Dynamics of a Model of a Supersolid

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A first order phase transition in a model of a superfluid is studied. The transition occurs when the energy gap for rotons decreases below a critical value. The ground state is a purely real, positive, and periodic wave function, which represents a new solid phase with long range phase order, as in superfluids. Properties and characteristics of this supersolid are discussed. Although uniform rotation occurs without dissipation, dissipationless flow around an obstacle is not possible. We show the existence of "supersolid vortices" and propose some experiments to test the theory.

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Penrose and Onsager (PO) [1] were the first to consider a Bose-Einstein condensation in solids and concluded that such a "supersolid" was impossible. Since then, this question has been revisited by various authors [2], both experimentally and theoretically, and there has been no compelling evidence of the existence of such a supersolid state. In [3], we studied a model of a superfluid with a roton minimum, the occurrence of this minimum arising from a nonlocal cubic interaction in a Gross-Pitaevskiĭ equation. In this model, when the flow speed increases, there is a supercritical bifurcation to a lamellar pattern of density and phase modulations, as was first suggested by Pitaevskiĭ [4]. This pattern may be seen as a kind of crystal, and the transition to this state is linked to an increase of the "thermodynamic" parameter which is the superfluid velocity. This suggests that a transition to a crystalline order could appear in the same model without velocity, by appropriately tuning another parameter, such as the density. It happens that, by increasing the density, the roton energy gap diminishes and there is a critical value for which the system undergoes a subcritical bifurcation (first order phase transition) to a regular pattern with a degenerate wave number. This pattern is hexagonal in two dimensions (2D) and has a bcc structure in 3D. This is a crystal in the sense of Landau: a many-body system where the lowest energy is a state of modulated density ([5], part 1, Sec. 145). It is a slightly different view of the usual crystal order, because there is *a priori* no simple relation for the number of atoms per unit cell. As shown by Chester [2] the proof by PO fails in that case, because it assumes each lattice site occupied by a particle. If the average number of atoms per cell is less than what is predicted by simple crystallographic enumeration, this system is a possible representation of a "superfluid" of vacancies, a system studied by Andreev and Lifshitz [2].

This "quantum crystal" is the ground state of the system and is described by a purely positive and real condensate wave function (up to a global phase). However, it is not exactly a crystal from the classical point of view, since the points in space are all correlated by the phase ϕ of the condensate wave function ψ as in a superfluid where collective motion of the atoms involves such a phase. This long range phase order is broken at finite temperature only in 3D, as shown by the Landau-Peierls argument ([5], part 1). A system of identical bosons at finite density seems to have only *two* possible ground states: a superfluid or a "supersolid," each with a uniform phase ϕ. The first order phase transition is controlled in the present model by a single parameter (later denoted as g), a combination of the de Boer parameter and the density, which describes well this transition, as it combines the zero-point motion and the number density.

For some time an accepted test for the supersolid state [2] was the possibility of nondissipative *matter flow* through a fixed crystal lattice. Such a flow is not possible in the present model at $T = 0$ K where a flow around an object leads to the creation of defects in the crystal which carry away some energy and yield plastic irreversible deformations. However, a possible way of detecting the supersolid state, other than thermodynamic measurements of the transition, would be to find evidence for the quantum phase, which can be done in a rotating supersolid. For low angular velocities the phase will not be constant and, for increasing rotation speed, the system could create vortices on the boundary of the container which move to the center when the speed increases.

Our starting point is the Gross-Pitaevskiĭ [6] equation:

$$i\hbar\frac{\partial\psi}{\partial t} = \frac{\hbar^2}{2m}\nabla^2\psi - \int d\mathbf{r}'\, U(|\mathbf{r}-\mathbf{r}'|)|\psi(\mathbf{r}',t)|^2\psi(\mathbf{r},t), \tag{1}$$

where ψ is the condensate wave function (a classical field); $U(r)$, the interparticle potential; m, the mass of the particles; and $2\pi\hbar$, Planck's constant. Equation (1) has a uniform solution: $\psi_0 e^{i\frac{E_0}{\hbar}t}$, where $E_0 = |\psi_0|^2\int d\mathbf{r}'\, U(r')$. A plane wave linear perturbation $\delta\psi_k e^{i\frac{E_0}{\hbar}t}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)}$ satisfies the dispersion relation [6]

$$\hbar\omega_k = \frac{\hbar^2}{m}\sqrt{\frac{k^4}{4} + \frac{m}{\hbar^2}|\psi_0|^2\hat{U}(k)k^2}\,, \tag{2}$$

where $\hat{U}(k) = \int d\mathbf{r}\, U(\mathbf{r})e^{i\mathbf{k}\cdot\mathbf{r}}$. Hereafter we take a soft sphere interaction [i.e., $U(r) = U_0 > 0$, $r < a$; $U(r) = 0$, $r > a$]. This not too realistic interaction makes nu-

merical simulations possible, however, our results depend mostly on the existence of a roton minimum.

Scaling arguments [k scales as $1/a$, and $U(k) \sim a^3 U_0$] suggest that the excitation spectrum of the liquid can be written as $\hbar\omega_k = \frac{\hbar^2}{ma^2} f_g(ka)$, where f_g is a function [defined directly from (2)] and g is the dimensionless quantity $g = 4\pi U_0 \frac{ma^2}{\hbar^2} na^3$, where $n = |\psi_0|^2$ is the number density of the uniform solution. The speed of sound is $c_s = \frac{\hbar}{ma}\sqrt{g/3}$. The spectrum ω_k may have four different shapes depending on g: (i) $g = 0$, which represents a perfect gas, with a ballistic spectrum $\omega_k \sim k^2$ and no superfluidity, as was first noticed by Landau; (ii) $0 < g < g_r$, the spectrum grows monotonically in k, without a roton minimum; (iii) $g_r < g < g_0$, a Landau spectrum with rotons; and (iv) $g = g_0$, such that $\omega_k = 0$ for some k_0, as at the edge of the phonon branch in solids. A reasonable value for k_0 is $2\pi/a$ (≈ 2.4 Å^{-1} for He II), roughly twice the wave number of the maximum of the Landau spectrum. (All the estimations concern helium, the energies will be in kelvin. It is useful to note that $\frac{\hbar^2}{m} = 12$ K Å^2, the quantum of circulation is $\frac{\hbar}{m} = 158$ m/s Å, and $a = 2.57$ Å.) This suggests that the roton minimum in He II is like a ghost of the solid state.

At $T = 0$ K, there is no stable liquid structure for large values of g, which is likely the case for all bosonic elements. On the other hand, $g \to \infty$ could mean that the zero-point motion vanishes or that quantum effects are negligible (neglecting the possibility of very dense states).

With a convenient value of g we obtain a Landau spectrum with rotons [3]. If we increase g, for instance, by keeping a, U_0 constant and increasing n, we observe that the roton gap decreases. We expect that there is a critical value $g_r < g_c < g_0$ for which the system crystallizes. The density increase might be achieved in a physical system by increasing the pressure and/or by cooling. Crystallization due to the roton minimum can be expected near the real solid phase since solid helium exists only for high pressures. The transition occurs when the roton minimum is near the k axis for zero frequency, if we use Landau's notation for rotons: $\hbar\omega_k = \Delta + \frac{\hbar^2}{2\mu}(|k| - k_r)^2$ for $k \approx k_r$. In our picture Δ, k_r, and μ are nontrivial functions of g. However, Δ decreases and the roton minimum k_r, increases as g increases. In our model, the functions $\Delta(g)$, $k_r(g)$, and $\mu_r(g)$ are known. In applications they should be determined experimentally. There is a first order phase transition for $\Delta = \Delta_M > 0$. The wave number for the bifurcation is k_r, which is the lattice number.

We will see now that this quantum crystal exists as a ground state of (1). Writing (1) as a function of the density, $\rho = |\psi|^2$, and the phase ϕ of ψ, we obtain a set of "perfect solidlike" equations:

$$\partial_t \rho = \frac{\hbar}{m} \nabla \cdot (\rho \nabla \phi), \tag{3}$$

$$\hbar \partial_t \phi = \frac{\hbar^2}{8m\rho^2}(\nabla\rho)^2 - \frac{\hbar^2}{4m\rho}\nabla^2\rho + \frac{\hbar^2}{2m}(\nabla\phi)^2 + \int d\mathbf{r}'\, U(|\mathbf{r} - \mathbf{r}'|)\rho(\mathbf{r}'). \tag{4}$$

In the ground state the wave function ψ is real (except for the global phase $E_0 t/\hbar$) because a nonuniform phase ϕ increases the energy via a $(\nabla\phi)^2$ term. This ground state can be determined by Eq. (4) with $\phi = (E_0/\hbar)t$, or by minimizing the functional

$$E[\rho_0] = \int d\mathbf{r} \frac{\hbar^2}{8m\rho_0}(\nabla\rho_0)^2 + \int d\mathbf{r}\, d\mathbf{r}'\, U(|\mathbf{r} - \mathbf{r}'|)[\rho_0(\mathbf{r}) - n][\rho_0(\mathbf{r}') - n]. \tag{5}$$

The solution ρ_0 is identical to the solution for a classical solid, since the phase ϕ is uniform.

We sketch the solution in 2D. We find an approximate solution ρ_0 as a modulation of the uniform solution n, i.e., $\rho_0(\mathbf{r}) = n + n\left(\sum_{j=1}^3 A_j e^{i\mathbf{k}_j\cdot\mathbf{r}} + \text{c.c.}\right)$, with the three complex amplitudes A_j such that $|A_j| \ll 1$, and the vectors $\mathbf{k}_j$ form an equilateral triangle ($\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3 = 0$) with a magnitude $|\mathbf{k}_j| = k_r$. If we put $\rho_0(\mathbf{r})$ in (5) and expand in powers of A_j, since $|A_j| \ll 1$, we obtain

$$\frac{E}{V} = n\frac{\hbar^2 k_r^2}{2m}\left(2\mu^2 \sum_{i=1}^3 |A_i|^2 - \frac{3}{4}(A_1A_2A_3 + A_1^*A_2^*A_3^*) + \frac{1}{2}\sum_{i=1}^3 |A_i|^4 + 2\sum_{i<j} |A_i|^2|A_j|^2 + \cdots,\right) \tag{6}$$

with $\mu = \frac{\hbar\omega_{k_r}}{\hbar^2 k_r^2/m}$. The ground state is at the minimum of the energy, i.e., $\frac{\delta E}{\delta A_i} = 0$. The hexagonal crystal solution is ($A_j = R_j e^{i\varphi_j}$) such that each of the three amplitudes is equal to

$$R_j = (3 \pm \sqrt{9 - 640\mu^2})/40, \quad j = 1, 2, 3; \tag{7}$$

and the phases satisfy $\varphi_1 + \varphi_2 + \varphi_3 = 0 (\text{mod} 2\pi)$. The negative sign in (7) is always unstable. The metastable solution [the positive sign in (7)] exists if $\mu^2 < 9/640$ equivalent to $\Delta < \Delta_c = \frac{3}{\sqrt{40}}\frac{\hbar^2 k_r^2}{4m}$ and it becomes stable at the Maxwell point (MP): $\Delta_M = \frac{3}{\sqrt{41.28}}\frac{\hbar^2 k_r^2}{4m}$, i.e., when the crystal and liquid energy are equal. For He II ($k_r = 1.95$ Å^{-1}) $\Delta_c = 5.4$ K and $\Delta_M = 6.0$ K. The corresponding energy barrier (per particle) at the MP is about $E/N \approx 1.2 \times 10^{-3}$ K. We note that this energy is reduced, by hybridization, by 3 orders of magnitude with respect to all energy scales in the data. $\Delta_{c/M}$ are bigger in 3D than in 2D. The relevant 3D crystal structures are bcc or hcp. In the bcc configuration we have six amplitudes

and the wave vectors form an octahedron [7]; each vector participates in *two* equilateral triangles, which produces the most stable configuration. The metastable bcc lattice exists if $\Delta < \Delta_c = \frac{3}{\sqrt{22}} \frac{\hbar^2 k_r^2}{4m}$, and becomes stable for $\Delta < \Delta_M = \frac{1}{\sqrt{44}} \frac{\hbar^2 k_r^2}{4m}$, which gives $\Delta_c = 7.3$ K, $\Delta_M = 8.11$ K, and $E/N \approx 3.2 \times 10^{-3}$ K.

Let us study now the dynamics of a crystalline ground state or supersolid. Following the principles of elasticity theory, we define the deformation vector $\mathbf{u}(\mathbf{r}, t)$, a slowly varying function of $\mathbf{r}$ and t, and solve (3) and (4) by a long wavelength expansion: $\rho(\mathbf{r}, t) = \rho_0(\mathbf{r} - \mathbf{u}(\mathbf{r}, t)) + \rho_1(\mathbf{r} - \mathbf{u}(\mathbf{r}, t)) + \cdots$. The solvability condition for ρ_1 gives

$$\partial_{tt} u_i = \lambda_{iklm} \partial_{kl} u_m + \cdots, \tag{8}$$

where $\partial_i \equiv \frac{\partial}{\partial x_i}$ and repeated indices are summed. The tensor λ_{iklm} for hexagonal symmetry in two dimensional systems has only two independent components [8] (the mode of uniform compression and the shear modulus) and λ can be written explicitly as integrals of functions of ρ_0 and of its gradients. The explicit calculations are long, but finally we get that both the compression and shear modulus lead to a speed of sound for elastic deformations of order $c_e \sim \frac{\hbar k_r}{m} \approx 310$ m/s. Equation (8) is the classical wave equation in crystals. A second equation allows us to compute the phase ϕ for a given u_i. In contrast to the displacement field this phase has both a slow and a fast space dependence. It is deduced from the mass conservation (3) and is such that, for $\partial_t u_i$ given, ϕ makes stationary the functional (with $\mathbf{A} = \dot{\mathbf{u}}$)

$$\mathcal{E}[\phi, \mathbf{A}] = \frac{1}{2} \int d\mathbf{r}\, \rho_0(\mathbf{r} - \mathbf{u})[(\hbar/m)\nabla\phi + \mathbf{A}(\mathbf{r})]^2, \tag{9}$$

which is positive definite because $\rho_0(\mathbf{r}) > 0$. In this way the nonstationary deformation is connected with the mass current $\mathbf{j} = \rho \frac{\hbar}{m} \nabla\phi$. The energy (9) is a Galilean invariant ($\mathbf{A} = \dot{\mathbf{u}}$, $u_i \to u_i + v_i t$, and $\partial_i \phi \to \partial_i \phi - \frac{m}{\hbar} v_i$, v_i uniform). As this Galilean invariance is global, there is no dissipationless superflow through the lattice in this model at $T = 0$ K. Imposing numerically a large scale phase gradient in the supersolid, where a fixed obstacle is immersed, this puts the lattice in motion (as predicted by our argument) and the displacement of the lattice around the obstacle produces defects and dissipation, even at very low velocities, as shown in Fig. 1. On the other hand, (9) allows us to compute $\nabla\phi$ for two different cases: for a given acoustical field $\mathbf{u}$ and for an adiabatic deformation imposed by the boundary conditions.

As noticed by Leggett [2] the phase in a rotating supersolid cannot be uniform. In our model, the phase of a rotating supersolid is given for low angular velocities (Ω) by the minimization of $\mathcal{E}[\phi, \mathbf{\Omega} \times \mathbf{x}]$. As $\mathbf{\Omega} \times \mathbf{x}$ changes slowly as a function of $\mathbf{x}$, compared to the lattice period, one can split $\nabla\phi$ into a slowly varying phase $\tilde{\phi}$ and a periodic part, both depending linearly on $\mathbf{\Omega} \times \mathbf{x}$. Homogenization techniques yield a uniform ρ_{eff}, a rank 2 tensor in general, which becomes a scalar for a hexagonal lattice. This allows us to write the energy (to be minimized) as $\frac{1}{2}\rho_{\text{eff}} \int d\mathbf{x}[(\hbar/m)\nabla\tilde{\phi} + \mathbf{\Omega} \times \mathbf{x}]^2$. This yields an effective inertia tensor by noting that this energy is quadratic in Ω. $\tilde{\phi}$ is linear in Ω and is determined by the solution of Laplace's equation, with the boundary condition $[(\hbar/m)\nabla\tilde{\phi} + \mathbf{\Omega} \times \mathbf{x}] \cdot \hat{\mathbf{n}} = 0$, where $\hat{\mathbf{n}}$ is normal to the boundary.

FIG. 1. Numerical simulation of a plastic flow around a disk. We plot the value of the density $|\psi|^2$. The wave function ψ vanishes on the black disk representing the obstacle. The flow velocity (from left to right) is much lower than the speed of sound. The arrows show the defects of the hexagonal pattern, which form a 5-7 pair. Insets show a numerical simulation of a supersolid vortex. We plot (a) $|\psi|^2$ (the small arrows pointing at the vortex core) and (b) ϕ.

As in Ref. [9] vortices appear spontaneously if the local speed on the boundary of the container becomes supersonic, i.e., $\Omega > \Omega_c = \frac{c_e}{L} = \frac{\hbar k_r}{mL}$, where L is the size of the container. "*Supersolid vortices*" are stationary solutions of Eq. (1) with a $\pm 2\pi$ phase jump around the vortex core, as can be seen in Fig. 1(b). Vortices are topological defects and cannot be removed by any infinitesimal perturbation of ψ. The vortex dynamics is quite different from the usual case of superfluids, since there is an interaction with the periodic distribution ρ_0; however, the calculation can be done for the two dimensional case (rectilinear vortices) in the usual way. The vortex velocity is

$$\mathbf{v}_{\text{vortex}} = -\frac{\hbar}{m}(\nabla\phi \pm \hat{\mathbf{z}} \times \nabla \ln \rho_0) + \mathbf{\Omega} \times \mathbf{x}, \tag{10}$$

where $\hat{\mathbf{z}}$ is parallel to the linear vortices which we consider also parallel to the angular velocity, and $\pm$ refers to the two possible choices of the quantum of circulation. The right hand side of (10) is evaluated at the position of the vortex. Note, however, that vortices are not, strictly speaking, described by minimization of (9), since $\psi = 0$ at the vortex core. Only the far field of a stationary vortex is given by minimization of $\mathcal{E}[\phi, 0]$, with the appropriate circulation condition. Insets of Fig. 1 show ϕ and $|\psi|^2$ near the core of such a vortex solution.

Another way to generate vortices is as follows. Imagine a superfluid vortex in He II, and freeze the system. If this solid He is not a supersolid, the vortex disappears. If it is a supersolid, the vortex cannot disappear in the crystallization process and one of two things may happen: (1)

the vortex in the fluid is pushed by the solid liquid interface and driven to the container boundary; or (2) the crystal phase grows inside the vortex core first, then it grows elsewhere to form a "supersolid vortex" [numerical simulations of (1) seem to confirm the latter]. Next we melt again the supersolid. The vortex cannot disappear and thus we obtain a vortex in He II which could be detected with the usual methods in superfluids. If this vortex has survived the freezing-melting process, we would have proof of the existence of a supersolid. However, the first possibility might happen and direct measurements of a nonuniform phase could fail. A way to avoid this problem could be by freezing a circulating superfluid in a multiconnected domain. (This idea was suggested by B. Castaing.)

We end this Letter with some remarks.

(i) The Landau-Peierls argument shows that the thermal phase fluctuations are lower for higher densities n, in the following way (see [5], part 2): $\langle\phi^2\rangle = \int \frac{d^D k}{(2\pi)^D}|\phi_k|^2$, with $|\phi_k|^2 = \frac{m e_k}{n\hbar^2 k^2}(\nu_k + 1/2)$, where $e_k = \hbar c_e k$ is the energy of elastic deformations, $\nu_k = (e^{\beta e_k} - 1)^{-1}$ is the Bose-Einstein distribution, and $\beta = 1/k_B T$. This leads (for $D = 3$) to $\langle\phi^2\rangle = \left(\frac{T}{T_0}\right)^2$, $T_0^2 = 12\hbar^3 n c_e/m$. This suggests that the transition line (between supersolid He and solid He) in the T-n phase diagram is given by $T(n) \sim \sqrt{n}$ (we neglect the dependence of c_e on n which means that k_r does not depend much on n). The long wavelength thermal phase fluctuations diverge logarithmically for 2D, as is well known.

(ii) We suggest that the He II supersolid transition occurs by increasing the density; therefore an estimate of the critical density n_c is possible, in principle, from the knowledge of $\Delta(g \sim n)$, together with the critical gap value Δ_c. In the absence of experimental data for $\Delta(n)$ we can fit, from [10], Δ as a function of the pressure (p). This gives $\Delta = \Delta(0)(1 - p/p_0)$, with $\Delta(0) = 8.74$ K and $p_0 = 157$ bars. The transition pressure for a bcc lattice would then be $p_c = 26$ bars.

(iii) The approach we present neglects quantum fluctuations, and one might wonder how they change this picture. One could imagine that dissipationless superflow is possible by quantum tunneling from one location of this crystal to another one shifted by one wavelength in the flow direction. This location indeed has the same energy, and so no dissipation is needed. Consider a flow around a macroscopic obstacle. Tunneling will be nonnegligible if the barrier height is independent of the size of the obstacle. This depends on the space dimension. The energy of the crystal interacting with the obstacle needs to be a periodic function of the location $\mathbf{x}_0$ of the obstacle. Let E_{int} be this energy. A Gibbs-like development shows that, for a macroscopic system, E_{int} expands like

$$E_{\text{int}} \approx eL^D + \gamma L^{D-1} + e'_{\text{int}}(\mathbf{x}_0),$$

where eL^D is the volume (bulk) energy and γL^{D-1} is the surface energy (both are independent of $\mathbf{x}_0$). The last term is the desired periodic function; as it is one order less than the surface term it is of order L^{D-2}, where L is the size of the obstacle. This implies that the energy barrier is independent of L in 2D (and thus that tunneling flow cannot be discarded). This barrier is of order L in 3D, where the tunneling flow can be neglected.

(iv) Let us comment on the finite temperature behavior of this model. Phonons are linear perturbations $\psi_1(\mathbf{r})e^{i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)}$ to ψ_0. As such they contribute to the density by an amount $|\psi_1(\mathbf{r})|^2$. Similarly, a plane wave carries some mass flow. This gives the idea that thermal excitations in the supersolid state behave like the normal fluid component of a superfluid. Thus "normal" mass flow should accompany a heat flux (without temperature gradient) as in a superfluid. Between two plates, the normal mass flow should be balanced by a supersolid displacement in the opposite direction. But, contrary to the superfluid case, and due to the supersolid lattice, the supersolid "flow" could be blocked by fixed obstacles, and in this case, the heat flux should be accompanied by a temperature difference between the plates. On the other hand, a response of a supersolid to imposed mechanical stress will be a deformation, as for an ordinary solid, not a normal flow through the lattice.

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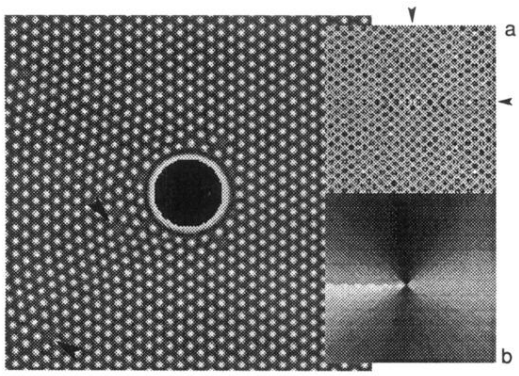

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