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MUON SPIN ROTATION

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Abstract: A short survey on the basic principles of the technique of muon spin rotation is given. The use of positive muons as probes in solids is illustrated by discussing the experimental information about the site and diffusion behavior of muons in metals.

1. Introduction

Within two days in January 1957 three manuscripts were submitted to the Physical Review, each one describing a separate novel experiment confirming the violation of parity in weak interactions, as suggested by Lee and Yang. The first by Wu et al. [1] reported on the experiments on β^- decay of polarized nuclei. In the second paper [2] Garwin, Lederman and Weinrich, by inventing the muon spin rotation technique, showed that parity is violated in both the

$$\pi^+ \to \mu^+ + \nu_\mu \qquad (1)$$

and the

$$\mu^+ \to e^+ + \nu_e + \bar{\nu}_\mu \qquad (2)$$

decays. The third paper [3] by Friedman and Telegdi introduced the notion of muonium and noted the spin-depolarizing effects arising from its hyperfine interaction and the rapid precession of its total magnetic moment in the external magnetic field. These last two papers set the stage for μSR (muon spin rotation, resonance, research, etc.) and established the principles on which most applications in solid-state physics and chemistry are based.

It is remarkable that in the paper by Garwin et al. appeared already the speculation "... it seems possible that polarized positive and negative muons will become a powerful tool for exploring magnetic fields in nuclei, atoms and interatomic regions". Applications to solid state physics were, however, rare until the construction of the meson factories about ten years ago. At present μSR is done at SIN (Villigen, Switzerland), TRIUMF (Vancouver), LAMPF (Los Alamos), CERN, DUBNA, KEK (Japan), BNL (Brookhaven), NIKHEF (Amsterdam), and SNS, the spallation neutron source at the Rutherford-Appleton Laboratory in England. Although the method is limited by the few existing muon beams, μSR activity has increased dramatically. The progress and status of μSR research can best be found by consulting the proceedings of the three μSR conferences [4]. Detailed information is given in Ref. [5] and [6].

2. Principles of the μSR technique

Muons are weakly and electromagnetically interacting particles which may be considered as heavy electrons. Some properties are

mass	$m_\mu = 105.659\ \mathrm{MeV}/c^2 = 206.768\ m_e$
	$= 0.112610\ m_p$
spin	$I = 1/2$
lifetime	$\tau_\mu = 2.19709\ \mu s$
magnetic moment	$\mu_\mu = 3.18335\ \mu_p$
gyromagnetic ratio	$\gamma_\mu/2\pi = 135.537\ \mathrm{MHz/T}$

Figure 1 shows in the upper part the pion decay (1) and the muon decay (2) at the maximum possible energy. When the muon is produced it is fully longitudinally polarized in the pion rest-frame. When it decays it follows from conservation laws that the positron is emitted in the muon spin direction. The angular correlation pattern for this case is shown on the left of the figure. If all the positrons in the decay are detected the angular correlation function is

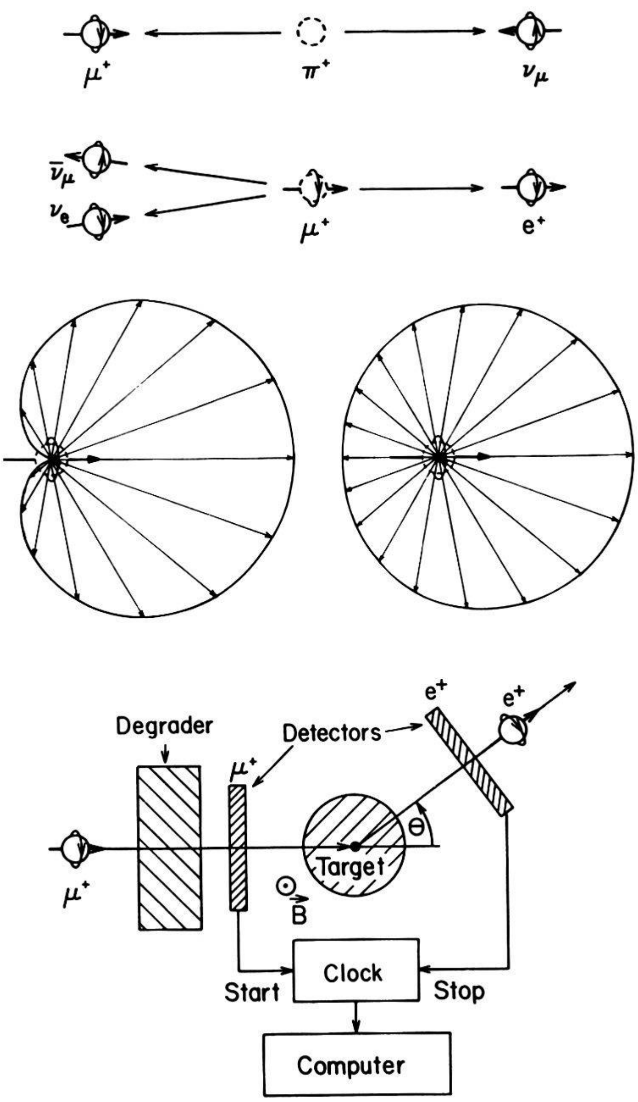

Figure 1

The upper part of the figure illustrates the π^+ and μ^+ decay. In the center, the angular distribution patterns of the decay positrons with respect to the μ^+ spin are shown for the case that only positrons at the maximum energy are registered (left) and for the case that all the positrons are detected (right). In the lower part a simplified diagram of a μSR apparatus is shown.

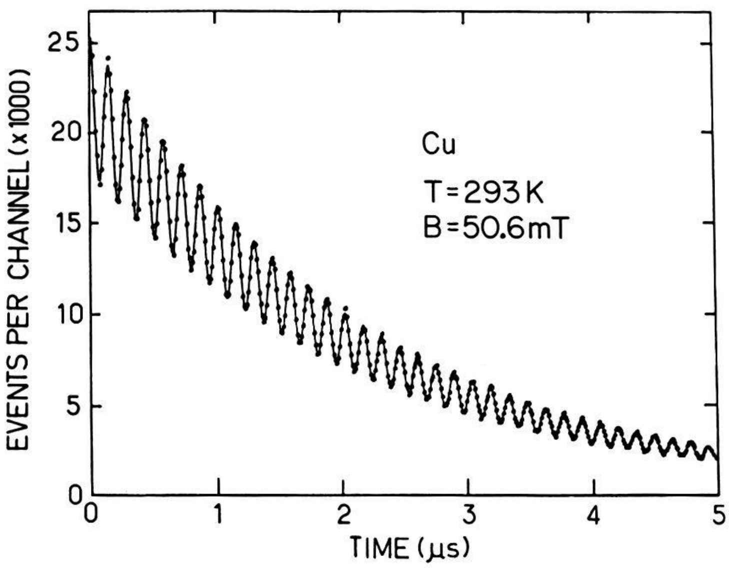

Figure 2

μSR spectrum of Cu in a transverse field of 50.6 mT. The oscillations superimposed on the exponential muon decay are due to the muon spin precession. The line plotted corresponds to the least square fit to Eq. (3).

$$N(\theta, t) = N_0 \exp(-t/\tau_\mu) \, [1 + A\, P(t)\cos(\theta + \omega_L t)], \tag{3}$$

where θ is the angle between the muon spin and the direction of the decay positron, giving the angular correlation pattern shown on the right. For negative muons the cosine term is negative. If an external magnetic field is applied perpendicular to the incoming muon spin direction, the spin will precess with the Larmor frequency

$$\omega_L = \gamma_\mu B. \tag{4}$$

The three quantities obtained from a μSR experiment are

1) the anisotropy A. If all muons are in the same state, $A = 1/3$.
2) the Larmor frequency ω_L which is a direct measure of the field B_μ at the muon.
3) the relaxation function $P(t)$ which in a transverse field measures field inhomogeneities e.g. due to nuclear dipolar fields. The form of $P(t)$ (exponential or Gaussian decay) gives information about the motion of the muons.

As an example Fig. 2 shows the μSR spectrum of polycrystalline copper measured at room temperature in an external transverse field of 50.6 mT. The observed frequency is $\omega_L = \gamma B$, the amplitude after corrections for solid angle effects is $A = 1/3$. No relaxation is observed, i.e. $P(t) = 1$.

3. Determination of the muon site and lattice relaxation

In non-magnetic metals the muon precession is in general disturbed by dipolar fields which are produced by the magnetic moments of neighboring nuclei. These nuclear spins are not ordered and thus produce at the interstitial sites variable dipolar fields of the order of 10^{-4} T. The Larmor precession of stationary muons in a transverse magnetic field of about 0.4 T is slightly disturbed by these additional randomly oriented dipolar fields and muons at sites with different fields precess at different frequencies. This results in an effective relaxation function which is given by

$$P(t) = \exp(-\sigma^2 t^2). \quad (5)$$

The quantity σ^2 is a measure of the width of the dipolar field distribution and depends on the site of the muon, its distance to the neighboring nuclei as well as on the orientation of the external field with respect to the crystal axes. This enabled the determination of the muon site in Cu. It was found that the μ^+ is stationary at the octahedral interstitial sites below 80 K and that the positions of the nearest Cu neighbors are slightly distorted. This lattice relaxation around the impurity lowers the energy. The muon is self-trapped and forms a small polaron. The non-vanishing electric field gradient at the nearby Cu nuclei has also been measured by observing the influence of the quadrupolar interaction of the Cu spins on the muon precession at lower external field strengths.

These results of μSR experiments on Cu and other metals have provoked considerable theoretical efforts to determine the electronic structure of protons or muons in metals from first principles. Various approaches have been developed to calculate potentials for hydrogen in metals. Most of them are based on the self-consistent density-functional formalism. Some progress has been achieved but there still remain uncertainties and open questions connected with this simplest impurity problem in metal physics. As an example, Fig. 3 shows a potential for a muon in Cu calculated for a rigid lattice. To account for the experimental results the influence of lattice relaxation should be considered, but the displacement of the neighboring ions depends on the extent of the muon wave function which depends on the potential. The latter is in turn a function of the lattice relaxation and a full calculation of the ground-state of the impurity system thus requires a self-consistent treatment. No such calculations have been performed yet.

4. Diffusion

At temperatures above 80 K the muons start to diffuse in the Cu lattice. Thereby the different dipolar field contributions are averaged out which results in a reduction of the relaxation rate and in the change of the form of the relaxation function to an exponential

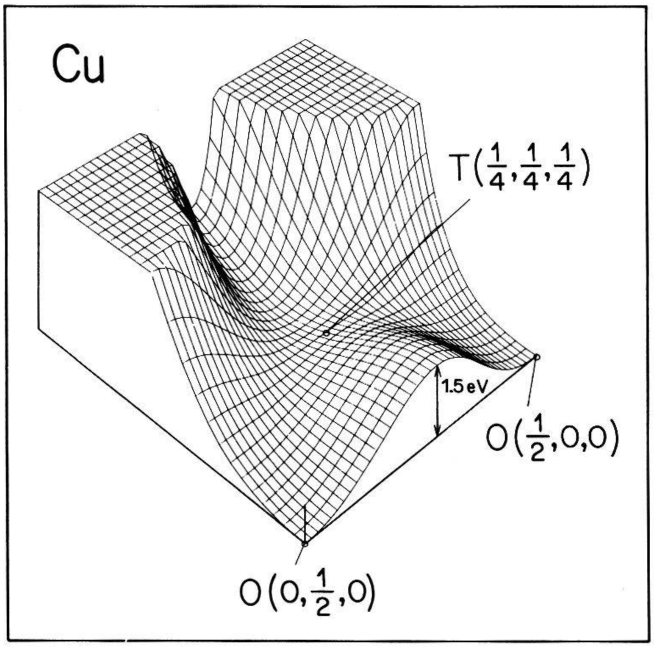

Figure 3

Calculated muon potential for Cu in a plane through two neighboring octahedral (O) and one tetrahedral (T) sites. The energy difference E(T)-E(O) is about 0.6 eV.

$$P(t) = \exp(-\lambda t). \tag{6}$$

In Fig. 4 the change of the relaxation rate in Cu as a function of temperature is shown. Above room temperature the diffusion is already so fast that within the experimental limits due to the finite muon lifetime no depolarization is observed.

The change-over from the stationary relaxation function (5) to the motional averaged one (6) is usually described by

$$P(t) = \exp\{-2\sigma^2\tau_c^2[\exp(-t/\tau_c) - 1 + t/\tau_c]\}. \tag{7}$$

The correlation time τ_c is directly related to the diffusion coefficient D. A fit of the time spectra to this relaxation function then yields D as a function of T. In Cu, where $D = a_o^2/12\ \tau_c$, it was found that D follows essentially an Arrhenius type temperature behavior $D = 4.4 \times 10^{-13} \exp[-48\ \mathrm{meV}/kT]\ \mathrm{m^2 s^{-1}}$. The activation energy of 48 meV is very low compared to the 400 meV measured for hydrogen diffusion in Cu. This can be explained qualitatively by phonon-assisted tunneling between ground states for which the isotope effect is rather large. A quantitative understanding is still missing.

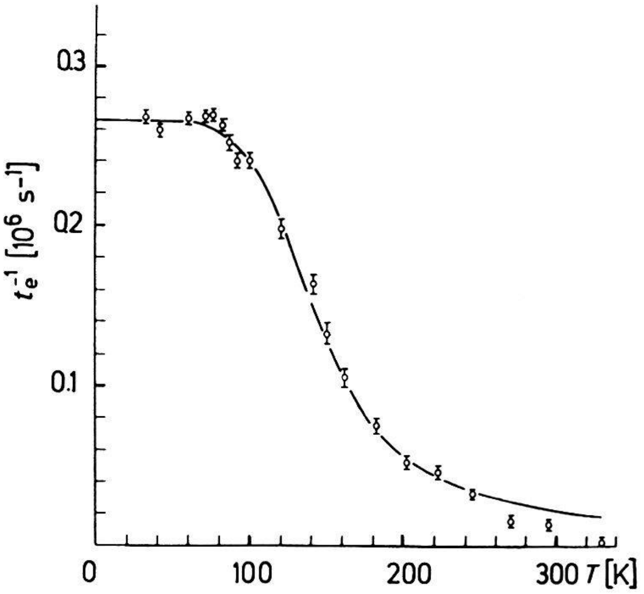

Figure 4

Reciprocal of time t_e in which the precession signal in Cu decays by a factor e^{-1} [$P(t_e) = {}^1/e$]. The solid line corresponds to the least square fit to Eq. (7) where τ_c follows an Arrhenius type behavior.

In recent experiments a decrease of the relaxation rate in Cu below 20 K was observed. This is an indication for the onset of coherent diffusion which also occurs for muons in other metals. In pure Al samples, e.g., no depolarization is observed down to 30 mK. Information about the diffusion can, however, be obtained indirectly from measurements on samples which are electron-irradiated or doped with impurities. Coherent diffusion is very sensitive to deviations from the periodic lattice potential produced by the strain fields. It is found that muons are trapped near vacancies or impurities but are released at elevated temperatures. The dependence of the relaxation rate on the impurity content of the sample then allows one to determine the diffusivity in the pure sample. These μSR experiments at low temperature have provoked a renewed interest in the theory of quantum diffusion of light particles. Kondo [7] has pointed out that non-adiabatic effects in the electron-muon interaction have to be considered but this point is still controversial [8].

5. Other techniques and applications

The μSR technique introduced above is called the individual time-differential μSR method in transverse fields since, as indicated in Fig. 1., the decay positrons from individual muons stopped in the sample are recorded one after the other. Other methods which may be favorable to study certain effects involve longitudinal or zero field methods. If pulsed muon beams are available, collective μSR and resonance methods can be used.

The use of muons as probes in solids is still growing. Among the various interesting applications the measurements of hyperfine fields in magnetically ordered materials, Knight shift experiments, μSR studies in spin glasses, and the investigation of the muonium states in semiconductors and insulators, should be mentioned.

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