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CURRENT PROBLEMS IN RESEARCH

Liquid Helium-3

The low-temperature properties of the rare isotope provide a basis for a new theory of liquids.

J. G. Daunt

Some of the more interesting problems in current research at very low temperatures concern the behavior and properties of pure liquid helium-3. Although this rare isotope of helium was discovered as long ago as 1939 by Alvarez and Cornog (*1*), a decade elapsed before its low-temperature properties could be studied on a macroscopic scale, the first liquefaction of pure He^3 having been made by Sydoriak, Grilly, and Hammel (*2*) in the Los Alamos Scientific Laboratory in 1949. At this date He^3 became available as the decay product of tritium, and the subsequent increasing availability of this rare isotope from the U.S. Atomic Energy Commission and in the U.S.S.R. has made possible a significant effort toward the solution of many problems in the past several years.

No attempt will be made here to make a complete review of past work or to discuss any of the problems associated with the solutions of He^3 in liquid He^4. I have already reviewed the earlier work, particularly that carried out during the difficult days when He^3 was available only in its natural abundance of about one part per million in He^4 (*3*), and many subsequent reviews have been published (*4–7*). The aim of the present article is to present some current research problems associated with the low-temperature properties of liquid He^3 from an experimental point of view.

Helium-3 differs from He^4 not only in its mass but also in possessing a nuclear spin (*8*) of ½. As was emphasized many years ago by the late Fritz London (*3, 5, 9*), this difference in nuclear spin might be expected to result in significant differences in the macroscopic behavior of the systems in their liquid phases, since He^3 would conform with Fermi-Dirac statistics whereas He^4 would be a Bose-Einstein system. On the other hand, the development of a quantum theory of corresponding states by de Boer and his coworkers (*10*) leads one to expect a marked similarity in the behavior of the two isotopes. The most striking similarity lies in the fact that under their saturated vapor pressure both He^3 and He^4 remain liquids down to absolute zero temperature. This circumstance of having both a Fermi and a Bose liquid existing at the very lowest temperatures, where one might expect unusual simplicity in their behavior, makes these liquids of particular interest. If a fitting picture of the liquid state is to be built up, it is surely here that the most promising investigations are to be made.

In considering the known and possible similarities and differences, two particular questions arise very significantly. The first question, which one would wish to answer by experimental methods, concerns the character of the elementary thermal excitations in the liquids. The second question concerns the occurrence of superfluidity, a phenomenon which is well known in liquid He^4 below its lambda temperature (*7, 11, 12*) and which theoretically may be expected also in a Fermi system, provided the attractive potential is sufficiently great (*13*). These two problems are closely interwoven and, from an experimental point of view, often are attacked by observations of the same type.

One of the earliest indications of an anomalous behavior in liquid He^4—a behavior later associated with the phenomenon of superfluidity—stemmed from measurements of the specific heat (*11, 12*). Figure 1 shows the specific heat of liquid He^4 as a function of temperature and illustrates impressively the sensitive indication which the specific heat yields of the collective transformation into the superfluid state as the temperature is reduced through the lambda point at 2.17°K. From such measurements conclusions could be drawn about the general character of the thermal excitations, as was done, for example, in the pioneering theoretical work of Landau (*14*) and in the subsequent development of that work by Feynman (*15*). Measurements of the specific heat at very low temperatures, below approximately 0.6°K (*16*), reveal a cubic dependence on temperature, indicating that at these temperatures the excitations are in the form of longitudinal acoustic waves, or phonons. At higher temperatures the rapid and almost exponential rise in the specific heat of the superfluid liquid characterizes excitations of much shorter wavelength, which have been termed rotons by Landau, and which must involve associated motion of small groups of atoms.

More recently this problem of the thermal excitations in liquid He^4 has been elegantly attacked in an entirely different manner, in accordance with a detailed theoretical proposal by Cohen and Feynman (*17*). The experiments were initiated by Palevsky and his coworkers (*18*) and further and independently developed by Yarnell, Arnold, Bendt, and Kerr (*19*) and by Henshaw (*20*). These workers observed the wavelength of slow neutrons inelastically scattered by liquid He^4, the scattering process being one in which the neutron creates a single thermal excitation in the liquid. By measurement of the

The author is professor of physics and astronomy at Ohio State University, Columbus.

change in wavelength due to scattering as a function of scattering angle, they were able to deduce the energy spectrum of the thermal excitations—that is, the energy as a function of momentum. The results strikingly confirmed and made more precise the conclusions outlined above, which had already been drawn from the earlier specific-heat data. Figure 2 shows graphically this energy spectrum.

No detail need be given here concerning the experimental establishment of the phenomenon of superfluidity in liquid He^4, first discovered in observations on fluid flow by Kapitza (*21*) and by Allen and Misener (*22*), since many reviews of this material exist (*7, 11, 12*).

Along the same lines of experimental attack, considerable attention has been paid to the observation of the specific heat of liquid He^3. This attention has been the more intense since the elegant method of slow neutron scattering is unavailable because of the extremely large absorption cross section of the He^3 nucleus (*23*). The first measurements of the specific heat of liquid He^3 were made by de Vries and Daunt (*24*), by Roberts and Sydoriak (*25*), and by Osborne, Abraham, and Weinstock (*26*); these measurements covered the range of temperature from 2.3° down to approximately 0.23°K. The results

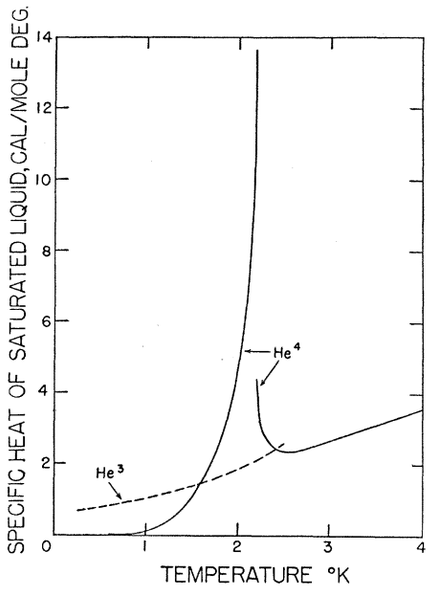


Fig. 1. The specific heat of liquid He^4 and liquid He^3 under their saturated vapor pressures as a function of temperature. Data for He^4 were taken from the work of W. H. Keesom and his co-workers (*11*) and for He^3, from the work of de Vries and Daunt (*24*), Roberts and Sydoriak (*25*), and Osborne, Abraham, and Weinstock (*26*).

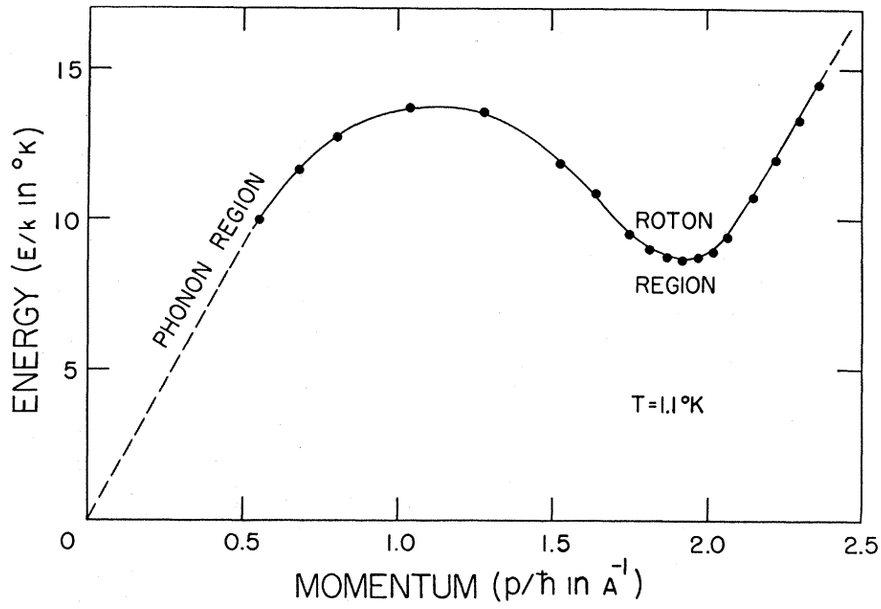


Fig. 2. The energy spectrum for thermal excitations in liquid He^4, the energy being plotted as a function of momentum. [From the work of Yarnell, Arnold, Bendt, and Kerr (*19*)]

are shown in Fig. 1 and give no indication of any lambda anomaly in the range of temperature covered. It was natural to conclude, therefore, that a transformation to superfluidity does not occur down to 0.23°K. This conclusion was supported by other experiments, particularly by the observation of the occurrence of only normal viscous flow in liquid He^3 in the range 1° to 3°K, by Osborne, Weinstock, and Abraham (*27*), and by the early observation of the monatomic decrease in the lambda temperature of He^4 in mixtures of He^3 and He^4 down to approximately 0.4°K, by Daunt and Heer (*28*).

It is interesting to note in Fig. 1 the very high absolute value of the specific heat of He^3 at the lower temperatures. For example, at 0.25°K it is 0.684 cal/moledeg, a value which is 1954 times larger than that for liquid He^4. Liquid He^3 near absolute zero, therefore, possesses an abundance of low-energy excitations not possessed by liquid He^4. As is clear from Fig. 1, the results down to 0.23°K do not permit unambiguous extrapolation of the specific heat to 0°K. There are many possible extrapolations consistent with the entropy requirements, and a variety of speculations have been made (*29*). In 1958 the results from measurement of the specific heat of liquid He^3 at approximately its saturated vapor pressure down to a temperature of 0.08°K were published by Brewer, Sreedhar, Kramers, and Daunt (*30*); these results are shown in Fig. 3. They permit a justifiable linear extrapolation to be made to 0°K, and they strongly support the theoretical picture of the liquid as a degenerate Fermi system of interacting particles, which had previously been put forward theoretically by Brueckner and Gammel (*31*) and by Landau (*32, 33*). In the region of the linear dependence of the specific heat on temperature, the degenerate Fermi liquid can be characterized by an effective mass m^*, given by:

$$m^*/m = C/C_F \qquad (1)$$

where C_F is the specific heat of an identical system of Fermi atoms without interaction and where m is the mass of the free He^3 atom. The experimental results yield a value of 2.0 for m^*/m, surprisingly close to the value of 1.76 obtained in the theoretical computations of Brueckner and Gammel.

A by-product of these results is the conclusion that one may set a still lower limit for the occurrence of superfluidity, the lack of a significant specific-heat anomaly indicating an absence of superfluidity down ot at least 0.08°K. It is not possible, however, to conclude from these results that superfluidity may not yet occur at still lower temperatures. If such a transformation were to occur, it is in principle possible for it to be such that the entropy of the liquid above the transformation temperature is the same as that which is obtained by the linear extrapolation of the known results. The occurrence of superconductivity in

metals is one example of a transformation fulfilling similar entropy requirements. A detailed theoretical investigation of the criterion for the occurrence of superfluidity in degenerate interacting Fermi systems and, in particular, in liquid He^3 has recently been made by Cooper, Mills, and Sessler (*13*). They concluded that superfluidity was in principle possible, but that owing to the very weak attractive forces in liquid He^3, it would be unlikely to occur unless, in some as yet unknown way, the number density of atoms could be considerably diminished. It is still, therefore, of considerable interest to make experimental studies of liquid He^3 at still lower temperatures, and it is hoped that it may not be too long before measurements down to 0.01°K will be technically feasible.

In the development of the theory of liquid He^3 as a Fermi liquid one of the most important early experimental landmarks was the elegant measurement by Fairbank and his co-workers (*34*) on the nuclear paramagnetic susceptibility. In making their observations of the susceptibility they used nuclear magnetic resonance techniques down to a temperature of 0.1°K and at a variety of pressures. Figure 4 gives some of the typical results for the liquid under its saturated vapor pressure; the quantity $\chi T/c$ is plotted as a function of temperature, where χ is the susceptibility and c is the Curie constant. It will be noted that at the lowest temperatures the curve approaches the origin linearly; this indicates that the susceptibility becomes independent of the temperature, as one would expect for a Fermi system.

Other predictions can be made concerning the behavior of liquid He^3 at the lowest temperatures—that is, below about 0.15°K—in the degenerate region of the Fermi liquid. The viscosity should be given by a/T^2, as has been pointed out by Abrikosov and Khalatnikov (*6, 35, 36*), with a lying between 10^{-5} and 10^{-6} centimeter-gram-second (cgs) units. The lowest temperature to which measurements of the viscosity have so far been made is about 0.35°K, in the work of Zinov'eva (*37*). She observed (see Fig. 5) that the viscosity increases with decreasing temperature but not as rapidly as $1/T^2$. The heat conductivity in the degenerate Fermi liquid region, according to Abrikosov and Khalatnikov (*6, 35*), should be given by $K = \beta/T$, with β lying between 10^2 and 10^3 cgs units—a prediction which has not yet been experimentally verified. The measurements of the heat conductivity which have been made—for example, by Fairbank and Lee (*38*)—do not extend below about 0.2°K and show a monatomical increase in heat conductivity with increasing temperature. As has been pointed out by Abrikosov and Khalatnikov (*6, 35*), the validity of these theoretical derivations of the viscosity and heat conductivity, and indeed of the whole model of the Fermi liquid, requires that the temperature be sufficiently low so that the quantum indeterminacy in the energy of the excitations due to their collisions is much smaller than the average excitation energy. Since the average excitation energy is of the order of kT, this yields the requirement:

$$kT \gg \hbar/\tau \qquad (2)$$

where τ, the time between collisions, is inversely proportional to T^2. By esti-

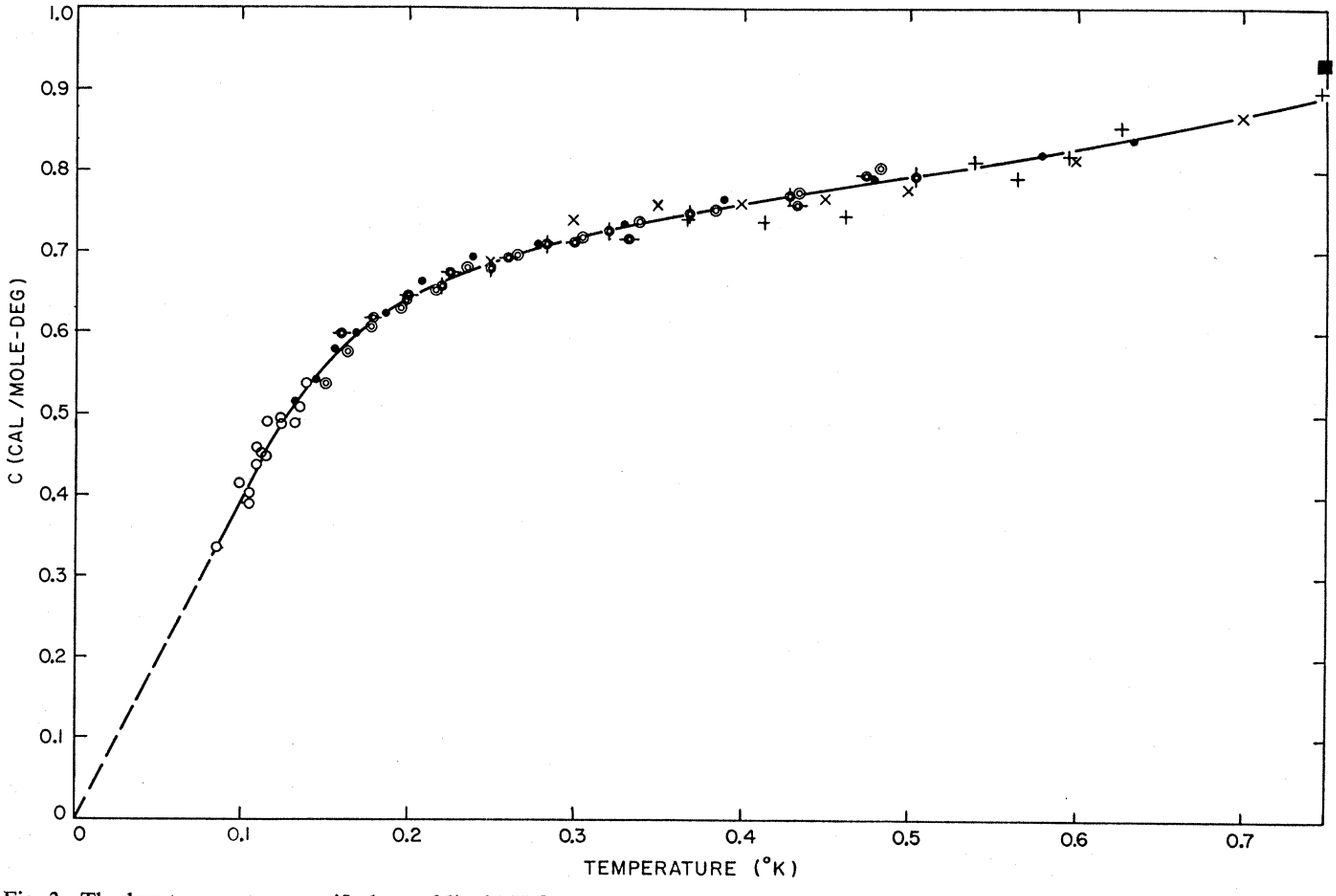


Fig. 3. The low-temperature specific heat of liquid He^3 under a pressure of a few centimeters of mercury. [From the work of Brewer, Sreedhar, Kramers, and Daunt (*30*); see also, Brewer, Daunt, and Sreedhar (*29*)]

mating the constant of proportionality from Zinov'eva's viscosity data, Abrikosov and Khalatnikov conclude that the validity requirement is

$$T \ll 0.3°\text{K} \qquad (3)$$

indicating that temperatures at least as low as 0.1°K must be attained in measurements of viscosity and heat conductivity in order to test the theory.

The propagation of sound has been theoretically investigated by Landau (*32*) and by Abrikosov and Khalatnikov (*6, 39*), and it is predicted that at the lowest temperatures it should exhibit several unusual features. The characteristic features of propagation are dependent on the relative values of the period of oscillation of the sound wave and of the collision time τ in the liquid.

For low-frequency sound, for which the period of the sound wave is large as compared with the collision time, sound propagation occurs in the usual manner and is due to the compressional waves in the liquid. Thus, when $\omega\tau \ll 1$, where ω is the angular frequency of the sound wave, normal first sound results, characterized by a displacement of the Fermi surface as a whole. Its absorption coefficient γ should be, according to Abrikosov and Khalatnikov (*3, 39*):

$$\gamma \approx 10^{-17}\ (\omega/T)^2\ \text{cm}^{-1} \qquad (4)$$

the increase in attenuation as the temperature is reduced being due to the corresponding increase in the viscosity. The velocity of first sound has been measured by Laquer, Sydoriak, and Roberts (*40*) and by Atkins and Flicker (*41*), Laquer and his co-workers having made measurements down to 0.34°K and having obtained a value for the velocity extrapolated to 0°K of c_1=183.4 m/sec. No measurements of the attenuation have yet been made.

As the frequency is increased or the temperature is reduced, or as both these changes occur, the situation will arise when $\omega\tau \approx 1$. In this event first sound is completely damped. Further increase in ω or decrease in T, or both, introduces, according to Landau (*32*), a new kind of sound propagation, called by him "zero sound," with a velocity of propagation different from that of first sound. For zero sound, $\omega\tau \gg 1$—that is, the period of the sound oscillations is shorter than the time between collisions, and the wave is characterized by an oscillating distortion of the Fermi surface only. Abrikosov and Khalatnikov (*6, 39*), using Zinov'eva's (*37*) viscosity data, estimate that:

$$\tau \approx 2.3 \times 10^{-12}\ T^{-2}\ \text{sec} \qquad (5)$$

so that for generation of zero sound with a readily available ultrasonic frequency of, say, 10 megacycles per second, temperatures at least as low as about 0.01°K would be required in the liquid He³. It would be of extreme interest to attempt the generation of zero sound experimentally, but clearly it will involve the marriage of two advanced techniques—namely, of very high-frequency sound generation and very low temperatures that can be attained only by magnetic cooling. Moreover, theoretical estimates have been made of the attenuation coefficient and velocity which clearly call for experimental verification.

A possible alternative to direct generation and observation of zero sound has been suggested by Abrikosov and Khalatnikov (*6, 42*). They proposed

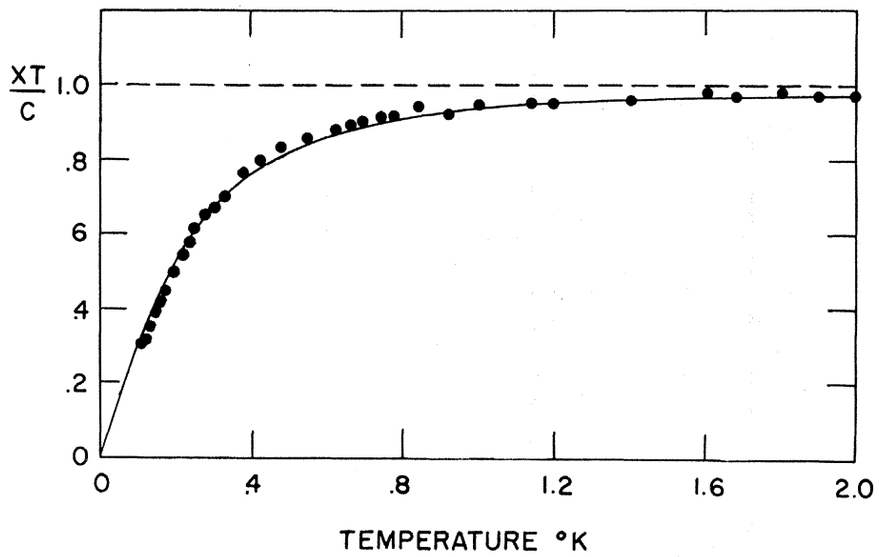


Fig. 4. The magnetic susceptibility χ of liquid He³ [from the work of Fairbank and Walters (see *34*)]. The ordinate plots the term $\chi T/c$, where c is the Curie constant. The full curve gives the value of $\chi T/c$ of an ideal Fermi-Dirac gas with a degeneracy temperature of 0.45°K.

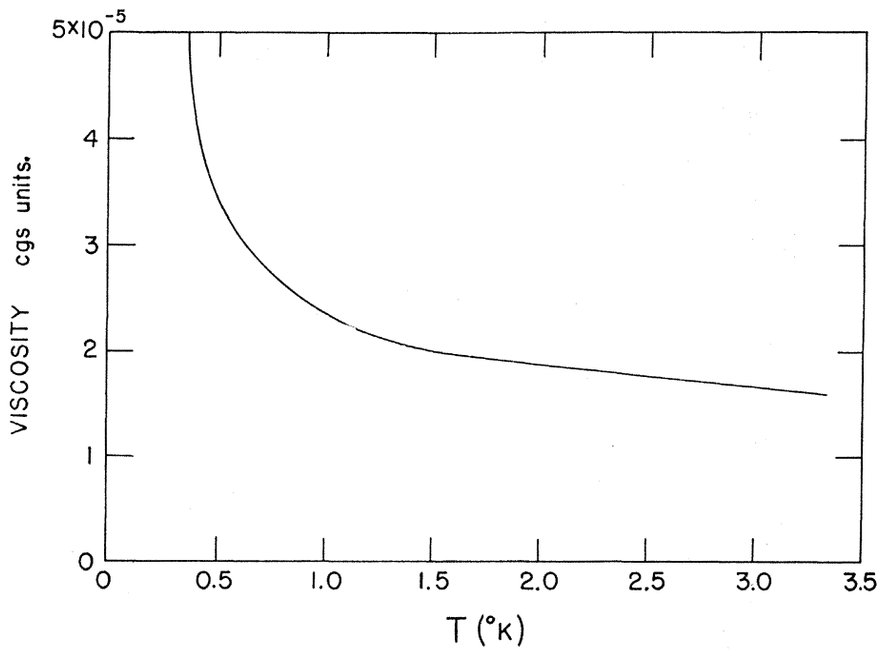


Fig. 5. The viscosity η of liquid He³ as a function of temperature. [From the work of Zinov'eva (*37*)]

that the velocity of zero sound could be deduced from observation of the intensity of satellite lines in Rayleigh scattering of light from the liquid. However, they estimate that, even for liquid temperatures of 0.01°K, light of wavelength well into the ultraviolet region would be required to produce an observable effect.

If it is possible to propagate zero sound in the liquid, the propagation would be possible even at $T = 0$°K. Therefore, one would conclude that at the lowest temperatures there must be Bose excitations such that the excitation energy ϵ is given by:

$$\epsilon = c_0 p \qquad (6)$$

where c_0 is the zero sound velocity and p is the excitation momentum. This, as in liquid He^4, would give a T^3 contribution to the specific heat. Numerical estimates (*6*, *39*) give $c_0 \approx 2 \times 10^4$ cm/sec, so that the contribution to the specific heat would be small compared with the linear term (*29*). Therefore, the neglect, to date, of this Bose term in thermodynamic considerations should not have introduced significant error.

One of the interesting features of the thermal excitations of the degenerate Fermi liquid is that with increase in density of the liquid the effective mass m^* should increase; this would mean that the specific heat and entropy at constant temperature should increase with increasing pressure. This behavior was first tentatively predicted from the measurements of susceptibility under pressure (*34*), which showed that at constant temperature the susceptibility became closer to its Curie value as the pressure was increased, and in 1957 the theory of Brueckner and Gammel (*31*) predicted this effect quantitatively. Recently measurements have been made on the specific heat of liquid He^3 as a function of pressure by Brewer, Daunt, and Sreedhar (*29*), and our results, together with measurements of the expansion coefficient by Brewer and Daunt (*43*), have been used to compute the entropy of the liquid as a function of T and p. Experimental evaluation of the expansion coefficients was necessary in order to compute entropies of compression. It is unnecessary to give here in detail the methods of computation. The results are presented in Fig. 6. It may been seen that in the low-temperature region the entropy does in fact increase with increasing pressure. Only at a very much higher temperature does the more normal negative sign of $(\partial S/\partial p)_T$ become evident. At the lowest temperature where the entropy is a linear function of temperature, this pressure effect can be conveniently described in terms of the effective mass m^*, and Fig. 7 shows the value of m^*/m as a function of the mean distance between atoms, both according to these experimental results and as computed by Brueckner and Gammel. Since the only experimental data inserted into the theory was the interatomic potential between free atoms, the general agreement between theory and experiment is noteworthy.

For temperatures above the linear specific-heat region of the Fermi liquid,

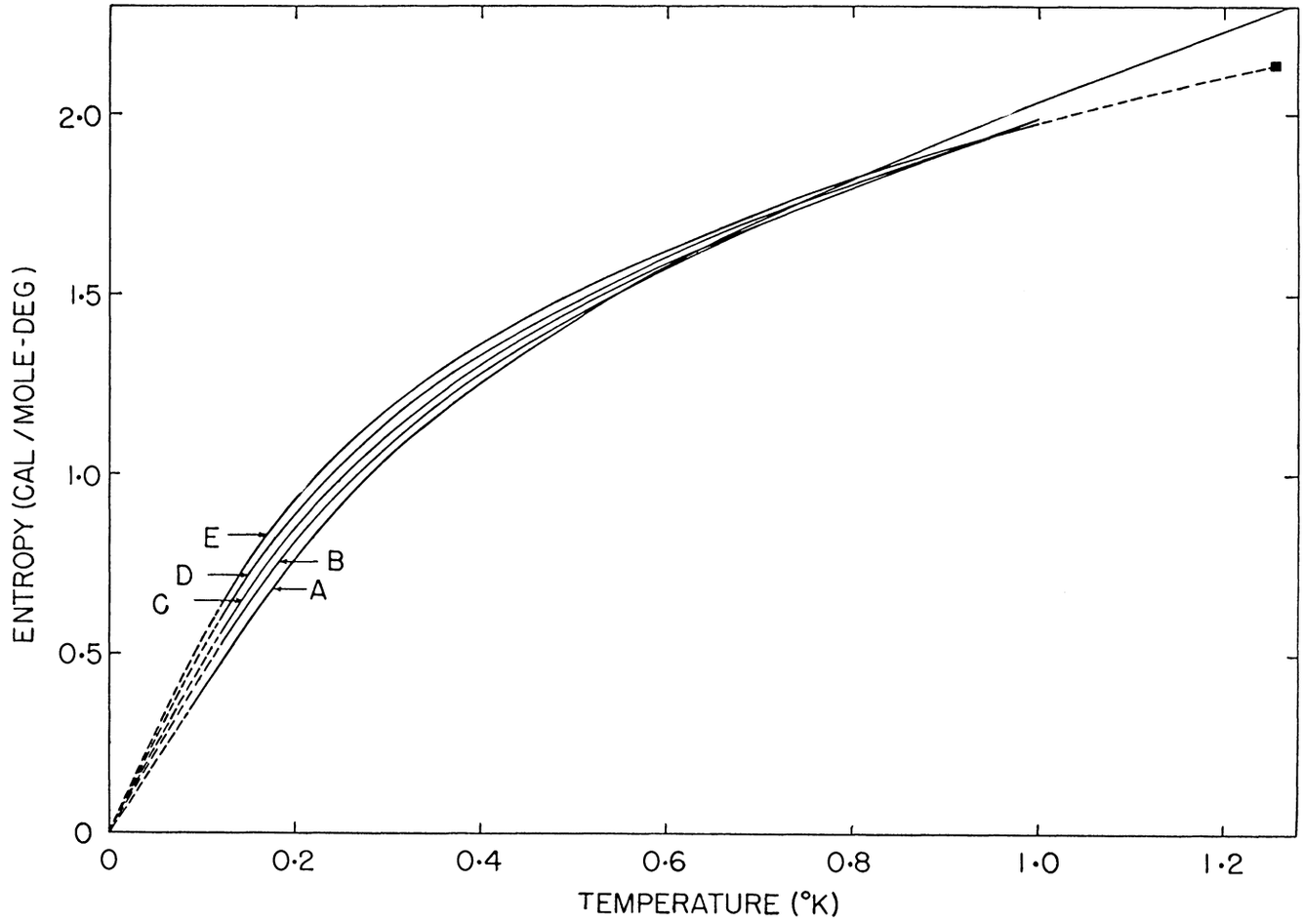
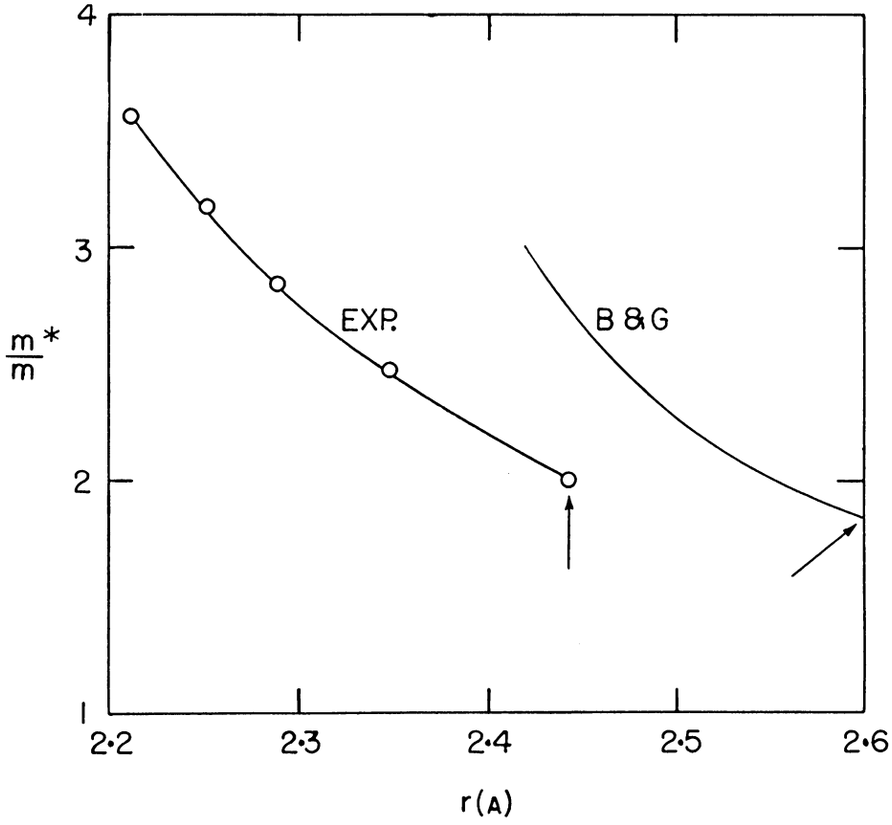
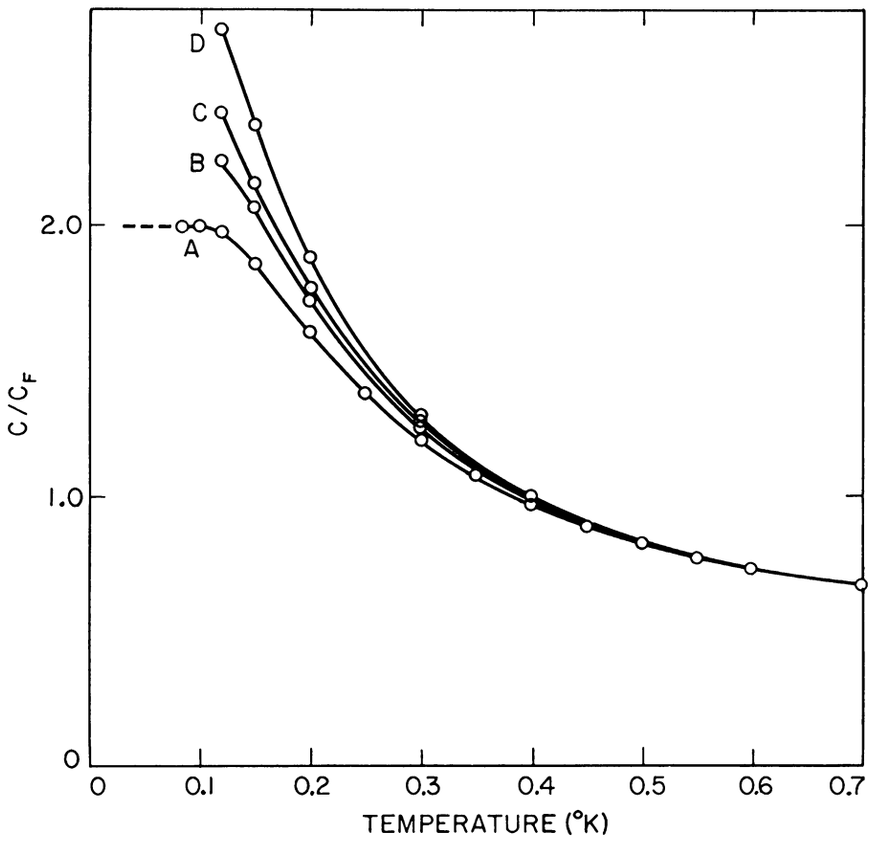


Fig. 6. Entropy of liquid He^3 as a function of temperature at various pressures [from the work of Brewer and Daunt (*43*)]. Curve *A*, at saturated vapor pressure; *B*, at 5 atmospheres; *C*, at 10 atmospheres; *D*, at 15 atmospheres; *E*, at 22 atmospheres.





the nature of the thermal excitations has not been investigated theoretically in any detail except in the work of Goldstein (*44*). The experimental results on the specific heat (*29*) in this region are presented in Fig. 8. In the figure the ratio of experimentally observed specific heat C to the specific heat of an ideal Fermi gas C_F having the same number density at each temperature and pressure is plotted as a function of temperature for a variety of different pressures. It will be seen that at the lowest temperature C/C_F increases rapidly with increasing pressure, as is evident also in Fig. 7. At higher temperatures C/C_F appears to reach values which are independent of pressure and which are less than 1. This does not necessarily mean, however, that m^* is less than 1, since terms involving $(\partial m^*/\partial T)$ may be of importance.

The broad features of liquid He^3 which have now been observed experimentally give a firm basis for regarding liquid He^3 as a degenerate Fermi liquid near 0°K, and the linear variation of the specific heat with temperature at the lowest temperatures allows us to make an interpretation concerning the thermal excitations. There remain, however, many problems requiring both experimental and theoretical investigation. From the experimental point of view it is clear that considerable effort should be made to extend the measurements of both specific heat and magnetic susceptibility to much lower temperatures, especially for the liquid under pressure. Such investigations would not only establish more precisely information which at the moment is based on extrapolation but would also explore more fully the question of the occurrence of superfluidity. Although the evidence that exists today on the superfluidity question points strongly to the assumption of complete absence of superfluidity in this rare isotope of helium, a firmer experimental conclusion on this is desirable. Experimental information on

Fig. 7 (top). Variation of m^*/m at 0°K with interatomic distance for liquid He^3. *B & G*, theory of Brueckner and Gammel (*31*); *Exp*, experiments of Brewer and Daunt (*43*). The arrows indicate the points corresponding to the saturated vapor pressure. Fig. 8 (bottom). The ratio C/C_F versus temperature, where C is the specific heat of liquid He^3 [from the work of Brewer, Daunt, and Sreedhar (see *29*)] and C_F is the specific heat of an ideal Fermi-Dirac gas of the same density as the liquid. Curve *A*, p = 6 to 14 cm-Hg; curve *B*, p = 5 atm; curve *C*, p = 10 atm; curve *D*, p = 25 atm.

other properties is necesary to provide a more detailed picture of the liquid. Measurements of the viscosity and thermal conductivity at very low temperatures would provide useful information. The predicted unusual behavior of sound propagation in the liquid at very low temperatures is of particular interest, and measurements of high-frequency sound propagation and attenuation at extremely low temperatures would be of great value, together with possible experiments on scattering of light. The fact that many of the critical experiments involve measurements at temperatures reached only by magnetic cooling provides a technical challenge which is being taken up today in many institutions specializing in low-temperature physics.

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Oxygen Transport through Hemoglobin Solutions

How does the presence of hemoglobin in a wet membrane mediate an eightfold increase in oxygen passage?

P. F. Scholander

Evolution from single cells to organisms is linked intimately with the development of a circulatory system. Without this, both size and activity would be severely limited by the slowness of diffusion. But even with a circulatory system, oxygen transport would be hampered by still another "unfitness of the environment"—namely, the very low solubility of oxygen in water. This difficulty was overcome by the evolution of oxygen-carrying pigments, which when circulated to the tissues could carry many times more oxygen than can water alone.

Oxygen-carrying pigments appeared not only in blood, however, but also in a vast area where visible means for transport of the pigment is lacking—namely, as myoglobin in the muscle system. Here it is found within the muscle cells, providing, so to speak, the last leg of the supply line to the oxygen-needy contractile machinery. But how could this myoglobin enhance oxygen transport unless it were circulated within the cell? Simple diffusion could hardly be aided by the pigment. It is true enough that the increased oxygen capacity could help to smooth out a fluctuating demand, as demonstrated by Millikan (*1*), but could it possibly also be that the pigment might

The author is professor of physiology at Scripps Institution of Oceanography, University of California, La Jolla.