

# Absence of Strict Crystalline Order in a Two-Dimensional Electron System

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It is shown that no long-range crystalline order is possible in a two-dimensional electron system, in spite of the long-range nature of the forces. The order is destroyed by the transverse phonons, as can be seen either by Peierls' argument or more rigorously by a modification of Mermin's argument.

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**KEY WORDS:** Long-range order; two-dimensional electron system; transverse phonons.

## 1. INTRODUCTION

Recently, two-dimensional electron systems have been widely studied, both theoretically and experimentally. A transition to a classical Wigner solid phase has been observed,<sup>(1)</sup> in agreement with a numerical simulation<sup>(2)</sup> and theoretical approaches.<sup>(3,4)</sup> The question arises of whether this two-dimensional "solid" phase has a genuine crystalline long-range order. Arguments according to which thermal fluctuations do not allow crystalline order in one and two dimensions have been given by Peierls<sup>(5)</sup> and Landau,<sup>(6)</sup> and later Mermin<sup>(7,8)</sup> gave a rigorous proof of the absence of order. Mermin's proof, however, is restricted to short-range potentials and therefore cannot be used, as it stands, for electrons. Very recently, Baus<sup>(9)</sup> extended Mermin's proof to some longer range potentials (at the expense of a lesser degree of rigor); however, for the case of a two-dimensional

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electron system, Baus' approach is inconclusive and crystalline order cannot be excluded by his argument.

In the present paper, we recall that the simple Peierls argument does apply to a two-dimensional system of electrons. Although the longitudinal phonons here become harmless "plasmons," the order is destroyed by the *transverse* soundlike phonons. Mermin's argument, as extended by Baus, is inconclusive because in that proof the roles of the transverse and longitudinal modes are not disentangled from one another. In the following, we show that, through a suitable modification of the argument, it is possible to separate the role of the transverse modes, which do prevent long-range order. Therefore, both the Peierls argument and a modified Mermin argument lead to the same conclusion: the absence of positional long-range order for an infinite two-dimensional electron system. This conclusion, of course, does not forbid the existence of a solid phase characterized by other properties, such as directional long-range order,<sup>(7)</sup> and approximate positional long-range order in the practical case of a finite system.<sup>(2,7)</sup>

The model which is considered here is a two-dimensional system of electrons of charge  $e$  and mass  $m$ . They interact with one another through the Coulomb potential  $e^2/r$ . Furthermore, they are embedded in a uniform background of opposite charge, which ensures overall neutrality.

## 2. THE PEIERLS ARGUMENT

This well-known argument starts from the assumption that a crystal exists. The vibrations of this crystal are studied within the harmonic approximation, and the mean square displacement of a particle is computed. When this mean square displacement is found to become infinite in the limiting case of an infinite system (the thermodynamic limit), it is concluded that the initial assumption that crystalline order exists is false.

This argument has been applied to the two-dimensional electron system by Chaplik<sup>(10)</sup> and Crandall.<sup>(11)</sup> For a system of  $N$  electrons, the mean square displacement of any of them is found to be

$$\langle u^2 \rangle = \frac{\hbar}{2Nm} \sum_{\mathbf{k}, s} \frac{\coth[\beta \hbar \omega_s(\mathbf{k})/2]}{\omega_s(\mathbf{k})} \quad (1)$$

where  $\beta = 1/k_B T$  ( $k_B$  is Boltzmann's constant and  $T$  is the temperature) and where  $\omega_s(\mathbf{k})$  is the frequency of a phonon of wave number  $\mathbf{k}$  and polarization  $s$ . Here, in the long-wavelength limit, the frequency  $\omega_l(\mathbf{k})$  of a longitudinal phonon behaves like  $k^{1/2}$ , while the frequency  $\omega_t(\mathbf{k})$  of a transverse phonon behaves like  $k$ . In the thermodynamic limit, the sum on  $\mathbf{k}$  in (1) becomes an integral on the two-dimensional  $\mathbf{k}$  space. While the

contribution from  $\omega_l(\mathbf{k})$  remains finite, the linear  $k$  behavior of  $\omega_l(\mathbf{k})$  results in a logarithmic divergence of  $\langle u^2 \rangle$  at any finite temperature.

Therefore, the long-range nature of the  $1/r$  potential, although it is responsible for the hardening of the longitudinal phonon dispersion law into  $k^{1/2}$ , does not invalidate the Peierls argument here, because transverse phonons with a normal linear dispersion law are still there. It should be noted that a different story may occur in *one*-dimensional models with long-range forces. For instance, the one-component plasma in one dimension, in which the particles interact through a potential proportional to their distance, has only longitudinal modes with a finite plasma frequency;  $\langle u^2 \rangle$  is then found to be finite in the harmonic approximation, in agreement with the existence of a rigorous proof,<sup>(12)</sup> in the classical case, that this model exhibits crystalline order.

### 3. A MODIFIED MERMIN ARGUMENT

The Peierls argument is based on a harmonic approximation. Although a completely rigorous approach is beyond the scope of the present paper, we now give an argument, inspired by Mermin's approach, where the harmonic approximation is not used. We consider only the classical case. We find it convenient to use periodic boundary conditions. If the electrons are supposed to form a crystal with a Bravais lattice generated by  $\mathbf{a}_1, \mathbf{a}_2$ , we take a box of sides  $N_1\mathbf{a}_1$  and  $N_2\mathbf{a}_2$  and wrap it on a torus in the usual way. Since we want any crystalline order to be signaled by a periodic one-body density, overall translations of the crystal must be prevented. This is achieved by introducing a periodic external potential  $V_e$  with periods  $\mathbf{a}_1$  and  $\mathbf{a}_2$ . The thermodynamic limit is taken *first*, and *afterwards* the external potential is brought to zero. The criterion for crystallinity is that the one-body density be a (nonconstant) periodic function with periods  $\mathbf{a}_1$  and  $\mathbf{a}_2$  after this double limit has been taken. We want to show that this is impossible.

We start from the Schwartz inequality

$$\langle |A|^2 \rangle \geq |\langle A^* B \rangle|^2 / \langle |B|^2 \rangle \quad (2)$$

where  $A$  and  $B$  are two dynamical variables and where  $\langle \dots \rangle$  denotes the canonical equilibrium average. We define one-body functions of the position  $\mathbf{r}$  by

$$\psi(\mathbf{r}) = \exp[i(\mathbf{k} + \mathbf{K}) \cdot \mathbf{r}] \quad (3)$$

and

$$\varphi(\mathbf{r}) = \exp(i\mathbf{k} \cdot \mathbf{r}) \quad (4)$$

where  $\mathbf{K}$  is some nonzero reciprocal-lattice vector (i.e.,  $\mathbf{K} \cdot \mathbf{a}_1$  and  $\mathbf{K} \cdot \mathbf{a}_2$  are multiples of  $2\pi$ ) and  $\mathbf{k}$  is of the form

$$\mathbf{k} = \frac{n_1}{N_1} \mathbf{b}_1 + \frac{n_2}{N_2} \mathbf{b}_2, \quad \mathbf{b}_\alpha \cdot \mathbf{a}_\gamma = 2\pi\delta_{\alpha\gamma} \quad (5)$$

so that  $\psi(\mathbf{r})$  and  $\varphi(\mathbf{r})$  obey periodic boundary conditions on the walls of the box. We now choose

$$A = \sum_{j=1}^N \psi(\mathbf{r}_j) \quad (6)$$

$$B = e^{\beta U} \sum_{j=1}^N \nabla_{y_j} [\varphi(\mathbf{r}_j) e^{-\beta U}] \quad (7)$$

$\mathbf{r}_j$  is the position of particle  $j$ ,  $U(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N)$  is the potential energy of the  $N$  particles, and  $\nabla_{y_j}$  denotes the derivative with respect to  $\mathbf{r}_j$  along a given direction marked by a unit vector  $\mathbf{e}_i$ . Using (6) and (7) in (2), and performing integrations by parts in which the surface terms vanish due to the periodic boundary conditions, we find

$$\langle |\sum_j \psi(\mathbf{r}_j)|^2 \rangle \geq \frac{|\langle \sum_j \varphi(\mathbf{r}_j) \nabla_{y_j} \psi^*(\mathbf{r}_j) \rangle|^2}{\langle \sum_j |\nabla_{y_j} \varphi(\mathbf{r}_j)|^2 + \beta \sum_{i,j} \varphi(\mathbf{r}_i) \varphi^*(\mathbf{r}_j) \nabla_{ii} \nabla_{jj} U \rangle} \quad (8)$$

Using (3) and (4) in (8), we obtain

$$S(\mathbf{K} + \mathbf{k}) \geq \frac{[(\mathbf{k} + \mathbf{K}) \cdot \mathbf{e}_i]^2 |\rho_{\mathbf{K}}|^2}{(\mathbf{k} \cdot \mathbf{e}_i)^2 + \beta D_{ii}(\mathbf{k})} \quad (9)$$

where

$$S(\mathbf{k}) = \frac{1}{N} \langle |\sum_j \exp(i\mathbf{k} \cdot \mathbf{r}_j)|^2 \rangle \quad (10)$$

is the structure factor,

$$\rho_{\mathbf{K}} = \frac{1}{N} \langle \sum_j \exp(i\mathbf{K} \cdot \mathbf{r}_j) \rangle \quad (11)$$

is the  $\mathbf{K}$  Fourier component of the one-body density (crystalline long-range order would be signaled by a nonzero value of  $\rho_{\mathbf{K}}$  for at least one nonzero reciprocal-lattice vector  $\mathbf{K}$ ), and

$$D_{ii}(\mathbf{k}) = \frac{1}{N} \langle \sum_{i,j} \{ \exp[i\mathbf{k} \cdot (\mathbf{r}_i - \mathbf{r}_j)] \} \nabla_{ii} \nabla_{jj} U \rangle \quad (12)$$

In (12), there are contributions from the particle-particle interactions, the particle-background interactions, and the external localizing potential. The particle-background contribution is of order  $\rho e^2/L$ , where  $\rho$  is the number density of the electrons and  $L$  a length which defines the size of the box;

therefore this contribution vanishes in the thermodynamic limit. In this limit,

$$D_{ii}(\mathbf{k}) = E + D_{ii}^{(0)}(\mathbf{k}) \quad (13)$$

where  $E$  is the contribution from the external potential and  $D_{ii}^{(0)}(\mathbf{k})$  the contribution from the particle-particle interactions  $e^2/r_{ij}$ . These contributions are

$$E = \lim_{N \rightarrow +\infty} \left[ \frac{1}{N} \int \rho(\mathbf{r}) \nabla_i^2 V_e(\mathbf{r}) d\mathbf{r} \right] \quad (14)$$

where  $\rho(\mathbf{r})$  is the one-body density, and

$$D_{ii}^{(0)}(\mathbf{k}) = \rho \int g(\mathbf{r}) [1 - \cos \mathbf{k} \cdot \mathbf{r}] \nabla_i^2 \frac{e^2}{r} d\mathbf{r} \quad (15)$$

where  $g(\mathbf{r})$  is the pair distribution function, which is defined as usual by

$$\rho g(\mathbf{r}) = \lim_{N \rightarrow +\infty} \left[ \frac{1}{N} \left\langle \sum_{i \neq j} \delta(\mathbf{r}_i - \mathbf{r}_j - \mathbf{r}) \right\rangle \right] \quad (16)$$

In a harmonic approximation,  $m^{-1}D_{ii}$  should be an element of the dynamical matrix. Here, no harmonic approximation is made, but, guided by the idea that the transverse modes are the relevant<sup>3</sup> ones, we choose the direction  $\mathbf{e}_t$  as transverse to  $\mathbf{k}$ :

$$\mathbf{k} \cdot \mathbf{e}_t = 0 \quad (17)$$

With this choice, one obtains, by an integration by parts,

$$\int (1 - \cos \mathbf{k} \cdot \mathbf{r}) \nabla_i^2 \frac{e^2}{r} d\mathbf{r} = 0 \quad (18)$$

Therefore, writing the pair distribution function  $g(\mathbf{r})$  as

$$g(\mathbf{r}) = 1 + h(\mathbf{r}) \quad (19)$$

one can recast (15) as

$$D_{ii}^{(0)}(\mathbf{k}) = \rho \int h(\mathbf{r}) [1 - \cos \mathbf{k} \cdot \mathbf{r}] \nabla_i^2 \frac{e^2}{r} d\mathbf{r} \quad (20)$$

In the small- $k$  limit, by expanding<sup>4</sup>  $\cos \mathbf{k} \cdot \mathbf{r}$ , we find  $D_{ii}^{(0)}(\mathbf{k})$  to behave like  $Ck^2$ , with

$$C = \frac{1}{2} \rho \int h(\mathbf{r}) (e^2/r) \cos^2 \theta (3 \sin^2 \theta - 1) d\mathbf{r} \quad (21)$$

<sup>3</sup> In the original Mermin argument, the gradient vector is used rather than its component  $\nabla_i$ , and the trace of the matrix  $D$  appears rather than its single element  $D_{ii}$ . In the case considered here, it is essential to have  $D_{ii}$  alone in the denominator of (9).

<sup>4</sup> We assume that an expansion of  $\cos \mathbf{k} \cdot \mathbf{r}$  is legitimate, at least for giving correctly the lowest order term. A rigorous proof would require more knowledge than we have about  $h(\mathbf{r})$ .

( $\theta$  is the angle between  $\mathbf{k}$  and  $\mathbf{r}$ ). We assume the electron lattice to be a triangular one;<sup>(13)</sup> this makes  $h(\mathbf{r})$  invariant when  $\theta$  is increased by  $\pi/3$ . Thus, if we use in (21) the identity

$$\cos^2\theta(3\sin^2\theta - 1) = -\frac{1}{8} - \frac{1}{2}\cos 2\theta - \frac{3}{8}\cos 4\theta \quad (22)$$

only the first term on the right-hand side of (22) contributes to (21), which becomes

$$C = -\frac{1}{16}\rho \int h(\mathbf{r})(e^2/r) d\mathbf{r} \quad (23)$$

Therefore,  $-8C$  is the potential energy per particle (not including the interaction energy with the external potential). The existence of the thermodynamic limit for the present system has not been rigorously proved. Here we assume that the potential energy per particle has a finite limit, or at least that  $C$  has finite lower and upper bounds when systems of increasing size are considered. Using (13) and (17) in (9), we obtain

$$S(\mathbf{K} + \mathbf{k}) \geq (\mathbf{K} \cdot \mathbf{e}_i)^2 |\rho_{\mathbf{k}}|^2 / \beta [E + D_{ii}^{(0)}(\mathbf{k})] \quad (24)$$

The denominator on the right-hand side of (24) is nonnegative, since it comes from the  $\langle |B|^2 \rangle$  of (2). Therefore, we can write<sup>5</sup>

$$S(\mathbf{K} + \mathbf{k}) \geq (\mathbf{K} \cdot \mathbf{e}_i)^2 |\rho_{\mathbf{k}}|^2 / \beta [ |E| + |D_{ii}^{(0)}(\mathbf{k})| ] \quad (25)$$

where  $D_{ii}^{(0)}(\mathbf{k})$  behaves like  $|C|k^2$  for small  $k$  values.

The final step follows the line of the original argument. Both sides of (25) are multiplied by a positive Gaussian function  $f(|\mathbf{K} + \mathbf{k}|)$ , divided by the surface  $S$  of the box, and summed on all  $\mathbf{k}$  values of the form (5). In terms of the Gaussian Fourier transform

$$F(r) = \frac{1}{(2\pi)^2} \int d\mathbf{q} [\exp(i\mathbf{q} \cdot \mathbf{r})] f(q) \quad (26)$$

we obtain in the thermodynamic limit

$$F(0) + \rho \int g(\mathbf{r}) F(r) d\mathbf{r} \geq \frac{|\rho_{\mathbf{k}}|^2}{(2\pi)^2 \beta} \int \frac{d\mathbf{k} (\mathbf{K} \cdot \mathbf{e}_i)^2 f(|\mathbf{K} + \mathbf{k}|)}{|E| + |D_{ii}^{(0)}(\mathbf{k})|} \quad (27)$$

From what we know about the general shape of the pair distribution function  $g(\mathbf{r})$ , it is rather obvious that the left-hand side of (25) is finite. [We could also argue that  $\frac{1}{2}\rho \int g(\mathbf{r}) F(r) d\mathbf{r}$  is bounded by the difference in free energies per particle for the pair potentials  $e^2/r$  and  $(e^2/r) - F(r)$ , but we must then assume that the free energy per particle exists in the thermodynamic limit for the latter case as well.] We can decrease the

<sup>5</sup> This step is perhaps unnecessary, because it is likely that  $E$  and  $D_{ii}^{(0)}$  are both positive.

right-hand side of (27) by restricting the integration domain to a small circle around the origin, where  $|D_n^{(0)}(\mathbf{k})|$  behaves like  $|C|k^2$ . The integral is then of the order of  $|\ln|E||$ . Therefore, when the external potential  $V_e$  is withdrawn and  $E$  goes to zero, the integral diverges, and the inequality (27) can be satisfied only if  $\rho_K$  goes to zero. No spontaneous crystalline order survives.

#### 4. CONCLUSION

Mainly due to the absence of information about the existence of the thermodynamic limit, we have not been able to reach complete mathematical rigor. We believe, however, that the main point in our argument is essentially correct: the transverse modes do not allow a positional long-range order in an infinite two-dimensional electron system, in spite of the long-range nature of the forces.

With the same limitations in rigor, the above conclusion can be easily generalized to a two-dimensional system of particles interacting through a repulsive potential  $1/r^n$ ,  $n$  being any positive real number (a background is needed for  $n \leq 2$ ; the case  $n > 2$  is covered by the original Mermin argument).<sup>6</sup>

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<sup>6</sup> Another paper on the same subject as the present one has been independently published (see Ref. 14).