## Supersolid Helium at High Pressure

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We have measured the pressure dependence of the supersolid fraction by a torsional oscillator technique. Superflow is found from 25.6 bar up to 136.9 bar. The supersolid fraction in the low temperature limit increases from 0.6% at 25.6 bar near the melting boundary up to a maximum of 1.5% near 55 bar before showing a monotonic decrease with pressure extrapolating to zero near 170 bar.

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Recently we reported the observation of superfluidity with the torsional oscillator technique in solid <sup>4</sup>He confined inside porous Vycor glass with a characteristic pore diameter of 7 nm [1], porous gold with a pore diameter of 490 nm [2] and also in bulk solid <sup>4</sup>He [3]. In the bulk experiment solid helium is confined in an annular channel inside the torsion bob (see inset III of Fig. 1). When the torsional oscillator is cooled below 230 mK the resonant period shows an abrupt drop from the expected, linearly extrapolated value from higher temperatures. The most simple explanation of the decrease in the period (which is proportional to the squared-root of the moment of inertia, *I*, of the torsion bob) is the onset of nonclassical rotational inertia (NCRI) or superfluidity in solid <sup>4</sup>He [4].

The supersolid fraction in the low temperature limit found in the Vycor and the bulk experiments is on the order of the 1% in spite of vastly different surface to volume ratio (a factor of  $2.5 \times 10^4$ ) of the space available for helium. This indicates the observed superfluidity is not a surface related phenomenon. It is also difficult to reconcile this observation with the suggestion that the superfluidity is due entirely to defects, dislocations, and other imperfections in the crystal since this would require the crystallite size in the bulk sample to be the same as that in Vycor or at most 7 nm.

Nevertheless, a number of theoretical papers suggest that superfluidity is unlikely to occur in a perfect crystal [5–8]. Andreev and Liftshitz suggested in 1969 a specific scenario that Bose condensation of zero-point vacancies and other defects can lead to superfluidity in solid helium [9]. If the observed superflow is in fact a simple consequence of condensation of zero-point vacancies then the supersolid fraction should decrease as the pressure (and hence density) of the solid sample is increased away from the melting boundary deep into the solid phase. In the bulk solid experiment a total of 17 samples of solid helium with pressure ranging from 26 to 65 bar were studied [3]. While superflow was found in every sample, the value of the supersolid fraction in the low temperature limit was found to vary between 0.6 and 1.7% with no obvious dependence on pressure (Fig. 4).

The set of measurements reported below was undertaken to understand and to reduce the scatter in the supersolid fraction. We made the assumption that the scatter in the supersolid fraction is a consequence of the random orientations of small crystallites inside the annular channel of the torsion cell. The solid samples studied in Ref. [3] were grown using the blocked capillary (constant volume) method. This method results in solid samples of reasonable

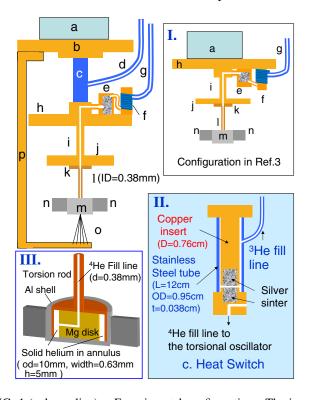


FIG. 1 (color online). Experimental configurations. The insets depict I: configuration in Ref. [3], II: details of heat switch, and III: annular channel in the cell. (a) is the mixing chamber; (b), thermal platform; (c), <sup>3</sup>He heat switch; (d), <sup>3</sup>He fill line; (e), silver sinter heat sink; (f), wound capillary heat sink; (g), <sup>4</sup>He fill line; (h), thermal platform; (i), vibration isolator; (j), thermal platform; (k), torsional oscillator base; (l), torsion rod; (m), torsion cell; (n), two electrodes; (o), copper wires; (p), cold finger.

quality [10–14]. There is consensus, however, that the crystal quality of solid grown under constant volume condition is inferior to that grown under constant temperature from superfluid [15–18] and constant pressure [19–23] conditions. In the constant volume method after liquid helium of the desired density has been introduced into the sample cell via a thin capillary, the temperature of the capillary at a certain point above the sample cell is lowered to solidify the helium within and form a plug. The liquid in the (constant) volume below the solid plug, including the sample cell, is then cooled into the solid phase with a concomitant drop in pressure.

The experimental configuration of Ref. [3] is shown in inset I, Fig. 1. Liquid helium from the capillary (g) is introduced from the base (k) of the oscillator through the torsion rod (1) to the torsion cell (m). The base of the oscillator was attached to a thermal platform (j) which was in turn connected to the mixing chamber (a) of the dilution refrigerator through another platform (h). Platforms h and j are connected through a vibration isolator (i) in the form of a hollow copper cylinder. In such a configuration, liquid helium in the torsion rod will freeze first. The solidification will then proceed through the long narrow hole (i.d. = 0.38 mm and length = 7.5 mm) drilled inside the magnesium disk (Fig. 1, inset III) before reaching the annulus of channel width = 0.63 mm. It is therefore plausible that such a solidification process through this long narrow path could result in polycrystalline samples with grains of size no larger than 0.38 mm with random and unreproducible orientations inside the annulus. We think this unfavorable growth process of the solid is the primary reason for the scatter in the supersolid fraction.

In the current experiment, we used the same torsional oscillator as that in Ref. [3]. However, we have installed a heat switch (c) (Fig. 1 inset II) between thermal platform h and the mixing chamber in an attempt to change the cooling path of the torsion cell during the growth of solid to facilitate the nucleation of solid helium from the bottom of the annulus. The heat switch can be opened (closed) by emptying (filling) the thin wall stainless steel tubing with liquid <sup>3</sup>He. When a solid is being grown in the torsion cell, the heat switch is opened and the latent heat of freezing is designed to be primarily carried from the torsion cell to the mixing chamber through 10 strands of 0.05 mm diameter copper wires (o) attached to the bottom of the torsion cell. The other ends of the wires are attached to a heavy copper bar (p) that is firmly anchored to the mixing chamber (Fig. 1).

During the growth of solid helium the pressure and the density of the sample in the cell were monitored by a resistance strain gauge (glued onto the wall of the torsion cell) and the increase in the period of the oscillator. In all solid samples we have grown for this study, we found the resonant period always shows an increase before the pres-

sure shows any noticeable decrease. This indicates that initially solid nucleates under the constant pressure growth condition and it is reasonable to speculate that the nucleation starts at the bottom of the annulus, close to the copper wires. In spite of repeated effort, we found it impossible to complete the solidification process under the constant pressure condition. What we found is that before the growth of solid in the torsion cell is completed the pressure always exhibits a drop indicating a solid block is formed cutting off the supply of helium into the torsion cell. The fraction of solid helium before the fill line is blocked is about 55% ( $\pm 5\%$ ) of total solid helium in the cell. It likely occurs when the solid in the annulus grows into the narrow hole in the magnesium disk. The copper wires were sufficient to cool the torsional oscillator down to 1.3 K.

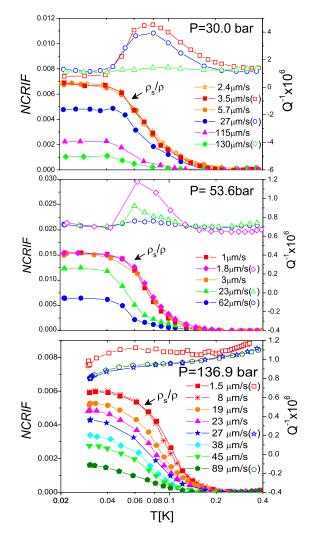


FIG. 2 (color online). NCRIF (closed symbols) and dissipation (in  $Q^{-1}$ , open symbols) as a function of temperature for three samples at P=30, 53.6, and 136.9 bar. The increases of the resonant period due to the filling of the cell are 2814, 3045, and 3700 ns, respectively. NCRIF measured at the limit of low oscillation speed corresponds to  $\rho_s/\rho$ .

Further cooling requires the introduction of liquid <sup>3</sup>He into the heat switch.

We have grown and studied 14 solid samples with pressure ranging from 25.6 to 136.9 bar. All samples, including the sample at 136.9 bar, exhibit supersolid decoupling. The temperature dependence of the supersolid fraction of each sample resembles those in Ref. [3]. Figure 2 shows the nonclassical rotational inertia fraction (NCRIF) as a function of temperature at different maximum oscillation speed of the annulus,  $v_{\text{max}}$ , for solid samples of 30.0, 53.6, and 136.9 bar. The NCRIF results are deduced from the resonant period following the same procedures outlined in Ref. [3]. The reproducibility of the resonant period readings is about 0.5 ns. The resonant period increases on the order of 3000 ns with the filling of the solid sample. This translates an error bar in NCRIF and  $\rho_s/\rho$  to about  $2 \times 10^{-4}$ . Since the determination of NCRIF involves the subtraction of the measured period from a temperature dependent background curve, there may be an additional systematic error in NCRIF (and  $\rho_s/\rho$ ) of comparable magnitude. The dissipation, in inverse quality factor  $(Q^{-1})$  of the oscillator, deduced from the amplitude of oscillation at three different  $v_{\rm max}$  of each sample are also shown with open symbols. Broad maxima centering near where NCRIF is changing most rapidly are found. These broad maxima in dissipation are more pronounced in low pressure solid samples and in data taken at low  $v_{\rm max}$ . The dissipation maximum fades with increasing pressure and it is barely discernible in samples with pressure exceeding 108 bar.

Figure 3 shows normalized NCRIF<sub>0</sub> (the low temperature limit of NCRIF at different  $v_{\rm max}$  divided by its value obtained at the lowest  $v_{\rm max}$ ) as a function of  $v_{\rm max}$  for solid samples at five different pressures. These five sets of data show a much better "collapse" onto a single curve compared to the data shown in Fig. 3 (panel D) of Ref. [3]. The NCRIF is independent of  $v_{\rm max}$ , provided  $v_{\rm max}$  does not exceed 10  $\mu$ m/s. Once exceeded, NCRIF decreases with  $v_{\rm max}$ . We interpret this as a critical velocity effect and as noted in Ref. [3], the result indicates that superflow in solid helium becomes dissipative with the appearance of a single vortex with unity quantum circulation (if the effective mass is a third of the atomic mass) or just a few vortices.

NCRIF measured with  $v_{\rm max}$  smaller than 10  $\mu$ m/s, being independent of oscillation speed, represents the supersolid fraction,  $\rho_s/\rho$ . We have used oscillation speed of 5  $\mu$ m/s or less to study the supersolid response of 9 additional solid samples at 25.6, 41.8, 48.7, 56.9, 60.1, 70.6, 87.1, 99.0, and 104.0 bar. The uncertainty in pressure determination is less than 0.5 bar. The low temperature supersolid fractions,  $\rho_{so}/\rho$ , of all 14 samples are plotted in Fig. 4 as a function of pressure. The new data show the supersolid fraction increases from 0.6% near the melting pressure up to a maximum of 1.5% near 55 bar before decreasing with further increase in pressure. A linear ex-

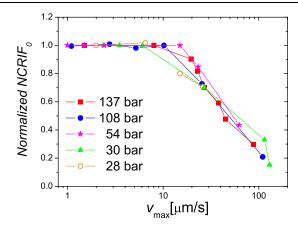


FIG. 3 (color online). Oscillation speed dependence of normalized NCRIF for five solid helium samples of P=28.1, 30, 53.6, 108.3, and 136.9 bar.  $v_{\rm max}$  is the maximum speed of the annulus holding solid <sup>4</sup>He. NCRIF is normalized by the low temperature supersolid fraction,  $\rho_{\rm so}/\rho$ .  $\rho_{\rm so}/\rho$  for each sample are 0.006 91, 0.006 79, 0.015 44, 0.006 12, and 0.005 76, respectively.

trapolation suggests the supersolid fraction will be reduced to zero for pressures exceeding 170 bar. Unfortunately the torsion cell exploded as we attempted to make a solid sample of 170 bar.

While the data taken with the new experimental configuration appears to be much improved over those shown in Ref. [3], the point to point scatter of the  $\rho_{so}/\rho$  values as shown in Fig. 4 is typically 0.15% and for the three data points near 55 bar it is as large as 0.5%. These values are many times larger than the uncertainty in  $\rho_s/\rho$  obtained in an individual sample. This suggested that we have not been growing solid samples in a completely reproducible manner and there is still substantial variation in the "crystal quality" of these solid samples. Measurements on solid samples contained in a torsion cell with simple cylindrical

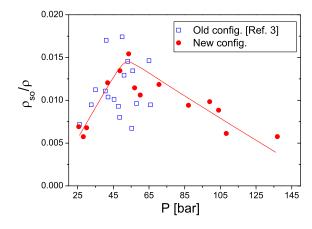


FIG. 4 (color online). Supersolid fraction in the low temperature limit,  $\rho_{so}/\rho$ , as a function of pressure. Solid samples prepared with heat switch (new configuration) yield data with less scatter. The solid line is a guide to the eyes.

geometry without an annulus and grown entirely under the constant pressure may reduce the scatter further.

The nonmonotonic dependence of the supersolid fraction on pressure indicates that, as noted above, the origin of supersolidity is more subtle than just the simple Bose condensation of zero-point vacancies. The fact that we found a supersolid fraction of up to 1.5% is also difficult to reconcile with the simple vacancy condensation model. A number of experiments [24,25] give indirect evidence that zero-point vacancies, if present below 0.2 K, would be much smaller than 1% of the lattice sites.

Solid helium at an elevated pressure is expected to be less quantum mechanical than that at a lower pressure [26,27]. X-ray diffraction studies measuring the zero-point energy induced motions of the  $^4$ He atoms from their lattice sites appear to confirm this expectation [22,28]. The declining supersolid fraction with pressure beyond 55 bar is also consistent with this expectation. However, we do not understand why there is no apparent change in  $T_c$  with pressure.

It has been suggested that a perfect solid helium crystal cannot support superflow [6-8]. This idea received support from the recent torsional oscillator experiment of Rittner and Reppy [29]. They found supersolid decoupling in a solid sample made by the same blocked capillary method. However, upon annealing the sample by cooling it much more slowly from about 1.5 K than when it was first grown, the supersolid decoupling is found to diminish and even disappear. We have looked for this annealing effect by cooling a number of solid samples from the liquid-solid coexistence region down to the lowest temperature at a cooling rate that is up to 5 times slower than that of Rittner and Reppy. We found the supersolid fraction due to different annealing procedure can differ by at most 15%. We have not been able to eliminate the superflow in any of the more than 50 bulk solid samples we have studied so far in our laboratory. In addition to Rittner and Reppy, our observation of superflow in solid helium with the torsional oscillator technique was also replicated by the Shirahama group at Keio University [30] and the Kubota group of the University of Tokyo [31]. These two groups have also tried but failed to eliminate superflow by annealing.

To conclude, we observed the supersolid phase extends at least up to 136.9 bar. The supersolid fraction appears to increase with pressure from the melting pressure up to 55 bar and then decreases with further increase with pressure. Linear extrapolation indicates the supersolid phase terminates near 170 bar. The critical velocity of the superflow is found to be on the order of  $10 \ \mu m/s$ .

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