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Absence of Long-Range Order with Long-Range Potentials

Marc Baus[1,2]

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A particular form of Mermin's inequality is analyzed for repulsive inverse power potentials $[V(r) = e^2 r^{-m}/m]$ in a d-dimensional space. For long-range potentials ($m \leqslant d$) the system is put into a stabilizing background. Long-range order is shown to be excluded for $d \leqslant (m + 2)/2$ when $m \leqslant d$, while for short-range potentials ($m > d$) we recover Mermin's result ($d \leqslant 2$). For Coulomb systems ($m = d - 2$) and the experimentally studied electron surface layer ($d = 2$, $m = 1$), long-range order cannot be excluded by the present argument.

KEY WORDS: Long-range order; Coulomb system; electron surface layer; Mermin's inequality.

1. INTRODUCTION

In 1936 it was pointed out by Peierls[(1)] that the mean-square deviation of an atom from its equilibrium position diverges in one and two dimensions for an infinite harmonic crystal. Somewhat later Landau[(2)] gave a general but *macroscopic* argument according to which fluctuations will destroy crystalline order possessing only a one- or two-dimensional periodicity. These conclusions were criticized, for instance, by Frenkel.[(3)] By including higher order terms with respect to the fluctuations Landau[(4)] later revised his result for the case of two-dimensional periodicity. Nowadays one often finds in the literature general statements according to which long-range order resulting from the breaking of a continuous symmetry will be destroyed by the fluctuations in one and two dimensions. Although this will often be the case, questions regarding the validity and the practical relevance of this statement generally cannot be overlooked. The first *microscopic* treatment of this problem for classical systems (to which the present considerations are

[1] Chimie–Physique II, Université Libre de Bruxelles, Bruxelles, Belgium.
[2] Chercheur Qualifié du Fonds National Belge de la Recherche Scientifique.

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restricted) is due to Mermin.[5] As is well known, the divergences found by Mermin in one and two dimensions are so weak that their practical relevance can be questioned. With respect to the general validity of the above statement, it should also be observed that Mermin's proof is explicitly restricted to short-range potentials. It is our purpose here to show that Mermin's conclusions are indeed sensitive to the range of the potential and hence that they are not completely general. To this end we will consider one-component systems of charged particles embedded into a stabilizing background. The particles are supposed to interact through a repulsive inverse-power pair potential. This includes the genuine Coulomb systems[6] as well as the electron surface layer to which much interest has been devoted recently.[6,7] With respect to the latter system it has been conjectured[8] that, although a proof was lacking, the general Peierls–Landau–Mermin argument should still be applicable. Below we will show that this is not the case. The major point is that Bogoliubov's $1/k^2$ singularity is weakened for long-range potentials and transformed in the latter system into a $1/k$ singularity. Consequently long-range crystalline order in an infinite electron sheet cannot be excluded by the Peierls–Landau–Mermin argument.

2. THE INEQUALITIES

We start from the well-known Schwarz inequality

$$\langle |A|^2\rangle\langle |B|^2\rangle \geqslant |\langle A^*B\rangle|^2 \tag{1}$$

where A^* denotes the complex conjugate of A, $|A|^2 = AA^*$, while $\langle\cdots\rangle$ stands for the canonical equilibrium average with respect to the Hamiltonian H at the inverse temperature $\beta = (k_B T)^{-1}$. As was shown by Mermin,[9] the classical version of Bogoliubov's inequality, namely

$$\langle |A|^2\rangle\langle [C, [C^*, \beta H]]\rangle \geqslant |\langle [C, A^*]\rangle|^2 \tag{2}$$

can be obtained from (1) by taking B equal to the Poisson bracket of C with H, $B = [C, H]$, and neglecting surface terms in momentum and position space. Although the present problem can be studied[10] on the basis of Eq. (2), a slightly different inequality was proposed by Mermin.[5] His result can be recovered from (1) and (2) by preaveraging the Poisson bracket $[C, H]$ over the momentum variables before substituting the result into (1). Hence, taking $A = \sum_j \psi(\mathbf{r}_j)$ and $\mathbf{C} = \sum_j \mathbf{p}_j\varphi(\mathbf{r}_j)$, with $\mathbf{r}_j$ and $\mathbf{p}_j$ the position and momentum of particle j, while the functions $\psi(\mathbf{r})$ and $\varphi(\mathbf{r})$ are such as to lead to vanishing surface terms, and defining $\mathbf{B} = \langle[\mathbf{C}, H]\rangle_K$, where $\langle\cdots\rangle_K$ stands for the momentum average with respect to the kinetic energy K of $H = K + U$, one obtains from the vector analog of (1), after neglecting spatial surface terms, the inequality originally proposed by Mermin[5]:

$$\left\langle\left|\sum_j \psi(\mathbf{r}_j)\right|^2\right\rangle\left[\left\langle\sum_j\left|\nabla_j\varphi(\mathbf{r}_j)\right|^2\right\rangle + \left\langle\sum_{i,j}\varphi(\mathbf{r}_i)\varphi^*(\mathbf{r}_j)\,\nabla_i\cdot\nabla_j\beta U\right\rangle\right]$$
$$\geqslant \left|\left\langle\sum_j \varphi(\mathbf{r}_j)\,\nabla_j\psi^*(\mathbf{r}_j)\right\rangle\right|^2 \tag{3}$$

which differs from Bogoliubov's inequality because

$$|\langle[\mathbf{C}, H]\rangle_K|^2 \neq \langle|[\mathbf{C}, H]|^2\rangle_K$$

Here and below all particle summations ($\sum_j$) run from $j = 1$ to $j = N$.

Next, we enclose the system of N particles into a d-dimensional cube of volume $\Omega = L^d$ with periodic boundary conditions. The particles are assumed to interact through a pair potential $V(|\mathbf{r}|)$, which can be represented by the following Fourier series:

$$V(|\mathbf{r}|) = \Omega^{-1}\sum_{\mathbf{k}} v(\mathbf{k})\exp(i\mathbf{k}\cdot\mathbf{r}) \tag{4}$$

with $\mathbf{k} = (2\pi/L)\mathbf{n}$, the d components of $\mathbf{n}$ being integers. The potential energy U can then be written as

$$U = U_0 + \frac{1}{2\Omega}\sum_{\mathbf{k}}\bar{v}(\mathbf{k})\sum_{\substack{i,j\\ i\neq j}}\exp[i\mathbf{k}\cdot(\mathbf{r}_i - \mathbf{r}_j)] \tag{5}$$

where U_0 is a constant. For the long-range, repulsive, Coulomb-like potentials we have in mind the system will not explode only when it is put into an oppositely "charged," smeared-out background, as is customary for one-component plasmas. We will assume this to be the case. The background then removes the $k = 0$ term from the summation in Eq. (4), or equivalently we have introduced in Eq. (5) the modified potential $\bar{v}(\mathbf{k})$:

$$\bar{v}(\mathbf{k}) = \begin{cases} 0, & \mathbf{k} = \mathbf{0} \\ v(\mathbf{k}), & \mathbf{k} \neq \mathbf{0} \end{cases} \tag{6}$$

whereas U_0 now becomes the Madelung energy of a cubic lattice with L as the spacing.[11] For short-range potentials (i.e., integrable at large distances) no background is required and we can use Eq. (5) with $\bar{v}(\mathbf{k}) = v(\mathbf{k})$ and $U_0 = 0$. To obtain the final inequality we take in Eq. (3)

$$\psi(\mathbf{r}) = \exp(i\mathbf{k}'\cdot\mathbf{r}) - \delta_{\mathbf{k}'} \qquad \text{and} \qquad \varphi(\mathbf{r}) = \exp(i\mathbf{k}\cdot\mathbf{r})$$

differing from Mermin's choice[5] mainly by the presence of the Kronecker delta $\delta_{\mathbf{k}}$ ($\delta_{\mathbf{k}} = 1$ if $\mathbf{k} = \mathbf{0}$, $\delta_{\mathbf{k}} = 0$ if $\mathbf{k} \neq \mathbf{0}$). For long-range potentials (i.e., not integrable at infinity), for which $v(k)$ is singular for small k, this difference is not immaterial. We now obtain from Eq. (3) for $k \neq 0$ (i.e., $\langle|\mathbf{B}|^2\rangle \neq 0$) the basic inequality:

$$S(\mathbf{k}') \geqslant \frac{|\mathbf{k}'|^2|\rho_{\mathbf{k}-\mathbf{k}'}|^2}{\mathbf{k}^2 + \nu(\mathbf{k}) + I(\mathbf{k})} \geqslant 0 \tag{7}$$

where $\rho_{\mathbf{k}}$ is the order parameter[5]:

$$\rho_{\mathbf{k}} = N^{-1}\left\langle \sum_j \exp(i\mathbf{k}\cdot\mathbf{r}_j) \right\rangle \tag{8}$$

$S(\mathbf{k})$ is the static structure factor:

$$S(\mathbf{k}) = N^{-1}\left\langle \left|\sum_j [\exp(i\mathbf{k}\cdot\mathbf{r}_j) - \delta_{\mathbf{k}}]\right|^2 \right\rangle \tag{9}$$

while $\nu(\mathbf{k})$ and $I(\mathbf{k})$ are defined by

$$\nu(\mathbf{k}) = \beta N \Omega^{-1} \sum_{\mathbf{q}} q^2 \bar{v}(\mathbf{q})(\delta_{\mathbf{k}-\mathbf{q}} - \delta_{\mathbf{q}}) \tag{10}$$

$$I(\mathbf{k}) = \beta \Omega^{-1} \sum_{\mathbf{q}} q^2 \bar{v}(\mathbf{q})[h(\mathbf{k} - \mathbf{q}) - h(\mathbf{q})] \tag{11}$$

with $S(\mathbf{k}) = 1 + h(\mathbf{k})$. From Eq. (5) and (9) we also have that

$$\langle (U - U_0)/N \rangle = (1/2\Omega) \sum_{\mathbf{k}} \bar{v}(\mathbf{k})[h(\mathbf{k}) + N\delta_{\mathbf{k}}] \tag{12}$$

which clearly shows that $\bar{v}(k = 0)$ has to be finite (short-range potentials) or to be removed by the background (long-range potentials) in order to obtain a finite result for the average energy per particle.

3. SOME APPLICATIONS

Two types of applications of the basic inequality (7) will be considered here. First we take $\mathbf{k}' = \mathbf{k}$, in which case (7) reduces to

$$S(\mathbf{k}) \geqslant k^2/[k^2 + \nu(\mathbf{k}) + I(\mathbf{k})] \geqslant 0 \tag{13}$$

which will be used below to obtain some information on the static structure factor $S(\mathbf{k})$. Next we take $\mathbf{k}' = \mathbf{k} + \mathbf{b}$ with $\mathbf{b}$ a nonzero reciprocal lattice vector, multiply both sides of (7) with a positive function $f(|\mathbf{k}|)$, and sum over all $\mathbf{k}$ values:

$$\Omega^{-1} \sum_{\mathbf{k}} S(\mathbf{k} + \mathbf{b}) f(|\mathbf{k}|) \geqslant |\rho_{\mathbf{b}}|^2 \Omega^{-1} \sum_{\mathbf{k}} \frac{|\mathbf{k} + \mathbf{b}|^2 f(|\mathbf{k}|)}{k^2 + \nu(\mathbf{k}) + I(\mathbf{k})} \geqslant 0 \tag{14}$$

If in the thermodynamic limit (TL: $N, \Omega \to \infty, N\Omega^{-1} \to n$) we have

$$\mathrm{TL}\, \Omega^{-1} \sum_{\mathbf{k}} S(\mathbf{k} + \mathbf{b}) f(|\mathbf{k}|) < \infty \tag{15}$$

whereas

$$\mathrm{TL}\, \Omega^{-1} \sum_{\mathbf{k}} \frac{|\mathbf{k} + \mathbf{b}|^2 f(|\mathbf{k}|)}{k^2 + v(\mathbf{k}) + I(\mathbf{k})} = \infty \tag{16}$$

then (14) implies TL $\rho_{\mathbf{b}} = 0$, which is the standard argument[5] for the absence of crystalline order in the infinite system. In order to establish the sensitivity of Eq. (16) to the range of the potential, it will be sufficient to consider repulsive inverse power potentials of the form $V(r) = e^2 r^{-m}/m$, e being the "charge" of the particles. This includes the Coulomb potential ($m = d - 2$) and the electron surface layer ($d = 2$, $m = 1$) as the most interesting cases. What will be needed here is $v(\mathbf{k})$ for large Ω or in the TL, the Fourier transform of $V(r)$. For many m values of interest, e.g., Coulomb systems, only the generalized Fourier transform of $V(r)$ exists (see Ref. 12 for a very readable account). Explicitly one has for $0 < m < d$ and by analytic continuation[12] also for the other values of m and d:

$$v(k) = \begin{cases} e^2 Z(d, m) k^{m-d}, & m - d \neq 2p \quad \text{(17a)} \\ e^2 X(d, m) k^{m-d}[1 + Y(d, m) \ln k], & m - d = 2p \quad \text{(17b)} \end{cases}$$

with $p = 0, 1, 2, \ldots$. Here it will be sufficient to quote[12] the value of $Z(d, m)$:

$$Z(d, m) = 2^{d-m-1} \pi^{d/2} \Gamma\left(\frac{d-m}{2}\right) \Big/ \Gamma\left(\frac{m+2}{2}\right) \tag{18}$$

where Γ is Euler's gamma function. From Eqs. (17a) and (18) the well-known results $v(k) = 4\pi e^2/k^2$ and $v(k) = 2\pi e^2/k$ are easily recovered for $m = 1$ and respectively $d = 3$ and $d = 2$. Here we can however consider m and d to be continuous real variables.

4. UPPER BOUNDS

Before analyzing what becomes of Bogoliubov's $1/k^2$ singularity, let us make sure that no other divergences will arise in the TL. For the short-range case ($m > d$) Mermin has already shown[5] that the various quantities appearing in (14) will remain bounded in the TL. In the long-range case we can make the following statements. Consider first (15). Adapting an argument of Sorokina,[10] we can write

$$\Omega^{-1} \sum_{\mathbf{k}} S(\mathbf{k} + \mathbf{b}) f(|\mathbf{k}|) = \frac{1}{N} \sum_{\substack{i,j \\ i \neq j}} \langle [\exp(i\mathbf{b} \cdot \mathbf{r}_{ij})] F(|\mathbf{r}_{ij}|) \rangle + F(r = 0) - \frac{N}{\Omega} f(\mathbf{k} = -\mathbf{b}) \tag{19}$$

where $F(|\mathbf{r}|)$ is the Fourier transform of $f(|\mathbf{k}|)$:

$$F(\mathbf{r}) = \Omega^{-1} \sum_{\mathbf{k}} f(\mathbf{k}) \exp(i\mathbf{k}\cdot\mathbf{r}) \tag{20a}$$

$$f(\mathbf{k}) = \int_{\Omega} d\mathbf{r}\, F(\mathbf{r}) \exp(-i\mathbf{k}\cdot\mathbf{r}) \tag{20b}$$

Since $f(|\mathbf{k}|)$ is positive we can rewrite Eq. (19) as

$$\Omega^{-1} \sum_{\mathbf{k}} S(\mathbf{k} + \mathbf{b}) f(|\mathbf{k}|) \leqslant \sum_{j \neq 1} \langle F(|\mathbf{r}_{1j}|)\rangle + F(0) \tag{21}$$

Now if the system is in a crystalline state, then the distance between two neighboring particles $|\mathbf{r}_{12}|$ is bounded from below, $|\mathbf{r}_{12}| > \alpha a$, by a fraction α, however small ($\alpha \neq 0$), of the lattice spacing a. Hence $|\mathbf{r}_{ij}| > \alpha a|\mathbf{i} - \mathbf{j}|$, where $\mathbf{j}$ is a vector with integer components labeling the equilibrium position of particle j. If for $F(|\mathbf{r}|)$ we select a monotonically decreasing positive function of $|\mathbf{r}|$, for instance a Gaussian, then we can replace $|\mathbf{r}_{1j}|$ in (21) by its lower bound and rewrite (21) as

$$\Omega^{-1} \sum_{\mathbf{k}} S(\mathbf{k} + \mathbf{b}) f(|\mathbf{k}|) \leqslant \sum_{\mathbf{j}} F(\alpha a|\mathbf{j}|) \leqslant \int d\mathbf{x}\, F(\alpha a|\mathbf{x}|) \equiv f(\mathbf{k} = 0)/(\alpha a)^d \tag{22}$$

which is finite for $\alpha \neq 0$ and hence (15) can always be realized in a solid phase whatever the potential which gave rise to it.

Consider now Eq. (16). From Eq. (10) we obtain

$$\nu(k) \underset{\mathrm{TL}}{=} \beta n k^2 v(k) \equiv \omega^2(k)/v_0^{\,2} \tag{23}$$

where $\omega(k)$ is the "plasma frequency" of a system of particles of thermal velocity $v_0 = (\beta M)^{-1/2}$, M being their mass. Notice that for short-range potentials $v(k = 0)$ will be finite and hence $\omega(k)$ will be soundlike and there is no reason in this case to separate it from $I(\mathbf{k})$. For $I(\mathbf{k})$ we obtain from (11)

$$I(\mathbf{k}) \underset{\mathrm{TL}}{=} \beta n \int d\mathbf{r}\, h(\mathbf{r})(1 - \exp i\mathbf{k}\cdot\mathbf{r})\, \nabla^2 V(r) \tag{24}$$

where $nh(\mathbf{r})$ is the Fourier transform of $h(\mathbf{k})$. Equation (24) differs from the corresponding quantity considered by Mermin[5] because of the appearance of $h(\mathbf{r})$ in it instead of the total correlation function $g(\mathbf{r}) = 1 + h(\mathbf{r})$. This difference is due to the presence of the background and becomes immaterial for short-range forces. Returning to (16), we see that if we take for $f(|\mathbf{k}|)$ a rapidly decreasing function of k, we only have to inquire for the behavior of the integrand of (16) for small k. As $h(\mathbf{k}) = h(-\mathbf{k})$, we have from Eq. (24) that $I(\mathbf{k}) = k^2C + O(k^4)$, with C given by

$$C = \tfrac{1}{2}n\beta \int d\mathbf{r}\, h(\mathbf{r})(\hat{\mathbf{k}}\cdot\mathbf{r})^2\, \nabla^2 V(r) \tag{25}$$

$$= (m/2d)(m + 2 - d)\beta\langle(U - U_0)/N\rangle_{\mathrm{TL}} \tag{26}$$

where in Eq. (26) we have used the fact that $V(r)$ is an inverse-power potential ($\sim r^{-m}$), whereas C cannot depend on the direction $\hat{\mathbf{k}}$ of $\mathbf{k}$ if the orientation of the crystal in space has not been fixed. Hence $I(\mathbf{k})$ will be of order k^2 for small k *provided* the average energy per particle $\langle(U - U_0)/N\rangle$ remains bounded in the TL. The latter property can be established rigorously both for the short-range potentials[5,6] and for the genuine Coulomb systems.[13,14] For the long-range inverse-power potentials considered here we retain from the latter proofs[14] that $\langle(U - U_0)/N\rangle$ can be bounded by the self-energy of finite cells. These self-energies will be finite whenever the potential is integrable at the origin, i.e., in the present case for $m < d$.

For all values of m (except eventually the boundary value $m = d$) we therefore expect $I(\mathbf{k})$ to behave for small k as k^2C with C finite. Hence the whole difference between the short-range and long-range potentials considered here ($\sim r^{-m}$) stems from the presence of the plasma frequency $\omega(k)$ of Eq. (23) in the basic inequality (7). For long-range potentials $\omega(k)$ will cease to be soundlike and will weaken Bogoliubov's $1/k^2$ singularity, as will now be seen.

5. RESULTS

Let us gradually decrease the value of m and inquire for the implications of Eqs. (13) and (14).

$m > d$. This corresponds to the short-ranged potentials already considered by Mermin.[5] Here $v(k = 0) = 0$ is automatically satisfied and no stabilizing background is needed. Moreover, $\nu(k)$ vanishes faster for small k than the k^2 terms, i.e., $k^2 + \nu(k) + I(k) \simeq k^2(1 + C)$, and we recover Bogoliubov's $1/k^2$ singularity, so that (16) is satisfied for $d \leqslant 2$, which is Mermin's result.[5]

As an interesting by-product, we observe that (13) implies that

$$\lim_{k\to 0} S(k) \equiv (n/\beta)\chi_T \geqslant (1 + C)^{-2} \tag{27}$$

yielding a lower bound for the isothermal compressibility χ_T in terms of C or via Eq. (26) in terms of the internal energy.

$d \geqslant m > d - 2$. This domain corresponds to long-range potentials decaying faster at large r than the genuine Coulomb potentials. This region includes the realistic case $d = 2$, $m = 1$ describing the electron sheets considered recently by various authors.[6–8] Here $v(k)$ exhibits a logarithmic ($m = d$) or algebraic ($m < d$) singularity for small k [see (17)]. In this case the background is essential in order to keep the system stable. The major point here is that $\nu(k)$ vanishes more *slowly* for small k than the k^2 terms [$\nu(k) \sim k^{2+m-d}$ for $m < d$] and Bogoliubov's $1/k^2$ singularity becomes a weaker $1/k^{2+m-d}$ singularity. Equation (16) is now satisfied for $d \leqslant (m + 2)/2$, i.e., $d \leqslant 2 - (d - m)$, as compared to $d \leqslant 2$ for the short-range forces, showing that (quite naturally) long-range forces ($d - m > 0$) make it more difficult to exclude long-range order. Notice also that here the plasma mode remains a low-frequency mode since $\omega(k)$ still vanishes with k. It is noteworthy that the peculiar dispersion relation $\omega(k) \sim k^{1/2}$ pertaining to the aforementioned electron sheet ($d = 2$, $m = 1$) has been checked experimentally.[6] Returning to (13), we also obtain for small k that $S(k) \geqslant [\beta n v(k)]^{-1}$ and observe that the equality corresponds precisely to the small-k behavior conjectured elsewhere[15] for $S(\mathbf{k})$ on quite different grounds.

$m = d - 2$. For this particular value of m, $V(r)$ is the fundamental solution of the d-dimensional Poisson equation:

$$\nabla^2 V(|\mathbf{r}|) = -Z(d, d-2)e^2\,\delta(\mathbf{r}) \tag{28}$$

i.e., $V(r) = e^2 r^{2-d}/(d-2)$ for $d \neq 2$ [and $V(r) = e^2 \ln r^{-1}$ for $d = 2$ by analytic continuation of the Fourier transform[12]]. For this genuine Coulomb case, $\omega(k)$ turns out to be a constant independent of k, whereas $I(\mathbf{k})$ vanishes identically [see Eqs. (24) and (28)]. Consequently, the Bogoliubov singularity is completely suppressed here and long-range order cannot be excluded[3] on the basis of (7) for any $d > 0$. The results obtained elsewhere by Mermin[16] for the $m = 1$, $d = 3$ Coulomb potential can now be easily generalized to variable d. From (13) we see that $S(\mathbf{k})$ is still bounded from below (for all k) by its "Debye–Hückel" approximation:

$$S(\mathbf{k}) \geqslant k^2/(k^2 + k_D{}^2) \tag{29}$$

where the d-dimensional generalization of the Debye wave number k_D and the corresponding plasma frequency $\omega_p = v_0 k_D$ can be obtained from the definition $k_D{}^2 = \beta e^2 n Z(d, d-2)$ together with Eq. (18). Using (29) in

[3] A more detailed analysis than the one provided by the Mermin inequality has been performed by Kunz[17] for the genuine Coulomb case in $d = 1$. His results indicate that even in the thermodynamic limit the system's state will depend on the applied boundary conditions. For fixed wall boundary conditions the system will be in a crystalline state, whereas it will be in a translationally invariant state for periodic boundary conditions.

Eq. (12) we also obtain the following generalization of Mermin's lower bound for the internal energy:

$$\left\langle \frac{U - U_0}{N} \right\rangle \geqslant -\frac{e^2}{4}(2\pi)^{-d}[Z(d, d - 2)]^2 k_D^{(d-2)/2}\Gamma\left(\frac{d-2}{2}\right)\Gamma\left(\frac{4-d}{2}\right), \quad 2 < d < 4 \qquad (30)$$

and similar results for some other thermodynamic functions.

$d - 2 > m$. In this somewhat exotic super-Coulomb case $\omega(k)$ will diverge for small k, reinforcing the above results for the Coulomb system, which clearly appear as a boundary case.

6. CONCLUSIONS

It has been shown that Bogoliubov's $1/k^2$ singularity leading to the absence of long-range positional order in the infinite crystal for $d \leqslant 2$ is affected by the range of the potential. For long-range inverse-power repulsive potentials of the form $V(r) = e^2 r^{-m}/m$ the system, suitably stabilized by an appropriately "charged" background, can exhibit no infinite crystalline order for $d \leqslant (m + 2)/2$ when $m \leqslant d$. Consequently, for genuine Coulomb systems ($m = d - 2$) and the electron surface layers ($m = 1$, $d = 2$) long-range order cannot be excluded by this argument.

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