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Proposed NMR determination of the exchange parameters in hexagonal-close-packed ^{3}He

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We calculate the relaxation times for an oriented single crystal of hexagonal-close-packed solid ^{3}He, which, according to the theory of Roger and co-workers, is dominated by triple exchange. The anisotropy of the relaxation times is a function of the ratio of exchange rates in the basal plane to those out of the plane and may be used to determine this ratio. Results are presented for T_1 and T_2 as a function of frequency as well as of crystal orientation.

The nature of exchange in solid ^{3}He has been a topic of investigation for many years. The low-pressure bcc phase has received most of the experimental and theoretical attention and it has been demonstrated that three- and four-body-exchange processes are important to the magnetic properties of that phase.[1] While much less attention has been paid to the hexagonal phase, a recent experiment[2] has shown that the Néel temperature of this phase is positive in agreement with the theoretical predictions that three-body exchanges should make this phase ferromagnetic.[1,3] Here we suggest that NMR experiments may be performed that can lead to better information concerning the exchange constants in the hexagonal-close-packed (hcp) phase.

Roger[3] has estimated the exchange constants in both bcc and hcp ^{3}He. He finds that magnetic processes in the hcp phase are dominated by three-particle exchanges which will lead to a ferromagnetic spin alignment at low temperatures. Pair and higher-order exchanges may also take place. There are three types of triangles made up of nearest neighbors in the hcp lattice: two completely in the basal plane and the other having two particles in the basal plane and the third out of the plane. The exchange constants for these triangles are not identical because of the different environments of the triangles.

A triple-exchange operator can be decomposed into *pair*-exchange operators[3] so that, if we neglect the four-particle and higher-order processes, we can write an effective exchange Hamiltonian for hcp ^{3}He in the usual form

$$H = -\tfrac{1}{2}\sum_{i<j} J_{ij}\boldsymbol{\sigma}_i\cdot\boldsymbol{\sigma}_j\,, \tag{1}$$

where $\boldsymbol{\sigma}_i$ is a Pauli matrix and $J_{ij}=J$ for a pair of particles in a basal plane and $J_{ij}=J'$ for a pair of particles having one member in the basal plane and the other out of the plane. Roger's calculations give $J\approx J'$. There has not yet been an experimental confirmation of this prediction. J and J' can include the effects of real pair-exchange integrals in the hcp lattice. However, we will neglect any second-neighbor pair exchanges.

We would like to show that anisotropy in NMR on an oriented hcp crystal of ^{3}He can lead to a strong indication of the relative sizes of J and J'. We calculate T_1 and T_2 by using Gaussian approximations for the correlation functions. To see the nature of our results we consider a special simple case. The adiabatic part of the spin-spin relaxation time is given by a standard expression[4]

$$(T_2^{-1})_{\mathrm{ad}} = \sqrt{(\pi/2)M_2^3/M_4}\,, \tag{2}$$

where M_2 and M_4 are moments of the resonance line.

To treat an oriented crystal, we make a restricted angular average. If the magnetic field has polar angle β with the c axis of the crystal and azimuthal angle ϕ_B with an arbitrary direction in the basal plane, then we average over just ϕ_B. T_2 will be a function of the angle β. Experimental verification of our predictions will require a single crystal with the c axis in a known direction. General expressions that include results for M_2 and M_4 in this averaging process will be given below.

Figure 1 indicates, that when the ratio $Y=J'/J$ is small, there is considerable anisotropy in T_2^{-1} while for larger Y values the dependence on β is considerably reduced. To understand this result physically we note that dipolar lattice sums in the hcp crystal are mildly dominated by the particles in the basal plane. In this regard, they behave somewhat like those in a triangular two-dimensional (2D) lattice.[5] Indeed the angular behavior of M_2 here and in

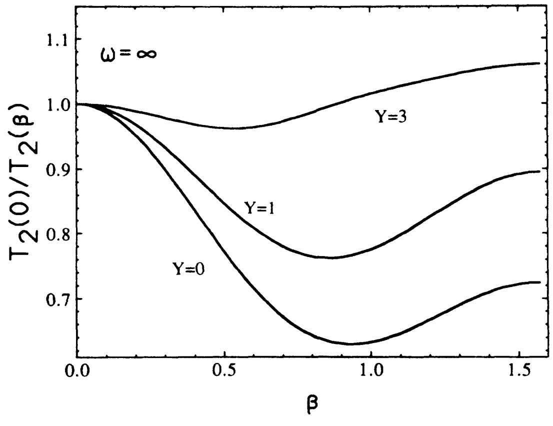

FIG. 1. The adiabatic portion ($\omega=\infty$) of the spin-spin relaxation time T_2 as a function of angle β in radians between the c axis and the external magnetic field [Eq. (2) or the first term of Eq. (5)]. Triple exchange is represented by effective pair exchange integrals. The variable Y is J'/J where J is the pair exchange integral for two near neighbors in the basal plane, and J' is that for particles in two different planes.

the 2D crystal are similar in that both reach a minimum around $\beta = \pi/4$. To see this behavior, note that the angular dependence of M_2 is just the average of $\frac{1}{4}(3z^2-1)^2$ where z is the component along the field of a unit interparticle vector $\hat{\mathbf{r}}_{ij}$. As shown in Ref. 5 in 2D, after an azimuthal angular average M_2 is proportional to $[1-3\sin^2\beta + \frac{27}{8}\sin^4\beta]$ which has a minimum at 42°.

Next, we repeat another argument from Ref. 5 in order to understand the behavior of $(T_2^{-1})_{\text{ad}} = M_2/\omega_c$, where ω_c is the correlation frequency. We expect that if there is anisotropy then the correlation frequency might be given by something like

$$\omega_c(\beta) = \tfrac{1}{2}(\omega_\parallel + \omega_\parallel \sin^2\beta + \omega_\perp \cos^2\beta)\,, \tag{3}$$

where $\omega_\parallel$ and $\omega_\perp$ are correlation frequencies for motion parallel and perpendicular to the basal plane. This result might arise in the following way: In a strictly 2D system ($\omega_\perp = 0$) with the magnetic field in the particle plane ($\beta = \pi/2$), motion of the spins both radially and axially modulates the interaction. On the other hand, at $\beta = 0$, the dipolar interaction is unchanged during rotational motion at constant $\hat{\mathbf{r}}_{ij}$ and we have effectively lost a degree of freedom with regard to motional narrowing. With $\omega_\perp$ set equal to zero, Eq. (3) has this expected qualitative dependence. On the other hand, in an isotropic 3D system with $\omega_\parallel = \omega_\perp$, ω_c loses its angular dependence as expected. The hcp system with $Y=0$ has (effective) two-particle exchange only in the basal plane and should behave somewhat like adsorbed ^{3}He. With the suggested functional forms we expect that $(T_2^{-1})_{\text{ad}}$ would drop off as β approaches $\pi/2$ and have a minimum near $\pi/4$. This is precisely what we see in Fig. 1.

When $\omega_\perp$ becomes larger than $\omega_\parallel$ we expect, from Eq. (3), that $(T_2^{-1})_{\text{ad}}$ will become large as β goes to $\pi/2$. Indeed, when Y is large, exchange out of the basal plane predominates and we do see the expected behavior in Fig. 1. Note, however, that very large Y never leads to an extreme anisotropy because J' implies both a parallel *and* a transverse part of ω_c. This is easy to see by examining the hcp lattice structure.

For the general case of both relaxation times for all frequencies, we follow Landesman[6] to show that

$$T_1^{-1} = J_1(\omega) + 4J_2(\omega)\,, \tag{4}$$

$$T_2^{-1} = \tfrac{3}{2}J_0(0) + \tfrac{5}{2}J_1(\omega) + J_2(\omega)\,, \tag{5}$$

where $J_m(\omega) = \sqrt{2\pi} g_m/\omega_m \exp(m^2\omega^2/2\omega_m^2)$ are the spectral functions and $\omega_m = (p_m/g_m)^{1/2}$ is a correlation frequency, and ω is the Larmor frequency. The spectral function J_m is the Fourier transform of the correlation function $G_m(t)$ corresponding to the m-spin-flip terms in the dipolar Hamiltonian. The quantities g_m and p_m are the coefficients of the zeroth- and second-order terms in the expansion of G_m in t. It is not difficult to show the relations $g_0 = \frac{1}{3}M_2$ and $p_0 = \frac{1}{3}M_4$. Equation (2) corresponds to the first term in Eq. (5).

After the restricted angular average described above we find that

$$g_m = \tfrac{3}{4}\hbar^2\gamma^4 \sum_{m'=0}^{2} \epsilon_{m'} D_{m'm}(\beta) u_{m'}\,, \tag{6}$$

where $\epsilon_{m'} = \frac{1}{2}$ for $m'=0$, $\frac{1}{6}$ for $m'=1$, and $\frac{1}{24}$ for $m'=2$. Also, we have $D_{m'm}(\beta) = \{[d^2_{m'm}(\beta)]^2 + [d^2_{-m'm}(\beta)]^2\}$; the $d^2_{m'm}(\beta)$ are the polar-angle part of the rotation matrices and can be found in many references.[7] Finally, the last factor is

$$u_m = (1/N)\sum_{ij} R_{ij}^{-6} P_2^m(\gamma_{ij})\,, \tag{7}$$

where R_{ij} is the interparticle separation at angle γ_{ij} with the c axis and $P_2^m(\gamma_{ij})$ is an associated Legendre polynomial. There are N particles in the lattice.

In a similar manner we find

$$p_m = \tfrac{3}{2}\hbar^2\gamma^4 \sum_{m'=0}^{2} \epsilon_{m'} D_{m'm}(\beta) v_{m'}\,,$$

where

$$v_m = (1/N)\sum_{ijk}\{R_{ij}^{-6}[P_2^m(\gamma_{ij})]^2 G_{ijk} + R_{ij}^{-3}R_{jk}^{-3}P_2^m(\gamma_{ij})P_2^m(\gamma_{jk})H_{ijk}\}\,. \tag{8}$$

The quantities G_{ijk} and H_{ijk} arise from doing complicated spin traces in the correlation functions. Because we are considering only effective pair exchanges, we can use the results derived in Van Vleck's fourth moment calculation.[4,8] We have

$$G_{ijk} = J_{ik}^2 + J_{jk}^2 + J_{ik}J_{jk} \tag{9}$$

and

$$H_{ijk} = -2J_{ij}^2 + J_{ik}J_{jk} - J_{ij}J_{jk} - J_{ij}J_{ik}\,. \tag{10}$$

Many of the lattice sums can be done analytically and others must be done numerically.

In 1975, Deville[9] treated the anisotropy of the NMR of a single crystal of ^{3}He both experimentally and theoretically. He assumed the isotropic case $Y=1$. He treated all possible orientations, in contrast with our restricted angular average, and as a consequence was unable to include the anisotropy of the three-body terms in M_4 ("unlike terms"). It is because we have averaged over the azimuthal angle ϕ_B that we are able to include these terms without difficulty.

Our result for the adiabatic part of T_2^{-1} corresponds to $\omega = \infty$. Figure 2 shows T_2^{-1} at the other extreme of $\omega = 0$. The anisotropy at small Y persists here although it is of a different character. Note that at the theoretically predicted value of $Y=1$, T_2^{-1} is quite independent of angle.

We also present results for T_1^{-1} at zero frequency in Fig. 3. Here, the anisotropy changes character as Y passes through 1. Actual experiments may be done at some intermediate frequency. We would be happy to use our programs to provide theoretical curves for the appropriate experimental situation or to provide the programs themselves.

We have shown curves in the form of ratios. Of course, the absolute values of T_1^{-1} and T_2^{-1} depend on Y as well. As J' increases for fixed J, each of these quantities decreases because the overall average exchange integral is getting larger.

Deville, by comparing his data[9] to his theoretical results, determined that all his hcp experiments were at

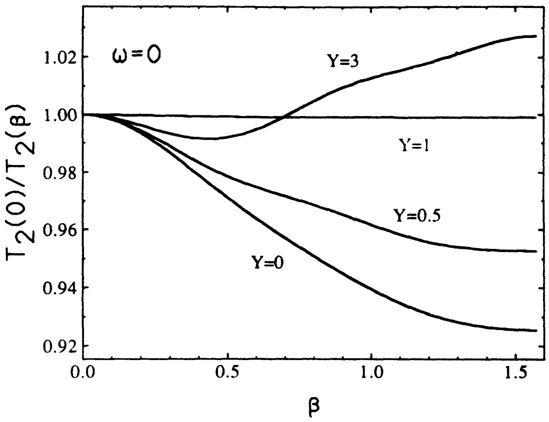

FIG. 2. T_2 from Eq. (5) in the limit $\omega=0$ as a function of β and Y. $Y=1$ corresponds to the theoretical prediction of Roger (Ref. 3).

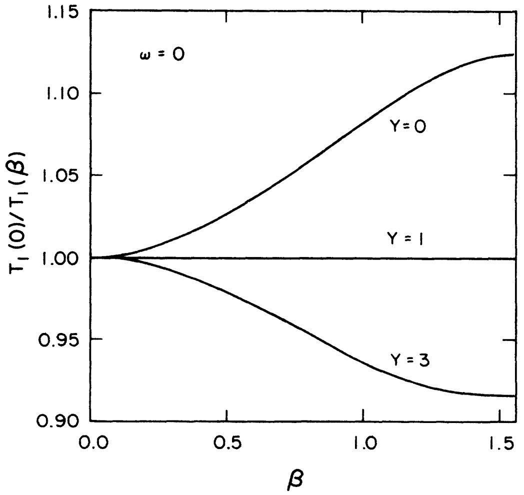

FIG. 3. The longitudinal spin relaxation time T_1 in the limit $\omega=0$ as a function of β and Y.

$\beta=\pi/2$. His angular dependence corresponds to a variation of ϕ_B. So, we are unfortunately unable to use his data to extract a value of Y.

We can easily reduce our results to a powder average by averaging over β. For example, we then find $M_4/M_2 = J^2(20.7+0.8Y+20.7Y^2)$.

Our results have been derived by neglect of the second-neighbor pair, four-particle, and any higher-order exchange integrals. After some of our work was done, we found that Matsumoto and Izuyama[10] had independently considered a somewhat similar treatment of T_1 in hcp ^{3}He. They did a complete angular average (i.e., they considered a powder) but included the effects of these higher-order exchange integrals. It should be possible to extend their work to the case of an oriented crystal.

We would like to thank Dr. Y. Takano who originally suggested to us that NMR on a single crystal might be a useful way to determine the exchange constants. We also appreciate useful conversations with Dr. B. Cowan, Dr. K. Matsumoto, Dr. G. Deville, and Dr. A. Landesman.

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