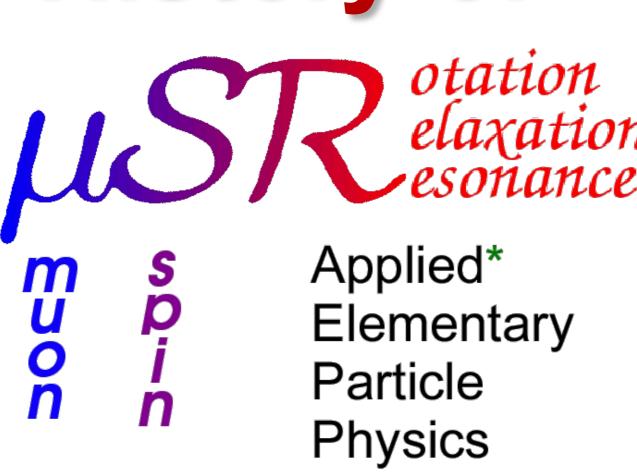




History of



A science fiction adventure story

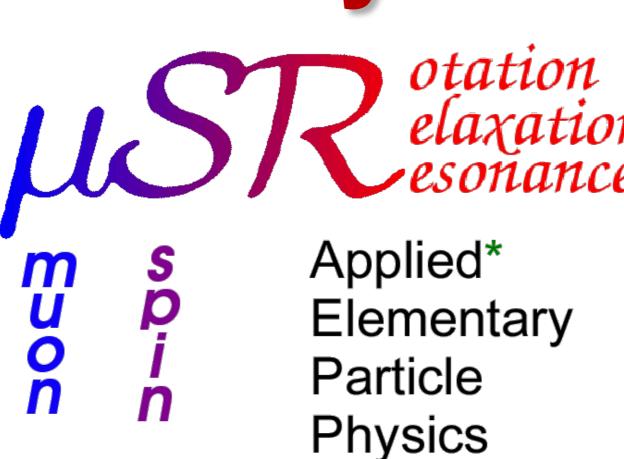
by

Jess H. Brewer





History of



*(to basic research in Materials Science and Chemistry)





OUTLINE

- Early History of μSR ("science fiction"?)
- Development of Advanced Muon Beams
- Research "Themes" in μ +SR
- µSR applications (interleaved among techniques...)









Fantasy: violates the "known laws of 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.")





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- Routine Physics: "We can do that . . ."
- Applied Science: "... and so can you!"





Before 1956: $\mu SR = Fantasy$

(violates "known laws of physics")

1930s: Mistaken Identity

Yukawa's "nuclear glue" mesons ≠ cosmic rays

1937 Rabi: Nuclear Magnetic Resonance

1940s: "Who Ordered That?"

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

1947 Richardson: produced $\pi \& \mu$ at Berkeley 184 in. Cyclotron

1949 Kuhn: "The Structure of Scientific Revolutions"





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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.





1958-1973: Science Fiction

9 1960s: Fundamental Physics Fun! — Tours de Force

Michel Parameters = Weak Interaction Laboratory

Heroic QED tests: $A_{HF}(Mu)$, μ_{μ} , g_{μ} – 2

All lead to refined μ SR techniques.

Applications: Muonium Chemistry, Semiconductors, Magnetism

mid-1970s: Meson Factories — Intensity Enables!

USA: **LAMPF** (now defunct) Switzerland: **SIN** (now **PSI**)

Canada: TRIUMF UK: RAL/ISIS

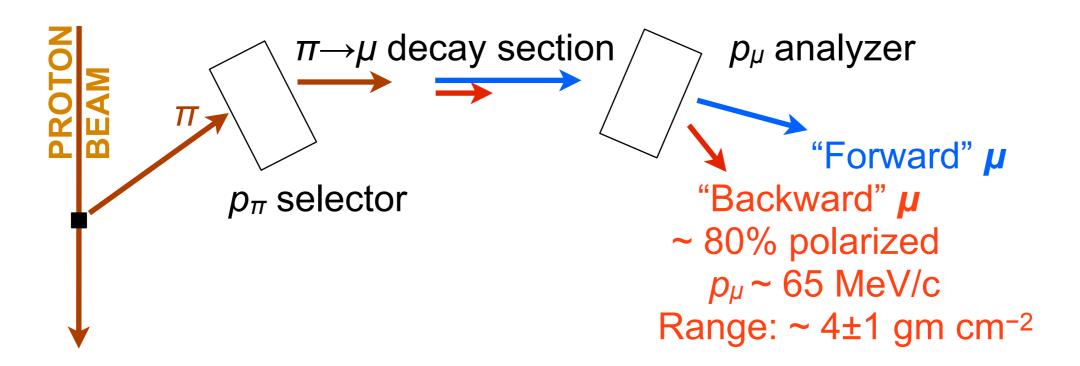
Japan: **KEK/BOOM** (→ **J-PARC**)





MUON BEAMS

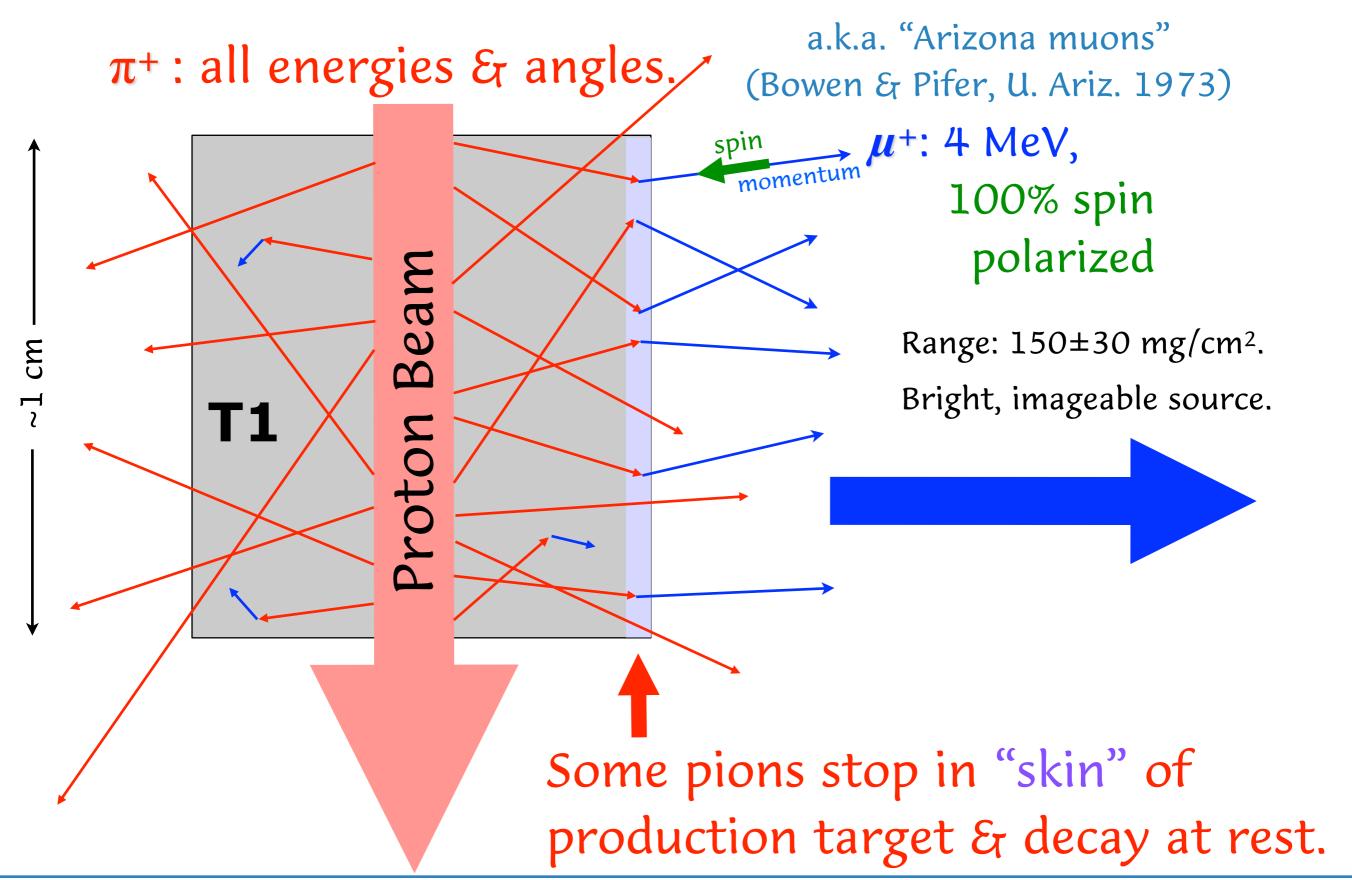
DECAY MUON CHANNEL (μ^+ or μ^-)





Surface Muons









µ+ Stopping Luminosity



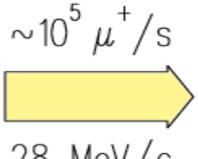
$$\sim$$
 10³ μ ⁺/s
 \sim 100 MeV/c

from low-current accel.

 $\sim 10^{5} \, \mu^{+}/\mathrm{s}$

BWD μ^{\pm} beams ~ 50 MeV/c from Meson Factories

Surface Muon beams from Meson Factories



28 MeV/c



 $\sim 1 \, \mathrm{g}$

~ 100 g



1965

1975 SIN

1985 TRIUMF

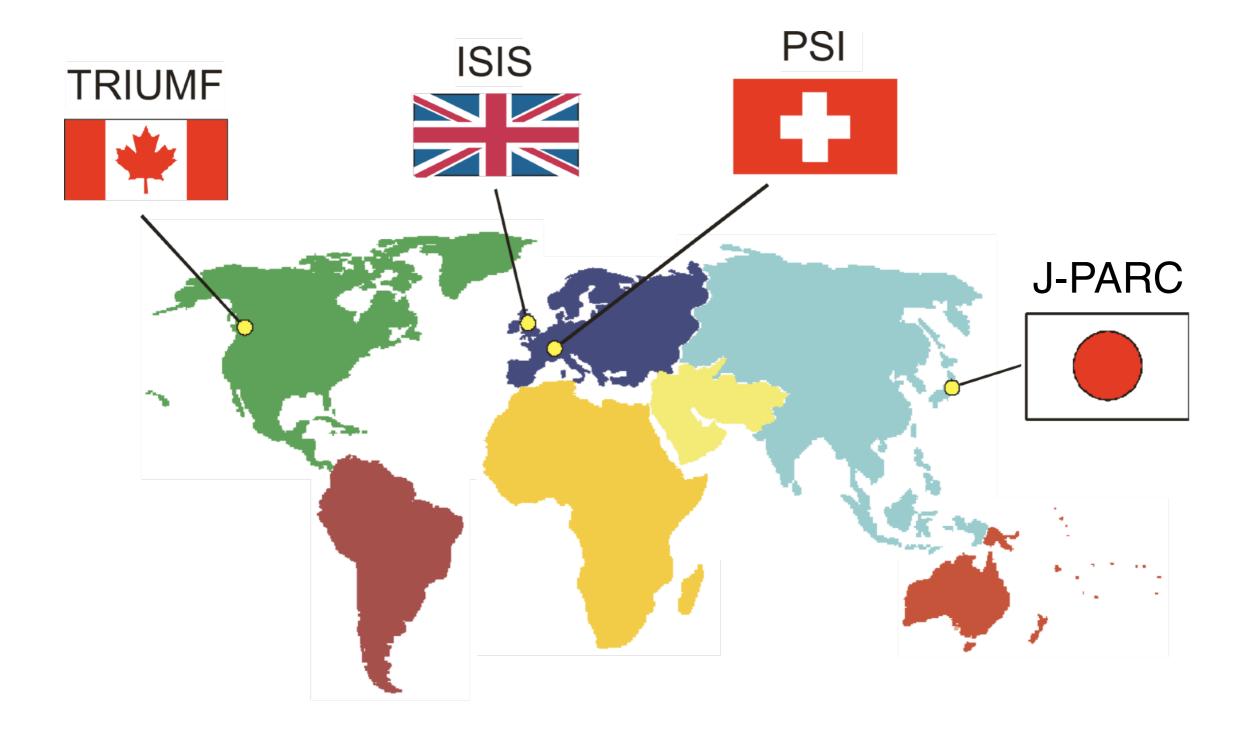
2005

Moderated surface
$$\mu^+$$
 $\sim 10^3 \, \mu^+/\mathrm{s}$ ~ 10 eV + re-accel. \longrightarrow for depth profiling





µSR today: Routine Science?







μSR today: Routine Science?

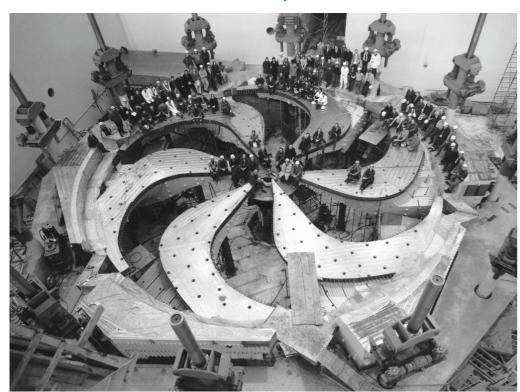




TRIUMF, 1972

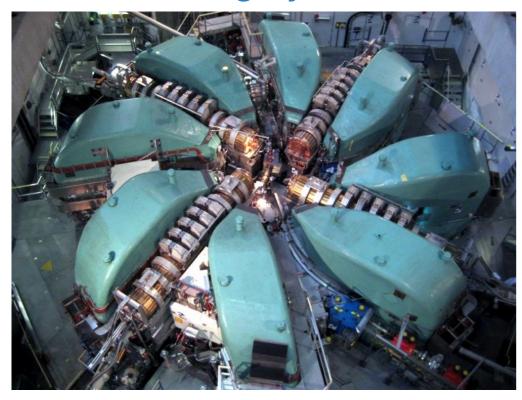








PSI Ring Cyclotron



J-PARC synchrotron





For newcomers...

How does it work?

... a brief introduction to



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

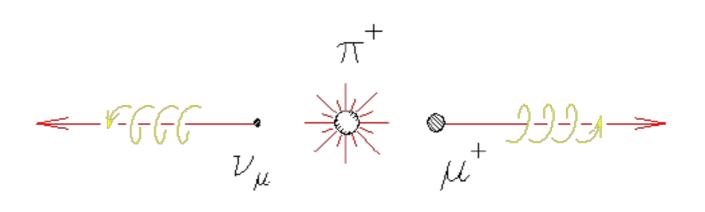
A spinless *pion* **stops** in the "skin" of the primary production target. It has zero linear momentum and zero angular momentum.

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
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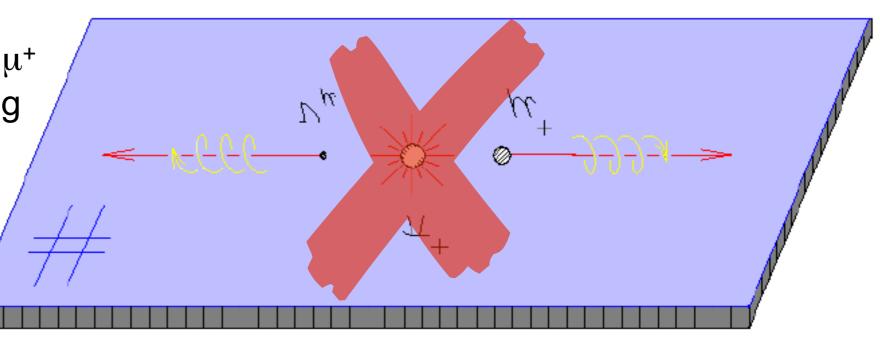
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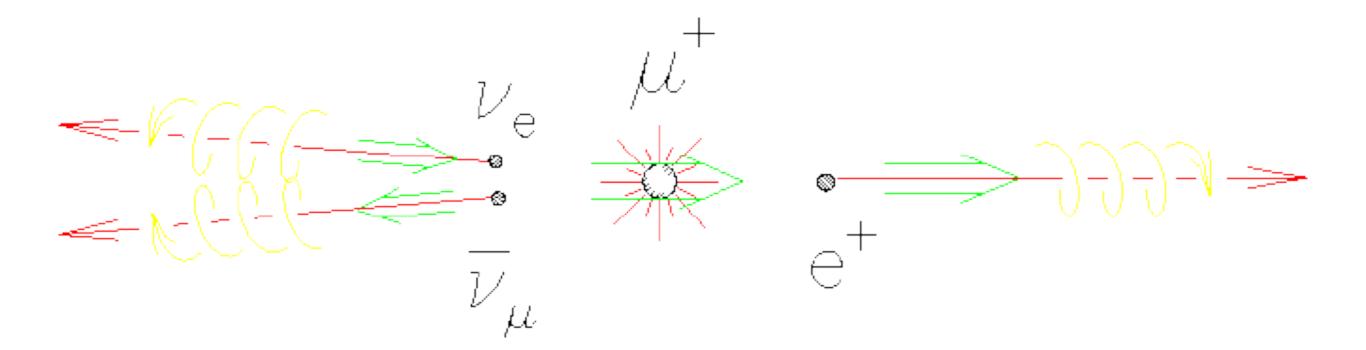
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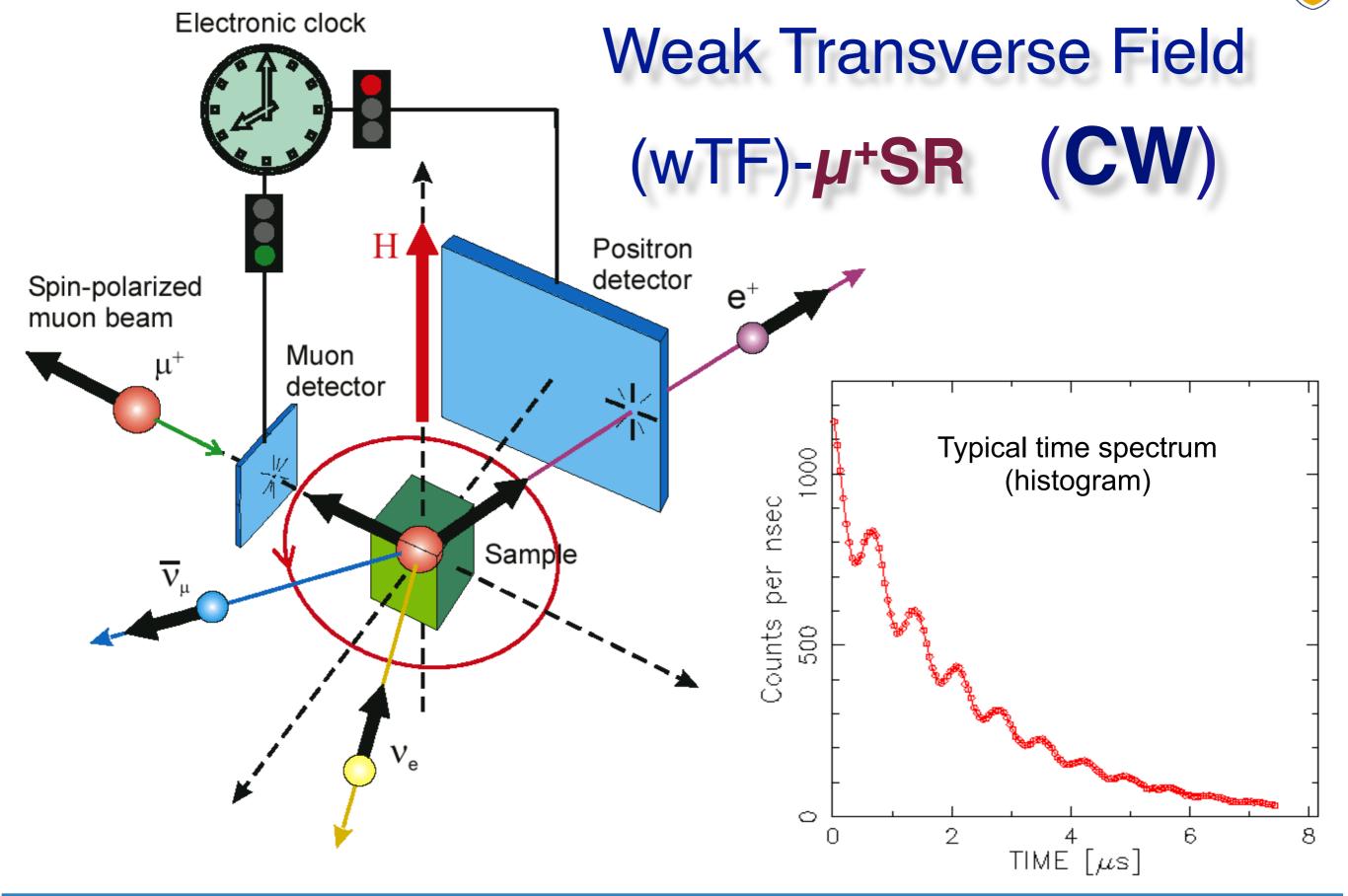
µ+ Decay



Neutrinos have negative helicity, antineutrinos positive. An ultrarelativistic positron behaves like an antineutrino. Thus the positron tends to be emitted along the μ^+ spin when $\nu_{\rm e}$ and $\overline{\nu}_{\rm \mu}$ go off together (highest energy e^+).









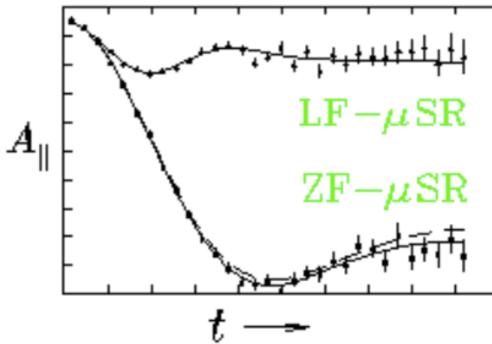


Zero or Longitudinal Field (ZF/LF)-\mu+SR



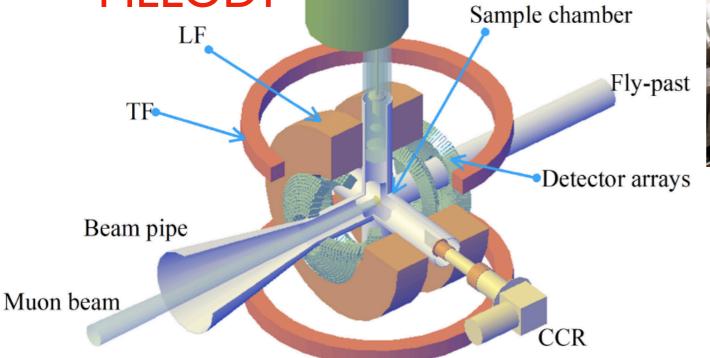


(pulsed)



$$A_{II} = (N_B - N_F)/(N_B + N_F)$$

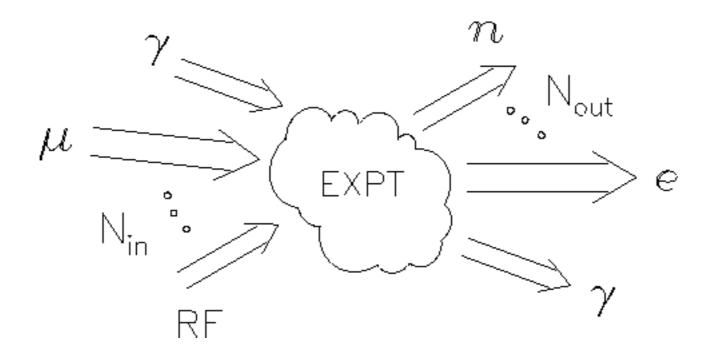
All the muons arrive in one pulse, requiring segmented detectors to avoid "pile-up" distortions.







CW vs. Pulsed µSR



"Advantage factor" for *pulsed* muons:

$$A_{\rm p} = log(N_{\rm in}/N_{\rm out})$$

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

*Dis*advantage of *CW* muons: rate (< 10⁴ s⁻¹ vs. "unlimited")

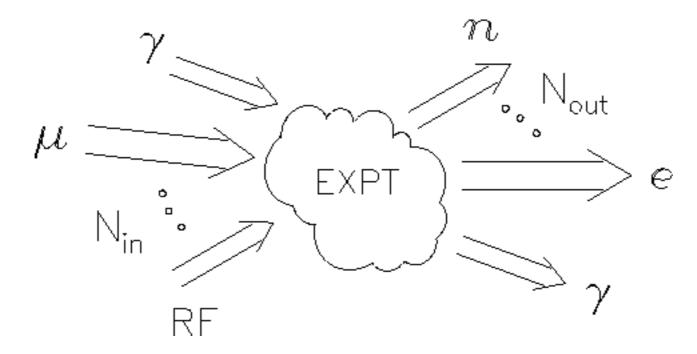




TRIUMF

CW vs. Pulsed µSR

PSI



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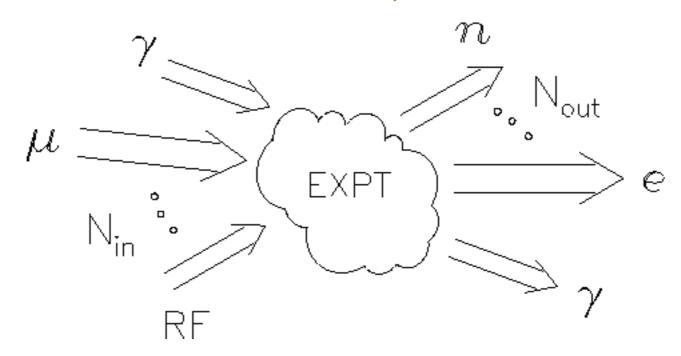




TRIUMF ISIS

CW vs. Pulsed µSR

PSI J-PARC, MELODY



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Research "Themes" in µ+SR

Muonium as light Hydrogen

 $(Mu = \mu^+e^-)$ $(H = p^+e^-)$

- 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, $\mu^+SR \rightarrow only$ 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 μ^-SR ...





And then there's μ^-SR ...

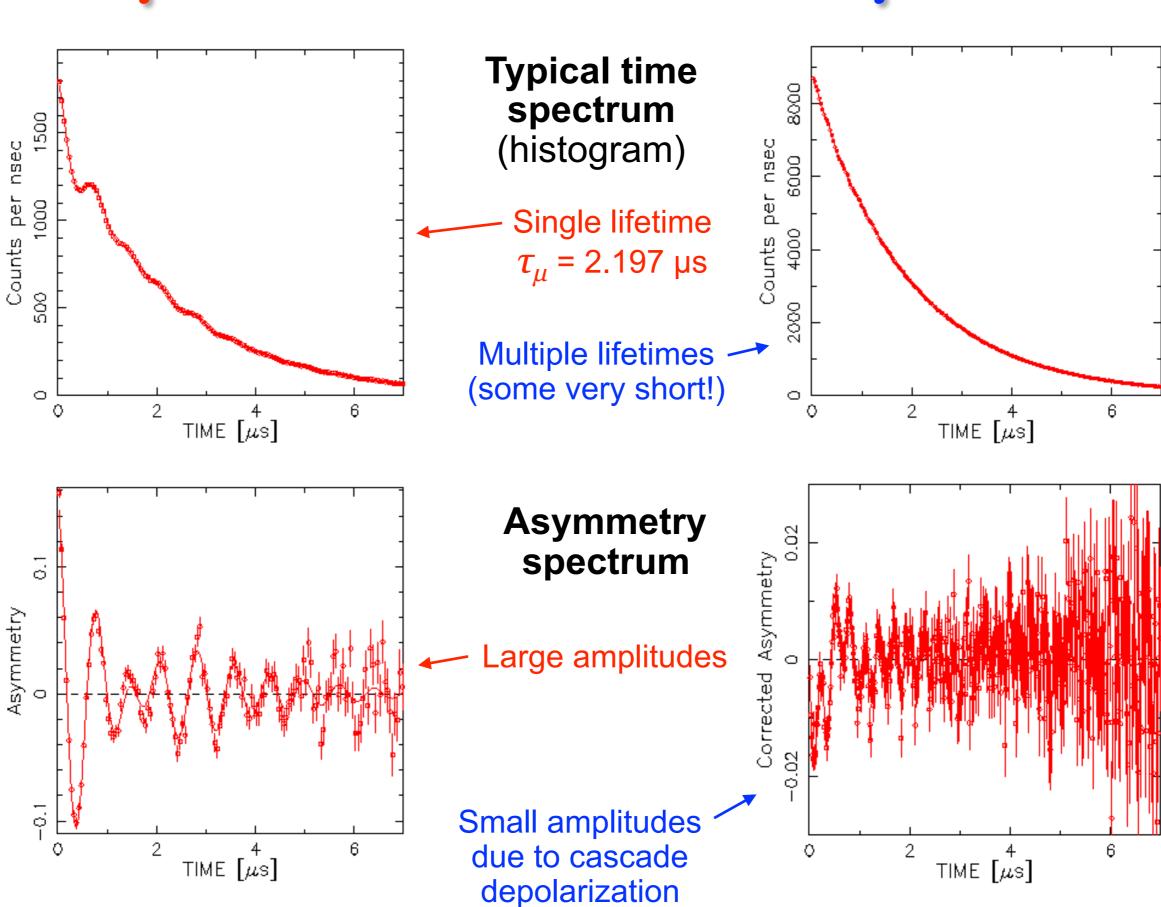
... but there's not enough time for all its methods & applications.



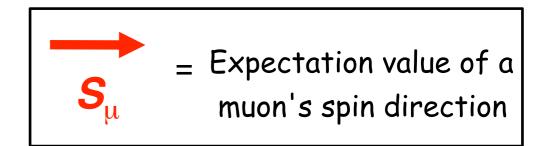


US.





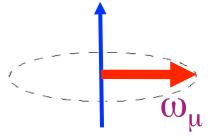
Motion of Muon Spins in Static Local Fields



(a) All muons "see" same field B: ————————————————————————for $B \parallel S_{\mu}$ nothing happens

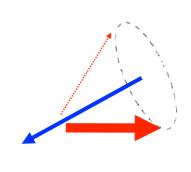
$$\omega_{\mu} = 2\pi \gamma_{\mu} |\mathbf{B}|$$
 $\gamma_{\mu} = 135.5 \text{ MHz/T}$

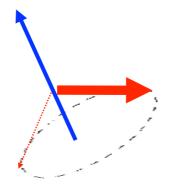
for $B \perp S_{\mu}$ Larmor precession:

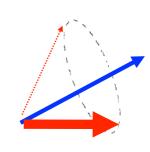


(b) All muons "see" same |B| but random direction:









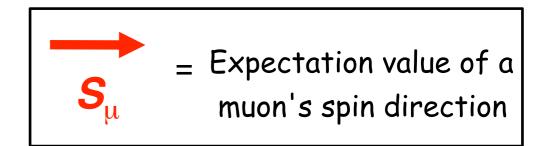
2/3 of $\boldsymbol{S}_{\!\mu}$ precesses at $\boldsymbol{\omega}_{\!\mu}$

1/3 of S_{μ} 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) $\Longrightarrow S_{\rm u}$ "relaxes" as $G_{zz}(t)$ [Kubo & Toyabe, 1960's]

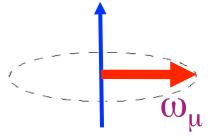
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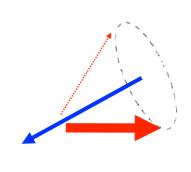
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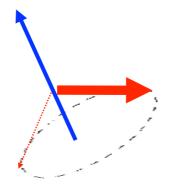
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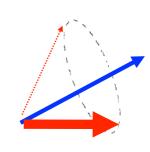


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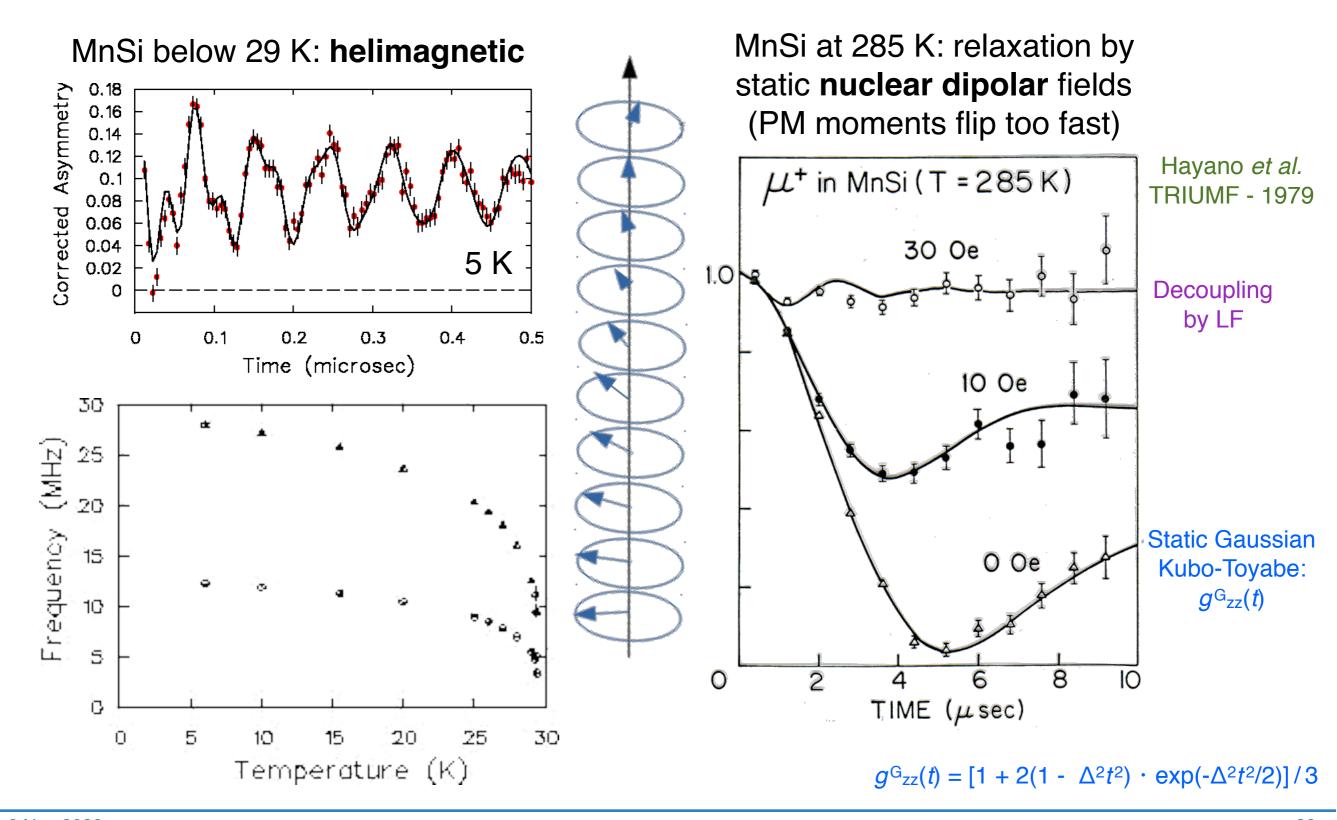
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ZF/LF-µSR & Local Magnetic Fields





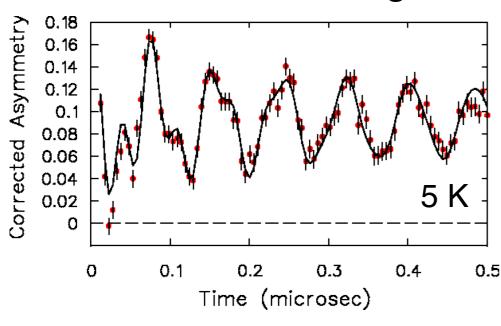


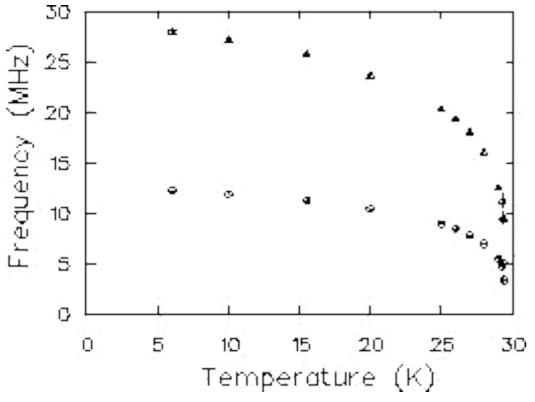
ZF/LF-µSR & Local Magnetic Fields

get **very interesting**

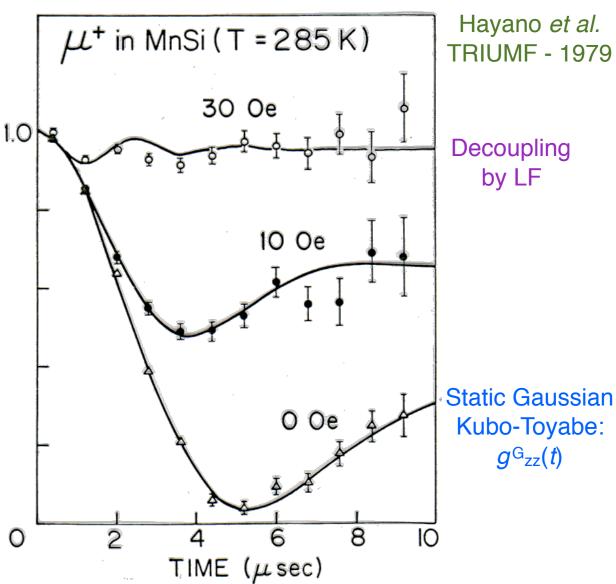
In between, things

MnSi below 29 K: helimagnetic





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



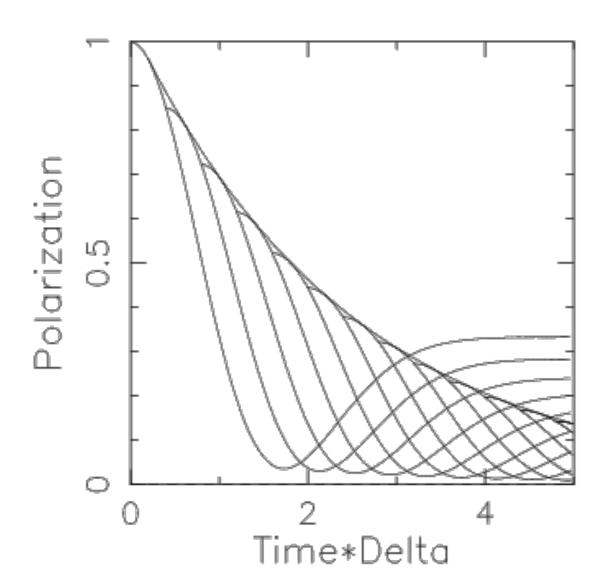
 $g^{G}_{zz}(t) = [1 + 2(1 - \Delta^{2}t^{2}) \cdot \exp(-\Delta^{2}t^{2}/2)]/3$





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:

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

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

Used to extract "hop" or fluctuation rate v.





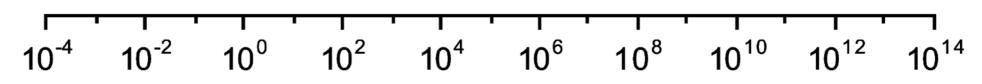
Time Scales



NMR

μSR

Neutron Scattering

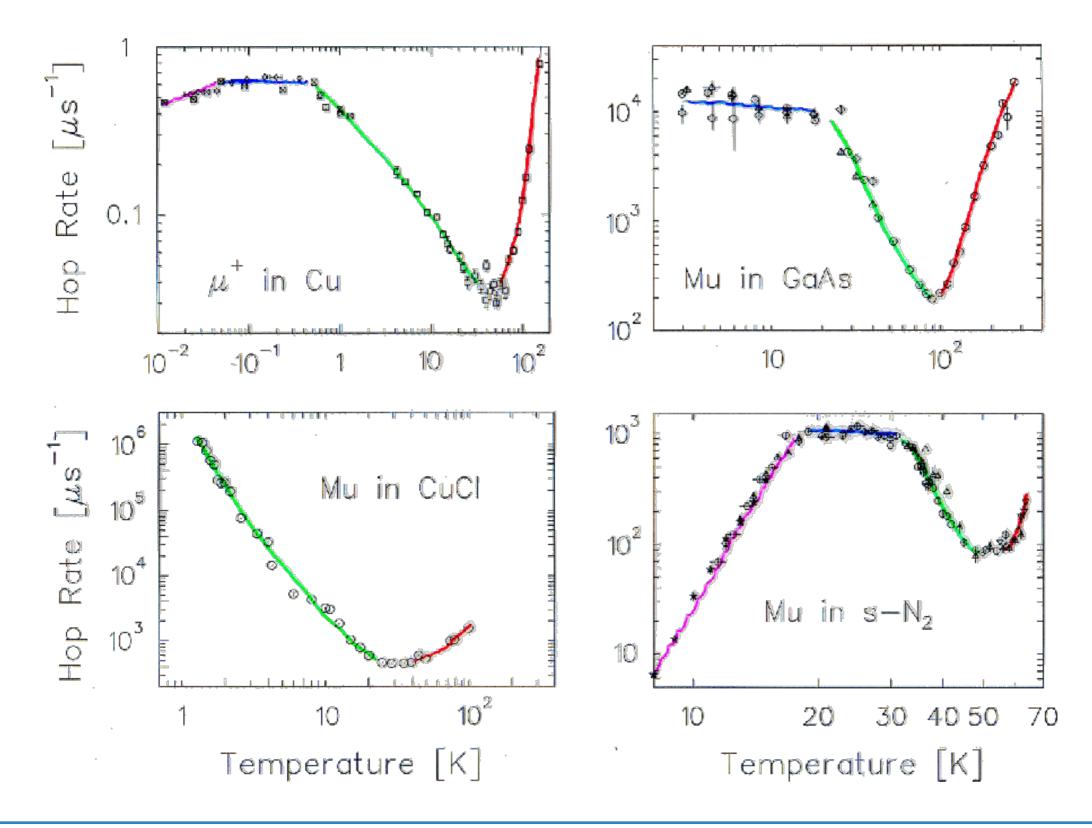


Fluctuation Rate (Hz)





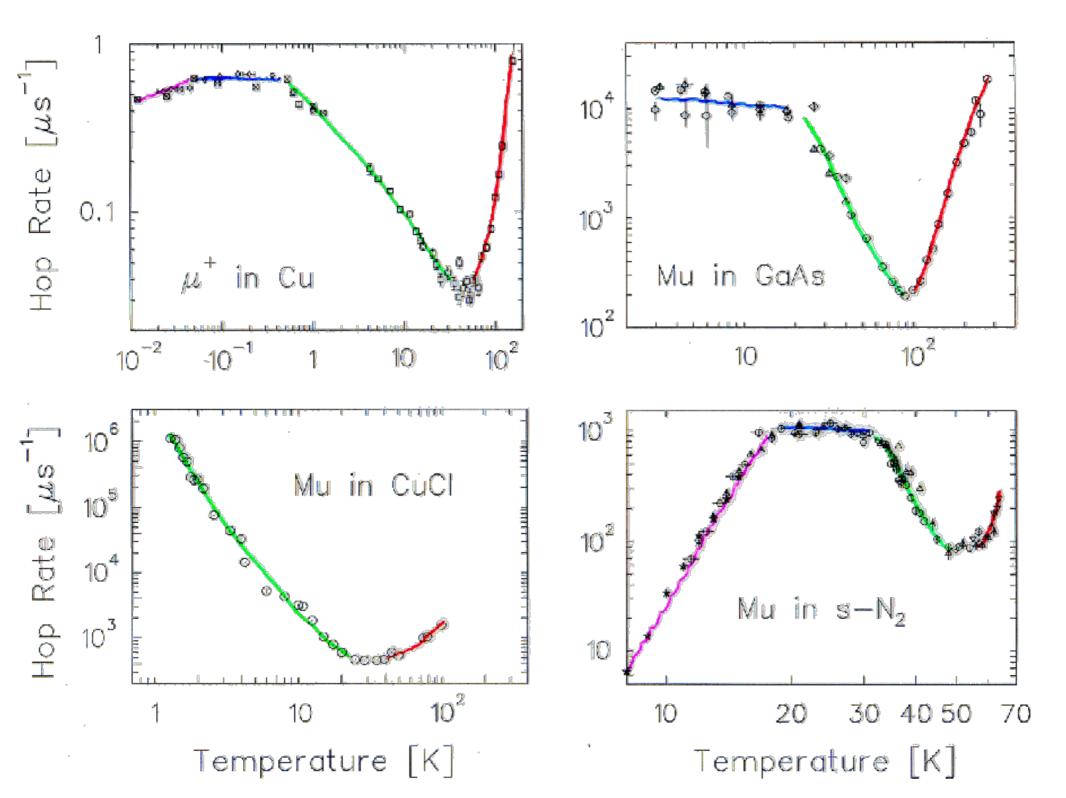
Quantum Diffusion







Quantum Diffusion



Thermally activated overbarrier *hopping* (incoherent).

↑ hot

Phonon scattering "spoils" coherent delocalization of lattice polarons.

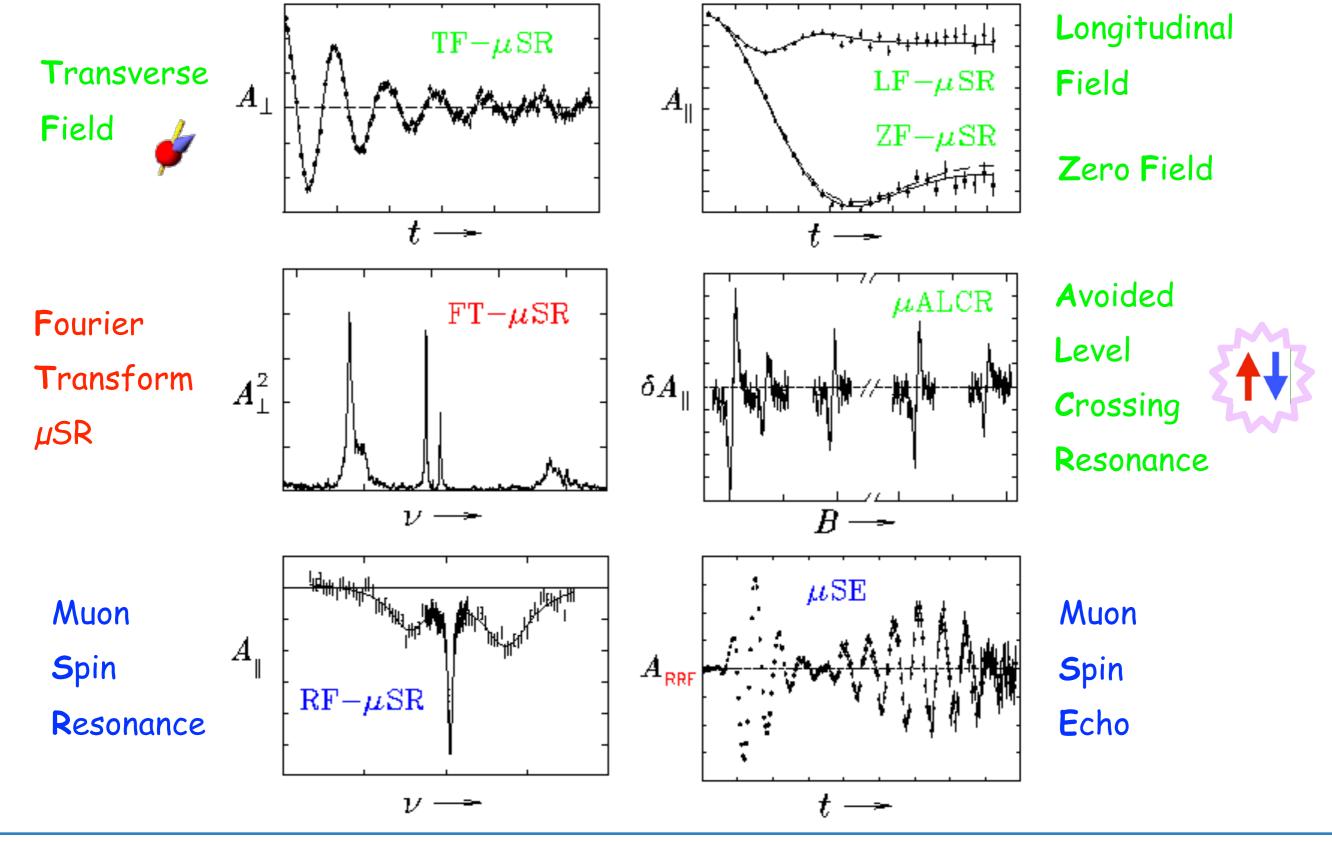
1 warm

Delocalized states find dilute defects and trap.

↓ cold

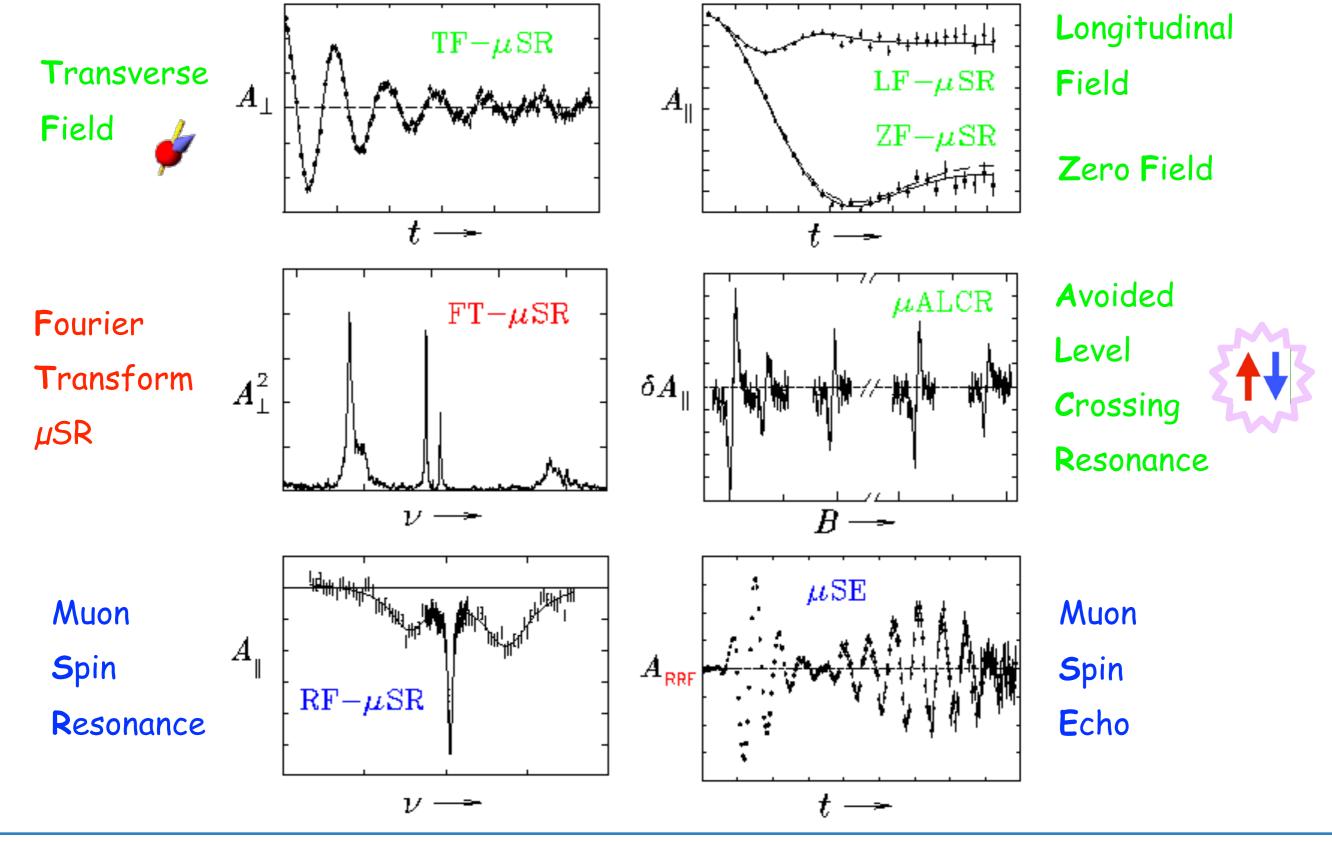






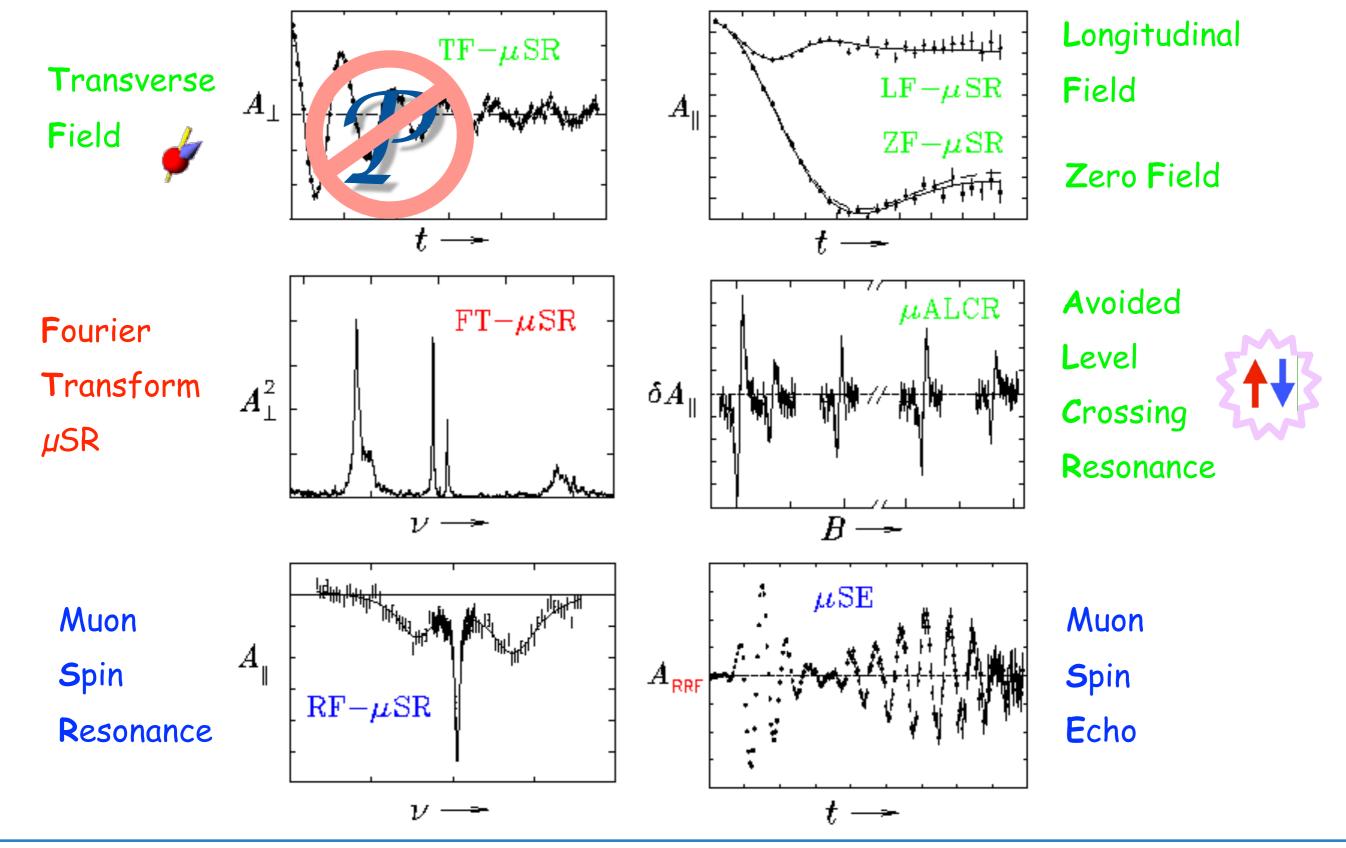






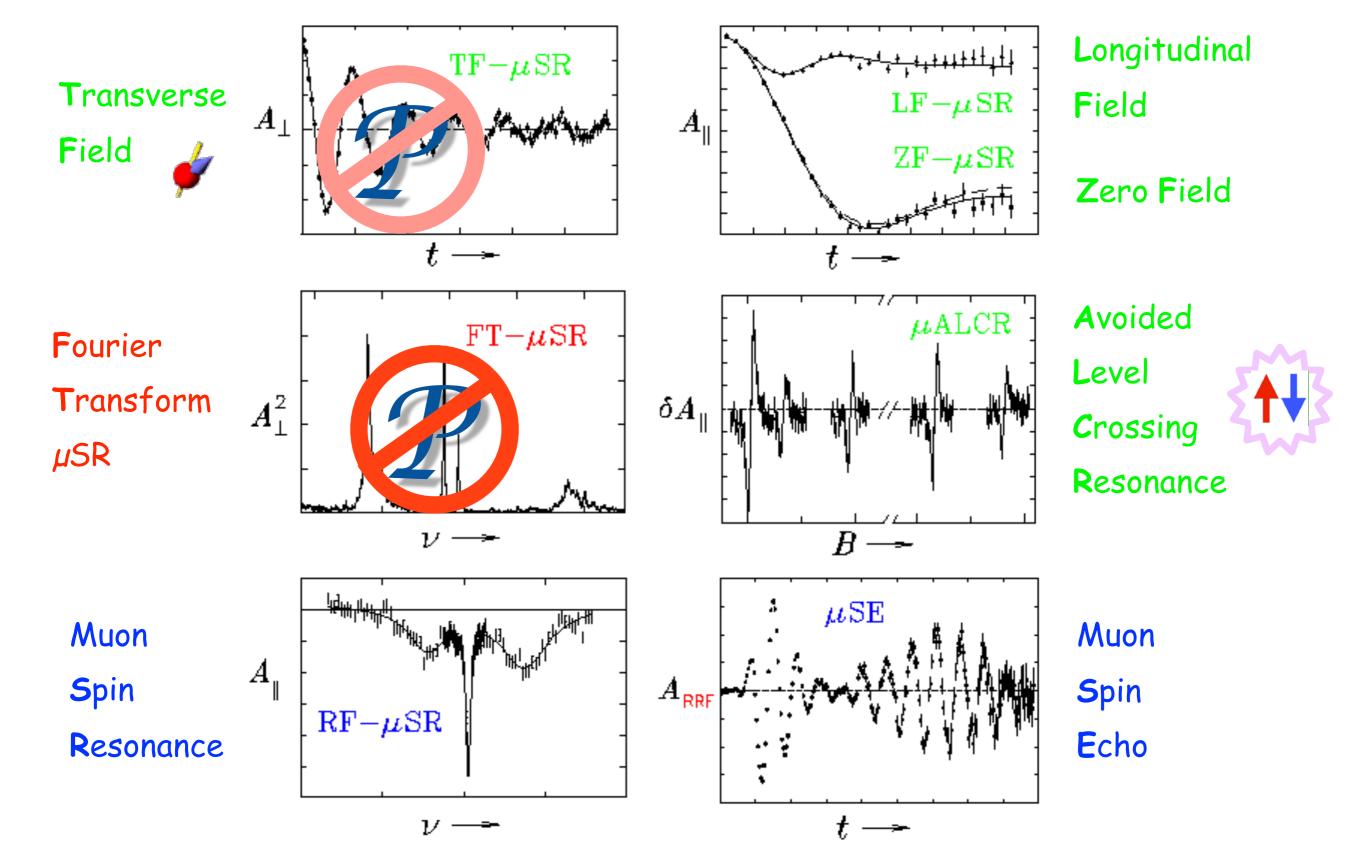








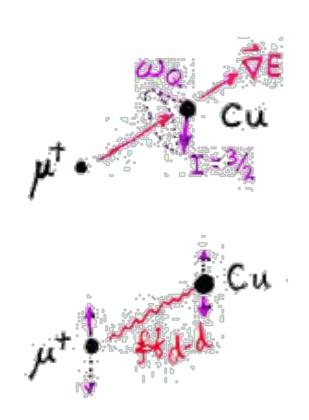


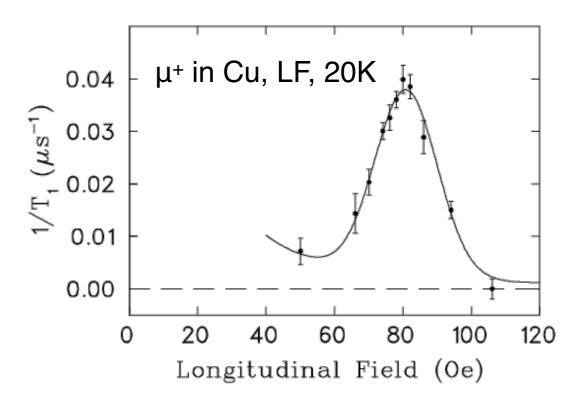






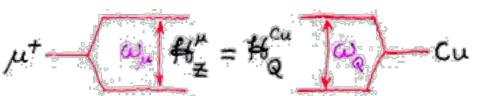
Avoided Level-Crossing Resonance

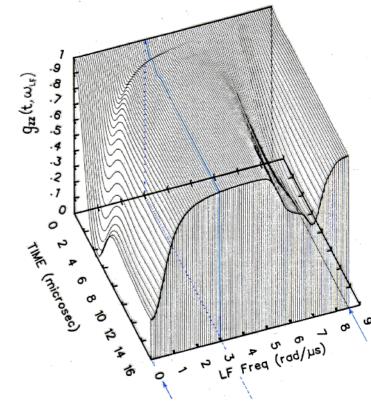




4

Nuclear Quadrupolar version

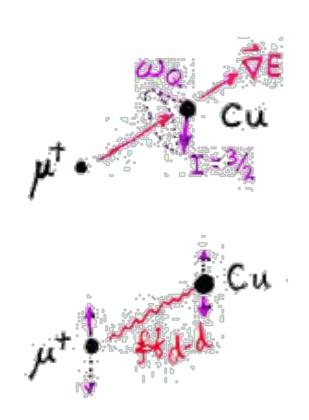


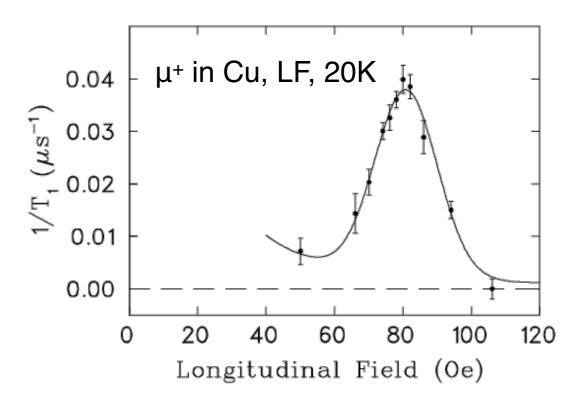






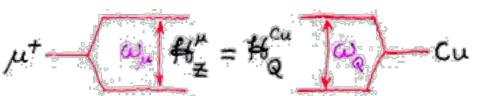
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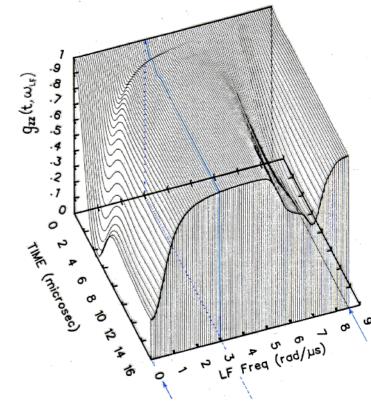




4

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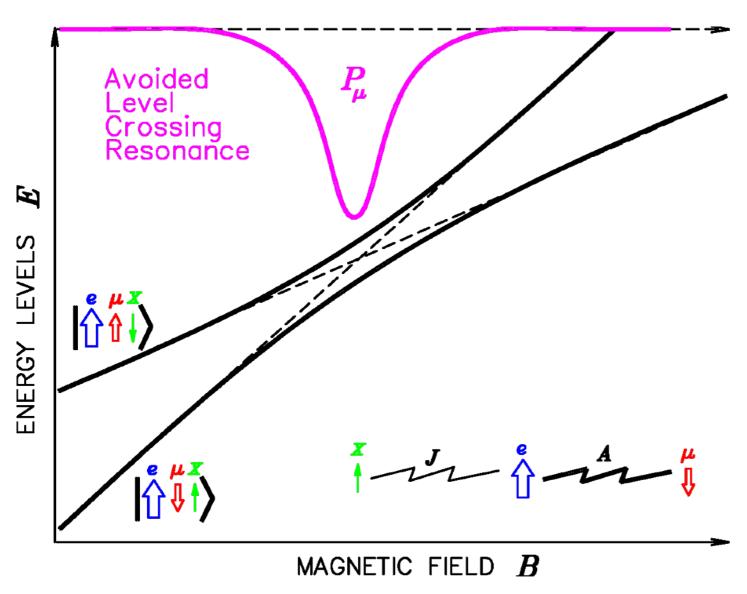




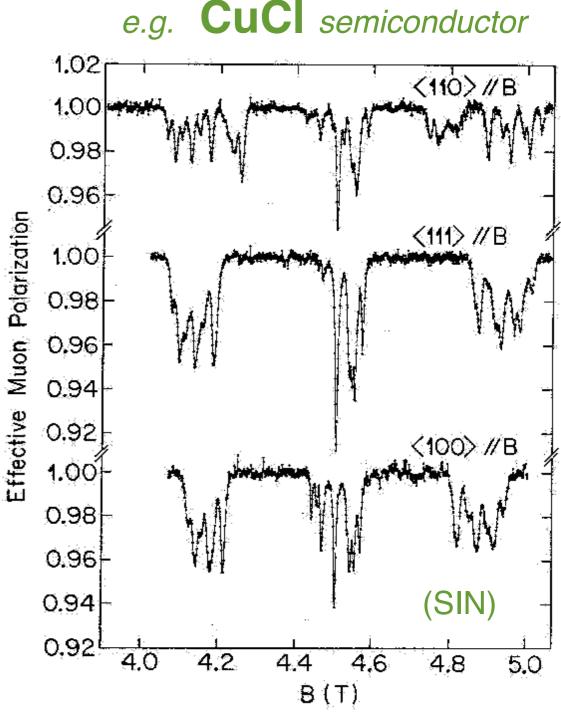




Avoided Level-Crossing Resonance



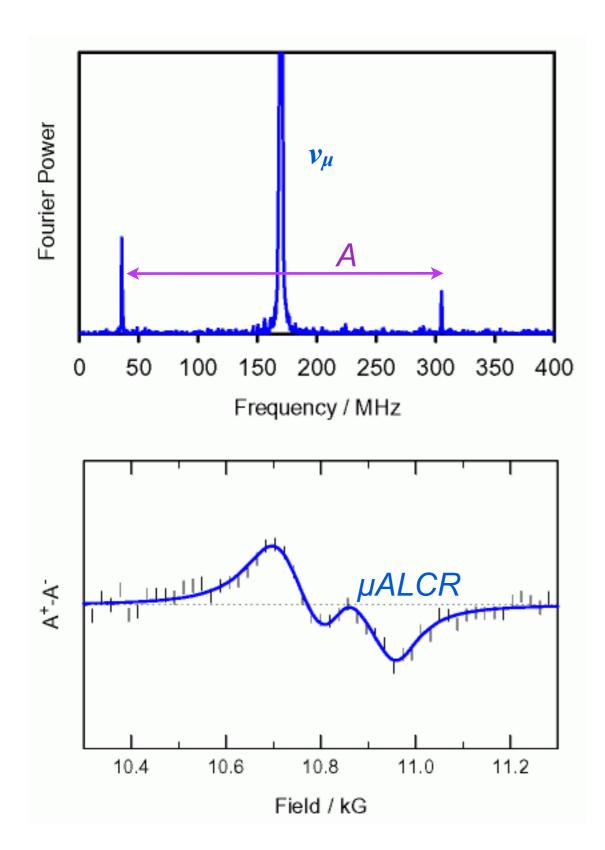
Nuclear Hyperfine version







Muonated Radicals



Organic Free Radicals in Superheated Water

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

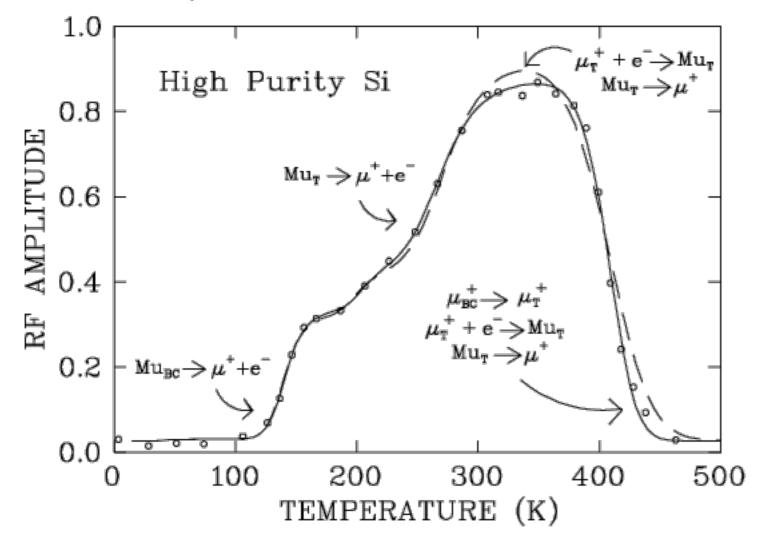
Apparatus has been developed to permit muon avoided levelcrossing spectroscopy (μ LCR) of organic free radicals in water at high temperatures and pressures. The combination of μ LCR with transverse-field muon spin rotation (TF- μ SR) provides the means to identify and characterize free radicals via their 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 hydrothermal 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 2hydroxy-2-propyl. The latter radical is the product of muonium addition to the enol form of acetone and is the subject of an earlier publication. The figure shows spectra for the 2-propyl radical detected in an aqueous solution of 2-propanol at 350°C and 250 bar.

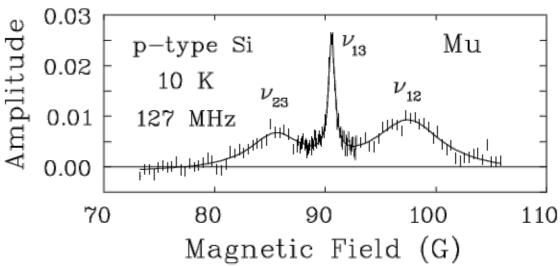




RF-µSR: muon Spin Resonance

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.





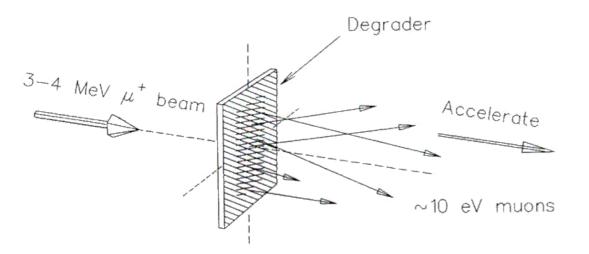
Muonium resonance at ω_{ij} shows fraction of muons in *paramagnetic* states such as Mu itself or a *radical* (paramagnetic molecule). In the above case the field-sweep shows broad ω_{12} and ω_{23} resonances as well as a *sharp two-photon* resonance at their average.

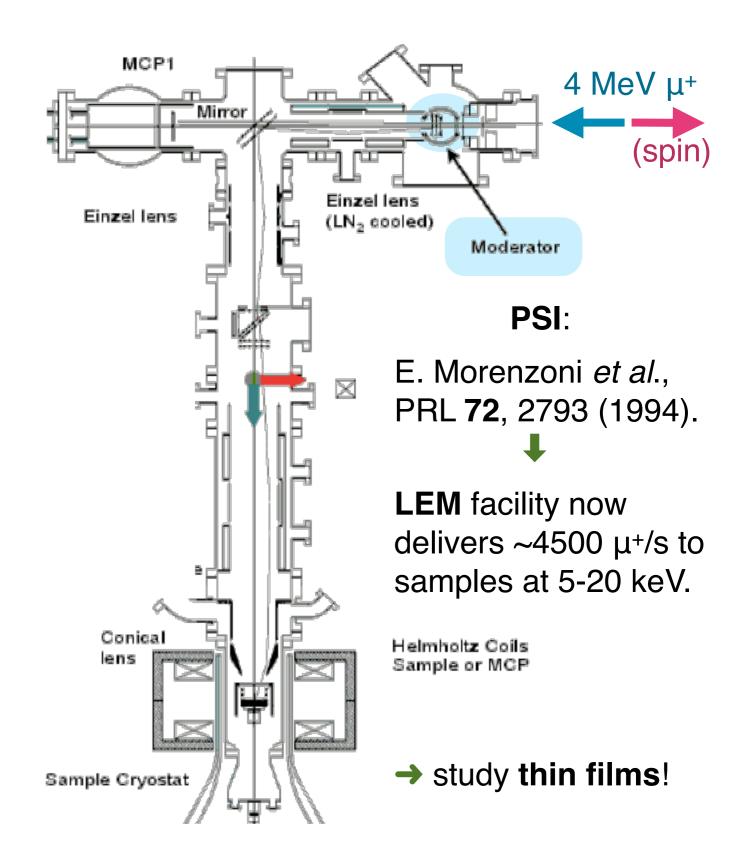
HO GO





Moderated Muons

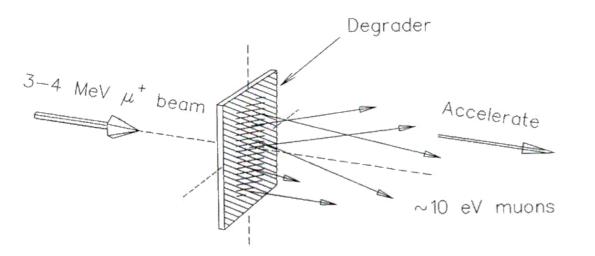






Moderated Muons





TRIUMF:

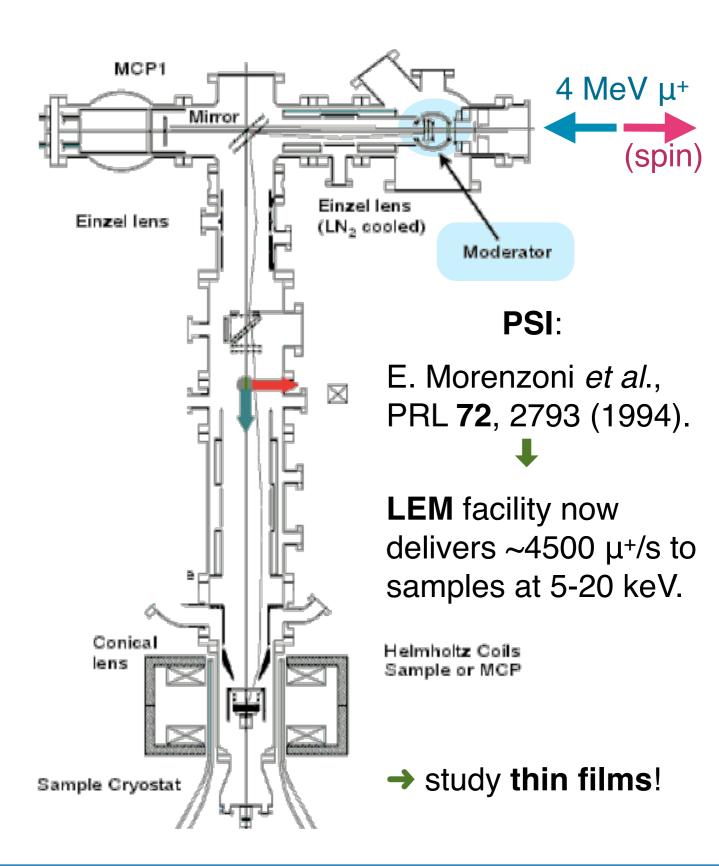
D.R. Harshman *et al.*, PRL **56**, 2850 (1986).



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



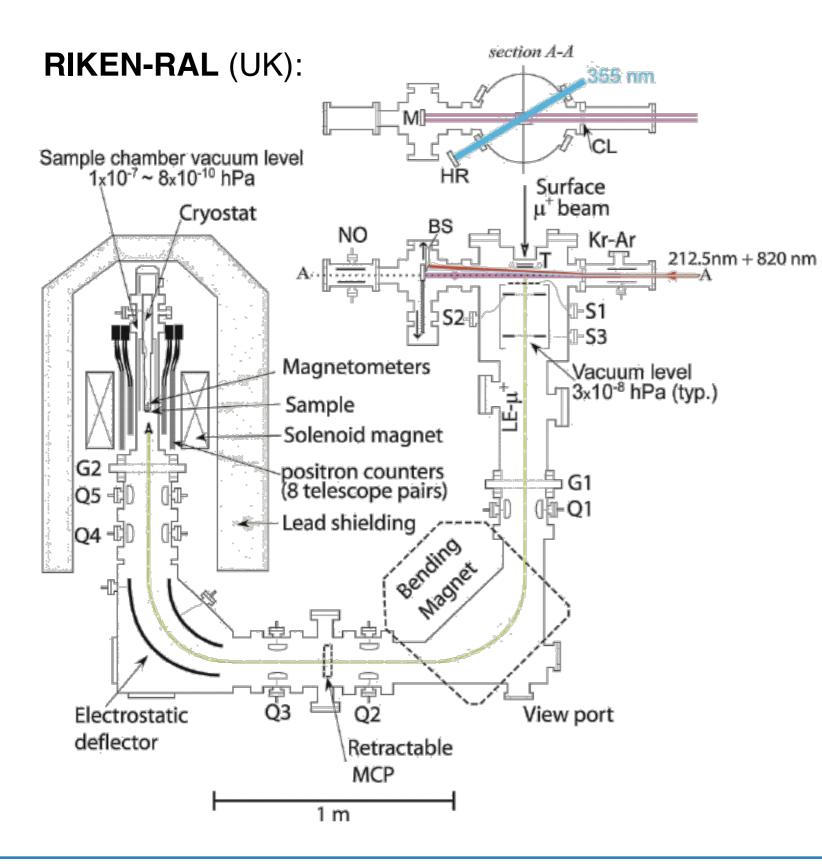
Unfortunately, yield of 1.75(4) epithermal µ+ per 10⁶ incident surface muons (for a solid Ar moderator) was too low to be practical at TRIUMF intensities.







Laser-Ionize Thermal Muonium



J-PARC (Japan):



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

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

- → More LE-µSR
- \rightarrow Improved muon g-2





High Field µSR



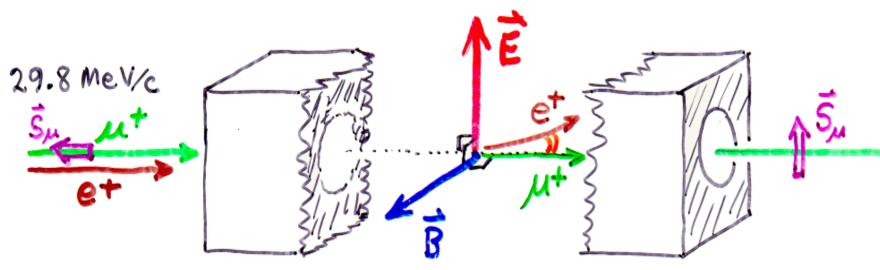
Fields of up to 10 T are now available, requiring a "business end" of the spectrometer < 3 cm in diameter (so that 30-50 MeV decay positron orbits don't "curl up" and miss the detectors) and a time resolution of ~ 150 ps. Muonium precession frequencies of over 2 GHz have been studied.



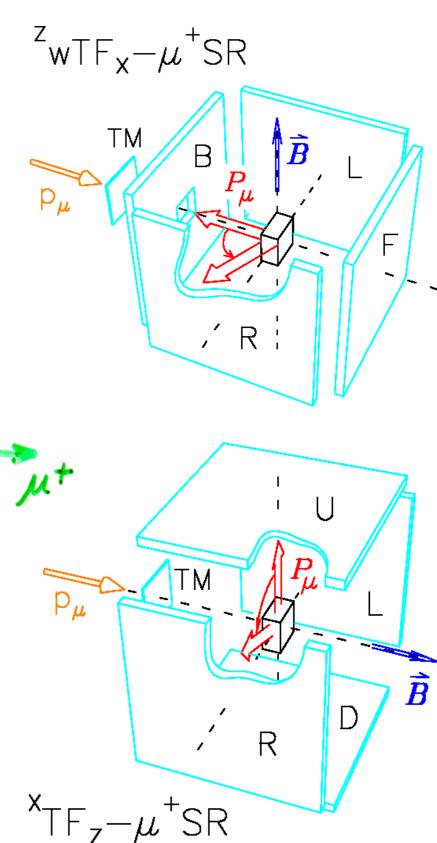


ExB velocity selector & Spin Rotator

("DC Separator" or Wien filter) for surface muons:



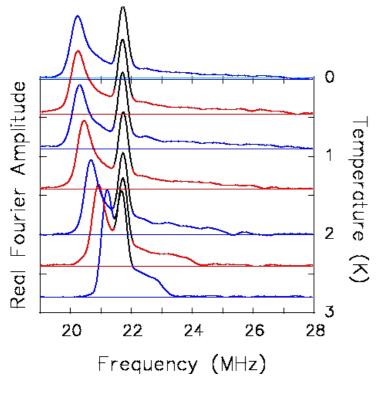
- Removes beam positrons
- Allows TF- μ +SR in high field (otherwise B deflects beam)

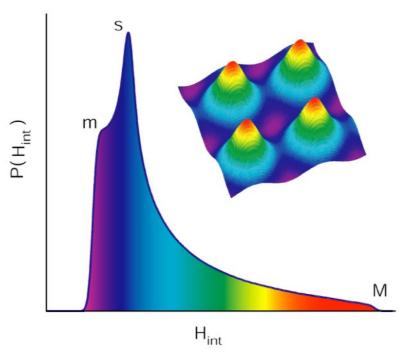


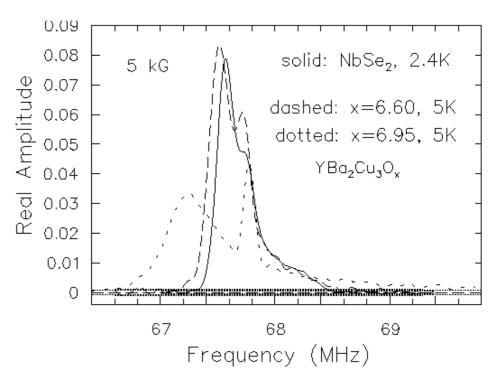


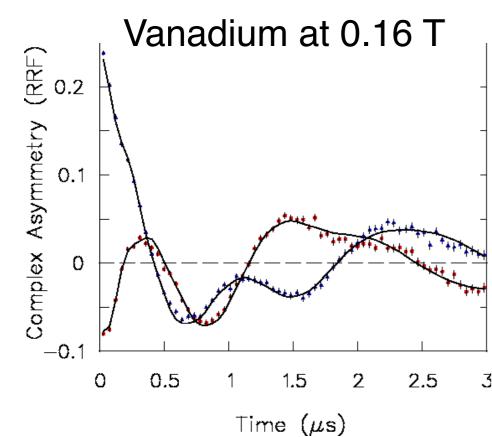


Type-II Superconductors



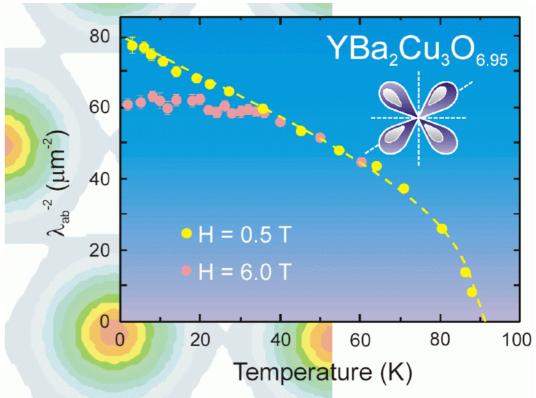






Fitting must always be done in the *time* domain, because of the "noise" from late times (low statistics).

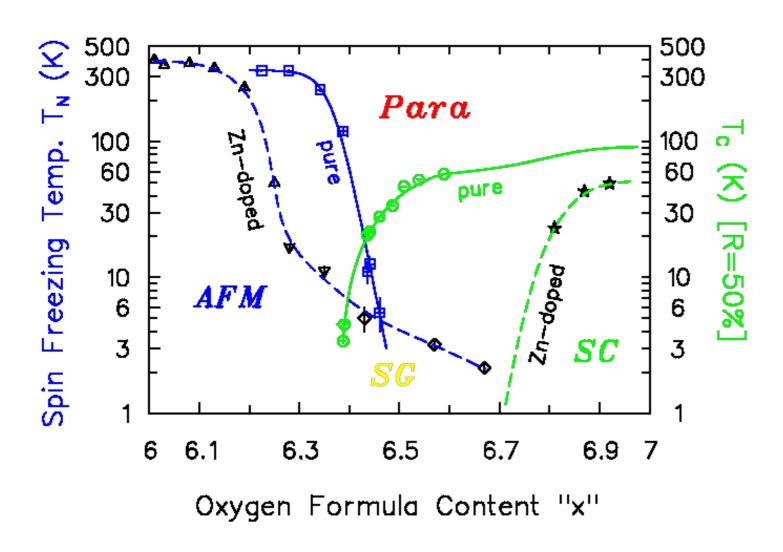
Extract magnetic penetration depth λ_{ab} $(\lambda_{ab}^{-2} \propto n_s)$.

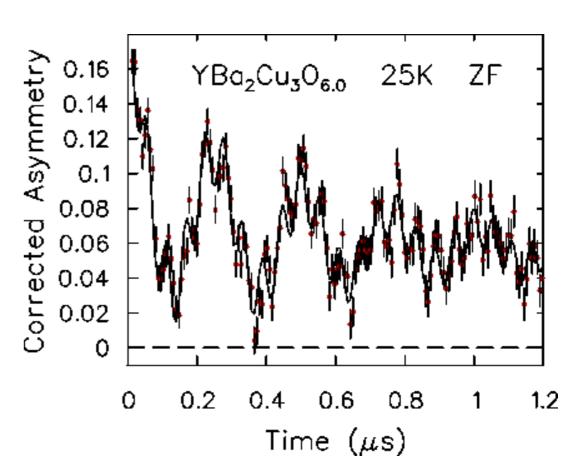






Coexistence of SC & Magnetism



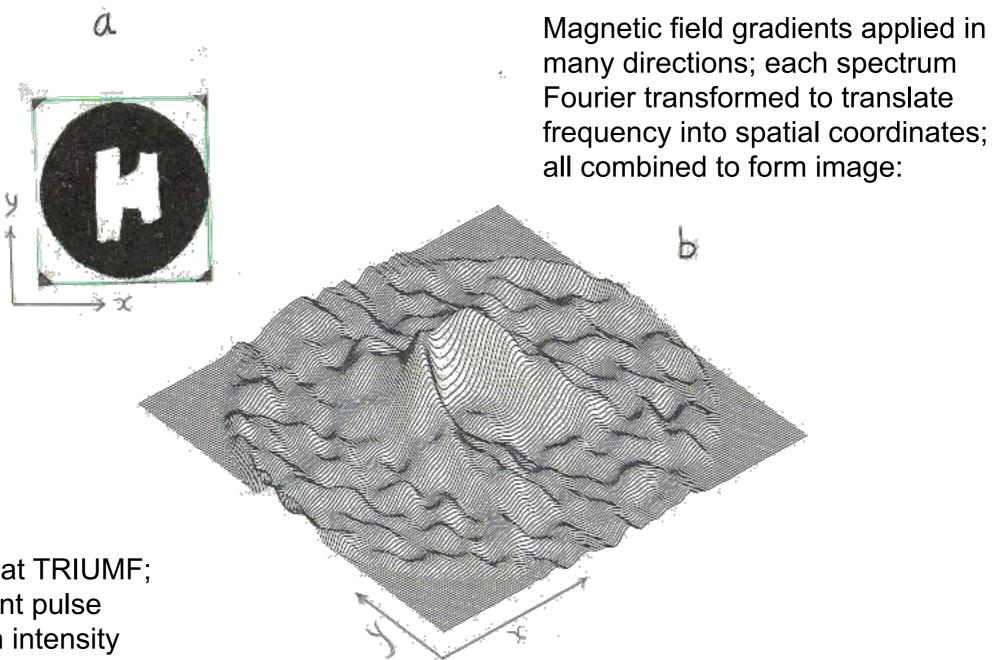






µSI: Muon Spin Imaging

"µ" cut from Al on depolarizing substrate:



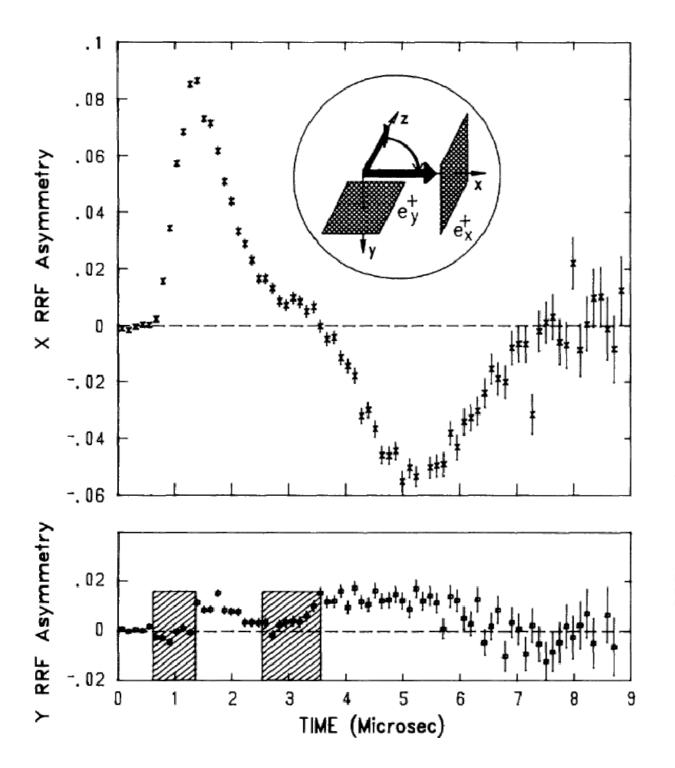
Deemed impractical at TRIUMF; but with RF & gradient pulse sequences at a high intensity pulsed facility, who knows...?

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



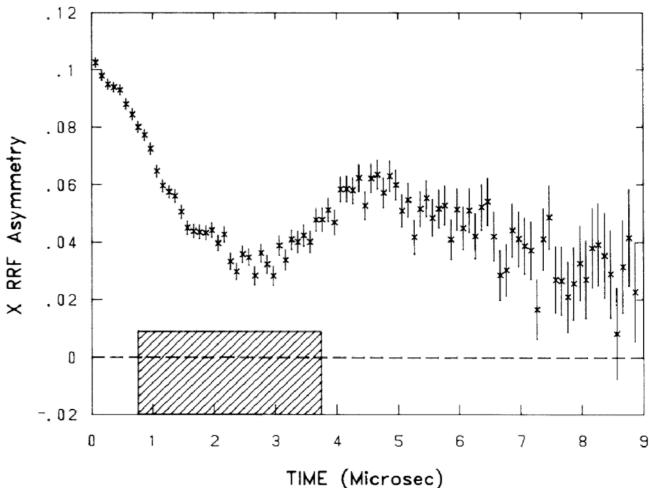


RF-µSE: muon Spin Echo



← Direct μSE in LaF3: muons enter with spins along B_0 . A $\Pi/2$ RF pulse at ω_{μ} "flips them up" and they precess and "dephase"; a Π pulse at time τ makes them "refocus".

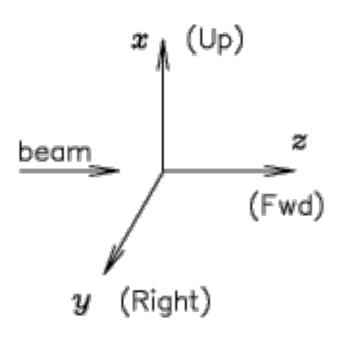
Indirect \muSE: muon spins initially $\perp B_0$ are refocused by a Π pulse on the ¹⁹F nuclei at frequency ω_F .

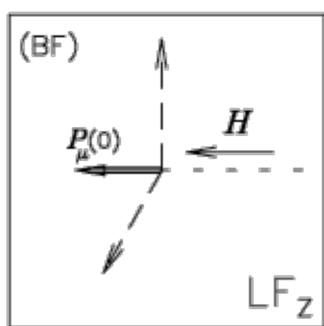


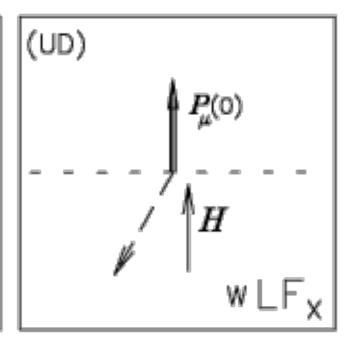


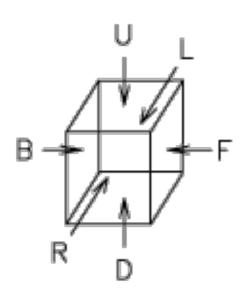


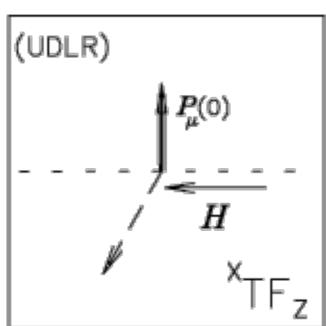
Coordinate Conventions

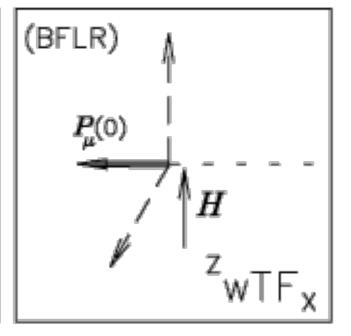










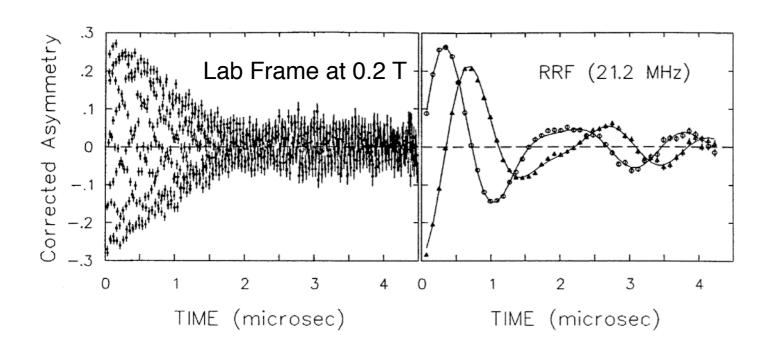




Rotating Reference Frame

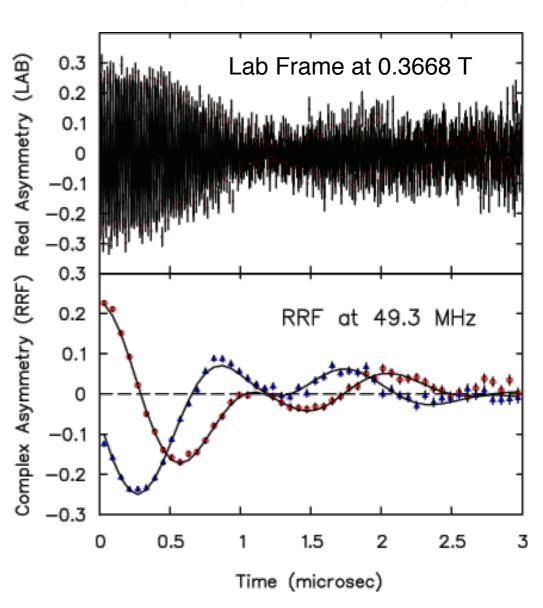
Muon spin precession in high transverse field (HTF-µSR) requires progressively smaller time bins to record the oscillations. These smaller bins capture fewer counts (lower statistics) and require more calculations for fitting. Worse yet, the essential characteristics of the data are not readily observed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

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



$$\mathcal{A}_{\text{LAB}}(t) = A_x(t) + iA_y(t)$$

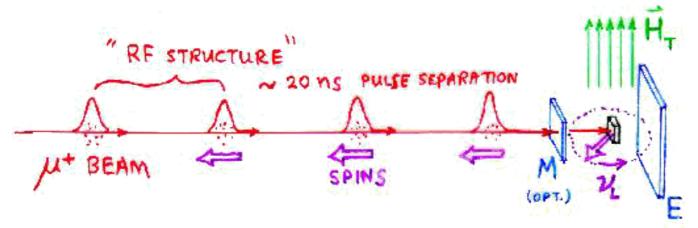
$$\mathcal{A}_{\text{RRF}}(\Omega,t) = e^{-i\Omega t} \mathcal{A}_{\text{LAB}}(t)$$







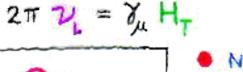
Stroboscopic µSR

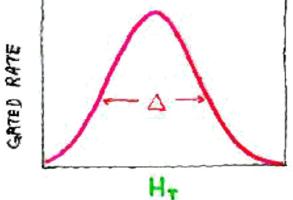


WHEN 1/2 (PRECESSION PERIOD) IS

1/n x INTERVAL BETWEEN MUON PULSES, Linteger (~1 or 2)

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



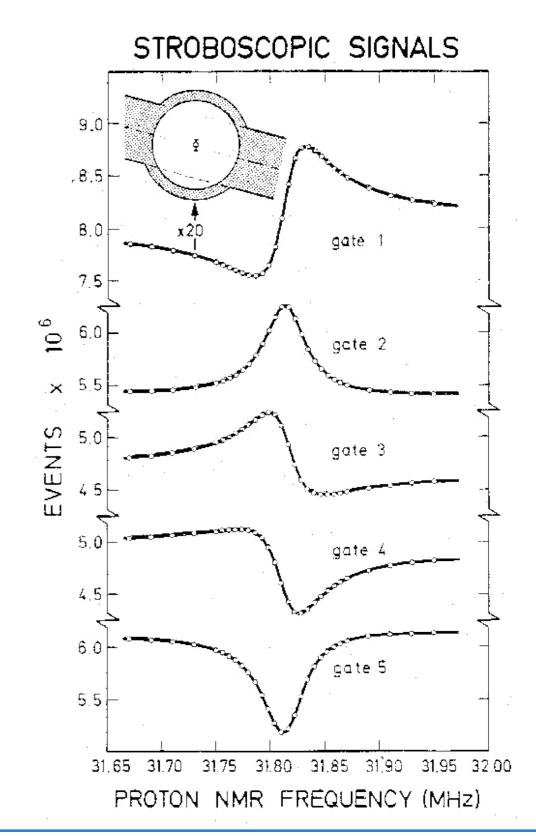


(MANY MUONS IN TARGET)

Gives KMCHT SHIFTS to ~1 ppm

PROBLEM: INTAINSIC WIDTH

$$\Delta = \frac{1}{2} = 0.454 \, \mu s^{-1}$$

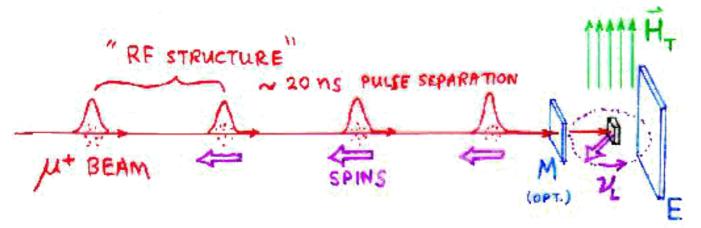






Stroboscopic µSR

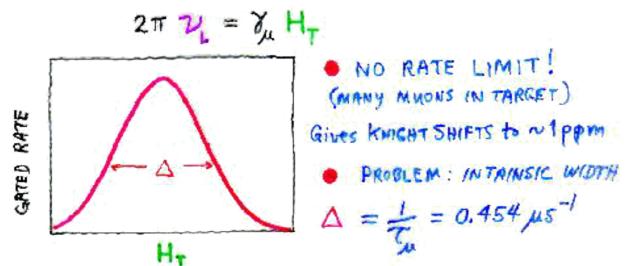
Schenck et al. - SIN

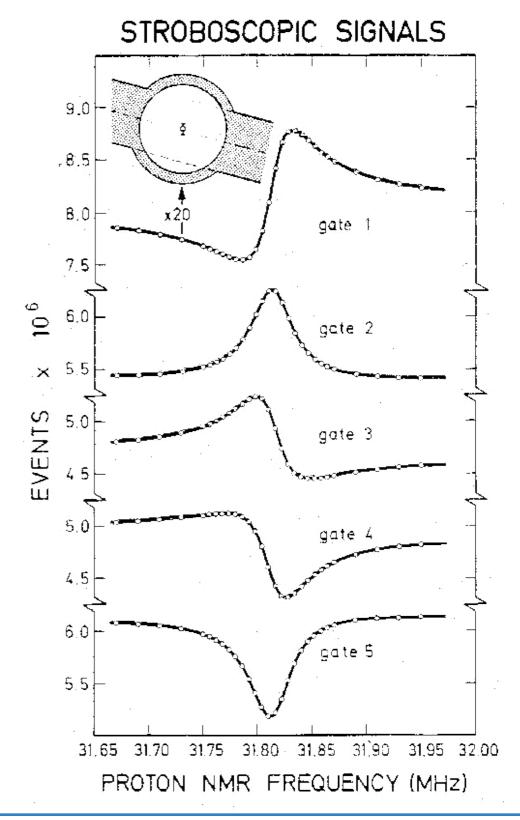


WHEN 1/1 (PRECESSION PERIOD) IS

1/n x INTERVAL BETWEEN MUON PULSES, Linteger (~1 or 2)

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

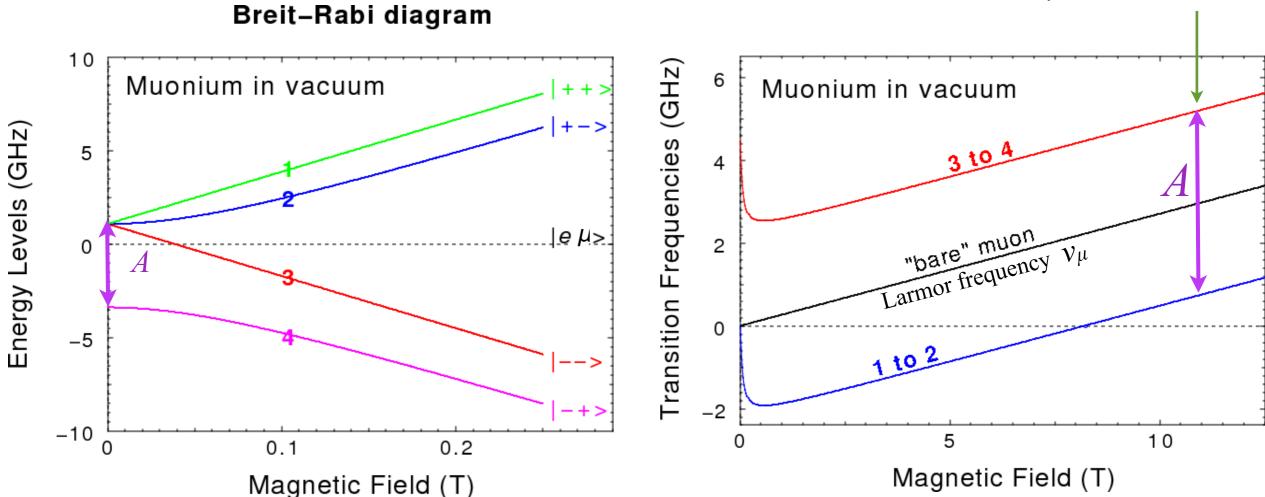






Muonium (Mu≡μ+e⁻) and FFT Spectroscopy

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

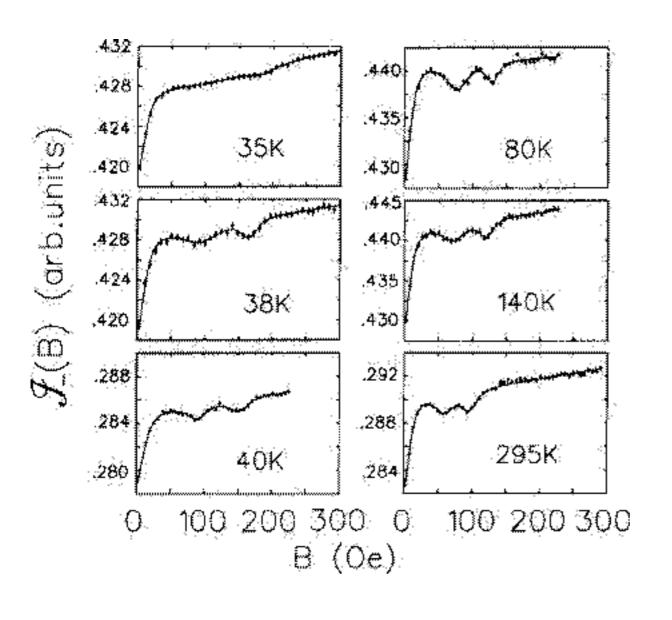


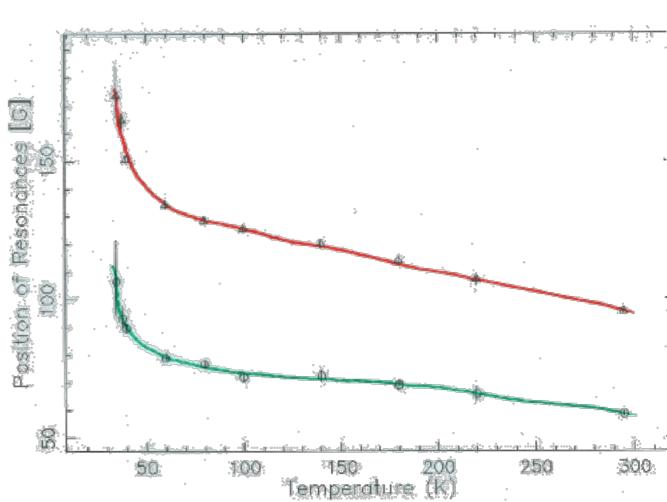
"Signature" of Mu (or other hyperfine-coupled μ^+e^- spin states) in high transverse field: two frequencies centred on v_μ and separated by the hyperfine splitting $\mathbf{A} \propto r^{-3}$.





Avoided Level-Crossing Resonance Nuclear Quadrupolar version: MnSi

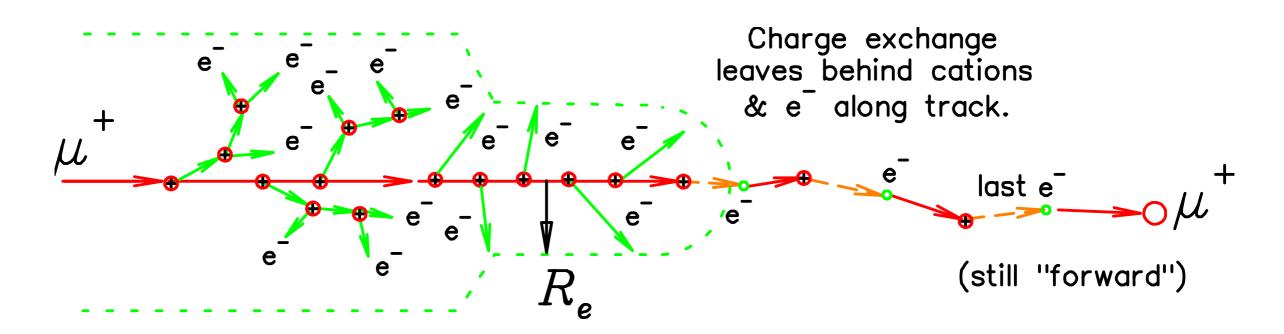




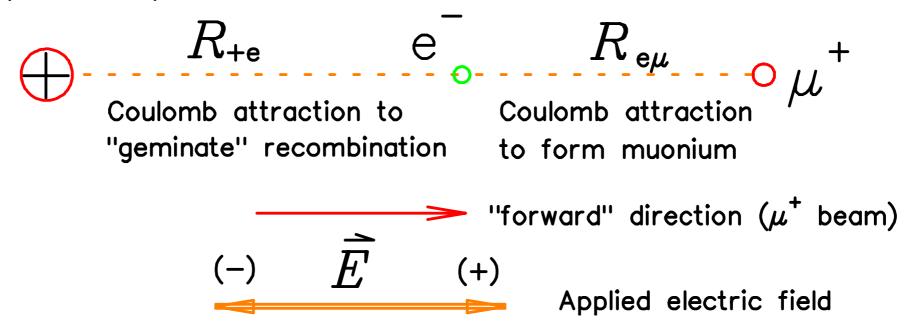




Radiolysis & "Delayed" Muonium Formation



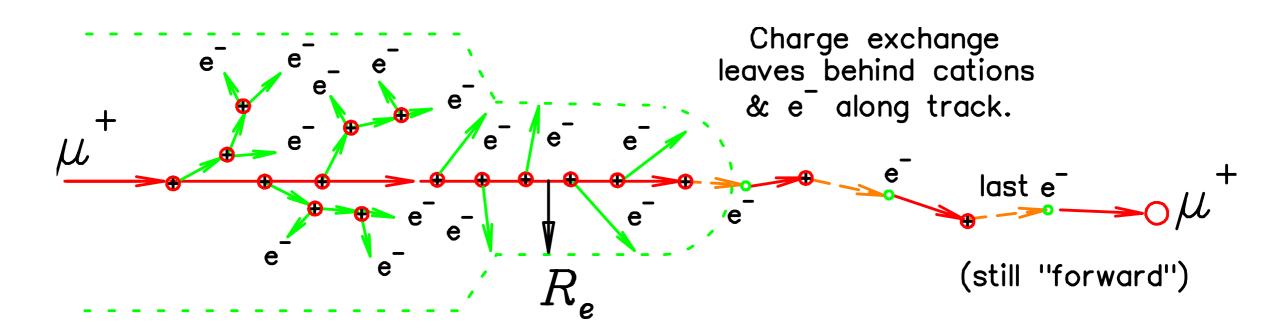
RESULT (sometimes):



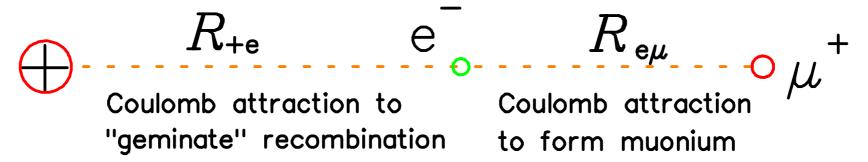




Radiolysis & "Delayed" Muonium Formation

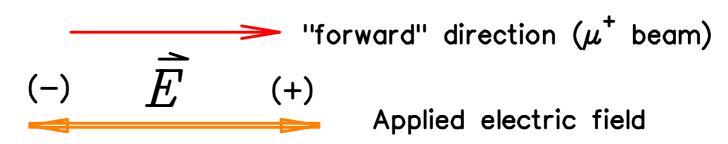


RESULT (sometimes):



Possible exceptions:

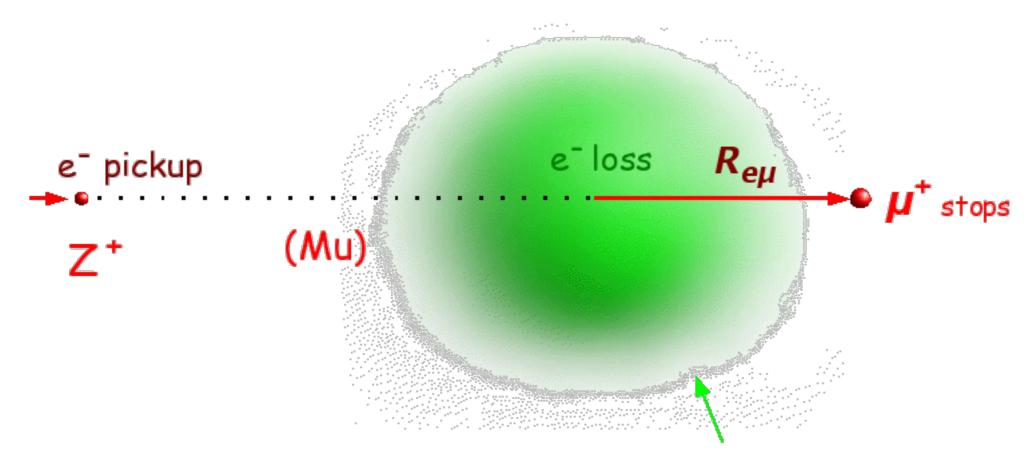
solids with extremely high electron mobilities.





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

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



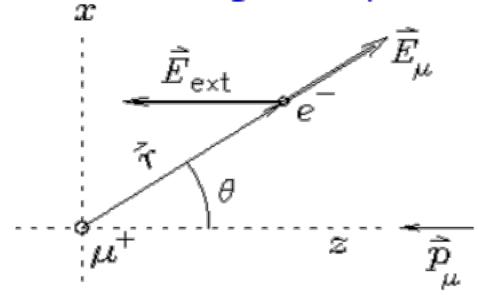
Distribution of "initial" e⁻ positions may <u>overlap</u> position where µ⁺ stops



Muonium Formation in an Electric Field



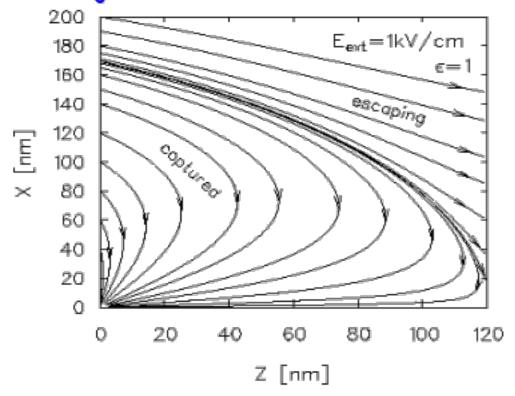




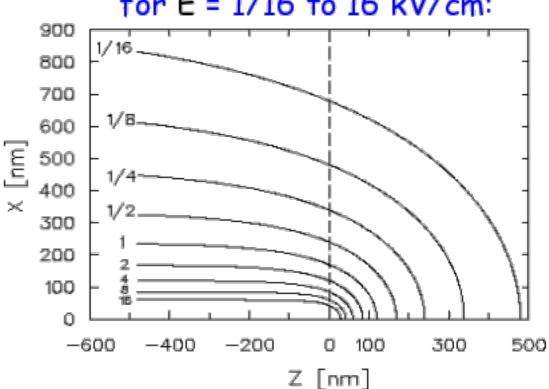
Viscous flow model:

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

Trajectories for E = 1 kV/cm:



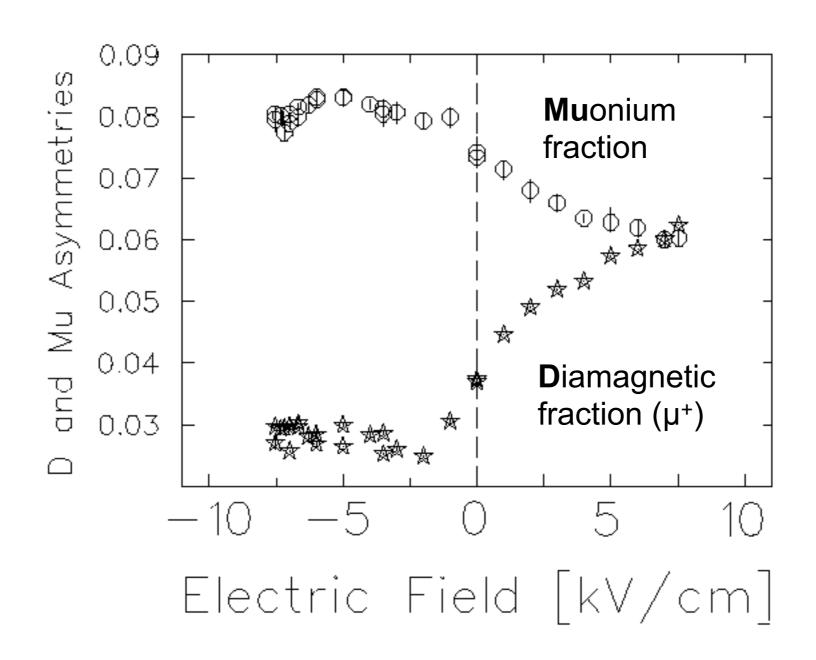
e capture boundaries for E = 1/16 to 16 kV/cm:





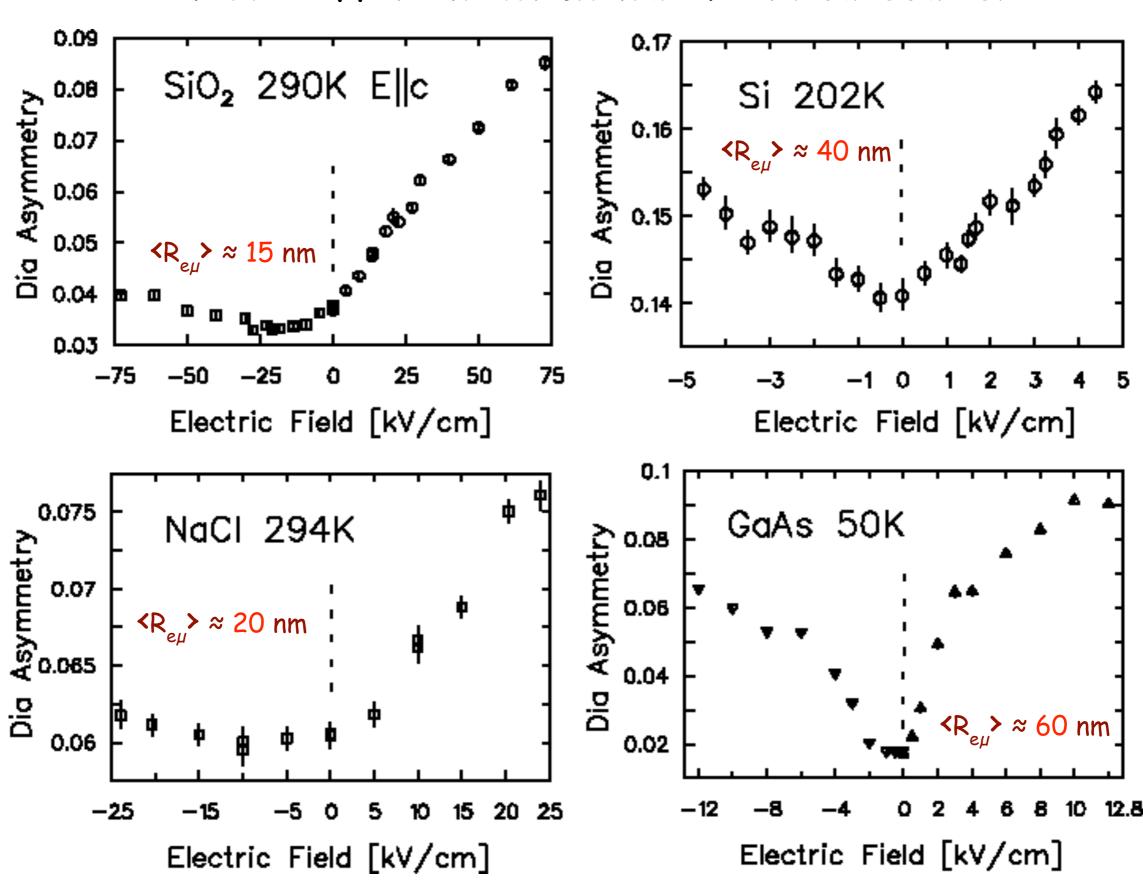


Delayed Mu Formation in Cryocrystals (e.g. s-N₂)



"Ordinary" Solids: Insulators & Semiconductors

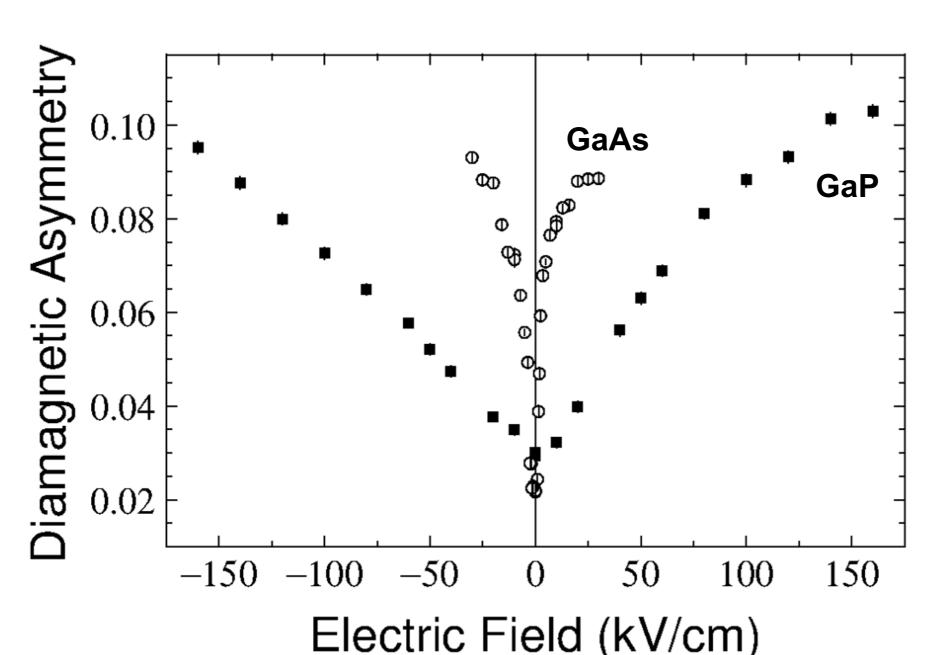
Note different horizontal & vertical scales!







Weakly Bound Mu States in High-Mobility Semiconductors



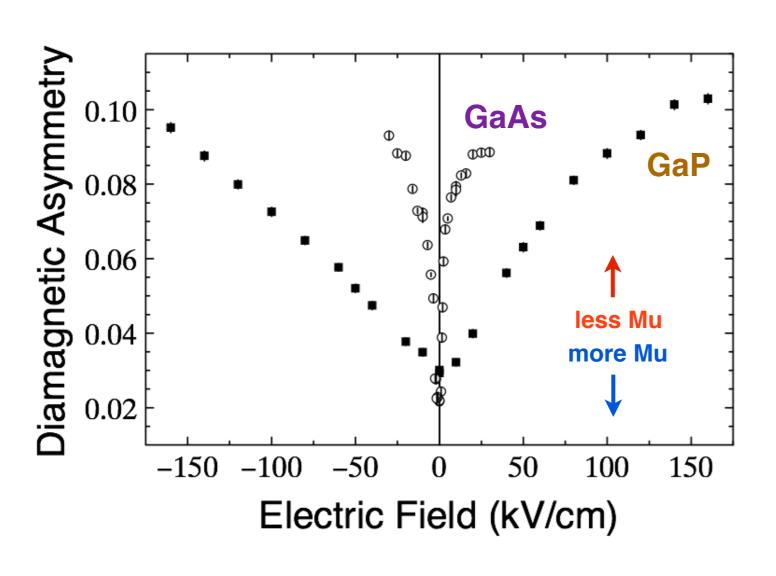
Initial states Mu_{wb} have electron orbitals "out in the lattice" with different effective masses; those with higher *m** are more strongly bound and harder to ionize with an applied *E* field.

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



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 μ SR Literature Entry # 2437

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

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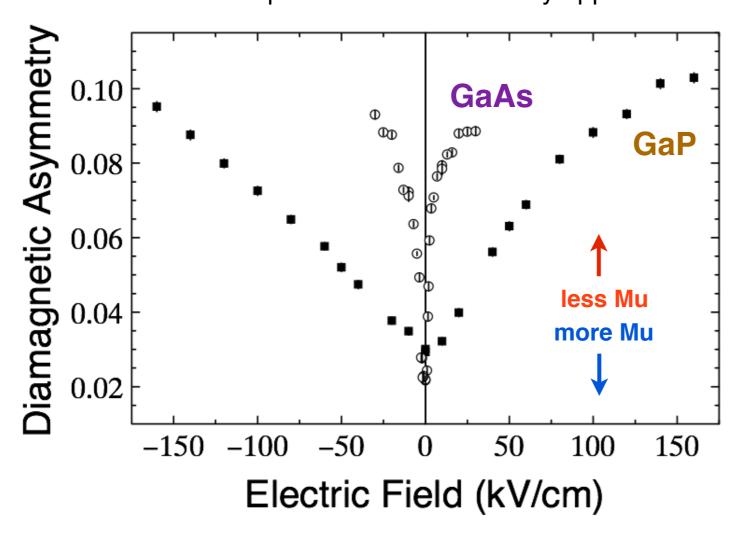




Weakly Bound Muonium States in GaAs & GaP

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

Lighter effective mass & higher mobility $e^- \Rightarrow$ 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.

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