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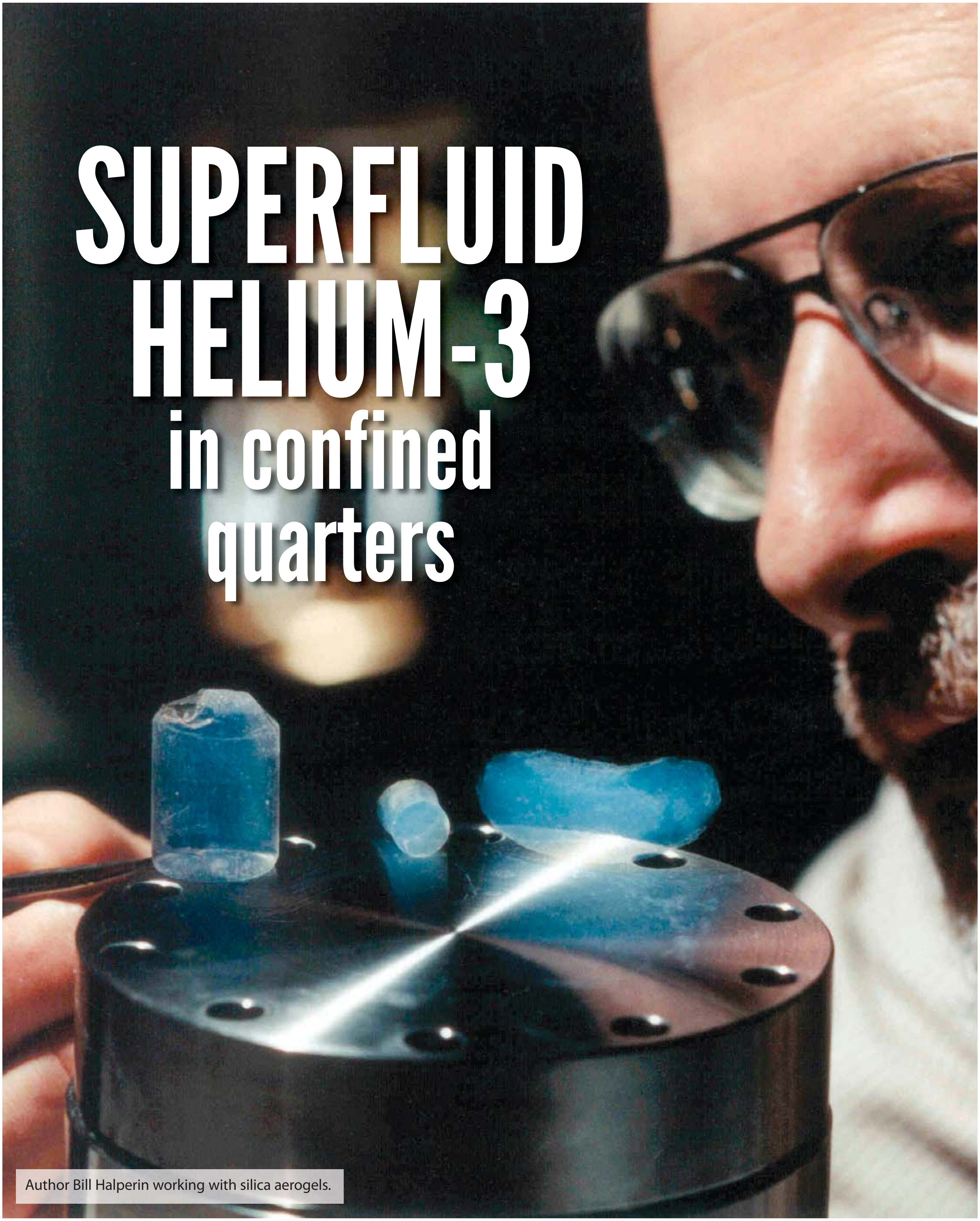

Author Bill Halperin working with silica aerogels.

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Disorder, confinement, and symmetry breaking contribute to the formation of new phases when liquid helium-3 is infused in a highly porous random solid such as a silica aerogel.

Liquid helium-3 and helium-4 are remarkable substances. They are quantum liquids, meaning that their behavior is governed by the laws of quantum mechanics. Because of their small atomic mass, each isotope exists in a liquid state down to the temperature of absolute zero. And at sufficiently low temperature, each becomes a superfluid. However, the two isotopes have very different properties because ^{3}He is a fermion and ^{4}He is a boson. As a result of their different statistics, superfluidity in ^{3}He appears at a temperature one-thousandth of that at which superfluid ^{4}He forms. A second difference is that ^{3}He has multiple thermodynamic phases.

The discovery of superfluid ^{3}He was recognized with two Nobel Prizes in Physics, one for experimental work and one for theory. The 1996 prize was awarded to David Lee, Douglas Osheroff, and Robert Richardson for their pioneering ^{3}He NMR measurements, carried out in 1971 at Cornell University.[1] (See PHYSICS TODAY, December 1996, page 17.) As the Cornell experiments were being conducted, Anthony Leggett, then at the University of Sussex, showed that superfluid ^{3}He is the realization of the Bardeen-Cooper-Schrieffer (BCS) pairing theory of superconductivity in a quantum liquid; he thus made an important connection that is still relevant to research on superconductivity.[2] Leggett's theory also accounted for the two observed phases of pure superfluid ^{3}He, and for it, he received a share of the 2003 Nobel Prize in Physics. (See PHYSICS TODAY, December 2003, page 21.)

In this article we highlight the discovery of new superfluid phases in ^{3}He infused into amorphous, extremely light, high-porosity solids known as aerogels. Before 1995 it was not clear that the superfluid could actually survive in such a messy environment. (See the article by Moses Chan, Norbert Mulders, and John Reppy, PHYSICS TODAY, August 1996, page 30.) And if it could survive, what would be the properties of the disordered superfluid?

Research by the three of us addressed those questions on several fronts. Parpia used a torsional oscillator (described below) to look for the superfluid mass fraction,[3] and Halperin performed NMR measurements[4] that were key to the identification of the new phase of ^{3}He as a superfluid of bound pairs with a nuclear spin of $\hbar$. Sauls and colleagues immediately began to develop a theory of superfluid ^{3}He confined in random media.[5] We found that not only does the superfluid exist but also its phase transitions are well defined in temperature, pressure, and magnetic field. More recently, various researchers have demonstrated that by engineering anisotropy in the mean free path of the confined superfluid, they can control which superfluid phases are stable and can even force the appearance of new phases. But before discussing those phases, we begin with a brief description of the properties of unconfined superfluid ^{3}He.

Pure helium-3

For a wide range of temperature below the Fermi temperature of about 1 K, ^{3}He is well described by Lev Landau's Fermi-liquid theory, which is formulated in terms of low-energy, fermionic excitations called quasiparticles. The thermodynamic and transport properties of the normal (not superfluid) Fermi liquid are determined by the quasiparticles and their interactions, which can lead to composite bosonic excitations. One such composite is a so-called paramagnon, a longed-lived spin excitation that is responsible for the binding of two spin-½ quasiparticles into a Cooper pair and thus for the formation of a superfluid.[6]

The experimental and theoretical developments that followed the initial discovery of superfluid ^{3}He have influenced many aspects of condensed-matter physics.[7] In particular, they have been central to developments in the field of unconventional superconductivity, which describes materials in which symmetries of the normal metallic state are broken in the superconducting phase. For ^{3}He, spin- and orbital-rotation symmetries, mirror symmetry, and time-reversal symmetry are all broken, as is the so-called $U(1)$ gauge symmetry, the characteristic broken symmetry of all superconductors and superfluids. Many ground states are possible in which states with orbital angular momentum equal to $\hbar$ (called p-wave) are spin-triplets with $\hbar$ spin angular momentum. Figure 1 shows the phase diagram for two of the p-wave, spin-triplet states—the A and B phases that exist in pure ^{3}He absent a magnetic field. Figures 2a and 2b show the energy gaps that separate the Fermi surface from the next-highest energy state in those two phases.

The A phase is a superposition with equal amplitudes for the oppositely spin-polarized triplet states $|\uparrow\uparrow\rangle$ and $|\downarrow\downarrow\rangle$, a combination called equal-spin pairing (ESP). The orbital wavefunction of the A phase breaks both time-reversal and mirror symmetries; for that reason, the A phase is chiral. The B phase is a superposition of all three triplet spin states: $|\uparrow\uparrow\rangle$, $|\downarrow\downarrow\rangle$, and $|\uparrow\downarrow + \downarrow\uparrow\rangle$. The spin states and orbital states are organized so that the B phase has zero total angular momentum. It is an isotropic state with an energy gap that is isotropic in momentum space. For most of the superfluid part of the zero-field phase diagram, the B phase is the stable one. The stability of the A phase at high pressure is a consequence of strong quasiparticle coupling to long-lived spin fluctuations.

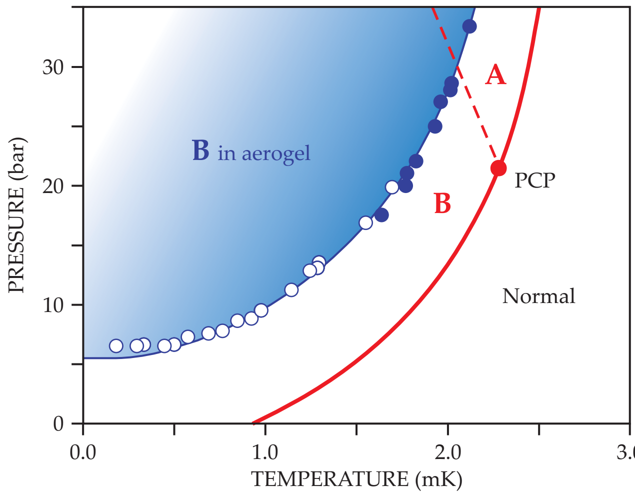

FIGURE 1. LIFE IS DIFFERENT IN AN AEROGEL. At zero magnetic field, pure helium-3 undergoes a transition from the normal state to the superfluid state along the red line. The superfluid state of the pure material can exist in either of two phases, called A and B. Normal ^{3}He and the A and B phases can all coexist at the marked polycritical point (PCP). For ^{3}He confined in a 98% porous silica aerogel, an amorphous, extremely light material, the phase transition occurs at lower temperature and higher pressure, and only the B phase (blue region) exists. Open circles are from superfluid-fraction measurements;[3] closed circles represent acoustic-impedance measurements.[4] The blue curve is a theoretical result that proceeds from the observation that the ^{3}He mean free path in an aerogel is much longer than the size of a superfluid Cooper pair.[5]

Because the B phase incorporates $|\uparrow\downarrow + \downarrow\uparrow\rangle$ pairs, its magnetic susceptibility is less than that of the A phase. The transition from A to B is first order and accompanied by a susceptibility jump. For B-phase ^{3}He in a magnetic field, the Zeeman energy can be comparable to the binding energy of a $|\uparrow\downarrow + \downarrow\uparrow\rangle$ pair, and the B phase is suppressed in favor of the A phase. For large magnetic fields, a third phase of the pure superfluid can exist. Called A_1, it is a chiral state like the A phase, but instead of including both the $|\uparrow\uparrow\rangle$ and $|\downarrow\downarrow\rangle$ states, it includes only the one that remains stable at high field. In 2015 Vladimir Dmitriev and colleagues reported a fourth p-wave phase.[8] That is the P (for polar) phase, also an ESP state like the A phase. The P phase has not been observed in pure ^{3}He. Rather, it was stabilized in a highly anisotropic alumina aerogel whose Al_2O_3 strands were nematically aligned—that is, oriented predominantly in parallel—and separated on average by about 50 nm. Liquid ^{3}He confined in that structure forms a phase with a single p orbital along the strands. The theoretical structure of the energy gap for the P phase in pure ^{3}He is shown in figure 2c.

Silica aerogel

Dry silica aerogels are low-density solids whose porosities may be as high as 99.5%. In aerogels used for superfluid ^{3}He inves-

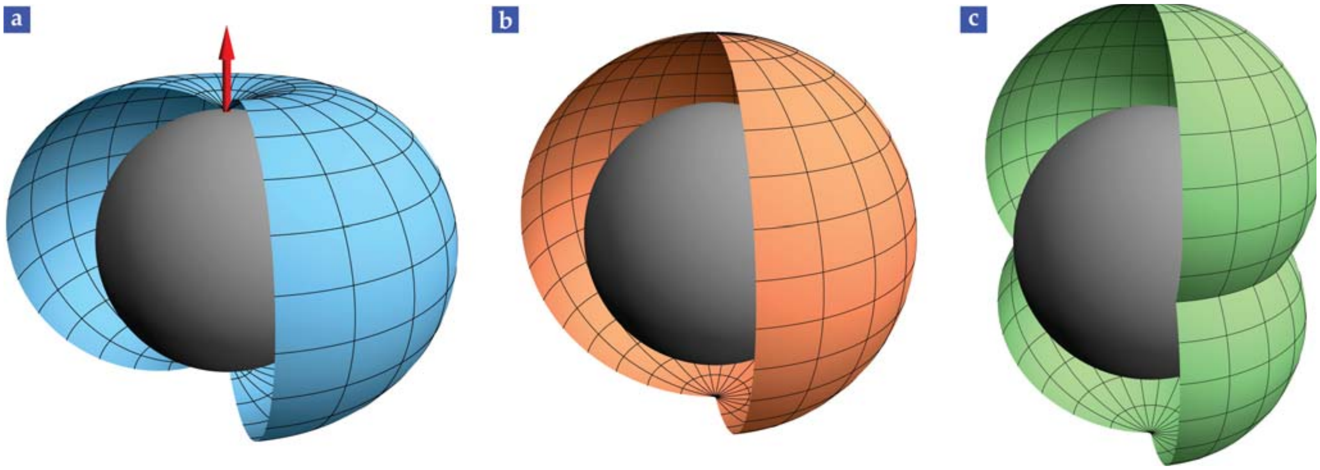

FIGURE 2. DIFFERENT PHASES HAVE DIFFERENT SYMMETRIES. In these sketches corresponding to different phases of pure superfluid helium-3, the gray spheres represent the Fermi surface in momentum space. Between each Fermi surface and the enveloping colored surface is an energy gap, in which no allowed momentum states exist. **(a)** The wavefunction of the chiral A phase breaks time-reversal and mirror symmetries and has angular momentum in the direction given by the red arrow. **(b)** The B phase is isotropic. **(c)** The newly discovered P (for polar) phase has at present only been observed in an anisotropic aerogel.

tigations, the porosity value is typically 98%. As shown in figure 3a, the aerogel structure consists of a dilute network of thin silicon dioxide strands whose diameter is typically 3–5 nm. Aerogels have found a wide range of applications that include Cherenkov counters used in particle physics; lightweight, transparent thermal insulation for space vehicles; and heterogeneous catalysis.

A silica aerogel can be fabricated with a base-catalyzed synthesis that produces nanometer-sized SiO_2 particles from tetramethylorthosilicate dissolved in alcohol. In a process called diffusion-limited cluster aggregation (DLCA), the particles diffuse in the fluid and aggregate to generate strands and clusters that bond to form a gel. That wet gel is dried supercritically in a high-pressure autoclave. Supercritical drying means that the wet gel never crosses the liquid–gas phase line; the technique ensures that the microstructure doesn't collapse from capillary forces at the liquid–gas interface.

The resulting material is hydrophobic and stable in air. A 98% porosity aerogel is fractal on length scales less than about 50 nm; on much longer length scales, it can be prepared to be very homogeneous and isotropic, as verified by small-angle x-ray scattering measurements and optical birefringence. According to our simulations, the strand-like structures of the gel have a spatial correlation length ξ of 30–50 nm; you can think of ξ as a measure of the largest voids, as shown in figure 3b.

A length scale even larger than ξ is associated with the open structure of an aerogel: the straight-line ballistic mean free path that terminates at aerogel surfaces. A DLCA simulation for 98% porous structure gives a mean-free-path length λ of about 180 nm, a value that agrees well with the measured mean free path for ^{3}He quasiparticles in the normal Fermi liquid. That agreement provided impetus for Sauls and colleagues to use scattering theory[5] with model parameters ξ and λ to describe the effects of an aerogel on the superfluid phases of ^{3}He.

Although aerogels can be homogeneous and isotropic on large scales, they need not be. As mentioned above, the P phase

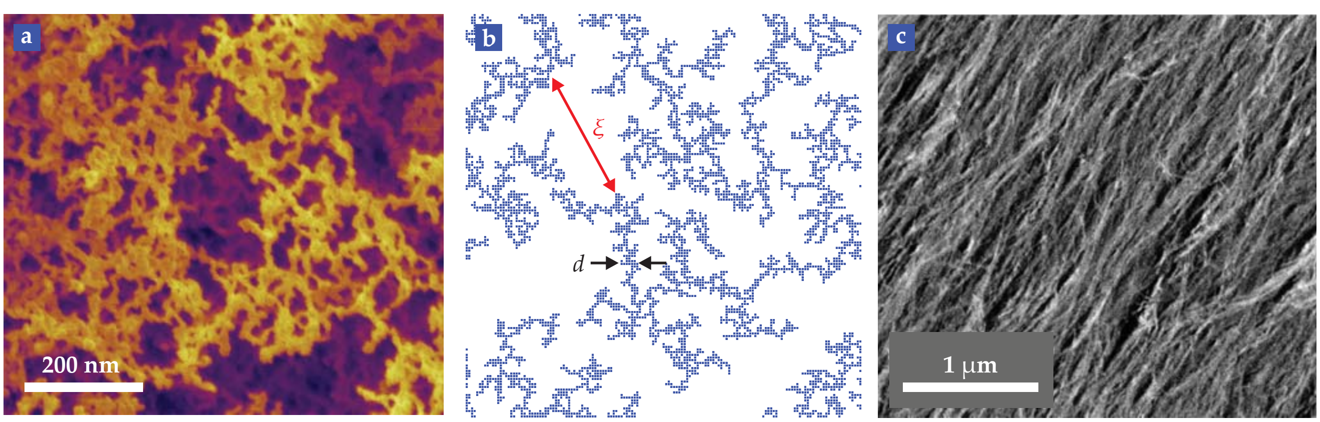

FIGURE 3. AEROGELS CAN BE ISOTROPIC OR NOT. (a) Typical superfluid helium-3 investigations use a 98% porous silica aerogel such as seen in this scanning electron microscope image. **(b)** A two-dimensional view of a 3D simulation of the aerogel illustrates the strand size *d* and the correlation length *ξ*, essentially the size of the largest voids. **(c)** The polar phase of superfluid was discovered in a nematically ordered aerogel formed from alumina strands such as shown in this scanning electron microscope image.

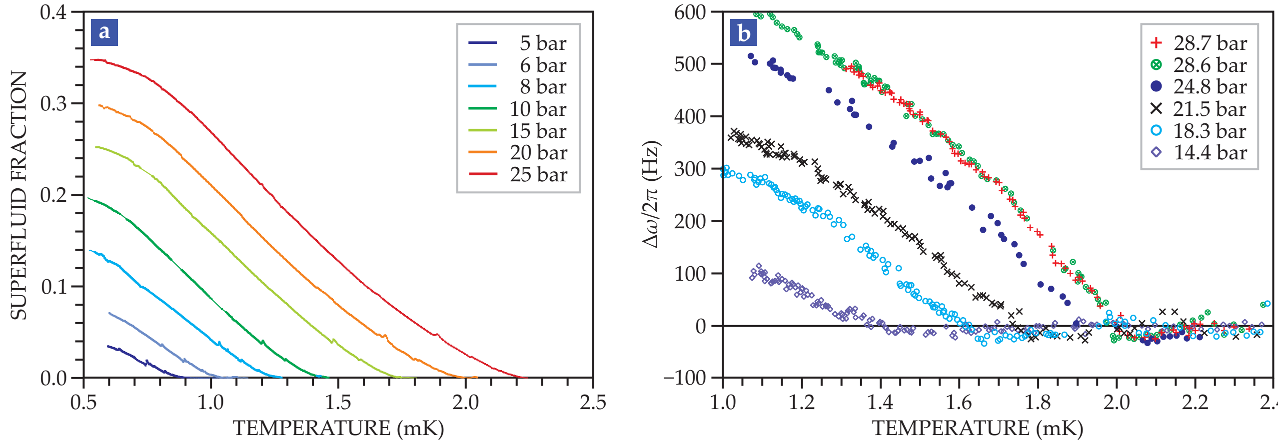

FIGURE 4. SUPERFLUIDS IN AEROGELS make themselves known. **(a)** The superfluid fraction is a function of temperature and pressure, as determined by torsional-oscillator experiments.[3] **(b)** The NMR frequency shift $\Delta\omega$ away from the value for a normal fluid is also a function of temperature and pressure.[4] Note the sharply defined onset of the superfluidity in the measurements, both of which were conducted with a 98% porous silica aerogel.

of superfluid ^{3}He was discovered in an anisotropic, nematically ordered aerogel; figure 3c shows an example.

Disorder in superfluid helium-3

Impurities and structural defects are unavoidable in almost all forms of condensed matter. For superconducting materials, including superfluids, relatively modest concentrations of chemical impurities or structural imperfections can mask intrinsic behavior or even destroy an unconventional superconducting state that has broken rotation or reflection symmetry.

In contrast, liquid ^{3}He at low temperature is the purest known form of matter. Even ^{4}He will not dissolve measurably in ^{3}He if the temperature is below 10 mK. The intrinsic properties of the normal and superfluid phases are rich, well studied, and quantitatively understood in terms of BCS pairing in a material free of defects and impurities.

Superfluid ^{3}He, presumed always to be pristine, had widely been considered unique among superconductors. That changed with the discovery of superfluidity of liquid ^{3}He in a silica aerogel, which opened wide-ranging opportunities to study the effects of disorder on an unconventional BCS condensate whose intrinsic properties were quantitatively understood.

Superfluid ^{3}He in a silica aerogel was discovered with the use of a torsional oscillator—a torsion rod attached perpendicular to a disk containing the ^{3}He–aerogel sample. In the so-called two-fluid model, the superfluid state of ^{3}He is a superposition of normal-fluid excitations that are viscously clamped to the porous structure and an inviscid, decoupled superfluid of Cooper pairs that does not contribute to the moment of inertia of the mechanical oscillator. At the onset of superfluidity, the moment of inertia and the resonant period of the oscillator decrease in a way that is directly related to the superfluid fraction. However, in contrast to pure superfluid ^{3}He, the superfluid fraction of the confined liquid is much less than unity as the temperature approaches absolute zero (see figure 4a).

As the superfluid fraction was being measured at Cornell, researchers at Northwestern University were obtaining a frequency shift $\Delta\omega$ of the superfluid's NMR spectrum from that of the normal fluid; figure 4b shows their result. For pure ^{3}He, $\Delta\omega$ was theoretically understood to be proportional to the square of the amplitude of the bound-pair wavefunction. For each specific p-wave state, according to theory, the shift depends in a characteristic way on the amplitude of the NMR excitation.[2] Thus a measurement of shift versus RF amplitude provides a powerful means for state identification.

The onset of the NMR frequency shift is at the critical temperature T_c of the superfluid transition. Proportional to the square of the amplitude of the pair wavefunction, $\Delta\omega$ grows

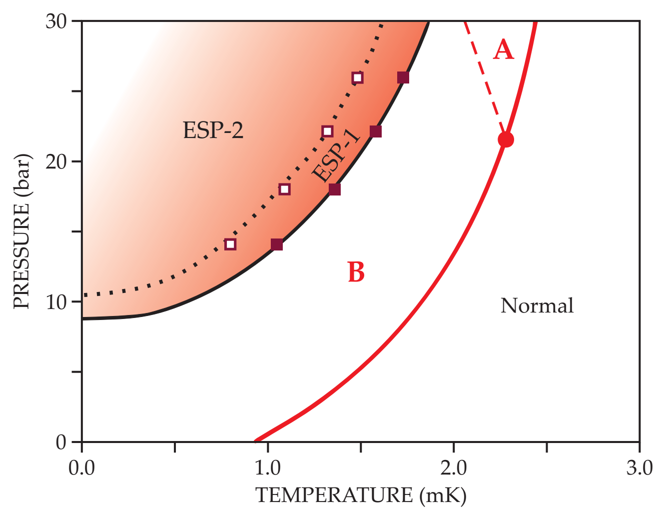

FIGURE 5. NEW PHASES ARISE in stretched aerogels. The phase-diagram data[10] (filled and open squares) are for superfluid helium-3 in a silica aerogel with a strain of 0.14, subject to an external magnetic field of 31.1 mT. The transition at relatively high temperature is from the normal fluid to an equal-spin pairing state labeled ESP-1. Like the A phase of pure ^{3}He, the ESP-1 phase is chiral. The solid black curve through the data is a fit based on scattering theory. At lower temperature, the ESP-1 phase transitions to a lower-symmetry chiral phase labeled ESP-2. The dotted curve is to guide the eye. For reference, we have reproduced the pure-superfluid phase-transition lines from figure 1.

with temperature below that second-order transition. Like the superfluid fraction, the frequency shift is smaller in an aerogel than in the pure superfluid. The reduction of both is due to ^{3}He quasiparticle scattering from the aerogel structure. The scattering not only breaks Cooper pairs, it also destroys the energy gap of the pure superfluid, even in the B phase.

Gapless superfluids in an isotropic aerogel

Pure ^{3}He in the B phase is a fully gapped BCS superfluid, "fully" meaning that the gap exists for momenta in all directions. When the temperature is much less than the gap energy divided by Boltzmann's constant—about 2 mK—essentially no normal fluid exists. The heat capacity, thermal conductivity, and normal fluid fraction are exponentially suppressed with temperature, as would be expected for an ideal BCS superconductor. However, the superfluid B phase in an isotropic aerogel is different: As the temperature approaches zero, the thermal conductivity and heat capacity go to zero linearly.

Those results are consistent with the theoretical prediction that superfluid ^{3}He in a high-porosity aerogel is "gapless"—that is, the superfluid has a nonzero density of states at the Fermi energy—over the entire pressure range such that a normal-fluid component coexists with the superfluid condensate at arbitrarily low temperature.[9] In fact, the calculated density of states as a function of energy shows only slight distinction among the various superfluid phases.

Acoustic-impedance measurements reveal that when no magnetic field is present, the equilibrium superfluid phase for isotropic aerogels is the gapless B phase, as shown in figure 1. The absence of the A phase in isotropic aerogel is a consequence of suppression of the strong-coupling effect, evident in pure ^{3}He, due to pair breaking by quasiparticle scattering. The observed stability of the B phase confirms the important prediction that isotropic quasiparticle scattering favors a more isotropic phase.[5] However, as with the pure superfluid, the A phase is more stable than the B phase in a magnetic field because the B phase has higher Zeeman energy. When the magnetic field is small, the transition temperature from the B phase to the A phase depends on the square of the magnetic field.

New phases in anisotropic aerogels

Perhaps the most remarkable effects of an aerogel on superfluid ^{3}He result from controlling quasiparticle scattering with engineered anisotropic aerogels. Uniaxial stretching or compression of the silica aerogel framework introduces in the quasiparticle mean free path a globally uniform anisotropy that can be conveniently quantified via optical birefringence.

Stretching is achieved by using extra catalyst during aerogel synthesis; the result is a uniform radial shrinkage of the aerogel away from the walls of a glass tube during the drying process.[10] Figure 5 shows the pressure–temperature phase diagram for superfluid ^{3}He in a stretched aerogel with a strain of 0.14. Equal-spin pairing phases, stabilized by anisotropic quasiparticle scattering, replace the B phase throughout the entire diagram.[5,11] NMR measurements with magnetic fields oriented parallel and perpendicular to the strain show that the ESP-1 phase is a chiral state with angular momentum aligned along the strain axis. At a temperature below the normal-to-ESP-1 transition is a second transition to a new ESP state, labeled ESP-2 in the figure. Theory predicts that phase to be a polar-distorted, biaxial chiral phase whose orbital structure is distorted away from the strain axis.[11] NMR measurements demonstrate that its angular momentum is perpendicular to the strain axis.

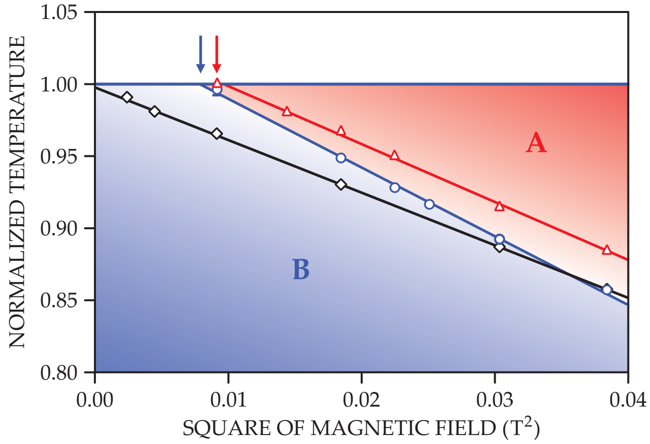

FIGURE 6. THE B PHASE MAKES A COMEBACK in a compressed silica aerogel. The phase-diagram data[12] shown here (open shapes), taken at a pressure of 26.3 bar, are for superfluid helium-3 in an isotropic silica aerogel (black) and in an anisotropic aerogel (blue and red) with a strain of −0.20. For the blue circles, the strain is parallel to the external magnetic field; for the red triangles, it is perpendicular. The temperature is normalized by the normal-to-superfluid transition temperature. The straight lines through the data are theoretical fits. At the transition temperature, the B phase in the isotropic aerogel is stable at zero applied field and unstable in any nonzero field. Compression, however, stabilizes the B phase at the transition temperature for fields up to a critical value of approximately 0.1 T. Arrows indicate polycritical points at which normal, A, and B phases can coexist in a nonzero magnetic field.

Anisotropy in an aerogel can also be created by compressing isotropic samples in the glass tube after drying. The unexpected result is that the B phase becomes more stable and preempts ESP A phases. As figure 6 shows, at a sufficiently high magnetic field the A phase reappears; thus there exists a new polycritical point describing conditions where the normal, A, and B phases can coexist in a nonzero magnetic field. The square of the field H_c at that point is proportional to the strain in the aerogel; a representative value is $H_c = 0.1$ T, which was obtained for a strain of −0.20 (negative for compression) and a pressure of 26.3 bar.[12] Further work has shown that the B phase in the compressed aerogel is no longer isotropic. The significant polar distortion of its orbital structure in the direction of the strain is a subject of ongoing investigation.

Topology and the polar phase

Evidence for the existence of a polar phase of superfluid ^{3}He came independently from experiments using NMR and torsional-oscillator techniques.[8,13] The investigations were conducted with nematically oriented alumina aerogels—sometimes called nafen or obninsk—manifesting extreme anisotropy: The ratio of the ballistic mean free path of quasiparticles moving along the nematic axis can be eight times that of the mean free path for perpendicular motion. The P phase has a single p-wave orbital state, illustrated in figure 2c for pure ^{3}He; the state's polar axis is aligned along the nematic axis of the aerogel.

The observation of the polar state is of great significance. It does not occur in pure superfluid ^{3}He, but theoretical work suggested it would be stable in anisotropic aerogels[14] and channels.[15] Moreover, the polar state was recognized to be a favorable candidate to support the elusive half-quantum vortex that Grigory Volovik and Vladimir Mineev predicted for *p*-wave, spin-triplet superfluids more than 40 years ago.[16] Indeed, the half-quantum vortex was experimentally detected[17] shortly after discovery of the P phase in ^{3}He confined in rotating nafen and cooled to as low as 300 μK.

But what is a half-quantum vortex? It is a topological defect that combines mass and spin vortices. For conventional superconductors, the change in the phase of the wavefunction around a closed path enclosing a quantized vortex is 2π; for the half-quantum vortex in superfluid ^{3}He, half of that phase change is associated with the circulation of superfluid mass current and half is associated with the circulation of spin current.[16]

The discoveries of new phases of ^{3}He confined in high-porosity disordered solids have demonstrated the effects of disorder on superfluids with complex symmetry breaking. Together with theoretical developments in topological condensed matter, the discoveries have opened up new research directions in the study of topological phases of ^{3}He confined on the nanoscale. Those include high-quality-factor nanofluidic mechanical oscillators, nanoscale NMR cells, ultrasonic cavities for quantum transport, and the dynamics in quantum fluids. Such investigations are helping to advance the frontier of hybrid quantum systems, a field that marries quantum systems with other nanoscale materials and nanofabrication techniques. To this day, one of the simplest of those quantum systems, superfluid ^{3}He, continues to astound.

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