Probable observation of a supersolid helium phase

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When liquid ^{4}He is cooled below 2.176 K, it undergoes a phase transition—Bose–Einstein condensation—and becomes a superfluid with zero viscosity[1]. Once in such a state, it can flow without dissipation even through pores of atomic dimensions. Although it is intuitive to associate superflow only with the liquid phase[2], it has been proposed theoretically[3–5] that superflow can also occur in the solid phase of ^{4}He. Owing to quantum mechanical fluctuations, delocalized vacancies and defects are expected to be present in crystalline solid ^{4}He, even in the limit of zero temperature. These zero-point vacancies can in principle allow the appearance of superfluidity in the solid[3,4]. However, in spite of many attempts[6], such a 'supersolid' phase has yet to be observed in bulk solid ^{4}He. Here we report torsional oscillator measurements on solid helium confined in a porous medium, a configuration that is likely to be more heavily populated with vacancies than bulk helium. We find an abrupt drop in the rotational inertia[5] of the confined solid below a certain critical temperature. The most likely interpretation of the inertia drop is entry into the supersolid phase. If confirmed, our results show that all three states of matter—gas[7], liquid[1] and solid—can undergo Bose–Einstein condensation.

The most direct experiment searching for the supersolid phase in bulk solid ^{4}He (performed by Bishop, Paalanen and Reppy[8]) also used the torsional oscillator technique. The resonant period of the high-Q oscillator shown in Fig. 1 is given by $2\pi\sqrt{I/G}$, where I is the moment of inertia of the torsion bob, which contains helium, and G is the torsional spring constant of the Be-Cu torsion rod. A small hole drilled through the centre of the torsion rod allows the introduction of helium into the torsion bob. The oscillator is driven and maintained at resonance by a pair of electrodes. The onset of superfluidity in the helium inside the torsion bob decreases I, and hence decreases the resonant period. Bishop *et al.*[8] made measurements of solid helium from 25 to 48 bar, and concluded that if there is a supersolid state, then either the supersolid fraction (the fraction of ^{4}He atoms participating in superflow) is less than 5×10^{-6} or the critical velocity is less than 5 μm s^{-1}. (The critical velocity is the maximum velocity of superflow without any detectable dissipation.)

In contrast to the results of Bishop *et al.*[8], our torsional oscillator measurements on solid helium grown inside a porous Vycor glass disk show a decrease in the resonant period, characteristic of entry into a supersolid state. The continuous pore space, constituting 30% of the total volume in Vycor, appears under the transmission electron microscope as a network of randomly and multiply interconnected cylindrical channels of about 7 nm diameter and 30 nm length[9]. There have been a number of experiments[10–15], including a torsional oscillator measurement by Brewer and collaborators[10,11], studying the solidification of ^{4}He inside Vycor glass. ^{4}He remains liquid down to temperature $T = 0$ K, unless a substantial pressure is applied to the sample. Below 1.3 K, this freezing pressure is essentially constant at 25 bar. Inside Vycor glass, however, a pressure close to 40 bar is required for solidification[10–15]. At low temperature in the presence of ^{4}He vapour, an amorphous surface film is adsorbed on the walls of the pores by the van der Waals potential. Because of lattice mismatch, this surface film is not favourable for the nucleation and continued growth of solid as the pressure is increased and brought towards the bulk freezing pressure. This means that freezing is initiated from the liquid in the centre of the pore by homogeneous nucleation of crystallites of radius limited by the pore size. The overpressure required to seed a crystallite of

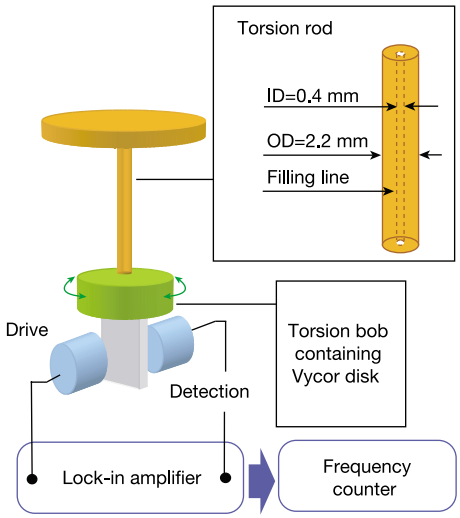

Figure 1 Torsional oscillator used in this experiment. The design of the oscillator follows those used by Reppy and collaborators[18]. The Vycor glass disk has a diameter of 15 mm and a thickness of 4 mm. The cylindrical drive and detection electrodes are aligned off-centre from, and are capacitively coupled to, the central electrode attached to the torsion bob. The signal from the detection electrode (proportional to the amplitude) is sent to the lock-in amplifier through a current preamplifier. The lock-in provides a driving voltage, which controls the amplitude of oscillation, to complete the phase-locked loop and keep the oscillator in resonance. The mechanical Q of the oscillator is 10^6 at low temperature, allowing the determination of the resonant period to a precision of 0.2 ns. The resonant period is 967,640 ns when the Vycor disk is empty, and is 971,900 ns near 0.2 K when pressurized with solid ^{4}He at 62 bar. Measurements were also made with a dummy torsional cell with the Vycor glass disk replaced by a solid brass disk.

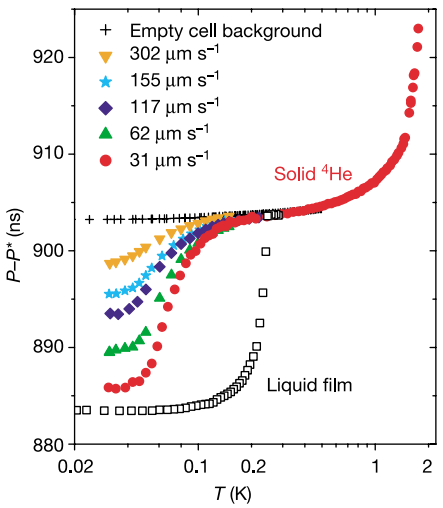

Figure 2 Resonant period as function of temperature of solid ^{4}He in Vycor glass. The resonant period for different oscillation amplitudes—and hence different velocities of the rim of the Vycor disk, v_{rim}—is shown. A drop in the period (ΔP), signifying the transition into the supersolid phase, is seen below 175 mK. Although the magnitude of ΔP depends strongly on the rim velocity, no such dependence of the period is seen above the transition temperature. For comparison, the empty (without helium) cell period, and the period of an atomically thin liquid film adsorbed on the walls of the internal pore space of Vycor, are also shown. The film measurement, showing a superfluid transition at 250 mK, is carried out with the same torsion cell. For easy comparison, 4,260 ns is added to the empty cell data and 3,290 ns to the film data. The ordinate shows $P - P^*$, the difference of the actual period P and $P^* = 971{,}000$ ns.

radius r in this geometric model of freezing is predicted to be proportional to the interfacial tension between the liquid and solid phases and inversely proportional to r. The large (15 bar) overpressure observed for solidification of ^{4}He in Vycor reflects its small pore diameter[10–15]. Solid ^{4}He grown inside Vycor with a tortuous porous structure and small characteristic pore diameter is likely to be heavily populated with vacancies.

The solid sample is grown and its density kept constant via the standard blocked capillary method[10–15]. We found that by cooling (from 3 K) a liquid sample of 75 bar in the torsion cell, the resultant pressure of the solid sample below 1 K is typically 62 ± 2 bar. The pressure is determined by an *in situ* strain gauge attached directly on the outside of the torsion cell. We have intentionally chosen this high pressure so that we can be sure that our helium sample in Vycor glass is deep in the solid phase. Figure 2 shows the resonant period of our Vycor disk torsional oscillator as a function of temperature. As the mechanical Q of the oscillator is 10^6, and the resonant period is of the order of 1 ms, the equilibration time for a new resonant period reading due to any change in the experimental condition—for example, a change in temperature—is expected and observed to be of the order of 1,000 s or 15 min.

Below 80 mK, however, the equilibration time of the resonant period depends on whether the measurement is taken during warming or cooling of the torsional cell. In a warming scan, the equilibration time is the same as that at higher temperatures; but in a cooling scan, this time lengthens noticeably with decreasing temperature and exceeds 60 min below 40 mK. Our period data are obtained during warming scans after waiting for complete equilibration at the lowest temperature. The resonant period was measured with different amplitudes of oscillations. The amplitude, proportional to the linear velocity of the rim of the Vycor disk, can be calculated from the a.c. voltage induced on the detection electrode. The resonant period above 0.2 K does not depend on the rim velocity, and for temperatures above 0.5 K it is similar to that found by Brewer and collaborators[10,11]. These authors did not extend their measurement below 0.5 K. The central result of our experiment is the additional drop in the period (ΔP) that begins at 175 mK. This drop is consistent with the confined solid entering the supersolid phase. ΔP is strongly attenuated with increasing amplitude of oscillation (that is, higher rim velocity of the Vycor). Figure 3 shows ΔP in the low-temperature limit as a function of the rim velocity. The transition temperature at 175 mK, however, does not appear to depend on the rim velocity.

For a disk oscillating at a fixed angular velocity, the local linear velocity ranges from zero up to a maximum rim velocity. Supersolid decoupling occurs for solid embedded in the region where the local velocity is smaller than the critical velocity. The temperature dependence of ΔP shown in Fig. 2 is probably a reflection of the velocity profile of the Vycor disk modulated by the temperature-dependent critical velocity of the supersolid. Although we do not know the exact functional form, the critical velocity is zero at the transition temperature (175 mK), increases with decreasing temperature, and saturates near 300 μm s^{-1} in the low-temperature (30 mK) limit. As a comparison, we also show in Fig. 2 the superfluid response of an atomically thin liquid film adsorbed in Vycor with zero-temperature ΔP of the same order as that of the solid sample. The measurements were carried out with the same torsional cell. In addition to the different temperature dependence, ΔP of the film as shown in Fig. 3 is independent of the rim velocity. This is not surprising, as the critical velocity of an atomically thin liquid ^{4}He film adsorbed in Vycor exceeds 200 mm s^{-1} (ref. 16), which is a factor of 700 higher than the maximum rim velocity of 300 μm s^{-1} used in this experiment.

Superfluid helium adsorbed inside an oscillating porous disk must execute a tortuous path of potential flow defined by pore structure[17]. As a result, a fraction, χ, of the superfluid moment of inertia, I_S, remains effectively locked to the porous disk and the observed period drop is proportional to the unlocked portion, $(1 - \chi)I_S$. If we make the assumption that the χ factor of supersolid in Vycor is zero, then the ΔP_0 of 22 ns shown in Fig. 3 is a direct measure of the zero-temperature and low-velocity supersolid

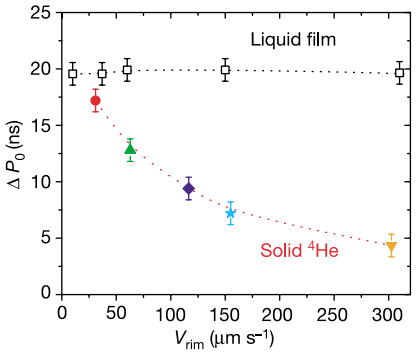

Figure 3 ΔP at the low temperature limit, ΔP_0, as a function of rim velocity of the Vycor disk. ΔP_0 values are deduced by subtracting the measured periods at 30 mK from the (shifted) empty cell period, as shown in Fig. 2. Whereas ΔP_0 of the liquid film is independent of rim velocity, it is strongly attenuated with higher rim velocity in solid ^{4}He. This plot shows that the critical velocity of supersolid at 30 mK is of the order of 300 μm s^{-1}. It also shows that ΔP_0 extrapolates to 22 ns in the limit of low rim velocity.

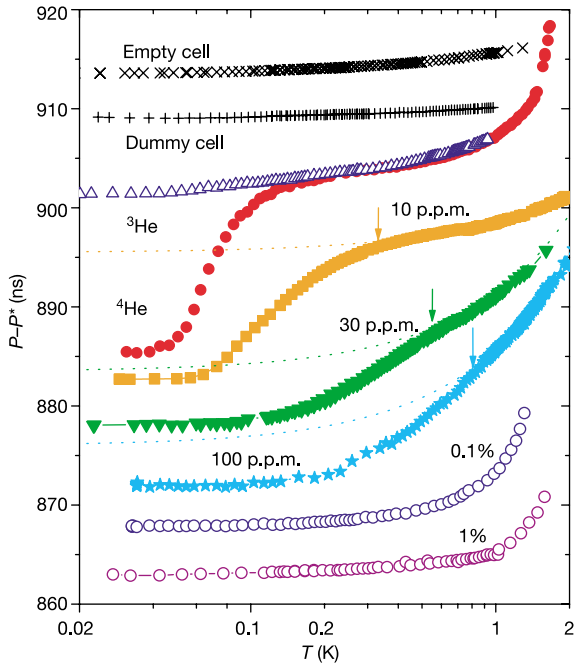

Figure 4 Resonant periods as a function of temperature for a variety of solid helium samples. The period scale shown corresponds to that for solid ^{4}He. As in Fig. 2, the ordinate shows $P - P^*$, the difference of the actual period P and $P^* = 971{,}000$ ns. The period data for other samples are shifted for clarity and easy comparisons. All measurements were made with the rim velocity of the Vycor disk near 30 μm s^{-1}. The plots show that the period drop effect is not related to the stiffening of bulk solid helium in the torsion rod. The effect is not seen in pure ^{3}He, and is not seen in solid mixtures with ^{3}He concentration exceeding 0.1%. The resonant periods in these samples are independent of the rim velocity of the Vycor disk. A period drop is found for mixtures with 10, 30 and 100 p.p.m. of ^{3}He. As in pure ^{4}He, the size of the drop in these samples with low ^{3}He concentrations is rim-velocity dependent. The dotted lines extrapolated smoothly from high temperature are the expected background period in the absence of period drops. The vertical arrows mark the transition temperatures of these samples. The ^{3}He concentrations listed are the average concentration of the solid inside the Vycor and in the capillary leading to the cell. The actual ^{3}He concentration inside the Vycor, particularly for low-concentration samples, could be quite different from the listed values.

moment of inertia. The resonant period increases by 4,260 ns when the porous Vycor glass disk in the torsion bob is filled with solid ^{4}He at 62 bar, therefore the supersolid fraction—the fraction of ^{4}He atoms that participate in superflow—is 5 parts in 10^3. On the other hand, if we assume that the χ factor of supersolid is 0.8, the same as that of superfluid in Vycor[18], then the supersolid fraction is 25 parts in 10^3. If we assume that the ^{4}He atoms in the supersolid fraction are uniformly distributed in the pore space and if we neglect interaction between these atoms, then we can use the ideal Bose gas theory to calculate the Bose–Einstein condensation temperature[19]. The calculated transition temperatures are 120 mK and 350 mK, respectively, for supersolid fractions of 5 and 25 parts in 10^3, bracketing the observed transition temperature of 175 mK.

In order to rule out non-supersolid mechanisms for the observed effect, we did a number of control experiments. We made measurements with the same torsional cell with pure solid ^{3}He and with solid ^{4}He diluted with 10, 30, 100, 1,000 and 10,000 p.p.m. of ^{3}He, all pressurized with the same procedures as that for pure ^{4}He, and resulting in a final pressure between 60 and 65 bar. The results of these measurements are shown in Fig. 4. The observed ΔP seen in pure ^{4}He is not seen for solid ^{3}He, and is also not seen for solid mixtures with ^{3}He concentrations exceeding 0.1%. The fact that the effect is not present in ^{3}He, a Fermi system, is reassuring. It is also reasonable that the addition of enough ^{3}He atoms is effective in quenching the supersolid phase. The behaviour found in samples with even lower ^{3}He concentrations is very intriguing. Besides reducing the magnitude of ΔP, the introduction of the minute amount of ^{3}He also broadens the transition and increases the transition temperature.

Figure 4 also shows measurements made with a dummy torsional oscillator consisting of a torsion rod that is identical to the normal torsional cell but with a torsion bob containing a solid brass disk rather than a Vycor glass disk. During measurements, the hole in the torsion rod is pressurized with solid ^{4}He (using the exact same procedure as that for the Vycor torsional cell) to a final pressure of 62 bar. The resonant period is temperature independent, showing no decrease at low temperature, similar to that of the empty cell. This means the effect that we have seen with pure solid ^{4}He and with ^{4}He diluted with small ^{3}He impurities occurs inside the torsion cell, and is not related to changes occurring in the bulk solid helium inside the torsion rod. Owing to the freezing of dislocations, solid helium inside the torsion rod stiffens at low temperature. This stiffening can contribute to the torsional spring constant of the torsion rod[20] and lower the resonant period. We have, however, taken the precaution of using an especially thick (2.2 mm diameter) torsion rod, so that this effect can be ignored. This assumption is confirmed by our measurements with the dummy cell.

It could be argued that solidification inside the Vycor glass is never complete, and that there is a persistent thin liquid film even at 62 bar, 20 bar above the reported solidification pressure. But there are observations that are not consistent with this picture. The temperature dependence of the supersolid is distinctly different from that of a liquid film, and the critical velocity observed for the solid is at least 700 times smaller than that in a film[16]. Figure 4 shows that the introduction of 0.1% of ^{3}He into solid ^{4}He completely eliminates the observed drop in the period. This is in strong contrast to the findings in liquid films. The addition of ^{3}He into a liquid ^{4}He film smoothly decreases the superfluid transition temperature, and a very high concentration of ^{3}He is needed to completely quench the transition[21]. The transition temperature of a pure ^{4}He film at 150 mK was found to decrease to below 20 mK only when the amount of ^{3}He exceeds 20% of all the ^{4}He in the adsorbed film, including that in the amorphous solid layer[21].

In conclusion, the most reasonable interpretation of the observed period drop is that it is a signature of transition into the supersolid state. The microscopic origin of this effect is not understood. We have noted that the solid ^{4}He grown in the Vycor pores is heavily populated with vacancies and defects. It seems that these vacancies may be responsible for enhancing Bose–Einstein condensation of the confined solid ^{4}He into the supersolid phase. □

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Partial order in the non-Fermi-liquid phase of MnSi

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Only a few metallic phases have been identified in pure crystalline materials. These include normal, ferromagnetic and antiferromagnetic metals, systems with spin and charge density wave order, and superconductors. Fermi-liquid theory provides a basis for the description of all of these phases. It has been suggested