Chapter 1

Introduction

This thesis is concerned with the problem of extending methods for force-detected nuclear magnetic resonance (NMR) methods to the nanoscale regime. Two fundamental problems must be solved for this goal to achieved. First, a sensitive detector of NMR signals is needed. In an NMR experiment, the signal consists of a collection of precessing nuclear magnetic dipoles. Because nuclear dipole magnetic moments are extremely weak, their interaction with a detector in general has a weak effect on the detector, and a detector with optimal coupling to the spins is needed in order to obtain a signal which is not negligible compared to the detector's thermal noise.

A second problem which must be solved is that fluctuations in the sample dipole become more pronounced in comparison with those of the mean dipole as the sample is scaled down. In a system of 10^5 room-temperature hydrogen nuclei placed in a 25 T static field, for example, the uncertainty $\Delta\mu$ is 37 times larger than $\langle\mu\rangle$. The response of a detector which has been designed to interact strongly with the nuclear dipole moment of this sample will be determined by the instantaneous state of the spins, which may be visualized as having an instantaneous dipole moment that fluctuates randomly around a mean value $\langle\mu\rangle$ that is much smaller than the instantaneous moment. For NMR experiments which obtain microscopic information about a sample by measuring $\langle\mu\rangle$, even a noiseless detector would be unable to extract microscopic information in an efficient way by detecting a signal in which the dipole fluctuations are many times larger than $\langle\mu\rangle$.

Both the thermal noise in the detector and the spin fluctuations decrease with

temperature. At the mK temperatures achievable with a dilution refrigerator, for instance, the fluctuations in the dipole moment of a single-proton sample are smaller than the mean dipole $\langle\mu\rangle$ if the proton is exposed to an applied field of a few tesla. However, detection of NMR signals in general depends on the acquisition of many transients, with the spin sample relaxing to a state near thermal equilibrium between transients. At mK temperatures, the spin-lattice interactions which restore the spins to thermal equilibrium between transients become "frozen out," and the time constant T_1 for relaxation to equilibrium increases by orders of magnitude over the room temperature value of T_1. Slow relaxation of the spins to thermal equilibrium translates to a pathologically long delay between transients, which makes acquisition of many transients impractical.

The results in this thesis suggest that a low-temperature mechanical resonator can be used both to induce longitudinal relaxation between transients and to detect the spectrum of samples consisting of a few nuclear spins. The use of a magnetic mechanical resonator as a sensitive detector of magnetic resonance has already been demonstrated in the applications of force microscopy [1] and NMR spectroscopy [2]. In particular, detection of a single electron spin by magnetic resonance force microscopy has been reported [3]. The additional role of a mechanical resonator as a replacement for spin-lattice interactions is portrayed schematically in figure 1.1. Because of the weak coupling between the spins and the cold bath, direct transfer of energy from the spins to the bath is inefficient. However, the magnetic mechanical resonator is coupled strongly both to the spins and to the cold bath, and energy transferred from the spins to the resonator is quickly dissipated into the bath, rather than being cycled back to the spins.

This thesis analyzes resonator-induced spin relaxation and the sensitivity of schemes which use a nanoscale resonator to detect nuclear spins. Chapter 2 derives relaxation equations from first principles and gives a physical interpretation of the processes contributing to relaxation. The intrinsically quantum mechanical nature of the relaxation is highlighted by comparing the quantum mechanical relaxation equations with analogous equations derived using a semiclassical model in which all spin compo-

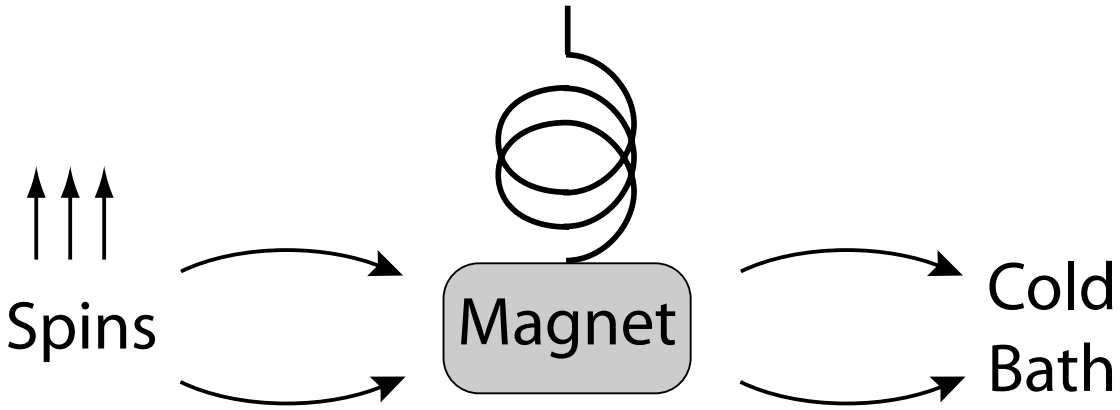

Figure 1.1: Schematic representation of the energy flow during resonator-induced spin polarization.

nents have a definite value simultaneously. In the case where the spins all experience the same field, the semiclassical spins cannot become polarized as a result of their interaction with the resonator, and a quantum mechanical model is necessary even for a qualitative description of the polarization process.

Chapter 3 analyzes resonator-induced relaxation. When multiple spins interact with the same resonator, the spins will not in general relax to a thermal state characterized by a spin temperature. Even in the case where spins are not directly coupled to each other, an indirect spin-spin interaction is present when all spins are coupled to the same resonator, since the resonator's field at a given spin is determined by the interactions which have occurred between the resonator and the other spins. We present a physical interpretation of the mechanism by which an indirect spin-spin torque develops during resonator-induced relaxation, and we estimate the magnitude of this torque and the time $T_{\rm corr}$ required for it to induce strong spin-spin correlations. A perturbation in the spin Hamiltonian which periodically reverses the direction of the indirect torques within a period $T_{\rm corr}$ will prevent the development of resonator-induced correlations and allow the spins to relax to a thermal state.

The longitudinal and transverse relaxation associated with different forms of the spin Hamiltonian H_s is characterized, and the mechanisms by which H_s modifies the relaxation are given a physical interpretation. In the case where the eigenstates of

H_s are weakly perturbed from product states, the system will relax exponentially to thermal equilibrium with the resonator, provided that resonator-induced couplings between populations and zero-quantum "flip-flop" coherences are suppressed by terms in H_s which shift the frequencies of these coherences sufficiently far from zero. Analysis of longitudinal relaxation in example systems of three dipole-dipole coupled spins shows that the relaxation occurs in two stages governed by different physical processes and that the three-spin systems do not relax to a thermal state. For substantially larger dipole-dipole coupled system (e.g., $N = 50$), we propose the hypotheses that the secular dipolar Hamiltonian H_D will quickly equalize the population of states which lie in the same eigenspace of I_z. Chapter 6 presents simulations of the longitudinal relaxation predicted by this hypothesis, and these simulations suggests that a single resonator could efficiently relax dipole-dipole coupled systems to a thermal state.

The analysis in chapter 3 suggests that the transverse relaxation induced by the mechanical resonator could occur on a shorter time scale than the longitudinal relaxation. We derive conditions which guarantee that the time constant for transverse relaxation will be $2/R_h$, where $1/R_h$ is the time constant for resonator-induced longitudinal relaxation of a single-spin sample to thermal equilibrium. Under these conditions, transverse relaxation can be interpreted as "lifetime broadening" associated with the shortened lifetime of energy eigenstates due to coupling with the resonator. For a two-spin system, however, we show analytically that "turning on" the dipolar coupling can accelerate resonator-induced transverse relaxation, and we give an interpretation of the mechanism by which this occurs. Chapter 6 presents simulations of four-spin systems which show that perturbations in the spin Hamiltonian can substantially accelerate resonator-induced transverse relaxation, and that this accelerated relaxation can be distinguished from so-called radiation damping. Analysis in chapter 3, as well as a simulation presented in chapter 6, show that spin-locking limits the rate of resonator-induced transverse relaxation. In the case where spin-locking field is large enough to average the terms of the dipolar Hamiltonian as

well as the superoperator responsible for resonator-induced relaxation, we find that

$$T_{1\rho} = 2/R_h. \tag{1.1}$$

The sensitivity of signal detection by the mechanical resonator is analyzed in chapter 4. Since the term "signal-to-noise ratio" is attached to a variety of different measures of sensitivity, we begin by defining the measures which we will use. We propose a general definition of signal-to-noise ratio (SNR) which can be used to compare the sensitivity of methods which measure the amplitude of a signal with the sensitivity of methods which yield a continuous record of a signal. This definition is used to compare the sensitivity of three schemes for detecting the NMR signal of a sample consisting of a few spins: spin-locked detection of a transverse dipole, detection of a freely-precessing dipole, and detection of a correlated product $\langle I_x(t_1) I_x(0)\rangle$. The dependence of SNR and acquisition time on resonator parameters is analyzed. We find that when the time constant for decay of the signal is $2/R_h$, with instrument noise substantially larger than spin noise, the only resonator parameter which appears in the SNR expression for spin-locked detection is ω_h/T_h, where ω_h is the mechanical frequency and T_h is the temperature. This result suggests that SNR for spin-locked detection will be insensitive to details of resonator design.

A torsional mechanical resonator design is presented in chapter 5. We discuss the advantages of using soft magnetic material and eliminating relative motion between the sample and the resonator, as well as the validity of the models used to characterize the resonator. The possibility of using non-metallic magnetic material as the source of the resonator's magnetic field is introduced. A numerical example is presented for which the calculated time constant for longitudinal relaxation of a single-spin sample is

$$1/R_h = 0.77\,\mathrm{s}\,.$$

In chapter 6, resonator-induced spin relaxation is simulated for various sample sizes and spin Hamiltonians, and the simulations are interpreted using the results obtained in chapter 3. In addition, we present simulations of NMR spectra for

samples containing two spins, with instrument noise and spin fluctuations included in the simulations. These simulations suggest the possibility of chemical studies in which force-detected NMR spectroscopy is used with single-spin sensitivity.

The final chapter studies the possibility of using hyperpolarized spins to cool a single mechanical mode. Numerical examples suggest that cooling would be negligible for resonators of size scale $\sim 10\,\mu$m or larger. In the regime characterized by these examples, substantial cooling requires sufficiently strong spin-resonator coupling that neither a mechanical mode nor a spin mode can be distinguished in the spin-resonator system; instead, the modes of the system include equal contributions from the spins and from the mechanical resonator. The spin-resonator correlations responsible for cooling make a significant contribution to the symmetric correlation function of the resonator coordinate, with the result that the noisy "thermal torque" acting on the resonator is increased rather than diminished by the presence of the hyperpolarized spins.