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

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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 μ, 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.

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We report the observation of a series of sharp, discrete resonance lines in the NMR spectrum of spin-polarized atomic hydrogen (H↓) (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↓ is expected[5] to remain a gas even at $T = 0$ K. Hence the condition $\lambda >> d$ 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↓ 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 $\vec{\sigma}$. This produces the magnetization oscillations discussed by Lhuillier and Laloë.

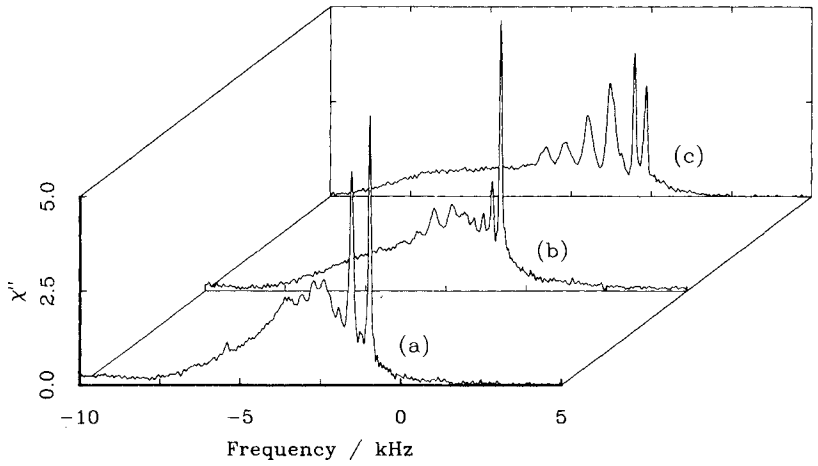

FIG. 1. H↓ NMR spectra showing the variation of the spectrum with the field gradient. The magnitude of the vectorial change in $\nabla\delta H_0(\vec{r})$ was about 0.4 G/cm between traces (*a*) and (*b*), and twice that much between (*b*) and (*c*). The initial magnitude and direction of $\nabla\delta H_0$ were not accurately known. The time between successive spectra is less than 2 min. All spectra were taken at $T = 245$ mK, $n = 3.2\times10^{16}$ atoms/cm^3, and $P \sim -1$ using tipping pulses of about 10°.

For our experimental conditions of $H_0 = 7.7$ T and $T < 0.8$ K, only the lowest two hyperfine states of H$\downarrow$, $|a\rangle = |\downarrow\ddagger\rangle - \eta|\uparrow\ddagger\rangle$ and $|b\rangle = |\downarrow\ddagger\rangle$, are thermally populated. Here $\downarrow$ denotes electron spin and $\ddagger$ denotes proton spin. The admixture $\eta \sim 3\times10^{-3}$ promotes recombination of $|a\rangle$-state atoms into molecular hydrogen,[7,8] 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 σ_+, which is in general nonlinear, can be linearized when σ_z is uniform and the component $\sigma_+(\vec{r},t) = \sigma_x + i\sigma_y$ perpendicular to $\vec{H}_0 = H_0\hat{z}$ is small compared to σ_z. In particular, this condition holds when the NMR tipping angle is small. The resulting equation of motion, in the rotating frame, is

$$i\frac{\partial\sigma_+}{\partial t} = \gamma\delta H_0\sigma_+ + iD_0\frac{1 - i\epsilon\mu P}{i + \mu^2 p^2}\nabla^2\sigma_+, \qquad (1)$$

where D_0 is the longitudinal spin diffusion coefficient (see below), $\delta H_0(\vec{r})$ is the deviation of H_0 from its average, and ϵ is $+1$ for bosons or -1 for fermions. The dimensionless parameter μ is of order λ/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[9] the polarization $P = (n_a - n_b)/(n_a + n_b) = \sigma_z/n$, where n_a and n_b are the number densities in the $|a\rangle$ and $|b\rangle$ states. The γ 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.[10] The walls of the sample cell are covered with a saturated film of pure ^{4}He. The 1-GHz NMR resonator is now a split-ring resonator[11] 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$\downarrow$ 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.

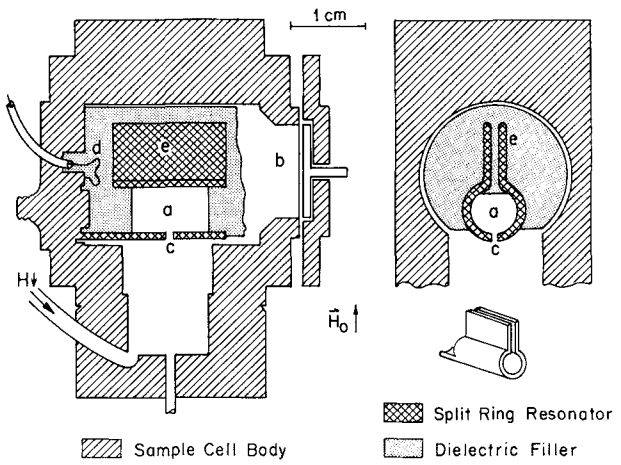

FIG. 2. Left and upper right: Two cross sections through the center of the sample region a; b is the strain gauge, c is the entrance hole, d is the coupling loop, and e is the capacitor. Lower right: perspective view of the resonator, on a smaller scale.

Typically, the spins are tipped by a single 20-mW pulse 10 μs long, giving a tipping angle of about 10°. The free-induction decay (FID) was normally digitized every 3 μs for 6 ms, and the Fourier transform χ 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 3×10^{15} and 5×10^{16} atoms/cm^3 and temperatures T between 160 and 700 mK.

At any given value of n, T, δH_0, and P, the spectrum is quite reproducible. Lowering the temperature, which also causes $|P|$ and presumably[4] $|\mu|$ 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 spetrum 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 δH_0 with the shim coils of our magnet. The variation of the total width of the spectrum with δH_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 δH_0, as predicted by Lévy and Ruckenstein.[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 δH_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$\downarrow$ atoms are loaded into the cell, P is initially positive, although it soon inverts because of preferential recombination $|a\rangle$ atoms. We can restore positive P with a tipping pulse of $\sim 180°$ 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 δH_0. We find $\epsilon\mu < 0$, as expected for an atomic H$\downarrow$ gas of bosons interacting via a net repulsive potential.

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

To measure D_0, the longitudinal spin diffusion was measured for temperatures between 394 and 520 mK and densities between 1×10^{16} and 9×10^{16} atoms/cm^3. To measure D_0, the magnetization was destroyed by a pulse of tipping angle $\theta\sim90°$ and the subsequent recovery of σ_z was probed by a second θ 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 s^{-1} at a density of 1×10^{16} at 400 mK. The time τ is long compared to the spin-wave lifetimes, but short compared to T_1 and to recombination times,[10,12] and so τ is entirely determined by diffusion of spins through the small hole in the resonator. The diffusion data were fitted by analytic solutions[13] of the diffusion equation $\partial\sigma_z/\partial t = D_0\nabla^2\sigma_z$ for our geometry. Our results vary from $nD_0 = (1.7\pm0.3)\times10^{18}$ cm^{-1} s^{-1} for n between 5×10^{16} and 9×10^{16} cm^{-3}, to $nD_0 = (1.3\pm0.3)\times10^{18}$ cm^{-1} s^{-1} for n between 1×10^{16} and 3×10^{16} cm^{-3}; this may be compared to the calculation of Lhuillier,[4] $nD_0 = 1.5\times10^{18}$ cm^{-1} s^{-1} and to the hard-sphere[5] value of $nD_0 = 0.5\times10^{18}$ cm^{-1} s^{-1}. There was no discernible dependence of D_0 on temperature over the temperature range of these data, as expected.[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, μP, and the magnetic field gradient components G_a and G_r parallel and perpendicular, respectively, to the axis of the resonator. Reflecting boundary conditions were used, i.e., no current of σ_+ normal to the wall.[6] The actual field profile δH_0 was only approximately

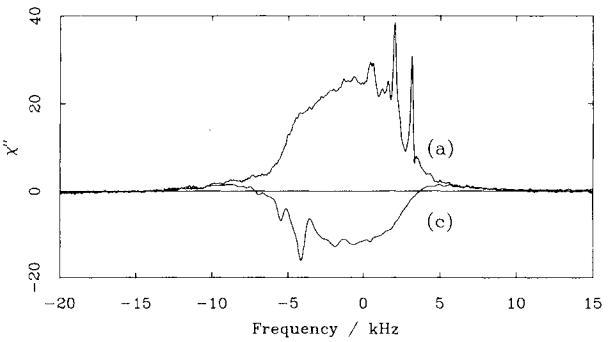

FIG. 3. Polarization reversal, using a three-pulse sequence: 45° (wait 10 ms) 180° (wait 12 ms) 45°. Trace (a) shows the response to pulse No. 1; trace (c) shows the response to pulse No. 3. Polarization is inverted by pulse No. 2, so that the spin-wave lines appear on the low-frequency side of trace (c). The first two pulses incidentally reduce $|P|$, so that the spectrum (c) is smaller and has broader spin-wave lines. The "wings" where trace (c) goes above zero are attributed to regions outside the resonator, where the spins are not inverted by pulse No. 2. The density was $n = 1.5\times10^{16}$ cm^{-3} at a temperature $T = 246$ mK.

known, and so G_a and G_r 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_2 processes).

The model spectra, and Eq. (1) itself, indicate that the spin-wave linewidths are much more sensitive to μP than to all other parameters. Fitting to the linewidths determined μP 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 μ by Lhuillier[4] and Lévy and Ruckenstein.[6]

In conclusion, exchange effects in a dilute H↓ 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.

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