Absence of crystalline order in two dimensions

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It is proven that in two dimensions a system of electrons embedded in a uniform neutralizing positive background and interacting by a potential given by e^2/r cannot exhibit long-range crystalline order at any finite temperature. The proof is conditional to the following prescription for taking the thermodynamic limit: The potential e^2/r is replaced by a screened potential $e^2e^{-\mu r}/r$ and μ is set to zero after the thermodynamic limit is taken.

Grimes and Adams[1] have recently presented evidence for a crystalline transition of electrons trapped on a surface of liquid helium. This quasi-two-dimensional system has been canonically modeled as a system of electrons in two dimensions, interacting by a potential given by e^2/r, and neutralized by a uniform positive background. Here r is the two-dimensional distance between two points. In this context it is important to know if at any finite temperature such a system can display true long-range crystalline order in the thermodynamic limit. In particular, there are attempts[2] to establish contacts between the experiment of Grimes and Adams and the melting theory of Kosterlitz and Thouless. For such a contact to be meaningful it remains to be shown that a conventional long-range order cannot exist in this system. This is precisely the subject of this paper.

We want to emphasize that the crystalline order in this system is not ruled out by the classic work of Mermin.[3] There exist highly plausible general arguments due to Landau[4] and Peierls[5] which indicate that crystalline long-range order cannot exist in two dimensions. These arguments, however, are not completely rigorous. Landau's argument is based on his general theory of second-order phase transitions, which is known to be misleading in the critical region. Peierls's argument is based on the harmonic approximation. The proof in this case consists of showing that the transverse part of the harmonic phonon frequency $\omega_t(\vec{k}) \simeq k^2a^2$ for $k \to 0$.[6] This then implies that the root-mean-square deviation of a particle from its equilibrium position increases indefinitely in the thermodynamic limit. Although these arguments can be challenged as not being rigorous enough, a recent Monte Carlo simulation of this system by Gann, Chakravarty, and Chester[7] indicated, although not conclusively, the validity of the conclusion drawn from them. On the basis of the numerical work it was conjectured (in Ref. 7) that although a rigorous proof does not exist, the general conclusion of Landau and Peierls should be valid.

In this paper we shall prove that a true long-range crystalline order cannot exist in this system in the thermodynamic limit. The proof does not invoke the harmonic approximation. It makes use of Bogoliubov's inequality[8] as discussed by Mermin[3] and is based on the prescription for the thermodynamic limit described below.

Some complications arise because of the long-range nature of the interaction. Although these complications are well known it is important to state them clearly: (a) In order to obtain physically meaningful results the interaction e^2/r is replaced by $e^2/r \exp(-\mu r)$. Since we are interested in the bulk properties of the neutral medium our limiting procedure will be first $N \to \infty$, $A \to \infty$, $N/A \to n =$ constant, and then $\mu \to 0$. Here N is the total number of electrons and A the area. This limiting procedure is a commonly accepted one, appearing in many textbooks.[9] However, we are unable to justify it rigorously. Curiously enough Mermin's proof does not hold for the screened interaction $e^2e^{-\mu r}/r$ either, since the interaction is not sufficiently divergent as $r \to 0$. (b) Another related assumption[10] is that the equilibrium state of the system does not have macroscopic surface charge. Although it is highly implausible that an equilibrium state violating this assumption could have lower free energy, we are not aware of a proof of this.

It will become evident later that even with these assumptions stated above, the original proof of Mermin does not go through because of the plasmons in the long-wavelength limit. Consider, then, N classical electrons enclosed in a box of area A. A uniform neutralizing positive background is assumed to exist.

The interaction energy U is

$$U=\frac{1}{2}\sum_{i\neq j}\frac{e^2}{|\vec{r}_i-\vec{r}_j|}\exp(-\mu|\vec{r}_i-\vec{r}_j|)-\frac{N}{A}\sum_{i=1}^{N}\int_A d\vec{r}\,\frac{e^2}{|\vec{r}-\vec{r}_i|}\exp(-\mu|\vec{r}-\vec{r}_i|)$$
$$+\frac{N^2}{2A^2}\int_A d\vec{r}\int_A d\vec{r}'\,\frac{e^2}{|\vec{r}-\vec{r}'|}\exp(-\mu|\vec{r}-\vec{r}'|)\ . \tag{1}$$

In Eq. (1) the first term is the electron-electron interaction, the second is the interaction between the electrons and the positive background, and the third is the self-energy of the background.

We adopt the same criterion for crystallinity as Mermin; i.e.,

$$\lim_{N\to\infty}\langle\rho_{\vec{k}}\rangle/N=0\ ,$$

$\vec{k}$ not a reciprocal-lattice vector

$$\lim_{N\to\infty}\frac{\langle\rho_{\vec{k}}\rangle}{N}\neq 0\ ,$$

for $\vec{k}$ equal to at least one reciprocal-lattice vector $\vec{G}$, where

$$\rho_{\vec{k}}=\sum_{i=1}^{N}e^{i\vec{k}\cdot\vec{r}_i}\ . \tag{2}$$

Here $\langle f\rangle$ denotes the canonical ensemble average with respect to the interaction energy U and the integrations are over the interior of a box of area A. We now consider the Schwartz inequality

$$\langle|C|^2\rangle\geq\langle|C^*\vec{B}|^2\rangle/\langle|\vec{B}|^2\rangle \tag{3}$$

where

and choose C and $\vec{B}$ to be

$$C=\sum_{i=1}^{N}e^{-i(\vec{k}+\vec{G})\cdot\vec{r}_i}\ , \tag{4}$$

$$\vec{B}=-k_BTe^{\beta U}\sum_{i=1}^{N}\left(\vec{\nabla}_i-\frac{\vec{k}(\vec{k}\cdot\vec{\nabla}_i)}{k^2}\right)\left(f(\vec{r}_i)e^{i\vec{k}\cdot\vec{r}_i}e^{-\beta U}\right)\ . \tag{5}$$

The function $f(\vec{r}_i)$ is chosen as follows.[10] Consider Ω_s to be a set of points within a distance $aL^{1/2}$ of the walls of the box, where a is independent of L. f is a continuous and differentiable function which vanishes on the surface and satisfies the following conditions: (1) $0\leq f\leq 1$ in Ω_s, (2) $f=1$ everywhere else, and (3) $|\vec{\nabla}f|<bL^{-1/2}$ in Ω_s, for some b independent of L. It will become clearer later on that we have gone to great pains in introducing the functions $f(\vec{r}_i)$.[11] The intention has been to make the surface terms arising from the integrations by parts vanish. The complications arising from $f(\vec{r}_i)$ could have been entirely avoided with a slightly careless use of the periodic boundary condition. The choice of $\vec{B}$ [Eq. (5)] is one of the key points of our proof. It serves to project out the longitudinal part of the force and suppresses the plasmons in the denominator of the right-hand side of the inequality [Eq. (3)]. With these choices of C and $\vec{B}$ [Eqs. (4) and (5)] it is straightforward to derive the inequality

$$\frac{1}{N}\langle|\rho_{\vec{k}+\vec{G}}|^2\rangle\geq k_BT\frac{\Delta(\vec{G})}{\Lambda(\vec{k})}\ , \tag{6}$$

where

$$\Delta(\vec{G})=[G^2-(\hat{k}\cdot\vec{G})^2]\left|\frac{1}{N}\int d\vec{r}_1P(\vec{r}_1)f(\vec{r}_1)e^{i\vec{G}\cdot\vec{r}_1}\right|^2\ , \tag{7}$$

$$\Lambda(\vec{k})=\frac{k_BT}{N}\int d\vec{r}_1P(\vec{r}_1)\{[\vec{\nabla}_1f(\vec{r}_1)]^2-[\hat{k}\cdot\vec{\nabla}_1f(\vec{r}_1)]^2\}-\frac{n}{N}\int d\vec{r}_1f^2(\vec{r}_1)P(\vec{r}_1)[\nabla_1^2-(\hat{k}\cdot\vec{\nabla}_1)^2]\int d\vec{r}_2\frac{e^{-\mu r_{12}}}{r_{12}}$$
$$+\frac{1}{2N}\int d\vec{r}_1\int d\vec{r}_2|f(\vec{r}_1)e^{i\vec{k}\cdot\vec{r}_1}-f(\vec{r}_2)e^{i\vec{k}\cdot\vec{r}_2}|^2\,P(\vec{r}_1,\vec{r}_2)[\nabla_1^2-(\hat{k}\cdot\vec{\nabla}_1)^2]\frac{e^{-\mu r_{12}}}{r_{12}}\ . \tag{8}$$

The distribution functions are defined to be

$$P(\vec{r}_1)=N\frac{\int e^{-\beta U}d\vec{r}_2\cdots d\vec{r}_N}{\int e^{-\beta U}d\vec{r}_1\cdots d\vec{r}_N}\ , \tag{9}$$

$$P(\vec{r}_1,\vec{r}_2)=N(N-1)\frac{\int e^{-\beta U}d\vec{r}_3\cdots d\vec{r}_N}{\int e^{-\beta U}d\vec{r}_1\cdots d\vec{r}_N}\ . \tag{10}$$

The inequality (6) is now multiplied by a positive Gaussian $g(\vec{k}+\vec{G})$ centered at $\vec{k}+\vec{G}=0$, and summed over $\vec{k}$ to obtain

$$\frac{1}{A}\sum_{\vec{k}} g(\vec{k}+\vec{G})\frac{\langle|\rho_{\vec{k}+\vec{G}}|^2\rangle}{N} \geq \frac{k_B T g(\frac{1}{2}G)}{A}\sum_{k<G/2}\frac{\Delta(\vec{G})}{\Lambda(\vec{k})} \quad . \tag{11}$$

The strategy now will be to proceed to the thermodynamic limit, show that $\Lambda(\vec{k}) \sim \lambda k^2$ for small k, and then take the limit $\mu \to 0$. The proof will then be completed by showing that the left-hand side of the inequality is bounded. Proceeding to the thermodynamic limit then allows us to write

$$\int_0^\infty kdk \int_0^{2\pi} d\theta_k g(\vec{k}+\vec{G})\frac{\langle|\rho_{\vec{k}+\vec{G}}|^2\rangle}{N} \geq k_B T g(\tfrac{1}{2}G)\int_{k<G/2} kdk \int_0^{2\pi} d\theta_k \frac{\Delta(\vec{G})}{\Lambda(\vec{k})} \quad . \tag{12}$$

The inequality (12) can be strengthened further with the help of methods similar to those used by Fernandez.[12] We obtain

$$\int d\vec{k} g(\vec{k}+\vec{G})\frac{\langle|\rho_{\vec{k}+\vec{G}}|^2\rangle}{N} \geq \tfrac{1}{8}\pi k_B T G^2 g(\tfrac{1}{2}G)\left|\frac{1}{N}\int d\vec{r}_1 P(\vec{r}_1) f(\vec{r}_1) e^{i\vec{G}\cdot\vec{r}_1}\right|^2 \int_{k<G/2} kdk\left\{4/\pi\int_0^{2\pi} d\theta_k \Lambda(\vec{k})\right\}^{-1} \tag{13}$$

Now

$$\begin{aligned}
\lim_{k\to 0}\frac{4}{\pi}\int_0^{2\pi} d\theta_k \Lambda(\vec{k}) = {} & \frac{4k_B T}{N}\int d\vec{r}_1 P(\vec{r}_1)[\vec{\nabla}_1 f(\vec{r}_1)]^2 - \frac{4n}{N}\int d\vec{r}_1 f^2(\vec{r}_1) P(\vec{r}_1)\nabla_1^2 \int d\vec{r}_2 \frac{e^{-\mu r_{12}}}{r_{12}} \\
& + \frac{2}{N}\int d\vec{r}_1 \int d\vec{r}_2 \frac{e^{-\mu r_{12}}}{r_{12}^3}(\mu^2 r_{12}^2 + \mu r_{12} + 1)[f(\vec{r}_1) - f(\vec{r}_2)]^2 P(\vec{r}_1, \vec{r}_2) \\
& + \frac{k^2}{2N}\int d\vec{r}_1 \int d\vec{r}_2 \frac{e^{-\mu r_{12}}}{r_{12}}(1 + \mu r_{12} - \mu^2 r_{12}^2)[1 + f(\vec{r}_2)][1 - f(\vec{r}_1)] P(\vec{r}_1, \vec{r}_2) \\
& + \frac{k^2}{2N}\int d\vec{r}_1 \int d\vec{r}_2 \frac{e^{-\mu r_{12}}}{r_{12}}(\mu^2 r_{12}^2 - \mu r_{12} - 1)\left[P(\vec{r}_1, \vec{r}_2) - \frac{N}{A}P(\vec{r}_1) - \frac{N}{A}P(\vec{r}_2) + \frac{N^2}{A^2}\right] \\
& + \frac{k^2}{2N}\int d\vec{r}_1 \int d\vec{r}_2 \left[\frac{N}{A}P(\vec{r}_1) + \frac{N}{A}P(\vec{r}_2) - \frac{N^2}{A^2}\right]\frac{e^{-\mu r_{12}}}{r_{12}}(\mu^2 r_{12}^2 - \mu r_{12} - 1) \quad .
\end{aligned} \tag{14}$$

The first term on the right-hand side of Eq. (14) vanishes since we are now in the thermodynamic limit. The argument is identical to that used by Mermin.[10] The second term can also be seen to vanish after converting it to a surface integral.

The next two terms also vanish but are trickier. Consider first the third term. Here at least one of the integrations get restricted to Ω_s and let us call that integration $\vec{r}_2$. Now

$$\begin{aligned}
& \frac{2}{N}\int_A d\vec{r}_1 \int_A d\vec{r}_2 \frac{e^{-\mu r_{12}}}{r_{12}^3}(\mu^2 r_{12}^2 + \mu r_{12} + 1)[f(\vec{r}_1) - f(\vec{r}_2)]^2 P(\vec{r}_1, \vec{r}_2) \\
& \qquad \leq \frac{2}{N}\frac{b^2}{L}\int_A d\vec{r}_1 \int_{\Omega_s} d\vec{r}_2 \frac{e^{-\mu r_{12}}}{r_{12}}(\mu^2 r_{12}^2 + \mu r_{12} + 1) P(\vec{r}_1, \vec{r}_2) \sim \left[\frac{N}{A}\right] L^{-3/2} \quad .
\end{aligned} \tag{15}$$

Similarly the fourth term can be estimated to be

$$\begin{aligned}
& \frac{k^2}{2N}\int_A d\vec{r}_1 \int_A d\vec{r}_2 \frac{e^{-\mu r_{12}}}{r_{12}}[1 + f(\vec{r}_2)][1 - f(\vec{r}_1)](1 + \mu r_{12} - \mu^2 r_{12}^2) P(\vec{r}_1, \vec{r}_2) \\
& \qquad \leq \frac{5k^2}{4N}\int_{\Omega_s} d\vec{r}_1 \int d\vec{r}_2 \frac{e^{-\mu r_{12}}}{r_{12}} P(\vec{r}_1, \vec{r}_2) \sim \left[\frac{N}{A}\right] L^{-1/2} \quad .
\end{aligned} \tag{16}$$

In both of the estimates [Eqs. (15) and (16)] we have followed the prescription that the thermodynamic limit is taken before $\mu \to 0$. If this were not the case both of these estimates would have diverged. Also, since we are in the thermodynamic limit the last term can also be seen to be zero by explicit integration. It is easy to verify that if we had made use of the function $\vec{B}$ as chosen by Mermin,[3] the term analogous to the last term in Eq. (14) would have diverged as $\mu \to 0$. A closer look would reveal that the leading term in that case is proportional to k and not k^2, thus substantiating our earlier remarks about the choice of $\vec{B}$. We then have

$$\lim_{k\to 0}\frac{4}{\pi}\int_0^{2\pi} d\theta_k \Lambda(\vec{k}) = \frac{k^2}{2N}\int d\vec{r}_1 \int d\vec{r}_2 \frac{e^{-\mu r_{12}}}{r_{12}}\left[P(\vec{r}_1,\vec{r}_2) - \frac{N}{A}P(\vec{r}_1) - \frac{N}{A}P(\vec{r}_2) + \frac{N^2}{A^2}\right](\mu^2 r_{12}^2 - \mu r_{12} - 1) \quad . \quad (17)$$

The limit $\mu \to 0$ now allows us to identify the right-hand side of Eq. (17) as proportional to the internal energy per electron of a system of electrons embedded in a neutralizing uniform positive background and interacting by a potential e^2/r in two dimensions. Hence,

$$\lim_{\mu\to 0}\lim_{k\to 0}\frac{4}{\pi}\int_0^{2\pi} d\theta_k \Lambda(\vec{k}) = -k^2(E_{\text{int}}/N) \quad . \quad (18)$$

For this system a rigorous lower bound has recently been obtained by Totsuji[13]

$$\frac{E_{\text{int}}}{N} \geq -\frac{1.1284e^2}{a} \quad , \quad (19)$$

where a is the average interparticle distance. Thus we have

$$\lim_{\mu\to 0}\lim_{k\to 0}\frac{4}{\pi}\int_0^{2\pi} d\theta_k \Lambda(\vec{k}) \leq k^2\left[\frac{1.1284e^2}{a}\right] \quad , \quad (20)$$

which can be used in the denominator to strengthen the inequality further. One additional piece of information is required. This is

$$\frac{\langle \rho_{\vec{G}} \rangle}{N} + \frac{1}{N}\int_{\Omega_s} d\vec{r}_1 P(\vec{r}_1) \geq \frac{1}{N}\int d\vec{r}_1 P(\vec{r}_1) f(\vec{r}_1) e^{i\vec{G}\cdot\vec{r}_1} \geq \frac{\langle \rho_{\vec{G}} \rangle}{N} - \frac{1}{N}\int_{\Omega_s} d\vec{r}_1 P(\vec{r}_1) \quad . \quad (21)$$

Therefore, $\frac{1}{N}\int d\vec{r}_1 P(\vec{r}_1) f(\vec{r}_1) e^{i\vec{G}\cdot\vec{r}_1}$ can be replaced in the thermodynamic limit by $\langle \rho_{\vec{G}} \rangle / N$ since by assumption the equilibrium state does not have macroscopic surface charge.[10]

To complete the proof it needs to be shown that the left-hand side of the inequality [Eq. (13)] is finite. This will then imply that a finite quantity is greater than or equal to infinity, and hence, the inescapable conclusion that $\langle \rho_{\vec{G}} \rangle / N$ vanishes at any finite temperature. One way to proceed is to follow Mermin.[3] The important ingredient is of course the free energy of the system considered in this paper. This, thanks to Totsuji,[13] can be shown to have both an upper and a lower bound. The other alternative is to follow Sorokina[14] word for word and arrive at the conclusion that the left-hand side of the inequality (13) is bounded.

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