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Spin Waves in Spin-Polarized Gases in the Knudsen Regime

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Boundary conditions are discussed for spin dynamics equations in dilute spinpolarized quantum gases. The bulk equations have the macroscopic "hydrodynamic" form even when mean free paths of the particles are large, and the main question is whether a supplementary "hydrodynamic" boundary condition is valid. Different boundary processes are considered including spin-conserving and non-conserving reflections, slip, formation of adsorbed surface layers, etc. The macroscopic boundary condition fails in the cases of very effective surface processes with violation of time-reversal symmetry (e.g. spin-lattice relaxation) or very high surface-induced diffusion rates. Otherwise, the surface processes are described by a simple boundary condition or by δ -type singularities in bulk equations. The meaning of different macroscopic parameters is clarified. The formation of dense adsorbed boundary layers changes the frequency shifts and linewidths of spin-wave resonances because of effective exchanges between surface and bulk particles and strong interactions within the boundary layers. Here the broadening of resonances is explained not only by additional surface dissipation (diffusion), but also by dephasing processes originating from a renormalization of the molecular field in the boundary layers. The results explain recent experiments by the Cornell group.

1. INTRODUCTION

Spin waves in spin-polarized quantum gases are usually studied (see review¹ and references therein) at low temperatures and densities of gases when the mean free paths of particles are quite long. In these conditions, one can easily reach a Knudsen regime where collisions with the walls may become more important than the interaction processes in the bulk.²

Below I discuss spin-wave resonances taking into account collisions of the particles with the cell walls. A general description of boundary scattering

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and relaxation should include spin-conserving reflections, spin non-conserving exchange and magnetic dipole processes, slip effects, sticking, the influence of possible adsorbed boundary layers, etc. Different complications are introduced step by step. For earlier results see Ref. 3 and references therein.

An appropriate way to describe different surface processes is to incorporate them into some reasonable boundary conditions. Spin dynamics of spin-polarized quantum gases is described by macroscopic equations of motion not only for a low-frequency "hydrodynamic" regime, but also for a high-frequency collisionless regime.¹ These equations in a uniform external magnetic field have the form of the well-known Leggett equations

$$(\omega - \Omega_0)\mathbf{M} - k_i \mathbf{J}_i = 0,$$

$$(\Omega - i/\tau) \mathbf{J}_i - (T/m)k_i \mathbf{M} = 0,$$
(1)

with the spin-wave spectrum

$$\omega = (T/m)k^2/(\Omega + i/\tau).$$
⁽²⁾

Here ω and **k** are the frequency and the wave vector of oscillations, *T* is the temperature, Ω_0 is the Larmor frequency in an external magnetic field, J_i and **M** are the densities of the spin current and magnetic moment, Ω is the frequency for the internal molecular field created by the coherent part of the interaction of the particles ($\Omega \gg kv$, $\omega - \Omega_0$), *m* and *v* are the particles' mass and velocity. Below we ignore the Larmor precession and assume, where necessary, that we are dealing with the rotating reference frame, $\omega \to \omega - \Omega_0$.

A natural boundary condition for these macroscopic equations of motion should be of the same order in gradients and have the form

$$\mathbf{M} + \Lambda n_i \nabla_i \mathbf{M} = 0, \qquad (3)$$

where **n** is the unit vector normal to the boundary (into the gas), and Λ is some coefficient with the dimensionality of length. When Eq. (3) is valid, this coefficient contains all the information about the boundary scattering.

Two main goals of this paper are to determine the limits of applicability for the boundary condition (3) including the Knudsen regime, and to express Λ through characteristic parameters of the interaction of the particles with the walls. If or when the boundary condition (3) fails, the whole macroscopic picture becomes meaningless even if the bulk equations (1) themselves are still valid. In this case one should return to the Boltzmann equation with a proper kinetic boundary condition instead of Eqs. (1) and (3).

In the simplest cases (see, e.g., Refs. 1-5) one finds either the zero spin current through the boundary, or the complete accommodation of magnetic moment at the non-magnetized wall. The spin current is proportional to a gradient of magnetization, and the former case corresponds to $\Lambda \rightarrow \infty$ and

 $\nabla_i \mathbf{M} = 0$. In the latter case $\Lambda \to 0$, and Eq. (3) reduces to $\mathbf{M} = 0$. Generally, the value of Λ depends on the structure of the walls, temperature, and the density of the gas N. In turn, the value of Λ determines the distribution of magnetization inside the cell, frequency shifts and linewidths. For example, in the simplest rectangular cell, the boundary conditions (3) lead for the standing wave $\mathbf{M} = \mathbf{M}_1 \exp(\mathbf{ikr}) + \mathbf{M}_2 \exp(-\mathbf{ikr})$ in the uniform field H to the dispersion relation

$$\tan k_i L_i = 2k_i \Lambda / (1 - k_i^2 \Lambda^2) \tag{4}$$

where indices characterize the directions in the rectangular cell with sizes L_i . The eigenvalues of the wave vector **k** are real (corresponding to a nondissipative standing wave) when Λ is real. As we will see later, the complex values of Λ and k originate not only from a surface dissipation, but also from some dissipationless dephasing surface processes.

The situation is more complicated in a non-uniform external magnetic field when the bulk magnetization is given not by a combination of plain waves, but by the Airy functions ϕ (cf. Refs. 1, 4)

$$\mathbf{M}(x) = \mathbf{M}_0 \phi \left(\left[-iG \right]^{1/3} \left[-x + \frac{\omega \tau T/Gm}{1 + i\Omega \tau} \left(1 + \Omega^2 \tau^2 \right) \right] \right)$$
(5)

where \mathbf{M}_0 is a scaling constant, and the field gradient **G** is directed along the x-axis. Here the frequency shifts and linewidths of the spin-wave resonances are given by the substitution of (5) into the boundary condition (3) at x = 0,

$$\phi(x=0) + \Lambda \phi'_x(x=0) = 0$$
 (6)

The boundary condition at the second wall, x=L, is irrelevant since the magnetization near this wall is very small, $M(L) \rightarrow 0$, due to strong inhomogeneity caused by the field gradient G.

Therefore, one has an additional reason to try to justify the use of macroscopic boundary conditions (3): the evaluation of the frequency eigenvalues (2), (5), (6) is rather cumbersome even if Eq. (3) is valid. If the macroscopic description fails, even only because of boundary conditions, calculations for inhomogeneous situations would become extremely difficult.

2. DIFFUSIVE AND SPECULAR REFLECTION BY NON-MAGNETIC WALLS

At low temperatures, the wavelengths of the particles are large in comparison with a characteristic atomic size, $\lambda \gg a$, and the particles are mostly scattered elastically by the walls. In most of the experiments, the walls are

magnetically protected, and the reflection is spin-conserving. However, there is a competing factor: an attraction to the walls becomes more and more important with lowering temperatures leading eventually to a formation of adsorbed boundary layers. The appearance of such layers results in a significant surface magnetic relaxation which, in turn, influences the bulk relaxation because of strong exchange between surface and bulk particles.

The adequate macroscopic boundary conditions should be formulated on the basis of a collision operator in the Boltzmann equation including collisions of the particles with the walls. The goal here is to determine what happens to the particle-surface collision operator in course of derivation of the macroscopic equations of motion by integration and parameterization of the Boltzmann equation. Since we are dealing with very dilute gases, the linearized collision integral may be separated into independent bulk and surface parts:

$$L(n) = L_v(n) + L_s(n).$$
 (7)

If the walls are non-magnetic, and particle-surface, as well as particle-particle, collisions are spin-conserving, the traces over the spin and spatial variables of the collision operators, multiplied by spin, are equal to zero,

$$\operatorname{Tr}_{\sigma} \int d^{3}p \, \boldsymbol{\sigma} \{L_{v}(n) + L_{s}(n)\} = 0, \qquad (8)$$

and the first macroscopic equation of motion—the magnetic moment conservation law—has exactly the same form as in the absence of the boundary collisions, Eq. (1),

$$\partial \mathbf{M}/\partial t + \operatorname{div} \mathbf{J} = 0.$$
 (9)

The second macroscopic equation, the equation in J_i , contains two types of interaction terms: the molecular field and dissipative terms. The molecular field originates from non-zero spin commutators for particles. If the walls are non-magnetic, all spin commutators containing the wall variables are equal to zero by definition, and the collisions with the non-magnetic walls do not renormalize the molecular field terms. This resembles the irrelevance of non-magnetic bulk impurities for the bulk moleclar field.¹ Then, if one is not interested in dissipative diffusion processes, the spin current conservation law retains its bulk form, and the boundary condition reduces to (3) with $\Lambda \rightarrow \infty$.

The boundary scattering changes diffusion currents near the walls, thus contributing to the spin diffusion. If we were dealing, instead of wall scattering, with a scattering by non-magnetic bulk impurities, the additional diffusion spin currents in the relaxation time approximation would look like

$$\operatorname{Tr}_{\sigma} \int d^{3}p \, v_{i} \sigma \{ L_{v}(n) + L_{s}(n) \} = -\mathbf{J}_{i}/\tau - \mathbf{J}_{i}/\tau_{s} = -\mathbf{J}_{i}/\tau_{e}$$
(10)

where the effective relaxation time τ_e is a combination of the particle-particle and particle-impurity diffusion relaxation times τ and τ_s :

$$\tau_e^{-1} = \tau^{-1} + \tau_s^{-1} \tag{11}$$

Such an approach would be quite appropriate when describing, for example, the effect of ⁴He impurities on spin waves in ³He[†] gas. The walls are also some δ -type imperfections in the gas which limit the mean free paths and lead to an additional diffusion in a manner similar to bulk impurities. It would be natural to try to introduce the wall-induced relaxation τ_s in the form of Eq. (10) with δ -type singularities. At first glance, the only precaution in using Eqs. (10) and (11) for the boundary scattering is to exclude the specular part of the reflection (non-diffusive scattering) and to be more careful with macroscopic boundary conditions on magnetic moment and spin current. As we will see, the influence of the boundary on spin waves in polarized gases does give rise to some additional δ -type spin diffusion terms, but is not equivalent to a simple renormalization of the bulk relaxation time τ . The proper parameterization differs from Eqs. (10) and (11) even in relaxation time approximations.

A more realistic approach is to integrate separately the Boltzmann equation in magnetic moment in the bulk of the system and in some boundary layer of the thickness a^* with the boundary conditions in the form of the continuity equations for the magnetic moment and the spin current in these two subsystems. The bulk magnetic moment is given by exponents $\exp(\pm ik_0x)$ with k_0

$$k_0^2 = \omega(\Omega + i/\tau)m/T - q^2, \quad \text{Im } k_0 > 0$$
 (12)

(cf. Eq.(2); q is the projection of the wave vector \mathbf{k} on the plane of the wall x = const), or by Airy functions (5), and within the boundary layer—by plain waves $exp(\pm ik_bx)$ with the value of k_b related to the scattering of particles by boundary imperfections. Here the main simplification would be to describe the scattering within the boundary layer not in the form of integral operators but as linear combinations of spin densities and currents with the coefficients of the proper symmetry. The last step is the momentum integration of the corresponding equations simultaneously with directing the thickness of the boundary layer $a^* \rightarrow 0$. As a result, the macroscopic equations of spin dynamics incorporate the boundary scattering in the form of different δ -type singularities. In this way, the meaning of different boundary parameters becomes rather straightforward, and their values can be evaluated using the densities of different types of "imperfections" within the boundary layers of the thickness a^* (usually, a^* is of the order of atomic size a). An additional benefit of keeping a^* in the equations is to have the boundary parameters with the same dimensionalities as their bulk counterparts.

Summarizing, one would expect the second macroscopic equation of motion (the equation in the transverse spin current J_{i}) to have the following form for two walls at x=0 and x=L (in the uniform magnetic field):

$$-i\Omega \mathbf{J}_i + (T/m)(\partial/\partial x_i)\mathbf{M} = -\{1/\tau + (a^*/\tau_s)\delta_{ix}[\delta(x-L) + \delta(x)]\}\mathbf{J}_i \quad (13)$$

where the "thickness" of the boundary $a^* \sim a$ is actually an auxiliary constant with a dimensionality of length. This constant is introduced in order to keep dimensionalities for all surface variables and densities the same as for their bulk counterparts; it never enters any equations as an independent parameter, but only in combination with other surface characteristics. Eq. (13) corresponds to the approximation somewhat similar to Eq. (10):

$$\operatorname{Tr}_{\sigma} \int d^{3}p \, v_{i} \sigma L_{s}(n) = -(\mathbf{J}_{i}/\tau_{s})a^{*}[\delta(x) + \delta(x-L)]. \tag{14}$$

After the Fourier transformation, Eqs. (9) and (13) reduce to

$$\omega \mathbf{M} - k_i \mathbf{J}_i = 0$$

$$(\Omega + i/\tau) \mathbf{J}_i - (T/m) k_i \mathbf{M}$$

$$= -(ia^*/\tau_s) \delta_{ix} [\mathbf{J}_i(x=0) + \mathbf{J}_i(x=L) \exp(ik_x L)]$$
(15)

(here and below, ω is not a bare frequency, but rather $\omega - \Omega_0$).

The solution of Eqs. (15) has the form

$$\mathbf{M}(x) = \frac{ima^*}{2\pi T\tau_s} \int \frac{k_x \, dk_x}{k_x^2 - k_0^2} \exp(-ik_x x) [\mathbf{J}_0 + \mathbf{J}_L \exp(ik_x L)] \tag{16}$$

where $\mathbf{J}_0 = \mathbf{J}_x(x=0)$, $\mathbf{J}_L = \mathbf{J}_x(x=L)$, and k_0 is given by Eq. (12).

The direct integration in Eq. (15) leads to

$$\mathbf{M}(x<0) = -\frac{ma^*}{2T\tau_s} \left[\mathbf{J}_0 + \mathbf{J}_L \exp(ik_0 L) \right] \exp(-ik_0 x)$$
(17a)

$$\mathbf{M}(x > L) = \frac{ma^*}{2T\tau_s} \left[\mathbf{J}_0 + \mathbf{J}_L \exp(-ik_0 L) \right] \exp(ik_0 x)$$
(17b)

$$\mathbf{M}(0 < x < L) = \frac{ma^*}{2T\tau_s} \left[\mathbf{J}_0 \exp(ik_0 x) - \mathbf{J}_L \exp(ik_0 (L-x)) \right]$$
(17c)

with the compatibility condition in the form of

$$\mathbf{M}(x<0) = \mathbf{M}(x>L) = 0, \ \mathbf{M}(x=0) = \mathbf{M}_0, \ \mathbf{M}(x=L) = \mathbf{M}_L$$
(18)

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Comparing Eqs. (17) and (18), one has

$$\mathbf{J}_0 + \mathbf{J}_L \exp(ik_0L) = \mathbf{J}_0 + \mathbf{J}_L \exp(-ik_0L) = 0,$$

while the magnetic moment distribution and the eigenvalues of the wave vector are reduced to

$$\exp(2ik_0L) = 1, k_0 = \pi n/L, \ \omega = \frac{T/m}{\Omega + i/\tau} \left(\frac{\pi^2 n^2}{L^2} + q^2\right),$$

$$\mathbf{M}(0 < x < a) = \frac{ma^*}{T\tau_s} \mathbf{J}_0 \cos(\pi nx/L)$$
(19)

Comparison with Eq. (4) demonstrates that this case is equivalent to the macroscopic boundary condition (3) with $\Lambda \rightarrow \infty$. This result is not unexpected since it is clear from the beginning that the simple reflection corresponds to the absence of the spin current through the boundary. This simplest case was discussed in such detail in order to skip lengthy calculations in analogous, but much more cumbersome and interesting situations.

Eqs. (13) and (14) do not reflect boundary-induced spin diffusion because the right hand side of Eqs. (13) and (14) is non-zero only for the x-component of the spin current. A more general equation should include changes in the y, z- components of J_i . In a system with one preferred direction (x), this more general equation should contain two scattering parameters rather than one, τ_s ;

$$-i(m/T)(\mathbf{\Omega}+i/\tau)\mathbf{J}_{i}+(\partial/\partial x_{i})\mathbf{M}$$

= $-a^{*}[\delta(x)+\delta(x-L)](\mathbf{\Omega}+i/\tau)^{2}(m/T)^{2}[(D'_{s}-D_{s})n_{i}n_{k}\mathbf{J}_{k}+D_{s}\mathbf{J}_{i}]$ (20)

where D'_s and D_s describe the wall-induced spin diffusion perpendicular and along the wall, and **n** is the unit vector normal to the wall. Due to a^* , both D'_s and D_s have the usual dimensionality of spin diffusion coefficients.

The non-zero value of D_s in Eq. (20) is associated with a non-specular (in a general sense) part of reflectons caused either by slip effects⁶

$$D_s \sim vl$$
,

(l is the particles' mean free path), or by a diffuseness of wall scattering,

$$D_s = v\zeta$$

related to microscopic, $\zeta \sim a^2/\lambda$, or macroscopic, $\zeta \sim G(0)/R$, inhomogeneities of the surface. Here $G(y-y', z-z') = \langle u(y, z)u(y', z') \rangle$ and R are the correlation function and radius of macroscopic inhomogeneities.

The form of coefficients in Eq. (20) corresponds to the diffusion spin current j and magnetization m in the boundary layer obeying the equations

$$\omega \mathbf{m} - q_i \mathbf{j}_i = i n_i \mathbf{J}_i(\mathbf{0}), \tag{21}$$

$$(i/\tau_w)\mathbf{j}_i - (T/m)q_i\mathbf{m} = 0, \qquad (22)$$

$$\mathbf{j}_k = ia^* D_s q_k \mathbf{M}(0) \tag{23}$$

In this particular case of simple diffusive (or slip) reflections, neither the exchange diffusion relaxation time within the boundary layer, τ_w in Eq. (2), nor ω in Eq. (21), are important.

The consequent procedure is completely analogous to the one described above. Now the integral (16) has the form

$$\frac{ia^*}{2\pi} \int dk_x \frac{\exp(-ikx)}{k_x^2 - k_0^2} \left\{ k_x [\mathbf{J}_0 + \mathbf{J}_L \exp(ik_x L)] + [\mathbf{J}_0' + \mathbf{J}_L' \exp(ik_x L)] \right\}$$
(24)

where $J'_0 = q_i J_i(x=0), J'_L = q_i J_i(x=L)$.

Direct integration leads to the equations analogous to (17), but with four, and not two, boundary values of the current $J_{0,L}$ and $J'_{0,L}$. Therefore one cannot simply use the boundary conditions (18) as above. One should rather derive an expression analogous to (16) and (24) for $q_i J_i$ from Eq. (20) and the first of Eqs. (15), calculate the corresponding integrals and use the conditions

$$q_i \mathbf{J}_i(x < 0) = q_i \mathbf{J}_i(x > L) = 0 \tag{25}$$

supplementary to (18). Then one obtains a system of four homogeneous equations in four variables $J_{0,L}$, $J'_{0,L}$ leading to the eigenspectrum of the type (4):

$$\exp(2ik_0L) = (1 - i/\Lambda k_0)^2 / (1 + i/\Lambda k_0)^2$$
(26)

if the parameter Λ for the effective boundary condition (3) is equal to

$$\Lambda = -\frac{iT/m}{q^2 a^* D_s(\Omega + i/\tau)}$$
(27)

On the other hand, Eqs. (21)-(23) result in

$$\Lambda = -i[(T/m)/a^*D_s]/[q^2(\Omega + i/\tau) - ik^2/\tau_w]$$
(28)

which is equivalent to Eq. (27) if the internal wall relaxation time is much longer than the bulk one, $\tau \ll \tau_w$.

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In the hydrodynamic regime for the bulk, $\Omega \tau \ll 1$, Eq. (27) reduces to

$$\Lambda(\Omega\tau\ll 1) = -\frac{\tau(T/m)}{a^*q^2D_s} = -\frac{D}{a^*q^2D_s}$$
(29)

 $D = \tau T/m$ is the bulk transverse spin diffusion coefficient. Though the values of Λ (29) and k_0 (4), (26) are real, the frequency (2), (12) remains imaginary,

$$\omega = -i\tau (T/m)(k_0^2 + q^2), \qquad (30)$$

and surface scattering leads only to an additional dissipation.

In the opposite spin-wave regime, $\Omega \tau \gg 1$, the parameter Λ is imaginary,

$$\Lambda(\Omega\tau \gg 1) = -\frac{i(T/m)}{a^*q^2 D_s \Omega},\tag{31}$$

and usually large, $|\Lambda| \gg 1$. In this case the inclusion of boundary scattering also results in some broadening of the spin-wave resonances with

$$\omega_n'' = -4\omega_n q^2 a^* L D_s \Omega m / \pi^2 n^2 T \tag{32}$$

 $(\omega_n \text{ is the frequency shift for the$ *n*th resonance). This expression should be compared with a purely bulk width

$$\omega'' = -\omega/\Omega\tau.$$

One should keep in mind that the eigenfrequency spectrum even in a rectangular 3D cell is rather complicated since eigenvectors k_0 for different directions depend on each other because of the dependence of effective Λ in Eq. (27) on the components of the wave vector parallel to the wall, **q**.

3. MORE GENERAL BOUNDARY CONDITIONS

The next step is to allow some magnetic relaxation of particles on the walls as a result of either magnetic dipole processes or exchanges with boundary particles. Such a relaxation corresponds to the addition of the term $-i\mathbf{m}/\tau_{sd}$ with a characteristic time τ_{sd} into the r.h.s. of Eq. (21).

These processes lead to a violation of the magnetic moment conservation law (9), (15) which should be re-written as

$$\omega \mathbf{M} - k_i \mathbf{J}_i = -[ia^* (\mathbf{M} - \mathbf{M}_e) / \tau_{sd}^*] [\delta(x) + \delta(x - L)]$$
(32)

with the renormalized relaxation time $\tau_{sd}^* = \tau_{sd}\tau_w(T/m)/D_s$. The eigenspectrum still has the form (26) with

$$\Lambda = -\frac{iT/m}{a^*(\Omega + i/\tau)} \left[\tau_{sd}^{*-1} + q^2 D_s\right]^{-1}$$
(33)

This equation is an adequate one for most of particular spin-wave problems in spin-polarized quantum gases.

However, Eqs. (20) and (32) are not the most general ones. As soon as one allows the spin relaxation on the walls and the corresponding violation of the time-reversal symmetry, one should include all other possible terms with the boundary values of the spin current J_i and the transverse magnetization **M** into the r.h.s. of Eqs. (20) and (32):

$$\omega \mathbf{M} + i(\partial/\partial x_i) \mathbf{J}_i = -ia^* [\delta(x) + \delta(x - L)] [\alpha_{11}\mathbf{M} + \alpha_{12}n_i \mathbf{J}_i], \quad (34a)$$

$$(\mathbf{\Omega} + i/\tau)\mathbf{J}_i + i(T/m)(\partial/\partial x_i)\mathbf{M}$$

= $-ia^*[\delta(x) + \delta(x-L)][\alpha_{21}n_i\mathbf{M} + (\alpha_{22} - \alpha_{23})n_in_k\mathbf{J}_k + \alpha_{23}\mathbf{J}_i].$ (34b)

where the meaning of all relaxation constants α_{ik} is quite clear. Then the spectrum obtains the form

$$\tan(k_0 L) = -(2ik_0 a^* T/m) [(\Omega + i/\tau)(\alpha_{11} - \alpha_{12}\alpha_{21}/\alpha_{23} + \alpha_{23}q^2 T/m(\Omega + i/\tau)]/[(T/m)^2(k^2 - (\Omega + i/\tau)^2\alpha_{12}^2/\alpha_{22}^2) + a^{*2} \{ [(\Omega + i/\tau)(\alpha_{11} - \alpha_{12}\alpha_{21}/\alpha_{22}) + \alpha_{23}q^2 T/m(\Omega + i/\tau) \}^2]$$
(35)

The comparison of Eq. (35) with macroscopic Eq. (4) demonstrates that the boundary condition may be reduced to Eq. (3) only if one of the off-diagonal terms in denominator, namely,

$$(T/m)^2(\Omega + i/\tau)^2 \alpha_{12}^2/\alpha_{22}^2,$$
 (36)

may be neglected. Then Λ reduces to

$$\Lambda = -i[(\Omega + i/\tau)(\alpha_{11} - \alpha_{12}\alpha_{21}/\alpha_{22})m/T + \alpha_{23}q^2/(\Omega + i/\tau)]^{-1}$$
(37)

which is essentially the same as Eq. (33).

These results show that the macroscopic description of spin dynamics of dilute spin-polarized quantum gases is incompatible in the Knudsen regime with strong spin non-conserving boundary scattering caused, for example, by magnetic dipole-dipole interaction. If the dipole interaction is rather weak, one is still able to take it into account using the macroscopic boundary condition (3) with the value of Λ given by Eq. (33) with long dipole relaxation time τ_{sd} . Other limitations on Eq. (3) arise when studying the integration of the Boltzmann equation leading to Eqs. (18) and (25). The transition from the 3D equations of spin dynamics in the boundary "layer" to the 2D ones (21)–(23) is possible when the x-component of the wave vector in the layer, k_b , is small,

$$k_b a^*, k_b^2 a^*/k_0 \ll 1$$
 (38)

meaning that Λ ,

$$\Lambda = \frac{i(T/m)}{D_s(\Omega + i/\tau)} \frac{1}{k_b^2 a^*},$$
(39)

should be rather large, $\Lambda k_0 \gg 1$, $\Lambda/a^* \gg 1$. This is true only if the surface depolarization and/or diffusion times τ_{sd} and mD_s/T are much longer than the bulk relaxation time τ , and the boundary condition on the "outer" side of the layer corresponds to the absence of spin current. These conditions are fulfilled if the density of surface imperfections is smaller than the density of the gas, and if the cell is made of non-magnetic material. As a result the meaning of the boundary parameters becomes rather straightforward, and their values can be easily estimated using the densities of different types of "imperfections" within the boundary "layer" of the thickness a^* .

4. ADSORBED BOUNDARY LAYERS

The situation changes at low temperatures when an adsorption of particles may lead to a formation of boundary layers with a noticeable density ρ :

$$\rho = N\lambda \exp(\varepsilon_0/T) \tag{40}$$

where ε_0 is the binding energy at the surface (1 K for hydrogen atoms and ⁴He coating of the walls, 0.35 K in case of ³He coating, and 2 K for ³He particles adsorbed by a free surface of ³He-⁴He liquid mixtures), and $\lambda \sim 2\pi\hbar/(mT)^{1/2}$ is the de Brogile wavelength of the particles. This leads not only to some numerical renormalizations of the boundary constants, but also to some major changes caused by a very effective exchange between identical bulk and surface particles. The interaction with boundary layers (and with the walls—via boundary layers) makes the boundary processes different from the above ones.

The adsorbed layers and the gas itself consist of the same identical particles. As a result, the exchange interaction between the bulk and boundary particles changes the effective molecular field (frequency Ω) at the boundary. This change and changes in relaxation parameters still may be incorporated into the above formalism. What is more, the strong exchange interaction between 3D and 2D particles makes the interaction with substrate more effective leading to an increase in relaxation rates (see comments in Ref. 7).

Furthermore, the magnetic exchange interaction within the boundary layer may be responsible for a propagation of peculiar 2D diffusive (or spinwave) spin modes along the walls. Now the boundary conditions should reflect a more complicated coupling of bulk and surface equations. However, one should avoid an effective increase in the order of the full set of differential equations of spin dynamics. The Leggett equations (1) for bulk spin dynamics were derived neglecting all higher order time derivatives (high order terms in ω) appearing after the momentum integrations of the bulk Boltzmann equation, and it would be beyond the accuracy to include higher-order time derivatives appearing as a result of boundary interactions. One should consider only the equilibrium (static) boundary coupling in the form of Eq. (23) restricting oneself to low-frequency spin oscillations (the lowest standing modes).

Boundary equations of spin dynamics (21)-(23) should be modified. Without the time derivatives and long dipole processes, the surface magnetic moment conservation law (21) assumes the form

$$\partial \mathbf{m}/\partial t + \operatorname{div} \mathbf{j} = n_i \mathbf{J}_i - (\mathbf{m} - \mathbf{m}_e)/\tau_{sd}$$
 (41)

where the transverse components of the equilibrium surface density of the magnetic moment, \mathbf{m}_e , are usually equal to zero. Eq. (41) should be supplemented by a modified equation (22) for a surface spin current, **j**,

$$(\boldsymbol{\Omega}_w + i/\tau_w) \mathbf{j}_i - (T/m) q_i \mathbf{m} = 0$$
(42)

where $\Omega_w \sim \alpha \hbar(\rho/m)(a/\lambda)$ is the molecular field frequency for the 2D boundary layers with the spin polarization α , and, in contrast to Eq. (41), the symmetry dictates the zero right hand side. If the dipole interaction is very strong and the time-reversal symmetry is broken, then one should add terms with **M** and J_i into the right hand sides of Eqs. (41) and (42) in the same manner as in Eqs. (34). This would complicate all the equations, but would not bring into being any new effects.

The constants in Eqs. (41) and (42) can be reduced to the coefficients of the type (20) and (32). Here again the 2D layer can be modeled as a δ type singularity (boundary condition) with some effective width a^* which is of the order $a^* \sim \lambda \ll a$ (now the thickness of a layer, a^* (23), is of the atomic size only in case of very deep boundary layers). The boundary condition again assumes the macroscopic form (3) with the coefficient Λ

$$\Lambda(\tau_{sd} \to \infty) = \frac{-iT/m}{a^* D_s(\Omega + i/\tau)} \left[q^2 - (\Omega_w + i/\tau_w) \omega(m/T) \right]^{-1}$$
(43)

Because of the high density of surface layers, the surface molecular field Ω_w and the frequency of collisions, $1/\tau_w$, are large in comparison with their bulk counterparts. Therefore, one should neglect in Eq. (43), in contrast to Eq. (28), the term with q^2 :

$$\Lambda = i(T/ma^*D_s)k^{-2}/(\Omega_w + i/\tau_w) \tag{44}$$

Both, $\Omega_w \sim \alpha \hbar(\rho/m)(a/\lambda)$ and $1/\tau_w \sim \rho v a^2/\lambda$, are proportional to the density of the adsorbed boundary layers, ρ , and increase exponentially with temperature, Eq. (40). As a result, the formation of the boundary layers leads to an exponential (in temperature) decrease in $|\Lambda|$. In a spin-wave regime for the boundary layers, $\Omega_w \tau_w \ll 1$, the value of Λ is imaginary,

$$\Lambda = iT/ma^* D_s k^2 \Omega_w \tag{45}$$

meaning that in case of a rectangular cell, Eq. (26), the formation of dense boundary layers results mostly in some shifts of spin-wave resonances but with a very noticeable change in width (cf. Eq. (32)):

$$\omega_n'' = -4\omega_n L(m/T) a^* D_s k^2 \Omega_w / \pi^2 n^2$$
(46)

In the opposite case of insignificant coherent interaction within the boundary layers, the value of Λ is real,

$$\Lambda = \tau_w T / ma^* D_s k^2, \tag{47}$$

and major effect in the bulk spin-wave regime $\Omega \tau \ll 1$ is a shift of resonances

$$\delta \omega_n = -4\omega_n L / \Lambda \pi^2 n^2 \tag{48}$$

while the attenuation is associated mostly with the bulk processes.

The comparison with the experimental data² of the Cornell group (exponential increase in linewidths with a proper exponent (40), and lack of large frequency shifts) allows one to conclude that the experimental conditions correspond to the formation of dense boundary layers with the internal collision rate much higher than in the bulk, and with a large coherent part of the interaction within the layers, Ω_w . The increase in linewidths was related to a strong surface molecular field (46) and was caused not by some additional surface dissipation, but by the dephasing associated with changes in precession frequencies near the boundary. The lack of noticeable frequency shifts indicates that the value of Λ was still large.

The available information is insufficient to make a conclusion on the existence of surface spin modes⁸ in experimental conditions² though the strong molecular field is a definite indication towards this possibility.

Boundary conditions for solid helium layers on cell walls⁹ differ from the above case of adsorbed layers for a hydrogen gas or ³He-⁴He mixtures. The major difference is a much lower (by three orders of magnitude)

exchange rate in solid layers making the molecular field Ω_w and diffusion rate $1/\tau_w$ (43), much smaller, and often negligible. Another factor is a smaller exchange rate between bulk and surface particles resulting in smaller D_s a larger Λ .

Some preliminary results have already been published earlier.¹⁰

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REFERENCES

- 1. A. E. Meyerovich, in *Helium Three*, W. P. Halperin and L. P. Pitaevcskii, eds. (North-Holland, Amsterdam, 1990), Chapter 3, pp. 757-879.
- 2. N. P. Bigelow, J. H. Freed, and D. M. Lee, Phys. Rev. Lett. 63, 1609 (1989).
- J. J. Berkhout, and J. T. M. Walraven, in Spin-Polarized Quantum Systems, S. Stringari, ed. (World Scientific, Singapore, 1989), pp. 201-208.
- 4. J. Freed, Ann. Phys. Fr. 10, 901 (1985).
- 5. D. Candela, N. Masuhara, D. S. Sherrill, and D. O. Edwards, J. Low Temp. Phys. 63, 369 (1986).
- 6. K. Ivanova, and A. E. Meyerovich, J. Low Temp. Phys. 72, 461 (1988).
- 7. A. Schuhl, S. Maegawa, M. W. Meisel, and M. Chapellier, Phys. Rev. Lett. 54, 1952 (1985).
- 8. V. Lefevre-Seguin, H. Guignes, and C. Lhuillier, Phys. Rev. B 36, 141 (1987).
- 9. P. C. Hammel and R. C. Richardson, Phys. Rev. Lett. 52, 1441 (1984).
- 10. A. E. Meyerovich, Physica B 165 & 166, 747 (1990).