

Nonlinear longitudinal NMR in superfluid ^3He

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The effect of a strong magnetic field on the relative orientation of the field, average spin, and order parameter vectors is considered for longitudinal NMR in superfluid ^3He . It is shown that the orientation is stabilized by strong static fields. Resonant effects are considered for nonlinear parallel “ringing” in which there is a slow broadening or stochastic modulation of the ringing frequency. The angle orientation is shown to be stable for the A phase, but in the B phase the stability depends on the remanent static field after the nonlinear ringing has been excited.

INTRODUCTION

Leggett first predicted¹ that a weak time-varying magnetic field $h(t)$ in superfluid ^3He can excite oscillations in the average spin vector S along the direction of the field. This phenomenon was subsequently detected in longitudinal NMR experiments as a resonant absorption of energy² from an ac field close to the resonance frequency Ω (dipole frequency) for longitudinal NMR. If a static field H_0 parallel to $h(t)$ is also present then the spin oscillates longitudinally about the equilibrium value $S_0 = \chi H_0 / |\gamma|$, which does not significantly alter the longitudinal NMR (here χ is the static susceptibility and γ is the gyromagnetic ratio).

The Leggett description of the magnetic properties of ^3He requires that one analyze the combined motion of the average spin vector S and the order parameter \mathbf{d} . In longitudinal NMR one usually assumes that in the A phase, \mathbf{d} moves perpendicular to the plane of the field: $\mathbf{d} \perp \mathbf{h}(t) \parallel S$, while in the B phase \mathbf{d} rotates about an axis \mathbf{n} parallel to $h(t)$ and S . We refer to this mutual orientation as the geometry of the longitudinal NMR; it is uniquely determined by $S = |S|$ and Φ , where Φ is the angle between \mathbf{d} and the plane normal to the field (for the A phase) or the rotation angle of \mathbf{d} about n (for the B phase). In either case, Φ is described by the equation for a pendulum which is driven by a nonlinear force and subject to nonlinear Leggett-Takagi damping. If the fields vary sufficiently slowly, the nonlinearity will be negligible and S and Φ will oscillate slightly about their equilibrium values, which for Φ are the points at which the dipole energy is a minimum.

It was shown in Refs. 3 and 4 by solving the equations numerically that for stronger ac fields, the behavior of S and Φ becomes very complicated and may be qualitatively different, depending on the field amplitude h and frequency ω . The specific nature of the Leggett-Takagi damping mechanism is largely responsible for this behavior. The geometry of the longitudinal NMR was assumed to be stable in Refs. 3 and 4 (so that the stability problem was not addressed), and it is only in this case that the longitudinal NMR is insensitive to static fields.

The stability problem for longitudinal NMR was first raised in Refs. 5 and 6, where it was shown that when no static field is present, fluctuations grow in amplitude and distort the longitudinal geometry, so that nonlinear longitu-

dinal NMR cannot be observed at all in the B phase⁶ and only with great difficulty in the A phase.⁵

There are grounds for believing that an applied static magnetic field might stabilize the geometry, so that the longitudinal resonance remains stable over a wider range. The purpose of the present paper is to investigate stabilization by a static field and to analyze longitudinal NMR for the case when the ac field frequency ω is equal to the rotation frequency of the “pendulum” \mathbf{d} when a static field changes instantaneously (during a time much shorter than the characteristic Leggett-Takagi relaxation time).

1. STABILITY ANALYSIS IN THE ADIABATIC APPROXIMATION

Following Fomin,⁷ we write

$$\begin{aligned} \dot{S} &= -\omega_L \frac{\partial V}{\partial \Phi}, \\ \dot{\Phi} &= \omega_L (S-1) - \omega_1 \sin \omega t + \omega_L \frac{\partial V}{\partial S} - k \omega_L^2 \frac{2S}{2S+P} \frac{\partial V}{\partial \Phi}, \\ \dot{P} &= 0 \end{aligned} \quad (1)$$

to first order in ε for the spin dynamics of the A and B phases of ^3He in the presence of an ac field $h(t) = h \sin \omega t$. We assume that the strong static magnetic field H_0 is parallel to $h(t)$ and that the necessary conditions for Fomin's adiabatic approximation $\varepsilon = \Omega^2 / \omega_L^2 \ll 1$ are satisfied ($\Omega = \Omega_{A,B}$ is the frequency of the linear longitudinal resonance in the A and B phases, and $\omega_L = |\gamma| H_0$ is the Larmor frequency). In Eqs. (1), $V = V_{A,B}$ is the dipole energy in the A and B phases, averaged over the period of the Larmor precession:

$$\begin{aligned} V_A &= -\frac{\Omega_A^2}{8\omega_L^2} \left[\left(1 + \frac{P}{S}\right)^2 + \frac{1}{2} \left(2 + \frac{P}{S}\right)^2 \cos 2\Phi \right], \\ V_B &= \frac{2\Omega_B^2}{15\omega_L^2} \left[\frac{1}{2} + \frac{P}{S} + \left(2 + \frac{P}{S}\right) \cos \Phi \right]^2. \end{aligned} \quad (2)$$

We take H_0 to lie along the z axis; $p = S_z - S$, where S_z is the projection on the z axis, $\omega_1 = |\gamma|h$, and k is the Leggett-Takagi (LT) relaxation coefficient. We use dimensionless quantities—spins are in units of S_0 , while the dipole energy is divided by χH_0^2 .

In this case, distortion of the longitudinal geometry is tantamount to self-excitation of the transverse spin compo-

nent S_{\perp} (in other words, the fluctuations in S_{\perp} grow in amplitude).¹⁾ Assuming $P \ll 1$, $S \sim 1$, and using $S_{\perp} = (S^2 - S_z^2)^{1/2}$, we get the following equations from Eqs. (1) for small S_{\perp} :

For the A phase,

$$\delta \dot{S}_{\perp} = -\frac{\Omega_A^2}{4\omega_L} \delta S_{\perp} \sin 2\Phi, \quad (3)$$

$$\ddot{\Phi} + \Gamma_A (\cos 2\Phi) \dot{\Phi} + \frac{1}{2}\Omega_A^2 \sin 2\Phi = -\omega_1 \omega \cos \omega t; \quad (4)$$

for the B phase,

$$\delta \dot{S}_{\perp} = \frac{\gamma \Omega^2}{15\omega_L} \delta S_{\perp} (1 + 4 \cos \Phi) \sin \Phi, \quad (5)$$

$$\ddot{\Phi} - \frac{4}{15} \Gamma_B (\cos \Phi + 4 \cos^2 \Phi) \dot{\Phi} - \frac{4}{15} \Omega_B^2 (1 + 4 \cos \Phi) \sin \Phi = -\omega_1 \omega \cos \omega t, \quad (6)$$

where $\Gamma_A = k\Omega_A^2 \approx 2 \cdot 10^4 \text{ s}^{-1}$ and $\Gamma_B = k\Omega_B^2 = 10^4 \text{ s}^{-1}$.

In view of Eqs. (3) and (5), we take the longitudinal geometry to be unstable in the A and B phases if and only if

$$\sin 2\Phi < 0 \quad (7)$$

and

$$\sin \Phi (1 + 4 \cos \Phi) > 0, \quad (8)$$

respectively, where Φ denotes solutions of the nonlinear equations (4), (6) such that $\Phi(0)$ is a minimum in V and $\dot{\Phi}(0) = 0$. We will analyze the existence of solutions of Eqs. (4), (6) satisfying (7) and (8) by using numerical results found for these equations in Refs. 3 and 4. In particular, the results in Ref. 3 imply that as the field "amplitude" ω_1 increases, bifurcations may occur in the A phase. The nature of the bifurcations depends on the frequency ω —transitions may occur from small oscillations Φ of frequency Ω_A to oscillatory behavior of Φ with period equal to a multiple of the period of the driving force, or to random motion with a correlation time $\sim 2\pi/\omega$. We note that the "negative friction" in the interval $\pi/4 < \Phi < 3\pi/4$ is responsible for the rotation or crossing of the pendulum (4) through the maximum "height" $\Omega_A^2/4$. As a result, the pendulum is not confined to the region defined by (7), and for all the cases considered in Ref. 3 the right-hand side of (3) averages to zero over times $t \gg 2\pi/\Omega_A, 2\pi/\omega$. For the A phase we thus conclude that: 1) The longitudinal geometry is stable, because the distorting fluctuations do not grow in amplitude if the static field is sufficiently strong; 2) The results in Ref. 3 are not correct⁵ unless a strong stabilizing static field is present.

The above arguments do not suffice to determine if the longitudinal geometry is asymptotically stable. However, they do indicate that it is stable for times $\sim 1/\varepsilon^2 \omega_L$, which may be comparable to the upper time bound for the validity of the Leggett-Takagi equations. We will therefore consider only these times.

Similar arguments based on the results in Ref. 4 for the B phase show that strong static magnetic fields also stabilize the longitudinal geometry in the B phase.

2. ANALYSIS OF THE STABILITY OF RAPID OSCILLATIONS OF THE ORDER PARAMETER

It is well known that if $\Gamma \ll \Omega, \omega_1 = 0, V$ has a minimum at $\Phi(0)$, and $\dot{\Phi}(0) = \Lambda \gg \Omega$, then the pendulum equations (4), (6) admit solutions of the form $\Phi = \Lambda t + O(\Omega^2/\omega_L^2)$, which describe quasiuniform rotations. Such a rotation can be induced in superfluid ^3He by instantaneously changing the static field H_0 by an amount $|\Delta H|$ comparable to H_0 . The order parameter then starts to rotate with angular frequency $\Lambda = |\gamma \Delta H|$, and the spin oscillates at frequency 2Λ . This phenomenon has been referred to as longitudinal nonlinear "ringing."⁸ In the rest of this paper we will use the LT equations to analyze stability for nonlinear ringing in a resonant ac field. The analysis will be carried out in two steps. First, in this section we examine the stability problem for fast rotations of the order parameter d , then in the next section we investigate the stability of the longitudinal geometry.

A. Free fast rotation of the order parameter is stable

Usually, rapid oscillations of pendulums become unstable due to relaxation, and in fact we will see that the LT relaxation mechanism does ultimately destabilize rapid rotations of d . Nevertheless, as might be expected, the instability grows much more slowly when d rotates rapidly because the relaxation terms depend periodically on the phase of the rotation [cf. Eqs. (4), (6)].

Using the method of slowly varying parameters developed by Moiseev⁹ to describe rapid oscillations of a pendulum, one can show that for $\Lambda \gg \Omega \gg \Gamma$ the solutions of Eqs. (4), (6) with $\omega_1 = 0$ are given by

$$\Phi = \psi(t) + a(t) \sin 2\psi(t) \quad (A\text{-phase}), \quad (9)$$

$$\Phi = \psi(t) - a(t) (\sin \psi(t) + \frac{1}{2} \sin 2\psi(t)) \quad (B\text{-phase}),$$

where $\psi(t)$ and $a(t)$ vary slowly with time and satisfy

$$\dot{a} = p\Gamma a^2, \quad \dot{\psi} = q\Omega/\sqrt{a}, \quad (10)$$

where

$$p_A = 2, \quad p_B = \frac{4}{3}, \quad q_A = 1/\sqrt{2}, \quad q_B = 4/\sqrt{15}, \quad a_A(0) = \Omega_A^2/2\Lambda^2, \\ a_B(0) = 16\Omega_B^2/15\Lambda^2, \quad \psi(0) = \Lambda.$$

It is clear from (10) that the "amplitudes" a increase with a logarithmic growth rate $\lambda \sim \Gamma a(0)$. We stress that λ is proportional to the small parameter $a(0) \ll 1$ because of the periodic dependence of the LT relaxation terms on the rapidly changing phase Φ . The rotation of d is thus damped over times $1/\lambda \sim \Lambda^2/\Gamma\Omega^2$ greatly exceeding the LT relaxation time, and d rotates many ($\sim \Lambda^3/\Gamma\Omega^2$) times before significant damping sets in.²⁾ If we consider the motion of d only for times $t \lesssim 1/\lambda$, we can thus neglect the destabilizing relaxation terms in Eqs. (4), (6) and regard free rapid rotation as stable.

B. Frequency broadening of nonlinear ringing induced by a periodic series of radio-frequency pulses

We now assume that $\omega_1 \neq 0$ in Eqs. (4), (6). Clearly, the ac field will then significantly alter the rapid rotations if its frequency $\omega \approx 2\Lambda$ is nearly twice the rotation frequency of

d. We will refer to this as the resonance condition.

A similar type of resonant pendulum rotation was analyzed in Ref. 9 for the case $\omega_1 \ll \Lambda$ by the methods of nonlinear mechanics. It was found that the rotation was unstable, regardless of the dissipation mechanism, and this result clearly applies to our situation. We will therefore analyze a somewhat different NMR problem in which a system is pumped by a periodic train of rf pulses; such pulses can induce stochastic behavior¹⁰ in spin systems through the "resonance interaction" effect. We will show that the rf pulses may produce a slow frequency broadening of the nonlinear ringing.

Let a train of pulses

$$\tilde{h}(t) = h \sum_{m=-\infty}^{+\infty} g\left(\frac{t}{T} - m\right) \sin \omega t, \quad \omega \tau_p \gg 1, \quad \tau_p \ll T \quad (11)$$

parallel to the static field act on the system, where $g(t)$ is the pulse shape function and τ_p and T are the pulse length and repetition period, respectively. We assume that the field (11) is weak in the sense that $\omega_1 \ll \omega$ and will consider the spin dynamics only for times $t \lesssim 1/\lambda$, i.e., the pulse train terminates before the relaxation starts. If we substitute (11) into Eqs. (4), (6) and impose the initial condition $\Lambda \gg \Omega$, we find that they have solutions of the form (9) in which the slowly varying functions $a(t)$ and $\phi(t) = 2\psi(t) = \omega t$ satisfy the Hamiltonian system

$$\dot{a} = \eta a^{3/2} \cos \varphi \sum_{m=-\infty}^{+\infty} g\left(\frac{t}{T} - m\right), \quad (12)$$

$$\dot{\varphi} = \frac{2q\Omega}{a^{3/2}} - \omega - \frac{1}{2} \eta a^{3/2} \sin \varphi \sum_{m=-\infty}^{+\infty} g\left(\frac{t}{T} - m\right),$$

where $\eta_A = \sqrt{8} \omega_1 \omega / \Omega_A$ and $\eta_B = -\sqrt{15} \omega_1 \omega / 2\Omega_B$. In deriving (12) we have assumed that $\omega \tau_f \gg 1$, where τ_f is the total rise and fall time of the pulses.

Since we may assume that $|2\Lambda - \omega| \gg |\eta| a^{5/2}(0)$ in (12), we see that it describes nonlinear oscillations of the phase ϕ and slow "amplitude" a . In general, these oscillations are unstable. We will consider only stochastically unstable oscillations generated by the interaction among the resonances; the analogous instability for Hamiltonian systems of the type (12) is well understood.¹⁰

Based on the results of Ref. 10, we can show that if the Chirikov condition $K = c\omega_1 \omega \tau_p T a(0) \gg 1$ for stochastic behavior is satisfied (with $c_A = 2$ and $c_B = 1$) then the phase becomes chaotic during times $\tau = 2T/\ln K$, while the distribution function of the "amplitude" a obeys the Fokker-Planck equation

$$\frac{\partial \rho}{\partial t} = D \frac{\partial^2}{\partial a^2} (a^3 \rho) + G \frac{\partial}{\partial a} (a^4 \rho) \quad (13)$$

for times $t \gg \tau$, where $D = (\eta \tau_p)^2 / 4T$, $G = -7D/2$. Equation (13) yields

$$\frac{d}{dt} \bar{a} = -G \bar{a}^4, \quad \frac{d}{dt} \overline{a^2} = \frac{9}{2} D \bar{a}^5 \quad (14)$$

for the first two moments of ρ . Since no method is available

for analyzing (14) in general, we consider the case $\bar{a}^4 \approx a^4(0)$, $\overline{a^2} \approx a^2(0)$, which corresponds to the initial growth stage of \bar{a} and $\overline{a^2}$. It is then obvious that

$$\overline{\Delta a} = |G| a^4(0) t, \quad \overline{(\Delta a)^2} = 2D a^5(0) t, \quad (15)$$

where $\Delta a = a - a(0) \ll a(0)$. The initial growth stage of the moments is thus confined to times $t \ll 1/D a^3(0)$.

We can use (15) to calculate the shift and linewidth of the nonlinear longitudinal NMR for times $t \gg \tau$. Indeed, expanding the frequency 2ψ in powers of $a - a(0)$ and averaging over ρ , we get

$$\sigma_1 = -2\Lambda D a^3(0) t, \quad \sigma_2 = \pm 2\Lambda [D a^3(0) t / 2]^{1/2} \quad (16)$$

for the shift and linewidth, respectively. Since we consider the case when $D a^3(0) t \ll 1$, (16) implies that $|\sigma_2| \gg |\sigma_1|$, i.e., the broadening completely obscures the shift. The effect of the periodic series of resonant rf pulses is thus to slowly broaden the frequency of the ringing signal (or else to modulate it stochastically), as described by (16).

We conclude this section with some quantitative estimates. If for example we suddenly decrease the field $H_0 \approx 200$ G by an amount $\Delta H \sim H_0$ and then apply a series of rf pulses with $h \sim 10$ G, $\tau_p \sim 10^{-5}$ s, $T \sim 10^{-3}$ s, and $\omega \approx 4 \cdot 10^6$ s⁻¹, we find that $K \sim 10$. The order parameter will thus rotate rapidly in a randomly modulated way, and hence the spin oscillations along the ac field $h(t)$ will also be stochastically modulated in the frequency range $2\Lambda \pm |\sigma_2|$. Toward the end of the pulse train, typical values of $|\sigma_2|$ in the *A* and *B* phases are $|\sigma_{2A}| \approx 6 \cdot 10^4$ s⁻¹ and $|\sigma_{2B}| \approx 2 \cdot 10^4$ s⁻¹.

3. STABILITY OF THE LONGITUDINAL GEOMETRY FOR A RAPIDLY ROTATING ORDER PARAMETER

Two cases must be considered: 1) the field H_0 changes instantaneously at time $t = 0$ but remains strong enough so that the adiabatic approximation is valid⁷; 2) H_0 turns off completely and instantaneously at $t = 0$. Although the rotation is rapid in either case [see Eqs. (9)], additional analysis is required to study the stability of the longitudinal geometry.

Indeed, because the adiabatic approximation holds for case 1) (Ref. 7), we can use Eqs. (3) and (5) to decide the stability question. Substituting Φ from (9) into (3), (5) and averaging over the fast phase ψ , we find that the longitudinal geometry is stable.

Since no static field is present, the adiabatic approximation breaks down in case 2) and the analysis must be based on the full system of Leggett-Takagi equations.

We first consider the *A* phase and assume that the field is completely turned off at $t = 0$. Then the phase (9) describes the rotation in the plane normal to the ac field, which for simplicity we take to be monochromatic. We follow Ref. 5 and consider the transverse spin (δS_x , δS_y) and the longitudinal order-parameter (δd_z) fluctuations, which distort the longitudinal geometry. The LT equations then lead to the system⁵

$$\begin{aligned}\delta\dot{S}_x &= \omega_1 \delta S_y \sin \omega t - \Omega_A \delta d_z \cos \Phi, \\ \delta\dot{S}_y &= -\omega_1 \delta S_x \sin \omega t,\end{aligned}\quad (17)$$

$$\delta\dot{d}_z = \Omega_A \delta S_x \cos \Phi + \Omega_A \delta S_y \sin \Phi - \Gamma_A \delta d_z \cos^2 \Phi,$$

where Φ satisfies (4) with the initial condition $\Lambda = |\gamma| H_0 \gg \Omega_A$. The problem clearly reduces to analyzing the stability of the trivial solution $\delta S_x = \delta S_y = \delta S_z = 0$ of system (17). Under our previous assumptions $\omega_1, \Omega_A \ll \omega \approx 2\Lambda$, we readily see that (17) describes oscillations in the fluctuations which are slow compared to the frequency of the ac field (the nonlinear ringing frequency). We can therefore examine the stability at the trivial point by averaging Eqs. (17) over a time $\sim 4\pi/\omega$ [cf. Ref. 11]. We note that because the small parameter Ω_A/ω is not present in the equations for oscillating d , (17) must be analyzed numerically.⁵

If we apply the averaging technique in Ref. 11 to (17), we find that the averaged fluctuations satisfy

$$\begin{aligned}\delta\dot{S}_x &= \frac{\Omega_A^2}{\omega} \delta S_y - \frac{\omega_1 \Omega_A^2}{2\omega^2} \left(1 + \frac{\Gamma}{\omega_1}\right) \delta S_x, \\ \delta\dot{S}_y &= -\frac{\omega_1 \Omega_A^2}{2\omega^2} \delta S_y, \quad \delta\dot{d}_z = -\frac{1}{2} \Gamma_A \delta d_z\end{aligned}\quad (18)$$

to third order in $2\omega_1/\omega \sim 2\Omega_A/\omega$. In deriving (18) we have taken $\Phi = \omega t/2$ as the solution of Eq. (4). It follows from (18) that the trivial solution of (17) is asymptotically stable, because all the eigenvalues of the coefficient matrix for (18) have negative real parts. One can show that the longitudinal geometry is also stable if a train of rf pulses acts on the system and the rapid rotation of d becomes stochastically modulated, because the large regular component of the rotation frequency of d is decisive. The longitudinal geometry in the A phase will thus be undisturbed if a strong static field is turned off instantaneously.

Let us now consider the B phase. Here the unperturbed longitudinal geometry corresponds to the situation when the magnetic field is parallel to the spin S and to the rotation axis n of d ; this configuration becomes distorted when components n_\perp and S_\perp transverse to the field are excited. Writing the dynamic spin equations for the B phase in Fomin's variables,¹² we find the system

$$\begin{aligned}\delta\dot{S}_\beta &= \frac{2\chi\Omega_B^2}{15\gamma^2} \delta\beta (1 + \cos \Phi) (1 + 4 \cos \Phi), \\ \delta\dot{\beta} &= \frac{\gamma^2}{\chi} \delta S_\beta + \frac{2}{15} \Gamma_B \delta\beta (1 + \cos \Phi) (1 + 4 \cos \Phi)\end{aligned}\quad (19)$$

for the fluctuations that distort the longitudinal geometry. Here S_β is the projection of the spin onto the moving β axis normal to the field, and $\sin \beta$ is proportional to the transverse components n_x, n_y of n ; we have used $\sin \delta\beta \approx \delta\beta$ in (19). We observe that (19) involves the magnetic field implicitly through the field-dependence of the phase Φ . If the strong static field is turned off instantaneously, Φ is given by (9) and describes a rapid rotation of d about the axis n .

We cannot use the averaging method in Ref. 11 to analyze the stability of the trivial point of system (19), because

the second equation contains no small parameter. We therefore apply the Liouville-Jacobi theorem to find the time behavior of the solutions of (19). If $M(t)$ is a matrix whose columns consist of two independent solutions of (19), then

$$\det M(t) = \det[M(0)] \exp\left(\int_0^t \text{Sp } \hat{P}(t') dt'\right), \quad (20)$$

where the elements of the matrix \hat{P} are the coefficients of the unknown functions in the right-hand sides of (19), so that

$$\text{Tr } \hat{P} = \frac{2}{15} \Gamma_B (1 + \cos \Phi) (1 + 4 \cos \Phi).$$

Since shortest time scale of the problem is $2\pi/\Lambda$, it is natural to study the behavior of (20) for $t \gg 2\pi/\Lambda$, which in mathematical terms requires evaluating the limit

$$\lim_{t \rightarrow \infty} \int_0^t \text{Tr } \hat{P}(t') dt' = \frac{2}{5} \Gamma_B(t). \quad (21)$$

Here we have used expression (9) for Φ and neglected small corrections of order a to the right-hand side of (21). The result (21) implies that when the static field is turned off instantaneously, the longitudinal geometry in the B phase becomes distorted rapidly, well before the rotation of the order parameter starts to become damped. Stochastic modulation of the ringing frequency in the B phase can thus be observed only in static fields that remain sufficiently strong.

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¹As in the adiabatic approximation, the angle between S and d is conserved,⁷ so that for our purposes it suffices to analyze the time dependence of the fluctuations in one of the variables, e.g., the spin.

²In the experiments on nonlinear ringing in Ref. 8, the ringing frequency was measured during a time $\ll 1/\lambda$, so that no instability was detected in the rotation.

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