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Spin Diffusion of Dilute He^3 - He^4 Solutions Under Pressure[†]

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The transport properties of dilute solutions of He^3 in He^4 provide information about the effective interaction between the He^3 quasiparticles. Of the transport properties the spin diffusion coefficient, which can be measured by NMR spin-echo techniques, is most sensitive to these interactions. Here we report such measurements on 4.97% and 1.15% He^3 in He^4 for pressures of 0.04, 9.2, and 20 atmospheres at temperatures between 1.5°K and 0.072°K. The effective interaction potentials at the three pressures deduced from these data are presented. From these potentials it appears that the temperature of the expected superfluid transition is highest for the lowest pressure and the smaller concentration studied.

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Most of the properties of the dilute mixtures of $\text{He}^3\text{-He}^4$ at low temperatures are characterized by the effective interaction among He^3 atoms. In order to obtain a fuller knowledge of this interaction, we have made measurements of the spin diffusion coefficient of 4.97% and 1.15% solutions of He^3 in He^4 at 0.04, 9.2, and 20.0 atmospheres, between 1.5°K and 0.072°K. Spin diffusion is selected because its mean free time relates directly to the interaction; secondly, because the quantum correction to its classical expression varies as $(T/T_F)^3$ while that for viscosity and thermal conductivity¹ varies as $(T/T_F)^{3/2}$; and thirdly, because there is no direct contribution from the He^4 background.

Figure 1 is a schematic representation of the low temperature segment of the cryostat. The refrigeration was provided by a dilution refrigerator which utilizes a capillaries-in-an-annular-space type of countercurrent heat exchanger. The mixing chamber and the sample cell were fabricated from Epibond 100 A. A needle valve was used to seal off the sample mixture at low temperatures and the pressure was generated by compressing a bellows with He^4 liquid diluted slightly with He^3 to reduce its thermal conductivity. The magnetic field was produced by a sixth-order super-conducting solenoid. The pulse burst generator which supplied the 90°-180°-180° pulse train at a resonance frequency of 301.1 KHz, is described elsewhere.² The echo signals were averaged to increase the signal-to-noise ratio with a fast digitizer and a 400-channel analyzer. Temperatures were measured with carbon resistors, which were calibrated against a powdered CMN salt pill both before and after the measurements.

In order to utilize most of the information contained in the echo, the amplitude of the echo was deduced by fitting the entire echo, obtained by

averaging several scans at each temperature, to the expected form³

$$h_i(t) = h_{oi} \frac{\sin A(t-t_1)}{A(t-t_0)}, \quad (1)$$

where h_{oi} is the desired amplitude. The parameter A is determined by the half-height of the sample cell and the total applied magnetic field gradient G , and t_0 is the time at which the maximum amplitude occurs. The diffusion coefficient D was then calculated from the expression³

$$D = \frac{12}{\gamma^2 \tau G^2} \ln \frac{h_{02}}{h_{01}} \quad (2)$$

where γ is the gyromagnetic ratio of He^3 and τ is the time interval between the two echoes. D was determined from echoes at each temperature with the applied gradient parallel and antiparallel to the uniform magnetic field. Averaging these values reduces the effect of any extraneous gradient field to second order.

The results of the measurements are displayed in Fig. 2. The overall uncertainty is about 10-11%, most of which comes from the uncertainty of the baseline and the external gradient value.

In the interpretation of data, we shall rely heavily on the work of Emery, and the equations hereafter can be found in Ref. 1. Starting from a phenomenological Hamiltonian for the dilute mixture, Emery shows that the He^3 and He^4 subsystems can be essentially decoupled by two canonical transformations. The net result is that the He^3 acquire an effective mass from

dragging the He^4 as they move about and interact with an effective potential resulting from the interaction of the "polarization" clouds around each He^3 atom. At low enough temperatures where the excitations of the He^4 can be neglected, the effective Hamiltonian of the system becomes

$$H' = \sum_{i=1}^{N_3} \frac{p_i^2}{2m_0^*} + \frac{1}{2} \sum_{i/j}^{N_3} v(r_{ij})$$

where m_0^* is the effective mass of a He^3 atom and r_{ij} is the separation of a pair of He^3 atoms. Since the He^3 quasiparticles form a dilute system, the two-body approximation is used throughout the theory.

The behavior of the spin-diffusion as a function of temperature can be divided into three regions. In the semiclassical region ($T \gg T_F$ where T_F is the Fermi temperature) the Boltzmann equation for spin diffusion reduces to the diffusion equation for a binary mixture of gases. The spin diffusion coefficient D is given by

$$\frac{Dx}{T^{3/2}} = \frac{3k_B^{3/2}}{32\hbar^2 N_0 \pi^{1/2}} v m_0^{*1/2} \left[\int_0^\infty dy e^{-y^2} \gamma^5 \sum_{\ell} (\ell+1) \sin^2(\delta_{\ell+1} - \delta_{\ell}) \right]^{-1}, \quad (3)$$

where x is the concentration, N_0 is Avogadro's number, v is molar volume, m_0^* is the effective mass of He^3 , δ_{ℓ} is the phase shift for the scattering at the appropriate relative kinetic energy $\frac{1}{2}k_B T$ and ℓ is the angular momentum.

We note that no expression for the spin diffusion coefficient in the intermediate region ($T \approx T_F$) has yet been derived. Consequently, no analysis of the present data in that region will be attempted here.

In the quantum mechanical region ($T \ll T_F$), the coefficient can be calculated within the framework of Landau Fermi liquid theory. The resultant expression is

$$D = 1/3 \frac{\chi_0}{\chi} v_F^2 \tau, \quad (4)$$

where

$$\frac{\chi}{\chi_0} = 1 + \left(\frac{m^*}{2\hbar p_F \pi} \right)^2 \int_0^{p_F} dp p [A_E(p,0) - A_0(p,0)],$$

$$\tau = \frac{\pi}{12} \tau_{\text{Op}} + \frac{3}{4} (\tau_A - \tau_{\text{Op}}),$$

$$\frac{\hbar}{\tau_A} = \frac{k_B T^2}{T_F} \left(\frac{m^* p_F}{4\pi\hbar^3} \right)^2 \int_0^{2\pi} d\varphi (1 - \cos\varphi) \int_0^{p_F} \frac{dp}{p_F} \frac{(p/p_F)^3}{(1 - p^2/p_F^2)^{1/2}} |A_E + A_0|^2,$$

$$\frac{\hbar}{\tau_{\text{Op}}} = \frac{k_B T^2}{2T_F} \left(\frac{m^* p_F}{4\pi\hbar^3} \right)^2 \int_0^{2\pi} d\varphi \int_0^{p_F} \frac{dp}{p_F} \frac{p/p_F}{(1 - p^2/p_F^2)^{1/2}} [3|A_0|^2 + |A_E|^2],$$

and

$$A_E(p, \varphi) = - \frac{4\pi\hbar^2}{m^*} \sum_{(\ell \text{ even})} (2\ell+1) P_\ell(\cos\varphi) \frac{\tan\delta_\ell(p)}{p}.$$

$A_0(p, \varphi)$ is given by a similar expression with the sum carried over odd values of ℓ . From the above equations, it is seen that in the quantum region DT^2 will be temperature independent. This behavior is indeed observed for a 5% solution when $T \lesssim 40\text{m}^\circ\text{K}$. For the present data, it is necessary to extrapolate into this region. Because of the symmetry between particles and holes, τ is an even function of T . Therefore, accounting for first order analytic corrections

$$DT^2 = \alpha + \beta T^2.$$

This relationship is well obeyed for a mixture with $T/T_F \lesssim 1/3^5$.

Our analysis proceeds as follows. The 4.97% data are extrapolated from $T \sim 0.1^\circ\text{K}$ on down to obtain α for the three pressures. Following Emery, we assume the effective potential to be an attractive square well of depth V_0 and range b with a repulsive hard core of radius r_c . In addition, r_c is taken to be $1.8 \text{ \AA}$, as determined by Emery for all pressures. Since the core is highly repulsive, it is improbable that the pressure can affect it significantly. Assuming a particular value for b , a curve of DT^2 as a function of V_0 is generated. A range of acceptable values of V_0 can be determined from this curve using the intercept value of DT^2 , α . With this value of b and the so determined values of V_0 , the semi-classical expression is used to plot a set of curves of $Dx/T^{3/2}$. For the effective mass at different pressures, we have used the slope derived from the second sound experiment,⁶ but chose to normalize the zero-pressure value to $2.34m_3$.⁷ The molar volume of mixtures is taken from the measurements of Watson *et al.*⁸ The high temperature data are then used to determine the best set of b and V_0 . In actual computation, only s and p state phase shifts have been included in the degenerate region, since the d -state impact parameter lies outside the range of force. In the semi-classical region, the first five phase shifts are used. Also in this region, the low concentration ($x < 1\%$) data of Biegelsen *et al.*⁹ have proved to be helpful in selecting b and V_0 , since they have a smaller quantum correction. The results of the analysis are tabulated in Table I.

As the pressure is increased, the effective potential appears to become deeper and to have a shorter range when the core is kept fixed. This trend is qualitatively expected, since an increase in pressure will lead to a decrease of atomic spacing and an accompanying increase in the potential energy. However, from 0.04 to 9.2 atmospheres the change in atomic spacing

is only 3%, while the corresponding change is 10% in range and 20% in depth. We believe this is due to the simplicity of the assumed shape of the potential and, as pointed out by Emery, it is more realistic to have a long range repulsion and an intermediate attractive region. The s and p state phase shifts for the three pressures are shown in Fig. 3. They are within about 10% of each other, although DT^2 differs more than 40%. The reason is that DT^2 is proportional to T_F^2/m_0^* , which accounts for most of the pressure dependence.

Since the transition temperature of the He^3 subsystem into the superfluid state is the highest when $\delta_\ell(p_F)$ is at its maximum value, our analysis shows that the more dilute mixture of 1.24% at He^3 at vapor pressure has the highest transition temperature of 10^{-8}°K . Under elevated pressure, the transition temperature is suppressed.

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TABLE I. Parameters deduced from spin diffusion measurements.

Pressure	α	β	v_o (K)	b_o (Å)	r_c (Å)
0.04	115.1 ± 10	6.64 ± 0.7	19.70	0.83	1.8
9.2	97.0 ± 9	5.61 ± 0.6	24.08	0.72	1.8
20.0	67.1 ± 6	5.55 ± 0.5	27.62	0.66	1.8

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FIGURE CAPTIONS

- Fig. 1 Low temperature segment of the cryostat and sample cell.
- Fig. 2 Experimental results for the spin diffusion coefficient as a function of temperature for 1.15 and 4.97% He^3 in He^4 .
- Fig. 3 The s- and p-state phase shifts for the best effective potential determined from spin diffusion data for 0.04, 9.2, and 20 atmospheres.

Fig. 1.

Fig. 2

Fig-3