^{6+\eta}$  when  $r \rightarrow \infty$ . Furthermore, it turns out that such decay has been independently obtained in [23] within a completely different approach. Accordingly, the  $1/r^{6+\eta}$ -decay of  $S(r)$  appears as a quite robust prediction, despite it is not yet rigorously established.

Since  $0 < \eta < 1$ ,  $S(r)$  decays faster than  $1/r^6$  and slower than  $1/r^8$ . Hence the second moment of  $S(r)$  is finite, while its fourth moment diverges. This is in agreement with the numerical results obtained within sophisticated Monte Carlo simulations [7]. These simulations also indicate that the Stillinger-Lovett second moment sum rule for  $S(r)$  is violated at the critical point. In fact, the slow decays of three- and four-body correlations present in the algebraic-decay scenario should prevent the Stillinger-Lovett sum rule to be satisfied, as strongly suggested by a conditional theorem [20]. The present analysis turns then to be also consistent with the simulation prediction concerning the dielectric nature of the critical point.

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