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Introduction to μSR: What, How, Where?

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INTRODUCTION TO μSR: WHAT, HOW, WHERE?

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μSR spectroscopy uses implanted muons to probe the structure and dynamics of matter at the microscopic level. The acronym stands for ***muon spin rotation, relaxation*** and ***resonance***, covering the various ways in which the muon spin polarization can evolve following implantation, in response to the local magnetic environment. The definition could now be extended to include muon *level crossing resonance* (μLCR), which is a double resonance technique informative on local electronic structure. Applications span a wide range of studies in physics, chemistry and materials science. Common features are the sensitivity of the muons' response to magnetic and hyperfine fields and the manner in which the positive muon behaves – in its chemical and elastic interactions with matter – as a lightweight proton. The experiments must be performed at accelerator laboratories where suitable muon beams are available. The beam is stopped in the material of interest, which may be gas, liquid or solid. Muons are short–lived particles, decaying with a lifetime of about $2\,\mu$s; they provide the classic illustration of parity violation in radioactive decay, and it is this phenomenon which is the basis for the μSR techniques.

This Lecture introduces the field by answering some obvious questions (or by indicating, at least, in which subject areas the answers are to be found). Most of the topics are treated in greater depth in subsequent Lectures.

1. WHAT ARE MUONS?

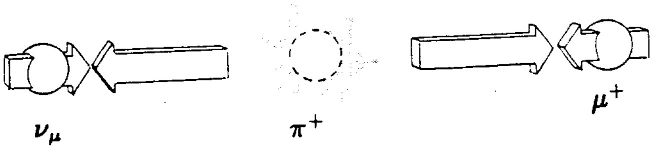
1.1 Particle physicists' view:

Muons are *leptons*. Like electrons and taus, they are elementary particles which do not feel the Strong Interaction, only the Electro–Weak. Both charge states exist (particle and antiparticle). So *negative* muons are like heavy electrons, *positive* muons like heavy positrons. In the *Standard Model*, electrons, muons and taus give their names to the 3 families of particles:

	Leptons:		Quarks:
Electron family	ν_e, $\bar{\nu}_e$	e^+, e^-	up, down
Muon family:	ν_μ, $\bar{\nu}_\mu$	μ^+, μ^-	strange, charm
Tau family:	ν_τ, $\bar{\nu}_\tau$	τ^+, τ^-	top, bottom

Muons are produced when a target of graphite or other light element is exposed to a sufficiently high–energy proton beam. Various proton–nucleon interactions, *e.g.* (1), generate pions in the first instance; these particles then decay quickly to muons (2).

$$
\begin{aligned}
p + p &\rightarrow p + n + \pi^+ && && (1)\\
\pi^+ &\rightarrow \mu^+ + \nu_\mu && (\tau_\pi = 26ns). && (2)
\end{aligned}
$$



Pion decay to spin-polarized muons (2)

The muon properties [1] relevant to μSR are:

Charge	$Q_\mu = Q_p = 1$ (a.u.)
Spin	$I_\mu = I_p = \frac{1}{2}$
Magnetic moment	$\mu_\mu \simeq 3.18\mu_p$
Gyromagnetic ratio	$\gamma_\mu \simeq 3.18\gamma_p \simeq 2\pi \times 136\,\mathrm{kHz/mT}$
g-factor	$g - 2 \simeq 10^{-4}$
Mass	$m_\mu \simeq 0.113\,m_p \simeq 207\,m_e$
Lifetime	$\tau_\mu = 2.197\,\mu\mathrm{s}$
Kinetic energy of "surface" muons	$\simeq 4\,\mathrm{MeV}$
Range in matter	$\simeq 100\,\mathrm{mg/cm^2}$.

μSR spectroscopy can be said to derive directly from the early experiments which determined these properties[1] and to benefit greatly, in terms of sensitivity, from the "nuclear technique" of counting individual particles. Success relies on two circumstances. The first is the high degree of spin polarization which can be achieved. This may be up to 100% for "surface" muons.[2] This high initial polarization is preserved during beam transport[3] and usually also when the muons are stopped or thermalized in the sample of interest (the spin state being insensitive to electrostatic interactions). The second is the manner in which the subsequent evolution of polarization within the sample may then be displayed. The muons are themselves unstable particles, and decay radioactively with a lifetime of just over 2 μs: negative muons decay to electrons, positive muons to positrons, *e.g:*

$$
\mu^+ \rightarrow e^+ + \bar{\nu}_e + \nu_\mu \qquad (\tau_\mu \simeq 2.2\,\mu s). \qquad (3)
$$

The β-emission is asymmetric with respect to the muon spin, allowing the muon polarization to be analyzed. Roughly stated, the electron or positron tends to be emitted preferentially along

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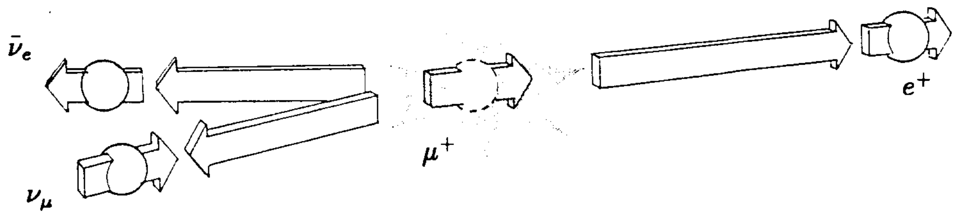
[1] In many cases current Muon Physics research is concerned with improving the precision of these measurements.

[2] *i.e.* for muons collected from the decay of pions at rest on the surface of the production target. This is a consequence of the neutrinos having "left handed" helicity; conservation of angular momentum in equation (2) then implies that the muon spin is antiparallel to its momentum vector (the beam direction) as illustrated in the sketch. Whether any *rare decays* giving right-handed neutrinos occur need not concern us, but is a current issue in Muon Physics.

[3] This is a consequence of $g - 2 \simeq 0$: the spin remains locked to the momentum vector through the bending and focussing magnets.



Positive muon decay (the positrons are detected in μSR experiments).

the instantaneous spin direction of the parent muon (more details are given in Section 3). This is the classic example of *parity violation*[4] in radioactive decay, published by Garwin, Lederman and Weinrich in 1957 [2]. These authors were well aware of the potential, and concluded their article with the perceptive statement: "*.....it seems possible that polarized positive and negative muons will become a powerful tool for exploring magnetic field in nuclei, atoms and interatomic regions*".

1.2 Atomic physicist' view:

Negative muons can substitute for ordinary electrons in *exotic* atoms and molecules. The process of capture in an outer orbital and *cascade* down to an inner generates characterisic X–ray spectra. Muons are some 200 times heavier than ordinary electrons, so the Bohr radii of their orbitals are correspondingly smaller. In the hydrogen and helium isotopes, the effective screening of the nuclear charge is exploited in *muon catalyzed fusion.* In heavier nuclei, premature capture by a nucleus can effectively reduce the negative muon's lifetime but, within this limitation, *muon spin rotation* experiments (μ^-SR) are possible. In their orbital ground states, the negative muons in effect transmute an element from atomic number Z to $Z-1$, and their precession frequencies probe the hyperfine fields close to the nuclei [4].

Muonic hydrogen *Muonium*

The bound state of a *positive* muon and an ordinary electron, Mu= μ^+e^-, closely resembles a hydrogen atom. Known as *muonium*, its importance in atomic physics is that measurement of its spectroscopic constants provides a valuable testing ground for bound–state *quantum electrodynamics* (QED). There is not the uncertainty in nuclear (proton) size which limits the comparison of experiment and theory for protium, muons being point–like in their interactions. Muonium resembles protium much more closely than does the other all–leptonic atom, *positronium*, since the mass ratio between muon and electron makes the positive muon essentially a central nucleus.

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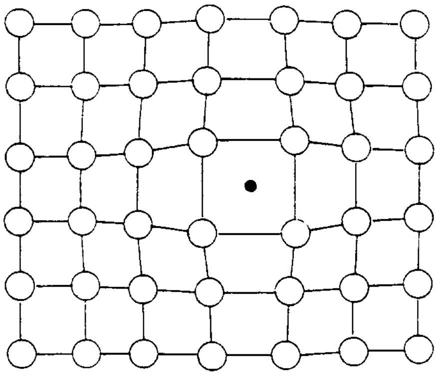
[4] The fact that neutrinos are "left–handed" and not "right–handed" is also an (extreme) example of parity violation; μSR can therefore be said to rely on parity violation both in the pion and muon decays, the former to produce the muon polarization, the latter to analyse it.

The remainder of this lecture (and most of the School) has to do with positive muons and muonium, not as the free particles or vacuum–state atoms, but in their interaction with matter.

1.3 Solid state physicists' view:

Implanted in matter, muons provide a microscopic probe with unique properties. They localise at specific sites in lattices or molecules and their μSR spectra characterize the local magnetic and hyperfine fields. The large magnetic dipole moment (about 3 times that of the proton) and the absence of an electric quadrupole moment (the muon has spin-$\frac{1}{2}$) make the muon a sensitive and accurate magnetometer. The nature of the muons' response and the way in which this is displayed is summarised in Sections 2 and 3 of this Lecture; subsequent Lectures give details and introduce applications in specific subject areas. Internal fields can be measured in magnetic materials and their fluctuations studied in the vicinity of magnetic phase transitions [5]. Spin density distribution can be mapped out in open–shell molecules [3, 6]. Flux penetration can be studied in superconductors [7].



Interstitial muon site in a lattice [3].

In a lattice, muons invariably adopt interstitial sites and create a particularly fundamental form of point defect, in which the immediate environment must respond to the introduction of a unit "test charge". In metals especially, charge and spin density are significantly disturbed [8, 5]. Modelling this response is a surprisingly severe and valuable test of the understanding of the properties of any material. The small mass, about one ninth that of the proton, make the muon the the prototype light interstitial for studies of dynamics and diffusion [10, 11].

1.4 Chemists' view:

In their chemical and elastic interactions with matter, positive muons may be considered to behave simply as lightweight protons and muonium as a pseudo–isotope of hydrogen [3, 6].[5] In support of the isotope analogy, the reduced mass of the electron in muonium differs from that in protium by only 0.5%, so that the size and binding energy of the two atoms are essentially identical [12]:

Bohr radius	$a_{Mu} = 1.004\, a_H$
Ionization potential	$I_{Mu} = 0.996\, I_H$

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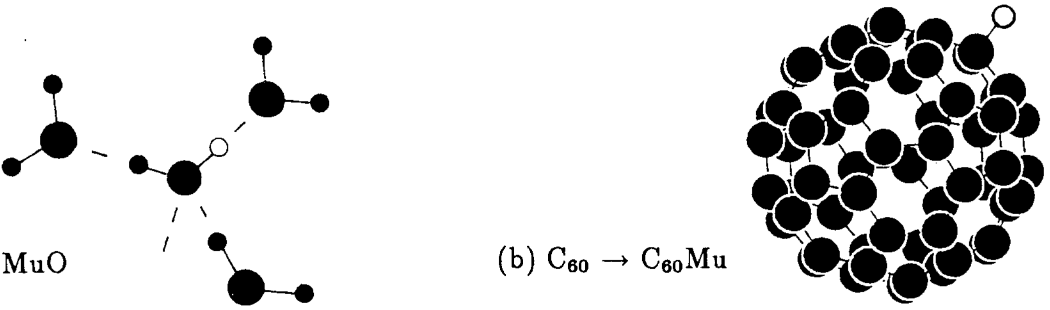
[5] The analogy would horrify the high energy particle physicists, but the point is that materials science and chemistry are concerned with relatively low energy phenomena – usually $< 100\,\mathrm{eV}$ – where the fact that the muon does not experience the Strong Interaction is unimportant.



(a) $H_2O \rightarrow HMuO$

(b) $C_{60} \rightarrow C_{60}Mu$

Muonium substituted in a closed shell molecule (a)
and incorporated in an open–shell molecule (b) [3].

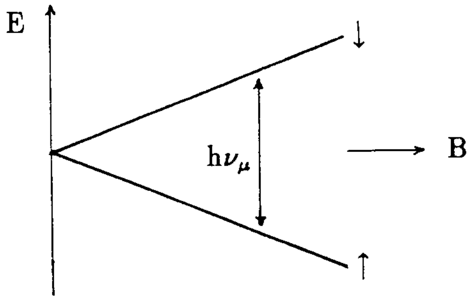
Their chemical behaviour is, in most reactions, qualitatively similar. The unprecedented isotopic mass ratio, $M_H/M_{Mu} = 9$, does however result in significant quantitative differences, and these are one of the major topics of interest in ***muonium chemistry***. Differences in reaction rate are referred to as ***kinetic isotope effects*** [6]; differences in the observable properties of molecules where hydrogen is replaced by muonium are referred to as ***dynamic isotope effects*** [3]. Both are much enhanced, compared with the corresponding isotope effects between protium and deuterium, and therefore more reliably interpreted.

2. WHAT IS MEASURED?

It is the response of the muon spins to their magnetic environment in the sample which is the source of information in all μSR experiments. This section introduces and summarizes the essential concepts. Many of these are also to be found in textbooks on ***magnetic resonance*** [13, 14] which should be consulted for a more rigorous treatment.

2.1 Zeeman energy

The muon has spin one–half. That is, it adopts one of two possible spin orientations with respect to any reference direction or ***quantization axis***. These are loosely referred to as the ***spin–up*** and ***spin–down*** states, a terminology which takes on some meaning when a magnetic field is applied and the quantization axis chosen to be along the field direction. Then the spin–up state (spin parallel to the field) has a lower magnetic energy than the spin–down state (antiparallel), the muon's magnetic moment being locked to its spin. The energy difference or splitting is known as the Zeeman energy and increases linearly with field.



Spin–$\frac{1}{2}$ energy levels.

2.2 Larmor frequency

Usually the Zeeman energy is measured in units of frequency, according to

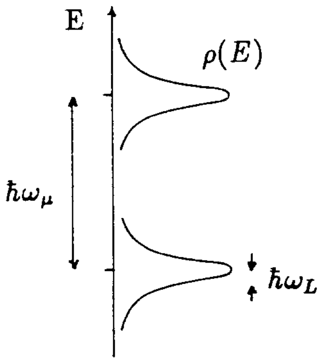
$$\delta E = h\nu_\mu = \hbar\omega_\mu = \hbar\gamma_\mu B. \tag{4}$$

The frequency ν_μ is called the Larmor frequency and is commonly expressed as an angular frequency $\omega_\mu = 2\pi\nu_\mu$. The constant of proportionality is the muon gyromagnetic (sometimes *magnetogyric*) ratio,[6]

$$\gamma = 2\pi \times 13.6\, kHz/Gauss \text{ or } 2\pi \times 0.136 MHz/mT. \tag{5}$$

2.3 Local fields

Normally there are nuclear or atomic moments on the host lattice. Consider a paramagnetic phase of localized moments. The dipolar fields from these moments give a *local field* at each muon site which adds vectorially to the applied field. Its value varies from site to site depending on how many neighbours are spin-up and how many spin-down. The result is a spread or distribution $\rho(E)$ of energy levels. This is roughly (but by no means exactly) Gaussian for a dense spin system, *e.g* with moments on every lattice site [15]. In the sketch the rms half-width is indicated as $\hbar\omega_L$. As an example, muons in copper metal experience a distribution of local fields from the Cu nuclear moments which has an rms half-width of about $\omega_L/\gamma_\mu \approx 0.4\,\text{mT}$ (4 Gauss) [10]. For a sparse or dilute spin system, the distribution becomes more Lorentzian.



Broadening of spin-$\frac{1}{2}$ energy levels by a distribution of local fields.

Additional effective fields known as ***hyperfine fields*** act on the muons when electrons with unpaired spin overlap onto the muon site [16]. This is so in itinerant or covalent magnetic systems [5, 8], or when the muon forms muonium or becomes incorporated in open-shell molecules [3, 6].

2.3 Polarization

Suppose that, in an ensemble of muons, a number $n_\downarrow$ are in the spin-down state and $n_\uparrow$ spin-up. *Polarization* is defined as the proportional difference in these populations:

$$P = (n_\uparrow - n_\downarrow)/(n_\uparrow + n_\downarrow). \tag{6}$$

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[6] This is about 3 times the proton value, and therefore larger than that of any other nucleus.

As long as the ensemble is sufficiently large, this applies whether the muons are implanted in bunches (as at a pulsed source), or singly (as at a continuous source). Thus for individual muons, the *probability* of occupying the spin–down state is $(1-P)/2$ and that of the spin–up state $(1+P)/2$.

2.4 μSR

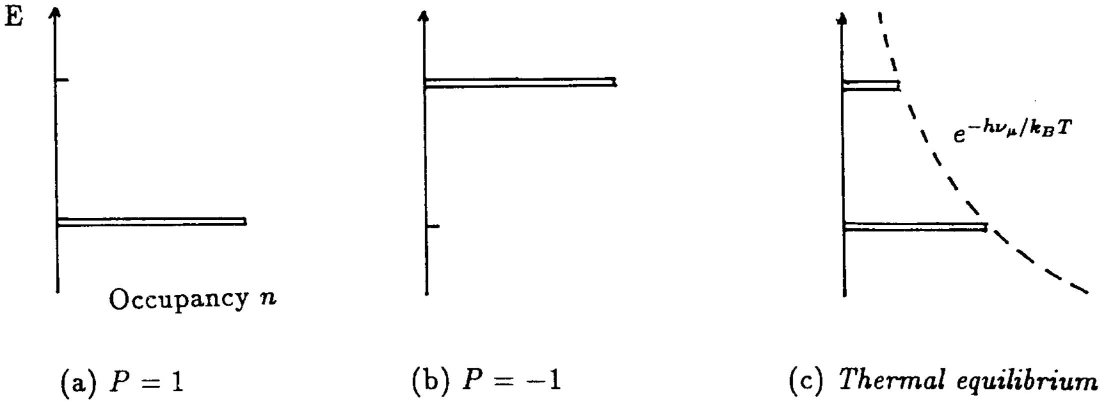
Muon polarization is the ***observable*** in all μSR experiments, monitored ***via*** the asymmetry in the muon decay (Section 3). A variety of behaviours is possible, including precession, oscillation and decay. The terms muon spin ***rotation, relaxation*** and ***resonance*** are used to cover the different types of observation and are introduced below.[7] Evolution of polarization is easiest to visualize for a bunch of muons implanted together at time zero, but an equivalent picture can be built up for muons implanted in successive bunches, or one by one [10].

Most early μSR experiments involved muon spin rotation and, in consequence, most reviews of μSR begin by describing this variant of the technique. Recently, however, ***longitudinal*** and ***zero–field*** experiments have gained considerably in popularity, *i.e.* experiments with the external magnetic field applied parallel to the initial polarization, or with null external field. It now seems appropriate to introduce first the concept of relaxation without precession, in a longitudinal field.

2.5 Longitudinal relaxation

The initial polarization is generally high in μSR experiments. The beam polarization can be as high as 100 % (Section 1.1) and in favourable circumstances this is largely preserved on implantation and thermalization in the sample. Consider the case of a static field which is large compared with the local fields in the sample, and applied so that it is parallel or antiparallel to the polarization direction. That is, the muons are initially all in the spin–up state (as in sketch (a), *i.e* with $P = 1$) or all in the spin–down state (as in (b), with $P = -1$). This is far from an equilibrium situation. In thermal equilibrium, the populations would be related as in sketch (c) by a Boltzmann factor:

$$\frac{n_\downarrow}{n_\uparrow} = e^{-h\nu_\mu/k_BT} \simeq 1 - \frac{h\nu_\mu}{k_BT} \;\ldots\ldots \tag{7}$$



(a) $P = 1$ (b) $P = -1$ (c) *Thermal equilibrium*

Occupancy and polarization.

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[7] The measurement known as ***repolarization*** is also used; this is introduced in subsequent Lectures [3, 9].

This implies that the equilibrium polarization is quite small in all but the most extreme conditions of high field or low temperature. For example, in 100 mT (1 kG), ν_μ =13.6 MHz so that $h\nu_\mu/k_BT \approx 10^{-3}$ at 1 K. The muon polarization should therefore decay away, or *relax*, essentially to zero. This is variously known as ***longitudinal*** or ***Zeeman*** or, in the solid state, ***spin–lattice*** relaxation. It must occur. The only question is whether it takes effect on the timescale of the muon lifetime.

2.6 Relaxation rate

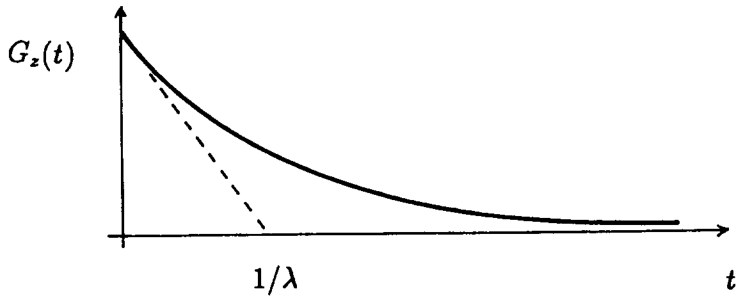
In a genuinely static longitudinal field, each muon is in a stationary state, and no mechanism for relaxation exists. A quantum of energy $\hbar\omega_\mu$ must be supplied or absorbed if the muon spin is to be *flipped*, *i.e.* if a transition is to be made between the spin–up and spin–down states. That is, the muon must experience some time–dependent perturbation which contains frequency components at its Larmor frequency. Fluctuation of the muons' magnetic environment provides the necessary coupling to the thermal ***reservoir*** or ***bath***.[8] This can be caused by fluctuation of the local fields, either a genuine fluctuation due to spin–flip transitions of the host spins [5] or, in the case of muon *diffusion*, an effective time dependence as the muon moves through the lattice [10, 11].

The decay of the polarization is described by a *relaxation function* $G_z(t)$:

$$P_z(t) = P_0.G_z(t) \tag{8}$$

This notation (with $G_z(0) = 1$) covers the situation when the initial polarization P_0 is less than unity. The subscript z indicates longitudinal relaxation, the $\hat{z}$ axis being chosen conventionally along the field direction. In a high external field ($\omega_\mu \gg \omega_L$), time dependent perturbation usually results in an exponential relaxation function [9]:

$$G_z(t) = e^{-\lambda t}. \tag{9}$$



Single exponential relaxation.

$\lambda = -\frac{d}{dt} ln P$ is the *relaxation rate* or inverse time constant, usually expressed in inverse microseconds (μs^{-1}). Other forms of the relaxation can occur, notably ***stretched exponential*** functions in spin-glass systems; in these cases a unique relaxation rate or time constant cannot be defined.

2.7 Dynamic range

The muon lifetime determines the timescale over which the evolution of polarization can be followed:

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[8] In solids, for instance, this is the phonon bath and the relaxation is known as *spin–lattice relaxation*.

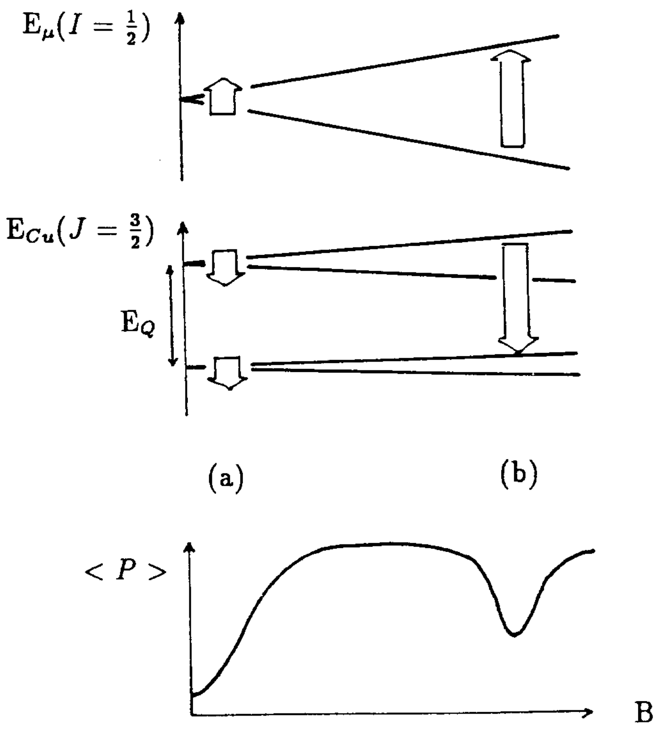
measurements of relaxation rate are most readily made in a 3–decade range, centred roughly on τ_μ^{-1}:

$$0.01 < \lambda < 10\,\mu s^{-1}. \tag{10}$$

This can be stretched or shifted to somewhat lower values at pulsed sources, and to higher values at continuous sources. The corresponding window on the *correlation times* which characterize the fluctuation depends on the strength of the perturbing interaction (usually on the strength of the local fields) [9]. For systems with only nuclear moments present, relaxation in a high longitudinal field is invariably slow on the μSR timescale. It is the proximity of atomic moments, *i.e.* electronic spins, which causes significant relaxation. Particularly potent mechanisms exist when the wave–function of an unpaired electron is centred on, or overlaps onto, the muon site [5, 9, 17]. A ***missing fraction***, that is, a proportion of the initial beam polarization which is unaccounted for in the various observed μSR signals, is invariably the result of depolarization which is too fast to monitor explicitly. A common example is depolarization of those muons which encounter radiolysis products, before these recombine or disperse [21].

2.8 Cross relaxation

Rather confusingly, the term ***relaxation*** is also used to describe the evolution of polarization under a *time–independent* (*i.e.* static) interaction. This is the case when the muons do not find themselves in a spin–eigenstate on implantation. The muons can lose polarization by *flip–flop* transitions when their Zeeman energies match level splittings on neighbouring spins. This is ***cross relaxation.*** The sketch shows two such cases, illustrated for muons in copper metal.



Depolarization by non-resonant (a) and resonant (b) cross–relaxation

At low (or zero) external fields, the spread in local fields allows the flip–flop conditions to be met and a depolarization of the muons is apparent (a). At higher fields ($\omega_\mu \gg \omega_L$) the muons are no longer "on speaking terms" with their neighbours in this way: the muons have a higher gyromagnetic ratio than all other nuclei, *i.e.* a very different Zeeman energy. Equality of the energy level separations is then only achieved if additional terms contribute to the energy level diagrams. For Cu neighbours, as for other nuclei with *quadrupole moments* it is the additional quadrupole energy E_q which allows the condition to be met. This is an example of *resonant* cross relaxation (b) or *level crossing resonance* [18, 19].

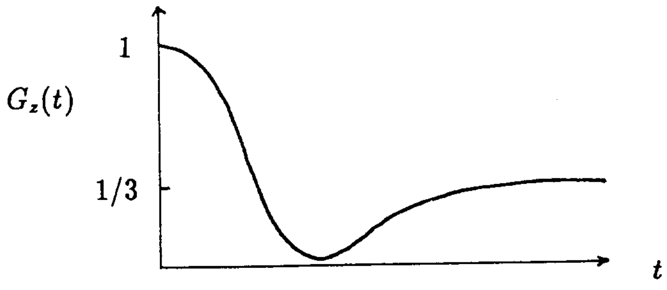
The figure illustrates the effect of non–resonant (a) and resonant (b) cross relaxation on the quantity

$$\langle P \rangle = \frac{1}{\tau_\mu} \int P(t).dt, \tag{11}$$

as measured in *integral counting* experiments [18]. The low–field variation, as non–resonant cross relaxation is suppressed by application of the external field is also called *repolarization.* The field *decouples* the muon and Cu spins. Other important examples of repolarization and level crossing resonance are found when hyperfine interactions are involved [3, 6].

2.9 Zero–field relaxation

The explicit time dependence of the cross relaxation function ($P(t) = P_0 G_z(t)$ in equation (11) above) rarely has a simple analytical form, unlike the high–field dynamical relaxation function of equation (9). But an important case is that of cross relaxation in null external field. This is the *Kubo–Toyabe* function, derived for the idealised case of static local fields which take all possible directions and have a Gaussian distribution of magnitudes [18].[9] The Kubo–Toyabe function takes a rather striking form:



Kubo–Toyabe zero–field relaxation function.

there is an initial Gaussian decay and wiggle which can be thought of as the superposition of oscillatory exchanges of polarization with individual neighbours (or as precession about the individual local fields: see below). Then the polarization recovers to 1/3 of its initial value: for individual muons, the components of their polarization along their local fields are preserved, and the 1/3 tail is the net projection of these components along the original direction. There can be no such long–term memory of the initial polarization if the internal fields fluctuate, or if the muon diffuses, so that this relaxation function is extremely sensitive to the onset of fluctuation. Again, data for copper illustrate the behaviour well.

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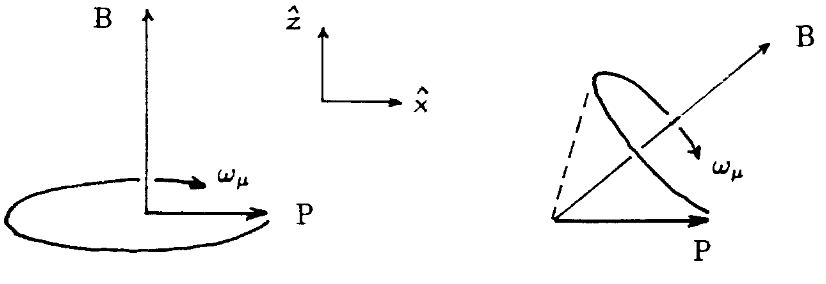
[9] Otherwise, *e.g.* non–Gaussian distribution, some fluctuation, and a small external field, numerical simulations are possible [20].

2.10 Muon spin rotation

The extreme example of muons implanted so that their initial spin state is not an eigenstate is the case of *transverse field*, that is, an external or internal field which is perpendicular to the initial polarization. The muons are neither spin–up nor spin–down with respect to such a field, but in a coherent superposition of the two states. The result is a *precession* or rotation of the polarization about the field direction *at the Larmor frequency corresponding to that field* [16, 10]. For a unique field direction the signal has the same amplitude, but different phase, for observation at different angles in the plane of the precession (a).



(a) (b)

Muon spin rotation, with field (a) perpendicular and (b) oblique to the initial polarization vector.

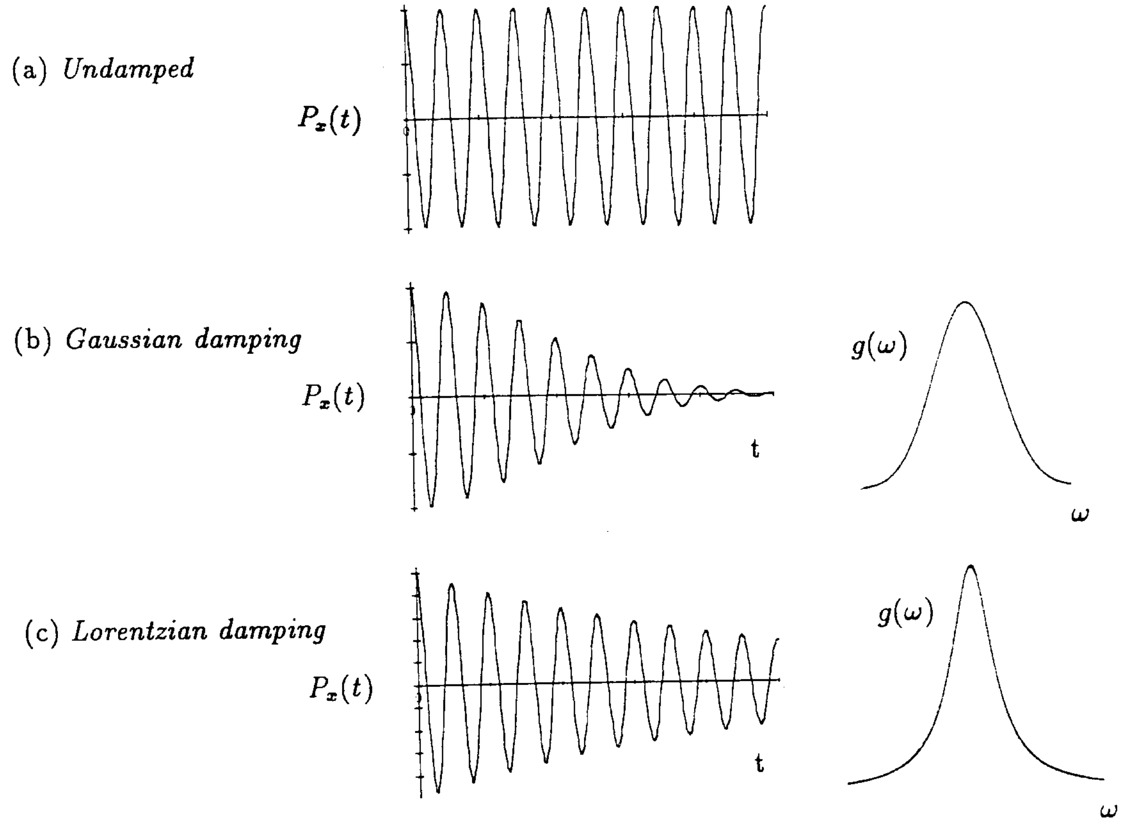
For arbitrary field direction with some transverse component the precession is on a cone rather than in a plane (b). The frequency is unchanged – still determined by the magnitude of the field; the amplitude of the signal is merely reduced somewhat (proportional to the appropriate projection of the oscillatory component of the polarization vector). This means that if the precession results from the internal field of an ordered ferromagnet, or antiferromagnet, a signal may still be seen in polycrystalline or powder samples. The superposition of all possible phases then implies that maximum amplitude is only seen in–line with the initial polarization (and nothing in the perpendicular directions).

2.11 Transverse relaxation

If the implanted muons experience identical transverse fields, they precess in phase (classically stated) so that their polarization is preserved. The μSR signal $P_x(t)$ sketched below, observed in a direction $\hat{x}$ perpendicular to the field, shows undiminished amplitude of the oscillations. If the field varies from site to site, the individual precessions dephase and the signal precession signal is damped. The envelope of the signal $G_x(t)$ is the *transverse relaxation function.* It is usually roughly Gaussian for a static distribution of local fields. Its Fourier transform $g(\omega)$ is the *lineshape*, and essentially maps this field distribution. Some remarkable lineshapes are encountered in type-II superconductors, where implanted muons map the distribution of flux in the vortex lattice.

2.12 Motional narrowing

In summary, μSR frequencies determine the magnetic field at the muon site, and μSR linewidths their variation. Fluctuation of the local fields, or diffusion of the muon, generally results in a *narrowing* of the linewidths, *i.e. less damping of the precession signal* [10]. In this situation the envelope of the time–domain signal changes from Gaussian to exponential and the frequency–domain signal, therefore, from Gaussian to Lorentzian (b – c below). Studies of muon diffusion in metals, especially, have expoited this effect.


(a) *Undamped*

(b) *Gaussian damping*

(c) *Lorentzian damping*

Time and frequency domain signals.

A good many other forms of relaxation functions, both longitudinal and transverse are encountered. Not all have analytical forms [20]. Those due to time dependent perturbation (dynamics) are generally less sensitive to the value of the applied field than those due to time–independent perturbation [5].

2.13 Dynamic range

The precession frequencies which are accessible in muon spin rotation experiments lie in the radiofrequency range. At continuous sources the upper limit is set by the timing resolution of the particle detectors and electronics (Section 3) and is typically about 500 MHz (corresponding to a magnetic or hyperfine field at the muon of 3.7 T). Curvature of the muon beam sets a limit to the the externally applied transverse fields which can be used:[10] for high frequency muon spin rotation experiments a *spin–rotated* beam must be used with the field applied in–line with the beam. With special equipment the high–frequency limit can be pushed to several GHz. At pulsed sources the timing resolution is set by the muon pulsewidth so that the maximum frequency which may be displayed directly may be quite low (roughly the inverse pulsewidth, *e.g.* about 10 MHz for pulses 100 ns wide). Access to high frequency spectra can be regained using RF or microwave *resonance* techniques.

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[10] As a guide, a surface muon beam has a radius of curvature of about 1 m in a field of 100 mT (1 kG).

2.14 RF resonance

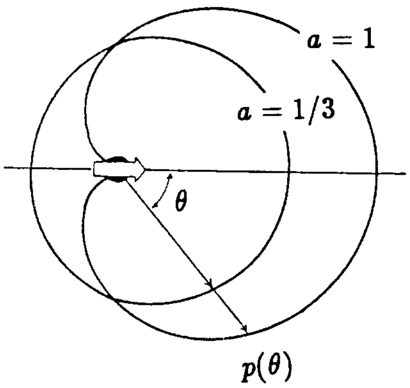
In *muon spin resonance*, muons are implanted with their initial polarization in a longitudinal field, and transitions induced between the Zeeman (or combined Zeeman and hyperfine) levels by applying an auxiliary field which oscillates at the appropriate frequency. (Except for certain hyperfine transitions, the time–dependent field must have some component perpendicular to the main static field.) Radiofrequency fields up to 500 MHz are in common use and microwave techniques for condensed phase studies are under development [19]. Most commonly, resonance is detected as depolarization (loss of forward–backward asymmetry), this technique being amenable to integral counting.

3. HOW IS IT DONE?

In every case, polarization is monitored *via* the anisotropy in the muon decay. For an individual muon, the probability that the positron is emitted at an angle θ to the instantaneous spin orientation takes the form

$$p(\theta) = 1 + a\,cos\theta, \tag{12}$$

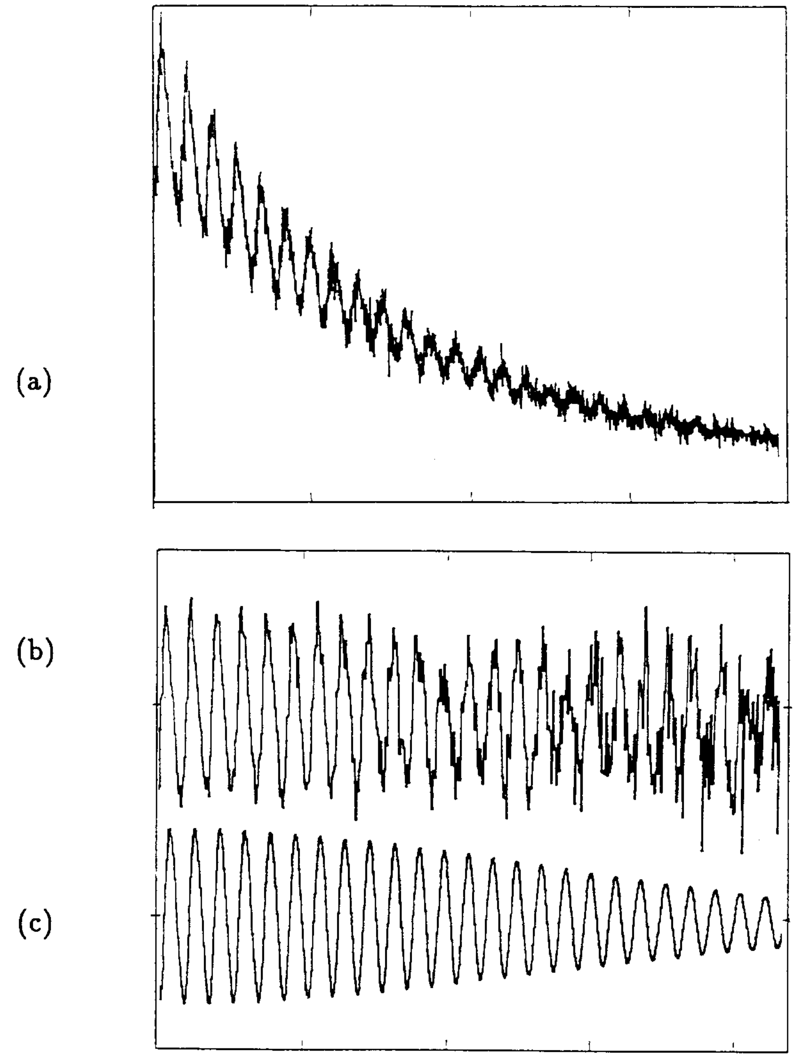
which may be represented as a polar diagram:



Anisotropy of β–emission (P = 1).

(This picture is a 2D cut through 3D surfaces which have axial symmetry about the muon spin direction; i.e. the $a = 1$ surface is apple–shaped.) Here a is the ***asymmetry*** parameter. It is a strong function of the positron energy, being unity for the highest energy positrons (corresponding to decays in which the two neutrinos are emitted in–line backwards) and in fact negative for low energy positrons. Effective values in the vicinity of one third are typical for μSR instruments, depending on the set–up [10, 18]. Thus the ***forward*** ($\theta = 0$) emission probability is about twice the ***backward*** ($\theta = 180$). For an ensemble of muons, therefore, there is a similar polar diagram for the angular distribution of the for positron emission, but with the asymmetry parameter scaled by the magnitude of the polarization. When the polarization vector precesses, as it does when the magnetic field is perpendicular or ***transverse*** to the initial vector, one can imagine the polar diagram locked to the polarization direction.

The positrons may be detected and counted in suitable detectors, usually ***scintillation detectors***. The raw data consists of records of the positron count rate in one or more of these detectors. For some resonance or repolarization experiments a time–average or ***integral*** count is adequate [18]; more usually the time dependence following muon implantation is required. A typical *histogram* is shown in the figure (a). At a pulsed source, the positron count rate is simply recorded following successive pulses to accumulate such histograms; at continuous sources they must be built up by correlating the detection of individual muons and positrons [10]. The histogram represents the muon decay curve ($\sim e^{-t/\tau_\mu}$), modulated by the signal of interest – in this case a precession signal. The *asymmetry* plot (b) can be extracted by multiplying by the inverse exponential (e^{+t/τ_μ}), after background subtraction [10]. Alternatively, it can be extracted from the *forward–backward* asymmetry between a pair of detectors, which is the preferred method for displaying non–oscillatory signals such as longitudinal or zero–field relaxation [18].



μSR histogram (a), asymmetry plot (b) and fit (c).

In the asymmetry plot (b), the divergence in statistical noise reflects the paucity of counts or *events* at long elapsed time. Rather noisy (low statistics) data are chosen in this example to illustrate the *fitting* of a theoretical model to the asymmetry plot [10, 20], in this case a single damped wave with a gaussian envelope or relaxation function (c).

4. BUT WHAT USE IS A PROBE WHICH ONLY LASTS $2\,\mu$s?

The muon decay clearly limits observation time to at most about 10 lifetimes (in practice to about $15\,\mu$s at continuous souces, $20\,\mu$s at pulsed sources): compared with its intensity on implantation ($t = 0$), the β–emission is reduced in intensity by a factor $e^{-10} = 4.5 \times 10^{-5}$ at $t = 10\tau_\mu = 22$ μs. This is a long time scale, however, compared with that for *thermalization* of the implanted muons in the sample (which may vary from nanoseconds in gases to picoseconds in condensed phases) [21]. Except for some cases of *metastability*, which are themselves of special interest, the muons can be said to reach their ground state promptly on implantation. By *state* is meant here the chemical environment – the various possibilities in different types of material are enumerated in the Electronic Structure Lecture [3]. It is not to be confused with the *spin state*, whose evolution provides the primary source of information in μSR experiments. The 10–20 μs timescale is quite adequate to follow this evolution to determine *precession frequencies* or *relaxation rates* over a wide range (sections 2.13, 2.7). In fact it sets a "window" on the timescale of dynamical phenomena which may be studied which proves to be particularly useful, in many instances bridging the gaps between those accessible to other techniques. This applies equally to spin dynamics in magnetism [5, 9], interstitial diffusion [10, 11] and chemical reaction [6].

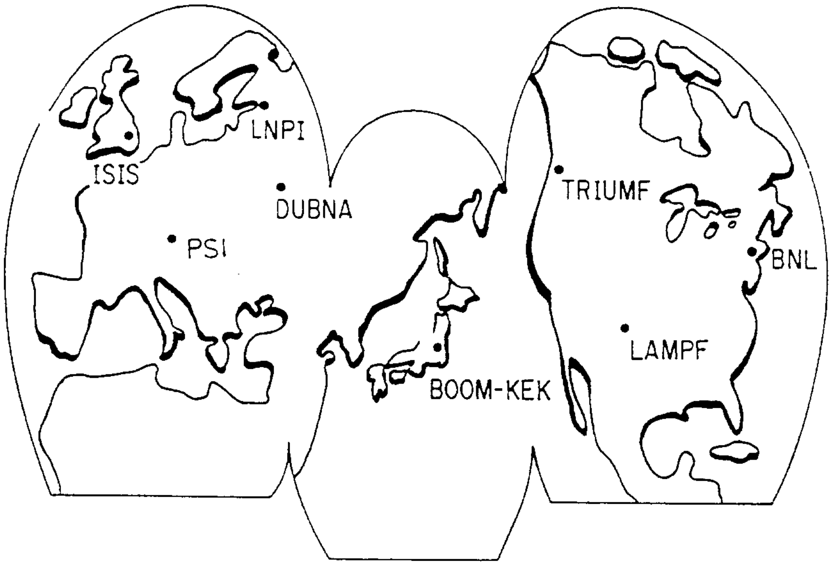
The detection of individual particles also gives μSR spectroscopy a remarkable selectivity and sensitivity. Attention is focussed on the muon alone, and its spin transitions (a Larmor frequency of 1MHz corresponds to an energy quantum of order 10^{-8} eV) are, in effect, detected at the energy of the muon decay (50 MeV). Thought of as a form of trigger–detection [13], this is an enormous enhancement in gain. Spectra are obtained from histograms which contain typically 5–50 million recorded muon decays, depending on the strength of the signal. Counting one muon per molecule, for instance, one can estimate that the whole of muonium chemistry has been established using less than a microgram of muonated material!

5. WHERE IS IT DONE?

μSR experiments can only be carried out at accelerator based muon sources. There are no "portable" sources. Modern sources giving a good intensity (10^3–10^6 or more muons per second) exist at the so–called *meson factories*, designed for pion and muon production, and also at spallation neutron sources, where muon production can be achieved in parallel with neutron production.

The laboratories which presently have the potential for suitable muon production are indicated on the map below. The most powerful of these which, at the time of writing, are scheduled regularly for μSR experiments, are TRIUMF (Vancouver), PSI (near Zürich), ISIS (near Oxford) and KEK (near Tokyo). The others marked, together with former sources at LBL (Berkeley) and CERN (Geneva) have been important in the development of μSR techniques to their present level.

TRIUMF and PSI are *continuous* sources, in the sense that the repetition periods of the primary accelerators are short compared with the muon lifetime; ISIS and KEK are *pulsed*, in the sense that muons are delivered in bursts whose duration is short, and repetition period long, compared with τ_μ. The pros and cons are examined elsewhere [22]. Broadly speaking, high frequencies and fast relaxation rates can only be displayed at continuous sources, pulsed sources have the advantage of virtually background–free spectra and favour the display of slow relaxation functions. They can also be used with synchronous excitation of the sample, notably in cases where the excitation could not be sustained continuously. Time structure of the beam is unimportant for *integral counting* experiments, notably level crossing resonance; LAMPF (New Mexico) holds the current record for overall flux in a single beam, used for this purpose.



Location of muon souces (1993).

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