HFI - Uppsala - June 1974:
My First Big International Conference!

Physica Scripta. Vol. 11, 144–148, 1974

μ^+SR Spectroscopy: The Positive Muon as a Magnetic Probe in Solids

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Received July 13, 1974

→ μ^+ in Ni, Fe

→ μ^+ quantum diffusion

→ Mu in SiO_2, Si, Ge; Mu* in Si

HOWTO of μSR methods developed at TRIUMF

A science fiction adventure story

by

Jess H. Brewer

UBC PHYSICS & ASTRONOMY

Our Motto:

μSR

Rotation
Relaxation
Resonance

muon spin

Applied*
Elementary
Particle
Physics

*(to basic research in
Materials Science
and Chemistry)

OUTLINE

- Research "**Themes**" in ***μSR***
- ***History*** of ***μSR*** ("*science fiction*"?)
- Development of ***Advanced Muon Beams***
- "**Workhorse**" ***μSR techniques***:

  wTF, ZF, LF, Strobo, FFT
- "**Exotic**" ***μSR techniques***:

  SR, HTF, RRF, ALCR, RF, μSI, E-field
- (***μSR applications*** interleaved among ***techniques***...)

Research "Themes" in μ^+SR

Muonium as light Hydrogen

(Mu = μ^+e^-) (H = p^+e^-)

- **Mu vs. H atom Chemistry:**
  - gases, liquids & solids
  - Best test of reaction rate theories.
  - Study "unobservable" H atom rxns.
  - Discover new radical species.
- **Mu vs. H in Semiconductors:**
  - Until recently, μ^+SR → **only** data on metastable H states in semiconductors!

The Muon as a Probe

- **Probing Magnetism:** unequalled sensitivity
  - Local fields: electronic structure; ordering
  - Dynamics: electronic, nuclear spins
- **Probing Superconductivity:** (esp. HT_cSC)
  - Coexistence of SC & Magnetism
  - Magnetic Penetration Depth λ
  - Coherence Length ξ

- **Quantum Diffusion:** μ^+ in metals (compare H^+); Mu in nonmetals (compare H).

And then there's $\mu^{-}SR$ ...

... but there's not enough time.

:-(

Evolution of μSR:

Fantasy ➜ Fiction ➜ Physics

- **Fantasy**: violates the "known laws of physics"
- **Science Fiction**: possible in principle, but impractical with existing technology. (**Clarke's Law**: *"Any sufficiently advanced technology is indistinguishable from magic."*)
- **Routine Physics**: "We can do that . . ."
- **Applied Science**: " . . . and so can you!"

Before 1956: ***μSR = Fantasy***

(violates "known laws of physics")

- ## 1930s: **Mistaken Identity**

  Yukawa's "nuclear glue" mesons ≠ cosmic rays
  1937 Rabi: Nuclear Magnetic Resonance

- ## 1940s: "**Who Ordered That?**"

  1940 Phys. Rev. Analytical Subject Index: "mesotron"
  1944 Rasetti: 1st application of muons to condensed matter physics
  1946 Bloch: Nuclear Induction (modern NMR with FID *etc.*)
  1946 Various: "two-meson" π-*μ* hypothesis ***Brewer: born***
  1947 Richardson: produced π & *μ* at Berkeley 184 in. Cyclotron
  1949 Kuhn: *"The Structure of Scientific Revolutions"*

- ## 1950s: "**Particle Paradise**"

  culminating in weird results with strange particles:
  1956 Cronin, Fitch, . . . : *"τ -θ puzzle"* (neutral kaons) → ***Revolution!***

1956-7: *Revolution*

- 1950s: "**Particle Paradise**"
  culminating in weird results with strange particles:
  1956 Cronin, Fitch, . . . : *"τ - θ puzzle"* (neutral **kaons**)

- 1956: Lee & Yang postulate
  ***P*-violation** in weak interactions

- 1957: Wu confirms *P*-violation in β decay;

  Friedman & Telegdi confirm *P*-violation in π-μ-e decay;

  so do Garwin, Lederman & Weinrich,
  using a prototype ***μSR*** technique.

For newcomers . . .

How does it work?

. . . a brief introduction to

Pion Decay: $\pi^+ \rightarrow \mu^+ + \nu_\mu$

A spinless *pion* **stops** in the “skin” of the primary production target.
It has zero linear momentum and zero angular momentum.

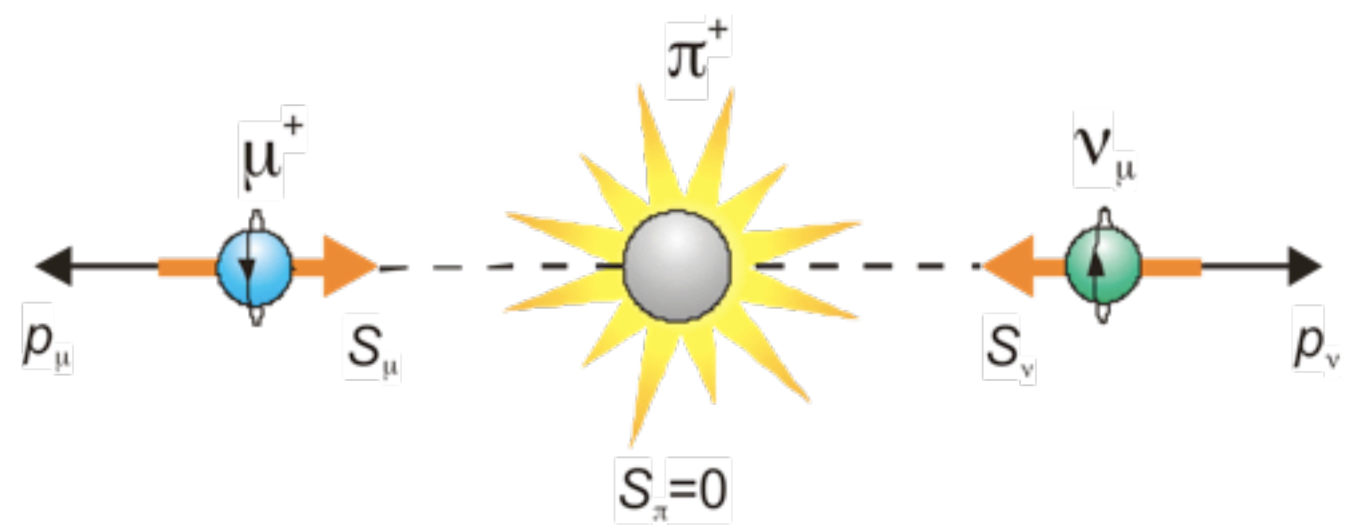
Conservation of Linear Momentum: The μ^+ is emitted with momentum equal and opposite to that of the ν_μ .

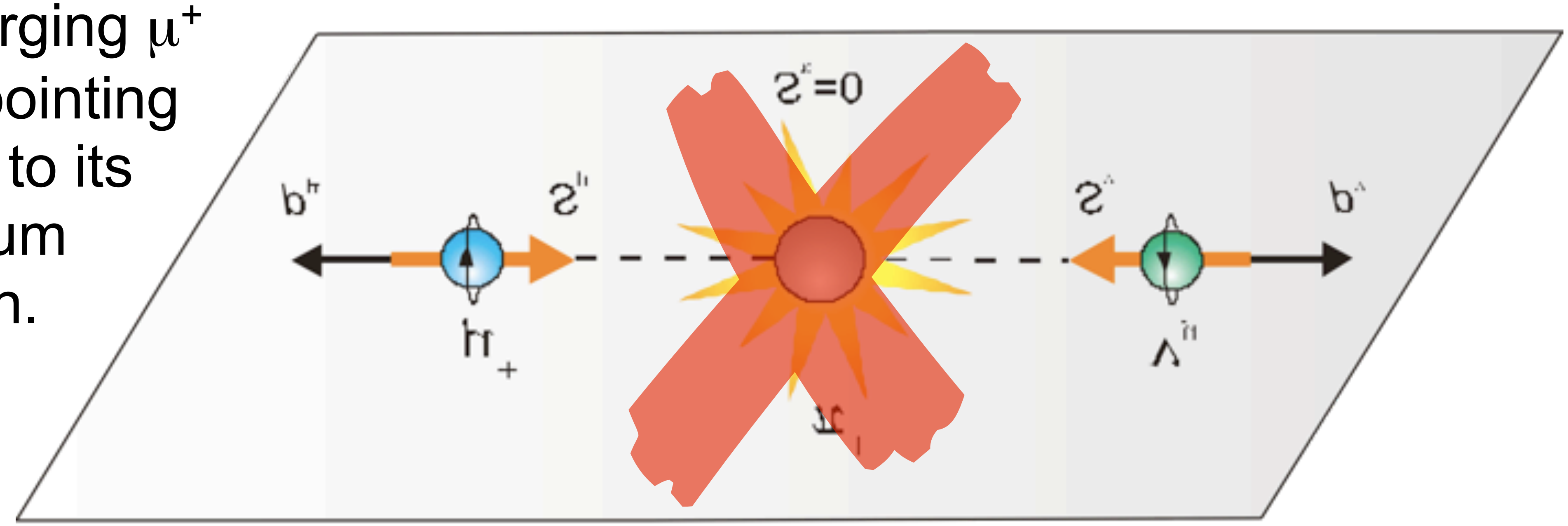
Conservation of Angular Momentum: μ^+ & ν_μ have equal & opposite spin.

Weak Interaction:

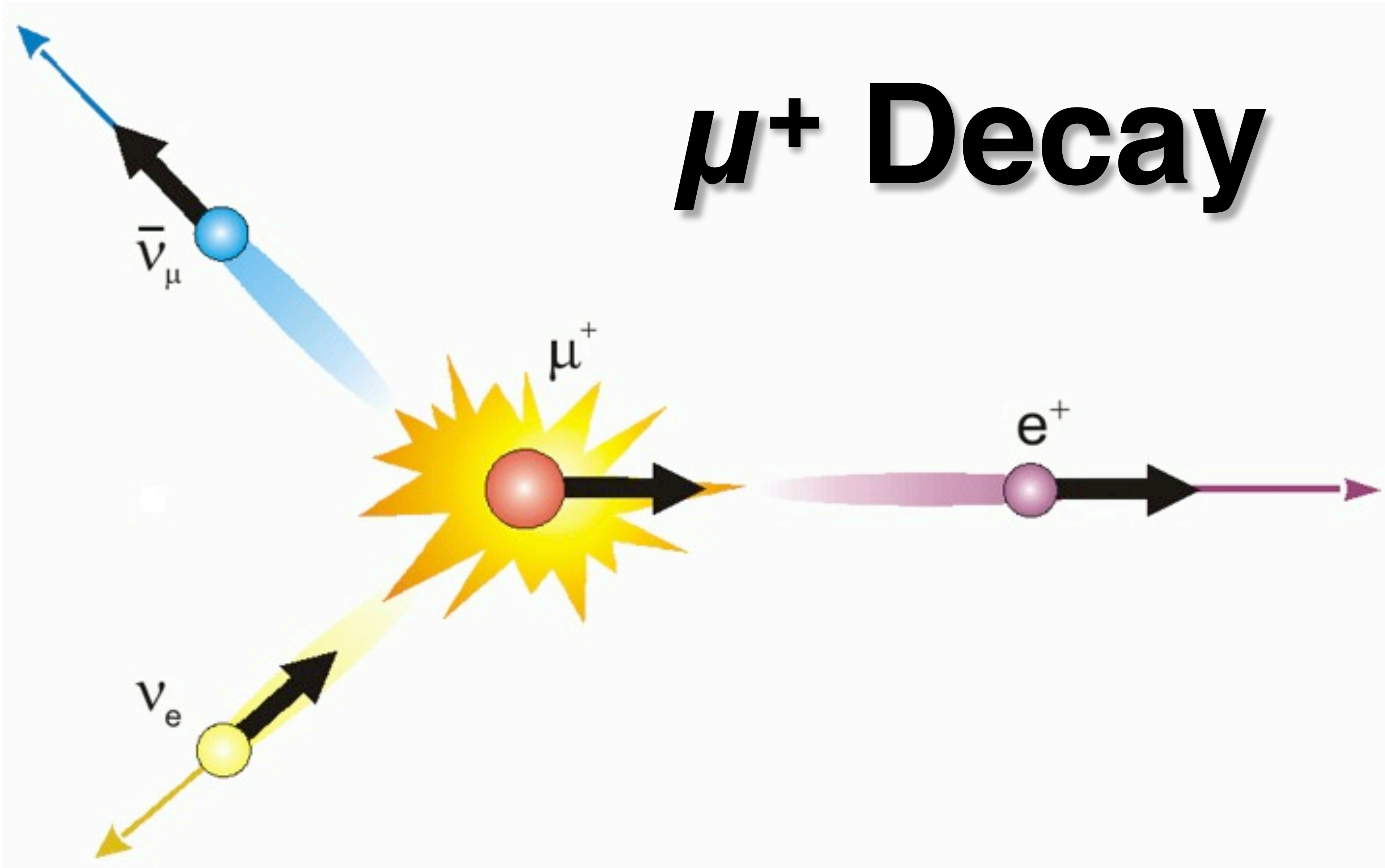
Only “left-handed” ν_μ are created.

Thus the emerging μ^+ has its spin pointing antiparallel to its momentum direction.





μ^+ Decay



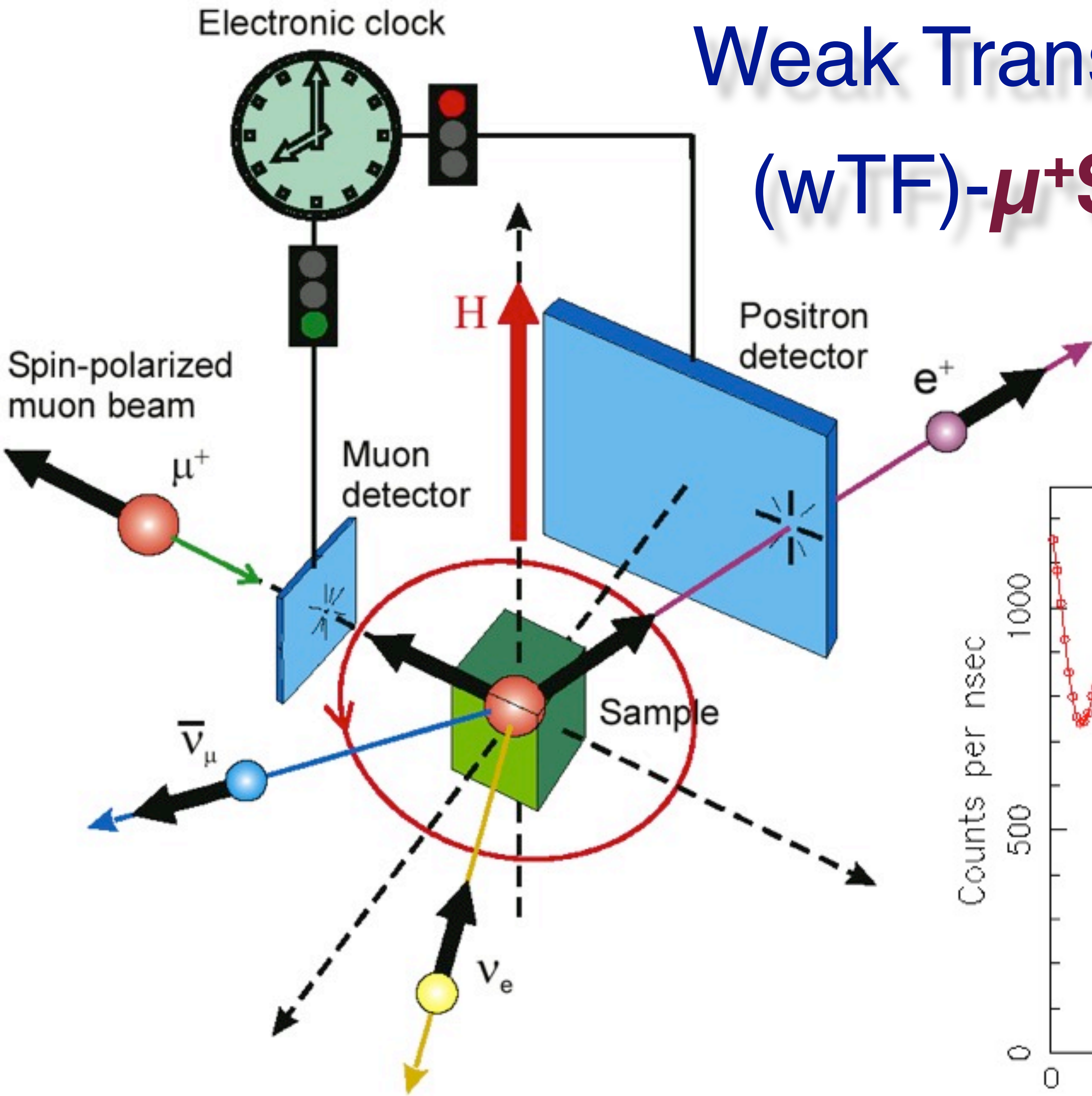
Neutrinos have negative helicity, antineutrinos positive.

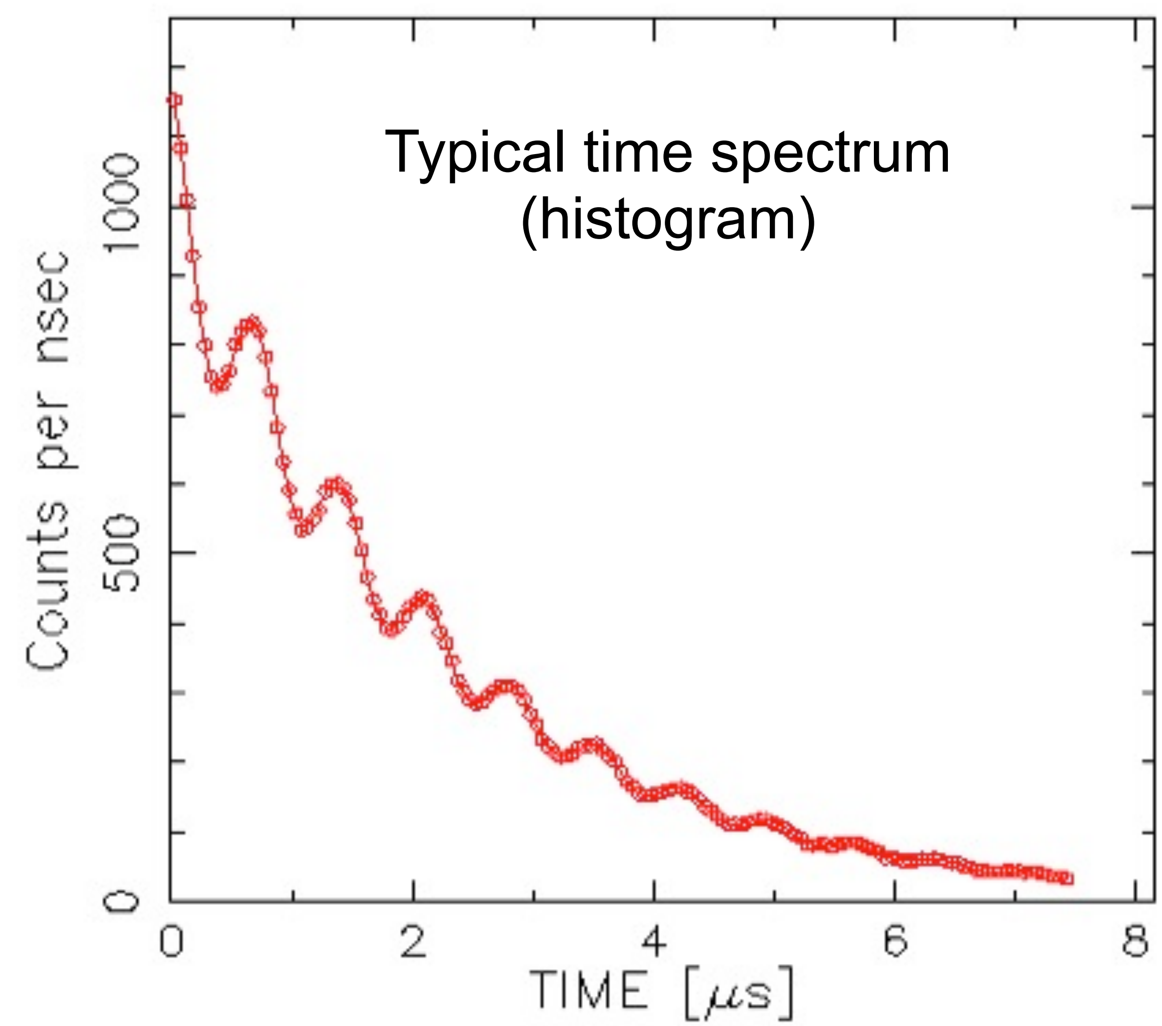
An ultrarelativistic positron behaves like an antineutrino.

Thus the positron <u>tends</u> to be emitted along the μ^+ spin

when ν_e and $\bar{\nu}_\mu$ go off together (highest energy e^+).

Weak Transverse Field (wTF)-μ^+SR (CW)





1958-1973: *Science Fiction*

- 1960s: **Fundamental Physics Fun!** – *Tours de Force*

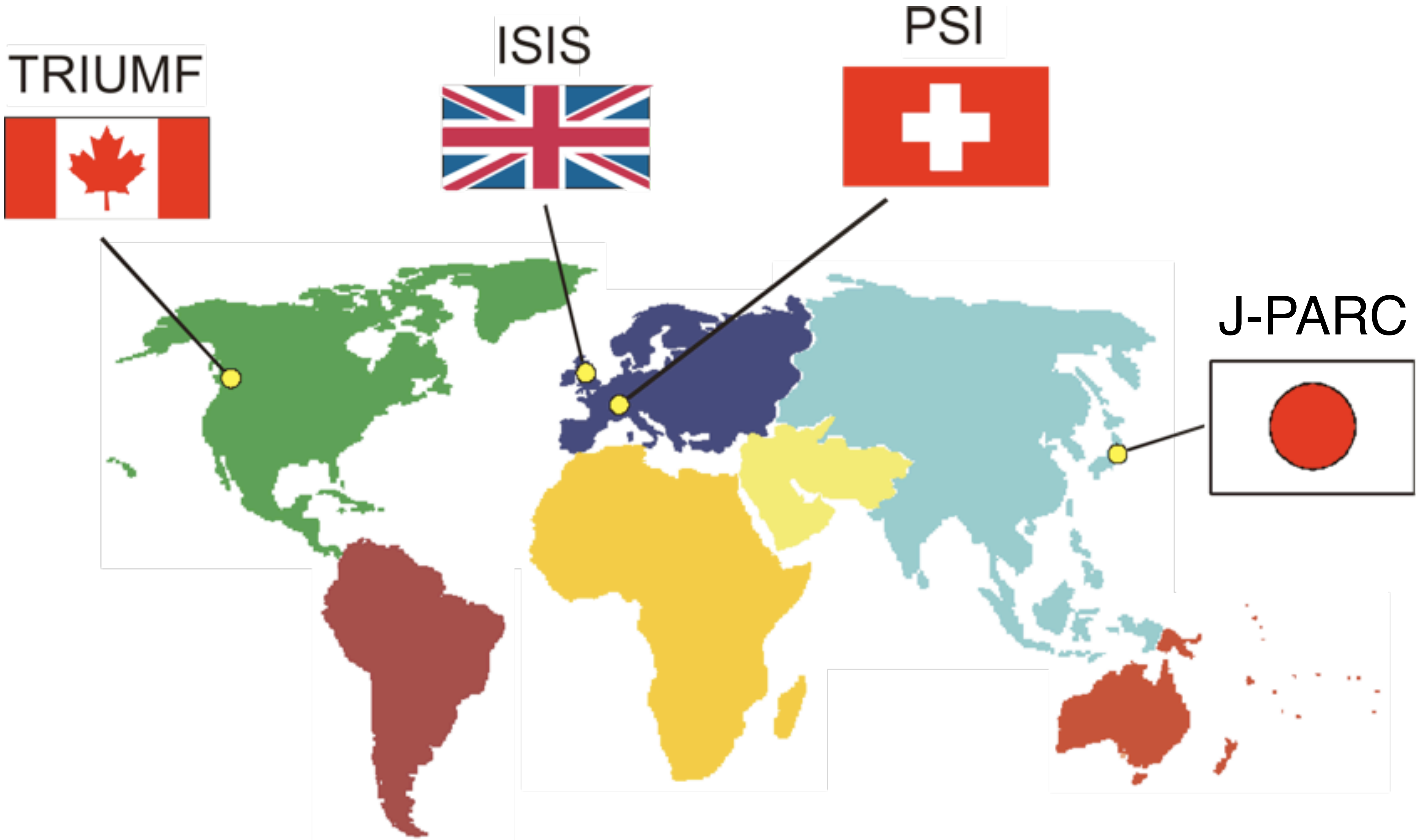
  Michel Parameters = Weak Interaction Laboratory
  Heroic **QED** tests: A_{HF}(Mu), μ_μ, $g_\mu - 2$
  All lead to *refined μSR techniques*.
  Applications: Muonium Chemistry, Semiconductors, Magnetism

- 1972: Bowen & Pifer build first Arizona/**surface muon beam** to search for for $\mu^+e^- \to \mu^-e^+$ conversion

- mid-1970s: **Meson Factories** – *Intensity Enables!*

  USA: **LAMPF** (now defunct)
  Canada: ***TRIUMF***
  Japan: **KEK/BOOM** (→ **J-PARC**)
  Switzerland: **SIN** (now **PSI**)
  UK: **RAL/ISIS**

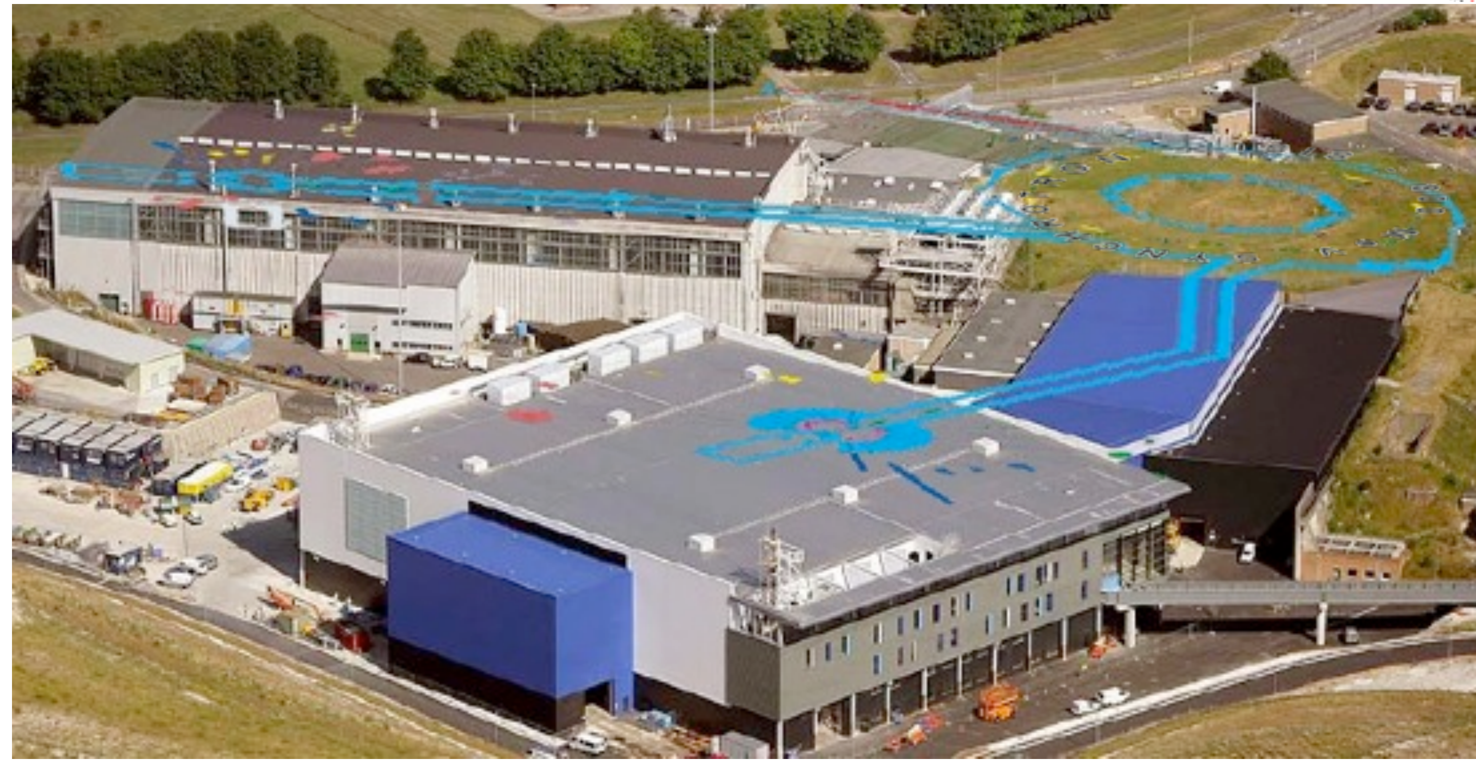
μSR today: Routine Science?



μSR today: Routine Science?

1972

ISIS site plan



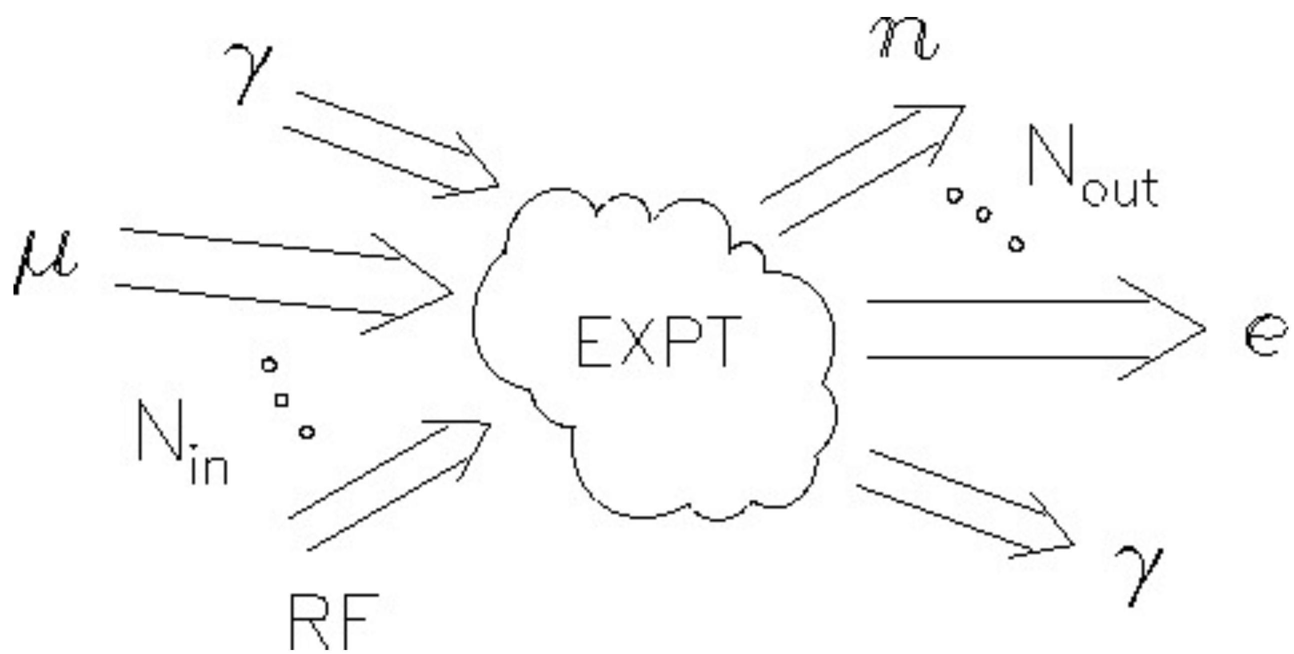
PSI Ring Cyclotron

J-PARC synchrotron

TRIUMF ISIS

CW *vs.* Pulsed μSR

PSI J-PARC



"Advantage factor" for *pulsed* muons:

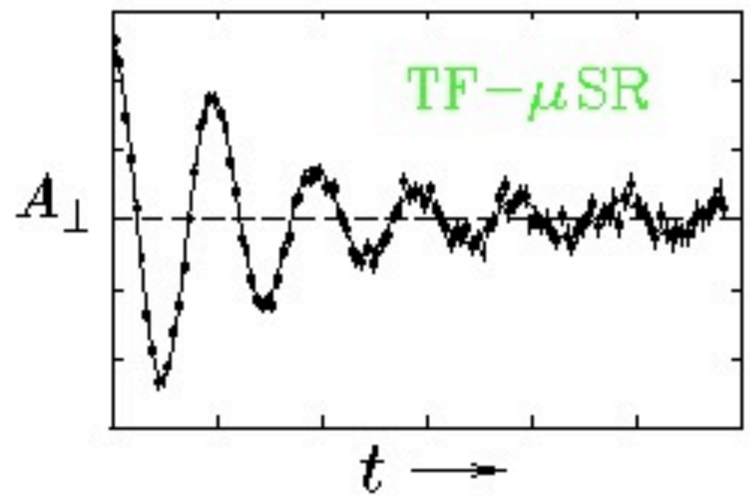
$$A_p = log(N_{in} / N_{out})$$

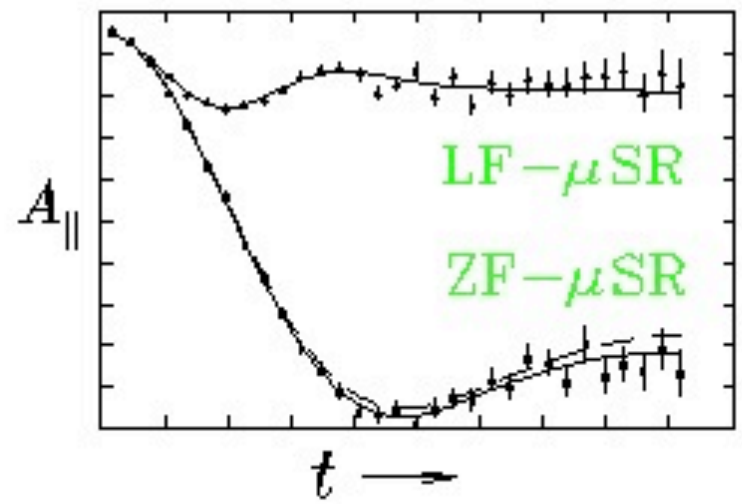
Advantage of *CW* muons: **time resolution** (< 1 ns *vs.* > 10 ns)

*Dis*advantage of *CW* muons: **rate** (< 10^4 s^{-1} *vs.* "unlimited")

μSR Acronyms

Transverse Field

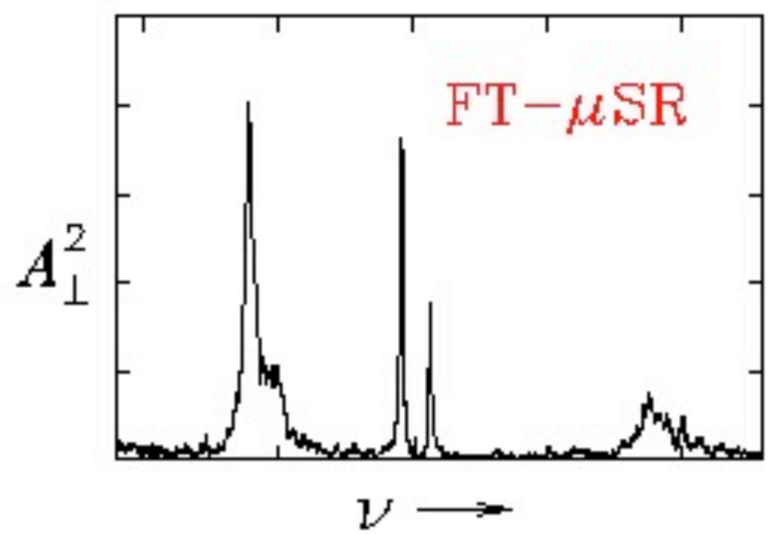


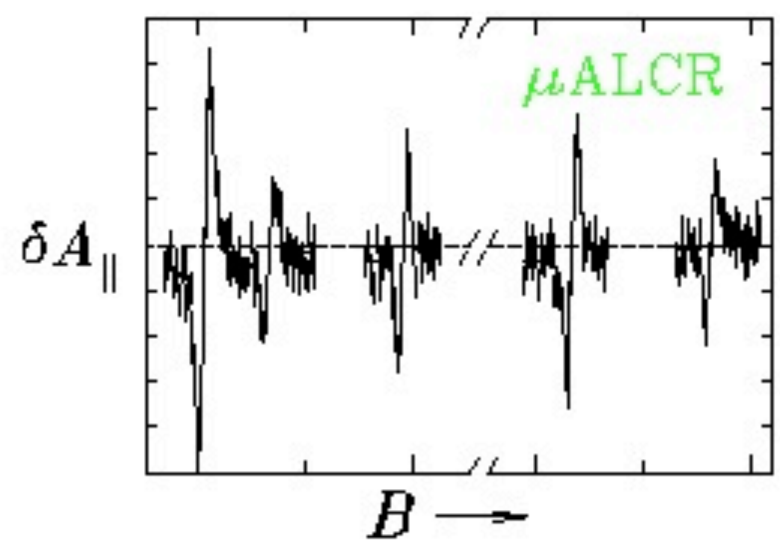


Longitudinal Field

Zero Field

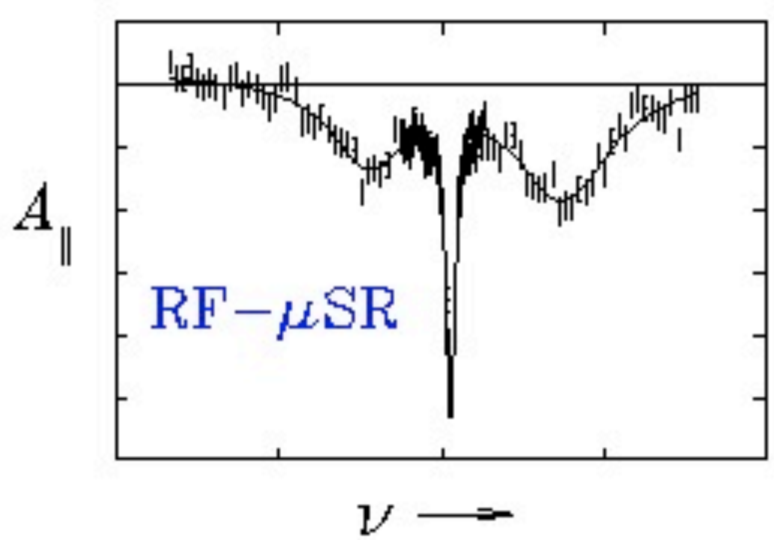
Fourier Transform μSR

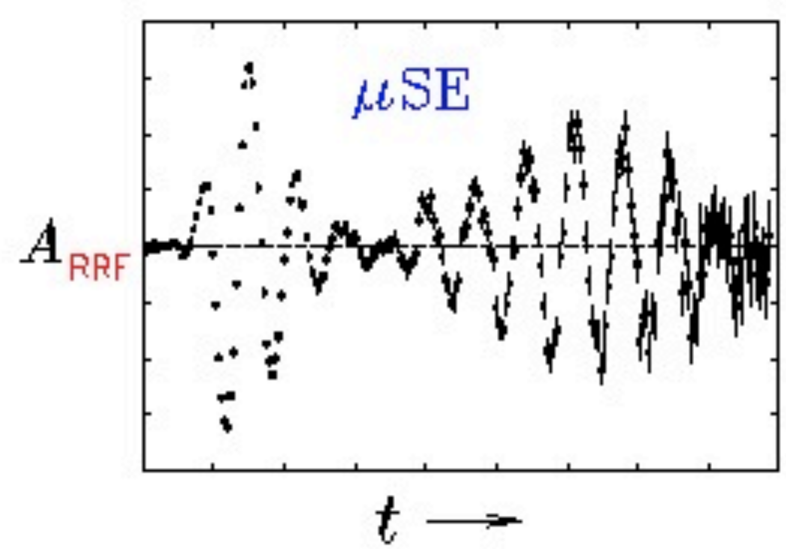




Avoided Level Crossing Resonance

Muon Spin Resonance

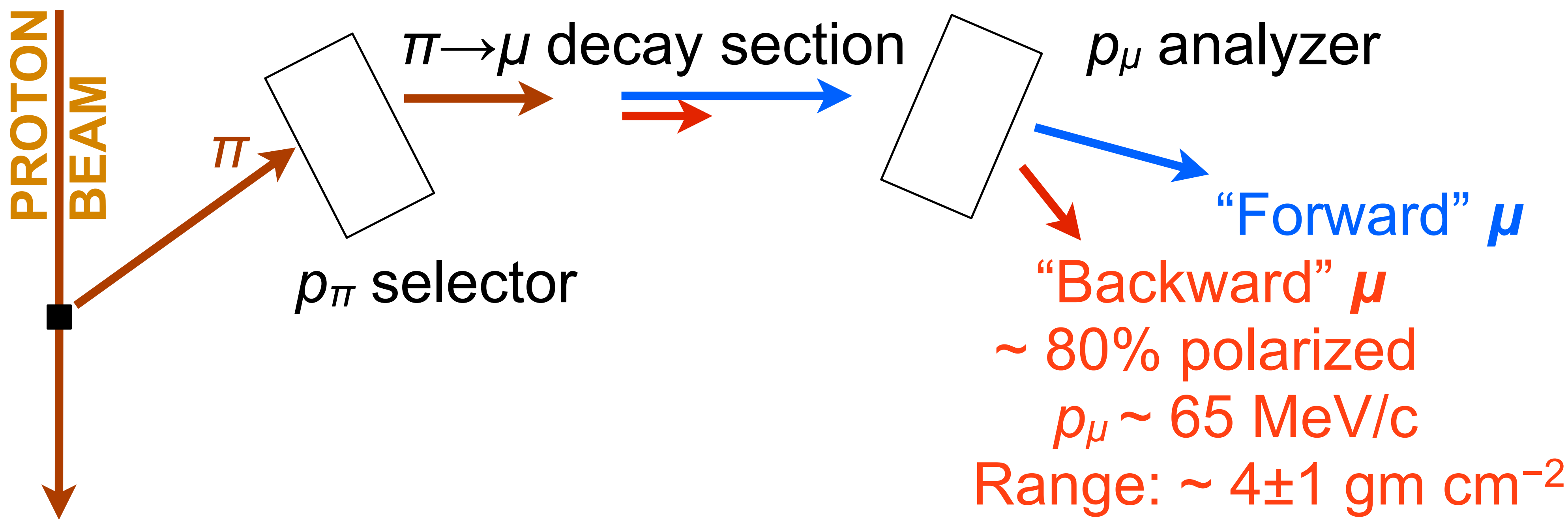




Muon Spin Echo

Forward & Backward Decay Muons

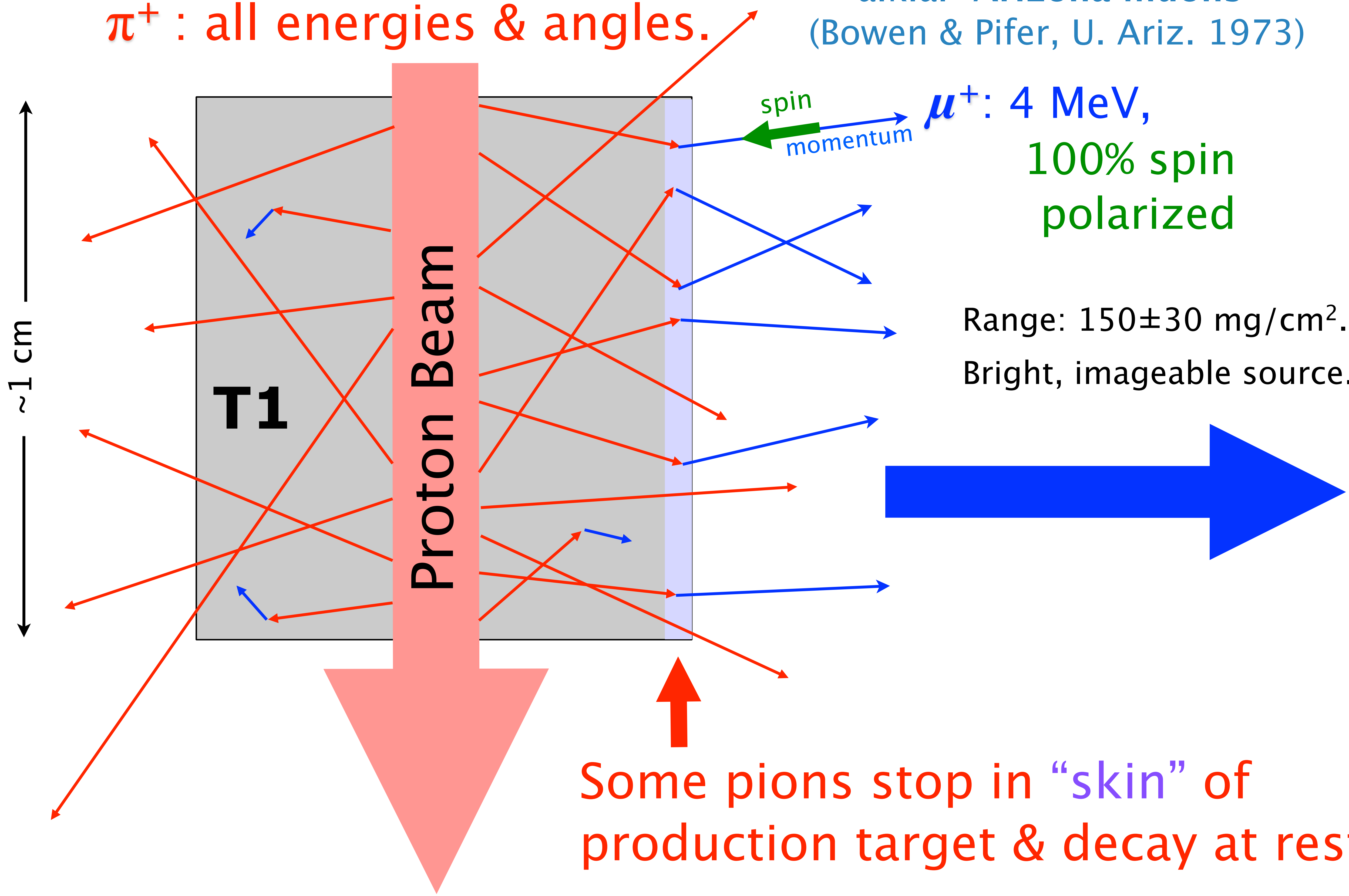
DECAY MUON CHANNEL (μ^+ or μ^-)



Surface Muons

π^+ : all energies & angles.

a.k.a. "**Arizona muons**"
(Bowen & Pifer, U. Ariz. 1973)



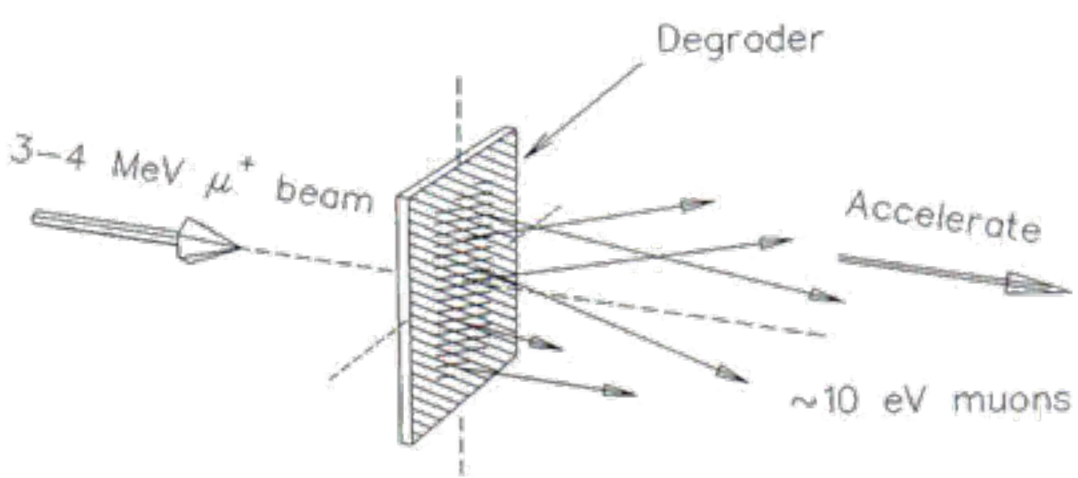
μ^+: 4 MeV,
100% spin polarized

Range: 150±30 mg/cm^2.

Bright, imageable source.

Some pions stop in "skin" of production target & decay at rest.

Moderated Muons



TRIUMF:

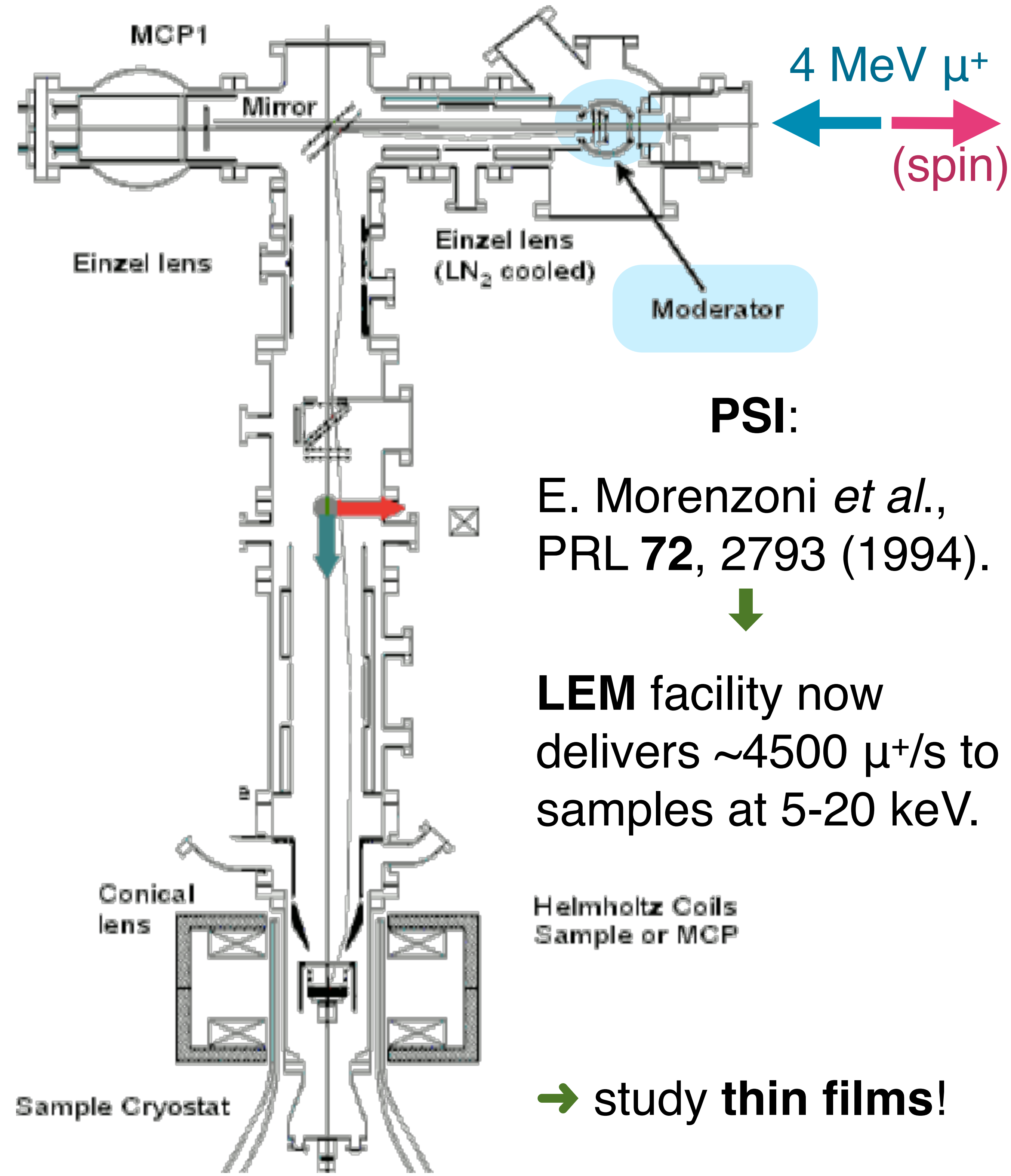
D.R. Harshman *et al.*, PRL **56**, 2850 (1986).

↓

G.D. Morris, M.Sc. thesis (1989).

↑

Unfortunately, yield of 1.75(4) epithermal μ^+ per 10^6 incident surface muons (for a solid Ar moderator) was too low to be practical at TRIUMF intensities.



PSI:

E. Morenzoni *et al.*, PRL **72**, 2793 (1994).

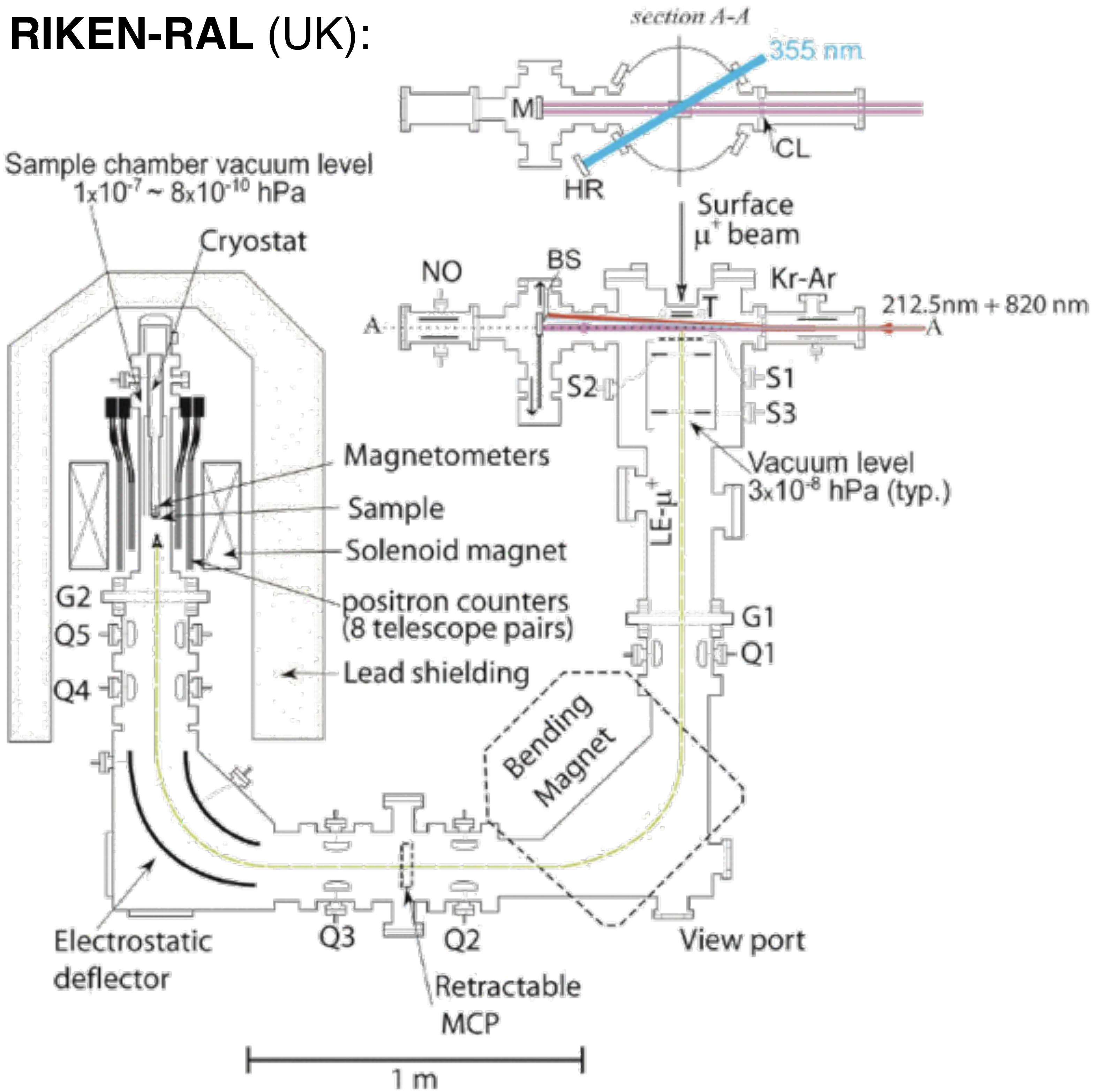
↓

LEM facility now delivers ~4500 μ^+/s to samples at 5-20 keV.

→ study **thin films**!

Laser-Ionize Thermal Muonium

RIKEN-RAL (UK):



J-PARC (Japan):

ULTRASLOW
MUON
MICROSCOPE

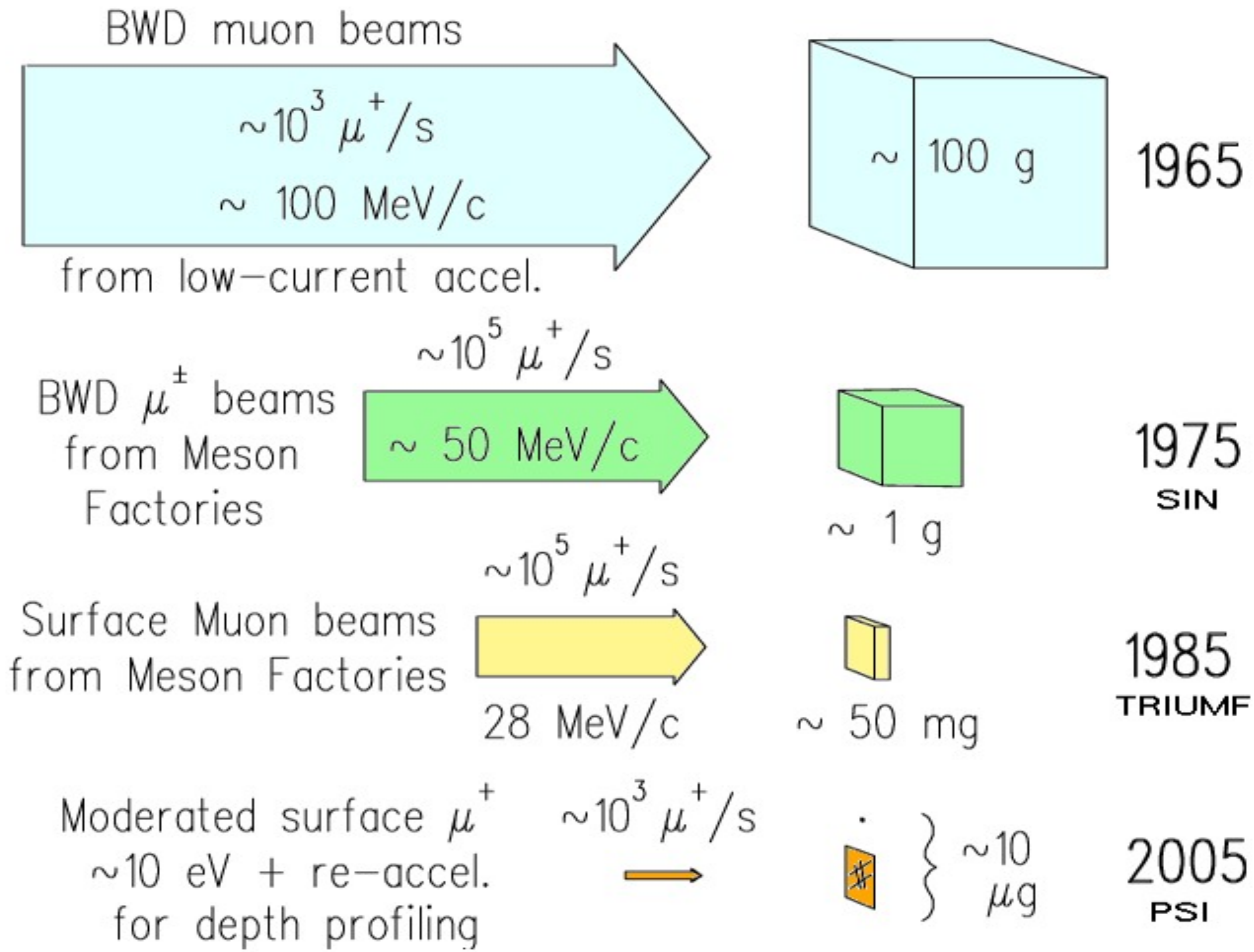
Advantage for **Pulsed** beams: re-accelerated pulse is *short* ⇒ improved time resolution!

Low emittance ⇒ very small final focus! ("Microscope")

➔ More LE-μSR

➔ Improved muon *g*–2 measurement

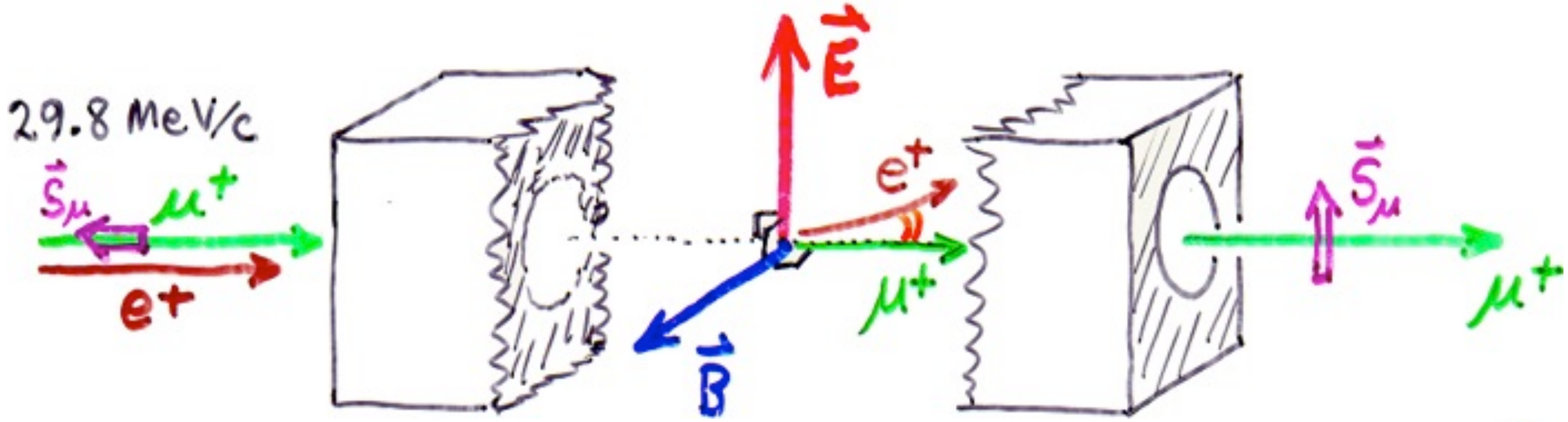
μ^+ Stopping Luminosity



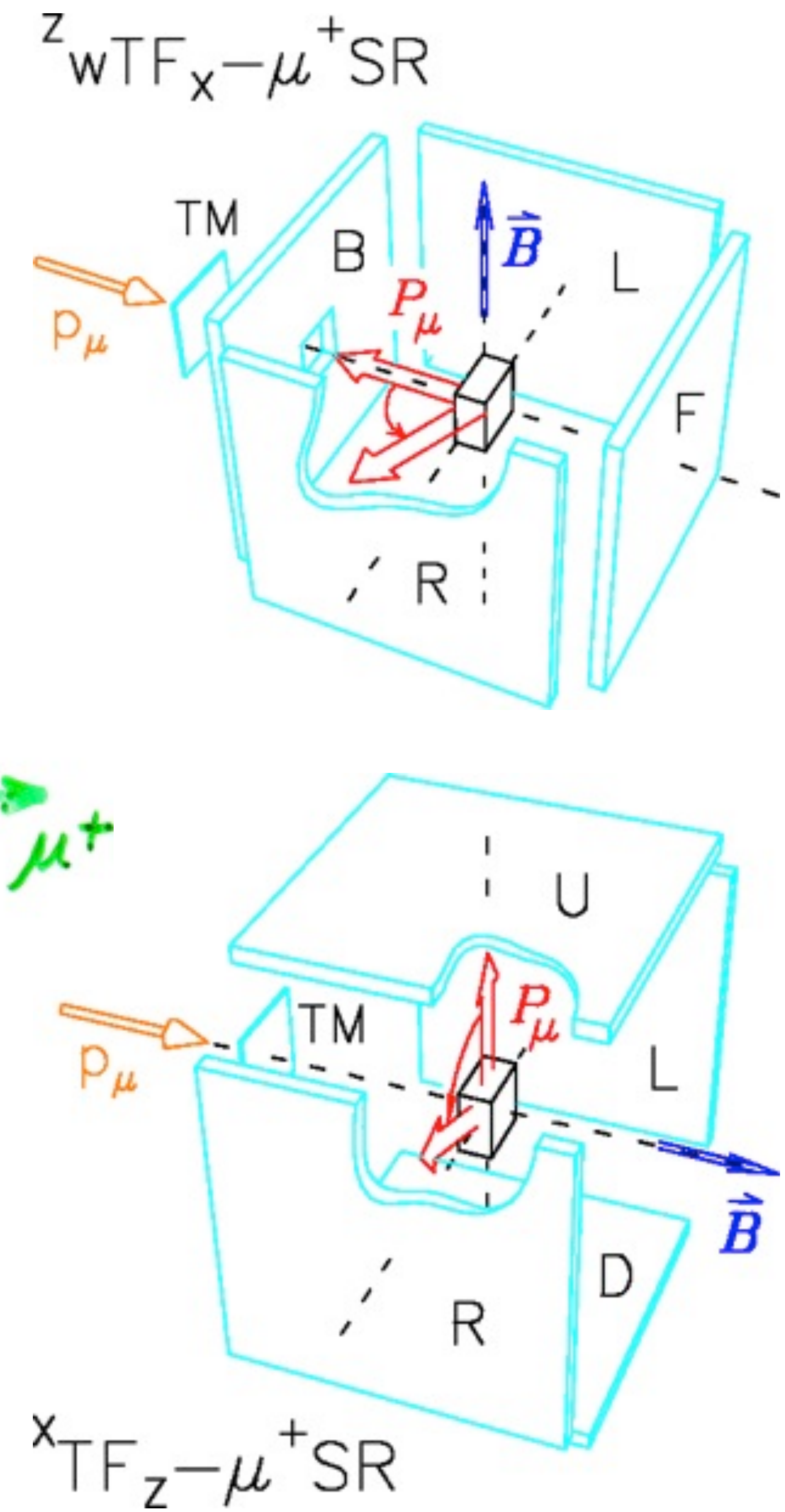
$E \times B$ velocity selector & Spin Rotator

("DC Separator" or Wien filter)

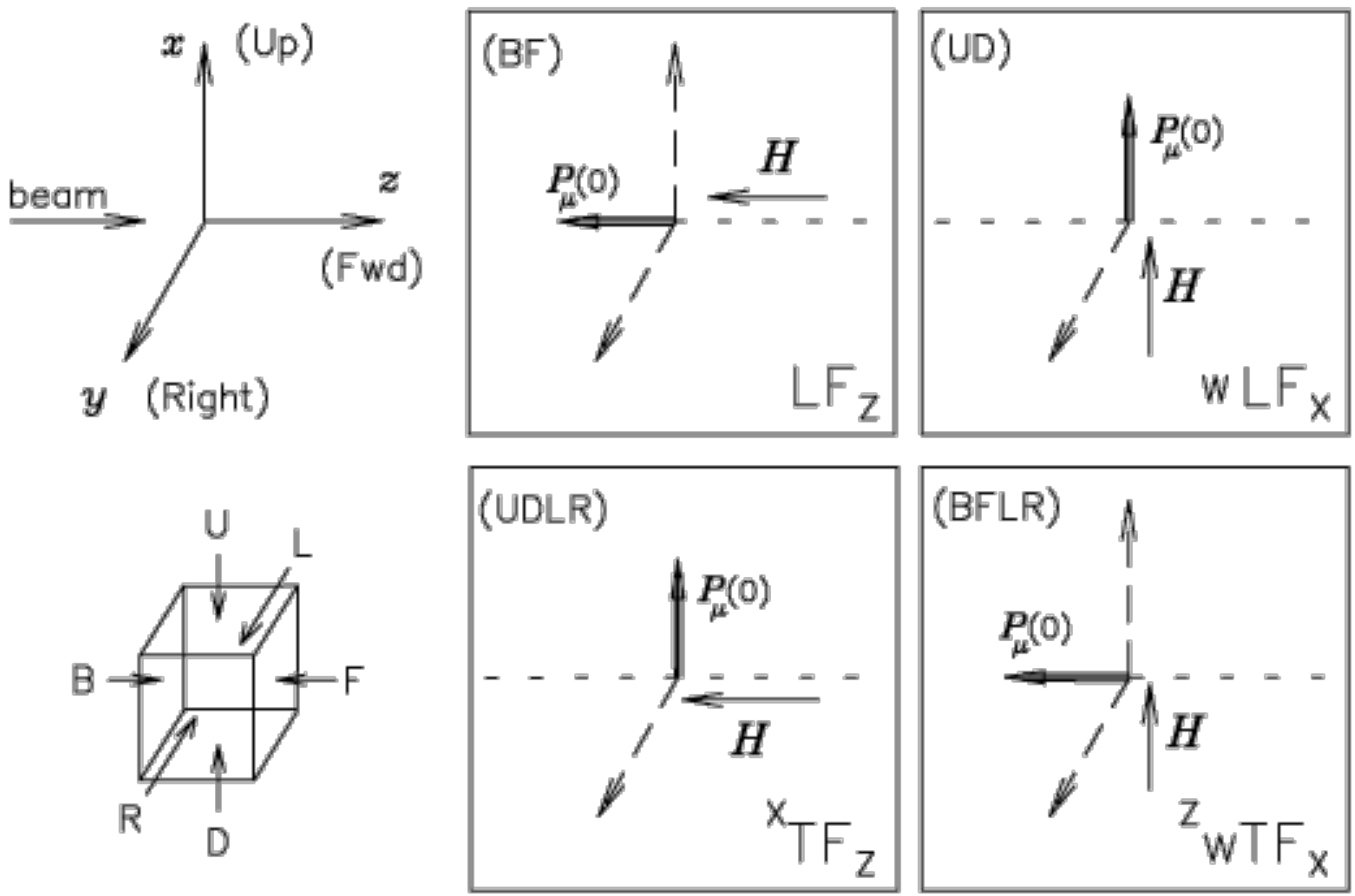
for **surface muons**:



- Removes beam positrons
- Allows TF-μ^+SR in high field
  (otherwise B deflects beam)



Coordinate Conventions



High Field μSR

Fields of up to 10 T are now available, requiring a “business end” of the spectrometer < 3 cm in diameter (so that 30-50 MeV decay positron orbits don’t “curl up” and miss the detectors) and a time resolution of ~ 150 ps. Muonium precession frequencies of over 2 GHz have been studied.

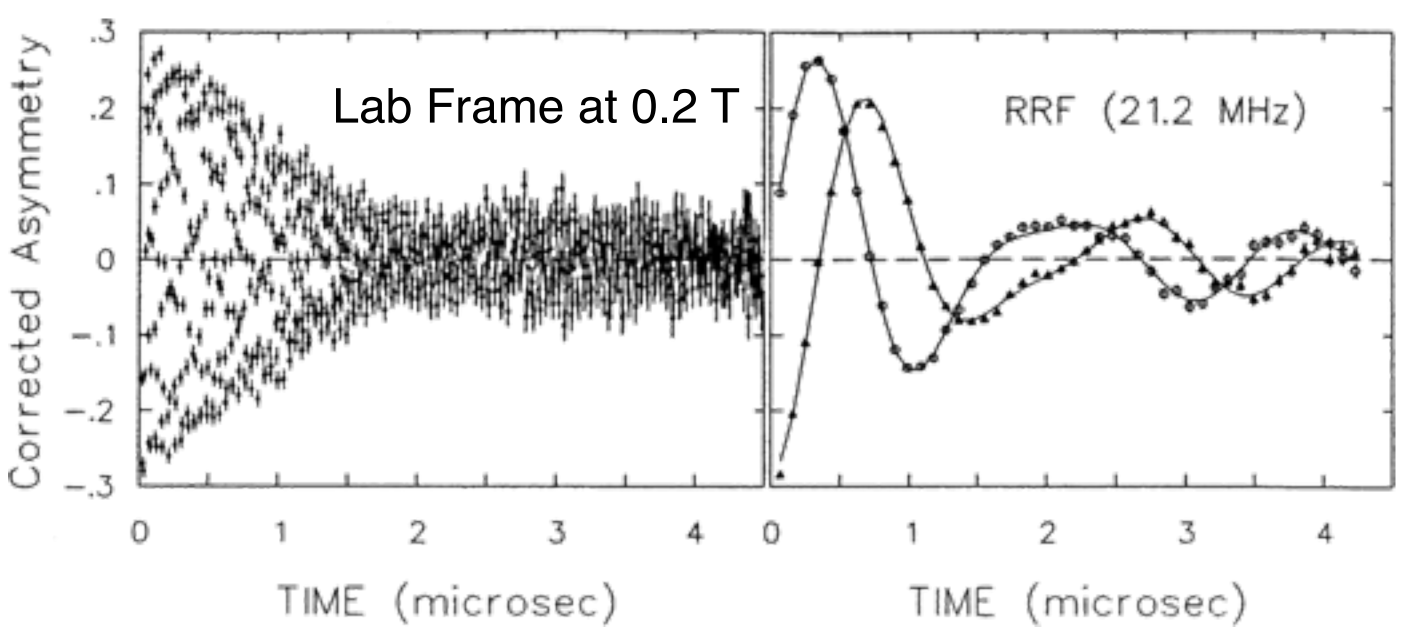
Rotating Reference Frame

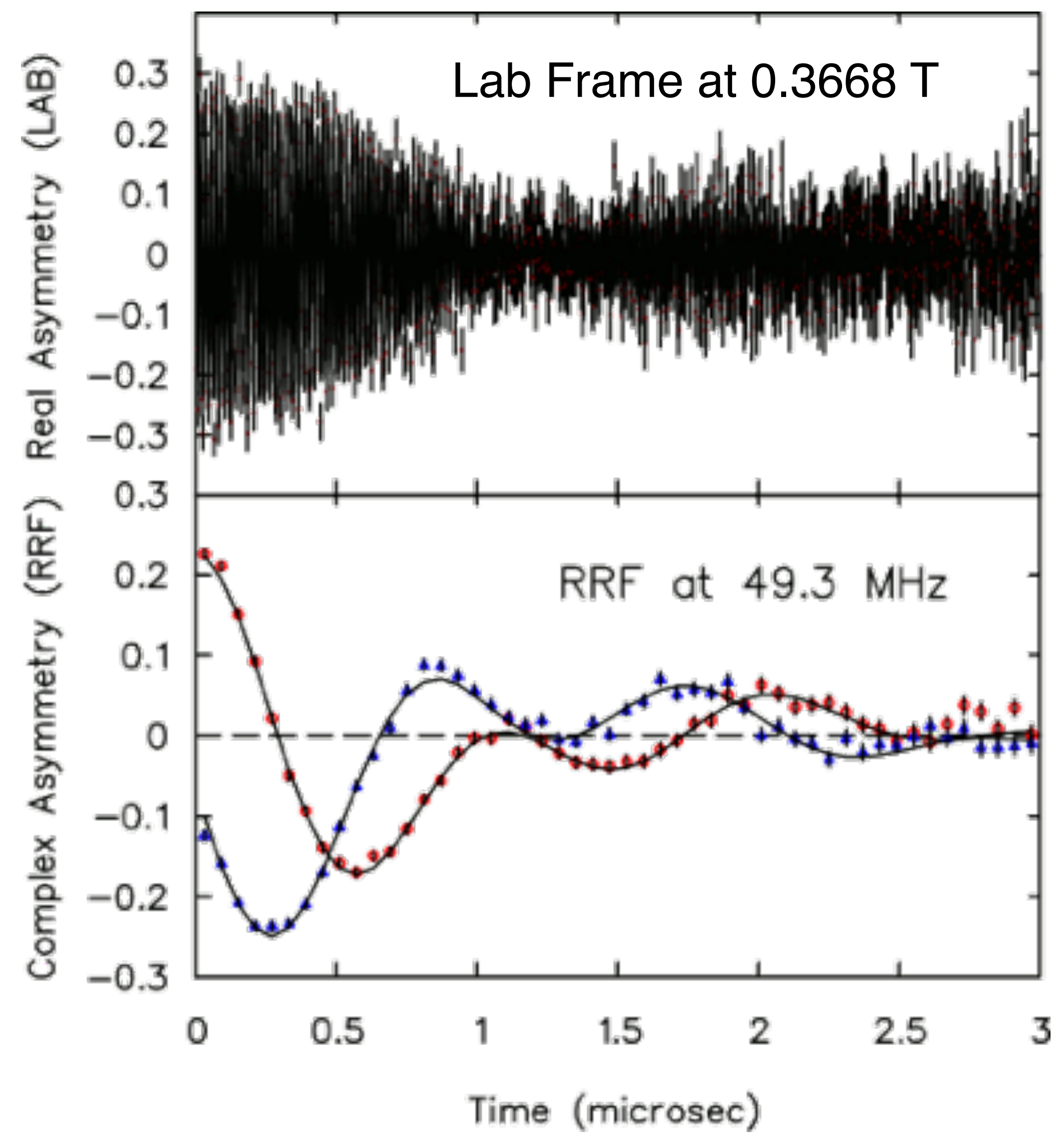
$$\mathcal{A}_{\mathrm{LAB}}(t) = \mathcal{A}_x(t) + i\mathcal{A}_y(t)$$

$$\mathcal{A}_{\mathrm{RRF}}(\Omega, t) = e^{-i\Omega t}\,\mathcal{A}_{\mathrm{LAB}}(t)$$

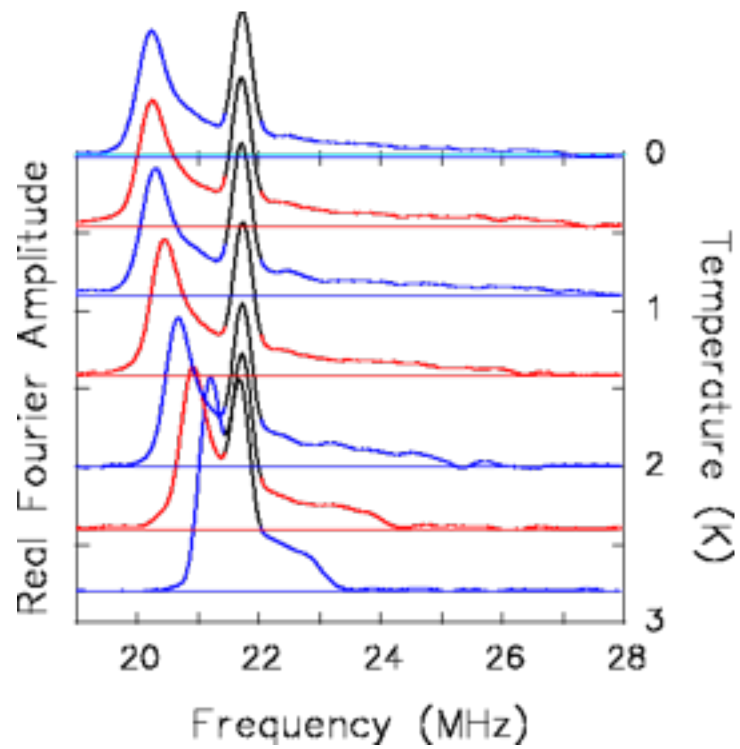
Muon spin precession in high transverse field (HTF-μSR) requires progressively smaller time bins to record the oscillations. These smaller bins capture fewer counts (lower statistics) and require more calculations for fitting. Worse yet, the essential characteristics of the data are not readily observed "by eye".

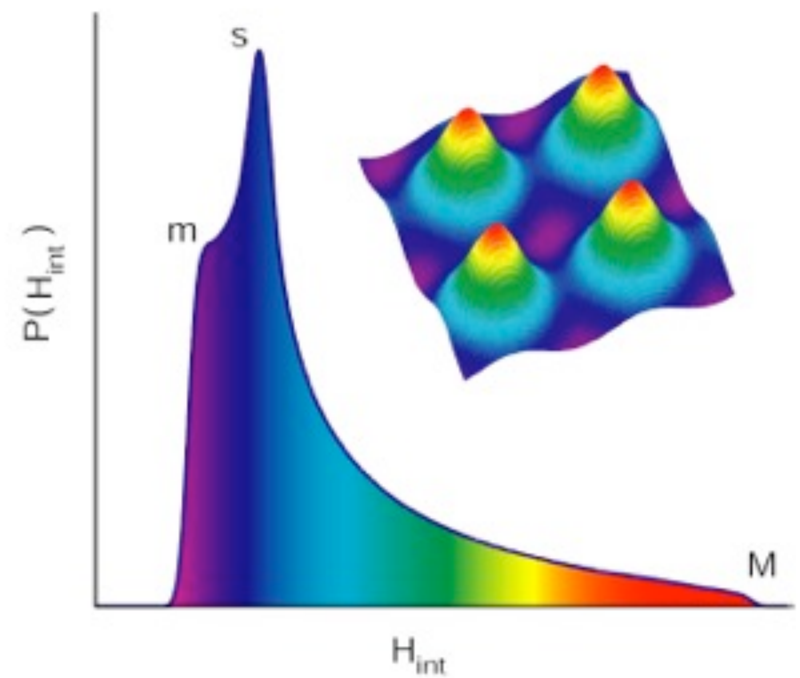
Fortunately, it is easy to convert the asymmetry spectrum into a Rotating Reference Frame (RRF) after the fact.

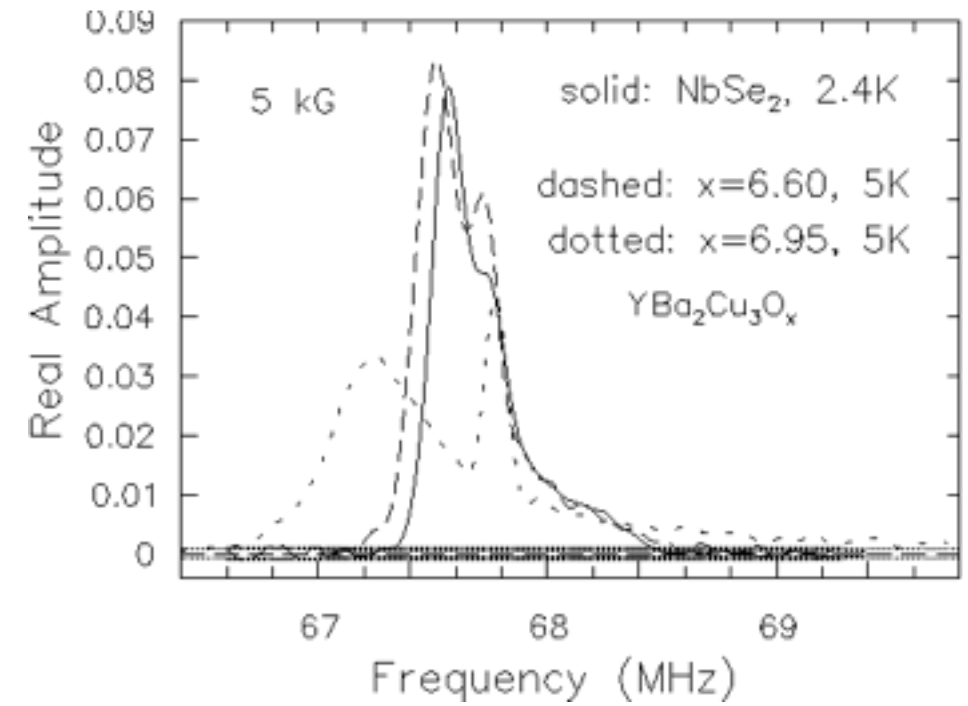


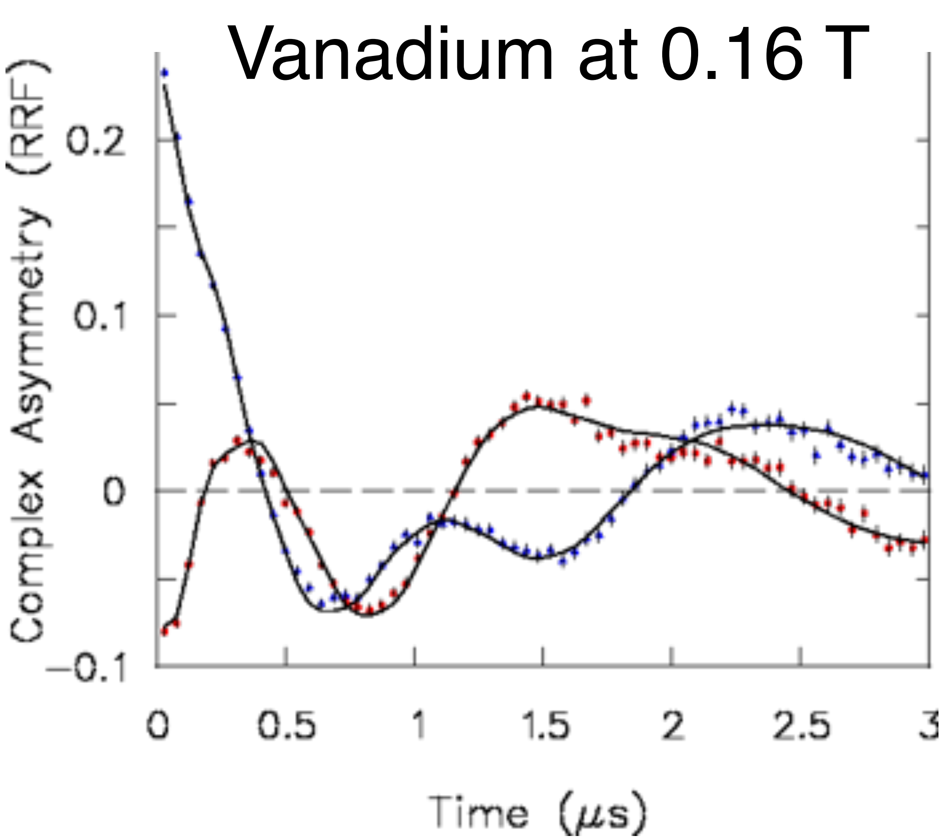


Type-II Superconductors





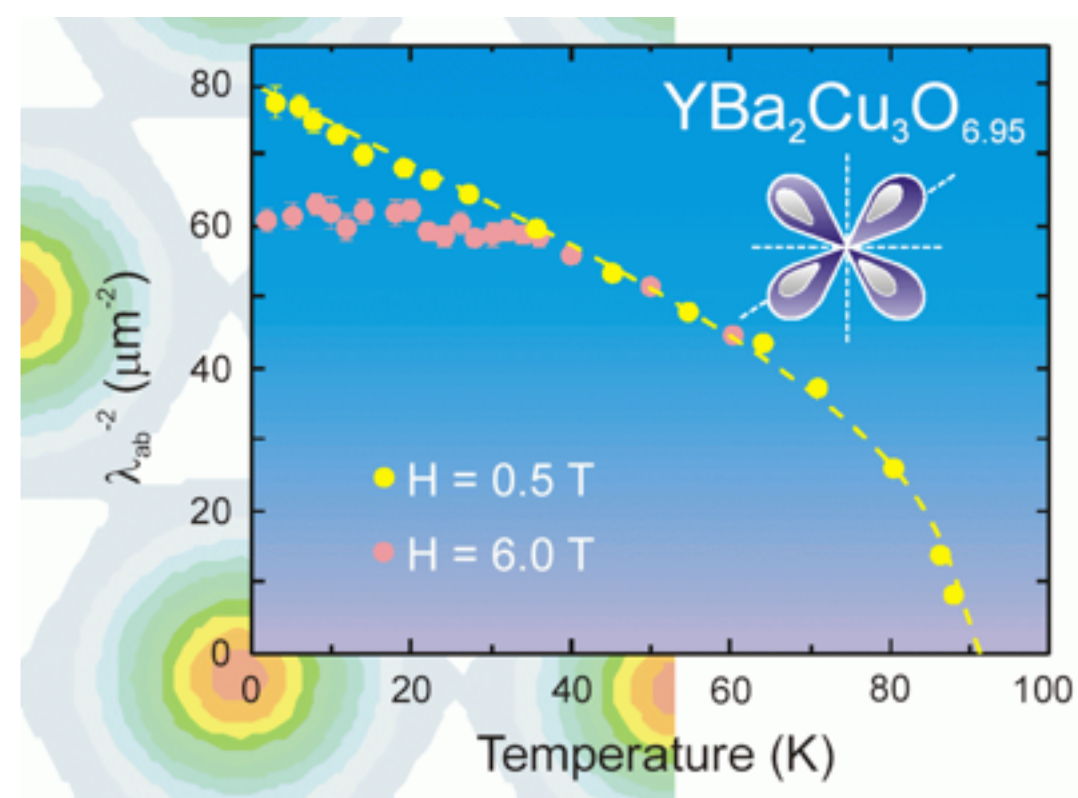




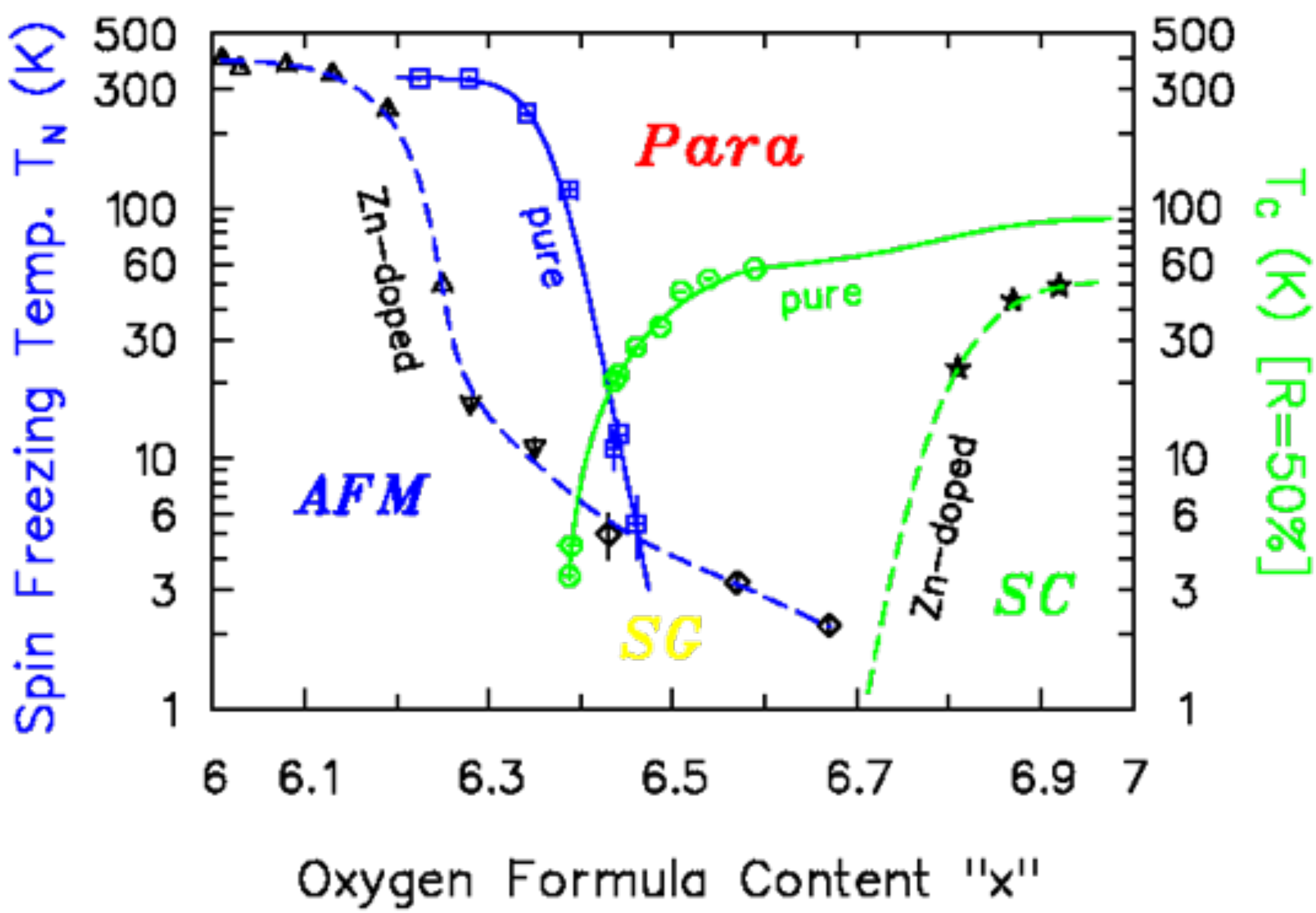
Fitting must always be done in the ***time*** domain, because of the "noise" from late times (low statistics).

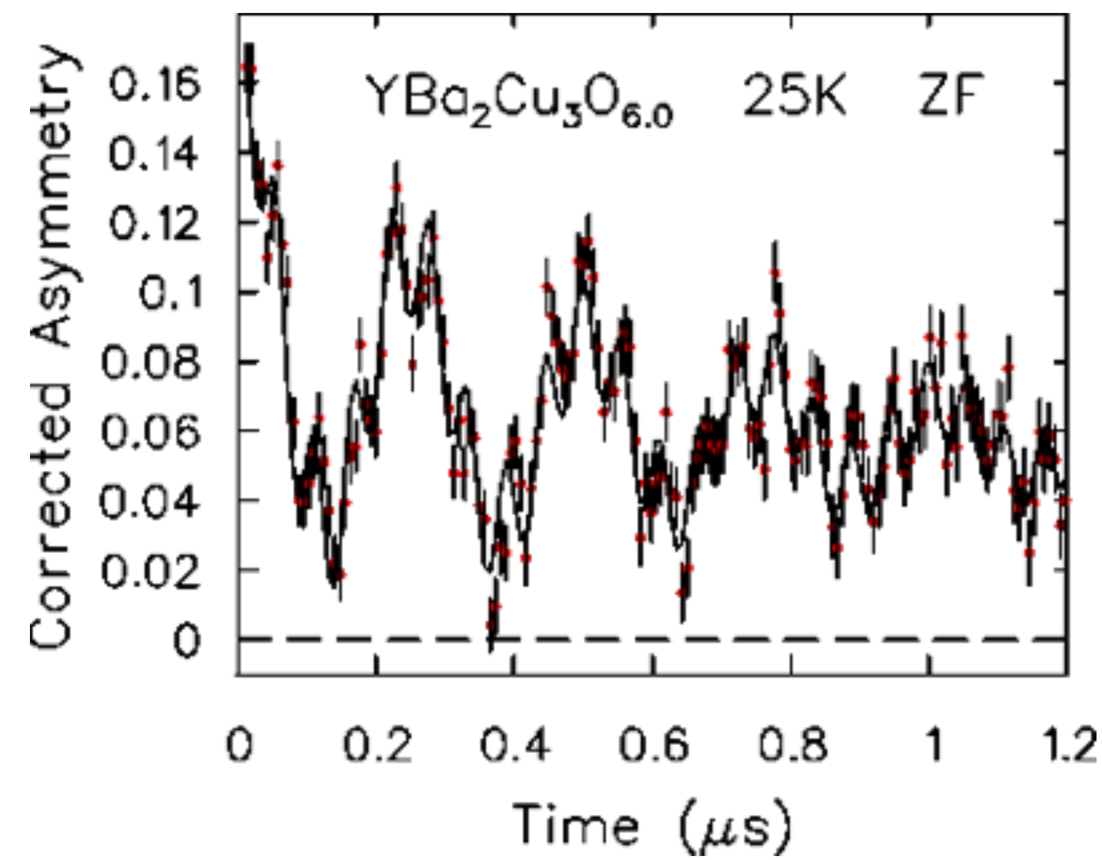
Extract magnetic penetration depth λ_{ab}

($\lambda_{ab}^{-2} \propto n_s$).



Coexistence of SC & Magnetism





Motion of Muon Spins in Static Local Fields

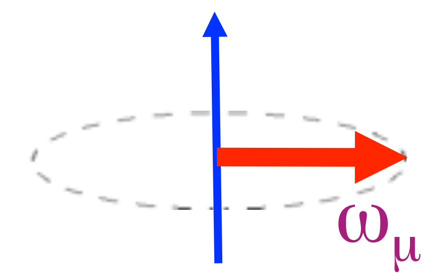
$\vec{S}_\mu$ = Expectation value of a muon's spin direction

(a) All muons "see" same field **B**: for **B** || $\mathbf{S}_\mu$ nothing happens

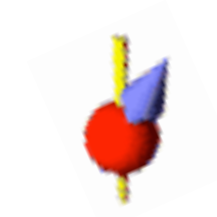
$\omega_\mu = 2\pi\, \gamma_\mu \, |\mathbf{B}|$

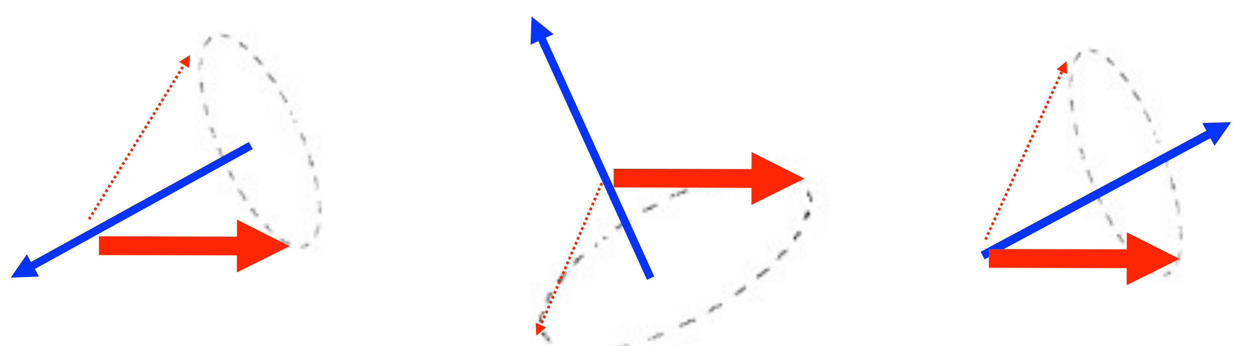
$\gamma_\mu = 135.5$ MHz/T

for **B** ⊥ $\mathbf{S}_\mu$ Larmor precession:



(b) All muons "see" same $|\mathbf{B}|$ but **random direction**:





2/3 of $\mathbf{S}_\mu$ **precesses** at ω_μ

1/3 of $\mathbf{S}_\mu$ **stays constant**

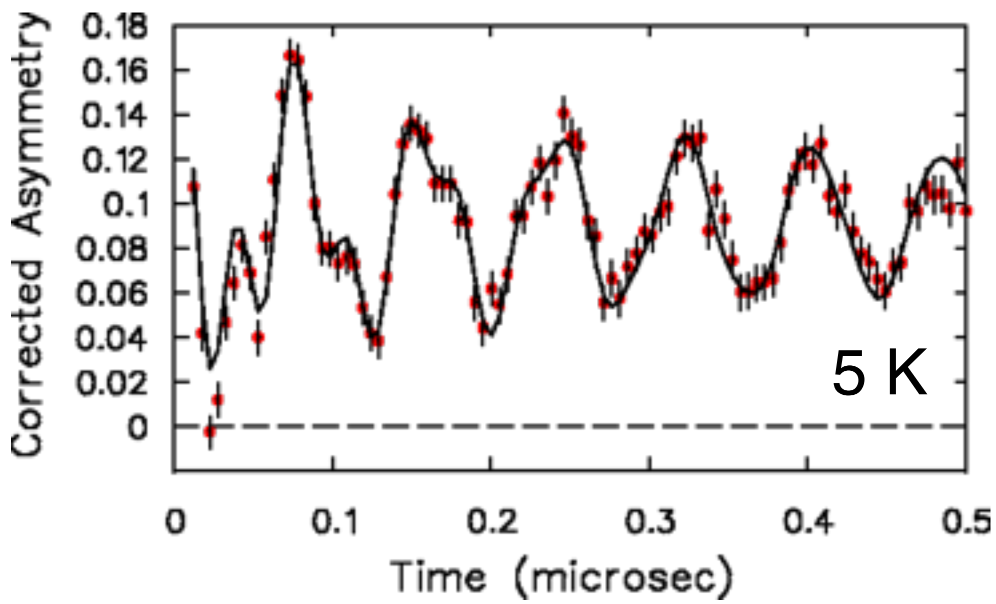
(c) Local field **B** **random** in **both** magnitude **and** direction:

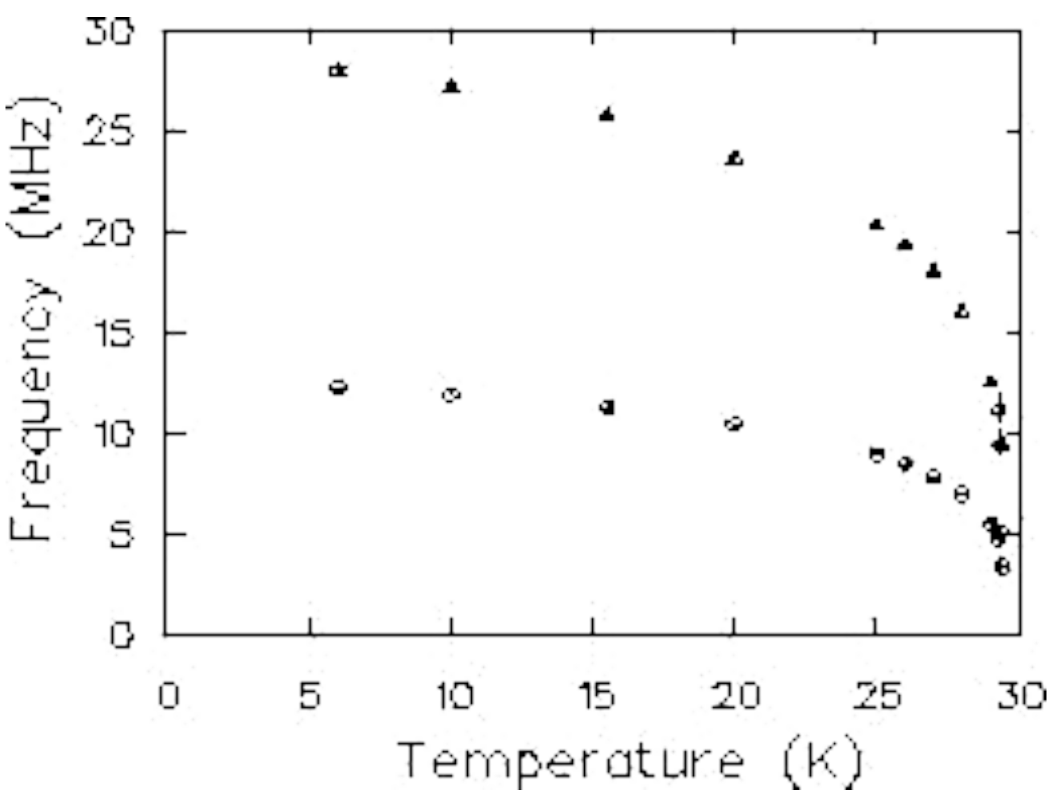
All ↗ do not return to the same orientation at the same time (dephasing) $\Rightarrow$ $\mathbf{S}_\mu$ "**relaxes**" as $G_{zz}(t)$ [Kubo & Toyabe, 1960's]

Saturday, August 30, 14

ZF/LF-μSR & Local Magnetic Fields

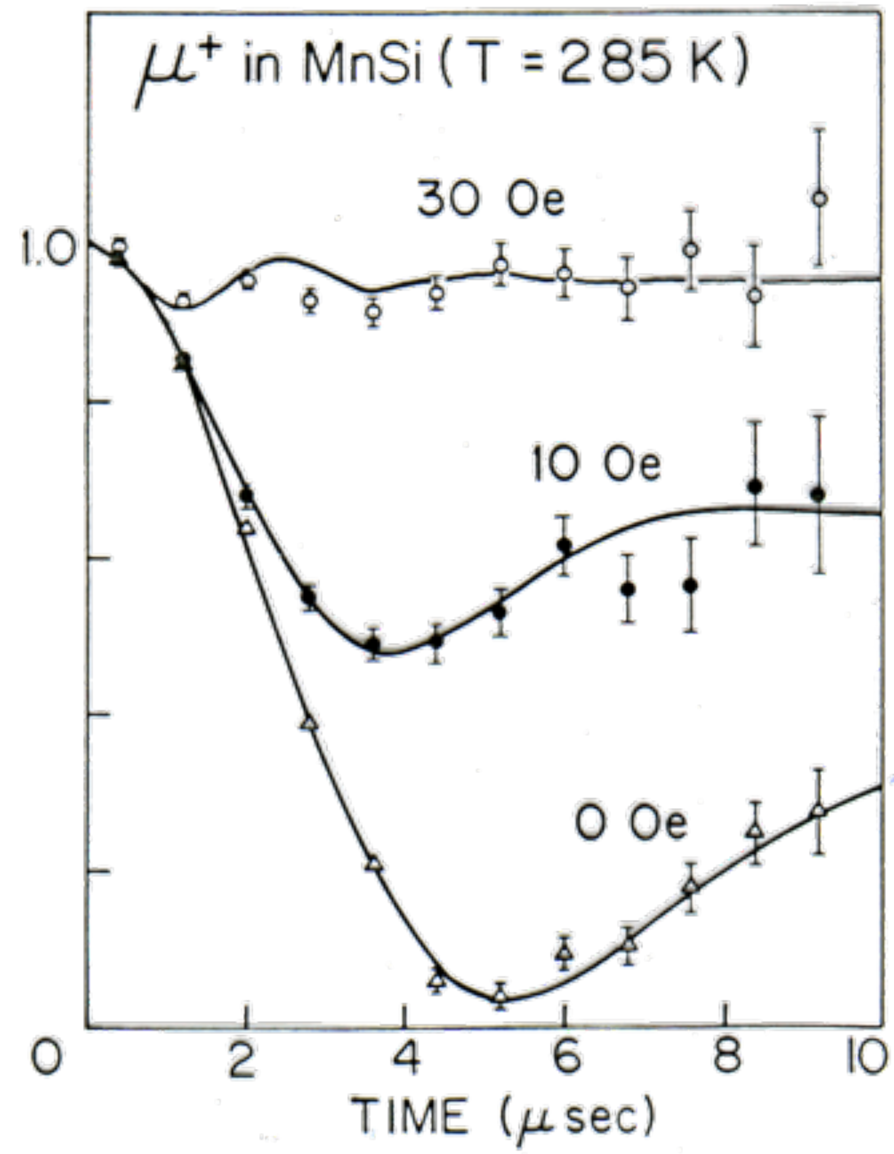
MnSi below 29 K: **helimagnetic**





In between, things get ***very interesting***...

MnSi at 285 K: relaxation by static **nuclear dipolar** fields (PM moments flip too fast)



Hayano *et al.* TRIUMF - 1987

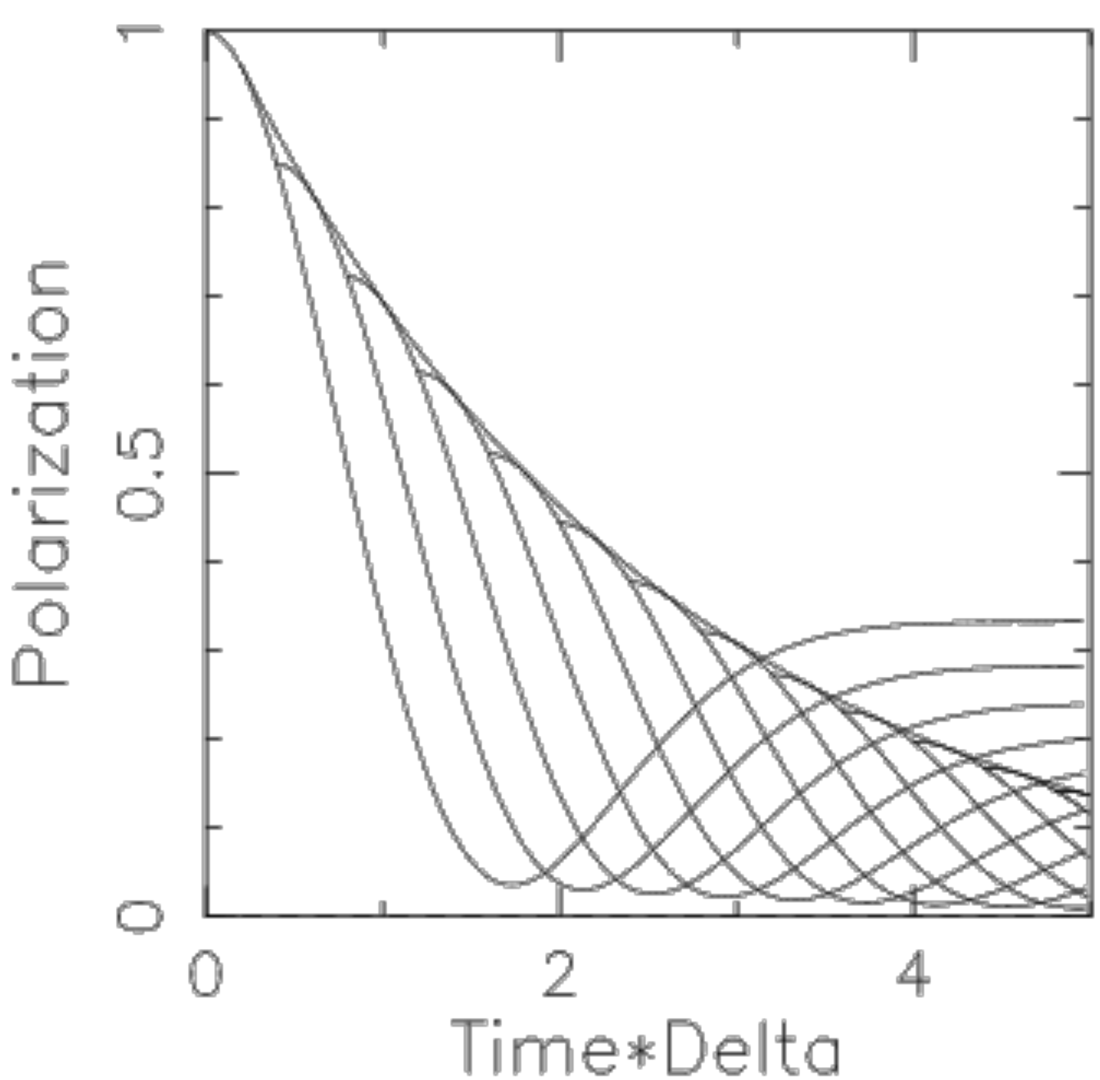
Decoupling by LF

Static Gaussian Kubo-Toyabe: $g^G_{zz}(t)$

$g^G_{zz}(t) = [1 + 2(1 - \Delta^2 t^2) \cdot \exp(-\Delta^2 t^2/2)] / 3$

Motion of μ⁺ Spins in Fluctuating Local Fields

"Strong Collision" model: local field is reselected at random from the same distribution each time a fluctuation takes place, either from muon hopping (plausible) or from reorientation of nearby moments (unlikely to change so completely).



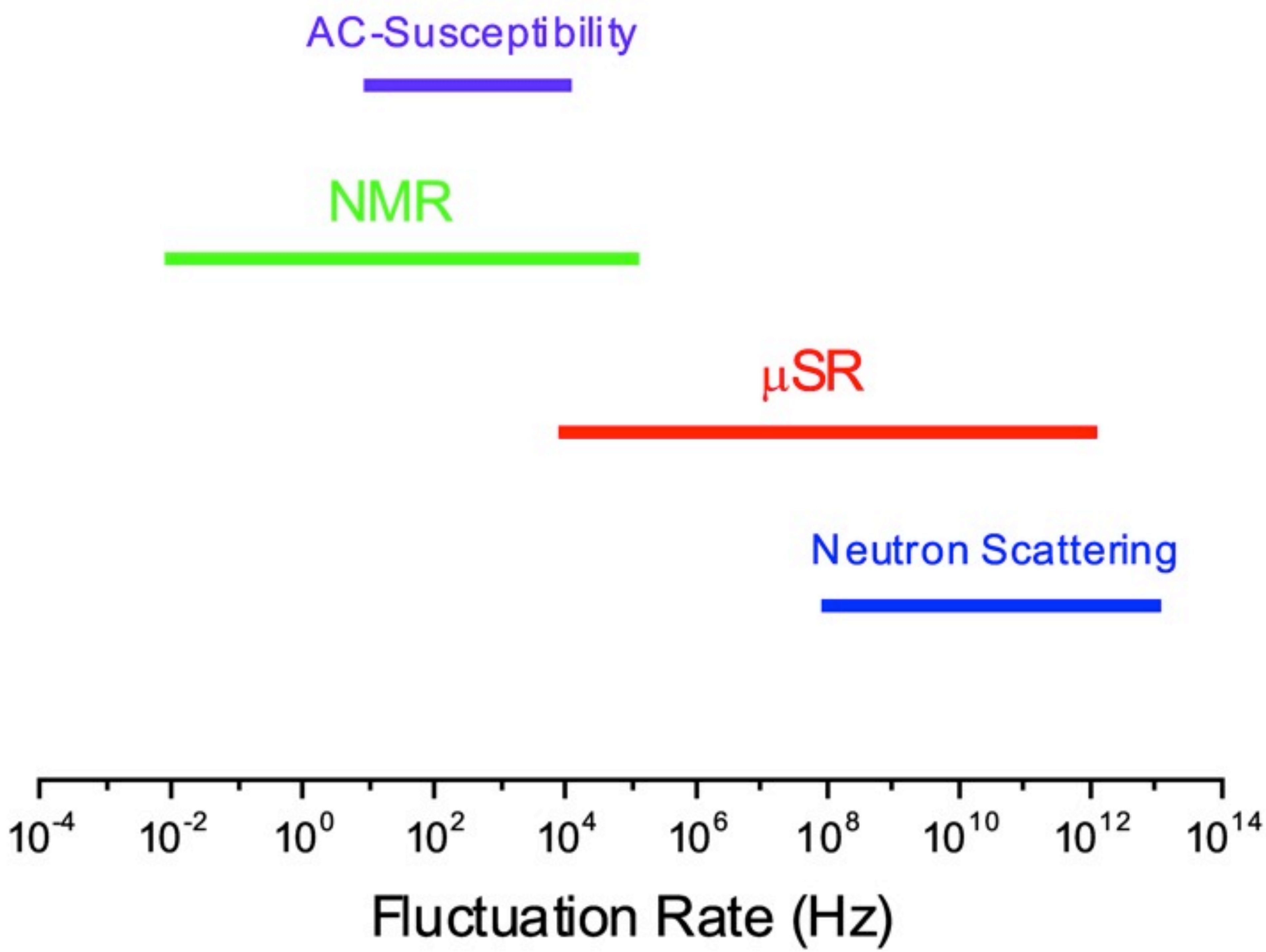
Kehr's recursion relation:

$$G(\Delta, t, \nu) = g(\Delta, t)e^{-\nu t} + \nu \int_0^t G(\Delta, t-\tau) g(\Delta, \tau)\, e^{-\nu\tau}\, d\tau$$

Sometimes solvable using Laplace transforms; numerical methods usually work too.

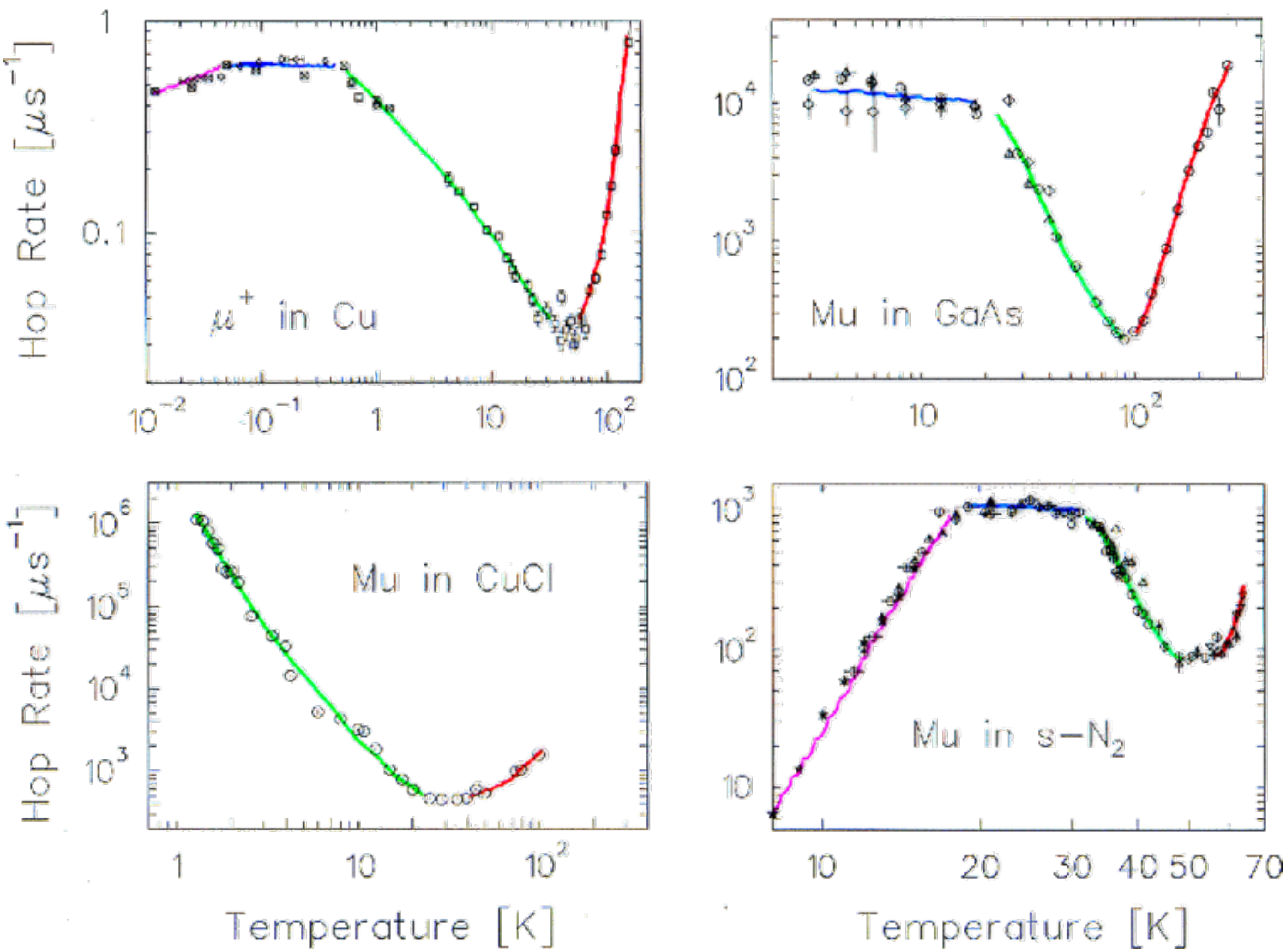
Used to extract "hop" or fluctuation rate ν.

Time Scales



AC-Susceptibility
NMR
μSR
Neutron Scattering
10-4 10-2 10^0 10^2 10^4 10^6 10^8 10^10 10^12 10^14
Fluctuation Rate (Hz)

Quantum Diffusion



Thermally activated over-barrier *hopping* (incoherent).

⬆ hot

Phonon scattering "spoils" coherent delocalization of *lattice polarons*.

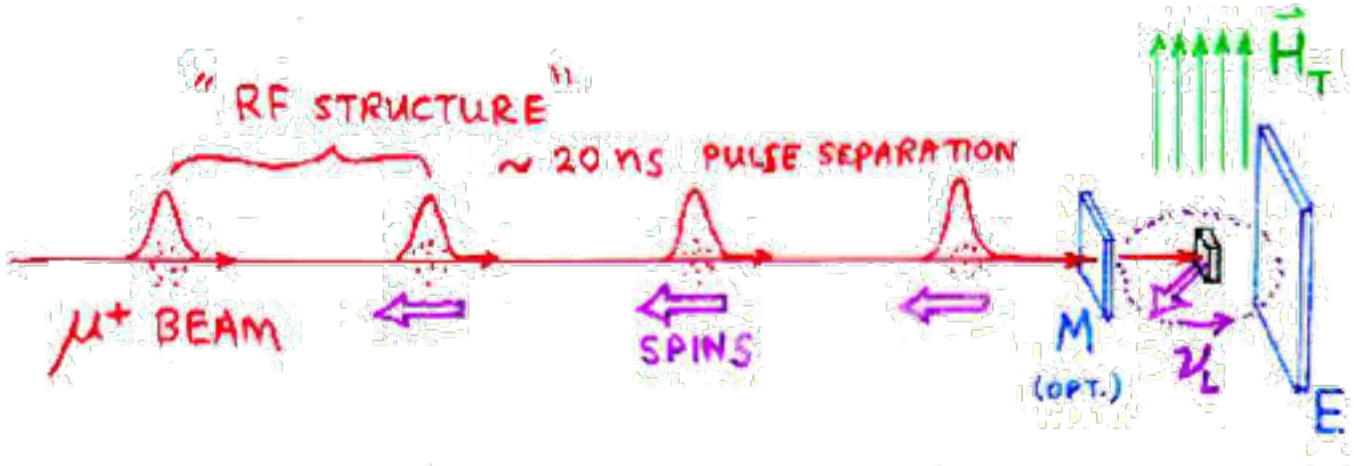
⬆ warm

Delocalized states find dilute defects and *trap*.

⬇ cold

Stroboscopic μSR

Schenck et al. - SIN



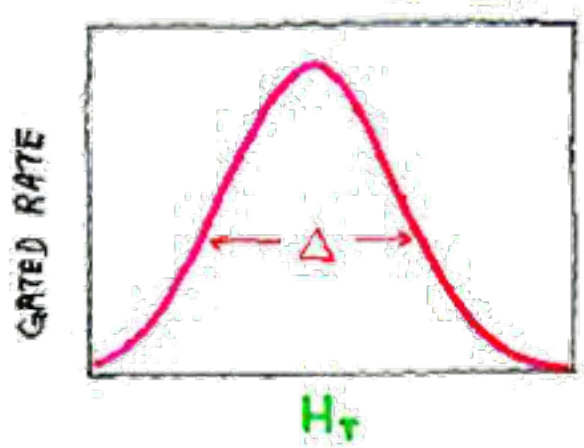
WHEN $1/\nu_L$ (PRECESSION PERIOD) is

$1/n$ × INTERVAL BETWEEN MUON PULSES,

↑ integer (~1 or 2)

ALL MUONS ARRIVE "IN PHASE" AND GATED COUNT RATE [AT APPROPRIATE TIME RELATIVE TO PULSE ARRIVAL] IS MAXIMIZED.

$$2\pi \nu_L = \gamma_\mu H_T$$

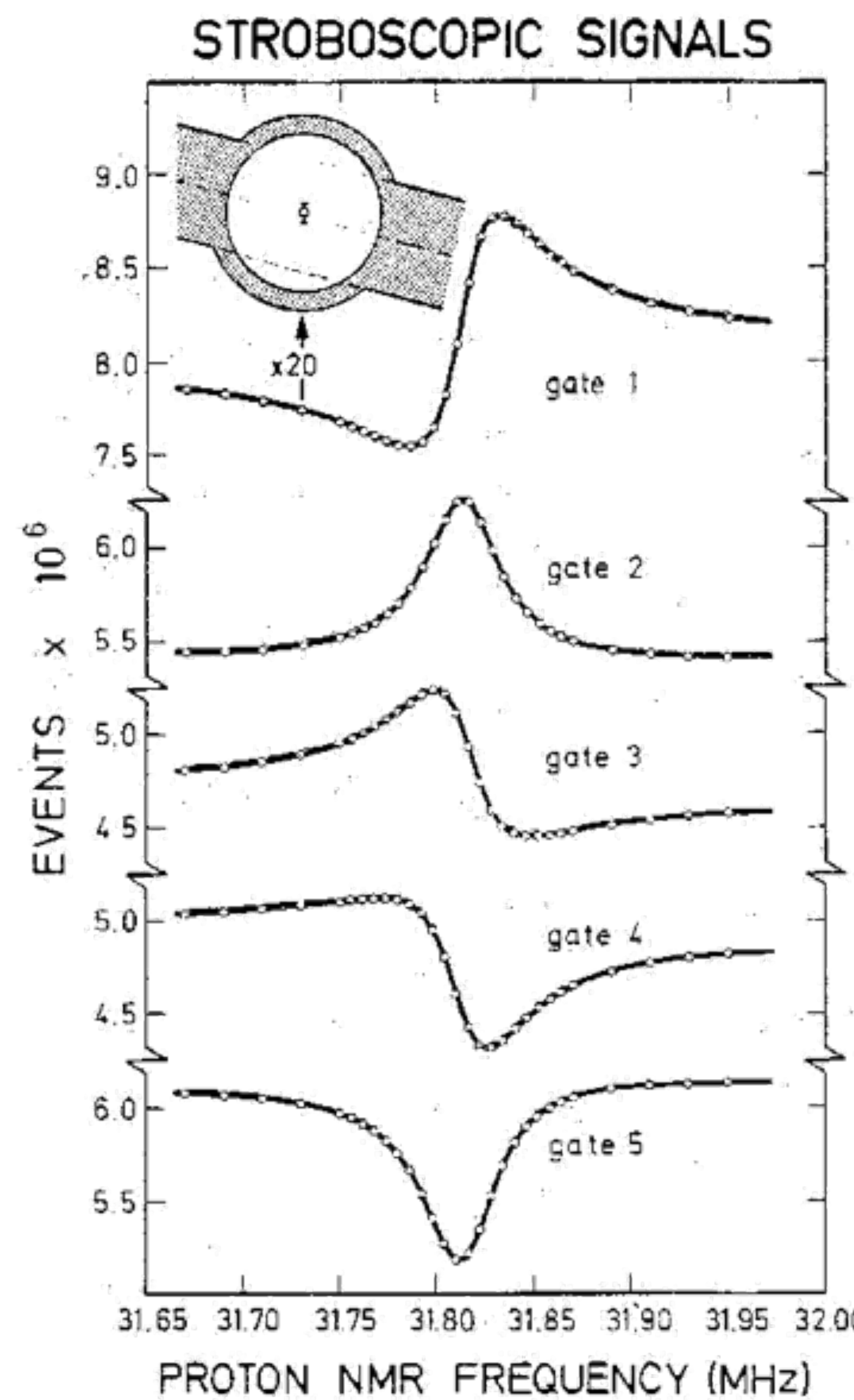


- NO RATE LIMIT! (MANY MUONS IN TARGET)

Gives KNIGHT SHIFTS to ~1 ppm

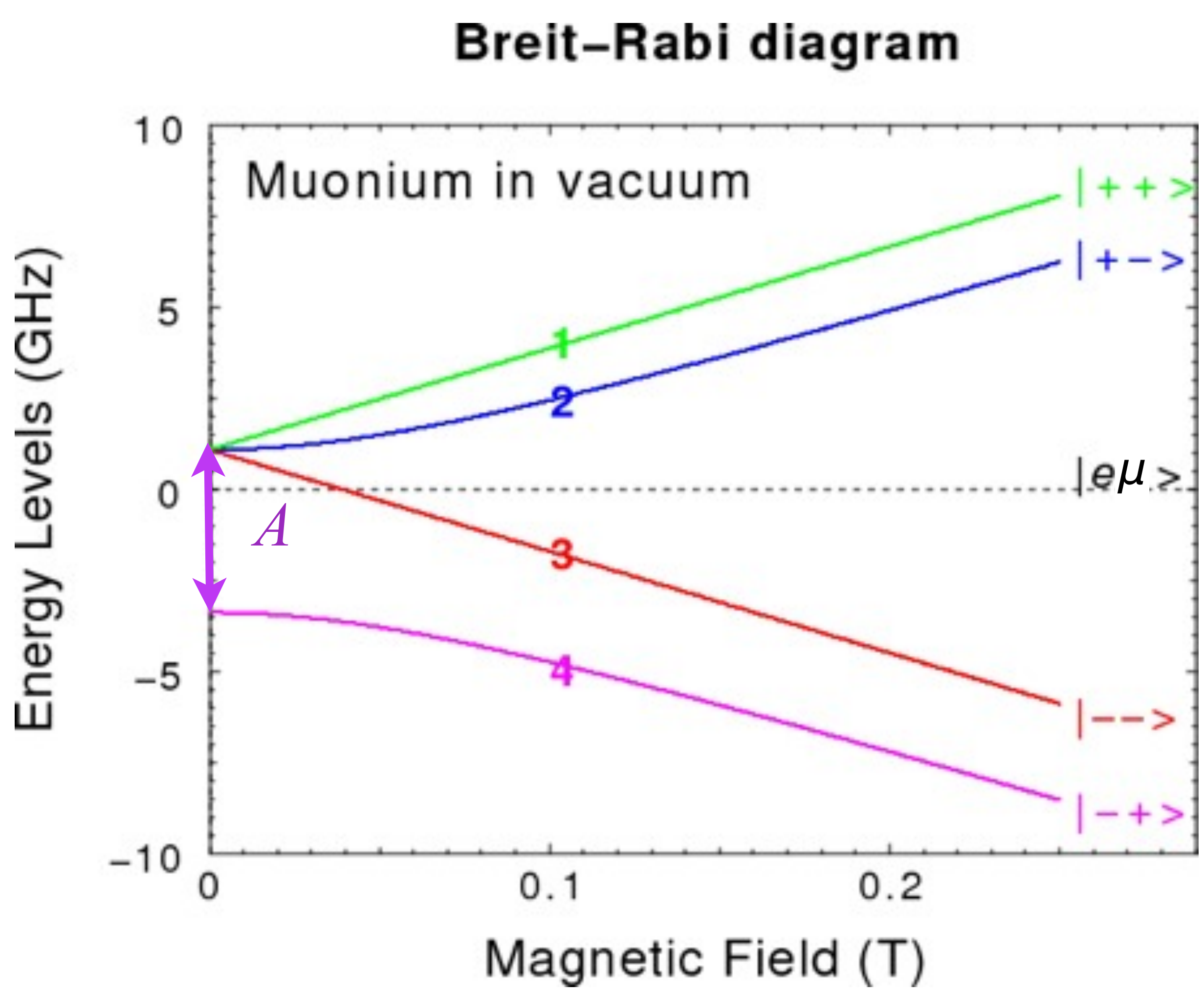
- PROBLEM: INTRINSIC WIDTH

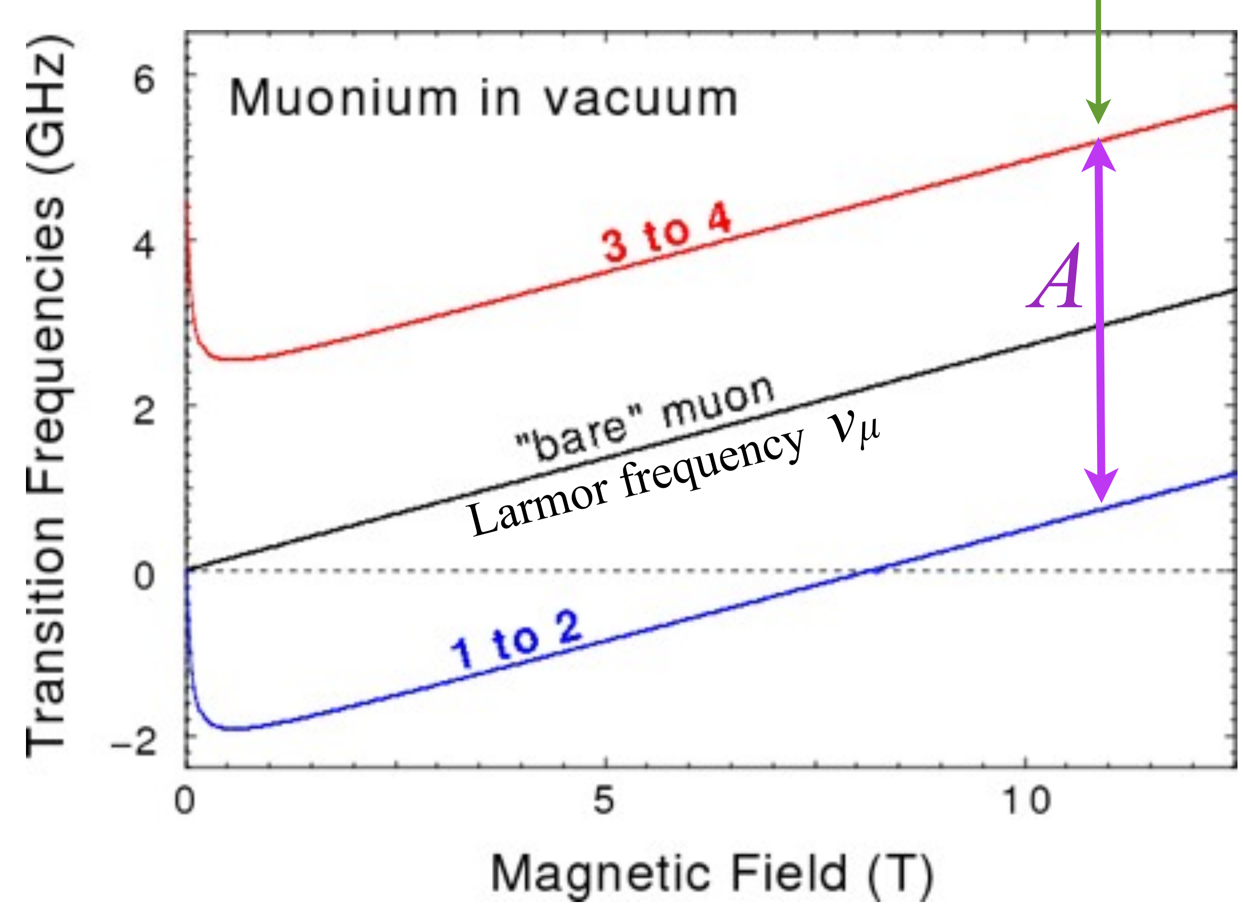
$$\Delta = \frac{1}{\tau_\mu} = 0.454\ \mu s^{-1}$$



Muonium (Mu≡μ^+e^-) *and FFT Spectroscopy*

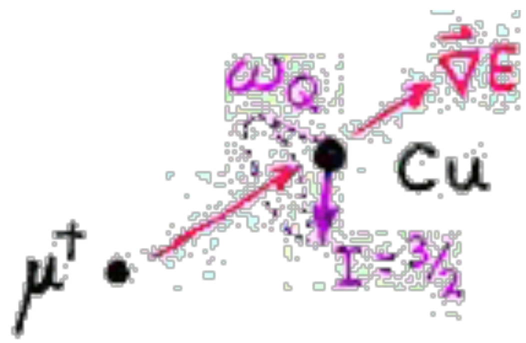
In a μSR experiment one measures a time spectrum at a given field and extracts ***all*** frequencies *via* FFT.

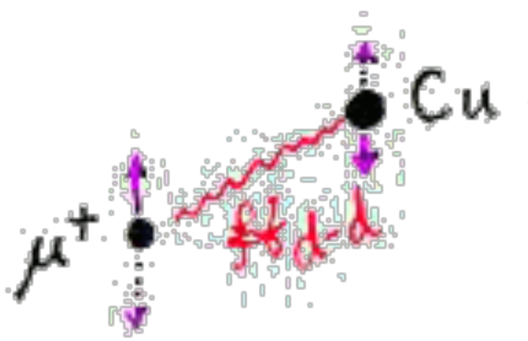


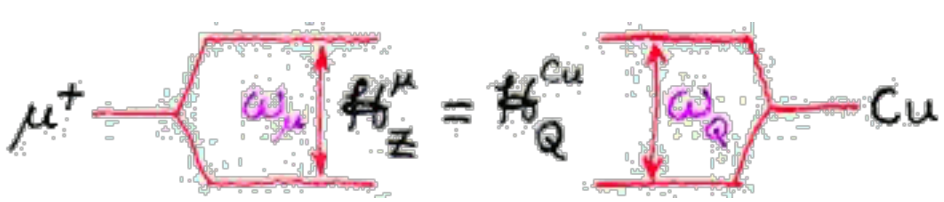


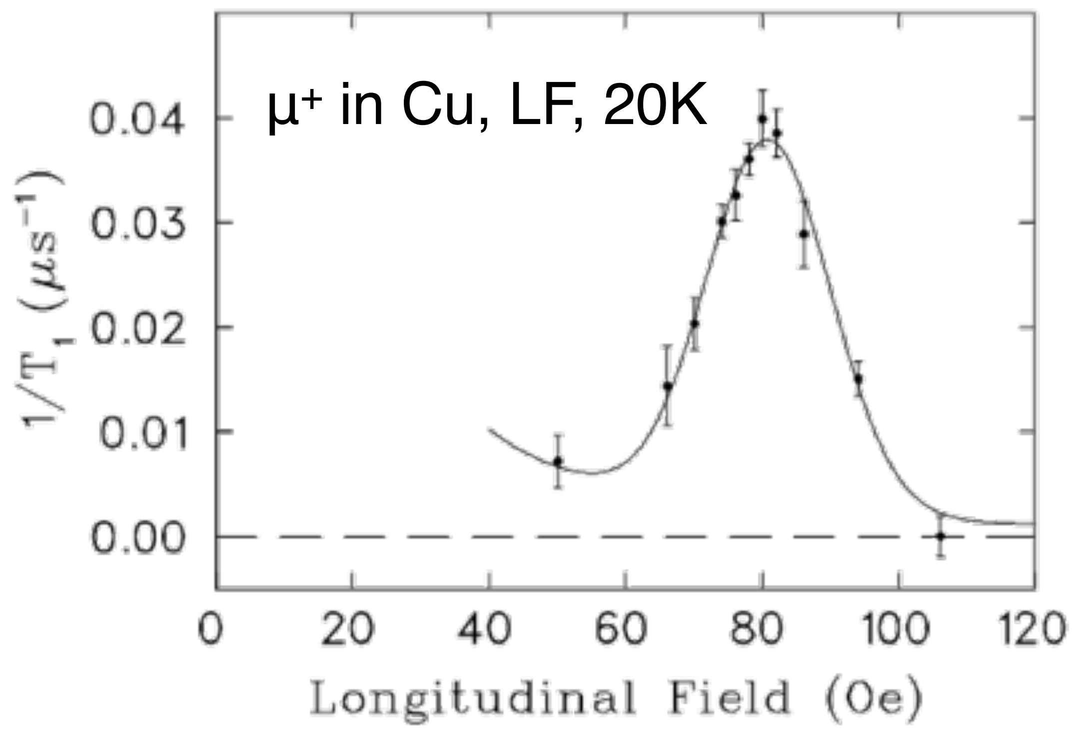
"**Signature**" of **Mu** (or other hyperfine-coupled μ^+e^- spin states) in **high transverse field**: ***two frequencies*** *centred on* ν_μ *and separated by the* ***hyperfine splitting A***$\propto r^{-3}$.

Avoided Level-Crossing Resonance

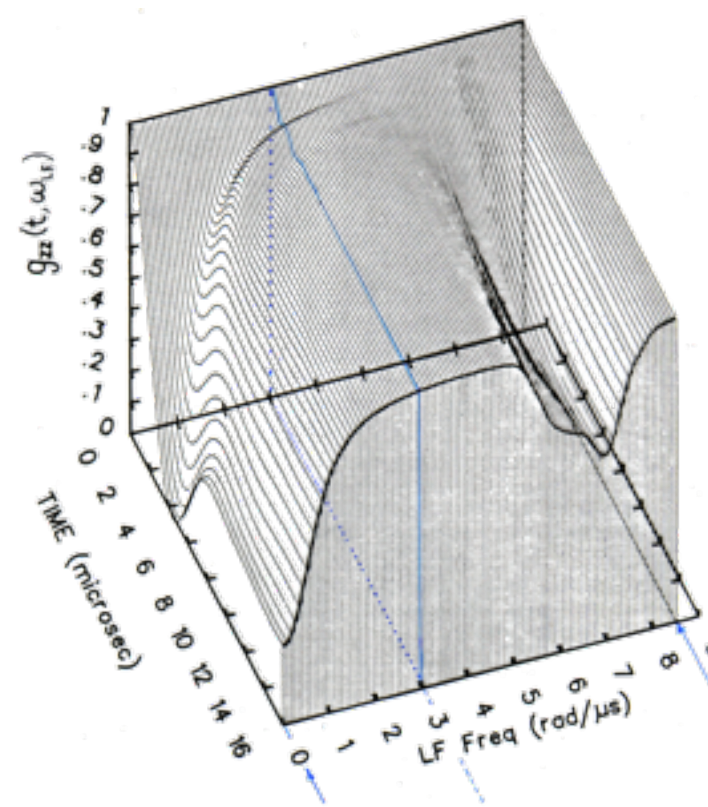




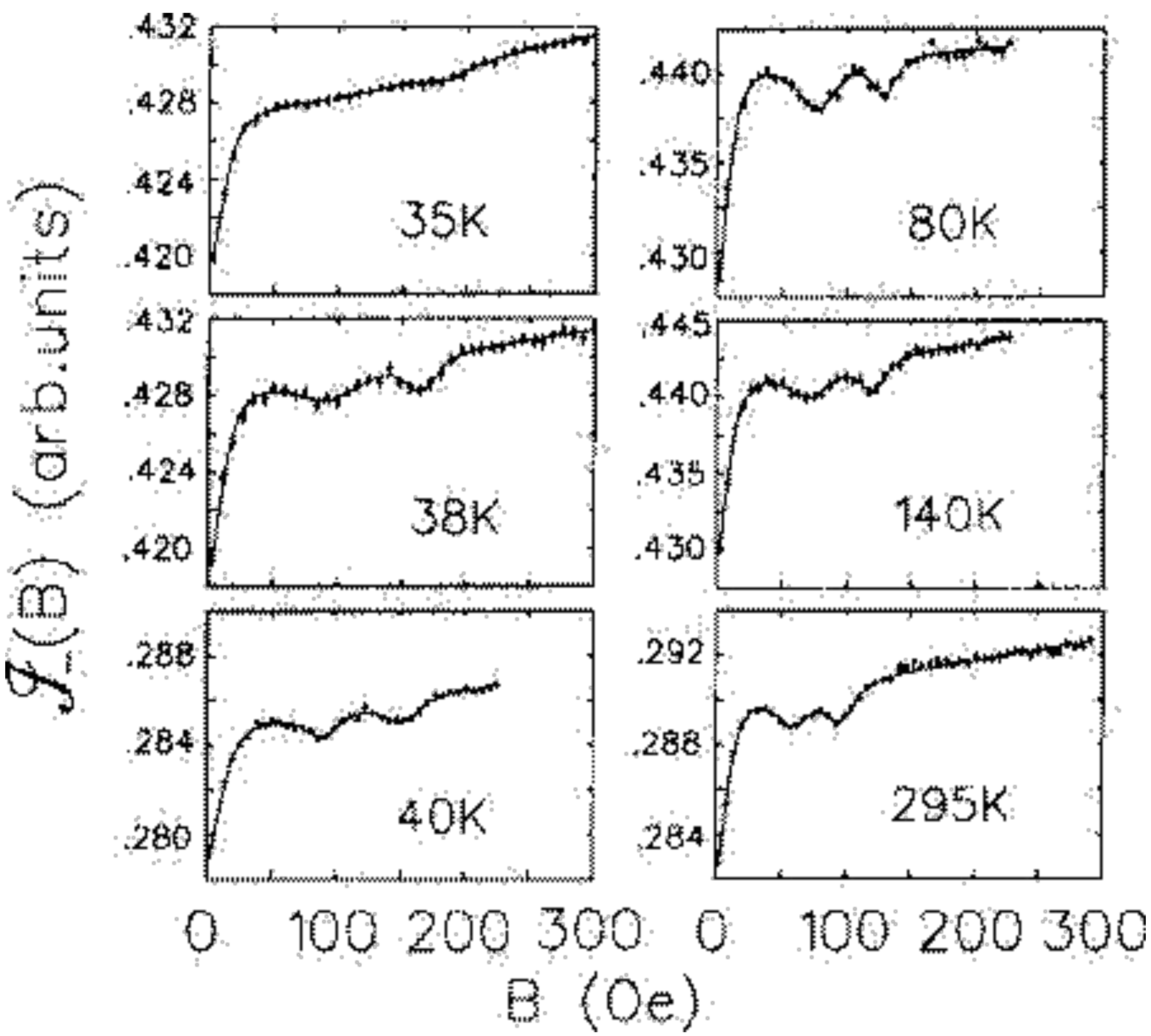


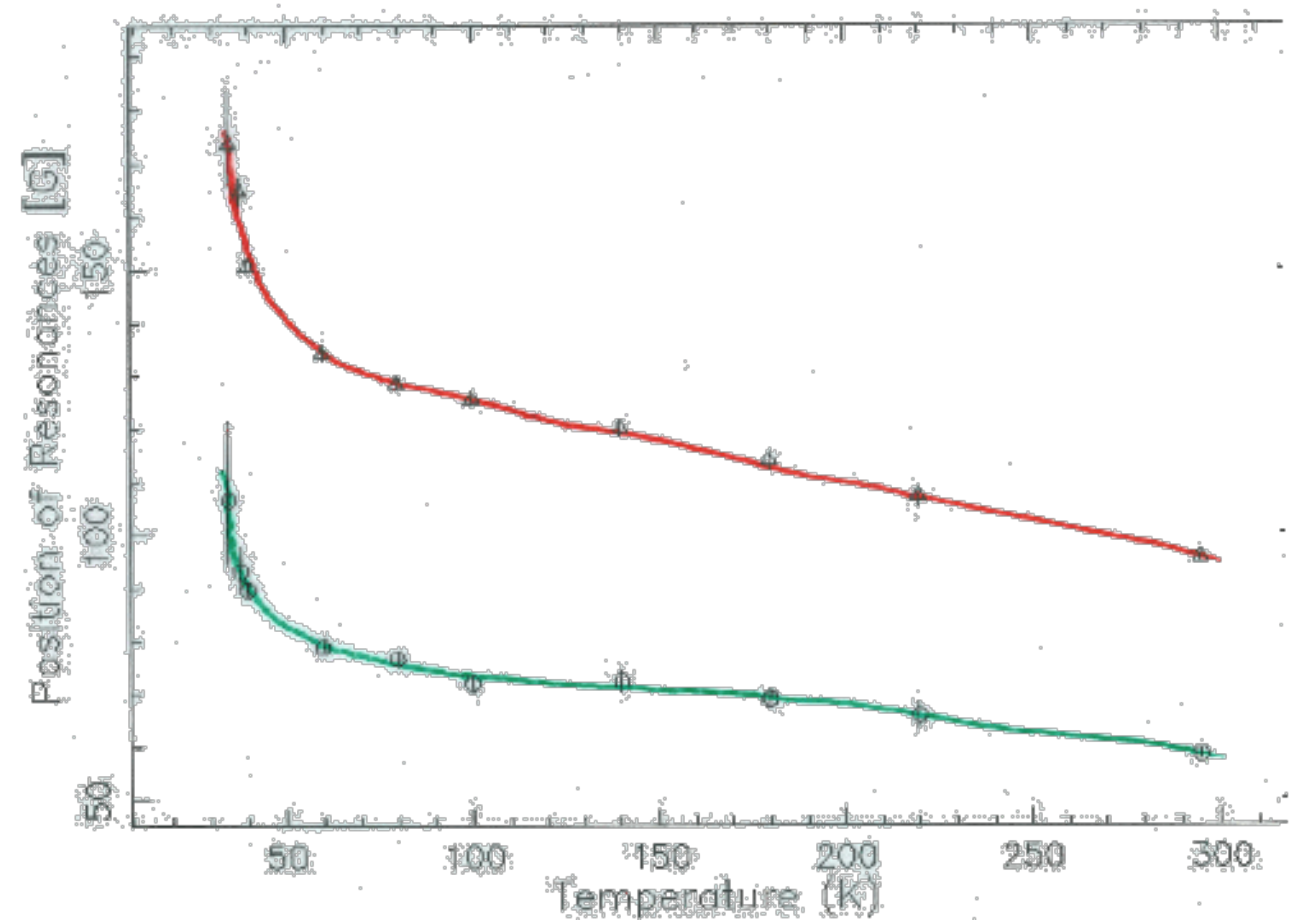


Nuclear Quadrupolar version

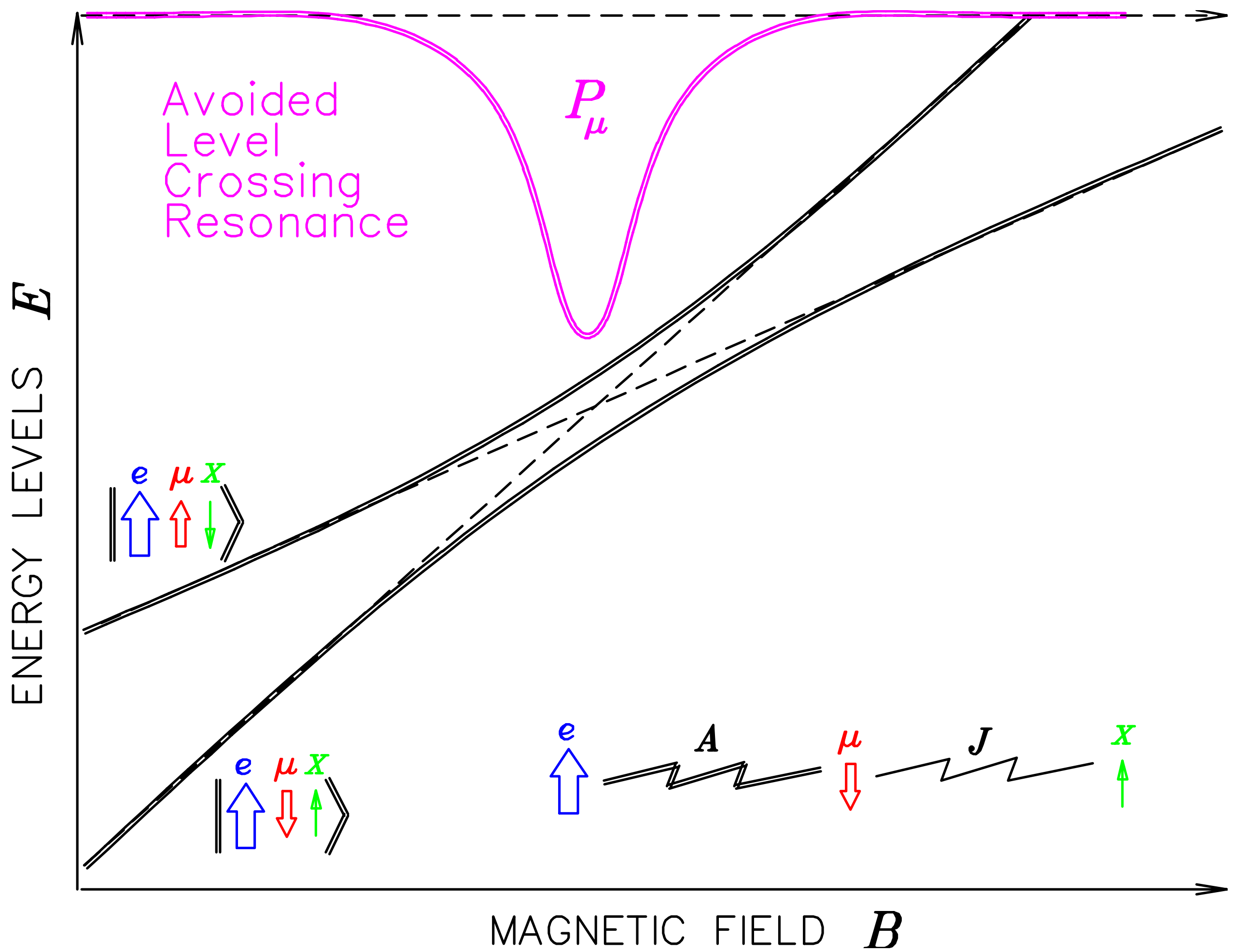


Avoided Level-Crossing Resonance Nuclear Quadrupolar version: MnSi



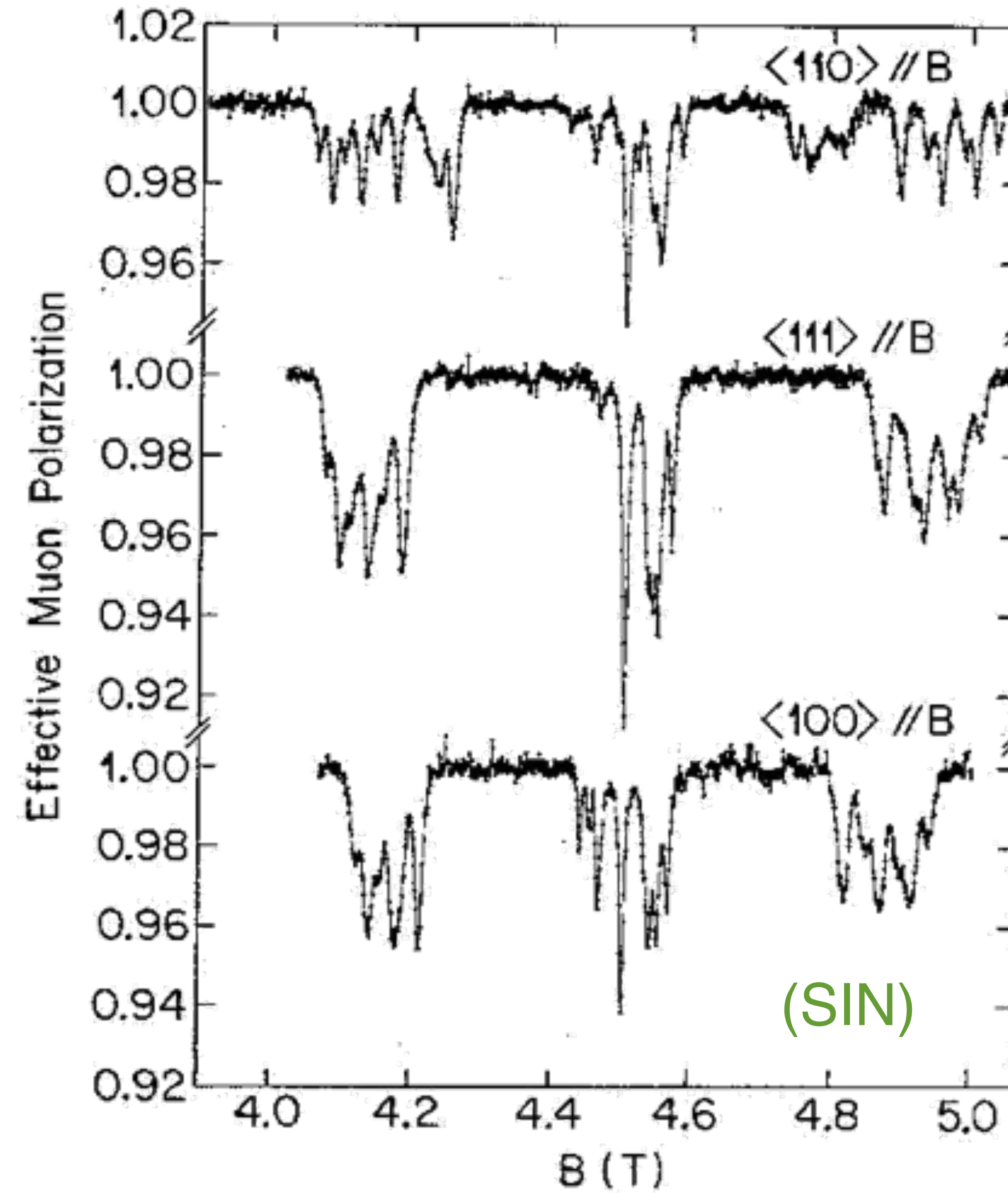


Avoided Level-Crossing Resonance

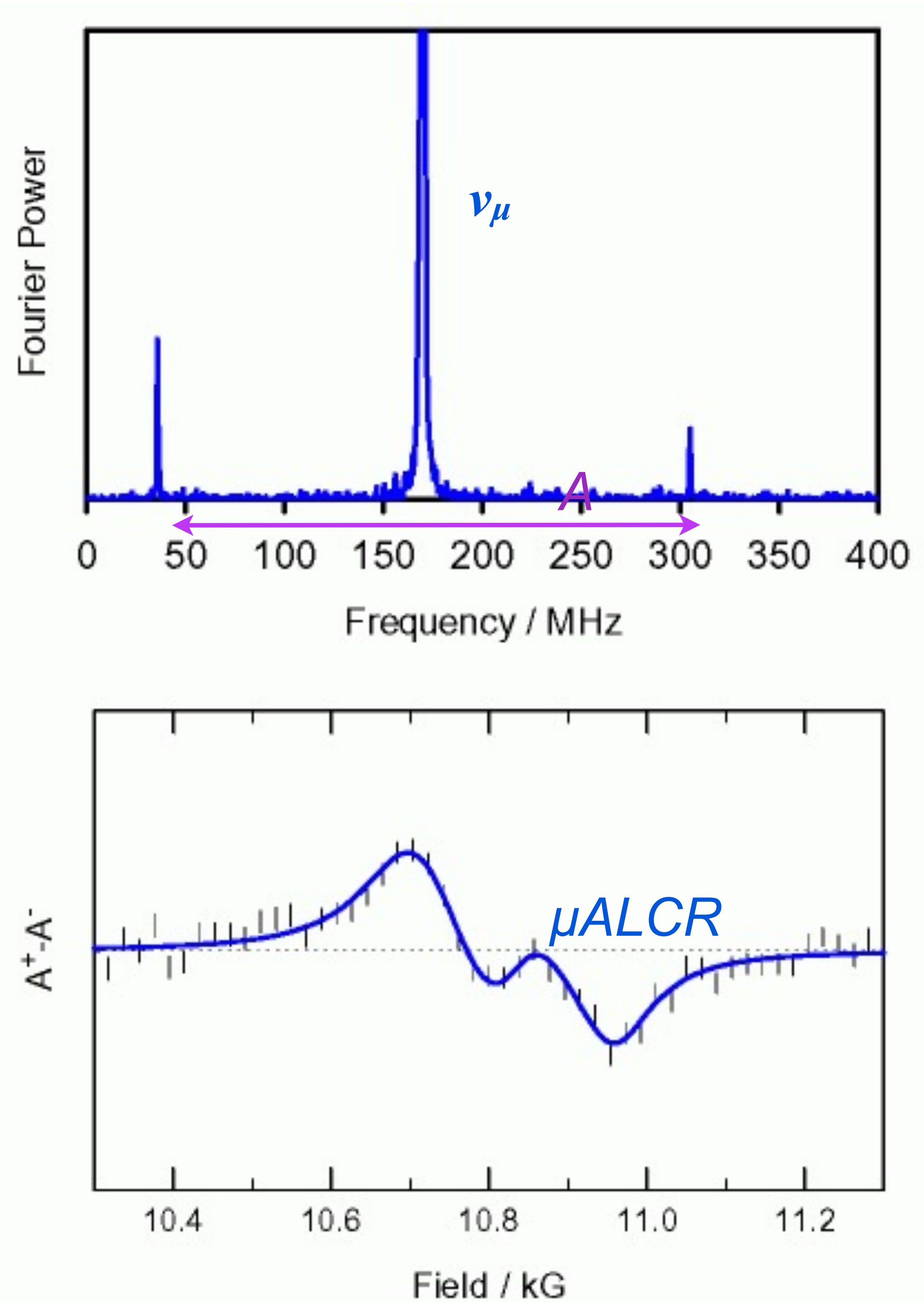


Nuclear Hyperfine version

e.g. **CuCl** *semiconductor*



Muonated Radicals



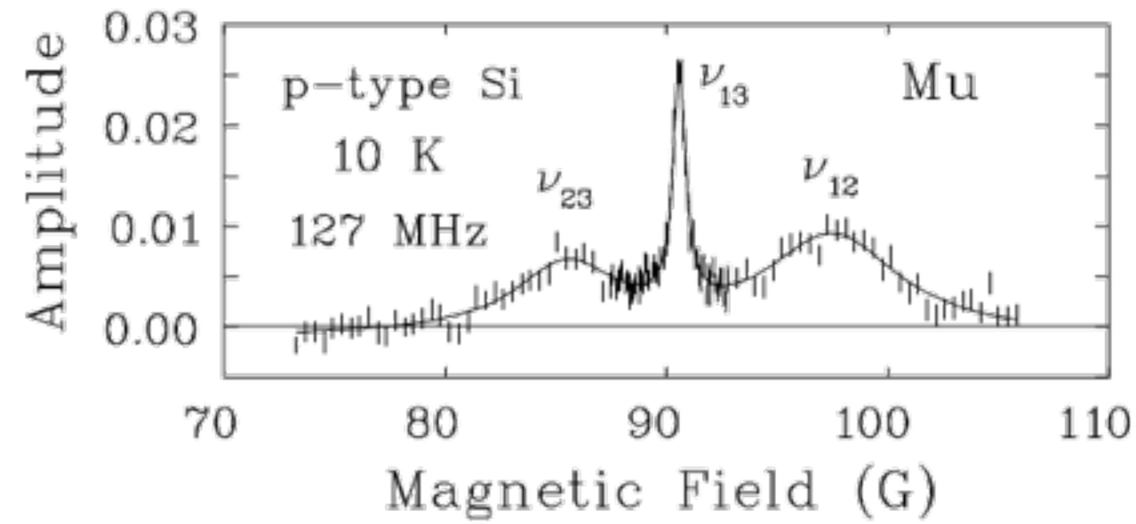
Organic Free Radicals in Superheated Water

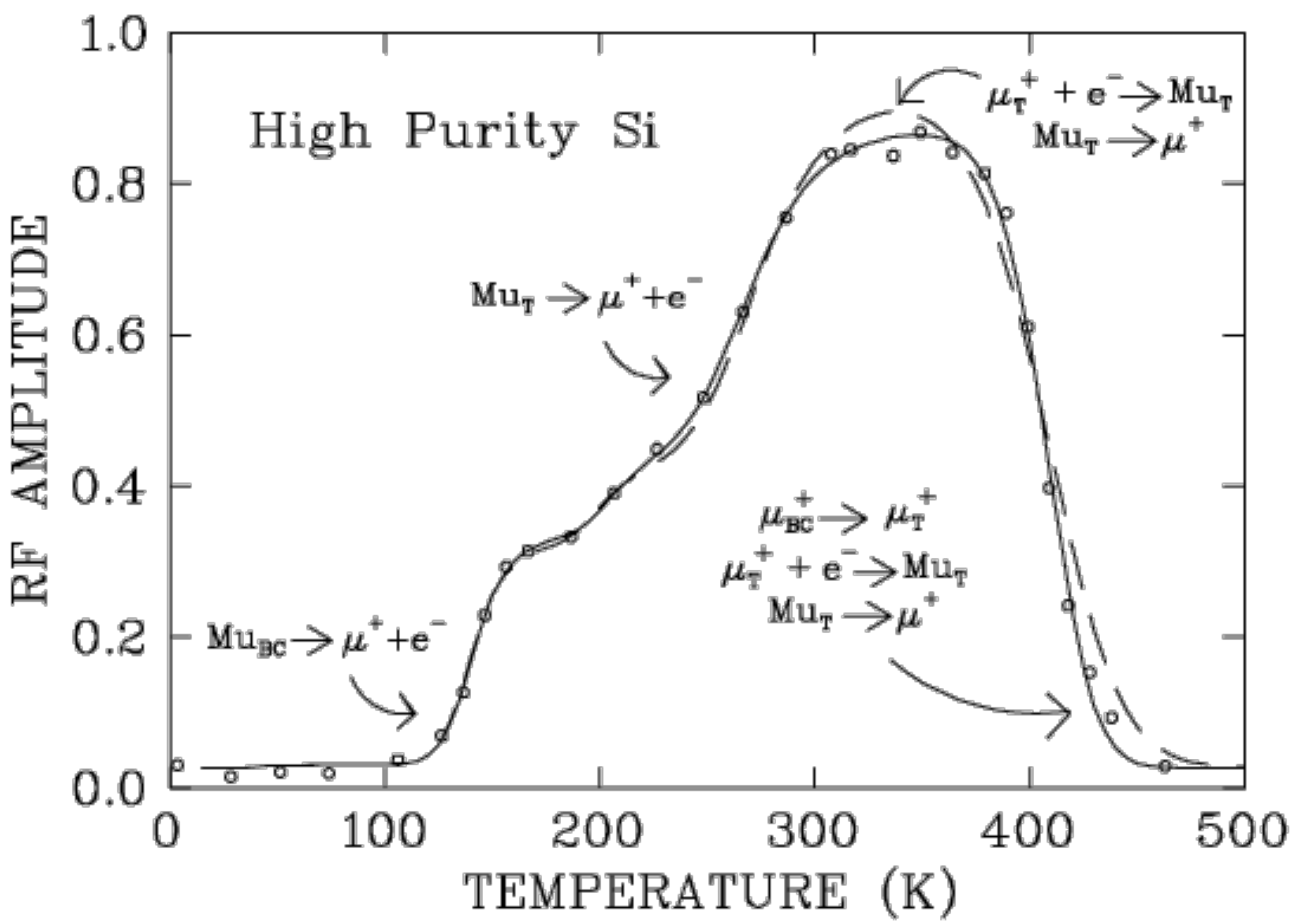
Paul W. Percival, Jean-Claude Brodovitch, Khashayar Ghandi, Brett M. McCollum, and Iain McKenzie

Apparatus has been developed to permit muon avoided level-crossing spectroscopy (μLCR) of organic free radicals in water at high temperatures and pressures. The combination of μLCR with transverse-field muon spin rotation (TF-μSR) provides the means to identify and characterize free radicals via their nuclear hyperfine constants. Muon spin spectroscopy is currently the only technique capable of studying transient free radicals under hydrothermal conditions in an unambiguous manner, free from interference from other reaction intermediates. We have utilized the technique to investigate hydrothermnal chemistry in two areas: dehydration of alcohols, and the enolization of acetone. Spectra have been recorded and hyperfine constants determined for the following free radicals in superheated water (typically 350°C at 250 bar): 2-propyl, 2-methyl-2-propyl (tert-butyl), and 2-hydroxy-2-propyl. The latter radical is the product of muonium addition to the enol form of acetone and is the subject of an earlier Research Highlight. The figure shows spectra for the **2-propyl** radical detected in an aqueous solution of 2-propanol at 350°C and 250 bar.

RF-μSR: muon Spin Resonance

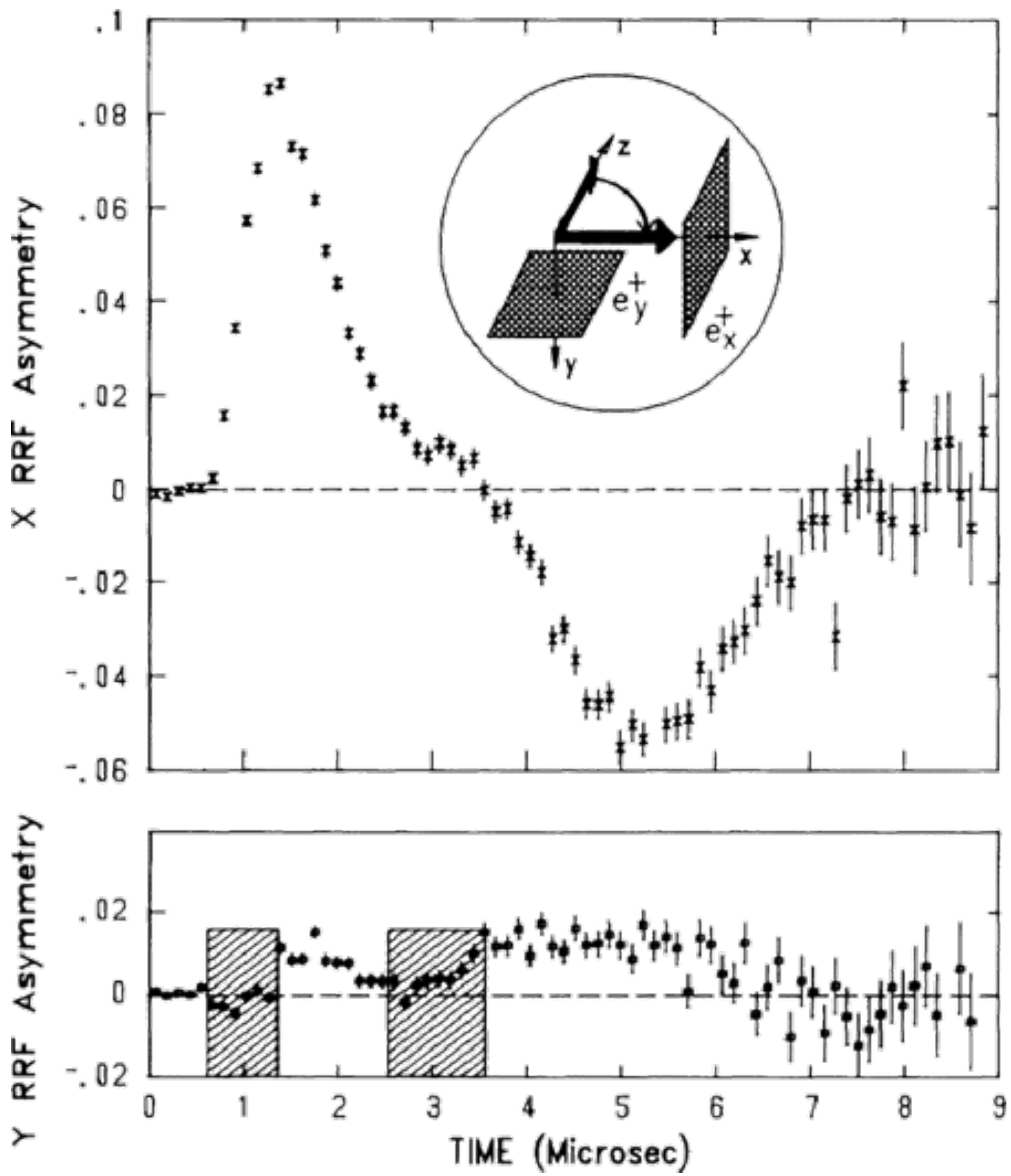
Resonance at ω_μ shows fraction of muons in ***diamagnetic*** states such as Mu^+ (= "bare" μ^+), Mu^- in various lattice sites even if it began as a paramagnetic state like Mu. Used to study formation and dissociation.





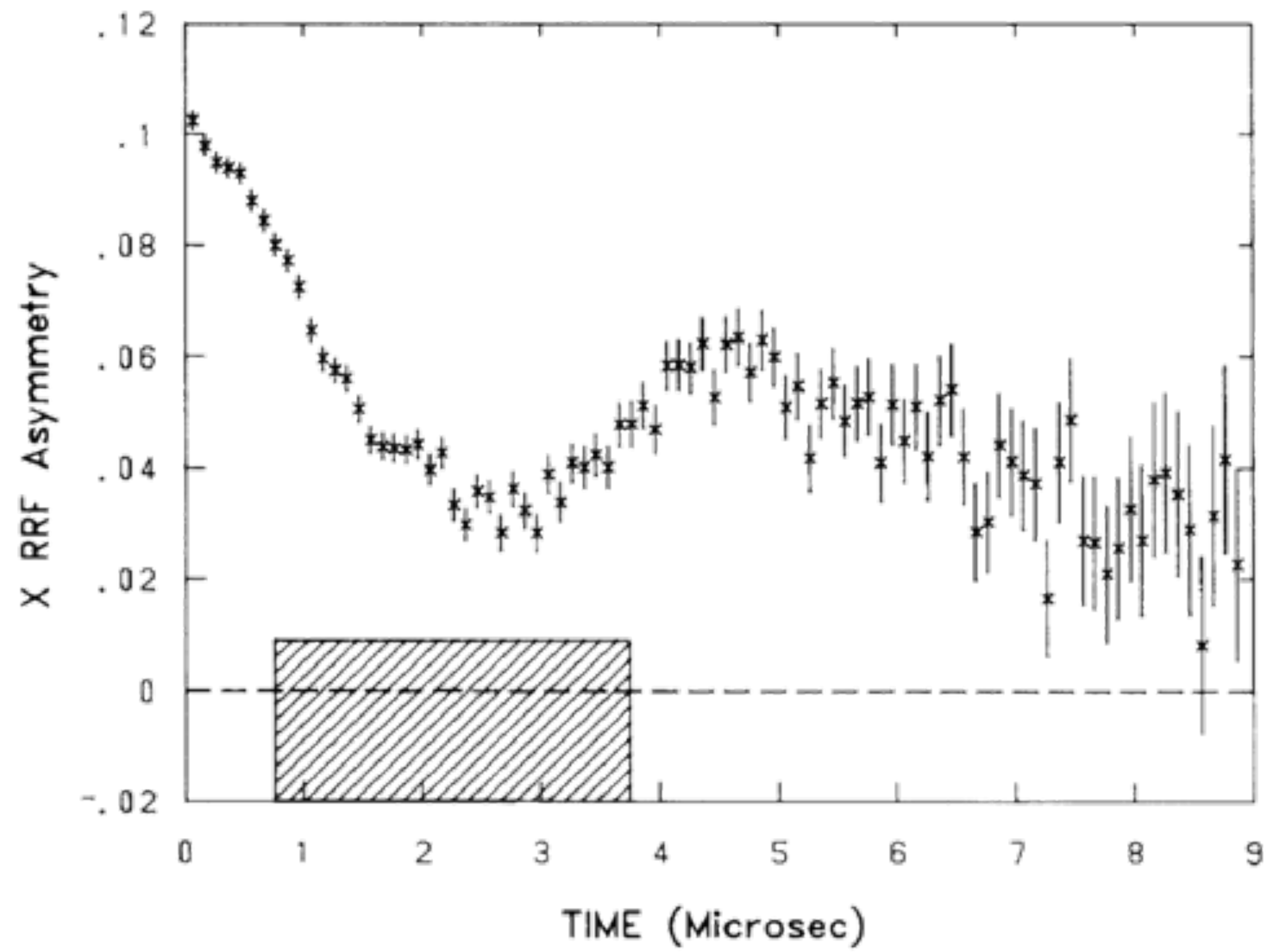
Muonium resonance at ω_{ij} shows fraction of muons in ***paramagnetic*** states such as Mu itself or a *radical* (paramagnetic molecule). In the above case the field-sweep shows broad ω_{12} and ω_{23} resonances as well as a *sharp* ***two-photon*** *resonance* at their average.

RF-*μSE*: muon *S*pin *E*cho



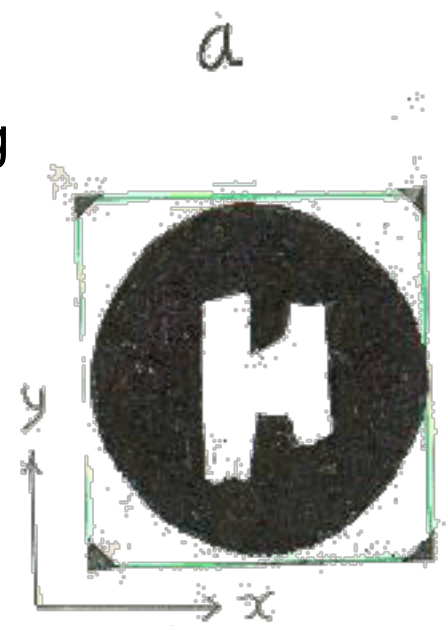
⬅ **Direct μSE** in LaF_3: muons enter with spins along $\boldsymbol{B}_0$. A $\Pi/2$ RF pulse at $\boldsymbol{\omega_\mu}$ "flips them up" and they precess and "dephase"; a Π pulse at time τ makes them "refocus".

Indirect μSE: muon spins initially $\perp$ $\boldsymbol{B}_0$ are refocused by a Π pulse *on the* ^{19}F *nuclei* at frequency $\boldsymbol{\omega_F}$. ⬇

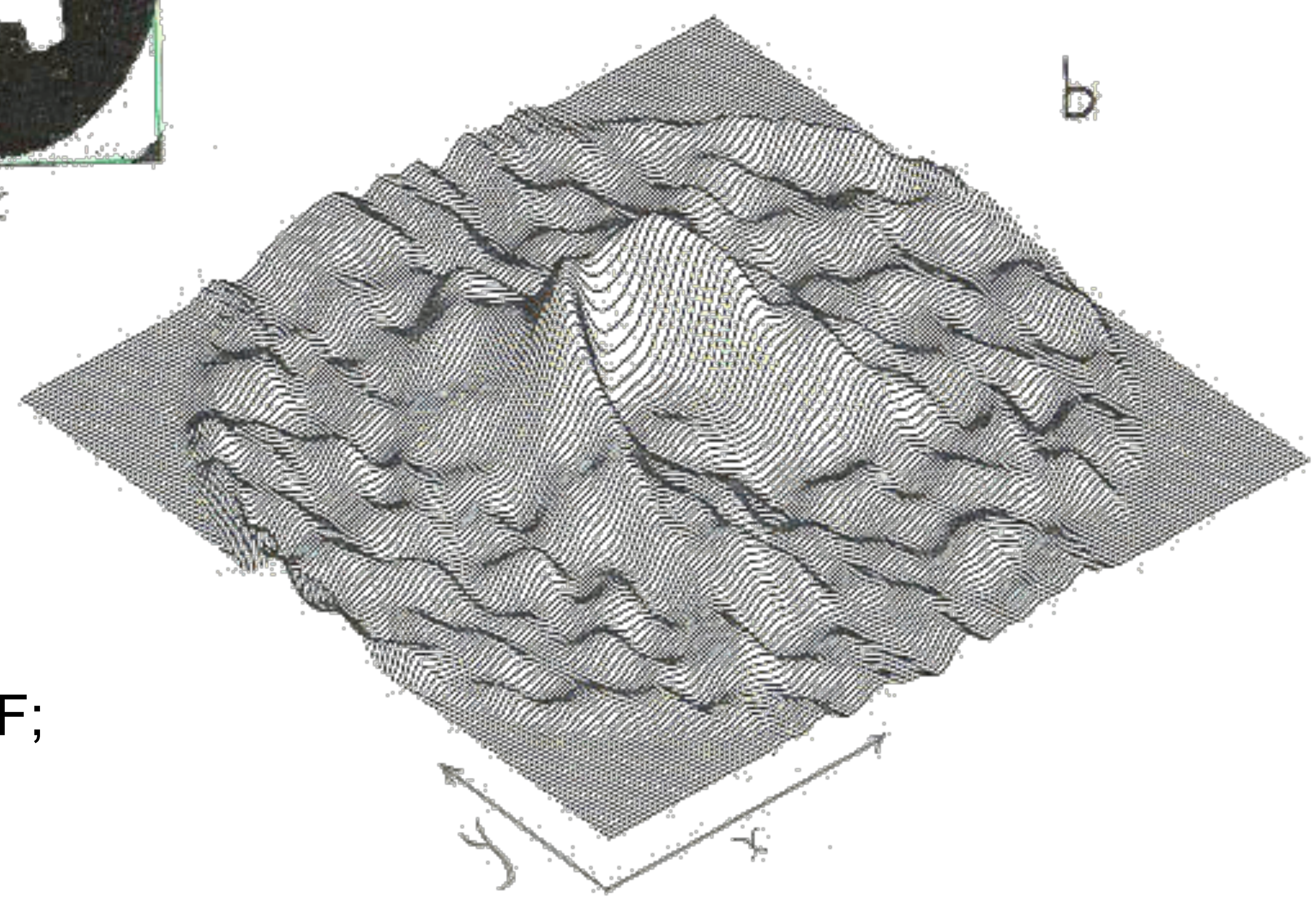


μSE : Muon Spin Imaging

"**μ**" cut from Al on depolarizing substrate:



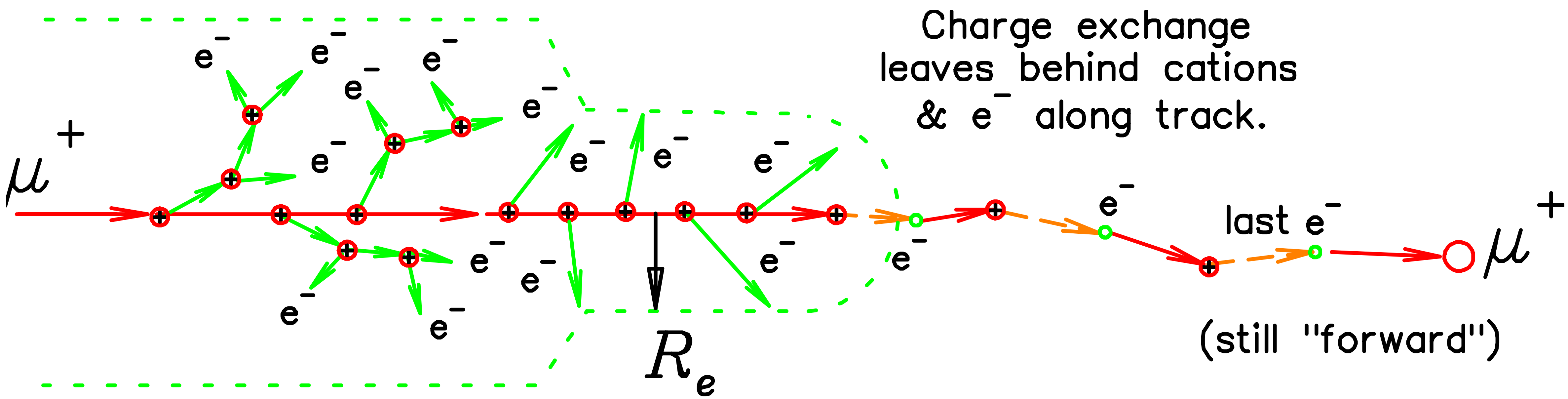
Magnetic field gradients applied in many directions; each spectrum Fourier transformed to translate frequency into spatial coordinates; all combined to form image:



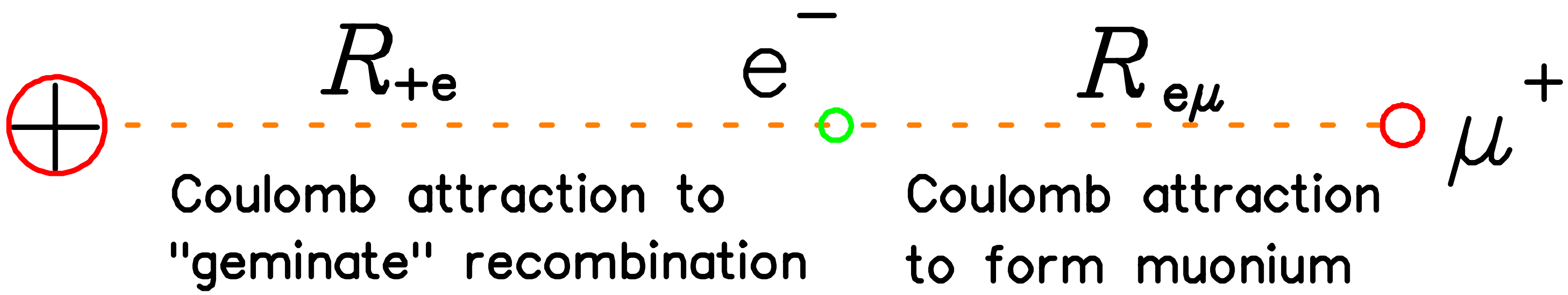
Deemed impractical at TRIUMF; but with RF & gradient pulse sequences at a high intensity pulsed facility, who knows...?

N. Kaplan *et al*., Hyperfine Int. **85**, 271 (1994)

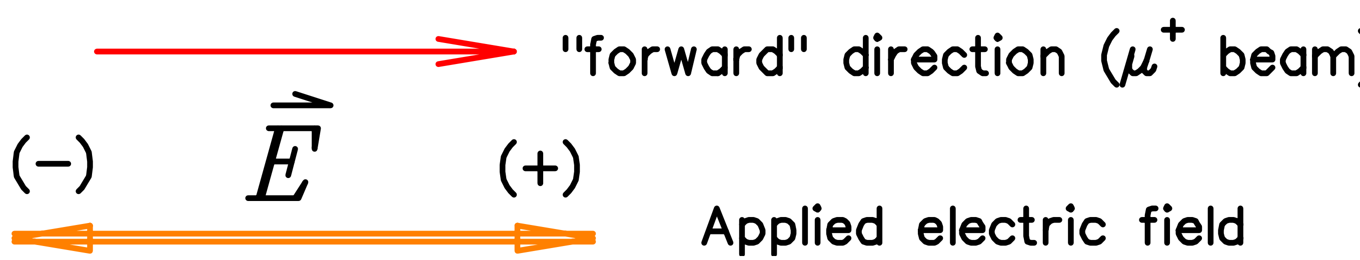
Radiolysis & "Delayed" Muonium Formation



RESULT (sometimes):

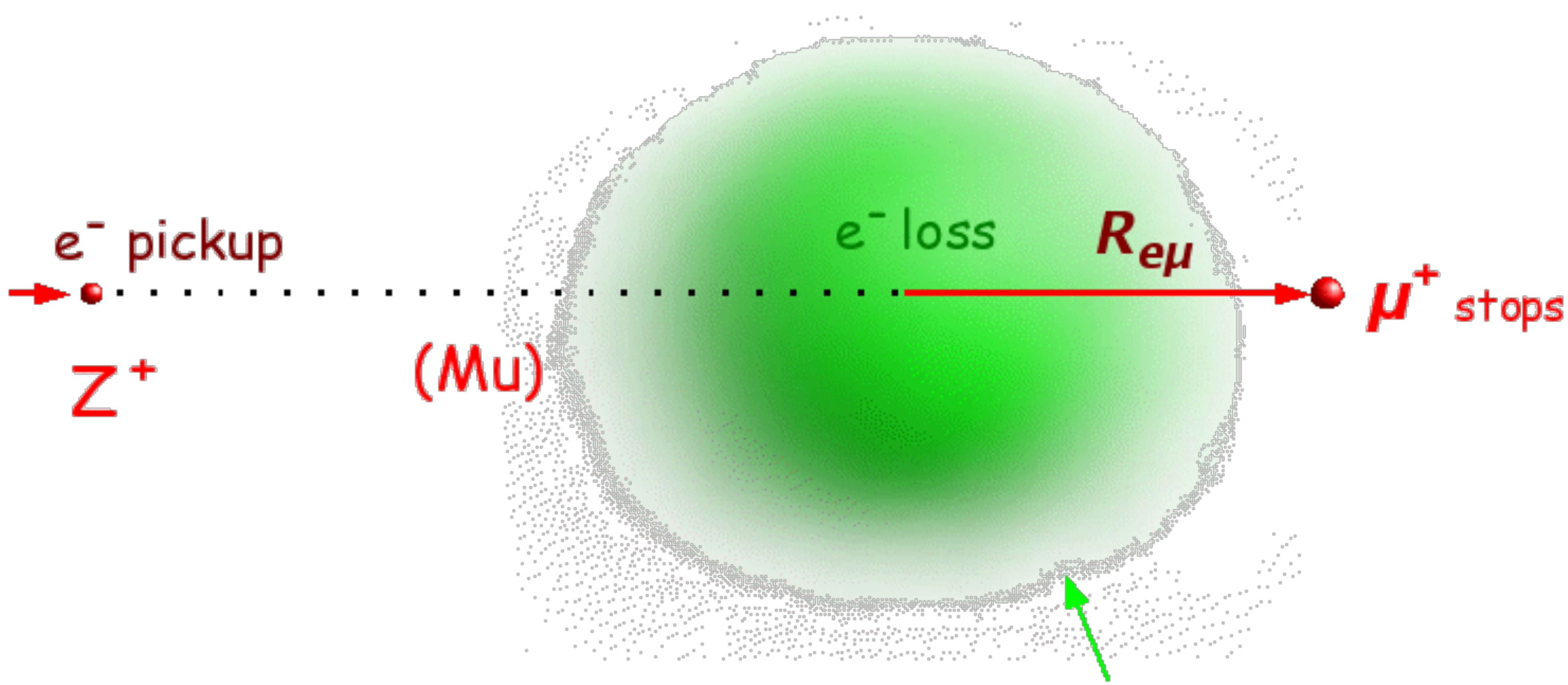


Possible **exceptions:**
solids with extremely high electron mobilities.



A closer look at the **final charge exchange:**

(Actual path is probably less straight, but still "forward".)



Distribution of "initial" e^- positions
may <u>overlap</u> position where μ^+ stops

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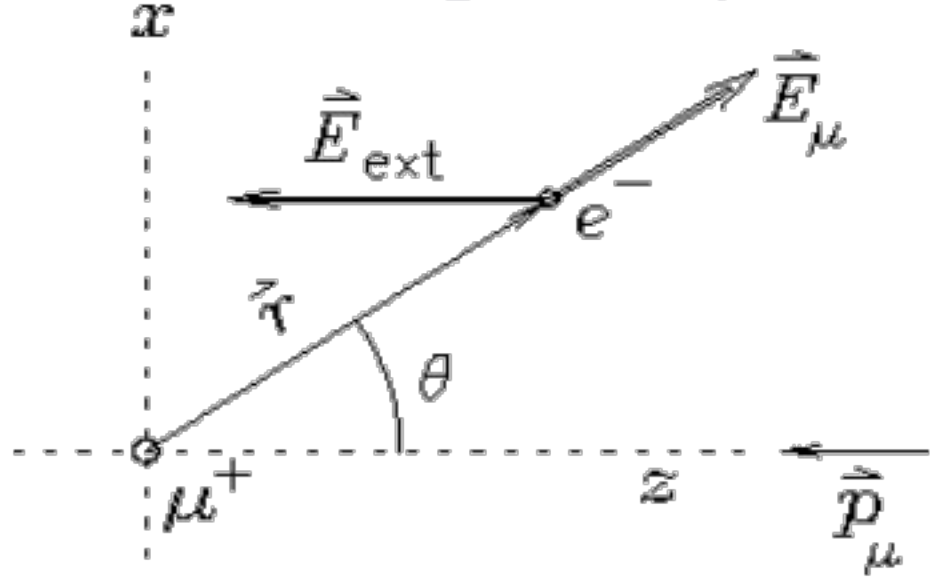
Muonium Formation in an Electric Field

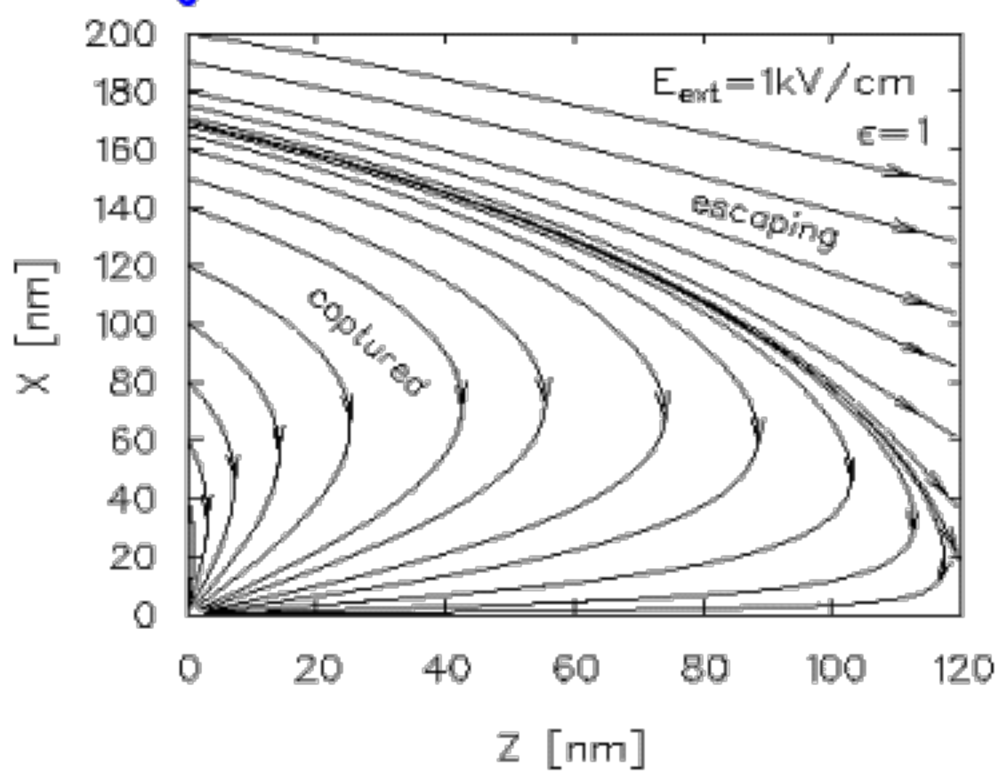
μ-e-**E** geometry:



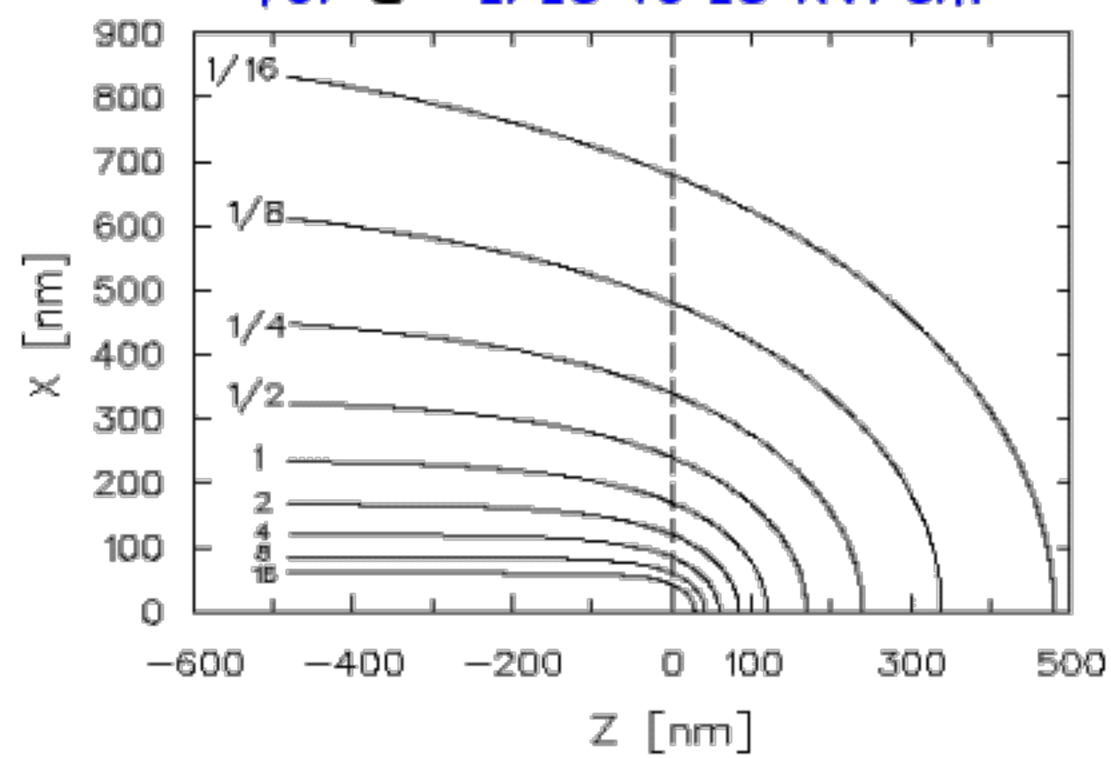
Viscous flow model:

e^- constantly loses momentum to the medium, and so follows "lines of **E**" at constant speed.

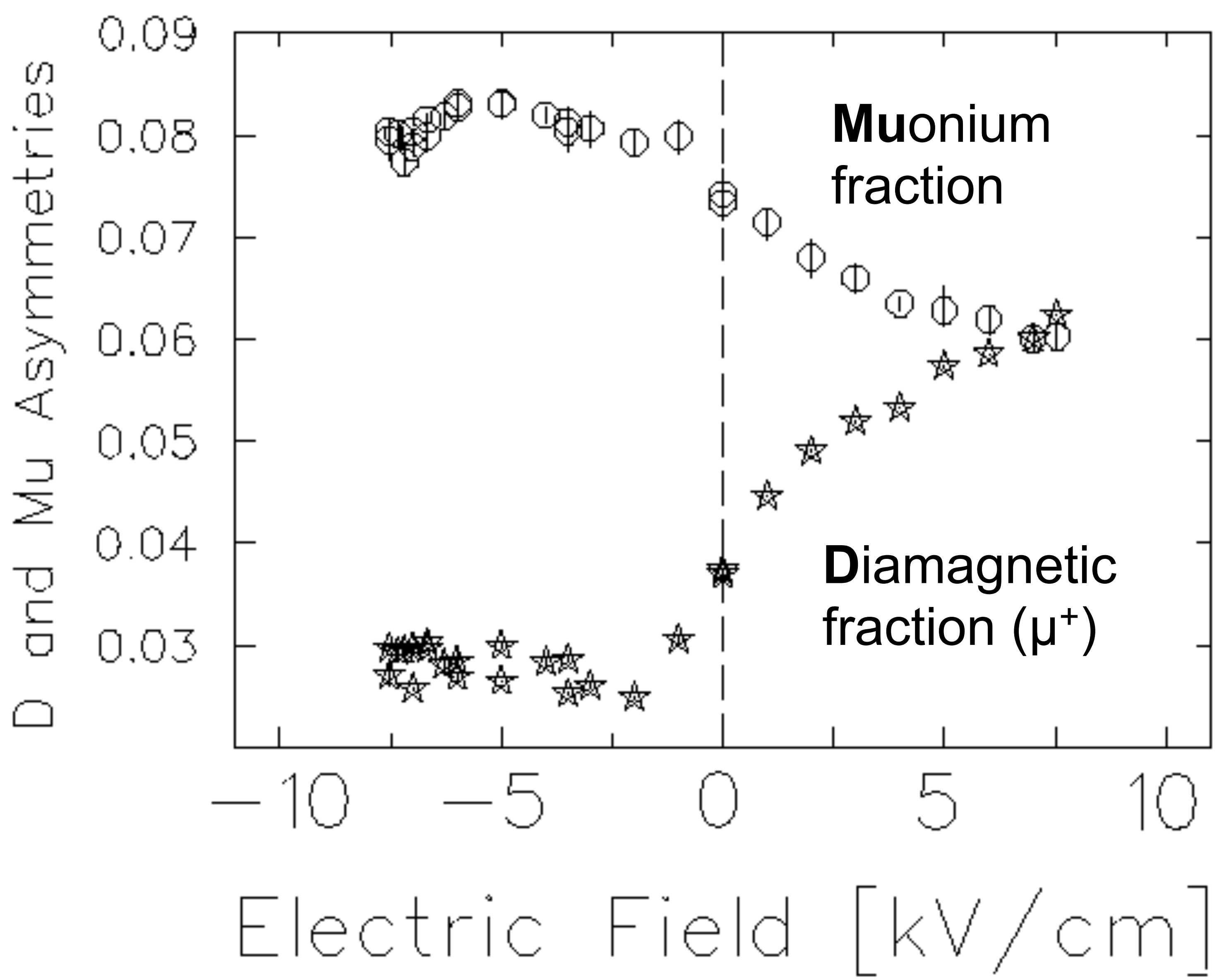
Trajectories for **E** = 1 kV/cm:



e^- capture boundaries for **E** = 1/16 to 16 kV/cm:

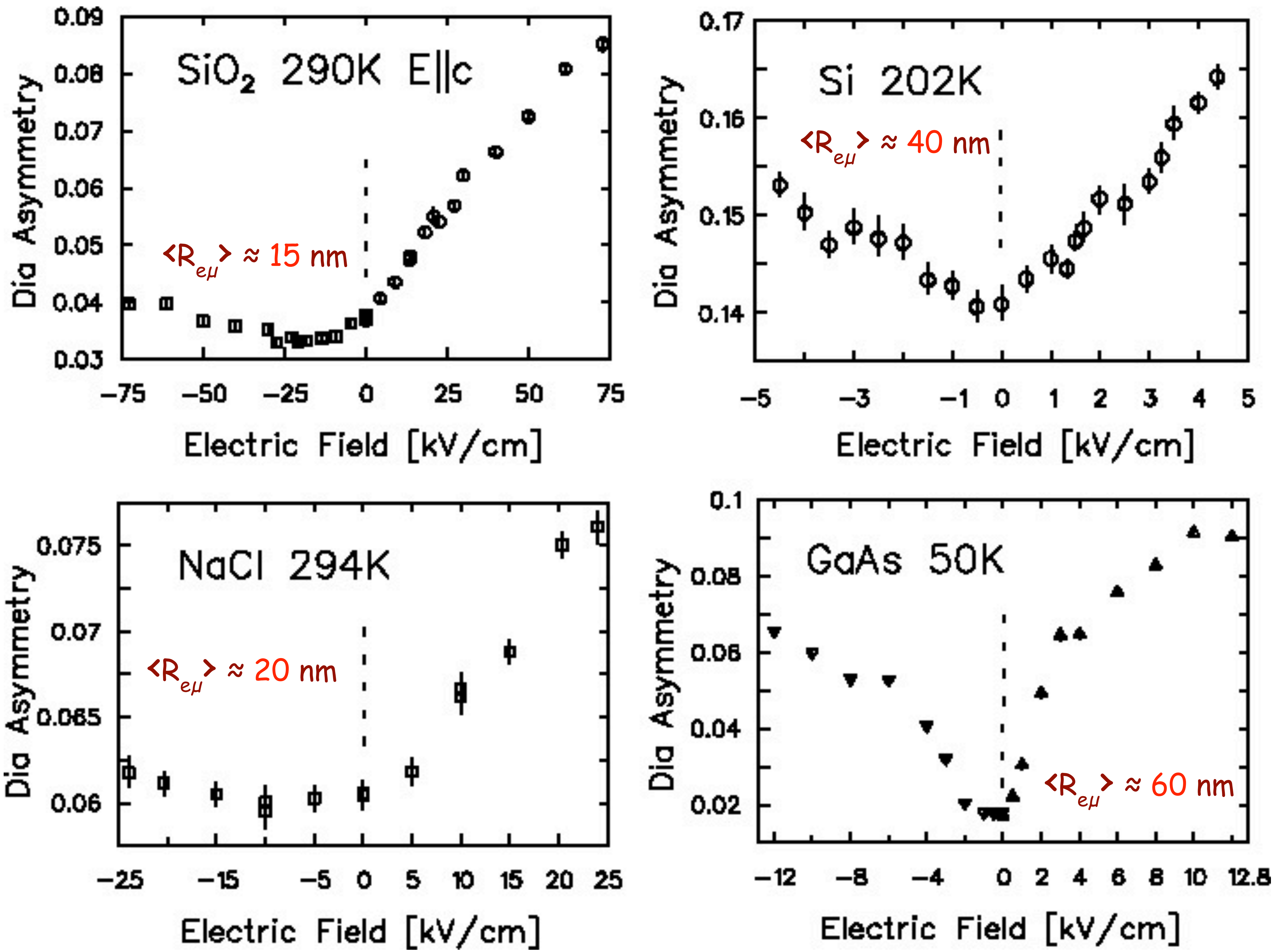


Delayed Mu Formation in Cryocrystals (*e.g.* s-N_2)

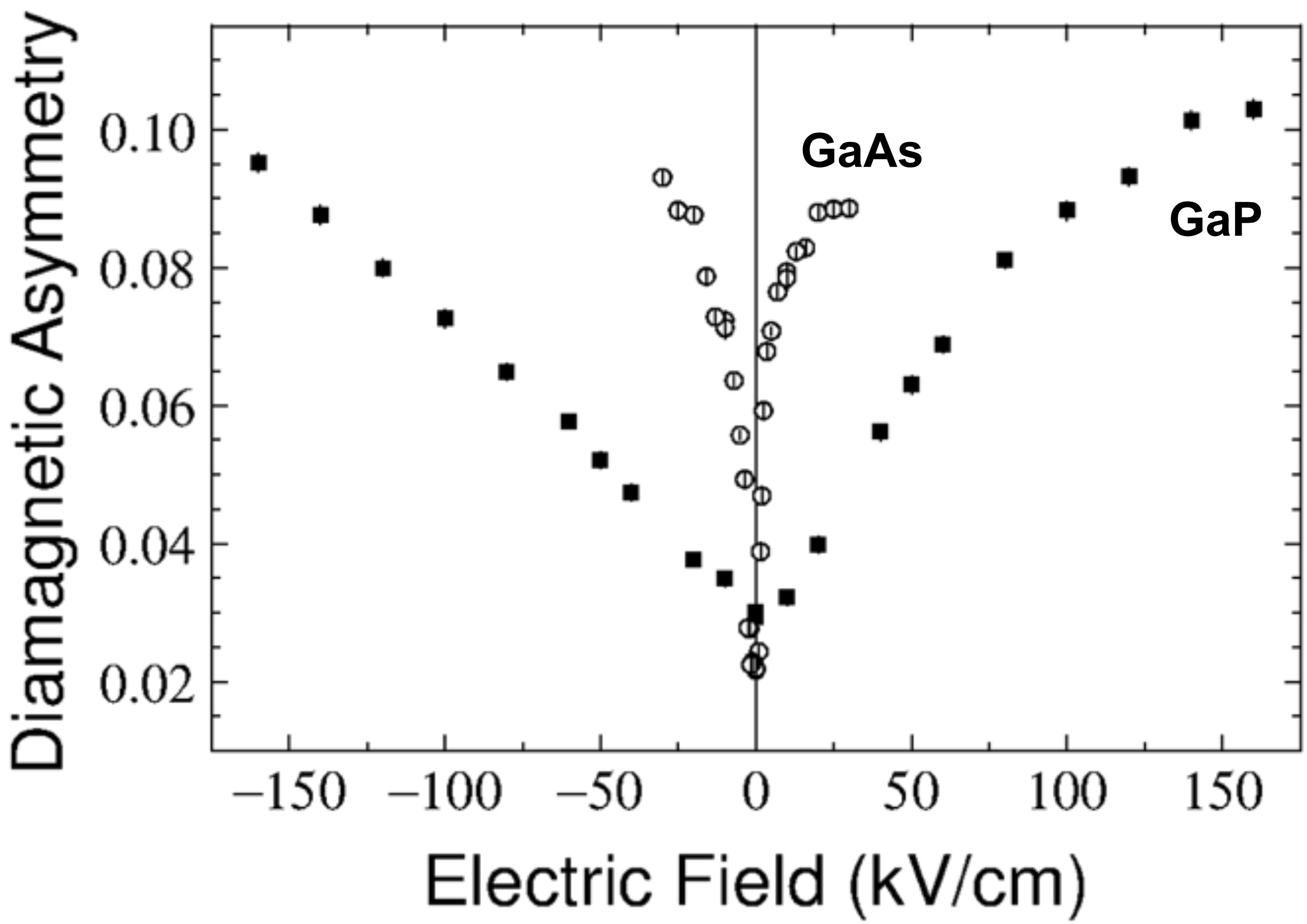


"Ordinary" Solids: Insulators & Semiconductors

Note different horizontal & vertical **scales**!



Weakly Bound Mu States in High-Mobility Semiconductors



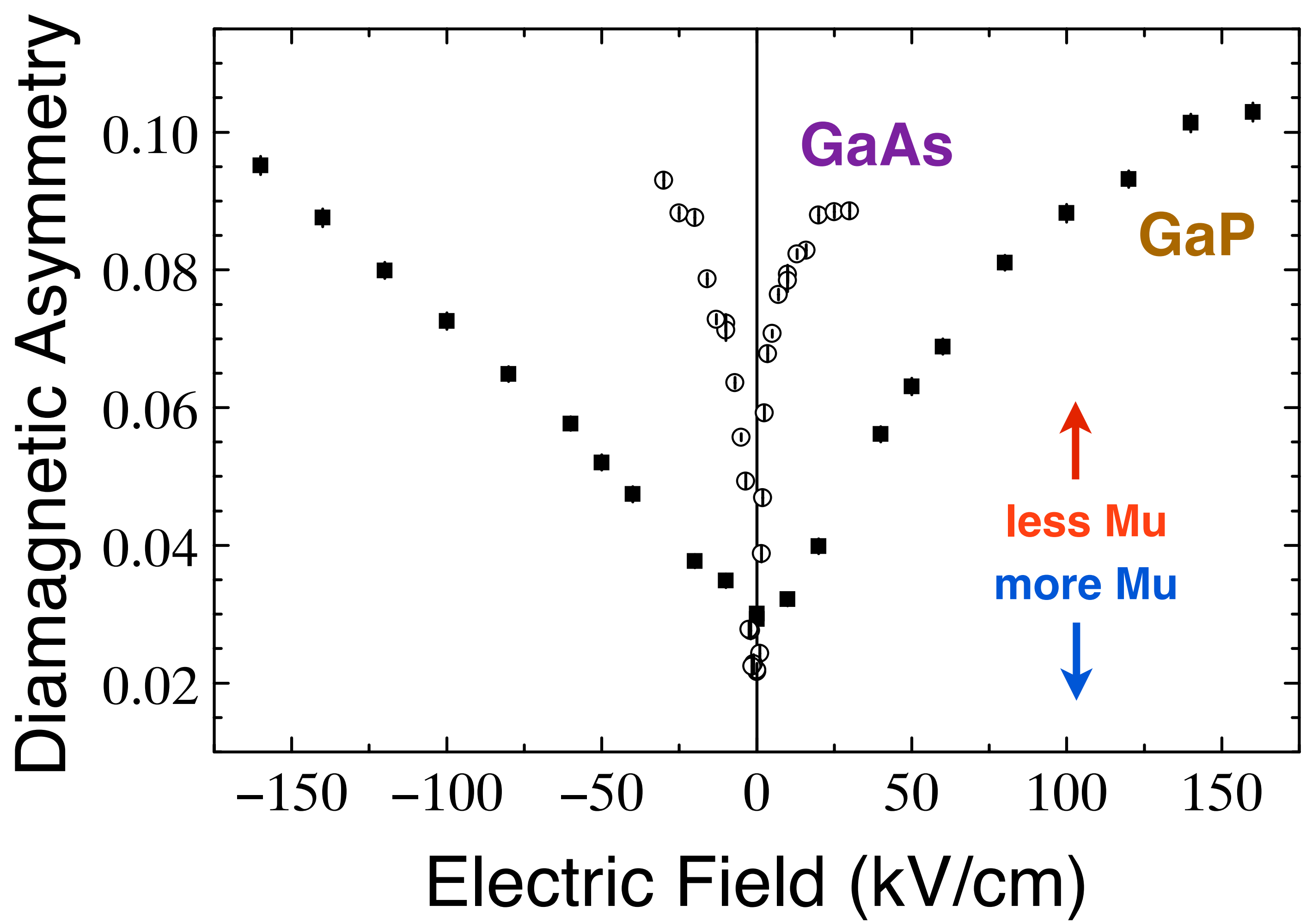
Initial states Mu_{wb} have electron orbitals "out in the lattice" with different effective masses; those with higher *m** are more strongly bound and harder to ionize with an applied ***E*** field.

V.G. Storchak et al., Phys. Rev. B **67**, 121201 (2003).

Weakly Bound Muonium States in GaAs & GaP

V.G. Storchak, D.G. Eshchenko, R.L. Lichti and J.H. Brewer

Lighter effective mass & higher mobility e^- ⇒ easier to prevent Mu formation by applied ***E***.



Muonium formation *via* electron transport to a positive muon implanted into semi-insulating GaP has been studied using muon spin rotation/relaxation with alternating electric fields up to 160 kV/cm. Formation of the muonium ground state is prohibited by a characteristic electric field of about 50 kV/cm in GaP compared to 5 kV/cm in GaAs, implying that formation of the Mu ground state may proceed through a weakly-bound intermediate state with a binding energy of about 23 meV in GaP or 7 meV in GaAs. These results are discussed and justified within the effective mass model.

See *μSR* Literature Entry # 2437

Phys. Rev. B **67**, 121201 (2003).

Finis