Novel Methods for Force-Detected Nuclear Magnetic Resonance

Thesis by

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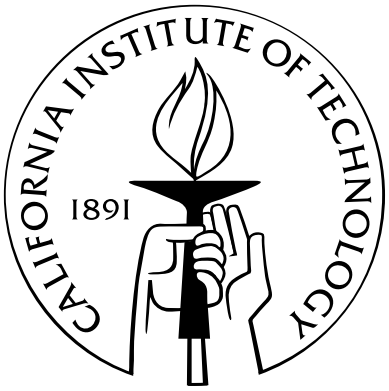

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To my mom

Acknowledgements

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Abstract

This thesis is concerned with the problem of extending methods for force-detected nuclear magnetic resonance (NMR) to the nanoscale regime. A magnetic mechanical resonator can be used both as a sensitive detector of spins and a means of inducing spin relaxation between detected transients. At the mK temperatures achievable in a dilution refrigerator, spin-lattice interactions are "frozen out," and resonator-induced relaxation can replace spin-lattice relaxation in returning the spins to equilibrium between detected transients. We analyze resonator-induced spin relaxation and the sensitivity of schemes which use a nanoscale mechanical resonator to detect spins.

Relaxation equations are derived from first principles, and a physical interpretation of the processes contributing to resonator-induced relaxation is given. 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 components 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.

Resonator-induced relaxation of spin systems is complicated by the fact that 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 of the system. This indirect interaction can prevent the spins from relaxing to a thermal state characterized by a spin temperature. 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_{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 time period shorter than T_{corr} will prevent the development of resonator-induced correlations and allow the spins to relax to a thermal state.

The mechanisms by which the spin Hamiltonian H_s modifies resonator-induced relaxation are characterized. 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 certain zero-quantum 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 containing three dipole-dipole coupled spins shows that the relaxation occurs in two stages governed by different physical processes, and 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 will quickly equalize the population of states which lie in the same eigenspace of I_z. Simulations of the longitudinal relaxation predicted by this hypothesis suggest that a single resonator could efficiently relax dipole-dipole coupled systems to a thermal state.

Arguments based on general properties of the master equation suggest that the transverse relaxation induced by the mechanical resonator could occur on a shorter time scale than that of 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 the "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. Simulations of four-spin systems also show that the presence of dipolar couplings can substantially accelerate resonator-induced transverse relaxation, and that this accelerated relaxation can be distinguished from so-called radiation damping. In addition, we find that spin-locking limits the rate of resonator-induced transverse relaxation. In the case where the spin-locking field is large enough to average the dipolar Hamiltonian and the superoperator responsible for resonator-induced relaxation, we have $T_{1\rho} = 2/R_h$.

We propose a general definition of signal-to-noise ratio (SNR) which can be used to compare the sensitivity of methods that measure the amplitude of a signal with the sensitivity of methods that 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 during the detection period is $2/R_h$, with instrument noise substantially larger than spin noise, the only resonator parameter which appears in the SNR expressions is ω_h/T_h, where ω_h is the mechanical frequency and T_h is the temperature. This result suggests, in particular, that SNR for spin-locked detection will be insensitive to details of resonator design.

A torsional mechanical resonator design is presented. 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 the longitudinal relaxation of a single-spin sample is $1/R_h = 0.77\,\mathrm{s}$. Simulations of detected NMR spectra for two-spin samples 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 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.

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