Journal of Statistical Physics, Vol. 41, Nos. 5/6, 1985

Time-Dependent Correlations in an Inhomogeneous One-Component Plasma

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Received July 8, 1985

Sum rules describing perfect screening at equilibrium in a classical plasma are extended to the time-displaced structure function of an inhomogeneous one-component plasma. We find that there are long-wavelength modes which oscillate undamped with a single frequency $\bar{\omega}$, $\bar{\omega}^2$ being an angular average of the squared plasma frequency at infinity. Our results are derived heuristically, allowing also for quantum effects, from linear response theory, and rigorously from the classical BBGKY hierarchy under some reasonable assumptions on the spatial decay of correlations. Special cases are investigated, in particular plasmas bounded by walls of varied shapes.

KEY WORDS: Sum rules; time-dependent correlations; one-component inhomogeneous plasma.

1. INTRODUCTION

Let $N(q)$ be the microscopic charge density at a point q of a Coulomb fluid (plasma, electrolyte,...), in equilibrium at the inverse temperature β. The static charge structure function is defined as

$$S(q_1 \mid q_2) = \langle N(q_1)\, N(q_2)\rangle_T = \langle N(q_1)\, N(q_2)\rangle - \langle N(q_1)\rangle\langle N(q_2)\rangle \quad (1.1)$$

where $\langle\ \rangle$ denotes an average over an equilibrium ensemble. In the homogeneous case, $\langle N(q)\rangle = 0$. The function S is known to obey a variety of sum rules, e.g., the Stillinger–Lovett rules[1]

$$\int dq\, S(q \mid 0) = 0 \quad (1.2)$$

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0022-4715/85/1200-0941$04.50/0

and

$$\frac{2\pi\beta}{3}\int dq\,|q|^2\,S(q\mid 0) = -1 \tag{1.3}$$

In terms of the Fourier transform

$$\tilde{S}(k) = \int dq\,\exp(-ik\cdot q)\,S(q\mid 0) \tag{1.4}$$

(1.2) and (1.3) are equivalent to

$$\frac{4\pi\beta\tilde{S}(k)}{|k|^2} \to 1, \qquad \text{as} \quad k \to 0 \tag{1.5}$$

These sum rules have been generalized to the case of an inhomogeneous Coulomb system by Carnie and Chan.[2,3] They take the form

$$\beta\int dq \int dq_1 \frac{1}{|q_1|}\,S(q_1\mid q) = 1 \tag{1.6}$$

The sum rules express the macroscopic property of *screening* which is possessed by Coulomb fluids. They can be derived by using linear response theory and assuming that an external charge introduced in the fluid induces a polarization charge which cancels the external one. More rigorous derivations using the BGY hierarchy can be given under suitable clustering assumptions.

In the present paper, we derive a dynamical and quantum mechanical generalization of (1.6) (and some other sum rules), for inhomogeneous *one-component* plasmas (OCP). The restriction to OCP's (i.e., to systems of identical particles of charge e and mass m in a fixed background of opposite charge) has to be made because we use a unique property of these systems: the long-wavelength plasma oscillations are not damped. From a microscopic point of view, this is related to the validity of a dipole sum rule [see (3.30)]. We can however deal with a large class of inhomogeneous OCP's: we allow the background density $\rho_b(q)$, a function of the position q, to have a value at infinity which may depend upon the direction Ω in which q recedes to infinity. More precisely, $\lim_{r\to\infty}\rho_b(r,\Omega) = \rho_\infty(\Omega)$ exists (for almost every Ω), with $q = (r, \Omega)$, $r = |q|$, Ω = angles of q. This allows in particular for OCP's bounded by walls of varied shapes.

Defining the usual dynamical structure factor $S(q_1, t\mid q)$ as the correlation function between the charge density at time t and point q_1 and the charge density at time 0 and point q, we find the proper dynamical and quantum-mechanical generalization of (1.6). It is Eq. (2.15), which reduces to (2.16) in the classical limit, and to (2.17) in the quantum mechanical static limit; the $\bar{\omega}^2$ appearing in these equations is defined by (2.12) as an angular average of the plasma frequency (squared) $\omega_p^2(\Omega)$ at infinity.

Note that a system of real atoms and molecules in which the nuclei are treated as *fixed* is included in our scheme, provided that they are confined to a bounded region of space. This region is then imagined surrounded by a more smeared out background $\rho_b(r, \Omega)$ which has the limit $\rho_\infty(\Omega)$ as $r \to \infty$.

In Section 2, we derive our generalization of the Carnie and Chan sum rule by using a linear response argument. We discuss in some detail a number of special cases. Through another linear response argument, we also obtain another family of sum rules involving the dipole moment of the charge structure factor. We also consider the effect of images forces created by a dielectric wall. The key ingredient is the assumption that macroscopic physics has to be valid on large length scales.

Section 3 reinvestigates the above-mentioned generalized sum rules, in the classical case, from a microscopic point of view, based upon the BBGKY hierarchy and reasonable spatial clustering assumptions. A preliminary account of this part has already been published.[4]

2. LINEAR RESPONSE APPROACH

2.1. Generalized Carnie and Chan Sum Rule

Let a system in equilibrium with a hamiltonian H_0 be subjected to a perturbation $A \cos \omega t$, where A is some observable. The linear response of some other observable B, i.e., the change of the average value of B, computed to first order in A, is of the form[5] $\mathrm{Re}[\chi_{BA}(\omega)\exp(-i\omega t)]$, which defines the response function $\chi_{BA}(\omega)$. We also consider, in the unperturbed system, the time dependent correlation function,

$$C_{AB}(t) = \langle A(t)\, B(0) \rangle_T \tag{2.1}$$

where $A(t)$ and $B(t)$ are the Heisenberg operators associated to A and B, and $\langle\ \rangle_T$ is a (truncated) canonical average for the nonperturbed system, and we define its Fourier transform

$$\tilde{C}_{AB}(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dt\, \exp(i\omega t)\, C_{AB}(t) \tag{2.2}$$

The response function and the correlation function are thus related by the fluctuation-dissipation theorem:

$$\tilde{C}_{AB}(\omega) = -\frac{\hbar}{\pi}\frac{1}{1-\exp(-\beta\hbar\omega)}\,\mathrm{Im}\,\chi_{BA}(\omega) \tag{2.3}$$

where we have assumed that A and B both have the same parity with respect to time reversal.

For our purposes here we choose H_0 to be the Hamiltonian of a OCP[6] and the perturbation as caused by an external oscillating charge $e_0 \cos \omega t$ located at the origin. Thus

$$A = e_0 \int dq_1 \frac{1}{|q_1|} N(q_1) \tag{2.4}$$

where $N(q_1)$ is the Schrödinger operator for the charge density at point q_1. Letting

$$B = B(q) = N(q) \tag{2.5}$$

we obtain from (2.2)

$$\tilde{C}_{AB(q)}(\omega) = \frac{e_0}{2\pi} \int_{-\infty}^{\infty} dt \exp(i\omega t) \int dq_1 \frac{1}{|q_1|} S(q_1, t \mid q) \tag{2.6}$$

where S is the time-dependent charge–charge correlation function

$$S(q_1, t \mid q) = \langle N(q_1, t)\, N(q, 0) \rangle_T \tag{2.7}$$

and $N(q_1, t)$ is the Heisenberg operator which corresponds to $N(q_1) = N(q_1, 0)$.

$\tilde{C}_{AB(q)}(\omega)$ in (2.6) is related through (2.3) to $\chi_{B(q)A}(\omega)$, the charge density at q. Now, the key point is that the integral

$$Q = \int dq\, \chi_{B(q)A}(\omega) \tag{2.8}$$

can be obtained by a macroscopic argument which goes as follows: $\mathrm{Re}[Q \exp(-i\omega t)]$ is the total net charge induced in the plasma by the external charge. In the static case ($\omega = 0$), one would have perfect screening and $Q + e_0$ would vanish. In the dynamical case ($\omega \neq 0$), $Q + e_0$ does not vanish because the plasma does not adjust itself instantaneously to the external charge. It is however still possible to compute Q, if we assume that the induced charge density decays fast enough at a large distance R from the origin for the electrical field to be asymptotically of the form $\mathrm{Re}[E \exp(-i\omega t)]$ with

$$E \sim \frac{e_0 + Q}{R^2} u \tag{2.9}$$

(u is the radial unit vector). When (2.9) holds and the background density $\rho_b(q)$ has a well-defined limit $\rho_\infty(\Omega)$ as q goes to infinity in the direction Ω, E will induce a radial current density $\mathrm{Re}[j \exp(-i\omega t)]$ with

$$-i\omega j \sim \frac{e^2 \rho_\infty(\Omega)}{m} E \tag{2.10}$$

Charge conservation then requires that

$$i\omega Q = R^2 \int j \cdot u \, d\Omega \tag{2.11}$$

Combining these equations, and introducing the averaged plasma frequency $\bar{\omega}$ defined by

$$\bar{\omega}^2 = \frac{e^2}{m} \int \rho_\infty(\Omega) \, d\Omega \tag{2.12}$$

we obtain

$$Q = \frac{\bar{\omega}^2}{\omega^2 - \bar{\omega}^2} e_0 \tag{2.13}$$

In Eq. (2.13) ω is to be understood as having an infinitesimal positive imaginary part which ensures that the perturbation is introduced adiabatically.[5] We find then from (2.8) and (2.13),

$$\frac{1}{e_0} \mathrm{Im} \int dq \, \chi_{B(q)A}(\omega) = -\frac{\pi\bar{\omega}}{2} [\delta(\omega - \bar{\omega}) - \delta(\omega + \bar{\omega})] \tag{2.14}$$

Using (2.6) and (2.14) in (2.3), and taking the Fourier transform with respect to ω, we obtain the final result

$$\int dq \int dq_1 \frac{1}{|q_1|} S(q_1, t \mid q) = \frac{\hbar\bar{\omega}}{2} \left[\frac{\exp(-i\bar{\omega}t)}{1 - \exp(-\beta\hbar\bar{\omega})} - \frac{\exp(i\bar{\omega}t)}{1 - \exp(\beta\hbar\bar{\omega})} \right] \tag{2.15}$$

This is the generalization of the Carnie and Chan sum rule to the dynamical quantum mechanical case, for OCP's. In the classical limit ($\hbar \to 0$), one finds

$$\beta \int dq \int dq_1 \frac{1}{|q_1|} S(q_1, t \mid q) = \cos \bar{\omega} t \tag{2.16}$$

In the static quantum mechanical limit ($t = 0$), one finds

$$\int dq \int dq_1 \frac{1}{|q_1|} S(q_1, 0 \mid q) = \frac{\hbar\bar{\omega}}{2} \coth \frac{\beta\hbar\bar{\omega}}{2} \tag{2.17}$$

An alternative, equivalent, derivation of (2.13) can be obtained by starting with the expression[6]

$$\varepsilon(\omega) = 1 - \frac{\omega_p^2}{\omega^2} \tag{2.18}$$

for the dielectric function $\varepsilon(\omega)$ of a homogeneous one-component plasma with density ρ_b, where ω_p is the plasma frequency:

$$\omega_p^2 = \frac{4\pi e^2 \rho_b}{m} \tag{2.19}$$

In the present inhomogeneous case, where ρ_b is a function of the position q, which has however a limit at infinity in each direction Ω, (2.18) and (2.19) still define a dielectric function at infinity $\varepsilon_\infty(\omega, \Omega)$. Hence, if an external charge $e_0 \cos \omega t$ is located at the origin, then, by Gauss's theorem, if we integrate E on a large sphere of radius R centered at the origin, we obtain

$$e_0 = \frac{R^2}{4\pi} \int \varepsilon_\infty(\omega, \Omega) E \cdot u \, d\Omega \tag{2.20}$$

where E must be of the form (2.9). This gives at once (2.13).

2.2. Special Cases

The general sum rule (2.15) can be applied to many special cases.

Uniform OCP. For a uniform plasma (constant background density ρ_b), $\bar{\omega}$ is the plasma frequency and (2.15), (2.17) reduce to a well-known long-wavelength sum rule[6-8]: the left-hand side of (2.15) becomes $\lim_{|k| \to 0} (4\pi/|k|^2)\, \tilde{S}(k, t)$, where

$$\tilde{S}(k, t) = \int dq \exp[ik \cdot (q - q_1)]\, S(q_1, t \mid q) \tag{2.21}$$

is the dynamical structure factor.

Semi-Infinite OCP. In the case of a semi-infinite plasma bounded by an impermeable plane wall,

$$\rho_b(q) = \begin{cases} 0, & x < 0 \\ \rho_b, & x > 0 \end{cases} \tag{2.22}$$

(the coordinate vector q of a point is now split into the component x normal to the wall and the component y parallel to the wall), the frequency

$$\bar{\omega} = \left(\frac{2\pi \rho_b e^2}{m} \right)^{1/2} = \omega_s \tag{2.23}$$

is the surface plasma frequency.[9] In terms of the two-dimensional partial Fourier transforms

$$\int dy \exp(-ik \cdot y) \frac{1}{(y^2 + x^2)^{1/2}} = \frac{2\pi}{|k|} \exp(-|k|\,|x|) \tag{2.24}$$

and

$$\tilde{S}(x_1, k, t \mid x) = \int dy \exp[ik \cdot (y - y_1)]\, S(x_1, y_1, t \mid x, y) \tag{2.25}$$

the left-hand side of (2.15) becomes

$$\lim_{|k| \to 0} \frac{2\pi}{|k|} \int_0^\infty dx \int_0^\infty dx_1 \exp(-|k|\, x_1)\, \tilde{S}(x_1, k, t \mid x)$$

Thus we recover a known sum-rule,[10] equivalent to Eq. (3.11) of Ref. 10. Through substraction of a bulk contribution, this sum rule gives information about the asymptotic form of $S(x_1, y_1, t \mid x, y)$ as $|y_1 - y| \to \infty$: for fixed x_1 and x, S decays like $|y_1 - y|^{-3}$. It is expected to decay much faster along any other Ω.

Two-Density OCP. For a two-density plasma,

$$\rho_b(q) = \begin{cases} \rho_+, & x > 0 \\ \rho_-, & x < 0 \end{cases} \tag{2.26}$$

(the plane $x = 0$ may be permeable or impermeable to the particles), the frequency $\bar{\omega}$ is

$$\bar{\omega} = \left[\frac{2\pi e^2}{m} (\rho_+ + \rho_-) \right]^{1/2} \tag{2.27}$$

the left-hand side of (2.15) becomes

$$\lim_{k \to 0} \frac{2\pi}{|k|} \int_{-\infty}^\infty dx \int_{-\infty}^\infty dx_1 \exp(-|k|\,|x_1|)\, \tilde{S}(x_1, k, t \mid x)$$

and we recover the sum rule (5.2) of Ref. 10. Again, through a substraction of bulk contributions this sum rule gives information about the asymptotic form of $S(x_1, y_1, t \mid x, y)$ as $|y_1 - y| \to \infty$.

Wedge. Let us now consider a plasma bounded by a wedge: the plasma is confined between two intersecting half-planes; the background density has a constant value ρ_b inside the wedge, 0 elsewhere. The z axis is the edge of the wedge, and the set of coordinates q can be chosen as cylindrical coordinates (z, r, θ). The frequency $\bar{\omega}$ is

$$\bar{\omega} = \left(\frac{2\alpha\rho_b e^2}{m}\right)^{1/2} \tag{2.28}$$

where α is the angle between the two half-planes; this is an edge plasmon[11] frequency. In terms of one-dimensional partial Fourier transforms

$$\int_{-\infty}^{\infty} dz \exp(ikz) \frac{1}{(z^2 + r^2)^{1/2}} = 2K_0(|k|\, r) \tag{2.29}$$

and

$$\tilde{S}(r_1, \theta_1, k, t \mid r, \theta) = \int_{-\infty}^{\infty} dz \exp[ik(z - z_1)]\, S(z_1, r_1, \theta_1, t \mid z, r, \theta) \tag{2.30}$$

the left-hand side of (2.15) becomes

$$\lim_{k \to 0} 2 \int_0^\alpha d\theta \int_0^\infty dr\, r \int_0^\alpha d\theta_1 \int_0^\infty dr_1\, r_1 K_0(|k|\, r_1)\, \tilde{S}(r_1, \theta_1, k, t \mid r, \theta)$$

Substracting from (2.15) surface and bulk contributions, as described in Appendix A, we can show that, as $|z - z_1| \to \infty$, $S(z_1, r_1, \theta_1, t \mid z, r, \theta)$ has the asymptotic form $f(r_1, \theta_1, t \mid r, \theta)/[|z - z_1| (\ln |z - z_1|)^2]$ obeying the sum rule

$$\begin{aligned}
&\int_0^\alpha d\theta \int_0^\infty dr\, r \int_0^\alpha d\theta_1 \int_0^\infty dr_1\, r_1 f(r_1, \theta_1, t \mid r, \theta) \\
&\quad = -\frac{\hbar\bar{\omega}}{8}\left[\frac{\exp(-i\bar{\omega}t)}{1 - \exp(-\beta\hbar\bar{\omega})} - \frac{\exp(i\bar{\omega}t)}{1 - \exp(\beta\hbar\bar{\omega})}\right] \\
&\quad\quad + \frac{\hbar\omega_s}{8}\left[\frac{\exp(-i\omega_s t)}{1 - \exp(-\beta\hbar\omega_s)} - \frac{\exp(i\omega_s t)}{1 - \exp(\beta\hbar\omega_s)}\right] \\
&\quad\quad + \frac{\alpha - \pi}{16\pi}\hbar\omega_p\left[\frac{\exp(-i\omega_p t)}{1 - \exp(-\beta\hbar\omega_p)} - \frac{\exp(i\omega_p t)}{1 - \exp(\beta\hbar\omega_p)}\right]
\end{aligned} \tag{2.31}$$

Cone. Obviously, (2.15) can be applied to a plasma confined in a cone, the apex of which is at the origin. The frequency is

$$\bar{\omega} = \left(\frac{\Omega \rho_b e^2}{m}\right)^{1/2} \tag{2.32}$$

where Ω is the solid angle of the cone.

Slab. For a plasma confined in a slab of thickness a, the averaged frequency $\bar{\omega}$ defined by (2.12) vanishes. If we again split q into x normal to the slab walls and y parallel to them, in terms of the two-dimensional Fourier transform (2.25), Eq. (2.15) becomes

$$\beta \lim_{|k| \to 0} \frac{2\pi}{|k|} \int_0^a dx \int_0^a dx_1\, \tilde{S}(x_1, k, t \mid x) = 1 \tag{2.33}$$

Equivalently, since the Fourier transform of $|k|$ is $-1/(2\pi\,|y|^3)$, we get the coordinate space asymptotic sum rule

$$\beta \int_0^a dx \int_0^a dx_1\, S(x_1, y_1, t \mid x, y)_{|y_1 - y| \to \infty} \sim -\frac{1}{4\pi^2\,|y_1 - y|^3} \tag{2.34}$$

It is remarkable that these sum rules have exactly the same form in the general dynamical and quantum mechanical case as in the static classical case: t and $\hbar$ do not appear in the right-hand side. This is so because a slab is essentially equivalent to a two-dimensional electron system, the collective oscillations of which have a frequency ω which behaves like $|k|^{1/2}$ for small wave numbers.[12] Therefore, in the small-k limit, these oscillations disappear.

In the case of a classical system, a more complete description of $\tilde{S}$ in the small-k limit has been given by Baus,[21] for a two-dimensional system of electrons. We believe that his results also hold for a slab of finite thickness, after $\tilde{S}(x_1, k, t \mid x)$ has been integrated upon x_1 and x. The results of Baus go beyond (2.33) and give information about terms of higher order in k.

Cylinder. Similar considerations apply for a plasma confined in a cylinder with a cross section S of arbitrary shape but finite area. Again, $\bar{\omega}$ vanishes. We now split the set of coordinates q into z parallel to the cylinder direction and r parallel to its cross section (in the present paragraph, r is a two-dimensional vector). In terms of the one-dimensional Fourier transform

$$\tilde{S}(r_1, k, t \mid r) = \int_{-\infty}^{\infty} dz \exp[ik(z - z_1)]\, S(r_1, z_1, t \mid r, z) \tag{2.35}$$

(2.15) becomes

$$-\beta \lim_{k \to 0} 2 \ln |k| \int_S dr \int_S dr_1\, \tilde{S}(r_1, k, t \mid r) = 1 \tag{2.36}$$

Equivalently, since the Fourier transform of $1/\ln |k|$ behaves asymptotically[13] like $1/[2\,|z|\,(\ln |z|)^2]$, we get the coordinate space sum rule that

$$\beta \int_S dr \int_S dr_1\, S(r_1, z_1, t \mid r, z)$$

has in its asymptotic form, as $|z_1 - z| \to \infty$, a term

$$-\frac{1}{4\,|z_1 - z|\,(\ln |z_1 - z|)^2}$$

(plus perhaps oscillating terms coming from possible singularities for finite real values of k).

Again these sum rules are the same as in the static classical case[13] owing to the fact that the frequency ω of the collective oscillations goes to zero with k (like $|k|\,|\ln |k||^{1/2}$).

2.3. Dipole Sum Rules

The sum rule (2.15) was obtained by analyzing the linear response to the radial electrical field of an external point charge. We now turn to another family of sum rules which can be obtained by analyzing the linear response to a homogeneous external electrical field; these sum rules involve the dipole moment of the pair correlation function.

For a uniform plasma, we would get once more the Stillinger–Lovett sum rule. Thus, we turn to other geometries.

Semi-Infinite OCP. We want to consider again the semi-infinite plasma with the background density (2.22). This semi-infinite geometry can be obtained as the limit of a slab geometry, in which the plasma is confined between two parallel walls at $x = 0$ and $x = L$; the limit $L \to \infty$ will be ultimately taken. We choose the perturbation as caused by charging the walls at $x = 0$ and L with oscillating uniform surface charge densities $\pm \alpha \cos \omega t$. Thus

$$A = -4\pi\alpha \int dy_1 \int_0^L dx_1\, x_1 N(x_1, y_1) \tag{2.37}$$

We look at the response of the charge density at $(x, y=0)$

$$B = B(x) = N(x, 0) \tag{2.38}$$

Now

$$\int_0^{L/2} dx\, \tilde{C}_{AB(x)}(\omega) = -4\pi\alpha \frac{1}{2\pi} \int_{-\infty}^{\infty} dt \exp(i\omega t) \int_0^{L/2} dx \int dy_1 \times \int_0^L dx_1\, x_1 S(x_1, y_1, t \mid x) \tag{2.39}$$

is related through (2.3) to

$$\sigma = \int_0^{L/2} dx\, \chi_{B(x)A}(\omega) \tag{2.40}$$

The quantity $\mathrm{Re}[\sigma \exp(-i\omega t)]$ is the surface charge density induced in the plasma along the wall at $x=0$ (as L becomes large, we actually expect this charge to be localized near the plane $x=0$, i.e., $\chi_{B(x)A}(\omega)$ will have a fast decay as x increases; thus the precise value of the upper integration limit in (2.39) is irrelevant. An opposite charge will be localized near the plane $x=L$). Therefore, σ can be obtained from a macroscopic argument, either by the same kind of reasoning as the one leading from (2.8) to (2.13), or, more rapidly, by using the dielectric function (2.18) and equating two alternative expressions for the electrical field in the bulk, after the limit $L \to \infty$ has been taken:

$$\mathrm{Re}[4\pi(\alpha + \sigma) \exp(-i\omega t)]\, u_x = \mathrm{Re}\left[\frac{4\pi\alpha}{\varepsilon(\omega)} \exp(-i\omega t)\right] u_x \tag{2.41}$$

(where u_x is the unit vector along the x axis). Thus we obtain at once

$$\sigma = \frac{\omega_p^2}{\omega^2 - \omega_p^2}\,\alpha \tag{2.42}$$

where ω_p is the bulk plasma frequency $(4\pi\rho_b e^2/m)^{1/2}$, and, from (2.3)

$$-4\pi \int_0^{\infty} dx \int dy_1 \int_0^{\infty} dx_1\, x_1 S(x_1, y_1, t \mid x) = \frac{\hbar\omega_p}{2}\left[\frac{\exp(-i\omega_p t)}{1 - \exp(-\beta\hbar\omega_p)} - \frac{\exp(i\omega_p t)}{1 - \exp(\beta\hbar\omega_p)}\right] \tag{2.43}$$

This is the generalization of the classical static dipole sum rule at a wall.[3,14]

OCP with Two Densities. We now consider the two-density plasma with the background density (2.26). Let

$$\omega_+ = \left(\frac{4\pi e^2 \rho_+}{m}\right)^{1/2}, \qquad \omega_- = \left(\frac{4\pi e^2 \rho_-}{m}\right)^{1/2} \tag{2.44}$$

be the plasma frequencies in the two regions. Choosing again the perturbation as caused by charging the plane $x=0$ with a surface charge density $\alpha \cos \omega t$ and the plane $x=L$ with the opposite charge, we can follow the same steps as in the previous paragraph, except that now σ must be defined as

$$\sigma = \int_{-L/2}^{L/2} dx\, \chi_{B(x)A}(\omega) \tag{2.45}$$

because there are induced charges on both sides of the wall at $x=0$. Thus we now obtain

$$\begin{aligned} -4\pi \int_{-\infty}^{\infty} dx \int dy_1 \int_0^{\infty} dx_1\, x_1 S(x_1, y_1, t \mid x) \\ = \frac{\hbar\omega_+}{2}\left[\frac{\exp(-i\omega_+ t)}{1-\exp(-\beta\hbar\omega_+)} - \frac{\exp(i\omega_+ t)}{1-\exp(\beta\hbar\omega_+)}\right] \end{aligned} \tag{2.46a}$$

and by a similar reasoning in the region $x<0$,

$$\begin{aligned} 4\pi \int_{-\infty}^{\infty} dx \int dy_1 \int_{-\infty}^{0} dx_1\, x_1 S(x_1, y_1, t \mid x) \\ = \frac{\hbar\omega_-}{2}\left[\frac{\exp(-i\omega_- t)}{1-\exp(-\beta\hbar\omega_-)} - \frac{\exp(i\omega_- t)}{1-\exp(\beta\hbar\omega_-)}\right] \end{aligned} \tag{2.46b}$$

In the classical static case, these dipole sum rules (2.46a) and (2.46b) reduce to

$$\begin{aligned} -4\pi\beta \int_{-\infty}^{\infty} dx \int dy_1 \int_0^{\infty} dx_1\, x_1 S(x_1, y_1 \mid x) \\ = 4\pi\beta \int_{-\infty}^{\infty} dx \int dy_1 \int_{-\infty}^{0} dx_1\, x_1 S(x_1, y_1 \mid x) = 1 \end{aligned} \tag{2.47}$$

We expect (2.47) to be valid also at the interface between two multicomponent plasmas. Note also that by combining (2.46a) and (2.46b) one obtains a sum rule for the total dipole moment

$$4\pi\int_{-\infty}^{\infty} dx \int dy_1 \int_{-\infty}^{\infty} dx_1\, x_1 S(x_1, y_1, t \mid x)$$
$$= \frac{\hbar\omega_-}{2}\left[\frac{\exp(-i\omega_- t)}{1-\exp(-\beta\hbar\omega_-)} - \frac{\exp(i\omega_- t)}{1-\exp(\beta\hbar\omega_-)}\right]$$
$$-\frac{\hbar\omega_+}{2}\left[\frac{\exp(-i\omega_+ t)}{1-\exp(-\beta\hbar\omega_+)} - \frac{\exp(i\omega_+ t)}{1-\exp(\beta\hbar\omega_+)}\right] \tag{2.48}$$

In the classical static case, the right-hand side of (2.48) vanishes, as it should since we expect the dipole moment $\int_{-\infty}^{\infty} dx_1\, x_1 S(x_1, y_1 \mid x)$ to vanish in a conducting medium, i.e., when the charges can adjust to give a good decay of correlations.[15]

The sum rules (2.43) and (2.46) can also be obtained by looking at the response to the radial electrical field created by a charged sphere of radius R; the limit $R \to \infty$ is taken at the end of the calculation. This approach, similar to the one which can be used in the classical static case,[14] is formally more correct for taking the thermodynamic limit.

2.4. Image Forces

Image effects can be incorporated in the above results about the semi-infinite plasma. We now assume that the semi-infinite plasma, which occupies the region $x > 0$, is bounded at $x = 0$ by a plane wall with a dielectric constant ε_W. We only consider the case when ε_W is finite.

The changes to be made in (2.15) are easily found. An external point charge e_0, located on the wall, at the origin, now creates a potential $2e_0/[(1+\varepsilon_W)\,|q_1|]$ rather than $e_0/|q_1|$. The frequency (2.23) has to be replaced by

$$\bar{\omega} = \left[\frac{4\pi\rho_b e^2}{(1+\varepsilon_W)m}\right]^{1/2} \tag{2.49}$$

and the sum rule (2.15) becomes

$$\int dq \int dq_1 \frac{1}{|q_1|} S(q_1, t \mid q)$$
$$= \lim_{k\to 0} \frac{2\pi}{|k|}\int_0^{\infty} dx \int_0^{\infty} dx_1\, e^{-|k|x_1}\tilde{S}(x_1, k, t \mid x)$$
$$= \frac{1+\varepsilon_W}{2}\frac{\hbar\bar{\omega}}{2}\left[\frac{\exp(-i\bar{\omega}t)}{1-\exp(-\beta\hbar\bar{\omega})} - \frac{\exp(i\bar{\omega}t)}{1-\exp(\beta\hbar\bar{\omega})}\right] \tag{2.50}$$

The argument leading to the sum rule (2.43) however is *not* sensitive to the value of ε_W, and (2.43) is not modified. Thus, image effects do not appear explicitly in the dipole sum rule[4] as already stated in Ref. 3 for the static classical case.

It may be noted that pertinent results for the case of an ideal conductor wall ($\varepsilon_W = \infty$) cannot be obtained by taking the limit $\varepsilon_W \to \infty$ in the above results. In that limit, the three expressions in (2.50) all diverge. As to (2.43), it is not valid in the case $\varepsilon_W = \infty$, because the limit $\varepsilon_W \to \infty$ and the integrations are operations which cannot be interchanged (this can be easily seen for the weak-coupling[(16)] explicit expression of $S(x_1, y_1, t=0 \mid x)$.

3. MICROSCOPIC THEORY

In this section we show that the dynamical sum rules previously derived from linear response and macroscopic screening arguments are exact consequences of the microscopic dynamics of the correlations. We treat the classical case: the quantum mechanical situation can be studied by analogous methods.

3.1. Time-Dependent Correlations

Let $u = (q, p)$ denote the position and momentum coordinates of a particle, and $U = (u_1, ..., u_n)$, $V = (v_1, ..., v_k)$ sets of particle coordinates. The time-dependent correlation functions involving n particles at time t and k particles at $t = 0$ are defined by

$$\rho(U, t \mid V) = \left\langle \left[\sum_{i_1 \neq \cdots \neq i_n} \delta(u_1 - \tilde{u}_{i_1}(t)) \cdots \delta(u_n - \tilde{u}_{i_n}(t)) \right] \times \left[\sum_{j_1 \neq \cdots \neq j_k} \delta(v_1 - \tilde{u}_{j_1}) \cdots \delta(v_k - \tilde{u}_{j_k}) \right] \right\rangle \tag{3.1}$$

$\tilde{u}_i(t) = (q_i(t), p_i(t))$ are coordinates of the ith particle at time t under the classical evolution, and $\langle \cdots \rangle$ is the thermal average on initial conditions $\tilde{u}_i(0) = \tilde{u}_i$.

From the stationarity of the equilibrium state, we have the symmetry relation

$$\rho(U, t \mid V) = \rho(V, -t \mid U) \tag{3.2}$$

[4] But image effects do appear explicitly in (2.50). Reference 3 might be misleading on this point.

When the set V is empty, the correlations reduce to their equilibrium (time-independent) values

$$\rho(u_1,\dots, u_n, t) = \rho(u_1,\dots, u_n) = \rho(q_1,\dots, q_n) \prod_{i=1}^{n} \left(\frac{\beta}{2\pi m}\right)^{3/2} \exp\left(-\beta \frac{p_i^2}{2m}\right) \tag{3.3}$$

where $\rho(q_1,\dots, q_n)$ are the usual equilibrium configurational distributions.

A quantity of particular interest is the position and momentum-dependent charge–charge correlation

$$S(u, t \mid v) = e^2[\rho(u, t \mid v) - \rho(u)\,\rho(v)] \tag{3.4}$$

from which we recover the configurational part (2.7) by integration over the momentum variables

$$S(q, t \mid q_1) = \int dp \int dp_1\, S(q, p, t \mid q_1, p_1) \tag{3.5}$$

(In the sequel, we simply suppress momentum arguments in the correlation functions when they have been integrated out).

More generally, we define the excess charge density at time t, when particles were fixed at $V = (q_1, p_1;\dots; q_k, p_k)$ at $t = 0$, by $e[\rho(q, t \mid V) - \rho(q)\,\rho(V)]$. This quantity is the time-dependent generalization of the static excess charge density introduced in Ref. 15

$$e[\rho(q, t=0 \mid V) - \rho(q)\,\rho(V)] = e\left[\rho(q, q_1,\dots, q_k) + \sum_{i=1}^{k} \delta(q - q_i)\,\rho(q_1,\dots, q_k) - \rho(q)\,\rho(q_1,\dots, q_k)\right] \times \prod_{i=1}^{k} \left(\frac{\beta}{2\pi m}\right)^{3/2} \exp\left(-\beta \frac{p_i^2}{2m}\right) \tag{3.6}$$

BBGKY Hierarchy. The BBGKY equations for the time-dependent correlations (3.1) of an inhomogeneous OCP with background density $\rho_b(q)$ have been discussed in Refs. 17 and 18. For $n = 1$ this has the form

$$\frac{\partial}{\partial t}\rho(u_1, t \mid V) = -\frac{p_1}{m}\cdot\nabla_{q_1}\rho(u_1, t \mid V) - eE(q_1)\cdot\nabla_{p_1}\rho(u_1, t \mid V) - e^2 \int dq_2\, F(q_1 - q_2)\cdot\nabla_{p_1}[\rho(u_1, q_2, t \mid V) - \rho(q_2)\,\rho(u_1, t \mid V)] \tag{3.7}$$

where $F(q) = -\nabla_q(1/|q|)$ is the Coulomb force and

$$E(q) = e \int dq_1 \, F(q - q_1)[\rho(q_1) - \rho_b(q_1)] \tag{3.8}$$

is the electric field due to the total charge density.

When the particles are constrained to move in a restricted domain D bounded by hard walls, the configurational integrals are restricted to D and we supplement equation (3.7) (valid inside D) by the condition of elastic collisions at the walls, i.e.,

$$\rho(q, p, t \mid V)\,|_{q \in \partial D} = \rho(q, \bar{p}, t \mid V)\,|_{q \in \partial D} \tag{3.9}$$

$\bar{p}$ is the momentum of an elastically reflected particle at q on the boundary ∂D of D, with incident momentum p.

We assume throughout this section that the time-dependent correlations (3.1) exist in the thermodynamic limit and satisfy the BBGKY equations for all times.

Decay Properties. We introduce the truncated correlations defined in the usual way

$$\rho_T(u_1, t \mid V) = \rho(u_1, t \mid V) - \rho(u_1)\,\rho(V) \tag{3.10}$$

$$\begin{aligned} \rho_T(u_1, u_2, t \mid V) = \rho(u_1, u_2, t \mid V) - \rho(u_1)\,\rho(u_2, t \mid V) - \rho(u_2)\,\rho(u_1, t \mid V) \\ - \rho(u_1, u_2)\,\rho(V) + 2\rho(u_1)\,\rho(u_2)\,\rho(V) \end{aligned} \tag{3.11}$$

At $t = 0$, these functions are a product of Maxwellians in momentum space and the equilibrium Ursell functions in configuration space. The latter are known to have good cluster properties in many cases, e.g., for an homogeneous OCP in the Debye–Hückel regime, they decay exponentially fast at large distances.[19]

We now assume that the time-dependent correlations still have reasonably good cluster properties. In particular we make the following assumptions for the homogeneous OCP:

(i) There is a sufficiently fast decay in momentum space

$$|\rho(q, p, t \mid V)| \leqslant \frac{M}{|p|^\eta}, \qquad \eta > 5 \tag{3.12}$$

for fixed q, t, V. This insures the existence of the second moment, so the kinetic energy density is finite.

(ii) The space and momentum distribution is bounded for $|q| \to \infty$ by

$$|\rho_T(q, p, t \mid V)| \leqslant \frac{M}{|q|^3} \tag{3.13}$$

for fixed p, t, V.

(iii) The charge density $\rho_T(q, t \mid V) = \int dp \, \rho_T(q, p, t \mid V)$ decays at least as

$$|\rho_T(q, t \mid V)| \leqslant \frac{M}{|q|^\eta}, \qquad \eta > 5 \tag{3.14}$$

for fixed t and V.

(iv) The three-point truncated spatial correlations are jointly integrable on two variables

$$\int dq_1 \int dq \, |q| \, |\rho_T(q_1, q_2, t \mid q)| < \infty \tag{3.15}$$

These cluster properties although not proven for any case, are in agreement with the small time expansion of the correlations; see Appendix B. [Notice that the density–density and density–momentum correlations (3.14) are expected to decay faster than the momentum–momentum correlations which are presumably not integrable on space for $t \neq 0$; see Appendix B.]

The precise assumptions required for the inhomogeneous systems we treat, i.e., those for which

$$\lim_{|q| \to \infty} \rho_b(|q|, \Omega) = \rho_\infty(\Omega) \tag{3.16}$$

are more cumbersome to write down explicitly in a concise way; we will generally state them when used. Basically we will assume that their correlations converge sufficiently rapidly to those of the homogeneous OCP with density $\rho_b(\Omega)$ whenever all particle coordinates go to infinity in a fixed direction Ω (see Section 3.3). We also assume the same properties as in (3.12)–(3.14) whenever q tends to infinity in a direction which is not parallel to the boundary of D. The semi-infinite case will be treated more explicitly in Appendix D.

Dynamical Equations. It will be useful to write the equations of motion in different forms. We first write the equivalent of (3.7) for the truncated functions. Substituting (3.10) and (3.11) in (3.7) gives

$$\frac{\partial}{\partial t}\rho_T(u_1, t \mid V) = -\frac{p_1}{m}\cdot\nabla_{q_1}\rho_T(u_1, t \mid V) - eE(q_1)\cdot\nabla_{p_1}\rho_T(u_1, t \mid V)$$
$$- e^2[\nabla_{p_1}\rho(u_1)]\cdot\int_D dq_2\, F(q_1-q_2)\,\rho_T(q_2, t \mid V)$$
$$- e^2\int_D dq_2\, F(q_1-q_2)\cdot\nabla_{p_1}\rho_T(u_1, q_2, t \mid V) \qquad (3.17)$$

In obtaining (3.17) we have used the equilibrium BGY equation

$$\beta^{-1}\nabla_{q_1}\rho(q_1) = eE(q_1)\,\rho(q_1) + e^2\int_D dq_2\, F(q_1-q_2)[\rho(q_1, q_2) - \rho(q_1)\,\rho(q_2)] \qquad (3.18)$$

to cancel the contribution of time-independent terms.

The evolution of the average of a momentum-independent function $f(q)$ is obtained by integrating out the momentum in (3.17). The integrals of the ∇_{p_1} terms gives no contribution because of the decay (3.12); thus we get

$$\frac{\partial}{\partial t}\int_D dq\, f(q)\,\rho_T(q, t \mid V) = -\int_D dq\, f(q)\,\nabla_q\cdot\int dp\,\frac{p}{m}\,\rho_T(q, p, t \mid V)$$
$$= \int_D dq[\nabla_q f(q)]\cdot\int dp\,\frac{p}{m}\,\rho_T(q, p, t \mid V) \qquad (3.19)$$

In the partial integration, there is no contribution at infinity by (3.13) and the surface contribution $\int_{\partial D} f(q)\, d\sigma\cdot\int dp(p/m)\,\rho_T(q, p, t \mid V)$ vanishes because of the reflection condition (3.9) [for $q\in\partial D$, change the variable $p\to\bar{p}$ and use with (3.9) the fact that $p\cdot d\sigma = -\bar{p}\cdot d\sigma$].

Finally, we find from (3.9), (3.17), and partial integration on momentum, that the second time derivative is given by

$$\frac{\partial^2}{\partial t^2}\int_D dq_1\, f(q_1)\,\rho_T(q_1, t \mid V)$$
$$= \frac{e^2}{m}\int_D dq_1[\nabla_{q_1} f(q_1)]\cdot\left[\rho(q_1)\int_D dq_2\, F(q_1-q_2)\,\rho_T(q_2, t \mid V)\right] \qquad (3.20)$$
$$+ \frac{1}{m}\int_D dq_1[\nabla_{q_1} f(q_1)]\cdot[eE(q_1)\,\rho_T(q_1, t \mid V)] \qquad (3.21)$$
$$- \frac{1}{m^2}\int_D dq_1\int dp_1[p_1\cdot\nabla_{q_1} f(q_1)][p_1\cdot\nabla_{q_1}\rho_T(q_1, p_1, t \mid V)] \qquad (3.22)$$
$$+ \frac{e^2}{m}\int_D dq_1\int_D dq_2\,\nabla_{q_1} f(q_1)\cdot F(q_1-q_2)\,\rho_T(q_1, q_2, t \mid V) \qquad (3.23)$$

3.2. Charge and Dipole Sum Rules

As mentioned in the introduction the plasma phase of the OCP is characterized by a set of sum rules expressing the perfect shielding of local charges[15,20]: in particular the static excess charge density of a (locally in-) homogeneous plasma carries no multipoles, i.e., with (3.6)

$$e \int dq\, \mathscr{Y}_l(q)\, \rho_T(q, t=0 \mid V) = 0 \tag{3.24}$$

where $\mathscr{Y}_l$ is a harmonic polynomial of order l. The case $l=0$ (resp. $l=1$) corresponds to the charge (resp. dipole) sum rule discussed earlier. We now look at the time-dependent generalizations of (3.24) from the point of view of the (formally) rigorous microscopic BBGKY hierarchy.

Charge Sum Rule. Choosing $f(q)=1$ in (3.19) gives immediately

$$e \frac{\partial}{\partial t} \int_D dq\, \rho_T(q, t \mid V) = 0$$

Hence, with the initial condition (3.24) for $l=0$, we find

$$e \int_D dq\, \rho_T(q, t \mid V) = 0 \tag{3.25}$$

for all times, or equivalently, with the symmetry (3.2),

$$e \int_D dq\, \rho_T(V, t \mid q) = 0$$

We therefore conclude that the charge sum rule remains true in the course of time for a general inhomogeneous OCP and for arbitrary positions and momenta of initial particles.

It is shown in Appendix C that the higher-order multipolar sum rules $l \geqslant 1$, $k \geqslant 2$ are not valid for $t \neq 0$. There is however one exception: the dipole sum rule in the uniform OCP, which we shall now prove.

Dipole Sum Rule in the Uniform OCP. We notice that in the uniform OCP $\rho(q) = \rho_b$ and $E(q) = 0$. Then Eq. (3.20) reduces to

$$\frac{\partial^2}{\partial t^2} \int dq_1\, f(q_1)\, \rho_T(q_1, t \mid V) = -\omega_p^2 \int dq_1\, f(q_1)\, \rho_T(q_1, t \mid V) \tag{3.26}$$

$$-\frac{1}{m^2}\int dq_1 \int dp_1[p_1 \cdot \nabla_{q_1} f(q_1)][p_1 \cdot \nabla_{q_1} \rho_T(q_1, p_1, t \mid V)] \tag{3.27}$$

$$+\frac{e^2}{m}\int dq_1 \int dq_2[\nabla_{q_1} f(q_1)] \cdot F(q_1 - q_2)\, \rho_T(q_1, q_2, t \mid V) \tag{3.28}$$

with

$$\omega_p^2 = \frac{4\pi e^2 \rho_b}{m}$$

We have integrated the term (3.20) by parts and used Poisson's equation $\nabla_q \cdot F(q) = 4\pi\delta(q)$.

If we now choose $f(q) = q$, the term (3.27) is the integral of a gradient which vanishes by the clustering (3.13). The term (3.28) is also zero because of the antisymmetry of $F(q)$, so Eq. (3.26) becomes an ordinary second-order differential equation. From (3.24) and (3.19), the initial conditions are found to be [with $V = (q_1, p_1;...; q_k, p_k)$]

$$\int dq\, q\rho_T(q, t=0 \mid q_1, p_1,..., q_k, p_k) = 0$$

$$\frac{\partial}{\partial t}\int dq\, q\rho_T(q, t \mid q_1, p_1,..., q_k, p_k)\bigg|_{t=0} = \frac{1}{m}\left(\sum_{j=1}^{k} p_j\right)\rho(q_1, p_1,..., q_k, p_k)$$

and the solution is thus

$$\int dq\, q\rho_T(q, t \mid q_1, p_1,..., q_k, p_k) = \frac{1}{m\omega_p}\left(\sum_{j=1}^{k} p_j\right)\rho(q_1, p_1,..., q_k, p_k)\sin \omega_p t \tag{3.29}$$

When we average (3.29) on initial momenta, we get

$$e\int dq\, q\rho_T(q, t \mid q_1,..., q_k) = 0 \tag{3.30}$$

for all t and all $q_1,..., q_k$, which is the time-dependent dipole sum rule. It can be understood as follows: in the OCP, the dipole (3.30) is proportional to the center of mass at time t of the local perturbation initially at $q_1,..., q_k$. Since the center of mass decouples from the relative coordinates, it is only subjected to the harmonic force of the background. Thus it oscillates at frequency ω_p [Eq. (3.29)] and remains constant if there is no initial velocity.

The Second Moment of the Structure Function in the Uniform OCP. As an application of the charge and dipole sum rules, we derive the second moment relation Eq. (2.16) for the uniform OCP,

$$-\frac{2\pi}{3}\int dq\,|q|^2\,S(q,t\mid 0)=\int dq\int dq_1\,\frac{1}{|q_1|}\,S(q_1,t\mid q)$$
$$=\beta^{-1}\cos\omega_p t \tag{3.31}$$

In the uniform OCP, the Carnie and Chan form (2.16) is identical with the second moment expression. It is useful to treat this case first since the same methods will apply also to the inhomogeneous OCP treated in the next subsection.

We take $f(q_1)=1/|q_1|$, $V=(q,p)$ in (3.26) and integrate over the position q and momentum p of the initial particle. We will establish below that, as a consequence of (3.25) and (3.30) the terms (3.27) and (3.28) vanish for all times, i.e.,

$$\int dq\left\{\int dq_1\int dp_1(p_1\cdot F(q_1))(p_1\cdot\nabla_{q_1}\rho_T(q_1,p_1,t\mid q))\right\}=0 \tag{3.32}$$

$$\int dq\left\{\int dq_1\int dq_2\,F(q_1)\cdot F(q_1-q_2)\,\rho_T(q_1,q_2,t\mid q)\right\}=0 \tag{3.33}$$

Then, with the definitions (3.4), (3.5), Eq. (3.26) reduces again to the simple differential equation

$$\frac{\partial^2}{\partial t^2}\int dq\int dq_1\,\frac{1}{|q_1|}\,S(q_1,t\mid q)=-\omega_p^2\int dq\int dq_1\,\frac{1}{|q_1|}\,S(q_1,t\mid q) \tag{3.34}$$

With the initial conditions $\int dq\int dq_1(1/|q_1|)\,S(q_1,t=0\mid q)=\beta^{-1}$ (the Stillinger–Lovett perfect screening relation) and $(\partial/\partial t)\int dq\int dq_1(1/|q_1|)\,S(q_1,t\mid q)\,|_{t=0}=0$ which follows from (3.19), the solution of Eq. (3.34) is identical to (3.31).

To show (3.32) and (3.33) we set

$$h(q_1,t\mid q)=\int dp_1\,p_1(p_1\cdot\nabla_{q_1}\rho_T(q_1,p_1,t\mid q))$$
$$=-\int dp_1\,p_1(p_1\cdot\nabla_q\rho_T(0,p_1,t\mid q-q_1)) \tag{3.35}$$

$$g(q_1,t\mid q)=\int dq_2\,F(q_1-q_2)\,\rho_T(q_1,q_2,t\mid q)$$
$$=-\int dq_2\,F(q_2)\,\rho_T(0,q_2,t\mid q-q_1) \tag{3.36}$$

where translation invariance has been used.

With this, the brackets in (3.32) and (3.33) can be written as gradient terms

$$\{\cdots\}(3.32) = \int dq_1\, F(q_1) \cdot h(0, t \mid q - q_1)$$

$$= \nabla_q \cdot \int dq_1 \frac{1}{|q - q_1|} h(0, t \mid q_1) \tag{3.37}$$

$$\{\cdots\}(3.33) = \nabla_q \cdot \int dq_1 \frac{1}{|q - q_1|} g(0, t \mid q_1) \tag{3.38}$$

Since $h(0, t \mid q)$ is itself a gradient, and with the decay (3.13), we have $\int dq\, h(0, t \mid q) = 0$. Moreover, it follows from the charge sum rule (3.25) that

$$\int dq\, qh(0, t \mid q) = \int dp_1\, p_1 p_1 \int dq\, \rho_T(0, p_1, t \mid q) = 0$$

Thus $h(0, t \mid q)$ carries no charge and no dipole. The same is true for $g(0, t \mid q)$ as a consequence of (3.25) and of the dipole sum rule (3.30) for $k = 2$

$$\int dq\, qg(0, t \mid q) = \int dq_2\, F(q_2) \int dq\, q\rho_T(0, q_2, t \mid q) = 0$$

[we have also used the integrability condition (3.15) and the symmetry (3.2)]. Hence, the potentials due to the distributions $h(0, t \mid q)$ and $g(0, t \mid q)$ are $o(1/|q|^2)$ as $|q| \to \infty$, showing that the integrals of the gradients (3.37) and (3.38) give no contributions at infinity.

3.3. Dynamical Sum Rules in the Inhomogeneous OCP

We consider in this section a general inhomogeneous OCP with an asymptotically constant background density (3.16). [We set $\rho_\infty(\Omega) = 0$ if the direction Ω does not belong to D.] We assume that

$$|\rho(r, \Omega) - \rho_\infty(\Omega)| \leqslant \frac{M}{r^\eta}, \qquad \eta > 1, \quad q = (r = |q|, \Omega) \tag{3.39}$$

the limit being obtained without fast oscillations

$$\int_0^\infty dr \left| \frac{\partial}{\partial r} \rho(r, \Omega) \right| < \infty \tag{3.40}$$

The Dynamical Carnie and Chan Sum Rule. Let

$$\phi(q_1, q, t) = \int_D dq_2 \frac{1}{|q_1 - q_2|} S(q_2, t \mid q) \tag{3.41}$$

be the potential at q_1 due to the distribution $S(q_2, t \mid q)$ for fixed q. We have

$$\phi(q_1, q, t) = o\left(\frac{1}{|q_1|}\right), \qquad q \text{ fixed} \tag{3.42}$$

since $S(q_2, t \mid q)$ carries no net charge.

We shall assume moreover that for fixed q_1, $\phi(q_1, q, t)$ is integrable with respect to q and the integral is a uniformly bounded function of q_1, i.e.,

$$\int_D dq\, |\phi(q_1, q, t)| < M, \qquad M \text{ independent of } q_1 \tag{3.43}$$

(See Appendix D for a discussion of this condition in the semi-infinite OCP.)

We notice the following property of $\phi(q_1, q, t)$: as a consequence of Poisson's equation and the electroneutrality (3.25)

$$\nabla_{q_1}^2 \phi(q_1, q, t) = -4\pi \int_D dq\, S(q_1, t \mid q) = 0 \tag{3.44}$$

Thus $\int_D dq\, \phi(q_1, q, t)$ is bounded and harmonic on the whole R^3; it is therefore constant with respect to q_1:

$$\int_D dq\, \phi(q_1, q, t) = \int_D dq\, \phi(0, q, t) \tag{3.45}$$

To establish (2.16) we set again $f(q_1) = 1/|q_1|$, $V = (q, p)$ in Eq. (3.20) and integrate it on q and p. We will show below that under some additional integrability conditions all terms in the right hand side of (3.20) vanish except the first one. Thus we are left with

$$\frac{\partial^2}{\partial t^2} \int_D dq\, \phi(0, q, t)$$

$$= \frac{e^2}{m} \int_D dq \left\{ \int_D dq_1\, F(q_1) \cdot [\rho(q_1) \nabla_{q_1} \phi(q_1, q, t)] \right\} \tag{3.46}$$

$$= -\frac{e^2}{m} \int_D dq \left\{ \int_D dq_1 [\nabla_{q_1} \cdot \rho(q_1)\, F(q_1)]\, \phi(q_1, q, t) \right\}$$

$$+ \frac{e^2}{m} \int_D dq \int_{\partial D} d\sigma_1 \cdot F(q_1)\, \rho(q_1)\, \phi(q_1, q, t) \tag{3.47}$$

Since by (3.42) $F(q_1)\,\rho(q_1)\,\phi(q_1, q, t) = o(1/|q_1|^2)$ for fixed q, there are no contributions at infinity in the integration by parts leading to (3.47), where ∂D denotes the finite distance boundaries of D. Under the assumptions (3.40), (3.43), the integrand in (3.47) is jointly integrable in q and q_1, hence integrals can be permuted. From the property (3.45), we get

$$\frac{\partial^2}{\partial t^2}\int_D dq\,\phi(0, q, t) = -\bar{\omega}^2 \int_D dq\,\phi(0, q, t) \tag{3.48}$$

with

$$\begin{aligned}\bar{\omega}^2 &= \frac{e^2}{m}\int_D dq_1\,\nabla_{q_1}\cdot[\rho(q_1)\,F(q_1)] - \int_{\partial D} d\sigma_1\cdot\rho(q_1)\,F(q_1)\\ &= -\frac{e^2}{m}\lim_{r\to\infty}\int_{|q_1|=r} d\sigma_1\cdot\left(\nabla_{q_1}\frac{1}{|q_1|}\right)\rho(q_1)\\ &= \frac{e^2}{m}\lim_{r\to\infty}\int d\Omega\,\rho(r,\Omega) = \frac{e^2}{m}\int d\Omega\,\rho_\infty(\Omega)\end{aligned} \tag{3.49}$$

where (3.39) and (3.40) have been used.

The solution of (3.48) with initial conditions $\int_D dq\,\phi(0, q, t=0) = \beta^{-1}$ (the inhomogeneous Stillinger–Lovett relation) and $(\partial/\partial t)\int_D dq\,\phi(0, q, t)\,|_{t=0} = 0$ [which follows from (3.19)] is precisely the formula (2.16).

It remains to explain why the terms (3.21), (3.22), and (3.23) vanish. Consider first the simple case of a local inhomogeneity: $\rho_\infty(\Omega) = \rho_\infty$ is independent of Ω. Let $\rho_T^{(\infty)}$ be the correlations of the uniform OCP with constant background density ρ_∞, and use the fact that the pure bulk contributions (3.32) and (3.33) vanish to write the terms (3.22) and (3.23) as

$$\int dq\left\{\int dq_1\int dp_1 (p_1\cdot F(q_1))[p_1\cdot\nabla_{q_1}(\rho_T(q_1, p_1, t\mid q) - \rho_T^{(\infty)}(q_1, p_1, t\mid q))]\right\} \tag{3.50}$$

$$\int dq\left\{\int dq_1\int dq_2\, F(q_1)\cdot F(q_1 - q_2)(\rho_T(q_1, q_2, t\mid q) - \rho_T^{(\infty)}(q_1, q_2, t\mid q))\right\} \tag{3.51}$$

If spatial arguments in $\rho_T - \rho_T^{(\infty)}$ are far apart, both ρ_T and $\rho_T^{(\infty)}$ vanish by clustering; when all arguments tend simultaneously to infinity in the same direction, $\rho_T - \rho_T^{(\infty)}$ tends to zero because of the convergence of the correlations of the inhomogeneous system to those of the uniform one. We assume at this point that this convergence is sufficiently fast to have the integrands (3.50) and (3.51) jointly integrable in all spatial variables, so that we can permute the q and the other integrals. Then these terms vanish in view of the charge sum rules (3.25). In (3.21), one has $E(q_1) = O(1/|q_1|^2)$ and thus $[\nabla_{q_1}(1/|q_1|)] \cdot E(q_1) = O(1/|q_1|^4)$. Hence with (3.14) the integrand is jointly integrable in q_1, q. Performing the q integral first, we get again zero by the charge sum rule.

In the general case, we define $\rho_T^{(\infty,\Omega)}$, the correlations of an uniform OCP with constant background density $\rho_\infty(\Omega)$. It follows from the rotation invariance of the uniform state that the brackets in (3.32) and (3.33) depend only on $|q|$. We have for instance from (3.33) for each fixed Ω

$$\int_0^\infty d|q|\, |q|^2 \left\{ \int dq_1 \int dq_2\, F(q_1) \cdot F(q_1 - q_2)\, \rho_T^{(\infty,\Omega)}(q_1, q_2, t \mid |q|, \Omega) \right\} = 0$$

and a similar identity corresponding to (3.32). We can now reproduce exactly the same argument as above replacing $\rho_T^{(\infty)}(q_1, p_1, t \mid q)$ and $\rho_T^{(\infty)}(q_1, q_2, t \mid q)$ by $\rho_T^{(\infty,\Omega)}(q_1, p_1, t \mid |q|, \Omega)$ and $\rho_T^{(\infty,\Omega)}(q_1, q_2, t \mid |q|, \Omega)$ in (3.50) and (3.51). The case of the semi-infinite OCP is treated in detail in Appendix C.

Dipole Sum Rule in the Semi-Infinite OCP. Under similar assumptions as in the preceding section, we easily recover the classical form of the dipole sum rule (2.43). Adding and substracting the corresponding bulk quantity, we get (with the same notations as in Section 2)

$$\begin{aligned}
&\int_0^\infty dx \int_0^\infty dx_1 \int dy_1\, x_1 S(x_1, y_1, t \mid x) \\
&\quad = \int_0^\infty dx \int_0^\infty dx_1 \int dy_1\, x_1 [S(x_1, y_1, t \mid x) - S^{(\infty)}(x_1, y_1, t \mid x)] \\
&\qquad + \int_0^\infty dx \int_0^\infty dx_1 \int dy_1\, x_1 S^{(\infty)}(x_1, y_1, t \mid x)
\end{aligned} \tag{3.52}$$

Assuming the joint integrability of the first moment of the difference of the semi-infinite and bulk structure function, we can permute the x and x_1 integrals in the first term of the right-hand side of (3.52). Doing this and applying then the charge sum rule (3.25), the contribution from the semi-infinite system on the right side of (3.52) vanishes:

$$\int_0^\infty dx \int dy_1\, S(x_1, y_1, t \mid x) = \int_0^\infty dx \int dy_1\, S(x_1, t \mid x, y_1) = 0$$

we then obtain

$$\begin{aligned}
&\int_0^\infty dx \int_0^\infty dx_1\, x_1 \int dy_1\, S(x_1, y_1, t \mid x) \\
&= \int_0^\infty dx \int_0^\infty dx_1\, x_1 \int dy_1\, S^{(\infty)}(x_1 - x, y_1, t \mid 0) \\
&\quad - \int_0^\infty dx_1 \int_0^\infty dx\, x_1 \int dy_1\, S^{(\infty)}(x_1 - x, y_1, t \mid 0) \\
&= \int_0^\infty dx \int_0^\infty dx_1 (x_1 - x) \int dy_1\, S^{(\infty)}(x_1 - x, y_1, t \mid 0) \\
&= \frac{1}{2} \int_{-\infty}^\infty dx\, x^2 \int dy\, S^{(\infty)}(x, y, t \mid 0) = -\frac{1}{4\pi\beta} \cos \omega_p t \qquad (3.53)
\end{aligned}$$

(3.53) results simply of the bulk second moment relation (3.31), and this gives the classical limit of (2.43).

With a treatment analogous to (3.52) for the half spaces $x_1 < 0$ and $x_1 > 0$ in the OCP with two densities, we get by the same reasoning the classical versions of (2.46a) and (2.46b).

ACKNOWLEDGMENTS

We thank A. Alastuey and M. L. Rosinberg for stimulating discussions. This research was supported in part by AFOSR under Grant No. 82-0016C at Rutgers University and the Swiss National Foundation for Science.

APPENDIX A: PLASMA IN A WEDGE

For a plasma in a wedge of angle α, (2.15) becomes

$$\begin{aligned}
&\lim_{k \to 0} 2 \int_0^\infty dr_1\, r_1 \int_0^\alpha d\theta_1 \int_0^\infty dr\, r \int_0^\alpha d\theta\, K_0(|k|\, r_1)\, \tilde{S}(r_1, \theta_1, k, t \mid r, \theta) \\
&= \frac{\hbar\bar{\omega}}{2} \left[\frac{\exp(-i\bar{\omega}t)}{1 - \exp(-\beta\hbar\bar{\omega})} - \frac{\exp(i\bar{\omega}t)}{1 - \exp(\beta\hbar\bar{\omega})} \right] \qquad (\text{A1})
\end{aligned}$$

where $\bar{\omega}$ is defined by (2.28). The Bessel function $K_0(|k|\, r_1)$ makes the integral absolutely convergent, and the order of the integrations is arbitrary. In the left-hand side of (A1), there are contributions from (r_1, θ_1) far away from the walls, with the bulk behavior

$$\lim_{r_1 \to \infty} \int_0^\infty dr\, r \int_0^\alpha d\theta\, \tilde{S}\left(r_1, \frac{\alpha}{2}, k, t \mid r, \theta\right)$$
$$\sim \frac{1}{8\pi} \hbar\omega_p \left[\frac{\exp(-i\omega_p t)}{1-\exp(-\beta\hbar\omega_p)} - \frac{\exp(i\omega_p t)}{1-\exp(\beta\hbar\omega_p)}\right] |k|^2, \qquad \text{when} \quad k \to 0 \tag{A2}$$

[the left-hand side of (A2) is (2.21) transposed to cylindrical coordinates]. There are also contributions from (r_1, θ_1) near one of the walls but far away from the edge of the wedge, with the semi-infinite geometry behavior

$$\lim_{r_1 \to \infty} r_1 \left[\int_0^\alpha d\theta_1 \int_0^\infty dr\, r \int_0^\alpha d\theta\, \tilde{S}(r_1, \theta_1, k, t \mid r, \theta) - \alpha \int_0^\infty dr\, r \int_0^\alpha d\theta\, \tilde{S}\left(r_1, \frac{\alpha}{2}, k, t \mid r, \theta\right)\right]$$
$$\sim \frac{1}{2\pi} \hbar\omega_s \left[\frac{\exp(-i\omega_s t)}{1-\exp(-\beta\hbar\omega_s)} - \frac{\exp(i\omega_s t)}{1-\exp(\beta\hbar\omega_s)}\right]$$
$$- \frac{1}{4\pi} \hbar\omega_p \left[\frac{\exp(-i\omega_p t)}{1-\exp(-\beta\hbar\omega_p)} - \frac{\exp(i\omega_p t)}{1-\exp(\beta\hbar\omega_p)}\right] |k|, \qquad \text{when} \quad k \to 0 \tag{A3}$$

[The left-hand side of (A3) is Eq. (3.13) of Ref. 10, transposed to cylindrical coordinates, and multiplied by a factor 2 because there are contributions from both walls of the wedge.] The contributions of (A2) and (A3) to (A1) are easily computed, since they involve the integrals

$$\int_0^\infty dr_1\, r_1 k^2 K_0(|k|\, r_1) = 1, \qquad \int_0^\infty dr_1\, |k|\, K_0(|k|\, r_1) = \frac{\pi}{2} \tag{A4}$$

Substracting in (A1) the contributions from (A2) and (A3), we find

$$\lim_{k \to 0} 2 \int_0^\infty dr_1\, K_0(|k|\, r_1) \left\{ r_1 \int_0^\alpha d\theta_1 \int_0^\infty dr\, r \int_0^\alpha d\theta\, \tilde{S}(r_1, \theta_1, k, t \mid r, \theta) \right.$$
$$- r_1 \alpha \lim_{r_1 \to \infty} \int_0^\infty dr\, r \int_0^\alpha d\theta\, \tilde{S}\left(r_1, \frac{\alpha}{2}, k, t \mid r, \theta\right)$$
$$- \lim_{r_1 \to \infty} r_1 \left[\int_0^\alpha d\theta_1 \int_0^\infty dr\, r \int_0^\alpha d\theta\, \tilde{S}(r_1, \theta_1, k, t \mid r, \theta)\right.$$
$$\left.\left. - \alpha \int_0^\infty dr\, r \int_0^\alpha d\theta\, \tilde{S}\left(r_1, \frac{\alpha}{2}, k, t \mid r, \theta\right)\right]\right\}$$

$$= \frac{\hbar\bar{\omega}}{2}\left[\frac{\exp(-i\bar{\omega}t)}{1-\exp(-\beta\hbar\bar{\omega})} - \frac{\exp(i\bar{\omega}t)}{1-\exp(\beta\hbar\bar{\omega})}\right]$$
$$- \frac{\hbar\omega_s}{2}\left[\frac{\exp(-i\omega_s t)}{1-\exp(-\beta\hbar\omega_s)} - \frac{\exp(i\omega_s t)}{1-\exp(\beta\hbar\omega_s)}\right]$$
$$+ \left(\frac{1}{2} - \frac{\alpha}{2\pi}\right)\frac{\hbar\omega_p}{2}\left[\frac{\exp(-i\omega_p t)}{1-\exp(-\beta\hbar\omega_p)} - \frac{\exp(i\omega_p t)}{1-\exp(\beta\hbar\omega_q)}\right] \tag{A5}$$

We now assume that the curly bracket in the left-hand side of (A5) goes rapidly to zero as r_1 increases, since we have substracted all contributions except those from the neighborhood of the edge. Thus we can replace K_0 by its leading term for small $|k|\ r_1$

$$K_0(|k|\ r_1) \sim -\ln(|k|\ r_1) \tag{A6}$$

Furthermore, the curly bracket in (A5) vanishes at $k=0$, because of the perfect screening condition

$$\int_0^\infty dr\, r \int_0^\alpha d\theta\, \tilde{S}(r_1, \theta_1, 0, t \mid r, \theta) = 0 \tag{A7}$$

Thus only the term $-\ln|k|$ of (A6) contributes in the limit $k=0$, and $\int_0^\infty dr_1\{\cdots\}$ behaves like $-C/(2\ln|k|)$, where C is the right-hand side of $(A5)$. If there is no other singularity on the reak k axis, a function of k which behaves like $-C/(2\ln|k|)$ has an inverse Fourier transform, a function of z, which behaves asymptotically[13] like $-C/[4\,|z|\,(\ln|z|)^2]$. Since the bulk and surface contributions on (A5) have a faster decay, and assuming that the asymptotic form of S with respect to $z-z_1$ is unchanged by the integration upon the variables r, θ, r_1, θ_1, we obtain (2.31).

APPENDIX B: TIME DERIVATIVES AT $t=0$

We compute the first and second time derivative of the correlations at $t=0$. For simplicity, we consider the homogeneous case $\rho(q)=\rho_b$, $E(q)=0$. The calculation involves the following steps.

(i) We single out the contribution of coincident points in the equilibrium correlations:

$$\rho(u, t=0 \mid V) = \rho(u, V) + \left[\sum_{j=1}^{k} \delta(u, v_j)\right]\rho(V) \tag{B1}$$

$$\rho(u, u', t=0 \mid V) = \rho(u, u', V) + \left[\sum_{j=1}^{k} \delta(u, v_j)\right] \rho(u^1, V) + \left[\sum_{j=1}^{k} \delta(u', v_j)\right] \rho(u, V) + \left[\sum_{i \neq j}^{k} \delta(u, v_i)\, \delta(u^1, v_j)\right] \rho(V) \qquad \text{(B2)}$$

and so on $[V = (q_1, p_1; ...; q_k, p_k), \delta(u, v_j) = \delta(q - q_j)\, \delta(p - p_j)]$.

(ii) We eliminate the higher-order correlations with the help of the equilibrium BGY equation:

$$\beta^{-1} \nabla_{q_i} \rho(q_1, ..., q_k) = \sum_{j \neq i}^{k} F(q_i - q_j)\, \rho(q_1, ..., q_k) + \int dq\, F(q_i - q)(\rho(q_1, ..., q_k, q) - \rho_b \rho(q_1, ..., q_k)) \qquad \text{(B3)}$$

(iii) We perform momentum integrations with the Maxwellian distribution. With this, it is found from (3.7) that for $q \neq q_1, ..., q_k$

$$\frac{\partial}{\partial t} \rho_T(u, t \mid V) \bigg|_{t=0} = 0$$

so the first derivative is strictly local in q.

We compute $(\partial^2/\partial t^2)\, \rho(u, t \mid V) \mid_{t=0}$ by iterating (3.7) and introducing the second BBGKY equation

$$\frac{\partial}{\partial t} \rho(u_1, u_2, t \mid V) = -\left(\frac{p_1}{m} \cdot \nabla_{q_1} + \frac{p_2}{m} \cdot \nabla_{q_2}\right) \rho(u_1, u_2, t \mid V) - e^2 [F(q_1 - q_2) \cdot \nabla_{p_1} + F(q_2 - q_1) \cdot \nabla_{p_2}]\, \rho(u_1, u_2, t \mid V) - e^2 \int dq_3 [F(q_1 - q_3) \cdot \nabla_{p_1} + F(q_2 - q_3) \cdot \nabla_{p_2}] \times [\rho(u_1, u_2, q_3, t \mid V) - \rho_b \rho(u_1, u_2, q_3, t \mid V)] \qquad \text{(B4)}$$

Using repeatedly (B2) and (B3) the final result takes the simple form for $q \neq q_1,..., q_k$

$$\frac{\partial^2}{\partial t^2} \rho_T(u, t \mid V) \bigg|_{t=0} = \frac{\beta e^2}{m^2} \rho(u, V) \sum_{j=1}^{k} (p \cdot \nabla_q)(p_j \cdot \nabla_q) \frac{1}{|q - q_j|} \quad \text{(B5)}$$

The right-hand side of (B5) can be interpreted as the instantaneous dipole potential due to the displacements $(p/m)\,dt$ and $(p_j/m)\,dt$ of the particles. (B5) shows that up to order t^2 there is a fast decay in momentum space [$\rho(u, V)$ is Maxwellian], but the decay in space is not faster than $|q|^{-3}$ [cf. (3.13)]. However, the right-hand side of (B5) vanishes when it is integrated on p, indicating that the charge correlations have faster decay properties [cf. (3.14)].

APPENDIX C: SUM RULES AT $t \neq 0$

To investigate the validity of the higher order sum rules ($l \geqslant 1$) at $t \neq 0$, we explicitly compute the time derivative of the excess particle density at $t = 0$. Separating the Maxwellian distribution from the configurational part, and using the fact that the average equilibrium momentum is zero, Eq. (3.19) gives for $t = 0$ [with $V = (q_1, p_1; ...; q_k, p_k)$]

$$e \frac{\partial}{\partial t} \int_D dq\, \mathscr{Y}_l(q)\, \rho_T(q, t \mid V) \bigg|_{t=0} = \frac{e}{m} \left[\sum_{j=1}^{k} p_j \cdot \nabla_{q_j} \mathscr{Y}_l(q_j) \right] \rho(V) \quad \text{(C1)}$$

The right-hand side of (C1) with $l \neq 0$ [$\mathscr{Y}_l(q) \neq \text{const}$] is obviously not zero, for a general V, showing that the higher-order multipoles are not conserved when we specify an initial distribution of particles with arbitrary velocities.

If we only specify the initial positions (averaging over $p_1,..., p_k$), (C1) vanishes, and we have to discuss the second-order time derivative. We consider the homogeneous case $\rho(q) = \rho_b$, $E(q) = 0$ and find from (3.20)

$$\frac{\partial^2}{\partial t^2} \int dq\, \mathscr{Y}_l(q)\, \rho_T(q, t \mid q_1,..., q_k) \bigg|_{t=0}$$

$$= -\frac{1}{m^2} \int dq \int dp [p \cdot \nabla_q \mathscr{Y}_l(q)][p \cdot \nabla_q \rho_T(q, p, 0 \mid q_1,..., q_k)] \quad \text{(C2)}$$

$$+ \frac{e^2}{m} \int dq \int dq' [\nabla_q \mathscr{Y}_l(q)] \cdot F(q - q')$$

$$\times [\rho(q, q', 0 \mid q_1,..., q_k) - \rho_b \rho(q, 0 \mid q_1,..., q_k)] \quad \text{(C3)}$$

(C3) results from the combination of the terms (3.20) and (3.23) with the definition of the truncated functions. In (C2) we can perform the p integration on the spherically symmetric Maxwellian distribution. After an integration by parts on q, we see that this term vanishes since $\nabla_q^2 \mathscr{Y}_l(q)=0$. In (C3), we tract the contribution of coincident points with the help of (B2). Then, we use the equilibrium equation (B3) together with $\nabla_q^2 \mathscr{Y}_l(q)=0$ to eliminate the occurrence of the force, and we are finally left with

$$\frac{\partial^2}{\partial t^2}\int dq\, \mathscr{Y}_l(q)\, \rho_T(q, t \mid q_1,..., q_k)\bigg|_{t=0} = \frac{1}{\beta m}\sum_{j=1}^{k} (\nabla_{q_j}\mathscr{Y}_l(q_j))\cdot\nabla_{q_j}\rho(q_1,..., q_k) \tag{C4}$$

In the uniform OCP, (C5) vanishes in the following three cases: (a) $l=0$, all k, corresponding to the charge sum rule (3.25); (b) $l=1$, all k (because of the translation invariance), corresponding to the dipole sum rule (3.30); (c) $k=1$, all l, corresponding to the spherically symmetric structure function. In all other cases (C5) is in general not zero, showing that the multiple sum rules for $l \geqslant 1$, $k \geqslant 2$ cannot hold in the uniform OCP at $t \neq 0$.

APPENDIX D: SEMI-INFINITE PLASMA

We discuss in some details the conditions (3.42) and (3.43) as well as the terms (3.50) and (3.51) for the semi-infinite OCP. Similar arguments apply to the OCP with two densities.

The working hypothesis is that the difference of the semi-infinite and bulk structure functions are jointly integrable in x_1, y_1 and $x>0$, i.e.,

$$\int_0^\infty dx \int dx_1 \int dy_1\, |\theta(x_1)\, S(x_1, y_1, t \mid x) - S^{(\infty)}(x_1, y_1, t \mid x)| < \infty$$

$$\theta(x_1) = \begin{cases} 1, & x_1 > 0 \\ 0, & x_1 < 0 \end{cases} \tag{D1}$$

[Note that in (D1), the contribution of $x_1 < 0$,

$$\int_0^\infty dx \int_{-\infty}^0 dx_1 \int dy_1\, |S^{(\infty)}(x_1 - x, y_1, t \mid 0)|$$

$$= \int_0^\infty dx_1 \int dy_1\, x_1\, |S^{(\infty)}(x_1, y_1, t \mid 0)|$$

which involves only the bulk function, is finite.]

Similarly, we assume that the functions h and g defined in (D3) and (D4) below are integrable in $x_1 y_1$ and $x>0$.

Notice that

$$\phi^{(\infty)}(q_1, q, t) = \int dq_2 \frac{1}{|q_1 - q - q_2|} S^{(\infty)}(q_2, t \mid 0)$$

is rapidly decreasing as $|q| \to \infty$ since the spherically symmetric function $S^{(\infty)}(q_2, t \mid 0)$ has no charge and multipoles. To get (3.42) it is therefore sufficient to show that

$$\begin{aligned} &\phi(q_1, x, y, t) - \phi^{(\infty)}(q_1, x, y, t) \\ &\quad = \int dx_2 \int dy_2 \left[\frac{1}{(x_1 - x_2)^2 + (y_1 - y - y_2)^2}\right]^{1/2} \\ &\qquad \times [\theta(x_2)\, S(x_2, y_2, t \mid x) - S^{(\infty)}(x_2, y_2, t \mid x)] \end{aligned} \tag{D2}$$

is integrable in y and $x>0$. Integrability on x is insured by (D1). Moreover, since $S(x_2, y_2, t \mid x) - S^{(\infty)}(x_2, y_2, t \mid x)$ has no dipole in the y direction (because of the $y \to -y$ invariance), the potential (D2) decays faster than $1/|y|^2$ as $|y| \to \infty$ providing integrability in the y plane.

We show that the terms (3.50) and (3.51) vanish in the semi-infinite OCP. We define as in (3.35) and (3.36)

$$\begin{aligned} &h(q_1, t \mid q) \\ &= \int dp_1\, p_1 [\theta(x_1)\, p_1 \cdot \nabla_{q_1} \rho_T(q_1, p_1, t \mid q) - p_1 \cdot \nabla_{q_1} \rho_T^{(\infty)}(q_1, p_1, t \mid q)] \end{aligned} \tag{D3}$$

$$\begin{aligned} &g(q_1, t \mid q) \\ &= \int dq_2\, F(q_1 - q_2) [\theta(x_1)\, \theta(x_2)\, \rho_T(q_1, q_2, t \mid q) - \rho_T^{(\infty)}(q_1, q_2, t \mid q)] \end{aligned} \tag{D4}$$

Then we have

$$\int dx_1 \int_0^\infty dx \int dy\, h(x_1, t \mid x, y) = 0 \tag{D5}$$

$$\int dx_1 \int_0^\infty dx \int dy\, g(x_1, t \mid x, y) = 0 \tag{D6}$$

In (D3) we distinguish between the y and x components of ∇_{q_1} and use the invariance under translations in the y direction to write

$$p_1 \cdot \nabla_{q_1} = p_1^y \cdot \nabla_{y_1} + p_1^x \frac{\partial}{\partial x_1} = -p_1^y \cdot \nabla_y + p_1^x \frac{\partial}{\partial x_1}$$

The y integral of the $-p_1^y \cdot \nabla_y$ term vanishes, and performing the x_1 integral first in the $p_1^x(\partial/\partial x_1)$ term gives

$$\int dx_1 \int_0^\infty dx \int dy\, h(x_1, t \mid x, y) = -\int dp_1\, p_1\, p_1^x \int_0^\infty dx \int dy\, \rho_T(0, p_1, t \mid x, y) = 0 \tag{D7}$$

because of the charge sum rule (3.25). This gives (D5), Eq. (D6) results of the antisymmetry of the force under the exchange of the q_1 and q_2 arguments.

With the definitions (D5) the term (3.50) reads

$$\int_0^\infty dx \int dy \int dx_1 \int dy_1\, F(x_1, y_1) \cdot h(x_1, t \mid x, y - y_1) = \int dy \int dx_1 \int dy_1\, F(x_1, y - y_1) \cdot \int_0^\infty dx\, h(x_1, t \mid x - y_1) \tag{D8}$$

where we have permuted the x and x_1, y_1 integrals in view of the integrability assumption on h. Writing explicitly the y and x components F^y and F^x of the force in (D8) gives rise to two terms

$$-\int dy\, \nabla_y \cdot \int dx_1 \int dy_1 \left[\frac{1}{x_1^2 + (y - y_1)^2}\right]^{1/2} \int_0^\infty dx\, h^x(x_1, t \mid x, y_1) \tag{D9}$$

and

$$\int dy \int dx_1 \int dy_1 \frac{x_1}{[x_1^2 + (y - y_1)^2]^{3/2}} \int_0^\infty dx\, h^x(x_1, t \mid x, y_1) = 2\pi \int dx_1\, \operatorname{sign} x_1 \int dy_1 \int_0^\infty h^x(x_1, t \mid x, y_1) \tag{D10}$$

where the y integral has been performed: $\int dy\, x_1/(x_1^2 + y^2)^{3/2} = 2\pi \operatorname{sign} x_1$.

The term (D9) is the y integral of the gradient of a potential which is $o(1/|y|)$ because of (D5): hence it vanishes.

The term (D10) is explicitly

$$-2\pi \int dp_1 (p_1^x)^2 \left\{\int_0^\infty dx \int dy [\rho_T(0, p_1, t \mid x, y) - 2\rho_T^{(\infty)}(0, p_1, t \mid x, y)]\right\} = -2\pi \int dp_1 (p_1^x)^2 \left[\int_0^\infty dx \int dy\, \rho_T(0, p_1, t \mid x, y) - \int dx \int dy\, \rho_T^{(\infty)}(0, p_1, t \mid x, y)\right] \tag{D11}$$

In the last term of (D11), the x integral has been extended from $-\infty$ to $+\infty$ by the space reflexion symmetry of the homogeneous state. Then both terms of (D11) vanish because of the charge sum rule (3.25). This proves that (3.50) does not contribute to the equation of motion in the semi-infinite OCP. The term (3.51) is treated in the same way with h replaced by g.

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