

Spin Waves in Liquid ^3He - ^4He Mixtures

J. R. Owers-Bradley,^(a) H. Chocholacs, R. M. Mueller, Ch. Buchal, M. Kubota, and F. Pobell^(b)
Institut für Festkörperforschung, Kernforschungsanlage Jülich, D-5170 Jülich, West Germany

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Continuous-wave nuclear magnetic resonance measurements have been performed on liquid ^3He - ^4He mixtures of 5.0% (at 0, 10, and 20 bars) and 9.5% (at 10 bars) ^3He between 0.3 and 25 mK. In the collisionless regime the NMR line is observed to shift and have a maximum in width, effects attributable to spin-wave excitations in the mixture and the first direct evidence for the existence of collective excitations in the ^3He - ^4He Fermi systems. Analysis of the data yields the Fermi-liquid parameter F_1^a for the 5%- ^3He mixture.

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In 1957 Landau¹ predicted the existence of collective modes in Fermi systems in the collisionless regime, $\omega\tau > 1$. Here τ is the fermion relaxation time and ω the frequency of the wave disturbance. Silin² applied Landau's Fermi-liquid theory to discuss the oscillations of a degenerate electron fluid and found that certain excitations could propagate in a finite magnetic field with a phononlike dispersion relation. In particular spin waves belong to these oscillations with $\tau = \tau_D$, the spin diffusion time. They correspond to oscillations in the quasiparticle density in which the spin-up and spin-down Fermi spheres have opposite phase. Platzman and Wolff³ considered the interacting electron gas and were able to calculate the dynamic susceptibility incorporating the Fermi-liquid parameters F_1^a . This theory successfully accounted for the sidebands in the ESR spectrum of thin metallic slabs and also explains the resonances observed recently in the cw NMR spectrum of pure ^3He .⁴

Leggett and Rice⁵ focused their attention on spin diffusion in the collisionless regime. They modified the usual hydrodynamic spin-diffusion coefficient, taking into account the effect of the molecular field produced by the polarization of spins in the interacting Fermi liquid. They predicted that the effective diffusion coefficient, D_{eff} , as measured by pulsed NMR, should have a maximum as a function of temperature whose position depends on the initial pulse angle and the external field. This effect was subsequently observed by Corruccini *et al.*⁶ in both ^3He and ^3He - ^4He mixtures, indirectly verifying the existence of spin waves. It was also possible to estimate F_1^a from these results for ^3He but not for the mixtures. A recent reanalysis of the ^3He diffusion data, including finite-temperature corrections, has led to more reliable and model-independent values for F_1^a and the interaction param-

eter⁷

$$\lambda = 1/(1 + F_0^a) - 1/(1 + F_1^a/3). \quad (1)$$

Unfortunately, the small size of F_0^a and the inability of the diffusion-coefficient measurements to give the sign of λ have prohibited the determination of F_1^a in the mixtures. In the present work cw NMR is employed to measure the frequency and linewidth at temperatures well above and below the crossover from the hydrodynamic to the collisionless regimes, enabling both the magnitude and the sign of λ , and hence a value for F_1^a , to be determined.

The dynamic Fermi-liquid susceptibility for small k is³

$$\chi(k, \omega) = \frac{(-m^*/m)\chi_0\Omega_0}{\omega - \omega_0 + iD^*k^2}, \quad (2)$$

where χ_0 is the static susceptibility of an ideal Fermi gas, $\Omega_0 = \omega_0/(1 + F_0^a)$, and $\omega_0/2\pi$ is the Larmor frequency. The term D^*k^2 contains the complex diffusion coefficient D^* and the wave number k for the spin-wave mode. In general several modes may be excited where for ^3He these appear as a number of sharp lines in the spectrum.⁴ For a particular k the frequency of the resonance is given by the imaginary part of D^* and the linewidth, $\Delta\omega$ (half width at half maximum), is determined by its real part:

$$\omega = \omega_0 + \text{Im}(D^*)k^2, \quad \Delta\omega = \text{Re}(D^*)k^2, \quad (3)$$

where

$$D^* = \frac{\frac{1}{3}v_F^2(1 + F_0^a)\tau_D}{1 + i\omega_0\tau_D\lambda}, \quad (4)$$

and v_F is the Fermi velocity ($\sim 27 \text{ ms}^{-1}$ for a 5% mixture at $P=0$ bar).⁸ The real part of D^* turns out to be essentially the same as D_{eff} given by Leggett and Rice for the spin-echo experiment. This is the case because the Leggett-Rice effect

involves the excitation of spin-wave modes, particularly for small-angle tipping of the magnetization vector.^{6,9} We expect to see a linewidth with a maximum when $\omega_0^2 \lambda^2 \tau_D^2 = 1$,

$$\Delta\omega = \frac{\frac{1}{3}v_F^2(1+F_0^a)\tau_D k^2}{1+\omega_0^2\lambda^2\tau_D^2}. \quad (5)$$

The shift in frequency is then

$$\omega - \omega_0 = -\frac{\frac{1}{3}v_F^2(1+F_0^a)\tau_D^2\omega_0\lambda k^2}{1+\omega_0^2\lambda^2\tau_D^2}. \quad (6)$$

At high temperatures $\text{Im}(D^*)$ is small, $\omega \rightarrow \omega_0$, and D^* reduces to the usual spin-diffusion constant. Fitting the cw NMR data with these two expressions yields values for λ and k^2 if v_F , F_0^a , and τ_D are known.

In our experiment we cooled the mixtures in a silver cell (Fig. 1) with approximately 50 m² of silver-sinter surface area and a volume of 23.5 cm³ (i.e., ~1 mole mixture). The cell was thermally connected to the copper stage of our two-stage nuclear demagnetization cryostat¹⁰ via a zinc heat switch allowing heat-capacity measurements to be performed as well.¹¹ Thermometry was by pulsed Pt NMR, calibrated against the National Bureau of Standards fixed-point device, and is considered accurate to within 5%. The NMR coil (2 mm long and 2 mm in diameter) consisted of 600 turns of 25- μ m Cu wire and was suspended in the open part of the cell as indicated. ³He NMR measurements were performed in the Pt NMR field of 280 G, corresponding to a ³He resonant frequency of 925 kHz, with excitation levels below 300 μ V. The spectrometer consisted of a low-noise home-built oscillator weakly coupled to the NMR tank circuit. The voltage appearing across the tank circuit was amplified by a low-noise field-effect transistor input amplifier and diode detected. A pair of large Helmholtz coils was placed outside the cryostat to sharpen up the NMR lines at high temperatures, improving the accuracy of the data but not otherwise affecting the results. Details of the experimental setup as well as the specific-heat results will be published elsewhere.¹¹ Measurements were taken for ³He concentrations of 9.5% at 10 bars and 5% at 0, 10, and 20 bars in the temperature range 0.3 to 25 mK. The resultant dc voltage output versus the oscillator frequency is shown for $x = 5\%$ and $P = 0$ bar in Fig. 1. Measurements at 1% ³He concentration were hampered by a poor signal-to-noise ratio.

Our cw NMR results for the mixtures do not

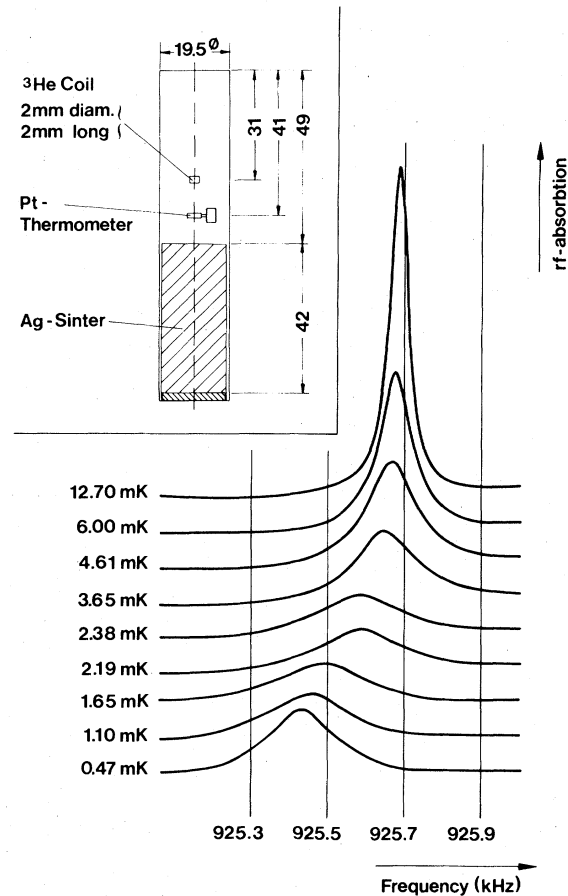


FIG. 1. The cw NMR lines for the 5% ³He-⁴He mixture at the indicated temperatures and $P = 0$ bar. Inset: Sample cell with dimensions in millimeters.

show the complex line structure observed by Scholtz for ³He.⁴ Only at the lowest temperatures and for the 9.5% mixture is there a small satellite line about 1% of the area of the main line and 23 Hz higher in frequency which rapidly broadens and disappears into the main line above 600 μ K. However, as can be clearly seen in Fig. 1 the main line shifts to lower frequency as we cool down and the width goes through a maximum. These data are plotted in Fig. 2. The sign of λ is directly related to the direction of the frequency shift, and is therefore positive. The full lines are Eqs. (5) and (6) using $F_0^a = 0.08$ and $\tau_D T^2 = 2.8 \times 10^{-11}$ s K² from Ref. 6 and v_F from Ref. 8. By fitting to the frequency data we find for the two free parameters $\lambda = +0.028 \pm 0.003$ and $k = 1.01 \pm 0.06$ mm⁻¹. The measured width is always greater than that predicted by Eq. (5) because it contains contributions from other mechanisms such as the inhomogeneities in the static field.

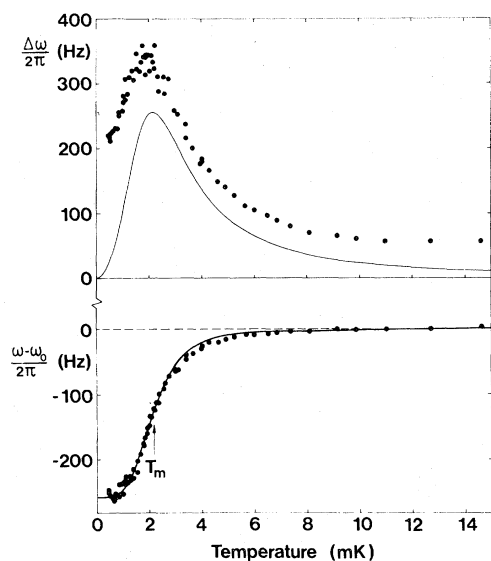


FIG. 2. The linewidth (top) and the frequency (bottom) of the NMR resonance of 5% ^3He in ^4He as a function of temperature at $P=0$ bar. The solid lines are derived from Eqs. (5) and (6) with use of the parameters given in the text.

It is not clear what determines k though we suppose that the sample-cell geometry and the rf-coil field profile are the important factors. Crudely speaking, for a mode in arbitrary geometry of characteristic dimensions d , we expect the lower limit on k to be $2\pi/d$. If we believe that the observed mode was contained only within the rf coil we would expect $k \approx 3 \text{ mm}^{-1}$. The measured $k=1.0 \text{ mm}^{-1}$ gives $d \approx 7 \text{ mm}$ which is of the order of the sample chamber size. Thus it appears that the mode is somehow locked in the whole cell and not restricted to the NMR coil. The other interesting feature is that there is no evidence at lower temperatures for a $k=0$ mode, normally the only value of k appropriate to an NMR experiment.

With the value of λ , whose magnitude agrees well with that determined by Corruccini, and taking $F_0^a = 0.08 \pm 0.03$, we find $F_1^a = 0.34 \pm 0.10$ for the 5% mixture at 0 bar. (The major source of error comes from F_0^a rather than λ .) This is in fair agreement with the prediction of Owen,¹² $F_1^a = 0.17$. Since our determination of F_1^a depends on a knowledge of τ_D and F_0^a we are only able to evaluate F_1^a for the 5%, 0-bar mixture where these quantities are known. However, we give in Table I the temperatures, T_m , where the linewidth has its maximum and the line shift its inflection point, i.e., where $\omega_0^2 \lambda^2 \tau_D^2 = 1$; the maximum linewidth; the total frequency shift; and the quantity $2 \text{Im}(D^*)T^2$ evaluated at T_m . The last quantity is calculated under the assumption that k is pressure and concentration independent. The value for 5% at 0 bar has been set equal to the DT^2 given by Murdock, Mountfield, and Corruccini.¹³ This same value was used to determine τ_D . The values for 5% at 10 and 20 bars show a pressure dependence that agrees well with that of Murdock, Mountfield, and Corruccini.¹³

Spin diffusion in the collisionless regime strongly depends on the angle φ through which the magnetization vector is tipped in a pulsed NMR experiment.^{5,6} The term $i\omega_0 \lambda \tau_D$ in the expression for D^* may be multiplied by $\cos \varphi$ to include this dependence. The analog to φ in a cw experiment is the rf excitation level. Normally in cw NMR for low levels we expect $\cos \varphi \approx 1$. To check whether we are in this regime, we have performed the experiment at a number of different rf levels in the range 7 to 200 μV and found no observable changes in the results, indicating that we were always in the limit of small φ .

Finally we comment that the NMR susceptibility for all mixtures remained constant within 10% and there were no discontinuities in the specific heat; hence to date we have no evidence for a

TABLE I. The temperature T_m at which the linewidth is greatest and at which the frequency shift has its inflection point (where $\omega_0^2 \lambda^2 \tau_D^2 = 1$), the maximum linewidth $\Delta\omega$, the total frequency shift $(\omega_{T=\infty} - \omega_{T=0})/2\pi$, and the quantity $2 \text{Im}(D^*)T^2$ for $x=5\%$ at $P=0, 10$, and 20 bars and $x=9.5\%$ at 10 bars.

x, P (bars)	T_m (mK)	$\Delta\omega/2\pi$ at T_m (Hz)	$(\omega_{T=\infty} - \omega_{T=0})/2\pi$ (Hz)	$2 \text{Im}(D^*)T^2$ at T_m ($10^{-6} \text{ cm}^2 \text{ K}^2 \text{ s}^{-1}$)
5%, 0	2.15 ± 0.1	350 ± 10	257 ± 5	75 ± 8
5%, 10	1.90 ± 0.1	290 ± 10	220 ± 10	50 ± 7
5%, 20	1.75 ± 0.1	340 ± 10	215 ± 5	42 ± 6
9.5%, 10	4.0 ± 0.5	84 ± 5	70 ± 10	70 ± 17

phase transition of the ^3He component in our mixtures under the conditions of these measurements down to 0.22 mK. Details of these experiments will be presented in a forthcoming publication.¹¹

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^(a)Present address: Physics Department, University of Nottingham, Nottingham NG7 2RD, England.

^(b)Present address: Physikalisches Institut, Universität Bayreuth, D-8580 Bayreuth, West Germany.

¹L. D. Landau, Zh. Eksp. Teor. Fiz. 30, 1058 (1956), and 33, 59 (1957) [Sov. Phys. JETP 3, 920 (1957), and 5, 101 (1957)].

²V. P. Silin, Zh. Eksp. Teor. Fiz. 35, 1243 (1958) [Sov. Phys. JETP 35, 879 (1959)].

³P. M. Platzman and P. A. Wolff, Phys. Rev. Lett. 18, 280 (1967).

⁴H. N. Scholtz, Ph.D. thesis, Ohio State University, 1981 (unpublished).

⁵A. J. Leggett and M. J. Rice, Phys. Rev. Lett. 20, 586 (1968), and 21, 506 (1968).

⁶L. R. Corruccini, D. D. Osheroff, D. M. Lee, and R. C. Richardson, J. Low Temp. Phys. 8, 229 (1972); L. R. Corruccini, Ph.D. thesis, Cornell University, 1971 (unpublished).

⁷A. Pal and P. Bhattacharyya, J. Low Temp. Phys. 51, 265 (1983).

⁸A. C. Anderson, W. R. Roach, R. E. Sarwinski, and J. C. Wheatley, Phys. Rev. Lett. 16, 263 (1968).

⁹S. Doniach, Phys. Rev. 177, 336 (1969).

¹⁰R. M. Mueller, Ch. Buchal, H. R. Folle, M. Kubota, and F. Pobell, Cryogenics 26, 395 (1980).

¹¹H. C. Chocholacs, R. M. Mueller, J. R. Owers-Bradley, Ch. Buchal, M. Kubota, and F. Pobell, to be published.

¹²J. C. Owen, Phys. Rev. Lett. 47, 586 (1981).

¹³E. S. Murdock, K. R. Mountfield, and L. R. Corruccini, J. Low Temp. Phys. 31, 581 (1978).