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Spin Diffusion in Doubly Spin Polarized Atomic Deuterium*

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Using an extension of the Boltzmann equation for the Wigner distribution appropriate for dilute spin 1 *systems, spin diffusion equations are derived in the limit of large nuclear polarization in the non-degenerate régime. As an example of a system to which this work may be applied, the domain of validity of the Boltzmann equation for doubly spin-polarized deuterium, D↓↓, is studied. The effect of a finite field gradient is discussed. A calculated spin wave spectrum for a model one-dimensional system in the presence of a gradient is presented. Analogous effects in spin* 1/2 *systems are compared and contrasted.*

1. INTRODUCTION

Spin diffusion in quantum systems produces effects that have no analogue in conventional diffusion systems. The spin-polarized quantum gases ^{3}He↓, H↓↓, and D↓↓ show these effects far from the degenerate régime. For D↓↓ and H↓↓, the arrows indicate that the electron and nucleus are polarized. In the case of He↓, only the nucleus is polarized. The momentum distribution is well described by a Boltzmann distribution, so that the systems under discussion bear some similarities to non-degenerate semiconductors. That this is so follows from a chain of inequalities, which defines a dilute

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non-degenerate, quantum gas:

$$a \ll \lambda \ll n^{-1/3} \ll l \ll L, \tag{1}$$

where a is the scattering range of the potential, λ is the thermal de Broglie wavelength, $\lambda = \sqrt{2\pi\hbar^2/mk_BT}$, $n^{-1/3}$ is the mean interparticle spacing, l is the mean free path in the gas, and L is the characteristic sample size.

The first inequality ensures that quantum-mechanical interference effects are important during collisions. The second ensures that one has a non-degenerate system. The third condition is the criterion for a dilute gas, and the final inequality enforces the hydrodynamic limit.

If the second inequality were reversed, one would be in the degenerate régime. In H↓↓, a bosonic system, one might see Bose-Einstein condensation for example. Similarly, in D↓↓, a fermionic system, one would expect to see the effects of Fermi degeneracy and ultimately 'gaseous superfluidity' via Cooper pairing.

Unfortunately, laboratory densities are far from the degenerate régime to date. If one wishes to study doubly spin polarized atomic deuterium gas, D↓↓, it is reasonable to consider the Boltzmann equation for the transport properties because the domain of validity coincides with the densities and polarizations that are experimentally feasible. The approach developed here is based on a Chapman-Enskog analysis of the Boltzmann equation when particle indistinguishability effects are built into the collision term.

In order to create doubly spin polarized hydrogen or deuterium gas, one relies on a strong magnetic field to polarize the electron spins. The nuclear spins remain unpolarized initially, because the Boltzmann factor is still nearly unity for achievable magnetic fields and temperatures in the nuclear spin space. Recombination events, however, can lead to profound modifications of the electron spin down nuclear spin manifold. The energy level diagram of atomic deuterium in low magnetic field is shown in Fig. 1. We are interested in the $|\alpha\rangle$, $|\beta\rangle$ and $|\gamma\rangle$ states. The $|\gamma\rangle$ state has no admixture of electron spin up and is described by $m_I = -1$ in nuclear spin space, where m_I is the nuclear magnetic quantum number; hence, the $|\gamma\rangle$ state is analogous to the $|b\rangle$ state in doubly spin polarized hydrogen. The remaining states in the nuclear spin manifold, the $|\beta\rangle$ and $|\alpha\rangle$ states, have a small admixture of electron spin up, due to the hyperfine interaction. If there is a third body to conserve energy and momentum, the $|\alpha\rangle$ and $|\beta\rangle$ states will recombine through the electronic singlet state bonding interaction. After recombination events have depleted the $|\alpha\rangle$ and $|\beta\rangle$ states, the nuclei are essentially completely polarized in the $|\gamma\rangle$ state. Note that this is a metastable state describable by a negative nuclear spin temperature. Because this is a rarefied gas, one expects that $T_{1,n}$ nuclear rates will be slow due to the $T_{1,n}$ bottleneck,[1] well-known from spin polarized hydrogen work.

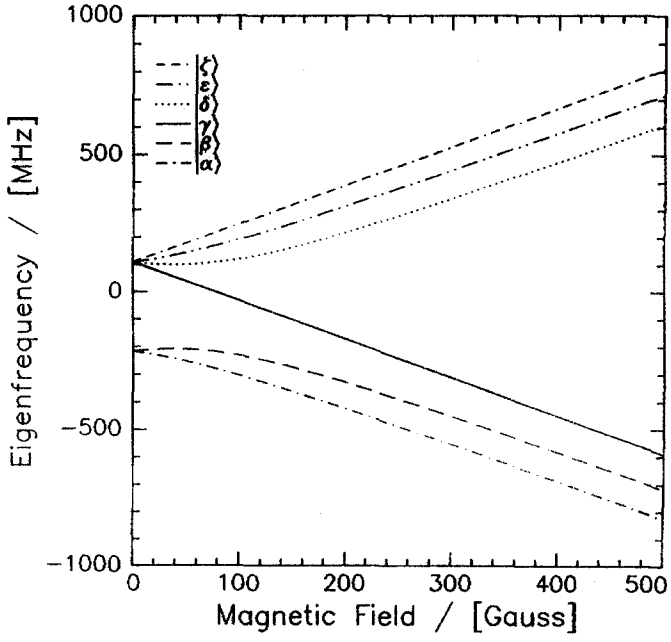

Fig. 1. Energy level diagram for atomic deuterium in a magnetic field.

Thus, a rarefied sample initially polarized only in the electronic states would become doubly spin polarized after short times, due to selective recombination, and would be metastable on the time scale of $T_{1,n}$.

The simplest NMR experiment that one could perform on doubly spin polarized atomic deuterium would be to look at the $|\gamma\rangle - |\beta\rangle$ transition. It is interesting to note that the hyperfine interaction introduces a small relative splitting between the two $\Delta m_I = 1$ transitions in the electron spin down manifold: $|\gamma\rangle - |\beta\rangle$ and $|\beta\rangle - |\alpha\rangle$, which is shown in Fig. 2. Figure 3 shows the $|\gamma\rangle - |\beta\rangle$ transition energy as a function of applied magnetic field. The spectrum that one would observe by irradiating the $|\gamma\rangle - |\beta\rangle$ transition will not have a significant admixture from the $|\beta\rangle - |\alpha\rangle$ transition for the following reasons. Initially, the $|\beta\rangle$ and $|\alpha\rangle$ states are nearly unpopulated, so that the $|\beta\rangle - |\alpha\rangle$ transition would be much weaker than the $|\gamma\rangle - |\beta\rangle$ transition. Secondly, the relative splitting of the $\Delta m_I = 1$ transition would prevent spectral overlap for the modest gradients that we propose in Sec. 6.

Reynolds, Hayden and Hardy[2] have reviewed attempts to produce and study D↓↓. The work of Shinkoda, *et al.*[3] in particular is important because their experiments indicate what densities and temperatures are feasible with conventional experimental techniques. They achieved densities appropriate

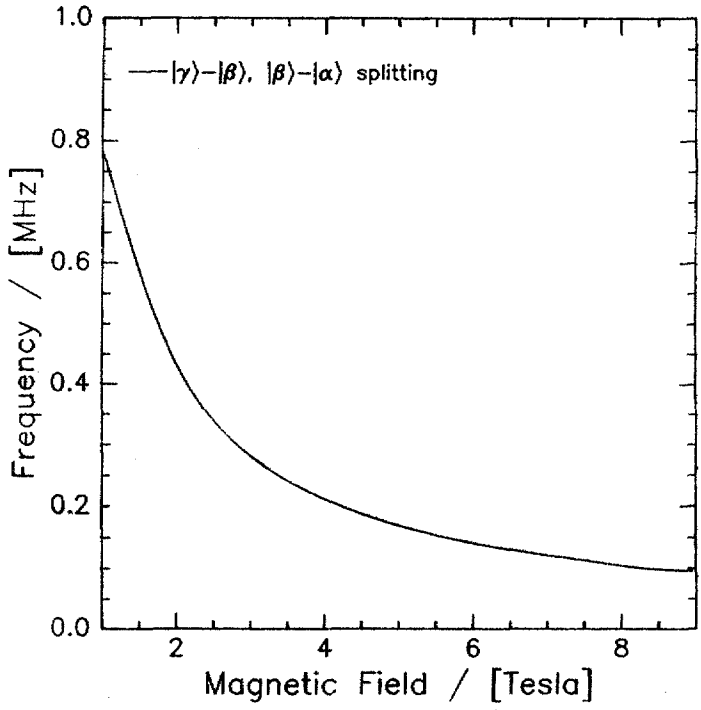

Fig. 2. Relative splitting of $\Delta m_l = 1$ transitions in D↓↓.

for a dilute, nondegenerate, hydrodynamic fluid at temperatures sufficiently high that the deuterium gas was primarily in the bulk phase, not the surface phase. These conditions correspond to the upper left hand corner of Fig. 4. Buckle[4] has developed a Fermi liquid theory for D↓↓, which assumes that the momentum distribution is degenerate. The domain of validity of such a theory as it stands is shown in the lower right hand side of Fig. 4, which has not yet been reached experimentally. As such, Buckle's theory does not apply to the dilute case considered here. Nevertheless, the Chapman-Enskog approach studied here, and the Fermi liquid theory of Buckle have some points in common, and analogies between the two approaches will be exploited as appropriate at various places in the sequel. In order to facilitate comparison between Buckle's treatment and this work, we have used a notation that is compatible with Buckle.[4]

The Chapman-Enskog approximation is expected to break down when the density is too high, e.g. in the degenerate régime, as well as when the density is too low so that the mean free path becomes comparable to the sample cell size, i.e. in the Knudsen régime. Buckle's results suggest that

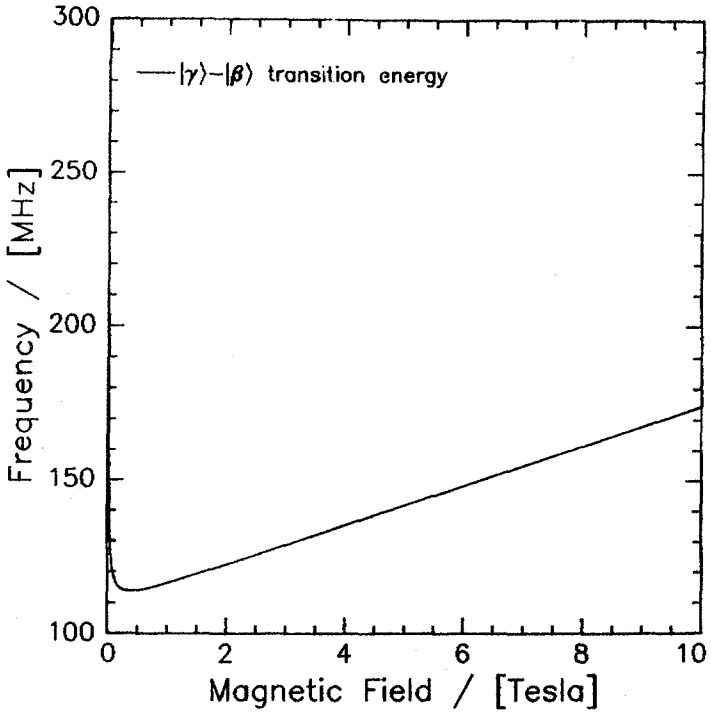

Fig. 3. $|\gamma\rangle$—$|\beta\rangle$ transition energy in D↓↓ as a function of applied magnetic field.

the behavior in the degenerate régime, at least not too far below T_F, will not be qualitatively different from the dilute limit considered here. Thus we find that the two approaches lead to similar results. Furthermore, recent work[5] in doubly spin polarized hydrogen, H↓↓, shows that the onset of the molecular or Knudsen régime does not lead to qualitatively different spin transport even though a hydrodynamic theory is no longer valid.

Spin transport in these systems is therefore amenable to study by several theoretical approaches. The method chosen here is, we believe, the one that corresponds most closely to the physical conditions that can be realized in the laboratory.

Spin diffusion is driven by gradients in the magnetization, which cause a spin current. As the spins diffuse, they interact with one another, which modifies the spin current, and thus the nonequilibrium magnetization *via* the continuity equation. The scattering processes will lead to quantum mechanical modifications of the spin current when the de Broglie wavelength becomes longer than the scattering range of the potential. The spin 1/2 case

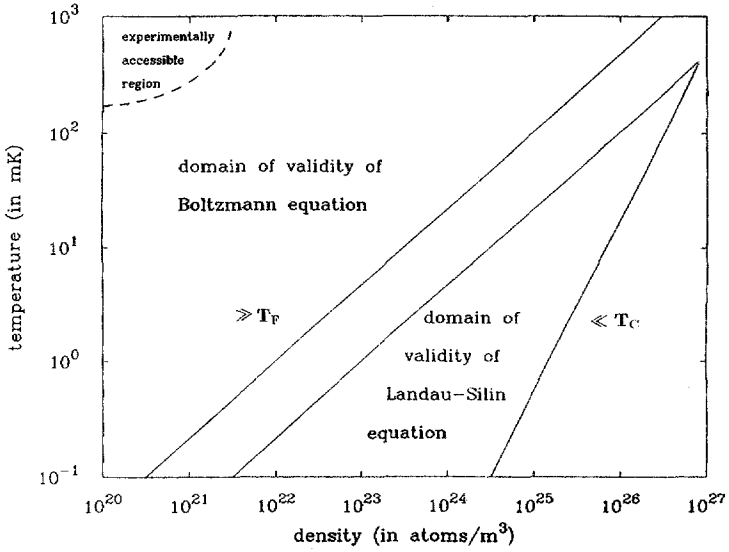

Fig. 4. The domains of validity of this work (low density region) and the work by Buckle (high density region) are plotted as a function of density and temperature. The temperatures and densities accessible by present-day cryogenic technology are also plotted. The Fermi temperature, T_F, runs between both domains. The domain of validity of Buckle's work is bounded at high densities by the gaseous superfluidity[1] transition temperature, T_c. See text for discussion.

is well known:

$$\mathbf{J}_i = -D_0 \operatorname{grad} M_i, \tag{2}$$

where $\mathbf{J}_i$ is the contribution to the spin current due to M_i, M_i is the ith component of the nuclear magnetization and D_0 is the classical diffusion coefficient. When the correct quantum mechanical description of scattering is incorporated into the theory, Eq. (2) becomes[6]

$$J_i(M_j) = -\frac{D_0}{1+\mu^2 M^2}\left\{\frac{\partial M_j}{\partial x_i} - \varepsilon\mu\left(M \wedge \frac{\partial \mathbf{M}}{\partial x_i}\right)_j + \mu^2 M_j\left(\mathbf{M}\cdot\frac{\partial \mathbf{M}}{\partial x_i}\right)\right\}, \tag{3}$$

where ε is $+1(-1)$ for bosons(fermions) and $\mathbf{M}$ is the nuclear magnetization; μ is a measure of the importance of exchange scattering relative to direct scattering and is analogous to the μ parameter defined by Silin[7] for degenerate Fermi liquids. We shall refer to μ as the spin wave quality factor. The importance of μ may be seen by taking the limit as $\mu \to 0$. One recovers the classical diffusion result of Eq (2).

One may derive a diffusion equation by inserting Eq. (3) into the continuity equation

$$\frac{\partial M_i}{\partial t} + \operatorname{div} \mathbf{J}_i = 0 \tag{4}$$

As will be shown *ut infra*, in the limit of small perturbations from a large static magnetization (small tip angle), the diffusion equation for the magnetization that one derives bears a resemblance to the Schrödinger equation.

The goal of this paper is to derive the spin 1 analogue of Eq. (3) in order to obtain a spin 1 diffusion equation for the magnetization by using a generalization of the Boltzmann equation approach developed by Lhuillier and Laloë.[8]

2. SPIN 1 BOLTZMANN EQUATION

In contrast to the usual derivation of spin waves for a degenerate Fermi liquid in an applied magnetic field,[9] which is based on a transport equation that neglects the collision term, we find that we must keep the collision term in the Boltzmann equation for a non-degenerate gas in order to generate the modifications of the spin current that lead to spin waves, as will be shown *ut infra*. The derivation of the Boltzmann equation with the appropriate collision term may be found elsewhere.[8] Some comments on the nature of the derivation are relevant, however. In deriving the quantum-mechanical two body cross-section, Lhuillier and Laloë[8] made no detailed assumptions about the nature of the spin structure of the colliding atoms. Hence, the equation that they derived may be applied to systems of arbitrary spin, once the appropriate spin-density matrix has been chosen.

The non-linear character of the Boltzmann equation makes it intractable, unfortunately, so that a well-defined approximation scheme must be used. Basically, Lhuillier and Laloë develop an expansion in l/L—the Chapman-Enskog approximation. The density matrix within this approximation becomes the sum of two parts: the equilibrium term and the first order correction. The correction term has a simple physical interpretation. In any case, the linearized Boltzmann equation, following the notation of Lhuillier and Laloë,[8] becomes:

$$\frac{d}{dt}\left(f_0(\mathbf{r}, \mathbf{p})\delta\bar{\rho}_s(\mathbf{r}, \mathbf{p})\right) + \frac{1}{m}\mathbf{p} \cdot \operatorname{grad}(\rho_s^0(\mathbf{r}, \mathbf{p}))$$

$$= f_0(\mathbf{r}, \mathbf{p}) I_{\text{coll}}(\delta\bar{\rho}_s(\mathbf{r}, \mathbf{p})), \tag{5}$$

where the density matrix, $\rho_s(\mathbf{r}, \mathbf{p}) \approx \rho_s^0(\mathbf{r}, \mathbf{p}) + \delta\rho_s(\mathbf{r}, \mathbf{p})$. The bar in Eq. (5) indicates that the Boltzmann distribution, $f_0(\mathbf{r}, \mathbf{p})$, has been factored out. Finally, note that the collision term is a matrix function of $\delta\bar{\rho}_s^0$ (cf. Eq (7)).

In order to calculate the effect of spin transport, we shall assume that spin transport reestablished the equilibrium momentum distribution. Under that assumption, $\delta\bar{\rho}_s$ depends only on the momentum:

$$\delta\bar{\rho}_s = \delta\bar{\rho}_s(\mathbf{p}) \tag{6}$$

Furthermore, our assumptions are consistent with associating a spin current with $\delta\rho$, os that we may take $\delta\bar{\rho}_s$ to be linear in p. The collision term under those approximations becomes

$$I_{\text{coll}}(\delta\bar{\rho}_s) = \int d^3q' \frac{q'}{m} \int d^2\hat{q} f_0(\mathbf{p}_2) \left\{ \sigma_k(\theta) \frac{1}{2} \delta\bar{\rho}_s(\mathbf{q}'-\mathbf{q}) + \frac{i\varepsilon}{2} \tau_k^{ex}(\theta)[\bar{\rho}_s^0, \delta\bar{\rho}_s(\mathbf{q}')] \right\}$$
$$+ \frac{i\varepsilon}{2} \int d^3q \frac{q}{m} \tau_{fwd}^{\text{ex}}(k) f_0(\mathbf{p}-\mathbf{q})[\bar{\rho}_s^0, \delta\bar{\rho}_s(\mathbf{q}')], \tag{7}$$

where the σ and τ cross-sections are defined in Ref. 8. The term in τ_{fwd}^{ex} is one of the contributions to the μ coefficient, which will be shown to be related to the spin wave 'quality factor' *ut infra.*

Once the commutators in the collision integral have been evaluated, all that is left are the momentum integrals, Eqs. (27) and (28) We shall see that they are related to integrals that have already been evaluated for H↓↓, ^{3}He↓, and D↓↓.[10]

3. SPIN 1 DENSITY MATRIX

For the case of spin 1/2, the spin-density matrix is

$$\rho_s = \frac{1}{2}(\mathbf{1}_2 + \boldsymbol{\sigma} \cdot \mathbf{M}(\mathbf{r})), \tag{8}$$

where $\mathbf{1}_2$ is the unit matrix in two dimensions, $\boldsymbol{\sigma}$ is the 'vector' of Pauli matrices, and $\mathbf{M}$ is the nuclear spin polarization vector.

For a spin 1 system, for which an interaction with an applied electric field gradient is possible, one must include additional terms in the density matrix proportional to spherical tensors of rank two. In order to facilitate comparison with Buckle's results, we will use his definitions.[4] A convenient form is:

$$\hat{Q}^0 = \frac{1}{\sqrt{6}}(3\hat{I}_0^2 - \mathbf{I}^2) \tag{9}$$

$$\hat{Q}^{\pm 1} = \frac{1}{\sqrt{2}} (\hat{I}^{\pm} \hat{I}^{0} + \hat{I}^{0} \hat{I}^{\pm}) \tag{10}$$

$$\hat{Q}^{\pm 2} = \frac{1}{2} [\hat{I}_x^2 - \hat{I}_y^2 \pm i(\hat{I}_x \hat{I}_y + \hat{I}_y \hat{I}_x)], \tag{11}$$

where **I** is the 'vector' of spin 1 angular momentum operators, and the caret indicates an operator. One finds

$$\hat{I}^{\pm} = \mp \frac{1}{\sqrt{2}} (\hat{I}_x \pm i\hat{I}_y) \tag{12}$$

$$\hat{I}^{0} = \hat{I}^{z} \tag{13}$$

Using the trace methods outlined in the appendix, one may write the spin density matrix for a spin 1 system in terms of an expansion in irreducible tensor operators, *viz.*

$$\rho_s = \tfrac{1}{3}\mathbf{1}_3 + \tfrac{1}{2}\mathbf{I} \cdot \mathbf{M}(\mathbf{r}) + \mathcal{Q} \cdot \mathcal{A}(\mathbf{r}), \tag{14}$$

where $\mathbf{1}_3$ is the three dimensional unit matrix, **M** is the nuclear spin polarization vector and $\mathcal{A}$ is the nuclear quadrupolar alignment tensor. The quantity $\mathcal{Q} \cdot \mathcal{A}$ is the rank two analogue of $\mathbf{I} \cdot \mathbf{M}$. One may define $\mathcal{A}$ as follows:

$$\mathcal{A}^q = \mathrm{Tr}\{\hat{Q}^q \rho_s\}, \tag{15}$$

where $\hat{Q}^q$ is one of the operators listed in Eq. (9)-(11). In terms of the contraction convention:

$$\mathbf{T}(1) \cdot \mathbf{T}(2) = \sum_{q=-k}^{q=k} (-1)^q T_k^q(1) T_k^{-q}(2), \tag{16}$$

where **T** is a spherical tensor of rank k, one may write

$$\mathcal{Q} \cdot \mathcal{A} = \sum_{q=-2}^{q=2} (-1)^q \hat{Q}^q A^{-q} \tag{17}$$

Let us choose a trial form of $\delta\bar{\rho}_s$ that is traceless and linearly dependent on momentum. One may write:

$$\delta\bar{\rho}_s(\mathbf{p}) = \tfrac{1}{2} \sum_{q''=-1}^{1} (-1)^{q''} (\mathbf{c}^1_{-q''} \cdot \mathbf{p}) \hat{S}_{q''} + \sum_{q'=-2}^{2} (-1)^{q'} (\mathbf{c}^2_{-q'} \cdot \mathbf{p}) \hat{Q}_{q'}, \tag{18}$$

where the **c** vectors will be shown to be related to the spin currents that are generated by the equilibrium restoring relaxation processes. We shall assume that the **c** vectors are momentum independent.

In order to get a feel for the physical meaning of the coefficients $\mathbf{c}_q^k$ in $\delta\bar{\rho}_s$, let us calculate the spin current associated with $\delta\bar{\rho}_s$, viz.

$$\begin{aligned}\mathbf{J}(\hat{S}_i) &\equiv \int d^3p\, f_0(\mathbf{p})\, \frac{\mathbf{p}}{m}\, [\mathrm{Tr}\{\hat{S}_i \delta\bar{\rho}_s\}] \\ &= \int d^3p\, f_0(\mathbf{p})\, \frac{\mathbf{p}}{m}\, \mathbf{p}\cdot\mathbf{c}^1_{-j}\, \frac{1}{2}(-1)^j Tr\{\hat{S}_i\hat{S}_j\} \\ &= \frac{1}{\beta}\mathbf{c}_i^1, \end{aligned} \tag{19}$$

similarly

$$\mathbf{J}(\hat{Q}_i) = \frac{1}{\beta}\mathbf{c}_i^2, \tag{20}$$

where we have used the summation convention and the trace relations in the appendix. Here, $1/\beta$ is k_BT, $\mathbf{J}(\mathbf{S})$ is the spin current associated with the nuclear polarization, $\mathbf{M}$, and $\mathbf{J}(\mathscr{Q})$ is the spin current associated with the nuclear quadrupolar alignment, $\mathscr{A}$.

This simple example illustrates a technique that we will use to evaluate the spin current due to the drift and collision terms in the kinetic equations. We operate on both sides of Eq. (5) in a generalized ket notation, viz.

$$\left\langle\!\left\langle \mathbf{p}, \hat{O} \left| \frac{\partial\delta\bar{\rho}_s}{\partial t} \right\rangle\!\right\rangle + \left\langle\!\left\langle \mathbf{p}, \hat{O} \left| \frac{1}{m}\mathbf{p}\cdot\mathrm{grad}\,\bar{\rho}_s \right\rangle\!\right\rangle = \ll \mathbf{p}, \hat{O} \,|\Gamma_{\mathrm{coll}}\delta\bar{\rho}_s \gg, \right. \tag{21}$$

where $\ll \mathbf{p}\hat{O}| \equiv \mathbf{p}\hat{O}$, an outer product of the momentum $\mathbf{p}$ and the spin operator $\hat{O}$, and the notation $\ll \cdot|\cdot \gg$ indicates a trace over spin operators and an integral over all momenta weighted by $f_0(\mathbf{p})$.

In the rotating frame approximation, we transform away the first term on the left hand side of Eq. (21). In Sec. 6, however, we shall examine the effect of a finite magnetic field gradient, which will require that we include the effect of a position dependent Larmor frequency in Eq. (21), because it is experimentally relevant. The finite gradient case is the analogue of spin wave spectroscopy[5] in H↓↓, which is performed in a magnetic field gradient.

4. SOLUTION OF THE BOLTZMANN EQUATION

When evaluating the collision term in Eq. (21), an examination of the collision integral in Eq. (7) shows that there will be $\mathbf{p}$-dependent factors in the integrand of the form

$$\mathrm{Tr}\{\hat{I}^q\delta\bar{\rho}_s\} = \mathbf{c}_q^1\cdot\mathbf{p} \tag{22}$$

$$\mathrm{Tr}\{\hat{Q}^q\delta\bar{\rho}_s\} = \mathbf{c}_q^2\cdot\mathbf{p}, \tag{23}$$

that are present in addition to the **p**-dependent cross-sections. In order to evaluate $[\bar{\rho}_s^0, \delta\bar{\rho}_s]$, we shall make the local frame approximation:

$$\mathbf{M} = M_0\hat{z} + \tilde{\mathbf{M}}(\mathbf{r}), \tag{24}$$

$$\mathscr{A} = A_0\hat{Q}_0 + \tilde{\mathscr{A}}(\mathbf{r}), \tag{25}$$

where $|\mathbf{M}| \approx M_0$ and $|\mathscr{A}| \approx A_0$. In this approximation we are limited to small gradients of magnetization, consistent with a small tipping angle NMR pulse, for example. In the local frame approximation, therefore, the commutator terms in the collision integral may be rewritten as

$$[\bar{\rho}_s^0, \delta\bar{\rho}_s] = \tfrac{1}{2}M^0[\hat{I}^0, \delta\bar{\rho}_s] + A^0[\hat{Q}^0, \delta\bar{\rho}_s], \tag{26}$$

where we have assumed that $\bar{\rho}_s^0$ is of the form of Eq. (14). We shall drop the tildes over the quantities $\mathbf{M}(\mathbf{r})$ and $\mathscr{A}(\mathbf{r})$ in the sequel. Inserting Eq. (7) into Eq. (21), we may rewrite the collision term of Eq. (21) in terms of the momentum integrals

$$X_1 = -\frac{8}{3}\frac{m}{\beta}\Omega^{(1,1)}_{[\sigma_k]}, \tag{27}$$

and

$$X_4 = -\frac{8}{3}\frac{m}{\beta}(\Omega^{(1,1)}_{[\tau^{ex}]} + \Xi^{(1)}_{[\tau^{ex}_{fwd}]}), \tag{28}$$

in the notation of Ref. 7 where

$$\Omega^{(1,1)}_{[\sigma_k]} = \frac{1}{\sqrt{\pi m\beta}}\int_0^\infty d\gamma\, e^{-\gamma^2}\gamma^5 Q^{(1)}_{[\sigma_k]}(k), \tag{29}$$

$$\Omega^{(1,1)}_{[\tau^{ex}_k]} = \frac{1}{\sqrt{\pi m\beta}}\int_0^\infty d\gamma\, e^{-\gamma^2}\gamma^5 \tilde{Q}^{(1)}_{[\tau^{ex}]}(k), \tag{30}$$

$$\Xi^{(1)}_{[\tau^{ex}_{fwd}]} = \frac{1}{\sqrt{\pi m\beta}}\int_0^\infty d\gamma\, e^{-\gamma^2}\gamma^5 \tau^{ex}_{fwd}(k), \tag{31}$$

and $k = (\sqrt{m/\beta})\gamma/\hbar$. The Q integrals are the angle-averaged cross-sections defined as follows

$$Q^{(1)}_{[\sigma_k]} = 2\pi\int_0^\pi \sin\vartheta\, d\vartheta(1-\cos\vartheta)\sigma_k(\vartheta), \tag{32}$$

and

$$\tilde{Q}^{(1)}_{[\tau^{ex}]}(k) = 2\pi \int_0^{\pi} \sin\vartheta \, d\vartheta \cos\vartheta \tau^{ex}(k) \tag{33}$$

These integrals have been evaluated[10] for spin-polarized ^{3}He, hydrogen and deuterium. The spin transport parameters are defined by the following quantities

$$nD_0 = -\frac{1}{\beta^2 X_1} = \frac{3}{8}\frac{1}{m\beta}\frac{1}{\Omega^{(1,1)}_{[\sigma_k]}} \tag{34}$$

$$\mu = \frac{X_4}{X_1} = \frac{\Omega^{(1,1)}_{[\tau^{ex}_k]} + \Xi^{(1)}_{[\tau^{ex}_{fwd}]}}{\Omega^{(1,1)}_{[\sigma_k]}}, \tag{35}$$

where n is the number density, D_0 is the longitudinal diffusion constant, and μ is a measure of the importance of exchange scattering to direct scattering. The quantities μ and nD_0 are plotted in Figs. 5 and 6 for spin polarized hydrogen and deuterium.

The dramatic upturn in μ at low temperatures for spin polarized deuterium is due to details of the atom–atom interaction potential. In the case of hydrogen, which has a larger zero-point energy than deuterium due to its lower mass, the minimum in the atom–atom potential is not as

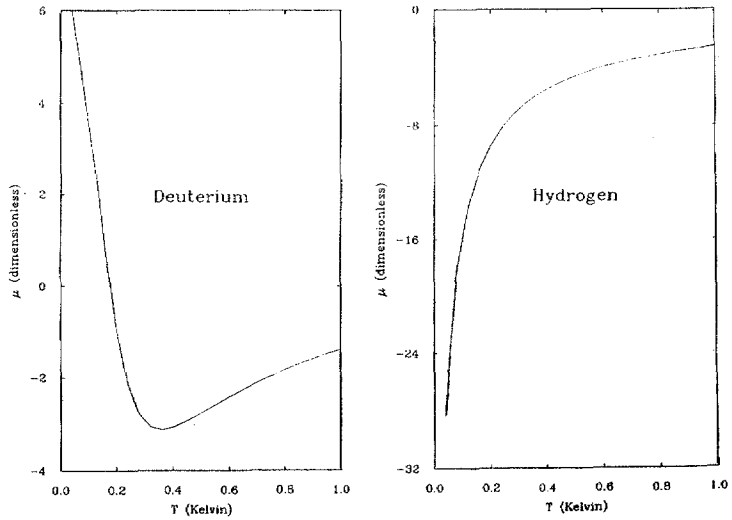

Fig. 5. The μ factor is plotted as a function of temperature for hydrogen and deuterium from 40 mK to 1 K. The difference in the low-temperature behavior of the two curves is due to the attractive interaction between two deuterium atoms, which becomes increasingly important at low temperatures. See text for discussion.

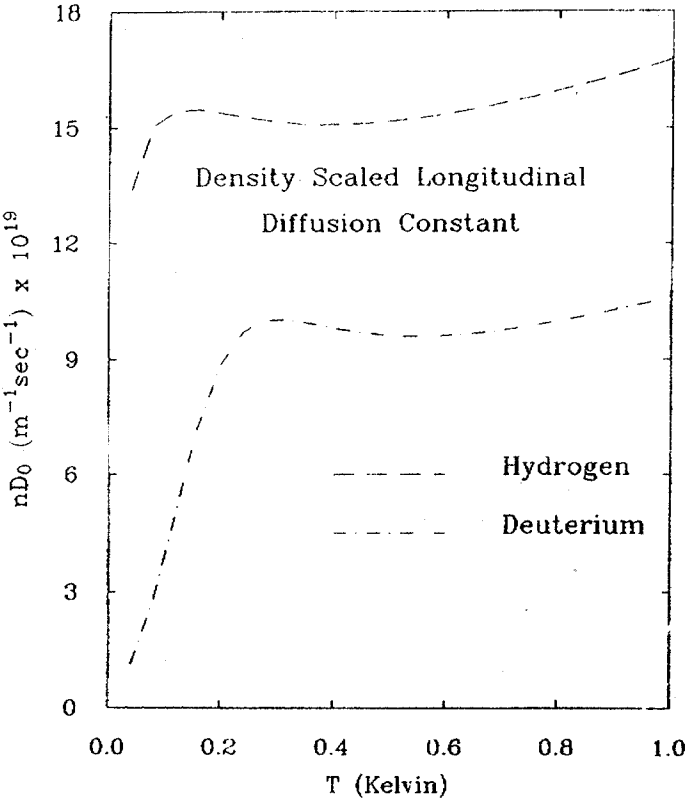

Fig. 6. The longitudinal diffusion constant, scaled by the density, is plotted as a function of temperature for hydrogen and deuterium from 40 mK to 1 K.

significant as the repulsive part of the triplet potential. Consequently, the repulsive triplet potential dominates at all temperatures for spin polarized hydrogen, so that μ decreases monotonically to $-\infty$ as one lowers the temperature. In the case of deuterium, however, the reduced zero point motion allows the atoms to sample the attractive part of the potential as one lowers the temperature, which causes μ to increase monotonically at sufficiently low temperatures. The point we wish to emphasize is that the different behavior of μ for hydrogen and deuterium results from differences in the atom-atom potential, not the different statistics.

Using the trace relations (73)-(79) and Eqs. (34) and (35), we may solve for the **c** vectors. We find

$$\frac{\mathbf{c}_{\pm 2}^2}{\beta} = -\frac{D_0}{1+\mu^2(2M_0)^2}(\mp i\varepsilon\mu(2M_0))\,\mathrm{grad}\,A_{\mp 2} \qquad (36)$$

$$-D_0 \operatorname{grad}\begin{pmatrix} A_\pm \\ M_\pm \end{pmatrix} = \begin{pmatrix} 1 \pm i\varepsilon\mu M_0 & \pm i\varepsilon\mu 2\sqrt{3}A_0 \\ \pm i\varepsilon\mu\sqrt{3}A_0 & 1 \pm i\varepsilon\mu M_0 \end{pmatrix}\begin{pmatrix} \mathbf{c}_\pm^2 \\ \mathbf{c}_\pm^1 \end{pmatrix}\frac{1}{\beta} \tag{37}$$

$$\frac{\mathbf{c}_0^2}{\beta} = -D_0 \operatorname{grad} A_0 \tag{38}$$

$$\frac{\mathbf{c}_0^1}{\beta} = -D_0 \operatorname{grad} M_0. \tag{39}$$

Recall that $\mathbf{c}_q^k/\beta = \mathbf{J}_q^k$, where $\mathbf{J}_q^k$ is the current associated with the expectation value of the operator $\hat{O}_q^k$. One can eliminate the currents by operating on both sides of Eqs. (36)–(39) with the operator $-\operatorname{div}$ and using the continuity equations $\operatorname{div} \mathbf{J}_q^k = -\partial/\partial t\langle \hat{O}_k^q \rangle$. The continuity equations incorporate the physical assumption that the spin currents drive the system back to equilibrium. The quantities M_0 and A_0 are constant in the local frame approximation and are unaffected by the $-\operatorname{div}$ operator. One finds after a simple calculation

$$\frac{\partial A_{\mp 2}}{\partial t} = \frac{D_0}{1+\mu^2(2M_0)^2}(1 \mp i\varepsilon\mu(2M_0))\Delta A_{\pm 2} \tag{40}$$

$$-D_0\Delta\begin{pmatrix} A_\pm \\ M_\pm \end{pmatrix} = \begin{pmatrix} 1 \pm i\varepsilon\mu M_0 & \pm i\varepsilon\mu 2\sqrt{3}A_0 \\ \pm i\varepsilon\mu\sqrt{3}A_0 & 1 \pm i\varepsilon\mu M_0 \end{pmatrix}\frac{\partial}{\partial t}\begin{pmatrix} A_\pm \\ M_\pm \end{pmatrix}, \tag{41}$$

as well as:

$$\frac{\partial A_0}{\partial t} = D_0\Delta A_0 \tag{42}$$

$$\frac{\partial M_0}{\partial t} = D_0\Delta M_0, \tag{43}$$

where Δ is the Laplacian operator.

5. DISCUSSION

We shall assume plane-wave solutions for all of the equations of the form

$$\hat{O}(\mathbf{r}, t) = e^{-i\omega t}\, e^{i\mathbf{k}\cdot\mathbf{r}}\hat{O} \tag{44}$$

This choice leads to dispersion relations for $A_{\pm 2}$, A_0 and M_0 of the form

$$-i\omega = -D_0 k^2 \quad \text{for } A_0 \quad \text{and} \quad M_0 \tag{45}$$

$$-i\omega_{\pm 2} = -\frac{D_0 k^2}{1+\mu^2(2M_0)^2}(1 \mp i\varepsilon\mu(2M_0)) \quad \text{for} \quad A_{\pm 2} \tag{46}$$

Note that A_0 and M_0 relax towards their equilibrium values on a time scale given by $1/D_0k^2$. The components $A_{\pm 2}$ have damped sinusoidal solutions where the 'quality factor' of the oscillation may be written as

$$Q_{\pm 2} = \left|\frac{\Re\{\omega_{\pm 2}\}}{\Im\{\omega_{\pm 2}\}}\right| = |2\mu M_0|. \tag{47}$$

These transitions correspond to two-quantum transitions. One could observe such transitions by performing a longitudinal resonance experiment in low magnetic fields. The signal to noise of such an experiment would be low, however, because the deuterium quadrupole moment is small. Moreover, the off-diagonal terms of the quadrupolar coupling to the electric field gradient, which are necessary for the observation of the longitudinal resonance, will cause the $|\gamma\rangle$ state to have an admixture of the $|\alpha\rangle$ state. The problem with the $|\alpha\rangle$ state is that it has an admixture of electron spin up, due to the hyperfine interaction. Thus, the quadrupole terms will cause the $|\gamma\rangle$ state to have an admixture of electron spin up which will reduce the sample lifetime and the nuclear polarization. We will concentrate on the one quantum transitions, therefore, which should be much easier to observe experimentally.

The coupled modes are more complex. We shall discuss the eigenvalues in terms of the quantity $M_0 \pm \sqrt{6}A_0$. Using the properties of the operators given in the appendix, one may show

$$\frac{1}{N}\sum_{I=\alpha,\beta,\gamma}\langle I|\hat{I}^0+\sqrt{6}\hat{Q}^0|I\rangle = 2\frac{N_\alpha - N_\beta}{N} = M_0+\sqrt{6}A_0, \tag{48}$$

$$\frac{1}{N}\sum_{I=\alpha,\beta,\gamma}\langle I|\hat{I}^0-\sqrt{6}\hat{Q}^0|I\rangle = 2\frac{N_\beta - N_\gamma}{N} = M_0-\sqrt{6}A_0, \tag{49}$$

where the sum is over all particles and N is the total number of particles. The ket, $|I\rangle$, is a nuclear pseudo-spin eigenstate and N_ν is the number of atoms in the state $|\nu\rangle$.

In this paper we are interested in those systems that have a polarization $M_0 \approx -1$, or $N_\gamma/N \approx -1$. In order to observe a non-vanishing transverse magnetization, therefore, we must calculate M^+ and A^+, which depend on raising operators. In that case, the only non-vanishing eigenvalue of Eq. (41) is

$$\lambda_+^- = 1 + i\varepsilon\mu(M_0 - \sqrt{6}A_0) \tag{50}$$

The '−' superscript indicates that we are interested in the $|\gamma\rangle - |\beta\rangle$ transition, which is described by the operator, u_-, defined in the appendix. Using the results derived in the appendix, one may show that u_- may be expressed in terms of the spherical tensor operators as

$$u_- = \left(\frac{1}{\sqrt{2}}\hat{Q}^+ - \frac{1}{2}\hat{I}^+\right) \tag{51}$$

A more thorough discussion of the transition operators is given in the appendix.

The equation that we must now solve is

$$D_0\Delta\langle u_-\rangle = \lambda_+^- \frac{\partial}{\partial t}\langle u_-\rangle, \tag{52}$$

where $\langle u_-\rangle$ is the expectation value of u_-. Just as in the two-quantum transitions, one may assume plane wave solutions, which leads to a dispersion relation of the form

$$-i\omega_+^- = -\frac{D_0 k^2}{\lambda_+^-}, \tag{53}$$

where the 'quality factor' of the underdamped diffusion mode is given by

$$\begin{aligned} Q_{\gamma\beta} &= \left|\frac{\Re\{\omega_+^-\}}{\Im\{\omega_+^-\}}\right| \\ &= |\mu(M_0 - \sqrt{6}A_0)| \end{aligned} \tag{54}$$

Note that the quantity $M_0 - \sqrt{6}A_0 \approx -2$ for a sample in which $|\gamma\rangle$ is the only significantly populated state. The μ parameter has a minimum at -3 in the experimentally accessible temperature régime. Hence, the sharpest observable modes corresponding to the $\gamma - \beta$ transition will have a quality factor $Q_{\gamma\beta} \approx 6$, which is comparable to the spin polarized hydrogen case.

6. FINITE GRADIENTS

In Sec. 2, we mentioned that a situation of great experimental interest occurs when the sample is in a finite field gradient. We are now in a position to make that statement more precise. There are two modifications that one must make to the Boltzmann equation in the presence of an applied external magnetic field.[8] First, one must add a precession term, $[\rho_s, \mathbf{I}\cdot\mathbf{B}]/i\hbar$. Second, one must add a 'Stern-Gerlach' term given by the anticommutator $[\partial\rho/\partial p_i, \partial(\mathbf{B}\cdot\mathbf{I})/\partial x_i]_+$. Using the equipartition theorem, which is valid in the non-degenerate régime, and the observation that a characteristic length over which $\mathbf{B}$ might vary is of the order of the sample cell size, L, we may

say that the 'Stern-Gerlach' term is of order $[\rho, \mathbf{B}\cdot\mathbf{I}]_+/L\sqrt{mk_BT}$. Hence, the ratio of the precession term to the 'Stern-Gerlach' term is of order $L\sqrt{mk_BT}/\hbar$. If we take T on the order of 100 mK, and L on the order of 1cm, then the ratio is of order 10^{18} and we may neglect the 'Stern-Gerlach' term.

We shall assume that the magnetic induction, $\mathbf{B}$, is given by

$$\mathbf{B} = B_0\hat{z} + (\mathbf{G}\cdot\mathbf{r})\hat{z}, \tag{55}$$

where $\mathbf{G}\cdot\mathbf{r}$ is of the order of a few gauss and B_0 is taken as, say, 10^5 gauss. Buckle[4] has treated the case of free precession in the absence of a field gradient in some detail. We are interested in the case of a finite gradient. In the local frame approximation of Sec. 4, we find that the continuity equation is modified in the presence of an external field.[11] In the Larmor frame defined by $\hbar\omega_0 = \gamma B_0$, we find

$$\frac{\partial \tilde{M}_\pm}{\partial t} \pm i\gamma\mathbf{G}\cdot\mathbf{r}\tilde{M}_\pm = 0 \tag{56}$$

$$\frac{\partial \tilde{A}_\pm}{\partial t} \pm i\gamma\mathbf{G}\cdot\mathbf{r}\tilde{A}_\pm = 0 \tag{57}$$

$$\frac{\partial \tilde{A}_{\pm 2}}{\partial t} \pm 2i\gamma\mathbf{G}\cdot\mathbf{r}\tilde{A}_{\pm 2} = 0, \tag{58}$$

where we have ignored the spin-flux term, div $\cdot\,\mathbf{J}$, for the moment. The components $\tilde{M}_0$ and $\tilde{A}_0$ are unaffected by external field gradients along the z direction, because they are assumed to be aligned along the external magnetic field B_0.

Using the assumptions discussed in Sec. 5, we may write the equations of motion for the two-quantum transitions and the $\gamma-\beta$ transition as

$$\left(\frac{\partial}{\partial t} \pm 2i\gamma\mathbf{G}\cdot\mathbf{r}\right)\tilde{A}_{\pm 2} = \frac{D_0}{1+\mu^2(2M_0)^2}(\mp i\varepsilon\mu(2M_0))\Delta\tilde{A}_{\pm 2} \tag{59}$$

$$\left(\frac{\partial}{\partial t} + i\gamma\mathbf{G}\cdot\mathbf{r}\right)\langle u_-\rangle = \frac{D_0}{\lambda_+^-}\Delta\langle u_-\rangle. \tag{60}$$

In order to interpret the equations of motion we have written down, let us first consider the $A_{\pm 2}$ modes in more detail. We shall assume that the time dependence is given by $e^{-i\omega_{\pm 2}^n t}$, where we anticipate the existence of a spectrum of eigenvalues labelled by n. If we multiply equation 59 by i and choose the upper sign, we find

$$\omega_{+2}^n A_{+2} = -\frac{D_0}{1+\mu^2(2M_0)^2}(-\varepsilon\mu(2M_0) - i)\frac{\partial^2}{\partial z^2}A_{+2} + 2\gamma G_z A_{+2}, \tag{61}$$

where we have limited attention to the $|\gamma\rangle - |\alpha\rangle$ transition and have assumed that the only significant spatial dependence is in the z direction. We have also assumed that the geometry of the sample cell allows us to use separation of variables.

In order to make further progress, we define the length, $l_\pm$, by

$$\frac{1}{l_\pm^3} = \frac{2\gamma G(1+\mu^2(2M_0)^2)}{\pm D_0(\varepsilon\mu(2M_0)+i)}. \tag{62}$$

Similarly,

$$\frac{\alpha_n}{l_\pm^2} = \frac{\omega_{+2}^n(1+\mu^2(2M_0)^2)}{D_0(\varepsilon\mu(2M_0)+i)}, \tag{63}$$

where α_n is determined below. We shall also define the dimensionless length, $\zeta_\pm$, as follows

$$\zeta_\pm = \frac{z}{l_\pm} + \alpha_n \tag{64}$$

In terms of these quantities, we may rewrite Eq. (61) in the much simpler form

$$\frac{d^2}{d\zeta_-^2} A_{+2} - \zeta_- A_{+2} = 0, \tag{65}$$

where the minus sign obtains for the case $\varepsilon\mu(2M_0) < 0$. The solutions of Eq. (65) are Airy functions[12] of complex argument. We may write an integral representation as follows

$$u_n(\zeta_-) = \frac{N_n}{\pi} \int_0^\infty d\eta \cos\left(\eta\zeta_- + \frac{1}{3}\eta^3\right) = N_n Ai(\zeta_-), \tag{66}$$

where N_n is a normalization constant.

We must now choose boundary conditions for $u_n(\zeta_-)$. We assume that there is a perfectly reflecting wall at $z = 0$. We also assume that the solution at the opposite wall, $u_n(z = L)$, has essentially decayed to zero, which means that the boundary condition at $z = 0$ is the only important one. This situation obtains when $\gamma GL > \Re\{\omega_{+2}^n\}$. The reflecting boundary condition in one dimension is

$$\left.\frac{\partial u_n}{\partial \zeta_-}\right|_{\zeta_- = \alpha_n} = 0, \tag{67}$$

where $\{\alpha_n\}$ are the roots of $u_n'(\zeta_-) = 0$. Tables of α_n and asymptotic expressions for α_n for large values of n may be found in the literature.[12]

We may rewrite Eq. (65) in terms of the definition of l_-. This gives us an eigenvalue equation, using the known roots, $\{\alpha_n\}$. We find

$$\omega_{+2}^n = -\alpha_n \left(-(2\gamma G)^2 D_0 \frac{(\varepsilon\mu(2M_0)+i)}{1+\mu^2(2M_0)^2} \right)^{1/3} \tag{68}$$

We have tacitly assumed that $\varepsilon\mu(2M_0)$ is negative. This is valid for D$\downarrow\downarrow$ in the quasi-equilibrium case.

If we make M_0 positive (following a π-pulse, say), then we must modify the eigenvalue equation and choose a new origin for z. The procedure is straightforward.[13] We find

$$\omega_{+2}^n = \begin{cases} -\alpha_n \left(-(2\gamma G)^2 D_0 \dfrac{(\varepsilon\mu(2M_0)+i)}{1+\mu^2(2M_0)^2} \right)^{1/3} & \text{for} \quad \varepsilon\mu(2M_0) < 0 \\ \\ \alpha_n \left((2\gamma G)^2 D_0 \dfrac{(\varepsilon\mu(2M_0)+i)}{1+\mu^2(2M_0)^2} \right)^{1/3} & \text{for} \quad \varepsilon\mu(2M_0) > 0, \end{cases} \tag{69}$$

and the new origin of z is the point $z = L$ in the old coordinate system.

Now that we have solved the equation of motion for A_{+2} in a field gradient, we can extend the discussion to $\langle u_- \rangle$ without difficulty. The only modifications to the treatment given *ut supra* that we need to make are to replace $2\gamma G$ by γG and $2M_0$ by $M_0 - \sqrt{6}A_0$. The eigenvalues then become

$$\omega_{\gamma\beta}^n = \begin{cases} -\alpha_n \left(-(\gamma G)^2 D_0 \dfrac{(\varepsilon\mu(M_0-\sqrt{6}A_0)+i)}{1+\mu^2(M_0-\sqrt{6}A_0)^2} \right)^{1/3} & \text{for the case:} \\ \varepsilon\mu(M_0-\sqrt{6}A_0) < 0; & \\ \\ \alpha_n \left((\gamma G)^2 D_0 \dfrac{(\varepsilon\mu(M_0-\sqrt{6}A_0)+i)}{1+\mu^2(M_0-\sqrt{6}A_0)^2} \right)^{1/3} & \text{for the case:} \\ \varepsilon\mu(M_0-\sqrt{6}A_0) > 0. & \end{cases} \tag{70}$$

In order to calculate a line-shape, we need to know the 'coupling strength' or 'transition moment' for each spin-wave mode. The line-shape is then the sum of the line-shapes of each spin-wave mode weighted by the 'transition moment'. We have assumed that the time dependence of each mode is an exponentially damped sinusoid; hence, the sum is a series of unequally weighted Lorentzian lines, where each spin-wave mode has a shift given by $\Re\{\omega_{\gamma\beta}^n\}$ and a width given by $\Im\{\omega_{\gamma\beta}^n\}$. The 'transition moment' is[13]

$$d_n = \frac{\gamma}{V} \int_V u_n(\zeta) H_1(z)\, dV \tag{71}$$

$$= \frac{\gamma H_1}{L} \int_0^L u_n(\zeta)\, dz, \tag{72}$$

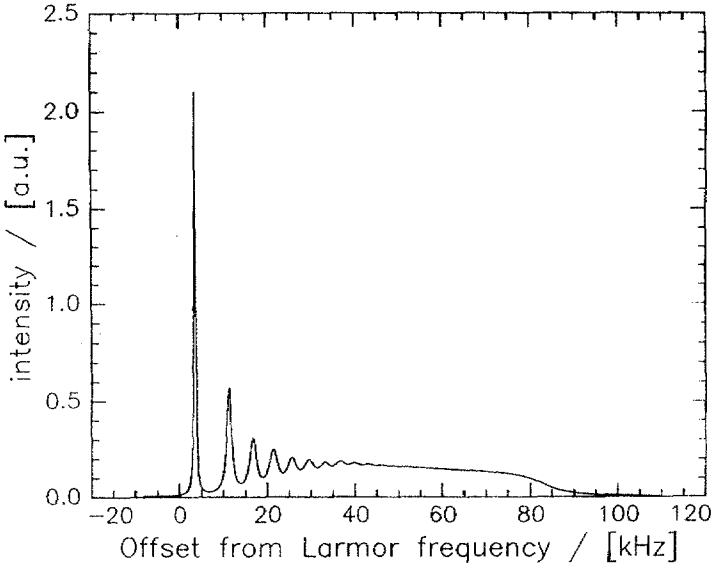

Fig. 7. A model spectrum for the $\gamma-\beta$ transition in D$\downarrow\downarrow$. The spectrum shows only the lowest order spin-waves, which are strongly 'trapped' in the one-dimensional model calculation.

where the three dimensional integral over the sample volume collapses to an integral over the only relevant dimension in our one-dimensional model.

In Fig. 7 we plot a model spectrum for the $\gamma-\beta$ transition with values of μ and nD_0 appropriate for 300 mK. If one compares the model spectrum with published data[14] for H$\downarrow$, one sees that the large-n spin wave modes of D$\downarrow\downarrow$ are shifted up in frequency vis-à-vis the sharp, small n modes. In H$\downarrow\downarrow$, the large-n spin-wave modes are shifted down in frequency vis-à-vis the sharp, small-n modes. The difference is due to the relative minus sign between the effective field in H$\downarrow\downarrow$, which behaves as a bosonic system with $\varepsilon=+1$, and D$\downarrow\downarrow$, which behaves as a fermionic system with $\varepsilon=-1$.

The model parameters used were $nD_0=10^{20}\,m^{-1}s^{-1}$, $\mu(M_0-\sqrt{6}A_0)=+6$, $\varepsilon=-1$, $G=1$ gauss/cm.

7. CONCLUSIONS

We have developed a spin 1 Boltzmann equation and applied it to D$\downarrow\downarrow$ for parameters that are experimentally relevant. Detailed comparisons of published hydrogen spin wave spectra with the simulated deuterium results are not particularly meaningful, however. The hydrogen experimental and theoretical spectra are in a three dimensional space. The deuterium calculation assumes a one dimensional model. What one can say unambiguously

is that the transport theory for spin polarized deuterium predicts resolvable spin wave structure for reasonable experimental conditions of density, field gradient, and diffusion coefficient even though the quantum mechanical transport parameter, μ, is smaller for deuterium than for hydrogen over all of the experimentally relevant temperature régime.[24]

The extension of the model to three dimensions is in principle straightforward. The underlying simplicity of the results is clearest in the one dimensional case, however. The philosophy behind the exhaustive analytical treatment given here is simply that models which one may solve analytically are useful paradigms in a variety of applications, as well as provide clear insights into the nature of the solutions. When experimental deuterium spectra are available for fitting, it will be straightforward to modify the existing hydrogen programs to handle deuterium spectra as well.

The normalization integral is calculated in the appendix following the discussion of spin operators. The program used to compute the model spectra was written in turbo PASCAL and is available from the authors.

APPENDIX

In this appendix we will discuss some useful properties of spherical tensor operators and their representation in terms of the transitions among the spin 1 nuclear manifold of D$\downarrow\downarrow$. Furthermore, we will evaluate the normalization integral for the eigenfunctions of the damped spin wave modes.

A.1. Transition Operators

In order to evaluate the traces that appear in Eq. (21), we note the following useful trace relations:

$$\mathrm{Tr}(\hat{I}^q \hat{I}^{-q'}) = (-1)^q 2\delta_{qq'} \tag{73}$$

$$\mathrm{Tr}(\hat{Q}^q \hat{Q}^{-q'}) = (-1)^q \delta_{qq'} \tag{74}$$

$$\mathrm{Tr}(\hat{I}^{\pm}[\hat{I}^0, \hat{I}^{\mp}]) = \pm 2 \tag{75}$$

$$\mathrm{Tr}(\hat{I}^{\pm}[\hat{Q}^0, \hat{Q}^{\mp}]) = \pm\sqrt{3} \tag{76}$$

$$\mathrm{Tr}(\hat{Q}^{\pm}[\hat{I}^0, \hat{Q}^{\mp}]) = \pm 1 \tag{77}$$

$$\mathrm{Tr}(\hat{Q}^{\pm}[\hat{Q}^0, \hat{I}^{\mp}]) = \pm\sqrt{3} \tag{78}$$

$$\mathrm{Tr}(\hat{Q}^{\pm 2}[\hat{I}^0, \hat{Q}^{\mp 2}]) = \mp 2, \tag{79}$$

where we have used the operators defined in Eqs. (9)-(13). The operators are equivalent to the set used in Buckle's work.[4] We have retained only those trace relations which are useful in the evaluation of Eq. (21) in the local frame approximation.

Before we actually write down a representation of the spherical tensor operators, we shall digress in order to develop a more natural basis for discussing magnetic resonance experiments in D↓↓. In order to do so, we introduce three spinors, $|\alpha\rangle$, $|\beta\rangle$, and $|\gamma\rangle$, which correspond to the projections of the pseudo-spin 1 system defined by the three lowest ground-state deuterium hyperfine levels.

We shall use Gell-Mann's λ matrices as a convenient representation of the transition inducing operators among the pseudo-spin 1 manifold levels. The λ matrices satisfy the following commutation relations

$$[\lambda_k, \lambda_l] = 2if_{klm}\lambda_m, \tag{80}$$

where f_{klm} is anti-symmetric under permutations of its indices. The λ matrices also satisfy anti-commutation relations, viz.

$$[\lambda_k, \lambda_l]_+ = \tfrac{4}{3}\delta_{kl} + 2d_{klm}\lambda_m, \tag{81}$$

where d_{klm} is symmetric under permutations of its indices. The λ matrices are also normalized according to

$$\mathrm{Tr}(\lambda_k\lambda_l) = 2\delta_{kl}, \tag{82}$$

where δ_{kl} is the Kronecker delta. The values of f_{klm} and d_{klm} are given in Itzykson and Zuber.[15]

A set of operators that describes the transitions among the pseudo-spin 1 levels is provided by the Sakata model, which is described in Lipkin.[16] We list the transition operators, their representation in terms of Gell-Mann

TABLE I

Connections between the Various Transition Operators and Their Representations

Transition	Transition operator	Gell-Mann representation	Spherical tensor
$\|\gamma\rangle \to \|\alpha\rangle$	t_+	$\frac{1}{2}(\lambda_1 + i\lambda 2)$	$-\hat{Q}^{+2}$
$\|\alpha\rangle \to \|\gamma\rangle$	t_-	$\frac{1}{2}(\lambda_1 - i\lambda 2)$	$-\hat{Q}^{-2}$
$\|\beta\rangle \to \|\gamma\rangle$	u_+	$\frac{1}{2}(\lambda_6 + i\lambda 7)$	$\frac{1}{2}\hat{I}^- - \frac{1}{\sqrt{2}}\hat{Q}^-$
$\|\gamma\rangle \to \|\beta\rangle$	u_-	$\frac{1}{2}(\lambda_6 - i\lambda 7)$	$-\frac{1}{2}\hat{I}^+ + \frac{1}{\sqrt{2}}\hat{Q}^+$
$\|\beta\rangle \to \|\alpha\rangle$	v_+	$\frac{1}{2}(\lambda_4 + i\lambda 5)$	$-\frac{1}{2}\hat{I}^+ - \frac{1}{\sqrt{2}}\hat{Q}^+$
$\|\alpha\rangle \to \|\beta\rangle$	v_-	$\frac{1}{2}(\lambda_4 - i\lambda 5)$	$\frac{1}{2}\hat{I}^- + \frac{1}{\sqrt{2}}\hat{Q}^-$
	t_0	$\frac{1}{2}\lambda_3$	$\frac{1}{2}\hat{I}^0$
	y_0	$\frac{1}{\sqrt{3}}\lambda 8$	$\sqrt{\frac{2}{3}}\hat{Q}^0$

matrices, and the connection to spherical tensor operators in Table 1. The transition operators also satisfy commutation relations, given in, *e.g.*, *Cahn.*[17]

A.2. Eigenfunction Normalization

The normalization of the eigenfunctions requires that one evaluate a contour integral. It is not convenient to use the standard inner product in order to calculate the normalization constant, because the eigenvalues E_n are complex. Instead of the usual Hermitian bilinear form for constructing scalar products, one may use a symmetric bilinear form, as discussed in Heuvers.[18] In this case, the normalization integral becomes,

$$\int_0^L u_n(z)u_n(z)\,dz = \int_0^L N_n^2\,\mathrm{Ai}^2(\zeta)\,dz. \tag{83}$$

For a given eigenvalue, E_n, integrating from $z=0$ to $z=L$ defines a line segment in the complex-ζ plane. Note, however, that in the limit of large gradients, the eigenfunction has a small modulus at $z=L$. In that case, one may extend the upper limit of integration to $z\to\infty$ without introducing significant error in the evaluation of the integral. At this point, the path of integration makes a small angle with respect to the $\Re(\zeta)$ axis for large values of $\mu(M_0-\sqrt{6}A_0)$. One may close the contour of integration by considering the contour shown in Fig. 8.

The Airy function, $\mathrm{Ai}(\zeta)$, is well behaved in the first quadrant for large values of ζ, so that the arc at infinity makes no contribution to the contour

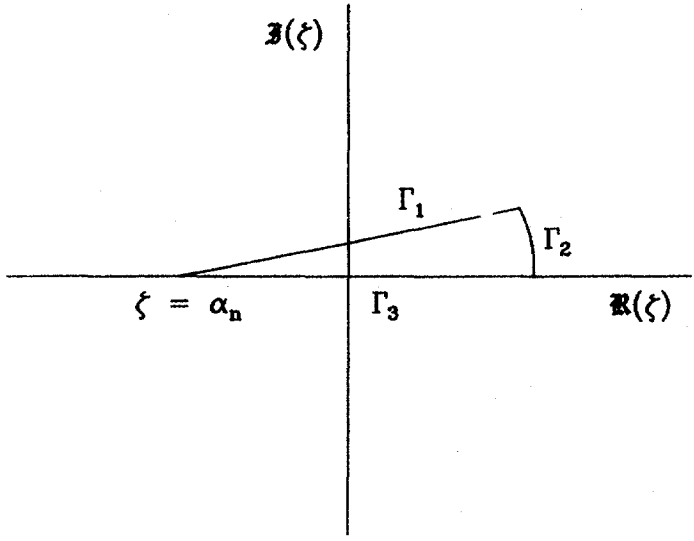

Fig. 8. Closed contour for the evaluation of the normalization integral.

integral. The contour encloses no singularities, which implies that the total integral vanishes by the fundamental theorem of contour integration.[19] Hence, one may write the normalization integral as

$$\int_0^L N_n^2 \operatorname{Ai}^2(\zeta_n)\, dz = \frac{1}{l_-} \int_{\alpha_n}^{C_\infty} N_n^2 \operatorname{Ai}^2(\zeta_n)\, d\zeta$$

$$\int_{\alpha_n}^{C_\infty} N_n^2 \operatorname{Ai}^2(\zeta_n)\, d\zeta = \int_{\Gamma_1} N_n^2 \operatorname{Ai}^2(\zeta_n)\, d\zeta$$

$$= I_1 \tag{84}$$

The integral along Γ_2 vanishes as argued above. If one defines

$$\int_{\alpha_n}^{\infty} N_n^2 \operatorname{Ai}^2(\zeta_n)\, d\zeta = -I_3, \tag{85}$$

then the contour integral, I_Γ, may be written

$$I_\Gamma = I_1 + I_2 + I_3$$

$$= 0, \tag{86}$$

by the fundamental theorem of contour integration; whence,

$$I_1 = -I_3,$$

or

$$\int_0^L N_n^2 \operatorname{Ai}^2(\zeta_n)\, dz = \frac{1}{l_-} \int_{l_-}^{\infty} N_n^2 \operatorname{Ai}^2(\zeta_n)\, d\zeta$$

$$= 1. \tag{87}$$

The problem is now reduced to evaluating the normalization integral along the real axis of the ζ plane. In order to do so, one may use the following trick. If one writes the integrand of the normalization integral as a total derivative of an analytic function, then the integral may be evaluated by evaluating the antiderivative at the endpoints.

Using the defining equation for the Airy function, $\operatorname{Ai}''(\zeta) = \zeta \operatorname{Ai}(\zeta)$, one may show

$$\operatorname{Ai}^2(\zeta) = \frac{d}{d\zeta} [\zeta(\operatorname{Ai}(\zeta))^2 - (\operatorname{Ai}'(\zeta))^2] \tag{88}$$

At the endpoints of the integral, the derivative term vanishes because of the boundary condition. As discussed in Sec. 6, one assumes that there is no spin-current into the walls, which implies $\operatorname{Ai}'(z=0) = \operatorname{Ai}'(z=L) = 0$. At the upper limit, the first term on the right hand side of Eq. (88) vanishes

because $\mathrm{Ai}(z) \to 0$, as $z \to \infty$. Hence, the normalization integral is determined by the values of the Airy function at a discrete set of points on the negative real axis corresponding to the eigenvalues of the problem. One finds,

$$\int_0^L N_n^2 Ai(\zeta_n) dz = \frac{1}{l_-} N_n^2(-\alpha_n)$$

$$= 1,$$

or

$$N_n = \sqrt{l_- \frac{1}{(-\alpha_n)\mathrm{Ai}^2(\alpha_n)}}. \tag{89}$$

In order to estimate the large n behavior of the normalization constants, it is useful to have asymptotic expansions for the roots of $\mathrm{Ai}'(\alpha_n) = 0$, as well as an asymptotic expansion of Ai for large negative real argument. For sufficiently large argument, one may show[20]

$$-\alpha_n \to \left(\frac{3\pi(4n-3)}{8}\right)^{2/3}. \tag{90}$$

Furthermore,[21]

$$\mathrm{Ai}(-z) \to \pi^{-1/2} z^{-1/4} \sin\left(\frac{2}{3}(z)^{3/2} + \frac{\pi}{4}\right) \tag{91}$$

Note that the boundary condition, $\mathrm{Ai}'(\alpha_n) = 0$, ensures that the Airy function is always at an extremum at $z = 0$. One may therefore approximate $\mathrm{Ai}(\alpha_n) \to \pm\pi^{-1/2}(-\alpha_n)^{-1/4}$ without introducing significant error in the asymptotic evaluation of $\mathrm{Ai}(\alpha_n)$. Putting the pieces together, one sees that the large n behavior of N_n is given by the following approximate formula

$$N_n \to \sqrt{\pi l_-}\left(\frac{3\pi(4n-3)}{8}\right)^{-1/6}. \tag{92}$$

In order to calculate the rf power absorbed by the excitation of the spin wave modes, one must calculate the coupling of each spin wave mode to the resonator rf field, H_1. One may define the coupling integral, $h_n = \int u_n H_1 dz$, where H_1 is assumed to be constant. This is a good approximation for the rf field in a loop gap resonator, for example. For constant H_1, therefore, the absorptive part of the transverse magnetization in the rotating frame is given by

$$\gamma\hbar\mathcal{M}_+''(\omega) = \chi(0) \sum_n \frac{1}{\pi} \frac{i\Gamma_n}{(\omega - \omega_n)^2 + \Gamma_n^2} h_n u_n, \tag{93}$$

where ω_n is the frequency shift of the spin wave mode in the presence of the gradient and Γ_n is the width. The integral over z may be handled in the same way as the integral for the normalization constant: the contour may be deformed onto the $\Re\zeta$ axis. Values of the Airy integral for real argument are tabulated in, e.g., Abramowitz and Stegun,[22] so that evaluation of the quadratures is not difficult.

If one defines the observation phase properly, the absorbed power gives a pure absorption signal, which may be calculated from

$$\mathcal{P} = -i\omega \int \mathcal{M}_+ H_1 \, dz$$

$$\propto \sum_n \frac{1}{\pi|l_-|} \frac{\Gamma_n}{(\omega-\omega_n)^2+\Gamma_n^2} \frac{1}{(-\alpha_n)\mathrm{Ai}^2(\alpha_n)} \left[\int_{\alpha_n}^{\infty} \mathrm{Ai}(x)\, dx\right]^2. \tag{94}$$

In order to facilitate the evaluation of the absorption signal from the tables of quadratures, one may use the integral representation of $\mathrm{Ai}(x)$ to show that[23]:

$$\int_0^{\infty} \mathrm{Ai}(x)\, dx = 1/3. \tag{95}$$

Tables of quadratures for Airy integrals of the form $\int_0^a \mathrm{Ai}(x)\, dx$, where $a<0$ are given in.[12] One may therefore compute the coupling integrals, h_n, from the tabulated values of the Airy integral and the identity 95. This completes the analysis of the one-dimensional model system considered in this paper.

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NOTE ADDED IN PROOF

We have recently received a preprint from E. P. Bashkin that discusses spin-wave modes in D↓↓ from an alternative point of view. Both approaches yield similar results for the gradient-free case that Bashkin discusses.

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