



Polarized muons as microscopic probes of materials, thin films and heterostructures

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1st RAON Users Workshop,
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Outline

- PSI μ SR facility (SpS, Swiss Muon Source) and instruments
- Selected scientific examples:
 - magnetism, superconductivity and spin transport
 - in bulk materials, thin films and heterostructures
- Summary and outlook

Accelerator Facilities PSI West

High Intensity
Proton Accelerator
0.59 GeV, 2.4 mA

UCN

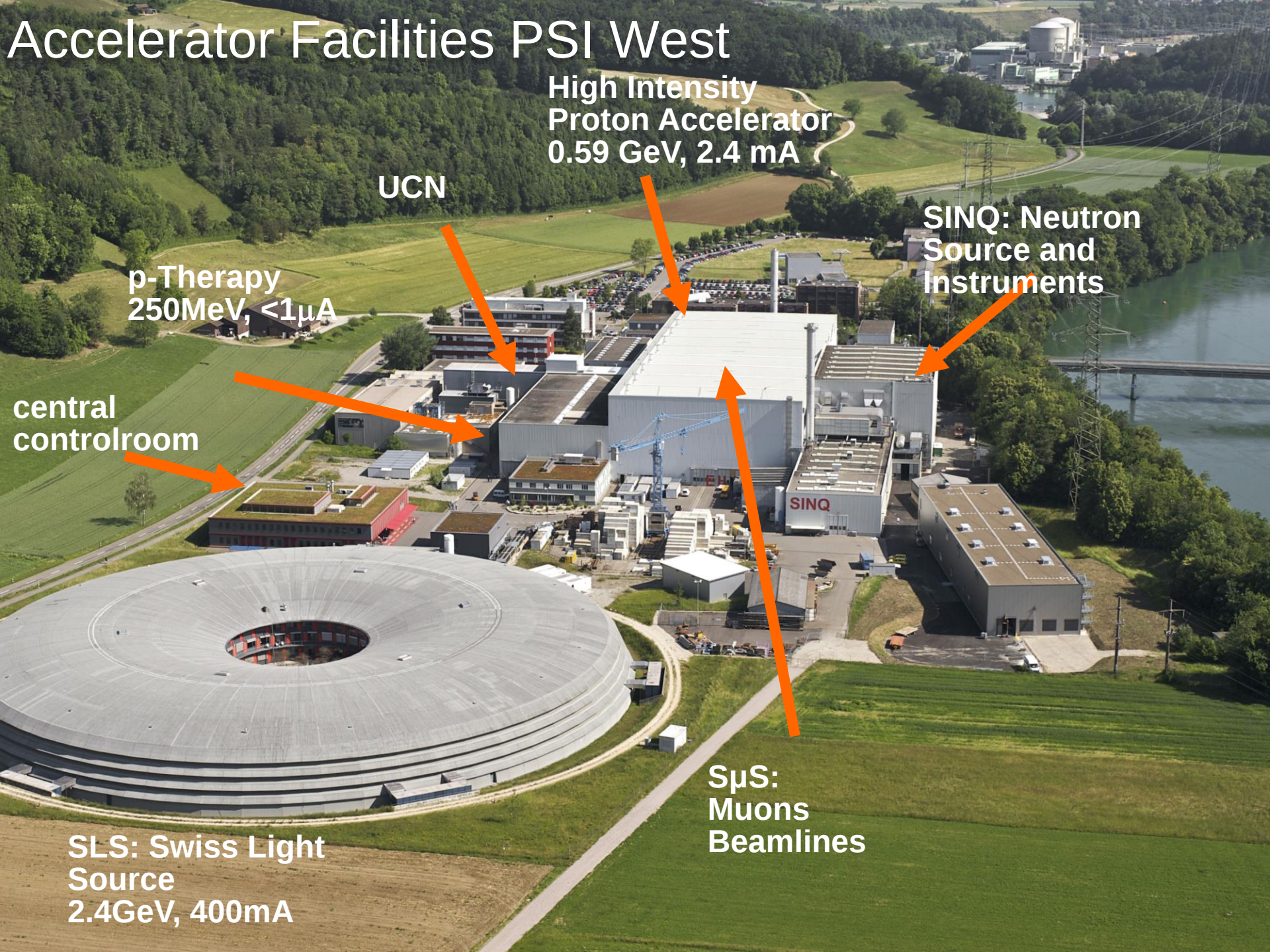
p-Therapy
250MeV, $<1\mu\text{A}$

SINQ: Neutron
Source and
Instruments

central
controlroom

S μ S:
Muons
Beamlines

SLS: Swiss Light
Source
2.4GeV, 400mA

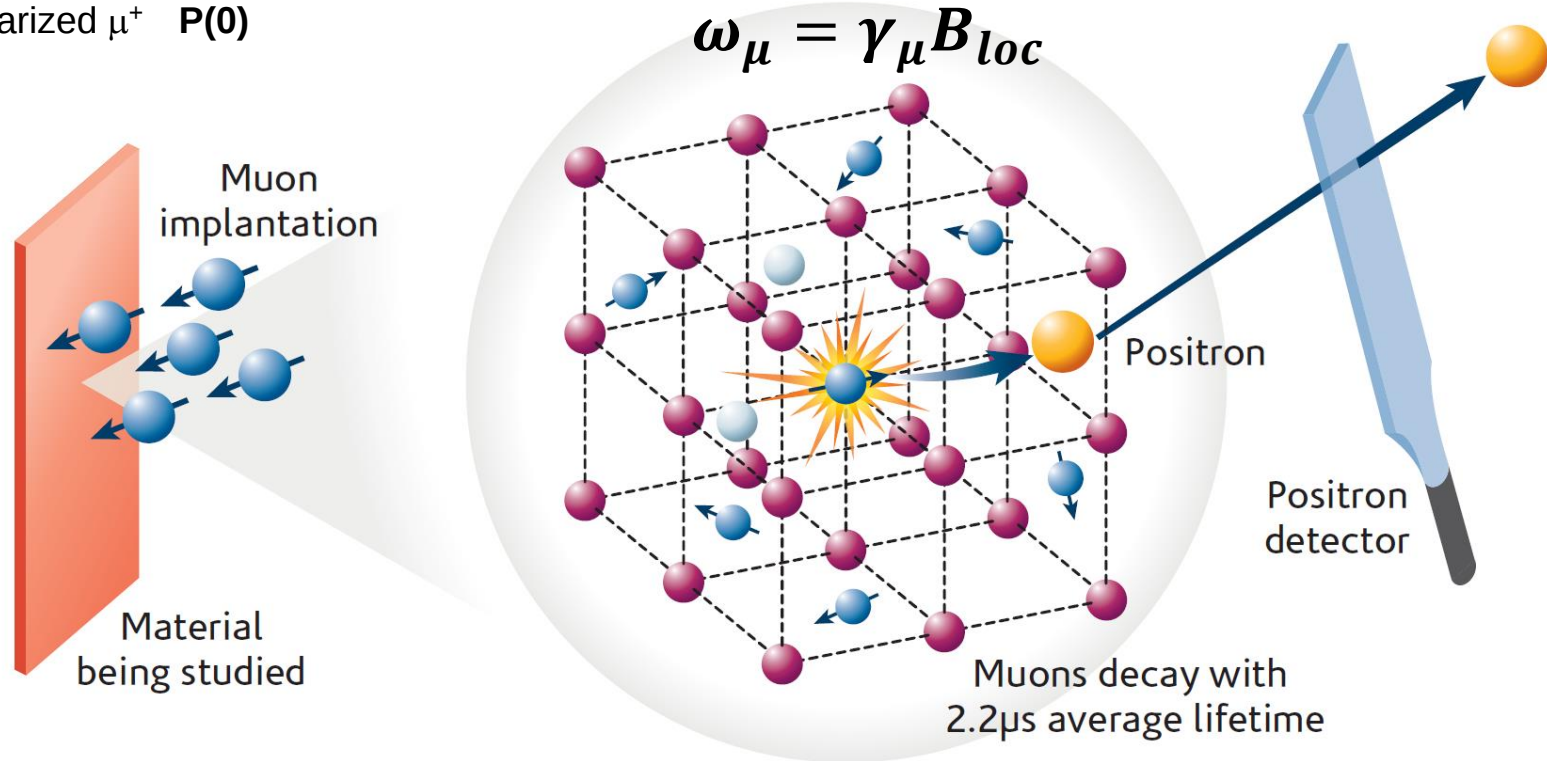


Polarized positive muons: magnetic microprobes of matter

Implant and thermalize ($\sim$ ps)
ensemble of 100% polarized μ^+ $\mathbf{P}(0)$

Muon spin interacts with local environment $\rightarrow \mathbf{P}(t)$

Measure $\mathbf{P}(t)$ via asymmetric muon decay



μ SR is sensitive to everything (static and dynamic) that changes the polarization of the muon: local magnetic fields, currents, spins, magnetic moments, fluctuations, chemical reactions (via muonium formation), electronic states

Muon Instruments at PSI : S_μS (Swiss Muon Source)

HAL9500

High Field and Low Temperature

Muon energy: **4.2 MeV** (μ^+)

9.5 T, < 20 mK

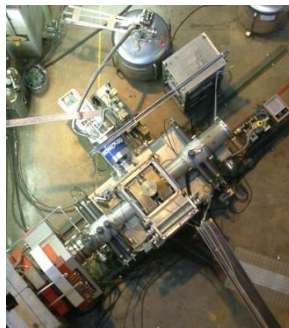


GPS

General Purpose Surface Muon Instrument

Muon energy: **4.2 MeV** (μ^+)

**0.6 T,
1.6 K 1200 K**



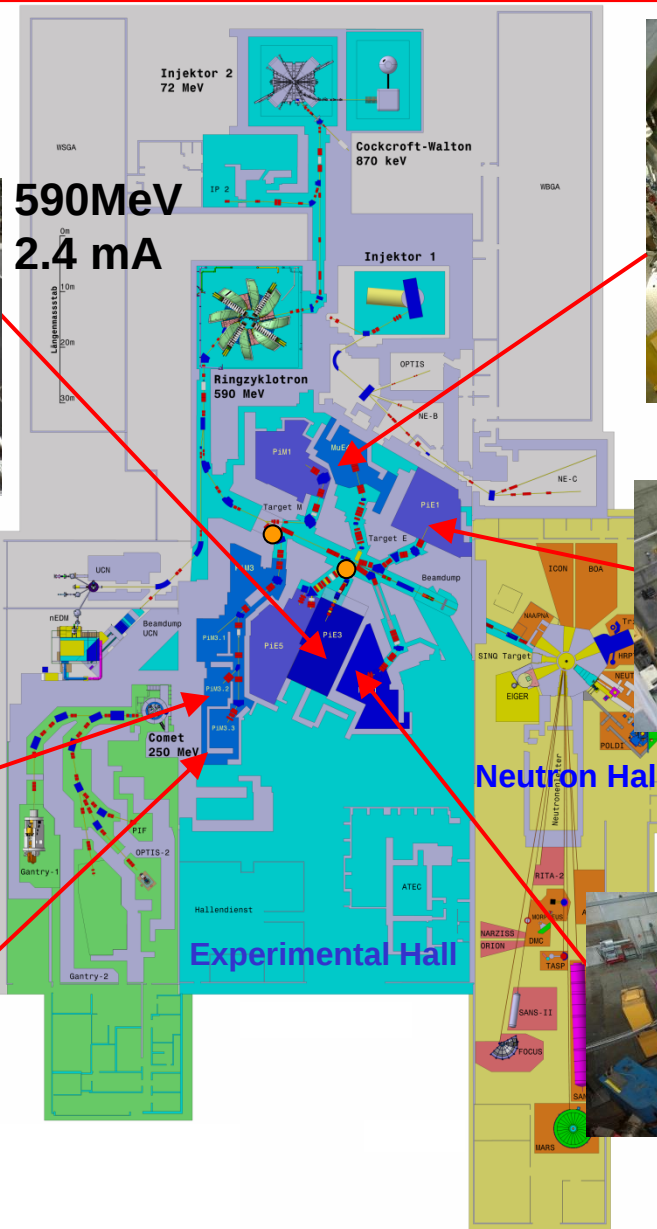
**Shared Beam Surface Muon Facility
(Muon On REquest)**

FLAME

Low Temperature Facility

Muon energy: **4.2 MeV** (μ^+)

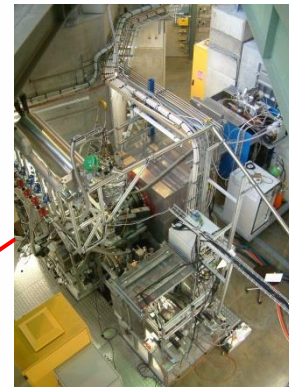
**3.5 T,
30 mK- 300 K
TF, LF**



LEM

Low-energy muon beam and instrument, tunable energy (**0.5-30 keV**, μ^+), thin-film, near-surface and multi-layer studies (1-300 nm)

**0.3 T
2.2 – 300 K**



DOLLY

General Purpose Surface Muon Instrument
 μ^+ energy: **4.2 MeV**

**0.5 T
250 mK**



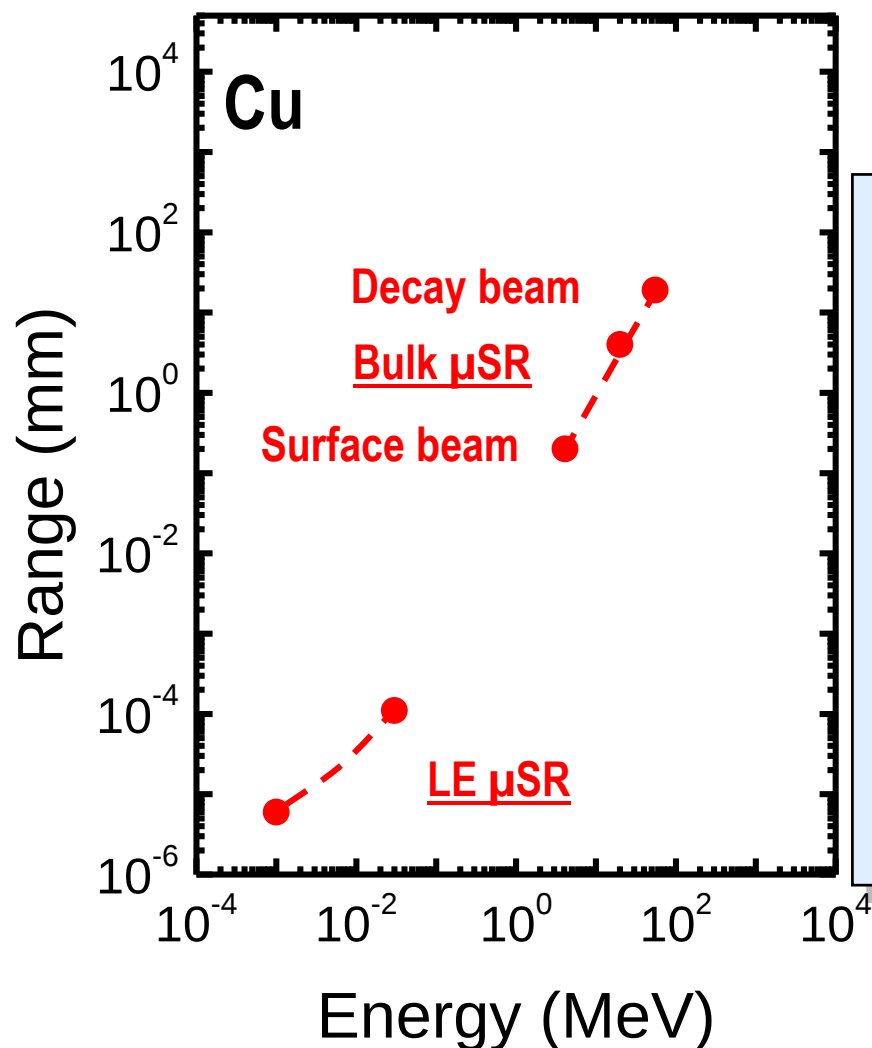
GPD

General Purpose Decay Channel Instrument
Pressure studies
Muon energy: **5 - 60 MeV** (μ^+ or μ^-)

**0.5 T,
250 mK
2.8 GPa**



Different muon energies for different studies



Bulk μ SR:

- ▶ “Surface” Muon Beams (4 MeV): “Normal” samples (sub-mm thick)
- ▶ Decay Muons beam (10-60 MeV): Bulky samples in pressure cells or in containers

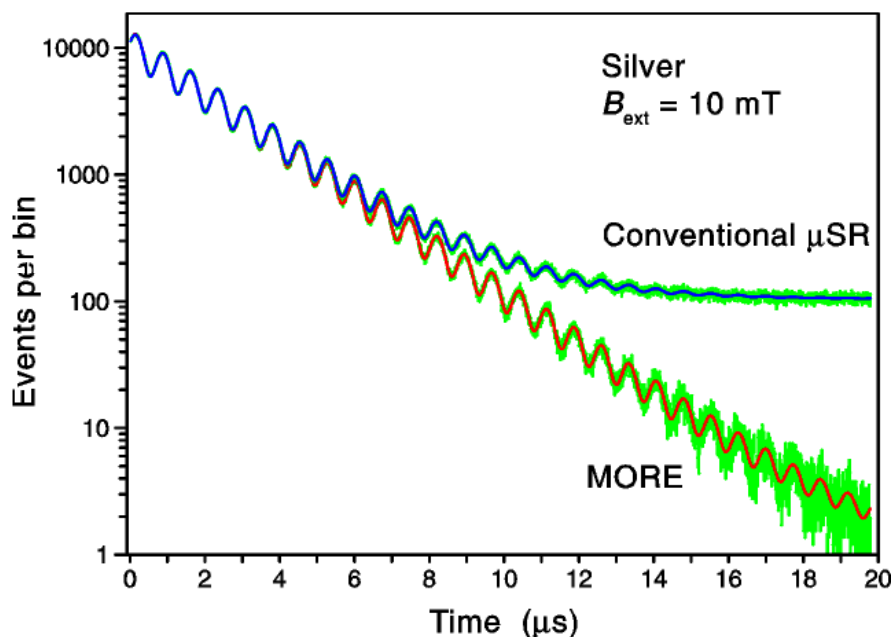
LE μ SR:

- ▶ Low energy muon beam (0.5-30 keV) for depth-dependent investigations (~ 2 –300 nm), thin films and heterostructures

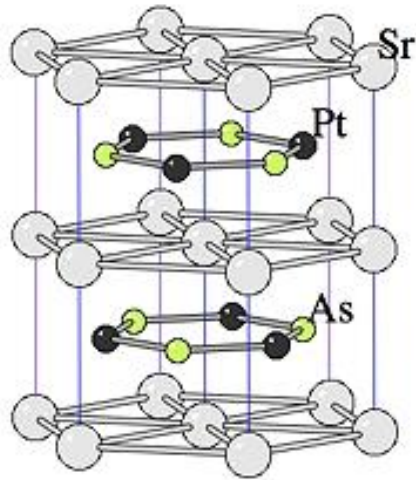
Muon On REquest: MORE on GPS/Flame

Extract one muon at a time out of a continuous beam using fast-switching kicker

Combines advantages of **continuous beam** (very fast timing → magnetism, superconductivity, high muon spin relaxation rate) and **pulsed beam** (very low background, → very weak magnetism, low muon spin relaxation rate)

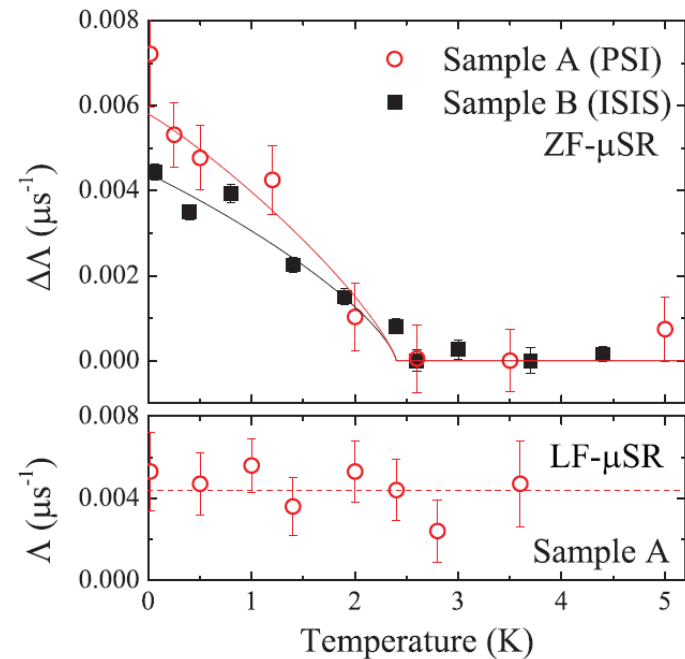
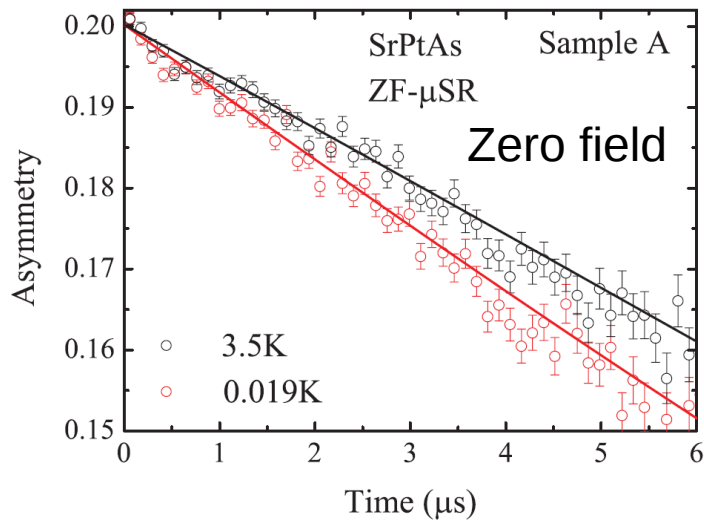


Time reversal symmetry breaking in SrPtAs



SrPtAs

$T_c = 2.4$ K
locally non-centrosymmetric
→ non trivial sc state, f, d+id,
TRSB

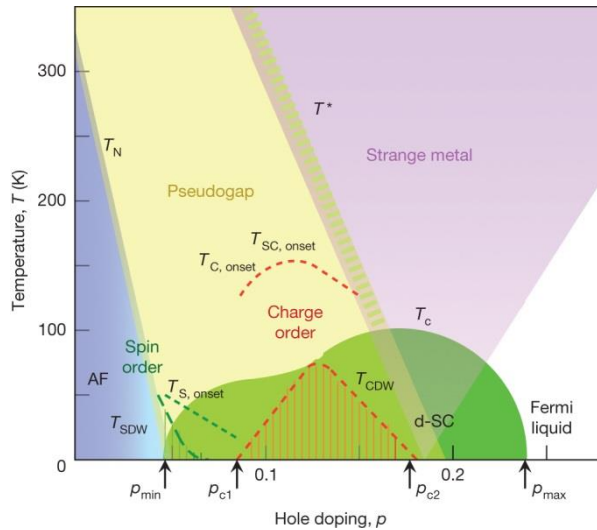


Spontaneous **very small magnetic field**
below $T_c \rightarrow \Delta B \approx 0.007$ mT $\rightarrow$ TRSB

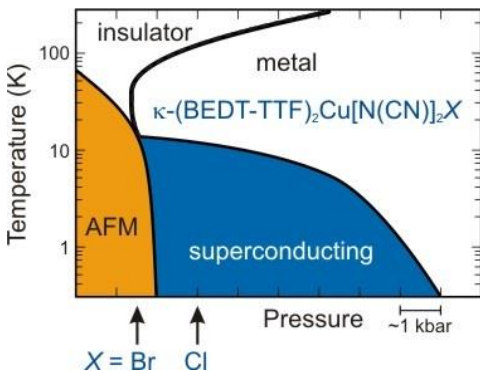
Detecting very small fields

Phase diagrams of superconductors

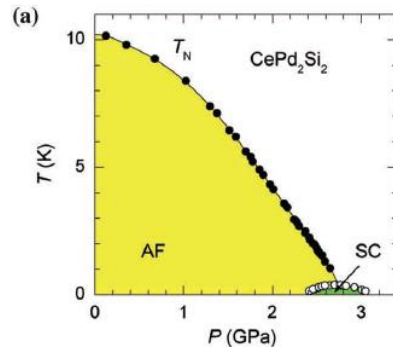
Electronic order/SC, magn., Phase competition/coexistence, QCP, interactions



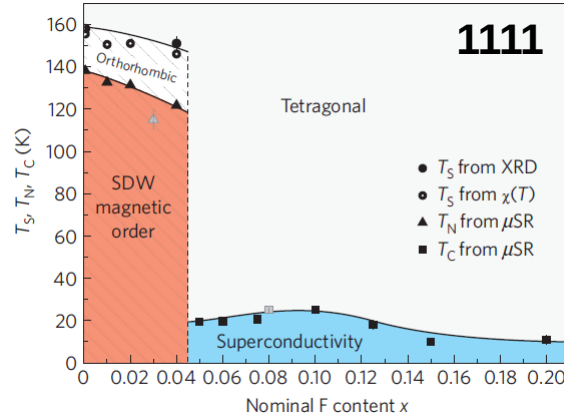
B Keimer et al. Nature **518**, 179-186 (2015)



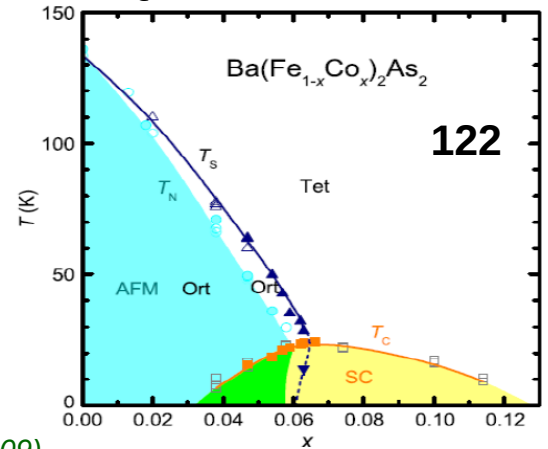
D. Faltermeier et al., Phys. Rev. B **76**, 165113 (2007)



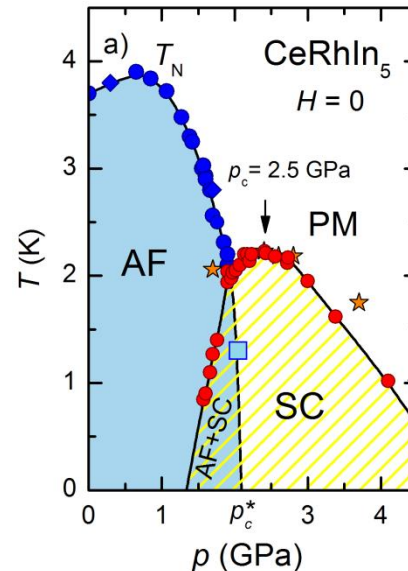
N.D. Mathur et al., Nature **1998**



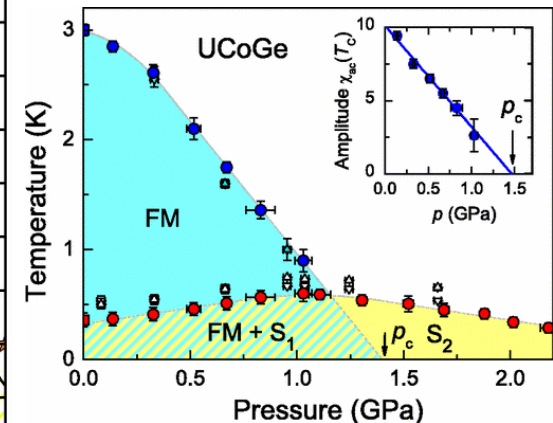
H. Luetkens et al., Nat. Mat. **8**, 305 (2009)



S. Nandi et al., PRL **104**, 057006 (2009)



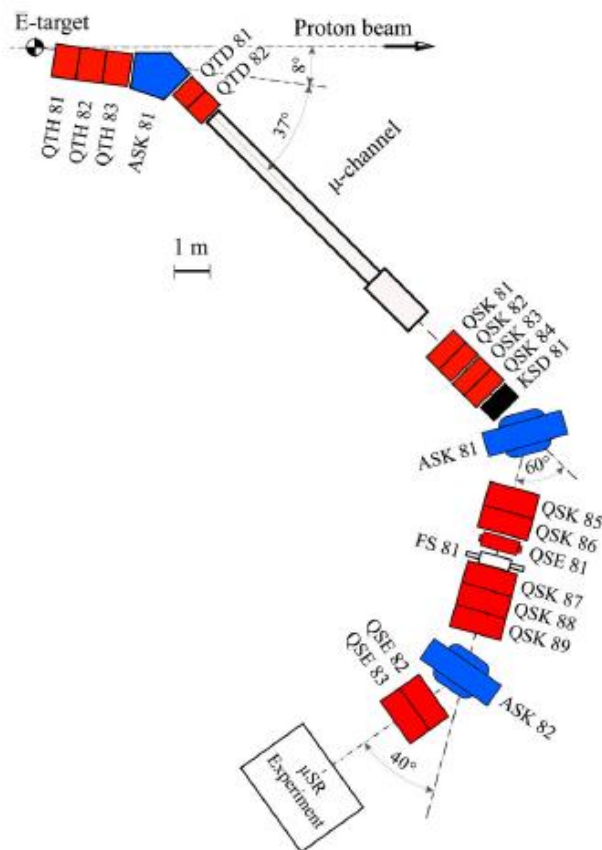
G. Knebel et al., Phys. Rev. B **74**, 020501(R) – (2006)



E. Slooten et al, PRL **2009**

μ SR studies under pressure

μ E1: Superconducting Decay Channel Beamline



- 10 - 60 MeV μ^+ or μ^- , 80% Polarization
- B_{ext} 0 - 0.6 T
- T 0.25 - 475 K
- Large sample chamber for **pressure cells** (diameter up to 40 mm)

μ SR studies under pressure

μ E1 Superconducting Decay Channel Beamline
High Energy Muon Instrument GPD



- 10 - 60 MeV μ^+ or μ^- , 80% Polarization
- B_{ext} 0 - 0.6 T
- T 0.25 - 475 K
- Large sample chamber for **pressure cells** (diameter up to 40 mm)

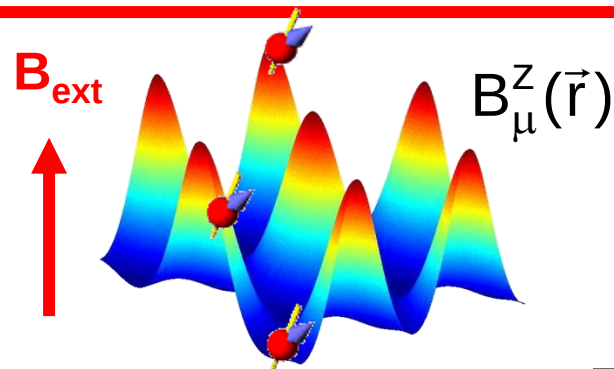
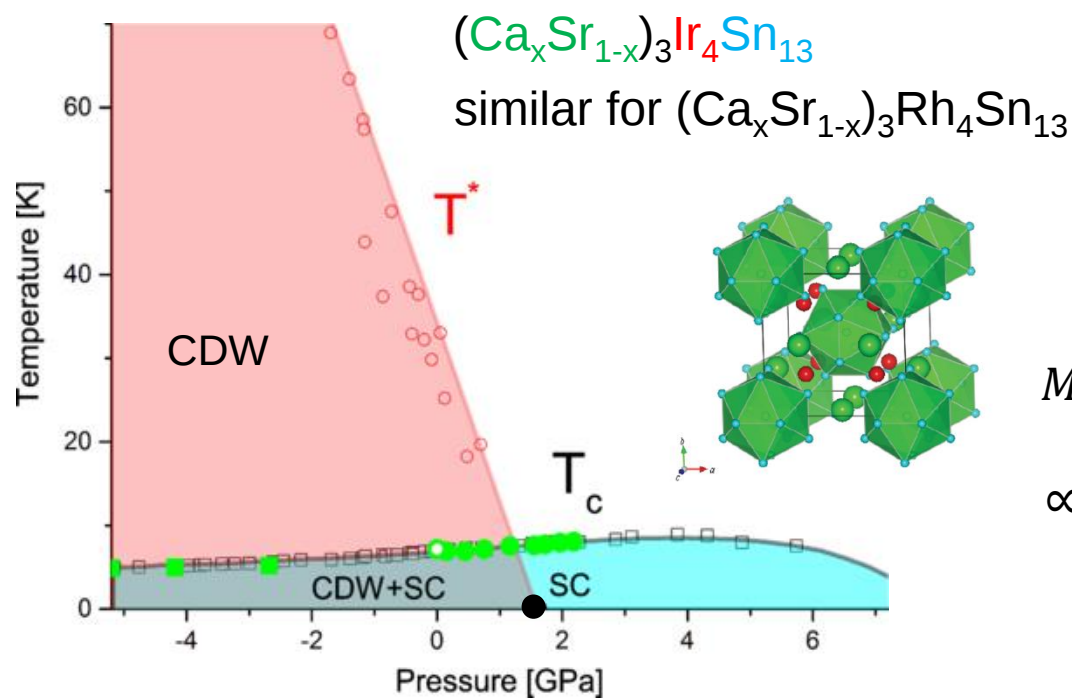
Pressure up to **2.8 GPa**
(@4K)

Double and single wall cells
Material: CuBe or MP35 alloy.
Diameter of the sample channel:
5.2 mm (7.2 mm).
Maximum sample height: 18mm
Pressure media: Daphne 7373 oil.



R. Khasanov et al.
High Pressure Research, **36**, 140 (2016)

QCP in the superconducting state of quasiskutteridites $R_3T_4X_{13}$

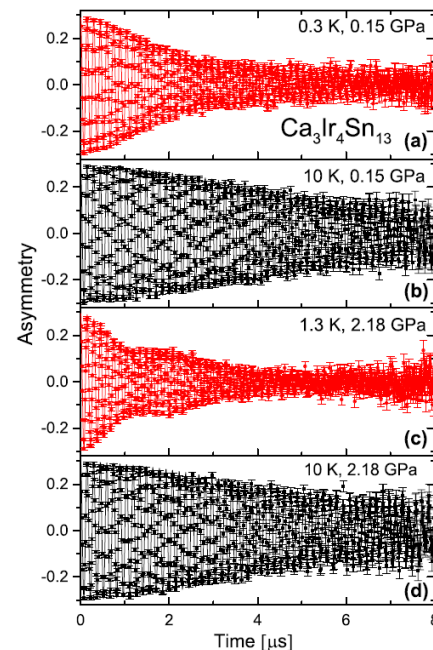


Muon spin relaxation $\sigma = \gamma_{\mu} \sqrt{\langle \Delta B_z^2 \rangle}$

$\propto$ superfluid density $\propto \frac{1}{\lambda^2} \propto \frac{n_s}{m^*}$

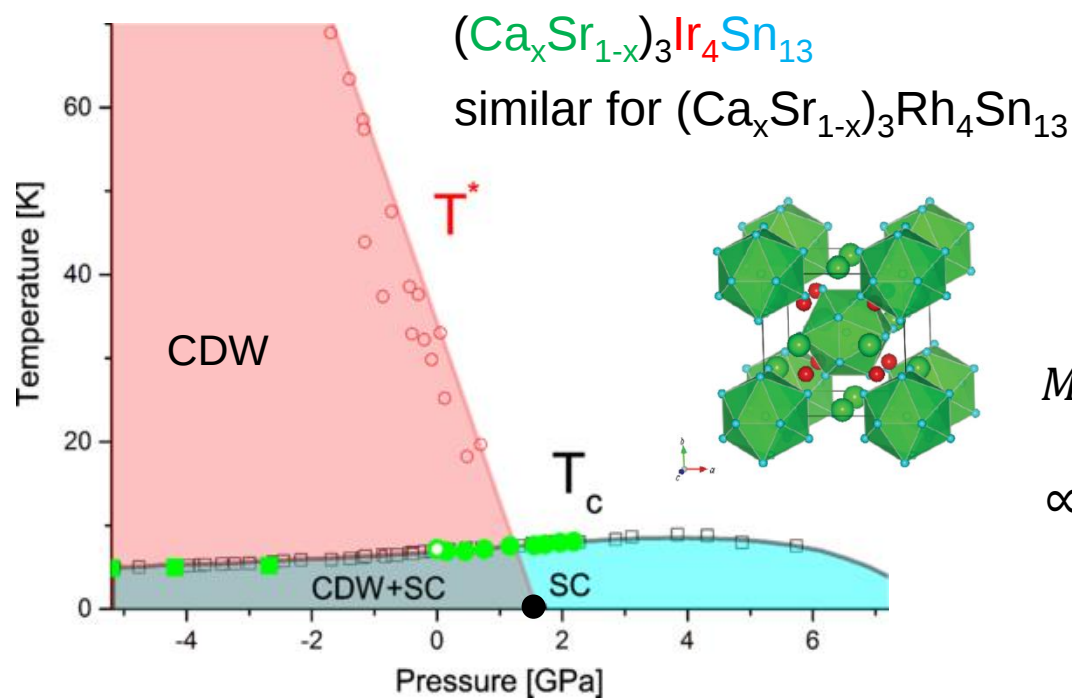
T^* : structural transition and FS reconstruction/CDW

$T^*(p)$ decreases with pressure suggesting **quantum critical point (QCP)** at $p_c \approx 1.8$ GPa for $Ca_3Ir_4Sn_{13}$ and -0.6 GPa for $Ca_3Rh_4Sn_{13}$ (corresponds to $(Ca_{0.9}Sr_{0.1})_3Rh_4Sn_{13}$)



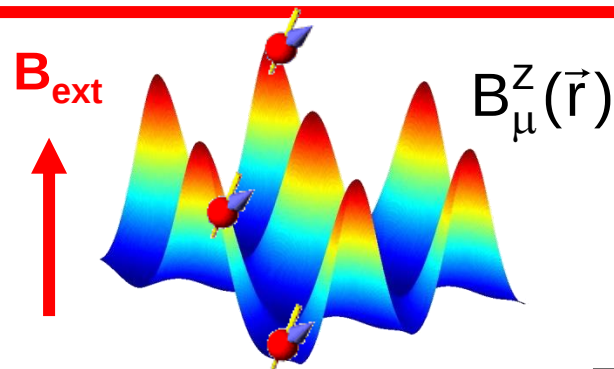
P. Biswas, J. Krieger, E. M. et al., PRB **92**, 195122 (2015), PRB **90**, 144505 (2014) and in preparation (2019)

QCP in the superconducting state of quasiskutteridites $R_3T_4X_{13}$



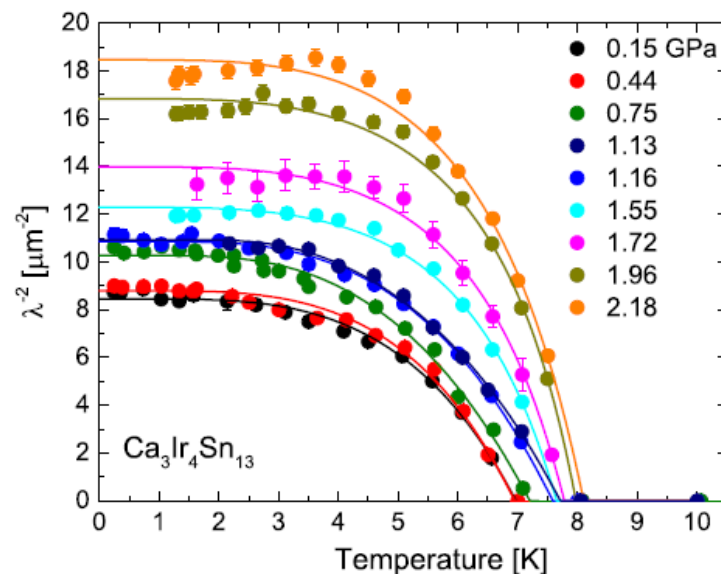
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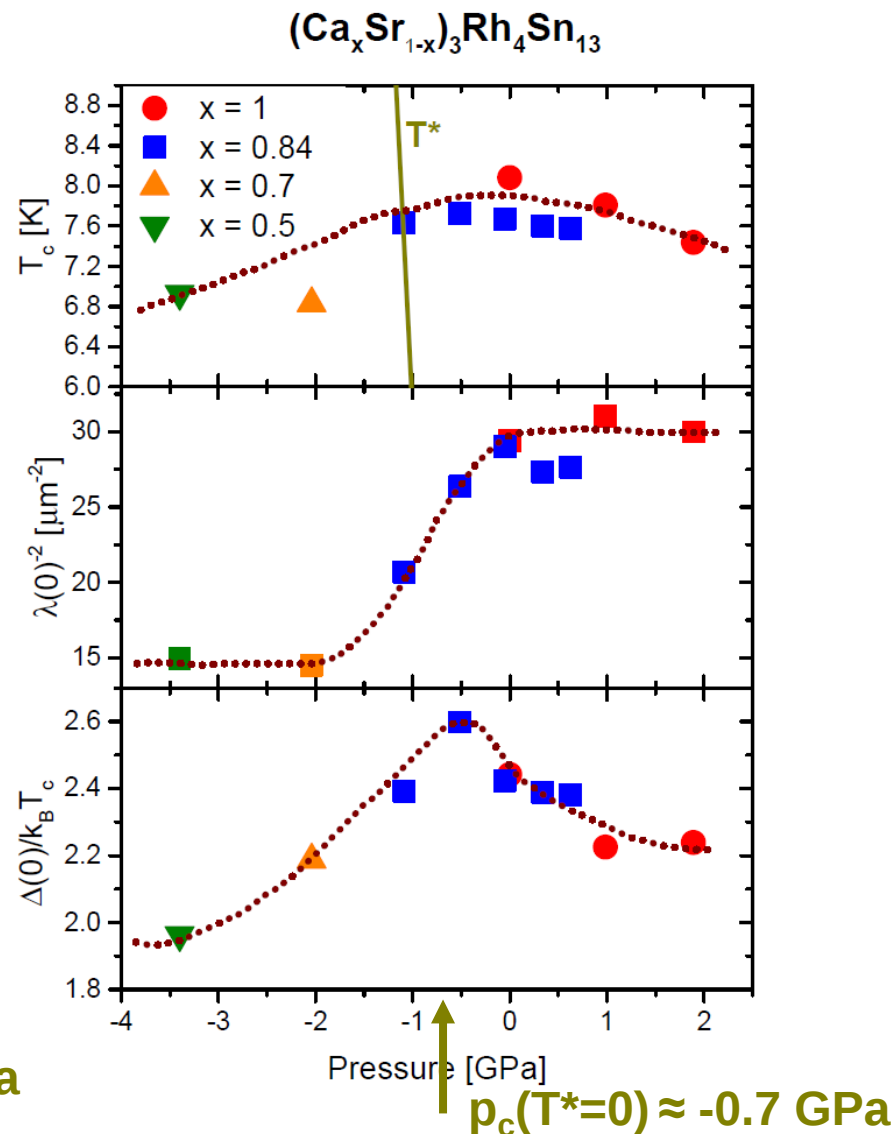
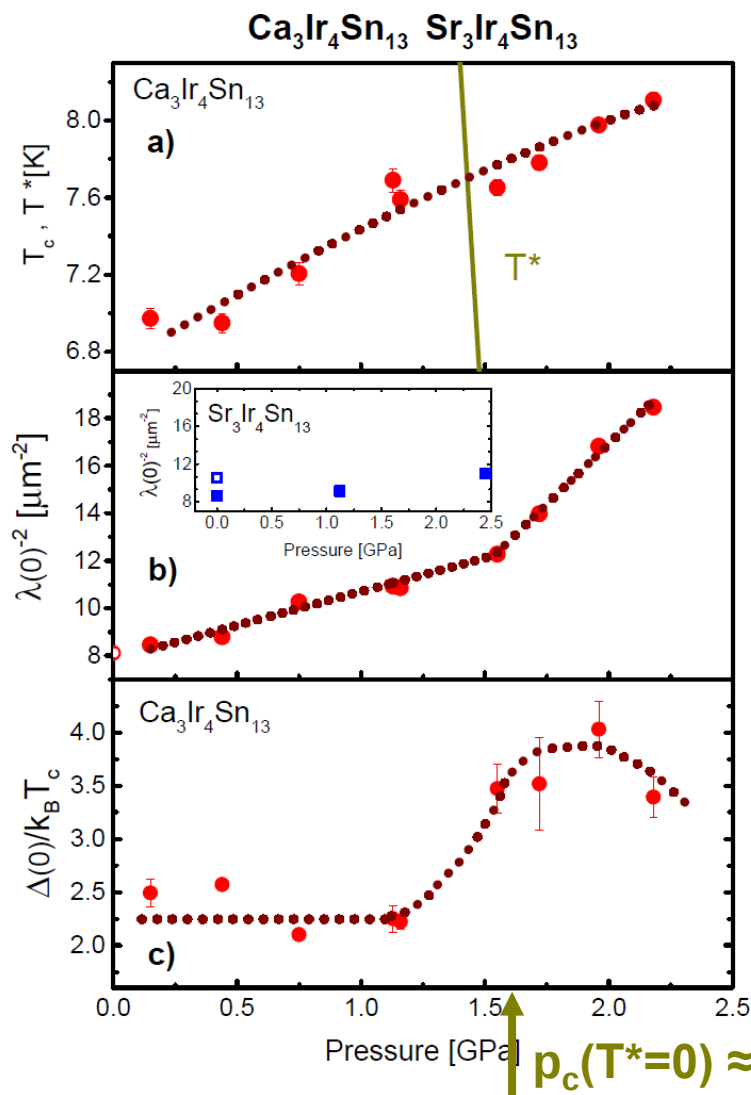


Muon spin relaxation $\sigma = \gamma_\mu \sqrt{\langle \Delta B_z^2 \rangle}$

$\propto$ superfluid density $\propto \frac{1}{\lambda^2} \propto \frac{n_s}{m^*}$



Strong enhancement of SC at QCP

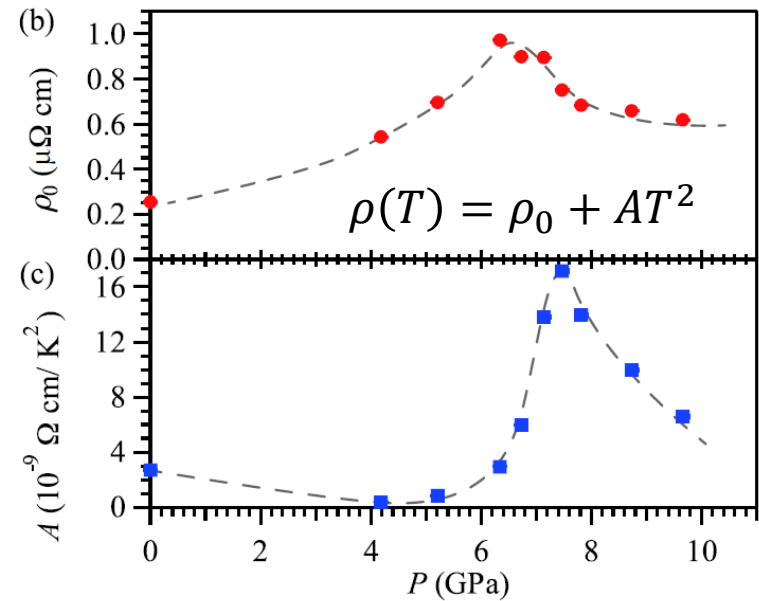
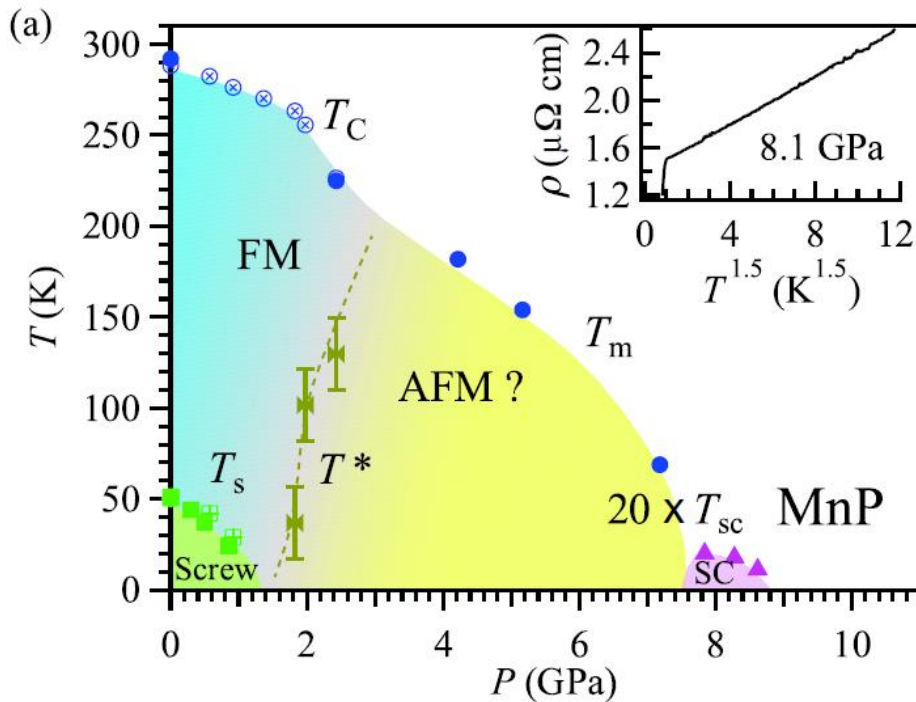
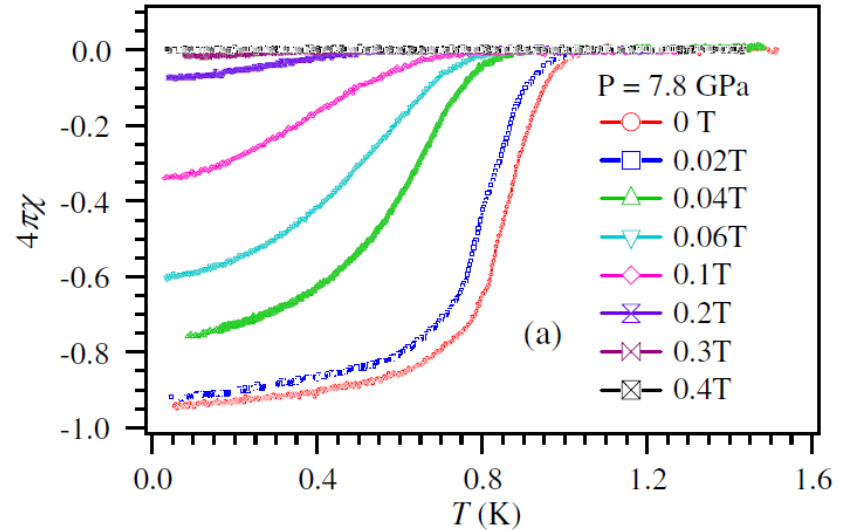
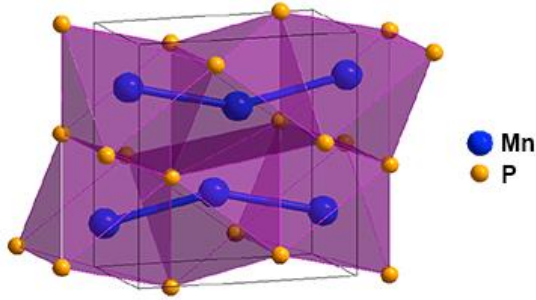


$$\frac{\Delta(0)}{k_B T_c} = 2.04(8) \quad @ \quad p = -5.2 \text{ GPa} \quad (Sr_3Ir_4Sn_{13})$$

$$\frac{\Delta(0)}{k_B T_c} = 1.93(8) \quad @ \quad p = -6.8 \text{ GPa} \quad (Sr_3Rh_4Sn_{13})$$

Pressure Induced Superconductivity in Mn based “helical” magnet

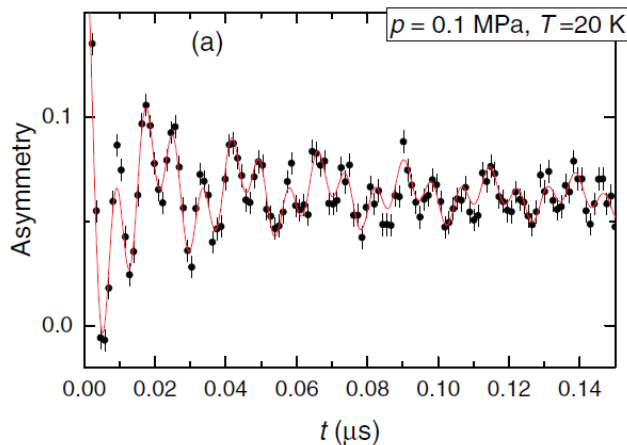
MnP: First Mn based SC $T_c \approx 1$ K



J. G. Cheng et al, PRL 114, 117001 (2015)

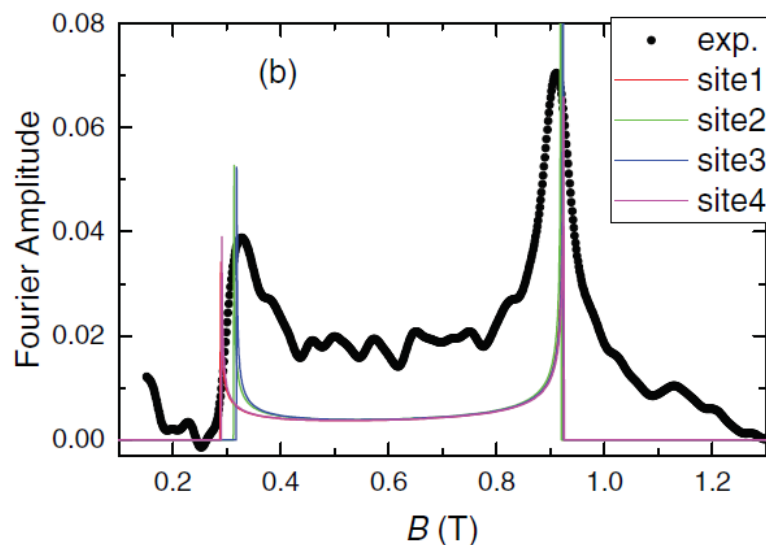
Magnetic structure of MnP under pressure

μ SR spectrum (ZF)



$$\omega = \gamma_{\mu} B_{loc}$$

Fourier transform



DFT calculation $\rightarrow$ muon sites

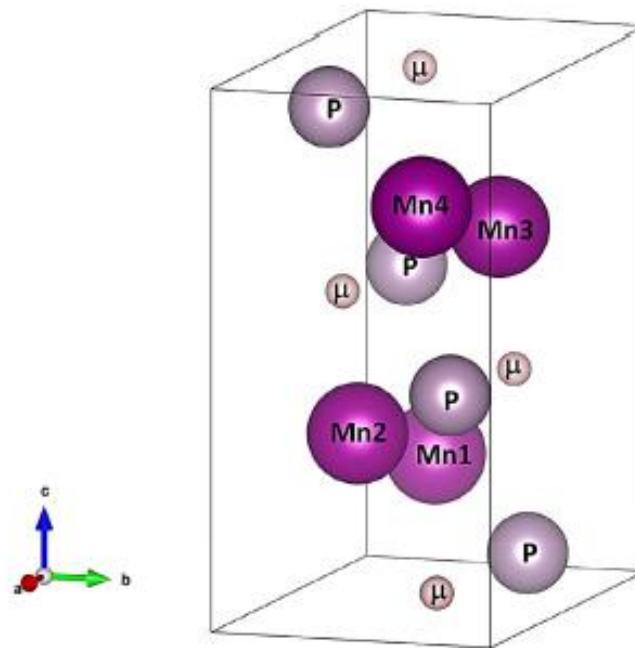
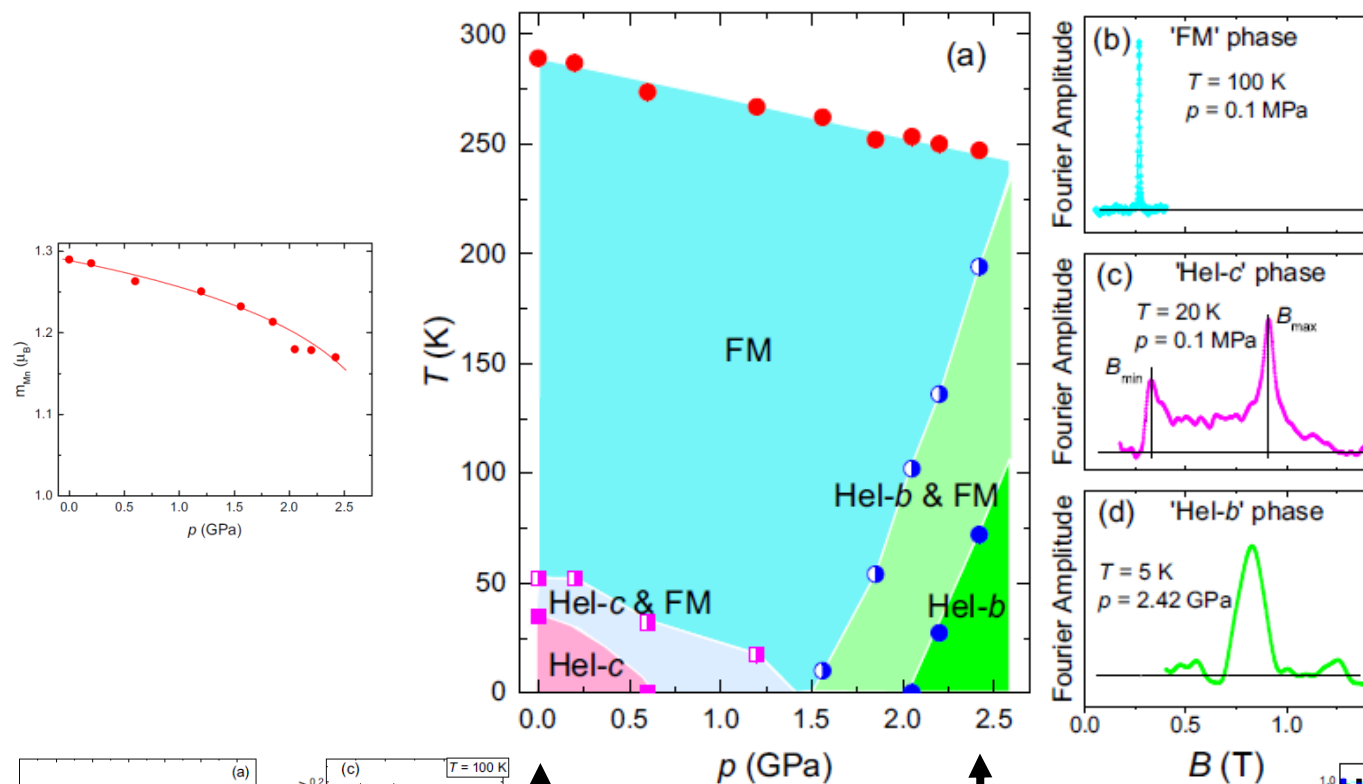


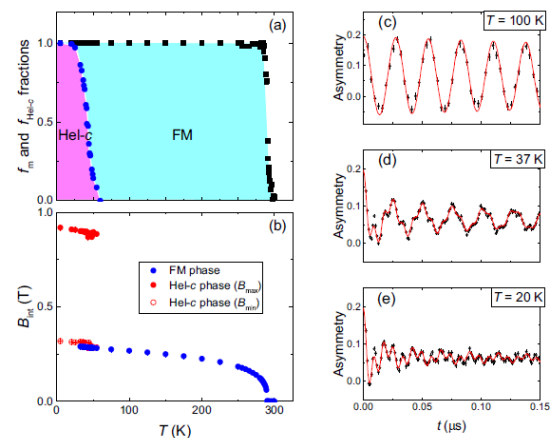
Figure 2. The orthorhombic crystal structure of MnP (*Pnma*, 62). The muon stopping positions (sites #1) were obtained by *ab initio* calculations (see section 3.2 and table 1). The structure was visualized by using VESTA [19].

Comparison local fields (determined by magnetic structure)
with experiment $\rightarrow$ magnetic structure

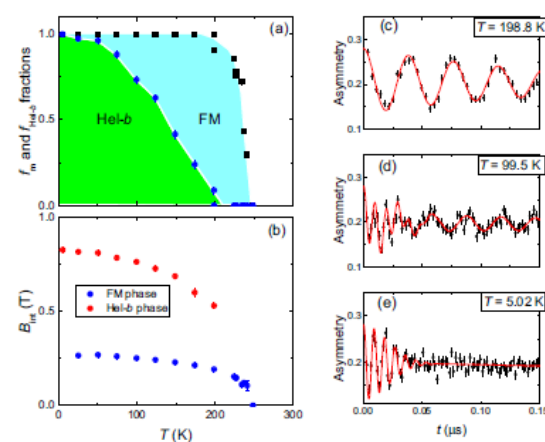
Magnetic structure of MnP under pressure



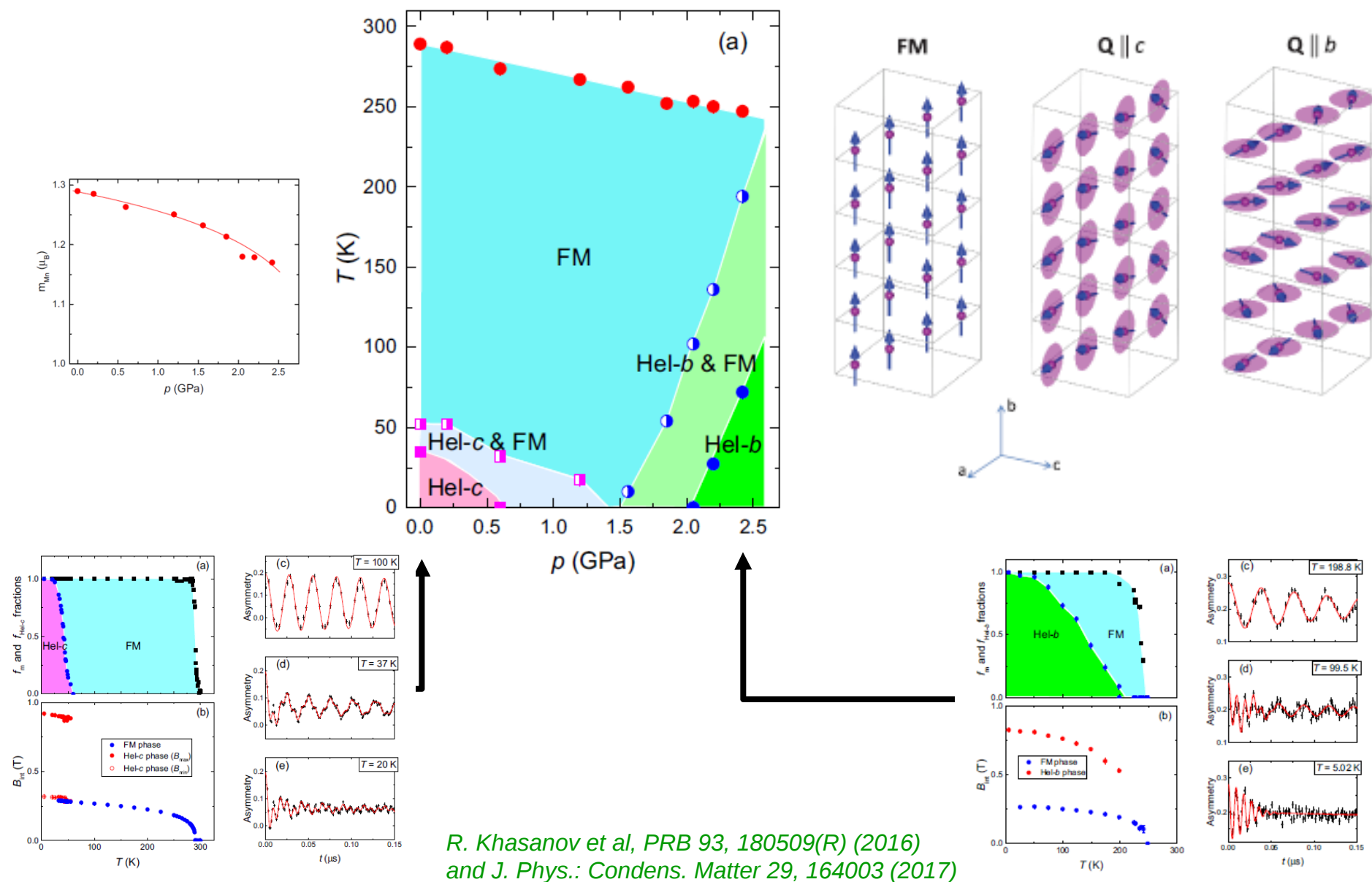
μ SR resolves discrepancy between neutron and X-ray structure data



*R. Khasanov et al, PRB 93, 180509(R) (2016)
and J. Phys.: Condens. Matter 29, 164003 (2017)*



Magnetic structure of MnP under pressure



Low energy μ^+ beam and instrument for LE- μ SR

- UHV system, 10^{-10} mbar
- some parts LN₂ cooled

Polarized Low Energy Muon Beam

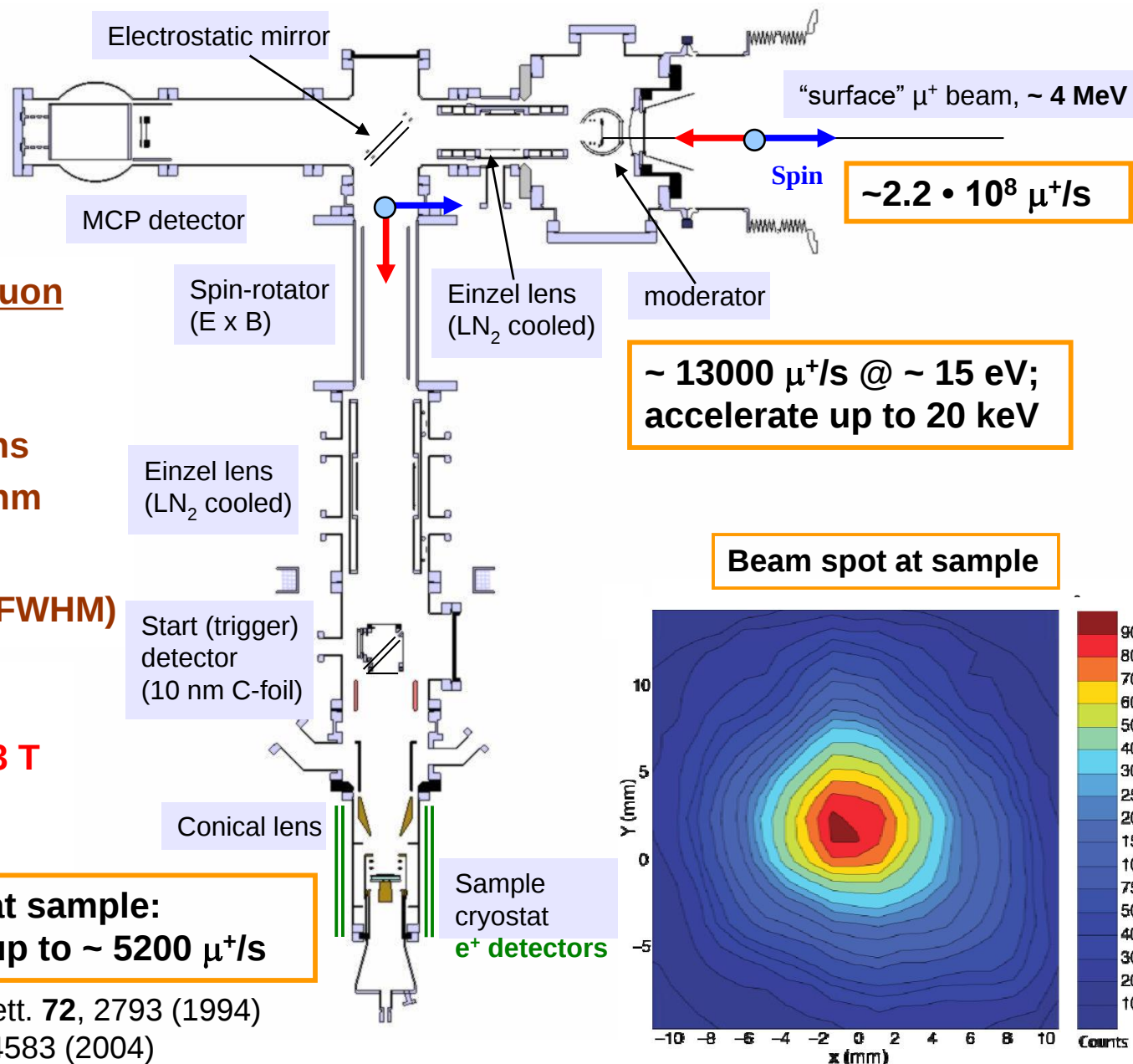
Energy: 0.5-30 keV
 ΔE , Δt : 400 eV, 5 ns
Depth: ~ 2 – 300 nm
Polarization: ~ 100 %
Beam Spot: ~ 12 mm (FWHM)

Sample environment:

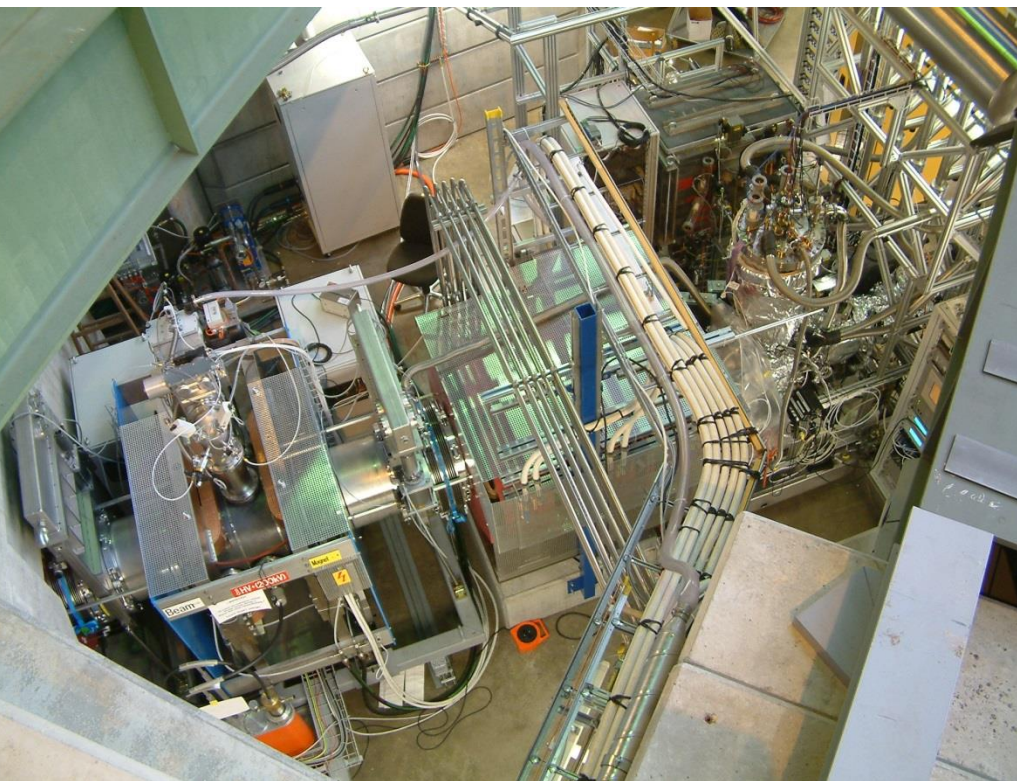
$B_{\perp} = 0 - 0.3$ T, $B_{\parallel} = 0 - 0.03$ T
(to sample surface)

$T = 2.5 - 320$ K

at sample:
up to ~ 5200 μ^+ /s



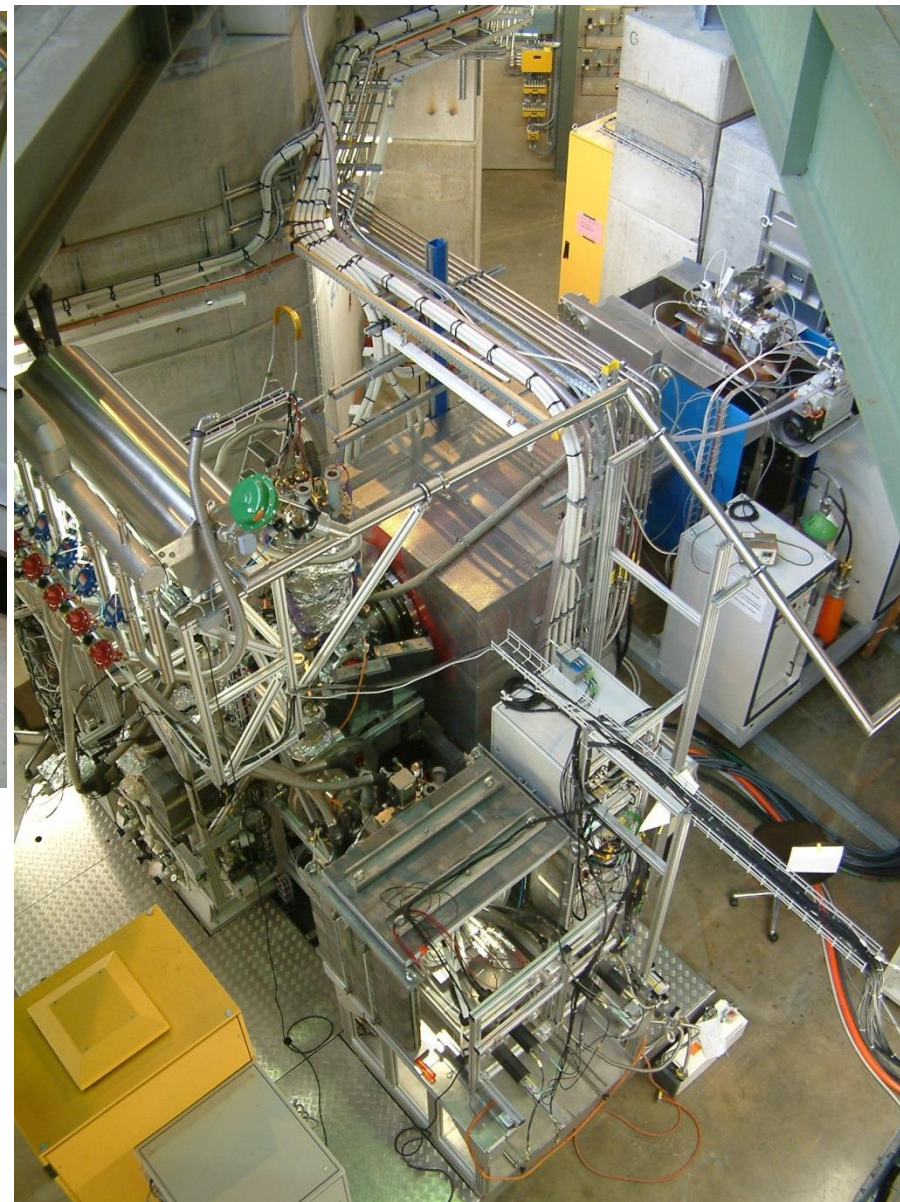
Low Energy- μ^+ Apparatus @ μ E4



$\sim 6.6 \cdot 10^8 \mu^+/\text{s}$ total

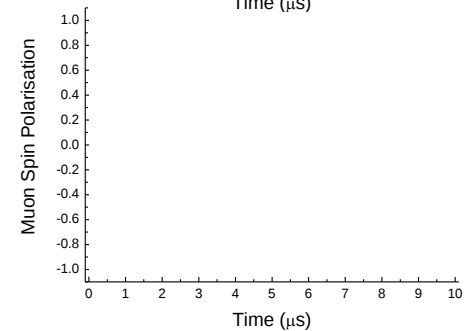
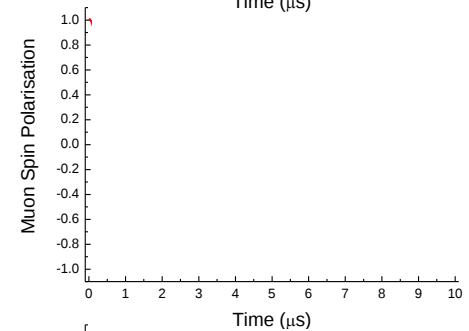
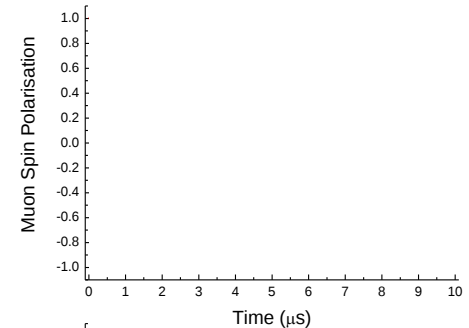
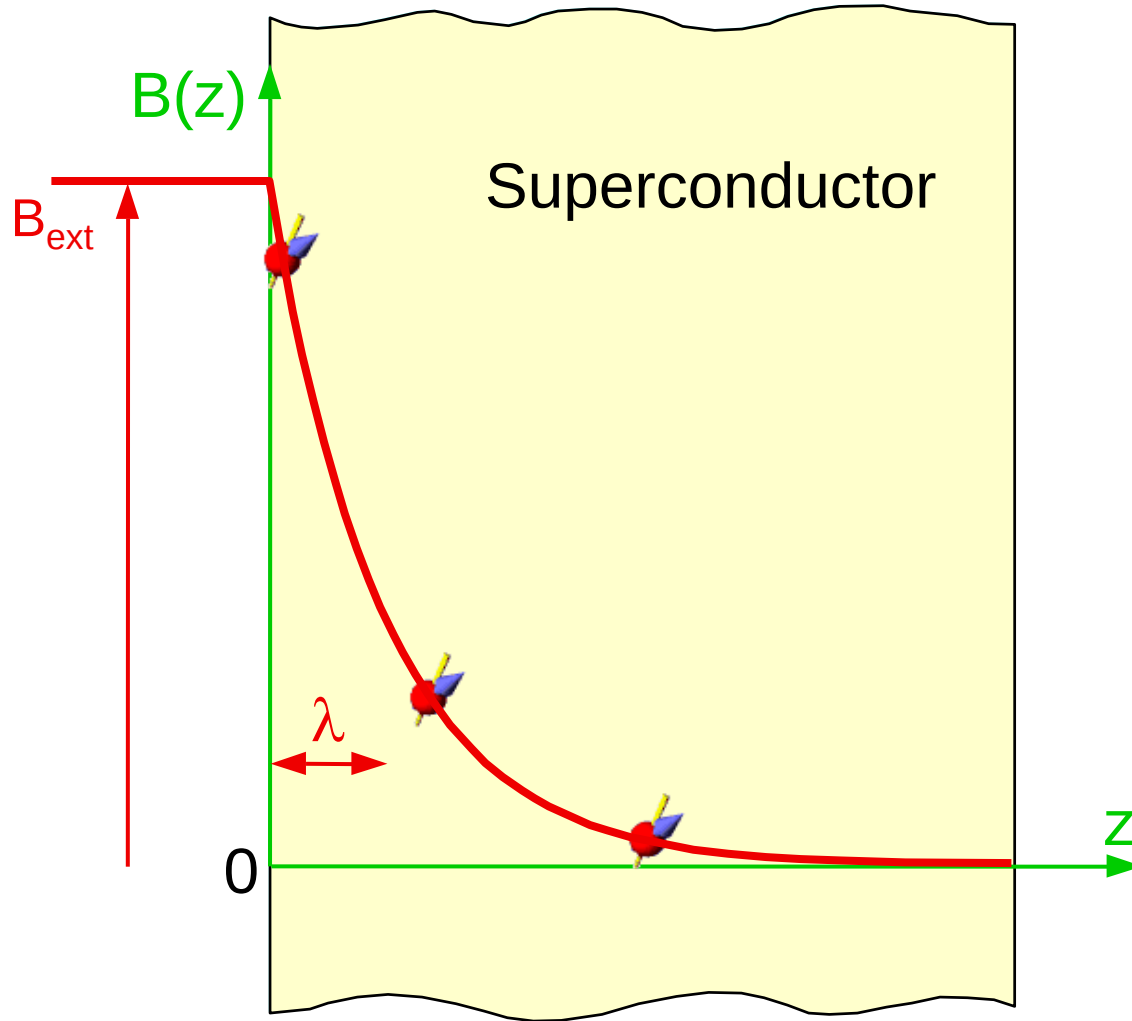
$\sim 2.2 \cdot 10^8 \mu^+/\text{s}$ on
LEM source

High flux “surface”
muon beam $\rightarrow$
Worldwide most
intense surface muon
beam



Th. Prokscha, E. Morenzoni, K. Deiters, F. Foroughi, D. George, R. Kobler, A. Suter and V. Vrankovic
Physica B 374-375, 460-464 (2006)
and Nucl. Instr. Meth. A 595, 317-331 (2008)

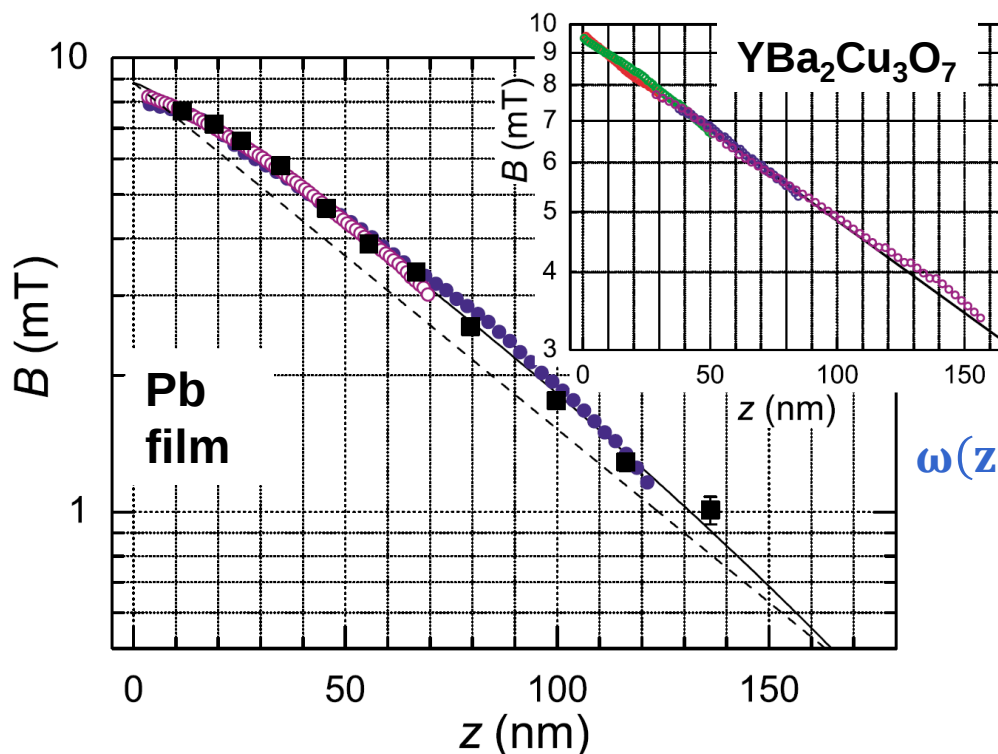
Depth dependent μ SR measurement



$$\omega_{\mu}(z) = \gamma_{\mu} B_{\text{loc}}(z)$$

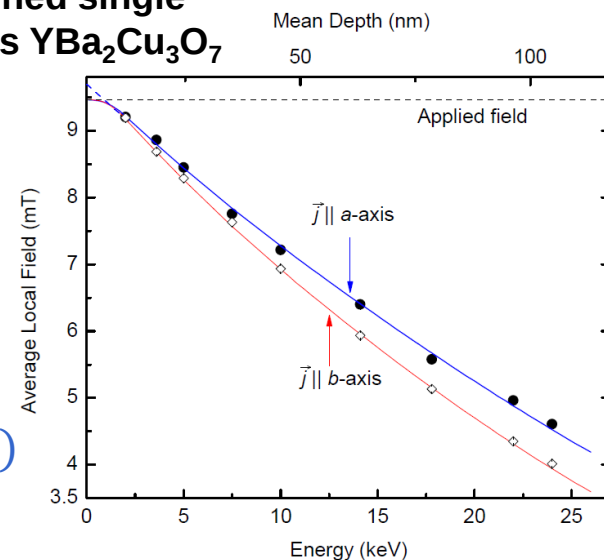
→ Magnetic field profile $B(z)$ over nm scale,
Direct measurements of SC length scales, λ , ξ

Magnetic field profiling at surfaces and in thin films

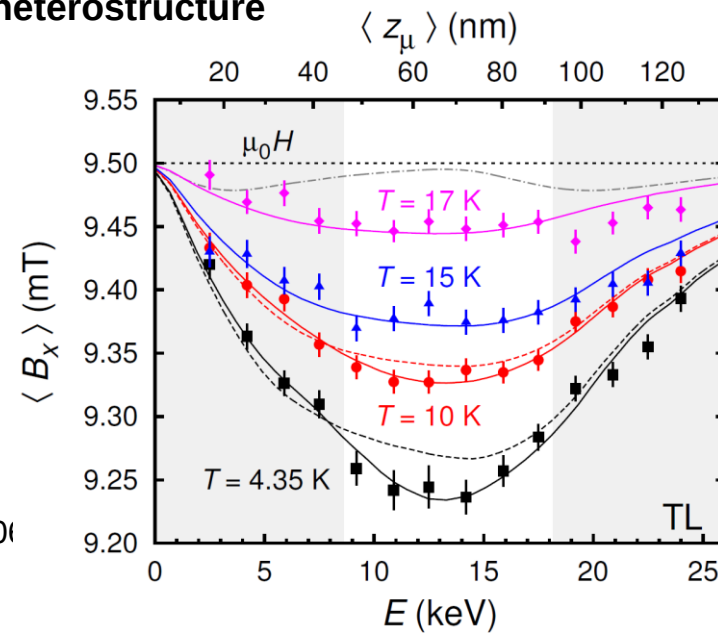


$$\omega(z) = \gamma_{\mu} B_{\text{loc}}(z)$$

Detwinned single crystals YBa₂Cu₃O₇



LSCO OP/UD/OP heterostructure



Magnetic field profiling on nm scale
Characteristics length scales λ , ξ
Anisotropies
Proximity effects

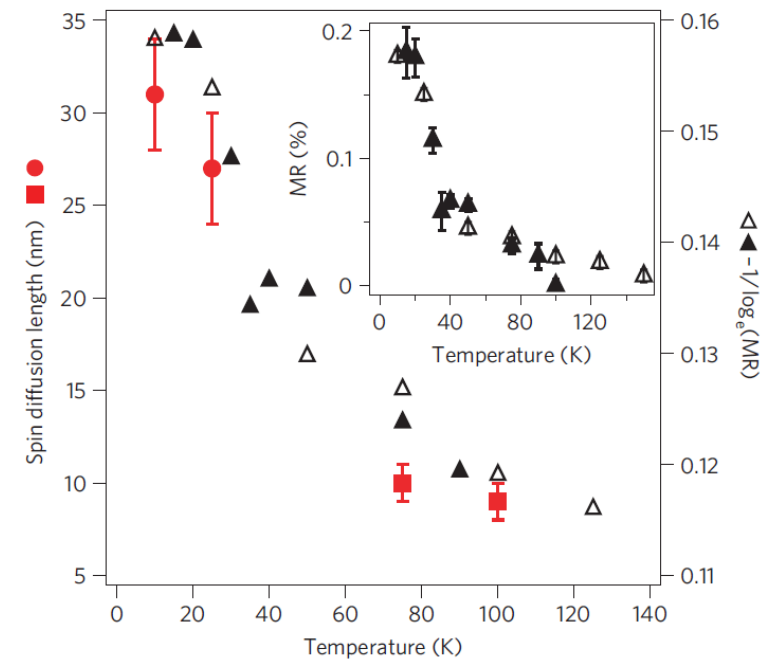
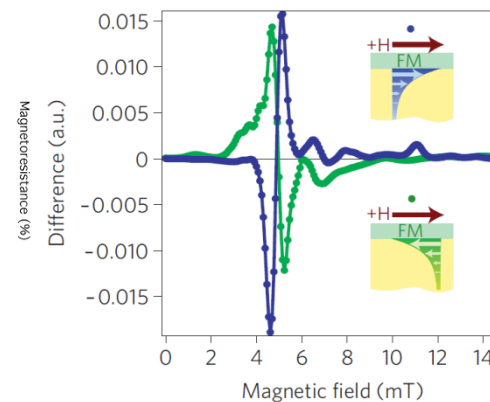
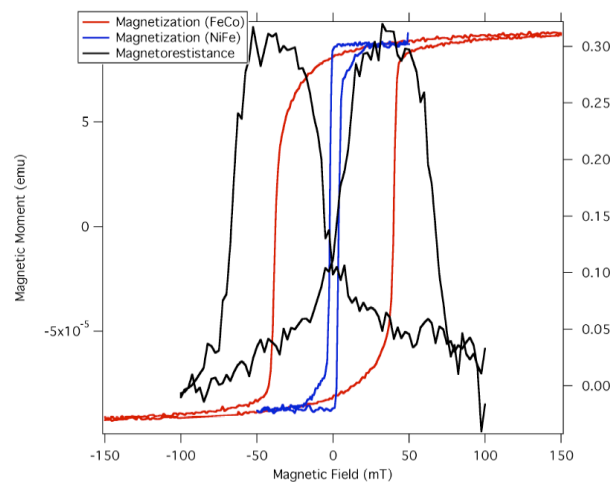
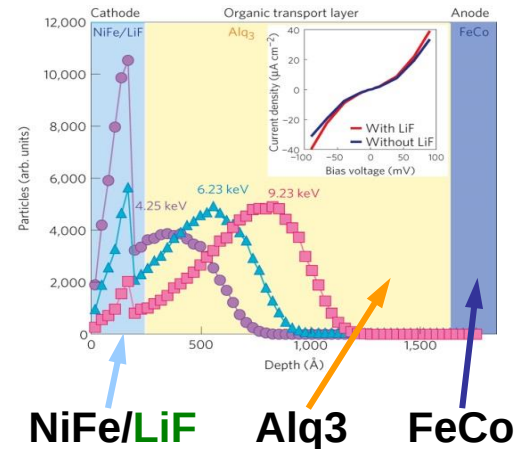
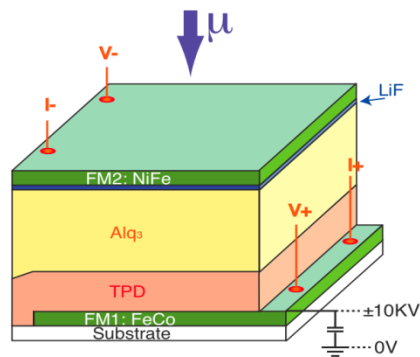
T.J. Jackson *et al.*, Phys. Rev. Lett. **84**, 4958 (2000)
A. Suter, E. M. *et al.* Phys. Rev. Lett. **92**, 087001 (2004) and Phys. Rev. B **72**, 024501 (2005)
R. F. Kiefl *et al.*, Phys. Rev. B **81**, 180502(R) (2010)
E. Morenzoni *et al.*, Nat. Commun. **2**:272 (2011).

Direct measurement of the electronic spin diffusion length in a fully functional organic spin valve by low-energy muon spin rotation

Probing spins in heterostructures

A. J. Drew^{1,2*}, J. Hoppler^{1,3}, L. Schulz¹, F. L. Pratt⁴, P. Desai², P. Shakya², T. Kreouzis², W. P. Gillin², A. Suter⁵, N. A. Morley⁶, V. K. Malik¹, A. Dubroka¹, K. W. Kim¹, H. Bouyanfif¹, F. Bourqui¹, C. Bernhard¹, R. Scheuermann⁵, G. J. Nieuwenhuys⁵, T. Prokscha⁵ and E. Morenzoni⁵

FM/sm/FM



Spin diffusion (decoherence) length
↔ Magnetoresistance

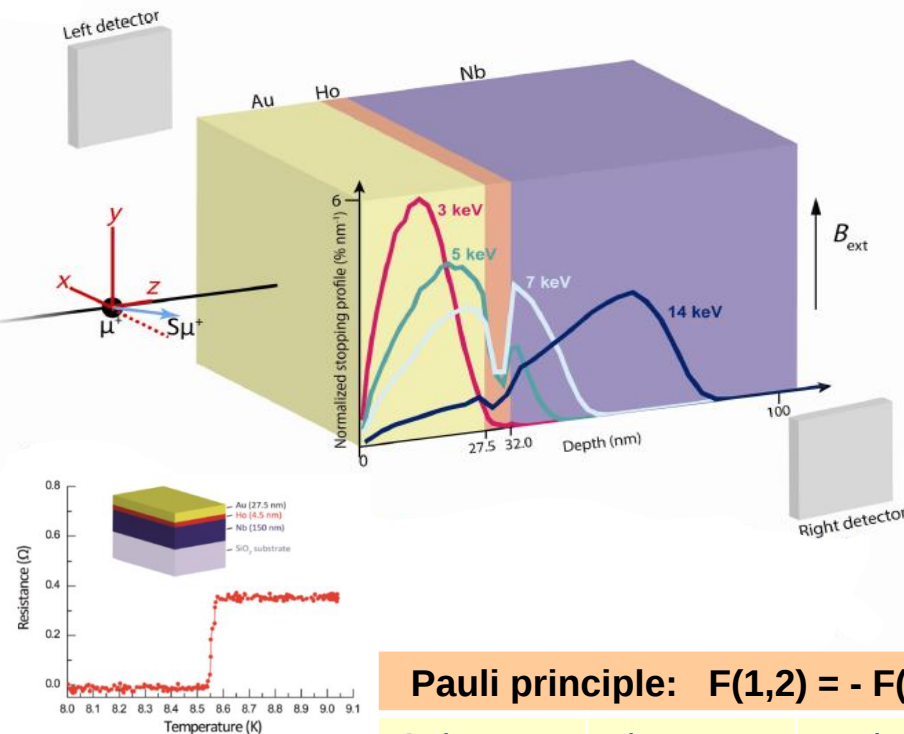
A. Drew et al. *Nature Materials* (2009)
L. Schultz et al. *Nature Materials* (2011)

Intrinsic Paramagnetic Meissner Effect Due to s-Wave Odd-Frequency Superconductivity

A. Di Bernardo,¹ Z. Salman,² X. L. Wang,³ M. Amado,¹ M. Egilmez,⁴ M. G. Flokstra,⁵ A. Suter,² S. L. Lee,⁵ J. H. Zhao,³ T. Prokscha,² E. Morenzoni,² M. G. Blamire,¹ J. Linder,⁶ and J. W. A. Robinson^{1,*}

NM/FM/SC

Cambridge-PSI-CAS-Sharjah-St.Andrews-NTNU



Spin singlet n_s : Meissner effect (diamagnetic)

$$\vec{J} = -\frac{e^2}{mc} n_s \vec{A}$$

Spin triplet component n_t :
paramagnetic Meissner effect
(increase of magn. flux instead of decrease)

$$\vec{J} = -\frac{e^2}{mc} (n_s - n_t) \vec{A}$$

Singlet and triplet components have
different decay lengths

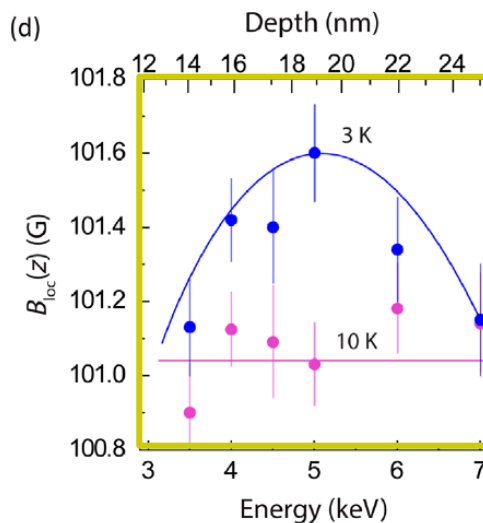
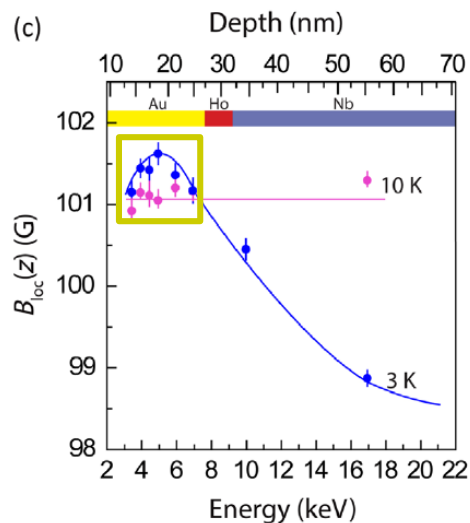
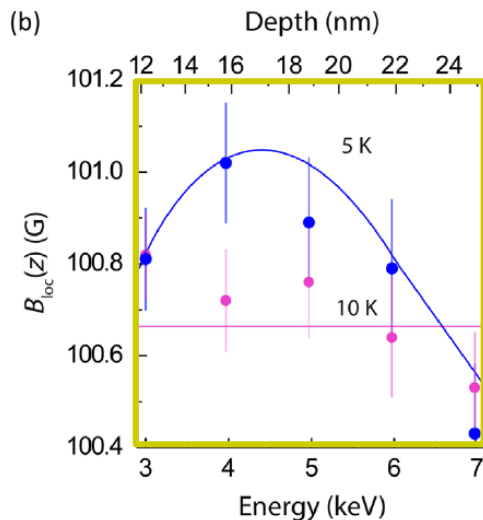
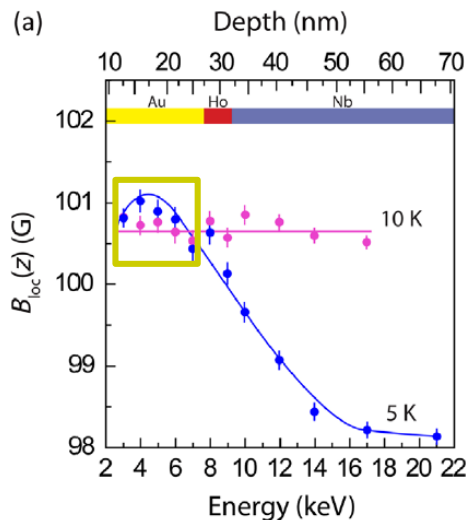
S. Mironov, A. Mel'nikov, and A. Buzdin, *Phys. Rev. Lett.* **109**, 23700 (2012)

V. L. Berezinskii *JETP* 20, 287 (1974)

J. Linder, A. Balatsky, *Rev. Mod. Phys.* (2019)

Magnetic field profiles

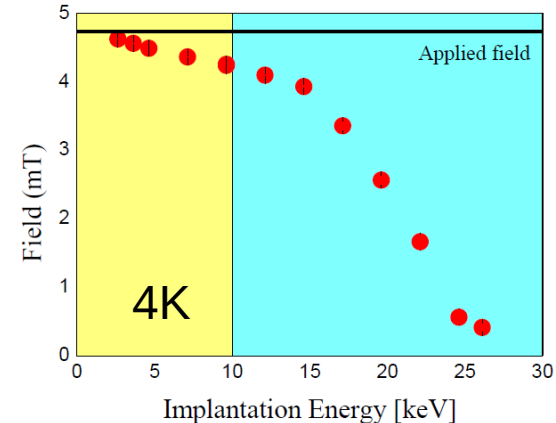
N/FM/SC



A. di Bernardo et al., Phys. Rev. X. 5, 041021 (2015)

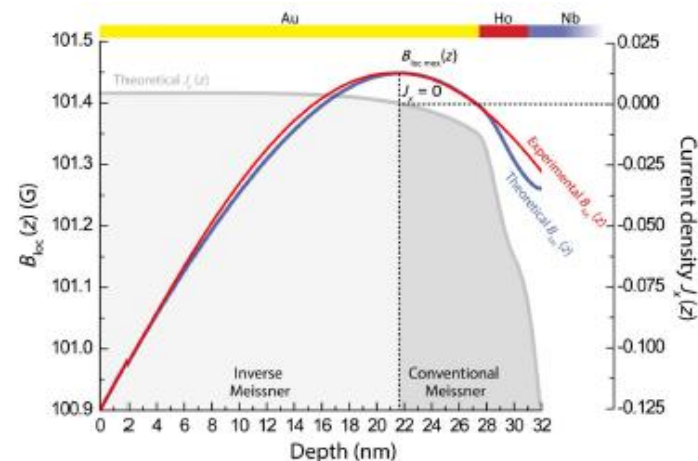
N/SC Al/Nb

Al(90nm)/Nb(270nm)



E. M et al., unpublished

Calculated field profile



Summary and outlook

- Some selected examples of use of μ SR in the study of magnetic and superconducting properties of bulk materials and thin films and heterostructures
- Not shown: many things
 - Applications in chemistry, soft matter, semiconductors (muonium spectroscopy), defect and diffusion studies, (quantum) impurities...
 - Resonance experiments or experiments with external stimulus, decoupling...
- μ SR now a standard technique making increasing significant contribution in Condensed Matter, Material Science research and other fields
- Synergy and complementary with other probes and facilities: neutrons, photons, β -NMR ... and with macroscopic techniques: transport, magnetization, thermodynamic measurements,...
- Increasing interest (> 200 proposals/year at PSI, overbooking factor > 2.5)
- New techniques, such as low energy μ SR are extending the already broad range of applications, new tools: high pressure, high field and low temperature instruments, external stimuli
- The new μ SR facility at RAON has great potential to address a new user community and enlarge the worldwide existing community (complementary to pulsed source at J-PARC)

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