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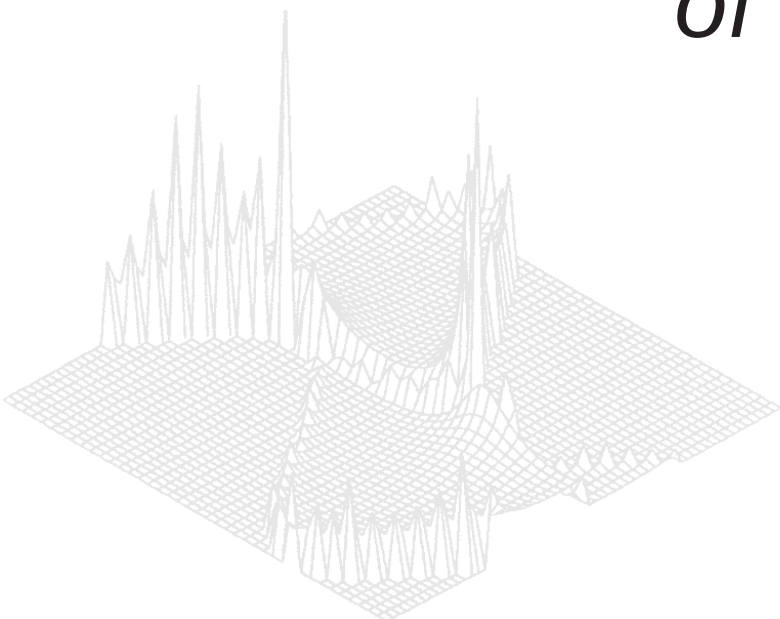

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Examples of the Application of Muon Spin Relaxation to Studies of Magnetism in Cuprates*

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Abstract

After a brief introduction to muon spin rotation and relaxation (μSR) some recently achieved results on $La_{2-x-y}Nd_xSr_yCuO_4$, $La_{2-x-y}Eu_xSr_yCuO_4$ and $Nd_{2-x}Ce_xCuO_4$ are presented demonstrating the high sensitivity of the method for the dynamics and ordering of small magnetic moments close to the superconducting phases. The μSR experiments give evidence for static and relatively slow dynamic magnetic correlations in these cuprates.

1. Introduction

The study of magnetic properties via implanted spin polarised positive muons μ^+ has become a standard method, which is complementary in its information to other nuclear probe methods like Mössbauer spectroscopy and NMR. In contrast to the latter, where usually substitutional sites are investigated, μ^+ is stopped at an interstitial site in a solid. Muon Spin Rotation and Relaxation (μSR) may probe local magnetic fields ranging between the dipolar contributions from nearby nuclei (some 10^{-4} Tesla) up to the strong dipolar and hyperfine fields met in magnetically ordered solids (several Tesla). It is mainly the sensitivity to very small fields which makes the method attractive for the study of very small ordered moments even down to the order of magnitude of nuclear magnetic moments which may escape observation by other methods. Typical examples are intermetallic heavy fermion systems (for a review see Schenck 1993), but very recently also the insulating oxide NpO_2 has been proven to show magnetic order of very small electronic moments (Kopmann *et al.* 1997). Depending on the experimental details and the specific conditions of a certain material the frequency window of μSR experiments ranges from about MHz to THz. Thus μSR is able to bridge the time scales of standard ac-susceptibility to the faster nuclear methods and neutron diffraction.

Introductions to the method can be found in the literature (e.g. Schenck 1985) and also on the webpages of the following muon facilities:

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PSI:	http://www.psi.ch
TRIUMF:	http://www.triumf.ca
ISIS:	http://ndafleetwood.nd.rl.ac.uk
KEK:	http://msl-www.kek.jp
LAMPF:	http://www.lanl.gov
JINR:	http://www.jinr.dubna.su.

In this paper we want to give some examples where the merits and advantages of μSR become apparent and may contribute to a better understanding of the physics involved. Instead of reviewing textbook examples we have chosen some very recent results obtained on cuprates. The μSR experiments described in the following have been performed at the muon facilities of PSI (Villigen, Switzerland), TRIUMF (Vancouver, Canada) and ISIS (Didcot, UK).

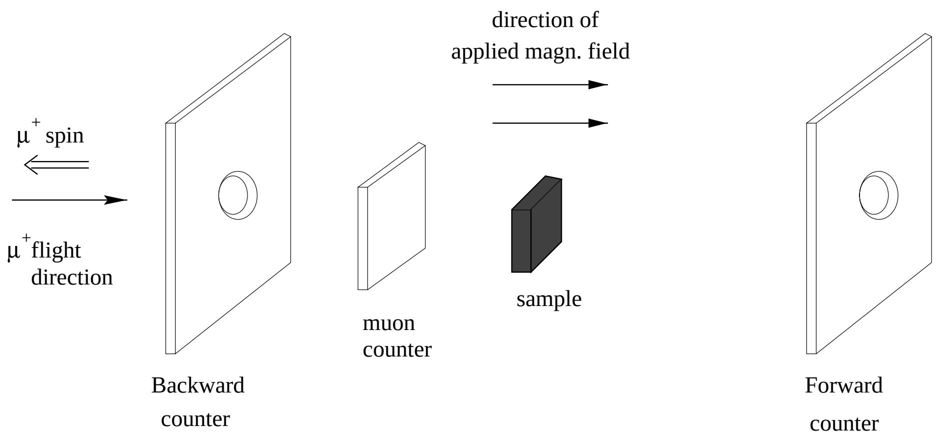

Fig. 1. Schematic set-up for zero field and longitudinal field μSR.

2. Experimental

Spin polarised μ^+ are usually produced from the decay of pions which are generated by a spallation process from medium energy protons (e.g. 590 MeV at PSI) in a stopping target (e.g. a carbon foil). The μ^+ are then implanted into the sample to be studied. Thermalisation occurs within 10^{-10}–10^{-9} s without loss of spin polarisation. The μ^+ come to rest at interstitial sites and decay with a lifetime of $2{\cdot}2\,\mu$s to a positron and a neutrino. A considerable asymmetry of the number of positrons is detected along the direction of the muon spin and opposite to it due to the parity violation in the weak interaction. In a standard μSR experiment (a schematic setup is depicted in Fig. 1) the time dependence of this backward–forward (b–f) asymmetry $(N_{\rm b}-N_{\bf f})/(N_{\rm b}+N_{\rm f})$ of the positron countrate is monitored via the backward and forward counter telescopes. The muon counter serves as start detector for the time histogram when a muon is implanted into the sample. Typical asymmetry spectra are shown in Fig. 2. If a magnetic field is acting at the site of the muon (this field may be an applied external one or an internally generated one by magnetic dipoles from nuclei and nearby atoms or a Fermi contact field) the muon spin will perform a precession leading to a sinusoidally modulated asymmetry pattern. A coherent spin precession even in a zero applied field (ZF) is expected for an ordered magnet resulting in a so-called 'spontaneous rotation pattern' (see e.g. the upper spectrum in Fig. 2). In a paramagnet the directions of the local fields will be randomly directed, thus no coherent spin precession can be observed and only a decay of asymmetry with time is found (lower spectrum in Fig. 2). This depolarisation may be caused by static local field distributions ('inhomogeneous broadening' in the language of NMR) or may be due to dynamic field fluctuations either caused by fluctuating electronic spins or by the changing magnetic surroundings met by a diffusing muon. For very fast fluctuations the damping of the signal vanishes corresponding to the limit of motional narrowing.

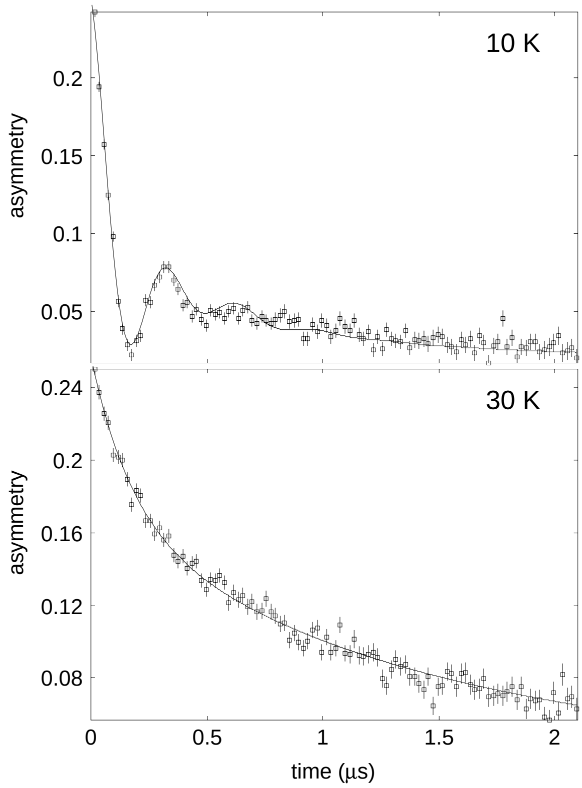

Fig. 2. Asymmetry plots for $La_{1\cdot25}Nd_{0\cdot60}Sr_{0\cdot15}CuO_4$ in the paramagnetic regime (lower spectrum) and ordered regime (upper).

Most informative in this respect is the analysis of the shape of the depolarisation function and its dependence on an applied field parallel to the initial orientation of the muon spin [the so-called longitudinal field (LF) arrangement—see Fig. 1]. With increasing strength of the applied LF the depolarisation of the muon spin is suppressed when the applied field becomes stronger than the static fields acting locally on the muon. The muon spin becomes 'decoupled' from its surrounding. Typical values of the LF necessary for decoupling static fields of nuclear dipolar origin are of the order 1–10 mT, whereas fields from electronic dipoles need $0\cdot1$–1 T. Fast dynamic fluctuations, however, will still cause relaxation of the muon spin. LF decoupling experiments thus allow us to distinguish between homogeneous and inhomogeneous origins of the depolarisation, a clear advantage for example compared to Mössbauer spectroscopy where this separation is usually impossible. In principle one may also decouple the muon spin from a dynamic surrounding. This, however, would require a still far stronger LF. Due to experimental difficulties in applying fields stronger than a few Tesla in a μSR experiment, this is hardly possible at the moment.

An admittedly often met problem in μSR is the determination of the stopping sites of the μ^+ in a certain solid. This can be solved by comparing the observed angular dependence of a static nuclear dipolar damping in a single crystal with quantum mechanically calculated values for various site candidates. Another possible way is to perform experiments under an applied magnetic field transverse to the muon spin. This allows one to follow the angular dependence of the muonic Knight shift in materials with non-ordered magnetic electronic moments. Also, spontaneous rotations observed in known ordered magnetic structures can be compared with dipolar sum calculations and thus may serve as an indicator for the stopping site. But even if the proper sites are uncertain, the observed magnetic response of the μ^+ is highly informative about the overall magnetic state of a specimen.

One also should keep in mind that the μ^+ may occupy different sites at different temperatures and may diffuse even at moderate temperatures. Then the magnetic information gained from μSR is an averaged one over the history of the muon's lifetime.

Another problem is faced if the amount of material available for an experimental target is very small, as is often the case for delicate compounds, especially if single crystals or thin layers are to be investigated. The commonly used muon beams typically provide μ^+ of about 4 MeV or higher making sample thicknesses of the order of mm necessary. At present there is very encouraging progress in the development of an efficient beam of very low energy muons which allow a shallow implantation. The presently achieved status of the very slow muon project at PSI was described by Morenzoni *et al.* (1996, 1997*a*, 1997*b*). With this new beam of muons in the energy range from eV to keV it will be possible to enter the nm range thus opening quite new fields for the investigations using μSR.

3. μSR on Cuprates

From the very beginning of the exciting developments following the discovery of superconductivity in cuprates, μSR has played a considerable role. Among the major successes of μSR in the field of high temperature superconductors are the discovery of the coexistence of magnetic correlations and superconductivity in the cuprate layers, the study of flux line patterns and their dynamics and of penetration depths (see Proc. Int. Conferences on Muon Spin Rotation, Relaxation and Resonance 1990, 1994, 1997). The information thus obtained is of considerable importance in attempts to understand the mechanisms of high temperature superconductivity. Especially, the appearance of small ordered magnetic moments has often escaped observation by neutron diffraction and μSR has proven to be the first method to detect them (Weidinger *et al.* 1989; Sternlieb *et al.* 1990; Torikai *et al.* 1990).

After a decade of intense research on high temperature superconductors the initially overwhelming enthusiasm has cooled down and a more careful and less hectic view has been allowed for revisiting e.g. the question of coexisting magnetism and superconductivity. Here we have in first place the antiferromagnetism of the CuO_2 planes carrying also the superconductivity. The ordered Cu moments are typically $\sim 0{\cdot}5\mu_B$ or smaller. But also the magnetism of rare-earth ions in the cuprates, their coupling to Cu and among themselves still offers open questions. In the rare-earth-123 compounds their interactions with Cu are weak, and so superconductivity is not suppressed. The rare-earth magnetism is also rather insensitive to the oxygen content, whereas Cu orders antiferromagnetically only in reduced samples.

In M_2CuO_4, with M = Nd, Pr, Sm, etc., which crystallises in the tetragonal T′ structure (see Fig. 3) complex non-collinear structures and spin reorientations are found. The interpretation of these magnetic phenomena is still under discussion (Sachidanandam *et al*. 1997 and references given there). Their influence on the superconductivity which is found e.g. in the Ce doped Sm and Nd compounds is also unclear. We will present here recent μSR results on the magnetic order of Nd_2CuO_4 and Pr_2CuO_4 and of the magnetic response of $Nd_{2-x}Ce_xCuO_4$ (Hillberg *et al*. 1997). The interpretation of specific heat data of the latter (Brugger *et al*. 1993) has caused controversial discussions. There is evidence that a new kind of heavy fermion behaviour appears in the overdoped n-type regime (Fulde and Zevin 1993; Fulde *et al*. 1993).

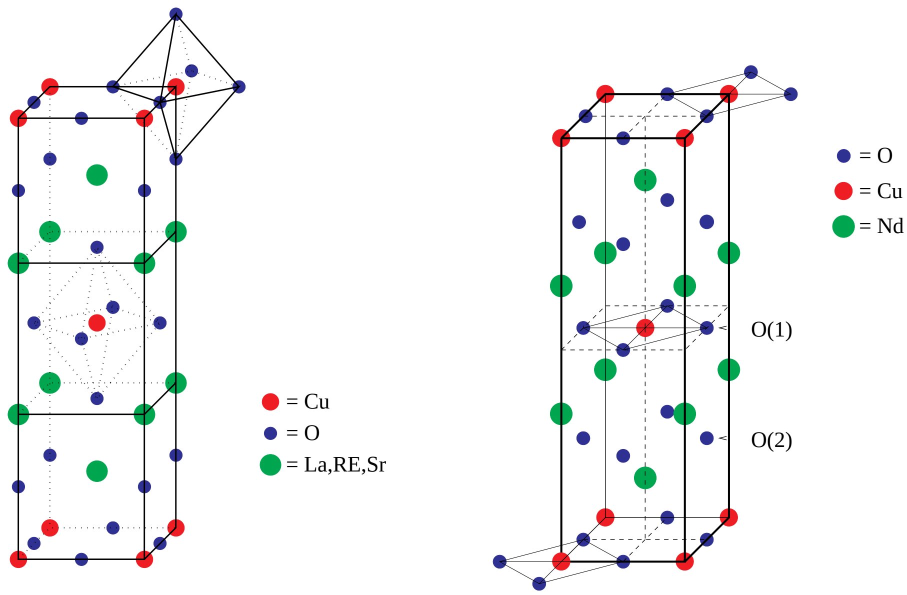

Fig. 3. Crystal structure of $La_{2-x-y}RE_xSr_yCuO_4$ (T structure *left*) and $Nd_{2-x}Ce_xCuO_4$ (T′ structure *right*).

(3a) Magnetic Behaviour of Sr Doped Tetragonal Systems

First, however, we want to turn to the ‘classical’ $La_{2-x-y}M_xSr_yCuO_4$ systems with M being a rare-earth ion. The most probable sites for muon stopping are about 1 Å from the apical oxygen atom. Without Sr doping these cuprates are antiferromagnetic due to the ordered Cu planes coupled weakly between each other. Even weak hole doping via Sr leads to a rapid suppression of T_N. Considerable effort has been devoted to understanding the interplay between magnetism and superconductivity in the orthorhombic phase of $La_{2-y}Sr_yCuO_4$ and its substitutional derivatives having the tetragonal T structure (see Fig. 3). The μSR experiments on $La_{2-y}Sr_yCuO_2$ were the first to actually prove the coexistence of two-dimensional magnetic correlations and superconductivity (Sternlieb *et al.* 1990). At higher temperatures the correlations are still dynamic, but freeze at a few Kelvin on the timescale of μSR, i.e. of the order of μs. In inelastic neutron scattering this freezing of Cu spins is also traced, however, at higher temperatures due to its shorter time window. All experiments provide evidence for a kind of spin glass. Judging from the μSR data nearly the whole sample takes part in the magnetic response thus excluding large scale phase separation into magnetic and superconducting fractions.

Phase segregation and structural and electronic instabilities have long been recognised as playing an essential role, not only from the preparational point of view, but also for a detailed understanding of the electric and magnetic behaviour of these materials. It has been proposed that holes introduced in the antiferromagnetic cuprate via Sr doping or oxygen deficiency might arrange themselves along lines separating antiferromagnetic regions (Emery and Kivelson 1993, 1994; Eskes *et al.* 1996; Zaanen *et al.* 1996). Inelastic neutron scattering in $La_{2-y}Sr_yCuO_4$ could in fact detect dynamic two-dimensional spin correlations, which are incommensurate with the crystal lattice (Mason *et al.* 1992). This incommensurability is explained by a modulation of both spin and charge in such a way that hole-rich domain walls separate antiferromagnetic domains; however, the charge order is not directly observable. Static stripe domains with spin and hole order have been revealed via neutron scattering (Tranquada *et al.* 1995, 1996) from superstructures in $La_{1\cdot 48}Nd_{0\cdot 4}Sr_{0\cdot 12}O_4$, which also has T structure but of the low temperature tetragonal (LTT) type and not the orthorhombically distorted one.

We have performed μSR on $La_{1\cdot 85-x}Nd_xSr_{0\cdot 15}O_4$. For this Sr concentration the LTT phase is found for Nd concentrations of $x \geq 0\cdot 2$; for smaller x the material has a low temperature orthorhombic (LTO) structure. Bulk superconductivity vanishes with the change from LTO to LTT for $x \geq 0\cdot 18$. Only some spurious superconductivity persists in the LTT compounds (Büchner *et al.* 1994). The μSR results reveal spontaneous rotation patterns (Fig. 2), i.e. coherent magnetic order for all Nd concentrations $x > 0\cdot 2$ below $T \sim 28$ K (Wagener *et al.* 1997). There is a superposition probably of several rotation frequencies, two being dominant. Their temperature dependencies are shown in Fig. 4. The behaviour of samples with different Nd content in the LTT phase is nearly identical. Down to temperatures of 4–5 K the frequencies follow roughly a magnetisation curve: below that one finds an upturn which is accompanied by an increase of the transverse damping rate. This goes parallel with an increase of the signal intensity of the magnetic peaks in neutron scattering (Tranquada *et al.* 1996) and is related to an increase of the Cu–Nd interaction which slows down the fluctuations of the Nd moments and is accompanied by a tilting of the Cu moments. It is tempting to interpret the two frequencies with muon sites in hole-rich and hole-depleted surroundings as expected for a magnetic stripe domain picture. One should stress that μSR by itself can only reveal that different magnetic surroundings develop. Whether these regions are stripes or have different shape cannot be decided.

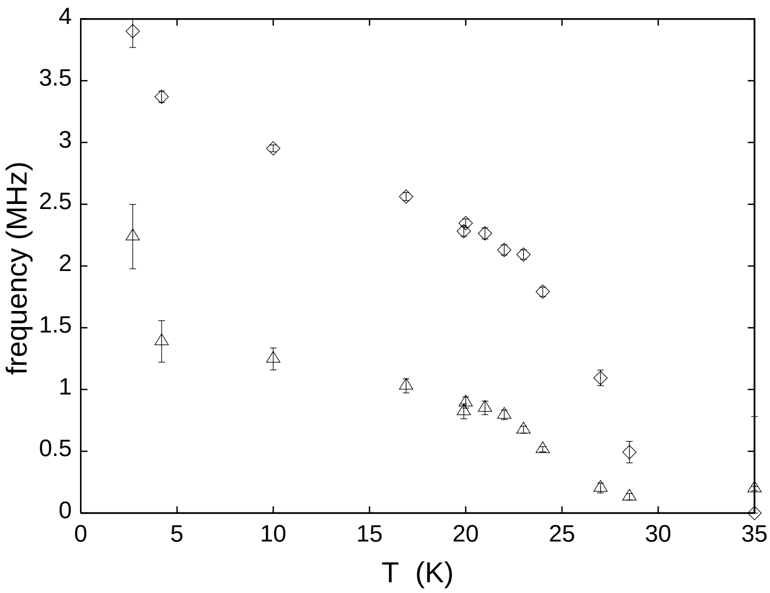

Fig. 4. Temperature dependence of spontaneous μSR frequencies in $La_{1\cdot 40}Nd_{0\cdot 45}Sr_{0\cdot 15}CuO_4$.

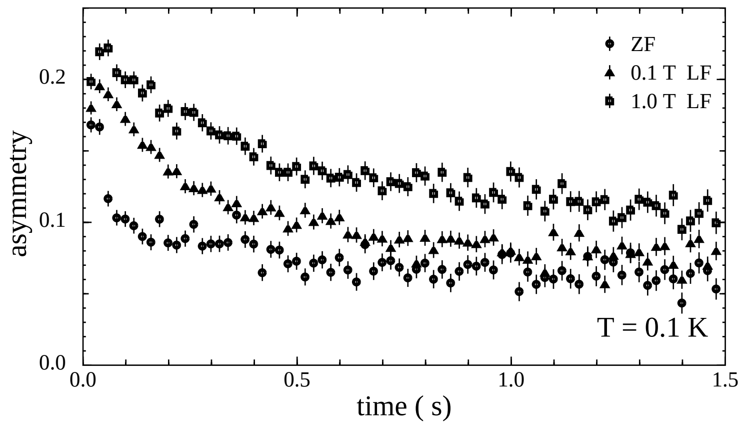

Fig. 5. LF decoupling of $La_{1\cdot 25}Nd_{0\cdot 60}Sr_{0\cdot 15}CuO_4$.

The observed μSR signals are not completely static but reveal fluctuations, as can be seen from LF decoupling experiments (Fig. 5). Even the application of only 1 T results in a partial decoupling. From these data a typical timescale of about 10^{-6} s is derived at $0 \cdot 1$ K which can be interpreted by a residual motion of the stripe structure. Note that this timescale would appear static in neutron diffraction studies.

Fig. 6 shows the peculiar phase diagram for $La_{1\cdot 85-x}Nd_xSr_{0\cdot 15}CuO_4$ where the superconducting transition temperature T_c, the magnetic ordering temperature T_M and the transition temperature T_{LT} from LTT to LTO meet in one point. It is suggestive that the change from superconductivity to magnetic order is related to the pinning of charge and spin domains and that their fast dynamics accompanied with dynamic lattice deformations is an ingredient of the mechanism of superconductivity.

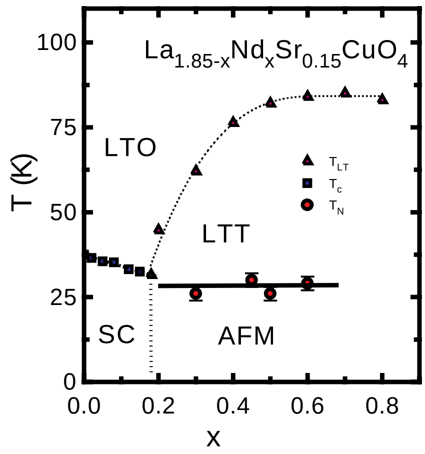

Fig. 6. Phase diagram of $La_{1\cdot 85-x}Nd_xSr_{0\cdot 15}CuO_4$.

We have also performed a series of μSR investigations on the system $La_{1\cdot 8-y}Eu_{0\cdot 2}Sr_yCuO_4$ which has LTT structure between $0 \leq y \leq 0\cdot 3$ at low temperatures. The LTO–LTT transition takes place above 100 K for $y < 0\cdot 23$. $Eu^{3+}(J = 0)$ does not carry a magnetic moment, so no residual magnetic interaction to Cu is expected in contrast to for example the Nd-doped compounds. Even weak Sr-doping ($y < 0\cdot 02$) leads to a rapid suppression of three-dimensional antiferromagnetic order similar to that observed in $La_{2-y}Sr_yCuO_4$ and $Y_{1-x}Ca_xBa_2Cu_3O_6$ (Borsa *et al*. 1995; Niedermayer *et al*. 1997). In the ordered regime μSR reveals spontaneous rotations (Fig. 7). The LTO–LTT transition is visible from a small step in frequency near 130 K (Fig. 8). In the range between about 20 K and $T_N = 190$ K the rotation frequencies are clearly reduced compared to the values found for La_2CuO_4. This can be understood via the fast mobility of the still low number of holes leading to a motional averaging. At low temperatures this dynamics freezes and the full local field at the muon site develops. Note that this increase of local field occurs here as a consequence of a dynamic effect, whereas that observed for LTT $La_{2-x-y}Nd_xSr_yCuO_2$ at lower temperatures is caused by the interaction of Cu and Nd moments.

The freezing of holes results in a spin glassy state which is superimposed on an antiferromagnetic order. For $La_{2-y}Sr_yCuO_4$ and $Y_{1-x}Ca_xBa_2Cu_3O_6$ this spin glass phase is also found and extends beyond $y = 0\cdot 02$ (Niedermayer *et al*. 1997; Torikai *et al*. 1990). For the Eu compound, however, clear spin rotations are visible even for higher Sr doping implying local coherent order. We interpret this order with the onset of static stripe formation. In $La_{2-y}Sr_yCuO_4$ superconductivity

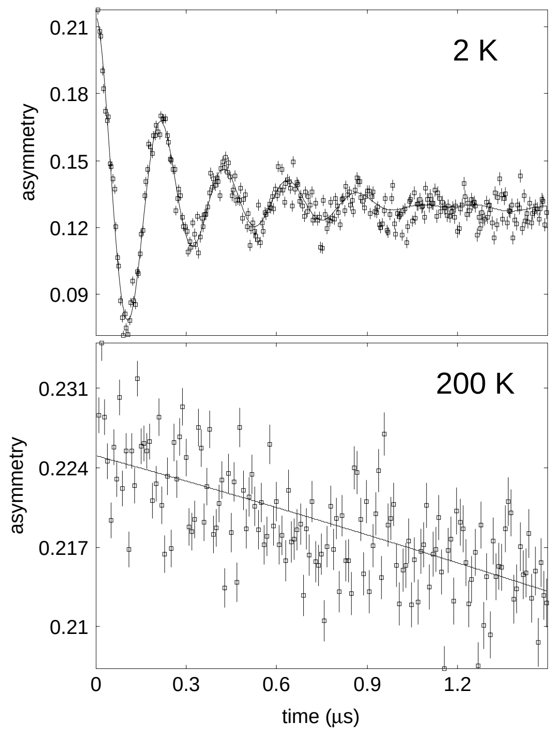

Fig. 7. Asymmetry plots of $La_{1\cdot786}Eu_{0\cdot20}Sr_{0\cdot014}CuO_4$ recorded below and above the onset temperature of magnetic order.

and magnetic correlations coexist for $y \geq 0\cdot06$. Random spin freezing occurs below ~8 K, but the dynamic magnetic correlations extend to nearly 20 K for $y = 0\cdot11$ (Torikai *et al.* 1990). Notably, for the largest $T_c \approx 38$ K, at $y = 0\cdot15$ the magnetic correlations vanish. In the Eu compounds the magnetic ordering temperature rises again for $y > 0\cdot10$ and reaches ~25 K for $y \approx 0\cdot12$ (Fig. 9). Superconductivity is found beyond $y = 0\cdot15$, bulk superconductivity only for $y > 0\cdot17$ with a maximum $T_c = 20$ K around $y = 0\cdot19$. In this concentration range two rotating signals are detected (see Fig. 10), as in $La_{1\cdot85-x}Nd_xSr_{0\cdot15}CuO_4$ which points again to hole-rich and hole-depleted areas. The spin rotations can be decoupled in an applied longitudinal field of $0\cdot2$ T thus proving that there is static order. No upturn of the rotation frequencies is found even for low temperatures. Since Eu carries no moment, no interaction between Cu and Eu will change the spin structure of the CuO planes. Nor is the freezing of hole motion present which we have proposed for low Sr doping. According to the value of asymmetry of the signals, practically the complete sample shows this magnetic response. For the highest Sr concentration studied, $y = 0\cdot2$, differences are found between the μSR signals after field cooling and zero-field cooling. Although this and also the shape of the depolarisation found at low temperature are reminiscent of a spin glass, they are rather caused by flux pinning of the superconducting phase. This sample is bulk superconducting ($T_c \sim 20$ K), but still reveals magnetic correlations up to 15 K. The stripe structure is expected to break down in this concentration range due to the high number of holes.

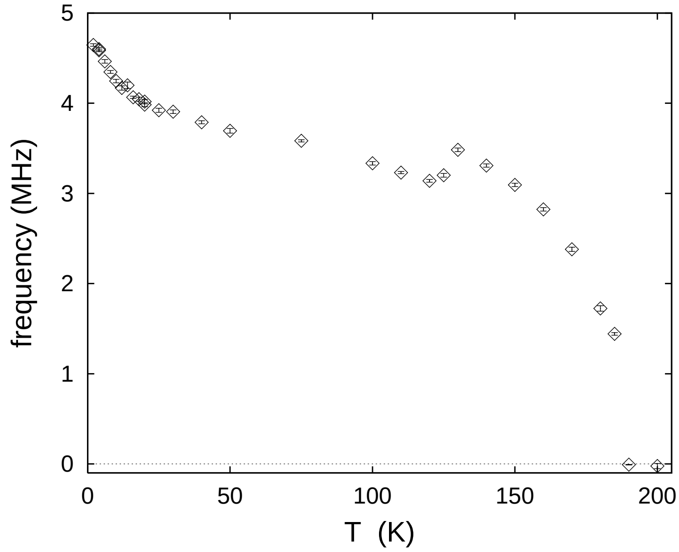

Fig. 8. Temperature dependence of the spontaneous rotation frequency in $La_{1 \cdot 786}Eu_{0 \cdot 20}Sr_{0 \cdot 014}CuO_4$.

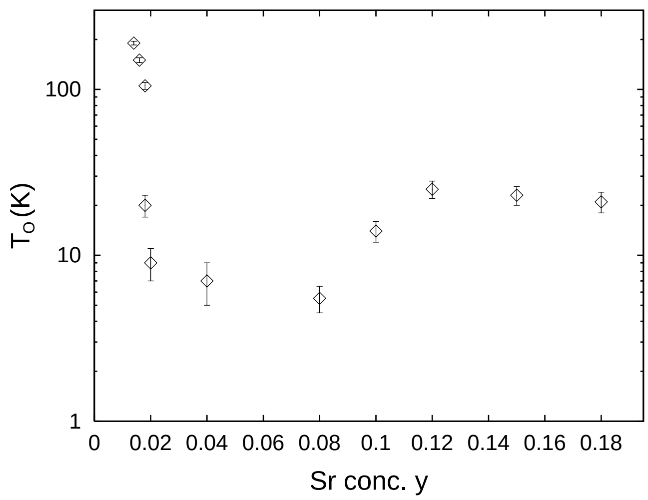

Fig. 9. Onset temperature of magnetic order of $La_{1 \cdot 80-y}Eu_{0 \cdot 20}Sr_yCuO_4$ as a function of Sr concentration y.

From these data it can be seen that stripe correlations and superconductivity can coexist. For the three compounds $La_{2-y}Sr_yCuO_4$, $La_{1 \cdot 8-y}Sr_yEu_{0 \cdot 2}CuO_4$ and $Y_{1-x}Ca_xBa_2Cu_3O_6$ with different structures, the magnetic correlations seem to become suppressed when superconductivity is most strongly developed. The differences in the dynamics of the magnetic correlations for the different materials appear to be related to the different strengths for pinning of the charge and spin domains.

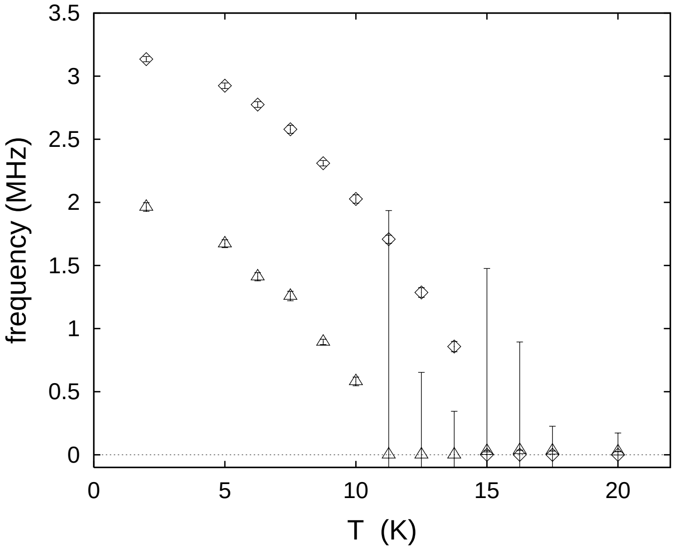

Fig. 10. Temperature dependence of the spontaneous rotation frequencies in $La_{1\cdot70}Eu_{0\cdot20}Sr_{0\cdot10}CuO_4$.

(3b) Magnetic Correlations in $Nd_{2-x}Ce_xCuO_4$

The undoped compound Nd_2CuO_4 reveals an antiferromagnetic order below $T_N = 270$ K with a non-collinear structure of the Cu moments. The magnetic structure undergoes several rearrangements at temperatures above 20 K which can be traced from neutron diffraction (Skanthakumar *et al.* 1993 and references given there) and also μSR (Hillberg *et al.* 1997 and references given there). The spin reorientations are visible from the changes of the frequency of spontaneous rotation (see Fig. 11 where I, II and III signify different non-collinear magnetic phases as characterised from neutron diffraction). From dipolar sum calculations for the known magnetic structures it is most probable that the muon is close to O(2), yet complete agreement between calculations and experimentally observed damping could not be achieved. It has been proposed (Sachidanandam *et al.* 1997) that the origin of the spin reorientation is a subtle interplay between Cu–Cu, Cu–Nd and Nd–Nd interactions of Cu planes with tightly bound neighbouring Nd planes. The increase of the muon rotation frequency below 10 K is accompanied by a strong increase in damping. In this temperature range the ground state Kramers doublet of Nd which is split by the molecular field from the ordered Cu becomes unequally populated and an ordered Nd moment develops. This is also supported by specific heat data revealing a Schottky anomaly (Brugger *et al.* 1993). The observed damping indicates competition between nearest neighbour Nd–Nd antiferromagnetic interactions and the ferromagnetic alignment induced by the Cu–Nd interaction. This competition causes a spin canting of the Nd moments and a broad static field distribution at the muon site similar to the observations made for LTT $La_{1\cdot85-x}Nd_xSr_{0\cdot15}CuO_4$. In contrast, μSR on Pr_2CuO_4 reveals a constant rotation pattern below 30 K (Hillberg *et al.* 1997). There the ground state crystal field level is a singlet carrying only a small induced moment. Thus the effect of the Cu–Pr interaction is negligible. Above 30 K, however, when the higher crystal field states are populated, spin reorientations are also found. Neither the spin reorientations in Nd_2CuO_4 nor those in Pr_2CuO_4 can be traced from magnetisation data. For Pr_2CuO_4 they have even escaped recent neutron scattering analysis (Sumarlin *et al.* 1995). Doping Nd_2CuO_4 with Ce leads to a reduction of the unit cell and a decrease of the CuO_2 plane distances. The electrons additionally available from Ce^{4+} doping reduce in part the magnetic Cu^{2+} to non-magnetic Cu^{1+}. The dilution of the CuO_2 spin system diminishes T_N and for Ce concentrations beyond $x = 0{\cdot}14$ the density of charge carriers in the CuO_2 planes becomes high enough that they become freely mobile and long range antiferromagnetic order is suppressed. Note that this suppression occurs at clearly higher electron concentrations as compared to the weak hole doping which is sufficient in $La_{2-x-y}M_xSr_yCuO$.

The superconductivity of $Nd_{2-x}Ce_xCuO_4$ is carried by electrons. The maximum T_c is about 20 K for $x = 0{\cdot}15$. Also in the superconducting concentration range a specific heat anomaly similar to the undoped compound is found. Upon increasing the doping this anomaly shifts to lower temperatures and a strong linear term T appears below $0{\cdot}3$ K with $\sim$4 J K^{-2} per mole of Nd for $x = 0{\cdot}2$. The high temperature Curie–Weiss susceptibility levels off in this temperature range (Brugger *et al.* 1993). This behaviour resembles that of heavy fermions. Recent magnetoresistance studies also support this (Maiser *et al.* 1997). We shall concentrate in the following on this overdoped compound $Nd_{1\cdot8}Ce_{0\cdot2}CuO_4$.

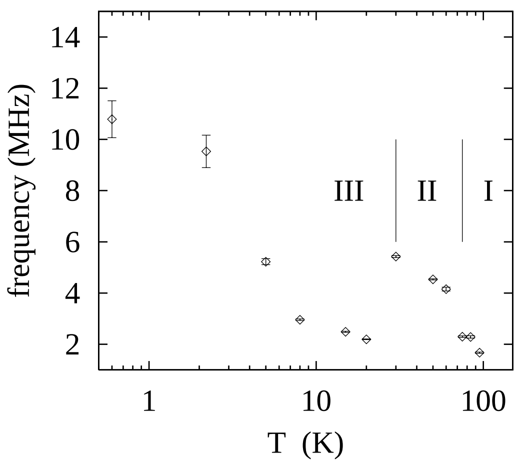

Fig. 11. Spontaneous rotation frequency in Nd_2CuO_4.

In usual heavy fermion systems a lattice of 4f or 5f ions is embedded in a sea of conduction electrons forming singlet states with a typical energy scale of $k_B T^*$ (where T^* is the Kondo temperature). The interaction of conduction electrons is comparatively small. In the presently discussed compound the coupling of Nd to the conduction electrons is, however, weak; i.e. the Nd^{3+}state is well localised and therefore T^* should be very low. In this new type of heavy fermion compound (Fulde and Zevin 1993; Fulde *et al.* 1993) the conduction electrons in the CuO_2 planes are highly correlated, which also causes the antiferromagnetic order in the undoped Nd_2CuO_4. The heavy fermion state is not affected by superconductivity.

Another model stresses the essential role of the contradictory Nd–Nd and Nd–Cu interactions already discussed (Thalmeier 1996; Henggeler *et al*. 1996, 1997). Ce-doping reduces the strength of the Nd–Cu coupling leading to a softening of Nd spin waves consistent with neutron data (Henggeler *et al*. 1997; Loewenhaupt *et al*. 1995, 1996). The connected closing of the spin wave gap thus gives rise to a high density of states at low energy. The ground state would be expected to be a spin glass with a wide distribution of configurations which explains the low temperature specific heat data. Also our muon data indicate a tendency of spin freezing from a distribution of relaxation frequencies leading to a depolarisation function of 'stretched' exponential type $\exp[-(\lambda t)^{\beta}]$. For temperatures above 2 K, β is close to 1, typical for uncorrelated electronic fluctuations causing the muon depolarisation. At lower temperatures β decreases to about 0·8–0·9. Yet even down to 70 mK the electronic spin bath stays dynamic (Fig. 12) with correlation times 2×10^{-9} s as derived from LF decoupling. No significant change of correlation time is detected below ~1 K. This means that neither static magnetic order, nor a randomly frozen spin glass structure is formed down to the lowest temperatures. For this temperature range a heavy fermion behaviour has been concluded from transport and caloric data. As indicated in Fig. 12 the absolute values of damping depend on sample preparation (mainly grain size), which is not, however, the case for the normalised temperature dependence. This points to mesoscopic effects affecting the local field distributions.

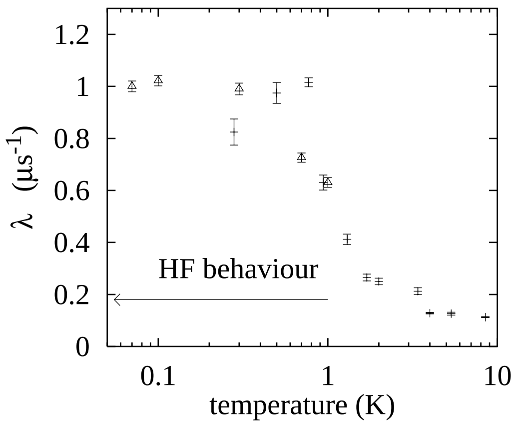

Fig. 12. Muon spin relaxation rate λ under ZF for $Nd_{1\cdot8}Ce_{0\cdot2}CuO_4$. The data from the sintered sample (triangles) are scaled to those from a sol-gel preparation (plusses) by multiplication by a factor of 0·25.

According to our LF experiments this heavy fermion state is not considerably influenced by fields up to 2·5 T, in agreement with specific heat data under magnetic fields. From the decoupling experiments one can also estimate the local field distribution which is mainly caused by the Nd moments. Assuming a random orientation and a μ^+ stopping site near an O(2) atom (Hillberg *et al*. 1997) the Nd moments can be estimated to be of the order of $\sim0\cdot1\mu_{\rm B}$. This is clearly reduced from the value of $\sim1\cdot7\mu_{\rm B}$ expected for the ground state Kramers doublet. In the case of correlated fluctuations this reduced value of the

Nd moments represents the lower limit. This relatively slow spin dynamics in overdoped $Nd_{1\cdot 8}Ce_{0\cdot 2}CuO_4$ also reminds us of the slow residual dynamics of the magnetic domain structure found for $La_{1\cdot 85-x}Nd_xSr_{0\cdot 15}CuO_4$. There, however, and also in the Eu doped La cuprate these domains are ordered and only reveal a very slow fluctuation. For $Nd_{1\cdot 8}Ce_{0\cdot 2}CuO_4$ no spontaneous μSR rotation is found and the fluctuations are faster by three orders of magnitude which may be related to the higher mobility of the doped electrons compared to the holes.

Igarashi *et al.* (1995) calculated fluctuation rates of the exchange field at localised Nd^{3+} produced by the two-dimensional correlated conduction electron system of the CuO_2 planes. At finite temperatures the interaction field at the Nd is fluctuating, but at sufficiently low temperatures these fluctuations are so slow that regions of short-range antiferromagnetic order are formed. In this model the undoped system is antiferromagnetically ordered for $T = 0$. For the doped compounds $Nd_{2-x}Ce_xCuO_4$, long-range order is suppressed by quantum fluctuations even for $T \to 0$ which agrees with our experiments. From a comparison of these calculations with the specific heat data a correlation time for the f-spin fluctuations on the order of about 10^{-11}–10^{-10} s has been derived. Keeping in mind that these calculations include many simplifications concerning the interaction of the 4f system with the conduction electron system, these values are remarkably close to the correlation time derived from μSR.

4. Conclusion

The μSR experiments reveal slow dynamics and also static magnetic correlations in and close to the superconducting concentration range of the hole doped tetragonal T structures of $La_{1\cdot 85-x}Nd_xSr_{0\cdot 15}CuO_4$ and $La_{1\cdot 8-y}Eu_{0\cdot 2}Sr_yCuO_4$. In contrast to the orthorhombic compounds of the $La_{2-y}Sr_yCuO_4$ and 1–2-3 families, coherent magnetic order is found. The heavy fermion-like electron doped $Nd_{2-x}Ce_xCuO_4$ reveals much faster dynamics. Its ground state is non-magnetic. The μSR data also provide evidence that areas with different magnetic response are formed in the Cu–O planes and support a model of stripe domains.

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