

The original evidence of supersolidity in helium-4 is explained away

The 2004 "discovery" experiment was long thought to be immune to elastic artifacts. But a careful repetition shows that it wasn't.

Helium, the lightest of the noble gases, liquefies at 4.2 K. But because its interatomic attraction is so weak and its zero-point quantum fluctuation so large, it never solidifies at pressures less than 25 atmospheres. At 2.2 K, a fraction of liquid helium-4, the dominant isotope, becomes a superfluid; it flows without viscosity or frictional dissipation.

Around 1970, a number of theorists suggested that even solid ^4He might exhibit superfluid-like behavior. Because lattice vacancies in crystalline ^4He were thought to persist and remain mobile down to absolute zero, it was conjectured that they would condense into a coherent quantum state in which vacancies, and therefore the atoms whose absence they mark, would flow unhindered through the solid lattice.

Experimenters soon undertook searches for a small "supersolid" phase in solid ^4He . But only in 2004 was there an apparently real sighting (see PHYSICS TODAY, April 2004, page 21).¹ Moses Chan and Eunseong Kim at the Penn-

sylvania State University found that the millisecond-scale resonant period of a solid ^4He sample oscillating in a torsion oscillator (TO; see figure 1) abruptly fell by about 18 ns as the sample was cooled below 200 mK.

Chan and Kim tentatively interpreted the period drop as a decrease in the oscillating sample's moment of inertia when about 1% of the solid ^4He enters a supersolid phase and thus decouples from the oscillation. The result provoked considerable interest, numerous other experiments worldwide that also seemed to see the transition, and some skepticism. The skepticism grew after John Beamish and James Day at the University of Alberta discovered in 2007 that the shear modulus of bulk solid ^4He increases substantially in precisely the temperature range where the supposed supersolid transition was being seen (see PHYSICS TODAY, February 2008, page 14).

That observed stiffening, an elastic effect with no suggestion of exotic quantum condensation, offered an alternative

explanation of the TO results: The period falls not because the sample's effective moment of inertia drops but rather because the effective torsion spring constant is increased by the abrupt stiffening of solid ^4He adhering to the oscillator.

At first glance, however, that prosaic alternative applies only to TO experiments with bulk solid ^4He samples. By contrast, the solid ^4He in the original Kim-Chan experiment¹ and in some of the later experiments was frozen within the nanopores of Vycor, a porous glass through which only the superfluid component of liquid helium can flow freely. Chan had chosen to look first for supersolidity in Vycor because he thought its tiny, irregular pores might maximize the abundance of lattice vacancies. For much the same reason, solid ^4He in Vycor should avoid the shear-modulus rise, whose mechanism involves the pinning of lattice dislocation lines much too long to fit in the pores.

But recently, careful analysis by Humphrey Maris at Brown University and numerical simulations by Chan's group have suggested that the stiffening of even a very small amount of bulk solid ^4He in the oscillator might account for all the anomalous period drops seen in Vycor experiments.

Now Chan and his postdoc Duk Kim report that they have repeated the 2004

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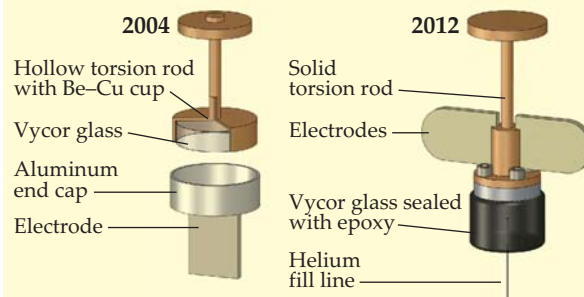
“discovery” experiment with a new TO design² that avoids essentially all bulk solid ⁴He. This time the anomalous period drop below 200 mK was gone, and so, perhaps, is supersolidity. As Beamish puts it, “Though there are still some unresolved issues, the new Chan paper seems nearly to close the circle.”

Oscillators old and new

In a TO, one determines a sample’s moment of inertia I by measuring the resonant oscillation period when the sample is attached to an elastic torsion rod. The resonant period is given by

$$P = 2\pi\sqrt{I/k},$$

where k is the rod’s torsion spring constant. The ultralow-dissipation oscillator systems used in the 2004 and 2012 Penn State Vycor experiments (shown in figure 1) could detect temperature-dependent changes in the oscillators’ millisecond-scale resonant periods with subnanosecond resolution. The high pressure needed to freeze the ⁴He



oscillation and measured temperature-dependent changes in the resonant period with subnanosecond precision. The crucial alteration in the 2012 experiment was the replacement of the metal sample-container assembly by a simple epoxy seal. That left no room for inadvertent solid ⁴He outside the Vycor whose abrupt stiffening might mimic the sought-after onset of supersolidity. (Adapted from ref. 2.)

and keep it frozen is maintained in both experiments by sealed transfer and containment systems.

In Chan’s 2004 oscillator, the Vycor disk, about 1.5 cm in diameter, was glued into a covering beryllium–copper cup, which was then fitted into the aluminum end cap. But a very thin space had to be left between the top of the Vycor and the Be–Cu cover to allow in-

fusion of liquid helium into the Vycor from the torsion rod’s hollow core, which served as a fill line.

Concerned that this space or other, inadvertent spaces in the assembly might have housed enough bulk solid ⁴He to stiffen the oscillator at 200 mK and thus raise its effective k , Chan and company chose what they call a “naked Vycor” design for the new experiment. They sim-

Figure 1. Torsion oscillators used in 2004 and 2012 experiments in Moses Chan’s laboratory.^{1,2} In both, helium-4 was frozen within the nanopores of a Vycor glass disk attached to a torsion rod. Electrode assemblies drove the rod to millisecond-period

Temperature-sensitive colloids show off an elusive melting mechanism

It’s been known for decades that colloids form ordered crystalline phases when packed together closely enough; when given more room to move, they behave like liquids. Colloidal systems thus offer a model for the microscopic mechanisms of phase transitions—processes that can’t be directly observed in normal crystals because molecules are too small and move too fast (see *PHYSICS TODAY*, December 1998, page 24). But because the parameter driving the phase transition is the colloid’s volume fraction, which is harder to homogeneously vary than temperature or pressure, some aspects of colloidal phase transitions have proved difficult to study.

In 2005 Arjun Yodh and colleagues at the University of Pennsylvania overcame that difficulty: They devised micron-sized polymer particles that reversibly shrink when heated, so the volume fraction can be tuned above and below the phase-transition threshold.¹ Yodh’s team used the particles to study heterogeneous melting, which begins at a grain boundary or other defect. Heterogeneous melting at a liquid–solid interface—the familiar case of an ice cube melting in a glass of water—had been studied and its mechanism understood: Particles near the surface begin to lose their crystalline order before the bulk material reaches its melting point. Yodh and colleagues were the first to observe the same mechanism at the interface between two crystalline domains.

Now Yilong Han and his colleagues at the Hong Kong University of Science and Technology have used the heat-sensitive particles to study the poorly understood process of homogeneous melting, in which a perfect crystal melts via the spontaneous nucleation of small liquid regions far from any preexisting defect or interface.² Because surface tension and strain between liquid and crystal compete with the bulk material’s free energy, a liquid nucleus is stable only when it’s larger than some critical size. Smaller nuclei just recrystallize—even when the bulk melting point is exceeded—so the crystal becomes superheated. Superheating and homogeneous melting are known to occur when a single crystal is heated internally with a laser or when surface melting is suppressed. But the nucleation mechanism has never before been observed

experimentally, and computer simulations have yielded inconsistent results.

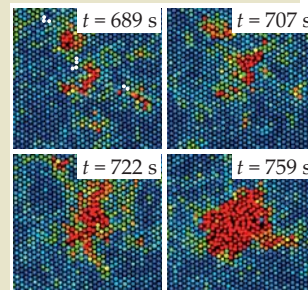
Han and company superheated a colloidal crystal—that is, they reduced the volume fraction below that which would normally trigger melting. Then they held it at constant temperature and watched it evolve. Snapshots from one of the 200 melting transitions they studied are shown in the figure. Blue, green, and orange dots represent solid-phase particles with progressively greater deviations from their lattice positions; liquid-phase regions, defined by a large deviation from hexagonal symmetry between a particle and its nearest neighbors, are shown in red. The researchers found that melting began not with the spontaneous formation of crystal defects, as some computer simulations had predicted, but rather with so-called loop rearrangements, first predicted by Xian-Ming Bai and Mo Li,³ in which particles swapped places while leaving the crystal structure intact.

What happened next depended on the degree of superheating. In weakly superheated crystals, liquid nuclei appeared and recrystallized until, by chance, one formed that was large enough to grow and melt the entire crystal. In more strongly superheated samples, two or more subcritical nuclei that formed near each other often coalesced into a nucleus of the critical size. At even more extreme superheating—when the volume fraction was reduced some 20% below the critical value for the melting transition—the crystalline phase was no longer metastable, and the entire crystal melted catastrophically.

Johanna Miller

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ply sealed the Vycor glass with epoxy. And the new torsion rod no longer has a hollow core to convey liquid helium. That function is replaced by a narrow fill line from below.

Figure 2 compares Chan's old and new results. The abrupt 18-ns period drop seen in the old experiment as the sample was cooled below 200 mK is gone. The only surviving steep feature, the period drop as the system is cooled to 1 K, is fully explained by the freezing of the initially liquid ^4He . The now very gradual period change below 1 K is accounted for by the stiffening of the Vycor itself. To facilitate comparison, period drops for all the curves in figure 2, with Vycor pores full or empty, use the period at 1 K as their zero reference.

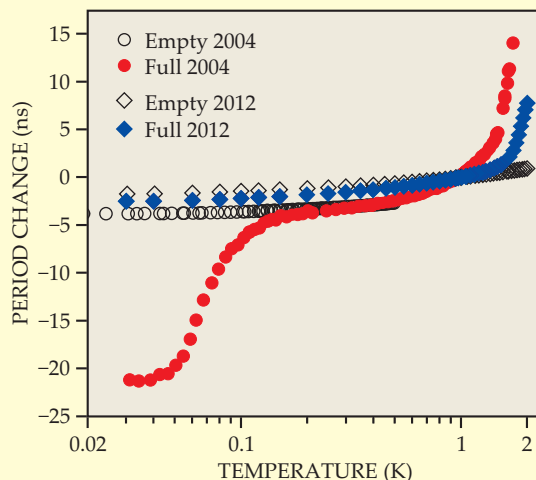
Based on their numerical simulations, Chan and company now attribute the period drop in the old data primarily to stiffening of the thin Be-Cu cover plate by an adhering solid ^4He layer about 50 μm thick. "In the new experiment, we see no remnant of the evidence for supersolidity," says Chan. "Playing detective has been interesting, but there are still loose ends to be resolved."

Loose ends

In 2010 a team at the Korea Advanced Institute of Science and Technology led by Chan's 2004 coauthor Eunseong Kim carried out a different sort of TO experiment, designed to distinguish evidence of supersolidity in bulk solid ^4He from mere elastic stiffening. Velocity fields in supersolids, just as in superfluids, should be perfectly irrotational (curl free). Therefore, a steady "DC" rotation of the entire oscillator apparatus—at a few radians per second—should measurably affect a real transition to supersolidity but not simple material properties like a shear modulus.

And indeed, Kim's team found a dependence of the oscillation-period drop on the rotation rate—and a lack of any

Figure 2. Changing resonant period of the torsion oscillators in figure 1 as the helium-4 in the porous Vycor glass disk was cooled below 2 K. The initial period drops above 1 K are explained by solidification of the ^4He . The second drop, below 200 mK in the 2004 data, was attributed to the onset of a supersolid phase. But the 2012 repetition, with modifications designed to eliminate inadvertent bulk solid ^4He outside the glass, exhibits no such drop. Also shown are resonant-period curves with the Vycor glass empty. For comparison, all the curves are set to zero at 1 K. (Adapted from ref. 2.)



such dependence in the shear modulus—that hints at transition to a supersolid phase.³ New TO experiments with DC rotation are currently in the works.

Beyond the experimental confines of torsion oscillators, heat-capacity measurements carried out by the Penn State group raise issues still unresolved. In 2009 Chan and company found that the specific heat of bulk solid ^4He peaks near 100 mK in a way that suggests a phase transition.⁴ "We're still investigating that," says Chan. "If it's not supersolidity, we'd like to know what it is."

Robert Hallock and coworkers at the University of Massachusetts Amherst have undertaken experiments of a different kind in search of atom transport through bulk solid ^4He . They attach reservoirs of superfluid ^4He to opposite ends of a long block of solid ^4He . Imposing a temperature difference between the reservoirs to create a chemical-potential difference, they look for evidence of resulting mass flow through the block.

The small mass flux observed in those experiments suggests that a network of edge dislocations in the ^4He lattice provides one-dimensional super-

fluid cores for the transport of atoms through the solid.⁵ "The relationship between the mass flux we seem to see and the torsion-oscillator results is unclear," says Hallock. "Its dependence on the imposed chemical potential is quite different from what one would expect from the quantum condensation of lattice vacancies proposed in the 1970s."

"Hallock's mass flux is much too small to produce an observable period drop in the torsion-oscillator experiments," says Chan. "And it happens at higher temperatures—around 600 mK. Maybe they're seeing the real supersolidity transition that our experiments are not sensitive enough to notice."

Bertram Schwarzschild

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physics update

These items, with supplementary material, first appeared at <http://www.physicstoday.org>.

Ultracold chemistry in supersonic beams. When two atoms or molecules interact at very low collision energies and therefore very cold effective temperatures, the reactants' de Broglie wavelengths become comparable to their separation. In that regime, theory says the classical notion of bouncing billiard balls breaks down; instead, one reactant can tunnel quantum mechanically—through an angular-momentum or energy barrier, for example—to chemically react with the other. But the experimental realization of such quantum effects lags far behind the theory, largely because of the difficulty of cooling neutral atoms and molecules. So quantum chemists make fast molecular beams of different species by letting high-pressure gas adiabati-

cally expand into vacuum through a small orifice. Researchers have set up reactive encounters by intersecting two such internally cold molecular beams. In that way, temperatures as low as 5 K have been reached, with hints of quantum behavior. Now a group of chemical physicists at the Weizmann Institute of Science in Israel have broken through to 10 mK and have seen clear quantum resonances. The trick? A beam of one species is magnetically curved to merge with a beam of the other species; in a comoving frame, the two reactants are practically motionless. The team looked at two systems: excited metastable helium in one beam and either molecular hydrogen or argon in the other. At ultralow energies, they saw well-resolved quantum-enhancement peaks in the reaction rates plotted against collision energy, for a process called Penning ionization. According to the group, the method should be applicable to many chemical reactions. (A. B. Henson et al., *Science* **338**, 234, 2012.) —SGB