Low temperature properties of solid ⁴He: Supersolidity or quantum "metallurgy"?

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And thanks to the NSF!

Where's Gainesville?



Supersolidity or Quantum Metallurgy?

Outline

□ Can BEC occur in a solid?

- A brief history of "supersolids"
- How would we know?
- Some curious experimental results
 - Torsional oscillator
 - Shear modulus anomalies
 - Specific heat
- Supersolidity or quantum metallurgy? The role of dislocations and other defects.

Superfluidity in ⁴He

- Discovered in 1937 by Kapitsa, and by Allen & Misener.
- Properties:
 - Zero viscosity-flow through narrow pores
 - Fountain effect
 - Creep along (up) surfaces
 - Second sound ("thermal waves")
- Related to BEC (but strong interactions!)



BEC in dilute atomic gases

- Discovered in 1995 by
 Wieman & Cornell (⁸⁷Rb)
 and Ketterle (²³Na).
- □ Temperature of 170 nK!
- Weak interactions (unlike ⁴He).
- Fermionic condensates also possible—⁴⁰K in a magnetic field (D. Jin, 2003).







Supersolidity or Quantum Metallurgy?

Can BEC occur in a solid?

- BEC occurs in the gas and liquid phases; can it also occur in a solid?
- Can off-diagonal long range order (Bose condensation) coexist simultaneously with shear rigidity (a solid)?
- No: BEC requires extended wavefunctions, solids have localized wavefunctions.
- □ Yes: why not?
- What do the theorists say? Are "supersolids" possible?

Penrose & Onsager (1956): no

PHYSICAL REVIEW

VOLUME 104, NUMBER 3

NOVEMBER 1, 1956

Bose-Einstein Condensation and Liquid Helium

OLIVER PENROSE* AND LARS ONSAGER Sterling Chemistry Laboratory, Yale University, New Haven, Connecticut (Received July 30, 1956)

The mathematical description of B.E. (Bose-Einstein) condensation is generalized so as to be applicable to a system of interacting particles. B.E. condensation is said to be present whenever the largest eigenvalue of the one-particle reduced density matrix is an extensive rather than an intensive quantity. Some transformations facilitating the practical use of this definition are given.

An argument based on first principles is given, indicating that liquid helium II in equilibrium shows B.E. condensation. For absolute zero, the argument is based on properties of the ground-state wave function derived from the assumption that there is no "long-range configurational order." A crude estimate indicates that roughly 8% of the atoms are "condensed" (note that the fraction of condensed particles need not be identified with ρ_s/ρ). Conversely, it is shown why one would not expect B.E. condensation in a solid. For finite temperatures Feynman's theory of the lambda-transition is applied: Feynman's approximations are shown to imply that our criterion of B.E. condensation is satisfied below the lambda-transition but not above it.





Chester (1970): well, maybe

PHYSICAL REVIEW A

VOLUME 2, NUMBER 1

JULY 1970

Speculations on Bose-Einstein Condensation and Quantum Crystals*

G. V. Chester

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It is shown, by almost rigorous arguments, that there exist many-body states of a system of interacting bosons which exhibit both crystalline order and Bose-Einstein condensation into the zero-momentum eigenstate of the single-particle density matrix. The implications of this result are discussed in relation to theories of superfluidity and the nature of quantum crystals.

type will give an accurate description of the exact states of solid and liquid helium four. We make this statement because the arguments presented above allow us to include in the wave function as complicated explicit correlations as we like. It is of course true that there are other states one can envisage which do not satisfy our condition, for example, a linear combination of Jastrow states. Indeed, if no state in this wide class of model wave functions adequately describes the spatial correlations of a real-quantum crystal, then these correlations must be fundamentally different from those which occur in a classical crystal. For these reasons, we believe that our speculation about real physical systems are on much firmer grounds than would appear at first sight.

Finally, we comment on the proof by Onsager and Penrose² that a state with crystalline order cannot have a Bose-Einstein condensate in the zero-momentum state. This proof is based on a particular class of model states in which each particle is localized on a lattice site, each site is occupied by a particle and symmetry is ignored. If either of the latter restrictions is removed, then the original proof fails.⁶ In particular, if there are vacancies in the model state and symmetry is ignored, then a condensate exists and this condensation would presumably persist if symmetry were taken into account. It is now interesting to note that we expect all the model states we have discussed to lead to crystalline order with vacancies present. This is because the equivalent classical system would be expected, on physical grounds, to lead to crystallization with a finite fraction of vacancies. We may therefore add one final speculation, namely, that a quantum crystal can only have a Bose-Einstein condensate if it has a finite fraction of vacancies. We see no reason, whatsoever, to suppose that a quantum crystal cannot have a finite fraction of vacancies at absolute zero. Liquid helium exists at absolute zero and this suggests that a crystal with a finite amount of spatial disorder could exist at absolute zero. If, on the other hand, a finite fraction of vacancies can only exist at elevated temperatures, then it might be impossible to have a Bose-Einstein condensate because of the high temperature. We pointed out in Sec. IV that it is almost impossible to predict the temperature at which such a condensation might occur.

Supersolidity or Quantum Metallurgy?

Vacancies and interstitials

Local density changes arise from either lattice fluctuations (with a displacement field u) or vacancies and interstitials.

$$\delta \rho = \delta \rho_{\Delta} - \rho_0 \nabla \cdot \mathbf{u}$$

In classical solids the density of vacancies is small at low temperatures.

 $n_{\text{vacancies}} = a^{-3} e^{-E_V/T}$

Does ⁴He have zero point vacancies?



$$\delta \rho_{\Delta} = (\delta \rho_{\text{interstitial}} - \delta \rho_{\text{vacancy}})$$

Supersolidity in Jastrownium[™]

□ Jastrow variational wavefunction for N bosons:

 $\Psi_N(\mathbf{r}_1,\ldots,\mathbf{r}_N) = \prod_{i\neq j} f(r_{ij}) = \exp\left[-\frac{1}{2}\sum_{i\neq j} u(r_{ij})\right]$

Probability distribution is identical to the classical Gibbs distribution for N particles interacting through a potential V(r_{ij})=T_{eff} u(r_{ij})

$$P_N = \Psi_N^2 = \exp\left[-\sum_{i \neq j} V(r_{ij})/T_{\text{eff}}\right]$$

- Expect a solid phase for some sufficiently low T_{eff} , with a defect density of $n_{vacancies} = a^{-3}e^{-E_V/T_{eff}}$
- Proof of principle: Jastrownium can have both ODLRO and crystalline order. What about ⁴He?
- Andreev and Lifshitz (1969): defects Bose condense, producing a condensate that lives within the solid phase.

Andreev & Lifshitz: here's one way

SOVIET PHYSICS JETP

VOLUME 29, NUMBER 6

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QUANTUM THEORY OF DEFECTS IN CRYSTALS

A. F. ANDREEV and I. M. LIFSHITZ

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Submitted January 15, 1969

Zh. Eksp. Teor. Fiz. 56, 2057-2068 (June, 1969)

At sufficiently low temperatures localized defects or impurities change into excitations that move practically freely through a crystal. As a result instead of the ordinary diffusion of defects, there arises a flow of a liquid consisting of "defectons" and "impuritons." It is shown that at absolute zero in crystals with a large amplitude of the zero-point oscillations (for example, in crystals of the solid helium type) zero-point defectons may exist, as a result of which the number of sites of an ideal crystal lattice may not coincide with the number of atoms. The thermodynamic and acoustic properties of crystals containing zero-point defectons are discussed. Such a crystal is neither a solid nor a liquid. Two kinds of motion are possible in it; one possesses the properties of motion in an elastic solid, the second possesses the properties of motion in a liquid. Under certain conditions the "liquid" type of crystal motion possesses the property of superfluidity. Similar effects should also be observed in quasiequilibrium states containing a given number of defectons.

Leggett (1970): look for NCRI!

Can a Solid Be "Superfluid"?

A. J. Leggett

School of Mathematical and Physical Sciences, University of Sussex, Falmer, Brighton, Sussex, England (Received 15 September 1970)



In this Letter we shall suggest that, on the contrary, it is impossible to exclude the occurence of NCRI also in insulating¹ solids (where "solid" is defined phenomenologically-see below), and that if it does occur it should produce a number of interesting phenomena analogous to those of superfluidity. However, we shall show that the associated "superfluid fraction" must be very small even at T = 0 (probably always $\leq 10^{-4}$). As a result, these phenomena could well have escaped notice even if "superfluid solids" do exist at temperatures already reached, since they have not (to the best of the author's knowledge) been specifically looked for. While the ideas discussed here are somewhat speculative, an experiment to test them should be relatively simple and seems well worthwhile.

If NCRI should indeed occur in a solid, how would it manifest itself? The most direct experiment to look for it would be to rotate the solid in the form of an annulus¹² below its transition temperature; then the apparent moment of inertia should be slightly less than the classical value I_0 (and, more relevantly, presumably temperature dependent). A second test would be to rotate the solid above its presumed critical angular velocity ω_c and then bring the container to rest; if we assume that NCRI is associated with the metastability of flow states as in other superfluid systems, we should expect a persistent residual angular momentum $(\rho_s/\rho)I_0\omega_c$. In view of the small value of ρ_s/ρ , it seems highly unlikely that these effects would have been discovered by accident even if "superfluid solids" do exist at attained temperatures.

Supersolidity or Quantum Metallurgy?

That's entertainment!



Supersolidity or Quantum Metallurgy?

Phase diagram of ⁴He

- Solid ⁴He is *soft*: shear modulus of 20 MPa (Al is 26 GPa, butter is 5 MPa).
- Solid ⁴He is *light*:density of 0.2 g/cm³ (like cork or balsa wood).
- $\Box \quad \theta_{\text{Debye}} = 25 \text{ K.}$
- de Boer parameter
 - $\Lambda\equiv\lambda/(2\pi\sigma)=0.282$

Solid Ar has $\Lambda = 0.019$. Quantum effects are important.



http://ltl.tkk.fi/research/theory/helium.html

Penn State experiments

(New York Times, 21 September 2004)



- ⁴He in vycor glass: Nature **427**, 225 (2004).
 Bulk ⁴He: Science **305**, 1941 (2004).
- Now reproduced by several groups: PSU, Cornell, Rutgers, Keio, Tokyo, Seoul, ...

$$\tau = 2\pi \sqrt{I/\alpha}$$

Supersolidity or Quantum Metallurgy?

Data from PSU (Science 2004)





Pressures from 26 to 66 bars. Amplitude of about 1 nm, maximum velocities of about 10 μ m/s. Frequency: 1 kHz (period of 10⁶ ns). Period shifts of order 40 ns. Q of order 10⁶.

Barrier inserted: no effect.

Effects of ³He impurities



Onset T decreases 250 to 75 mK

Transition sharpens as ³He is reduced

Supersolidity or Quantum Metallurgy?

Shear stiffening



- Day and Beamish (Nature 2007): shear modulus increases by 10-20% at low temperature
- Onset temperature of stiffening for pure samples at 80 mK, similar to torsional oscillator.
- Shape of stiffening similar to period shift.
- \Box Sensitive to ³He impurities.
- See ATD and DA Huse (Nature 2007).

Summary so far...

- Effect observed by several different groups.
- Sample preparation is important: annealing reduces effect (Rittner & Reppy).
- Small quantities of ³He affect the magnitude of NCRI.
- No pressure driven flow (Day & Beamish), but chemical potential driven flow (Ray and Hallock)?
- Mechanical changes with reduced temperature: shear stiffening.
- Monte Carlo: perfect crystal is insulating. Vacancy creation energy about 13K.

- Are the mechanical changes and NCRI related?
- What is the role of defects dislocations or grain boundaries?
- □ Why is ³He important?
- What is the origin of the low critical velocity?
- For non-superfluid explanations, what about the c-shaped sample cells?

Torsional oscillator: rigid body

- **Equation of motion for a rigid solid:** $\left[(I_{cell} + I_{He}) \frac{d^2}{dt^2} + \gamma \frac{d}{dt} + \alpha \right] \theta(t) = \tau_{ext}(t)$
- Resonant period:

$$P = P_0 \left(1 + \frac{1}{8Q^2} + \dots \right), \quad Q = \frac{\sqrt{\alpha I_{\text{total}}}}{\gamma} = \mathcal{O}(10^6)$$
$$P_0 = 2\pi \sqrt{\frac{I_{\text{cell}} + I_{\text{He}}}{\alpha}}$$
Be-Cu torsion

$$\frac{\Delta P}{P_0} = \frac{1}{2} \frac{\Delta I_{\text{tot}}}{I_{\text{tot}}} = \mathcal{O}(10^{-5})$$

What happens if the solid ⁴He is not rigid?





Torsional oscillator: elastic solid



$$\underbrace{\left(I_{\text{cell}}\frac{d^2}{dt^2} + \gamma \frac{d}{dt} + \alpha\right)\theta(t) = \tau_{\text{ext}}(t)}_{\text{back reaction from elastic solid}} + \underbrace{\frac{M(t)}{t}}_{\text{back reaction from elastic solid}}$$

■ Back action: moment that the solid ⁴He exerts on the walls of the cell (linear response): $M(t) = \int dt'g(t - t')\theta(t'), \quad M(\omega) = g(\omega)\theta(\omega)$ □ Oscillator response $f\chi(\omega) = \theta(\omega)/\tau_{ext}(\omega)$ $\chi^{-1}(\omega) = -I_{cell}\omega^2 - i\gamma\omega + \alpha - q(\omega)$

The complex poles of the response function determine the resonant frequency and dissipation of the system.

Elastic response of the solid

□ *All* of the information about the solid ⁴He is contained in $g(\omega)$. It has the following properties:

□ analytic in upper half frequency plane;

real and imaginary parts obey Kramers-Kronig relations;

□ low frequency behavior must be a rigid solid: $g(\omega) = I_{\rm He}\omega^2 + O(\omega^3)$

□ To calculate $g(\omega)$ we need to solve the equation of motion for an elastic solid:

$$\rho \partial_t^2 u_i = \partial_j \sigma_{ij}, \quad u_{ik} = \left(\partial_k u_i + \partial_i u_k\right)/2$$

 $\Box \text{ Hooke's Law (nonlocal in time):} \\ \sigma_{ij}(t) = \int dt' K_{ijkl}(t-t')u_{kl}(t'), \quad \sigma_{ij}(\omega) = K_{ijkl}(\omega)u_{kl}(\omega)$

Viscoelasticity

□ Isotropic elasticity:

 $K_{ijkl}(\omega) = \lambda(\omega)\delta_{ij}\delta_{kl} + \mu(\omega)(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})$

 $-\rho\omega^2\mathbf{u} = B(\omega)\nabla(\nabla\cdot\mathbf{u}) - \mu(\omega)\nabla\times\nabla\times\mathbf{u}, \qquad B = \lambda + 2\mu$

- □ Shear motion of an elastic solid: $\rho \partial_t^2 \mathbf{u} = \mu_0 \nabla^2 \mathbf{u}$
- □ Navier-Stokes for a viscous fluid: $\rho \partial_t \mathbf{v} = \eta \nabla^2 \mathbf{v} \xrightarrow{\mathbf{v} = \partial_t \mathbf{u}} \rho \partial_t^2 \mathbf{u} = \eta \partial_t \nabla^2 \mathbf{u}$
- Combining (in "parallel"): $\rho \partial_t^2 \mathbf{u} = (\mu_0 + \eta \partial_t) \nabla^2 \mathbf{u}$
- □ Kelvin-Voigt model (internal friction): $\mu(\omega) = \mu_0 + i\eta\omega = \mu_0(1 + i\omega\tau), \quad \tau = \eta/\mu_0$

μ

Boundary value problem



□ Shear stress exerted by the solid on the cell:

$$\sigma_{\theta r} = \mu(\omega) \left(\partial_r - \frac{1}{r} \right) u_{\theta} \bigg|_{r=R} = -\theta_0 R^2 \rho \omega^2 \frac{J_2(kR)}{kR J_1(kR)}$$

Supersolidity or Quantum Metallurgy?

Properties of results

TO is a probe of the shear modulus. The period shift and the dissipation are related! $\frac{\Delta P}{P} \propto \operatorname{Re} \left| \frac{1}{\mu(\omega)} \right|, \quad \Delta Q^{-1} \propto \operatorname{Im} \left| \frac{1}{\mu(\omega)} \right|$ Corrections vanish for a rigid solid. \Box The peak value of ΔQ^{-1} is independent of τ : $\Delta Q^{-1}\Big|_{\max} = \underbrace{\frac{1}{48}}_{10^{-2}} \underbrace{\frac{I_{\text{He}}}{I_{\text{tot}}}}_{10^{-2} - 10^{-3}} \underbrace{\left(\frac{\omega_0 R}{c_T}\right)^2}_{10^{-2}} = \mathcal{O}(10^{-6} - 10^{-7})$ \Box At the peak, $\Delta Q^{-1}\big|_{\rm max} = 2\Delta P/P\big|_{\rm max}$

□ For no dissipation, changing the shear modulus changes the period (inertial overshoot):

$$\frac{\Delta P}{P} = -\frac{1}{48} \left(\frac{\omega_0 R}{c_T}\right)^2 \frac{I_{\rm He}}{I_{\rm tot}} \frac{\Delta \mu}{\mu}$$

Data fits (Yoo & ATD, PRB 2009)



- Dissipation peak identifies long relaxation time on the order of $1 \text{ms } \tau = \tau_0 \exp(E_0/T)$. Dislocations?
- Model seems to only account for 10% of the period shift. Suitable *distributions* of relaxation times allow a fit of both the period shift and dissipation peak (Su *et al.*, PRL 2010).
- □ Can dislocations be superfluid?

Landau theory for a superfluid

- Symmetry of order parameter \$\psi(\mathbf{x}) \rightarrow \psi(\mathbf{x}) e^{i\phi}\$
 Broken U(1) symmetry for T<T_c.
 Coarse-grained free energy:
 \$\mathcal{F}_{sf} = \int_{\mathbf{x}} \{ \frac{1}{2}c |\nabla \psi|^2 + \frac{1}{2}a(T)|\psi|^2 + \frac{w}{4!}|\psi|^4 \}, \$a(T) = a_0(T - T_0)\$
- Average over configurations:

$$Z = \int D\psi D\psi^* e^{-\mathcal{F}_{\rm sf}/T}, \qquad F = -T \ln Z$$

- □ Fluctuations shift $T_0 \rightarrow T_c$, produce singularities as a function of the reduced temperature t=|(T-T_c)/T_c|.
- Universal exponents and amplitude ratios.





Coupling superfluidity & elasticity

ATD, Goldbart & Toner (PRL 2006): Landau model with coupling between superfluidity and elasticity (strain dependent T_c):

$$\mathcal{F}_{ss} = \int_{\mathbf{x}} \left\{ \frac{1}{2} c_{ij} \partial_i \psi \partial_j \psi^* + \frac{1}{2} a^{(0)} |\psi|^2 + \frac{w}{4!} |\psi|^4 + \frac{1}{2} K_{ijkl} u_{ij} u_{kl} + \frac{1}{2} a^{(1)}_{ij} u_{ij} |\psi|^2 \right\}.$$

- Predictions
 - XY anomaly in specific heat (lambda transition)
 - Anomalies in elastic constants; shows up as a dip in the sound speed at the transition:

$$K_{ijkl} = -T \frac{\partial^2 F}{\partial u_{ij} \partial u_{kl}}$$

= $K_{ijkl}^{(0)} - \frac{1}{4T} a_{ij}^{(1)} a_{kl}^{(1)} \int_{\mathbf{x}} \int_{\mathbf{x}'} \langle |\psi(\mathbf{x})|^2 |\psi(\mathbf{x}')|^2 \rangle_0$

Supersolidity from dislocations?

- Dislocations: topological defects in a crystal (a string). Orowan, Polanyi and Taylor (1934) proposed that plastic deformation of solids can be described by dislocations.
- Dislocations can promote superfluidity. Recall model: $\mathcal{F}_{ss} = \int_{\mathbf{x}} \left\{ \frac{1}{2}c |\nabla \psi|^2 + \frac{1}{2}t(\mathbf{r})|\psi|^2 + \frac{w}{4}|\psi|^4 \right\},\$ $t(\mathbf{r}) = t_0 + gu_{ii}$
- Quenched dislocations produce large, long-ranged strains. For an edge dislocation (isotropic elasticity)

$$u_{ii} = \nabla \cdot \mathbf{u} = \frac{4\mu}{2\mu + \lambda} \frac{a\cos\theta}{r_{\perp}}$$

Even if t₀>0 (PIMC), can have t<0 near the dislocation! Solve linearized Landau equation.





Edge dislocation



Screw dislocation

Supersolidity or Quantum Metallurgy?

Details: 2d quantum dipole

Schrodinger equation for edge dislocation:

$$-\frac{\hbar^2}{2m_3}\nabla^2\psi + p\frac{\cos\theta}{r}\psi = E\psi$$



Numerical methods (see K. Dasbiswas *et al.*, PRB 2010)

$$E_0 = -0.28m_3 p^2/\hbar^2 = -0.28\frac{m_3}{\hbar^2} \left(\frac{\mu b}{3\pi}\frac{1+\nu}{1-\nu}\delta V\right)^2 \simeq -1 \text{ K}$$

□ What about screw dislocations? Need nonlinear strains, $U(r) \sim 1/r^2$. See recent Corboz et al., PRL 2008; binding energy $-E_0 = 0.8 \pm 0.1$ K.

Reduction to a 1D model

Integrate out gapped transverse degrees of freedom in time-dependent Landau theory $\frac{\partial \psi}{\partial t} = \bar{\epsilon}\psi + \hat{L}\psi + \partial_z^2\psi - \epsilon|\psi|^2\psi + \zeta$ $\hat{L} \equiv \nabla_T^2 - u(x, y) + \epsilon_0$ b □ Threshold Ansatz: ξ_{\perp} $\psi(x, y, z, t) = A(z, t)\phi_0(x, y)$ Amplitude equation: $\frac{\partial A}{\partial t} = \bar{\epsilon}A + \partial_z^2 A - g|A|^2 A + \zeta_1 \qquad g = \epsilon \int dx dy |\phi_0|^4$

Supersolidity or Quantum Metallurgy?

Network model

1D superfluid order along a single dislocation, can overlap with neighboring dislocations

Network of dislocations in solid ⁴He bulk superfluid order (Shevchenko 1988, Toner 2008)



Derivation of the network model

- Superfluid coupling between sites i, j
- □ Integrate over fluctuations along 1D
- Effective coupling between neighboring sites obtained from a "propagator":

$$K(\psi_i, \psi_j; L) = \int D\psi D\psi^* \exp\left\{-\frac{\beta}{2} \int_0^L dz \left[a|\psi|^2 + \frac{1}{2}b|\psi|^4 + c \left|\frac{d\psi}{dz}\right|^2\right]\right\}$$

 □ Effective XY model: H(ψ_i, ψ_j; L) = J_{ij}(L) cos(θ_i − θ_j) J_{ij}(L) = c|ψ_i||ψ_j| ξ sinh(L/ξ)

 □ Exponential dependence of coupling on separation between nodes.

- A dislocation in a Bose solid may have a superfluid core (Landau theory, PIMC).
- Dislocation motion leads to shear softening, dislocation pinning to stiffening.
- Dislocations bind solute atoms (e.g., ³He impurities)—could lead to specific heat anomalies.
- Perhaps the NCRI and shear stiffening are correlated—superfluid dislocations?

Summary

- New experiments have revealed a novel phenomena in solid ⁴He that is likely connected to the superfluid behavior of extended defects (low dimensional superfluidity).
- This superfluidity is coupled to changes in the elastic properties, due to defect motion. Impurities can pin the defects and enhance the superfluid response.
- Dislocation motion + BEC = "quantum metallurgy"?

The three amigos



Debajit, Kinjal and Chi-Deuk

Supersolidity or Quantum Metallurgy?
Torsional oscillator: rigid body

- **Equation of motion for a rigid solid:** $\left[(I_{cell} + I_{He}) \frac{d^2}{dt^2} + \gamma \frac{d}{dt} + \alpha \right] \theta(t) = \tau_{ext}(t)$
- Resonant period:

$$P = P_0 \left(1 + \frac{1}{8Q^2} + \dots \right), \quad Q = \frac{\sqrt{\alpha I_{\text{total}}}}{\gamma} = \mathcal{O}(10^6)$$
$$P_0 = 2\pi \sqrt{\frac{I_{\text{cell}} + I_{\text{He}}}{\alpha}}$$
Be-Cu torsion

$$\frac{\Delta P}{P_0} = \frac{1}{2} \frac{\Delta I_{\text{tot}}}{I_{\text{tot}}} = \mathcal{O}(10^{-5})$$

What happens if the solid ⁴He is not rigid?





Specific heat near the λ transition

□ The singular part of the specific heat is a correlation function:

 $S = -\partial F / \partial T \propto -\partial F / \partial a(T) = \int_{\mathbf{x}} \langle |\psi(\mathbf{x})|^2 \rangle$

 $C = T(\partial S/\partial T) \propto \int_{\mathbf{x}} \int_{\mathbf{x}'} \langle |\psi(\mathbf{x})|^2 |\psi(\mathbf{x}')|^2 \rangle \sim A_{\pm} |t|^{-\alpha}$



Lipa et al., Phys. Rev. B (2003).

Barmatz & Rudnick, Phys. Rev. (1968)

Supersolidity or Quantum Metallurgy?

Specific heat I

High resolution specific heat measurements of the lambda transition in zero gravity.



J.A. Lipa et al., Phys. Rev. B **68**, 174518 (2003).

Specific heat near the putative supersolid transition in solid ⁴He.



Figure 5 Specific heat peak of the 1 ppb, 0.3 ppm, and 10 ppm samples. The x_3 -independent peak centred around 75 mK is revealed when the phonon contribution is subtracted. The red dashed lines indicate the uncertainty in the 1 ppb data. The uncertainty for $x_3 = 0.3$ ppm is comparable. For $x_3 = 10$ ppm, it is similar above 200 mK but decreases more gradually with decreasing temperature. For T < 100 mK the uncertainty is four times larger than that of the 1 ppb sample. The inset compares the specific heat of the three samples without the subtraction of the impuriton term of the 10 ppm sample (dotted line, 59 μ J mol⁻¹ K⁻¹).

Lin, Clark, and Chan, Nature (2007)

Supersolidity or Quantum Metallurgy?

An alternative: lattice gas model

- Edge dislocations in ⁴He provide an attractive potential for ³He impurities.
- Bound ³He impurities "evaporate" from the dislocations, increasing entropy and producing a bump in the specific heat.
- Divide the impurities into bound and free; two systems are in chemical equilibrium.
- □ Treat both systems classically.
- See T. N. Antsygina et al., Low Temp. Phys. 21, 453 (1995).



bound to dislocation

particles in the bulk

Supersolidity or Quantum Metallurgy?

Binding of ³He to dislocations

- □ Hydrostatic pressure due to an edge dislocation (continuum theory): $p = -\frac{1}{3}\sigma_{ii} = \frac{\mu b}{3\pi}\frac{1+\nu}{1-\nu}\frac{\sin\theta}{r}$
- Effective potential due to a volume defect *δV* (Cottrell "atmosphere"):

$$U(r,\theta) = p\delta V = U_0 \frac{\sin\theta}{r}, \quad U_0 = \frac{\mu b}{3\pi} \frac{1+\nu}{1-\nu} \delta V$$

Breaks down in the core due to diverging strains; need a cut off. The cut off will reduce the binding energy.

Specific heat II: some details

- \square N ³He impurities, M defect sites that bind the impurities with energy ϵ .
- The defect sites have 0 or 1 ³He impurities (two level system). Ignore correlations among sites and quantum statistics.
- Assume particles that have evaporated form a noninteracting gas.

$$N = \langle N_{\text{gas}} \rangle + M \langle n_{\text{site}} \rangle, \quad \langle n_{\text{site}} \rangle = \left[1 + e^{-\beta(\epsilon + \mu)} \right]^{-1}$$

Equate chemical potentials of the gas and the adsorbed particles:

$$\langle N_{\rm gas} \rangle = \frac{1}{2} \left[N - M - p(T) + \sqrt{(N - M - p(T))^2 + 4Np(T)} \right]$$

 $p(T) = V e^{-\epsilon/T} \left(\frac{2\pi m_3 T}{h^2} \right)^{3/2}$

Supersolidity or Quantum Metallurgy?

Properties

Calculate the molar specific heat at constant N; complicated expression. Roughly, there is a background piece (from the gas particles) and a bump (Schottky anomaly) from the adsorbates.

Two limits:

- N>M ("saturated" case): size of the bump scales as M.
- N<M ("unsaturated" case): size of bump scales with N.
- \Box Peak appears at a temperature T^{*} such that
 - $\epsilon + \mu(T^*) = T^*$. The peak position depends on the ³He concentration (weakly).

Sample comparison with data



Supersolidity or Quantum Metallurgy?

Hydrodynamics I: simple fluid

Conservation laws and broken symmetries lead to long-lived "hydrodynamic" modes (lifetime diverges at long wavelengths).

□ Simple fluid:

- Conserved quantities are ρ , g_i , e. $\partial_t \rho + \partial_i g_i = 0$ (conservation of mass), $\partial_t g_i + \partial_j \sigma_{ij} = 0$ (conservation of momentum), $\partial_t e + \partial_i J_i^Q = 0$ (conservation of energy).
- No broken symmetries.
- 5 conserved densities \Rightarrow 5 hydrodynamic modes.
 - □ 2 transverse momentum diffusion modes $\omega = -iD_t q^2$.
 - 1 longitudinal thermal diffusion mode

$$\omega = -iD_T q^2$$

□ 2 longitudinal sound modes

$$\omega = \pm c_1 q$$

Light scattering from a simple fluid



P. A. Fleury and J. P. Boon, Phys. Rev. **186**, 244 (1969)

Intensity of scattered light: $I(\mathbf{q},\omega) \propto S(\mathbf{q},\omega) \qquad S(\mathbf{q},\omega) = \langle \delta\rho(\mathbf{q},\omega)\delta\rho(-\mathbf{q},-\omega) \rangle$

□ Longitudinal modes couple to density fluctuations.

- Sound produces the Brillouin peaks.
- Thermal diffusion produces the Rayleigh peak (coupling of thermal fluctuations to the density through thermal expansion).

Hydrodynamics II: superfluid

- Conserved densities ρ , g_i , e.
- Broken U(1) gauge symmetry

$$\psi(\mathbf{r}) = |\psi(\mathbf{r})|e^{i\theta(\mathbf{r})}, \qquad \mathbf{v}_s = \frac{\hbar}{m}\nabla\theta.$$

Another equation of motion:

$$\partial_t \theta = \mu/\hbar$$
 Josephson relation.

- □ 6 hydrodynamic modes:
 - 2 transverse momentum diffusion modes.
 - 2 longitudinal (first) sound modes.
 - 2 longitudinal second sound modes.
- Central Rayleigh peak splits into two new Brillouin peaks.

Light scattering in a superfluid







FIG. 6. Four Brillouin spectra taken at $P_{\lambda} = 23.1$ bars near the λ transition of liquid ⁴He. In the top spectrum the frequency shifts of first and second sound are marked. For second sound, zero-frequency shift is at the center of the marked free spectral range (149.7 MHz). The instrumental linewidth (3 MHz) is also shown.

Winterling, Holmes & Greytak PRL 1973

Tarvin, Vidal & Greytak 1977

Supersolidity or Quantum Metallurgy?

Solid "hydrodynamics"

- \Box Conserved quantities: ρ , g_i , e.
- □ Broken translation symmetry: u_i , i=1,2,3
- □ Mode counting: 5 conserved densities and 3 broken symmetry variables \Rightarrow 8 hydrodynamic modes. For an isotropic solid (two Lame constants λ and μ):
 - 2 pairs of transverse sound modes (4),
 - 1 pair of longitudinal sound modes (2),
 - 1 thermal diffusion mode (1).
- What's missing? Martin, Parodi, and Pershan (1972): diffusion of vacancies and interstitials.

Supersolid hydrodynamics

- **Conserved quantities:** ρ , g_i , e
- \Box Broken symmetries: u_i , gauge symmetry.
- - 2 pairs of transverse sound modes (4).
 - 1 pair of longitudinal sound modes (2).
 - 1 pair of longitudinal "fourth sound" modes (2).
 - 1 longitudinal thermal diffusion mode.
- Use Andreev & Lifshitz hydrodynamics to derive the structure function (isothermal, isotropic solid). New Brillouin peaks below T_c.

Structure function for supersolid

$$\frac{c_L^4}{D_\Delta\rho_0}\frac{\beta S(\tilde{\mathbf{q}},\tilde{\omega})}{2} \simeq \frac{\left[\delta+\Omega\left(2-\delta\right)\right]\tilde{q}^4}{\left[\tilde{\omega}^2-\left(1+\Omega\right)\tilde{q}^2\right]^2+\left[\left(\delta+2\Omega\right)\tilde{q}^2\tilde{\omega}\right]^2} + \frac{\delta^2\Omega\tilde{q}^4}{\left(\tilde{\omega}^2-\delta\Omega\tilde{q}^2\right)^2+\left[\left(1-2\Omega\right)\tilde{q}^2\tilde{\omega}\right]^2} + \frac{\tilde{q}^2\left[\delta+2\Omega\left(2-\delta\right)\right]\left(\tilde{\omega}^2-\delta\Omega\tilde{q}^2\right)}{\left(\tilde{\omega}^2-\delta\Omega\tilde{q}^2\right)^2+\left[\left(1-2\Omega\right)\tilde{q}^2\tilde{\omega}\right]^2},$$

 $\tilde{\omega} = D_{\Delta}\omega/c_L^2, \ \tilde{q} = D_{\Delta}q/c_L, \ \Omega = \rho_{s0}/\rho_0, \ \delta = 1/(\rho_0\chi c_L^2).$



Supersolidity or Quantum Metallurgy?

Supersolid Lagrangian

- Lagrangian
 - Reversible dynamics for the phase and lattice displacement fields
 - Lagrangian coordinates R_i , Eulerian coordinates x_i , deformation tensor $R_{ij} = \partial R_j / \partial x_i$
 - Respect symmetries (conservation laws): rotational symmetry, Galilean invariance, gauge symmetry.

$$\mathcal{L}_{SS} = -\rho \partial_t \phi - \frac{1}{2} \rho_s (\nabla \phi)^2 + \frac{1}{2} \rho_n v_n^2 - f(\rho, \rho_s, T, R_{ij}) - \rho_n \mathbf{v}_n \cdot \nabla \phi$$

$$\rho = \rho_n + \rho_s, \qquad v_{ni} = -R_{ij}^{-1}\partial_t R_j$$

- Reproduces Andreev-Lifshitz hydrodynamics. Agrees with recent work by Son (2005) [disagrees with Josserand (2007), Ye (2007)].
- Good starting point for studying vortex dynamics in supersolids (Yoo and Dorsey, unpublished). Question: do vortices in supersolids behave differently than in superfluids?

Effective Euclidean Action I

We start from the Lagrangian density of non-dissipative Andreev and Lifshitz hydrodynamics (Yoo & ATD, PRB 2010)

- For simplicity, consider 2-dim isotropic supersolids
- Expand up to quadratic order and integrate out the density fluctuations

$$S_{\rm E}[\phi, \mathbf{u}] = \frac{1}{2} \int d\tau \int d^2x \left[\underbrace{2i\rho\partial_{\tau}\phi + 2\lambda_{ij}u_{ij}}_{\text{linear terms}} + \underbrace{\rho^2\chi(\partial_{\tau}\phi)^2 + \rho_{\rm s}(\partial_i\phi)^2}_{\text{superfluid}} + \underbrace{\rho_{\rm n}(\partial_{\tau}u_i)^2 + (\tilde{\lambda} - \rho^2\gamma^2\chi)u_{ii}^2 + 2\tilde{\mu}u_{ij}^2}_{\text{lattice}} + i(\rho_{\rm n} - \rho^2\gamma\chi)(\partial_{\tau}u_i\partial_i\phi + u_{ii}\partial_{\tau}\phi) \right]_{\text{coupling}}$$

Supersolidity or Quantum Metallurgy?

Effective Euclidean Action II

Incorporate vortices and edge dislocations

$$\phi^{\rm V} = \frac{\hbar}{m} \sum_{\alpha} e^{\alpha} \arctan\left[\frac{y - y_{\rm V}^{\alpha}(\tau)}{x - x_{\rm V}^{\alpha}(\tau)}\right], \qquad u_i^{\rm D} = \sum_{\bar{\alpha}} \frac{b_i^{\bar{\alpha}}}{2\pi} \arctan\left[\frac{y - y_{\rm D}^{\bar{\alpha}}(\tau)}{x - x_{\rm D}^{\bar{\alpha}}(\tau)}\right]$$

Integrate out the degrees of freedom from the superfluid $\phi^{\rm S}$ and the lattice ${\bf u}^{\rm S}$

 $S_{\rm E}[\mathbf{x}_{\rm V}(\tau), \mathbf{x}_{\rm D}(\tau)] = S_{\rm vortex}[\mathbf{x}_{\rm V}(\tau)] + S_{\rm dislocation}[\mathbf{x}_{\rm D}(\tau)] + S_{\rm coupling}[\mathbf{x}_{\rm V}(\tau), \mathbf{x}_{\rm D}(\tau)]$

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Vortex Dynamics I

Action for vortices: $S_{\text{vortex}}[\mathbf{x}_{V}(\tau)] = S_{\text{Magnus}}[\mathbf{x}_{V}(\tau)] + S_{V}[\mathbf{x}_{V}(\tau)]$ $S_{\text{Magnus}} = -\frac{2i\pi\hbar\rho}{m}\sum \mathrm{e}^{lpha}\int d au \frac{dx_{\mathrm{V}}^{lpha}(au)}{d au}y_{\mathrm{V}}^{lpha}(au)$ $S_{\rm V} = -\frac{\pi\hbar^2(\rho_{\rm n} - \rho^2\chi\gamma)^2}{8m^2\rho_{\rm n}c_T}\sum_{\alpha\beta} e^{\alpha}e^{\beta}\int d\tau \int d\tau' \frac{c_T^2}{\sqrt{c_T^2(\tau - \tau')^2 + |\mathbf{x}_{\rm V}^{\alpha}(\tau) - \mathbf{x}_{\rm V}^{\beta}(\tau')|^2}}$ coupling to the transverse sound modes $+\frac{\pi\hbar^2 A_L}{2m^2\rho_{\mathrm{n}}c_L}\sum_{\alpha\beta}\mathrm{e}^{\alpha}\mathrm{e}^{\beta}\int d\tau\int d\tau' \frac{s_i^{\alpha}(\tau)s_i^{\beta}(\tau')+c_L^2}{\sqrt{c_L^2(\tau-\tau')^2+|\mathbf{x}_{\mathrm{V}}^{\alpha}(\tau)-\mathbf{x}_{\mathrm{V}}^{\beta}(\tau')|^2}}$ coupling to the longitudinal first sound modes $+\frac{\pi\hbar^2 A_2}{2m^2\rho_{\mathrm{n}}c_2}\sum_{\alpha\beta}\mathrm{e}^{\alpha}\mathrm{e}^{\beta}\int d\tau\int d\tau' \frac{s_i^{\alpha}(\tau)s_i^{\rho}(\tau')+c_2^2}{\sqrt{c_2^2(\tau-\tau')^2+|\mathbf{x}_{\mathrm{V}}^{\alpha}(\tau)-\mathbf{x}_{\mathrm{V}}^{\beta}(\tau')|^2}},$ Eckern-Schimd action (1989) for vortices in superfluid

Similarly, action for edge dislocations

 $S_{\text{dislocation}}[\mathbf{x}_{\mathrm{D}}(\tau)] = S_{\mathrm{P-K}}[\mathbf{x}_{\mathrm{D}}(\tau)] + S_{\mathrm{D}}[\mathbf{x}_{\mathrm{D}}(\tau)]$

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Vortex Dynamics II

D For small frequencies $c_{T,L,2}^2(\tau - \tau')^2 \gg |\mathbf{x}_V^{\alpha}(\tau) - \mathbf{x}_V^{\beta}(\tau')|^2$

$$S_{\text{vortex}} \simeq S_{\text{static}} + \int \frac{d\omega}{2\pi} \left[-2\pi\rho\omega x_{\text{V}}(\omega)y_{\text{V}}(-\omega) + \frac{1}{2}M_{\text{V}}(\omega)\omega^2 |\mathbf{x}(\omega)|^2 \right]$$

$$M_{\rm V}(\omega) = \underbrace{M_{c_T}(\omega) + M_{c_L}(\omega)}_{+M_{c_2}(\omega)} + M_{c_2}(\omega)$$

due to coupling with elasticity

$$M_{\rm V}(\omega) \simeq \frac{3\pi\hbar^2(\rho_{\rm n}-\rho^2\chi\gamma)^2}{4m^2\rho_{\rm n}c_T^2} - \frac{\pi\hbar^2}{m^2} \left[\rho^2\chi + \frac{1}{4}(\rho_{\rm n}-\rho^2\chi\gamma)^2\left(\frac{1}{\tilde{\mu}} + \frac{1}{\lambda-\rho^2\chi\gamma^2}\right)\right] \left[\tilde{\gamma} + \frac{3}{2} + \ln(|\omega|\epsilon)\right]$$

□ Similarly, for dislocations

$$M_{ij}^{\mathrm{D}} \simeq 4\pi^{3} \ln(|\omega|\epsilon) \left\{ \frac{2b_{i}b_{j}}{\rho_{\mathrm{s}}(\tilde{\Lambda}+2\tilde{\mu})^{2}} \left[\rho_{\mathrm{s}}\rho_{\mathrm{n}}(\tilde{\Lambda}+\tilde{\mu})^{2} + \frac{(\rho_{\mathrm{n}}-\rho^{2}\chi\gamma)^{2}}{4}\tilde{\Lambda}^{2} \right] -\delta_{ij}b_{k}b_{k} \left[2\rho_{\mathrm{n}} + \frac{\rho_{\mathrm{n}}(\tilde{\Lambda}^{2}-\tilde{\mu}^{2})}{(\tilde{\Lambda}+2\tilde{\mu})^{2}} + \frac{(\rho_{\mathrm{n}}-\rho^{2}\chi\gamma)^{2}}{2\rho_{\mathrm{s}}}\frac{\tilde{\Lambda}^{2}+2\tilde{\mu}^{2}}{(\tilde{\Lambda}+2\tilde{\mu})^{2}} \right] \right\}$$

Supersolidity or Quantum Metallurgy?

Summary

- Dissipation peak in the TO response is well described by a simple viscoelastic model. A long time scale is identified, probably from dislocation physics.
- The viscoelastic model only accounts for about 10% period shift. Is the rest of the "spectral weight" at zero frequency? Is there a superfluid response?
- Specific heat feature appears to have a natural interpretation as a Schottky anomaly due to evaporation of ³He impurities from dislocations. Is the binding energy related to the Arrhenius behavior of the viscoelastic model?

Experiments (circa 1992)

Review: M. W. Meisel, Physica B 178, 121 (1992).

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M.W. Meisel / Supersolid ⁴He

Table 1 Summary of experimental searches for supersolid ⁴He.

Group	Method	³ He/ ⁴ He concentration	Temperature range	Pressure	Conclusion
Andreev et al. (1969) [23]	Plastic flow	Unstated	$T_{\rm min} = 500 {\rm mK}$	25–154 bar	$\frac{\frac{\rho_{\rm s}}{\rho} < 1 \times 10^{-3}}{v_{\rm c} < 2 \times 10^{-7}} {\rm cm/s}$
Suzuki (1973) [24]	Plastic flow	Unstated	1.38–2.12 K	29-51 bar	$v_{\rm c} < 10^{-4} {\rm cm/s}$ $v_{\rm c} < 10^{-4} {\rm cm/s}$
Tsymbalenko (1976) [25]	Plastic flow	Unstated	0.6–2.1 K	2540 bar	$v_{\rm c} < 5 \times 10^{-10} {\rm cm/s}$
Dyumin et al. (1989) [28]	Plastic flow	Unstated	1.0–1.4 K	On the melting curve	$v_{\rm c} < 10^{-9} {\rm cm/s}$
Greywall (1977) [26]	ΔP across weak-link	Unstated	$T_{\rm min} = 30 \ {\rm mK}$	25-50 bar	$\frac{\rho_{\rm s}}{\rho} v_{\rm c} < 2.5 \times 10^{-9} {\rm cm/s}$
Bishop et al. (1981) [27]	Torsional oscillator	$0.3 - 411 \times 10^{-6}$	$T_{\rm min} = 25 {\rm mK}$	25-48 bar	$\frac{\rho_{\rm s}}{\rho} < 5 \times 10^{-6}$ $v_{\rm c} < 5 \times 10^{-8}$ cm/s
Bonfait et al. (1989) [29]	Cylindric "U"-tube	Standard commercial ⁴He	$T_{\rm min} = 4 \ {\rm mK}$	On the melting curve	$\frac{\rho_{\rm s}}{\rho} v_{\rm c} < 1.8 \times 10^{-9} \rm cm/s$
Adams et al. (1990) [31]	$P_{\rm v}(T)$	~10 ⁻⁶	$T_{\rm min} = 1 {\rm mK}$	26 bar	$\frac{\rho_{\rm s}}{\rho} < 10^{-5}$
van de Haar et al. (1991) [32]	$P_{v}(T)$	~10 ^{−7} (1)	1.5–120 mK	On the melting curve and 26 bar	$\frac{\rho_{\rm s}}{\rho} < 2 \times 10^{-5} \text{ at } 26 \text{ bar}$ $<6 \times 10^{-7} \text{ on melting}$ curve
Lengua and Goodkind (1990) [30]	Ultrasound	1.5×10^{-9}	30 mK-1.0 K	~25 bar	$T_{\rm c} = 117 {\rm mK}(?)$ $\frac{\rho_{\rm s}}{\rho} \sim 10^{-3}(?)$

The initial attempts by Andreev et al. [23], Suzuki [24] and Tsymbalenko [25] and a recent effort by Dyumin et al. [28] to observe a supersolid ⁴He state involved the study of plastic flow in crystals in which physical objects were moved. Later, techniques more reminiscent of superfluid helium studies were employed. Greywall [26] attempted to detect mass flow through a weak link which was subjected to a chemical potential difference between two mass reservoirs. Bishop et al. [27] used a sensitive torsional oscillator to search for a change of the moment of inertia of the system. Bonfait et al. [29] searched for mass flow in a cylindric "U"-tube experiment. Although some of the experiments [24, 25, 27, 28, 57] were able to study dislocations in the crystal and other effects, none of the investigations resulted in a positive identification of the supersolid state. The experimental conditions and results are summarized in table 1.

There are always a number of possible explanations for a null result; however, the most obvious explanation for the results of the aforementioned experiments involves the critical temperature, T_c , and the critical velocity, v_c . Either the work was not performed at temperatures below T_c , or the mass transport was being driven by a velocity field higher than v_c . Although Bonfait et al. [29] argue that their ex-

searches. As emphasized above, thermodynamic studies, which avoid problems associated with the critical velocity, are preferable to ones which are intrinsically sensitive to v_e . Finally, the experimentalists should prepare for the extension of the temperature range down to the sub-millikelvin region where new challenges in cooling the samples and in measuring the temperature of the system will be encountered.

Supersolidity or Quantum Metallurgy?

Details

- □ Resonant period of oscillation is $\tau = 2\pi \sqrt{I/\alpha}$
- Changes in the period can be due to either I or α .
- Pressures ranged from 26 to 66 bars.
- Decoupling observed below 230 mK.
- Amplitude of about 1 nm, maximum velocities of about 10 μ m/s.
- Frequency: 1 kHz (period of 10⁶ ns). Period shifts of order 40 ns. Q of order 10⁶.
- Cell size: 10 mm OD, annulus 0.63 mm width, 5 mm height.
- Barrier inserted: no effect.
- □ 400 ppm of ³He quenches effect.

