

# $\mu$ SR - Muon spin rotation, relaxation and resonance

The muon is an extremely sensitive probe ( $\sim 10^{-3} \mu_B$ ) for static and fluctuating magnetic fields and field distributions (spin 1/2 , no quadrupole interaction) in condensed matter.

- Muon and its properties
- Muon production and muon beams
- Muon interaction with matter
- Technical details of  $\mu$ SR experiments
- Areas of  $\mu$ SR applications

# Some Bibliography on $\mu$ SR

## Books

- A. Schenck, Muon Spin Rotation Spectroscopy, (Adam Hilger, Bristol, 1985)
- E. Karlsson, Solid State Phenomena, As Seen by Muons, Protons, And Excited Nuclei, (Clarendon, Oxford 1995)
- A. Yaouanc and P. Dalmás de Réotier, Muon Spin Rotation, Relaxation and Resonance: Applications to Condensed Matter, (Oxford University Press, Oxford, 2011)

## Reviews

- [http://inac.cea.fr/Pisp/pierre.dalmas-de-reotier/introduction\\_muSR.pdf](http://inac.cea.fr/Pisp/pierre.dalmas-de-reotier/introduction_muSR.pdf)
- <http://musr.ca/intro/musr/muSRBrochure.pdf>

## Web sites:

The TRIUMF  $\mu$ SR User Facility <http://musr.triumf.ca/>

Paul Scherrer Institut (PSI) Laboratory for Muon Spin Spectroscopy <http://lmu.web.psi.ch/>

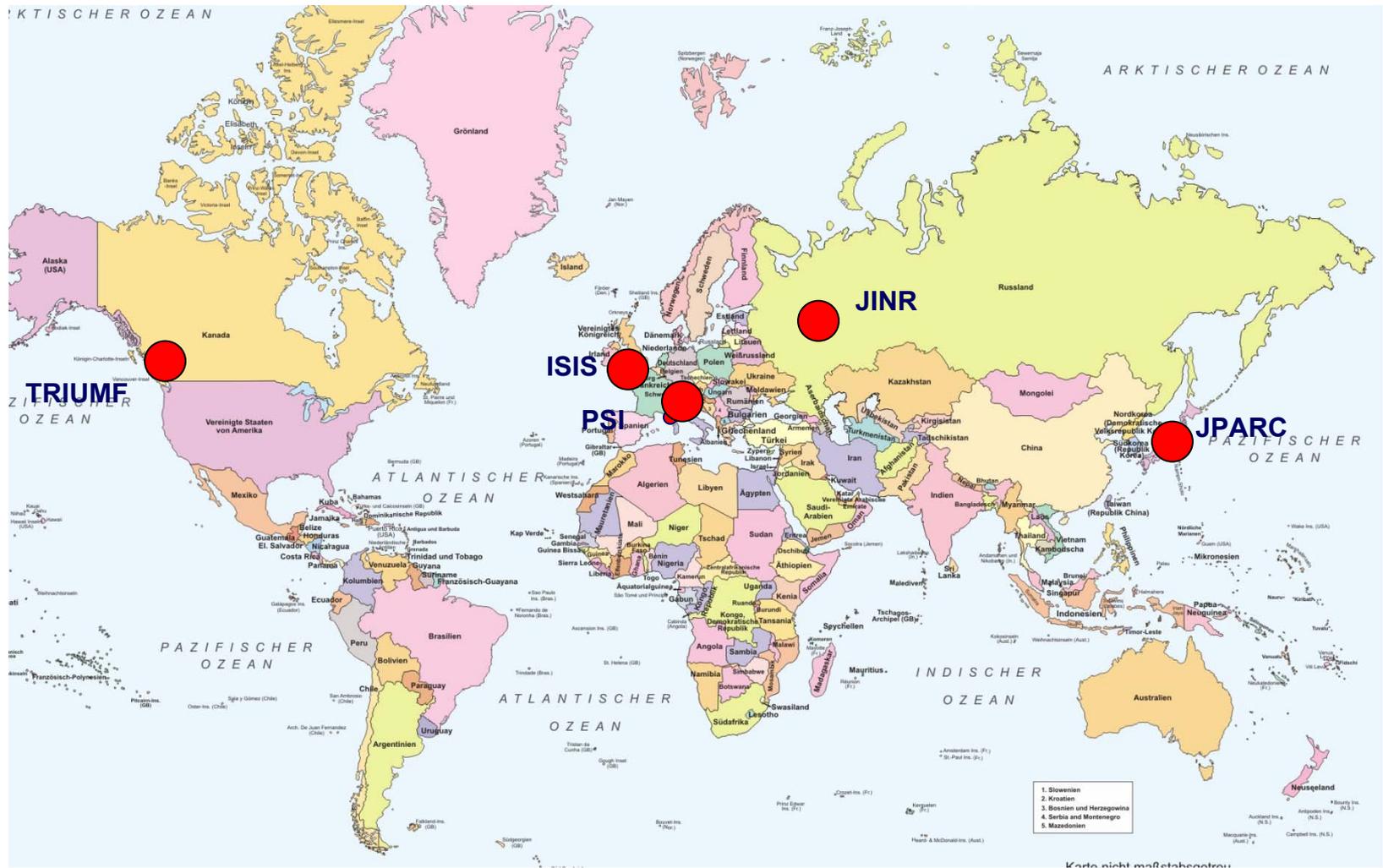
The ISIS Pulsed Muon Facility <http://www.isis.rl.ac.uk/muons/>

KEK Meson Science Laboratory <http://msl-www.kek.jp/>

The RIKEN-RAL Muon Facility <http://nectar.nd.rl.ac.uk/~rikenral/>

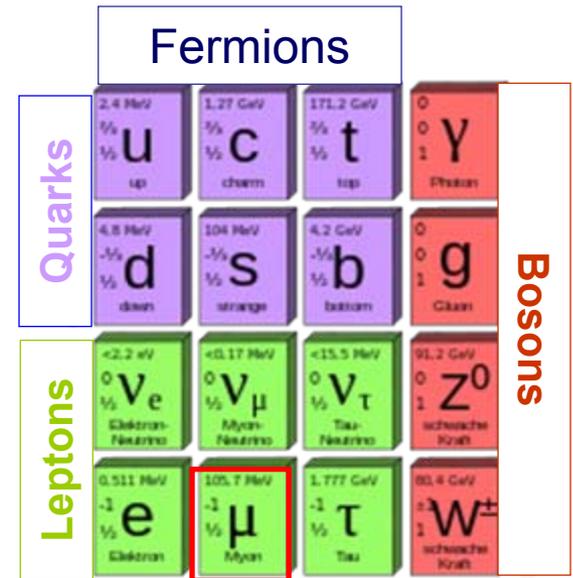
High Intensity Proton Accelerator Facility <http://jkj.tokai.jaeri.go.jp/>

# Global Muon Facilities



# Muon Properties

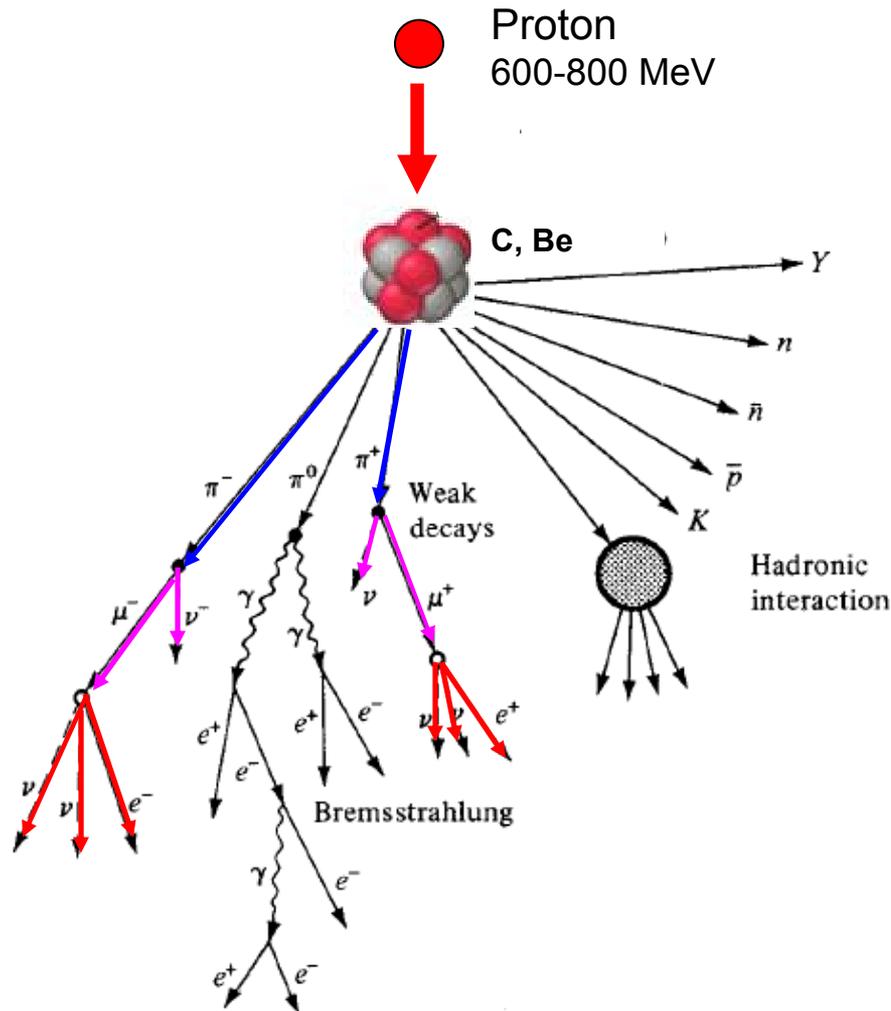
**Leptons** and **quarks** are the basic building blocks of matter, i.e., they are seen as the "elementary particles". There are six leptons in the present structure, the electron, muon and tau particles and their associated neutrinos.



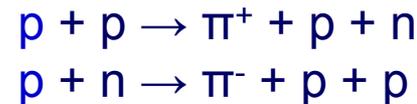
Particle/antiparticle	Spin	Charge	Mass	Magnetic moment	lifetime (μs) (free particle)
muon/antimuon	1/2	-/+ e	105.7 MeV 207 m <sub>e</sub> 0.11 m <sub>p</sub>	3.18 μ <sub>p</sub>	2.19
electron/positron	1/2	-/+ e	511 keV m <sub>e</sub>	657 μ <sub>p</sub>	stable
Proton/antiproton	1/2	+/- e	938 MeV m <sub>p</sub> = 1836 m <sub>e</sub>	μ <sub>p</sub> (1,410 606 · 10 <sup>-26</sup> J/T)	Stable (>10 <sup>33</sup> a)

**A positive muon μ<sup>+</sup> can be considered as an unstable light isotope of hydrogen (0.11 mp), a negative muon μ<sup>-</sup> as an unstable heavy electron (207 me)**

# Muon production



Protons of 600 to 800 MeV kinetic energy interact with protons or neutrons of the nuclei of a light element target (typically graphite) to produce pions ( $\pi^+$ ) as intermediate particles .



Pions are unstable (lifetime 26 ns) and decay into muons (and neutrinos):

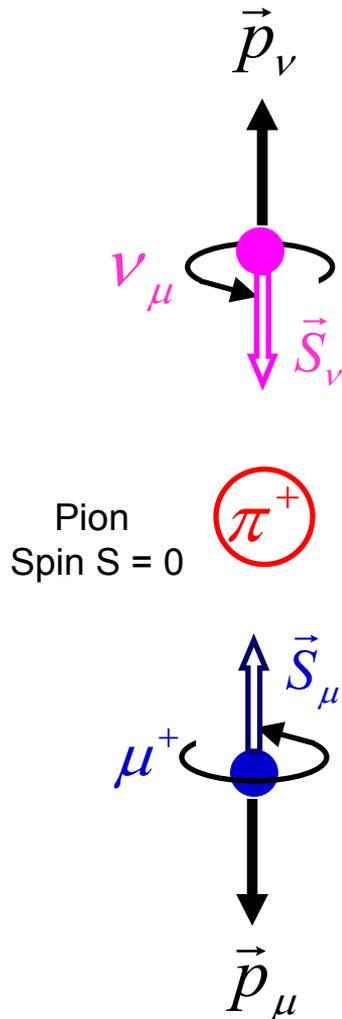


Muons are unstable (free particle lifetime 2.197  $\mu$ s) and decay into electrons/positrons and neutrinos



For  $\pi$  at rest:  $E_{\text{kin}}(\mu) = 4.12 \text{ MeV}$

# Muon polarization and decay kinematics



The neutrino of the weak interaction decay



has helicity  $-1$ , i.e. its spin is opposite to the momentum

Momentum conservation of the pion decay at rest leads to **opposite momenta of muon and neutrino**. Since the pion has spin 0, **the fermions  $\mu^+$  and  $\nu_\mu$  have opposite spins**.

As a consequence, the muon beam is **polarized**, i.e. the muon spin is **opposite to the momentum**

## Kinematics of pion decay at rest:

Energy and momentum conservation result in:

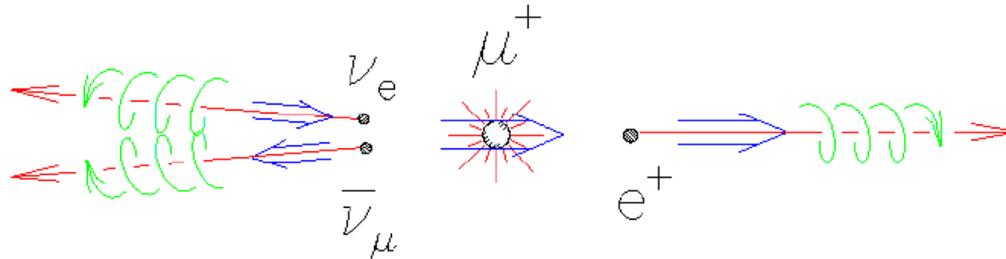
$\mu$  momentum:  $p = 29.79 \text{ MeV}/c$

$\mu$  kinetic energy:  $E = 4.12 \text{ MeV}$

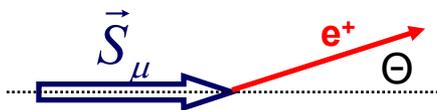
# Muon polarization and correlation with the positron trajectory



Due to the parity violation of the muon decay, the muon emits a fast positron (electron) preferentially along the direction of its spin.

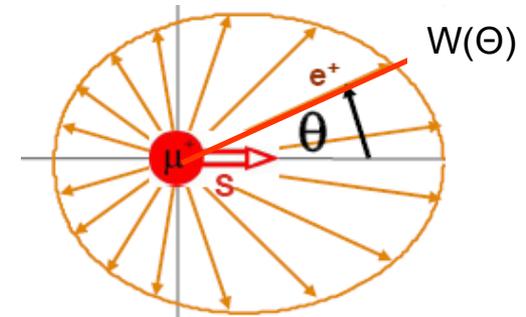


The positron emission probability is given by:



$$W(\Theta)d\Theta \propto (1 + a(E)\cos(\Theta))d\Theta$$

$$\text{Energy average: } \overline{a(E)} = 1/3$$

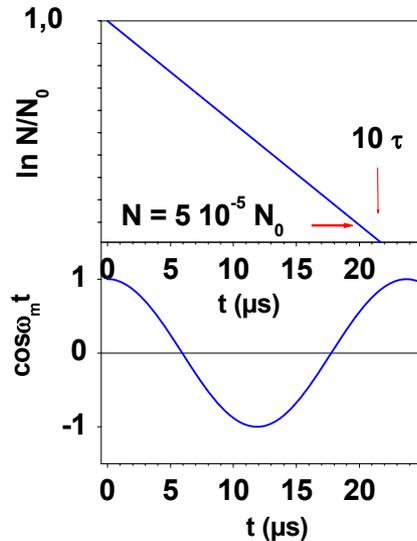


**This anisotropic emission (*polarisation*) constitutes the basis for the  $\mu$ SR technique.**

**As muon spin and positron direction are correlated, detection of the positron carries information on the orientation of the muon spin at the time of decay, similar to perturbed  $\gamma\gamma$ -angular correlations (PAC).**

# The muon as a probe of magnetic fields in condensed matter

Assume: At least one Larmor precession fits into the observation time window  $\Delta t = 20 \mu\text{s}$   
 (~ 10 muon life times)



$\tau = 2.19 \mu\text{s}$
$\mu_\mu = 4.8 \cdot 10^{-26} \text{ J/T}$
$\mu_p = 1.4 \cdot 10^{-26} \text{ J/T}$

The corresponding Larmor frequency:

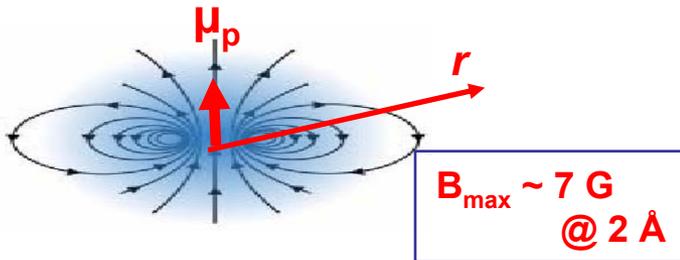
$$\nu_\mu = \frac{\mu_\mu}{h} B = \frac{1}{T} = \frac{1}{2} 10^6 \text{ s}^{-1} \approx 5 \cdot 10^4 \text{ s}^{-1}$$



The lower limit of a magnetic field to be detected by  $\mu\text{SR}$

$$B \geq 5 \cdot 10^4 \text{ s}^{-1} \frac{h}{\mu_\mu} \approx 0.7 \text{ mT} \approx 7 \text{ Gauss}$$

For comparison: the dipolar field of a **proton**



Because its the long life time (= long observation time window) the muon is an **extremely sensitive probe** (~ 0.001  $\mu_B$ ) for static and fluctuating magnetic fields and field distributions in condensed matter

$$B_{dip} = \frac{\mu_0}{4\pi} \frac{3\vec{r} \cdot (\vec{\mu} \cdot \vec{r}) - \mu \cdot r}{r^5}$$

## Muons in matter

### The negative muon $\mu^-$

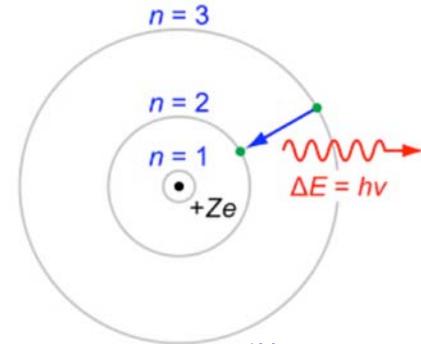
$\mu^-$  behaves as a **heavy electron** and is easily captured into the atomic orbitals.

Because it is "heavy", the  $\mu^-$  quickly cascades to the atomic 1s ground state in close proximity to host nuclei, producing **muonic X-rays**.

The significant overlap between muon and nuclear wave functions can result in *nuclear capture*. The measured mean lifetime of  $\mu^-$  in matter is considerably shorter than that of  $\mu^+$  due to capture by nuclei.

Because of reduced polarization and shorter lifetime,  $\mu^-$  SR is used only in selected cases for solid state studies.

Negative muons are, however, of great interest in nuclear physics



$$r_n(\mu) = n^2 \frac{m_e}{m_\mu} a_0$$

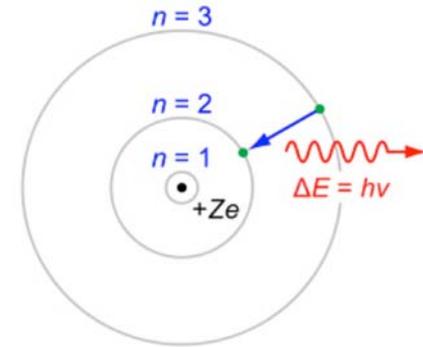
$$a_0 = 0.5 \times 10^{-10} m$$

$$r_1(\mu) \approx 10^{-12} m$$

$\mu^+$ in anything $\sim 2 \mu s$
$\mu^-$ in C: $\sim 2 \mu s$
$\mu^-$ in Pb: $\sim 0.07 \mu s$
$R(\text{Pb}) \sim 0.7 \cdot 10^{-12} m$

# X-rays of muonic atoms

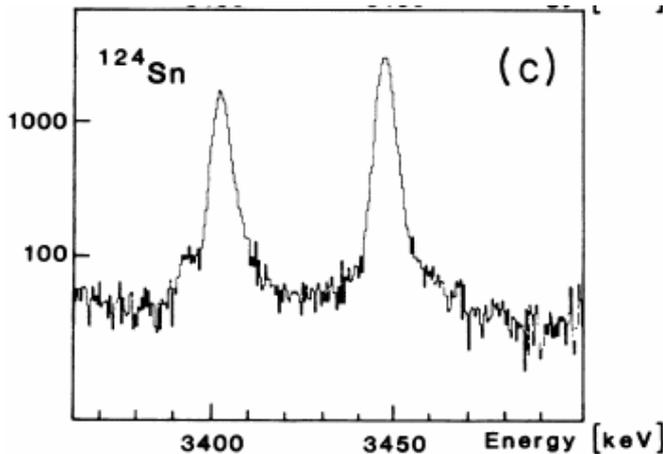
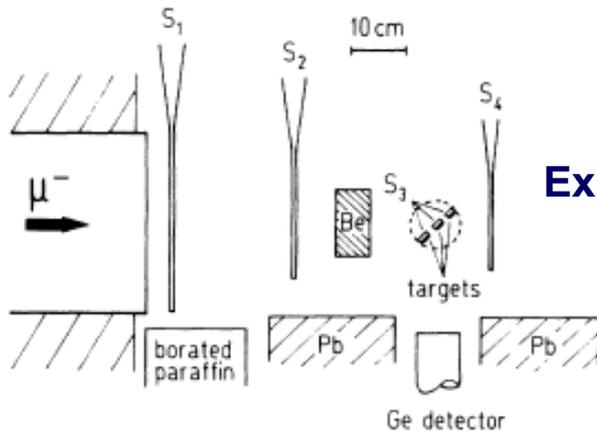
Muonic X-rays provide valuable information on nuclear charge distributions and nuclear deformation parameters: Quadrupole splitting of muonic X-rays yield **absolute values of nuclear quadrupole moments**



$$E_n(\mu) \propto \frac{m_\mu}{m_e} \frac{1}{n^2}$$

$$r_n(\mu) = n^2 \frac{m_e}{m_\mu} a_0$$

## Experimental set-up



Prompt muonic x-ray spectra showing the  $2p_{1/2}-1s$  and the  $2p_{3/2}-1s$  transitions in  $^{124}\text{Sn}$

# Muons in matter

## The positive muon $\mu^+$

In crystalline solids the  $\mu^+$  is *repelled* by the charge of the host nuclei but in a molecular gas or liquid  $\mu^+$  is *attracted* to the electron cloud around the host molecules.

In a solid  $\mu^+$  generally comes to rest at an **interstitial site** of high symmetry between the lattice ions, where it exists in a diamagnetic state as a "quasi-free" probe.

However, in oxides the  $\mu^+$  may localize near an oxygen atom, forming a  $\mu$ -O bond similar to an OH "hydrogen" bond.

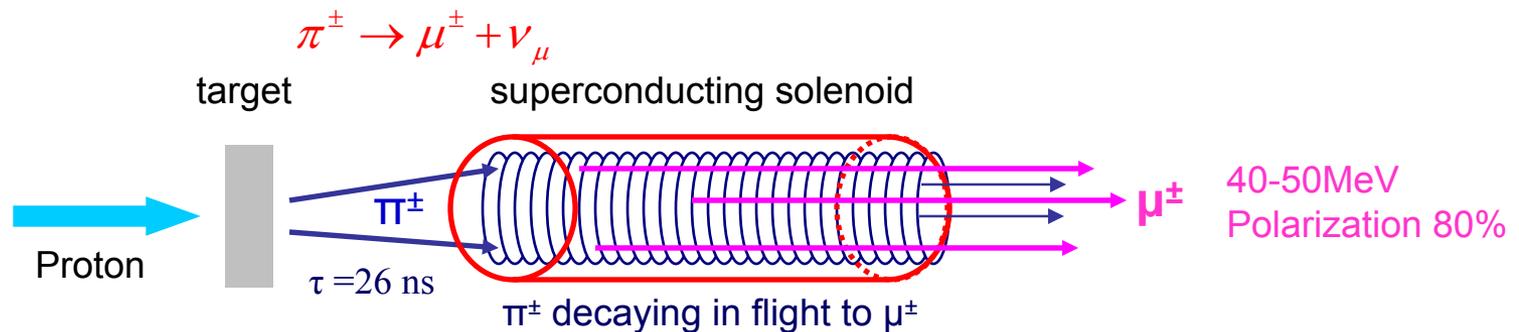
## ***Muonium*** ( $\text{Mu} \equiv \mu^+e^-$ )

In certain materials (e.g. semiconductors) a  $\mu^+$  can pick up an electron to form muonium which has almost the same Bohr radius and ionization potential, but a mass 9 times smaller than hydrogen ( $\text{H} \equiv p^+e^-$ ).

# Different types of muon beams

## I. High-energy beams (available at PSI, TRIUMF, J-PARC and RIKEN-RAL)

Pions escaping the production target at high energies are collected over a certain solid angle and directed on to a decay section consisting of a long superconducting solenoid with a field of several Tesla. If the pion momentum is not too high, a large fraction of the pions will have decayed before they reach the end of the solenoid.



**Advantage:** Homogeneous implantation of the muons into **large sample volumes**. Such beams are also used to study specimens inside of recipients, e.g. samples inside pressure cells.

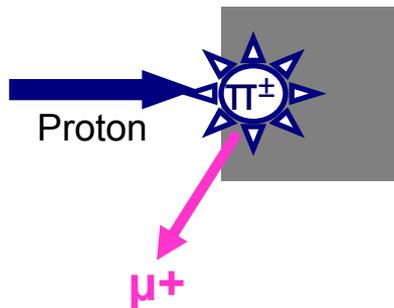
# Different types of muon beams

## II. Surface (or Arizona) $\mu^+$ beams

(available at PSI, TRIUMF, J-PARC, ISIS and RIKEN-RAL).

### Used for the investigation of relatively thin samples

Muons arising from pions slowing down in the production target and decaying **at rest** near the target surface



Ideally monochromatic muons,

- 100 % polarized
- kinetic energy of 4.1 MeV  $\rightarrow$
- small range in matter of  $\sim 180$  mg/cm<sup>2</sup>.

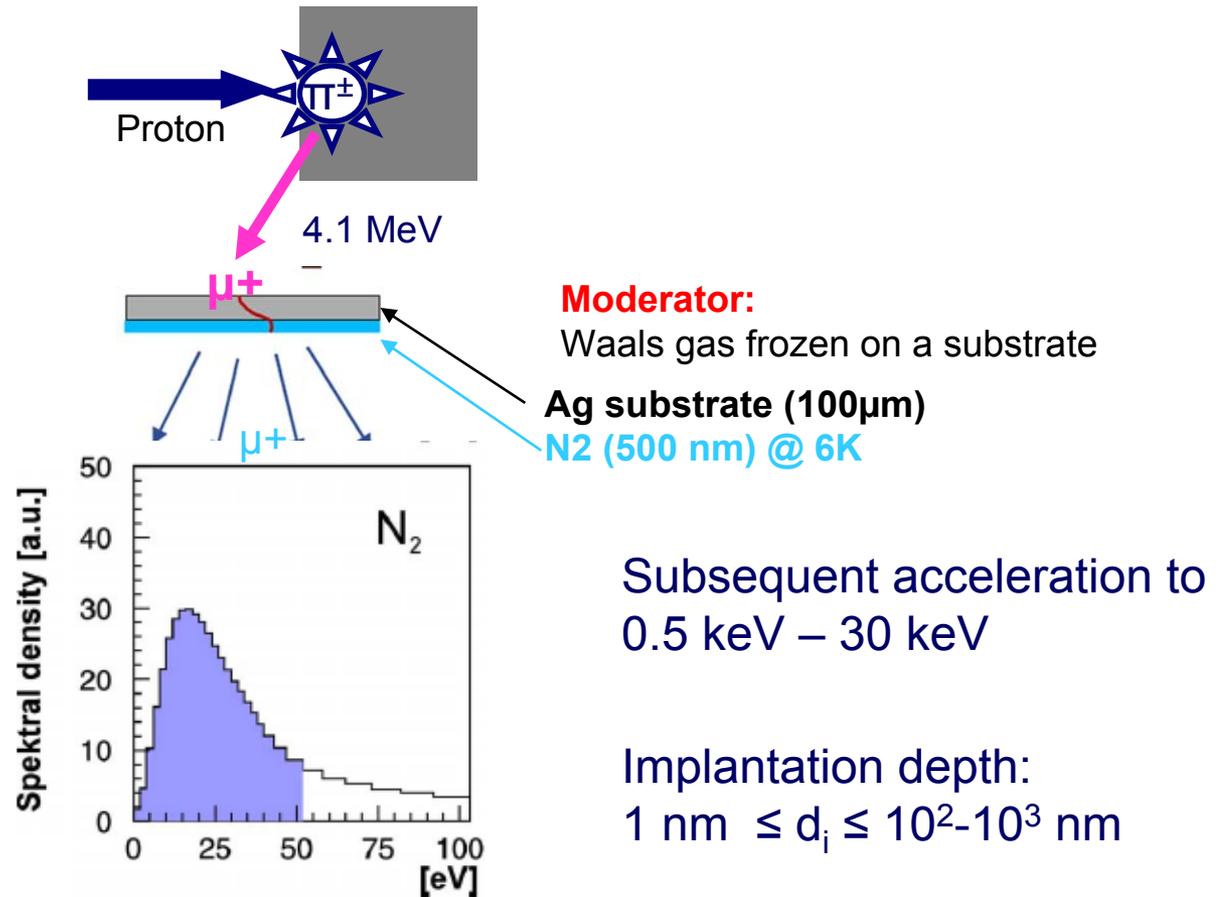
**Works only for positive pions**, because a negative pion stopping in the production target almost always undergoes nuclear capture from low-lying orbitals of pionic atoms (replacing an electron) before it has a chance to decay.

# Different types of muon beams

## III. Low-energy muon beams Surface

(available at PSI, projected at J-PARC, and RIKEN-RAL).

### Moderation of Arizona beams



## The mechanism of muon moderation

$4 \text{ MeV} \leq E_\mu \leq 10\text{-}50 \text{ keV}$ : energy loss by ionization processes

$E_\mu \leq 10 \text{ keV}$ : energy loss by charge-exchange processes

$E_\mu < 50 \text{ eV}$

Solid Ar, N<sub>2</sub>, ... are broad band isolators →

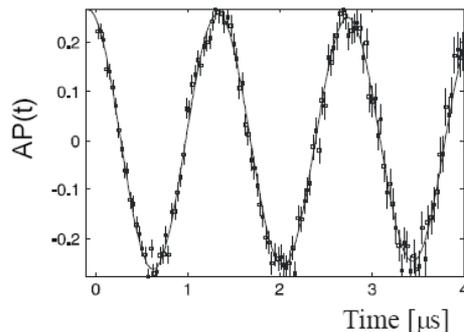
Suppression of electronic energy loss for  $E_\mu \approx E_{\text{gap}}$   
 no efficient inelastic energy loss channels available →

Epithermal muons can move through the solid  
 without efficient energy loss → **escape depth of up to 100 nm.**

**Moderation efficiency:**  $N_{\text{epi}}/N_{4\text{MeV}} \approx 10^{-4} - 10^{-5}$

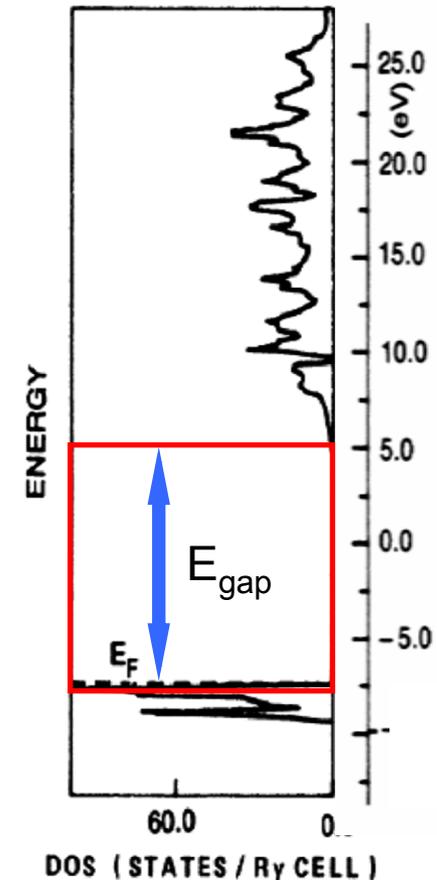
### IMPORTANT:

μSR of epithermal μ<sup>+</sup>



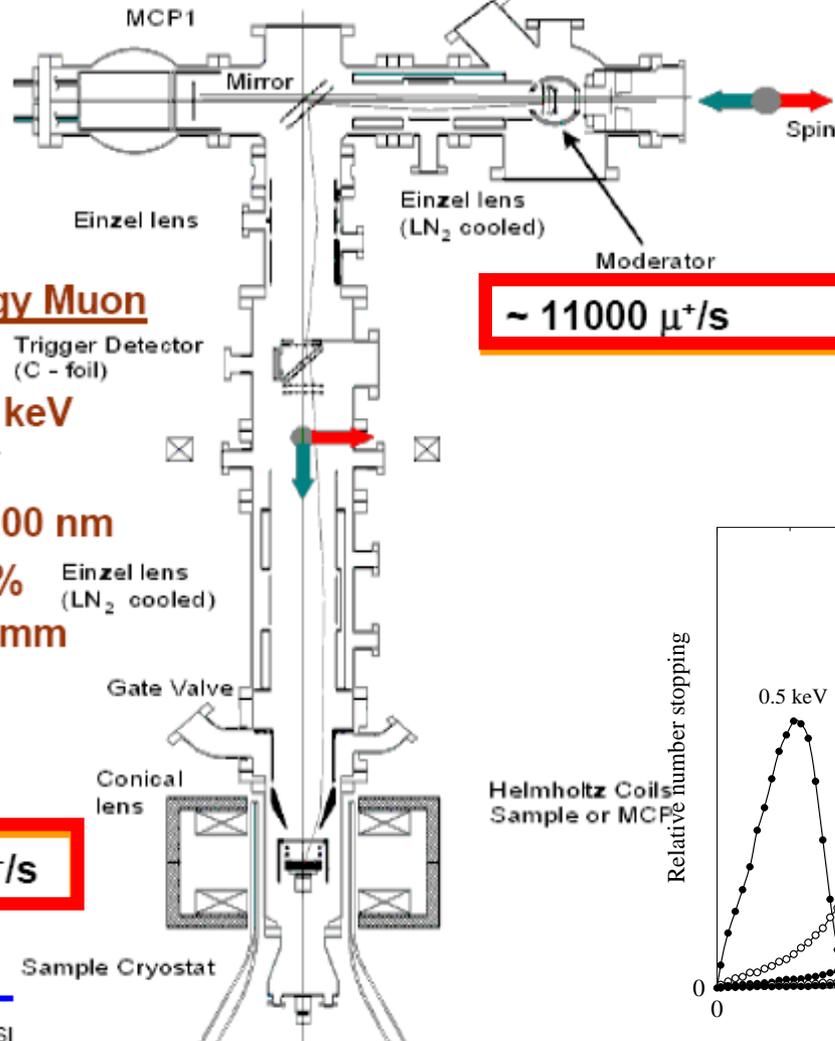
The polarization survives moderation

Density of states of solid Ar



# Low energy $\mu^+$ beam at Paul Scherrer Institute

Surface  $\mu$  beam  
E ~ 4 MeV



$\sim 1.9 \cdot 10^8 \mu^+/s$

from new  $\mu$ E4 beam line

$\sim 11000 \mu^+/s$

- UHV system ( $\sim 10^{-10}$  mbar)

- Electrostatic transport, focussing and energy selection.

- All transport elements LN<sub>2</sub> cooled

## Polarized Low Energy Muon

### Beam

Energy: 0.5-30 keV

$\Delta E$ : 400 eV

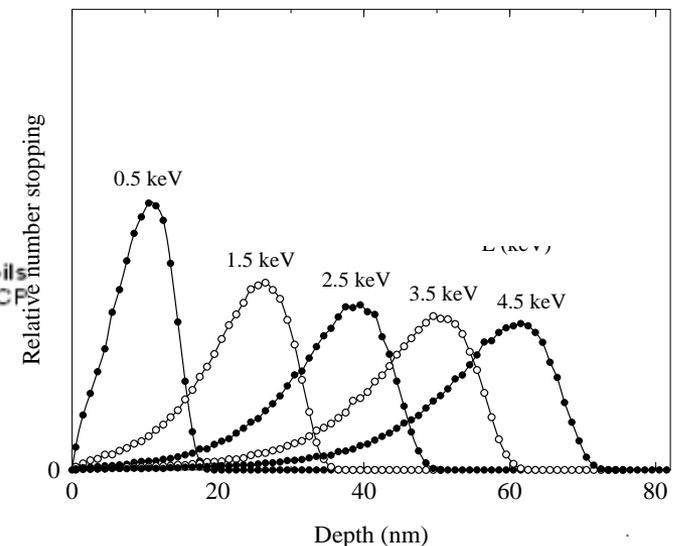
Depth:  $\sim 1 - 200$  nm

Polarization  $\sim 100\%$

Beam Spot: 10-20 mm

$\sim 4500 \mu^+/s$

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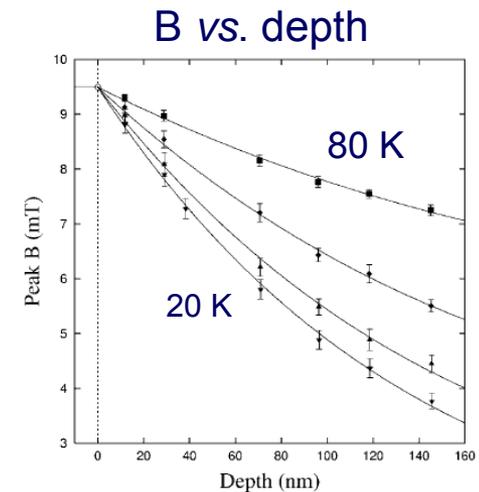
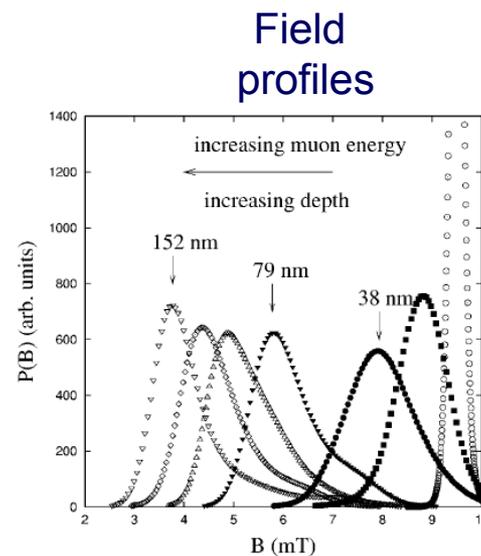
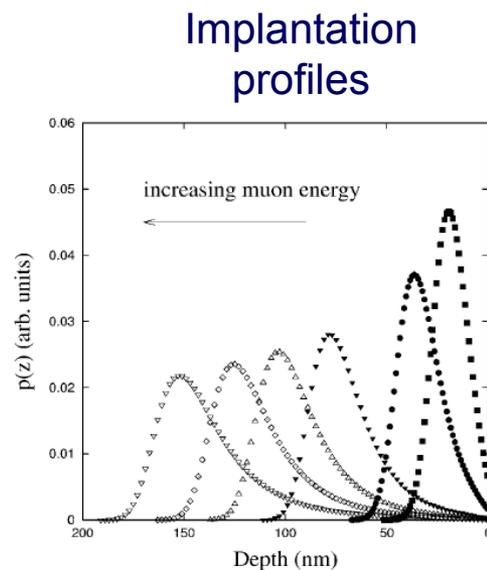


Tunable energy range : 0.5 -30 keV; Implantation depth:  $1 \text{ nm} \leq d_i \leq 10^2\text{-}10^3 \text{ nm}$

# Profile of the Magnetic Field beneath the Surface of a High-Tc Superconductor with a few nm Resolution

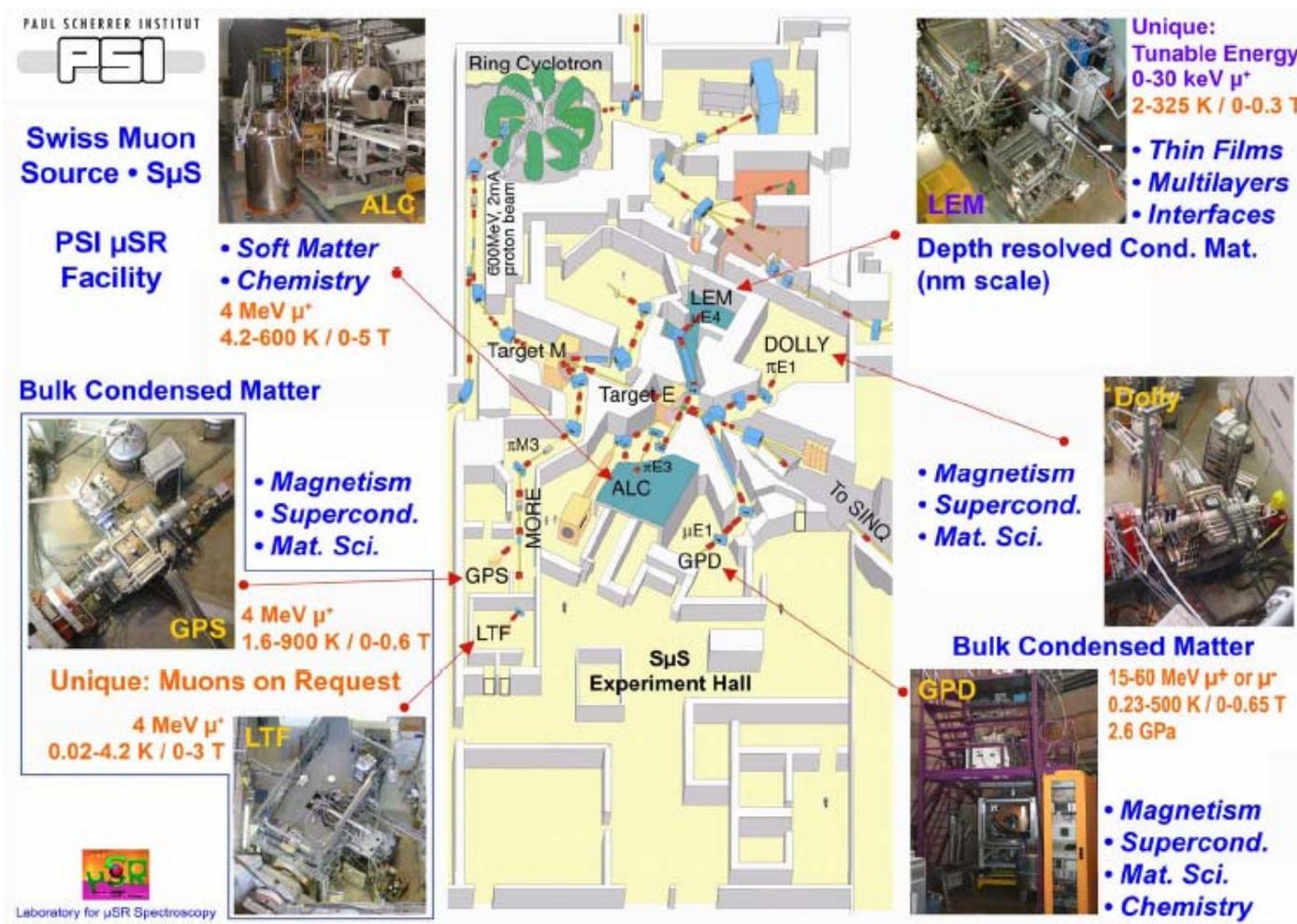
Thin film of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO) at  $T < T_C$  ( $\sim 90$  K)

Muons were implanted at depths from 20–150 nm by varying the energy of the incident muons from 3–30 keV

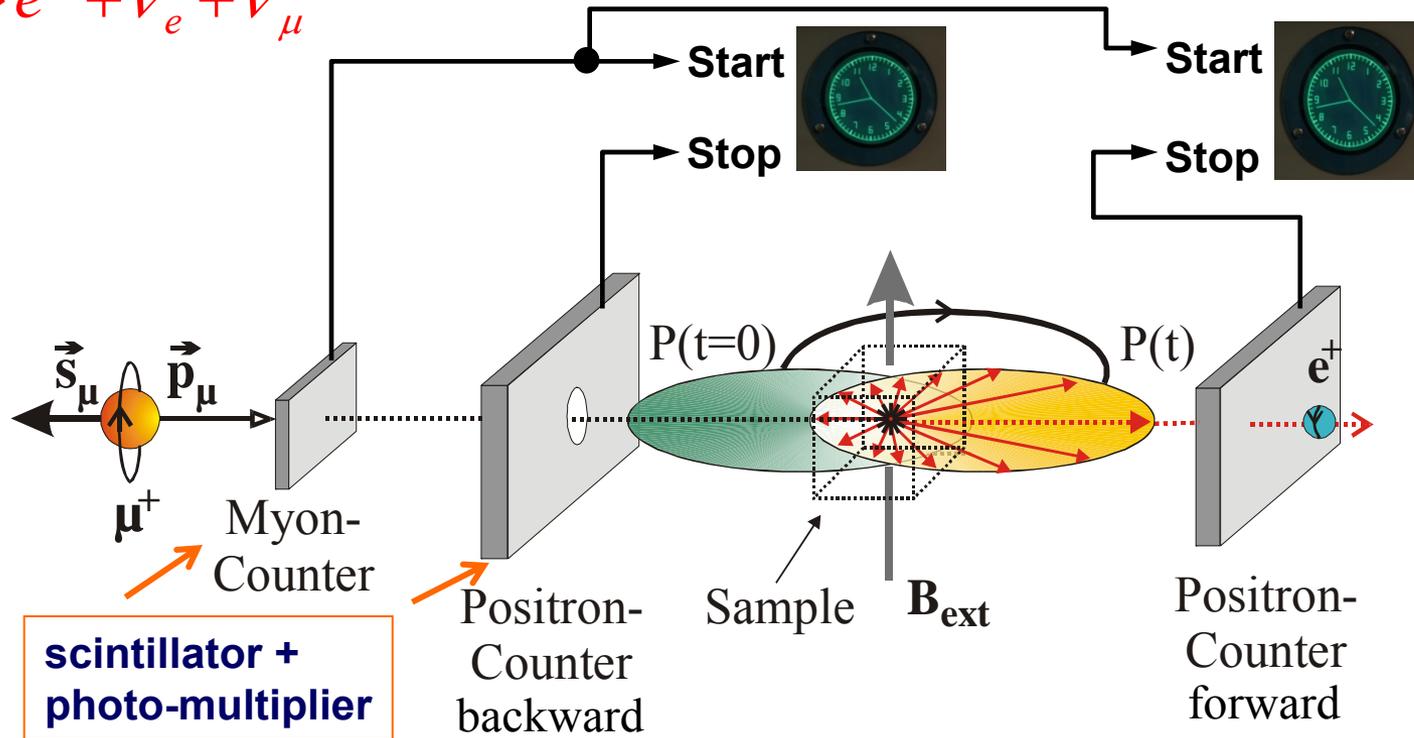


# Muon beams at S $\mu$ S – Swiss Muon Source

- **High energy beam (15-60 MeV  $\mu^+$  or  $\mu^-$ )** for bulk matter studies
- **Surface (Arizona) beam (4 MeV  $\mu^+$ )** – Magnetism, superconductivity, soft matter, chemistry in relatively thin samples
- **Low-energy muon beam (0-30 keV  $\mu^+$ )** – Thin films, multilayers, interfaces on a nm scale



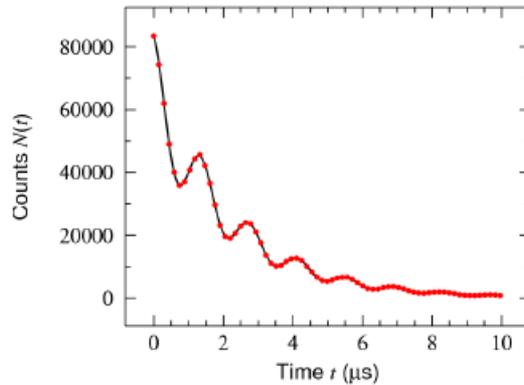
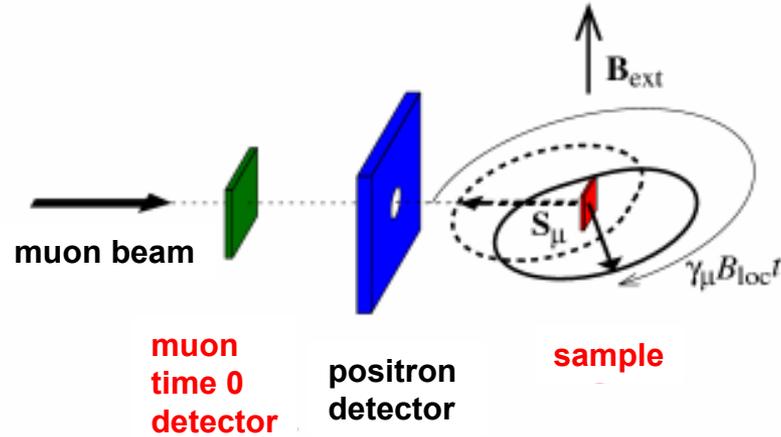
# Principle of a Muon-Spin-Rotation ( $\mu$ SR) experiment



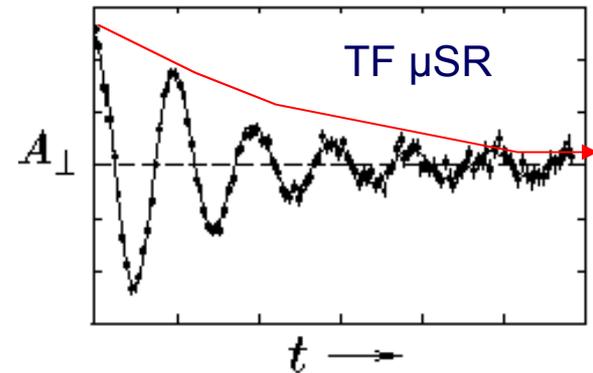
## 3 possibilities :

- TF  $\mu$ SR (transverse field) – Knight shift measurements, penetration depth (superconductivity)
- LF  $\mu$ SR (longitudinal field) – spin dynamics (different time scales than neutron scattering)
- ZF  $\mu$ SR (zero field) – weak magnetism, search for magnetic ordering

# Transverse field (TF) $\mu$ SR geometry



$$A(t) = \frac{N_\uparrow - N_\downarrow}{N_\uparrow + N_\downarrow}$$



Damping due to

- static field distribution
- dynamic interaction

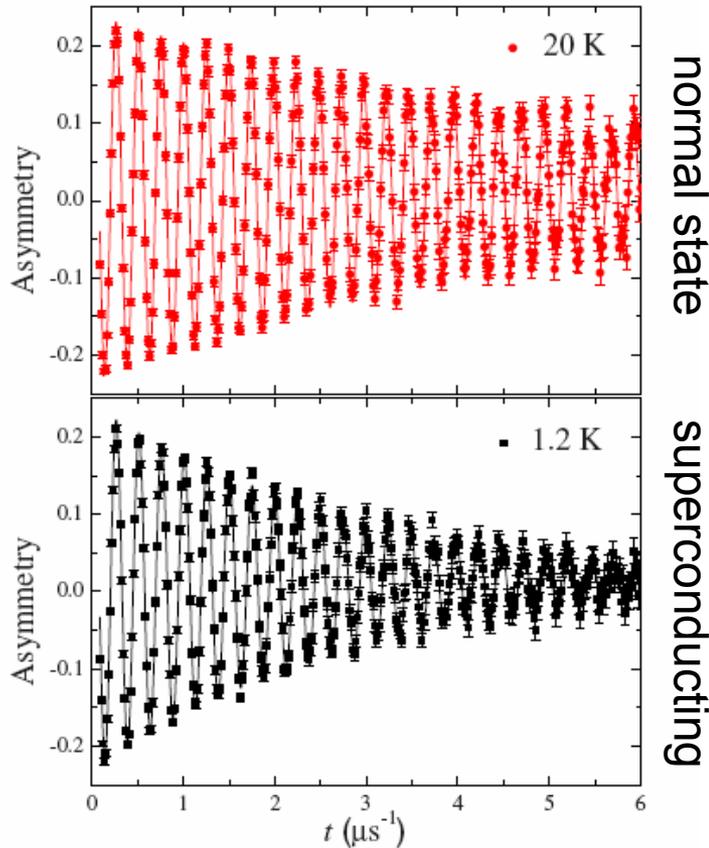
Larmor precession

$$A_0 P(t) = A_0 \left[ \frac{1}{3} e^{-\lambda_L t} + \frac{2}{3} e^{-\lambda_T t} \cos(\gamma_\mu B t) \right]$$

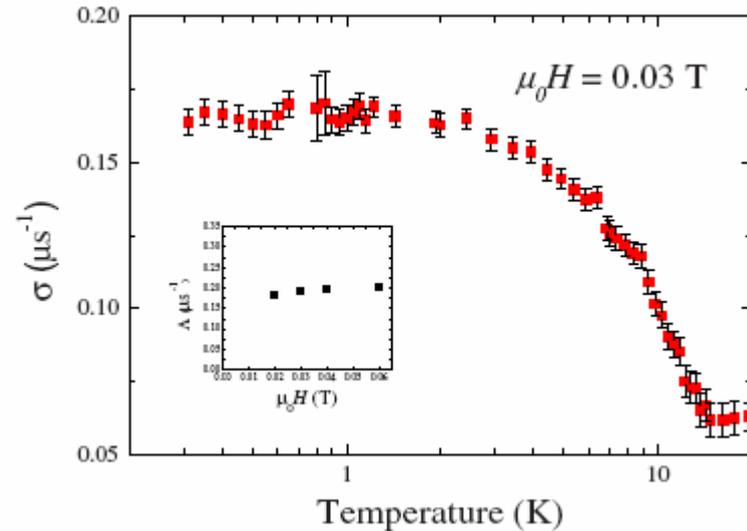
# Muon-spin-spectroscopy study of superconducting $\text{FeTe}_{0.5}\text{Se}_{0.5}$ ( $T_c = 14.4$ K)

P. K. Biswas et al., PRB 81, 092510 (2010)

TF- $\mu$ SR spectra

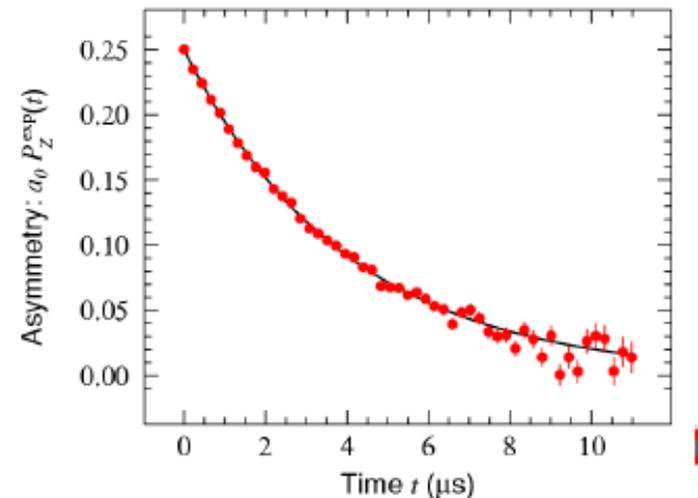
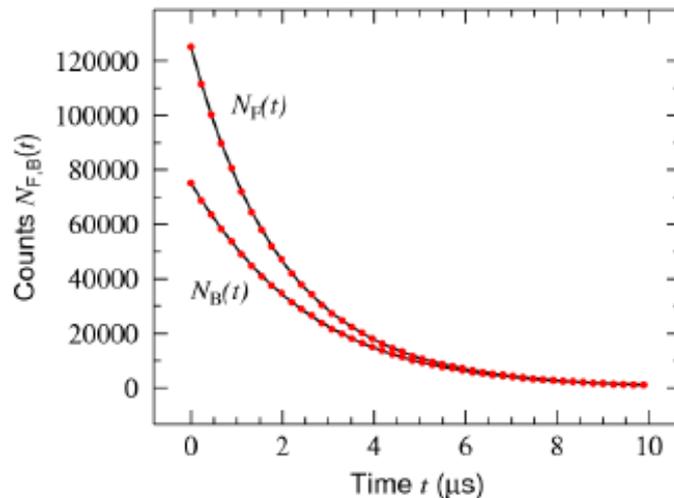
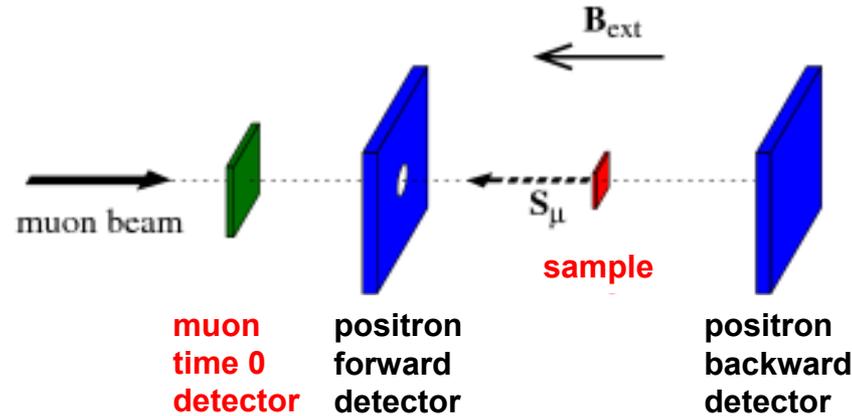


Gaussian depolarization rate



The relaxation rate increases below  $T_c$  due to the **inhomogeneous field distribution** of the flux-line lattice  $\rightarrow$   
 Information on the London penetration depth

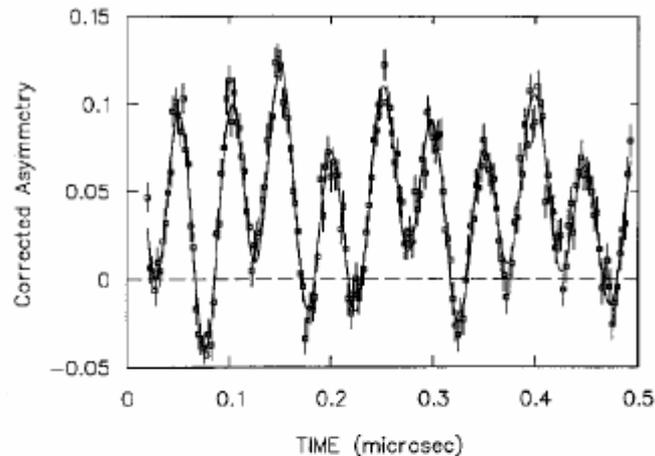
# Zero and longitudinal field $\mu$ SR geometry



ZF- $\mu$ SR is a very sensitive method of detecting **weak internal magnetism**, that arises due to ordered magnetic moments, or **random fields that are static or fluctuating with time**.

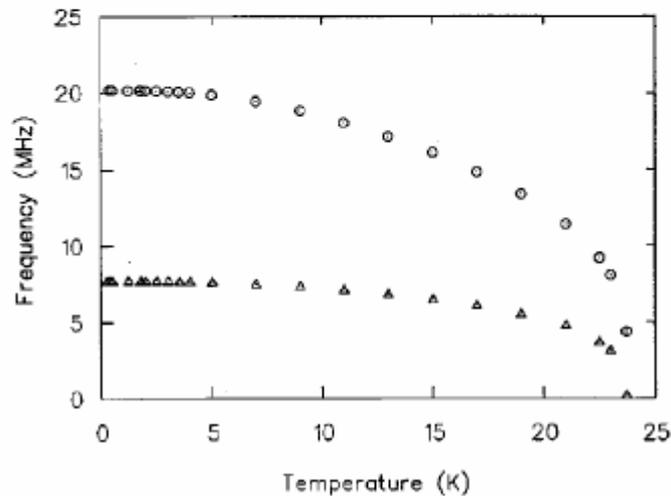
ZF- $\mu$ SR provided the first evidence for static magnetic order in the undoped parent compound  $\text{La}_2\text{CuO}_4$ . Since then  $\mu$ SR has played a major role in determining magnetic phase diagrams, particularly in high- $T_c$  and heavy-fermion systems.

## An example of ZF $\mu$ SR spectra: $\mu^+$ in antiferromagnetic $\text{CaV}_3\text{O}_7$



Two precession signals corresponding to two different muon sites

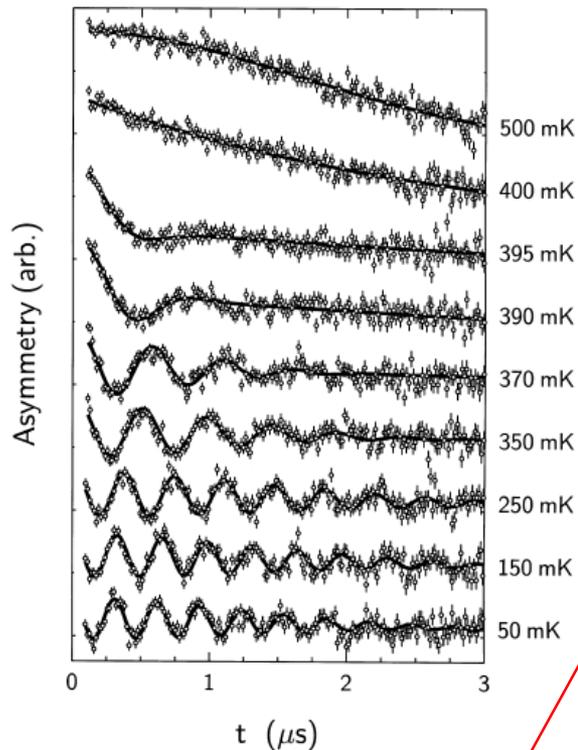
The local field is a consequence of the AF order of the V moments.



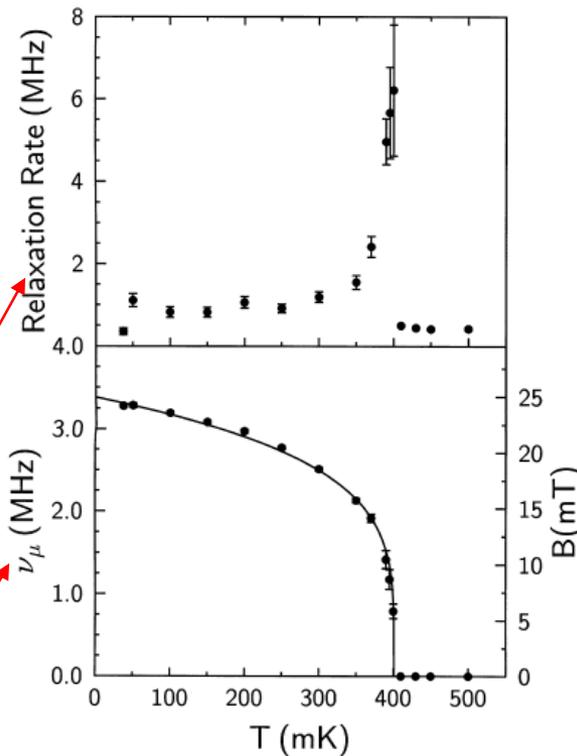
The magnetization curves

# ZF- $\mu$ SR and ordered organic ferromagnets and antiferromagnets

Antiferromagnetic Tanol suberate ( $C_{13}H_{23}O_2NO$ )<sub>2</sub>



Raw  $\mu$ SR data measured on tanol suberate in zero magnetic field.



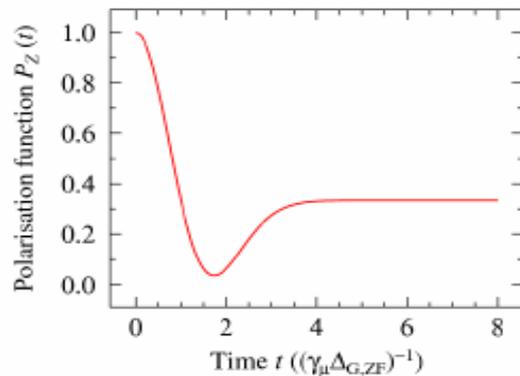
Temperature dependence of the relaxation rate and frequency of the muon-spin precession.

$$P(t) = a_L(t) + a_T e^{-\lambda t} \cos(2\pi\nu_\mu t)$$

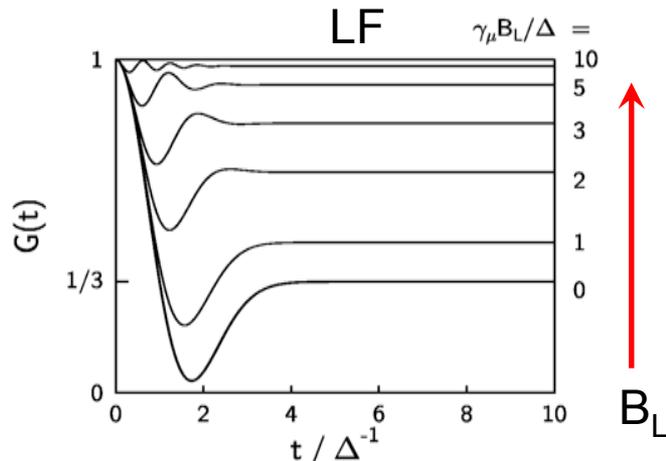
# Spectra of static field distributions in ZF- and LF- geometry

Kubo Toyabe functions for static Gaussian field distribution - width  $\Delta$

ZF

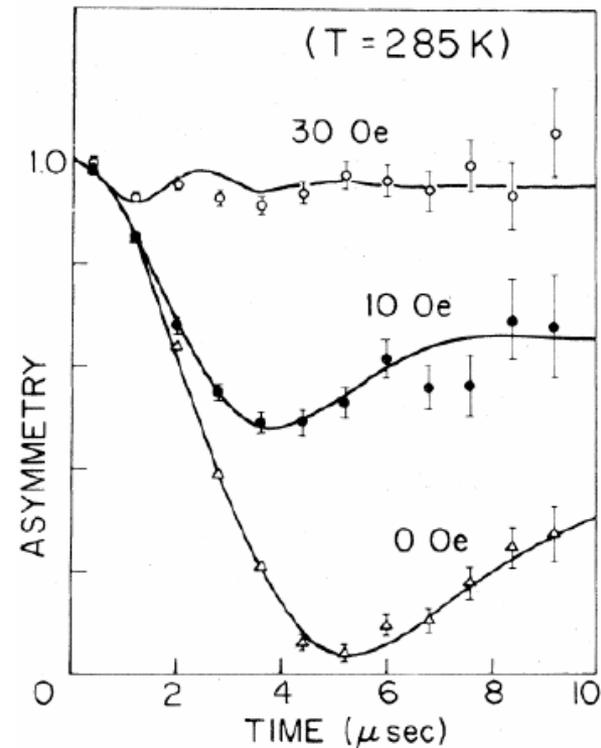


LF



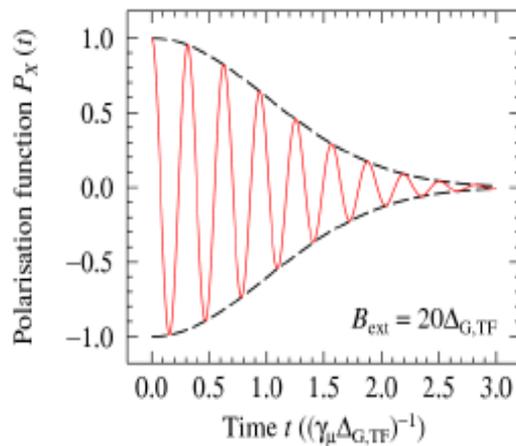
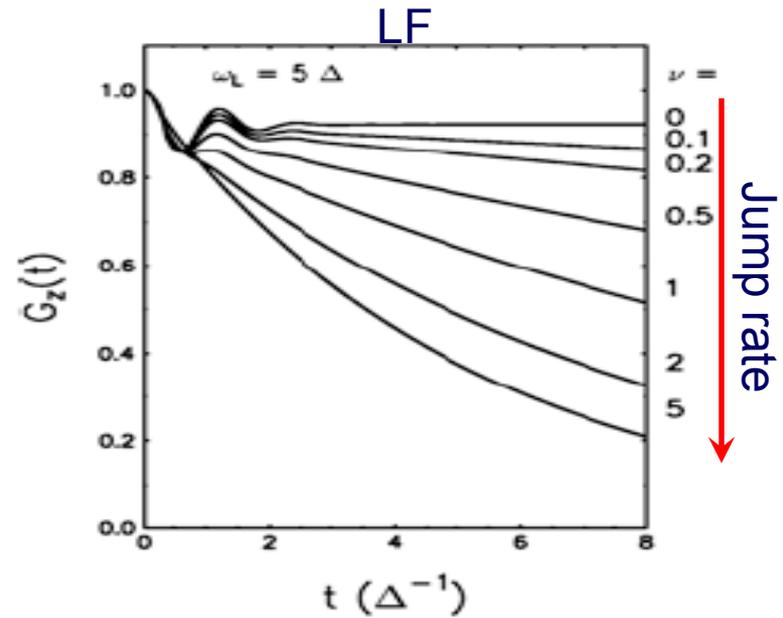
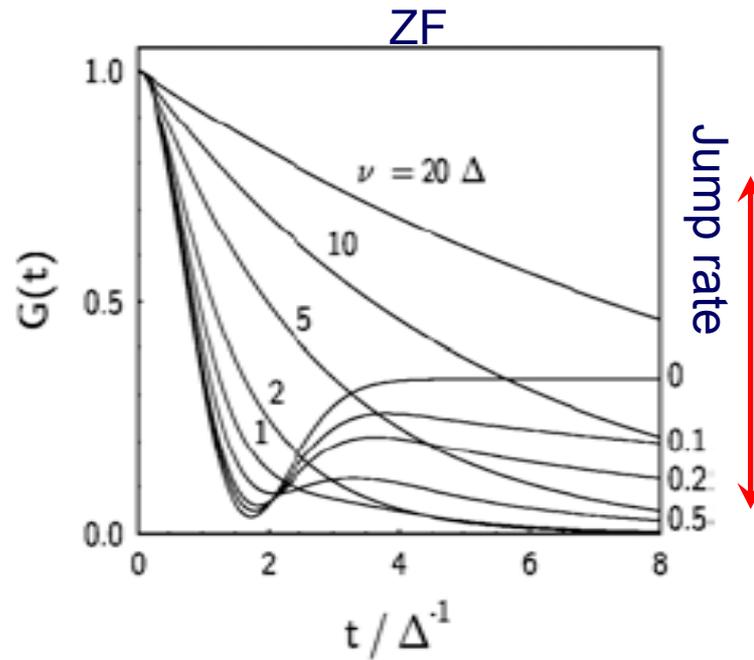
The longitudinal field decouples the spins from the field distribution

An example: Muon spin relaxation and longitudinal field-decoupling in the paramagnetic phase of MnSi (R.S. Hayano et al., Phys. Rev **B20**, 850 (1979)).

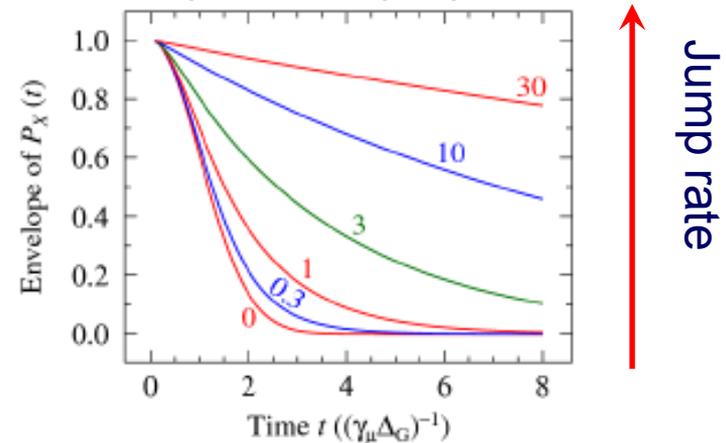


In this case the local field is produced mainly by the Mn nuclear moments.

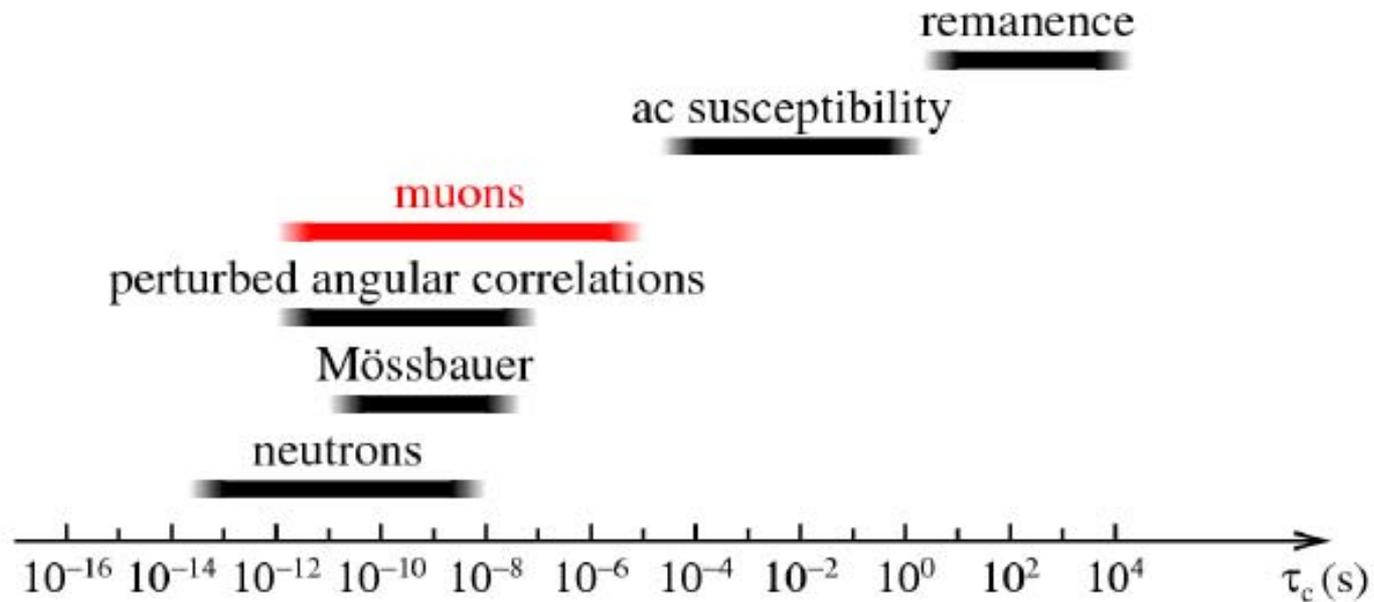
# The effect of dynamics in ZF, LF -and TF-geometry



## Envelope of TF spin precession



# The dynamical ranges of different techniques



# Areas of $\mu$ SR applications

## **Magnetic Systems**

*Magnetically Ordered Systems*  
*Spin-Glass Systems*  
*Frustrated Spin Systems*  
*Colossal Magnetoresistance*  
*Low-Dimensional Systems*  
*Heavy Fermion Systems*  
*Quasicrystals*  
*Molecular Magnets & Clusters*

## **Superconductors**

*Magnetic Phases & Phase Separation*  
*Weak Magnetism*  
*Characteristic Length Scales*  
*Pairing Properties*

## **Transport**

*Quantum Diffusion*  
*Electron Transport in Non-Metals*  
*Conducting Polymers*

## **Semiconductors**

## **Chemistry**

*Chemical Reaction Kinetics and H Isotopes*  
*Free Radical Systems*

## **Biological Applications**