Observation of Nuclear Spin Waves in Spin-Polarized Atomic Hydrogen Gas

B. R. Johnson, J. S. Denker, N. Bigelow, L. P. Lévy, J. H. Freed, and D. M. Lee
Laboratory of Atomic and Solid State Physics and Baker Laboratory of Chemistry,
Cornell University, Ithaca, New York 14853
(Received 24 January 1984)

We have observed narrow, distinct resonances in the NMR spectrum of dilute spin-polarized atomic hydrogen gas (\(n \sim 10^{16}\) atoms/cm\(^3\)). The dependence of the observed spectra on temperature, density, polarization, and magnetic field gradient is consistent with theoretical predictions for spin-wave excitations damped by diffusion. We have measured the parameter \(\mu\), which is a measure of the importance of exchange effects in spin-transport processes, and the diffusion coefficient \(D_0\), both of which are in reasonable agreement with theory.

PACS numbers: 67.40.-w, 67.70.+n

We report the observation of a series of sharp, discrete resonance lines in the NMR spectrum of spin-polarized atomic hydrogen (H\(_\uparrow\)) (see Fig. 1). We interpret the observed spectra in terms of collective nuclear spin-wave oscillations in this rarefied gas.

Quantum mechanical exchange in degenerate systems (e.g., liquid \(^3\)He and electrons in metals\(^1,2\)) greatly alters the spin-transport properties and in particular causes these systems to exhibit spin-wave oscillations. Lhuillier and Laloë\(^3,4\) have shown, however, that in a nuclear spin-polarized gas, even when the interactions are spin independent, exchange effects have a significant influence on the nuclear spin-transport properties when the thermal de Broglie wavelength \(\lambda = \hbar (2\pi/mkT)^{1/2}\) becomes significantly larger than the atomic size \(d\). Because of its low atomic mass and negligible attractive interaction, H\(_\uparrow\) is expected\(^5\) to remain a gas even at \(T = 0\) K. Hence the condition \(\lambda \gg d\) can easily be satisfied long before degeneracy effects become important.

When two atoms with different nuclear spin orientations pass within one de Broglie wavelength of each other, exchange effects cause their spins to precess about one another. In the following letter,\(^6\) Lévy and Ruckenstein discuss spin transport in H\(_\uparrow\) in terms of a quasiparticle description similar to Landau Fermi-liquid theory. They show that at the macroscopic level, exchange effects give rise to a precession of the spin current about a molecular field parallel to the nuclear spin density \(\sigma\). This produces the magnetization oscillations discussed by Lhuillier and Laloë.

![FIG. 1. H\(_\uparrow\) NMR spectra showing the variation of the spectrum with the field gradient. The magnitude of the vectorial change in \(\nabla \delta H_0(\mathbf{r})\) was about 0.4 G/cm between traces (a) and (b), and twice that much between (b) and (c). The initial magnitude and direction of \(\nabla \delta H_0\) were not accurately known. The time between successive spectra is less than 2 min. All spectra were taken at \(T = 245\) mK, \(n = 3.2 \times 10^{16}\) atoms/cm\(^3\), and \(P \sim -1\) using tipping pulses of about 10°.](image-url)
For our experimental conditions of $H_0 = 7.7$ T and $T < 0.8$ K, only the lowest two hyperfine states of $H_1$, $|a\rangle = |\downarrow\rangle - \eta |\uparrow\rangle$ and $|b\rangle = |\downarrow\rangle + \eta |\uparrow\rangle$, are thermally populated. Here $\downarrow$ denotes electron spin and $\uparrow$ denotes proton spin. The admixture $\eta = 3 \times 10^{-3}$ promotes recombination of $|a\rangle$-state atoms into molecular hydrogen, creating a large negative nuclear spin polarization, which in turn creates a large molecular field. The experiment probes the presence of a molecular field by disturbing the atoms from equilibrium with cw or pulsed NMR at the $|a\rangle - |b\rangle$ transition frequency.

The equation of motion for $\sigma_+$, which is in general nonlinear, can be linearized when $\sigma_z$ is uniform and the component $\sigma_+ (\vec{T}, t) = \sigma_z + i \sigma_y$ perpendicular to $H_0 = H_0^\perp$ is small compared to $\sigma_z$. In particular, this condition holds when the NMR tipping angle is small. The resulting equation of motion, in the rotating frame, is

$$i \frac{\partial \sigma_+}{\partial t} = \gamma \delta H_0 \sigma_+ + i D_0 \frac{1 - i \epsilon \mu P}{i + \mu^2 p^2} \nabla^2 \sigma_+,$$

where $D_0$ is the longitudinal spin diffusion coefficient (see below), $\delta H_0 (T)$ is the deviation of $H_0$ from its average, and $\epsilon$ is +1 for bosons or -1 for fermions. The dimensionless parameter $\mu$ is of order $\lambda/d$, and measures the ratio of the spin-current precession frequency in the molecular field to the decay rate of the spin current. We define the polarization $P_+ = (n_a - n_b)/(n_a + n_b) = \sigma_y/n$, where $n_a$ and $n_b$ are the number densities in the $|a\rangle$ and $|b\rangle$ states. The $\gamma$ used in Eq. (1) is nearly equal to the gyromagnetic ratio of the proton.

This equation, with appropriate boundary conditions, predicts discrete resonances in the NMR spectrum at frequencies corresponding to standing spin-wave modes.

The cryostat is similar to the one used in earlier work. The walls of the sample cell are covered with a saturated film of pure $^3$He. The 1-GHz NMR resonator is now a split-ring resonator made of copper which is heat sunk to the main body of the sample cell (see Fig. 2). A combination of boron nitride and silicone vacuum grease fills most of the region just outside the resonator, plugging the capacitor gap and the ends of the resonator. This ensures that most of the NMR signal comes from the region inside the resonator, where the rf magnetic field $H_1$ is uniform. One can show that when $H_1$ is uniform, a nonuniform $H_0$ is necessary to allow the spin-wave modes to couple to the NMR resonator. The sample region inside the resonator is nearly cylindrical, with diameter 0.63 cm and length 1.04 cm. $H_1$ can travel between the resonator and the much larger outer reservoir of gas only via a hole of diameter 0.081 cm and length 0.132 cm in the copper wall of the resonator. The resonator center frequency is 1042.4 MHz, its loaded $Q$ is 650, and it is overcoupled to minimize the effect of radiation damping on the NMR spectrum.

Typically, the spins are tipped by a single 20-mW pulse 10 $\mu$s long, giving a tipping angle of about 10°. The free-induction decay (FID) was normally digitized every 3 $\mu$s for 6 ms, and the Fourier transform $X$ was computed. Some cw derivative spectra were also taken, giving results similar to the derivatives of the Fourier-transform spectra. The pulse method was preferred because of uncertainties about the changes in polarization and density during the time required to sweep out a complete cw spectrum.

For tipping pulses of $< 20°$, a typical spectrum has the appearance of a broad resonance (of order 10 kHz wide) with several narrow lines superimposed on it. Usually the first line at the edge of the spectrum is the most prominent one, and is about 200 Hz wide; lines narrower than 100 Hz have been seen. Subsequent lines become successively broader. These lines are prominent features of all spectra except under some extreme conditions of low magnetization or high temperature (about 700 mK). Most spectra were taken with $P$ negative, at densities $n$ between $3 \times 10^{15}$ and $5 \times 10^{16}$ atoms/cm$^3$ and temperatures $T$ between 160 and 700 mK.

At any given value of $n$, $T$, $\delta H_0$, and $P$, the spectrum is quite reproducible. Lowering the temperature, which also causes $|P|$ and presumably $|\mu|$ to
increase, makes the lines narrower, more numerous, and more intense. The same trend can be observed by allowing $|P|$ to increase while holding $T$ constant. As the density is reduced, the lines become somewhat broader and more widely separated. In certain cases, the appearance of the spectrum changes markedly with small changes in $\nabla \delta H_0$ (see Fig. 1).

The total width and center frequency of the spectrum are nearly independent of $n$, $T$, and $P$, but the width has been varied from 3 to 15 kHz by changing the field $\delta H_0$ with the shim coils of our magnet. The variation of the total width of the spectrum with $\delta H_0$ is as expected for inhomogeneous broadening. Given that $H_1$ is uniform, the weak variation of the overall width with $n$, $T$, and $P$ is consistent with localization of the spin waves by $\delta H_0$, as predicted by Lévy and Ruckenstien.\textsuperscript{6}

We attribute the sharp lines in the spectra to damped spin waves in the sample region inside the resonator. If we draw an analogy between Eq. (1) and the Schrödinger equation, this means that the applied “potential” due to $\gamma \delta H_0$ is large compared to the spin-wave “kinetic energy” resulting from the term containing $\nabla^2 \sigma_+$. The sign of $\epsilon \mu P$ determines whether the spin-wave modes will be confined in the region of most positive or most negative $\delta H_0$. The narrowest lines (which have the smallest “kinetic energy”) should appear on the high-frequency side of the spectrum when $\epsilon \mu P$ is positive, whereas they should appear on the low-frequency side when $\epsilon \mu P$ is negative.

When $H_1$, atoms are loaded into the cell, $P$ is initially positive, although it soon inverts because of preferential recombination on $|a\rangle$ atoms. We can restore positive $P$ with a tipping pulse of $\sim 180^\circ$ and then probe the system with a small pulse. The change of sign of $P$ can be verified by observing the sign of the initial voltage of the FID. In either case, spectra obtained under the condition of positive polarization have the narrowest lines flipped to the low-frequency side of the spectra; the edge frequencies of the spectra are not shifted (see Fig. 3). This confirms the applicability of Eq. (1) and the confinement of the spin waves by $\delta H_0$. We find $\epsilon \mu < 0$, as expected for an atomic $H_1$ gas of bosons interacting via a net repulsive potential.

For large tipping angle $\theta$, the spectrum is markedly different from the small-$\theta$ spectrum. This is probably due to the expected nonlinear spin response at large $\theta$.

To measure $D_0$, the longitudinal spin diffusion was measured for temperatures between 394 and 520 mK and densities between $1 \times 10^{16}$ and $9 \times 10^{16}$ atoms/cm$^3$. To measure $D_0$, the magnetization was destroyed by a pulse of tipping angle $\theta \sim 90^\circ$ and the subsequent recovery of $\sigma_+$ was probed by a second $\theta$ pulse a time $t_0$ later. The ratio of the FID initial amplitudes was analyzed as a function of $t_0$, yielding a recovery rate $1/\tau$ of about $10^{-3}$ s$^{-1}$ at a density of $1 \times 10^{16}$ at 400 mK. The time $\tau$ is long compared to the spin-wave lifetimes, but short compared to $T_1$ and to recombination times,\textsuperscript{10,11} and so $\tau$ is entirely determined by diffusion of spins through the small hole in the resonator. The diffusion data were fitted by analytic solutions\textsuperscript{13} of the diffusion equation $\partial \sigma_+/\partial t = D_0 \nabla^2 \sigma_+$ for our geometry. Our results vary from $nD_0 = (1.7 \pm 0.3) \times 10^{18}$ cm$^{-1}$ s$^{-1}$ for $n$ between $5 \times 10^{16}$ and $9 \times 10^{16}$ cm$^{-3}$, to $nD_0 = (1.3 \pm 0.3) \times 10^{18}$ cm$^{-1}$ s$^{-1}$ for $n$ between $1 \times 10^{16}$ and $3 \times 10^{16}$ cm$^{-3}$, this may be compared to the calculation of Lhuillier,\textsuperscript{4} $nD_0 = 1.5 \times 10^{18}$ cm$^{-1}$ s$^{-1}$ and to the hard-sphere\textsuperscript{4} value of $nD_0 = 0.5 \times 10^{18}$ cm$^{-1}$ s$^{-1}$. There was no discernible dependence of $D_0$ on temperature over the temperature range of these data, as expected.\textsuperscript{4}

Some typical spectra were fitted by model spectra computed from Eq. (1) for a cylindrical region, with use of given values of $D_0$, $\mu P$, and the magnetic field gradient components $G_a$ and $G_\phi$, parallel and perpendicular, respectively, to the axis of the resonator. Reflecting boundary conditions were used, i.e., no current of $\sigma_+$ normal to the wall.\textsuperscript{6} The actual field profile $\delta H_0$ was only approximately
known, and so $G_{S}$ and $G_{S}$ were chosen to optimize the fit. The chosen value of $D_0$ was close to the independently determined value described above.

Qualitative agreement between the model spectra and the data was obtained. Good quantitative agreement was not always obtained. In particular, the linewidths and positions agreed better than the intensities, which tended to be greater in the experimental spectra. This may have been due to nonuniform $H_{1}$, nonuniform $\nabla \delta H_{0}$, nonideal shape of the sample region, or nonzero spin current normal to the walls (conceivably due to $T_{J}$ processes).

The model spectra, and Eq. (1) itself, indicate that the spin-wave linewidths are much more sensitive to $\mu P$ than to all other parameters. Fitting to the linewidths determined $\mu P$ within 20%. For spectra at 227 and 245 mK, we find $\mu P = 6 \pm 1$. We estimate $P = -0.8 \pm 0.15$ for these spectra, and so $\mu = 7.5 \pm 2$, in agreement with calculations of $\mu$ by Lhuillier and Lévy and Ruckenstein.\(^6\)

In conclusion, exchange effects in a dilute $H_{1}$ gas cause damped spin waves to be a prominent aspect of the spin transport properties. The main features of the observed phenomena are in good agreement with the theory.

We thank L. S. Goldner, K. Feldmann, K. Earle, and D. Thompson for valuable assistance, and A. E. Ruckenstein for illuminating discussions. One of us (L.P.L.) wishes to thank General Motors Corporation for fellowship support during the course of this work. This work was supported by National Science Foundation Grant No. DMR-8305284.

---


\(^{9}\)This is the negative of the definition used in Ref. 7, but is the same as the $M_{s}$ used in Refs. 3 and 4.


