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Correlations in the Kosterlitz–Thouless Phase of the Two-Dimensional Coulomb Gas

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The particle and charge correlations of the two-dimensional Coulomb gas are studied in the dielectric phase. A term-by-term analysis of the low-fugacity expansions suggests that the large-distance behaviors of the particle correlations are governed by multipolar interactions, similar to what happens in a system of permanent dipoles. These behaviors are compatible with the asymptotic structure of the BGY hierarchy equations; on the other hand, a new identity for the dielectric constant ε is used to show that the four-particle correlations decay as the dipole–dipole potential $1/r^2$ when two neutral pairs are separated by a large distance r. Near the zero-density critical point of the Kosterlitz–Thouless transition, we resum the low-fugacity expansions of both $1/\varepsilon$ and the charge correlation $C(r)$. We thus retrieve the coupling constant flow equations of the renormalization group as well as the effective interaction energy of the iterated mean-field theory by Kosterlitz and Thouless. The coupling constant at the RG fixed point is then identified with $1/\varepsilon$. The nonanalyticity of $1/\varepsilon$ at the transition turns out to coincide with the divergence of the low-fugacity series for this quantity. The leading term in the large-distance behavior of $C(r)$ is found to be the same as for external charges. Moreover, we exhibit the subleading terms which also contribute to $1/\varepsilon$.

KEY WORDS: Kosterlitz–Thouless transition; Coulomb gas; dielectric phase; correlations; fugacity expansions; BGY hierarchy.

1. INTRODUCTION

The two-dimensional Coulomb gas (CG) is a neutral system made up of two species of charges interacting via a logarithmic potential. In this model (Section 2), the attraction between oppositely charged particles competes

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with the thermal motion. If the particles are point charges[1] ($\pm e$), the attraction at short distances makes the system collapse for coupling constants $\Gamma \geqslant 2$ ($\Gamma = \beta e^2$ and β is the inverse temperature). On the contrary, if the collapse is avoided by some short-range repulsion, the model remains well-behaved for any temperature. At high temperatures, the system is in a conductive phase: the correlations decay exponentially at large distance.[3] At low temperatures, $\Gamma \geqslant 4$, the system undergoes the so-called Kosterlitz–Thouless (KT) transition[7] to a dielectric phase, where the long-range logarithmic potential binds opposite charges in pairs (in this phase, the free energy variation associated with the creation of a macroscopic pair is positive). In the conductive phase the external charges are perfectly screened and the dielectric constant ε is infinite, whereas in the dielectric phase the screening is only partial[4] and ε is finite.

The above binding mechanism was devised by Kosterlitz and Thouless in order to explain a special class of two-dimensional transitions. Indeed, in two-dimensional systems with local interactions and continuous symmetry, there is no spontaneous symmetry breaking.[9] However, high- and low-temperature expansions show that in some systems (XY model, defects in pseudosolids, superfluid helium films,...), the correlations change from an exponential decay to an algebraic behavior at large distances. This transition from a disordered phase to a quasi-ordered phase is due to the condensation of topological excitations interacting via a logarithmic potential. The corresponding universality class is characterized by exponents which continuously vary with some microscopic parameters. More recently, generalized Coulomb gases made up of both electrical and magnetic charges have been introduced[10]; for such systems, the critical exponents on the transition line can be exactly calculated through the renormalization group (RG) methods. This approach is complementary to the results of the conformally invariant theory.[11] In this paper, we are interested in the particle and charge correlations of the CG in the dielectric phase, which are not studied in the previous methods, nor in the field-theory methods applied to the sine-Gordon representation of the CG.[12]

In the literature, the intrinsic correlations of the CG are dealt with only in the iterated mean-field theory, introduced by Kosterlitz and

[3] For a rigorous proof at sufficiently high temperatures, see Yang[2]; this work extends the analysis of Brydges and Federbush[3] and Imbrie[4] to two-dimensional Coulomb systems. A lattice version of the CG has been exactly solved at $\Gamma = 2$ by Gaudin[5]; the corresponding correlations decay exponentially and this system is indeed in a conductive phase, as shown by Cornu and Jancovici.[6]

[4] In refs. 8 it is rigorously shown that, at sufficiently low temperatures and small activities, the correlations between external noninteger charges are bounded by an inverse power law and go to zero at large distances.

Thouless[7] in order to describe the transition at finite density. In this theory, the two charges of a given neutral pair polarize the smaller pairs which are located between them. Then the effective energy of the pair is equal to the interaction potential in the vacuum renormalized by an effective dielectric constant $\varepsilon(r)$ which depends on the size r of the pair. The intrinsic charge correlation $C(r)$ is identified with an effective pair correlation proportional to the Boltzmann factor associated with the above effective energy. Thus, in the framework of this heuristic approach, this charge correlation in the dielectric phase would decay as $1/r^{\Gamma/\varepsilon}$, where ε is the macroscopic dielectric constant of the medium [ε appears as the limit of $\varepsilon(r)$ when $r \to \infty$]. This result is not *a priori* obvious at all. First, $C(r)$ is the difference between the particle correlations $\rho^T_{++}(r)$ and $\rho^T_{+-}(r)$; $\rho^T_{++}(r)$ [$\rho^T_{+-}(r)$] denotes the correlation between charges with the same (opposite) sign. The point is that these particle correlations are expected to decay as $1/r^4$, similar to what happens in a system of permanent dipoles.[13] Second, even if compensations of the $1/r^4$ terms occur, the intrinsic charge correlation does not usually behave as the correlation between infinitesimal external charges q_α and q_γ which, by definition of ε, decays as $\exp[-\beta(q_\alpha q_\gamma/\varepsilon)\ln r] = 1/r^{(\beta q_\alpha q_\gamma/\varepsilon)}$.

In this paper, we study the large-distance behaviors of the particle correlations in the dielectric phase by both an analysis of the low-fugacity expansions (Section 3) and a survey of the BGY hierarchy equations at finite densities (Section 5). The term-by-term analysis indicates that these behaviors are governed by multipolar interactions. In particular, $\rho^T_{++}(r)$ and $\rho^T_{+-}(r)$ indeed decay as $1/r^4$, i.e., as the square of the dipole–dipole potential. These perturbative results are shown to be compatible with the asymptotic structure of the BGY equations. More precisely, we first derive a new exact expression of $1/\varepsilon$ in terms of the dipole associated with the cloud surrounding two opposite charges of the medium. Then, this identity is used to show that the four-particle correlation ρ^T_{-+-+} decays as $1/r^2$ when two neutral pairs are separated by a large distance r.

Another part of this paper (Section 4) is devoted to a survey of both $1/\varepsilon$ and the intrinsic charge correlation $C(r)$ in the regime where both the fugacity z and $\Gamma-4$ are small, i.e., near the critical point at zero density ($\Gamma=4$). First, according to the survey in Section 3, the multipolar $1/r^4$ contributions cancel out in $C(r)$. Our analysis consists in the simultaneous resummations of the low-fugacity expansions of $C(r)$ and $1/\varepsilon$, which is related to the second moment of $C(r)$ via the linear response theory. In the considered regime, we have to retain in the expansion for $1/\varepsilon$ all the terms $z^{2N}/(\Gamma-4)^{2N-1}$ which are of the same order. These terms arise from the large-distance behavior of $C(r)$. We stress that both leading and subleading parts of the asymptotic expansion of $C(r)$ do contribute. The study of $C(r)$

at the order z^4 exhibits the partial screening of a given pair by a smaller pair which behaves as a polarizable dipole. Then, the generalization of this mechanism of nested pairs gives a recurrence scheme for handling the higher-order terms. This leads to a system of coupled differential equations for $C(r)$ and the above quantity $1/\varepsilon(r)$, which comes out in a very natural way. In fact, this system proves to be equivalent to the coupling constant flow equations of the RG[14,15]; thus, the coupling constant at the fixed point is to be identified with Γ/ε, as already suggested in the literature. Moreover, the solution $C(r)$ of our resummation equations turns out to have the same structure as the effective pair correlation postulated by Kosterlitz and Thouless. Hence, the basic idea of these authors is supported by the present analysis starting from first principles, at least near the zero-density critical point. On the other hand, the asymptotic behavior of $C(r)$ takes the form $A(r)/r^{\Gamma/\varepsilon}$, where $A(r)$ goes to a constant; this ensures that $C(r)$ indeed behaves as the correlation between external charges. However, $A(r)$ is an infinite sum of terms $1/r^{N(\Gamma/\varepsilon-4)}$ ($N \geqslant 0$) which all contribute to $1/\varepsilon$. Finally, in our approach, the signal of the transition directly appears as the nonanalyticity of $1/\varepsilon$, which happens to coincide with the divergence of the low-fugacity series of this quantity.

2. DESCRIPTION OF THE MODEL

The two-dimensional Coulomb gas (CG) is a neutral system made up of two species of particles with the same mass but opposite charges $\pm e$ ($e > 0$), which move in a plane surface and interact via the pairwise two-dimensional Coulomb potential $v_C(r)$ (r is the distance between the interacting particles). This potential is defined as the solution of the two-dimensional Poisson equation

$$\nabla^2 v_C(r) + 2\pi\delta(\mathbf{r}) = 0 \tag{2.1}$$

and it takes the logarithmic form

$$v_C(r) = -\ln\left(\frac{r}{L}\right) \tag{2.2}$$

(L is an irrelevant length scale which fixes the origin of the potential). The attraction between oppositely charged particles competes with the thermal motion and gives rise to two kinds of phenomena.

In the first place the point particle model introduced by Hauge and Hemmer[1] is unstable for the temperatures lower than $T_u = e^2/2k_B$, because at short distances the attraction between oppositely charged particles makes the system collapse. In order to prevent this collapse, we introduce a sufficiently repulsive short-range pairwise interaction $w^{\rm SR}_{+-}$ between these particles. Thus the basic two-body potentials are (with obvious notations)

$$\begin{aligned} v_{++}(r) &= e^2 v_{\rm C}(r) + w^{\rm SR}_{++}(r) \\ v_{+-}(r) &= -e^2 v_{\rm C}(r) + w^{\rm SR}_{+-}(r) \\ v_{--}(r) &= e^2 v_{\rm C}(r) + w^{\rm SR}_{--}(r) \end{aligned} \tag{2.3}$$

The short-range interactions $w^{\rm SR}_{++}(r)$ and $w^{\rm SR}_{--}(r)$ are not essential for the stability of the system and may be omitted. Moreover, as far as long-range effects are concerned, the precise form of the short-range potentials is not crucial; so, for technical reasons, we shall consider various kinds of $w^{\rm SR}_{++}(r)$, $w^{\rm SR}_{--}(r)$, and $w^{\rm SR}_{+-}(r)$ in the following sections. For instance, the use of a hard-disk model is most appropriate for the resummation of the low-fugacity expansions (see Section 4). On the contrary, in order to handle the BGY hierarchy (see Section 5), it is convenient to define the short-range potentials in such a way that the whole potentials v_{++}, v_{+-}, and v_{--} are differentiable everywhere. Thus, the finite CG, with $2N$ particles moving in a surface of area A and interacting via the potentials given by (2.3), has a well-behaved thermodynamic limit, whatever the values of the temperature T and the density of each species $\rho \equiv \rho_+ = \rho_- = N/A$ may be.[16]

Second, for the temperatures lower than T_u, the long range of the logarithmic Coulomb attraction between positive and negative charges binds them in pairs with a finite polarizability; for fixed small values of the density, the infinite system undergoes a Kosterlitz–Thouless (KT) transition between a high-temperature conductive phase and a low-temperature dielectric phase. The transition is not signaled by thermodynamic singularities, but by the discontinuity of the inverse of the dielectric constant ε which characterizes the electrical properties of the system. The present dielectric constant ε is defined through the linear response of the infinite system to an infinitesimal external charge distribution $q_{\rm ext}$. When the external charge distribution $q_{\rm ext}(\mathbf{r})$ is immersed into the infinite fluid, there appears a total charge distribution $\delta q_{\rm tot}(\mathbf{r})$, which is the sum of $q_{\rm ext}(\mathbf{r})$ and of the corresponding charge density induced in the system. In the linear regime, the Fourier transforms $q_{\rm ext}(\mathbf{k})$ and $\delta q_{\rm tot}(\mathbf{k})$ are proportional to each other,

$$\delta q_{\rm tot}(\mathbf{k}) = \frac{1}{\varepsilon(\mathbf{k})}\, q_{\rm ext}(\mathbf{k}) \tag{2.4}$$

and ε is defined through the response function $\varepsilon(\mathbf{k})$ in the limit of a uniform external distribution (that is, an extended distribution with spatial variations which are very slow with respect to the mean distance between the particles of the system),

$$\varepsilon \equiv \lim_{k \to 0} \varepsilon(\mathbf{k}) \tag{2.5}$$

In a conductive phase, the infinitesimal uniform external charge is perfectly screened, $\delta q_{\text{tot}}(\mathbf{k}) = 0$ and then ε is infinite, whereas in the dielectric phase, the screening is only partial, $\delta q_{\text{tot}}(\mathbf{k}) \neq 0$ and ε is finite.

Using the linear response theory, one easily relates ε to the internal charge–charge correlation $C(r)$ of the infinite system,

$$C(r) \equiv \lim_{\text{TL}} \left\langle \left[e \sum_{i=1}^{N} \delta(\mathbf{x}_i - \mathbf{0}) - e \sum_{i=1}^{N} \delta(\mathbf{y}_i - \mathbf{0}) \right] \times \left[e \sum_{j=1}^{N} \delta(\mathbf{x}_j - \mathbf{r}) - e \sum_{j=1}^{N} \delta(\mathbf{y}_j - \mathbf{r}) \right] \right\rangle \tag{2.6}$$

[In (2.6) TL means "thermodynamic limit" and $\langle \cdots \rangle$ denotes the thermal equilibrium average; the positions of the charges with positive (negative) sign is denoted by $\mathbf{x}_i$ ($\mathbf{y}_i$).] The relation is

$$\frac{1}{\varepsilon(\mathbf{k})} = 1 - \beta v_c(\mathbf{k})\, C(\mathbf{k}) \tag{2.7}$$

so that

$$\frac{1}{\varepsilon} = 1 + \frac{\pi\beta}{2} \int d\mathbf{r}\, r^2 C(r) \tag{2.8}$$

Notice that there exists another definition for ε, which proves to be equivalent to the previous one. In this definition the linear response theory is used to calculate the effective potential $v_{\alpha\beta}^{\text{eff}}$ between two infinitesimal point charges q_α and q_β immersed in the infinite system. This potential is defined through the first-order difference between the free energy of the fluid with both charges in it and the sum of the free energies associated with the creation of the polarization clouds when the charges are immersed separately into the system. According to the linear response theory, one finds

$$v_{\alpha\beta}^{\text{eff}}(\mathbf{k}) = q_\alpha q_\beta \frac{v_c(\mathbf{k})}{\varepsilon(\mathbf{k})} \tag{2.9}$$

where we have used (2.7) to make $1/\varepsilon(\mathbf{k})$ appear. In the dielectric phase, $1/\varepsilon$ is finite and the linearly screened effective potential becomes proportional to the bare Coulomb interaction at large distances. On the contrary, in the conductive phase, $1/\varepsilon(\mathbf{k})$ goes to zero as $\mathbf{k}$ vanishes and the large-distance behavior of the linearly screened potential differs from the Coulomb law; both localized and extended infinitesimal external charge distributions are perfectly screened in this phase.

According to (2.8), the KT transition is also characterized by a qualitative change in the behavior of the correlations of the CG. In the conductive phase $C(r)$ obeys the so-called Stillinger–Lovett sum rule[17]

$$\int d\mathbf{r}\, r^2 C(r) = -\frac{2}{\pi\beta} \tag{2.10}$$

which is a direct consequence of (2.8) with $\varepsilon = \infty$. This sum rule no longer holds in the dielectric phase. According to the analysis by Martin and Gruber,[18] this breakdown of the Stillinger–Lovett sum rule implies that some particle correlations decay algebraically at large distances in this phase. Indeed, by using the BGY equations, these authors have shown that, if the two-body correlations decay faster than $1/r^4$ and if the three- and four-body correlations decay faster than $1/r^3$, then the sum rule (2.10) is satisfied (these conditions are fulfilled in the conductive phase where all the correlations are expected to decay exponentially). The particle correlations of interest, $\rho^T_{s_1 \cdots s_n}(\mathbf{r}_1,..., \mathbf{r}_n)$, are defined as

$$\rho^T_{s_1 \cdots s_n}(\mathbf{r}_1,..., \mathbf{r}_n) = \lim_{\mathrm{TL}} \left\langle \left[\prod_{i=1}^{n} \left(\delta_{+,s_i} \sum_{j=1}^{N} \delta(\mathbf{x}_j - \mathbf{r}_i) + \delta_{-,s_i} \sum_{j=1}^{N} \delta(\mathbf{y}_j - \mathbf{r}_i) \right) \right]_{\mathrm{nc}} \right\rangle^T \tag{2.11}$$

where s_i denotes the sign of the considered species, the superscript T indicates a full truncation with respect to all the partitions of $\{\mathbf{r}_1,..., \mathbf{r}_n\}$, and the notation $[\cdots]_{\mathrm{nc}}$ means that only the contributions of points which do not coincide must be retained. These particle correlations are related to the usual Ursell functions by

$$\rho^T_{s_1 \cdots s_n}(\mathbf{r}_1,..., \mathbf{r}_n) = \rho^n h_{s_1 \cdots s_n}(\mathbf{r}_1,..., \mathbf{r}_n) \tag{2.12}$$

Notice that the charge–charge correlation $C(r)$ can be expressed in terms of the particle correlation as

$$C(r) = 2e^2 \rho \delta(\mathbf{r}) + e^2[\rho^T_{++}(\mathbf{0}, \mathbf{r}) + \rho^T_{--}(\mathbf{0}, \mathbf{r}) - 2\rho^T_{+-}(\mathbf{0}, \mathbf{r})] \tag{2.13}$$

3. ANALYSIS OF THE LOW-FUGACITY EXPANSIONS

In this section, we consider a Coulomb gas made up of charged hard disks with a diameter σ. Then the short-range potentials in (2.3) reduce to

$$w^{SR}_{++}(r) = w^{SR}_{--}(r) = w^{SR}_{+-}(r) = \begin{cases} \infty, & r < \sigma \\ 0, & \sigma < r \end{cases} \tag{3.1}$$

Furthermore, for the sake of simplicity, we shall take $L = \sigma$. For the present symmetric version of the CG,

$$\rho^T_{++}(r) = \rho^T_{--}(r) \tag{3.2}$$

and similar identities hold for correlations between more than two particles. The charge–charge correlation given by (2.13) can then be rewritten as

$$C(r) = 2e^2\rho\delta(\mathbf{r}) + 2e^2[\rho^T_{++}(\mathbf{0}, \mathbf{r}) - \rho^T_{+-}(\mathbf{0}, \mathbf{r})] \tag{3.3}$$

The large-distance behavior of the particle and charge correlations will be studied by expanding the formers in powers of the fugacity z. This analysis is allowed in the dielectric phase because all the terms of these expansions are well defined[19] for $\Gamma > 4$. In Section 3.1 we introduce the grand canonical ensemble. In Section 3.2 we show that the particle correlations decay algebraically at each order in the fugacity, and we determine the dominant powers for the two-, three-, and four-body correlations. Some comments are given in Section 3.3.

3.1. Grand Canonical Ensemble

We start from the grand canonical ensemble representation for a finite system which occupies a surface with an area A. We introduce space-dependent dimensionless fugacities $z_+(\mathbf{r})$ and $z_-(\mathbf{r})$ for each species. The corresponding grand partition function Ξ_A is

$$\Xi_A = \sum_{N=0}^{\infty} \frac{1}{(N!)^2} \int_{\mathcal{D}_N} \left[\prod_{i=1}^{N} \frac{d\mathbf{x}_i}{\sigma^2} \frac{d\mathbf{y}_i}{\sigma^2} z_+(\mathbf{x}_i)\, z_-(\mathbf{y}_i) \right] \times \exp[-\beta V_{2N}(\mathbf{x}_1,..., \mathbf{x}_N; \mathbf{y}_1,..., \mathbf{y}_N)] \tag{3.4}$$

where V_{2N} is the electrostatic interaction energy of $2N$ charged hard disks,

$$V_{2N}(\mathbf{x}_1,..., \mathbf{x}_N; \mathbf{y}_1,..., \mathbf{y}_N) = -\frac{e^2}{2} \sum_{i \neq j} \ln\left(\frac{|\mathbf{x}_i - \mathbf{x}_j|}{\sigma}\right) - \frac{e^2}{2} \sum_{i \neq j} \ln\left(\frac{|\mathbf{y}_i - \mathbf{y}_j|}{\sigma}\right) + \frac{e^2}{2} \sum_{i,j} \ln\left(\frac{|\mathbf{x}_i - \mathbf{y}_j|}{\sigma}\right) \tag{3.5}$$

The integration domain $\mathscr{D}_N$ is included in A^{2N} and is such that all the particles are separated by a distance larger than σ. In the definition (3.4) the summation is performed over neutral systems. One could define another grand partition function by summing also the contributions from the non-neutral systems. Both definitions become equivalent in the thermodynamic limit. The particle correlations (2.11) of the homogeneous (neutral) infinite system are given by

$$\rho^T_{s_1 \cdots s_n}(\mathbf{r}_1,..., \mathbf{r}_n) = z^n \lim_{\mathrm{TL}} \frac{\delta^n \ln \Xi_A}{\delta z_{s_1}(\mathbf{r}_1) \cdots \delta z_{s_n}(\mathbf{r}_n)}\Bigg|_{z_+(\mathbf{r}) = z_-(\mathbf{r}) = z} \tag{3.6}$$

In (3.6) the functional derivative is calculated for uniform fugacities $z_+(\mathbf{r}) = z_-(\mathbf{r}) = z$ and the thermodynamic limit is taken at fixed z, β, and $\mathbf{r}_i$ $(i = 1,..., n)$. The equilibrium state of the infinite system is entirely specified by the two dimensionless parameters z and $\Gamma = \beta e^2$.

The expansions of the particle correlations in entire powers of the fugacity z are obtained by using the definition (3.4) in (3.6) and by taking the thermodynamic limit in each term. The expression of the term of order z^{n+p} is an integral over p field points (with the n root points $\mathbf{r}_1,..., \mathbf{r}_n$ kept fixed). In general, these integrals are well defined only for systems with short-range integrable potentials. On the contrary, for three-dimensional Coulomb systems with the familiar $1/r$ potential, all the coefficients of these expansions diverge. However, in two dimensions, the situation is quite different because of the confining character of the logarithmic Coulomb potential. Indeed, Speer[19] has shown that all the above coefficients are finite for $\Gamma > 4$. However, the related integrals are only conditionally convergent, because there are residual dipole–dipole interactions between neutral clusters of charges which are far away from one another (these interactions decay only as $1/r^2$, which is the border power for integrability). The analysis of the way in which the contributions of the boundary regions of the finite system vanish in the thermodynamic limit gives a prescription for handling the contributions from these configurations: one must first perform the angular integration over the orientations of the neutral cluster which is sent to infinity.[19]

3.2. Term-by-Term Analysis

In the dielectric phase Γ is always larger than 4. Consequently the above fugacity expansions can be used for studying the large-distance behavior of the particle correlations. In this subsection, we restrict ourselves to a term-by-term qualitative inspection of such behaviors. This survey is carried out with some detail for the terms with the lowest order in z and the main arguments are briefly sketched for the higher-order terms. We explicitly consider only the two-, three-, and four-body particle correlations.

At the order z^4 included, we have (for $r_{12} \equiv |\mathbf{r}_1 - \mathbf{r}_2| > \sigma$),

$$\rho^T_{++}(\mathbf{r}_1, \mathbf{r}_2) = \frac{z^4}{\sigma^4} \int_{\mathscr{D}^2_\sigma} \frac{d\mathbf{y}_1}{\sigma^2} \frac{d\mathbf{y}_2}{\sigma^2} \left\{ \frac{1}{2} \exp[-\beta V_4(\mathbf{r}_1, \mathbf{r}_2; \mathbf{y}_1, \mathbf{y}_2)] - \exp[-\beta V_2(\mathbf{r}_1; \mathbf{y}_1)] \exp[-\beta V_2(\mathbf{r}_2; \mathbf{y}_2)] \right\} + O(z^6) \tag{3.7}$$

and

$$\rho^T_{+-}(\mathbf{r}_1, \mathbf{r}_2) = \frac{z^2}{\sigma^4} \exp[-\beta V_2(\mathbf{r}_1; \mathbf{r}_2)] + \frac{z^4}{\sigma^4} \int_{\mathscr{D}^2_\sigma} \frac{d\mathbf{x}_1}{\sigma^2} \frac{d\mathbf{y}_1}{\sigma^2} \{\exp[-\beta V_4(\mathbf{r}_1, \mathbf{x}_1; \mathbf{r}_2, \mathbf{y}_1)] - \exp[-\beta V_2(\mathbf{r}_1; \mathbf{r}_2)] \exp[-\beta V_2(\mathbf{x}_1; \mathbf{y}_1)] - \exp[-\beta V_2(\mathbf{r}_1; \mathbf{y}_1)] \exp[-\beta V_2(\mathbf{x}_1; \mathbf{r}_2)]\} + O(z^6) \tag{3.8}$$

The expansion (3.7) begins at the order z^4 only, because there are no neutral configurations involving two identical charges at the order z^2. The integration domain $\mathscr{D}^p_\sigma$ is R^{2p} minus the region corresponding to the configurations where one (or more) relative distances between the field or root points is less than σ. Notice that the integral in the right-hand side of (3.8) is only conditionally convergent and must be calculated according to the general prescription given in Section 3.1.

The large-r_{12} behavior of the z^2 term in (3.8) is obvious, since the latter reduces to $(z^2/\sigma^4)(\sigma/r_{12})^\Gamma$. In order to study the z^4 terms in (3.7) and (3.8), we first remark that the most probable configurations of the field points are such that each of them is close to another field or root point with an opposite charge. These configurations are drawn in Figs. 1a and 1b for ρ^T_{++} and in Figs. 2a and 2b for ρ^T_{+-}. For such configurations, the pairs of opposite charges behave as dipoles, as far as their electrostatic interactions with the other charges are concerned. Their contributions to the asymptotic behaviors of ρ^T_{++} and ρ^T_{+-} are obtained by expanding V_4 in powers of the sizes of the neutral pairs. This method is illustrated below for every configuration.

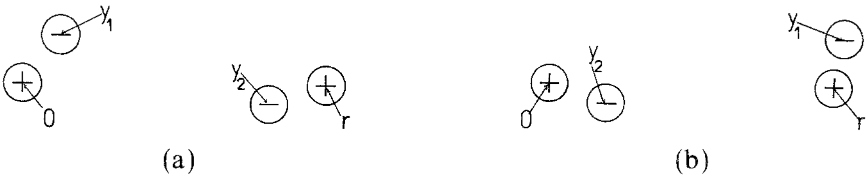

Fig. 1. The most probable configurations of the field points $(\mathbf{y}_1, \mathbf{y}_2)$ which contribute to $\rho^T_{++}(\mathbf{0}, \mathbf{r})$ at the order z^4. A circle with a positive (negative) sign inside represents a hard disk carrying a positive (negative) charge.

3.2.1. Configuration of Fig. 2a. First, V_4 is rewritten as

$$V_4(\mathbf{r}_1, \mathbf{x}_1; \mathbf{r}_2, \mathbf{y}_1) = e^2 \ln t_1 + e^2 \ln t_2 + U(\mathscr{P}_1, \mathscr{P}_2) \tag{3.9}$$

where $\mathbf{t}_1 \equiv \mathbf{r}_1 - \mathbf{y}_1$ and $\mathbf{t}_2 \equiv \mathbf{x}_1 - \mathbf{r}_2$; $\mathscr{P}_1$ ($\mathscr{P}_2$) denotes the neutral pair $\{\oplus\, \mathbf{r}_1; \ominus\, \mathbf{y}_1\}$ ($\{\oplus\, \mathbf{x}_1; \ominus\, \mathbf{r}_2\}$) and $U(\mathscr{P}_1, \mathscr{P}_2)$ is the electrostatic interaction between these pairs. When the pairs 1 and 2 are small compared to their relative distance r_{12}, $U(\mathscr{P}_1, \mathscr{P}_2)$ can be represented by the multipole expansion

$$U(\mathscr{P}_1, \mathscr{P}_2) = e^2 \sum_{n=2}^{\infty} \frac{(-1)^n}{n!} [(\mathbf{t}_1 \cdot \nabla + \mathbf{t}_2 \cdot \nabla)^n - (\mathbf{t}_1 \cdot \nabla)^n - (\mathbf{t}_2 \cdot \nabla)^n] \times \ln\left(\frac{|\mathbf{r}_1 - \mathbf{r}_2|}{\sigma}\right) \tag{3.10a}$$

$$= e^2 (\mathbf{t}_1 \cdot \nabla)(\mathbf{t}_2 \cdot \nabla) \ln\left(\frac{|\mathbf{r}_1 - \mathbf{r}_2|}{\sigma}\right) + O\left(\frac{1}{r_{12}^3}\right) \tag{3.10b}$$

The first term in (3.10b), i.e., the term $n = 2$ in (3.10a), is the usual dipole–dipole interaction potential, which decays as $1/r_{12}^2$ when $r_{12} \to \infty$. In this limit the electrostatic interaction $U(\mathscr{P}_1, \mathscr{P}_2)$ goes to zero, so that the Boltzmann factor $\exp[-\beta U(\mathscr{P}_1, \mathscr{P}_2)]$ can be expanded in a Taylor series

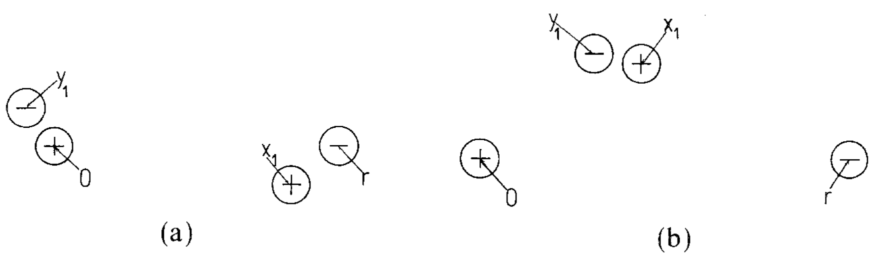

Fig. 2. The same as Fig. 1, for $\rho^T_{+-}(\mathbf{0}, \mathbf{r})$ and the field points $(\mathbf{x}_1, \mathbf{y}_1)$.

with respect to $\beta U(\mathscr{P}_1, \mathscr{P}_2)$. The contribution of the considered configuration to $\rho^T_{+-}(\mathbf{r}_1, \mathbf{r}_2)$ at the order z^4 then becomes, according to (3.8),

$$\frac{z^4}{\sigma^4}\int_{t_1>\sigma}\frac{d\mathbf{t}_1}{\sigma^2}\int_{t_2>\sigma}\frac{d\mathbf{t}_2}{\sigma^2}\left(\frac{\sigma}{t_1}\right)^\Gamma\left(\frac{\sigma}{t_2}\right)^\Gamma\left(-\beta U+\frac{\beta^2U^2}{2!}+\cdots\right) \tag{3.11}$$

where we have taken into account that the zeroth-order term in βU in the expansion of $\exp(-\beta V_4)$ is exactly compensated by the term

$$\exp[-\beta V_2(\mathbf{r}_1;\mathbf{y}_1)]\exp[-\beta V_2(\mathbf{x}_1;\mathbf{r}_2)] = (\sigma/t_1)^\Gamma(\sigma/t_2)^\Gamma$$

Moreover, we have replaced $\mathscr{D}^2_\sigma$ by $\mathscr{D}^1_\sigma\otimes\mathscr{D}^1_\sigma$ and we have omitted the term $\exp[-\beta V_2(\mathbf{r}_1;\mathbf{r}_2)]\exp[-\beta V_2(\mathbf{x}_1;\mathbf{y}_1)]$. Such approximations can be justified *a posteriori*, since they introduce corrections which decay faster than the expression (3.11) [see also Appendix A for a detailed survey of the large-r_{12} behavior of the integrals involved in (3.7) and (3.8)]. The linear term in U does not give any contribution to (3.11), as a consequence of the rotational invariance of the weighting factors $(\sigma/t_1)^\Gamma$ and $(\sigma/t_2)^\Gamma$ combined with the harmonicity of the Coulomb potential. Indeed, once the angular integrations over the orientations of $\mathbf{t}_1$ and $\mathbf{t}_2$ have been performed, the operators $(\mathbf{t}_1\cdot\nabla)^n$ and $(\mathbf{t}_2\cdot\nabla)^n$ either vanish, if n is odd, or become proportional to $(\nabla^2)^{n/2}$, if n is even: in both cases the action of these operators onto $\ln(|\mathbf{r}_1-\mathbf{r}_2|/\sigma)$ gives zero for $r_{12}\neq 0$ by virtue of the Poisson equation (2.1). The first nonvanishing contribution to (3.11) arises from the quadratic term in U. According to (3.10b), the latter reduces to the square of the dipole–dipole potential between the pairs $\mathscr{P}_1$ and $\mathscr{P}_2$ when $r_{12}\to\infty$. After a straightforward integration over the sizes and orientations of the dipoles, we find that the contribution of the configuration in Fig. 2a to $\rho^T_{+-}(\mathbf{r}_1, \mathbf{r}_2)$ behaves as

$$\frac{z^4}{\sigma^4}\frac{\pi^2\Gamma^2}{(\Gamma-4)^2}\left(\frac{\sigma}{r_{12}}\right)^4 + o\left(\frac{1}{r_{12}^4}\right) \tag{3.12}$$

when $r_{12}\to\infty$. In deriving (3.12), we have implicitly used that Γ is larger than 4 when computing the integral

$$\int_{t>\sigma}\frac{d\mathbf{t}}{\sigma^2}\left(\frac{\sigma}{t}\right)^\Gamma t^2 = \frac{2\pi\sigma^2}{\Gamma-4} \tag{3.13}$$

This integral is related to the polarizability of a single pair in the vacuum. When $\Gamma\to 4^+$ this quantity diverges and it is infinite for $\Gamma<4$.

3.2.2. Configurations of Figs. 1a and 1b. These configurations are similar to the configuration of Fig. 2a since they consist of two far away interacting neutral pairs. They give identical contributions to ρ^T_{++} and the sum of these contributions behaves as (3.12).

3.2.3. Configuration of Fig. 2b. In this configuration there is only one neutral pair $\mathscr{P} = \{\oplus\ \mathbf{x}_1;\ \ominus\ \mathbf{y}_1\}$ interacting with the two far away charges $\{\oplus\ \mathbf{r}_1\}$ and $\{\ominus\ \mathbf{r}_2\}$. Using the notations $\mathbf{t} \equiv \mathbf{x}_1 - \mathbf{y}_1$ and $\mathbf{R} \equiv (\mathbf{x}_1 + \mathbf{y}_1)/2$, the part of V_4 which has to be treated perturbatively behaves as

$$e^2(\mathbf{t} \cdot \nabla_{\mathbf{R}}) \left[\ln\left(\frac{|\mathbf{R}-\mathbf{r}_1|}{\sigma}\right) - \ln\left(\frac{|\mathbf{R}-\mathbf{r}_2|}{\sigma}\right) \right] \tag{3.14}$$

for t small compared to $|\mathbf{R}-\mathbf{r}_1|$ and $|\mathbf{R}-\mathbf{r}_2|$. For the same reasons as those exposed in the study of (3.11), the leading behavior of the configuration of Fig. 2b is entirely governed by the quadratic term in the expansion of the Boltzmann factor associated with (3.14). Changing from the variables $(\mathbf{x}_1, \mathbf{y}_1)$ to $(\mathbf{t}, \mathbf{R})$, we find that the leading term in this expansion is

$$\frac{z^4}{\sigma^4}\frac{\Gamma^2}{2}\left(\frac{\sigma}{r_{12}}\right)^{\Gamma} \int_{\mathscr{D}_\sigma^2} \frac{d\mathbf{R}}{\sigma^2}\frac{d\mathbf{t}}{\sigma^2}\left(\frac{\sigma}{t}\right)^{\Gamma} \times \left\{ \mathbf{t} \cdot \nabla_{\mathbf{R}} \left[\ln\left(\frac{|\mathbf{R}-\mathbf{r}_1|}{\sigma}\right) - \ln\left(\frac{|\mathbf{R}-\mathbf{r}_2|}{\sigma}\right) \right] \right\}^2 \tag{3.15}$$

The dominant contributions to the integral in (3.15) arise from the region where $|\mathbf{R}-\mathbf{r}_1|$ and $|\mathbf{R}-\mathbf{r}_2|$ are large (but smaller than r_{12}). Consequently, the leading behavior of (3.15) is not affected by the replacement of $\mathscr{D}_\sigma^2$ by the disentangled conditions $t > \sigma$ and $|\mathbf{R}-\mathbf{r}_i| > \sigma$ ($i = 1, 2$). The latter condition on $\mathbf{R}$ can be viewed as an arbitrary cutoff which prevents spurious divergences associated with the nonintegrability of $[\nabla_{\mathbf{R}} \ln(|\mathbf{R}-\mathbf{r}_i|/\sigma)]^2$ at $\mathbf{R} = \mathbf{r}_i$. With this prescription, we find that (3.15) behaves as[5]

$$\begin{aligned}
&\frac{z^4}{\sigma^4}\frac{\Gamma^2}{2}\left(\frac{\sigma}{r_{12}}\right)^{\Gamma} \int_{|\mathbf{R}-\mathbf{r}_1|, |\mathbf{R}-\mathbf{r}_2| > \sigma} \frac{d\mathbf{R}}{\sigma^2} \int_{t>\sigma} \frac{d\mathbf{t}}{\sigma^2}\left(\frac{\sigma}{t}\right)^{\Gamma} \\
&\qquad \times \left\{ \mathbf{t} \cdot \nabla_{\mathbf{R}} \left[\ln\left(\frac{|\mathbf{R}-\mathbf{r}_1|}{\sigma}\right) - \ln\left(\frac{|\mathbf{R}-\mathbf{r}_2|}{\sigma}\right) \right] \right\}^2 \\
&= \frac{z^4}{\sigma^4}\frac{\pi\Gamma^2}{2(\Gamma-4)}\left(\frac{\sigma}{r_{12}}\right)^{\Gamma} \int_{|\mathbf{R}-\mathbf{r}_1|, |\mathbf{R}-\mathbf{r}_2| > \sigma} d\mathbf{R} \\
&\qquad \times \left\{ \nabla_{\mathbf{R}} \left[\ln\left(\frac{|\mathbf{R}-\mathbf{r}_1|}{\sigma}\right) - \ln\left(\frac{|\mathbf{R}-\mathbf{r}_2|}{\sigma}\right) \right] \right\}^2 \\
&\sim \frac{z^4}{\sigma^4}\frac{2\pi^2\Gamma^2}{\Gamma-4}\left(\frac{\sigma}{r_{12}}\right)^{\Gamma} \ln\left(\frac{r_{12}}{\sigma}\right)
\end{aligned} \tag{3.16}$$

[5] This calculation already appeared in the evaluation of the first nontrivial contribution (in a low-temperature expansion) to the vortex correlation of a classical planar Heisenberg model made by Jose *et al.*[20]

when $r_{12} \to \infty$ [the last line of (3.16) follows from an integration by parts and the use of the Poisson equation (2.1)].

It can be checked that the above configurations indeed give the dominant contributions to the large-r_{12} behavior of $\rho^T_{++}(\mathbf{r}_1, \mathbf{r}_2)$ and $\rho^T_{+-}(\mathbf{r}_1, \mathbf{r}_2)$ at the order z^4. More precisely, we get (with obvious notations)

$$\rho^{T(4)}_{++}(\mathbf{r}_1, \mathbf{r}_2) \sim \rho^{T(4)}_{+-}(\mathbf{r}_1, \mathbf{r}_2) \sim \frac{z^4}{\sigma^4} \frac{\pi^2 \Gamma^2}{(\Gamma - 4)^2} \left(\frac{\sigma}{r_{12}}\right)^4, \qquad r_{12} \to \infty \quad (3.17)$$

The asymptotic behavior (3.17) does not depend on the sign of the charges, because it is entirely determined by the fluctuations of the dipole carried by a root charge and its opposite binding charge. Furthermore, the expression (3.16) is the first correction to the leading term (3.17) of the asymptotic expansion of $\rho^{T(4)}_{+-}(\mathbf{r}_1, \mathbf{r}_2)$; the corresponding correction to $\rho^{T(4)}_{++}(\mathbf{r}_1, \mathbf{r}_2)$ is at least $O(1/r_{12}^{\Gamma})$ and decays faster than (3.16).

The analysis of the terms of order z^{2N}, $\rho^{T(2N)}_{++}(\mathbf{r}_1, \mathbf{r}_2)$ and $\rho^{T(2N)}_{+-}(\mathbf{r}_1, \mathbf{r}_2)$, can be performed through a straightforward generalization of the previous method. The dominant contributions to the large-r_{12} behaviors of these terms[6] arise from the configurations $\mathscr{C}^{\text{neutral}}_{ab}$, where all the particles belong to neutral ensembles $\mathscr{E}^{\text{neutral}}_a$ and $\mathscr{E}^{\text{neutral}}_b$ which contain the particles located at $\mathbf{r}_1$ and $\mathbf{r}_2$, respectively, and whose sizes are small compared to r_{12} (these configurations are similar to those of Figs. 1a, 1b, and 2b). In order to estimate these contributions, we split the total potential V_{2N} as

$$V_{2N} = W_a + W_b + U_{ab} \quad (3.18)$$

where W_a and W_b are the electrostatic energies of $\mathscr{E}^{\text{neutral}}_a$ and $\mathscr{E}^{\text{neutral}}_b$, respectively, and U_{ab} is their mutual interaction potential. Since the distance r_{12} between $\mathscr{E}^{\text{neutral}}_a$ and $\mathscr{E}^{\text{neutral}}_b$ is large compared to their extension, the potential U_{ab} behaves as a dipole–dipole potential, i.e.,

$$U_{ab} \sim (\mathbf{P}_a \cdot \nabla)(\mathbf{P}_b \cdot \nabla) \ln\left(\frac{|\mathbf{r}_1 - \mathbf{r}_2|}{\sigma}\right) \quad (3.19)$$

where $\mathbf{P}_a$ and $\mathbf{P}_b$ are the dipoles of $\mathscr{E}^{\text{neutral}}_a$ and $\mathscr{E}^{\text{neutral}}_b$, respectively. In the large-r_{12} limit, U_{ab} goes to zero and the Boltzmann factor $\exp(-\beta U_{ab})$ can

[6] *A priori*, there are also contributions from configurations $\{\mathscr{E}^{\text{neutral}}_a, \mathscr{E}^{\text{neutral}}_b, \mathscr{E}^{\text{neutral}}_1, ..., \mathscr{E}^{\text{neutral}}_n\}$, where the neutral ensembles $\mathscr{E}^{\text{neutral}}_i$ $(i = 1, ..., n)$ contain only field points and are located between $\mathbf{r}_1$ and $\mathbf{r}_2$. These contributions involve products of dipole–dipole convolution chains connecting $\mathscr{E}^{\text{neutral}}_a$ to $\mathscr{E}^{\text{neutral}}_b$. It turns out that these convolution chains vanish in two dimensions, in contrast to the three-dimensional case, where they behave as the dipole–dipole potential itself, as shown by Høye and Stell.[21]

be expanded in powers of βU_{ab}. The contribution of $\mathscr{C}_{ab}^{\text{neutral}}$ then takes the form (up to numerical multiplicative constants arising from combinatories)

$$z^{2N} \int d\mathscr{E}_a^{\text{neutral}}\, d\mathscr{E}_b^{\text{neutral}} [\exp(-\beta W_a)]^T [\exp(-\beta W_b)]^T \times \left(-\beta U_{ab} + \frac{\beta^2 U_{ab}^2}{2!} + \cdots \right) \tag{3.20}$$

In (3.20), $d\mathscr{E}_a^{\text{neutral}}$ and $d\mathscr{E}_b^{\text{neutral}}$ denote spatial integrations over the positions of the field points belonging to $\mathscr{E}_a^{\text{neutral}}$ and $\mathscr{E}_b^{\text{neutral}}$, respectively; furthermore, the weighting factors $[\exp(-\beta W_a)]^T$ and $[\exp(-\beta W_b)]^T$ are truncated with respect to all the partitions of the truncation processes of $\exp(-\beta V_{2N})$ involved in the integrals which define $\varrho_{++}^{T(2N)}(\mathbf{r}_1, \mathbf{r}_2)$ and $\varrho_{+-}^{T(2N)}(\mathbf{r}_1, \mathbf{r}_2)$. The expression (3.20) has the same structure as (3.11). Using again the rotational invariance of $[\exp(-\beta W_a)]^T$ and $[\exp(-\beta W_b)]^T$ as well as the harmonicity of the Coulomb potential, we find that the linear term in U_{ab} in (3.20) does not give any contribution [as in (3.11)]. The leading term of the asymptotic behavior of (3.20) then arises from the quadratic term in U_{ab}. According to (3.19), it behaves as

$$\frac{\beta^2 z^{2N}}{2} \int d\mathscr{E}_a^{\text{neutral}}\, d\mathscr{E}_b^{\text{neutral}} [\exp(-\beta W_a)]^T [\exp(-\beta W_b)]^T \times \left[(\mathbf{P}_a \cdot \nabla)(\mathbf{P}_b \cdot \nabla) \ln \left(\frac{|\mathbf{r}_1 - \mathbf{r}_2|}{\sigma} \right) \right]^2$$

$$= \frac{\beta^2 z^{2N}}{4} \left\{ \int d\mathscr{E}_a^{\text{neutral}} [\exp(-\beta W_a)]^T P_a^2 \right\} \times \left\{ \int d\mathscr{E}_b^{\text{neutral}} [\exp(-\beta W_b)]^T P_b^2 \right\} \frac{1}{r_{12}^4} \tag{3.21}$$

since the integrals related to the average of P^2 with the weighting factor $[\exp(-\beta W)]^T$ are convergent for $\Gamma > 4$ and for any considered neutral ensemble $\mathscr{E}^{\text{neutral}}$ [as the integral (3.13) in the case where $\mathscr{E}$ reduces to a single pair]. The asymptotic behavior of $\rho_{++}^{T(2N)}(\mathbf{r}_1, \mathbf{r}_2)$ and $\rho_{+-}^{T(2N)}(\mathbf{r}_1, \mathbf{r}_2)$ is obtained by summing all the contributions (3.21). In this summation one must properly take into account all the ways of labeling the field points which belong to $\mathscr{E}_a^{\text{neutral}}$ and $\mathscr{E}_b^{\text{neutral}}$, respectively, as well as the symmetry factors attached to each diagram defining $\rho_{++}^{T(2N)}(\mathbf{r}_1, \mathbf{r}_2)$ and $\rho_{+-}^{T(2N)}(\mathbf{r}_1, \mathbf{r}_2)$. Using also the invariance of $[\exp(-\beta W)]^T P^2$ with respect to the change of sign of all the charges belonging to $\mathscr{E}^{\text{neutral}}$, we find that the total $1/r_{12}^4$ contributions to $\rho_{++}^{T(2N)}(\mathbf{r}_1, \mathbf{r}_2)$ and $\rho_{+-}^{T(2N)}(\mathbf{r}_1, \mathbf{r}_2)$ are identical, i.e.,

$$\rho_{++}^{T(2N)}(\mathbf{r}_1, \mathbf{r}_2) \sim \rho_{+-}^{T(2N)}(\mathbf{r}_1, \mathbf{r}_2) \sim z^{2N} \frac{\alpha_{2N}(\Gamma)}{r_{12}^4} \tag{3.22}$$

In (3.22) $\alpha_{2N}(\Gamma)$ is a dimensionless coefficient which diverges when $\Gamma \to 4^+$. Its evaluation becomes rapidly intricate as N increases, because of the combinatories and the difficulty in explicitly computing the averages of P^2 weighted by $[\exp(-\beta W)]^T$.

The previous analysis can also be applied to the three- and four-particle correlations (and to any higher-order correlations of course). Because of the symmetric nature of the present CG, we only have to consider $\rho_{+++}^T(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)$, $\rho_{++-}^T(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)$, $\rho_{++++}^T(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4)$, $\rho_{+++-}^T(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4)$, and $\rho_{++--}^T(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4)$. Again, at any order z^{2N}, the dominant contributions to the large-distance behaviors arise from configurations where all the field points belong to neutral clusters attached to one isolated root point or to a set of several close root points. The corresponding potential V_{2N} is split in a way similar to (3.18), and the Boltzmann factor $\exp(-\beta V_{2N})$ is expanded with respect to the multipole–multipole interaction potentials between the neutral clusters. Finally we get (omitting the coefficients and the possible angular dependences)

$$\rho_{+++}^{T(2N)}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) \sim \rho_{++-}^{T(2N)}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) \sim \frac{1}{r_{13}^4}, \quad r_{13} \to \infty, \ r_{12} \text{ fixed} \tag{3.23}$$

$$\rho_{++-}^{T(2N)}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) \sim \frac{1}{r_{12}^4}, \qquad r_{12} \to \infty, \quad r_{13} \text{ fixed} \tag{3.24}$$

$$\rho_{+++}^{T(2N)}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) \sim \rho_{++-}^{T(2N)}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) \sim \frac{1}{r_{12}^2 r_{13}^2 r_{23}^2}, \qquad r_{12}, r_{13}, r_{23} \to \infty \tag{3.25}$$

$$\rho_{++++}^{T(2N)}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4) \sim \rho_{+++-}^{T(2N)}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4) \sim \frac{1}{r_{14}^4}, \qquad r_{14} \to \infty, \quad r_{12}, r_{13} \text{ fixed} \tag{3.26}$$

$$\rho_{+++-}^{T(2N)}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4) \sim \rho_{++--}^{T(2N)}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4) \sim \frac{1}{r_{13}^4}, \qquad r_{13} \to \infty, \quad r_{12}, r_{14} \text{ fixed} \tag{3.27}$$

$$\rho_{++--}^{T(2N)}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4) \sim \frac{1}{r_{12}^4}, \qquad r_{12} \to \infty, \quad r_{13}, r_{14} \text{ fixed} \tag{3.28}$$

$$\rho^{T(2N)}_{++++}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4) \sim \rho^{T(2N)}_{++--}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4)$$

$$\sim \frac{1}{r_{13}^4}, \qquad r_{13} \to \infty, \quad r_{12}, r_{34} \text{ fixed} \tag{3.29}$$

$$\rho^{T(2N)}_{+++-}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4) \sim \frac{1}{r_{13}^3}, \qquad r_{13} \to \infty, \quad r_{12}, r_{34} \text{ fixed} \tag{3.30}$$

$$\rho^{T(2N)}_{++--}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4) \sim \frac{1}{r_{12}^2}, \qquad r_{12} \to \infty, \quad r_{13}, r_{24} \text{ fixed} \tag{3.31}$$

$$\rho^{T(2N)}_{++++}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4) \sim \rho^{T(2N)}_{+++-}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4)$$

$$\sim \rho^{T(2N)}_{++--}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4)$$

$$\sim \frac{1}{r_{13}^2 r_{34}^2 r_{14}^2}, \qquad r_{13}, r_{14}, r_{34} \to \infty, \quad r_{12} \text{ fixed} \tag{3.32}$$

$$\rho^{T(2N)}_{+++-}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4) \sim \rho^{T(2N)}_{++--}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4)$$

$$\sim \frac{1}{r_{12}^2 r_{13}^2 r_{23}^2}, \qquad r_{12}, r_{13}, r_{23} \to \infty, \quad r_{14} \text{ fixed} \tag{3.33}$$

$$\rho^{T(2N)}_{++++}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4) \sim \rho^{T(2N)}_{+++-}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4)$$

$$\sim \rho^{T(2N)}_{++--}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4)$$

$$\sim \frac{1}{r_{12}^2 r_{23}^2 r_{34}^2 r_{41}^2} + \frac{1}{r_{13}^2 r_{32}^2 r_{24}^2 r_{41}^2}$$

$$+ \frac{1}{r_{13}^2 r_{34}^2 r_{42}^2 r_{21}^2}, \qquad r_{ij} \to \infty \tag{3.34}$$

The above behaviors are understood to be valid at any order z^{2N}, with the possible exception of the first-order terms, which may decay faster [for instance, $\varrho^{T(4)}_{++-}$ decays faster than (3.23)]. Furthermore, for some special asymptotic configurations of the root points, the three- and four-particle correlations decay slower than the two-particle ones: in these configurations, either there exists at least three (or more) far away sets with at least one root point in each of them, or there are two far away sets with two (or more) root points in them. For the first kind of configuration, such as (3.25), the products of the $1/r^2$ terms arise from the products of dipole–dipole interactions between a given neutral cluster and two different other ones. For the second kind of configuration, the two neutral clusters attached to the two sets of root points are no longer invariant under rotations and the resulting averages of the multipole–multipole interactions do not necessarily vanish. For instance, the $1/r^3$ and $1/r^2$ terms in (3.30) and (3.31) arise from quadrupole–dipole interactions and from dipole–dipole interactions, respectively.

3.3. Comments

In order to make a rigorous estimation of the large-distance behaviors of the particle correlations by starting from the above term-by-term analysis of the low-fugacity expansions, one should calculate explicitly the coefficients involved in (3.22)–(3.34), resum the corresponding series in z^2, and control the series of the rests. This is a formidable task, which is far beyond the scope of the present paper. At this level, and as will be discussed in Section 4.3, it is reasonable to assume that the z-expansions converge for $\Gamma > 4$ fixed and z sufficiently small[7]: the particle correlations should then decay as each term of the expansions. In Section 5, the corresponding decays will be shown to be compatible with the asymptotic structure of the BGY equations. Moreover, they do not satisfy the conditions found by Martin and Gruber[18] for a conducting system: the phase is dielectric.

The large-distance behaviors predicted by our analysis are very similar to those relative to classical neutral molecules carrying permanent dipoles in three dimensions.[13] The correlations between two molecules with given orientations of their dipoles indeed decay as $1/r^3$, i.e., as the dipole–dipole potential.[25] Once the integrations over these orientations have been performed, the residual position correlations decay faster, as $1/r^6$, which is the square of the dipole–dipole potential.[13] In our system, the four-particle correlations ρ^T_{-+-+} and the two-body ones (either ρ^T_{++} or ρ^T_{+-}) are the analogs of the full molecule–molecule correlations and of the reduced position–position correlations, respectively.

The above behaviors are not special to the present symmetric version of the CG. In fact, one always has the same kind of power-law decays, whatever the precise form of the short-range potentials w^{SR}_{++}, w^{SR}_{+-}, and w^{SR}_{--} may be. Only the coefficients in front of these power laws depend on the latter, through integrals of the form (3.13), for instance, with $\exp[-\beta v_{+-}(t)]$ in place of $(\sigma/t)^\Gamma$. Furthermore, we point out that $\rho^T_{++}(r)$, $\rho^T_{+-}(r)$, and $\rho^T_{--}(r)$ have exactly the same decay when $r \to \infty$,

[7] For a gas of permanent dipoles, Gawedski and Kupiainen[22] have proved that the fugacity expansion of the pressure has a finite radius of convergence. This result has been extended to the hierarchical dipole gas by Benfatto *et al.*[23]. Dimock and Hurd should be able to prove the analyticity of the pressure in z near $z = 0$ for the CG, on the basis of their recent paper.[24]

even in the case of nonsymmetric CG in which $\rho^T_{++}(r)$ and $\rho^T_{--}(r)$ are not identical at finite distances. Similar large-distance identities between higher-order correlations hold as in the symmetric case.

4. RESUMMATIONS NEAR THE ZERO-DENSITY CRITICAL POINT

As in Section 3, we consider a symmetric CG made of charged hard disks. We study the large-distance behavior of the charge correlation $C(r)$ in the dielectric phase near the zero-density critical point, where both z and $(\Gamma-4)$ are small parameters. This analysis starts from the low-fugacity expansion of $C(r)$: at a given order z^{2N}, the large-distance behavior of $C^{(2N)}(r)$ is determined by replacing $\rho^{T(2N)}_{++}$ and $\rho^{T(2N)}_{+-}$ in (3.3) by their asymptotic expansions. According to (3.22), the multipolar $1/r^4$ contributions from the particle correlations cancel out. The detailed study of the orders z^2 and z^4 suggests that the remaining terms should decay as $1/r^\Gamma$ multiplied by powers of $\ln(r/\sigma)$ with coefficients which diverge when $\Gamma \to 4^+$. In the regime where z and $(\Gamma-4)$ are of the same order, these divergent terms are resummed via a recurrence method which leads to a system of coupled differential equations. In the latter, the dielectric constant ε, which appears in a very natural way, must be calculated self-consistently with the asymptotic behavior of $C(r)$. This is due to the fact that the dominant contributions to the integral expression of ε given by (2.8) arise from the large distances, in the limit $\Gamma \to 4^+$. At every order z^{2N}, these contributions diverge like $1/(\Gamma-4)^{2N-1}$, as a result of the non-integrability of $r^2 C^{(2N)}(r)$ at $\Gamma=4$ when $r\to\infty$. In the present limit, all these contributions are equivalent and consequently must be indeed resummed.

In Section 4.1 we first describe the above method, which is composed of four steps. The first two steps deal with the rigorous study of the z^2 and z^4 terms. A recurrence scheme for handling the higher-order terms is then introduced. The final step consists in deriving the required differential equations which incorporate the full resummation of the low-fugacity expansions in a systematic way. The results are displayed in Section 4.2 and are discussed in Section 4.3.

4.1. Method

Step 1. The Order z^2. The z^2 term in the expansion of $C(r)$ obviously reduces to

$$C^{(2)}(r) = -2e^2 \frac{z^2}{\sigma^4}\left(\frac{\sigma}{r}\right)^\Gamma \tag{4.1}$$

Its contribution to $1/\varepsilon$ is readily obtained by inserting (4.1) in (2.8), with the result

$$\left(\frac{1}{\varepsilon}\right)^{(2)}=\frac{-2\pi^2\Gamma}{\Gamma-4}z^2 \tag{4.2}$$

where $(1/\varepsilon)^{(2N)}$ denotes the term of order z^{2N} in the expansion of $1/\varepsilon$. In the limit $\Gamma\to 4^+$, (4.2) behaves as

$$\left(\frac{1}{\varepsilon}\right)^{(2)}=-8\pi^2\frac{z^2}{\Gamma-4}+o\left(\frac{z^2}{\Gamma-4}\right) \tag{4.3}$$

The point is that the divergent behavior (4.3) arises from the large-distance contributions of $C^{(2)}(r)$ to $1/\varepsilon$. Indeed, when splitting the integral

$$\pi^2\beta\int_\sigma^\infty dr\, r^3 C^{(2)}(r) \tag{4.4}$$

into

$$\pi^2\beta\int_\sigma^l dr\, r^3 C^{(2)}(r)+\pi^2\beta\int_l^\infty dr\, r^3 C^{(2)}(r) \tag{4.5}$$

where l is some given distance, we see that, when $\Gamma\to 4^+$, the first integral in (4.5) behaves as $-8\pi^2 z^2\ln(l/\sigma)$, whereas the second one exactly behaves as (4.3) whatever the value of l may be. This suggests that the leading $z^2/(\Gamma-4)$ contributions to $(1/\varepsilon)^{(2)}$ can be calculated by using in (2.8) the limit form $C_{\mathrm{AS}}^{(2)}(r)$ when $\Gamma\to 4^+$ of the leading term in the large-distance expansion of $C^{(2)}(r)$. Since $C_{\mathrm{AS}}^{(2)}(r)$ reduces in fact to $C^{(2)}(r)$ itself, we obviously can write at the order z^2 included

$$\frac{1}{\varepsilon}=1+\pi^2\beta\int_0^\infty dr\, r^3[C_{\mathrm{AS}}^{(2)}(r)+\cdots]+o\left(\frac{z^2}{\Gamma-4}\right) \tag{4.6}$$

and

$$C_{\mathrm{AS}}(r)=-2e^2\frac{z^2}{\sigma^4}(1+\cdots)\left(\frac{\sigma}{r}\right)^{\Gamma[(1/\varepsilon)^{(0)}+\cdots]} \tag{4.7}$$

Equations (4.6) and (4.7) indicate that the calculation of $C_{\mathrm{AS}}(r)$ at all orders in z^2 is linked to the corresponding determination of $1/\varepsilon$. This connection will appear more explicitly at the order z^4.

Step 2. The Order z^4. According to the detailed study of $\rho_{++}^{T(4)}$ and $\rho_{+-}^{T(4)}$ in Section 3.2, $C_{\mathrm{AS}}^{(4)}(r)$ is directly obtained from (3.16),

$$C_{\mathrm{AS}}^{(4)}(r)=-\frac{64\pi^2}{\Gamma-4}e^2\frac{z^4}{\sigma^4}\left(\frac{\sigma}{r}\right)^{\Gamma}\ln\left(\frac{r}{\sigma}\right) \tag{4.8}$$

The coefficient $1/(\Gamma-4)$ arises from the integral (3.13), which is related to the z^2 term of the expansion of $1/\varepsilon$. Indeed, noting that

$$\left(\frac{\sigma}{r}\right)^{\Gamma/\varepsilon}=\left(\frac{\sigma}{r}\right)^{\Gamma}\exp\left[\left(1-\frac{1}{\varepsilon}\right)\Gamma\ln\left(\frac{r}{\sigma}\right)\right]$$
$$=\left(\frac{\sigma}{r}\right)^{\Gamma}\left[1+\frac{2\pi^2\Gamma^2}{\Gamma-4}z^2\ln\left(\frac{r}{\sigma}\right)+O(z^4)\right] \tag{4.9}$$

we then see that it is tempting to reexponentiate $[C_{\text{AS}}^{(2)}(r)+C_{\text{AS}}^{(4)}(r)]$ through a formula similar to (4.7). Furthermore, taking into account the identity

$$\int_\sigma^\infty \frac{dr\, r^3}{\sigma^4}\left(\frac{\sigma}{r}\right)^{\Gamma}\ln\left(\frac{r}{\sigma}\right)=\frac{1}{(\Gamma-4)^2} \tag{4.10}$$

we see that $C_{\text{AS}}^{(4)}(r)$ indeed contributes a term of order $z^4/(\Gamma-4)^3$ to $1/\varepsilon$. This contribution reads

$$\pi^2\beta\int_\sigma^\infty dr\, r^3 C_{\text{AS}}^{(4)}(r)=-\frac{256\pi^4 z^4}{(\Gamma-4)^3}+o\left(\frac{z^4}{(\Gamma-4)^3}\right) \tag{4.11}$$

if we *a priori* assume a formula similar to (4.6). In fact, in order to extend Eqs. (4.6), (4.7) to the present order, one must first carefully control all the terms in the large-r expansion of $C^{(4)}(r)$ which decay like $1/r^\Gamma$ [apart from possible powers of $\ln(r/\sigma)$] in the limit $\Gamma\to 4^+$ and which might give contributions to $1/\varepsilon$ of order $z^4/(\Gamma-4)^3$. Second, one must prove that these contributions can be calculated by replacing in (2.8) $C^{(4)}(r)$ by its corresponding truncated large-r expansion over the whole integration domain from σ to infinity. These two points are investigated in Appendix A by starting from the integral representation of $C^{(4)}(r)$,

$$C^{(4)}(r)=e^2\frac{z^4}{\sigma^4}\left(\frac{\sigma}{r}\right)^{\Gamma}\int^* d\mathbf{v}_1\, d\mathbf{v}_2\left\{\left(\frac{r}{\sigma}\right)^{2\Gamma}\frac{|\mathbf{v}_1-\mathbf{v}_2|^\Gamma}{v_1^\Gamma v_2^\Gamma\,|\mathbf{v}_1-\mathbf{r}/\sigma|^\Gamma\,|\mathbf{v}_2-\mathbf{r}/\sigma|^\Gamma}\right.$$
$$-\frac{v_2^\Gamma\,|\mathbf{v}_1-\mathbf{r}/\sigma|^\Gamma}{v_1^\Gamma\,|\mathbf{v}_2-\mathbf{r}/\sigma|^\Gamma\,|\mathbf{v}_1-\mathbf{v}_2|^\Gamma}$$
$$\left.-\frac{v_1^\Gamma\,|\mathbf{v}_2-\mathbf{r}/\sigma|^\Gamma}{v_2^\Gamma\,|\mathbf{v}_1-\mathbf{r}/\sigma|^\Gamma\,|\mathbf{v}_1-\mathbf{v}_2|^\Gamma}+\frac{2}{|\mathbf{v}_1-\mathbf{v}_2|^\Gamma}\right\} \tag{4.12}$$

where the notation $\int^*$ means that the integration domain for $\mathbf{v}_1$ and $\mathbf{v}_2$ excludes the configurations such that any of the relative distances between the points $\mathbf{0}$, $\mathbf{r}/\sigma$, $\mathbf{v}_1$, $\mathbf{v}_2$ is smaller than 1. This expression is easily obtained from the integrals defining $\rho_{++}^{T(4)}$ and $\rho_{+-}^{T(4)}$ in (3.7) and (3.8), where we have symmetrized the corresponding integrands and introduced the dimensionless variables $\mathbf{v}_1$ and $\mathbf{v}_2$, which denote the positions of the field points in units of σ. The method consists in rewriting $C^{(4)}(r)$ as a part $C_\varepsilon^{(4)}(r)$ which gives contributions to $1/\varepsilon$ of order $z^4/(\Gamma-4)^3$, plus a remainder which is uniformly bounded with respect to r and Γ. These uniform bounds are used to prove that this rest gives contributions to $1/\varepsilon$ bounded by $\text{cst} \times z^4/(\Gamma-4)^2$ and that $C_\varepsilon^{(4)}(r)$ reduces to the first three terms in the large-r expansion of $C^{(4)}(r)$ where the involved coefficients are replaced by their limit forms when $\Gamma \to 4^+$. We find

$$C_\varepsilon^{(4)}(r) = -\frac{64\pi^2 e^2 z^4}{(\Gamma-4)\,\sigma^4}\left(\frac{\sigma}{r}\right)^{\Gamma}\left[\ln\left(\frac{r}{\sigma}\right) - \frac{1}{\Gamma-4} + \frac{1}{\Gamma-4}\left(\frac{\sigma}{r}\right)^{\Gamma-4}\right] \tag{4.13}$$

while the contributions to $1/\varepsilon$ of order $z^4/(\Gamma-4)^3$ can be calculated from

$$\pi^2\beta \int_\sigma^\infty dr\, r^3 C_\varepsilon^{(4)}(r) \tag{4.14}$$

as

$$\left(\frac{1}{\varepsilon}\right)^{(4)} = -\frac{128\pi^2 z^4}{(\Gamma-4)^3} + o\left(\frac{z^4}{(\Gamma-4)^3}\right) \tag{4.15}$$

Note that $C_{\text{AS}}^{(4)}(r)$ indeed is the leading term (when $r \to \infty$) in (4.13) and its contribution (4.11) to $1/\varepsilon$ is twice the total contribution (4.15). The required extensions of (4.6), (4.7) at the order z^4 then read

$$\left(\frac{1}{\varepsilon}\right) = 1 + \pi^2\beta \int_\sigma^\infty dr\, r^3 [C_\varepsilon^{(2)}(r) + C_\varepsilon^{(4)}(r) + \cdots] + o\left(\frac{z^2}{\Gamma-4}\right) \tag{4.16}$$

and

$$C_{\text{AS}}(r) = -2e^2\frac{z^2}{\sigma^4}\left[1 - \frac{32\pi^2 z^2}{(\Gamma-4)^2} + \cdots\right]\left(\frac{\sigma}{r}\right)^{\Gamma[1 + (1/\varepsilon)^{(2)} + \cdots]} \tag{4.17}$$

where we have used the obvious relation $C_\varepsilon^{(2)}(r) = C_{\text{AS}}^{(2)}(r)$. We stress that the subleading correction which behaves as $(\sigma/r)^\Gamma$ in (4.13) contributes to the resummation process of $C_{\text{AS}}(r)$ through the term of order z^4 in the constant prefactor in front of $(\sigma/r)^{\Gamma[1+\cdots]}$. Moreover, it is understood that $(1/\varepsilon)^{(2)}$ is replaced by its limit form (4.3) when $\Gamma \to 4^+$.

Equations (4.16), (4.17) clearly illustrate the interplay between the resummation processes of $1/\varepsilon$ and $C_{\text{AS}}(r)$. However, since $1/\varepsilon$ is determined by $C_\varepsilon(r)$ rather than $C_{\text{AS}}(r)$, it is crucial to derive a resummation equation for $C_\varepsilon(r)$. The latter could be formulated in a similar way to the expansion for $C_{AS}(r)$ given by (4.17). However, such a formulation is not very convenient because it introduces constant prefactors whose relation with $1/\varepsilon$ is not *a priori* obvious, and furthermore it involves subleading terms, the number of which increases with N. For instance, the equivalent formula to (4.17) for $C_\varepsilon(r)$ reads at the order z^4

$$\begin{aligned} C_\varepsilon(r) = & -2\frac{e^2z^2}{\sigma^4}\left[1-\frac{32\pi^2z^2}{(\Gamma-4)^2}+\cdots\right]\left(\frac{\sigma}{r}\right)^{\Gamma[1+(1/\varepsilon)^{(2)}+\cdots]} \\ & -2\frac{e^2z^2}{\sigma^4}\left[\frac{32\pi^2z^2}{(\Gamma-4)^2}+\cdots\right]\left(\frac{\sigma}{r}\right)^{2\Gamma[1+\cdots]-4} \\ & +\cdots \end{aligned} \tag{4.18}$$

In fact, the required equation for $C_\varepsilon(r)$ will be further expressed in an integral form. The derivation of this equation starts from the following central observation originating from the present study of the z^4 term. As far as the calculations of $(1/\varepsilon)^{(4)}$ and of the large-r expansion of $C^{(4)}(r)$ in the limit $\Gamma\to 4^+$ are concerned, it is legitimate to replace the expression (4.12) by the integral (see Appendix A)

$$\begin{aligned} -2\frac{e^2z^4}{\sigma^4}\left(\frac{\sigma}{r}\right)^\Gamma & \left\{\int_{\sigma<|\mathbf{x}|<r/2}\frac{d\mathbf{x}}{\sigma^2}\int_{\sigma<|\mathbf{y}-\mathbf{x}|<|\mathbf{x}|}\frac{d\mathbf{y}}{\sigma^2}\left(\frac{\sigma}{|\mathbf{x}-\mathbf{y}|}\right)^\Gamma\right. \\ & \times\frac{\Gamma^2}{2}\left[(\mathbf{x}-\mathbf{y})\cdot\left(\frac{\mathbf{x}}{|\mathbf{x}|^2}-\frac{\mathbf{x}-\mathbf{r}}{|\mathbf{x}-\mathbf{r}|^2}\right)\right]^2 \\ & +\int_{\sigma<|\mathbf{x}-\mathbf{r}|<r/2}\frac{d\mathbf{x}}{\sigma^2}\int_{\sigma<|\mathbf{y}-\mathbf{x}|<|\mathbf{x}-\mathbf{r}|}\frac{d\mathbf{y}}{\sigma^2}\left(\frac{\sigma}{|\mathbf{x}-\mathbf{y}|}\right)^\Gamma \\ & \left.\times\frac{\Gamma^2}{2}\left[(\mathbf{x}-\mathbf{y})\cdot\left(\frac{\mathbf{x}}{|\mathbf{x}|^2}-\frac{\mathbf{x}-\mathbf{r}}{|\mathbf{x}-\mathbf{r}|^2}\right)\right]^2\right\} \end{aligned} \tag{4.19}$$

Since the quantity

$$\frac{\Gamma^2}{2}\left[(\mathbf{x}-\mathbf{y})\cdot\left(\frac{\mathbf{x}}{|\mathbf{x}|^2}-\frac{\mathbf{x}-\mathbf{r}}{|\mathbf{x}-\mathbf{r}|^2}\right)\right]^2 \tag{4.20}$$

is nothing but the quadratic term in the expansion of the Boltzmann factor associated with the dipole-charge interaction potential between $\mathscr{P}$ and two opposite charges located at $\mathbf{0}$ and $\mathbf{r}$, respectively, the configurations of $(\mathbf{v}_1, \mathbf{v}_2)$ in (4.12) which contribute to $C_\varepsilon^{(4)}(r)$ are associated with the physical situation described in Fig. 3: a fixed neutral pair $\mathscr{P}_0=\{\oplus\,\mathbf{0}, \ominus\,\mathbf{r}\}$ of size r interacts with a smaller pair $\mathscr{P}=\{\oplus\,\mathbf{x}, \ominus\,\mathbf{y}\}$ which is essentially

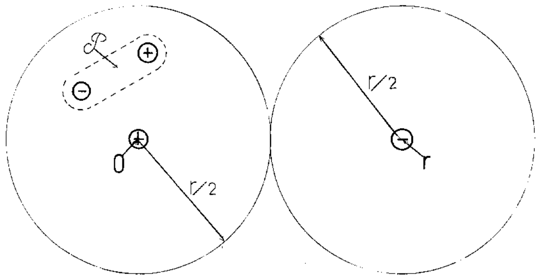

Fig. 3. A typical configuration of the field pair $\mathcal{P}$ which contributes to $C_\varepsilon^{(4)}(\mathbf{r})$. The strip delimited by the dashed line represents the pair $\mathcal{P}$.

located in one of the two disks with radius $r/2$ and centered on either $\mathbf{0}$ or $\mathbf{r}$. The size of $\mathcal{P}$ is large compared to σ, but remains small compared to r. Indeed, $C_\varepsilon^{(4)}(r)$ can be immediately computed from (4.19) by neglecting $|\mathbf{x}|$ ($|\mathbf{x}-\mathbf{r}|$) with respect to r ($|\mathbf{x}|$) in the disk $|\mathbf{x}|<r/2$ ($|\mathbf{x}-\mathbf{r}|<r/2$) and the final result (in the limit $\Gamma\to 4^+$) does not depend on the precise radius of the disks, which could be any fraction of r. Thus, $C_\varepsilon^{(4)}(r)$ can be interpreted as resulting from the sole contribution of the single pair $\mathcal{P}_0$ partially screened by the polarizable pair $\mathcal{P}$ via the operator

$$\mathcal{S}_{\mathcal{P}_0}(\mathcal{P})=\left(\frac{\sigma}{|\mathbf{x}-\mathbf{y}|}\right)^{\Gamma}\frac{\Gamma^2}{2}\times\begin{cases}\left[(\mathbf{x}-\mathbf{y})\cdot\dfrac{\mathbf{x}}{|\mathbf{x}|^2}\right]^2, & |\mathbf{x}|<\dfrac{r}{2}\\[2ex] \left[(\mathbf{x}-\mathbf{y})\cdot\dfrac{(\mathbf{x}-\mathbf{r})}{|\mathbf{x}-\mathbf{r}|^2}\right]^2, & |\mathbf{x}-\mathbf{r}|<\dfrac{r}{2}\end{cases} \tag{4.21}$$

The corresponding expression for $C_\varepsilon^{(4)}(r)$ then is

$$C_\varepsilon^{(4)}(r)=-2\frac{e^2z^4}{\sigma^4}\left(\frac{\sigma}{r}\right)^{\Gamma}\int_{|\mathcal{P}_0|>|\mathcal{P}|}d\mathcal{P}\,\mathcal{S}_{\mathcal{P}_0}(\mathcal{P}) \tag{4.22}$$

where $\int_{|\mathcal{P}_0|>|\mathcal{P}|}d\mathcal{P}$ means that one has to consider all the sizes, orientations, and locations of $\mathcal{P}$ which satisfy the geometrical constraints defined in (4.19). Note that (4.22) exactly reduces to (4.13) only if one replaces the prefactor Γ^2 in the definition (4.21) of $\mathcal{S}_{\mathcal{P}_0}(\mathcal{P})$ by 16 and $2^{\Gamma-4}$ by 1. However, it is legitimate to keep this prefactor Γ^2 in our further calculations because this does not affect the final form of $C_\varepsilon(r)$ in the limit of interest. For a similar reason, one can replace the upper integration bound $r/2$ by r in the integral over $\mathcal{P}$.

Step 3. The Recurrence Scheme at the Order z^{2N}. We shall assume that the part $C_\varepsilon^{(2N)}(r)$ of $C^{(2N)}(r)$ which contributes to $(1/\varepsilon)^{(2N)}$, the term of order $z^{2N}/(\Gamma-4)^{2N-1}$ in $1/\varepsilon$, still coincides with the limit form when

$\Gamma \to 4^+$ of the truncated large-r expansion of $C^{(2N)}(r)$. A rigorous derivation could be in principle carried out by extending the method introduced in Appendix A to the integral representation of $C^{(2N)}(r)$ with $2N-2$ field points. Here, we only guess the configurations of both root and field points which contribute to $C_\varepsilon^{(2N)}(r)$ from the above physical interpretation of $C_\varepsilon^{(4)}(r)$. Then, we briefly sketch the main arguments which lead to the generalization of the expression (4.22) at the present order z^{2N}.

In the configurations which contribute to $C_\varepsilon^{(2N)}(r)$, there are one fixed neutral pair $\mathscr{P}_0$ and $N-1$ field neutral pairs $\mathscr{P}_i$ ($i=1,\dots, N-1$). Each field pair $\mathscr{P}_i$ ($i=1,\dots, N-1$) has a large size [this induces the $1/(\Gamma-4)$ divergences] and can be viewed as nested in only one larger pair $\mathscr{P}_j$ (field or root), in the geometrical sense made explicit in Fig. 3 (with $\mathscr{P}_j$ in place of $\mathscr{P}_0$ and $\mathscr{P}_i$ in place of $\mathscr{P}$). In other words, all the field pairs $\mathscr{P}_i$ belong to chains of nested pairs, which all start with the largest root pair $\mathscr{P}_0$. We denote these as the chain $\mathscr{C}^\alpha_{\mathscr{P}_0}$ ($\alpha=1, 2,\dots$) with the symbolical notation

$$\mathscr{C}^\alpha_{\mathscr{P}_0} = \{\mathscr{P}_0 > \mathscr{P}_1^\alpha > \cdots > \mathscr{P}^\alpha_{n_\alpha}\} \qquad (n_\alpha \geqslant 1)$$

where each pair $\mathscr{P}_i^\alpha$ ($i=1,\dots, n_\alpha$) is nested in the pair $\mathscr{P}^\alpha_{i-1}$ ($\mathscr{P}_0^\alpha = \mathscr{P}_0$). Let us emphasize that a given pair may belong to various chains, i.e., one may have $\mathscr{P}_i^\alpha = \mathscr{P}_j^\gamma$ with $\alpha \neq \gamma$. In the corresponding potential V_{2N}, it is then legitimate to retain only the dipole–charge interaction potential between the dipole carried by $\mathscr{P}_i^\alpha$ and the two opposite charges constituting the larger pair $\mathscr{P}^\alpha_{i-1}$ in which it is nested (in addition to the self-energies of the pairs, of course). The dipole–charge interactions of $\mathscr{P}_i^\alpha$ with the charges constituting the pairs which belong to the same chain but which are larger than $\mathscr{P}^\alpha_{i-1}$ are negligible compared to the analogous interactions of $\mathscr{P}^\alpha_{i-1}$ with these charges, because the dipole size of $\mathscr{P}_i^\alpha$ is much smaller than that of $\mathscr{P}^\alpha_{i-1}$. The dipole–dipole interactions between pairs belonging to different chains $\mathscr{C}^\alpha_{\mathscr{P}_0}$ and $\mathscr{C}^\gamma_{\mathscr{P}_0}$ are omitted since they do not give contributions which diverge sufficiently fast with respect to $1/(\Gamma-4)$. Thus, in the expansion of the Boltzmann factor $\exp(-\beta V_{2N})$, the part which depends on $\mathscr{P}_i^\alpha$ and which contributes to C_ε^{2N} reduces to the product of the screening operators $\mathscr{S}_{\mathscr{P}^\alpha_{i-1}}(\mathscr{P}_i^\alpha)$ and $\mathscr{S}_{\mathscr{P}^\alpha_i}(\mathscr{P}^\alpha_{i+1})$ if $1 \leqslant i < n_\alpha$, and to $\mathscr{S}_{\mathscr{P}^\alpha_{n_\alpha-1}}(\mathscr{P}^\alpha_{n_\alpha})$ if $i=n_\alpha$. Summing the contributions of all the chains, we then obtain

$$C_\varepsilon^{2N}(r) = -2e^2 \frac{z^{2N}}{\sigma^4} \frac{1}{(N-1)!} \left(\frac{\sigma}{r}\right)^\Gamma \sum_{\text{chains}} \int_> d\mathscr{C}_{\mathscr{P}_0} \times \widetilde{\prod_\alpha} \left[\mathscr{S}_{\mathscr{P}_0}(\mathscr{P}_1^\alpha) \cdots \mathscr{S}_{\mathscr{P}^\alpha_{n_\alpha-1}}(\mathscr{P}^\alpha_{n_\alpha})\right] \tag{4.23}$$

where $\int_> d\mathscr{C}_{\mathscr{P}_0}$ denotes a spatial integration over all the sizes, orientations, and locations of the neutral field pairs satisfying the set of geometrical constraints $|\mathscr{P}_0| > |\mathscr{P}_1^\alpha| > \cdots > |\mathscr{P}_n^\alpha|$, and the notation $\tilde{\prod}_\alpha$ means that the screening operator $\mathscr{S}^o_{\mathscr{P}'}(\mathscr{P})$ for a given pair $\mathscr{P}$ must be counted only once in the product $\tilde{\prod}_\alpha \cdots$: a given pair screens only one larger pair [if $\mathscr{P}$ belongs to several chains, one may have the product of an arbitrary number of screening operators $\mathscr{S}_{\mathscr{P}}(\mathscr{P}')$, $\mathscr{S}_{\mathscr{P}}(\mathscr{P}'')$,..., corresponding to the screening of $\mathscr{P}$ by smaller pairs $\mathscr{P}'$, $\mathscr{P}''$,...]. In (4.23), the factor $1/(N-1)!$ arises from the product of the symmetry factor $1/[(N-1)!]^2$ attached to the diagram defining ρ_{+-}, by the number $(N-1)!$ of neutral pairs which can be formed among the $2(N-1)$ labeled charges $\{\mathbf{x}_1,..., \mathbf{x}_{N-1}; \mathbf{y}_1,..., \mathbf{y}_{N-1}\}$. Furthermore, the sum over the chains involves all the ways of building the chains $\mathscr{C}^\alpha_{\mathscr{P}_0}$ with the above neutral pairs. For instance, at the order z^6, the chains, pairs, and screening operators are represented in Figs. 4a and 4b, and the sum over the chains reduces to

$$\left[\int_{|\mathscr{P}_0| > |\mathscr{P}_1^1|} d\mathscr{P}_1^1\, \mathscr{S}_{\mathscr{P}_0}(\mathscr{P}_1^1)\right]\left[\int_{|\mathscr{P}_0| > |\mathscr{P}_1^2|} d\mathscr{P}_1^2\, \mathscr{S}_{\mathscr{P}_0}(\mathscr{P}_1^2)\right] \tag{4.24}$$

plus twice

$$\int_{|\mathscr{P}_0| > |\mathscr{P}_1^1|} d\mathscr{P}_1^1\, \mathscr{S}_{\mathscr{P}_0}(\mathscr{P}_1^1) \int_{|\mathscr{P}_1^1| > |\mathscr{P}_2^1|} d\mathscr{P}_2^1\, \mathscr{S}_{\mathscr{P}_1^1}(\mathscr{P}_2^1) \tag{4.25}$$

[the contribution (4.25) is multiplied by 2 because of the two ways of building the chain $\mathscr{C}^1_{\mathscr{P}_0} = \{\mathscr{P}_0 > \mathscr{P}_1^1 > \mathscr{P}_1^2\}$ with two given neutral pairs].

The inspection of the structure of (4.23) suggests a very natural way to derive a recurrence equation which links $C_\varepsilon^{(2N)}(r)$ to $C_\varepsilon^{(2p)}(r)$ with

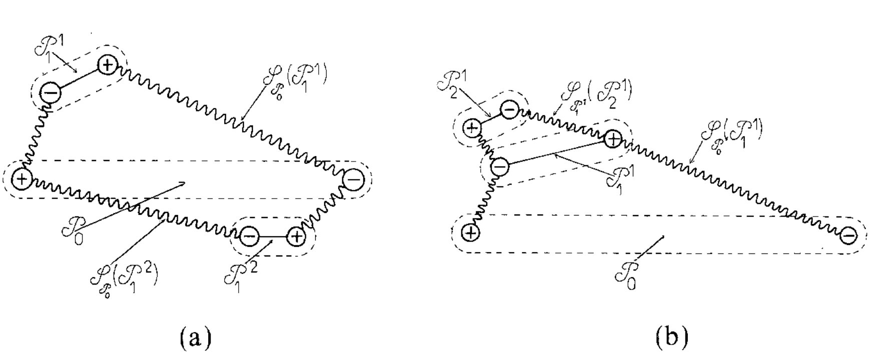

Fig. 4. The two kinds of chain configurations which contribute to $C_\varepsilon(r)$ at the order z^6. The strips delimited by the dashed lines represent the neutral pairs. The straight line connecting the two opposite charges in a pair $\mathscr{P}'$, together with the wavy lines connecting this pair to the larger one $\mathscr{P}$ in which it is nested, represent the screening operator $\mathscr{S}_{\mathscr{P}}(\mathscr{P}')$.

$1 \leqslant p \leqslant N-1$. Let $S_\varepsilon^{(2(N-1))}(r)$ be the sum over the chains appearing in (4.23),

$$C_\varepsilon^{(2N)}(r) = -2e^2 \frac{z^2}{\sigma^4} \frac{z^{2(N-1)}}{(N-1)!} \left(\frac{\sigma}{r}\right)^\Gamma S_\varepsilon^{(2(N-1))}(r) \tag{4.26}$$

We can rewrite this quantity as

$$S_\varepsilon^{(2(N-1))}(r) = \sum_{p=1}^{N-1} \frac{(N-1)!}{p!\,(N-1-p)!} \sum_{\substack{q_\alpha \geqslant 0 \\ q_1 + \cdots + q_p = N-1-p}} \frac{(N-1-p)!}{q_1! \cdots q_p!} \times I_{2q_1}(r) \cdots I_{2q_p}(r) \tag{4.27}$$

with

$$I_{2q}(r) = \int_{|\mathscr{P}_0| > |\mathscr{P}|} d\mathscr{P}\, \mathscr{S}_{\mathscr{P}_0}(\mathscr{P})\, S_\varepsilon^{(2q)}(t) \tag{4.28}$$

and where $t = |\mathbf{x} - \mathbf{y}|$ is the size of the pair $\mathscr{P} = \{\oplus\, \mathbf{x}, \ominus\, \mathbf{y}\}$. In (4.27), p is the number of different pairs $\mathscr{P}_1^\alpha$ directly nested in $\mathscr{P}_0$ and $(N-1)!/[p!\,(N-1-p)!]$ is the number of ways of choosing these p pairs among $(N-1)$. The quantity q_α is the number of pairs which belong to all the subchains starting with $\mathscr{P}_1^\alpha$, and $(N-1-p)!/(q_1! \cdots q_p!)$ is the number of ways of distributing $N-1-p$ pairs among p ensembles containing $q_1,\ldots, q_p$ pairs, respectively. The contribution of all the subchains starting with $\mathscr{P}_1^\alpha$ reduces, by definition, to $S_\varepsilon^{(2q_\alpha)}(t_1^\alpha)$, where t_1^α is the size of the pair $\mathscr{P}_1^\alpha$. Then the contribution $I_{2q_\alpha}(r)$ to $S_\varepsilon^{(2(N-1))}(r)$ from the corresponding complete chains nested in $\mathscr{P}_0$ through $\mathscr{P}_1^\alpha$ obviously takes the form (4.28). Each quantity $I_{2q_\alpha}(r)$ factorizes in (4.27) [in a similar way to (4.24), for instance] because there are no coupling terms in (4.23) between pairs belonging to chains $\{\mathscr{P}_0 > \mathscr{P}_1^\alpha > \cdots\}$ and $\{\mathscr{P}_0 > \mathscr{P}_1^\gamma > \cdots\}$ (with $\gamma \neq \alpha$), respectively. Equations (4.26)–(4.28) provide the basis of the required recurrence scheme for $C_\varepsilon^{(2N)}(r)$.

Step 4. The Coupled Differential Equations for $C_\varepsilon(r)$ and $\epsilon(r)$. Using the definition of $S_\varepsilon^{(2(N-1))}(r)$ given by (4.26) in the z^2 series representation of $C_\varepsilon(r)$, we get

$$C_\varepsilon(r) = -2e^2 \frac{z^2}{\sigma^4} \left(\frac{\sigma}{r}\right)^\Gamma \left\{1 + \sum_{N=1}^{\infty} \frac{z^{2N}}{N!} S_\varepsilon^{(2N)}(r)\right\} \tag{4.29}$$

where we have taken into account that $S_\varepsilon^{(0)}(r) = 1$. Replacing $S_\varepsilon^{(2N)}(r)$ by the recurrent expression (4.27) (with N in place of $N-1$) in each term of order z^{2N} in (4.29), we obtain

$$C_\varepsilon(r) = -2e^2 \frac{z^2}{\sigma^4}\left(\frac{\sigma}{r}\right)^\Gamma \left\{1 + \sum_{N=1}^{\infty} z^{2N} \times\left[\sum_{p=1}^{N} \frac{1}{p!} \sum_{\substack{q_\alpha \geq 0 \\ q_1+\cdots+q_p=N-p}} \frac{I_{2q_1}(r)\cdots I_{2q_p}(r)}{q_1!\cdots q_p!}\right]\right\}$$

$$= -2e^2 \frac{z^2}{\sigma^4}\left(\frac{\sigma}{r}\right)^\Gamma \times\left\{1 + \sum_{p=1}^{\infty} \frac{z^{2p}}{p!} \sum_{q_1,\ldots,q_p=0}^{\infty} \frac{z^{2q_1}}{q_1!} I_{2q_1}(r)\cdots \frac{z^{2q_p}}{q_p!} I_{2q_p}(r)\right\}$$

$$= -2e^2 \frac{z^2}{\sigma^4}\left(\frac{\sigma}{r}\right)^\Gamma \exp\left\{z^2 \sum_{q=0}^{\infty} \frac{z^{2q}}{q!} I_{2q}(r)\right\} \tag{4.30}$$

The second line in (4.30) follows from a first summation over N, whose result is both to extend the upper summation bounds relative to $p, q_1,\ldots, q_p$ to infinity, and to suppress the constraint over $q_1 + \cdots + q_p$. Replacing $I_{2q}(r)$ by (4.28) in (4.30), and inverting the sum over q and the integral over $\mathscr{P}$ in order to make $C_\varepsilon(t)$ appear, we finally get

$$C_\varepsilon(r) = -2e^2 \frac{z^2}{\sigma^4}\left(\frac{\sigma}{r}\right)^\Gamma \exp\left[-\frac{\sigma^4}{2e^2}\int_{|\mathscr{P}_0| > |\mathscr{P}|} d\mathscr{P}\, \mathscr{S}_{\mathscr{P}_0}(\mathscr{P})\, C_\varepsilon(t)\left(\frac{t}{\sigma}\right)^\Gamma\right] \tag{4.31}$$

Using the expression (4.21) of $\mathscr{S}_{\mathscr{P}_0}(\mathscr{P})$ and proceeding to an integration by parts in $\int_{|\mathscr{P}_0| > |\mathscr{P}|} d\mathscr{P}\ldots$, we can rewrite (4.31) as

$$C_\varepsilon(r) = -2e^2 \frac{z^2}{\sigma^4}\left(\frac{\sigma}{r}\right)^\Gamma \exp\left[-\pi^2\beta\Gamma \ln\left(\frac{r}{\sigma}\right)\int_\sigma^r dt\, t^3 C_\varepsilon(t) + \pi^2\beta\Gamma\int_\sigma^r dt\, t^3 \ln\left(\frac{t}{\sigma}\right) C_\varepsilon(t)\right] \tag{4.32}$$

The integral equation (4.32) for $C_\varepsilon(r)$ incorporates in a systematic way the full resummation of the whole z^2 expansion of $C_\varepsilon(r)$. In order to solve this equation, it is convenient to introduce the spatial-dependent dielectric constant $\varepsilon(r)$ defined by

$$\frac{1}{\varepsilon(r)} = 1 + \pi^2\beta\int_\sigma^r dt\, t^3 C_\varepsilon(t) \tag{4.33}$$

This quantity comes out from (4.32) in a very natural way, and is identified as above because it reduces, apart from terms $o(z^2/(\Gamma-4))$, to the dielectric constant ε in the limit $r \to \infty$. The latter is indeed related to C_ε via

$$\frac{1}{\varepsilon} = 1 + \pi^2 \beta \int_\sigma^\infty dt\, t^3 C_\varepsilon(t) \tag{4.34}$$

which is the immediate extension of (4.16) to all orders in z^2 [the terms $o(z^{2N}/(\Gamma-4)^{2N-1})$ for arbitrary N and which are left over in (4.34) are not, and will no longer be, explicitly specified, for the sake of notational convenience]. Differentiating both sides of (4.32) and (4.33) with respect to $\ln(r/\sigma)$, we easily find

$$\frac{d}{d[\ln(r/\sigma)]} \frac{1}{\varepsilon(r)} = \pi^2 \beta r^4 C_\varepsilon(r) \tag{4.35}$$

$$\frac{d}{d[\ln(r/\sigma)]} C_\varepsilon(r) = -\frac{\Gamma}{\varepsilon(r)} C_\varepsilon(r) \tag{4.36}$$

with the boundary conditions $\varepsilon(\sigma) = 1$ and $C_\varepsilon(\sigma) = -2e^2 z^2/\sigma^4$, which directly follow from (4.33) and (4.32) for $r = \sigma$. The integration of (4.36) leads to

$$C_\varepsilon(r) = -2e^2 \frac{z^2}{\sigma^4} \exp\left[-\int_\sigma^r dr' \frac{\Gamma}{r' \varepsilon(r')} \right] \tag{4.37}$$

This integral equation and the definition of $1/\varepsilon(r)$ are equivalent to the heuristic iterated mean-field theory devised by Kosterlitz and Thouless.[7] Thus, the coupled differential equations (4.35), (4.36) are equivalent to the flow equations.[15] Now we are able to express the required forms of $1/\varepsilon$, $C_\varepsilon(r)$, and $C_{AS}(r)$ in the present small-z and small-$(\Gamma-4)$ limit.

4.2. Results

The resolution of (4.35), (4.36) gives[15]

$$\frac{1}{\varepsilon} = 1 + \frac{\Gamma-4}{4} \left\{ \left[1 - \frac{(8\pi z)^2}{(\Gamma-4)^2} \right]^{1/2} - 1 \right\} \tag{4.38}$$

The full resummation of the z^2 expansion of $1/\varepsilon$ is incorporated in the function

$$\left[1 - \frac{(8\pi z)^2}{(\Gamma-4)^2} \right]^{1/2} - 1 \tag{4.39}$$

It is easily checked that the expansion of (4.39) in powers of z^2 allows us to retrieve the terms (4.3) and (4.15) of order z^2 and z^4, respectively, in the expansion of $1/\varepsilon$, as it should be. The radius of convergence of the corresponding entire series in z^2 is given by the equation

$$8\pi z = \Gamma - 4 \tag{4.40}$$

which is nothing but the equation of the transition line between the dielectric and conductive phases, predicted by the renormalization group methods.[12,15]

Integrating (4.36) from σ to r, we find

$$C_\varepsilon(r) = -2e^2 \frac{z^2}{\sigma^4} \left(\frac{\sigma}{r}\right)^{\Gamma/\varepsilon} A(r) \tag{4.41}$$

with

$$A(r) = A_0 \exp\left\{\Gamma \int_r^\infty dt \frac{1}{t} \left[\frac{1}{\varepsilon(t)} - \frac{1}{\varepsilon}\right]\right\} \tag{4.42}$$

and where A_0 is a constant independent of r,

$$A_0 = \exp\left\{-\Gamma \int_\sigma^\infty dt \frac{1}{t} \left[\frac{1}{\varepsilon(t)} - \frac{1}{\varepsilon}\right]\right\} \tag{4.43}$$

The integrals involved in the rhs of (4.42), (4.43) do converge in the dielectric phase, i.e., for $8\pi z < \Gamma - 4$, because then, according to (4.33) and (4.41), the quantity $[1/\varepsilon(t) - 1/\varepsilon]$ goes to zero as $1/t^{\Gamma/\varepsilon - 4}$ when $t \to \infty$ with Γ/ε strictly larger than 4. The amplitude coefficient A_0 is a function of $z/(\Gamma - 4)$, whose explicit calculation requires us to solve the coupled differential equations (4.35), (4.36) for all the values of r. The leading term in the large-distance expansion of $C(r)$ is readily obtained from (4.41),

$$C_{\mathrm{AS}}(r) = -2e^2 \frac{z^2}{\sigma^4} A_0 \left(\frac{\sigma}{r}\right)^{\Gamma/\varepsilon} \tag{4.44}$$

This formula is the extension of the expression (4.17) to all orders in z^2.

The structure of the subleading corrections to (4.44) can be obtained from the following recursive method. The leading term in the large-distance behavior of

$$\frac{1}{\varepsilon(r)} - \frac{1}{\varepsilon} = -\pi^2 \beta \int_r^\infty dt\, t^3 C_\varepsilon(t) \tag{4.45}$$

is given by replacing $C_\varepsilon(t)$ by $C_{\mathrm{AS}}(t)$. Using (4.44) and (4.38), we then get

$$A(r)=A_0\left\{1+\frac{[8\pi z/(\Gamma-4)]^2}{2\{1-[8\pi z/(\Gamma-4)]^2\}^{1/2}}A_0\left(\frac{\sigma}{r}\right)^{\Gamma/\varepsilon-4}+o\left(\left(\frac{\sigma}{r}\right)^{\Gamma/\varepsilon-4}\right)\right\} \tag{4.46}$$

Replacing $A(r)$ by (4.46) in (4.41), we find that the first subleading correction to $C_{\mathrm{AS}}(r)$ in the large-r expansion of $C(r)$ takes the form

$$-2\frac{e^2z^2}{\sigma^4}A_1\left(\frac{\sigma}{r}\right)^{2\Gamma/\varepsilon-4} \tag{4.47}$$

with

$$A_1=\frac{[8\pi z/(\Gamma-4)]^2}{\{1-[8\pi z/(\Gamma-4)]^2\}^{1/2}}\frac{A_0^2}{2} \tag{4.48}$$

This method can be extended to the calculation of all the subleading terms involved in $C_\varepsilon(r)$, by starting from the integrodifferential equation

$$\frac{dA}{d[\ln(r/\sigma)]}=-32\pi^2\frac{z^2}{\sigma^4}A(r)\int_r^\infty dt\,t^3\left(\frac{\sigma}{t}\right)^{\Gamma/\varepsilon}A(t) \tag{4.49}$$

which follows from the differentiation of the definition (4.42) of $A(r)$ and the combination of (4.41), (4.42), and (4.45). The final result is

$$C_\varepsilon(r)=-2\frac{e^2z^2}{\sigma^4}\left\{A_0\left(\frac{\sigma}{r}\right)^{\Gamma/\varepsilon}+\sum_{N=1}^{\infty}A_N\left(\frac{\sigma}{r}\right)^{\Gamma/\varepsilon+N(\Gamma/\varepsilon-4)}\right\} \tag{4.50}$$

which is the resummed form to all orders in z^2 of the expression (4.18). The amplitude coefficients A_N ($N\geqslant 1$) can be recursively calculated in terms of A_0 and of the function $[8\pi z/(\Gamma-4)]^2/\{1-[8\pi z/(\Gamma-4)]^2\}^{1/2}$. Their z^2 expansions only start at the order z^{2N}. All the terms in the rhs of (4.50) indeed contribute to $1/\varepsilon$, in agreement with the definition of $C_\varepsilon(r)$. At a given order z^{2N}, $C_\varepsilon^{2N}(r)$ is a linear combination of the quantities

$$-2\frac{e^2z^2}{\sigma^4}z^{2(N-1)}\frac{1}{(\Gamma-4)^{2(N-1)-n}}\left(\frac{\sigma}{r}\right)^{\Gamma+m(\Gamma-4)}\left[\ln\left(\frac{r}{\sigma}\right)\right]^n \tag{4.51}$$

with $0\leqslant m\leqslant N-1$ and $0\leqslant n\leqslant N-1$, whose precise form can be found from the z^2 expansions of A_N and $1/\varepsilon$ in (4.50). Such quantities indeed give contributions to $1/\varepsilon$ of order $z^{2N}/(\Gamma-4)^{2N-1}$ by virtue of

$$\int_\sigma^\infty\frac{dr\,r^3}{\sigma^4}\left(\frac{\sigma}{r}\right)^{\Gamma+m(\Gamma-4)}\left[\ln\left(\frac{r}{\sigma}\right)\right]^n=\frac{n!}{m^{n+1}}\frac{1}{(\Gamma-4)^{n+1}} \tag{4.52}$$

This linear combination is the extension of the expression (4.13) for $C_{\varepsilon}^{(4)}(r)$ to higher orders.

We stress again that the above results must be understood as the limit forms of the quantities of interest, in the dielectric region close to the critical point ($\Gamma = 4$, $z = 0$), where both z and $(\Gamma - 4)$ are small parameters. For instance, the interpretation of (4.50) is the following: at small but finite values of z and $(\Gamma - 4)$, the charge–charge correlation $C(r)$ in the dielectric phase has a large-r expansion of the form

$$-2\frac{e^2 z^2}{\sigma^4} \sum_{N=0}^{\infty} \mathscr{A}_N(z, \Gamma - 4)\left(\frac{\sigma}{r}\right)^{p_N} + \mathscr{R}(r) \tag{4.53}$$

with $p_0 < p_1 < \cdots$ and where the remainder $\mathscr{R}(r)$ decays faster than $(\sigma/r)^{p_N}$ for any N in the whole considered region. In the limit where both z and $(\Gamma - 4)$ go to zero, with the ratio $8\pi z/(\Gamma - 4)$ kept fixed, the amplitudes $\mathscr{A}_N$ behave as

$$\mathscr{A}_N(z, \Gamma - 4) \sim A_N(8\pi z/(\Gamma - 4)) \tag{4.54}$$

and the powers p_N collapse to 4 according to

$$p_N - 4 \sim (N+1)\left(\frac{\Gamma}{\varepsilon} - 4\right) \sim (N+1)(\Gamma - 4)\left[1 - \left(\frac{8\pi z}{\Gamma - 4}\right)^2\right]^{1/2} \tag{4.55}$$

for all N and $8\pi z < \Gamma - 4$.

4.3. Comments

The systematic resummation of the low-fugacity expansions provides exact expressions for the limit forms of the quantities of interest in the dielectric region, near the zero-density critical point. These exact calculations starting from first principles are in perfect agreement with the predictions of the renormalization group. For instance, we find that the dielectric constant does have a singularity on the transition line calculated with the RG methods.[12,15] It turns out that this singularity coincides with the divergence of the low-fugacity series for $1/\varepsilon$. This situation is a bit unusual, since, in general, the divergences of such expansions arise from singularities in the complex plane which are not associated to physical phase transitions (as in the liquid–gas problem). Furthermore, our study confirms that, at least for small values of $(\Gamma - 4)$, the effective coupling constant characterizing the fixed point of the RG flow equations can be identified with Γ/ε, as suggested in the literature. Indeed, such an identification exactly gives the resummed expression (4.38) of $1/\varepsilon$ and the power Γ/ε involved in the large-distance behavior (4.44) of $C(r)$.

Our approach also provides a more rigorous foundation to the idea of the iterated mean-field approach originally introduced by Kosterlitz and Thouless.[7] This theory takes into account the polarization of the smaller pairs by a larger one: the interaction energy of a given pair is equal to the potential of a single pair in the vacuum multiplied by an effective coupling constant which depends on the size of the pair, as if the latter were immersed in a polarizable medium. In our approach, the screening of large pairs by smaller ones is described by the operator $\mathscr{S}_{\mathscr{P}_0}(\mathscr{P})$, and the equations of the iterated mean-field theory are recovered in a very natural way, without any *a priori* assumptions. In fact, our study shows that the effective pair correlation appearing in the latter theory can be indeed identified with the charge–charge correlations of the system. We stress that one of the crucial points which allow this identification is the cancellation of the $1/r^4$ leading terms in the large-distance behaviors of the particle correlations ρ^T_{++} and ρ^T_{+-}.

At finite values of z and $(\Gamma-4)$, i.e., at finite densities, the extension of the above results is rather questionable. Then the critical value of z (for a given $\Gamma>4$) may still coincide with the radius of convergence of the low-fugacity expansion of $1/\varepsilon$, but the asymptotic behavior of $C(r)$ should no longer be proportional to $(\sigma/r)^{\Gamma/\varepsilon}$. Indeed, the behavior (4.44) is the consequence of two circumstances, (i) $1/\varepsilon$ is entirely determined by the large-distance behavior of $C(r)$, and (ii) a given pair is only screened by smaller pairs.

At finite values of $(\Gamma-4)$, these circumstances are not met, since the contributions to $1/\varepsilon$ from the finite distances in (2.8) as well as the screening by larger pairs cannot be neglected. In fact, the similarity of (4.44) to the corresponding behavior of the pair correlation between two infinitesimal external opposite charges is rather accidental and specific to the limit $\Gamma\to 4^+$. Another argument supports the above considerations: for Γ sufficiently large and z small enough, the possible $(\sigma/r)^{\Gamma/\varepsilon}$ terms decay faster than the $1/r^5$ terms which arise in the multipolar expansions of ρ^T_{++} and ρ^T_{+-} and which do not cancel out in $C(r)$. For such values of Γ, the KT transition should become a first-order transition between a dielectric gas and a conductive liquid.[26]

All our results obviously apply to any CG, i.e., with any kind of short-range potentials. The key quantity in the corresponding resummations is the integral

$$\int d\mathbf{t}\,\exp[-\beta v_{+-}(t)]\,t^2 \tag{4.56}$$

which is the equivalent of (3.13). In the limit $\Gamma\to 4^+$, this integral diverges as

$$2\pi L^4/(\Gamma-4) \tag{4.57}$$

In (4.57), L is the length which appears in the asymptotic logarithmic behavior of the potential: it also characterizes the scale where short-range interactions die out and Coulomb forces take over. Consequently the resummed expressions of $1/\varepsilon$ and of $C_\varepsilon(r)$ are merely obtained by changing σ into L.

On principle the resummation techniques should also allow one to calculate the coefficient of the $1/r^4$ terms in the large-distance behaviors of ρ^T_{++} and ρ^T_{+-} near the zero-density critical point. This amounts to calculating the limit form of the dimensionless coefficient $\alpha_{2N}(\Gamma)$ involved in (3.22) when $\Gamma \to 4^+$, for any N. We expect that, according to an analysis similar to that for $C(r)$, $\alpha_{2N}(\Gamma)$ should be proportional to $1/(\Gamma-4)^{2N-2}$, and the sizes in $[z/(\Gamma-4)]^2$ for ρ^T_{++} and ρ^T_{+-} should have the same radius of convergence as $1/\varepsilon$, or maybe a smaller one. Indeed, for $8\pi z > \Gamma-4$ the system is in its conductive phase and the particle correlations are expected to decay exponentially, a behavior which is not compatible with the convergence of the low-fugacity expansions. Notice that the amplitude coefficients A_N in the large-distance expansion (4.50) of $C(r)$ do vanish on the transition line, i.e., for $8\pi z = \Gamma - 4$: this is in agreement with the appearance of the above exponential decay.

5. ANALYSIS OF THE BGY HIERARCHY

In this section, we study the large-distance behavior of the particle correlations starting from the BGY hierarchy equations. For this purpose, we consider a symmetric version of the CG, with potentials

$$v_{++}(r) = v_{--}(r) = -v_{+-}(r) = e^2 v(r) \tag{5.1}$$

where $v(r)$ is differentiable everywhere. For instance, one may choose

$$v(r) = \begin{cases} -\ln(r/\sigma), & r > \sigma \\ \frac{1}{2}[1-(r/\sigma)^2], & r < \sigma \end{cases} \tag{5.2}$$

which corresponds to a parabolic regularization at the origin of the Coulomb potential (2.3) (this potential is the same as that created by a uniformly charged disk). In Section 5.1, we derive a new exact expression of $1/\varepsilon$ in terms of the dipole of the charge cloud surrounding two opposite fixed charges of the medium. This identity, valid in both conductive and dielectric phases, is obtained through manipulations of the BGY equations for ρ^T_{++} and ρ^T_{+-}, which are inspired by those by Martin and Gruber.[18] In Section 5.2 this identity is used to show that some particle correlations cannot decay faster than $1/r^2$ in the dielectric phase. In Section 5.3 we show that the decay scenario for the particle correlations suggested by the analysis of the low-fugacity expansions is compatible with the large-distance structure of the BGY equations. Throughout all this section, we shall assume that the BGY equations are satisfied in the homogeneous infinite system.[8]

5.1. The Dielectric Constant in Terms of a Dipole

Our starting point is the linear-response expression (2.8) which relates $1/\varepsilon$ to the second moment of the charge correlation $C(r)$. The required identity for $1/\varepsilon$ is obtained by calculating this second moment in terms of the dipoles of three-body correlations, through manipulations of the BGY equation for $C(r)$. First, we derive the latter from the BGY equations for ρ^T_{++} and ρ^T_{+-}. Then, we formulate a set of clustering assumptions which ensure the convergence of the integrals appearing in the BGY hierarchy as well as the validity of the above manipulations. The new expression obtained for $1/\varepsilon$ is finally checked in the zero-density limit.

The BGY equations for the two-body densities ρ^T_{++} and ρ^T_{+-} read

$$\begin{aligned}\nabla_2 \rho^T_{++}(\mathbf{r}_1, \mathbf{r}_2) = {} & \beta e^2 \mathbf{F}(\mathbf{r}_2 - \mathbf{r}_1)\, \rho^T_{++}(\mathbf{r}_1, \mathbf{r}_2) \\ & + \beta e^2 \int d\mathbf{r}_3\, \mathbf{F}(\mathbf{r}_2 - \mathbf{r}_3)[\rho^T_{+++}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) \\ & - \rho^T_{++-}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)] \end{aligned} \tag{5.3a}$$

$$\begin{aligned}\nabla_2 \rho^T_{+-}(\mathbf{r}_1, \mathbf{r}_2) = {} & -\beta e^2 \mathbf{F}(\mathbf{r}_2 - \mathbf{r}_1)\, \rho^T_{+-}(\mathbf{r}_1, \mathbf{r}_2) \\ & + \beta e^2 \int d\mathbf{r}_3\, \mathbf{F}(\mathbf{r}_2 - \mathbf{r}_3)[\rho^T_{+--}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) \\ & - \rho^T_{+-+}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)] \end{aligned} \tag{5.3b}$$

where $\mathbf{F}(\mathbf{r})$ is the force deriving from the potential $v(r)$, i.e.,

$$\mathbf{F}(\mathbf{r}) = -\nabla v(r) \tag{5.4}$$

It is useful to rewrite (5.3a), (5.3b) in terms of the fully truncated densities, i.e., the particle correlations. For this purpose, we use the well-known relations

$$\rho_{s_1 s_2}(\mathbf{r}_1, \mathbf{r}_2) = \rho^2 + \rho^T_{s_1 s_2}(\mathbf{r}_1, \mathbf{r}_2) \tag{5.5}$$

$$\begin{aligned}\rho_{s_1 s_2 s_3}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) = {} & \rho^3 + \rho\rho^T_{s_1 s_2}(\mathbf{r}_1, \mathbf{r}_2) + \rho\rho^T_{s_1 s_3}(\mathbf{r}_1, \mathbf{r}_3) + \rho\rho^T_{s_2 s_3}(\mathbf{r}_2, \mathbf{r}_3) \\ & + \rho^T_{s_1 s_2 s_3}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) \end{aligned} \tag{5.6}$$

[8] The validity of the BGY equations for the correlations of the infinite system has been proved, at sufficiently high temperature, by Fontaine and Martin.[27]

and we introduce the total charge density $Q_{s_1 \cdots s_n}(\mathbf{r}_1 \cdots \mathbf{r}_n | \mathbf{r})$ induced at $\mathbf{r}$ in the medium by n charges $s_i e$ fixed at $\mathbf{r}_i$ ($i = 1,..., n$), i.e.,

$$Q_{s_1 \cdots s_n}(\mathbf{r}_1 \cdots \mathbf{r}_n | \mathbf{r}) = \frac{e[\rho_{s_1 \cdots s_n +}(\mathbf{r}_1,..., \mathbf{r}_n, \mathbf{r}) - \rho_{s_1 \cdots s_n -}(\mathbf{r}_1,..., \mathbf{r}_n, \mathbf{r})]}{\rho_{s_1 \cdots s_n}(\mathbf{r}_1,..., \mathbf{r}_n)} + \sum_{i=1}^{n} s_i e \delta(\mathbf{r} - \mathbf{r}_i) \tag{5.7}$$

From these definitions, we easily find

$$\begin{aligned} \nabla_2 \rho^T_{++}(\mathbf{r}_1, \mathbf{r}_2) = {} & \beta e^2 \mathbf{F}(\mathbf{r}_2 - \mathbf{r}_1)\, \rho^T_{++}(\mathbf{r}_1, \mathbf{r}_2) \\ & + \beta e \rho^2 \int d\mathbf{r}_3\, \mathbf{F}(\mathbf{r}_2 - \mathbf{r}_3)\, Q_+(\mathbf{r}_1 | \mathbf{r}_3) \\ & + \beta e^2 \int d\mathbf{r}_3\, \mathbf{F}(\mathbf{r}_2 - \mathbf{r}_3)[\rho^T_{+++}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) \\ & - \rho^T_{++-}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)] \end{aligned} \tag{5.8a}$$

$$\begin{aligned} \nabla_2 \rho^T_{+-}(\mathbf{r}_1, \mathbf{r}_2) = {} & -\beta e^2 \mathbf{F}(\mathbf{r}_2 - \mathbf{r}_1)\, \rho^T_{+-}(\mathbf{r}_1, \mathbf{r}_2) \\ & - \beta e \rho^2 \int d\mathbf{r}_3\, \mathbf{F}(\mathbf{r}_2 - \mathbf{r}_3)\, Q_+(\mathbf{r}_1 | \mathbf{r}_3) \\ & + \beta e^2 \int d\mathbf{r}_3\, \mathbf{F}(\mathbf{r}_2 - \mathbf{r}_3)[\rho^T_{+--}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) \\ & - \rho^T_{+-+}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)] \end{aligned} \tag{5.8b}$$

For the present symmetric CG,

$$C(r_{12}) = 2e^2[\rho^T_{++}(\mathbf{r}_1, \mathbf{r}_2) - \rho^T_{+-}(\mathbf{r}_1, \mathbf{r}_2)] + 2e^2 \rho \delta(\mathbf{r}_1 - \mathbf{r}_2) \tag{5.9}$$

Then the BGY equation for the charge correlation directly follows from the subtraction of (5.8b) to (5.8a), i.e.,

$$\begin{aligned} & \nabla_2[C(r_{12}) - 2e^2 \rho \delta(\mathbf{r}_1 - \mathbf{r}_2)] \\ & = 2\beta e^2 \rho \int d\mathbf{r}_3\, \mathbf{F}(\mathbf{r}_2 - \mathbf{r}_3)\, C(r_{13}) + 2\beta e^4 \int d\mathbf{r}_3\, \mathbf{F}(\mathbf{r}_2 - \mathbf{r}_3) \\ & \quad \times [\rho^T_{+++}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) - \rho^T_{++-}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) + \rho^T_{++}(\mathbf{r}_1, \mathbf{r}_2)\, \delta(\mathbf{r}_3 - \mathbf{r}_1) \\ & \quad + \rho^T_{+-+}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) - \rho^T_{+--}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) + \rho^T_{+-}(\mathbf{r}_1, \mathbf{r}_2)\, \delta(\mathbf{r}_3 - \mathbf{r}_1)] \end{aligned} \tag{5.10}$$

In deriving (5.10), we have also used

$$C(r_{13}) = 2e\rho Q_{+}(\mathbf{r}_1 | \mathbf{r}_3) \tag{5.11}$$

which is an obvious consequence of (5.7) with ($n = 1$, $s_1 = +$) and (5.9).

In order to ensure the convergence of the integrals in the rhs of (5.8a), (5.8b), and (5.10), the two- and three-particle correlations must have a minimal decay when one particle is sent to infinity. It is easy to check that a $1/r^{1+\delta}$ decay ($\delta > 0$) makes the above integrals absolutely convergent. In the following, we shall assume stronger clustering properties, namely

$$|\rho^T_{s_1 s_2}(\mathbf{r}_1, \mathbf{r}_2)| < \frac{\text{cst}}{r_{12}^{3+\delta}} \tag{5.12a}$$

$$C(r_{12}) < \frac{\text{cst}}{r_{12}^{4+\delta}} \tag{5.12b}$$

$$|\rho^T_{s_1 s_2 s_3}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)| < \frac{N_3(\text{Inf}(r_{ik}, r_{kj}))}{r_{ij}^{3+\delta}} \tag{5.12c}$$

$$|\rho^T_{s_1 s_2 s_3 s_4}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4)| < \frac{N_4(\text{Inf}(r_{ik}, r_{kj}), \text{Inf}(r_{il}, r_{lj}))}{r_{ij}^{1+\delta}} \tag{5.12d}$$

where $N_3(r)$ and $N_4(r, s)$ are bounded functions of r and s such that $r^{2+\delta} N_3(r)$, $r^{1+\delta} N_4(r, s)$, and $s^{1+\delta} N_4(r, s)$ are also bounded. The clustering assumptions (5.12) should be always satisfied. Indeed, an exponential decay of the correlations is expected in the conductive phase, while the algebraic decays in the dielectric phase suggested by the low-fugacity expansions are compatible with the bounds (5.12). Note that the latter imply the perfect screening sum rules (see, e.g., ref. 28)

$$\int d\mathbf{r}\, C(r) = 0 \tag{5.13a}$$

$$\int d\mathbf{r}\, Q_{s_1 s_2}(\mathbf{r}_1, \mathbf{r}_2 | \mathbf{r}) = 0 \tag{5.13b}$$

which are indeed expected to hold in both phases.

In order to calculate the second moment of $C(r)$, we use a method first introduced by Martin and Gruber.[18] Let us multiply each side of Eq. (5.10) by $(\mathbf{r}_1 - \mathbf{r}_2)$ and integrate the resulting equality over $\mathbf{r}_1$. If we take into account the translation invariance of the homogeneous infinite system which allows us to change ∇_2 into $-\nabla_1$ in the lhs of (5.10), we then obtain

$$-\int d\mathbf{r}_1\,(\mathbf{r}_1-\mathbf{r}_2)\cdot\nabla_1[C(r_{12})-2e^2\rho\delta(\mathbf{r}_1-\mathbf{r}_2)]$$

$$=2\beta e^2\rho\int d\mathbf{r}_1\,(\mathbf{r}_1-\mathbf{r}_2)\cdot\int d\mathbf{r}_3\,\mathbf{F}(\mathbf{r}_2-\mathbf{r}_3)\,C(r_{13})$$

$$+2\beta e^4\int d\mathbf{r}_1\,(\mathbf{r}_1-\mathbf{r}_2)\cdot\int d\mathbf{r}_3\,\mathbf{F}(\mathbf{r}_2-\mathbf{r}_3)$$

$$\times\{\rho^T_{+++}(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)+\rho^T_{+-+}(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)-\rho^T_{++-}(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)$$
$$-\rho^T_{+--}(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)+[\rho^T_{++}(\mathbf{r}_1,\mathbf{r}_2)+\rho^T_{+-}(\mathbf{r}_1,\mathbf{r}_2)]\,\delta(\mathbf{r}_1-\mathbf{r}_3)\} \tag{5.14}$$

The left-hand side of (5.14) is easily calculated via an integration by parts,

$$-\int d\mathbf{r}_1\,(\mathbf{r}_1-\mathbf{r}_2)\cdot\nabla_1[C(r_{12})-2e^2\rho\delta(r_{12})]$$

$$=\int d\mathbf{r}_1\,[C(r_{12})-2e^2\rho\delta(r_{12})]\,\nabla_1\cdot(\mathbf{r}_1-\mathbf{r}_2)$$

$$=2\int d\mathbf{r}_1\,C(r_{12})-4e^2\rho$$

$$=-4e^2\rho \tag{5.15}$$

where the last line follows from the perfect screening sum rule (5.13a). Note that the contribution from the surface terms in the integration by parts vanishes by virtue of (5.12b). For calculating the first term of the right-hand side of (5.14), we split the force $\mathbf{F}$ into a short-range part $\mathbf{F}^{\mathrm{SR}}$ and the long-range Coulomb part $\mathbf{F}_{\mathrm{C}}=-\nabla v_{\mathrm{C}}$:

$$2\beta e^2\rho\int d\mathbf{r}_1\,(\mathbf{r}_1-\mathbf{r}_2)\cdot\int d\mathbf{r}_3\,\mathbf{F}(\mathbf{r}_2-\mathbf{r}_3)\,C(r_{13})$$

$$=2\beta e^2\rho\int d\mathbf{r}_1\,(\mathbf{r}_1-\mathbf{r}_2)\cdot\int d\mathbf{r}_3\,\mathbf{F}^{\mathrm{SR}}(\mathbf{r}_2-\mathbf{r}_3)\,C(r_{13})$$

$$+2\beta e^2\rho\int d\mathbf{r}_1\,(\mathbf{r}_1-\mathbf{r}_2)\cdot\int d\mathbf{r}_3\,\mathbf{F}_{\mathrm{C}}(\mathbf{r}_2-\mathbf{r}_3)\,C(r_{13}) \tag{5.16}$$

In the first integral of the rhs of (5.16), we can invert the integrals over $\mathbf{r}_1$ and $\mathbf{r}_3$, because the decay of $(\mathbf{r}_1-\mathbf{r}_2)\cdot\mathbf{F}^{\mathrm{SR}}(\mathbf{r}_2-\mathbf{r}_3)\,C(r_{13})$ at infinity for any configuration $(\mathbf{r}_1,\mathbf{r}_3)$ is sufficiently fast to ensure absolute convergence. We then obtain

$$2\beta e^2\rho \int d\mathbf{r}_1\,(\mathbf{r}_1-\mathbf{r}_2)\cdot \int d\mathbf{r}_3\,\mathbf{F}^{SR}(\mathbf{r}_2-\mathbf{r}_3)\,C(r_{13})$$
$$=2\beta e^2\rho \int d\mathbf{r}_3\,\mathbf{F}^{SR}(\mathbf{r}_2-\mathbf{r}_3)\cdot \int d\mathbf{r}_1\,(\mathbf{r}_1-\mathbf{r}_2)\,C(r_{13})$$
$$=2\beta e^2\rho \int d\mathbf{r}_3\,\mathbf{F}^{SR}(\mathbf{r}_2-\mathbf{r}_3)$$
$$\times\left[\int d\mathbf{r}_1\,(\mathbf{r}_1-\mathbf{r}_3)\,C(r_{13})+(\mathbf{r}_3-\mathbf{r}_2)\int d\mathbf{r}_1\,C(r_{13})\right]$$
$$=0 \tag{5.17}$$

since the net charge $\int d\mathbf{r}_1\,C(r_{13})$ identically vanishes by virtue of the perfect screening rule (5.13a), while the dipole $\int d\mathbf{r}_1\,(\mathbf{r}_1-\mathbf{r}_3)\,C(r_{13})$ also vanishes because of the rotational invariance of the homogeneous infinite system. In the second integral of (5.16), we write $(\mathbf{r}_1-\mathbf{r}_2)=\nabla_1(r_{12}^2/2)$ and we perform an integration by parts. The surface term does not give any contribution because the bound (5.12b) implies that $\int d\mathbf{r}_3\,\mathbf{F}_C(\mathbf{r}_2-\mathbf{r}_3)\,C(r_{13})$ decays faster than $1/r_{12}^{3+\delta}$, and consequently $|r_{12}^3\int d\mathbf{r}_3\,\mathbf{F}_C(\mathbf{r}_2-\mathbf{r}_3)\,C(r_{13})|$ goes to zero when $r_{12}\to\infty$. Using also

$$\nabla_1\int d\mathbf{r}_3\,\mathbf{F}_C(\mathbf{r}_2-\mathbf{r}_3)\,C(r_{13})=-\nabla_2\int d\mathbf{r}_3\,\mathbf{F}_C(\mathbf{r}_2-\mathbf{r}_3)\,C(r_{13})$$
$$=-\int d\mathbf{r}_3\,C(r_{13})\,\nabla_2\cdot\mathbf{F}_C(\mathbf{r}_2-\mathbf{r}_3) \tag{5.18}$$

which follows from the translation invariance of the homogeneous infinite system, and

$$\nabla_2\cdot\mathbf{F}_C(\mathbf{r}_2-\mathbf{r}_3)=2\pi\delta(\mathbf{r}_2-\mathbf{r}_3) \tag{5.19}$$

we finally find

$$2\beta e^2\rho \int d\mathbf{r}_1\,(\mathbf{r}_1-\mathbf{r}_2)\cdot \int d\mathbf{r}_3\,\mathbf{F}_C(\mathbf{r}_2-\mathbf{r}_3)\,C(r_{13})$$
$$=2\pi\beta e^2\rho \int d\mathbf{r}\,r^2C(r) \tag{5.20}$$

In the last integral of the rhs of (5.14), we can first perform the integration over $\mathbf{r}_1$, because the full truncation of the distribution densities inside the brackets $[\cdots]$ together with the bounds (5.12a), (5.12c) and the $1/r$ decay of $\mathbf{F}$ ensure absolute convergence. This leads to the sum of the following integrals:

$$\int d\mathbf{r}_1\,(\mathbf{r}_1-\mathbf{r}_2)[\rho^T_{+++}(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)-\rho^T_{-++}(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)+\rho^T_{++}(\mathbf{r}_1,\mathbf{r}_2)\,\delta(\mathbf{r}_1-\mathbf{r}_3)] \tag{5.21a}$$

$$\int d\mathbf{r}_1\,(\mathbf{r}_1-\mathbf{r}_2)[\rho^T_{+-+}(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)-\rho^T_{--+}(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)+\rho^T_{+-}(\mathbf{r}_1,\mathbf{r}_2)\,\delta(\mathbf{r}_1-\mathbf{r}_3)] \tag{5.21b}$$

where we have used the symmetry relations $\rho^T_{+--}=\rho^T_{-++}$ and $\rho^T_{++-}=\rho^T_{--+}$. According to the definitions (5.5)–(5.7), $Q_{++}(\mathbf{r}_2,\mathbf{r}_3\,|\,\mathbf{r}_1)$ and $Q_{-+}(\mathbf{r}_2,\mathbf{r}_3\,|\,\mathbf{r}_1)$ can be rewritten as

$$\begin{aligned}Q_{++}(\mathbf{r}_2,\mathbf{r}_3\,|\,\mathbf{r}_1)=\frac{e}{\rho_{++}(\mathbf{r}_2,\mathbf{r}_3)}\,&[\rho^T_{+++}(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)-\rho^T_{-++}(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)\\&+\rho\rho^T_{++}(\mathbf{r}_1,\mathbf{r}_2)+\rho\rho^T_{++}(\mathbf{r}_1,\mathbf{r}_3)\\&-\rho\rho^T_{-+}(\mathbf{r}_1,\mathbf{r}_2)-\rho\rho^T_{-+}(\mathbf{r}_1,\mathbf{r}_3)]\\&+e[\delta(\mathbf{r}_1-\mathbf{r}_3)+\delta(\mathbf{r}_1-\mathbf{r}_2)]\end{aligned} \tag{5.22a}$$

$$\begin{aligned}Q_{-+}(\mathbf{r}_2,\mathbf{r}_3\,|\,\mathbf{r}_1)=\frac{e}{\rho_{-+}(\mathbf{r}_2,\mathbf{r}_3)}\,&[\rho^T_{+-+}(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)-\rho^T_{--+}(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)\\&+\rho\rho^T_{+-}(\mathbf{r}_1,\mathbf{r}_2)+\rho\rho^T_{++}(\mathbf{r}_1,\mathbf{r}_3)\\&-\rho\rho^T_{--}(\mathbf{r}_1,\mathbf{r}_2)-\rho\rho^T_{-+}(\mathbf{r}_1,\mathbf{r}_3)]\\&+e[\delta(\mathbf{r}_1-\mathbf{r}_3)-\delta(\mathbf{r}_1-\mathbf{r}_2)]\end{aligned} \tag{5.22b}$$

Let $\mathbf{p}_{++}(\mathbf{r}_2,\mathbf{r}_3)$ and $\mathbf{p}_{-+}(\mathbf{r}_2,\mathbf{r}_3)$ be the dipoles carried by the charge distributions $Q_{++}(\mathbf{r}_2,\mathbf{r}_3\,|\,\mathbf{r}_1)$ and $Q_{-+}(\mathbf{r}_2,\mathbf{r}_3\,|\,\mathbf{r}_1)$, namely

$$\mathbf{p}_{++}(\mathbf{r}_2,\mathbf{r}_3)=\int d\mathbf{r}_1\,\mathbf{r}_1 Q_{++}(\mathbf{r}_2,\mathbf{r}_3\,|\,\mathbf{r}_1) \tag{5.23a}$$

$$\mathbf{p}_{-+}(\mathbf{r}_2,\mathbf{r}_3)=\int d\mathbf{r}_1\,\mathbf{r}_1 Q_{-+}(\mathbf{r}_2,\mathbf{r}_3\,|\,\mathbf{r}_1) \tag{5.23b}$$

it is easily checked, by using (5.22), that the integrals (5.21a) and (5.21b), respectively, reduce to

$$\frac{1}{e}\,\rho_{++}(\mathbf{r}_2,\mathbf{r}_3)\,\mathbf{p}_{++}(\mathbf{r}_2,\mathbf{r}_3) \tag{5.24}$$

and

$$\frac{1}{e}\rho_{-+}(\mathbf{r}_2, \mathbf{r}_3)\, \mathbf{p}_{-+}(\mathbf{r}_2, \mathbf{r}_3) \tag{5.25}$$

[in this identification the perfect screening rules (5.13) are also taken into account as well as the rotational invariance of the homogeneous infinite system]. Thus, the last integral of the rhs of (5.14) becomes

$$\begin{aligned} 2\beta e^4 \int d\mathbf{r}_1\, (\mathbf{r}_1 - \mathbf{r}_2) \cdot \int d\mathbf{r}_3\, \mathbf{F}(\mathbf{r}_2 - \mathbf{r}_3) [&\rho^T_{+++}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) + \rho^T_{+-+}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) \\ &- \rho^T_{++-}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) - \rho^T_{+--}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) \\ &+ \rho^T_{++}(\mathbf{r}_1, \mathbf{r}_2)\, \delta(\mathbf{r}_1 - \mathbf{r}_3) + \rho^T_{+-}(\mathbf{r}_1, \mathbf{r}_2)\, \delta(\mathbf{r}_1 - \mathbf{r}_3)] \\ = 2\beta e^2 \int d\mathbf{r}\, \mathbf{F}(\mathbf{r}) \cdot [\rho_{++}(r)\, \mathbf{p}_{++}(\mathbf{r}) + \rho_{-+}(r)\, \mathbf{p}_{-+}(\mathbf{r})] \end{aligned} \tag{5.26}$$

with $\mathbf{r} = \mathbf{r}_2 - \mathbf{r}_3$ and $\mathbf{p}_{++}(\mathbf{r}) = \mathbf{p}_{++}(\mathbf{r}_2, \mathbf{r}_3)$, $\mathbf{p}_{-+}(\mathbf{r}) = \mathbf{p}_{-+}(\mathbf{r}_2, \mathbf{r}_3)$. Replacing the various terms of Eq. (5.14) by their expressions (5.15), (5.17), (5.20), and (5.26), we find

$$\frac{\pi\beta}{2} \int d\mathbf{r}\, r^2 C(r) = -1 - \frac{\beta e}{2\rho} \int d\mathbf{r}\, \mathbf{F}(\mathbf{r}) \cdot [\rho_{++}(r)\, \mathbf{p}_{++}(\mathbf{r}) + \rho_{-+}(r)\, \mathbf{p}_{-+}(\mathbf{r})] \tag{5.27}$$

If we insert this expression for the second moment of $C(r)$ in (2.8), we finally obtain the required expression for $1/\varepsilon$,

$$\frac{1}{\varepsilon} = \frac{-\beta e}{2\rho} \int d\mathbf{r}\, \mathbf{F}(\mathbf{r}) \cdot [\rho_{++}(r)\, \mathbf{p}_{++}(\mathbf{r}) + \rho_{-+}(r)\, \mathbf{p}_{-+}(\mathbf{r})] \tag{5.28}$$

The integral in (5.28) does converge since $\mathbf{p}_{++}(\mathbf{r})$ and $\mathbf{p}_{-+}(\mathbf{r})$ decay faster than $1/r^{1+\delta}$ as a consequence of (5.12c).

We stress that the identity (5.28) is valid in both conductive and dielectric phases. In the conductive phase, the dipoles $\mathbf{p}_{++}(\mathbf{r})$ and $\mathbf{p}_{-+}(\mathbf{r})$ identically vanish as a consequence of the multipole sum rules.[28,29] Then, the rhs of (5.28) also vanishes, and we recover that ε is infinite. In the dielectric phase, $\mathbf{p}_{++}(\mathbf{r})$ still vanishes for symmetry reasons. However, since $1/\varepsilon$ is finite, there exists a nonzero measure ensemble of values of r, such that $\mathbf{p}_{-+}(\mathbf{r})$ is different from zero. This result is well supported by the physical picture which describes the dielectric phase as a system of neutral dipolar molecules.

The identity (5.28) can be checked in the dielectric phase at the lowest orders in z^2 by replacing the quantities of interest by their low-fugacity expansions in (5.28) and (2.8), respectively. For instance, one finds at the zeroth order from (2.8)

$$\left(\frac{1}{\varepsilon}\right)^{(0)} = 1 \tag{5.29a}$$

On the other hand, the identity (5.28) gives

$$\begin{aligned}\left(\frac{1}{\varepsilon}\right)^{(0)} &= -\frac{\beta e}{2\rho^{(2)}} \int d\mathbf{r}\, \rho^{(2)}_{-+}(r)\, \mathbf{F}(\mathbf{r}) \cdot \mathbf{p}^{(0)}_{-+}(\mathbf{r}) \\ &= -\frac{\beta e}{2z^2 \int d\mathbf{r} \exp[\beta e^2 v(r)]} \int d\mathbf{r}\, z^2 \exp[\beta e^2 v(r)]\, [-\nabla v(r)] \cdot (-e\mathbf{r}) \\ &= -\frac{1}{2 \int d\mathbf{r} \exp[\beta e^2 v(r)]} \int d\mathbf{r}\, \mathbf{r} \cdot \nabla \exp[\beta e^2 v(r)] \\ &= 1 \end{aligned} \tag{5.29b}$$

which does coincide with (5.29a), as it should be. Checking the identity at the order z^2 is more cumbersome and involves the manipulations of non-absolutely convergent integrals (see Appendix B).

5.2. Existence of Dipole–Dipole Correlations

As mentioned in Section 2, the violation of the Stillinger–Lovett sum rule implies that some correlations have an algebraic decay in the dielectric phase. Here, starting from the existence of the nonvanishing dipole $\mathbf{p}_{-+}$, we give strong arguments which suggest that ρ^T_{-+-+} should decay as $1/r^2$ (similarly to the dipole–dipole potential) when two neutral pairs are separated by a large distance r. This nonperturbative result corroborates the findings relative to the term-by-term analysis of the low-fugacity expansions of the particle correlations (see Section 3). The coefficient of the above $1/r^2$ behaviour is related to $\mathbf{p}_{-+}$ and $1/\varepsilon$ via integral expressions, which can be viewed as sum rules. The whole study is carried out through the inspection of the asymptotic structure of the BGY equations for the three-particle correlations. We give the detailed inspection only for ρ^T_{-++}.

The BGY equation for $\rho^T_{-++}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)$ directly follows from the corresponding equation for the three-body density $\rho_{-++}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)$.

Expressing also the four-body densities in terms of the particle correlations and using (5.7), (5.8), one finds

$$\nabla_3 \rho^T_{-++}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)$$
$$= \beta e^2 [\mathbf{F}(\mathbf{r}_3 - \mathbf{r}_2) - \mathbf{F}(\mathbf{r}_3 - \mathbf{r}_1)]\, \rho^T_{-++}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) \quad (5.30\text{a})$$
$$+ \beta e \rho^T_{-+}(\mathbf{r}_1, \mathbf{r}_3) \int d\mathbf{r}_4\, \mathbf{F}(\mathbf{r}_3 - \mathbf{r}_4)\, \rho Q_+(\mathbf{r}_2 \mid \mathbf{r}_4) \quad (5.30\text{b})$$
$$+ \beta e \rho^T_{++}(\mathbf{r}_2, \mathbf{r}_3) \int d\mathbf{r}_4\, \mathbf{F}(\mathbf{r}_3 - \mathbf{r}_4)\, \rho Q_-(\mathbf{r}_1 \mid \mathbf{r}_4) \quad (5.30\text{c})$$
$$+ \beta e \rho \int d\mathbf{r}_4\, \mathbf{F}(\mathbf{r}_3 - \mathbf{r}_4)[\rho_{-+}(\mathbf{r}_1, \mathbf{r}_2)\, Q_{-+}(\mathbf{r}_1, \mathbf{r}_2 \mid \mathbf{r}_4)$$
$$- \rho^2 Q_-(\mathbf{r}_1 \mid \mathbf{r}_4) - \rho^2 Q_+(\mathbf{r}_2 \mid \mathbf{r}_4)] \quad (5.30\text{d})$$
$$+ \beta e^2 \int d\mathbf{r}_4\, \mathbf{F}(\mathbf{r}_3 - \mathbf{r}_4)$$
$$\times [\rho^T_{-+++}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4) - \rho^T_{-++-}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4)] \quad (5.30\text{e})$$

Let us study the asymptotic behavior of the three-body mean-field term (5.30d) when $\mathbf{r}_3$ is sent to infinity, $\mathbf{r}_1$ and $\mathbf{r}_2$ being kept fixed. For this, we first split $\mathbf{F}$ into a short-range part $\mathbf{F}^{\mathrm{SR}}$ and the long-range Coulomb part $\mathbf{F}_{\mathrm{C}}$. The term (5.30d) can then be rewritten as

$$\beta e \rho \int d\mathbf{r}_4\, \mathbf{F}^{\mathrm{SR}}(\mathbf{r}_3 - \mathbf{r}_4)[\rho_{-+}(\mathbf{r}_1, \mathbf{r}_2)\, Q_{-+}(\mathbf{r}_1, \mathbf{r}_2 \mid \mathbf{r}_4)$$
$$- \rho^2 Q_-(\mathbf{r}_1 \mid \mathbf{r}_4) - \rho^2 Q_+(\mathbf{r}_2 \mid \mathbf{r}_4)] \quad (5.31\text{a})$$
$$+ \beta e \rho \int d\mathbf{r}_4\, \mathbf{F}_{\mathrm{C}}(\mathbf{r}_3 - \mathbf{r}_4)[\rho_{-+}(\mathbf{r}_1, \mathbf{r}_2)\, Q_{-+}(\mathbf{r}_1, \mathbf{r}_2 \mid \mathbf{r}_4)$$
$$- \rho^2 Q_-(\mathbf{r}_1 \mid \mathbf{r}_4) - \rho^2 Q_+(\mathbf{r}_2 \mid \mathbf{r}_4)] \quad (5.31\text{b})$$

Since $\mathbf{F}^{\mathrm{SR}}$ has a compact support, (5.31a) decays as

$$[\rho_{-+}(\mathbf{r}_1, \mathbf{r}_2)\, Q_{-+}(\mathbf{r}_1, \mathbf{r}_2 \mid \mathbf{r}_3) - \rho^2 Q_-(\mathbf{r}_1 \mid \mathbf{r}_3) - \rho^2 Q_+(\mathbf{r}_2 \mid \mathbf{r}_3)]$$

i.e., faster than $1/r_3^3$ by virtue of the bounds (5.12a), (5.12c). The quantity (5.31b) is proportional to the Coulomb field at $\mathbf{r}_3$ created by the localized charge distribution

$$\rho_{-+}(\mathbf{r}_1, \mathbf{r}_2)\, Q_{-+}(\mathbf{r}_1, \mathbf{r}_2 \mid \mathbf{r}_4) - \rho^2 Q_-(\mathbf{r}_1 \mid \mathbf{r}_4) - \rho^2 Q_+(\mathbf{r}_2 \mid \mathbf{r}_4) \quad (5.32)$$

Therefore its asymptotic expansion for r_3 large is given by the familiar multipolar expansion with respect to the successive multipoles of (5.32), plus a remainder $\mathbf{E}^{(R)}_{-+}(\mathbf{r}_1, \mathbf{r}_2 | \mathbf{r}_3)$ which obeys the Poisson equation

$$\nabla_3 \cdot \mathbf{E}^{(R)}_{-+}(\mathbf{r}_1, \mathbf{r}_2 | \mathbf{r}_3) \tag{5.33}$$
$$= 2\pi\beta e\rho[\rho_{-+}(\mathbf{r}_1, \mathbf{r}_2)\, Q_{-+}(\mathbf{r}_1, \mathbf{r}_2 | \mathbf{r}_3) - \rho^2 Q_-(\mathbf{r}_1 | \mathbf{r}_3) - \rho^2 Q_+(\mathbf{r}_2 | \mathbf{r}_3)]$$

The bounds (5.12a), (5.12c) imply that $\mathbf{E}^{(R)}_{-+}(\mathbf{r}_1, \mathbf{r}_2 | \mathbf{r}_3)$ decays faster than $1/r_3^3$. In the multipole expansion, the monopole $1/r_3$ term vanishes because of the perfect screening sum rules (5.13). However, the dipole $1/r_3^2$ term does not vanish in general, because the dipole of the charge distribution (5.32) reduces to $\rho_{-+}(\mathbf{r}_1, \mathbf{r}_2)\, \mathbf{p}_{-+}(\mathbf{r}_1, \mathbf{r}_2)$. Consequently, we find that (5.30d) behaves as $(\hat{\mathbf{r}}_3 \equiv \mathbf{r}_3/r_3)$

$$-\beta e\rho\rho_{-+}(\mathbf{r}_1, \mathbf{r}_2)\, \frac{[\mathbf{p}_{-+}(\mathbf{r}_1, \mathbf{r}_2) - 2\hat{\mathbf{r}}_3 \cdot \mathbf{p}_{-+}(\mathbf{r}_1, \mathbf{r}_2)\, \hat{\mathbf{r}}_3]}{r_3^2} \tag{5.34}$$

when $r_3 \to \infty$ with $(\mathbf{r}_1, \mathbf{r}_2)$ fixed.

The leading $1/r_3^2$ term (5.34) of the asymptotic expansion of (5.30d) must be compensated by another $1/r_3^2$ contribution arising either from the lhs of (5.30) or from the quantities (5.30a)–(5.30d). Taking into account the bounds (5.12a), (5.12c), we immediately see[9] that the lhs of (5.30) as well as (5.30a) decay faster than $1/r_3^4$. The asymptotic behavior of the two-body mean-field terms (5.30b), (5.30c) can be studied like that of (5.30d). Since $Q_+(\mathbf{r}_2 | \mathbf{r}_4)$ and $Q_-(\mathbf{r}_2 | \mathbf{r}_4)$ do not carry any monopole or dipole, these terms decay faster than $1/r_3^5$. Thus, the sole $1/r_3^2$ contribution which can cancel (5.34) arises from the four-body term (5.30e), i.e.,

$$e \int d\mathbf{r}_4\, \mathbf{F}(\mathbf{r}_3 - \mathbf{r}_4)[\rho^T_{-+++}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_2, \mathbf{r}_4) - \rho^T_{-++-}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4)]$$
$$\sim -\rho\rho_{-+}(\mathbf{r}_1, \mathbf{r}_2)\, \frac{\mathbf{p}_{-+}(\mathbf{r}_1, \mathbf{r}_2) - 2[\hat{\mathbf{r}}_3 \cdot \mathbf{p}_{-+}(\mathbf{r}_1, \mathbf{r}_2)]\, \hat{\mathbf{r}}_3}{r_3^2} \tag{5.35}$$

when $r_3 \to \infty$, $(\mathbf{r}_1, \mathbf{r}_2)$ being kept fixed.

The above analysis can be repeated for the BGY equations relative to $\rho^T_{-+-}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)$, $\rho^T_{+++}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)$ and $\rho^T_{++-}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)$. Similarly to (5.35), we find

$$e \int d\mathbf{r}_4\, \mathbf{F}(\mathbf{r}_3 - \mathbf{r}_4)[\rho^T_{-+-+}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4) - \rho^T_{-+--}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4)]$$
$$\sim -\rho\rho_{-+}(\mathbf{r}_1, \mathbf{r}_2)\, \frac{\mathbf{p}_{-+}(\mathbf{r}_1, \mathbf{r}_2) - 2[\hat{\mathbf{r}}_3 \cdot \mathbf{p}_{-+}(\mathbf{r}_1, \mathbf{r}_2)]\, \hat{\mathbf{r}}_3}{r_3^2} \tag{5.36}$$

[9] For this, we implicitly assume that the large-distance expansions of the particle correlations do not involve oscillating terms like $(\cos r^\mu)/r^\nu$ with $\nu - \mu \leqslant 3$.

whereas, since $\mathbf{p}_{++}(\mathbf{r}_1, \mathbf{r}_2) = \mathbf{0}$,

$$e\int d\mathbf{r}_4\, \mathbf{F}(\mathbf{r}_3 - \mathbf{r}_4)[\rho^T_{++++}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4) - \rho^T_{+++-}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4)] = o(1/r_3^2) \tag{5.37}$$

and

$$e\int d\mathbf{r}_4\, \mathbf{F}(\mathbf{r}_3 - \mathbf{r}_4)[\rho^T_{++-+}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4) - \rho^T_{++--}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4)] = o(1/r_3^2) \tag{5.38}$$

From the asymptotic behaviors (5.35), (5.36), it is very natural to guess that there exist four-particle correlations which only decay as $1/r^2$ for some specific configurations of $\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4$. In fact, we conjecture that such correlations and configurations correspond to two far away neutral pairs, i.e., one should have ($\hat{\mathbf{R}} \equiv \mathbf{R}/R$)

$$\rho^T_{-+-+}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4) \sim \frac{D_{-+,-+}(\mathbf{r}_1 - \mathbf{r}_2, \mathbf{r}_3 - \mathbf{r}_4; \hat{\mathbf{R}})}{R^2} \tag{5.39}$$

when $\mathbf{R} = (\mathbf{r}_3 + \mathbf{r}_4 - \mathbf{r}_1 - \mathbf{r}_2)/2$ is sent to infinity, $(\mathbf{r}_1 - \mathbf{r}_2)$ and $(\mathbf{r}_3 - \mathbf{r}_4)$ being kept fixed. All the other four-particle correlations or configurations should lead to decays faster than $1/r^2$. This conjecture is compatible with the exact behaviors (5.35)–(5.38) and with the term-by-term analysis of the low-fugacity expansions. Therefore we believe that (5.39) is the true behavior in the whole dielectric phase, although this form cannot be derived in a rigorous deductive way from the exact results (5.35)–(5.38).

The function $D_{-+,-+}$, coefficient of the $1/R^2$ term in (5.39), obviously satisfies the symmetry relations

$$D_{-+,-+}(\mathbf{t}', \mathbf{t}''; \hat{\mathbf{R}}) = D_{-+,-+}(\mathbf{t}'', \mathbf{t}'; -\hat{\mathbf{R}}) \tag{5.40a}$$

$$D_{-+,-+}(-\mathbf{t}', -\mathbf{t}''; \hat{\mathbf{R}}) = D_{-+,-+}(\mathbf{t}', \mathbf{t}''; \hat{\mathbf{R}}) \tag{5.40b}$$

$$D_{-+,-+}(\mathbf{t}', -\mathbf{t}''; \hat{\mathbf{R}}) = -D_{-+,-+}(\mathbf{t}', \mathbf{t}''; \hat{\mathbf{R}}) \tag{5.40c}$$

where $e\mathbf{t}' = e(\mathbf{r}_2 - \mathbf{r}_1)$ and $e\mathbf{t}'' = e(\mathbf{r}_4 - \mathbf{r}_3)$ are the dipoles carried by the pairs $\mathscr{P}' = \{\oplus\, \mathbf{r}_2, \ominus\, \mathbf{r}_1\}$ and $\mathscr{P}'' = \{\oplus\, \mathbf{r}_4, \ominus\, \mathbf{r}_3\}$. Furthermore, we easily obtain from (5.36)

$$e\int d\mathbf{t}''\, \mathbf{F}(\mathbf{t}'')\, D_{-+,-+}(\mathbf{t}', \mathbf{t}''; \hat{\mathbf{R}}) = -\rho\rho_{-+}(t')[\mathbf{p}_{-+}(\mathbf{t}') - 2(\hat{\mathbf{R}} \cdot \mathbf{p}_{-+}(\mathbf{t}'))\hat{\mathbf{R}}] \tag{5.41}$$

Multiplying each side of (5.41) by $\mathbf{F}(\mathbf{t}')$ and integrating over $\mathbf{t}'$, we get

$$\int d\mathbf{t}'\, d\mathbf{t}''\, \mathbf{F}(\mathbf{t}') \cdot \mathbf{F}(\mathbf{t}'')\, D_{-+,-+}(\mathbf{t}', \mathbf{t}''; \hat{\mathbf{R}}) = 0 \tag{5.42}$$

which follows from the colinearity of $\mathbf{F}(\mathbf{t}')$ with $\mathbf{p}_{-+}(\mathbf{t}')$. On the other hand, a first projection of (5.41) over $\hat{\mathbf{R}}$, followed by the multiplication of the resulting equality by $\hat{\mathbf{R}} \cdot \mathbf{F}(\mathbf{t}')$ and an integration over $\mathbf{t}'$, leads to

$$\int d\mathbf{t}'\, d\mathbf{t}''\, [\hat{\mathbf{R}} \cdot \mathbf{F}(\mathbf{t}')][\hat{\mathbf{R}} \cdot \mathbf{F}(\mathbf{t}'')]\, D_{-+,-+}(\mathbf{t}', \mathbf{t}''; \hat{\mathbf{R}}) = \frac{\rho^2}{\beta e^2 \varepsilon} \tag{5.43}$$

In deriving (5.43), we have also used the relation (5.28). The identities (5.41)–(5.43) are expected to be valid at any finite density in the dielectric phase. In Appendix C, we check that the identity (5.41) is indeed satisfied at the order z^6 included for $D_{-+,-+}$ [the identities (5.42) and (5.43), which are immediate consequences of (5.41), are then also satisfied].

The above $1/R^2$ decays can be interpreted as resulting from dipole–dipole interactions. However, we stress that the precise form of (5.39) cannot be rewritten as the dipole–dipole potential between the two pairs $\mathscr{P}'$ and $\mathscr{P}''$ renormalized by the multiplicative factor $1/\varepsilon$, even near the zero-density critical point [see the expression (C.8) for $D^{(6)}_{-+,-+}$]. Thus, the intrinsic correlations between two far away dipoles of the medium do not behave as the effective correlations between infinitesimal external dipoles immersed in the medium. This confirms that the $1/r^{\Gamma/\varepsilon}$ decay of $C(r)$ is very peculiar to this quantity and to the limit $\Gamma \to 4^+$.

5.3. A Plausible Decay Scenario

In the previous subsection we gave strong arguments indicating that ρ^T_{-+-+} decays algebraically as $1/r^2$ when two neutral pairs are separated by a large distance r. In fact, all the particle correlations should decay as power laws in the dielectric phase, as suggested by the analysis of the low-fugacity expansions. Here, we present an algebraic decay scenario for the two- and three-particle correlations inspired from this analysis. This scenario is shown to be consistent with the asymptotic structure of the BGY equations (5.8). Furthermore, we discuss the hypotheses of the conditional theorem of Alastuey and Martin,[30] which are violated in the present scenario.

The two-particle correlations should decay as the square of the dipole–dipole potential (see Section 3), i.e.,

$$\rho^T_{++}(\mathbf{r}_1, \mathbf{r}_2) \sim \frac{B_{+,+}}{r_{12}^4} \tag{5.44a}$$

$$\rho^T_{+-}(\mathbf{r}_1, \mathbf{r}_2) \sim \frac{B_{+,-}}{r_{12}^4} \tag{5.44b}$$

when $r_{12} \to \infty$, where the constant coefficients $B_{+,+}$ and $B_{+,-}$ are identical. Consequently, the charge correlation $C(r_{12})$ decays faster than $1/r_{12}^4$, i.e.,

$$C(r_{12}) \sim \frac{C_0}{r_{12}^{p_0}}, \qquad r_{12} \to \infty \tag{5.45}$$

where C_0 is a constant coefficient and the power p_0 is strictly larger than 4. When one particle is sent to infinity, the three-particle correlations should decay like $1/r^4$. Typically,

$$\rho^T_{+++}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) \sim \frac{B_{++,+}(\mathbf{r}_1 - \mathbf{r}_3; \hat{\mathbf{r}}_2)}{r_2^4} \tag{5.46}$$

when $r_2 \to \infty$, with $\mathbf{r}_1$, $\mathbf{r}_3$, $\hat{\mathbf{r}}_2$ kept fixed, and the other three-particle correlations have similar behaviors. When the three relative distances r_{12}, r_{13}, r_{23} become large, these three-particle correlations should decay as the product $1/(r_{12}^2 r_{13}^2 r_{23}^2)$ of the dipole–dipole potentials. For instance,

$$\rho^T_{+++}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) \sim \frac{D_{+,+,+}(\theta_1, \theta_2, \theta_3)}{r_{12}^2 r_{13}^2 r_{23}^2} \tag{5.47}$$

when $r_{12}, r_{13}, r_{23} \to \infty$, the angles θ_1, θ_2, θ_3 of the triangle $(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)$ being kept fixed. Finally, there exist uniform bounds of the kind

$$|\rho^T_{+++}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)| < \frac{M(r)}{r_{12}^4} \tag{5.48}$$

with $r = \text{Inf}(\mathbf{r}_{13}, \mathbf{r}_{32})$ and $M(r)$ a bounded function which goes to zero as $r \to \infty$. Note that the behaviors (5.46), (5.47) are compatible with the bound (5.48).

The large-distance behaviors of the various terms of the BGY equation (5.8a) for $\rho^T_{++}(\mathbf{r}_1, \mathbf{r}_2)$ are now determined from the above decay scenario. Assuming that the corrections to the leading term (5.44a) in the asymptotic expansion of ρ^T_{++} are monotonic, we see that the lhs of (5.8a) behaves as

$$\nabla_2 \rho^T_{++}(\mathbf{0}, \mathbf{r}_2) \sim -\frac{4B_{+,+}}{r_2^5} \hat{\mathbf{r}}_2 \tag{5.49}$$

when $r_2 \to \infty$ with $\mathbf{r}_1 = \mathbf{0}$ fixed. In the rhs of (5.8a), the behavior of the self-term is obvious,

$$\beta e^2 \mathbf{F}(\mathbf{r}_2)\, \rho^T_{++}(\mathbf{0}, \mathbf{r}_2) \sim \frac{\beta e^2 B_{+,+}}{r_2^5} \hat{\mathbf{r}}_2, \qquad r_2 \to \infty \tag{5.50}$$

The mean-field term proportional to the electric field created at $\mathbf{r}_2$ by the charge distribution $Q_+(\mathbf{0}|\mathbf{r}_3)$ can be computed from Newton's theorem, since $Q_+(\mathbf{0}|\mathbf{r}_3)$ is invariant under rotations. This gives

$$\beta e \rho^2 \int d\mathbf{r}_3 \, \mathbf{F}(\mathbf{r}_2-\mathbf{r}_3)\, Q_+(\mathbf{0}|\mathbf{r}_3) \sim -\frac{\pi\beta\rho C_0}{(p_0-2)\, r_2^{p_0-1}}\hat{\mathbf{r}}_2, \qquad r_2\to\infty \tag{5.51}$$

where we have taken into account the relation (5.11), the perfect screening rule (5.13a), and the decay (5.45). The determination of the precise behavior of the three-body term would require additional assumptions on the decay of ρ^T_{+++} and ρ^T_{++-}, which cannot be easily guessed from the term-by-term analysis. However, Lemma L1 in ref. 30, whose hypotheses are satisfied by the uniform bound (5.48), can be applied here, with the result

$$\begin{aligned}\beta e^2 \int d\mathbf{r}_3 \, \mathbf{F}(\mathbf{r}_2-\mathbf{r}_3)[\rho^T_{+++}(\mathbf{0},\mathbf{r}_2,\mathbf{r}_3)-\rho^T_{++-}(\mathbf{0},\mathbf{r}_2,\mathbf{r}_3)]\\ = o(1/r_2^3), \qquad r_2\to\infty\end{aligned} \tag{5.52}$$

The large-distance behaviors (5.49)–(5.52) are consistent with the BGY equation (5.8a). Indeed, the multipolar $1/r_2^5$ behaviors (5.49), (5.50) should be compensated by the contributions to the three-body term of the regions $\mathbf{r}_3$ close to $\mathbf{0}$ and $\mathbf{r}_3$ close to $\mathbf{r}_2$, respectively. On the other hand, the $1/r_2^{p_0-1}$ behavior (5.51) should be compensated by contributions to the three-body term arising from the intermediate region between $\mathbf{0}$ and $\mathbf{r}_2$ [this cancellation is compatible with the decay (5.52), since p_0-1 is strictly larger than 3].

The algebraic decay scenario corresponding to Eqs. (5.44)–(5.48) must violate at least one of the hypotheses of a conditional theorem which precisely excludes monotonic power-law behaviors in a multicomponent Coulomb fluid.[30] This theorem specifies that if there exists some real power $\nu > 2$ such that

(i) $\rho^T_{s_1 s_2}(\mathbf{r}_1,\mathbf{r}_2) \sim A_{s_1 s_2}/r_2^\nu, \qquad r_2\to\infty$

(ii) $\rho^T_{s_1 s_2 s_3}(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3) < M(r)/r_2^\nu, \qquad r_2\to\infty$

(iii) $s_1 s_2 A_{s_1 s_2} < 0$

then the constants $A_{s_1 s_2}$ necessarily vanish whatever ν is. The clustering assumptions (i) and (ii) with $\nu=4$ are satisfied by the above decay scenario. However, the assumption (iii), which stipulates that ρ^T_{++} and ρ^T_{+-} have opposite signs at large distances, is not fulfilled here, since ρ^T_{++} and ρ^T_{+-} become in fact identical when $r_2\to\infty$ ($B_{+,+}=B_{+,-}$). This assumption is crucial for the derivation of the conditional theorem, because it implies that the charge correlation decays like the particle correlations: the mean-field term in the BGY equation for $\rho^T_{s_1 s_2}(\mathbf{0}, \mathbf{r}_2)$ then behaves as $1/r_2^{\nu-1}$ when $\mathbf{r}_2 \to \infty$ and cannot be compensated by any other term. In the present scenario, the charge correlation decays faster than the particle ones, and this allows cancellations between the mean-field and three-body terms. Physically, the hypothesis (iii) amounts to assuming that the sign of the particle correlations is determined by the electrostatic attraction or repulsion between the considered charges. This should be true at high temperatures, but no longer holds in the dielectric phase, where the formation of neutral molecules is the crucial phenomenon.

The above algebraic decay scenario can be extended to the four- and higher-order particle correlations. When one particle is sent to infinity, these correlations always decay as $1/r^4$. When two groups of two or more particles are separated by a large distance r, the corresponding correlations decay as $1/r^2$ if each group carries a dipole, as $1/r^3$ if one of these dipoles vanishes, or as $1/r^4$ if both vanish. The slower decay of the particle correlations for these specific configurations is essential for ensuring the consistency of the whole algebraic scenario with the full BGY hierarchy. In particular, in the BGY equation relative to $\nabla_n \rho^T_{s_1 \cdots s_n}(\mathbf{r}_1, ..., \mathbf{r}_n)$, the multipolar field created at $\mathbf{r}_n$ by the charge distribution $Q_{s_1 \cdots s_{n-1}}(\mathbf{r}_1, ..., \mathbf{r}_{n-1} \mid \mathbf{r}_{n+1})$ is compensated by the contribution to the integral term,

$$\int d\mathbf{r}_{n+1}\, \mathbf{F}(\mathbf{r}_n - \mathbf{r}_{n+1})[\rho^T_{s_1 \cdots s_n +}(\mathbf{r}_1, ..., \mathbf{r}_{n+1}) - \rho^T_{s_1 \cdots s_n -}(\mathbf{r}_1, ..., \mathbf{r}_{n+1})]$$

of the region $\mathbf{r}_{n+1}$ close to $\mathbf{r}_n$ (this cancellation is studied in detail for $n = 3$ in the preceding subsection).

6. CONCLUSION

Resumming the usual low-fugacity expansions near the zero-density critical point, we have calculated the large-distance expansion of the charge correlations in the dielectric phase as a sum of inverse power laws. Our first-principles approach allows us to retrieve some predictions of the RG. In particular, the exponent of the leading term in the above expansion is found to be the coupling constant Γ renormalized by the dielectric constant ε, in agreement with the conjectured value of the effective coupling constant at the fixed point of the RG flow equations (the latter has to be identified with the above critical exponent). Moreover, we give a precise mathematical foundation to the physical idea of screening of a large pair by smaller pairs introduced by Kosterlitz and Thouless in the heuristic iterated mean-field theory. We also have studied the large-distance behavior of the particle correlations in the dielectric phase at finite densities. A term-by-term analysis of the low-fugacity expansions combined with a survey of the BGY hierarchy equations provides a plausible algebraic decay scenario very similar[10] to what happens in a system of permanent dipoles. We stress that the four- and higher-order particle correlations may decay slower than the two-particle ones for some specific configurations, typically as slow as the $1/r^2$ dipole–dipole interaction potential. This slower decay of the higher-order particle correlations is essential for ensuring the consistency of the algebraic scenario with the BGY hierarchy.

Our analysis of the particle and charge correlations is of course no longer valid in the conductive phase, where an exponential clustering is expected. For instance, the nested-pair resummation processes near the zero-density critical point break down for $8\pi z > \Gamma - 4$, as signaled by the corresponding divergences of the quantities of interest. At the transition line $8\pi z = \Gamma - 4$, the coefficients of the inverse power laws involved in the large-distance expansion of $C(r)$ vanish: this announces the exponential decay of $C(r)$ in the conductive phase. A precise study of the latter behavior would require resummations of the low-fugacity expansions "à la Debye–Hückel." These resummations should be much more intricate than the usual resummation of the chain diagrams valid in the weak-coupling regime[32] because the charges are strongly correlated near the zero-density critical point however small their density is. Heuristic theories[15,33] provide estimations for the divergent behavior of the correlation length when the transition line is approached from the conductive phase. They rely on the physical idea that the effective density of free charges goes to zero in this limit (almost all the positive and negative charges are bound in neutral molecules). A precise formulation of this idea in a rigorous statistical description should be a key step in the systematic resummations near the transition line.

The KT transition characterizes a change in the formal linear response of the infinite system to infinitesimal external charges. This change corresponds to different large-distance behavior of the translationally invariant intrinsic correlations and allows an unambiguous definition of both dielectric and conductive phases. However, a natural question concerns the status of the KT dielectric phase with respect to the usual macroscopic definition of a dielectric material. In particular, does the total

[10] It turns out that this kind of algebraic decay scenario also should be observed in quantum Coulomb fluids for any values of the thermodynamic parameters, as shown by Alastuey and Martin.[31] In the quantum case, the existence of algebraic tails in the equilibrium particle correlations is linked to intrinsic quantum fluctuations and does not require the formation of neutral entities.

polarization of a large finite sample submitted to a weak external electric field behave in the thermodynamic limit as predicted by the laws of macroscopic electrostatics[11]? In fact, the validity of such laws is ensured by a very peculiar behavior of the finite-volume correlations near the boundaries of the sample, as shown by Choquard *et al.*[(34)] These authors start from the microscopic representation of the dielectric susceptibility tensor and determine its behavior in the thermodynamic limit for some simple models where explicit expressions of the correlations are available. Such an analysis applied to the present system should provide a better understanding of the macroscopic electrical properties of the KT dielectric phase.

A special version of the CG where one species (the "ions") is fixed at the sites of a lattice has been studied by molecular-dynamics simulations.[(35)] At small densities, this model exhibits a KT transition near $\Gamma = 4$. This can be shown[(36)] by rephrasing the original thermodynamic argument by Kosterlitz and Thouless[(7)] or by using a "spin-block" method. Our resummation techniques could be applied to this model in the canonical ensemble. We expect the charge and two-body "electron" correlations to have similar decays as those found here, when these correlations are averaged over the cells.[(36)] Without any average, the electron–electron correlation is the analog of ρ^T_{-+-+} and should decay as $1/r^2$. At high densities the KT transition should become a ferroelectric one.[(37)]

In the conductive phase, all the particle correlations have the same kind of exponential decay at large distances. This phase can then be qualitatively described by approximate theories based on the familiar closures of either the BBGKY or the BGY hierarchy. For instance, the well-known Vlasov approximation has been applied[(38)] to the fixed-ion version of the CG. Such theories fail in reproducing the appearance of the KT dielectric phase at low temperatures because they automatically exclude possible slower decays of the higher-order correlations.

Finally, we mention the existence of various solvable one-dimensional models with logarithmic interactions.[(39)] These models undergo transitions of the KT type characterized by a qualitative change in the large-distance behavior of the correlations (however, in both high- and low-temperature phases, the decay of the latter is algebraic). The study of these transitions within the RG techniques or the methods introduced in the present paper should usefully complete some available explicit exact results.[(39)]

[11] Here we only consider the linear response of the sample. However, the true response might be seriously affected by the following ionization mechanism, which is not taken into account by a first-order perturbative treatment of the external field. Indeed, however small the latter is, it is sufficient to break large, weakly-bound neutral pairs. The corresponding ionized free charges should prevent the sample from behaving as a truly insulating and dielectric material.

APPENDIX A

In this appendix, we extract the part $C_\varepsilon^{(4)}(r)$ of $C^{(4)}(r)$ which gives contribution to $1/\varepsilon$ of order $z^4/(\Gamma-4)^3$ in the limit $\Gamma \to 4^+$. For this, we start from the integral expression (4.12) of $C^{(4)}(r)$, which is first rewritten as the sum of the conditionally convergent integral

$$e^2 \frac{z^4}{\sigma^4}\left(\frac{\sigma}{r}\right)^\Gamma \int^* d\mathbf{v}_1\, d\mathbf{v}_2 \left\{\frac{\Gamma}{|\mathbf{v}_1-\mathbf{v}_2|^\Gamma}(\mathbf{v}_1-\mathbf{v}_2)\cdot\left[\frac{\mathbf{v}_2}{v_2^2}-\frac{(\mathbf{v}_2-\mathbf{r}/\sigma)}{(\mathbf{v}_2-\mathbf{r}/\sigma)^2}\right] - \frac{\Gamma}{|\mathbf{v}_1-\mathbf{v}_2|^\Gamma}(\mathbf{v}_1-\mathbf{v}_2)\cdot\left[\frac{\mathbf{v}_1}{v_1^2}-\frac{(\mathbf{v}_1-\mathbf{r}/\sigma)}{(\mathbf{v}_1-\mathbf{r}/\sigma)^2}\right]\right\} \tag{A.1}$$

plus the two absolutely convergent integrals

$$e^2 \frac{z^4}{\sigma^4}\left(\frac{\sigma}{r}\right)^\Gamma \int^* d\mathbf{v}_1\, d\mathbf{v}_2 \left\{-\frac{\Gamma^2}{2\,|\mathbf{v}_1-\mathbf{v}_2|^\Gamma}\left[(\mathbf{v}_1-\mathbf{v}_2)\cdot\left(\frac{\mathbf{v}_1}{v_1^2}-\frac{(\mathbf{v}_1-\mathbf{r}/\sigma)}{(\mathbf{v}_1-\mathbf{r}/\sigma)^2}\right)\right]^2 - \frac{\Gamma^2}{2\,|\mathbf{v}_1-\mathbf{v}_2|^\Gamma}\left[(\mathbf{v}_1-\mathbf{v}_2)\cdot\left(\frac{\mathbf{v}_2}{v_2^2}-\frac{(\mathbf{v}_2-\mathbf{r}/\sigma)}{(\mathbf{v}_2-\mathbf{r}/\sigma)^2}\right)\right]^2\right\} \tag{A.2}$$

and

$$\begin{aligned}
&e^2 \frac{z^4}{\sigma^4}\left(\frac{\sigma}{r}\right)^\Gamma \int^* d\mathbf{v}_1\, d\mathbf{v}_2 \\
&\times\left\{\left(\frac{r}{\sigma}\right)^{2\Gamma} \frac{|\mathbf{v}_1-\mathbf{v}_2|^\Gamma}{v_1^\Gamma v_2^\Gamma\, |\mathbf{v}_1-\mathbf{r}/\sigma|^\Gamma\, |\mathbf{v}_2-\mathbf{r}/\sigma|^\Gamma}\right. \\
&-\frac{v_1^\Gamma\, |\mathbf{v}_2-\mathbf{r}/\sigma|^\Gamma}{v_2^\Gamma\, |\mathbf{v}_1-\mathbf{v}_2|^\Gamma\, |\mathbf{v}_1-\mathbf{r}/\sigma|^\Gamma} \\
&-\frac{v_2^\Gamma\, |\mathbf{v}_1-\mathbf{r}/\sigma|^\Gamma}{v_1^\Gamma\, |\mathbf{v}_1-\mathbf{v}_2|^\Gamma\, |\mathbf{v}_2-\mathbf{r}/\sigma|^\Gamma} \\
&+\frac{2}{|\mathbf{v}_1-\mathbf{v}_2|^\Gamma} \\
&+\frac{\Gamma}{|\mathbf{v}_1-\mathbf{v}_2|^\Gamma}(\mathbf{v}_1-\mathbf{v}_2)\cdot\left[\frac{\mathbf{v}_1}{v_1^2}-\frac{(\mathbf{v}_1-\mathbf{r}/\sigma)}{(\mathbf{v}_1-\mathbf{r}/\sigma)^2}\right] \\
&-\frac{\Gamma}{|\mathbf{v}_1-\mathbf{v}_2|^\Gamma}(\mathbf{v}_1-\mathbf{v}_2)\cdot\left[\frac{\mathbf{v}_2}{v_2^2}-\frac{(\mathbf{v}_2-\mathbf{r}/\sigma)}{(\mathbf{v}_2-\mathbf{r}/\sigma)^2}\right] \\
&+\frac{\Gamma^2}{2\,|\mathbf{v}_1-\mathbf{v}_2|^\Gamma}\left[(\mathbf{v}_1-\mathbf{v}_2)\cdot\left(\frac{\mathbf{v}_1}{v_1^2}-\frac{(\mathbf{v}_1-\mathbf{r}/\sigma)}{(\mathbf{v}_1-\mathbf{r}/\sigma)^2}\right)\right]^2 \\
&\left.+\frac{\Gamma^2}{2\,|\mathbf{v}_1-\mathbf{v}_2|^\Gamma}\left[(\mathbf{v}_1-\mathbf{v}_2)\cdot\left(\frac{\mathbf{v}_2}{v_2^2}-\frac{(\mathbf{v}_2-\mathbf{r}/\sigma)}{(\mathbf{v}_2-\mathbf{r}/\sigma)^2}\right)\right]^2\right\}
\end{aligned} \tag{A.3}$$

where the notation $\int^*$ means that the integration domain for $\mathbf{v}_1$ and $\mathbf{v}_2$ excludes the configurations such that any of the relative distances between the points $\mathbf{0}$, $\mathbf{r}/\sigma$, $\mathbf{v}_1$, $\mathbf{v}_2$ is smaller than 1. It turns out that the integral (A.1) vanishes, as shown by the transformation of variables $(\mathbf{v}_1, \mathbf{v}_2) \to (\mathbf{v}_1 - \mathbf{r}/\sigma, \mathbf{v}_2 - \mathbf{r}/\sigma)$, which transforms the integrand into its opposite. In order to calculate the contributions of (A.2) and (A.3) to $C_\varepsilon^{(4)}(r)$, we proceed as follows. We first integrate over $\mathbf{v}_2$ at fixed $\mathbf{v}_1$ and we split the integration domains into various regions. For each region, we extract the possible part of the corresponding integrand which gives contributions to $C_\varepsilon^{(4)}(r)$. The remaining part is then uniformly bounded over the considered region by functions which may also depend on r and Γ. This provides uniform bounds with respect to r and Γ (in a finite neighborhood of $\Gamma = 4$) for the contribution to $C^{(4)}(r)$ of this remaining part. These uniform bounds are used to prove that the latter does not contribute to $C_\varepsilon^{(4)}(r)$, and that furthermore $C_\varepsilon^{(4)}(r)$ indeed reduces to the first terms of the large-distance expansion of $C^{(4)}(r)$, where the involved coefficients are replaced by their limit forms when $\Gamma \to 4^+$. The above method is first applied to the integral (A.2).

The integral (A.2) can be rewritten as

$$-\frac{e^2 z^4}{\sigma^4}\left(\frac{\sigma}{r}\right)^\Gamma \Gamma^2 \int_{\mathbf{v}_1, |\mathbf{v}_1 - \mathbf{r}/\sigma| > 1} d\mathbf{v}_1 \int_{|\mathbf{v}_1 - \mathbf{v}_2|, |\mathbf{v}_2 - \mathbf{r}/\sigma|, v_2 > 1} d\mathbf{v}_2 \frac{1}{|\mathbf{v}_1 - \mathbf{v}_2|^\Gamma}$$
$$\times \left[(\mathbf{v}_1 - \mathbf{v}_2) \cdot \left(\frac{\mathbf{v}_1}{v_1^2} - \frac{(\mathbf{v}_1 - \mathbf{r}/\sigma)}{(\mathbf{v}_1 - \mathbf{r}/\sigma)^2} \right) \right]^2$$

$$= -\frac{e^2 z^4}{\sigma^4}\left(\frac{\sigma}{r}\right)^\Gamma \left(\frac{\pi \Gamma^2}{\Gamma - 4}\right) \int_{v_1, |\mathbf{v}_1 - \mathbf{r}/\sigma| > v_0} d\mathbf{v}_1$$
$$\times \left[\frac{\mathbf{v}_1}{v_1^2} - \frac{(\mathbf{v}_1 - \mathbf{r}/\sigma)}{(v_1 - \mathbf{r}/\sigma)^2} \right]^2 \tag{A.4a}$$

$$+ 2\frac{e^2 z^4}{\sigma^4}\left(\frac{\sigma}{r}\right)^\Gamma \Gamma^2 \int_{v_1, |\mathbf{v}_1 - \mathbf{r}/\sigma| > v_0} d\mathbf{v}_1 \int_{u < 1} d\mathbf{u} \frac{1}{|\mathbf{v}_1 - \mathbf{u}|^\Gamma}$$
$$\times \left[(\mathbf{v}_1 - \mathbf{u}) \cdot \left(\frac{\mathbf{v}_1}{v_1^2} - \frac{(\mathbf{v}_1 - \mathbf{r}/\sigma)}{(\mathbf{v}_1 - r/\sigma)^2} \right) \right]^2 \tag{A.4b}$$

$$- 2\frac{e^2 z^4}{\sigma^4}\left(\frac{\sigma}{r}\right)^\Gamma \Gamma^2 \int_{1 < v_1 < v_0} d\mathbf{v}_1 \int_{|\mathbf{v}_1 - \mathbf{v}_2|, |\mathbf{v}_2 - \mathbf{r}/\sigma|, v_2 > 1} d\mathbf{v}_2 \frac{1}{|\mathbf{v}_1 - \mathbf{v}_2|^\Gamma}$$
$$\times \left[(\mathbf{v}_1 - \mathbf{v}_2) \cdot \left(\frac{\mathbf{v}_1}{v_1^2} - \frac{(\mathbf{v}_1 - \mathbf{r}/\sigma)}{(\mathbf{v}_1 - \mathbf{r}/\sigma)^2} \right) \right]^2 \tag{A.4c}$$

for a given $v_0 > 2$ and r/σ sufficiently large ($r/\sigma > 2v_0$). The integral (A.4a) is evaluated through an integration by parts and the use of the Poisson equation as

$$2\frac{e^2z^4}{\sigma^4}\left(\frac{\sigma}{r}\right)^{\Gamma}\left(\frac{\pi\Gamma^2}{\Gamma-4}\right)\int_0^{2\pi} d\theta_0\, \mathbf{v}_0\cdot\left[\frac{\mathbf{v}_0}{v_0^2}-\frac{(\mathbf{v}_0-\mathbf{r}/\sigma)}{(\mathbf{v}_0-\mathbf{r}/\sigma)^2}\right]$$
$$\times\left(\ln v_0-\ln\left|\mathbf{v}_0-\frac{\mathbf{r}}{\sigma}\right|\right) \qquad \text{(A.5)}$$

where θ_0 is the polar angle of $\mathbf{v}_0$ in a given frame. The integrand of (A.5) can be rewritten as $-\ln(r/\sigma)$ plus a term bounded by $\text{cst}\times(\sigma/r)\ln(r/\sigma)$ and a bounded function of $\mathbf{v}_0$. Thus, (A.5) is equal to

$$-64\pi^2e^2\frac{z^4}{\sigma^4}\left(\frac{1}{\Gamma-4}\right)\left(\frac{\sigma}{r}\right)^{\Gamma}\ln\left(\frac{r}{\sigma}\right) \qquad \text{(A.6)}$$

plus three terms bounded respectively by $\text{cst}\times z^4(\sigma/r)^{\Gamma}\ln(r/\sigma)$, $\text{cst}\times z^4(\sigma/r)^{\Gamma+1}\ln(r/\sigma)/(\Gamma-4)$, and $\text{cst}\times z^4(\sigma/r)^{\Gamma}/(\Gamma-4)$. Since the integral over $\mathbf{u}$ in (A.4b) is bounded by $\text{cst}/v_1^{\Gamma-2}$ and the integral over $\mathbf{v}_2$ in (A.4c) is bounded by $\text{cst}/(\Gamma-4)$, the quantities (A.4b) and (A.4c) are both bounded by $\text{cst}\times z^4(\sigma/r)^{\Gamma}/(\Gamma-4)$. Thus, the sole contribution to $C_\varepsilon^4(r)$ of (A.2) arises from (A.4a) and reduces to (A.6). Indeed, all the other terms give contributions to $1/\varepsilon$ which are bounded by $z^4/(\Gamma-4)^2$.

Now, we turn to the contribution of the integral (A.3). After the transformation of variable $\mathbf{v}_2=v_1\mathbf{w}$ in the integral over $\mathbf{v}_2$, (A.3) becomes

$$e^2\frac{z^4}{\sigma^4}\left(\frac{\sigma}{r}\right)^{\Gamma}\int_{v_1,\,|\mathbf{v}_1-\mathbf{r}/\sigma|>1} d\mathbf{v}_1\,\frac{1}{v_1^{\Gamma-2}}$$
$$\times\int_{w,\,|\mathbf{w}-\mathbf{v}_1|,\,|\mathbf{w}-\mathbf{r}/(\sigma v_1)|>1/v_1}\{f_a+f_b+\cdots+f_h\} \qquad \text{(A.7)}$$

with

$$f_a=\frac{(r/\sigma v_1)^{2\Gamma}}{|\hat{\mathbf{v}}_1-\mathbf{r}/\sigma v_1|^{\Gamma}}\frac{|\hat{\mathbf{v}}_1-\mathbf{w}|^{\Gamma}}{w^{\Gamma}\,|\mathbf{w}-\mathbf{r}/\sigma v_1|^{\Gamma}} \qquad \text{(A.8a)}$$

$$f_b=-\frac{1}{|\hat{\mathbf{v}}_1-\mathbf{r}/\sigma v_1|^{\Gamma}}\frac{|\mathbf{w}-\mathbf{r}/\sigma v_1|^{\Gamma}}{w^{\Gamma}\,|\hat{\mathbf{v}}_1-\mathbf{w}|^{\Gamma}} \qquad \text{(A.8b)}$$

$$f_c=-\left|\hat{\mathbf{v}}_1-\frac{\mathbf{r}}{\sigma v_1}\right|^{\Gamma}\frac{\mathbf{w}^{\Gamma}}{|\hat{\mathbf{v}}_1-\mathbf{w}|^{\Gamma}\,|\mathbf{w}-\mathbf{r}/\sigma v_1|^{\Gamma}} \qquad \text{(A.8c)}$$

$$f_d=\frac{2}{|\hat{\mathbf{v}}_1-\mathbf{w}|^{\Gamma}} \qquad \text{(A.8d)}$$

$$f_e=\frac{\Gamma}{|\hat{\mathbf{v}}_1-\mathbf{w}|^{\Gamma}}(\hat{\mathbf{v}}_1-\mathbf{w})\cdot\left(\mathbf{v}_1-\frac{\hat{\mathbf{v}}_1-\mathbf{r}/\sigma v_1}{|\hat{\mathbf{v}}_1-\mathbf{r}/\sigma v_1|^2}\right) \qquad \text{(A.8e)}$$

$$f_f = -\frac{\Gamma}{|\hat{\mathbf{v}}_1 - \mathbf{w}|^\Gamma} (\hat{\mathbf{v}}_1 - \mathbf{w}) \cdot \left(\frac{\mathbf{w}}{w^2} - \frac{\mathbf{w} - \mathbf{r}/\sigma v_1}{|\mathbf{w} - \mathbf{r}/\sigma v_1|^2} \right) \tag{A.8f}$$

$$f_g = \frac{\Gamma^2}{2} \frac{1}{|\hat{\mathbf{v}}_1 - \mathbf{w}|^\Gamma} \left[(\hat{\mathbf{v}}_1 - \mathbf{w}) \cdot \left(\hat{\mathbf{v}}_1 - \frac{\hat{\mathbf{v}}_1 - \mathbf{r}/\sigma v_1}{|\hat{\mathbf{v}}_1 - \mathbf{r}/\sigma v_1|^2} \right) \right]^2 \tag{A.8g}$$

$$f_h = \frac{\Gamma^2}{2} \frac{1}{|\hat{\mathbf{v}}_1 - \mathbf{w}|^\Gamma} \left[(\hat{\mathbf{v}}_1 - \mathbf{w}) \cdot \left(\frac{\mathbf{w}}{w^2} - \frac{\mathbf{w} - \mathbf{r}/\sigma v_1}{|\mathbf{w} - \mathbf{r}/\sigma v_1|^2} \right) \right]^2 \tag{A.8h}$$

In (A.7), the integration domain for $\mathbf{v}_1$ is split into the two disks with radius $r/(2\sigma)$ centered at $\mathbf{0}$ $[1 < v_1 < r/(2\sigma)]$ and $\mathbf{r}/\sigma$ $[1 < |\mathbf{v}_1 - \mathbf{r}/\sigma| < r/(2\sigma)]$, respectively, plus the region $(v_1, |\mathbf{v}_1 - \mathbf{r}/\sigma| > r/(2\sigma))$ outside these disks. For each given $\mathbf{v}_1$, the integration domain for $\mathbf{w}$ is split into three disks with arbitrary constant radius centered at $\mathbf{0}$ $(1/v_1 < w < \text{cst})$, $\mathbf{r}/\sigma v_1$ $(1/v_1 < |\mathbf{w} - \mathbf{r}/\sigma v_1| < \text{cst})$, and $\hat{\mathbf{v}}_1$ $(1/v_1 < |\mathbf{w} - \hat{\mathbf{v}}_1| < \text{cst})$, respectively, plus the region $(w, |\mathbf{w} - \hat{\mathbf{v}}_1|, |\mathbf{w} - \mathbf{r}/\sigma v_1| > \text{cst})$ outside these disks. The contributions of the various regions are now evaluated.

(1) $1 < v_1 < r/(2\sigma)$

(1.i) $1/v_1 < w < \text{cst}$

The functions f_c, f_d, f_e, and f_g remain bounded near $\mathbf{w} = \mathbf{0}$. The integral of f_f over the angle of $\mathbf{w}$ is also bounded. Furthermore, using a convergent multipolar expansion in powers of $\mathbf{w}$ and combining the harmonicity of the Coulomb potential with the rotational invariance of the weighting factor $1/w^\Gamma$, we find that the angular integral of $(f_a + f_b)$ reduces to a singular term bounded by $\text{cst}/w^{\Gamma-4}$ plus a bounded function of w. This implies that $\int_{1/v_1 < w < \text{cst}} d\mathbf{w} \, \{f_a + f_b \cdots + f_g\}$ is bounded. On the other hand, since f_h behaves as $1/w^2$ when $w \to 0$, $\int_{1/v_1 < w < \text{cst}} d\mathbf{w} \, f_h$ can be rewritten as a sum of the logarithmic singular term

$$\int_{1/v_1 < w < \text{cst}} d\mathbf{w} \, \frac{\Gamma^2}{2} \frac{(\hat{\mathbf{v}}_1 \cdot \mathbf{w})^2}{w^4} = \frac{\pi \Gamma^2}{2} \ln v_1 + \text{cst} \tag{A.9}$$

plus a bounded function of v_1. The contribution to $C_\varepsilon^4(r)$ of the considered region then entirely arises from the above logarithmic term and reads

$$\begin{aligned} e^2 \frac{z^4}{\sigma^4} \left(\frac{\sigma}{r}\right)^\Gamma & \int_{1 < v_1 < r/2\sigma} d\mathbf{v}_1 \, \frac{\pi \Gamma^2}{2} \frac{\ln v_1}{v_1^{\Gamma-2}} \\ &= -\pi^2 \Gamma^2 e^2 \frac{z^4}{\sigma^4} \left(\frac{\sigma}{r}\right)^\Gamma \frac{\partial}{\partial \Gamma} \int_1^{r/2\sigma} dv_1 \, \frac{1}{v_1^{\Gamma-3}} \\ &= 16\pi^2 e^2 \frac{z^4}{\sigma^4} \frac{1}{(\Gamma-4)^2} \left[\left(\frac{\sigma}{r}\right)^\Gamma - \left(\frac{\sigma}{r}\right)^{2\Gamma-4} \right] \\ &\quad - 16\pi^2 e^2 \frac{z^4}{\sigma^4} \frac{1}{\Gamma-4} \left(\frac{\sigma}{r}\right)^{2\Gamma-4} \ln\left(\frac{r}{\sigma}\right) \end{aligned} \tag{A.10}$$

In the last line of (A.10), we have omitted the terms arising from the replacements of the prefactor Γ^2 by 16 and of $2^{\Gamma-4}$ by 1 because they are bounded by $\mathrm{cst} \times z^4(\sigma/r)^{\Gamma}/(\Gamma-4)$, $\mathrm{cst} \times z^4(\sigma/r)^{2\Gamma-4}/(\Gamma-4)$, and $\mathrm{cst} \times z^4(\sigma/r)^{2\Gamma-4}\ln(r/\sigma)$, and consequently do not contribute to $C_\varepsilon^4(r)$. Moreover, the other contributions to $C^{(4)}(r)$ arising from $(f_a+f_b+\cdots+f_g)$ and from the integrable (at $\mathbf{w}=0$) part of f_h are bounded by $\mathrm{cst} \times z^4(\sigma/r)^{\Gamma}/(\Gamma-4)$.

(1.ii) $1/v_1 < |\mathbf{w} - \mathbf{r}/\sigma v_1| < \mathrm{cst}$

We set $\mathbf{w} = \mathbf{r}/\sigma v_1 + \mathbf{w}'$. The functions f_b, f_d, f_e, and f_g remain bounded near $\mathbf{w}' = \mathbf{0}$. The integral of f_f over the angle of $\mathbf{w}'$ is also bounded for w' small. An analysis similar to that of case (1.i) shows that the angular integral of (f_a+f_c) reduces to a singular term bounded by $\mathrm{cst}/(w')^{\Gamma-4}$ plus a bounded function of w'. Finally, the angular integral of f_h is the sum of a singular term bounded by $\mathrm{cst} \times (\sigma v_1/r)^{\Gamma-2}/w'^2$ plus a bounded function of w'. The part of (A.7) corresponding to the considered region can then be rewritten as the sum of two terms which are bounded by $\mathrm{cst} \times z^4(\sigma/r)^{\Gamma}/(\Gamma-4)$ and $\mathrm{cst} \times z^4(\sigma/r)^{\Gamma}\ln(r/\sigma)$, respectively. Thus, this region does not contribute to $C_\varepsilon^4(r)$.

(1.iii) $1/v_1 < |\mathbf{w} - \hat{\mathbf{v}}_1| < \mathrm{cst}$

We set $\mathbf{w} = \hat{\mathbf{v}}_1 + \mathbf{w}''$. The function f_a remains bounded near $\mathbf{w}'' = \mathbf{0}$. The integral of $(f_b + \cdots + f_h)$ over the angle of $\mathbf{w}''$ reduces to a singular term bounded by $\mathrm{cst}/(w'')^{\Gamma-4}$ plus a bounded function of w''. Consequently, there is no contribution to $C_\varepsilon^4(r)$, and the contribution to $C^4(r)$ is bounded by $\mathrm{cst} \times z^4(\sigma/r)^{\Gamma}/(\Gamma-4)$.

(1.iiii) w, $|\mathbf{w} - \hat{\mathbf{v}}_1|$, $|\mathbf{w} - \mathbf{r}/\sigma v_1| > \mathrm{cst}$

The functions f_b, f_d, f_e, f_f, and f_h are bounded by functions of $\mathbf{w}$ which are integrable over $\mathbb{R}^2$. In the region $w > r/(2\sigma v_1)$, the integral of (f_a+f_c) over the angle of $\mathbf{v}_1$ is bounded by a function of $\mathbf{w}$ integrable over $\mathbb{R}_2$, while the corresponding angular integral of

$$\int_{\substack{w < r/(2\sigma v_1) \\ w, |\mathbf{w} - \hat{\mathbf{v}}_1| > \mathrm{cst}}} d\mathbf{w}\, (f_a + f_c)$$

is bounded. Finally, f_g can be rewritten as the sum of an integrable function of $\mathbf{w}$ plus the function

$$\Gamma^2[\mathbf{w} \cdot (\hat{\mathbf{v}}_1 - (\hat{\mathbf{v}}_1 - \mathbf{r}/\sigma v_1)/|\hat{\mathbf{v}}_1 - \mathbf{r}/\sigma v_1|^2)]^2/(2w^{\Gamma})$$

whose integral over $\mathbf{w}$ reduces to a bounded term plus

$$\frac{\pi\Gamma^2}{2(\Gamma-4)}\left[\hat{\mathbf{v}}_1 - \frac{\hat{\mathbf{v}}_1 - \mathbf{r}/\sigma v_1}{|\hat{\mathbf{v}}_1 - \mathbf{r}/\sigma v_1|^2}\right]^2$$

Consequently, the sole contribution to $C_\varepsilon^4(r)$ arises from the latter singular term and reads

$$e^2 \frac{z^4}{\sigma^4}\left(\frac{\sigma}{r}\right)^\Gamma \int_{1 < v_1 < r/(2\sigma)} d\mathbf{v}_1 \frac{\pi\Gamma^2}{2(\Gamma-4)} \frac{1}{v_1^{\Gamma-2}} \times\left[\hat{\mathbf{v}}_1 - \frac{\hat{\mathbf{v}}_1 - \mathbf{r}/\sigma v_1}{|\hat{\mathbf{v}}_1 - \mathbf{r}/\sigma v_1|^2}\right]^2 = 16\pi^2 e^2 \frac{z^4}{\sigma^4} \frac{1}{(\Gamma-4)^2}\left[\left(\frac{\sigma}{r}\right)^\Gamma - \left(\frac{\sigma}{r}\right)^{2\Gamma-4}\right] \tag{A.11}$$

In the last line of (A.11) we have omitted terms bounded by $\text{cst} \times z^4(\sigma/r)^\Gamma/(\Gamma-4)$, a function which also bounds all the other contributions to $C^{(4)}(r)$ of the present region.

(2) $1 < |\hat{\mathbf{v}}_1 - \mathbf{r}/\sigma| < r/(2\sigma)$

For obvious symmetry reasons, the contributions to $C_\varepsilon^4(r)$ and $C^{(4)}(r)$ are identical to those of the region (1).

(3) $v_1, |\mathbf{v}_1 - \mathbf{r}/\sigma| > r/(2\sigma)$

(3.i) $1/v_1 < w < \text{cst}$ or $1/v_1 < |\mathbf{w} - \mathbf{r}/\sigma v_1| < \text{cst}$

The functions f_d, f_e, and f_g are bounded, while f_f is bounded by a function which remains integrable at $\mathbf{w} = 0$ and $\mathbf{w} = \mathbf{r}/\sigma v_1$. On the other hand, f_a, f_b, and f_c are bounded outside the disks centered at $\mathbf{0}$ and $\mathbf{r}/\sigma v_1$ with radius $\mathbf{r}/(2\sigma v_1)$. In the disk $w < r/(2\sigma v_1)$ $[|\mathbf{w} - \mathbf{r}/\sigma v_1| < r/(2\sigma v_1)]$, f_c (f_b) remains bounded, while the angular integral of $(f_a + f_b)$ $[(f_a + f_c)]$ over the angle of $\mathbf{w}$ $(\mathbf{w} - \mathbf{r}/\sigma v_1)$ can be rewritten as a singular term bounded by $\text{cst}/w^{\Gamma-4}$ $[\text{cst}/|\mathbf{w} - \mathbf{r}/\sigma v_1|^{\Gamma-4}]$ plus a bounded function of w $(|\mathbf{w} - \mathbf{r}/\sigma v_1|)$. Therefore the integral of $(f_a + f_b + \cdots + f_g)$ over the considered region is bounded and does not contribute to $C_\varepsilon^4(r)$. The integral of f_h outside the disks $w < r/(2\sigma v_1)$ and $|\mathbf{w} - \mathbf{r}/\sigma v_1| < r/(2\sigma v_1)$ is bounded by a constant times

$$\int_{w, |\mathbf{w} - \mathbf{r}/\sigma v_1| > r/(2\sigma v_1)} d\mathbf{w} \left[\frac{\mathbf{w}}{w^2} - \frac{\mathbf{w} - \mathbf{r}/\sigma v_1}{|\mathbf{w} - \mathbf{r}/\sigma v_1|^2}\right]^2$$

which is a pure number, as shown by the transformation of variable $\mathbf{w} = \mathbf{u}r/\sigma v_1$. In the disk $w < r/(2\sigma v_1)$ $[|\mathbf{w} - \mathbf{r}/\sigma v_1| < r/(2\sigma v_1)]$, f_h is equal to the singular term

$$\tfrac{1}{2}\Gamma^2(\hat{\mathbf{v}}_1 \cdot \mathbf{w})^2/w^4$$

$$\left[\frac{1}{2}\Gamma^2 \frac{1}{|\hat{\mathbf{v}}_1 - \mathbf{r}/\sigma v_1|^\Gamma} [(\hat{\mathbf{v}}_1 - \mathbf{r}/\sigma v_1) \cdot (\mathbf{w} - \mathbf{r}/\sigma v_1)]^2/|\mathbf{w} - \mathbf{r}/\sigma v_1|^4\right]$$

plus a function integrable at $\mathbf{w}=0$ ($\mathbf{w}=\mathbf{r}/\sigma v_1$) and bounded by

$$[\text{cst}\times \sigma v_1/rw + \text{cst}\times(\sigma v_1/r)^2]$$
$$\{[\text{cst}\times \sigma v_1/(r\,|\mathbf{w}-\mathbf{r}/\sigma v_1|)+\text{cst}\times(\sigma v_1/r)^2]\}$$

Thus the integral of f_h over the above disks reduces to

$$\frac{\Gamma^2}{2}\left\{\int_{1/v_1<w<r/(2\sigma v_1)} d\mathbf{w}\,\frac{(\mathbf{w}\cdot\hat{\mathbf{v}}_1)^2}{w^4} + \frac{1}{|\hat{\mathbf{v}}_1-\mathbf{r}/\sigma v_1|^{\Gamma}}\int_{1/v_1<w'<r/(2\sigma v_1)} d\mathbf{w}'\,\frac{[\mathbf{w}'\cdot(\hat{\mathbf{v}}_1-\mathbf{r}/\sigma v_1)]^2}{w'^4}\right\}$$
$$=\frac{\pi\Gamma^2}{2}\left[1+\frac{1}{|\hat{\mathbf{v}}_1-r/\sigma v_1|^{\Gamma-2}}\right]\ln\frac{r}{2\sigma} \tag{A.12}$$

plus a bounded quantity. It then follows that the sole contribution to $C_\varepsilon^4(r)$ arises from the logarithmic term (A.12) and reads

$$e^2\frac{z^4}{\sigma^4}\left(\frac{\sigma}{r}\right)^{\Gamma}\int_{v_1,\,|\mathbf{v}_1-r/\sigma|>r/(2\sigma)} d\mathbf{v}_1\,\frac{\pi\Gamma^2\ln(r/\sigma)}{2v_1^{\Gamma-2}}\left[1+\frac{1}{|\hat{\mathbf{v}}_1-\mathbf{r}/\sigma v_1|^{\Gamma-2}}\right]$$
$$=16\pi e^2\frac{z^4}{\sigma^4}\left(\frac{\sigma}{r}\right)^{\Gamma}\ln\left(\frac{r}{\sigma}\right)\int_{v_1>r/\sigma} d\mathbf{v}_1\,\frac{1}{v_1^{\Gamma-2}}$$
$$=32\pi^2e^2\frac{z^4}{\sigma^4}\frac{1}{\Gamma-4}\frac{\ln(r/\sigma)}{r^{2\Gamma-4}} \tag{A.13}$$

In the second line of (A.13) the replacements of $|\hat{\mathbf{v}}_1-r/\sigma v_1|^{\Gamma-2}$ by 1 and of the condition v_1, $|\mathbf{v}_1-\mathbf{r}/\sigma|>r/\sigma$ by $v_1>r/\sigma$ are legitimate because the $1/(\Gamma-4)$ divergent term arises from the values of v_1 large compared to r/σ. The terms omitted in the second line of (A.13) are bounded by $\text{cst}\times z^4(\sigma/r)^{2\Gamma-4}\ln(r/\sigma)$, while the other contributions to $C^{(4)}(r)$ are bounded by $\text{cst}\times z^4\times(\sigma/r)^{2\Gamma-4}/(\Gamma-4)$.

(3.ii) $1/v_1<|\mathbf{w}-\hat{\mathbf{v}}_1|<\text{cst}$

As in the case $v_1<r/(2\sigma)$ and for the same reasons, this region does not contribute to $C_\varepsilon^4(r)$, and its contribution to $C^{(4)}(r)$ is bounded by $\text{cst}\times z^4\times(\sigma/r)^{2\Gamma-4}/(\Gamma-4)$.

(3.iii) w, $|\mathbf{w}-\mathbf{r}/\sigma v_1|$, $|\mathbf{w}-\hat{\mathbf{v}}_1|>\text{cst}$

The functions f_a, f_b, f_c, f_d, f_e, f_f, and f_h are bounded by integrable functions of $\mathbf{w}$, while f_g can be rewritten as a term bounded by an integrable function plus

$$\frac{\Gamma^2}{2w^\Gamma}\left[\mathbf{w}\cdot\left(\hat{\mathbf{v}}_1-\frac{\hat{\mathbf{v}}_1-\mathbf{r}/\sigma v_1}{(\hat{\mathbf{v}}_1-\mathbf{r}/\sigma v_1)^2}\right)\right]^2 \tag{A.14}$$

The integral of (A.14) over $\mathbf{w}$ is bounded by $\text{cst}\times(r/\sigma v_1)^2/(\Gamma-4)$. Thus there is no contribution of the considered region to $C_\varepsilon^4(r)$, and its contribution to $C^{(4)}(r)$ is bounded by $\text{cst}\times z^4(\sigma/r)^{2\Gamma-4}/(\Gamma-4)$.

The total contribution of the integral (A.3) to $C_\varepsilon^4(r)$ is equal to (A.13) plus twice the sum of (A.11) and (A.10). Adding the contribution to that (A.6) of the integral (A.2), we finally obtain the expression (4.13) of $C_\varepsilon^4(r)$. Taking into account the above uniform bounds for the various contributions to $C^{(4)}(r)$, we see that $C_\varepsilon^4(r)$ indeed is the limit form when $\Gamma\to 4^+$ of the first three terms in the large-distance expansion of $C^{(4)}(r)$ [note that the $\ln(r/\sigma)/r^{2\Gamma-4}$ terms arising from (A.9) and (A.12) exactly cancel out].

Finally, we derive a simple integral representation for $C_\varepsilon^4(r)$ which involves the coordinates $\mathbf{x}$ and $\mathbf{y}$ of the field pair $\mathscr{P}=[\oplus\ \mathbf{x}, \ominus\ \mathbf{y}]$. Since

$$\int_{|\mathbf{x}|,|\mathbf{x}-\mathbf{r}|>r/2}\frac{d\mathbf{x}}{\sigma^2}\int_{|\mathbf{x}-\mathbf{y}|>\sigma}\frac{d\mathbf{y}}{\sigma^2}\frac{\sigma^\Gamma}{|\mathbf{x}-\mathbf{y}|^\Gamma}\left[(\mathbf{x}-\mathbf{y})\cdot\left(\frac{\mathbf{x}}{|\mathbf{x}|^2}-\frac{\mathbf{x}-\mathbf{r}}{|\mathbf{x}-\mathbf{r}|^2}\right)\right]^2$$

is bounded by $\text{cst}/(\Gamma-4)$, the contribution (A.6) of the integral (A.2) to $C_\varepsilon^4(r)$ entirely arises from

$$-2e^2\frac{z^4}{\sigma^4}\left(\frac{\sigma}{r}\right)^\Gamma\int_{\substack{|\mathbf{x}|\text{ or }|\mathbf{x}-\mathbf{r}|<r/2\\|\mathbf{x}|,|\mathbf{x}-\mathbf{r}|>\sigma}}\frac{d\mathbf{x}}{\sigma^2}$$
$$\times\int_{|\mathbf{x}-\mathbf{y}|>\sigma}\frac{d\mathbf{y}}{\sigma^2}\frac{\Gamma^2}{2}\frac{\sigma^\Gamma}{|\mathbf{x}-\mathbf{y}|^\Gamma}\left[(\mathbf{x}-\mathbf{y})\cdot\left(\frac{\mathbf{x}}{|\mathbf{x}|^2}-\frac{\mathbf{x}-\mathbf{r}}{|\mathbf{x}-\mathbf{r}|^2}\right)\right]^2 \tag{A.15}$$

Furthermore, returning to the integral expressions over $\mathbf{v}_1$ and $\mathbf{w}$ in (A.10), (A.11), and (A.13) and introducing the variables $\mathbf{x}$ and $\mathbf{y}$, we find after simple manipulations that the contribution to $C_\varepsilon^4(r)$ of the integral (A.3) is identical to that of

$$2e^2\frac{z^4}{\sigma^4}\left(\frac{\sigma}{r}\right)^\Gamma\int_{\substack{|\mathbf{x}|\text{ or }|\mathbf{x}-\mathbf{r}|<r/2\\|\mathbf{x}|,|\mathbf{x}-\mathbf{r}|>\sigma}}\frac{d\mathbf{x}}{\sigma^2}$$
$$\times\int_{|\mathbf{x}-\mathbf{y}|>|\mathbf{x}|}\frac{d\mathbf{y}}{\sigma^2}\frac{\Gamma^2}{2}\left(\frac{\sigma}{|\mathbf{x}-\mathbf{y}|}\right)^\Gamma\left[(\mathbf{x}-\mathbf{y})\cdot\left(\frac{\mathbf{x}}{|\mathbf{x}|^2}-\frac{\mathbf{x}-\mathbf{r}}{|\mathbf{x}-\mathbf{r}|^2}\right)\right]^2 \tag{A.16}$$

Thus, the sum of (A.15) and (A.16) provides the integral representation (4.19) of $C_\varepsilon^4(r)$, which must be understood in the following mathematical sense: it reduces to $C_\varepsilon^4(r)$ plus a remainder which has the same uniform (with respect to r and Γ) upper bounds as the difference $[C^{(4)}(r) - C_\varepsilon^4(r)]$.

APPENDIX B

In this appendix, we show that the z^2 terms in the expansions of the expressions (5.28) and (2.8) for $1/\varepsilon$ do coincide. For this, we have to calculate ρ and $\rho_{-+}(|\mathbf{y}_1|)\,\mathbf{p}_{-+}(\mathbf{y}_1)$ at the order z^4, i.e.,

$$\begin{aligned}\rho = {} & \frac{z^2}{\sigma^4}\int d\mathbf{y}_1 \exp[\beta e^2 v(|\mathbf{y}_1|)] \\ & + \frac{z^4}{2\sigma^8}\int d\mathbf{y}_1\, d\mathbf{x}_2\, d\mathbf{y}_2\, \{\exp[-\beta V_4(\mathbf{0}, \mathbf{x}_2; \mathbf{y}_1, \mathbf{y}_2)] \\ & - \exp[\beta e^2 v(|\mathbf{y}_1|)]\exp[\beta e^2 v(|\mathbf{x}_2 - \mathbf{y}_2|)] \\ & - \exp[\beta e^2 v(|\mathbf{y}_2|)]\exp[\beta e^2 v(|\mathbf{x}_2 - \mathbf{y}_1|)]\} \\ & + O(z^6)\end{aligned} \tag{B.1}$$

and

$$\begin{aligned}\rho_{-+}(|\mathbf{y}_1|)\, p_{-+}(\mathbf{y}_1) = {} & -e\frac{z^2}{\sigma^4}\mathbf{y}_1 \exp[\beta e^2 v(|\mathbf{y}_1|)] \\ & - e\frac{z^4}{\sigma^8}\mathbf{y}_1 \int d\mathbf{x}_2\, d\mathbf{y}_2\, \{\exp[-\beta V_4(\mathbf{0}, \mathbf{x}_2; \mathbf{y}_1, \mathbf{y}_2)] \\ & - \exp[\beta e^2 v(|\mathbf{y}_1|)]\exp[\beta e^2 v(|\mathbf{x}_2 - \mathbf{y}_2|)]\} \\ & + e\frac{z^4}{\sigma^8}\int d\mathbf{x}_2\, \mathbf{x}_2 \int d\mathbf{y}_2\, \{\exp[-\beta V_4(\mathbf{0}, \mathbf{x}_2; \mathbf{y}_1, \mathbf{y}_2)] \\ & - \exp[-\beta V_4(\mathbf{0}, \mathbf{y}_2; \mathbf{y}_1, \mathbf{x}_2)]\} \\ & + O(z^6)\end{aligned} \tag{B.2}$$

The rhs of (5.28) then becomes, after using (5.29b),

$$\begin{aligned} 1 - {} & \frac{z^2}{\sigma^4 \int d\mathbf{y}_1 \exp[\beta e^2 v(|\mathbf{y}_1|)]} \\ & \times\left(\frac{1}{2}\int d\mathbf{y}_1\, d\mathbf{x}_2\, d\mathbf{y}_2\, \{\exp[-\beta V_4(\mathbf{0}, \mathbf{x}_2; \mathbf{y}_1, \mathbf{y}_2)]\right. \\ & - \exp[\beta e^2 v(|\mathbf{y}_1|)]\exp[\beta e^2 v(|\mathbf{x}_2 - \mathbf{y}_2|)] \\ & - \exp[\beta e^2 v(|\mathbf{y}_2|)]\exp[\beta e^2 v(|\mathbf{x}_2 - \mathbf{y}_1|)]\} \\ & \left. + \frac{\beta e^2}{2}\int d\mathbf{y}_1\, \nabla v(\mathbf{y}_1)\cdot \mathbf{P}(\mathbf{y}_1)\right) + O(z^4)\end{aligned} \tag{B.3}$$

with

$$\mathbf{P}(\mathbf{y}_1)=\mathbf{y}_1\int d\mathbf{x}_2\,d\mathbf{y}_2\,\{\exp-[\beta V_4(\mathbf{0},\mathbf{x}_2;\mathbf{y}_1,\mathbf{y}_2)] \\ -\exp[\beta e^2 v(|\mathbf{y}_1|)]\exp[\beta e^2 v(|\mathbf{x}_2-\mathbf{y}_2|)]\} \\ -\int d\mathbf{x}_2\mathbf{x}_2\int d\mathbf{y}_2\,\{\exp[-\beta V_4(\mathbf{0},\mathbf{x}_2;\mathbf{y}_1,\mathbf{y}_2)] \\ \times\exp[-\beta V_4(\mathbf{0},\mathbf{y}_2;\mathbf{y}_1,\mathbf{x}_2)]\} \tag{B.4}$$

In order to manipulate the conditionally convergent integrals which appear in (B.3) and (B.4), in particular to invert the successive integrals over $\mathbf{y}_1$, $\mathbf{x}_2$, and $\mathbf{y}_2$, it is convenient to introduce the function

$$G(\mathbf{0},\mathbf{x}_2;\mathbf{y}_1,\mathbf{y}_2)=\exp[-\beta V_4(\mathbf{0},\mathbf{x}_2;\mathbf{y}_1,\mathbf{y}_2)] \\ -\exp[\beta e^2 v(|\mathbf{y}_1|)]\exp[\beta e^2 v(|\mathbf{x}_2-\mathbf{y}_2|)] \\ \times[1+\beta e^2(\mathbf{y}_1\cdot\nabla)(\mathbf{y}_2-\mathbf{x}_2)\cdot\nabla v(\mathbf{x}_2)] \\ -\exp[\beta e^2 v(|\mathbf{y}_2|)]\exp[\beta e^2 v(|\mathbf{x}_2-\mathbf{y}_1|)] \\ \times[1+\beta e^2(\mathbf{y}_2\cdot\nabla)(\mathbf{y}_1-\mathbf{x}_2)\cdot\nabla v(\mathbf{x}_2)] \tag{B.5}$$

Taking into account the general prescription given in the main text for calculating such integrals, we then obtain

$$\int d\mathbf{y}_1\,d\mathbf{x}_2\,d\mathbf{y}_2\,\{\exp[-\beta V_4(\mathbf{0},\mathbf{x}_2;\mathbf{y}_1,\mathbf{y}_2)] \\ -\exp[\beta e^2 v(|\mathbf{y}_1|)]\exp[\beta e^2 v(|\mathbf{x}_2-\mathbf{y}_2|)] \\ -\exp[\beta e^2 v(|\mathbf{y}_2|)]\exp[\beta e^2 v(|\mathbf{x}_2-\mathbf{y}_1|)]\} \\ =\int d\mathbf{y}_1\,d\mathbf{x}_2\,d\mathbf{y}_2\,G(\mathbf{0},\mathbf{x}_2;\mathbf{y}_1,\mathbf{y}_2) \tag{B.6}$$

and

$$\beta e^2\int d\mathbf{y}_1\,\nabla v(\mathbf{y}_1)\cdot\mathbf{P}(\mathbf{y}_1) \\ =-\beta e^2\int d\mathbf{y}_1\,d\mathbf{x}_2\,d\mathbf{y}_2\,G(\mathbf{0},\mathbf{x}_2;\mathbf{y}_1,\mathbf{y}_2)(\mathbf{x}_2-\mathbf{y}_2-\mathbf{y}_1)\cdot\nabla v(\mathbf{y}_1) \\ +2\pi\beta e^2\int d\mathbf{y}_1\exp[\beta e^2 v(|\mathbf{y}_1|)]\int dt\,t^2\exp[\beta e^2 v(t)] \\ -\frac{\beta^2 e^4}{2}\int dt\,t^2\exp[\beta e^2 v(t)]\int d\mathbf{y}_1\,d\mathbf{t}'\exp[\beta e^2 v(t')] \\ \times[\nabla^2 v(\mathbf{y}_1)](\mathbf{t}'\cdot\nabla)\,v(\mathbf{y}_1-\mathbf{t}') \tag{B.7}$$

In (B.6) and (B.7), the integrals involving G are absolutely convergent because G has a sufficiently fast decay for any large separation of its arguments. The last two integrals in the rhs of (B.7) arise from the dipole–dipole interaction terms appearing in the definition (B.5) of G [these terms do not contribute to the rhs of (B.6) for obvious symmetry reasons]. On the other hand, an integration by parts gives

$$\beta e^2 \int d\mathbf{y}_1\, \nabla v(\mathbf{y}_1) \cdot \mathbf{P}(\mathbf{y}_1) = -\int d\mathbf{y}_1 \exp[\beta e^2 v(|\mathbf{y}_1|)]\, \nabla \cdot \{\mathbf{P}(\mathbf{y}_1) \exp[-\beta e^2 v(\mathbf{y}_1)]\} \quad \text{(B.8)}$$

If we replace $\mathbf{P}(\mathbf{y}_1)$ by (B.4), we can calculate $\nabla \cdot \{\mathbf{P}(\mathbf{y}_1) \exp[-\beta e^2 v(\mathbf{y}_1)]\}$ by inverting the differentiations with respect to $\mathbf{y}_1$ and the integrals over $\mathbf{x}_2$ and $\mathbf{y}_2$. Rewriting the corresponding result in terms of G, the identity (B.8) then becomes

$$\begin{aligned}
\beta e^2 \int d\mathbf{y}_1\, \nabla v(\mathbf{y}_1) \cdot \mathbf{P}(\mathbf{y}_1)
&= \beta e^2 \int d\mathbf{y}_1\, d\mathbf{x}_2\, d\mathbf{y}_2\, G(\mathbf{0}, \mathbf{x}_2; \mathbf{y}_1, \mathbf{y}_2)(\mathbf{x}_2 - \mathbf{y}_1 - \mathbf{y}_2) \cdot \nabla v(\mathbf{y}_1) \\
&\quad + 2\pi\beta e^2 \int d\mathbf{y}_1 \exp[\beta e^2 v(|\mathbf{y}_1|)] \int dt\, t^2 \exp[\beta e^2 v(t)] \\
&\quad - 2 \int d\mathbf{y}_1\, d\mathbf{x}_2\, d\mathbf{y}_2\, G(\mathbf{0}, \mathbf{x}_2; \mathbf{y}_1, \mathbf{y}_2) \\
&\quad + \frac{\beta^2 e^4}{2} \int d\mathbf{t}\, t^2 \exp[\beta e^2 v(t)] \\
&\quad \times \int d\mathbf{y}_1\, d\mathbf{t}' \exp[\beta e^2 v(t')]\, [\nabla^2 v(\mathbf{y}_1)](\mathbf{t}' \cdot \nabla)\, v(\mathbf{y}_1 - \mathbf{t}') \qquad \text{(B.9)}
\end{aligned}$$

where we have used the symmetry relations

$$G(\mathbf{0}, \mathbf{x}_2; \mathbf{y}_1, \mathbf{y}_2) = G(\mathbf{0}, \mathbf{x}_2; \mathbf{y}_2, \mathbf{y}_1) = G(\mathbf{0}, \mathbf{x}_2; \mathbf{x}_2 - \mathbf{y}_1, \mathbf{x}_2 - \mathbf{y}_2)$$

Adding (B.7) to (B.9), we find

$$\begin{aligned}
\beta e^2 \int d\mathbf{y}_1\, \nabla v(\mathbf{y}_1) \cdot \mathbf{P}(\mathbf{y}_1)
&= -\int d\mathbf{y}_1\, d\mathbf{x}_2\, d\mathbf{y}_2\, G(\mathbf{0}, \mathbf{x}_2; \mathbf{y}_1, \mathbf{y}_2) \\
&\quad + 2\pi\beta e^2 \int d\mathbf{y}_1 \exp[\beta e^2 v(|\mathbf{y}_1|)] \int d\mathbf{t}\, t^2 \exp[\beta e^2 v(t)] \qquad \text{(B.10)}
\end{aligned}$$

Replacing the integrals involved in (B.3) by their respective expressions (B.6) and (B.10), we finally get for the rhs of (5.28)

$$1 - \frac{\pi \beta e^2 z^2}{\sigma^4} \int dt\, t^2 \exp[\beta e^2 v(t)] + O(z^4) \qquad \text{(B.11)}$$

which indeed is identical to a truncated expansion at the order z^2 of the rhs of (2.8).

APPENDIX C

In this appendix, we calculate the function $D_{-+,-+}(\mathbf{t}', \mathbf{t}'', \hat{\mathbf{R}})$ at the order z^6 included and we show that it indeed satisfies the identity (5.41). The expression for $\rho^T_{-+-+}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4)$ at the order z^6 reads

$$
\begin{aligned}
&\rho^T_{-+-+}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4)\\
&\quad = \frac{z^4}{\sigma^8} \{\exp[-\beta V_4(\mathbf{r}_2, \mathbf{r}_4; \mathbf{r}_1, \mathbf{r}_3)]\\
&\qquad - \exp[\beta e^2 v(r_{12})] \exp[\beta e^2 v(r_{34})]\}\\
&\qquad + \frac{z^6}{\sigma^{12}} \int d\mathbf{x}\, d\mathbf{y}\, \{\exp[-\beta V_6(\mathbf{r}_2, \mathbf{r}_4, \mathbf{x}; \mathbf{r}_1, \mathbf{r}_3, \mathbf{y})]\\
&\qquad - \exp[-\beta V_4(\mathbf{r}_2, \mathbf{r}_4; \mathbf{r}_1, \mathbf{r}_3)] \exp[\beta e^2 v(|\mathbf{x}-\mathbf{y}|)]\\
&\qquad - \exp[-\beta V_4(\mathbf{r}_2, \mathbf{x}; \mathbf{r}_1, \mathbf{r}_3)] \exp[\beta e^2 v(|\mathbf{r}_4-\mathbf{y}|)]\\
&\qquad - \exp[-\beta V_4(\mathbf{r}_2, \mathbf{r}_4; \mathbf{r}_1, \mathbf{y})] \exp[\beta e^2 v(|\mathbf{x}-\mathbf{r}_3|)]\\
&\qquad - \exp[-\beta V_4(\mathbf{x}, \mathbf{r}_4; \mathbf{r}_1, \mathbf{r}_3)] \exp[\beta e^2 v(|\mathbf{r}_2-\mathbf{y}|)]\\
&\qquad - \exp[-\beta V_4(\mathbf{r}_2, \mathbf{r}_4; \mathbf{y}, \mathbf{r}_3)] \exp[\beta e^2 v(|\mathbf{x}-\mathbf{r}_1|)]\\
&\qquad - \exp[-\beta V_4(\mathbf{x}, \mathbf{r}_4; \mathbf{y}, \mathbf{r}_3)] \exp[\beta e^2 v(\mathbf{r}_{12})]\\
&\qquad - \exp[-\beta V_4(\mathbf{r}_2, \mathbf{x}; \mathbf{r}_1, \mathbf{y})] \exp[\beta e^2 v(\mathbf{r}_{34})]\\
&\qquad - \exp[-\beta V_4(\mathbf{x}, \mathbf{r}_4; \mathbf{r}_1, \mathbf{y})] \exp[\beta e^2 v(\mathbf{r}_{23})]\\
&\qquad - \exp[-\beta V_4(\mathbf{r}_2, \mathbf{x}; \mathbf{y}, \mathbf{r}_3)] \exp[\beta e^2 v(\mathbf{r}_{14})]\\
&\qquad + 2 \exp[\beta e^2 v(|\mathbf{x}-\mathbf{r}_1|)] \exp[\beta e^2 v(|\mathbf{r}_2-\mathbf{y}|)] \exp[\beta e^2 v(\mathbf{r}_{34})]\\
&\qquad + 2 \exp[\beta e^2 v(|\mathbf{x}-\mathbf{r}_3|)] \exp[\beta e^2 v(|\mathbf{r}_4-\mathbf{y}|)] \exp[\beta e^2 v(\mathbf{r}_{12})]\\
&\qquad + 2 \exp[\beta e^2 v(|\mathbf{x}-\mathbf{r}_3|)] \exp[\beta e^2 v(|\mathbf{r}_2-\mathbf{y}|)] \exp[\beta e^2 v(\mathbf{r}_{14})]\\
&\qquad + 2 \exp[\beta e^2 v(|\mathbf{x}-\mathbf{r}_1|)] \exp[\beta e^2 v(|\mathbf{r}_4-\mathbf{y}|)] \exp[\beta e^2 v(\mathbf{r}_{23})]\\
&\qquad + 2 \exp[\beta e^2 v(|\mathbf{x}-\mathbf{y}|)] \exp[\beta e^2 v(\mathbf{r}_{12})] \exp[\beta e^2 v(\mathbf{r}_{34})]\\
&\qquad + 2 \exp[\beta e^2 v(|\mathbf{x}-\mathbf{y}|)] \exp[\beta e^2 v(\mathbf{r}_{14})] \exp[\beta e^2 v(\mathbf{r}_{23})]\}\\
&\qquad + O(z^8)
\end{aligned}
\tag{C.1}
$$

We set $\mathbf{t}' = \mathbf{r}_2 - \mathbf{r}_1$, $\mathbf{t}'' = \mathbf{r}_4 - \mathbf{r}_3$, and $\mathbf{R} = (\mathbf{r}_3 + \mathbf{r}_4 - \mathbf{r}_1 - \mathbf{r}_2)/2$, and we study the limit of (C.1) when $R \to \infty$ with $\hat{\mathbf{R}}$, $\mathbf{t}'$, and $\mathbf{t}''$ being kept fixed. As expected, each term of order z^{2N} decays as $D^{(2N)}_{-+,-+}(\mathbf{t}', \mathbf{t}'', \hat{\mathbf{R}})/R^2$. Using a multipolar expansion of $V_4(\mathbf{r}_2, \mathbf{r}_4; \mathbf{r}_1, \mathbf{r}_3)$ with respect to $\mathbf{t}'$ and $\mathbf{t}''$, we easily obtain

$$D^{(4)}_{-+,-+}(\mathbf{t}', \mathbf{t}'', \hat{\mathbf{R}}) = -\beta e^2 \frac{z^4}{\sigma^8} \exp[\beta e^2 v(t')] \exp[\beta e^2 v(t'')] \times [(\mathbf{t}' \cdot \mathbf{t}'') - 2(\hat{\mathbf{R}} \cdot \mathbf{t}')(\hat{\mathbf{R}} \cdot \mathbf{t}'')] \tag{C.2}$$

The coefficient $D^{(6)}_{-+,-+}$ is entirely determined by the large-R behavior of the conditionally convergent integral

$$\begin{aligned} \int d\mathbf{x}\, d\mathbf{y}\, \{&\exp[-\beta V_6(\mathbf{r}_2, \mathbf{r}_4, \mathbf{x}; \mathbf{r}_1, \mathbf{r}_3, \mathbf{y})] \\ &- \exp[\beta e^2 v(|\mathbf{x}-\mathbf{y}|)] \exp[-\beta V_4(\mathbf{r}_2, \mathbf{r}_4; \mathbf{r}_1, \mathbf{r}_3)] \\ &- \exp[-\beta V_4(\mathbf{x}, \mathbf{r}_4; \mathbf{y}, \mathbf{r}_3)] \exp[\beta e^2 v(t')] \\ &- \exp[-\beta V_4(\mathbf{r}_2, \mathbf{x}; \mathbf{r}_1, \mathbf{y})] \exp[\beta e^2 v(t'')] \\ &+ 2 \exp[\beta e^2 v(|\mathbf{x}-\mathbf{y}|)] \exp[\beta e^2 v(t')] \exp[\beta e^2 v(t'')]\} \end{aligned} \tag{C.3}$$

[all the other terms in the integral coefficient of z^6 in (C.1) give contributions which decay faster than $1/R^2$]. It is convenient to rewrite (C.3) as the absolutely convergent integral $(\mathbf{t} = \mathbf{x} - \mathbf{y})$

$$\begin{aligned} \int d\mathbf{x}\, d\mathbf{y}\, (&\exp[-\beta V_6(\mathbf{r}_2, \mathbf{r}_4, \mathbf{x}; \mathbf{r}_1, \mathbf{r}_3, \mathbf{y})] \\ &- \exp[\beta e^2 v(t)] \exp[-\beta V_4(\mathbf{r}_2, \mathbf{r}_4; \mathbf{r}_1, \mathbf{r}_3)] \\ &- \exp[-\beta V_4(\mathbf{x}, \mathbf{r}_4; \mathbf{y}, \mathbf{r}_3)] \exp[\beta e^2 v(t')] \\ &- \exp[-\beta V_4(\mathbf{r}_2, \mathbf{x}; \mathbf{r}_1, \mathbf{y})] \exp[\beta e^2 v(t'')] \\ &+ \exp[\beta e^2 v(t)] \exp[\beta e^2 v(t')] \exp[\beta e^2 v(t'')] \\ &\times \{2 - \beta^2 e^4 [(\mathbf{t}' \cdot \nabla)(\mathbf{t}'' \cdot \nabla)\, v(\mathbf{r}_{24})][(\mathbf{t}' \cdot \nabla)(\mathbf{t} \cdot \nabla)\, v(\mathbf{x} - \mathbf{r}_2)] \\ &- \beta^2 e^4 [(\mathbf{t}' \cdot \nabla)(\mathbf{t}'' \cdot \nabla)\, v(\mathbf{r}_{24})][(\mathbf{t}'' \cdot \nabla)(\mathbf{t} \cdot \nabla)\, v(\mathbf{x} - \mathbf{r}_4)] \\ &- \beta^2 e^4 [(\mathbf{t}' \cdot \nabla)(\mathbf{t} \cdot \nabla)\, v(\mathbf{x} - \mathbf{r}_2)][(\mathbf{t}'' \cdot \nabla)(\mathbf{t} \cdot \nabla)\, v(\mathbf{x} - \mathbf{r}_4)]\}) \end{aligned} \tag{C.4}$$

plus

$$\beta^2 e^4 \exp[\beta e^2 v(t')] \exp[\beta e^2 v(t'')] \int d\mathbf{x}\, d\mathbf{y}\, \exp[\beta e^2 v(t)] \times [(\mathbf{t}' \cdot \nabla)(\mathbf{t} \cdot \nabla)\, v(\mathbf{x} - \mathbf{r}_2)][(\mathbf{t}'' \cdot \nabla)(\mathbf{t} \cdot \nabla)\, v(\mathbf{x} - \mathbf{r}_4)] \tag{C.5}$$

The large-R behavior of (C.5) is found to be

$$-\pi\beta^2 e^4 \exp[\beta e^2 v(t')] \exp[\beta e^2 v(t'')] \int d\mathbf{t}\, t^2 \exp[\beta e^2 v(t)]$$
$$\times (\mathbf{t}' \cdot \nabla)(\mathbf{t}'' \cdot \nabla)\, v(\mathbf{R}) \qquad \text{(C.6)}$$

after an integration by parts and use of the identity $\int d\mathbf{x}\, \nabla^2 v(x) = -2\pi$. The leading contributions to the large-R behavior of (C.4) arise from configurations where the field pair $\mathscr{P} = \{\oplus\, \mathbf{x}, \ominus\, \mathbf{y}\}$ is close either to the pairs $\mathscr{P}'$ or $\mathscr{P}''$. For $\mathscr{P}$ close to $\mathscr{P}''$ we get the $1/R^2$ contribution

$$\beta e^2 \exp[\beta e^2 v(t')] \int d\mathbf{x}\, d\mathbf{y}\, \{\exp[-\beta V_4(\mathbf{x}, \mathbf{r}_4; \mathbf{y}, \mathbf{r}_3)]$$
$$- \exp[\beta e^2 v(t)] \exp[\beta e^2 v(t'')][1 + \beta e^2 (\mathbf{t}'' \cdot \nabla)(\mathbf{t} \cdot \nabla)\, v(\mathbf{x} - \mathbf{r}_4)\}$$
$$\times ((\mathbf{t} + \mathbf{t}'') \cdot \nabla)(\mathbf{t}' \cdot \nabla)\, v(\mathbf{R})$$
$$= \beta e^2 \exp[\beta e^2 v(t')][\mathbf{P}(\mathbf{t}'') \cdot \nabla](\mathbf{t}' \cdot \nabla)\, v(\mathbf{R})$$
$$+ \pi\beta^2 e^4 \exp[\beta e^2 v(t')] \exp[\beta e^2 v(t'')] \int d\mathbf{t}\, t^2 \exp[\beta e^2 v(t)]$$
$$\times (\mathbf{t}' \cdot \nabla)(\mathbf{t}'' \cdot \nabla)\, v(\mathbf{R}) \qquad \text{(C.7)}$$

where $\mathbf{P}(\mathbf{t}'')$ is defined by (B.4) with $\mathbf{y}_1 = -\mathbf{t}''$. The contribution of the region $\mathscr{P}$ close to $\mathscr{P}'$ reduces to (C.7) with $(\mathbf{t}'', \mathbf{t}')$ in place of $(\mathbf{t}', \mathbf{t}'')$ for obvious symmetry reasons. Thus, the large-R behavior of (C.3) is equal to (C.6) plus twice the symmetrized (with respect to $\mathbf{t}'$ and $\mathbf{t}''$) form of (C.7). This leads to

$$D^{(6)}_{-+,-+}(\mathbf{t}', \mathbf{t}''; \hat{\mathbf{R}})$$
$$= -\pi\beta^2 e^4 \frac{z^6}{\sigma^{12}} \exp[\beta e^2 v(t')] \exp[\beta e^2 v(t'')] \int d\mathbf{t}\, t^2 \exp[\beta e^2 v(t)]$$
$$\times [(\mathbf{t}' \cdot \mathbf{t}'') - 2(\hat{\mathbf{R}} \cdot \mathbf{t}')(\hat{\mathbf{R}} \cdot \mathbf{t}'')]$$
$$- \beta e^2 \frac{z^6}{\sigma_{12}} (\exp[\beta e^2 v(t')]\, \{[\mathbf{P}(\mathbf{t}'') \cdot \mathbf{t}'] - 2[\hat{\mathbf{R}} \cdot \mathbf{P}(\mathbf{t}'')](\hat{\mathbf{R}} \cdot \mathbf{t}')\}$$
$$+ \exp[\beta e^2 v(t'')]\{[\mathbf{P}(\mathbf{t}') \cdot \mathbf{t}''] - 2(\hat{\mathbf{R}} \cdot \mathbf{t}'')[\hat{\mathbf{R}} \cdot \mathbf{P}(\mathbf{t}')]\}) \qquad \text{(C.8)}$$

At the order z^6 included the rhs of the identity (5.41) reads

$$-\frac{ez^4}{\sigma^8}\exp[\beta e^2 v(t')]\int dt\,\exp[\beta e^2 v(t)]\,[\mathbf{t}'-2(\hat{\mathbf{R}}\cdot\mathbf{t}')\,\hat{\mathbf{R}}]$$

$$-\frac{ez^6}{\sigma^{12}}\exp[\beta e^2 v(t')]\int d\mathbf{y}_1\,d\mathbf{x}_2\,d\mathbf{y}_2\,G(\mathbf{0},\mathbf{x}_2;\mathbf{y}_1,\mathbf{y}_2)[\mathbf{t}'-2(\hat{\mathbf{R}}\cdot\mathbf{t}')\,\hat{\mathbf{R}}]$$

$$-\frac{ez^6}{\sigma^{12}}\int dt\,\exp[\beta e^2 v(t)][\mathbf{P}(\mathbf{t}')-2(\hat{\mathbf{R}}\cdot\mathbf{t}')\cdot\mathbf{P}(\mathbf{t}')]$$

$$+O(z^8) \qquad \text{(C.9)}$$

At the same order, the lhs of (5.41) is determined from the expressions (C.2), (C.8) of $D^{(4,6)}_{-+,-+}$. Using the colinearity of the vectors $\mathbf{t}''$, $\mathbf{P}(\mathbf{t}'')$, and $\mathbf{F}(\mathbf{t}'')=-\nabla v(\mathbf{t}'')$, as well as the identity (B.10) with $\mathbf{y}_1=-\mathbf{t}''$, we find that this lhs indeed reduces to (C.9).

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