The Application of Intense Beams of Positive Pions and Muons in Solid-State Physics

Alfred Seeger

Max-Planck-Institut für Metallforschung, Institut für Physik, Stuttgart, and Universität Stuttgart, Institut für theoretische und angewandte Physik, Postfach 80 06 65, D - 7000 Stuttgart 80, Germany.

Abstract. It is pointed out that while the proton accelerator requirements for meson factories and for spallation sources are very similar, the target requirements (thin targets of light nuclei for π-meson (pion) production, thick targets of very heavy nuclei for spallation) are complimentary. It is therefore possible to use, with very little interference, the same proton beam for neutron production by the spallation process and pion production. From the pion decay muon beams may be obtained. The muon and pion beams of a high-intensity spallation installation may possess properties that make them extremely interesting for solid-state (and neutrino) physics. This is particularly true for beams with appropriate time structures, namely pulse lengths $\Delta t_1$ that are short compared with the lifetimes $\tau_{\mu} = 2.6 \times 10^{-8}$ s or $\tau_{\pi} = 2.2 \times 10^{-6}$ s, and periods $\Delta t_2$ that are long compared with these lifetimes.

The present paper outlines the basic physics of positively charged pions and muons and of their behaviour in condensed matter. This behaviour may be studied by two quite different techniques, viz. lattice steering (channelling) of the positive muons ($\mu^+$) resulting from the $\pi^+$ decay or of the relativistic positrons ($e^+$) resulting from the $\mu^+$ decay, and muon spin rotation ($\mu$SR). Both $\pi^+/\mu^+$ and $\mu^+/e^+$ lattice steering have recently been demonstrated experimentally. The lattice-steering techniques have the advantage that they are universally applicable. With more intense $\mu^+$ and $\pi^+$ beams of suitable time structure the time-differential versions of the lattice-steering techniques are very promising for the study of crystal defects that can be decorated by $\pi^+$ and $\mu^+$.

$\mu$SR constitutes a well established technique based on the non-conservation of parity in Weak Interactions. It may be applied to a wide variety of materials with suitable magnetic properties. These include nuclear magnetism, ferro- and antiferromagnetism, spin polarization of conduction electrons, orbital magnetism of electrons in superconductors with non-homogeneous magnetic structures, and muonium atoms. So far the great majority of the $\mu$SR experiments were done in the so-called individual time-differential mode. When stronger beams with suitable time structures become available, resonance and collective-mode $\mu$SR methods will give access to problems that could not be investigated with the classical $\mu$SR methods.
1. Introduction

Muons (originally called mesotrons or $\mu$-mesons) and pions ($\pi$-mesons) were discovered in the study of the cosmic radiation. Incoming high-energy protons react in the upper atmosphere with light nuclei and produce positively charged ($\pi^+$), neutral ($\pi^0$), and negatively charged ($\pi^-$) pions together with heavier particle such as K mesons etc. The neutral pions decay very rapidly mainly into two $\gamma$ quanta (lifetime at rest $\tau_{\pi^0}=8.4\times10^{-6}$ s). They have so far not found any application in condensed-matter physics and will not be discussed here further. The charged pions decay with a much longer rest lifetime $\tau_{\pi^\pm}=2.60\times10^{-8}$ s through a so-called semi-leptonic decay

$$\pi^\pm \rightarrow \mu^\mp + \nu_\mu$$

into muons ($\mu^-, \mu^+$) and muon (anti-) neutrinos ($\nu_\mu, \bar{\nu}_\mu$). The muons (lifetime at rest $\tau_\mu=2.197\times10^{-6}$ s) are sufficiently long-lived for many of them to reach (with the help of the relativistic time dilatation) the ground level of the Earth. The cosmic-ray intensity at ground level is almost entirely due to high-energy muons (Fig. 1, upper left-hand corner).

In meson factories the processes occurring in the cosmic radiation are reproduced, though on different energy and intensity scales. Protons from accelerators with typical energies between about 0.5 and 1.1 GeV fall on thin targets of light nuclei (e.g., Be or graphite), where $\pi^+$, $\pi^0$, and $\pi^-$ are generated. At the time of the foundation of CERN (~1950) the largest accelerators conceived were mainly planned for the production of $\pi$-mesons to be used in nuclear and elementary-particle physics. Since then the development of the large proton accelerators has branched into that of high-energy accelerators, aiming at higher and higher energies, usually at the cost of intensity, and that of "middle-energy" accelerators or meson factories, whose purpose is to provide intense beams of charged pions and muons. The three meson factories in operation are SIN (Schweizerisches Institut für Nuklearforschung = Swiss Institute of Nuclear Research, Villigen, Aargau, Switzerland), LAMPF (Los Alamos Meson Physics Facility, Los Alamos, New Mexico, USA), and TRIUMF (Tri-University Meson Facility, Vancouver, Canada). An intermediate place is occupied by the booster machines of high-energy proton accelerators, which may operate in energy and intensity ranges comparable to those of the meson factories or at least of meson factorettes such as those at CERN, Dubna and Gatchina (USSR). At one of them, viz. the Japanese high-energy physics laboratory KEK at Tsukuba, Ibaraki, Japan, the Booster Meson facility BOOM has recently gone into operation [1,2].

The proton accelerators of spallation neutron sources work in the same energy range as meson factories and should produce, in order for the spallation sources to be competitive with high-flux reactors, proton currents which are comparable with or larger than those of the existing meson factories. In spallation sources the neutrons are produced in thick targets of very heavy nuclei (e.g., Pb-Bi, U). This means that while the accelerator requirements for spallation sources and meson factories are essentially the same with regard to proton energy and intensity (time structure will be considered separately below), the target requirements are complementary. It is thus feasible to use simultaneously and with little interference the same proton beam for both the production of neutrons by the spallation process and the generation of intense beams of charged pions and, through the decay reaction (1), of muons (comp. Fig. 1). In view of the very widespread and rapidly growing applications of pions and muons this is an extremely interesting possibility. Without going into further detail we mention that according to (1) intense
The present paper will concentrate on the use of positive pions and muons in solid state physics, emphasizing the new possibilities that will be opened up if very intense and/or pulsed beams become available. Fig. 1 gives a survey of the main configurations envisaged and discussed in detail below. We should nevertheless like to draw attention to just one application of negative pions, viz. their use in cancer therapy [4]. This development (in the view of the present author of enormous future potential) has recently been started at LAMPF and SIN and might be greatly accelerated by the availability of more intense beams.

2. Basic Physics

Since (1) is a two-body decay, in the pion rest system the muons obtained from (1) possess a fixed energy (corresponding to kinetic energy $E_{\mu}^{\text{kin}} = 4.12$ MeV, linear momentum $p_{\mu} = 29.79$ MeV/c, and velocity $v_{\mu} = 0.27 c$). Pions are spinless particles. Since by their nature neutrinos are 100% left-handed polarized, conservation of angular momentum demands that in the $\pi^+$ rest system the $\mu^+$ generated by (1) are 100% left-handed polarized, too (Fig. 2). (This is opposite to the "natural" polarization of anti-leptons such as $\mu^-$ and $e^+$, and hence the reason why only $1.2 \times 10^{-4}$ of the $\pi^+$ decay into positrons $e^+$ rather than into positive muons.)
Positive muons decay according to the leptonic reaction (β-decay)

\[ \mu^+ \rightarrow e^+ + \nu_e + \bar{\nu}_\mu + \nu_e \tag{2} \]

where \( \nu_e \) denotes the electron neutrino. The decay positrons possess an energy spectrum with maximum values (in the \( \mu^+ \) rest system) \( p_{e_{\text{max}}} = 52.8 \) MeV/c and \( v_{e_{\text{max}}} = 0.99999 c \). Because of parity non-conservation the emission probability of the positrons is asymmetric with respect to the direction of the muon spin \( \mu \). Averaged over all positron energies its angular dependence is given by (comp. Fig. 2)

\[ \bar{p}(e^+) \propto 1 + \frac{1}{3} \cos \theta \tag{3} \]

This permits us to study, by observing the directional distribution of the decay positrons, the muon polarization at the time of the decay and to obtain, on account of the coupling between spin and magnetic moment of the muons, information on the "magnetic history" of the muons. This is the basic idea of the μSR (Muon Spin Rotation) technique, which was already employed in the work [5] establishing the parity non-conservation in (2) and which has since found wide application in nuclear, atomic, and solid-state physics [6-8] as well as in chemistry [9] and biophysics [10]. Like neutrons, \( \mu^+ \) and \( \pi^+ \) may be used as probes for the study of condensed matter. In contrast to neutrons, however, \( \mu^+ \) and \( \pi^+ \) interact with their environment predominantly through the electrostatic interaction and are in this respect analogous to positrons [11]. This has two important
thermalization: $10^{-12}$s-$10^{-14}$s $\ll T_{\pi^+}, T_{\pi^0}, T_{\mu}$

| principal | electrostatic $\rightarrow$ interstitial sites trapping at defects $\mu^-$ "atom" formation |
| interaction: | |
| diffusion: | strongly mass-dependent (isotope effect) may lead to trapping at defects |
| information transmitted in decay or annihilation: | $e^+$ | $\mu^+$ | $\pi^+$ |
| | $e^-$ charge and momentum density ($e^+$ lifetime, angular correlation, Doppler broadening) | "magnetic history" ($\mu$SR) | location in crystal (lattice steering: blocking or flux enhancement) |
| interpretation: | involved | direct |

Fig. 3: Positively charged probes in condensed matter

Consequences. First, in condensed matter positive pions and muons as well as positrons are slowed down to thermal velocities ("thermalized") in times that are short compared with their lifetimes. With respect to their kinetic and potential energy they are hence most of the time in thermal equilibrium with their environment. This means that they constitute indeed, like neutrons but for different reasons, well-defined probes. Second, the Coulomb repulsion by the nuclei keeps the positively charged probes as far away from the nuclei as possible. In perfect crystals this leads to the localization of the probes in lattice interstices. In imperfect crystals, in addition bound states may be formed at sites where nuclei are missing (e.g., vacancies) or where extra space is available (e.g., on the dilation side of edge dislocations).

In thermal equilibrium $\pi^+$ and $\mu^+$ diffuse by hopping from interstice to interstice. If their diffusivity $D^+$ is high enough, they may be trapped at imperfections providing bound states for them. Because of the ratio of 85 in their mean lifetimes the probability that this occurs is much higher for $\mu^+$ than for $\pi^+$ under otherwise equal conditions. This may be used to determine the mass dependence (isotope effect) of the diffusivity of point particles of unit positive charge (mass ratios proton: pion:muon $m_p : m_{\pi^+} : m_\mu = 1 : 1/7 : 1/9$), which is of great interest for the theory of quantum diffusion in crystals [12-14] and for a deeper understanding of hydrogen diffusion.

As indicated in Fig. 3 the information on location and trapping of the probes is transmitted by the decay or annihilation products, in the case of

1) In the case of positive pions this suppresses the Strong Interaction of these hadrons with the nuclei. During the implantation the suppression is achieved by keeping the pion momenta at or below about 150 MeV/c and thus below the pion-nucleon resonances.
by the $\mu^+$, in the case of $\mu^+$ by the $e^+$, and in the case of $e^+$ by the annihilation $\gamma$'s (usually in the $2\gamma$-annihilation mode). With regard to the ease of interpretation of this information big differences between the three probes exist. Since in the case of positrons the behaviour of the probes in condensed matter and the annihilation process are determined by the same interaction (viz. the electromagnetic interaction) the quantitative interpretation of the information transmitted by the annihilation $\gamma$'s requires rather involved calculations. These are made particularly difficult by the fact that electrons and positrons possess the same mass, so that the recoil of the probes has to be allowed for and the usual approximations in the electron theory of "impurities" in crystals become inapplicable. The situation is much simpler for muons and pions, since the decay reactions (1) or (2) are controlled by the Weak interaction and therefore (for all practical purposes) independent of the electrostatic interaction which governs the behaviour of the probes in condensed matter. The latter is many orders of magnitude stronger than the interaction between the magnetic moments of the muons and those of the constituents of matter - nuclei and electrons - so that the tracing of the "magnetic history" of the muons by the $\mu$SR technique (see Sect. 3) does not interfere with their behaviour as charged probes.

The situation is even more striking for the other technique that can be used to obtain information on $\pi^+$ or $\pi^+$ in crystals, i.e. the lattice steering of the charged decay products $e^+$ or $\mu^+$ \[15\] (see Sect. 4). Here the "message" carried by the decay particles is accumulated only after the decay and disappearance of the probes, so that any interference with their behaviour is impossible.

The combination of the properties just described with the fact that the diffusivities of $\mu^+$ and $\pi^+$ can be varied within fairly wide limits by varying the temperature makes these particles ideal probes, in particular for "decorating" defects in crystalline solids \[11\]. It is true that on account of their larger masses the diffusivities of $\mu^+$ and $\pi^+$ are substantially smaller than those of positrons, but this is compensated by their much longer lifetimes. (Typical positron lifetimes in metals are about $2 \cdot 10^{-10}$ s.)

3. The $\mu^+$SR Technique

The various $\mu$SR techniques all make use of three basic facts, namely (i) that muons possess a magnetic moment (of a magnitude 4.65 times that of the neutron, corresponding to a gyromagnetic ratio $\gamma_\mu = 8.516 \cdot 10^8$ T·s⁻¹g⁻¹), (ii) that they are generated according to (1) with longitudinal polarization, and (iii) that the polarization at the time of decay may be detected by observing the directional distribution of the positrons from the decay reaction (2). $\mu$SR is similar to NMR (Nuclear Magnetic Resonance) or EPR (Electron Paramagnetic Resonance) but much simpler in several respects. Because of (i) high magnetic fields in order to produce a detectable polarization are not needed. Because of (ii) changes in the polarization may be detected without having to employ high-frequency radiation. Moreover, the theoretical interpretation is often much more straightforward than in NMR, since the interaction between muon spins is completely negligible on account of the extremely small number of muons in the samples at any one time.

In the discussion of the various $\mu$SR techniques it is convenient to distinguish between transverse measurements, in which the average field $B_\mu$ acting on the magnetic moments of the muons is perpendicular to the muon polarization $P$, and longitudinal measurements, in which the muon polarization precesses in the plane perpendicular to $B_\mu$ with the angular velocity

$$\omega_\mu = \gamma_\mu B_\mu$$

(4)
The transverse polarization $P_2$, that is the polarization component perpendicular to $B$, stays constant if all muon moments experience the same magnetic field. If this is not the case, the muon spins may precess at somewhat different angular velocities, and the transverse polarization $P_2$ "relaxes" gradually towards its equilibrium value zero. This may be described by the transverse relaxation function

$$R_2(t) = \frac{P_2(t)}{P_2(0)},$$

where $P_2(t)$ is the transverse polarization of an ensemble of thermalized muons at time $t$ after thermalisation.

A longitudinal field $B_\mu$ gives rise to Zeeman levels separated by

$$\Delta E = \hbar \omega_\mu,$$

where $\omega_\mu$ is given by (4). Transitions between the Zeeman levels are introduced by magnetic fields with circular frequency $\omega_\mu$ and non-vanishing component perpendicular to the muon polarization. Such time-dependent fields may either be external fields, or may result from different magnetic fields at different muon sites, so that diffusing muons experience magnetic fields that vary with time.

The relaxation of the initial longitudinal polarization $P_1(0)$ of a muon ensemble to its equilibrium value $P_{eq}$ may be described by means of a longitudinal relaxation function $R_1(t)$:

$$R_1(t) = \frac{P_1(t) - P_{eq}}{P_1(0) - P_{eq}}.$$

If we consider transitions between the Zeeman levels of the $\mu^+$ only, we are dealing with a two-level system. Hence the longitudinal relaxation function has, to a very good approximation, the form

$$R_1(t) = \exp(-\Gamma_1 t),$$

so that the information obtainable from longitudinal relaxation measurements is contained in the field and temperature dependence of the longitudinal relaxation rate $\Gamma_1$. By contrast, the functional form of the transverse relaxation function $R_2(t)$ depends on the type of magnetic interaction involved and on the motion of the muons. Important special cases are the exponential function

$$R_2(t) = \exp(-\Gamma_2 t),$$

which is usually encountered when the muons diffuse very rapidly, and the Gaussian

$$R_2(t) = \exp(-\Gamma_0 t),$$

which is characteristic for immobile muons and a Gaussian distribution of the magnetic fields at the muon sites [16,17].

How are precession or Zeeman frequencies $\omega_\mu$ and relaxation functions $R_i(t)$ measured? As indicated in Fig. 4, the available techniques may be grouped into the individual mode, the collective mode, and the high-frequency or resonance technique, in which an external high-frequency field is employed.
In the individual mode (which is the one used by most groups in the field and responsible for most of the μSR results so far obtained) the entry and the decay times of individual muons as well as the directions in which the decay positrons emerge are recorded. From the observation of many such events [typically (1 - 30) \times 10^6] the decay rate \( \dot{N}_1 \) in each of the so-called positron telescopes (scintillation counters) is recorded as a function of the time difference between entry and exit (Fig. 4). From this information the relaxation functions (and in the transverse case also the precession frequency \( \omega_m \)) may be obtained. A prerequisite of the method is that with high probability there is only one muon in the sample at any one time (unless one uses counters which enable him to differentiate between different parts of the sample from which the positrons are coming). This limits sharply the rate at which useful events can be accumulated and, because of stability problems, also the smallest relaxation rate that can be detected. At the present time this limit appears to be \( \Gamma_2 \approx 1/10 \ddot{N}_1 \approx 0.05 \cdot 10^{-6} \text{s}^{-1} \).

The collective mode requires pulsed muon beams. If the pulse length \( \Delta t_1 \) is small compared with the muon lifetime \( \tau \) and if the repetition period \( \Delta t_2 \) of the pulses is much longer than \( \tau \), it is possible to use the collective entry of the muons of a pulse as zero-time signal. The decay times and the directions of positron emission may either be determined as in the individual mode with the help of positron telescopes and fast electronics by the time-differential technique, or "macroscopically" by measuring in a Cerenkov counter the superposition of all delayed events following a muon burst [2]. In the collective mode there is no inherent limit to the rate of data accumulation. It is therefore feasible to detect substantially smaller relaxation rates and precession frequencies than in the individual mode simply because

<table>
<thead>
<tr>
<th>Mode</th>
<th>Optimizing</th>
<th>Transverse measurement</th>
<th>Longitudinal measurement</th>
</tr>
</thead>
<tbody>
<tr>
<td>Individual</td>
<td>START: entry of individual ( \mu^+ )</td>
<td>( \dot{N}_1 = \frac{m_e \Gamma_1}{\tau} )</td>
<td>( \dot{N}_1 = \frac{m_e \Gamma_1}{\tau} )</td>
</tr>
<tr>
<td>STOP: generation and direction of ( e^- ) from same ( \mu^+ )</td>
<td>( \tau )</td>
<td>( \tau )</td>
<td></td>
</tr>
<tr>
<td>Collective</td>
<td>START: entry ( \mu^+ ) pulse at length ( \Delta t_1 )</td>
<td>( \dot{N}_1 = \frac{m_e \Gamma_1}{\tau} )</td>
<td>( \dot{N}_1 = \frac{m_e \Gamma_1}{\tau} )</td>
</tr>
<tr>
<td>STOP: generation and direction of ( e^- ) from same ( \mu^+ ) in pulse</td>
<td>( \Delta t_1 )</td>
<td>( \Delta t_1 )</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 4: \( \mu^+ \)SR techniques
there may be more muons that live long enough. The use of pulsed beams is particularly advantageous in longitudinal measurements, where the elimination of the background constitutes a serious problem when continuous $\mu^+$ beams are employed. The main disadvantage of the collective measurements is that they are not suited for the study of fast relaxation processes or rapid muon spin precession.\footnote{The shortest relaxation times or spin precession periods that can be measured in the collective mode are determined by the time interval $\Delta t_0$ over which the muons coming from the same pulse enter the sample. $\Delta t_0$ may be made shorter than $\Delta t_1$ by letting the muons pass through time-dependent magnetic fields arranged in such a way that the paths of muons generated at the beginning of the pulse are longer than those generated towards the end. The compression of a pulse of muons with $v/\gamma = 0.27c$ by $1 \cdot 10^{-7}$ s requires a path difference of $8 \, \text{m.}$}

Of course, the advantages of the collective mode can only be fully realized if the muon beams not only possess the right time structure and high fluxes within the muon pulses but also a sufficiently large average flux. In this regard it should be noted that because of the exponential time law of the muon $\beta$-decay the minimum relaxation rates or precession frequencies that may be still be detected increases only logarithmically with the muon flux.

Collective-mode measurements have recently been performed at BOOM with $\Delta t_1 = 5 \cdot 10^{-8}$ s, $\Delta t_2 = 5 \cdot 10^{-2}$ s, and $3.5 \cdot 10^5 \, \mu^+$ per pulse\footnote{The shortest relaxation times or spin precession periods that can be measured in the collective mode are determined by the time interval $\Delta t_0$ over which the muons coming from the same pulse enter the sample. $\Delta t_0$ may be made shorter than $\Delta t_1$ by letting the muons pass through time-dependent magnetic fields arranged in such a way that the paths of muons generated at the beginning of the pulse are longer than those generated towards the end. The compression of a pulse of muons with $v/\gamma = 0.27c$ by $1 \cdot 10^{-7}$ s requires a path difference of $8 \, \text{m.}$}. It has indeed been possible to obtain a preliminary relaxation rate $\Gamma_2 > 10^{-6}$ s$^{-1}$ for indium at 393 K, a quantity that due to its smallness could not be reliably determined in earlier individual-mode measurements.

The high-frequency or resonance $\mu$SR technique comes closest to the familiar NMR or EPR techniques\footnote{The shortest relaxation times or spin precession periods that can be measured in the collective mode are determined by the time interval $\Delta t_0$ over which the muons coming from the same pulse enter the sample. $\Delta t_0$ may be made shorter than $\Delta t_1$ by letting the muons pass through time-dependent magnetic fields arranged in such a way that the paths of muons generated at the beginning of the pulse are longer than those generated towards the end. The compression of a pulse of muons with $v/\gamma = 0.27c$ by $1 \cdot 10^{-7}$ s requires a path difference of $8 \, \text{m.}$}. An external hf field with a non-vanishing magnetic component perpendicular to the muon polarization $P$ induces transitions between the Zeeman levels if its circular frequency $\omega_{hf}$ satisfies the condition $\omega_{hf} \approx \omega_0$. The muon-spin reversals associated with these transitions reduce the asymmetry $(N_+ - N_-)/(N_+ + N_-)$ between the counting rates $N_+$ in the positron telescopes in the direction of $P$ and $N_-$ in the opposite direction. By sweeping the frequency $\omega_{hf}$ or by varying the applied magnetic field $B_{app}$, and hence the energy difference $\Delta E$ we may determine the resonance conditions and thus obtain the spectrum of Zeeman frequencies $\omega_j$.

Resonance measurements have been carried out on the triplet state of muonium atoms $\text{Mu} = (\mu^+e^-)$\footnote{The shortest relaxation times or spin precession periods that can be measured in the collective mode are determined by the time interval $\Delta t_0$ over which the muons coming from the same pulse enter the sample. $\Delta t_0$ may be made shorter than $\Delta t_1$ by letting the muons pass through time-dependent magnetic fields arranged in such a way that the paths of muons generated at the beginning of the pulse are longer than those generated towards the end. The compression of a pulse of muons with $v/\gamma = 0.27c$ by $1 \cdot 10^{-7}$ s requires a path difference of $8 \, \text{m.}$}. In this state electron and muon spins are parallel, so that the system behaves as a spin-one particle with gyromagnetic ratio $(\gamma_e - \gamma_\mu)/2$, where $\gamma_e = 1760.8 \cdot 10^8 \, \text{s}^{-1} \cdot \text{T}^{-1}$ denotes the electron gyromagnetic ratio. The physical situation may be described qualitatively by saying that the torque exerted on the spin momentum is not determined by the magnetic moment of the muon but by the much larger magnetic moment of the electron. It is therefore not particularly difficult to achieve the hf field strength necessary to reverse the muon spins within times of the order of magnitude of the muon lifetime.

From the practical point of view the situation is quite different if we attempt to study "naked" $\mu^+$ or negative muonium ions $\text{Mu}^- = (\mu^-e^-)$ by means of the resonance technique. Then the hf field has to act directly on the magnetic moments of the muons. The hf field strengths required for the spin reversal are now larger by a factor $10^2$ than in the triplet $\text{Mu}$ case. Under otherwise equal conditions (same frequency and sample volume) this means that the hf power would have to be increased by a factor of $10^4$. The problem of
supplying and cooling away the necessary power is very much reduced if
the muon beam is pulsed with a period $\Delta t_2 \gg \tau_{\mu}$, since then the high-
frequency field needs to be applied only as long as the sample still con-
tains a detectable number of muons. On the other hand, there is no limita-
tion to the number of muons that may be present in the sample at any
one time. This means that in the resonance technique best use is made
of the muons and of the hf energy if the muons are produced in intense
pulses of length $\Delta t_1 \leq \tau_{\mu}$ and if both the muon and the hf pulses possess
long periods $\Delta t_2 \gg \tau_{\mu}$. Similar considerations hold for the effects of
laser light on $\mu$SR, which are of interest for the study of muonium atoms
in non-metallic materials.

A $\mu$SR resonance facility satisfying the above requirements has re-
cently been installed at BOOM [2]. A radio-frequency field with a peak
power of about 20 kW is applied during $10^4$ s after each muon beam burst.
As a first application of the facility the $\mu^+$ resonance in $\text{H}_2\text{O}$ was de-
tected by irradiating at a fixed frequency of $20 \cdot 10^6$ Hz and sweeping the
magnetic fields. It should be emphasized, however, that in the resonance
technique the use of pulsed muon beams is not essential but merely a
matter of economy. A letter of intent for $\mu^+$ spin resonance experiments
at SIN (where $\Delta t_1 \ll \Delta t_2 < \tau_{\mu}$) has been submitted by a Stuttgart-Heidel-
berg collaboration in November 1979.

All $\mu$SR techniques require magnetic interactions between the magnetic
moments of the implanted muons and the host material. As indicated in Table I,
the possible interactions may be divided into five major groups. Table I
lists examples of the materials that might be investigated through the
various interactions, of the physical quantities that may be observed, and
of the information that may be gained from the measurements. The multitude
of existing possibilities forbids a systematic discussion in this paper.
We have to confine ourselves to a few remarks on each of the five inter-
actions.

I) The dipolar interaction between the nuclear magnetic moments
and the muon magnetic moments is rather weak. Longitudinal or transverse
relaxation rates that can be investigated with good precision require a
high abundance of nuclei with large magnetic moments. Fortunately a large
number of elements satisfy these requirements. Quite detailed information
on the behaviour of muons may be obtained through the dependence of the trans-
verse rate on the crystallographic direction of the applied field $B_{\text{appl}}$. A
particularly interesting possibility is the use as analytical tool for de-
tecting and characterizing intrinsic or extrinsic (impurity) defects cap-
able of trapping positive muons [19,20].

II) The interaction of magnetic moments of d- and f- electrons with
those of muons is the strongest of the interactions considered in Table I.
Its consequences for $\mu$SR measurements are strongly dependent not only on the
magnetic structure of the material (ferromagnetic, antiferromagnetic, ferri-
magnetic, strongly paramagnetic ions in diamagnetic or weakly paramagnetic
hosts) but also on its crystallographic structure and the symmetry of the
sites occupied by the $\mu^+$ [8,21,22]. In many cases the strength of the inter-
action allows us to carry out measurements on internal fields, muon diffu-
sion, and muon trapping over much wider temperatures ranges than is feasible
with other techniques [21,23].

III) From the theoretical point of view the contact interaction between
conduction electrons and muon spins is just a special case of the interaction
between electron and muon magnetic moments. The practical manifestations,
however, are quite different from those of II). Because of the Pauli principle
<table>
<thead>
<tr>
<th>Magnetic Interaction</th>
<th>Examples</th>
<th>Observations</th>
<th>Information to be Gained</th>
</tr>
</thead>
<tbody>
<tr>
<td>nuclear magnetic moments</td>
<td>Li, ..., Cs, Be, Al, In, Cu Co, As, Bi, V, Nb, Ta</td>
<td>longitudinal or transverse relaxation</td>
<td>( \mu^+ ) location, lattice distortion around ( \mu^+ ), ( \mu^+ ) diffusion, study of defects and trace impurities</td>
</tr>
<tr>
<td>magnetic moments of d- or f- electrons</td>
<td>Fe, Ni, Co, Gd, Cr, Mn, Dy, Fe\textsubscript{3}O\textsubscript{4}</td>
<td>( B_\mu = \omega_\mu / \gamma_\mu ), longitudinal or transverse relaxation</td>
<td>internal magnetic fields, ( \mu^+ ) diffusion over wide T range, trapping at defects</td>
</tr>
<tr>
<td>contact interaction with conduction electrons</td>
<td>normal metals, ferromagnets</td>
<td>Knight-shift ( B_\mu = \omega_\mu / \gamma_\mu )</td>
<td>local density of states, conduction-electron spin polarization, trapping at defects</td>
</tr>
<tr>
<td>orbital magnetism of conduction electrons</td>
<td>Type I + II superconductors</td>
<td>( B_\mu = \omega_\mu / \gamma_\mu ), transverse relaxation</td>
<td>fast low-temperature diffusion of ( \mu^+ ), ( B_\mu ) distribution, flux-line flow</td>
</tr>
<tr>
<td>( \mu = (\mu^+e^-) ): hyperfine interaction</td>
<td>semiconductors, insulators, gases</td>
<td>( j_\mu ), longitudinal or transverse relaxation</td>
<td>( \mu ) chemistry, reaction rates, simulation of H behaviour, isotope effects</td>
</tr>
</tbody>
</table>

Table I: Magnetic Interaction in \( \mu^+ \)SR Applications
the spin polarizability and hence the Knight shift in normal metals is very small. By means of a special stroboscopic μSR technique [24] in many "normal" metals the muon Knight shift may be determined more accurately than is possible for the hydrogen Knight shift. In this way interesting information on the electronic structure of point-charge impurities in normal metals has been obtained [25]. In ferromagnetic metals there exists a spontaneous spin polarization of the conduction electrons which gives rise to the so-called Fermi field acting on the muon magnetic moments. Whereas neutron diffraction can give information on the spin polarization of conduction electrons in ideal ferromagnetic metals [26], the information obtainable from μSR measurements pertains to the presence of positive point charges and is thus complementary to the neutron-diffraction information.

IV) The interaction between the muon magnetic moments and the orbital magnetism of conduction electrons in superconductors may be used to study the magnetic field distribution in magnetically inhomogeneous superconducting states, i.e. in the intermediate or in the mixed state [27]. Particularly interesting possibilities exist in the mixed state of Type-II superconductors, where the very fast low-temperature diffusion of muons in pure Type-II superconductors may be studied [28,29] and where information on the flow of flux lines may be obtained.

V) In materials in which muonium atoms $\text{Mu} = (\mu^+e^-)$ are formed the hyperfine interaction between the muon spin and that of the bound electron is usually the most important magnetic interaction of the muons. This interaction may be employed in a wide variety of ways in order to obtain information on the physics and chemistry of the host material. The study of the magnetic properties of Mu atoms has given rise to a field of its own, muonium chemistry [9].

4. Lattice Steering ("Channelling")

The basic ideas of the lattice-steering technique, which is applicable to both reactions (1) and (2), are indicated in Fig. 5. For simplicity we shall first describe the technique as based on reaction (1), briefly called $\pi^-/\mu^+$ lattice steering or channelling, and subsequently discuss the differences between $\pi^-/\mu^+$ and $\mu^+e^-$ lattice steering.

Suppose that the implanted positive pions are located on interstitial sites in an otherwise perfect crystal. Then the monoenergetic positive muons emitted into one of the open "channels" going out from the $\pi^-$ sites along the low-index crystallographic directions undergo sequences of correlated repulsive small-angle collisions with the nuclei bordering the channel. They keep the direction of the $\mu^+$ propagation, on the average, parallel to the channel direction (Fig. 5, top). This "lattice steering" effect leads to a flux-enhancement in the channel directions compared to random directions, in which the $\mu^+$ are deflected by large-angle scattering.

The crystallographic directions in which the flux enhancement may be observed in position-sensitive detectors placed sufficiently far from the sample depend on the interstitial sites on which the emitting particles are located, so that from observations in different crystallographic directions these sites may be deduced. Moreover, from the precise shape of the enhancement profile one may draw conclusions on the spatial distribution of the emission probability, i.e. on the quantum-mechanical and thermal vibration amplitudes of the $\pi^-$ in their interstices.
In those directions in which the emitted particles encounter an entire row of atoms, the phenomenon of blocking takes place. As indicated in Fig. 5, this results in a diminution of the muon flux relative to that observed in a random direction. If the $\mu^+$ are located on substitutional sites because they have been trapped in vacancies, the blocking phenomenon occurs in all major crystallographic directions. Thus vacancies (and, provided observations in a sufficient number of crystallographic directions are available, also divacancies, etc.) may be monitored fairly directly by this "decoration" technique.

The angular range around a crystallographic direction in which the information on the location of the probes is contained may be estimated from Lindhard's critical angle

$$\psi_{cr} = 2(Z e^2 / 4\pi \varepsilon_0 p v d)^{1/2} ,$$

where $Z$ denotes the atomic number of the host material, $e$ the elementary electric charge, $\varepsilon_0 = 8.85 \times 10^{-12}$ A s V$^{-1}$ m$^{-1}$, $p$ and $v$ linear momentum and velocity of the particles undergoing channelling (assumed to have unit charge), and $d$ the interatomic distance in the channelling direction. For positive muons emitted by pions at rest (11) becomes

$$\psi_{cr} = 9 (Z / d_{Na})^{1/2} ,$$

where $\psi_{cr}$ is measured in angular minutes. This means that for medium heavy elements the solid angle within which the lattice-steering phenomenon takes place is comparable with that under which the full Moon is seen from the Earth. The information on the pion location is thus carried by a very small fraction of the muons that are emitted, so that lattice-steering experiments, informative and easy to interpret as they are, require large pion fluxes.

Closer inspection shows that the fraction of "useful" $\mu^+$ is even smaller than might appear from the preceding estimate. The small-angle scattering of channelled particles by conduction and valence electrons leads to an increase of $\langle \Delta \psi^2 \rangle$ proportional to the distance travelled by the channelled particles ($\Delta \psi = \text{deviation of the propagation direction from the channel axis}$). It is thus necessary to limit the depth from which the particles are emitted. Since in the case of $\pi^+ / \mu^+$ lattice steering the channelled particles are originally
Fig. 6: Flux enhancement of positive muons emitted in <100> directions from therma-

dized \( \pi^+ \) in a tantalum single crystal at

200 K \((x,y = \text{coordinates on position-}

sensitive detector)\).

The first \( \pi^+/\mu^+ \) lattice-

steering experiments have recently been performed on tan-

talum single crystals by Maier et al. \cite{15,32,33}. Fig. 5 indi-

cates schematically the experi-

mental arrangement employed.

Because of the short pion life-

time linear momentum and ve-

locity of the \( \pi^+ \) were chosen

close to the highest values

admissible from the point of

view of nuclear reaction cross-

sections \(\text{cf. footnote}\).

By means of a degrader foil the pions were

slowed down in front of the

sample so that they were deposited close to the specimen surface. Fig. 6 shows

two flux-enhancement patterns A and B obtained in these experiments \cite{33}.

Two patterns rather than just one were observed because the sample was rotated

with respect to the beam in order to eliminate the characteristics of the po-

sition-sensitive detector. The separation of the two peaks agrees with the

angle of rotation. The width of the flux-enhancement peak corresponds to the

theoretical expectations.

Experiments on \( \mu^+/e^+ \) lattice steering, in which the muons serve as the

probe and the \( e^+ \) undergo channelling, present several additional problems: (i)

Generally speaking, the available \( \mu^+ \) fluxes are smaller than the \( \pi^+ \) fluxes.

(ii) The quantity \( p\cdot v \), which according to (11) determines the critical angle

for channelling, is smaller for \( e^+ \) from (2) than for \( \mu^+ \) from (1)\(^3\). Hence in

\( \mu^+/e^+ \) lattice steering the critical angles are smaller than those of \( \pi^+/\mu^+ \)

channelling and the demands on the perfection of the crystals used as samples

accordingly higher. In addition, the fraction of the particles carrying the

lattice-steering information becomes even smaller than in \( \pi^+/\mu^+ \) experiments.

(iii) Since the \( e^+ \) emitted by \( \mu^+ \) are not monoenergetic, the depth limitation

necessary to keep the straggling effect below a critical level cannot be

achieved by energy selection but must be done by limiting the depth at which

the \( \mu^+ \) are implanted.

An experimental arrangement for a \( \mu^+/e^+ \) lattice–steering experiment is

shown in Fig. 5 (bottom). It makes use of the fact that from \( \pi^+ \) stopped near

the surface of the proton target a well-collimated beam of so-called surface

muons with kinetic energies \( E_{\text{kin}} \lesssim 4.12 \text{ MeV} \) ("Arizona beam") may be obtained.

The experimental difficulties described above have been overcome in recent

experiments of Maier et al. on Au \cite{34}, in which \( \mu^+/e^+ \) lattice steering was

demonstrated for the first time. In these experiments the surface muons were

\(^3\)At the upper end of the \( e^+ \) spectrum we have \( p_{e^+}v_e = 53 \text{ MeV} \), which is to be

compared with \( p_{\mu^+}v_\mu = 8.0 \text{ MeV} \).
Fig. 7: Blocking of positrons emitted in a \langle 100 \rangle direction from thermalized \mu^+ in gold single crystal at 100 K (averaged over angle around \langle 100 \rangle).

lattice-steering method very promising for the future. It does, however, require single crystals of good quality and sufficient size. The know-how of growing such crystals is available for a large number of materials. We expect that in the years to come further progress in crystal growing and in the application of the lattice-steering technique will go hand in hand.


Let us return to Fig. 1, where the main uses of a future combined spallation-meson-production facility are indicated. The charged pions produced in a thin target of light nuclei may be utilized either directly for biomedical applications and for elementary-particle, nuclear, atomic, and solid-state physics, or for the production of muon beams.

Muon beams may be obtained either from pions decaying at rest or from pions decaying in flight. In the first case the muons are emitted from pions that have been stopped near the surface of the proton target. The \mu^+ beams produced in this way are known as "surface-muon beams" or (for historical reasons [35]) as "Arizona beams". Such beams possess the attractive feature that they are virtually 100 % longitudinally polarized. The kinetic energy of the muons, \( E_{\mu^+ \text{kin}} \), varies between zero and 4.12 MeV. This means that the penetration of the muons is small and that low-density targets (gases and vapors!) as well as very thin samples may be investigated. Furthermore, because of the low \( E_{\mu^+ \text{kin}} \) values Arizona beams are very useful if one wishes to implant the muons only near the sample surface. Examples for this are the \mu^+ /e^+ lattice-steering experiments discussed in Sect. 4 or the investigation of near-surface radiation damage produced by ion or electron bombardment.

A definite drawback of the surface muons is that because of their small linear momenta (\( p_{\mu^+} < 30 \text{ MeV/c} \)) they are easily deflected by transverse magnetic fields. This makes it difficult to carry out transverse \( \mu \text{SR} \) measurements in
applied fields exceeding, say, 0.1 T. This weakness can be remedied by means of a spin rotator, a combination of magnetic and strong electric fields that rotates the muon spin by 90° with respect to the muon momentum and thus gives us transversely polarized beams. The geometry of transverse μSR experiments can be arranged in such a way that beams with transverse polarization are not deflected by the applied magnetic field. A low momentum beam with transverse polarization would also be of great help for μ⁺/e⁺ lattice-steering experiments, since then full advantage could be taken of the fact that in (2) the asymmetry of the positron emission depends on the positron energy and that the emission of low-energy positrons opposite to the muon spin direction is suppressed. In all future Arizona-beam installations provisions should be made for setting up a spin rotator, which necessarily is several meters long.

In order to obtain polarized muons from pions in flight one needs a so-called muon channel. In the case of SIN and BOOM this consists of a superconducting solenoid, in which the spiralling path of the pions is so long that virtually all of them decay in the channel. In this way high-momentum muon beams with very little pion and positron contamination have been achieved. However, because of the so-called kinematic depolarization [36] the polarization of high-momentum beams is necessarily smaller than that of Arizona beams and amounts in practice to not more than 0.75 - 0.80.

The kinematic depolarization is due to the fact that a muon beam extracted in the laboratory system in, say, backward direction from the flight direction of the pions contains a certain fraction of muons that have been emitted in the forward direction as seen from the pion rest system and which hence in the laboratory system possess the "wrong" spin direction.

So far the only application of positive pions outside of nuclear and elementary-particle physics is the μ⁺/μ⁺ lattice steering described in Sect. 4. In this application the μ⁺ should be implanted into the samples at energies as low as possible in order to keep the path of the emerging μ⁺ inside the samples short. On the other hand, in order not to lose too many pions by decay the pion velocity and hence the pion energy should be as high as possible. The distance over which the fraction e⁻ of pions with velocity c/4 have not yet decayed is only 2 m. In the experiments on Ta performed at SIN [15,33] a flux of 2·10⁸μ⁺/s at a momentum p⁺ = 150 MeV/c was available. This resulted in 8 counts per second in the useful ranges of direction and energy. Under these conditions, which by present standards must be considered quite favourable, about 5 hours were required in order to obtain the information contained in Fig. 6.

The two important time constants in the physics of pions and muons are the lifetimes \( \tau_{\text{μ⁺}} = 2.6 \cdot 10^{-8} \) s and \( \tau_{\text{π⁺}} = 2.2 \cdot 10^{-8} \) s. From this it follows that interesting time structures are \( \Delta t_1 \ll \tau_{\text{μ⁺}} \ll \Delta t_2 \) for pion beams and \( \Delta t_1 \ll \tau_{\text{μ⁺}} \ll \Delta t_2 \) for muon beams. The time-structure requirements for pulsed μSR measurements have been discussed in Sect. 3. In the following we add some remarks on the use of beams with the above time structures for the study of pion and muon trapping at crystal defects by means of time-differential lattice steering.

In order to reach defects at which they may be trapped, thermalized pions and muons must diffuse a certain distance. An estimate of the time it takes till the fraction e⁻ is trapped (neglecting the possibility of detrapping) by an atomic concentration C of defects with capture radius \( r_0 \) is given by

\[
t = \frac{V_A}{4\pi r_0 C D^+},
\]

(13)
where $D^+$ denotes the diffusivity of the positively charged probes and $V_A$ the atomic volume. The majority of the probes that decay at times short compared to (13) will do so at interstitial sites in a perfect lattice environment and will give rise to the corresponding flux-enhancement pattern, whereas those which decay at times longer than (13) are likely to have been trapped. The flux-enhancement and blocking patterns should then be those of the trapping sites.

Using pulses with lengths $\Delta t_1$ satisfying one of the above conditions as collective entry signals we may record the lattice-steering patterns as a function of the time the probes have spent in the sample. If the trapping time $t$ is comparable with the lifetime of the probes this allows us to obtain in the same run the patterns both of the tree and the trapped probes. From this the trapping time $t$ and, with the help of (13), the quantity $r,C D^+$ may be derived. For a fixed defect concentration it will thus be possible to deduce the temperature dependence of $D^+$. Alternatively, if $D^+$ is known, one may determine $r,C$ experimentally.

The possibility just described is particularly attractive in view of the fact that the ratio of the muon to the pion lifetime is 85 and that the diffusion coefficients of the two probes are not expected to be too different. The combination of $\pi^+ / \mu^+$ and $\mu^+ / e^+$ lattice steering should enable us to follow variations of $D^+$ or $C$ over almost four powers of ten.

The normal operation conditions of SIN are $\Delta t_1 = 1 \cdot 10^{-6}s$ and $\Delta t_2 = 2 \cdot 10^{-8}s$. This means that they satisfy $\Delta t_1 << \tau_{\pi^+}$ but not $\tau_{\pi^+} << \Delta t_2$. However, at the cost of reducing the average pion flux to one third it is possible to operate the SIN accelerator at $\Delta t_2 = 5.9 \cdot 10^{-8}s$, corresponding to $t_2 / \tau_{\pi^+} = 2.3$. Under these conditions the time-differential $\pi^+ / \mu^+$ lattice-steering experiment described above appears feasible, although the time structure is not ideal and a single run is expected to take more than 24 hours. After the increase of the $\pi^+$ flux foreseen in a few years from now [37] it should be possible to perform time-differential $\pi^+ / \mu^+$ experiments routinely at SIN.

With the present facilities at SIN a non-time-differential $\mu^+ / e^+$ lattice-steering experiment would take about a factor of ten longer than the corresponding $\pi^+ / \mu^+$ experiment if the same statistical accuracy is to be achieved. This means that it goes beyond what can be routinely done now but that routine experiments should become feasible after the SIN extension program is completed.

To the writer's knowledge, at present there is nowhere a realistic possibility to perform time-differential $\mu^+ / e^+$ lattice-steering experiments. However, an accelerator with an average 1.1 GeV proton current of 5 mA and time structure $\Delta t_1 = 1.5 \cdot 10^{-7}s$ and $\Delta t_2 = 2 \cdot 10^{-2}s$ as it is under discussion for the German neutron spallation source would provide close-to-ideal conditions for setting up such experiments. This would allow us to reach the "ultimate goal" of the application of positive pions and muons to solid-state physics, namely the possibility to perform $\mu$SR and time-differential $\mu^+ / e^+$ lattice-steering experiments on the same samples and thus to obtain experimental information in unprecedented completeness and detail on crystal defects capable of trapping muons.

Fig. 8 summarizes our discussions on flux strength and time-structure.

4) In Fig. 4 the interrupted lines mean that the time structure inside the lines is advantageous but not necessary.
<table>
<thead>
<tr>
<th>Time structure</th>
<th>Direct current</th>
<th>$\Delta t_1 \ll \tau_\mu \ll \Delta t_2$</th>
<th>$\Delta t_1 \ll \tau_\pi \ll \Delta t_2$</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Individual time-differential $\mu$SR (trans., long.)</td>
<td></td>
<td></td>
<td></td>
<td>Classical technique, only marginal improvements possible</td>
</tr>
<tr>
<td>Non-frequency $\mu$SR, coupling through $\mu$spin (Mu)</td>
<td></td>
<td></td>
<td></td>
<td>Time structure desirable but not necessary</td>
</tr>
<tr>
<td>$\pi^+$/e$^+$ lattice steering</td>
<td></td>
<td></td>
<td></td>
<td>Demonstrated at SIN</td>
</tr>
<tr>
<td>High-frequency $\mu$SR, coupling through $\mu$spin (long)</td>
<td></td>
<td></td>
<td></td>
<td>Feasible at SIN (5-HD letter of intent) and BOOM</td>
</tr>
<tr>
<td>Collective time-differential $\mu$SR (trans., long.)</td>
<td></td>
<td></td>
<td></td>
<td>Principal technique at BOOM (Japan)</td>
</tr>
<tr>
<td>Time-differential $\pi^+$/e$^+$ lattice steering</td>
<td></td>
<td></td>
<td></td>
<td>Feasible at SIN, requires larger $\pi^+$ flux</td>
</tr>
<tr>
<td>$\mu^+$/e$^+$ lattice steering</td>
<td></td>
<td></td>
<td></td>
<td>Demonstrated at SIN, larger flux required</td>
</tr>
<tr>
<td>Time-differential $\mu^+$/e$^+$ lattice steering</td>
<td></td>
<td></td>
<td></td>
<td>Much larger fluxes than available required</td>
</tr>
<tr>
<td>T-$d\mu^+$/e$^+$ lattice steering $\mu$SR on same sample</td>
<td></td>
<td></td>
<td></td>
<td>&quot;ULTIMATE&quot; AIM</td>
</tr>
</tbody>
</table>

Fig. 8: Time-structure and flux requirements for $\mu$SR and lattice steering

The "classical" individual-mode $\mu$SR makes best use of the muons produced if a direct current of muons is available. (Direct current in this context means that $\Delta t_2 \ll \tau_\mu$, so that from the point of view of muon physics SIN is a direct current facility.) As explained in Sect. 3 the count rates that can be achieved in this mode are sharply limited, so that a further increase in the muon flux will only give marginal improvements (e.g., narrower momentum range of the beams, higher polarization of high-momentum beams, possibility to use smaller samples). All other techniques, however, will gain from higher muon and pion fluxes, most strikingly so the lattice-steering experiments in which the count-rate is strictly and without limitation proportional to the particle flux.

The various techniques have been arranged in Fig. 8 roughly in the sequence of increasing flux required. Only the first two techniques may be said to be in good shape with the present facilities. All other techniques will substantially profit from future increases in intensity or, as the time-differential $\mu^+$/e$^+$ experiments, will only become possible when fluxes at least one order of magnitude higher than those of to-day will be available.

5) An exception is the stroboscopic $\mu$SR technique [24] mentioned in Sect. 3, which does make use of the finiteness of $\Delta t_2$. 
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