# Spin waves and spin diffusion in Fermi liquids: Bounds on effective diffusion coefficients

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(Received 10 September 1985)

We investigate the accuracy of the usual relaxation-time approximations, involving the spindiffusion lifetime  $\tau_D$ , which are generally made in analyses of spin waves and the Leggett-Rice effect in Fermi liquids. By employing the variational methods of Ah-Sam, Højgaard-Jensen, and Smith and of Egilsson and Pethick, we are able to determine upper and lower bounds on the effective diffusion coefficient resulting from spin-wave phenomena which are accurate in the whole Fermi-liquid regime ( $T \ll T_F$ ). Our results indicate that the usual approximations break down for  $T \leq 7$  mK in <sup>3</sup>He, but are accurate to within  $\sim 2\%$  in 5% <sup>3</sup>He-<sup>4</sup>He mixtures. Our results are consistent with values of  $F_1^a \approx -0.6$  at 0 bar,  $\approx -0.4$  at 27 bars in <sup>3</sup>He, but these have considerable uncertainties.

### I. INTRODUCTION

The possible existence of propagating collective modes of transverse spin polarization in Fermi liquids was first pointed out by Silin.<sup>1</sup> These were called "spin waves" in analogy to the collective modes in ferromagnetic systems. Silin considered a homogeneous, infinite system immersed in a uniform, static magnetic field, and found the normal modes of the system by considering small perturbations from the equilibrium polarization. The frequencies of these free oscillations were determined in part by the effect of the internal "molecular" field due to quasiparticle interactions, since the local spin polarization would not simply precess about the external field, but about the net field which resulted from the internal and external fields together.<sup>2</sup> Silin found expressions for the eigenfrequencies associated with distortions of the Fermi surface of all partial waves *l*. He pointed out that propagating wavelike modes could exist which would have, in lowest order, quadratic dependence on wavenumber q.

Later, Platzman and Wolff<sup>3</sup> explicitly calculated the  $q^2$  dependence of the l=0 Silin mode—i.e., the spin-wave spectrum in the long-wavelength limit—by including the effects of spatial inhomogeneities through the drift terms in the kinetic equation. They also included the effects of collisional damping of these waves by introducing a relaxation-time approximation for the collision integral (Silin had considered the collisionless case). The eigenfrequency of the l=0 mode was shown by Silin to be equal to the Larmor frequency  $\omega_0$ —that is, it was unaffected by the molecular field terms—and so the frequencies of this branch of the spin-wave spectra are  $\approx \omega_0$  in the long-wavelength regime.

Platzman and Wolff applied their result to the problem of microwave transmission through a metal slab of thickness L by conduction-electron spin resonance, in which standing spin-wave modes of wavelength  $n\pi/L$  would be created. They were able to calculate the transmitted signal as a function of applied magnetic field and therefore of spin-wave frequency. Since the microwave energy is transported through the slab in the form of a transverse magnetization, the spectra of the collective modes of transverse magnetization determine the shape of the observed signal. These spin-wave modes were observed for the first time in alkali metals by Schultz and Dunifer.<sup>4</sup>

Platzman and Wolff presented their result in the form of an expression for the transverse susceptibility,

$$\chi(q,\omega) = \frac{(-m^*/m)\chi_0[\omega_0/(1+F_0^a)]}{\omega - \omega_0 + iD^*q^2} , \qquad (1.1)$$

where  $m^*$  is the effective mass,  $\chi_0$  is the static susceptibility for a noninteracting Fermi gas,  $F_0^a$  is the usual Landau parameter, and  $D^*$  is a complex diffusion coefficient. For neutral systems, it is given by

$$D^* = \frac{(v_F^2/3)(1+F_0^a)\tau_D}{1+i\omega_0\lambda\tau_D} , \qquad (1.2)$$

where  $v_F$  is the quasiparticle velocity at the Fermi surface,  $\tau_D$  is the spin-diffusion relaxation time, and  $\lambda \equiv (1+F_0^a)^{-1} - (1+F_1^a/3)^{-1}$  is a parameter which characterizes the strength of quasiparticle interactions. The poles of  $\chi(q,\omega)$  determine the collective modes, so  $(\omega - \omega_0) = -iD^*q^2$ . For  $\omega_0\lambda\tau_D \ll 1$  (the hightemperature regime, since  $\tau_D \sim 1/T^2$ ),  $D^*$  is purely real, and there is only diffusional broadening of the resonance at the Larmor frequency  $\omega_0$ . However, for  $\omega_0\lambda\tau_D \gg 1$  at very low temperatures— $D^*$  is purely imaginary and freely propagating collective modes with a  $q^2$  spectrum the spin waves—will be present.

Another analysis of the effects of quasiparticle interactions in the precession-dominated regime was provided by Leggett and Rice.<sup>5</sup> They considered the case of a Fermi liquid in a nonuniform external magnetic field in which the net spin polarization had been tipped away from the direction of the external field. The gradient in the magnetic field would cause the transverse component of the spin polarization to precess at different rates depending on its position. The resulting gradient in spin polariza-

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tion, they showed, would drive a (transverse) spin current which would have a net precession about the internal molecular field. Through the continuity equation relating spin density and spin current, they obtained an expression which related the time dependence of the transverse spin density to the quasiparticle interaction (embodied in the parameter  $\lambda$ ) which could then be observed in a spin-echo experiment employing pulsed NMR.

For the case of small tipping angles, Leggett and Rice showed that the effects of the quasiparticle molecular field would be observable in the form of an effective diffusion coefficient  $D_{\text{eff}}$ . This  $D_{\text{eff}}$  would cause an exponential decay in the amplitude A of successive spin echoes according to

$$A \sim \exp[(-\frac{1}{12})D_{\rm eff}\gamma^2 G^2 t_0^2], \qquad (1.3)$$

where  $\gamma$  is the gyromagnetic ratio, G is the magnetic field gradient, and  $t_0$  is the time between spin-echo signals. The effective diffusion coefficient is given by

$$D_{\rm eff} = \frac{D_{\sigma}}{1 + \omega_0^2 \lambda^2 \tau_D^2 \cos^2 \phi} , \qquad (1.4)$$

where  $\phi$  is the tipping angle and  $D_{\sigma} = [(v_F^2/3)(1+F_0^a)\tau_D]$  is the diffusion coefficient in the high-temperature regime.

It turns out that this  $D_{\text{eff}}$  differs from the real part of the  $D^*$  derived by Platzman and Wolff for spin waves only by an additional factor of  $\cos^2\phi$  in the denominator. This is perhaps not so surprising, since the same precessional effects of the quasiparticle field are the key feature. Indeed, as shown explicitly by Doniach,<sup>6</sup> the magnetic field gradient causes l=0 spin-wave modes to be excited for a whole range of wave numbers q in the longwavelength regime. Therefore, here, as before, we have the case of propagating spin-wave modes with diffusional damping.

The  $1/T^2$  dependence of  $\tau_D$  causes a maximum to occur in the temperature dependence of the effective diffusion coefficient and this clear prediction was experimentally confirmed by Corruccini, Osheroff, Lee, and Richardson<sup>7</sup> in both pure <sup>3</sup>He and <sup>3</sup>He-<sup>4</sup>He mixtures. In this first observation of spin-wave phenomena in helium, they used the Leggett-Rice expressions to obtain a value for the Landau parameter  $F_1^a$  from their data.

Actual standing spin-wave modes, characterized by particular values for q, have been observed much more recently. Observations by Owers-Bradley *et al.*<sup>8</sup> in <sup>3</sup>He-<sup>4</sup>He mixtures, and by Masuhara *et al.*<sup>9</sup> in <sup>3</sup>He have been reported in the past two years. In addition, related observations of multiple spin echoes have recently been made by Einzel *et al.*<sup>10</sup>

A common feature of all of the theoretical treatments described above is the use of a relaxation-time approximation for the collision integral in the Landau kinetic equation, which employs the spin-diffusion relaxation time  $\tau_D$ to characterize the collisional damping of the spin waves. In the high-temperature regime ( $\omega_0 \lambda \tau_D \ll 1$ ), in which normal diffusion is the dominant process, this approximation should be adequate. However, for the very-lowtemperature regime in which spin waves propagate, this is a much more questionable assumption. As  $T \rightarrow 0$ ,  $D_{Re}^* \rightarrow 0$  ( $D_{Re}^* \equiv real$  part of  $D^*$ ;  $D_{Im}^* \equiv imaginary part of <math>D^*$ ) and we have freely propagating, undamped spin waves. As T increases and collisional effects can no longer be neglected, diffusional damping of the collective modes begins to become significant. However, as discussed above, the physical process underlying the diffusion in the low-temperature regime is quite different from that at high T. When the collisional relaxation time is much longer than the time required for the spins to precess about the local effective (external plus internal) magnetic field, the strong precessional effects on the transverse spin current will be the determining influence on the nature of the spin diffusion. One would not expect this different physical process to be characterized by the same relaxation time as in ordinary diffusion.

This problem was recently examined by Pal and Bhattacharyya,<sup>11</sup> who analyzed the Leggett-Rice effect by using an approximation for the form of the collision integral. This was in effect an approximate interpolation between an exact low-temperature expression and an approximate high-temperature one, which they used to reanalyze the data of the experiment of Corruccini *et al.* 

Our approach here will be to use the variational methods which have been applied to the study of the Landau kinetic equation to derive exact limiting expressions for the effective diffusion coefficients which are associated with spin-wave phenomena in Fermi liquids. The limitation will be that we are only able to obtain upper and lower bounds, which however will be valid over the whole Fermi-liquid regime ( $T \ll T_F$ ). The methods we use have been developed and discussed in detail by Højgaard-Jensen, Smith, and Wilkins,<sup>12</sup> Ah-Sam, Højgaard-Jensen, and Smith,<sup>13</sup> and Egilsson and Pethick.<sup>14</sup> In particular, Egilsson and Pethick used these methods to treat the closely analogous problem of the transition from zero sound to first sound in Fermi liquids.

The rest of this paper is organized as follows. Section II discusses the case of spin waves, and is analogous to the treatment of Platzman and Wolff. We use the method of Ah-Sam et al. to calculate bounds on the effective diffusion coefficient in the long-wavelength limit, which may then be compared to the results of Platzman and Wolff. Section III discusses the Leggett-Rice effect in the context of the spin-echo experiment performed by Corruccini et al. We use the same method as before to calculate the bounds on the real part of the effective diffusion coefficient, which is measured in that experiment. These bounds may then be compared to the expression of Leggett and Rice. In Sec. IV we carry out the comparison of our bounds to the results derived by Platzman and Wolff and Leggett and Rice, and then reanalyze the available experimental data to make several tentative statements about some of the parameters in normal <sup>3</sup>He and <sup>3</sup>He-<sup>4</sup>He mixtures. Section V is a short summary. Appendix A outlines the reduction of the collision integral to a form used in the method of Ah-Sam et al., while Appendix B summarizes that method.

### **II. SPIN WAVES**

We start from the kinetic equation for the spin density in the case where the axis of net spin polarization is located in an arbitrary direction. This may be written as<sup>15</sup>

$$\frac{\partial \sigma_{\mathbf{p}}}{\partial t} + \frac{\partial}{\partial r_{i}} \left[ \frac{\partial \epsilon_{\mathbf{p}}}{\partial p_{i}} \sigma_{\mathbf{p}} + \frac{\partial \mathbf{h}_{\mathbf{p}}}{\partial p_{i}} n_{\mathbf{p}} \right] \\ + \frac{\partial}{\partial p_{i}} \left[ \frac{-\partial \epsilon_{\mathbf{p}}}{\partial r_{i}} \sigma_{\mathbf{p}} - \frac{\partial \mathbf{h}_{\mathbf{p}}}{\partial r_{i}} n_{\mathbf{p}} \right] \\ = \left[ \frac{\partial \sigma_{\mathbf{p}}}{\partial t} \right]_{\text{precession}} + \left[ \frac{\partial \sigma_{\mathbf{p}}}{\partial t} \right]_{\text{collision}}, \quad (2.1)$$

where

$$\mathbf{h}_{\mathbf{p}} = \frac{-\gamma \hbar}{2} \mathbf{H} + 2 \int \frac{d^3 p'}{(2\pi\hbar)^3} f^a_{\mathbf{p}\mathbf{p}'} \boldsymbol{\sigma}_{\mathbf{p}'}$$
(2.2)

describes the local effective magnetic field, and includes coupling to both the external field and the effective magnetic field produced by quasiparticle interactions. (Here,  $\gamma$  is the gyromagnetic ratio; the density of polarized spins is considered to be a small fraction of the total density of particles, so spin polarization effects on  $f_{pp'}^a$  are ignored.)

We want to focus on the behavior of small perturbations of the spin polarization from an equilibrium polarization  $\sigma_p^0$  in the direction of the external field. We define  $\sigma_p = \sigma_p^0 + \delta \sigma_p$  and proceed to linearize the equation, dropping higher-order terms, to obtain

$$\frac{\partial \delta \boldsymbol{\sigma}_{\mathbf{p}}}{\partial t} + \mathbf{v}_{\mathbf{p}} \cdot \boldsymbol{\nabla}_{\mathbf{r}} \left[ \delta \boldsymbol{\sigma}_{\mathbf{p}} - \frac{\partial n_{p}^{0}}{\partial \boldsymbol{\epsilon}_{p}} \left[ \frac{-\gamma \hbar}{2} \delta \mathbf{H} + 2 \int \frac{d^{3} p'}{(2\pi\hbar)^{3}} f_{\mathbf{pp}'}^{a} \, \delta \boldsymbol{\sigma}_{\mathbf{p}'} \right] \right] = \left[ \frac{\partial \boldsymbol{\sigma}_{\mathbf{p}}}{\partial t} \right]_{\text{precession}} + \left[ \frac{\partial \boldsymbol{\sigma}_{\mathbf{p}}}{\partial t} \right]_{\text{collision}}, \quad (2.3)$$

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where  $(\partial n_p^0 / \partial \epsilon_p)$  refers to the equilibrium functions. The precessional term is given (before linearization) by

$$\left[\frac{\partial \boldsymbol{\sigma}_{\mathbf{p}}}{\partial t}\right]_{\text{precession}} = \frac{-2}{\hbar} \boldsymbol{\sigma}_{\mathbf{p}} \times \mathbf{h}_{\mathbf{p}}$$
$$= \gamma \boldsymbol{\sigma}_{\mathbf{p}} \times \mathbf{H} - \frac{4}{\hbar} \int \frac{d^{3} \boldsymbol{p}'}{(2\pi\hbar)^{3}} f^{a}_{\mathbf{p}\mathbf{p}'}(\boldsymbol{\sigma}_{\mathbf{p}} \times \boldsymbol{\sigma}_{\mathbf{p}'}) , \qquad (2.4)$$

since one must include precession about the local effective field, as well as about the external field. The collision term will be equal to  $I[\overline{\sigma}_p]$ , the collision integral, where the overbar indicates the local equilibrium values.

We proceed to linearize the precession term, where we make use of the fact that

$$\boldsymbol{\sigma}_{\mathbf{p}}^{0} = \frac{-\partial \boldsymbol{n}_{p}^{0}}{\partial \boldsymbol{\epsilon}_{p}} \frac{\boldsymbol{\sigma}^{0}}{N(0)} , \qquad (2.5)$$

where  $\sigma^0$  is the total equilibrium magnetization and N(0) is the density of states at the Fermi surface. We take the transverse component of these equations in order to study the behavior of the transverse spin polarization  $\delta\sigma_p^+ \equiv (\delta\sigma_p)_x + i(\delta\sigma_p)_y$ . (There is an analogous equation for  $\delta\sigma_p^-$ .) Then we define

$$\delta\sigma_{\mathbf{p}}^{+} = \frac{-\partial n_{p}^{0}}{\partial\epsilon_{p}} v_{\mathbf{p}}^{+} , \qquad (2.6)$$

and we obtain

$$\frac{\partial \mathbf{v}_{\mathbf{p}}^{+}}{\partial t} + \mathbf{v}_{\mathbf{p}} \cdot \nabla_{\mathbf{r}} \left[ \mathbf{v}_{\mathbf{p}}^{+} + \frac{N(0)}{2} \left[ -\gamma \hbar \delta H^{+} + 2\int \frac{d\Omega'}{4\pi} f^{a}_{\mathbf{pp}'} \mathbf{v}_{\mathbf{p}'}^{+} \right] \right] \\ + i \left[ \left[ \omega_{0} - \frac{2}{\hbar} f^{a}_{0} \sigma^{0} \right] \mathbf{v}_{\mathbf{p}}^{+} + \frac{2}{\hbar} \sigma^{0} \int \frac{d\Omega'}{4\pi} f^{a}_{\mathbf{pp}'} \mathbf{v}_{\mathbf{p}'}^{+} \right] \\ = I \left[ \overline{\mathbf{v}}_{\mathbf{p}}^{+} \right]. \quad (2.7)$$

Here,  $f^{a}_{pp'}$  is evaluated at the Fermi surface. We now write

$$v_{\rm p}(\mathbf{r},t) = \int d^3q \, d\omega v_{\rm p}(\mathbf{q},\omega) e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)}$$

and we obtain (where we take  $\delta H \rightarrow 0$ , since we are looking for the free oscillations of the system)

$$\left[\omega - \mathbf{v}_{\mathbf{p}} \cdot \mathbf{q} - \omega_{0} + \frac{2}{\hbar} f_{0}^{a} \sigma^{0} \right] v_{\mathbf{p}}^{+} - \left[\frac{2}{\hbar} \sigma^{0} + N(0) \mathbf{v}_{\mathbf{p}} \cdot \mathbf{q}\right] \left[\int \frac{d\Omega'}{4\pi} f_{\mathbf{pp}'}^{a} v_{\mathbf{p}'}^{+}\right] = iI[\bar{\mathbf{v}}_{\mathbf{p}}^{+}] .$$

$$(2.8)$$

We then expand  $v_p^+$  in a series of spherical harmonics, so

$$v_{\mathbf{p}}^{+} = \sum_{n,m'} A_{nm'}(\epsilon) Y_{nm'}(\theta,\phi) . \qquad (2.9)$$

We make the usual expansion of the Landau parameters and express it in terms of spherical harmonics

$$f_{\mathbf{pp}'}^{a} = \sum_{l} f_{l}^{a} P_{l}(\chi_{\mathbf{pp}'}) = \sum_{l,m} f_{l}^{a} Y_{lm}(\theta, \phi) Y_{lm}^{*}(\theta', \phi') \frac{4\pi}{2l+1} ,$$
(2.10)

where we use  $\mathbf{p}(\theta, \phi) \equiv \mathbf{p}(\Omega)$ , and  $\mathbf{p}'(\theta', \phi') \equiv \mathbf{p}'(\Omega')$ ; here the  $\hat{\mathbf{z}}$  axis is defined by the direction of the external magnetic field. Then we get

$$\int \frac{d\Omega'}{4\pi} f^{a}_{pp'} v^{+}_{p'} = \sum_{\substack{l,m, \\ n,m'}} \frac{1}{2l+1} A_{nm'} Y_{lm}(\theta,\phi) \delta_{ln} \delta_{mm'}$$
$$= \sum_{l,m} \frac{f^{a}_{l}}{2l+1} A_{lm} Y_{lm}(\theta,\phi) . \qquad (2.11)$$

Now the factor  $\mathbf{v}_{\mathbf{p}} \cdot \mathbf{q}$  will bring in another angle, and so we may write the equation as follows:

$$\sum_{l,m} \left[ \omega - \omega_0 + \frac{2}{\hbar} f_0^a \sigma^0 - \frac{2\sigma^0}{\hbar} \frac{f_l^a}{2l+1} \right] A_{lm} Y_{lm}(\theta, \phi)$$
$$- (\mathbf{v_p} \cdot \mathbf{q}) \sum_{l,m} \left[ 1 + \frac{F_l^a}{2l+1} \right] A_{lm} Y_{lm}(\theta, \phi)$$
$$= \sum_{n,s} i I_{ns} Y_{ns}(\theta, \phi) . \quad (2.12)$$

Here we are defining

$$I[\bar{\nu}_{\rm p}^+] = \sum_{n,s} I_{ns}[A_{ns}(\epsilon)]Y_{ns}(\theta,\phi) , \qquad (2.13)$$

where

$$\overline{v}_{p}^{+} = \sum_{n,s} \overline{A}_{ns}(\epsilon) Y_{ns}(\theta, \phi)$$
$$= \sum_{n,s} \left[ 1 + \frac{F_{n}^{a}}{2n+1} \right] A_{ns}(\epsilon) Y_{ns}(\theta, \phi)$$
(2.14)

relates the "local equilibrium" spin density  $\delta \overline{\sigma}$  to the spin density  $\delta \sigma$ . Now,  $\mathbf{v_p} \cdot \mathbf{q} \rightarrow v_F q \cos(\theta_{pq})$  for excitations near the Fermi surface. We expand this in spherical harmonics, where the direction of the wave vector  $\mathbf{q}$  is defined by the angles  $\Omega'' = (\theta'', \phi'')$ . Then, after multiplying by  $Y_{l'm'}^{\bullet}(\theta, \phi)$  and integrating with respect to  $\Omega$ , we obtain

$$\Omega_{l'}A_{l'm'} - \left\{ \left[ \frac{4\pi}{3} \right] v_{F}q \left[ Y_{1,-1}^{*}(\Omega'') \sum_{l,m} \left[ 1 + \frac{F_{l}^{a}}{2l+1} \right] A_{lm} \int d\Omega \ Y_{lm}(\Omega) Y_{l'm'}^{*}(\Omega) Y_{1,-1}(\Omega) \right. \\ \left. + Y_{10}^{*}(\Omega'') \sum_{l,m} \left[ 1 + \frac{F_{l}^{a}}{2l+1} \right] A_{lm} \int d\Omega \ Y_{lm}(\Omega) Y_{l'm'}^{*}(\Omega) Y_{10}(\Omega) \right. \\ \left. + Y_{11}^{*}(\Omega'') \sum_{l,m} \left[ 1 + \frac{F_{l}^{a}}{2l+1} \right] A_{lm} \int d\Omega \ Y_{lm}(\Omega) Y_{l'm'}^{*}(\Omega) Y_{10}(\Omega) \right] \right] = iI_{l'm'}.$$

$$(2.15)$$

Here we have defined

$$\Omega_{l'} = \left[ \omega - \omega_0 + \frac{2\sigma^0}{\hbar} \left[ f_0^a - \frac{f_{l'}^a}{2l' + 1} \right] \right].$$
(2.16)

Now,

$$\int d\Omega Y_{lm}(\Omega) Y_{l'm'}^{*}(\Omega) Y_{1m''}(\Omega) = (-1)^{m} \left[ \frac{3(2l+1)(2l'+1)}{4\pi} \right]^{1/2} \begin{bmatrix} 1 & l & l' \\ 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} 1 & l' & l' \\ m'' & m & -m' \end{bmatrix}$$
$$= 0 \quad \text{if } l+l'=\text{even} .$$
(2.17)

To estimate the relative sizes of the coefficients for different l, we first assume that  $A_{l+1,m} \ll A_{lm}$ , and then find that for  $l \neq 0$ ,

$$\frac{A_{lm}}{A_{l-1,m}} \sim \frac{v_F q}{(\omega_0 + i/\tau_D)} , \qquad (2.18)$$

where we have taken  $I_{lm} \sim (-A_{lm}/\tau_D)$ . In the longwavelength limit, where  $v_Fq \ll \omega_0$ ,  $A_{lm} \ll A_{l-1,m}$  and so we may ignore coefficients of  $l \ge 2$ . In this case, we will then have only the following allowed terms in the sums:  $(l',m')=(0,0) \rightarrow (l,m)=(1,-1)$ , (1,0), and (1,1); also (l',m')=(1,1), (1,0), and (1,-1) all allow only (l,m)=(0,0). These then lead to the following equations:

$$\Omega_0 A_{00} = \frac{Q_1}{\sqrt{4\pi}} \left[ A_{10} Y_{10}^*(\Omega'') - A_{11} Y_{1,-1}^*(\Omega'') - A_{1,-1} Y_{11}^*(\Omega'') \right], \qquad (2.19)$$

$$\Omega_1 A_{11} - \frac{Q_0 A_{00}}{\sqrt{4\pi}} Y_{11}^*(\Omega'') = i I_{11} , \qquad (2.20)$$

$$\Omega_1 A_{10} - \frac{Q_0 A_{00}}{\sqrt{4\pi}} Y_{10}^*(\Omega'') = i I_{10} , \qquad (2.21)$$

$$\Omega_1 A_{1,-1} - \frac{Q_0 A_{00}}{\sqrt{4\pi}} Y^*_{1,-1}(\Omega'') = i I_{1,-1} , \qquad (2.22)$$

where we have defined

$$Q_l = \left[\frac{4\pi}{3}v_F q \left[1 + \frac{F_l^a}{2l+1}\right]\right].$$
 (2.23)

Here we have used the fact that  $I_{lm} = 0$  for l=0, since quasiparticle spin is conserved in collisions. In Appendix A we show that

$$I_{1m} = \frac{-(1+\frac{1}{3}F_1^a)}{2\tau} GA_{1m}(\epsilon) , \qquad (2.24)$$

where G is the collision operator (not to be confused with the same symbol used before for the magnetic field gradient), and obeys

$$Gf(t) = (\pi^2 + t^2)f(t) - \lambda_D \int_{-\infty}^{+\infty} dt' \frac{(t - t')f(t')}{\sinh[(t - t')/2]} .$$
(2.25)

Here,  $\tau = 8\pi^4 \hbar^6 / (m^*)^3 \langle W \rangle (k_B T)^2$  is the characteristic quasiparticle relaxation time,

$$\langle W \rangle = \int \frac{d\Omega}{4\pi} \frac{W(\theta,\phi)}{\cos(\theta/2)} ,$$

where  $W(\theta, \phi)$  is the spin-averaged scattering probability as a function of the usual Abrikosov-Khalatnikov angles  $(\theta, \phi)$ ,<sup>16</sup> whose definition is discussed in Ref. 15 (they are not the same as those we have previously symbolized by  $\Omega$ ).  $\lambda_D$  is given by

$$\lambda_D = 1 - \frac{1}{\langle W \rangle} \int \frac{d\Omega}{4\pi} \frac{W_{\uparrow\downarrow}(1 - \cos\theta)(1 - \cos\phi)}{2\cos(\theta/2)} , \quad (2.26)$$

where again  $(\theta, \phi)$  have the meaning defined in Ref. 15, and  $W_{\uparrow\downarrow}$  is the scattering probability for particles colliding with oppositely aligned spin. The value of  $\lambda_D$  lies between -3 and 1.

Now if we define  $Q(\epsilon) = iK(\epsilon)$  where

$$\frac{A_{11}(\epsilon)}{A_{00}(\epsilon)Y_{11}^{*}} \equiv \frac{-1}{\alpha} K(\epsilon) \cosh\left[\frac{\epsilon}{2k_{B}T}\right]$$
$$= \frac{2\sqrt{4\pi}}{3} v_{F}q\tau \frac{1+F_{0}^{a}}{1+F_{1}^{a}/3} K(\epsilon) \cosh\left[\frac{\epsilon}{2k_{B}T}\right]$$
(2.27)

(where formally we mean  $[1/A_{00}(\epsilon)] \equiv A_{00}^{-1}(\epsilon)$ , and we have omitted the  $\Omega''$  argument of the spherical harmonics) and write  $\Omega'_1 = \{[-2\tau/(1+F_1^a/3)]\Omega_1\}$ , we obtain from Eq. (2.20)

$$(i\Omega_1'+G)Q(\epsilon) = X = \frac{1}{\cosh(\epsilon/2k_BT)} .$$
 (2.28)

Then, in analogy to the notation of Ref. 13 and using the variable  $t = \epsilon/k_B T$ , we will be able to find bounds on

$$\sigma_{\rm Re} = \int_{-\infty}^{+\infty} \frac{-K_{\rm Im}(t)}{\cosh(t/2)} dt \qquad (2.29a)$$

and

$$\sigma_{\rm Im} = \int_{-\infty}^{+\infty} \frac{-K_{\rm Re}(t)}{\cosh(t/2)} dt , \qquad (2.29b)$$

where the real and imaginary parts are indicated. Now let us write

$$R(\epsilon) \equiv \frac{A_{11}(\epsilon)}{A_{00}(\epsilon)Y_{11}^*} . \tag{2.30}$$

Then we may look for  $R \equiv R(\epsilon = \epsilon_F)$  and we find that it leads to the form of Eq. (2.29) as follows:

$$4\alpha R_{\rm Im} = \frac{-6}{\sqrt{4\pi}} \frac{1 + F_1^a/3}{1 + F_0^a} \frac{1}{v_F q \tau} R_{\rm Im}$$
  
=  $4\alpha \int_{-\infty}^{+\infty} R_{\rm Im}(\epsilon) \left[ \frac{-\partial n_p^0}{\partial \epsilon_p} \right] d\epsilon$   
=  $\int_{-\infty}^{+\infty} \alpha R_{\rm Im}(t) \frac{dt}{\cosh^2(t/2)}$   
=  $\sigma_{\rm Re}$ . (2.31)

We may repeat this procedure for Eqs. (2.21) and (2.22) to get identical relations for the quantities  $(A_{10}/A_{00}Y_{11}^*)_{\rm Im}$  and  $(A_{1,-1}/A_{00}Y_{1,-1}^*)_{\rm Im}$ . We also obtain corresponding relations for  $\sigma_{\rm Im}$  in terms of the real parts of the expansion coefficients. Then, by using these bounds in Eq. (2.19), we finally obtain upper and lower bounds on  $[\sqrt{4\pi}(\Omega_0/Q_1)(4\alpha)(4\pi/3)] \equiv A(\omega-\omega_0)$ , where

$$A = \{-6/[(v_F q)^2 \tau (1 + F_0^a)]\}$$

The method of Ah-Sam *et al.* is summarized in Appendix B. Taking the bounds found by them we finally have

$$(\omega - \omega_0)_{\mathrm{Im}} \ge \frac{1}{A} \left[ \frac{(\Omega_1')^2}{a_1} + \frac{a_3}{a_1^2} \frac{(\Omega_1')^2 + (a_1 b_{32} / a_3 b_{12})}{(\Omega_1')^2 + (a_{-1} b_{32} / a_1 b_{12})} \right]^{-1},$$
(2.32)

$$(\omega - \omega_0)_{\rm Im} \le \frac{1}{A} \left[ \frac{(\Omega_1')^2}{a_1} + \frac{a_3}{a_1^2} \frac{(\Omega_1')^2 + (a_1b_{21}/a_3b_{01})}{(\Omega_1')^2 + [(a_1^3 + a_0^2a_3 - 2a_0a_1a_2)/(a_1b_{01})]} \right]^{-1},$$
(2.33)

$$(\omega - \omega_0)_{\text{Re}} \ge \frac{\Omega_1'}{A} \left[ \frac{(\Omega_1')^2}{a_0} + \frac{a_2}{a_0^2} \frac{(\Omega_1')^2 + [(a_0 b_{22})/(a_2 b_{02})]}{(\Omega_1')^2 + [(a_{-2} b_{22})/(a_0 b_{02})]} \right]^{-1},$$
(2.34)

$$(\omega - \omega_0)_{\text{Re}} \le \frac{\Omega_1'}{A} \left[ \frac{(\Omega_1')^2}{a_0} + \frac{a_2}{a_0^2} \frac{(\Omega_1')^2 + [(a_0b_{11})/(a_2b_{-11})]}{(\Omega_1')^2 + [(a_0^3 + a_{-1}^2 a_2 - a_{-1}a_0a_1)/(a_0b_{-11})]} \right]^{-1}.$$
(2.35)

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Here,  $a_n = (X, G^n X)$  where  $(A, B) = \int_{-\infty}^{+\infty} dt A(t)B(t)$  and  $b_{nm} = a_{n+m}a_{n-m} - a_n^2$ . The values of the matrix elements  $a_n$  are listed in Appendix B.

$$\Omega_1' = \frac{-2\tau}{1 + F_1^a/3} \left[ \omega - \omega_0 + \frac{2\sigma^0}{\hbar} (f_0^a - f_1^a/3) \right], \quad (2.36)$$

We also need

where

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$$\sigma^{0} = \delta n_{\uparrow} - \delta n_{\downarrow} = [(\gamma \hbar/2) N(0) H] / (1 + F_{0}^{a})$$
$$= (\hbar/2) N(0) \omega_{0} / (1 + F_{0}^{a}) ,$$

where N(0) is the density of states at the Fermi surface. Then, since  $(\omega - \omega_0) \ll \omega_0$ , we get  $\Omega'_1 = 2\tau \omega_0 \lambda$ , where

$$\lambda = \frac{1}{1 + F_0^a} - \frac{1}{1 + F_1^a/3} \tag{2.37}$$

is the parameter which characterizes the strength of the quasiparticle interaction.

In the low-temperature limit (this corresponds to  $\omega_0 \tau \lambda \gg 1$ ; note that  $\tau \sim 1/T^2$ ), the upper and lower bounds agree and we have

$$\frac{\Omega_1'}{A(\omega-\omega_0)_{\rm Re}} = \frac{(\Omega_1')^2}{a_0} + \left[\frac{a_2}{a_0^2}\right] + O((\Omega_1')^{-2}), \quad (2.38)$$

$$\frac{1}{A(\omega-\omega_0)_{\rm Im}} = \frac{(\Omega_1')^2}{a_1} + \left(\frac{a_3}{a_1^2}\right) + O((\Omega_1')^{-2}) . \quad (2.39)$$

These lead to

$$(\omega - \omega_0)_{\rm Re}(T \to 0) = \frac{-\frac{1}{3}(1 + F_0^a)(v_F q)^2}{\omega_0 \lambda} , \qquad (2.40)$$

 $(\omega - \omega_0)_{\text{Im}}(T \rightarrow 0)$ 

$$=\frac{-\frac{1}{3}(1+F_0^a)(v_F q)^2}{\omega_0^2 \lambda^2 \tau} [(2\pi^2/3)(1-\lambda_D)]. \quad (2.41)$$

The expression for  $(\omega - \omega_0)_{Re}$  is identical to that of Platzman and Wolff for  $T \rightarrow 0$ . The result for  $(\omega - \omega_0)_{Im}$ differs from Platzman and Wolff only in that  $\tau_D$  is replaced by a different relaxation time, which is given by

$$\tau_D \to \tau \left[ \frac{3}{2\pi^2} \frac{1}{(1-\lambda_D)} \right]. \tag{2.42}$$

This is the same as the  $\tau_{\rm pre}$  found by Pal and Bhattacharyya in their analysis of the Leggett-Rice effect [note that our  $\tau$  differs from their  $\tau(0)$  by a factor  $\pi^2/2$ ].

The high-temperature  $(\omega_0 \tau \lambda \ll 1)$  limit of the lower bounds gives

$$(\omega - \omega_0)_{\text{Re}}(T \to \infty) = (-\frac{1}{3})(v_F q)^2 (1 + F_0^a) \omega_0 \lambda \tau^2 a_{-2} , \qquad (2.43)$$

$$(\omega - \omega_0)_{\text{Im}}(T \to \infty) = [(-\frac{1}{6})(v_F q)^2 (1 + F_0^a) \tau] \left[ \frac{1}{6} + \frac{8\lambda_D}{\pi^2} \sum_{n=1,3,\dots} \frac{2n+1}{n^2(n+1)^2} \frac{1}{n(n+1) - 2\lambda_D} \right]$$

$$= (-\frac{1}{3})(v_F q)^2 (1 + F_0^a) \tau_D . \qquad (2.44)$$

(Note that the high-temperature limit of the upper bounds is not expected to lead to the exact result due to the choice of trial function. See Ref. 14.) The expression for  $(\omega - \omega_0)_{\text{Im}}$  agrees with that of Platzman and Wolff. The expression for  $(\omega - \omega_0)_{\text{Re}}$ , although in agreement to order  $(T^{-2})$ , is not  $\sim \tau_D^2$ , as is the Platzman-Wolff (PW) expression, and the discrepancy depends strongly on the value of  $\lambda_D$ . (This is analogous to the result of Egilsson and Pethick for the sound velocity.)

Our upper and lower bounds thus provide an interpolation between the high- and low-temperature limits, which is accurate and reliable for *all* temperatures in the Fermiliquid ( $T \ll T_F$ ) regime.

## **III. LEGGETT-RICE EFFECT**

(In this discussion of the Leggett-Rice effect, we will be following the notation of Ref. 15 fairly closely.) By starting with the kinetic equation for the spin density, an equation for the linearized spin current may be found which includes the effects of precession arising from the molecular field due to quasiparticle interaction. This equation for the spin current may then be related, through the equation for spin conservation, to the transverse component of the spin density. The time dependence of the transverse spin density may then be found, and from this, the amplitude of the echoes in a spin-echo experiment.

The equation for the spin current may be written in the following form:<sup>15</sup>

$$\frac{\partial}{\partial t} j_{\sigma,i}(\mathbf{r},t) + \frac{\partial}{\partial r_k} (\Pi_{\sigma})_{ik} = \left[ \frac{\partial j_{\sigma,i}}{\partial t} \right]_{\text{precession}} + \left[ \frac{\partial j_{\sigma,i}}{\partial t} \right]_{\text{collision}}$$
(3.1)

In the long-wavelength limit,

$$(\Pi_{\sigma})_{ik} = \delta_{ik} (1 + F_1^a / 3)(1 + F_0^a) \frac{v_F^2}{3} \delta\sigma(\mathbf{r}, t) , \qquad (3.2)$$

where  $\delta \sigma(\mathbf{r},t) \equiv \sigma(\mathbf{r},t) - \sigma^0(\mathbf{r},t)$ , and

$$\boldsymbol{\sigma}^{0}(\mathbf{r},t) \equiv \frac{\gamma \hbar}{2} \frac{N(0)\mathbf{H}(\mathbf{r},t)}{1+F_{0}^{a}}$$
(3.3)

is the local equilibrium magnetization that would be produced by a static field of the same value as the instantaneous external field.

For the precession term, one obtains<sup>5,15</sup>

$$\left|\frac{\partial j_{\sigma,i}}{\partial t}\right|_{\text{precession}} = \gamma j_{\sigma,i} \times \mathbf{H} - \frac{2}{\hbar} (f_0^a - f_1^a/3) (j_{\sigma,i} \times \sigma) , \qquad (3.4)$$

where the cross products involve the spin components of  $j_{\sigma}$ . Instead of using a relaxation-time approximation for the collision term, we write

$$\left(\frac{\partial j_{\sigma,i}}{\partial t}\right)_{\text{collision}} = (1 + F_1^a/3)I[j_{\sigma,i}], \qquad (3.5)$$

where, as before, the  $(1 + F_1^a/3)$  factor relates the l=1

moments of the spin density and the local equilibrium spin density (and so we leave off the overbar from the  $j_{\sigma,i}$ inside the I). This gives the following equation for the spin current:

$$\frac{\partial}{\partial t} j_{\sigma,i}(\mathbf{r},t) + (1 + F_1^a/3)(1 + F_0^a) \frac{v_F^2}{3} \frac{\partial}{\partial r_i} \delta \sigma = \gamma j_{\sigma,i}(\mathbf{r},t) \times \mathbf{H}(\mathbf{r},t) - \frac{2}{\tilde{\pi}} (f_0^a - f_1^a/3) j_{\sigma,i}(\mathbf{r},t) \times \sigma(\mathbf{r},t) + (1 + F_1^a/3) I[j_{\sigma,i}] .$$

$$(3.6)$$

The spin polarization precesses with the local Larmor frequency, and therefore, in the quasisteady state, the spin current will also precess about H(r) with the local Larmor frequency. That is, its explicit time dependence-apart from the time dependence contributed by the precession about the internal field, and by the collisional terms-will be given by

$$\frac{\partial}{\partial t} j_{\sigma,i}(\mathbf{r},t) = \gamma j_{\sigma,i}(\mathbf{r},t) \times \mathbf{H}(\mathbf{r}) . \qquad (3.7)$$

Using this in Eq. (3.6), we finally get

$$(1+F_{1}^{a}/3)(1+F_{0}^{a})\frac{v_{F}^{2}}{3}\frac{\partial}{\partial r_{i}}\delta\sigma$$

$$+\frac{2}{\hbar}(f_{0}^{a}-f_{1}^{a}/3)j_{\sigma,i}(\mathbf{r},t)\times\sigma(\mathbf{r},t)=(1+F_{1}^{a}/3)I[j_{\sigma,i}].$$
(3.8)

Taking the transverse components of this equation, where  $\sigma^+ = \sigma_x + i\sigma_y$ , we get

$$(1+F_1^a/3)(1+F_0^a)\frac{v_F^2}{3}\frac{\partial\sigma^+}{\partial r_i} - \frac{2i}{\hbar}(f_0^a - f_1^a/3)j_i^+\sigma_z$$
  
=(1+F\_1^a/3)I[j\_i^+] (3.9)

(where we have ignored a component  $\mathbf{j}_{\sigma_z} \sim \nabla \delta \sigma_z$ , which is very small since  $\delta \sigma_z \sim \text{const}$ ; see Leggett<sup>5</sup>). Then, with  $I[j_i^+] = (-G/2\tau)j_i^+$ , we get the equation

$$(G+i\Omega)Q(t) = X = \frac{1}{\cosh(t/2)}$$
, (3.10)

where

$$D^{+} = (2v_F^2/3)\tau(1+F_0^a)Q\cosh(t/2) , \qquad (3.11)$$

$$\Omega = \frac{4\tau}{\hbar} \left[ \frac{(f_1^a/3) - f_0^a}{1 + F_1^a/3} \right] \sigma_z \tag{3.12}$$

and  $D^+$  is defined by

$$j_i^+ = -D^+ \frac{\partial \,\delta\sigma^+}{\partial r_i} \,. \tag{3.13}$$

In the spin-echo experiment, the spins are tipped at an angle  $\phi$  from the external magnetic field and  $\sigma_z = \sigma^0 \cos \phi$ , while  $\Omega = 2\tau \lambda \omega_0 \cos \phi$ . Then we have virtually the identical problem as before, where we may find bounds on

$$\sigma_{\rm Re} = \int_{-\infty}^{+\infty} \frac{Q_{\rm Re}}{X} dt = \frac{6D_{\rm Re}^+}{\tau(1+F_0^0)v_F^2} . \tag{3.14}$$

Then the upper and lower bounds of  $D_{Re}^+$  will be the same

as those on  $(\omega_0 - \omega)_{\text{Im}}$ , except that  $\Omega^2$  contains an additional factor of  $\cos^2 \phi$ , and there is no factor q. Now we show how the quantity  $D^+$  relates to the spin-echo experiment.

When the kinetic equation for the spin density is summed over momenta, the contribution of the internal field term to the precession vanishes and the precession term becomes simply  $\gamma \sigma(\mathbf{r},t) \times \mathbf{H}(\mathbf{r},t)$ , i.e., precession only about the external magnetic field. Then one obtains the net spin conservation law

$$\frac{\partial \boldsymbol{\sigma}(\mathbf{r},t)}{\partial t} + \frac{\partial}{\partial r_i} j_{\boldsymbol{\sigma},i}(\mathbf{r},t) = \gamma \boldsymbol{\sigma}(\mathbf{r},t) \times \mathbf{H}(\mathbf{r},t) . \qquad (3.15)$$

Using  $\sigma = \sigma^0 + \delta \sigma$  and then taking the transverse component of this equation, we get

$$\frac{\partial \delta \sigma^+}{\partial t} + i\gamma H \,\delta \sigma^+ = D^+ \nabla^2 \delta \sigma^+ \,, \qquad (3.16)$$

where we have used the definition of  $D^+$  (and the terms in  $\sigma^0$  have dropped out). Now we follow Leggett in writing

$$\delta\sigma^{+}(\mathbf{r},t) = A(t) \exp[i\theta(t)] \exp[-i\xi(\mathbf{r},t)], \qquad (3.17)$$

where during any one free precession period,

$$\xi(\mathbf{r},t) = \xi(\mathbf{r},t_i) + \frac{\gamma}{\hbar} H(\mathbf{r})(t-t_i) . \qquad (3.18)$$

Here,  $t_i$  indicates the beginning of the period of free precession. Making this substitution and taking the real part gives us

$$\frac{\partial A}{\partial t} = -D_{\text{Re}}^{+} (\nabla \xi)^2 A , \qquad (3.19)$$

where only linear gradients in the magnetic field are considered significant. Then we get

$$\ln\left[\frac{A(t)}{A(t_i)}\right] = -D_{\text{Re}}^+ \int_{t_i}^t (\nabla \xi)^2(t') dt' . \qquad (3.20)$$

We have removed  $D_{Re}^+$  from the integral on the righthand side of 3.20. More exactly,  $D_{Re}^+$  also depends on  $A^{2}(t)$ , but in the limit of small tipping angle, this is a relatively insignificant term (see Leggett's derivation). Now we write with Leggett

$$\nabla \xi = \gamma G(t - t_i) , \qquad (3.21)$$

where  $G \equiv |\nabla H|$  is the standard notation for the gradient of the external field (not to be confused with the collision operator). We define  $h \equiv [A(t_2)/A(t_1)]$  as the ratio of the amplitudes of the transverse magnetization at

two different times. This will correspond to the amplitude of the echoes in a spin-echo experiment. In this experiment, the tipping pulse (of magnitude  $\phi$ ) occurs at t=0, the first 180° pulse takes place at  $t=t_0/2$ , and the n=1 echo occurs at  $t=t_0$ , where then  $t_0$  is the period between pulses and also between echoes. Then the magnetization will undergo the same precession during two periods: (i) the period between the first echo and the next spin flip, and (ii) the period between that spin flip and the next echo. Each period is of length  $t_0/2$ , and so we get

$$\ln h = -\frac{1}{12} D_{\rm Re}^{+} \gamma^2 G^2 t_0^2 . \qquad (3.22)$$

This is the equation for the decay of the echo amplitude written down by Leggett and Rice in the regime of small tipping angle. Here our  $D_{Re}^+$  corresponds to their  $D_{eff}$ , to which it may therefore be compared.

Note that to get results for large tipping angles, we would need to numerically integrate

$$\int \frac{dA}{D_{\rm Re}^+(A^2)A} , \qquad (3.23)$$

which we have not done.

## IV. RESULTS AND COMPARISON WITH EXPERIMENT

The principle application of these results is in the two Fermi liquids, pure <sup>3</sup>He and dilute mixtures of <sup>3</sup>He in <sup>4</sup>He. This discussion will be in the context of these systems.

There are only two experiments which are directly relevant to these results, those of Corruccini *et al.* on pure <sup>3</sup>He and on <sup>3</sup>He-<sup>4</sup>He mixtures, and that of Owers-Bradley *et al.* more recently on <sup>3</sup>He-<sup>4</sup>He mixtures. The recent experiment of Masuhara *et al.* cannot be easily compared, as will be discussed below. The experiment by Einzel *et al.* on multiple spin echoes is also not immediately relevant.

There are a number of different parameters which enter into our results. The Larmor frequency  $\omega_0$  just depends on the applied external field and the gyromagnetic ratio  $\gamma$ . The tipping angle  $\phi$  in the spin-echo experiments can be considered to be well specified. The wave number q in the spin-wave experiment, however, may be quite uncertain, as will be discussed further.

The parameters  $v_F$  and  $F_0^a$  are accessible through other experiments and may in principle be specified. There are considerable discrepancies in different measurements, however. Currently the most widely used values for <sup>3</sup>He seem to be those of Wheatley<sup>17</sup> and Greywall.<sup>18</sup>

The parameter  $F_1^a$  is not directly accessible from other experiments and its value is quite uncertain. It has generally been considered the "target" parameter of the spinwave experiments, whose value will be determined in the experiment.

The parameters  $\tau$  and  $\lambda_D$  in our expressions replace the spin-diffusion relaxation time  $\tau_D$  which appears in the previous derivations. There is a rigorous relation which links the three parameters (this relation is  $2\tau_D/\tau = a_{-1}$ , where  $a_{-1}$  is defined in Appendix B<sup>15</sup>), but  $\lambda_D$  is not itself directly accessible in experiments, apart from measurements of the other two. In <sup>3</sup>He,  $\tau$  has been measured

experimentally; in <sup>3</sup>He-<sup>4</sup>He mixtures, it has not. The various experimental determinations of  $\tau_D$  and the related spin-diffusion coefficient  $D_{\sigma}$  differ significantly from each other, and so it is not possible to consider this parameter as precisely determined.

Our first task is to compare our results with the usual expressions—those derived by Platzman and Wolff and by Leggett and Rice—for some self-consistent set of appropriate parameters in the two systems, <sup>3</sup>He and <sup>3</sup>He-<sup>4</sup>He mixtures. The object here is to examine the validity, as a function of temperature, of the relaxation-time approximation employing  $\tau_D$ . Our expectation is that, while this approximation should be increasingly adequate at higher temperatures—the regime of normal diffusion—it may not be accurate at low temperatures, where the precession-al effects due to the quasiparticle interactions are dominant.

The next goal is to directly compare our expressions with the available experimental data to see whether any new knowledge of the various parameters may be extracted. There are very limited data in the low-temperature regime, and the experimental uncertainties associated with many of the parameters are considerable, so any conclusions we draw must be very tentative. (We point out here that in this very-low-temperature regime, finite temperature corrections should be quite insignificant.)

Another possible task, which we have not carried out here, is to attempt to use our results in the large tippingangle regime of the Leggett-Rice effect, i.e., where the echo amplitude attenuation is not a simple exponential decay.

## A. <sup>3</sup>He

The quasiparticle lifetime at the Fermi surface, the parameter  $\tau$ , may be measured for pure <sup>3</sup>He in experiments on the superfluid phase.<sup>19</sup> We have used the values of  $\tau$  at 0 and 27 bars interpolated from these experiments.<sup>20,21</sup> The values depend somewhat on which values are adopted for the effective mass. (Note that Pal and Bhattacharyya use a pressure-independent value for  $\tau$ , which appears to differ by a factor  $\sim 2$  at 0 bar from the experimental curve.)

When a value of  $\tau_D$  is adopted, a value for the parameter  $\lambda_D$  may be extracted from the relation mentioned above. The problem is that there is considerable uncertainty in the value of  $\tau_D$ , since different experiments give substantially different results for the spin-diffusion coefficient. Note that once a spin-diffusion coefficient is measured, the value of  $\tau_D$  depends on the values adopted for  $v_F$  and  $F_0^a$ , through the relation

$$D_{\sigma} = (v_F^2/3)(1+F_0^a)\tau_D$$
,

so these must form a consistent set.

First we compare our results to that of Leggett and Rice. To do this we plot our equations and theirs, using the same set of parameters. The set we choose is simply the set used by Corruccini *et al.* to fit their data for <sup>3</sup>He; therefore, we use the  $\tau_D$  measured by this experiment. In addition, we use the recent values of  $\tau$  mentioned above, in particular, that value consistent with the Wheatley values for  $v_F$  and  $F_0^a$ , which are employed by Corruccini.

The results for 0 bar at a Larmor frequency of 23.5 MHz are shown in Figs. 1(a) and 1(b). A very similar figure results at 36.7 MHz, the other experimental frequency.

At increasingly high temperatures, the use of  $\tau_D$  is seen to be correct, as our upper bound becomes essentially identical to the Leggett-Rice expression (as discussed in Sec. II for the analogous spin-wave expression). We see that at temperatures below  $\sim 7 \text{ mK}$  ( $\sim 9 \text{ mK}$  at 36.7 MHz), the approximation using  $\tau_D$  breaks down, as our upper and lower bounds join above the Leggett-Rice plot. Figure 1(b) shows that the discrepancy is  $\sim 16\%$  as  $T \rightarrow 0$ . At 27 bars, using a frequency of 36.7 MHz, the breakdown occurs at  $\sim 6 \text{ mK}$  and the discrepancy is  $\sim 19\%$ .

The implication of these results is, that in the Leggett-Rice expressions,  $\tau_D$  should be replaced by a relaxation time characteristic of the precessional regime, designated by Pal and Bhattacharyya as  $\tau_{\rm pre}$ , where  $\tau_{\rm pre}=0.86\tau_D$  at 0 bar and  $0.84\tau_D$  at 27 bars.

The magnitude of the discrepancy is most strongly dependent on the ratio of the values of  $\tau_D$  and  $\tau$  which are used (which determine the value of the parameter  $\lambda_D$ ), and is not particularly sensitive to the other parameters. That is, a substantially smaller value for  $\tau$  (which implies

 $\lambda_D \rightarrow 1$ ) would decrease the discrepancy, while similar changes in the other parameters would not. (The analogous situation for sound attenuation is discussed by Egilsson and Pethick.)

Note that the above discussion is not directed at a comparison with the actual *data* for the Leggett-Rice effect measured by Corruccini *et al.*, but just a comparison with the theoretical expressions using some typical experimental parameters.

We have not made a similar comparison with the Platzman-Wolff expressions for spin waves in <sup>3</sup>He, since the only available experimental parameters are for measurements which are not directly related to our results. In particular, the experiment by Masuhara *et al.* measured only the imaginary part of the effective diffusion coefficient in the very-low-temperature regime, where the value of the relaxation time used has essentially no effect [see Eq. (2.40)]. Their attempt to fit their data in this regime with a relaxation time is not quantitatively reliable (cf. Masuhara<sup>9(b)</sup>). Note, that in any case, the results for the *real* part of the effective diffusion coefficient  $[(\omega_0 - \omega)_{Im}]$  are essentially the same for both the spin-wave case and the Leggett-Rice effect.

We have attempted to make an improved fit to the data of Corruccini *et al.* by using the most recent values for the various parameters (those of Greywall for  $v_F$  and  $F_0^a$ , and of Sachrajda *et al.*<sup>22</sup> for  $\tau_D$ ), while adjusting  $F_1^a$  for



FIG. 1. <sup>3</sup>He, 0 bar: (a) Comparison of variational bounds with the Leggett-Rice expression. Parameter values used are from data of Corruccini *et al.*:  $D_{\sigma}T^2 = 1.17 \times 10^{-6}$  cm<sup>2</sup> K<sup>2</sup>/sec;  $\lambda = 1.95$ ;  $\omega_0/2\pi = 23.5$  MHz;  $\phi = 18^{\circ}$ . Also used was  $\tau T^2 = 3.7 \times 10^{-12}$  sec K<sup>2</sup>. Dotted line is the upper bound, dashed line, the lower bound. (b) Fractional deviation of bounds from the Leggett-Rice result, using above parameters.



FIG. 2. <sup>3</sup>He, 0 bar: Fractional deviation of variational bounds from data fit line of Corruccini *et al.* Parameters used for bounds are from recent measurements:  $v_F = 5.99 \times 10^3$  cm/sec;  $F_0^a = -0.7$ ;  $\tau_D T^2 = 3.95 \times 10^{-13}$  sec K<sup>2</sup>. Also  $\phi = 18^\circ$ ;  $\omega_0/2\pi = 23.5$  MHz;  $\tau T^2 = 3.5 \times 10^{-12}$  sec K<sup>2</sup>. (a)  $F_1^a = -0.4$ , (b)  $F_1^a = -0.6$ .

the best fit at low temperatures. Note that due to the discrepancy in measurements of the diffusion coefficient from that of Corruccini *et al.*, it is not possible to fit the high-temperature part of the curve well for any choice of the other parameters. Therefore, our "best fit" is of questionable value. In addition, the data at low temperatures are very limited indeed.

Keeping these qualifications in mind, we show in Fig. 2, fits to the data at 0 bar, where in Fig. 2(a),  $F_1^a = -0.4$  is used and in Fig. 2(b)  $F_1^a = -0.6$  is used. These give the closest fits we were able to obtain. In Fig. 3, we plot results for <sup>3</sup>He at 27 bars, where in Fig. 3(a) a fit with the original parameters employed by Corruccini et al. is attempted, while in Fig. 3(b) more recent measurements are used and  $F_1^a = -0.4$  is assumed; other values for this parameter produced somewhat poorer fits. Precisely what constitutes a best fit is somewhat subjective, but we may say that our fits are consistent with values of  $F_1^a \approx -0.6$ at 0 bar and  $F_1^a \approx -0.4$  at 27 bars. These may be compared to values found by other workers, which include many different values in the range  $|F_1^a| < 1$  (see Refs. 22) and 23, and references therein). (It should be noted that the values for  $F_1^a$  extracted by Sachrajda et al. and Masuhara et al. from the Corruccini data using the



FIG. 3. <sup>3</sup>He, 27 bars: Fractional deviation of variational bounds from the data fit line of Corruccini *et al.* for  $\omega_0/2\pi = 36.7$  MHz,  $\phi = 20^\circ$ : (a) using parameters of Corruccini *et al.*:  $D_{\sigma}T^2 = 1.25 \times 10^{-7}$  cm<sup>2</sup>K<sup>2</sup>/sec;  $\lambda = 2.9$ ; also  $\tau T^2 = 1.6 \times 10^{-12}$  sec K<sup>2</sup>; (b) using recent measurements:  $v_F = 3.57 \times 10^3$  cm/sec;  $F_0^a = -0.76$ ;  $\tau_D T^2 = 1.44 \times 10^{-12}$  sec K<sup>2</sup>; also,  $\tau T^2 = 1.7 \times 10^{-12}$  sec K<sup>2</sup>. Fit using  $F_1^a = -0.4$ .

Greywall parameters are simply a rescaling of  $\lambda$  with respect to  $\tau_D$  and are not a refit of the data.)

## B. <sup>3</sup>He-<sup>4</sup>He mixtures

In the <sup>3</sup>He-<sup>4</sup>He mixtures, the parameters  $\tau$  and  $\lambda_D$  are experimentally unknown, although a theoretical calculation of  $\lambda_D$  by Fu and Pethick<sup>24</sup> yielded a value of 0.81 for a 5% solution. Since the validity of the approximation employing  $\tau_D$  is strongly dependent on the value of  $\lambda_D$ , as discussed in the preceding section, it is not possible to state an unambiguous result in this system, while it was, essentially, possible to do so in <sup>3</sup>He.

For a value of  $\lambda_D \sim 0.8$ , the discrepancy between our results and the results of both Leggett and Rice and Platzman and Wolff for the real part of the effective diffusion coefficient is  $\leq 2\%$ . For the imaginary part, the discrepancy between the results of Platzman and Wolff and ours is even smaller than that, by an order of magnitude. A typical plot is shown in Fig. 4. The other parameters we have used for these comparisons are those adopted by Corruccini et al. and Owers-Bradley et al. to fit the data in their respective experiments. These experiments were at 6.4% concentration and 5% concentration, respectively, both observed at 0 bar. We have also used the value of q obtained by Owers-Bradley *et al.* as a fit to their data. However, this may not be very reliable, see below. (Note also that although the Owers-Bradley data for the real part of the diffusion coefficient is well fit in terms of the shape of the curve by Platzman and Wolff, there is a large, systematic discrepancy in amplitude.)

If one does not use the above values of  $\lambda_D$  for <sup>3</sup>He-<sup>4</sup>He mixtures, but instead substitutes values which are typical of those in the pure <sup>3</sup>He system (i.e.,  $\sim -1$ ), the discrepancy between our bounds and the Platzman-Wolff results becomes very much larger, not surprisingly. Indeed, the proportional discrepancy becomes roughly the same as was found in pure <sup>3</sup>He.

The discrepancy in the high-temperature limit of the imaginary part of the diffusion constant, mentioned at the end of Sec. II, varies from  $\sim 10\%$  for  $\lambda_D = -1$ , to  $\sim 0.2\%$  for  $\lambda_D = 0.8$ .



FIG. 4. <sup>3</sup>He-<sup>4</sup>He, 5%, 0 bar: Fractional deviation of the variational bounds from the Platzmann-Wolff expression for  $(\omega_0 - \omega)_{Im}$ , for  $\lambda_D = 0.8$ . Parameters used are those of Owers-Bradley *et al.*:  $v_F = 2.7 \times 10^3$  cm/sec;  $F_0^a = 0.08$ ;  $F_1^a = 0.34$ ;  $\tau_D T^2 = 2.8 \times 10^{-11}$  sec K<sup>2</sup>; q = 10.1 cm<sup>-1</sup>;  $\omega_0/2\pi = 0.925$  MHz.



FIG. 5. <sup>3</sup>He-<sup>4</sup>He, 5%, 0 bar: Fractional deviation of the variational bounds from the Platzmann-Wolff expression for  $(\omega - \omega_0)_{\text{Re}}$ , as a function of  $\lambda_D$ , at the temperature corresponding to  $\omega_0 \tau_D \lambda = 4$ .

This can be understood from Fig. 5, where the deviation of the variational bounds from the Platzman-Wolff expressions will be inaccurate and the actual  $\lambda$  (and thus  $F_0^a$  and  $F_1^a$ ) will differ from the value fit from the PW expression. In principle, one would have to fit the experiproach those of Platzman and Wolff. (This is analogous to the results of Egilsson and Pethick for sound attenuation.)

Now, the data of Owers-Bradley *et al.*, which is fit by the Platzman-Wolff expression, would imply a value for  $\lambda$  (the combination of  $F_0^a$  and  $F_1^a$ ), if the values of q,  $\omega_0$ ,  $v_F$ , and  $\tau_D$  are considered to be precisely known. Then this value of  $\lambda$  will be accurate to the extent that  $\lambda_D \sim 1$ .

If, however,  $\lambda_D$  is far from 1, then the Platzman-Wolff expressions will be inaccurate and the actual  $\lambda$  (and thus  $F_0^a$  and  $F_1^a$ ) will differ from the value fit from the PW expression. In principle, one would have to fit the experimental data from our expressions, leaving both  $\lambda$  and  $\lambda_D$ as free parameters. In effect, this would allow one to fit the quasiparticle relaxation time  $\tau$  as well, since this parameter follows directly from  $\lambda_D$ .

If one simply fits the data to the PW expressions, then different values of  $\lambda_D$ —i.e., different  $\tau$ 's—will result in much bigger disagreements between our expressions and the data. This is illustrated in Fig. 6. Here it can be seen that the agreement using  $\lambda_D = 0.8$  is an order of magnitude better than the agreement using  $\lambda_D = 0.5$ , and several orders of magnitude better than with  $\lambda_D < 0$ .

If one could somehow pin down the value of  $\lambda$  independently (and the other relevant parameters mentioned above were precisely known), then one could, in principle, extract  $\tau$ . For instance, if in this case  $\lambda$  were equal to the value of Owers-Bradley *et al.* with less than a 0.3% error, it would imply a value of  $\lambda_D \sim 0.8$ . This would be consistent with a value for  $\tau T^2 \leq 3 \times 10^{-11} \sec K^2$ .

However, the recent work of Masuhara *et al.* casts some doubt on a naive interpretation of the data of Owers-Bradley *et al.* such as ours. This is because of their determination that a correct treatment of cell geometry and boundary conditions is extremely important in a quantitative analysis of spin-wave modes, such as those observed by Owers-Bradley *et al.* In particular, the value of q used by Owers-Bradley *et al.* to fit their data is not clearly related to their cell geometry and may be substantially inaccurate. Masuhara *et al.* do not use the Platzman-Wolff results to analyze their data, but instead have derived detailed numerical fits which incorporate the effect of boundary conditions.

In view of this we cannot make any quantitatively reliable statement about  $\tau$ . However, it is interesting to note that the  $\lambda_D$  value of Fu and Pethick would be consistent with high reliability of the parameter values obtained from fits to Platzman-Wolff expressions.

The value of  $F_0^a$  used by both Corruccini *et al.* and Owers-Bradley *et al.* is 0.08. Recent results suggest that this should perhaps by replaced by values of 0.0 at 6.4% concentration and 0.3 for a 5%-concentration solution,



FIG. 6. <sup>3</sup>He-<sup>4</sup>He, 5%, 0 bar: Fractional deviation of the variational bounds from data of Owers-Bradley *et al.* for  $(\omega - \omega_0)_{\text{Re}}$ . Parameters are as in Fig. 4. (a)  $\lambda_D = -1.0$ , (b)  $\lambda_D = 0.5$ , (c)  $\lambda_D = 0.8$ .

and also that  $D_{\sigma}$  may be somewhat different than the value used by these groups.<sup>25</sup> These would then also require a rescaling of  $\tau_D$ . However, in these cases the  $\lambda_D$  parameter is still the determining unknown quantity, and our discussion above regarding its value still is valid.

#### V. SUMMARY

We have calculated upper and lower bounds on the effective diffusion coefficient for spin-wave phenomena in Fermi liquids, including those measured in a spin-echo observation of the Leggett-Rice effect at small tipping angle. By treating the collision integral exactly, we avoid the approximation made in using the spin-diffusion relaxation time  $\tau_D$  in the low-temperature, precession-dominated regime. Therefore, our bounds should hold throughout the whole Fermi-liquid ( $T \ll T_F$ ) regime (in the long-wavelength limit).

Our results indicate that the relaxation-time approximation employing  $\tau_D$  is inaccurate at low temperatures, and that as  $T \rightarrow 0$  the effective relaxation time approaches a value of  $\{\tau(3/2\pi^2)[1-\lambda_D]^{-1}\}$ , where  $\tau$  is the characteristic quasiparticle relaxation time and  $\lambda_D$  is a function of the scattering amplitude.

In <sup>3</sup>He we find that the approximation using  $\tau_D$  breaks down below ~7 mK, and the error approaches 16–20%. In <sup>3</sup>He-<sup>4</sup>He mixtures at concentrations of ~5%, the error due to the use of  $\tau_D$  appears to be small (<2%).

Our results are consistent with values of  $F_1^a$  in <sup>3</sup>He of  $\approx -0.6$  at 0 bar and  $\approx -0.4$  at 27 bars. Experimental difficulties make these values substantially uncertain.

### **ACKNOWLEDGMENTS**

We thank Chris Pethick for useful suggestions and for helpful discussions. We are grateful to Naoto Masuhara and David Edwards for detailed discussions of the work done at Ohio State University. This work was supported in part by U.S. Department of Energy Contract No. DE-AC02-76ER13001.

### APPENDIX A

We sketch below the reduction of the collision integral. Many details can be found in Ref. 15.

The linearized collision integral can be written as

$$I[\bar{\sigma}_{\mathbf{p}'}] = \frac{-1}{k_B T V^2} \sum_{2} \sum_{3,4}' n_1 n_2 (1-n_3) (1-n_4) W(1,2;3,4) \delta_{\mathbf{p}_1 + \mathbf{p}_2, \mathbf{p}_3 + \mathbf{p}_4} \delta_{\sigma_1 + \sigma_2, \sigma_3 + \sigma_4} \delta(\epsilon_1 + \epsilon_2 - \epsilon_3 - \epsilon_4) (\bar{\Phi}_1 + \bar{\Phi}_2 - \bar{\Phi}_3 - \bar{\Phi}_4) ,$$
(A1)

where  $\overline{\Phi}_i \equiv \delta \overline{\sigma}_i / (-\partial n_i^0 / \partial \epsilon_i)$  and W(1,2;3,4) is a transition probability for the scattering  $|\mathbf{p}_1 \sigma_1, \mathbf{p}_2 \sigma_2 \rangle \rightarrow |\mathbf{p}_3 \sigma_3, \mathbf{p}_4 \sigma_4 \rangle$ . (Our notation  $I[\overline{\sigma}_{\mathbf{p}'}]$  is unconventional.) Then, in the method of Ref. 16, this becomes

$$I[\bar{\sigma}_{\mathbf{p}'}] = \frac{-1}{k_B T} \frac{(m^*)^3}{(2\pi\hbar)^6} \int d\epsilon_2 \int d\epsilon_3 \int d\epsilon_4 n_1 n_2 (1-n_3)(1-n_4) \\ \times \int_0^{\pi} d\theta \sin\theta \int_0^{2\pi} d\phi \int_0^{2\pi} d\phi_2 \frac{W(\theta,\phi)}{\cos(\theta/2)} (\bar{\Phi}_1 + \bar{\Phi}_2 - \bar{\Phi}_3 - \bar{\Phi}_4) .$$
(A2)

After changing to the variables  $x_i = \epsilon_i / (k_B T)$  and defining the variable  $\psi(x_i)$  by writing

$$\overline{\Phi}_i = -\mathbf{v}_i \cdot \nabla \mu(\mathbf{r}, t) \tau \psi(\epsilon_i) ,$$

where  $\mu(\mathbf{r},t)$  is the chemical potential and  $\tau$  is the relaxation time defined in Sec. II, one obtains

$$I[\bar{\sigma}_{\mathbf{p}'}] = \frac{-1}{k_B T \tau} \int_{-\infty}^{\infty} dx_2 \int_{-\infty}^{\infty} dx_3 \int_{-\infty}^{\infty} dx_4 n_1 n_2 n_3 n_4 \delta(x_1 + x_2 - x_3 - x_4) [\psi(x_1) - \lambda_D \psi(x_2)] .$$
(A3)

[In the above equations the local spin quantization axis has been taken to be along the net spin polarization vector  $\hat{\sigma}_{p}(\mathbf{r},t)$ .] This becomes

$$I[\bar{\sigma}_{\mathbf{p}'}] = \frac{-1}{k_B T \tau} \left[ \frac{x_1^2 + \pi^2}{2} n_1 (1 - n_1) \psi(x_1) -\lambda_D n_1 \int_{-\infty}^{\infty} dx_2 \frac{(x_1 + x_2) n_2}{1 - e^{-(x_1 + x_2)}} \psi(x_2) \right].$$
(A4)

When we expand the collision integral in a series of spherical harmonics (by expanding the  $\delta \overline{\sigma}_{p'}$  function contained in it) and switch to the variable  $\overline{\nu}_{p'}$  we find from this equation that the l=1 moment gives

$$I_{1m} = (-1/2\tau) G \overline{A}_{1m}(\epsilon) \; .$$

Finally, Eq. (2.24) follows from the relation

$$\delta \bar{n}_{\mathbf{p}} \equiv \delta n_{\mathbf{p}} - \frac{\partial n_{p}^{0}}{\partial \epsilon_{p}} \delta \epsilon_{\mathbf{p}} = \delta n_{\mathbf{p}} - \frac{\partial n_{p}^{0}}{\delta \epsilon_{p}} \int \frac{d^{3}p'}{(2\pi\hbar)^{3}} f_{\mathbf{p}\mathbf{p}'} \delta n_{\mathbf{p}'} , \qquad (A5)$$

which defines the overbar notation, and which generates the factor  $(1 + F_1^a/3)$ .

#### APPENDIX B

Here we summarize the variational method of Ah-Sam et al.<sup>13</sup> This method is directed at solving the linear, inhomogeneous integrodifferential equation

$$HQ = X , (B1)$$

where Q is an unknown function of the variable t, X is the inhomogeneous term, and H is a linear integrodifferential operator. The goal will be to find variational bounds on a scalar product of Q and X, denoted by

$$T = (Q^*, X) , \qquad (B2)$$

where

$$(U^*, V) = \int dt \ U^*(t) V(t) \ . \tag{B3}$$

It has long been known<sup>26</sup> that if H is Hermitian, that is,  $(U^*,HV)=(V^*,HU)^*$  and is positive—i.e., only has nonnegative eigenvalues—then T may be bounded from below according to

$$T = (Q^*, X) \ge \frac{[\operatorname{Re}(U^*, X)]^2}{(U^*, HU)} , \qquad (B4)$$

where U is an arbitrary trial function.

It was shown by Jensen *et al.* that if H can be separated into two positive Hermitian operators J and L,

$$H = J + L , (B5)$$

then, if either J or L has an inverse, T may also have an upper bound. If  $J^{-1}$  exists, this upper bound is

$$T = (Q^*, X) \le (X^*, J^{-1}X) - \frac{\{\operatorname{Re}[U^*, (HJ^{-1} - 1)X]\}^2}{[U^*, (HJ^{-1} - 1)HU]}.$$
(B6)

Ah-Sam *et al.* consider the situation when H is non-Hermitian. They write it as

$$H = G + A , \tag{B7}$$

where G is Hermitian, and A is anti-Hermitian. (We will assume here that G and X are real.) They introduce the functions

$$f = \frac{1}{2}(Q + Q^*), \quad g = (1/2i)(Q - Q^*)$$
 (B8)

and obtain separate equations for f and g,

$$(G+iAG^{-1}iA)f=X, (B9)$$

$$[iA + G(iA)^{-1}G]g = X, (B10)$$

where the existence of  $G^{-1}$  and  $A^{-1}$  is assumed. This al-

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lows them to obtain upper and lower bounds on the quantities

$$\sigma_{\text{Re}} = (f, X) = (Q_{\text{Re}}, X), \ \sigma_{\text{Im}} = -(g, X) = -(Q_{\text{Im}}, X).$$
  
(B11)

Using the trial functions U = pX + c(1-p)GX,  $c = a_0/a_1$ (for the lower bounds) and  $U = pG^{-1}X + c(1-p)GX$ ,  $c = a_1/a_3$  (for the upper bounds), Ah-Sam *et al.* then find the upper and lower bounds as a function of the matrix elements

$$a_n = (X, G^n X) = \int_{-\infty}^{+\infty} ds \, X(s) G^n X(s) \,. \tag{B12}$$

Ah-Sam *et al.* then apply this to obtain bounds related to the equation  $(G+i\Omega)Q = X$ , where G is the collision operator [Eq. (2.25)] and X is as defined in Eq. (2.28). They also obtained the matrix elements, which are as follows (in terms of  $\lambda_D$ ):

$$\begin{aligned} a_{-2} &= \pi^{-4} \left[ 2.847 + 2.250 \left[ \frac{4\lambda_D^2}{(2 - 2\lambda_D)^2} \right] \right. \\ &+ 4.935 \left[ \frac{2\lambda_D}{(2 - 2\lambda_D)} \right] + 0.461 \left[ \frac{2\lambda_D}{(12 - 2\lambda_D)} \right] \\ &+ 0.292 \left[ \frac{4\lambda_D^2}{(2 - 2\lambda_D)(12 - 2\lambda_D)} \right] \\ &+ 0.057 \left[ \frac{4\lambda_D^2}{(12 - 2\lambda_D)^2} \right] + \cdots \right], \\ a_{-1} &= \frac{1}{3} + \frac{8}{\pi^2} \lambda_D \sum_{n=1,3,\dots} \frac{2n + 1}{n^2(n+1)^2} \frac{1}{n(n+1) - 2\lambda_D} \\ a_0 &= 4, \\ a_1 &= \pi^2 \frac{16}{3} (1 - \lambda_D), \\ a_2 &= \pi^4 \frac{128}{15} (1 - \lambda_D)^2, \\ a_3 &= \pi^6 \frac{256}{105} (1 - \lambda_D)^2 (8 - 6\lambda_D), \\ a_4 &= \pi^8 \frac{2048}{315} (1 - \lambda_D)^2 (480 - 456\lambda_D + 200\lambda_D^2 - 40\lambda_D^3). \end{aligned}$$

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