Observation of Spin Rotation Effects in Polarized ^{3}He-^{4}He Mixtures

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Quantum exchange effects, predicted to occur in the spin diffusion of polarized nondegenerate gases, have been observed in a ^{3}He-^{4}He mixture by spin-echo techniques. Measurements of the spin rotation parameter $(\mu M_0)^2$ and diffusion constant D are in reasonable agreement with theoretical predictions until the degeneracy temperature is approached.

The transport properties of dilute gases are determined by binary collisions. These collisions involve particle-exchange effects which are quite sensitive to the spin orientations of the atoms. Consequently large changes in the transport properties of nondegenerate gases can occur as they become spin polarized. Lhuillier and Laloe[1] (LL) have developed a spin hydrodynamic theory for these systems. Experimentally, these effects are being investigated in ^{3}He gas polarized by optical pumping techniques,[2] and spin waves associated with the phenomenon have been found in spin-aligned hydrogen.[3] In the present work modifications of the usual spin-echo[4] behavior by this effect have been found in a dilute ^{3}He-^{4}He mixture polarized to $\sim 30\%$ by the brute force use of a large magnetic field (8.9 T) and low temperatures.

Dilute mixtures were studied because the solute ^{3}He atoms form a gas of weakly interacting quasi-particles. The interaction ordinarily leads to a scattering cross section between unlike spins $\sigma \sim a_0^2$, where a_0 is the s-wave scattering length of the quasiparticles. When the liquid becomes partially polarized and particles interact with a particular relative spin orientation, a second scattering process can become important. It involves an interference between the transmitted wave and the exchange portion of the scattered wave, and has a cross section $\tau \sim \lambda a_0$, where λ is the deBroglie wavelength. The relevant parameter is the relative strength of this type of scattering, $\mu = \tau / \sigma$. It can be calculated directly from the interparticle potential, and is large when $\lambda \gg a_0$. This exchange scattering effectively reduces the transmitted amplitudes of the particles and rotates their spin directions, as if by an internal torque, about the direction of their total spin.

When incorporated into a hydrodynamic theory these effects lead to analogous terms in the spin current. This modifies the response of the mixture to the spin-echo sequence, in which an initial rf pulse tips the polarization M by an angle θ away

from the static field, an inverting rf pulse at $t/2$ reverses the dephasing of M due to the field gradient G, and a transverse polarization echo forms at time t. For the helical configuration of M appropriate in our spin-echo experiment, the spin current is

$$J_z(M_j) = -\frac{D}{1+\mu^2 M^2}\left[\frac{dM_j}{dz} + \mu\left(\vec{M} \times \frac{d\vec{M}}{dz}\right)_j\right].$$

$$(1)$$

The first term is the ordinary diffusion term, here reduced from its classical value D because of the spin rotation effect by an amount that depends upon the total polarization $M(t)$. The second term is the additional transport of longitudinal and transverse polarization in the vertical direction due to the rotation of the spins about M during collisions.

The equation for the spin evolution in the rotating frame becomes

$$\frac{\partial M_+}{\partial t} = -i\gamma G_z M_+ + \frac{D(1+i\mu M_z)}{1+\mu^2 M^2}\nabla^2 M_+.$$

$$(2)$$

This equation can be solved[5] for the echo produced in the experiment described above. If $h(t)$ is the peak amplitude of the echo relative to its amplitude at $t = 0$, and ϕ is the phase of the echo signal relative to the value of π it usually has a result of the 180° pulse, then (2) leads to

$$(1+\mu^2 M_0^2 \cos^2\theta)\ln h(t)$$
$$+ \tfrac{1}{2}(\mu M_0)^2 \sin^2\theta[h^2(t)-1]$$
$$= -\tfrac{1}{12}(\gamma G)^2 Dt^3 \qquad (3)$$

and

$$\phi = -\mu M_0 \cos\theta \ln h(t), \qquad (4)$$

where M_0 is the initial polarizatoin of the system. The phase shift is caused by the spin-rotation-

induced vertical transport of the polarization helix, which at any point will appear to be the rotation of the local direction of M_+.

The spin-echo experiment (see Fig. 1) was carried out with a 290-MHz spectrometer[6] and a static gradient of 0.26 T/m. Phase insensitive measurements of the echo amplitudes were made by mixing the signal with a reference oscillator to produce a 50-kHz i.f. signal and measuring the peak amplitude of its envelope. At each temperature measurements were made for various times and tip angles. The parameters D, μM_0, and a normalization factor were obtained by making a least-squares fit[7] of Eq. (3) simultaneously to the entire data set at that temperature. The results are shown in Fig. 2. In the absence of the spin rotation effect, the data would fall on two parallel straight lines on the plot whose slope would be related to the diffusion constant. The echo decay is curved because the diffusion extinguishes the transverse polarization which in turn suppresses the spin rotation effect and increases the rate of decay. The destruction of the polarization by diffusion is complete in the 90° case and the effect is most pronounced.

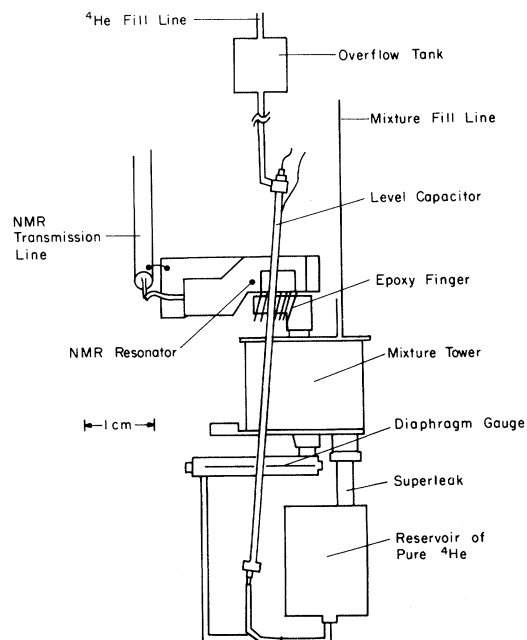The phase shift of the sample magnetization by diffusion can be measured with a phase sensitive detector. For this, the phase of the reference oscillator is set by matching it to that of the free induction decay. With this setting, the echos appear as in Fig. 3. In the tipping angle $\theta = 90°$ case, the decay in amplitude is the same as that measured with a phase insensitive detector, indicating that there is no phase shift in agreement with Eq. (4). But in the $\theta = 45°$ case the signal is seen to distort, nearly disappear, and reappear inverted because the magnetization has shifted in phase by more than $\frac{1}{4}$ of a cycle with respect to the reference signal. Eq. (4) predicts the echo to be shifted by $\frac{1}{4}$ cycle near signal C in Fig. 3, which is quite acceptable given the accuracy with which μM_0 is known. This phase shift is related to the frequency of spin waves in mixtures.[8]

The results of the fits for the spin rotation parameter $(\mu M_0)^2$ and diffusion constant D are shown in Figs. 4 and 5. We have not fit the data in Fig. 4, but provide various calculations of $(\mu M_0)^2$ for comparison. We take the Fermi gas polarization for M_0, and evaluate the cross-section ratio μ for different forms of the quasiparticle interaction. We contrast an evaluation of $(\mu M_0)^2$ in the s-wave limit with a calculation that includes the momentum dependence of the effective interaction. We have used the "effective potential" between quasiparticles,[9] and its associated s-wave scattering length $(a_0 \simeq 1.2 \text{ Å})$, as obtained by Ebner[10] from other spin diffusion data at higher concentrations, in the calculation. Increasing the interaction strength by using 1.5 Å for a_0, as suggested by Bashkin and

FIG. 1. The sample cell contained a 370-ppm mixture of ^{3}He in ^{4}He. The cerium magnesium nitrate thermometer (not shown) had to be located outside the high-field region and be connected to the cell via a wire braid and sintered exchanger. The NMR coil, thermally anchored to a surrounding shield, slipped onto the epoxy finger.
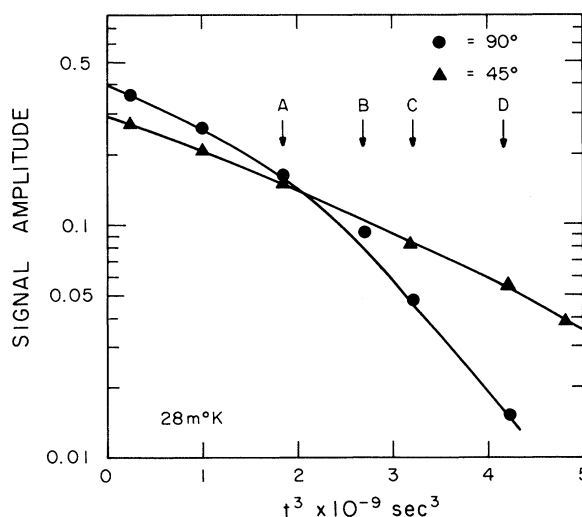
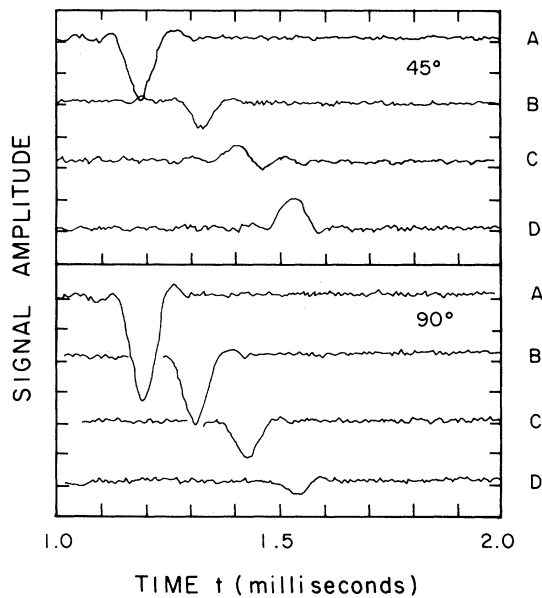FIG. 2. The NMR spin-echo amplitudes at various times and tip angles can be fit reasonably well by Eq. (3).

FIG. 3. Traces of spin echo envelopes taken at 28 mK showing the phase shift due to the spin rotation effect. The trace identification A, B, C, D refers to the time at which the echo was measured, as indicated in Fig. 2.
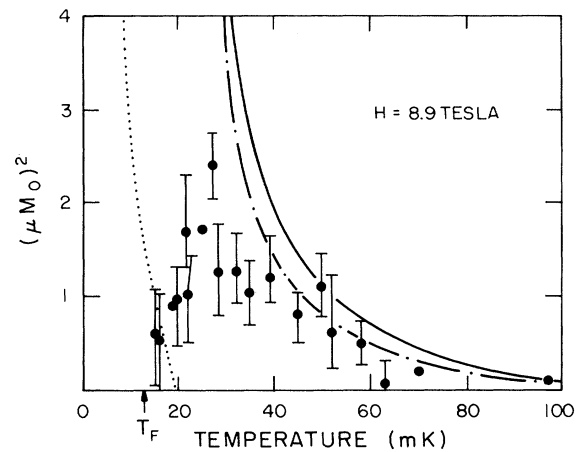

FIG. 4. The spin rotation parameter $(\mu M_0)^2$ extracted from the data. The error bars indicate the variation in $(\mu M_0)^2$ necessary to increase the χ^2 of the fit by 1.

Meyerovich,[11] lowers the curves by about a third. Although better measurements will be needed to extract details of the interaction, the results down to 25 mK have the approximate magnitude and temperature dependence expected.

Below 25 mK the drop in $(\mu M_0)^2$ is unexpected and makes our continued interpretation in the LL framework uncertain. We proceed because the fits are still good and depict the response. The LL theory, which does not include degeneracy effects, can only be expected to be valid until λ approaches the interparticle spacing, which would occur at 13 mK for our sample if it were unpolarized. Eventually the spin rotation effect evolves into the Leggett-Rice[5,12] effect when the system becomes fully degenerate.[13] If we evaluate a Leggett-Rice expression for an unpolarized degenerate Fermi liquid with an s-wave interaction we find the dotted curve in Fig. 4. We may be in a transition region between the two limiting behaviors, but a full calculation for a half-polarized, half-degenerate mixture is called for.

There is also the possibility that the apparent drop in $(\mu M_0)^2$ is the result of an extraneous process such as the heat leak. We note that data taken below 25 mK differed from those above in that the sample chamber was cooled relative to its immediate surroundings by a Pomeranchuk refrigerator,

and that a heat leak down the filling capillary did prevent cooling below 10 mK. But to change $(\mu M_0)^2$ it would have to affect the relative behavior of polarization tipped at different angles. Further the mixture's osmotic pressure and nuclear susceptibility varied properly over the same temperature interval, which argues against gross internal temperature or concentration gradients, or problems with establishing an equilibrium polarization.

The diffusion constant is given in Fig. 5. The solid curve is the expression for D given by Ebner,[10] evaluated for our concentration. The
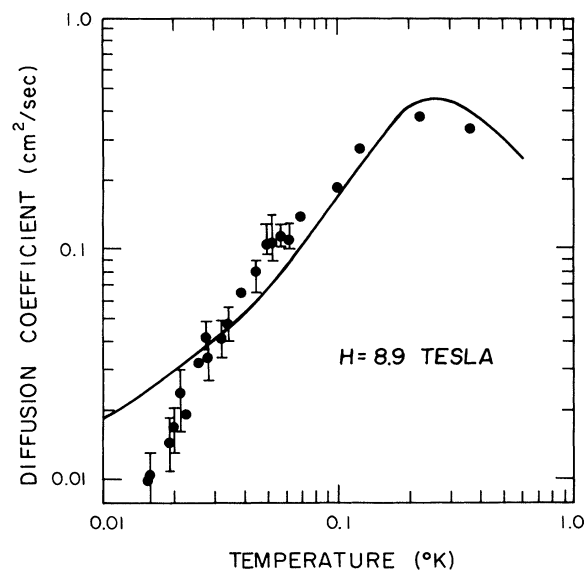

FIG. 5. The spin diffusion coefficient extracted from the data.

agreement is satisfactory at high temperatures where a peak in D occurs because the effective interaction goes through zero. Below the peak the cross section tends to a constant and D should vary as $T^{1/2}$. The data follow this trend, but seem to drop more quickly at the lowest temperatures.

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