Single Spin Magnetic Resonance Force Microscopy

Volume 1: Technical and Management Proposal

BAA 01-39
Molecular Observation, Spectroscopy and Imaging using Cantilevers (MOSAIC)

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GE Medical Systems - Dr. James Tropp

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1 Summary of Proposal

IBM and its team (involving researchers from Stanford University, University of Washington, University of Michigan and GE Medical Systems) propose a focused effort on extending magnetic resonance force microscopy (MRFM) to single spin sensitivity and angstrom spatial resolution. If successful, the effort could lead to three-dimensional imaging of atomic structure below surfaces in a non-destructive manner. Such a capability would have broad, even revolutionary, applications in fields ranging from protein structure determination to semiconductor device characterization (e.g., three dimensional dopant imaging). The effort is highly interdisciplinary and pushes the state-of-the-art in quantum mechanical measurement, nanomechanical device fabrication, ultrasensitive force detection, magnetic nanostructure fabrication, spin manipulation techniques, signal processing and high frequency mechanical control. Both electron spin and nuclear spin detection techniques will be investigated.

Because of the team's extensive experience in MRFM and large investment in MRFM-related capital equipment, we are well positioned to pursue the work proposed here. IBM pioneered the very first MRFM experiments in 1992 and, together with the University of Washington, has set the pace for progress in the field. In addition, IBM and Stanford University pioneered the field of ultrasensitive force detection. The IBM-Stanford collaboration has resulted in a succession of world records for detecting small (atto- and sub-attonewton) forces. Since IBM has had an active experimental program pursuing single spin detection for the past five years, it is initimately familiar with the many complex experimental and theoretical issues.

Much of the infrastructure required for the proposed work is already in place, including the capability to fabricate ultrasensitive single-crystal silicon cantilevers, extensive instrumentation for attonewton force detection, rf and microwave equipment for stimulating magnetic resonance and a dilution refrigerator specifically designed for MRFM experiments at millikelvin temperatures.

1.1 Innovative claims and technical approach

The proposed work is divided into five main tasks. Task 1 directly addresses the challenge of detecting individual electron spins, while Tasks 2-5 lay the groundwork for future efforts on single nuclear spin detection and molecular imaging.

Task 1: Single electron spin detection and imaging

We propose to perform experiments aimed at detecting and imaging individual electron spins with nanometer-scale spatial resolution in three dimensions. The experiments will be performed at millikelvin temperature (e.g., $T = 0.1$ K) because previous experiments at higher temperature (4K) taught us that spin relaxation due to thermomagnetic noise from the MRFM tip is a significant issue. The low operating temperature will dramatically reduce spin relaxation effects and significantly lower the cantilever Brownian motion noise. A dilution refrigerator customized for MRFM experiments is already in place at IBM and the first millikelvin MRFM experiments were recently performed successfully using micron-size samples. This experience should greatly accelerate the work proposed here.

The proposed experiments will use ultrasensitive silicon cantilevers previously developed by an IBM-Stanford collaboration. These cantilevers have demonstrated force noise as low as 0.8 attonewton in a 1 Hz bandwidth. Because of their low resonant frequency, in the kilohertz range, the MRFM experiments will use cyclic adiabatic inversion to create a low frequency oscillating spin signal.

Initially, the proposed experiments will use the most idealized samples available (e.g., dangling bonds in SiO_2 known as E' centers), so as to optimize the magnetic resonance behavior of the spins. If the initial single-spin experiments are successful, then extension to other spin systems can be explored. Of particular interest is the imaging of individual dopants in semiconductor devices or detecting electron spin labels attached to biomolecules.

The key work items that will be performed under this portion of the contract include:

- Lithographic fabrication of high anisotropy magnetic tips capable of producing field gradients greater than 2 Gauss per angstrom. This work will supplement our previous effort on using focused ion beam techniques for producing high anisotropy tips.
- Construction of miniature superconducting microwave resonators capable of producing a strong microwave magnetic field at millikelvin temperatures. Since the allowable power dissipation at an operating temperature of 100 mK is only 400 microwatts, the resonator development is a significant challenge and poses some technical risk.
- Experimental investigation of electron spin relaxation near the MRFM tip as a function of tip proximity and temperature. Understanding spin relaxation in the context of MRFM is key to rational design of single-spin experiments.
- The construction of MRFM apparatus optimized for single-spin detection.
- An intensive search for a single-spin signal.
- If a single-spin signal is detected with adequate signal-to-noise ratio, then three-dimensional data will be taken and spin images formed.

The behavior of the electron spins in the presence of the MRFM magnetic tip is probably the most significant unknown factor and thus presents substantial risk. (Note: similar decoherence and relaxation issues are present for all quantum readout devices, not just MRFM.) For the single-spin experiments to be successful, the target spin must be responsive to manipulation by cyclic adiabatic inversion. Hundreds or thousands of inversion cycles are required in order to allow sufficient signal-to-noise ratio to be achieved. Thus, a study of the behavior of spins while undergoing cyclic inversion will be a significant outcome of this work. Since the initial state (up or down) of an individual spin is unknown, signal processing techniques designed to accomodate the unknown, fluctuating signal polarity will be incorporated.

Task 2: Larmor frequency detection of NMR

All previous MRFM experiments have been performed with cantilevers that respond at kilohertz frequencies. Because of their low frequency, these cantilevers cannot be used to directly detect spin precession. Thus previous MRFM experiments have been performed by using some type of spin modulation, such as cyclic adiabatic inversion, in order to create a detectable signal at kilohertz frequency.

By designing and fabricating special high frequency cantilevers, direct (Larmor frequency) detection of nuclear spin precession should be possible. Direct detection has several advantages and is the most promising technique for extension to single-spin NMR. Perhaps the biggest advantage is a practical one: it eliminates the need for an external rf field source. Thus a significant source of heat and disturbance can be eliminated from the system, allowing operation at the lowest possible temperatures with the least disturbance to the measurement. This advantage may prove to be crucial when pursuing the detection of individual nuclear spins. Thus this work can be considered as a prelude to single nuclear spin detection.

We propose to perform experiments aimed at demonstrating MRFM detection of nuclear magnetic resonance at the Larmor frequency. Because of the small magnetic moment of nuclear spins, the initial demonstrations will be performed using spin ensembles, rather than individual spins. A major challenge of the work is to demonstrate that high frequency cantilevers can be fabricated with adequate force sensitivity (in the attonewton range). To achieve the combination of high mechanical resonance frequency and low force noise, the cantilevers must be much smaller than conventional MRFM cantilevers and the detection system must be optimized for the small cantilever size. A second challenge is to demonstrate that a suitably large rf magnetic field can be generated via cantilever oscillation. In order to lock the spins to the cantilever vibration, the vibrating cantilever should produce an oscillating field strength on the order of 10 Gauss.

Key elements of this work include:

- Fabrication of ultrasensitive cantilevers with resonant frequencies in the range of 0.5 - 5 MHz.
- Fabrication of suitable magnetic tips for Larmor-frequency NMR experiments. The field gradient must be sufficient to allow “spin-locking” of the nuclear spins when the cantilever vibrates.
- Development of experimental protocols appropriate for Larmor-frequency NMR experiments
- An intensive search for a Larmor frequency NMR signal

- An experimental study of spin behavior (i.e., relaxation and decoherence) in Larmor-frequency NMR experiments (assuming that a Larmor-frequency NMR signal is detected). Characteristics that are unique to Larmor-frequency MRFM detection will be examined.

Task 3: Pushing the limits of force detection

MRFM has been proposed as a means to determine three-dimensional atomic structures by detecting NMR signals from individual nuclear spins. Making such measurements will require force detection capability well beyond the current state-of-the-art. For example, a hydrogen nucleus (i.e., a proton) in a 20 Gauss per angstrom field gradient will generate a force of only 0.2 attonewtons-rms. This force is 4× smaller than has ever been detected using ultrathin, low frequency cantilevers (in a 1 Hz bandwidth) and at least 50× smaller than can be detected presently using megahertz-frequency cantilevers.

We propose to explore the limits of force detection by developing ultrasmall cross-section, high Q cantilevers and operating them at millikelvin temperautes.. Both low-frequency (5 - 50 kHz) and high-frequency (0.5 - 5 MHz) cantilevers will be studied. Typical cantilevers will have sub-100 nm thickness and widths between 100 and 500 nm. The high frequency cantilevers will be suitable for future single nuclear spin detection experiments, if force noise below 0.2 aN per root hertz can be achieved.

In addition to cantilever fabrication techniques, optimized means of detecting the cantilever vibration signal will be investigated. The goal is for the force noise to be limited only by intrinsic thermal vibration, not detector noise. Achieving this noise limit is extremely challenging due to the small physical size of the cantilevers, the small amplitude of the thermal vibrations at millikelvin temperature and the very low power dissipation required to avoid cantilever heating.

Key work items include:

- Fabrication of ultrasmall cross-section (USC) cantilevers, with special emphasis on designs that maximize the quality factor Q and minimize cantilever stiffness. Techniques to fabricate sub-picogram cantilevers with sub-100 nm thickness and submicron widths will be developed. Techniques for *in situ* cleaning of the cantilever will be incorporated, if necessary.

- Development of advanced detection techniques that are suitable for sensing sub-angstrom motion of high frequency cantilevers at millikelvin temperature. To maintain the low operating temperature, self-heating of the cantilever must be minimized by keeping power dissipation extremely low (in the nanowatt range or less). Two detection techniques are prime candidates: piezoresistive detection and optimized fiberoptic interferometry. In addition, mechanical parametric amplification techniques will be explored as a means to improve the signal-to-noise ratio for sub-attonewton force detection.

- Characterization of force noise near surfaces at millikelvin temperature. Previous experience with low frequency cantilevers at T = 4K has shown that cantilevers operating near a sample surface can suffer from excess force noise induced by electric field fluctuations emanating from the sample surface. Characterization of this surface-induced force noise at millikelvin temperatures is a key step in assessing the realizability of single spin NMR detection.

Task 4: Optimal control of high frequency cantilevers

Larmor-frequency MRFM experiments require megahertz-frequency cantilevers that pose special challenges for monitoring and control: their high frequencies will require fast digital signal processing (DSP) hardware, their increased sensitivity will mean that data will be acquired at faster rates, and the exploration of new temperature and imaging regimes will require advanced algorithms for continuously and accurately monitoring the status of the experiment.

Recent advances in commercial off-the-shelf (COTS) hardware and software, coupled with novel processing algorithms recently developed at the University of Washington (UW), will allow the above MRFM challenges to be met in a timely and efficient manner.

The key work items that will be performed under the contract include:

- *Acquisition and validation of an advanced DSP processing system*
  A "downconversion-upconversion" design will be implemented using high speed DSP technology. The proposed design approach has been verifed to work well by direct "breadboard" experiments performed at UW. The DSP implementation will be subcontracted by UW to a DSP design house (DSPcon Inc.).

- *Cantilever emulation*
  The first application of the DSP system will be to emulate Larmor cantilevers; this allows software development to proceed without waiting upon cantilever fabrication.

- *Optimal Larmor control*
  Control in MRFM is governed by a simple rule: a device that does a good job of emulating a cantilever can also be made to serve as a good feedback controller. By closing the loop between a controller and an emulator, the entire control system can be exercised and debugged over a broad range of operating conditions.

- *Larmor RF synthesis*
  As noted for Task 2, a Larmor-frequency cantilever allows spins to be manipulated by controlling the cantilever motion, rather than by applying (undesirable heat producing) external fields. Thus, the software will be required to apply control forces such that specified "spin-flip" B-field sequences are generated by the tip motion.

- *Reference design integration*
  Under an in-place DARPA/DSO contract called *The Accelerated Development of MRFM*, Profs. Sidles and Garbini are developing a reference design and performance analysis (RDPA) for a molecular observation technology based on Larmor cantilevers. To further accelerate this effort, the UW group will emulate the RDPA

> reference cantilever in one DSP, control it with the RDPA reference controller design, as embedded in a second DSP, and then inject signals emulating those deriving from single-spin challenge targets. This strategy of emulating the reference design in DSP software will avoid wasteful duplication of effort, and allow software development to begin immediately as part of the in-place UW DARPA/DSO effort.

After proper operation of the controller is established, the work will then focus on the areas of real-time diagnostics, adaptive control, and signal multiplexing. Algorithms and control strategies developed and simulated in the initial phases of the program will then be applied to real cantilevers and integrated into MRFM apparatuses at UW and IBM.

Task 5: Spin dynamics in large field gradients

Achieving the ultimate goal of determining three-dimensional atomic structure in biomolecules will depend on suitable behavior of spins in the large field gradient of the MRFM tip. Because the nuclear spins in a biomolecule have spacings on the order of 1-2 Å, the possibility of spin diffusion via rapid flip-flops between adjacent spins must be considered. In principle, spin diffusion will be suppressed by the large gradient from the MRFM tip. The degree of suppression is not fully understood. We propose to perform quantum mechanical simulations of closely spaced spins in a field gradient to elucidate the spin dynamics in the context of MRFM. This understanding will be crucial for future molecular imaging experiments.

Task 6: Signal and image processing for MRFM

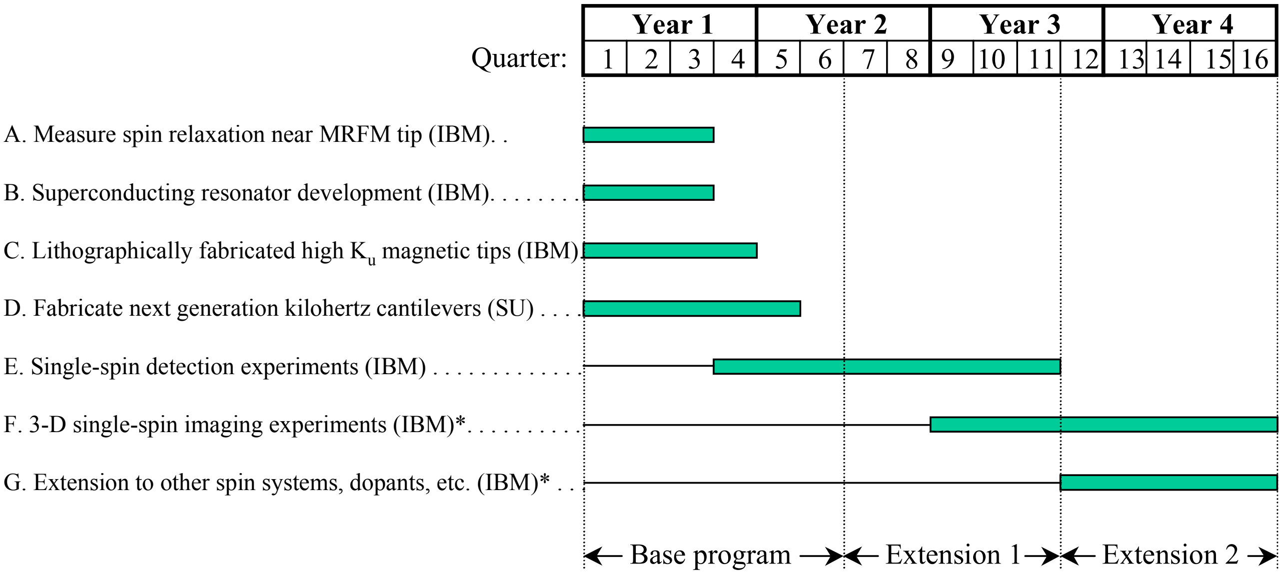
Signals from single-spin MRFM experiments are anticipated to be very small and have statistical characteristics that will require specialized signal processing techniques. The key issue is that the initial quantum state of the spin is unknown. When the detection process collapses the spin wavefunction to an eigenstate of the effective field, the spin will be found in either the spin up or the spin down states with almost equal probability. Thus, there is no *a priori* knowledge of the polarity of the single spin signal. Furthermore, there is substantial likelihood that the polarity of the spin signal will spontaneously flip during the course of the measurement due relaxation effects. Standard signal averaging techniques are ineffective in handling signals of randomly fluctuating polarity. The issue of fluctuating signal polarity becomes even more acute when one considers the challenges of reconstructing multispin images containing fluctuating spins.

Task 6 addresses the signal processing issues for single-spin MRFM. Professors Jeff Fessler and Al Hero at the University of Michigan (UM) will work on signal and image processing problems associated with development of single spin MRFM. Initially, the work will be focussed on development of model-based algorithms for optimal single-electron spin and Larmor NMR detection from cantilever measurements. This will involve mathematical and statistical modeling of the cantilever-based measurement system, developing statistically optimal detection algorithms, and developing lower bounds on the minimum required SNR thresholds.

Issues related to MRFM image reconstruction will also be addressed, both on the macroscale (non-single-spin) and on the microscopic (single-spin) scale. Image reconstruction algorithms for detecting and localizing a small number of superimposed spins will be developed and analyzed.

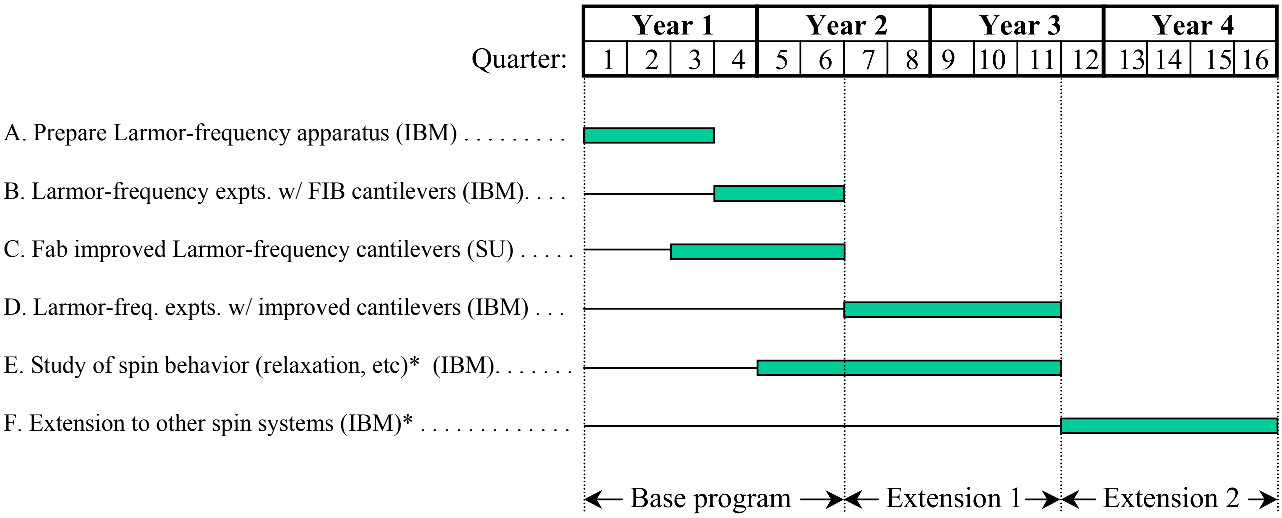
1.2 Cost, schedule and milestones

Task 1 - Single electron spin detection



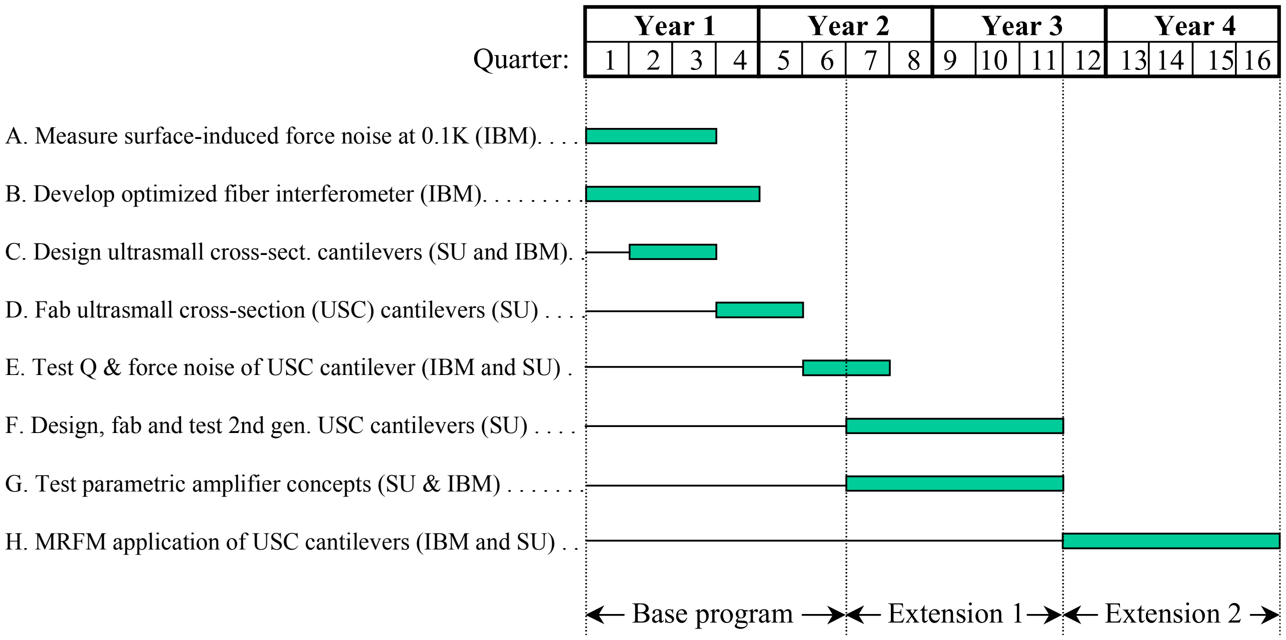
*assuming single-spin detection experiments are successful

Task 2 - Larmor-frequency detection of NMR

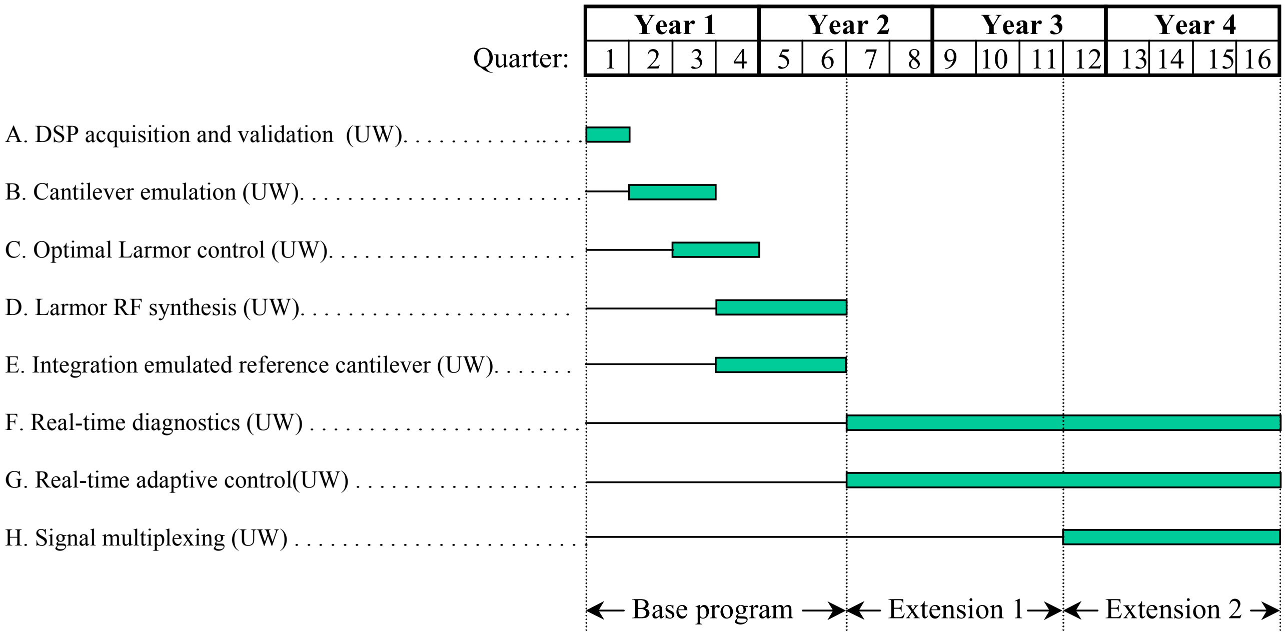


*assuming earlier Larmor-frequency experiments are successful

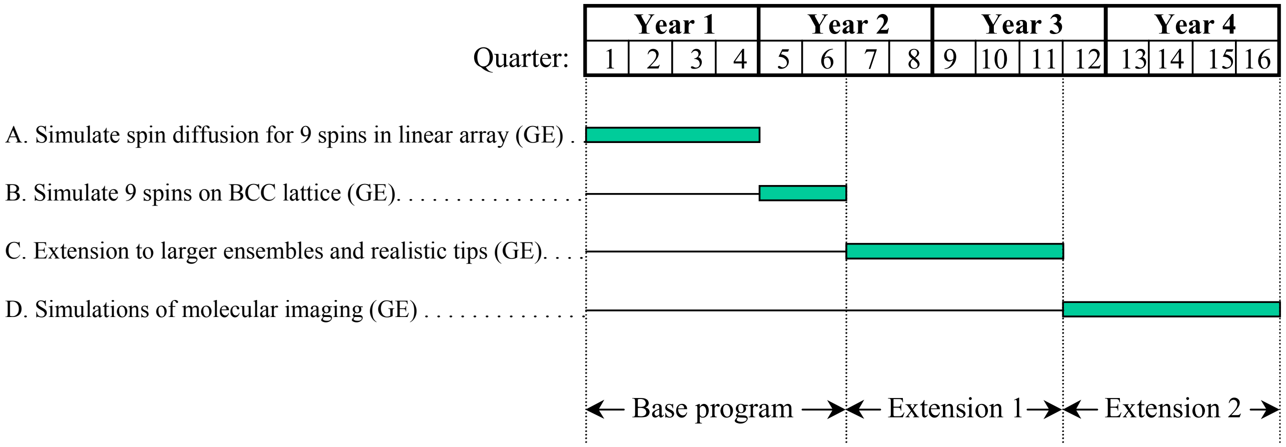
Task 3 - Pushing the Limits of Force Detection



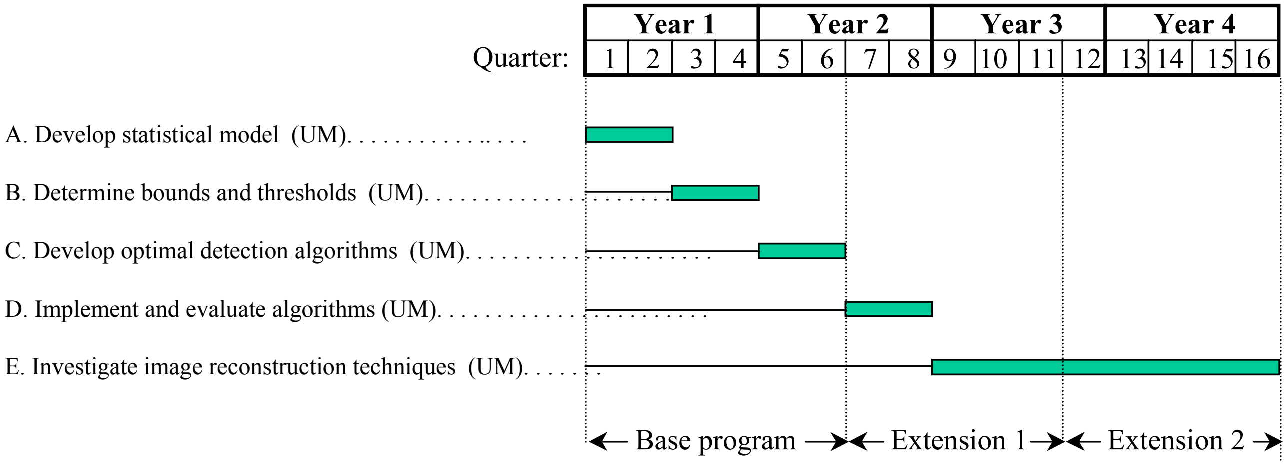
Task 4 - Optimal control of high-frequency cantilevers



Task 5 - Quantum simulations of spins in tip field gradient



Task 6 - Signal and image processing for MRFM



1.3 Organization and Management of the Team

The contract will be administered through the IBM Almaden Research Center with the overall technical effort managed by Dr. Daniel Rugar. Sub-contracts will be let to Stanford University (Prof. Thomas Kenny), University of Washington (Profs. John Sidles and Joseph Garbini), University of Michigan (Profs. Al Hero and Jeff Fessler) and GE Medical Systems (Dr. James Tropp).

An organization chart is shown in Fig. 1 on the next page. This chart specifies general areas of responsibility for the team members. Specific task responsibilities of each team member are denoted by the schedule charts in section 1.2.

This contract proposal is predicated on a strong and close interaction between IBM and its subcontractors. The close geographical proximity of IBM (San Jose, CA) to Stanford University (Stanford, CA) and GE Medical Systems (Fremont, CA) will facilitate close interactions with those institutions. Frequent telephone and e-mail contact, supplemented by regular on-site visitation will allow efficient collaboration with University of Washington (UW) and University of Michigan (UM).

Cantilever design, fabrication and testing will entail especially close collaboration between IBM and Stanford, where the cantilevers will be fabricated. This collaboration is facilitated by the fact that the Rugar group at IBM and the Kenny group at Stanford have worked closely together for the past 7 years on previous projects and already have Joint Study agreements in place. In addition, Daniel Rugar is a Consulting Professor in the Applied Physics department at Stanford University, which further facilitates interactions with Stanford University and its students. It is anticipated that the Stanford students will spend a significant fraction of their time at IBM interacting with IBM personnel.

IBM and the University of Washington also have a long history of fruitful cooperation. In fact, the first experimental demonstration of MRFM in 1992 was the result of a collaboration between Rugar at IBM and Sidles at University of Washington. The two groups have had a number of both formal and informal collaborations since 1992. In this contract, UW will take primary responsibility for high speed controller development.

The collaboration with Dr. Jim Tropp at GE Medical Systems will add an additional perspective on the issue of spin dynamics and other technical issues regarding angstrom-scale magnetic resonance imaging. Theoretical results from GE under the sub-contract will complement theoretical activities at UW being performed under separate contract. Sidles and Tropp will have frequent interactions to compare theoretical conclusions.

The collaboration with Profs. Hero and Fessler at the University of Michigan will add signal and imaging processing expertise to the team.

Figure 1 - Organization Chart

IBM Almaden Research Center

- Administers contract and coordinates overall technical work.
- Focal point of the main MRFM experiments. Develops necessary experimental capability for the low temperature MRFM experiments, including the millikelvin microscope mechanics, force noise measurements, magnetic tips, superconducting resonators, interferometer systems, data collection systems, spin manipulation protocols, etc.
- Extensive experience in MRFM experiments. Complete facility for MRFM already in place, including dilution refrigerator for millikelvin experiments.
- Key personnel:
  - Dr. Daniel Rugar, Manager - oversees the overall effort, with specific responsibilities for MRFM mechanical design, millikelvin experimental techniques and spin manipulation protocols. Approx. 70% effort.
  - Dr. John Mamin, Research Staff Member - involved in all aspects of the MRFM experiments, with specific responsibilities for nanowatt interferometry, superconducting resonators and magnetic tip fabrication. Approx. 70% effort.
  - Mr. Dean Pearson, Technician - technical support, especially vacuum systems and mechanical issues. Approx. 30%
  - Post-doc #1 - single-spin MRFM experiments, including detailed experimental setup, dilution refrigerator operation, data collection and analysis. Approx. 100% effort.
  - Post-doc #2 - high frequency force detection, especially interferometer optimized for detecting ultrasmall cross-section (USC) cantilevers. Assist w/ cantilever fabrication, as necessary. Approx. 100% effort.

Stanford University (SU) - sub-contractor

- Design, fabrication and testing of micromechanical components, including ultrasensitive cantilevers and piezoresistive sensors.
- Extensive experience in micromachining, especially ultrasensitive cantilever and piezoresistive sensors. State-of-the-art micro- and nanofabrication capabilities are available at the Center for Integrated Systems.
- Key personnel:
  - Prof. Thomas Kenny, PI for sub-contract - provides micromachining expertise and supervises graduate student researchers. Approx. 6% effort.
  - Graduate student #1 - design and fabrication of ultrasensitive, high frequency cantilevers.
  - Graduate student #2 - characterization and testing of high frequency cantilevers, including force noise evaluations.

University of Washington (UW) - subcontractor

- Design, construction and testing of high speed digital signal processor for optimal control of high frequency cantilevers.
- Extensive experience in MRFM theory and experiment, especially digital control of MRFM cantilevers.
- Key personnel:
  - Prof. John Sidles, co-PI for sub-contract - provides general expertise for MRFM theory and experiments. Approx. 8% effort.
  - Prof. Joseph Garbini, co-PI for sub-contract - has extensive expertise in digital control. Approx. 8% effort.
  - Graduate student - Digital controller design, construction and programming.

University of Michigan (UM) - subcontractor

- Signal and image processing studies, including algorithm design and evaluation.
- Extensive experience in signal processing, optimal detection methods and image reconstruction.
- Key personnel:
  - Prof. Albert Hero, PI for sub-contract - extensive experience in signal processing and optimal detection techniques. Approx. 16% effort.
  - Prof. Jeff Fessler - extensive experience in image reconstruction, including magnetic resonanceimaging (MRI). Approx. 8% effort.
  - Graduate students (2) - Signal processing design, implementation and evaluation.

GE Medical Systems - sub-contractor

- Quantum mechanical calculations of spin dynamics, including evaluation of spin diffusion in tip field gradient
- Extensive experience in magnetic resonance imaging (MRI) theory and hardware. Familiarity with issues related to atomic structure determination.
- Key personnel:
  - Dr. James Tropp, PI for sub-contract - theoretical study of interacting spins, including quantum mechanical computer simulations of spin-spin interactions. Approx. 10% effort.

Figure 1:

1.4 Deliverables

The deliverable item under this proposed contract will be a Final Technical Report. It shall include a description of the methodology of the experimentation and the results obtained under this proposed contract. No devices, materials or computer programs created as a portion of the work in the proposed contract, or technical data beyond that specifically contained in the Final Technical Report, shall be deliverable items under this proposed contract.

2 Detailed Proposal Information

2.1 Statement of Work in Plain English

We propose a focused effort aimed at extending MRFM toward single spin sensitivity and angstrom-scale spatial resolution. The proposed work can be divided into five main areas:

Task 1: Single electron spin detection and imaging

We propose to perform experiments aimed at detecting and imaging individual electron spins with angstrom-scale spatial resolution in three dimensions. This work involves constructing a complex MRFM experiment and integrating all of the critical elements. These elements include ultrasensitive cantilever force sensors, a sensisitive fiberoptic interferometer system, high field gradient magnetic tips, a superconducting microwave resonator and an appropriate sample containing unpaired electron spins.

Previous experiments at $T = 4$K showed that electron spins are poorly behaved and undergo rapid relaxation when the magnetic tip is positioned near the sample. To overcome this issue, we propose to operate our MRFM experiment at very low temperatures (as low as 100 millikelvin).

Intensive experiments to detect single spin signals will be performed.

There is some risk that, despite our best efforts, the spins may still undergo an unacceptable rate of relaxation and decoherence, preventing successful single spin detection. In this case, experiments will be performed to elucidate the physics of the MRFM measurement process, including tip-spin interaction. Strategies to overcome single-spin measurement issues will be examined and implemented, if possible.

Task 2: Larmor frequency detection of NMR

We propose to perform experiments aimed at directly detecting nuclear spin precession using specially fabricated high frequency cantilevers. Key elements of this work include: 1) Fabrication of ultrasensitive cantilevers with resonant frequencies in the range of 0.5 - 5 MHz, 2) fabrication of suitable magnetic tips for Larmor-frequency NMR experiments, 3) development of experimental protocols appropriate for Larmor-frequency NMR experiments, and 4) an intensive search for a Larmor-frequency NMR signal. Characteristics that are unique to Larmor-frequency MRFM detection will be examined.

Task 3: Pushing the limits of force detection

Ultrasmall cross-section, high Q cantilevers will be developed in order to push the technological limits of force detection. Cantilevers will be fabricated with sub-100 nm thickness and submicron width. The cantilevers will be operated and characterized at temperatures down to 100 mK. Optimized means of detecting the cantilever vibration signal will also be investigated. Force noise will be evaluated for both isolated cantilevers and cantilevers operating near a sample suface.

Task 4: Optimal control of high frequency cantilevers

A high speed digital control system will be developed for optimally controlling the dynamics of high frequency cantilevers. The controller will be adaptive and have built-in diagnostic capability.

Task 5: Spin dynamics in large field gradients

The ability of MRFM to achieve the ultimate goal of determining three-dimensional atomic structure in biomolecules depends critically on the behavior of the spins in the large field gradient of the MRFM tip. Because the nuclear spins in a biomolecule have spacings on the order of 1-2 Å, the possibility of spin diffusion via rapid flip-flops between adjacent spins must be considered. In principal, spin diffusion will be suppressed by the large gradient from the MRFM tip. The degree of suppression is not yet fully understood. We propose to perform quantum mechanical simulations of closely spaced spins in a field gradient to elucidate the spin dynamics in the context of MRFM.

Task 6: Signal and image processing for MRFM

Specialized algorithms for MRFM signal and image processing will be developed and analyzed.

2.2 Technical Rationale and Background Information

2.2.1 MRFM Basics

As pointed out in the MOSAIC workshop, the determination of three-dimensional atomic structure is key to structural molecular biology. Furthermore, as electronic devices shrink to nanometer dimensions, detailed atomic-scale structural information is increasingly critical for many important hardware technologies. Although the field of microscopy has made great progress during recent decades, no presently available imaging technique can directly determine complex three-dimensional atomic structures.

Magnetic resonance force microscopy (MRFM), proposed by Sidles in 1991[1], is a promising approach for three-dimensional atomic structure determination. The technique combines elements of atomic force microscopy (AFM) and magnetic resonance imaging (MRI). The basic elements of MRFM are illustrated in Fig. 2. A sample whose atomic structure is to be determined (e.g., a molecule) is placed below a sharp magnetic tip that is attached to a sensitive micromechanical cantilever. The magnetic tip generates a strong magnetic field $B_t(x, y, z)$ that is spatially inhomogeneous. In the classical picture, the presence of B_t causes the spins in the sample to precess with a Larmor frequency $\omega_L = \gamma B_t(x, y, z)$, where γ is the gyromagnetic ratio.

Because of the spatial inhomogeneity of B_t, spins at different locations in the sample have different Larmor frequencies. An external coil generates a radiofrequency (rf) magnetic field of frequency ω_0, which excites magnetic resonance in those spins located within a thin resonant slice where $\omega_L(x, y, z) = \omega_0$. The thickness of the resonant slice determines the spatial resolution of the technique. For magnetic tips with radii on the order of 500 Å, the gradient of the tip field can exceed 10 Gauss/Å . Since magnetic

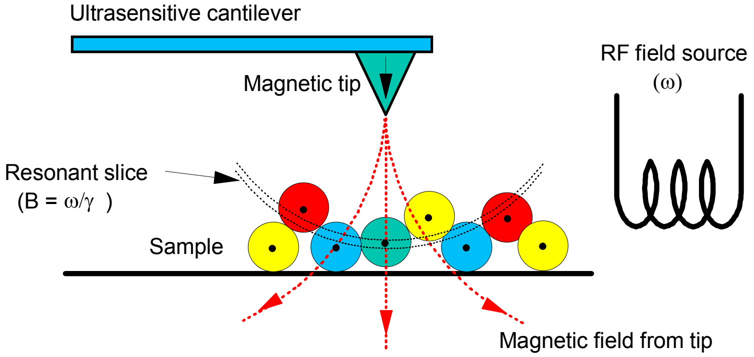


Figure 2: A magnetic resonance force microscope for detecting individual nuclear spins within a molecule.

resonance linewidths ΔB are typically on the order of 1 Gauss, the resonant slice thickness can be substantially less than 1 Å, suggesting that sub-angstrom spatial resolution may be possible. Three-dimensional maps of nuclear spins are formed by scanning the sample in three dimensions and then mathematically transforming the data using image reconstruction techniques[2, 3, 4].

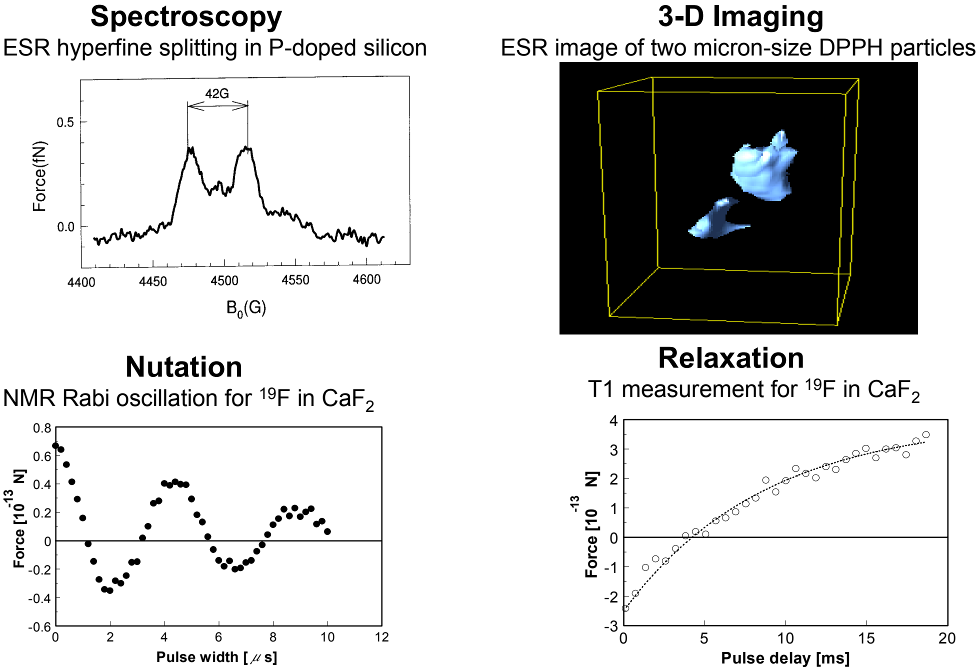
Several means for generating a detectable force signal are possible. For example, frequency modulation of the rf field can induce cyclic adiabatic inversion. The spins in the slice are repeatedly reversed in orientation, creating an oscillating magnetic force on the magnetic tip and causing the cantilever to vibrate. To achieve maximum vibrational response, the frequency of the spin manipulation is chosen to be at the mechanical resonance of the cantilever (typically in the kilohertz range).

In the case of nuclear spins, it may be possible to directly detect the Larmor frequency precession of the spin if high frequency (multi-megahertz) cantilevers can be fabricated with sufficient sensitivity. In this case, the coil is unnecessary since the cantilever vibration can be used to directly excite magnetic resonance of the spins.

Although single-spin detection has not yet been achieved, MRFM has already been very successful for studying spin behavior in micrometer-size samples. Figure 3 shows a sampling of current MRFM capabilities.

2.2.2 Signal-to-noise basics

The paramount objective in designing a single-spin MRFM experiment is achieving adequate signal-to-noise ratio (SNR). For a sinusoidal force signal, the rms signal amplitude is given by $F_{signal} = (1/\sqrt{2})\mu G$, where μ is the spin magnetic moment and G is the field gradient from magnetic tip. For the first demonstration of single-spin detection, we maximize μ by using electron spins rather than nuclear spins, and maximize G by choosing an optimal geometry and material for the magnetic tip. (Magnetic tips will be discussed


Spectroscopy
ESR hyperfine splitting in P-doped silicon

3-D Imaging
ESR image of two micron-size DPPH particles

Nutation
NMR Rabi oscillation for ^{19}F in CaF$_2$

Relaxation
T1 measurement for ^{19}F in CaF$_2$

Figure 3: Examples of various types of MRFM measurements on micron-scale samples. All data taken at IBM.

in more detail in section 2.3.1.) Assuming $G = 2$ Gauss/Å, then the force signal resulting from a single electron spin is $F_{signal} = 1.3 \times 10^{-17}$ N = 13 aN-rms.

Now consider the noise. The force noise for soft, low frequency cantilevers is typically dominated by thermal vibrations (Brownian motion) of the cantilever. The thermal noise is often analyzed by approximating the cantilever dynamics using a simple harmonic oscillator model with a Langevin-type thermal noise driving term $F_{noise}(t)$:

$$m\frac{d^2x}{dt^2} + \Gamma\frac{dx}{dt} + kx = F_{signal}(t) + F_{noise}(t), \tag{1}$$

where x is the displacement at the tip, m is the cantilever effective mass, k is the cantilever spring constant and Γ is the friction coefficient that characterizes the dissipation. For a cantilever with resonance frequency ω_0 and quality factor Q, then $\omega_0^2 = k/m$ and $\Gamma = k/\omega_0 Q$.

In order to maintain thermal equilibrium (i.e., average kinetic plus potential energy equal to k_BT), the spectral density of the force noise S_F necessarily depends on the cantilever dissipation according to[5]

$$S_F = 4\Gamma k_B T. \tag{2}$$

This is the essence of the fluctuation-dissipation theorem as applied to cantilevers. (Note that we use the convention that S_F is a single-sided spectral density.) For detection in a

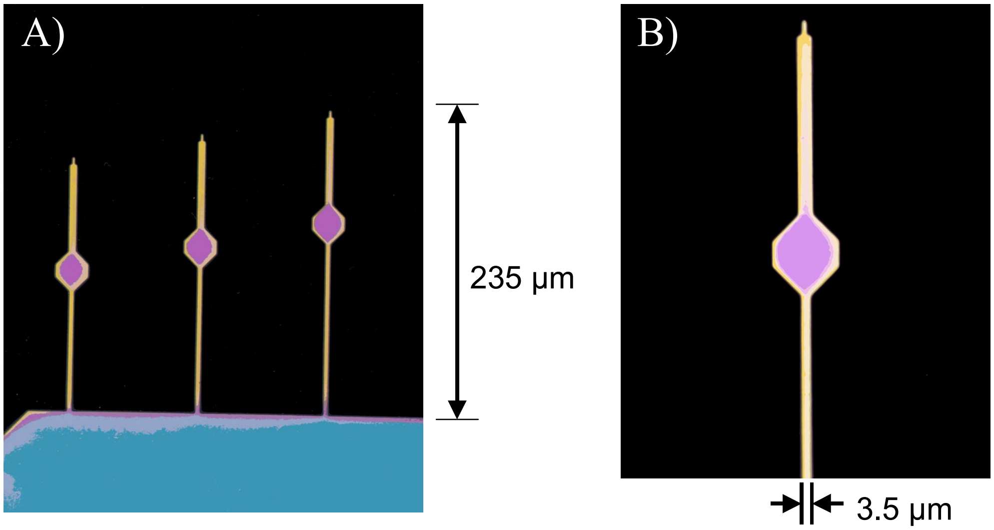


Figure 4: A) Ultrasensitive single-crystal silicon cantilevers for MRFM. These cantilevers are only 65 nm thick and are a million times more flexible than cantilevers used for atomic force microscopy. B) This cantilever exhibited a force noise of 1.3 attonewtons per root hertz at 2.6K. A similar cantilever achieved a force noise of 0.82 aN/Hz$^{1/2}$ at 100 mK. This is the lowest noise ever measured for a mechanical force sensor.

bandwidth $\Delta\nu$, this spectral density gives the minimum detectable force

$$F_{\text{min}} = S_F^{1/2} \Delta\nu^{1/2} = \left(\frac{4kk_B T \Delta\nu}{\omega_0 Q}\right)^{1/2} . \tag{3}$$

From equation (2) it is clear that there are just two ways to reduce the force noise spectral density: lower the temperature or reduce the cantilever dissipation Γ. Focusing for now on the latter, we consider Γ in terms of cantilever geometry and material properties. We first note that for the fundamental flexural mode of a simple rectangular cantilever, $k = 0.257\, E\, w\, t/L^3$ and $\omega = 1.01\ (t/L^2)\,(E/\rho)^{1/2}$, where E is the Young's modulus of the cantilever material, ρ is the density and L, w, and t are the length, width and thickness, respectively[6, 7]. Combining these with (2) and (3), we find

$$\Gamma = 0.254 \left(\frac{wt^2}{L\,Q}\right) (E\rho)^{1/2} . \tag{4}$$

For a given Q, Γ is minimized by making the cantilever narrow, thin and long. For sufficiently thin cantilevers (e.g., $t < 1$ μm), however, Q is found to decrease with decreasing thickness because of contamination and oxidation of the cantilever surfaces[8]. Thus maintaining clean cantilever surfaces is a key requirement for achieving the lowest possible force noise.

Figure 4 shows optical micrographs of IBM-Stanford cantilevers that epitomize our “narrow, thin and long” cantilever strategy [5]. A typical cantilever is 65 nm thick, 235

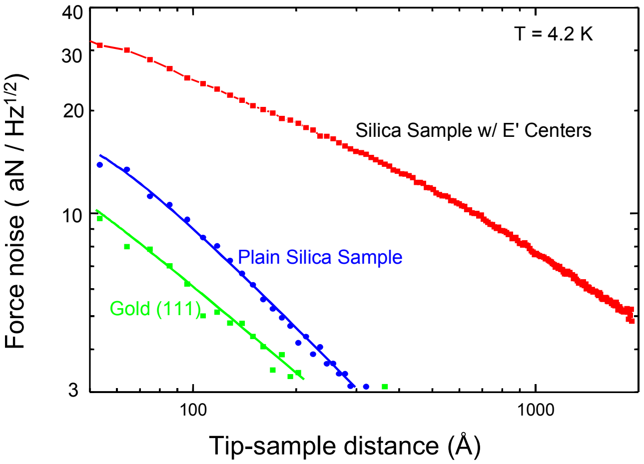


Figure 5: Force noise near surfaces at 4.2K. The silica sample with E' centers generates significantly larger force noise because of the charged nature of the E' center.

μm long and, at the base, is approximately 3.5 μm wide. Because the cantilever is so thin, the resulting spring constant is extraordinarily small, about 3 μN/m. This is roughly a million times "softer" than is typically used for atomic force microscopy. If a macroscopic spring had this spring constant, then a 1 gram weight would result in a 3 kilometer deflection!

The cantilever in Fig. 4B was characterized at temperatures down to 2.6 K. At this temperature, a Q of 4.6×10^4 was measured with a resonance frequency of 950 Hz, yielding a dissipation level of $\Gamma = 1.1 \times 10^{-14}$ kg/s. This is the lowest dissipation ever measured for a cantilever. At 4K, this dissipation corresponds to a force noise of 2 attonewtons in a 1 Hz bandwidth.

2.2.3 Single Spin SNR - Surface-induced force noise is important!

Since the force signal for a single electron spin is expected to be ~13 aN-rms and the cantilever force noise at 4K is ~2 aN in a 1 Hz bandwidth, one might think that single-spin detection with SNR on the order of 6 is readily achievable. However, two additional effects must be considered: surface-induced force noise and detection bandwidth.

When a MRFM tip approaches the surface of a sample, a non-contact friction effect has been observed [9]. The cantilever Q is found to drop dramatically, accompanied by a corresponding increase in force noise. A series of careful experiments were performed at IBM to characterize non-contact friction and the corresponding force noise as a function of tip-sample distance [9]. Figure 5 shows the increased noise obtained at 4.2K. At a distance of 200Å from an E' center sample, the surface-induced force noise is approximately 19 aN per root Hz. Thus, even in a 1 Hz bandwidth (which is unrealistically narrow - see next section), the force noise exceeds the expected magnitude of a single spin signal (SNR = 0.7).

The temperature dependence of non-contact friction has not yet been studied at temperatures less than 4K. Extrapolating the trend from higher temperatures, it is likely that lowering the temperature will result in a further reduction of non-contact friction. Even if the friction coefficient $\Gamma_{surface}$ stays constant at the lower temperature, the force noise will still drop significantly since the force noise spectral density is proportional to T [see equation (2)]. By dropping the operating temperature to 100 mK, the resulting force noise will decrease from 19 $\mathrm{aN/Hz}^{1/2}$ to 3 $\mathrm{aN/Hz}^{1/2}$. This noise reduction will prove to be crucial for single-spin detection.

2.2.4 Single Spin SNR - Don't forget the detection bandwidth!

Detection bandwidth is an important (and too often neglected) parameter that must be taken into account when estimating SNR. Ideally one would like to use a narrow detection bandwidth (i.e., long integration time) to minimize the noise. However, in single-spin experiments, the detection bandwidth is dictated by the random flip rate of the spin (i.e., spin relaxation). A detailed study was performed at $T = 4\mathrm{K}$ to measure the influence of the magnetic tip on spin relaxation while a small ensemble of electron spins was undergoing cyclic adiabatic inversion. The essence of the experiment was to monitor the MRFM signal and determine how the long the spins remained "spin-locked" during the cyclic adiabatic inversion. As the tip-to-spin distance was reduced from 1.6 μm to 0.75 μm, the spin locking time τ_m decreased from 275 ms to 65 millisecond (see Fig. 6). It is believed that this effect is due to magnetic field fluctuations emanating from the magnetic tip [10]. Since single-spin experiments will require even closer approach of the tip (e.g., 200Å), then τ_m would be expected to drop even further. Assuming that τ_m =10 ms at 200Å distance, then the single-shot detection bandwidth would roughly correspond to 100 Hz.

Assuming that the force noise density is 19 aN per root Hz (corresponding to the surface-induced noise at 4K), then a 100 Hz detection bandwidth results in a force noise of 190 aN-rms. Assuming the single spin signal is 13 aN-rms, then the single-shot SNR is only 0.07 at 4K.

This SNR is too low to be useful. One might consider doing signal averaging to improve the net SNR. Unfortunately standard signal averaging cannot be applied because the polarity of the single-shot signal is not constant (i.e., it is not known *a priori* whether the spin state is up or down and the spin state flips randomly at a rate given roughly by $1/\tau_m$). Thus, a polarity-independent method of signal averaging must be used (e.g., averaging the square of the signal). Polarity independent averaging techniques have been investigated at IBM [11] and were found to be ineffective unless the single-shot SNR is close to unity. Thus, it is unlikely that single-spin MRFM can be successful at 4K.

2.2.5 Rationale for millikelvin temperature

Reducing the operating temperature to millikelvin temperature (e.g., 100 mK) will have three important beneficial effects:

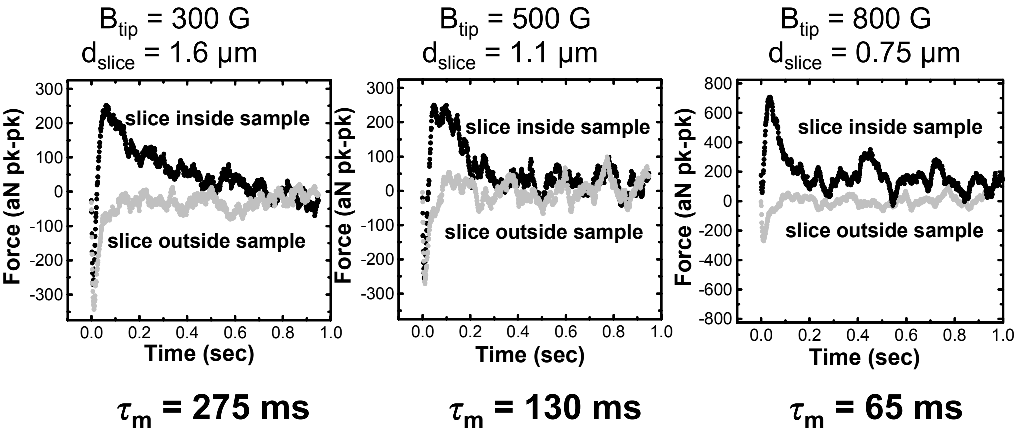


Figure 6: Force signals and the corresponding spin-lock lifetimes obtained for tip-to-spin distances ranging from 1.6 μm to 0.75 μm.

- The cantilever Brownian motion noise will be reduced.
- The surface-induced force noise will be reduced.
- The random flip rate of the spin during adiabatic passages will be reduced. This increases the spin-lock time τ_m and allows a narrower detection bandwidth.

The surface-induced force noise at 100 mK is expected to be approximately 3 aN per root Hz (or less). If a spin-lock time of τ_m =100 ms is achieved, roughly corresponding to a 10 Hz detection bandwidth, then the net force noise will be 10 aN-rms. If the spin signal is 13 aN-rms, then the single-shot SNR is 13 /10 = 1.3.

Since the single-shot SNR is above unity, then polarity-independent averaging can be nearly as effective as normal signal averaging [11]. For a 10 s averaging time, the resulting net SNR would be on the order of 10.

A summary of the SNR analysis is shown in Table I.

The conclusion is clear: **based on our best estimates of the realistic experimental parameters, millikelvin temperature is required for single spin detection.** The biggest unknown factor in estimating the SNR is τ_m. Experiments to explicitly measure τ_m at millikelvin temperature are planned as part of the proposed work in Task 1.

2.3 Detailed Technical Approach - Task 1

2.3.1 Basic elements of a single-spin experiment at 100 mK

Task 1 of this proposal is to develop a single electron spin experiment operating at millikelvin temperature. Figure 7 shows the experimental configuration presently favored at

Table I - Comparison of Single-Electron-Spin SNR at 4K and 100 mK

Temperature	T	**4 K**	**100 mK**
Field gradient	$\partial B_z/\partial x$	2 G/Å	2 G/Å
Signal force	F_{signal}	13 aN-rms	13 aN-rms
Spin-lock relaxation time	τ_m	10 ms	100 ms
Detection bandwidth ($\sim 1/\tau_m$)	$\Delta\nu$	100 Hz	10 Hz
Cantilever thermal noise density	$S^{1/2}_{cantilever}$	6.3 aN/$\sqrt{\text{Hz}}$	1 aN/$\sqrt{\text{Hz}}$
Surface-induced noise density	$S^{1/2}_{surface}$	19 aN/$\sqrt{\text{Hz}}$	3 aN/$\sqrt{\text{Hz}}$
Total noise in bandwidth $\Delta\nu$	F_{noise}	200 aN	10 aN
SNR for bandwidth $\Delta\nu$	$\text{SNR}_{\Delta\nu}$	0.07	1.3
SNR after 10 s polarity-independent averaging	SNR_{ave}	~0.1	~10

IBM. The experiment uses an ultrasensitive cantilever similar to those shown in Fig. 4. The spring constant is on the order of 10^{-4} N/m, which allows attonewton force resolution. Because the cantilever is so compliant and the tip must be approached to within 10 nm of the sample surface, the more traditional horizontal cantilever configuration (Fig. 2) would be prone to "snap-in" by van der Waals forces. To avoid snap-in, the vertical orientation is chosen because even a "soft" cantilever is very stiff in the longitudinal direction. IBM has had extensive experience with this configuration and we are therefore confident of reliable operation close to the sample surface.

The basic operating principles of the experiment are similar to those described in section 2.2.1. The main difference is that the cantilever responds only to lateral forces, so that the detected spin must be located slightly offset from the cantilever axis. Adiabatic inversion of the target spin will be induced by slightly vibrating the cantilever (e.g., 1 nm amplitude) while maintaining a constant microwave frequency. The oscillating field strength from the magnetic tip due to the cantilever vibration will induce the spin inversion. The signal is detected via a small shift of the cantilever frequency. A detailed analysis of this frequency-based detection scheme shows that the basic SNR considerations discussed in sections 2.2.2 - 2.2.5 are still valid.

Magnetic tips The magnetic tip is a critical element of the experiment since it must supply the very large gradient ($\geq$ 2 Gauss/Å) required to generate a detectable force signal. Figure 8 shows two basic tip types that have been developed at IBM: cobalt "nanowire" tips and high anisotropy particle tips. Electron holography measurements revealed that the particle tips, which are fabricated using focused ion beam (FIB) techniques, produce much larger field gradients. The gradient strength $\partial B_z/\partial x$ at a distance of 200Å from the tip was found to be on the order of 2 G/Å.

Further tip investigations are planned under the proposed contract. In particular, lithographically fabricated tips of epitaxially grown FePt will be investigated. FePt has

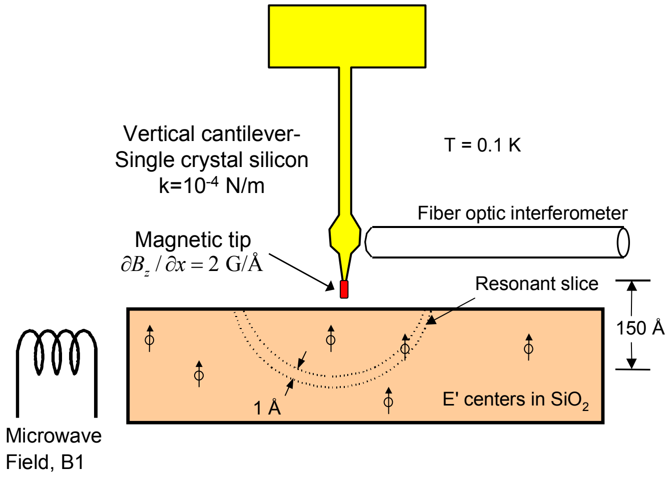
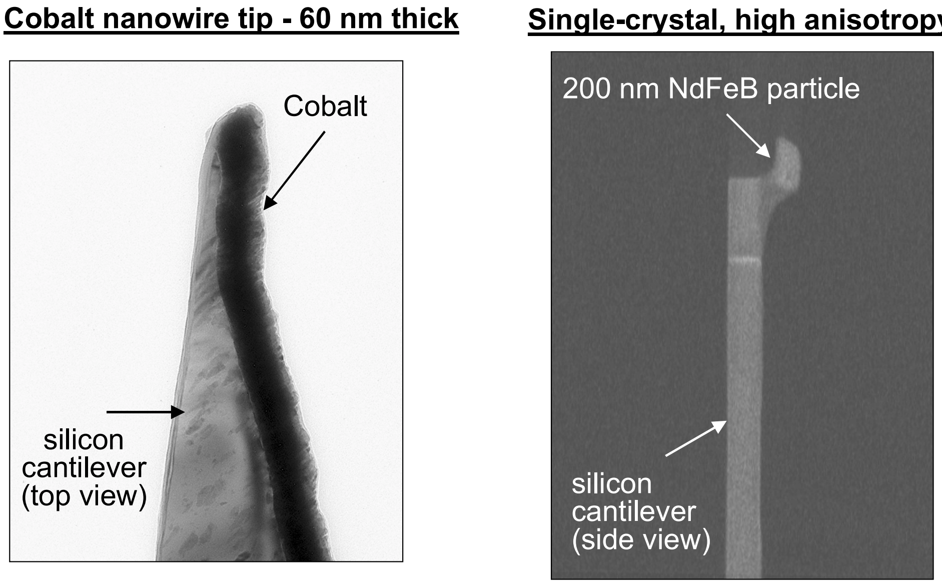


Figure 7: Diagram of proposed single electron spin experiment. The vertical cantilever configuration allows the tip to approach the sample without being “snapped in” by van der Waals forces.


Cobalt nanowire tip - 60 nm thick

Single-crystal, high anisotropy tip

Figure 8: Two types of magnetic tips developed for MRFM. The cobalt nanowire tips are fabricated by evaporating cobalt onto the cantilever sidewall. The high anisotropy particle tips are fabricated by gluing micron-size particles of NdFeB or SmCo onto the cantilever and then using a focused ion beam to shape the tip to the desired size. The particle tips are preferred because they produce much larger field gradients than nanowire tips.

large moment and very high anisotropy, making it a suitable tip material. By using lithographic fabrication techniques, it is anticipated that the tips will avoid "dead layer" problems that result from ion-beam damage incurred during FIB fabrication.

Spin system The sample will consist of a low concentration of unpaired electron spins associated with silicon dangling bonds in silica (E′ centers). The dangling bonds are created with controllable concentration by gamma irradiation. IBM has studied this spin system extensively and the magnetic resonance properties are nearly ideal for an initial single-spin experiment. In particular, the ability to cyclically invert the spins for thousands of cycles using adiabatic rapid passage has been demonstrated. The only major drawback of this system is that the E′ centers are charged. This leads to internal charge inhomogeneity, resulting in significant stray electric field that emanates from the sample surface. This stray electric field results in increased surface-induced force noise. This noise should be acceptably small at millikelvin temperatures.

After single-spin signals are successfully acquired using our most idealized samples, alternative spin systems can be considered, including dopants in silicon and bio-compatible spin labels.

2.3.2 Technical challenges at millikelvin temperatures

Operation at millikelvin temperature presents some serious technical issues that must be addressed. The two most serious issues are: 1) heating of the experiment by the microwave power and 2) heating of the cantilever by optical radiation from the interferometer.

Microwave power issue The cooling power of a large dilution refrigerator (such as the one at IBM) is typically 400 microwatts for an operating temperature of 100 mK. This small cooling power can be easily overwhelmed when generating the microwave magnetic field. For example, in the 4K experiment previously operated at IBM, a microwave power of roughly 100 milliwatts was required to generate a microwave magnetic field of 2 Gauss using a 200 μm diameter coil. This is a factor of 2500 larger than can be tolerated in the dilution refrigerator!

To obtain the necessary large reduction in power dissipation, a superconducting resonator for MRFM is under development at IBM. As shown in Figs. 9 and 10, the resonator consists of a 220 μm diameter niobium coil wound with 25 μm niobium wire that is wire-bonded to a short section of niobium microstripline. Initial tests at temperatures down to 100 mK have demonstrated resonator quality factors of several thousand. Pulsed microwave fields as large as 1.6 Gauss were measured based on observation of the electron spin Rabi frequency (i.e., spin nutation).

Although this is a promising result, further improvements in critical current characteristics of the resonator are needed in order to increase the field above 2 Gauss. (2 Gauss is considered the minimum field necessary to achieve efficient cyclic adiabatic inversion.) In addition, the power dissipation is still too large for continuous (cw) operation by about a factor of 20. Although low duty cycle techniques could mitigate this issue, continuous

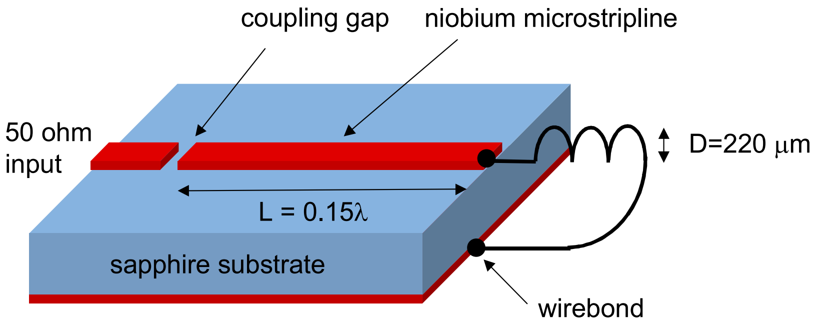


Figure 9: Superconducting resonator that generates the microwave magnetic field. Both the coil and the microstripline are fabricated from niobium.

operation is preferred. Further development of the superconducting resonator is planned under the proposed contract.

Nanowatt interferometry Another key consideration at millikelvin temperature is the heating of the cantilever by optical radiation from the interferometer. At millikelvin temperature, the thermal conductivity of silicon drops rapidly, following a T^3 law as the phonons freeze out. This fact, combined with the long, narrow geometry of the cantilever, makes the cantilever an extremely poor conductor of heat. To avoid significant heating of the cantilever, the absorbed optical power must be in the femtowatt range.

IBM has developed an ultralow power fiberoptic interferometer for millikelvin MRFM applications (see Fig. 11). The basic fiberoptic interferometer, originally developed by Rugar et al. [12] for atomic force microscopy (AFM), relies on the interference between the reference light reflected from the cleaved fiber end and the light reflected by the cantilever. To improve performance at millikelvin temperature, a wavelength of 1550 nm was chosen so that the photon energy is less than the bandgap of silicon. At this wavelength, silicon has very little absorption (the residual absorption is mainly due to impurity and surface effects). To achieve good reflectivity from the bare silicon cantilever, the thickness of the cantilever is tuned to an odd multiple of quarter-wavelength. Because the refractive index of silicon is so high ($n = 3.5$), the reflectivity can be as high as 70% for cantilevers as thin as 100 nm.

To improve the interferometer SNR, two additional steps are taken: 1) the reference reflection from the end of the fiber is optimized by applying a 70 nm silicon coating. This coating acts as a partially reflecting dielectric mirror; 2) an avalanche photodiode is used in order to boost the signal and shot noise above the electronic noise of the photodiode preamplifier.

To demonstrate operation of the interferometer, measurements of cantilever vibrational noise were made at temperatures down to 100 mK [13]. As can be seen in the noise

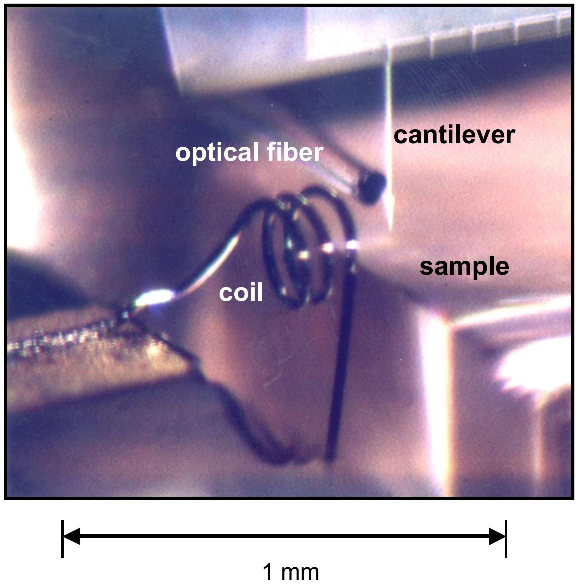


Figure 10: Photograph of an MRFM setup showing the configuration of the coil with respect to the sample, cantilever and optical fiber. The coil diameter is 220 μm.

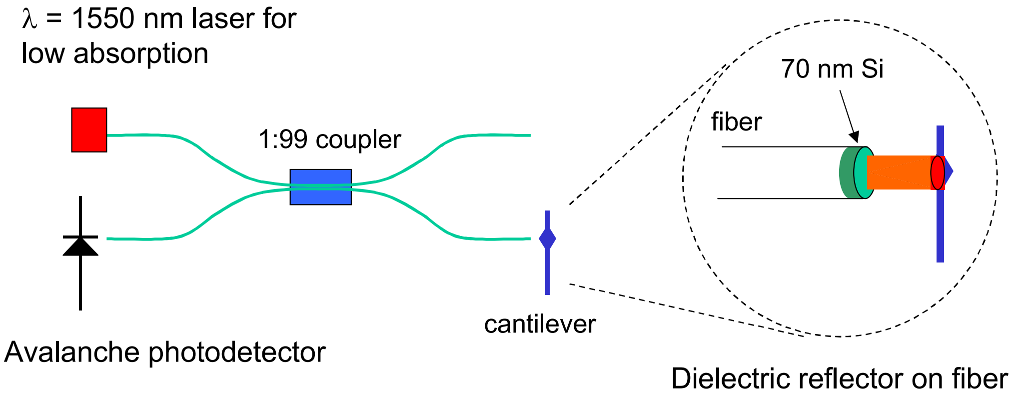


Figure 11: Ultralow power (nanowatt) interferometer for millikelvin MRFM applications.

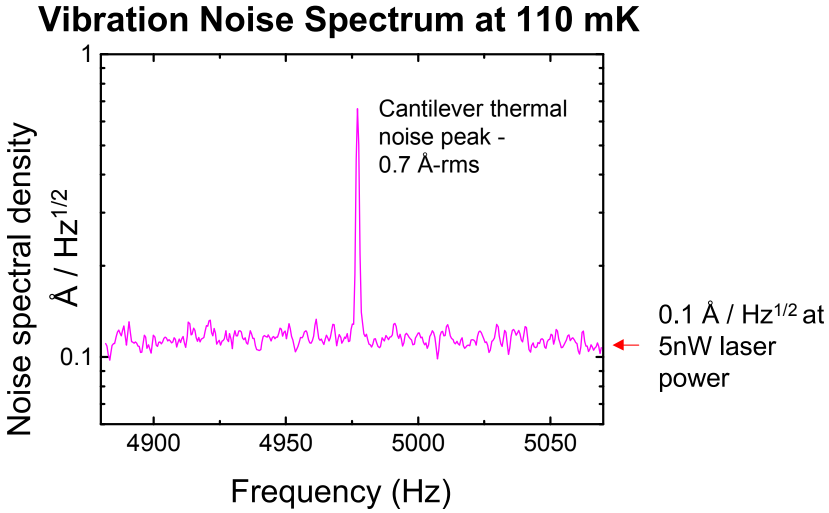


Figure 12: Thermal noise of cantilever measured at 110 mK using only 5 nanowatts of interferometer power.

spectrum shown in Fig. 12, the thermal vibrations of the cantilever can be easily seen with only 5 nanowatts of optical power incident on the cantilever.

The dependence of the thermal vibration amplitude on temperature was carefully measured in order to determine the cantilever noise temperature. As shown in Fig. 13, the thermal vibration amplitude followed the expected $T^{1/2}$ dependence until the very lowest temperature was reached. The measured vibration noise at 100 mK was 0.6 Å-rms, which is equivalent to a noise temperature of only 220 mK. This demonstrates that the optical heating of the cantilever was at most 120 mK. Based on the expected cantilever thermal resistance, this suggests that the absorbed optical power was on the order of 5 femtowatts or less.

2.3.3 Preliminary work demonstrating millikelvin MRFM

Preliminary work has been performed to demonstrate MRFM at millikelvin temperature using micrometer-size E$'$ center samples. As shown in Fig. 14A, the MRFM signal was detected with excellent signal-to-noise ratio. In addition, a number of basic magnetic resonance measurements were made, including measurement of the spin-lattice relaxation time (T_1). As seen in Fig. 14B, extraordinarily long T_1's were observed (e.g., nearly 5 minutes at 100 mK).

The experience gained from these preliminary millikelvin experiments will greatly accelerate the development of single-spin MRFM under the MOSAIC program.

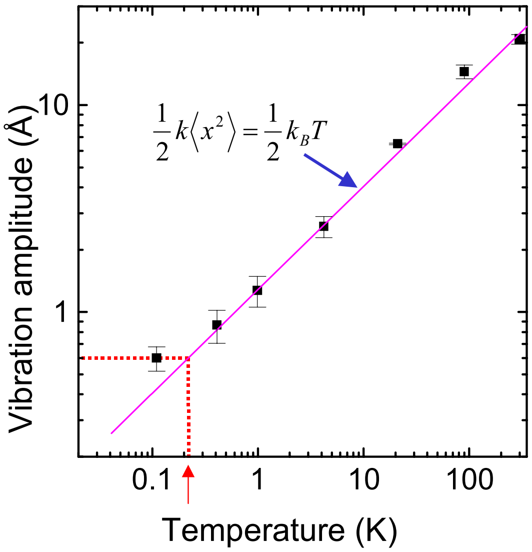


Figure 13: Cantilever vibration amplitude vs temperature showing dependence predicted by the equipartition theorem. To avoid cantilever heating, an interferometer wavelength of 1550 nm was used and the power was limited to a few nanowatts. At the 100 mK base temperature, the observed vibration noise was 0.6Å, equivalent to a noise temperature of 220 mK. The corresponding force noise was 820 zeptonewtons per root Hz, which is the lowest force noise ever observed for a mechanical force sensor.

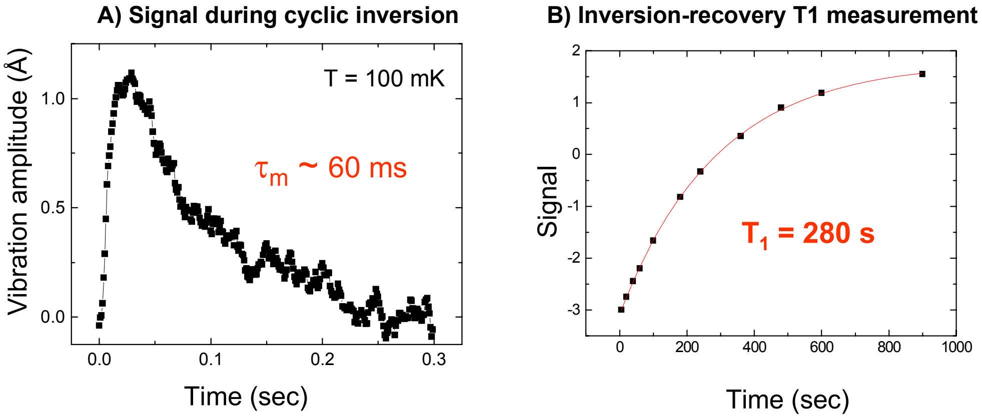


Figure 14: Results from the first millikelvin MRFM experiment. A) Cantilever vibration signal resulting from cyclic inversion of E′ centers at $T = 100$ mK. B) Inversion-recovery measurement of the spin-lattice relaxation time at 100 mK. T_1 was found to be nearly 5 minutes.

Table II - Parameters for initial Larmor detection demonstration

Temperature	T	100 mK
Cantilever dimensions	$l \times w \times t$	20 μm × 3 μm × 0.28 μm
Spring constant	k	0.3 N/m
Frequency	f	0.75 MHz
Quality factor	Q	50,000
Force noise density (preamp-limited)	$\mathrm{S}_f^{1/2}$	6 aN/$\sqrt{\mathrm{Hz}}$
Tip dimensions	$l_{tip} \times w_{tip} \times t_{tip}$	2 μm × 2 μm × 2 μm
Lateral field gradient	$\partial B_x/\partial x$	0.8 Gauss/nm
Resonant slice thickness	$\Delta z_{slice} = B_1/(\partial B_z/\partial z)$	9 nm
Net number of nuclear spins (^{19}F)	N_{net}	10^5
Signal amplitude	F_{signal}	100 aN
SNR in 1 Hz bandwidth	SNR	∼16

2.4 Detailed Technical Approach - Task 2

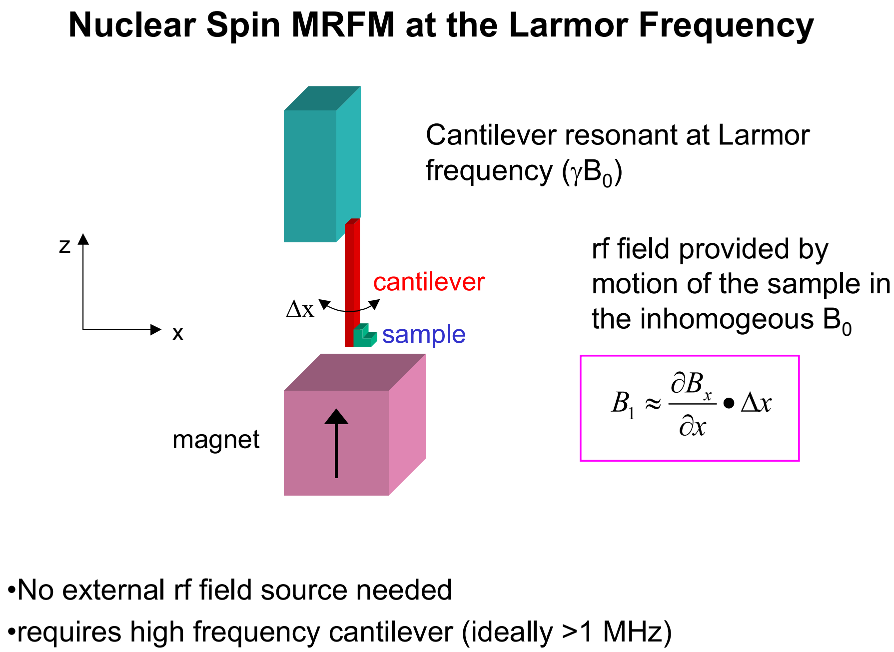
2.4.1 Larmor frequency detection of NMR

Task 2 of this proposal is to demonstrate direct Larmor-frequency detection of nuclear spin precession. Direct detection is believed to be the most promising technique for eventually achieving single nuclear spin sensitivity since it eliminates the coil as a source of heat and disturbance.

The key component in a Larmor frequency experiment is the high frequency cantilever. The cantilever should have the highest possible resonance frequency while maintaining excellent force resolution. To achieve this combination of properties, the cantilever must have the smallest possible mass (i.e., short, narrow and thin). Task 3 addresses the ultimate limits of high frequency cantilever performance. The present task (Task 2) concentrates on an initial demonstation of Larmor-freqency detection using a submicron-size ensemble of spins.

Two possible configurations are suitable for an initial demonstration: sample-on-cantilever or tip-on-cantilever. Fig. 15 shows the sample-on-cantilever configuration. An external coil is not required since the rf field is generated by oscillating the cantilever. For a cantilever vibrating with amplitude Δx, the effective rf field strength is $B_1 = (\partial B_x/\partial x)\Delta x$, where B_x is the transverse component of the tip field. B_1 should be larger than local fields within the sample in order to achieve a good "spin-lock" (i.e., long rotating-frame spin-lattice relaxation time, $\mathrm{T}_{1\rho}$). For materials with high nuclear spin density, $B_1 \geq 10$ Gauss is required.

Table II shows the key parameters for an initial demonstration experiment. An easily fabricated tip size is assumed (2 μm × 2 μm × 2 μm), yielding a lateral gradient $\partial B_x/\partial x =$ 0.8 Gauss/nm. With a driven vibration amplitude of 100Å, an effective slice thickness $\Delta z_{slice} = B_1/(\partial Bz/\partial z) = 10$ nm is expected.



Nuclear Spin MRFM at the Larmor Frequency

$$B_1 \approx \frac{\partial B_x}{\partial x} \bullet \Delta x$$

- No external rf field source needed
- requires high frequency cantilever (ideally >1 MHz)

Figure 15: A Larmor-frequency MRFM experiment with the sample-on-cantilever configuration.

The experiment assumes a sample of CaF_2, though many other samples would also be suitable (e.g., GaAs, Cu, Pt, etc.). The main sample requirements are a large spin density and a relaxation time ratio $T_{1\rho}/T_1$ close to unity. Biological samples are also possible, but, first, relaxation time measurements should be performed at millikelvin temperature to verify that $T_{1\rho}/T_1$ is reasonable.

After the first Larmor-frequency experiment is successful, then rapid progress should be possible. The extension of this technique toward single nuclear spin detection depends primarily on progress in ultrasensitive force detection. This is the subject of the next task.

2.5 Detailed Technical Approach - Task 3

2.5.1 Pushing the limits of force detection

The ultimate goal of the MOSAIC program is to develop a technique capable of three-dimensional molecular imaging. If MRFM is to fulfill this goal, the capability to detect individual nuclear spins must be achieved.

As discussed in Section 2.2.1, the force from a single nuclear spin is extremely small. Even if we assume a tip field gradient of 20 Gauss/Å (10× larger than that assumed in Task 1 for single electron spin detection), the force on a proton (^{1}H nucleus) is only 0.2 aN-rms. This force signal is 4× smaller than has ever been detected with low frequency

Table III - Future Ultrasmall cross-section (USC) Cantilevers

Parameter	Symbol	Present Cantilever Design	Future Low frequency Design	Future High frequency Design
Width	w	3000 nm	400 nm	100 nm
Thickness	t	270 nm	100 nm	50 nm
Length	L	200 μm	100 μm	5 μm
Reflector size	w_r	25 μm	2 μm	0.7 μm
Spring constant	k	100 μN/m	7 μN/m	4700 μN/m
Frequency	f	3 kHz	8 kHz	1.3 MHz
Quality factor	Q	150,000	500,000	200,000
Force noise @ 100 mK	$S_F^{1/2}$	0.82 aN/$\sqrt{\text{Hz}}$	0.05 aN/$\sqrt{\text{Hz}}$	0.13 aN/$\sqrt{\text{Hz}}$

cantilevers and at least 50× smaller than has been demonstrated with high frequency cantilevers (assuming unity SNR in a 1 Hz bandwidth).

The objective of Task 3 is to develop technology and methodology that pushes cantilever-based force detection substantially beyond current limits. We consider three approaches: 1) fabricating cantilevers with ultrasmall cross-section, 2) reducing dissipation via surface cleaning and 3) improved optical detection.

2.5.2 Cantilevers with ultrasmall cross-section (USC)

Based on equations (2) and (4) in section 2.2.2, the cantilever force noise density can be written as

$$S_F^{1/2} = \left(\frac{wt^2}{L\,Q}\right)^{1/2} (E\rho)^{1/4} (k_BT)^{1/2}. \tag{5}$$

As discussed previously, the geometrical dependence embodied in this equation was the basis of our very successful "narrow, thin and long" cantilever strategy.

By using electron beam lithography (or advanced optical lithography), this approach can be extended to even narrower and thinner cantilevers. Table III shows a comparison between our present cantilevers and possible future cantilevers with ultrasmall cross-sections (USC).

As can be seen in the table, force noise well below 0.1 aN/$\sqrt{\text{Hz}}$ may be achievable with low frequency USC cantilevers, provided that very high Q can be maintained. High frequency cantilevers are at some disadvantage compared to low frequency cantilevers because of their greater stiffness. Nevertheless, it appears that force noise below 0.2 aN/$\sqrt{\text{Hz}}$ may be achievable. A demonstration of cantilever performance with this level of force noise would represent important progress toward single nuclear spin MRFM.

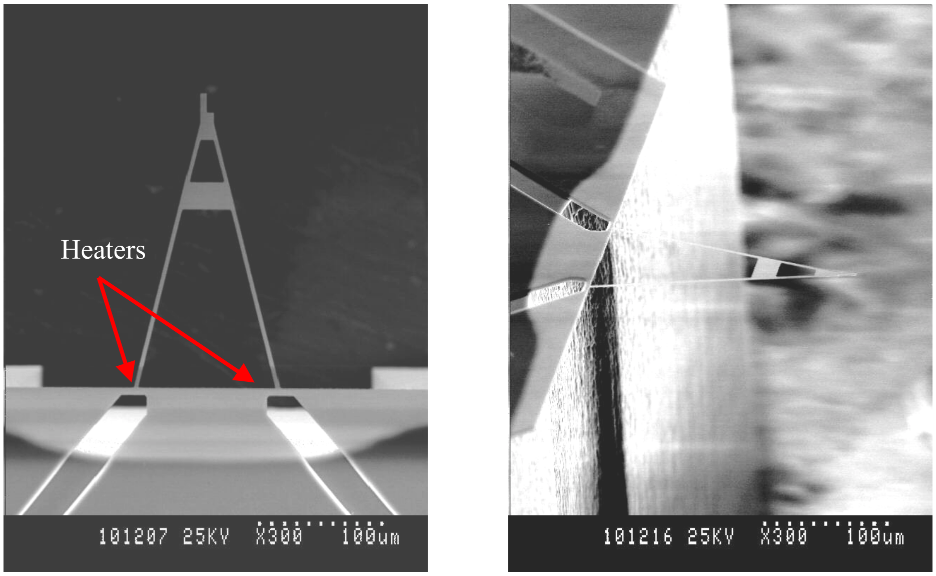


Figure 16: Two views of a cantilever with integrated electrical heaters for desorbing contaminants from the cantilever surface.

2.5.3 Achieving high Q in ultrasmall cross-section cantilevers

The sub-attonewton force noise predicted in Table III for USC cantilevers depends on maintaining high quality factor, despite the very small cross-sectional dimensions. Maintaining high Q is expected to be a major challenge since previous studies have found that Q typically decreases with decreasing cantilever dimensions (especially thickness). Careful studies by the Stanford-IBM team [8] and others[14, 15] have demonstrated that the decline in Q with oscillator size is due to oxidation and contamination of the oscillator's surfaces. As the surface-to-volume ratio of an oscillator becomes larger, this effect becomes more important. With this knowledge in hand, the path to reduced oscillator dissipation is clear: **keep the cantilever surface clean!**

One simple method for cantilever cleaning is to desorb surface adsorbates (water and organic contamination) by heating the cantilever in vacuum. The most elegant way to do this is to incorporate a heater element into the structure of the cantilever. Figure 16 shows a cantilever fabricated by the Kenny group at Stanford that has built-in micromachined heaters. Cantilever temperatures up to 700°C can be achieved. After heat treatment, significantly enhanced cantilever Q was found, as shown in Fig. 17.

This basic idea should be extendable to ultrasmall cross-section cantilevers. Fig. 18 shows one possible cantilever design.

2.5.4 Detection issues for USC cantilevers

As cantilever dimensions shrink, detection of the cantilever motion becomes a major issue. In the case of fiberoptic interferometry, it becomes difficult to obtain a good reflection

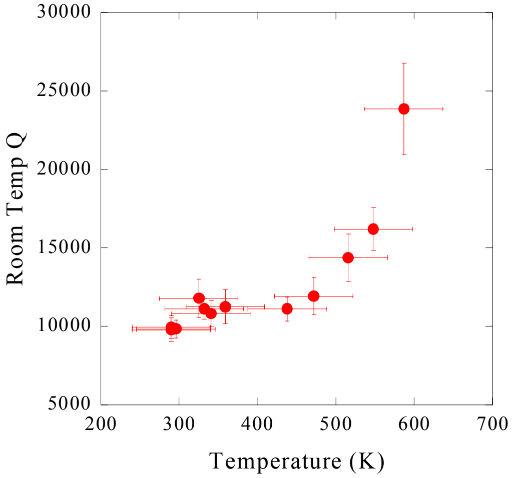


Figure 17: Room temperature Q following cantilever heating.

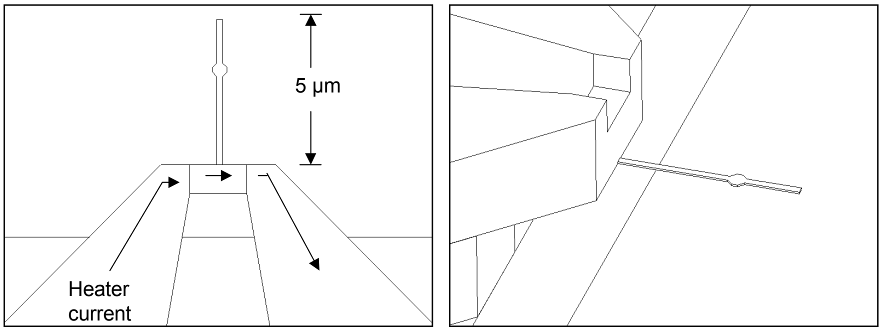


Figure 18: Possible design of ultrasmall cross-section cantilever with built-in heater element.

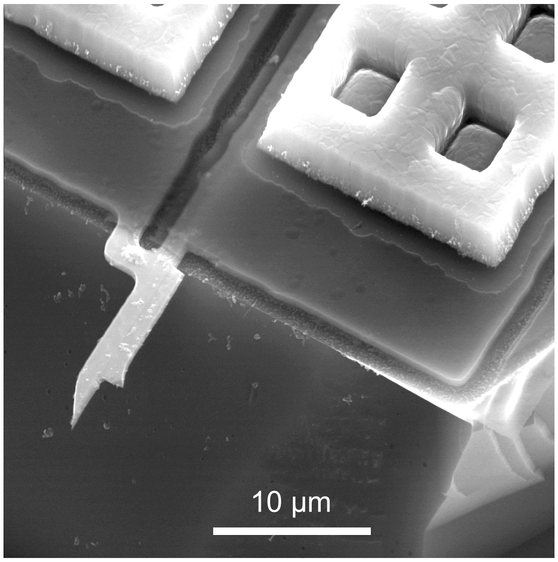


Figure 19: Scanning electron micrograph of 12 μm long piezoresistive cantilever with 4.6 MHz resonance frequency. The cantilever was designed at IBM and fabricated at Stanford.

signal from the cantilever since the beam size, typically 5 - 10 μm in diameter, is so much larger than the cantilever width. Previous IBM-Stanford cantilevers incorporated a 25 μm-wide reflector to facilitate good reflectance back into the fiber. For ultrasmall cross-section cantilever, we will design a much smaller reflector, on the order of 1 μm in diameter. To get good coupling efficiency with the optical fiber, we will incorporate a high numerical aperture objective lens to improve the coupling efficiency and build a three-dimensional piezo-actuation stage to align the focused optical beam onto the reflector.

Piezoresistive strain sensors are another means for sensitively detecting cantilever motion Great strides have been made recently in fabricating extremely thin silicon piezoresistors by shallow ion implantation or, better yet, by growing very thin doped epi-layers. Cantilevers with integrated sensors have been fabricated as thin as 900Å by the Kenny group at Stanford [16]. Figure 19 shows a high frequency (4.6 MHz) piezoresistive cantilever designed at IBM [17].

The primary disadvantage of the piezoresistive sensor is the heating of the piezoresistor by the measurement current. Calculations suggest that self-heating of the cantilever will limit force the force resolution to 1 aN or greater. Nevertheless, these types of sensors are worthy of further study.

As the ability to detect cantilever motion becomes limited by detection issues rather than cantilever Brownian motion, mechanical parametric amplification techniques may be helpful. The concept of using mechanical parametric amplification to overcome detection noise was developed by Rugar and Grütter at IBM in 1991[18]. The basic idea is that mechanical sigal gain can be achieved by modulating (i.e., “pumping”) the cantilever spring constant at twice the signal frequency. The amplification is virtually noise-free and allows both the signal and Brownian motion noise to be increased above the noise level of the detection system. This idea will be explored in more detail as part of this proposed contract.

2.6 Detailed Technical Approach - Task 4

Task 4 of this proposal is to develop a high speed digital controller for optimal feedback control of high frequency cantilever dynamics.

2.6.1 Fundamentals of Optimal Control

The theory and experimental practice of optimal control have been addressed in articles by the UW group. [19, 20]; these principles have been very successful in practical MRFM experiments.

Here we will review only the main results as they relate to the control of high-frequency (Larmor) cantilevers. With reference to Fig. 20, we begin by asking, what is the optimal form of the controller transfer function $H(s)$? The answer is mainly determined by the relative strength of the measurement noise $v(t)$, which we regard as a random function with spectral density V, and the process (thermal) noise $w(t)$ having spectral density W. As shown in [19], the key parameter is a dimensionless ratio α given by

$$\alpha \simeq \frac{W}{k^2 V} \tag{6}$$

where k is the spring constant of the cantilever.[1] In principle larger values of α are desirable, values in the range $10^{-3} - 10^{-2}$ are achievable in practice.

2.6.2 Downconversion/Upconversion Strategies

Digital signal processing offers a flexible and convenient means of achieving dynamic control of force microscope cantilevers.

Given a system with $\alpha \ll 1$, the optimal controller $H(s)$ takes a particularly simple form when it is implemented via a digital downconversion/upconversion strategy as shown in Fig. 21. In this strategy, the high-frequency cantilever signal is downcoverted to a low-frequency signal, filtered in the low-frequency domain, then upconverted to a control signal.

An important advantage of the downconversion/upconversion method is that the sampling rates in the low frequency stage of the controller are slow enough that sophisticated on-the-fly calculations can be carried out. Starting with the formalism of [19], the optimal low-pass transfer function $H'(s)$ can be shown to be a simple single-pole filter:

$$H'(s) \simeq k \, \frac{\alpha\beta}{\alpha + \beta} \, \frac{1}{1 + s\tau_{oc}} \tag{7}$$

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[1] Close reading of [19] will show that Eq. 6 is an approximation, but it is a *very* accurate approximation for practical MRFM experiments.

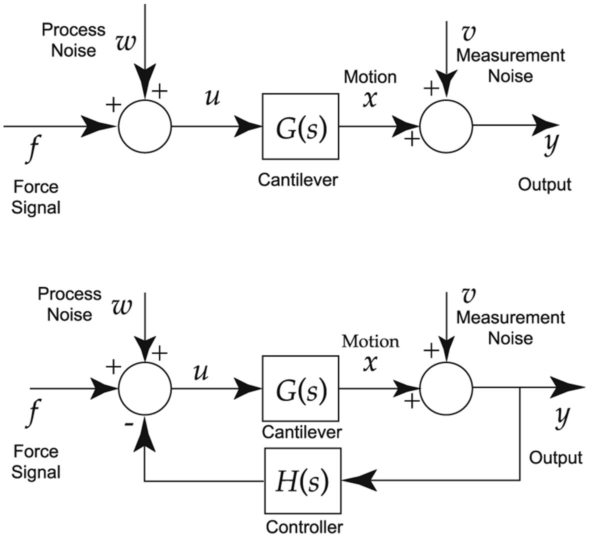


Figure 20: Uncontrolled versus controlled cantilevers.

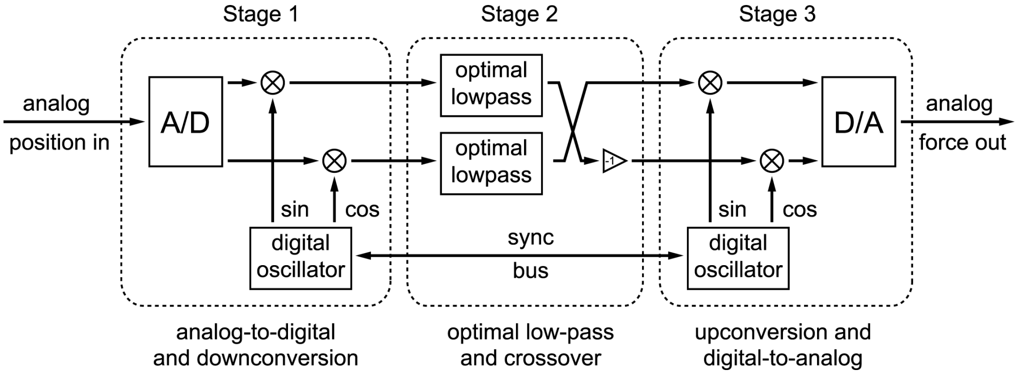


Figure 21: Digital signal processing for Larmor-frequency controllers via a downconversion/upconversion strategy.

which is a single-pole filter whose time constant τ_{oc} is given by

$$\omega_n \tau_{oc} = \frac{2}{\alpha + \beta} = 2 Q_{oc} \tag{8}$$

where ω_n is the natural frequency of the cantilever, and Q_{oc} is the closed-loop quality factor in the limit $\{\alpha, \beta\} \ll 1$.

The main engineering parameter in the above equations is β. Larger values of β lead to larger control forces, and a broader cantilever frequency response.

2.6.3 DSP Hardware Considerations

DSP devices like that shown in Fig. 21 are characterized by a digital latency τ_L, which can be compensated by standard phase-lead techniques. At downconverted frequencies ω such that higher-order terms of order $(\omega \tau_L)^2$ are negligible, compensation is most readily achieved by the simple substitution $\tau_{oc} \to \tau_{oc} - \tau_L$, combined with the insertion of a phase lead $\phi = \omega_n \tau_L$ into the upconversion mixer. For this method to yield accurate latency compensation within the controller bandwidth, the inequality $\tau_L \ll \tau_{oc}$ must be satisfied, *i.e.*, the digital latency must be short compared to the time constant of the optimal controller. This is a physically reasonable requirement.

In a typical MRFM experiment the desired control bandwidth $b_{oc} = \omega_n / Q_{oc} = 2/\tau_{oc}$ is of order 300 Hz (at present) to 3000 Hz (for advanced Larmor devices), corresponding to a low-pass filter time constant τ_{oc} of order one millisecond to 100 microseconds. Thus, in designing a general-purpose MRFM optimal controller of the downconversion/upconversion type shown in Fig. 21, the digital latency τ_L should be appreciably less than 100 microseconds, with 10–20 microseconds being a reasonable goal. Modern DSP devices readily achieve such latencies.

To test these ideas, a downconversion/upconversion optimal controller was “breadboarded” at UW and this prototype was used to successfully control the existing UW MRFM apparatus. Again with reference to Fig. 21, Stages 1 and 2 were implemented in a Stanford Instruments SR830 digital lockin amplifier, which supplied the downconversion and low-pass filtering functions, while Stage 3 was implemented by using the SR830 X-channel and Y-channel analog amplitudes to amplitude-modulate a pair of Hewlett-Packard DS345 signal generators, which supplied the upconversion function; the DS345 output signals were then combined via a Kronheit unit-gain differential amplifier.

Downconversion/upconversion controllers for Larmor cantilevers will require megahertz carrier frequencies and full programmability, neither of which could be achieved with our SR830/DS345 prototype.

After a review of available DSP systems, we determined that Pentek offers a commercial off-the-shelf (COTS) product (see Fig. 22) capable of implementing a single-card, fully programmable downconversion/upconversion controllers, suitable for cantilever frequencies up to $\sim$ 35 MHz, with a digital latency of no more than 10–20 μs, which is adequate for our purposes

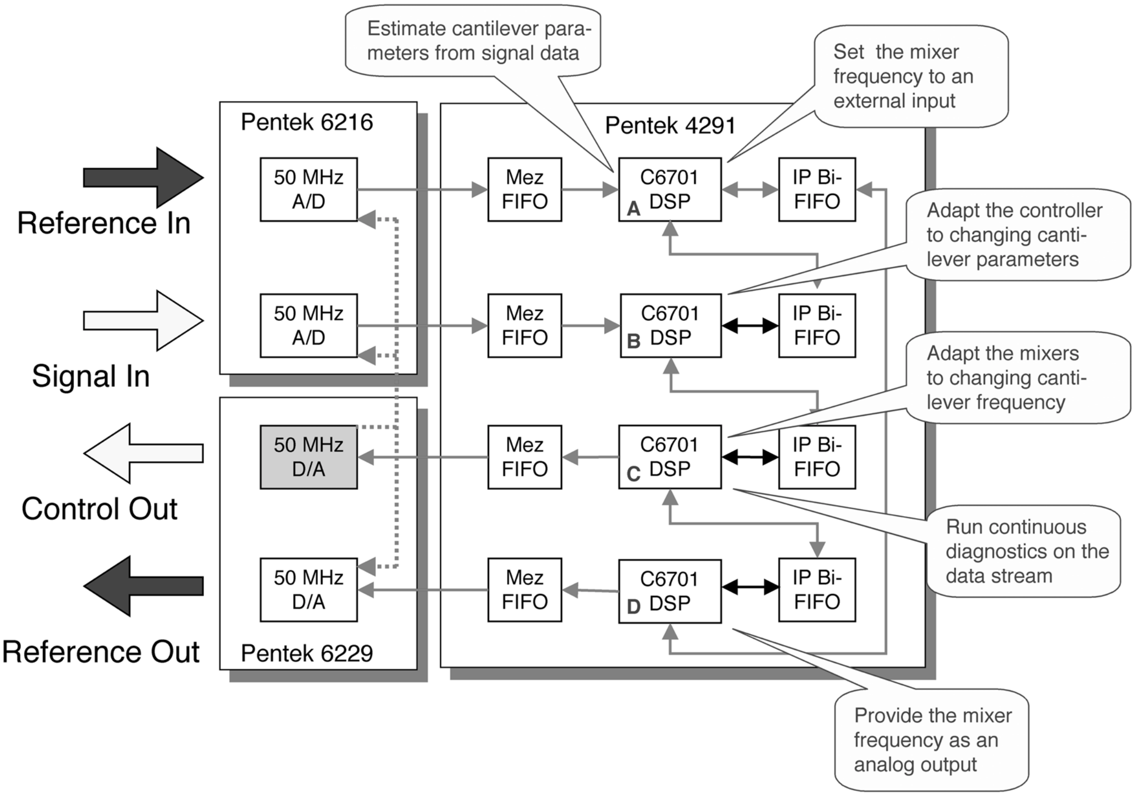


Figure 22: Implementation of an advanced Larmor controller using Pentek DSP products. In addition to providing basic control of high frequency cantilevers, the controller will perform diagnostic tests and be adaptive to changing conditions (i.e., tip-sample interactions).

The UW group will acquire a single Pentek board of the type shown in Fig. 22, and then work with consulting engineers from the DSP design firm DSPcon, Inc. to demonstrate that this hardware can achieve the following critical milestones: (1) synchronization of the downconversion and upconversion oscillators, and (2) reducing digital latency to below 20 μs, and (3) implementation of a single-pole filter in Stage 2.

Once these milestones have been achieved, UW will acquire three additional DSP systems, for a total of four; this will supply IBM and the UW each with a Larmor emulator/controller pair.

2.6.4 Reference Design Emulation

Under a separate DARPA/DSO contract *The Accelerated Development of MRFM*, the UW MRFM group is developing a reference design and performance analysis (RDPA) for a molecular observation technology. This reference design is based on Larmor cantilevers similar to those proposed for development under Tasks 2 and 3 of the present proposal. In order to accelerate testing of the controller before actual RDPA Larmor cantilevers are available, the UW group will emulate the cantilever characteristics in one DSP, control the emulated cantilever with a second DSP, and then inject test signals characteristic of single-spin challenge targets. This approach will greatly accelerate controller testing, and allow software development to begin at a very early stage.

Using an aircraft analogy, this strategy ensures that when the airframe is ready (*i.e.*, the Larmor cantilevers and tips are ready), the avionics (*i.e.*, the control system) is ready too.

2.6.5 Diagnostics

Control techniques serve many purposes in MRFM above and beyond the strictly literal notion of “control”. Since all the noise in an MRFM experiment flows through and is processed by the controller, controllers are the most natural site for on-line diagnostics.

In UW MRFM experiments, twenty-two different statistical tests are routinely performed on the cantilever signal to give instant feedback to the MRFM scientist. These tests include: mean, trend, skew, kurtosis, χ-square, Anderson-Darling (in both the time and frequency domains), mean power, decorrelation rate, quantization noise, and signal-to-noise ratio. The results are presented on a LabView computer screen in a “dashboard” format: A green light indicates that a given test provides no grounds to suspect the noise is non-thermal, a yellow light indicates a less-than-5% chance that the test result could arise from thermal noise, and red lights indicate less than 1% chance that the result is random—so a red light is a strong indication of a problem with the experiment!

The dashboard display format is borrowed directly from avionics. We find that it provides indispensible diagnostic information when we are “flying” our MRFM experiments.

2.6.6 Advanced DSP applications

As the MRFM tip approaches the sample surface, the cantilever transfer function (resonance frequency, Q, etc.) can undergo significant change. In addition, the noise environment is influenced by tip-sample interactions. Thus it is important for MRFM controllers to be not only diagnostic, but also adaptive.

Such an adaptive, autocalibrating capability exists in the present UW digital DSP controller, but communication between this controller and the outside world is slow and awkward; we are eager for the greater bandwidth and flexibility that the downconversion/upconversion controllers will provide.

2.7 Detailed Technical Approach - Task 5

2.7.1 Spin dynamics in large field gradients

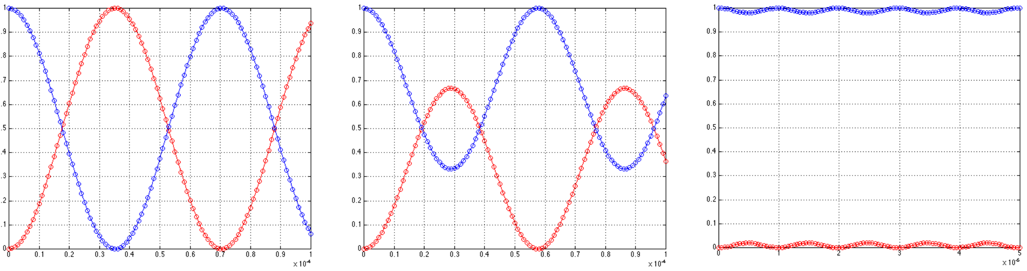
Achieving the ultimate goal of determining three-dimensional atomic structure in biomolecules will depend on suitable behavior of spins in the large field gradient of the MRFM tip. Because nuclear spins in a biomolecule have spacings on the order of 1 - 2 Å, there exists the possibility that rapid flip-flops (i.e., spin exchange) between adjacent spins will disrupt the imaging process. In principle, spin exchange will be suppressed by the large field gradient from the MRFM tip.

Task 5 addresses the issue of spin-spin interactions in the context of MRFM. The calculations will start with simple model systems (e.g., a linear array of spins in a one-dimensional gradient) and then work towards more realistic models. Among the questions to be addressed in this work are:

- What is the minimum linear gradient required to quench spin exchange to a degree sufficient to permit the resolved observation of a single spin in a one-dimensional lattice?
- Does the minimum strength depend upon the presence of remote neighbor spins?
- For spins in a cubic lattice, subject to the gradient of a spherical MRFM tip, how does the number of spins within the ‘sensitive slice’ vary with the distance from the tip?
- Do the spin dynamics point towards the prospect of extracting molecular-structural information from any components of the spin Hamiltonian? Do the spin dynamics themselves suggest a detection scheme?

2.7.2 Methodological Issues and Progress to Date

Preliminary calculations have shown that the evolution due to dipole-mediated spin exchange of a single member of a proton pair, subjected to a linear gradient along the



grad = 500 Hz/Å ~ 0.12 G/Å
$\omega_{\mathrm{eff}}/(2\pi)$ = 14 kHz δ=1.5

grad = 5000 Hz/Å ~ 1.2 G/Å
$\omega_{\mathrm{eff}}/(2\pi)$ = 17 kHz δ= 0.96

grad = 50000 Hz/Å ~ 12 G/Å
$\omega_{\mathrm{eff}}/(2\pi)$ = 101 kHz δ= 0.14

Figure 23: Time course of spin z component for a proton pair in three different field gradients. Spin exchange is nearly fully quenched for a gradient of 12 G/Å.

inter-proton axis, can be written in closed form as:

$$\langle I_{z1}(t)\rangle = \frac{1}{2} \pm \frac{1}{2}\left[\cos^2(\omega_{eff}t/2) + \cos 2\phi \sin^2\left(\omega_{eff}t/2\right)\right], \tag{9}$$

where $\langle I_{z1}(t)\rangle$ is the time dependent z component expectation value of one proton, $\tan\phi$ is given by $k/2\delta$, where k is the angle-dependent dipolar coupling constant in Hz and δ is the gradient strength also in Hz, and $\omega_{eff} = \left(k^2/4 + \delta^2\right)^{1/2}$. Calculations using this formula show that spin exchange is effectively quenched, and single spin detection thereby enabled, at a gradient strength of 12 G/Å (see Fig. 23).

Unfortunately, the simple derivation used in this case is not extensible to multiple spins, and it appears that brute force diagonalization of the Hamiltonian, and calculation of the time dependent propagator will be required. While this will lead to large matrices (e.g. 1024 × 1024 for 10 protons), these are expected to be band-diagonal, if (as anticipated) it develops that spin interactions between remote neighbors can be neglected beyond, say 15 Å. (The fall off in the energy of interaction, relative to a near neighbor at 2 Å, would be ≈99%.)

The Hamiltonian matrices will be written in an interaction representation in which the external gradient (plus any uniform polarizing field) both vanish at the position of the 'reference spin' whose evolution will be calculated. The calculations will start with a linear array of spins and then progress to a three-dimension body-centered cubic configuration. Finally, spin configuations representative of simple molecules will be studied.

2.8 Detailed Technical Approach - Task 6

2.8.1 Signal and Image Processing for MRFM

Signals from single-spin MRFM experiments are anticipated to be very small and have statistical characteristics that will require specialized signal processing techniques. The key issue is that the initial quantum state of the spin is unknown. When the detection process collapses the spin wavefunction to an eigenstate of the effective field, the spin will be found in either the spin up or the spin down states with almost equal probability. Thus, there is no *a priori* knowledge of the polarity of the single spin signal. Furthermore, there is substantial likelihood that the polarity of the spin signal will spontaneously flip during the course of the measurement due relaxation effects. Standard signal averaging techniques are ineffective in handling signals of randomly fluctuating polarity. The issue of fluctuating signal polarity becomes even more acute when one considers the challenges of reconstructing multispin images containing fluctuating spins.

Task 6 addresses the signal processing issues for single-spin MRFM. Professors Jeff Fessler and Al Hero at the University of Michigan will work on signal and image processing problems associated with development of single spin MRFM. Initially, the work will be focussed on development of model-based algorithms for optimal single-electron spin and Larmor NMR detection from cantilever measurements. This will involve mathematical and statistical modeling of the cantilever-based measurement system, developing statistically optimal detection algorithms, and developing lower bounds on the minimum required SNR thresholds.

Issues related to MRFM image reconstruction will also be addressed, both on the macroscale (non-single-spin) and on the microscopic (single-spin) scale. Image reconstruction algorithms for detecting and localizing a small number of superimposed spins will be developed and analyzed.

The proposed work for the first two years is described below. The time lines for each of the tasks are only approximate and some tasks may be performed earlier or later than scheduled depending on progress.

Modeling (months 1-6)**:**

The University of Michigan team members will develop and validate a statistical model for the single-spin detection problem using measured input/output characteristics of the cantilever. Profs. Fessler and Hero will visit IBM in the first couple of months for face-to-face meetings with the other co-I's and to visit the facilities. Among the issues to be considered are: statistical models of cantilever response, spin relaxation, and laser metrology. At this meeting we will also try to characterize the experimental data which will be needed for model validation.

Bounds and thresholds (Months 6-12):

The statistical model will be used to investigate fundamental factors limiting optimal detection performance. This will be accomplished by developing decision-theoretic lower bounds on detection and localization errors and specification of algorithm-independent SNR thresholds. Instrument factors influencing these thresholds will be determined and compared to previous thresholds computed by Rugar and his collaborators for both linear phase coherent (heterodyne) and square law (energy) detectors. Non-linear detectors which improve upon energy detection will also be investigated.

Optimal detection algorithms (Months 12-18)

We will extend the single electron spin measurement models to the single Larmor frequency MRFM detection and revaluate fundamental limits on performance. On the basis of the models we will develop detection algorithms that come close to attaining statistically optimal performance. We will investigate maximum likelihood, Bayes optimal, and other methods for developing these detection algorithms. Algorithms performance will be analyzed using software modules developed by IBM and University of Washington for simulating the MRFM cantilever system. We will also determine the effect of parameter variations in the model on our bound predictions and on simulated detector performance.

Implementation (Months 18-24)

Optimal and suboptimal algorithms will be run on real data and any discrepancies between experimental results and benchmark (bound) predictions will be accounted for. The goal in this period will be to closely collaborate with IBM to experimentally demonstrate detection of single-spin and Larmor frequency using MRFM.

MRFM Imaging (Months 24-48 - [Contingent])

If we are sucessful in demonstrating that single spin detection is feasible, then microscale subsurface imaging algorithms which simultaneously detect, deconvolve, and localize small numbers of single-spin moments will be developed.

2.9 Previous accomplishments and CVs

Dr. Daniel Rugar

Dr. Rugar is manager of the Nanoscale Studies group at the IBM Almaden Research Center. His current interests include magnetic resonance force microscopy, ultrasensitive force detection, micro- and nanomechanics, and ultrahigh density data storage. Dr. Rugar performed the first experimental demonstration of MRFM[21] and has been a leader in the field ever since. As part of the MRFM effort, Dr. Rugar pioneered the field of ultrasensitive force detection using micromechanical cantilevers and has set a series of force detection records[5, 22, 13]. He has published over 100 papers and holds 13 patents. He has received IBM internal awards for work on magnetic force microscopy, magnetic resonance force microscopy, near-field optical data storage and ultrahigh density data storage based on the atomic force microscope.

Education

Stanford University	Applied Physics	Ph.D.	1982
Pomona College	Physics	B.A.	1975

Experience

Manager, Nanoscale Studies	IBM Research Division	1997 - present
Manager, Exploratory Storage Studies	IBM Research Division	1988 - 1997
Research Staff Member	IBM Research Division	1984 - 1988

Honors and Awards:

Distinguished Lecturer of IEEE Magnetic Society, 1999-2000
IBM Almaden Innovation Grant - award to initiate MRFM project.
IBM Outstanding Innovation Award - for fiberoptic interferometer invention

Selected publications:

1. "Mechanical Detection of Magnetic Resonance", D. Rugar, C.S. Yannoni and J. A. Sidles, Nature **360**, 563 (1992).

2. "First images from a magnetic resonance force microscope", O. Zueger and D. Rugar, Appl. Phys. Lett. **63**, 2496 (1993).

3. "Attonewton force detection using ultrathin silicon cantilevers", T.D. Stowe, K. Yasumura, T.W. Kenny, D. Botkin, K. Wago and D. Rugar, Appl. Phys. Lett. **71**, 288 (1997).

4. "Force-detected electron spin resonance: adiabatic inversion, nutation and spin echo", K. Wago, D. Botkin, O. Zueger, R. Kendrick, C.S. Yannoni and D. Rugar, Phys. Rev. B **57**, 1108 (1998).

5. "Thermal fluctuations in a magnetic tip: Implications for magnetic resonance force microscopy", J. D. Hannay, R.W. Chantrell and D. Rugar, J. Appl. Phys. **87**, 6827 (2000)

Prof. Thomas W. Kenny

Professor Kenny is an Associate Professor in the Mechanical Engineering department at Stanford University. Prof. Kenny develops micromachined sensors, actuators, and biofluidic devices for industrial and scientific applications. His group has expertise in fabricating complicated 3-dimensional structures with shapes optimized for various applications. Examples include development of attonewton sensing cantilevers for MRFM, ultrathin piezoresistive cantilevers, and multi-axis force sensors for biological applications. In this program, Kenny and his group will provide access to unique fabrication facilities at Stanford, and substantial experience with the design, fabrication and optimization of ultrasensitive cantilevers.

Education

University of California, Berkeley	Physics	Ph.D.	1989
University of Minnesota, Minneapolis	Physics	B.S.	1983

Experience

Associate Professor	Stanford University	2000 - present
Assistant Professor	Stanford University	1994 - 2000
Staff Scientist, Group Leader	Jet Propulsion Lab	1989 - 1994
Research Assistant	Univ. of California, Berkeley	1984 - 1989

Honors

NSF CAREER Award, 7/95
Terman Fellowship, 7/94
R+D 100 Award, 5/93
Distinguished Teaching Award, 6/89

Selected Publications:

1. J.A. Harley, and T.W. Kenny, "High Sensitivity Piezoresistive Cantilevers under 1000Å Thick", Appl. Phys. Lett. **75**, 289 (1999).

2. T.D. Stowe, K. Yasumura, T. Pfafman, T. Kenny, D. Botkin, and D. Rugar, "Attonewton Force Detection using Ultrathin Silicon Cantilevers", Appl. Phys. Lett. **71**, 288 (1997).

3. B.W. Chui, et al., "Low-stiffness Silicon Cantilevers with Integrated Heaters and Piezoresistive Sensors for High-Density AFM Thermomechanical Data Storage", Journal of MicroElectroMechanical Sys. **7**, 69 (1998)

4. B.W. Chui, T.W. Kenny, H.J. Mamin, B.D. Terris, and D. Rugar," Independent Detection of Vertical and Lateral Forces with a Sidewall-Implanted Dual-Axis Piezoresistive Cantilever", *Appl. Phys Lett.* **72**, 1388 (1998).

5. B. C. Stipe, H.J. Mamin, T.D.Stowe, T.W. Kenny and D. Rugar, "Magnetic dissipation and fluctuations in individual nanomagnets measured by ultrasensitive cantilever magnetometry", *Phys. Rev. Lett.* **86**, 2874 (2001).

Prof. John A. Sidles

Dr. Sidles is a full Professor at the University of Washington, School of Medicine, Department of Orthopædics. Dr. Sidles published the first MRFM article in 1991 [23], coauthored the first MRFM experiment article in 1992 [21], and with NIH funding has been active ever since in MRFM research. Dr. Sidles also maintains a very active program in clinical orthopædic research.

Education

University of Washington	Physics	Ph.D.	1982
University of Iowa	Physics	B.A.	1974

Employment History

Professor	U of W	1998 - present
Associate Professor	" . . . "	1992-1998
Assistant Professor	" . . . "	1986 - 1992
Acting Asst. Professor	" . . . "	1984 - 1986
Research Assistant	" . . . "	1984

Selected publications:

1. J. A. Sidles, "Folded Stern-Gerlach experiment as a means for detecting nuclear magnetic resonance in individual nuclei", *Physical Review Letters* **68**, 1124-7 (1992)

2. J. A. Sidles, J. L. Garbini and G. P. Drobny, "The theory of oscillator-coupled magnetic resonance with potential applications to molecular imaging", *Review of Scientific Instruments* **63**, 3881-99 (1992).

3. J. A. Sidles, J. L. Garbini, K. J. Bruland, D. Rugar, O. Züger, S. Hoen, and C. S. Yannoni, "Magnetic Resonance Force Microscopy", *Reviews of Modern Physics* **67**, 249-265 (1995).

4. J. A. Sidles and D. Rugar, "Signal-to-noise ratios in inductive and mechanical detection of magnetic resonance", *Physical Review Letters* **70**, 3506-9, 1993

5. K. J. Bruland, W. M. Dougherty, J. L. Garbini, S. H. Chao and J. A. Sidles, "Force-detected magnetic resonance in a field gradient of 250,000 tesla per meter", *Applied Physics Letters* 73, 3542-4 (1999).

Prof. Joseph L. Garbini

Dr. Garbini is a full Professor at the University of Washington, College of Engineering, Department of Mechanical Engineering. Dr. Garbini published one of the first MRFM theory articles in 1991 [24], and with NSF funding has been active ever since in MRFM research. His specialty is design and control.

Education

University of Washington	Mechanical Engineering	Ph.D.	1977
University of Washington	Mechanical Engineering	M.S.	1973
University of Washington	Mechanical Engineering	B.S.	1971

Experience

Professor	U of W	1991 - present
Associate Professor	" . . . "	1985 - 1991
Assistant Professor	" . . . "	1979 - 1985

Selected publications:

1. J. A. Sidles, J. L. Garbini and G. P. Drobny, "The theory of oscillator-coupled magnetic resonance with potential applications to molecular imaging", *Review of Scientific Instruments* **63**, 3881-99 (1992).

2. J. L. Garbini, K. J. Bruland, W. M. Dougherty and J. A. Sidles, "Optimal control of force microscope cantilever. I. controller design", *Journal of Applied Physics* **80**, 1951-8 (1996).

3. K. J. Bruland, J. L. Garbini, W. M. Dougherty and J. A. Sidles, "Optimal control of force microscope cantilever. II. magnetic coupling implementation", *Journal of Applied Physics* **80**, 1951-8 (1996).

4. K. J. Bruland, J. L. Garbini, W. M. Dougherty, S. H. Chao and J. A. Sidles, "Thermal tuning of a fiber-optic interferometer for maximum sensitivity", *Review of Scientific Instruments* **70**, 3542-4 (1999).

5. K. J. Bruland, W. M. Dougherty, J. L. Garbini, S. H. Chao and J. A. Sidles, "Force-detected magnetic resonance in a field gradient of 250,000 tesla per meter", *Applied Physics Letters* 73, 3542-4 (1999).

Alfred O. Hero

Professor, Electrical Engineering and Computer Science, Biomedical Engineering, and Statistics
The University of Michigan, Ann Arbor

EDUCATION

Boston University, Boston, Massachusetts, B.S.E.E., 1980, Electrical Engineering
Princeton University, Princeton, New Jersey, M.S., 1982, Electrical Engineering
Princeton University, Princeton, New Jersey, M.A., 1982, Electrical Engineering
Princeton University, Princeton, New Jersey, Ph.D., 1984, Electrical Engineering

RESEARCH AND PROFESSIONAL EXPERIENCE:

Sept. 1984-Aug. 1990, Assistant Professor of Electrical Engineering and Computer Science, University of Michigan, Ann Arbor, MI.

Aug. 1987, 1988, 1989, Visiting Scientist, M.I.T. Lincoln Laboratory, Lexington, MA.

Sept. 1990-present, Associate Professor of Electrical Engineering and Computer Science, University of Michigan, Ann Arbor, MI.

July 1991 - Jan. 1992, Research Scientist, Ecole Nationale Supérieure de Techniques Avancées (ENSTA).

July - Aug. 1993, William Clay Ford Fellow, Scientific Research Laboratory, Ford Motor Co.

Sept. 1996-present, Professor of Electrical Engineering and Computer Science, Biomedical Engineering, and Statistics, University of Michigan, Ann Arbor, MI

Jan. - Apr. 1999, Research Scientist, Centre Nationale de Recherche Scientifique (CNRS), Ecole Normale Supérieure, Lyon, France

June-Aug. 1999, Research Scientist, Ecole Nationale Supérieure de Télécommunications (ENST), Paris, France

May-June 2001, Visiting Professor, University of Nice, Sophia-Antipolis, France

FELLOWSHIPS AND HONORS (past 5 years):

1997	Fellow of IEEE
1998	Meritorious Service Award, IEEE Signal Processing Society
1998	Best Paper Award, IEEE Signal Processing Society
2000	3rd Millenium Medal, IEEE

PROFESSIONAL ACTIVITIES (past 5 years):

1996	Treasurer, Conference Board of IEEE Signal Processing Society
1996	Chair, Statistical Signal Processing and Arrays Technical Committee, IEEE Signal Proccessing Society
1996	Associate Editor, IEEE Transactions on Information Theory
1999	Chair, Commission C, Int'l Union of Radio Sciences (URSI)
2000	Vice President (Finance), IEEE Signal Processing Society

RELEVANT PUBLICATIONS:

"Kullback proximal algorithms for maximum likelihood estimation," S. Chretien and A. O. Hero, IEEE Trans. on Inform. Theory, Vol IT-46, No. 5, pp. 1800-1810, Aug. 2000.

"Tree structured non-linear signal modeling and prediction," O. Michel, A. O . Hero and A.-E. Badel, IEEE Trans. on Signal Processing, Vol. SP-47, No. 11, pp. 3027-3041, Nov. 1999.

"Minimax emission computed tomography using high-resolution anatomical side information and B-spline models," A. O. Hero, R. Piramuthu, J. A. Fessler and S. R. Titus, IEEE Trans. on Information Theory (Special issue on Multiscale Statistical Signal Analysis and its Applications), Vol 45, No. 3, pp. 920-938, April 1999.

"Exploring estimator bias-variance tradeoffs using the uniform CR bound," A.O. Hero, J.A. Fessler, and M. Usman, IEEE Trans. on Signal Processing, Vol. SP-44, No. 8, pp. 2026-2041, Aug. 1996. (Best Paper Award, IEEE Signal Processing Society})

"Penalized maximum likelihood image reconstruction using space alternating generalized EM algorithms," J.A. Fessler and A.O. Hero, IEEE Trans. on Image Processing, Vol 4, No. 10, pp. 1417-1429, Oct 1995.

Dr. James Tropp

Dr. Tropp is an expert in nuclear magnetic resonance with achievements in theory, experiment and hardware development. He has led research teams in both industrial and academic settings. NMR experience includes magnetic resonance imaging, localized spectroscopy *in vivo,* and high resolution spectroscopy of liquids and solids. For this MOSAIC proposal, Dr. Tropp will study spin diffusion processes in the context of MRFM and molecular imaging (Task 5).

Education

University of Chicago	Biochemistry	Ph.D.	1976
New York University	English	B.A.	1970

Experience

Scientist	General Electric Medical Systems	1991 - present
Scientist	Toshiba America MRI	1982 - 1991
Engineer	GE NMR Instruments (formerly Nicolet Magnetics)	1982 - 1985
Post-doc	MIT, Dept. of Chemistry	1980 - 1982
Post-doc	Brandeis University, Dept. of Biochemistry	1976 - 1980

Selected publications:

1. “Spatial Dependence of a Differential Shading Artifact in Images from Coil Arrays with Reactive Cross-talk at 1.5 T”, J. Tropp and T. Schirmer, J. Magn. Reson. **151**, 146 (2001)

2. “Very Selective Suppression Pulses for Clinical MRSI Studies of Brain and Prostate Cancer”, T-K. C. Tran, D. B. Vigneron, N. Sailasuta, J. Tropp, P. Le Roux, J. Kurhanewics, S. Nelson, and R. Hurd, Magn. Reson. in Med. **43**, 23 (2000).

3. “Mutual Inductance in the Bird Cage Resonator”, J. Tropp, J. Magn. Reson. **126**, 9 (1997).

4. “Dipolar Cross Correlation and the Nuclear Overhauser Effect in Systems with Strong Scalar Coupling”, J. Tropp, J. Magn. Reson. A **103**, 90 (1993)

5. “The Theory of an Arbitrarily Perturbed Bird-Cage Resonator, and a Simple Method for Restoring It to Full Symmetry”, J. Tropp J. Magn. Reson. **95**, 235 (1991).

2.10 Facilities

IBM Almaden Research Center

IBM has MRFM apparatuses operational at 4K and 100 millikelvin and two other cantilever testing setups operable at temperatures between 300K and 1.5K. All of the setups are equipped with fiberoptic interferometry and have mechanics for mounting and aligning micromachined cantilevers.

The millikelvin apparatus is based on a customized Oxford Kelvinox 400 dilution refrigerator. The refrigerator has a vacuum chamber attached to the mixing chamber in order to accomodate the MRFM apparatus. Other special features include an oversize 1K pot that allows extended one-shot operation to minimize vibration noise, an oil-free helium circulation system that uses a magnetically levitated turbopump to minimize vibration and an 8 Tesla superconducting magnet. The vacuum chamber housing the MRFM experiment is served by three microwave-capable coaxes (one of which is superconducting) and 24 high voltage lines for controlling piezoelectric positioners.

A full set of electronics for MRFM experiments is available, including extensive microwave and rf instrumentation, multichannel pulse generators, spectrum analyzers, lock-in amplifiers, high voltage amplifiers for piezoelectric positioning, and data collection electronics. Additional custom-designed electronics will be incorporated, as necessary.

To characterize magnetic resonance properties of samples used in MRFM experiments, a variable temperature X-band cw EPR spectrometer and several pulsed NMR spectrometers are available.

For fabrication of magnetic and superconducting components, IBM has extensive facilities for thin film deposition, including numerous sputtering and evaporation systems. Microstructure and nanostructure fabrication facilities are available, including advanced optical lithography, reactive ion etching and ion milling. A state-of-the-art electron beam lithography facility centered around a Leica Nanowriter is planned for installation during the first quarter of 2002. A focused ion beam (FEI 830XL dual-beam instrument) is also available for fabricating custom nanostructures, including magnetic tips based on bulk rare-earth magnets. A micromanipulator system combined with a high numerical aperture, long working distance microscope allows precision mounting of magnetic tips or samples onto cantilevers. Custom-designed jigs have been built for use in conjunction with the micromanipulator in order to facilitate the manipulation and mounting of small particles on cantilevers.

Stanford University

The Center for Integrated Systems (CIS) houses a complete state-of-the-art silicon and GaAs nano-fabrication facility. This facility is part of the new NSF National Nanofabrication Users Network and its capabilities are believed to be among the best in U.S. universities. The clean room facilities are housed in a 10,000 square foot, class 100 laboratory. Particularly notable is the Hitachi E-beam writer which can achieve 10 Å resolution for direct pattern writing, new Lam Research and STS RIE systems and a Digital Instruments AFM. In addition, the facility includes E-beam mask making, wafer steppers for photolithography, LPCVD systems for poly, oxide, nitride, and tungsten, dry etching systems for all thin film materials used in device fabrication, sputtering systems for metals, dielectrics and silicides, six MBE systems for heterojunction and superlattice structures in Si and the III-V alloys. The entire facility is open to any trained student and is computer automated to make certain that only authorized users operate specific pieces of equipment. Testing facilities adjacent to the laboratory permit measurement of process parameters, both optical and electronic device characteristics, and functional circuit performance using computer controlled systems. A dedicated microwave test setup includes a HP 8510 Vector Network Analyzer and Cascade-Microtech microwave probe station for S-parameter measurements of microwave devices. A full time staff of 6 process and 6 maintenance technicians maintain the lab and characterize new equipment. Students are allowed full access to the facility after completing a series of lecture and laboratory courses. The replacement value of the laboratory equipment alone is approximately $50 million.

2.11 Current and pending proposals

IBM - D. Rugar, PI

Current support

ONR Contract No. N00014-98-C-0070, “Toward Single Spin MRFM”, 04/98 - 09/02, $755,066 total. Supports development of techniques for single-spin MRFM.

University of Washington - J. Sidles and/or J. Garbini, PIs

Current support

NIH #R01-RR08820-09, “Imaging of Molecules by Oscillator Coupled Resonance”, 08/01 - 7/06, $957,802 total. A MRFM theory-only grant. (J.S.)

NSF #0097544, “Direct 3D imaging of molecular structure: Quantum sensing and control”, 06/01 - 505/04, $399,071 total. Fundamental research in MRFM-related quantum sensing and control. (J.G)

U.S. Army (DARPA/DSO) #DAA19-01-1-0679, “The accelerated development of MRFM”, 7/01 - 6/03, $439,859 total. A two-year project to accelerate the pace of MRFM development. (J.G. & J.S.)

Pending support

DARPA/DSO (BAA01-39: MOSAIC), “Achieving molecular observation in four years”, 4/02 - 3/06, $3,355,853 total. A four-year program to achieve a comprehensive Defense molecular observation capability. (J.G. & J.S.)

Stanford University - T. Kenny, PI

Current support

NSF GOALI, “Ultrasensitive Cantilevers for MRFM”, 8/99 - 7/02, $60K/year. This is a collaboration with the Rugar-IBM group focused on developing and characterizing ultrathin cantilevers for detection of attonewton forces, and for gaining an understanding of how materials, geometry, surfaces, and other controllable parameters affect the ability to measure small forces.

NSF XYZ, “MEMS Devices for Experiments in Biology”, 11/99 - 10/02, $175K/year. This grant supports the development of MEMS force sensors for measurements of interesting biological forces. Examples include measurements of molecular adhesion forces between cadherin molecules (with James Nelson of Stanford’s Cell Biology Department), measurements of the adhesive forces of hairs from the feet of Geckos (With Bob Full of the UCB Integrative Biology Department), and measurements of Actin-Myosin interactions (With James Spudich of the Stanford Molecular Biology Department).

DARPA HERETIC, “Electrokinetic Closed-Loop Cooling for Integrated Circuits”, 6/99 - 5/02, $350K/year. This project is a collaboration with Professors Goodson and Santiago at Stanford to develop pumps, heat exchangers, and thermal modeling in support of a new fluidic chip-cooling technology. This project has collaborators at Intel, and is working towards a capability for removal of 200W from an integrated circuit.

NSF NANO, “Cantilevers Integrated with Nanostructures for MFM” (w/ Prof. K. Moler), 7/01 - 6/04, $50K/year. This is a collaboration with Prof. Moler and researchers at IBM to develop high-resolution magnetic force microscope probes based on the integration of cantilevers, nanomagnets, and other sensing approaches.

NSF Civil, “Damage Detection in Civil Structures” (w/ Prof. Law), 10/00 - 11/03, $30K/year. This project is focused on the development of miniature sensors suitable for integration within buildings and other civil structures for detection of changes that indicate minor or major damage during events such as earthquakes.

NSF Civil-II, “ Wireless Monitoring of Civil Structures” (w/ Prof. Kiremidjian), 6/04, $50K/year. This project extends the research on sensors for civil structures to include wireless stations, integrated signal processing and data reduction, and other technologies as needed to allow long-term wireless monitoring.

Los Alamos National Labs/DOE, “Accelerometers for Vibration Monitoring”, 11/00 - 10/01, $70K/year. Sensors for Damage Prognosis. In this project, sensors are being provided for integration within a Composite Wing on a ultralight autonomous aircraft to detect damage that might lead to failure.

Pending support

DARPA Nano-Oscillators,“ Ultra-High-Q Oscillators”, 11/01 - 10/05, $600K/year. This project seeks fabrication and encapsulation methods for GHz resonators that feature high Q and high stability.

DARPA Mixed-signal integration, “Chip-Integrated Microchannel Cooling for Mixed-Signal ICs”, (w/ Prof. Goodson), 11/01 - 10/05, $500K/year. This project seeks methods for integration of CMOS, RF, MEMS, and cooling into multi-layer ICs for future applications.

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