

Pressure-Driven Flow of Solid Helium

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The recent torsional oscillator results of Kim and Chan show an anomalous mass decoupling, interpreted by the authors as a supersolid phase transition, in solid ^4He . We have used a piezoelectrically driven diaphragm to study the flow of solid helium through an array of capillaries. Our measurements showed no indication of low temperature flow, placing stringent restrictions on supersolid flow in response to a pressure difference. The average flow speed at low temperatures was less than 1.2×10^{-14} m/s, corresponding to a supersolid velocity at least 7 orders of magnitude smaller than the critical velocities inferred from the torsional oscillator measurements.

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Recent experiments by Kim and Chan [1,2] showed that solid helium decouples from a torsional oscillator at temperatures below about 0.2 K. In liquid ^4He , such decoupling reflects the nonclassical rotational inertia (NCRI) associated with superfluidity and these experiments suggest that ^4He also exhibits “supersolidity.” The possibility of supersolidity in helium has been discussed for many years [3–5] but previous experimental searches [6–8] were unsuccessful. Following Kim and Chan’s experiments, a number of papers have discussed the possible microscopic origins of supersolidity [9–16] and the properties that such a state might exhibit [12,17]. However, there is not yet a consensus on whether supersolidity can occur in a defect-free crystal and further experiments are needed to establish whether solid helium displays any of the other unusual properties associated with superfluidity. We recently [18] used a capacitive method to look for pressure-driven flow of solid helium confined in the pores of vycor glass, but saw no evidence of superflow at temperatures down to 30 mK, nor has supersolidity been seen in recent ultrasonic experiments in vycor [19]. In this Letter, we report measurements of dc and low frequency ac flow of solid ^4He through an array of glass capillaries. Near the melting temperature, applying a pressure difference caused solid helium to flow through the capillaries, but the rate decreased with temperature; below about 1 K no flow was detected. Our experiments extended to 35 mK, well into the temperature range where Kim and Chan observed NCRI, and used isotopically purified ^4He . Our results place stringent limits on possible pressure-induced supersolid flow.

The essential results of the torsional oscillator measurements were similar for ^4He confined in the nanometer pores of vycor glass [1] and for bulk ^4He [2]. Each showed a gradual transition at $T_c \approx 0.2$ K with about 1% of the helium (the “supersolid fraction” ρ_s/ρ) decoupling at the lowest temperatures and amplitudes. The decoupling was smaller at large oscillation amplitudes, suggesting a supersolid critical velocity $v_c \sim 10 \mu\text{m/s}$ in both systems. The similarities support the interpretation that NCRI is an

intrinsic property of solid helium rather than, for example, occurring in a liquid layer at pore surfaces. The measurements in vycor revealed a remarkable sensitivity to ^3He impurities; concentrations as low as 10 ppm significantly reduced the NCRI. In bulk ^4He , ρ_s was shown to vary with pressure, going through a maximum around 55 bar [20]. However, T_c was nearly pressure independent.

Early suggestions [3] that ^4He could exhibit supersolidity were based on the idea that quantum solids might contain “zero point vacancies” (ZPV) which would Bose condense and produce supersolidity. However, both measurements [5] and calculations [21] indicate that vacancies in helium have an activation energy of at least 15 K, with no evidence of vacancies at zero temperature. Nonetheless, direct comparisons of density to lattice constants from x-ray measurements can only rule out ZPV at the 0.1% level and Anderson recently suggested [13] that solid helium may be incommensurate, with vacancies in a highly correlated ground state. Even in the absence of ZPV, Leggett showed [4] that atomic exchange could lead to supersolidity. He estimated $\rho_s/\rho \lesssim 10^{-4}$ for ^4He but subsequent calculations [11,12] have predicted supersolid fractions ranging from 10^{-5} to 1%. However, recent path integral Monte Carlo calculations [14] found that exchange frequencies decrease exponentially with ring length and thus that supersolidity is not expected in a perfect, commensurate ^4He crystal. The conclusion that supersolidity in ^4He must involve defects is supported by very general path integral arguments [9,10]. Vacancy-interstitial pairs (VIP) may be needed for supersolidity [9–11,15] but interstitials in ^4He have large activation energies (around 48 K [14]) and VIP appear to be strongly bound [16] and so cannot transport mass or produce supersolidity. Extended defects such as dislocations, stacking faults, or grain boundaries may be essential.

While there is not yet a consensus on the microscopic origin of supersolidity, the similarity between the NCRI seen for ^4He in the pores of vycor and for bulk ^4He constrains models. For example, it is difficult to imagine mechanisms involving grain boundaries that would not be

affected by confinement in nm pores. Recent calculations [22] for ^4He in vycor-like pores provide evidence of a mobile liquid-like layer near the pore surface where the superfluid response might originate. This could be related to the NCRI's sensitivity to ^3He , since impurities would preferentially go to the delocalized layer and disrupt superfluidity, but it would not explain the bulk helium results.

The long-standing interest in quantum crystals inspired a number of earlier searches for supersolidity in ^4He , although many of them did not reach the temperature range where Kim and Chan observed NCRI. One that did extend to 25 mK saw no decoupling from a torsional oscillator [7], leading the authors to conclude that either the transition temperature was below 25 mK or else the supersolid density or the critical velocity was very small ($\rho_s/\rho < 5 \times 10^{-6}$ or $v_c < 5 \mu\text{m/s}$). There have also been attempts to look for flow of solid helium in capillaries but pressure differences of order 1 bar did not produce measurable flow [6] down to 30 mK, nor was flow seen in a subsequent U-tube experiment [8] which extended to 4 mK. These measurements put similar limits on possible superflow in bulk helium ($\frac{\rho_s}{\rho} v_c \lesssim 2 \times 10^{-11}$ m/s) and our recent experiments [18] put a comparable limit on pressure-induced flow solid of ^4He in the pores of vycor ($\frac{\rho_s}{\rho} v_c \lesssim 1.5 \times 10^{-11}$ m/s). One group of experiments [23–25] that did show unusual behavior involved ultrasound and heat pulses. The interpretation was complicated but, like the observation of NCRI of Kim and Chan, the results were sensitive to ^3He impurities at the ppm level.

Other than the torsional oscillator experiments, there have not yet been direct observations of supersolid behavior either in bulk or in small pores. However, the small critical velocities and the sensitivity to ^3He impurities may affect dc flow or other properties even more strongly than the torsional oscillator measurements. Also, solids have properties not shared by liquids (e.g., a lattice with shear rigidity) and a supersolid may not exhibit all of the effects we associate with superfluidity (e.g., superleaks, persistent currents, thermomechanical effects, quantized vortices, second sound, etc.). Below we describe a set of experiments to look for one such property in solid ^4He : superflow in response to pressure. We applied small pressure differences (3 to 100 mbar) at low temperatures (down to 35 mK) and used both isotopically purified ^4He (^3He concentration < 0.002 ppm [26]) and ^4He with the natural isotopic composition (typically 0.3 ppm ^3He). We made both dc and low frequency ac (below 1 Hz) measurements, but did not see any evidence of flow below about 1 K.

Our beryllium copper cell consisted of two cylindrical chambers connected by a “superleak” of about 36 000 parallel glass capillaries (25 μm in diameter) which were fused into a 3 mm thick “glass capillary array” (GCA) (from Collimated Holes Inc.) [27] with an open cross-sectional area $A = 0.18 \text{ cm}^2$. The outer wall of the larger chamber (diameter 25 mm, height ≈ 1 mm, volume $V_1 =$

0.49 cm^3) included a flexible diaphragm which could be moved with an external piezoelectric actuator (APC International, model PSt 150/10 \times 10 \times 18) [28] to compress the helium. The smaller chamber (diameter 7 mm, height 0.3 mm, $V_2 = 0.01 \text{ cm}^3$) included a capacitive pressure gauge which, when used with a 1 kHz automatic bridge (Andeen-Hagerling 2550A), had a resolution and stability better than 0.2 mbar. If helium moves a distance dx through the capillaries, the resulting pressure change is $dP = \frac{A}{\kappa V_2} dx$, where κ is the helium's compressibility, so we typically could detect a 0.3 nm displacement of solid ^4He through the GCA. The cell, which had a total volume (including the GCA channels and fill line) $V_{\text{total}} = 0.79 \text{ cm}^3$, was mounted on the mixing chamber of a dilution refrigerator. Temperatures were measured with a germanium thermometer, with a ^{60}Co nuclear orientation thermometer for calibration below 50 mK.

We started by filling and pressurizing the cell at 4.2 K, using a room temperature gauge to calibrate our capacitive pressure gauge. We calibrated our actuator and diaphragm in the liquid phase at 1.95 K and 36.4 bar, just below the melting curve. The bottom set of data in Fig. 1 shows the pressure response (right axis) when the full voltage (150 Vdc) was applied to the actuator. As expected, the pressure increased immediately (within the few seconds the capacitance bridge took to respond) and returned to its original value when the diaphragm was released after about half an hour. The pressure change due to the compression was $\Delta P_{\text{liquid}} \approx 84 \text{ mbar}$. Using the liquid's compressibility ($\kappa_{\text{liquid}} = 3.6 \times 10^{-3} \text{ bar}^{-1}$ [29]) gives a volume change $\Delta V/V_{\text{total}} \approx 0.03\%$, corresponding to a diaphragm deflection of about 1 μm .

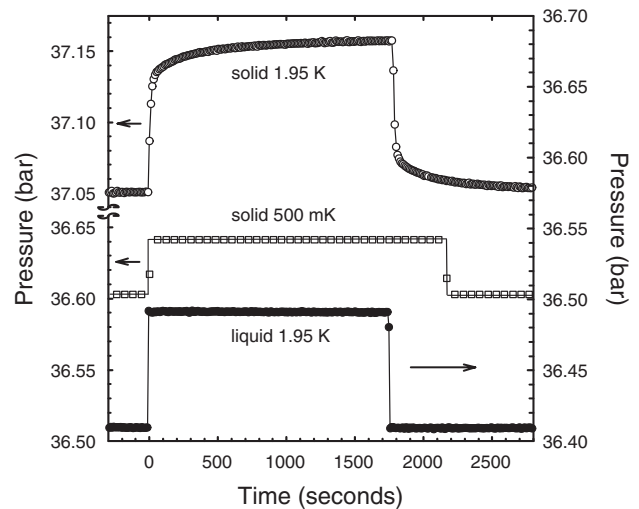


FIG. 1. Pressure response to “squeezes.” Lower curve (solid circles): liquid ^4He at 1.95 K, 36.4 bar. Middle curve (open squares): solid ^4He at 500 mK, 36.6 bar. Upper curve (open circles): solid ^4He near melting at 1.95 K, 37.1 bar. Lines are guides to the eye. Note the different pressure axes.

Crystals were grown using the blocked capillary, constant volume technique. We started with liquid at high pressure and monitored the cell pressure as it was cooled. At a pressure of 61.7 bar, freezing began at 2.60 K and was complete at a final pressure of 37.1 bar. We then annealed the solid by keeping it within 50 mK of its melting temperature for at least 2 hours, thereby eliminating pressure gradients created during freezing and thus producing a sharp melting onset (at $T_m = 1.96$ K) characteristic of a uniform density crystal. Our initial experiments used ^4He with the natural isotopic composition and were consistent with the measurements shown in this Letter which were made using isotopically purified ^4He .

Our basic flow measurement was made at temperatures below T_m by quickly (over about 5 seconds) applying a dc voltage to the actuator to squeeze the solid ^4He , thus increasing the pressure in the large chamber. In contrast to the case where the cell contained liquid, the solid helium may flow through the GCA channels slowly, or not at all, so the pressures in the two chambers may not equilibrate. However, even without flow, some pressure is transmitted to the second chamber, since a pressure difference will cause the GCA plate separating the chambers to flex elastically. This small deflection appears as an immediate pressure step in the other chamber. Any subsequent flow through the channels will further increase the pressure, but more slowly.

The upper two sets of data in Fig. 1 show the response to a pressure step when the cell contains solid helium. At 0.5 K (middle curve) the pressure in the second chamber immediately changed by about 38 mbar, corresponding to the GCA flexing by about 30 nm. Above about half the melting temperature, this initial jump was followed by a slower, temperature-dependent change due to flow. The top curve in Fig. 1 shows the response at 1.95 K, very close to melting. After the initial jump, the pressure continued to increase due to flow of solid through the channels and relaxation of the GCA, but stabilized within about half an hour. The total increase of 105 mbar is slightly larger than the corresponding change with liquid helium, as expected given the solid's smaller compressibility ($\kappa_{\text{solid}} \approx 3.1 \times 10^{-3} \text{ bar}^{-1}$ [29]), and indicates that, near melting, flow through the channels can maintain pressure equilibrium between the two chambers. For all three sets of data, we confirmed the linearity of the response; i.e., the pressure changes were proportional to the voltage applied to the diaphragm actuator.

The most interesting question is whether solid helium will flow through the capillaries in the temperature range where Kim and Chan saw decoupling. Figure 2 compares the pressure response at 35 mK to that at 500 mK. They are essentially identical, with no indication of flow over a period of about 20 hours. The rate of pressure change is

$$\frac{dP}{dt} = \frac{A\bar{v}}{\kappa_{\text{solid}}V_2} < \frac{0.5 \text{ mbar}}{20 \text{ hours}} \quad (1)$$

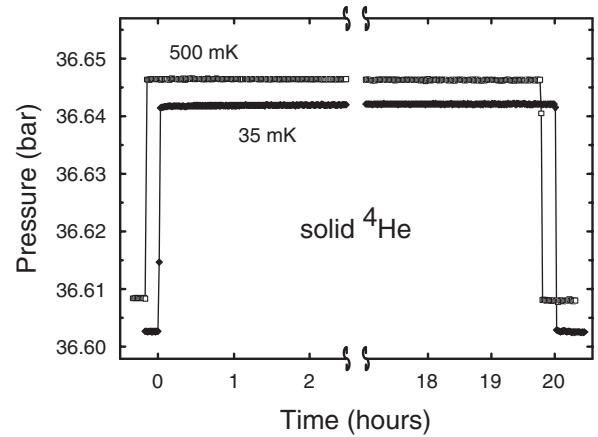


FIG. 2. Solid ^4He response at 500 mK (upper curve, open symbols) and 35 mK (lower curve, solid symbols). Lines are guides to the eye and the curves are offset for clarity. Note the time scale, which is much longer than in Fig. 1.

giving a limit on the average flow velocity

$$\bar{v} = \frac{\rho_s}{\rho} v_c < 1.2 \times 10^{-14} \text{ m/s}. \quad (2)$$

We also made low frequency ac measurements using the piezoelectric actuator to produce smaller pressure oscillations (± 4 V, corresponding to ± 3 mbar). The pressure was measured using a manual capacitance bridge (General Radio 1615-A operating at 10 kHz) with an analog lock-in amplifier, and the ac response was monitored with a digital lock-in (Stanford Research SR830 DSP). At 0.5 K the amplitude of the pressure oscillations was independent of frequency up to about 1 Hz, as expected since the GCA can flex very rapidly. Close to melting, the frequency dependence was more complicated since, as Fig. 1 shows, solid can flow through the capillaries even on a time scale of a few seconds. We looked for ac flow at low temperatures by cooling the cell below 0.5 K. Figure 3 shows the amplitude

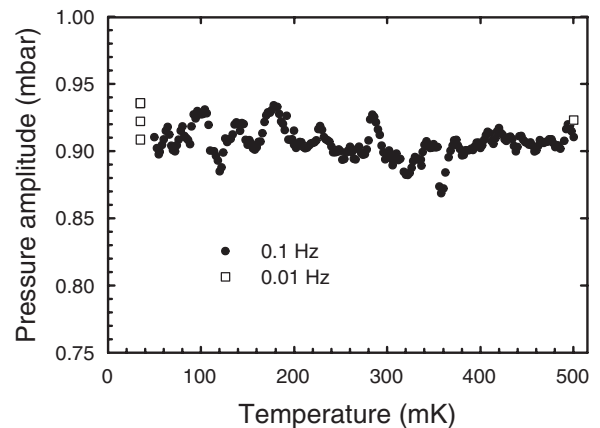


FIG. 3. ac pressure response in solid ^4He at low temperatures. Solid symbols are taken at 0.1 Hz during cooling. Open squares at 35 and 500 mK were taken at 0.01 Hz.

of the pressure oscillations at a frequency of 0.1 Hz. It also shows 0.01 Hz data at 35 mK and at 0.5 K, illustrating the frequency independence over this temperature range. The resolution is better than for dc flow and the pressure amplitude is constant within ± 0.02 mbar, with no evidence of temperature dependence that could be attributed to the onset of flow through the capillaries. Sample heating limited these measurements to frequencies below 1 Hz so we were not able to make direct comparison to the Kim-Chan torsional oscillator measurements at 1 kHz.

For a supersolid fraction $\frac{\rho_s}{\rho} = 1\%$, our dc flow limit (2) implies $v_c < 1.2 \times 10^{-12}$ m/s, 7 orders of magnitude smaller than the critical velocity inferred from Kim and Chan's torsional oscillator measurements and more than a thousand times smaller than the limits set by previous flow experiments [6,8]. Flow in solids often involves dislocations or grain boundaries, which can be immobilized by small concentrations of impurities. Our measurements using isotopically purified ^4He were essentially identical to our initial results with natural ^4He so the absence of pressure-induced superflow is not due to impurity pinning of such defects. There has also been a suggestion [30] that a surface melted layer could allow solid helium in a torsional oscillator to slip, providing an alternative, nonsupersolid explanation of the bulk ^4He decoupling. Our measurements appear to rule out such behavior at low temperatures, although it may occur near melting.

The torsional oscillator results were also consistent with the displacement, rather than the velocity, being limited to a critical value. We can put limits on possible displacements of the solid helium at low temperatures from the data in Figs. 2 and 3. Since the pressure jumps at 35 and 500 mK agree within 1 mbar, the corresponding displacements cannot differ by more than 2 nm. Our ac measurements are less sensitive to flow, but more sensitive to displacements, and rule out movements of solid helium through the capillaries larger than 0.03 nm. If we again assume that only a 1% supersolid fraction moves, this would imply supersolid displacements less than 3 nm, comparable to the amplitude of Kim and Chan's torsional oscillator at their critical velocity (for their 1 kHz oscillator, $v_c \sim 10 \mu\text{m/s}$ corresponds to an amplitude $\frac{v_c}{\omega} \sim 2$ nm).

These experiments show that static or low frequency pressure differences do not produce either superflow or unusual displacements at low temperatures in solid ^4He . If the helium forms a supersolid, then its flow properties must be quite different from those of a superfluid, in which the chemical potential difference created by a pressure change would cause superflow.

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