

SPIN ECHOES IN LIQUID He<sup>3</sup> AND MIXTURES: A PREDICTED NEW EFFECT

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We deduce that the "apparent" spin-diffusion coefficient  $D_{\text{eff}}$  measured by the spin-echo technique should show a maximum as a function of temperature. The effect is estimated to lie in the experimentally attainable range for both pure He<sup>3</sup> and dilute mixtures, and should give a direct value of the Fermi-liquid parameter  $Z_1$ .

NMR at low temperatures in liquid He<sup>3</sup>, and in dilute solutions of He<sup>3</sup> in superfluid He<sup>4</sup>, has two peculiar features: The molecular field generated by exclusion-principle effects is quite comparable with the external field, and the diffusion lifetime  $\tau_D$  is very long for a liquid.<sup>1,2</sup> We shall show that these two factors should combine to produce a striking effect on the "apparent" spin-diffusion coefficient measured by the conventional spin-echo technique, provided that the initial pulse corresponds to any angle other than the usual 90°. Under this condition the magnetization gradient  $\partial\vec{M}/\partial z$ , which is in the plane (in spin space) normal to the external field owing to inhomogeneous precession, will not be parallel to the magnetization  $M$  itself, which has a component along the external field. Consequently the spin current, which is driven by the magnetization gradient, can precess in spin space around the molecular field generated by  $\vec{M}$ , as well as around the external field. This leads to a complex diffusion coefficient whose real part is measured by a (phase-insensitive) echo detector. Thus the "apparent" diffusion coefficient depends both on the initial spin polarization and on the flip angle; moreover we shall show that if these variables are held constant, it has a maximum as a function of temperature.

The idea of a complex diffusion coefficient is, of course, not entirely new; in particular, Platzman and Wolff<sup>3</sup> in their study of conduction-electron spin-resonance transmission in metals found an effect closely related to the one predicted here. However, conventional spin resonance is only a rather special case

of the much more general type of situation available in a spin-echo experiment, and their results are therefore too restricted for our purpose. We shall show that the comparison of the experimental results with the theory is very much more immediate in our case than in the one considered by them; this is due both to the absence of coupling to the orbital motion in He<sup>3</sup>, and to the fact that in a spin-echo experiment it is the natural precession of the system itself, rather than the boundary conditions, which imposes the necessary inhomogeneity of the magnetization.

On the basis of the Landau<sup>4</sup> theory of a Fermi liquid, Silin<sup>5</sup> has written down the general kinetic equation in the presence of a magnetic field [his Eq. (1.8)].<sup>6</sup> We shall assume that the fractional spin polarization is always small, in which case the quantity  $f$  in that equation can be put equal to its value in the ground state. We introduce the quantity

$$\vec{m}(\vec{n}, \vec{r}, t) \equiv (dn/d\epsilon) \int d\epsilon_p \vec{\sigma}(\vec{p}, \vec{r}, t), \quad (1)$$

where as usual  $\vec{\sigma}(\vec{p}, \vec{r}, t)$  is the spin operator of a quasiparticle of momentum  $\vec{p}$  at the point  $(\vec{r}, t)$ ,  $dn/d\epsilon$  is the density of states near the Fermi surface, and  $\vec{n}$  is a unit vector specifying direction on the Fermi surface. We assume that for what follows the external magnetic field  $\vec{H}_0(\vec{r}) \equiv \gamma^{-1} \vec{\nabla}_0(\vec{r})$  may be taken as constant in space except when it determines a precession frequency; that is, we neglect the small force terms which arise from the gradient of  $\vec{H}_0$ . After the usual algebraic manipulations, we then get the general equation of motion of  $\vec{m}(\vec{n}, \vec{r}, t)$ :

$$\frac{\partial \vec{m}}{\partial t} + \sum_{i=1}^3 v_i \frac{\partial}{\partial x_i} \{ \vec{m}(\vec{n}) + \vec{Q}(\vec{n}) \} - \vec{m} \times \vec{\nabla}_0(\vec{r}) + 4 \left( \frac{dn}{d\epsilon} \right)^{-1} \vec{m}(\vec{n}) \times \vec{Q}(\vec{n}) = \left( \frac{\partial \vec{m}}{\partial t} \right)_{\text{coll}}, \quad (2)$$

where

$$\vec{Q}(\vec{n}) \equiv \frac{1}{4}(dn/d\epsilon) \int \zeta(\vec{n} \cdot \vec{n}') \vec{m}(\vec{n}') d\Omega/4\pi, \quad (3)$$

$\zeta$  being, as usual, the spin-dependent part of the Landau quasiparticle interaction function. Equation (2) is valid for any situation in which the fractional spin polarization is small and the external field effectively constant; Eq. (2) of Ref. 3, in the limit of zero orbital coupling and rf field, is the special case obtained by linearizing around the equilibrium situation.

We look for a solution of (2) in the form (the external field is taken along the  $z$  axis)

$$\begin{aligned} m_z(\vec{n}, \vec{r}, t) &= \text{const}, \quad Q_z(\vec{n}, \vec{r}, t) = \text{const} = \frac{1}{4} Z_0 m_z^+, \\ m_x(\vec{n}, \vec{r}, t) &= -im_y(\vec{n}, \vec{r}, t) = m^+(\vec{n}, \vec{r}, t), \\ Q_x(\vec{n}, \vec{r}, t) &= -iQ_y(\vec{n}, \vec{r}, t) = Q^+(\vec{n}, \vec{r}, t), \end{aligned} \quad (4)$$

where we have introduced the conventional dimensionless parameters  $Z_l$  defined by

$$\zeta(\vec{n} \cdot \vec{n}') \equiv (dn/d\epsilon)^{-1} \sum_l Z_l P_l(\cos\theta). \quad (5)$$

We then have

$$\frac{\partial m^+}{\partial t}(\vec{n}, \vec{r}, t) + i\Omega_0(\vec{r})m^+ + \sum_{i=1}^3 v_i \frac{\partial}{\partial x_i} \{m^+(\vec{n}) + Q^+(\vec{n})\} - \left[ 4 \left( \frac{dn}{d\epsilon} \right)^{-1} m_z \left( \frac{1}{4} Z_0 m^+ - Q^+ \right) \right] = \left( \frac{\partial m^+}{\partial t} \right)_{\text{coll}}. \quad (6)$$

With fields and field gradients of the order of those normally used in spin-echo experiments, the inhomogeneity of  $m^+$  in space over a diffusion mean free path is always small, i.e., "normal-diffusion" conditions obtain. We may then look for a solution of Eq. (6) in the form

$$m^+(\vec{n}, \vec{r}, t) = A(\vec{r}, t) + B(\vec{r}, t) \vec{n} \cdot \nabla A(\vec{r}, t),$$

where  $A$  and  $B$  are slowly varying in space. Then defining the total precessing spin magnetization

$$M^+(\vec{r}, t) \equiv \int (d\Omega/4\pi) m^+(\vec{n}, \vec{r}, t)$$

and its current

$$\vec{J}^+(\vec{r}, t) \equiv \int (d\Omega/4\pi) v_F \vec{n} [m^+(\vec{n}, \vec{r}, t) + Q^+(\vec{n}, \vec{r}, t)],$$

we get (cf. footnote 6)

$$\frac{\partial M^+}{\partial t}(\vec{r}, t) + i\Omega_0(\vec{r})M^+ + \nabla \cdot \vec{J}^+(\vec{r}, t) = 0, \quad (7)$$

$$\frac{\partial \vec{J}^+}{\partial t}(\vec{r}, t) + i\Omega_0(\vec{r})\vec{J}^+ + \frac{1}{3}v_F^2 \left( 1 + \frac{1}{4}Z_0 \right) \left( 1 + \frac{1}{12}Z_0 \right) \nabla M^+ = -i\lambda K \left( 1 + \frac{1}{12}Z_1 \right) \vec{J}^+ + \left( \frac{\partial \vec{J}^+}{\partial t} \right)_{\text{coll}}, \quad (8)$$

where

$$\lambda \equiv \left( 1 + \frac{1}{4}Z_0 \right)^{-1} - \left( 1 + \frac{1}{12}Z_1 \right)^{-1}, \quad (9)$$

$$K \equiv 4 \left( 1 + \frac{1}{4}Z_0 \right) (dn/d\epsilon)^{-1} m_z^+. \quad (10)$$

Finally we assume that the collision term can be described by a relaxation time:

$$\left( \frac{\partial \vec{J}^+}{\partial t} \right)_{\text{coll}} = - \left( 1 + \frac{1}{12}Z_1 \right) \vec{J}^+ / \tau_D, \quad (11)$$

where the factor  $1 + \frac{1}{12}Z_1$  is artificially introduced so that our definition of  $\tau_D$  agrees with what has conventionally been determined<sup>7</sup> from the experimental diffusion coefficient in the

90°-180°-180° method. We assume that  $\tau_D$  is independent of  $m_z^+$ .<sup>8</sup> Then the nearly-steady-state solution of Eqs. (7) and (8) obeys the diffusion-type equation

$$\frac{\partial M^+}{\partial t}(\vec{r}, t) + i\Omega_0(\vec{r})M^+ - \tilde{D} \nabla^2 M^+ = 0 \quad (12)$$

with

$$\tilde{D}(K) \equiv \frac{\frac{1}{3}v_F^2 \left( 1 + \frac{1}{4}Z_0 \right) \tau_D}{1 + i\lambda K \tau_D}, \quad (13)$$

where  $\lambda$  and  $K$  are given by Eqs. (9) and (10).

Our result reduces to the usual one<sup>7</sup> when either  $\lambda = 0$  (no interaction between spins) or  $K = 0$  (as in a  $90^\circ$ - $180^\circ$ - $180^\circ$  experiment with long  $T_1$ ), and to that of Ref. 3 (taking into account the slightly different definition of  $\tau$  used there) when  $m_z$  has its equilibrium value in the field  $H_0$  (the usual spin-resonance situation); in that case we have  $K = \Omega_0$ .

We now apply Eqs. (12) and (13) to a spin-echo experiment.<sup>9</sup> At time  $t < 0$  we have some nearly uniform spin polarization along the  $z$  axis; since  $T_1$  in the systems considered is very long (of the order of minutes) this need not necessarily be the equilibrium polarization in the field  $\vec{H}_0$  in which the experiment is actually conducted. It is convenient to define an equivalent Larmor frequency  $\Omega_p$ , which is the Larmor frequency in the field necessary to produce the actual initial polarization under equilibrium conditions, by

$$\Omega_p \equiv 4(1 + \frac{1}{4}Z_0)(dn/d\epsilon)^{-1}m_z(t < 0). \quad (14)$$

At time  $t = 0$  we apply an rf pulse which rotates the magnetization through some angle  $\varphi$ ; thereafter,  $K$  is approximately constant at  $\Omega_p \cos\varphi$ . Since the third term in (12) is very small compared with the second,  $M^+$  is given to a good approximation, for all times  $\gg \Omega_0^{-1}$ , by

$$M^+(\mathbf{r}, t) = M^+(0) \exp[-i\Omega_0(\mathbf{r})t - \frac{1}{3}\tilde{D}\gamma^2 G^2 t^3], \quad (15)$$

where  $G \equiv |\nabla H_0|$ . When we apply a  $180^\circ$  rf pulse at time  $\frac{1}{2}t_e$ , we both reverse the phase of  $M^+$  and change  $K$  to  $-K$ . For  $t > \frac{1}{2}t_e$  the magnetization therefore obeys Eq. (12) with  $\tilde{D}^*$  instead of  $\tilde{D}$ . (The  $180^\circ$  pulse will of course affect the spin current  $J^+$  in a somewhat complicated way in the presence of molecular-field effects, but any transients so induced will die out in a time  $\tau_D$ , which is short compared with the times of interest.) Consequently the magnetization at time  $t_e$ , when an echo is expected, will be

$$M^+(\vec{\mathbf{r}}, t) = M^+(0) \times \exp[-\frac{1}{12}\tilde{D}^*(\Omega_p \cos\varphi)\gamma^2 G^2 t_e^3] \quad (16)$$

and will therefore be in phase over the whole sample apart from the negligible dephasing caused by possible weak space dependence of  $\Omega_p$  and  $G$ . A similar analysis applies to subsequent echoes. Since we assume that the echo detector is not phase sensitive, we therefore find that the "apparent" diffusion coefficient  $D_{\text{eff}}$  determined from experiment by the usual

relation  $D_{\text{eff}} = 12 \ln(h_1/h_2)/\gamma^2 G^2 (t_2 - t_1)^3$  will be given by

$$D_{\text{eff}} = \text{Re}\tilde{D}(\Omega_p \cos\varphi) = \frac{\frac{1}{3}V_F^2(1 + \frac{1}{4}Z_0)\tau_D}{1 + \lambda^2\Omega_p^2\tau_D^2\cos^2\varphi}. \quad (17)$$

We emphasize that it is  $\Omega_p$  rather than  $\Omega_0$  which enters the result; this means that, once having produced a large polarization, we are free to do the experiment in as low a field as we choose.

The most striking aspect of the result (17) is that it predicts a maximum of  $D_{\text{eff}}$  as a function of temperature if  $\Omega_p$  and  $\varphi$  are held fixed, since  $\tau_D$  is proportional to  $T^{-2}$  at low temperatures. We have estimated the temperature  $T_m$  of this maximum as a function of  $\nu_p (\equiv \Omega_p/2\pi)$  for pure  $\text{He}^3$  and for 1.3 and 5% solutions, using the experimental values<sup>1,2</sup> for  $\tau_D$  and reasonable estimates for the unknown Landau parameter  $Z_1$ , which occurs in  $\lambda$ . We find that for all three systems  $T_m$  is of the order (2-3 mdeg)  $\times \nu_p^{1/2}$ , where  $\nu_p$  is in Mc/sec.

Apart from the intrinsic interest of observing this effect, it would give a direct value of the hitherto unknown Landau parameter  $Z_1$  (note that nowhere in the derivation have we assumed that  $Z_l = 0$  for  $l \geq 2$ ). This is of some interest since various guesses and predictions have been made concerning this quantity.<sup>10</sup> In particular, in the case of dilute mixtures the theory of Bardeen, Baym, and Pines<sup>11</sup> assumes an effective interaction between  $\text{He}^3$  quasiparticles which is spin independent and weak; one would therefore predict  $\frac{1}{4}Z_1 \equiv F_1$  (the Hartree term is spherically symmetric and the Fock term couples only parallel spins), whatever the concrete form of the interaction.  $F_1$  is approximately available<sup>11</sup> from the concentration dependence of the  $\text{He}^3$  effective mass,<sup>12</sup> and so the value of  $Z_1$  should provide a rather stringent test of the theory. Appropriate experiments are currently being designed at the University of Sussex.

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<sup>1</sup>J. C. Wheatley, in *Quantum Fluids*, edited by D. F. Brewer (North-Holland Publishing Company, Amsterdam, The Netherlands, 1966), p. 206.

<sup>2</sup>A. C. Anderson, D. O. Edwards, W. R. Roach, R. E. Sarwinski, and J. C. Wheatley, Phys. Rev. Lett. **17**, 367 (1966).

<sup>3</sup>P. M. Platzman and P. A. Wolff, Phys. Rev. Letters **18**, 280 (1967).

<sup>4</sup>L. D. Landau, Zh. Eksperim. i Teor. Fiz. **30**, 1058 (1956) [translation: Soviet Phys.-JETP **3**, 920 (1957)].

<sup>5</sup>V. P. Silin, Zh. Eksperim. i Teor. Fiz. **33**, 1227 (1957) [translation: Soviet Phys.-JETP **6**, 945 (1958)].

<sup>6</sup>In what follows we put the spin relaxation times  $T_1$  and  $T_2$  equal to infinity.

<sup>7</sup>D. Hone, Phys. Rev. **121**, 669 (1961). The difference between the diffusion coefficients governing the decay of  $J_z$  and  $J^+$  entails complications which there is no

space to discuss here; Eq. (11) is simply to be regarded as a convenient definition of  $\tau_D$ .

<sup>8</sup>This is not quite obvious if  $Z_1 \neq 0$  and deserves theoretical and experimental checking. Our preliminary calculations appear to indicate that it is so.

<sup>9</sup>For details of the theory we refer to the standard treatments; provided the detector is not phase-sensitive the new features introduced here give rise to no special difficulty.

<sup>10</sup>Cf. A. J. Leggett, to be published.

<sup>11</sup>J. Bardeen, G. Baym, and D. Pines, Phys. Rev. **156**, 1, 207 (1967).

<sup>12</sup>Or in principle without any approximation from the density of the normal component at  $T=0$ .

## DEPENDENCE OF CRITICAL PROPERTIES ON DIMENSIONALITY OF SPINS

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We consider a new model Hamiltonian  $\mathcal{H}^{(\nu)}$  for interacting  $\nu$ -dimensional classical "spins";  $\mathcal{H}^{(\nu)}$  reduces to the Ising, planar, and Heisenberg models, respectively, for  $\nu=1, 2$ , and 3. Certain critical properties of  $\mathcal{H}^{(\nu)}$  are found to be monotonic functions of  $\nu$ .

Although it was first introduced as a simple model of ferromagnetism, the nearest-neighbor  $S=\frac{1}{2}$  Ising model has come to serve as a practical model for a binary alloy and a classical gas.<sup>1</sup> More recently, the classical planar model has received attention as a fairly crude lattice model for the  $\lambda$  transition in a Bose fluid.<sup>2,3</sup> The classical Heisenberg model has been proposed<sup>4</sup> as a realistic model for isotropically interacting spins at temperatures in the neighborhood of the critical temperature  $T_c$ . Finally, the spherical model<sup>5</sup> has long been attractive, especially since it is exactly soluble. The Ising, planar, and Heisenberg models are special cases (for  $\nu=1, 2$ , and 3) of

$$\mathcal{H}^{(\nu)} = -2J \sum_{\langle ij \rangle} \vec{S}_i^{(\nu)} \cdot \vec{S}_j^{(\nu)}, \quad (1)$$

where the  $\vec{S}_i^{(\nu)}$  are  $\nu$ -dimensional vectors of magnitude  $\sqrt{\nu}$ , and  $-2J\nu$  is the energy of a nearest-neighbor pair  $\langle ij \rangle$  of parallel "spins" localized on sites  $i$  and  $j$ . Here we apply high-temperature expansion methods to obtain some critical properties of the model Hamiltonian (1).

We have calculated, for general  $\nu$  and for general lattice structure, the coefficients  $\hat{a}_n^{(\nu)}$  in the expansion for the zero-field reduced

susceptibility

$$\bar{\chi}^{(\nu)} = 1 + \sum_{n=1}^{\infty} \hat{a}_n^{(\nu)} x^n \quad (2)$$

[through order  $n=8$  for close-packed and through order  $n=9$  for loose-packed lattices], and the coefficients  $\hat{c}_n^{(\nu)}$  in the specific-heat series

$$C^{(\nu)} = Nk \sum_{n=2}^{\infty} \hat{c}_n^{(\nu)} x^n \quad (3)$$

[through orders  $n=9, 10$  for close- and loose-packed lattices, respectively]. Here  $x \equiv 2J/kT$  and  $k$  is the Boltzmann constant. The coefficients were computed directly from a diagrammatic representation of the zero-field spin correlation function  $\langle \vec{S}_0 \cdot \vec{S}_R \rangle_{\beta}^{(\nu)}$  for  $\mathcal{H}^{(\nu)}$ .<sup>6</sup> The requisite diagrams<sup>7</sup> are the same regardless of the dimensionality of the spins. Moreover, certain topological similarities among the diagrams may be exploited to reduce from 298 to only 15 the number of averages or "traces" which must actually be calculated! We obtain complete agreement with previous calculations<sup>1,3,4</sup> of the coefficients  $\hat{a}_n^{(\nu)}$  and  $\hat{c}_n^{(\nu)}$  when we specialize to the cases  $\nu=1, 2$ , and 3; hence our work serves as an independent and very thorough check on these previous calculations. More-