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Superfluid liquid hydrogen?

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Recent work by Peter Toennies' group in Göttingen has revealed convincing evidence for superfluidity in liquid hydrogen – thus adding a third liquid to the two, liquid ^{4}He and liquid ^{3}He, in which superfluididy is already known.

Superfluids are numbered among the most peculiar and counter-intuitive of all materials. They have no viscosity at all. Thus, an object travelling in a pure superfluid moves frictionlessly. Its behaviour is inertial, much as it would be in a vacuum (apart from an increased effective mass). Similarly, superfluids can flow effortlessly through narrow channels or pores that are virtually impermeable to conventional liquids.

Superfluids are relatively rare, and inaccessible. The first liquid in which superfluidity was recognised was ^{4}He, in 1938. It was found to undergo a phase transition to the superfluid state below the so-called lambda temperature $T_\lambda = 2.12$K. Fritz London suggested at the time that the phenomenon was associated with Bose-Einstein condensation (BEC), which is expected to occur in any assembly of freely mobile bosons (particles with zero or integer spin) if cooled below a critical temperature T_B. In BEC, a macroscopic fraction of all the particles in the assembly congregate together in the zero-momentum ground state. In 1972 superfluidity was also discovered in the rare isotope, liquid ^{3}He, below a critical temperature about a thousand times smaller, at $T_c = 2.4$mK. The mechanism here is somewhat different because, with half-integral spin (being composed of an odd number of fundamental particles), the ^{3}He atom is a fermion, not a boson. But, at T_c, the atoms can link up to form Cooper pairs. Because each pair is a boson (being composed of an even number of fundamental particles), the system can then undergo BEC like its liquid ^{4}He cousin.

Where else may superfluididy exist? The most long-standing example is the electron gas in a superconductor – the original instance of a Cooper pair system (electrons being fermions). More recently, superfluidity has been observed in laser-cooled alkali atomic gases. It is also believed that the neutron and proton fluids in neutron stars are superfluid but the idea is, to say the least, rather difficult to test experimentally.

Although it is generally agreed that there is a close association between BEC and superfluidity, their exact relationship has yet to be established – the ground state condensate of an ideal Bose-condensed system would clearly not be a superfluid, because there is no reason why condensate atoms should not undergo collisions with walls or with a moving object, leading to the usual frictional effects. The nonidealities – the inter-particle forces – must in some way also be important. So there has been much effort over the years to find new laboratory superfluids for experimental investigation. A major difficulty is that, with the exception of helium, other materials have solidified long before they have been cooled down to their calculated T_B.

Even the best candidate, hydrogen, has its triple point at 13.8K, whereas its T_B is calculated to be much lower, at about 6K (for para-H_2, with its nuclear spins antiparallel) where the material would normally be solid. A possible approach explored by Humphrey Maris (Brown University) has been to try to supercool the liquid below its normal solidification point – but this has not succeeded. The alternative approach pursued by Slava Grebenev and colleagues has been radically different: it is based on the laser-droplet beam depletion apparatus developed by the Toennies group.

The structure of the droplets for the hydrogen experiment is as indicated schematically in Fig 1. A tiny quantity – between 14 and 16 molecules – of para hydrogen (pH_2) is held within a microscopic helium droplet containing a large impurity molecule (linear carbonyl sulphide, or OCS). In the stable state the OCS molecule is at the centre, surrounded by a thin layer of hydrogen, followed by a relatively thick shell of liquid ^{4}He as shown in Fig. 1(a). It was also possible to add an outer shell of liquid ^{3}He as shown in (b). In the vacuum of the beam tube, the droplets cooled almost instantly by evaporation to 0.38K for droplets with a ^{4}He outer shell, or to 0.15K for the ^{3}He coated droplets. In both cases, the ^{4}He shell would have been a pure superfluid, with no viscous "grip" on the rest of the droplet. The state of the hydrogen was unknown.

While travelling in a beam from the source region to a detector (quadrupole mass spectrometer), the droplets were exposed to infra-red photons from a laser. When the laser was tuned to a wavelength such that the droplet could absorb photons, the detector signal decreased markedly. A variety of different modes of the OCS molecule can in principle be excited. Of particular interest here are rotational states of the OCS/pH_2 complex. If the complex rotates as a whole, it has a relatively large moment of inertia. If however the pH_2 layer becomes superfluid, it can be expected largely to decouple from the OCS, so that its net moment of inertia falls almost to zero in the case of axial rotations of the OCS.

Examples of the spectra reported by the Göttingen group are shown in Fig 2. The upper part is for a 0.38K droplet (without ^{3}He), and the lower part is for a 0.15K droplet (further cooled by ^{3}He evaporation). In each case, the spectra repeat, depending on the number of pH_2 molecules present, but their shapes at these two temperatures are totally different. The sharp peaks at 0.38K are related to rotation of the complex, and their absence at 0.15K strongly suggests that rotation is no longer being excited. The most likely explanation is that the moment of inertia of the OCS/pH_2 complex has fallen to a very small value at 0.15K because the onset of superfluidity in the hydrogen at some temperature intermediate between 0.38K and 0.15K has caused it to decouple from the OCS. When the experiment was repeated with deuterium in place of the hydrogen, the sharp peaks still persisted at 0.15K, indicating an absence of superfluidity in deuterium at this temperature. The authors have carried out several checks and simulations, and all of them seem to confirm the central conclusion of superfluidity in para-H_2 at 0.15K.

How does this result relate to more conventional types of experiment? It should be emphasized that superfluidity in a shell containing about 15 hydrogen molecules does not necessarily mean that the phenomenon will ever be observed in bulk liquid. Probably, it will not, because of the difficulty of maintaining the liquid state at low enough temperatures. But the results for droplets seem to have given us a new superfluid, presumably quite different from the liquid heliums with which low temperature physicists are so familiar. It will be an interesting challenge to find ways of studying it in order to establish its properties.

Figures

1. Sketch indicating schematically the structure of droplets used in the quest for superfluidity in hydrogen. The core is an OCS molecule surrounded by 14–16 molecules of para-hydrogen (pH_2). (a) With an outer coating of $\sim$8000 ^{4}He atoms, the droplet reaches a temperature of 0.38K. (b) With $\sim$500 ^{4}He atoms, and $\sim$10000 ^{3}He atoms as an outer coating, the increased evaporative cooling takes the droplet down to 0.15K.

   [Sketch as below]

2. Infra-red absorption spectra for $OCS(pH_2)_n$ complex in ^{4}He droplets at 0.38K (top) and in ^{3}He/^{4}He droplets at 0.15K (bottom), showing contributions from droplets with $n = 14, 15$ and 16 molecules of para-hydrogen. Note that the sharp spectral peaks seen at 0.38K are absent at 0.15K.

   [Right hand column (parts C and D) from Fig. 2 of Grebenev et al, *Science* **289**, 1532 (2000).]