Absence of Supersolidity in Solid Helium in Porous Vycor Glass

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In 2004, Kim and Chan carried out torsional oscillator measurements of solid helium confined in porous Vycor glass and found an abrupt drop in the resonant period below 200 mK. The period drop was interpreted as probable experimental evidence of nonclassical rotational inertia. This experiment sparked considerable activities in the studies of superfluidity in solid helium. More recent ultrasound and torsional oscillator studies, however, found evidence that shear modulus stiffening is responsible for at least a fraction of the period drop found in bulk solid helium samples. The experimental configuration of Kim and Chan makes it unavoidable to have a small amount of bulk solid inside the torsion cell containing the Vycor disk. We report here the results of a new helium in Vycor experiment with a design that is completely free from any bulk solid shear modulus stiffening effect. We found no measurable period drop that can be attributed to nonclassical rotational inertia.

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In 2004, Kim and Chan (KC) carried out a torsional oscillator (TO) experiment of solid helium confined in porous Vycor glass [1]. When the torsion cell was cooled below 200 mK, an abrupt drop in the resonant period of the TO was found. The period drop was interpreted as probable evidence of nonclassical rotational inertia (NCRI) in solid ^{4}He. A similar period drop was found in bulk solid ^{4}He [2]. While these results were replicated in over 25 other TO experiments, the magnitude of NCRI varies from experiment to experiment [3–11].

Interestingly, the shear modulus of bulk solid helium showed an increase at the same onset temperature of NCRI with identical temperature and ^{3}He concentration dependences [12]. For polycrystalline samples, a 10%–20% increase was found, and for single crystal samples the increase can be as large as 80% [13]. Since solid helium is a constituent of the TO, an increase in its shear modulus will stiffen the TO and cause the resonant period to drop, thus mimicking NCRI. The shear modulus effect on the resonant period of TO was analyzed in detail in two different studies [14,15]. Simulations with the finite element method (FEM) give consistent results with their analytic calculations [16]. For an "infinitely" rigid TO containing a cylindrical isotropic solid helium sample of 1 cm in diameter and in height and oscillating at 1 kHz, a 20% increase of the shear modulus of helium results in a drop in the resonant period of approximately 1×10^{-7}, or 0.1 ns. For a TO cell that is not rigid, the shear modulus effect can be greatly amplified. In some TOs, solid helium contributes significantly to the mechanical coupling of the different parts of the torsion cell. The coupling is strengthened when the solid helium sample is stiffened at low temperature resulting in a lower resonant period [4,17]. The torsion rod is usually attached at the center of one end of the (cylindrical) torsion cell. Although the shear modulus of the metal is 3 orders of magnitude larger (5×10^{10} versus 2×10^{7} Pa) than solid helium, if the plate to which the torsion rod is attached is sufficiently thin, the supplemental effect of solid helium in transmitting the torque from the torsion rod to the rest of the torsion cell will not be negligible. In this case the resonant period may show a measurable abrupt drop when solid helium is stiffened at low temperature [18]. Last, in most TOs, torsion rods are hollow and used as fill lines. Solid helium inside the torsion rod contributes to the spring constant of the torsion rod. If the ratio of the outer and inner diameters of the torsion rod is not sufficiently large, the shear modulus increase at low temperature of the solid helium in the torsion rod can again induce a measurable period drop. This torsion rod effect may be responsible for the apparent NCRI reported in a number of TO experiments [19].

While there is considerable uncertainty on the claim of NCRI in bulk solid helium, this should not be the case, at first glance, for the 2004 KC Vycor glass experiment [20]. The microscopic mechanism responsible for the shear modulus increase at low temperature in solid helium is the pinning of the dislocation lines to ^{3}He impurities at low temperatures. This is not applicable in Vycor glass, since the typical distance between neighboring nodes in a dislocation line network is a few microns [21], orders of magnitude longer than the diameter (7 nm) of the pores in Vycor glass. However, all porous media experiments to date, including KC, were carried out by placing the porous samples inside sealed metal torsion cells [1,22–25]. Since helium must be allowed to infuse into the porous samples, it is necessary to maintain an open empty space, however small, in the torsion cells. When the porous sample is pressurized with solid helium, there will be bulk solid helium in this open space. This raises the question of whether this small quantity of bulk solid in the TO can be responsible for the observed period drop that has been interpreted as the NCRI signature. To clarify this issue, we built a TO out of a Vycor glass disk without a metal container and with a configuration that reduces the shear modulus stiffening effect of bulk solid helium to be orders of magnitude below that can be detected. Drawings of the current and the KC torsional oscillators are shown in Fig. 1.

The Vycor glass disk (diameter 14 mm and height 10 mm) was sealed by a thin layer of epoxy (Stycast 2850) painted on the exterior of the disk. A small hole was drilled into the center of the Vycor disk to accommodate a filling capillary (outer diameter 0.3 mm and inner diameter 0.1 mm). The capillary-Vycor joint is also sealed with epoxy. The other end of the capillary is secured in position 6 cm below the Vycor disk. The volume of the empty space inside the Vycor disk surrounding and inside the capillary is estimated to be less than 1×10^{-4} cc, or 0.02% of the total volume of solid helium inside the porous structure. The expected period changes due to the stiffening of the bulk solid helium inside and outside of the capillary are at most 3×10^{-5} and 5×10^{-3} ns, respectively. These values are orders of magnitude smaller than the resolution of the experiment. We know there is no other bulk space in the cell, because, if there is any gap between the Vycor disk and the thin epoxy layer, a leak will develop when the sample is pressurized. The Vycor glass disk is glued onto an Invar plate with thermal expansion that matches Vycor. The Vycor-Invar cell was then secured to a solid BeCu torsion rod by screws. The mechanical Q of the empty TO above 0.1 K is 8×10^5, and, as is commonly found in many other TOs, the Q shoots up below 0.1 K. The resonant period of the TO is 1.15 ms, and the mass loading due to a solid helium sample is approximately 5000 ns. Measurements were made on three different liquid helium films adsorbed on the walls of the Vycor pores, and superfluid transitions with T_c's of 70, 450, and 630 mK were found. These measurements showed the expected Kosterlitz-Thouless-like behavior confirming that the Vycor disk is free from any crack and capable of supporting superflow. A careful comparison of the temperature dependence of the superfluid density of the lowest coverage film to that of an earlier experiment showed that our temperature scale is reliable to within 1 mK down to 30 mK [26].

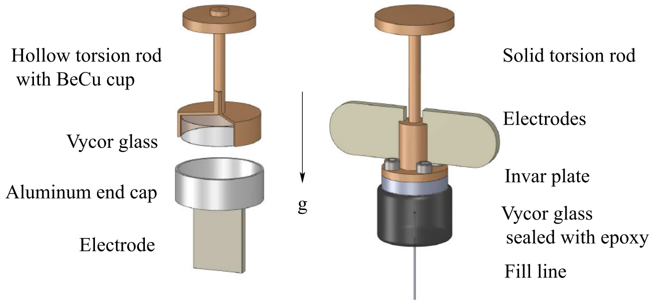

FIG. 1 (color online). Torsional oscillators with porous Vycor glass used in 2004 (left) and the current experiment (right).

The principal results of this experiment are shown in Fig. 2. In contrast to KC in 2004, the "naked" Vycor TO shows no period drop within the experimental resolution (0.1 ns) near and below 200 mK. Since the mass loading is 5000 ns, if there is NCRI, it is less than 2×10^{-5}.

Both the 2004 and 2012 TOs show a period drop with decreasing temperature above 1 K. This drop is the signature of solidification of liquid helium in the Vycor pores. In a TO housing a bulk liquid-solid coexistence sample, the resonant period increases when liquid solidifies and adheres to the walls of the torsion cell. However, during the solidification process in Vycor, liquid helium, being entrained in small pores with solid helium "plugs," is always coupled to the oscillation. Therefore the resonant period does not increase with solidification but rather decreases, because solid helium stiffens the silica structure of the Vycor glass and hence the TO. This is shown more clearly in Fig. 3. The cell was pressurized with 65 bar liquid helium at 2.7 K and cooled down below 1 K rapidly. The sample was melted and refrozen a few times with the capillary fill line blocked with solid helium. Figure 3 shows data taken during the fourth warming and cooling cycle and also the fifth warming scan, while Fig. 2 shows the data of the same sample during the fourth cooling scan. The thermal cycles show a melting and freezing hysteresis similar to that found in other studies [27]. Melting was found to commence near 1.9 K and completed at 2.5 K, while freezing is found to commence near 2.0 K and completed near 1.6 K. The period readings show a more rapid change with temperature in the liquid-solid coexistence region than that when the Vycor is filled with either all liquid or all solid. The data taken during the fifth

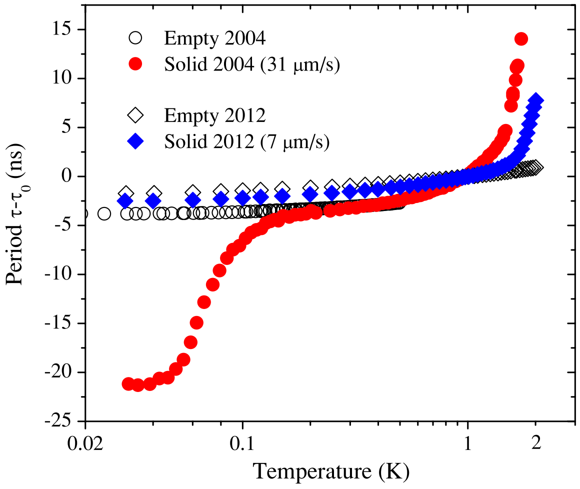

FIG. 2 (color online). Resonant period versus temperature of the 2004 and 2012 Vycor TOs infused with solid ^{4}He. Empty cell curves are also shown. The period drop found in the 2004 experiment appears to be the consequence of a bulk solid ^{4}He layer inside the TO. The curves are shifted for easy comparison by adjusting the values of τ_0.

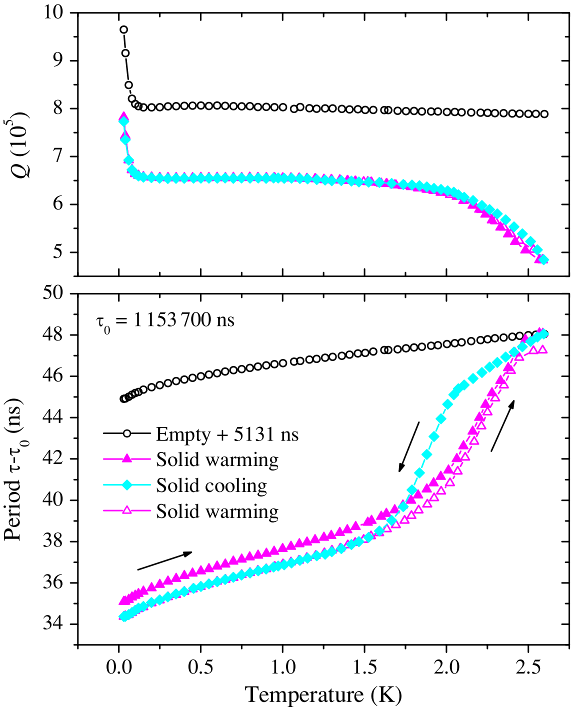

FIG. 3 (color online). Resonant period and Q value of warming and cooling scans of the same solid helium sample shown in Fig. 2. Oscillation speeds are between 7 and 11 μm/s.

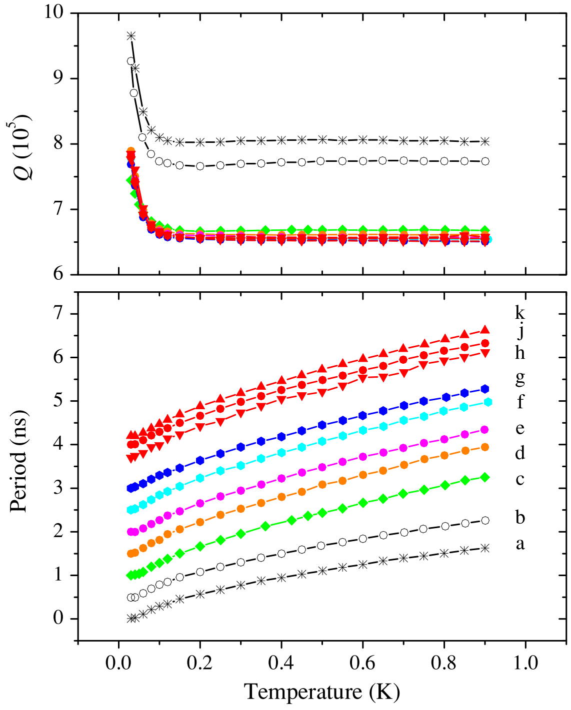

FIG. 4 (color online). Resonant period and Q value as a function of temperature for different samples. (a) Empty cell, (b) inert layer film (21.5μ mol/m^2), (c) liquid-solid coexistence sample, (d) high purity solid ^{4}He (0.3 ppm ^{4}He), freezing completed at 1.4 K, (e) high purity solid, freezing completed at 1.9 K, (f) solid ^{4}He with 30 ppm ^{3}He, freezing completed at 1.5 K, (g) solid ^{4}He with 300 ppm ^{3}He, freezing completed at 1.6 K, (h),(j),(k) high purity solid, freezing completed at 1.6 K with oscillation speed of 1.8, 7.3, and 73 μm/s, respectively. Scan (j) is also shown in Fig. 3. The speeds for all other data sets are between 7 and 11 μm/s. Period readings are shifted for clarity. The mechanical Q's of the different samples are shown in the top panel with the same symbols as the period data. The Q's for all the solid samples overlap each other.

warming scan (from 30 mK) overlap perfectly with the fourth cooling scan up to 1.5 K. When the scan was extended to 2.6 K and cooled down, as in the fourth cycle, the period below 1.5 K was found to be reduced by 1 ns. We think this is a consequence of the lowering of the freezing temperature of helium or other liquid in a confined geometry [27]. When the solid in the Vycor pores is melted as the cell is warmed above 2 K, the liquid immediately refreezes into the bulk space in and outside the capillary inserted inside the Vycor disk. Thus each warming-cooling cycle up to 2.6 K results in the loss of a small fraction (1 out of 5000) of the solid inside the Vycor pores. The solid helium from the Vycor pores that accumulated near the capillary at the center of the TO does not contribute any measurable moment of inertia. Other than the 1 ns drop, the period results of the cooling and warming scans between 30 mK and 1.5 K are perfectly reproducible, and there is no sign of any abrupt drop within the resolution of the period measurement. The mechanical Q of the TO loaded with a solid sample is 20% lower than that of the empty cell, and there is no sign of any dissipation "peak" below 200 mK.

Figure 4 shows the low temperature period and Q readings of two other solid samples, a liquid-solid coexistence sample, an adsorbed (inert layer) film sample at a surface coverage below superfluid onset, and solid samples diluted with 30 and 300 ppm of ^{3}He. Since the freezing of ^{4}He in Vycor takes place over a range of pressure and we have no means of recording the pressure, we identified each sample shown in Fig. 4 by the temperature at which freezing is completed. One of the samples we studied shows conversion of liquid to solid down to 1.2 K, where the coexistence boundary is essentially flat. This sample is likely to have a small fraction of liquid down to the lowest temperature. The period versus temperature plots for this liquid-solid coexistence sample and all solid samples including those diluted with ^{3}He impurities below 900 mK are reproducible in cooling and warming scans and are perfectly parallel with each other. The temperature dependence of the period is completely insensitive to the oscillation speed (from 1.8 to 78 μm/s) of the TO. The total change in period between 30 and 900 mK of these samples is 2.5 ns, whereas it is 1.6 ns for the empty TO and the inert layer film sample. These observations are consistent with the interpretation mentioned above that the solidification of helium in the porous structure stiffens the Vycor disk and that the stiffness of the Vycor-solid helium composite continue to increase gradually with decreasing temperature down to 30 mK.

How can we understand the differences in the results of the current and the 2004 KC experiments? We think the bulk solid in the metallic torsion cell is responsible for the period drop found below 200 mK in KC. There is solid helium in the hollow torsion rod in the TO of KC. The contribution of solid helium towards the spring constant of the torsion rod can be calculated from the shear moduli of BeCu and solid helium and also the outer diameter (2.2 mm) and inner diameter (0.4 mm) of the torsion rod. Such a calculation found that a 20% increase in the shear modulus of solid helium below 200 mK will result in a period drop of only ~0.1 ns. Therefore this is not the primary source of the 17 ns drop. In KC, the Vycor disk was first glued with epoxy inside the upper half of the TO, namely, an open cylindrical BeCu cup with its top plate connected to the hollow torsion rod. Then an aluminum end cap was slipped over the container and glued with epoxy (Fig. 1). In gluing the Vycor disk, care must be exercised to keep a thin open gap between the top of the disk and the BeCu plate to allow the infusion of helium into the Vycor disk from the torsion rod. This bulk solid helium layer in the gap has a non-negligible contribution to the rigidity of the TO, because the thickness of the BeCu plate is only 0.5 mm. FEM simulations show that the effect on the resonant period due to the shear modulus increase is inversely proportional to the thickness of the helium layer. This is sensible, since the solid helium layer can be regarded as a glue between the BeCu plate and the rest of the TO, and it is more "effective" if the layer is thin. For a 0.1 mm thick bulk solid helium layer, a 9 ns period change is found for a 20% increase of shear modulus. For a 50 μm layer, a period drop of 16 ns, in good agreement with the experimental value of 17 ns, is found. It is also possible that a small gap may open up between the Vycor and the epoxy or between the epoxy and the cylindrical wall of the torsion cell upon cooling and/or pressurization of the TO cell with the helium sample. Solid helium in such a gap will add to the shear modulus stiffening effect. If a bulk solid ^{4}He layer is in fact responsible for the apparent NCRI, then it is easy to understand why KC found similar ^{3}He impurity and oscillation speed dependences as bulk solid samples [1,2]. The reason why no period drop was found when the cell was filled with a solid ^{3}He sample is that the bulk ^{3}He layer in the KC cell has bcc crystal structure which does not show any shear modulus stiffening at low temperature [28].

Recently, Mi and Reppy carried out a study of solid ^{4}He in Vycor with an aluminum TO with two resonant frequencies [25]. They found very small period drops of 1.4 ns in the low frequency mode and 0.9 ns in the high frequency mode. The authors showed that their results are not consistent with the simple superfluid interpretation [25]. Instead, these period drops are very likely also consequences of the bulk ^{4}He solid in the torsion cell and in the hollow torsion rod. The solid helium in their torsion rod can contribute period drops of 0.3 and 0.1 ns in the low and high frequency modes, respectively. The effect due to the bulk solid layer in the Mi-Reppy cell is much smaller than that in KC, because the thickness of the aluminum plate holding the torsion rod is 10 times thicker. FEM calculations find a solid ^{4}He layer of 0.1 mm will contribute period drops of 0.6 and 0.4 ns, respectively, in the low and high frequency modes for a 20% increase in the shear modulus. Since we do not know the actual thicknesses of the bulk helium layers in the torsion cells, the results of the FEM calculations cannot be quantitatively compared with the experimental results. However, the calculations do find period drops in reasonable agreement to both the KC and Mi-Reppy experiments. In the case of the Mi-Reppy experiment, the FEM results also reflect properly the frequency dependence.

In conclusion, solid ^{4}He confined in Vycor shows no evidence of NCRI. The period drop found in the 2004 experiment has its origin in the stiffening of bulk solid ^{4}He layer in the torsion cell. This experiment raises further doubt on the claim of NCRI in bulk solid ^{4}He. As noted above, if the TOs used in the bulk solid experiments are not as rigid as one assumed, the observed period drops can be easily explained as consequences of shear modulus stiffening.

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[1] E. Kim and M. H. W. Chan, Nature (London) **427**, 225 (2004).

[2] E. Kim and M. H. W. Chan, Science **305**, 1941 (2004).

[3] M. Kondo, S. Takada, Y. Shibayama, and K. Shirahama, J. Low Temp. Phys. **148**, 695 (2007).

[4] A. S. C. Rittner and J. D. Reppy, Phys. Rev. Lett. **98**, 175302 (2007).

[5] Y. Aoki, J. C. Graves, and H. Kojima, Phys. Rev. Lett. **99**, 015301 (2007).

[6] A. Penzev, Y. Yasuta, and M. Kubota, Phys. Rev. Lett. **101**, 065301 (2008).

[7] B. Hunt, E. Pratt, V. Gadagkar, M. Yamashita, A. V. Balatsky, and J. C. Davis, Science **324**, 632 (2009).

[8] H. Choi, D. Takahashi, K. Kono, and E. Kim, Science **330**, 1512 (2010).

[9] R. Toda, P. Gumann, K. Kosaka, M. Kanemoto, W. Onoe, and Y. Sasaki, Phys. Rev. B **81**, 214515 (2010).

[10] D. E. Zmeev and A. I. Golov, Phys. Rev. Lett. **107**, 065302 (2011).

[11] A. D. Fefferman, X. Rojas, A. Haziot, S. Balibar, J. T. West, and M. H. W. Chan, Phys. Rev. B **85**, 094103 (2012).

[12] J. Day and J. Beamish, Nature (London) **450**, 853 (2007).

[13] X. Rojas, A. Haziot, V. Bapst, S. Balibar, and H. J. Maris, Phys. Rev. Lett. **105**, 145302 (2010).

[14] H. Maris and S. Balibar, J. Low Temp. Phys. **162**, 12 (2011).
[15] J. D. Reppy, X. Mi, A. Justin, and E. J. Mueller, J. Low Temp. Phys. **168**, 175 (2012).
[16] A. C. Clark, J. D. Maynard, and M. H. W. Chan, Phys. Rev. B **77**, 184513 (2008).
[17] D. Y. Kim, J. T. West, T. A. Engstrom, N. Mulders, and M. H. W. Chan, Phys. Rev. B **85**, 024533 (2012).
[18] H. J. Maris, Phys. Rev. B **86**, 020502 (2012).
[19] J. R. Beamish, A. D. Fefferman, A. Haziot, X. Rojas, and S. Balibar, Phys. Rev. B **85**, 180501 (2012).
[20] M. H. W. Chan, Science **319**, 1207 (2008).
[21] R. Wanner, I. Iwasa, and S. Wales, Solid State Commun. **18**, 853 (1976).
[22] E. Kim and M. H. W. Chan, J. Low Temp. Phys. **138**, 859 (2005).
[23] D. Y. Kim, S. Kwon, H. Choi, H. C. Kim, and E. Kim, New J. Phys. **12**, 033004 (2010).
[24] N. Mulders, J. T. West, M. H. W. Chan, C. N. Kodituwakku, C. A. Burns, and L. B. Lurio, Phys. Rev. Lett. **101**, 165303 (2008).
[25] X. Mi and J. D. Reppy, Phys. Rev. Lett. **108**, 225305 (2012).
[26] B. C. Crooker, B. Hebral, E. N. Smith, Y. Takano, and J. D. Reppy, Phys. Rev. Lett. **51**, 666 (1983).
[27] E. B. Molz and J. R. Beamish, J. Low Temp. Phys. **101**, 1055 (1995).
[28] J. T. West, O. Syshchenko, J. Beamish, and M. H. W. Chan, Nature Phys. **5**, 598 (2009).