

Classical Rotational Inertia of Solid ^4He

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The observation of reduced rotational inertia in a cell containing solid ^4He has been interpreted as evidence for superfluidity of the solid. We propose an alternative explanation: slippage of the solid, due to grain boundary premelting between the solid and dense adsorbed layers at the container wall. We calculate the range of film thickness, and determine the viscosity that will account for the missing rotational inertia. Grain boundary premelting also explains inertial anomalies in an earlier study of solid helium in porous glass and indicates that the liquid is partially superfluid.

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A recent paper by Kim and Chan [1] describes the observation of inertial anomalies, termed “nonclassical rotational inertial fraction (NCRIF)” in solid ^4He , which are taken to demonstrate superfluidity of the solid. The exciting possibilities raised by the experiment and their earlier study [2] wherein ^4He was solidified in Vycor, a microporous glass, span a range of fundamental issues in quantum materials. Leggett notes [3] the possibility of supersolid behavior as a long-standing speculation, based on the hypothesis of Bose-Einstein condensation of zero-point vacancies, but Prokof'ev and Svistunov argue [4] that the vacancy density in well ordered solid ^4He is insufficient for Bose-Einstein condensation at the experimental temperatures, speculating instead that the NCRIF in the Vycor study [2] may be due to the superfluidity of vacancies in a defect-laden layer of solid ^4He at the Vycor surfaces. Finally, Beamish [5] noted the possibility that a disordered thin liquidlike layer at the Vycor walls may persist at low temperature and argued that its superfluid properties would be different from those found at pressures well below solidification pressure. Indeed, Khairallah and Ceperley show [6], by path integral Monte Carlo simulation, that a *liquid* film exists between solid ^4He and a Vycor wall, and that the superfluidity of the film can account for the missing inertia in [2].

Here we suggest an alternative explanation of the experimental NCRIF: slippage of the solid at the wall of the container due to a liquid film caused by grain boundary premelting. Grain boundary premelting is a common feature of virtually all crystalline solids, and it has been observed in solid ^4He , in the high pressure hcp phase [7]. The premelting in the case discussed here is not at *ordinary* grain boundaries, but at the interface between the bulk solid and *dense adsorbed layers* at the container wall. The dense layers, due to strong adsorption forces, are responsible for nonzero wetting angles between solid ^4He and copper and glass walls [8,9]; rather than an ordinary grain boundary, the contacting surface in question more nearly resembles the interface between two different materials. The liquid film separates the bulk solid from the torsional balance and replaces the shear strength of the dry

interface by viscous drag. Our model allows the possibility that the film is superfluid, in which case the calculation is equally applicable to its normal fluid component. Indeed, when we apply our model to the earlier experiment of the solid in Vycor, we conclude that the film is partially superfluid. In the following we first estimate the thickness of the premelted film, and then calculate the dynamical coupling.

The nature of premelting in any system is determined by the competition between bulk and surface free energies. *Complete premelting*, in which the thickness of the melt layer diverges as temperature approaches the bulk transition, requires that the total excess surface free energy per unit area, $F(d)$, be a positive monotonically decreasing function of the film thickness with a global minimum at infinite film thickness. In *incomplete premelting*, the melt thickness remains finite at the bulk transition, such as in recent studies of ice [10]. It is a general result [11–13] that in a symmetrical system (e.g., solid/liquid/solid) the long range interactions are attractive, and consequently grain boundary melting must be incomplete. The most complete formulation of the excess surface free energy per unit area, $F(d)$, for systems *entirely* controlled by frequency dependent dispersion forces is that of Dyzaleshinskii, Lifshitz, and Pitaevskii (DLP) [14]. However, it requires as input the frequency dependent dielectric properties of the layers in the system under consideration. Because we are dealing with bulk solid ^4He and the dense adsorbed solid at the container wall, the input data for the DLP theory are not available. Therefore, we proceed with reasonable ranges of the Lennard-Jones parameters [15,16].

The total free energy of the system at a given temperature T and pressure P is written

$$G_T(T, P, d) = \rho_\ell \mu_\ell(T, P)d + F(d), \quad (1)$$

where the liquid density and chemical potential are ρ_ℓ and $\mu_\ell(T, P)$. In grain boundary premelting

$$F(d) \equiv 2\gamma_{s\ell} + \rho_\ell \int_d^\infty V(z)dz, \quad (2)$$

where $F(d)$ is the total excess interfacial free energy of the solid-solid interface, and $\gamma_{s\ell}$ is the interfacial free energy

per unit area of the solid-liquid interfaces, with implicit reference to the crystallographic orientation present at an interface. In lieu of the DLP theory, the most general phenomenological mean field model considers $V(z)$ as the Lennard-Jones potential [15] but augmented to embody the effects of retardation *vis-à-vis*

$$V(z) = \frac{4C_3^3}{27D^2} \frac{1}{z^9} - \frac{C_3}{z^3} - \frac{B}{z^4}, \quad (3)$$

where z is the coordinate normal to the surfaces, $C_3(B)$ is the nonretarded (retarded) van der Waals attraction, and D is the well depth. At each temperature and pressure below T_λ the bulk and interfacial free energies strike a balance, and one can show, e.g., [12,17], that a unique equilibrium film thickness obtains from

$$\frac{1}{\rho_\ell} \frac{\partial F(d)}{\partial d} = -q_m \frac{T_\lambda - T}{T} \equiv -q_m t, \quad (4)$$

where the latent heat of fusion is q_m and t is the reduced temperature. From this we can simply write the equilibrium film thickness-temperature relation as $t = q_m^{-1} V(d)$. Because $q_m = q_m(t)$ such that $q_m \rightarrow 0$ as $T \rightarrow 0$ ($t \rightarrow 1$), we find the latent heat to be a key function in the problem: It embodies the bulk free energy penalty for converting some solid to liquid, against the melt driving interactions of the repulsive part of the potential. The continuous temperature dependence of q_m is extrapolated to 0 K using the data of Swenson [18]. In Fig. 1 we plot $d = d(t)$ for a range of potential parameters suggested from the detailed analysis of the wetting of wide classes of substrates by liquid helium [15]. We find that although the magnitude of the thickness of a premelted layer depends on the parameters used in the calculation, (a) the temperature dependence itself is not monotonic as found in the usual circumstance with constant q_m and (b) the film thicknesses (≈ 4 –8 atomic layers) represent a conservative range given the uncertainty in the parameters of the potential [14], and yet (c) are sufficient to accommodate flow and/or superflow (e.g., [19,20]).

We now estimate the dynamical coupling of the solid to the container, via viscous drag due to the normal fluid component of the premelted layer. Although the bulk liquid may be completely superfluid at the experimental temperatures and pressures [21], there is appreciable depletion of superfluid at the walls of the narrow gap [22], which produces a normal fluid fraction ρ_n/ρ_ℓ with viscosity η . The solid is a thin walled hollow cylinder, of mean radius a , wall thickness s , and height h , bathed on both sides by a layer of liquid thickness d . The liquid is driven by the container's torsional oscillations of frequency ω and of angular displacement $\theta(t) = \theta_o e^{i\omega t}$, driving the solid at the same frequency, and angular amplitude θ'_o . The hydrodynamic regime [23–25] is governed by the relative magnitudes of d and the decay length of transverse viscous waves $\lambda = \sqrt{2\eta/\rho_n\omega}$. We provisionally assume that λ is much greater than the gap width d , which simplifies the regime to

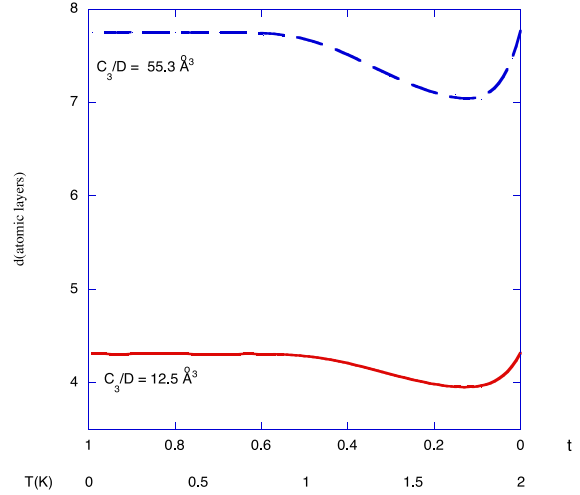


FIG. 1 (color online). Plots of liquid ${}^4\text{He}$ film thickness as a function of temperature $T(\text{K})$ and reduced temperature, $t \equiv (T_\lambda - T)/T$, and values of the Lennard-Jones potential parameters C_3/D chosen for expected ranges [14]. Because retardation typically occurs at ranges that are much larger than is found, we let $B = 0$. The data from Swenson [20] for $q_m = q_m(t)$ are extrapolated to absolute zero. At long range *any* grain boundary film will not diverge under this potential because the long ranged contributions are attractive under dispersion forces, and hence the melting is incomplete. A particular novelty of this situation is that the film thickness does not vary monotonically with temperature.

one of nearly steady flow. The ratio of d to the cylinder radius a ; $d/a \ll 1$, provides a second simplification that makes the problem equivalent to the drag between parallel plates. Therefore, the fluid velocity varies linearly between the surface of the cell and the solid helium, so that the viscous drag per unit area on the solid helium is $f = (a\omega\eta/d)(\theta_o - \theta'_o)e^{i\omega t}$. The torque \mathcal{T}_T due to the total force on both inner and outer surfaces of the solid cylinder is

$$\mathcal{T}_T = \frac{4\pi a^3 h \omega \eta}{d} (\theta_o - \theta'_o) e^{i\omega t}. \quad (5)$$

The torque induces the solid's inertial response, the time rate of change of angular momentum: $\dot{\mathbf{L}}_s = I\omega^2 \theta'_o e^{i\omega t}$, where $I = 2\pi a^3 h s \rho_{\text{sol}}$, in which we emphasize that ρ_{sol} is the density of the solid to avoid confusion with the conventional nomenclature for the superfluid density. Thus, the fractional difference in the amplitudes of rotational motion between the bulk solid and the premelted film is written as $\frac{\theta_o - \theta'_o}{\theta'_o} \equiv \delta$, wherein the controlling parameter is the value of δ , which is written as follows:

$$\delta = \frac{ds}{\lambda^2} \frac{\rho_{\text{sol}}}{\rho_\ell} = \frac{ds\omega\rho_{\text{sol}}}{2\eta}. \quad (6)$$

We now estimate δ and equate this to the experimental values of NCRIF.

The experimental values for $\rho_{\text{sol}} = 0.2 \text{ g cm}^{-3}$ [18], $s = 0.063 \text{ cm}$, $\omega = 6.5 \times 10^3 \text{ s}^{-1}$ [1], and the theoretical values for d of 4–8 atomic layers are used. Since the viscosity of the premelted liquid is not known, we estimate it by applying the theoretical slippage to the experimental NCRIF. For the maximum NCRIF ≈ 0.02 and the minimum theoretical gap of 4 atomic layers we obtain an effective viscosity $\eta \approx 300 \mu\text{P}$; for the larger gap, $\eta \approx 600 \mu\text{P}$. These values yield a penetration depth between 10 and 20 μm , validating the assumption that $\lambda \gg d$. The range of the effective viscosity is considerably greater than is typical of normal fluid at saturated vapor pressure [24]. Two distinct mechanisms, both due to the narrow gap, may account for the increase: (i) the healing length of superfluid depletion is on the order of a few molecular distances [22], and (ii) the effective viscosity of the liquid within the depletion zone may be subject to the reduced fluidity that occurs for all liquids in narrow channels [25]. The depleted superfluid evidently acts as a Newtonian fluid with a viscosity consistent with liquid ^4He , and enhanced by confinement. Within this framework, the estimated slippage is consistent with the range of observed NCRIF.

Additional observations of Kim and Chan are that the NCRIF decreases at higher amplitudes of oscillation, which is given as strong supporting evidence of superfluid solid behavior, attributing it to exceeding the critical velocity of superfluidity in the solid [1]. We suggest that it can, indeed, be evidence of superfluidity, but in the liquid film rather than in the solid. The slippage is controlled, at low relative speeds, by the viscosity of the normal fluid component and is augmented, at higher speed, by the excitation of the superfluid fraction.

An experiment to test whether NCRIF is due to solid superfluidity or to slippage of bulk solid that is superfluid could be readily made by the insertion of a ridge in the cell wall. An appropriate ridge dimension would tend to lock the solid but would not greatly restrict the superflow of vacancies.

The results of the earlier work with solid helium in Vycor [2] are relevant here. Because of pinning of the solid by the matrix, the loss of rotational inertia indicates superfluidity. It may be, as Prokof'ev and Svistunov [4] and Beamish [5] suggest, in a disordered solid layer at the Vycor interface. But it can be expected that grain boundary melting occurs in the Vycor study, just as in the experiment with bulk solid. Thus the superfluidity would be associated with a liquid film, rather than the solid. The pore geometry is described as 7 nm diameter cylinders, multiply connected with a tortuosity factor of 4 [2]. We assume that the pore surfaces are plated with 2 dense layers of solid ^4He , and that the grain boundary between the plating and the solid is wetted by a partially superfluid premelted film. The missing inertia depends on the product of thickness of premelt and its superfluid fraction; if the thickness is four atomic layers, the observed missing inertia can be accounted for by a superfluid fraction of about 12%.

Khairallah and Ceperley's demonstration of *liquidity* of the interface is consistent with such a scenario [6].

Finally, although we propose an alternative explanation to a superfluid solid, we consider that decoupling due to a premelted superfluid film offers interesting possibilities. The conjectured superfluid exists in a region of pressure and temperature not otherwise accessible, and the experiment suggests interesting and entirely new possibilities in studies of premelting and liquid confinement.

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