

## Phenomenological Ginzburg-Landau theory for supersolidity

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A Ginzburg-Landau theory is proposed in which the supersolid state is viewed as a system displaying features of an ordinary solid and of a superfluid. The theory shows that the superfluid part is responsible for a nonclassical rotational inertia (NCRI) behavior, but the ordinary part (the lattice) is responsible for elastic behaviors usually seen in solids. Moreover, the superfluid part contributes to an excess of heat capacity near the supersolid-ordinary solid transition. The theory provides a coherent picture, at least at the macroscopic scale, of supersolidity that reconciles (NCRI) and the heat-capacity measurements. The parameters of the Ginzburg-Landau free energy are estimated using experimental data, hence a healing length of the order of 100 nm and a critical speed of the order of 0.1 m/s are predicted, both results consistent with recent studies by Kubota and co-workers.

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### I. INTRODUCTION

Solid helium below temperatures of the order of 0.1 K displays very anomalous properties.<sup>1</sup> Among them, the anomalous drop of the effective (measurable) moment of inertia of solid helium has generated enormous speculation about the existence of supersolidity, first predicted forty years ago.<sup>2–4</sup> Indeed, recent experiments by Kim, Chan, and co-workers<sup>5–8</sup> may be interpreted as a nonclassical rotational inertia (NCRI), one of the characteristic of supersolidity.<sup>4</sup> The “experimentum crucis,” the so-called “blocked annulus experiment,”<sup>6,9</sup> placed evidence consistent with the existence of a macroscopic superflow. In the same line, Kim and collaborators have found that supersolidity in a steady rotating frame is in agreement with a superfluid behavior.<sup>10</sup> Although, to this day, eight groups<sup>9,11–15</sup> confirm Kim and Chan’s findings, the value of the NCRI depends on many factors such as crystal sample preparation, cell material, etc. Moreover, annealing was shown to lower systematically the superfluid fraction.<sup>8,11</sup> Today, the zero-temperature limit of the NCRI fraction (NCRIF) is spanned over three orders of magnitude, from 20% to 0.02%, without any rational explanation. Furthermore, an alternative view that unifies the NCRI and shear modulus experiments under the assumption of a complex rheology<sup>16</sup> has been considered.

In the context of mechanical properties, up to date, the responses of solid <sup>4</sup>He to pressure<sup>17,18</sup> and chemical potential<sup>19,20</sup> gradients showed no evidence of superflow in the solid. More important, the mechanical response to shear shows an increase of the effective shear modulus of solid helium with an intriguing similar dependence on temperature of the observed NCRI.<sup>21,22</sup> Similarly, sound measurements of cavity resonances maybe interpreted in terms of a shear modulus anomaly.<sup>23,24</sup> Finally, thermodynamical measurements show an anomaly by an excess of heat capacity at temperatures lower than 0.1 K.<sup>25–27</sup> For more details, the reader may consult the reviews.<sup>28–32</sup>

This issue remains controversial, partly due to the difficulty of investigating the helium system both theoretically and experimentally, for instance, the apparent contradictory feature of solid helium below 100 mK: Supersolid helium behaves like a coherent superfluid in a nonclassical rotational

inertia experiment but its behavior is of an ordinary solid in pressure/external force driven experiments.

This observation indicates that supersolidity possesses a quite complex behavior: indeed there are two different large scale collective motions, in some sense similar to the original two fluid model of Landau for superfluid helium: one is the lattice displacements as in ordinary solids, while the second is a superfluid motion. The lattice and the superfluid motion maybe realized independently. In some situations, depending on the boundary conditions, the system responds as an ordinary solid, and in other cases as a superfluid type of motion. In this sense, because of a rotation the superfluid mode is needed by the boundary conditions, while a gradient pressure does not require a superflow to be at equilibrium, because elasticity balances pressure. This point was satisfactorily explained,<sup>33</sup> where a theory for macroscopic equation of a supersolid was proposed. In Ref. 33 the existence of a nonclassical rotational inertia fraction in the limit of small rotation speed and no superflow under small (but finite) stress nor external force is shown.

On the other hand, nowadays there is no satisfactory explanation of the observed excess of heat capacity near the transition temperature. It may be a consequence of a structural (lattice) property or because of the existence of a superfluid behavior. One may ask *is this excess of heat capacity related to the superfluid fraction?*

The problem of supersolidity represents an ambitious challenge; we shall restrict ourselves to only a few aspects of supersolidity, namely the aforementioned hypothetical relation between the excess of heat capacity and the superfluid fraction. This paper may reach, in the opinion of the author, a coherent understanding of these two macroscopic properties of the supersolid state, which could be tested in experiments. Although of a phenomenological character, we think that the predictions should be pertinent to solid helium-4 at low temperatures (below 0.1 K). More precisely, we develop a Ginzburg-Landau (GL) theory and provide a self-consistent approach valid near the transition temperature characterized by an excess of the specific heat at the transition temperature and an increase of the supersolid fraction. The model displays a NCRI in a rotating container, as well as ordinary elastic behavior. We fit the Landau-Ginzburg parameters from the current experimental

data from NCRI and heat-capacity experiments and we predict values for the coherence length, critical velocity, as well other quantities. These predictions depend on the zero-temperature limit of the superfluid density, which is considered as a given parameter. The main properties predicted by the theory are the existence of a NCRI, four sound modes instead of the usual three normal modes in solids, quantized vortices, and persistent currents. Similar works by Dorsey, Goldbart and Toner,<sup>34</sup> Ye,<sup>35</sup> Toner,<sup>36</sup> and Anderson<sup>37</sup> were considered previously but with the obvious differences for the reader.

## II. GINZBURG-LANDAU ORDER,PARAMETER EXPANSION

We shall make the hypothesis that a supersolid behaves as a “two fluid model,” that is composed by a superfluid part (responsible of NCRI and of an excess of heat capacity) and the lattice structure responsible for the usual solid behavior. Near the transition ordinary solid-supersolid phase it is expected that the free energy of the superfluid part  $F_{ss}$  (meanwhile, we shall consider only the supersolid part of the free energy) could be expanded in series of an order parameter  $\psi$ ,

$$F_{ss}(T) = \int \left( -\alpha(T)|\psi|^2 + \frac{\beta(T)}{2}|\psi|^4 + \frac{\hbar^2}{2M}|\nabla\psi|^2 \right) dV. \quad (1)$$

The minimum free energy provides the equilibrium order parameter, which satisfies the time-independent GL equation:

$$\frac{\hbar^2}{2M}\nabla^2\psi + \alpha(T)\psi - \beta(T)|\psi|^2\psi = 0. \quad (2)$$

For a homogeneous state one has  $|\psi_{ss}|^2 = \alpha(T)/\beta(T)$ . The three parameters  $\alpha(T)$ ,  $\beta(T)$ , and  $M$  are functions of the temperature  $T$ , pressure, etc. However, only two of them are independent because the order parameter  $\psi$  has an arbitrary normalization, thus we need only two measurements to fit the parameters. Those parameters may be computed with the aid of the experimental data of the NCRI and specific heat as follows (this scheme was originally proposed by Ginzburg and Pitaevskii<sup>38</sup>). First, consider the total free energy of a homogeneous system,  $F_{ss} = -\frac{\alpha(T)^2}{2\beta(T)}V$ , thus the excess of specific heat of the supersolid phase is

$$C_{\text{peak}} = -T \frac{\partial^2 F_{ss}}{\partial T^2} = T V \frac{\partial^2}{\partial T^2} \left( \frac{\alpha(T)^2}{2\beta(T)} \right).$$

Next, consider a superflow as a cause of a nonuniform phase, namely  $\psi_{\text{flow}} = \sqrt{\alpha(T)/\beta(T)}e^{i\phi}$ . The superflow energy of this state is  $E = \frac{\hbar^2}{2M} \frac{\alpha(T)}{\beta(T)} \int |\nabla\phi|^2 dV$ . Identifying the superfluid velocity by  $\mathbf{v}_s = \frac{\hbar}{m} \nabla\phi$  and the superflow energy with  $E = \frac{1}{2} f^{ss} m\rho \int |\mathbf{v}_s|^2 dV$ , where  $m$  is the atomic mass of helium,  $\rho = N/V$  is the number density of the solid, and  $f^{ss}(T)$  is the superfluid fraction, one obtains

$$f^{ss}(T) = \frac{m}{M} \frac{\alpha(T)}{\rho\beta(T)}.$$

Finally, the Ginzburg-Landau expansion has meaning only if  $\alpha(T)$  vanishes at the critical temperature  $T_c$  (about 0.1 K in experiments) and all terms of the free energy (1) are of the same order. As usual, one expands in Taylor series the parameters

and we shall keep in the free energy only the terms of the right order in  $(T_c - T)$ , i.e.,  $\alpha(T) = \alpha_0(T_c - T)/T_c$ , while  $\beta$  and  $M$  are independent of  $T$ .

We rewrite the relations for the specific heat and supersolid fraction in a convenient form. For instance, we express the specific heat in terms of the number of mol,  $N/N_A$  such that  $R = N_A k_B = 8.31 \text{ J}/(\text{mol K})$ , and  $N_A$  is the Avogadro constant:

$$C_{\text{peak}} = R \left( \frac{\alpha_0}{\rho\beta} \right) \left( \frac{\alpha_0}{k_B T_c} \right). \quad (3)$$

Similarly, for the superfluid fraction one has

$$f^{ss} = \frac{m}{M} \left( \frac{\alpha_0}{\rho\beta} \right) \frac{T_c - T}{T_c}. \quad (4)$$

In expressions (3) and (4), the following dimensionless quantities appear naturally:

$$\Pi_1 = \frac{C_{\text{peak}}}{R} \Big|_{T=T_c} = \left( \frac{\alpha_0}{\rho\beta} \right) \left( \frac{\alpha_0}{k_B T_c} \right), \quad (5)$$

$$\Pi_2 = -T_c \frac{df^{ss}}{dT} \Big|_{T=T_c} = \left( \frac{m}{M} \right) \left( \frac{\alpha_0}{\rho\beta} \right), \quad (6)$$

$$\Pi_3 = \frac{\Pi_1}{\Pi_2} = \frac{C_{\text{peak}}/R}{-T_c df^{ss}/dT} \Big|_{T=T_c} = \left( \frac{M}{m} \right) \left( \frac{\alpha_0}{k_B T_c} \right), \quad (7)$$

which could be estimated via the experimental data of the specific heat and the superfluid density near the transition (notice that these relations are not independent). From various experimental data one has the values in Table I for the dimensionless relations.

From these data, one concludes that

$$-\frac{T_c}{f^{ss}(0)} \frac{df^{ss}}{dT} \Big|_{T=T_c} \approx 1.5 - 2,$$

therefore  $\Pi_2 \sim f^{ss}(0)$ . Moreover, it is tempting to conjecture that  $\Pi_1 \sim [f^{ss}(0)]^2$ , however there is no simultaneous measurement yet of nonclassical rotational inertia and heat capacity to conclude such a scaling law.

### A. Entropy of the supersolid part

The contribution of the superfluid part to the entropy excess increases linearly in temperature until the critical temperature  $T_c$ ; for  $T > T_c$  the superfluid entropy part keeps a constant value. This superfluid part of the entropy follows directly from the Landau free energy (1):

$$\Delta S = -\frac{\partial F}{\partial T} = V \frac{\alpha_0^2}{\beta} \frac{T}{T_c^2}.$$

The order of magnitude of the entropy excess per atom is

$$\frac{\Delta S_{\text{excess}}}{N} = k_B \left( \frac{\alpha_0}{\rho\beta} \right) \left( \frac{\alpha_0}{k_B T_c} \right) \approx 10^{-6} k_B,$$

which is consistent with the experimental data.<sup>25</sup> It should be noticed that this excess is over the usual phonon contribution in solids.

In Ref. 44, it is claimed that specific-heat measurements are contrary to entropy predictions based on a Bose-Einstein type of supersolidity, however, we may see that in our Ginzburg-Landau model there is no such problem.

TABLE I. A summary of possible values for the dimensionless parameters for various experimental data. The value of the dimensionless parameter  $\Pi_3$  is of the order of  $10^{-4}$ . The value of  $\Pi_2$  fluctuates around  $10^{-2}$  and  $10^{-4}$ . It is important to notice that  $\Pi_2/f^{ss}(0)$  is a number between 1 and 2. The value of  $\Pi_3$  in aerogel (\*) is obtained combining Refs. 42 and 43, and it is higher than in the case of bulk samples.

Reference	$\approx T_c$ (mK)	$\approx f^{ss}(T=0)$	$\Pi_1$	$\Pi_2$	$\Pi_3$	$\Pi_2/f^{ss}(0)$
25	80		$2.3 \times 10^{-6}$			
CP, 26	60		$6 \times 10^{-7}$			
BC2007, 26	100		$2.1 \times 10^{-6}$			
BC 1 ppb, 26	120		$5.6 \times 10^{-6}$			
27	130	$9 \times 10^{-3}$	$1.2 \times 10^{-6}$	$1.7 \times 10^{-2}$	$7 \times 10^{-5}$	1.88
27	130	$4 \times 10^{-3}$	$2 \times 10^{-6}$	$8 \times 10^{-3}$	$2.5 \times 10^{-4}$	2
27	300	$3.7 \times 10^{-3}$	$5 \times 10^{-6}$	$3.3 \times 10^{-3}$	$1.5 \times 10^{-3}$	0.9
AgCu, 300 ppb, 8	150	$1.45 \times 10^{-3}$		$2.1 \times 10^{-3}$		1.44
AgCu, 300 ppb, 8	115	$4 \times 10^{-4}$		$8 \times 10^{-4}$		2
BeCu, 1 ppb, 8	75	$1.35 \times 10^{-2}$		$2.1 \times 10^{-2}$		1.55
BeCu, 1 ppb, 8	65	$3.5 \times 10^{-3}$		$6 \times 10^{-3}$		1.7
10	160	$2.2 \times 10^{-2}$		$3 \times 10^{-2}$		1.5
39,40	150	$9 \times 10^{-4}$		$1.5 \times 10^{-3}$		1.66
16,41	56					1.3
Porous, 5	100	$5 \times 10^{-3}$		$10^{-2}$		2
Porous, 300 ppb, 42	100	$4 \times 10^{-4}$		$6.6 \times 10^{-3}$		1.65
Porous, 1–300 ppb, 43	220		$2.4 \times 10^{-5}$		$3.6 \times 10^{-3}$ (*)	
Porous, 300 ppm, 43	200		$2.7 \times 10^{-4}$			

### B. Coherence length

The coherence length is defined from the balance of the two first terms of the GL free energy:

$$\ell^2(T) \equiv \frac{\hbar^2}{M\alpha(T)},$$

expressing  $\alpha(T)$  in terms of the other constants, one gets

$$\ell(T) = \ell_0 \left( \frac{T_c - T}{T_c} \right)^{-1/2}, \text{ with}$$

$$\ell_0 = \lambda_c \sqrt{\frac{m}{M} \frac{k_B T_c}{\alpha_0}} = \lambda_c \sqrt{\Pi_2 / \Pi_1} = \frac{\lambda_c}{\sqrt{\Pi_3}}, \quad (8)$$

and the constant  $\lambda_c = \frac{\hbar}{\sqrt{mk_B T_c}}$  is the de Broglie thermal wavelength of helium at the transition temperature, which is about  $\lambda_c \approx 1.2$  nm at  $T_c \approx 0.08$  K (in the estimations throughout the paper we shall consider  $T_c \approx 0.08$  K,  $k_B T_c \approx 1.1 \times 10^{-24}$  J,  $m\rho = 194$  kg/m<sup>3</sup>,  $m = 6.52 \times 10^{-27}$  kg,  $\hbar/m = 1.58 \times 10^{-8}$  m<sup>2</sup>/s). Finally, using the data collected in Table I, one obtains that the healing length is approximately

$$\ell_0 \approx (40 - 110) \text{ nm},$$

which is consistent with the estimations done by Kubota and collaborators.<sup>40</sup> Therefore  $\ell_0$  is large enough to be interpreted as a macroscopic quantity, which at least does not invalidate a macroscopical approach. In the case of an aerogel this coherence length would be of the order of the typical separation between the silica strands, however the value of  $\Pi_3$  appears as large as  $\Pi_3 \approx 0.0036$  (see Table I), so that  $\ell_0 \approx 20$  nm, thus superfluidity may manifest over distances of the order of 100 nm.

In a more general way, one shall expect that the following length plays the role of a healing length at any temperature:

$$\ell(T) = \frac{\hbar}{\sqrt{mk_B T}} \sqrt{\frac{-T df^{ss}/dT}{C_{\text{peak}}/R}}.$$

This model conjectures the existence of topological vortices in a supersolid because of the existence of a coherent long-range-order phase in the system. As in superfluids, “supersolid vortices” are stationary solutions of Eq. (2) with a  $\pm 2\pi$  phase jump around the vortex core.<sup>38</sup> Vortices cannot be removed by any infinitesimal perturbation of the order parameter, since there are topological defects. Vortices maybe generated in various ways, the most common is via the mechanism of critical velocity.

Similarly persistent currents are also present in the frame of our model. A persistent current maybe easily obtained in a supersolid if it is in a multiconnected domain. Indeed, imposing a phase jump of  $2\pi$  as one turns around the hole in the multiconnected domain we assure the existence of a nonuniform phase as in the case of a vortex. Vortices and persistent currents are a nonambiguous property that would confirm the existence of a supersolid as a coherent state. Up to date, there is no direct experimental evidence of such behaviors.

### C. Supersolid–ordinary solid interface energy

The superfluid bulk-free energy density is  $f(T) = -f_0 \left( \frac{T_c - T}{T_c} \right)^2$ , with  $f_0 = \frac{\alpha_0^2}{2\beta} = \frac{k_B T_c \rho}{2} \left( \frac{\alpha_0}{\rho\beta} \right) \left( \frac{\alpha_0}{k_B T_c} \right) = \rho \frac{k_B T_c}{2} \Pi_1 \approx (0.02 \text{ up to } 0.08) \text{ Pa}$ . Next, as in the original Ginzburg-Landau work, we shall consider the existence of an interface composed of an ordinary solid ( $\psi = 0$ ) and supersolid state [ $\psi_{ss} = \sqrt{\alpha(T)/\beta(T)}$ ]. Notice that this interface is imposed by a boundary or by an object. Thus the energy (per unit surface)

that we shall compute represents the energy deficit because of the existence of this boundary. For simplicity, the interface is settled at  $x = 0$ , an equilibrium planar interface follows from Eq. (2), with the adequate boundary conditions. The interface solution is  $\psi_I(x) = \sqrt{\alpha(T)/\beta(T)} \tanh(x/\ell)$ . The surface energy is the energy difference between the energy of the interface solution  $\psi_I$  and the homogeneous solution  $\psi_{ss}$ , that is

$$\begin{aligned} \sigma_{ss} &= -\frac{4}{3} f(T) \ell(T) \\ &= \frac{2}{3} k_B T_c (\rho \ell_0) \left( \frac{\alpha_0}{\rho \beta} \right) \left( \frac{\alpha_0}{k_B T_c} \right) \left( \frac{T_c - T}{T_c} \right)^{3/2}. \end{aligned}$$

With the above estimations, one has [ $\frac{2}{3} k_B T_c (\rho \ell_0) \approx 2.6 \times 10^{-5} \text{ kg/s}^2$ ]

$$\sigma_{ss} = 2 \times 10^{-9} \text{ kg/s}^2 \left( \frac{T_c - T}{T_c} \right)^{3/2},$$

that is at least  $10^{-5}$  times smaller than the solid-superfluid helium surface energy. This small factor is not surprising because the only quantity with an energy scale in the theory is the specific heat, which is a millionth smaller than in superfluid helium.

A consequence of the existence of surface energy is a delay of the ordinary solid-supersolid transition. Indeed if the solid is inside of a small volume  $\Omega$  then the gradient term in the GL free energy increases significantly, so that the quadratic term of the free energy compensates the negative contribution of the  $\alpha|\psi|^2$  term. The ordinary solid state is therefore stable. More precisely, the supersolid phase exists if  $T/T_c \leq 1 - \ell_0^2 \lambda_0$  where  $\lambda_0$  is the lowest eigenvalue (which has dimensions of the inverse of an area  $\sim \Omega^{-2/3}$ ) of the linear equation  $-\nabla^2 u = \lambda u$  with  $u = 0$  on  $\partial\Omega$ . So if the geometry and porosity of the domain provide a  $\lambda_0 > 1/\ell_0^2$  then the supersolid transition may never occur even at zero temperature. The value of the healing length of the order of 100 nm prevents supersolidity in domains smaller (in all directions) than 100 nm. However, in the case of solid helium in a porous media, the healing length becomes as small as 20 nm so that supersolidity may manifest.

### III. SUPERSOLID AND ELASTIC COUPLING AT EQUILIBRIUM

The free energy (1) does not consider the existence of a lattice nor its elasticity. We shall therefore extend this Ginzburg-Landau approach coupling the superfluid part with the lattice. Last, at some point this theory should also match the Andreev-Lifshitz equations.<sup>2,33,45,46</sup> The thermodynamical variables will be the number density  $\rho$  of solid helium, the complex order parameter  $\psi$ , the elastic deformation  $\mathbf{u}$ , and the entropy per unit volume  $s$ .

To derive the set of equations we use the standard approach of the Landau two fluid model, namely the conservation laws, and we impose the Galilean invariance. We shall go step by step to sketch well the different aspects of the proposed macroscopic equations.

The total (that is, the superfluid plus the ordinary solid part) energy and momentum densities are

$$\mathcal{E} = \frac{\hbar^2}{2M} |\nabla\psi|^2 + \frac{m}{2} (\rho - \rho^{ss}) \dot{\mathbf{u}}^2 + \mathcal{E}_0(|\psi|^2, \rho, s, u_{ik}), \quad (9)$$

$$\mathbf{j} = m \left( -\frac{i\hbar}{2M} (\psi^* \nabla\psi - \psi \nabla\psi^*) + (\rho - \rho^{ss}) \dot{\mathbf{u}} \right). \quad (10)$$

The kinetic terms in Eq. (9) are of the form  $\frac{1}{2} m \rho^{ss} \mathbf{v}^2 + \frac{m}{2} (\rho - \rho^{ss}) \dot{\mathbf{u}}^2$ , while the momentum (10) takes the usual expression:  $\mathbf{j} = m \rho^{ss} \mathbf{v}^s + m (\rho - \rho^{ss}) \dot{\mathbf{u}}_k$ . These quantities come from the Galilean invariance. Note that  $\rho^{ss} = (m/M) |\psi|^2$  is the supersolid number density (4). Here  $\mathcal{E}_0(|\psi|^2, \rho, s, u_{ik})$  is the internal energy of the body that depends explicitly on the order parameter  $|\psi|^2$  (the Landau expansion of previous section), the number density  $\rho$ , the entropy density  $s$ , and the elastic strain  $u_{ik} = (\partial_i u_k + \partial_k u_i)/2$ . We shall come back to it later.

The equilibrium solution is obtained minimizing the total energy under the constrain of a constant momentum. Using the Lagrange multiplier technique varying  $\int [\mathcal{E} - \lambda(\mathbf{x}, t) \cdot \mathbf{j}] d^D \mathbf{x}$  with respect to  $\psi$  and  $\dot{\mathbf{u}}$ , one has that the equilibrium equation coupled to the lattice motion is [it comes out that  $\lambda(\mathbf{x}, t) = \dot{\mathbf{u}}$ ]<sup>47</sup>

$$\frac{1}{2M} (-i\hbar \nabla - m \dot{\mathbf{u}})^2 \psi + \frac{\partial \mathcal{E}_0}{\partial |\psi|^2} \psi = 0,$$

where the derivative in the last term in Eq. (11) is formally at constant density mass, entropy, and strain, that is,  $\frac{\partial \mathcal{E}_0}{\partial |\psi|^2} |_{\rho, s, u_{ik}}$  and this term should be understood as  $[\alpha(T) - \beta(T) |\psi|^2] \psi$ , thus

$$\frac{1}{2M} (-i\hbar \nabla - m \dot{\mathbf{u}})^2 \psi - (\alpha - \beta |\psi|^2) \psi = 0. \quad (11)$$

Equation (11) replaces Eq. (2). A uniform rotation follows easily by taking  $\dot{\mathbf{u}} = \boldsymbol{\omega} \times \mathbf{r}$  and solving Eq. (11). This is the well-known problem of a superconductor under an external uniform magnetic flux. The equilibrium state is  $\psi_{\text{flow}} = \sqrt{\alpha(T)/\beta(T)} e^{i\phi}$  where  $\phi$  satisfies  $\nabla^2 \phi = 0$  inside the domain or container and  $\hat{\mathbf{n}} \cdot (\hbar \nabla \phi - m \boldsymbol{\omega} \times \mathbf{r}) = 0$  in the boundaries, that is, the usual problem of a rotating perfect fluid. This presents a nonclassical rotational behavior that depends on the geometry of the container, as in the ‘‘blocked annulus experiment,’’<sup>6,9</sup> and more importantly the supersolid density is exactly  $\rho^{ss}(T)$  defined by Eq. (4). In the case of a rigid rotation, Eq. (11) becomes the usual Ginzburg-Landau equation for superconductivity in the presence of a uniform magnetic field. Moreover, as the steady rotation  $\omega$  increases, vortices appear as  $\omega > \omega_{c_1}$ , then more and more vortices are nucleated at regular intervals  $\Delta\omega = \frac{\hbar}{mS}$  ( $S$  is the effective surface of the container). Finally, NCRI decreases ultimately to zero as  $\omega > \omega_{c_2} = \frac{\hbar}{m\ell(T)^2}$ , a frequency that is of the order of  $\omega_{c_2} = \frac{\hbar}{m\ell_0^2} \approx 1.5 \times 10^6 \text{ s}^{-1}$ , that is, a frequency of the order of  $f_{c_2} = 250 \text{ kHz}$ . This view is consistent with experiments in steady rotation by Kim and collaborators.<sup>10</sup>

#### A. Macroscopic equations for a supersolid at $T \neq 0$

Next we shall write the dynamical equations for  $\psi$ ,  $\rho$ ,  $\mathbf{u}$ , and  $s$ . A simple count indicates that the present approach presents two extra variables than our previous model at  $T = 0$ .<sup>33</sup> One of these new variables is the entropy, absent



at  $T = 0$ . The other variable is the modulus of the order parameter, that is, the superfluid density  $\varrho^{ss}$ . In our previous macroscopical equations,<sup>33</sup> the superfluid or supersolid density was a parameter of the problem. Notice finally that the supersolid density is also a parameter in the Andreev-Lifshitz model.<sup>2</sup> In the present context the supersolid density evolves in time and, indeed, it is fixed by the thermodynamical equilibrium (11).

We shall write an equation for  $\psi$  that should relax to equilibrium after long times to Eq. (11) and, more importantly, it should be Galilean invariant. This phenomenological approach was undertaken by Pitaevskii in 1958.<sup>47</sup> Pitaevskii considers

$$i\hbar \frac{\partial \psi}{\partial t} = -\frac{\hbar^2}{2m} \nabla^2 \psi + \varphi \psi - i\Lambda \times \left( \frac{1}{2M} (-i\hbar \nabla - m\dot{\mathbf{u}})^2 \psi - (\alpha - \beta |\psi|^2) \psi \right), \quad (12)$$

where the first term (with  $m$  the atomic mass of helium and not  $M$ !) is responsible for a Galilean invariance, and  $\varphi$  is a potential energy that should be determined. The parameter  $\Lambda$  is a dimensionless quantity that measures the rate of relaxation to equilibrium. This parameter  $\Lambda$  will be considered as a real number.<sup>48</sup> Finally, we keep, for generality, the  $\frac{\partial \mathcal{E}_0}{\partial |\psi|^2}$  term, instead of the usual Ginzburg-Landau expansion  $-(\alpha - \beta |\psi|^2)$ .

For the other pertinent variables we write the conservation equation for the total mass density, momentum, and entropy:<sup>2,47</sup>

$$\partial_t \rho + \nabla \cdot (\mathbf{j}/m) = 0, \quad (13)$$

$$\partial_t j_i + \partial_k T_{ik} = 0, \quad (14)$$

$$\partial_t s + \nabla \cdot (s\dot{\mathbf{u}} - \mathbf{q}) = R \geq 0. \quad (15)$$

These three equations are the usual conservation laws of the number of particles, the momentum, and the entropy<sup>2</sup> [note that this is the total entropy of the solid and not just the superfluid excess coming from  $F_{ss}(T)$ ]. Finally the potential energy  $\varphi$ , the energy momentum tensor  $T_{ik}$ , the heat flux  $\mathbf{q}$ , and the positive entropy rate  $R$  are chosen by imposing the conservation of the total energy (9):  $\partial_t \mathcal{E} + \nabla \cdot \mathbf{Q} \equiv 0$ . Taking the time derivative of Eq. (9), after a long, but straightforward, calculation (details are presented in the Supplemental Material<sup>49</sup>), one obtains

$$\begin{aligned} & \frac{\partial}{\partial t} \mathcal{E} + \partial_i Q_i \\ &= \left( R - \mathbf{q} \cdot \nabla T - \frac{2\Lambda}{\hbar} |\hat{B}\psi|^2 \right) \\ &+ \dot{\mathbf{u}} \cdot \left[ (\rho - \varrho^{ss}) \nabla \left( \frac{\partial \mathcal{E}_0}{\partial \rho} \right) + s \nabla T - \nabla p + \varrho^{ss} \nabla \varphi \right] \\ &+ \frac{1}{m} (\nabla \cdot \mathbf{j}^s) \left( \varphi - \frac{\partial \mathcal{E}_0}{\partial \rho} - \frac{M}{m} \frac{\partial \mathcal{E}_0}{\partial |\psi|^2} \right). \end{aligned} \quad (16)$$

To write Eq. (16), we have explicitly defined

$$\begin{aligned} T_{ik} &= \frac{\hbar^2}{4M} (\partial_i \psi \partial_k \psi^* - \psi^* \partial_{ik} \psi + \text{c.c.}) \\ &+ m(\rho - \varrho^{ss}) \dot{u}_i \dot{u}_k - p \delta_{ik} - \frac{\partial \mathcal{E}_0}{\partial u_{ik}}, \end{aligned} \quad (17)$$

$$\begin{aligned} Q_i &= -T q_i + \frac{1}{2M} [(i\hbar \partial_i \psi^* \hat{A} \psi + \Lambda \hbar \partial_i \psi^* \hat{B} \psi + \text{c.c.})] \\ &+ \left[ \left( \frac{1}{2} m(\rho - \varrho^{ss}) \dot{\mathbf{u}}^2 + T s + (\rho - \varrho^{ss}) \frac{\partial \mathcal{E}_0}{\partial \rho} \right. \right. \\ &\left. \left. + \frac{\Lambda \hbar}{2M} [\nabla \cdot (\mathbf{j}^s - m \varrho^{ss} \dot{\mathbf{u}})] \right) \delta_{ik} - \frac{\partial \mathcal{E}_0}{\partial u_{ik}} \right] \dot{u}_k. \end{aligned} \quad (18)$$

The shape of the stress tensor above Eq. (17) follows directly from the theory of superfluids plus an elasticity contribution  $-\frac{\partial \mathcal{E}_0}{\partial u_{ik}}$ , which follows from the ordinary elasticity. Finally, the expression for  $\hat{A}$  and  $\hat{B}$  are given by (see details in the Supplemental Material<sup>49</sup>)

$$\hat{A} = -\frac{\hbar^2}{2m} \nabla^2 + \varphi \quad \text{and} \quad (19)$$

$$\hat{B} = \frac{1}{2M} (-i\hbar \nabla - m\dot{\mathbf{u}})^2 + \frac{\partial \mathcal{E}_0}{\partial |\psi|^2}. \quad (20)$$

The requirement of conservation of energy imposes restrictions on the auxiliary fields  $\varphi$ ,  $p$ , and the entropy production  $R$ ,

$$\nabla p = s \nabla T + (\rho - \varrho^{ss}) \nabla \frac{\partial \mathcal{E}_0}{\partial \rho} + \varrho^{ss} \nabla \varphi, \quad (21)$$

$$\varphi = \left( \frac{\partial \mathcal{E}_0}{\partial \rho} + \frac{M}{m} \frac{\partial \mathcal{E}_0}{\partial |\psi|^2} \right), \quad (22)$$

$$R = \mathbf{q} \cdot \nabla T + \frac{2\Lambda}{\hbar} |\hat{B}\psi|^2 \geq 0, \quad (23)$$

$$\mathbf{q} = \kappa \nabla T. \quad (24)$$

Here Eq. (21) is a Gibbs-Duhem relation, thus one defines the chemical potential by  $\mu = \frac{\partial \mathcal{E}_0}{\partial \rho}$ . Finally, Fourier's law (24) follows from the positiveness of entropy production (23), and we notice that the equilibrium holds for a uniform temperature on the sample and by having  $\hat{B}\psi = 0$ , that is, if  $\psi$  satisfies Eq. (11), which is consistent with the original assumptions.

Finally, let us point out that for practical purposes one expands the energy in power of the order parameter and of the strain,

$$\begin{aligned} \mathcal{E}_0(\rho, |\psi|^2, s, u_{ik}) &= e_0(\rho) - \alpha(T) |\psi|^2 + \frac{\beta(T)}{2} |\psi|^4 \\ &+ a u_{ii} |\psi|^2 + \frac{1}{2} (K - 2\mu_s/3) u_{ii}^2 \\ &+ \frac{1}{2} \mu_s u_{ik} u_{ki} + \dots, \end{aligned} \quad (25)$$

where  $e_0(\rho)$  is an internal energy,  $K$  and  $\mu_s$  are the bulk and shear modulus, respectively, the term  $a u_{ii} |\psi|^2$  is of the same order as  $(T_c - T)^2$ , and it has been considered by Ref. 34 that this term would be finally responsible for a shear modulus stiffening as the supersolid phase takes place ( $T < T_c$ ).

## B. Sound

We shall consider briefly the oscillations around the equilibrium state, characterized by uniform values of the density  $\rho$ , entropy  $s$ , superfluid density  $\varrho^{ss}$ , and phase  $\phi$ . Moreover, there is no strain  $u_{ik} = 0$  as well as no lattice displacements  $\dot{\mathbf{u}} = 0$ . The perturbation scheme is more subtle than expected because the total density has two types of

variations: one is the usual change of density in ordinary solids, that is, because of a change of the strain  $u_{ik}$ , we denote this by  $\rho_{cl}(u_{ik})$ ; the second change comes because of the long-range coherent superfluid behavior. This second change is responsible for phase coherence and ultimately of the existence of a supersolid density. We consider  $\rho = \rho_{cl}(u_{ik}) + \delta\rho$ , then one notices that  $\partial_t \rho_{cl} + \nabla \cdot (\rho_{cl} \dot{\mathbf{u}}) \approx (\partial \rho_{cl} / \partial u_{ik} + \rho_{cl} \delta_{ik}) \partial_t u_{ik} \approx 0$  because  $\rho_{cl}(T, u_{ik})$  behaves as an ordinary or classical solid.<sup>2</sup> Thus in Eq. (13) only nonordinary variation of mass in helium survives,

$$\partial_t \delta\rho + \nabla \cdot [\rho^{ss}(v^s - \dot{\mathbf{u}})] = 0,$$

where  $\delta\rho$  is the variation of the number of particles per unit volume.

Following the usual linear theory of sound modes, one expresses the variables as exponentials  $\sim e^{i(k \cdot x - \omega t)}$ , and one derives equations for the four linear normal modes of oscillations, as in the Andreev-Lifshitz model.<sup>2,50</sup> These modes are (up to linear order in  $f^{ss}$ ) as follows:

(i) Two shear modes (decoupled up to linear order in an infinite space) with a dispersion relation,

$$\omega_k = c_s(1 + f^{ss}/2)k,$$

where  $c_s^2 = \mu_s/(m\rho)$  is the usual shear waves' sound speed; the factor  $(1 + f^{ss}/2)$  is because the effective inertia is less than the total solid. The effective sound speed of shear waves  $\sqrt{\mu_s/(m\rho)}(1 + f^{ss}/2)$  presents an increase because of the shear modulus stiffness<sup>21</sup> and because of the factor  $(1 + f^{ss}/2)$  too. The former effect is about 8%, while the second is in general less than 1%. Independent measurements of shear modulus<sup>22</sup> and sound speed<sup>23</sup> do not agree with the present result. Despite the great accuracy there is no evidence of a superfluid behavior through sound speed measurements.

(ii) One compression mode, which in the zero-temperature limit reduces to

$$\omega_k = k c_K \left( 1 + \frac{f^{ss}}{2} + \frac{f^{ss}}{2} \frac{(\rho \partial^2 \mathcal{E}_0 / \partial \rho^2)}{m c_K^2} \right) - i \Lambda f^{ss} \frac{\hbar k^2}{4M} \left( 1 + \frac{(\partial \mathcal{E}_0 / \partial \rho + \rho \partial^2 \mathcal{E}_0 / \partial \rho^2)}{m c_K^2} \right),$$

where  $c_K^2 = (K + 4\mu_s/3)/(m\rho)$  is the ordinary velocity of sound of the compression mode,  $K$  is the bulk modulus of solid helium. The second term represents a damping, which may fix the value of the dimensionless parameter  $\Lambda \frac{m}{M}$ .

(iii) Last, a superfluid mode, also known as the fourth sound.<sup>50</sup> As in previous work<sup>2,33,50</sup> a new propagating mode appears besides the usual longitudinal and transverse modes in regular crystals. The speed of propagation of this mode is smaller than the usual elastic sound waves' speed; indeed the dispersion relation of the superfluid mode behaves as

$$\omega_k = c_B \sqrt{f^{ss}} k + i \Lambda \frac{\hbar}{4M} k^2, \quad (26)$$

with

$$c_B^2 = \frac{\rho}{m} \left( \mu'(\rho) + \beta \frac{M^2}{m^2} \right), \quad (27)$$

where  $\mu(\rho) = \frac{\partial \epsilon_0}{\partial \rho}$  is the chemical potential. This slow mode is a signal of a modulation of the coherent quantum phase. Although there is no conclusive evidence; in Ref. 51 a slow mode was perhaps observed. Among the two contributions in  $c_B^2$ , Eq. (27), the Bogoliubov one,  $\rho \mu'(\rho)/m$ , is of the order of the bulk sound speed of solid helium, therefore is definitively much larger than the second one, the latter being a speed of the order of  $\sqrt{\frac{k_B T_c}{m} \Pi_1 / \Pi_2^2}$ , that is, a few meters/second with the above estimates in Table I. The final speed of the fourth mode is of the order of  $c_B \sqrt{f^{ss}}$ .

### C. Critical speed for vortex nucleation

In the frame of the Ginzburg-Landau equation (12) a critical speed for vortex nucleation appears as an instability of a uniform relative flow between the lattice and the superflow. We shall perturb the homogeneous solution  $\psi = \sqrt{\alpha/\beta}$  of Eq. (12). To do this, it is useful to transform Eq. (12) into an amplitude-phase dynamics. Writing  $\psi = R e^{i\phi}$  one may split Eq. (12) in the real and imaginary parts:

$$\partial_t R^2 + \nabla \cdot \left( R^2 \frac{\hbar}{m} \nabla \phi \right) = \frac{2\Lambda}{\hbar} \left[ \frac{\hbar^2}{2M} \nabla^2 R + \alpha R - \beta R^3 - \frac{m^2}{2M} \left( \frac{\hbar}{m} \nabla \phi - \dot{\mathbf{u}} \right)^2 R \right] R \quad (28)$$

$$\partial_t \phi = \frac{\hbar}{2m} \left( \frac{1}{R} \nabla^2 R - (\nabla \phi)^2 \right) - \frac{\varphi}{\hbar} + \frac{\Lambda m}{2MR^2} \nabla \cdot \left[ R^2 \left( \frac{\hbar}{m} \nabla \phi - \dot{\mathbf{u}} \right) \right]. \quad (29)$$

Because of the dissipative term, proportional to  $\frac{2\Lambda\alpha}{\hbar}$  in Eq. (28), the superfluid density is settled by the fast dissipative dynamics, thus the amplitude  $R$  follows simply the dynamics of the superfluid coherent phase  $\phi$ . Neglecting  $\partial_t R$  in Eq. (28), one can obtain this dependence with an asymptotic expansion in small gradients of  $\nabla \phi$  and small elastic displacements  $\dot{\mathbf{u}}$ ; one obtains up to first nontrivial order

$$R^2 = \frac{1}{\beta} \left[ \alpha - \frac{m^2}{2M} \left( \frac{\hbar}{m} \nabla \phi - \dot{\mathbf{u}} \right)^2 - \frac{\hbar^2}{2m\Lambda} \nabla^2 \phi \right].$$

Introducing this expression for the dynamical behavior of the superfluid phase (29), and then expanding, re-arranging and keeping the lowest order, one obtains that the phase equation rules,

$$\begin{aligned} \partial_t \phi = & \frac{\hbar}{2m} \left( \frac{\Lambda m}{M} + \frac{M}{\Lambda m} \right) \nabla^2 \phi - \frac{\hbar}{2m} (\nabla \phi)^2 + \frac{m}{2\hbar} A^2 \\ & - \frac{\Lambda m^3}{2M^2} \frac{A_i A_j}{\alpha - m^2 A^2 / 2M} \partial_i A_j - \frac{m}{2\Lambda \alpha} \left( \frac{\hbar}{m} \partial_i \phi \right) \\ & \times A_j \partial_i A_j - \mu(\rho) / \hbar, \end{aligned} \quad (30)$$

where we have defined the relative superfluid-lattice speed,

$$A_i = \frac{\hbar}{m} \nabla_i \phi - \dot{u}_i.$$

If the crystal is moving with a constant speed  $v$  along the  $x$  axis ( $\dot{u}_x = v$ ) one gets the linear dynamics for the phase perturbation:

$$\partial_t \phi = \frac{\hbar}{2m} \left( \frac{\Lambda m}{M} + \frac{M}{\Lambda m} \right) \nabla^2 \phi - \frac{\hbar}{2m} \frac{\Lambda m^2}{M^2} \frac{v^2}{\alpha - m^2 v^2 / 2M} \partial_{xx} \phi + \text{h.o.t.} \quad (31)$$

The condition for the stability of the superflow is that the prefactor of  $\partial_{xx} \phi$  term must be positive, so that

$$\left( \frac{\Lambda m}{M} + \frac{M}{\Lambda m} \right) - \frac{\Lambda m^3}{M^2} \frac{v^2}{\alpha - m^2 v^2 / 2M} > 0,$$

thus a stable superflow holds if

$$v^2 < \frac{2M\alpha}{m^2} \frac{1 + \left(\frac{\Lambda m}{M}\right)^2}{1 + \frac{3}{2} \left(\frac{\Lambda m}{M}\right)^2}.$$

Because the parameter  $\sqrt{\frac{1 + (\Lambda m/M)^2}{1 + (3/2)(\Lambda m/M)^2}}$  is a number between  $\sqrt{2/3} \approx 0.8$  and 1, thus the relevant value for the critical speed is of the order of

$$v_c = \sqrt{\frac{2M\alpha}{m^2}} = \sqrt{\frac{\alpha_0}{k_B T_c} \frac{M}{m}} \sqrt{\frac{2k_B T_c}{m}} = \sqrt{\Pi_3} \sqrt{\frac{2k_B T_c}{m}}.$$

Because  $\sqrt{\frac{2k_B T_c}{m}} \approx 12$  m/s, and according to previous values of the dimensionless quantities one has that  $0.1$  m/s  $\lesssim v_c \lesssim 0.6$  m/s, in close agreement with the experimental value.<sup>40</sup>

If  $v < v_c$  a stable superflow is possible, however, if  $v > v_c$  a vortex behavior dominates the dynamics. Naturally thermal fluctuations may change the threshold for vortex nucleation.

We conclude with the following remark: Taking gradient of Eq. (30) one gets a Navier-Stokes equation for the superfluid velocity, the kinematic viscosity being  $\frac{\hbar}{2m} \left( \frac{\Lambda m}{M} + \frac{M}{\Lambda m} \right)$ , a number proportional to  $\hbar/m$ .

#### IV. DISCUSSION

Supersolidity is viewed as a complex system that displays together ordinary elasticity and superfluidity. *Ab initio* superfluidity is understood in the frame of a Ginzburg-Landau theory near the transition temperature. Ultimately, the whole theory requires a nontrivial coupling of elastic and superfluid motion. The free parameters of the theory may be estimated from the current experimental data of nonclassical rotational inertia fraction and heat capacity. The theory predicts a

macroscopic coherence length, which would be of the order of 50–100 nm (in bulk experiments), and a critical speed of the order of 10–60 cm/s, both predictions in agreement with the expected values.<sup>39,40</sup> Moreover, the theory predicts a slow Bogoliubov-like superfluid sound mode with a propagation speed that scales as the square root of the superfluid fraction,  $\sqrt{f_{ss}}$ , and may be estimated between 4 and 40 m/s for the current data of the observed superfluid fraction.

Although the theory cannot predict a value for the dimensionless parameter  $\frac{m}{M}$ , because of the extra intrinsic free parameter, which is the order parameter normalization, the theory predicts the right values of thermodynamical quantities as entropy, or surface energy without any supplementary assumption. In conclusion, the present theory provides a unified view of three different behaviors of supersolidity supported by experiments: NCRI, anomaly of heat capacity, and the ordinary mechanical behavior under external stress.

Finally, we emphasize that there are many open questions, which are not contemplated in the present theory, that deserve some attention. For instance, *why is the superfluid or supersolid fraction  $f_{ss}$  so small?* Contrary to superfluid liquid helium, and superfluids in general, the zero-temperature limit gives a supersolid fraction, between 20% and 0.02%, while in superfluids the superfluid fraction is always 100%. Leggett, in a series of papers,<sup>4</sup> indicates that the ground-state wave function of crystalline structure provides a natural lower value for the superfluid fraction at  $T = 0$  K. Presumably, the value of the superfluid density is of microscopic origin and may depend on the crystallography of the solid. This drives us to a more important question: *Why does the supersolid fraction vary by three orders of magnitude from one sample to another?* It has been noticed that the superfluid density depends strongly on the sample preparation, that is, experimental procedure and the cell material. There is abundant evidence that crystalline defects, like grain boundaries, dislocations, vacancies, interstitially enhance supersolidity, however a method to control it precisely is not known. Given that at present, we do not possess a rational theory that predicts the superfluid fraction, the proposed theory uses the superfluid density as a given parameter.

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