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SPIN ECHOES IN SPIN POLARIZED HYDROGEN

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We have conducted preliminary spin echo experiments in spin polarized hydrogen. The heights of the echoes follow a remarkable pattern, and we often see a regular series of additional peaks in the FID envelope which resemble multiple echoes. We conjecture that molecular fields, with due regard for boundary conditions, could account for most of the observed effects.

A typical ideal spin echo experiment would consist of a $\pi/2$ pulse applied at time $t = 0$ and a π pulse applied at $t = \tau$, forming an echo at $t = 2\tau$. Another π pulse applied at $t = 2\tau + \tau'$ forms an echo at $t = 2\tau + 2\tau'$.

Figure 1a shows some typical H↓ spin echo signals. There are normal echoes at $t = 2\tau$ and $t = 4\tau$, followed by several prominent peaks in the FID envelope at multiples of τ thereafter. This is the basic pattern of the data, which can be seen over a fair range of magnetic field gradient (≈0 to 2 G/cm) and spacing ($\tau \leq 300$ μsec). This pattern is also observed in $\pi/2$-π pulse experiments. Sometimes the recurrent peaks appear to ride atop a nonzero but gently varying background which may be due to spin waves excited by imperfections in the π pulses.

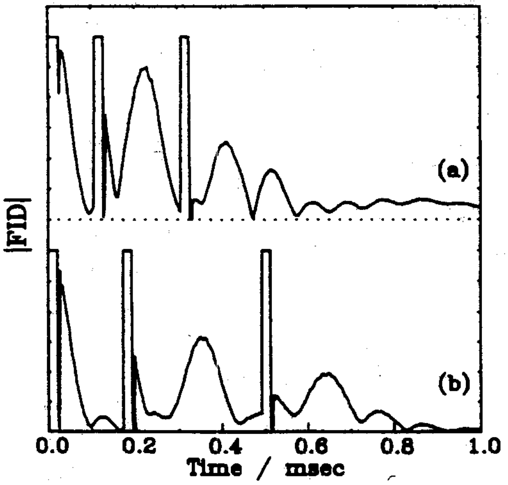

Figure 1. Typical H↓ spin echoes resulting from a $\pi/2$ - π - π pulse sequence. T ≈ 245 mK, n ≈ 4.3×10^{16}cm^{-3}, G ≈ 23 kHz/cm.

A variation from the simple pattern can be seen in figure 1b: although the first echo forms at the expected time, the second echo forms slightly earlier than expected, and then the multiple echoes form at progressively shorter intervals, all slightly shorter than τ and τ'. Sometimes the reverse is true: the second echo forms late and the intervals between echoes are progressively longer.

Often there is a prominent FID immediately following the first π pulse, as in figure 1b. This is despite the fact that we adjusted the π pulses to minimize the FID following an isolated π pulse. We do not observe any echo-like structures following an isolated π pulse. Note that FID following the second π pulse is much smaller, although we are reasonably certain that the two pulses were identical.

The magnitude of H_1 is the same for all pulses; the tipping angle was determined by the pulse duration. Nominally, all tipping pulses were applied about the same axis, although there was a slight drift of the tipping axis in the x--y plane. Magnet drift may have detracted from the accuracy of a few of the tipping pulses.

We do not have a clear picture of what causes the recurrent peaks. They may not be genuine multiple echoes, but we emphasize that the close correlation between peak position and τ is observed in a large fraction of FIDs, over the entire range of the data. It appears that these peaks are unrelated to the multiple echoes seen in superfluid ^{3}He (1). The theory (2) which explains the multiple echoes in solid ^{3}He (3) predicts no multiple echos for a $\pi/2$-π sequence, and it is unlikely that imperfections in our π pulses could give rise to the large effects we see. Perhaps in the absence of boundary effects (see below) we wouldn't see the additional peaks.

It is instructive to study the height of the first echo as a function of τ. We start with the hypothesis that the decrease of echo height is due to spin diffusion in a gradient $G = \vec{\nabla}(\gamma H_0)$. The formula of Torrey (4) is $s_+(2\tau)/s_+(0) = \exp\{-(2/3)\tau^3 G^2 D_{eff}\}$, where we are using the notation of reference (5). D_{eff} is some effective diffusion constant (see below). We have analyzed our data by plotting $\ln[s_+(2\tau)/s_+(0)]$ versus $\tau^3 G^2 D_{eff}$. If we neglect molecular field effects and set D_{eff} equal to the diffusion constant D_0 determined by other means (6), the data points do not fall on a straight line. Furthermore, they consistently fall at least a factor of 10 below the prediction of Torrey, i.e. the observed decay of transverse magnetization is anomalously fast.

We can try to account for molecular field effects by using a reduced diffusion constant $D_{eff} = D_0/(1+\mu^2 P^2)$. This collapses all the data from a variety of G and μP values onto a fairly straight line. This supports the idea that the echoes decay because of dephased spins diffusing in a gradient -- as opposed to some other process, e.g. diffusion limited surface relaxation. It also indicates that the molecular field has important effects on the diffusion. The fact that it is the slope (not the intercept) that is anomalous indicates that imperfections in the tipping pulses can not account for the decay.

On the other hand, the reduced diffusion constant makes the theoretical decay rate even **slower** than the observed rate. If one adjusts the value of D_0 to get agreement with the observed ratios of (echo/FID), then the theory would predict a much **faster** decay rate of the ratios of (second echo/first echo) than we observe. The full theory of Leggett and Rice (7) predicts an entirely different form of the echo decay anyway, which we can approximate by $1-[s_+(2\tau)/s_+(0)]^2 = \exp[-(2/3)\tau^3 G^2 D_{eff}]$. Our data does not fit this form.

There are three factors which complicate the analysis of this situation: the large molecular fields, the nonlinear effects of the large tipping angle, and the effects of the boundaries. Removing any one of these factors produces a relatively tractable system: Robertson (8) discussed spin echoes in the presence of boundaries, Lévy and Ruckenstein (9) discussed the effect of molecular fields and boundaries (for small tipping angles), and Leggett and Rice (7) and Leggett (10) discussed spin echoes in the presence of molecular fields. Leggett's equations do not have any boundary terms, which corresponds to the case of very distant or perfectly absorbing walls. Our studies (6) of spin waves indicate that reflecting boundary conditions are appropriate for our cell, and the large values of μ and D_0 for H↓ cause boundary effects to be important farther from the boundary than in ^{3}He.

We conjecture that the observed echo behavior might be explained by including appropriate boundary terms in eq. 3 in ref. (5).

For example, consider spins that have been tipped into the x--y plane, and subjected to a gradient $G_{xz} = \partial(\gamma H_z)/\partial x$, that is, H_0 is parallel to z and its z component varies with x. The dephased spins will have a gradient $\vec{\nabla}\underset{\sim}{s}$ in the x--y plane and perpendicular to $\underset{\sim}{s}$. Equation 3 of reference (5) predicts a spin current that carries z magnetization in the x direction. Except at the walls, this current is uniform and divergence free, contributing nothing to the equation of motion of $\underset{\sim}{s}(\vec{r}, t)$. For reflecting boundary conditions, though, there will be an accumulation of s_z at the +x boundary and an accumulation of $-s_z$ at the -x boundary. Applying a π pulse rotates the triad $(\underset{\sim}{s}, \vec{\nabla}\underset{\sim}{s}, \vec{J})$ by π, reversing the direction of the current, but also reversing the sign of the accumulated s_z. Therefore the accumulation continues to grow while the echo is forming.

This process causes the magnetization to rotate out of the x--y plane. This reduces the size of the received NMR signal, but does not necessarily destroy the magnetization. Estimates of the size of this effect indicate that it could be significant in our experimental situation. It produces a very inhomogeneous distribution of spin density, producing a very inhomogeneous molecular field that can affect the equation of motion of the spins in very complicated ways.

It is clear that much more work needs to be done before the spin dynamics of H↓ are fully understood. We are continuing our investigations.

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