

Muon-spin rotation experiments in high- T_c superconductors and related materials

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Recent muon-spin rotation μ SR experiments in high- T_c superconductors and related antiferromagnetic materials are reviewed. The possibilities and the limitations of the μ SR method for investigating these materials are briefly discussed. In a high- T_c superconductor, μ SR is an ideal tool with which to study the local magnetic field distribution at the muon site, allowing a determination of the London penetration depth. It is further shown that μ SR experiments may contribute to the microscopic understanding of the superconducting glass state in the high- T_c oxides. In the related antiferromagnetic materials μ SR is a sensitive method for detecting frozen local magnetic moments.

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1. Introduction

The discovery of the high- T_c copper oxides by Bednorz and Müller has stimulated an enormous number of experimental and theoretical investigations of these and related antiferromagnetic materials in order to search for possible mechanisms responsible for superconductivity in these materials. Most experimental work has been done with standard techniques (susceptibility, resistivity, specific heat). In contrast to such experiments, the muon-spin rotation (μ SR) technique provides a sensitive *microscopic* probe of the local magnetic field (at the muon) in high- T_c superconductors and related antiferromagnetic materials. The μ SR method has been used successfully to investigate various magnetic properties of these materials, and a large number of papers on this subject have already appeared in the literature [1]. In this paper some typical μ SR experiments are reviewed. Since the field is growing so rapidly, a comprehensive treatment is not possible here, but the examples presented are representative. It will be demonstrated that μ SR experiments can make relevant contributions to the microscopic understanding of these materials.

The organization of the paper is as follows: In Section 2 the basic principles of the μ SR technique are briefly reviewed, insofar as is relevant to the interpretation of the following experiments. In Section 3 some recent μ SR measurements of the London penetration depth λ in La-Sr-Cu-O and Y-Ba-Cu-O are reported. The possibilities and limits of the μ SR technique concerning the determination of λ are discussed. The glasslike superconducting properties of the high- T_c oxides are discussed in Section 4. It is shown that recent μ SR results on Y-Ba-Cu-O are consistent with results of macroscopic experiments (susceptibility, resistivity) and with the predictions obtained from models of weakly coupled superconducting domains. Section 5 is devoted to μ SR experiments involving the interplay between magnetism and superconductivity in the La-Sr-Cu-O system. Such experiments are of special importance, since it is generally agreed that magnetism may play an essential role in these CuO₂-based materials [1]. The conclusions follow in Section 6.

2. The muon-spin rotation (μ SR) technique

In a μ SR experiment, spin-polarized positive muons are implanted into a sample to be investigated. The sample is located in an external magnetic field that is perpendicular to the initial muon-spin polarization. During the short thermalization process, the muons retain their polarization and subsequently undergo Larmor precession at 135.5 MHz/T in the local magnetic field. After a mean lifetime of 2.2 μ s, the muon decays into a positron and two neutrinos. As the decay positrons are emitted preferentially along the muon-spin direction (due to parity violation), the muon-spin precession can therefore be observed by detecting the number of decay positrons as a function of time after a muon has stopped in the sample.

The observed decay-time histogram of muons stopped in the sample may be written in the form

$$N(t) = N_0 \exp(-t/\tau_\mu) [1 + AR(t) \cos(2\pi\nu_\mu t + \alpha)] + B, \quad (1)$$

where N_0 is a normalization constant, τ_μ is the muon lifetime (2.2 μ s), A is the precession amplitude, ν_μ is the muon precession frequency, α is the initial phase, and B is a time-independent background. The relaxation function $R(t)$ describes the damping of the precession signal. In the case of a Gaussian distribution of internal fields, the relaxation function has the form

$$R(t) = \exp(-\sigma^2 t^2), \quad (2)$$

where σ is the muon-spin depolarization rate. In general, the data are analyzed by performing least-squares fits to the time-histogram $N(t)$ defined in Equation (1).

In a magnetic solid a measurement of the Larmor precession frequency $\omega_\mu = 2\pi\nu_\mu$ allows a determination of the local magnetic field B_μ at the muon site according to the

relation $\omega_\mu = \gamma_\mu B_\mu$, where $\gamma_\mu = 2\pi \times 135.5$ MHz/T is the gyromagnetic ratio of the muon. In the case of a superconductor the average local magnetic field $\langle \tilde{B}_\mu \rangle$ at the muon site is given by

$$\langle \tilde{B}_\mu \rangle = \tilde{B}_{\text{ext}} + \mu_0(1 - N)\tilde{M}, \quad (3)$$

where $\tilde{B}_{\text{ext}}$ is the external field, N is the demagnetization factor, and $\tilde{M}$ is the magnetization of the sample. The precession amplitude A is a measure of the fraction of muons precessing in the field, and the depolarization rate σ yields information on the local field distribution at the muon site. For a Gaussian distribution of internal fields, the second moment of the field distribution $\langle \Delta B_\mu^2 \rangle$ can be determined from the depolarization rate σ using the relation

$$\langle \Delta B_\mu^2 \rangle = 2\sigma^2/\gamma_\mu^2. \quad (4)$$

One should note, however, that in general (see below) the local field distribution is *not* Gaussian [2]. In this case a more sophisticated data analysis is required in order to determine the first and second moment of the local field distribution from the measured μ SR time spectrum. In magnetically ordered solids, the μ SR technique can be used to study frozen local magnetic moments. Ferromagnetic and antiferromagnetic ordering give rise to local magnetic fields at the muon site which yield muon-spin precession, even in zero external field. The μ SR method is also sensitive to fluctuations in local magnetic fields, yielding information on the dynamics of magnetic moments in a solid. A detailed description of the μ SR technique is given in [3].

3. London penetration depth

From a μ SR experiment it is in principle possible to determine the London penetration depth λ , one of the fundamental quantities of a superconductor. The muon serves as a microscopic probe of the local field distribution in the bulk of a superconductor. In the mixed phase of a type-II superconductor, the external magnetic field ($B_{\text{ext}} > B_{c1}$) penetrates the sample in the form of a regular vortex lattice. This results in a broad distribution of local fields, the width of which is determined by the penetration depth λ . Assuming that a perfect triangular vortex lattice is formed in an isotropic superconductor, the second moment of the local field distribution is field-independent and is given [3] by

$$\langle \Delta B^2 \rangle \approx (\sqrt{3}/32\pi^3) \Phi_0^2/\lambda^4, \quad (5)$$

where Φ_0 is the elementary flux quantum. Here it is assumed that λ is larger than the distance between vortices. For the superconducting oxides this condition is satisfied for $B_{\text{ext}} > 0.1$ T. For an ordinary BCS-like superconductor, the temperature dependence of λ is given by the empirical formula

$$\lambda(T) = \lambda(0)[1 - (T/T_c)^4]^{-1/2}, \quad (6)$$

where

$$\lambda(0) \propto \sqrt{m^*/n_s}. \quad (7)$$

Here m^* is the effective mass and n_s is the carrier density. Equation (6) usually holds for BCS-like superconductors where the coherence length is larger than λ , a condition which is certainly not true for the high- T_c superconducting oxides, which have an extremely short coherence length [4].

By combining Equations (4) and (5), the penetration depth λ can in principle be extracted from the measured depolarization rate σ according to the relation

$$\sigma^2 \approx (\sqrt{3}/64\pi^3)\gamma_\mu^2\Phi_0^2/\lambda^4. \quad (8)$$

However, in order to get a reliable value for λ , the following conditions must be satisfied: (1) A perfect vortex lattice must be present (no flux pinning due to sample inhomogeneities); (2) λ must be larger than the vortex spacing; (3) the local field distribution must be Gaussian-like; and (4) λ must be isotropic. A critical discussion concerning the determination of λ from μ SR experiments is given by Brandt [2]. For instance, he shows that approximating the asymmetric field distribution by a Gaussian distribution, as assumed in most μ SR experiments, leads to a value of λ which is too large for a perfect vortex lattice and to a value of λ which is too small for a vortex lattice distorted by flux-pinning.

Several μ SR groups have reported measurements of λ in ceramic high- T_c oxide superconductors [5–12]. In most experiments, λ was determined from the temperature-dependent depolarization rate σ using the simple relation in Equation (8). Here only two typical examples of these experiments are presented.

Kiefl et al. [10] carefully studied the temperature dependence of σ in sintered $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ after field-cooling the samples in an external field of 1.5 T. It is important to note that the measurements were done on field-cooled samples in order to attain a regular vortex lattice in the sample. For the field-cooled case the magnetic flux density is almost constant above and below T_c . Any change of the external magnetic field in the superconducting state gives rise to flux-pinning in these inhomogeneous materials. Flux-pinning effects are in general temperature-dependent, and may therefore lead to an inhomogeneous line broadening. The measurements were performed in a high external field $B_{\text{ext}} = 1.5$ T, since it was found that at low temperature the depolarization rate is almost constant for $B_{\text{ext}} > 1$ T, indicating that flux-pinning effects are negligible at high fields. From the low-temperature limit for $\sigma(0)$, the effective isotropic average of the penetration depth in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ ($T_c = 90.7$ K) was estimated to be $\lambda(0) \approx 1450$ Å. For $T/T_c < 0.2$, the temperature behavior of λ was found to be well described by the BCS-like behavior given in Equation (6), suggesting an s-wave pairing mechanism. However, at higher temperatures there is a serious deviation from the conventional temperature behavior which may be explained by assuming a very strong coupling [10].

Recently, Uemura et al. [11] studied the temperature dependence of the depolarization rate σ in sintered samples of $\text{YBa}_2\text{Cu}_3\text{O}_x$ ($x = 6.66, 6.95, 7.0$) and $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$. For $\text{YBa}_2\text{Cu}_3\text{O}_{7.0}$ ($T_c = 91.1$ K) the effective isotropic average of the penetration depth was found to be $\lambda(0) \approx 1470$ Å, in agreement with $\lambda(0) \approx 1450$ Å reported in [10] (see above). However, in contrast to Kiefl et al. [10], they observed that the temperature behavior of λ in $\text{YBa}_2\text{Cu}_3\text{O}_x$ ($x = 6.95, 7.0$) is well described by the conventional formula in Equation (6). In addition, they found empirically that the depolarization rate $\sigma(0)$ at zero temperature is approximately proportional to the critical temperature T_c for the different samples investigated [$\sigma(0) \propto T_c$]. Using Equations (7) and (8), they obtained the “universal” relation

$$\sigma(0) \propto 1/\lambda(0)^2 \propto n_s/m^* \propto T_c. \quad (9)$$

This empirical relation implies that the critical temperatures T_c of various high- T_c oxide superconducting systems (even with different crystallographic structures) seem to be proportional to the carrier concentration n_s divided by the effective mass m^* . They argued that a change in T_c is mainly due to a change of n_s rather than to a change of m^* . This is supported by the experimental fact that the Sommerfeld constant of the low-temperature specific heat, which for a two-dimensional model is proportional to m^* , does not differ very much for various high- T_c superconducting oxides. In the framework of BCS theory, the “universal” relation in Equation (9) suggests a high-energy scale of the interaction which mediates the coupling between the superconducting carriers [11].

So far, we have neglected the extreme electronic anisotropy in the high- T_c superconducting oxides. In these layered materials the supercurrents are expected to flow dominantly in the CuO_2 planes, giving rise to an *anisotropic* penetration depth. The screening of an external field perpendicular to the CuO_2 plane is expected to be much larger ($\lambda_\perp$) than for a field parallel to the plane ($\lambda_\parallel \gg \lambda_\perp$). The μ SR lineshape of an anisotropic type-II superconductor in the vortex phase was calculated by Celio et al. [13] using a simple model. Schopohl and Baratoff [14] used a more sophisticated approach to calculate the field distribution in an anisotropic extreme type-II superconductor. From an experimental point of view, the situation is less clear, since it is difficult to extract an anisotropic penetration depth from μ SR experiments on ceramic samples. In order to determine the anisotropy of λ in these materials, μ SR experiments on high-quality single crystals are required. Such experiments are now in progress.

4. Glasslike properties of superconductors with a short coherence length

Shortly after the discovery of the high- T_c superconducting oxides, it was suggested [15] that these materials may have glasslike superconducting properties [16, 17]. The first

experimental evidence for glassy behavior in these materials was obtained from susceptibility measurements on the La-Ba-Cu-O system [15]. Several features that are typical of glasslike behavior were observed: (a) pronounced differences in the magnetic susceptibility of field-cooled and zero-field-cooled samples (metastable effects); (b) the existence of a phase line separating reversible from irreversible behavior ("quasi-de-Almeida-Thouless line"); and (c) nonexponential time dependence of trapped flux. Several other experiments [1] have also been discussed in terms of a glasslike superconducting state, including magnetic relaxation, magnetic hysteresis, electrical resistivity in a magnetic field, and μ SR experiments.

To describe the glassy behavior in these granular superconductors, we consider an assembly of superconducting domains of uniform phase that are weakly coupled through Josephson junctions or the proximity effect. The behavior of this weakly coupled system may be studied in an external magnetic field by using the Hamiltonian [16, 17]

$$\mathcal{H} = - \sum_{\langle i,j \rangle} J_{ij} \cdot \cos(\phi_i - \phi_j - A_{ij}). \quad (10)$$

Here J_{ij} is the coupling constant between clusters i and j , $\phi_{i,j}$ are the superconducting phases, and the phase factor A_{ij} is defined by

$$A_{ij} = \frac{2\pi}{\Phi_0} \int_i^j \vec{A} \cdot d\vec{l}, \quad (11)$$

where $\vec{A}$ is the vector potential and Φ_0 is the elementary flux quantum. The integral is taken from domains i to j . Positional disorder of the domains leads to frustration (glassy behavior) in the system via the phase factors A_{ij} [17].

For ceramic superconducting oxides, it is very likely that Josephson junctions may occur *inside* the physical grains at defects such as twin boundaries and oxygen defect planes [4]. The appearance of these junctions is a consequence of the extremely *short* coherence length in these materials [4]. Therefore, it is expected that glassy behavior may also occur in *single crystals* containing twin planes [17].

The model in Equation (10) has been studied in detail by Morgenstern et al. [17] on a two-dimensional lattice where glassy behavior is introduced by site disorder of the superconducting domains. Schneider et al. [18] have considered a more general model of weakly coupled superconducting domains where the screening effects are also taken into account. The predictions of these models are consistent with several experimental findings [1], including μ SR experiments presented in this paper.

μ SR measurements on ceramic $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ that support the "glass picture" were performed by the University of Zurich group [19, 20]. The measurements were done on field-cooled (FC) and zero-field-cooled (ZFC), earth-field compensated samples in an external magnetic field ranging

from 20 to 300 mT using low-momentum muons (28 to 40 MeV/c). A detailed discussion of the results is given in [19, 20].

Some typical Fourier power spectra of μ SR time-histograms taken in an external magnetic field of 20 mT above and below the transition temperature T_c are shown in Figure 1. Above T_c the muons precess in the external magnetic field, as indicated by the dashed lines in Figure 1. Below T_c the lines are shifted to lower fields with respect to the external field, indicating that the sample is magnetized in the superconducting state [see Equation (3)]. Well below T_c the Fourier spectra look quite different for ZFC and FC samples. For example, at 8 K the line for the FC case is rather narrow and Gaussian-like, whereas for the ZFC case the signal is very broad and by no means Gaussian. The fact that the local field distribution is very broad for ZFC samples may be considered as direct evidence of a *metastable* state. Zero-field-cooling the system to a low temperature and switching on the field suddenly leads to a *metastable* state which slowly decays as the system equilibrates. The measured muon precession frequency ν_μ and the depolarization rate σ as a function of temperature in a field of 20 mT (FC versus ZFC) are shown in Figure 2.

Above T_c the constant value of $\nu_\mu = 2.69$ MHz corresponds to the external field of 20 mT, and the small constant value of $\sigma \approx 0.1 \mu\text{s}^{-1}$ is very likely due to nuclear moments. Below T_c a pronounced temperature-dependent frequency shift and an enhanced muon depolarization rate are observed. Note that well below T_c the precession frequency ν_μ and the depolarization rate σ are quite different for the FC and ZFC cases. One can clearly observe *irreversible* effects in the μ SR data as demonstrated in Figure 2: In a first step the sample is cooled in zero field (ZFC) to 8 K. After a magnetic field is turned on, the system is in a *metastable* state (see also Figure 1). When the temperature is subsequently raised to any temperature T_A below a certain temperature T^* (which depends on the external field) and then lowered again, the system responds *irreversibly*. However, when the sample temperature is raised above T^* , the system responds *reversibly* to any temperature change, as indicated by the FC curves in Figure 2. Note that the metastable effects are reflected in both the first (precession frequency) and the second moment (depolarization rate) of the local field distribution. Measurements taken at higher fields (100 and 200 mT) as well as on a different sample exhibit similar irreversible effects. The agreement of the μ SR results in Figure 2 with previous *macroscopic* susceptibility measurements [15] on the La-Ba-Cu-O system is striking. A comparison with temperature-dependent susceptibility measurements taken on the same sample indicates that the relative frequency shift $\Delta\nu_\mu/\nu_\mu^{\text{ext}} = (B_\mu - B_{\text{ext}})/B_{\text{ext}}$ [see Equation (3)] is proportional to the macroscopic susceptibility $\chi = M/H_{\text{ext}}$, suggesting that the muon probes the local susceptibility in the superconductor. In addition to

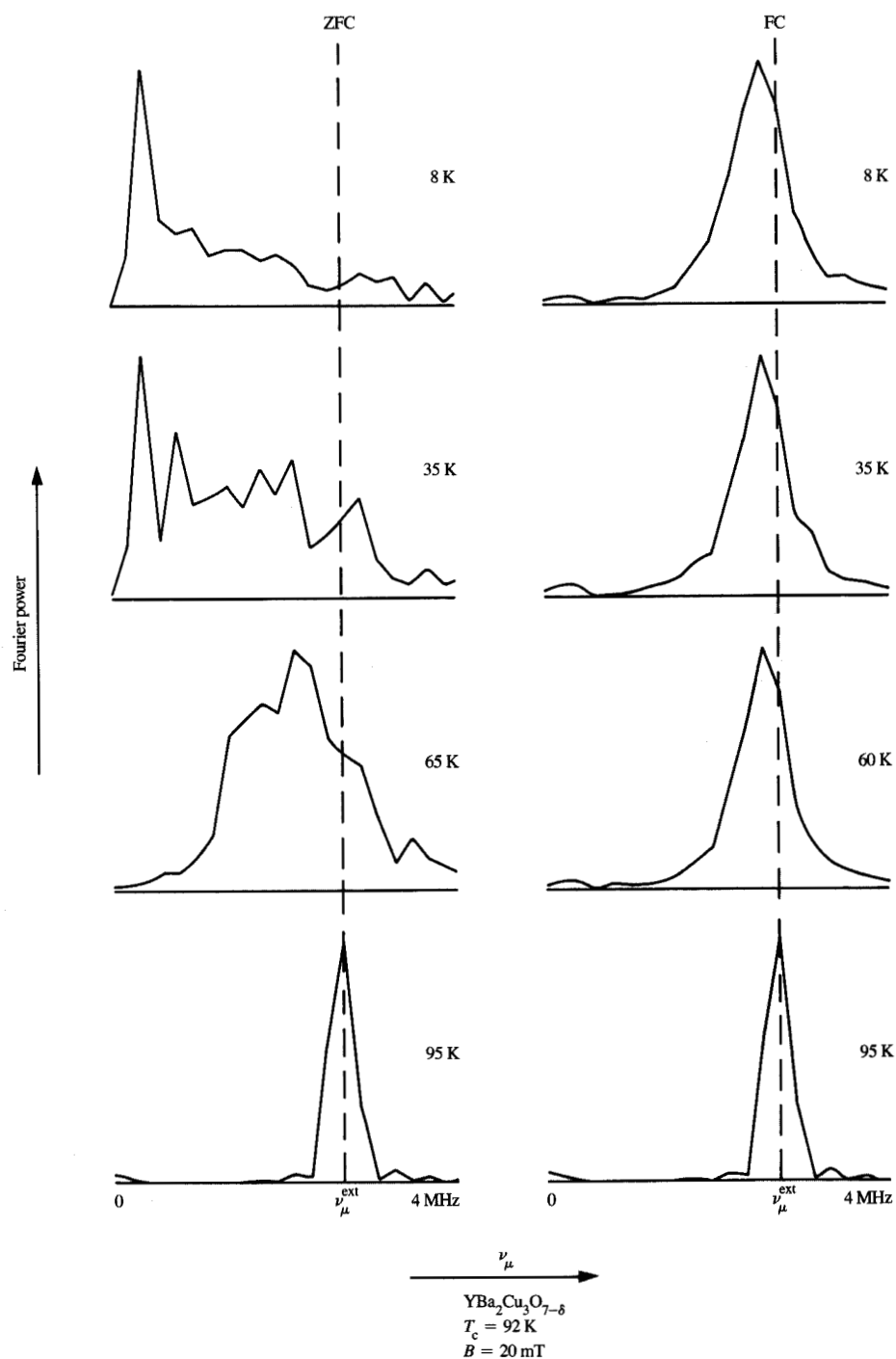


Figure 1

Fourier power spectra of μSR time-histograms taken at various temperatures above and below T_c in a field of 20 mT (ZFC vs. FC case).

standard susceptibility measurements, μSR experiments provide important information on the local magnetization distribution in a superconductor.

Another metastable feature of a "glassy" superconductor is magnetic hysteresis. A pronounced hysteresis was observed in the μSR depolarization rate σ when the magnetic field was

changed at a constant temperature (Figure 3). In this experiment the sample was cooled in zero field to 40 K, and then σ was measured as a function of field according to the loop indicated by the arrows in Figure 3. As a result, a "butterfly-like" hysteresis loop was obtained. For comparison, magnetization measurements taken on the same sample with a SQUID magnetometer are also shown in Figure 3 (right-hand scale). It is evident that the μ SR depolarization rate $\sigma(B)$ and the magnitude of the macroscopic magnetization $|M(B)|$ exhibit the same hysteresis effect. We found empirically that $\sigma(B)$, which is a measure of the second moment of the local field distribution [Equation (4)], is a linear function of $|M(B)|$: $\sigma(B) - \sigma_0 = a\gamma_\mu\mu_0|M(B)|$, where $\sigma_0 = 2.2(1) \mu\text{s}^{-1}$ and $a = 0.25(1)$ are constants. This empirical finding is consistent with the model of weakly coupled superconducting domains [18], which predicts that the variance of the induced field $\langle \Delta B_s^2 \rangle$ is proportional to the square of the magnetization M according to the relation $\langle \Delta B_s^2 \rangle = (\mu_0 M)^2 (\beta_A - 1)$, where β_A characterizes the induced field distribution. The present μ SR experiments yield $\beta_A = 1.2(1)$ in agreement with theory [18]. The field dependence of the second moment of the local magnetization distribution was also studied by Morgenstern [21] for the two-dimensional glass model of weakly coupled superconducting domains [17]. As a result, a butterfly-like hysteresis loop of the second moment, similar to that of the μ SR experiments, was obtained.

It has been found experimentally [15, 22, 23], as well as for models of weakly coupled superconducting domains [17, 18], that the phase line $T^*(B)$, separating reversible from irreversible behavior (see Figure 2), may be written in terms of a power law:

$$T^*(0) - T^*(B) \propto B^{2/3}. \quad (12)$$

The temperature $T^*(B)$ is defined by the appearance of irreversible behavior. Strictly speaking, this temperature is different from the critical temperature $T_c(B)$. However, according to experiments [15, 22] and theoretical considerations [17, 18], these two temperatures are almost indistinguishable. The phase line in Equation (12) was first found experimentally in the La-Ba-Cu-O system [15] and has been called the "quasi-de-Almeida-Thouless" (AT) line in analogy with spin glasses. For sintered $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, the AT line has been confirmed by means of electrical resistance measurements [22] in an external magnetic field, as well as by μ SR experiments, as shown in Figure 3 of [19]. Recently, Yeshurun and Malozemoff [23] have reported FC and ZFC magnetic measurements on a *single crystal* of Y-Ba-Cu-O. As a result, they also found an irreversibility line which is consistent with the AT line in Equation (12). The observed irreversibility line was interpreted in terms of a single-barrier thermally activated flux-creep model (giant flux-creep) using a simple scaling argument. Here the question arises: Is the glass model or the giant flux-creep model the more

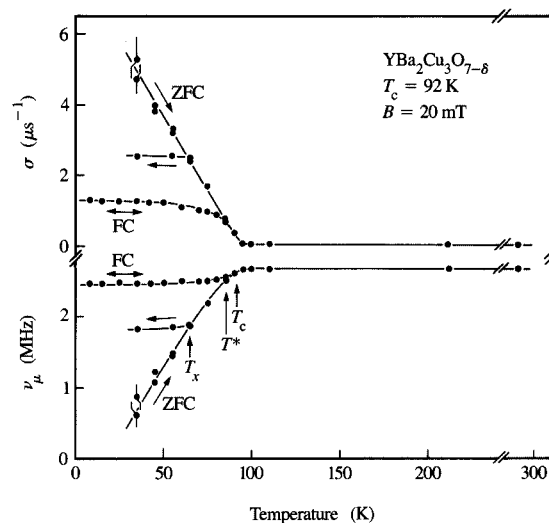


Figure 2

Muon precession frequency ν_μ and depolarization rate σ as a function of temperature in a field of 20 mT (ZFC vs. FC case). Lines are guides to the eye. See text for a detailed explanation.

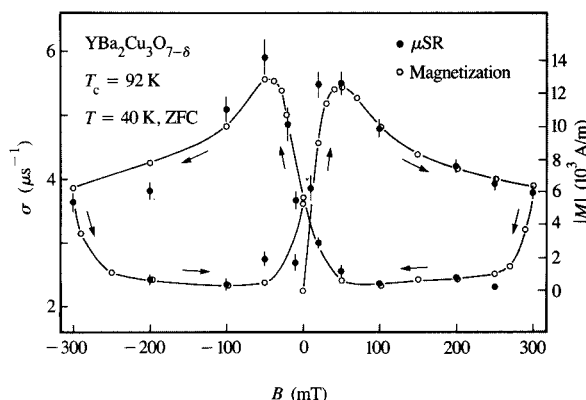


Figure 3

Field dependence of the depolarization rate σ (left-hand scale) and of the magnitude of the magnetization $|M|$ (right-hand scale) after ZFC at 40 K. The arrows indicate how the field was changed.

appropriate description of the glasslike phenomena observed in the superconducting oxides? It has been pointed out by Morgenstern [24] that for superconductors with a *short* coherence length, the glass model may be considered as a

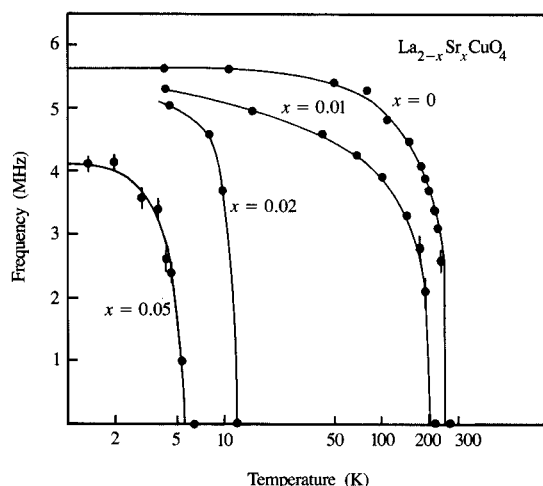


Figure 4

Temperature dependence of the muon precession frequency for sintered samples of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ measured in zero external field. Lines are guides to the eye (from Budnick et al. [27]).

generalization of the giant flux-creep model. At low temperatures both models essentially describe the same physics (single-barrier model). However, at higher temperatures (not too close to T_c) the simple flux-creep model fails, since one has to take into account a whole "hierarchy of barriers" giving rise to a broad distribution of relaxation times which is a characteristic feature of a glass.

5. Magnetism and superconductivity

Several experiments [1] including neutron scattering, transport, and μSR measurements suggest that magnetism plays an important role in the CuO_2 -based superconductors such as La-Sr-Cu-O and Y-Ba-Cu-O . Due to its rather simple structure (no interplay between CuO chains and planes), $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ is the most ideal high- T_c oxide system to search for a possible mechanism of superconductivity in the copper oxides. This system has been investigated by several μSR groups [25–28], and only part of this work is discussed here. Similar μSR experiments have also been performed in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ as a function of oxygen deficiency δ [29, 30].

The parent nonsuperconducting compound La_2CuO_4 is antiferromagnetic, with a Néel temperature around 250 K [26]. When this system is doped with the appropriate amount of Sr (or Ba), it becomes the famous superconductor discovered by Bednorz and Müller. In order to investigate the possible interplay between antiferromagnetism and superconductivity in this system, it is important to study the

magnetic phase diagram of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ in detail. The magnetic phase diagram of $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ was first studied by Japanese groups [31] by means of NMR. Independently, Aharony et al. [32] proposed a model in which the disorder in the Cu^{2+} spin system is introduced by the frustration of the oxygen holes. It is assumed that electronic holes on the oxygens induce local ferromagnetic exchange between neighboring Cu^{2+} spins and destroy the antiferromagnetic order in the copper spin system (competing ferromagnetic and antiferromagnetic interaction). This frustration model predicts a phase diagram in the insulating region ($x < 0.05$), including an antiferromagnetic and spin-glass phase, which is consistent with several experiments [27, 28, 31].

The use of μSR is a sensitive method for detecting magnetic ordering, and two groups [27, 28] have studied the magnetic properties of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ as a function of Sr concentration. Budnick et al. [27] performed μSR experiments on $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ($x = 0, 0.01, 0.02, 0.05, 0.1$) in zero external field. At low temperatures a clear precession signal corresponding to a single precession frequency was seen for $x \leq 0.05$, indicating the existence of a magnetically ordered state. However, it should be emphasized that because μSR is sensitive only to local fields, the type of magnetic ordering (long-range ferromagnetic or antiferromagnetic ordering or freezing of short-range correlations) cannot in general be determined from a μSR experiment [27, 28]. Here we define the "spin-freezing (Néel) temperature T_f " as the temperature where muon precession sets in (time scale $\approx 5 \mu\text{s}$). Figure 4 shows the temperature dependence of the measured precession frequency for various Sr-doped sintered samples. We note a dramatic decrease of the ordering temperature T_f when x is only slightly increased (for $x = 0$, $T_f \approx 250 \text{ K}$, whereas for $x = 0.05$, $T_f \approx 6 \text{ K}$). For $x = 0.1$ no precession signal was found. It is evident from Figure 4 that T_f decreases as the Sr concentration is approached where the system becomes superconducting for $x \geq 0.07$. On the other hand, the measured frequencies depend only weakly on the Sr concentration at low temperatures, indicating that the Cu^{2+} magnetic moments are not much affected by Sr doping. Therefore, the low ordering temperatures are due to a weakening of the exchange interaction between the Cu^{2+} moments, rather than to a reduction of the copper moments themselves. Similar results were also reported by Harshman et al. [28] for $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ and by Uemura et al. [25] for $\text{La}_2\text{CuO}_{4-\delta}$ where the ordering temperature is varied by δ . In addition, it was found [28] that for $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ the damping of the muon precession signal at low temperatures increases with increasing x , indicating that the local field is made more inhomogeneous by Sr doping.

Harshman et al. [28] performed μSR experiments on sintered $\text{La}_{1.95}\text{Sr}_{0.05}\text{CuO}_4$ in an external field parallel to the initial muon-spin polarization. They apparently observed two distinct components in the longitudinal time spectra:

Component I is due to fluctuating local fields ($B_\mu \approx 30$ mT) which freeze below T_f to yield muon-spin precession (see Figure 4). A longitudinal field of the order of 40 mT is needed to essentially quench the fluctuating local fields above T_f . Component II, on the other hand, is not quenched above T_f , even in a field as large as 160 mT. Thus, component II is most likely due to a set of much stronger magnetic moments which, however, do not yield a precession signal below T_f . They suggest that the two components may be associated with muons stopping at the two different oxygen sites in the sample, namely oxygens in the CuO_2 planes (site II) and oxygens between the planes (site I). Muons at site I will experience a net local magnetic field in the antiferromagnetic phase, giving rise to a muon-spin precession signal. On the other hand, muons located at site II in the CuO_2 planes will be exposed to zero magnetic field in the antiferromagnetic phase, since at this lattice site the total contribution from the two Néel sublattices is zero. However, these muons are very sensitive to spin configurations where two neighboring copper spins are parallel. The fact that the relaxation rate of component II, which is associated with muons at site II, is increasing with increasing Sr concentration is probably good evidence for the existence of ferromagnetically aligned pairs of Cu^{2+} moments, as proposed in the model of Aharony et al. [32].

An important conclusion of the present μSR experiments [27, 28] is that the local magnetic field at low temperatures is almost independent of the Sr concentration (see Figure 4), although the inhomogeneity in the local field increases with increasing x [28]. On the other hand, neutron-scattering experiments show that the three-dimensional magnetic moment decreases with increasing x . This contradictory result lies in the fact that the muon is a local probe of frozen magnetic moments, whereas in a neutron-scattering experiment one measures the sublattice magnetization. A comparison between μSR and neutron-scattering experiments suggests that at $x \approx 0.02$ a crossover occurs from antiferromagnetism to spin-glass behavior [28], in agreement with the proposed phase diagram [31, 32]. Spin-glass behavior is usually observed in systems with random competing interactions. In the model of [32], frustration, which leads to spin-glass behavior, arises naturally when the antiferromagnetic copper spin system is doped with the appropriate quantity of oxygen holes.

6. Conclusions

In conclusion, we have demonstrated that μSR is in many respects a unique method for investigating various magnetic properties of high- T_c superconductors and related antiferromagnetic materials.

In a superconductor μSR provides information on the local field distribution at the muon, allowing a determination of the London penetration depth $\lambda(0)$. So far, most experiments have been performed on sintered samples,

and values of the effective isotropic average of $\lambda(0)$ have been reported for various high- T_c superconductors [5–12]. It was found empirically by means of μSR that the “universal” relation $T_c \propto n_s/m^*$ [see Equation (9)] holds for various high- T_c superconducting systems (even with different crystallographic structures), suggesting a high-energy scale for the coupling between superconducting carriers [11]. μSR measurements on high-quality single crystals which allow a determination of the *anisotropic* penetration depth in these layered materials are now in progress.

μSR experiments [19, 20] of sintered samples of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ show several features that are characteristic of a glasslike superconducting state [15–17]: (a) irreversible effects and differences between FC and ZFC experiments (precession frequency, depolarization rate); (b) hysteresis effects in the depolarization rate and the magnetization; and (c) the existence of a “quasi-de-Almeida–Thouless” line separating reversible from irreversible behavior. The present experiments are consistent with other experiments (susceptibility, resistivity) which have been interpreted in terms of glasslike behavior, as well as with the predictions of models of weakly coupled superconducting domains [16–18]. The glassy behavior in the high- T_c oxides is a consequence of the extremely short coherence length [4] in these materials and therefore occurs also in *single crystals* containing twin planes. The superconducting glass model may be considered as a generalization of the giant flux-creep model [24].

Zero-field and longitudinal-field μSR measurements on $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ yield important information on the magnetic phase diagram of this system [25–28]. The antiferromagnetic ordering temperature drops drastically with increasing Sr concentration x . At low temperatures the local field at the muon is almost independent of x , indicating that the Cu^{2+} moments are not much affected by Sr doping. A comparison with neutron-scattering experiments [28] suggests that at $x \approx 0.02$, spin-glass ordering takes the place of antiferromagnetic ordering, as predicted by the simple model of Aharony et al. [32]. Longitudinal-field μSR measurements [28] indicate the appearance of “strong” magnetic moments; the population of these moments increases with x . The existence of “strong” moments is probably good evidence for ferromagnetically aligned Cu^{2+} spins, as proposed in the magnetic frustration model [32]. However, more detailed experiments are required in order to support this hypothesis.

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