

Disorder and the Supersolid State of Solid ^4He

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(Received 13 March 2007; published 26 April 2007)

We report torsional oscillator supersolid studies of highly disordered samples of solid ^4He . In an attempt to approach the amorphous or glassy state of the solid, we prepare our samples by rapid freezing from the normal phase of liquid ^4He . Less than two minutes is required for the entire process of freezing and the subsequent cooling of the sample to below 1 K. The supersolid signals observed for such samples are remarkably large, exceeding 20% of the entire solid helium moment of inertia. These results, taken with the finding that the magnitude of the small supersolid signals observed in our earlier experiments can be reduced to an unobservable level by annealing, strongly suggest that the supersolid state exists for the disordered or glassy state of helium and is absent in high quality crystals of solid ^4He .

DOI: [10.1103/PhysRevLett.98.175302](https://doi.org/10.1103/PhysRevLett.98.175302)

PACS numbers: 67.80.Mg, 67.90.+z

Following the discovery by Kim and Chan (KC) [1,2] of the supersolid or nonclassical rotational inertia state of bulk solid ^4He , several independent groups using the same torsional oscillator technique have confirmed the KC supersolid results. These include the Japanese groups of Shirahama *et al.* [3], working at Keio University, and Penvev *et al.* [4] at the Institute for Solid State Physics, Tokyo University, as well as our group [5] at Cornell University. In these early experiments, the solid samples were formed by the blocked capillary technique. In this method, the fill line to the cell is first allowed to freeze, ensuring that solidification in the cell occurs under a condition of constant average density. This technique is known to produce relatively disordered polycrystalline samples. The signals observed in the early experiments were small, representing, at most, a few percent of the total solid helium mass. The Cornell experiments [5] also demonstrated that the supersolid signal could be substantially reduced through annealing of the sample. In some cases, the annealing process appeared to eliminate the supersolid signal; i.e., it reduced the supersolid fraction below the 0.05% level of experimental detection. This signal reduction upon annealing strongly suggested that sample disorder plays an important role in supersolid phenomena. This inference is supported by more recent work by Chan's group [6], where high quality crystals were grown under conditions of constant pressure. The supersolid signals observed for these constant pressure samples were somewhat smaller in magnitude than those obtained for more disordered samples created in the same cell by the blocked capillary method.

Recently, there has been a growing consensus in the theoretical community [7–10] that an ideal hcp helium crystal will not exhibit the supersolid phenomenon but rather that some form of disorder such as vacancies, interstitials, superfluid grain boundaries [11,12], or perhaps a glassy or superglass phase [13,14] is required for the ex-

istence of the supersolid state. A summary of the current theoretical literature is given in a recent review [15].

In the measurements reported here, we investigate the role of disorder in supersolid phenomena. In order to maximize sample disorder, we have confined the solid within a narrow annular region. The small volume and large surface area to volume ratio (S/V) provided by this geometry allow rapid freezing and subsequent cooling of the sample to low temperatures, ensuring a high degree of frozen-in disorder. To provide a contrast to the disordered annular samples, we have also studied the supersolid phenomena in an open cylindrical geometry where slow freezing and cooling are employed to promote the growth of large helium crystals with a relatively low level of disorder.

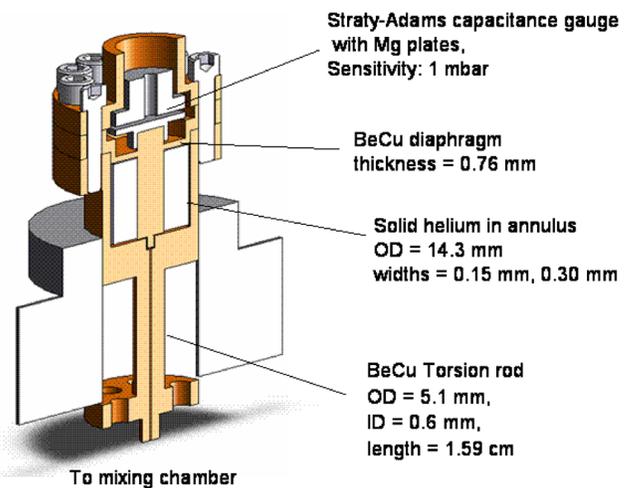


FIG. 1 (color online). Torsional oscillator: The motion of the torsion bob is excited and detected electrostatically. A Straty-Adams capacitance gauge, a heater, and a thermometer are attached on top of the torsion bob. At 4 K, the resonance frequency with the biggest insert is 874 Hz, and the mechanical quality factor is $Q = 5 \times 10^5$.

The torsional oscillator cell employed for these measurements is shown in Fig. 1. A special feature is a removable top plate sealed to the body of the oscillator with a lead o-ring. This feature allows us to vary the internal geometry of the cell between runs. A capacitance pressure gauge, along with a thermometer and heater, is incorporated into the top structure of the oscillator. Cylindrical magnesium inserts can be mounted inside the oscillator to provide a range of annular spacings. The sample volume of the cell with the insert structure removed is 1.8 cm^3 with a surface area to volume ratio $S/V = 4.6 \text{ cm}^{-1}$. With a magnesium insert in place, the open volume of the cell is much reduced, and the surface area in contact with the solid helium is nearly doubled. At this time, we have investigated samples formed with two different annular gaps: 0.30 and 0.15 mm. For the cell with the smallest gap, $S/V = 131 \text{ cm}^{-1}$.

We determined the moment of inertia of the cell to be 51 gcm^2 based on the change in frequency produced by a calibrated variation in the cell moment of inertia. We obtain the solid ^4He moment of inertia I_s by a calculation based on the density of the solid helium at the melting temperature and the measured geometry of the cell interior. In the case of the cell with the 0.15 mm gap, $I_s = 9.8 \times 10^{-3} \text{ gcm}^2$ for an assumed ^4He molar volume of 19.5 cm^3 . The addition of this moment of inertia to the oscillator would produce an increase in the oscillator period of magnitude $\Delta P_0 = 109 \pm 4 \text{ ns}$. The uncertainty in ΔP_0 arises almost entirely from the measurement error in determining the dimension of the 0.15 mm gap.

In previous torsional oscillator supersolid experiments, the quantity ΔP_0 is obtained from the period shift seen upon freezing. In our case, the period shift that occurs during freezing is complicated by a shift of opposite sign caused by the drop in pressure that occurs during solid formation. The pressure sensitivity of the oscillator period is discussed below. In the case of our 0.15 mm gap samples, the period shift due to the pressure effect is on the order of 20%–30% of ΔP_0 . Therefore, we have chosen to calculate ΔP_0 rather than to rely on a pressure correction to the period shift seen on freezing.

Otherwise, the experimental procedures followed in these measurements are similar to those employed in our earlier work [5]. As a first step, an empty cell run is made to determine the temperature-dependent backgrounds for the torsional oscillator period, dissipation, and the capacitance pressure gauge. The helium used for our samples is commercial grade helium with a reported ^4He concentration of 0.3 ppm. After the cell is filled with liquid ^4He , the capacitance pressure gauge is calibrated at 4 K for pressures up to 70 bar. As the pressure is raised during this calibration, the oscillator period is observed to increase linearly with pressure, with a sensitivity of 1.14 ns/bar. In our measurements discussed in this Letter, the samples were all formed at pressures above the hcp-bcc triple point to avoid complications due to the bcc phase.

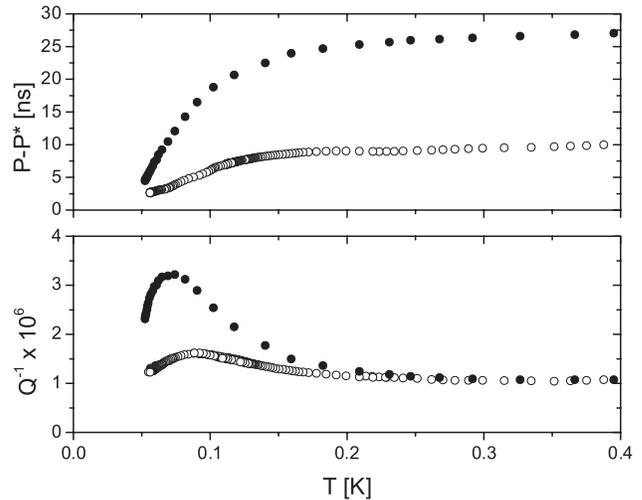


FIG. 2. Period and dissipation are shown as a function of temperature for the annular cell with 0.15 mm width. The first sample (open circles) was cooled to below 1 K over a period of three hours. Following this low temperature run, the sample was rapidly melted and quench-cooled (solid circles) to below 1 K in 90 s. The sample pressures were 41 (open circles) and 51 bar (solid circles), and the rim velocities at 300 mK were 9.7 (open circles) and $5.9 \mu\text{m/s}$ (solid circles), respectively. The value of P^* for both samples is 1.144 785 ms.

In Fig. 2, we show period and dissipation Q^{-1} data as a function of temperature for two runs with the 0.15 mm gap cell. The lower set of period data was obtained for the first run following the initial freezing of the sample. In this case, the freezing and subsequent cooling to below 1 K took place over a three-hour period. During this relatively slow cooling process, we believe that a certain amount of sample annealing can take place, resulting in a relatively small supersolid signal obtained by taking the difference between the period data and a linear fit to period data above the supersolid transition. A further decrease in cooling rate, taking 14 hours to cool below 1 K, did not result in an additional decrease in signal size. The upper data set was obtained after a “quench” cool of the sample. In this procedure, the sample is melted by a heat pulse applied to either the cell heater or the cell thermometer. As soon as melting has occurred, as indicated by the pressure and period signals, the heat is turned off, and the sample rapidly freezes and cools to a temperature below 1 K in a time interval of approximately 90 seconds.

The most important feature of these quench-cooled data is the large increase in the magnitude of the supersolid signal. For this sample, the period reduction $\Delta P(T)$ at 50 mK is about 22 ns and corresponds to a supersolid fraction $\rho_s/\rho \equiv \Delta P(T)/\Delta P_0$, amounting to a 20% fraction of the solid helium moment of inertia opposed to 6% for the slowly frozen sample. Similarly, the supersolid fraction in the 0.3 mm annulus varied from 4% (for slowly frozen samples) to 6% after a quench cool. Although this

signal is more than an order of magnitude larger than any reported for previous experiments [1–3,5,4], the general temperature dependence of both the supersolid signal and the dissipation data are similar to that of the earlier data.

In a significant recent experiment, Sasaki *et al.* [11] have observed grain-boundary mediated superflow in solid samples in contact with the superfluid phase. These authors suggest that superflow along the surface of grain boundaries may be a possible explanation for the supersolid signals observed by KC. Although the small signals observed in the early experiments might be explained by this mechanism, it is difficult to imagine grain boundaries occupying 20% or more of the sample volume, as would be required to explain the signals in the present experiments. A solid helium sample with such a high concentration of grain boundaries might better be described as a glass.

Another feature of the data shown in Fig. 2 is an increase in the period of the oscillator following the quench cool. This increase in the period is principally associated with the substantial increase in the sample pressure of 9.7 bar resulting from the quench-cool process. We believe that this increase is a result of inflationary pressure arising from the increased disorder in the sample. Vacancies, microscopic voids, and a possible glassy phase are the most obvious candidates for the source for the inflationary pressure. If so, the excess pressure provides a convenient measure of the relative degree of disorder in the sample.

Annealing of this sample at temperatures near the melting value will lead to a reduction in the level of disorder and should also lead to a reduction in the sample pressure. To test this idea, we have raised the temperature of the sample to 1.57 K, where thermally activated annealing is expected on the basis of our earlier work [5]. In Fig. 3, we plot both the oscillator period and the sample pressure

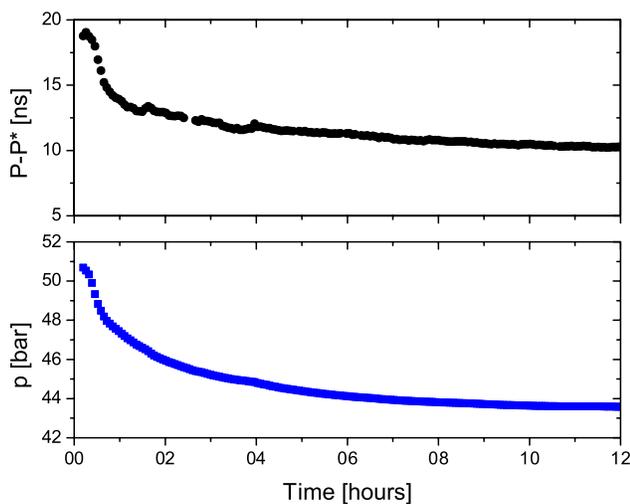


FIG. 3 (color online). Pressure and period are shown as a function of time during annealing. The temperature is set to 1.57 K; the melting temperature of the sample is 2.35 K.

as functions of time during the annealing process. Both the pressure and the period are seen to relax with a time constant of approximately 1 h. After waiting 10 h, the pressure approaches a constant value, 7 bar below the preannealed value. A further increase in the temperature to 2.0 K produces a further drop in pressure with a much faster relaxation time. The reduction in pressure during the annealing of disordered solid ^4He samples is not a new phenomenon but has been reported a number of times by other experimenters, most recently in a paper [16] reporting evidence for a glassy phase in ^4He samples grown by the blocked capillary method.

We have also studied the influence of velocity on the magnitude of the supersolid signal for the 0.15 and 0.30 mm annular cells. In general, the results are similar to those reported by KC for their 0.63 mm annular cell; however, we find a critical velocity which is a factor of 4 higher than for the KC data.

To further investigate the influence of disorder on the magnitude of the supersolid signal, we have formed solid samples in a 951 Hz oscillator with a relatively large (1.8 cm^3) open cylindrical volume and S/V of 5.84 cm^{-1} . The samples formed in this cell require two hours for the freezing of the sample and the subsequent cooling to below 1 K. Given the relatively slow cooling through the annealing temperature range $T > 1 \text{ K}$, we expect a polycrystalline sample with relatively large crystals and a significantly reduced level of disorder as compared to the quench-cooled samples.

In Fig. 4, we display period and dissipation signals for these samples. The supersolid signals are very small, on the

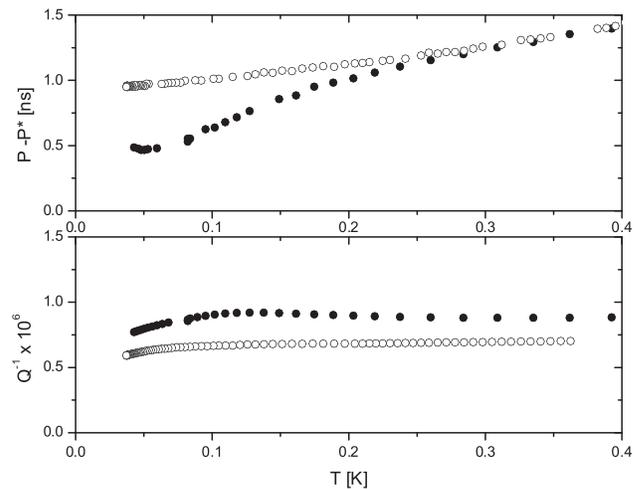


FIG. 4. Period and dissipation are shown as a function of temperature in a cylindrical cell with a volume of 1.8 cm^3 . The sample (solid circles) was frozen and cooled below 1 K in 2 hours. The sample pressure was 32 bar, and the rim velocity at 300 mK was $24.7 \mu\text{m/s}$. At a velocity of $9 \mu\text{m/s}$, the graphs are identical. We also display the empty cell period and dissipation (open circles). The values of P^* are 1.054 064 (solid circles) and 1.052 100 ms (empty cell).

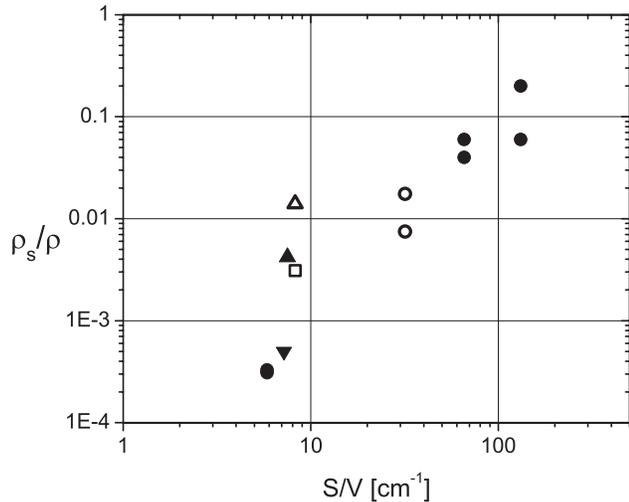


FIG. 5. Overview for a range of supersolid fractions as a function of surface to volume ratios from different researchers. All of the data obtained by the blocked capillary method are above 31 bar in order to avoid complications by the bcc phase. Cornell (solid circles); Penn State, annular cell (open circles) [2,6], cylindrical cell $T_{\text{melting}} = 2.17$ K (open triangle) [17], and cylindrical cell constant pressure growth 26 bar (open square) [17]; Shirahama *et al.* 41 bar (closed triangle) [3]. An upper bound on the size of the supersolid signal set by our earlier measurements in a square cell at 32 bar [5] is displayed as an inverted closed triangle.

order of 3.1×10^{-4} of the total solid helium moment of inertia for velocities as low as $9 \mu\text{m/s}$. Thus, an alteration in sample preparation and the surface to volume ratio can lead to a reduction in the amplitude of the supersolid signal of 3 orders of magnitude. We believe that the extremely small signals observed for these large open volume samples can most likely be attributed to remnant vestigial disorder in the sample and do not represent supersolid flow within the helium crystallites themselves.

In Fig. 5, we give an overview of the relation between the surface to volume ratio and the maximum and minimum observed supersolid signals. The data shown in this plot demonstrate a clear trend for increasing supersolid signal with increasing surface to volume ratio. As S/V increases, the possibility of greater frozen-in disorder increases, since the samples can be frozen and cooled more quickly and also because the disorder may be stabilized by a more confining geometry.

At this time, it is still unclear what the exact mechanism for the supersolid phenomenon may be. It is clear, however, that disorder plays a key role in the phenomenon: Increasing disorder leads to larger supersolid signals, while reducing disorder by growing higher quality crystals has the

effect of reducing the supersolid signal, in some cases below the detectable level. An unresolved puzzle is presented by the results of KC in Vycor [1] and porous gold [18]; in both cases, the S/V ratios are orders of magnitude larger than for our cells, yet the supersolid fractions are on the order of 2%.

We acknowledge useful conversations with M. H. W. Chan, A. C. Clark, E. Mueller, K. Hazzard, V. Elser, and G. V. Chester, and we also thank J. V. Reppy for editorial assistance. The work reported here has been supported by Cornell University, the National Science Foundation under Grant No. DMR-0203244, and the Cornell Center for Materials Research under Grant No. DMR-0520404.

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