

Wigner Crystallization and Its Relation to the Poor Decay of Pair Correlations in One-Component Plasmas of Arbitrary Dimension

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We study various consequences of crystalline ordering in one-component (ordinary and surface) plasmas of arbitrary dimension, especially its relation to the poor decay of the pair correlation function. We considerably improve and presumably optimize the various known bounds on its decay at infinity and scrutinize the usual arguments claiming absence of crystallization in two-dimensional plasmas.

KEY WORDS: Wigner crystallization; one-component Coulomb plasmas; Mermin argument.

1. INTRODUCTION

In contrast to systems of classical particles interacting via short-range potentials, where crystallinity is ruled out by the original Mermin argument in one or two dimensions,⁽¹⁾ matters are not so transparent in the case of long-range interactions (e.g., Coulomb). In fact, there exists an old conjecture by Wigner⁽²⁾ that electron systems may crystallize at sufficiently low temperatures.

This has been rigorously confirmed by Kunz for the one-dimensional ordinary jellium⁽³⁾ and is due both to the peculiar properties of the one-dimensional Coulomb potential, $V(x) = -2\pi q^2 |x|$, and the obvious absence of transversal phonons. In higher dimensions two types of phonons exist, where the longitudinal branch turns out to be harmless, while the

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transverse ones exhibit the typical k^{-2} singularity, which, at least in harmonic approximation, rules out crystallinity in $d=2$ due to the usual Peierls argument.⁽⁴⁾

In more recent times there have been various attempts to go beyond the harmonic approximation by employing generalizations of the above-mentioned Mermin argument. A first step in this direction is due to Baus,⁽⁵⁾ who managed to incorporate the effects of the neutralizing background into various quantities, e.g., the structure factor $S(k)$. The essential ingredient is, however, an observation made independently by Chakravarty and Dasgupta⁽⁶⁾ and Alastuey and Jancovici⁽⁷⁾ that it is in fact the transverse phonons which matter and that one has to disentangle them from the longitudinal ones in the Mermin inequality. This was not done, e.g., in the paper by Baus, so that the crystallinity of neither ordinary nor surface jellium systems could be ruled out.

In ref. 6 the authors started with particles interacting via the three-dimensional Yukawa potential, being captured on a two-dimensional surface, and made the transition to the $1/r$ potential after the thermodynamic limit (TL) has been taken. In ref. 7 the two-dimensional surface jellium is treated for the special case of a triangular lattice with periodic boundary conditions. The authors admit, however, that several manipulations and limiting procedures are not completely rigorous.

In a subsequent paper Martinelli and Merlini⁽⁸⁾ tried to circumvent these problems for the two-dimensional ordinary jellium, i.e., $V(r) = -2q^2 \ln|r|$, by replacing possibly uncontrollable limit procedures with a decay assumption for a certain pair correlation function. However, as we will demonstrate in the following, a closer inspection shows that the pair correlation function employed by them does *not* decay at all in a crystalline state, which spoils their whole reasoning at a crucial point!

To put it in a nutshell: the question of whether crystalline states can exist appears, on a more rigorous level, to be not fully settled for the one-dimensional surface jellium [$V(r) \sim \ln|r|$] and in two dimensions for the general case both of ordinary and surface jellium [$V(r) \sim \ln|r|$, resp. $\sim 1/|r|$]. In the following we will reexamine this interesting question, exploiting (as did Martinelli and Merlini) the well-known connection between crystallinity and poor clustering of particle correlations and extend our methods also to the case $d=3$ (and, in principle, to arbitrary d).

As to this wider context, see, e.g., ref. 9, where the BBGKY hierarchy has been employed, and for the situation of shorter ranged interactions, also refs. 10 and 11, where other approaches have been used. In any case one should, however, remark that, typically, all strategies which do not employ the so-called Bogoliubov inequality do not lead to optimal results in this (however special) situation. Before we embark on the main subject

matter of the paper, we mention a very recent review of the whole field of Coulomb systems by Martin.⁽¹²⁾

As a first step, we develop in the following section a general method to calculate in an effective way the overall potential energy of jellium systems for *periodic boundary conditions* leading to modified effective pair potentials along the lines of ref. 13. In Section 3 we investigate the pair correlation function as suggested by Martinelli and Merlini and show that, in contrast to their assumption, it definitely does not decay in a crystalline state, but becomes asymptotically nontrivially periodic. We then treat in Section 4 in successive order the two- and three-dimensional Coulomb plasma and the two- and one-dimensional surface jellium, one of our main ideas being to split off the periodic contribution of the above-mentioned correlation function and analyze its details in *k* space rather than position space.

Our results are the following: If

$$|\rho_{T,A}^{(2)}(r_1, r_2)| \lesssim \begin{cases} \text{(a)} & |r_1 - r_2|^{-(1+\epsilon)} \\ \text{(b)} & |r_1 - r_2|^{-(3/2+\epsilon)} \\ \text{(c)} & |r_1 - r_2|^{-(1/2+\epsilon)} \\ \text{(d)} & |r_1 - r_2|^0 \end{cases} \quad (1.1)$$

uniformly in *A* (with *A*, the volume of the system, to ∞) for (a) two-dimensional ordinary jellium, (b) three-dimensional ordinary jellium, (c) two-dimensional surface jellium, and (d) one-dimensional surface jellium, then the corresponding equilibrium state cannot be crystalline. Here $\rho_T^{(2)}$ is the truncated two-point function, which definitely has to decay in a pure phase on *a priori* grounds. Thus, as a direct consequence of (1.1)(d), one-dimensional surface jellium can never crystallize.

2. EFFECTIVE POTENTIALS FOR COULOMB SYSTEMS

As we study Coulomb systems with periodic boundary conditions, the appropriate first step consists in solving the Poisson equation for *N* point particles, each carrying charge *q* and being immersed in a homogeneous neutralizing background, by Fourier methods.

For ordinary jellium⁽¹³⁾ the Poisson equation reads

$$\Delta\Phi(r) = -4\pi q \left[\sum_{i=1}^N \delta(r-r_i) - N/A \right] \quad (2.1)$$

the solution being

$$\Phi(r) = \Phi_0 + 4\pi q/\Lambda \cdot \sum_{k \neq 0} \sum_i 1/k^2 \cdot \exp[ik(r - r_i)], \quad k \in \Lambda^* \quad (2.2)$$

the reciprocal lattice to Λ .

The corresponding potential energy is

$$U = 1/2 \int d^d r q \left[\sum_i \delta(r - r_i) - N/\Lambda \right] \cdot \Phi(r) - 1/2 \int d^d r \sum_i \delta(r - r_i) \cdot V_c(r - r_i) \quad (2.3)$$

the second (formally infinite) term of which represents the usual self-energy counterterm occurring in the electrodynamics of ideal point charges.

Inserting (2.2) into (2.3), we get

$$U = U_0 + 1/2 \sum_{i \neq j} \sum_{k \neq 0} 4\pi q^2/\Lambda \cdot 1/k^2 \cdot \exp[ik(r_i - r_j)] \quad (2.4)$$

with

$$U_0 := \lim_{r \rightarrow 0} N/2 \cdot \left[\sum_{k \neq 0} 4\pi q^2/\Lambda \cdot 1/k^2 \cdot e^{ikr} - V_c(r) \right]$$

$V_c(r)$ being the standard solution of $\Delta V(r) = -4\pi q^2 \delta(r)$.

In the case of d -dimensional surface jellium we solve the $(d+1)$ -dimensional Poisson equation⁽¹⁴⁾

$$(\Delta + \partial^2/\partial z^2) \Phi(r, z) = -4\pi q \left[\sum_i \delta(r - r_i) - N/\Lambda \right] \cdot \delta(z) \quad (2.5)$$

The solution is

$$\begin{aligned} \Phi(r, z) &= \Phi_0 + \sum_{k \neq 0} \int dk 2q/\Lambda \cdot 1/(k^2 + \kappa^2) \cdot \sum_i \exp[ik(r - r_i)] \cdot \exp(ikz) \\ &= \Phi_0 + \sum_{k \neq 0} 2\pi q/\Lambda \cdot 1/|k| \cdot \sum_i \exp[ik(r_i - r_j)] \cdot \exp(-|k| \cdot |z|) \end{aligned} \quad (2.6)$$

The corresponding potential energy reads

$$U = U_0 + 1/2 \sum_{i \neq j} \sum_{k \neq 0} 2\pi q^2/\Lambda \cdot 1/|k| \cdot \exp[ik(r_i - r_j)] \quad (2.7)$$

3. THE MARTINELLI-MERLINI ARGUMENT REVISITED

We do not intend to repeat the Mermin argument for Coulomb systems as used in refs. 5-8 in every detail, but restrict ourselves to the necessary minimum of notions and notations. The usual point of departure is the modified Mermin inequality:

$$S(K+k) \geq \frac{[(K+k) e_i]^2 |\rho_K|^2}{(k e_i)^2 + D_{ii}(k)} \quad (3.1)$$

e_i is a unit vector pointing into an arbitrary direction which will be specified later. K is a vector of the reciprocal lattice G^* of the crystalline state, which is presumed to exist in the TL, and k is chosen from its first Brillouin zone $\cap A^* = L^{-1}G^*$ (A being the L^d -fold copy of the unit cell of the crystal lattice). Here

$$\rho(K) = \langle N^{-1} \sum_i \exp(iKr_i) \rangle \quad (3.2)$$

the Fourier transform of the particle density divided by N ,³ is assumed to be different from zero for at least one $K \neq 0$ in the TL if a crystalline state exists.

To elucidate the physical meaning of $S(q)$, we start from the averaged two-particle density

$$\rho g(r) := 1/N \cdot \int_A dr' \rho^{(2)}(r', r' - r) = \left\langle 1/N \sum_{i \neq j} \delta(r_i - r_j - r) \right\rangle \quad (3.3)$$

We transform this into an averaged charge-charge correlation (up to a factor q):

$$\rho h(r) = \rho g(r) - \rho \quad (3.4)$$

Fourier transforming (F.tr.) $\rho h(r)$, we get

$$\begin{aligned} S(q) - 1 &= \text{F.tr.}[\rho h(r)] \\ &= 1/N \cdot \left\langle \sum_{i \neq j} \exp[iq(r_i - r_j)] \right\rangle - N\delta_q \\ &= 1/N \left\langle \left| \sum_i [\exp(iqr_i) - \delta_q] \right|^2 \right\rangle - 1 \end{aligned} \quad (3.5)$$

³ With the convention $\rho(r)/\rho = \sum e^{iKr} \rho(K)$, implying $\rho(K) = 1/N \cdot \int_A \rho(r) e^{-iKr} dr$.

The last quantity to mention is

$$\begin{aligned}
 D_{ii}(k) = & \beta/N \cdot \left\langle \sum_{i,j} \exp[ik(r_i - r_j)] (e_i \nabla_i)(e_i \nabla_j) U \right\rangle \\
 & + \beta/N \cdot \int dr \rho(r) (e_i \nabla)^2 v_{\text{ex}}(r)
 \end{aligned} \tag{3.6}$$

with U defined in (2.4) [resp. (2.7)] and v_{ex} being an external field fixing the location of the crystal and which will be switched off after the TL has been performed.

The term stemming from the exterior field does not contribute if the limits are taken in the stated order. The first term can be written as

$$\begin{aligned}
 & \beta/\Lambda \cdot \sum_{p \neq 0} \Phi(p) (pe_i)^2 [S(p-k) - S(p)] \\
 & + \beta\rho \sum_{p \neq 0} \Phi(p) (pe_i)^2 [\delta_{p-k} - \delta_p]
 \end{aligned} \tag{3.7}$$

with

$$\Phi(p) = \begin{cases} 4\pi q^2/p^2 & \text{for ordinary jellium} \\ 2\pi q^2/|p| & \text{for surface jellium} \end{cases}$$

For $d \neq 1$, e_i can be chosen perpendicular to k . This eliminates the second term in (3.7). The remaining term can be expressed in position space as

$$\beta\rho \int_{\Lambda} d^d h(r) (1 - \cos kr) (e_i \nabla)^2 \phi_{\Lambda}(r) \tag{3.8}$$

with $\phi_{\Lambda}(r) = 1/\Lambda \cdot \sum_{k \neq 0} \Phi(k) e^{ikr}$ the effective pair potential occurring in (2.4), (2.7).

The strategy of Martinelli and Merlini has been to show that (3.8) goes as $\lesssim k^2 \ln |k|$ for small k in the TL under the proviso that $h(r)$ is both bounded and decaying $\lesssim r^{-2}$ uniformly in Λ . However, this proviso is definitely *not* true for states exhibiting crystalline order in the TL. This can be seen as follows: We write

$$\begin{aligned}
 \rho h(r) = & 1/N \cdot \int_{\Lambda} dr' \rho^{(2)}(r', r' - r) - \rho \\
 = & 1/N \cdot \int_{\Lambda} dr' \rho_T^{(2)}(r', r' - r) + 1/N \cdot \int_{\Lambda} dr' \rho^{(1)}(r') \rho^{(1)}(r' - r) - \rho
 \end{aligned} \tag{3.9}$$

Let $A \rightarrow \infty$ and afterward $r \rightarrow \infty$. If the TL is attained sufficiently uniformly,

$$\lim_{r \rightarrow \infty} \lim_{A \rightarrow \infty} 1/N \cdot \int_A dr' \rho_{T,A}^{(2)}(r', r' - r) = 0 \tag{3.10}$$

since in a pure phase $\rho_T^{(2)}(r_1, r_2)$ has to decay in the difference variable $(r_1 - r_2)$ anyhow.

In the TL the second term acquires the form (C_0 is the unit cell)

$$\begin{aligned} & 1/(\rho C_0) \cdot \int_{C_0} dr' \rho^{(1)}(r') \rho^{(1)}(r' - r) - \rho \\ & = \rho \sum_{K \neq 0} |\rho(K)|^2 e^{-iKr} \end{aligned} \tag{3.11}$$

If the equilibrium state is nontrivially periodic, $\rho(K) \neq 0$ for at least one $K \neq 0$. Due to the uniqueness theorem of Fourier series, we can infer that in that case $\rho h(r)$ is asymptotically *nontrivially* periodic as $r \rightarrow \infty$ (i.e., not decaying!). This shows that the procedure of ref. 8 breaks down at a central point. In the following section we will develop a strategy which, among other things, circumvents this snag.

4. IMPROVED BOUNDS FOR VARIOUS "JELLIUM SYSTEMS"

In a first step we split off in $\rho h(r)$ the term asymptotically oscillating in the TL, i.e.,

$$\begin{aligned} \rho h^{(o)}(r) & := 1/N \cdot \int_A dr' \rho^{(1)}(r') \rho^{(1)}(r' - r) - \rho \\ & = \rho \sum_{k \neq 0} |\rho(k)|^2 e^{-ikr} \end{aligned} \tag{4.1}$$

The corresponding contribution in $S(p) - 1$ is

$$S^{(o)}(p) := \text{F.tr.}(\rho h^{(o)}(r)) = N |\rho(p)|^2 - N \delta_p \tag{4.2}$$

and leads in (3.7) to the part

$$\begin{aligned} & \beta/A \cdot \sum_{p \neq 0} \Phi(p)(pe_i)^2 [S^{(o)}(p - k) - S^{(o)}(p)] \\ & = \beta \rho \sum_{p \neq 0} \Phi(p)(pe_i)^2 [|\rho(p - k)|^2 - |\rho(p)|^2] \\ & = \beta \rho \sum_{p \neq 0, -k} (pe_i)^2 |\rho(p)|^2 [\Phi(p + k) - \Phi(p)], \quad \text{with } p \in A^* \end{aligned} \tag{4.3}$$

Assuming sufficient uniformity (as above) for $A \rightarrow \infty$, this goes over in the TL into

$$\beta\rho \sum_{K \neq 0} (Ke_i)^2 \rho(K) \rho(-K) [\Phi(K+k) - \Phi(K)], \quad \text{where } K \in G^* \quad (4.4)$$

We now estimate (4.4) in the small- k limit both for ordinary and surface jellium. To this end, we expand $\Phi(K+k) - \Phi(K)$ into a Taylor series with remainder:

$$\Phi(K+k) - \Phi(K) = (k\nabla) \Phi(K) + 1/2 \cdot (k\nabla)^2 \Phi(K + \theta k) \quad (4.5)$$

This leads in the two cases to:

(i) $\Phi(K) = 4\pi q^2 / K^2$:

$$(k\nabla) \Phi(K) = -4\pi q^2 \cdot 2kK / |K|^4$$

$$\frac{1}{2}(k\nabla)^2 \Phi(p) = 4\pi q^2 [4(kp)^2 / |p|^6 - k^2 / |p|^4]$$

(ii) $\Phi(K) = 2\pi q^2 / |K|$:

$$(k\nabla) \Phi(K) = -2\pi q^2 kK / |K|^3$$

$$\frac{1}{2}(k\nabla)^2 \Phi(p) = \pi q^2 [3(kp)^2 / |p|^5 - k^2 / |p|^3]$$

(4.6)

In both cases $(k\nabla) \Phi(K)$ is antisymmetric with respect to K and thus does not contribute in (4.4). Inserting $(K + \theta k)$ for p , we have (with $|k| \ll |K|$)

$$|1/2 \cdot (k\nabla)^2 \Phi(K + \theta k)| < \begin{cases} \text{(i)} & \text{const} \cdot k^2 |K|^{-4} \\ \text{(ii)} & \text{const} \cdot k^2 |K|^{-3} \end{cases} \quad (4.7)$$

Hence (4.4) can be estimated for small k as

$$|(4.4)| \leq \begin{cases} \text{(i)} & \text{const} \cdot k^2 \sum_{K \neq 0} |\rho(K)|^2 |K|^{-2} \\ \text{(ii)} & \text{const} \cdot k^2 \sum_{K \neq 0} |\rho(K)|^2 |K|^{-1} \end{cases} \quad (4.8)$$

Remark. Since we have explicitly used that $e_i k = 0$, the above calculations hold only for $d \geq 2$. It will, however, turn out in the following that for $d = 1$ the above splitting of $h(r)$ into a decaying and an oscillating part is not necessary.

As to the remaining, now explicitly decaying part

$$\rho h^{(d)}(r) = \rho [h(r) - h^{(o)}(r)] = 1/N \cdot \int_A dr' \rho_T^{(2)}(r', r' - r) \quad (4.9)$$

our procedure is comparable to the one in ref. 8.

Note first that the decay of $h^{(d)}(r)$ as $|r| \rightarrow \infty$ is the same as the asymptotic behavior of $\rho_T^{(2)}(r_1, r_2)$ in the difference variable $r_1 - r_2$. We now replace $h(r)$ in (3.8) by $h^{(d)}(r)$ and treat the following four cases.

4.1. Ordinary Jellium; $d = 2$

We assume that $h^{(d)}(r)$ can be bounded uniformly in A both like

$$|h^{(d)}(r)| \leq C_1 \quad \text{and} \quad |h^{(d)}(r)| \leq \frac{C_2}{|r|^{1+\varepsilon}}, \quad \varepsilon > 0 \quad (4.10)$$

From the explicit form of $\varphi_A(r)$,

$$\varphi_A(r) = \frac{4\pi q^2}{A} \sum_{k \neq 0} \frac{1}{k^2} e^{ikr}; \quad k \in A^* \quad (4.11)$$

we get the following identity:

$$(e_t \nabla)^2 \varphi_A(r) = \frac{1}{L^2} (e_t \nabla_{r/L})^2 \varphi_{C_0}(r/L) \quad (4.12)$$

(just as in ref. 8).

In order to get a strong estimate of (4.12), one has to control the singularity of the above expression at the origin. The technical trick consists in subtracting a suitable function—which is given in closed form—from φ_{C_0} so that the difference is harmonic. This means especially (via “elliptic regularity”) that arbitrary derivatives remain bounded on C_0 . In the above case we have

$$\Delta \varphi_{C_0}(r) = -4\pi q^2 \left[\delta(r) - \frac{1}{C_0} \right]$$

and therefore

$$\Delta \left(\varphi_{C_0}(r) + 2q^2 \ln |r| - \frac{\pi q^2}{C_0} r^2 \right) \equiv 0 \quad (4.13)$$

Remark. Note that the choice made by Martinelli and Merlini,⁽⁸⁾ in contrast to their claim, does not lead to a harmonic function.

From (4.13) we now infer

$$\begin{aligned} \text{(i)} \quad & \left| \frac{1}{L^2} (e_t \nabla_{r/L})^2 \left[\varphi_{C_0} \left(\frac{r}{L} \right) + 2q^2 \ln \left| \frac{r}{L} \right| - \frac{\pi q^2}{C_0} \left(\frac{r}{L} \right)^2 \right] \right| \leq \frac{C_3}{L^2} \\ \text{(ii)} \quad & \left| \frac{1}{L^2} (e_t \nabla_{r/L})^2 \left[2q^2 \ln \left| \frac{r}{L} \right| - \frac{\pi q^2}{C_0} \left(\frac{r}{L} \right)^2 \right] \right| \leq \frac{C_4}{r^2} + \frac{C_5}{L^2} \end{aligned} \quad (4.14)$$

and therefore

$$|(e_i \nabla)^2 \varphi_A(r)| \leq \frac{C}{r^2} + \frac{C'}{L^2} \tag{4.15}$$

In a last step we have the following estimate:

$$\begin{aligned} & \left| \int_A d^2r h^{(d)}(r) (1 - \cos kr) (e_i \nabla)^2 \varphi_A(r) \right| \\ & \leq A_1 \int_A d^2r \frac{1}{|r|} |k| \cdot |r| \frac{1}{L^2} + A_2 \int_{|r| \leq 1/|k|} d^2r \frac{1}{|r|} k^2 r^2 \frac{1}{r^2} \\ & \quad + A_3 \int_{|r| \geq 1/|k|} d^2r \frac{1}{|r|} \cdot 2 \frac{1}{r^2} = O(|k|) \end{aligned} \tag{4.16}$$

as $|k| \rightarrow 0$ uniformly in A . The conclusion is

$$S(K+k) \geq \text{const} \cdot (Ke_t)^2 |\rho(K)|^2 \frac{1}{|k|} \quad \text{in the TL} \tag{4.17}$$

In (4.10) we assumed an upper bound on the decay of $h^{(d)}(r)$. We are now able to show by employing (4.17) that surprisingly the poor decay of (4.10) is still too fast to allow the existence of a $\rho(K) \neq 0$ for a $K \neq 0$! To this end, we proceed as follows: In a first step we extract that part from $S(K+k)$ which stems from $\rho h^{(o)}(r) = \rho_k \sum_{K \neq 0} |\rho(K)|^2 e^{-iKr}$ [see (3.11)], having the form (now with $p \in \mathbb{R}^2$)

$$S^{(o)}(p) = \text{const} \cdot \sum_{K \neq 0} |\rho(K)|^2 \delta(p - K) \tag{4.18}$$

This part evidently does not contribute in (4.17).

We then introduce a smooth nonnegative cutoff function $f_\varepsilon(k)$, localized in an ε -neighborhood of $k = 0$.

Since the usual Mermin argument relies on nonintegrable singularities, we rewrite (4.17), with $S^{(d)} := S - S^{(o)}$, as

$$S^{(d)}(K+k) \frac{f_\varepsilon(k)}{|k|} \geq \text{const} \cdot (Ke_t)^2 |\rho(K)|^2 \frac{f_\varepsilon(k)}{k^2} \tag{4.19}$$

Integrating now with respect to k , we get on the lhs

$$\begin{aligned}
 & \int d^2k [S^{(d)}(K+k) - 1] \frac{f_\varepsilon(k)}{|k|} + \int d^2k \frac{f_\varepsilon(k)}{|k|} \\
 &= \int d^2r e^{iKr} h^{(d)}(r) F(r) + \int d^2k \frac{f_\varepsilon(k)}{|k|} \\
 &\leq a_1 + a_2 \int_{|r| \geq R_0} d^2r \frac{1}{|r|^{1+\varepsilon}} \frac{1}{|r|} < \infty \tag{4.20}
 \end{aligned}$$

where $F(r)$ is, up to a multiplicative constant, the Fourier transform of $f_\varepsilon(k)/|k|$, which, by asymptotic Fourier analysis (cf., e.g., ref. 15), behaves as $|r|^{-1}$ for large $|r|$. On the other hand, an integration with respect to k would lead to an infinite rhs in (4.19) if $|\rho(K)| \neq 0$ for some $K \neq 0$. In other words, crystallinity is incompatible with a decay of $|h^{(d)}(r)| \lesssim |r|^{-(1+\varepsilon)}$, $\varepsilon > 0$!

4.2. Ordinary Jellium; $d = 3$

We now assume uniformly in \mathcal{A}

$$|h^{(d)}(r)| \leq C_1 \quad \text{and} \quad |h^{(d)}(r)| \leq \frac{C_2}{|r|^{3/2+\varepsilon}} \tag{4.21}$$

Instead of (4.12) we get

$$(e_i \nabla)^2 \varphi_{\mathcal{A}}(r) = (1/L^3) (e_i \nabla_{r/L})^2 \varphi_{C_0}(r/L) \tag{4.22}$$

Among other things, $2 \ln |r|$ in (4.13) has to be replaced with $-1/|r|$, and one is finally led to the estimate

$$|(e_i \nabla)^2 \varphi_{\mathcal{A}}(r)| \leq \frac{C}{|r|^3} + \frac{C'}{L^3} \tag{4.23}$$

The analog of (4.16) is

$$\begin{aligned}
 & \left| \int_{\mathcal{A}} d^2r h^{(d)}(r) (1 - \cos kr) (e_i \nabla)^2 \varphi_{\mathcal{A}}(r) \right| \\
 & \leq A_1 \int_{\mathcal{A}} d^3r \frac{1}{|r|^{3/2}} |k|^{3/2} |r|^{3/2} \frac{1}{L^3} \\
 & \quad + A_2 \int_{|r| \leq 1/|k|} d^3r \frac{1}{|r|^{3/2}} k^2 r^2 \frac{1}{|r|^3} \\
 & \quad + A_3 \int_{|r| \geq 1/|k|} d^3r \frac{1}{|r|^{3/2}} 2 \frac{1}{|r|^3} \\
 & = O(|k|^{3/2}) \quad \text{as} \quad |k| \rightarrow 0 \tag{4.24}
 \end{aligned}$$

leading, in the TL, to the inequality

$$S^{(d)}(K+k) \frac{f_\varepsilon(k)}{|k|^{3/2}} \geq \text{const} \cdot (Ke_t)^2 |\rho(K)|^2 \frac{f_\varepsilon(k)}{|k|^3} \tag{4.25}$$

Since the Fourier transform of $f_\varepsilon(k)/|k|^{3/2}$ behaves asymptotically as $|r|^{-3/2}$ for large $|r|$, the k integral on the lhs is finite due to our assumption (4.21). As in (a) above, we arrive at the conclusion that crystallinity is incompatible with a decay of $|h^{(d)}(r)| \lesssim |r|^{-(3/2+\varepsilon)}$

4.3. Surface Jellium; $d = 2$

We assume that $h^{(d)}(r)$ can be bounded uniformly in A both like

$$|h^{(d)}(r)| \leq C_1 \quad \text{and} \quad |h^{(d)}(r)| \leq \frac{C_2}{|r|^{1/2+\varepsilon}} \tag{4.26}$$

The effective potential $\varphi_A(r)$ reads

$$\varphi_A(r) = \frac{2\pi q^2}{A} \sum_{k \neq 0} \frac{1}{|k|} e^{ikr} = \tilde{\varphi}_A(r, z=0) \tag{4.27}$$

with

$$\tilde{\varphi}_A(r, z) := \frac{2\pi q^2}{A} \sum_{k \neq 0} \frac{1}{|k|} e^{-|k| \cdot |z|}$$

The potential $\tilde{\varphi}_A$ is introduced in order to exploit harmonicity arguments in $d=3$ rather than $d=2$.

In analogy to case (a), we get

$$\begin{aligned} (e_t \nabla_r)^2 \tilde{\varphi}_A(r, z) &= \frac{1}{L^3} (e_t \nabla_{r/L})^2 \tilde{\varphi}_{C_0} \left(\frac{r}{L}, \frac{z}{L} \right) \\ \left(A_r + \frac{\partial^2}{\partial z^2} \right) \tilde{\varphi}_{C_0}(r, z) &= -4\pi q^2 \left[\delta(r) - \frac{1}{C_0} \right] \delta(z) \\ \left(A_r + \frac{\partial^2}{\partial z^2} \right) \left[\tilde{\varphi}_{C_0}(r, z) - \frac{q^2}{(r^2+z^2)^{1/2}} - \frac{2\pi q^2}{C_0} |z| \right] &\equiv 0 \end{aligned} \tag{4.28}$$

and therefore

$$|(e_t \nabla)^2 \varphi_A(r)| = |(e_t \nabla_r)^2 \tilde{\varphi}_A(r, z=0)| \leq \frac{C}{|r|^3} + \frac{C'}{L^3} \tag{4.29}$$

As in (4.16), (4.24), this yields (with the same splitting of the integration domain)

$$\left| \int_A d^2r h^{(d)}(r)(1 - \cos kr)(e_r \nabla)^2 \varphi_A(r) \right| \leq \text{const} \cdot |k|^{3/2} \tag{4.30}$$

uniformly in A .

Proceeding as in (a), (b), we have now in the TL

$$S^{(d)}(K+k) \frac{f_\varepsilon(k)}{|k|^{1/2}} \geq \text{const} \cdot (Ke_\varepsilon)^2 |\rho(K)|^2 \frac{f_\varepsilon(k)}{k^2} \tag{4.31}$$

Since the Fourier transform of $f_\varepsilon(k)/|k|^{1/2}$ behaves as $|r|^{-3/2}$ for large $|r|$, (4.26) leads to a contradiction if $\rho(K) \neq 0$ for some $K \neq 0$. In other words, crystallinity is excluded for $|h^{(d)}(r)| \lesssim |r|^{-(1/2+\varepsilon)}$!

4.4. Surface Jellium; $d = 1$

As mentioned above, a disentangling of directions parallel and orthogonal to k is of course not possible in $d = 1$. On the other hand, it will turn out that this procedure—as the splitting of h into $h^{(o)} + h^{(d)}$ —is not necessary anyhow. Instead we only use uniform boundedness of h as $A \rightarrow \mathbb{R}$.

Again we start from expression (3.7):

$$\begin{aligned} & \frac{2\pi q^2}{A} \beta \sum_{p \neq 0} |p| [S(p-k) - S(p)] + 2\pi q^2 \beta \rho \sum_{p \neq 0} |p| (\delta_{p-k} - \delta_p) \\ &= \beta \rho \int_A dr h(r)(1 - \cos kr) \frac{d^2}{dr^2} \varphi_A(r) + 2\pi q^2 \beta \rho |k| \end{aligned} \tag{4.32}$$

The same harmonicity arguments as in (4.27) and what followed lead to the estimate

$$\left| \frac{d^2}{dr^2} \varphi_A(r) \right| \leq \frac{C}{r^2} + \frac{C'}{L^2} \tag{4.33}$$

and we get

$$\left| \int_A dr h(r)(1 - \cos kr) \frac{d^2}{dr^2} \varphi_A(r) \right| = O(|k|) \tag{4.34}$$

as $|k| \rightarrow 0$ uniformly in A .

The analog of (4.17) reads now

$$S(K+k) \geq \text{const} \cdot K^2 |\rho(K)|^2 \frac{1}{|k|} \quad (4.35)$$

the rhs of which is already nonintegrable at $k=0$.

Thus, crystallinity is excluded by standard arguments.

5. SUMMARY

As already remarked in the introduction, our analysis has shown that, in order to exclude crystallinity, one indeed arrives, by means of the techniques presented above, at the considerably stronger estimates

$$\begin{aligned} |\rho_T^{(2)}(r_1, r_2)| &\lesssim \text{(a)} \quad |r_1 - r_2|^{-(1+\varepsilon)} \\ &\text{(b)} \quad |r_1 - r_2|^{-(3/2+\varepsilon)} \\ &\text{(c)} \quad |r_1 - r_2|^{-(1/2+\varepsilon)} \\ &\text{(d)} \quad |r_1 - r_2|^0 \end{aligned}$$

as compared to the older ones given in ref. 9, i.e.,

$$\begin{aligned} |\rho_T^{(2)}| \quad \text{or} \quad |\rho_T^{(3)}| &\lesssim \text{(a)} \quad |r_1 - r_2|^{-(3+\varepsilon)} \\ &\text{(b)} \quad |r_1 - r_2|^{-(4+\varepsilon)} \\ &\text{(c)} \quad |r_1 - r_2|^{-(3+\varepsilon)} \\ &\text{(d)} \quad |r_1 - r_2|^{-(2+\varepsilon)} \end{aligned}$$

The merit of the method of Martinelli and Merlini which we partly have employed in this paper is that it avoids uncontrollable manipulations with possibly ill-defined expressions. However, their approach contained a serious flaw due to the fact that the correlation function $h(r)$ does not decay as $|r| \rightarrow \infty$. One of our contributions consists in the observation that $h(r)$ can be naturally split into a nondecaying oscillatory and a decaying part which can be estimated separately in k (resp. r) space. Furthermore, we improve their approach to derive, up to logarithmic corrections, possibly optimal bounds also for the case $d=3$ and for the surface jellia.

As a last point we want to briefly comment on the observation of Alastuey and Jancovici⁽⁷⁾ that a two-dimensional surface plasma should not be able to crystallize into a triangular lattice. To arrive at this conclu-

sion, the authors employed some extra hypotheses which, admittedly, look plausible from a physical point of view. Under this proviso they get an asymptotic behavior $D_{ii}(k) \approx Ck^2$ as $|k| \rightarrow 0$, which excludes crystallinity via the usual Mermin argument. Their reasoning may be supported by their observation that the prefactor C turns out to be, in the however special case of a triangular lattice symmetry, a multiple of the excess internal energy per particle, which may be expected to remain finite if the thermodynamic limit exists at all.

On the other hand, the definitely not decaying $h(r)$, consisting of the oscillating $h^{(o)}(r)$ and the (as we have shown above) exceedingly slowly decaying $h^{(d)}(r)$, appears in all relevant integral expressions of ref. 7 such as those for $D_{ii}(k)$ or the internal energy per particle. This makes it necessary that very special cancellations have to take place in these integrals due to suitable oscillations not only of $h^{(o)}(r)$, but also of $h^{(d)}(r)$! That this may happen can be inferred from observations we made elsewhere in a related context (cf. ref. 17 p. 193–194, where the “freezing problem” was briefly addressed, or p. 202 in connection with the surface tension).

Nevertheless, we must admit that we feel still a little bit uneasy, since intuition may well be deceiving in the case of quantities with a very slow decay. Thus it might happen that (e.g., due to effects of something like “observables at infinity”) the “naive” energy per particle acquires some extra term. However, as long as the thermodynamic limit is not really under control, these considerations remain mere speculations.

We conclude this paper with the remark that similar features [(non)-analyticities and (non)integrable singularities] also occur in another field of current active research, namely the theory of liquid–vapor interfaces in $d=3$ (cf. ref. 16 and references given there).

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