Classical rotational inertia of solid $^4$He

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The observation of reduced rotational inertia in a cell containing solid $^4$He has been interpreted as evidence for superfluidity of the solid. An alternative explanation is slippage of the solid at the container wall due to grain boundary premelting between the solid and dense adsorbed layers at the container wall. We calculate the range of film thickness and the viscous drag, and find that the slippage can account for the observations.

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A recent report by Kim and Chan$^1$ describes the observation of inertial anomalies, termed “non classical rotational inertial fraction (NCRIF)” in solid $^4$He, which are taken to demonstrate superfluidity of the solid. The exciting possibilities raised by the experiment and their earlier study$^2$ wherein $^4$He 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, and Prokof’ev and Svistunov argue$^4$ that the vacancy density in well ordered solid $^4$He 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 $^4$He at the Vycor surfaces. Finally, Beamish$^5$ noted the possibility that a disordered thin liquid like 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.

Here we suggest an alternative explanation of the experiment on bulk solid$^1$: slippage of the solid at the wall of the container due to a liquid film caused by grain boundary premelting. The premelting in this case 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 $^4$He and copper and glass walls$^6$; 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. 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$^8$. It is a general result$^9$-11 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 Dzyaloshinskii, Lifshitz and Pitaevskii (DLP)$^{12}$. 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 $^4$He 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$^{13}$.

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

$$G_T(T, P, d) = \rho_t \mu(T, P)d + F(d), \quad (1)$$

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

$$F(d) \equiv \gamma_{ss}(d) = 2\gamma_{st} + \rho_t \int_d^\infty V(z)dz, \quad (2)$$

where $\gamma_{ss}(d)$ is the interfacial free energy of the solid–solid interface, and $\gamma_{st}$ 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$^{13}$ but augmented to embody the effects of retardation viz a viz

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

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 $T_3$ the bulk and interfacial free energies strike a balance and one can show [e.g., 10, 12] that a unique equilibrium film thickness obtains from

$$\frac{1}{\rho_e} \frac{\partial F(d)}{\partial d} = -q_m \frac{T_3 - T}{T} \equiv -q_m t,$$  \hspace{1cm} (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 = \frac{q_m}{1} V(d).

In the figure 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 [13]. Curves for three values of the latent heat of fusion $q_m$ are shown and the value at the NCRIF onset temperature of 175 mK is extrapolated to lower temperatures from 1 K using the data of Swenson [14]. Because $t = q_m^{-1} V(d)$, we find $q_m$ to be the most important parameter in the problem: The latent heat embodies the bulk free energy penalty for converting solid to liquid, against the melt pressure and temperatures below about 1K, leaves open important quantitative questions. Thus, we view our film thickness calculations as conservative–thinner than is likely. 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 rather weak and (b) the film thicknesses (1-4 atomic layers) are sufficient to accommodate flow and superflow (e.g., 14, 15).

We now estimate the dynamical coupling of the solid to the container, through the premelted layer. 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 angular displacement $\theta(t) = \theta_0 e^{i\omega t}$, driving the solid at the same frequency, and angular amplitude $\theta_0$. The hydrodynamic regime is governed by the relative magnitudes of $d$ and the decay length of transverse viscous waves $\lambda = \sqrt{2\eta/\rho_0 \omega}$, where the dynamic viscosity is $\eta$ and the density of the liquid $\rho_e$. To evaluate $\lambda$ we have the experimental frequency $\omega = 2.3 \times 10^3$ s$^{-1}$ and the liquid density $\rho_s \approx 0.2 g/cm^3$ in the range of experimental pressures. Since the viscosity of the premelted liquid is not known, we must estimate it from the properties of the bulk liquid. The viscosity of normal liquid HeI is about $20 \mu P$; the viscosity of the normal component of HeII between 1.2 K and $T_3$ ranges between 10$\mu P$ and 20$\mu P$ [20]. However, the viscosity may be enhanced in the very narrow gap $d$ [18], thereby placing an uncertainty on the experimental value of $\eta$. As a conservative measure, we will allow a possible enhancement of an order of magnitude. Accordingly, we estimate $2 \times 10^{-4} < \lambda < 7 \times 10^{-4}$ cm, which is on the order of one thousand times the thickness $d$ of the grain boundary melted liquid, thereby reducing the problem to one of slow, nearly steady flow. A second simplification stems from the ratio of $d$ to the cylinder radius $a$; $d/a << 1$, which 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 $f = (\omega \eta / d)(\theta_0 - \theta_0') e^{i\omega t}$. The torque $T_T$ due to the total force on both inner and outer surfaces of the solid cylinder is

$$T_T = \frac{4\pi a^3 h \eta}{d} (\theta_0 - \theta_0') e^{i\omega t}.$$  \hspace{1cm} (5)

The torque induces the solid’s inertial response, the time rate of change of angular momentum: $L_s = I \omega^2 \theta_0 e^{i\omega t}$, where $I = 2\pi a^3 h \rho_s e$ in which we emphasize that $\rho_s$ 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 $\frac{\theta_0 - \theta_0'}{\theta_0} = \delta$, and hence the fractional inertial response is $\frac{\theta_0 - \theta_0'}{\theta_0} = \frac{\delta}{1+\delta}$. Wherein the controlling parameter is the value of $\delta$ which is written as follows

$$\delta = \frac{ds}{\chi^2 \rho_s / \rho_e}.$$  \hspace{1cm} (6)

The ranges of possible values of $d$ and $\eta$ impose wide limits on the estimate of slippage: 0.004 $< \delta < 0.26$. These extremes span the maximum values of the NCRIF measured at low temperature and low amplitude, roughly 0.005 to 0.02, depending on the pressure [11].

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 propose 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.

The results of the earlier work with Vycor [2] are relevant here. Because of pinning of the solid by the Vycor matrix [2], the loss of rotational inertia is interpreted as an indication of superfluidity. It may be, as Prokof’ev and Svistunov [3] 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, as in the experiment with bulk solid. Thus the superfluidity would be associated with the liquid film, rather than the solid. Indeed, the nm scale disorder of Vycor was used in order to enhance the density of vacancies [2], and depending on pressure and the assumed fraction of material locked by the tortuosity of the Vycor, the estimate of the BEC fraction could be less than that in the bulk solid. In the framework of our theory, the difference between the experiments may be explained in
terms of the wall materials inducing a different density of adsorbed solid. Interesting tests that span the existing range of behavior would involve the insertion of a ridge in the cell wall of the most recent experiment [1] or changing the wall material. An appropriate ridge dimension would lock the solid and thereby offer a possible test of whether slippage had explained the NCRIF. An additional observation is an extreme sensitivity of the NCRIF to 

Finally, although we propose an alternative explanation to a superfluid solid, we consider that decoupling due to a premelted, and possibly superfluid, film offers interesting possibilities. The film 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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