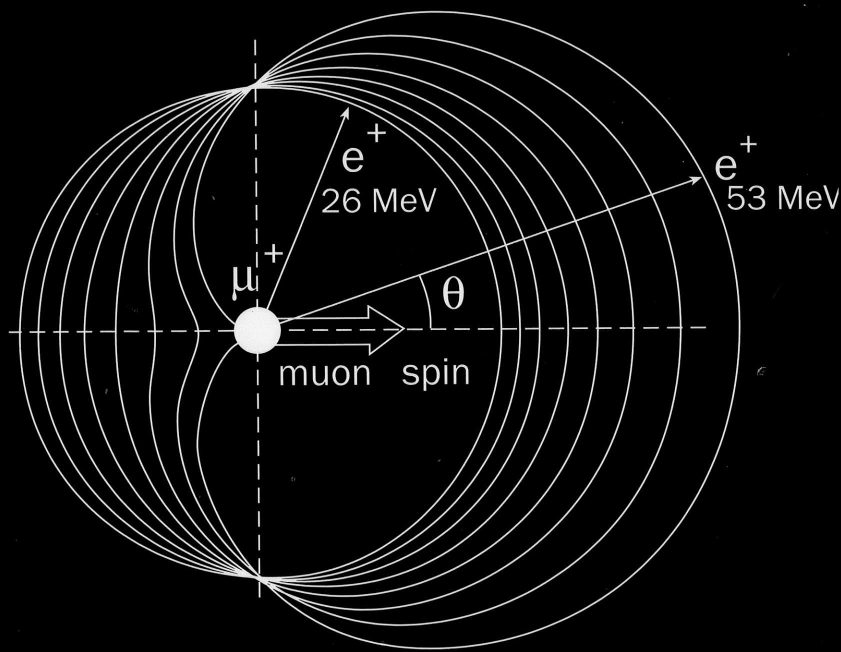
MUON SCIENCE

Muons in Physics, Chemistry and Materials

edited by
S. L. LEE
S. H. KILCOYNE
R. CYWINSKI



Proceedings of the Fifty First Scottish Universities
Summer School in Physics, August 1998

A NATO Advanced Study Institute

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MUONS IN PHYSICS, CHEMISTRY AND MATERIALS

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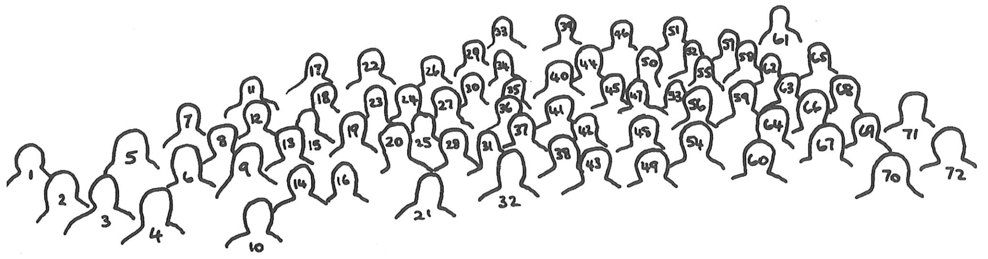

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Director's Preface

Over the last decade the extremely sensitive muon beam techniques known collectively as μSR have become increasingly popular probes of condensed matter, and are now assuming a central role in scientific and technological studies of the solid state within the disciplines of physics, chemistry and materials science. μSR is providing fundamental and often unique information on phenomena as diverse as flux distributions and dynamics in superconductors, moment localization and spin fluctuations in itinerant magnets, heavy fermion and spin glass magnetism, and diffusion processes in metals and semiconductors. Muonium chemistry is being used to probe reaction kinetics and hydrogen bonding at the microscopic level and on nanosecond time scales. Recent developments in the production of slow muons have opened up the possibility of depth profiling in nanostructured materials. Doppler-free laser spectroscopy is being used to excite transitions between the 1S and 2S states in muonium, enabling the most stringent test of QED theory. We are also hearing the first reports of the use of muon telescopes to probe inside active volcanoes for advanced warning of eruptions, the use of muon beam techniques to explore cognitive processes and information transfer within the brain and, perhaps most importantly, of the potential of cold muon catalysed fusion as a new energy source for the third millennium. In addition intense, third generation muon sources are already on the drawing board, promising ever more powerful and sophisticated muon facilities for a growing international community.

Feeling a little overwhelmed by the pace of developments in the apparently all encompassing research field known loosely as Muon Science, Sue Kilcoyne, Steve Lee and I decided that it was perhaps time to take stock. We considered that a summer school, which would bring together young researchers and internationally renowned experts, would be the ideal vehicle for formalizing and publicizing recent key developments in the experimental and theoretical aspects of muon science. With this aim in mind we approached the committee of the Scottish Universities Summer Schools in Physics (SUSSP) and NATO for financial backing. Thanks to the tremendous generosity and support of these two bodies we were able to launch the first NATO Advanced Study Institute in Muon Science and the 51st Summer School in the SUSSP series.

SUSSP51 was held at St Andrews University between 17 and 28 August 1998. The School attracted 80 scientists from 19 countries, who attended 44 lectures, two data analysis workshops, four discussion groups and two poster sessions at which a total of 52 posters were presented. The tour of a whisky distillery, a visit to William Wallace country and the obligatory round of golf were equally well attended! On behalf of the Organizing Committee (Sue, Steve and myself) I have the greatest pleasure in thanking the extremely distinguished international team of lecturers and all the participants at the Summer School for their tremendous support, interest and enthusiasm which made the School such a success, both scientifically and socially.

As Director of the School, and on behalf of the lecturers and the participants, I would also like to extend my warmest thanks to my co-organizers, Sue Kilcoyne (School Secretary) and Steve Lee (School Treasurer) without whose organizational skills, hard work and dedication the Summer School would simply not have been possible.

Finally, as Editors of these proceedings, Steve, Sue and I would like to express our gratitude to all the Summer School lecturers, each of whom has contributed a timely and authoritative chapter to this volume. Our special thanks are also due to Dr Peter Osborne, the SUSSP Series Editor, who has overseen the whole editorial process and expertly handled the final stages of the editing.

Bob Cywinski

St Andrews, April 1999

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μSR: an introduction

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1 Introduction

As even the most cursory inspection of the contents of these Summer School proceedings will reveal, muon science is extraordinarily diverse in its nature and application. On the one hand its practitioners are rigorously probing the fundamental tenets of quantum electrodynamics and exploring and developing the potential of muon catalysed fusion. On the other hand, solid state physicists, chemists and materials scientists are increasingly utilizing an implanted muon's remarkable sensitivity to static and dynamic microscopic magnetic fields to investigate scientifically important and technologically relevant aspects of structural, magnetic and electronic phenomena in magnets, superconductors, semiconductors and insulators. It is these latter studies of the solid state that are generally grouped together under the generic name of "μSR". This acronym was coined by Toshi Yamazaki, Ken Nagamine, Ken Crowe and Jess Brewer in 1974 to grace the cover of the first issue of the μSR Newsletter in which the following definition and explanation was given:

> "μSR stands for **M**uon **S**pin **R**elaxation, **R**otation, **R**esonance, **R**esearch or what have you. The intention of the mnemonic acronym is to draw attention to the analogy with NMR and ESR, the range of whose applications is well known. Any study of the interactions of the muon spin by virtue of the asymmetric decay is considered μSR, but this definition is not intended to exclude any peripherally related phenomena, especially if relevant to the use of the muon's magnetic moment as a delicate probe of matter".

Over the intervening twenty-five years solid state scientists have come to appreciate the tremendous potential of μSR. Correspondingly the number of exponents of the various forms of the μSR technique has grown enormously. Indeed, μSR has now become an almost ubiquitous probe of the solid state. As a sophisticated experimental tool it sits comfortably alongside NMR, ESR, Mössbauer spectroscopy, photoemission, Raman spectroscopy, x-ray scattering and that other ubiquitous probe, neutron scattering, sometimes complementing and sometimes competing, but always providing new and often unique insights into the nature of fundamental physical and chemical processes in materials. Moreover, unlike many other experimental techniques, μSR itself continues to evolve. It is already clear, for example, that entirely new avenues in thin film, multilayer and surface science are being opened and explored through the growing exploitation of newly developed ultra slow muon facilities (Morenzoni, this volume).

In these proceedings many of the key aspects of the application of μSR in solid state science (as well as the more fundamental aspects of muon science) will be addressed. Here, by way of introduction, we intend only to provide an overview of the methodology and realization of the μSR technique, focusing upon the use of the positive muon in rotation and relaxation measurements.

2 The basis of μSR spectroscopy

In marked contrast to microscopic probes such as NMR, Mössbauer spectroscopy and $\gamma\gamma$PAC, for which specific target nuclei are required, μSR is universally applicable as muons can be implanted in any material. However, in order to observe the relaxation, rotation or resonance of muon spins within a sample, it is clear that the incident muon beam should be polarized and of sufficiently low energy to stop within a reasonable thickness of sample. It is extremely fortuitous that both of these two prerequisites are met by positive muons produced in the ordinary two-body decay of charged pions (see for example, Eaton and Kilcoyne, this volume). A remarkable feature of positive pion decay is the maximal violation of parity symmetry which causes the μ^+ to be emitted with perfect spin polarization. In the pion decay process, which occurs with a pion half-life of 26.03ns, the muon emerges in the rest frame of the pion with a momentum of 29.79MeV/c and a kinetic energy of 4.119 MeV. At these energies muons rapidly thermalise within a sample, as shown schematically in Figure 1. Indeed the muon implantation and thermalisation processes occur so rapidly that depolarization is insignificant. This provides μSR with a tremendous advantage as a magnetic resonance technique. Whereas NMR and ESR rely upon a thermal equilibrium spin polarization, usually achieved at low temperatures and high magnetic fields, μSR begins with a perfectly polarised probe, regardless of the conditions in the medium to be studied, and the muon spin degrees of freedom usually start their evolution as far as conceivable from thermal equilibrium.

Once the muons are implanted within a sample their local magnetic environment dictates the subsequent evolution of their spin vectors. If the muons experience a unique off-axis magnetic field (either internal or applied) the spins precess coherently around the field at their Larmor frequency. However any spatial or temporal, site to site, variation of the magnetic field results in a dephasing or depolarization of the muon spin ensemble (see for example, Uemura this volume). This precessional motion and/or depolarisation of the muons spins can be monitored because of the propensity of the muon decay positron to be emitted preferentially along the spin direction of the muon, a further consequence of parity violation in the weak interaction (Figure 2). In a μSR experiment the information on the distribution of local magnetic environments over all muon sites is thus delivered to

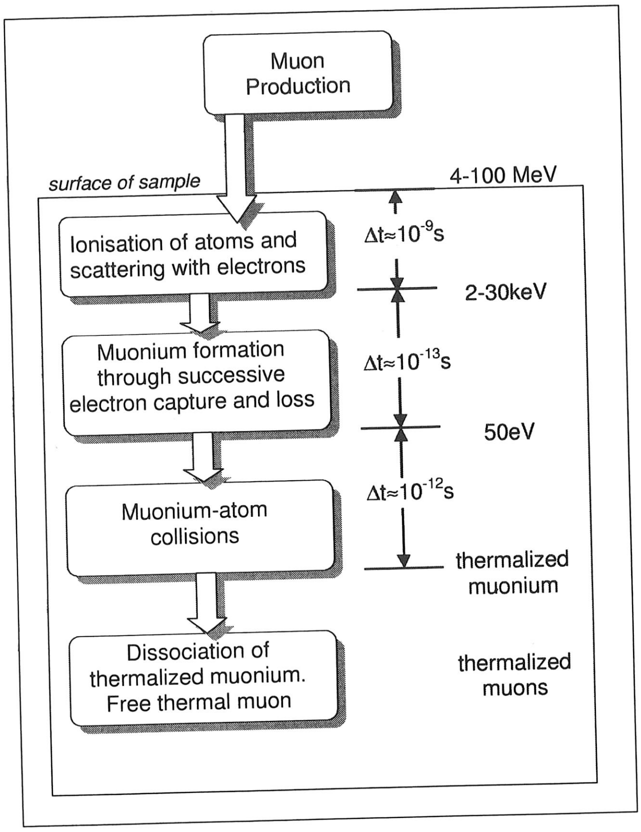

Figure 1. *Schematic representation of the processes of implantation and thermalisation of a muon within a sample. An indication of the time scales and muon energies at each stage of the process is provided. the final stage, that of the dissociation of thermal muonium, does not always occur, in which case it is the behaviour of the muonium atom within the sample that is studied.*

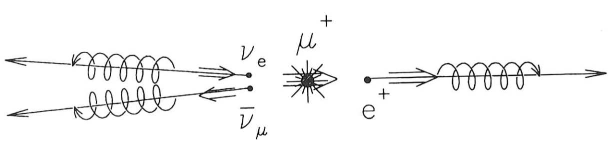

Figure 2. *Positive muon decay; this example gives the highest positron energy and an "asymmetry" of 100% (ie the positron has zero probability of being emitted opposite to the muon spin vector. The half-life for muon decay is 2.197μs*

the experimentalist in the form of relatively high energy (up to 53MeV) decay positrons which readily penetrate sample holders, cryostats or ovens and the detectors used to establish the time and direction of the muon spin at the instant of decay.

Unfortunately the decay positrons are not emitted precisely along the muon spin

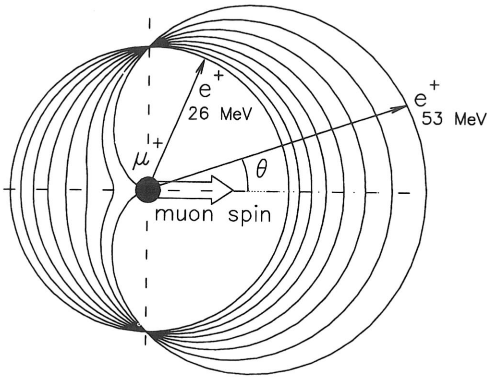

Figure 3. *The angular distribution of positrons from positively charged muons for various positron energies.*

direction but instead are distributed about this direction according to the probability function

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

where θ is the angle between the muon spin and the direction of positron emission. The factor a, known as the asymmetry factor, increases monotonically with the positron energy up to a value of $a = 1$ for the maximum energy of 52.83MeV. The variation of the angular probability function $W(\theta)$ is shown in Figure 3 for a number of decay positron energy. Note that changes sign at low energy. However, very few positrons are emitted with such low energies and those that are will usually not be detected: In any real experiment, some of the lower energy positrons do not penetrate intervening material to reach the detector, whilst others are deflected by applied magnetic fields. In many cases "degraders" are used between the sample and detectors specifically to absorb the low energy positrons and hence maximize the measured asymmetry. The experimentally observed maximum asymmetry A is dependent upon an appropriate integration over the energy dependent probabilities of positron emission and detection, the energy dependent asymmetry and the solid angle of the detector. A is therefore an empirical parameter which must be determined by measurement on a sample known not to produce any muon depolarization but otherwise identical in every respect to the sample of interest. In many cases ultra-pure silver provides a suitable calibrant providing that the self-attenuation of positrons by the silver sample is identical to that of the sample itself. Clearly this calibration can never be perfect, and in general no absolute calibration of A to better that 5% can be obtained. The relative precision in the measurement of A is, however, usually at least an order of magnitude better than this.

3 Experimental geometries

The experimentalist usually has control over the orientation of the detectors, the applied magnetic field and (within some range) the muons' spin polarization. The beam momentum can also be deflected, but this is rarely desirable. In order to designate consistently

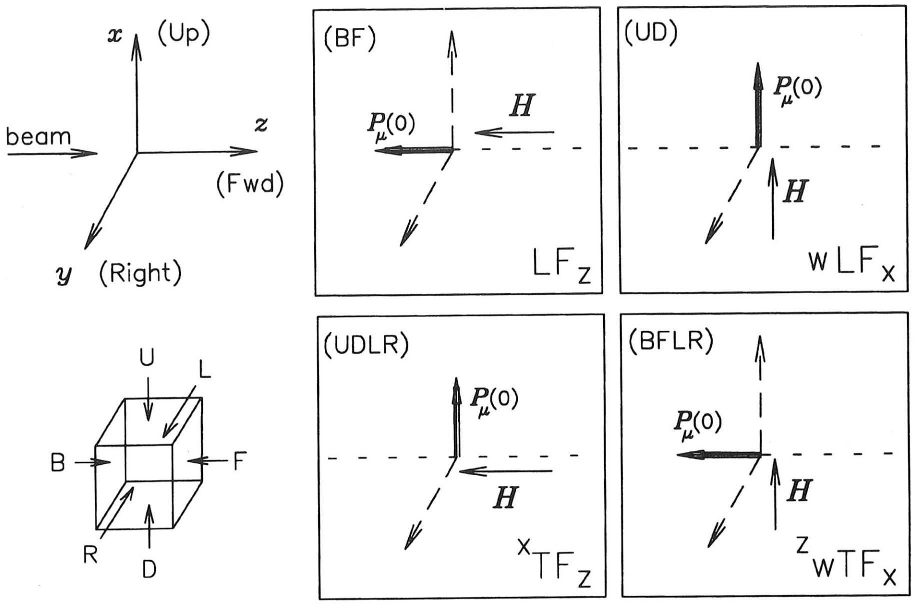

Figure 4. *Conventions for labeling the coordinates in μSR experiments using surface muons. The superscript on the left indicates the direction of polarization of the incoming muon, whereas the subscript on the right indicates the direction of any applied magnetic field. For longitudinal field (LF) and by continuation, for zero field (ZF) both will always be the same, but for transverse (TF) geometry there are in principle two possible arrangements for each choice of super(sub) script, for example* ${}^z wTF_y$ *and* ${}^x wTF_y$. *The letter 'w' indicates a "weak" field, that is a field which is not strong enough to deflect the muon beam appreciably.*

the different orientation choices, the labeling conventions defined in Figure 4 above have been devised specifically for μSR experiments using surface muons which are initially polarized opposite to their momentum vector (and whose spins could be rotated by 90° when a "Wien filter" is present in the beamline). Such flexibility is not available for conventional or "backward" $\mu^\pm$ beams, which will be neglected here partly for that reason. The standard detector array consists of six counters aligned with the positive and negative coordinate axes and labelled F (forward), B (backward), U (up), D (down), L (left) and R (right) according to a "beam's-eye view" naming convention. Note that the unrotated muon polarization points toward the B counter and in the spin-rotated mode toward the U counter; the latter depends, of course, on the orientation of the fields in the Wien filter.

3.1 Transverse field μSR

The simplest and most familiar time-differential (TD) μSR technique is the transverse field (TF) muon spin *rotation* experiment, in which an external magnetic field is applied perpendicular (transverse) to the muon polarization, causing the muon spins to *precess* about the field. Typical transverse field geometries are shown below in Figures 5 and 6.

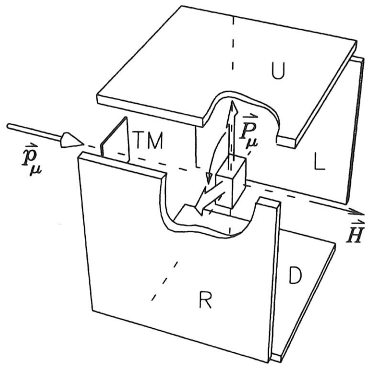

Figure 5. *Sketch of ^xTF_z μSR experimental geometry. p_μ and P_μ refer to the momentum and polarisation vectors of the muon beam respectively. H is the direction of the applied magnetic field.*

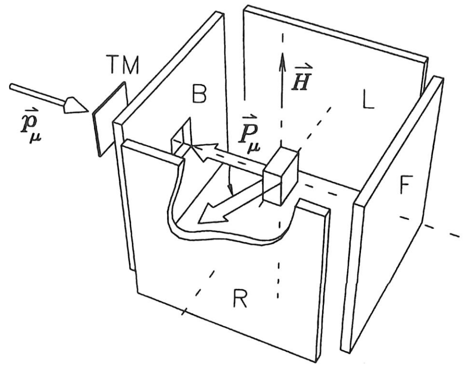

Figure 6. *Sketch of $^zwTF_x$ μSR experimental geometry.*

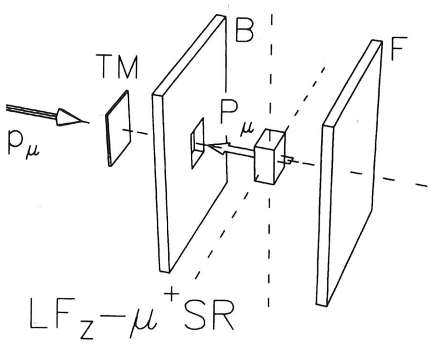

Figure 7. *Sketch of wLF_z μSR experimental geometry*

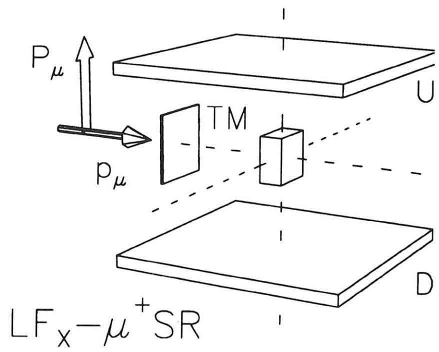

Figure 8. *Sketch of wLF_x μSR experimental geometry*

3.2 Longitudinal and zero field μSR

Consider now the time evolution of the muon polarization in a magnetic field parallel to its initial direction (longitudinal field or LF). If the muon polarization initially has no components perpendicular to the local field then none will develop (although exotic exceptions such as paramagnetic systems in single crystals with highly anisotropic hyperfine interactions may be thought of as having an effective transverse field component) and only the polarization along the initial direction needs to be measured. Two particular experimental geometries for LF μSR are shown in Figures 7 and 8. It should be noted that zero field μSR, which is generally called muon spin relaxation, is simply LF μSR in the zero field limit.

4 The asymmetry spectrum

Extraction of μSR asymmetry spectra from the time histogrammed decay positron spectra taken in a longitudinal geometry requires knowledge of the raw positron time spectrum's normalization N_0 [count/bin] and time-independent background fraction B, neither of which can be extracted numerically from the data without some model of the time dependence of the longitudinal polarization. Instead, one combines the time spectra from two detectors on opposite sides of the sample (such as "B" and "F") in the following way:

Firstly we define the parameters:

$\epsilon_{B,F}$ = efficiency of B or F positron detector

$B_{B,F}$ = beam-borne background in the positron detector

$A_{B,F}$ = intrinsic asymmetry of the positron detector (the count rate is proportional to $1 \pm A_{B,F}$ for incident muons fully polarised along (or opposite to) the symmetry axis of the detector)

$P_z(t)$ = time dependent muon polarization along the z-axis

Thus

$$N_{B,F} = B_{B,F} + N_0 \epsilon_{B,F} \left[1 \pm A_{B,F} P_z(t)\right]$$

where N_0 is a common normalisation. At a continuous muon source it is generally assumed that $B_{B,F}$ is time independent and can be determined using "$t < 0$" time bins in positron spectra, whereas for pulsed muon sources $B_{B,F}$ is vanishingly small (Eaton and Kilcoyne, this volume). The experimental asymmetry $a_0(t)$ is then obtained from either

$$a_0(t) = \frac{[N_B(t) - B_B] - [N_F(t) - B_F]}{[N_F(t) - B_B] + [N_F(t) - B_F]}$$

or

$$a_0(t) = \frac{(1-\alpha) + (1+\alpha\beta) A_B P_{B,F}(t)}{(1+\alpha) + (1-\alpha\beta) A_B P_{B,F}(t)}$$

where $\alpha = \epsilon_F / \epsilon_B$ and $\beta = A_F / A_B$ and $(1-\alpha)/(1+\alpha)$ is the "baseline" asymmetry for totally unpolarised muons.

In addition to the obvious "baseline-shift" there is also a more subtle distortion in $a_0(t)$ for $\alpha\beta \neq 1$ where it appears that a plot of $a_0(t)$ has a non-linear scale for the abscissa. To remove these distortions completely α and β must be determined independently, usually by fitting spectra taken in a weak transverse field, but with the muon beam, sample and detector geometry. The corrected asymmetry is then

$$A_B P_z(t) = \frac{(\alpha - 1) + (\alpha + 1) a_0(t)}{(\alpha\beta + 1) + (\alpha\beta - 1) a_0(t)}.$$

These corrections apply equally to transverse field μSR asymmetry spectra formed from opposing pairs of detectors. However, β must be determined from simultaneous fits to the "raw" spectra, $N_{B,F}(t)$, collected in TF geometry in opposite detectors. It is not

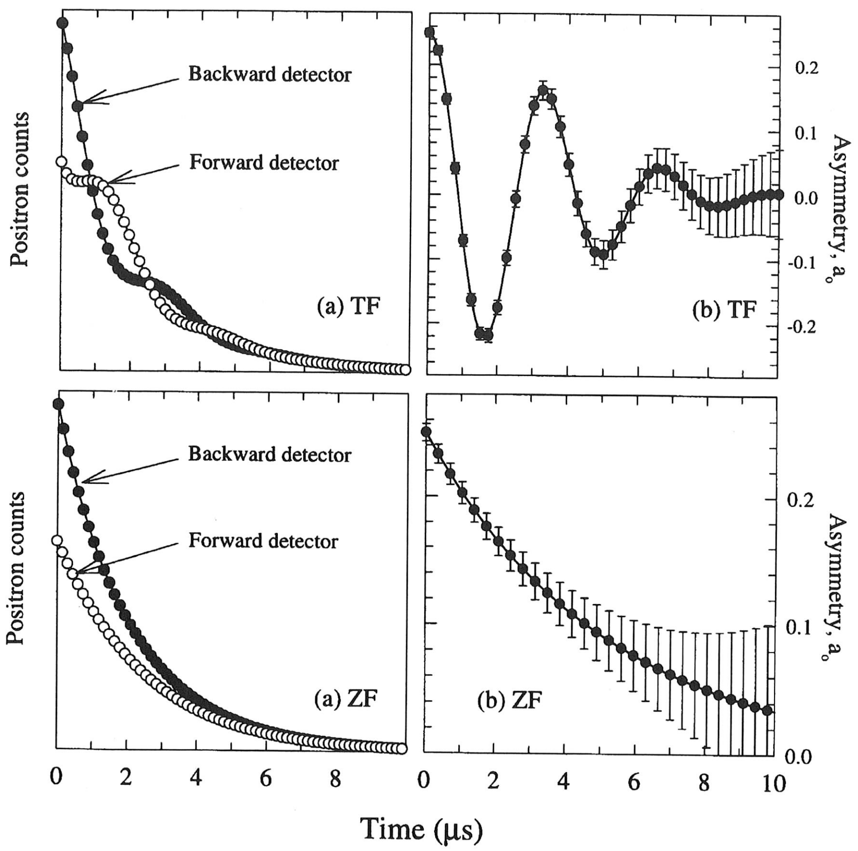

Figure 9. *(a) Simulated raw positron spectra in forward and backward detectors for transverse field (TF) and zero field (ZF) geometries with (b) the associated corrected asymmetry spectra. The simulation assumes $N_{B,F}(t=0) \approx 10^4$ counts.*

unusual to assume that $\beta = 1$, although in principle one should always determining this empirical parameter as accurately as possible. On a pulsed muon source, for which the beam-borne background is negligible, it is usual to make the simple calculation

$$a_0(t) = \frac{N_B(t) - \alpha N_F(t)}{N_B(t) + \alpha N_F(t)}.$$

α is obtained from a fit to transverse field data measured in a field of 2–4mT. The appropriate α is found when the transverse field spectrum calculated with this expression oscillates symmetrically about the time axis. For typical μSR experiments a_0 will take a value of between 0.2 and 0.27.

Raw positron histograms, $N_{B,F}(t)$, simulated for transverse field muon spin relaxation and zero field muon spin relaxation, together with their associated corrected asymmetry spectra, $a_0P(t)$, are shown in Figure 9. Note the rapidly increasing experimental uncertainties in $a_0P(t)$ at longer times. Some important consequences of these experimental statistics will be discussed in the Appendix to this volume (Rainford).

It is generally the goal of the experimentalist to extract the spectral parameters a_0 and $P(t)$ from TF, LF or ZF data for a given sample as a function of temperature, magnetic field, pressure etc. It is these spectral parameters that embody crucial information on the spatial distributions and dynamical fluctuations of the muons' magnetic environment averaged over the sample volume. In many of the following chapters the methods of modelling and interpreting $P(t)$ will be presented and discussed in detail for a wide range of topical problems in solid state science.

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