Is solid helium a SUPERSOLID?

Robert Hallock

Recent experiments suggest that helium-4 atoms can flow through an experimental cell filled with solid helium. But that incompletely understood flow is quite different from the reported superfluid-like motion that so excited physicists a decade ago.

The road to scientific discovery and verification is typically not the clean, smooth path portrayed in textbooks. Rather, it often includes wrong turns, controversy, unsubstantiated claims, misinterpretations, and even real surprises. Eventually scientists sort it all out, and the story gets distilled in the retelling to the point that the life is taken out of it and the reader is left with a few brief facts.

In the end, the pursuit of new science is stimulating, exciting, and often a great pleasure. But frankly, it is also at times frustrating. So has been the case with solid helium. And the story of that material is a classic tale of the reality of scientific work—a fascinating, wide-ranging, still incomplete account that demonstrates the power of science to discover.

A strange transition

Helium-4 is a remarkable element. At a pressure of 1 atmosphere (1.01 bar), it remains a gas until cooled to 4.2 K, at which point the ^{4}He vapor pressure and atmospheric pressure are equal and the ^{4}He liquefies. Upon further cooling in equilibrium with its vapor, at about 2.2 K the liquid undergoes a spectacular phase transition to a superfluid state. And due to quantum mechanical zero-point motion, it remains a liquid upon cooling to as close to absolute zero as physicists have been able to reach. It solidifies only with the application of some 25 atm pressure at very low temperature.

Theorists, notably Alexander Andreev and Ilya Lifshitz at the Institute for Physical Problems in Moscow (now the P. L. Kapitza Institute for Physical Problems) and Geoffrey Chester at Cornell University, investigated solid ^{4}He from several angles. By around 1970 they had concluded that it might be possible for the solid to undergo a rather strange transition to a state of matter in which crystalline order and Bose condensation coexist. At the time, some theorists thought that the solid ^{4}He possesses mobile ground-state vacancies. Since the motion of vacancies comes with motion of atoms, Chester and other theorists speculated that the solid could demonstrate superfluid-like properties: Atoms of ^{4}He might be able to move coherently through an otherwise well-ordered solid.

The startling prediction soon aroused experimental interest. Motivated by the idea of superfluid-like motion in a solid, Anthony Leggett, then at the University of Sussex, proposed a simple experiment that could put it to the test: Fill a container with solid ^{4}He, place it at the end of a torsion rod, and measure the period of torsional oscillation as a function of temperature.[1] If some of the ^{4}He atoms were to decouple from the rest of the solid, that decoupling would manifest itself as a change in the oscillation period. The basic technique had been pioneered in the 1940s in a famous experiment by Elevter Andronikashvili,[2] then a visiting scientist at the Institute for Physical Problems, who used a stack of closely placed disks at the end of a torsion rod and detected a temperature-dependent superfluid component for liquid ^{4}He as mass between the plates decoupled from them.

By the late 1970s, experimentalists were investigating whether there was something unusual about solid ^{4}He. In one of those studies, Dennis Greywall, then at Bell Labs, put the solid in two chambers that could communicate through narrow, solid-filled channels.[3] If the solid were to transition to an un-

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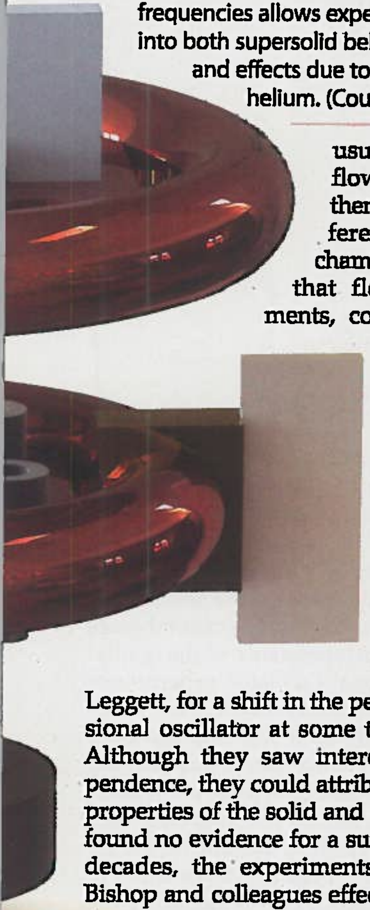


A torsional oscillator with two resonant frequencies allows experimenters to gain insight into both supersolid behavior in solid helium-4 and effects due to the stiffness of the helium. (Courtesy of Eunseong Kim.)

usual state in which it could flow through the channels, then an initial pressure difference between the two chambers would relax due to that flow. Greywall's experiments, conducted at several average cell pressures, resulted in the unambiguous conclusion that pressure differences did not relax: There was no supersolid. Within a few years of Greywall's investigations, David Bishop and Mikko Paalanen at Bell Labs and John Reppy at Cornell looked, per Leggett, for a shift in the period of a solid-filled torsional oscillator at some transition temperature.[4] Although they saw interesting temperature dependence, they could attribute it to other, nonsuper properties of the solid and concluded that they had found no evidence for a supersolid. For nearly two decades, the experiments by Greywall and by Bishop and colleagues effectively killed any enthusiasm to search for a supersolid state of matter.

Then a new result with an unusual interpretation rekindled enthusiasm for further study of the solid. Working at the University of California, San Diego, Pei-Chun Ho, Ian Bindloss, and John Goodkind reported an acoustic anomaly present in their studies of solid ^{4}He with a ^{3}He impurity of 14.5 parts per million: a small shift in the velocity of sound and a peak in the sound attenuation at a specific frequency-dependent temperature.[5] They interpreted their result to be consistent with the presence of a Bose-condensed state above the shift temperature.

Few paid much attention, but Moses Chan at the Pennsylvania State University did. In 2004 he and Eunseong Kim reported on an extensive series of torsional-oscillator measurements[6] that once again explored the behavior of solid ^{4}He in the manner recommended by Leggett.

Since the early theoretical work had suggested that vacancies were likely important for a supersolid to exist, Kim and Chan first filled an experimental cell with Vycor, a nanoporous glass with a large surface area. They reasoned that since the solid ^{4}He lattice and Vycor surface were incommensurate, the introduction of the glass would cause many vacancies to be present in the ^{4}He.

The two researchers quickly saw something unusual. As illustrated in figure 1, as they lowered the temperature below 0.15 K or so, the period of the torsional oscillator shifted in a manner consistent with a temperature-dependent decoupling of roughly 1% of the ^{4}He mass from the oscillator. Their interpretation, anticipated by Leggett in his paper outlining the oscillator technique[1] and consistent with what was then known about liquid and solid ^{4}He, was that they had observed evidence for a transition to the elusive supersolid. After a series of careful control experiments to rule out various possible experimental artifacts and document the effect of ^{3}He impurities, they announced the likely discovery of a new state of matter, the supersolid.

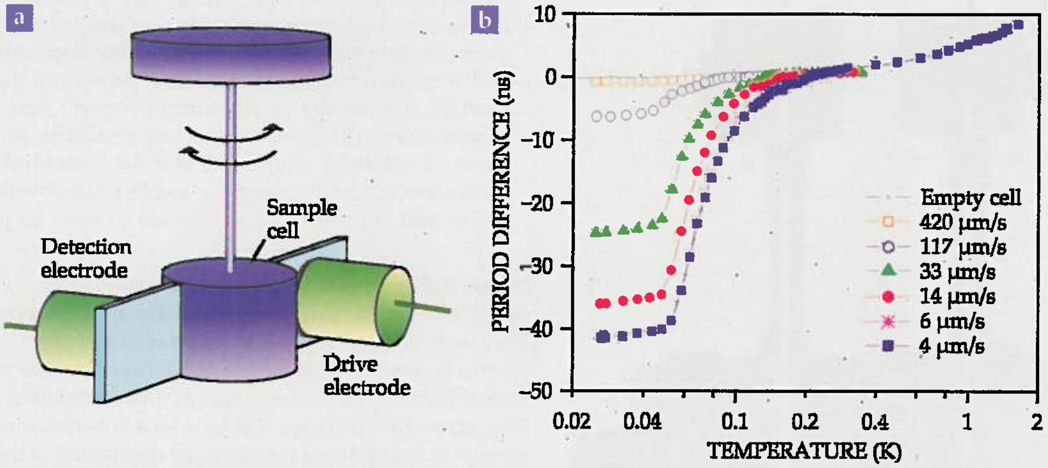
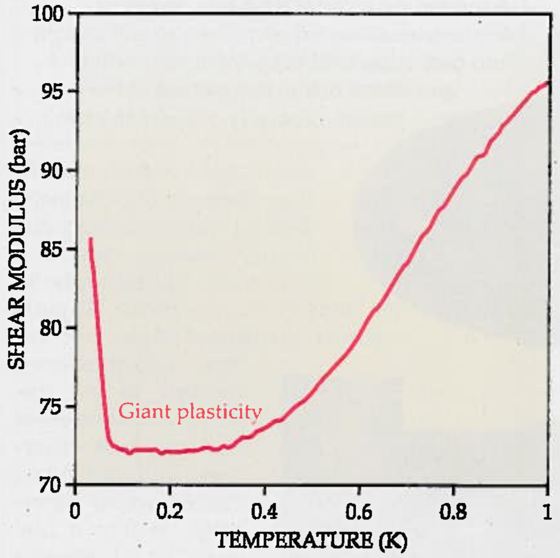


Figure 1. A sensitive torsional oscillator filled with solid helium appeared to indicate supersolid behavior in a 2004 experiment by Eunseong Kim and Moses Chan. **(a)** In a torsional-oscillator experiment, the period of the torsional oscillator is related to the moment of inertia of the sample cell and its contents and to the spring constant of the torsion rod. If the contents decouple from the motion, the oscillation period shifts. The capacitively coupled electrodes shown in this schematic of the Kim and Chan oscillator helped to maintain the oscillation and measure its frequency. **(b)** Kim and Chan found a period shift that depended on temperature and the peak oscillator velocity and that was consistent with expectations based on many years of experience with the superfluid transition in liquid helium. A cell filled with ^{4}He frozen in porous Vycor glass and one filled only with bulk solid ^{4}He yielded similar results.[6] (Adapted from PHYSICS TODAY, November 2004, page 23.)

Figure 2. The shear modulus of solid helium-4 at first falls when the temperature is decreased below 1 K. But at around 0.3 K the fall levels off, and near 0.1 K the modulus abruptly rises. That sharp increase has a temperature dependence much like that observed[6] by Eunseong Kim and Moses Chan for the period shift in a torsional oscillator filled with solid ^{4}He (see figure 1). At temperatures approaching absolute zero, solid ^{4}He has a stiffness of 127 bar (1 bar = 0.987 atm). Thus the trough centered at 0.2 K represents a substantial softening, called giant plasticity by Ariel Haziot and colleagues, who took the data shown here. (Adapted from ref. 15.)



The elusive nature of supersolidity

Several research groups worldwide followed up on the work of Kim and Chan with similar torsional-oscillator experiments. Most reproduced a frequency shift that could be interpreted as a manifestation of decoupled mass, consistent with the presence of a supersolid. Those confirmations became big news in the world of condensed-matter physics. But, curiously, the percentages of decoupled mass in the replication experiments spanned two orders of magnitude (and eventually would grow to more than four orders of magnitude). Not everyone in the field was convinced that a supersolid was present; Alexander Balatsky (Los Alamos National Laboratory) and his colleagues were particularly noteworthy critics of the supersolid interpretation. More important, the original theoretical scenario with ground-state vacancies was shown to be irrelevant to ^{4}He, but disorder-induced superfluidity was observed in numerical simulation.[7] The claim that a supersolid had been spotted certainly began to look robust to many.[8]

Among the criticisms of the supersolid interpretation was that dislocations or structural effects could be responsible for the observed frequency shifts. By 2007, James Day and John Beamish at the University of Alberta had measured the shear modulus of solid ^{4}He and found a striking resemblance between the temperature dependence of the oscillation period in the torsional-oscillator experiments and the temperature dependence of the shear modulus in their own work.[9] To their surprise, the shear modulus increased as the temperature was lowered below 0.15 K, and the increase took place at the same temperature at which the period shift had been seen in the torsional oscillators. They attributed the increase in stiffness to the temperature dependence of the mobility of dislocations in the solid. After all, dislocations could be pinned by impurities such as the ^{3}He present at about 200 parts per billion in ordinary helium from natural-gas wells.

The Day and Beamish work immediately gave rise to the question, Was supersolidity responsible for the shift in the shear modulus, or was it a change in structural properties that yielded the supersolidity? A more troubling variant also presented itself: Perhaps the change in structural property was the whole story and there was no supersolidity at all. Early calculations suggested that the structural effects were not large enough to explain the observed shifts, and supersolidity continued to have its pro-

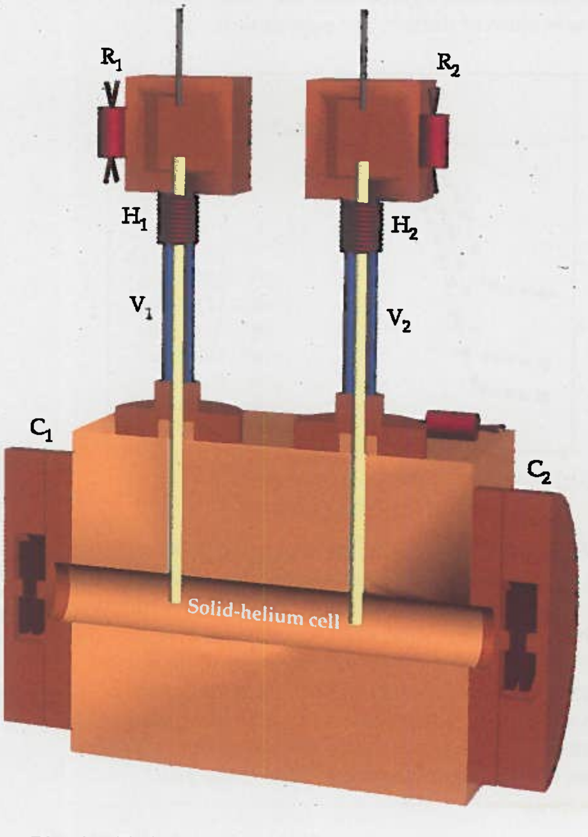


Figure 3. The UMass sandwich was designed to determine if mass could be made to flow through a cell filled with solid helium-4. The idea was to place two warmer reservoirs (R_1, R_2) of superfluid on either side of the colder solid ^{4}He, create a chemical-potential difference across the reservoirs, and look for a flux-indicating change in the reservoir pressures. As mentioned in the text, the solid pressure should be above the value for which bulk superfluid and solid coexist in equilibrium; therefore, the superfluid is confined to regions of porous Vycor glass (V_1, V_2) that penetrate halfway into the solid-filled cell. The experimental cell has a cryogenic capacitive pressure gauge (C_1, C_2) on each end, and heaters (H_1, H_2) control the temperature in the reservoirs. Other gauges and thermometers not labeled measure the pressure and temperature in the reservoirs. (Adapted from ref. 17.)

ponents. But the Day and Beamish work raised caution flags, as did other experiments, notably an unpublished investigation by Reppy.[10]

Structural effects came to the fore again after a new round of calculations by Humphrey Maris (Brown University) and others. The theorists showed that the moment of inertia of the experimental torsion cell could be subtly modified if the structure of the solid ^{4}He inside it changed, and various researchers argued about whether such structural changes could influence the oscillation period enough to cause the experimentally observed period shifts.

Experimentalists responded by creating stiffer cells and found that investigations with those cells showed less of a supersolid signal. By 2012 the Penn State group had completely redesigned its torsional oscillator, and the apparent mass-decoupling signal observed with the new apparatus was greatly reduced—in fact, it disappeared to within experimental error. The Penn State group retracted its interpretation of its earlier work and announced that it now thought that supersolid was not present in a cell filled with Vycor. More recently Chan and colleagues announced a similar conclusion for bulk solid ^{4}He: No supersolid is present at a level greater than approximately four parts per million.[11]

Torsional oscillators with two resonant frequencies were first used in studies of solid helium by Haruo Kojima and his collaborators at Rutgers University. Since then they have allowed experimenters to make progress in separating the behavior expected due to a supersolid from effects due to the stiffness of the solid ^{4}He. In recent work with the double-frequency cell shown on page 30, Kim (now at the Korea Advanced Institute of Science and Technology) and collaborators saw no evidence for a supersolid.[12] But Reppy and his Cornell team found a persistent though small supersolid-like signal in their double-frequency oscillator experiment.[13] In ongoing work, a group led by John Saunders at Royal Holloway, University of London, has seen a signal similar to that observed at Cornell, but the London researchers are not convinced that it is due to a supersolid.

The observed signals appear in the same temperature range as does the shear modulus change, and so the interpretation of the observations warrants considerable caution. There remains a lingering disagreement among some in the torsional-oscillator community about the evidence for a supersolid state, but the consensus is that the torsional-oscillator work has surely failed to provide convincing evidence for the presence of a supersolid and perhaps has failed to provide any evidence at all. But solid ^{4}He has yet to reveal all of its secrets.

Outside the torsional-oscillator community, other experiments have brought different techniques to bear on the study of solid ^{4}He. Some have attempted to squeeze a solid ^{4}He lattice with a diaphragm and thus drive a flow of solid ^{4}He through small solid-filled channels.[14] Once the experimenters eliminated artifacts, those studies confirmed to very high accuracy Greywall's original null result: No flow of the solid ^{4}He had taken place. Kojima took a classic experiment from the study of superfluids and applied it to solid ^{4}He. He looked for the propagation of fourth sound—that is, a wave that propagates in a ^{4}He-filled microporous media but for which only the superfluid component of the ^{4}He is in motion—but was unable to see any evidence of it in a solid-filled cell.

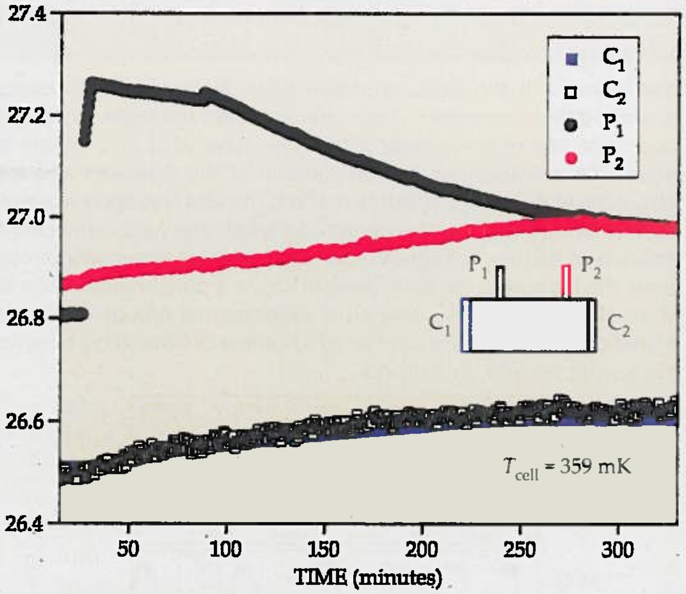


Figure 4. Evidence for mass flow through the UMass sandwich cell filled with solid helium. At a time of about 25 minutes, atoms are injected into the left superfluid-filled reservoir of the device, as reflected by the sharp increase in pressure registered by gauge P_1. The subsequent increase in pressure seen by gauge P_2 at the right-most reservoir demonstrates the mass flow. During the flow, the density of the solid increases, as recorded by *in situ* pressure gauges C_1 and C_2. In several experiments, including the one shown here, the pressure measured by P_2 grew nearly linearly at a rate independent of the pressure difference that drove the flow. Such behavior is consistent with expectations for superfluid flow at a limiting, or critical, velocity.

More and more curious

Following up on the observation of an increase in the shear modulus at low temperatures, Ariel Haziot, Sébastien Balibar (École Normale Supérieure in Paris), Beamish, and collaborators studied in great detail how the ^{4}He stiffness depends on temperature.[15] The researchers confirmed that when solid ^{4}He is cooled to low temperatures, the shear modulus decreases before it eventually increases again; figure 2 shows some of their data. They termed the softening of the solid represented by the resulting broad minimum "giant plasticity." Possibly the effect will have implications in the wider realm of materials science.

A classic signature of the phase transition to a traditional superfluid is a peak in the heat capacity. Chan and his group looked for an analogous signal in solid ^{4}He. They found a small peak in the heat capacity near the temperatures that in previous experiments marked the changes in torsional-oscillator frequency and shear modulus. But in contrast to the earlier results, the temperature at which the peak was observed did not change when the concentration of ^{3}He impurities in the solid was varied. The physics behind the heat capacity peak still needs to be fully understood.

Other aspects of solid ^{4}He also lack an explanation. For example, Kimitoshi Kono and coworkers at the RIKEN research institute in Japan saw various effects in torsional-oscillator experiments in which an overall rotation of the entire apparatus was superposed on the usual torsional oscillation. Some rotation studies, notably those of Minoru Kubota at the University of Tokyo, have been interpreted in terms of a vortex model proposed by Philip Anderson at Princeton University, but a detailed overall understanding is still lacking. Donald Candela (University of Massachusetts Amherst), Neil Sullivan (University of Florida), and their collaborators have recently studied the temperature dependence of the nuclear magnetic resonance relaxation time of ^{3}He in a solid ^{4}He sample containing an admixture of 16 parts per million of ^{3}He. They found a peak but concluded it was not related to supersolidity. Curiously, the peak appeared at about the same temperature as the shift in sound velocity seen much earlier by Goodkind's group.[5] It's not yet known whether the two effects are related.

The UMass sandwich

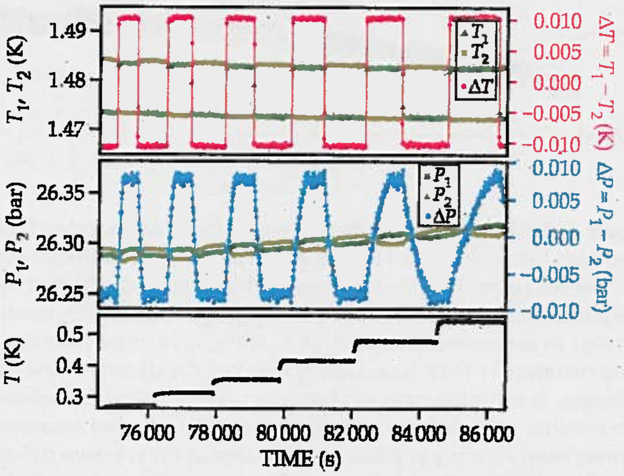
My colleagues and I at UMass took a conceptually different approach to the study of solid ^{4}He. Rather than to squeeze the solid directly, as some have done, our notion was to sandwich the solid between two regions of superfluid liquid and then create a chemical-potential difference between the two reservoirs—for example, by increasing the pressure in one of the superfluid-filled regions. If ^{4}He atoms were to pass through the solid, we would observe a pressure increase in the other superfluid-filled region. Figure 3 shows our device, the UMass sandwich.

One tricky issue we had to work around was that we could not conduct the experiment with the solid on the melting curve—that is, at a temperature and pressure for which bulk superfluid and solid ^{4}He coexist in equilibrium. If we were to do so, we would leave open the possibility that any mass flux we found was due to well-understood liquid flow, not solid flow. In the UMass sandwich, therefore, the superfluid liquid in contact with solid ^{4}He is confined in the pores of Vycor, an environment that raises the freezing pressure of the superfluid, and the solid is at a pressure above that of the melting curve. Initial observations, presented in figure 4, showed that a mass flux was indeed present.[16] Moreover, the flux was nearly independent of the driving pressure gradient—a classic signature of superfluid flow at its limiting, or critical, velocity. And, unexpectedly, an increase in the solid density accompanied the mass flux.

Just what carries the flux and why the density increases is still uncertain and remains a bit controversial. To explain both phenomena, Şebnem Söyler (then at UMass) and colleagues proposed that the cores, or boundaries, of edge dislocations, such as the one illustrated in figure 5, might be responsible for the flux.[7] From their simulations they concluded that such cores would have a superfluid density along them. Their explanation of why flux is accompanied by density increase is that the flow is due to effectively one-dimensional superfluidity along the cores of edge dislocations but that some of the flow-

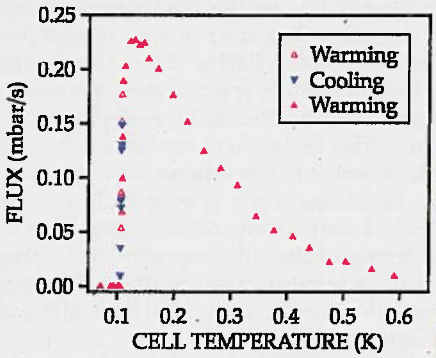
Solid helium and the fountain effect

In early work with the UMass sandwich (figure 3), we created a chemical-potential difference between two superfluid-filled reservoirs by injecting atoms into one of them. More recently we have used an alternate approach: We manipulated the temperature of the reservoirs and took advantage of the so-called fountain effect. The idea is to apply a temperature difference between two superfluid-filled reservoirs connected by channels so narrow that only a superfluid component can flow through them; the induced superfluid flow will create a pressure difference between the reservoirs. We, and other experimental groups conducting related experiments, have uncovered a number of fascinating behaviors that are not yet fully understood.



The two colored plots above, obtained with ordinary helium from natural-gas wells, show that an alternation of the reservoir temperatures T_1 and T_2 results in an oscillating pressure difference, $\Delta P = P_1 - P_2$. The rate at which ΔP changes is a measure of the flux of ^{4}He through a cell filled with solid helium—the meat of the UMass sandwich—and it depends on the temperature T of the cell, indicated in the bottom plot of the panel. The curvature in the ΔP data suggests that the flux can be a nonlinear function of the driving chemical-potential difference.

For the data presented in the graph below, the magnitude of the alternating temperature difference across the reservoirs is fixed. As the temperature of the solid-filled cell decreases, the flux through the cell (more precisely, the initial rate of change of ΔP across the superfluid reservoirs) at first increases with a universal functional dependence on temperature—that is, one independent of sample history or impurity concentration. At a lower, concentration-dependent temperature, the flux drops. For ordinary well helium, which has a ^{3}He impurity of about 200 parts per billion, the extinction is not complete, and with further reduction in temperature the flux rises again. For the data shown here, the ^{3}He concentration is larger, about 19.5 parts per million, and the extinction appears to be complete. Evidently, ^{3}He blocks some part of the flux pathway once the solid-filled cell reaches a specific temperature determined by the ^{3}He concentration.



The drop in flux is rather dramatic: A temperature decrease of 1.5 mK is enough to choke off the superfluid flow. But if the temperature is then increased above the extinction temperature, T_d, the flux recovers within a few hundred seconds.

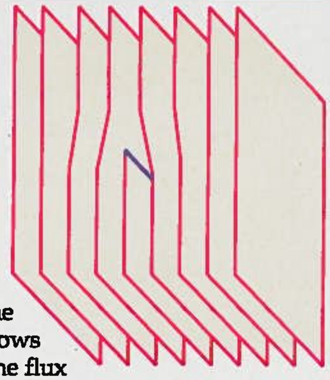


Figure 5. An edge dislocation in an otherwise perfect crystal is analogous to a sheet of paper that is partially inserted into an existing sheaf. The planes illustrated here represent planes of atoms in the solid helium lattice, and the blue line represents the terminus, or core, of the edge dislocation. The flow of atoms along the core is one suggestion put forth to explain recent experiments.

ing atoms don't make it to the end of the core. As a result, the edge dislocation grows farther into the solid, whose density thereby increases. Others have suggested, in contrast, that the result is due to liquid channels that can form where crystal boundaries meet a container wall.

In more recent experiments with the UMass sandwich, we discovered that the flux can depend on the driving chemical-potential difference; the box on page 34 gives some of the details. Moreover, in contrast to ordinary electric currents, which vary linearly with the applied potential difference, the flux depended on the driving chemical potential raised to a pressure-dependent power near ⅓. When the potential difference was large, the mass flow approached a maximum, temperature-dependent velocity, in a manner similar to what is observed[17] for superfluid ^{4}He. We concluded that whatever carries the flux through the solid seems to have a 1D character. Our results are consistent with the notion of flow along dislocation cores, but they don't prove it.

We also found (and discuss further in the box) that as a sample is cooled below about 0.6 K, the flow increases with decreasing temperature in a universal way—that is, the temperature dependence is independent of sample characteristics. However, the flow then decreases abruptly at a temperature T_d near 0.1 K—the precise value depends on the concentration of ^{3}He impurity—and it can be extinguished completely if the impurity concentration is high enough.[16] In follow-up experiments, we determined that the decrease in flux takes place over a temperature range of just a few millikelvin. More recently Beamish and his colleagues[12] have used a variation of the UMass sandwich to confirm a number of properties that we had observed. Chan's group has undertaken related work[12] and has also observed flow through a thin slab of solid ^{4}He.

A convoluted path

Intriguing questions remain. For example, What carries the flux, and how does it do it? What is the specific behavior of the ^{3}He that allows it to block the flux? The observations to date appear to be consistent with the notion that the flow takes place along edge-dislocation cores. Apparently, the ^{3}He blocks the flux at a concentration-dependent temperature, perhaps by moving to places where dislocations intersect. On the other hand, Beamish and colleagues have pointed out that it is energetically favorable for the blockage to occur at the interface where the solid ^{4}He meets the superfluid in the Vycor pores.[12] Experiments show that the flux recovers within a few hundred seconds when the temperature is raised above T_d, a result that constrains models for what blocks the flux.

We in the field have yet to understand what gives rise to the universal behavior above T_d and what limits the flux at a given temperature. New work with the UMass sandwich shows that the limitation to the flux above T_d is in the solid ^{4}He itself and unrelated to the solid interface with the superfluid in the Vycor.[12]

The story of solid ^{4}He is still unfolding. Can the material exist as a supersolid? If so, the supersolid is nothing like the state originally envisioned by Chester and other theorists more than four decades ago. The pathway to understanding has been convoluted; the real story is unlike the historical summaries in textbooks or review articles that describe in a more direct way our final understanding of a topic. But the journey along a real scientific pathway to discovery can be exciting, especially when one travels with open and friendly colleagues.

I appreciate the numerous conversations with many colleagues, from whom I have benefited in various ways. I am indebted to my collaborators Michael Ray and Yegor Vekhov, who took much of the data produced at the University of Massachusetts Amherst. Our work has been supported by NSF and by trust funds provided by the university.

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