Response to National Science Foundation
Program Solicitation 00-106:
Initiative on Sensing and Imaging Technologies

Direct 3D Imaging of Molecular Structure: Quantum Sensing and Control

Principal Investigator: Joseph L. Garbini, Ph.D.
Professor, Department of Mechanical Engineering
College of Engineering, Box 352600
University of Washington, Seattle WA 98195
email: garbini@u.washington.edu
phone: 206.543.5399

Co-Investigator: John A. Sidles, Ph.D.
Professor, Department of Orthopædics
School of Medicine, Box 356500
University of Washington, Seattle WA 98195
email: sidles@u.washington.edu
phone: 206.543.3690

The proposal is freely available on-line at the URL:

`ftp://ftp.u.washington.edu`
`/public/sidles/QUMOD.dir/QUMOD_NSF.pdf`

It is provided as a service to anyone writing a proposal relating to quantum molecular observation. A CD containing an MRFM BibTeX database, graphical material, and TeX source files is available upon request. We particularly welcome inquiries relating to collaborative research and MRFM pilot projects.

A Summary of Objectives

One of the oldest and most enduring dreams of the scientific community is to directly observe molecular structure nondestructively, *in situ*, in three dimensions, with Angstrom-scale resolution. Such an imaging technology would immediately address urgent needs in nanoscale engineering, materials science, molecular biology, and medicine.

The objective of the proposed research is to create such a technology.

A.1 Methods to Be Employed

The proposed method is magnetic resonance force microscopy (MRFM), which was conceived in 1991, by the proposers [53, 54, 57], specifically as a means for 3D molecular imaging. The central concept of MRFM is to combine three-dimensional magnetic resonance imaging with angstrom-scale probe microscopy [56]. MRFM was first experimentally demonstrated in 1992 in collaboration with Dan Rugar's IBM group [48]. Subsequently, MRFM has developed into a worldwide sensing and imaging research effort (see Sections C.2 and C.7).

A well-defined and technically feasible path to 3D molecular observation by MRFM has now emerged from NSF- and NIH-funded research by the proposers [13, 15, 20, 23, 48, 53–58] and by other MRFM groups [24,27,48,49,66,69,74–78]. The critical path combines elements of cryogenic technology, nanoscale technology, and quantum signal processing (see Section C.3).

The proposers' MRFM research is focused upon two critical path goals: first, establishing reliable, experimentally validated design principles for single-spin quantum control and imaging (this NSF proposal); and second, establishing firm theoretical foundations for controlling spin decoherence in the MRFM environment (under NIH support) (Sections C.4 and C.8).

Achieving these two goals will allow a feasible path to practical molecular imaging to be specified in detail and with confidence. The logical next step will be to launch a coordinated national research initiative for quantum molecular observation (Section C.5).

A.2 Specific Aims

The specific aims of this NSF proposal are to:

- demonstrate nanoscale resolution in 3D MRFM imaging;
- achieve a reliable, experimentally validated understanding of electron and proton spin relaxation in the MRFM environment;
- extend present design principles for optimal control and estimation to the quantum environment appropriate to single spin imaging.

The proposed means are to:

- validate and calibrate the proposers' newly completed 3D MRFM scanner, via force, parametric, and multiplex imaging experiments;
- design and operate a next-generation adaptive digital controller, incorporating optimal control, estimation, and diagnostic algorithms;
- survey electron and proton spin relaxation in a variety of target samples.

A.3 Broader Impacts

Practical quantum molecular observation will mark the coming of age of a new engineering discipline, *quantum engineering*. The proposed opening of a new imaging window onto the largely unobserved world of 3D molecular structure will be among the first "grand challenge" achievements of quantum engineering, and will revolutionize the fields of nanoscale engineering, materials science, biology, medicine, and engineering education.

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C Project Description

Directly visualizing molecular structure is one of the most important and enduring dreams of the scientific community. As Feynman foresaw in 1959 [22]:

> It is very easy to answer many of these fundamental biological questions; you just look at the thing! You will see the order of bases in the chain; you will see the structure of the microsome ... I put this out as a challenge: is there no way to make the electron microscope more powerful?

Only in the past decade have scientists appreciated that Feynman's challenge is unlikely to be met using any form of energetic quanta. As reviewed by Henderson in 1995 [26]:

> Radiation damage ... prevents the determination of the structure of a single biological macromolecule at atomic resolution using any kind of microscopy. This is true whether neutrons, electrons, or x-rays are used as the illumination.

Other studies report similar findings [28, 29, 42, 56].[1] In short, there is at present no known way to directly observe molecular structure by energetic particle imaging.

C.1 Quantum Molecular Observation

How can Henderson's limits be surmounted? During the 1970s and 80s, two new technologies, magnetic resonance imaging [34, 38] and scanning probe microscopy [7–9], proved that high spatial resolution can be achieved using low-energy quanta that cause no radiation damage. Another revolutionary idea emerged in the 1980s and 90s: quantum observation technologies, like single-ion clocks [10, 17, 30] and quantum computers [21, 31, 44, 50], which obtain valuable information from the direct observation of isolated quantum systems.

These new sensing and imaging technologies are not subject to Henderson's limits. A central theme of this proposal is that when these technologies—magnetic resonance imaging, probe microscopy, and quantum observation—are applied in combination, they constitute a powerful new *quantum molecular observation* technology for fulfilling Feynman's challenge.

Our overall goal is to make quantum molecular observation a practical reality.

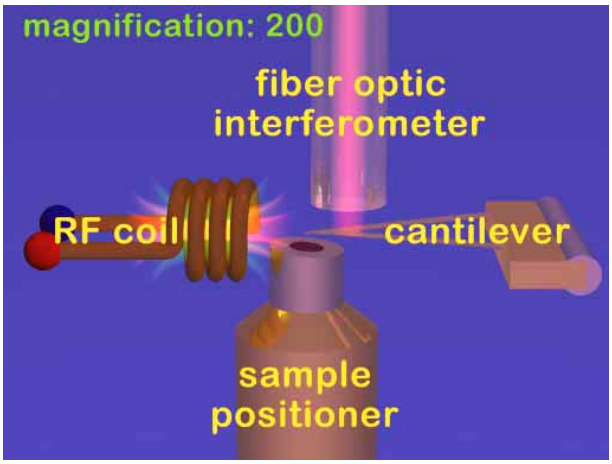
C.2 Overview of MRFM

Magnetic resonance force microscopy (MRFM) was conceived from the outset [53] as a quantum observation technology [54] that would combine magnetic resonance imaging and scanning probe microscopy [57] to create a technology for achieving [56]:

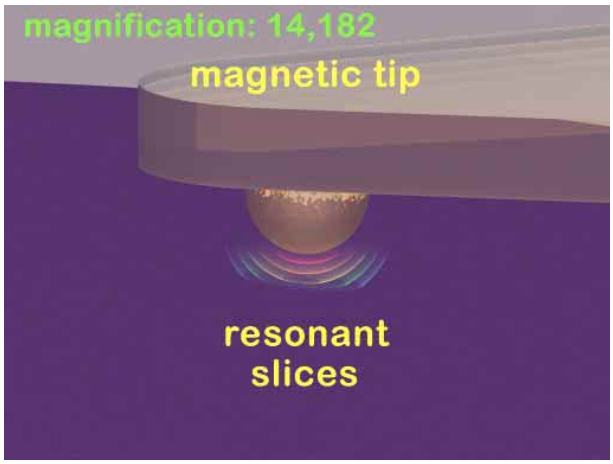
- the direct observation of individual molecules,
- *in situ*, in their native forms and native environments,
- with three-dimensional atomic-scale resolution,
- by a nondestructive observation process.

Such a technology would function as a true *quantum molecular microscope*, allowing researchers to observe atomic-scale structure and environments in nanoelectronic devices, materials, and biological tissues as readily as present-day optical microscopes observe the structure and behavior of living cells.

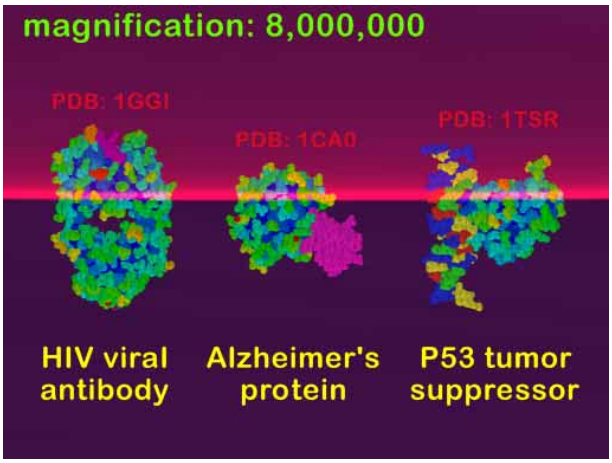
[1] For example, from computer simulations, Neutze *et al.* [42] derive a resolution limit of ~ 15 Angstroms for x-ray laser imaging of individual protein molecules. They assume an optimized x-ray pulse with duration 1 fs, focal spot of 100 nm, and intensity of 8×10^{22} W/cm^2 (!). Each pulse results in the complete ionization of a single target molecule.



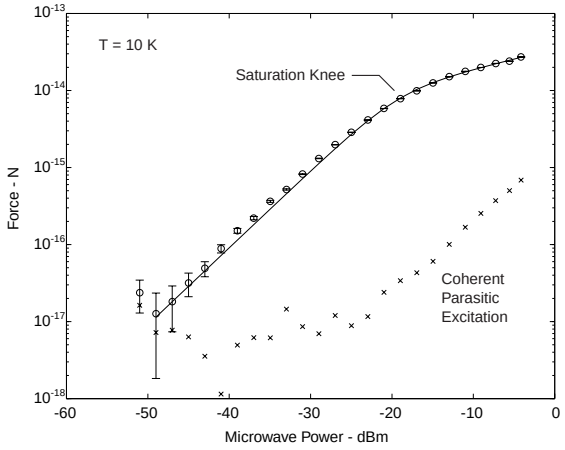
A cantilever with a magnetic tip (center) approaches a sample positioner (entering from below). A radio-frequency coil (left) modulates the sample spins, and a fiber-optic interferometer (top) detects the resulting cantilever motion.



The tip-sample region, with resonant slices. The tip is 2.9 μm in diameter. Slices closer to the tip have higher resonant frequency. Note that the magnetic slices reach into the sample, allowing nondestructive subsurface 3D imaging.



Close-up of a resonant slice. Protein data bank entries 1GGI, 1CAO, and 1TSR are shown only for scale; the sample that generated the data at right was a 5% solution of 1,1-diphenyl-2-picrylhydrazil (DPPH) in polystyrene [18].



Electron spin resonance data acquired at 10 K [18]. The Bloch equations predict a resonant slice thickness of $\sim$ 13 nm in this experiment. Note the large SNR ratio and low noise floor of < 10 aN that arise from this thin, nanometer-scale resonant slice.

Figure 1: Our present MRFM apparatus, as described in [18].

With reference to Fig. 1 (above),[2] MRFM signals arise when spins in a sample are modulated by an applied radio-frequency field, precisely as in conventional magnetic resonance imaging. Only spins within a thin resonant slice are affected; the spins closer to the tip are in too strong a field for resonance, while spins farther from the tip are in too weak a field. The thickness of the slice is described by Bloch-type equations [18]; the stronger the gradient of the magnetic field, the thinner the resonant slice.

[2] The Fig. 1 pictures are from a 3D animation of MRFM imaging which can be accessed by web browser at the URL: `ftp://ftp.u.washington.edu/public/sidles/QUMOD.dir/QUMOD.mov`

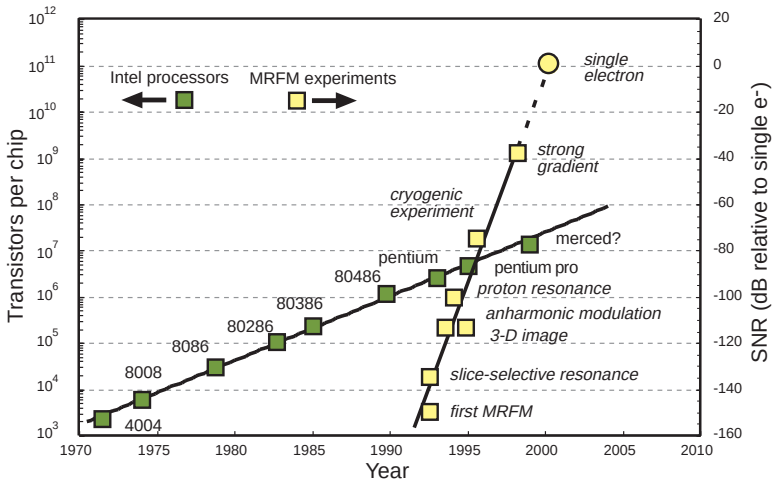


Figure 2: Published MRFM sensitivity contrasted with transistors per integrated circuit. See Table 1 (p. C.8) for the MRFM device parameters; Intel transistor counts are from [1].

The spins within a resonant slice are detected by force microscopy, by virtue of the magnetic force between the spins and the nearby tip, which excites the cantilever into motion which is detected by the interferometer. As in conventional magnetic resonance, there are many different ways to modulate this tip-sample interaction: cyclic saturation, adiabatic inversion, spin echos, and pulse inversion have all been demonstrated in the context of MRFM, for both nuclear and electron spins.

MRFM has two main limitations. First, it is most effective at cryogenic temperatures, such that all molecular motion ceases. Thus, the dynamical behavior of observed molecules will have to be inferred from frozen structural "snapshots", augmented by dynamical computer simulations. Second, quantum observation requires nuclei with nonzero spin. In some cases this will require isotopic labeling, *e.g.* the ^{13}C labeling assumed in [57].

Since 1992, MRFM sensitivity has doubled every six months on average (see Fig. 2 and Table 1) . This exponential doubling is known as "Moore's Law". How does Moore's Law coexist with the equally well known "Murphy's Law"—the law that what can go wrong *will* go wrong, particularly when developing a new technology? In Moore's words [40]:

> The reason we have a violation of Murphy's Law is that we're exploiting the technology. By making things smaller, everything gets better simultaneously.

MRFM devices enjoy the same favorable "Moore Scaling" as semiconductor devices: making MRFM devices smaller makes them work better. Murphy's Law still applies; there are always plenty of technical challenges in each new device generation. But because the physical scaling is favorable, solutions to these challenges have—to date—always been found.

C.3 MRFM Design Principles

The design principles of MRFM have been developed in many articles by the proposers' group [11–15, 18–20, 23, 51–58] and other by other MRFM research groups [2–6, 16, 24, 25, 27, 35–37, 39, 41, 45–49, 59–78]; The most complete single reference is our [56], and the most complete discussions of spin decoherence and quantum observation—which are emerging as fundamental research issues in MRFM—are in [51, 52, 54, 57].

Up until the present year, the MRFM community has mainly focused on design principles relating to device sensitivity. These principles are conveniently summarized by expressing thermal noise (the limiting noise in all MRFM experiments to date) as equivalent to a fluctuating spin magnetic moment $\mu(t)$ with spectral density S_μ. Improving MRFM spin sensitivity is then equivalent to making S_μ as small as possible. According to [56, 58], S_μ is given by:

$$S_\mu = \frac{m}{g^2\tau}\, 2k_{\rm B}T, \quad \text{where} \begin{cases} m, & \text{cantilever motional mass;} \\ g, & \text{tip magnetic field gradient;} \\ \tau, & \text{cantilever damping time;} \\ k_{\rm B}T, & \text{thermal energy.} \end{cases} \tag{1}$$

From this we read off four main design options for increasing MRFM sensitivity:

- reduce the temperature T
- reduce the mass m
- increase the magnetic gradient g
- increase the damping time τ

These design principles work extremely well in practice. As illustrated in Fig. 2 and as summarized in Table 1, MRFM signal-to-noise ratios have increased by 112 dB since 1992. The design path has been precisely as specified above: lower temperature, decreased mass, increased gradient, and increased damping time.[3]

Equation 1 suggests the following "quick" route to practical molecular observation. Relative to our present device [18] (see Fig. 1 and the final entry in Table 1):

- reduce the size of the cantilever from 300 μm to 3 μm, and thereby gain 60 dB of SNR by virtue of reduced cantilever mass;
- reduce the radius of the tip to 300 Å, and thereby gain 66 dB of SNR by virtue of the increased magnetic gradient;
- operate the device in a dilution refrigerator at 30 mK, and thereby gain 25 dB of SNR by virtue of the reduced temperature.

The resulting device would operate with $T = 30$ mK, $m = 6.3$ fg, $\tau = 1$ s, and $g = 84$ G/Å.[4]

This device would achieve impressive performance in observing individual proton moments: 54 dB of SNR in 1 Hz of bandwidth, or equivalently, 10 dB of SNR in a bandwith of 10 kHz. With appropriate multiplexing, the device could observe many thousands of atomic coordinates per second. *It would therefore function as a true molecular microscope, and bring to fruition the enduring dream of directly observing molecular structure.*

Although such a molecular observation technology would be sophisticated, the devices themselves would be neither large nor particularly expensive. The heart of each device would occupy less than one cubic millimeter, and an entire instrument would be table-top scale, such that individual researchers could in principle afford them.

[3] Reviewers should appreciate that each 6 dB of SNR gain represents a doubling of sensitivity of MRFM technology. Thus, the 112 dB of increased SNR from the first MRFM experiment to the present state-of-the-art represents eighteen sensitivity doublings. We are not aware of any other imaging technology in recent years that has demonstrated similar exponential gains.

[4] This device closely resembles the device we described in 1992 [57]; this article extensively discusses design issues, and contains the only published quantum simulation of a molecular scan.

Article	Device Parameters	Sensitivity
July 1992a: D. Rugar, C. S. Yannoni, and J. A. Sidles. Mechanical detection of magnetic resonance. *Nature*, 360(6404):563–6, 1992. First detected MRFM signal.	$T = 300$ K $m = 39$ ng $\tau = 0.04$ sec $g = 10^{-5}$ G/Å	$S_f^{1/2} = 2.9 \times 10^{-15}\,\mathrm{N}/\sqrt{\mathrm{Hz}}$ $S_\mu^{1/2} = 3.1 \times 10^{7}\,\mu_\mathrm{B}/\sqrt{\mathrm{Hz}}$ SNR $= -149$ dB
July 1992b: Same article as above, but with a stronger gradient applied to the sample.	$T = 300$ K $m = 39$ ng $\tau = 0.04$ sec $g = 6.0 \times 10^{-5}$ G/Å	$S_f^{1/2} = 2.9 \times 10^{-15}\,\mathrm{N}/\sqrt{\mathrm{Hz}}$ $S_\mu^{1/2} = 5.2 \times 10^{6}\,\mu_\mathrm{B}/\sqrt{\mathrm{Hz}}$ SNR $= -134$ dB
July 1993: O. Züger and D. Rugar. First images from a magnetic resonance force microscope. *Appl. Phys. Lett.*, 63(18):2496–8, 1993. First Fourier transform imaging techniques.	$T = 300$ K $m = 13$ ng $\tau = 0.03$ sec $g = 4.2 \times 10^{-4}$ G/Å	$S_f^{1/2} = 1.8 \times 10^{-15}\,\mathrm{N}/\sqrt{\mathrm{Hz}}$ $S_\mu^{1/2} = 4.6 \times 10^{5}\,\mu_\mathrm{B}/\sqrt{\mathrm{Hz}}$ SNR $= -113$ dB
November 1994: K. J. Bruland, J. Krzystek, J. L. Garbini, and J. A. Sidles. Anharmonic modulation for noise reduction in magnetic resonance force microscopy. *Rev. Sci. Intrum.*, 66(4):2853–6, 1995. First UW experiment.	$T = 300$ K $m = 17$ ng $\tau = 0.05$ sec $g = 3.8 \times 10^{-4}$ G/Å	$S_f^{1/2} = 1.6 \times 10^{-15}\,\mathrm{N}/\sqrt{\mathrm{Hz}}$ $S_\mu^{1/2} = 4.6 \times 10^{5}\,\mu_\mathrm{B}/\sqrt{\mathrm{Hz}}$ SNR $= -113$ dB
January 1994: D. Rugar, O. Züger, S. Hoen, C. S. Yannoni, H. M. Vieth, and R. D. Kendrick. Force detection of nuclear magnetic resonance. *Science*, 264(5165):1560–3, 1994. First nuclear-spin MRFM.	$T = 300$ K $m = 13$ ng $\tau = 0.34$ sec $g = 6 \times 10^{-4}$ G/Å	$S_f^{1/2} = 5.6 \times 10^{-16}\,\mathrm{N}/\sqrt{\mathrm{Hz}}$ $S_\mu^{1/2} = 1.0 \times 10^{5}\,\mu_\mathrm{B}/\sqrt{\mathrm{Hz}}$ SNR $= -100$ dB
May 1995: K. Wago, O. Züger, R. Kendrick, C. S. Yannoni, and D. Rugar. Low-temperature magnetic resonance force detection. *J. Va. Sci. Tech. B*, 14(2):1197–201, 1996. First cryogenic MRFM experiment.	$T = 6$ K $m = 18$ ng $\tau = 3.2$ sec $g = 6 \times 10^{-4}$ G/Å	$S_f^{1/2} = 3.068\,10^{-17}\,\mathrm{N}/\sqrt{\mathrm{Hz}}$ $S_\mu^{1/2} = 5515\ \mu_\mathrm{B}/\sqrt{\mathrm{Hz}}$ SNR $= -74.8$dB
March 1998: 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. *Appl. Phys. Lett.*, 73:1959–1964, 1998. The strongest-gradient MRFM experiment to date.	$T = 77$ K $m = 6.3$ ng $\tau = 0.45$ sec $g = 0.25$ G/Å	$S_f^{1/2} = 1.7\,10^{-16}\,\mathrm{N}/\sqrt{\mathrm{Hz}}$ $S_\mu^{1/2} = 75\,\mu_\mathrm{B}/\sqrt{\mathrm{Hz}}$ SNR $= -37$ dB
June 2000: W. M. Dougherty, K. J. Bruland, S. H. Chao, J. L. Garbini, S. E. Jensen, and J. A. Sidles. The Bloch equations in high-gradient magnetic resonance force microscopy: theory and experiment. *J. Mag. Res.*, 143:106–119, 2000. Emphasis shifting from sensitivity to control and decoherence.	$T = 10$ K $m = 6.3$ ng $\tau = 1.0$ sec $g = 0.044$ G/Å	$S_f^{1/2} = 8.2\,10^{-17}\,\mathrm{N}/\sqrt{\mathrm{Hz}}$ $S_\mu^{1/2} = 203\,\mu_\mathrm{B}/\sqrt{\mathrm{Hz}}$ SNR $= -46$ dB

Table 1: Improvements in MRFM sensitivity with time. Experimental parameters and device sensitivities are tabulated for eight MRFM experiments in the period 1992-2000. Here $\mu_\mathrm{B} = \hbar\gamma_\mathrm{e}/2$ is the magnetic moment of a single electron, *i.e.*, one Bohr magneton. The signal-to-noise (SNR) values in this table are those plotted in Fig. 2, as computed for the detection of one μ_B in one Hertz of bandwidth.

With the benefit of eight years of MRFM experience, we can identify two main obstacles on this "quick" path. The first challenge relates to fabricating cryogenic MEMS hardware: the required state-of-the-art cryogenic, microfabrication, optical, and micromagnetic engineering is beyond the scale (and budget) of small university research groups like ours. This is why our long-term goal is to launch a national research initiative for quantum molecular observation (see Section C.5).

The second challenge is more subtle and relates to the emerging discipline of quantum engineering. Even if the above-specified MEMS hardware were freely available, and achieved the stated noise level, *no one would know how best to use this hardware to image molecular structure.* As discussed in the following section, the engineering challenges involved are linked to quantum phenomena: spin decoherence, quantum measurement back-action, the Casimir effect, and the Stern-Gerlach effect. It is these *quantum engineering challenges* that we propose to address with NSF support.

C.4 Objectives for the Proposed Work

The specific objectives of the proposed research are:

- **Spin Relaxation:** Spin decoherence impairs the detection of polarized spins, and in our recent experiments has emerged as a critical path issue for molecular observation. In MRFM, short spin relaxation times may arise from a variety of sources; some are intrinsic to the sample, others are associated with molecules adsorbed onto the surface of the sample, and still others are related to properties of the magnetic tip, as reviewed in [18, 52]. A specific aim of this project is to achieve a reliable, experimentally validated understanding of electron and proton spin relaxation in the MRFM environment.
- **3D MRFM Imaging:** Three-dimensional imaging with sub-Angstrom resolution is the essential activity by which molecular structure will be determined. The MRFM environment necessarily includes three factors that complicate accurate imaging: high vacuum, cryogenic temperatures, and a close approach between the magnetic tip and the sample, which creates Casimir forces between the tip and the sample. In addition, noise processes including thermal noise acting on the cantilever, shot noise within the interferometer detector, and back-action noise caused by photons impinging on the cantilever must all be managed [56]. Therefore, the second project goal is to demonstrate 3D imaging with nanoscale resolution in the unique MRFM environment.
- **Quantum Observation and Control:** Control and estimation of the cantilever dynamic motion are key elements of MRFM operation. As we approach the goal of single-spin detection, classical models must be abandoned, and quantum phenomena such as the Stern-Gerlach effect will appear [53, 54]. Both the motion of the spin-cantilever system, and the interferometric process by which that motion is observed are subject to rigorous quantum mechanical treatment [51, 57]. The third objective of this investigation is to extend present classical design principles for optimal estimation and control to the quantum environment appropriate to single spin detection and imaging.

A thread which unites these objectives—aside from their relevance to molecular observation—is that each explores the boundary between classical and quantum engineering.

C.5 Relation to Longer-Term Goals

Our long-term goal is to ensure that quantum molecular observation becomes a practical reality in the years 2005–2010. In service of this goal, the deliverable of this proposal—considered as a whole—will be a technical path to a practical quantum molecular imaging technology, specified in detail and with confidence in its feasibility.

The logical next step will be to launch a coordinated national research initiative for quantum molecular observation. In a recent white paper[5] we have proposed that DoD fund a series of three Quantum Molecular Observation Workshops, to take place in the years 2001-2003. In coming months we will also seek NIH and NSF support for these workshops.

C.6 Significance

A desktop-scale quantum molecular imaging technology, as described in Section C.3 and in [56, 57], would broadly impact almost every area of science and engineering.

In materials science, researchers could directly observe phenomena like early fracture initiation. In nanoscale technologies, researchers would directly observe the structures they were fabricating. And in molecular biology, there would be less emphasis on experiments, and more emphasis upon direct observation, similar to present-day disciplines like astronomy and field ecology. A Human Proteome project, analogous to the Human Genome Project, could immediately be launched, with the objective of observing, cataloging and annotating every human protein.

Uniquely, direct molecular observation would allow every protein species to be observed *in situ*, so that all its structural variants, habitats, and interactions could be studied systematically. This capability would accelerate every branch of medical research.

C.7 Results from Prior NSF and NIH Support

Our MRFM research has been supported by the NSF and NIH since 1992, beginning with an 1992 one-year NIH Small Grant for Innovative Research. We are grateful.

Many of our most important NSF- and NIH-supported research results were reviewed in previous sections. The MRFM design rules were first established by our research (Section C.3), we have experimentally demonstrated that applying these rules leads to exponential improvements in MRFM sensitivity (as illustrated Fig. 2 and summarized in detail in Table 1), and we are presently at the forefront of addressing the quantum engineering challenges that are emerging as central issues in MRFM (Section C.4).

Table 2 (following page) shows the thirteen MRFM groups presently known to us, ten in the U.S. and three international. These groups represent a substantial leverage of the initial NSF and NIH investment in MRFM technology.

These successes, and our UW publications [11–15, 18–20, 23, 48, 51–58, 68] represent substantial progress toward a goal that we set forth in our first (1991) MRFM article [53]:

> The techniques described herein might eventually be extended to allow the imaging of biological molecules, and in fact were devised with this goal in mind. ...
>
> It is therefore clear that developing a practical molecular imager would require a substantial effort by many scientists, and that there would be no absolute assurance of success. Nonetheless, present and projected medical needs might justify such an effort.

[5] This white paper can be accessed at the URL:
`ftp://ftp.u.washington.edu/public/sidles/QUMOD.dir/QUMOD.pdf`

> Of the proteins encoded by the AIDS genome, only HIV-l protease has a known three-dimensional structure. Recently, a partial structure for HIV-l reverse transcriptase has also been obtained. The remaining proteins have so far proven refractory to x-ray crystallography. The missing structural information is a significant obstacle to the rational design of drugs and vaccines.
>
> As a faculty member in a school of medicine, the author frequently observes the sequelæ to our present lack of knowledge. This letter is offered in the hope that it may eventually contribute to better treatments for intractable disorders.

To the extent that diseases like HIV-AIDS, malaria, and tuberculosis are still global scourges today, nine years later, the original MRFM goal is still unrealized. Yet to the extent that quantum molecular observation is increasingly regarded as technically feasible, and to the extent that the envisioned "substantial effort by many scientists" is being mobilized (Section C.5), there are reasonable grounds for hope that the original MRFM goal of quantum molecular observation *will* be achieved.

C.8 Proposed Research Plan

Our overall research strategy regarding MRFM development is summarized in the following passage from our 1995 *Reviews of Modern Physics* article [56]:

> Present research in MRFM emphasizes experiments that are doable and scalable. Here "doable" means that a working experiment can be built on a benchtop. "Scalable" means that making the apparatus smaller makes the experiment work better. This reflects our pragmatic opinion that progress is most likely to be achieved by cumulative improvement of working devices.

Since this passage appeared in print, our UW group has advanced by a succession of "doable" and "scalable" experiments to the recent completion (under an NSF MRI grant, DBI-9724426) of a cryogenic 3D MRFM scanner designed for nanoscale resolution. As described in the

Personnel	Institution	Email Address	MRFM Interests
John Sidles Joe Garbini Bill Dougherty	University of Washington	sidles@u.washington.edu garbini@u.washington.edu dougherty@me.washington.edu	Biomolecular Structure
Dan Rugar Nino Yanoni	IBM Almaden Research Center	rugar@almaden.ibm.com yanoni@almaden.ibm.com	Magnetic Storage and Single Spins
Chris Hammel Gennady Berman	Los Alamos	pch@lanl.gov gpb@lanl.gov	Ferromagnetic Resonance Quantum Computing
John Moreland	NIST Boulder	moreland@boulder.nist.gov	MRFM Metrology
Doran Smith	University of Maryland, U. S. Army Research Lab	dsmith@squid.umd.edu	Nuclear MRFM; Optical Pumping
Daniel Weitecamp Michael Roukes	Cal Tech	weitekamp@romeo.caltech.edu roukes@caltech.edu	MRFM Spectroscopy
John Markert	Univ. of Texas, Austin	markert@physics.utexas.edu	Bulk Nuclear MRFM
Raul Fainchtein	Johns Hopkins	raul.fainchtein@jhuapl.edu	Bulk Nuclear MRFM
John Mahron	Cornell University	jam99@cornell.edu	Bulk Nuclear MRFM
Lorenzo Lenci Massimo Martinelli	Consiglio Nazionale delle Ricerche, Pisa	lorenzo@risc.pi.cnr.it massimo@risc.pi.cnr.it	Angular Momentum Absorption
Wiebren Veeman	University of Duisburg	w.s.veeman@uniduisburg.de	Bulk Nuclear MRFM
H. J. Güntherodt E. Meyer	University of Basel, Switzerland	guentherodt@ubaclu@unibas.ch	MRFM Instrumentation
Phillip Wigen	Ohio State University	wigen@mps.ohio-state.edu	Quantum Computing

Table 2: Active MRFM Researchers

following sections, this scanner will be the experimental vehicle for investigating our three, interrelated objectives: spin relaxation, 3D imaging, and quantum observation.

C.8.1 Research Activities

We propose a three-year plan in which we combine our ongoing theoretical and experimental techniques for studying spin relaxation with two new activities: first, 3D imaging with a next-generation digital signal processor (DSP) cantilever controller and, second, quantum analysis of MRFM estimation and control. All three activities will begin at the start of the grant period. The research activities will be scheduled as follows.

Year One: By the end of the first year we will have completed the calibration of our new 3D MRFM scanner. Calibration will be accomplished by making MRFM images of cast replicas of commercial force microscope calibration standards. The replicas will be made with a paramagnetic medium, *e.g.*, DPPH-doped polystyrene. Using samples of known geometry we will investigate the three most promising imaging techniques: (1) direct force imaging, (2) parametric imaging, in which variations in the spring constant are detected, and (3) multiplexed imaging, which produces images from multiple resonant slices simultaneously.

Year Two: By the mid-point of the grant period the next-generation adaptive digital controller, incorporating optimal control and diagnostic algorithms, will be completed and evaluated. Adaptive control will allow reliable operation at smaller tip-sample separations, resulting in a higher magnetic field gradient and therefore a higher signal-to-noise ratio. We anticipate substantial improvement over the first-year images.

Year Three: By the end of the grant period the results of all three activities will come together: (1) The 3D MRFM imaging studies using the advanced controller will be evaluated, (2) a survey of electron and proton spin relaxation in a variety of target samples will have been completed, and (3) the quantum effects on estimation, control, and imaging in MRFM will be better understood.

Our MRFM laboratory at the University of Washington has provided an educational venue for excellent education of a succession of students: undergraduate (2), masters (4), Ph.D. (2) and postdoctoral (2). The research plan articulated here offers exciting research opportunities for two well-prepared Ph.D. students: one working on imaging and control, and the other working on spin relaxation and quantum analysis.

In the following sections we describe the MRFM apparatus and review the experimental methods that will be used to complete these research activities.

C.8.2 Experimental Methods: MRFM Apparatus

A block diagram of our present system appears in Fig. 3. A complete description of this apparatus can be found in our recent *Journal of Magnetic Resonance* article [18]. The MRFM force microscope assembly is housed in a turbo-pumped cryostat that achieves a vacuum of better than 10^{-7} Torr at 10 K. The force-detecting element is presently a soft commercial cantilever ($k = 0.015$ N/m, resonance frequency 7792 Hz), mounted with a 5.8 μm-diameter SmCo magnetic tip. The cryogenic 1D stick-slip shear-piezo sample positioner is driven by a high voltage amplifier. Resonant microwave or RF fields are produced by a three-turn microcoil 120 μm in diameter. Micro-coaxial cable connects the coil via a vacuum feedthrough to microwave (1–20 GHz) and/or RF (DC–40 MHz) synthesizers, which are

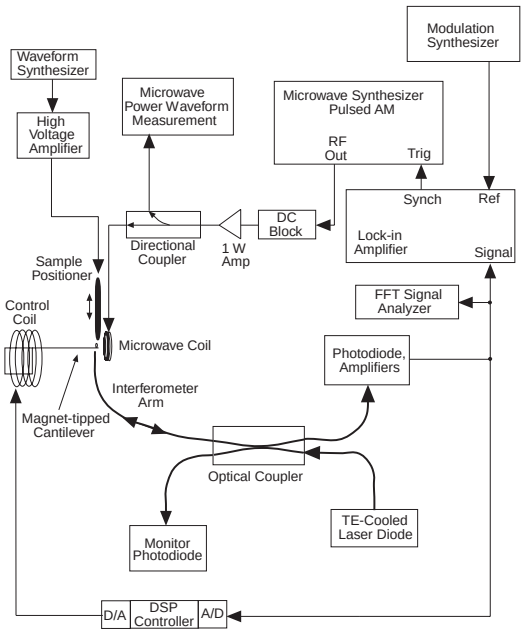


Figure 3: Schematic of the presently-installed 1D MRFM apparatus. Our newly completed NSF MRI 3D scanner will inherit most of this support instrumentation.

amplitude and/or frequency modulated. Another, larger coil provides audio-frequency force-feedback from a DSP controller.

An all-fiber interferometer detects the cantilever motion. The effective interferometer noise floor, expressed in terms of cantilever displacement is 0.016 $\text{Å}/\sqrt{\text{Hz}}$, corresponding to an equivalent noise temperature of 0.3 mK; thermal (Langevin) motion of the cantilever is the dominant noise process. The interferometer is fringe-centered by a novel thermal-tuning technique [12]. A battery-isolated photoreceiver converts the stablized interferometer output into a robust voltage signal which is detected by a lock-in amplifier. The instruments in the experiment are operated by LabView software via GPIB.

Our newly completed NSF/MRI 3D scanning device is depicted in Fig. 4. The four-quadrant piezo tube device features a lateral (X,Y) scanning distance of approximately 2.6

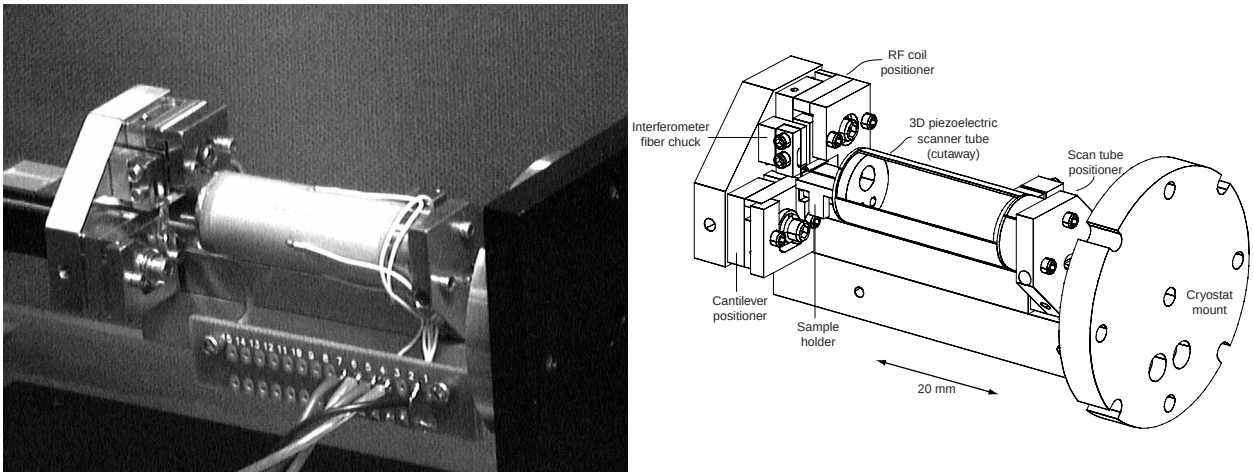


Figure 4: Photograph and drawing of the new NSF MRI 3D piezo scan head.

μm at 4 K. Effectively unlimited vertical displacement (Z) is obtained by a new stick-slip sample holder design. An accompanying computer-controlled, low-noise, high-bandwidth $\pm$150 V driver has been completed. The tube and driver combination is designed to achieve sub-Angstrom voxel scanning precision.

C.8.3 Experimental Methods: Spin Relaxation

Spin relaxation rates in MRFM are, generally speaking, straightforward to measure experimentally, because many of the standard methods of magnetic resonance can be adapted to the high gradient MRFM environment. Spin relaxation is conventionally characterized in terms of relaxation times $\{T_1, T_2\}$ which appear as parameters in the Bloch equations. By a detailed analysis of Bloch equations in high-gradient force microscopy [18], we have shown that T_1 can be determined from the phase lag of the force signal relative to an applied RF modulation, while T_2 can be determined from the "knee" in a force-RF power curve (note: the data in Fig. 1 show a typical knee).

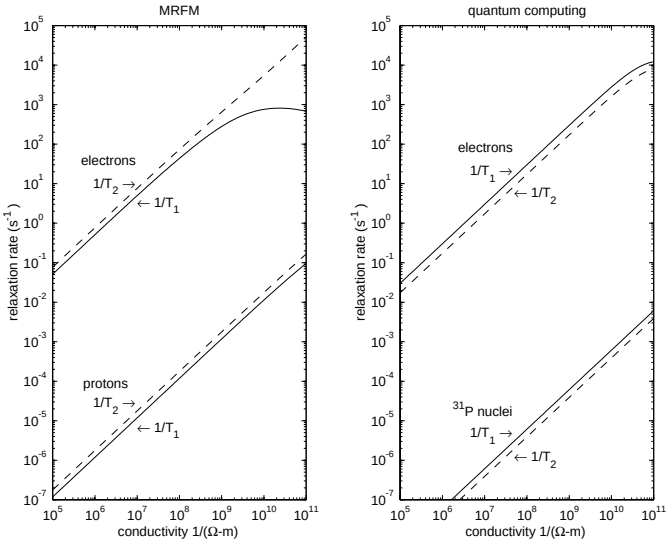


Figure 5: Predicted electron and proton spin relaxation rates in MRFM and quantum computing, as a function of tip conductivity, from [52]. Operating parameters: tip diameter 1 μm, tip-sample separation 50 nm, ambient B-field 0.58 T, temperature 4 K. Note the predicted rapid relaxation rates for tip conductivities in excess of 10^6 $(\Omega{\cdot}\mathrm{m})^{-1}$.

Observed relaxation rates in MRFM are typically more rapid than theory and/or bulk measurements would lead one to expect. This situation has ample historical precedent. In a review of ESR relaxation in traditional bulk samples, Orbach and Stapleton [43] emphasize that achieving a firm understanding of relaxation mechanisms in ESR spectroscopy required a forty-year iterative struggle.

In MRFM experiments the nearby magnetic tip contains at least four novel thermal reservoirs which might plausibly couple to spins in the nearby sample. These thermal reservoirs are: (1) paramagnetic spins in the passivating oxide layer of the tip, (2) ferromagnetic spin wave excitations in the tip itself, (3) thermally excited domain wall motions, and (4) thermally excited currents in the conduction band.

In a recent theoretical study of thermal magnetic noise [52, see Fig. 5] originating in the tip conduction band, we predicted electron spin relaxation rates sufficient to require considerable care in the design of single-spin MRFM experiments.

We propose to begin our study of MRFM relaxation mechanisms in a sequential measurement of ESR relaxation in samples plated onto (1) plain silica, (2) silica plated with gold

(for conduction band relaxation), (3) silica plated with unoxidized cobalt (for ferromagnetic relaxation), (4) silica plated with oxidized iron (for passivation layer relaxation). We will particularly seek to identify samples with long electron spin relaxation times, because such samples are suitable for the "grand challenge" MRFM goal of observing single electron spin moments. With these pilot experiments accomplished, we will seek partners for MRFM-based ESR studies of passivating layers in, *e.g.*, magnetic recording materials.

C.8.4 Experimental Methods: Control and Imaging

A critical element of MRFM imaging is the active control of cantilever dynamics via a torque applied to the tip. Control accomplishes three goals which greatly enhance the practical effectiveness of soft (high sensitivity), high-Q cantilevers: (1) it broadens the cantilever response bandwidth, (2) it reduces the system damping time and, (3) it lowers the thermal vibration amplitude, allowing instrument designs of inherently high sensitivity to achieve the required positional accuracy, without a penalty in signal-to-noise-ratio.

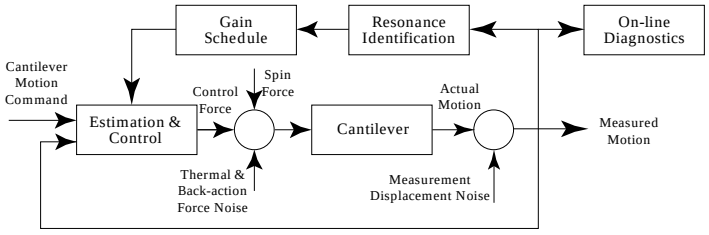


Figure 6: Adaptive cantilever control with online diagnostics

As shown in Fig. 6 the cantilever is subject to thermal (Langevin) and back-action (photon flux) force noises, as well as displacement (measurement) noise resulting from shot noise in the interferometer. The control force is generated through a magnetic field acting on the magnetic tip. All of these noise processes are well characterized by theory. An optimal controller/observer strategy that balances the control effort and the position accuracy requirements, while minimizing the covariance of estimated position satisfies the requirement of most magnetic resonance experiments [11, 13, 14, 23].

Three-dimensional imaging gives rise to new control requirements. First, as the tip-sample separation diminishes, electrostatic forces (which may be attractive or repulsive), and the attractive Casimir force [32, 33] alter the effective spring constant of the cantilever. At close approach, the Casimir force dominates and tends to counterbalance the spring restoring force, substantially lowering the resonance frequency. As the resonance frequency shifts, a fixed-parameter controller falls out of tune with the cantilever. The presence of noise, and the necessarily limited control effort allowed while measuring attoNewton forces generally precludes the use of wide bandwidth control.

We have recently completed a series of experiments using the adaptive technique (Fig. 6). A discrete Hilbert transform, implemented in the DSP along with the controller/estimator, identifies the shift in resonance frequency. The identified frequency is used to continually index the gain scheduler without breaking the control loop. Unlike least mean square (LMS) identification, the Hilbert transform technique proved to be robust to measurement noise, even when operated in closed loop. The adaptive controller tracked changes as small as 0.1% of the resonance frequency, while accommodating changes in the effective stiffness greater

than 50%. The results of these preliminary experiments are promising, yet substantial work remains to integrate adaptive control into the imaging environment, and to develop general design criteria for fully adaptive MRFM.

The second requirement of imaging is for continuous diagnostic assessment of the experiment. An important lesson of the past five years of measuring attoNewton forces has been that parasitic effects can be detected and managed by monitoring the statistical characteristics of the measured cantilever motion. The DSP is the natural choice for implementing a suite of online diagnostic algorithms.

Recent advances in digital signal processors now allow both the adaptive control and diagnostic functions to be accomplished in the DSP. This "next-generation" cantilever controller will be the key element of our investigation of nanoscale MRFM imaging.

C.8.5 Experimental Methods: Quantum Control and Observation

The goal of this portion of our research plan is, to our knowledge, entirely novel. We propose to create an analytical quantum model, and an end-to-end computer simulation of quantum dynamics, for the combined spin-cantilever-interferometer-controller system described on the previous pages.

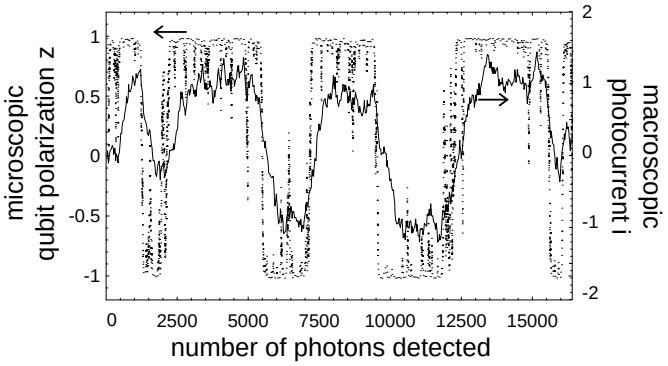


Figure 7: Dynamics of a spin system continuously observed by interferometry while weakly coupled to thermal noise, from [51]. The interferometer photocurrent (solid line) is strongly but not perfectly correlated with the microscopic quantum polarization (dotted line). Note that coarse-grained "quantum jumps" emerge from fine-grained stochastic "jitter".

Our motivation comes from a question that Dan Rugar asked us in 1996: "How does the Stern-Gerlach effect work in MRFM?" We remind reviewers that the Stern-Gerlach effect is the empirical fact that a carefully measured single quantum spin is always found to be spin-up or spin-down; never in a mixed state.

To address this question at a level of detail that would satisfy Rugar's curiosity, and our own, we first analyzed a simpler problem: that of a single spin continuously, but weakly, observed by an interferometer, in the presence of thermal noise. The results of this analysis were extremely useful from an quantum engineering point of view [51, see Fig. 7]. Seemingly mysterious quantum phenomena like quantum "jumps", the Stern-Gerlach effect, the quantum Zeno effect, and the AC Stark Effect emerged naturally as coarse-grained aspects of a Fokker-Planck description of the interferometric measurement process competing with the thermal decoherence. It turned out that both an analytic description and a computer simulation of the measurement process were possible; these two approaches were (naturally) in complete agreement, and each illuminated the other (Fig. 7).

We propose to apply this same approach in a system with two added elements: a cantilever and a feedback control loop. As in [51], a computer simulation of the entire closed loop quantum system is feasible, albeit with some added bookkeeping to keep track of the cantilever dynamics. At high temperatures, or in weakly observed systems, we confidently

expect the simulated behavior of the controlled spin-cantilever-interferometer system to be of the classical Langevin type. But as the temperature is lowered, or the strength of the observational interaction is increased, or the coupling to the thermal reservoir is reduced, phenomena like quantum jumps and the Stern-Gerlach effect should emerge, again as the result of competition between the measurement process and the thermal reservoir.

From the viewpoint of classical control theory, it is not even clear what state variables should be assigned to a weakly observed quantum system; much work remains to be done.

This proposed simulation will be far more intricate than that of [51], both analytically and in computer terms. It will also be of far greater practical consequence. Following upon our ongoing studies of spin relaxation, imaging, and control, the proposed quantum analysis will be the final piece of the puzzle needed to justify the contemplated national initiative for quantum molecular observation (Section C.5).

C.9 Integrating Research and Education

A major educational objective of our MRFM group is to establish quantum engineering as a degree-granting program or specialty within the University of Washington College of Engineering. If administrative approval is forthcoming, the two Ph.D. students supported under this proposal will graduate as the University of Washington's first degreed Quantum Engineers.

For this to happen, we will have create for the College of Engineering a well-defined quantum engineering curriculum. To this end, we plan to create a one year course, entitled "Practical Quantum Engineering", which will cover the research topics set forth in Section C.8.5, at a level appropriate to senior or first year graduate students in engineering.

C.10 Broader Impacts

Practical quantum molecular observation will mark the coming of age of a new engineering discipline, *quantum engineering*. The proposed opening of a new imaging window onto the largely unobserved world of 3D molecular structure will be among the first "grand challenge" achievements of quantum engineering, and will revolutionize the fields of nanoscale engineering, materials science, biology, medicine, and engineering education.

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