



All in a spin – An introduction to muon spectroscopy

Adrian Hillier
ISIS Muon Group

Plan

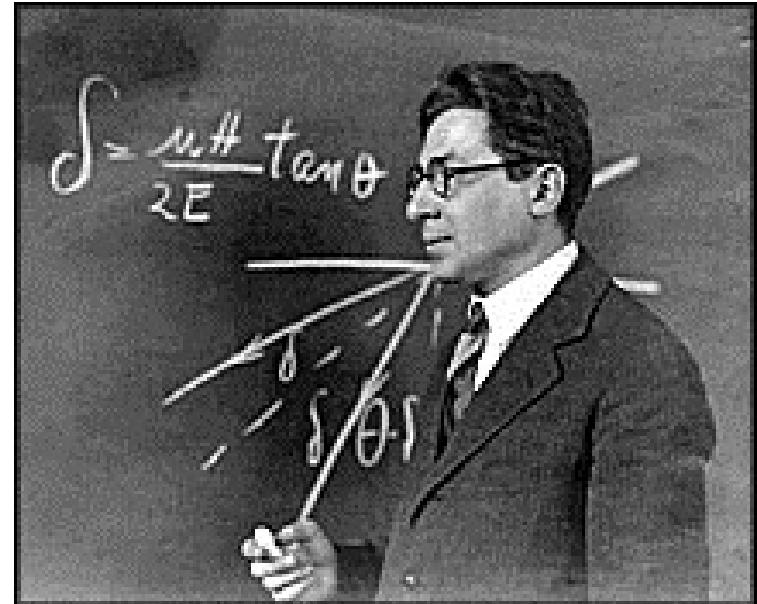
1. An Introduction to Muons
2. The μ SR technique
3. The ISIS μ SR Facility
4. Condensed matter studies with muons
5. Muons elsewhere in the world



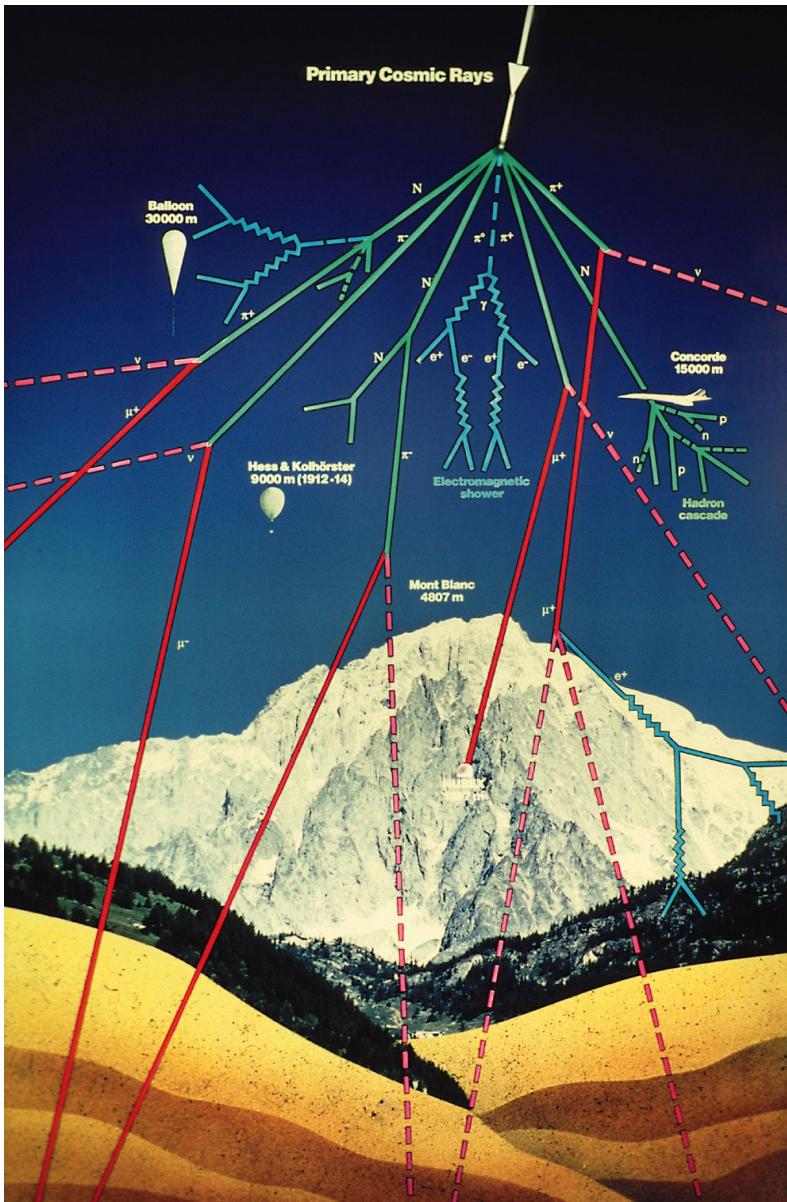
1. An Introduction to Muons

‘Who ordered that?!’

*I.I. Rabi, Physics Nobel prize
winner, 1944*



Discovery of the muon

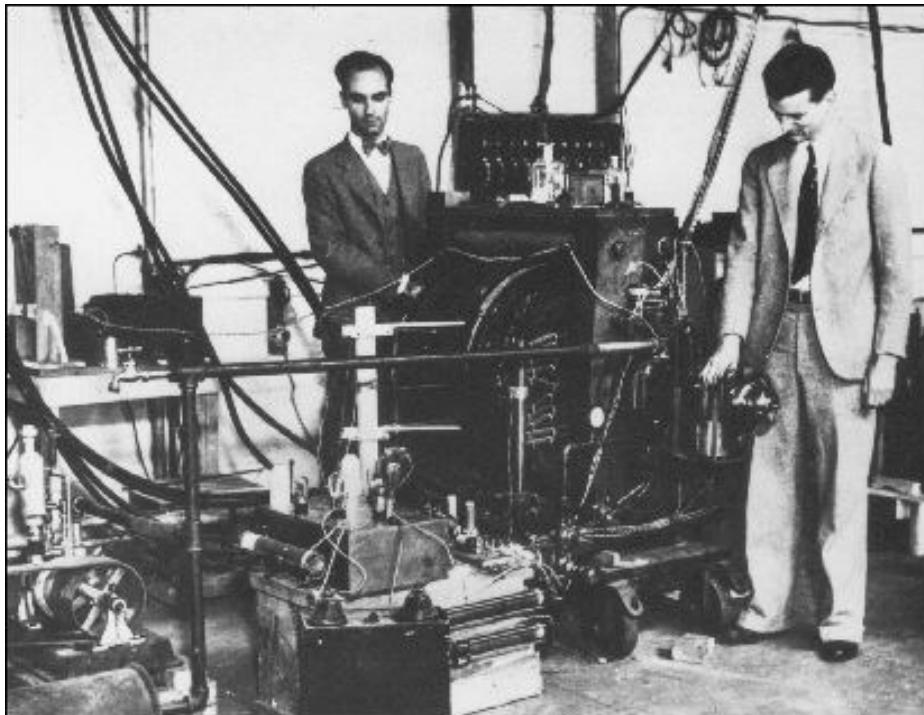


Cosmic rays

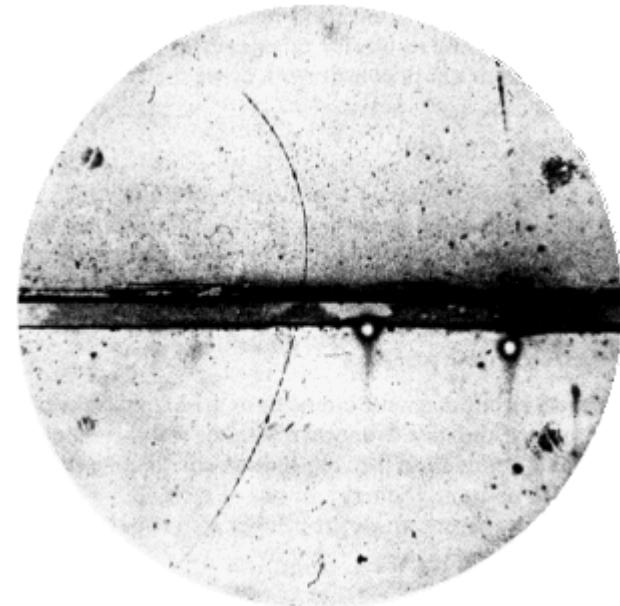
- Discovered 1912 - Victor Hess (Nobel Prize 1936)
- 87% protons, 12% He, 1% heavier nuclei

Discovery of the muon

Carl Anderson - Caltech
1936 Nobel prize for discovery of positron



*Carl Anderson and Seth
Neddermeyer with cloud chamber
courtesy Caltech Archives*



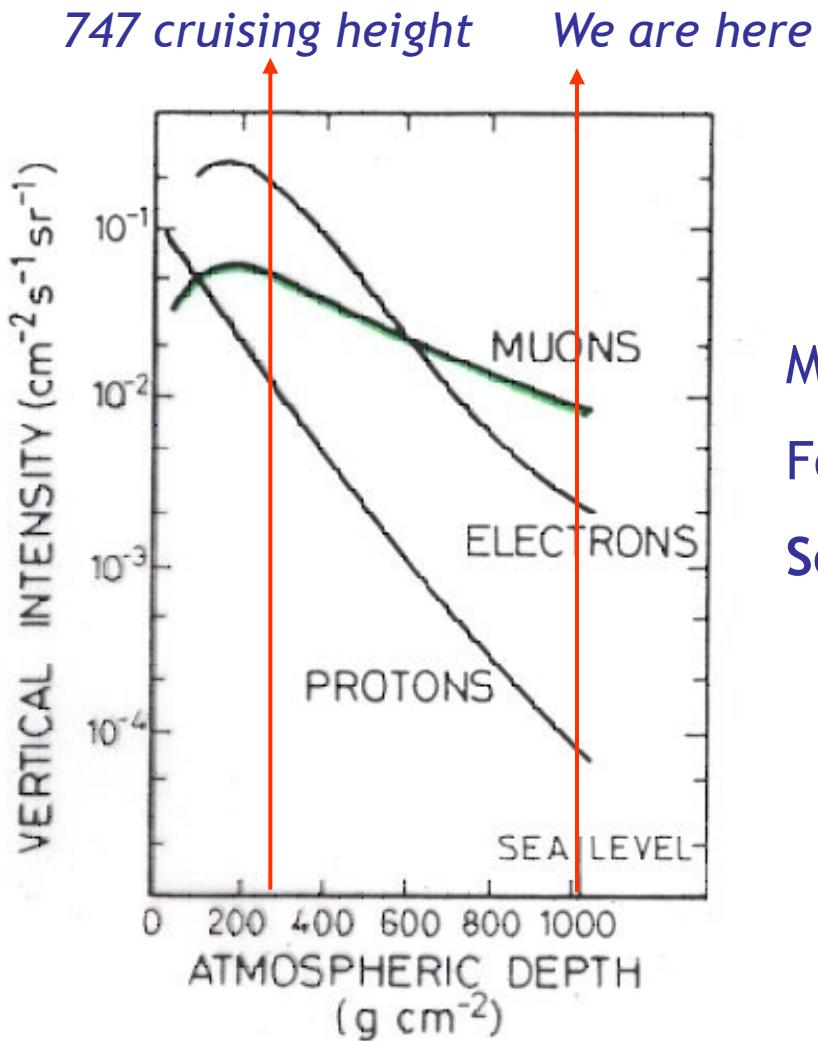
1937, with Seth
Neddermeyer - charged
particle 200 x electron
mass - the muon

Discovery of the muon



Physics at the farm: discovery of the muon.

Muons in cosmic rays



Muons - average energy 4 GeV.

For muons $E > 1 \text{ GeV}$, flux is $1 \text{ cm}^{-2} \text{ min}^{-2}$

So - we get hit by $\sim 1 \text{ muon s}^{-1}$!



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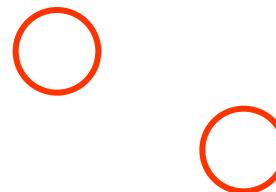
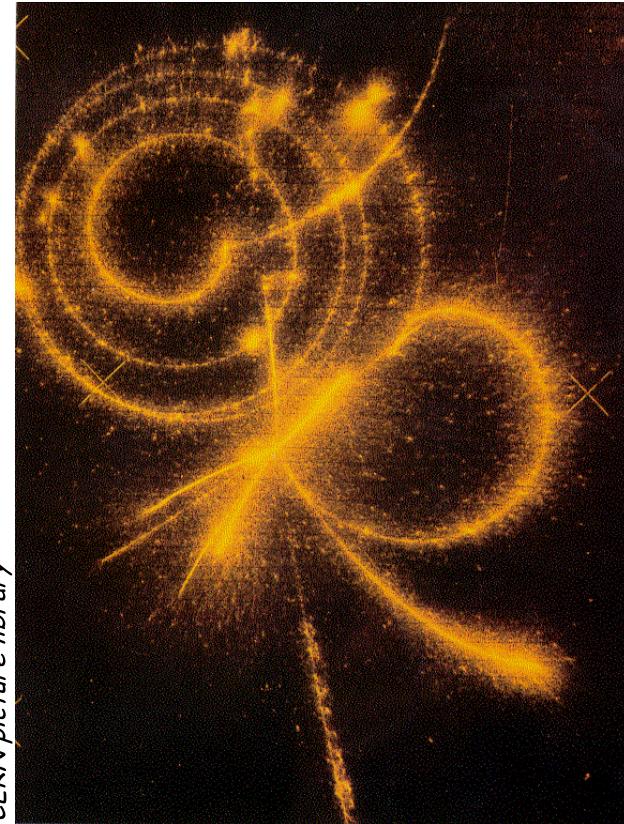
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Muon properties

Muons:

- fundamental, charged particles
- heavy electrons
- spin 1/2
- magnetic moment $3.2 \times m_p$
- mass $0.11 \times m_p$
- produced from pion decays
- lifetime $2.2 \mu s$
- decay into a positron (+ 2xν)

The life of a muon



2. The μ SR technique

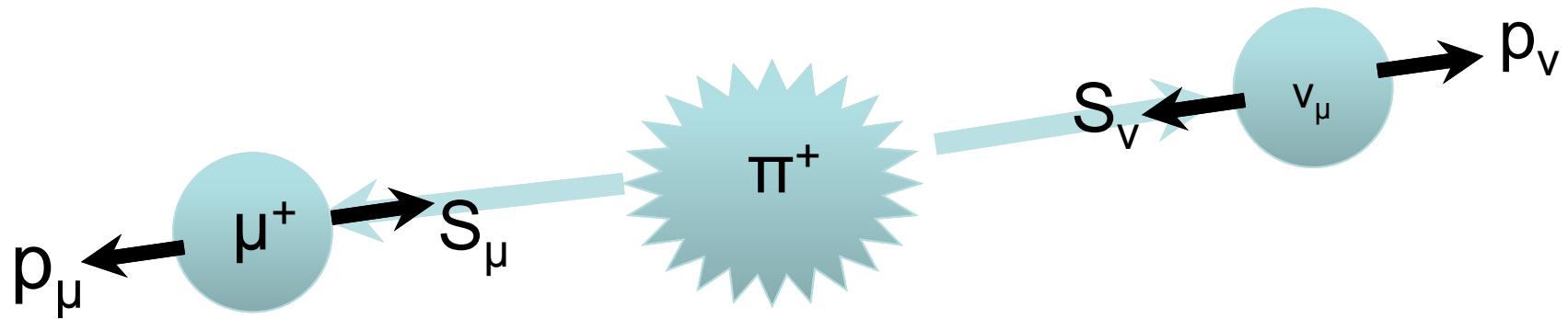
μ^+

μ^+



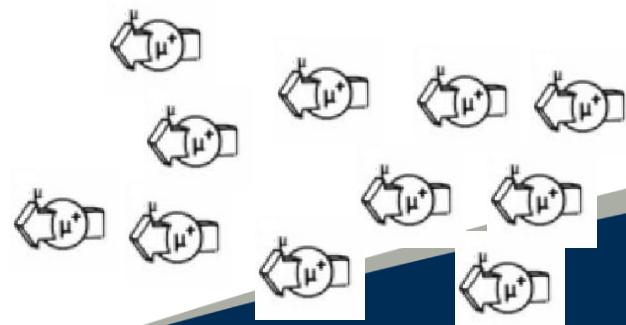
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Muon birth

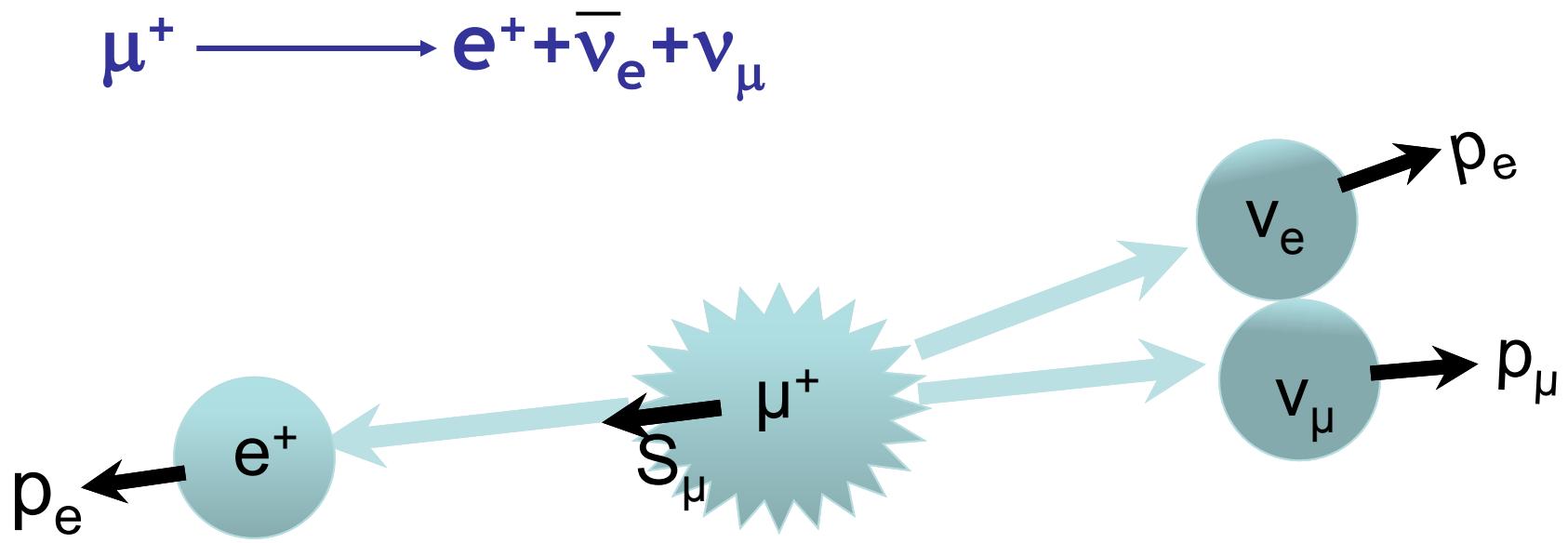


π^+ at rest, $I=0$

Result: a fully spin-polarised muon beam



Muon death



Positrons emitted preferentially in muon spin direction



Muon death

$$W(\theta) = 1 + a \cos \theta$$

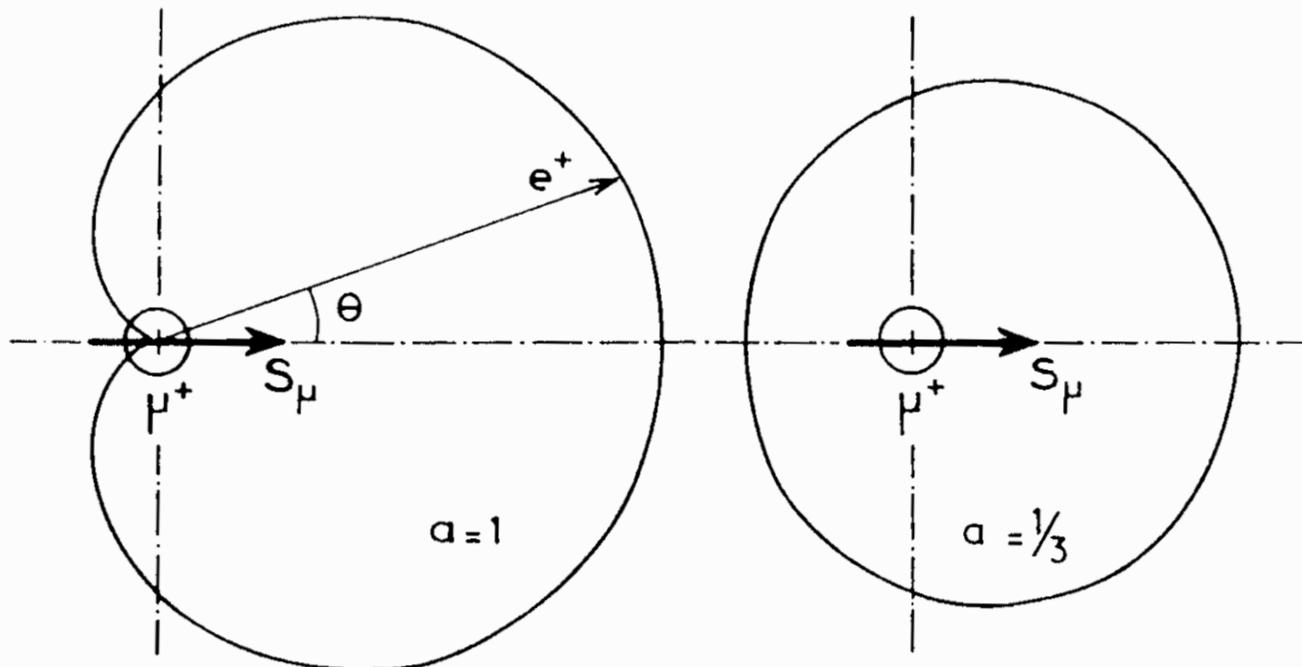


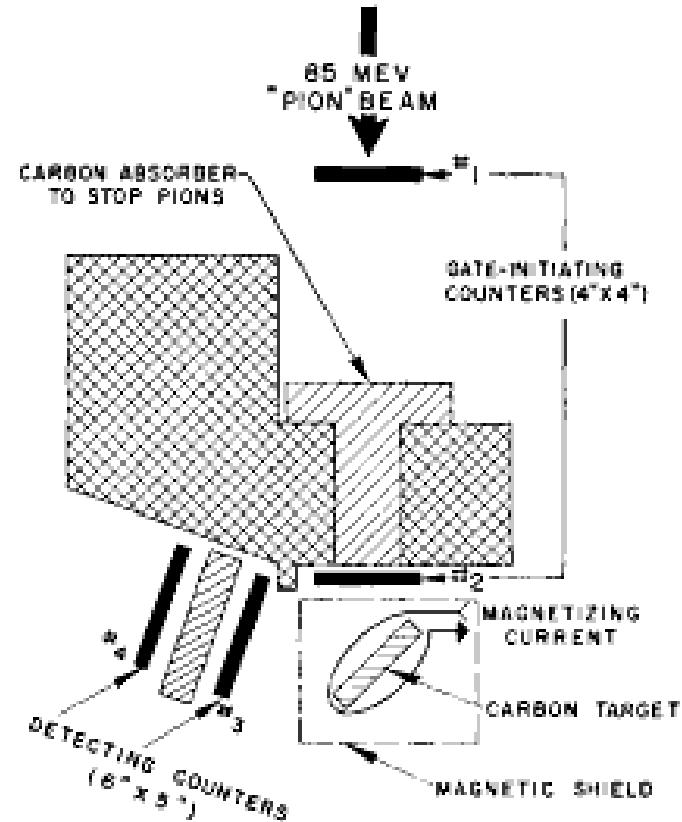
Fig. 1 Angular distribution of positrons for asymmetry parameters 1 and $\frac{1}{3}$

Muon death

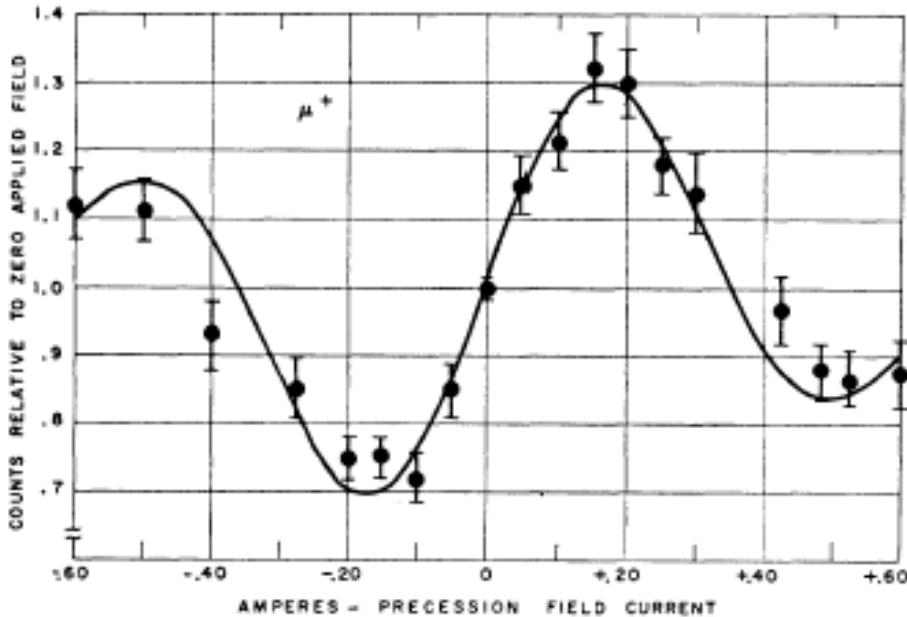
Observations of the Failure of Conservation of Parity and Charge Conjugation in Meson Decays: the Magnetic Moment of the Free Muon*

RICHARD L. GARWIN,[†] LEON M. LEDERMAN,
AND MARCEL WEINRICH

*Physics Department, Nevis Cyclotron Laboratories,
Columbia University, Irvington-on-Hudson,
New York, New York*



Muon death



'It seems possible that polarised positive and negative muons will become a powerful tool for exploring magnetic fields in nuclei, atoms and interatomic regions'.

Garwin et al., Phys Rev
1957

The muon technique was born!

μ SR . . . muon spin rotation, relaxation and resonance -
or just muon spin research

The μ SR technique

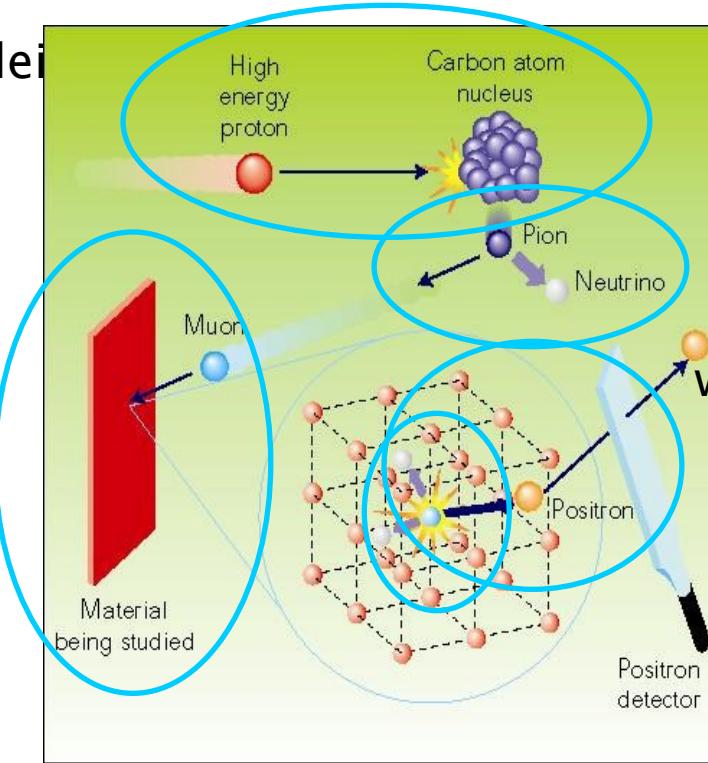
High energy protons

(800 MeV at ISIS)

collide with carbon nuclei
producing pions

Implantation,
(stopped in \sim 1 mm
water)

Muons interact with
local magnetic
environment



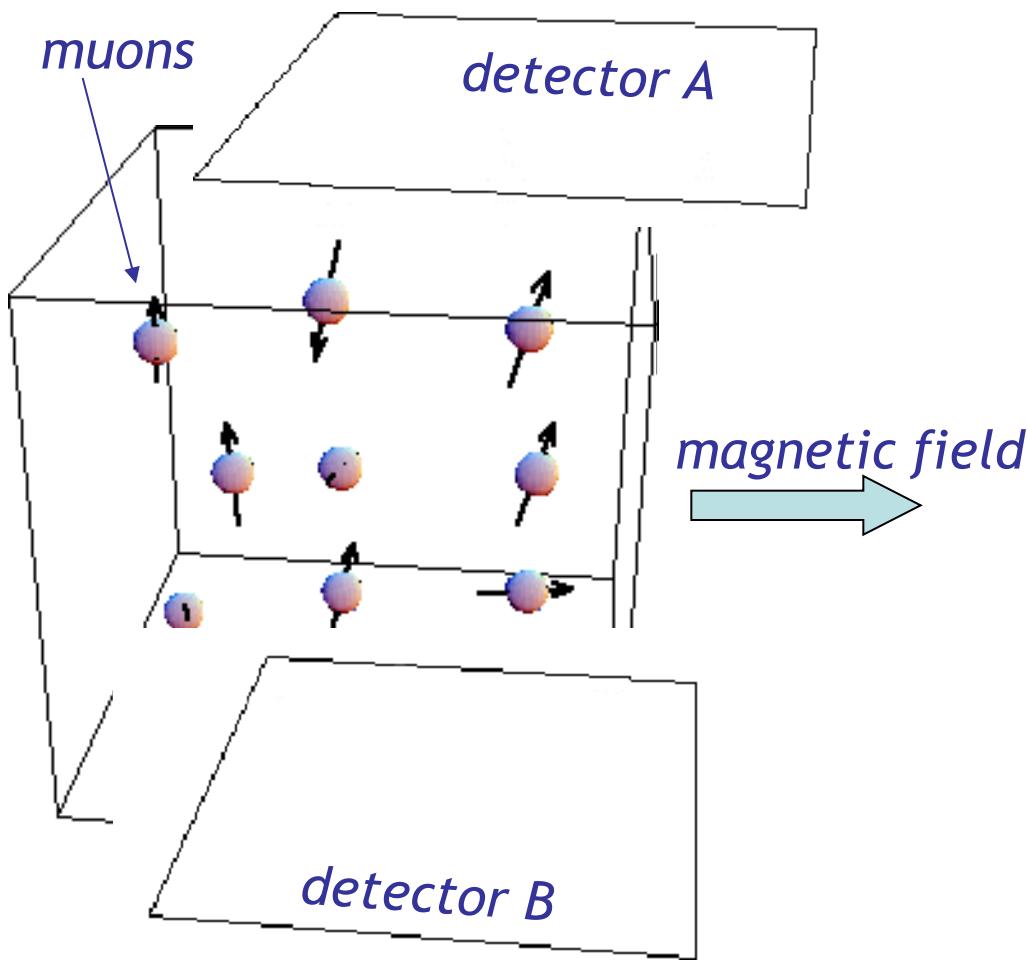
Monitor the positron distribution to infer the muons' polarisation after implantation. Learn about the muons' local environment or the muon behaviour itself.

$\pi^+ \rightarrow \mu^+ + \nu_\mu$
**4 MeV muons are
100% spin
polarised**

Decay, lifetime $2.2\mu\text{s}$
 $\mu^+ \rightarrow e^+ + \nu_e + \nu_\mu$
we detect decay positrons

**The positrons are
preferentially
emitted in muon
spin direction**

The μ SR technique

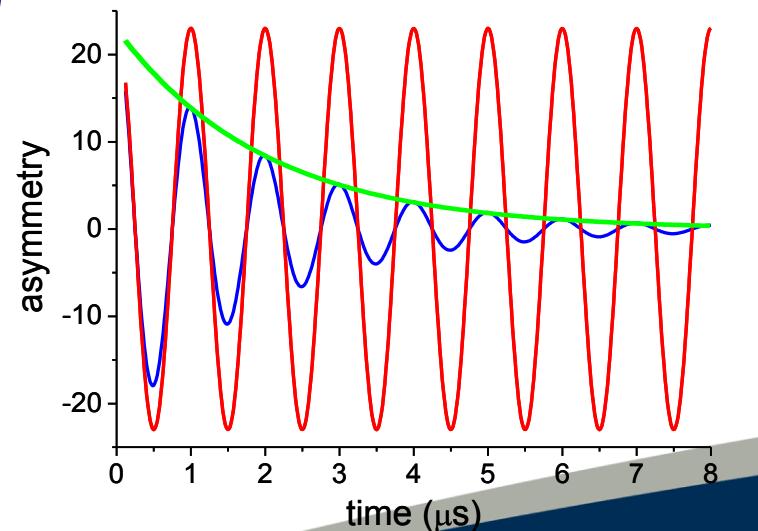


'asymmetry' =

$$\frac{N_A(t) - N_B(t)}{N_A(t) + N_B(t)}$$

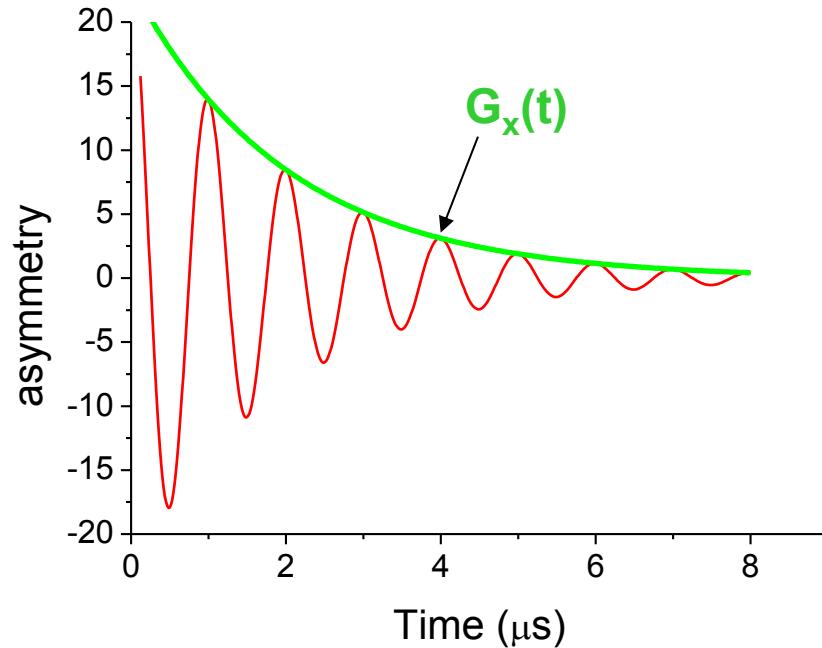
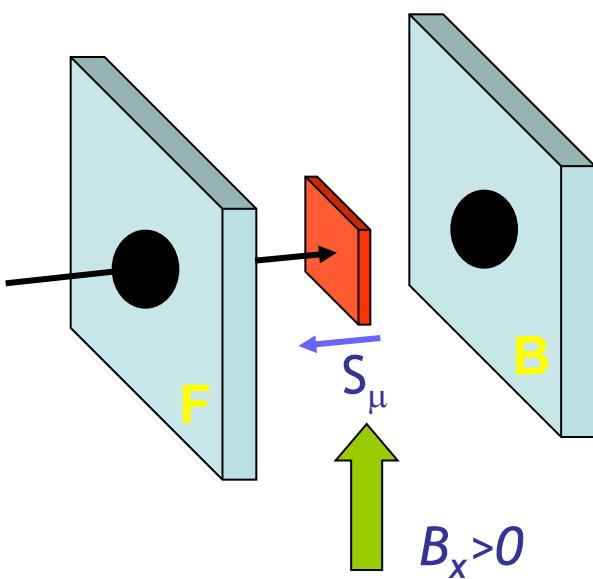
non-relaxing polarisation

relaxing polarisation



The μ SR technique

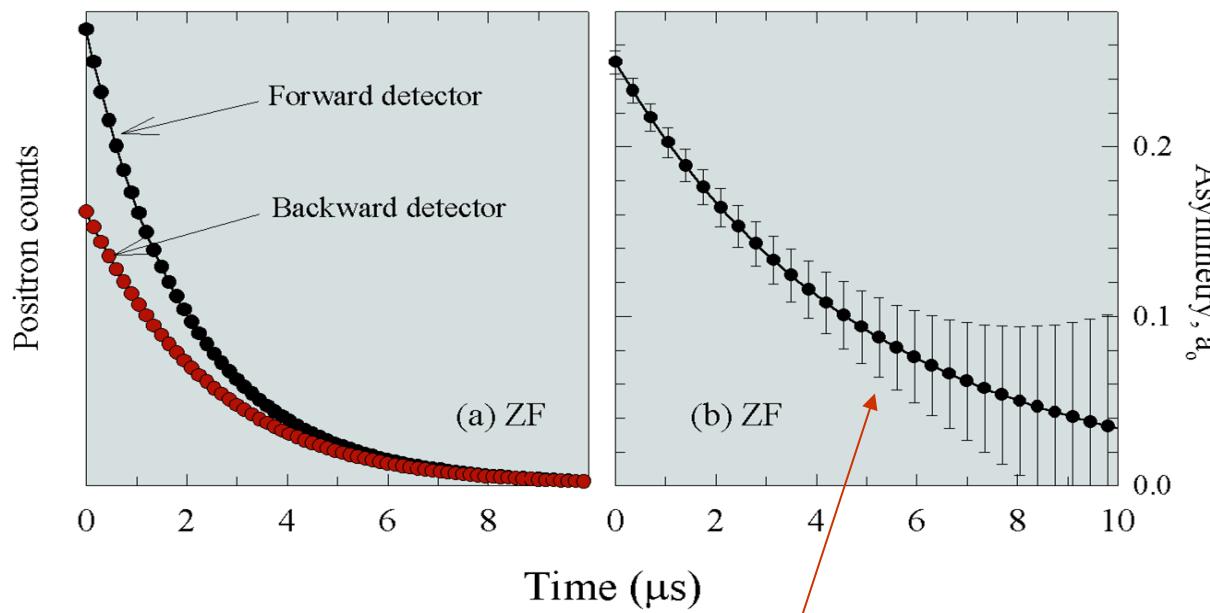
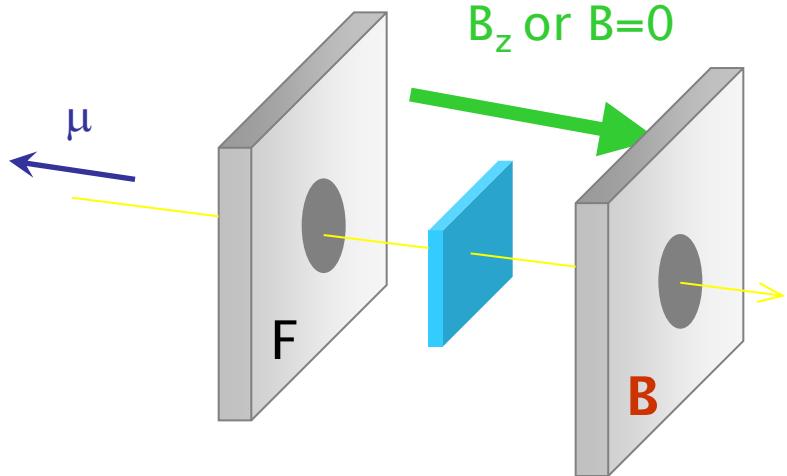
'Transverse' field



$$\text{asymmetry} = \frac{N_F(t) - N_B(t)}{N_F(t) + N_B(t)} = a_0 G_x(t) \cos(\omega t)$$



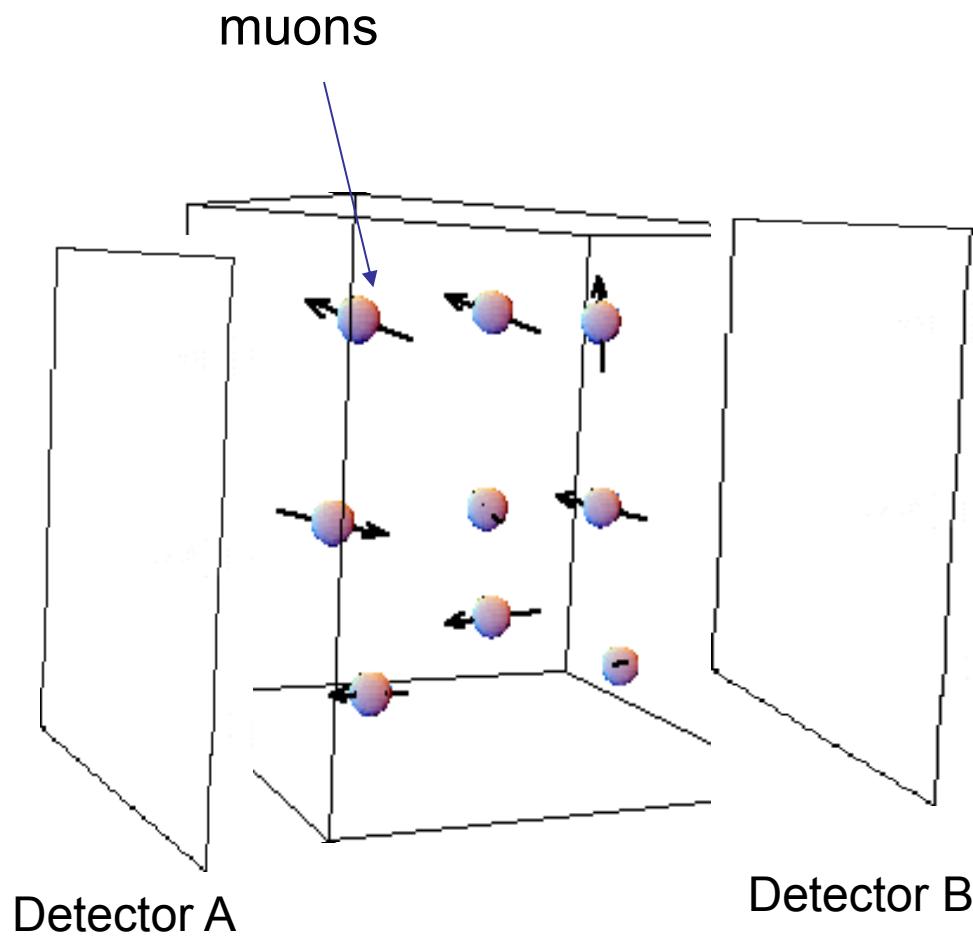
The μ SR technique



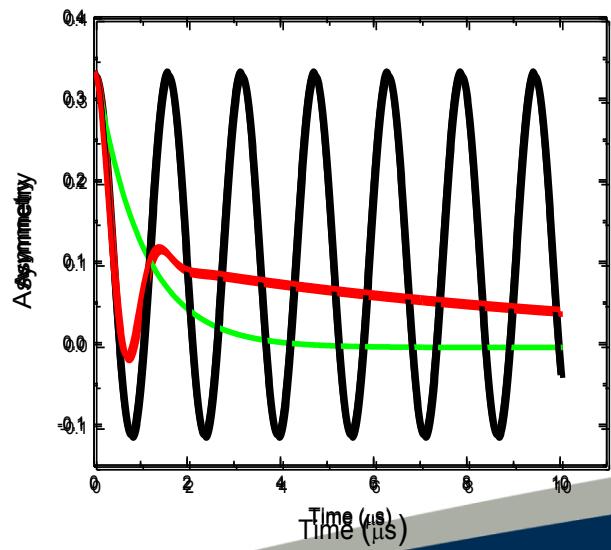
$$R_z(t) = \frac{F(t) - B(t)}{F(t) + B(t)} = a_0 G_z(t)$$



The μ SR technique

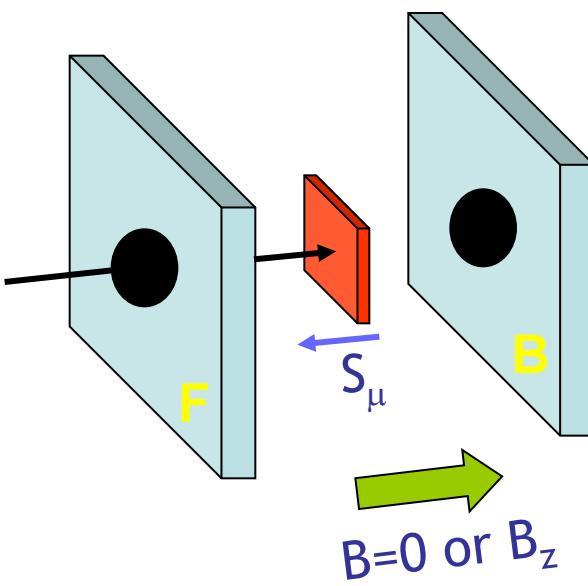


- Precessing polarisation
- Precessing and relaxing polarisation
- Relaxing signal

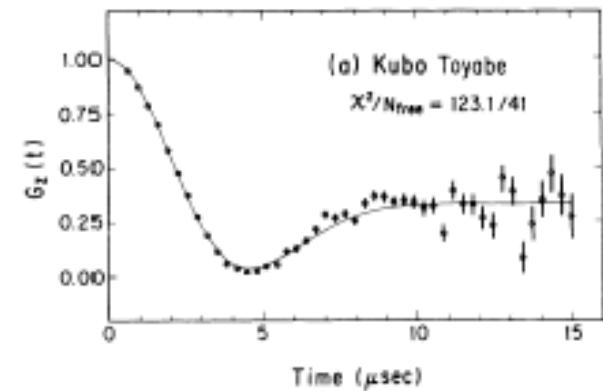
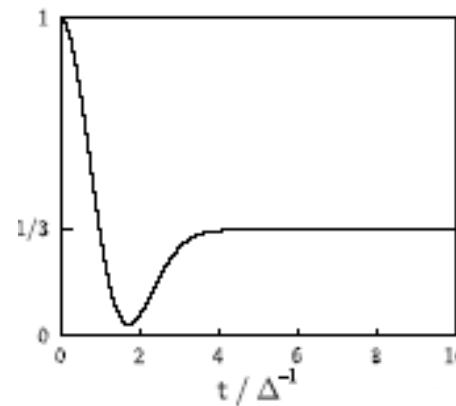


The μ SR technique

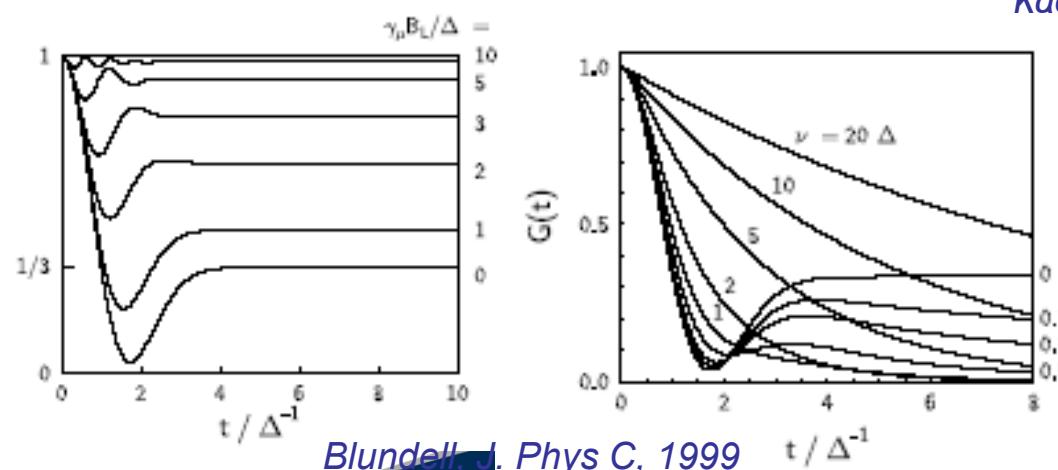
Zero or ‘longitudinal’ field



$$\text{asymmetry} = \frac{N_F(t) - N_B(t)}{N_F(t) + N_B(t)} = a_0 G_z(t)$$

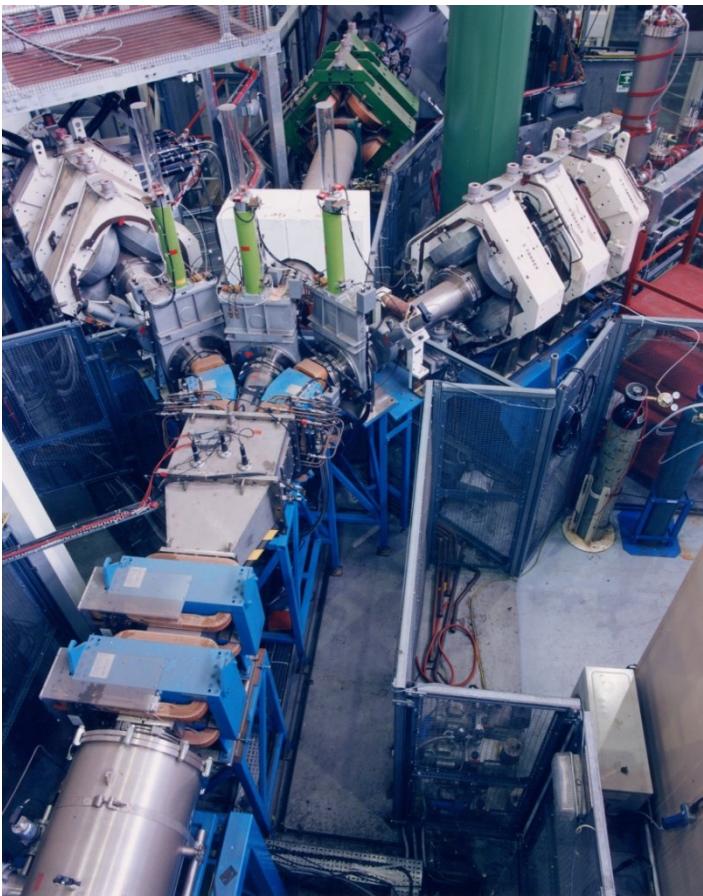


Kadono et al., PRB 1989



Blundell, J. Phys C, 1999

3. The ISIS Muon Facility



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The ISIS Facility



The Rutherford Appleton Laboratory

The ISIS Facility



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Spallation neutron source

800 MeV proton beam

Neutrons produced for 25 instruments

7 muon experimental areas

2000 users/yr

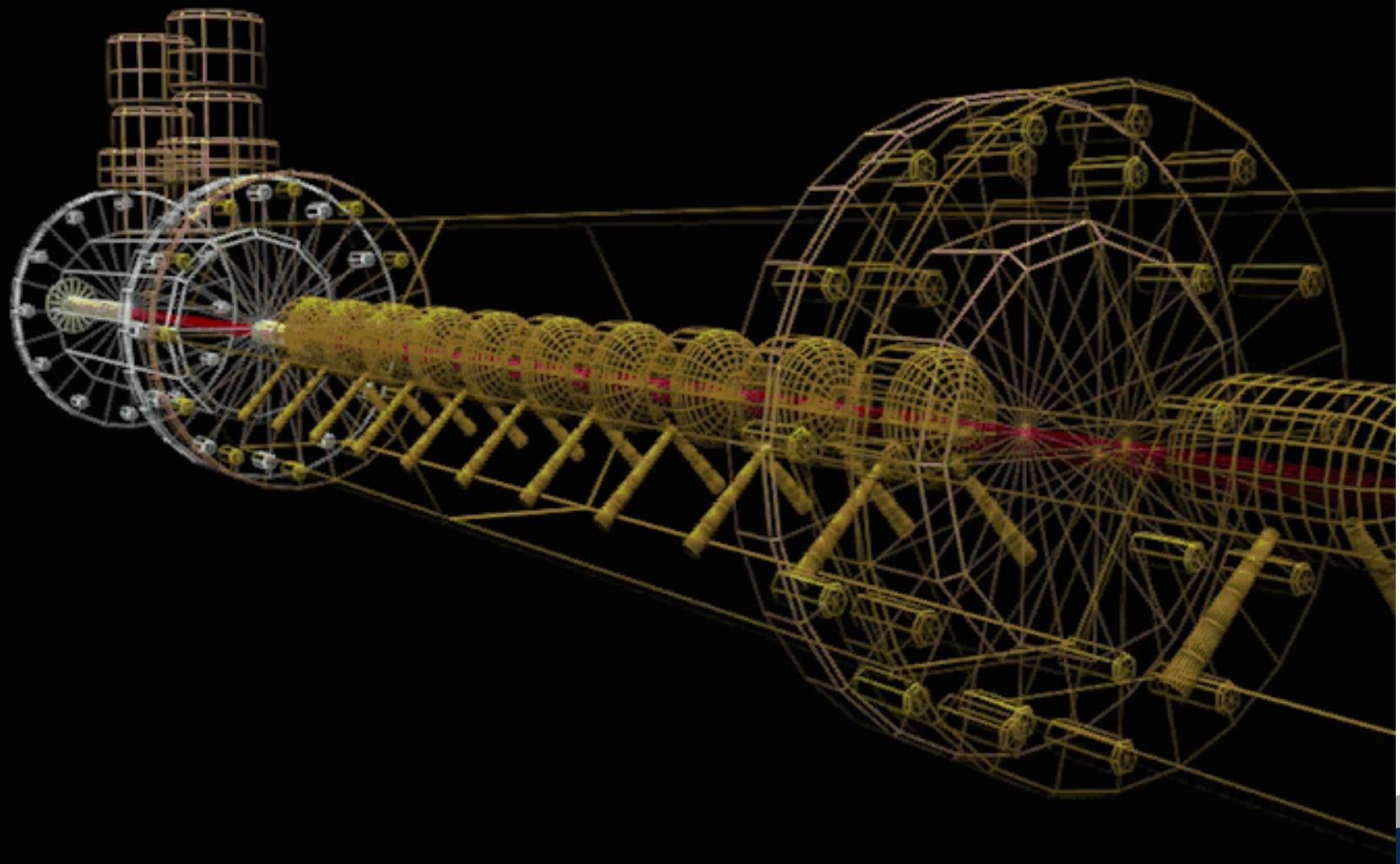
~800 experiments/yr

~500 publications/yr

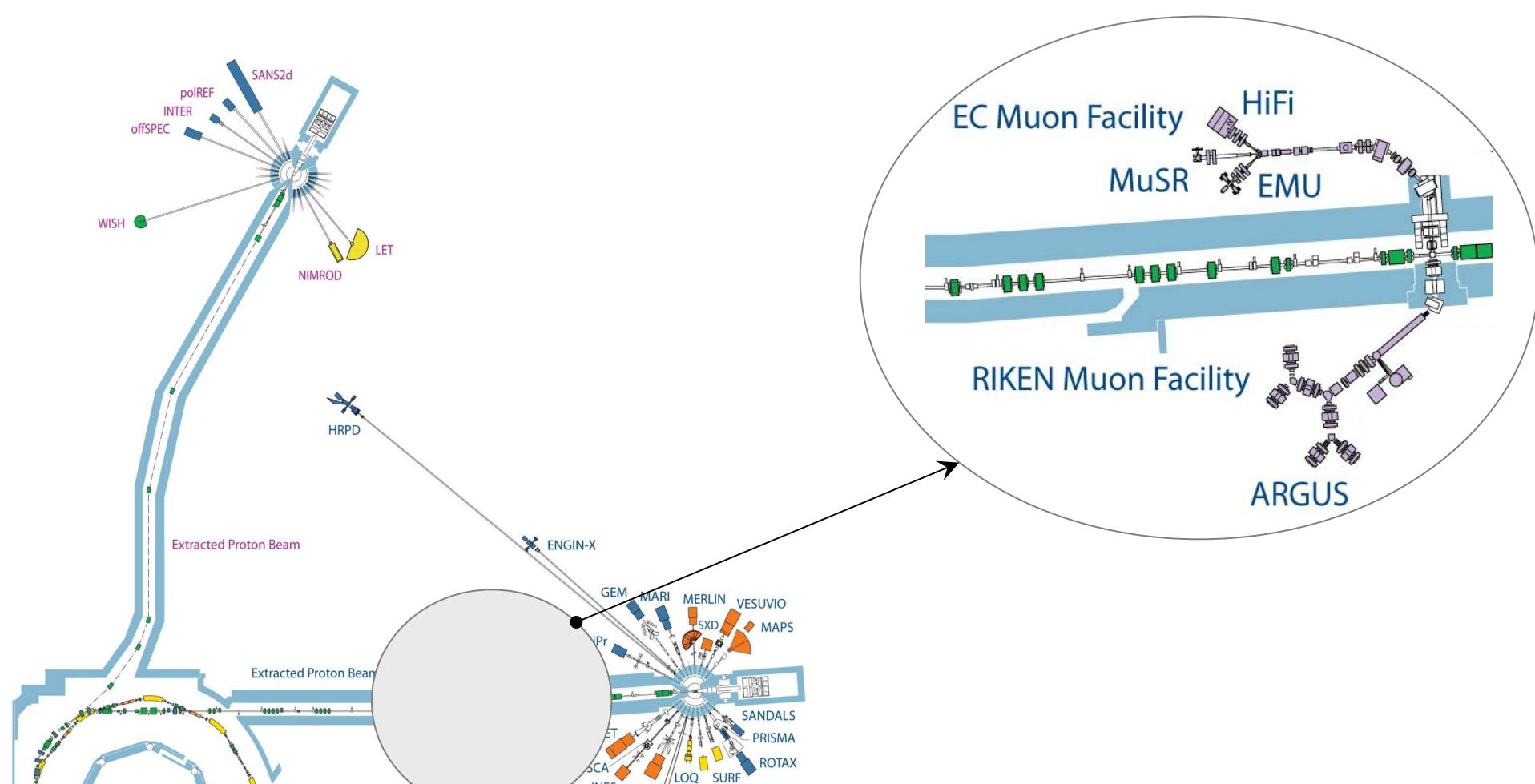


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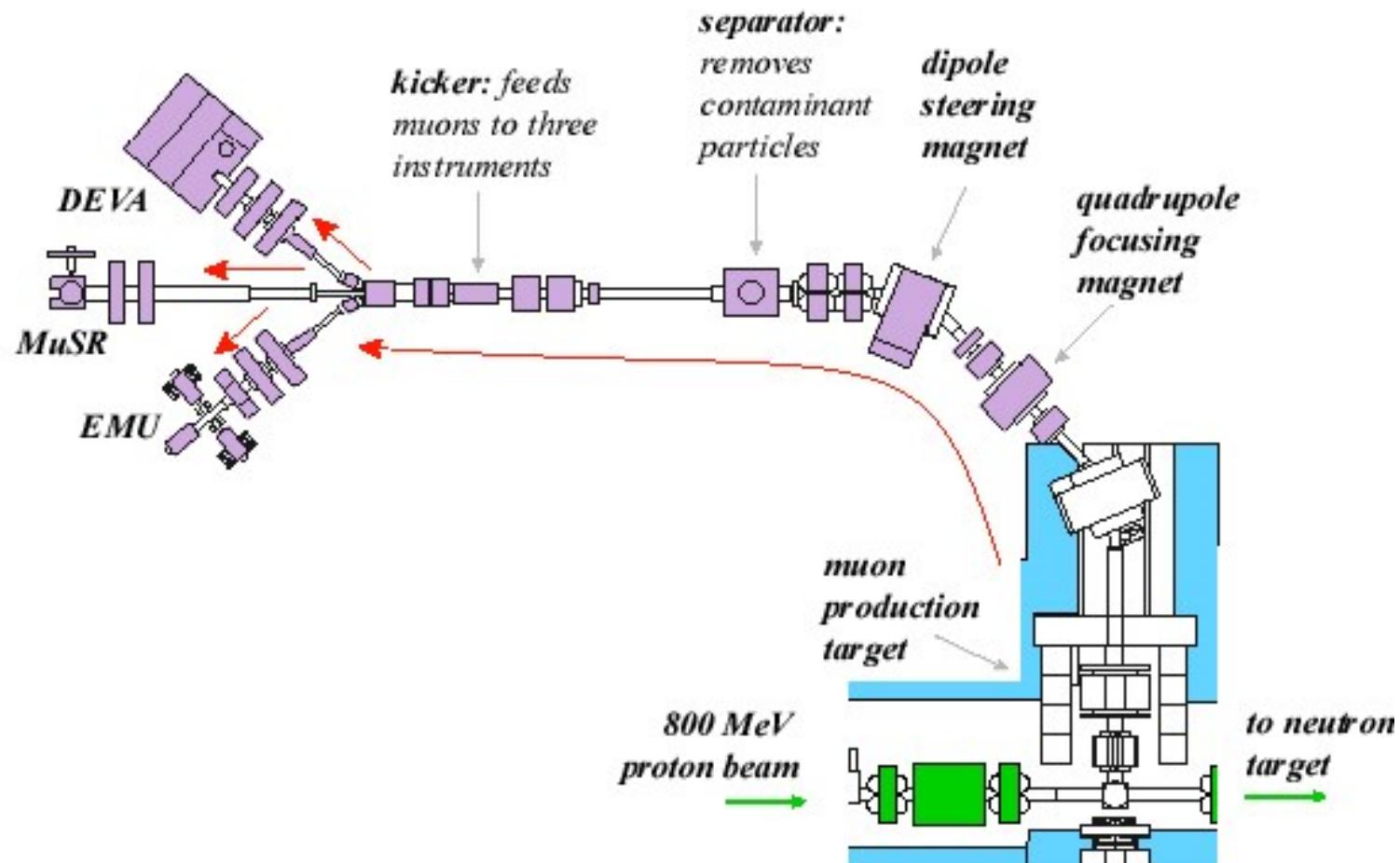
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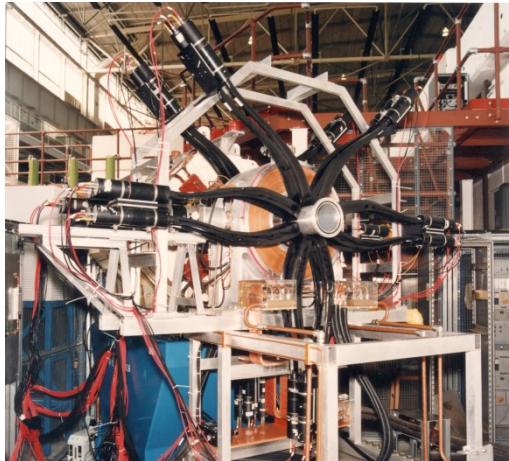
Muons at ISIS



Muons at ISIS

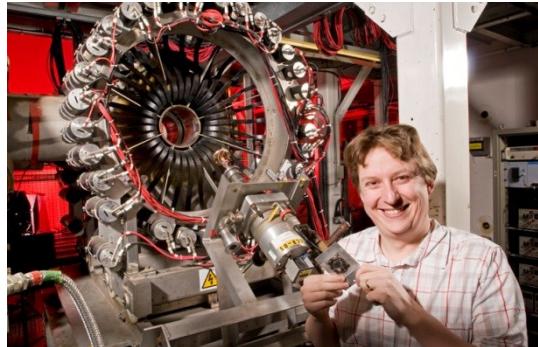


Muons at ISIS



EMU

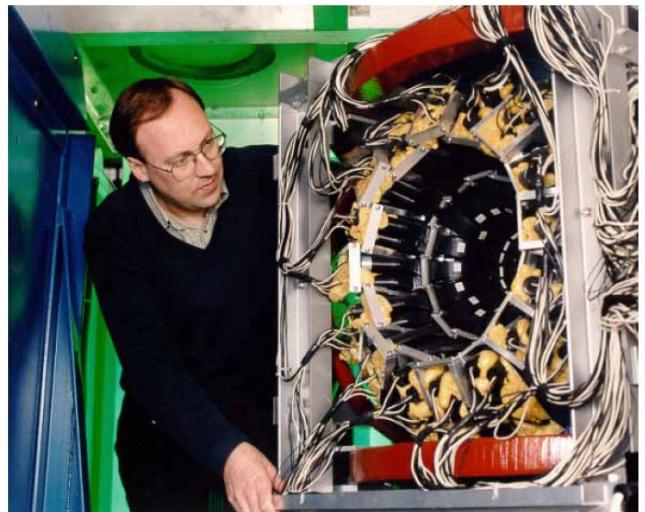
- Fields: 0G - 5 T
- Temperatures: 30 mK - 1500 K
- Pressure: up to 6.4kbar
- + and - muons



MuSR



ARGUS



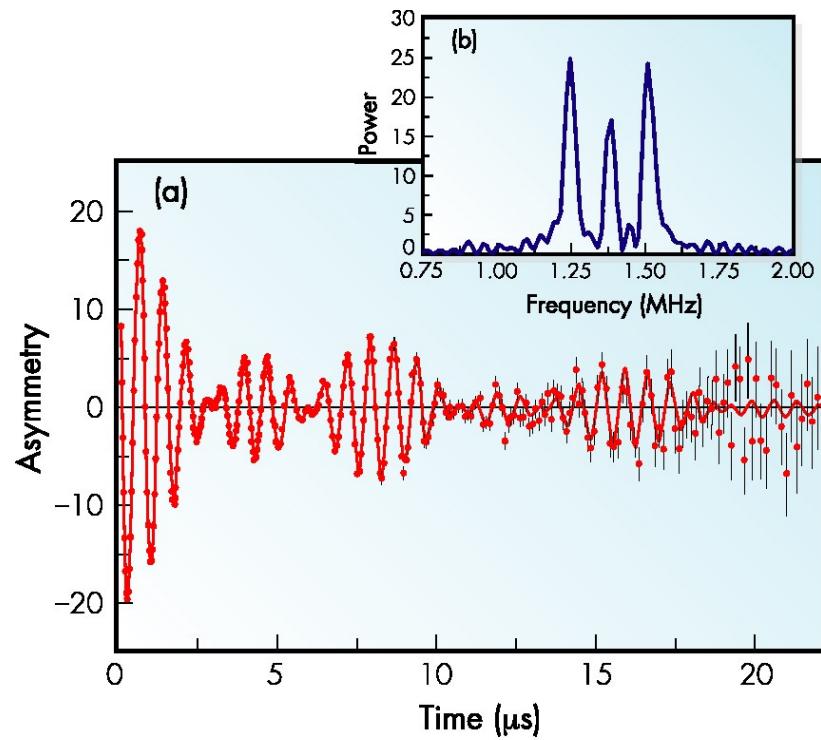
HiFi

- Gas/liquid samples
- Pulsed stimuli, e.g. light, E/B-fields, RF



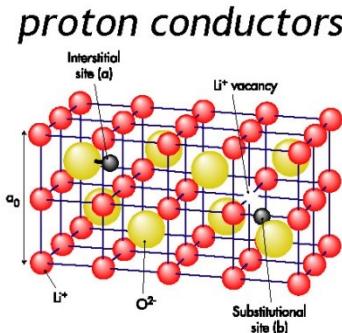
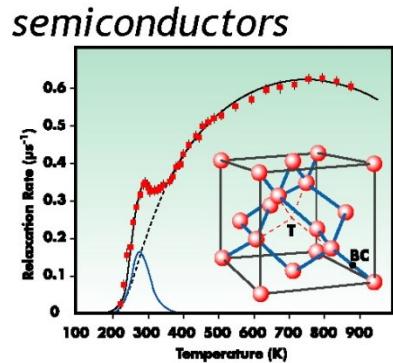
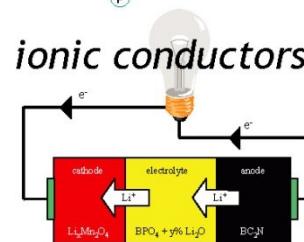
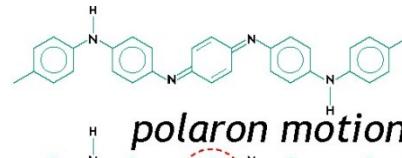
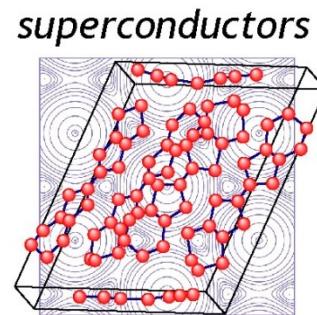
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4. Science with muons

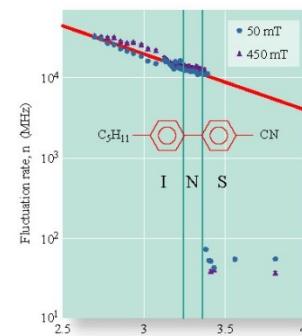


Muon science areas

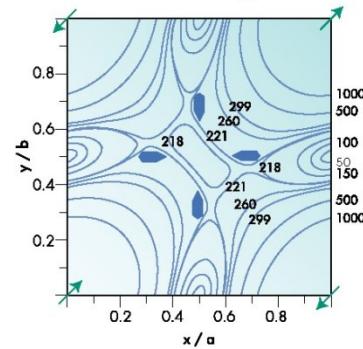
- Muons as passive probes in superconductivity, magnetism, molecular dynamics, charge transport.
- Muons as active probes: proton analogues in semiconductors, proton conductors, light particle diffusion, etc.



molecular dynamics

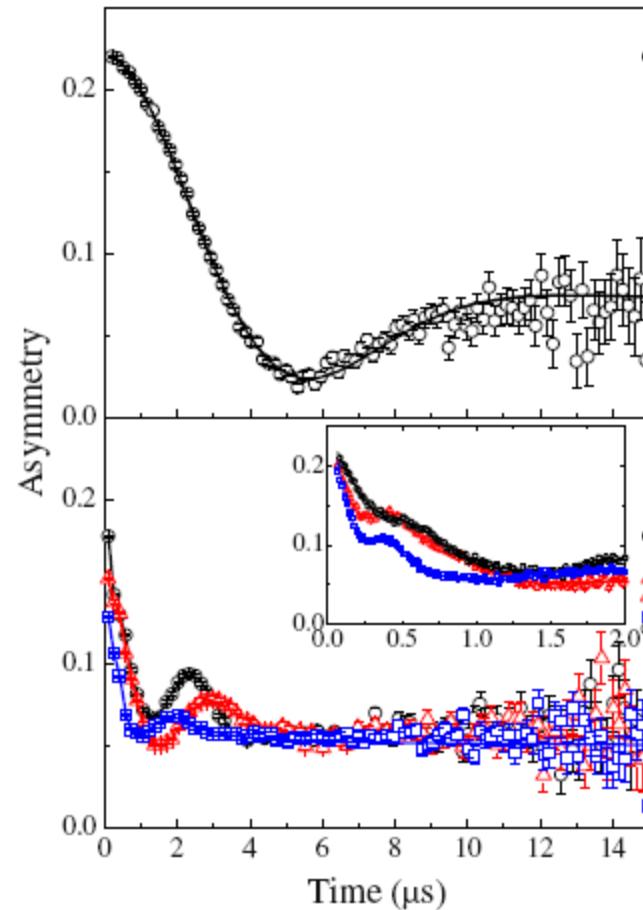
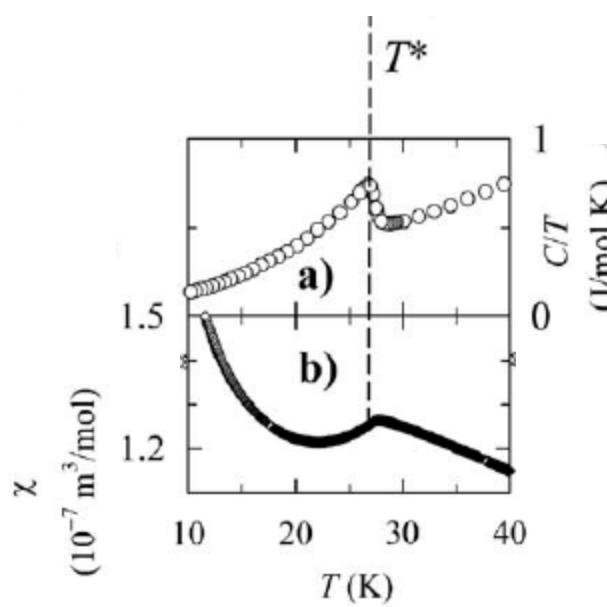


magnetism



Magnetism

CeRu₂Al₁₀

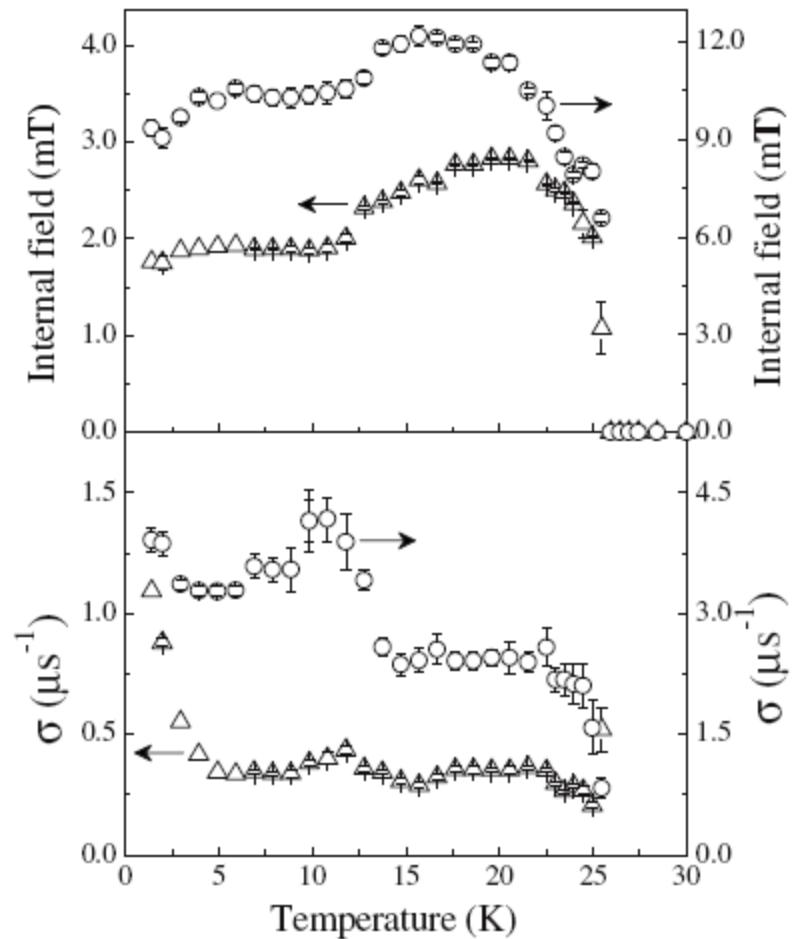


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Magnetism

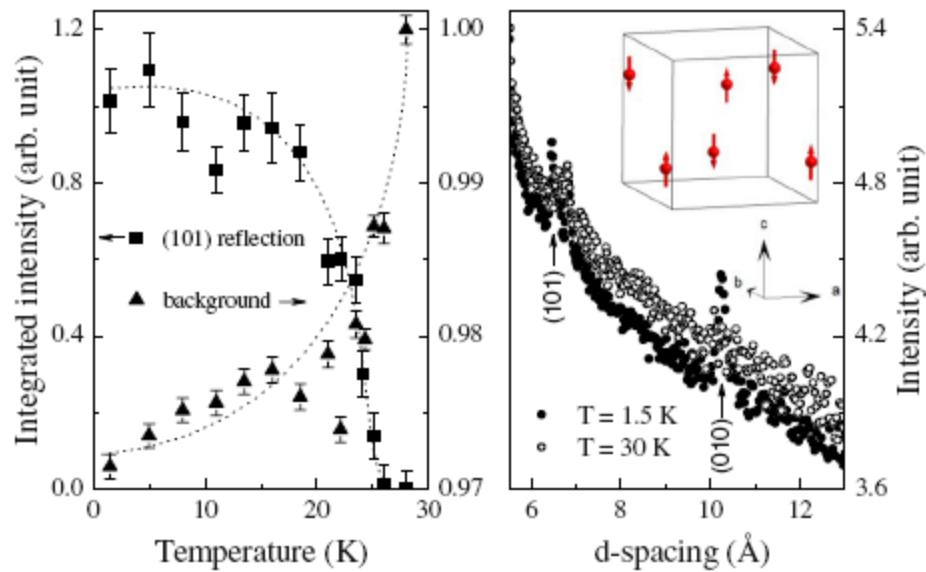
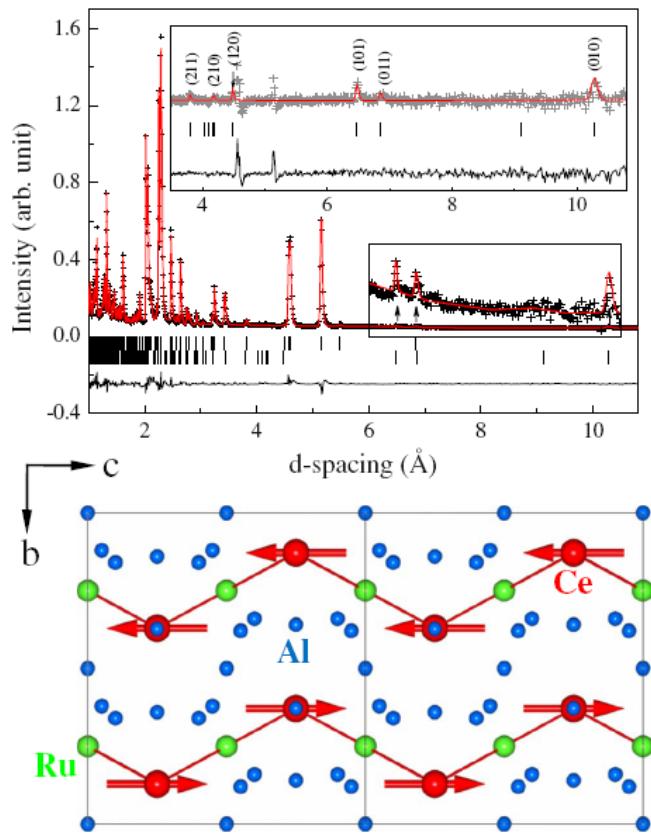
CeRu₂Al₁₀



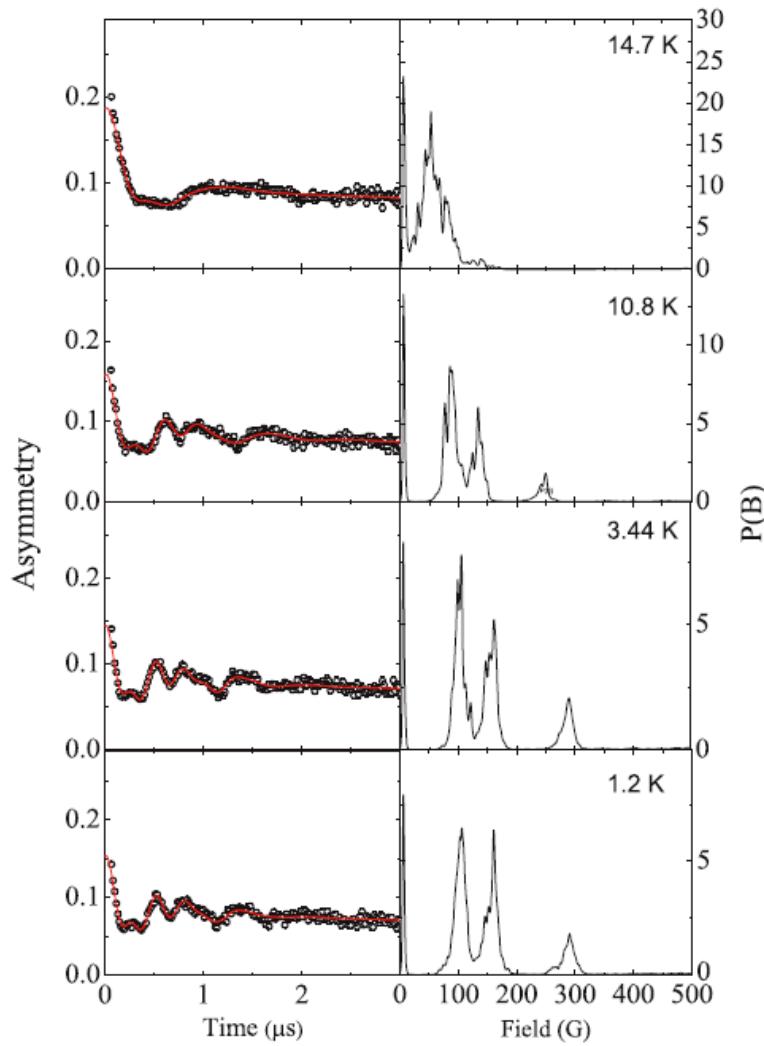
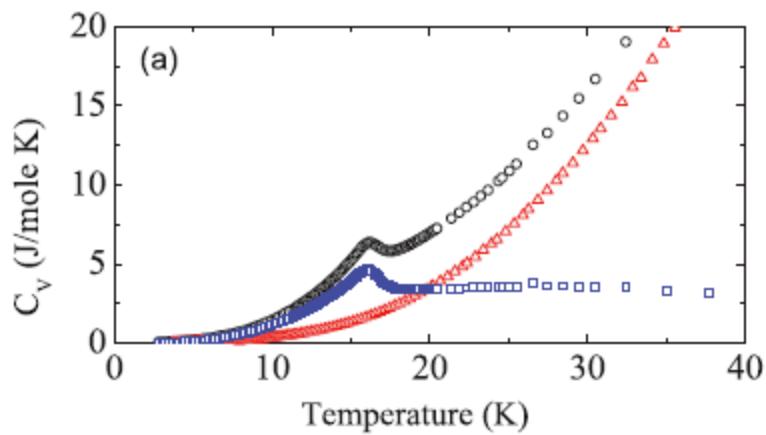
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Magnetism

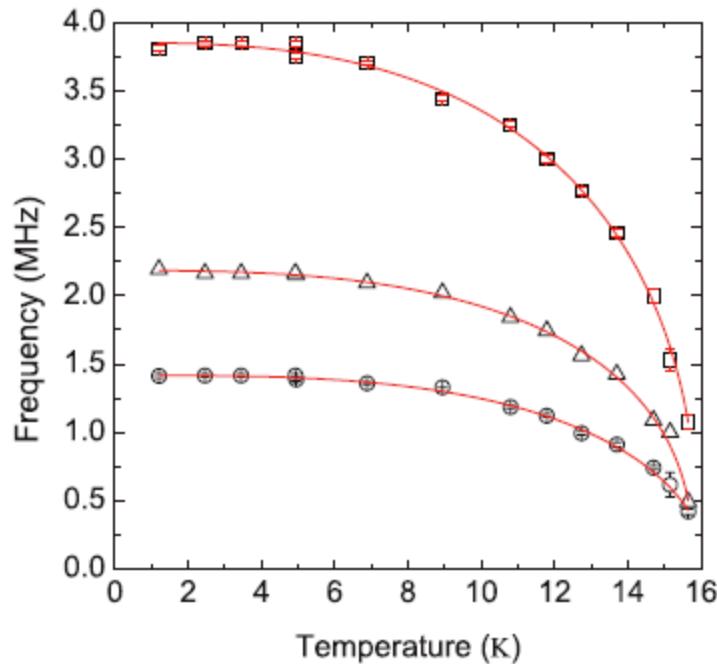
CeRu₂Al₁₀



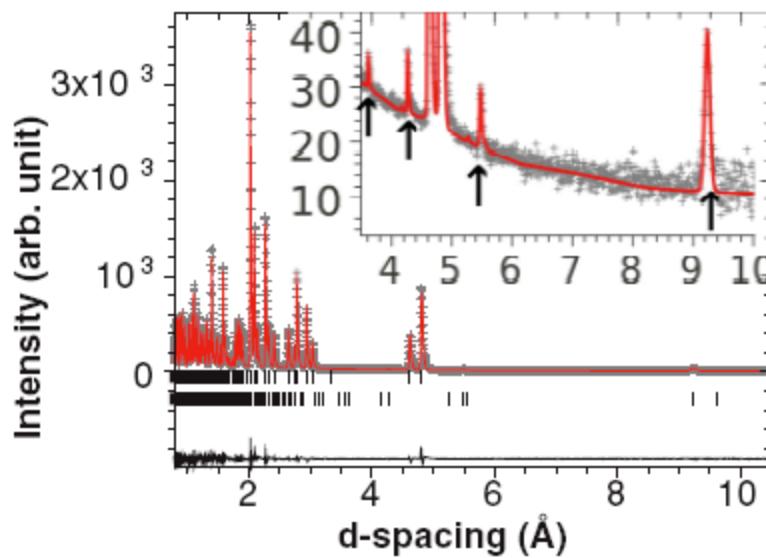
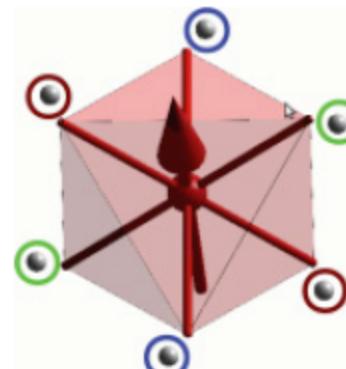
Magnetism



Magnetism



$\text{Sr}_3\text{ZnRhO}_6$

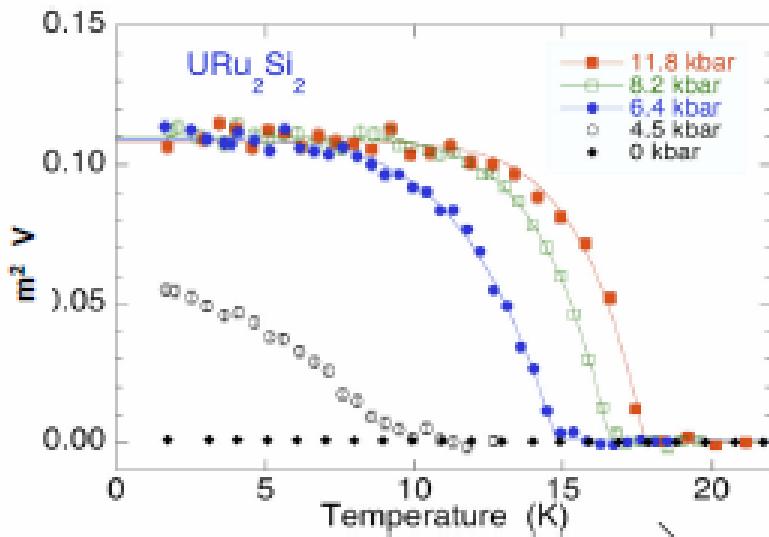


Magnetism

Example URu_2Si_2

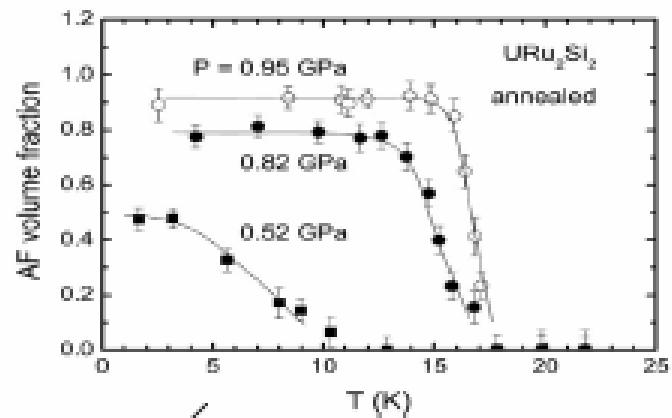
Neutron scattering:

F. Bourdarot et al., condmat/0312205



Muon Spin Rotation:

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



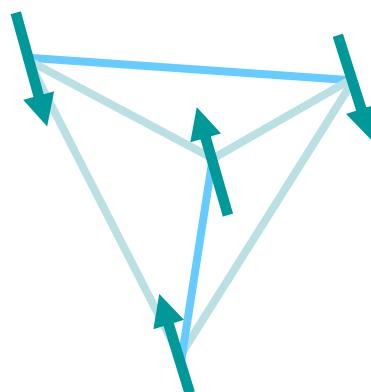
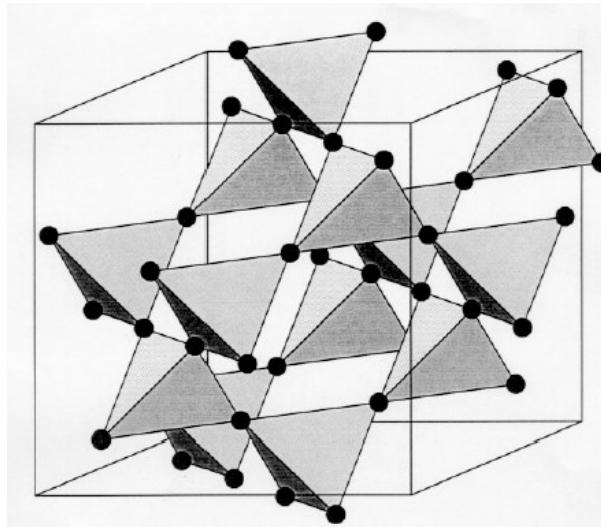
Phase separation in magnetic and non magnetic volumes

Only the combination of neutron and muon data allows the correct interpretation of the data

Frustrated magnetism

Geometrical frustration:
anti-ferromagnetism,
spins cannot all
be satisfied

Pyrochlore compounds:
Corner-sharing
tetrahedra lead to
geometrical
frustration

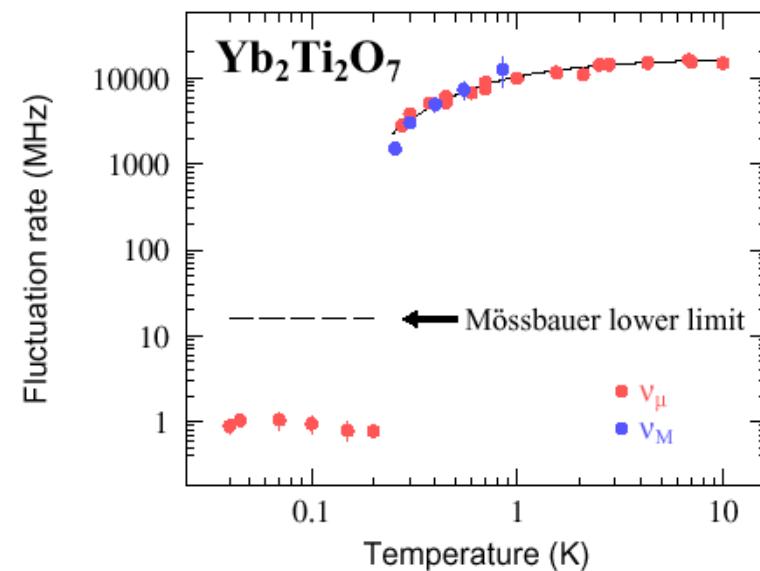
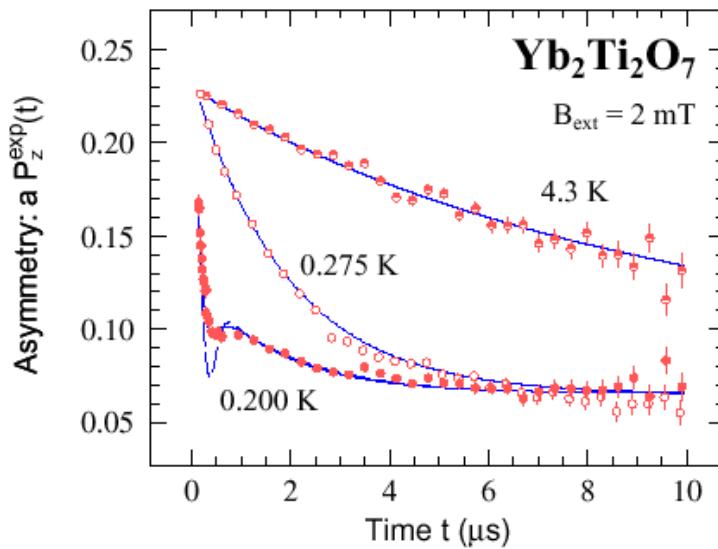


The μ SR technique

- is highly sensitive to local fields,
- is able to characterise the timescale of local field fluctuations,
- is well suited to the investigation of compounds which do not display long-range magnetic correlations,
- requires no application of external fields.

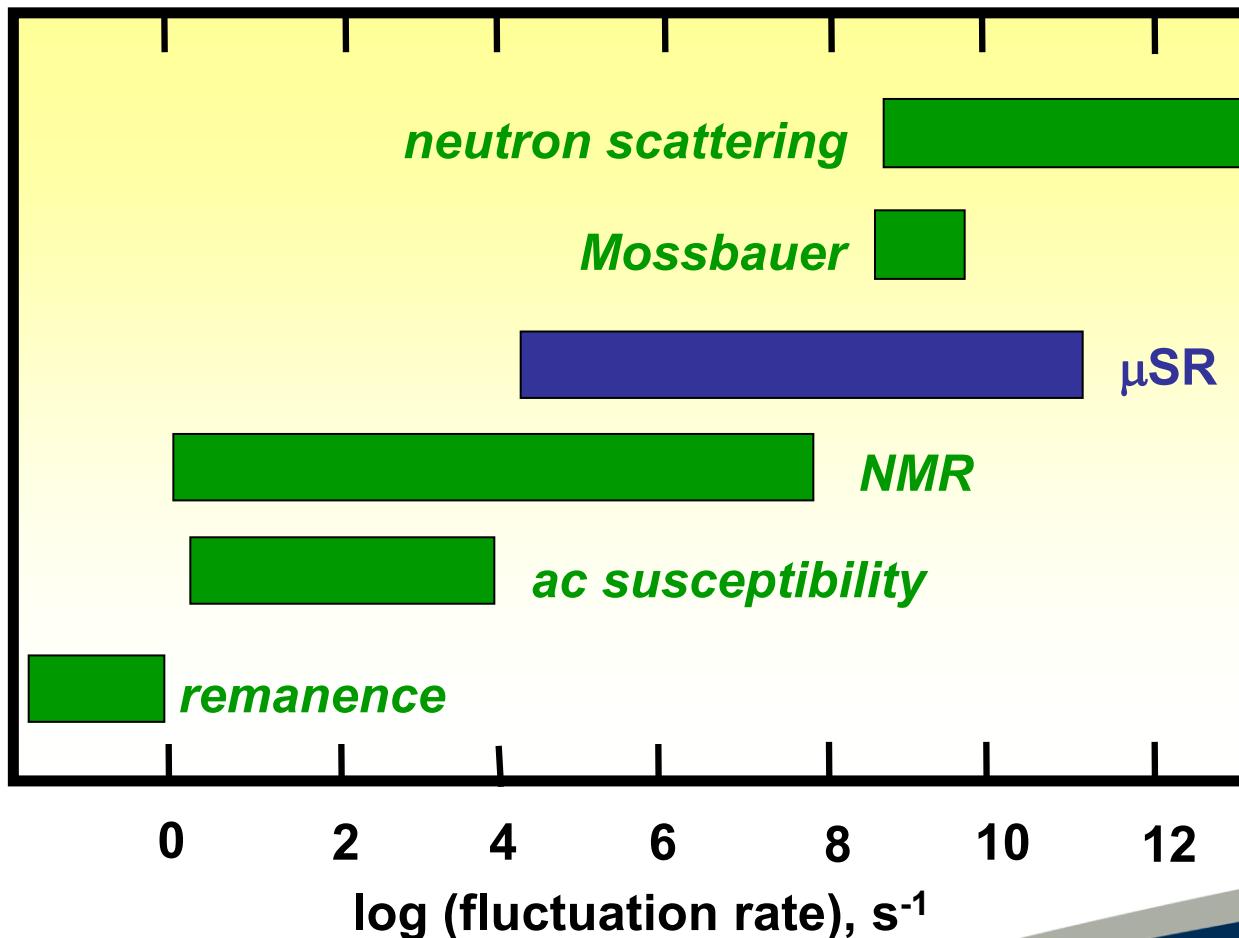
Frustrated magnetism

First order transition in the spin dynamics of
geometrically frustrated $\text{Yb}_2\text{Ti}_2\text{O}_7$
J Hedges et al, PRL 88 (2002) 077204

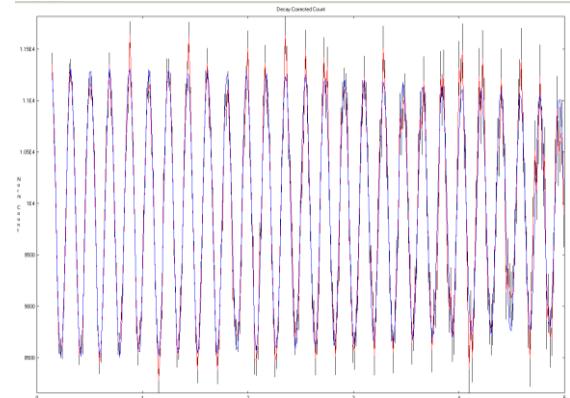
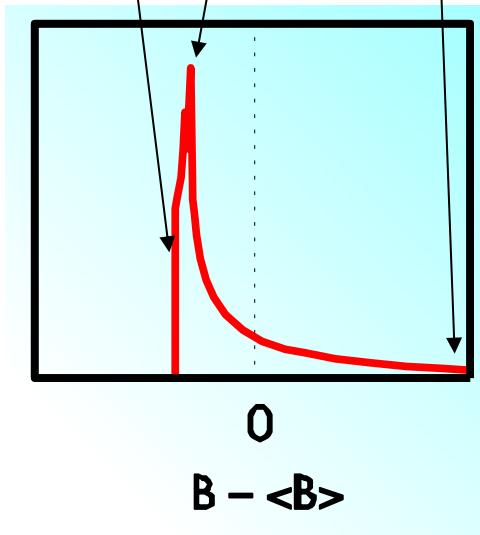
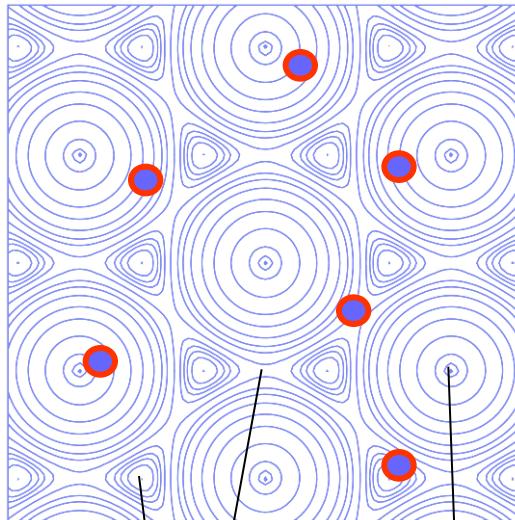


- First order transition at 0.24 K is not to long (or short) range order, but is a change in the Yb^{3+} fluctuation rate.
- Neutron diffraction, Mossbauer and μ SR used

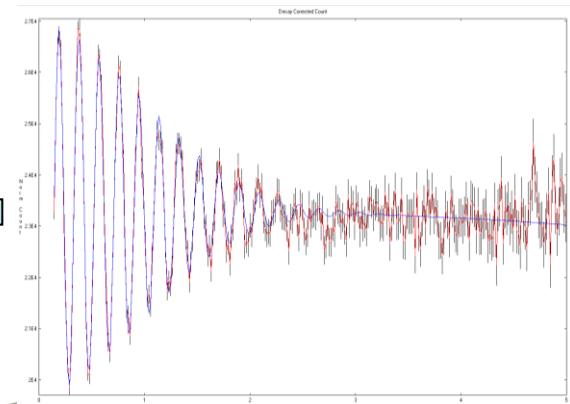
Complementary



Superconductors



Above T_c
Below T_c

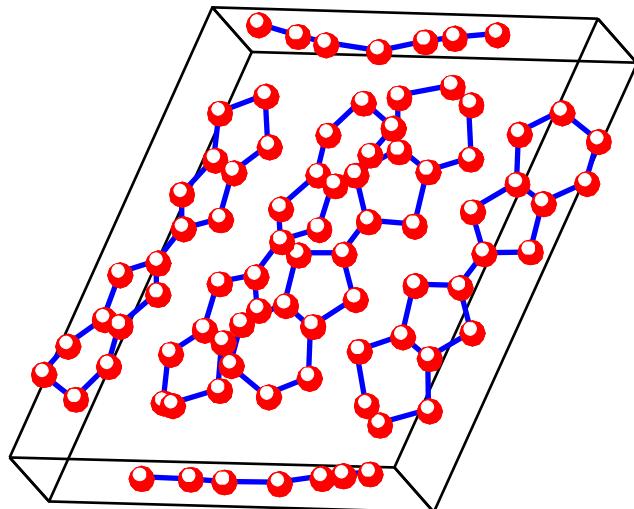


- Interplay of superconductivity and magnetism;
- flux lattice studies;
- measurement of fundamental superconducting parameters

Superconductors

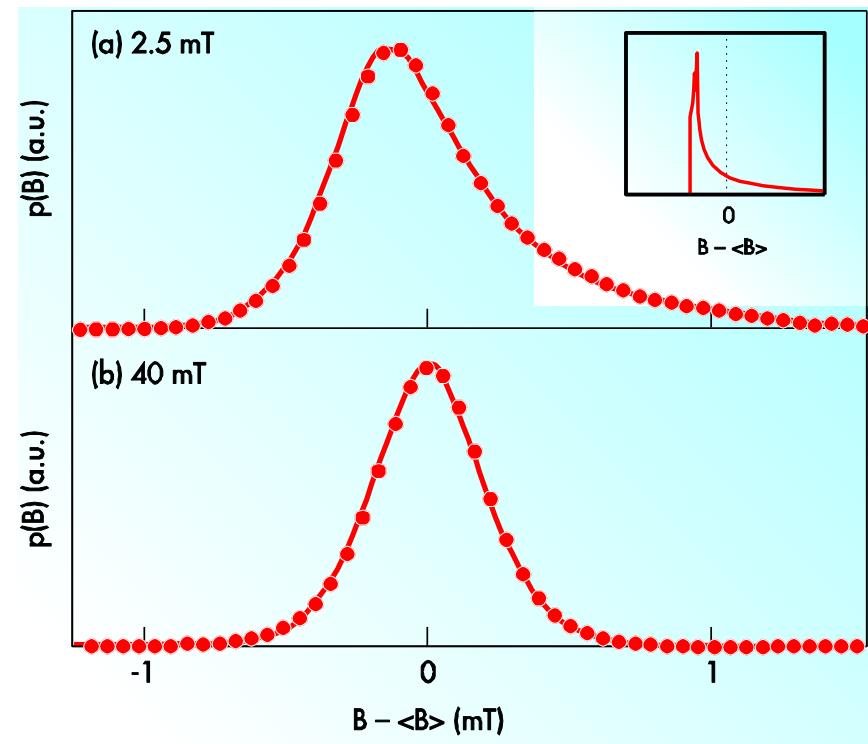
Investigation of Vortex Behavior
in the Organic Superconductor
 κ -(BEDT-TTF)₂Cu(SCN)₂ Using
Muon Spin Rotation

Lee *et al*, PRL 1997



$T_c = 10.4$ K

Highly anisotropic

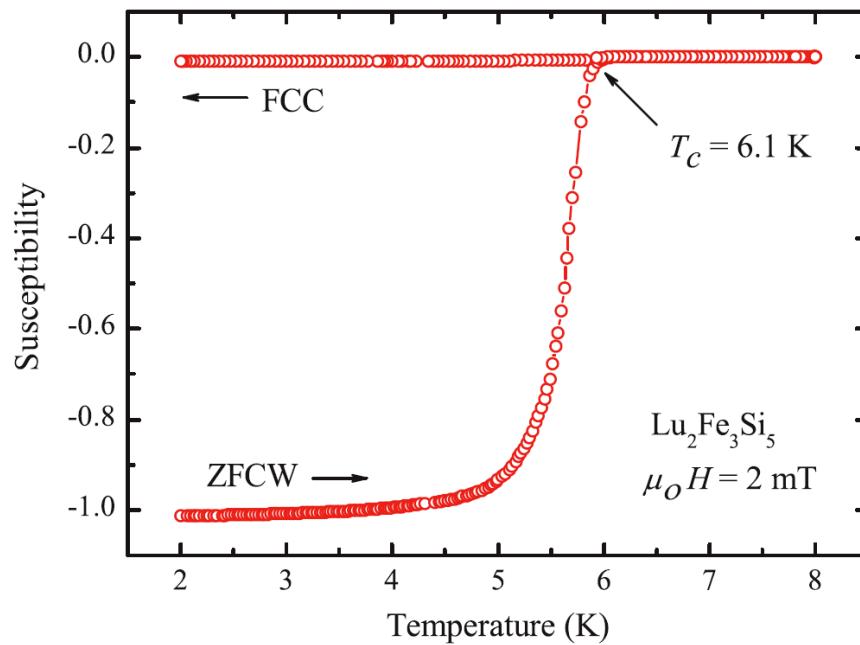


Measurements at 1.8K
Breakdown of 3D order with B and T

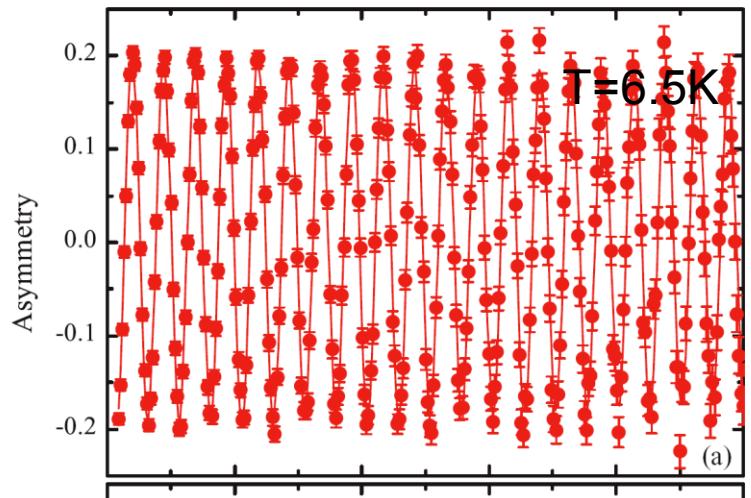


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Superconductors

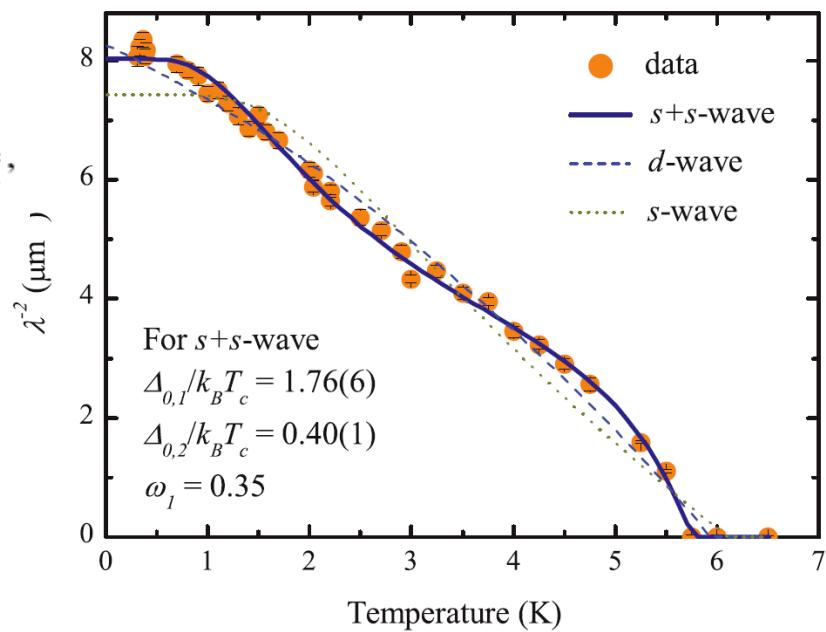
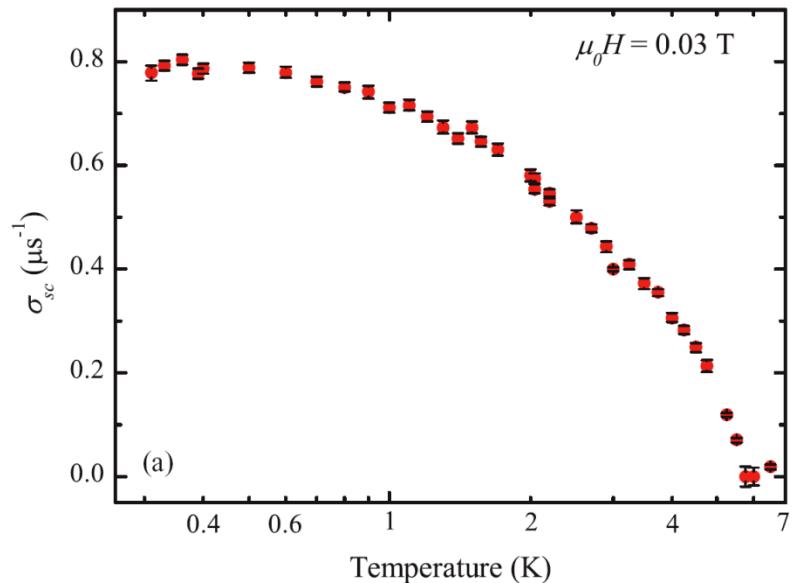


Superconductors



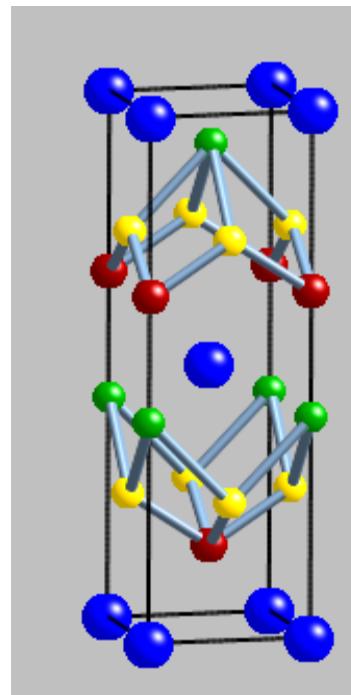
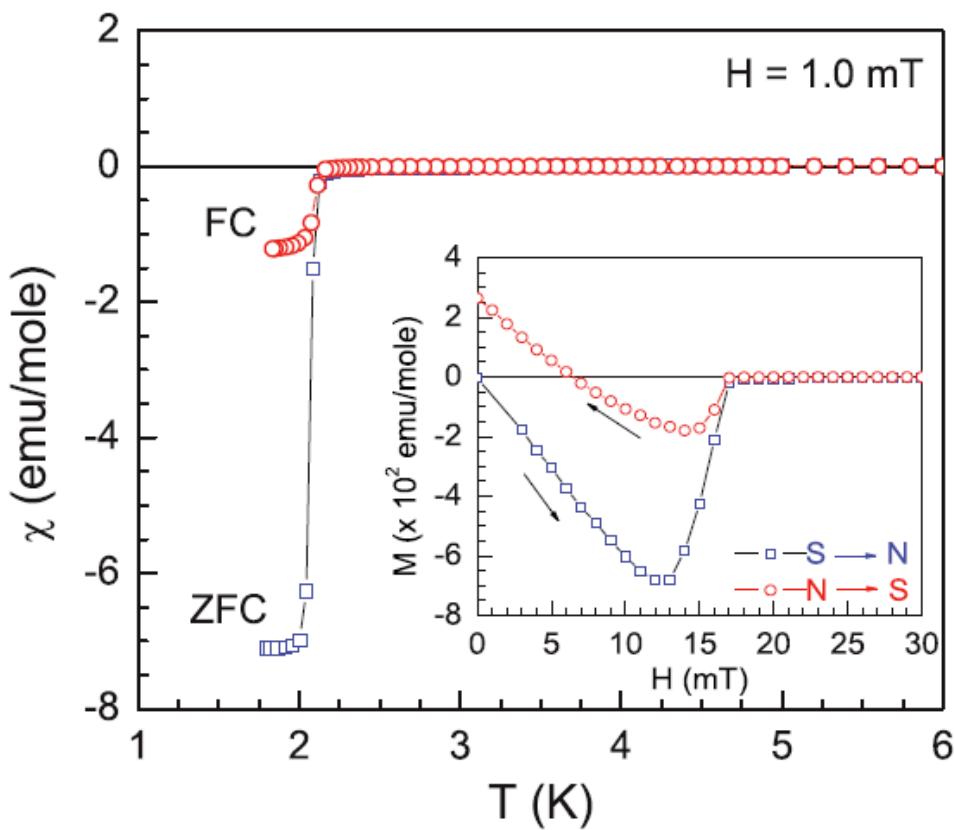
$$\frac{\lambda^{-2}(T)}{\lambda^{-2}(0)} = \omega_1 \frac{\lambda^{-2}(T, \Delta_{0,1})}{\lambda^{-2}(0, \Delta_{0,1})} + \omega_2 \frac{\lambda^{-2}(T, \Delta_{0,2})}{\lambda^{-2}(0, \Delta_{0,2})},$$

$$\frac{\lambda^{-2}(T, \Delta_{0,i})}{\lambda^{-2}(0, \Delta_{0,i})} = 1 + \frac{1}{\pi} \int_0^{2\pi} \int_{\Delta(T, \varphi)}^{\infty} \left(\frac{\partial f}{\partial E} \right) \frac{EdEd\varphi}{\sqrt{E^2 - \Delta_i(T, \varphi)^2}},$$

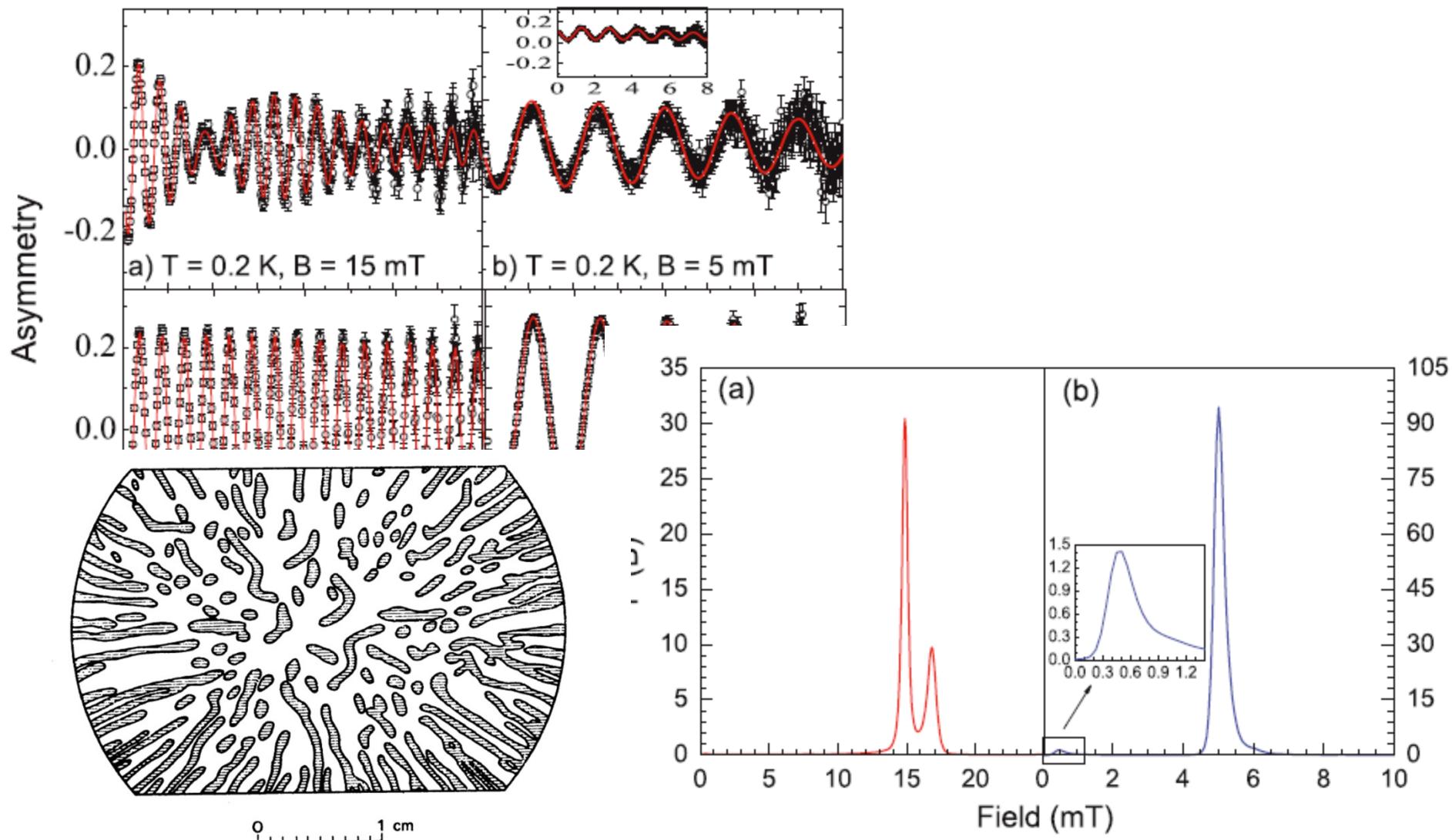


Superconductors

LaRhSi₃

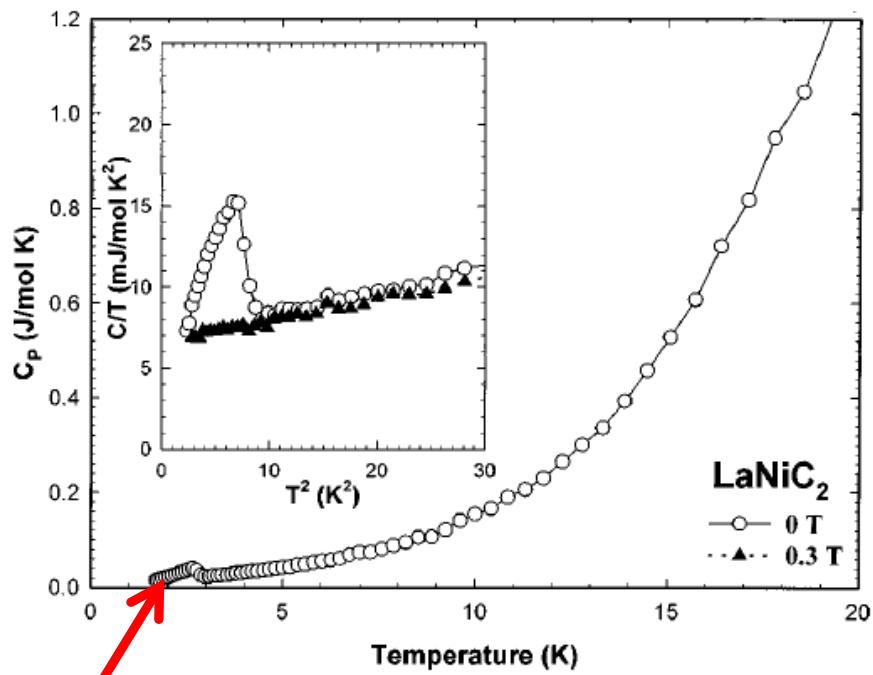


Superconductors



Superconductors

LaNiC₂

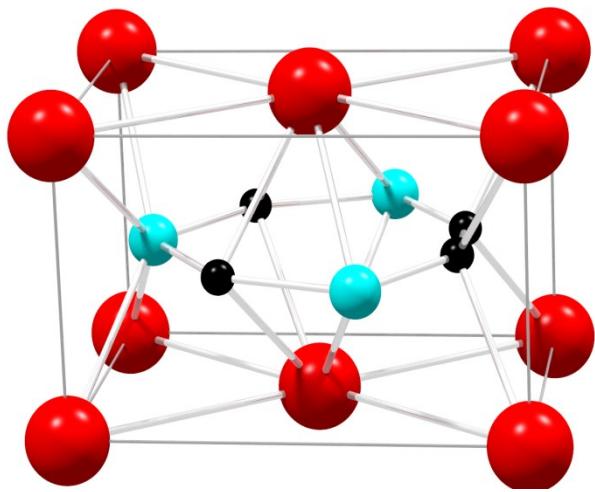


$$T_c = 2.7 \text{ K} \quad \Delta C/\gamma T_c = 1.26 \text{ c.f. (BCS: 1.43)}$$

Note no inversion centre.

C.f. CePt₃Si ⁽¹⁾, Li₂Pt₃B & Li₂Pd₃B ⁽²⁾, ...

(1) Bauer et al. PRL'04 (2) Yuan et al. PRL'06

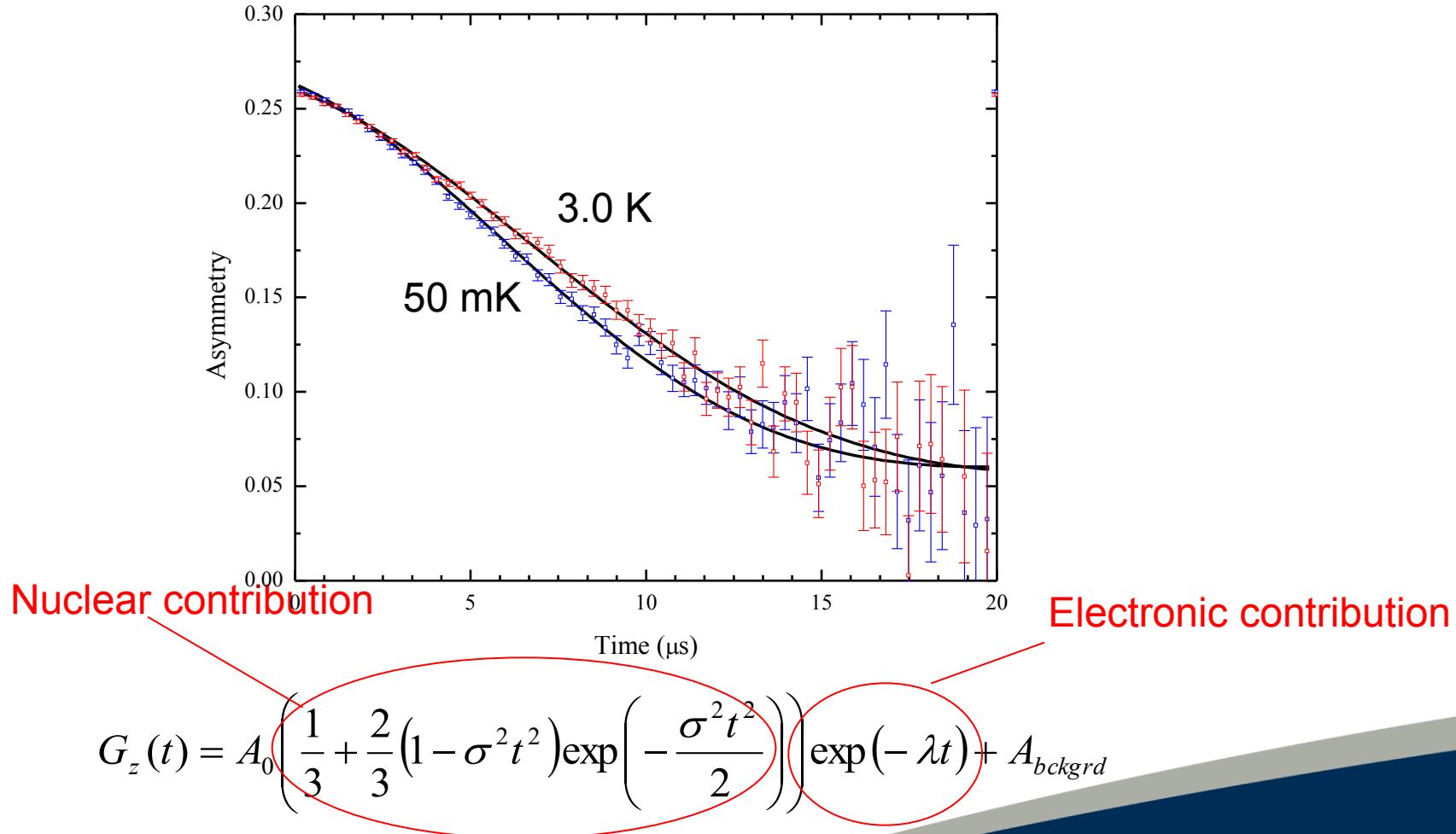


Orthorhombic Amm2 C_{2v}

$a=3.96 \text{ \AA}$
 $b=4.58 \text{ \AA}$
 $c=6.20 \text{ \AA}$

Superconductors

ZF μ SR on LaNiC₂

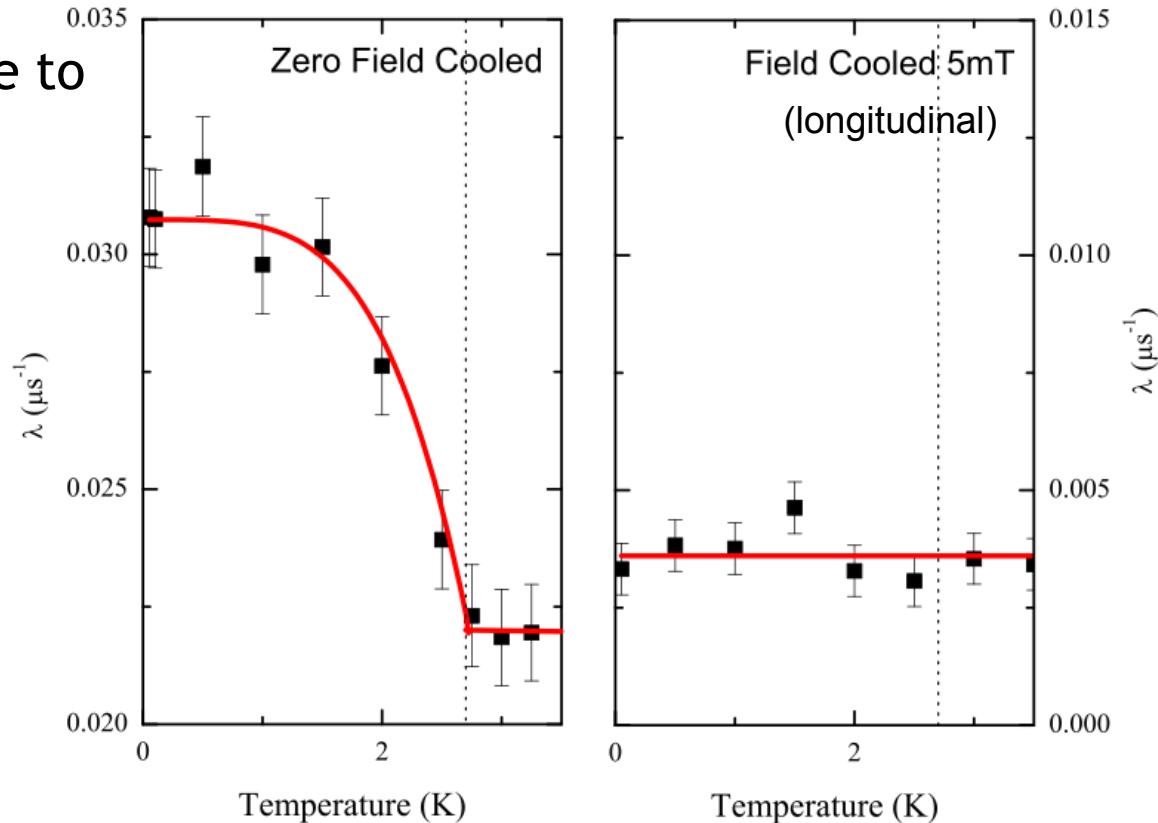


Hillier *et al* PRL 102 117007 (2009)

Superconductors

Relaxation due to
electronic
moments

Moment
size
 $\sim 0.1G$
($\sim 0.01\mu_B$)



Spontaneous, quasi-static fields appearing at T_c
⇒ superconducting state breaks time-reversal symmetry

Superconductors

$SO(3) \times C_{2v}$	Gap function (unitary)	Gap function (non-unitary)
1A_1	$\Delta(\mathbf{k})=1$	-
1A_2	$\Delta(\mathbf{k})=k_x k_y$	breaks only $U(1) \times SO(3)$ *
1B_1	$\Delta(\mathbf{k})=k_x k_z$	-
1B_2	$\Delta(\mathbf{k})=k_y k_z$	-
3A_1	$d(\mathbf{k})=(0,0,1)k_z$	$d(\mathbf{k})=(1,i,0)k_z$
3A_2	$d(\mathbf{k})=(0,0,1)k_x k_y k_z$	$d(\mathbf{k})=(1,i,0)k_x k_y k_z$
3B_1	$d(\mathbf{k})=(0,0,1)k_x$	$d(\mathbf{k})=(1,i,0)k_x$
3B_2	$d(\mathbf{k})=(0,0,1)k_y$	$d(\mathbf{k})=(1,i,0)k_y$

Non-unitary

* C.f. Li₂Pd₃B & Li₂Pt₃B,
H. Q. Yuan et al. PRL'06

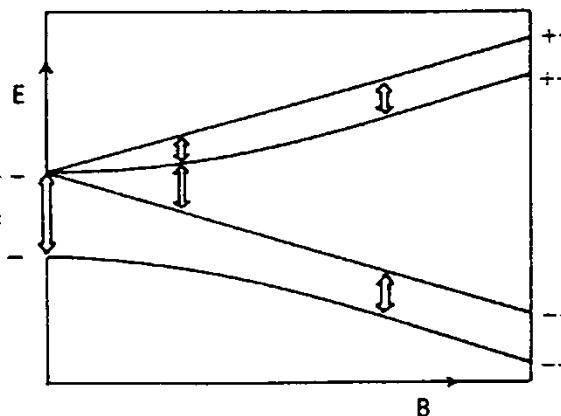
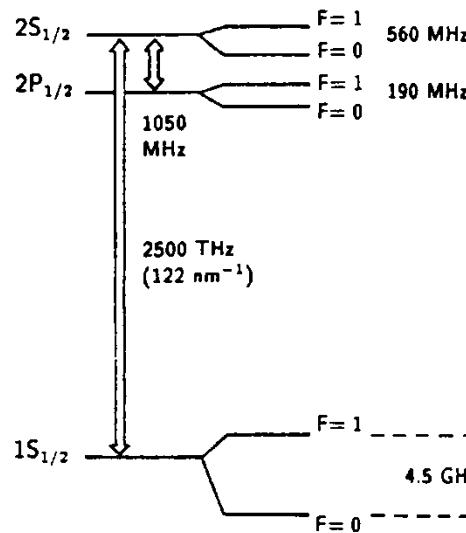
Hillier *et al* PRL 102 117007 (2009)

H and Semiconductors

'Muonium' (Mu): light hydrogen atom



e^-



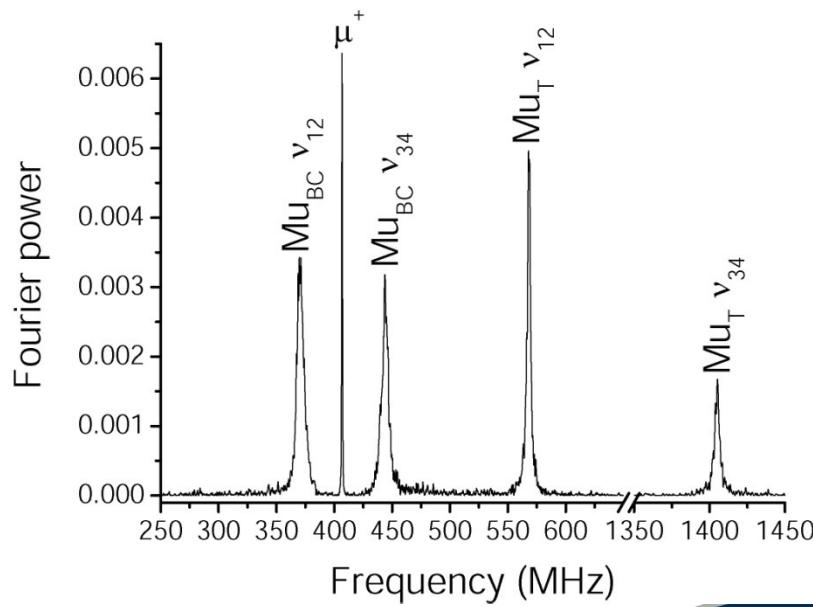
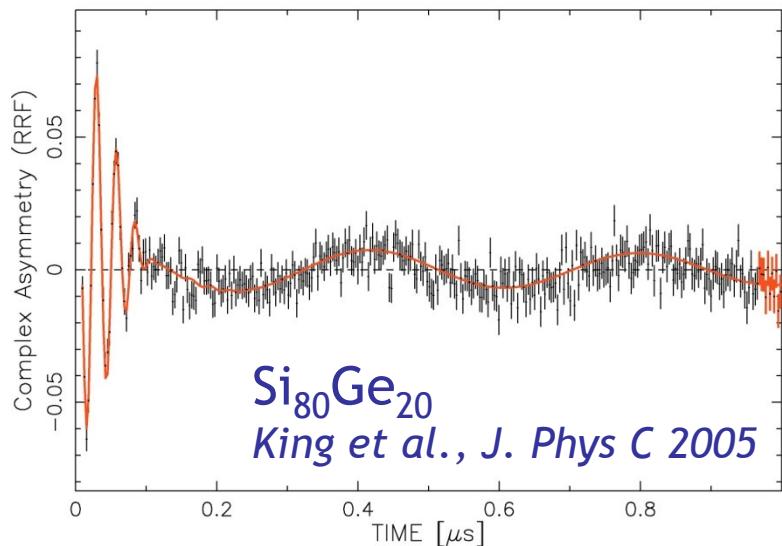
Mass: $m_{\text{Mu}} = 1/9 m_H$

Bohr radius: $a_{\text{Mu}} = 1.004 a_H$

Ionisation potential: $I_{\text{Mu}} = 0.996 I_H$

Chemical behaviour very similar - dynamics different

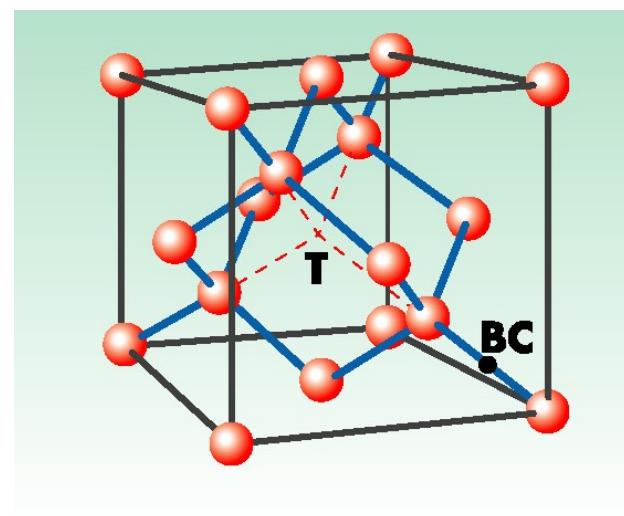
H and Semiconductors



Silicon: two muon species exist at low temperatures.

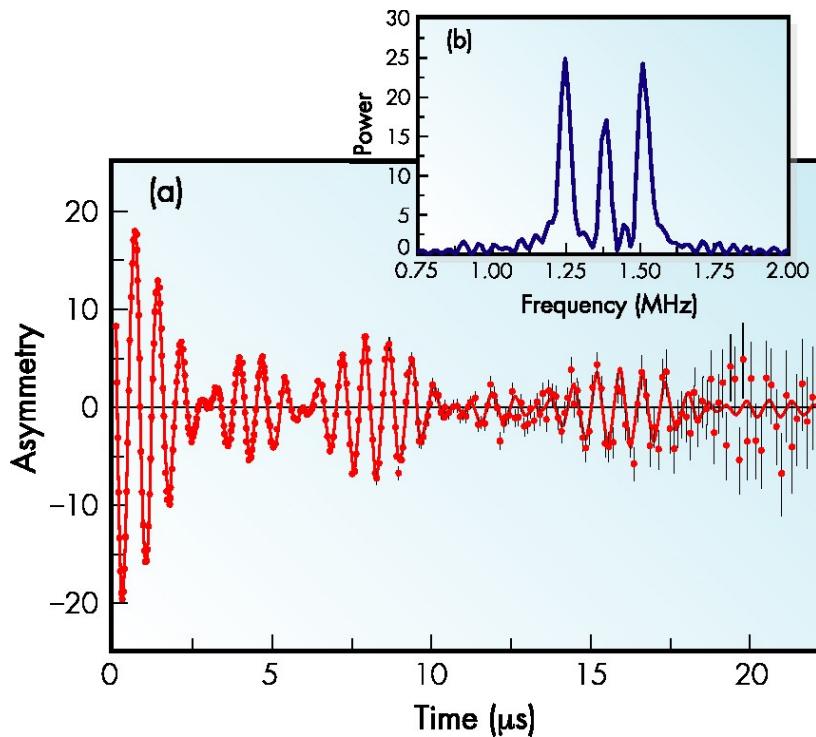
Mu_{T} : Cage-centred, isotropic, mobile

Mu_{BC} : Bond centred, axially symmetric, immobile

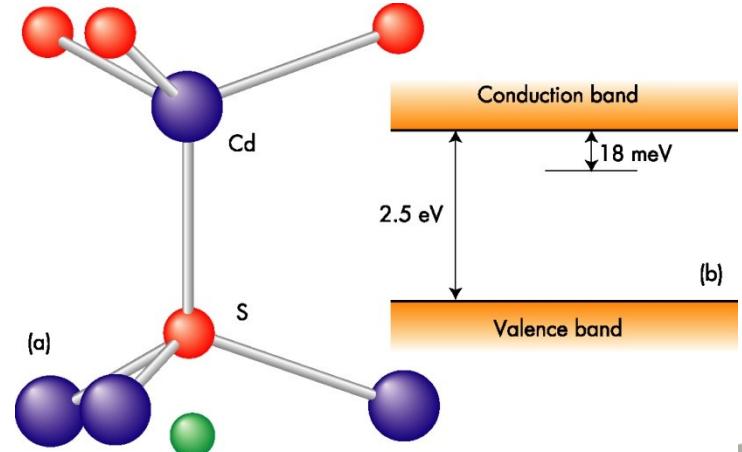


H and Semiconductors

CdS: II-VI semiconductor



- Hyperfine parameter 10^{-4} of vacuum value
 - Extended wavefunction
 - Ionises at 22 K
 - **Shallow donor state**
- Gil et al, PRL 1999



H and Semiconductors

VOLUME 86, NUMBER 12

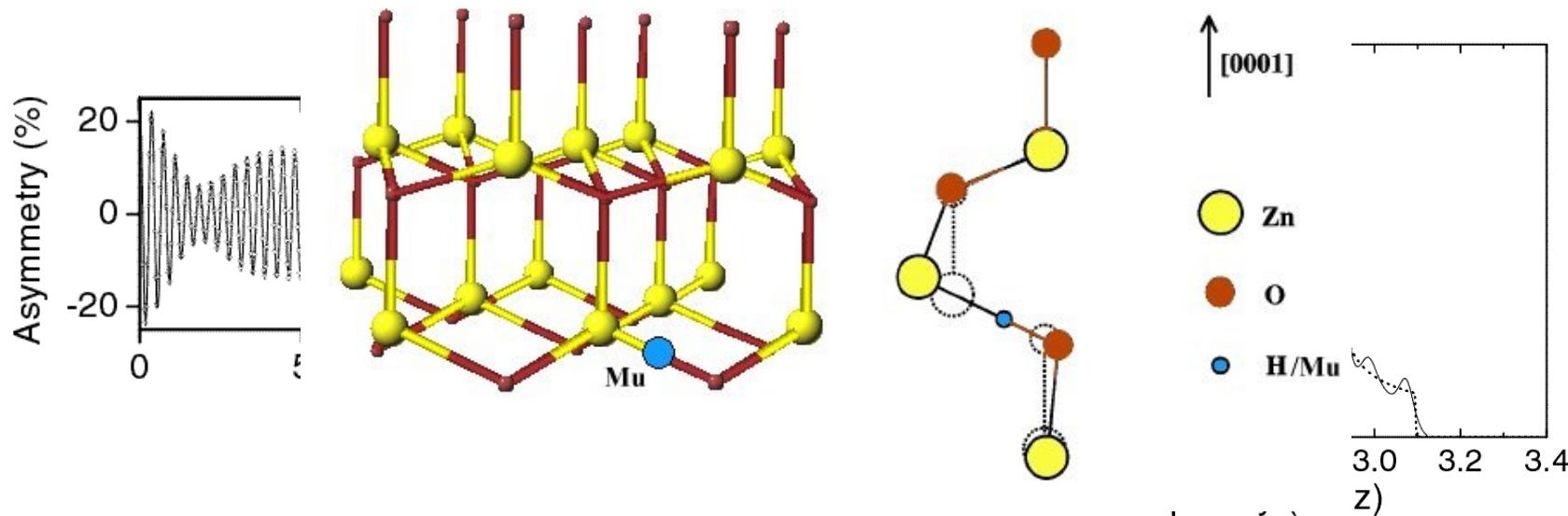
PHYSICAL REVIEW LETTERS

19 MARCH 2001

Experimental Confirmation of the Predicted Shallow Donor Hydrogen State in Zinc Oxide

S. F. J. Cox,^{1,2} E. A. Davis,³ S. P. Cottrell,¹ P. J. C. King,¹ J. S. Lord,¹ J. M. Gil,⁴ H. V. Alberto,⁴ R. C. Vilão,⁴ J. Pirotto Duarte,⁴ N. Ayres de Campos,⁴ A. Weidinger,⁵ R. L. Lichti,⁶ and S. J. C. Irvine⁷

¹ISIS Facility, Rutherford Appleton Laboratory, Chilton OX11 0QX, United Kingdom



Science & Technology Facilities Council

ISIS

Organic Spintronics

PHYSICAL REVIEW B 84, 085209 (2011)



Importance of intramolecular electron spin relaxation in small molecule semiconductors

L. Schulz,¹ M. Willis,² L. Nuccio,² P. Shusharov,² S. Fratini,³ F. L. Pratt,⁴ W. P. Gillin,² T. Kreouzis,² M. Heeney,⁵ N. Stingelin,⁵ C. A. Stafford,⁶ D. J. Beesley,⁵ C. Bernhard,¹ J. E. Anthony,⁷ I. McKenzie,⁴ J. S. Lord,⁴ and A. J. Drew^{1,2}

¹Department of Physics and Fribourg Center of Nanomaterials, University of Fribourg, CH-1700 Fribourg, Switzerland

²Queen Mary University of London, Department of Physics, London, E1 4NS, United Kingdom

³Institut Néel CNRS, F-38042 Grenoble, France

⁴ISIS Muon Facility, Rutherford Appleton Laboratory, Didcot, OX11 0QX, United Kingdom

⁵Centre for Plastic Electronics, Imperial College London, London, SW7 2AZ, United Kingdom

⁶Department of Physics, University of Arizona, Tucson, Arizona, USA

⁷Department of Chemistry, U

Small molecule
semiconductors used in
molecular electronic devices
such as:

Spin valves
OLEDs
FETs

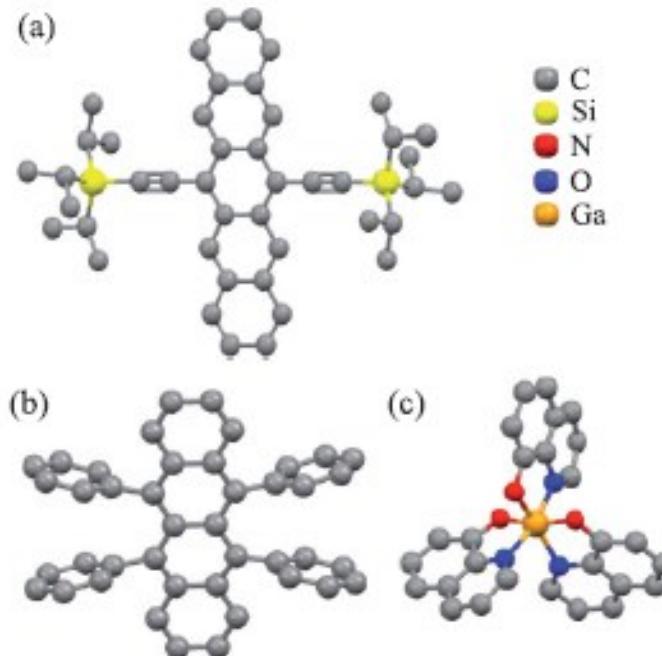
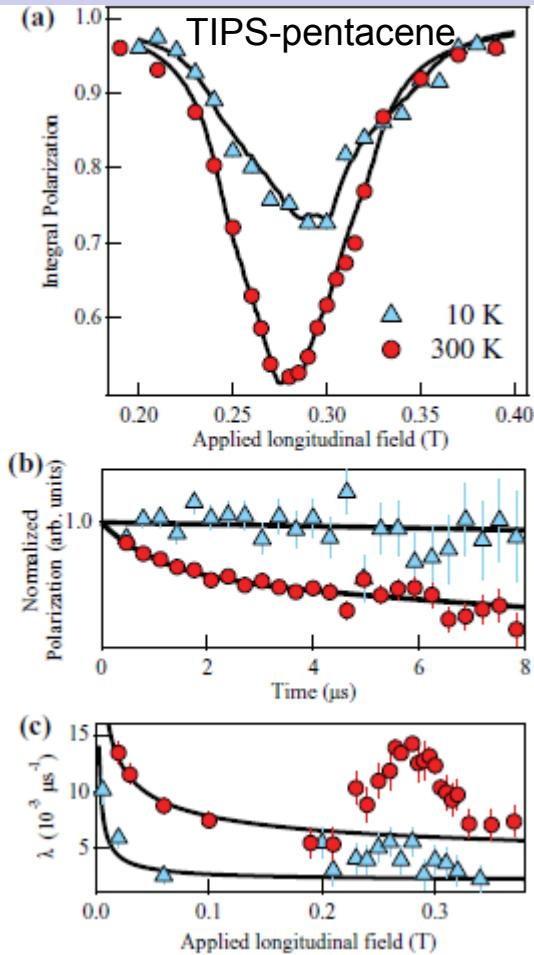


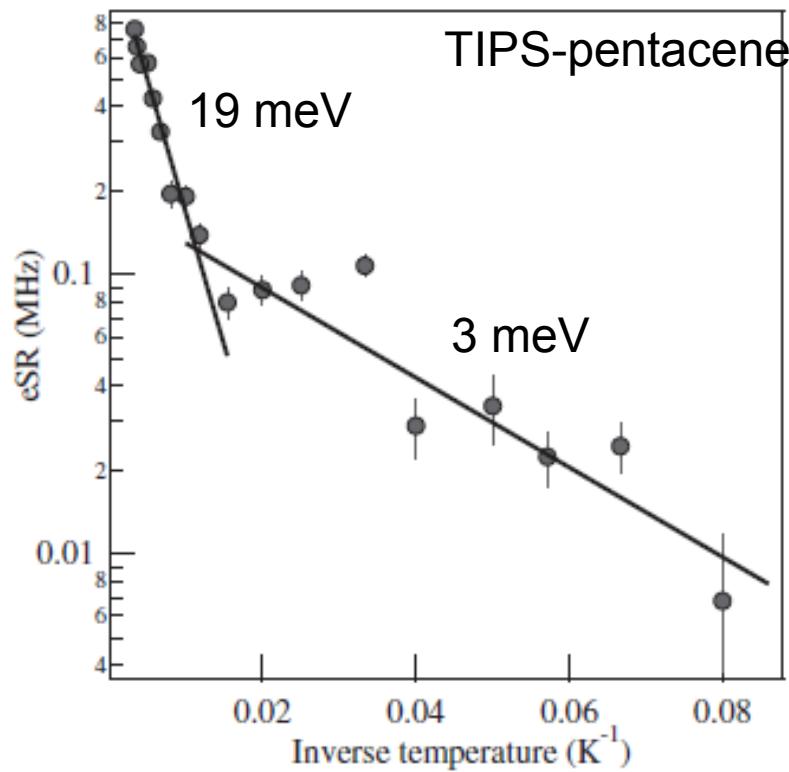
FIG. 1. (Color online) Molecular structures of (a) TIPS-pentacene, (b) rubrene, and (c) Gaq_3 (Refs. 13, 14, and 22–24). Hydrogen has been omitted for clarity.

Organic Spintronics

ALC resonance gives a sensitive measure of the electron spin relaxation (eSR)

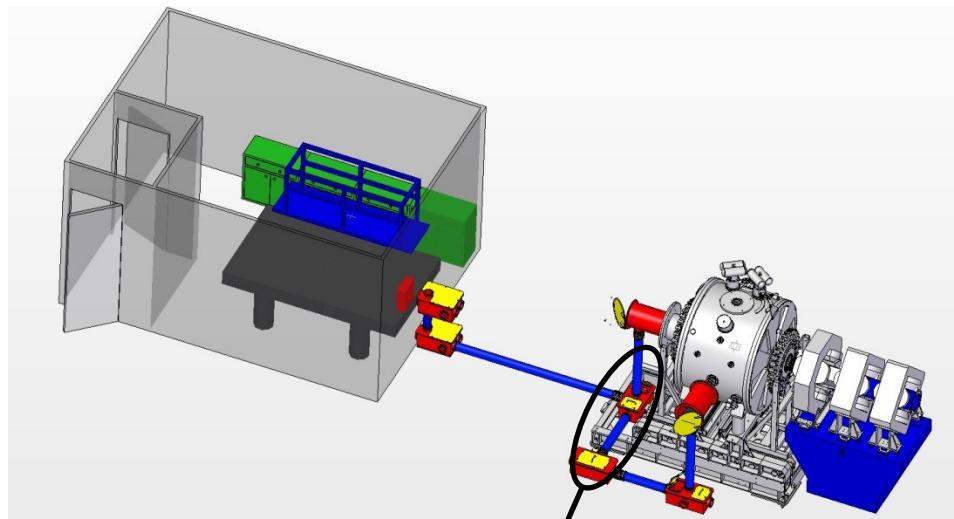


T dependence of the eSR is governed by intramolecular vibrational modes



Vibration-modulated spin-orbit coupling emerges as the dominant spin relaxation mechanism in this type of system (rather than hyperfine interaction)

Organic Spintronics - Exciting muons



Alan Drew from QMUL has been awarded 1.5MEuros European Research Council grant to develop the HiFi muon spectrometer.

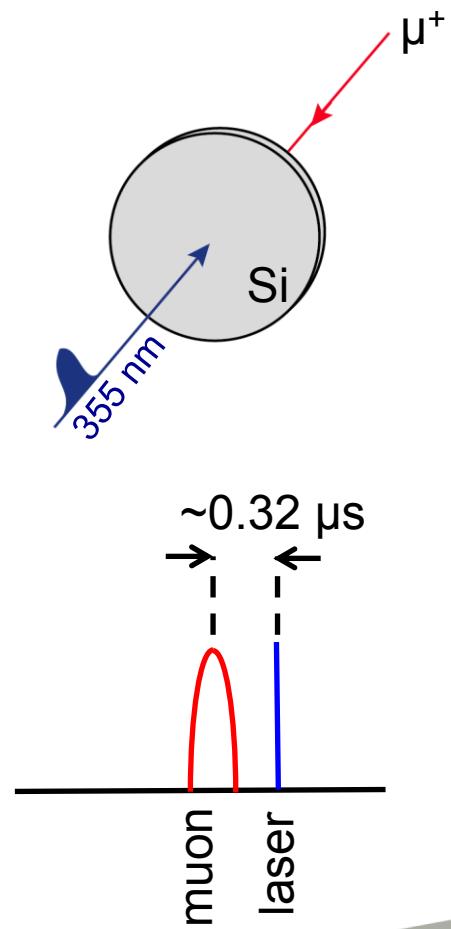
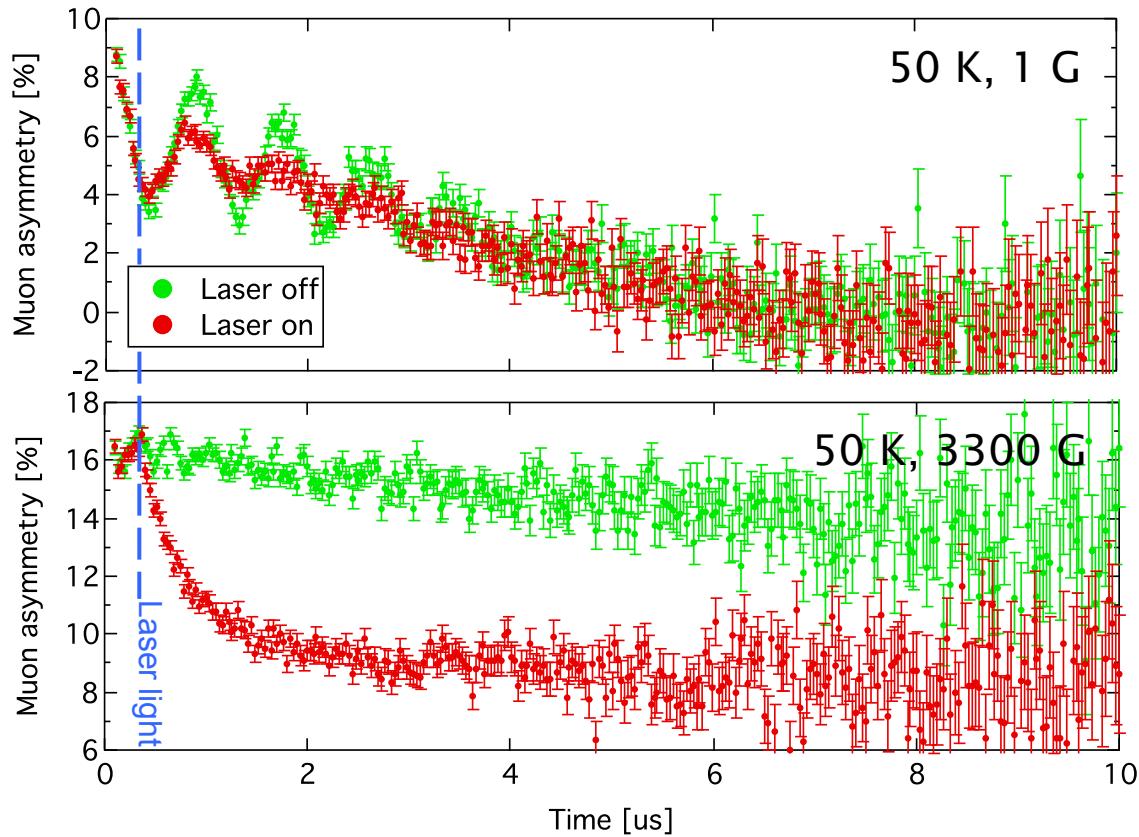
By installing a powerful laser system on to the existing HIFI instrument – the ability to perform muon experiments enhanced by a laser in magnetic fields as high as 5T is not available anywhere else.

This research has potential impact in fields ranging from green energy production to biology and medicine.

Transport system installed and first beam Dec. First experiments already completed

Commissioning experiment on Silicon

n-type Si: well-studied system



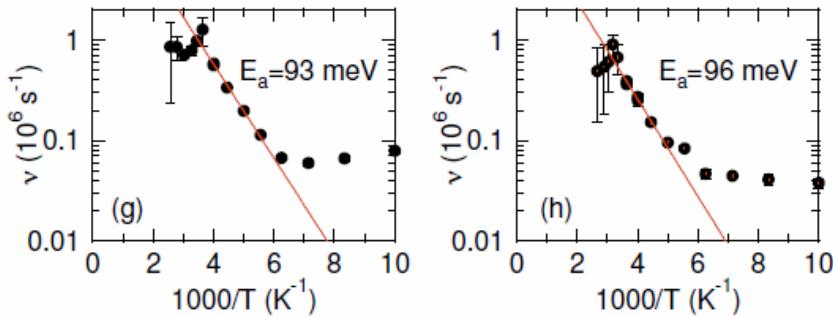
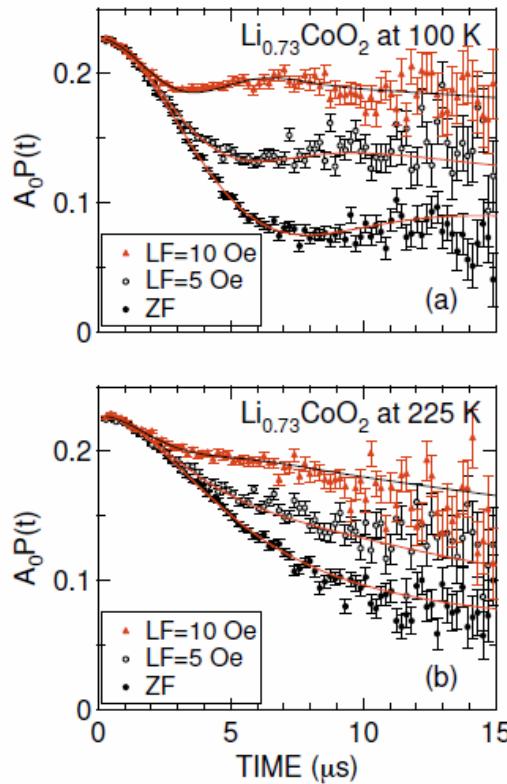
- Site exchange conversion induces relaxation: $\text{Mu}_T^0 \rightarrow \text{Mu}_{\text{BC}}^0$



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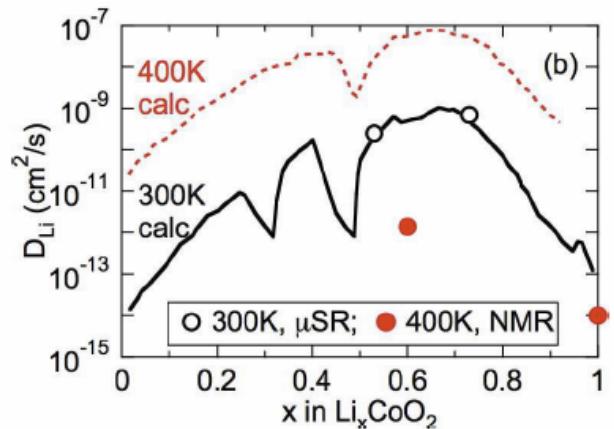
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Ionic conduction



Li diffusion constant determined from the fluctuation rate

$$D_{\text{Li}} = \sum_{i=1}^n \frac{1}{N_i} Z_{v,i} s_i^2 \nu,$$

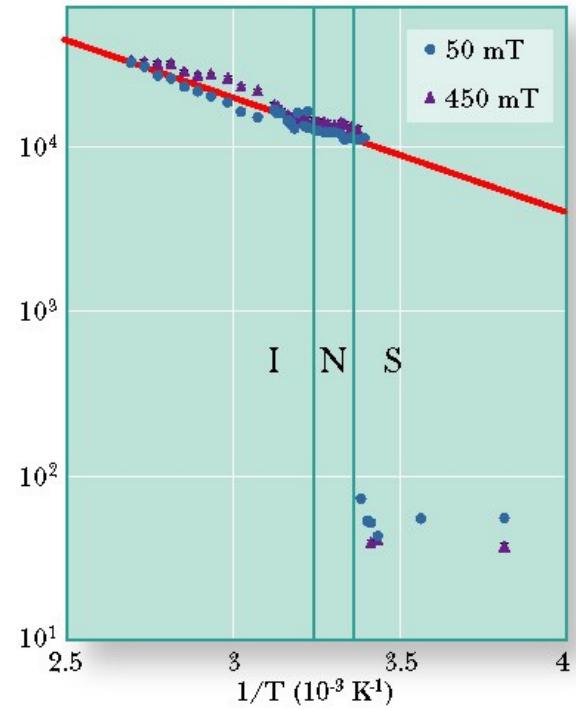
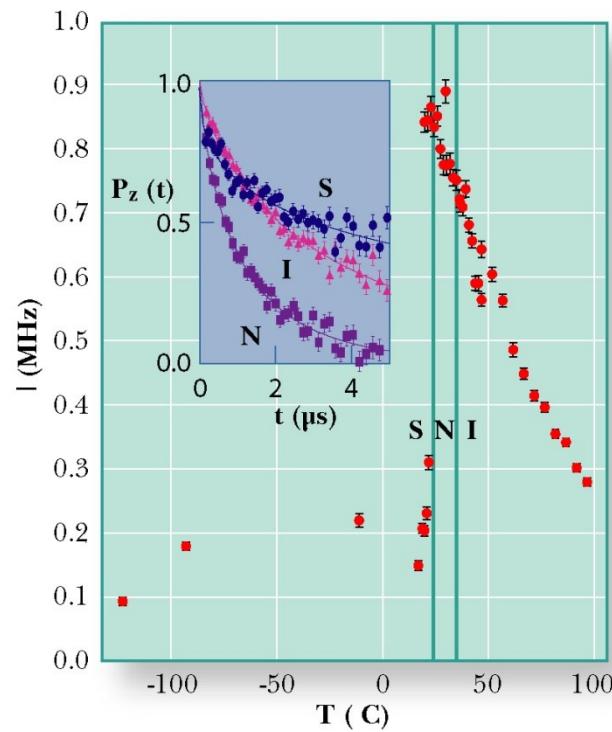
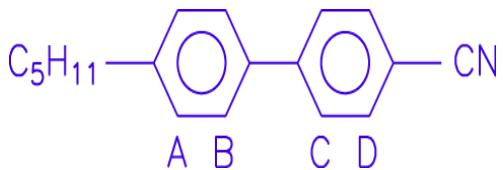


Fit to a dynamic Gaussian KT function

$$A_0 P_{\text{LF}}(t) = A_{\text{KT}} G^{\text{DGKT}}(\Delta, \nu, t, H_{\text{LF}}) + A_{\text{BG}}$$

Molecular dynamics

liquid crystal
4'-n-pentyl-4-
cyanobiphenyl

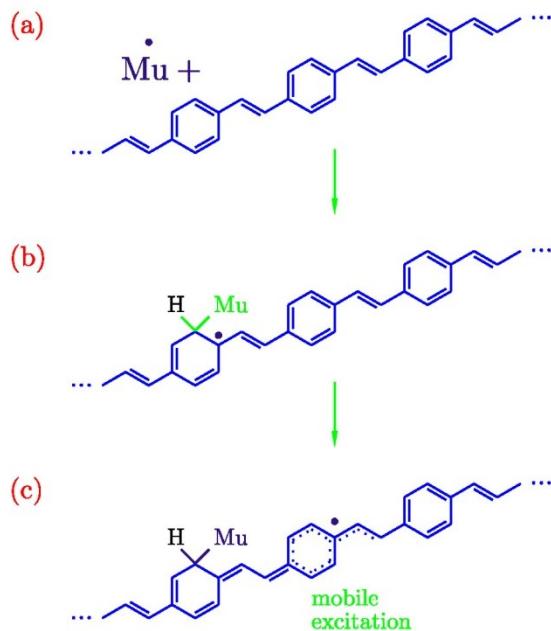


- Molecular fluctuation rate extracted from muon depolarisation
- Large increase at solid-nematic phase transition.
- Motion is consistent with rotation of the whole molecule.

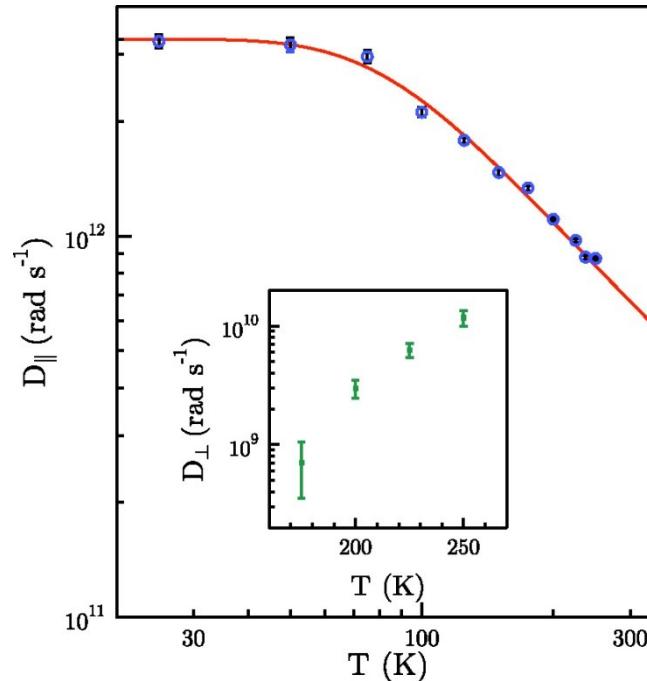
*Molecular dynamics in a nematic liquid crystal probed by muons; BW Lovett *et al*, PRB 63 (2000) 054204.*

Carrier motion in polymers

Polyphenylvinylene (PPV)



- Polymer LEDs based upon PPV
- muons can study polaron motion in *undoped* polymers; they provide information on intrinsic mobility processes

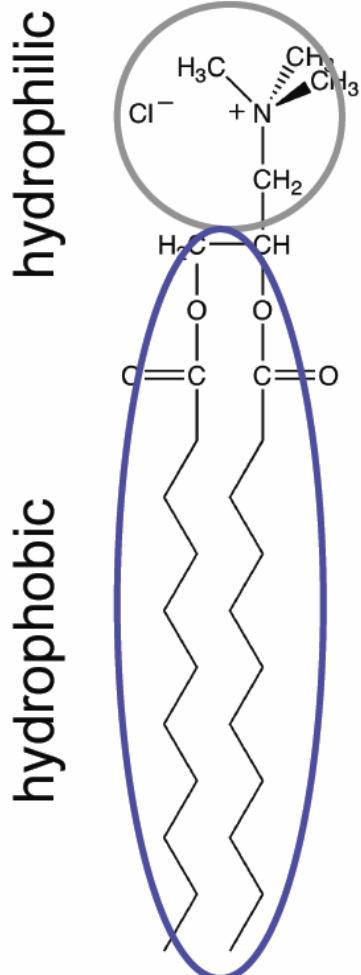


For PPV,
intrachain
diffusion is
phonon-limited,
metallic-style;
interchain
diffusion is
thermally-assisted.

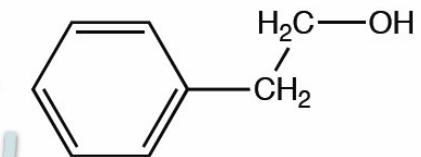
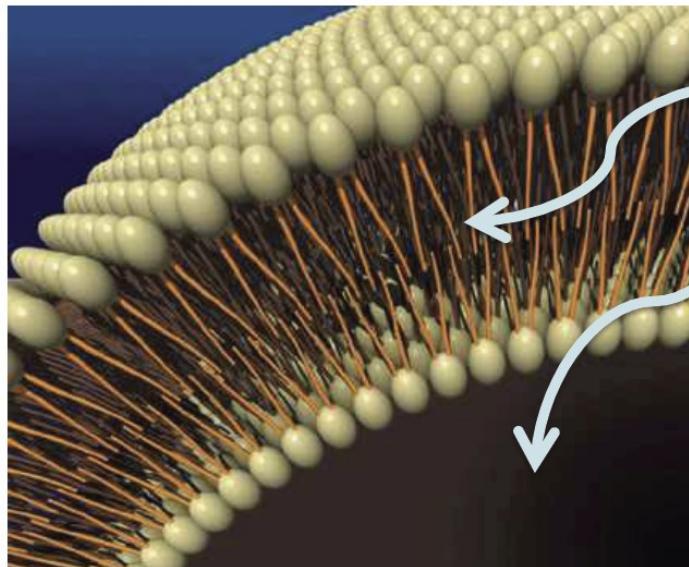
Muon-spin relaxation study of charge carrier dynamics in the conducting polymer PPV, SJ Blundell et al, *Synth Met.* 119 (2001) 205

Partitioning of co-surfactants

Surfactant



Co-surfactant



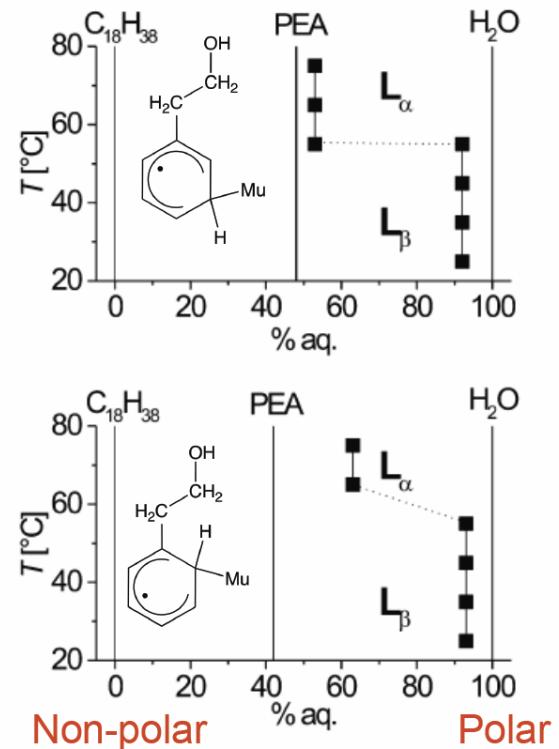
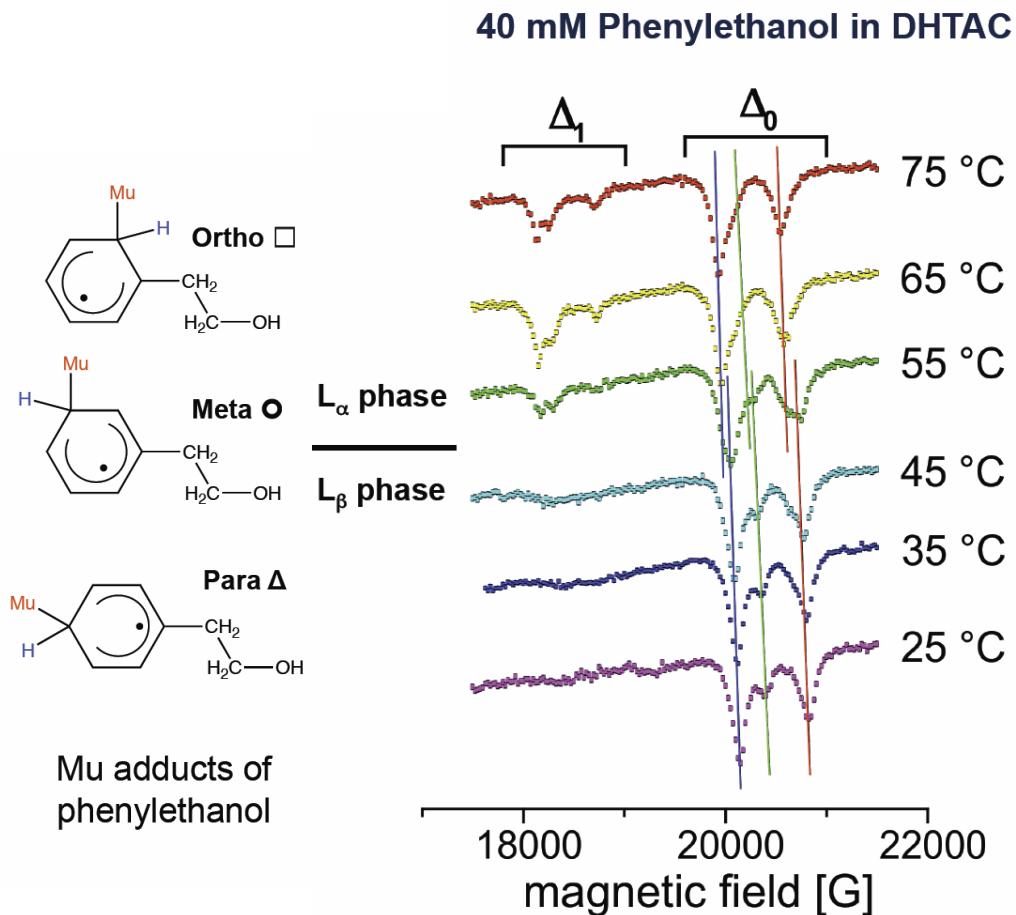
2-phenylethanol

- Fragrances
- Food additives.
- Drug delivery

Surfactants can form bilayers, micelles, vesicles, etc.



Partitioning of co-surfactants

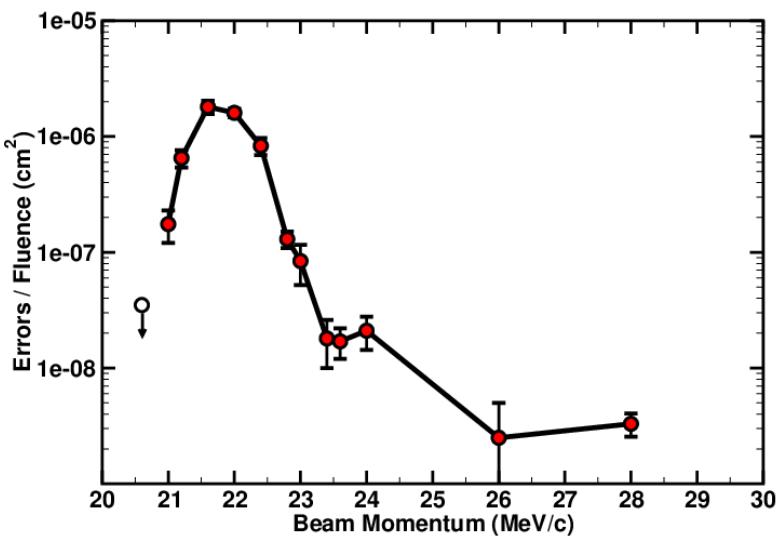
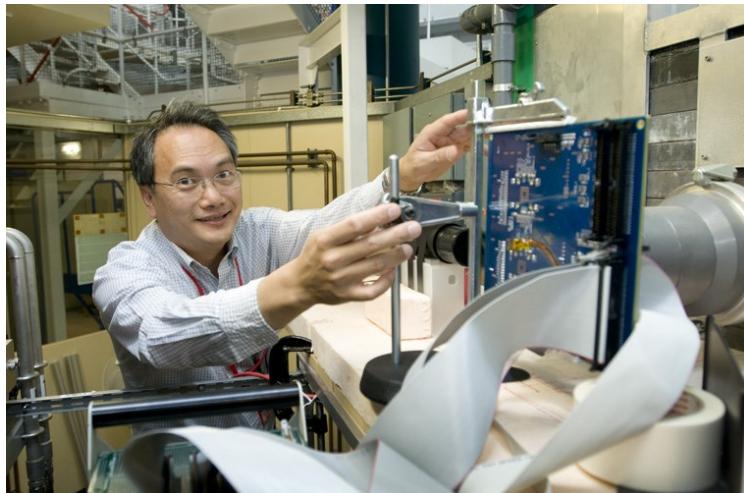


Scheuermann *et al.* PCCP 2002, 4, 1510



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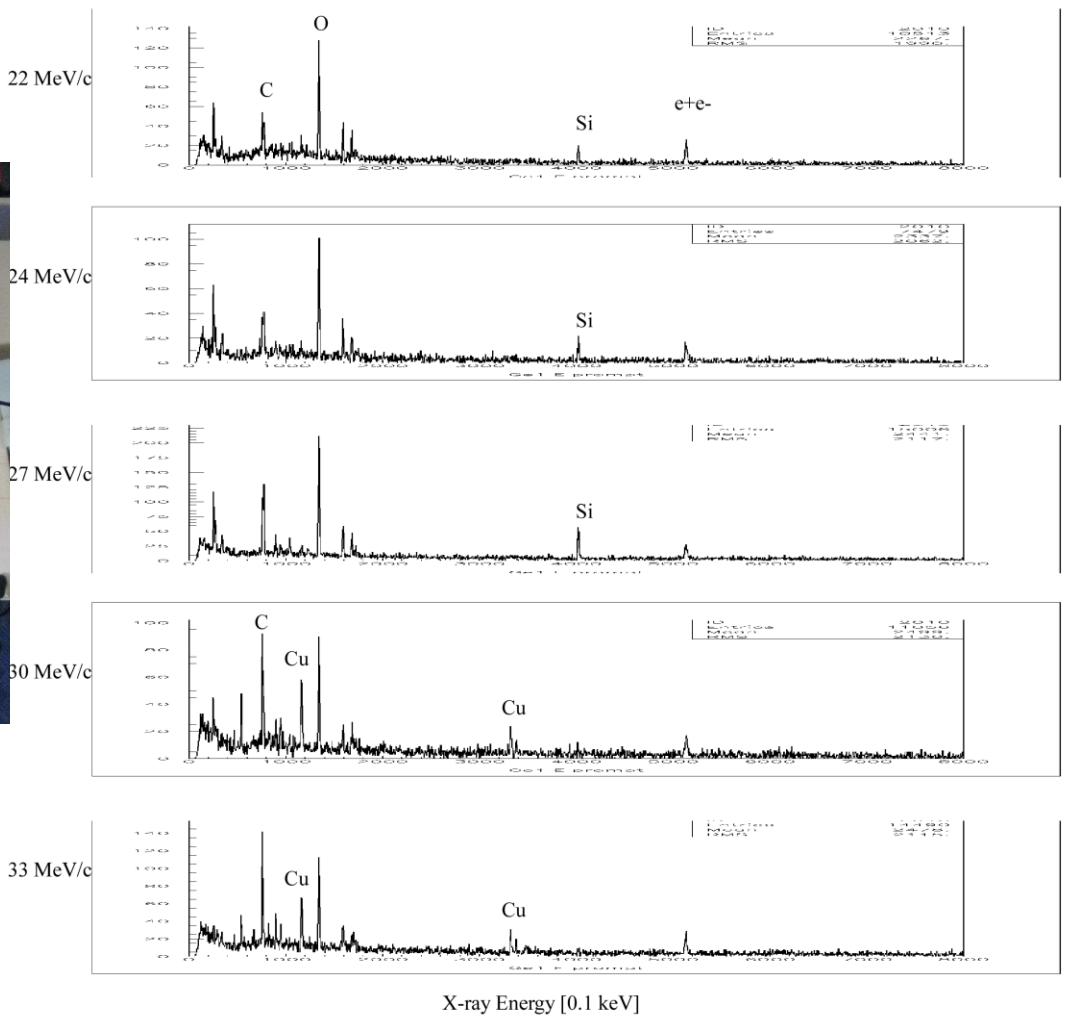
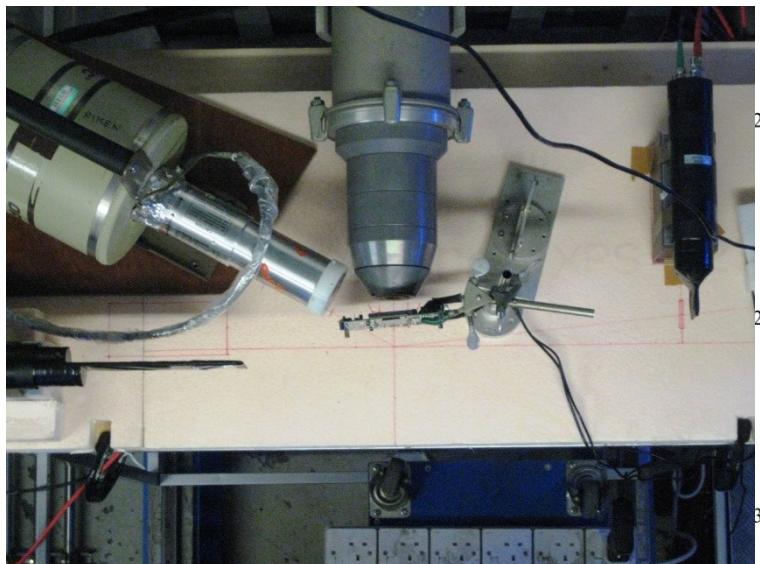
Muon Electronics Irradiation



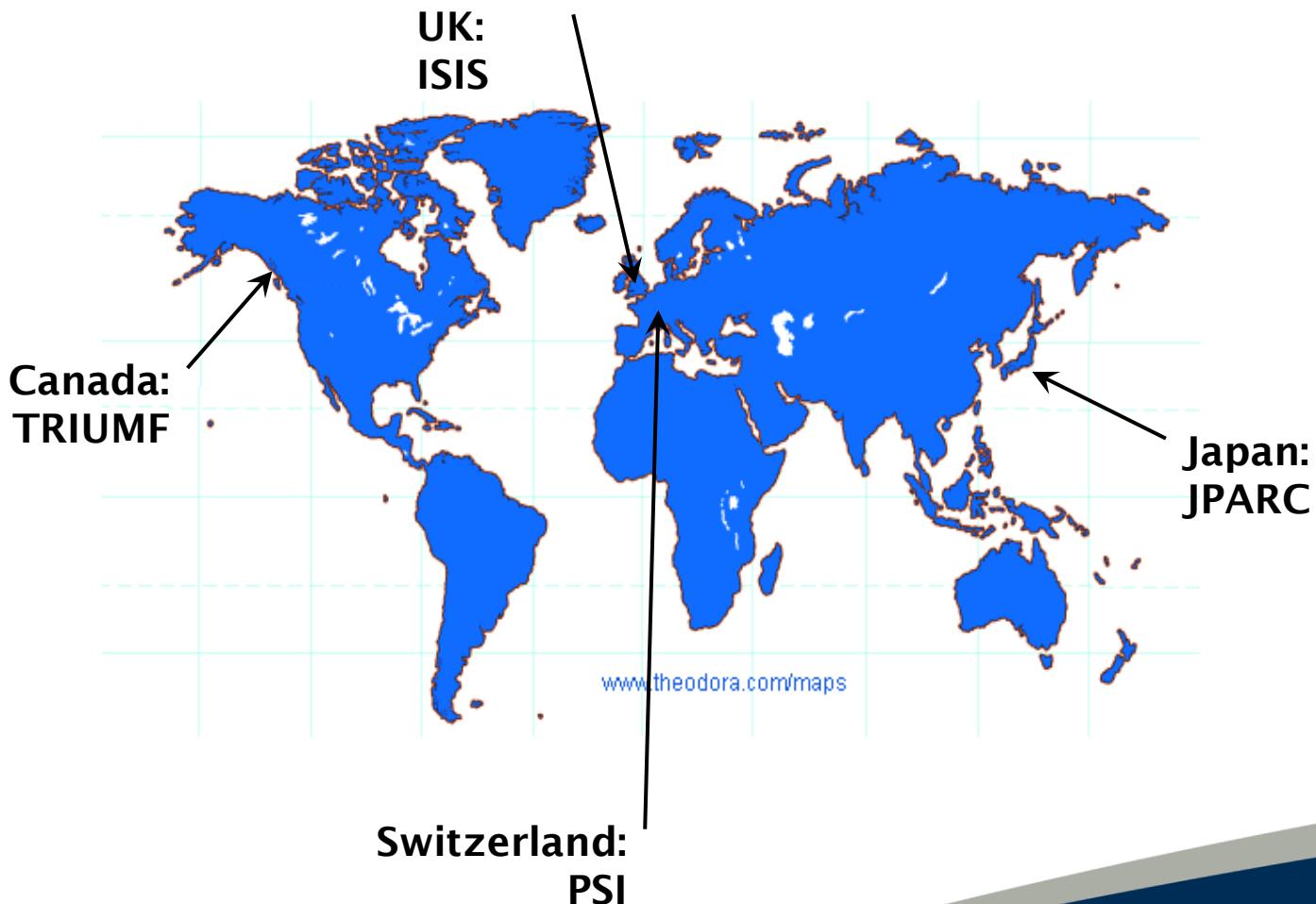
- Researchers at Vanderbilt University used RIKEN-RAL facility to investigate effects of muons on state-of-the-art microelectronic memories
- Collaborators from industry including Marvell Semiconductors, Cisco Systems, and Texas Instruments
- Muon-induced data errors clearly observed in multiple devices
- Results will support investigation into error rates of commercial electronics
- Recently tested electronics for CERN

Muon Electronics Irradiation (2)

We can use negative muons
finding the active component

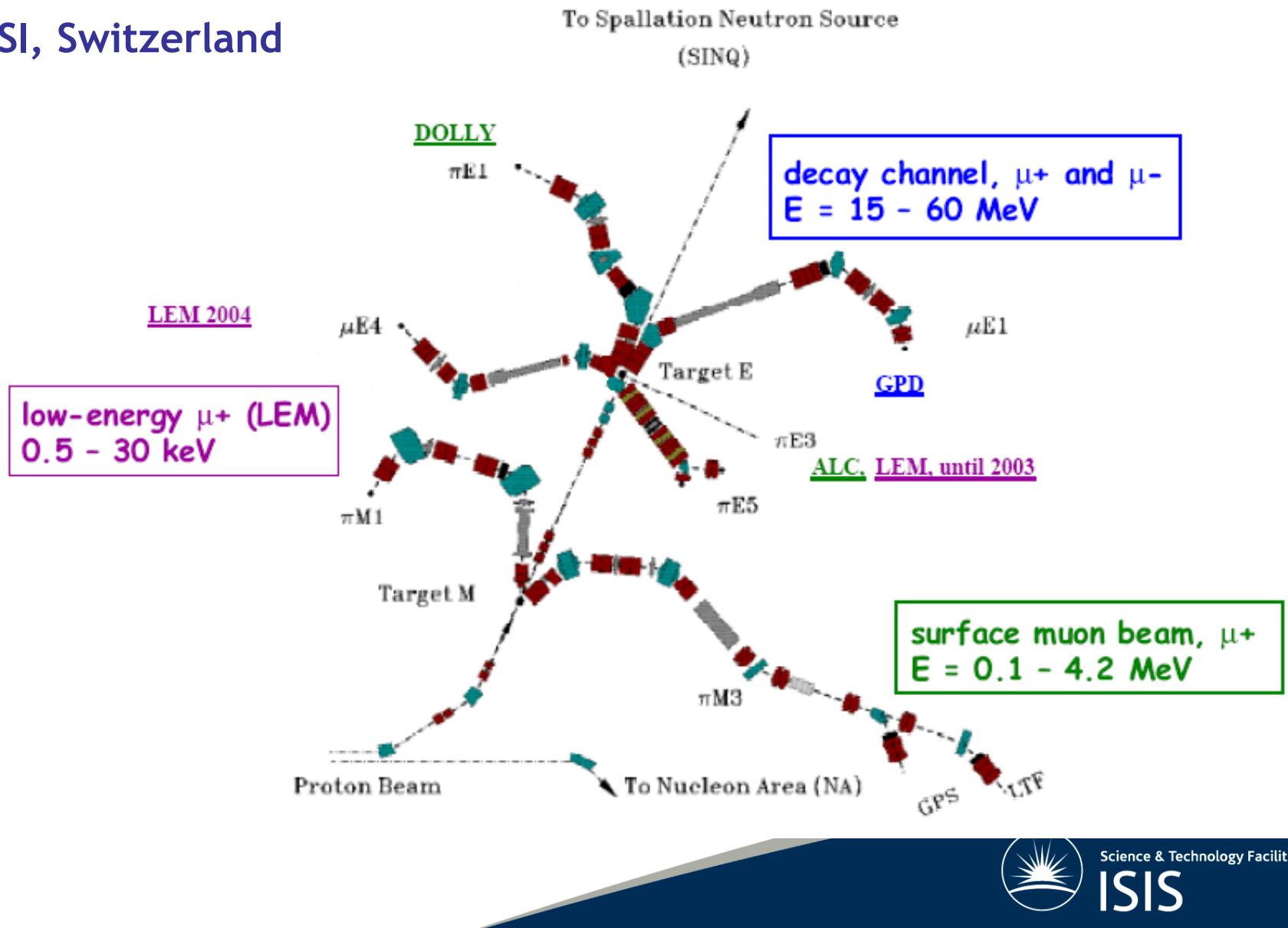


μ SR facilities



μ SR facilities

PSI, Switzerland



Science & Technology Facilities Council

ISIS

μ SR facilities

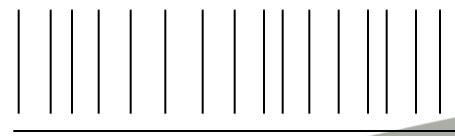
Pulsed (e.g. ISIS, J-PARC)

- Bursts of muons, ~50 Hz
- Low intrinsic background
- Weak relaxations, slow precession
- No fundamental rate limit
- Big detector arrays
- *Pulsed environments*



Continuous (e.g. PSI, TRIUMF)

- Single muons
- Higher intrinsic background
- Fast relaxations, rapid precession
- Data rates limited
- Small, compact detector arrays
- *PSI: low energy muons for surface studies, + pressure*



6. Summary

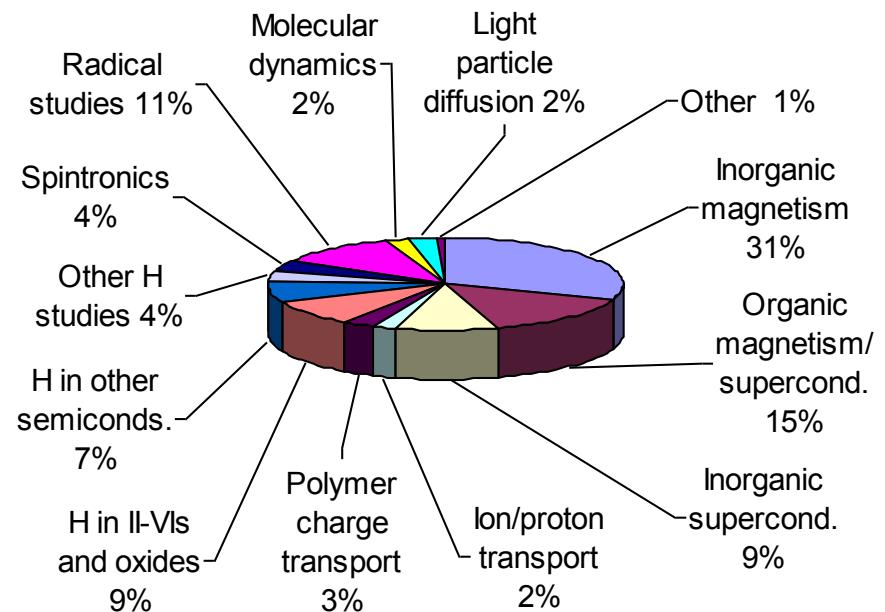
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**"You're not allowed to use
the sprinkler system to keep
your audience awake."**

MUONS:

- Versatile probes of magnetic, superconducting, molecular systems
- Analogues of protons/hydrogen in semiconductors
- Complementary to other techniques ISIS / PSI / J-PARC all have n and μ facilities
- Around 60 groups from 20 countries using ISIS muons
- Further details:



www.isis.stfc.ac.uk/groups/muons/



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