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A New Proof of the Stillinger–Lovett Complete Shielding Condition

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It is shown that any equilibrium state of classical charged particles with correlation having a spatial decay faster than $1/|x|^{\nu+2}$ in dimension $\nu = 2, 3$ obeys the Stillinger–Lovett second moment condition. Under the same clustering hypothesis, arbitrary localized external charge distributions are completely shielded.

KEY WORDS: Charged systems; BGY hierarchy; screening; spatial clustering; electrostatic sum rules.

1. INTRODUCTION

Much information of interest regarding the equilibrium properties of fluids of charged particles can be derived from the structure factor $\tilde{S}(k)$, the Fourier transform of $S(x)$, the truncated charge–charge correlation function. A fundamental property of $\tilde{S}(k)$ for charged systems is its definite short wave length behavior:

$$\lim_{|k|\to 0} \frac{\tilde{S}(k)}{|k|^2} = \frac{1}{\omega_\nu \beta} \tag{1.1}$$

which is equivalent to the sum rules

$$\int dx\, S(x) = 0 \tag{1.2}$$

$$\int dx\, x S(x) = 0 \tag{1.3}$$

$$\int dx\, |x|^2 S(x) = -\frac{2\nu}{\omega_\nu \beta} \tag{1.4}$$

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with $\omega_2 = 2\pi$, $\omega_3 = 4\pi$ in dimensions $\nu = 2$ and $\nu = 3$, respectively, and $\beta = (k_B T)^{-1}$ is the inverse temperature.

Equation (1.2) is the usual electroneutrality condition, whereas Eq. (1.3) is trivially satisfied in uniform states. The sum rule (1.4), called the second moment Stillinger–Lovett condition (S.L. condition), is equivalent with the property that the inverse dielectric constant vanishes. This follows from the well-known relation[1,2], [2]

$$\epsilon^{-1} = \lim_{|k|\to 0}\left[1 - \frac{\omega_\nu \beta}{|k|^2}\tilde{S}(k)\right] = 1 + \frac{\omega_\nu \beta}{2\nu}\int dx\, |x|^2 S(x) \tag{1.5}$$

$\epsilon = \infty$ expresses the fact that a charged system satisfying (1.4) behaves as a perfect conductor from the viewpoint of electrostatics. In other words, the S.L. condition characterizes a plasma phase, by opposition to a dielectric phase, where $1 \leqslant \epsilon < \infty$. It is therefore of interest to know precisely when the S.L. condition holds.

The purpose of this note is to present a simple first principle proof of (1.4), based on the only assumptions that the system is at thermal equilibrium and that its correlations have a spatial decay faster than $|x|^{-(\nu+2)}$ in dimension $\nu = 2, 3$ (see Proposition 1). We emphasize that the proof does not involve any additional assumption on the behavior of the direct correlation function or the dielectric function itself, but relies on some new sum rules which have recently been found to be true in Coulomb systems.[5–7]

Moreover, under essentially the same hypothesis, we have previously shown that any state of charged particles, in the absence of external forces, has to be invariant under translations and rotations.[8,9] We therefore obtain the general result that *classical Coulomb states which cluster faster than* $|x|^{-(\nu+2)}$ *are necessarily homogeneous, isotropic, and "perfect conductors."*

There are two instances where homogeneous phases of classical charged systems have been rigorously shown not to be of the plasma type. These are the Kösterlitz–Thouless phase of the two-dimensional low-temperature Coulomb gas[10] and the two-component one-dimensional Coulomb gas at all temperatures.[11] In both cases dipoles form spontaneously, owing to the confining nature of the Coulomb potential in one and two dimensions. The reasons for which our proof does not apply to these cases are the weak decay of the particle correlations in the Kösterlitz–

[2] Alternatively, Eq. (1.4) is a consequence of the assumption that the direct correlation function behaves as $-\beta(e_1 e_2/|x|)$ as $|x| \to \infty$ plus a short range contribution.[3,4] Eq. (1.4) has been rigorously shown from the cluster expansion in the high temperature phase[15] and follows in general from certain assumptions in the Sine–Gordon representation of the Coulomb gas[17].

Thouless phase (presumably like $|x|^{-\nu}$ as expected for pure dipole gases) and the peculiar nature of the Coulomb force in one dimension.

Crystalline phases (or phases with directional order) exhibit long-range order (i.e., weak clustering) in dimension $\nu \geqslant 2$,[9] and therefore we cannot draw any conclusion on the behavior of their dielectric functions from the present analysis.

In the Section 2, we define the general setting and recall the relevant sum rules expressing the screening properties in charged systems. We give the proof of the S.L. condition in Section 3 for the case of a homogeneous state. We extend our proof to mixtures of ions and dipoles in Section 4 and deduce the version of the S.L. condition appropriate to this situation. This relation has also been found in Ref. 12 from reasonable assumptions on the direct correlation functions.

In Section 5, we show that charges which are fractional with respect to the system's charges are shielded under the same hypothesis which are needed for the derivation of the S.L. condition. This point is of interest since fractional charges are precisely not screened in the Kösterlitz–Thouless phase and in one dimension.[10,11] On the other hand, the shielding of the system's charges occurs under weaker conditions (i.e., integrable clustering) and is expected to be generally true. Furthermore, these considerations enable us to discuss the S.L. condition for inhomogeneous systems.

The extension of these results to quantum systems will be given elsewhere.

2. GENERAL SETTING

Here and in the following section we will consider homogeneous and isotropic equilibrium states of charged particles in $\mathbb{R}^\nu$, $\nu \geqslant 2$. The particles of species α carries a charge e_α ($e_\alpha \neq 0$ for some species); we introduce the abbreviated notation $q = (\alpha, x)$ where x denotes the position of the particle, and $\int dq \cdots = \int_{\mathbb{R}^\nu} dx \sum_\alpha \cdots$.

The correlation functions $\rho(q_1), \rho(q_1 q_2)$ (singlet, doublet densities ...) describing the state of the system at temperature β^{-1} are assumed to be bounded differentiable functions satisfying the usual BGY hierarchy[13]:

$$\beta^{-1}\nabla_1 \rho(q_1) = \int dq\, F^S(q_1 q)\rho(q_1 q) + \int dq\, F^L(q_1 q)\left[\rho(q_1 q) - \rho(q_1)\rho(q)\right]$$
$$= 0 \tag{2.1}$$

$$\beta^{-1}\nabla_1 \rho(q_1 q_2) = \left[F^S(q_1 q_2) + F^L(q_1 q_2)\right]\rho(q_1 q_2) + \int dq\, F^S(q_1 q)\rho(q_1 q_2 q)$$
$$+ \int dq\, F^L(q_1 q)\left[\rho(q_1 q_2 q) - \rho(q)\rho(q_1 q_2)\right] \tag{2.2}$$

In (2.1) and (2.2), we have singled out the contribution of the Coulomb force:

$$
\begin{gathered}
F^L(q_1q_2) = -e_{\alpha_1}e_{\alpha_2}(\nabla_1\phi)(x_1 - x_2) \\
\phi(x) = \begin{cases} \dfrac{1}{|x|}, & \nu = 3 \\ -\ln|x|, & \nu = 2 \end{cases}
\end{gathered}
\tag{2.3}
$$

$F^S(q_1q_2) = F^S_{\alpha_1\alpha_2}(x_1 - x_2)$ is a short-range two-body force, covariant under rotations and antisymmetric under the exchange of particles, which includes the local repulsion effect needed for thermodynamic stability. Here F^S is assumed to be integrable on $\mathbb{R}^\nu$, but more singular repulsions or hard cores can be allowed (see comments at the end of Section 3). Because of the local neutrality, we have always (see comments in Section 3)

$$\sum_\alpha e_\alpha\rho_\alpha + \rho_B = 0$$

where ρ_α are the (constant) particle densities and ρ_B is a uniform external charge density characterizing "jellium" systems.

We introduce also the truncated (Ursell) functions defined in the usual way:

$$
\begin{gathered}
\rho^T(q_1q_2) = \rho(q_1q_2) - \rho(q_1)\rho(q_2) \\
\rho^T(q_1q_2q_3) = \rho(q_1q_2q_3) - \rho(q_1)\rho^T(q_1q_2) - \rho(q_2)\rho^T(q_1q_3) - \rho(q_3)\rho(q_1q_2)
\end{gathered}
\tag{2.4}
$$

Their rate of decay is specified by a parameter η such that

$$|r^\eta\rho^T(q_1 \ldots q_k)| \leqslant M_k < \infty \tag{2.5}$$

with $r = \sup_{i,j}|x_i - x_j|$, $i, j = 1 \ldots k$.

The screening properties of charged systems are conveniently expressed in terms of the excess particle density $\rho(q \,|\, q_1 \ldots q_n)$ when particles of type $\alpha_1 \ldots \alpha_n$ are fixed at $x_1 \ldots x_n$:

$$
\begin{gathered}
\rho(q \,|\, q_1 \ldots q_n) = \frac{\rho(qq_1 \ldots q_n)}{\rho(q_1 \ldots q_n)} + \sum_{i=1}^n \delta_{qq_i} - \rho(q) \\
\delta_{qq_i} = \delta_{\alpha\alpha_i}\delta(x - x_i)
\end{gathered}
\tag{2.6}
$$

It has been proven (see Refs. 6–8) that if the clustering condition (2.5) holds with $\eta > \nu + l$ for some nonnegative integer l and $k = 2, \ldots, n+2$, then the excess particle density $\rho(q \,|\, q_1 \ldots q_n)$ carries no multipole moments up to order l. In particular when $l = 0$ and $l = 1$, respectively, we have the charge sum rule

$$\int dq\, e_\alpha\rho(q \,|\, q_1 \ldots q_n) = 0 \tag{2.7}$$

and the dipole sum rule

$$\int dq\, e_\alpha x \rho(q \mid q_1 \ldots q_n) = 0 \tag{2.8}$$

Finally, let us define the function $\hat{\rho}^T(q_1 q_2)$:

$$\hat{\rho}^T(q_1 q_2) = \rho^T(q_1 q_2) + \delta_{q_1 q_2}\rho(q_1) = \rho(q_1 \mid q_2)\rho(q_2) = \rho(q_2 \mid q_1)\rho(q_1) \tag{2.9}$$

in terms of which the charge–charge correlation reads

$$S(x_2 - x_1) = \sum_{\alpha_1 \alpha_2} e_{\alpha_1} e_{\alpha_2} \hat{\rho}^T_{\alpha_1 \alpha_2}(x_1, x_2) \tag{2.10}$$

The charge sum rule implies (1.2), whereas in uniform states all higher-order multipole moments of $S(x)$ have to vanish (when they exist) because of spherical symmetry.

3. PROOF OF STILLINGER-LOVETT SECOND MOMENT CONDITION

The precise clustering assumptions for the validity of the S.L. condition are stated in the following proposition.

Proposition 1. The S.L. condition (1.4) is satisfied in any homogeneous isotropic neutral state obeying the BGY equations (2.1), (2.2), provided that (2.5) holds with

(i) $\eta > \nu + 2, \quad k = 2$

(ii) $\eta > \nu + 1, \quad k = 3, 4$

$\nu \geqslant 2$

and

(iii) $$\int dq_1 \int dq_2\, |e_{\alpha_2} x_2|\, |\rho^T(q_1 q_2 q_3)| < \infty$$

Before going into the proof, we give in Lemma 1 two equivalent formulations of (1.1) and (1.4) for uniform states.

Lemma 1. Under the assumption (i) of Proposition 1, the formulas (1.1), (1.4) and (3.1), (3.2) are equivalent for any uniform state satisfying the electroneutrality condition (1.2), with

$$\beta \int dx \left[\int dy\, \phi(x-y) S(y) \right] = 1 \tag{3.1}$$

$$\frac{\beta}{\nu} \int dx\, x \cdot \left[\int dy\, (-(\nabla\phi)(x-y)) S(y) \right] = 1 \tag{3.2}$$

where ϕ is the Coulomb potential (2.3).

The proof of Lemma 1 can be found in Appendix B.

Proof of Proposition 1. Because of translation invariance, the left-hand side of (2.2) can be written

$$\nabla_1 \rho(q_1 q_2) = -\nabla_2 \rho(q_1 q_2) = -\nabla_2 \rho^T(q_1 q_2) \tag{3.3}$$

Multiplying the first BGY equation (2.1) by $\rho(q_1 q_2)/\rho(q_1)$ and subtracting it from the second equation (2.2) gives, with (3.3),

$$-\nabla_2 \rho^T(q_1 q_2) = \beta \int dq\, F(q_1 q) \left[\rho(q_1 q_2 q) - \frac{\rho(q_1 q)\rho(q_1 q_2)}{\rho(q_1)} + \delta_{qq_2} \rho(q_1 q) \right] \tag{3.4}$$

where $F(q_1 q_2) = F^S(q_1 q_2) + F^L(q_1 q_2)$ is the total force.

We take now the scalar product of (3.4) with $e_{\alpha_2} x_2$, sum over the species α_2, and integrate x_2 in a sphere of radius R centered at the origin.

Integrating by parts, the left-hand side of (3.4) gives then

$$\begin{aligned}
&\int_{|x_2| \leqslant R} dx_2 \sum_{\alpha_2} e_{\alpha_2} x_2 \cdot \nabla_2 \rho^T(q_1 q_2) \\
&\quad = -\nu \int_{|x_2| \leqslant R} dx_2 \sum_{\alpha_2} e_{\alpha_2} \rho^T(q_1 q_2) + \int_{|x_2| = R} dS_2 \cdot \sum_{\alpha_2} e_{\alpha_2} x_2 \rho^T(q_1 q_2)
\end{aligned} \tag{3.5}$$

The assumption (i) implies that the surface term in (3.5) vanishes as $R \to \infty$, and the first term of the right-hand side converges to $\nu e_{\alpha_1} \rho(q_1)$ by the charge sum rule, which is true when (i) and (ii) hold.[5,6]

We get thus from (3.4) and (3.5)

$$\begin{aligned}
&\lim_{R \to \infty} \beta \int_{|x_2| \leqslant R} dq_2\, e_{\alpha_2} x_2 \\
&\quad \cdot \left[\int dq\, F(q_1 q) \left(\rho(q_1 q_2 q) - \frac{\rho(q_1 q)\rho(q_1 q_2)}{\rho(q_1)} + \delta_{qq_2} \rho(q_1 q) \right) \right] \\
&\quad = -\nu e_{\alpha_1} \rho(q_1)
\end{aligned} \tag{3.6}$$

In order to obtain a sum rule for the two-point function, it is necessary to eliminate the contribution of the three-point function from (3.6). This can be done with the help of the following identities obtained from (2.4) and (2.6), respectively:

$$\rho(q_1 q_2 q) - \frac{\rho(q_1 q)\rho(q_1 q_2)}{\rho(q_1)} + \delta_{qq_2} \rho(q_1 q)$$

$$= \rho(q_1) \hat{\rho}^T(q_2 q) + r(q_1 q_2 q) \tag{3.7}$$

$$= (\rho(q_2 \mid q_1 q) - \rho(q_2 \mid q_1)) \rho(q_1 q) \tag{3.8}$$

with

$$r(q_1q_2q) = \rho^T(q_1q_2q) - \frac{\rho^T(q_1q)\rho^T(q_1q_2)}{\rho(q_1)} + \delta_{qq_2}\rho^T(q_1q) \tag{3.9}$$

The dipole sum rule (2.8) for $n = 1, 2$, which is valid by assumption (ii) (see Ref. 6), implies with (3.8) that

$$\lim_{R\to\infty} \int_{|x_2|\leqslant R} dq_2\, e_{\alpha_2}x_2 \left(\rho(q_1q_2q) - \frac{\rho(q_1q)\rho(q_1q_2)}{\rho(q_1)} + \delta_{qq_2}\rho(q_1q) \right) = 0 \tag{3.10}$$

Since the short-range force is integrable, the clustering assumptions imply that the part of the integral of (3.6) involving $F^S(q_1q)$ is jointly integrable for x and x_2 in $\mathbb{R}^\nu \times \mathbb{R}^\nu$. This allows to exchange the order of integrations in (3.6) and to take the limit $R \to \infty$ under the q-integral sign. Hence, by virtue of (3.10), the short-range force does not contribute to (3.6).

To evaluate the contribution of the Coulomb force to (3.6), we notice from (3.7), (3.8) and the dipole sum rules that we have also

$$\lim_{R\to\infty} \int_{|x_2|\leqslant R} dq_2\, e_{\alpha_2}x_2 r(q_1q_2q) = 0 \tag{3.11}$$

Moreover, the expression (3.9) with (iii) shows that $e_{\alpha_2}x_2 r(q_1q_2q)$ is itself jointly integrable in q and q_2. Exchanging as before the q integral and the limit $R \to \infty$, we conclude from (3.11) that the contribution of $e_{\alpha_2}x_2 \cdot F^L(q_1q_2)r(q_1q_2q)$ to (3.6) also vanishes as $R \to \infty$. Thus we are left in (3.6) with

$$\lim_{R\to\infty} \beta \int_{|x_2|\leqslant R} dq_2\, e_{\alpha_2}x_2 \cdot \left[\int dq\, F^L(q_1q)\hat{\rho}^T(q_2q) \right] = -\nu e_{\alpha_1} \tag{3.12}$$

When we introduce the definition (2.10) of the charge–charge correlation function and set $x_1 = 0$, we see that (3.12) is identical to (3.2), and thus to (1.4) or (1.1) by the lemma.

Comments

1. There is no loss in generality in assuming that the state is translation and rotation invariant. In fact, with essentially the same hypothesis as in Proposition 1 [precisely, (2.5) holds with $\eta > \nu + 2$, $k = 2, 3, 4$ and $\int dq_1 \int dq_2 |e_{\alpha_2}x_2| |\rho^T(q_1q_2 \ldots q_n)| < \infty$ for $n = 3, 4$[3]] it has been proven that any solution $\rho(q_1), \rho(q_1q_2)$ of the BGY equations (2.1), (2.2) has to be Euclidean invariant (see Proposition 3, of Ref. 9).

[3] Here, $\eta > \nu + 1$ is sufficient when there is no background density, i.e., $\rho_B = 0$.

The same remark applies to the neutrality $\sum_\alpha \rho_\alpha + \rho_B = 0$: any translation-invariant equilibrium state having integrable clustering is necessarily locally neutral (Ref. 5, Proposition 6).

2. Since $E = \epsilon^{-1}D$, $\epsilon^{-1} = 0$ means phenomenologically that there cannot exist a constant bulk electric field $E \neq 0$ in the system. This agrees with the result of Ref. 9, Corollary 2 of Proposition 3, showing that under the same clustering conditions, there does not exist any solution of the equilibrium equations with nonvanishing bulk electric field.

3. As already mentioned in the introduction, the S.L. condition is violated in the two-dimensional Kösterlitz–Thouless phase and in the one-dimensional (two component) Coulomb gas. The reason for the nonapplicability of our proof is different in the two cases. The clustering of the Kösterlitz–Thouless phase is presumably too weak. However, in the one-dimensional Coulomb gas, although the clustering is known to be exponentially fast, the dipole sum rule (2.8) with $n = 2$ cannot be established because of the peculiar nature of the one-dimensional Coulomb force (i.e., nondecreasing at infinity; see Ref. 6, Section C). In fact, the use of the dipole sum rule for $n = 2$ in (3.8) is the nontrivial part of our proof. [Equation (2.8) with $n = 1$ is trivially true in uniform systems.]

4. If we define

$$\rho_0(q_1 \cdots q_n) = \frac{\rho(q_0 q_1 \cdots q_n)}{\rho(q_0)} \qquad q_0 = (\alpha_0, x_0) \tag{3.13}$$

then $\rho_0(q_1 \ldots q_n)$ are the correlation functions of an equilibrium state in the presence of an external particle α_0 fixed at x_0.

We see that (3.4) can be written as

$$\nabla_1 \rho_0(q_1) = \beta\Big(F^{(e)}(q_1) + \int dq\, F^{(e)}(q)\rho_0^T(qq_1)\Big) \tag{3.14}$$

where we have set $q_1 \to q_0$, $q_2 \to q_1$, and $F^{(e)}(q) = F(qq_0)$ in (3.4).

Equation (3.14) is a special case of a more general equation for nonuniform systems, the Wertheim, Lovett, Mou and Buff (WLMB) equation, which relates the density gradient to an integral of the external force $F^{(e)}$ over the pair correlation function (the external force being here due to one of the system's particle α_0 fixed at x_0).

The WLMB equation for charged systems in the case of arbitrary boundaries and general external forces has been recently established and discussed in Ref. 14, and will be used in the Section 5.

5. The case where the two-body short-range force $F^S_{\alpha_1\alpha_2}(x)$ has a nonintegrable singularity at the origin can be treated in the same way

provided that the correlations vanish sufficiently fast at coincident points, i.e., $F^S(q_1q_2)\rho(q_1q_2), F^S(q_1q_2)\rho(q_1q_2q_3), \ldots,$ are integrable in q_1. It can then be checked that the same proof can be carried through.

4. MIXTURE OF IONS AND DIPOLES

The results of the preceding section can be extended without difficulty to the case of a uniform dipolar solvant. We indicate here the needed modifications of the proof.

The particle of species α carries now either a charge e_α or a permanent dipole moment $\mu_\alpha = d_\alpha \omega$ with orientation ω. We set $q = (\alpha, x, \omega)$ and $\int dq \cdots = \int_{\mathbb{R}^\nu} dx \int d\omega \sum_\alpha \cdots$. We normalize the integration over dipole angles to one, i.e., $\int d\omega = 1$.

We assume that the ionic densities do not vanish. (The consideration of this section does not apply to a pure dipole fluid which has a very different physical behavior.) With these notations, the BGY hierarchy keeps the same form (2.1) and (2.2), with the following definition of the short- and long-range part of the force:

F^S includes any integrable force, antisymmetric and covariant under rotations, as well as the dipole–dipole force (which has also an integrable decay).

F^L consists of the charge–charge and the charge–dipole forces

$$F^L(q_1q_2) = -\nabla_1(e_{\alpha_1}e_{\alpha_2} + e_{\alpha_1}\mu_{\alpha_2}\cdot\nabla_2 + e_{\alpha_2}\mu_{\alpha_1}\cdot\nabla_1)\phi^r(x_1 - x_2) \tag{4.1}$$

$$\phi^r(x) = \begin{cases} \displaystyle\int dy \frac{1}{|x-y|} f(|y|), & \nu = 3 \\ \displaystyle -\int dy \ln|x-y| f(|y|), & \nu = 2 \end{cases} \tag{4.2}$$

where f is a smooth function with compact support such that $\int dy\, f(|y|) = 1$.

We have introduced a regularization of the Coulomb potential at the origin to suppress the $|x|^{-\nu}$ singularity in the charge–dipole force at $x = 0$. The difference between this cutoff force and the exact Coulomb force can be included as a contribution to $F^s(q_1q_2)$.

In addition to the charge–charge correlation function

$$S(x_2 - x_1) = \int d\omega_1 \int d\omega_2 \sum_{\alpha_1\alpha_2} e_{\alpha_1}e_{\alpha_2}\rho^T_{\alpha_1\alpha_2}(x_1\omega_1, x_2\omega_2) + \sum_\alpha e_\alpha^2 \rho_\alpha \delta(x_2 - x_1) \tag{4.3}$$

we define the charge–dipole correlation function

$$P(x_2 - x_1) = \int d\omega_1 \int d\omega_2 \sum_{\alpha_1\alpha_2} e_{\alpha_1} \mu_{\alpha_2} \rho^T_{\alpha_1\alpha_2}(x_1\omega_1, x_2\omega_2) \tag{4.4}$$

Then, the S.L. conditions (1.1), (1.4), (3.1), (3.2) are again equivalent provided that one replaces $S(x)$ by $S(x) - (\nabla \cdot P)(x)$, and we have the analog of Proposition 1.

Proposition 2. The S.L. condition $\int dx\, |x|^2 (S(x) - (\nabla \cdot P)(x)) = -2\nu/\omega_\nu \beta$ is satisfied in any homogeneous isotropic state obeying the equations (2.1), (2.2) provided that (2.5) holds with

(i) $\eta > \nu + 2$, $k = 2$ when α_1 and α_2 are charges; $\eta > \nu + 1$, $k = 2$ when α_1 is a charge, α_2 a dipole;

(ii) $\eta > \nu + 1$, $k = 3, 4$ when α_i are all charges or charges and dipoles; $\eta > \nu$, $k = 2, 3, 4$ when all α_i are dipoles;

(iii) $\int dq_1 \int dq_2\, |e_{\alpha_2} x_2|\, |\rho^T(q_1 q_2 q_3)| < \infty$.

(See the proof of Lemma 2 in Appendix B.)

Lemma 2. Under the assumption (i) of Proposition 2, the formulas (1.1), (1.4), (3.1), and (3.2) are equivalent [with $S(x) - (\nabla \cdot P)(x)$ in place of $S(x)$] for any uniform state satisfying the electroneutrality condition (1.2).

The lemma also holds if we have the regularized Coulomb potential ϕ^r (4.2) in (3.1) and (3.2).

For the proof of the Proposition 2, we first establish Eq. (3.4) as in Proposition 1. Let then the particle of type α_1 be charged ($e_{\alpha_1} \neq 0$, $d_{\alpha_1} = 0$). In view of the clustering property of the charge–dipole correlation one has

$$\lim_{R\to\infty} \int_{|x_2| \leqslant R} dx_2 \int d\omega_2 \sum_{\alpha_2} \mu_{\alpha_2} \cdot \nabla_2 \rho^T(q_1 q_2) = 0$$

Using the charge sum rule (2.7) which is valid when (i) and (ii) hold (Appendix A of Ref. 14), we get

$$\lim_{R\to\infty} \int_{|x_2| \leqslant R} dx_2 \int d\omega_2 \sum_{\alpha_2} (e_{\alpha_2} x_2 + \mu_{\alpha_2}) \cdot \nabla_2 \rho^T(q_1 q_2) = \nu e_{\alpha_1} \rho(q_1) \tag{4.5}$$

Hence the analog of (3.6) becomes

$$\begin{aligned} &\lim_{R\to\infty} \beta \int_{|x_2| \leqslant R} dq_2 (e_{\alpha_2} x_2 + \mu_{\alpha_2}) \\ &\qquad \cdot \left[\int dq\, F(q_1 q) \left(\rho(q_1 q_2 q) - \frac{\rho(q_1 q)\rho(q_1 q_2)}{\rho(q_1)} + \delta_{q q_2} \rho(q_1 q) \right) \right] \\ &\quad = -\nu e_{\alpha_1} \rho(q_1) \end{aligned} \tag{4.6}$$

The rest of the argument is now identical to (3.7)–(3.11). We use here the dipole-sum rules

$$\int dq\,(e_\alpha x + \mu_\alpha)\rho(q \mid q_1 \ldots q_n) = 0, \qquad n = 1, 2 \tag{4.7}$$

which are also true under the assumptions (i) and (ii) (see Appendix A of Ref 14). We are thus left with

$$\lim_{R\to\infty} \beta \int_{|x_2| \leqslant R} dq_2\,(e_{\alpha_2} x_2 + \mu_{\alpha_2}) \cdot \left[\int dq\, F^L(q_1 q)\hat{\rho}^T(q_2 q) \right] = -\nu e_{\alpha_1} \tag{4.8}$$

After the change of variable $x \to x + x_2$ in the q-integral, taking into account that $\mu_{\alpha_1} = 0$ and setting $x_1 = 0$, (4.8) takes the more explicit form

$$\lim_{R\to\infty} \beta \int_{|x_2| \leqslant R} dq_2\,(e_{\alpha_2} x_2 + \mu_{\alpha_2}) \cdot \nabla_2 \left[\int dq\,(e_\alpha + \mu_\alpha \cdot \nabla_x)\phi^r(x - x_2)\hat{\rho}(q, \alpha_2 0 \omega_2) \right] = -\nu \tag{4.9}$$

The bracket $[\cdots]$ in (4.9) is the potential at x_2 due to the excess particle distribution $\rho(q \mid \alpha_2 0 \omega_2)$ with a particle α_2 fixed at the origin. Since this distribution has zero total charge and no dipole moment, the corresponding potential has to decrease faster as $|x_2|^{-(\nu-1)}$ as $|x_2| \leqslant \infty$ (see Lemma 4, Appendix A).

As a consequence, the term in (4.9) proportional to μ_{α_2} vanishes as $R \to \infty$ and when the definitions (4.3) and (4.4) are introduced, (4.9) reduces to one of the equivalent formulas (3.1) and (3.2) with $S(x) - (\nabla \cdot P)(x)$ replacing $S(x)$. The S.L. condition (4.1) is now a consequence of Lemma 2.

5. COMPLETE SHIELDING

In this section, we show that complete shielding occurs under the same conditions as those necessary to derive the S.L. condition. We then extend the S.L. condition to locally inhomogeneous systems in $\mathbb{R}^\nu$.

We consider here an infinitely extended, pure charged system (no dipoles) in the presence of a localized distribution of arbitrary fixed charges $C^{(e)}(x)$. The equilibrium correlations $\rho^*(q_1 \ldots q_n)$ of the inhomogeneous system are solution of the BGY hierarchy whose first equation is

$$\beta^{-1}\nabla_1 \rho^*(q_1) = e_{\alpha_1} E(x_1)\rho^*(q_1) + \int dq\, F^L(q_1 q)\rho^{*T}(q_1 q) \tag{5.1}$$

$$E(x_1) = E_0 + \int dx \left(\frac{x_1 - x}{|x_1 - x|^\nu} + \frac{x}{|x|^\nu} \right) \left(C^*(x) + C^{(e)}(x) \right) \tag{5.2}$$

is the electric field due to all charges, i.e., the system's charge density

$C^*(x) = \rho_B + \sum_\alpha e_\alpha \rho_\alpha^*(x)$ and the external distribution $C^{(e)}(x)$, and $E_0 = E(x_1 = 0)$.

Throughout this section, we study asymptotically neutral states, assuming that the system's charge density decays as

$$C^*(x) = O\left(\frac{1}{|x|^\epsilon}\right), \qquad |x| \to \infty, \quad \text{for some} \quad \epsilon > 0 \tag{5.3}$$

With (5.3), $E(x_1)$ is well defined by Eq. (5.2) and everywhere bounded (see Ref. 14).

The system has the *complete shielding property* if any external distribution is screened by the system's charges, i.e.,

$$\int dx\left(C^*(x) + C^{(e)}(x)\right) = 0 \tag{5.4}$$

In the particular case where the external distribution consists of point particles of the same species as those which constitute the system itself, located at $q_1, \ldots, q_n$, one has

$$\rho^*(q) = \frac{\rho(qq_1 \cdots q_n)}{\rho(q_1 \cdots q_n)}$$

and therefore (5.4) is identical to the n-charge sum rule (2.7). The shielding of the system's charge occurs when the clustering is integrable (since then the charge sum rule holds), and is likely to be even more generally true.

The situation is, however, quite different when the external charges are not integer multiples of the system's charges. In fact, fractional charges are not shielded and the S.L. condition is violated in the one-dimensional Coulomb gas at all temperatures (even though the clustering is exponentially fast).[11] The same remark applies to the Kösterlitz–Thouless phase in two dimensions at low temperature.[10] On the other hand, it follows from the cluster expansion that both properties (i.e., complete shielding and S.L. conditions) are true in the high-temperature phase in dimension $\nu \geqslant 2$.[15] It is therefore natural to conjecture that the S.L. relation and the complete shielding property are equivalent statements.

The following proposition shows that in fact, both properties hold under essentially identical clustering assumptions.

Proposition 3. Let $\rho^*(q_1 \ldots q_n)$ be an equilibrium state of a Coulomb system in the presence of an external (point) charge Q^*. Then the complete shielding of Q^* occurs if the condition (2.5) holds with

(i) $\qquad \eta > \nu + 2, \qquad k = 2, 3, 4, \quad \nu \geqslant 2$

(ii) $\qquad \int dq_1 \int dq_2 \, |\rho^T(q_1 q_2 \cdots q_k)|, \qquad k = 3, 4$

To establish Proposition 3, we first show that the state ρ^* is homogeneous at infinity and that the local perturbation induced by the external charge decays in an integrable way.

Lemma 3. Under the hypothesis of Proposition 3, there exist homogeneous densities ρ_α and correlations $\rho(q_1 q_2) = \rho_{\alpha_1\alpha_2}(x_1 - x_2)$ such that

$$\rho^*_\alpha(x) - \rho_\alpha = O\left(\frac{1}{|x|^{\nu+\epsilon}}\right), \qquad \epsilon > 0 \tag{5.5}$$

$$\rho^*_{\alpha_1\alpha_2}(x_1, x_2) - \rho_{\alpha_1\alpha_2}(x_1 - x_2) = O\left(\frac{1}{|x_1 + x_2|^{\nu+\epsilon}}\right) \tag{5.6}$$

uniformly with respect to $x_l - x_2$.

Proof of Lemma 3. Information on the asymptotic behavior of the density can be obtained from the first WLMB equation (Wertheim–Lovett–Mou–Buff):

$$\nabla_1 \rho^*(q_1) = \beta \int F^{(e)}(q) \hat{\rho}^{*T}(q_1 q)\, dq \tag{5.7}$$

$F^{(e)}(Q) = e_\alpha Q^*(x/|x|^\nu)$ is the external force.

It has been shown in Ref. 14 that Eq. (5.7) [and Eq. (5.11) below] can be derived rigorously from the BGY hierarchy under the conditions of the proposition and (5.3).

Introducing the definition

$$g_{x_1}(x) = \sum_\alpha e_\alpha \hat{\rho}^{*T}_{\alpha\alpha_1}(x x_1) \tag{5.8}$$

Equation (5.7) takes the form

$$\nabla_1 \rho^*_{\alpha_1}(x_1) = \beta Q^* \int dx \frac{x}{|x|^\nu} g_{x_1}(x) = \beta Q^* \int dx \frac{x + x_1}{|x + x_1|^\nu} g_{x_1}(x + x_1) \tag{5.9}$$

From the clustering assumptions (i) and the results of Ref. 6, $g_{x_1}(x)$ has no multipoles up to order 2. Hence, the same is true for the shifted distribution $g_{x_1}(x + x_1)$ for each fixed x_1. Moreover, it follows from (i) and (5.8) that $g_{x_1}(x + x_1)$ is $O(1/|x|^{\nu+2+\epsilon})$ uniformly with respect to x_1. We can therefore conclude from the Lemma 4 of Appendix A that

$$\nabla_1 \rho^*_{\alpha_1}(x_1) = O\left(\frac{1}{|x_1|^{\nu+1+\epsilon}}\right) \tag{5.10}$$

which in turn implies the result (5.5) by integration.

To prove (5.6) we use similar arguments starting from the WLMB equation for the two-point function,[14]

$$\begin{aligned}(\nabla_1 + \nabla_2)\rho^*(q_1 q_2) &= \beta\big(F^{(e)}(q_1) + F^{(e)}(q_2)\big)\rho^*(q_1 q_2) \\ &\quad + \beta \int dq\, F^{(e)}(q)(\rho^*(q q_1 q_2) - \rho^*(q)\rho^*(q_1 q_2))\end{aligned} \tag{5.11}$$

The details are in Appendix C.

Proof of Proposition 3. As a consequence of Lemma 3, we note that $\rho^T_{\alpha_1\alpha_2}(x_1 - x_2)$ has the same clustering properties as $\rho^{*T}_{\alpha_1\alpha_2}(x_1, x_2)$. Indeed, using (5.5), (5.6), and (i), one gets

$$|\rho^T_{\alpha_1\alpha_2}(x_1 - x_2)| \leqslant |\rho^{*T}_{\alpha_1\alpha_2}(x_1 + a, x_2 + a)| + O\left(\frac{1}{|a|^{\nu+\epsilon}}\right)$$
$$\leqslant \frac{M}{|x_1 - x_2|^{\nu+2+\epsilon}} + O\left(\frac{1}{|a|^{\nu+\epsilon}}\right)$$

Letting $|a| \to \infty$ with x_1, x_2 fixed, we obtain

$$|\rho^T_{\alpha_1\alpha_2}(x_1 - x_2)| \leqslant \frac{M}{|x_1 - x_2|^{\nu+2+\epsilon}} \tag{5.12}$$

We deduce from (5.6) and (5.12) that

$$\rho^{*T}_{\alpha_1\alpha_2}(x_1, x_2) - \rho^T_{\alpha_1\alpha_2}(x_1 - x_2) = O\left(\frac{1}{|x_1|^{\nu+1}}\right) \tag{5.13}$$

uniformly with respect to x_2.

The statement of Proposition 3 is then obtained using a result of Ref. 6, Section IV showing precisely that complete shielding occurs whenever (5.5) and (5.13) are true.

Comments

1. Integrating (5.7) on x_1 and summing on charges e_{α_1}, we get in the same way as (3.6) what did follow from (3.4)

$$\frac{\beta}{\nu} \int dx\, \mathbf{x} \cdot \int dy\, (-\nabla\phi(y)) S^*(x, y) = 1 \tag{5.14}$$

where $S^*(x, y)$ is the charge–charge correlation of the inhomogeneous state, and ϕ the Coulomb potential (2.3) (see also Ref. 16).

Equation (5.14) is the proper generalization of (3.2) to nonuniform systems. It cannot in general be reduced to the form (1.1) or (1.2). However, for a locally perturbed state, we expect that the spatial average of the charge–charge correlation

$$\bar{S}(x - y) = \lim_{V \to \mathbb{R}^\nu} \frac{1}{V} \int_V da\, S^*(x + a, y + a) = \lim_{|a| \to \infty} S^*(x + a, y + a)$$

still satisfies the S.L. condition in the form (1.1) or (1.4). Starting from (5.14) and using the same arguments as in Proposition 1, this will certainly be the case whenever the difference $\bar{S}(x - y) - S^*(x, y)$ is jointly integrable in x and y.

2. The result of Proposition 3 can be generalized to arbitrary localized external charge distributions, as well as to mixtures of ions and

permanent dipoles. For systems limited by infinitely extended walls (as a charged planar electrode) this analysis can be carried through with some modifications due to additional surface terms which enter in the WLMB equation (see Ref. 14).

3. It is interesting to remark that one must also use the dipole sum rule with $n = 2$ in the proof of Proposition 3 [see (B7)]. The quadrupole sum rule (which was automatic for $\rho(q \mid q_1)$ in the homogeneous case) is also used nontrivially to derive Proposition 3.

4. It can be checked that the following more general result is true. If ν is replaced by $\nu + l$, l a positive integer, in the condition (i) of Proposition 3, then $C^*(x) + C^{(e)}(x)$ carries no multipoles up to order l. In particular, if the ρ^{*T} have exponential clustering, the screening of $C^{(e)}(x)$ is perfect, i.e., without the occurrence of any multipole moment. This extends the perfect screening property discussed in Ref. 7 to arbitrary external charges.

APPENDIX A

The following lemma (essentially the same as Lemma 1 of Ref. 6) is always used to estimate the asymptotic behavior of the correlation functions. We state it here again for convenience.

Lemma 4. Let $F(x)$ be a locally integrable function on $\mathbb{R}^\nu$, continuously differentiable up to order $l + 1$ in a neighborhood of $x = \infty$ with

$$\text{(i)} \quad \left(\partial^{(k)}_{i_1 \ldots i_k} F\right)(x) = O\left(\frac{1}{|x|^{\gamma+k}}\right), \qquad k = 0, 1, \ldots, l+1, \quad \gamma > 0$$

and $g(x, y)$ a bounded function such that

$$\text{(ii)} \qquad g(x, y) = O\left(\frac{1}{|y|^{\nu+l+\epsilon}}\right), \qquad \epsilon > 0$$

uniformly with respect to x.

Then

$$\int dy\, R_l(x, y) g(x, y) = O\left(\frac{1}{|x|^{\gamma+l+\delta}}\right) \tag{A1}$$

for some $\delta > 0$ and where

$$R_l(x, y) = F(x - y) - \sum_{k=0}^{l} \frac{(-1)^k}{k!} y^{i_1} \ldots y^{i_k} \left(\partial^{(k)}_{i_1 \ldots i_k} F\right)(x) \tag{A2}$$

Proof. Consider first in (A1) the integration domain $|y| \leqslant |x|/2$. An estimation of the rest of the Taylor expansion of $F(x - y)$ around x gives with (i)

$$|R_l(x,y)| \leqslant M \frac{|y|^{l+1}}{|x|^{\gamma+l+1}} \leqslant M' \frac{|y|^{l+\delta}}{|x|^{\gamma+l+\delta}} \tag{A3}$$

with $0 < \delta \leqslant 1$, $M' = M(1/2)^{1-\delta}$.

Thus, choosing $0 < \delta < \epsilon$,

$$\left| \int_{|y| \leqslant |x|/2} dy\, R_l(x,y) g(x,y) \right| \leqslant \frac{M'}{|x|^{\gamma+l+\delta}} \int_{|y| \leqslant |x|/2} dy\, |y|^{l+\delta} |g(x,y)|$$
$$= O\left(\frac{1}{|x|^{\gamma+l+\delta}} \right)$$

since by (ii) $|y|^{l+\delta} g(x,y)$ is integrable in y uniformly with respect to x for $0 < \delta < \epsilon$.

The contribution of the domain $|y| \geqslant |x|/2$ in (A2) is estimated exactly as in Lemma 1 of Ref. 6.

Let us note that this lemma still holds in the case $\gamma = 0$ under the conditions

$$F(x) = O(\ln|x|)$$
$$\left(\partial^{(k)}_{i_1 \ldots i_k} F\right)(x) = O\left(\frac{1}{|x|^k}\right), \qquad k = 1, \ldots, e+1$$

APPENDIX B. "PROOF OF LEMMAS 1 AND 2"

A. Equivalence of (1.1) and (2.2)–(4)

$\lim_{|k|\to 0} S(k) = 0$ is equivalent to $\int dx\, S(x) = 0$, i.e., (1.2). $\lim_{|k|\to 0}(1/|k|) S(k) = 0$ is equivalent to $\int dx\, x S(x) = 0$, i.e., (1.3).

Using (1.2) and (1.3)

$$\frac{1}{\omega_\nu \beta} = \lim_{|k|\to 0} \frac{1}{|k|^2} S(k) = \lim_{|k|\to 0} \int dx \left(\frac{e^{ikx} - 1 - ikx}{|k|^2} \right)$$
$$= -\frac{1}{2} \int dx (\hat{k} \cdot x)^2 S(x) = -\frac{1}{2\nu} \int dx\, |x|^2 S(x) \tag{B1}$$

where the exchange of the limit and the integral is allowed by dominated convergence.

B. Equivalence of (1.2)–(4), (3.1), and (3.2)

Under the clustering assumption (i), $S(x) = O(1/|x|^{\nu+2+\epsilon})$ and since the state is isotropic, $S(x)$ has neither dipole nor quadrupole moment.

Therefore, using $\sum_i^\nu(\partial_{ii}^{(2)}\phi)(x_1)=(\Delta\phi)(x_1)=0$, $x_1\neq 0$, and applying Lemma 4 with $\gamma=\nu-2$ and $l=2$ we find

$$\int dx\,\phi(x_1-x)S(x)=\int dx\Bigg[\phi(x_1-x)-\phi(x_1)+x\cdot(\nabla\phi)(x_1)$$
$$-\frac{1}{2}\sum_{ij}^{\nu}\left(x^ix^j-\frac{1}{\nu}\delta^{ij}\right)(\partial_{ij}^{(2)}\phi)(x_1)\Bigg]S(x)$$
$$=\int dx\,R_2(x_1,x)S(x)=O\left(\frac{1}{|x_1|^{\nu+\delta}}\right)\tag{B2}$$

In the same way, for the force $F=-\nabla\phi$, Lemma 4 with $\gamma=\nu-1$ and $l=2$ yields

$$\int dx\,F(x_1-x)S(x)=\int dx\,R_2(x_1,x)S(x)=O\left(\frac{1}{|x_1|^{\nu+1+\delta}}\right)\tag{B3}$$

Consider now

$$-\omega_\nu\int dx_1\,|x_1|^2S(x_1)=\lim_{R\to\infty}\int_{|x_1|\leqslant R}dx_1\,|x_1|^2\nabla_1^2\int dx\,\phi(x_1-x)S(x)\tag{B4}$$

(3.2) and (3.1) follow when we perform one and two integration by parts in (B4), noting that the surface terms vanish in view of (B3) and (B2).

The proof of Lemma 2 is identical to that of Lemma 1 when one notices that the covariance of $P(x)$ under rotations implies

$$\int dx\,P(x)=0\tag{B5}$$

and

$$\int dx\left(\nu x^iP^k(x)-\delta^{ik}x\cdot P(x)\right)=0\tag{B6}$$

With (B5), (B6), and $\sum_i^\nu(\partial_iF^i)(x_1)=(\nabla\cdot F)(x_1)=0$, $x_1\neq 0$, we can write

$$\int dx\,\phi(x_1-x)(\nabla\cdot P)(x)$$
$$=\int dx\Bigg\{(F(x_1-x)-F(x_1))\cdot P(x)$$
$$+\sum_{ik}^{\nu}(\partial_iF^k)(x_1)\left(x^iP^k(x)-\frac{1}{\nu}\delta^{ik}x\cdot P(x)\right)\Bigg\}$$
$$=\int dx(F(x_1-x)-F(x_1)+(x\cdot\nabla)F(x_1))\cdot P(x)$$
$$=\int dx\,R_1(x_1,x)P(x)\tag{B7}$$

Since $P(x)=O(1/|x|^{\nu+1+\epsilon})$ [assumption (i)], we conclude from Lemma 4 with $\gamma = \nu - 1$ and $l = 1$ that (B7) is $O(1/|x_1|^{\nu+\delta})$. Similarly,

$$\int dx\, F(x_1 - x)(\nabla \cdot P)(x) = \nabla_1 \int dx\, F(x_1 - x) \cdot P(x) = O\left(1/|x_1|^{\nu+1+\delta}\right) \tag{B8}$$

Notice finally that if ϕ is replaced by ϕ^r in (3.1) and (3.2), these equations can as well be written in terms of ϕ and S^r, P^r with

$$\begin{aligned} S^r(x) &= \int dy\, S(x-y) f(y) \\ P^r(x) &= \int dy\, P(x-y) f(y) \end{aligned} \tag{B9}$$

It is not difficult to check that S^r, P^r have the same properties as S and P. Therefore, using again the sum rules, we get also

$$\begin{aligned} -\frac{2\nu}{\omega_\nu \beta} &= \int dx\, |x|^2 \int dy\, (S(x-y) - (\nabla \cdot P)(x-y)) f(y) \\ &= \int dy\, f(y) \int dx\, (|x|^2 + 2x \cdot y + |y|^2)(S(x) - (\nabla \cdot P)(x)) \\ &= \int dx\, |x|^2 (S(x) - (\nabla \cdot P)(x)) \end{aligned}$$

APPENDIX C. "PROOF OF LEMMA 3"

Lemma 4 is applied as follows in the proof of Lemma 3.

A. Since $g_{x_1}(x + x_1)$ [(5.8)] has no multipole moments up to order $l = 2$, one has as in Appendix B

$$\int dx\, \frac{x_1 + x}{|x_1 + x|^\nu}\, g_{x_1}(x + x_1) = \int dx\, R_2(x_1, -x) g_{x_1}(x + x_1)$$

(5.10) follows then from Lemma 4 with $\gamma = \nu - 1$ and $l = 2$.

B. We write (5.11) in terms of the truncated functions

$$(\nabla_1 + \nabla_2)\rho^{*T}(q_1 q_2) = \beta \int dq\, F^{(e)}(q) \left[\rho^{*T}(q_1 q_2 q) + (\delta_{qq_1} + \delta_{qq_2}) \rho^{*T}(q_1 q_2) \right] \tag{C1}$$

With the introduction of the variables

$$\xi = \frac{x_1 + x_2}{2}, \qquad \eta = \frac{x_1 - x_2}{2}$$

and the definitions

$$\begin{aligned} g(\xi, \eta) &= \rho^{*T}_{\alpha_1 \alpha_2}(x_1, x_2) \\ h(x, \xi, \eta) &= \sum_\alpha e_\alpha \rho^{*T}_{\alpha \alpha_1 \alpha_2}(x_1, x_1, x_2) \end{aligned} \tag{C2}$$

(C1) takes the simple form

$$\nabla_\xi g(\xi,\eta) = \beta Q^* \int dx \frac{x}{|x|^\nu} \big[h(x,\xi,\eta) + \big(e_{\alpha_1}\delta(x-\xi-\eta) + e_{\alpha_2}\delta(x-\xi+\eta)\big) g(\xi,\eta) \big] \tag{C3}$$

From (2.6) we have the identity

$$\rho^{*T}(qq_1q_2) + (\delta_{qq_1} + \delta_{qq_2})\rho^{*T}(q_1q_2) = \rho^*(q \mid q_1q_2)\rho^*(q_1q_2) - (\rho^*(q \mid q_1) + \rho^*(q \mid q_2))\rho^*(q_1)\rho^*(q_2) \tag{C4}$$

which shows that the left-hand side of (C4) satisfies the $l = 0, 1, 2$ sum rules for fixed q_1 and q_2 since $\eta > \nu + 2$ by assumption (i). This implies that $h(x,\xi,\eta) + (e_{\alpha_1}\delta(x-\xi-\eta) + e_{\alpha_2}\delta(x-\xi+\eta))g(\xi,\eta)$ has no multipole moments up to order 2 for fixed ξ and η. So does the shifted distribution obtained by replacing x by $x+\xi$ for each fixed ξ.

This allows to write (C3) as

$$\begin{aligned}
\nabla_\xi g(\xi,\eta) &= \beta Q^* \int dx \frac{x+\xi}{|x+\xi|^\nu} \big[h(x+\xi,\xi,\eta) + \big(e_{\alpha_1}\delta(x-\eta) + e_{\alpha_2}\delta(x+\eta)\big) g(\xi,\eta) \big] \\
&= \beta Q^* \int dx\, R_2(\xi,-x) \big[h(x+\xi,\xi,\eta) + \big(e_{\alpha_1}\delta(x-\eta) + e_{\alpha_2}\delta(x+\eta)\big) g(\xi,\eta) \big] \\
&= \beta Q^* \big(e_{\alpha_1} R_2(\xi,-\eta) + e_{\alpha_2} R_2(\xi,\eta)\big)\, g(\xi,\eta) \\
&\quad + \beta Q^* \int dx\, R_2(\xi,-x) h(x+\xi,\xi,\eta)
\end{aligned} \tag{C5}$$

Since $|R_2(\xi,\eta)| \leqslant M'(|\eta|^{2+\delta}/|\xi|^{\nu+1+\delta})$ for fixed η, and ξ large enough [see (A3)] and since $|g(\xi,\eta)| \leqslant M/|\eta|^{\nu+2+\epsilon}$ with $\delta < \epsilon$ by the clustering assumption, we have

$$R_2(\xi,\eta)\, g(\xi,\eta) = O\left(\frac{1}{|\xi|^{\nu+1+\delta}} \right)$$

uniformly with respect to η.

The same is true for $R_2(\xi,-\eta) g(\xi,\eta)$.

Moreover, it follows from the definition (C4), (2.7), and the condition (i) of Proposition 3 that

$$|h(x+\xi,\xi,\eta)| \leqslant \frac{M}{r^{\nu+2+\epsilon}}, \qquad r = \sup(|x+\eta|, |x-\eta|, 2|\eta|)$$

Since $|x| \leqslant \sup(|x+\eta|, |x-\eta|) \leqslant r$ one has $h(x+\xi,\xi,\eta) = O(1/|x|^{\nu+2+\epsilon})$

uniformly with respect to ξ and η. The Lemma 4 implies then that the last term of (C5) is $O(1/|\xi|^{\nu+1+\delta})$ uniformly with respect to η.

We find thus $\nabla_\xi g(\xi, \eta) = O(1/|\xi|^{\nu+1+\delta})$ and with the definition (C2), this implies the result (5.6) of Lemma 3.

REFERENCES

1. F. Stillinger and R. Lovett, *J. Chem. Phys.* **48**:3858 (1968).
2. F. Stillinger and R. Lovett, *J. Chem. Phys.* **49**:1991 (1968).
3. D. J. Mitchell, D. A. McQuarrie, A. Szabo, and J. Groneveld, *J. Stat. Phys.* **17**:15 (1977).
4. M. Baus and J. P. Hansen, *Phys. Rep.* **59**:1 (1980).
5. Ch. Gruber, Ph. A. Martin, Ch. Lugrin, *J. Stat. Phys.* **22**:193 (1980).
6. Ch. Gruber, J. L. Lebowitz, and Ph. A. Martin, *J. Chem. Phys.* **72**:944 (1981).
7. L. Blum, Ch. Gruber, J. L. Lebowitz, and Ph. A. Martin, *Phys. Rev. Lett.* **48**:1769 (1982).
8. Ch. Gruber and Ph. A. Martin, *Ann. Phys.* **131**:56 (1981).
9. Ch. Gruber, Ph. A. Martin, and Ch. Oguey, *Commun. Math. Phys.* **84**:55 (1982).
10. J. Fröhlich and T. Spencer, *Commun. Math. Phys.* **81**:527 (1981); J. Fröchlich, in *Proceedings of the Int. School of Math. Phys. Ettore Majorano*, G. Velo and A. Wightmann, eds. (Plenum Press, New York, 1980).
11. M. Aizenman and Ph. A. Martin, *Commun. Math. Phys.* **78**:99 (1980); M. Aizenman, in *Proceedings of the Int. School of Math. Phys. Ettore Majorano*, G. Velo and A. Wightmann, eds. (Plenum Press, New York, 1980).
12. D. Chan, D. Mitchell, B. Ninham, and B. Pailthorpe, *J. Chem. Phys.* **69**:691 (1978).
13. J. Yvon, *J. Phys. Radium* **19**:733 (1959); R. Balescu, *Equilibrium and Nonequilibrium Statistical Mechanics* (Wiley, New York, 1975).
14. L. Blum, Ch. Gruber, D. Henderson, J. L. Lebowitz, and Ph. A. Martin, to appear in *J. Chem. Phys.*
15. D. Bridges and P. Federbush, *Commun. Math. Phys.* **73**:197 (1980).
16. S. L. Carnie and D. Chan, *Chem. Phys. Lett.* **77**:437 (1981).
17. J. Fröhlich and T. Spencer, *J. Stat. Phys.* **24**:617 (1981).