



## <sup>3</sup>He - Two Superfluid Phases

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A new and exciting era in the study of liquid <sup>3</sup>He began two and a half years ago when Osheroff, Richardson, and Lee<sup>(1)</sup> at Cornell University announced the discovery of two features on the melting curve of <sup>3</sup>He, occurring at 2.6 and 2.0 mK, respectively. It was first believed that the new transitions were associated with solid <sup>3</sup>He but it soon became apparent that they are properties of the liquid.

In spite of great experimental difficulties, a large amount of data is now available on liquid <sup>3</sup>He below 3 mK. We know today that there are two new low temperature phases, the A-liquid and the B-liquid, in addition to the normal Fermi-liquid; the *P,T*-diagram of <sup>3</sup>He is illustrated in Fig. 1. Experimental data, together with many theoretical investigations, show that the liquid in the A- and B-phases displays properties characteristic of a superfluid. The transition temperature for <sup>3</sup>He is about three orders of magnitude lower than for <sup>4</sup>He.

In this paper I shall first describe several experimental facts about liquid

<sup>3</sup>He in the A- and B-phases and then discuss the relevant theory. For additional information more comprehensive reviews<sup>(2),(3)</sup> should be consulted.

For investigating liquid <sup>3</sup>He below 3 mK many new methods, involving both refrigeration and thermometry, had to be developed. Pomeranchuk cooling and adiabatic demagnetization of both electronic and nuclear spins have been employed for reaching the necessary low temperature. For details I refer to other publications<sup>(4),(5),(6)</sup> and to Fig. 2.

I shall begin with the phase diagram<sup>(6),(7)</sup> of Fig. 1. The boundary between the Fermi-liquid region and the A- or B-liquids, starting at 2.6 mK (point A) on the melting curve, moves towards lower temperatures with decreasing pressure; at *P* = 0 the transition occurs at 0.93 mK. The boundary separating the A- and B-liquids has a negative slope. It starts from the melting curve at 2.0 mK (point B) and terminates at a polycritical point (PCP) at *P* = 21.5 bar and *T* = 2.4 mK. The transition from the normal Fermi-liquid to the A- or B-phases is of second order whereas the phase change between the A- and B-liquids is of first order with a small latent heat associated with it. Supercooling of 0.1 - 0.3 mK has often been observed at the AB-transition from the A-liquid to the B-liquid. We notice that the A-phase occupies only a small part of the *P,T*-diagram.

Gully, Osheroff, Lawson, Richardson, and Lee<sup>(8)</sup> have investigated the transition between the Fermi- and A-liquids as a function of *B*, the externally applied magnetic field. As *B* is increased, the AF-transition, which

is quite narrow in low fields, first broadens and then clearly divides itself into two distinct branches; the separation increases linearly as a function of *B*. The situation is different with the first order phase change between the A- and B-liquids. Osheroff, Gully, Richardson, and Lee<sup>(9)</sup> discovered that the BA-transition moves towards lower temperatures in proportion to *B*<sup>2</sup> as the applied field is increased. Both these measurements were done on the melting curve. Paulson, Kojima, and Wheatley<sup>(10)</sup> found that in a field of 30 - 40 mT (300 - 400 gauss) a narrow tail of the A-phase extends between the normal Fermi-liquid and the B-phase down to 10 bar at least.

A considerable amount of information about the superfluid phases of <sup>3</sup>He has been obtained through NMR experiments. The early measurements by Osheroff et al.<sup>(9)</sup> already showed that the signal amplitude remained approximately constant in the A-phase but, as the temperature was lowered, the resonance peak shifted steadily towards higher frequencies by a relatively large amount. When the transition to the B-phase occurred the signal moved back to its original position.

Leggett<sup>(11)</sup> was able to explain these phenomena and he predicted,

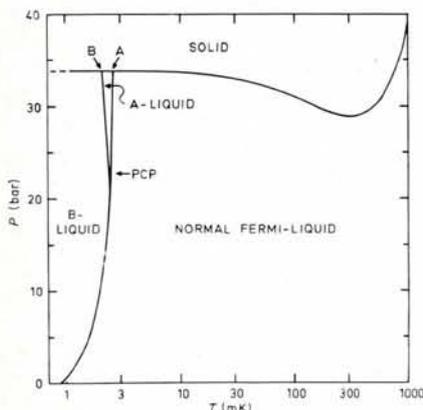


Fig. 1. The phase diagram of <sup>3</sup>He in zero external magnetic field<sup>(1)</sup> (1). Note the logarithmic temperature scale.

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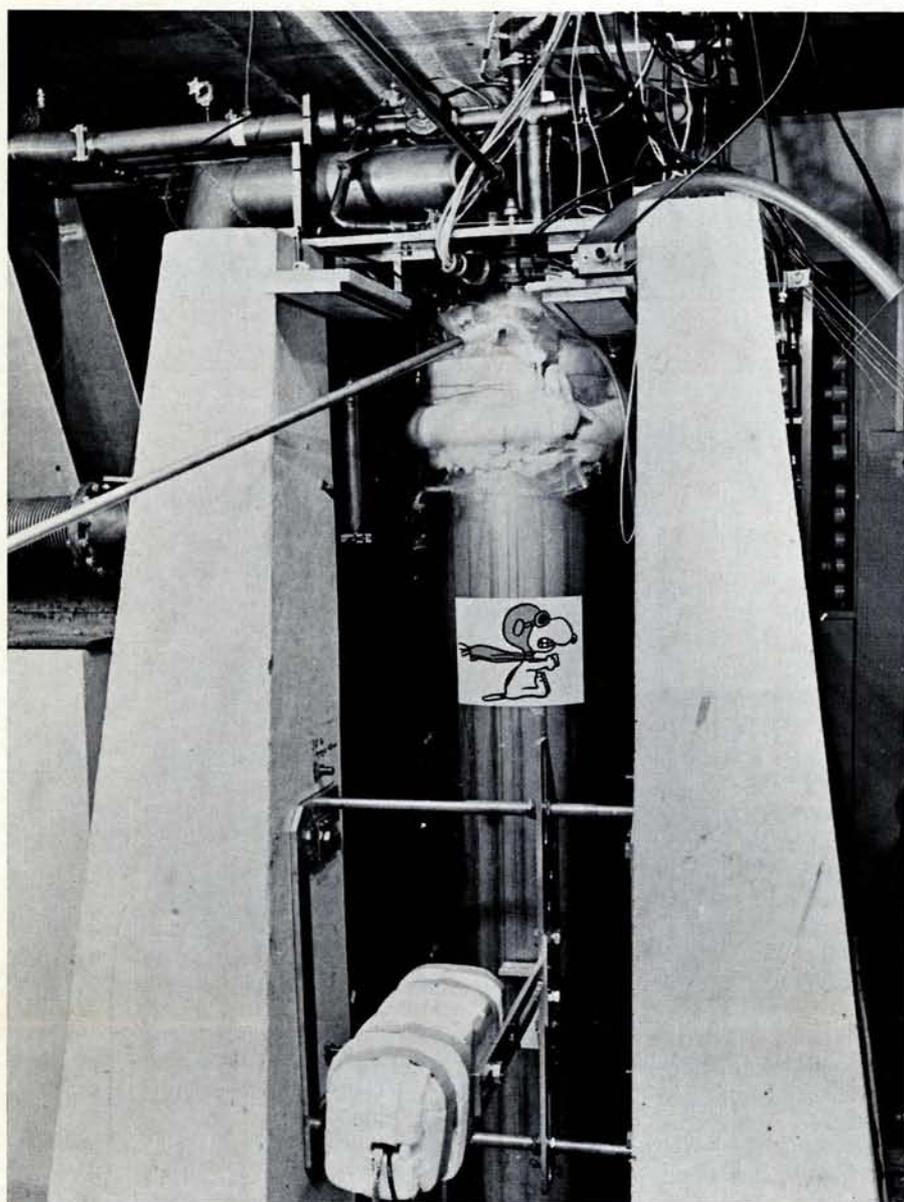


Fig. 2. The nuclear refrigeration apparatus at the Helsinki University of Technology. In order to reduce the vibrational heat leak the cryostat was mounted on a 12 ton concrete block which rests on springs. The outer liquid  $N_2$  dewar is 200 cm long and 30 cm in diameter. Precooling to 17 mK is done by a powerful dilution refrigerator. The nuclear stage consists of 22 moles of fine copper wire which can be magnetized to 7 T (70 kgauss) by a superconducting solenoid. In this apparatus liquid  $^3\text{He}$  has recently been cooled to 0.6 mK, lower than ever before. The temperature remained below 1 mK for over 20 hours<sup>(4)</sup>.

in addition, a longitudinal resonance which was subsequently first found by Osheroff and Brinkman<sup>(12)</sup>. In observing this NMR mode, which is a peculiarity of superfluid  $^3\text{He}$ , the rf-coil and the static magnetic field must be parallel to each other, in contrast to the usual perpendicular arrangement. The longitudinal and transverse resonance frequencies,  $\omega_L$  and  $\omega_T$  respectively, were found to be accurately related by the equation

$$\omega_T^2 - (\gamma B)^2 = \omega_L^2, \quad (1)$$

where  $\gamma B$  is the unshifted Larmor frequency in the Fermi-liquid region. These measurements were made at various magnetic fields along the melting curve at 34 bar pressure; simi-

lar but more comprehensive experiments have also been published by Bozler, Bernier, Gully, Richardson, and Lee<sup>(13)</sup>. Webb, Kleinberg, and Wheatley<sup>(14)</sup> have recently reported the "ringing" of magnetization in superfluid  $^3\text{He}$  when a small field  $\Delta B$ , parallel to the steady field  $B$ , is suddenly turned off.

NMR data far below the superfluid transition temperature are important for establishing the properties of the A- and B-liquids. Ahonen, Haikala, Krusius, and Lounasmaa<sup>(15)</sup> have recently made transverse NMR measurements on  $^3\text{He}$  between 0.7 and 2.6 mK and from zero to 27 bar pressure. The low temperatures were reached by means of adiabatic nuclear demagnetization of copper (cf. Fig. 2). In the A-

phase, again according to Leggett<sup>(11)</sup>, a universal curve should result by plotting

$$[\omega_T^2 - (\gamma B)^2]/T_{AF}^2$$

versus the reduced temperature  $T/T_{AF}$ ; this, indeed, was the case. In the B-phase it was found that the static susceptibility  $\chi$ , as determined by integrating under the NMR curves, saturates at 25% of its value in the Fermi-liquid region. Measurements were also made on  $^3\text{He}$  intermixed with fine platinum powder; the results were quite different from those obtained with bulk liquid.

Paulson, Kojima, and Wheatley<sup>(16)</sup> have investigated the static nuclear susceptibility of superfluid  $^3\text{He}$ . Measurements were made in a field of 38 mT and at pressures between 10 and 33 bar. It was found that in the A-liquid  $\chi$  is independent of temperature within the experimental uncertainty of only a few tenths percent. An 0.5% decrease in  $\chi$  was observed as the liquid warmed through  $T_{AF}$ .

In the B-phase  $\chi$  decreased approximately as a linear function of temperature. The lowest measured susceptibilities, at  $T/T_{AF} = 0.8$ , were about 35% of  $\chi$  in the Fermi-liquid region, without any clear indication of saturation to a finite value at still lower temperatures.

Webb, Greytak, Johnson, and Wheatley<sup>(17)</sup> have measured the specific heat of  $^3\text{He}$  near the AF-boundary. The shape of the curve was quite similar to that found for the heat capacity of a superconductor around the critical point. The transition occurred over a temperature interval which was less than  $3 \mu\text{K}$ . Precise measurements revealed no traces of a latent heat, i.e. the phase change appears to be of second order.

Fourth sound is a pressure wave propagating through a superfluid in which the normal liquid (density  $\rho_n$ ) is locked by a porous medium. The relative density of the superfluid component can be calculated from the equation

$$\rho_s/\rho = c_4^2/c_1^2, \quad (2)$$

where  $c_1$  and  $c_4$  are the velocities of the first (= ordinary) and fourth sound, respectively, and  $\rho = \rho_n + \rho_s$ .

Kojima, Paulson, and Wheatley<sup>(18)</sup> have observed fourth sound by pulse and resonance methods; the results prove that  $^3\text{He}$  is a superfluid. Their experimental chamber was packed with cerium magnesium nitrate powder; the salt was employed as the refrigerant, superleak, and thermometer. The data show that  $\rho_s/\rho$  is

quite small, varies slowly with temperature, and displays little or no anomalous behaviour at the boundary between the A- and B-liquids. Similar results have recently been obtained by Yanof and Reppy<sup>(19)</sup>.

I shall next explain, little more in detail, measurements of viscous damping in liquid <sup>3</sup>He by Alvesalo, Anufriyev, Collan, Lounasmaa, and Wennerström<sup>(20)</sup>; the experiments were restricted to the melting curve. The <sup>3</sup>He cell was equipped with a vibrating wire viscometer. The string, 25 mm long, 0.25 mm thick, and made of superconducting Nb-Ti wire, was under a tension which gave the system a natural resonant frequency of about 1200 Hz. During measurements an 0.1 T magnetic field was applied perpendicular to the string and an ac-current of 2 mA was passed through it. The Faraday voltage appearing across the ends of the vibrating wire was measured as the frequency of the excitation current was swept semi-continuously through the resonance. The bulk liquid viscosity  $\eta$  is inversely proportional to the square of the signal amplitude.

Results of several runs are shown in Fig. 3. It is observed that as the Fermi-liquid cools from 15 mK, the amplitude first decreases, reaches a minimum at A (cf. Fig. 1), and then starts to increase. After a fast initial growth in the A-liquid the signal continues to increase more slowly while the temperature is further reduced, until at B the vibration amplitude jumps discontinuously and then in-

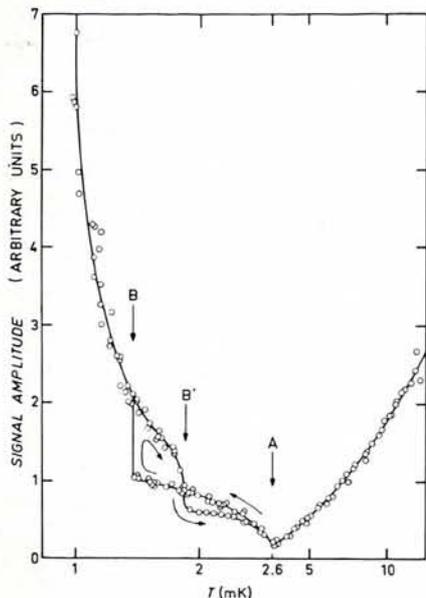


Fig. 3. The signal amplitude of the vibrating wire viscometer at resonance<sup>(20)</sup>. Points labeled A and B refer to Fig. 1. The hysteresis loops are marked by arrows; during warming the BA-transition occurred at B'. Note that the temperature scale is expanded below 2.6 mK.

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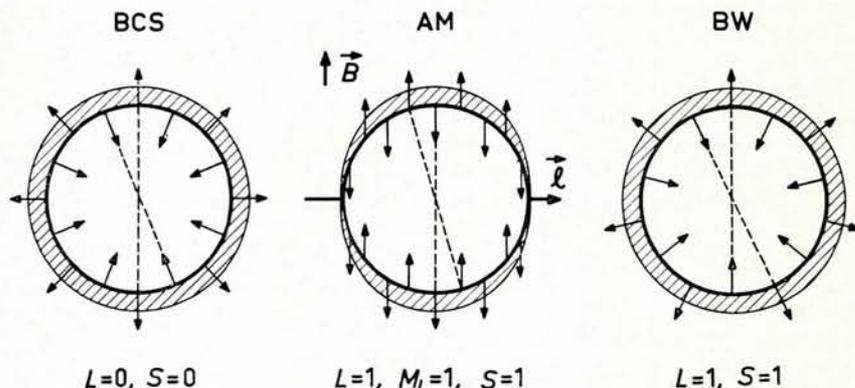


Fig. 4. An "over-simplified" picture of the BCS, AM, and BW ground states. Pairing occurs between two  $^3\text{He}$  atoms on opposite sides of the Fermi-sphere; two pairs are indicated in each case with a dashed line. In the AM- and BW-states the spins (shown by arrows) of paired atoms are parallel to each other, in contrast with the antiparallel arrangement of the BCS-state. Shaded areas illustrate the energy gap  $\Delta$ . In the AM-state the anisotropic  $\Delta$  is symmetric about the  $l_z$ -axis.

creases rapidly in the B-liquid. The height of the viscometer signal at the lowest experimental temperature, probably near 1.0 mK, is almost four times the amplitude at 10 mK and is increasing with decreasing temperature very quickly indeed. Two hysteresis loops, associated with the transition between the A- and B-liquids, are also clearly seen. These measurements provided the first dramatic indication of superfluidity in  $^3\text{He}$ .

The experiments have recently been repeated with improved techniques<sup>(21)</sup>. The new data show that resistive flow of liquid  $^3\text{He}$  in the A- and B-phases is accompanied by a flow of zero viscosity; this proves superfluidity of  $^3\text{He}$ . In terms of the two-fluid model, damping of a vibrating string is caused by the normal fraction of the liquid; the superfluid component manifests itself only through the hydrodynamic mass of the wire. By measuring accurately the amplitude or the width and the frequency shift of the resonance curve it is possible to calculate separately the viscosity  $\eta_n$  and the density of the normal component.

The results show that as liquid  $^3\text{He}$  cools through the FA-transition,  $\eta_n$  decreases rapidly, within 0.3 mK, to about 25% of its value in the Fermi-liquid region and then stays essentially constant. At the AB-boundary  $\eta_n$  drops an additional 5% and then starts to increase as the temperature is further reduced. The viscosity of liquid  $^3\text{He}$  thus behaves qualitatively in a manner familiar from experiments on liquid  $^4\text{He}$  below 2.2 K.

According to Alvesalo et al.<sup>(21)</sup> the normal density decreases relatively slowly with temperature in the A-phase, approximately as  $T/T_{AF}$ ; this is in fair agreement with the fourth sound measurements<sup>(18), (19)</sup> discussed

earlier. At the AB-boundary, however,  $\rho_n$  drops abruptly from about 60% of the bulk liquid density to 25% of  $\rho$  and then decreases rapidly upon further cooling. Below 1.3 mK,  $\rho_n$  is less than 1% of  $\rho$ . The rapid variation of  $\rho_n$  as a function of temperature in the B-phase is in disagreement with the data of Kojima et al.<sup>(18)</sup> and of Yanof and Reppy<sup>(19)</sup>; these authors observed no appreciable change in the temperature dependence of  $\rho_s/\rho$  at the AB-boundary. The discrepancy may indicate that the properties of superfluid  $^3\text{He}$  are changed when the liquid is confined inside a porous medium necessary for the observation of fourth sound<sup>(15)</sup>.

The discovery of superfluidity in liquid  $^3\text{He}$  did not come unexpectedly; it had been foreseen by several individuals ever since the BCS-theory of superconductivity was published in 1957. However, predictions for the transition temperature varied many orders of magnitude. Superfluidity in  $^3\text{He}$  is more closely related to superconductivity in metals than to superfluidity in  $^4\text{He}$ ; the first two phenomena are characterized by pairing of Fermi-Dirac particles whereas superfluidity in  $^4\text{He}$  is related to Bose-Einstein condensation.

According to the BCS-theory of superconductivity, two electrons on opposite sides of the Fermi-surface and of opposite spins feel a slight attraction. If this interaction is not too weak the electrons form a Cooper pair and fall into the superconducting ground state. The attractive potential can be described as a slight deformation of the crystalline lattice by one of the electrons which is sensed by its partner. The BCS-ground state (cf. Fig. 4), separated from the single-electron excited states by an energy

gap  $\Delta$ , is spherically symmetric with both the orbital angular momentum and the spin of the pair vanishing, i.e.  $L = 0$  and  $S = 0$ .

A rather analogous situation, with a Lennard-Jones type potential, exists between quasiparticles in liquid  $^3\text{He}$ . However, the strong repulsive core associated with molecular interaction prevents  $L = 0$  pairing and a higher value of the orbital angular momentum is required for an attractive potential.  $^3\text{He}$  atoms on the Fermi surface have a relatively high speed,  $v_F = 50$  m/s. The liquid is thus able to lower its energy if pairs of atoms circle around each other; centrifugal forces then keep them outside the range of the repulsive core.

Because  $^3\text{He}$  is a fermion the total wave function of the pair must be antisymmetric. Thus, for an even  $L$ ,  $S = 0$ , whereas if  $L$  is odd,  $S = 1$ . The nearly ferromagnetic character of liquid  $^3\text{He}$  tends to create clusters of atoms with parallel spins; this favors states with an odd  $L$ . For a sufficiently large  $L$  the interaction is always attractive. In the ground state the orbital angular momentum naturally assumes the value which produces the strongest pair.

Rather soon after the BCS-theory was accepted, Anderson and Morel<sup>(22)</sup> applied it to the case  $L = 1$  and  $S = 1$ , with spin components  $S_z = \pm 1$  only taken into account. Somewhat later Balian and Werthamer<sup>(23)</sup> considered the same case with the component  $S_z = 0$  included. It is now believed that the A-liquid is in an Anderson-Morel (AM) phase and, with some uncertainty, that the B-liquid is in the state suggested by Balian and Werthamer (BW).

The AM-state is anisotropic; its energy gap  $\Delta$  varies as  $\sin \Theta$  where  $\Theta$  is measured from the gap axis  $\ell$  (cf. Fig. 4). Pairing occurs separately between spin-up and spin-down particles. Each pair has  $M_L = 1$  in the direction of  $\ell$  and  $S_d = 0$  along another axis  $\mathbf{d}$ . In order to minimize the dipolar energy,  $\ell$  and  $\mathbf{d}$  orient parallel to each other. In this way the spin vectors are in the plane of pair rotation and thus, part of the time, behind each other which lowers the energy; if the spins were perpendicular to the plane of rotation they would always be side-by-side. The Zeeman energy turns  $\mathbf{d}$  perpendicular to the applied magnetic field  $\mathbf{B}$ . Depairing effects will tend to orient  $\ell$  perpendicular to any surface in contact with the liquid; the pair thus rotates in a plane parallel to the surface. It is likely that in small diameter geometries surface effects do-

minate over magnetic energy at low fields.

In the isotropic BW-state (cf. Fig. 4) pairs with  $L = 1$ ,  $S = 1$ , and total angular momentum  $J = 0$  are first formed. The spin variables are then rotated around an arbitrary axis; dipolar forces fix the angle of rotation to  $104^\circ$ . Depairing effects weakly align the axis of rotation parallel to  $\mathbf{B}$  and perpendicular to surfaces. The field energy seems to dominate above 25 mT.

Leggett<sup>(11)</sup> first showed that NMR data<sup>(9)</sup> in the A-liquid could be explained in terms of an AM-state; his theory also predicted a longitudinal resonance which was later observed<sup>(12), (13)</sup>. In transverse NMR the precession of magnetization around the static field  $\mathbf{B}$  causes  $\mathbf{d}$  to oscillate about the direction of  $\ell$ . The increased resonance frequency, as observed in the A-liquid [cf. Eq. (2)], implies odd- $L$  pairing and is caused by the restoring force between  $\mathbf{d}$  and  $\ell$ , which acts in addition to the magnetic field. In longitudinal resonance the angle between  $\mathbf{d}$  and  $\ell$  is forced to oscillate directly by applying a sinusoidal signal parallel to  $\mathbf{B}$ . In the particular AM-state proposed by Anderson and Brinkman<sup>(24)</sup> the longitudinal and transverse resonance frequencies are related exactly by Eq. (1). Experimental confirmation is really excellent<sup>(9), (12), (13), (15)</sup>.

Mermin and Ambegaokar<sup>(25)</sup> have shown that an anisotropic phase with odd- $L$  pairing, such as the AM-state, explains the splitting of the AF-transition in a magnetic field. Pairing occurs independently within the  $S_z = +1$  and  $S_z = -1$  spin systems. The transition temperature of the  $S_z = +1$  population is raised and that of the  $S_z = -1$  system is depressed by amounts which are linear functions of the external magnetic field. In this system  $\chi$  behaves like the Pauli susceptibility and is independent of temperature in agreement with experiments.

The quadratic depression of the AB-phase boundary as a function of the applied magnetic field is in agreement with spherically symmetric  $S = 1$  pairing, i.e. with the BW-state. The same field dependence is obtained, however, if even- $L$  pairing is assumed. In this case the  $^3\text{He}$  atoms have opposite spins, i.e.  $S = 0$ , and the field induced imbalance in the spin system competes with pairing. The net effect is, because the magnetic energy is proportional to  $B^2$ , that  $T_{AB}$  is lowered quadratically as a function of  $B$ .

Which, if either, of these two possibilities actually occurs in the B-liquid? This can be determined from magnetic susceptibility data if measurements are extended sufficiently far into the B-phase. For  $S = 1$  pairing  $\chi$  saturates with decreasing temperature to 35-40% of its value in the Fermi-liquid region, whereas in the  $S = 0$  case  $\chi$  is reduced to zero. The measurements of Osheroff and Brinkman<sup>(12)</sup> are in favor of  $S = 1$  pairing whereas the data of Paulson et al.<sup>(16)</sup> suggest  $S = 0$  pairing presumably in an  $L = 2$  state. The recent results of Ahonen et al.<sup>(15)</sup>, giving a 25% limiting value, point to a more complex situation. There is thus a possibility that the B-liquid is not in a BW-state after all.

Many other experimental results are also in general agreement with pairing theories of the BCS-type. The heat capacity immediately below the AF-transition should be a factor of 2.4 larger than immediately above it. Experimentally<sup>(17)</sup> the ratio is 2.9; inclusion of spin fluctuation effects may remove this discrepancy. Shumeiko's<sup>(26)</sup> calculations of the viscosity are in fair agreement with experiments<sup>(21)</sup>.

Similarly measurements<sup>(27), (28)</sup> on the attenuation of zero sound are in rough accord with theory<sup>(29)</sup>.

It is truly amazing how much information has been obtained about liquid  $^3\text{He}$  below 3 mK, both experimentally and theoretically, in a little more than two years. Superfluidity has been proven and many properties of the new phases are already known quite precisely. This rapid progress shows both the vitality of current research at ultralow temperatures and the importance of these phenomena for modern theory of collective systems. However, as further developments and refinements occur, many of the statements in this review must surely be modified.

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# Bose-Einstein Condensation of Excitons

Interest has been revived in high density excitons in semiconductors by some recent experiments that seem to indicate the existence of a new phase where excitons, or biexcitons undergo Bose-Einstein condensation.

If light of photon energy larger than the band gap is sent onto a semiconductor, free electron-hole pairs are formed. At low enough temperatures the electron-hole Coulomb attraction leads to a bound pair called exciton similar to a hydrogen atom. Excitement about high density excitons started in 1968, when V.M. Asnin and A.A. Rogachev<sup>1)</sup> from the Ioffe Physico-Technical Instit., Leningrad, showed that when germanium was hit by laser light of increasing intensity — thus creating excitons in the sample — a change of state of the exciton gas was taking place at some critical exciton density. This was very suggestive of a Mott-type insulator-metal transition of the exciton system which is expected when the exciton spacing is comparable with the exciton diameter. The idea that the lowest energy state of the exciton system was in fact "metallic" droplets was first suggested by L.V. Keldysh<sup>2)</sup> from the Lebedev Physical Institute in Moscow. Subsequent luminescence and light scattering experiments were soon carried out by Ya. Pokrovskii<sup>3)</sup> from the Institute of Radioengineering and Electronics of the Academy of Sciences in Moscow, and brought out that in the high density phase the excitons had dissolved into free electrons and holes, indeed forming "metallic" droplets, with typical radii of some microns, in thermal equilibrium with the exciton gas. Very good agreement was obtained by subsequent detailed work by Nozières and Combescot<sup>4)</sup> of Ecole Normale, Paris, and W.F. Brinkman and T.M. Rice<sup>5)</sup> of Bell Labs. Though this was quite satisfactory, it has also been clear that the free exciton gas and the electron-hole plasma droplet are not the only possible states for a collection of excitons. In the first place, excitons bear an extraordinary resemblance with hydrogen atoms, and can bind in pairs to form molecules, or "biexcitons", analogous to H<sub>2</sub>. Predicted very long ago, biexcitons have been observed in luminescence spectra of many crystals. The energetic competition between a biexciton gas and the droplet state has also been investigated by W.F. Brinkman and T.M. Rice<sup>6)</sup>

and the conclusion was that the details of the band structure play a crucial role.

Another possibility comes from Bose-Einstein statistics, which both excitons and biexcitons follow approximately, being made up with even numbers of fermions. The phenomenon of Bose-Einstein condensation (BEC), which occurs at a temperature  $T_c = \hbar^2/12 k m (N/V)^{2/3}$  for  $N$  noninteracting perfect bosons of mass  $m$  in a volume  $V$  consists in a macroscopic population of particles in the lowest energy state. BEC might occur also for excitons (and biexcitons), as has been pointed out more than 10 years ago by Moskalenko<sup>6)</sup> from the Institute of Physics and Mathematics in Kishinev, Blatt and coworkers from New York University and Casella from IBM Watson Research Center, Yorktown Heights. Other systems, in which BEC is in discussion, are superfluids such as <sup>4</sup>He below the  $\lambda$ -point and Cooper pairs in a superconductor. The significance of experiments on exciton BEC lies in the new approach to this fundamental problem. One of the obstacles in the way of a possible condensation is the Pauli principle, which, by requiring antisymmetry of the total wave function to fermion exchange, prevents macroscopic occupancy of the exciton states, and thus removes that feature of Bose statistics which is crucial to BEC. It is, however, clear that this problem is not necessarily a serious one, e.g., as long as the exciton spacing is large enough to allow for the neglect of the exciton (or biexciton) size in comparison. The low temperature state in this case is only in principle distinct, but thermodynamically indistinguishable, from a Bose-Einstein condensate.

A second obstacle is interaction between the excitons, or biexcitons. Even in the large distance limit specified before, the net effect of the Pauli principle is equivalent to a repulsive exciton-exciton force. In addition to this, theorists like P.W. Anderson, S.T. Chui, and W.F. Brinkman<sup>7)</sup> of Bell Labs envisage at least another force, due to screening of the electron-ion potential, which seems to cancel the Pauli repulsion and leads to a net attractive exciton-exciton force. At the opposite limit of short distances, excitons, of course, attract each other, in the attempt to bind and form a biexciton. This overall attraction is

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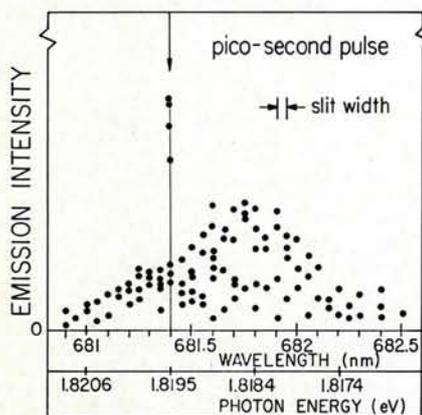


Fig. 1. Luminescence spectrum of a CdSe crystal at 1.8 K under the pico-second light pulse excitation (slit width: 0.05 nm); from ref. 9.