Decay of Correlations and Related Sum Rules in a Layered Classical Plasma.

R. FANTONI and M. P. TOSI
Scuola Normale Superiore - Piazza dei Cavalieri 7, I-56126 Pisa, Italy

(ricevuto il 22 Novembre 1994; approvato il 30 Gennaio 1995)

Summary. — The asymptotic behaviours of particle correlation functions and the related sum rules are discussed for a layered classical plasma with e^2/r interactions in the fluid state, in dependence on the number of layers. These properties derive from consistency conditions imposed by screening on the hierarchical equations, as already treated by A. Alastuey and P. A. Martin (*J. Stat. Phys.*, **39**, 405 (1985)) for various Coulomb fluids. The main results concern i) the type of clustering of correlations needed for the validity of multipolar sum rules at various orders, ii) the proof that the pair correlation function in a finite multilayer may carry an electric dipole moment and the calculation of its partioning among the layers, and iii) the dimensionality crossover in an infinitely extended or periodically repeated multilayer with varying interlayer spacing and wave vector.

PACS 61.20.Gy - Statistical theories of liquid structure.

1. - Introduction.

Systems of electrons with two-dimensional dynamics have long been useful as models for a variety of physical systems such as inversion layers in semiconductors and semiconductor heterostructures [1, 2], surface electrons on liquid He [3], intercalated graphite [4] and transition-metal dichalcogenides [5]. The thermodynamic states of physical interest may range from extreme degeneracy to quasi-classical, and the electronic system may be confined to a single layer or form a multiplicity of layers up to a periodic stacking. While most of the theoretical treatments in the literature have taken account of intralayer correlations, specific attention has recently been brought to the role of the interlayer correlations in two-layer structures in relation to Wigner crystallization [6] and collective excitations [7].

Correlations in both homogeneous and inhomogeneous Coulomb fluids have a number of exactly determinable asymptotic properties, which may be conveniently expressed in the form of sum rules arising as consistency relations imposed by the long-range Coulomb interactions [8]. In particular, Alastuey and Martin [9] have shown that correlations in a two-dimensional classical plasma with e^2/r interactions have an algebraic r^{-3} decay as an exact lower bound. In the present work we extend their treatment to a system consisting of an arbitrary number N_p of equispaced layers confining a classical one-component plasma with e^2/r interactions. In view of the known differences in the asymptotic behaviours of correlations in the classical and quantal three-dimensional plasma [10, 11] we do not expect that our results should be generally applicable without further analysis to layered systems of electrons in the quantal regime.

2. – Equilibrium equations and multipolar sum rules.

The model consists of a stack of N_p layers with interlayer spacing d, each layer having area S and containing a classical fluid of N point-like charges embedded in a uniform neutralizing background. The normal to the stack is taken along the z-direction and the z-coordinate of each layer is denoted by md with m an integer. All vectors are decomposed into their in-plane and z components, with the notation $R = (\boldsymbol{r}, z)$ and $K = (\boldsymbol{k}, k_z)$. The medium has a uniform dielectric constant equal to unity, so that image forces are absent and the particles interact via the potential $e^2\phi(|R|) = e^2(r^2+z^2)^{-1/2}$, with Fourier transform $\tilde{\phi}(k, z) = (2\pi/k)\exp[-k|z|]$.

The usual assumption is made that in the thermodynamic limit ($N \to \infty$ and $S \to \infty$ at fixed average density $\rho = N/S$) the instantaneous density correlation functions exist and continue to obey the equilibrium equations of the Born-Green-Yvon (BGY) hierarchy. Starting from the n-body density distribution functions,

$$\rho(R_1, \ldots, R_n) = \langle [N(R_1)\ldots N(R_n)]_{\mathrm{SL}} \rangle, \tag{2.1}$$

where $N(R)$ is the particle density operator and the suffix SL indicates that the self-terms are omitted, we introduce [8] the density of excess particles at R when n particles are fixed at $R_1, \ldots, R_n$ as

$$\rho_{\mathrm{e}}(R|R_1, \ldots R_n) = \rho(R, R_1, \ldots, R_n) - \rho(R)\rho(R_1, \ldots, R_n) + \sum_{i=1}^{n} \delta(R-R_i)\rho(R_1, \ldots, R_n). \tag{2.2}$$

Furthermore, denoting by $Q = (R_2, \ldots, R_n)$ the positions of a set of $(n-1)$ particles we define the truncated n-body and $(n+1)$-body correlation functions by

$$\rho_{\mathrm{T}}(R_1, Q) = \rho(R_1, Q) - \rho(R_1)\rho(Q) \tag{2.3}$$

and

$$\rho_{\mathrm{T}}(R, R_1, Q) = \rho(R, R_1, Q) - \rho(R)\rho_{\mathrm{T}}(R_1, Q) - \rho(R_1)\rho_{\mathrm{T}}(R, Q) - \rho(Q)\rho(R, R_1). \tag{2.4}$$

The equilibrium equations of the BGY hierarchy can then be written as

$$(\beta e^2)^{-1}\nabla_{r_1}\rho_{\mathrm{T}}(R_1, Q) = \rho(R_1)\boldsymbol{E}_{\parallel}(R_1|Q) + \sum_{i=2}^{n} \boldsymbol{F}_{\parallel}(R_1-R_i)\rho_{\mathrm{T}}(R_1, Q) + \sum_m \int \mathrm{d}\boldsymbol{r}_m \boldsymbol{F}_{\parallel}(R_1-R_m)\rho_{\mathrm{T}}(R_m, R_1, Q), \tag{2.5}$$

where $\beta = (k_{\mathrm{B}}T)^{-1}$ and R_m denotes the position of a particle in the m-th layer. In eq. (2.5) we have defined $\boldsymbol{F}_{\parallel}(R) = -\nabla_r\phi(|R|)$ and introduced the electric field $\boldsymbol{E}_{\parallel}(R_1|Q)$

generated at R_1 when $(n-1)$ particles are at positions Q,

$$E_{\parallel}(R_1\,|\,Q) = \sum_m \int \mathrm{d}\boldsymbol{r}_m \boldsymbol{F}_{\parallel}(R_1 - R_m)\rho_{\mathrm{e}}(R_m\,|\,Q)\,. \tag{2.6}$$

The absolute convergence of the integral in eq. (2.6) and of the last integral on the right-hand side of eq. (2.5) requires that the correlations between a particle and any set of other particles should vanish as the particle is moved to infinite distance. The appropriate clustering condition is

$$|\rho(R_m, R_1, Q) - \rho(R_m)\rho(R_1, Q)| \leqslant M|\boldsymbol{r}_m|^{-\eta} \tag{2.7}$$

with M finite and $\eta > 0$. We also note that

$$\int \mathrm{d}\boldsymbol{r}_m \rho_{\mathrm{e}}(R_m\,|\,R_1, \ldots, R_n) = 0\,, \tag{2.8}$$

from the normalization condition relating the integral of the $(n+1)$-body distribution function to the n-body one. Both these properties will be taken to be valid to all orders in what follows.

Additional sum rules, relating to multipolar moments of correlations, can be shown to be valid [12, 13] if the decay of correlations is sufficiently rapid. Specifically, assume that the clustering conditions

$$|D^{\eta}\rho_{\mathrm{T}}(R_1, \ldots, R_n)| \leqslant M < \infty\,, \qquad D = \sup_{i,j}(|R_i - R_j|) \tag{2.9}$$

hold for $n = 2, \ldots, n_0 + 1$ and $\eta > 2 + l_0$ (for N_p finite) or $\eta > 3 + l_0$ (for $N_p \to \infty$). Then the (l, n) multipolar sum rules,

$$\sum_m \int \mathrm{d}\boldsymbol{r}_m \rho_{\mathrm{e}}(R_m\,|\,Q)\left[(R_m\cdot\nabla)^l \frac{R\cdot\hat{u}}{R^3}\right]_{R=\hat{u}} = 0\,, \tag{2.10}$$

where $\hat{u}$ is a unit vector in the plane of the layers, hold for $0 \leqslant l \leqslant l_0$ and $1 \leqslant n \leqslant n_0$. Equation (2.8) ensures that the charge sum rules (eq. (2.10) for $l = 0$) are always valid. If $l_0 = 1$ the dipole sum rules

$$\sum_m \int \mathrm{d}\boldsymbol{r}_m \boldsymbol{r}_m \rho_{\mathrm{e}}(R_m\,|\,Q) = 0 \tag{2.11}$$

also hold for the correlation functions up to the n_0-body one.

Proposition (2.9), (2.10) follows from studying the asymptotic behaviour of the BGY equations for $R_1 = (\lambda\hat{u}, 0)$ with $\lambda \to \infty$. An integration over the area A of a circle $C(\lambda\hat{u}, r_0)$ centred in R_1 and of given radius r_0 is first carried out to handle the gradient terms, yielding in particular

$$\int_A \mathrm{d}\boldsymbol{r}_1 \nabla_{\boldsymbol{r}_1} \rho_{\mathrm{T}}(R_1, Q) = \int_C \mathrm{d}\boldsymbol{y}\, \rho_{\mathrm{T}}(\lambda\hat{u} + \boldsymbol{y}, Q) = O(\lambda^{-\eta}) \tag{2.12}$$

for the term on the left-hand side of eq. (2.5). It is easily seen that the second term on the right-hand side decays faster than $\lambda^{-\eta}$, while it shown in appendix A that the third term decays faster than $\lambda^{-(l_0+2)}$ irrespectively of the number of layers. Hence, the electric potential $\Phi(R_1\,|\,Q)$ associated with the field $\boldsymbol{E}_{\parallel}(R_1\,|\,Q)$

must also decay faster than $\lambda^{-(l_0+2)}$. Comparison with a multipolar expansion yields eq. (2.10) for all $l \leqslant l_0$.

We shall focus in the following sections on the two-body correlation function. It is therefore useful to show at this point the form taken by the above general formalism in this case. We have

$$\rho_{\mathrm{T}}(R_m, R'_{m'}) = \rho(R_m)\rho(R'_{m'})[g(R_m, R'_{m'}) - 1] \tag{2.13}$$

and

$$\rho_{\mathrm{e}}(R_m | R'_{m'}) = \rho_{\mathrm{T}}(R_m, R'_{m'}) + \rho(R_m)\delta(R_m - R'_{m'}), \tag{2.14}$$

the first particle being in the m-th layer and the second in the m'-th layer, and $g(R, R')$ being the usual pair distribution function. Equation (2.8) yields

$$\int \mathrm{d}\boldsymbol{r}_m \rho_{\mathrm{T}}(R_m, R'_{m'}) = -\rho\,\delta_{m,m'}, \tag{2.15}$$

which may be viewed as a set of charge sum rules holding layer by layer. Finally, the appropriate BGY equilibrium equation involves the three-body correlation function,

$$(\beta e^2)^{-1}\nabla_{r_m}\rho_{\mathrm{T}}(R_m, R'_{m'}) = \rho(R_m)\boldsymbol{E}_{\parallel}(R_m | R'_{m'}) + \boldsymbol{F}_{\parallel}(R_m - R'_{m'})\rho_{\mathrm{T}}(R_m, R'_{m'}) + \\ + \sum_{m''}\int \mathrm{d}\boldsymbol{r}_{m''}\boldsymbol{F}_{\parallel}(R_m - R''_{m''})\rho_{\mathrm{T}}(R''_{m''}, R_m, R'_{m'}). \tag{2.16}$$

The electric field $\boldsymbol{E}_{\parallel}$ entering eq. (2.16) is to be determined from the Poisson equation. It should also be remarked that in the limit $N_p \to \infty$, according to the proof of proposition (2.9), (2.10) given above and in appendix A, an algebraic decay of two-body correlations implies a slower algebraic decay of the electric field. Therefore, an algebraic decay would not be compatible with eq. (2.16) if the three-body correlations were to decay more rapidly than the two-body ones.

For a homogeneous fluid confined to a single layer Alastuey and Martin [9] have shown that under appropriate clustering conditions on the two, three- and four-body correlation functions the structure factor $S(k)$ at long wavelengths is related to the interaction potential by

$$\lim_{k\to 0} S(k) = [\rho\beta e^2\tilde{\phi}(k, 0)]^{-1} = k/k_{\mathrm{D}} \tag{2.17}$$

with $k_{\mathrm{D}} = 2\pi\rho\beta e^2$. However, such a behaviour of $S(k)$ implies that the asymptotic form of the pair correlations contains a term behaving like r^{-3}, which contradicts the assumed validity of the clustering conditions. While the charge sum rule suffices to ensure that $S(k)$ vanishes for $k \to 0$, a dipole moment arising from the three-body correlation function must supplement the k_{D}^{-1} term in determining the value of $S(k)/k$ for $k \to 0$.

Taking $S(k)/k$ as a finite constant for $k \to 0$ and bearing in mind the possibility of other singularities arising at finite k, the conclusion is that the pair correlations cannot decay asymptotically faster than r^{-3}. According to the BGY equation for the pair correlation function, the excess potential $\Phi(r, z)$ associated with the field $\boldsymbol{E}(r, z)$

then cannot decay faster than r^{-3} for $z=0$. In fact, from the Poisson equation

$$\frac{1}{r}\frac{\partial}{\partial r}[rE_{\parallel}(r,z)]+\frac{\partial}{\partial z}E_z(r,z)=4\pi\delta(z)\rho_e(r|0) \tag{2.18}$$

and assuming only the charge sum rule, the first two terms of a multipolar expansion for $\Phi(r,z)$ have the form $P_l(|\cos\theta|)/|R|^{(l+1)}$ where $l=1$ or 2, $\cos\theta=z/|R|$ and $P_l(x)$ are the Legendre polynomials. Using $P_1(0)=0$ and $P_1(1)=1$ one sees that $\Phi(r,0)$ decays like r^{-3} and $\Phi(0,z)$ decays like $|z|^{-2}$. These behaviours were derived in early work by Fetter [14] within a hydrodynamic approach, which reduces in the static case to the Debye-Hückel approximation and thus assumes that the relation in eq. (2.17) is valid. The magnitude of the dipole moment associated with the pair correlation function is given in this approximation by the Debye screening length $1/k_D$.

The important point to be stressed is that for a monolayer, at variance from the case of the three-dimensional classical plasma, an algebraic decay of correlations and an algebraic decay of the potential are mutually consistent. We carry out below the same analysis for a multilayered system.

3. – Asymptotic behaviour of correlations in a multilayered plasma: the case of finite N_p.

We have seen in sect. 2 that the type of clustering which ensures the validity of multipolar sum rules up to order (l_0, n_0) is independent of the number N_p of layers provided that N_p is finite. We examine in this section the asymptotic behaviour of the pair correlations in this case. The limit $N_p\to\infty$ will be discussed in the next section.

As a first step we rewrite the BGY eq. (2.16) as

$$(\beta e^2)^{-1}\nabla_r\rho_T(R_m,0)=\rho\boldsymbol{E}_{\parallel}(R_m|0)+\boldsymbol{W}_m(\boldsymbol{r}), \tag{3.1}$$

where one of the particles has been taken at the origin and we have defined

$$\boldsymbol{W}_m(\boldsymbol{r})=\sum_{m'}\int d\boldsymbol{r}'\,\boldsymbol{F}_{\parallel}(R'_{m'})H(R_m,R'_{m'}) \tag{3.2}$$

with

$$H(R_m,R'_{m'})=\rho_T(R_m,R'_{m'},0)+\delta(\boldsymbol{r}-\boldsymbol{r}')\delta_{mm'}\rho_T(R_m,0). \tag{3.3}$$

Use has been made of the symmetry properties of the three-body correlation function.

We introduce the structure factor $S(K)$ as

$$S(K)=\sum_m S_m(k)\exp[-ik_z md], \tag{3.4}$$

where

$$S_m(k)=\delta_{m0}+\rho^{-1}\int d\boldsymbol{r}\exp[-i\boldsymbol{k}\cdot\boldsymbol{r}]\rho_T(R_m,0) \tag{3.5}$$

are the partial structure factors describing intralayer ($m=0$) and interlayer ($m\neq 0$) correlations. Using eq. (2.6) for the excess electric field, the Fourier transform of eq. (3.1) then is

$$S_m(k) - \delta_{m0} = -(k_D/k)\sum_{m'} \exp[-|m-m'|kd] S_{m'}(k) + k_D \Delta_m(k), \tag{3.6}$$

where

$$\Delta_m(k) = -i(2\pi\rho^2 k^2)^{-1} \sum_{m'} \int d\boldsymbol{r} \exp[-i\boldsymbol{k}\cdot\boldsymbol{r}] \int d\boldsymbol{r}'\, \boldsymbol{k}\cdot\boldsymbol{F}_{\parallel}(R'_{m'}) H(R_m, R'_{m'}). \tag{3.7}$$

Hence,

$$S_m(k)/k = \sum_{m'} A^{-1}_{mm'}(k)\left[\frac{\delta_{m'0}}{k_D} + \Delta_{m'}(k)\right], \tag{3.8}$$

where $A^{-1}_{mm'}(k)$ is the inverse of a matrix $A_{mm'}(k)$ which is defined by

$$A_{mm'}(k) = (k/k_D)\delta_{mm'} + \exp[-|m-m'|kd]. \tag{3.9}$$

The charge sum rule (2.15) yields

$$\lim_{k\to 0} S_m(k) = 0 \tag{3.10}$$

for all values of m. Using it in eq. (3.6) we find

$$\lim_{k\to 0} \sum_{m'} \frac{S_{m'}(k)}{k} = \frac{\delta_{m0}}{k_D} + \lim_{k\to 0} \Delta_m(k). \tag{3.11}$$

Clearly, the quantity on the right-hand side of this equation must be independent of the index m. We denote it thereafter by the symbol Λ. Namely,

$$\Lambda \equiv \lim_{k\to 0} \sum_m \frac{S_m(k)}{k} = \frac{1}{k_D} + \lim_{k\to 0} \Delta_0(k) = \lim_{k\to 0} \Delta_{m\neq 0}(k). \tag{3.12}$$

It is evident from eqs. (3.12) and (3.5) that Λ gives the length of the electric dipole moment associated with the total pair correlation function $\sum_m \rho_e(R_m|0)/\rho$. Equation (3.12) implies very strong correlations: we can obtain the dipole moment of the whole stack from a three-body correlation function involving a particle in *anyone* of the layers and the particle at the origin, provided that we add the quantity k_D^{-1} when the first particle lies in the same layer as the particle at the origin.

We can now examine the solution of eq. (3.6) in the long-wavelength limit. Using eq. (3.12) in eq. (3.8) we have

$$\lim_{k\to 0} \frac{S_m(k)}{k} = \alpha_m \Lambda, \tag{3.13}$$

where the coefficients α_m are given by

$$\alpha_m = \lim_{k\to 0} \sum_{m'} A^{-1}_{mm'}(k) \tag{3.14}$$

and satisfy the sum rule $\sum_m \alpha_m = 1$. For instance, for a bilayer $\alpha_m = 1/2$, while for a trilayer we find

$$\alpha_1 = \alpha_{-1} = \frac{1 + 3\,dk_D + 2(dk_D)^2}{3 + 8\,dk_D + 4(dk_D)^2} \tag{3.15}$$

and

$$\alpha_0 = \frac{1 + 2\,dk_D}{3 + 8\,dk_D + 4(dk_D)^2}\,. \tag{3.16}$$

We see that the partitioning of the total dipole moment Λ among the various layers is exactly known from eq. (3.13) and (3.14). The coefficients α_m are functions of dk_D which depend only on the number of layers.

The values taken by the quantities $\Delta_m(k)$ at long wavelengths remain to be discussed. It is evident from eq. (3.12) that they cannot be all equal to zero. A more formal argument, relating the behaviour of $\sum_m \Delta_m(k \to 0)$ to the clustering of correlation functions, is given in appendix B. The result is that, if $\Lambda \neq 0$, the intralayer and interlayer pair correlation functions cannot decay asymptotically faster than r^{-3}.

The discussion given in appendix B does not exclude the possibility $\Lambda = 0$. This would imply $\Delta_m(k \to 0) = 0$ for all $m \neq 0$ and $\Delta_0(k \to 0) = -k_D^{-1}$. Evidently, the linear term in the low-k expansion of the intralayer and interlayer structure factors would then be absent and the leading term would presumably have a regular k^2 behaviour, completely invalidating a Debye-Hückel approximation. As is shown in appendix B, in such a case a slow asymptotic decay would still be present in the three-body and/or four-body correlation functions.

4. – Asymptotic behaviour of correlations in the limit $N_p \to \infty$.

We return to eqs. (3.6) and (3.7), in which we have to take the limit $N_p \to \infty$ in the sums over the layer index m'. We first take Fourier transforms with respect to the z-coordinate, by multiplying both sides of eq. (3.6) by $\exp[-ik_z md]$ and summing over the layer index m. In the limit $N_p \to \infty$ we find

$$S(K) - 1 = -(k_D/k) \sum_{m=-\infty}^{\infty} \Gamma_m(K) S_m(K) + k_D \Delta(K)\,, \tag{4.1}$$

where

$$\Gamma_m(K) = \exp[-ik_z md] \sum_{m'=-\infty}^{\infty} \exp[-ik_z m'd - k|m'|d] = \exp[-ik_z md] \frac{\sinh(kd)}{\cosh(kd) - \cos(k_z d)} \tag{4.2}$$

and

$$\Delta(K) = -i(2\pi\rho^2 k^2)^{-1} \sum_{m'=-\infty}^{\infty} \int d\boldsymbol{r}'\, \boldsymbol{k}\cdot\boldsymbol{F}_{\parallel}(R'_{m'}) \cdot \sum_{m=-\infty}^{\infty} \exp[-ik_z md] \int d\boldsymbol{r} \exp[-i\boldsymbol{k}\cdot\boldsymbol{r}] H(R_m, R'_{m'})\,. \tag{4.3}$$

We have assumed that the two integrals in eq. (3.7) can be interchanged (see appendix B). The sum over m in eq. (4.1) can now be carried out, with the result

$$S(K) - 1 = -\rho\beta v(K) S(K) + k_D \Delta(K), \tag{4.4}$$

where [15]

$$v(K) = \frac{2\pi e^2}{k} \frac{\sinh(kd)}{\cosh(kd) - \cos(k_z d)}. \tag{4.5}$$

We may remark that the same result (4.4) is obtained when, instead of taking the limit $N_p \to \infty$, one imposes periodic boundary conditions along the z-direction on a stack of N_p layers. In this case $S(K)$ and $\Delta(K)$ are the sums of $S_m(\boldsymbol{k}, k_z)$ and of $\Delta_m(\boldsymbol{k}, k_z)$ over the layers included in the Born-von Karman periodicity cell.

The effective potential $v(K)$ in eq. (4.5) shows dimensional crossover with varying d, tending to $2\pi e^2/k$ in the limit $d \to \infty$ (an infinite stack of independent monolayers) and to $4\pi e^2/(K^2 d)$ in the limit $d \to 0$ (a three-dimensional plasma with mean particle density ρ/d and two-dimensional dynamics). In the latter limit the Poisson equation becomes a local differential equation and one can apply the argument developed by Martin [8] to analyse the clustering of correlations in a fully three-dimensional plasma. In brief, if one assumes an algebraic decay of the total charge density, the Poisson equation yields a slower algebraic decay of the total electric field. This result is not compatible with the asymptotic behaviour of the BGY equation for the pair correlation function, leading to the conclusion that correlations must asymptotically decay more rapidly than any finite inverse power of the distance.

Expression (4.5) for $v(K)$ yields $v(K) \to 4\pi e^2/(k^2 d)$ in the limit $k \to 0$ at $k_z = 0$, for any finite value of the layer spacing d. Equation (4.4) yields

$$\lim_{k \to 0} \frac{2}{d} \frac{S(k, 0)}{k^2} = \frac{1}{k_D} + \lim_{k \to 0} \Delta(k, 0). \tag{4.6}$$

This relation should be contrasted with the analogous relation which can easily be obtained for the case of a finite number of layers from the results in sect. 3,

$$\lim_{k \to 0} N_p \frac{S(k, 0)}{k} = \frac{1}{k_D} + \lim_{k \to 0} \Delta(k, 0). \tag{4.7}$$

The charge sum rules suffice to ensure that the last term on the right-hand side of eq. (4.6) is at most a finite constant, so that $S(k, 0)$ is proportional to k^2 in the limit $kd \ll 1$. Such an analytic behaviour of $S(k, 0)$ at the origin precludes the possibility of drawing conclusions on the existence of algebraic terms in the asymptotic behaviour of the pair correlations. If in addition the dipole sum rule holds for the three-body correlation function $\sum_m H(R_m, R'_{m'})$ in eq. (4.3), then $\Delta(k \to 0, 0)$ vanishes and the further sum rule

$$\lim_{k \to 0} \frac{S(k, 0)}{k^2} = L_D^2 \tag{4.8}$$

holds. Here,

$$L_D = (4\pi\rho\beta e^2/d)^{-1/2} \tag{4.9}$$

is the three-dimensional Debye screening length. Equation (4.8), which may also be written as the integral relation

$$\sum_{m=-\infty}^{\infty} \rho \int \mathrm{d}\boldsymbol{r}\, r^2 [g(r, md) - 1] = -4L_{\mathrm{D}}^2 \tag{4.10}$$

on the total pair correlation function, is the form presently taken by the Stillinger-Lovett sum rule [16, 17]. It ensures that the plasma is capable of screening completely any static distribution of external charges having spatial dependence of the form $\rho_{\mathrm{ext}}(\boldsymbol{r})$.

The partial structure factors $S_m(k)$ are related to the total structure factor $S(K)$ by

$$S_m(k) = \frac{2\pi}{d} \int_{-\pi/d}^{\pi/d} \mathrm{d}k_z \exp[ik_z md] S(K), \tag{4.11}$$

thus requiring full knowledge of the k_z dependence of $S(K)$ even in the limit $k \to 0$. The multipolar sum rules provide no information on the behaviour of $\Delta(0, k_z)$. On the assumption that $\Delta(K)$ can be neglected, Fetter [15] has solved eqs. (4.4) and (4.11) in conjunction with the Poisson equation. Within this approximation he has shown that the partial pair correlation function $\rho_{\mathrm{T}}(r, z)$ decays exponentially both as a function of r at fixed z and as a function of z at fixed r, such a decay being anisotropic except in the limit $dk_{\mathrm{D}} \ll 1$.

4.1. *Dynamical implications.* – We next wish to point out how the foregoing discussion may be related to the dynamics of the classical layered plasma at long wavelengths. The effective potential $v(K)$ determines a characteristic frequency $\omega_0(K)$ given by

$$\omega_0^2(K) = \frac{\rho k^2}{M} v(K), \tag{4.12}$$

where M is the mass of the particles. In the limit $k \to 0$ the dispersion relation (4.12) describes an optic mode at $k_z = 0$ and an acoustic one at $k_z \neq 0$. The hydrodynamic treatment given by Fetter [15] leads to a collective mode with a dispersion relation given by (4.12) supplemented by a k^2 term with a coefficient determined by the adiabatic free-gas speed of sound. Olego *et al.* [18] have found that the dispersion relation (4.12) is in good agreement with the results of their inelastic light scattering experiments from GaAs-(AlGa)As heterostructures.

A simple connection between structure and dynamics can be made on the assumption that the *f*-sum rule on the dynamic structure factor $S(K; \omega)$,

$$\int_{-\infty}^{\infty} \frac{\mathrm{d}\omega}{2\pi} \omega^2 S(K; \omega) = \frac{k^2}{\beta M}, \tag{4.13}$$

is exhausted by a single collective mode. Since $S(K)$ is the integral of $S(K; \omega)$ over frequency, this would lead one to express $S(K)$ in terms of the collective-mode frequency as

$$S(K) = \frac{k^2}{M\beta\omega_0^2(K)} . \tag{4.14}$$

Comparison of eq. (4.14) with eq. (4.4) shows that such a single-mode representation of the spectrum is allowed in the limit $k \to 0$ only if $k_z = 0$, where $\Delta(k \to 0, 0)$ vanishes. In fact, the work of Totsuji[19] on the dynamics of a classical two-dimensional plasma shows that excitational electron-electron collisions give a relevant spectral contribution at long wavelengths (a Landau-type contribution associated with single-particle excitations is exponentially small in this limit). The collisional damping of the collective mode as calculated by Totsuji is linear in k for $k \to 0$, *i.e.* of the same order as the frequency of the acoustic mode. It thus appears that a single-mode representation of the spectrum at $k_z \neq 0$ is invalid for a layered classical plasma.

5. – Summary and concluding remarks.

In this work we have applied to a layered classical plasma methods of analysis previously developed to examine the asymptotic behaviours of the correlation functions in Coulomb fluids and the sum rules that are consistent with these behaviours. Our main results concern the conditions for the validity of multipolar sum rules, the dipolar structure of a finite multilayer and the dimensionality crossover in an infinitely extended (or periodically repeated) multilayer with varying interlayer spacing and wave vector.

The theoretical possibility of crystalline order is notoriously related to poor clustering of particle correlations and has drawn considerable attention for Coulomb systems in low dimensionalities. In particular, from an analysis of the BGY hierarchy for a monolayer Gruber and Martin[20] have shown that the pair or three-body correlation functions should decay asymptotically more rapidly than r^{-3} in order to exclude crystallinity. Their analysis is easily extended to a finite multilayer, leading to the same conclusion. However, Requardt and Wagner[21] have recently been able to obtain more stringent conditions through the use of the Mermin inequality for a variety of Coulomb systems including the monolayer with r^{-1} interactions.

* * *

Sponsorship and support by the Istituto Nazionale di Fisica della Materia is gratefully acknowledged.

Appendix A

Asymptotic behaviour of the $(n+1)$-body term in the BGY equations for layered plasmas.

We prove in this appendix that the $(n+1)$-body term in eq. (2.5), under the clustering condition (2.9), decays faster than $\lambda^{-(l_0+2)}$ when $R_1 = (\lambda\hat{u}, 0)$ with $\lambda \to \infty$, irrespectively of the number N_p of layers. Since the group of particles at Q is kept fixed in taking the limit, it is sufficient to examine the behaviour of the three-body term.

In the indicated limit we can write the following inequality:

$$\left| \sum_m \int \mathrm{d}\boldsymbol{r}_m \boldsymbol{F}_\parallel(R_m) \rho_\mathrm{T}(\lambda\hat{u}, R_m, 0) \right| \leqslant$$

$$\leqslant \sum_{m \neq 0} \int \mathrm{d}\boldsymbol{r}_m \frac{r_m}{|R_m|^3} \frac{M}{[\sup(\lambda, |R_m|)]^\eta} + \int \mathrm{d}\boldsymbol{r}_m r_m^{-2} |\rho_\mathrm{T}(\lambda\hat{u}, \boldsymbol{r}_m, 0)| ,$$

where, from the results of Gruber *et al.* [12], the second term on the right-hand side decays like $\lambda^{-\eta}$. Hence,

$$\text{(A.1)} \qquad \left| \sum_m \int \mathrm{d}\boldsymbol{r}_m \boldsymbol{F}_\parallel(R_m) \rho_\mathrm{T}(\lambda\hat{u}, R_m, 0) \right| \leqslant \sum_{m \neq 0} \int_{r \geqslant |m| d} \mathrm{d}\boldsymbol{r}\, r^{-2} \frac{M}{[\sup(\lambda, r)]^\eta} + O(\lambda^{-\eta}).$$

In the case where N_p is finite, it is evident that the first term on the right-hand side of eq. (A.1) also decays like $\lambda^{-\eta}$, *i.e.* faster than $\lambda^{-(l_0+2)}$ if $\eta > l_0 + 2$.

In the case $N_p \to \infty$, on the other hand, the above term can be rewritten as

$$2 \sum_{m=1}^{\infty} \int_{r \geqslant md} \mathrm{d}\boldsymbol{r}\, r^{-2} \frac{M}{[\sup(\lambda, r)]^\eta} =$$

$$= 4\pi \left\{ \sum_{m=1}^{[\lambda/d]} \left[\lambda^{-\eta} \int_{md}^{\lambda} r^{-1} \mathrm{d}r + \int_{\lambda}^{\infty} r^{-(1+\eta)} \mathrm{d}r \right] + \sum_{m=[\lambda/d]+1}^{\infty} \int_{md}^{\infty} r^{-(1+\eta)} \mathrm{d}r \right\}.$$

For $\lambda \to \infty$ we have

$$\sum_m f(md/\lambda) = \frac{\lambda}{d} \sum_n f(n)\, \Delta n \to \frac{\lambda}{d} \int f(x)\, \mathrm{d}x$$

with $n = md/\lambda$ and $\Delta n = d/\lambda \to 0$, if $f(x)$ does not change appreciably with x in the range Δn. Hence, in the case $N_p \to \infty$ the $(n+1)$-body term decays faster than $\lambda^{-(l_0+2)}$ if $\eta > l_0 + 3$.

Appendix B

Clustering of correlations and behaviour of $\Delta_m(k)$ at long wavelengths.

Starting from the definition of $\Delta_m(k)$ in eq. (3.7), we first prove that $\sum_m \Delta_m(k \to 0) = 0$ if i) the clustering condition (2.9) holds for $\eta > 3$ and $n = 2$, 3 and 4; and ii) for sufficiently large $|x|$ we have $|x|^\eta \int \mathrm{d}y\, \rho_\mathrm{T}(xy0) \leqslant M < \infty$, x and y being any two position coordinates in the layers.

Following the line of argument given by Alastuey and Martin [9] for a similar proposition regarding a monolayer, we use the condition ii) above to interchange the order of the two integrals in eq. (3.7). We can then write

$$\text{(B.1)}\qquad \Delta_m(k) = -i(2\pi\rho^2k^2)^{-1}\sum_{m'}\int \mathrm{d}\boldsymbol{r}'\ \boldsymbol{k}\cdot\boldsymbol{F}_{\parallel}(R'_{m'})\int \mathrm{d}\boldsymbol{r}\exp[-i\boldsymbol{k}\cdot\boldsymbol{r}]H'(R_m, R'_{m'}),$$

where

$$\text{(B.2)}\qquad H'(R_m, R'_{m'}) = H(R_m, R'_{m'}) + \delta_{m0}\delta(\boldsymbol{r})\rho_{\mathrm{T}}(R'_{m'}, 0) =$$
$$= \rho_{\mathrm{e}}(R_m\,|R'_{m'}, 0) - \rho\rho_{\mathrm{e}}(R_m\,|R'_{m'}) - \rho\rho_{\mathrm{e}}(R_m\,|0).$$

The difference between H' and H in eq. (B.2) does not contribute to the integral in eq. (B.1) and has been included so that we may make use of the multipolar sum rules given in eq. (2.10). After expanding the factor $\exp[-i\boldsymbol{k}\cdot\boldsymbol{r}]$ in eq. (B.1) and using the charge sum rules, we find

$$\text{(B.3)}\qquad \sum_m \Delta_m(k\to 0) = -i(2\pi\rho^2)^{-1}\sum_{m'}\int \mathrm{d}\boldsymbol{r}'\ \hat{\boldsymbol{k}}\cdot\boldsymbol{F}_{\parallel}(R'_{m'})\sum_m\int \mathrm{d}\boldsymbol{r}\,\hat{\boldsymbol{k}}\cdot\boldsymbol{r}H'(R_m, R'_{m'}) + o(1).$$

However, under the condition i) the dipolar sum rule holds for both two-body and three-body correlation functions, so that the first term on the right-hand side of eq. (B.3) vanishes.

By summing eq. (3.11) over all layers we then find that

$$\text{(B.4)}\qquad \Lambda \equiv \lim_{k\to 0}\frac{\sum_m S_m(k)}{k} = (N_p k_{\mathrm{D}})^{-1} + o(1)$$

under the same conditions i) and ii) stated above. It would be natural to assume this result in a Debye-Hückel treatment, its implication being that the dipole moment as read from the large-z behaviour of the electric potential created by a stack of N_p layers is that of a monolayer with particle density $N_p\rho$. However, it follows from eq. (B.4) that the asymptotic form of the total pair correlation function $\sum_m \rho_{\mathrm{T}}(R_m, 0)$ would contain a term behaving like r^{-3}. We have thus reached a contradiction: the clustering condition i) must hold for the validity of eq. (B.4), but eq. (B.4) refutes the validity of condition i).

We therefore conclude that

$$\text{(B.5)}\qquad \lim_{k\to 0}\sum_m \Delta_m(k) = O(1)$$

and that the clustering cannot be faster than r^{-3} for at least one among the two-, three- and four-body correlation functions. It would seem reasonable to expect that the higher-body correlations should not decay more slowly than the two-body ones, leading to the conclusion that the asymptotic decay of the total pair correlation function should not be faster than r^{-3}. In such a case, $\Lambda \neq 0$ and the discussion given in the main text shows that each one of the partial pair correlations cannot decay faster than r^{-3}. However, it seems to us that the possibility $\Lambda = 0$, implying slow decay of correlations at higher order, cannot be excluded.

REFERENCES

[1] Stern F., *Phys. Rev. Lett.*, **18** (1967) 546; **30** (1973) 278.
[2] See, for instance, Butcher P., March N. H. and Tosi M. P. (Editors), *Physics of Low-Dimensional Semiconductor Structures* (Plenum, New York, N.Y.) 1993.
[3] Brown T. R. and Grimes C. C., *Phys. Rev. Lett.*, **29** (1972) 1233.
[4] Visscher P. B. and Falicov L. M., *Phys. Rev. B*, **3** (1971) 2541.
[5] Gamble F. R., DiSalvo F. L., Klemm R. A. and Geballe T. H., *Science*, **168** (1970) 568.
[6] Swierkowski L., Neilson D. and Szymanski J., *Phys. Rev. Lett.*, **67** (1991) 241.
[7] Neilson D., Swierkowski L., Szymanski J. and Liu L., *Phys. Rev. Lett.*, **71** (1993) 4035.
[8] Martin P. A., *Rev. Mod. Phys.*, **60** (1988) 1075.
[9] Alastuey A. and Martin P. A., *J. Stat. Phys.*, **39** (1985) 405.
[10] Alastuey A. and Martin P. A., *Europhys. Lett.*, **6** (1988) 385.
[11] Cornu F. and Martin P. A., *Phys. Rev. A*, **44** (1991) 4893.
[12] Gruber C., Lebowitz J. L. and Martin P. A., *J. Chem. Phys.*, **75** (1981) 944.
[13] Blum L., Gruber C., Lebowitz J. L. and Martin P. A., *Phys. Rev. Lett.*, 48 (1982) 1769.
[14] Fetter A. L., *Ann. Phys. (N.Y.)*, 88 (1974) 1.
[15] Fetter A. L., *Ann. Phys. (N.Y.)*, 81 (1973) 367.
[16] Stillinger F. H. and Lovett R., *J. Chem. Phys.*, 48 (1968) 385; 49 (1968) 1991.
[17] Carnie S. L. and Chan D. Y. C., *Chem. Phys. Lett.*, 77 (1981) 437; Carnie S. L., *J. Chem. Phys.*, 78 (1983) 2742.
[18] Olego D., Pinczuk A., Gossard A. C. and Wiegmann W., *Phys. Rev. B*, **25** (1982) 7867.
[19] Totsuji H., *J. Phys. Soc. Jpn.*, **40** (1976) 857.
[20] Gruber C. and Martin P. A., *Ann. Phys. (N.Y.)*, **131** (1981) 56.
[21] Requardt M. and Wagner H. J., *J. Stat. Phys.*, 58 (1990) 1165.