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THERMAL AND MAGNETIC PROPERTIES OF DILUTE SOLUTIONS OF He^3 IN He^4 AT LOW TEMPERATURES*

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The prediction by Edwards *et al.*[1] that there should be no phase separation down to $T = 0$°K in dilute solutions of He^3 in He^4 for concentrations of less than about 6% He^3 was confirmed at least in part through heat-capacity measurements by Anderson, Roach, Sarwinski, and Wheatley[2] for a concentration of about 5% down to a temperature of nearly 10 mdeg K. The idea[3] that the He^3 quasiparticles in the He^4 at low temperatures constituted a weakly interacting Fermi fluid was also confirmed in these experiments. This Letter describes the results of a set of experiments at saturated vapor pressure on the thermal and magnetic properties of two dilute solutions of He^3 in He^4. It shows that the He^3 in dilute solutions does indeed have several of the properties associated with a normal Fermi fluid. Dilute solutions of He^3 in He^4 are particularly advantageous in studying the weakly interacting Fermi fluid since the Fermi momentum may be varied by changing concentration. Hence the momentum dependence of the quasiparticle interactions may be tested. Moreover, the present experiments form a quantitative basis for theories of the quasiparticle interactions and for predictions of a low-temperature cooperative state.[4]

In the present work the earlier heat-capacity measurements[1,2] have been confirmed and extended to lower temperatures, and measurements have also been made of the spin-diffusion coefficient and nuclear susceptibility of the *same* two dilute solutions, of nominal concentrations 1.3 and 5.0%. It is important to measure both heat capacity and magnetic properties on the same concentration since parameters such as effective mass and Fermi temperature determined by the former are used in interpreting the latter measurements.

Only a few brief remarks can be given here on the experimental method. The refrigeration problem and methods used for thermal and magnetic measurements are given by Abel, Anderson, Black, and Wheatley.[5] The method of thermal isolation while using superfluid He^4 is discussed by Vilches and Wheatley.[6] We had one important experimental difficulty. We normally cool He^3 by means of powdered cerium magnesium nitrate (CMN) which passes through an NBS 40 sieve (particle size less than 0.42 mm). CMN power prepared in this way cools pure He^3 anomalously well.[7] In the present work it was necessary to pass the CMN through an NBS 400 sieve (particle size less than 37 μ) to obtain an adequately low thermal time constant at low temperatures. The background heat capacity for the thermal measurements was obtained with pure He^4 in the cell. Above about 5 mdeg K the resulting molar heat capacity (attributed entirely to the CMN) was the same for different powder sizes (varying over a factor of 40 in particle size) and also the same as determined by Abel, Anderson, Black, and Wheatley[8] by a difference method in which He^3 provided thermal contact within the cell. The latter measurements of CMN heat capacity were used in the final data analysis since they lead to the most consistent treatment of the very low-temperature heat-capacity data. The temperature scale used here is the magnetic temperature scale valid for powered CMN in the shape of a right circular cylinder with diameter equal to height. At least above 2 mdeg K this scale is independent of particle size for sizes less than those which pass through an NBS 40 sieve.

Results of the heat-capacity measurements are shown in Fig. 1, where heat capacity per mole of solution has been divided by R, the gas constant, to render it dimensionless, and by T^* to remove most of the temperature dependence at low temperatures. These data have been fitted to the heat capacity calculated by Stoner[9] and McDougall and Stoner[10] for the ideal Fermi-Dirac gas by means of two parameters, x and T_F, where $T_F = p_F^2/2m^*k$ is the Fermi temperature, p_F is the Fermi momentum, m^* is the effective mass, k is Boltzmann's constant, and $x \equiv N_3/(N_3+N_4)$ is the He^3 concentration. The concentration x is not experimentally determined with precision, but the values of x determined by curve fitting agree with the measurements within their experimental error of about 5%. The scatter in heat capacity at low temperatures is reasonable when one recalls that the plotted points are obtained as the difference between the measured heat capacity and a rather large background heat capacity due to the CMN. For purposes of comparison, the low-temperature heat-capacity data[8] for pure He^3 at 0.28 atm are also plotted on Fig. 1.

After the heat-capacity measurements were completed, the cell was modified for magnetic measurements, which were made using the method of spin echoes at a frequency of about 100 kc/sec. The results of the measurements are shown in Figs. 2 and 3. The self-diffusion coefficient D was determined from the damp-

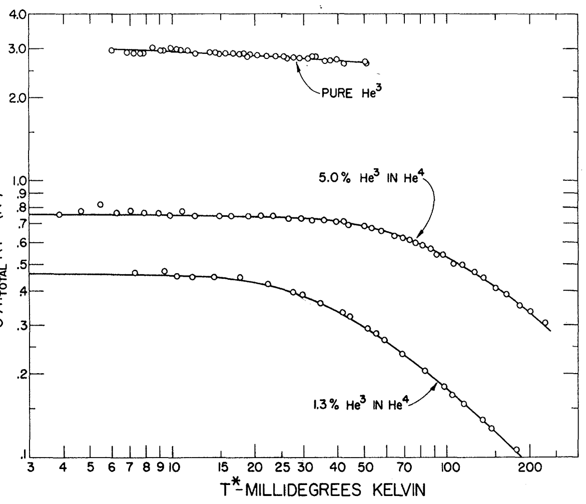

FIG. 1. Ratio of heat capacity per mole of solution to RT^*, where R is the gas constant and T^* is the magnetic temperature, for the dilute solutions of He^3 in He^4 of nominal concentration 1.3 and 5.0% He^3. Lines through the experimental data are theoretical ones based on the heat capacity of an ideal gas of Fermi-Dirac particles as computed in Refs. 9 and 10. For the nominal 1.3% concentration the theoretical line was drawn for the parameters $x = 0.0132$, $m^*/m = 2.38$, $T_F = 0.141$°K. For the nominal 5.0% concentration these parameters are $x = 0.0502$, $m^*/m = 2.45$, $T_F = 0.331$°K. The molar volumes determined by Kerr[15] were used in analyzing the data. Data from Ref. 8 for the heat capacity of pure He^3 at 0.28 atm are shown for comparison.

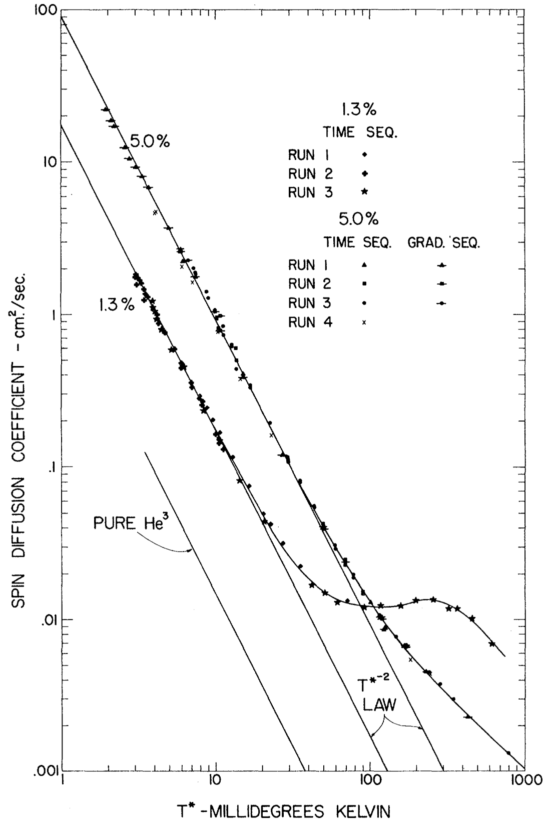

FIG. 2. Spin-diffusion coefficient for dilute solutions of He^3 in He^4 of nominal concentrations 1.3 and 5.0% He^3. The smoothed curve of the spin-diffusion coefficient for pure He^3 at 0.28 atm is shown for comparison.

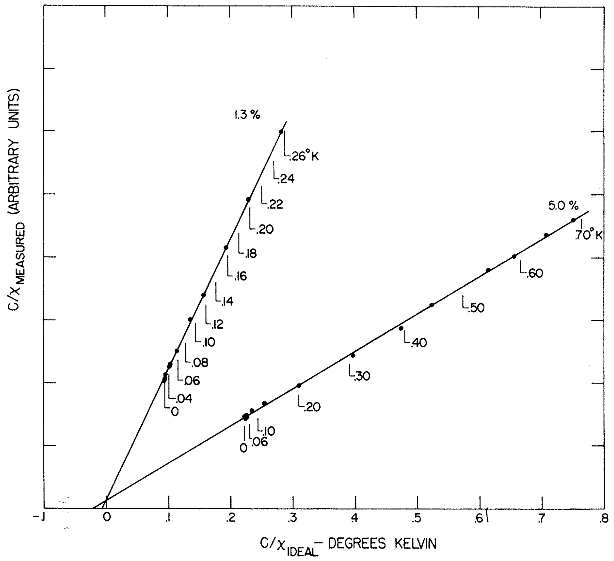

FIG. 3. A quantity proportional to C/χ_{meas} for dilute solutions of He^3 in He^4 of nominal concentrations 1.3 and 5.0% He^3 plotted against the ratio C/χ_{ideal}, where C is the Curie constant and χ_{ideal} is the susceptibility of an ideal Fermi-Dirac gas having the single Fermi temperature T_F determined by the heat capacity measurements. At low temperatures $C/\chi_{\text{ideal}} \to \frac{2}{3}T_F$, while at high temperatures $C/\chi_{\text{ideal}} \to T$. A Kelvin temperature scale is given along each curve to facilitate a comparison of these data with those on the heat capacity and self-diffusion coefficient.

ing of spin echoes in a field gradient G by fitting the data to the formula $\ln(h_1/h_2) = [(\gamma^2 G^2 D t^3/12) + t/T_2]$, where γ is the gyromagnetic ratio and t is the time between two successive echoes of heights h_1 and h_2. Corrections were made for the effect of unwanted field gradients, determined by reversing G, and for the width of the 180° pulse. Data were obtained both from sequences of photographs (at a constant temperature) in which G was varied at fixed t (gradient sequences) and from sequences in which t was varied at fixed G (time sequences). At low temperatures we experienced some difficulties in the intercomparison of these types of data. Moreover the origin of the T_2-type damping, which was considerable, is obscure. For most of the data the lowest magnetic temperature was about 3 mdeg K. An attempt was made to lower the temperature of the measurements further by a method in which the field at the CMN was held nearly at zero for most of the time by an auxiliary coil, which was turned off rapidly when a diffusion measurement was made. The method was based on the assumption of a short T_1 (less than 1 sec) for the He^3 and an assumed long (several hundred sec) thermal relaxation time. The method worked for one run on the 5.0% solution but failed later, possibly due to a shortening of the thermal relaxation time during the progress of the experiment. The results of the successful run, in which some rather large diffusion coefficients were measured, are shown in Fig. 2.

The values of D fit T^{*-2} laws within experimental scatter below about 30 mdeg K for the 5.0% solution and below about 10 mdeg K for the 1.3% solution. For comparison, the diffusion coefficient[11] for pure He^3 at 0.28 atm is also shown on Fig. 2. These data can be analyzed[11,12] to find a relaxation time for diffusion τ_D by means of the Fermi liquid expression

$$D = \tfrac{1}{3} v_F^2 \tau_D (1 + \tfrac{1}{4} Z_0), \tag{1}$$

where $v_F = p_F/m^*$ is the Fermi velocity and Z_0 is an average spin-dependent Fermi liquid parameter. On the basis of Eq. (1), the nature of the results in Fig. 2 is qualitatively reasonable. At low temperatures one expects $D_{1.3\,\%} < D_{5.0\,\%}$ since v_F^2 varies approximately as $x^{2/3}$. At higher temperatures one expects $D_{1.3\,\%} > D_{5.0\,\%}$, for in this temperature region (assumed still low enough that He^3-roton and He^3-phonon scattering is unimportant) the He^3 quasiparticles should behave like a classical gas. At all concentrations they are roughly equally excited in k space while the probability per unit time for scattering is proportional to x^{-1}. Above about 0.4°K and below about 1°K one does find in fact that xD depends approximately on temperature only. The present data at 0.5°K give $xD = (1.1 \pm 0.1) \times 10^{-4}$ cm^2/sec, in reasonable agreement with earlier data.[13] The increase of D with decreasing T in this range is no doubt due to the momentum dependence of the interactions.

The data for the nuclear susceptibility χ shown on Fig. 3 very nearly obey the ideal Fermi-Dirac gas law, numerical values being computed from the tables given in Ref. 10 for values of T_F determined from the heat-capacity measurements. As shown by Husa, Edwards, and Gaines,[15] it is useful to plot C/χ_{meas} (or a quantity proportional to it) against C/χ_{ideal}, where C is the Curie constant and χ_{ideal} is the calculated susceptibility of an ideal Fermi-Dirac gas having a single Fermi temperature given by the

Table I. Summary of measured and derived parameters for two dilute solutions of He^3 in He^4 and for pure He^3. Probable errors listed in parentheses are to be used for comparison of entries in the same row. Data for pure He^3 at 0.28 atm are taken from Ref. 11.

	1.3 %	5.0 %	Pure (0.28 atm)
$T_F \equiv P_F^2/2m^*k$	0.141 ± 0.002°K	0.331 ± 0.005°K	1.62°K
$x \equiv N_3/(N_3+N_4)$	0.0132 ± 0.0001	0.0502 ± 0.0003	1
m^*/m	2.38 ± 0.04 (0.02)	2.46 ± 0.04 (0.02)	3.08
F_1	$F_{1,5.0\%} - F_{1,1.3\%} = 0.11 \pm 0.04$		6.25
$k_F \equiv P_F/\hbar$	2.04×10^7/cm	3.17×10^7/cm	7.88×10^7/cm
$v_F \equiv P_F/m^*$	1.80×10^3 cm/sec	2.71×10^3 cm/sec	5.38×10^3 cm/sec
$1 + \frac{1}{4}Z_0 \equiv (\chi_{\text{ideal}}/\chi)_{0°\text{K}}$	1.09 ± 0.03	1.08 ± 0.03	0.34
Z_0	$+0.35 \pm 0.1$	$+0.34 \pm 0.1$	-2.66
DT^2	$[17.2 \pm 1.7(0.5)] \times 10^{-6}$ cm² K°²/sec	$[90 \pm 9(4)] \times 10^{-6}$ cm² K°²/sec	$(1.52 \pm 0.15) \times 10^{-6}$ cm² K°²/sec
$\tau_D T^2 \equiv 3DT^2/v_F^2(1+Z_0/4)$	1.46×10^{-11} sec K°²	3.4×10^{-11} sec K°²	4.7×10^{-13} sec K°²
a	$-(0.65 \pm 0.2) \times 10^{-8}$ cm	$-(0.45 \pm 0.2) \times 10^{-8}$ cm	•••

results of the heat-capacity measurements. Following Huang,[14] an imperfect Fermi gas would follow a straight line on this plot as long as the de Broglie wavelength of the particles is long compared with the range of their interaction. The intercept on the horizontal axis, $(4/3\pi)k_{\rm F}a$ where a is the scattering length, is a measure of the strength of the interaction. As Fig. 3 shows, the data for the solutions are very close to such straight lines, although there are some deviations which may be outside experimental errors. The values of a, shown in Table I, from the small, negative intercepts are in agreement with the previous high-temperature measurements, and they show that the interaction is attractive. We have also compared the data with the Fermi liquid equation[11,12]

$$C/\chi_{\rm meas}(0^\circ{\rm K}) = \tfrac{2}{3}T_{\rm F}(1+\tfrac{1}{4}Z_0). \tag{2}$$

In calculating the value of Z_0 from this equation, the limiting value of $C/\chi_{\rm meas}$ at zero temperature was obtained by plotting $C/\chi_{\rm meas}$ vs T^2 and extrapolating to zero temperature. The effective value of the Curie constant was obtained from the straight lines in Fig. 3.

The results of a numerical analysis of the data are given in Table I. For comparison the corresponding values[11] for pure $\rm He^3$ at 0.28 atm are also given. There is a small dependence of effective mass on x. Some of this may arise from changes in the $\rm He^4$ background with x. However, if all the change were due to $\rm He^3$ quasiparticle interaction, one could write[11,12]

$$m^*(x)/m^*(x\to 0) = 1+\tfrac{1}{3}F_1. \tag{3}$$

The increase in the Fermi-liquid parameter F_1 as x changes from 1.3 to 5.0% would be 0.11 ± 0.04. The value of F_1 itself should not be much different. This is to be compared with $F_1 = 6.25$ for pure $\rm He^3$. The parameter Z_0, which measures the spin dependence of the interactions, is small (~0.3) and positive in the solutions measured while it is large and negative (−2.7) for pure $\rm He^3$. This reflects a weak attractive exchange energy for $\rm He^3$ quasiparticles in the dilute solutions. The low-temperature relaxation time τ_D (inverse of scattering probability per unit time) is much larger for the dilute solutions than for pure $\rm He^3$. However, τ_D is approximately 2.3 times larger for the 5.0% than for the 1.3% solution, indicating that the scattering probability for antiparallel spins is 2.3 times less when they scatter around a Fermi sphere of radius $k_{\rm F,\,5.0\%} = 3.17\times10^7\ {\rm cm}^{-1}$ than it is when they scatter around a sphere of radius $k_{\rm F,\,1.3\%} = 2.04\times10^7\ {\rm cm}^{-1}$. The conclusion that there is an average attractive interaction between $\rm He^3$ quasiparticles is also in agreement with the quantitative results of Ref. 1.

In conclusion, we find that dilute solutions of $\rm He^3$ in $\rm He^4$ offer a unique advantage for the study of a Fermi fluid under conditions where the Fermi momentum may be varied by changing the concentration. The range of solutions available seems to limit studies to the region of weak interactions. Hence, studies of dilute solutions complement the study of pure $\rm He^3$, in which the interactions are strong (compare values of F_1 and Z_0 in Table I). The transport properties allow a detailed study of the quasiparticle interactions in the dilute solutions. The spin-diffusion coefficient measures the averaged scattering probability $\langle w_D(\theta,\varphi) \times (1-\cos\theta)(1-\cos\varphi)/\cos(\theta/2)\rangle$, in which $w_D(\theta,\varphi)$ is the scattering probability for antiparallel spins, the angle θ being that between the momenta of the colliding particles and the angle φ being that between the initial and final planes of scattering. The spin-diffusion coefficient is not very sensitive to forward scattering ($\varphi = 0^\circ$) because of the factor $1-\cos\varphi$ in the above average. On the other hand, the thermal conductivity depends on the averaged scattering probability $\langle w_K(\theta,\varphi)(1-\cos\theta)/\cos(\theta/2)\rangle$, in which $w_K(\theta,\varphi)$ is an appropriately averaged scattering probability for parallel and antiparallel spins. The thermal conductivity is thus more sensitive to forward scattering. Measurements of thermal conductivity are, therefore, of first importance in further attempts to obtain a more quantitative understanding of the interaction between $\rm He^3$ quasiparticles in dilute solutions.

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INTERACTIONS BETWEEN He^3 ATOMS IN DILUTE SOLUTIONS OF He^3 IN SUPERFLUID He^4

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The magnitude, sign, and momentum dependence of the effective interaction between He^3 atoms in superfluid He^4 are derived from the experimental data. Calculations of the Fermi-liquid parameters for the normal state of He^3 in solution yield results in agreement with experiment; the temperature for a superfluid transition associated with the He^3 is estimated to be $\sim 2\times10^{-6}$ °K.

The experiments of Edwards *et al.*[1] and of Anderson *et al.*[2] indicate that there is a small attractive interaction between He^3 atoms in dilute solution in He^4 and thus there should be a superfluid transition associated with the He^3 at sufficiently low temperatures. The purpose of this Letter is to derive from the data an approximation to the magnitude, sign, and momentum dependence of the interaction and to estimate the transition temperature.

It has been suggested earlier[3] that there should be an attractive interaction arising from the exchange of He^4 phonons between two He^3 atoms in solution, and that this interaction might lead to a superfluid transition. There are additional contributions from the van der Waals interaction and the short-range repulsion between He^3 atoms, both modified by the surrounding He^4. A calculation of the net interaction from microscopic theory is difficult. Emery has made some progress in this direction and has discussed the problems involved.[4] The empirical interaction is considerably smaller than one might have estimated; Emery points out that there is considerable cancellation in each term of the perturbation expansion. The physical reason seems to be that to a first approximation, when a He^3 atom replaces a He^4 atom in the solution, the force fields are identical and it is only effects which come from a difference in zero-point motion or in statistics that can lead to an interaction.

To estimate an empirical interaction $V(r)$ from the data, we assume that it is spin and velocity independent, and so depends only on the separation r between two He^3 atoms in solution. We shall use the measurements of the phase-separation curve[1] for $T \geqslant 0.2$°K and the spin-diffusion experiments in the millidegree region[2] to determine $V_{\vec{k}}$, the spatial Fourier transform of $V(r)$.

From their phase-separation measurements, Edwards *et al.* have deduced μ_3, the chemical potential of the He^3, as a function of x, the concentration of He^3 in He^4. They plot $\mu_3'(x)$, the difference between μ_3 and μ_F, the chemical potential of a *free* Fermi gas of the same effective mass and density as the He^3 in the mixture. Apart from an exchange correction, which is relatively small at $T \geqslant 0.2$°K, V_0 is determined by the slope of the $\mu_3'(x)$ curve; thus

$$V_0 \sim (\partial\mu_3'/\partial x)/n_4 \simeq -0.1 m_4 s^2/n_4, \qquad (1)$$

where m_4, n_4, and s are the mass, number density, and sound velocity of pure He^4. The