Magnetic Resonance Force Microscopy

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Purpose

• Imaging mechanisms have many uses in our society
  – Common
    • X-Ray, MRI
  – Specialized
    • AFM, Electron Microscopy, NMR

• Better resolution can open up new possible applications
  – Quantum computing, Molecular imaging
Atomic Force Microscopy

• AFM was invented in 1986 and is one of the most popular tools for imaging
• AFM can function in 2 primary modes: Contact and non-contact
Problems with AFM

- Contact mode AFM techniques cannot be used for imaging at a scale that is needed to detect single spins.
- The contact between the needle and the surface can damage both if not used with extreme care.
- Although many competing imaging techniques have been developed, AFM is still a robust technique.
- AFM can only scan the top surface of the sample, thus limiting its use in sub-surface imaging.
Origins of MRFM

• MRFM was originally proposed in the early 1990s
  – “as a means of obtaining three-dimensional images of individual biological molecules”

• This technique showed potential of imaging at a single spin level but was limited by the apparatus

• Recent advances in ultra sensitive Cantilever-based force sensors and better understanding of the physical processes have made Single Spin detection possible

• Using MRFM, the authors report that they were able to observe a 25nm spatial resolution
Principles behind MRI and MFRI

• MRI and MFRI are both based on the same physics of the sample
  – Spin of electrically charged particles

• They differ in the technique used to measure the spin
  – MRI utilizes induction
  – MFRI utilizes mechanical force
Spin and magnetic resonance

• Example system - Hydrogen atom
  – *Nuclei used to create clinical MRI images*
• Nucleus has a net positive charge due to the proton

Spin and magnetic resonance

- Proton has a spin and a mass
  - Rotates like a spinning top
  - Angular momentum associated with it
    - Behaves like a gyroscope and retains spatial direction of its axis of rotation
- Proton has a magnetic moment
  - Due to it being a rotating electrical charge
  - A tiny magnet
  - Affected by magnetic fields and electromagnetic waves
  - Can induces an electrical potential if it moves
  - Can’t directly measure spin direction of the proton but can measure the resulting magnetic axis
Spin and magnetic resonance

- By applying a magnetic field the spins will try to align along the field direction.
- The spins react with an avoiding action called precessional motion.
- *Larmor frequency* is the characteristic frequency associated with the precessional motion of spins located in a magnetic field.
  - MRI and MRFI are based on the larmor frequency.
  - Exactly proportional to the strength of the magnetic field.
- *Larmor equation*

\[ \omega_0 = \gamma \times B_0 \]

- \( \omega_0 \) – larmor frequency (MHz)
- \( \gamma \) – gyromatic ratio (constant determined by the material)
- \( B_0 \) – magnetic field strength (T)
Spin and magnetic resonance

- The majority of spins align in the applied magnetic field (z-direction)
- The magnetic vectors of the aligned spins add together to create a longitudinal magnetization in the z-direction, $M_z$
Spin and magnetic resonance

- Possible to flip the spin direction
- An electromagnetic wave having the same frequency as the Larmor frequency, $\omega_0$, can be used to transfer energy to the spins
  - *Resonance condition*
- Applying a RF pulse with the correct pulse and duration can cause the spins to flip
- As the spins flip so does their longitudinal magnetization, $M_z$
Spin and magnetic resonance

Spins align in direction of magnetic field

RF changes alignment of spins

Magnetization of spins now in xy-plane (transverse magnetization $M_{xy}$)
How MRI and MRFI differ

- The motion of $M_z$ can be measured and used to determine information about the system

**MRI** - motion of $M_z$ induces an alternating voltage in the receiving coil with a frequency equal to the Larmor frequency

**MRFI** - motion of $M_z$ acts to change the frequency at which the cantilever vibrates at

SIDLES JA, GARBINI JL, BRULAND KJ, et al. REVIEWS OF MODERN PHYSICS
MRFI - setup overview

- A ferromagnetic tip is attached to a cantilever that is sensitive enough to bend in response to very small forces
- Apply a RF magnetic field at the Larmor frequency the magnetic moments of either the nucleus or electrons within a slice of the sample can be flipped up or down

SIDLES JA, GARBINI JL, BRULAND KJ, et al.REVIEWS OF MODERN PHYSICS
MRFI - setup overview

- This flipping generates an alternating force on the magnetic tip that causes the cantilever to vibrate
- Vibrations are detected using an interferometer

Rugar D, Budakian R, Mamin HJ, et al. NATURE
T1 - longitudinal relaxation

- Over time spins will gradually return to being oriented along the external magnetic field, $B_0$, – **longitudinal relaxation**
- The magnitude of the transverse magnetization, $M_{xy}$, decreases
- The magnitude of the longitudinal relaxation, $M_z$, will increase
T1 - longitudinal relaxation

- Energy is emitted into the surroundings
- T1 - time constant of longitudinal relaxation
  - Independent of strength of $B_0$ and internal movement of molecules
- Determines how fast the spins will return to their original starting positions oriented along $B_0$ and be able to be excited again
Phase

- Phase refers to an angle

Spin A precesses in the XY plane

Spin B precesses with the same speed as A but 20° behind A, *phase difference = -30°*
Phase coherence

- Directly after excitation all spins are in phase
  - *Phase coherence*
- Phase coherence vanishes following excitation
- Individual magnetic vectors cancel each other out
- The transverse magnetization vector $M_{xy}$ becomes smaller and eventually vanishes
T2 - transverse relaxation

- Loss of transverse magnetization, $M_{xy}$, due to loss of phase coherence
- No energy emission to the surroundings
- Energy is exchange between spins
  - Neighboring spins set up local magnetic fields, $B_L$
  - The precession frequency of a spin changes based on $B_L$
  - Phase coherence is lost
Advantages of MRFM

• The review article by Sidles et al. summarized the appeal of MRFM in 3 simple for very important points:
  – The magnetic imaging is non-contact and specific to electron and nuclear spins
  – The imaging magnetic field is 3-Dimensional and reaches below the scanned surface allowing for imaging of subsurface structures
  – The mathematics and theory behind magnetic resonance is well understood and the algorithms involved in image deconvolution are well conditioned
Using MRFM

- The fundamental challenge to achieving single-spin sensitivity is the magnitude of the force exerted by an electron.
  - This force is measured in attonewton (1 aN = 10^{-18} Newtons)
- In comparison to the AFM, force is 1 Million times smaller
- MRFM can be used to scan beneath the topographic surface of a sample (100nm)
- Successful application at this scale requires very sensitive equipment and small tolerances
Experimental Setup

• MRFM uses:
  – Mass-loaded Si cantilever (150nm wide SmCo magnetic tip)
  – A sample of vitreous Silica
  – A external magnetic field source (coil)
  – The experiment was performed in a small vacuum chamber at 1.6Kelvin
    • Sm => Samarium
    • Co => Cobalt
Procedure

• At first, the sample is irradiated with 2-Gy dose of Co60 gamma rays
  – This produces a small concentration of dangling bonds containing unpaired electrons
• The estimated concentration of spins is approx. $10^{14}$ cm$^{-3}$
  – For simplification, it is assumed that the unpaired electrons are far enough to not interfere with each other
• An external microwave magnetic field is applied to the system to create a resonant slice within the sample
• The spin must be slightly in front or slightly behind the tip in the x direction to create a noticeable change in the cantilever (for a vertical tip)
Resonant Slice

- Due to the deposit of SmCo on the tip, the tip has a magnetic field \( B_{\text{tip}}(x,y,z) \)
- A static magnetic field \( B_{\text{ext}}(z) \) is applied to the system
- A “resonant slice” is formed at the position where the sum of the two magnetic fields is equal to the condition for electron spin resonance
  \[ B_0(x,y,z) = |B_{\text{tip}}(x,y,z) + \hat{z} B_{\text{ext}}| = \omega_{\text{rf}} / \gamma \]
  \[ \gamma / 2\pi = 2.8 \times 10^{10} \text{ Hz T}^{-1} \quad \omega_{\text{rf}} / 2\pi = 2.96 \text{ GHz} \]
- \( B_0(x,y,z) = 106 \text{ mT} \) and \( B_{\text{ext}} = 30 \text{ mT} \)
- The thickness of the slice is inversely proportional to the gradient of the magnetic field
- Typically, the resonant slice is a surface that extends 250nm below the tip
Force Microscopy

• map force gradients near surfaces w/o contact
• Force gradients are detected as shifts in the resonant frequency of the mechanical vibration of a cantilever that is positioned near the surface of interest
• Common detection schemes:
  – Cantilever is driven at a constant frequency
  – Force gradient detected as variation in amplitude or phase of the cantilever vibration.
Improvements on Force Microscopy

• Signal to noise ratio (S/N) and sensitivity can be increased by increasing Q of cantilever
• High Q means smaller max available BW
• Small BW means a slow system

Need an improved detection method that increases sensitivity through high Q w/o decreasing BW
Slope Detection

- Cantilever is driven at a fixed frequency $w_d$ slightly off resonance frequency, $w_0$.

  \[ \omega_0^2 = \frac{k_{\text{eff}}}{m} \]
  \[ k_{\text{eff}} = k_L + \frac{\partial F}{\partial z} \]

- $m$: effective mass, $k_L$: force constant, $\frac{\partial F}{\partial z}$ force gradient
- Change in $\frac{\partial F}{\partial z} \rightarrow$ shift in resonant frequency $\rightarrow$ shift in vibration amplitude
- Derive signal by measuring change in amplitude

Albercht, 1991
Slope Detection Limitations

Minimum detectable force gradient:

\[ \delta F_{\text{min}}' = \sqrt{2 k_L k_B TB} / \omega_0 Q < z_{osc}^2 \]

Maximize sensitivity by using high Q?

Increasing Q restricts BW: \( t = 2Q/w_0 \)

Low Q: fast response, low sensitivity

High Q: slow response, high sensitivity

Albercht, 1991
Frequency Modulation Technique

- Cantilever serves as frequency-determining element (constant amplitude)
- The frequency of the cantilever is instantaneously modulated by variations in the force gradient acting on the cantilever
- S/N for a given BW depends on Q
- BW is governed only by the characteristics of the FM demodulator
- Can increase Q w/o decreasing BW
FM Detection

- High Q cantilever
- Changes in force gradient cause change in oscillator frequency which are detected by a FM demodulator
- AGC: maintains vibration amplitude at constant level
- Frequency detection: tunable analog FM detector

Albercht, 1991
Comparison

Minimum detectable force gradient:

\[ \delta F'_{\text{min}} = \sqrt{\frac{4k_Lk_BT\beta}{\omega_0Q}} \langle z_{osc}^2 \rangle \]

min detectable \( \frac{\partial F}{\partial z} \sim \) same as slope detection method

✓ Similar sensitivity
✓ Independent BW and Q
✓ Increase sensitivity by higher Q without affecting BW

Albercht, 1991
OSCAR

Oscillating Cantilever-driven Adiabatic Reversals

- Cantilever acts as frequency determining element
- Gain-controlled positive feedback loop drives the cantilever to oscillate at a set amplitude.
- As the cantilever vibrates, position of resonant slice oscillates through a region of the sample
- Spins in the resonant slice cyclically invert due to the effect of adiabatic rapid passage
OSCAR

- Cyclic inversion generates an oscillatory interaction force
- modifies the cantilever restoring force
- change in spring constant: $\Delta k \approx \frac{F_{\text{spin}}}{\Delta z}$

$F_{\text{rms}}$: rms amplitude of oscillating force from spins

$\Delta z$: rms cantilever amplitude

- change in oscillation frequency $\frac{\Delta f}{f} \approx \left(\frac{1}{2}\right)(\frac{\Delta k}{k})$

- detected by analog frequency demodulator
• In other words, the alternating magnetic force on the cantilever mimics a change in cantilever stiffness:

\[
\delta f_c = \pm \frac{2 f_c G \mu_B}{\pi k x_{\text{peak}}} \quad G \equiv \frac{\partial B_0}{\partial x}
\]

• sign of frequency change depends on relative phase of the spin inversion wrt the cantilever motion

• Rugar’s experiment: \(|\delta f_c| = 3.7 \pm 1.3 \text{mHz}|

OSCAR
Interrupted OSCAR

Microwave field, $B$, is turned off for one-half to a cantilever cycle every 64 cycles, $f_{\text{int}} = f_c/64 = 86$Hz

Interrupt $B \Rightarrow$ relative phase of spin and cantilever reverses (frequency shift $\Rightarrow$ reverse polarity) $\Rightarrow$

frequency shift alternates between positive and negative values in a square-wave with

$$f_{\text{sig}} = f_{\text{int}}/2 \sim 43\text{Hz}$$

Rugar, 2004
iOSCAR Data Analysis

- Fourier series of a square wave: \( f(x) = \frac{4}{\pi} \sum_{n=1,3,5,...}^{\infty} \frac{1}{n} \sin\left(\frac{\pi n x}{L}\right) \).

- Frequency shift signal:

\[
\Delta f(t) = \frac{4}{\pi} |\delta f_c| A(t) \sin(2\pi f_{\text{sig}} t) + \text{higher harmonics}
\]

- \( A(t) \): signal will not be perfectly periodic cause of extra random spin flips induced by the environment

\(<A(t)> = 0, \quad <|A(t)|^2> = 1\)

- relatively large frequency noise of cantilever due to thermal motion and tip-sample interaction \(\rightarrow\) signal averaging (square of signal amplitude)
iOSCAR Data Analysis

- analog frequency demodulator and lock-in amplifier determine the energy variance of in-phase and quadrature component of frequency shift.

**Lock in Amplifier:**
measures a small signal even in presence of noise

\[
\begin{align*}
  v_{\text{in}} &= V_0 \sin(\omega t) \\
  v_{\text{sq}} &= \frac{4}{\pi} \left( \sin(\omega t) + \frac{1}{3} \sin(3\omega t) + \frac{1}{5} \sin(5\omega t) + \ldots \right) \\
  v_{\text{out}} &= \frac{2V_0}{\pi} \left( 1 - \frac{2}{3} \cos(2\omega t) - \frac{2}{15} \cos(4\omega t) - \frac{2}{35} \cos(6\omega t) \ldots \right)
\end{align*}
\]
iOSCAR Data Analysis

- spin signal and measurement noise uncorrelated $\Rightarrow$
  \[ \sigma_I^2 = \sigma_{spin}^2 + \sigma_{noise}^2 \]
- quadrature variance contains noise data only $\Rightarrow$
  \[ \sigma_{spin}^2 = \sigma_I^2 - \sigma_Q^2 \]

Lateral scan:
Peak $\Rightarrow$ single spin
Low S/N: had to use considerable averaging

Rugar, 2004
Field Dependence of Spin Signal

- Reduce external field $\rightarrow$ shrink resonance slice $\rightarrow$ shift in scan position of signal peak
- $B$: 34 to 30 mT $\rightarrow$ peak shift of 19nm
  $\Delta B / \Delta x \rightarrow G \sim 2 \times 10^5$ Tm$^{-1}$, field gradient

Rugar, 2004
Magnetic Resonance Dependence

• signal disappeared if the microwaves were absent or turned on continuously
• varying the timing of microwave interruptions → different outcome
• signal disappeared if the starting time of interruption was shifted by \( \frac{1}{4} \) of the cantilever cycle
• signal disappeared when the interruption duration was a full cantilever cycle
Single Spin Detection

Spatial isolation of the spin signal \(\rightarrow\) single spin

Low spin density: \(10^{13}\) to \(10^{14}\) cm\(^{-3}\)

- 200 to 500 nm spacing between spins
- most sample locations have no spin interacting with the resonant slice \(\rightarrow\) zero baseline in previous plot

A spin signal sample was scanned through \(\sim 30\) independent locations in order to locate a well-positioned spin and hence obtain a strong signal.
Quantum Computation: an Application

• single spin qubit state readout is a big challenge
• detecting single electronic moment is crucial
• MRFM: directly measure the spin of single moment
• magnetic resonance imaging of MRFM: able to select the individual electron moment that is to be detected

References


Spectral Analysis

- The following plots are the result of 2 scans of the sample (laterally in the x direction) with 2 magnitudes of the external field.
Position vs. Frequency

- The following false color plot shows the power spectral density as a function of position.
- The graph shows that the spin signal is localized both spatially and spectrally.
Results

• From the experiment, the authors were able to determine that the spectrum can be fitted with a Lorentzian function

\[ S(f) = \frac{4\tau_m \langle [\Delta f_1(t)]^2 \rangle}{[1 + 4\pi^2 \tau_m^2 f^2]} \]

• The spectral width at half-maximum was found to be 0.21 Hz and \( \tau_m = 760\text{ms} \)

• The total magnitude of the spin signal (by integrating the spectrum): \( \langle [\Delta f_1(t)]^2 \rangle = 28\text{mHz}^2 \)

• Solving for \( |\delta f_c| \) resulted in the value \( |\delta f_c| = 4.2\text{mHz} \) which is very close to the expected value of 3.7mHz
Improvements

• Although the results are an astounding success, further improvements are still possible (and in some cases, needed)
• The authors suggest that the following improvements are needed:
  – A higher field gradient, resulting in a dramatic speed increase in the acquisition time (possibly enabling 2-D and 3-D imaging)
  – A decrease in the measurement time to below Tm allowing for a real time readout of the spin quantum state
  – Extension to single nuclear spin detection – requires a 1,000 fold improvement in magnetic moment sensitivity