Royal Holloway
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NMR on ^{3}He using DC SQUIDS

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ULTI III, Lammi, Jan04

Motivation

Detect precessing magnetization with a SQUID

Two "flavours"

tuned

High sensitivity

⇓

Study samples with low spin density

untuned

Broadband

⇓

NMR in ultralow magnetic fields

Outline

1. NMR and the use of SQUIDs - some history
2. Tuned and untuned input circuits
3. Operation of broadband SQUID amplifiers
4. Noise and sensitivity
5. Applications
6. Future prospects

Applications

1. Unconventional superfluidity in submicron ^{3}He slabs

(Measurement of the superfluid transition in a single slab resting on a polished silver surface)

2. Ferromagnetism of 2D solid ^{3}He

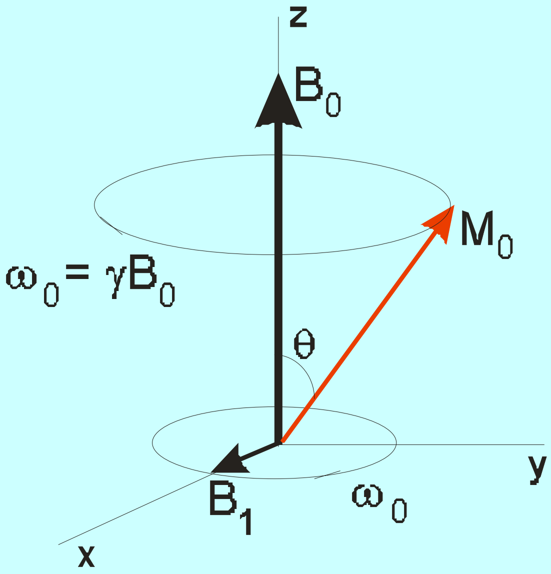
(Studies of ^{3}He adsorbed on graphite to low magnetic fields. Two dimensional frustrated magnetic system)

3. Knight shift in superconducting UPt_3

(To help determine equilibrium order parameter)

4. NMR in ultralow fields on room temperature samples

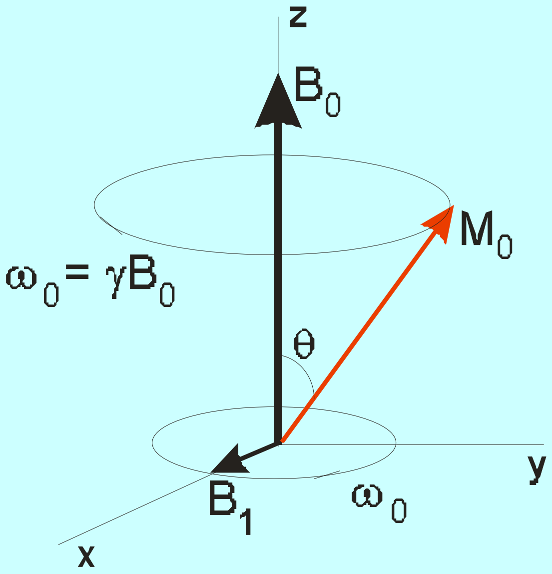
Detection of NMR using SQUID



1. Measure M_z

2. Measure M_y (free precession)

Detection of NMR using SQUID



1. Measure M_z

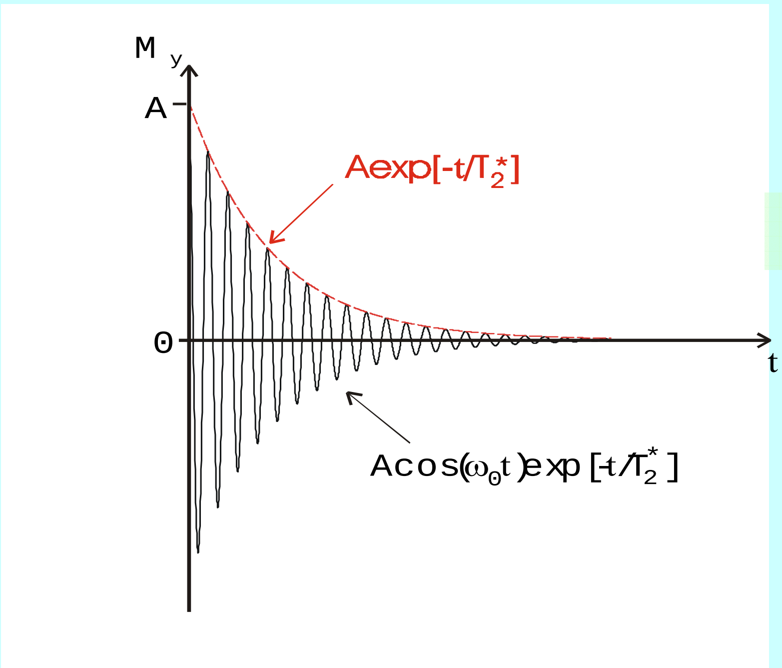
Typical bandwidth $1/T_1$

2. Measure M_y (free precession)

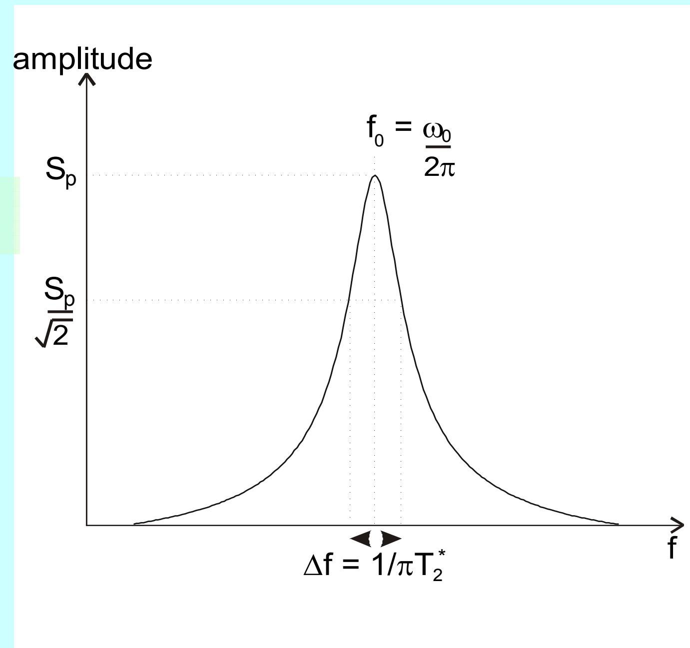
Typical bandwidth ω_0

Free induction decay

NMR lineshape



Fourier transform

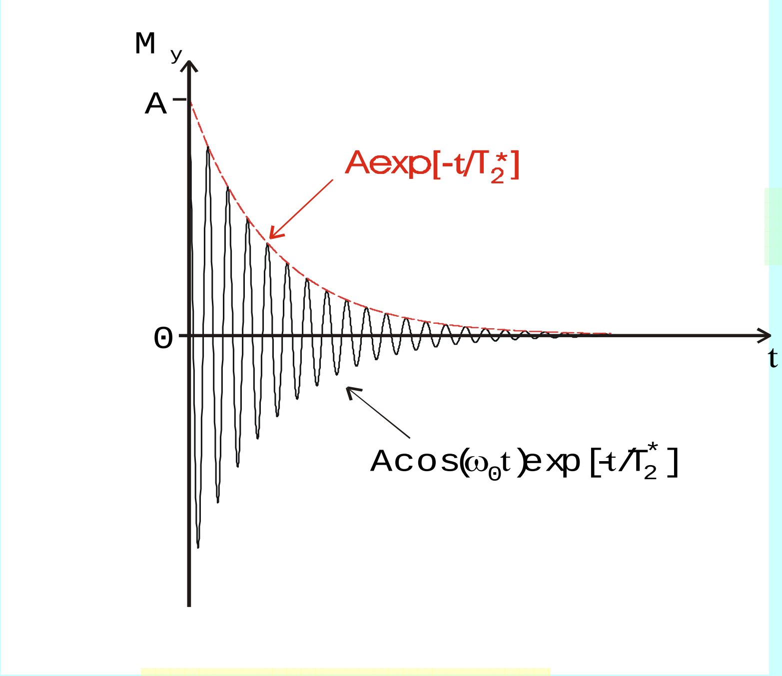


Time domain

Frequency domain

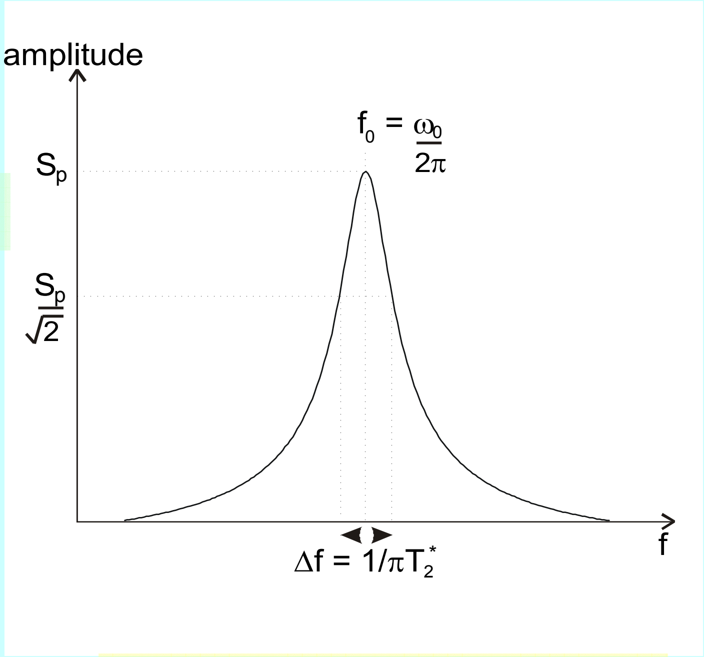
Free induction decay

NMR lineshape



Time domain

Fourier transform

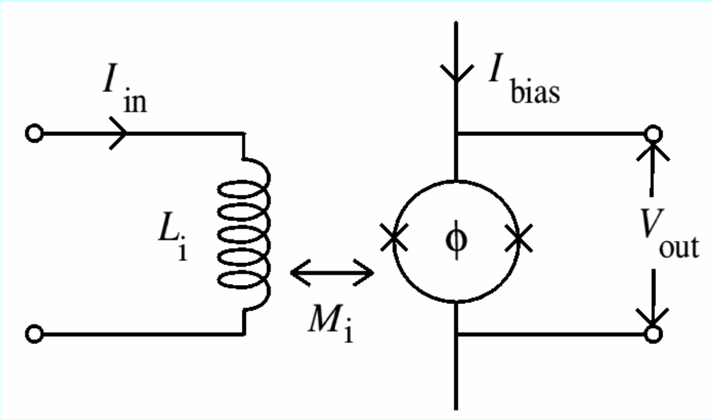


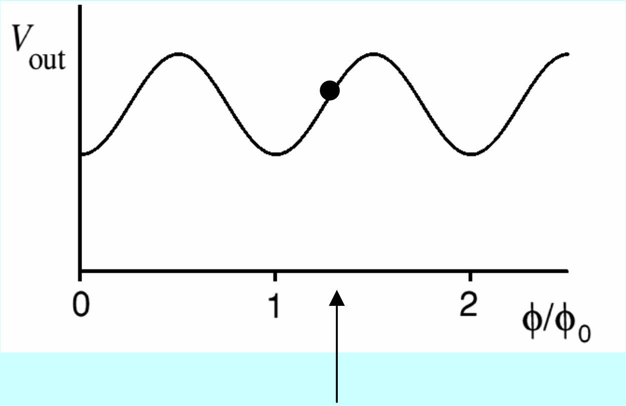
Frequency domain

T_2^* **determines** →

- Required signal capture time
- Noise bandwidth

DC SQUID converts magnetic flux to voltage



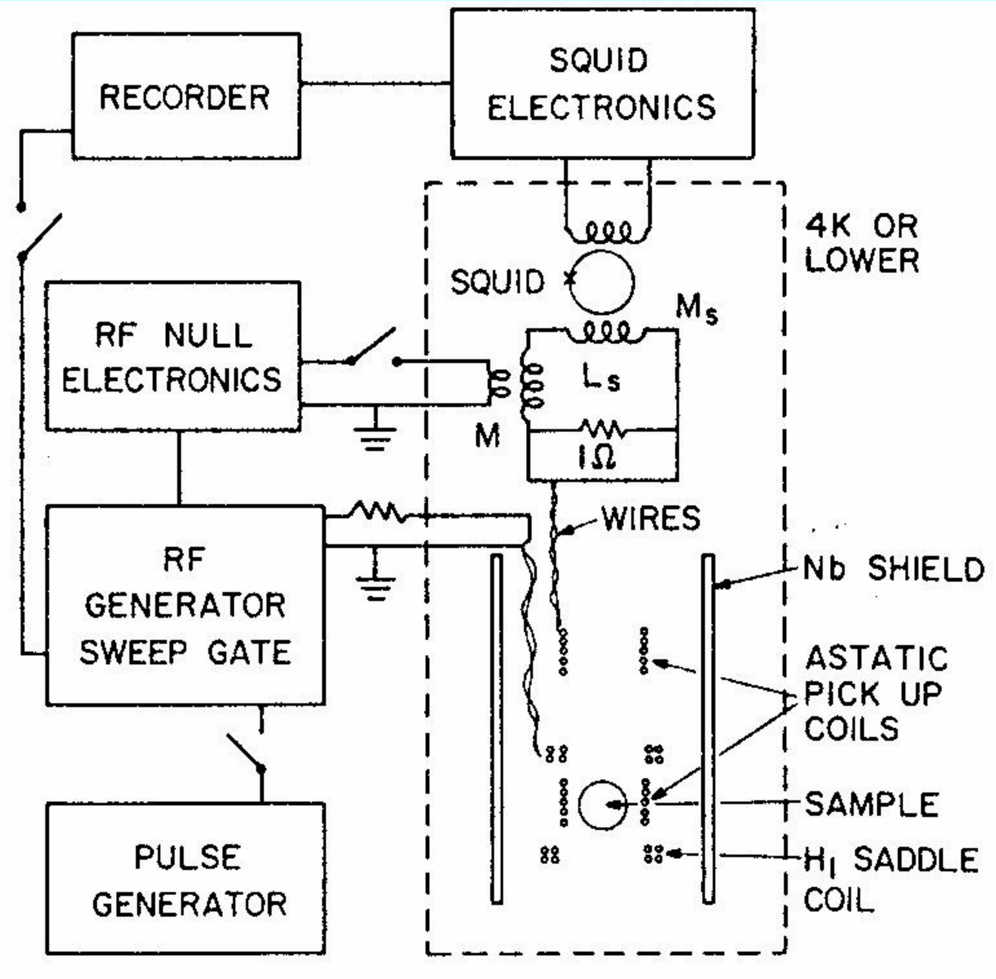


Modes of operation:

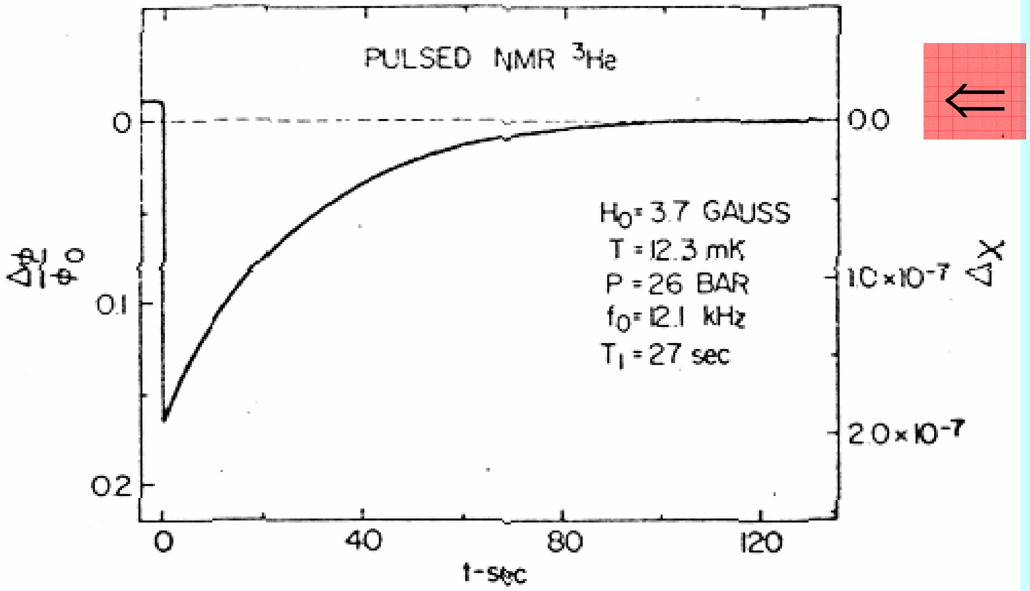
1. Small signal amplifier
   - small dynamic range
   - poor gain stability

2. Linear amplifier : Flux locked loop mode
   - wide dynamic range
   - good gain stability
   - reduced bandwidth

Detection of M_z



applications include: metals, ^{3}He
drug detection (eg. ^{14}N and ^{35}Cl in cocaine)

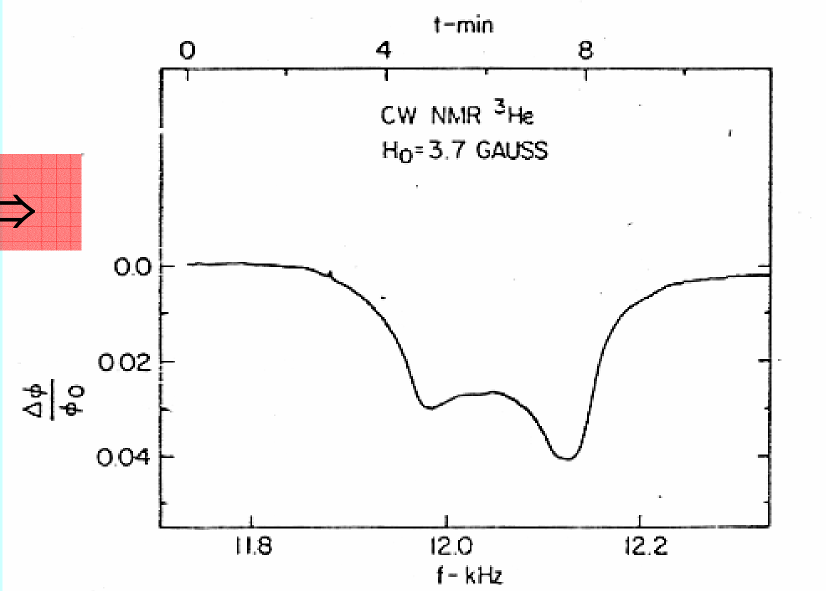


Measure M_z :
spin-lattice relaxation
after 180° pulse

R. A Webb Rev. Sci. Inst. 48 (1977) 1585

Sample :
Bulk liquid ^{3}He

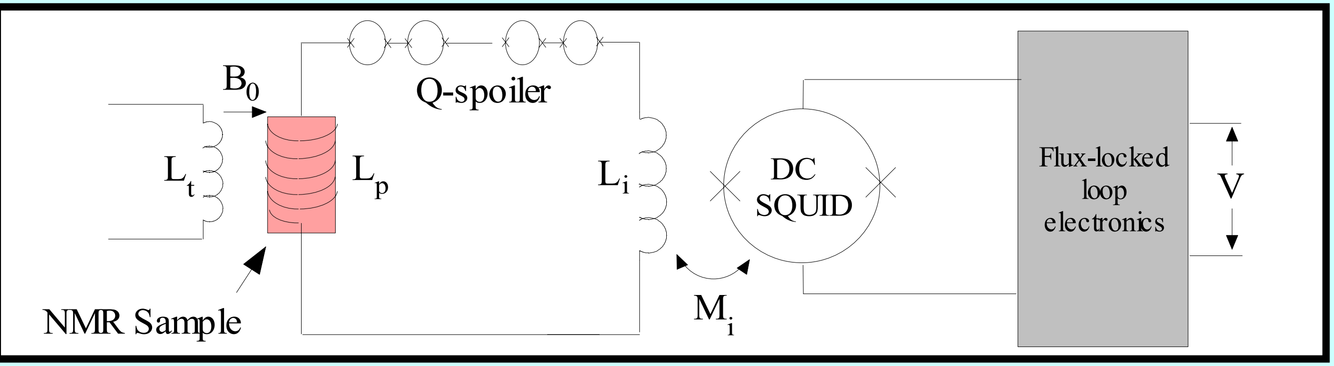
Measure M_z :
sweep frequency of transverse
rf field slowly through resonance



1. How do we couple NMR signal into SQUID?

2. How do we read-out voltage across SQUID?
(operation of flux locked loop)

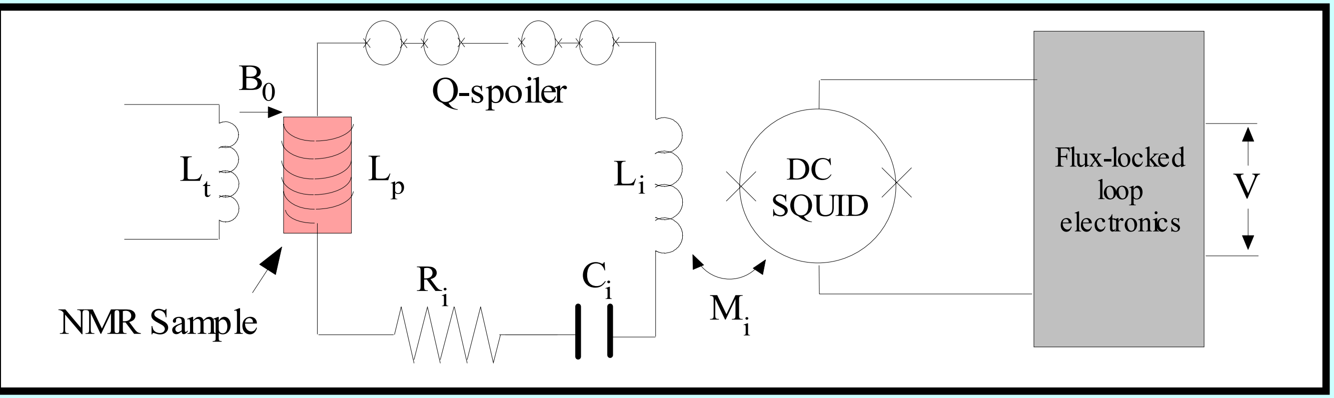
Broadband: superconducting flux transformer input circuit



Friedman et al.
Rev. Sci. Inst.
57 (1986) 410

Detect flux : signal ∝ magnetisation ∝ frequency

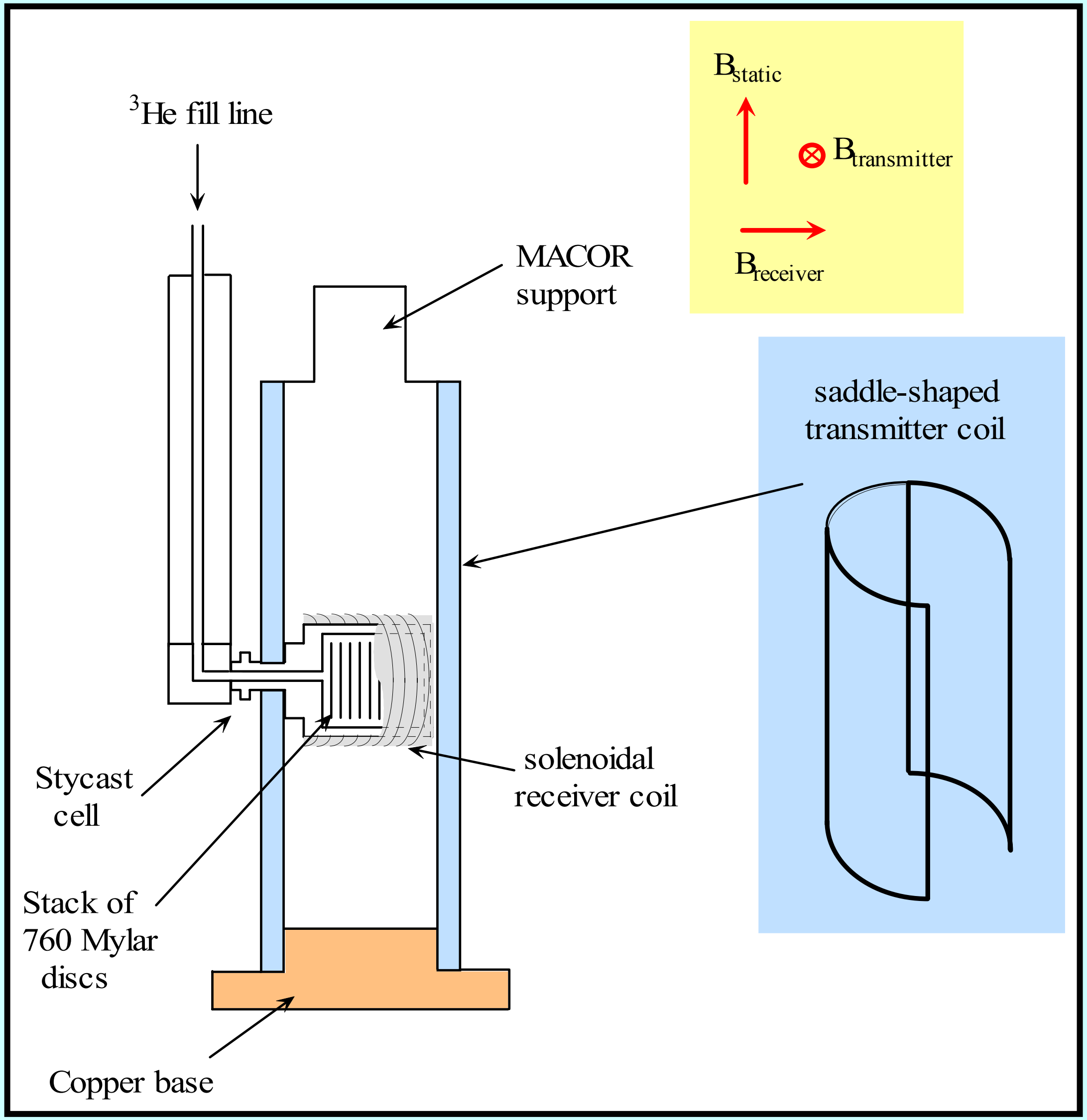
Tuned: series tuned input circuit



Freeman et al.
App. Phys. Lett.
46 (1986) 300

Detect rate of change of flux : signal ∝ (frequency)^2 × Q

Typical NMR coil set

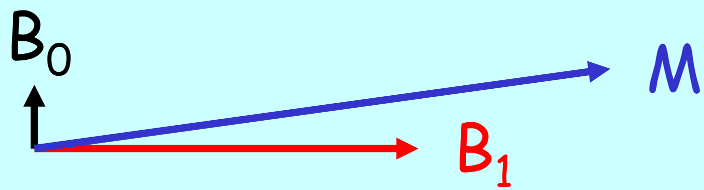


Orthogonal transmitter and receiver coils

Apply tipping pulse using saddle shaped transmitter coil

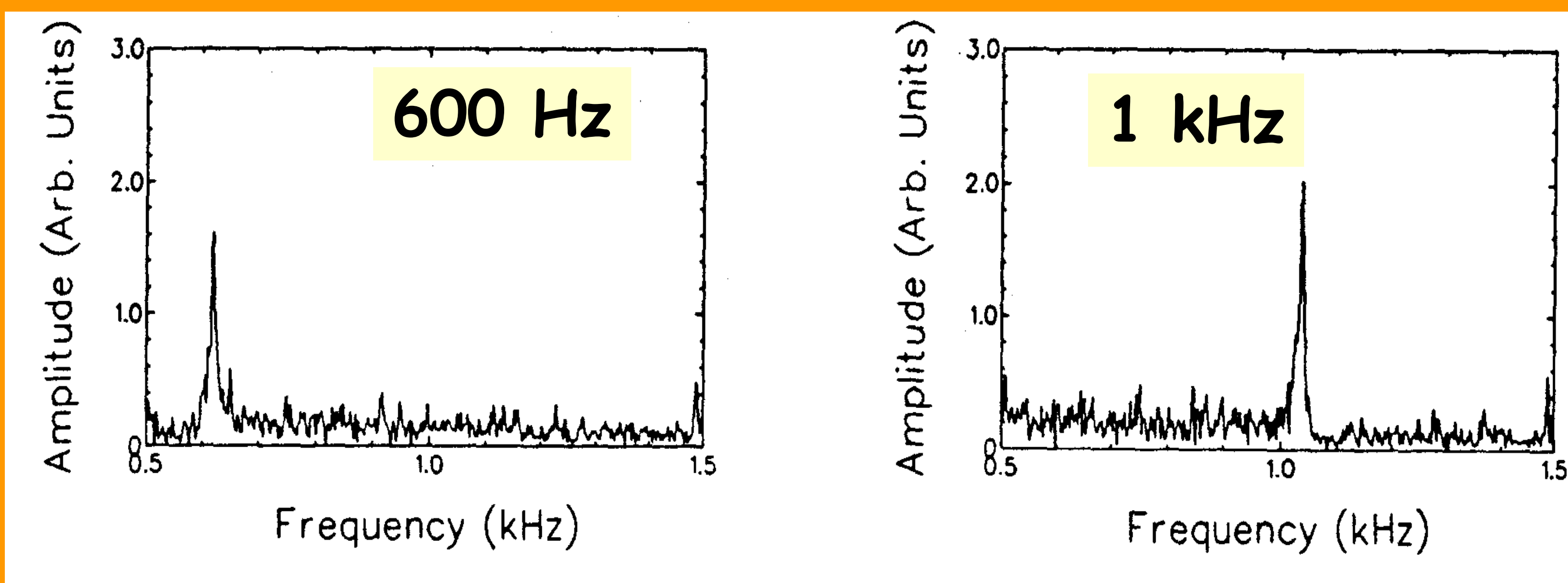
Receiver coil (saddle or solenoid) is mounted orthogonally to reduce cross coupling

1. Polarise by B_1 (= 6 G)
2. Turn off B_1
3. Spins precess around B_0 (= 0.19 or 0.32 G)

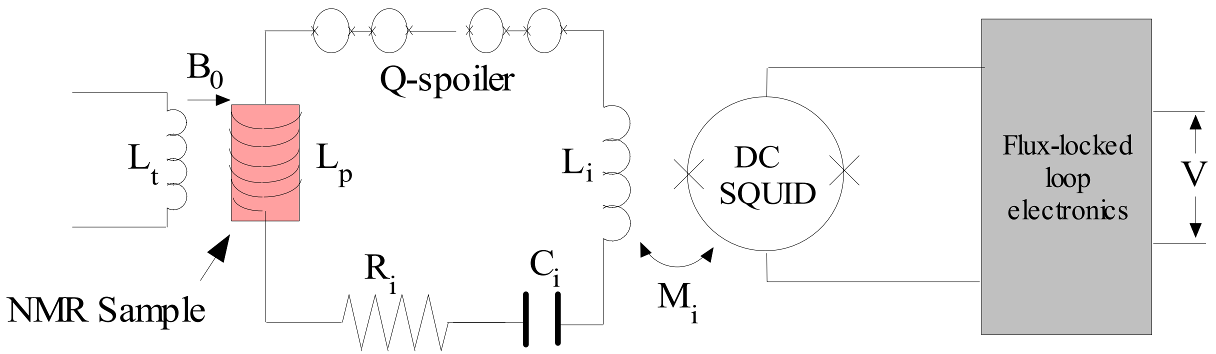


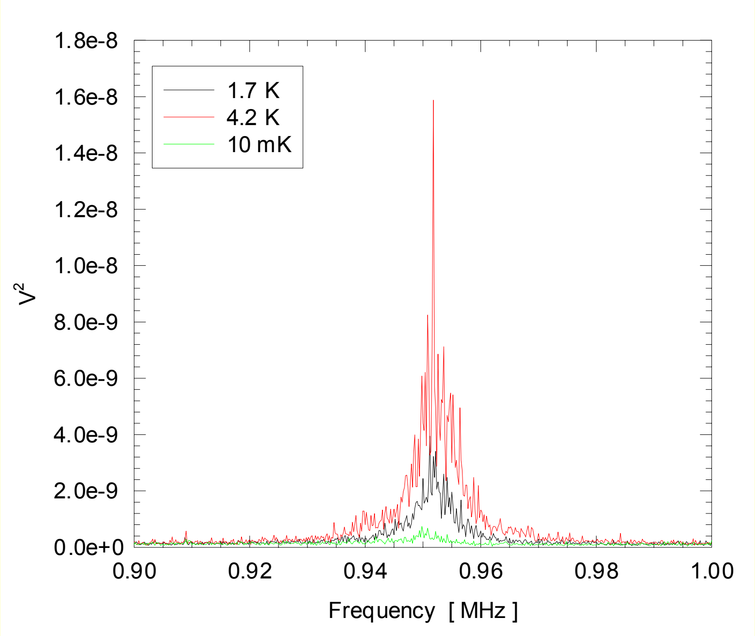
Ultralow frequency NMR on ^{3}He gas at 4.2K

using commercial (Quantum Design) SQUID with 50 kHz bandwidth



Tuned spectrometer: optimising noise





Johnson noise peak as a function of sample coil temperature

Noise temperature T_N.

At $T_N \Rightarrow$ SQUID amplifier noise = Johnson noise from input circuit

Optimising noise temperature

SQUID energy sensitivity

$$\varepsilon = \frac{\langle \phi_N^2 \rangle}{2L_s}$$

$$(T_N)_{opt} = \frac{\varepsilon \omega_0}{k_B}$$

when $x = \frac{\omega_0 M_i^2}{R_i L_s} = 1$

$$Q \frac{M_i^2}{(L_p + L_s)L_s} = 1$$

For W9M SQUID $\langle \phi_N^2 \rangle^{1/2} = 1.1$ $\mu\phi_0/\sqrt{Hz}$; L_s=210 pH

$\Rightarrow \varepsilon$ = 20 h

$\Rightarrow$ $(T_N)_{opt}$ = 6.0 mK

More completely: $T_N = \sqrt{K} \frac{(T_N)_{opt}}{2} \left(\frac{1}{x} + x \right)$

Conventional flux-locked loop scheme

- Cryogenic output transformer
- Flux modulation

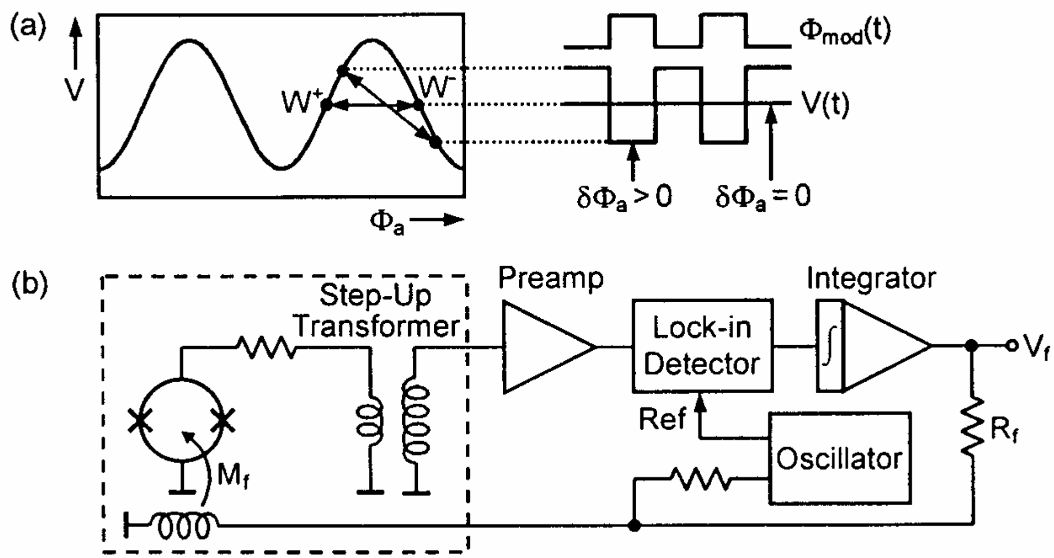
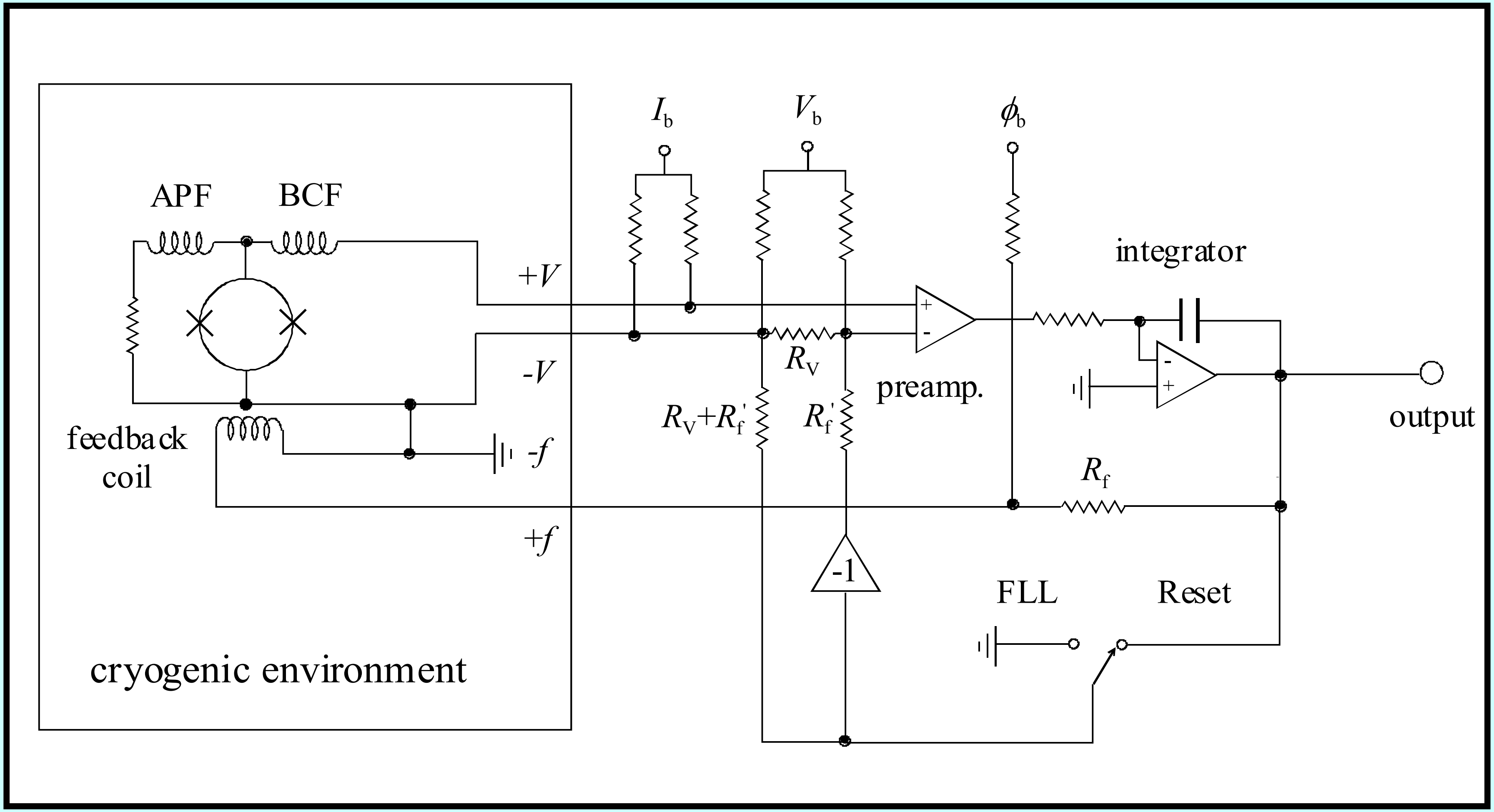


Fig. 4.3. SQUID readout with flux modulation: (a) V-Φ_a characteristic and (b) FLL circuit. A square-wave modulation flux Φ_{mod} switches the SQUID periodically between the working points W^+ and W^-. Components inside the dashed box are at cryogenic temperature.

Drung and Mück

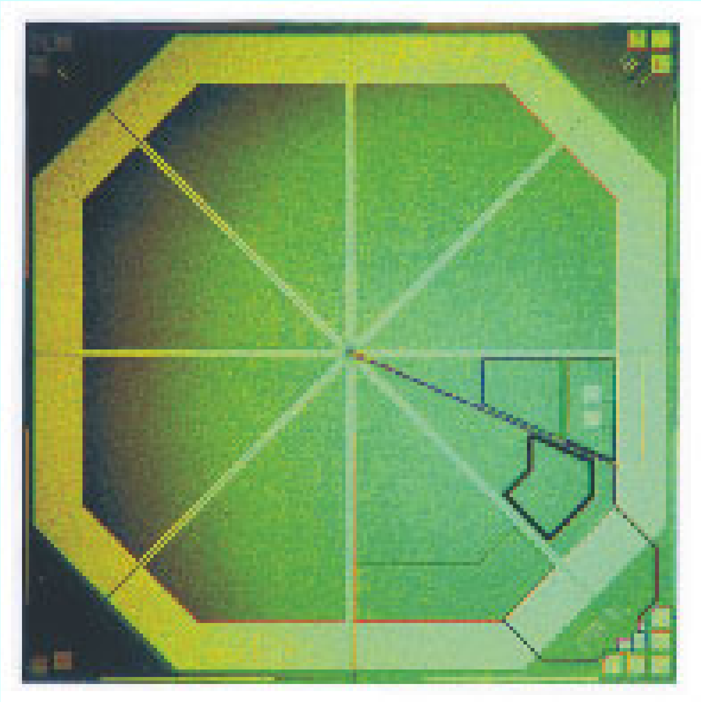
Flux locked loop

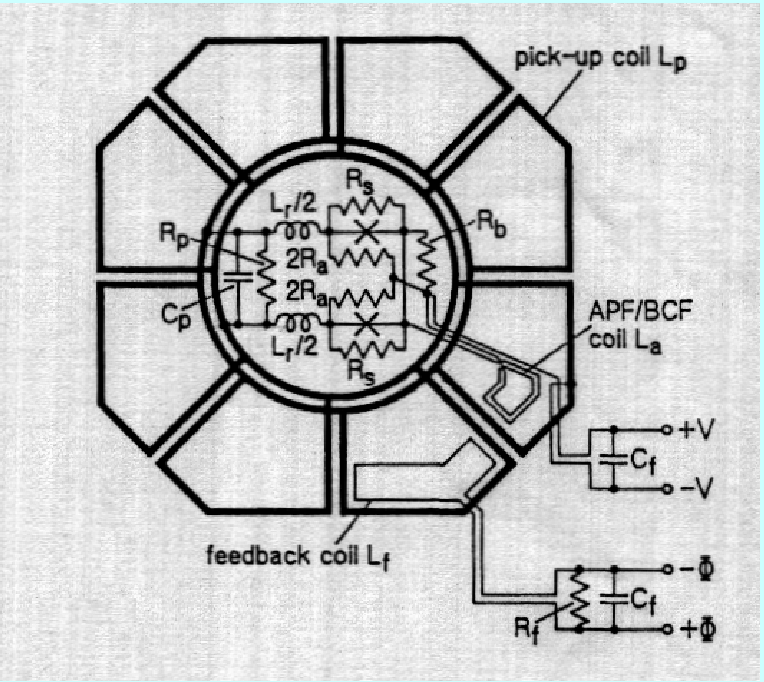
- SQUID output coupled directly to room temperature amplifier
- SQUID characteristic modified by APF



Additional **P**ositive **F**eedback : D Drung et al.
App. Phys. Lett. 57 (1990) 406

PTB SQUID





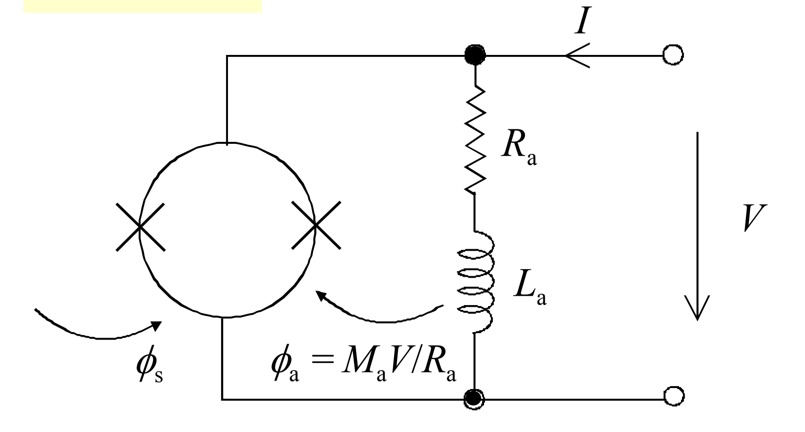
SQUID constructed from 8 parallel loops
⇒ combines large area with low inductance

J Zimmerman
J.App.Phys. **42**, 4483 (1971)

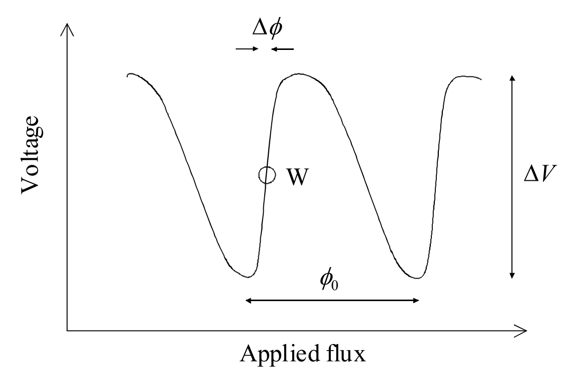
High field sensitivity (biomagnetic measurements)
⇒ "flip-chip" input coil

Noise and Bandwidth

APF coil



Skewed characteristic



$$\left\langle \phi_N^2 \right\rangle_{effective} = \left\langle \phi_N^2 \right\rangle + \frac{\left\langle V_N^2 \right\rangle}{V_\phi^2}$$

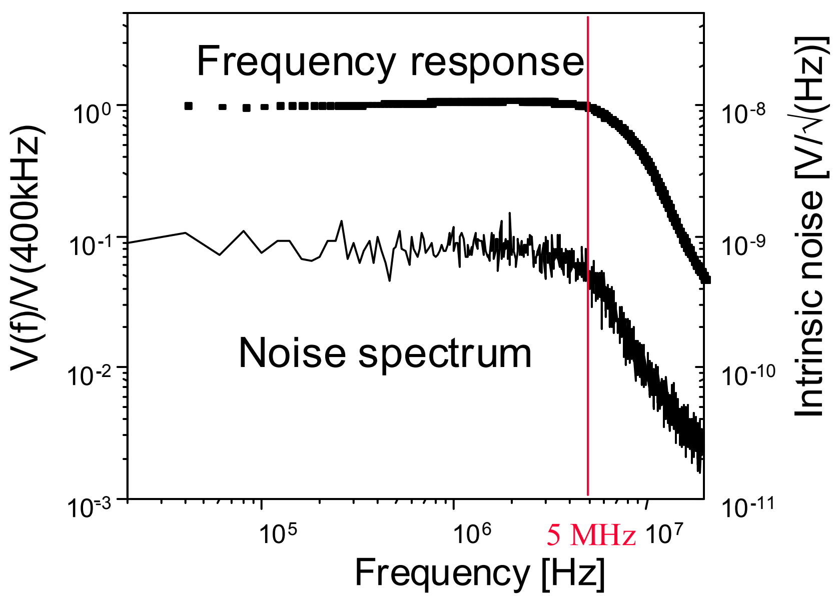
$$\left\langle \phi_N^2 \right\rangle^{1/2} = 1.1 \ \mu\phi_0/\sqrt{\text{Hz}}$$

$$\left\langle V_N^2 \right\rangle^{1/2} = 0.45 \ \text{nV}/\sqrt{\text{Hz}}$$

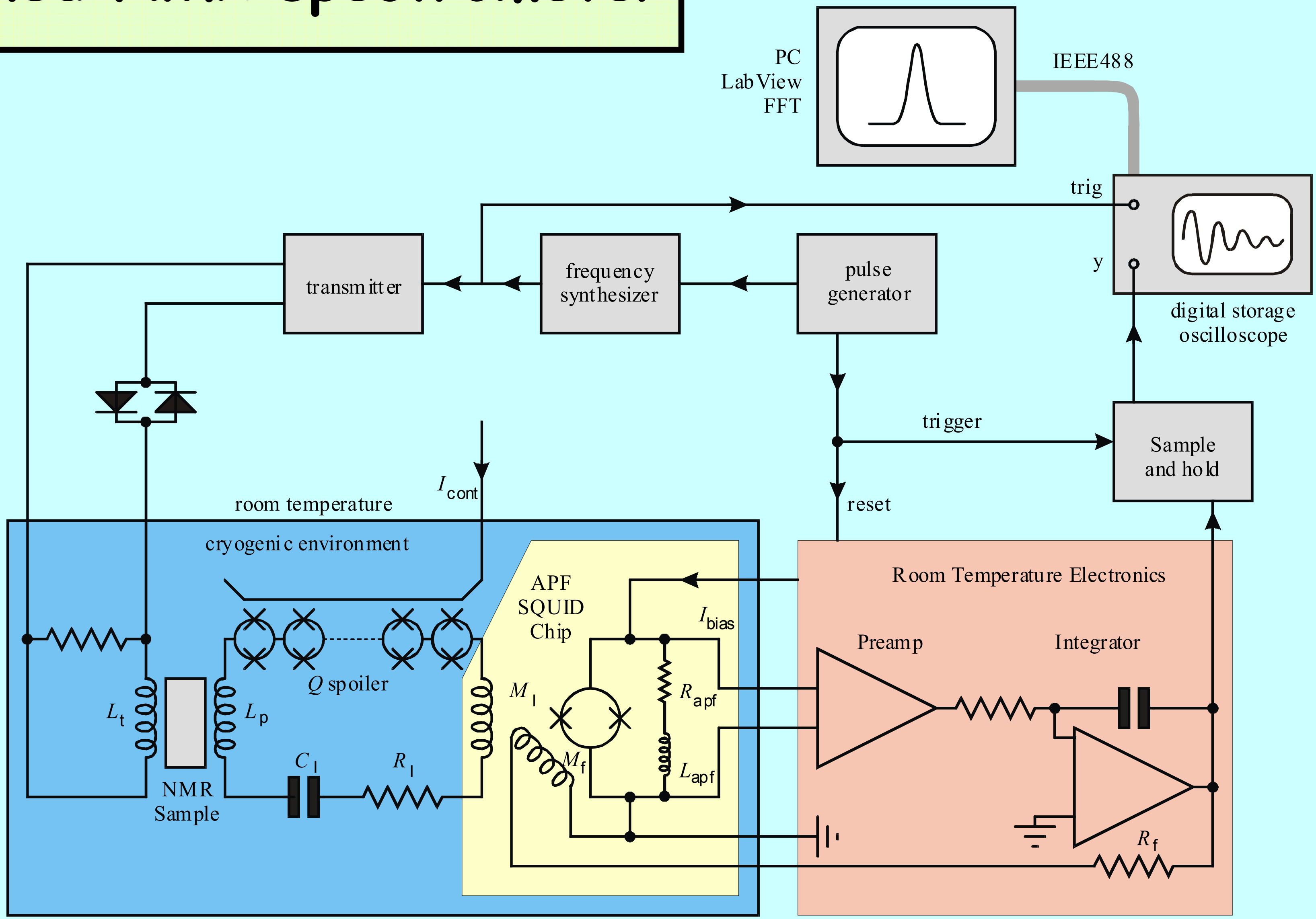
$$V_\phi = 415 \ \mu\text{V}/\phi_0$$

SQUID noise = amplifier noise

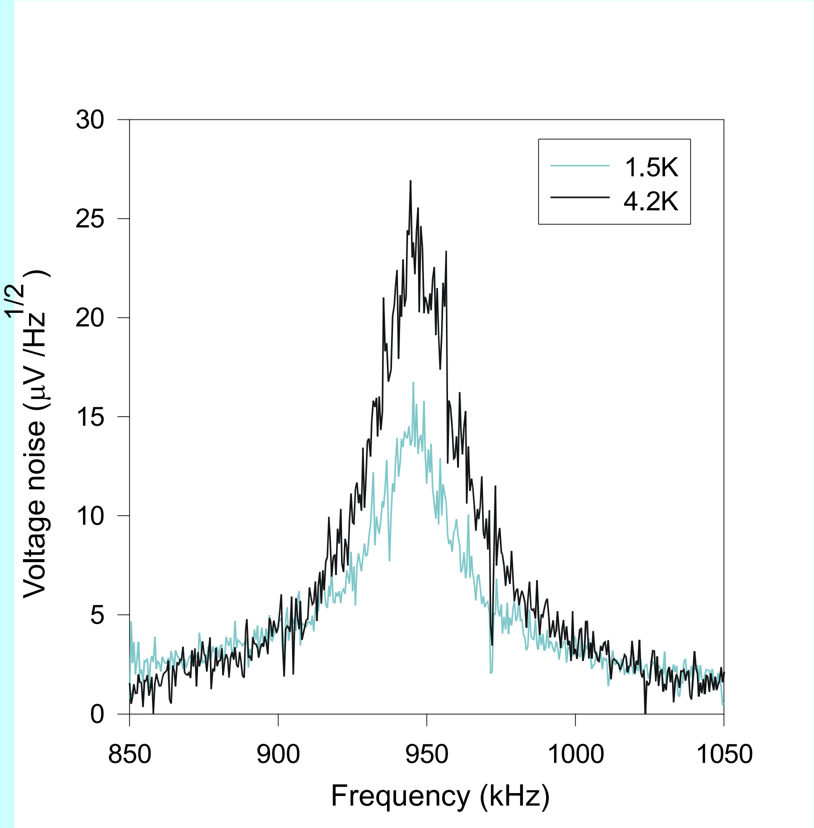
Typical bandwidth = 5 MHz

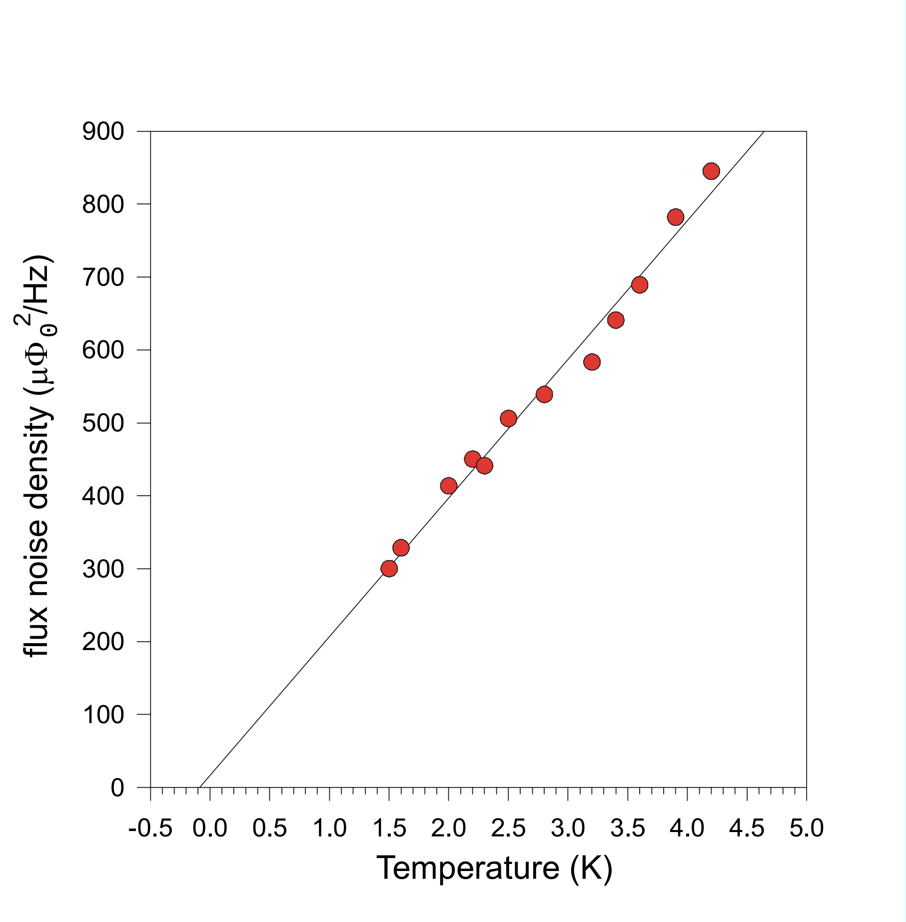


Tuned NMR spectrometer



Estimate of achieved noise temperature from tests to 1.5 K





Some headaches with tuned spectrometers

gain, bandwidth and effective Q depended in an unexpected way on feedback resistor, R_f: Introduce weak feedback directly into input circuit.

Spurious noise source limits noise temperature. Model by an effective resistance at some T_{eff} in input circuit. Study at 4K using a high Q tuned circuit (superconducting coil).

Appears to arise from coupling to APF coil. Removing APF coil, we can achieve T_N ~30 mK under optimum conditions

Background resonances

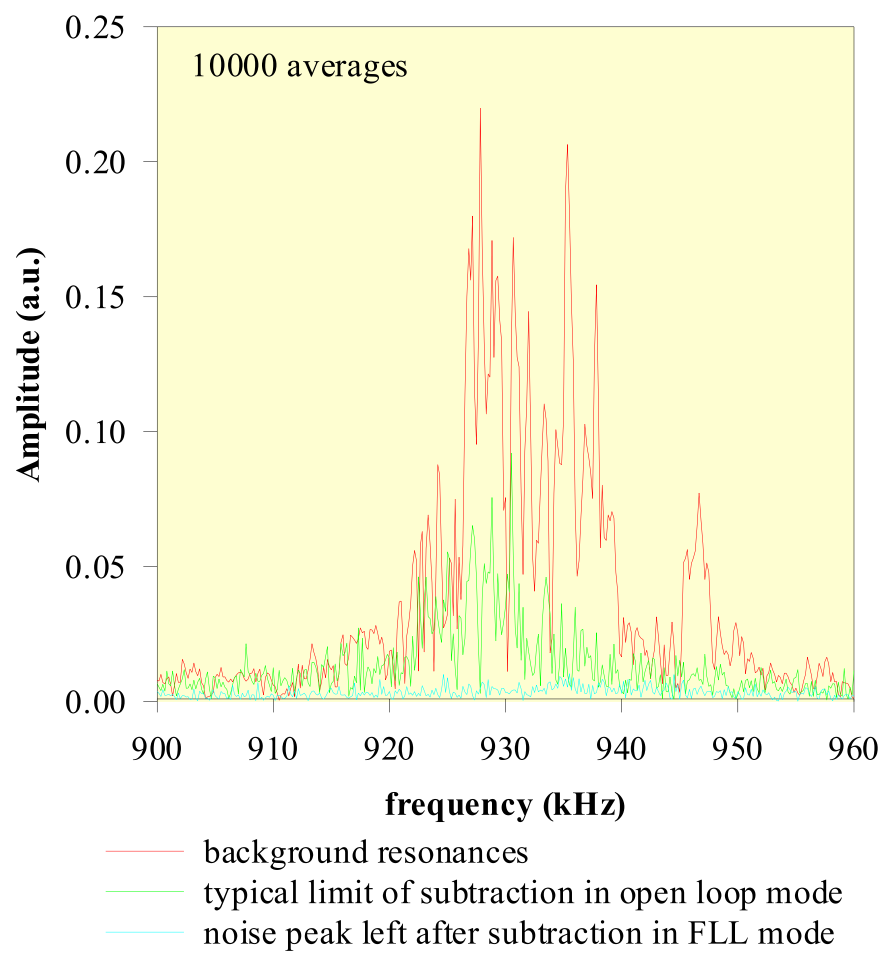
cf “coil disease” in conventional NMR

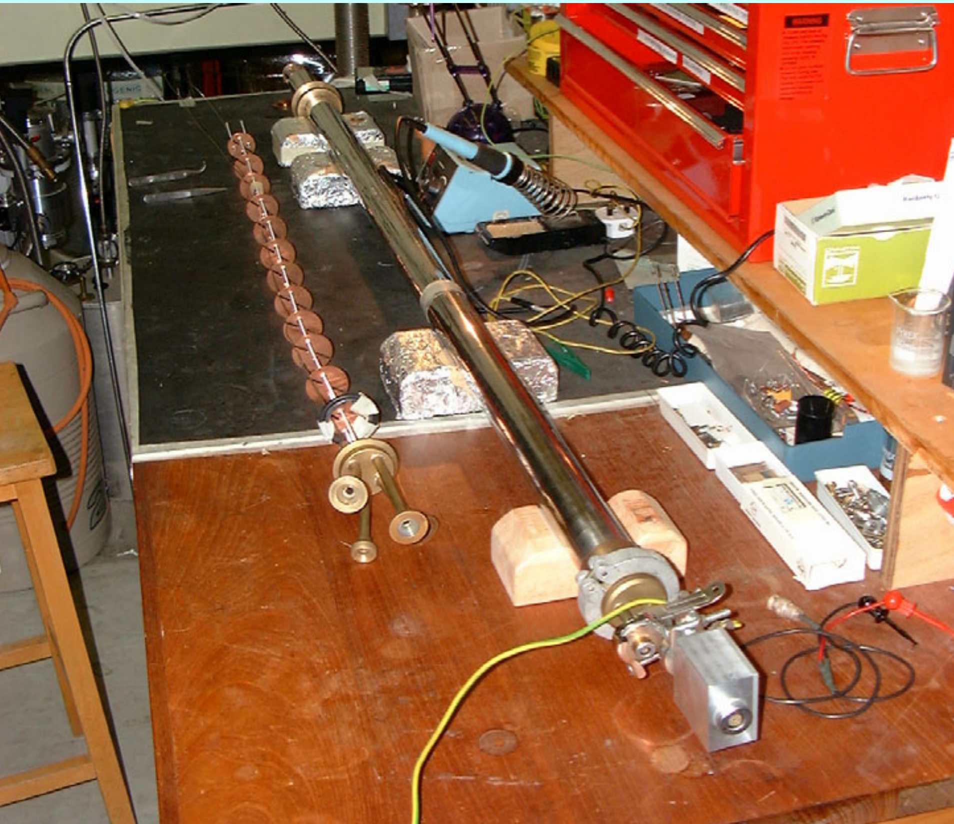
A coherent background signal due to magneto-acoustic resonances is observed.

Subtracted by taking magnetic field “off-tune”.

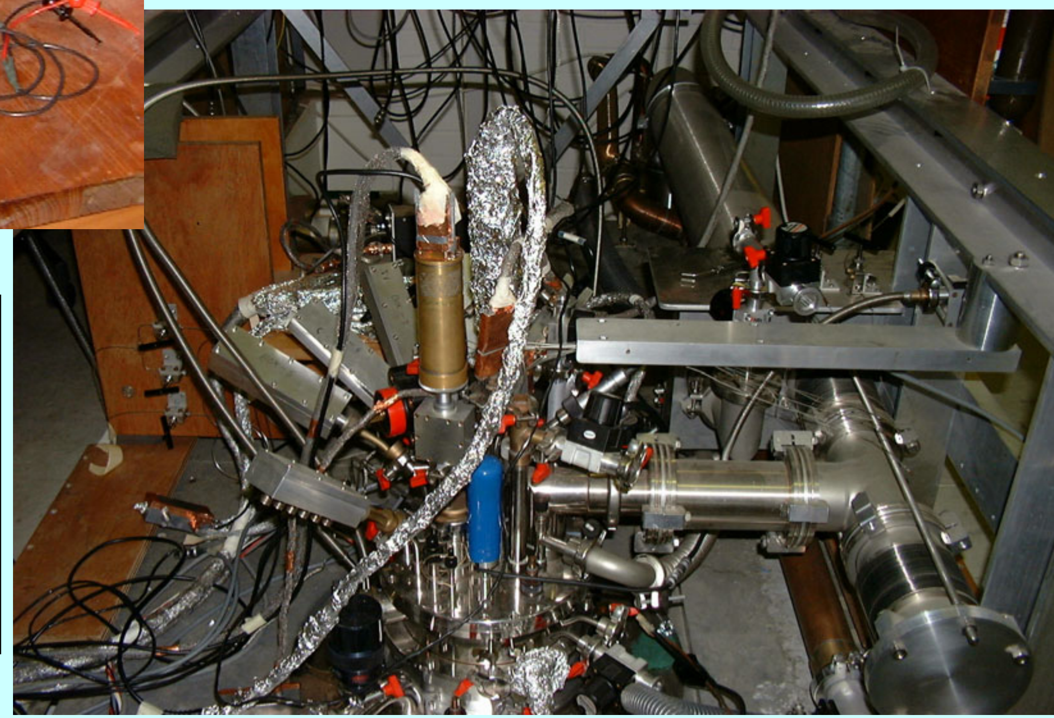
↑

High gain stability achieved by operation in FLL mode is crucial

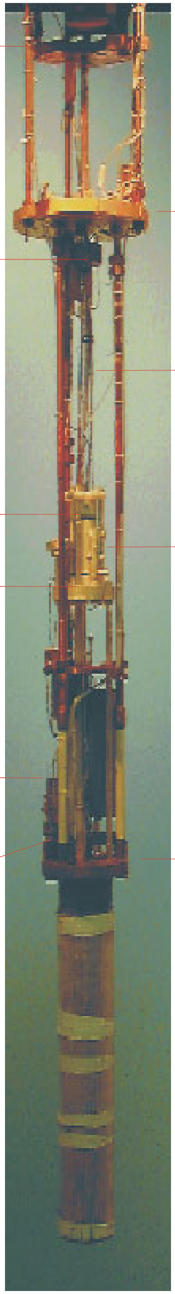




Assembly of SQUID amplifier probe
for test at 4.2K



Fully assembled probe installed in cryostat, through one of three 50mm diameter line of sight access ports



Nuclear Demagnetisation Cryostat for SQUID NMR and other things

Calculating the NMR signal size

Principle of reciprocity

Flux coupled to sample coil

$$\phi_p = \int_{sample} \underline{B}_1 \cdot \underline{M}_{xy}\, dv$$

Integrate over sample volume

B_1 = field per unit current of sample coil

Hoult and Richards, *J. Mag. Res.*, **24**, 71 (1976)

⇓

Estimate minimum number of detectable spins

Minimum number of detectable spins

Peak signal = peak noise in frequency domain, in a single shot.

$$N_m = K_N^F \left(\frac{4k_B T_{spin}}{\hbar\omega_0}\right)\left(\frac{4k_B T_{sys}}{\hbar\omega_0}\right)^{1/2}\left(\frac{1}{Q}\right)^{1/2}\left[\frac{\hbar\left(\frac{2K_T}{T_2^*}\right)L_p}{\hbar^2\gamma^2\left\langle B_1^2\right\rangle}\right]^{1/2}$$

$$T_{sys} = T_{coil} + T_N$$

Minimum number of detectable spins

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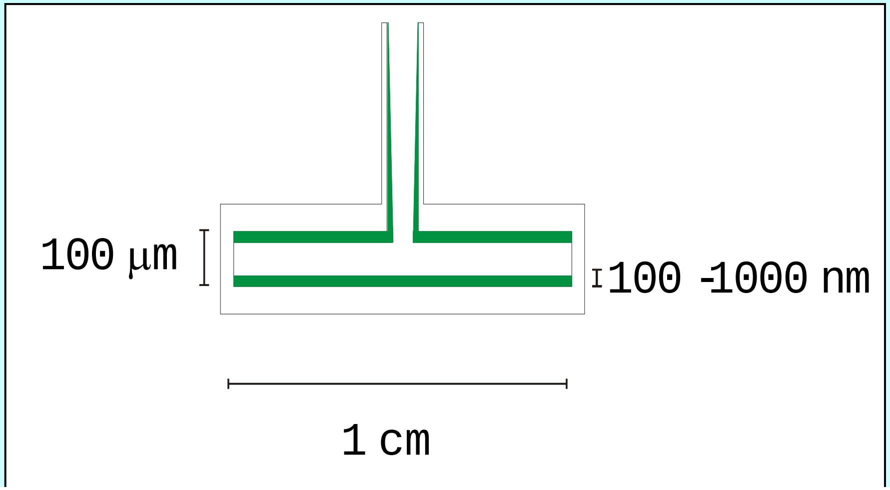
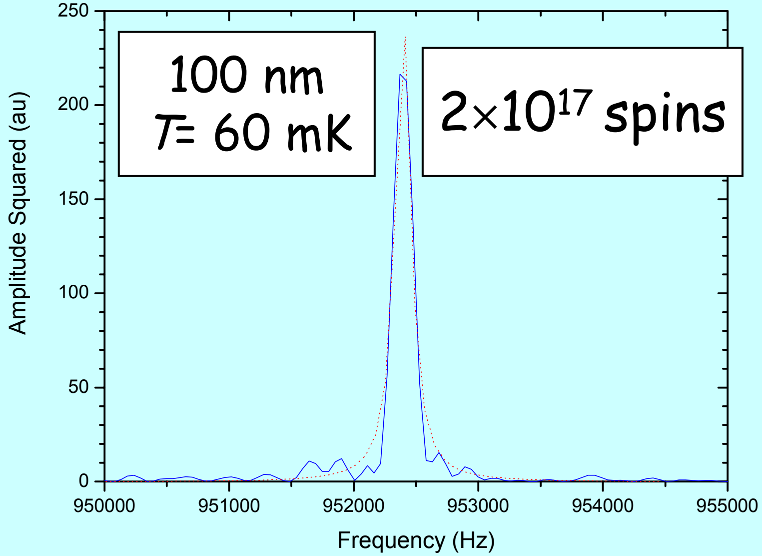


Eg 100 nm film, spin density 10^{-4} x bulk

For superfluid film coil geometry

$N_m = 3.\,10^{16}$ ($\rightarrow 5.10^{15}$) **(1MHz)**

^{3}He monolayer ⇔ **6.10^{14} cm^{-2}**



Observation of signal from a nominal "100 nm thick film"

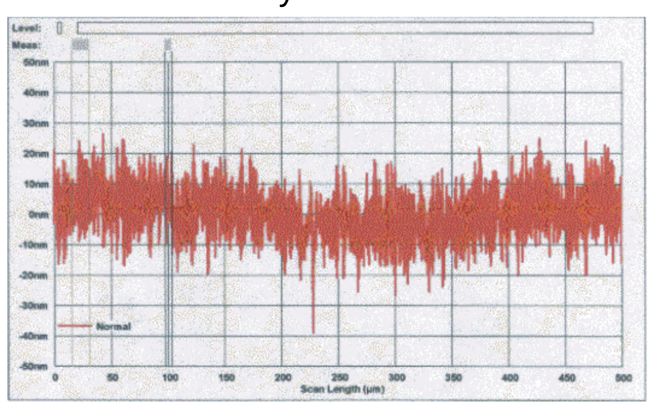
- stability of film ?
- cooling the film ?

Mechanical polishing achieves a measured roughness of ± 10 nm over 50mm

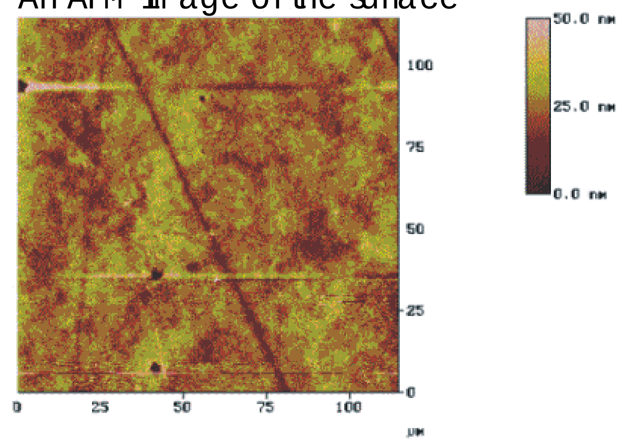
Model the surface as $h = h_0 \cos(kx)$, then h_0 = 10 nm and k = 4 mm^{-1}

Surface Scan by a Tencor Profilometer

±10 nm



An AFM image of the surface



Superfluid ^{3}He films

1. 1987 Freeman *et al*

NMR on ^{3}He in channels

2. 1990 Crooker *et al*

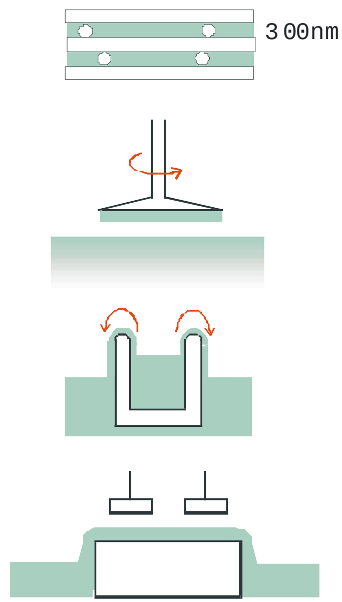
Metastable films

3. 1991 Harrison *et al*

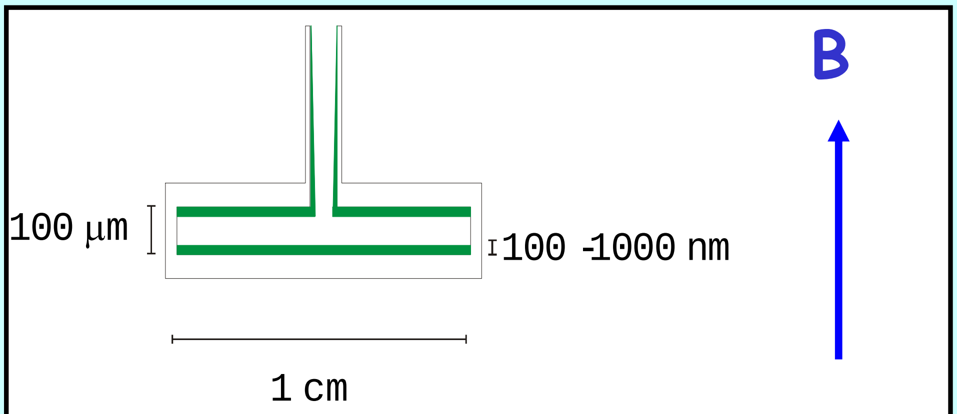
'Self emptying beaker'

4. 1998/99 Davis *et al*

Third sound



- Freeman et al clearly observed a transition into the A-phase.
- All observed a reduction in T_c as a function of film thickness
- The equilibrium phase diagram as a function of film thickness has not been mapped out.



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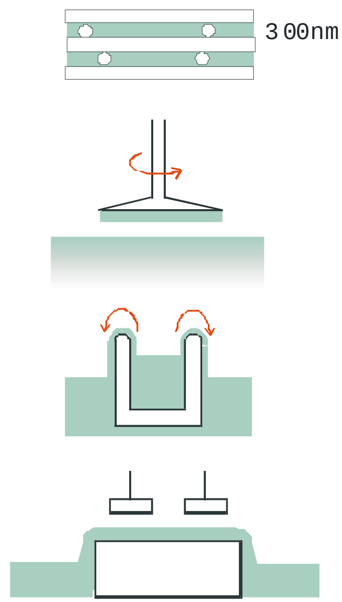

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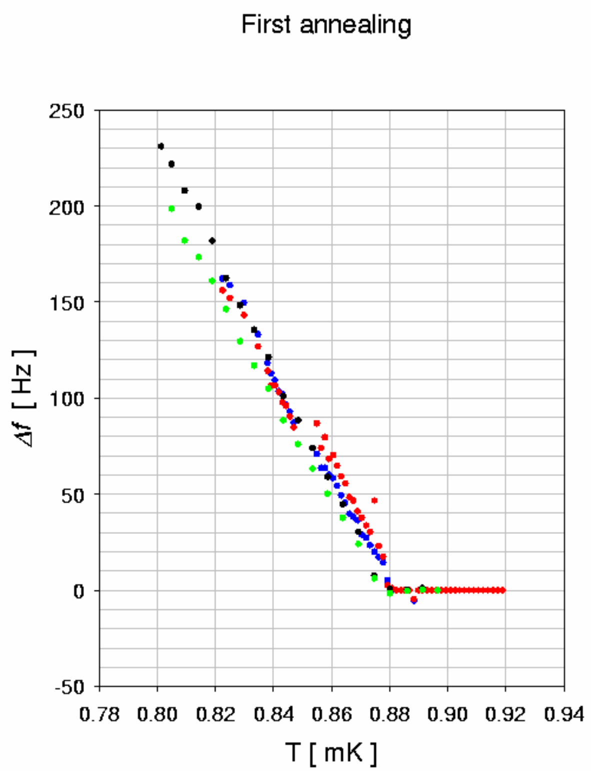
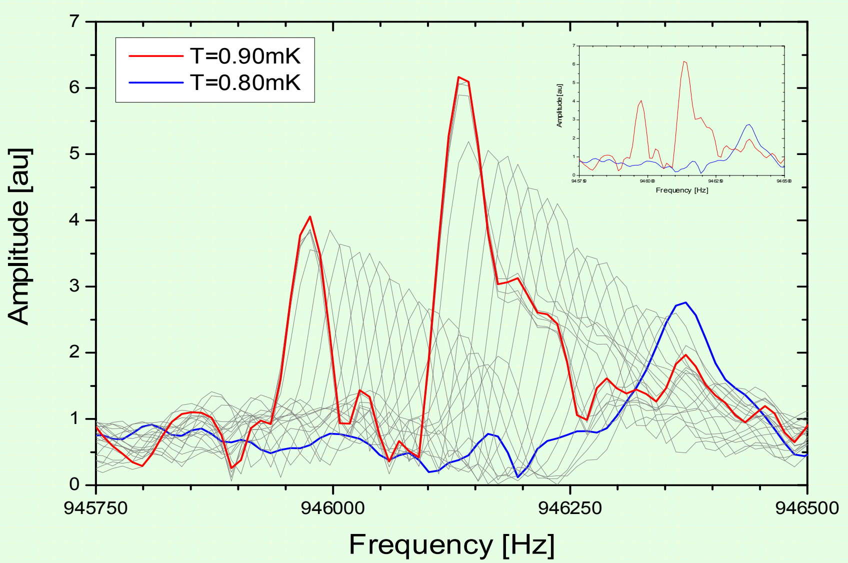
NMR ⇒ "fingerprint" to identify superfluid ground state, as a function of film thickness

- observe a superfluid transition

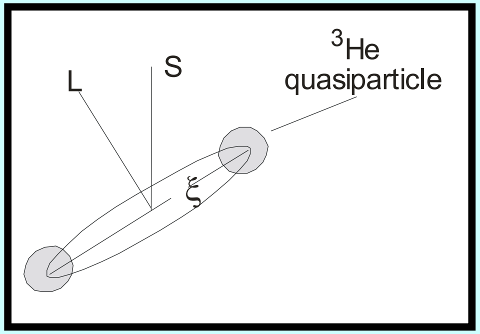
  ⇒ film cools through 1 cm^2 surface

- frequency shift is positive



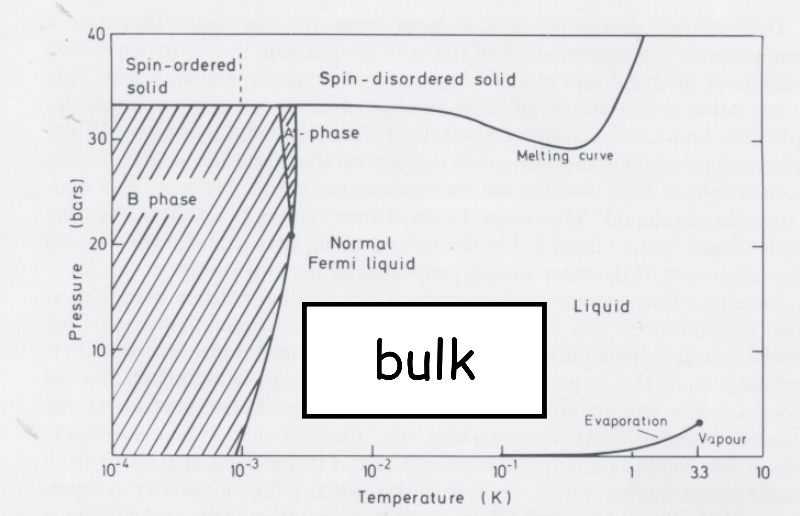


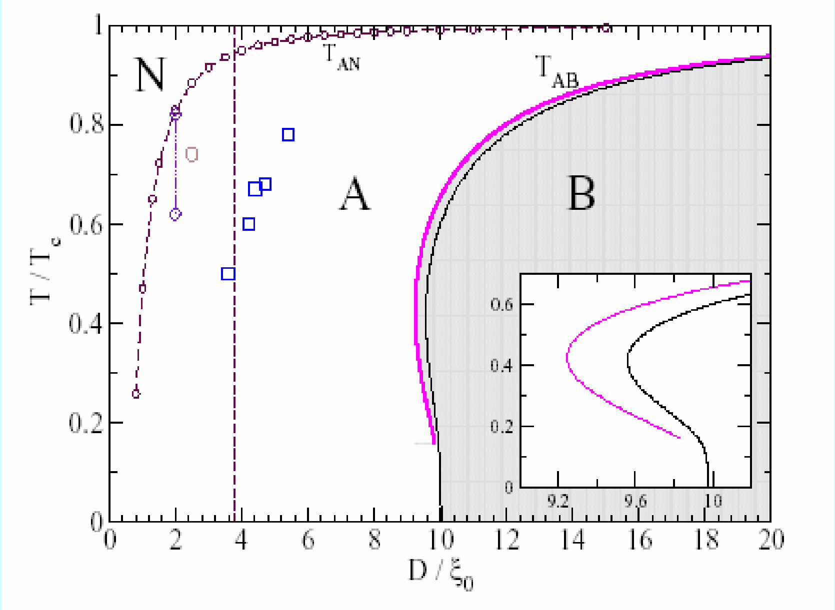
Unconventional (p-wave) superfluidity in thin slabs



$$\xi_0 = \frac{\hbar v_F}{2\pi k_B T_c}$$

72 nm → 14 nm

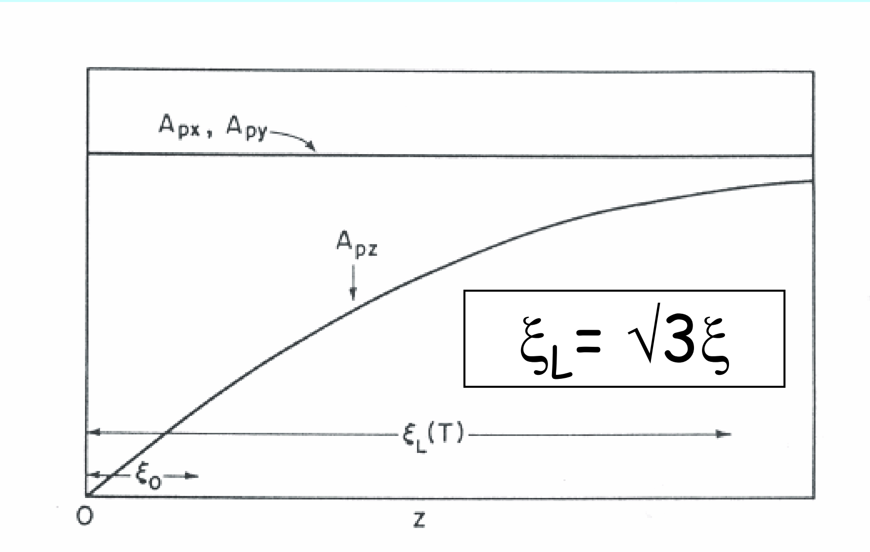




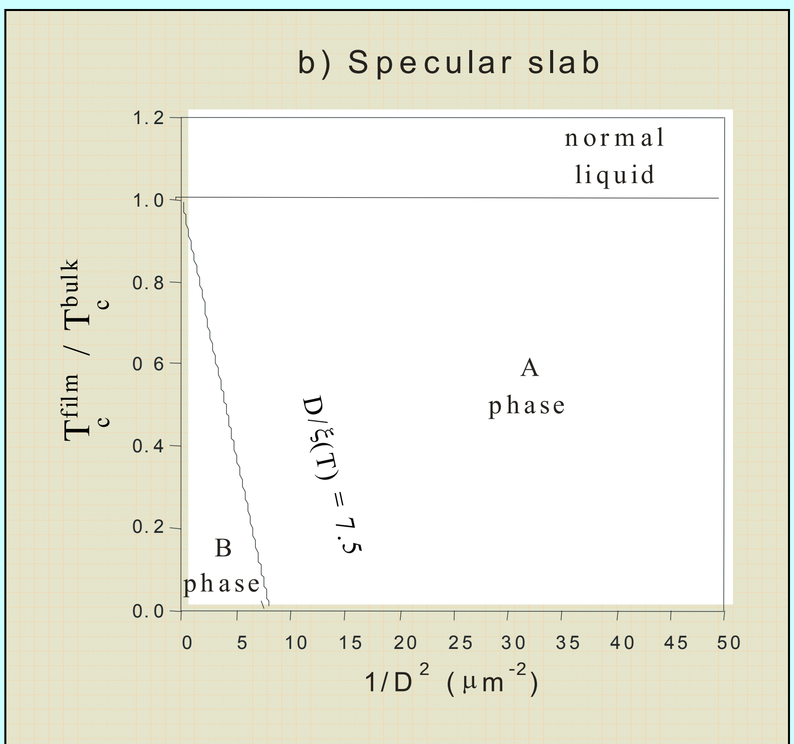
Superfluid phase diagram of thin film

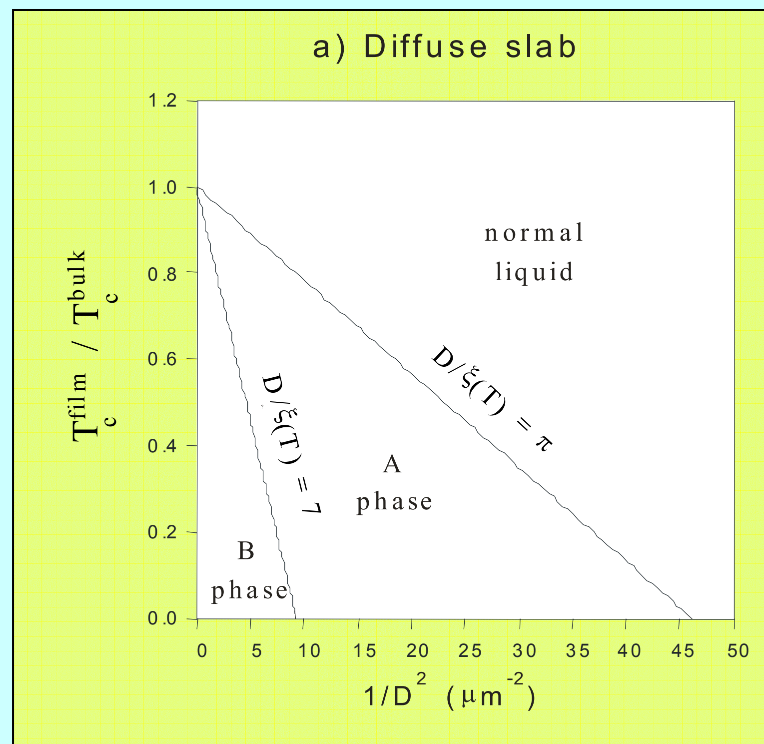
Suppression of components of order parameter at a wall

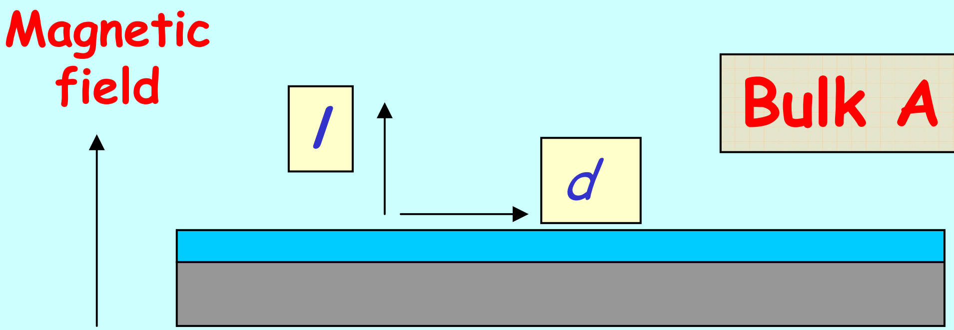
Ambegaokar, deGennes, Rainer (1974)



$$\xi(T) = 0.649\xi_0 \left(1 - \frac{T}{T_c}\right)^{1/2}$$

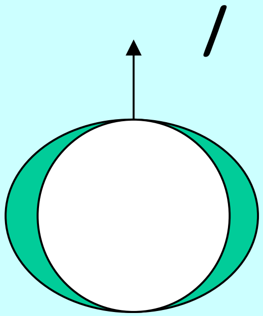






Bulk A phase

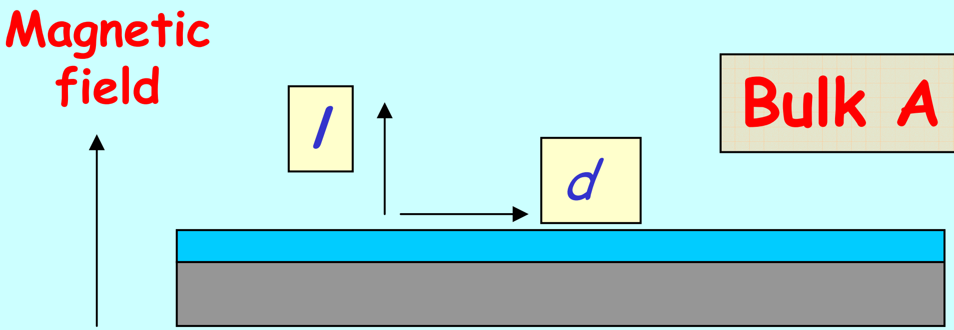
$$\underline{d} = \Delta \begin{bmatrix} 1 & i & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \underline{k}$$



This orientation of *l* and *d* maximises dipole energy

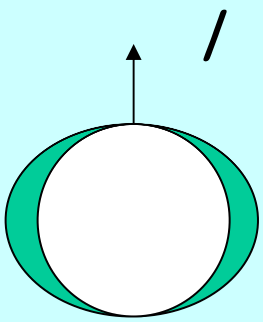
$\Rightarrow$ negative frequency shift

A new phase of superfluid ^{3}He?



Bulk A phase

$$\underline{d} = \Delta \begin{bmatrix} 1 & i & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \underline{k}$$

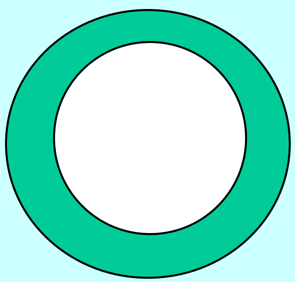


This orientation of *l* and *d* maximises dipole energy

⇒ negative frequency shift

2D "a-phase"

$$\underline{d} = \Delta \begin{bmatrix} 1 & i \\ 0 & 0 \\ 0 & 0 \end{bmatrix} \underline{k}$$



l normal to plane

ESP state
isotropic gap

Sr_2RuO_4

This orientation of *l* and *d* minimises dipole energy
⇒ positive frequency shift

Brusov and Popov (1981)

Stein and Cross (1979)

Tešanović and Valls, Phys. Rev. B31, 1374 (1985)

Clash of dimensionalities

$$k_F \square \frac{1}{a} << d$$

Normal fluid is 3D

$$d \square \xi$$

Superfluid is 2D

Clash of dimensionalities

$$k_F \sim \frac{1}{a} << d$$

Normal fluid is 3D

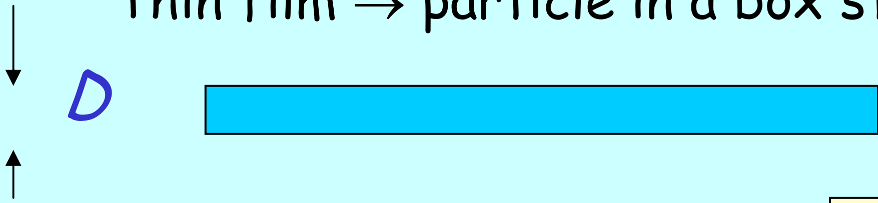
$$d \sim \xi$$

Superfluid is 2D

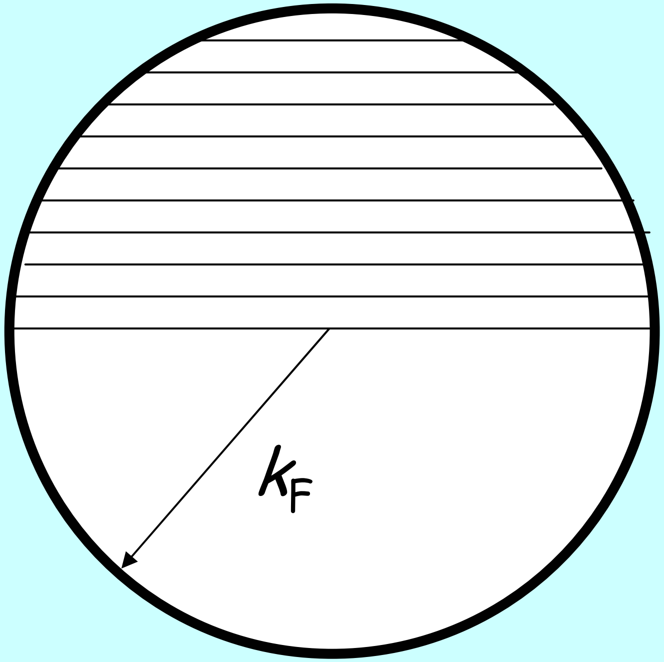
?

Quantum size effects

Thin film → particle in a box states



$$\varepsilon = \frac{\hbar^2}{2m}(k_x^2 + k_y^2) + \frac{n^2 h^2}{8mD^2}$$



Number of Fermi discs $N = \frac{k_F D}{\pi}$

[~ 200 discs/100nm thickness]

Energy scales

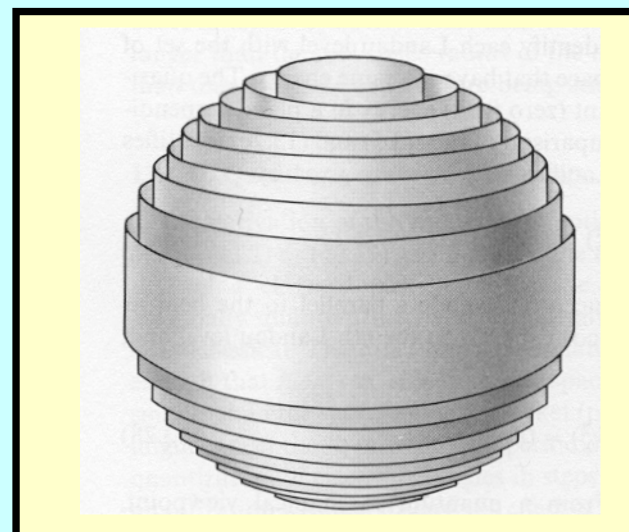
$\frac{1}{k_B}\frac{h^2}{8mD^2}$ ~ 1.5 μK for D =300 nm

$\frac{1}{k_B}\frac{2E_F}{N}$ ~ 5 mK for D = 300 nm

(N → N + 1)

Lifetime broadening h/τ ~ 10 μK at *T* ~ 1 mK

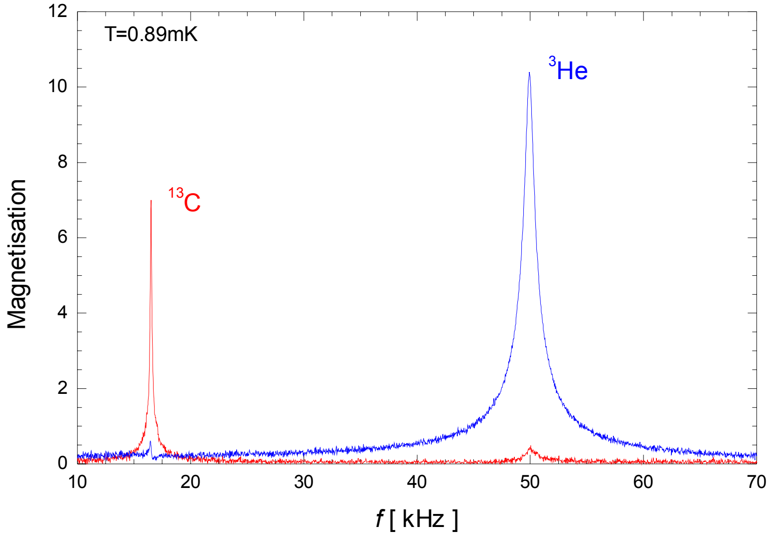
cf dHvA →



⇒ coherent 2D superfluidity of discs

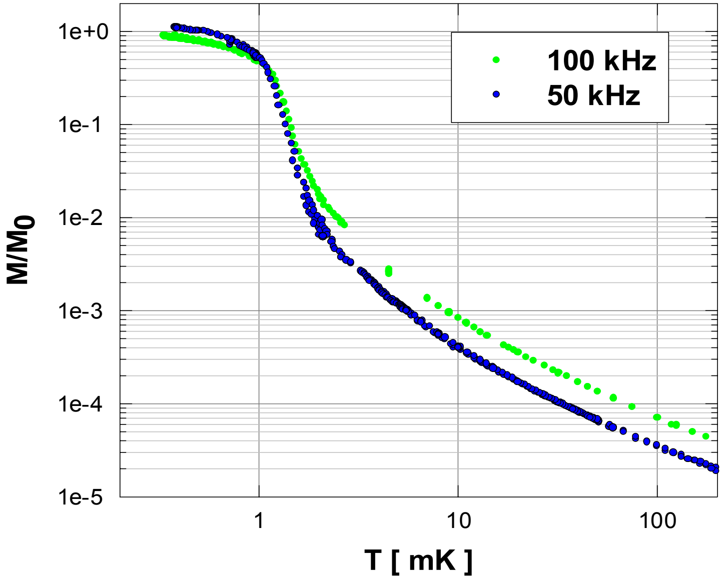
Broadband SQUID NMR: 2D ferromagnetic ^3He on graphite

In an applied field of 1.54 mT, by simply resetting the frequency synthesiser, the ^{13}C line from the graphite substrate can clearly be observed. Dipolar broadening of the ^{13}C line is small due to low isotopic abundance. Useful for thermometry at T < 1 mK.

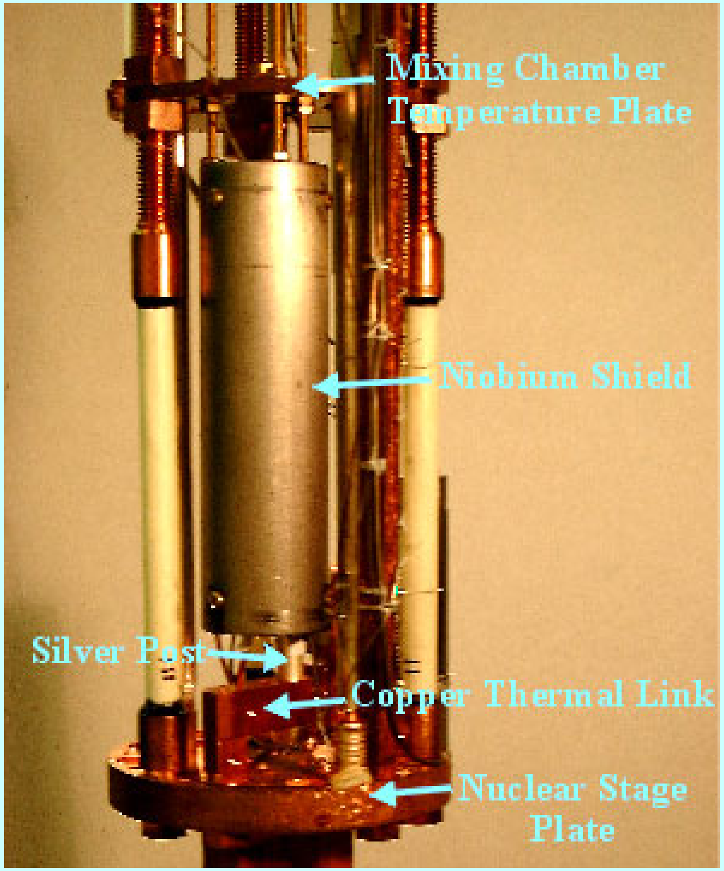


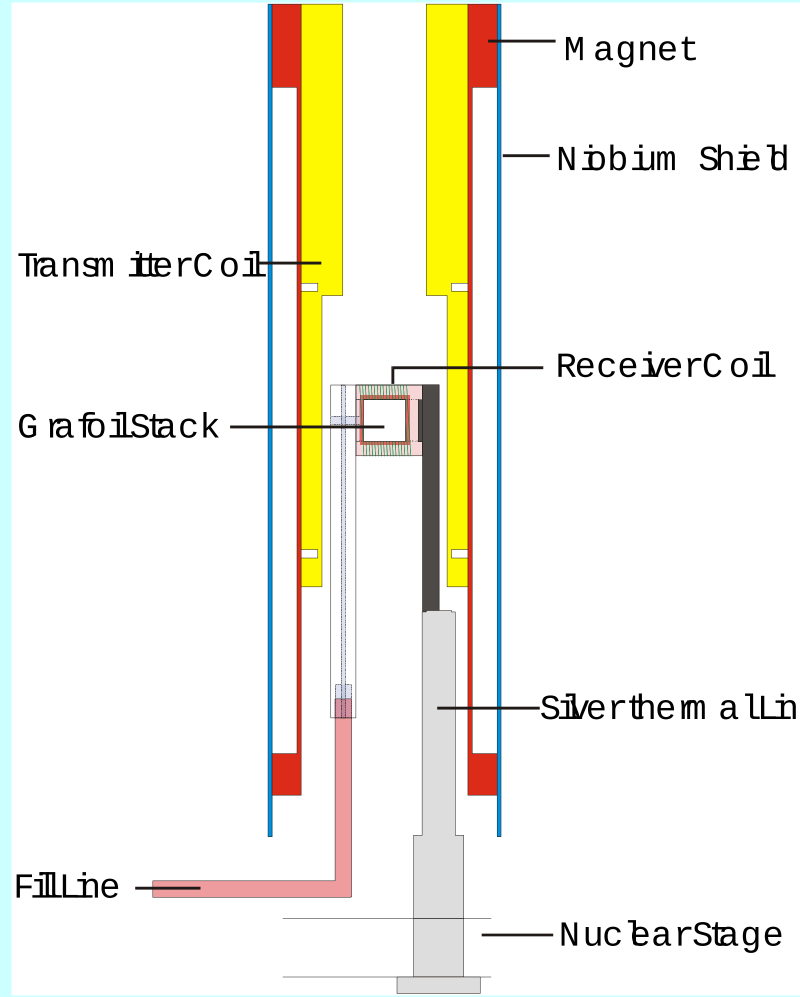
Magnetization approaches saturation at lowest T

Measured magnetization ranges over a factor approaching 10^5

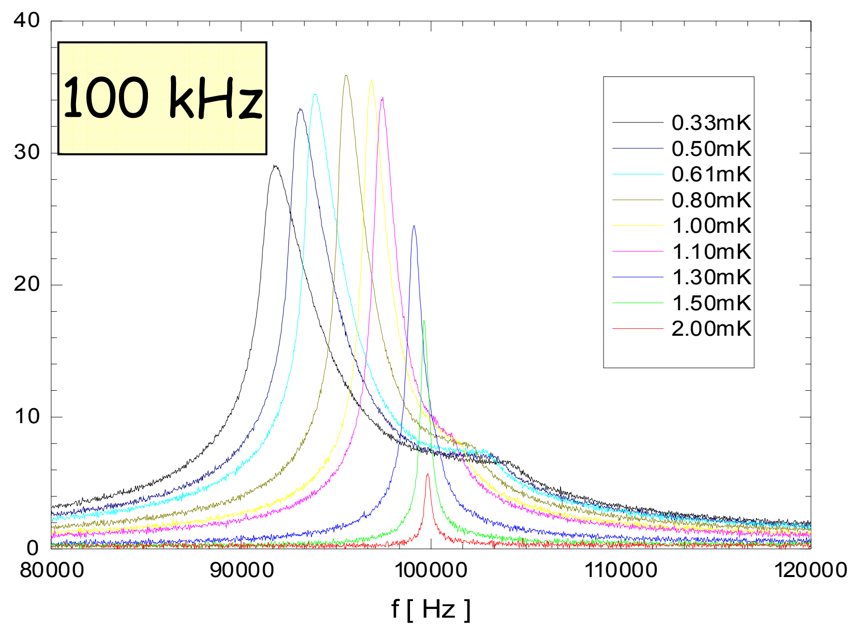


Cell details



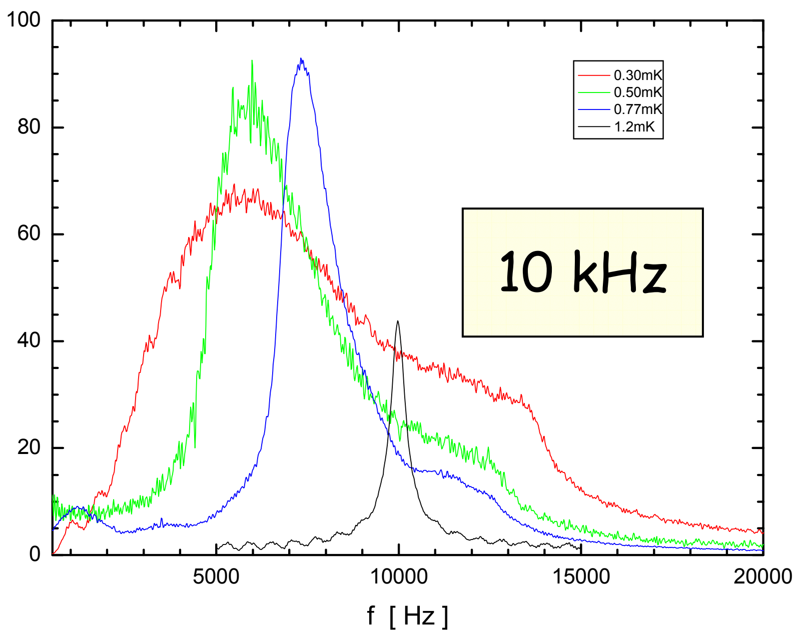


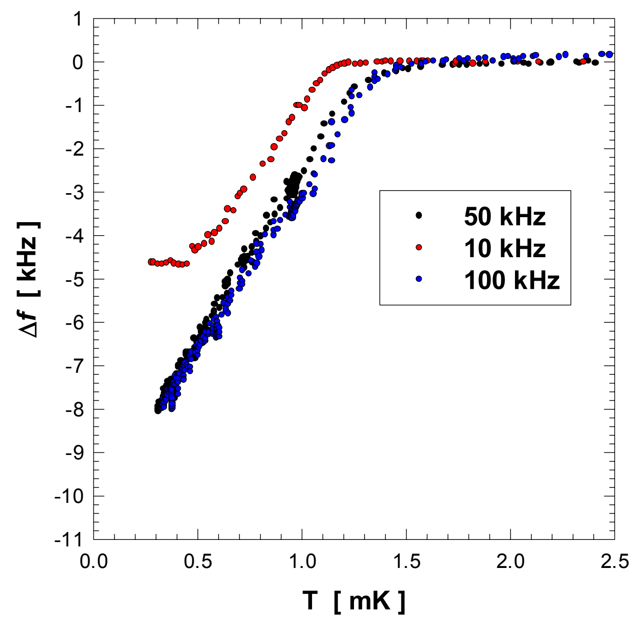
Incorporating inner “overlapping niobium shield” to eliminate eddy current transient from magnet former



Large frequency shifts due to dipolar field

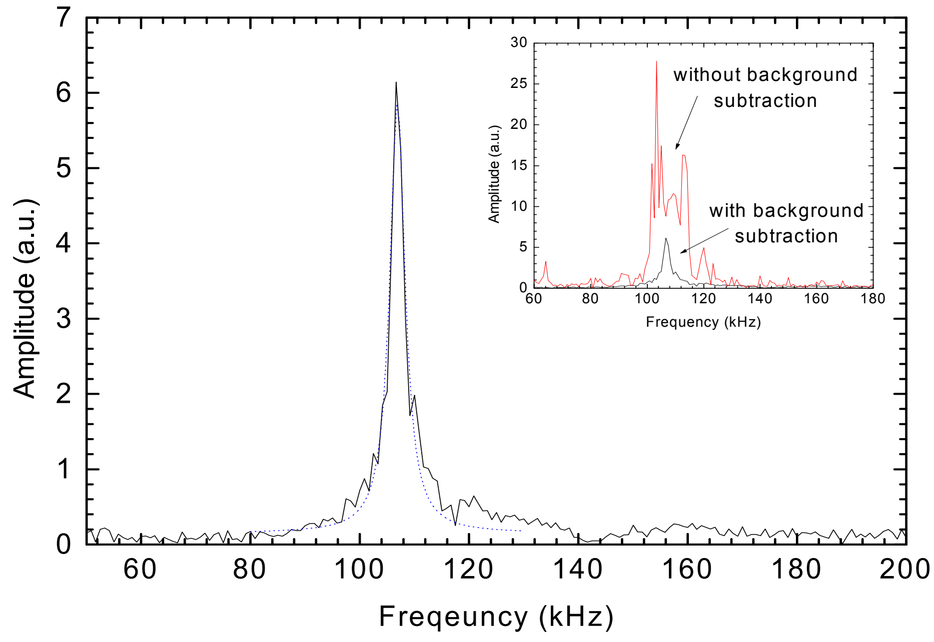
Spin waves in a 2D frustrated ferromagnet





Broadband SQUID NMR on UPt_3

Objective: determination of pairing state



6 single crystals
(0.7 × 0.5 × 5 mm)

Signal from skin/penetration depth
⇒ small signals. Need to minimise background signal.

Measurements in 12-61 mT (previous lowest field 190 mT, H. Tou *et al.*, *Phys. Rev. Lett.* **80**, 9791 (1998))

Surface quality is an issue

Future directions

New p-wave order parameters in confined geometries, for example thin slab

Ultralow field NMR on biological systems (porous media) using broadband spectrometers

Improvements in sensitivity