Muon Spin Rotation and Relaxation Experiments on Thin Films [★]

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Abstract. The recent development at the Paul Scherrer Institute of a beam of low energy muons allows depth dependent muon spin rotation and relaxation investigations in thin samples, multilayers and near surface regions (low energy μ SR, LE- μ SR). After a brief overview of the LE- μ SR method, some representative experiments performed with this technique will be presented. The first direct determination of the field profile just below the surface of a high-temperature superconductor in the Meissner phase illustrates the power and sensitivity of low energy muons as near-surface probe and is an example of general application to depth profiling of magnetic fields. The evolution of the flux line lattice distribution across the surface of a YBa₂Cu₃O₇ film in the vortex phase has been investigated by implanting muons on both sides of a normal-superconducting boundary. A determination of the relaxation time and energy barrier to thermal activation in iron nanoclusters, embedded in a silver thin film matrix (500 nm), demonstrates the use of slow muons to measure the properties of samples that cannot be made thick enough for the use of conventional μ SR. Other experiments investigated the magnetic properties of thin Cr(001) layers at thicknesses above and below the collapse of the spin density wave.

Key words: muon spin rotation, LE- μ SR method, muon spin relaxation.

1. Introduction

Highly polarized positive muons (μ^+), implanted with about 4 MeV kinetic energy in matter (so-called surface muons) are widely used as microscopic magnetic probes or spin labels (Muon Spin Rotation or Relaxation (μ SR)). However, because of the high energy, mean and variance of the implantation depth are of the order of fraction of mm, thus limiting the application of the μ SR technique to the study of bulk matter properties without any depth resolution. On the other hand, thin films, multilayers and near surface regions are of increasing importance in contemporary condensed matter science, with the reduced dimensionality

[★] Invited.

providing a new approach into fundamental and emergent physical behavior and novel technological applications. The muon, as a local sensitive probe with complementary observational time window to other probes or techniques, offers new insights into these objects of investigation. To be used for thin film studies and to be able to perform depth dependent scanning of a probe, the energy of the particle must be tunable in the interval between a few tens of keV down to a few eV for surface investigations. At these energies, implantation depths in matter typically extend from 200–300 nm down to the subnanometer region. Such a low energy (LE) beam with μ SR spectrometer has been developed over the recent years at the Paul Scherrer Institute thus setting the basis of the LE- μ SR method and opening new fields of μ SR investigations [1]. Basically, LE- μ SR allows depth dependent scanning of samples and the study of samples, which are too thin to be investigated with standard μ SR techniques.

2. Generation of epithermal muons

At present, in analogy with the similar case in positron physics, the moderation technique is the most simple and efficient method [2, 3] to create a sufficient amount of very slow particles starting from particles that are intrinsically of energetic nature (the muon energy from the pion decay is about 4 MeV). Briefly, an intense beam of readily available surface μ^+ is slowed down to epithermal energies (~ 15 eV) in an appropriate moderator consisting in a thin layer (few hundreds nm) of a van der Waals bound solid deposited on a metal substrate. The generation of epithermal muons relies on the specific behavior of charged particle in wide band gap cryosolids, particularly in the regime preceding thermalization. The general picture that emerges from various investigations is as follows. In these weakly bound solids, in the final stage before thermalization, slowing down and scattering are dominated by soft μ^+ -atom elastic collisions. In this energy regime, electronic slowing down processes such as ionization, excitation or electron capture are strongly suppressed or even energetically not allowed because of positive threshold energies. Therefore, muons, which by the statistical nature of the energy loss process in matter fall within the appropriate energy range in the condensed layer are likely to avoid neutralization (muonium formation) and thermalization and escape from the frozen gas at epithermal energies. The reduced energy-loss rate in these perfect insulators leads to a large escape depth of the order of 50 nm and to efficiencies of the order of 10^{-5} – 10^{-4} to convert an energetic fast muon into a very slow one [4]. The moderation mechanism is also reflected by the correlation between efficiency and band-gap energy and by the energy distribution of the emitted epithermal muons peaking at ~ 15 eV. The initial almost 100% longitudinal polarization of the incoming surface muons is conserved, because of the fastness of the moderation process (~ 10 ps) [3]. The moderation efficiency can be optimized by choosing appropriate growth conditions of the solid gas layer. The long-term stability of the moderator is ensured by ultrahigh vacuum (UHV) conditions.

3. A low energy μ SR experiment

Experiments are performed with the apparatus schematically shown in Figure 1. The UHV apparatus combines the source of epithermal muons, the low energy beam and the μ SR spectrometer. Its operation is as follows. Surface muons of energy ~ 4 MeV are incident at a present rate of $\sim 2 \cdot 10^7$ /s onto a cryogenic moderator consisting of ~ 300 nm of solid nitrogen condensed onto a microstructured silver substrate held at ~ 10 K and at a positive potential of 15 or 20 kV [5]. Epithermal

Figure 1. Layout of the apparatus used to generate the polarized low energy muon beam and to perform muon spin rotation and relaxation experiments.

muons emerging from the moderator are accelerated in its potential and transported by electrostatic “*einzel*” lenses and a mirror to the sample, where they are focused to a beam spot with typical diameter 10–15 mm (FWHM) and arrive at a rate of about 300/s. The electrostatic mirror separates low energy muons from any other particles such as beam positrons or fast muons exiting the moderator. The low energy muons are detected where they pass through a 10-nm thin carbon foil placed at an intermediate focus of the beam transport system (“trigger detector” in Figure 1). At the foil, the muons release a few electrons, which give the required muon implantation signal for time differential μ SR experiments in a microchannel plate. On passing through the foil, the muons lose 1.6 keV and acquire an rms energy spread of 0.5 keV. The trigger signal is also used to measure the time-of-flight (TOF) of each low energy muon after it was detected at a scintillator on entering the apparatus. By selecting on the TOF, we discard muons coming from the moderator with energies outside the epithermal region, but with low enough energy to be reflected by the mirror. The final kinetic energy of the muons arriving at the sample may be varied over the range ~ 0 –30 keV by applying an accelerating or decelerating potential of up to 12.5 kV to the sample, which is mounted via a sapphire plate on a continuous-flow helium cryostat or another sample holder. Due to the 90° deflection at the electrostatic mirror, the muons are horizontally polarized, transverse to their direction of motion, when they arrive at the sample. Hence, the muon spins will precess in the horizontal plane if a vertical magnetic field is applied, or alternatively in a vertical plane about a horizontal field parallel to their direction of motion. Both geometries have been used in experiments on this apparatus. The decay positrons from the muons implanted in the sample are normally detected by a set of four scintillator “telescopes” placed left, right, above and below the beam axis.

4. Microscopic magnetic field distributions near the surface of a $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin film

4.1. VORTEX STATE

Over the past 20 years the μ SR technique has been applied to measure various aspects of the magnetic behavior of high-temperature superconductors whose superconducting properties are intimately related to their magnetic properties. For instance, μ^+ can measure the microscopic field distribution in the vortex state of a type II superconductor, which is characterized by a large internal spatial inhomogeneity due to the penetration (for $B_{c1} < B_{\text{ext}} < B_{c2}$) of quantized flux lines and the formation of a flux line lattice (FLL). Since the muons are randomly distributed over the flux line lattice, the internal field has a simple relationship with $P(t)$, the time evolution of the muon spin polarization. The microscopic field distribution is obtained by Fourier transforming $P(t)$. Muon spin relaxation is one of the most applied bulk methods to absolutely determine λ , the London magnetic penetration depth, a fundamental property of the superconductor, since its value re-

flects the number density n_s and effective mass m_s^* of the superconducting carriers: $\mu_0 \lambda^2 = m_s^* / 4e^2 n_s$.

We used LE- μ^+ to investigate the evolution of the magnetic field distribution generated by the FLL as the vortices emerge through the surface of a superconducting film [6]. The experiment was performed with a 700 nm thick, c -axis oriented, homogenous $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO) film with transition temperature 87.5 K, epitaxially grown by thermal coevaporation on a single crystal LaAlO_3 substrate. A magnetic field was applied parallel to the c -axis and transverse to the μ^+ spin. By varying the energy of the slow muon beam we were able to adjust the implantation depth into the film and thus to measure the evolution of the field distribution as the vortex lines emerge from the surface of the film. To access the region outside the superconductor a second YBCO film was grown under exactly the same conditions and covered ex situ with a 70 nm thick Ag layer. By stopping the muons within this Ag layer we can monitor the spatial evolution of the field distribution outside the superconductor. The field distribution $P(B)dB$, i.e., the fraction of the sample which has a local field between B and $B + dB$, is the Fourier transform of the muon polarization $P(t)$ as obtained from the measured asymmetry spectra:

$$P(t) = \int P(B) \cos[\gamma_\mu Bt + \Phi] dB \quad (1)$$

(γ_μ – muon gyromagnetic ratio: $\gamma_\mu/2\pi = 0.1355 \mu\text{s}^{-1} \text{ mT}^{-1}$, Φ – initial spin phase). The transform is better performed using a so-called maximum entropy technique, which reduces the effect of Poisson noise in decay spectra with low statistics [7]. In Figure 2(a), we display three representative field distributions (background corrected), which were obtained by implanting muons with energies of 29 and 3 keV into the uncovered YBCO film and with an energy of 3 keV into the Ag covered film. According to Monte Carlo simulations with the TRIM.SP code [8], whose reliability to predict muon implantation profiles has been tested in separate experiments [9], muons with these energies come to rest at a mean depth of 135 and 18 nm below the YBCO surface in case of the uncovered film and stop 55 nm in front of the superconductor film within the Ag layer in case of the Ag covered film.

The field variation $B(x, y, z)$ within the triangular vortex lattice produces a distinctive asymmetric field distribution $P(B)$. It displays a pronounced tail towards high fields arising from regions within the vortex lattice close to the vortex cores, a maximum which corresponds to the most probable field B_{sad} at the saddle point between adjacent vortices, and a cutoff on the low field side corresponding to the field minimum at the point which is most remote from the vortex cores. Figure 2(a) shows that, as the flux lines emerge towards and through the surface of the superconducting film, B_{sad} moves towards the applied field and the overall width of the field distribution diminishes indicating a more homogenous magnetic field distribution. On the outside of the superconducting film, the field distribution is still asymmetric and reminiscent of the internal field distribution but the tail

Figure 2. (a) Measured magnetic field distribution at $T = 20$ K and $B_{\text{ext}} = 10.4$ mT at distances of $z = 135$ nm and $z = 18$ nm below and $z = -55$ nm in front of the surface of a $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ film. As the flux lines emerge through the surface of the film, both the difference between most probable field B_{saddle} and B_{ext} and the tail on the high field side are drastically reduced (the solid curves are a fit from the London model). (b) Depth dependence of $B_{\text{ext}} - B_{\text{saddle}}$ obtained at $T = 20$ K and $B_{\text{ext}} = 10.4$ mT. The dashed curves are predictions of the London model for different values of the London penetration depth λ_{ab} and $B_{\text{ext}} = 10.4$ mT. Excellent agreement between the experimental and calculated values is obtained for $\lambda_{ab} \approx 150$ nm.

towards high fields is drastically reduced. Overall, the data provide evidence that the size of a vortex increases at the surface of a superconductor. The line shapes show appreciable smearing relative to the theoretical field distributions, which have been obtained by assuming that inside the superconductor ($z > 0$), the London equation describes the spatial variation of $B_z(x, y, z)$, with source terms representing the flux line cores, whereas on the outside B_z obeys Laplace's equation. The smearing effects are taken into account by folding the theoretical field distribution with the $\text{LE-}\mu$ stopping profile and a Gaussian. The resulting fit with the London penetration depth along the ab -plane, λ_{ab} , as single free parameter is shown as a solid line in Figure 2(a) and well reproduces the experimental data yielding $\lambda_{ab} = 145(5)$ nm. The penetration depth can also be obtained from an integral quantity such as the difference between the mean field B_0 and the most probable

field B_{sad} of the field distribution [10]. With the field applied perpendicular to the thin film above T_c , demagnetizing effects ensure that $B_0 = B_{\text{ext}}$. In Figure 2(b) we plot the measured depth dependence of $B_{\text{ext}} - B_{\text{sad}}$ and compare it to the results of calculations for three different values of the London penetration depth λ_{ab} of 140, 150 and 160 nm (dashed lines). It can be seen that λ_{ab} controls the magnitude of the cusp shift, but has very little effect on the depth dependence. The deviations of the field distribution from uniform (outside) or bulk flux lattice (inside) die away to zero over the distance $\sim a_0/2\pi$ from the boundary at $z = 0$, where a_0 is the flux line spacing. Excellent agreement between the measured and calculated shift is obtained for a penetration depth of about 150 nm, in good agreement with the value determined from the exact analysis of the $P(B)$ distributions.

4.2. MEISSNER STATE

The determination of λ from the moments of the magnetic field distribution in the vortex state by muon relaxation gave the first broadly accepted value for this fundamental quantity. This method is not free of limitation, however. The second and (higher) moments of the magnetic field distribution depend on all sources of magnetic field inhomogeneities. Furthermore, one has to rely on the use of the modified London model, where λ describes how the magnetic field decays away from the vortex core, and know the vortex lattice symmetry and structure. Flux line core effects, deviations of the flux line lattice from its ideal configuration and other effects such as finite time range and ringing suppression have to be included in the data analysis.

The depth sensitivity in the nm range of LE μ^+ penetrating into the surface region allows to directly measure values of magnetic fields as a function of depth, thus avoiding the difficulties mentioned above [11]. For this experiment a magnetic field of 9.5 mT was applied parallel to the surface of the YBCO film (i.e., perpendicular to the c -axis) after zero field cooling to 20 K. The large value of B_{c1} at low T , surface barriers and flux pinning, all prevent flux line entry, so in these measurements we are measuring Meissner screening. It is well known that superconductors in the Meissner state do not completely exclude a magnetic field applied parallel to the surface, but that the field penetrates a short distance into the superconductor. In the simplest case, it is expected that an external field B_{ext} , applied parallel to the surface, penetrates according the equation

$$B = B_{\text{ext}} e^{-z/\lambda}, \quad (2)$$

where z is the depth perpendicular to the surface, and λ is the London magnetic penetration depth, describing here the magnetic field decay from the superconductor surface. LE- μ^+ can provide an experimental proof of Equation (2), which is expected to apply in those superconductors (such as high- T_c materials including YBCO) which have a large value of the ratio $\kappa = \lambda/\xi$, where ξ is the coherence length. Equation (2) was first predicted in 1935 [12], but never exper-

imentally tested at microscopic level. This is of relevance, if we consider that microwave techniques used to measure with high precision the temperature dependence of the change in the London penetration depth, $\Delta\lambda(T)$, implicitly assume Equation (2) [13].

It should be stressed that low energy muons provide a direct measure of the spatial variation of the magnetic field beneath the surface. This technique differs in principle and not just in methodology from the various methods used to deduce the London penetration length in that no assumptions have to be made about the functional form of the field penetration law. By tuning the energy from 3 to 30 keV the depth of implantation was varied between ~ 20 and ~ 150 nm. The muon implantation depth distribution $p(z)$ corresponding to each of the implantation energies was calculated using the Monte-Carlo code TRIM.SP (Figure 3(a)). Figure 3(b) shows the corresponding magnetic field distributions obtained at 20 K from various implantation energies. The higher the incident muon energy, the lower the average field they experience. It can also be seen that muons of a given energy stop over a certain range of depths, giving a small range of fields in $P(B)$. To an excellent first approximation, to plot the field B as a function of depth z , we simply read off the peak values in Figure 3(b) and plot them versus the peak values in Figure 3(a). This plot allows one to iterate rapidly to the correct relationship between $P(B)$ and $p(z)$:

$$P(B) = \frac{p(z(B))}{|dB/dz|}. \quad (3)$$

The values of B_{peak} versus z_{peak} obtained from the iteration are plotted in Figure 4(a) for several sample temperatures. The theoretical lines are plots of the function:

$$B(z) = B_{\text{ext}} \frac{\cosh((t - z)/\lambda)}{\cosh(t/\lambda)}, \quad (4)$$

which is the form taken by Equation (2) for a thin film of thickness $2t$, with the flux penetrating from both surfaces. The actual variation of field with depth in Figure 4(a) is well fitted by the theoretical expression with no adjustable parameter other than the temperature dependent λ . For 20 K we find for instance a value of 146 ± 3 nm. Since the shielding currents essentially flow in the ab -planes the penetration depth we are measuring is λ_{ab} . The data at the different temperatures agree well with the values from the vortex state measurement described in the preceding section and from single crystal measurements by bulk μ SR [14].

In conclusion, we have used low energy muons to make the first direct microscopic measurement of magnetic field values near the surface of a superconductor in the Meissner state, providing a direct confirmation of the London equation and the possibility of investigating cases where Equation (2) does not apply. The functional form of the spatial decay of the magnetic field beneath the surface of a superconductor is of particular interest in condensed matter research. For example,

Figure 3. (a) Depth distributions of implanted muons for various energies, calculated using the Monte Carlo code TRIM.SP. These distributions have been convoluted with a Gaussian of width 5 nm to represent the depth straggling due to surface roughness of the film. It will be noted that the asymmetrical shape of the stopping distributions is reflected in the asymmetry of the field distributions in (b). (b) Field distributions observed in YBCO at 20 K in a parallel field of 9.5 mT, with muons implanted at the same energies as in (a). As the muons are implanted deeper within the sample in the Meissner state, there is a reduction in the average field experienced by the muons. The narrow field distribution at 9.5 mT shown with open circles was taken at 100 K – i.e., in the normal state, and is independent of implantation energy.

Figure 4. Values of field versus corresponding depth for various values of sample temperature: The solid lines represent fits to Equation (4), with the value of λ as the free parameter. The insert shows the result of the fit.

the exponential decay law is not expected to apply in the case of low- κ materials such as aluminum, lead or tin (type I superconductors), where the attenuation of the field is weaker due to nonlocal effects arising from the large coherence length of the superconducting charge carriers. In an extreme type I superconductor ($\xi \gg \lambda$), for instance, Pippard model predicts for the magnetic penetration depth

$$\lambda = \lambda_{\text{London}} \left(\frac{\xi}{\lambda_{\text{London}}} \right)^{1/3} > \lambda_{\text{London}}$$

[15]. Furthermore in such “nonlocal” superconductors a sign reversal of the field is expected at depths of order $z/\xi = 0.2$ [16]. Also, in high- T_c materials, the d-wave nature of the pairing may give nonlinear and nonlocal corrections to Equation (3), particularly at low temperatures.

5. Relaxation of iron nanoclusters embedded in a thin nonmagnetic matrix

In this study, we implanted 16 keV muons in thin film samples of 500 nm thickness, consisting of a silver matrix containing iron clusters of diameter 2.8(5) nm with an equivalent iron concentration of 0.1% by volume [17]. This experiment is an example of investigations, where a variable implantation depth is not necessarily required but LE- μ^+ are needed because of the reduced thickness of the system to be investigated.

The magnetization vector of these single domain ferromagnetic particles may be observed to be unstable against thermal activation, a behavior known as superpara-

Figure 5. The muon spin relaxation rate over temperature range 4.7–300 K. The open symbols show the results of fits of the asymmetry data with the low temperature form of the relaxation function, $\beta = 1$. The full symbols correspond to the high temperature form, $\beta = 0.5$. At high temperatures (20–300 K), an Arrhenius law fits the relaxation rate. The slope gives an energy barrier of 51 ± 9 K. The intercept corresponds to an intrinsic cluster relaxation time of $\tau_0 = 12 \pm 4$ ns.

magnetism [18]. Superparamagnetism is interesting both as a fundamental limit of magnetic storage stability and as a signature of the microscopic mechanisms of magnetic reversal. Small single domain nanoclusters are an almost ideal system with which the phenomena can be studied. Metallurgical methods usually used to produce nanoparticles give rise to wide dispersion of sizes, variety of shapes and particle separation that varies considerably. This limits the interpretation of the results since quantities, such as relaxation time and activation energy, depend critically on these parameters. Narrow distribution of sizes and shapes may be obtained in thin film samples, too thin to be investigated by standard μ SR methods, containing clusters formed from an evaporated mass-filtered flux in a gas condensation source [19]. In the samples used in this first experiment, the filtration step was omitted but the method still gives a tighter size distribution than can be obtained, for instance, by precipitation.

The implanted particles sense the Fe cluster moments indirectly via the distribution of dipolar magnetic fields resulting from the cluster ensemble. Transverse field measurements in 25 mT were collected as a function of temperature after field cooling the sample temperatures down to 4.7 K. The decay positron time spectra were fitted in the time domain with a signal of the form

$$N_i(t) = N_i^0 e^{-t/\tau_\mu} [1 + A_i e^{(-\sigma t)^\beta} \cos(\omega_\mu t + \phi_i)]. \quad (5)$$

A_i and ϕ_i are the amplitude and phase of the original muon asymmetry spectra, and the stretched exponential represents the spin relaxation function with σ the

relaxation rate (the stretched form is a consequence of averaging over different fluctuating local fields at different muon sites). This quantity is plotted as a function of $1/T$ in Figure 5. At low temperatures, where the exponent takes the value of $\beta = 1$, a constant, large, exponential relaxation is observed. This behavior reflects a static dilute distribution of dipoles producing a Lorentzian field distribution for the spatial components of the internal field [20]. In this regime the muon-spin relaxation rate σ_{Lorentz} is related to the half-width half-maximum (HWHM) ΔB of the field distribution by $\Delta B = \sigma_{\text{Lorentz}}/\gamma_{\mu}$. The average damping rate of $0.4 \mu\text{s}^{-1}$ corresponds to $\Delta B = 0.5 \text{ mT}$.

The expected changeover from a constant relaxation rate at low temperatures to a temperature dependent rate at high temperatures occurs around 15 K, which defines the so-called blocking temperature, i.e., the temperature where the spin dynamics as seen by the μ^+ is blocked. This value is consistent with SQUID magnetometry measurements showing that magnetization curves at 10 and 25 K are non-hysteretic and superpose when plotted against B/T . On the effective time scale of a SQUID measurement moments begin to freeze at 5 K. At higher temperatures, the implanted muons experience rapidly fluctuating random fields, characterized by a relaxation time τ . The correct form of relaxation function in this case is an exponential with the relaxation exponent $\beta = 0.5$ [20]. In this regime the muon-spin relaxation rate $\sigma(T)$ is related to the cluster relaxation time $\tau(T)$ by $\sigma(T) = 4(\gamma_{\mu}\Delta B)^2\tau(T)$.

Figure 5 shows that, for temperatures above $\sim 15 \text{ K}$ the temperature dependence of the relaxation follows closely an Arrhenius law. Thermally activated, the magnetic moments fluctuate spontaneously giving rise to a damping rate of the form $\sigma(T) = \sigma_0 \exp(E_A/k_B T)$. From the intercept of the fitted curve in Figure 5 we obtain an intrinsic cluster relaxation time (inverse of the attempt frequency) of $\tau_0 = 1/\sigma_0 = 12 \pm 4 \text{ ns}$. The linear slope of the data gives an activation energy of $E_A = 51 \pm 9 \text{ K}$. If we assume that the cluster volume is $1.2 \cdot 10^{-26} \text{ m}^3$, as suggested by magnetometry data we find a cubic anisotropy constant of $K = 2.3 \pm 0.4 \cdot 10^5 \text{ J m}^{-3}$. This enhancement by a factor of 5 over the bulk value is consistent with the enhanced orbital moment found previously for similar clusters [21]. The enhancement arises from the reduced coordination of the atoms at the surface of a cluster, which results in an incomplete quenching of the orbital moment. However, the possibility of deviations of the nanoclusters from spherical shape should also be considered.

This experiment shows that it is possible to study the temperature dependence of nanocluster dynamics in the superparamagnetic state by LE- μ SR and that this technique can be used to extract parameters such as activation energy, anisotropy energy and relaxation times. Recent developments of the gas condensation source used to deposit the clusters now allow the deposition of clusters of diameter 2 nm with 5% resolution in time periods appropriate for thin film deposition. Furthermore, the kinetic energy with which these highly selected clusters are deposited onto the substrate may now be controlled with electrostatic acceleration grids. This

affords control over deformation of the clusters when they lose their kinetic energy and come to rest on the substrate, and the possibility of control over the shape anisotropy [21].

6. Magnetism of thin chromium films

Whereas magnetic properties of overlayers can be investigated by various techniques, buried layers are more difficult to access experimentally especially when antiferromagnetic (AF) order is involved. Direct methods such as neutron and X-ray scattering also encounter difficulties because of the small scattering volume. The low energy and the relative narrow stop distribution of the muons allow studying buried thin layers with a local probe, thus complementing investigations employing radioactive isotopes [22]. Recently, the magnetic structure of thin Cr films has attracted much interest because of its use in giant magnetic resistance materials like Fe/Cr multilayers, where Cr acts as a mediating spacer between the exchange coupled Fe layers (for a recent review see [23]). In these multilayers, by reducing the thickness t_{Cr} , the Néel temperature decreases from the bulk value of 311 K and, finally, drops to zero at a critical thickness $t_0 \approx 4.2$ nm. This value is close to the modulation period of the spin density wave (SDW) which is formed in the bulk below the Néel temperature with the antiferromagnetic ordered Cr moments showing spatial sinusoidal modulation. The thickness dependence is well described by a scaling theory assuming a magnetically dead layer of the thickness t_0 below which the incommensurate SDW does not fit into the Cr layer anymore [24]. However, the strong dependence of the magnetic structure on parameters such as specific structural properties resulting from different sample preparation conditions has led to contradictory pictures about the magnetic state of thin Cr films. Below the critical thickness, paramagnetic or commensurate AF phases, for thicknesses up to 25 nm longitudinal SDW versus transverse SDW can be found in the literature.

We performed first investigations with LE- μ^+ of the magnetic properties of thin Cr films buried by nonmagnetic boundary layers. We studied two thicknesses one above and one below the collapse of SDW where different effects are expected [25]. The Cr samples (one with $t_{\text{Cr}} = 3.4$ nm and another with $t_{\text{Cr}} = 9.1$ nm) were prepared by molecular beam epitaxy with non-magnetic boundary layers deposited on a clean MgO (0 0 1) surface. X-ray reflectometry measurements, gave the following layer sequences in nm:

Sample 1: MgO(0 0 1)/3.4(3)Cr/21.3(4)Au/9.1(4)Cr/34.5(10)Al,

Sample 2: MgO(0 0 1)/4.3(3)Cr/21.4(3)Au/3.4(3)Cr/10.2(3)Au/3.4(4)Cr/
35.0(10)Al

(the numbers in parentheses denote the mean roughness).

By varying the muon implantation energy between 1.8 and 29.3 keV we can change the fraction of muons stopping in Cr or in the nonmagnetic layers. Zero field measurements show that muons stopping in Cr in the SDW state, because of the

Figure 6. Dependence of the total asymmetry as a function of implantation energy at two temperatures for (a) 9.1 nm thin Cr layer, (b) 3.1 nm thin Cr layer.

broad field distribution sensed by them, very rapidly depolarize with a relaxation rate of the order of $10 \mu\text{s}^{-1}$. In transverse field measurements, the asymmetry A_{TF} reflects the fraction of muons which have thermalized in a nonmagnetic surrounding. For the 9.1 nm Cr layer, the observation of a significant drop in A_{TF} as the temperature is lowered associated with a concomitant peak in the relaxation rate clearly indicates a magnetic phase transition at 285 ± 5 K. The crossing of the temperature boundary is also observed in the energy dependence of A_{TF} with the same layer, which shows an almost flat energy independent maximum asymmetry at 320 K (above the transition temperature), in contrast to a decrease of A_{TF} with E_{impl} at 20 K, reflecting the increasing muon fraction stopping in the magnetically

ordered buried Cr layer (Figure 6(a)). Differently from these results, for the 3.4 nm layer no difference between the energy scans at 320 and 20 K is observed. In this case, at both temperatures a reduction of A_{TF} is observed with increasing E_{impl} , parallel to the increasing probability for muons to enter the Cr layer (Figure 6(b)). These results show that, whereas the 9.1 nm thin Cr layer reveals a magnetic phase transition at 285 K, the one with a thickness of 3.4 nm exhibits a static magnetic ordered state in the temperature range from 20 to 320 K. This observation is consistent with recent experiments of thin Cr layers ($t_{Cr} < 4$ nm), grown adjacent to a monolayer of Sn enriched with the ^{119}Sn Mössbauer nuclei. For all thicknesses investigated, the hyperfine field at the Sn nucleus site shows magnetic ordering at temperatures much higher the Néel temperature in the bulk [26]. By contrast, PAC studies of Ag/Cr multilayers [27] find Cr to remain in the longitudinal SDW phase up to a temperature of 500 K and observe, below a critical thickness of ~ 5.1 nm, the collapse of the SDW and a transition to paramagnetic behavior. At present, it is not clear whether different strains originating from diverse sample preparation conditions or other extrinsic factors may account for the observed variance.

7. Conclusion

The development of a full understanding of many magnetic and/or superconducting systems requires the use of experimental probes that access the physical quantities of concern on a local scale within the material. The availability of low energy polarized muons has now made possible the development of one of the most desired and natural extension of μSR , i.e., the use of muons for depth resolved analysis of thin films, interfaces and surfaces. This work provides a brief overview of some experiments recently performed, indicating the potential of the new low energetic magnetic microprobe. For instance, direct measurements of the magnetic field distribution or of the field penetration law have the potential to yield information crucial to understanding the detail of the superconducting state in superlattice systems, consisting of superconducting and magnetic or insulating layers.

Measurements of the properties of thin magnetic layers have to cope very often with the detection of small asymmetry signals of high frequency and large relaxation rate. In spite of these difficulties, the experiment on thin multilayered Cr samples has given evidence that open questions concerning the magnetism of these systems can be addressed with LE- μSR . Comparable exploratory measurements of thin films of colossal-magnetoresistance material and of dimensional effects in spin glasses have also been initiated [28].

Low energy μSR is still in its infancy and the experiments described here have to be considered as first generation experiments. However, the first steps have been done. The intensity of the source has been steadily evolving over the past years and higher fluxes will certainly contribute to the full realization of the potential. Different promising directions to increase the flux can be envisaged. Intensity increases may be obtained at the new proposed μSR facilities or with specially designed

muon production targets. A recent study shows that also at existing muon factories, such as PSI, about a factor of 10 higher fluxes of surface and consequently of $\text{LE-}\mu^+$ can be reached with the construction of a new beam designed for optimum collection and transport of surface and low momenta muons [29].

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