Crystalline Stability and Order in One and Two Dimensions

JULIO F. FERNÁNDEZ
Physics Department, University of South Carolina, Columbia, South Carolina 29208
(Received 30 June 1969)

It is shown that no quantum system of particles, regardless of their interactions, can form a stable solid in one or two dimensions, if both the temperature and the compressibility are different from zero. By stable solid we mean that $\langle u^2\rangle$ be finite. It is also proven that a neutral system of electrons and nuclei cannot exhibit crystalline order, of the usual kind, in one or two dimensions. This last proof is based on Bogolyubov's inequality.

I. INTRODUCTION

It was proven long ago, by Peierls,[1] that a two-dimensional system of particles connected by linear springs will not form a solid. Landau[2] obtained the same result using phenomenological arguments. More recently, Mermin[3] has been able to prove from first principles that a system of particles interacting through a two-body potential $\phi(r)$ satisfying the conditions

$$\phi(r)\rightarrow 1/r^{2+\epsilon} \quad \text{as} \quad r\rightarrow\infty\,,$$
$$\phi(r)\rightarrow |A|/r^{2+\epsilon} \quad \text{as} \quad r\rightarrow 0$$

will not form a solid.

Two outstanding examples of systems covered by the proofs from first principles are the system of hard cores, and the system of particles interacting through the Coulomb potential. The computer experiments of Alder and Wainright[4] performed on a two-dimensional system of hard cores indicate a change of phase, from a fluid to a crystalline state.

The case of the Coulomb potential is of great interest because it is the source of all pertinent nonrelativistic forces in macroscopic physics. It has been studied in the realm of models, and it has been predicted that a system of nuclei and electrons can undergo a liquid-solid phase transition in one dimension.[5]

We shall discuss now three different criteria that may be used to diagnose the existence of the solid phase before we describe the contents of this paper. These well-known criteria are:

(a) The stability condition is

$$\langle u_i^2\rangle<\infty\,, \tag{1}$$

where $\mathbf{u}_i$ is the displacement of the ith particle from its equilibrium position, and the brackets denote thermal averages. To speak about $\langle u_i^2\rangle$ requires distinguishability of particles. One of the objections that one can raise against this condition is that even in a system which one knows to be in a solid phase, $\langle u_i^2\rangle$ may be infinite, meaning only that particles diffuse. But one can have a solid with diffusion. One way to avoid this criticism, simultaneously making the particles distinguishable, is to join the particles in the system by extremely weak springs, which would now act in addition to the previously existing intermolecular forces. If one still cannot satisfy Eq. (1), one does not have a stable solid. This criterion was used by Peierls[1] and Landau.[2]

(b) A less stringent view of crystalline order is one in which $\rho(\mathbf{r})$, the particle density as a function of position, has the periodicity of some lattice. Then if $\mathbf{G}$ is a reciprocal-lattice vector,

$$\lim_{N\rightarrow\infty}\frac{\langle\rho_{\mathbf{G}}\rangle}{N}\neq 0\,, \tag{2}$$

where N is the total number of particles and

$$\rho_{\mathbf{G}}=\int\rho(\mathbf{r})e^{-i\mathbf{G}\cdot\mathbf{r}}d\mathbf{r}$$

is associated with crystalline order.

The criterion given here was used by Landau[2] and Mermin.[3] It is necessary for the existence of Bragg peaks in x-ray[6] or neutron scattering[7] by the system under consideration.

In Appendix A, the connection between the criteria (a) and (b) is discussed. It is shown that Eq. (1) implies Eq. (2), although the converse is not true.

(c) We give the last example of a condition that one may impose on a system to be accepted as a solid. That is, that the zero-frequency shear modules of elasticity not be zero. This imposes a condition on the shear-stress–shear-stress time-dependent correlation function.[8]

As Frenkel[9] stressed, the high-frequency shear modulus is not qualitatively different in fluids and solids. In the infinite-frequency limit, the shear and bulk moduli, in fact, obey the Cauchy relation.[10,11] It makes sense,

[1] R. E. Peierls, Helv. Phys. Acta Suppl. **7**, 81 (1936); Ann. Inst. Henri Poincaré **5**, 177 (1935).
[2] L. D. Landau, Phys. Z. Soviet **11**, 26 (1937); *Collected Papers of Landau* (Pergamon Press, Ltd., London, 1967), p. 193.
[3] N. D. Mermin, Phys. Rev. **176**, 250 (1968).
[4] B. J. Alder and R. E. Wainwright, Phys. Rev. **127**, 359 (1962).
[5] G. Carmi, Phys. Rev. **176**, 521 (1968).
[6] C. Kittel, *Introduction to Solid State Physics* (John Wiley & Sons, New York, 1966), 3rd ed., p. 63.
[7] L. Van Hove, Phys. Rev. **95**, 249 (1954).
[8] M. S. Green, J. Chem. Phys. **14**, 180 (1946); H. Mori, Progr. Theoret. Phys. (Kyoto) **28**, 273 (1962); J. M. Luttinger, Phys. Rev. **135**, 1505 (1964).
[9] J. Frenkel, *Kinetic Theory of Liquids* (Dover Publications, Inc., New York, 1955), 1st ed., p. 188.
[10] J. F. Fernández, Phys. Letters **27A**, 263 (1968).
[11] R. Zwanzig and R. D. Mountain, J. Chem. Phys. **43**, 4464 (1965).

though, to associate a nonvanishing zero-frequency shear modulus with a solid, and a vanishing one with a fluid. This creterion goes along with Alder's[12] idea of the solid-fluid transition. We will only use criteria (a) and (b).

In Sec. II we give an argument, from first principles, to show that no system, regardless of the interaction potential, can satisfy Eq. (1) in one or two dimensions if the compressibility is not zero at $T\neq 0$. This argument is valid for a system of hard cores, particles interacting through the Coulomb or any other potential. In Sec. III we prove that a neutral system of nuclei and electrons interacting through the Coulomb potential cannot satisfy Eq. (2) in one or two dimensions at $T\neq 0$. In Sec. IV we discuss the results.

II. INSTABILITY OF SOLIDS IN TWO DIMENSIONS

In this section we treat a quantum system of distinguishable particles. It will be shown that Eq. (1) is not satisfied in two dimensions. The argument given here is not rigourous in one regard, namely the, limit interchange shown in Eq. (6). Our procedure is to assume $\langle u_i^2\rangle<\infty$ and arrive at a contradiction. For simplicity we treat the case of one particle per primitive cell.

The definition of

$$\rho_{\mathbf{k}}=\int\rho(\mathbf{r})e^{-i\mathbf{k}\cdot\mathbf{r}}d\mathbf{r}$$

and

$$\rho(\mathbf{r})=\sum_i\delta(\mathbf{r}-\mathbf{r}_i) \tag{3}$$

allows us to write

$$\langle\rho_{\mathbf{k}}^+\rho_{\mathbf{k}}\rangle=\sum_{ij}\langle e^{-i\mathbf{k}\cdot(\mathbf{X}_i-\mathbf{X}_i)}e^{-i\mathbf{k}\cdot(\mathbf{u}_i-\mathbf{u}_j)}\rangle, \tag{4}$$

where $\mathbf{X}_i$ is the position vector of the ith lattice site, and $\mathbf{u}_i$ is the displacement of the ith particle away from the site.

If we substitute into Eq. (4) the expression

$$e^{-i\mathbf{k}\cdot(\mathbf{u}_i-\mathbf{u}_j)}=\cos[\mathbf{k}\cdot(\mathbf{u}_i-\mathbf{u}_j)]-i\sin[\mathbf{k}\cdot(\mathbf{u}_i\cdot\mathbf{u}_j)]$$

and

$$\cos\alpha=1-\tfrac{1}{2}\alpha^2+(\cos\alpha-1+\tfrac{1}{2}\alpha^2),\qquad \sin\alpha=\alpha+(\sin\alpha-\alpha),$$

we obtain

$$\langle\rho_{\mathbf{k}}^+\rho_{\mathbf{k}}\rangle=N^2\delta_{\mathbf{k},\mathbf{G}}[1-\langle(\mathbf{k}\cdot\mathbf{u})^2\rangle]+\mathrm{N}\langle\mathbf{k}\cdot\mathbf{Q}_k{}^+\mathbf{k}\cdot\mathbf{Q}_k\rangle+f_{\mathbf{k}}, \tag{5}$$

where $\mathbf{G}$ is a reciprocal-lattice vector,

$$\mathbf{Q}_{\mathbf{k}}=N^{-1/2}\sum_i\mathbf{u}_ie^{-i\mathbf{k}\cdot\mathbf{X}_i}$$

and

$$f_{\mathbf{k}}=\sum_{ij}e^{-i\mathbf{k}\cdot(\mathbf{X}_i-\mathbf{X}_j)}\{\langle\cos\mathbf{k}\cdot(\mathbf{u}_i-\mathbf{u}_j)\rangle-1+\tfrac{1}{2}\langle[k\cdot(\mathbf{u}_i-\mathbf{u}_j)]^2\rangle-i(\langle\sin[\mathbf{k}\cdot(\mathbf{u}_i-\mathbf{u}_j)]\rangle-\langle\mathbf{k}\cdot(\mathbf{u}_i-\mathbf{u}_j)\rangle)\}.$$

We now divide Eq. (5) by N, take the limit $N\to\infty$, and finally, let $k\to 0$. Since $\langle u_i^2\rangle<\infty$, by hypothesis, it is reasonable to *assume* that

$$\lim_{k\to 0}\lim_{N\to\infty}\frac{f_{\mathbf{k}}}{N}=0. \tag{6}$$

We are then left with

$$\lim_{k\to 0}\lim_{N\to\infty}\frac{\langle\rho_{\mathbf{k}}^+\rho_{\mathbf{k}}\rangle}{N}=\lim_{k\to 0}\lim_{N\to\infty}\langle\mathbf{k}\cdot\mathbf{Q}_{\mathbf{k}}\mathbf{k}\cdot\mathbf{Q}_{\mathbf{k}}\rangle.$$

Using a well-known relation, the left-hand side of the previous expression may be replaced by

$$(N/\Omega)\chi_TK_BT,$$

where Ω is the volume of the system, χ_T is the isothermal compressibility, K_B is Boltzmann's constant, and T is the temperature. With Eq. (6) we obtain

$$\lim_{k\to 0}\lim_{N\to\infty}\langle\mathbf{k}\cdot\mathbf{Q}_{\mathbf{k}}{}^+\mathbf{k}\cdot\mathbf{Q}_{\mathbf{k}}\rangle=\frac{N}{\Omega}\chi_TK_BT. \tag{7}$$

From the definition of $Q_{\mathbf{k}}$ it follows that

$$\langle\mathbf{u}_i^2\rangle=\frac{1}{N}\sum_k\langle\mathbf{Q}_{\mathbf{k}}{}^+\mathbf{Q}_{\mathbf{k}}\rangle \tag{8}$$

In the thermodynamic limit $\Omega\to\infty$, $N\to\infty$, $N/\Omega=\text{const}$, Eq. (7) and (8) imply that

$$\langle u_i^2\rangle\geq\frac{1}{(2\pi)^3}\int\frac{\chi_TK_BT}{k^2}dk. \tag{9}$$

This equation implies that if $T\neq 0$ and $\chi_T\neq 0$, then $\langle u_i^2\rangle\geq\infty$. It contradicts our hypothesis. $\chi_T=0$ implies that the particles are jammed against each other, so it is geometrically impossible for $\langle u_i^2\rangle$ to go to infinity. At $T=0$ the entropy plays no role, so again it is not surprising to find $\langle u_i^2\rangle$ not infinite. Equations (7) and (8) also imply, of course, that

$$\lim_{|i-j|\to\infty}\langle(\mathbf{u}_i-\mathbf{u}_j)^2\rangle=\infty,\quad\text{if}\quad T\neq 0,\chi\neq 0.$$

If each particle is not "permanently bound" to its nearest neighbor, $\langle u^2\rangle=\infty$ may simply imply diffusion. If, however, in addition to the given intermolecular potential, one connects the nearest-neighbor particles by extremely weak springs, and $\langle u^2\rangle=\infty$ persists, then it must imply only infinite fluctuations in the absence of

[12] B. J. Alder, W. R. Gardner, J. K. Hoffer, N. E. Phillips, and D. A. Young, Phys. Rev. Letters 21, 732 (1968).

diffusion. The Hamiltonian of the system plays no role at all in the derivation of Eq. (9). It follows, then, that this equation is still valid whether we join particles by springs or not. So $\langle u^2\rangle=\infty$ persists, after we join particles by springs to prevent diffusion. For a discussion of the relation between $\langle u^2\rangle=\infty$ and Eq. (2) see Appendix A.

We point out that the results of this section are independent of the interaction potential. It is also worth noticing that the results are useless for a system of charged particles in an oppositely charged static background, since the compressibility is zero in this case.

III. NUCLEI AND ELECTRONS

Here we treat a system of electrons and nuclei interacting through the Coulomb potential in one and two dimensions. The system is treated quantum mechanically. (Classically, such systems are unstable and collapse.) It is proven in this section that Eq. (2), the condition for crystalline order, it not satisfied by such system.[13] The proof fails for a system of charged particles in *static* charge background. The proof given here is based on the Bogolyubov[14,15] inequality.

The system is placed in a "soft box," i.e., in the one-dimensional case every particle is subjected to the external potential

$$H_w=fN^{1/4}\sum_i\left[-x_i\theta(-x_i)+(x_i-L_x)\theta(x_i-L_x)\right],$$

where $\theta(x)$ is the step function, x is the particle position, f is a finite quantity, and N is the number of nuclei. In the thermodynamic limit, $N\rightarrow\infty$, $\Omega\rightarrow\infty$, N/Ω finite, we get impenetrable walls. (The external potential in the two-dimensional case is the obvious generalization of this.)

To start with, we invoke Bogolyubov's inequality

$$\tfrac{1}{2}\langle[A,A^+]_+\rangle\langle[[C,H],C^+]\rangle\geq K_BT|\langle[C,A]\rangle|^2 \quad (10)$$

and choose

$$C=J_{\mathbf{k}}=\sum_i(\mathbf{k}\cdot\mathbf{p}_i+\tfrac{1}{2}k^2)e^{-i\mathbf{k}\cdot\mathbf{r}_i}+\sum_\nu(\mathbf{k}\cdot\mathbf{P}_\nu+\tfrac{1}{2}k^2)e^{-i\mathbf{k}\cdot\mathbf{R}_\nu} \quad (11)$$

and

$$A=\rho_{\mathbf{G}-\mathbf{k}}{}^N=\sum_\nu e^{-i(\mathbf{G}-\mathbf{k})\cdot\mathbf{R}_\nu}. \quad (12)$$

Here the brackets denote thermal averages; $\mathbf{p}_i$ and $\mathbf{r}_i$ are the momentum and position, respectively, of the ith electron; $\mathbf{P}_\nu$ and $\mathbf{R}_\nu$ are the momentum and position of the νth nucleus. To avoid confusion, Latin indices are used for electrons and Greek ones for nuclei. C is the $\mathbf{k}$th Fourier component of the mass current density of the electrons plus nuclei. A is the $\mathbf{G}-\mathbf{k}$ Fourier component of the nuclear number density. (In Sec. IV the physical idea motivating the choices of C and A is discussed.)

Let the wave vector $\mathbf{k}$ take the values $k_x=2\pi n_x/L_x$, $k_y=2\pi n_y/L_y$, where n_x and n_y are integers. The functions $e^{i\mathbf{k}\cdot\mathbf{x}}$ do not make up a complete set for the soft box, although they do form a complete set in the restricted region where the external field is zero. However, in the thermodynamic limit, the set $e^{i\mathbf{k}\cdot\mathbf{x}}$ with out choice of $\mathbf{k}$'s, goes into a complete set.

The various terms in Bogolyubov's inequality must now be computed. The term on the right-hand side is

$$|\langle[C,A]\rangle|^2=[(\mathbf{k}-\mathbf{G})\cdot\mathbf{k}]^2|\langle\rho_{\mathbf{G}}|^2. \quad (13)$$

We wish to show that no crystalline order exists by showing that $\langle\psi_{\mathbf{G}}\rangle=0$, where $\langle\psi_{\mathbf{G}}\rangle=\lim_{N\to\infty}N^{-1}\langle\rho_G\rangle$, for $\mathbf{G}\neq0$. At $T\neq0$, $\langle\psi_{\mathbf{G}}\rangle=0$ in any number of dimensions if $[P,H]=0$, where P is the total momentum. In order to enable the system to condense into a crystalline state if periodic boundary conditions are used, one adds a term to the Hamiltonian which breaks the system's translational symmetry. (See Wagner,[16] for example.) However, this procedure is not necessary in our case since the walls break the translational symmetry of the Hamiltonian. We now compute the term $[[C,H],C^+]$ in inequality (10). First we obtain

$$\begin{aligned}\langle[[J_{\mathbf{k}},H],J_{\mathbf{k}}{}^+]\rangle=&\tfrac{1}{4}N[Z/m+1/M]k^6+6\beta(\alpha)N\langle\mathrm{KE}\rangle k^4\\&+\sum_{\mu\nu}(\mathbf{k}\cdot\nabla_\mu\mathbf{k}\cdot\nabla_\nu H)e^{i\mathbf{k}\cdot(\mathbf{R}_\mu-\mathbf{R}_\nu)}\\&+\sum_{ij}\langle(\mathbf{k}\cdot\nabla_i\mathbf{k}\cdot\nabla_jH)e^{i\mathbf{k}\cdot(\mathbf{r}_i-\mathbf{r}_j)}\\&+2\sum_{i\nu}\langle(\mathbf{k}\cdot\nabla_i\mathbf{k}\cdot\nabla_\nu H)e^{i\mathbf{k}\cdot(\mathbf{r}_i-\mathbf{R}_\nu)}\\&+fN^{1/4}\begin{pmatrix}0(1)\\0(N^{1/2})\end{pmatrix},\quad\text{in }\begin{pmatrix}1\\2\end{pmatrix}\text{dimensions},\end{aligned} \quad (14)$$

where $N\langle\mathrm{KE}\rangle$ is the total kinetic energy of the system, β is given by

$$\beta(\alpha)=(Z\langle\mathrm{KE}^{\mathrm{elect}}\rangle\langle(\mathbf{k}\cdot\mathbf{p}_i)^2\rangle/k^2\langle p_i{}^2\rangle\langle\mathrm{KE}^{\mathrm{nucl}}\rangle\times\langle(\mathbf{k}\cdot\mathbf{P}_\nu)^2\rangle/k^2\langle P_\nu{}^2\rangle)\langle\mathrm{KE}\rangle^{-1},$$

where α is the angle between $\mathbf{k}$ and $\mathbf{G}$, and the last term in the equation is due to the contribution of the soft walls to H. The explicit form of H is

$$H=\sum_i\frac{p_i{}^2}{2m}+\sum_\nu\frac{P_\nu{}^2}{2M}+\tfrac{1}{2}\sum_{ij}{}'U_{ij}{}^{EE}+\tfrac{1}{2}\sum_{\nu\mu}{}'U_{\mu\nu}{}^{NN}\sum_{i\nu}U_{i\nu}{}^{NE}+H_w, \quad (15)$$

[13] The results of this section were reported at the Twenty-First Yeshiva University Statistical Mechanics Meeting, March 31, 1969 (unpublished).

[14] N. N. Bogolyubov, Physik Abhandl Sowjetunion 6, 1 (1962); 6, 113 (1962); 6, 229 (1962).

[15] N. D. Mermin and H. Wagner, Phys. Rev. Letters 17, 1133 (1966).

[16] H. Wagner, Z. Physik 195, 273 (1966).

where

$$U_{(|r|)}{}^{EE}=Z^{-2}U_{(|r|)}{}^{NN}=-Z^{-1}U_{(|r|)}{}^{NE}$$
$$=-2\pi e^2|r|, \quad \text{in 1 dimension}$$
$$=-2e^2\ln|r|, \quad \text{in 2 dimensions.} \quad (16)$$

Substituting this Hamiltonian into Eq. (14) gives

$$N\Lambda(\mathbf{k})k^2=\tfrac{1}{4}N[Z/m+1/M]k^4+6_\beta(\alpha)N\langle\text{KE}\rangle k^2$$
$$+N\int[1-2\sin^2(\theta-\alpha)]\binom{0}{2}r^{-2}$$
$$\times[1-\cos(\mathbf{k}\cdot\mathbf{r})]g(\mathbf{r})d\mathbf{r}$$
$$+fN^{1/4}\begin{pmatrix}0(1)\\0(N^{1/2})\end{pmatrix}, \quad \text{in } \binom{1}{2}\text{dimensions}, \quad (17)$$

where

$$g(\mathbf{r})=\frac{1}{N}\left[\int\langle q(\mathbf{R})q(\mathbf{R}+\mathbf{r})\rangle-\frac{N}{\Omega}Z(Z+1)\delta(\mathbf{r})\right]d\mathbf{R}, \quad (18)$$

and $q(\mathbf{R})$ is the *charge* density at $\mathbf{R}$, i.e.,

$$q(\mathbf{R})=Ze\rho^N(\mathbf{R})-e\rho^E(\mathbf{R}). \quad (19)$$

$\rho^N(\mathbf{R})$ and $\rho^E(\mathbf{R})$ are, respectively, the nuclear and electronic number density at $\mathbf{R}$, Ze is the nuclear charge; $N\Lambda(\mathbf{k})k^4=\langle[[J_\mathbf{k},H],J_\mathbf{k}^+]\rangle$, and θ is the angle between $\mathbf{r}$ and $\mathbf{G}$.

Using the definition of Λ. Eqs. (11)–(13), Eq. (10) may be cast into the following form:

$$\frac{1}{N}\langle\rho_{-\mathbf{G}+\mathbf{k}}{}^N\rho_{\mathbf{G}-\mathbf{k}}{}^N\rangle\geq K_BT\frac{|\langle\rho_\mathbf{G}\rangle|^2}{N^2}\frac{[(\mathbf{k}-\mathbf{G})\cdot\mathbf{k}]^2}{\Lambda(\mathbf{k})k^4}.$$

Multiplying both sides of the above inequality by $e^{-2\sigma k^2/2}$, summing over all the allowed values of $\mathbf{k}$, and taking the thermodynamic limit results in

$$D\geq k_BT|\langle\psi_\mathbf{G}\rangle|^2\int d\mathbf{k}\, e^{(-\sigma^2k^2/2)}\frac{[(\mathbf{k}-\mathbf{G})\cdot\mathbf{k}]^2}{\Lambda(k)k^4}, \quad (20)$$

where

$$D=\int S(\mathbf{G}-\mathbf{k})e^{-\sigma^2k^2/2}d\mathbf{k} \quad (21)$$

and

$$S(\mathbf{G}-\mathbf{k})=\lim_{N\to\infty}\frac{1}{N}\langle\rho_{-\mathbf{G}+\mathbf{k}}{}^N\rho_{\mathbf{G}-\mathbf{k}}{}^N\rangle.$$

To conclude the proof, we have to show that D is finite and how $\Lambda(\mathbf{k})$ depends on $\mathbf{k}$. It follows from the definition of D that

$$D=\frac{2\pi}{\sigma^2}\lim_{N\to\infty}\frac{1}{N}\int d\mathbf{R}\,d\mathbf{r}\,\langle\rho^N(\mathbf{R})\rho^N(\mathbf{R}+\mathbf{r})\rangle$$
$$e^{-i\vec{G}\cdot\vec{r}}e^{-r^2/2\sigma^2}. \quad (22)$$

We assume that the system is nonpathological, in the sense that

$$\int_{\Delta\Omega_1}\int_{\Delta\Omega_2}\langle\rho^N(\mathbf{r})\rho^N(\mathbf{R}+\mathbf{r})\rangle d\mathbf{R}\,d\mathbf{r}$$

is finite, where $\Delta\Omega_1$ and $\Delta\Omega_2$ are any two finite domains of integration for $\mathbf{r}$ and $\mathbf{R}$. It then follows that D is finite.

In one dimension, it follows from Eqs. (20) and (17), and the assumption that the kinetic energy per particle is finite, that there exists a $k'\neq 0$ such that

$$D\geq|\langle\psi_\mathbf{G}\rangle|^2\frac{k_BTG^2}{6\langle\text{KE}\rangle}\int_0^{k'}\frac{d|k|}{k^2}e^{-k^2\sigma^2/2} \quad (23)$$

and therefore, that Eq. (2) is not satisfied.

The two-dimensional case is not as simple. It is shown in Appendix B, where we take care of $\Lambda(\mathbf{k})$, that there exists a $\mathbf{k}_1\neq 0$ and a finite quantity b such that inequality (20) implies

$$D\geq|\langle\psi_\mathbf{G}\rangle|^2\frac{k_BTG^2}{b}\int_0^{|k_1|}\frac{d|k|}{|k|}e^{-k^2\sigma^2/2}, \quad (24)$$

which again shows that Eq. (2) is not satisfied in two dimensions.

IV. DISCUSSION

We will first indicate the physical ideas that motivate the choice of C and A in Bogolyubov's inequality. One wants to have $[C,A]\sim\rho_\mathbf{G}/N$ on the right-hand side of the inequality, since this is the operator whose expectation value we want to show vanishes. Now $[J_\mathbf{k}{}^N,\rho_{\mathbf{G}-\mathbf{k}}{}^N]\sim\rho_G{}^N$ and $[J_{\mathbf{G}-\mathbf{k}}{}^N,\rho_\mathbf{k}{}^N]\sim\rho_G{}^N$. One has to decide which one of these, J or ρ, is to be identified with C, and which one with A. $[[\rho,H],\rho^+]$ is independent of the spatial part of H, and nowhere else in Bogolyubov's inequality do the properties of H enter. Therefore, if the proof would go through with this choice of C it would imply no crystalline ordering even in the presence of an external field having the periodicity of the lattice. Therefore, the proof cannot go through with such a choice. One should then choose C to be the current and A the density. That is not enough for the proof to go. We are dealing with two speciew of charged particles, nuclei and electrons. Each of these species has approximately two modes, the plasma mode and the sound mode. The plasma is mode undesirable for the proof to go through for the reasons that follow. We note first that

$$\left[\frac{if(t)}{\partial t}\right]_{t=0}=\frac{1}{2\pi}\int_{-\infty}^{\infty}\omega f(\omega)d\omega$$

and

$$\left[\frac{i\partial}{\partial t}\langle[A(t),A^+(0)]\rangle\right]=\langle[[A,H],A^+]\rangle.$$

Furthermore,

$$J_{\mathbf{k}}^{N}(t)=Mi\frac{\partial\rho_{\mathbf{k}}^{N}}{\partial t},$$

and, from the f sum rule[17]

$$\int_{-\infty}^{\infty}\omega X^{NN}(\mathbf{k},\omega)d\omega=Nk^2/M,$$

where

$$X^{NN}(\mathbf{k},\omega)=\int\langle[\rho_{\mathbf{k}}^{N}(t),\rho_{\mathbf{k}}^{N+}(0)]\rangle e^{i\omega t}dt,$$

it follows that

$$\int\omega^3 X^{NN}(\mathbf{k},\omega)d\omega\sim Nk^2$$

for small k.

This last expression is proportional to $\langle[[C,H],C^+]\rangle$ if we choose $C=J_{\mathbf{k}}^{N}$, and the $\mathbf{k}$ dependence will ruin the proof. On the other hand, if only sound waves were present the previous expression would go as k^4 for small k, which is the desired k dependence. The way to circumvent the plasma mode is to choose C equal to the total mass current density, i.e., electronic plus nuclear, as in Eq. (11). The local center of mass of the two charged species does not feel the plasma motion; in the plasma mode the two charge species move 180° out of phase as in an optical mode in a solid. This is the reason why the choice of C made in Eq. (11) yields $\langle[[C,H],C^+]\rangle\sim Nk^4$.

The previous discussion makes clear why the proof of Sec. III does not go through for a system of electrons in a static positive background, in two dimensions.

If one tries to prove lack of crystalline order for a system of hard cores in a fashion similar to that of Sec. III, one gets $\Lambda=\infty$. The inequality seems useless in this case.

The stability proof of Sec. II implies that for large N,

$$\langle u_i^2\rangle\gtrsim k_BT\chi_T\ln N$$

in two dimensions, which for systems such as a harmonic solid, where Eq. (A2) is valid, implies that

$$|\langle\rho_G\rangle|/N\lesssim N^{-G^2k_BT\chi_T}. \tag{25}$$

On the other hand, the proof of Sec. III implies that

$$|\langle\rho_G\rangle|/N\lesssim(\ln N)^{-1/2} \tag{26}$$

in two dimensions for a system of N nuclei and ZN electrons. It follows that either Eq. (A2) is not a good approximation in this case, or Bogolyubov's inequality provides a poor bound. The latter is probably the case, since the application of the Bogolyubov inequality to a harmonic solid yields an equation like (26), but one knows that Eq. (A2) is valid in this case and therefore Eq. (25) holds for this particular system.

With some modifications, the proof of Sec. III can be extended to partially finite geometries.[18] The extension of Sec. II to partially finite geometries follows the work of Krueger[19] in a trivial way.

Although the results of Sec. II are useless for a system of electrons in a positive background in one dimension, it follows from the results of Sec. III, i.e., that there can be no crystalline order for this system, and Appendix A, that this system is unstable, i.e., it does not satisfy Eq. (1). No such statement can be made about the two-dimensional system.

ACKNOWLEDGMENTS

We would like to thank Professor E. C. Lerner and Professor H. A. Gersch for their comments.

APPENDIX A

We discuss here the connections between Eqs. (1) and (2), i.e., the relations between the stability condition for a solid, $\langle u_i^2\rangle<\infty$, and the crystalline-order condition

$$\lim_{N\to\infty}\frac{|\langle\rho_{\mathbf{G}}\rangle|}{N}\neq 0.$$

It follows from the definitions of $\rho_{\mathbf{G}}$ and $\mathbf{G}$ that

$$\langle\rho_{\mathbf{G}}\rangle/N=\langle e^{i\mathbf{k}\cdot\mathbf{u}}\rangle. \tag{A1}$$

For a harmonic solid[20]

$$\langle\rho_{\mathbf{G}}\rangle/N=e^{-\langle(\mathbf{G}\cdot\mathbf{u}_i)^2\rangle/2} \tag{A2}$$

and the connection is then very clear. Unfortunately, no similar equation can be written in general. Glauber[21] proved Eq. (A2) in "general," but under the assumption of independent linear fields which has only been justified for the harmonic solid. For a reflection invariant system Eq. (A1) may be written as follows:

$$\langle\rho_{\mathbf{G}}\rangle/N=\langle\cos(\mathbf{G}\cdot\mathbf{u}_i)\rangle=\int\cos(\mathbf{G}\cdot\mathbf{u}_i)P(\mathbf{u}_i)d\mathbf{u}_i, \tag{A3}$$

where $P(\mathbf{u}_i)$ is the probability that the ith particle is at a point $\mathbf{u}_i$ away from its lattice position.

It follows from Eq. (A3) that if $P(\mathbf{u})$ is a reasonable function of $\mathbf{u}$ then $\langle u_i^2\rangle<\infty$ implies Eq. (2) but the converse does not follow necessarily. In fact, an example can be given in which Eq. (2) is satisfied but Eq. (1) is not. A system under the influence of an external potential such as $c\rho_{\mathbf{G}}+\bar{c}\rho_G^+$ will satisfy Eq. (2) in any

[17] P. Nozières and D. Pines, Nuovo Cimento 9, 4 (1958).

[18] J. F. Fernández (to be published).

[19] D. A. Krueger, Phys. Rev. Letters 19, 563 (1967).

[20] D. Pines, *Elementary Excitations in Solids* (W. A. Benjamin, Inc., New York, 1963), pp. 40–44.

[21] R. Glauber, Phys. Rev. 98, 1692 (1955).

number of dimensions, but, as shown in Sec. II, $\langle u_i^2\rangle$ would equal infinity in two dimensions even in the presence of the external potential, as long as the compressibility remained finite. Therefore, Eq. (2) implies Eq. (1), but the converse is not true.

APPENDIX B

It is shown in this appendix that there exist a $k_1\neq 0$ and a finite quantity b such that

$$\int \frac{d\mathbf{k}\, e^{-k^2\sigma^2/2}[(\mathbf{k}-\mathbf{G})\cdot\mathbf{k}]^2}{\Lambda(k)k^4} \geq \int_0^{|k_1|} \frac{d|k|\, e^{-k^2\sigma^2/2}G^2}{b|k|} \tag{B1}$$

which enables one to go from inequality (20) to inequality (24). Consider the integral

$$\int_0^{2\pi} d\alpha\, [(\mathbf{k}-\mathbf{G})\cdot\mathbf{k}]^2/\Lambda(\mathbf{k}),$$

where α has been defined as the angle between $\mathbf{k}$ and $\mathbf{G}$. Since $\Lambda(\mathbf{k})>0$, it follows that

$$\int_0^{2\pi} d\alpha \frac{[(\mathbf{k}-\mathbf{G})\cdot\mathbf{k}]^2}{\Lambda(\mathbf{k})} \geq \int_{\frac{3}{4}\pi}^{\pi} d\alpha \frac{[(\mathbf{k}-\mathbf{G})\cdot\mathbf{k}]^2}{\Lambda(\mathbf{k})}.$$

Also,

$$[(\vec{\mathbf{k}}-\vec{\mathbf{G}})\cdot\vec{\mathbf{k}}]^2 > \tfrac{1}{2}k^2G^2 \quad \text{for} \quad \tfrac{3}{4}\leq\alpha\leq\pi.$$

It follows, then, that

$$\int_0^{2\pi} d\alpha \frac{[(\mathbf{k}-\mathbf{G})\cdot\mathbf{k}]^2}{\Lambda(\mathbf{k})} > \int_{\frac{3}{4}\pi}^{\pi} d\alpha \frac{k^2G^2}{2\Lambda(\mathbf{k})}. \tag{B2}$$

It follows from the fact that, in general,

$$\int_{x_1}^{x_2} \frac{dx}{f(x)} \geq \int_{x_1}^{x_2} \frac{dx}{\langle f(x)\rangle}$$

if $f>0$, that the inequality (B2) is strengthened if we replace $\Lambda(\mathbf{k})$ by

$$\Lambda^1(k) = \frac{4}{\pi}\int_0^{2\pi} \Lambda(\mathbf{k})d\alpha > \frac{4}{\pi}\int_{\frac{3}{4}\pi}^{\pi} \Lambda(\mathbf{k})d\alpha.$$

Therefore,

$$\int_0^{2\pi} d\alpha \frac{[(\mathbf{k}-\mathbf{G})\cdot\mathbf{k}]^2}{\Lambda(\mathbf{k})} \geq \tfrac{1}{4}\pi\tfrac{1}{2}k^2G^2\frac{1}{\Lambda^1(k)}. \tag{B3}$$

In order to arrive at inequality (B1), we now have to consider the behavior of $\Lambda^1(k)$ for small k. From the definition of $\Lambda^1(k)$, and Eq. (17) we write

$$\lim_{k\to 0}\Lambda^1(k) = 8\left\{\frac{1}{4}\left[\frac{Z}{m}+\frac{1}{M}\right]+3\text{KE}\right\} + \lim_{\alpha\to 0}\lim_{k\to 0}\frac{4}{\pi}\frac{1}{k^2} \times \int e^{-\alpha|r|}[1-2\sin^2(\theta-\alpha)] \times[1-\cos\mathbf{k}\cdot\mathbf{r}]\cdot g(\mathbf{r})d\alpha\, d\theta\, r\, dr$$

with the help of virial theorem[22] in two dimensions

$$2p\Omega/N = 2\langle\text{KE}\rangle + \tfrac{1}{2}\int g(\mathbf{r})d\mathbf{r},$$

where $g(\mathbf{r})$ has been defined in Eq. (18), we get

$$\lim_{k\to 0}\Lambda^1(k) = 8\left[p\frac{\Omega}{\text{N}}+2\langle\text{KE}\rangle\right].$$

Therefore, assuming the pressure and kinetic energy per nucleus to be finite, then there exists a finite quantity b and a $\mathbf{k}_1\neq 0$ such that $\Lambda^1(k)<8b/\pi$ for $|\mathbf{k}|\leq|\mathbf{k}_1|$. Inequality (B1) follows.

[22] J. C. Slater, J. Chem. Phys. 1, 687 (1933); J. de Boer, Physica **15**, 843 (1949); P. N. Argyres, Phys. Rev. **154**, 410 (1967).