Squeezing Superfluid from a Stone: Coupling Superfluidity and Elasticity in a Supersolid

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Starting from the assumption that the normal solid to supersolid (NS-SS) phase transition is continuous, we develop a phenomenological Landau theory of the transition in which superfluidity is coupled to the elasticity of the crystalline ^{4}He lattice. We find that the elasticity does not affect the universal properties of the superfluid transition, so that in an unstressed crystal the well-known λ anomaly in the heat capacity of the superfluid transition should also appear at the NS-SS transition. We also find that the onset of supersolidity leads to anomalies in the elastic moduli and thermal expansion coefficients near the transition and, conversely, that inhomogeneous lattice strains can induce local variations of the superfluid transition temperature, leading to a broadened transition.

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Superfluidity—the ability of *liquid* ^{4}He, when cooled below 2.176 K, to flow without resistance [1,2] through narrow pores—has long served as a paradigm for the phenomenon of ''off-diagonal long-range order'' (ODLRO) in quantum liquids and superconductors [3]. Supersolidity—the coexistence of ODLRO with the crystalline order of a solid—was proposed theoretically [4–12] as an even more exotic phase of *solid* ^{4}He, but it has eluded detection [13,14]. Recently, Kim and Chan [15,16] have reported the onset of ''nonclassical rotational inertia'' [6] in a torsional oscillator experiment with solid ^{4}He, and they interpret their results as indicating the onset of supersolidity. However, their interpretation remains controversial [17–22], so it is important to complement the nonequilibrium torsional oscillator measurements with *equilibrium* thermodynamic measurements, e.g., of the specific heat. In this work we start from the assumption that normal solid to supersolid (NS-SS) phase transition is continuous, and develop a phenomenological Landau theory of the transition in which superfluidity is coupled to the elasticity of the crystalline ^{4}He lattice. We find that the elasticity does not affect the universal properties of the superfluid transition, so that in an unstressed crystal the well-known λ anomaly in the heat capacity of the superfluid transition should also appear at the NS-SS transition. We also find that the onset of supersolidity leads to anomalies in the elastic moduli and thermal expansion coefficients near the transition; conversely, inhomogeneous strains in the lattice can induce local variations of the superfluid transition temperature, leading to a broadened transition. As our theory is rooted in a few simple assumptions and symmetry principles, we expect our results to be robust and insensitive to the details of a microscopic model for the supersolidity.

We hypothesize that, as with the *superfluid* ^{4}He, the appropriate order parameter describing the onset of supersolidity is a complex scalar field $\psi(\mathbf{r})$, depending on the location $\mathbf{r}$. We shall assume that the phase transition to the supersolid state is continuous, as is the *superfluid* ^{4}He transition. Then, as is well known from the theory of critical phenomena [23], the *universal* properties of the supersolid transition may be obtained via a model that retains only those terms in the free energy up to leading (relevant) order in powers of the fields and their spatial gradients, resulting in the Landau form:

$$\mathcal{F}_s = \int d^3r \left\{ \frac{1}{2} c_{\alpha\beta} \partial_\alpha \psi \partial_\beta \psi^* + \frac{1}{2} a(T) |\psi|^2 + \frac{1}{4!} w |\psi|^4 \right\}. \tag{1}$$

Here, $a(T)$ depends smoothly on the temperature T; it is negative at low temperatures, changing sign slightly above the transition temperature T_c. [If fluctuations of ψ are ignored, T_c would be exactly the temperature at which $a(T)$ changes sign.] For $T < T_c$ ($T > T_c$) the thermal expectation value of $\langle\psi\rangle$ is nonzero (zero). The constant w measures the strength of the nonlinearity, and the symmetric tensor $c_{\alpha\beta}$ characterizes the spatial anisotropy inherited from the crystallinity of the normal solid. For an isotropic superfluid or for a cubic crystal $c_{\alpha\beta} = c\delta_{\alpha\beta}$, with c a constant, whereas for an hcp crystal (such as solid helium [24]) $c_{\alpha\beta}$ is uniaxial, such that $c_{\alpha\beta} = c_z n_\alpha n_\beta + c_\perp(\delta_{\alpha\beta} - n_\alpha n_\beta)$, with $\mathbf{n}$ a unit vector that points along the preferred axis of the crystal, and c_z and $c_\perp$ independent constants. It is important to note that the symmetry of the superfluid density tensor $\rho^S_{\alpha\beta}$ (which relates the superfluid velocity to the momentum density) is the same as that of $c_{\alpha\beta}$.

If we were dealing with the normal-to-*superfluid* transition, Eq. (1) (with $c_{\alpha\beta} = c\delta_{\alpha\beta}$) would be the entire story. However, because we are dealing with the normal-to-*supersolid* transition, it is not: when formulating theories of continuous phase transitions, it is necessary to keep *all* of the degrees of freedom that are soft (i.e., exhibit large thermal fluctuations) at the transition [23]. For a superfluid, ψ *is* the only such degree of freedom. However, in a supersolid there are additional phonon degrees of freedom

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that are soft not only at the transition but throughout both the normal and supersolid phases. These are associated with displacements $\mathbf{u}(\mathbf{r})$ of the positions $\mathbf{r}$ of the undistorted crystal lattice. (The normal solidification of the ^{4}He occurs at a temperature substantially higher than T_c so that amplitude fluctuations of the density waves are not soft and may be neglected.)

As the free energy must be invariant under spatial translations and rotations [25], it can only depend upon $\mathbf{u}(\mathbf{r})$ through the symmetric strain tensor $u_{\alpha\beta} \equiv \frac{1}{2} \times (\partial_\alpha u_\beta + \partial_\beta u_\alpha + \partial_\alpha u_\gamma \partial_\beta u_\gamma)$. Thus, at long wavelengths the relevant terms in the free energy involving $u_{\alpha\beta}$ alone are simply those of standard elastic theory: $\mathcal{F}_e = \frac{1}{2} \times \int d^3 r \lambda_{\alpha\beta\gamma\delta} u_{\alpha\beta} u_{\gamma\delta}$, where $\lambda_{\alpha\beta\gamma\delta}$ are the bare elastic constants, and repeated indices are summed over. The form of $\lambda_{\alpha\beta\gamma\delta}$ is dictated by the symmetry of the crystal; for an hcp crystal such as ^{4}He, it is parametrized by 5 independent elastic constants [25].

To determine the form of the coupling between the supersolid order parameter ψ and the local displacement field $\mathbf{u}(\mathbf{r})$, we follow the work of Aronovitz *et al.* [26] by allowing the (formerly constant) parameters in $\mathcal{F}_s$ [i.e., $a(T)$ and w] to depend on the local value of $\mathbf{u}$ in a manner consistent with the symmetries of the system. Thus, we expand in powers of the strain tensor, $a(T) \to a^{(0)} + a^{(1)}_{\alpha\beta} u_{\alpha\beta} + a^{(2)}_{\alpha\beta\gamma\delta} u_{\alpha\beta} u_{\gamma\delta} + \cdots$, and similarly for w. Here, the tensor $a_{\alpha\beta}$ has the same symmetry as $c_{\alpha\beta}$, and hence for a uniaxial crystal (such as the hcp phase of solid helium [24]) $a^{(1)}_{\alpha\beta} = a_z n_\alpha n_\beta + a_\perp (\delta_{\alpha\beta} - n_\alpha n_\beta)$, with a_z and $a_\perp$ independent coupling constants.

The terms of $O(u^2_{\alpha\beta})$ in the expansion of a and of $O(u_{\alpha\beta})$ in the expansion of w all prove, by naïve power counting [23], to be irrelevant; i.e., they do *not* affect the universal critical properties of the transition. Hence, these properties follow from the following minimal model:

$$\mathcal{F}_{\rm rel} = \int d^3 r \Big\{ \frac{1}{2} c_{\alpha\beta} \partial_\alpha \psi \partial_\beta \psi^* + \frac{1}{2} a^{(0)} |\psi|^2 + \frac{1}{4!} w |\psi|^4 + \frac{1}{2} \lambda_{\alpha\beta\gamma\delta} u_{\alpha\beta} u_{\gamma\delta} + \frac{1}{2} a^{(1)}_{\alpha\beta} u_{\alpha\beta} |\psi|^2 \Big\}. \quad (2)$$

In our minimal model the effect of the elasticity is to produce a local, strain-dependent critical temperature for the superfluid. In fact, our minimal model is formally equivalent to that of a magnetic system of planar spins [an $O(2)$ ferromagnet] on a compressible lattice, for which locally dilating or compressing the lattice causes the exchange couplings to decrease or increase, resulting in a local change of the critical temperature. Compressible models of this sort have a long history [27], but the most relevant work is that of De Moura *et al.* [28], which shows that the elasticity is irrelevant to the critical properties of the $O(2)$ ferromagnet, provided the specific-heat exponent α of the decoupled (i.e., $a^{(1)}_{\alpha\beta} = 0$) system is negative, which it is at $d = 3$ [29]. Thus, we may conclude that the universality class, and hence universal properties, associated with the supersolid transition are unaltered by the coupling of elastic degrees of freedom to the supersolid order parameter. In particular, this implies a λ anomaly in the specific heat C_p near the transition:

$$C_p(t) = \frac{A^\pm}{\alpha} |t|^{-\alpha} (1 + a^\pm_c |t|^\Delta + b^\pm_c |t|^{2\Delta} + \cdots) + B, \quad (3)$$

where $t \equiv (T - T_c)/T_c$ denotes the reduced temperature, $\alpha = -0.0127 \pm 0.0003$ is the universal specific-heat exponent of the superfluid transition, and $\Delta = 0.529 \pm 0.009$ is the equally universal correction to the scaling exponent [29]. The subscripts $+$ and $-$ in Eq. (3) denote behaviors above ($T > T_c$) and below ($T < T_c$) the transition, respectively. Although the constants $A_\pm$ are nonuniversal (they will, e.g., change as one moves along the SS-NS phase boundary in the pressure-temperature phase diagram), their *ratio is* universal; the current best estimate of its value is $A_+/A_- = 1.053 \pm 0.002$ [29]. The superfluid density tensor $\rho^{\rm S}_{\alpha\beta}$ also exhibits universal scaling with reduced temperature t, i.e., $\rho^{\rm S}_{\alpha\beta} = {}^0\rho^{\rm S}_{\alpha\beta} |t|^\nu$, where the tensor ${}^0\rho^{\rm S}_{\alpha\beta}$ is temperature independent, and $\nu = 0.671\,55 \pm 0.000\,27$ is [29] the universal correlation-length exponent.

Despite the irrelevance of the elastic couplings to ψ for the universality class of the transition, these couplings *do* have important, experimentally observable consequences: because of them, the elastic properties inherit singularities in their temperature dependence from parent singularities associated with the critical fluctuations of the supersolid order parameter. Following Ref. [26], we construct the effective free energy $\mathcal{F}_{\rm eff}$ governing the elastic fluctuations by integrating out the supersolid fluctuations:

$$e^{-\mathcal{F}_{\rm eff}[u_{\alpha\beta}]/k_{\rm B}T} \equiv \int \mathcal{D}(\psi, \psi^*) e^{-\mathcal{F}_{\rm rel}[\psi, u_{\alpha\beta}]/k_{\rm B}T}. \quad (4)$$

Proceeding perturbatively, in powers of the $\psi - u_{\alpha\beta}$ coupling term, we obtain (neglecting an additive constant)

$$\mathcal{F}_{\rm eff}[u_{\alpha\beta}] = \int d^3 r \Big\{ \frac{1}{2} (\lambda_{\alpha\beta\gamma\delta} + \delta\lambda_{\alpha\beta\gamma\delta}) u_{\alpha\beta} u_{\gamma\delta} + \sigma_{\alpha\beta} u_{\alpha\beta} \Big\}, \quad (5)$$

where the singular parts of the fluctuation corrections to the bare elastic constant and stress tensors are given, to leading order, by

$$\delta\lambda_{\alpha\beta\gamma\delta} = -a^{(1)}_{\alpha\beta} a^{(1)}_{\gamma\delta} \hat{C}(t)/k_{\rm B}T, \quad (6a)$$

$$\sigma_{\alpha\beta} = +a^{(1)}_{\alpha\beta} D(t). \quad (6b)$$

Here, the governing functions, $\hat{C}$ (which proves to be proportional to the singular part of the specific heat) and D, are given by

$$D(t) \equiv \left\langle \frac{|\psi(\mathbf{r})|^2}{2} \right\rangle_0 = \frac{\partial \tilde{\mathcal{F}}_{\rm rig}}{\partial a^{(0)}}, \tag{7a}$$

$$\hat{C}(t) \equiv \lim_{k\to 0} \int \frac{d^d k'}{(2\pi)^d} \left\langle \frac{|\psi|^2_{\mathbf{k}}}{2} \frac{|\psi|^2_{\mathbf{k}'}}{2} \right\rangle_0^c = -k_{\rm B} T \frac{\partial^2 \tilde{\mathcal{F}}_{\rm rig}}{\partial a^{(0)2}}, \tag{7b}$$

where the expectation values $\langle \cdots \rangle_0$ are taken with respect to the *rigid* ψ system (i.e., a system in which all $u_{\alpha\beta}$'s are frozen at zero), whose free energy $\mathcal{F}_{\rm rig}$ is just $\mathcal{F}_{\rm eff}[u_{\alpha\beta}]$ with all $u_{\alpha\beta}$'s set to zero. Moreover, $|\psi|^2_{\mathbf{k}}$ denotes the Fourier transform of $|\psi(\mathbf{r})|^2$, and $\langle \cdots \rangle^c$ indicates a connected correlator. In addition, $\tilde{\mathcal{F}}_{\rm rig}$ is the free energy density associated with $\mathcal{F}_{\rm rig}$.

$D(t)$ and $\hat{C}(t)$ can readily be related to the specific-heat singularity of the rigid system, by noting that $a^{(0)}$ is a linear function of T close to T_c, so that derivatives with respect to $a^{(0)}$ are proportional (near T_c) to derivatives with respect to T. Hence, simple thermodynamic identities imply that $D(t)$ and $\hat{C}(t)$ are proportional to the entropy and the specific heat of the rigid system, respectively, near T_c. This immediately determines their critical behavior: $D(t) = G^{\pm}|t|^{1-\alpha}(1 + a_c^{\pm}|t|^{\Delta} + \cdots)$ and $\hat{C}(t) = A'^{\pm}|t|^{-\alpha}(1 + a_c^{\pm}|t|^{\Delta} + \cdots)$, where $G^+/G^- = A'^+/A'^- = A^+/A^- = 1.053 \pm 0.002$ are universal.

It is evidently valuable to estimate the size of the expected elastic and thermal expansion anomalies. To do this, we need an estimate of the couplings $a^{(1)}_{\alpha\beta}$ in Eq. (2), and this can be obtained, following Ref. [30], from the form of the SS-NS phase boundary, $T_c(P)$, in the pressure-temperature phase diagram. In particular, the couplings are related to the slope of the boundary via

$$\partial T_c(P)/\partial P \sim a^{(1)}/(\lambda a'), \tag{8}$$

where $a' \equiv da^{(0)}/dT$, and λ is a typical elastic constant. With this in hand, we can now estimate the size of the elastic and thermal expansion anomalies. This can be accomplished by using Eq. (7) to estimate $\hat{C}$ and D well away from the critical point (say, at $T = 2T_c$), where we can make the Gaussian approximation to the correlation functions, which gives, e.g.,

$$D(T)|_{T=2T_c} \sim \int d^3k \frac{k_{\rm B}T}{a^{(0)}(T)} \sim \frac{k_{\rm B}}{a'\xi_0^3}, \tag{9}$$

where ξ_0 is the high-temperature correlation length for ψ fluctuations. We have taken the integral over k to have an ultraviolet cutoff comparable to ξ_0, used the fact that, well above T_c, $a^{(0)}(T)/c \sim \xi_0^{-2}$ to replace the propagator [up to $O(1)$ factors] by $a^{(0)}$ for all k, and estimated $a(T = 2T_c) \sim a'T_c$. For superfluid ^{4}He, the length ξ_0 is known to be comparable to the atomic size: $\xi_0 \sim 0.2$ nm; for want of better information, we shall assume that this is also true for the supersolid.

By using this estimate for $D(T)$ in Eq. (6b), and then minimizing Eq. (5) over $u_{\alpha\beta}$, we arrive at a typical value for the thermal expansion:

$$\delta u_{\alpha\beta} \sim \frac{a^{(1)}}{\lambda} D(T) \sim \frac{\partial}{\partial P} \frac{k_{\rm B}T_c(P)}{\xi_0^3}, \tag{10}$$

where in the last step we have used Eq. (8). Estimating $|\partial T_c(P)/\partial P| \sim 10^{-2}$ K/atm by its value in the *liquid* state of ^{4}He [24] gives a typical value of $\delta u_{\alpha\beta} \sim 0.16$. For the fractional anomaly in the elastic constants, arguments essentially identical to those just used give

$$\frac{\delta\lambda}{\lambda} \sim \frac{\lambda k_{\rm B}}{T_c \xi_0^3} \left(\frac{\partial T_c(P)}{\partial P} \right)^2 \sim 0.17. \tag{11}$$

Perhaps the best way to observe the predicted anomalies in the elastic constants is through sound speed measurements. In *single crystals,* the polarization of the sound modes studied must be chosen judiciously: the uniaxial form for the expansion coefficients, $a^{(1)}_{\alpha\beta} = a_z n_\alpha n_\beta + a_\perp(\delta_{\alpha\beta} - n_\alpha n_\beta)$, implies that only "bulk" elastic moduli, specifically, the terms $\lambda_{zz}(n_\alpha n_\beta u_{\alpha\beta})^2 + \lambda_{\perp\perp}((\delta_{\alpha\beta} - n_\alpha n_\beta)u_{\alpha\beta})^2 + \lambda_{\perp z}((\delta_{\alpha\beta} - n_\alpha n_\beta)u_{\alpha\beta})(n_\alpha n_\beta u_{\alpha\beta})$, acquire anomalous temperature dependence. The elastic constants λ_{zz}, $\lambda_{\perp\perp}$, and $\lambda_{\perp z}$ are readily shown by a standard sound mode analysis [31] to affect only the sound speeds of modes polarized in the plane formed by the normal to the hexagonal layers and the direction of propagation. Furthermore, in this plane, transverse modes propagating either along, or orthogonal to, the layers also have sound speeds independent of the anomaly displaying elastic constants λ_{zz}, $\lambda_{\perp\perp}$, and $\lambda_{\perp z}$. Hence, to observe the anomaly in the sound speeds in a single crystal of supersolid hcp helium, one should study modes polarized in the plane formed by the hexagonal layers and the direction of propagation, *and* choose that propagation direction *not* to lie in, or orthogonal to, the layers.

It is, however, unlikely that experiments will be performed on single crystals of helium. It is far more likely that they will be performed on polycrystalline samples, which are macroscopically isotropic, due to the random orientations of the constituent crystallites. Calculating the isotropic shear modulus μ and Lamé coefficient λ of such an ensemble of randomly oriented crystallites is well known to be a formidable problem (see, e.g., Ref. [32]). Nonetheless, using the exact bounds of Hill [33], we can show [31] that *both* the shear modulus and the bulk modulus of a macroscopically isotropic polycrystalline helium sample will exhibit the $|t|^{-\alpha}$ anomaly we predict here. As a result, *both* the transverse *and* the longitudinal sound speeds of such a sample will show the $|t|^{-\alpha}$ anomaly.

Beyond the critical properties described above, our model has the important implication that in a helium crystal the SS-NS transition would be *rounded* by any spatially inhomogeneous internal stresses that make $a^{(1)}_{\alpha\beta}u_{\alpha\beta} \neq 0$. As such stresses are almost unavoidable in any crystal (and are believed to be present in the experiments of Kim and Chan [15,16]), this rounding is almost certain to be present in all experiments performed to date.

The reason for such broadening is very simple: stresses that make $a^{(1)}_{\alpha\beta}u_{\alpha\beta}$ spatially inhomogeneous make T_c of the SS-NS transition spatially inhomogeneous as well; cf. Eq. (2). Therefore, roughly speaking, distinct parts of the sample would become supersolid at distinct temperatures, broadening the transition. Such broadening is evident in the $\rho^S(T)$ vs T plots of Kim and Chan [16], which clearly *do not* show the expected $|t|^\nu$ singularity near the putative T_c, with ν being the correlation-length exponent. [If they did, $\rho^S(T)$ vs T would hit the horizontal axis *perpendicularly,* rather than—as in those data—tangentially.] This broadening may also explain the apparent absence of the expected λ anomaly in the specific heat [34] near the putative T_c in those experiments: this peak is simply "smeared away."

One intriguing, albeit highly speculative, final implication of the coupling of strains to the supersolid order parameter ψ has to do with the very existence of the supersolid state in ^{4}He. A number of microscopic calculations [17] suggest that ^{4}He does *not* have a supersolid state at all, in contradiction with the experiments of Kim and Chan [15]. These calculations were presumably done under conditions of purely *hydrostatic* stress (i.e., simple pressure), for which only the three diagonal components of the strain tensor are nonzero and equal (i.e., $u_{xx} = u_{yy} = u_{zz} \neq 0$). If it were the case that the coefficients a_z and $a_\perp$ in the uniaxial coupling tensor for solid hcp ^{4}He happened to obey $a_z \approx -2a_\perp$, then the effect of this pressure on the effective T_c for supersolid order would be very small. If the *unstrained* T_c were negative [i.e., the coefficient $a^{(0)}$ in Eq. (2) were positive for all T], this would imply that, under such a hydrostatic stress, the crystal would never enter the supersolid state. If, however, the crystal were to be subjected to an *anisotropic* stress (i.e., one for which the relation $u_{xx} = u_{yy} = u_{zz}$ is *not* satisfied), the near cancellation of $a^{(1)}_{\alpha\beta}u_{\alpha\beta}$ would *not* occur, and this term might be able to make the effective T_c positive [i.e., change the overall sign of the coefficient of $|\psi|^2$ in Eq. (2)]. That is, it is possible in our model that, although a *hydrostatically* stressed sample would *not* show a supersolid phase, an *anisotropically* stressed one might. This could be highly significant, as Kim and Chan [15] believe that such anisotropic stresses *are* present in their samples. Although the above argument is obviously quite speculative, it seems to us, nonetheless, a possibility that these random stresses, far from being an experimental nuisance, might just be what is responsible for the presence of supersolidity. Support for this idea comes from two other facts: (1) Many experiments [13,14] do not see supersolidity. Perhaps these samples simply lacked sufficiently large inhomogeneous, anisotropic stresses. (2) The superfluid fraction in the experiments of Kim and Chan [15] is extremely low. Could this be because only very small, highly anisotropically stressed regions of the sample are going supersolid?

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