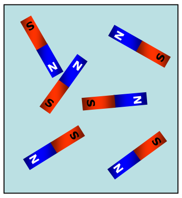
Muon Spin Rotation / Relaxation on Magnetic Materials

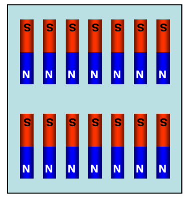
Magnetism

Magnetism

Paramagnetism | **Ferromagnetism**



fluctuating
$T > T_C$

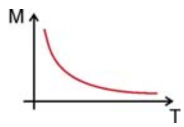


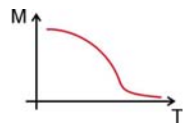
static
$T < T_C$

The interesting property of magnetically ordered system is the size and temperature dependence of the magnetic moment (order parameter).

How do you measure this?

Macroscopic techniques (average over the whole sample):

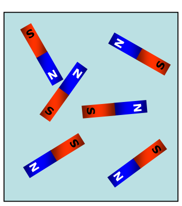




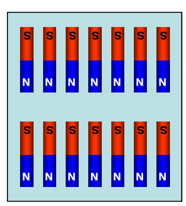
SQUID, PPMS, …

Magnetism

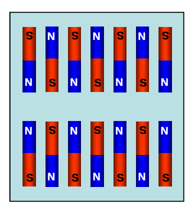
Paramagnetism | **Ferromagnetism** | **Antiferromagnetism**



fluctuating
$T > T_C$



static
$T < T_C$

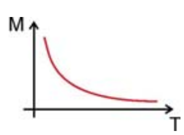


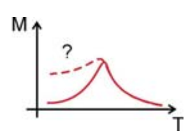
static
$T < T_N$

The interesting property of magnetically ordered system is the size and temperature dependence of the magnetic moment (order parameter).

How do you measure this?

Macroscopic techniques (average over the whole sample):





Magnetism

Scattering techniques:
(neutrons, X-rays)

Local probes:
(µSR, NMR, …)

Strength of muon spin rotation / relaxation:

- study of very weak magnetism
- Investigation of magnetically inhomogeneous materials:
  - Chemical inhomogeneity (“dirty samples”)
  - **Competing interactions** (interesting!)

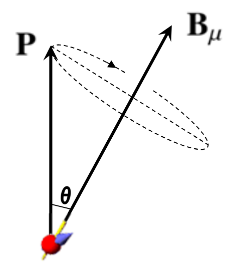
Alex Amato Masterschool 2017

Relation
µSR frequency ↔ field

Alex Amato Masterschool 2017

Muon Spin Precession – Larmor frequency

$$\mathbf{P} = \frac{\langle \mathbf{S} \rangle}{\frac{\hbar}{2}}$$

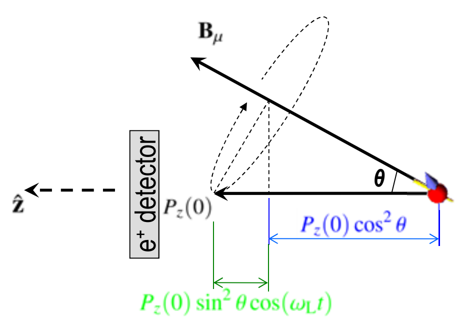


Larmor precessions with angular velocity: $\omega_\mathrm{L} = \gamma_\mu B_\mu$

with $\gamma_\mu = \frac{e}{2m_\mu} g_\mu = 8.51615 \times 10^8$ rad/sT

Frequency: $\frac{\gamma_\mu}{2\pi} = 135.539$ MHz/T

Field Direction -- Field Distribution



$$P_z(t) = \int f(\mathbf{B}_\mu) \left[\cos^2\theta + \sin^2\theta \cos(\gamma_\mu B_\mu t) \right] d\mathbf{B}_\mu$$

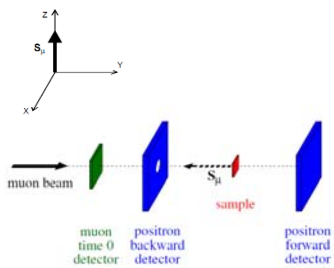
- Static part
- Oscillating part

θ angle between magnetic field and muon polarization at $t = 0$

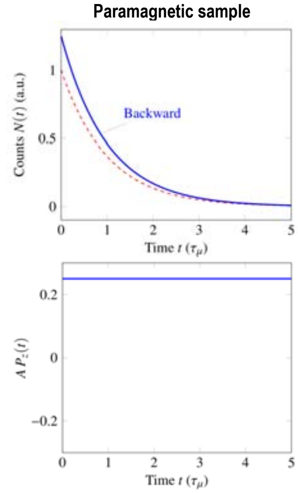
PAUL SCHERRER INSTITUT

Typical Measurement Geometry

ZF: Zero field geometry



Paramagnetic sample

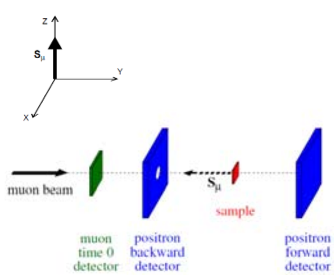


Alex Amato Masterschool 2017

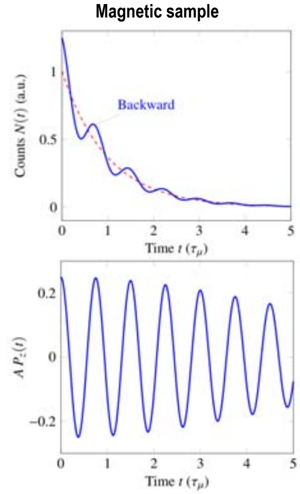
PAUL SCHERRER INSTITUT

Typical Measurement Geometry

ZF: Zero field geometry



Magnetic sample

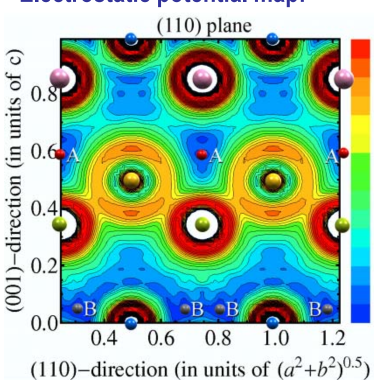


Alex Amato Masterschool 2017

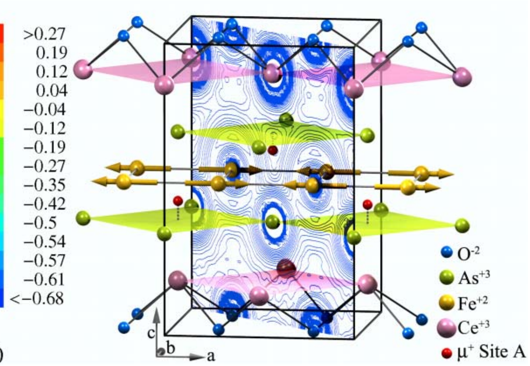
The field at the muon site

Interstitial Muon Stopping Sites

Electrostatic potential map:



CeFeAsO structure:



Positive muon likes to stop:

- In the potential minimum
- High symmetry sites
- Large spaces in the crystal structure

Experimental determination:

► *A. Amato et al.,*
Phys. Rev. B 89, 184225 (2014).

Theoretical predictions (based on DFT + μ):

► *J. S. Moeller et al., Phys. Scr. 88 068510 (2013)*
F. Bernardini et al., Phys. Rev. B 87, 115148 (2013)
P. Bonfà et al., J. Phys. Chem. C 119, 4278 (2015)

Field at the muon site

Internal field at the muon site:

$$\mathbf{B}_\mu = \mathbf{B}_c + \mathbf{B}_{\text{dip}}$$

- Contact field $\propto e|\Psi(\mathbf{r}_\mu)|^2$
- Dipolar contribution

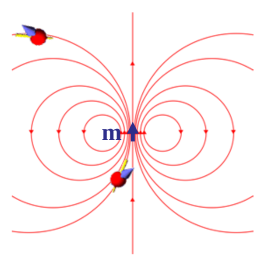
$$\mathbf{B}_{\text{dip}} = \sum_i \frac{1}{r_i^3}\left[\frac{(3\mathbf{m}_i \cdot \mathbf{r}_i)}{r_i^2}\mathbf{r}_i - \mathbf{m}_i\right]$$

Field at the muon site

- Contact field $\propto e|\Psi(\mathbf{r}_\mu)|^2$
- Dipolar contribution

$$\mathbf{B}_{\text{dip}} = \sum_i \frac{1}{r_i^3}\left[\frac{(3\mathbf{m}_i \cdot \mathbf{r}_i)}{r_i^2}\mathbf{r}_i - \mathbf{m}_i\right]$$

Both dependent on the muon site.

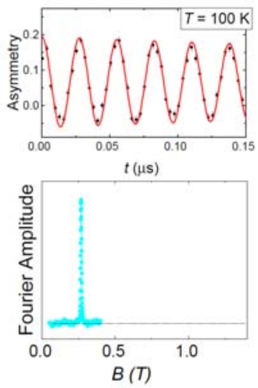


- **One µSR spontaneous frequency**:
  → Only one type of muon stopping site
  → All the crystallographically equivalent stopping sites are also magnetically equivalent
- **More than one µSR spontaneous frequencies**:
  → More than one type of muon stopping site
  → And/or the crystallographically equivalent stopping sites are magnetically inequivalent

Examples: Field at the muon site

MnP

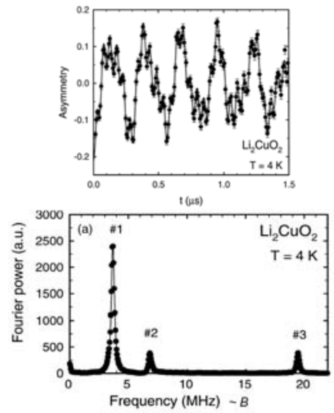
Ferromagnet
One muon stopping site



R. Khasanov et al.,
Phys. Rev. B ***93****, 180509(R) (2016)*

Li_2CuO_2

Antiferromagnet
3 muon stopping sites



U. Staub, B. Roessli and A. Amato,
Physica B ***289-290****, 299 (2000)*

But in both cases:

The observed frequencies scale
with the value of the ordered moment (ordered parameter):

$$\nu_\mu = \frac{\omega_L}{2\pi} = \frac{\gamma_\mu}{2\pi} B_\mu \propto m_{ord}$$

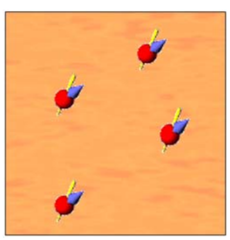
Advantages of the µSR technique to investigate magnets

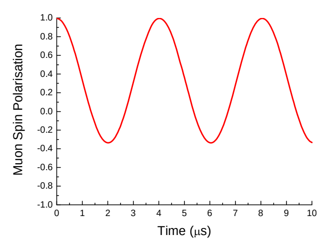
Alex Amato — Masterschool 2017

Advantage: Test the Magnetic Homogeneity

Homogen:

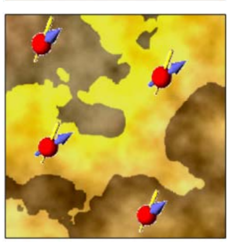
M_{hom}

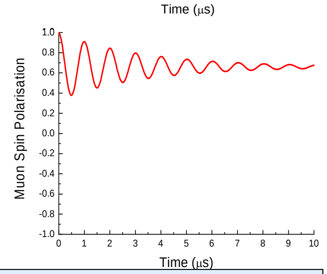




Inhomogeneous:

$M_{inhom} = M_{hom}$





Frequency	= Size of the magnetic moments (order parameter)
Damping	= Inhomogeneity within the magnetic areas
Amplitude	= Magnetic volume fraction

Advantage: Sensitivity of the Technique

Internal field at the muon site:

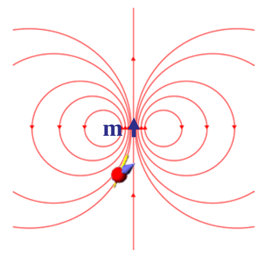
$$\mathbf{B}_\mu = \mathbf{B}_c + \mathbf{B}_{\rm dip}$$

- Contact field $\propto e|\Psi(\mathbf{r}_\mu)|^2$
- Dipolar contribution

$$\mathbf{B}_{\rm dip} = \sum_i \frac{1}{r_i^3}\left[\frac{(3\mathbf{m}_i \cdot \mathbf{r}_i)}{r_i^2}\mathbf{r}_i - \mathbf{m}_i\right]$$

$$B_{\rm dip} \simeq \frac{m}{r^3}$$

For $m = 1\,\mu_{\rm B}$ and $r = 1\,\text{Å} \Rightarrow\ B_{\rm dip} \simeq 1$ T



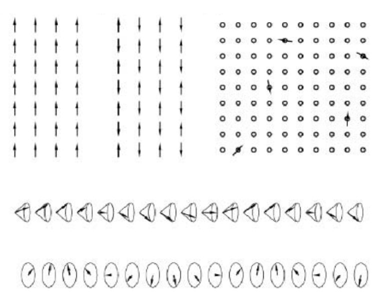
μSR time window: 10-20 μsec

➡ Frequencies down to 50 kHz detectable
Fields of few Gauss (10^{-4} T)

➡ *static moments as low as 0.001 μ_B can be detected by μSR*

Different Depolarization Functions

ferro antiferro dilute



helical

spiral

- nuclear moments
- electronic moments

- static moments
- fluctuating moments

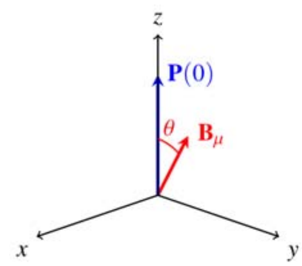
- various spin structures
- spin glasses (randomness)
- variety of field distributions

dedicated $P(t)$ for each problem

Magnetism of Single Crystals

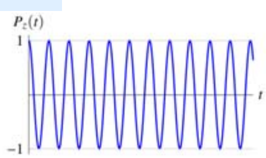
Simple Magnetic Sample – Single Crystal

$$P_z(t) = \int f(\mathbf{B}_\mu)\left[\cos^2\theta + \sin^2\theta\cos(\gamma_\mu B_\mu t)\right]d\mathbf{B}_\mu$$



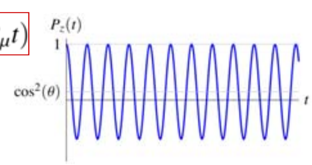
Single Crystal with $\theta = \pi/2$

$$P_z(t) = \cos(\gamma_\mu B_\mu t)$$



Single Crystal with $\theta \neq \pi/2$

$$P_z(t) = \cos^2\theta + \sin^2\theta\cos(\gamma_\mu B_\mu t)$$

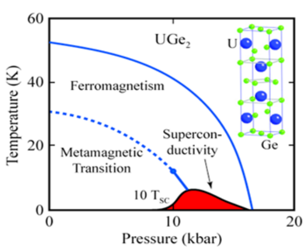


→ in a single crystal the amplitude of the oscillatory component indicates the direction of the internal field

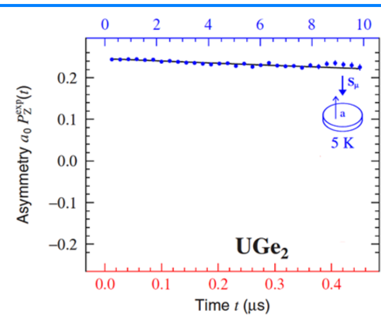
Examples:

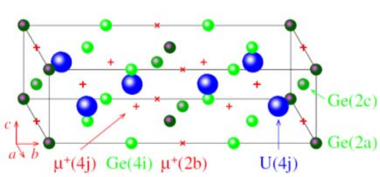
Magnetism on Single Crystal: UGe_2
Magnetism on Single Crystal: Li_2CuO_2

Example Single Crystal: UGe_2



Cavendish Lab.

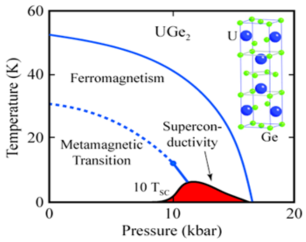




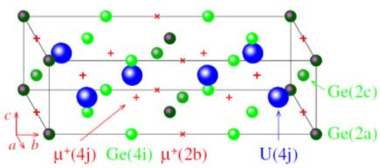
- Two magnetically inequivalent muon stopping sites
- No oscillations for $\mathbf{P}(0)$ pointing along *a*-axis
  → Internal fields parallel to *a*-axis (θ = 0)
- $\mathbf{P}(0)$ perp. to *a*-axis: oscillations around zero
  → because single crystal (θ = 90 deg.)

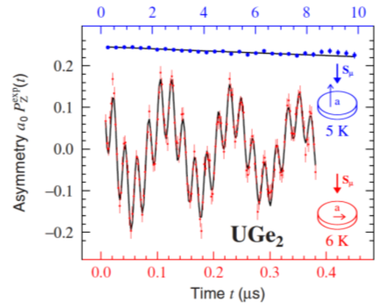
S. Sakarya et al., Phys. Rev. B 81, 024429 (2010)

Example Single Crystal: UGe_2



Cavendish Lab.





- Two magnetically inequivalent muon stopping sites
- No oscillations for $\mathbf{P}(0)$ pointing along *a*-axis
  → Internal fields parallel to *a*-axis (θ = 0)
- $\mathbf{P}(0)$ perp. to *a*-axis: oscillations around zero
  → because single crystal (θ = 90 deg.)

S. Sakarya et al., Phys. Rev. B 81, 024429 (2010)

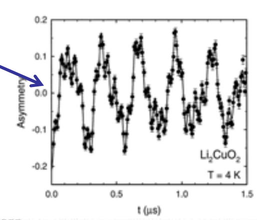
Example Single Crystal : Li_2CuO_2

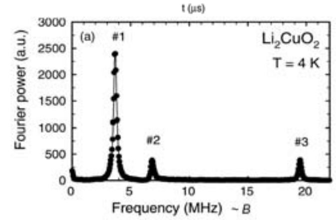
- oscillations around zero
  → because single crystal

Li_2CuO_2

Antiferromagnet

3 muon stopping sites





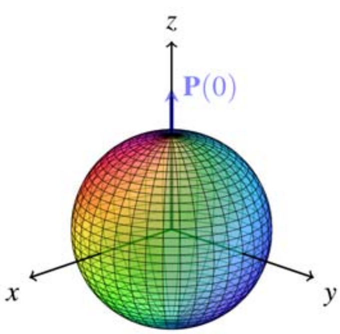
*U. Staub, B. Roessli and A. Amato, Physica B **289-290**, 299 (2000)*

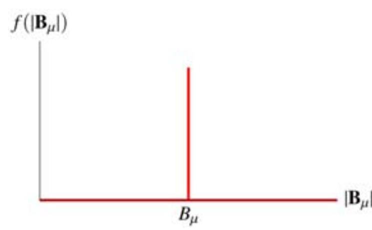
Magnetism of Polycrystals (Powders)

Simple Magnetic Sample – Polycrystal

$$P_z(t) = \int f(\mathbf{B}_\mu)\left[\cos^2\theta + \sin^2\theta\cos(\gamma_\mu B_\mu t)\right]d\mathbf{B}_\mu$$

Polycrystal (powder)





If isotropic:

$$f(|\mathbf{B}_\mu|) = f(\mathbf{B}_\mu)\,4\pi B_\mu^2$$

$$f(\mathbf{B}_\mu)\,d\mathbf{B}_\mu = \frac{f(|\mathbf{B}_\mu|)}{4\pi}\sin(\theta)\,d\theta\,d\phi\,dB_\mu$$

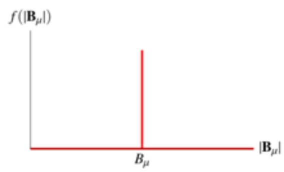
$$P_z(t) = \frac{1}{3} + \frac{2}{3}\cos(\gamma_\mu B_\mu t)$$

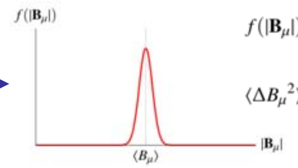
Simple Magnetic Sample – Polycrystal

$$P_z(t) = \int f(\mathbf{B}_\mu)\left[\cos^2\theta + \sin^2\theta\cos(\gamma_\mu B_\mu t)\right]d\mathbf{B}_\mu$$

Polycrystal: Ideal Case

Polycrystal: Real Case



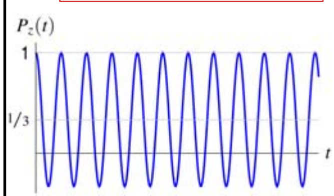


$$f(|\mathbf{B}_\mu|) = \frac{1}{\sqrt{2\pi\langle\Delta B_\mu^2\rangle}}\exp\left[-\frac{(B_\mu - \langle B_\mu\rangle)^2}{2\langle\Delta B_\mu{}^2\rangle}\right]$$

$$\langle\Delta B_\mu{}^2\rangle = \int (B_\mu - \langle B_\mu\rangle)^2 f(|\mathbf{B}_\mu|)dB_\mu$$

$$P_z(t) = \frac{1}{3} + \frac{2}{3}\cos(\gamma_\mu B_\mu t)$$

$$P_z(t) = \frac{1}{3} + \frac{2}{3}\exp\left[-\frac{1}{2}\gamma_\mu^2\langle\Delta B_\mu{}^2\rangle t^2\right]\cos\left[\gamma_\mu\langle B_\mu\rangle t\right]$$



Examples:

Magnetism of Polycrystalline LaOFeAs
Magnetism of MnP

Example Polycrystal: LaFeAsO

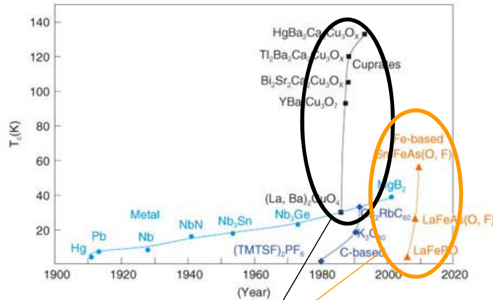
Cu-based superconductors

J. Georg Bednorz

K. Alexander Müller

Fe-based superconductors

Hideo Hosono

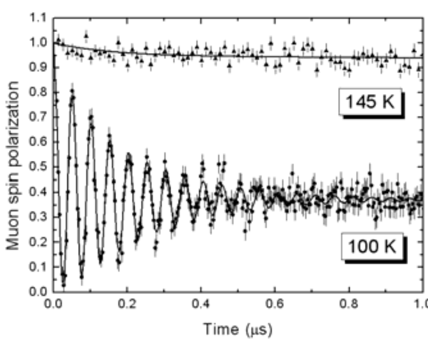


Study of the interplay
Magnetism/SC by µSR

From NTT Basic Research Lab.

Example Polycrystal: LaFeAsO

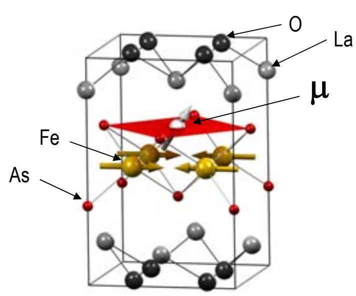
Muon spin rotation:



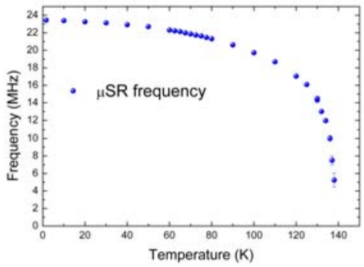
H.H. Klauss, et al., Phys. Rev. Lett. ***101****, 077005 (2008)*

Muon Spin Rotation

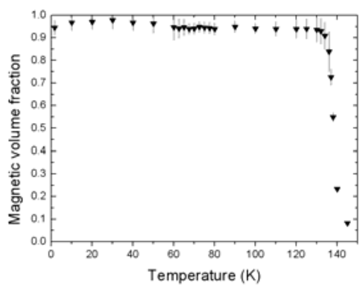
- Oscillations around 1/3
  → because polycrystal



Example Polycrystal: LaFeAsO

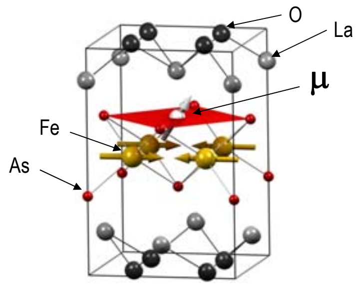


H.H. Klauss, et al., Phys. Rev. Lett. ***101****, 077005 (2008)*

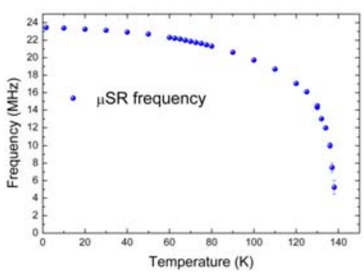


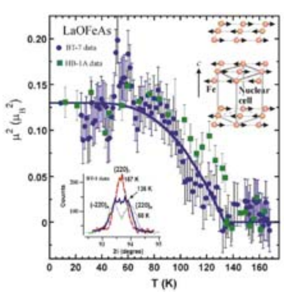
Muon Spin Rotation

- Oscillations around 1/3
  → because polycrystal
- T-dependence of the Fe magnetization with high precision, T_N = 138 K
- 100% of the sample volume is magnetic



Example Polycrystal: LaFeAsO

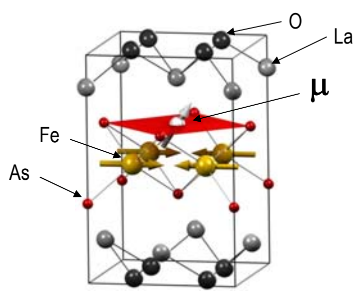




C. de la Cruz et al., Nature 453, 899 (2008)

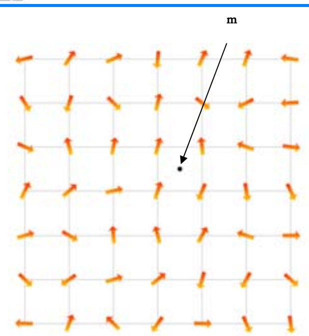
Muon Spin Rotation

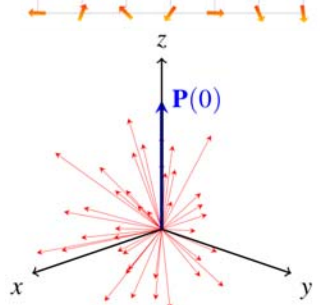
- Oscillations around 1/3
  → because polycrystal
- T-dependence of the Fe magnetization with high precision, T_N = 138 K
- 100% of the sample volume is magnetic

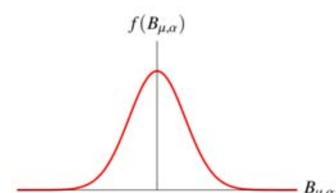


Randomly Oriented Magnetic Moments
-
Short Range Magnetism
-
Magnetic Disorder

Randomly Oriented Moments







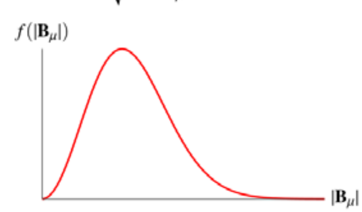
$$f(B_{\mu,\alpha}) = \frac{1}{\sqrt{2\pi\langle\Delta B_\mu^2\rangle}} \exp\left[-\frac{B_{\mu,\alpha}^2}{2\langle\Delta B_\mu{}^2\rangle}\right]$$

If isotropic:

$$f(|\mathbf{B}_\mu|) = f(\mathbf{B}_\mu)\, 4\pi B_\mu^2$$

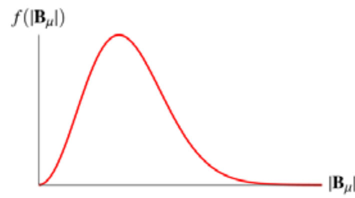
Maxwell distribution:

$$f(|\mathbf{B}_\mu|) = \frac{1}{\sqrt{2\pi\langle\Delta B_\mu^2\rangle^3}} 4\pi B_\mu^2 \exp\left[-\frac{B_\mu^2}{2\langle\Delta B_\mu^2\rangle}\right]$$



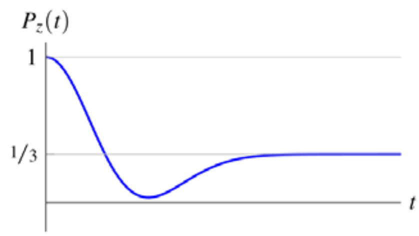
Randomly Oriented Moments

$$P_z(t) = \int f(\mathbf{B}_\mu)\left[\cos^2\theta + \sin^2\theta\cos(\gamma_\mu B_\mu t)\right]d\mathbf{B}_\mu$$



Kubo-Toyabe function

$$P_z(t) = \frac{1}{3} + \frac{2}{3}\left[1 - \gamma_\mu^2\langle\Delta B_\mu^2\rangle t^2\right]\exp\left[-\frac{\gamma_\mu^2\langle\Delta B_\mu^2\rangle t^2}{2}\right]$$



Example:

Kubo-Toyabe depolarization due to nuclear moments InN and MnSi

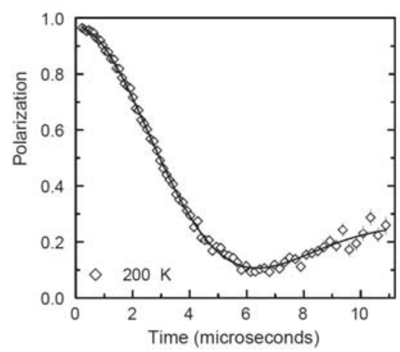
InN & MnSi

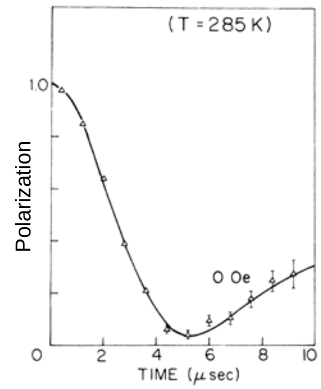
InN
Semiconductor
Study of the hydrogen-related defect chemistry

Y.G. Celebi et al., Physica B 340-342, 385 (2003)

MnSi
system lacking inversion symmetry
itinerant-electron magnet MnSi

R.S. Hayano et al., Phys. Rev. B 20, 850 (1979)





In the paramagnetic state, a KT function is very often observed reflecting the small field distribution created by the nuclear moments

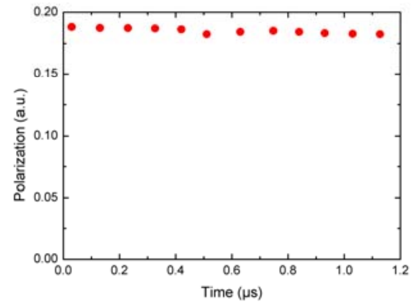
Example:

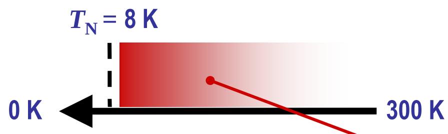
CePtSn
Transition from paramagnetism to antiferromagnetism through a disordered static moments phase

CePtSn

CePtSn: metallic Kondo system
- small hybridization of $4f$ – electrons (RKKY interaction > Kondo effect)
- Magnetic order at about 8 K
- Non-centrosymmetric system

G.M. Kalvius et al., Europhys. Lett. 29, 501 (1995)



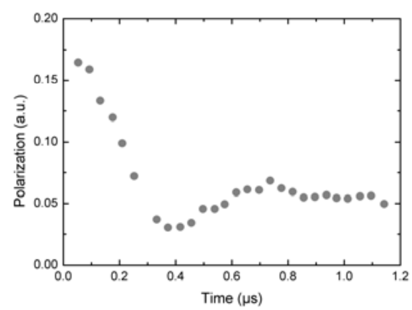


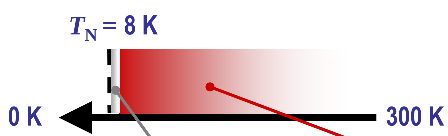
Paramagnetic state (fig. 9 K): weak depolarization due to nuclear moments (→ Kubo-Toyabe function with very weak depolarization)

CePtSn

CePtSn: metallic Kondo system
- small hybridization of $4f$ – electrons (RKKY interaction > Kondo effect)
- Magnetic order at about 8 K
- Non-centrosymmetric system

G.M. Kalvius et al., Europhys. Lett. 29, 501 (1995)





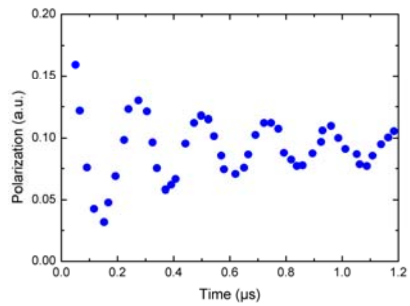
Just above AF order (fig.: 8.3 K): strong depolarization due to ramdomly oriented static electronic magnetic moments (→ Kubo-Toyabe function)

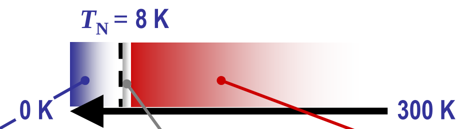
Paramagnetic state (fig. 9 K): weak depolarization due to nuclear moments (→ Kubo-Toyabe function with very weak depolarization)

CePtSn

CePtSn: metallic Kondo system
- small hybridization of 4f – electrons (RKKY interaction > Kondo effect)
- Magnetic order at about 8 K
- Non-centrosymmetric system

G.M. Kalvius et al., Europhys. Lett. 29, 501 (1995)





In the AF state (fig. 7 K): spatial ordering of the magnetic moments (→ well defined muon-spin precessions)

Just above AF order (fig.: 8.3 K): strong depolarization due to ramdomly oriented static electronic magnetic moments (→ Kubo-Toyabe function)

Paramagnetic state (fig. 9 K): weak depolarization due to nuclear moments (→ Kubo-Toyabe function with very weak depolarization)

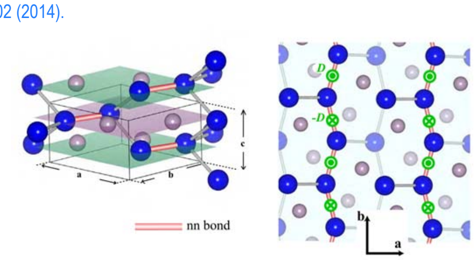
Detection of Magnetic Phase Separation
-
Coexistence of Different Magnetic Phases

Example:

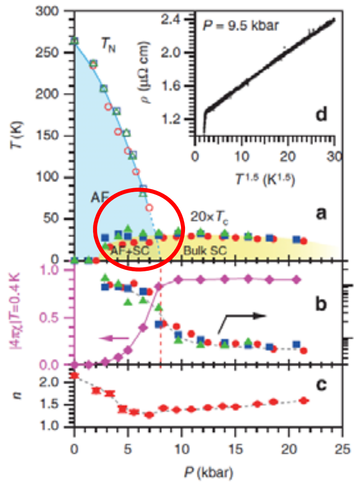
Magnetic Phase Separation in CrAs

CrAs

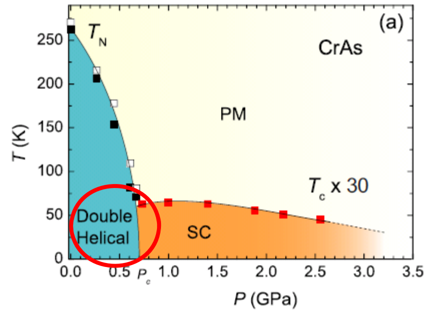
- *Pnma* structure
- Magnetic transition at 265 K (helimagnetic state)
  propagation vector k along [001], 1.7 μ_B
- T_N decreases with pressure
  Magnetic order suppressed above $p_c \approx 0.7$ GPa
- **Superconductivity** occurs when the magnetic phase is suppressed
  ($T_c \approx 2$ K)
  W. Wu et al., Nature Commun. **5**, 5508 (2014)
  H. Kotegawa et al., J. Phys. Soc. Jpn. **83**, 093702 (2014).



CrAs

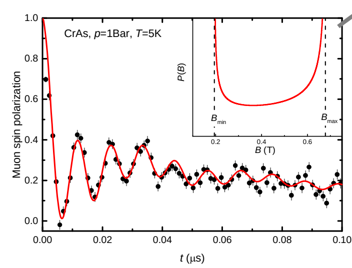


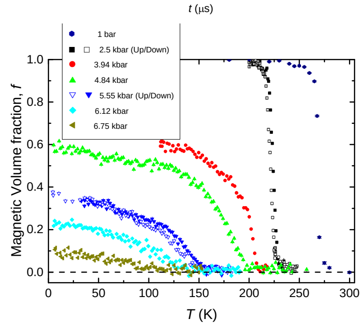
Wu et al., Nat. Comm. **5**, 5508 (2014)
Resistivity & Magnetization

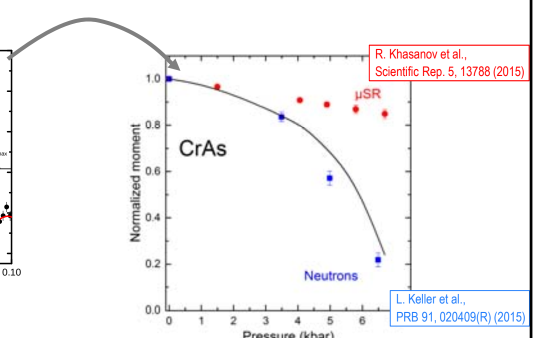


Kotegawa et al., J. Phys. Soc. Jpn. 83, 093702 (2014)
Resistivity

CrAs: ordered moment







From μSR:

- Ordered moment
- Magnetic Volume

From Neutrons:
Average moment

$$m_{\text{average}} = m\sqrt{f}$$

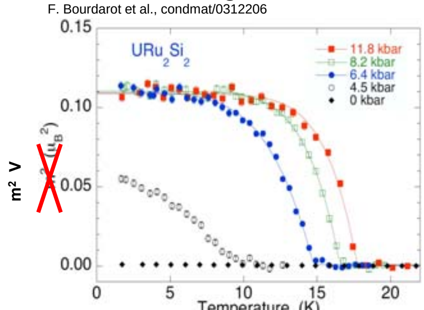
Q = (0,0, 0.3562)

Example:

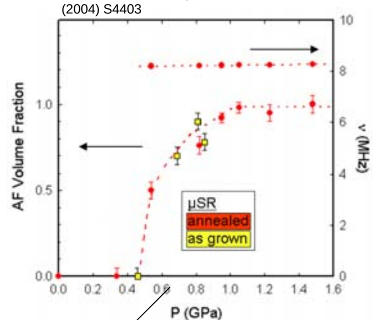
Magnetic Phase Separation in URu_2Si_2

URu_2Si_2

Neutron scattering:
F. Bourdarot et al., condmat/0312206



Muon spin relaxation:
A. Amato et al., J. Phys.: Condens. Matter **16** (2004) S4403



Phase separation into magnetic and non-magnetic regions.

Only the combination of neutron scattering and muon spin relaxation allowed the correct interpretation of the data.

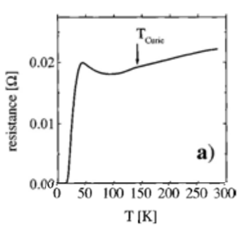
Example:

Microscopic Coexistence of Superconductivity and Magnetism in $RuSr_2GdCu_2O_8$

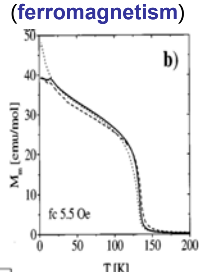
Alex Amato Masterschool 2017

$RuSr_2GdCu_2O_8$

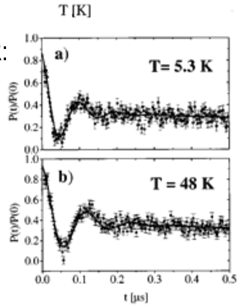
Resistivity:
(**superconductivity**)



Magnetization:
(**ferromagnetism**)

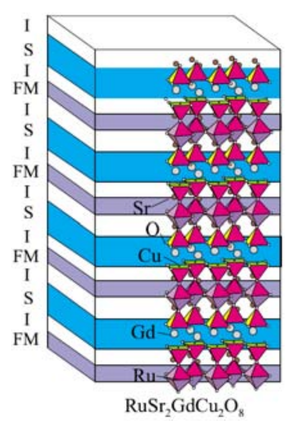


μSR:



~100%
Magnetic volume

Microscopic coexistence of superconductivity and magnetism



$RuSr_2GdCu_2O_8$

Structure:
T. Nachtrab et al.,
Phys. Rev. Lett. ***92*** *(2004) 117001*

C. Bernhard et al., Phys. Rev. B 59 (1999) 14099

Alex Amato Masterschool 2017

Summary
Magnetism detection by μSR

Magnetism:

- **Local probe**
  - Magnetic volume fraction
- **μSR frequency**
  - Magnetic order parameter (10^{-3}–10^{-4} μ_B)
  - Temperature dependence
- **μSR relaxation rate**
  - Homogeneity of magnetism
- **Magnetic fluctuations**
  - Time window: 10^5 – 10^9 Hz

Summary – ZF µSR

Single Crystals – Magnet

$$P_z(t) = \exp\left[-\frac{1}{2}\gamma_\mu^2\langle\Delta B_\mu{}^2\rangle t^2\right]\cos(\gamma_\mu B_\mu t)$$

- **Frequency:** Size of the magnetic moments
- **Damping:** Inhomogeneity
- **Amplitude:** Magnetic volume fraction

Polycrystals – Magnet

$$P_z(t) = \frac{1}{3} + \frac{2}{3}\exp\left[-\frac{1}{2}\gamma_\mu^2\langle\Delta B_\mu{}^2\rangle t^2\right]\cos\left[\gamma_\mu\langle B_\mu\rangle t\right]$$

Randomly oriented static moments

$$P_z(t) = \frac{1}{3} + \frac{2}{3}\left[1 - \gamma_\mu^2\langle\Delta B_\mu^2\rangle t^2\right]\exp\left[-\frac{\gamma_\mu^2\langle\Delta B_\mu^2\rangle t^2}{2}\right]$$

Thanks

Figures, slides, and ideas taken from:
H. Luetkens, A. Suter, E. Morenzoni,
Th. Prokscha, *PSI*

Thank you for your attention!