

Particle Accelerator-driven Muon Spectroscopy: An invaluable tool to understand our material world

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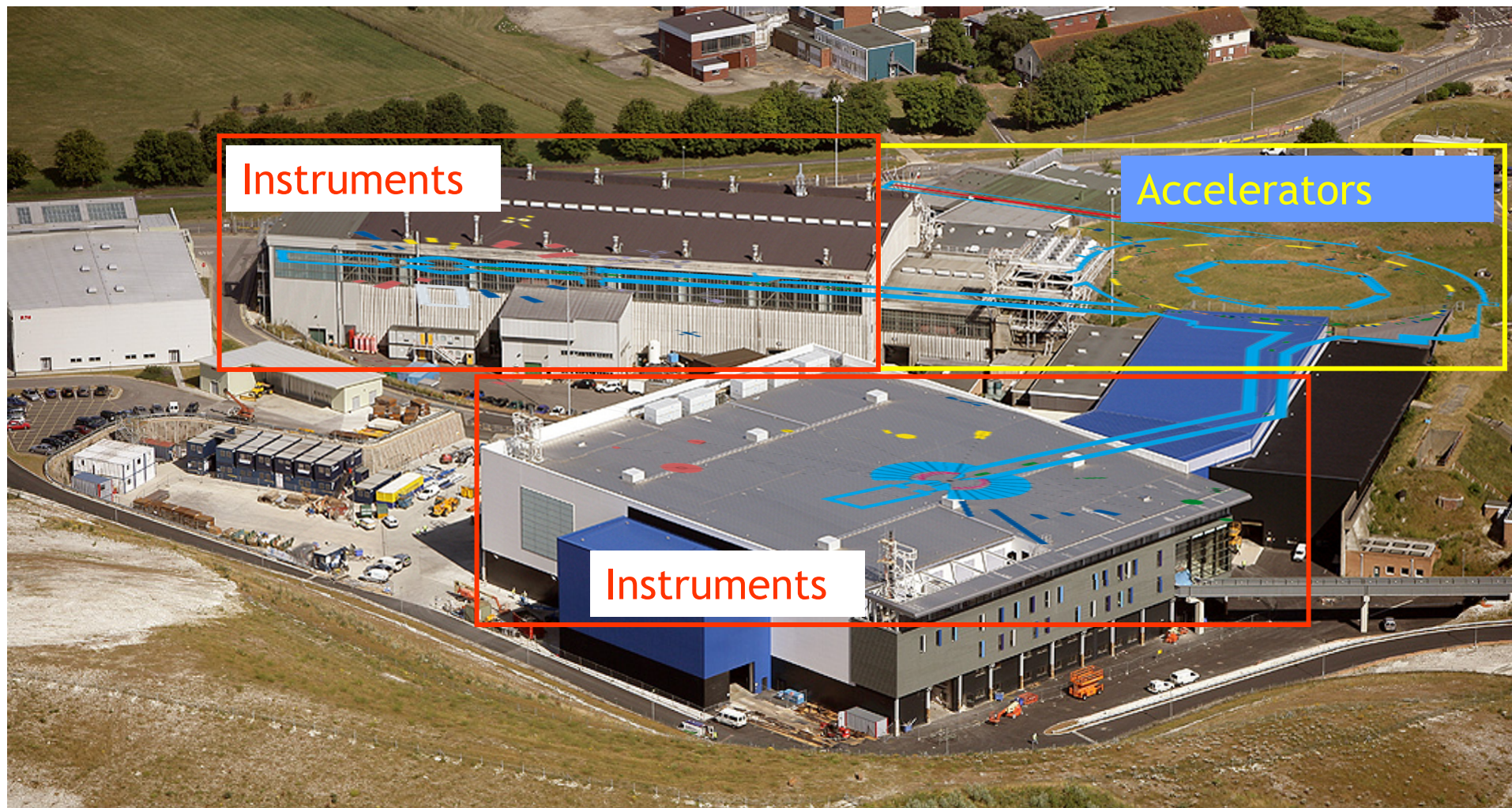
ISIS Neutron and Muon Source



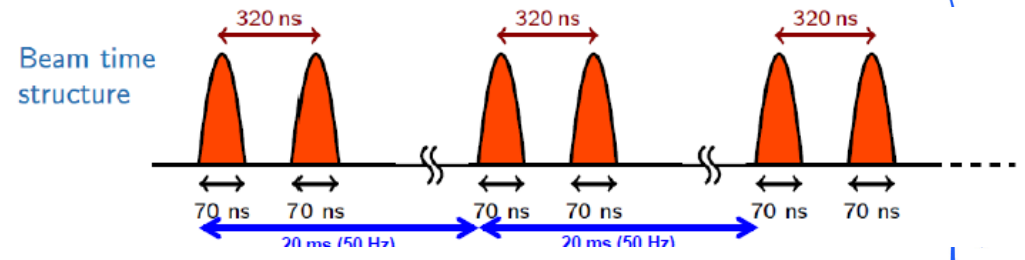
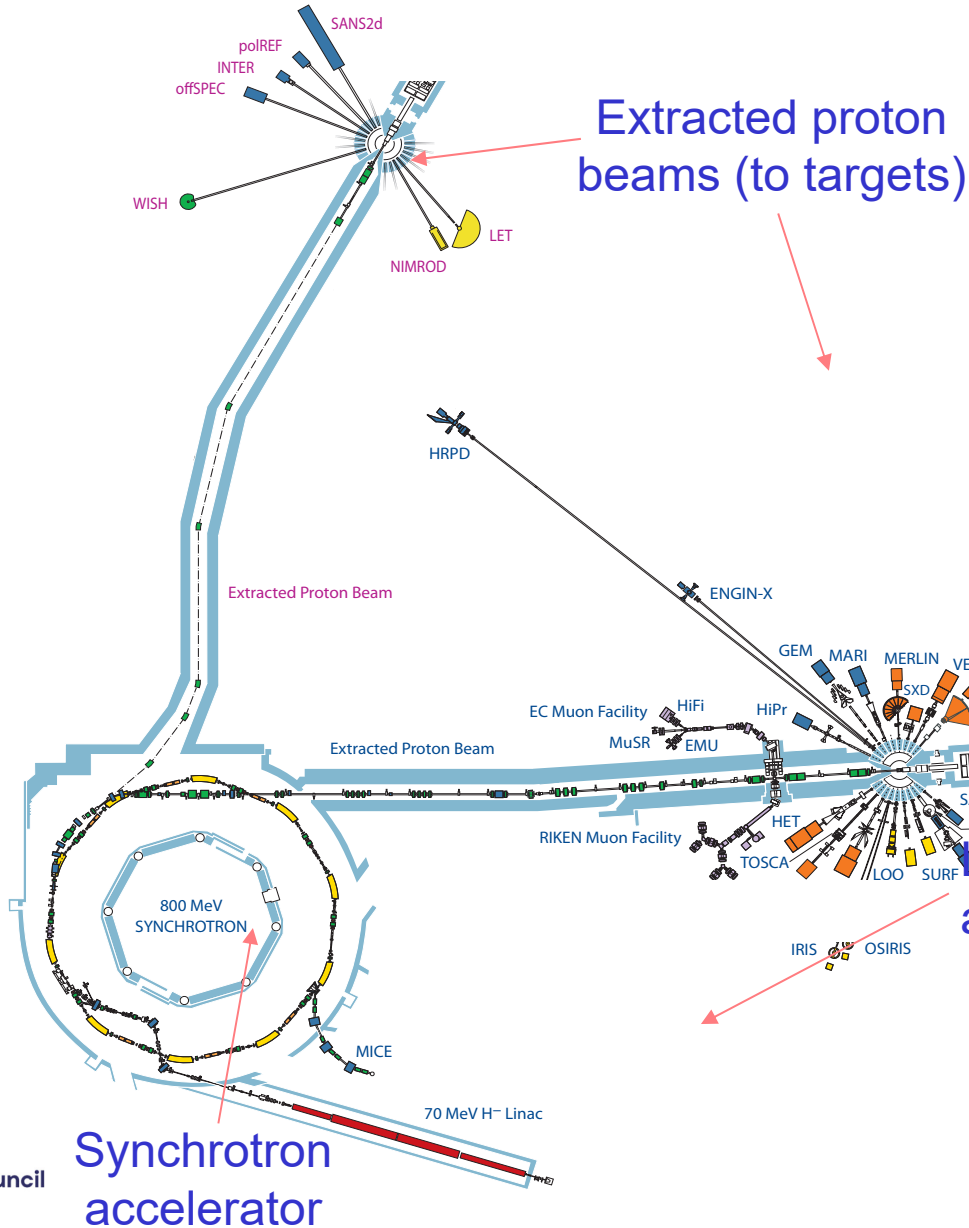
The Rutherford Appleton Laboratory



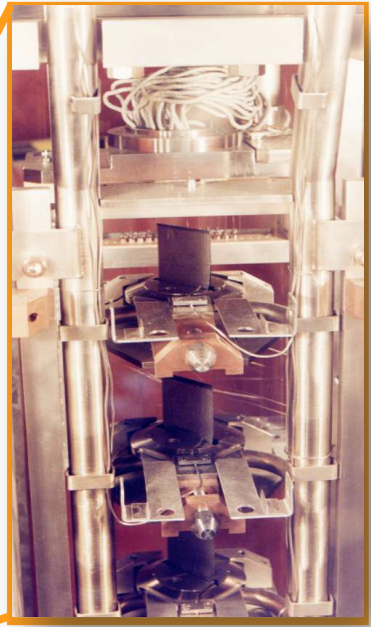
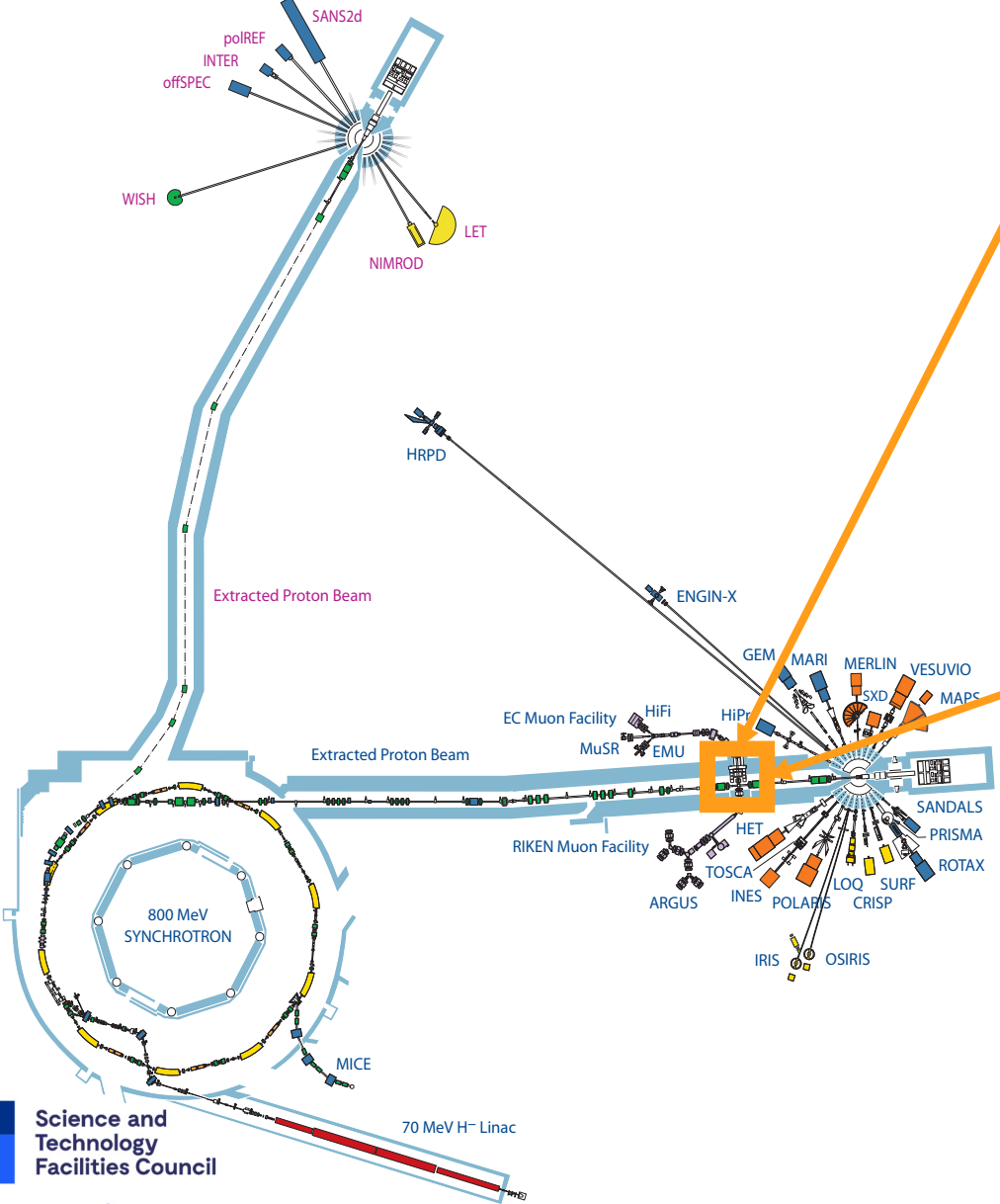
The ISIS Pulsed Neutron and Muon Source



The ISIS Accelerators

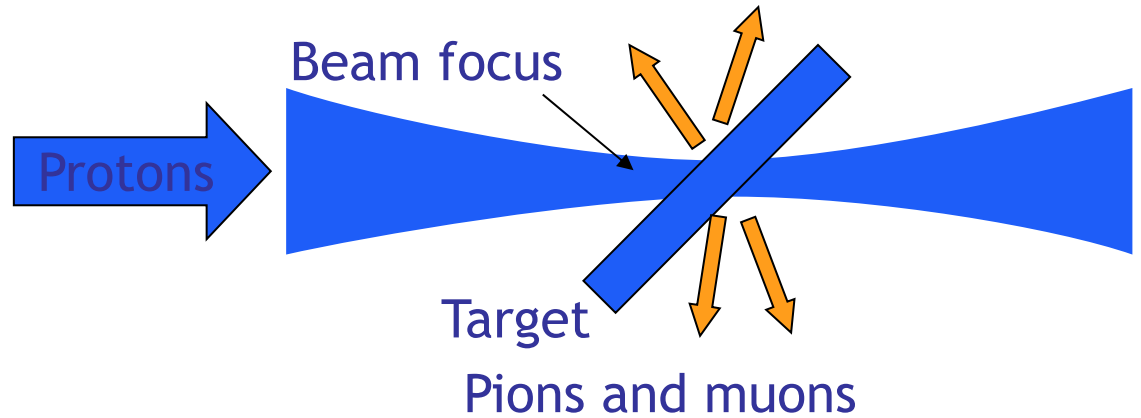


The ISIS pulsed neutron and muon source



Muon target

- Graphite target
- Takes ~3-5% of the proton beam



Key properties of the muon II

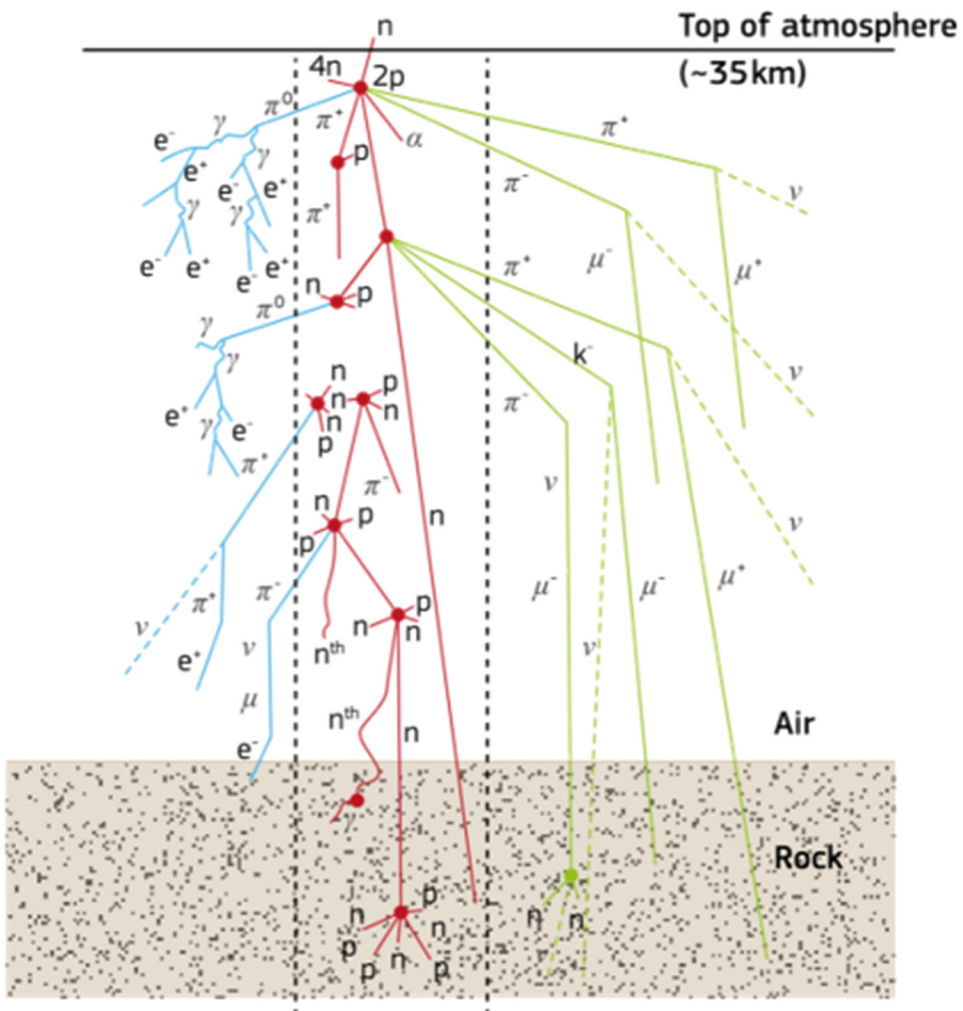
	p (NMR)	μ (μSR)	e (ESR)
Charge	$\pm e$	$\pm e$	$\pm e$
Spin	1/2	1/2	1/2
Mass	$1836m_e$ $=938\text{MeV}/c^2$	$207m_e$ $=105.7\text{MeV}/c^2$	m_e $=0.511\text{MeV}/c^2$
Lifetime	$>10^{33}$ years	2.2 μ s (except for μ^- capture)	$>10^{26}$ years
Magnetic moment	μ_p	3.18 μ_p	657 μ_p
Gyromagnetic ratio (kHz/G)	4.26	13.5	2800
Spin polarization	Needs $\sim 2.5\text{-}5\text{T}$ for $P < 73\%$	Produce μ^+ with nearly $P=100\%$	Needs some magnetic field

Major constituent of cosmic rays AND produced at accelerator facilities for particle physics and condensed matter research.

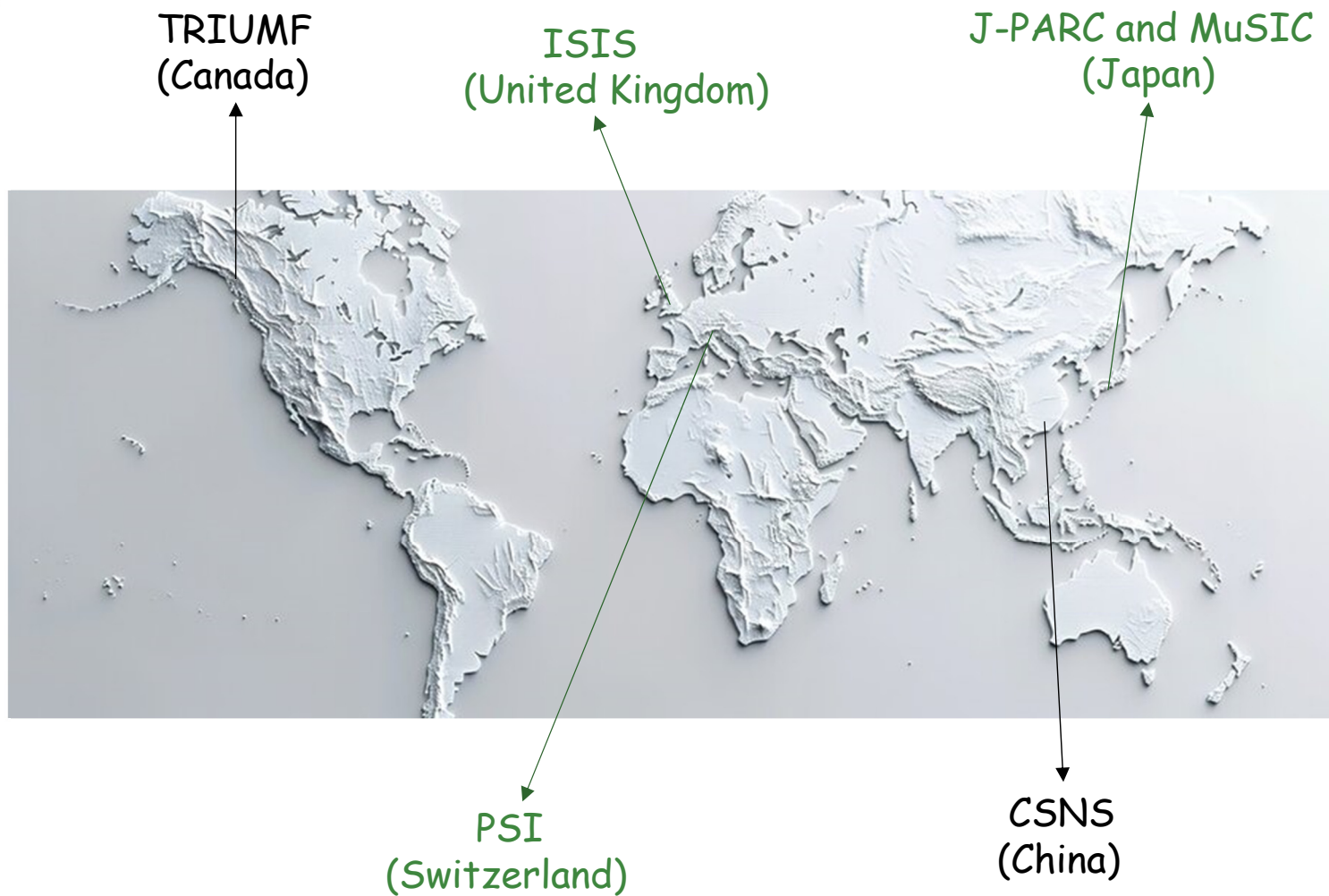


Muon Sources

Natural source: Cosmic rays

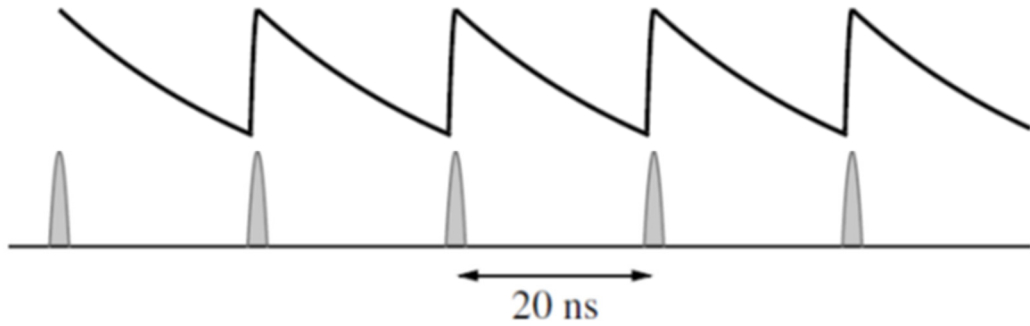


Human-made source: Accelerators



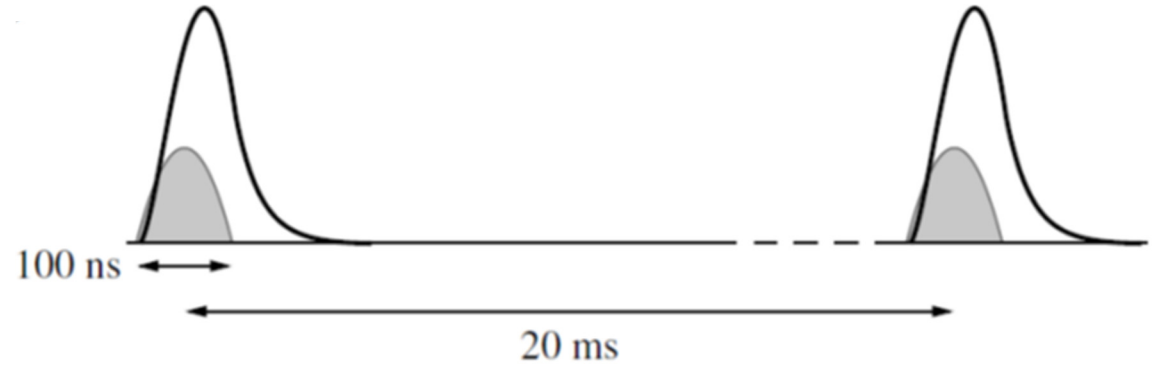
Pulsed and continuous sources of muons

Continuous source (e.g. PSI, TRIUMF)



- Single muons
- Higher intrinsic background
- Time resolution set by electronics (~ 10 ps)
-> fast relaxations, rapid precession
- Rate limit set by single muon at a time constraint
- More compact detector arrays

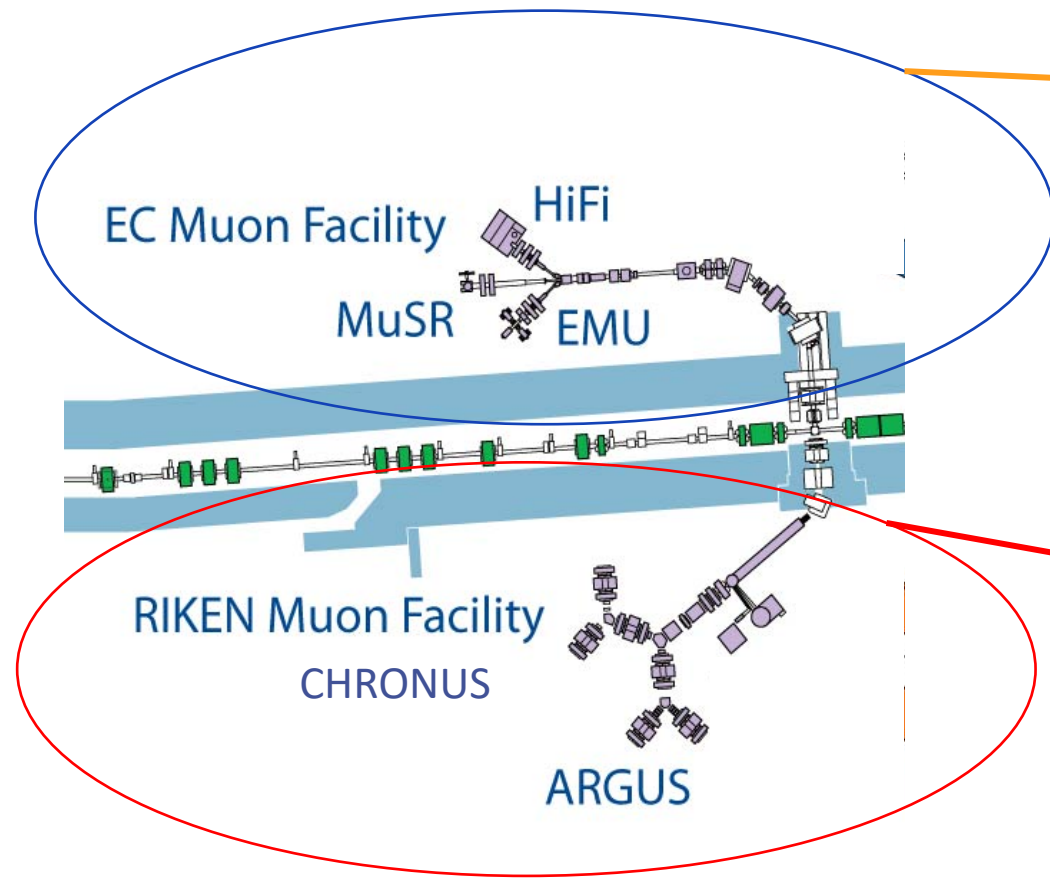
Pulsed source (e.g. ISIS, J-PARC)



- Bursts of many muons
- Low intrinsic background
- Time resolution set by muon pulse width (~ 50 ns)
-> Weak relaxations, slow precession
- Rate limit set by detector dead time
- Big detector arrays to cope with high instantaneous rate



Muons at ISIS



Surface Muon beam
100% spin polarised
Fixed momentum ~ 27 MeV/c
Pulses split between beamlines

Decay Muon beam
Positive or negative muons
Spin polarised
Variable momentum ~ 15 - 120 MeV/c
Pulses can be split between beamlines



Muon production – surface, decay and low energy muons

Surface -> 4MeV (10-100 μm)

- Decay of π^+ at rest near the target surface.
- Optimal energy 4MeV (29MeV/c)
- Nearly 100% spin polarised
- Large fraction of muon experiments use surface muons

Decay * -> 1-100 MeV (1-1000 μm)

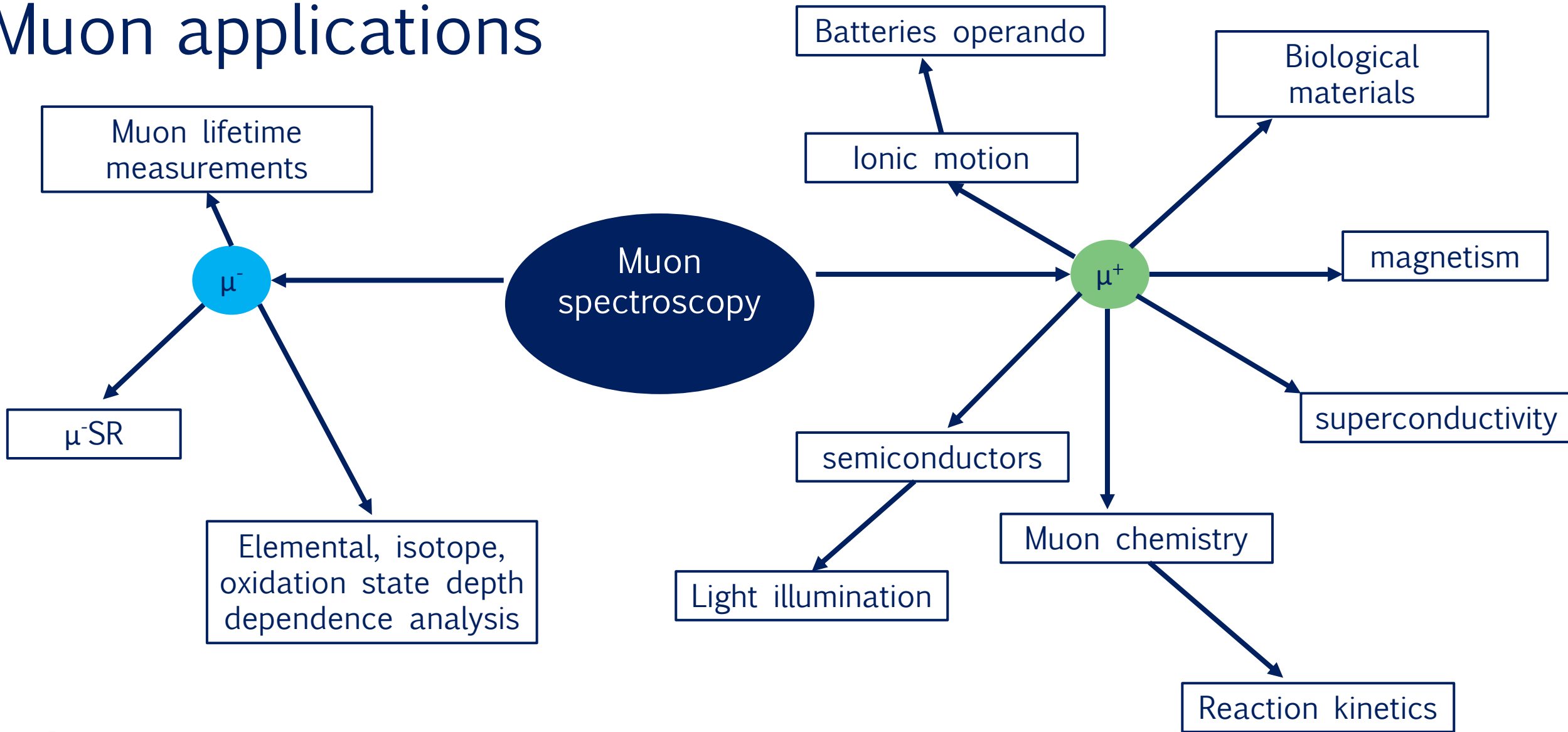
- Produced when finite momentum pions leave the target and decay in flight.
- Can be μ^+ or μ^-
- Range of momentum possible (0 – 120MeV/c)
- Up to 80% spin polarised
- Enables measurements of samples in complex sample environments.

Low E -> 1-30keV (1-100 nm)

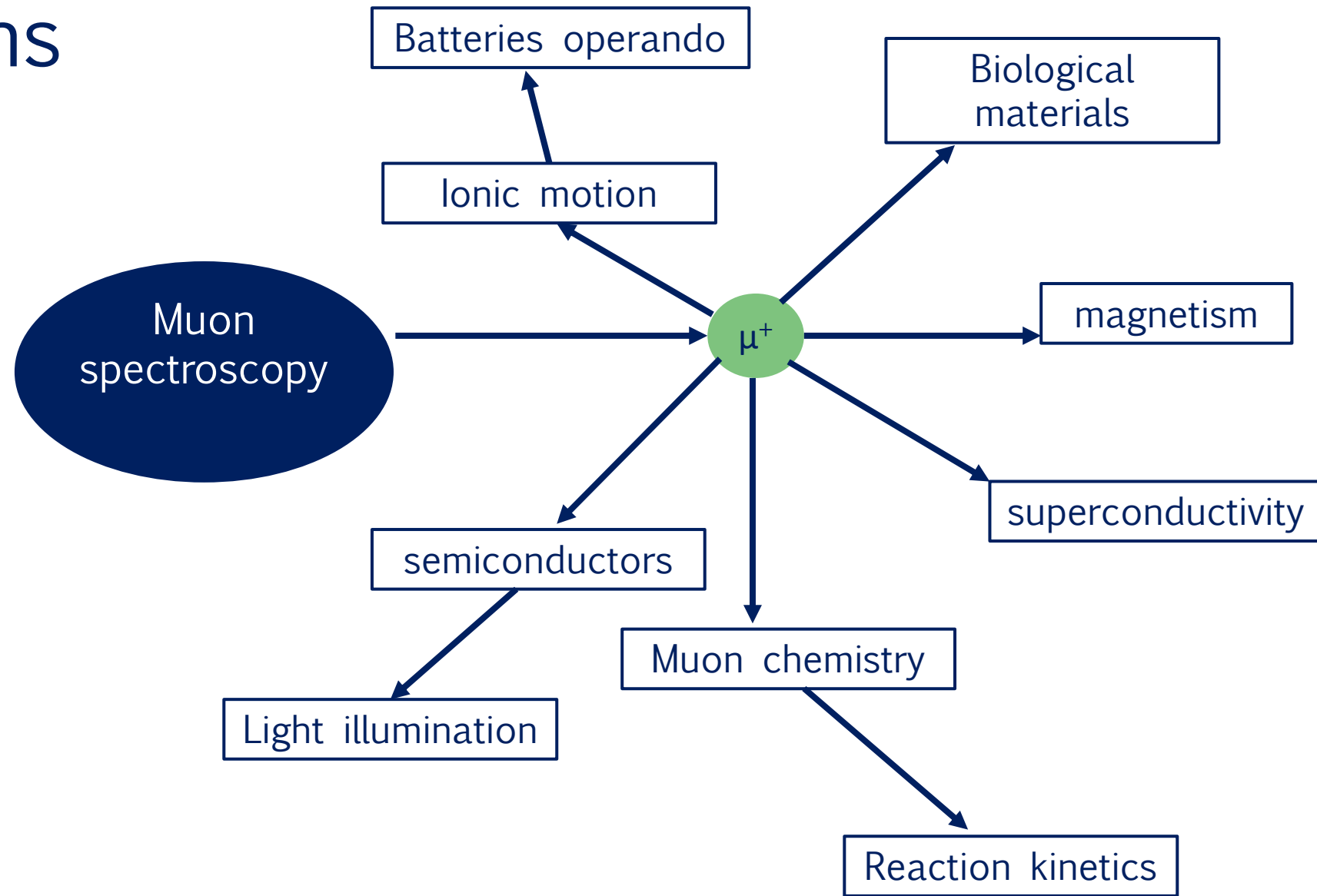
- Surface muons can be moderated down to lower energies.
- Range of energy possible by acceleration with electric fields (\sim 1-30keV)
- Nearly 100% spin polarised
- Enables measurement of thin films, surfaces and interfaces.
- Currently only at PSI (in development at J-PARC)



Muon applications

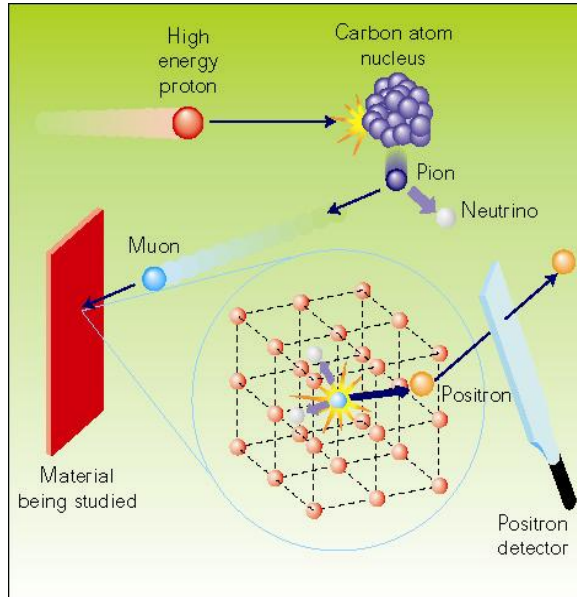


Muon applications

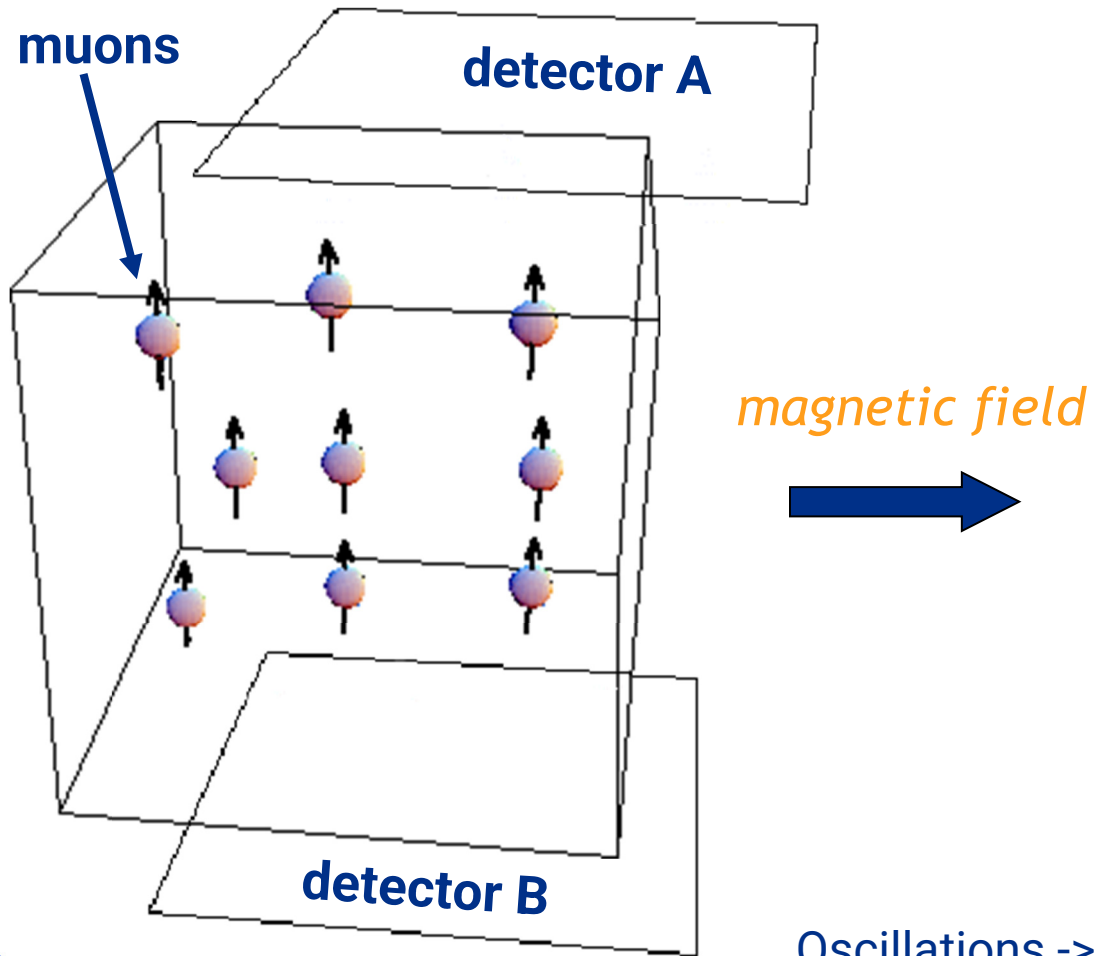


The muon spectroscopy technique – the muon lifecycle

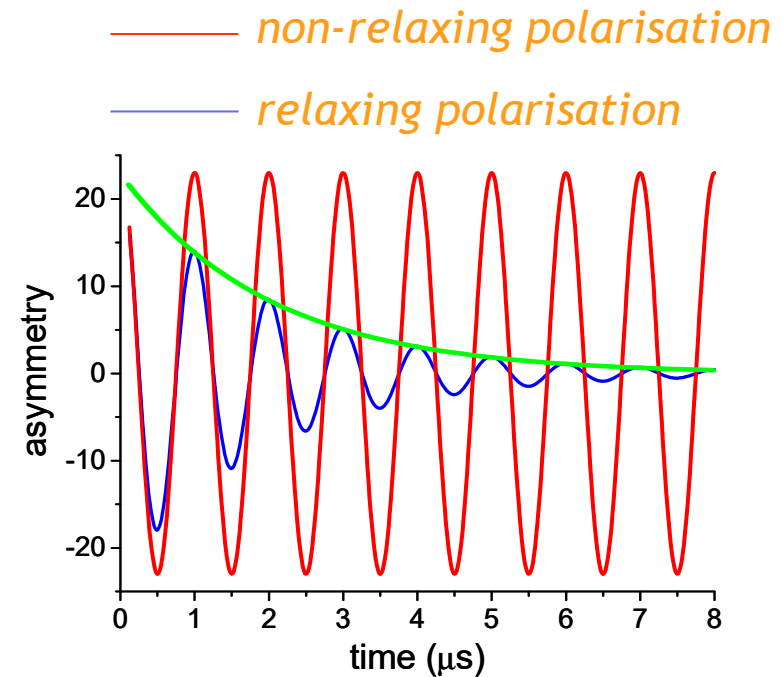
- 1 Pion production: high energy protons collide with C nuclei in graphite target.
- 2 $\pi^+ \rightarrow \mu^+ + \nu_\mu$
muons are 100% spin polarised
- 3 Implantation,
surface muons \rightarrow 0.1-1mm,
decay muons \rightarrow 0.1-10mm
and Low Energy \rightarrow 10-100nm.
- 4 Muons interact with local magnetic environment
- 5 Decay, lifetime $2.2\mu\text{s}$
 $\mu^+ \rightarrow e^+ + \nu_e + \nu_\mu$
we detect the decay positrons
- 6 **The positrons are preferentially emitted in muon spin direction**
- 7 Measure the positron distribution to determine the muons' polarisation in time. Learn about the muons' local environment or the muon behaviour itself.



The muon spectroscopy technique



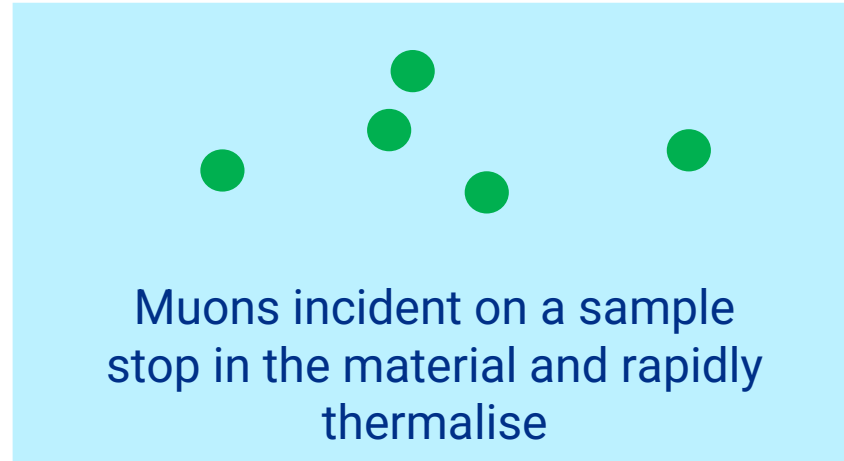
$$\text{'asymmetry' } (t) = \frac{N_A(t) - N_B(t)}{N_A(t) + N_B(t)} = a_0 \mathbf{G}_x(t) \cos(\omega t)$$



Oscillations -> magnetic order across muon stopping sites
 Damping -> inhomogeneous fields that depolarise the muon spins



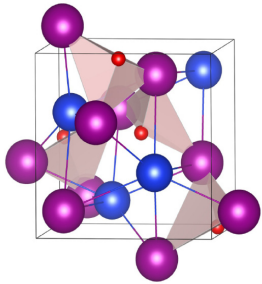
Properties of the muon – muon states in matter



1

Muon implants at interstitial site.

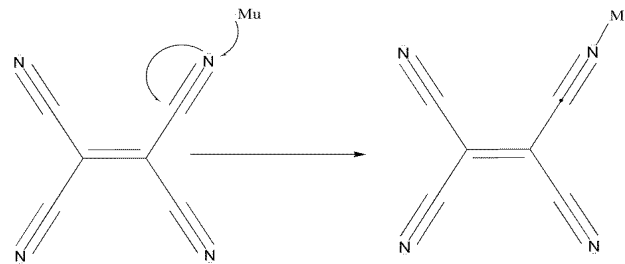
Attracted to regions of negative potential.



2

Diamagnetic muonium.

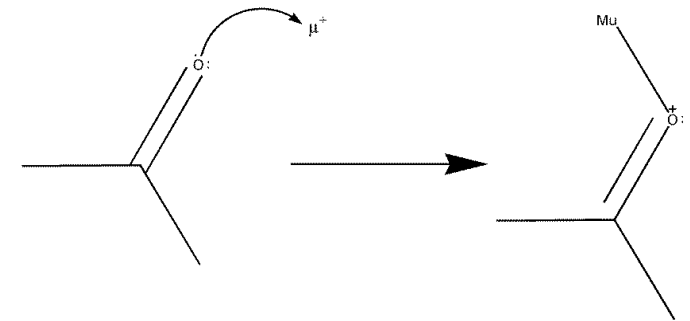
Muon captured by electron lone pair on an atom.



3

Paramagnetic muonium.

Muon mimics hydrogen ion and interacts with sample.

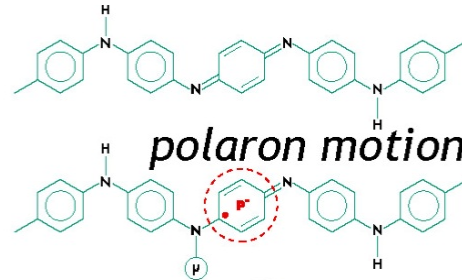
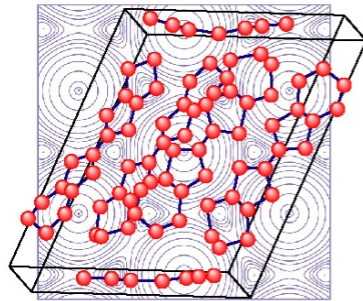


Muon science areas

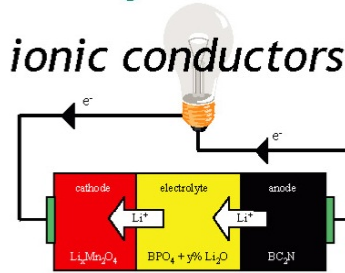
Muons as passive probes in superconductivity, magnetism, molecular dynamics, charge transport.

Muons as active probes: proton analogues in semiconductors, proton conductors, light particle diffusion, etc.

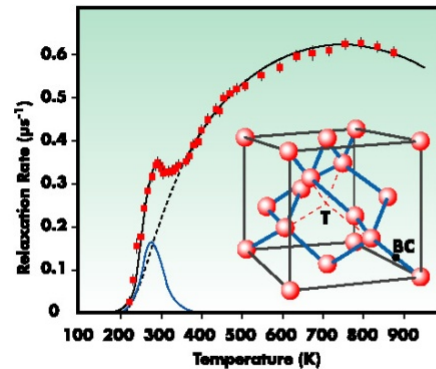
superconductors



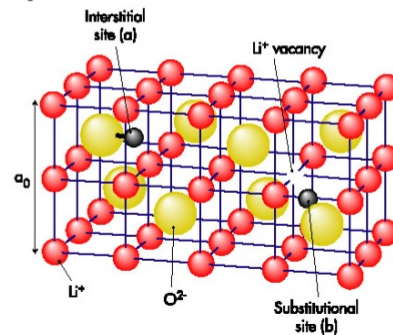
ionic conductors



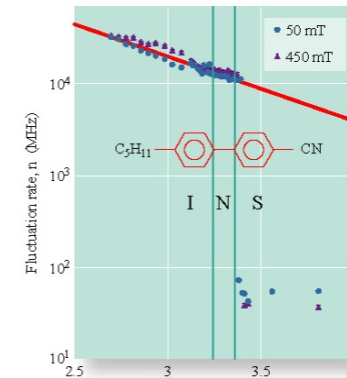
semiconductors



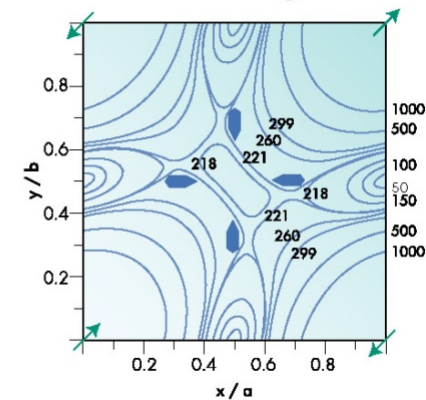
proton conductors



molecular dynamics

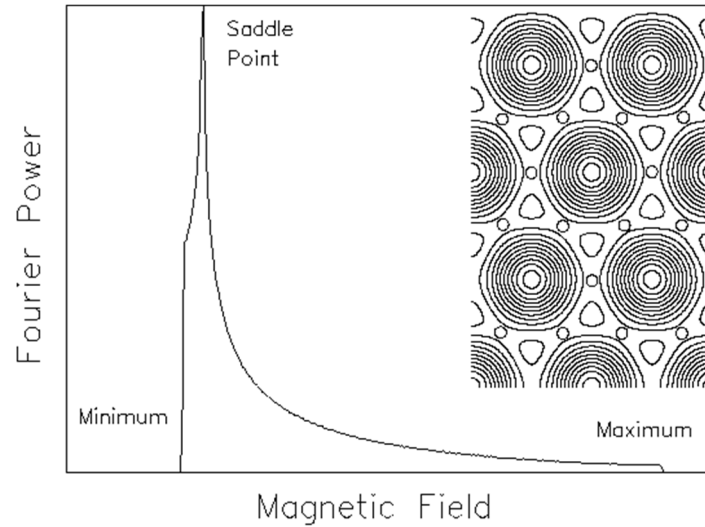
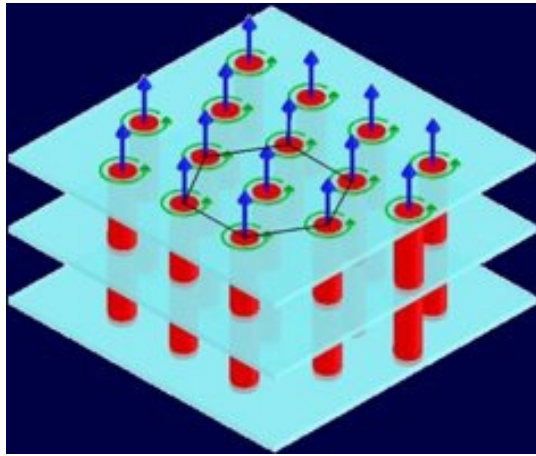


magnetism

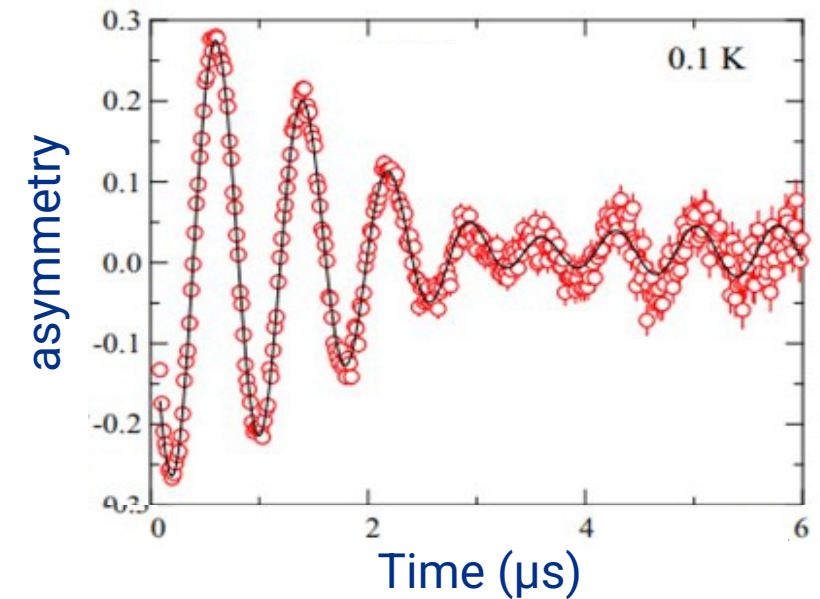
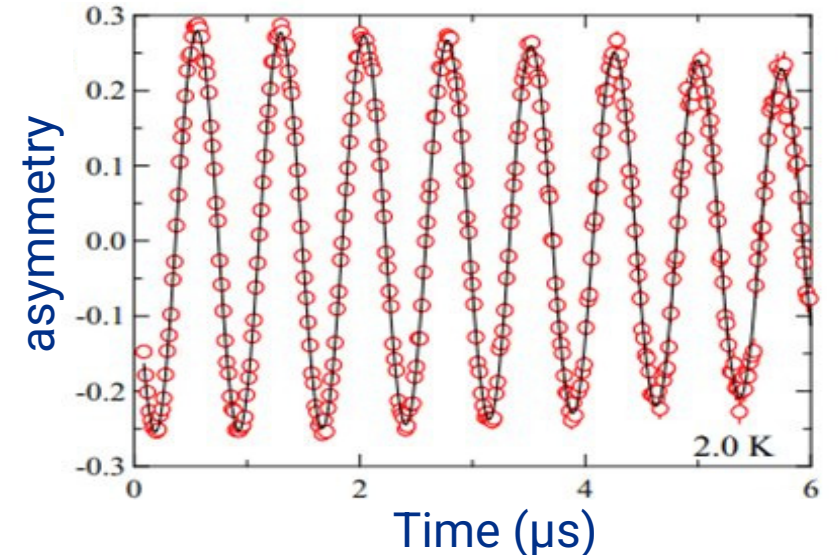


Scientific Highlights I – Superconductivity

TF- μ SR - measures the vortex lattice field distributions





Muons measure a distribution of fields that depends on the penetration depth λ and the coherence length ξ .

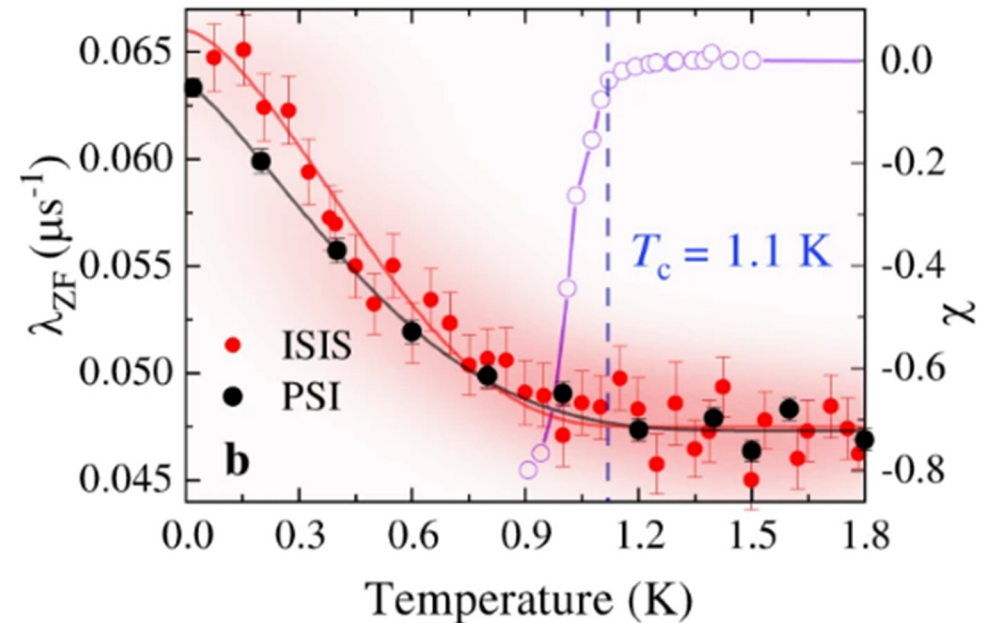
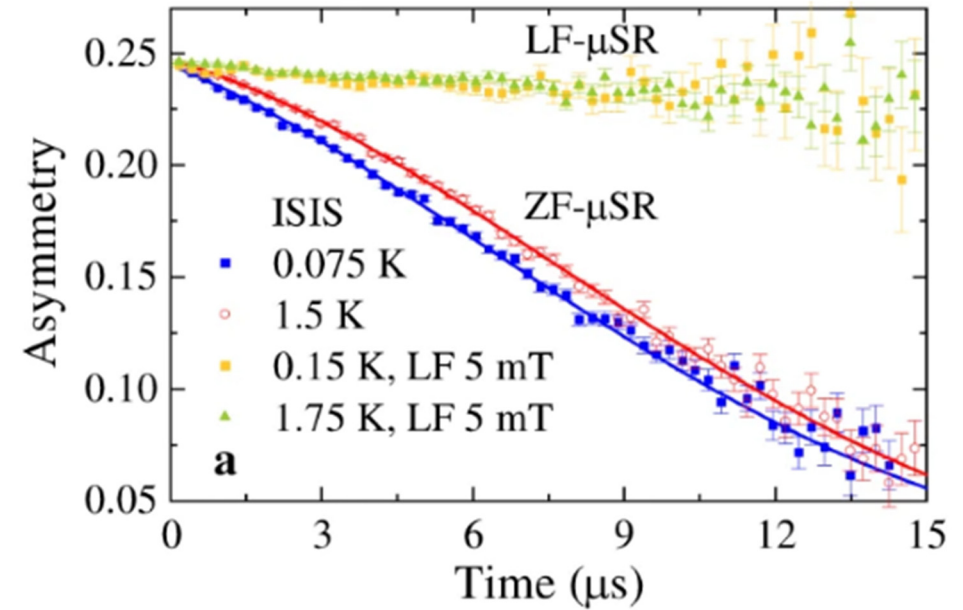
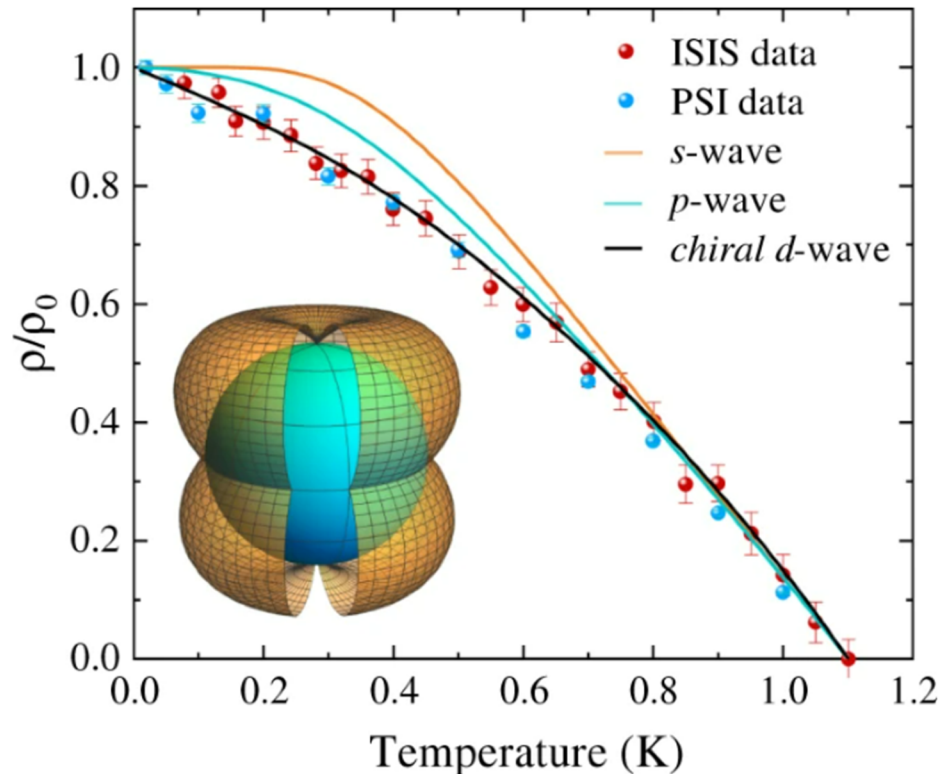


Science highlights I: chiral superconductivity in LaPt_3P – ZF and TF μSR

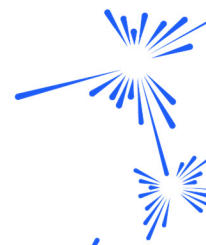
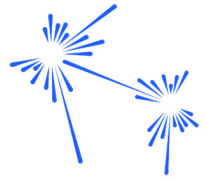
Chiral singlet superconductivity in the weakly correlated metal LaPt_3P

[P. K. Biswas](#) , [S. K. Ghosh](#) , [J. Z. Zhao](#), [D. A. Mayoh](#), [N. D. Zhigadlo](#), [Xiaofeng Xu](#), [C. Baines](#), [A. D. Hillier](#),
[G. Balakrishnan](#) & [M. R. Lees](#)

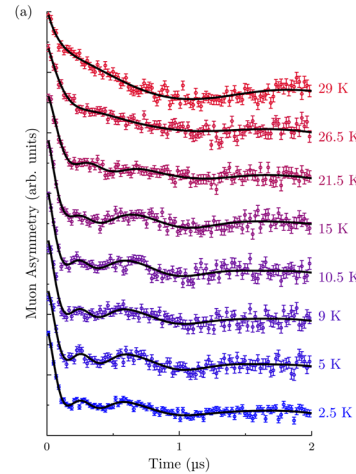
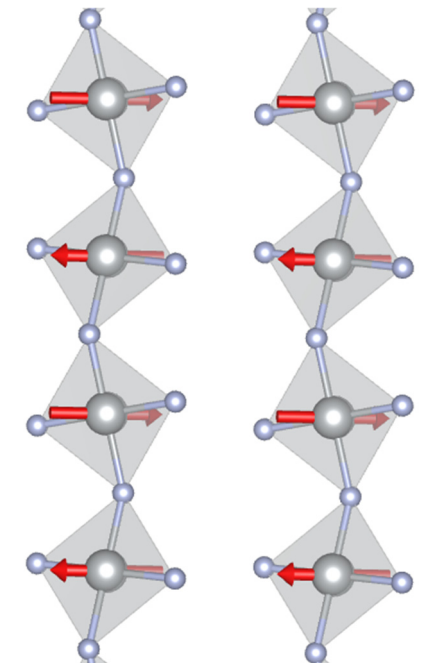
[Nature Communications](#) **12**, Article number: 2504 (2021) | [Cite this article](#)



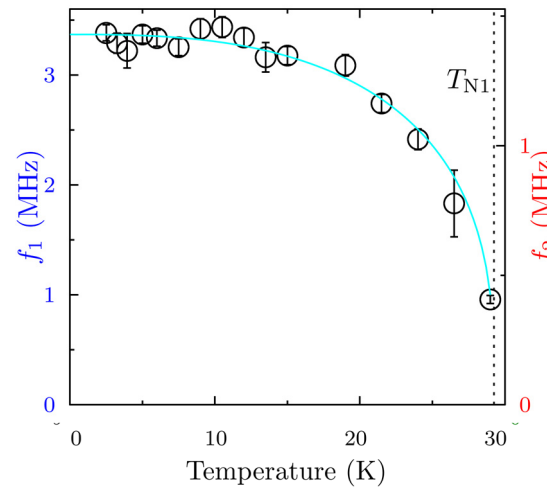
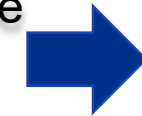
Science highlights II – KAgF_3 : Using F-- μ --F states to measure magnetic materials



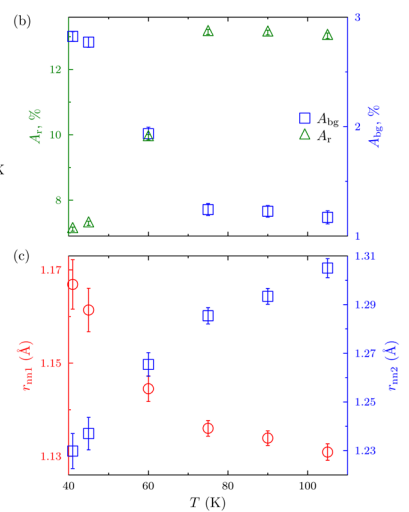
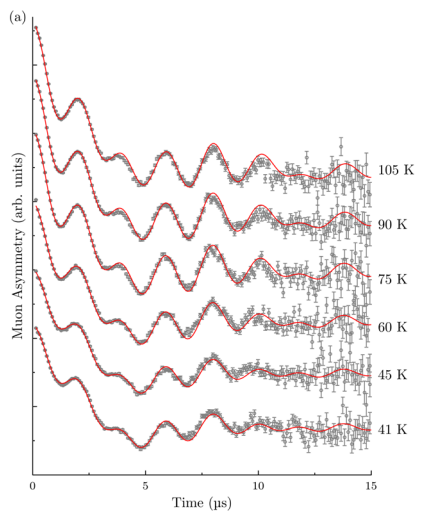
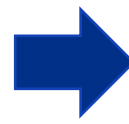
Discover an A-type AFM structure with $\mu_{\text{Ag}} = 0.47\mu_{\text{B}}$



Use magnetic oscillations to find the magnetic structure below T_c



Use F-- μ --F states above T_c and DFT to find the muon site



Scientific Highlights III – Battery materials

Dynamics from Ionic Motion

- Spin-polarised muon stops in the sample
- Fields from nearby nuclei cause incoherent spin precession
- Ion with nuclear moment moves near muon and flips the muon spin (or vice versa)
- Muon decays and positron detected
- Signal depends on how often the muon spin was flipped

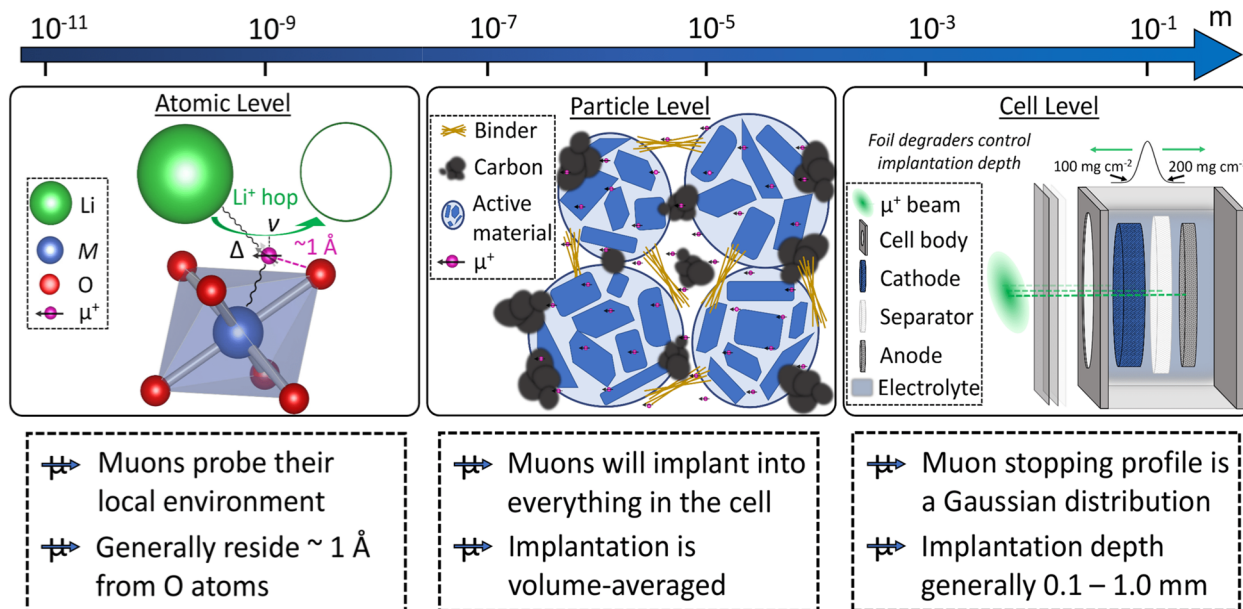
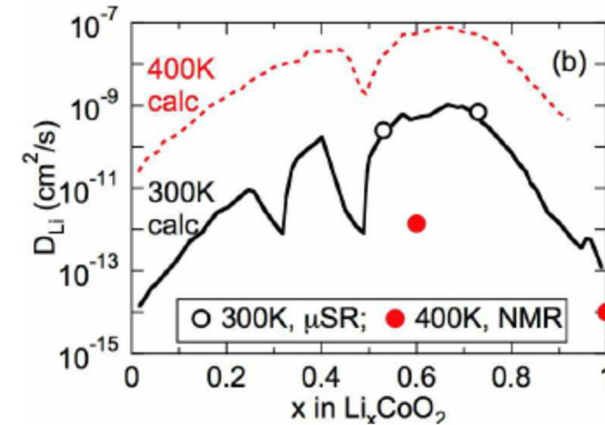
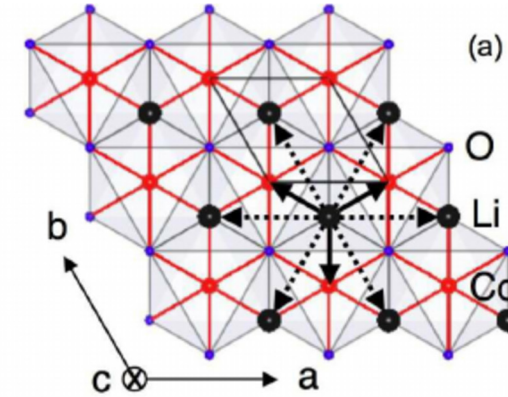


Figure from Innes McClelland



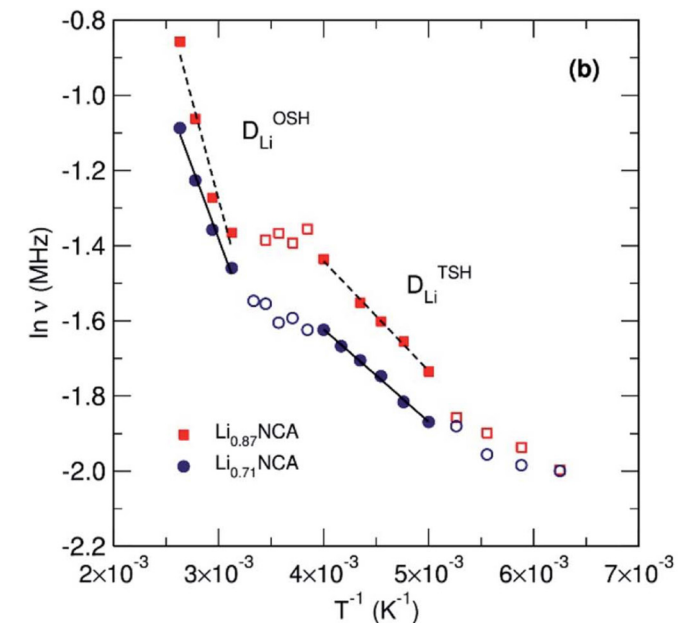
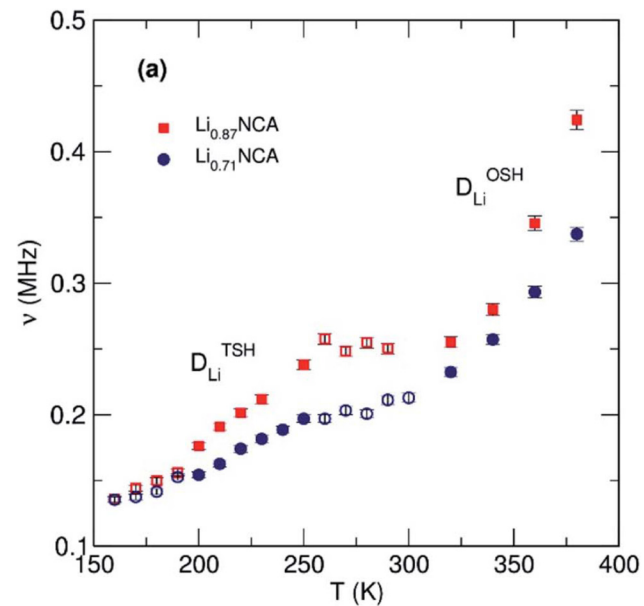
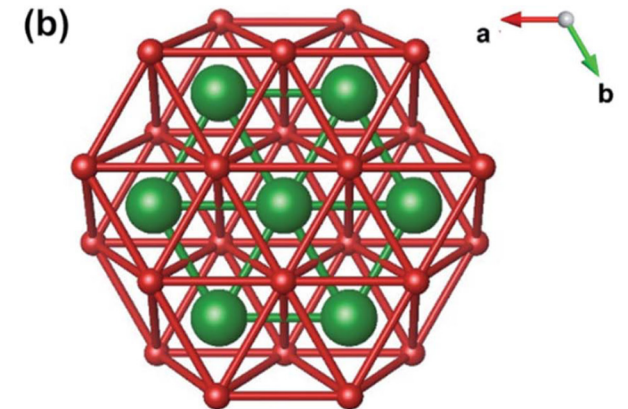
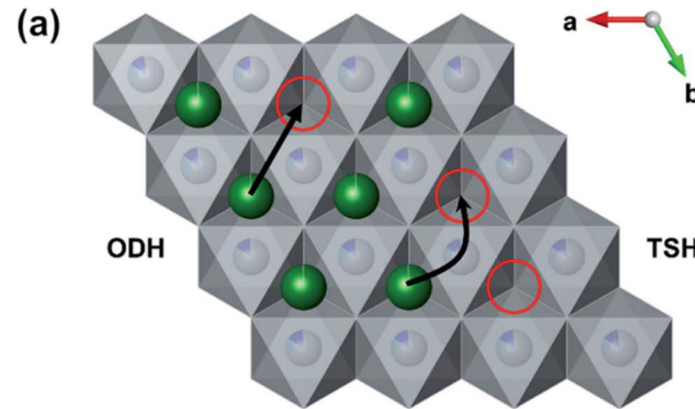
J. Sugiyama et al, Phys. Rev. Lett. 103, 147601 (2009)

Muons (both positive and negative) allow these effects to be probed over a range of lengthscales in, for example, battery materials

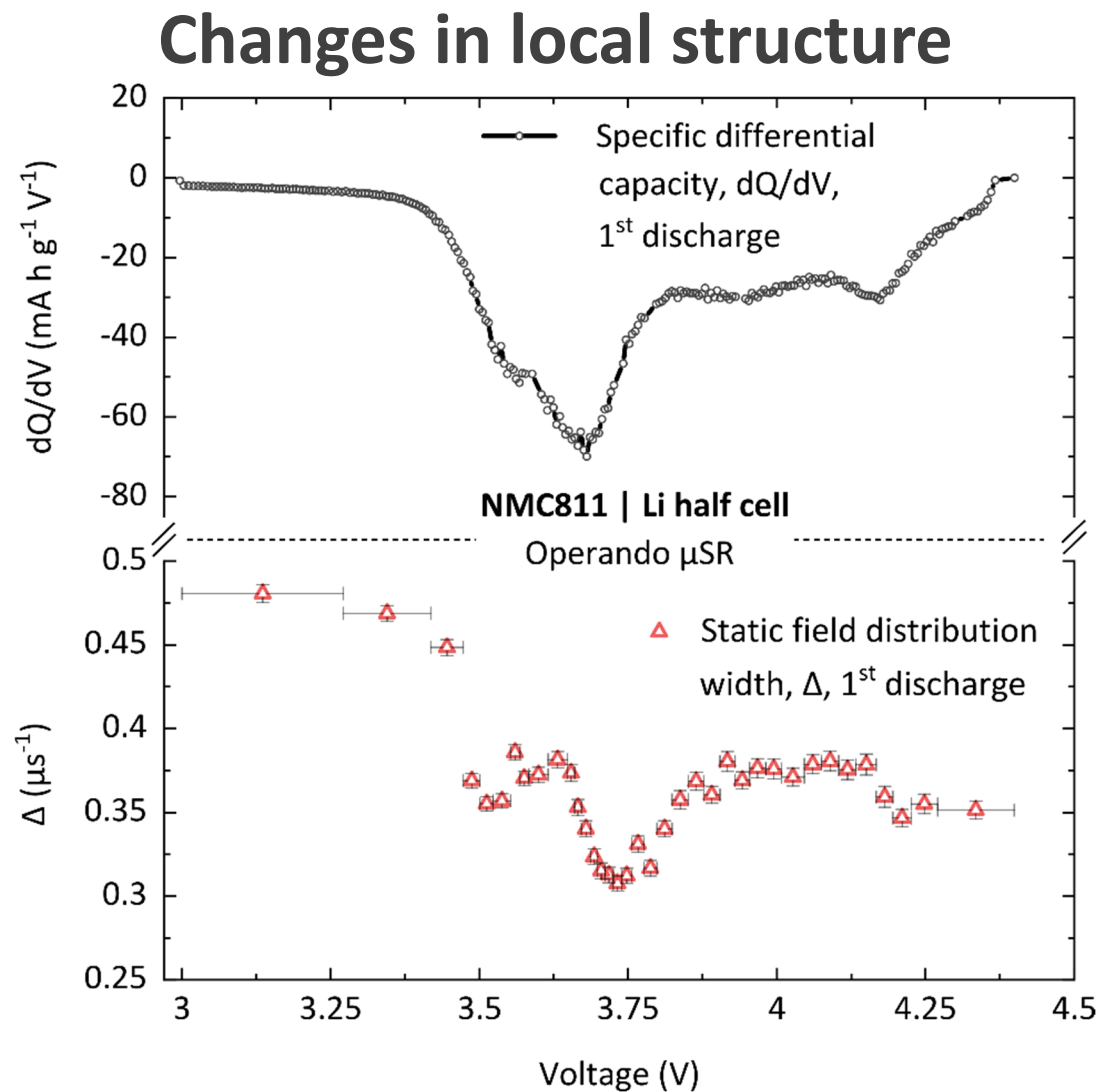
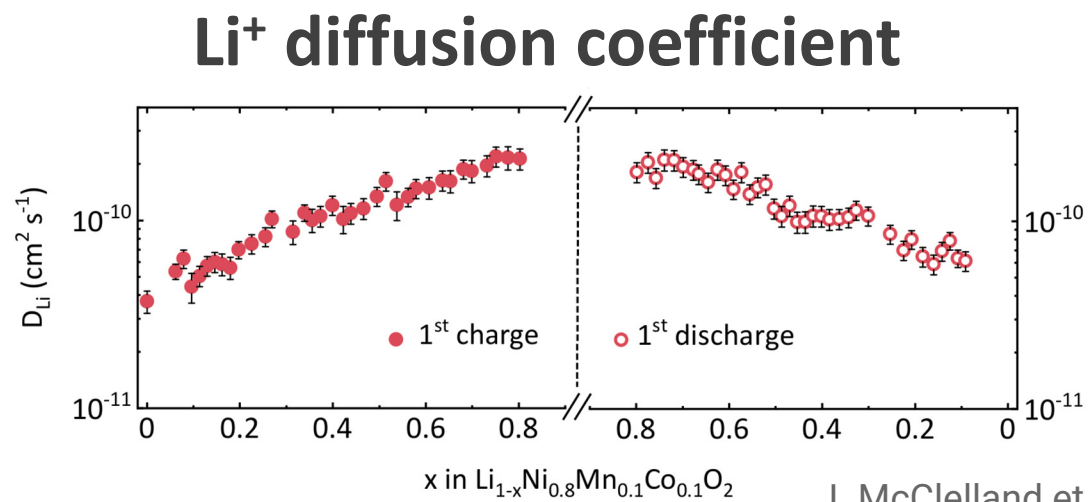
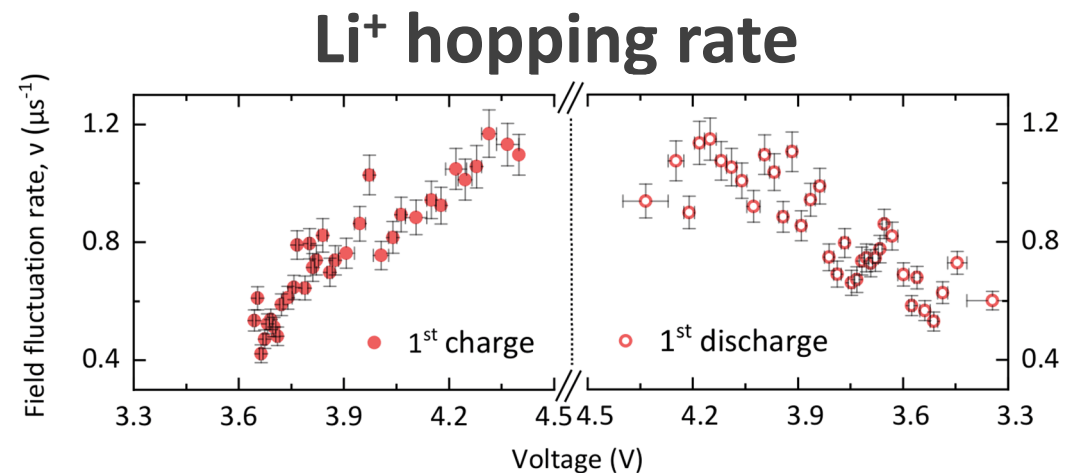


Scientific Highlights III – Battery materials (Ex Situ)

- Primary use of muons is to measure local ionic motion
 - Works in paramagnetic materials
 - Material doesn't have to be in a cell
 - Ion must have a nuclear magnetic moment, e.g. Li, Na, K, Mg, I
- Measure the hopping rate as a function of temperature
 - Extract energy barrier(s)
 - Find hopping rate \sim MHz and derive local diffusivity for available pathways

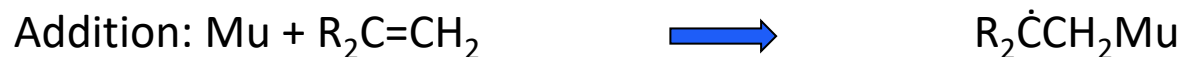
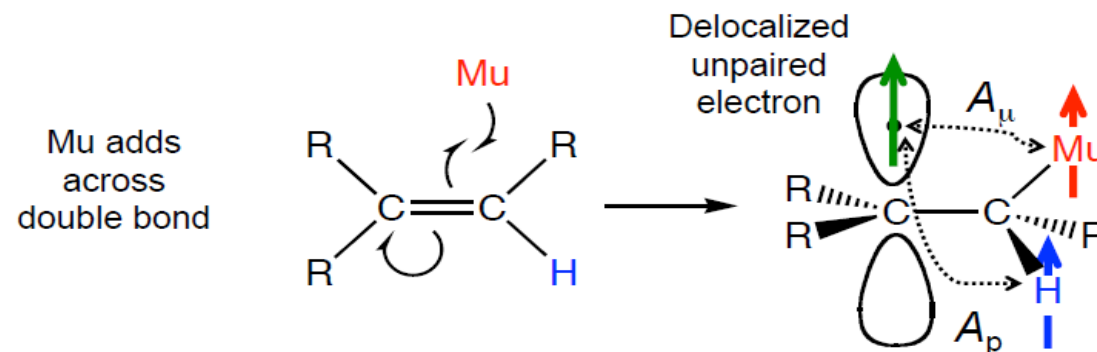
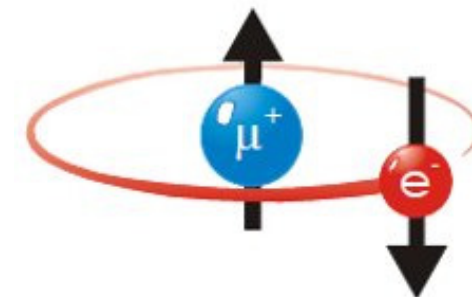


Scientific Highlights III – Battery materials (In Operando)



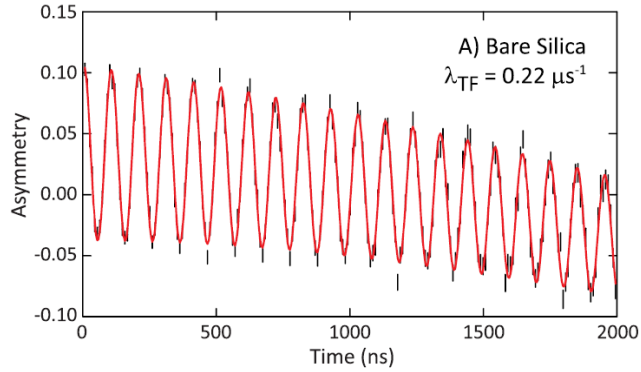
Scientific Highlights IV – Muonium Chemistry

- Positive muons can capture electrons to form a light hydrogen isotope: Muonium (Mu)
- Mu acts as a radioactively labelled light isotope of hydrogen (similar Bohr radius, ionisation energy and reduced mass)
- Mu can undergo the same reactions as a H-atom:

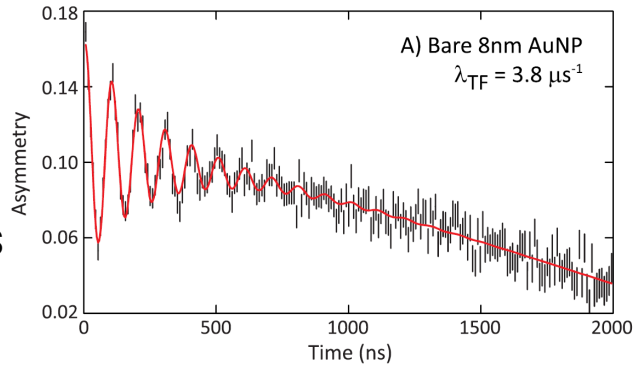


Scientific Highlights IV – Muonium Chemistry

Mu formed in bare silica



Mu reacts with gold nanoparticles



Catalytic effect of Benzene

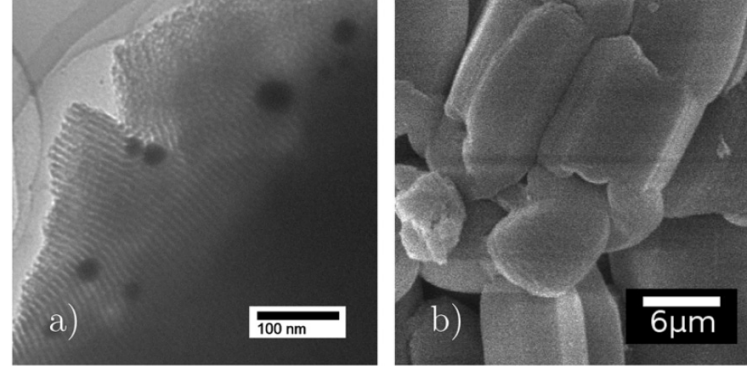
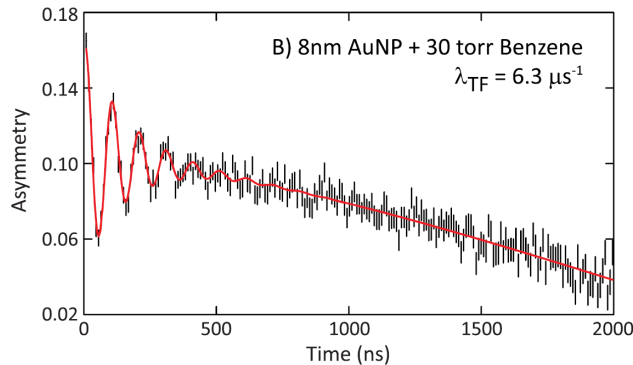


FIG. 1. (a) TEM image showing GNPs embedded in the silica mesopores. Note the apparent size distribution of GNPs. (b) SEM image of the silica grains.

Mu reacting with 8 nm GNPs (6G TF)

Sequence of events:

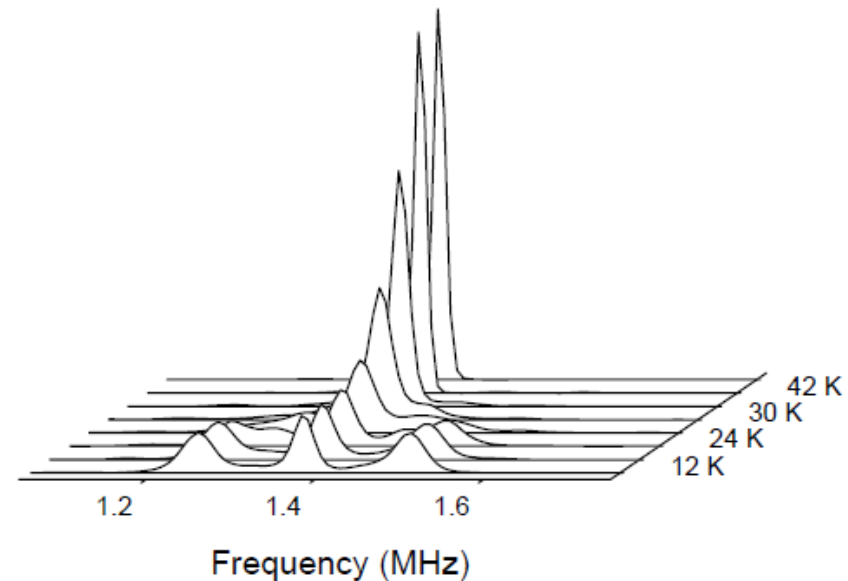
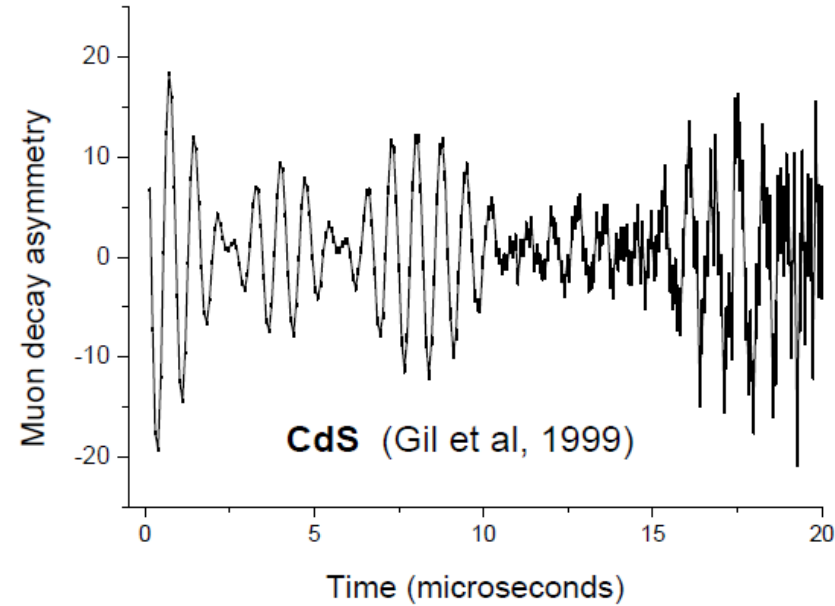
- Mu formed in silica escapes the mesopores,
- Quickly diffuses to a GNP,
- Loses e^- spin to metal, i.e. $\text{Mu} \rightarrow \mu^+$ (chemisorption reaction), hence Mu relaxation

2 - 3 fold catalytic effect from benzene on metal surface



Scientific Highlights VI - Semiconductors

- Generally studying muonium in the semiconductor
- Hydrogen defect analogue
 - Where it forms in the material
 - What dynamics it experiences
- Investigating paramagnetic centres
 - e.g. NV centres in diamond
- Charge carrier dynamics
 - Thermal effects
 - Light induced effects

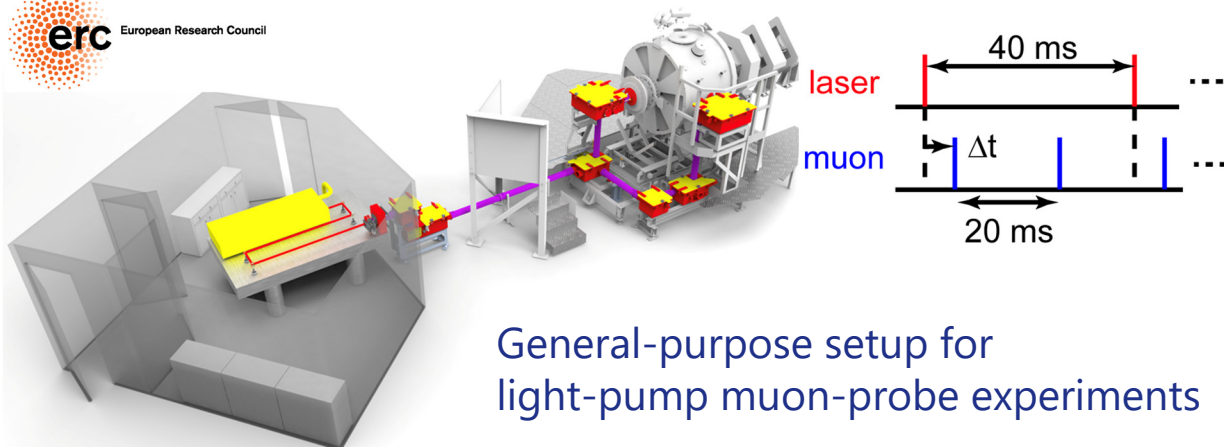
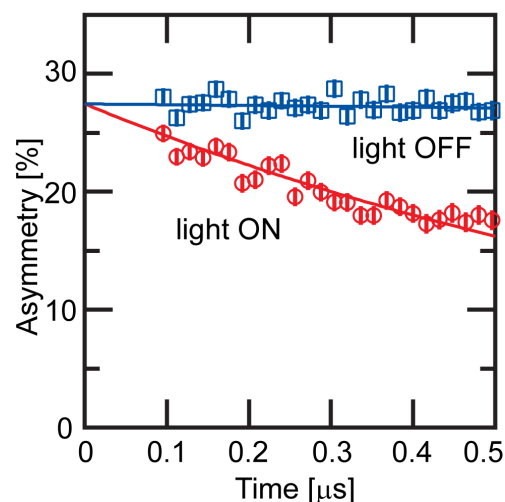


Scientific Highlights VI - Semiconductors

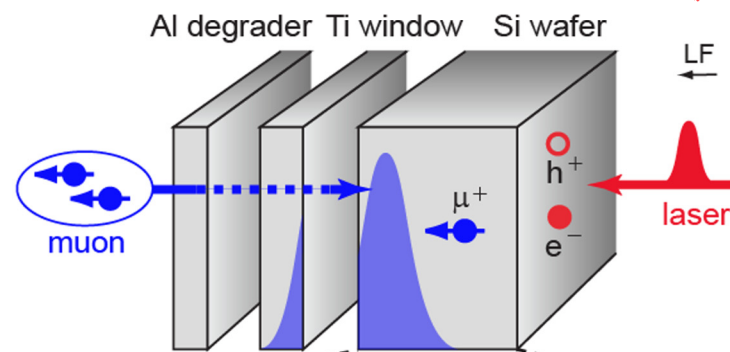
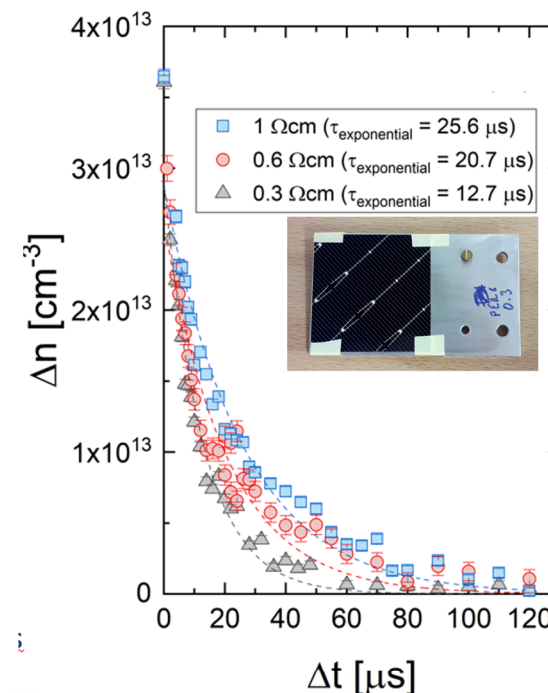
koji.yokoyama@stfc.ac.uk



- Carrier-Mu interaction depolarises muon signal
→ Relaxation rate tells carrier density
- HIFI Laser for optically generating carriers in semiconductors



- By changing the pulse delay, one can measure carrier lifetime spectrum
 - Even in completed solar cells
- Depth-dependent measurements enable carrier kinetics study within a wafer



Applicable to many semiconductors. So far: Si, Ge, SiC, GaAs



ISIS Neutron and Muon Source

Review of Scientific Instruments 87, 125111 (2016).
 Phys. Rev. Lett. 119, 226601 (2017).
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Appl. Phys. Lett. 118, 252105 (2021).
 Journal of Applied Physics 132, 065704 (2022).
 Journal of Applied Physics 136, 055707 (2024).

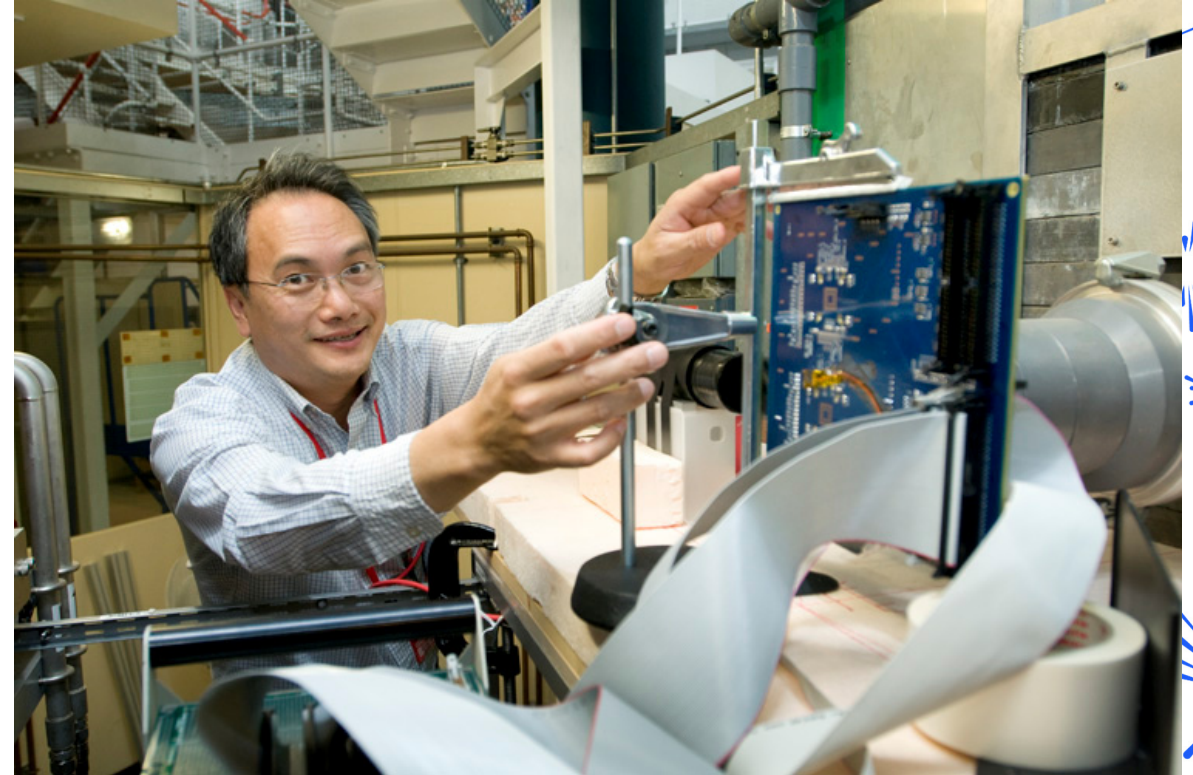


Protecting electronics from cosmic invaders

Single event upsets (SEUs) occur when high energy particles hit microelectronics, causing devices to malfunction. Solar flares and cosmic rays are the main sources of such particles and, with the ubiquitous nature and rapid advances in microelectronics; it's an area of intense interest. The semiconductor industry is looking ahead by understanding how and why devices might become vulnerable.

“Our experiments are about understanding the probability of these events, which represents an important contribution to the semiconductor industry. This will inform future design decisions, enabling manufacturers to develop devices that are less susceptible to muon SEUs.”

Professor Bhuva, Vanderbilt University



***STOP: 0x000000D1 (0x00000000, 0xF73120AE, 0xC0000008, 0xC0000000)

A problem has been detected and Windows has been shut down to prevent damage to your computer

DRIVER_IRQL_NOT_LESS_OR_EQUAL

If this is the first time you've seen this Stop error screen, restart your computer. If this screen appears again, follow these steps:

Check to make sure any new hardware or software is properly installed. If this is a new installation, ask your hardware or software manufacturer for any windows updates you might need.

If problems continue, disable or remove any newly installed hardware or software. Disable BIOS memory options such as caching or shadowing. If you need to use Safe Mode to remove or disable components, restart your computer, press F8 to select Advanced Startup Options, and then select Safe Mode.

*** ABCD.SYS - Address F73120AE base at C0000000, DateStamp 36B072A3

Kernel1 Debugger Using: COM2 (Port 0x2F8, Baud Rate 19200)

Beginning dump of physical memory

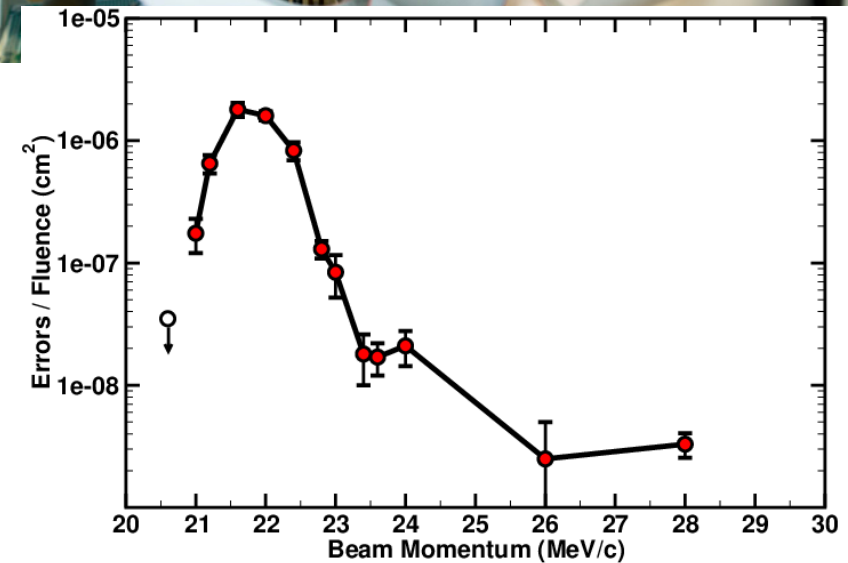
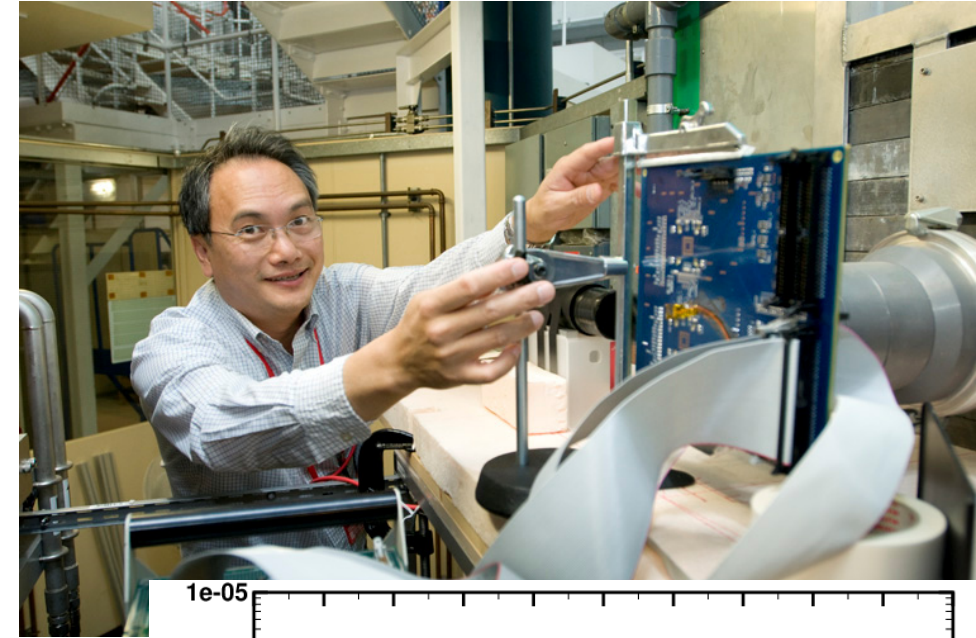
Physical memory dump complete. Contact your system administrator or technical support group.

Protecting electronics from cosmic invaders

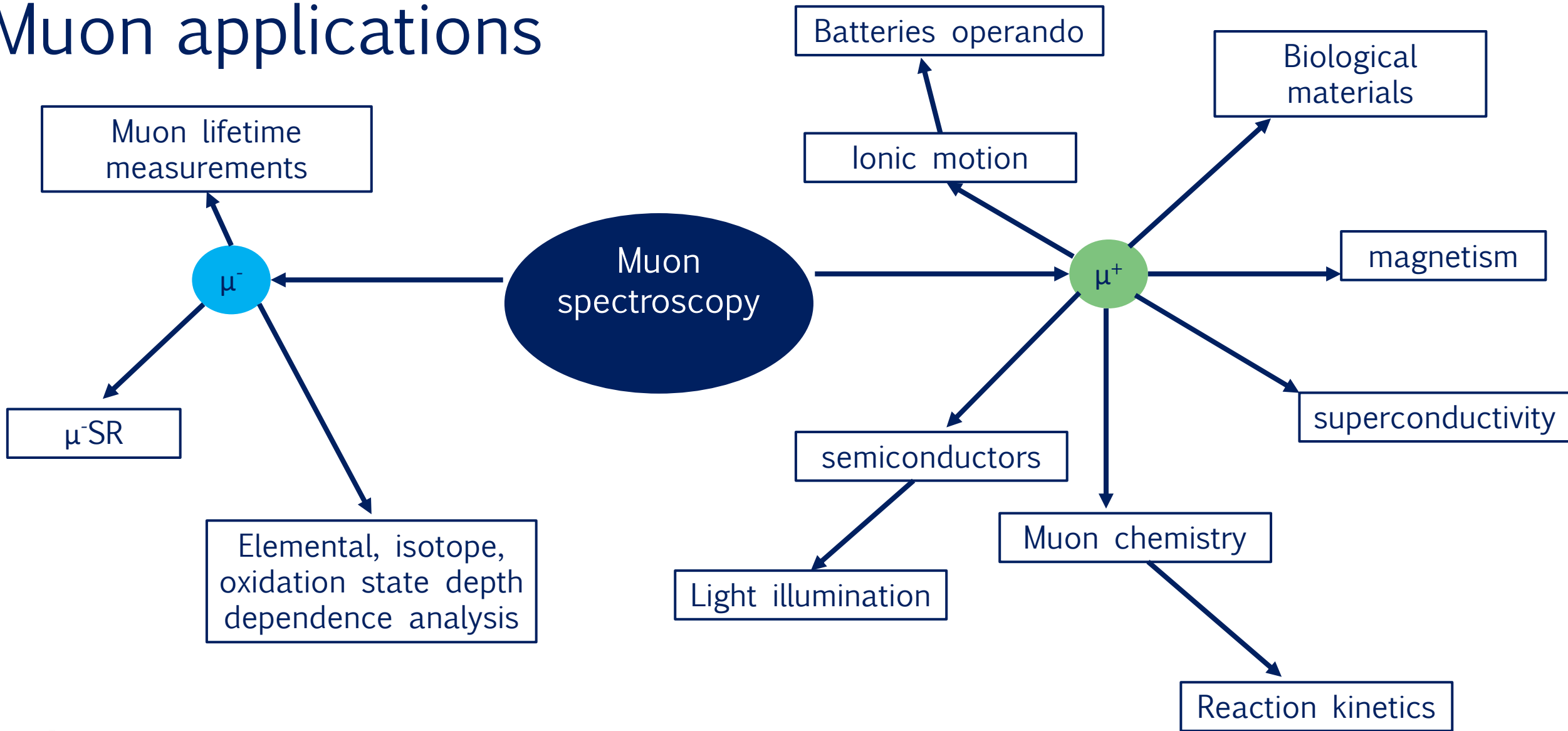
Single event upsets (SEUs) occur when high energy particles hit microelectronics, causing devices to malfunction. Solar flares and cosmic rays are the main sources of such particles and, with the ubiquitous nature and rapid advances in microelectronics; it's an area of intense interest. The semiconductor industry is looking ahead by understanding how and why devices might become vulnerable.

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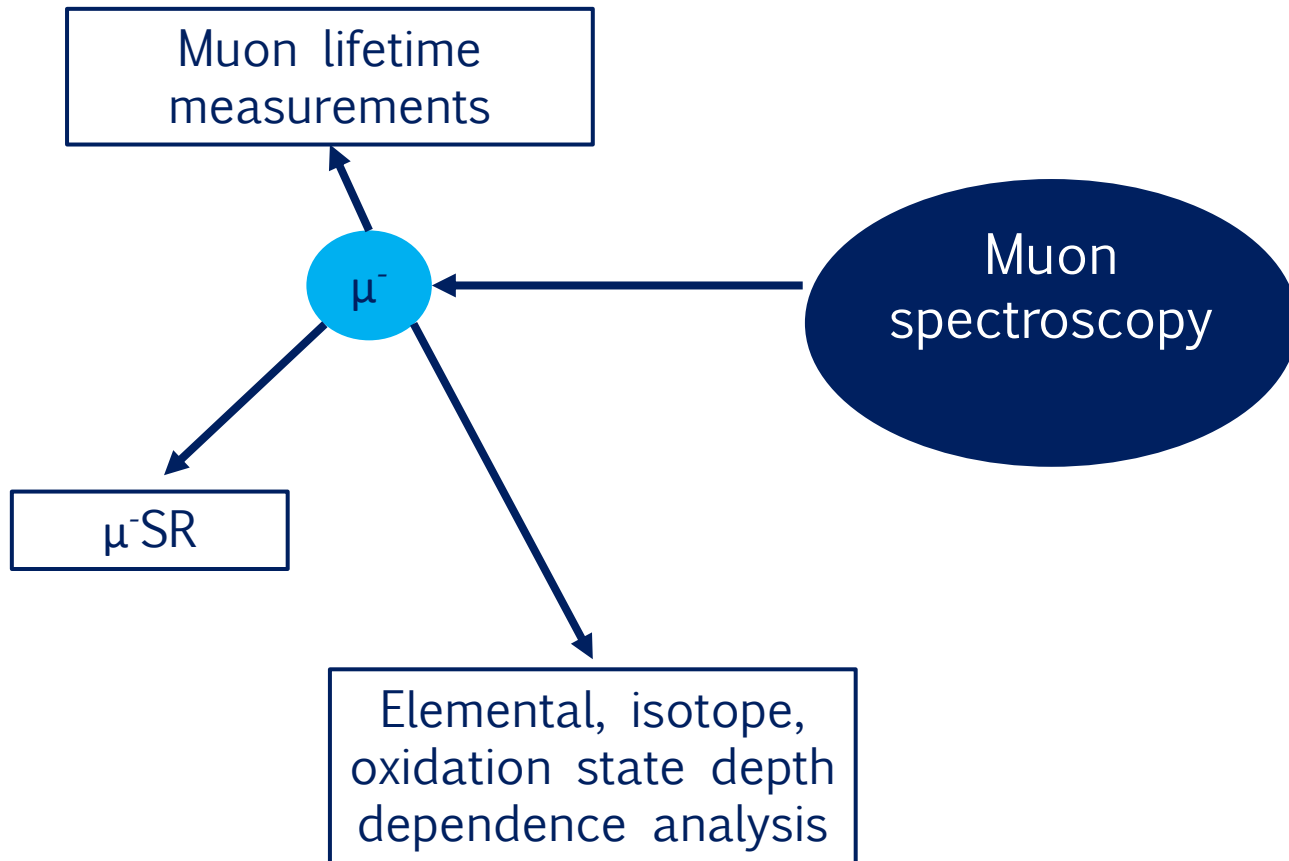
Professor Bhuvu, Vanderbilt University



Muon applications



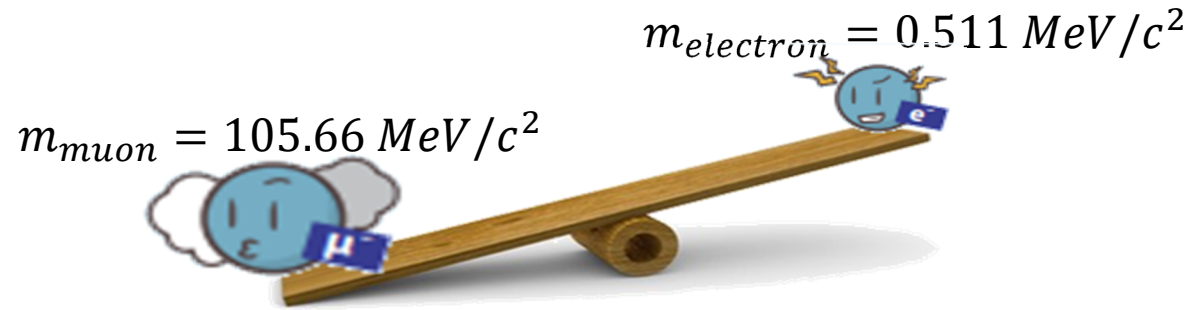
Muon applications



Muon properties

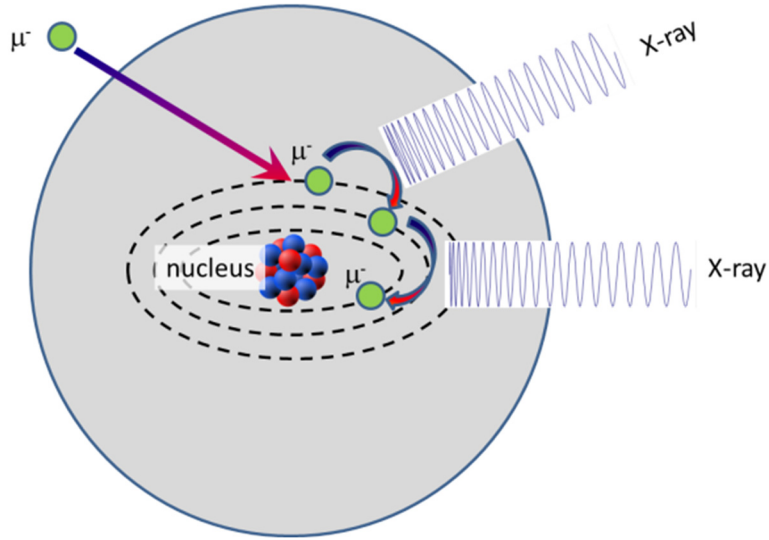
- fundamental, charged particles
- heavy electrons
- spin 1/2
- magnetic moment $3.2 \times m_p$
- mass $0.11 \times m_p$
- mass $200 \times m_e$
- produced from pion decays
- lifetime $2.2 \mu s$
- decay into a positron or electron (+ 2ν)
- energy of the muon is controllable

	Three generations of matter (fermions)			Interactions/force (bosons)	
QUARKS	2.2 MeV/c ² 2/3 1/2 u up	1.28 GeV/c ² 2/3 1/2 c charm	173.1 GeV/c ² 2/3 1/2 t top	0 0 1 g gluon	124.97 GeV/c ² 0 1 H higgs
	4.7 MeV/c ² -1/3 1/2 d down	96 MeV/c ² -1/3 1/2 s strange	4.18 GeV/c ² -1/3 1/2 b bottom	0 0 1 γ photon	SCALAR BOSONS
	0.511 MeV/c ² -1 1/2 e electron	105.66 MeV/c ² -1 1/2 μ muon	1.7768 GeV/c ² -1 1/2 τ tau	91.19 GeV/c ² 0 1 Z Z boson	
LEPTONS	<1.0 eV/c ² 0 1/2 ν _e electron neutrino	<0.17 MeV/c ² 0 1/2 ν _μ muon neutrino	<18.2 MeV/c ² 0 1/2 ν _τ tau neutrino	80.39 GeV/c ² ±1 1 W W boson	

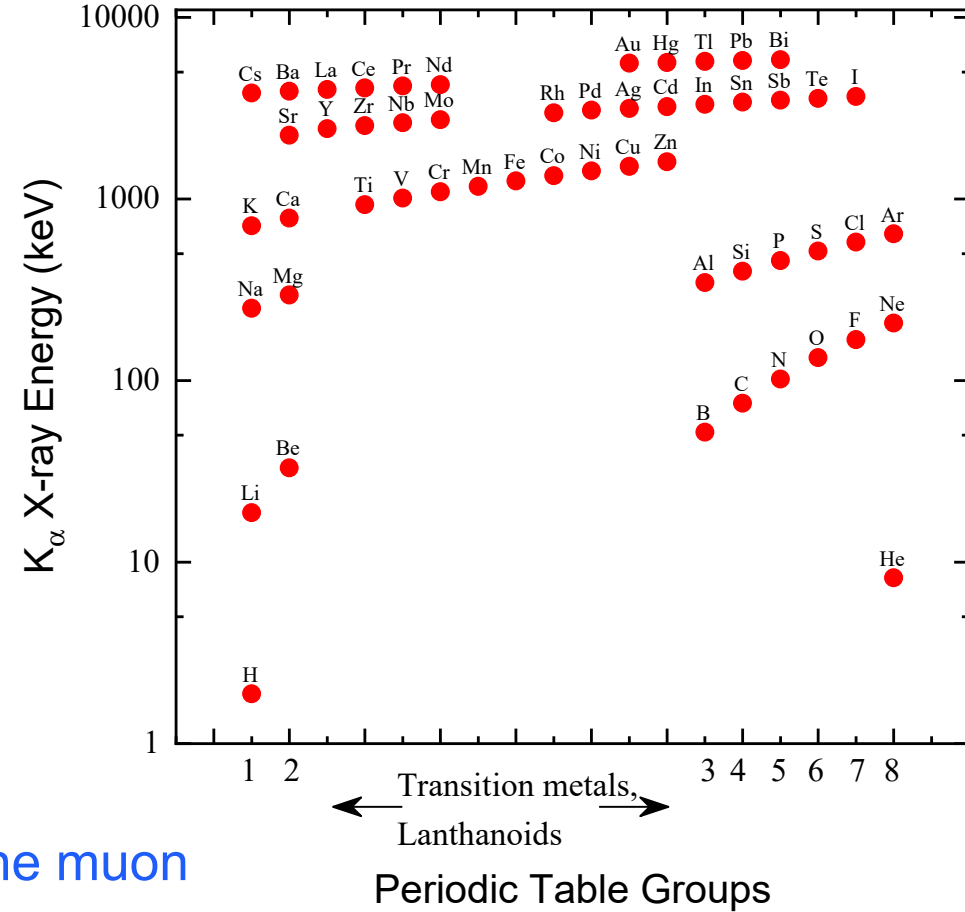


$$\frac{m_{muon}}{m_{electron}} = 206.77 \sim 207$$

Negative Muon Capture



- High Energy X-rays emitted
 - 0.01- 6 MeV
- Energy dependent of the atom which captures the muon
- Isotope and oxidation states can also be determined



Negative muons – usage at ISIS

Diverse science areas

- Cultural Heritage
 - Bronze Age artefacts
 - Mary Rose artefacts
 - Baptismal Gates
 - Swords
 - Cannon Balls
 - Sundials
 - Coins
 - Faience and glass
- Solar cells
- Piezoelectric devices
- Li batteries
- Advanced engineering

Hillier et al in Handbook of Cultural Heritage analysis
Cataldo et al, Appl. Sci 2022



Understanding Roman Gold Coinage Inside Out

George Alexander Green^{a,1}, Katsu Ishida^b, Bethany V. Hampshire^c, Kevin Butcher^a, A. Mark Pollard^d, Adrian D. Hillier^e

Archaeological and Anthropological Science
<https://doi.org/10.1007/s12520-022-01631-3>

ORIGINAL PAPER

Negative muons reveal the economic chaos of Rome's AD 68/9 Civil Wars

G. A. Green^{1,2} · K. Ishida³ · K. Domoney¹ · T. Agoro^{4,5} · A. D. Hillier²



Microchemical Journal
Volume 125, March 2016, Pages 20

Probing beneath the surface without a destructive elemental analysis using negative muons



Probing the past with negative muons

Using negative muons as a tool for elemental analysis is now possible at ISIS. Muon scientists, together with the Universities of Oxford and Warwick, are currently developing the technique on the RIKEN Port 4 to non-destructively probe the composition of archaeological objects – including gold and silver coins from the Roman period and the Mary Rose – paving the way for future studies on other archaeological artefacts, engineering materials, bio-systems, and battery materials.



Figure 2. Gold Roman coin dated AD 74-75, analysed on RIKEN Port 4. [View full-size image](#)

Elemental analysis is a process in which the composition of a sample is determined. These techniques are often destructive and unsuitable for the analysis of valuable samples such as those of archaeological artefacts, engineering materials, bio-systems, and battery materials. Other non-destructive measurement techniques such as those based on muons can offer non-destructive measurement of characteristic elements.

IOP Publishing
Journal of Physics: Condensed Matter
J. Phys.: Condens. Matter 30 (2018) 125703 (5pp)
<https://doi.org/10.1088/1361-648X/aaade3>

Depth dependant element analysis of $PbMg_{1/3}Nb_{2/3}O_3$ using muonic x-rays

K L Brown¹, C P J Stockdale¹, H Luo², X Zhao³, J-F Li⁴, D Viehland⁴, G Xu⁵, P M Gehring³, K Ishida⁶, A D Hillier⁷ and C Stock^{1,8}

applied sciences

Review

A Novel Non-Destructive Technique for Cultural Heritage: Depth Profiling and Elemental Analysis Underneath the Surface with Negative Muons

Matteo Cataldo^{1,2,3,*}, Massimiliano Clemenza^{1,2}, Katsuko Ishida⁴ and Adrian D. Hillier³

Home > Handbook of Cultural Heritage Analysis > Reference work entry

Depth-Dependent Bulk Elemental Analysis Using Negative Muons

Adrian D. Hillier Beth Hampshire & Katsu Ishida

JAAS

PAPER [View Article Online](#) [View Journal](#) [View Issue](#)

[Check for updates](#)

A new multidisciplinary non-destructive protocol for the analysis of stony meteorites: gamma spectroscopy, neutron and muon techniques supported by Raman microscopy and SEM-EDS

Cite this: *J. Anal. At. Spectrom.* 2023, 38, 293

nanomaterials

Using the Emission of Muonic X-rays as a Spectroscopic Tool for the Investigation of the Local Chemistry of Elements

Matteo Aramini¹, Chiara Milanese^{2,*}, Adrian D. Hillier¹, Alessandro Girella², Christian Horstmann³, Thomas Klassen^{3,4}, Katsuo Ishida^{1,5}, Martin Dornheim³ and Claudio Pistidda^{3,6}



Roman silver coins:



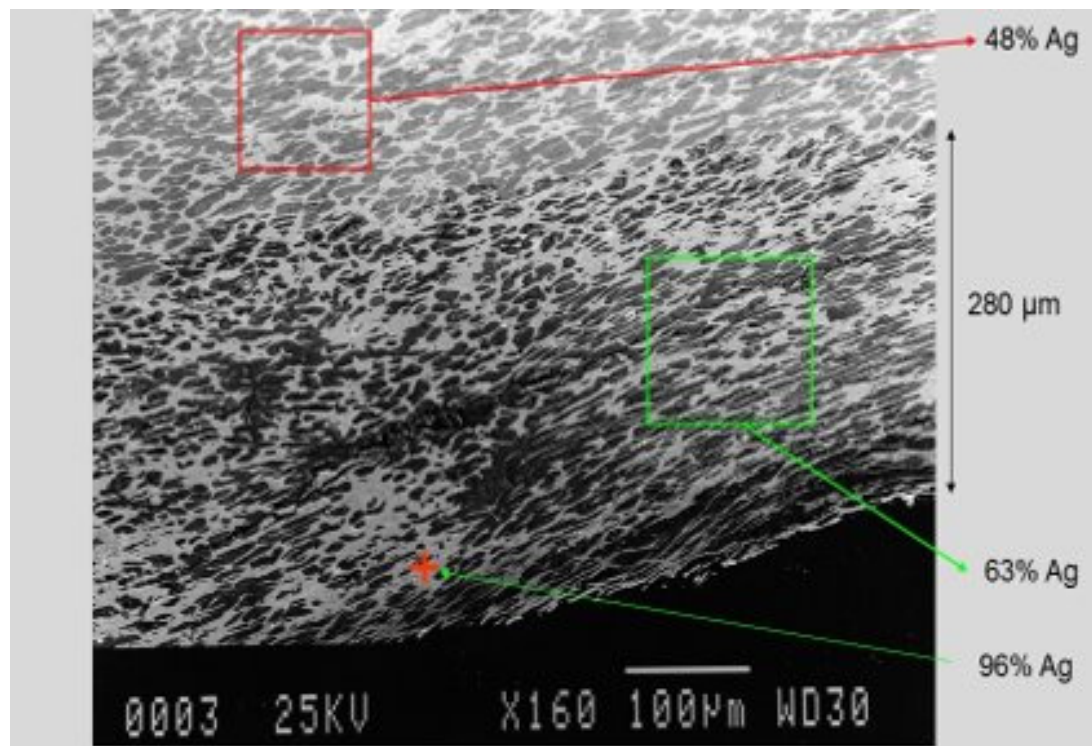
denarius of Julia Domna

- 3rd century (211-217 CE)

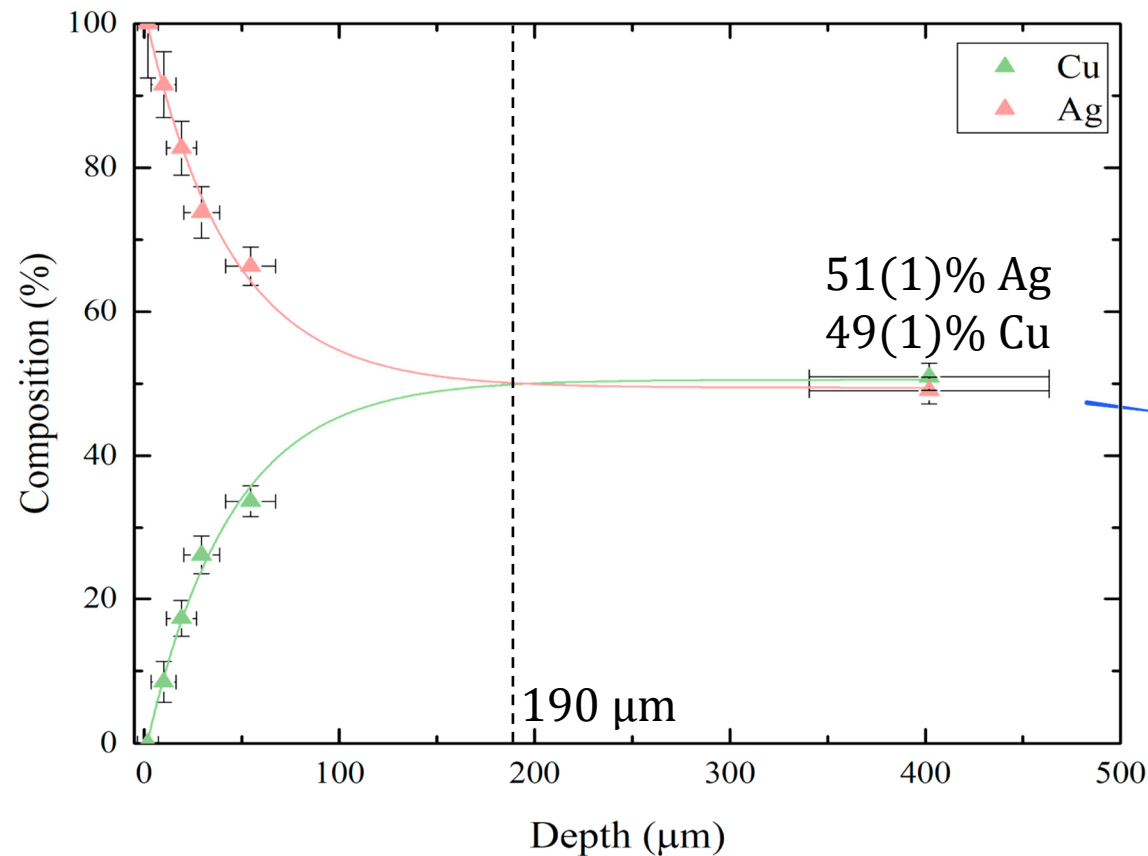
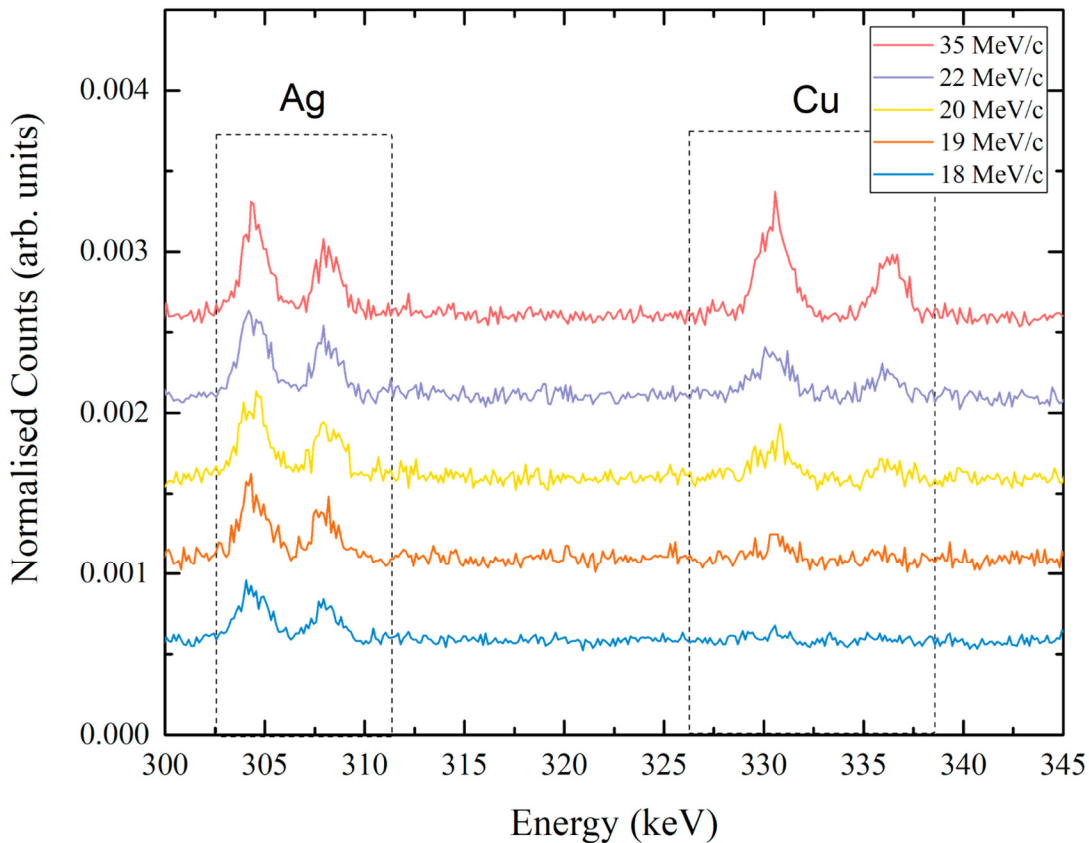
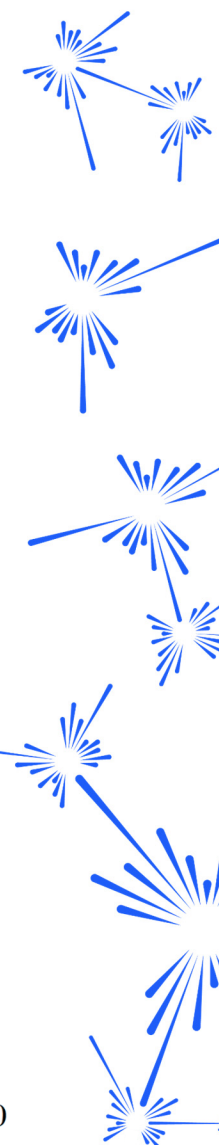


The fineness and quality of a coinage is often taken by historians to be a comment on the fiscal health of the issuing state

Yet very little is really known about its fineness and chemical composition



Roman silver coins:



Roman gold coins



During the AD 68/9 Civil Wars: Galba, Otho, Vitellius and then Vespasian fought for — and gained — control

A period of serious and sustained disruption.

Existing analyses shows a minor reduction in the purity of the gold coinage

Using X-ray fluorescence and u-XES, a number of heavily debased gold coins issued during the AD 68/9 and many slightly debased coins issued in their immediate aftermath.

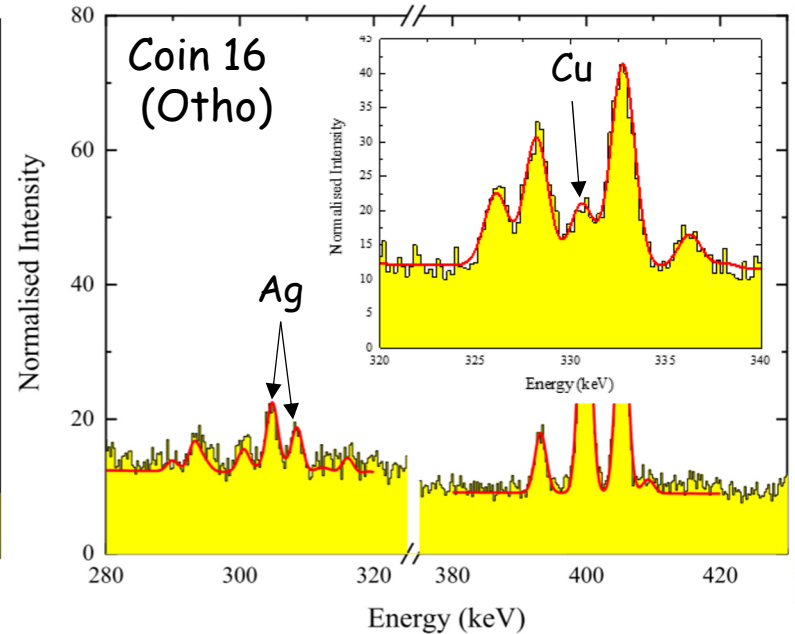
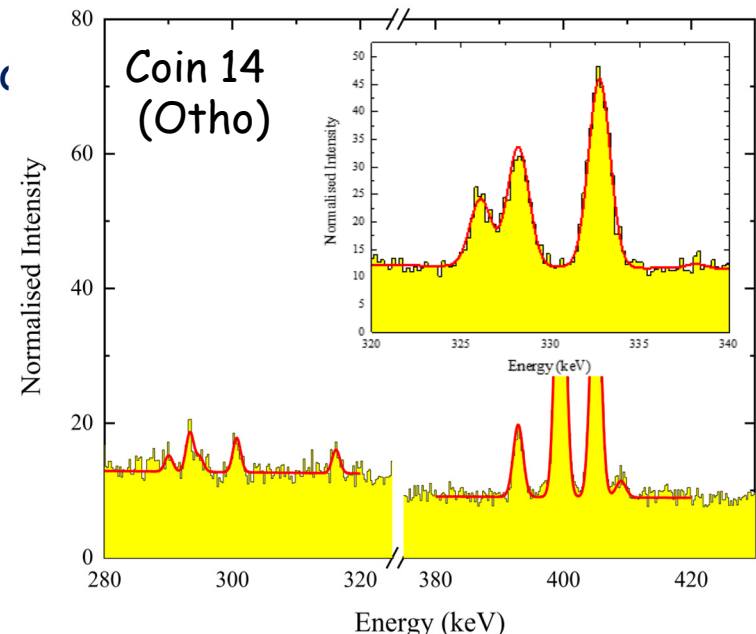
aurei from the "year of four emperors"

- AD 68/9

G. A. Green et al, *Archaeological and Anthropological Sciences* 14, 165 (2022)



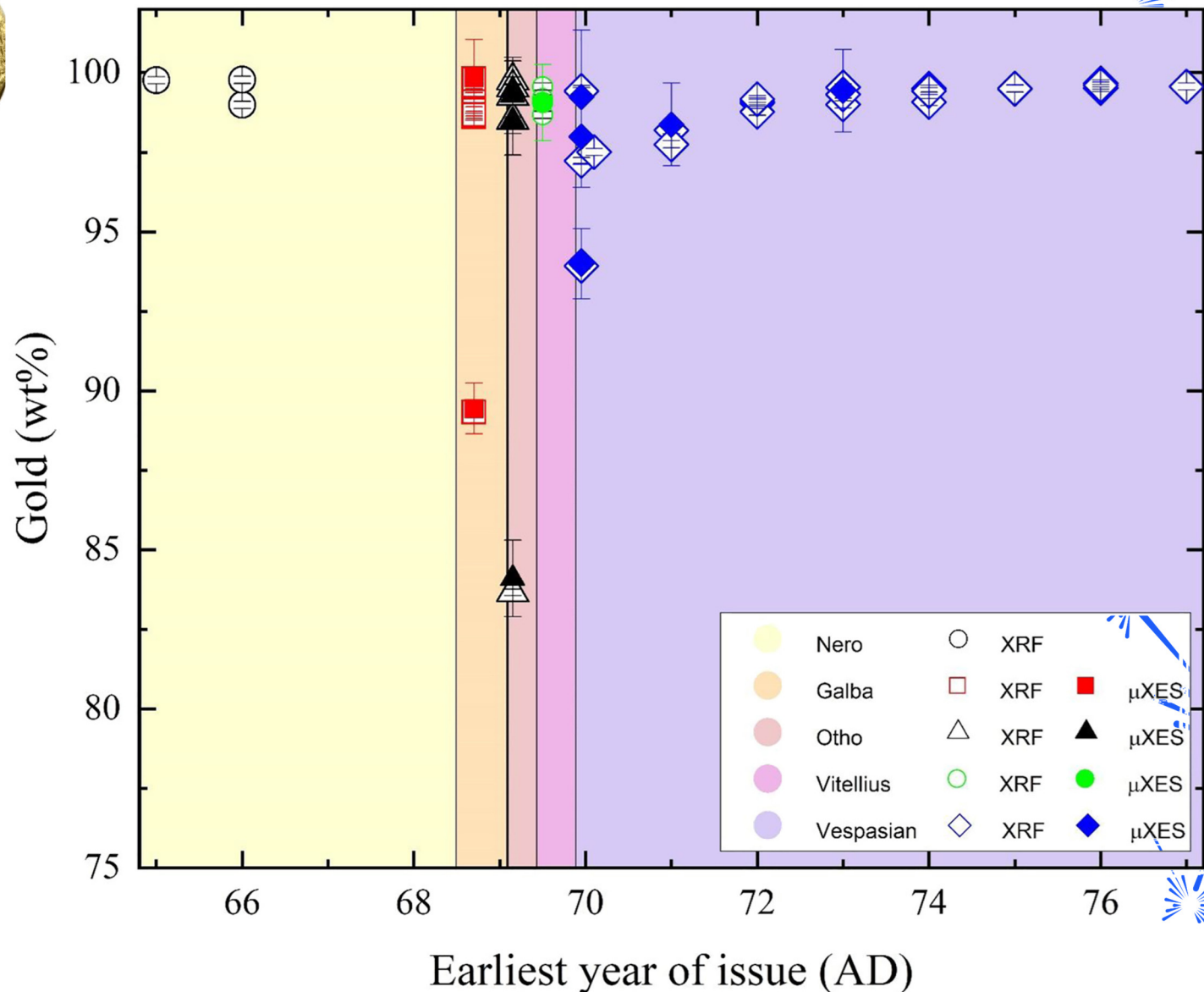
ISIS Neutron and Muon Source



Roman gold coins:



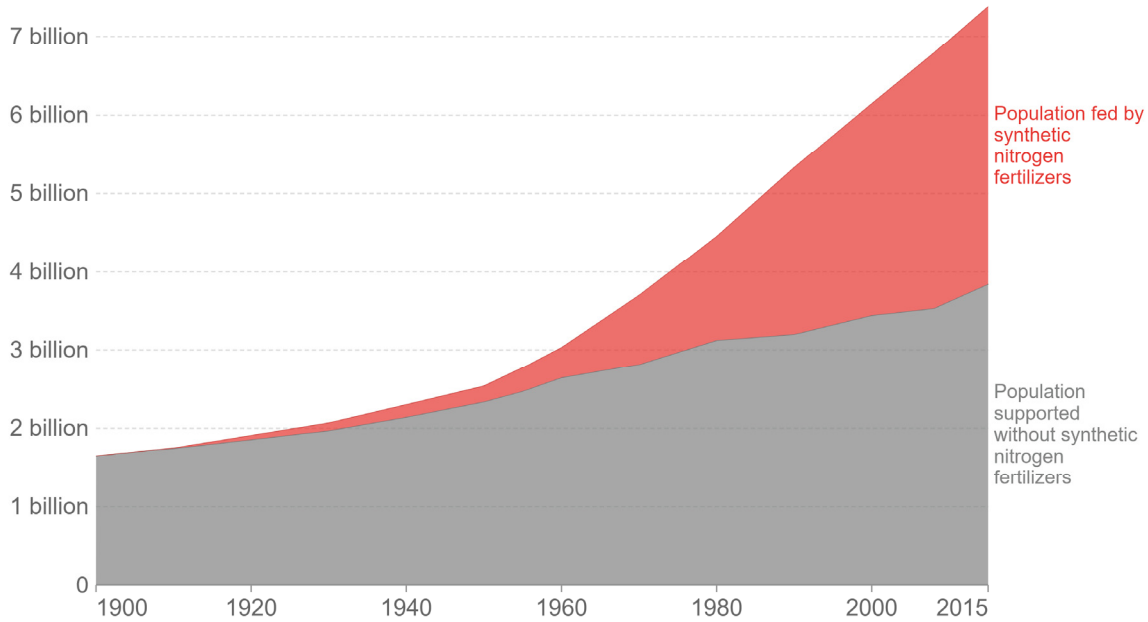
- Using X-ray fluorescence and μ -XES, a number of heavily debased gold coins issued during the AD 68/9 and many slightly debased coins issued in their immediate aftermath.
- Debased Civil War gold coinages were indeed produced; that copper was used to debase Roman gold coins during this time, c. 185 years earlier than first shown
- The metallurgical evidence from the gold coinage show the AD 68/9 Civil Wars caused significant and sustained disruption to the Roman economic system.



Green fertiliser: Nitrogen fertilisers

World population supported by synthetic nitrogen fertilizers

Estimates of the share of the global population which could be supported with and without the production of synthetic nitrogen fertilizers (via the Haber-Bosch process) for food production. Best estimates project that just over half of the global population could be sustained without reactive nitrogen fertilizer derived from the Haber-Bosch process.

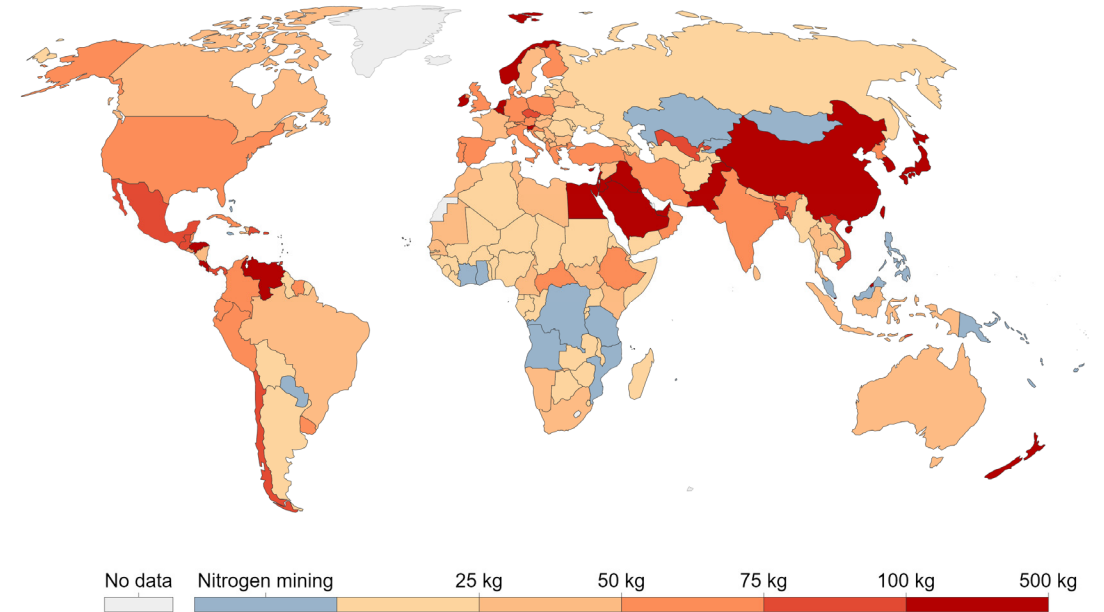


Source: Erisman et al. (2008); Smil (2002); Stewart (2005)

OurWorldInData.org/how-many-people-does-synthetic-fertilizer-feed/ • CC BY

Excess nitrogen per hectare of cropland

Amount of excess nitrogen per hectare of cropland. This is the difference between nitrogen inputs, and amount harvested in crops.



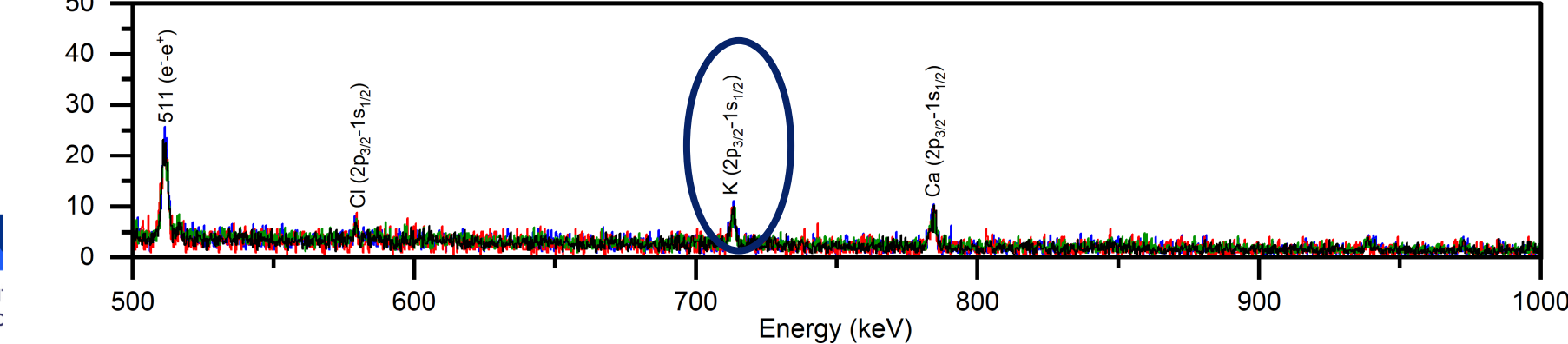
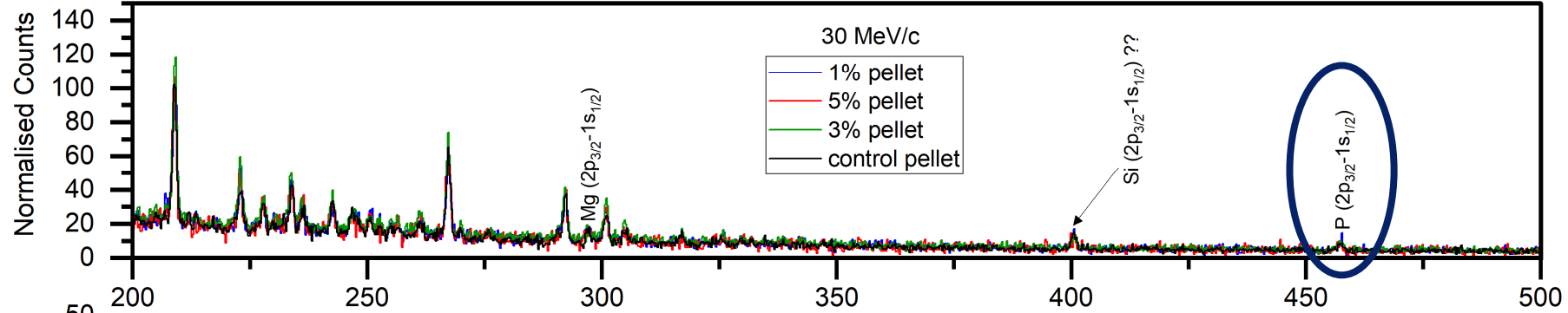
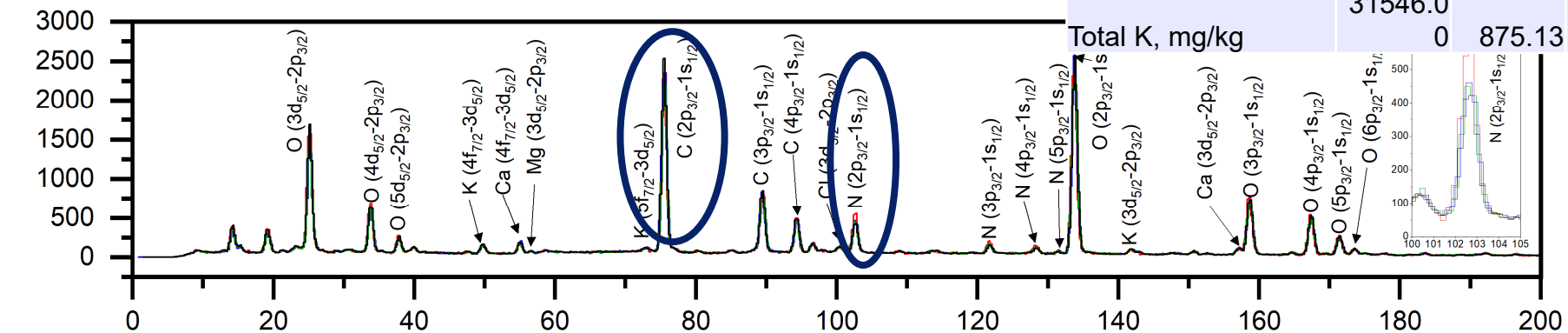
Source: West, Gerber, Engstrom, Mueller, Brauman, Carlson, Cassidy, Johnston, MacDonald, Ray & Siebert (2014). Leverage points for improving global food security and the environment. <i>Science</i>. OurWorldInData.org/fertilizers • CC BY

- typically, only 45-55% of the nitrogen applied is taken up by crops¹

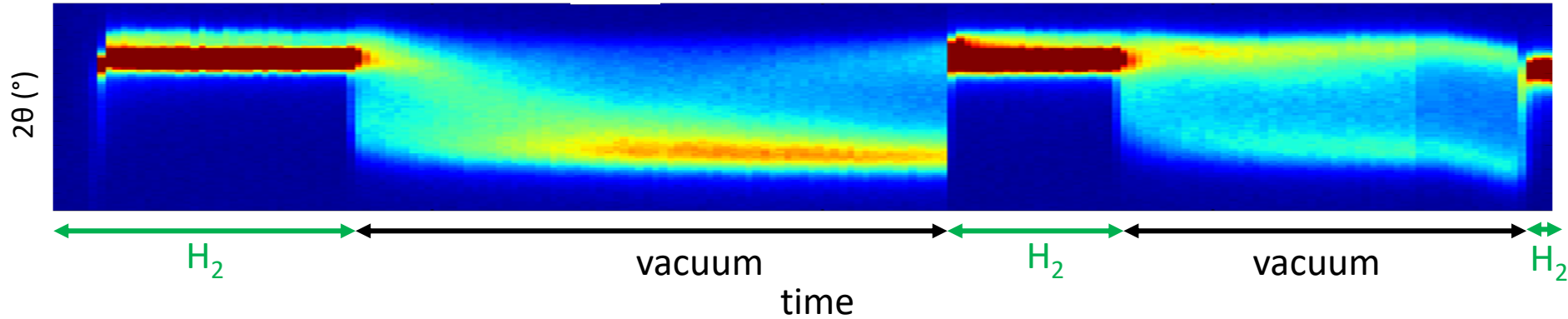
¹Oluwadunsin et al (2022). J. Env Management 317 : 115395



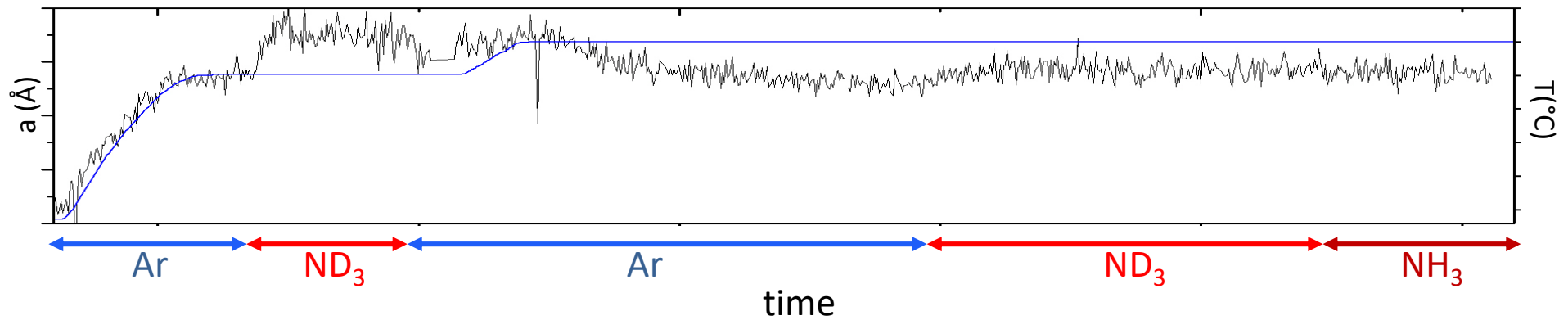
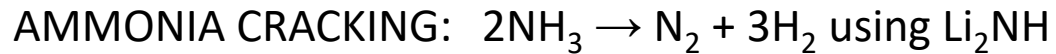
	Control Pellet		1% BS Pellet		5% BS Pellet	
	Mean	Std Dev	Mean	Std Dev	Mean	Std Dev
Total N, % w/w	4.70	0.16	4.84	0.02	5.98	0.08
Total C, % w/w	38.00	1.46	37.35	0.30	36.75	0.39
Total P, mg/kg	13023.7		12929.7		12606.5	
	5	237.27	5	202.86	0	118.70
Total K, mg/kg	31546.0		30863.5		29622.0	
	0	875.13	0	217.77	0	220.09



Energy Materials: Lithium amide imide (LiNH₂ – Li₂NH)



Synchrotron X-ray powder diffraction shows hydrogen release occurs via range of intermediate stoichiometry values – key to easily reversible hydrogen storage



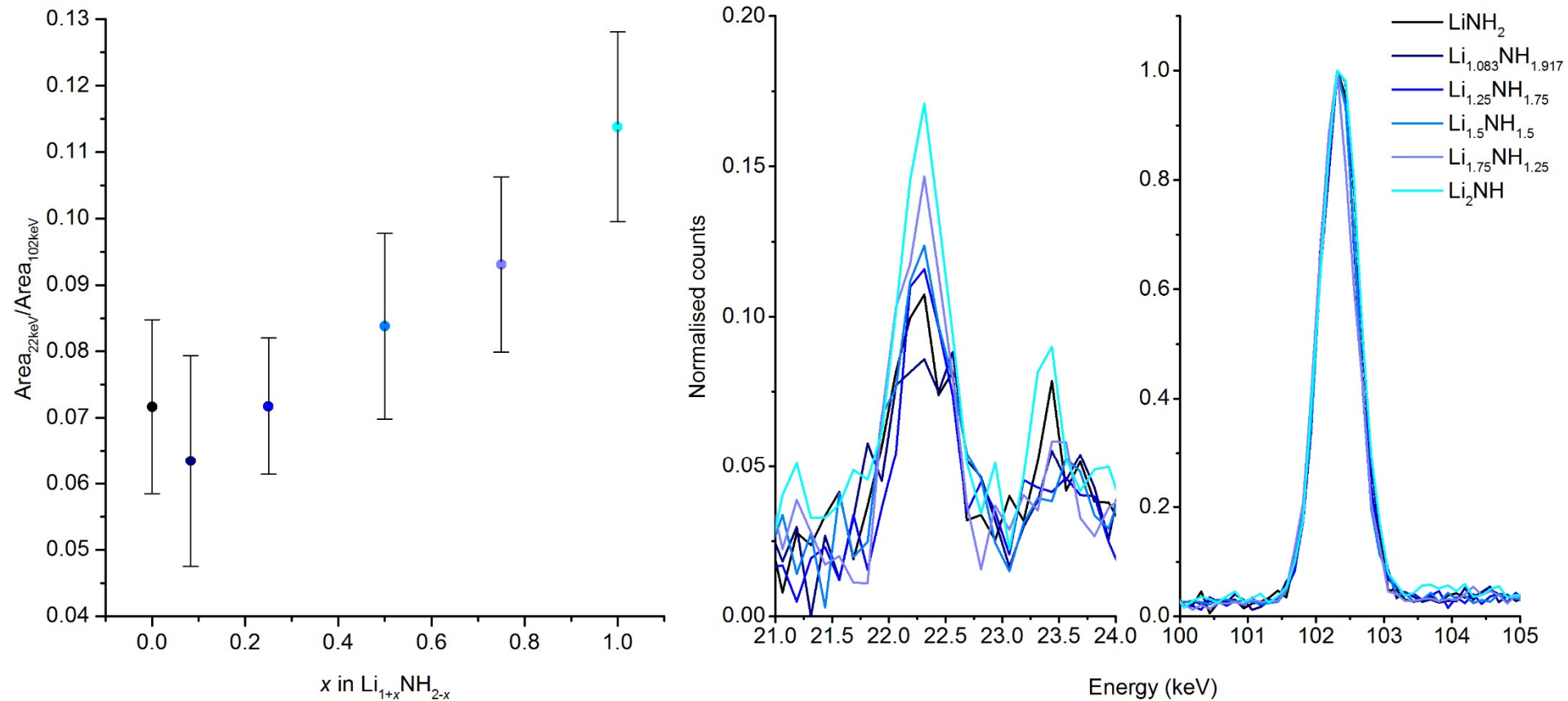
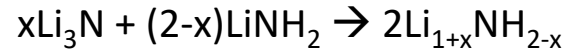
Variation in lattice parameter of Li_2ND on exposure to ND_3 indicates non-stoichiometry during ammonia decomposition reaction – active form of the catalyst is non-stoichiometric.

Energy Materials: Lithium amide imide (LiNH₂ – Li₂NH)

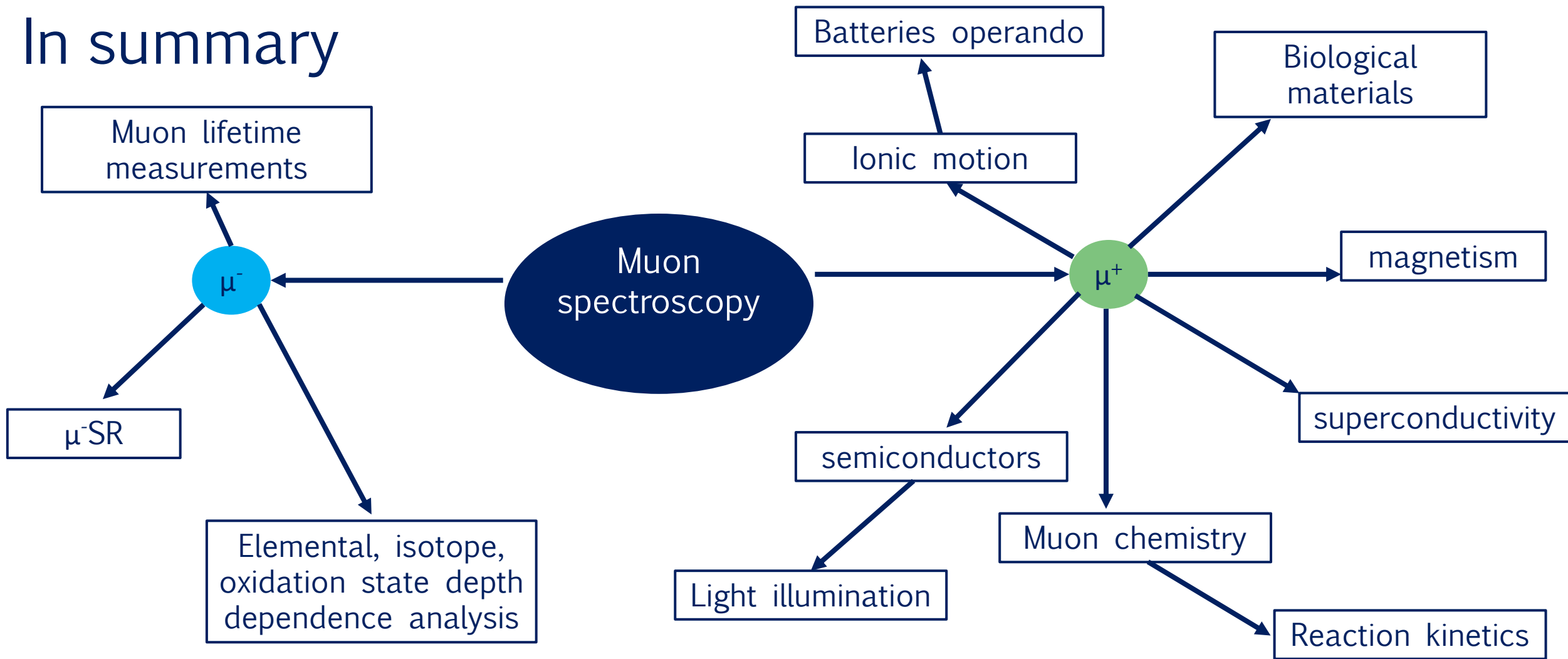
Quantifying the stoichiometry is difficult. By diffraction the scattering (both X-ray and neutron) is dominated by the nitrogen.

QUANTIFYING STOICHIOMETRY WITH NEGATIVE MUONS

Synthesised a series of lithium amide-imide samples with varying stoichiometry:



In summary



Muons are a powerful probe to understand our material world



Bye! It's time for me to decay.