

New York, 1971), Vol. 26, p. 338.

⁴One expects the molecules to be incident on the target in various excited states primarily determined by conditions in the accelerator ion source. The average internuclear separation \bar{r}_0 will therefore differ slightly from the value for the molecular ground state. The curves shown in Fig. 1 were calculated with $\bar{r}_0 = 0.8$ and 1.1 \AA and with Gaussian widths (fwhm) of 0.5 and 0.7 \AA for $(^4\text{HeH})^+$ and D_2^+ , respectively. For further details, see the discussion by J. Remillieux, in Proceedings of the Fifth International Radiation Congress, Seattle, Washington, 1974 (to be published).

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NMR in $A\text{-}^3\text{He}$ and $B\text{-}^3\text{He}$: The Intrinsic Relaxation Mechanism

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(Received 18 February 1975)

A previously developed theory of NMR in superfluid ^3He is generalized to include relaxation effects. The physical mechanism envisaged is the relaxation of nonequilibrium Cooper-pair concentrations in the different spin bands. The results should describe both linear and nonlinear phenomena for all frequency regimes. The results for continuous-wave linewidths disagree with those of Combescot and Ebisawa as to temperature dependence.

If we assume, as is by now conventional, that the A phase of liquid ^3He is the ABM (Anderson-Brinkman-Morel) state¹ and the B phase the BW (Balian-Werthamer) state,² then most of the gross features of the dynamic nuclear magnetism in both phases³ can be understood in terms of the equations derived previously⁴ from a Born-Oppenheimer type of approximation. These equations are equations of motion for the total spin \vec{S} and the vector $\vec{T}(\vec{n})$ [or $\vec{d}(\vec{n})$] which describes the amplitude and axis of spin polarization of the Cooper pairs at point \vec{n} on the Fermi surface:

$$d\vec{S}/dt = \gamma\vec{S} \times \vec{\mathcal{H}}(t) + \vec{R}_D \{ \vec{T}(\vec{n}) \}, \quad (1)$$

$$d\vec{T}(\vec{n})/dt = \vec{T}(\vec{n}) \times \vec{H}. \quad (2)$$

Here $\vec{\mathcal{H}}(t)$ is the external magnetic field (including the radio-frequency field, if any) and \vec{R}_D is the "dipole torque" and is a bilinear function of $\vec{T}(\vec{n})$ and $\vec{T}^\dagger(\vec{n})$ for whose specific form we refer to Ref. 4. \vec{H} is defined as $-(\partial E/\partial \vec{S})$; in particular, in the case of longitudinal resonance, it is the difference of the up- and down-spin chemical potentials. To get a closed set of equations we need to supplement Eqs. (1) and (2) with an expression for \vec{H} in terms of \vec{S} and $\vec{\mathcal{H}}$. In the zeroth-order adiabatic approximation used in Ref.

4 the system is taken to be always in equilibrium for the given values of \vec{S} and $\vec{T}(\vec{n})$; then (neglecting for the moment the question of "susceptibility anisotropy") the requisite relation is

$$\vec{H} = \gamma\vec{\mathcal{H}}(t) - \gamma^2\chi^{-1}\vec{S}. \quad (3)$$

The theory based on Eqs. (1)–(3) seems to be reasonably successful in accounting for a variety of phenomena observed in the dynamic nuclear magnetism of the new phases.^{3–6} However, it has the major defect that there is no point at which relaxation is introduced. Experimentally, on the other hand, very substantial effects of damping and relaxation are seen, although it is not always easy to disentangle intrinsic effects from those which may be due to inhomogeneity of sample orientation and/or rf field. A theory of the damping of the cw resonance has been given by Combescot and Ebisawa⁷ on the basis of a kinetic-equation approach; however, the generalization to nonlinear phenomena, though possible,⁸ is not trivial, and there may be, therefore, some value in an attempt to set up a more intuitive, less mathematically complex approach to the problem.

In this Letter we attempt to generalize Eq. (3)

so as to give a general theory of relaxation which will apply equally to linear and nonlinear phenomena. The resulting theory is only slightly more complicated than the original zero-order adiabatic approximation, and enables us to treat all frequency regimes in a unified way. In the cw limit the results, although similar in many respects to those of Ref. 7, give a different temperature dependence for the linewidths near T_c .

To see the basic physical idea, let us for the moment concentrate on the A -phase longitudinal resonance in zero external field. This may be regarded as a Josephson-like phenomenon in which the Cooper pairs tunnel between the up- and down-spin bands under the influence of the weak, spin-nonconserving, dipole force.⁴ Imagine that we do a thought experiment as follows: Starting from a situation in which \vec{S} is 0 but the $\vec{T}(\vec{n})$ are slightly rotated away from their equilibrium configuration around the z axis, for time $0 < t < t_1$ we allow the dipole forces to act but switch off all collision processes, while for $t_1 < t < \infty$ we switch off the dipole forces (thereby enforcing rigorous spin conservation) but allow spin-conserving collision processes to occur. Since the normal component is completely unaffected by the tunneling process, the number of up-spin normal particles at t_1 will be the same as at $t=0$ (and, of course, the same as the number of down-spin normal particles). On the other hand, a number of Cooper pairs will have tunneled (say) from the down to the up band. The up-spin band is therefore *not in internal equilibrium* at $t=t_1$: There are too many Cooper pairs relative to the normal component.⁹ Consequently, the field $\vec{H} = -\partial E/\partial \vec{S}$ (the difference in the up- and down-spin chemical potentials) is not related to the spin polarization \vec{S} by the equilibrium susceptibility χ but by some "susceptibility at constant normal component" χ_c : $\vec{H} = -\gamma^2 \chi_c^{-1} \vec{S}$. Now when for $t > t_1$ collision processes begin to act, they will of course not change the total spin (i.e., the total number of particles in each band) but they will convert the surplus Cooper pairs in (say) the up-spin band into normal particles, until at $t = \infty$ the two bands are in complete internal equilibrium and we have $\vec{H} = -\gamma^2 \chi^{-1} \vec{S}$, where χ is the true equilibrium susceptibility. In the real situation, of course, the dipole forces and the collision processes act simultaneously, the latter with some characteristic relaxation time τ . Then if ω is the longitudinal resonance frequency, we see that in the limit $\omega\tau \rightarrow 0$ ("hydrodynamic" limit) Eq. (3) is correct, whereas in the opposite limit $\omega\tau \rightarrow \infty$ ("collisionless" limit) χ should be

replaced there by χ_c . In the intermediate regime \vec{H} is relaxing between $-\gamma^2 \chi_c^{-1} \vec{S}$ and $-\gamma^2 \chi^{-1} \vec{S}$ and is therefore partially out of phase with \vec{S} ; it is this which gives rise to damping.

Let us try to make this idea quantitative, considering now a quite general situation. Define a vector \vec{S}_p which represents the contribution to the spin polarization "from Cooper pairs" (a precise definition of \vec{S}_p will be given below) and suppose that in the limit of zero relaxation ($\vec{S} = \vec{S}_p$) the field $\vec{H} \equiv -\partial E/\partial \vec{S}$ is related to the spin by $\vec{H} = \gamma \vec{\mathcal{C}}(t) - \gamma^2 \chi_c^{-1} \vec{S}$, thereby defining the "Cooper-pair susceptibility" χ_c . Then define also a vector $\vec{\eta}$ by

$$\vec{\eta} \equiv \vec{S}_p - \vec{S}_{p0}(\vec{S}), \quad (4)$$

where \vec{S}_{p0} is the equilibrium value of \vec{S}_p for the given value of \vec{S} . (That is, $\vec{\eta}$ represents the extent to which the pairs are out of equilibrium with the normal component.) Evidently we have $\vec{S}_{p0}(\vec{S}) = (\chi_{c0}/\chi_0) \vec{S} \equiv \lambda(T) \vec{S}$, where the subscript zero indicates that the susceptibilities are *not* corrected for Fermi-liquid effects (cf. below). Provided that the small-field condition $H \ll \Delta/\hbar$ is fulfilled, we can take the field \vec{H} to be a linear function of \vec{S} and $\vec{\eta}$ [and of course of the external field $\vec{\mathcal{C}}(t)$]:

$$\vec{H} = \gamma \vec{\mathcal{C}}(t) - \alpha \vec{S} - \beta \vec{\eta}. \quad (5)$$

By considering the case of complete relaxation ($\vec{\eta} = 0$) and also the case of no relaxation ($\vec{S}_p = \vec{S}$) when we must get the result given above, we find

$$\alpha = \gamma^2 \chi^{-1},$$

$$\beta = \gamma^2 \frac{\chi_c^{-1} - \chi^{-1}}{1 - \lambda} = \gamma^2 \chi_{c0}^{-1} (\lambda \equiv \chi_{c0}/\chi_0). \quad (6)$$

In obtaining the last form for β we assumed that the Fermi-liquid effects on χ_c and χ have the simple form $\chi_c^{-1} = \chi_{c0}^{-1} + \frac{1}{4} Z_0 \chi_{n0}^{-1}$, etc. [where $\chi_{n0} \equiv \frac{1}{4} \gamma^2 \hbar^2 (dn/d\epsilon)$ and Z_0 is the usual Landau parameter ($\equiv 4F_0$)]. This is not necessarily strictly true if higher Landau parameters are important, but the error is likely to be small (and always negligible near T_c).

To complete the theory we need the equation of motion of $\vec{\eta}$. This has three terms: First of all, $\vec{\eta}$ [like \vec{S} but unlike $\vec{T}(\vec{n})$] represents a physical spin vector and hence precesses around the *physical* field, that is the sum of the external field and the molecular field $\vec{\mathcal{C}}_{mol} = -\frac{1}{4} Z_0 \gamma \chi_{n0}^{-1} \vec{S}$. Secondly, there is a contribution from the dipole torque; since the latter induces a change in \vec{S} only through changing \vec{S}_p (cf. above), the contribution to $d\vec{\eta}/dt$ is just $(1 - \lambda)$ times that to $d\vec{S}/dt$. Finally, $\vec{\eta}$ will tend to relax back to its equilibrium value, namely zero; we shall assume that

this relaxation is exponential with some lifetime $\tau(T)$ which is of the order of the normal-particle lifetime. Taking all these terms into account we find

$$d\vec{\eta}/dt = \vec{\eta} \times \{ \gamma \vec{\mathcal{C}}(t) - \gamma^2 \chi_{n0}^{-1} (\frac{1}{4} Z_0) \vec{\mathcal{S}} \} + (1 - \lambda) \vec{R}_D - \vec{\eta}/\tau \quad (7)$$

while a combination of (5) and (6) gives

$$\vec{H} = \gamma \vec{\mathcal{C}}(t) - \gamma^2 \chi^{-1} [\vec{\mathcal{S}} + \lambda^{-1} (\chi/\chi_0) \vec{\eta}]. \quad (8)$$

Equations (7) and (8) replace (3) and, with (1) and (2), are assumed to give a complete description of the spin dynamics including relaxation.¹⁰

We must next determine the parameter λ occurring in Eqs. (7) and (8). To do this we need a precise definition of the quantity $\vec{\mathcal{S}}_p$. We will consider first the ABM state and in particular the case of longitudinal oscillation; then the vector \vec{d} remains in the x - y plane and $\vec{\mathcal{S}}_p$ is along the z axis. If we neglect the weak dipole forces, the up- and down-spin bands can be considered separately: Let us focus on a given pair of plane-wave states ($\vec{k}\uparrow, -\vec{k}\uparrow$) in the up band. The space associated with these two plane-wave states is spanned by the four orthogonal basis vectors $|00\rangle, |10\rangle, |01\rangle, |11\rangle$, where for instance $|10\rangle$ denotes the occupation-number eigenstate in which $\vec{k}\uparrow$ is full and $-\vec{k}\uparrow$ empty.

In BCS-type theories, the energy eigenstates associated with the pair are the two broken-pair states $|10\rangle$ and $|01\rangle$, and two orthogonal linear combinations (ground pair and excited pair) of $|00\rangle$ and $|11\rangle$ with amplitudes ($u_{\vec{k}\uparrow}, v_{\vec{k}\uparrow}$) and ($v_{\vec{k}\uparrow}^*, -u_{\vec{k}\uparrow}^*$), respectively. The coefficients $u_{\vec{k}\uparrow}, v_{\vec{k}\uparrow}$ are determined in the usual way in terms of the gap $\Delta_{\vec{k}\uparrow}$ and the energy $\epsilon_{\vec{k}\uparrow}$ measured from the up-spin chemical potential μ_{\uparrow} . Relative to the ground-pair state the broken-pair states have energy $E_{\vec{k}\uparrow} \equiv (\epsilon_{\vec{k}\uparrow}^2 + |\Delta_{\vec{k}\uparrow}|^2)^{1/2}$ and the excited-pair state $2E_{\vec{k}\uparrow}$, and in thermal equilibrium the occupation probabilities are given by the corresponding Boltzmann factors. The total number of particles in the up-spin band (hence the total spin) is a function both of the coefficients $u_{\vec{k}\uparrow}, v_{\vec{k}\uparrow}$ and of the occupation probabilities, and if μ_{\uparrow} is changed (e.g., so that $\delta\mu_{\uparrow} = -\delta\mu_{\downarrow} \ll \Delta$) then both these factors must adjust in order to produce the new equilibrium. We shall define $\vec{\mathcal{S}}_p$ as the change in spin induced by the change of $u_{\vec{k}\uparrow}$ and $v_{\vec{k}\uparrow}$ (and $u_{\vec{k}\uparrow}$ and $v_{\vec{k}\uparrow}$) alone without change of the occupation probabilities. Noticing now that the field $\vec{H} \equiv -\partial E/\partial \vec{\mathcal{S}}$ is just $(\mu_{\uparrow} - \mu_{\downarrow})/\hbar$, recalling the definition $\vec{\mathcal{S}}_p \equiv \gamma^{-2} \chi_c \vec{H}$ appropriate to the situation considered, and neglecting

Fermi-liquid effects (so that what is calculated is χ_{c0} rather than χ_c) we find after a straightforward calculation

$$\chi_{c0} = \frac{1}{4} \gamma^2 \hbar^2 (dn/d\epsilon) [1 - f(T)], \quad (9)$$

where $f(T)$ is the function defined by Combescot and Ebisawa⁷:

$$f(T) \equiv \int \frac{d\Omega}{4\pi} \int_0^\infty d\epsilon \frac{\epsilon_{\vec{k}}^{-2}}{E_{\vec{k}}^{-2}} \frac{1}{2} \beta \operatorname{sech}^2 \frac{1}{2} \beta E_{\vec{k}}. \quad (10)$$

Hence the function $\lambda(T)$ appearing in Eqs. (7) and (8) is given for the ABM state by

$$\lambda(T)_{\text{ABM}} = 1 - f(T). \quad (11)$$

We notice that λ tends to 1 in the low-temperature limit, but near T_c is proportional to $(T_c - T)^{1/2}$.

For the BW state we use the fact that for any given point \vec{n} on the Fermi surface with pair-quantization vector $\vec{d}(\vec{n})$ defined in the usual way, the contribution to χ_{c0} must be a tensor quantity of the form $\text{const} [\delta_{ij} |\vec{d}(\vec{n})|^2 - d_i(\vec{n}) d_j^*(\vec{n})]$ (this follows because from its definition, χ_{c0} has no contribution from the normal component). After averaging over the Fermi surface, we find the simple result

$$\lambda(T)_{\text{BW}} = \frac{2[1 - f(T)]}{2 + Y(T)}. \quad (12)$$

[Of course, $f(T)$ is not exactly the same function for the two states since $E_{\vec{k}}$ has a different form.] Note also that χ/χ_0 is simply $(1 + \frac{1}{4} Z_0)^{-1}$ for the ABM state and $\{1 + \frac{1}{4} Z_0 [\frac{2}{3} + \frac{1}{3} Y(T)]\}^{-1}$ for the BW state, where $Y(T)$ is the Yosida function.

Equations (1), (2), (7), and (8) have numerous obvious applications. There is space here only to quote a few without derivation (we hope to give a more extensive discussion elsewhere).

cw-resonance linewidths.—In the hydrodynamic limit ($\omega_D \tau \ll 1$, $\omega_L \tau \ll 1$, where ω_D is the longitudinal resonance frequency) we find results identical to those of Combescot and Ebisawa [Eqs. (9) and (11) of Ref. 7], *except*¹¹ for an extra factor of λ^{-1} [see Eqs. (11) and (12) above] in each case. As a result we predict that near T_c the longitudinal linewidth behaves as $(T_c - T)^{1/2}$ rather than as⁷ $(T_c - T)$. This seems to indicate that our assumption about relaxation of the nonequilibrium Cooper-pair concentration cannot be equivalent to that presumably embodied in their Eq. (1).

Nonlinear longitudinal resonance.—If θ represents the angle of rotation of the vectors $\vec{d}(\vec{n})$ in the x - y plane (or half the phase difference between the up- and down-spin pairs)^{3,4} and $E_D(\theta)$ is the

dipole energy as a function of this angle, we obtain¹²

$$\ddot{\theta} + \frac{1}{\tau} \dot{\theta} + \gamma^2 \chi_c^{-1} \dot{\theta} \frac{\partial^2 E_D}{\partial \theta^2} + \gamma^2 \chi^{-1} \frac{\partial E_D}{\partial \theta} = 0. \quad (13)$$

This equation can be applied to either phase by a suitable choice of the function $E_D(\theta)$.

Relaxation in pulse-type experiments.—In the limit $\omega_D \ll \omega_L$, $\omega_L \tau \ll 1$ (which should describe most pulse experiments conducted to date) we find that after a large-angle pulse the recovery of the energy (when averaged over several Larmor periods) is linear rather than exponential¹³ with an effective recovery time $T_{1,\text{eff}}$ given by

$$T_{1,\text{eff}} = \alpha \left(\frac{\omega_L}{\omega_D} \right)^2 \Gamma_{\parallel}^{-1}, \quad \Gamma_{\parallel} \equiv \frac{\chi}{\chi_0} \left(\frac{1-\lambda}{\lambda} \right) \omega_D^2 \tau, \quad (14)$$

where Γ_{\parallel} is in fact the width of the cw longitudinal resonance in this limit [cf. Eq. (13) above] and α is a numerical factor of order unity. It seems reasonable to assume that M_z , the z component of magnetization, recovers in a similar way. Since Γ_{\parallel} varies as $(T_c - T)^{1/2}$ near T_c , we see that $T_{1,\text{eff}} \propto H^2 (T_c - T)^{-3/2}$. Very recently, Corruccini and Osheroff¹⁴ conducted systematic pulse experiments and observed, in the A phase, just such a linear relaxation of M_z ; however, they found $T_{1,\text{eff}} \propto H (T_c - T)^{-1}$. It seems probable, therefore, that the relaxation they saw was not the intrinsic relaxation considered in this paper but was, as they suggest, due to the inflow of magnetization supercurrents from the unperturbed region outside the rf coil, or to some other extrinsic effect.

One of us (A.J.L.) is grateful to Pierre Hohenberg, Phil Anderson, and Bill Brinkman for a very stimulating discussion which sowed the seeds of some of the above ideas. We are also grateful to Roland Combescot and Vinay Ambegaokar for

very helpful correspondence.

*Work supported by the Nishina Memorial Foundation.

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¹⁰The above argument, and in particular Eq. (5), ignores the effects of susceptibility anisotropy (Ref. 4). There is unfortunately no space to discuss this question here but we believe that in all cases the corrections are at most of order $\Gamma_{\parallel} / \omega_D (\ll 1)$, where Γ_{\parallel} and ω_D are defined below.

¹¹That is, provided our τ is identified with theirs (as seems natural, since both are taken to be of the order of the normal-quasiparticle lifetime).

¹²Equations of a similar form have been obtained by R. Combescot and V. Ambegaokar (private communication).

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