Absence of exponential clustering in quantum Coulomb fluids

A. Alastuey
Laboratoire de Physique Théorique et Hautes Energies, Université de Paris-Sud, Bâtiment 211, F-91405 Orsay, France

Ph. A. Martin
Institut de Physique Théorique, Ecole Polytechnique Fédérale de Lausanne, PHB-Ecublens-CH-1015 Lausanne, Switzerland

(Received 10 April 1989)

We show that the quantum corrections to the classical correlations of a Coulomb fluid do not decay exponentially fast for all values of the thermodynamical parameters. Specifically, the $\hbar^4$ term in the Wigner-Kirkwood expansion of the equilibrium charge-charge correlations of the quantum one-component plasma is found to decay like $|\mathbf{r}|^{-10}$. More generally, using functional integration, we present a diagrammatic representation of the $\hbar$ expansion of the correlations in a multicomponent fluid with a locally regularized Coulomb potential and Maxwell-Boltzmann statistics. The $\hbar^{2n}$ terms are found to decay algebraically for all $n \geq 2$. Furthermore, an analysis of the hierarchy equations for the correlations provides upper bounds that are compatible with the findings of the perturbative expansion. Except for the monopole, all higher-order multipole sum rules do not hold, in general, in the quantum system. This violation of the multipole sum rules as well as the related algebraic tails are due to the intrinsic quantum fluctuations that prevent a perfect organization of the screening clouds. This phenomenon is illustrated in a simpler model where the large-distance correlations between two quantum particles embedded in a classical plasma can be exactly computed.

I. INTRODUCTION

One of the most fundamental properties of a system of charged particles in thermal equilibrium is the screening of the Coulomb potential $\phi(\mathbf{r})=|\mathbf{r}|^{-1}$. The following familiar and widely accepted picture is part of the standard background knowledge. A particle is surrounded by a screening cloud of opposite charge having an extension λ, the screening length. The charge distribution of the particle together with its cloud produces, in the medium, an effective potential

$$\phi_{\text{eff}}(\mathbf{r})\sim\frac{1}{|\mathbf{r}|}\exp(-|\mathbf{r}|/\lambda)\ ,$$

which becomes negligible at distances $|\mathbf{r}|>\lambda$. Then, for practical purposes, one may take the screening effects into account by replacing the Coulomb potential $\phi(\mathbf{r})$ by the exponential potential $\phi_{\text{eff}}(\mathbf{r})$. This picture is supported by all the mean-field treatments of the collective behavior of charges, starting with the Debye-Hückel theory of classical electrolytes.[1] Soon after the emergence of quantum mechanics, the Thomas-Fermi theory of the electron fluid has led to the same form of the effective potential.[2] An exponential effective potential is also obtained in the random-phase approximation (RPA) extensively developed in the years 1950–1960.[3,4] These assertions are strictly valid at nonzero temperatures. In the ground state, one finds an additional oscillatory algebraic term, the Friedel oscillations, due to the sharpness of the Fermi distribution. The application of mean-field theories to the calculation of the correlations of the fluid itself (see, for instance, Appendix H) also predicts an exponential clustering.

But what is rigorously known on this question? It has been firmly established in recent years that the Debye-Hückel picture is indeed correct in a plasma phase of classical charges. At sufficiently high temperature and low densities, Brydges and Federbush,[5] Imbrie,[6] and Yang[7] rigorously show that the particle correlations have a decay which is bounded by an exponential. Also all the studies of the solvable two-dimensional classical Coulomb models at a special value of the temperature exhibit a fast decay of the correlations.[8,9] However, when quantum mechanics is taken into account, the results are still scarce. The existence of the thermodynamic limit for the pressure[10] and the correlations[11] has been proven in a number of cases, but essentially nothing is known exactly on the asymptotic behavior of the particle distributions. Even doubt has been raised on the possible exponential falloff of the quantum-mechanical charge-charge correlations.[12,13] In Ref. 13 the authors rigorously show that certain imaginary-time Green's functions have an algebraic decay and they conjecture that the same should be true for the charge-charge correlation function. The point of this paper is to present strong evidences that the equilibrium correlations of quantum-mechanical charges do not cluster exponentially fast, irrespective of the value of the density and of the temperature T.

Let us give a first qualitative understanding for this lack of exponential clustering. It is known both classically[14] and quantum mechanically[15] that a falloff of the correlations faster than any inverse power is equivalent to strong screening properties: the screening clouds are so perfectly organized at equilibrium as to shield not only the total charge but all multipoles of any given particle configuration. The central observation is that this perfect organization does occur in a classical plasma phase, but is always destroyed by the quantum fluctuations. In the quantum system the monopole still vanishes (there is no bare Coulomb potential seen in equilibrium matter); however, the higher-order multipoles do not, in general, vanish. The latter generate then multipole forces which in turn induce algebraic tails in the correlations. We immediately emphasize that these multipole forces are different from the van der Waals forces due to the occurrence of complex polarizable entities, atoms or molecules, that can now be formed by the quantum binding process. The phenomenon is really due to the intrinsic quantum nature of the particles, as it is exemplified in the one-component plasma (OCP) where only one species of structureless charges with the same sign is present and no binding occurs (Sec. IV). It is clear that the mean-field theories (Thomas-Fermi, RPA) do not account for these fine quantum effects, and in this respect, do not reproduce even qualitatively the true behavior of the system.

We develop our arguments along three lines. First, in Secs. II and III, we examine the general constraints that are imposed on the correlations by the structure of the equilibrium equations. More precisely, in Sec. II, we perform an asymptotic analysis of the "evolution equations" for the imaginary-time Green functions of the OCP as a charge is sent to infinity. Under reasonable assumptions (existence of integer inverse power-law expansions starting with a $|\mathbf{r}|^{-3}$ term), we show that the Kubo-Martin-Schwinger (KMS) equilibrium condition and the locality imply upper bounds on the decay of the correlations. For instance, the charge-charge correlations decay faster than $|\mathbf{r}|^{-6}$. These considerations are nonperturbative and valid for Fermi statistics, but rely on a number of *a priori* assumptions (existence of the thermodynamic limit, monotonous decay). In Sec. III we derive various sum rules for the quantum OCP. Most of them have been obtained earlier by the linear-response theory. Here they appear as consistency relations imposed by the long range of the Coulomb force in the equilibrium equations. It is, however, not possible to pursue the analysis as in the high-temperature classical phase[16] to exclude any monotonous inverse power-law decay and establish the validity of all multipole sum rules.

This leads us to the second aspect of our work, an investigation of the quantum corrections to the classical correlation functions using the Wigner-Kirkwood expansion formalism in powers of the Planck constant $\hbar$. From the fourth order on, we find that these corrections have algebraic tails which can reasonably be considered as lower bounds to the decay in a semiclassical regime (but we do not control the possible convergence of the series). In Sec. IV we present a detailed calculation of the $\hbar^4$ term of the charge-charge correlation function of the OCP with Maxwell-Boltzmann statistics and obtain that this term behaves as $|\mathbf{r}|^{-10}$ as $|\mathbf{r}|\to\infty$. In Sec. V we briefly indicate how the results of the preceding sections for the OCP generalize to multicomponent systems (with a Coulomb potential regularized at short distances). Section VI is devoted to a more thorough study of the $\hbar$ expansion for multicomponent systems from the viewpoint of functional integration. Here we generate the terms more systematically by diagrammatic rules and give prescriptions to single out algebraically decaying diagrams. A qualitative analysis of these diagrams reveals that there is a slow decay at all order $\hbar^{2n}$, $n\geq 2$. In particular, this analysis indicates that the particle-particle correlations should have a large-distance behavior as $|\mathbf{r}|^{-6}$ while the several point correlations should have even the slower decay $|\mathbf{r}|^{-3}$ as groups of particles are separated.

Finally, in Sec. VII, we study a simplified model where only two quantum charges are immersed in a classical plasma. It is then possible to determine the exact asymptotic behavior of the correlations of the two charges, which is found as $|\mathbf{r}|^{-6}$, $|\mathbf{r}|\to\infty$. The model also enables us to illustrate explicitly the role played by the quantum fluctuations in the occurrence of the algebraic tails. Discussions and conclusions are presented in Sec. VIII. A preliminary account of this work is published in Ref. 17, and part of it is also reviewed in Ref. 18.

II. EQUILIBRIUM EQUATIONS AND CLUSTER PROPERTIES

A. General setting

In this section we investigate what kind of cluster properties are compatible with the equilibrium equations. For simplicity we consider the quantum-mechanical OCP and give the modifications for the multicomponent systems in Sec. V. The OCP consists of quantum particles of charge e and mass m in a classical background with charge density $-e\rho$. Since the spin plays no role in the sequel, it will not be taken into account.

Relevant quantities belonging to a single particle are the number density

$$N(\mathbf{r})=\delta(\mathbf{r}-\mathbf{q}) \tag{2.1}$$

and the current-density

$$\mathbf{J}(\mathbf{r})=\frac{e}{2m}[\mathbf{p}\delta(\mathbf{r}-\mathbf{q})+\delta(\mathbf{r}-\mathbf{q})\mathbf{p}] , \tag{2.2}$$

where $\mathbf{p}$ and $\mathbf{q}$ are the momentum and position operators of the particle. We keep the same notation $N(\mathbf{r})$ and $\mathbf{J}(\mathbf{r})$ for the second quantized densities in the many-particle system. In particular, the particle and charge density are

$$N(\mathbf{r})=a^*(\mathbf{r})a(\mathbf{r}) , \tag{2.3}$$

$$Q(\mathbf{r})=e[N(\mathbf{r})-\rho] , \tag{2.4}$$

where $a^*(\mathbf{r})$ and $a(\mathbf{r})$ are the creation and annihilation of a particle with Fermi statistics, $a^*(\mathbf{r})a(\mathbf{r}')+a(\mathbf{r}')a^*(\mathbf{r})=\delta(\mathbf{r}-\mathbf{r}')$.

The total energy is $H=K+U$ with K and U the kinetic and potential energy formally given by

$$K=\int d\mathbf{p}\frac{|\mathbf{p}|^2}{2m}\tilde{a}^*(\mathbf{p})\tilde{a}(\mathbf{p}) , \tag{2.5}$$

$$U=\tfrac{1}{2}\int d\mathbf{r}\int d\mathbf{r}'\phi(\mathbf{r}-\mathbf{r}'):Q(\mathbf{r})Q(\mathbf{r}'): , \tag{2.6}$$

where $\phi(\mathbf{r})=1/|\mathbf{r}|$ is the Coulomb potential, $\tilde{a}(\mathbf{p})$ the Fourier transform of $a(\mathbf{r})$, and : : means Wick ordering.

The imaginary-time correlation $\langle A_\tau B\rangle$ of two local observables A and B is defined in the thermodynamic limit by

$$\langle B_\tau A \rangle = \lim_{\Lambda\to\mathbb{R}^3} \frac{1}{\Xi_\Lambda} \mathrm{Tr}_\Lambda (e^{\beta\mu N} e^{-(\beta-\tau)H} B e^{-\tau H} A) , \tag{2.7}$$

where $\Xi_\Lambda = \mathrm{Tr}_\Lambda e^{-\beta(H-\mu N)}$ is the grand partition function, μ is the chemical potential, and N the total number of particles. In the right-hand side (rhs) of (2.7), the finite volume Hamiltonian has to be defined with appropriate conditions at the boundaries of Λ. The existence of the thermodynamic for a system of electrons and nuclei is established in Ref. 10. The infinite volume limit of the correlations (2.7) has been shown to exist for a charge symmetric Coulomb gas without statistics and for Bose statistics at sufficiently low activity.[11] We assume here that the limit (2.7) exists for the OCP with Fermi statistics and still obeys the same relations that hold at finite volume. One has, in particular, the Kubo-Martin-Schwinger condition

$$\langle B_\tau A \rangle = \langle A_{\beta-\tau} B \rangle, \quad 0 \le \tau \le \beta \tag{2.8}$$

and the imaginary-time equations of motion

$$\frac{d}{d\tau} \langle B_\tau A \rangle = \langle [H,B]_\tau A \rangle = \langle A_{\beta-\tau} [H,B] \rangle . \tag{2.9}$$

In particular, one has

$$\langle B_\tau \rangle = \langle B \rangle, \quad 0 \le \tau \le \beta . \tag{2.10}$$

Specifying $B = Q(\mathbf{r})$ in (2.9) and working out the commutator $[H,Q(\mathbf{r})] = [K,Q(\mathbf{r})]$ gives the "continuity equation"

$$\frac{d}{d\tau} \langle Q_\tau(\mathbf{r}) A \rangle_T - i\hbar \nabla \cdot \langle \mathbf{J}_\tau(\mathbf{r}) A \rangle_T = 0 , \tag{2.11}$$

where the truncated expectation is defined by

$$\langle BA \rangle_T = \langle BA \rangle - \langle B \rangle \langle A \rangle . \tag{2.12}$$

Notice that by neutrality and translation invariance

$$\begin{aligned} &\langle Q_\tau(\mathbf{r}) \rangle = \langle Q(\mathbf{r}) \rangle = e(\langle N(\mathbf{r}) \rangle - \rho) = 0 , \\ &\langle J^\mu_\tau(\mathbf{r}) \rangle = \langle J^\mu(\mathbf{r}) \rangle = 0, \quad \mu = 1,2,3 . \end{aligned} \tag{2.13}$$

Choosing now $B = \mathbf{J}(\mathbf{r})$ in (2.9) gives the "law of force"

$$\frac{d}{d\tau} \langle J^\mu_\tau(\mathbf{r}) A \rangle = i\hbar \sum_{\nu=1}^{3} \nabla^\nu \langle K^{\mu\nu}_\tau(\mathbf{r}) A \rangle - i\hbar \frac{e^2}{m} \int d\mathbf{r}' F^\mu(\mathbf{r}-\mathbf{r}') \langle :N_\tau(\mathbf{r}) Q_\tau(\mathbf{r}'): A \rangle . \tag{2.14}$$

In Eq. (2.14) $\mathbf{F}(\mathbf{r}) = -\nabla\phi(\mathbf{r})$ is the Coulomb force and $K^{\mu\nu}(\mathbf{r})$ is a "kinetic energy density tensor"

$$K^{\mu\nu}(\mathbf{r}) = \frac{1}{2m} [p^\mu J^\nu(\mathbf{r}) + J^\nu(\mathbf{r}) p^\mu] . \tag{2.15}$$

Introducing the fully truncated expectation

$$\langle CBA \rangle_T = \langle (C - \langle C \rangle)(B - \langle B \rangle)(A - \langle A \rangle) \rangle \tag{2.16}$$

and using the neutrality (2.13) again, one finds

$$\langle :N_\tau(\mathbf{r}) Q_\tau(\mathbf{r}'): A \rangle = \langle :N_\tau(\mathbf{r}) Q_\tau(\mathbf{r}'): A \rangle_T + \rho \langle Q_\tau(\mathbf{r}') A \rangle_T + \langle A \rangle \langle :N(\mathbf{r}) Q(\mathbf{r}'): \rangle_T . \tag{2.17}$$

With the help of (2.13) and (2.17), Eq. (2.14) can be written in terms of truncated expectations only,

$$\frac{d}{d\tau} \langle J^\mu_\tau(\mathbf{r}) A \rangle_T = i\hbar \sum_{\nu=1}^{3} \nabla^\nu \langle K^{\mu\nu}_\tau(\mathbf{r}) A \rangle_T \tag{2.18a}$$

$$- i\hbar \frac{\rho e^2}{m} \int d\mathbf{r}' F^\mu(\mathbf{r}-\mathbf{r}') \langle Q_\tau(\mathbf{r}') A \rangle_T \tag{2.18b}$$

$$- i\hbar \frac{e^2}{m} \int d\mathbf{r}' F^\mu(\mathbf{r}-\mathbf{r}') \langle :N_\tau(\mathbf{r}) Q_\tau(\mathbf{r}'): A \rangle_T, \quad \mu = 1,2,3 . \tag{2.18c}$$

Because of translation and rotation invariance, the correlation $\langle :N(\mathbf{r}) Q(\mathbf{r}'): \rangle$ depends only on $|\mathbf{r}-\mathbf{r}'|$ and it does not contribute to the integral with the force in (2.14). Moreover, $\nabla^\nu \langle K^{\mu\nu}(\mathbf{r}) \rangle = 0$ since $\langle K^{\mu\nu}(\mathbf{r}) \rangle$ is constant with respect to $\mathbf{r}$.

The correlation of the charge $\langle Q_\tau(\mathbf{r}) A \rangle_T = \langle A_{\beta-\tau} Q(\mathbf{r}) \rangle_T$ with an observable A can be interpreted as an excess charge density when the observable A has been specified. It will also be called the charge cloud attached to A. The term (2.18b) is proportional to the electric field

$$\mathbf{E}(\mathbf{r}|A) = \int d\mathbf{r}' \mathbf{F}(\mathbf{r}-\mathbf{r}') \langle Q_\tau(\mathbf{r}') A \rangle_T \tag{2.19}$$

at the point $\mathbf{r}$ due to this charge cloud and it satisfies the Poisson equation

$$\nabla \cdot \mathbf{E}(\mathbf{r}|A) = 4\pi \langle Q_\tau(\mathbf{r}) A \rangle_T . \tag{2.20}$$

Finally, the combination of (2.11), (2.18), and (2.20) leads to the second-order differential equation

$$\frac{d^2}{d\tau^2} \langle Q_\tau(\mathbf{r}) A \rangle_T - \hbar^2 \omega_p^2 \langle Q_\tau(\mathbf{r}) A \rangle_T \tag{2.21a}$$

$$= -\hbar^2 \sum_{\mu,\nu}^{3} \nabla^\mu \nabla^\nu \langle K^{\mu\nu}_\tau(\mathbf{r}) A \rangle_T \tag{2.21b}$$

$$+ \frac{\hbar^2 e^2}{m} \nabla \cdot \int d\mathbf{r}' \mathbf{F}(\mathbf{r}-\mathbf{r}') \langle :N_\tau(\mathbf{r}) Q_\tau(\mathbf{r}'): A \rangle_T , \tag{2.21c}$$

where $\omega_p = (4\pi e^2 \rho / m)^{1/2}$ is the plasma frequency.

The integrals in (2.18), (2.19), and (2.21) are absolutely convergent if the correlations decay at least as

$$|\langle Q_\tau(\mathbf{r}')A\rangle_T| \le \frac{M_1}{|\mathbf{r}'|^\delta} , \quad |\langle :N_\tau(\mathbf{r})Q_\tau(\mathbf{r}'):A\rangle_T| \le \frac{M_2}{|\mathbf{r}'|^\delta} , \tag{2.22}$$

with $\delta > 1$ and fixed τ, A, and $\mathbf{r}$. We shall assume this minimal cluster property holds throughout this section and derive some exact consequences of the equilibrium equations.

B. Cluster properties

We investigate the constraints which are imposed by Eqs. (2.11), (2.18), and (2.21) on the decay of the correlations. These equations can obviously not be solved explicitly and our analysis relies on the *a priori* assumption that this decay is like an integer inverse power law without oscillations at infinity. We remark that this assumption is plausible physically in so far as nonzero multipole moments of the charge clouds will induce long-range multipole forces with algebraic tails.

The monopole of the charge cloud (the total charge) is expected to vanish,

$$\int d\mathbf{r}\langle Q_\tau(\mathbf{r})A\rangle_T = \int d\mathbf{r}\langle A_{\beta-\tau}Q(\mathbf{r})\rangle_T = 0 , \tag{2.23}$$

otherwise localized charges in matter would produce a bare Coulomb potential at large distances. But the dipole $\int d\mathbf{r}\,\mathbf{r}\langle Q_\tau(\mathbf{r})A\rangle_T$ has no fundamental reasons to vanish for a general A and in fact it does not [see Eqs. (3.12) and (3.15) of Sec. III]. As a consequence, the electric field (2.19) will not decay faster than $|\mathbf{r}|^{-3}$,

$$\mathbf{E}(\mathbf{r}|A) \sim \frac{1}{|\mathbf{r}|^3}\int d\mathbf{r}'[3\hat{\mathbf{r}}(\hat{\mathbf{r}}\cdot\mathbf{r}') - \mathbf{r}']\langle Q_\tau(\mathbf{r}')A\rangle_T , \quad |\mathbf{r}| \to \infty, \quad \hat{\mathbf{r}} = \frac{\mathbf{r}}{|\mathbf{r}|} . \tag{2.24}$$

For the consistency of Eq. (2.18), it is necessary that some other terms of this equation behave also as $|\mathbf{r}|^{-3}$ for large $|\mathbf{r}|$. It is therefore natural to assume that all terms and all correlations occurring in Eq. (2.18) have an asymptotic development starting with a $|\mathbf{r}|^{-3}$ contribution. Then the structure of Eqs. (2.18) and (2.21) imposes several constraints by equating the coefficients of these developments at a given order. We do not claim that other possible behaviors excluded by this scheme cannot occur (e.g., oscillations, fractional inverse power law, decay even slower than $|\mathbf{r}|^{-3}$). We expect, however, that at sufficiently high temperature and low density at least, the decay will be monotonous and the description given below is meaningful.

We now formalize our assumption in more precise terms.

(i) Let

$$A = A^1_{\tau_1} A^2_{\tau_2} \cdots A^k_{\tau_k}, \quad 0 \le \tau_j \le \beta, \quad j = i, \ldots, k \tag{2.25}$$

be a product of imaginary time evolved local observables A^j (like particle and current densities). Then for any fixed such A, $\langle Q_\tau(\mathbf{r})A\rangle_T$ has an asymptotic expansion

$$\langle Q_\tau(\mathbf{r})A\rangle_T = \frac{w_3(\tau,A)}{|\mathbf{r}|^3} + \frac{w_4(\tau,A)}{|\mathbf{r}|^4} + \frac{w_5(\tau,A)}{|\mathbf{r}|^5} + \cdots . \tag{2.26}$$

We assume similarly that all the truncated correlations functions involved in Eqs. (2.11), (2.18), and (2.21) have inverse power-law asymptotic expansions as a point $\mathbf{r}$ is sent to infinity.

(ii) For fixed τ and A, we can find $n_1 \ge 3$, $n_2 \ge 3$ and ϵ, $0 \le \epsilon < 2$, such that the fully truncated function $g(\mathbf{r},\mathbf{r}') = \langle :N_\tau(\mathbf{r})Q_\tau(\mathbf{r}'):A\rangle_T$ satisfies bounds of the form

$$\begin{aligned} ||\mathbf{r}|^{n_1} g(\mathbf{r},\mathbf{r}')| &\le M(t), \quad |\mathbf{r}-\mathbf{r}'| \le \frac{|\mathbf{r}|}{2} \\ ||\mathbf{r}|^{n_2} g(\mathbf{r},\mathbf{r}')| &\le M(t), \quad |\mathbf{r}-\mathbf{r}'| \ge \frac{|\mathbf{r}|}{2} \end{aligned} \tag{2.27}$$

with $t = \min(|\mathbf{r}'|, |\mathbf{r}-\mathbf{r}'|)$ and

$$\int d\mathbf{r}\frac{M(|\mathbf{r}|)}{|\mathbf{r}|^\epsilon} < \infty . \tag{2.28}$$

The distinction between the two cases (2.27) allows for the possibility of different cluster properties depending on whether the distance $|\mathbf{r}-\mathbf{r}'|$ remains finite or not. The condition (2.28) on $M(t)$ expresses the usual property of the fully truncated function, that is, has some joint decay as two of the distances $|\mathbf{r}|$, $|\mathbf{r}'|$, and $|\mathbf{r}-\mathbf{r}'|$ grow to infinity. According to the general scheme, $M(|\mathbf{x}|)$ decays not slower than $1/|\mathbf{x}|^3$ and (2.28) is fulfilled for any ϵ strictly positive.

The next proposition shows that the correlation of the charge always has a faster decay than the threshold behavior $|\mathbf{r}|^{-3}$.

Proposition 1. Assume that properties (i) and (ii) hold with $n_1 = n_2 = 3$; then the correlation of the charge with a general observable A of the form (2.25) satisfies

$$|\langle Q_\tau(\mathbf{r})A\rangle_T| \le \frac{C_1(\tau,A)}{|\mathbf{r}|^4} . \tag{2.29}$$

Moreover, if A is strictly local one has

$$|\langle Q_\tau(\mathbf{r})A\rangle_T| \le \frac{C_2(\tau,A)}{|\mathbf{r}|^5} . \tag{2.30}$$

We first state the following lemma which is used to control the behavior for large $\mathbf{r}$ of the integral occurring in the term (2.21c) (proof in Appendix A).

Lemma 1. Set $h(\mathbf{r}') = \lim_{|\mathbf{r}|\to\infty} |\mathbf{r}|^{n_1} g(\mathbf{r},\mathbf{r}+\mathbf{r}')$ for fixed $\mathbf{r}'$ and suppose that (2.27) and (2.28) hold; then

$$\int d\mathbf{r}'\mathbf{F}(\mathbf{r}-\mathbf{r}')g(\mathbf{r},\mathbf{r}') = \frac{1}{|\mathbf{r}|^{n_1}}\int d\mathbf{r}'\mathbf{F}(\mathbf{r}')h(\mathbf{r}') + R_1(\mathbf{r}) + R_2(\mathbf{r}) , \tag{2.31}$$

with $R_1(\mathbf{r})=o(1/|\mathbf{r}|^{n_1})$ and $R_2(\mathbf{r})=O(1/|\mathbf{r}|^{n_2+2-\epsilon})$.

Proof of the proposition. According to the inverse power-law expansion starting with $|\mathbf{r}|^{-3}$, the term (2.21b), a second derivative, is $O(|\mathbf{r}|^{-5})$. For the term (2.21c), one notes from the definition (2.16) and because of the neutrality and of the commutation $[Q(\mathbf{r}),Q(\mathbf{r}')]=0$, that one has the symmetry relation

$$eg(\mathbf{r},\mathbf{r}')=e\langle :N_\tau(\mathbf{r})Q_\tau(\mathbf{r}'):A\rangle_T$$
$$=\langle :Q_\tau(\mathbf{r})Q_\tau(\mathbf{r}'):A\rangle_T=eg(\mathbf{r}',\mathbf{r})\ . \tag{2.32}$$

This implies in turn $h(\mathbf{r}')=h(-\mathbf{r}')$ since one can write for fixed $\mathbf{r}'$

$$\begin{aligned}h(\mathbf{r}')&=\lim_{|\mathbf{r}|\to\infty}|\mathbf{r}|^{n_1}g(\mathbf{r},\mathbf{r}+\mathbf{r}')\\&=\lim_{|\mathbf{r}|\to\infty}|\mathbf{r}-\mathbf{r}'|^{n_1}g(\mathbf{r}-\mathbf{r}',\mathbf{r})\\&=\lim_{|\mathbf{r}|\to\infty}\left[\frac{|\mathbf{r}-\mathbf{r}'|}{|\mathbf{r}|}\right]^{n_1}\lim_{|\mathbf{r}|\to\infty}|\mathbf{r}|^{n_1}g(\mathbf{r}-\mathbf{r}',\mathbf{r})\\&=\lim_{|\mathbf{r}|\to\infty}|\mathbf{r}|^{n_1}g(\mathbf{r},\mathbf{r}-\mathbf{r}')=h(-\mathbf{r}')\ .\end{aligned} \tag{2.33}$$

Thus $\int d\mathbf{r}'\mathbf{F}(\mathbf{r}')h(\mathbf{r}')=0$ because of the antisymmetry of the force. Then the application of Lemma 1 with $n_1=n_2=3$ and $\epsilon<2$ to the integral in (2.21c) implies that this integral decays faster than $1/|\mathbf{r}|^3$ and hence its gradient faster than $1/|\mathbf{r}|^4$ (assuming monotonous decay).

Inserting the expansion (2.26) in Eq. (2.21), we conclude that the coefficients $w_n(\tau,A)$, $n=3,4$, obey the differential equation

$$\frac{d^2}{d\tau^2}w_n(\tau,A)-\hbar^2\omega_p^2w_n(\tau,A)=0,\quad 0\le\tau\le\beta\ . \tag{2.34}$$

Moreover, the continuity equation (2.11) implies with the assumption (i) that $(d/d\tau)w_3(\tau,A)=0$, and hence from (2.34) $w_3(\tau,A)=0$. This shows (2.29).

To obtain (2.30), we apply the KMS condition (2.8)

$$\langle Q_\beta(\mathbf{r})A\rangle_T-\langle Q(\mathbf{r})A\rangle_T=\langle[A,Q(\mathbf{r})]\rangle \tag{2.35}$$

and from (2.11)

$$\frac{d}{d\tau}\langle Q_\tau(\mathbf{r})A\rangle_T\bigg|_{\tau=\beta}-\frac{d}{d\tau}\langle Q_\tau(\mathbf{r})A\rangle_T\bigg|_{\tau=0}=i\hbar\nabla\cdot\langle[A,\mathbf{J}(\mathbf{r})]\rangle\ . \tag{2.36}$$

If A is local, the commutators $[A,Q(\mathbf{r})]$ and $[A,\mathbf{J}(\mathbf{r})]$ vanish when $|\mathbf{r}|$ is large enough. This implies that the coefficient $w_4(\tau,A)$ satisfies the boundary conditions

$$\begin{aligned}&w_4(\beta,A)=w_4(0,A)\ ,\\&\frac{d}{d\tau}w_4(\tau,A)\bigg|_{\tau=\beta}=\frac{d}{d\tau}w_4(\tau,A)\bigg|_{\tau=0}\ .\end{aligned} \tag{2.37}$$

The solution of Eq. (2.34) with the conditions (2.37) is $w_4(\tau,A)=0$, thus showing (2.30).

A stronger result can be obtained for the charge-charge correlation $\langle Q_\tau(\mathbf{r})Q(\mathbf{0})\rangle$. If we specify $A=Q(\mathbf{0})$ in Eq. (2.21), we observe that using the KMS relation and translation invariance, one can write

$$\langle K_\tau^{\mu\nu}(\mathbf{r})Q(\mathbf{0})\rangle_T=\langle Q_{\beta-\tau}(-\mathbf{r})K^{\mu\nu}(\mathbf{0})\rangle_T\ , \tag{2.38}$$

$$\begin{aligned}e\langle :N_\tau(\mathbf{r})Q_\tau(\mathbf{r}'):Q(\mathbf{0})\rangle_T&\\=\langle :Q_\tau(\mathbf{r})Q_\tau(\mathbf{r}'):Q(\mathbf{0})\rangle_T&\\=\langle Q_{\beta-\tau}(-\mathbf{r}):Q(\mathbf{0})Q(\mathbf{r}'-\mathbf{r}):\rangle_T&\ .\end{aligned} \tag{2.39}$$

The result (2.30) shows that the correlation (2.38) decays at least as $|\mathbf{r}|^{-5}$, and its second derivative [term (2.21b)] as $|\mathbf{r}|^{-7}$. By the same result (2.30), the correlation (2.39) does not decay slower than $|\mathbf{r}|^{-5}$ as $|\mathbf{r}|\to\infty$ with $|\mathbf{r}'-\mathbf{r}|$ fixed. Moreover, by (2.29) it decays not slower than t^{-4} as the distances $t=|\mathbf{r}'|$ or $t=|\mathbf{r}-\mathbf{r}'|$ tend to infinity. This allows us to apply the lemma with $n_1=5$, $n_2=4$, and $\epsilon=0$ to the integral occurring in (2.21c). Taking into account the symmetry (2.33), this integral decays faster than $|\mathbf{r}|^{-5}$ and its gradient faster than $|\mathbf{r}|^{-6}$. Thus the coefficients $w_n(\tau,Q(\mathbf{0}))$, $n=5,6$, in the asymptotic development (2.26) of $\langle Q_\tau(\mathbf{r})Q(\mathbf{0})\rangle$ obey also the second-order differential equation (2.34), as well as the boundary conditions (2.37) for the same locality reason as found from (2.35) and (2.36). Hence $w_n(\tau,Q(\mathbf{0}))=0$, $n=5,6$, implying

$$|\langle Q_\tau(\mathbf{r})Q(\mathbf{0})\rangle|=o\left[\frac{1}{|\mathbf{r}|^6}\right],\quad|\mathbf{r}|\to\infty\ . \tag{2.40}$$

In particular, the charge-charge correlation $S(\mathbf{r})=\langle Q(\mathbf{r})Q(\mathbf{0})\rangle$ decays faster than $|\mathbf{r}|^{-6}$. We emphasize again that the upperbounds (2.29), (2.30), and (2.40) should not be considered as rigorous results on the correlations of the quantum OCP. As already said in the beginning of Sec. II, they follow if we admit the validity of the hierarchy equations for the Green functions in the thermodynamic limit, integer inverse power-law decay and bounds having the structure (2.27), (2.28). But for the lack of any mathematically rigorous information on the decay of the correlations in quantum-mechanical Coulombic matter, we feel that it would be interesting to investigate what are the simplest conceivable compatible scenarios. In fact, one sees that the KMS equilibrium condition together with the locality play a nontrivial compelling role in reducing the bare Coulombic decay (as $|\mathbf{r}|^{-1}$) to the faster ones (2.29), (2.30), or (2.40), a manifestation of screening. In the classical case, the corresponding investigation of the equilibrium equations enables exclusion of any monotonous inverse power-law decay, allowing thus for exponential clustering.[16] In the quantum case, it does not appear possible to pursue the analysis beyond the present point. The calculation of the quantum corrections presented in the rest of the paper reveals indeed that all quantum-mechanical correlations have algebraic tails.

III. SUM RULES

A. Charge and dipole sum rules in the OCP

The existence of algebraic decay is intimately connected to the multipolar structure of the charge screening clouds. It is convenient to discuss the screening properties of these clouds in terms of the excess charge density at $\mathbf{r}$ when a configuration of charges is fixed at $\mathbf{r}_1, \mathbf{r}_2, \ldots \mathbf{r}_k$, $\mathbf{r}_1 \neq \mathbf{r}_2, \neq \cdots \neq \mathbf{r}_k$, defined by

$$C(\mathbf{r}|\mathbf{r}_1, \ldots, \mathbf{r}_k) = \langle Q(\mathbf{r}) N(\mathbf{r}_1) \ldots N(\mathbf{r}_k) \rangle \ . \tag{3.1}$$

The properly normalized excess charge density would be the quantity (3.1) divided by $\langle N(\mathbf{r}_1) \cdots N(\mathbf{r}_k) \rangle$. We recall the general theorem that exponential clustering implies the vanishing of all multipoles of the excess charge density (3.1), i.e.,

$$\int d\mathbf{r}\, \mathcal{Y}_l(\mathbf{r}) C(\mathbf{r}|\mathbf{r}_1, \ldots, \mathbf{r}_k) = 0 \tag{3.2}$$

for all harmonic polynomials $\mathcal{Y}_l(\mathbf{r})$ of degree $l = 0, 1, 2, \ldots$ and all particle configurations $\mathbf{r}_1, \ldots, \mathbf{r}_k$ (see Refs. 14 and 18 in the classical case and Ref. 15 in the quantum case). The sum rules (3.2) are indeed true in the Debye screening phase of a classical plasma where exponential clustering is known to take place.[5,6,19]

It turns out that the rules (3.2) are violated in the quantum OCP for $l \geq 2$ [the first quantum correction to (3.2) does not vanish, see Sec. IV C]. We must therefore conclude from the above-mentioned general theorem that the clustering of the quantum OCP cannot be exponentially fast. However, the charge ($l = 0$) and the dipole ($l = 1$) sum rule

$$\int d\mathbf{r}\, C(\mathbf{r}|\mathbf{r}_1, \ldots, \mathbf{r}_k) = 0 \ , \tag{3.3}$$

$$\int d\mathbf{r}\, \mathbf{r} C(\mathbf{r}|\mathbf{r}_1, \ldots, \mathbf{r}_k) = 0 \ , \tag{3.4}$$

hold in the OCP and can easily be derived from the equilibrium equations (2.11) and (2.21), assuming the validity of the analysis of Sec. II B. In particular, the decays found in Sec. II B ensure the convergence of spatial integrals and that integrals of gradient terms give no surface contributions.

To establish (3.3), we consider the more general correlations $\langle Q_\tau(\mathbf{r}) A \rangle_T$ where A is any local observable [(3.1) corresponds to $\tau = 0$ and $A = N(\mathbf{r}_1) \cdots N(\mathbf{r}_k)$] and note that the "continuity equation" (2.11) implies

$$\frac{d}{d\tau} \int d\mathbf{r} \langle Q_\tau(\mathbf{r}) A \rangle_T = 0 \ . \tag{3.5}$$

Moreover, Eq. (2.21) gives simply after integration on $\mathbf{r}$

$$\frac{d^2}{d\tau^2} \int d\mathbf{r} \langle Q_\tau(\mathbf{r}) A \rangle_T - \hbar^2 \omega_p^2 \int dr \langle Q_\tau(\mathbf{r}) A \rangle_T = 0 \ . \tag{3.6}$$

The combination of (3.5) and (3.6) leads to

$$\int d\mathbf{r} \langle Q_\tau(\mathbf{r}) A \rangle_T = 0 \ , \tag{3.7}$$

giving the charge sum rule (3.3) as a special case.

To establish (3.4), we remark from (2.21) that the dipole of $\langle Q_\tau(\mathbf{r}) A \rangle_T$ obeys the second-order differential equation

$$\frac{d^2}{d\tau^2} \int d\mathbf{r}\, \mathbf{r} \langle Q_\tau(r) A \rangle_T - \hbar^2 \omega_p^2 \int d\mathbf{r}\, \mathbf{r} \langle Q_\tau(\mathbf{r}) A \rangle_T = 0 \ . \tag{3.8}$$

This is because (2.21b) and (2.21c) do not contribute after integration by parts. (Note that

$$\int d\mathbf{r} \int d\mathbf{r}' \mathbf{F}(\mathbf{r} - \mathbf{r}') g(\mathbf{r}, \mathbf{r}') = 0$$

since $g(\mathbf{r}, \mathbf{r}')$ is symmetric [see (2.32)] and $\mathbf{F}(\mathbf{r} - \mathbf{r}')$ is antisymmetric.) One deduces from the KMS condition that

$$\int d\mathbf{r}\, \mathbf{r} \langle Q_\beta(\mathbf{r}) A \rangle_T - \int d\mathbf{r}\, \mathbf{r} \langle Q(\mathbf{r}) A \rangle_T = \int d\mathbf{r}\, \mathbf{r} \langle [A, Q(\mathbf{r})] \rangle_T \tag{3.9}$$

and from (2.11), using the KMS condition again,

$$\begin{aligned} \frac{d}{d\tau} \int d\mathbf{r}\, \mathbf{r} \langle Q_\tau(\mathbf{r}) A \rangle_T \Big|_{\tau=\beta} - \frac{d}{d\tau} \int d\mathbf{r}\, \mathbf{r} \langle Q_\tau(\mathbf{r}) A \rangle_T \Big|_{\tau=0} &= i\hbar \int d\mathbf{r}\, \mathbf{r} \{ \nabla \cdot \langle [A, \mathbf{J}(\mathbf{r})] \rangle_T \} \\ &= -i\hbar \int d\mathbf{r} \langle [A, \mathbf{J}(\mathbf{r})] \rangle_T = 0 \ . \end{aligned} \tag{3.10}$$

This last integral vanishes because of the translational invariance of the state. Indeed, from the definition (2.2), $\int d\mathbf{r}\, \mathbf{J}(\mathbf{r}) = (e/m) \mathbf{P}$ is proportional to the formal generator of space translations $\mathbf{P}$. If $A(\mathbf{y})$ is the space translate of A to the point $\mathbf{y}$, one has

$$\int d\mathbf{r} \langle [A, \mathbf{J}(\mathbf{r})] \rangle_T = \frac{e}{m} \langle [A, \mathbf{P}] \rangle = i \frac{e}{m} \nabla \langle A(\mathbf{y}) \rangle = 0 \tag{3.11}$$

since $\langle A(\mathbf{y}) \rangle$ is constant in a homogeneous state. For a local A, the commutator $[A, \mathbf{P}]$ is still a local observable, and the average $\langle [A, \mathbf{P}] \rangle$ is well defined. The solution of Eq. (3.8) with the conditions (3.9) and (3.10) is

$$\int d\mathbf{r}\, \mathbf{r} \langle Q_\tau(\mathbf{r}) A \rangle_T = \frac{1}{2} \left[\frac{e^{\hbar\omega_p \tau}}{e^{\hbar\omega_p \beta} - 1} - \frac{e^{-\hbar\omega_p \tau}}{1 - e^{-\hbar\omega_p \beta}} \right] \int d\mathbf{r}\, \mathbf{r} \langle [A, Q(\mathbf{r})] \rangle_T \ . \tag{3.12}$$

If A is a purely configurational observable, it commutes with $Q(\mathbf{r})$, and one finds

$$\int d\mathbf{r}\, \mathbf{r} \langle Q_\tau(\mathbf{r}) A \rangle_T = 0 \quad \text{when } [A, Q(\mathbf{r})] = 0 \tag{3.13}$$

with (3.4) as a special case.

The right-hand side of (3.12) does not vanish in general. For instance, if $A = J^\nu(0)$ is the ν component of the current (2.2), one obtains from the canonical commutation relations

$$\int d\mathbf{r}\, r^{\mu}\langle [J^{\nu}(0),Q(\mathbf{r})]\rangle = -i\hbar\frac{e^2}{m}\rho\delta\mu\nu \;, \tag{3.14}$$

leading to the sum rule for the charge-current correlations

$$\int d\mathbf{r}\, r^{\mu}\langle Q_{\tau}(\mathbf{r})J^{\nu}(0)\rangle = -i\delta\mu\nu\frac{\hbar e^2}{2m}\rho\left[\frac{e^{\hbar\omega_p\tau}}{e^{\hbar\omega_p\beta}-1}-\frac{e^{-\hbar\omega_p\tau}}{1-e^{-\hbar\omega_p\beta}}\right] . \tag{3.15}$$

We remark finally from (3.12) that the response function $\int_0^{\beta}d\tau\langle Q_{\tau}(\mathbf{r})A\rangle_T$ has no dipole

$$\int d\mathbf{r}\,\mathbf{r}\int_0^{\beta}d\tau\langle Q_{\tau}(\mathbf{r})A\rangle_T=0 \tag{3.16}$$

for any local A.

B. Second moment

The second moment of the imaginary-time charge-charge correlation obeys again the simple differential equation

$$\frac{d^2}{d\tau^2}\int d\mathbf{r}|\mathbf{r}|^2\langle Q_{\tau}(\mathbf{r})Q(0)\rangle_T-\hbar^2\omega_p^2\int d\mathbf{r}|\mathbf{r}|^2\langle Q_{\tau}(\mathbf{r})Q(0)\rangle_T=0 \;. \tag{3.17}$$

This follows from Eq. (2.21) [with $A=Q(0)$] once multiplied by $|\mathbf{r}|^2$ and integrated on $\mathbf{r}$. The terms (2.21b) and (2.21c) do not contribute. After integration by parts, using translation invariance and the KMS condition, one finds

$$\int d\mathbf{r}|\mathbf{r}|^2\nabla^{\mu}\nabla^{\nu}\langle K_{\tau}^{\mu\nu}(\mathbf{r})Q(0)\rangle_T=2\delta\mu\nu\int d\mathbf{r}\langle Q_{\beta-\tau}(\mathbf{r})K^{\mu\mu}(0)\rangle_T \;, \tag{3.18}$$

$$\int d\mathbf{r}|\mathbf{r}|^2\nabla\cdot\int d\mathbf{r}'\mathbf{F}(\mathbf{r}-\mathbf{r}')\langle :N_{\tau}(\mathbf{r})Q_{\tau}(\mathbf{r}'):Q(0)\rangle_T=2\int d\mathbf{r}'\mathbf{F}(\mathbf{r}')\cdot\int d\mathbf{r}\,\mathbf{r}\langle Q_{\beta-\tau}(\mathbf{r}):N(0)Q(\mathbf{r}')\rangle_T \;. \tag{3.19}$$

Both (3.18) and (3.19) vanish because of the sum rules (3.7) and (3.13). Two conditions are needed to determine uniquely the solution of the second-order differential equation (3.17). The first one follows from the KMS relation and from translation and rotation invariance,

$$\int d\mathbf{r}|\mathbf{r}|^2\langle Q_{\beta}(\mathbf{r})Q(0)\rangle_T=\int d\mathbf{r}|\mathbf{r}|^2\langle Q(\mathbf{r})Q(0)\rangle_T \;. \tag{3.20}$$

A second condition is obtained by integrating (3.17) on $\tau(0\le\tau\le\beta)$ and applying successively the continuity equation (2.11), the KMS relation, and the identity (3.14),

$$\begin{aligned}\int d\mathbf{r}|\mathbf{r}|^2\int_0^{\beta}d\tau\langle Q_{\tau}(\mathbf{r})Q(0)\rangle_T&=\frac{i}{\hbar\omega_p^2}\int d\mathbf{r}|\mathbf{r}|^2\nabla\cdot\langle \mathbf{J}_{\beta}(\mathbf{r})Q(0)-\mathbf{J}(\mathbf{r})Q(0)\rangle\\&=-\frac{2i}{\hbar\omega_p^2}\int d\mathbf{r}\,\mathbf{r}\cdot\langle[\mathbf{J}(\mathbf{r}),Q(0)]\rangle=-\frac{3}{2\pi}\;.\end{aligned} \tag{3.21}$$

The solution of (3.17) under the conditions (3.20) and (3.21) is

$$\int d\mathbf{r}|\mathbf{r}|^2\langle Q_{\tau}(\mathbf{r})Q(0)\rangle_T = -\frac{3}{4\pi}\hbar\omega_p\left[\frac{e^{\hbar\omega_p\tau}}{e^{\hbar\omega_p\beta}-1}+\frac{e^{-\hbar\omega_p\tau}}{1-e^{-\hbar\omega_p\beta}}\right] . \tag{3.22}$$

Equation (3.22) is the generalization for arbitrary τ of a known sum rule for the static structure function $S(\mathbf{r})=\langle Q(\mathbf{r})Q(0)\rangle_T$ of the OCP, usually derived by linear-response arguments,[3]

$$\int d\mathbf{r}|\mathbf{r}|^2S(\mathbf{r})=-\frac{3}{4\pi}\hbar\omega_p\coth\left[\frac{\hbar\omega_p\beta}{2}\right] . \tag{3.23}$$

We note that Eq. (3.21) is a sum rule for the quantum-mechanical response function

$$\chi(\mathbf{r})=\int_0^{\beta}d\tau\langle Q_{\tau}(\mathbf{r})Q(0)\rangle$$

to an infinitesimal static external point charge in the OCP. In linear response, Eq. (3.21) expresses the shielding of this external charge and is the quantum analog of the classical Stillinger-Lovett second moment condition. In the classical limit, the response function $\chi(\mathbf{r})$ and the structure function $S(\mathbf{r})$ are proportional and both (3.21) and (3.22) reduce to the classical Stillinger-Lovett condition. [For alternative derivations of (3.23) and (3.21), see Refs. 20 and 21.]

IV. WIGNER-KIRKWOOD EXPANSION

A. General formalism

In this section we compute the first terms of the $\hbar$ expansion of the static charge-charge correlation function

$$S^{\mathrm{qm}}(\mathbf{r})=\langle Q(\mathbf{r})Q(0)\rangle^{\mathrm{qm}}=e^2\langle N(\mathbf{r})N(0)\rangle_T^{\mathrm{qm}} \;, \tag{4.1}$$

$$S^{\mathrm{qm}}(\mathbf{r})=S^{(0)}(\mathbf{r})+\hbar^2S^{(2)}(\mathbf{r})+\hbar^4S^{(4)}(\mathbf{r})+\cdots \;. \tag{4.2}$$

For now on, quantities with the index qm are quantum mechanical, and quantities without the index qm are classical, e.g., $\langle\ \rangle^{\rm qm}$ and $\langle\ \rangle$ denote, respectively, the quantum and classical averages. In (4.2), $S^{(0)}(\mathbf{r}) = S(\mathbf{r}) = \langle Q(\mathbf{r})Q(\mathbf{0})\rangle$ is the corresponding classical charge-charge correlation function of the OCP and $S^{(2)}(\mathbf{r}), S^{(4)}(\mathbf{r}), \ldots$ are also expressible in terms of higher-order distribution functions $\rho(\mathbf{r}_1, \ldots, \mathbf{r}_k)$, $k=1,2,\ldots$, of the classical OCP. The second-order term has been calculated[22]

$$S^{(2)}(\mathbf{r}) = \frac{\beta e^2}{12m}\nabla^2\rho_T(\mathbf{r},\mathbf{0}) \tag{4.3}$$

and decays fast when $\rho_T(\mathbf{r},\mathbf{0})$ does so. Our main result is that even in the regime where all classical truncated (Ursell) correlation functions of the OCP decay exponentially fast, the fourth-order term is algebraic,

$$S^{(4)}(\mathbf{r}) \sim \frac{7}{16\pi^2}\left[\frac{\beta e}{m}\right]^2 \frac{1}{|\mathbf{r}|^{10}}, \quad |\mathbf{r}| \to \infty \ . \tag{4.4}$$

To establish (4.4), we proceed as follows. We consider first a quantum OCP with a finite number N of charges in $\mathbb{R}^3$ submitted to the potential of a uniform infinitely extended background of density $-e\rho$. The Hamiltonian of this system is

$$H_N = -\frac{\hbar^2}{2m}\sum_{i=1}^{N}\nabla_i^2 + V(\mathbf{r}_1, \ldots, \mathbf{r}_N) \ , \tag{4.5}$$

$$V(\mathbf{r}_1, \ldots, \mathbf{r}_N) = e^2\tfrac{1}{2}\sum_{\substack{i=1\\ i\neq j}}^{N}\phi(\mathbf{r}_i - \mathbf{r}_j) + e\sum_{j=1}^{N}\phi_b(\mathbf{r}_i) \ . \tag{4.6}$$

$\phi(\mathbf{r}) = 1/|\mathbf{r}|$ is the Coulomb potential and H_N acts on $\mathcal{L}^2(\mathbb{R}^{3N})$. In (4.6), the background potential formally given by $\phi_b(\mathbf{r}) = -e\rho\int d\mathbf{r}'\phi(\mathbf{r}-\mathbf{r}')$ is defined as the spherically symmetric solution of the corresponding Poisson equation $\nabla^2\phi_b(\mathbf{r}) = 4\pi e\rho$, i.e. (up to an irrelevant additive constant),

$$\phi_b(\mathbf{r}) = \frac{2\pi}{3}e\rho|\mathbf{r}|^2 \ . \tag{4.7}$$

This potential confines the particles in a bounded spatial region and no finite distance boundary conditions are needed for the Laplacian. Thus the equilibrium charge-charge correlations

$$\langle Q(\mathbf{r})Q(\mathbf{r}')\rangle^{\rm qm} = \frac{\mathrm{Tr}[Q(\mathbf{r})Q(\mathbf{r}')\exp(-\beta H_N)]}{\mathrm{Tr}\exp(-\beta H_N)} \ , \tag{4.8}$$

with

$$Q(\mathbf{r}) = e[N(\mathbf{r}) - \rho], \quad N(\mathbf{r}) = \sum_{i=1}^{N}\delta(\mathbf{r}-\mathbf{r}_i) \ ,$$

are well defined for finite N, as are the higher-order correlation functions. The same remark applies to the corresponding classical correlations, e.g.,

$$\langle Q(\mathbf{r})Q(\mathbf{r}')\rangle = \frac{\int d\mathbf{r}_1\cdots\int d\mathbf{r}_N Q(\mathbf{r})Q(\mathbf{r}')\exp[-\beta V(\mathbf{r}_1, \ldots, \mathbf{r}_N)]}{\int d\mathbf{r}_1\cdots d\mathbf{r}_N\exp[-\beta V(\mathbf{r}_1, \ldots, \mathbf{r}_N)]} \ , \tag{4.9}$$

which converge as $N\to\infty$ to those of the classical translationally invariant and locally neutral OCP with density ρ. The background potential acts as a soft wall. In the plasma phase, the bulk correlations are independent of the boundary conditions in the thermodynamic limit, as it can be checked in a solvable model.[23] To obtain (4.2) we shall use the Wigner-Kirkwood (WK) formalism, expanding the quantum-mechanical functions in powers of $\hbar$ and then letting $N\to\infty$ term by term.[24]

For a Hamiltonian of the form (4.5) with a general potential energy $V(\mathbf{r}_1, \ldots, \mathbf{r}_N)$ the first terms of the Wigner-Kirkwood expansion are given by the formulas (4.10)–(4.13) below. To abbreviate the notation, one sets

$$\mathbf{R} = \{r_i^\mu, i=1,\ldots,N, \mu=1,2,3\} = \{R_p, p=1,\ldots,3N\}$$

a point in configuration space $\mathbb{R}^{3N}$ where $i=1,\ldots,N$, $\mu=1,2,3$ are, respectively, the particle and vector indices. The corresponding gradient on $\mathbb{R}^{3N}$ is

$$\boldsymbol{\nabla} = \{\nabla_i^\mu = \partial/\partial r_i^\mu, i=1,\ldots,N, \mu=1,2,3\} = \{\nabla_p = \partial/\partial R_p, p=1,\ldots,3N\} \ .$$

Then, the diagonal part of the kernel of $e^{-\beta H}$ is expanded in powers of $\hbar$ with the result [formula (2.10) of Refs. 25 and 26]

$$(\mathbf{R}|e^{-\beta H_N}|\mathbf{R}) = \left[\frac{1}{\sqrt{2\pi}\lambda}\right]^{3N}[1+G(\mathbf{R})]e^{-\beta V(\mathbf{R})} \ , \tag{4.10}$$

$$G(\mathbf{R}) = \hbar^2 G^{(2)}(\mathbf{R}) + \hbar^4 G^{(4)}(\mathbf{R}) + \cdots \ , \tag{4.11}$$

$$G^{(2)} = \frac{\beta}{m}\left[-\tfrac{1}{12}\beta\nabla^2 V + \tfrac{1}{24}\beta^2(\boldsymbol{\nabla}V)^2\right] \ , \tag{4.12}$$

$$\begin{aligned} G^{(4)} = \tfrac{1}{2}\left[\frac{\beta}{24m}\right]^2 \{&4\beta^2(\nabla^2 V)^2 - 4\beta^3(\nabla^2 V)(\boldsymbol{\nabla}V)^2 + \beta^4[(\boldsymbol{\nabla}V)^2]^2 - \tfrac{24}{5}\beta(\nabla^2)^2 V + \tfrac{32}{5}\beta^2\boldsymbol{\nabla}V\cdot\boldsymbol{\nabla}(\nabla^2 V) \\ &+ \tfrac{8}{5}\beta^2\nabla^2(\boldsymbol{\nabla}V)^2 - \tfrac{12}{5}\beta^3\boldsymbol{\nabla}V\cdot\boldsymbol{\nabla}(\boldsymbol{\nabla}V)^2\} \ . \end{aligned} \tag{4.13}$$

In (4.10), $\lambda=(\hbar^2\beta/m)^{1/2}$ is the thermal de Broglie wave length. In (4.12) and (4.13), the dot means the scalar product on $\mathbb{R}^{3N}$, $(\nabla V)^2=\nabla V\cdot\nabla V$ and $\nabla^2=\nabla\cdot\nabla$ is the Laplacian on $\mathbb{R}^{3N}$.

If $A=A(\mathbf{R})$ is a configurational observable [acting as a multiplication operator on $\mathcal{L}^2(\mathbb{R}^{3N})$], its quantum-mechanical average $\langle A\rangle^{\mathrm{qm}}$ is first developed around its classical value in powers of the quantum correction (4.11). This gives (dropping the index N from now on)

$$\begin{aligned}\langle A\rangle^{\mathrm{qm}}&=\frac{\mathrm{Tr}\,A\exp(-\beta H)}{\mathrm{Tr}\exp(-\beta H)}\\&=\frac{\int d\mathbf{R}\,A(\mathbf{R})(\mathbf{R}|e^{-\beta H}|\mathbf{R})}{\int d\mathbf{R}(\mathbf{R}|e^{-\beta H}|\mathbf{R})}\\&=\frac{\langle A\rangle+\langle AG\rangle}{1+\langle G\rangle}\\&=(\langle A\rangle+\langle AG\rangle)(1-\langle G\rangle+\langle G\rangle^2+\cdots)\ ,\end{aligned}\tag{4.14}$$

where $\langle\ \rangle$ denote the corresponding classical averages. Inserting the expansion (4.11) for G in (4.14) and collecting the powers of $\hbar$ leads to

$$\langle A\rangle^{\mathrm{qm}}=\langle A\rangle+\hbar^2\langle A\rangle^{(2)}+\hbar^4\langle A\rangle^{(4)}+\cdots\ ,\tag{4.15}$$

$$\langle A\rangle^{(2)}=\langle AG^{(2)}\rangle-\langle A\rangle\langle G^{(2)}\rangle\ ,\tag{4.16}$$

$$\langle A\rangle^{(4)}=\langle AG^{(4)}\rangle-\langle A\rangle\langle G^{(4)}\rangle-\langle A\rangle^{(2)}\langle G^{(2)}\rangle\ ,\tag{4.17}$$

where $G^{(2)}$ and $G^{(4)}$ are defined by (4.12) and (4.13).

Applying the general formalism to the case where $A=Q(\mathbf{r}_1)Q(\mathbf{r}_2)$ is the product of charge densities at $\mathbf{r}_1$ and $\mathbf{r}_2$, the second- and fourth-order corrections (4.2) are

$$S^{(k)}(\mathbf{r})=\lim_{N\to\infty}\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle^{(k)}\ ,\qquad \mathbf{r}=\mathbf{r}_1-\mathbf{r}_2,\quad k=2,4\tag{4.18}$$

with

$$\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle^{(2)}=\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)G^{(2)}\rangle-\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle\langle G^{(2)}\rangle\ ,\tag{4.19}$$

$$\begin{aligned}\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle^{(4)}=&\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)G^{(4)}\rangle\\&-\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle\langle G^{(4)}\rangle\\&-\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle^{(2)}\langle G^{(2)}\rangle\ .\end{aligned}\tag{4.20}$$

According to (4.13) and if $V(\mathbf{r}_1,\ldots,\mathbf{r}_N)$ has the form (4.6), one notices that $G^{(4)}$ involves the fourth power of the two-body Coulomb potential, i.e., an eight-body observable. Therefore the term (4.20) involves, in principle, classical correlations up to order 10. Fortunately, after appropriate reduction of the formulas, and using specific properties of the OCP, this number can be reduced to 5.

B. Reduction of the formulas

In the OCP, the formulas simplify considerably with the observation that the Gibbs factor $\exp[-\beta V(\mathbf{r}_1,\ldots,\mathbf{r}_N)]$ vanishes faster than any inverse power when two arguments coincide as a consequence of the infinite repulsion between two particles at the same point. This allows us to apply the following rule: for any pair of particles i,j

$$\delta(\mathbf{r}_i-\mathbf{r}_j)e^{-\beta V(\mathbf{r}_1,\ldots,\mathbf{r}_N)}=0\ ,\qquad i\neq j,\quad i,j=1,\ldots,N\tag{4.21}$$

and the same rule holds for the product of any derivative of $\delta(\mathbf{r}_i-\mathbf{r}_j)$ with the Gibbs factor.

Consider first the second-order term (4.12). The square of the potential can be eliminated with the help of the identity

$$\nabla^2e^{-\beta V}=-\beta(\nabla^2V)e^{-\beta V}+\beta^2(\nabla V)^2e^{-\beta V}\ .\tag{4.22}$$

Introducing (4.22) with the Poisson equation

$$\nabla^2V=-4\pi e^2\sum_{i\neq j}^{N}\delta(\mathbf{r}_i-\mathbf{r}_j)+4\pi e^2\rho N\tag{4.23}$$

and applying the rule (4.21), Eq. (4.12) takes the simpler form

$$G^{(2)}=\frac{\beta}{24m}(\nabla^2-4\pi e^2\rho N\beta)\ .\tag{4.24}$$

In (4.24), the Laplacian ∇^2 is understood as acting on the Gibbs factor $e^{-\beta V}$ when calculating a classical average.

We proceed in the same way for the fourth-order term. We first eliminate in (4.13) the highest power $[(\nabla V)^2]^2$ of the interaction by means of the identity

$$\begin{aligned}(\nabla^2)^2e^{-\beta V}=\{&-\beta(\nabla^2)^2V+2\beta^2\nabla(\nabla^2V)\cdot\nabla V+\beta^2(\nabla^2V)^2+\beta^2\nabla^2(\nabla V)^2-2\beta^3(\nabla^2V)(\nabla V)^2\\&-2\beta^3\nabla(\nabla V)^2\cdot\nabla V+\beta^4[(\nabla V)^2]^2\}e^{-\beta V}\ ,\end{aligned}\tag{4.25}$$

obtaining in this way

$$\begin{aligned}G^{(4)}=\frac{1}{2}\left[\frac{\beta}{24m}\right]^2&[3\beta^2(\nabla^2V)^2-2\beta^3(\nabla^2V)(\nabla V)^2-\tfrac{19}{5}\beta(\nabla^2)^2V+\tfrac{22}{5}\beta^2\nabla V\cdot\nabla(\nabla^2V)+\tfrac{3}{5}\beta^2\nabla^2(\nabla V)^2\\&-\tfrac{2}{5}\beta^3\nabla V\cdot\nabla(\nabla V)^2+(\nabla^2)^2]\ .\end{aligned}\tag{4.26}$$

Then the factor $(\nabla V)^2$ in the second term of the right-hand side of (4.26) is also eliminated with the help of (4.22), leading to

$$G^{(4)}=\frac{1}{2}\left[\frac{\beta}{24m}\right]^2[\beta^2(\nabla^2 V)^2-\tfrac{19}{5}\beta(\nabla^2)^2V+\tfrac{22}{5}\beta^2\nabla V\cdot\nabla(\nabla^2 V)+\tfrac{3}{5}\beta^2\nabla^2(\nabla V)^2-\tfrac{2}{5}\beta^3\nabla V\cdot\nabla(\nabla V)^2-2\beta(\nabla^2 V)\nabla^2+(\nabla^2)^2] \; . \tag{4.27}$$

As in (4.24), the differential operators occurring in the last terms of (4.26) and (4.27) act on $e^{-\beta V}$ when calculating a classical average.

Let us examine in more detail the terms $\nabla^2(\nabla V)^2$ and $\nabla V\cdot\nabla(\nabla V)^2$ in (4.27). One has

$$\nabla^2(\nabla V)^2=\sum_{p,q}\nabla_p^2(\nabla_q V)^2=2\sum_{p,q}\nabla_p[(\nabla_p\nabla_q V)\nabla_q V]=2\nabla(\nabla^2 V)\cdot\nabla V+2\sum_{p,q}(\nabla_p\nabla_q V)^2 \; , \tag{4.28}$$

where p,q run on all components in $\mathbb{R}^{3N}$. With the identity

$$\nabla_p\nabla_q e^{-\beta V}=-\beta(\nabla_p\nabla_q V)e^{-\beta V}+\beta^2(\nabla_p V)(\nabla_q V)e^{-\beta V} \tag{4.29}$$

the term $\nabla V\cdot\nabla(\nabla V)^2$ is transformed to

$$\nabla V\cdot\nabla(\nabla V)^2=\sum_{p,q}(\nabla_p V)\nabla_p(\nabla_q V)^2=2\sum_{p,q}(\nabla_p V)(\nabla_q V)(\nabla_p\nabla_q V)=\frac{2}{\beta}\sum_{p,q}(\nabla_p\nabla_q V)^2+\frac{2}{\beta^2}\sum_{p,q}(\nabla_p\nabla_q V)\nabla_p\nabla_q \; . \tag{4.30}$$

All the other terms in (4.27) (except the last one) involve the Laplacian of the potential $\nabla^2 V$. By (4.23) and the rule (4.21), we can replace everywhere $\nabla^2 V$ by $4\pi e^2\rho N$ (and hence $[(\nabla^2)]^2 V$ and $\nabla(\nabla^2 V)$ by zero). With this, and together with (4.28) and (4.30), the expression (4.27) takes the final form

$$G^{(4)}=\frac{1}{5}\frac{\beta^4}{(24m)^2}\sum_{p,q}(\nabla_p\nabla_q V)^2 \tag{4.31a}$$

$$+\frac{1}{2}\frac{\beta^4}{(24m)^2}(4\pi e^2\rho N)^2 \tag{4.31b}$$

$$-\frac{\beta^3}{(24m)^2}4\pi e^2\rho N\nabla^2 \tag{4.31c}$$

$$+\frac{1}{2}\left[\frac{\beta}{24m}\right]^2\left[(\nabla^2)^2-\tfrac{4}{5}\beta\sum_{p,q}(\nabla_p\nabla_q V)\nabla_p\nabla_q\right] . \tag{4.31d}$$

C. The second order

We consider the second-order term in the usual quantum configurational k point distribution functions $\rho^{\rm qm}(\mathbf{r}_1,\ldots,\mathbf{r}_k)$ defined by

$$\rho^{\rm qm}(\mathbf{r}_1,\ldots,\mathbf{r}_k)=\langle[N(\mathbf{r}_1)\cdots N(\mathbf{r}_k)]_{\rm nc}\rangle^{\rm qm} \; , \tag{4.32}$$

$$\rho^{\rm qm}(\mathbf{r}_1,\ldots,\mathbf{r}_k)=\rho(\mathbf{r}_1,\ldots,\mathbf{r}_k)+\hbar^2\rho^{(2)}(\mathbf{r}_1,\ldots,\mathbf{r}_k)+\cdots \; . \tag{4.33}$$

The notation $[N(\mathbf{r}_1)\cdots N(\mathbf{r}_k)]_{\rm nc}$ means that the contribution of coincident points $\mathbf{r}_i=\mathbf{r}_j$, $i,j=1,\ldots,k$, is not included, and $\rho(\mathbf{r}_1,\ldots,\mathbf{r}_k)$ are the corresponding classical functions. The correction $\rho^{(2)}(\mathbf{r}_1,\ldots,\mathbf{r}_k)$ is obtained from (4.16) and (4.24) with the choice $A=[N(\mathbf{r}_1)\cdots N(\mathbf{r}_k)]_{\rm nc}$.

We first note that the integral of any derivative or product of derivatives of the Gibbs factor vanishes,

$$\int d\mathbf{r}_i(\nabla_i^\mu\nabla_i^\nu\cdots)e^{-\beta V(\mathbf{r}_1,\ldots,\mathbf{r}_i,\ldots,\mathbf{r}_N)}=0 \; , \tag{4.34}$$

since $e^{-\beta V}$ decreases as a Gaussian in every direction in $\mathbb{R}^{3N}$ [see (4.7)]. So we find from (4.16) and (4.24)

$$\langle G^{(2)}\rangle=-\frac{\pi}{6m}\beta^2 e^2\rho N \; , \tag{4.35}$$

$$\rho^{(2)}(\mathbf{r}_1,\ldots,\mathbf{r}_k)=\frac{\beta}{24m}\sum_{j=1}^{k}\nabla_j^2\rho(\mathbf{r}_1,\ldots,\mathbf{r}_k) \; . \tag{4.36}$$

There are no quantum corrections to the one-point function to any order since its value is fixed to ρ by the local neutrality. For the two-point function, one recovers immediately the result (4.3) if one uses translation invariance.

Let us calculate from (4.36) the second-order quantum correction $c^{(2)}(\mathbf{r}|\mathbf{r}_1,\ldots,\mathbf{r}_k)$ to the classical excess charge density (3.1) for an arbitrary particle configuration $\mathbf{r}_1,\ldots,\mathbf{r}_k$,

$$c^{(2)}(\mathbf{r}|\mathbf{r}_1,\ldots,\mathbf{r}_k)=e\rho^{(2)}(\mathbf{r},\mathbf{r}_1,\ldots,\mathbf{r}_k)-e\rho\rho^{(2)}(\mathbf{r}_1,\ldots,\mathbf{r}_k)+e\sum_{j=1}^{k}\delta(\mathbf{r}-\mathbf{r}_j)\rho^{(2)}(\mathbf{r}_1,\ldots,\mathbf{r}_k) \; . \tag{4.37}$$

Using (4.36), $\nabla_j^2\mathcal{Y}_l(\mathbf{r}_j)=0$, and the fact that the classical excess charge density verifies the multipole sum rules (3.2), one finds

$$\int d\mathbf{r}\,\mathcal{Y}_l(\mathbf{r})c^{(2)}(\mathbf{r}|\mathbf{r}_1,\ldots,\mathbf{r}_k)=-\frac{\beta e}{12m}\sum_{j=1}^{k}\nabla_j\mathcal{Y}_l(\mathbf{r}_j)\cdot\nabla_j\rho(\mathbf{r}_1,\ldots,\mathbf{r}_k) \; . \tag{4.38}$$

The right-hand side of (4.38) is different from zero only if $l\neq 0,1$ [$\sum_{j=1}^{k}\nabla_j\rho(\mathbf{r}_1,\ldots,\mathbf{r}_k)=0$ because of translation invariance]. Thus, only the charge and dipole sum rules are verified at the order $\hbar^2$, a result compatible with Eqs. (3.3) and

(3.4) of Sec. III A. No higher-order sum rules hold in the quantum OCP for the excess charge density (3.1).

Finally, it is also possible to find the second-order term of the excess charge density $c_\tau(\mathbf{r}|\mathbf{r}_1,\ldots,\mathbf{r}_k)=\langle Q_\tau(\mathbf{r})N(\mathbf{r}_1)\cdots N(\mathbf{r}_k)\rangle$ at a nonzero imaginary time τ. A calculation left to the reader shows that (4.38) must be modified to

$$\int d\mathbf{r}\,\mathcal{Y}_l(\mathbf{r})c_\tau^{(2)}(\mathbf{r}|\mathbf{r}_1,\ldots,\mathbf{r}_k)=\left[\frac{\tau(\beta-\tau)}{2\beta}-\beta\right]\frac{e}{12m}\sum_{j=1}^{k}\nabla_j\mathcal{Y}_l(\mathbf{r}_j)\cdot\nabla_j\rho(\mathbf{r}_1,\ldots,\mathbf{r}_k)\ . \tag{4.39}$$

The rhs of (4.39) does not vanish for $1\geq 2$ and $0\leq\tau\leq\beta$. But it is interesting to observe that

$$\int_0^\beta d\tau\left[\frac{\tau(\beta-\tau)}{2}-\beta\right]=0$$

and therefore the response function $\int_0^\beta d\tau\langle Q_\tau(\mathbf{r})N(\mathbf{r}_1)\cdots N(\mathbf{r}_k)\rangle$ obeys all the multipole sum rules to order $\hbar^2$. This is an indication that the response function of the quantum system to classical external charges might have better screening and cluster properties than the Green's functions themselves.

D. The fourth order

We now insert (4.24), (4.31), and (4.35) in (4.20). The term (4.31b) drops because of the truncation and the term (4.31c) is canceled by the last term in the right-hand side of (4.20):

$$\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle^{(4)}=\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle_a^{(4)}+\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle_b^{(4)}\ ,$$

$$\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle_a^{(4)}=\frac{1}{5}\frac{\beta^4}{(24m)^2}\left[\left\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\sum_{p,q}(\nabla_p\nabla_q V)^2\right\rangle-\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle\left\langle\sum_{p,q}(\nabla_p\nabla_q V)^2\right\rangle\right]\ , \tag{4.40a}$$

$$\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle_b^{(4)}=\frac{1}{2}\left[\frac{\beta}{24m}\right]^2\left[\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)(\nabla^2)^2\rangle-\frac{4}{5}\beta\left[\left\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\sum_{p,q}(\nabla_p\nabla_q V)\nabla_p\nabla_q\right\rangle\right.\right.$$
$$\left.\left.-\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle\left\langle\sum_{p,q}(\nabla_p\nabla_q V)\nabla_p\nabla_q\right\rangle\right]\right]\ . \tag{4.40b}$$

The expression (4.40b), involving derivatives of the Gibbs factor, is shown to have a fast decay (Appendix B). The algebraic tail comes from the term (4.40a), which we now investigate more closely.

The first step is to single out the contribution of coincident particles in the second derivatives of the total potential (4.6)

$$\nabla_i^\mu\nabla_j^\nu V=\begin{cases}-e^2\phi^{\mu\nu}(\mathbf{r}_i-\mathbf{r}_j), & i\neq j\\ e^2\displaystyle\sum_{\substack{k=1\\k\neq i}}^{N}\phi^{\mu\nu}(\mathbf{r}_i-\mathbf{r}_k)-e^2\rho\int d\mathbf{r}\,\phi^{\mu\nu}(\mathbf{r}-\mathbf{r}_i), & i=j\end{cases} \tag{4.41}$$

where $\phi^{\mu\nu}(\mathbf{r})=(\nabla^\mu\nabla^\nu\phi)(\mathbf{r})$ is the dipole-dipole potential. This allows us to express $\sum_{p,q}(\nabla_p\nabla_q V)^2$ in terms of the charge density $Q(\mathbf{r})$ and particle densities $N(\mathbf{r})$ as a sum of a two- and a three-point contribution,

$$\sum_{p,q}(\nabla_p\nabla_q V)^2=\sum_{\mu,\nu=1}^{3}\left[\sum_{\substack{i,j=1\\i\neq j}}^{N}(\nabla_i^\mu\nabla_j^\nu V)^2+\sum_{i=1}^{N}(\nabla_i^\mu\nabla_i^\nu V)^2\right]$$
$$=e^4\int d\mathbf{r}_3\int d\mathbf{r}_4 f(\mathbf{r}_3-\mathbf{r}_4)[N(\mathbf{r}_3)N(\mathbf{r}_4)]_{\text{nc}} \tag{4.42a}$$
$$+e^2\sum_{\mu,\nu=1}^{3}\int d\mathbf{r}_3\int d\mathbf{r}_4\int d\mathbf{r}_5\phi^{\mu\nu}(\mathbf{r}_3-\mathbf{r}_4)\phi^{\mu\nu}(\mathbf{r}_3-\mathbf{r}_5)[N(\mathbf{r}_3)|Q(\mathbf{r}_4)Q(\mathbf{r}_5)]_{\text{nc}}\ . \tag{4.42b}$$

The notation $[N(\mathbf{r}_1)|Q(\mathbf{r}_2)Q(\mathbf{r}_3)]_{\text{nc}}$ means that the contributions of coincident particles at $\mathbf{r}_1=\mathbf{r}_2$ and $\mathbf{r}_1=\mathbf{r}_3$ are suppressed, and $f(\mathbf{r})$ is defined by

$$f(\mathbf{r})=\sum_{\mu,\nu=1}^{3}[\phi^{\mu\nu}(\mathbf{r})]^2\ . \tag{4.43}$$

Inserting (4.42) into (4.40a) gives

$$\langle Q(r_1)Q(r_2)\rangle_a^{(4)}=\frac{1}{5}\frac{\beta^4e^4}{(24m)^2}\int d\mathbf{r}_3 d\mathbf{r}_4 f(\mathbf{r}_3-\mathbf{r}_4)K_1(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4) \tag{4.44a}$$

$$+\frac{1}{5}\frac{\beta^4e^2}{(24m)^2}\sum_{\mu,\nu=1}^{3}\int d\mathbf{r}_3\int d\mathbf{r}_4\int d\mathbf{r}_5\phi^{\mu\nu}(\mathbf{r}_3-\mathbf{r}_4)\phi^{\mu\nu}(\mathbf{r}_3-\mathbf{r}_5)K_2(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4,\mathbf{r}_5)\ , \tag{4.44b}$$

with the four- and five-point correlation functions

$$K_1(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4)=\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)[N(\mathbf{r}_3)N(\mathbf{r}_4)]_{\rm nc}\rangle-\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle\langle [N(\mathbf{r}_3)N(\mathbf{r}_4)]_{\rm nc}\rangle\ , \tag{4.45}$$

$$K_2(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4,\mathbf{r}_5)=\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)[N(\mathbf{r}_3)|Q(\mathbf{r}_4)Q(\mathbf{r}_5)]_{\rm nc}\rangle-\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle\langle [N(\mathbf{r}_3)|Q(\mathbf{r}_4)Q(\mathbf{r}_5)]_{\rm nc}\rangle\ . \tag{4.46}$$

Notice in (4.44a) that $f(\mathbf{r})$ behaves as the square of the dipole potential, i.e., $f(\mathbf{r})\sim 1/|r|^6$ is integrable at infinity but not at the origin. This will cause no problem since the distribution functions vanish sufficiently fast whenever two arguments coincide. In (4.44b), $\phi^{\mu\nu}(\mathbf{r})$ behaves as $|\mathbf{r}|^{-3}$ as $|\mathbf{r}|\to 0$ and the same remark applies (see Appendix D).

To determine the asymptotic behavior of (4.44a) as $|\mathbf{r}_1-\mathbf{r}_2|\to\infty$, it will be convenient to decompose

$$f(\mathbf{r})=f_s(\mathbf{r})+f_l(\mathbf{r}) \tag{4.47}$$

into a short-range part $f_s(\mathbf{r})$ and long-range part $f_l(\mathbf{r})$ with $f_s(\mathbf{r})$ of compact support and $f_l(\mathbf{r})$ regular at the origin. We shall discuss below the long-range part only (the short-range part, which does not contribute to the algebraic decay, is discussed in Appendix C). One needs, moreover, to decompose the expressions (4.45) and (4.46) into fully truncated correlations by means of the formula

$$\langle A_1\cdots A_n\rangle=\sum_{\mathcal{P}}\langle A_{i_1}A_{i_2}\cdots\rangle_T\cdots\langle\cdots A_{i_n}\rangle_T\ . \tag{4.48}$$

The sum runs over all partitions $\mathcal{P}=[m_1,\ldots,m_k]$ of $1,2,\ldots,n$ into k subsets of $m_1,\ldots,m_k$ elements, $k=1,\ldots,n,m_1+\cdots+m_k=n$. When (4.48) is applied to (4.45), and taking into account the neutrality $\langle Q(\mathbf{r})\rangle=0$ and $\langle N(r)\rangle=\rho$, one obtains the decomposition

$$K_1(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4)=\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)[N(\mathbf{r}_3)N(\mathbf{r}_4)]_{\rm nc}\rangle_T \tag{4.48a}$$

$$+\rho\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)N(\mathbf{r}_3)\rangle_T+\rho\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)N(\mathbf{r}_4)\rangle_T \tag{4.48b}$$

$$+\langle Q(\mathbf{r}_1)N(\mathbf{r}_3)\rangle_T\langle Q(\mathbf{r}_2)N(\mathbf{r}_4)\rangle_T+\langle Q(\mathbf{r}_1)N(\mathbf{r}_4)\rangle_T\langle Q(\mathbf{r}_2)N(\mathbf{r}_3)\rangle_T\ , \tag{4.48c}$$

where only the partitions [4], [1,3], and [2,2] occur.

Under the assumption that the fully truncated correlations decay exponentially fast when any group of particles is separated, it is clear that (4.48a) gives a short-range contribution to (4.44a). Using translation invariance, the contribution (4.48b) can be written as

$$\frac{2}{5}\frac{\beta^4e^3\rho}{(24m)^2}\left[\int d\mathbf{r}\,f_l(\mathbf{r})\right]\int d\mathbf{r}_3\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)Q(\mathbf{r}_3)\rangle_T=0\ . \tag{4.49}$$

Note that because of neutrality, one has $e\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)N(\mathbf{r}_3)\rangle_T=\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)Q(\mathbf{r}_3)\rangle_T$ and $e\langle Q(\mathbf{r}_1)N(\mathbf{r}_2)\rangle_T=\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle_s=S(\mathbf{r}_1-\mathbf{r}_2)$. The contribution of (4.48b) vanishes as a consequence of the charge sum rule (3.3). The contribution of (4.48c) to (4.44a) is

$$\frac{2}{5}\frac{\beta^4e^2}{(24m)^2}\int d\mathbf{r}_3\int d\mathbf{r}_4 S(\mathbf{r}_1-\mathbf{r}_3)f_l(\mathbf{r}_3-\mathbf{r}_4)S(\mathbf{r}_4-\mathbf{r}_2) \tag{4.50}$$

and will be responsible for the algebraic tail. It is shown in Appendix D that (4.44b) is rapidly decaying as $|\mathbf{r}_1-\mathbf{r}_2|\to\infty$ [as is the term (4.40b)], hence (4.50) is the leading asymptotic term at the order $\hbar^4$:

$$S^{(4)}(\mathbf{r})=\frac{2}{5}\frac{\beta^4e^2}{(24m)^2}\int d\mathbf{r}_1\int d\mathbf{r}_2 S(\mathbf{r}_1)f_l(\mathbf{r}-\mathbf{r}_1+\mathbf{r}_2)S(\mathbf{r}_2)+\cdots\ , \tag{4.51}$$

where the ellipses represents exponential terms as $|\mathbf{r}|\to\infty$.

It remains to find the exact power law as $|\mathbf{r}|\to\infty$ in (4.51). For $\mathbf{r}_1-\mathbf{r}_2=\mathbf{a}$ fixed, $f_l(\mathbf{r}-\mathbf{a})$ has an asymptotic expansion of the form

$$f_l(\mathbf{r}-\mathbf{a})=\sum_{n\ge 0}(-1)^n\Gamma_{\mu_1\cdots\mu_n}(\hat{\mathbf{a}})\frac{a^{\mu_1}\cdots a^{\mu_n}}{|\mathbf{r}|^{6+n}},\quad |\mathbf{r}|\to\infty \tag{4.52}$$

(with summation on repeated indices) where the tensors $\Gamma_{\mu_1\cdots\mu_n}(\hat{\mathbf{a}})$ depend on the unit vector $\hat{\mathbf{a}}=\mathbf{a}/|\mathbf{a}|$ and can be obtained by squaring the multipole expansion of the dipole potential. Inserting (4.52) in (4.51), one has to calculate the moments

$$\int d\mathbf{r}_1 \int d\mathbf{r}_2 S(\mathbf{r}_1)(r_1-r_2)^{\mu_1}\cdots(r_1-r_2)^{\mu_n}S(\mathbf{r}_2) . \quad (4.53)$$

The spherical symmetry of $S(\mathbf{r})$ together with the electroneutrality $\int d\mathbf{r}\, S(\mathbf{r})=0$ implies that the moments (4.53) vanish for $n=1,2,3$. The fourth moment (4.53) is equal to

$$2(\delta_{\mu_1\mu_2}\delta_{\mu_3\mu_4}+\delta_{\mu_1\mu_3}\delta_{\mu_2\mu_4}+\delta_{\mu_1\mu_4}\delta_{\mu_2\mu_3})\left[\tfrac{1}{3}\int d\mathbf{r}|\mathbf{r}|^2 S(\mathbf{r})\right]^2 . \quad (4.54)$$

One finds therefore with (4.54) and (4.52) that the slowest term in (4.51) is

$$S^{(4)}(\mathbf{r})\sim \frac{4}{5}\frac{\beta^4 e^2}{(24m)^2} A\left[\tfrac{1}{3}\int d\mathbf{r}'|\mathbf{r}'|^2 S(\mathbf{r}')\right]^2 \frac{1}{|\mathbf{r}|^{10}} , \quad |\mathbf{r}|\to\infty \quad (4.55)$$

with

$$A=\sum_{\mu_1,\mu_2}^{3}(\Gamma_{\mu_1\mu_1\mu_2\mu_2}+\Gamma_{\mu_1\mu_2\mu_1\mu_2}+\Gamma_{\mu_1\mu_2\mu_2\mu_1}) . \quad (4.56)$$

One knows from the Stillinger-Lovett second moment condition for a classical OCP that[27]

$$\tfrac{1}{3}\int d\mathbf{r}|\mathbf{r}|^2 S(\mathbf{r})=\frac{1}{2\pi\beta} \quad (4.57)$$

and a calculation found in Appendix E gives the value $A=1260$, hence the final result (4.4) follows.

We add that we have checked that the second-order term $S^{(2)}(\mathbf{r})$ (4.3) and the fourth-order term $S^{(4)}(\mathbf{r})$ [found by collecting all the contributions (4.40a) and (4.40b)] verify the charge sum rule and the second moment conditions

$$\int d\mathbf{r}|\mathbf{r}|^2 S^{(2)}(\mathbf{r})=-\frac{\beta\rho e^2}{2m} , \quad (4.58)$$

$$\int d\mathbf{r}|\mathbf{r}|^2 S^{(4)}(\mathbf{r})=\frac{\pi}{30}\frac{\beta^3\rho^2 e^4}{m^2} , \quad (4.59)$$

inferred by expanding the right-hand side of (3.23) in powers of $\hbar$. In particular, the value (4.59) of the second moment comes from the term (B5) of Appendix B, and it can be verified that all the other parts of $S^{(4)}(\mathbf{r})$ do not contribute to it. This last verification involves the use of generalized classical Stillinger-Lovett conditions given in Ref. 28.

V. MULTICOMPONENT SYSTEMS

We consider a multicomponent system made of M species of particles with charges e_α and masses m_α, $\alpha=1,\ldots,M$. The finite system, contained in a box of volume Λ, has total number of particles $N=\sum_\alpha N_\alpha$ with N_α the number of particles of species α, and it is overall neutral, $\sum_\alpha e_\alpha N_\alpha=0$.

The total Hamiltonian of the system is

$$H=-\sum_i \frac{\hbar^2}{2m_i}\nabla_i^2+\tfrac{1}{2}\sum_{\substack{i,j\\ i\neq j}} e_i e_j \phi(\mathbf{r}_i-\mathbf{r}_j) , \quad (5.1)$$

where $\mathbf{r}_i$, e_i, and m_i are, respectively, the position, the charge, and the mass of the particle labeled by $i=(\alpha,k)$, and the sums run on all species $\alpha=1,\ldots,M$ and on $k=1,\ldots,N_\alpha$ for each species. $\phi(\mathbf{r})$ is the Coulomb potential, or a regularized Coulomb potential if Fermi statistics is not taken into account. The regularized potential reduces to $1/|\mathbf{r}|$ for $|\mathbf{r}|>\sigma$, and to some short-range potential regular at the origin for $|\mathbf{r}|<\sigma$ (the precise form of the latter does not need to be specified). In this section we indicate briefly how the analysis of the preceding sections extends to the system (5.1). A more thorough study will be presented in the next section by means of functional integration. In general, multicomponent systems with different charges and masses have less symmetries than the OCP. In particular, the mass and electric currents are no longer proportional [for instance, the argument (3.11) used to establish the dipole sum rule (3.4) does not apply any more]. Therefore we expect weaker bounds for the correlations than in the OCP.

We first give the generalization of the equilibrium equations (2.11) and (2.21) for charged fermions, assuming the existence of the thermodynamic limit of Green's functions

$$e_\alpha \frac{d}{d\tau}\langle N_\tau(\alpha\mathbf{r})A\rangle_T - i\hbar\nabla\cdot\langle \mathbf{J}(\alpha\mathbf{r})A\rangle_T=0 , \quad (5.2)$$

$$e_\alpha \frac{d^2}{d\tau^2}\langle N(\alpha\mathbf{r})A\rangle_T-\hbar^2\omega_{p\alpha}^2\langle Q_\tau(\mathbf{r})A\rangle_T \quad (5.3a)$$

$$=-\hbar^2\sum_{\mu\nu}^{3}\nabla^\mu\nabla^\nu\langle K_\tau^{\mu\nu}(\alpha\mathbf{r})A\rangle_T \quad (5.3b)$$

$$+\hbar^2\frac{e_\alpha^2}{m_\alpha}\nabla\cdot\int d\mathbf{r}' F(\mathbf{r}-\mathbf{r}')\langle :N_\tau(\alpha\mathbf{r})Q_\tau(\mathbf{r}'):A\rangle_T . \quad (5.3c)$$

In the "continuity equation" (5.2), $N(\alpha\mathbf{r})=a^*(\alpha\mathbf{r})a(\alpha\mathbf{r})$ is the microscopic density of particles of type α and $\mathbf{J}(\alpha\mathbf{r})$ is the corresponding electric current density. In Eq. (5.3), $\omega_{p\alpha}$ is the plasmon frequency of the α particle. The partial "kinetic energy tensor" $K^{\mu\nu}(\alpha\mathbf{r})$ is formed as in (2.15) and $Q(\mathbf{r})=\sum e_\alpha N(\alpha\mathbf{r})$ is the total charge density.

Under the same assumptions (i) and (ii) of Sec. II B we can proceed as in Proposition 1. The main difference is that the function

$$g_\alpha(\mathbf{r},\mathbf{r}')=\langle :N_\tau(\alpha\mathbf{r})Q_\tau(\mathbf{r}'):A\rangle_T$$

is no more symmetric under the exchange of $\mathbf{r}$ and $\mathbf{r}'$, and (2.33) does not apply. Lemma 1 with $n_1=n_2=3$ and $\epsilon<2$ gives that the integral in (5.3c) is $O(|\mathbf{r}|^{-3})$, hence its gradient is $O(|\mathbf{r}|^{-4})$. Then Eq. (5.3) summed on α implies the differential equation

$$\frac{d^2}{d\tau^2}w_3(\tau,A)-\hbar^2\left[\sum_\alpha \omega_{p\alpha}^2\right]w_3(\tau,A)=0 . \tag{5.4}$$

The continuity equation (5.2) gives again $(d/d\tau)w_3(\tau,A)=0$, hence $w_3(\tau,A)=0$ and

$$|\langle Q_\tau(\mathbf{r})A\rangle_T|\le\frac{C_1(\tau,A)}{|\mathbf{r}|^4} . \tag{5.5}$$

Setting $A=Q(\mathbf{0})$ in (5.3), one argues again from the KMS condition, translation invariance, and (5.5) that the term (5.3b) is $O(|\mathbf{r}|^{-6})$. One can apply Lemma 1 with $n_1=4$, $n_2=3$, and $\epsilon<1$ to the term (5.3c), which is $O(|\mathbf{r}|^{-5})$. Thus $w_4(\tau,Q(\mathbf{0}))$ obeys (5.4), and by the KMS boundary conditions, $w_4(\tau,Q(\mathbf{0}))=0$, leading thus to

$$|\langle Q_\tau(\mathbf{r})Q(\mathbf{0})\rangle_T|\le\frac{C_2(\tau,A)}{|\mathbf{r}|^5} . \tag{5.6}$$

If we consider now the particle-particle correlations $\langle N_\tau(\alpha_1\mathbf{r}_2)N(\alpha_2\mathbf{r}_1)\rangle_T$ we can obtain the same result as (5.6) under the additional assumption that

$$e_{\alpha_1}e_{\alpha_2}\langle N_\tau(\alpha_1\mathbf{r}_2)N(\alpha_2\mathbf{r}_1)\rangle_T<0 \quad \text{for } |\mathbf{r}_1-\mathbf{r}_2| \text{ large enough} \tag{5.7}$$

and similar inequalities for higher-order particle correlations. The inequality (5.7) expresses the electrostatic attraction or repulsion of charges at large distances, and should be satisfied in a monotonous regime (high temperature and low density).

By the same arguments which lead to (3.7), one finds immediately that the charge sum rule holds in the multicomponent system. This is no more the case for the dipole sum rule (3.13) as can be seen from the $\hbar^2$-quantum correction (5.9). The second moment relation (3.23) for the charge-charge correlation holds only in the OCP. However, as in the OCP, one can conjecture that the response functions have better screening and cluster properties, in particular that the dipole sum rule (3.16) remains true for multicomponent systems [see comment after Eq. (5.9)]. One expects on the physical grounds that the relation (3.21)

$$\int d\mathbf{r}|\mathbf{r}|^2\int_0^\beta d\tau\langle Q_\tau(\mathbf{r})Q(\mathbf{0})\rangle=-\frac{3}{2\pi} , \tag{5.8}$$

expressing the shielding of an infinitesimal external classical charge, is true in general. In fact, the sum rule (5.8) is also an exact consequence of the equilibrium equations of the multicomponent system when (3.16) holds.[21]

We now indicate the modifications of the Wigner-Kirkwood formalism given in Sec. IV that are needed to treat the multicomponent system. Since Maxwell-Boltzmann statistics is used here, a regularized Coulomb potential is needed to provide stability. In the configuration space of coordinates of all particles, the gradient

$$\frac{1}{\sqrt{m}}\nabla=\left\{\frac{1}{\sqrt{m}}\nabla_i,\ i=1,2,\ldots,N\right\}$$

must be replaced by the differential operator

$$\left\{\frac{1}{\sqrt{m_\alpha}}\nabla_{\alpha,k},\ \alpha=1,\ldots,M;k=1,\ldots,N_\alpha\right\} ,$$

where $\nabla_{\alpha,k}$ is the three-dimensional gradient associated with the kth particle of species α. With this modification one can go through the algebra of Secs. III A and III B, which has a similar structure. The main result at the order $\hbar^2$ is that the multipole of the excess charge density (4.39) is replaced by

$$\int d\mathbf{r}\,\mathcal{Y}_l(\mathbf{r})c_\tau^{(2)}(\mathbf{r}|\alpha_1\mathbf{r}_1,\ldots,\alpha_k\mathbf{r}_k)=\left[\frac{\tau(\beta-\tau)}{2\beta}-\beta\right]\frac{1}{12}\sum_{j=1}^k\frac{e_{\alpha_j}}{m_{\alpha_j}}\nabla_j\mathcal{Y}_l(\mathbf{r}_j)\cdot\nabla_j\rho(\alpha_1\mathbf{r}_1,\ldots,\alpha_k\mathbf{r}_k) , \tag{5.9}$$

where $\rho(\alpha_1\mathbf{r}_1,\ldots,\alpha_k\mathbf{r}_k)$ is the classical distribution function of particles of species $\alpha_1,\ldots,\alpha_k$ at $\mathbf{r}_1,\ldots,\mathbf{r}_k$. The rhs of (5.9) is, in general, different from zero if $l\ge1$, thus all the multipole sum rules, including the dipole, are violated at the order $\hbar^2$ in a multicomponent quantum system. However, one sees on (5.9) that the multipoles of the response function $\int_0^\beta d\tau\langle Q_\tau(\mathbf{r})N(\alpha_1\mathbf{r}_1)\cdots N(\alpha_k\mathbf{r}_k)\rangle$ vanish at the order $\hbar^2$, and the same remarks made after Eq. (4.39) apply here also.

One can again calculate the quantum corrections to the classical truncated particle-particle correlations $\rho_T(\alpha_1\mathbf{r}_1,\alpha_2\mathbf{r}_2)$ at order $\hbar^4$, i.e.,

$$\begin{aligned}\rho_T^{\mathrm{qm}}(\alpha_1\mathbf{r}_1,\alpha_2\mathbf{r}_2)&=\langle N(\alpha_1\mathbf{r}_1)N(\alpha_2\mathbf{r}_2)\rangle_T^{\mathrm{qm}}\\&=\rho_T(\alpha_1\mathbf{r}_1,\alpha_2\mathbf{r}_2)+\hbar^2\rho_T^{(2)}(\alpha_1\mathbf{r}_1,\alpha_2\mathbf{r}_2)+\hbar^4\rho_T^{(4)}(\alpha_1\mathbf{r}_1,\alpha_2\mathbf{r}_2)+\cdots .\end{aligned} \tag{5.10}$$

There are now contributions involving $\nabla^2\phi(\mathbf{r})$ [which were vanishing in the OCP with the strict Coulomb potential by the rule (4.21)], but it is easily checked that all these contributions are short range. As in the OCP, only the analog of the term (4.50),

$$\frac{2}{5}\left[\frac{\beta^2}{24}\right]^2\sum_{\alpha_3\alpha_4}\int d\mathbf{r}_3\int d\mathbf{r}_4\frac{e_{\alpha_3}^2}{m_{\alpha_3}}\langle N(\alpha_1\mathbf{r}_1)N(\alpha_3\mathbf{r}_3)\rangle_T f_l(\mathbf{r}_3-\mathbf{r}_4)\frac{e_{\alpha_4}^2}{m_{\alpha_4}}\langle N(\alpha_4\mathbf{r}_4)N(\alpha_2\mathbf{r}_2)\rangle_T , \tag{5.11}$$

has an algebraic decay, i.e.,

$$\rho_T^{(4)}(\alpha_1\mathbf{r}_1,\alpha_2 0)\sim\frac{\beta^4}{240}\frac{1}{|r_1|^6}\left[\sum_\alpha\frac{e_\alpha^2}{m_\alpha}\int d\mathbf{r}\langle N(\alpha\mathbf{r})N(\alpha_1 0)\rangle_T\right]\left[\sum_\alpha\frac{e_\alpha^2}{m_\alpha}\int d\mathbf{r}\langle N(\alpha\mathbf{r})N(\alpha_2 0)\rangle_T\right],\quad |\mathbf{r}_1|\to\infty\ . \tag{5.12}$$

The quantities in large parentheses, which vanish in a one-component system because of the charge sum rule, are now different from zero. Thus the behavior of the particle-particle correlations at the order $\hbar^4$ is $|\mathbf{r}|^{-6}$, i.e., as the square of the dipole potential itself. However, forming the charge-charge correlations $S^{\rm qm}=\langle Q(\mathbf{r})Q(0)\rangle_T^{\rm qm}$ one notes in (5.12) that the coefficient of the leading term $|\mathbf{r}|^{-6}$ vanishes because of the charge sum rule $\int d\mathbf{r}\langle N(\alpha 0)Q(\mathbf{r})\rangle_T=0$. Then by a calculation similar to that leading to (4.55) one recovers the same $|\mathbf{r}|^{-10}$ behavior as in the OCP,

$$S^{(4)}(\mathbf{r})\sim\frac{4}{5}\left[\frac{\beta^2}{24}\right]^2 A\left[\frac{1}{3}\sum_\alpha\frac{e_\alpha^2}{m_\alpha}\int d\mathbf{r}'|\mathbf{r}'|^2\langle N(\alpha 0)Q(\mathbf{r}')\rangle_T\right]^2\frac{1}{|\mathbf{r}|^{10}},\quad |\mathbf{r}|\to\infty \tag{5.13}$$

with $A=1260$.

VI. FUNCTIONAL INTEGRATION FORMALISM

In this section we extend the analysis of Sec. V to all the terms of the Wigner-Kirkwood expansion of the equilibrium correlations of a multicomponent quantum system. We start from a finite system described by the canonical ensemble. Using the functional integration formalism, we introduce a diagrammatic-expansion scheme which generates the Wigner-Kirkwood expansion in a systematic way. This scheme allows a qualitative analysis of the large-distance behaviors of the correlations of the infinite system, up to any order in $\hbar$. All the $\hbar^{2n}$ terms ($n\geq 2$) are found to decay algebraically with powers depending on the nature of the considered correlations (particle-particle, charge-charge, etc.). In this part of the section, for the sake of simplicity, we only sketch the main arguments. Explicit calculations of the numerical coefficients involved in the asymptotic behaviors are restricted to the $\hbar^4$ terms.

A. Formal representation of the Wigner-Kirkwood expansion

We consider the multicomponent system $\mathcal{S}$ made of M species defined in the preceding section. We assume that the particles obey Maxwell-Boltzmann statistics. The Hamiltonian is given by Eq. (5.1) with a regularized Coulomb potential. It is well known[29,30] that the canonical partition function

$$Z_\Lambda=\frac{1}{\prod_\alpha(N_\alpha)!}\mathrm{Tr}[\exp(-\beta H)] \tag{6.1}$$

can be rewritten as

$$Z_\Lambda=\frac{1}{\prod_\alpha(2\pi\beta\hbar^2/m_\alpha)^{3N_\alpha/2}(N_\alpha)!}\int_{\Lambda^N}\prod_i d\mathbf{q}_i\int\prod_i\mathcal{D}(\xi_i)\exp\left[-\frac{\beta}{2}\sum_{i\neq j}e_ie_j\int_0^1 ds\,\phi(|\mathbf{q}_i-\mathbf{q}_j+\lambda_i\xi_i(s)-\lambda_j\xi_j(s)|)\right]. \tag{6.2}$$

In (6.2), the sums and products over the index $i=(\alpha,k)$ run on $\alpha=1,\dots,M$, $k=1,\dots,N_\alpha$, and $\lambda_i=(\beta\hbar^2/m_i)^{1/2}$ is the thermal de Broglie wavelength of the ith particle. $\mathcal{D}(\xi_i)$ is the Gaussian measure of the Brownian bridge process which defines the functional integration over all the dimensionless paths $\xi_i(s)$ subjected to the constraint $\xi_i(0)=\xi_i(1)=0$. It is normalized to 1 and its covariance is given by

$$\overline{\xi^\mu(s)\xi^\nu(t)}=\int\mathcal{D}(\xi)\xi^\mu(s)\xi^\nu(t)=\delta_{\mu\nu}\times\begin{cases}s(1-t), & s\leq t\\ t(1-s), & t\leq s\ .\end{cases} \tag{6.3}$$

The canonical distribution functions of the system have functional integration representations similar to (6.2). In particular, the n-body distribution function

$$\rho_\Lambda^{\rm qm}(\alpha_1\mathbf{r}_1,\dots,\alpha_n\mathbf{r}_n)=\left\langle\left[\prod_{l=1}^n N(\alpha_l\mathbf{r}_l)\right]_{\rm nc}\right\rangle_\Lambda^{\rm qm},\quad N(\alpha\mathbf{r})=\sum_j\delta_{\gamma\alpha}\delta(\mathbf{q}_j-\mathbf{r}),\quad j=(\gamma,k) \tag{6.4}$$

reads

$$\begin{aligned}\rho_\Lambda^{\rm qm}&(\alpha_1\mathbf{r}_1,\dots,\alpha_n\mathbf{r}_n)\\&=\int_{\Lambda^N}\prod_i d\mathbf{q}_i\left[\prod_{l=1}^n N(\alpha_l\mathbf{r}_l)\right]_{\rm nc}\\&\qquad\times\int\prod_i\mathcal{D}(\xi_i)\exp\left[-\frac{\beta}{2}\sum_{i\neq j}e_ie_j\int_0^1 ds\,\phi(|\mathbf{q}_i-\mathbf{q}_j+\lambda_i\xi_i(s)-\lambda_j\xi_j(s)|)\right]\\&\qquad\qquad\times\left[\int_{\Lambda^N}\prod_i d\mathbf{q}_i\int\prod_i\mathcal{D}(\xi_i)\exp\left[-\frac{\beta}{2}\sum_{i\neq j}e_ie_j\int_0^1 ds\,\phi(|\mathbf{q}_i-\mathbf{q}_j+\lambda_i\xi_i(s)-\lambda_j\xi_j(s)|)\right]\right]^{-1}.\end{aligned} \tag{6.5}$$

In the present description of the finite system, one should, in principle, take into account the boundary terms arising from the interactions of the particles with the walls of the box. We have omitted such terms in the expressions (6.2) and (6.5) because we are ultimately interested in the correlations of the infinite system obtained by taking the thermodynamic limit (N_α, $V\to\infty$, N_α/V fixed) which is assumed to be well defined, i.e., independent of boundary effects (for a charged symmetric Coulomb gas without statistics, the existence of the infinite volume limit is established in Fröhlich and Park[11]).

The representations (6.2) and (6.5) lead us to introduce, in a very natural way, an auxiliary classical system $\mathcal{S}^*$ made of the following M species of filaments. Each species is characterized by the parameters $(e_\alpha,\lambda_\alpha)$. The state of the filament i is characterized by its spatial position $\mathbf{q}_i$, and by an internal degree of freedom $\xi_i(s)$ associated with its shape. The measure $d\mathbf{r}\,\mathcal{D}(\xi)$ defines the summation over all the possible states of a filament. Two filaments i and j of $\mathcal{S}^*$ interact via the two-body potential

$$v_{ij}^*=e_ie_j\int_0^1 ds\,\phi(|\mathbf{q}_i-\mathbf{q}_j+\lambda_i\xi_i(s)-\lambda_j\xi_j(s)|)\ , \tag{6.6}$$

which depends on their positions, their shapes, and the species to which they belong. Note that v_{ij}^* is different from the electrostatic interaction energy between two uniformly charged filaments (for additional comments about this point see Sec. VII). The total interaction potential of $\mathcal{S}^*$ is

$$V^*=\tfrac{1}{2}\sum_{i\neq j} v_{ij}^*\ . \tag{6.7}$$

At equilibrium, the n-body classical distribution functions of $\mathcal{S}^*$, which depend on both the position $\mathbf{r}_l$ and the shape $\boldsymbol{\eta}_l(s)$ of the filaments, are given by

$$\rho_\Lambda^*(\alpha_1\mathbf{r}_1\boldsymbol{\eta}_1,\ldots,\alpha_n\mathbf{r}_n\boldsymbol{\eta}_n)=\left\langle\left[\prod_{l=1}^{n}N^*(\alpha_l\mathbf{r}_l\boldsymbol{\eta}_l)\right]_{\rm nc}\right\rangle_\Lambda^*$$
$$=\frac{\displaystyle\int_{\Lambda^N}\prod_i d\mathbf{q}_i\int\prod_i\mathcal{D}(\xi_i)\left[\prod_{l=1}^{n}N^*(\alpha_l\mathbf{r}_l\boldsymbol{\eta}_l)\right]_{\rm nc}\exp(-\beta V^*)}{\displaystyle\int_{\Lambda^N}\prod_i d\mathbf{q}_i\int\prod_i\mathcal{D}(\xi_i)\exp(-\beta V^*)}\ , \tag{6.8}$$

where $N^*(\alpha\mathbf{r}\boldsymbol{\eta})$ is the microscopic phase-space filament density of species α,

$$N^*(\alpha\mathbf{r}\boldsymbol{\eta})=\sum_j\delta_{\gamma\alpha}\delta(\mathbf{q}_j-\mathbf{r})\delta(\xi_j-\boldsymbol{\eta})\ . \tag{6.9}$$

In (6.9) $\delta(\xi-\boldsymbol{\eta})$ is a formal delta functional such that

$$\int\mathcal{D}(\xi)\delta(\xi-\boldsymbol{\eta})\mathcal{F}\{\xi\}=\mathcal{F}\{\boldsymbol{\eta}\} \tag{6.10}$$

for any functional $\mathcal{F}$ of $\xi(s)$. If one integrates $\rho_\Lambda^*(\alpha_1\mathbf{r}_1\boldsymbol{\eta}_1,\ldots,\alpha_n\mathbf{r}_n\boldsymbol{\eta}_n)$ over all the shapes $\boldsymbol{\eta}_l(s)$ of the filaments, one finds

$$\int\mathcal{D}(\boldsymbol{\eta}_1)\cdots\mathcal{D}(\boldsymbol{\eta}_n)\rho_\Lambda^*(\alpha_1\mathbf{r}_1\boldsymbol{\eta}_1,\ldots,\alpha_n\mathbf{r}_n\boldsymbol{\eta}_n)=\rho_\Lambda^{\rm qm}(\alpha_1\mathbf{r}_1,\ldots,\alpha_n\mathbf{r}_n)\ , \tag{6.11}$$

which is an obvious consequence of the definitions (6.6)–(6.10). Since the n-body correlations $\rho_{\Lambda,T}^*$ and $\rho_{\Lambda,T}^{\rm qm}$ of $\mathcal{S}^*$ and $\mathcal{S}$ are defined through the full truncation of the corresponding n-body distribution functions, one immediately obtains from (6.11)

$$\int\mathcal{D}(\boldsymbol{\eta}_1)\cdots\mathcal{D}(\boldsymbol{\eta}_n)\rho_{\Lambda,T}^*(\alpha_1\mathbf{r}_1\boldsymbol{\eta}_1,\ldots,\alpha_n\mathbf{r}_n\boldsymbol{\eta}_n)=\rho_{\Lambda,T}^{\rm qm}(\alpha_1\mathbf{r}_1,\ldots,\alpha_n\mathbf{r}_n)\ . \tag{6.12}$$

The identity (6.12) shows that the quantum equilibrium correlations of $\mathcal{S}$ can be obtained from the classical positions correlations of $\mathcal{S}^*$. This is particularly useful for our purpose, because the $\hbar$ dependence of the correlations of $\mathcal{S}^*$ only occurs through the de Broglie length λ_i in the arguments of the two-body potential v_{ij}^*. Rewriting the latter as

$$v_{ij}^*=e_ie_j\phi(|\mathbf{q}_i-\mathbf{q}_j|)+e_ie_jw_{ij}^*\ , \tag{6.13}$$

with

$$w_{ij}^*=\sum_{n=1}^{\infty}\frac{1}{n!}\int_0^1 ds[\lambda_i\xi_i(s)\cdot\nabla_i+\lambda_j\xi_j(s)\cdot\nabla_j]^n\phi(\mathbf{q}_i-\mathbf{q}_j) \tag{6.14}$$

we see that the $\hbar$ expansion of $\rho_{\Lambda,T}^{\rm qm}$ can be inferred from the perturbative expansion of $\rho_{\Lambda,T}^*$ with respect to w_{ij}^*. Thus we are left with the standard problem of perturbation of the classical equilibrium state with respect to the two-body potential. This can be treated as follows. Let $\mathcal{E}$ be the ensemble of parameters $(\mathbf{X}\boldsymbol{\omega}\gamma)$ with $\mathbf{X}$ a position, $\boldsymbol{\omega}(s)$ a Brownian bridge, and γ a species index, and let $Q^*(\mathcal{E})$ be the N-body operator

$$Q^*(\mathcal{E})=\sum_i e_i\delta_{\alpha\gamma}\delta(\mathbf{q}_i-\mathbf{X})\delta(\xi_i-\boldsymbol{\omega})\ . \tag{6.15}$$

Replacing v_{ij}^* by (6.13) in (6.7), we rewrite V^* as

$$V^*=V_0^*+W^*\ , \tag{6.16}$$

with

$$V_0^*=\tfrac{1}{2}\sum_{i\neq j}e_ie_j\phi(|\mathbf{q}_i-\mathbf{q}_j|) \tag{6.17}$$

and

$$W^*=\frac{1}{2}\int d\mathcal{E}\,d\mathcal{E}'[Q^*(\mathcal{E})Q^*(\mathcal{E}')]_{\rm nc}w^*(\mathcal{E},\mathcal{E}') \ . \qquad (6.18)$$

In (6.18), $\int d\mathcal{E}$ means $\sum_\gamma \int d\mathbf{X}\,\mathcal{D}(\boldsymbol{\omega})$ while $w^*(\mathcal{E},\mathcal{E}')$ is given by (6.14) with $(\lambda_\gamma\boldsymbol{\omega}\mathbf{X},\lambda_{\gamma'}\boldsymbol{\omega}'\mathbf{X}')$ in place of $(\lambda_i\xi_i\mathbf{q}_i,\lambda_j\xi_j\mathbf{q}_j)$. Calling $\mathcal{S}_0^*$ the unperturbed system with total interaction potential V_0^*, we see that the position correlations of $\mathcal{S}_0^*$ obviously coincide with the classical correlations of $\mathcal{S}$. Furthermore, the perturbative representation of the n-body correlations of $\mathcal{S}$ in terms of the correlations of $\mathcal{S}_0^*$ formally reads

$$\rho^*_{\Lambda,T}(\alpha_1\mathbf{r}_1\boldsymbol{\eta}_1,\ldots,\alpha_n\mathbf{r}_n\boldsymbol{\eta}_n) = \rho^{*,0}_{\Lambda,T}(\alpha_1\mathbf{r}_1\boldsymbol{\eta}_1,\ldots,\alpha_n\mathbf{r}_n\boldsymbol{\eta}_n) + \sum_{p=1}^{\infty}\frac{(-1)^p\beta^p}{2^p p!}\int\prod_{l=1}^{p}d\mathcal{E}_l\,d\mathcal{E}'_l\,w^*(\mathcal{E}_l,\mathcal{E}'_l)\left\langle\left[\prod_{l=1}^{n}N^*(\alpha_l\mathbf{r}_l\boldsymbol{\eta}_l)\right]_{\rm nc}\prod_{l=1}^{p}[Q^*(\mathcal{E}_l)Q^*(\mathcal{E}'_l)]_{\rm nc}\right\rangle^{*,0}_{\Lambda,\rm PT} . \qquad (6.19)$$

In (6.19), $\langle\ \rangle^{*,0}_{\Lambda,\rm PT}$ means a thermal average with the unperturbed Boltzmann factor $\exp(-\beta V_0^*)$; furthermore, this average is partially truncated with respect to all the partitions of the ensemble $\{\mathbf{r}_1,\ldots,\mathbf{r}_n,\mathbf{X}_1,\mathbf{X}'_1,\ldots,\mathbf{X}_p,\mathbf{X}'_p\}$ which do not split an interacting pair $(\mathbf{X}_l,\mathbf{X}'_l)$. The required representation of the quantum correlations $\rho_T^{\rm qm}$ of the infinite system $\mathcal{S}$, directly follows from (6.19), by integrating with respect to all the shapes $\boldsymbol{\eta}_l(s)$ $(l=1,\ldots,n)$ and by taking the thermodynamic limit of the truncated averages $\langle\ \rangle^{*,0}_{\Lambda,\rm PT}$ for fixed configurations (both operations are applied term to term). This gives

$$\rho_T^{\rm qm}(\alpha_1\mathbf{r}_1,\ldots,\alpha_n\mathbf{r}_n) = \rho_T(\alpha_1\mathbf{r}_1,\ldots,\alpha_n\mathbf{r}_n)+\sum_{p=1}^{\infty}\frac{(-1)^p\beta^p}{2^p p!}\int\prod_{l=1}^{p}d\mathcal{E}_l\,d\mathcal{E}'_l\,w^*(\mathcal{E}_l,\mathcal{E}'_l)\left\langle\left[\prod_{l=1}^{n}N(\alpha_l\mathbf{r}_l)\right]_{\rm nc}\prod_{l=1}^{p}[Q^*(\mathcal{E}_l)Q^*(\mathcal{E}'_l)]_{\rm nc}\right\rangle^{*,0}_{\rm PT} , \qquad (6.20)$$

which constitutes a formal representation of the Wigner-Kirkwood (WK) expansion of $\rho_T^{\rm qm}(\alpha_1\mathbf{r}_1,\ldots,\alpha_n\mathbf{r}_n)$ around its classical value $\rho_T(\alpha_1\mathbf{r}_1,\ldots,\alpha_n\mathbf{r}_n)$. This procedure also provides an expansion, similar to (6.20), of the one-body species densities of the quantum system around their classical counterparts. Consequently, the classical reference quantities appearing in (6.20) must be calculated at densities which are different from those of the quantum system. This peculiarity is a well-known feature of the perturbation theory in the canonical ensemble and is linked to the $1/N$ tails in the canonical distribution functions of the finite system. Such tails disappear in a grand-canonical formulation, and it can be easily seen that the representation (6.20) corresponds, in fact, to a perturbative expansion at fixed fugacities (and fixed temperature of course). This point is not relevant for our purpose.

B. The diagrammatic-expansion scheme

In order to study the large-distance behaviors, it is convenient to reformulate the WK expansion generated by the formal representation (6.20), in a diagrammatic language. For this purpose, we first replace w^* by its Taylor's series (6.14) and the partially truncated averages

$$\left\langle\left[\prod_{l=1}^{n}N(\alpha_l\mathbf{r}_l)\right]_{\rm nc}\prod_{l=1}^{p}[Q^*(\mathcal{E}_l)Q^*(\mathcal{E}'_l)]_{\rm nc}\right\rangle^{*,0}_{\rm PT} \qquad (6.21)$$

by their expression in terms of the fully truncated averages

$$\left\langle\left[\prod_{\Omega}N(\alpha_l\mathbf{r}_l)\right]_{\rm nc}\prod_{\Omega'}Q^*(\mathcal{E}_l)\prod_{\Omega''}Q^*(\mathcal{E}'_l)\right\rangle^{*,0}_{T} . \qquad (6.22)$$

In (6.22), Ω is a subset of $[1,\ldots,n]$, Ω' and Ω'' are subsets of $[1,\ldots,p]$, and the truncation is taken with respect to all the partitions of the arguments [in (6.22), the contribution of coincident interacting points $\mathcal{E}_l$ and $\mathcal{E}'_l$ must be excluded]. Each contribution to the WK expansion obtained by the above operations can be associated with a diagram with n root points $\{\mathbf{r}_1,\ldots,\mathbf{r}_n\}$ and $2p$ field points $\{\mathcal{E}_1,\mathcal{E}'_1,\ldots,\mathcal{E}_p,\mathcal{E}'_p\}$. In such a diagram, two field points $\mathcal{E}$ and $\mathcal{E}'$ are connected by an interaction bond $[(n,n')\neq(0,0)]$

$$\frac{1}{n!n'!}\lambda_\gamma^n\lambda_{\gamma'}^{n'}\int_0^1 ds[\boldsymbol{\omega}(s)\cdot\nabla]^n[\boldsymbol{\omega}'(s)\cdot\nabla']^{n'}\phi(\mathbf{X}-\mathbf{X}') \ . \qquad (6.23)$$

We call correlated group the set of arguments occurring in a given fully truncated average (6.22). Each root or field point belongs to one and only one correlated group whose statistical weight has the general form (6.22). Furthermore, when the diagram contains more than one correlated group, its topological structure has the following two properties.

(1) There exists at least one pair of interacting field points which belong to different correlated groups.

(2) For any pair of points, there exists at least one connecting chain made of interaction bonds and of correlated groups (the diagram cannot be separated into two or more disconnected parts).

In Fig. 1 we have drawn a typical diagram which contributes to $\rho_T^{qm}(\alpha_1\mathbf{r}_1,\alpha_2\mathbf{r}_2)$.

At this level, we have obtained a representation of the WK expansion in terms of diagrams. In these diagrams, the field points have a complex structure, because they are characterized as usual by their spatial positions, but also by their Brownian bridges (and their species labels). In order to simplify the present diagrammatic representation, in particular to perform the functional integrations upon the Brownian bridges, we introduce the notions of nude and dressed field points. A field point $\mathcal{E}$ (or $\mathcal{E}'$) is nude if the interaction bond (6.23) does not depend on $(\boldsymbol{\omega}\lambda_\gamma)$ [or $(\boldsymbol{\omega}'\lambda_{\gamma'})$], i.e., if n (or n') is zero; it is dressed otherwise. Using

$$\sum_\gamma \int \mathcal{D}(\boldsymbol{\omega}) Q^*(\mathcal{E}) = Q(\mathbf{X}) , \tag{6.24}$$

we see that the operation $\sum_\gamma \int \mathcal{D}(\boldsymbol{\omega})$ applied to a nude field point $\mathcal{E}$, transforms the operator $Q^*(\mathcal{E})$ into the charge-density operator $Q(\mathbf{X})$, while the other elements and the structure of the considered diagram remain unchanged. The case of the dressed field points can be treated as follows. Since the Boltzmann factor of $\mathcal{S}_0^*$ only depends upon the spatial positions, the truncated averages (6.22) depend upon the Brownian bridges through products of δ functionals arising from the contributions of coincident points in

$$\prod_{\Omega'} Q^*(\mathcal{E}_l) \prod_{\Omega''} Q^*(\mathcal{E}'_l) .$$

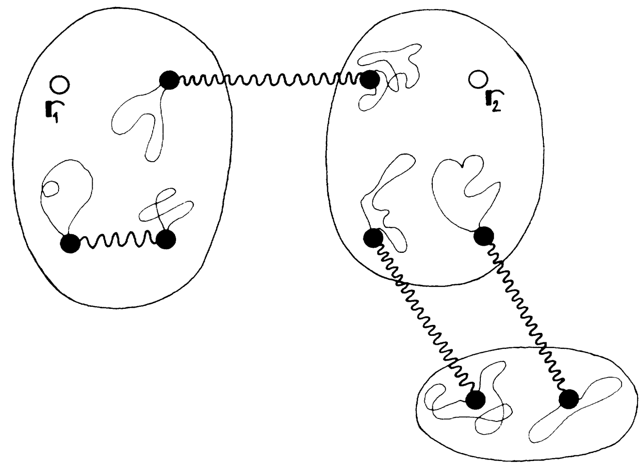

FIG. 1. A typical diagram which contributes to the first perturbative representation of $\rho_T^{qm}(\alpha_1\mathbf{r}_1,\alpha_2\mathbf{r}_2)$. The white circles are root points. The black circles with the random lines are filamentous field points $\mathcal{E}$ and $\mathcal{E}'$. The wavy lines connecting two black circles are interactions bonds (6.23). The bubbles are correlated groups whose statistical weights have the general form (6.22).

Consequently, the operation $\int \mathcal{D}(\boldsymbol{\omega}) \int d\mathcal{E}_1 \cdots \mathcal{E}_q$ applied to any set of $(q+1)$ coincident dressed field points, transforms $Q^*(\mathcal{E})Q^*(\mathcal{E}_1)\cdots Q^*(\mathcal{E}_q)$ into $e_\gamma^{q+1}N(\gamma\mathbf{X})$ while the corresponding product of the $(q+1)$ interaction bonds becomes

$$\int \mathcal{D}(\boldsymbol{\omega}) \frac{1}{n!n'!}\lambda_\gamma^n \lambda_{\gamma'}^{n'} \int_0^1 ds\, [\boldsymbol{\omega}(s)\cdot\boldsymbol{\nabla}]^n [\boldsymbol{\omega}'(s)\cdot\boldsymbol{\nabla}']^{n'} \phi(\mathbf{X}-\mathbf{X}') \frac{1}{n_1!n_1'!}\lambda_\gamma^{n_1}\lambda_{\gamma_1'}^{n_1'} \int_0^1 ds_1 [\boldsymbol{\omega}(s_1)\cdot\boldsymbol{\nabla}]^{n_1} [\boldsymbol{\omega}_1'(s_1)\cdot\boldsymbol{\nabla}_1']^{n_1'} \phi(\mathbf{X}-\mathbf{X}_1')$$

$$\times \cdots \frac{1}{n_q!n_q'!}\lambda_\gamma^{n_q}\lambda_{\gamma_q'}^{n_q'} \int_0^1 ds_q [\boldsymbol{\omega}(s_q)\cdot\boldsymbol{\nabla}]^{n_q} [\boldsymbol{\omega}_q'(s_q)\cdot\boldsymbol{\nabla}_q']^{n_q'} \phi(\mathbf{X}-\mathbf{X}_q') . \tag{6.25}$$

Furthermore, since the Boltzmann factor of $\mathcal{S}_0^*$ is identical to the classical Boltzmann factor of $\mathcal{S}$, the truncated averages (6.22) are replaced by the usual classical position correlations of $\mathcal{S}$.

Once the above operations have been applied to all the nude field points and to all the sets of coincident dressed field points, the diagrammatic expansion of $\rho_T^{qm}(\alpha_1\mathbf{r}_1,\ldots,\alpha_n\mathbf{r}_n)$ has the following structure. Each diagram is made of n root points $(\mathbf{r}_1,\ldots,\mathbf{r}_n)$, p nude field points $(\mathbf{X}_1,\ldots,\mathbf{X}_p)$, and q dressed field-points $(\gamma_1\mathbf{Y}_1,\ldots,\gamma_q\mathbf{Y}_q)$. A nude field point is connected to one and only one dressed field point through one interaction bond, while a dressed field point interacts with an arbitrary number of field points (nude or dressed). Two interacting dressed field points are connected by one or more interaction bonds. An interaction bond reduces symbolically to $\lambda_{\gamma'}^{n'} D_\mathbf{Y}\phi(\mathbf{X}-\mathbf{Y})$ $(n\neq 0)$, or to $\lambda_\gamma^n\lambda_{\gamma'}^{n'} D_\mathbf{Y}^n D_{\mathbf{Y}'}^{n'}\phi(\mathbf{Y}-\mathbf{Y}')$ $(n,n'\neq 0)$ where $D_\mathbf{Y}^n$ is an nth-order differential operator with respect to $\mathbf{Y}$. In fact, the interaction bonds attached to $\mathbf{Y}$ are sums of products of derivatives of the Coulomb potential, whose precise form is determined from (6.25) by applying the standard rules of calculation of the moments of a Gaussian measure [see, for instance, (6.36) and (6.37)]; the sum of the powers $n,n_1,\ldots$ in (6.25) is an even nonzero integer because the odd moments vanish (consequently the WK expansion only contains even powers of $\hbar$). Each root or field point belongs to one and only one correlated group with a statistical weight proportional to the classical correlation

$$\left\langle \left[\prod_\Omega N(\alpha_l\mathbf{r}_l)\right]_{\rm nc} \left[\prod_{\Omega'} N(\gamma_l\mathbf{Y}_l)\right]_{\rm nc} \prod_{\Omega''} Q(\mathbf{X}_l)\right\rangle_T \tag{6.26}$$

[in (6.26), the contributions of coincident interacting field points are excluded as in (6.22)]. If a correlated group contains only one dressed field point, its statistical weight

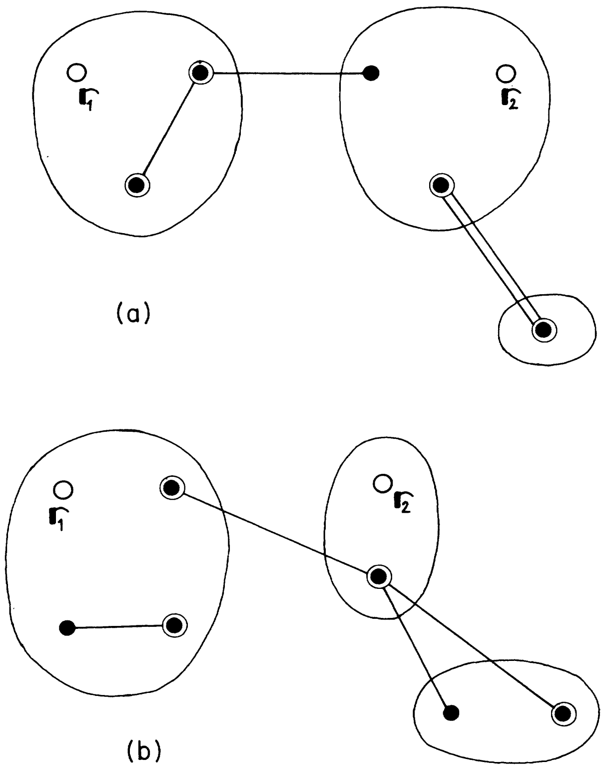

FIG. 2. Two diagrams which arise from the diagram shown in Fig. 1 after integration over the Brownian variables. The small black circles are nude field points $\mathbf{X}$. The black circles with white rings are dressed field points $(\gamma\mathbf{Y})$. Each straight line connecting two black circles with white rings is an interaction bond $\lambda_\gamma^n \lambda_{\gamma'}^{n'} D_{\mathbf{Y}}^n D_{\mathbf{Y}'}^{n'} \phi(\mathbf{Y}-\mathbf{Y}')$. The straight line connecting a black circle with a white ring to a small black circle is an interaction bond $\lambda_\gamma^n D_{\mathbf{Y}}^n \phi(\mathbf{Y}-\mathbf{X})$. The bubbles are correlated groups whose statistical weight have the general form (6.26).

reduces to a mean particle density; if it contains one and only one nude field point, the corresponding diagram does not contribute because the mean charge density $\langle Q(\mathbf{X})\rangle$ vanishes by virtue of overall neutrality. Finally, if a diagram contains more than one correlated group, its topological structure satisfies the two properties (1) and (2) (in other words, the topological structure of the genuine diagrammatic representation is conserved through the reduction operations). In Figs. 2(a) and 2(b), we have drawn two diagrams which arise from the diagram of Fig. 1 by the reduction process.

C. General qualitative analysis of the large-distance behaviors of the correlations

In the above diagrammatic expansion, the contribution of a given diagram of order $\hbar^{2p}$ to $\rho_T^{\mathrm{qm}}(\alpha_1\mathbf{r}_1, \ldots, \alpha_n\mathbf{r}_n)$, is determined up to a numerical multiplicative factor, whose evaluation becomes rapidly inextricable as p increases because of combinatory problems (see Sec. VI D for explicit calculations at the order $\hbar^4$). Therefore we restrict ourselves to a qualitative study of the large-distance behavior of the term of order $\hbar^{2p}$ in the WK expansion of $\rho_T^{\mathrm{qm}}(\alpha_1\mathbf{r}_1, \ldots, \alpha_n\mathbf{r}_n)$. More precisely, we formulate an ensemble of necessary conditions which must be fulfilled by the slow-decaying diagrams defined as those giving algebraic contributions to this asymptotic behavior. This allows us to estimate the minimal powers involved in these contributions. In the analysis, we assume that all the classical correlations of $\mathcal{S}$ decay fast, i.e., faster than any inverse power. For the sake of simplicity, we first consider the case of the two-point correlations $\rho_T^{\mathrm{qm}}(\alpha_1\mathbf{r}_1, \alpha_2\mathbf{r}_2)$. The three-point and higher-order correlations will be briefly examined at the end of this subsection.

If in a given diagram, the two root points $\mathbf{r}_1$ and $\mathbf{r}_2$ belong to the same correlated group, this diagram obviously decays fast when $r_{12}=|\mathbf{r}_1-\mathbf{r}_2|$ goes to infinity. Thus, the slow-decaying diagrams necessarily belong to the class Γ_{12} defined as follows. In a Γ_{12} diagram, $\mathbf{r}_1$ and $\mathbf{r}_2$ belong to two different correlated groups $\mathcal{C}_a$ and $\mathcal{C}_b$ connected by one or more chains made with interaction bonds and correlated groups. In these chains, a connecting path L is an ensemble of field points

$$\{\mathbf{Z}_a, \mathbf{Z}_1, \mathbf{Z}_1', \ldots, \mathbf{Z}_l, \mathbf{Z}_l', \ldots, \mathbf{Z}_b\} \quad (\mathbf{Z}=\mathbf{X} \text{ or } \mathbf{Y})$$

such that

$$\mathbf{Z}_a \in \mathcal{C}_a, \quad \mathbf{Z}_b \in \mathcal{C}_b, \quad \mathbf{Z}_l \in \mathcal{C}_l, \quad \mathbf{Z}_l' \in \mathcal{C}_l$$

and $\mathcal{C}_l$ are intermediary correlated groups which are all different, and

$$(\mathbf{Z}_a, \mathbf{Z}_1), (\mathbf{Z}_1', \mathbf{Z}_2), \ldots, (\mathbf{Z}_{l-1}', \mathbf{Z}_l), (\mathbf{Z}_l', \mathbf{Z}_{l+1})$$

and so on, are pairs of interacting field-points. A field point belonging to a connecting path is called a connecting field point. A connecting field point may belong to various connecting paths. In Fig. 3, we have drawn a typical Γ_{12} diagram with several connecting paths.

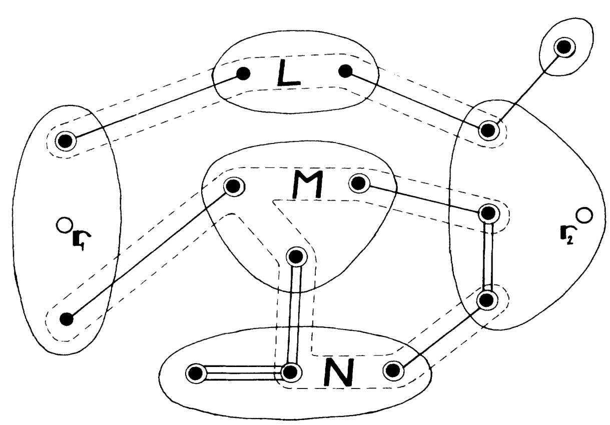

FIG. 3. A typical Γ_{12} diagram with three connecting paths L, M, and N. The strips delimited by the dashed lines are connecting paths. The small black circles and the black circles with white rings inside the strips are connecting field points.

It is important to stress that the slow-decaying diagrams constitute a subclass of Γ_{12}, i.e., some Γ_{12} diagrams still have a fast decay. A number of such cases are given below in the simple situation where there exists a single interaction bond which connects two field points belonging to $\mathcal{C}_a$ and $\mathcal{C}_b$, respectively.

(i) If an interaction bond connects a nude field point $\mathbf{X}$ belonging to $\mathcal{C}_a$ (or $\mathcal{C}_b$) to a dressed field point $(\gamma\mathbf{Y})$ belonging to $\mathcal{C}_b$ (or $\mathcal{C}_a$), the contribution of the considered diagram involves the integral

$$\int d\mathbf{X}\, D_{\mathbf{Y}}^{n}\phi(\mathbf{X}-\mathbf{Y})\langle Q(\mathbf{X})N(\alpha_1\mathbf{r}_1)\cdots\rangle_T = D_{\mathbf{Y}}^{n}\int d\mathbf{X}\,\phi(\mathbf{X}-\mathbf{Y})\langle Q(\mathbf{X})N(\alpha_1\mathbf{r}_1)\cdots\rangle_T \,. \tag{6.27}$$

Since $\langle Q(\mathbf{X})N(\alpha_1\mathbf{r}_1)\cdots\rangle_T$ decays fast when $|\mathbf{X}|\to\infty$, all the multipoles with respect to $\mathbf{X}$ of this truncated charge density vanish.[14] Thus the Coulomb potential

$$\int d\mathbf{X}\,\phi(\mathbf{X}-\mathbf{Y})\langle Q(\mathbf{X})N(\alpha_1\mathbf{r}_1)\cdots\rangle_T \tag{6.28}$$

created at $\mathbf{Y}$ by the truncated charge density $\langle Q(\mathbf{X})N(\alpha_1\mathbf{r}_1)\cdots\rangle_T$, decays fast when $|\mathbf{Y}-\mathbf{r}_1|\to\infty$. So does the integral (6.27). Then, the considered diagram decays fast when $r_{12}\to\infty$, since one has the convolution of (6.27) with the rapidly decreasing function $\langle N(\gamma\mathbf{Y})N(\alpha_2\mathbf{r}_2)\cdots\rangle_T$.

(ii) If an interaction bond connects two dressed field points $(\gamma_1\mathbf{Y}_1)$ and $(\gamma_2\mathbf{Y}_2)$ belonging to $\mathcal{C}_a$ and $\mathcal{C}_b$, respectively, and if $(\gamma_1\mathbf{Y}_1)$ is singly connected (i.e., interacts with one and only one field point), the former bond reads

$$\lambda_{\gamma_1}^{n_1}\lambda_{\gamma_2}^{n_2}D_{\mathbf{Y}_1}^{n_1}D_{\mathbf{Y}_2}^{n_2}\phi(\mathbf{Y}_1-\mathbf{Y}_2)\,, \tag{6.29}$$

with

$$D_{\mathbf{Y}_1}^{n_1}=\frac{1}{n_1!}\int_0^1 ds\int\mathcal{D}(\boldsymbol{\omega}_1)[\boldsymbol{\omega}_1(s)\cdot\nabla_{\mathbf{Y}_1}]^{n_1}\,. \tag{6.30}$$

Since the Gaussian measure $\mathcal{D}(\boldsymbol{\omega})$ is invariant under rotations, $D_{\mathbf{Y}_1}^{n_1}$ is proportional to $(\nabla_{\mathbf{Y}_1}^2)^{n_1/2}$ (n_1 is even). Using also

$$\nabla^2\phi(r)=0,\quad r>\sigma \tag{6.31}$$

we find that the interaction bond (6.29) is short-ranged. Therefore the corresponding diagram decays fast when $r_{12}\to\infty$ because $(\mathbf{Y}_1,\mathbf{r}_1)$ as well as $(\mathbf{Y}_2,\mathbf{r}_2)$ belong to the same correlated groups.

From the above examples, we infer that, in a slow-decaying diagram, each of two-interacting field points which belong to $\mathcal{C}_a$ and $\mathcal{C}_b$ are necessarily doubly connected (a field point $\mathbf{Y}$ is doubly connected if there exist at least two interaction bonds which connect $\mathbf{Y}$ to one or more field points). When $\mathcal{C}_a$ and $\mathcal{C}_b$ are connected by chains involving intermediary correlated groups, we find by a straightforward inspection that a slow decay of the considered diagram implies that, in each connecting path, there exist at least two connecting field points $\mathbf{Y}'$ and $\mathbf{Y}''$ which fulfill the following conditions. First they belong to different correlated groups and both are doubly connected. Furthermore, they are connected either directly through one or more interaction bonds or indirectly through the convolution

$$\int d\mathbf{X}'d\mathbf{X}''D_{\mathbf{Y}'}^{n'}\phi(\mathbf{Y}'-\mathbf{X}')\langle Q(\mathbf{X}')Q(\mathbf{X}'')\rangle_T \times D_{\mathbf{Y}''}^{n''}\phi(\mathbf{Y}''-\mathbf{X}'')\,, \tag{6.32}$$

which is shown (see Appendix F) to behave, when $|\mathbf{Y}'-\mathbf{Y}''|\to\infty$, as

$$\beta^{-1}D_{\mathbf{Y}'}^{n'}D_{\mathbf{Y}''}^{n''}\phi(\mathbf{Y}'-\mathbf{Y}'') \tag{6.33}$$

apart from exponentially decaying terms. Such connecting field points $\mathbf{Y}'$ and $\mathbf{Y}''$ are called algebraic field points, while the pair $(\mathbf{Y}',\mathbf{Y}'')$ is called an algebraic pair. Let us emphasize that the previous conditions are necessary, but not sufficient, for a slow decay of the considered diagram. In other words, a diagram which fulfills these conditions may still have a fast decay as illustrated below.

Now, we want to determine the minimal powers which appear in the algebraic contributions of the slow-decaying diagrams. We first consider the simplest diagram which, *a priori*, gives an algebraic contribution. In this diagram, $\mathcal{C}_a$ and $\mathcal{C}_b$ are connected by one and only one interaction bond. The contribution of the former is proportional to

$$\int d\mathbf{Y}_1 d\mathbf{Y}_2 D_{\mathbf{Y}_1}^{n_1}D_{\mathbf{Y}_2}^{n_2}\phi(\mathbf{Y}_1-\mathbf{Y}_2)\int d\{\mathcal{P}_1\}d\{\mathcal{P}_2\}\prod_1(\mathcal{B})\prod_2(\mathcal{B})\langle N(\gamma_1\mathbf{Y}_1)N(\alpha_1\mathbf{r}_1)\cdots\rangle_T\langle N(\gamma_2\mathbf{Y}_2)N(\alpha_2\mathbf{r}_2)\cdots\rangle_T\prod\langle\;\rangle_T\,. \tag{6.34}$$

In (6.34), $\mathbf{Y}_1$ and $\mathbf{Y}_2$ are doubly connected, $\{\mathcal{P}_1\}$ and $\{\mathcal{P}_2\}$ are two disconnected ensembles of field points, $\prod_1(\mathcal{B})$ and $\prod_2(\mathcal{B})$ are the products of the interaction bonds connecting the field points of $\{\mathbf{Y}_1,\mathcal{P}_1\}$ and $\{\mathbf{Y}_2,\mathcal{P}_2\}$, respectively, and $\prod\langle\;\rangle_T$ is the product of statistical weights associated with correlated groups made with parts of $\{\mathcal{P}_1\}$ or parts of $\{\mathcal{P}_2\}$. The precise forms of the interaction bonds which appear in (6.34) are determined according to the general rules described in Sec. VI B. Taking into account the rotation invariance of the Gaussian measure $\mathcal{D}(\boldsymbol{\omega})$ and of the reference classical system, we find that the contribution (6.34) can be rewritten as a sum of terms of the form

$$\int d\mathbf{Y}_1 d\mathbf{Y}_2 G_1(|\mathbf{Y}_1-\mathbf{r}_1|)G_2(|\mathbf{Y}_2-\mathbf{r}_2|)P_{n_1}((\mathbf{Y}_1-\mathbf{r}_1)\cdot\nabla_{\mathbf{Y}_1},\nabla_{\mathbf{Y}_1}^2)P_{n_2}((\mathbf{Y}_2-\mathbf{r}_2)\cdot\nabla_{\mathbf{Y}_2},\nabla_{\mathbf{Y}_2}^2)\phi(\mathbf{Y}_1-\mathbf{Y}_2)\,, \tag{6.35}$$

where $G_1(r)$ and $G_2(r)$ are fast-decaying functions of r, while $P_{n_1}(a,b)$ and $P_{n_2}(a,b)$ are polynomials in the two variables a and b. The asymptotic behavior of (6.35) when $r_{12}\to\infty$, is given, apart from exponentially decaying terms, by the expansion of

$$P_{n_1}((\mathbf{Y}_1-\mathbf{r}_1)\cdot\nabla_{\mathbf{Y}_1},\nabla^2_{\mathbf{Y}_1})P_{n_2}((\mathbf{Y}_2-\mathbf{r}_2)\cdot\nabla_{\mathbf{Y}_2}\nabla^2_{\mathbf{Y}_2})\phi(\mathbf{Y}_1-\mathbf{Y}_2)$$

around

$$P_{n_1}((\mathbf{Y}_1-\mathbf{r}_1)\cdot\nabla_{\mathbf{r}_1},\nabla^2_{\mathbf{r}_1})P_{n_2}((\mathbf{Y}_2-\mathbf{r}_2)\cdot\nabla_{\mathbf{r}_2},\nabla^2_{\mathbf{r}_2})\phi(\mathbf{r}_1-\mathbf{r}_2)$$

with respect to $(\mathbf{Y}_1-\mathbf{r}_1)$ and $(\mathbf{Y}_2-\mathbf{r}_2)$. Since G_1 and G_2 are spherically symmetric functions, the resulting multipolar expansion of (6.35) only involves terms proportional to

$$(\nabla^2_{\mathbf{r}_1})^{l_1}(\nabla^2_{\mathbf{r}_2})^{l_2}\phi(\mathbf{r}_1-\mathbf{r}_2)$$

with l_1 and l_2 strictly positive integers (the differential operators appearing in P_{n_1} and P_{n_2} are of order $n_1>0$ and $n_2>0$, respectively). Because of (6.31), all these terms vanish, and we finally conclude that the considered diagram decays fast when $r_{12}\to\infty$.

We turn now to the general case of a diagram which fulfills the set of necessary algebraic conditions described above. In such a diagram, $\mathcal{C}_a$ and $\mathcal{C}_b$ are connected by one or more chains (a chain may reduce to one or more interaction bonds without any intermediary correlated groups). In each connecting path L, there exist $p(L)$ algebraic pairs

$$\{(\mathbf{Y}'_{L,1},\mathbf{Y}''_{L,1}),\ldots,(Y'_{L,p(L)},\mathbf{Y}''_{L,p(L)})\}$$

with $p(L)\geq 1$. Note that two successive pairs in a given connecting path may have one common field point (i.e., one may have $Y''_{L,l}=\mathbf{Y}'_{L,l+1}$), while an algebraic point may belong to various connecting paths [for fixing ideas, see the diagram shown in Fig. 4(a), which is of the general type considered here]. In order to estimate the large-r_{12} behavior of the present diagram, we first perform the integrations over the nonalgebraic field points. The eventual algebraic terms in this behavior then arise from a convolution of fast-decaying functions ψ_{fast} whose arguments are subsets of $\{\mathbf{r}_1,\mathbf{r}_2;\mathbf{Y}'_{L,l},\mathbf{Y}''_{L,l};\ L=\text{connecting paths}\}$, and of two-point slow-decaying functions φ_{slow} whose arguments are algebraic pairs. The fast decay of ψ_{fast} is a consequence of (6.33), of the fast decay of the classical correlations, and of the harmonicity of the Coulomb potential [for similar reasons as those explained above in the detailed study of the particular diagrams discussed in (i) and (ii)]. $\varphi_{\text{slow}}(\mathbf{Y}'_{L,l},\mathbf{Y}''_{L,l})$ is nothing but the product of one or more interaction bonds connecting $\mathbf{Y}'_{L,l}$ and $\mathbf{Y}''_{L,l}$ [as a consequence of (6.33)]. The actual convolution can be represented by a diagram, where all the field points are algebraic field points while the weights associated with the correlation clouds are functions ψ_{fast} [which may reduce to classical correlations (6.26)]. In Fig. 4(b), we have drawn the corresponding diagram which arises from the diagram shown in Fig. 4(a) after integration over the nonalgebraic field points. The

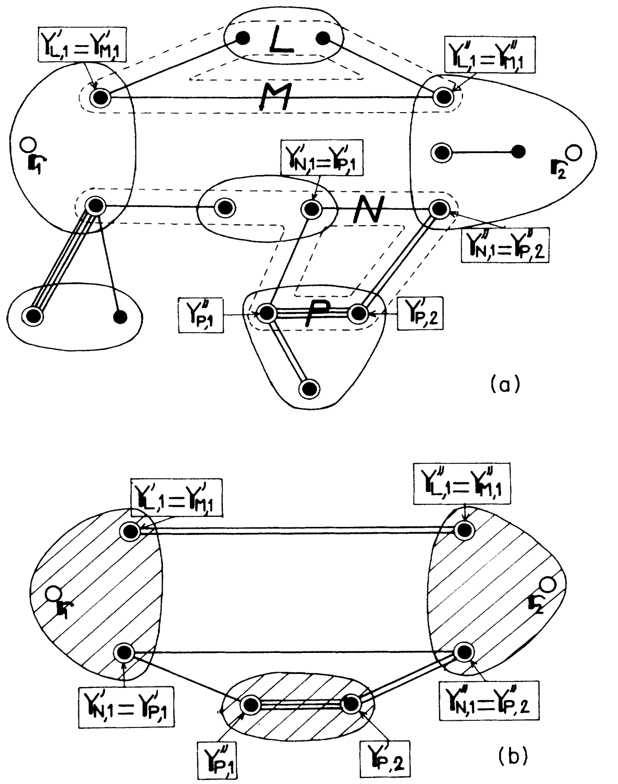

FIG. 4. (a) A typical diagram with four connecting paths L, M, N, and P, which fulfills the set of necessary algebraic conditions. The algebraic field points $\mathbf{Y}'$ and $\mathbf{Y}''$ are explicitly indicated on the figure. (b) The diagram which arises from the diagram shown in (a) after integration over the nonalgebraic field points. The hatched bubbles represent functions ψ_{fast}.

configurations of the algebraic field points which may give algebraic contributions to the large-r_{12} behavior of the previous convolution, are such that there exist q_i pairs $(\mathbf{Y}'_{l,i},\mathbf{Y}''_{l,i})$ $(i=1,2;l=1,\ldots,q_i)$ with $|\mathbf{Y}'_{l,i}-\mathbf{r}_i|$ finite (of the order of the mean-interparticle distance) and $|\mathbf{Y}'_{l,i}-Y''_{l,i}|$ large (of the order of r_{12}). If for $i=1$ or $i=2$, one has $q_i=1$ and $\varphi_{\text{slow}}(\mathbf{Y}'_{1,i},\mathbf{Y}''_{1,i})$ reduces to a single interaction bond, the considered configuration does not give, in fact, any algebraic contribution, for similar reasons as those exposed in detail for the simplest *a priori* slow-decaying diagram. Thus the algebraic contributions are given by configurations where, for $i=1$ and 2, one has $q_i=2$, or $q_i=1$ and $\varphi_{\text{slow}}(\mathbf{Y}'_{1,i},\mathbf{Y}''_{1,i})$ is the product of at least two-interaction bonds. Since all the interaction bonds involved in the functions φ_{slow} decay at least like $1/r^3$ at large distances r, we finally conclude that the leading term of the large-r_{12} expansion of the slow-decaying diagrams, is at least of order $1/r_{12}^6$. Some of these diagrams decay exactly as $1/r_{12}^6$ when $r_{12}\to\infty$ (see below and Sec. VI D), while others decay faster. For instance, the diagram shown in Fig. 4(a) decays at least as $(r_{12})^{-12}$, as can be easily seen from Fig. 4(b). Because of rotation invariance, the pure $1/r^3 * 1/r^3$ convolutions take the form

$$\int d\mathbf{Y}_3[\nabla \mathbf{Y}_3 D_{\mathbf{Y}_1}\phi(\mathbf{Y}_1-\mathbf{Y}_3)]\cdot[\nabla_{\mathbf{Y}_3} D_{\mathbf{Y}_2}\phi(\mathbf{Y}_3-\mathbf{Y}_2)] \ .$$

These convolutions do behave as $1/|\mathbf{Y}_1-\mathbf{Y}_2|^3$ for $|\mathbf{Y}_1-\mathbf{Y}_2|$ large, and do not involve

$$(\ln|\mathbf{Y}_1-\mathbf{Y}_2|)/|\mathbf{Y}_1-\mathbf{Y}_2|^3$$

terms as can be checked through integrations by parts and by using (6.31).

The above analysis shows that all the terms of the WK expansion of $\rho_T^{\mathrm{qm}}(\alpha_1\mathbf{r}_1,\alpha_2\mathbf{r}_2)$ decay at least like $1/r_{12}^6$ when $r_{12}\to\infty$. The $\hbar^2$ term decays faster and the $\hbar^4$ term behaves exactly as $1/r_{12}^6$, as can be checked through explicit calculations (see Sec. VI D). At any order $\hbar^{2n}$, $n\geq 3$, one has the slow-decaying diagram shown in Fig. 5, which does behave as $1/r_{12}^6$ when $r_{12}\to\infty$, as can be checked through a multipolar expansion of the two interaction bonds which connect $\mathcal{C}_a$ and $\mathcal{C}_b$. At this order, there are other slow-decaying diagrams which have a similar large-r_{12} behavior. As illustrated by the explicit calculations at the order $\hbar^4$, the $1/r_{12}^6$ contributions of all these diagrams should not cancel out in general, and consequently should be the leading terms at the order $\hbar^{2n}$, $n\geq 2$. This suggests that $\rho_T^{\mathrm{qm}}(\alpha_1\mathbf{r}_1,\alpha_2\mathbf{r}_2)$ should decay like $1/r_{12}^6$ for sufficiently high temperatures and low densities (under these conditions, the quantum effects are small and it is reasonable to treat them perturbatively). Of course, a rigorous proof of the latter statement would require a detailed control of all the terms of the WK expansion (see Sec. VII for an exact resummation of this expansion in a particular limit).

The above analysis can be also applied to the particle-charge correlations $\langle N(\alpha_1\mathbf{r}_1)Q(\mathbf{r}_2)\rangle^{\mathrm{qm}}$ and to the charge-charge correlation $S^{\mathrm{qm}}(r_{12})=\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle^{\mathrm{qm}}$. The large-distance behavior of the corresponding slow-decaying diagrams is determined through multipolar expansions of the interaction bonds connecting the algebraic field points. Using the translation invariance of the reference classical system, as well as the classical charge and dipole sum rules, we find that $\langle N(\alpha_1\mathbf{r}_1)Q(\mathbf{r}_2)\rangle^{\mathrm{qm}}$ should decay like $1/r_{12}^8$ when $r_{12}\to\infty$, while $S^{\mathrm{qm}}(r_{12})$ should decay like $1/r_{12}^{10}$ (under the same assumptions as

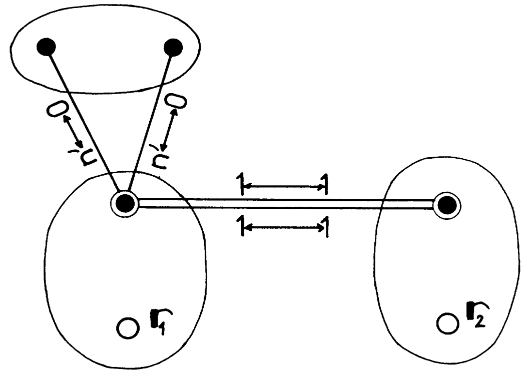

FIG. 5. A diagram of order $\hbar^{2n}$, $n\geq 3$, which gives $1/r^6$ tails to the large-distance behavior of $\rho_T^{\mathrm{qm}}(\alpha_1\mathbf{r}_1,\alpha_2\mathbf{r}_2)$. The notation $p\leftrightarrow q$ (p and q integers) over (or below) a straight line connecting two black circles means that the corresponding interaction bond involves products of differential operators of orders p and q applied to the Coulomb potential. Here, one has $n'=n-2$.

TABLE I. Large-distance behaviors of the quantum correlations predicted by the Wigner-Kirkwood expansion.

Correlations	Asymptotic configurations	Asymptotic behaviors
$\langle N(\alpha_1\mathbf{r}_1)N(\alpha_2\mathbf{r}_2)\rangle_T^{\mathrm{qm}}$	$r_{12}=r\to\infty$	$1/r^6$
$\langle N(\alpha_1\mathbf{r}_1)Q(\mathbf{r}_2)\rangle_T^{\mathrm{qm}}$	$r_{12}=r\to\infty$	$1/r^8$
$\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle_T^{\mathrm{qm}}$	$r_{12}=r\to\infty$	$1/r^{10}$
$\langle N(\alpha_1\mathbf{r}_1)N(\alpha_2\mathbf{r}_2)N(\alpha_3\mathbf{r}_3)\rangle_T^{\mathrm{qm}}$	$\mathbf{r}_{12}$ fixed $r_{13}=r\to\infty$	$1/r^6$
	$r_{12}=r$, $r_{13}=ar$, $r_{23}=br$ $r\to\infty$; a and b fixed	$1/r^9$
$\langle N(\alpha_1\mathbf{r}_1)N(\alpha_2\mathbf{r}_2)N(\alpha_3\mathbf{r}_3)N(\alpha_4\mathbf{r}_r)\rangle_T^{\mathrm{qm}}$	$\mathbf{r}_{12}$ and $\mathbf{r}_{13}$ fixed $r_{14}=r\to\infty$	$1/r^6$
	$\mathbf{r}_{12}$ and $\mathbf{r}_{34}$ fixed $r_{13}=r\to\infty$	$1/r^3$
	$\mathbf{r}_{12}$, a and b fixed $r_{13}=r$, $r_{14}=ar$, $r_{34}=br$, $r\to\infty$	$1/r^9$
	$r_{12}=r$, $r_{13}=ar$, $r_{14}=br$ $r_{23}=cr$, $r_{34}=dr$ $r\to\infty$; a, b, c, and d fixed	$1/r^{12}$

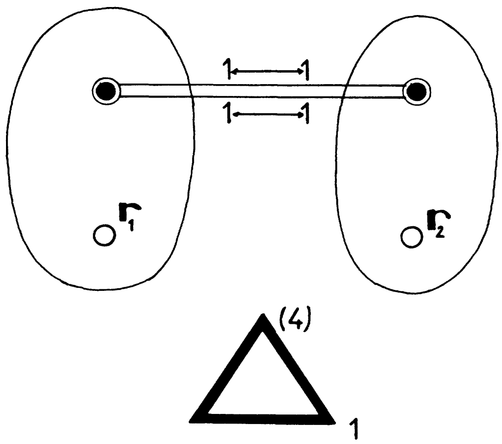

FIG. 6. The diagram $\Delta_1^{(4)}$ of order $\hbar^4$ which gives an algebraic contribution to the large-distance behavior of $\rho_T^{\rm qm}(\alpha_1\mathbf{r}_1,\alpha_2\mathbf{r}_2)$.

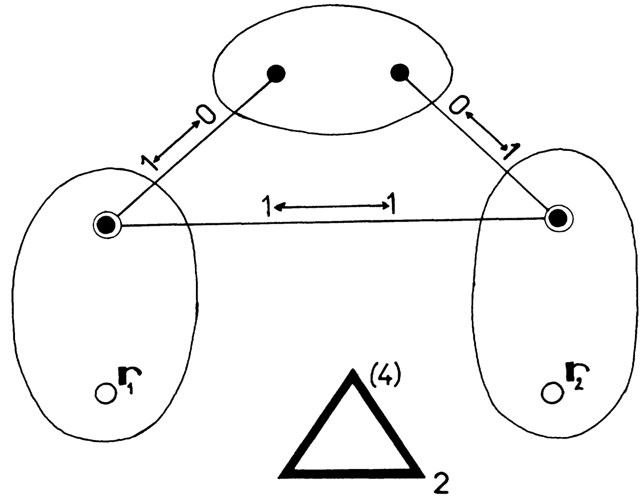

FIG. 7. Same as Fig. 6 but for $\Delta_2^{(4)}$.

above relative to the convergence of the WK expansion). A similar analysis shows that the three-point correlations $\rho_T^{\rm qm}(\alpha_1\mathbf{r}_1,\alpha_2\mathbf{r}_2,\alpha_3\mathbf{r}_3)$ should decay at least as $1/r^6$ for any large separation of its arguments, while the four-point correlations $\rho_T^{\rm qm}(\alpha_1\mathbf{r}_1,\alpha_2\mathbf{r}_3,\alpha_3\mathbf{r}_3,\alpha_4\mathbf{r}_4)$ have a slower decay for the following asymptotic configurations. When $|\mathbf{r}_1-\mathbf{r}_3|\to\infty$, $|\mathbf{r}_1-\mathbf{r}_2|$ and $|\mathbf{r}_3-\mathbf{r}_4|$ being kept fixed, $\rho_T^{\rm qm}(\alpha_1\mathbf{r}_1,\alpha_2\mathbf{r}_2,\alpha_3\mathbf{r}_3,\alpha_4\mathbf{r}_4)$ indeed decays like $1/r_{13}^3$. This $1/r_{13}^3$ term arises in part from the contributions of the simplest slow-decaying diagrams, where $(\mathbf{r}_1,\mathbf{r}_2)$ and $(\mathbf{r}_3,\mathbf{r}_4)$, respectively, belong to correlated groups $\mathcal{C}_a$ and $\mathcal{C}_b$ which are connected by one and only one interaction bond of the form $D_{\mathbf{Y}_1}D_{\mathbf{Y}_2}\phi(\mathbf{Y}_1-\mathbf{Y}_2)$: in the multipolar expansion of this bond with respect to $(\mathbf{Y}_1-\mathbf{r}_1)$ and $(\mathbf{Y}_2-\mathbf{r}_2)$, the zeroth-order term does not vanish in general because the statistical weights associated with $\mathcal{C}_a$ and $\mathcal{C}_b$ are no longer invariant under rotations for nonzero fixed vectors $\mathbf{r}_{12}$ and $\mathbf{r}_{34}$. Physically, the $1/r^3$ tails can be related to the dipole-dipole interaction between polarization clouds surrounding groups of two or more charges. These $1/r^3$ tails appear in any correlations (particle or charge) between two groups of two or more points separated by a large distance r. We have summarized the asymptotic behavior of the various correlations, suggested by the WK expansion, in Table I. Note that the faster decay of the two-point charge correlations, as well as the slower decay of the four-point and higher-order correlations, are compatible with the nonperturbative analysis of the hierarchy equations (see Sec. V).

D. Explicit calculations at the orders $\hbar^2$ and $\hbar^4$

According to the general rules derived in Sec. VI C, we exhibit all the slow-decaying diagrams at the orders $\hbar^2$ and $\hbar^4$, and we explicitly compute their algebraic contributions to the large-distance behavior of the correlations. This allows us to recover the results found in Sec. V by a different method. We consider only the two-point correlations.

In any slow-decaying diagram, there are at least two algebraic field points. Since each of these field points is at least doubly connected, any such diagram is at least of order $\hbar^4$. Therefore there are no slow-decaying diagrams of order $\hbar^2$, and the corresponding quantum corrections to the classical correlations decay fast, as already noticed in Sec. V. At the order $\hbar^4$, one has three slow-decaying diagrams $\Delta_1^{(4)}$, $\Delta_2^{(4)}$, and $\Delta_3^{(4)}$ which are shown in Figs. 6, 7, and 8, respectively. For calculating the contribution of each of these diagrams, one has to determine the following.

- The order p of the term in the representation (6.20) from which the diagram arises.
- The symmetry factor associated with all the ways of labeling the field points and of generating the dressed field points $\mathbf{Y}$ through the collapse of one or more field points $\mathcal{E}$.
- The precise form of the interaction bonds coming from Taylor's series (6.14) and the functional integrations over the Brownian variables.

For $\Delta_1^{(4)}$, $p=2$, the symmetry factor is 4, and the product of the two-bonds connecting $\mathbf{Y}_1$ and $\mathbf{Y}_2$ is

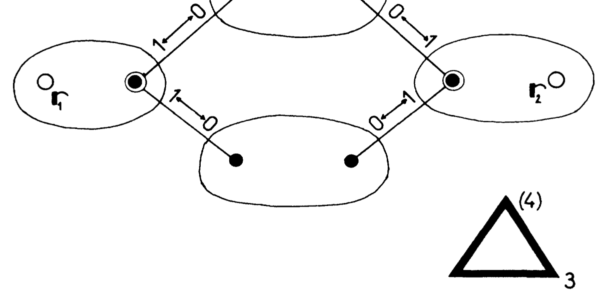

FIG. 8. Same as Fig. 6 but for $\Delta_3^{(4)}$.

$$\lambda_{\gamma_1}^2\lambda_{\gamma_2}^2\int_0^1 ds\int_0^1 dt\int\mathcal{D}(\omega_1)\mathcal{D}(\omega_2)\frac{1}{1!1!}[\omega_1(s)\cdot\nabla_{Y_1}][\omega_2(s)\cdot\nabla_{Y_2}]\phi(\mathbf{Y}_1-\mathbf{Y}_2)\frac{1}{1!1!}[\omega_1(t)\cdot\nabla_{Y_1}][\omega_2(t)\cdot\nabla_{Y_2}]\phi(\mathbf{Y}_1-\mathbf{Y}_2)\ , \tag{6.36}$$

which reduces to

$$\frac{1}{90}\lambda_{\gamma_1}^2\lambda_{\gamma_2}^2\sum_{\mu,\nu}[\nabla^\mu\nabla^\nu\phi(\mathbf{Y}_1-\mathbf{Y}_2)]^2 \tag{6.37}$$

by use of the covariance formula (6.3). The contribution of $\Delta_1^{(4)}$ then becomes

$$\frac{(-1)^2\beta^2}{2^2\times 2!}4\sum_{\gamma_1,\gamma_2}\int d\mathbf{Y}_1 d\mathbf{Y}_2 e_{\gamma_1}^2\langle N(\alpha_1\mathbf{r}_1)N(\gamma_1\mathbf{Y}_1)\rangle_T e_{\gamma_2}^2\langle N(\alpha_2\mathbf{r}_2)N(\gamma_2\mathbf{Y}_2)\rangle_T\tfrac{1}{90}\lambda_{\gamma_1}^2\lambda_{\gamma_2}^2\sum_{\mu,\nu}[\nabla^\mu\nabla^\nu\phi(\mathbf{Y}_1-\mathbf{Y}_2)]^2$$

$$=\frac{\beta^4\hbar^4}{180}\sum_{\gamma_1,\gamma_2}\frac{e_{\gamma_1}^2e_{\gamma_2}^2}{m_{\gamma_1}m_{\gamma_2}}\int d\mathbf{Y}_1 d\mathbf{Y}_2\langle N(\alpha_1\mathbf{r}_1)N(\gamma_1\mathbf{Y}_1)\rangle_T\langle N(\alpha_2\mathbf{r}_2)N(\gamma_2\mathbf{Y}_2)\rangle_T f(|\mathbf{Y}_1-\mathbf{Y}_2|)\ , \tag{6.38}$$

where we recall that $f(r)=\sum_{\mu,\nu}[\nabla^\mu\nabla^\nu\phi(\mathbf{r})]^2$. Similarly, we find that the contributions of $\Delta_2^{(4)}$ and $\Delta_3^{(4)}$ are, respectively,

$$\frac{(-1)^3\beta^3}{2^3\times 3!}48\sum_{\gamma_1,\gamma_2}\int d\mathbf{Y}_1 d\mathbf{Y}_2 d\mathbf{X}_1 d\mathbf{X}_2 e_{\gamma_1}^2\langle N(\alpha_1\mathbf{r}_1)N(\gamma_1\mathbf{Y}_1)\rangle_T e_{\gamma_2}^2\langle N(\alpha_2\mathbf{r}_2)N(\gamma_2\mathbf{Y}_2)\rangle_T\langle Q(\mathbf{X}_1)Q(\mathbf{X}_2)\rangle_T$$

$$\times\lambda_{\gamma_1}^2\lambda_{\gamma_2}^2\int_0^1 ds\int_0^1 dt\int_0^1 du\int\mathcal{D}(\omega_1)\mathcal{D}(\omega_2)\frac{1}{1!1!}[\omega_1(s)\cdot\nabla_{Y_1}][\omega_2(s)\cdot\nabla_{Y_2}]\phi(\mathbf{Y}_1-\mathbf{Y}_2)\frac{1}{1!}[\omega_1(t)\cdot\nabla_{Y_1}]$$

$$\times\phi(\mathbf{Y}_1-\mathbf{X}_1)\frac{1}{1!}[\omega_2(u)\cdot\nabla_{Y_2}]\phi(\mathbf{Y}_2-\mathbf{X}_2)$$

$$=\frac{-\beta^5\hbar^4}{120}\sum_{\gamma_1,\gamma_2}\frac{e_{\gamma_1}^2e_{\gamma_2}^2}{m_{\gamma_1}m_{\gamma_2}}\int d\mathbf{Y}_1 d\mathbf{Y}_2 d\mathbf{X}_1 d\mathbf{X}_2\langle N(\alpha_1\mathbf{r}_1)N(\gamma_1\mathbf{Y}_1)\rangle_T\langle N(\alpha_2\mathbf{r}_2)N(\gamma_2\mathbf{Y}_2)\rangle_T\langle Q(\mathbf{X}_1)Q(\mathbf{X}_2)\rangle_T$$

$$\times\sum_{\mu,\nu}[\nabla^\mu\nabla^\nu\phi(\mathbf{Y}_1-\mathbf{Y}_2)][\nabla^\mu\phi(\mathbf{Y}_1-\mathbf{X}_1)][\nabla^\nu\phi(\mathbf{Y}_2-\mathbf{X}_2)] \tag{6.39}$$

and

$$\frac{(-1)^4\beta^4}{2^4\times 4!}192\sum_{\gamma_1,\gamma_2}\int d\mathbf{Y}_1 d\mathbf{Y}_2 d\mathbf{X}_1 d\mathbf{X}_2 d\mathbf{X}_3 d\mathbf{X}_4 e_{\gamma_1}^2\langle N(\alpha_1\mathbf{r}_1)N(\gamma_1\mathbf{Y}_1)\rangle_T e_{\gamma_2}^2\langle N(\alpha_2\mathbf{r}_2)N(\gamma_2\mathbf{Y}_2)\rangle_T\langle Q(\mathbf{X}_1)Q(\mathbf{X}_2)\rangle_T$$

$$\times\langle Q(\mathbf{x}_3)Q(\mathbf{x}_4)\rangle_T\lambda_{\gamma_1}^2\lambda_{\gamma_2}^2\int_0^1 ds\int_0^1 dt\int_0^1 du\int_0^1 dv\int\mathcal{D}(\omega_1)\mathcal{D}(\omega_2)\frac{1}{1!}[\omega_1(s)\cdot\nabla_{Y_1}]\phi(\mathbf{Y}_1-\mathbf{X}_1)\frac{1}{1!}$$

$$\times[\omega_1(t)\cdot\nabla_{Y_1}]\phi(\mathbf{Y}_1-\mathbf{X}_3)\frac{1}{1!}[\omega_2(u)\cdot\nabla_{Y_2}]\phi(\mathbf{Y}_2-\mathbf{X}_2)\frac{1}{1!}[\omega_2(v)\cdot\nabla_{Y_2}]\phi(\mathbf{Y}_2-\mathbf{X}_4)$$

$$=\frac{\beta^6\hbar^4}{288}\sum_{\gamma_1,\gamma_2}\frac{e_{\gamma_1}^2e_{\gamma_2}^2}{m_{\gamma_1}m_{\gamma_2}}\int d\mathbf{Y}_1 d\mathbf{Y}_2 d\mathbf{X}_1 d\mathbf{X}_2 d\mathbf{X}_3 d\mathbf{X}_4\langle N(\alpha_1\mathbf{r}_1)N(\gamma_1\mathbf{Y}_1)\rangle_T\langle N(\alpha_2\mathbf{r}_2)N(\gamma_2\mathbf{Y}_2)\rangle_T\langle Q(\mathbf{x}_1)Q(\mathbf{x}_2)\rangle_T$$

$$\times\langle Q(\mathbf{x}_3)Q(\mathbf{x}_4)\rangle_T[\nabla\phi(\mathbf{Y}_1-\mathbf{X}_1)\cdot\nabla\phi(\mathbf{Y}_1-\mathbf{X}_3)][\nabla\phi(\mathbf{Y}_2-\mathbf{X}_2)\cdot\nabla\phi(\mathbf{Y}_2-\mathbf{X}_4)]\ . \tag{6.40}$$

In (6.39) and (6.40), the convolution integrals of the type (6.32) over the pairs of nude field points $(\mathbf{X}_1,\mathbf{X}_2)$ and $(\mathbf{X}_3,\mathbf{X}_4)$ can be performed in terms of (6.33) plus exponentially decaying terms when $r_{12}\to\infty$. Therefore the algebraic contributions of $\Delta_2^{(4)}$ and $\Delta_3^{(4)}$ can be rewritten as

$$\frac{-\beta^4\hbar^4}{120}\sum_{\gamma_1,\gamma_2}\frac{e_{\gamma_1}^2e_{\gamma_2}^2}{m_{\gamma_1}m_{\gamma_2}}\int d\mathbf{Y}_1 d\mathbf{Y}_2\langle N(\alpha_1\mathbf{r}_1)N(\gamma_1\mathbf{Y}_1)\rangle_T\langle N(\alpha_2\mathbf{r}_2)N(\gamma_2\mathbf{Y}_2)\rangle_T f(|\mathbf{Y}_1-\mathbf{Y}_2|) \tag{6.41}$$

and

$$\frac{\beta^4\hbar^4}{288}\sum_{\gamma_1,\gamma_2}\frac{e_{\gamma_1}^2e_{\gamma_2}^2}{m_{\gamma_1}m_{\gamma_2}}\int d\mathbf{Y}_1 d\mathbf{Y}_2\langle N(\alpha_1\mathbf{r}_1)N(\gamma_1\mathbf{Y}_1)\rangle_T\langle N(\alpha_2\mathbf{r}_2)N(\gamma_2\mathbf{Y}_2)\rangle_T f(|\mathbf{Y}_1-\mathbf{Y}_2|)\ , \tag{6.42}$$

respectively. The total algebraic contribution of $\Delta_1^{(4)}$, $\Delta_2^{(4)}$, and $\Delta_3^{(4)}$, given by the sum of (6.38), (6.41), and (6.42), then does reduce to the expression (5.11), as it should. Consequently, the leading terms in the large-distance behaviors of $\rho_T^{(4)}(\alpha_1\mathbf{r}_1,\alpha_2\mathbf{r}_2)$ and $S^{(4)}(r_{12})$ are proportional to $1/r_{12}^6$ and $1/r_{12}^{10}$, respectively (see Sec. V).

Although the present diagrammatic analysis is more powerful than the method of Sec. V for evaluating the algebraic contributions at a given order in $\hbar$, the explicit calculations at the order $\hbar^6$ remain rather tedious, since one has 40 slow-decaying diagrams at this order.

VII. A MODEL: THE HYDROGEN ATOM IN A CLASSICAL PLASMA

It is of pedagogical value to study a simplified model in which all particles but two are classical. In this model, the effects of the intrinsic quantum fluctuations on screening can be more clearly displayed and discussed in a nonperturbative way. Two quantum-mechanical particles of charges e_1,e_2 and masses m_1,m_2 (e.g., an electron $-e$ and a proton e) are imbedded in a classical plasma and in thermal equilibrium with it. It is not important here to specify the detailed composition of the classical gas (an OCP or a multicomponent system) provided that it possesses all the strong screening properties of a classical plasma phase. The Hamiltonian of the two quantum particles in the presence of a configuration C of the classical gas in volume Λ is

$$H(C)=\frac{|\mathbf{p}_1|^2}{2m_1}+\frac{|\mathbf{p}_2|^2}{2m_2}+e_1e_2\phi(\mathbf{r}_1-\mathbf{r}_2)+e_1\int_\Lambda d\mathbf{r}\,\phi(\mathbf{r}_1-\mathbf{r})Q(\mathbf{r},C)+e_2\int_\Lambda d\mathbf{r}\,\phi(\mathbf{r}_2-\mathbf{r})Q(\mathbf{r},C)\ , \tag{7.1}$$

where $Q(\mathbf{r},C)$ is the microscopic charge density of the classical gas corresponding to this configuration.

We consider the equilibrium distribution of the two quantum charges at $\mathbf{r}_1$ and $\mathbf{r}_2$

$$\rho_\Lambda(\mathbf{r}_1,\mathbf{r}_2)=\frac{\int_\Lambda dC(\mathbf{r}_1,\mathbf{r}_2|\exp[-\beta H(C)]|\mathbf{r}_1,\mathbf{r}_2)\exp[-\beta U_0(C)]}{\int_\Lambda dC\,\exp[-\beta U_0(C)]}\ , \tag{7.2}$$

where $U_0(C)$ is the Coulomb energy of the classical gas and $(\mathbf{r}_1,\mathbf{r}_2|\exp[-\beta H(C)]|\mathbf{r}_1,\mathbf{r}_2)$ is the diagonal configurational kernel of $\exp[-\beta H(C)]$. According to the Feynman-Kac formula and the Brownian-bridge representation, this kernel is given by the functional integral [see (6.2)]

$$(\mathbf{r}_1,\mathbf{r}_2|\exp[-\beta H(C)]|\mathbf{r}_1,\mathbf{r}_2)=\frac{1}{(2\pi\lambda_1\lambda_2)^3}\int\mathcal{D}(\xi_1)\int\mathcal{D}(\xi_2)\exp[-\beta E(\mathbf{r}_1,\xi_1;\mathbf{r}_2,\xi_2,\xi_2;C)]\ , \tag{7.3}$$

with

$$E(\mathbf{r}_1,\xi_1;\mathbf{r}_2,\xi_2;C)=e_1e_2\int_0^1 ds\,\phi(\mathbf{r}_1-\mathbf{r}_2+\lambda_1\xi_1(s)-\lambda_2\xi_2(s))+\sum_{i=1,2}\int_\Lambda d\mathbf{r}\int_0^1 ds\,\phi(\mathbf{r}_i+\lambda_i\xi_i(s)-\mathbf{r})Q(\mathbf{r},C)\ . \tag{7.4}$$

As discussed in Sec. VI, one can think of the quantum particles as random charged filaments at $\mathbf{r}_1$ and $\mathbf{r}_2$ with charge densities

$$e_in_i(\mathbf{r})=e_i\int_0^1 ds\,\delta(\mathbf{r}_i+\lambda_i\xi_i(s)-\mathbf{r}),\quad i=1,2\ . \tag{7.5}$$

It is of interest to compare the quantum energy (7.4) for fixed ξ_1,ξ_2 with the corresponding classical interaction electrostatic energy of the two filamentous charges (7.5) in the configuration C, i.e.,

$$E_{\rm cl}(\mathbf{r}_1,\xi_1;\mathbf{r}_2,\xi_2;C)=e_1e_2\int_\Lambda d\mathbf{r}\int_\Lambda d\mathbf{r}'n_1(\mathbf{r})\phi(\mathbf{r}-\mathbf{r}')n_2(\mathbf{r}')+\sum_{i=1}^{2}e_i\int_\Lambda d\mathbf{r}\int_\Lambda d\mathbf{r}'n_i(\mathbf{r})\phi(\mathbf{r}-\mathbf{r}')Q(\mathbf{r}',C)\ . \tag{7.6}$$

The self energy of the filaments is not included in (7.6). The comparison of (7.4) and (7.6) gives

$$E(\mathbf{r}_1,\xi_1;\mathbf{r}_2,\xi_2;C)=E_{\rm cl}(\mathbf{r}_1,\xi_1;\mathbf{r}_2,\xi_2;C)+W(\mathbf{r}_1,\xi_1;\mathbf{r}_2,\xi_2)\ , \tag{7.7}$$

with

$$W(\mathbf{r}_1,\xi_1;\mathbf{r}_2,\xi_2)=e_1e_2\int_0^1 ds_1\int_0^1 ds_2[\delta(s_1-s_2)-1]\phi(\mathbf{r}_1-\mathbf{r}_2+\lambda_1\xi_1(s_1)-\lambda_2\xi_s(s_2))\ . \tag{7.8}$$

For fixed ξ_1,ξ_2, we note that $U_0(C)+E_{\rm cl}(\mathbf{r}_1,\xi_1;\mathbf{r}_2,\xi_2;C)=U(C)$ is precisely the total electrostatic energy of the configuration C of the classical gas in the presence of the two additional charges (7.5). So inserting the representation (7.3) with the decomposition (7.7) into (7.2), we can write

$$\rho_\Lambda(\mathbf{r}_1,\mathbf{r}_2)=\frac{1}{(2\pi\lambda_1\lambda_2)^3}\int\mathcal{D}(\xi_1)\int\mathcal{D}(\xi_2)\exp[-\beta W(\mathbf{r}_1,\xi_1;\mathbf{r}_2,\xi_2)]\frac{Z_\Lambda(\mathbf{r}_1,\xi_1;\mathbf{r}_2,\xi_2)}{Z_\Lambda}\ , \tag{7.9}$$

where $\mathcal{Z}_\Lambda = \int_\Lambda dC \exp[-\beta U_0(C)]$ is the partition function of the classical gas and

$$\mathcal{Z}_\Lambda = \int_\Lambda dC \exp\{-\beta[U_0(C) + E_{\rm cl}(\mathbf{r}_1,\xi_1;\mathbf{r}_2,\xi_2;C)]\}$$

is the partition function of the same gas in the presence of the two additional external charges (7.5).

In the same way, we define the one-point distribution functions

$$\rho_\Lambda(\mathbf{r}_1) = \frac{\int_\Lambda dC (\mathbf{r}_i|\exp[-\beta H_i(C)]|\mathbf{r}_i)\exp[-\beta U_0(C)]}{\int_\Lambda dC \exp[-\beta U_0(C)]}, \tag{7.10}$$

with

$$H_i(C) = \frac{|\mathbf{p}_i|^2}{2m_i} + e_i \int_\Lambda d\mathbf{r}\, \phi(\mathbf{r}_i - \mathbf{r}) Q(\mathbf{r}, C), \quad i = 1,2 \tag{7.11}$$

corresponding to the immersion of a single quantum particle in the classical gas. They have a representation analogous to (7.9),

$$\rho(\mathbf{r}_i) = \left[\frac{1}{\sqrt{2\pi}\lambda_i}\right]^3 \int \mathcal{D}(\xi_i) \frac{\mathcal{Z}_\Lambda(\mathbf{r}_i,\xi_i)}{\mathcal{Z}_\Lambda}, \tag{7.12}$$

where $\mathcal{Z}_\Lambda(\mathbf{r}_i,\xi_i)$ is the partition function of the classical gas in the presence of a single external filamentous charge (7.5).

We introduce finally the excess free energies (considered from now on in the infinite volume limit)

$$F(\mathbf{r}_1-\mathbf{r}_2,\xi_1,\xi_2) = -\lim_{|\Lambda|\to\infty} \frac{1}{\beta} \ln\left[\frac{\mathcal{Z}_\Lambda(\mathbf{r}_1,\xi_2;\mathbf{r}_2,\xi_2)}{\mathcal{Z}_\Lambda}\right], \tag{7.13}$$

$$F(\xi_i) = -\lim_{|\Lambda|\to\infty} \frac{1}{\beta} \ln\left[\frac{\mathcal{Z}_\Lambda(\mathbf{r}_i,\xi_i)}{\mathcal{Z}_\Lambda}\right], \quad i = 1,2 \tag{7.14}$$

and the effective classical potential (the energy needed to separate the charged filaments at $\mathbf{r}_1$ and $\mathbf{r}_2$ to infinity in the gas)

$$\phi_{\rm eff}(\mathbf{r}_1-\mathbf{r}_2,\xi_1,\xi_2) = F(\mathbf{r}_1-\mathbf{r}_2,\xi_1,\xi_2) - F(\xi_1) - F(\xi_2). \tag{7.15}$$

With (7.9), (7.12), and these definitions, the dimensionless truncated correlation of the quantum charges

$$g(\mathbf{r}_1-\mathbf{r}_2) = \lim_{|\Lambda|\to\infty} \left[\frac{\rho_\Lambda(\mathbf{r}_1,\mathbf{r}_2)}{\rho_\Lambda(\mathbf{r}_1)\rho_\Lambda(\mathbf{r}_2)} - 1\right] \tag{7.16}$$

takes the simple form

$$g(\mathbf{r}) = \int \bar{\mathcal{D}}(\xi_1) \int \bar{\mathcal{D}}(\xi_2) (\exp\{-\beta[\phi_{\rm eff}(\mathbf{r},\xi_1,\xi_2) + W(\mathbf{r},\xi_1,\xi_2)]\} - 1). \tag{7.17}$$

In (7.17), $\bar{\mathcal{D}}(\xi_i)$ are non-Gaussian measures renormalized by the one particle excess free energy (7.14),

$$\bar{\mathcal{D}}(\xi_i) = \frac{\exp[-\beta F(\xi_i)]}{\int \mathcal{D}(\xi_i)\exp[-\beta F(\xi_i)]} \mathcal{D}(\xi_i). \tag{7.18}$$

The quantities $F(\xi_i)$ and $\phi_{\rm eff}(\mathbf{r},\xi_1,\xi_2)$ pertain to the purely classical system, which is assumed to be in a perfectly screening exponentially clustering plasma phase. Thus for fixed ξ_1 and ξ_2 and $|\mathbf{r}_1-\mathbf{r}_2|$ large, the filamentous charges are localized in the neighborhood of $\mathbf{r}_1$ and $\mathbf{r}_2$, respectively, and their effective potential $\phi_{\rm eff}(\mathbf{r}_1-\mathbf{r}_2,\xi_1,\xi_2)$ decays exponentially fast as $|\mathbf{r}_1-\mathbf{r}_2|\to\infty$. To make the model completely definite let us write down the expressions of $F(\xi_i)$ and $\phi_{\rm eff}(\mathbf{r},\xi_1,\xi_2)$, when the classical plasma is treated in the Debye-Hückel approximation. This approximation is legitimate when the dimensionless coupling parameter Γ of the plasma is small. One finds (see Appendix G)

$$F^{\rm DH}(\xi_i) = \frac{e_i^2}{2} \int_0^1 ds_1 \int_0^1 ds_2 \frac{\exp[-\varkappa\lambda_i|\xi_i(s_1)-\xi_i(s_2)|]-1}{\lambda_i|\xi_i(s_1)-\xi_i(s_2)|}, \tag{7.19}$$

$$\phi_{\rm eff}^{\rm DH}(\mathbf{r},\xi_1,\xi_2) = e_1 e_2 \int_0^1 ds_1 \int_0^1 ds_2 \frac{\exp[-\varkappa|\mathbf{r}+\lambda_1\xi_1(s_1)-\lambda_2\xi_2(s_2)|]}{|\mathbf{r}+\lambda_1\xi_1(s_1)-\lambda_2\xi_2(s_2)|}, \tag{7.20}$$

where $\varkappa$ is the inverse Debye length of the classical plasma.

The potential $W(\mathbf{r},\xi_1,\xi_2)$ has a purely quantum-mechanical origin and is easily seen from (7.8) to be dipolar [the terms of order $|\mathbf{r}|^{-1}$ and $|\mathbf{r}|^{-2}$ vanish because $\int_0^1 ds[\delta(t-s)-1]=0$]

$$W(\mathbf{r},\xi_1,\xi_2) \sim \frac{e_1e_2}{2} \int_0^1 ds_1 \int_0^1 ds_2 [\delta(s_1-s_2)-1]\{[\lambda_1\xi_1(s_1)-\lambda_2\xi_2(s_2)]\cdot\nabla\}^2 \frac{1}{|\mathbf{r}|}, \quad |\mathbf{r}|\to\infty. \tag{7.21}$$

Since $\phi_{\rm eff}(\mathbf{r},\xi_1,\xi_2)$ is exponential, it is precisely this quantum-mechanical part $W(\mathbf{r},\xi_1,\xi_2)$ which governs the asymptotic behavior of $g(\mathbf{r})$ as $|\mathbf{r}|\to\infty$. We can thus generate the inverse power asymptotic development of $g(r)$ by neglecting $\phi_{\rm eff}(\mathbf{r},\xi_1,\xi_2)$ and expanding $\exp[-\beta W(\mathbf{r},\xi_1,\xi_2)]$ in (7.17),

$$g(\mathbf{r}) \simeq -\beta \int \bar{\mathcal{D}}(\xi_1) \int \bar{\mathcal{D}}(\xi_2) W(\mathbf{r},\xi_1,\xi_2) + \frac{\beta^2}{2} \int \bar{\mathcal{D}}(\xi_1) \int \bar{\mathcal{D}}(\xi_2) [W(\mathbf{r},\xi_1,\xi_2)]^2 + \cdots. \tag{7.22}$$

The first term in the rhs of (7.22) has no algebraic term. Indeed, using the formula

$$\phi(\mathbf{r}+\mathbf{a})=\sum_{n=0}^{\infty}\frac{(\mathbf{a}\cdot\nabla)^n}{n!}\phi(\mathbf{r}) \tag{7.23}$$

the nth-order term of its multipole expansion involves the quantity

$$\int\bar{\mathcal{D}}(\xi_1)\int\bar{\mathcal{D}}(\xi_2)\{[\lambda_1\xi_1(s_1)-\lambda_2\xi_2(s_2)]\cdot\nabla\}^n\phi(\mathbf{r}) \ . \tag{7.24}$$

It is clear from the definition (6.3) of the Brownian-bridge process and from (7.14) that the measure $\bar{\mathcal{D}}(\xi_i)$ is invariant under the spatial inversion and rotations of the path ξ_i. Hence the integral of any odd product of $\xi_i(s_i)$ vanishes and the integral of an even product

$$\int\bar{\mathcal{D}}(\xi_i)[\xi_i(s_i)\cdot\nabla]^{2m}$$

is necessarily proportional to $(\nabla^2)^m$. Since the functional $F(\xi_i)$ (7.14) is bounded and $\mathcal{D}(\xi_i)$ is Gaussian, all moments of $\bar{\mathcal{D}}(\xi_i)$ exist. Thus the expression (7.24) vanishes for $n\geq 2$ and $\mathbf{r}\neq 0$ because of the harmonicity of the Coulomb potential [there is obviously no monopole contribution in (7.8)].

The slowest asymptotic term of $g(\mathbf{r})$ comes from the second term of the development (7.22), when $W(\mathbf{r},\xi_1,\xi_2)$ is taken at the dipolar order (7.21). Using again the fact that certain terms vanish for the same symmetry reasons as (7.24) and $\int_0^1 ds[\delta(s-t)-1]=0$ one finds after some algebra that

$$\frac{\beta^2}{2}\int\bar{\mathcal{D}}(\xi_1)\int\bar{\mathcal{D}}(\xi_2)[W(\mathbf{r},\xi_1,\xi_2)]^2\sim\frac{B}{|\mathbf{r}|^6}\ , \quad |\mathbf{r}|\to\infty \tag{7.25}$$

with

$$B=\frac{\beta^2}{2}(e_1\lambda_1e_2\lambda_2)^2\int\bar{\mathcal{D}}(\xi_1)\int\bar{\mathcal{D}}(\xi_2)\left[\int_0^1 ds_1\int_0^1 ds_2[\delta(s_1-s_2)-1][\xi_1(s_1)\cdot\nabla][\xi_2(s_2)\cdot\nabla]\frac{1}{|\mathbf{r}|}\bigg|_{|\mathbf{r}|=1}\right]^2 . \tag{7.26}$$

The coefficient B (the integral of a positive function) is obviously different from zero, and thus (7.25) represents the exact asymptotic behavior of the correlation between the two quantum particles in the classical gas. An expression of B in terms of the physical constants would require us to perform explicitly the functional integral (7.26) with the non-Gaussian measures $\bar{\mathcal{D}}(\xi_i)$ involving the one-particle excess free energy (7.14). It can be noted from (7.14) that B is not a polynomial function of the Planck constant so there are $|\mathbf{r}|^{-6}$ contributions at arbitrary high orders in $\hbar$. Since the prefactor $(\lambda_1\lambda_2)^2$ in (7.26) is already of order $\hbar^4$, to obtain B at this order it suffices to replace $F(\xi_i)$ by its classical value F_{cl}^i which is independent of ξ_i, and then, according to (7.18), $\bar{\mathcal{D}}(\xi_i)$ by the Brownian-bridge measures $\mathcal{D}(\xi_i)$ [in the Debye-Hückel approximation (7.19), $F_{\text{cl}}^i=-\varkappa e^2/2$]. Using the rules for Gaussian moments and the covariance (6.3) one finds as in Sec. VI D

$$B=\hbar^4\frac{\beta^4}{240}\frac{e_1^2}{m_1}\frac{e_2^2}{m_2}+o(\hbar^4)\ . \tag{7.27}$$

The special model studied in this section can be viewed as a particular limit of the general quantum multicomponent system described in Secs. V and VI. In this limit the densities ρ_1 and ρ_2 of the species 1 and 2 go to zero, while the masses m_α, $\alpha=3,\ldots,M$, of the other species go to infinity. We have checked that the expression (7.27) of the coefficient B at the order $\hbar^4$ is indeed recovered from the general expression (5.12) by taking the limit of $\rho_T^{(4)}(\alpha_1\mathbf{r},\alpha_2 0)/\rho_1\rho_2$ when $\rho_1,\rho_2\to 0$ and $m_\alpha\to\infty$, $\alpha=3,\ldots,M$ (the other parameters being kept fixed). In this verification, we have used $(i=1,2)$

$$\begin{aligned}
&\lim_{\rho_1,\rho_2\to 0}\frac{[\langle N(\alpha_i\mathbf{r})N(\alpha_i\mathbf{0})\rangle_T-\rho_i\delta(\mathbf{r})]}{\rho_i^2}=h_{ii}(r)\ ,\\
&\lim_{\rho_1,\rho_2\to 0}\frac{\langle N(\alpha_1\mathbf{r})N(\alpha_2\mathbf{0})\rangle_T}{\rho_1\rho_2}=h_{12}(r)\ ,\\
&\lim_{\rho_1,\rho_2\to 0}\frac{\langle N(\alpha_i\mathbf{0})N(\alpha\mathbf{r})\rangle_T}{\rho_i}=\rho_\alpha h_{i\alpha}(r)\ ,\quad \alpha=3,\ldots,M
\end{aligned} \tag{7.28}$$

where h_{ii}, h_{12}, and $h_{i\alpha}$, $\alpha=3,\ldots,M$, are well-behaved classical Ursell functions, whose fast decay is governed by finite correlation lengths (in the present limit, the classical screening of the charges of the infinitely dilute species 1 and 2, is ensured by the charges of the other species which have nonzero fixed densities). Furthermore, the nonperturbative calculation of the present section shows that, in the above limit, the findings relative to the resummation of the large-distance behavior of the $\hbar^{2n}$ terms of the WK expansion are indeed satisfied (see Sec. VI).

VIII. CONCLUSION

In this paper we have given strong evidences that there is no exponential clustering for the equilibrium correlations of quantum charged fluids. These evidences rely on the perturbative Wigner-Kirkwood expansion (Secs. IV and VI), the imaginary-time evolution equations (Secs. II and V), and the model of two quantum charges immersed in a classical charged fluid (Sec. VII). Each term of the WK expansion of the correlations decays algebraically at large distances. This expansion then provides algebraic lower bounds (under assumptions relative to the convergence of the former). The analysis of the large-distance structure of the imaginary-time evolution equations provides upper bounds which are compatible with the perturbative results. The model allows an exact calculation of the asymptotic behavior of the correlations of the quantum charges, which is found to be algebraic (like $1/|\mathbf{r}|^6$) and to satisfy the above bounds. Thus, from this ensemble of confluent arguments, we infer that the Debye screening does not exist, strictly speaking, in real matter.

Of course, the absence of exponential clustering does not mean that there is no screening at all. The equilibrium equations of Sec. II are formulated in terms of the bare Coulomb potential, and at first sight, particle correlations could decay as slow as $|\mathbf{r}|^{-3}$. The analysis of Sec. II precisely shows that the slowest decaying terms do not occur for fundamental reasons (KMS, locality). The same remark applies to the diagrammatic analysis of Sec. VI where the longest-range contributions are also excluded by general rules. As a result, the charge sum rule, which states that the total net charge vanishes, still holds as in the classical case (as well as the dipole sum rule for the OCP). But the higher multipole sum rules no longer hold in general, as shown in Secs. III–V. Furthermore, the screening of external classical charges should be better than the screening of the quantum charges of the medium. In particular, the response functions should decay faster than the internal correlations of the system.

The basic mechanism which induces the algebraic tails in the quantum correlations is linked to the intrinsic quantum fluctuations. In the functional integration formalism, these fluctuations can be represented by filamentous charge distributions. The interaction potential between the quantum charges involves multipolelike interactions, arising from the "ghost" multipoles associated with the filamentous distributions. As illustrated by the perturbative analysis of Sec. VI, or by the exact calculation of Sec. VII, it is precisely these multipolelike interactions which are responsible for the algebraic tails (this mechanism is similar to the pollution scenario conjectured by Brydges and Seiler[13]). Since the above mechanism is an intrinsic feature of quantum mechanics, the algebraic decay of the correlations should occur at any value of the thermodynamic parameters. We have checked that the same effects occur in a classical system when all the ions have an internal structure giving rise to permanent dipoles. In this case also, this internal degree of freedom prevents the screening clouds from being perfectly organized, and exponential decay is lost. However, the correlations of structureless charges in a dipolar solvent are expected to decay exponentially fast.[31] Another analogy, more natural, can be made between quantum statics and classical dynamics of point charges. In both cases, velocity and positional distributions cannot be disentangled. In the classical dynamical evolution, any perfect arrangement of the clouds is broken by the collision processes, and the system cannot instantaneously restore such an arrangement because of inertia effects. In the quantum static case, the quantum fluctuations themselves are the disturbing factor.

We now comment on the role played by Fermi statistics. We have given the explicit forms of the corresponding algebraic tails for several models, in the semiclassical regime (high temperatures and low densities) and in the framework of Maxwell-Boltzmann statistics. For the OCP, these statistics can be used without modifying the bare Coulomb potential, because all the mobile charges repel themselves. In the classical limit, the exchange effects arising from Fermi statistics are expected to be exponentially small with $\hbar$.[32] Therefore the expressions computed in Sec. IV are indeed the dominant terms in the semiclassical regime for an OCP described by Fermi statistics. For a multicomponent system, with positive and negative mobile charges, the use of Maxwell-Boltzmann statistics implies a regularization of the Coulomb potential at short distances, in order to prevent the collapse between oppositely charged particles. For a real system described by Fermi statistics (such as mixture of protons and electrons), this regularization procedure is introduced on semiheuristic grounds[33] with the help of an effective $\hbar$-dependent potential. Thus the expressions derived in Secs. V and VI cannot be viewed as the leading terms of a systematic expansion for the real system.

Furthermore, in a system of nuclei and electrons, atoms or molecules can be formed with the familiar van der Waals forces between them. Notice that these forces are usually computed in vacuum.[34] Our results show that the van der Waals forces cannot be exponentially screened by free quantum charges that are always present in a nonzero density state. Therefore, in the presence of atoms or molecules, there are two different mechanisms providing algebraic tails. The first one, described in this paper, exists intrinsically and independently of any binding process, while the second one (the van der Waals forces) is due to the polarizability of quantum bound states. The determination, starting from first principles, of the precise form of the total algebraic tails induced by the combination of the intrinsic and van der Waals mechanisms is a difficult problem, even in the semiclassical regime.

Our techniques obviously do not apply to the ground state. At zero temperature, it is well known[3] that the discontinuity of the Fermi distribution induces algebraic Friedel oscillations (like $1/|r|^3$) in the correlations. As far as the screening mechanisms are concerned, we expect that the intrinsic quantum fluctuations should certainly be not less disturbing than at finite temperatures, inducing also algebraic tails in addition to the Friedel term. This is supported by the observation that the coefficients of the algebraic tails computed in Secs. IV and VI diverge at $T=0$.

It is important to stress again that the usual mean-field theories such as RPA or Thomas-Fermi do not take into account properly the intrinsic quantum fluctuations since they predict an exponential clustering (see Appendix H). The situation is similar in classical dynamics where the Vlasov approximation[35] does not reproduce the spatial algebraic decay of time-dependent correlations.[17,18] This is because the Vlasov dynamics reduces to a one-body motion in a mean-field potential and consequently does not include the collision process. A better understanding of the failure of the RPA approximation, as well as how it should be improved to incorporate the effects discussed in this paper, is an open problem.

We finally discuss the possible observable implications of our findings. Since our explicit calculations pertain to the semiclassical regime, we only consider real systems under conditions such that the quantum effects are small. The first one is a sodium chloride electrolyte at room temperature, and the second one is the interior of some white dwarf made of ^{12}C nuclei embedded in a nonpolarizable degenerate electron gas. In both cases, we determine the crossover distance r_0 above which quantum algebraic tails dominate the classical behavior, equating the expression (4.4) to the usual exponential law, i.e.,

$$\frac{\rho e^2}{4\pi\xi^2}\frac{\exp(-r_0/\xi)}{r_0}=\frac{7}{16\pi^2}\left[\frac{\beta e}{m}\right]^2\frac{\hbar^4}{r_0^{10}} . \tag{8.1}$$

In (8.1), ξ is the screening length, and the classical exponential is normalized in order to obey the charge sum rule. This normalization as well as the use of (4.4) for a two-component system give correctly the order of magnitude r_0. Setting $x_0=r_0/\xi$, the crossover equation can be rewritten in the form (up to a dimensionless multiplicative numerical constant)

$$x_0^9e^{-x_0}\sim\Gamma\left[\frac{a}{\xi}\right]^7\left[\frac{a_B}{a}\right]\left[\frac{\lambda_{\mathrm{dB}}}{a}\right]^2 , \tag{8.2}$$

where $a\sim\rho^{-1/3}$ is the mean interparticle distance, $\Gamma=\beta e^2/a$ is the coupling constant, $a_B=\hbar^2/me^2$ is the Bohr radius, and $\lambda_{\mathrm{dB}}\sim(\beta\hbar^2/m)^{1/2}$ is the de Broglie thermal wavelength. In a semiclassical regime, the two last factors in the rhs of (8.2) are obviously small, implying r_0 large compared to ξ [ξ is of the order of few interparticle distances and the classical factor $(a/\xi)^7/\Gamma$ is of order 1]. For instance, for the first system, $T=300$ K, $a\sim16$ Å (for a decimolar solution), $\xi\sim a$, $\Gamma\sim1$ (e^2 must be divided by the dielectric constant of water which is close to 80), $a_B\sim0.76\times10^{-3}$ Å (here m is taken equal to the average of the atomic masses of Na and Cl) and $\lambda_{\mathrm{dB}}\sim0.073$ Å. Then (8.2) gives $x_0\sim60$. For the second system, $T=10^8$ K, $a\sim360$ F ($\rho\sim10^8$ g/cm^3), $\xi\sim a$, $\Gamma\sim16$, $a_B\sim0.066$ F (e^2 must be multiplied by Z^2 with $Z=6$ for ^{12}C, and m is the atomic mass of ^{12}C), and $\lambda_{\mathrm{dB}}\sim200$ F. Then, (8.2) gives $x_0\sim40$. In both cases, the quantum effects on the clustering turn out to be very small, and consequently the use of exponentially screened effective potentials is legitimate from a quantitative point of view. However, if we apply crudely (8.2) for the electrons in a metal (replacing ξ by the Thomas-Fermi screening length λ_{TF}), all lengths occurring in (8.2) have the same order of magnitude and r_0 is of the order λ_{TF}. This indicates that the use of an exponential effective potential might be less reliable in this case.

ACKNOWLEDGMENTS

Laboratoire de Physique Theorique et Hautes Energies associé au Centre National de la Recherche Scientifique. One of us (P.A.M.) was partially supported by the Swiss National Foundation for Science.

APPENDIX A: PROOF OF LEMMA 1

We split the $\mathbf{r}'$ integral over the two regions $D_{\mathbf{r}}=\{\mathbf{r}'\,|\,|\mathbf{r}-\mathbf{r}'|\le|\mathbf{r}|/2\}$ and its complement $\bar{D}_{\mathbf{r}}$. In $D_{\mathbf{r}}$ one has $|\mathbf{r}'|\ge|\mathbf{r}|/2$, and by (2.27), $||\mathbf{r}|^{n_1}g(\mathbf{r}-\mathbf{r}')|\le M(|\mathbf{r}-\mathbf{r}'|)$. This shows that for $|\mathbf{r}'|\ge|\mathbf{r}|/2$, $|\mathbf{r}|^{n_1}|\mathbf{F}(\mathbf{r}')g(\mathbf{r},\mathbf{r}+\mathbf{r}')|$ is bounded uniformly with respect to $\mathbf{r}$ by the function $|\mathbf{F}(\mathbf{r}')|M(|\mathbf{r}'|)$, which is integrable when (2.28) holds. Hence it follows by dominated convergence that

$$\begin{aligned}\lim_{|\mathbf{r}|\to\infty}|\mathbf{r}|^{n_1}\int_{D_{\mathbf{r}}}d\mathbf{r}'\mathbf{F}(\mathbf{r}-\mathbf{r}')g(\mathbf{r},\mathbf{r}')&=\lim_{|\mathbf{r}|\to\infty}|\mathbf{r}|^{n_1}\int_{|\mathbf{r}'|\le|\mathbf{r}|/2}d\mathbf{r}'\mathbf{F}(\mathbf{r}')g(\mathbf{r},\mathbf{r}+\mathbf{r}')\\&=\int d\mathbf{r}'\mathbf{F}(\mathbf{r}')h(\mathbf{r}')\end{aligned} \tag{A1}$$

We divide again $\bar{D}_{\mathbf{r}}=\bar{D}_{\mathbf{r}}^{(1)}\cup\bar{D}_{\mathbf{r}}^{(2)}$ into the two regions

$$\begin{aligned}\bar{D}_{\mathbf{r}}^{(1)}&=\{\mathbf{r}'\,|\,|\mathbf{r}-\mathbf{r}'|\ge|\mathbf{r}|/2,|\mathbf{r}'|\le|\mathbf{r}-\mathbf{r}'|\} ,\\ \bar{D}_{\mathbf{r}}^{(2)}&=\{\mathbf{r}'\,|\,|\mathbf{r}-\mathbf{r}'|\ge|\mathbf{r}|/2,|\mathbf{r}'|\ge|\mathbf{r}-\mathbf{r}'|\} .\end{aligned} \tag{A2}$$

In $\bar{D}_{\mathbf{r}}^{(1)}$ we have, when (2.28) holds,

$$\begin{aligned}|\mathbf{r}|^{n_2}\left|\int_{\bar{D}_{\mathbf{r}}^{(1)}}d\mathbf{r}'\mathbf{F}(\mathbf{r}-\mathbf{r}')g(\mathbf{r},\mathbf{r}')\right|&\le\int_{\bar{D}_{\mathbf{r}}^{(1)}}d\mathbf{r}'\frac{M(|\mathbf{r}'|)}{|\mathbf{r}-\mathbf{r}'|^2}\le\left[\frac{2}{|\mathbf{r}|}\right]^{2-\epsilon}\int_{|\mathbf{r}'|\le|\mathbf{r}-\mathbf{r}'|}d\mathbf{r}'\frac{M(|\mathbf{r}'|)}{|\mathbf{r}-\mathbf{r}'|^{\epsilon}}\\&\le\left[\frac{2}{|\mathbf{r}|}\right]^{2-\epsilon}\int d\mathbf{r}'\frac{M(|\mathbf{r}'|)}{|\mathbf{r}'|^{\epsilon}}=O\left[\frac{1}{|\mathbf{r}|^{2-\epsilon}}\right] .\end{aligned} \tag{A3}$$

Similarly we find in $\bar{D}_{\mathbf{r}}^{(2)}$

$$\begin{aligned}|\mathbf{r}|^{n_2}\left|\int_{\bar{D}_{\mathbf{r}}^{(2)}}d\mathbf{r}'\mathbf{F}(\mathbf{r}-\mathbf{r}')g(\mathbf{r},\mathbf{r}')\right|&\le\int_{\bar{D}_{\mathbf{r}}^{(2)}}d\mathbf{r}'\frac{M(|\mathbf{r}-\mathbf{r}'|)}{|\mathbf{r}-\mathbf{r}'|^2}\\&\le\left[\frac{2}{|\mathbf{r}|}\right]^{2-\epsilon}\int_{|\mathbf{r}'|\ge|\mathbf{r}-\mathbf{r}'|}d\mathbf{r}'\frac{M(|\mathbf{r}-\mathbf{r}'|)}{|\mathbf{r}-\mathbf{r}'|^{\epsilon}}\\&\le\left[\frac{2}{|\mathbf{r}|}\right]^{2-\epsilon}\int d\mathbf{r}'\frac{M(|\mathbf{r}'|)}{|\mathbf{r}'|^{\epsilon}}=O\left[\frac{1}{|\mathbf{r}|^{2-\epsilon}}\right] .\end{aligned} \tag{A4}$$

The combination of (A1), (A3), and (A4) gives the result of the lemma.

APPENDIX B: TERM (4.40b)

Using the property (4.34), the first term in the right-hand side of (4.40b) is expressed with the help of the two-point classical correlations

$$\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)(\nabla^2)^2\rangle=(\nabla_1^2+\nabla_2^2)^2\rho(\mathbf{r}_1,\mathbf{r}_2)=(\nabla_1^2+\nabla_2^2)^2\rho_T(\mathbf{r}_1,\mathbf{r}_2)\ . \tag{B1}$$

This quantity is rapidly decaying as $|\mathbf{r}_1-\mathbf{r}_2|\to\infty$.

To treat the second term (4.40b) in compact form, it is useful to introduce the operators

$$D^{\mu}(\mathbf{r})=\sum_{i=1}^{n}\delta(\mathbf{r}-\mathbf{r}_i)\nabla_i^{\mu}\ , \tag{B2}$$

$$D^{\mu\nu}(\mathbf{r})=\sum_{i=1}^{n}\delta(\mathbf{r}-\mathbf{r}_i)\nabla_i^{\mu}\nabla_i^{\nu}\ . \tag{B3}$$

Singling out the contribution of coincident particles

$$\sum_{p,q}\sum_{\mu,\nu}^{3}\left[\sum_{\substack{i,j=1\\ i\neq j}}^{n}+\sum_{\substack{i,j=1\\ i=j}}^{n}\right]$$

and taking (4.41) into account, the second term in (4.40b) can be written as

$$\begin{aligned}
&\sum_{p,q}[\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)(\nabla_p\nabla_q V)\nabla_p\nabla_q\rangle-\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle\langle(\nabla_p\nabla_q V)\nabla_p\nabla_q\rangle]\\
&\quad=-e^2\sum_{\mu,\nu=1}^{3}\int d\mathbf{r}_3\int d\mathbf{r}_4\phi^{\mu\nu}(\mathbf{r}_3-\mathbf{r}_4)\\
&\qquad\qquad\times\{\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)[D^{\mu}(\mathbf{r}_3)D^{\nu}(\mathbf{r}_4)]_{\mathrm{nc}}\rangle-\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle\langle[D^{\mu}(\mathbf{r}_3)D^{\nu}(\mathbf{r}_4)]_{\mathrm{nc}}\rangle\} &&\text{(B4a)}\\
&\quad\quad+e^2\sum_{\mu,\nu=1}^{3}\int d\mathbf{r}_3\int d\mathbf{r}_4\phi^{\mu\nu}(\mathbf{r}_3-\mathbf{r}_4)\{\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)[Q(\mathbf{r}_3)D^{\mu\nu}(\mathbf{r}_4)]_{\mathrm{nc}}\rangle-\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)\rangle\langle[Q(\mathbf{r}_3)D^{\mu\nu}(\mathbf{r}_4)]_{\mathrm{nc}}\rangle\}\ . &&\text{(B4b)}
\end{aligned}$$

The decomposition of (B4a) in fully truncated correlations is easily done with (4.48). Since $\langle Q(\mathbf{r})\rangle=\langle D^{\mu}(\mathbf{r})\rangle=0$ by neutrality and translation invariance, only the partitions [4] and [2,2] contribute. The partition [4] involves the fully truncated correlation $\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)[D^{\mu}(\mathbf{r}_3)D^{\nu}(\mathbf{r}_4)]_{\mathrm{nc}}\rangle_T$ which is rapidly decreasing as $|\mathbf{r}_1-\mathbf{r}_2|\to\infty$. The contribution of the partitions [2,2] to (B4a) is equal to

$$\begin{aligned}
&-2e^2\sum_{\mu,\nu=1}^{3}\int d\mathbf{r}_3\int d\mathbf{r}_4\phi_l^{\mu\nu}(\mathbf{r}_3-\mathbf{r}_4)\langle Q(\mathbf{r}_1)D^{\mu}(\mathbf{r}_3)\rangle_T\langle Q(\mathbf{r}_2)D^{\nu}(\mathbf{r}_4)\rangle_T\\
&\quad=-2e^4\sum_{\mu,\nu=1}^{3}\int d\mathbf{r}_3\int d\mathbf{r}_4\phi_l^{\mu\nu}(\mathbf{r}_3-\mathbf{r}_4)\nabla_3^{\mu}\rho_T(\mathbf{r}_1,\mathbf{r}_3)\nabla_4^{\nu}\rho_T(\mathbf{r}_2,\mathbf{r}_4)=2e^4\int d\mathbf{r}_3\int d\mathbf{r}_4[(\nabla^2)^2\phi_l](\mathbf{r}_3-\mathbf{r}_4)\rho_T(\mathbf{r}_1,\mathbf{r}_3)\rho_T(\mathbf{r}_2,\mathbf{r}_4)\ .
\end{aligned} \tag{B5}$$

The second line results from the definition (B2) and from the property (4.34), and the third line from integration by parts. The Coulomb potential $\phi(\mathbf{r})=\phi_s(\mathbf{r})+\phi_l(\mathbf{r})$ has been decomposed into a long-range $\phi_l(\mathbf{r})$ regularized at the origin and a short-range part $\phi_s(\mathbf{r})$. In (B5), only the long-range part has been considered (contributions due to the short-range part have a fast decay; see Appendix C). Then the function $(\nabla^2)^2\phi_l(\mathbf{r})$ has a fast decay as well as $\rho_T(\mathbf{r}_1,\mathbf{r}_2)=\rho(\mathbf{r}_1,\mathbf{r}_2)-\rho^2$. This implies that (B5) is rapidly decreasing as $|\mathbf{r}_1-\mathbf{r}_2|\to\infty$.

The term (B4b) is similar. The fully truncated correlation $\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)[Q(\mathbf{r}_3)D^{\mu\nu}(\mathbf{r}_4)]_{\mathrm{nc}}\rangle_T$ is rapidly decreasing as $|\mathbf{r}_1-\mathbf{r}_2|\to\infty$. The partitions [2,2] give the following contribution to (B4b):

$$\begin{aligned}
&2e^2\sum_{\mu,\nu=1}^{3}\int d\mathbf{r}_3\int d\mathbf{r}_4\phi_l^{\mu\nu}(\mathbf{r}_3-\mathbf{r}_4)\langle Q(\mathbf{r}_1)Q(\mathbf{r}_3)\rangle_T\langle Q(\mathbf{r}_2)D^{\mu\nu}(\mathbf{r}_4)\rangle_T\\
&\quad=2e^2\sum_{\mu,\nu=1}^{3}\int d\mathbf{r}_3\int d\mathbf{r}_4\phi_l^{\mu\nu}(\mathbf{r}_3-\mathbf{r}_4)S(\mathbf{r}_1-\mathbf{r}_3)\nabla_4^{\mu}\nabla_4^{\nu}\rho_T(\mathbf{r}_2,\mathbf{r}_4)=2e^2\int d\mathbf{r}_3\int d\mathbf{r}_4[(\nabla^2)^2\phi_l](\mathbf{r}_3-\mathbf{r}_4)S(\mathbf{r}_1-\mathbf{r}_3)\rho_T(\mathbf{r}_2,\mathbf{r}_4)\ ,
\end{aligned} \tag{B6}$$

which is short range for the same reasons as for (B5).

APPENDIX C: CONTRIBUTIONS FROM THE SHORT-RANGE PART OF THE POTENTIAL

In any expression, the short-range singular part at $\mathbf{r}_i=\mathbf{r}_j$ of $\phi_s^{\mu\nu}(\mathbf{r}_i-\mathbf{r}_j)$ or $f_s(\mathbf{r}_i-\mathbf{r}_j)$ occurs always in conjunction with a correlation function which vanishes at $\mathbf{r}_i=\mathbf{r}_j$, so that integrals are locally convergent. When examining the asymptotic behavior of quantities involving $\phi_s^{\mu\nu}(\mathbf{r}_i-\mathbf{r}_j)$ or $f_s(\mathbf{r}_i-\mathbf{r}_j)$ by truncating the correlation functions, it suffices to omit the truncations involving the arguments $\mathbf{r}_i-\mathbf{r}_j$ occurring in the potential. Then, the integrals over these variables are also convergent at infinity because of the short range of the potential itself. As an example, we treat the singular short-range function $f_s(\mathbf{r})$ in (4.44a).

For this, we truncate (4.45) without splitting the product $P(\mathbf{r}_3,\mathbf{r}_4)=[N(\mathbf{r}_3)N(\mathbf{r}_4)]_{\rm nc}$ and obtain the contribution

$$\int d\mathbf{r}_3\int d\mathbf{r}_4 f_s(\mathbf{r}_3)\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)P(\mathbf{r}_3+\mathbf{r}_4,\mathbf{r}_4)\rangle_T \; . \tag{C1}$$

The function $\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)P(\mathbf{r}_3+\mathbf{r}_4,\mathbf{r}_4)\rangle_T$ vanishes at $\mathbf{r}_3=0$ and tends to zero rapidly as the points $\mathbf{r}_1$, $\mathbf{r}_2$, and $\mathbf{r}_4$ are separated. Thus the integral (C1) is convergent and has a fast decay as $(\mathbf{r}_1-\mathbf{r}_2)\to\infty$. Similar arguments apply to all contributions due to the short-range part of the potential which have been left over in Appendixes B and D.

APPENDIX D: STUDY OF THE TERM (4.44b)

Let us show first that the integrals are well defined locally. We decompose $\phi(\mathbf{r})=\phi_s(\mathbf{r})+\phi_l(\mathbf{r})$ with $\phi_l(\mathbf{r})$ regular at the origin and $\phi_s(\mathbf{r})$ integrable, and note that the short-range part does not contribute to

$$\rho\nabla^\mu\nabla^\nu\int d\mathbf{r}'\phi_s(\mathbf{r}-\mathbf{r}')=0 \; . \tag{D1}$$

This implies, for instance, that

$$\int d\mathbf{r}_4\phi_s^{\mu\nu}(\mathbf{r}_3-\mathbf{r}_4)\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)[N(\mathbf{r}_3)|Q(\mathbf{r}_4)Q(\mathbf{r}_5)]_{\rm nc}\rangle=\int d\mathbf{r}_4\phi_s^{\mu\nu}(\mathbf{r}_3-\mathbf{r}_4)\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)[N(\mathbf{r}_3)|N(\mathbf{r}_4)Q(\mathbf{r}_5)]_{\rm nc}\rangle \; . \tag{D2}$$

This integral is well defined at $\mathbf{r}_4=\mathbf{r}_3$ since the distribution function in (D2) vanishes there. The same argument applies to all short-range parts $\phi_s^{\mu\nu}(\mathbf{r})$ occurring in (4.44b). Integrability at infinity will be ensured by the cluster properties. From now on, we consider only the long-range part of the dipole potential.

We decompose (4.46) into fully truncated correlations with the abbreviated notation $\langle Q(\mathbf{r}_1)Q(\mathbf{r}_2)[N(\mathbf{r}_3)|Q(\mathbf{r}_4)Q(\mathbf{r}_5)]_{\rm nc}\rangle=\langle 12345\rangle$. Since $\langle m\rangle=\langle Q(\mathbf{r}_m)\rangle=0$ for $m=1,2,4,5$ and $\langle 3\rangle=\langle N(\mathbf{r}_3)\rangle=\rho$, only the partitions [5],[1,4],[2,3],[1,2,2] contribute:

$$K_2(12345)=\langle 12345\rangle_T+\langle 3\rangle\langle 1245\rangle_T \tag{D3a}$$

$$+\langle 13\rangle_T\langle 245\rangle_T+\langle 23\rangle_T\langle 145\rangle_T \tag{D3b}$$

$$+\langle 14\rangle_T\langle 235\rangle_T+\langle 24\rangle_T\langle 135\rangle_T+\langle 15\rangle_T\langle 234\rangle_T+\langle 25\rangle_T\langle 134\rangle_T \tag{D3c}$$

$$+\langle 34\rangle_T\langle 125\rangle_T+\langle 35\rangle_T\langle 124\rangle_T \tag{D3d}$$

$$+\langle 45\rangle_T\langle 123\rangle_T \tag{D3e}$$

$$+\langle 3\rangle\langle 14\rangle_T\langle 25\rangle_T+\langle 3\rangle\langle 15\rangle_T\langle 24\rangle_T \; . \tag{D3f}$$

In (D3a), the arguments $\mathbf{r}_1$ and $\mathbf{r}_2$ occur in fully truncated functions, thus these terms have a fast decay [note that the integral on $\mathbf{r}_3$ in the second term (D3a) is convergent since $\phi_l^{\mu\nu}(\mathbf{r}_3-\mathbf{r}_4)\phi_l^{\mu\nu}(\mathbf{r}_3-\mathbf{r}_5)\sim 1/|\mathbf{r}_3|^6$, $|\mathbf{r}_3|\to\infty$].

Using translation invariance the contribution of the terms (D3b) to (4.44b) is of the form (up to a constant factor, and with $\mathbf{r}_{12}=\mathbf{r}_1-\mathbf{r}_2$)

$$\sum_{\mu,\nu=1}^{3}\int d\mathbf{r}_3 S(\mathbf{r}_{12}-\mathbf{r}_3)\left[\int d\mathbf{r}_4\int d\mathbf{r}_5\phi_l^{\mu\nu}(\mathbf{r}_3-\mathbf{r}_4)\phi_l^{\mu\nu}(\mathbf{r}_3-\mathbf{r}_5)\langle Q(0)Q(\mathbf{r}_4)Q(\mathbf{r}_5)\rangle_T\right] . \tag{D4}$$

Since the charge clouds labeled by $\mathbf{r}_4$ and $\mathbf{r}_5$ in $\langle Q(0)Q(\mathbf{r}_4)Q(\mathbf{r}_5)\rangle_T$ have no multipoles [the classical multipole sum rules (3.2)], the expression in large parentheses in (D4) decreases faster that any inverse power of $\mathbf{r}_3$. Therefore the expression (D4), which is the convolution of two functions with rapid decrease, has also a fast decay as $|\mathbf{r}_{12}|\to\infty$.

The contributions of the terms (D3c) are of the form

$$\sum_{\mu,\nu=1}^{3}\int d\mathbf{r}_3\left[\int d\mathbf{r}_4\phi_l^{\mu\nu}(\mathbf{r}_{12}-\mathbf{r}_3-\mathbf{r}_4)S(\mathbf{r}_4)\right]\left[\int d\mathbf{r}_5\phi_l^{\mu\nu}(\mathbf{r}_3-\mathbf{r}_5)\langle Q(\mathrm{O})[N(\mathbf{r}_3)N(\mathbf{r}_5)]_{\rm nc}\rangle_T\right] . \tag{D5}$$

Both expressions in large parentheses have a fast decay as $|\mathbf{r}_3|\to\infty$, the first one because $S(\mathbf{r})$ has no multipoles and the second one because $\mathbf{r}_3$ occurs as the argument of a fully truncated function. Thus the convolution (D5) is rapidly decreasing as $|\mathbf{r}_{12}|\to\infty$.

The contribution of the terms (D3d) can be written as

$$\sum_{\mu,\nu=1}^{3} \int d\mathbf{r}_4 \phi_l^{\mu\nu}(\mathbf{r}_4) S(\mathbf{r}_4) \int d\mathbf{r}_3 \int d\mathbf{r}_5 \phi_l^{\mu\nu}(\mathbf{r}_3-\mathbf{r}_5) \langle Q(\mathbf{r}_1) Q(\mathbf{r}_2) Q(\mathbf{r}_5)\rangle_T$$

$$= \left[\frac{1}{3}\int d\mathbf{r}_4 (\nabla^2\phi_l)(\mathbf{r}_4) S(\mathbf{r}_4)\right]\left[\int d\mathbf{r}_3 \nabla^2 \phi_l(\mathbf{r}_3)\right]\int d\mathbf{r}_5 \langle Q(\mathbf{r}_1) Q(\mathbf{r}_2) Q(\mathbf{r}_5)\rangle_T \quad \text{(D6)}$$

since $\int d\mathbf{r} \nabla^\mu \nabla^\nu \phi_l(\mathbf{r}) S(\mathbf{r}) = \delta_{\mu\nu} \frac{1}{3} \int d\mathbf{r} (\nabla^2 \phi_l)(\mathbf{r}) S(\mathbf{r})$ by spherical symmetry. The function $(\nabla^2\phi_l)(\mathbf{r})$ is integrable and the contribution (D6) vanishes because of the charge sum rule.

The term (D3e) can be written as

$$\sum_{\mu,\nu=1}^{3} \left[\int d\mathbf{r}_4 \phi_l^{\mu\nu}(\mathbf{r}_4) \int d\mathbf{r}_5 \phi_l^{\mu\nu}(\mathbf{r}_4-\mathbf{r}_5) S(\mathbf{r}_5)\right] \int d\mathbf{r}_3 \langle Q(\mathbf{r}_1) Q(\mathbf{r}_2) Q(\mathbf{r}_3)\rangle_T \ . \quad \text{(D7)}$$

The integrals in the large parentheses are convergent and (D7) vanishes again by the charge sum rule.

The term (D3f),

$$2\rho \sum_{\mu,\nu=1}^{3} \int d\mathbf{r}_3 \left[\int d\mathbf{r}_4 \phi_l^{\mu\nu}(\mathbf{r}_{12}-\mathbf{r}_3-\mathbf{r}_4) S(\mathbf{r}_4)\right]\left[\int d\mathbf{r}_5 \phi_l^{\mu\nu}(\mathbf{r}_3-\mathbf{r}_5) S(\mathbf{r}_5)\right] , \quad \text{(D8)}$$

being the convolution of two rapidly decreasing function [$S(\mathbf{r})$ has no multipoles], has a fast decay.

APPENDIX E: CALCULATION OF THE NUMBER A IN (4.56)

One writes the multipole expansion of the dipole potential as $|\mathbf{r}| \to \infty$

$$\phi^{\mu_1\mu_2}(\mathbf{r}-\mathbf{a}) = \nabla^{\mu_1}\nabla^{\mu_2}\frac{1}{|\mathbf{r}-\mathbf{a}|} = \sum_{n\,(\geq 0)} (-1)^n \gamma_{\mu_1\mu_2\nu_1\cdots\nu_n} \frac{a^{\nu_1}\cdots a^{\nu_n}}{|\mathbf{r}|^{3+n}} \quad \text{(E1)}$$

(summation on repeated indices) with the completely symmetric tensors

$$\gamma_{\mu_1\mu_2\nu_1\cdots\nu_n} = \frac{1}{n!}\nabla^{\mu_1}\nabla^{\mu_2}\nabla^{\nu_1}\cdots\nabla^{\nu_n}\frac{1}{|\mathbf{r}|}\bigg|_{|\mathbf{r}|=1} . \quad \text{(E2)}$$

Notice that the harmonicity of the Coulomb potential implies

$$\sum_{\mu=1}^{3} \gamma_{\mu\mu\nu_1\cdots\nu_n} = 0 \ . \quad \text{(E3)}$$

Introducing (E1) into $f(\mathbf{r}-\mathbf{a}) = \sum_{\mu,\nu=1}^{3} [\phi^{\mu\nu}(\mathbf{r}-\mathbf{a})]^2$ one finds that the coefficients of the terms of order $|\mathbf{r}|^{-10}$ in the development (4.52) are expressed by

$$\Gamma_{\mu_1\mu_2\mu_3\mu_4} = \sum_{\nu_1,\nu_2} (\gamma_{\nu_1\nu_2\mu_1\mu_2}\gamma_{\nu_1\nu_2\mu_3\mu_4} + 2\gamma_{\nu_1\nu_2\mu_1}\gamma_{\nu_1\nu_2\mu_2\mu_3\mu_4} + 2\gamma_{\nu_1\nu_2}\gamma_{\nu_1\nu_2\mu_1\mu_2\mu_3\mu_4}) \ . \quad \text{(E4)}$$

Taking (E3) into account, the number A (4.56) is therefore given by

$$A = 2\sum_{\mu_1,\mu_2,\nu_1,\nu_2=1}^{3} (\gamma_{\mu_1\mu_2\nu_1\nu_2})^2 \ . \quad \text{(E5)}$$

One calculates $\gamma_{\mu_1\mu_2\nu_1\nu_2}$ from (E2)

$$\gamma_{\mu_1\mu_2\nu_1\nu_2} = \tfrac{105}{2}\hat{\mathbf{r}}_{\mu_1}\hat{\mathbf{r}}_{\mu_2}\hat{\mathbf{r}}_{\nu_1}\hat{\mathbf{r}}_{\nu_2} - \tfrac{15}{2}(\hat{\mathbf{r}}_{\mu_1}\hat{\mathbf{r}}_{\mu_2}\delta_{\nu_1\nu_2} + \hat{\mathbf{r}}_{\mu_1}\hat{\mathbf{r}}_{\nu_1}\delta_{\mu_2\nu_2} + \hat{\mathbf{r}}_{\mu_1}\hat{\mathbf{r}}_{\nu_2}\delta_{\mu_2\nu_1} + \hat{\mathbf{r}}_{\mu_2}\hat{\mathbf{r}}_{\nu_1}\delta_{\mu_1\nu_2} + \hat{\mathbf{r}}_{\mu_2}\hat{\mathbf{r}}_{\nu_2}\delta_{\mu_1\nu_1} + \hat{\mathbf{r}}_{\nu_1}\hat{\mathbf{r}}_{\nu_2}\delta_{\mu_1\mu_2})$$
$$+ \tfrac{3}{2}(\delta_{\mu_1\mu_2}\delta_{\nu_1\nu_2} + \delta_{\mu_1\nu_1}\delta_{\mu_2\nu_2} + \delta_{\mu_1\nu_2}\delta_{\mu_2\nu_1}) , \quad \text{(E6)}$$

where $\hat{\mathbf{r}} = \mathbf{r}/|\mathbf{r}|$ is a unit vector, and one finds $A = 1260$.

APPENDIX F: DERIVATION OF THE ASYMPTOTIC BEHAVIOR (6.33)

In this appendix, we study the asymptotic behavior of the convolution integral

$$\int d\mathbf{X}' d\mathbf{X}'' \phi(\mathbf{Y}'-\mathbf{X}') \langle Q(\mathbf{X}') Q(\mathbf{X}'')\rangle \phi(\mathbf{Y}''-\mathbf{X}'') \quad \text{(F1)}$$

when $|\mathbf{Y}'-\mathbf{Y}''| \to \infty$. Introducing the electrostatic potential

$$V_{\text{eff}}(\mathbf{Y}''|\mathbf{X}') = \int d\mathbf{X}'' \phi(\mathbf{X}''-\mathbf{Y}'') \langle Q(\mathbf{X}') Q(\mathbf{X}'')\rangle \quad \text{(F2)}$$

created at $\mathbf{Y}''$ by the charge distribution $\langle Q(\mathbf{X}') Q(\mathbf{X}'')\rangle$ centered at $\mathbf{X}'$, we can rewrite (F1) as

$$\int d\mathbf{X}' \phi(\mathbf{Y}'-\mathbf{X}') V_{\text{eff}}(\mathbf{Y}''|\mathbf{X}') \ . \quad \text{(F3)}$$

Since $\langle Q(\mathbf{X}')Q(\mathbf{X}'')\rangle$ decays fast when $|\mathbf{X}'-\mathbf{X}''| \to\infty$ and does not carry any multipole, $V_{\text{eff}}(\mathbf{Y}''|\mathbf{X}')$ decays fast when $|\mathbf{Y}''-\mathbf{X}'|\to\infty$. Therefore the asymptotic behavior of (F3) when $|\mathbf{Y}'-\mathbf{Y}''|\to\infty$ can be obtained through the expansion of $\phi(\mathbf{Y}'-\mathbf{X}')$ around $\phi(\mathbf{Y}'-\mathbf{Y}'')$, apart from exponentially decaying terms. Taking into account that $V_{\text{eff}}(\mathbf{Y}''|\mathbf{X}')$ is spherically symmetric, i.e., only depends on $|\mathbf{Y}''-\mathbf{X}'|$, we see that the resulting multipolar expansion reduces to the monopole term

$$\phi(\mathbf{Y}'-\mathbf{Y}'')\int d\mathbf{X}'V_{\text{eff}}(\mathbf{Y}''|\mathbf{X}') \; . \tag{F4}$$

Using the classical Carnie and Chan sum rule[36] which states

$$\int d\mathbf{X}'V_{\text{eff}}(\mathbf{Y}''|\mathbf{X}')=\beta^{-1} \tag{F5}$$

we finally find that (F1) behaves as $\beta^{-1}\phi(\mathbf{Y}'-\mathbf{Y}'')$ when $|\mathbf{Y}'-\mathbf{Y}''|\to\infty$, apart from exponentially decaying terms. This leads to the asymptotic behavior (6.33) because the expression (6.32) is merely given by the application of the operator $D^{n'}_{\mathbf{Y}'}D^{n''}_{\mathbf{Y}''}$ to (F1).

APPENDIX G: DERIVATION OF EXPRESSIONS (7.19) AND (7.20)

In this appendix, we derive the expressions (7.19) and (7.20) in the Debye-Hückel approximation. The derivation is standard except for the fact that the energy is temperature dependent through the densities (7.5) and the de Broglie thermal lengths λ_i. We consider first the electrostatic energy of the classical gas in the presence of a single external charge density $n_1(\mathbf{r})$ (7.5). We obtain the excess free energy $F(\xi_i)$ (7.19) from the formula

$$\beta F=\int_0^{\beta}d\beta'\left[\left\langle\frac{\partial}{\partial\beta'}(\beta' U)\right\rangle-\langle U_0\rangle_0\right] , \tag{G1}$$

where $\langle\ \rangle$ and $\langle\ \rangle_0$ denote, respectively, the thermal averages with respect to the perturbed and unperturbed energies U and U_0. In the Debye-Hückel approximation, the correlations are neglected,

$$\left\langle\frac{\partial}{\partial\beta}(\beta U)\right\rangle^{\text{DH}}-\langle U_0\rangle_0^{\text{DH}}=\tfrac{1}{2}\int d\mathbf{r}\int d\mathbf{r}'\phi(\mathbf{r}-\mathbf{r}')\langle Q(\mathbf{r})\rangle^{\text{DH}}\langle Q(\mathbf{r}')\rangle^{\text{DH}}+e_1\int d\mathbf{r}\int d\mathbf{r}'\frac{\partial}{\partial\beta}[\beta n_1(\mathbf{r})]\phi(\mathbf{r}-\mathbf{r}')\langle Q(\mathbf{r}')\rangle^{\text{DH}} , \tag{G2}$$

and the induced charge density $\langle Q(\mathbf{r})\rangle^{\text{DH}}$ is calculated from the linearized Poisson-Boltzmann equations with external source $e_1n_1(\mathbf{r})$ [notice that $\langle Q(\mathbf{r})\rangle_0=0$ in the uniform system] with the standard result

$$\langle Q(\mathbf{r})\rangle^{\text{DH}}=-\frac{\varkappa^2}{4\pi}\int d\mathbf{r}'\frac{e^{-\varkappa|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|}e_1n_1(\mathbf{r}') , \tag{G3}$$

$$\varkappa^2=4\pi e^2\rho\beta \; . \tag{G4}$$

$\varkappa$ is the inverse Debye length. The quantity (G2) is easily calculated with the help of Fourier transforms

$$\begin{aligned}\left\langle\frac{\partial}{\partial\beta}(\beta U)\right\rangle^{\text{DH}}-\langle U_0\rangle_0^{\text{DH}}=&\tfrac{1}{2}\int d\mathbf{k}\langle Q(\mathbf{k})\rangle^{\text{DH}}\frac{4\pi}{|\mathbf{k}|^2}\langle Q(-\mathbf{k})\rangle^{\text{DH}}\\&+\frac{e_1}{2}\int d\mathbf{k}\left[\frac{\partial}{\partial\beta}[\beta n_1(\mathbf{k})]\frac{4\pi}{|\mathbf{k}|^2}\langle Q(-\mathbf{k})\rangle^{\text{DH}}+\frac{\partial}{\partial\beta}[\beta n_1(-\mathbf{k})]\frac{4\pi}{|\mathbf{k}|^2}\langle Q(\mathbf{k})\rangle^{\text{DH}}\right]\end{aligned} \tag{G5}$$

and

$$\langle Q(\mathbf{k})\rangle^{\text{DH}}=-\frac{\varkappa^2}{|\mathbf{k}|^2+\varkappa^2}e_1n_1(\mathbf{k}) \; . \tag{G6}$$

Substituting (G6) into (G5), and noting the relations

$$\beta\frac{\partial}{\partial\beta}\varkappa^2=\varkappa^2,\quad \frac{\partial}{\partial\beta}\left[\frac{\beta\varkappa^2}{|\mathbf{k}|^2+\varkappa^2}\right]=\frac{2\varkappa^2}{|\mathbf{k}|^2+\varkappa^2}-\left[\frac{\varkappa^2}{|\mathbf{k}|^2+\varkappa^2}\right]^2 , \tag{G7}$$

one finds after some algebra

$$\left\langle\frac{\partial}{\partial\beta}(\beta U)\right\rangle^{\text{DH}}-\langle U_0\rangle_0^{\text{DH}}=\frac{e_1^2}{2}\frac{\partial}{\partial\beta}\left[\beta\int d\mathbf{k}\,n_1(\mathbf{k})n_1(-\mathbf{k})\left[\frac{4\pi}{|\mathbf{k}|^2+\varkappa^2}-\frac{4\pi}{|k|^2}\right]\right] . \tag{G8}$$

Hence it follows from (G1) that

$$F=\frac{e_1^2}{2}\int d\mathbf{k}\, n_1(\mathbf{k})n_1(-\mathbf{k})\left[\frac{4\pi}{|\mathbf{k}|^2+\varkappa^2}-\frac{4\pi}{|\mathbf{k}|^2}\right]$$
$$=\frac{e_1^2}{2}\int d\mathbf{r}\int d\mathbf{r}'\frac{(e^{-\varkappa|\mathbf{r}-\mathbf{r}'|}-1)}{|\mathbf{r}-\mathbf{r}'|}n_1(\mathbf{r})n_1(\mathbf{r}') . \tag{G9}$$

When the expression (7.5) of $n_1(\mathbf{r})$ is used, (G9) is identical to (7.19). The calculation of the effective potential ϕ_{eff} (7.15) proceeds along the same line. One evaluates (G1) in the Debye-Hückel approximation in the presence of the external distribution $n_1(\mathbf{r})+n_2(\mathbf{r})$ due to two filaments, and this leads to (7.20).

APPENDIX H: DECAY RATE OF $S_{\text{RPA}}(r)$

In this appendix, we show that the quantum charge-charge correlation of the OCP computed in the framework of the RPA, $S_{\text{RPA}}(r)$, decays faster than any inverse power at large distances r, at finite temperature.

In the RPA, one first computes the wave-number- and frequency-dependent dielectric constant $\epsilon_{\text{RPA}}(k,\omega)$, as[4]

$$\epsilon_{\text{RPA}}(k,\omega)=1-\frac{4\pi e^2}{\hbar k^2}\frac{1}{(2\pi)^3}\int dq\frac{[f_{\text{FD}}(\mathbf{q})-f_{\text{FD}}(\mathbf{q}+\mathbf{k})]}{\left[\omega-\left[\frac{\hbar\mathbf{q}\cdot\mathbf{k}}{m}+\frac{\hbar k^2}{2m}\right]+i\delta\right]} , \tag{H1}$$

where $f_{\text{FD}}(\mathbf{q})$ is the Fermi-Dirac distribution (z is the fugacity)

$$f_{\text{FD}}(\mathbf{q})=\frac{1}{[z^{-1}\exp(\beta\hbar^2q^2/2m)+1]} . \tag{H2}$$

The expression (H1) must be understood as a limit when the small real positive number δ goes to zero. This limit is easily computed using the theory of analytical functions, with the result

$$\epsilon_{\text{RPA}}(k,\omega)=1+R(k,\omega)+iI(k,\omega) , \tag{H3}$$

where R and I are real functions given by

$$R(k,\omega)=\frac{4\pi me^2}{\hbar^2k^3}\left[P_R\left[\frac{m\omega}{\hbar k}-\frac{k}{2}\right]-P_R\left[\frac{m\omega}{\hbar k}+\frac{k}{2}\right]\right] \tag{H4}$$

and

$$I(k,\omega)=\frac{4\pi^2me^2}{\hbar^2k^3}\left[P_I\left[\frac{m\omega}{\hbar k}-\frac{k}{2}\right]-P_I\left[\frac{m\omega}{\hbar k}+\frac{k}{2}\right]\right] . \tag{H5}$$

In (H4) and (H5), the functions P_1 and P_R are defined by

$$P_I(\xi)=\frac{1}{(2\pi)^3}\int d\mathbf{q}_\perp f_{\text{FD}}(\xi\hat{\mathbf{k}}+\mathbf{q}_\perp) \tag{H6}$$

and

$$P_R(\xi)=\mathrm{P}\int_{-\infty}^{\infty}d\eta\frac{P_I(\xi+\eta)}{\eta} , \tag{H7}$$

respectively, with $\hat{\mathbf{k}}$ a given unit vector, $\mathbf{q}_\perp$ a two-dimensional vector orthogonal to $\hat{\mathbf{k}}$ and P the principal part.

The quantum version of the fluctuation-dissipation theorem links the dynamical structure factor $\tilde{S}(k,\omega)$ to the dielectric constant $\epsilon(k,\omega)$ through

$$\tilde{S}(k,\omega)=\frac{-\hbar k^2}{4\pi^2[1-\exp(-\beta\hbar\omega)]}\mathrm{Im}\frac{1}{\epsilon(k,\omega)} , \tag{H8}$$

where Im denotes the imaginary part. Replacing $\epsilon(k,\omega)$ by (H3) in (H8), one finds

$$\tilde{S}_{\text{RP}}(k,\omega)=\frac{\hbar k^2}{4\pi^2[1-\exp(-\beta\hbar\omega)]}\times\frac{I(k,\omega)}{\{[1+R(k,\omega)]^2+[I(k,\omega)]^2\}} . \tag{H9}$$

The corresponding expression of the static structure factor

$$\tilde{S}(k)=\int d\mathbf{r}\exp(i\mathbf{k}\cdot\mathbf{r})S(r) \tag{H10}$$

directly follows from

$$\tilde{S}(k)=\int_{-\infty}^{\infty}d\omega\,\tilde{S}(k,\omega) \tag{H11}$$

with the result

$$\tilde{S}_{\text{RPA}}(k)=\frac{\hbar k^2}{4\pi^2}\int_{-\infty}^{\infty}d\omega\frac{I(k,\omega)}{[1-\exp(-\beta\hbar\omega)]\{[1+R(k,\omega)]^2+[I(k,\omega)]^2\}} . \tag{H12}$$

For our purpose, it is sufficient to study the analytic properties of $\tilde{S}_{\text{RPA}}(k)$ on the real axis. It is immediately seen from (H4)–(H7) and (H12) that $\tilde{S}_{\text{RPA}}(k)$ is analytic at any $k>0$. At $k=0$, we have to investigate the structure of the small-k expansion of (H12). The various powers of k which appear in this expansion arise from the contributions in the integral $\int_{-\infty}^{\infty}d\omega\cdots$ of the following three regions: (i) ω small, (ii) ω close to $-\omega_p$, (iii) ω close to ω_p, where ω_p is the plasma frequency, $\omega_p=(4\pi e^2\rho/m)^{1/2}$. The contribution of the region (i) is easily obtained through the variable change $\omega=vk$ and the expansion of the integrant in (H12) in powers of k at fixed v. From the expansions

$$R(k,vk)=\frac{-4\pi me^2}{\hbar^2k^2}\sum_{n=0}^{\infty}\frac{k^{2n}}{2^{2n}(2n+1)!}\frac{d^{2n+1}P_R}{d\xi^{2n+1}}\left[\frac{mv}{\hbar}\right],$$

$$I(k,vk)=\frac{-4\pi me^2}{\hbar^2k^2}\sum_{n=0}^{\infty}\frac{k^{2n}}{2^{2n}(2n+1)!}\frac{d^{2n+1}P_I}{d\xi^{2n+1}}\left[\frac{mv}{\hbar}\right]$$

$$\tag{H13}$$

$$1-\exp(-\beta\hbar vk)=\beta\hbar vk\sum_{n=0}^{\infty}\frac{(-1)^n}{(n+1)!}(\beta\hbar vk)^n,$$

we infer the contribution of the region (i) takes the form

$$\frac{\hbar^2k^4}{16\pi^3m\beta e^2}\int_{-\infty}^{\infty}dv\left[\sum_{n=0}^{\infty}s_n(v)k^n\right]. \tag{H14}$$

Taking into account the parity relations, $P_R(-mv/\hbar)=-P_R(mv/\hbar)$ and $P_I(-mv/\hbar)=-P_I(mv/\hbar)$, we find that $s_n(v)$ is an odd function of v if n is odd, and an even function otherwise. Furthermore, for any n, $s_n(v)$ is integrable everywhere and decays like a Gaussian when $|v|\to\infty$ because so does $P_I(mv/\hbar)$ in this limit [$P_R(mv/\hbar)$ decays algebraically when $|v|\to\infty$].

Thus we can perform the integral $\int_{-\infty}^{\infty}dv$ in (H14) term to term. The resulting expression is an entire series in k^2, with a first term proportional to k^4. We turn now to the contributions of the regions (ii) and (iii). For small values of k, the integrant in (H12) has two plasmon peaks centered at $\omega=\pm\omega_m(k)$, with $\omega_m(k)$ close to ω_p and such that

$$1+R(k,\omega_m(k))=0\ . \tag{H15}$$

The height of these peaks diverges as $\exp[\beta m\omega_m^2(k)/2k^2]$ when $k\to 0$, because for ω finite and k small, $I(k,\omega)$ is Gaussianly small and behaves (apart from a multiplicative power of k) as $\exp(-\beta m\omega^2/2k^2)$. Furthermore, the width of these peaks is proportional to

$$|I(k,\omega_m(k))/(\partial R/\partial\omega)(k,\omega_m(k))|\ ,$$

which is also Gaussianly small and behaves as $\exp[-\beta m\omega_m^2(k)/2k^2]$. Therefore the contributions of the plasmon peaks to $\tilde{S}_{\mathrm{RPA}}(k)$ in the small-k limit, can be computed, apart from exponentially small terms, by replacing the integrant of (H12) by the Lorentzians in $\nu=\omega-[\pm\omega_m(k)]$

$$\frac{1}{\{1-\exp[\mp\beta\hbar\omega_m(k)]\}}\,\frac{I(k,\pm\omega_m(k))}{\left[[I(k,\pm\omega_m(k))]^2+\left[\dfrac{\partial R}{\partial\omega}(k,\pm\omega_m(k))\right]^2\nu^2\right]} \tag{H16}$$

for ω close to $\pm\omega_m(k)$, respectively. After a straightforward integration over ν running from $-\infty$ to ∞, these contributions become

$$\frac{\hbar k^2}{4\pi^2}\left[\frac{1}{\{1-\exp[-\beta\hbar\omega_m(k)]\}}\,\frac{\pi}{\dfrac{\partial R}{\partial\omega}(k,\omega_m(k))}-\frac{1}{\{1-\exp[\beta\hbar\omega_m(k)]\}}\,\frac{\pi}{\dfrac{\partial R}{\partial\omega}(k,\omega_m(k))}\right]$$
$$=\frac{\hbar k^2}{4\pi}\,\frac{1}{\dfrac{\partial R}{\partial\omega}(k,\omega_m(k))}\coth\left[\frac{\beta\hbar\omega_m(k)}{2}\right]. \tag{H17}$$

As can be checked from the definitions (H4), (H6), and (H7), the small-k expansion of $R(k,\omega)$ for ω fixed and finite reads

$$R(k,\omega)=-\frac{\omega_p^2}{\omega^2}\left[1+\sum_{n=1}^{\infty}r_n(\omega)k^{2n}\right] \tag{H18}$$

apart from exponentially small terms with k, where the functions $r_n(\omega)$ are polynomials in $1/\omega^2$; for deriving (H18) we have used the parity of $P_I(\xi)$ and the identity

$$\rho=\int_{-\infty}^{\infty}d\xi\,P_I(\xi)=\frac{1}{(2\pi)^3}\int d\mathbf{q}f_{\mathrm{FD}}(\mathbf{q})\ , \tag{H19}$$

which links the density to the fugacity for an ideal Fermi gas. Inserting (H18) in (H15), we find that

$$\omega_m(k)=\omega_p+\sum_{n=1}^{\infty}c_nk^{2n} \tag{H20}$$

plus exponentially small terms with k. Replacing $\omega_m(k)$ by (H20) in (H17), and taking into account (H18), we see that the contribution of the regions (ii) and (iii) to the small-k behavior of $\tilde{S}_{\mathrm{RPA}}(k)$ can be represented by an entire series in k^2, apart from exponentially small terms with k. Thus the small-k expansion of $\tilde{S}_{\mathrm{RPA}}(k)$ has a similar structure, and since $\tilde{S}_{\mathrm{RPA}}(k)$ is analytic at any $k>0$, we finally conclude that $S_{\mathrm{RPA}}(r)$ decays faster than any inverse power of the distance r when $r\to\infty$.

Note that at zero temperature, the Fermi-Dirac distribution becomes singular at the Fermi wave number. This induces a singularity in $\tilde{S}_{\mathrm{RPA}}(k)$ at this wave number, which leads to the well-known algebraic Friedel oscillations in $S_{\mathrm{RPA}}(r)$ at large distances. Furthermore, the k^2 term of the small-k expansion of $\tilde{S}_{\mathrm{RPA}}(k)$ is entirely determined by the contribution of the plasmon peaks. At this order, we can replace, in (H17), $\omega_m(k)$ by ω_p and $\partial R/\partial\omega(k,\omega_p)$ by $2/\omega_p$ according to the expressions (H20) and (H18). This gives

$$\tilde{S}_{\mathrm{RPA}}(k)\sim\frac{k^2}{4\pi}\frac{\hbar\omega_p}{2}\coth\left[\frac{\beta\hbar\omega_p}{2}\right],\quad k\to 0 \tag{H21}$$

which shows that the RPA preserves the sum rule (3.23).

[1]P. Debye and E. Hückel, Phys. Z. **9**, 185 (1923).
[2]N. F. Mott, Proc. Camb. Phil. Soc. **32**, 281 (1936); N. H. March, in *Theory of the Inhomogeneous Electron Gas,* edited by S. Lundqvist and N. H. March (Plenum, New York, 1983).
[3]D. Pines and Ph. Nozières, *The Theory of Quantum Liquids* (Benjamin, New York, 1986).
[4]D. Pines, Physica **26**, S103 (1960), and references quoted therein.
[5]D. Brydges and P. Federbush, Commun. Math. Phys. **73**, 197 (1980).
[6]J. Imbrie, Commun. Math. Phys. **87**, 515 (1983).
[7]W. Yang, J. Stat. Phys. **49**, 1 (1987).
[8]B. Jancovici, Phys. Rev. Lett. **46**, 386 (1981); A. Alastuey, in *Strongly Coupled Plasma Physics,* Vol. 154 of *NATO Advanced Study Institute, Series B: Physics,* edited by F. J. Rogers and H. E. DeWitt (Plenum, New York, 1986).
[9]M. Gaudin, J. Phys. (Paris) **46**, 1027 (1985); F. Cornu and B. Jancovici, J. Stat. Phys. **49**, 33 (1987); E. R. Smith, J. Stat. Phys. **50**, 813 (1988).
[10]J. L. Lebowitz and E. Lieb, Adv. Math. **9**, 316 (1972).
[11]J. Fröhlich and Y. M. Park, Commun. Math. Phys. **59**, 235 (1978); J. Fröhlich and Y. M. Park, J. Stat. Phys. **23**, 701 (1980).
[12]D. Brydges and P. Federbush, in *Rigorous Atomic and Molecular Physics,* Vol. 74 of *NATO Advanced Study Institute, Series B: Physics,* edited by G. Velo and A. S. Wightman (Plenum, New York, 1981).
[13]D. Brydges and E. Seiler, J. Stat. Phys. **42**, 405 (1986).
[14]L. Blum, Ch. Gruber, J. L. Lebowitz, and Ph. A. Martin, Phys. Rev. Lett. **48**, 1769 (1982); Ch. Gruber, J. L. Lebowitz, and Ph. A. Martin, J. Chem. Phys. **75**, 944 (1981).
[15]Ph. A. Martin and Ch. Gruber, Phys. Rev. A **30**, 512 (1984).
[16]A. Alastuey and Ph. A. Martin, J. Stat. Phys. **39**, 405 (1985).
[17]A. Alastuey and Ph. A. Martin, Europhys. Lett. **6**, 385 (1988).
[18]Ph. A. Martin, Rev. Mod. Phys. **60**, 1075 (1988).
[19]J. R. Fontaine and Ph. A. Martin, J. Stat. Phys. **36**, 163 (1984).
[20]Ph. A. Martin and Ch. Oguey, J. Phys. A **18**, 1995 (1985).
[21]Ph. A. Martin and Ch. Oguey, Phys. Rev. A **33**, 4191 (1986).
[22]B. Jancovici, Mol. Phys. **32**, 1177 (1976).
[23]B. Jancovici, J. Stat. Phys. **34**, 803 (1984); see also A. Alastuey, Ann. Phys. (Paris) **11**, 653 (1986).
[24]E. P. Wigner, Phys. Rev. **40**, 749 (1932); J. G. Kirkwood, *ibid.* **44**, 31 (1933); K. Imre, E. Özizmir, M. Rosenbaum, and P. F. Zweifel, J. Math. Phys. **8**, 1097 (1967); L. D. Landau and E. M. Lifshitz, *Statistical Physics,* Vol. 5 of *Course of Theoretical Physics* (Pergamon, London, 1959).
[25]A. Alastuey and B. Jancovici, Physica **97A**, 349 (1979).
[26]T. Kihara, Y. Midzuno, and T. Shizume, J. Phys. Soc. Jpn. **10**, 249 (1955).
[27]F. H. Stillinger and R. Lovett, J. Chem. Phys. **49**, 1991 (1968).
[28]A. Alastuey, J. Phys. (Paris) **49**, 1507 (1988); M. Brajon and P. Vieillefosse, J. Stat. Phys. (to be published).
[29]J. Ginibre, in *Statistical Mechanics and Quantum Field Theory,* 1971 Les Houches Lectures, edited by C. de Witt and R. Stora (Gordon and Breach, New York, 1971).
[30]B. Simon, *Functional Integration and Quantum Physics* (Academic, New York, 1979).
[31]J. S. Hoye and G. Stell, J. Chem. Phys. **68**, 4145 (1978).
[32]B. Jancovici, Physica **91A**, 152 (1978).
[33]H. Minoo, M. M. Gombert, and C. Deutsch, Phys. Rev. A **23**, 924 (1981), and references quoted therein.
[34]See, for instance, L. I. Schiff, *Quantum Mechanics* (McGraw-Hill, New York, 1955).
[35]S. Ichimaru, *Basic Principles of Plasma Physics* (Benjamin, Reading, MA, 1973).
[36]S. L. Carnie and D. Y. C. Chan, Chem. Phys. Lett. **77**, 437 (1981).