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Studies of Nonlinear Spin Dynamics of Polarized Atomic Hydrogen Using NMR Pulses of Finite Duration in an Applied Magnetic Field Gradient

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We describe the results of experiments on the effects of pulses of finite duration on the spin wave spectra of a sample of spin polarized atomic hydrogen in a static magnetic field gradient and for a range NMR tipping angles. As the NMR pulse width is increased the spectral width of the pulse decreases and the spatial distribution of magnetization following the pulse is affected. Experiments have covered a range of spectral widths for the pulse from much narrower than to much wider than the inhomogeneous spectral width of the sample. Significant changes are observed in the Fourier transform spectra as a function of pulse length and will be compared to computer simulation and theory.

INTRODUCTION

The transport of magnetization in spin polarized atomic hydrogen can be described by a set of highly nonlinear equations of motion which couple all components of magnetization in all spatial directions [1-3]. For the case of small tipping angle NMR experiments the equations can be linearized and successfully describe experimental observations [4] The fully nonlinear equations of motion have been considered analytically by Lévy [5], for the case of no static field gradient, one spatial degree of freedom and in the low temperature high polarization limit. The theoretical predictions include the presence of a family of new nonlinear modes which are characterized by nonuniform magnetization distributions within the sample. However, there has not yet been a definitive connection made between experimental observations and theoretical predictions in the nonlinear regime. In an earlier paper we described the effects of increased tipping angles, and hence increased nonlinearity, on the frequency spectrum [6]. These experiments were performed using minimal static field gradients and highly uniform initial magnetization distributions. In this report we describe some results of experiments which probe the effect of nonuniform initial magnetization distributions on the coupling to nonlinear modes of the system. In particular we examine the changes in the relative phases of the observed modes which are attributed to the nonlinear character of the transport equations.

EXPERIMENT: Technique

Our initial experiments in the linear regime were performed using a uniform rf tipping field in the presence of an applied static field gradient. The pulse lengths were always chosen to be short as compared with dephasing times as well as spin wave lifetimes. Under these conditions, the coupling to spin wave modes is determined by the rf field profile and the modes are initially in phase. However, as the duration of the tipping pulse is allowed to increase, the magnetization immediately after the pulse is no longer uniform across the sample. As the pulse length is increased, the pulse power (and therefore the strength of the tipping field H_1) must be decreased if the tipping angle of the spins which are on resonance is to be held constant. In a static field gradient the Larmor frequency depends on the position in the sample. Consequently, in a frame rotating at the Larmor frequency of the magnetization in the center of the cell, there is a residual static field component in other regions of the sample. As the residual static field increases, the effective field rotates out of the transverse plane. The result is that the effective field about which the magnetization processes during the tipping pulse varies in magnitude and direction across the sample. The spins which are off resonance experience a smaller tipping angle from the longitudinal axis and are rotated in the transverse plane. In the limit of extremely long duration pulses, only the spins whose Larmor frequencies are located within the spectral width of the pulse are affected. The idea in these experiments was to intentionally create a nonuniform initial magnetization distribution in order to couple to some of the nonlinear modes of the system.

The experiments were performed by adjusting the rf tipping frequency to be in resonance with the mean Larmor frequency of the sample in an applied static field gradient. This was done by using short duration (about 10 μsec) small tipping angle pulses (6 degrees) and adjusting the rf frequency for so-called 'zero beat' of the NMR free induction decay. In this manner the tipping frequency is set to the center of the inhomogeneously broadened background lineshape. The static field gradients were typically 1 gauss/cm linear gradients applied along the longitudinal axis of the NMR resonator. The experimental geometry was identical to that used in earlier work [4] and will not be discussed here. Under these conditions we performed a series of isolated NMR tipping pulses of increasing duration and decreasing power, keeping the on resonance tipping angle constant, as described earlier. Sweeps were performed for both small and large tipping angles. By staggering the pulse durations during each sweep, we were able to check for changes in the sample density which arise from NMR induced recombination. Care was taken to allow sufficient time between individual pulses for recovery of polarization of the sample contained within the NMR resonator by diffusion [7]. Sweeps were performed at fixed temperature, constant density and high nuclear polarization ($P_z \approx 1$).

EXPERIMENT: Results

Distinct changes have been observed in the relative phases of the modes in the NMR spectra for a variety of initial conditions. For finite duration pulses (typically 200 μsec) effects have been observed in the spectra from large, intermediate and small tipping angle experiments. For short duration pulses (10 μsec) effects on the phases have only been observed for large tipping angles and are therefore attributed to nonlinear effects.

For the longer duration pulses, the orientation of the transverse magnetization varies across the cell. As a result there is variation in the phase at which the distribution couples to successive linear modes as well creating an initial distribution which can couple to nonlinear modes of the system. In the linear limit, the equations of motion for the transverse magnetization take on the form of a Schroedinger equation with damping [2,4,5] where the role of the potential is played by the gradient in the static field. The linear modes couple to the rf field at their classical turning point in the effective potential. For the lowest order and most spatially localized mode the turning point is far from the center of the cell and hence from the center of the overall inhomogeneously broadened line. The result is that the lower order modes couple to the initial magnetization distribution with a larger relative phase shift than the higher order modes. If the phase shifts between the modes were due exclusively to the initial magnetization distribution, the phase shifts in adjacent modes should change monotonically. However, as shown in Figure 1 this is not the case for many of the experimental spectra. Furthermore, the presence of phase shifts in the modes of the short duration large tipping angle spectra implies that the phase shifts observed in the longer pulse duration spectra cannot be attributed exclusively to the effects of the initial conditions on the linear modes. The conclusion is that the observed effects cannot be ascribed to the linear model.

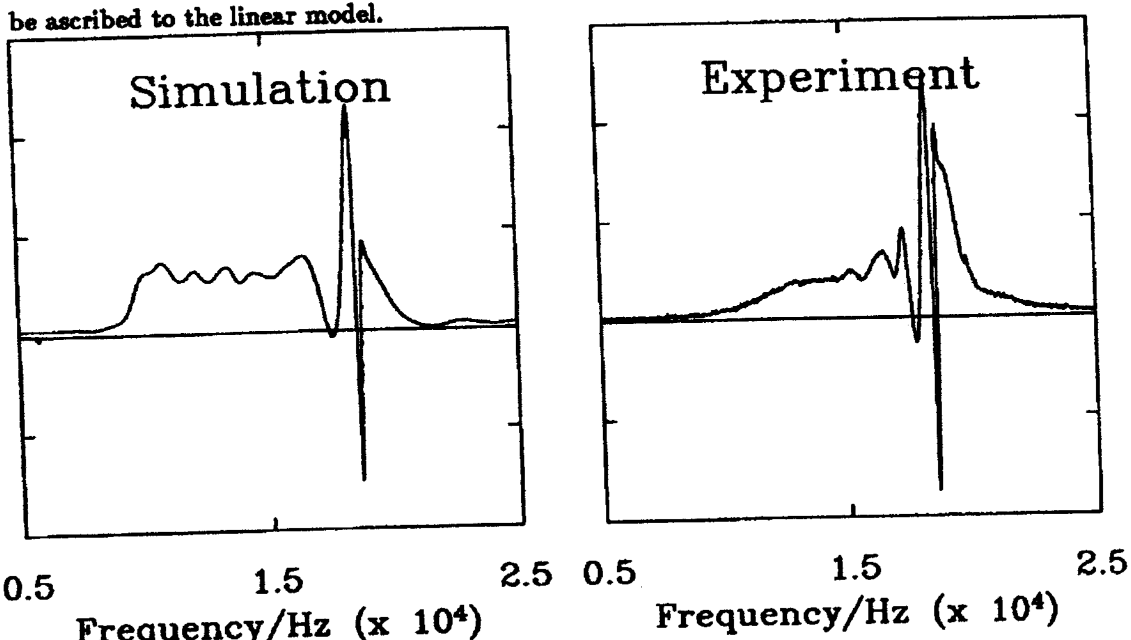

Figure 1. Real part of Fourier spectrum for pulsed NMR experiment in polarized atomic hydrogen. Experiment performed at density $n \approx 10^{16} cm^{-3}$, 340 mK, 7.6 Tesla and 1 gauss/cm linear gradient using a 200 μsec pulse to produce a 45° tipping angle at the center of the sample. Computer simulation based on experimental conditions but assuming uniform initial magnetization.

In order to better understand the observed effects, we have performed computer simulations based on the fully nonlinear equations of motion, as described in reference 6. In Figure 1 we show a comparison between the real parts of an experimental spectrum produced using a 200 μsec pulse and a simulation which assumes a uniform initial magnetization distribution but otherwise identical conditions to the experiment. The spectra are qualitatively very similar, in particular they both exhibit a drastic phase shift between the lowest order spin wave modes and the remaining spectrum. Although such a dramatic relative phase shift was not observed in any short pulse large tipping angle experiments, this may be attributed to a destabilization of the nonlinear mode in three dimensions when the mode is not strongly excited.

CONCLUSION

The observed experimental effects are very suggestive of some of the modes predicted by Lévy, in particular the apparent narrowing and dispersive appearance of the modes with increasing nonlinearity. However a more quantitative analysis is required. As described in reference 8 in this volume we have developed a linear predictive technique for spectral analysis which will help us to disentangle the relative phases and linewidths of the modes. In addition by modeling the experiments with both nonlinear and linearized equations of motion we can further isolate nonlinear effects from linear effects for a more detailed comparison with theory. In future experiments we plan to probe the system with smaller increments in tipping angle which will allow us to examine the details of the coupling to the nonlinear terms in the transport equations. We also plan to investigate pulse shaping techniques which can be used to irradiate selected portions of the frequency spectrum.

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REFERENCES

1) C. Lhuillier and F. Läloe, J. de Physique 43, 197 (1982) and 43, 225 (1982).
2) L. Lévy and A. Ruckenstein, Phys. Rev. Lett. 52, 1512 (1984).
3) E. Bashkin, JETP Lett. 33, 8 (1981).
4) B. R. Johnson, J. S. Denker, N. P. Bigelow, L. P. Lévy, J. H. Freed and D. M. Lee, Phys. Rev. Lett. 52, 1508 (1984).
5) L. P. Lévy, Phys. Rev. B 31, 7077 (1985).
6) N. P. Bigelow, B. W. Statt, J. H. Freed and D. M. Lee, Jap. J. Appl. Phys. 26-3, 233 (1987).
7) B. Yurke, J. S. Denker, B. R. Johnson, N. P. Bigelow, L. P. Lévy, D. M. Lee and J. H. Freed, Phys. Rev. Lett. 50, 1137 (1983).
8) N. P. Bigelow, B. W. Statt, J. H. Freed and D. M. Lee, this vol.