First μ^+ SR studies on thin films with a new beam of low energy positive muons at energies below 20 keV

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At the Paul Scherrer Institute slow positive muons (μ^+) with nearly 100% polarization and an energy of about 10 eV are generated by moderation of an intense secondary beam of surface muons in an appropriate condensed gas layer. These epithermal muons are used as a source of a tertiary beam of tunable energy between 10 eV and 20 keV. The range of these muons in solids is up to 100 nm which allows the extension of the μ^+ SR techniques (muon spin rotation, relaxation, resonance) to the study of thin films. A basic requirement for the proper interpretation of μ^+ SR results on thin films and multi-layers is the knowledge of the depth distribution of muons in matter. To date, no data are available concerning this topic. Therefore, we investigated the penetration depth of μ^+ with energies between 8 keV and 16 keV in Cu/SiO₂ samples. The experimental data are in agreement with simulated predictions. Additionally, we present two examples of first applications of low energy μ^+ in μ^+ SR investigations. We measured the magnetic field distribution inside a 500-nm thin High- T_c superconductor (YBa₂Cu₃O_{7- δ}), as well as the depth dependence of the field distribution near the surface. In another experiment a 500-nm thin sample of Fe-nanoclusters (diameter 2.4(4) nm), embedded in an Ag matrix with a volume concentration of 0.1%, was investigated with transverse field μ^+ SR.

During the last two decades the muon spin rotation techniques (μ SR) [1] have been developed into a powerful research tool for condensed matter studies. These techniques normally use polarized positive muons (μ^+), which are implanted into a sample where they act as microscopic probes of magnetic properties at the stopping sites of the μ^+ . The information about local magnetic fields, field variations, magnetic phase transitions and muon diffusion is obtained by measuring the time-dependent angular distribution of the positron originating from the anisotropic μ^+ decay. In semi-conductors and insulators the μ^+ may thermalize as muonium ($\text{Mu} \equiv \mu^+e^-$), which is used as a light hydrogen isotope to study diffusion processes, to obtain information about defects, and as a probe in free radical chemistry. The available muon beams for μ^+ SR deliver μ^+ with energies in the MeV range. The range of these μ^+ in condensed

matter is about several hundred μm with a range straggling of the order of 100 μm . Therefore, investigations on thin films, multi-layers, interfaces and surfaces are not accessible to the conventional μ^+ SR method. In order to extend the applicability of μ^+ SR to these growing fields, we have developed a beam of polarized low energy μ^+ with energies between a few eV and 20 keV. These energies correspond to ranges between a few nm and about 100 nm. The new promising technique (LE- μ^+ SR, low energy μ^+ SR [2]) becomes now available at the Paul Scherrer Institute (PSI) and we present in this paper first LE- μ^+ SR measurements on thin film samples.

To date, the most efficient method to generate nearly 100%-polarized μ^+ with energies of a few eV is based on a moderation technique. An intense beam of surface muons with an energy of approximately 4 MeV is decelerated in a moderator consisting of a metal substrate at a temperature of about 10 K, which is covered by a condensed $\sim 500\text{-nm}$ thin van der Waals layer (generally Ar or N_2). In the final moderation stage before thermalization, the slowing down of μ^+ in the layer is completely dominated by soft muon-atom elastic collisions. These μ^+ are likely not to thermalize but escape from the frozen layer at epithermal energies of about 10 eV with an energy spread of 20 eV (FWHM). The efficiency for the conversion of a fast μ^+ into a slow one amounts to 10^{-5} – 10^{-4} . The slow μ^+ are extracted from the moderator region by applying a high voltage of up to 20 kV to the electrically insulated substrate. Then, the μ^+ are focused onto the sample using an electrostatic transport system. Details on the moderation process and the PSI apparatus can be found in [2–4].

In order to fully exploit the potential of LE- μ^+ SR the interaction of μ^+ in solids must be better understood. For example, the knowledge of the range distribution is of fundamental importance for the interpretation of LE- μ^+ SR data as well as for the design of thin samples for future experiments. Until now, no data are available for μ^+ due to a lack of suitable beams. Various Monte Carlo simulation programs are available to calculate the transport of particles in matter. We examined the reliability of predictions of ranges for μ^+ in matter based on Monte Carlo simulations by investigating the implantation depth of μ^+ in Cu/SiO₂ samples with different Cu thicknesses (see figure 1(a)). In a transverse magnetic field (5 mT) μ^+ SR experiment the frac-

Figure 1. (a) Method to measure the implantation profile of μ^+ in matter. (b) Fraction of μ^+ stopped in a 50-nm Cu film as a function of the implantation energy. The curve represents the prediction of the modified version of the program TRIM [6].

tion of μ^+ stopping in the thin Cu layer can be easily distinguished from the fraction stopping in SiO_2 , where nearly all μ^+ form Mu which precesses about hundred times faster. Figure 1(b) shows the measured fraction of μ^+ stopped in a 50-nm Cu layer. The data are compared with a modified version of the program TRIM (Transport of Ions in Matter) [5,6]. Within the present experimental errors the measured fractions are well reproduced by the simulation.

The conventional μ^+ SR technique has proven its ability to measure the local field distribution inside the bulk of poly- and single-crystalline superconductors (SC) to yield information on the dynamics of the vortex state. During the last few years of intense research on the field of high- T_c SCs it has become more and more clear that some of the applications most likely to be realized will be based on thin films. Such samples are accessible to LE- μ^+ SR, but not to the conventional μ^+ SR technique. In a first experiment we studied a 500-nm thin $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ sample, where we applied an external magnetic field of 28 mT perpendicular to the sample surface and parallel to the c -axis. Muons of 16 keV kinetic energy, corresponding to a mean range of 70 nm, were implanted in the sample. The distribution of local fields experienced by the μ^+ (called line shape) is determined using the maximum entropy method. Figure 2 shows the results obtained at different temperatures. Above the transition temperature of $T_c = 87.5$ K only the external field B_{ext} is detected. With decreasing temperature the maximum B_{saddle} of the field distribution decreases and the line shape develops the typical features with a high field tail. This indicates the formation of the flux line lattice in the SC. From the shape and the value of $(B_{\text{ext}} - B_{\text{saddle}})$ the penetration depth can be determined. It agrees with the values found for bulk samples. Additionally, by varying the energy of the μ^+ and by using an YBCO sample with a 90-nm Ag capping layer, we were able to measure the variation of the field distribution as a function of depth near the surface of the SC. When approaching the surface from the inside of the SC the shape of the field distribution is narrowed and B_{saddle} is shifted closer to B_{ext} . Outside the SC (i.e., in the Ag layer) the field distribution due to the vortex structure is still measurable since the length scale of the variation of the field distribution is about $0.2a$ (a is a distance between the vortices).

Figure 2. Temperature dependence of the magnetic field distribution inside a 500-nm thin $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ layer measured with 16 keV μ^+ in an external transverse field of 28 mT.

A smooth transition to the homogenous external field is observed when the distance to the surface of the SC is further increased. The preliminary analysis of the data shows that the measured variation of the field distribution is well described by the London model [7].

Samples of nano-sized, ferromagnetic single domain clusters, embedded in a matrix with low concentration, are an almost ideal system to study the phenomenon of superparamagnetism. Superparamagnetic relaxation is mostly investigated by Mössbauer spectroscopy and magnetization measurements. The μ SR technique offers a very different observational time window of the dynamics of the cluster moments. Especially interesting for LE- μ^+ SR are well prepared samples such as mass selected clusters whose preparation is limited to thicknesses of the order of a few hundred nm [8]. We investigated with transverse field μ^+ SR a 500-nm thin sample of Fe-nanoclusters (diameter of 2.4(4) nm), embedded in an Ag matrix with a volume concentration of 0.1% and a magnetic moment of $\sim 1200\mu_B$ of each cluster. Due to the low concentration Mössbauer spectroscopy is not applicable in this case. The implanted μ^+ sense the Fe-cluster moments indirectly via the distribution of dipolar fields resulting from the cluster ensemble. Thermal activation flips the cluster moments with a characteristic time $\tau = \tau_0 e^{E_A/kT}$, where τ_0 is the high temperature limit, E_A – the activation energy, k – the Boltzmann constant and T – the temperature. This behavior is reflected in the relaxation rate λ as a function of temperature shown in figure 3. At low temperatures the muon senses a static distribution of dipolar fields giving rise to a large, constant relaxation rate. At the blocking temperature of about 15 K the magnetic moments are still frozen on the time scale of the μ^+ . Above 20 K the clusters show superparamagnetic behavior and the slope of $\log \lambda$ becomes proportional to E_A . From a fit to the data the activation energy and τ_0 can be determined. We obtain $E_A = 63(5)$ K and $\tau_0 = 1.2(4) \times 10^{-8}$ s.

The reported experiments have partially exploratory character, but clearly demonstrate the present potential of LE- μ^+ SR at PSI. They have been obtained with a surface muon beam rate of $1.2 \times 10^7 \text{ s}^{-1}$ and 5% momentum bite, yielding a slow μ^+ rate of about 400 s^{-1} at the moderator and about 60 s^{-1} at the sample position. With

Figure 3. Relaxation rate λ as a function of temperature T of 16 keV μ^+ implanted in an Fe-nanocluster sample. $\lambda_0 = \lambda(T = 0)$. An external transverse field of 25 mT is applied.

currently implemented improvements of the LE- μ^+ beam the last number is predicted to increase to about 150 s^{-1} .

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