Magnetic Resonance Force Microscopy

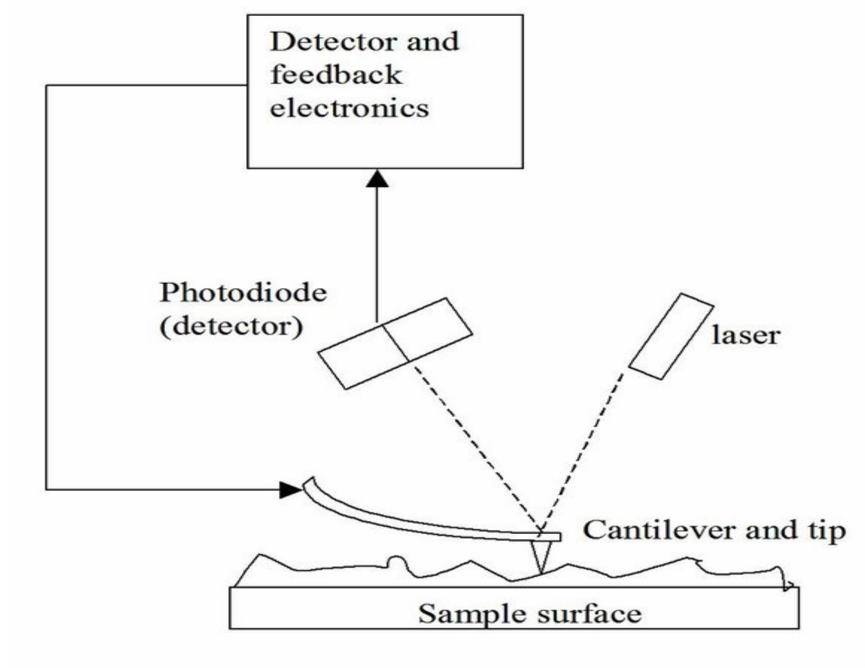
By:
Jaspreet Wadhwa
Stephanie Teich-McGoldrick
Zeinab Mousavi

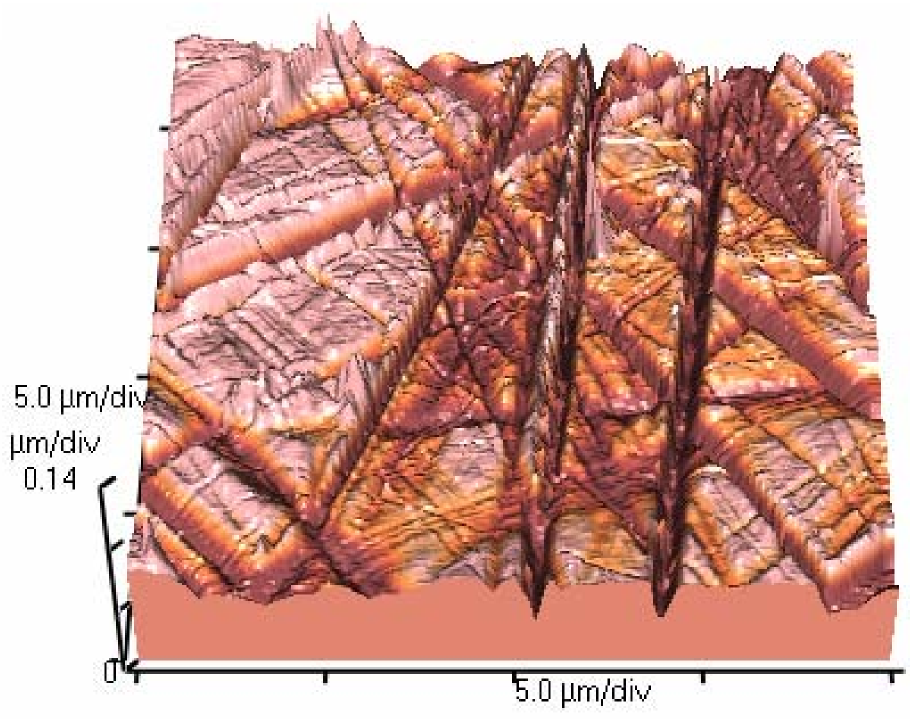
Purpose

- 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

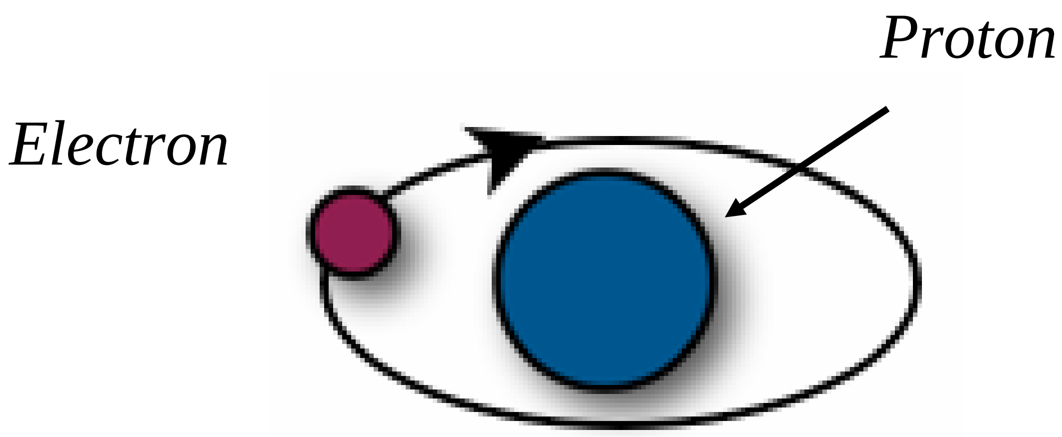
- MRFM was originally proposed in the early 1990s
  - “as a means of obtaining three-dimensional images of individual biological molecules”
- This technique showed potential of imaging at a single spin level but was limited by the apparatus
- Recent advances in ultra sensitive Cantilever-based force 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*
  - *MRFI utilizes mechanical force*

Spin and magnetic resonance

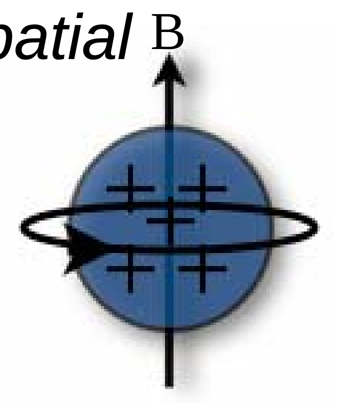
- Example system - Hydrogen atom
  - *Nuclei used to create clinical MRI images*
- Nucleus has a net positive charge due to the proton



Weishaupt, Kochli, Marinek, *How does MRI work?,* Springer, 2003

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*

$$\omega_0 = \gamma * B_0$$

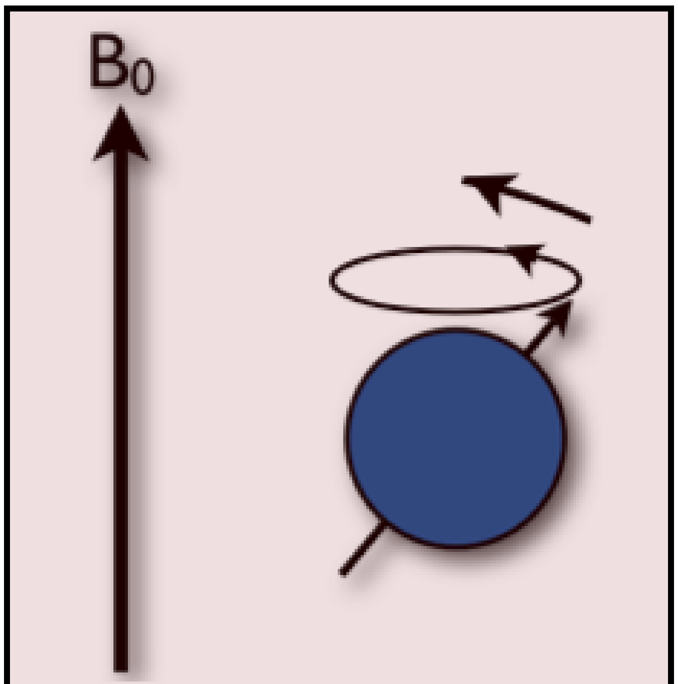
ω_0 – larmor freqency (MHz)

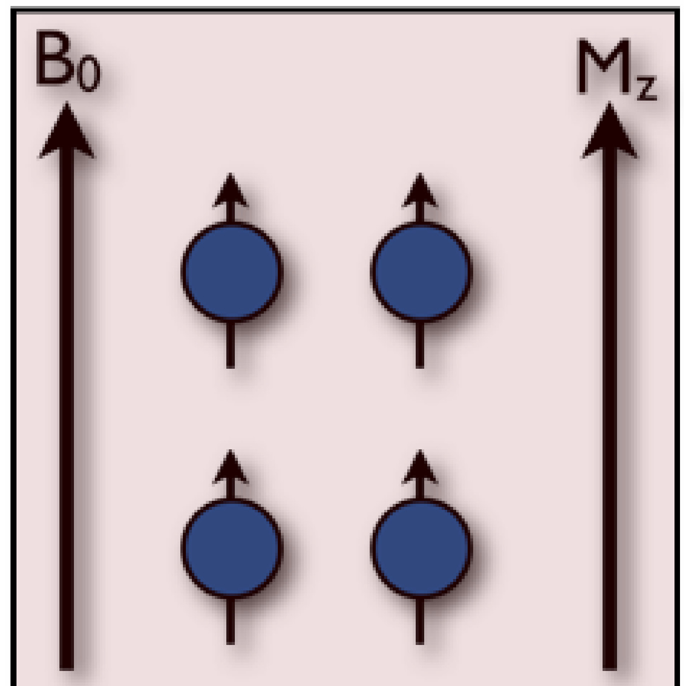
γ – gyromatic ratio (constant determined by the material)

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

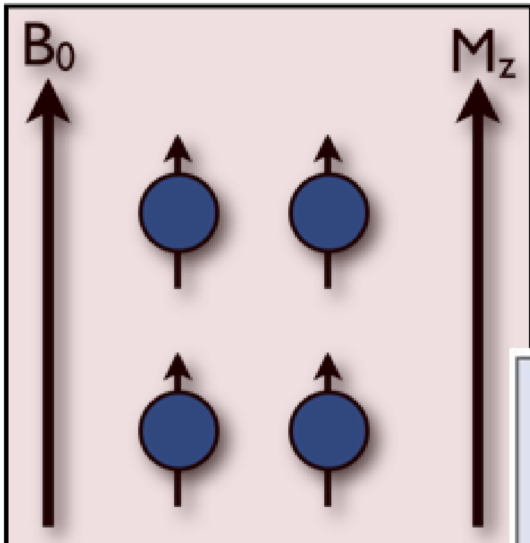




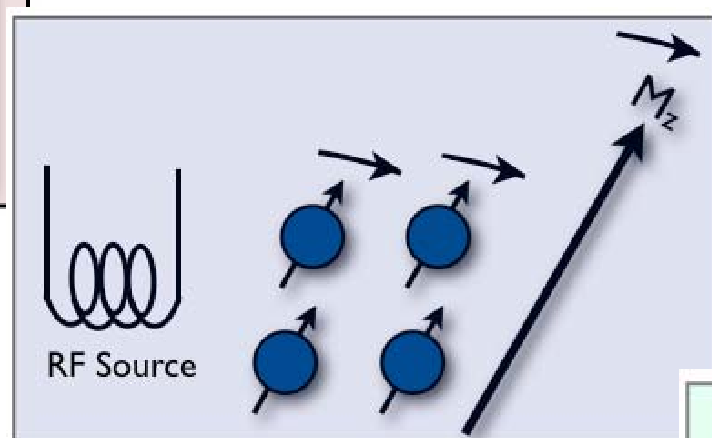
Spin and magnetic resonance

- Possible to flip the spin direction
- An electromagnetic wave having the same frequency as the Larmor frequency, ω_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

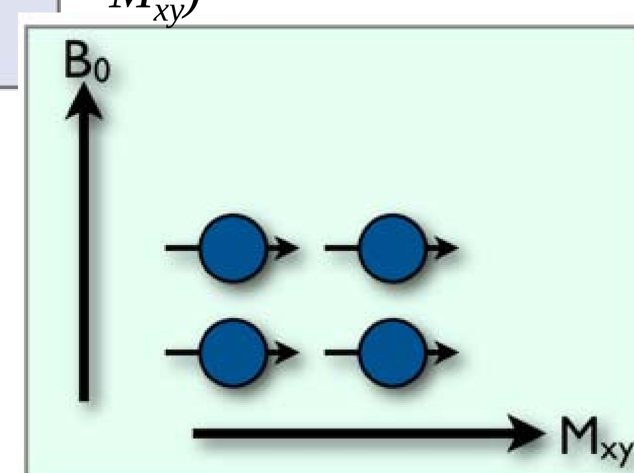


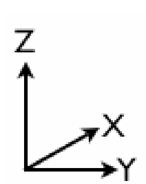
Spins align in direction of magnetic field



RF changes alignment of spins

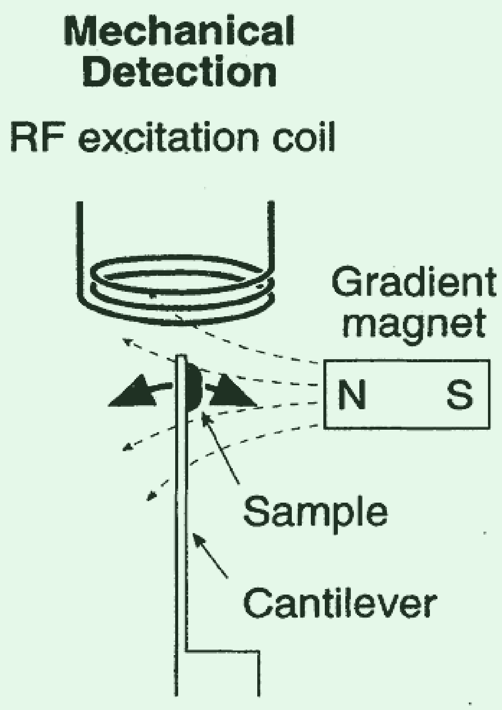
Magnetization of spins now in xy-plane (transverse magnetization M_{xy})



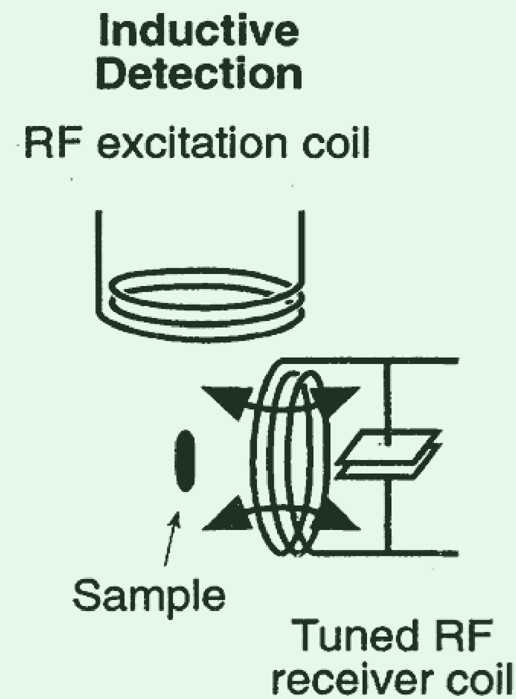


How MRI and MRFI differ

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



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

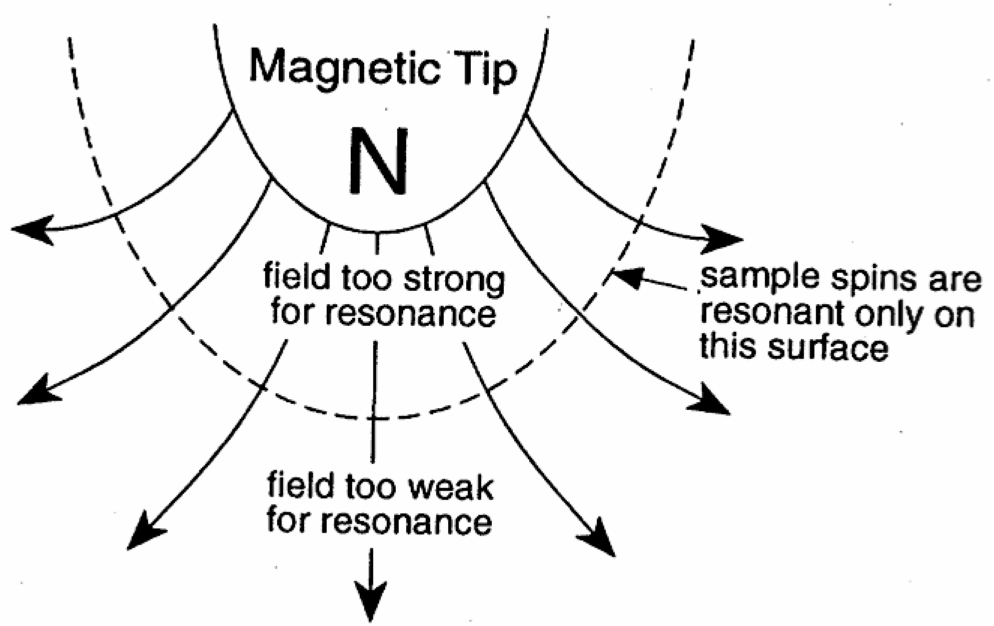


•**MRI** - motion of M_z induces an alternating voltage in the receiving coil with a frequency equal to the Larmor frequency

SIDLES JA, GARBINI JL, BRULAND KJ, et al.REVIEWS OF MODERN PHYSICS

MRFI - setup overview

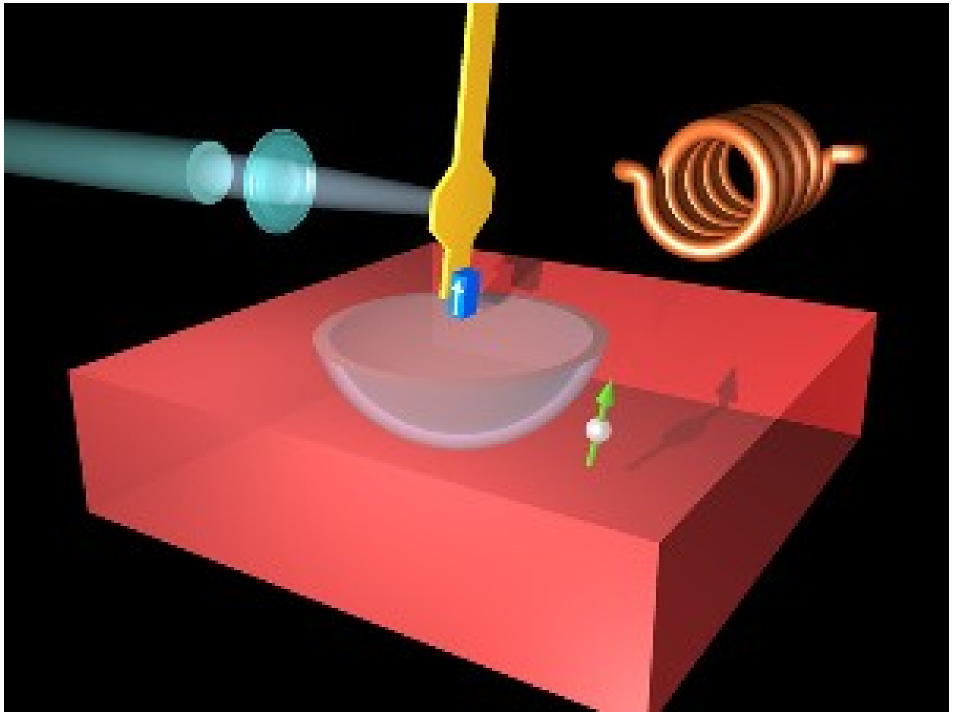
- A ferromagnetic tip is attached to a cantilever that is sensitive enough to bend in response to very small forces
- Apply a RF magnetic field at the Larmor frequency the magnetic moments of either the nucleus or electrons within a slice of the sample can be flipped up or down



SIDLES JA, GARBINI JL, BRULAND KJ, et al.REVIEWS OF MODERN PHYSICS

MRFI - setup overview

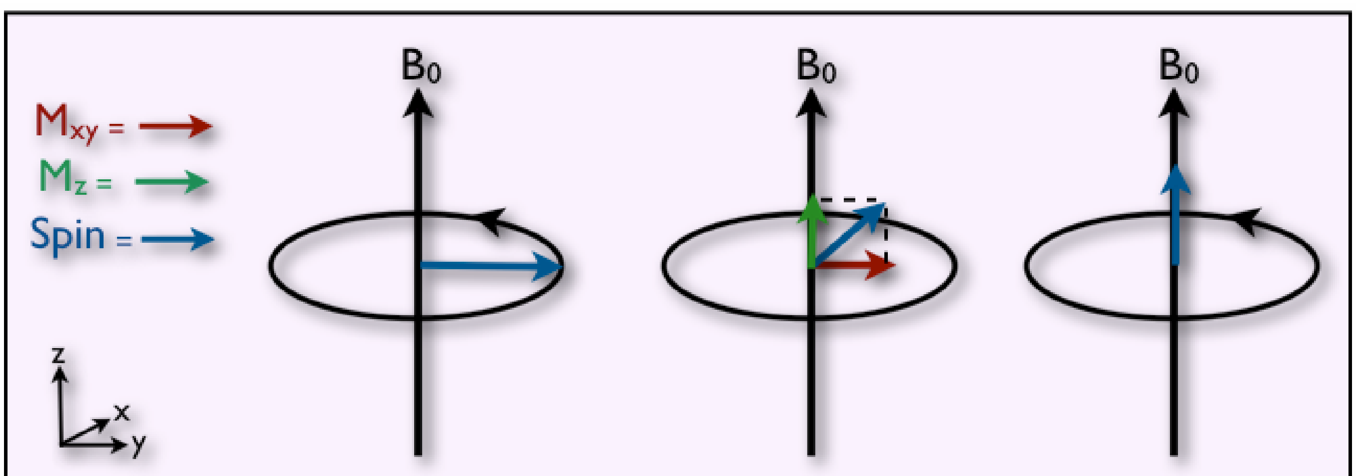
- This flipping generates an alternating force on the magnetic tip that causes the cantilever to vibrate
- Vibrations are detected using an interferometer



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

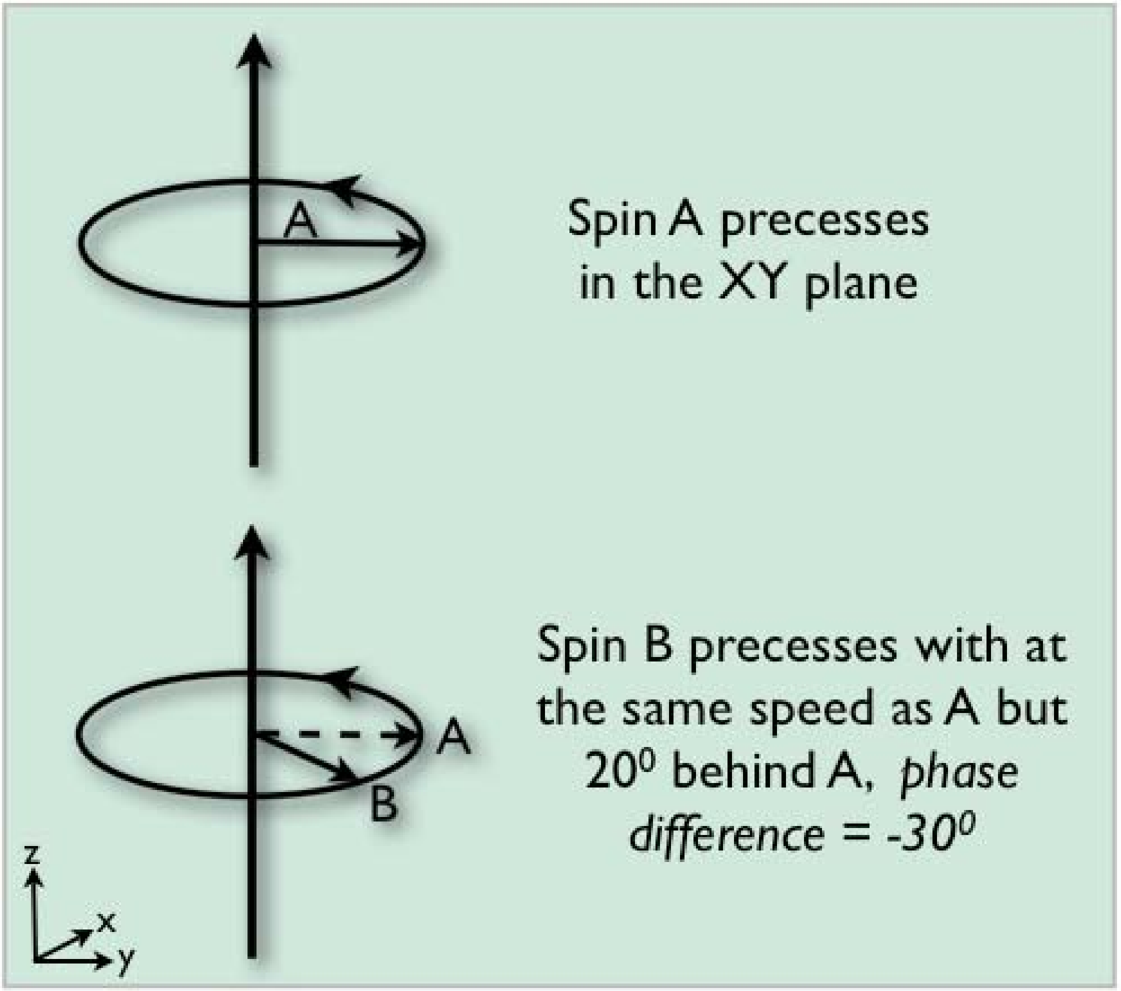


T1 - longitudinal relaxation

- 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

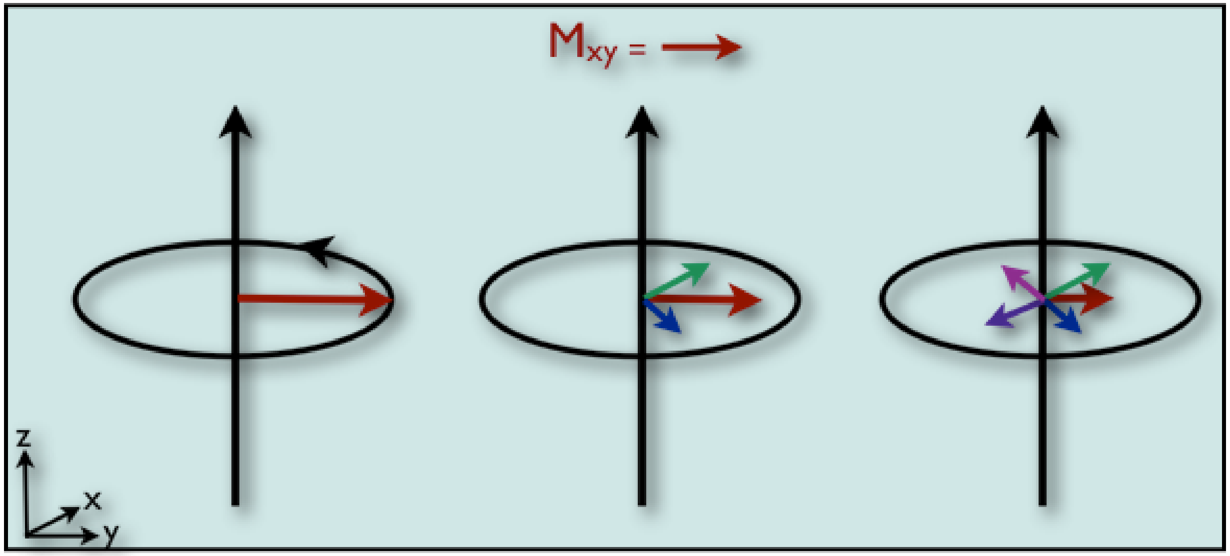
Phase

- Phase refers to an angle



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

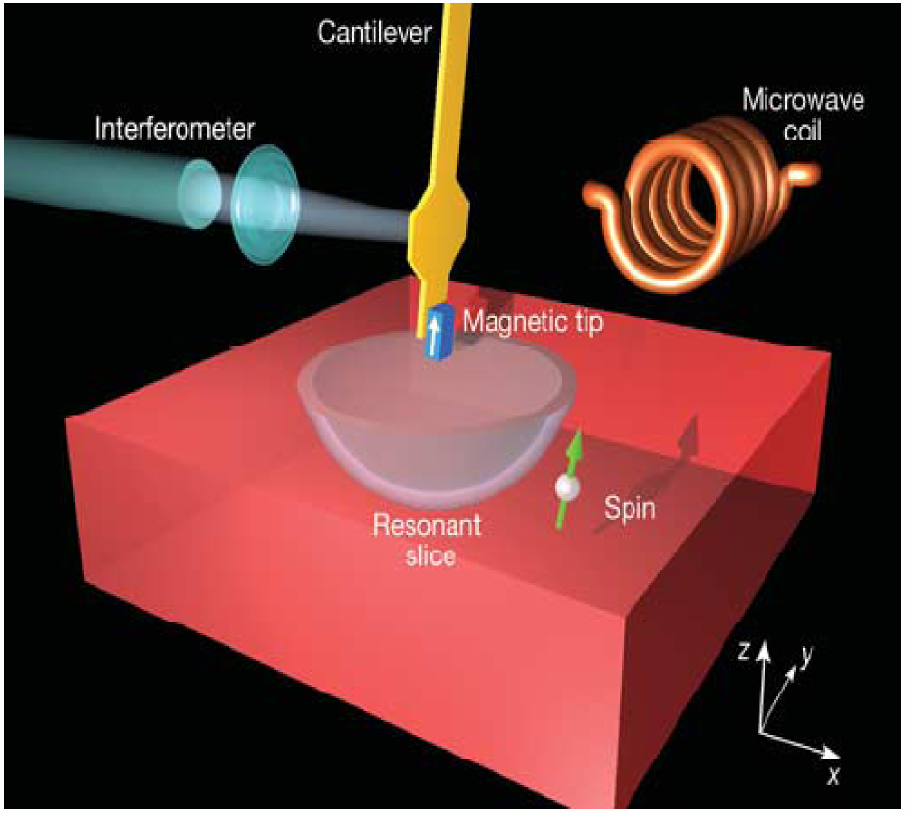
- The review article by Sidles et al. summarized the appeal of MRFM in 3 simple for very important points:
  - The magnetic imaging is non-contact and specific to electron and nuclear spins
  - The imaging magnetic field is 3-Dimensional and reaches below the scanned surface allowing for imaging of subsurface 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 electrons
  - 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 ($B_{tip}(x,y,z)$)
- A static magnetic field ($B_{ext}(z)$) 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

$$B_0(x,y,z) \equiv |\mathbf{B}_{\text{tip}}(x,y,z) + \hat{\mathbf{z}}\ B_{\text{ext}}| = \omega_{\text{rf}}/\gamma$$

$$\gamma/2\pi = 2.8 \times 10^{10}\,\text{Hz}\,\text{T}^{-1} \qquad \omega_{\text{rf}}/2\pi = 2.96\,\text{GHz}$$

  - $B0(x,y,z) = 106$ mT and $Bext = 30$ 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

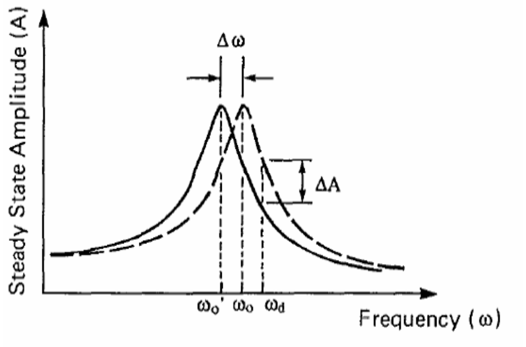
Need an improved detection method that increases sensitivity through high Q w/o decreasing BW

Slope Detection

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

$$\omega_0^2 = \frac{k_{eff}}{m} \qquad k_{eff} = k_L + \frac{\partial F}{\partial z}$$

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



Albercht, 1991

Slope Detection Limitations

Minimum detectable force gradient:

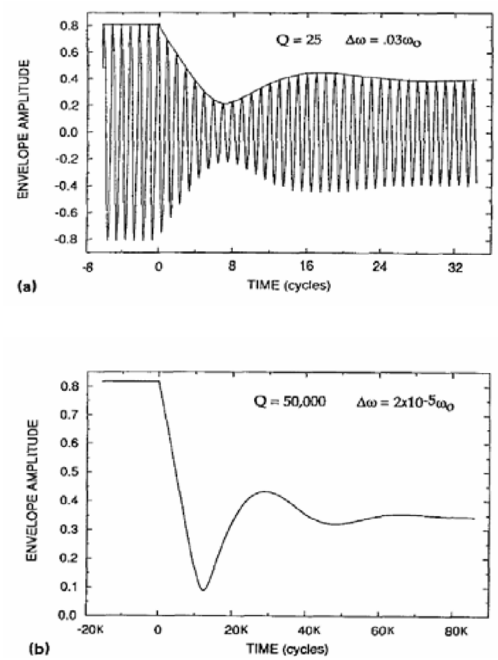
$$\delta F'_{\min} = \sqrt{2 k_L k_B T B / \omega_0 Q < z_{osc}^2 >}$$

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



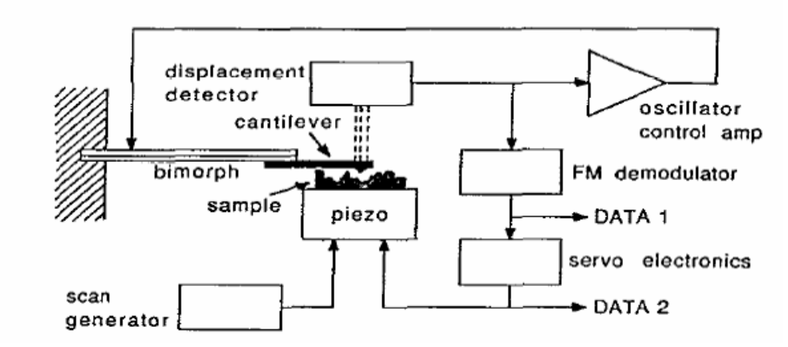
Albercht, 1991

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

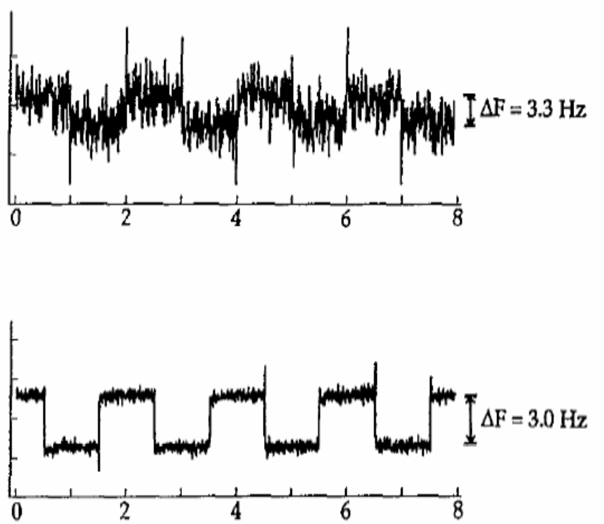
Comparison

Minimum detectable force gradient:

$$\delta F'_{\min} = \sqrt{4k_L k_B TB / \omega_0 Q < z_{osc}^2 >}$$

min detectable $\frac{\partial F}{\partial z}$ ~ same as slope detection method

- ✓ Similar sensitivity
- ✓ Independent BW and Q
- ✓ Increase sensitivity by higher Q w/o affecting BW



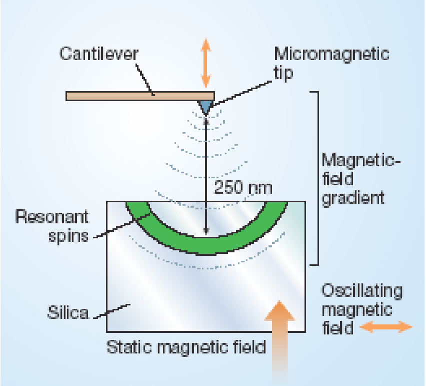
Albercht, 1991

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: $\Delta k \approx \frac{F_{spin}}{\Delta z}$

  F_{rms}: rms amplitude of oscillating force from spins

  Δz :rms cantilever amplitude
- change in oscillation frequency $\frac{\Delta f}{f} \approx (\frac{1}{2})(\frac{\Delta k}{k})$
- detected by analog frequency demodulator

OSCAR

- In other words, the alternating magnetic force on the cantilever mimics a change in cantilever stiffness:

$$\delta f_c = \pm \frac{2 f_c G \mu_B}{\pi k x_{peak}} \qquad G \equiv \frac{\partial B_0}{\partial x}$$

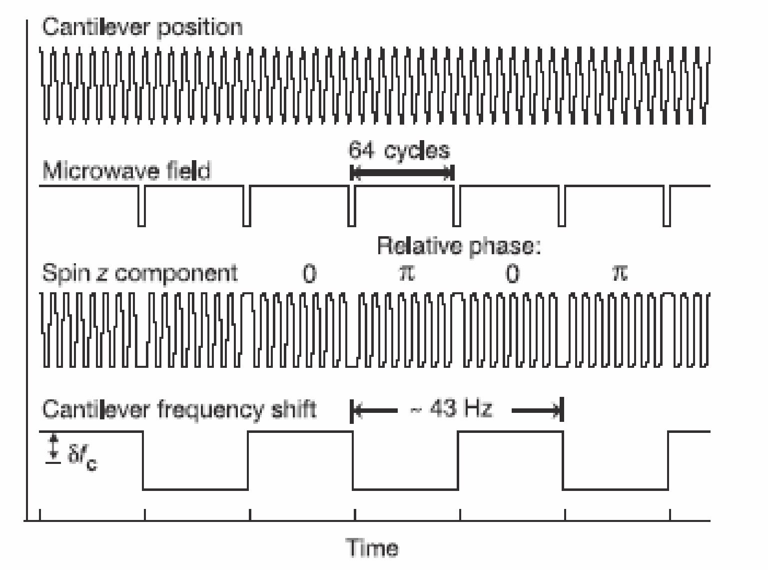
- sign of frequency change depends on relative phase of the spin inversion wrt the cantilever motion
- Rugar's experiment: $|\delta f_c| = 3.7 \pm 1.3 mHz$

Interrupted OSCAR

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

Interrupt B → relative phase of spin and cantilever reverses (frequency shift → reverse polarity) → frequency shift alternates between positive and negative values in a square-wave with

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



Rugar, 2004

iOSCAR Data Analysis

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

$$\Delta f(t) = \frac{4}{\pi} |\delta f_c| A(t) \sin(2\pi f_{sig} t) + higherharm\ onics$$

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

  $<A(t)>=0$, $<|A(t)|^2>=1$
- relatively large frequency noise of cantilever due to thermal motion and tip-sample interaction → 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.

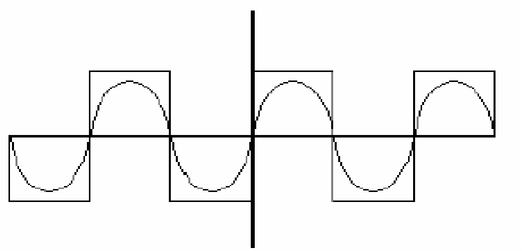
Lock in Amplifier:

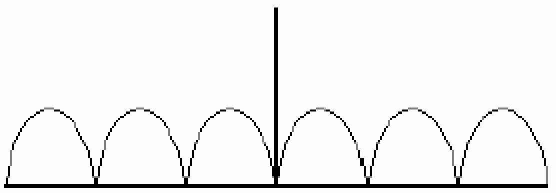
measures a small signal even in presence of noise

$$v_{in} = V_0 \sin(\omega t)$$

$$v_{sq} = \frac{4}{\pi}\left(\sin(\omega t) + \frac{1}{3}\sin(3\omega t) + \frac{1}{5}\sin(5\omega t) + \dots \right)$$

$$v_{out} = \frac{2V_0}{\pi}\left(1 - \frac{2}{3}\cos(2\omega t) - \frac{2}{15}\cos(4\omega t) - \frac{2}{35}\cos(6\omega t) \dots \right)$$





iOSCAR Data Analysis

- spin signal and measurement noise uncorrelated →

$$\sigma_I^2 = \sigma_{spin}^2 + \sigma_{noise}^2$$

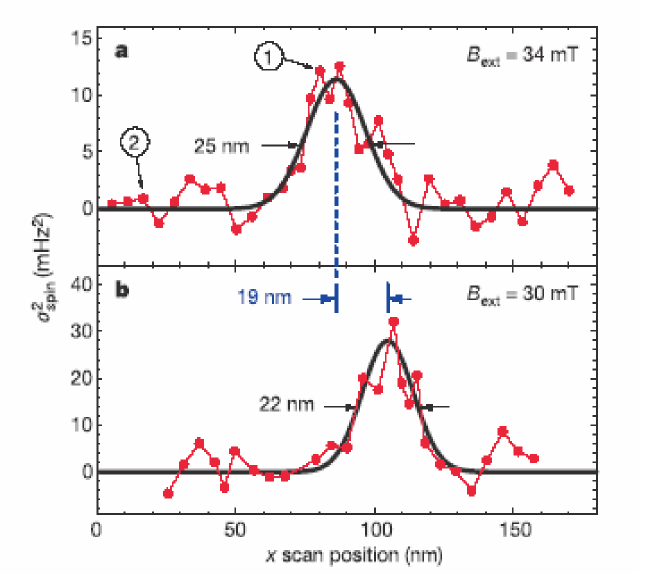
- quadrature variance contains noise data only →

$$\sigma_{spin}^2 = \sigma_I^2 - \sigma_Q^2$$

Lateral scan:

Peak → single spin

Low S/N: had to use considerable averaging

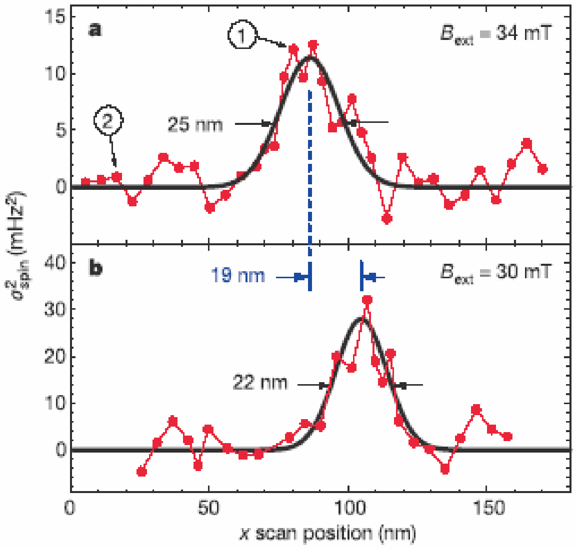


Rugar, 2004

Field Dependence of Spin Signal

- Reduce external field → shrink resonance slice → shift in scan position of signal peak
- B: 34 to 30 mT → peak shift of 19nm

  $\Delta B / \Delta x$ → G ~2x10^5 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 ¼ of the cantilever cycle
- signal disappeared when the interruption duration was a full cantilever cycle

Single Spin Detection

Spatial isolation of the spin signal → single spin

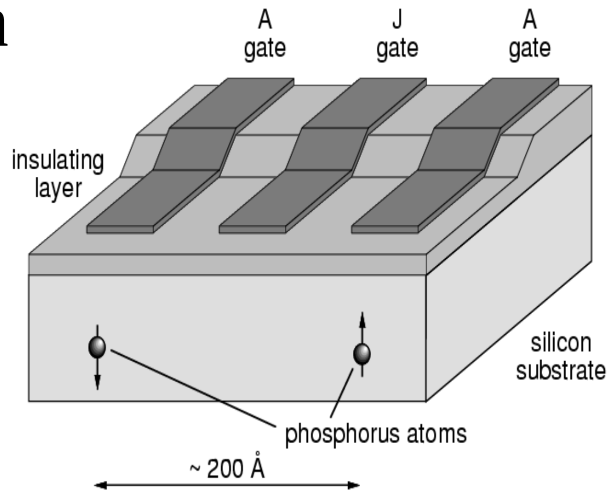
Low spin density: 10^{13} to 10^{14} cm^{-3}

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

A spin signal sample was scanned through ~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



B.E. Kane, *Nature* **393**, 133 (1998)

References

Albrecht, T. R., Grutter, P., Horne, D. Rugar, D. Frequency modulation detection using high-Q cantilevers for enhanced force microscopy sensitivity. J. Appl. Phys. **69**, 668-673 (1991).

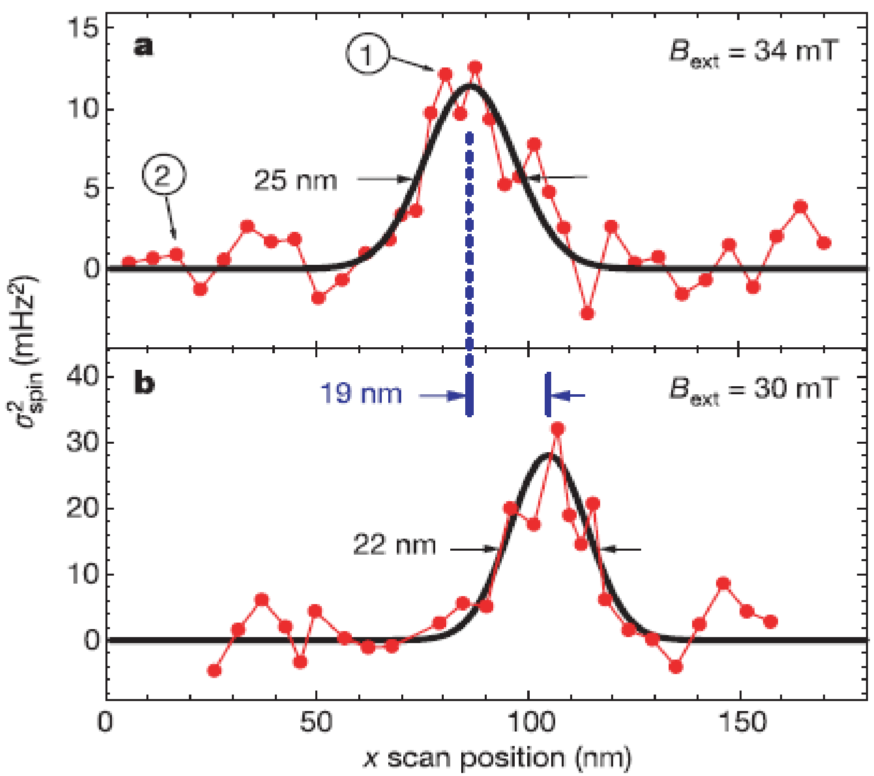
Hammel, P. C. Seeing single spins. Nature. **430**, 300-301 (2004).

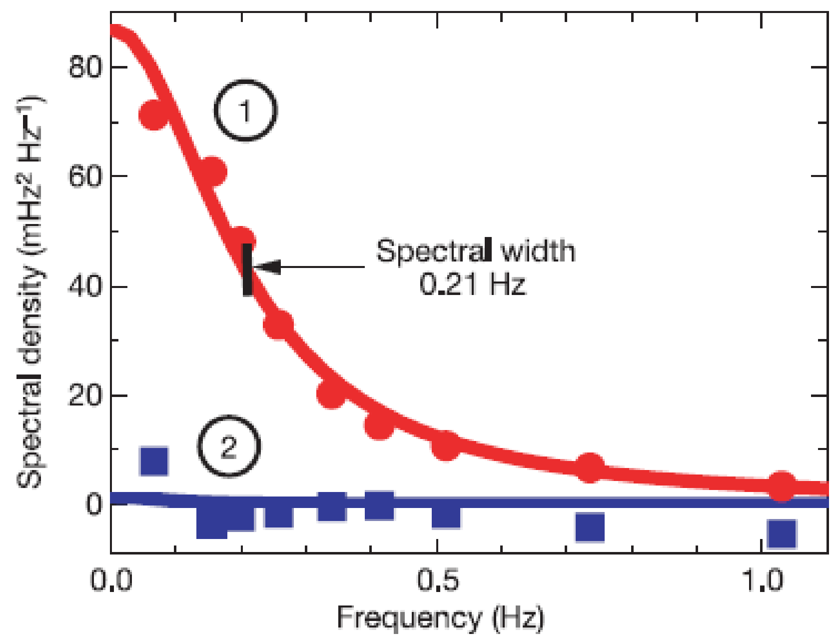
Rugar, D. Budakian, R. Mamin, H. J. Chui, B. W. Single spin detection by magnetic resonance force microscopy. Nature. **430**, 329-332 (2004).

Stipe, B. C., Mamin H. J., Yannoni C. S., Stowe T. D. , Kenny, T. W., Rugar, D. electron spin relaxation near a Micron-size ferromagnet. Phys. Rev. Lett. **87**, 277602 (2001).

Spectral Analysis

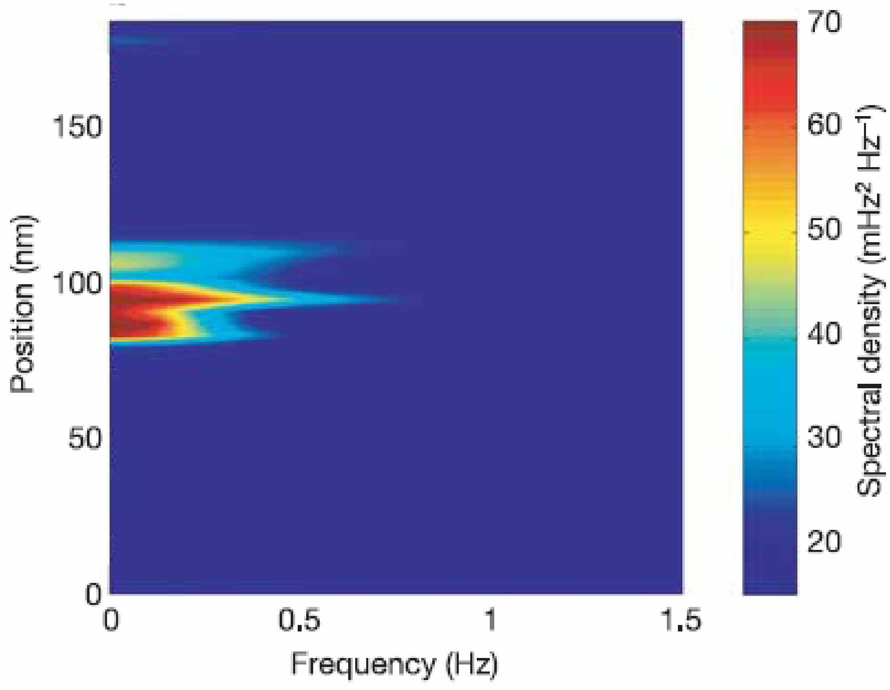
- The following plots are the result of 2 scans of the sample (laterally in the x direction) with 2 magnitudes of the external field





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) = $4\tau_m \langle [\Delta f_1(t)]^2 \rangle / [1 + 4\pi^2 \tau_m^2 f^2]$
- The spectral width at half-maximum was found to be 0.21 Hz and τ_m = 760ms
- The total magnitude of the spin signal (by integrating the spectrum): $\langle [\Delta f_1(t)]^2 \rangle = 28\,\mathrm{mHz}^2$
- Solving for $|\delta f_c|$ resulted in the value $|\delta f_c| = 4.2\,\mathrm{mHz}$ 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