Superconductivity and Spin Fluctuations

by David Pines

Electrons in normal metals are normal Fermi liquid in that, no matter how strong their interaction, the particles retain the essential properties of a non-interacting fermion system: the ground-state distribution of particles can be characterized by a Fermi surface in momentum space (which is spherical if one neglects anisotropy introduced by the lattice); excited states can be placed in one-to-one correspondence with those of a free-electron gas: the specific heat varies linearly with the temperature; and so on. In superconductors, on the other hand, as first shown by Bardeen, Cooper, and Schrieffer in their microscopic theory developed in 1957, both the ground state and the excited states of the system are altered in a fundamental way. A net attractive interaction between pairs of particles near the Fermi surface gives rise to an instability of the normal state, and the superconducting ground state becomes a single quantum state, the condensate, which is a coherent superposition of bound particle pairs and which can flow without resistance. To produce single-particle excitations from the superconducting ground state requires a finite amount of energy, the energy gap, so that the specific heat of a superconductor is drastically altered from that of a normal metal.

In BCS theory the net attractive interaction between conduction electrons near the Fermi surface arises from the exchange of phonons, the quanta of crystal lattice vibrations. The coherent pairs that make up the condensate are in $^3S$ states (that is, states with zero total spin and angular momentum), corresponding to pairs of particles with opposite spins and momenta. Other pairings, such as p-state pairing (in which the condensate would be a coherent superposition of pairs of particles with parallel spins) or d-state pairing, are in principle possible; however, both experiment and microscopic calculations to date suggest that where electron-group on various magnetic and nearly magnetic systems, Among the papers was one by P. H. Frings and coworkers entitled "Magnetic Properties of U,Pt Compounds," which had been presented during the summer at a magnetism conference in Kyoto that none of us had been able to attend. In this paper were data on the specific heat and magnetic susceptibility of UPt, at temperatures above 1 kelvin. These data clearly hinted at enhanced spin fluctuations.

One sign of such behavior is a magnetic susceptibility whose order of magnitude lies approximately midway between that of a nonmagnetic metal ($10^{-4}$ electromagnetic unit per mole) and that of a ferromagnetic metal ($10^{-1}$ emu/mole). Frings et al. reported a susceptibility of 0.8 x $10^{-4}$ emu/mole for UPt, a value of the right order of magnitude and similar to those of the two other known metallic spin fluctuators TiBe$_2$ (discovered at Los Alamos) and UA$_1$.

(Liquid helium-3, the other spin fluctuator known at the time, is nonmetallic.) Another sign of near magnetism is an increase in the susceptibility at some high magnetic field, indicating the transition from near magnetism to magnetism. Such an increase occurs for UPt, according to Frings et al., between 150 and 200 kilogauss. A final, sure sign of enhanced spin fluctuations is an increase, rather than a steady decrease, in the specific heat with decreasing temperature.
phonon interactions are sufficiently strong as to bring about superconductivity, an s-state condensate will be energetically favorable.

Under some circumstances a normal Fermi liquid may become almost ferromagnetic in that particle interactions give rise to internal magnetic fields that act to enhance substantially the usual Pauli paramagnetic susceptibility. In such a system low-frequency spin fluctuation excitations are likewise greatly enhanced, and the strong coupling of particles near the Fermi surface to these spin fluctuations (sometimes called paramagnons) leads to an effective mass that is frequency (and temperature) dependent. A signature of this dependence is a term in the specific heat that varies with temperature as \( T^3 \ln T \) (compared to the \( T^3 \) variation characteristic of normal Fermi liquids). Three such almost ferromagnetic metallic Fermi liquids have thus far been discovered: TiBe\(_3\), UA\(_1\), and, most recently, UP\(_3\).

Liquid helium-3 is an example of a fermion system that is both nearly ferromagnetic and, at temperatures less than 2 millikelvins, superfluid (the analogue for neutral systems of superconductivity). Its specific heat contains a \( T^3 \ln T \) term, and neutron-scattering experiments provide direct evidence for the strongly enhanced low-frequency spin fluctuation excitations responsible for that behavior. It is moreover a \( p \) -state superfluid; that is, the condensate is formed from coherent combinations of pairs of particles of parallel spins in \( p \) -states. This \( p \) -state superfluidity is not an accident: the short-range repulsion between helium-3 atoms is so strong that \( s \) -state pairing is strongly suppressed, and the interplay between that strong repulsion and the Pauli principle is responsible for the almost ferromagnetic behavior. Put another way, the particle correlations responsible for the enhanced spin fluctuations tend to oppose \( s \) -state superfluidity and to favor formation of a \( p \) -state condensate, in part as a result of the particle-spin fluctuation coupling.

It is natural therefore to hope that in metals exhibiting strongly enhanced spin fluctuations, one might possibly have \( p \) -state superconductivity of purely electronic origin. UP\(_3\), appears to be a particularly promising candidate for such an electronic analogue of liquid helium-3. Not only might it be the first metal for which electron interactions alone give rise to superconductivity, but its identification as an anisotropic superfluid could open the way to a quite new family of superconducting phenomena, in much the same way as the study of superfluid helium-3 has vastly expanded our understanding of neutral superfluid phenomena.

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This increase follows from the presence of a term proportional to \( T^3 \ln T \) in the electronic specific heat, Frings et al, reported an upturn in (the low-temperature specific heat of UP\(_3\), but gave no detailed analysis of its temperature dependence. In light of these suggestive data, we planned a more thorough investigation of UP\(_3\).

We were also interested in UP\(_3\) because of our research on the new class of materials described in the sidebar “Heavy-Fermion Superconductors.” The intermetallic compound CeA\(_1\), was regarded as a likely member of this class and yet showed no superconductivity. A study of UP\(_3\), might help explain why, since UP\(_3\) and CeA\(_1\), have the same crystal structure.

The Serendipitous Experiment

Before proceeding with our plans for UP\(_3\), we wanted single crystals of very high quality. By June of ’83 we had grown some crystals in the form of tiny whiskers (see “Single Crystals from Metal Solutions”). The best measure of the quality of a metallic crystal is its chemical resistance near absolute zero. At such low temperatures the resistance is due primarily to scattering of electrons from lattice defects since scattering from lattice vibrations is suppressed. The resistance of the whiskers was still dropping at 1.3 kelvins, our lowest easily obtainable