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Magnetic order in $La_{1.85-x}Nd_xSr_{0.15}CuO_4$ with $0.30\leqslant x\leqslant 0.60$

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Spontaneous muon spin rotation is observed in muon spin relaxation experiments on $La_{1.85-x}Nd_xSr_{0.15}CuO_4$ with $0.30\leqslant x\leqslant 0.60$, proving the onset of magnetic order below 30 K. The rotation frequencies and the damping rate will be compared to those of La_2CuO_4 and $La_{1.7}Nd_{0.3}CuO_4$. Below ≈ 4 K, there is an increase of the coupling between the moments of Nd and Cu, which can be observed in the muon-rotation frequency and the damping rate. [S0163-1829(97)50126-3]

I. INTRODUCTION

Depending on hole concentration y, rare-earth concentration x, and temperature, four different structural phases can be observed in the high-temperature superconductor (HTSC) system $La_{2-x-y}R_xSr_yCuO_4$ using x-ray diffraction: a high-temperature tetragonal (HTT) phase (space group *I4/mm*), a low-temperature orthorhombic (LTO) phase (space group *Bmab*), a second low-temperature orthorhombic (LTO2) phase (space group *Pccn*), and a low- temperature tetragonal (LTT) phase (space group $P4_2/ncm$).[1,2] Although the changes in lattice parameters are only of the order of 1%, the effects on the electronic ground state are very strong.[3–5]

Both superconductivity and antiferromagnetism have been observed in the system $La_{2-x-y}Nd_xSr_yCuO_4$. A lot of research has been done by many groups to elaborate the whole phase diagram including the electronic order transitions. Büchner *et al.*[3] have given a schematic structural phase diagram: above a Nd concentration $x>0.18$ the structure at low temperatures is no longer LTO. Depending on the Sr concentration, a transition to LTO2 or LTT is observed. The tilting of the CuO_6 octahedra, which depends on both the Sr and the Nd concentration, seems to have an important influence on the electronic properties. Superconductivity is found[3] below a critical tilting angle $\Phi_c=3.6°$. According to Büchner *et al.*,[3] there is no bulk superconductivity in the LTT phase for $\Phi>\Phi_c$. Local antiferromagnetic order was discussed as the electronic ground state in this region.

Based on the observation of superlattice peaks in neutron-scattering experiments on $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$, Tranquada *et al.* have proposed a static stripe correlation model in the LTT phase:[4,5] the LTO-LTT phase-transition temperature is supposed to be accompanied by the onset of charge order. The holes segregate forming a superlattice. In the hole-poor regions the coupling of the Cu spins is strong enough to lead to antiferromagnetic order at lower temperatures. The antiferromagnetic domains are separated by hole-rich domain walls. The spin ordering temperature was found at about 30 K, which is well below the charge ordering temperature.

In the following, we restrict ourselves to a constant Sr concentration of $y=0.15$. The compound $La_{1.85}Sr_{0.15}CuO_4$ (i.e., without Nd) shows only two structural phases, a HTT phase and a LTO phase. Bulk superconductivity appears below $T_c=40$ K. When substituting La by Nd in $La_{1.85-x}Nd_xSr_{0.15}CuO_4$, T_c decreases. A phase transition to the LTT phase occurs at $x=0.18$. No bulk superconductivity is found in this LTT phase.[1] The LTO-LTT transition temperature increases with the Nd concentration (see Fig. 1 and Ref. 6). Mössbauer experiments[7] on iron and tin doped samples of $La_{1.25}Nd_{0.60}Sr_{0.15}CuO_4$ show signs for local magnetic order below about 32 K. This motivated an investigation of the compounds without Fe or Sn dopants using muon spin relaxation (μ^+SR). The results will be discussed in view of the stripe correlation model.[4,5]

II. EXPERIMENT

μ^+SR is a method for microscopically probing the magnetism in a sample. The spin polarized positive muon is implanted in the sample and comes to rest at an interstitial site. During its lifetime ($\tau=2.2\ \mu$s), the spin of the muon pre-

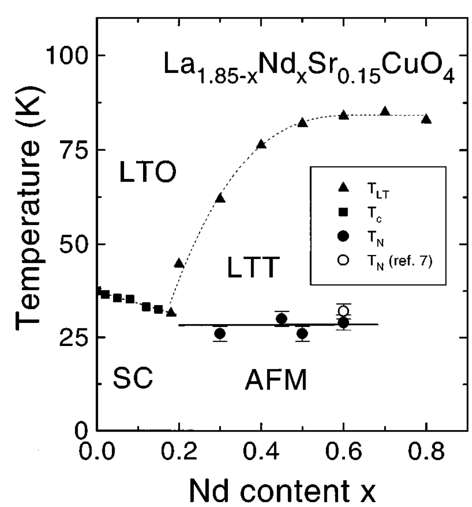

FIG. 1. Phase diagram of $La_{1.85-x}Nd_xSr_{0.15}CuO_4$. The filled circles in the diagram mark the onset temperature T_O of magnetic order as derived from our experiments; the open circle marks T_O as derived from Mössbauer data (see Ref. 7).

cesses in the field produced by its neighborhood or by applied external fields [zero field (ZF) or transverse field (TF) μ^+SR]. Random and fluctuating fields lead to a depolarization of the muon spin resulting in a damping of the signal. When the muon decays a positron is emitted. The emission is spatially anisotropic making it possible to derive the direction of the muon spin at the time of decay. The decay positrons emitted from the sample parallel (N_b) and antiparallel (N_f) with respect to the direction of the initial muon polarization allow us to determine an asymmetry $A=(N_b-N_f)/(N_b+N_f)$, which is plotted as a function of the time elapsed after the implantation of the muon (see Fig. 2). The asymmetry of the counts in two detectors on opposite sides of the sample is plotted versus the time elapsed since implantation of the muon. Thus μ^+SR allows the measurement of magnetic distributions and fluctuations. Decoupling experiments in applied longitudinal fields (LF μ^+SR) can distinguish between static and dynamic magnetic moments (in the time window of the μ^+SR) and the size of the static moments can be estimated.

Powder samples of $La_{1.85-x}Nd_xSr_{0.15}CuO_4$ with x ranging from 0.30 to 0.60 were prepared by standard solid-state reaction.[8] Their structural and superconducting properties have already been published.[1] ZF, LF, and TF μ^+SR experiments were performed at the GPS and LTF spectrometers of PSI, Switzerland. The temperature was varied between 0.1 K and 100 K; fields up to 1 T were applied.

III. RESULTS

The contribution to the depolarization of the muon spin which is caused by quasistatic nuclear spins in the surroundings of the muon was found to be compatible with the apical oxygen muon site. This is the expected muon site (see Ref. 9). Below 100 K, the depolarization due to electronic spins slowly increases with decreasing temperature indicating a slowing down process of the electronic fluctuations. The structural phase transition (LTO-LTT) taking place at about 75 K as confirmed by x-ray diffraction[1] is not reflected in our experiments.

Below 30 K, spontaneous muon spin rotations are observed (see Fig. 2). The frequency distribution of the μ^+SR spectra is rather broad. In powder samples the direction of the field at the muon site is isotropically distributed, which was taken into account by the fit function we used:

$$A(t)=A_0 D_{\mathrm{GKT}}(t)\left[\tfrac{2}{3}e^{-R_2 t}\cos(2\pi\nu t)+\tfrac{1}{3}e^{-R_1 t}\right]. \quad (1)$$

The Gaussian-Kubo-Toyabe function $D_{\mathrm{GKT}}(t)$ stands for the nuclear depolarization; the term in brackets describes the electronic part, i.e., the transverse component rotating with the frequency ν and damped with the transverse relaxation rate R_2 and the longitudinal component damped with the longitudinal relaxation rate R_1. A fit with only one powder signal resulted in very poor agreement with the spectra just below the onset temperature of the muon spin rotations. Incommensurabilities of the spin structure are expected to result in a Bessel function instead of the cosine in Eq. (1) (compare Ref. 10). We found, however, that the Bessel function did not improve the fit. Two of the powder signals [of the form given in Eq. (1)] had to be used to satisfactorily fit the spectra in the whole temperature range below 30 K. The asymmetries of the two signals could be kept constant for all the spectra of each sample. These asymmetries, the rotation frequencies ν at 10 K, and the estimated ordering temperatures T_O are given in Table I. The temperature dependence of the rotation frequencies for the samples $La_{1.4}Nd_{0.45}Sr_{0.15}CuO_4$ and $La_{1.25}Nd_{0.60}Sr_{0.15}CuO_4$ is shown in Fig. 3. When decreasing temperature, the transverse damping rate R_2 of the rotating signal rises from about 0.3 μs^{-1} at

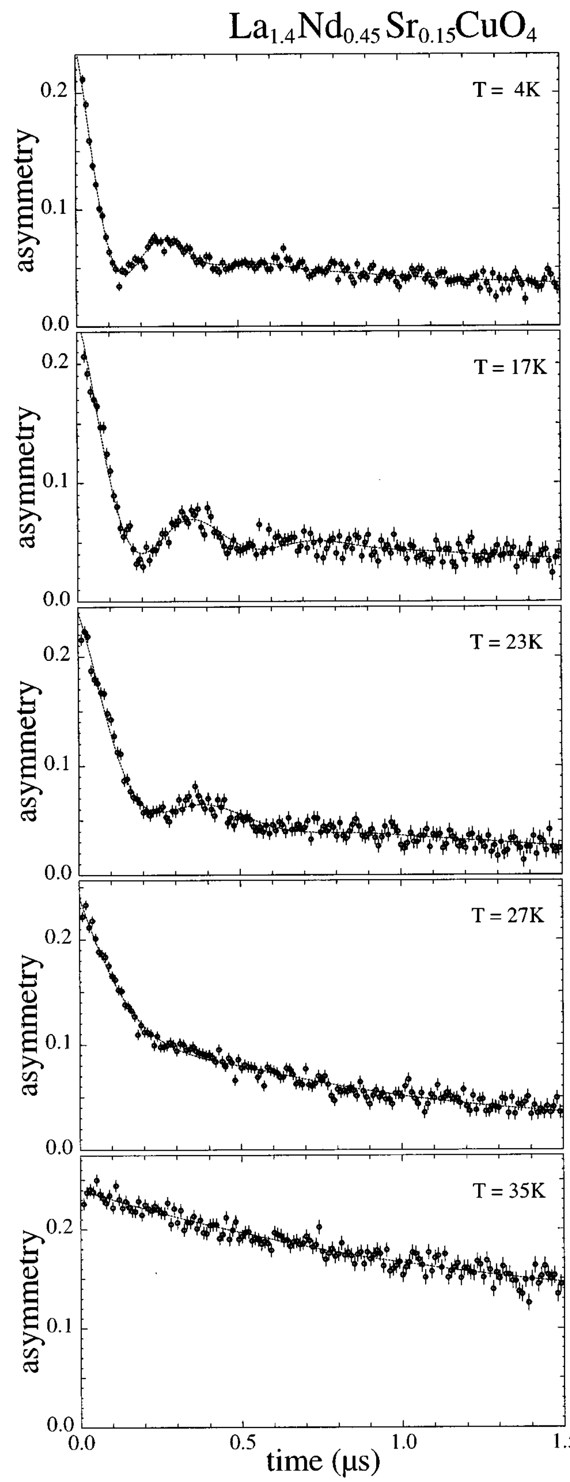

FIG. 2. Asymmetry plots of $La_{1.4}Nd_{0.45}Sr_{0.15}CuO_4$ at different temperatures.

TABLE I. Muon spin rotation frequencies at 10 K, asymmetries of the two signals, and estimated ordering temperatures T_O.

Nd content	1st signal		2nd signal		Ordering temp.
x	freq. (MHz)	asym.	freq. (MHz)	asym.	T_O (K)
0.30	2.80±0.05	0.10±0.01	1.12±0.07	0.11±0.01	26±2
0.45	2.95±0.03	0.15±0.01	1.25±0.09	0.08±0.01	30±2
0.50	2.95±0.05	0.04±0.01	1.70±0.05	0.11±0.01	26±2
0.60	3.15±0.06	0.05±0.01	2.00±0.10	0.09±0.01	29±2

≈35 K to more than 5 μs^{-1} at T_O. There is a broad hump in the region of T_O and R_2 only slowly decreases with temperature.

Below 5 K, a further increase is found in the muon rotation frequencies (see Fig. 3) and in the damping rates. This effect is more pronounced for the samples with higher Nd concentrations. Applying longitudinal fields up to 1 T at 0.1 K does not lead to a complete recovery of the polarization (see Fig. 4). This reveals that even at 0.1 K the electronic spin system does not become totally static.

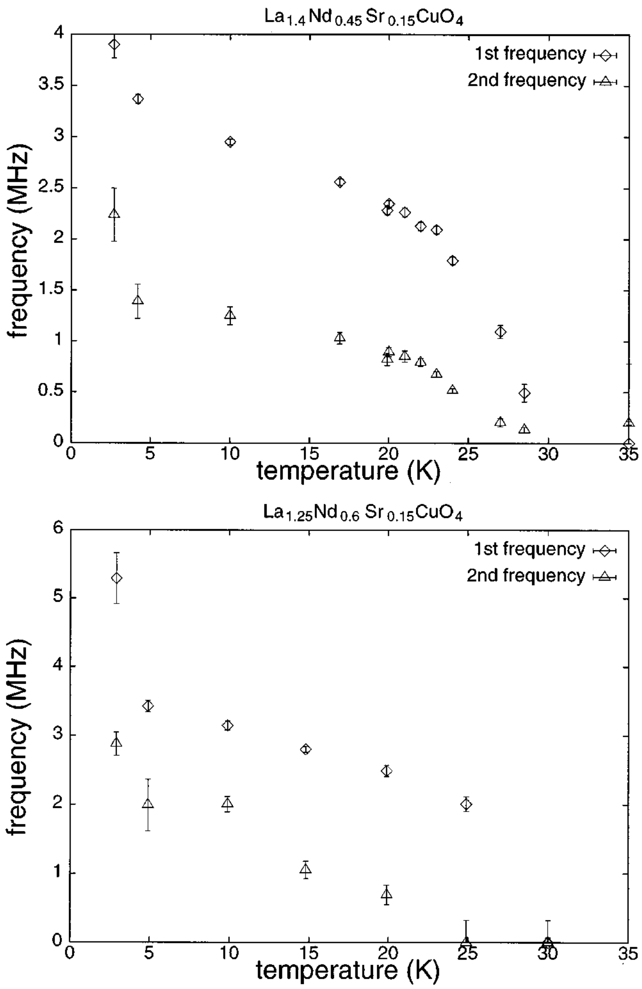

FIG. 3. Temperature dependence of the two muon spin rotation frequencies for the samples $La_{1.4}Nd_{0.45}Sr_{0.15}CuO_4$ and $La_{1.25}Nd_{0.60}Sr_{0.15}CuO_4$.

IV. DISCUSSION

The observation of spontaneous muon spin rotations proves the onset of magnetic order in $La_{1.85-x}Nd_xSr_{0.15}CuO_4$ with $0.30 \leqslant x \leqslant 0.60$. The onset temperature T_O of magnetic order is 28±2 K for all Nd concentrations. This is in good agreement with the T_O found for Fe and Sn doped samples of $La_{1.25}Nd_{0.60}Sr_{0.15}CuO_4$ using Mössbauer spectroscopy.[7] The antiferromagnetic order in $La_{1.85-x}Nd_xSr_{0.15}CuO_4$ is restricted on the LTT phase ($x \geqslant 0.18$), but the onset temperature T_O is independent of the Nd concentration x (see phase diagram in Fig. 1). Particularly, we want to point out that the magnetic order is not bound to the Sr concentration of $y=\frac{1}{8}$. Our value for T_O also roughly agrees with the value found for $La_{1.48}Nd_{0.40}Sr_{0.12}CuO_4$ using neutron scattering.[5] For $La_{1.475}Nd_{0.4}Sr_{0.125}CuO_4$, Luke *et al.* report a value of $T_O \approx 30$ K determined by μ^+SR experiments.[11]

The analysis of our μ^+SR experiments shows that within an uncertainty of about 5% every implanted muon is exposed to a static hyperfine field. Thus the magnetic order is not restricted to certain clusters. Superconducting regions of the sample should show a weakly dampened nonrotating signal. Such a component is not found in our data. Therefore, only spurious superconductivity can be present in our samples. A comparison of the muon rotation frequencies in La_2CuO_4 (LTO phase) (Refs. 12 and 13) and $La_{1.7}Nd_{0.3}CuO_4$ (LTT phase) (Ref. 14) reveals that in $La_{1.85-x}Nd_xSr_{0.15}CuO_4$ the frequency is remarkably reduced. For La_2CuO_4, a value of 5.6–5.7 MHz was reported; experiments on $La_{1.7}Nd_{0.3}CuO_4$ show a frequency of about 4.9 MHz. The average frequency in our samples with Nd and Sr dopants is smaller than 2.5 MHz. Another interesting feature is the temperature dependence of the damping rate: at T_O, $La_{1.7}Nd_{0.3}CuO_4$ shows a sharp peak in the damping rate. This is typical for a magnetic second-order phase transition. The Sr doping in $La_{1.85-x}Nd_xSr_{0.15}CuO_4$ seems to smear out this sharp peak resulting in a broad hump in the temperature plot of the damping rate.

The fact that a fit with only one powder signal resulted in poor agreement with the spectra in the temperature range between 20 and 30 K, is caused by a distribution of the magnetic field at the muon site. To keep the number of parameters as small as possible we restricted ourselves to a fit with two powder signals. This fit strategy turned out successful, since the relative weight of both signal contributions could be fixed for the temperature range below 35 K. These two contributions can be interpreted in the stripe model of Tranquada *et al.*:[4,5] the antiferromagnetic order is restricted to

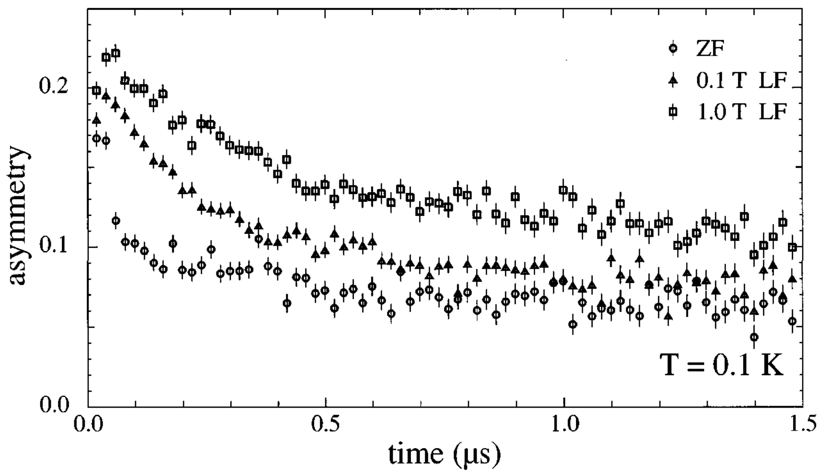

FIG. 4. LF decoupling experiment on $La_{1.25}Nd_{0.6}Sr_{0.15}CuO_4$ at $T=0.1$ K.

small domains separated by hole-rich domain walls. Signal 1 is tentatively attributed to the muons in the antiferromagnetic domains, whereas signal 2 to those in the less magnetic wall areas. Both signals are severely affected by inhomogeneous broadening due to the distribution of the magnetic field ranging between the values for walls and domains. Notably, in $La_{1.85-x}Nd_xSr_{0.15}CuO_4$, there are still spin fluctuations down to 0.1 K as shown by the LF decoupling experiments. The estimated electronic correlation times are of the order of 10^{-6} s. This indicates a still finite fluctuation rate of the Nd and/or Cu spins similar to $Nd_{1.8}Ce_{0.2}CuO_4$ (see Ref. 15).

We finally report on our observations at very low temperatures: below ≈ 4 K, a further increase of the rotation frequencies and of the transverse damping rate R_2 is found. This coincides with an increase of the signal intensity of the magnetic peaks in neutron scattering.[5] An increase of the muon spin rotation frequency means an increase of the average of the magnetic field at the muon site. An ordering of the Nd moments due to Nd-Nd interactions can be excluded due to the large distances. We rather propose an increase of the Nd-Cu interactions leading to a slowing down of the fluctuations of the Nd moments accompanied by a tilting of the Cu moments. This picture agrees with the neutron-scattering data of Tranquada *et al.*:[5] below 3 K, a strong increase of the intensity of the magnetic superlattice peaks has been found. Experiments with different directions of the incident neutron beam on the single crystal were interpreted as a rotation of the orientation of the magnetic moments. The Cu moments, which lie in the a-b plane above 5 K as in undoped La_2CuO_4, are assumed to rotate towards the [001] direction due to a stronger coupling with the Nd moments. In μ^+SR experiments on Nd_2CuO_4, a similar increase of the muon spin rotation frequency was found below about 10 K, which is also related to a reorientation of the Cu moments due to their coupling to Nd.[15] For Pr_2CuO_4 in contrast, no change is found due to the only small induced moment at Pr.

V. CONCLUSION

Our μ^+SR experiments have proved the existence of magnetic order in the Cu-O planes in the system $La_{1.85-x}Nd_xSr_{0.15}CuO_4$ with $0.30 \leqslant x \leqslant 0.60$. The onset temperature T_O was found to be independent of the Nd concentration x. This leads to the rather peculiar phase diagram (see Fig. 1), where T_c, T_N, and $T_{LTO \rightarrow LTT}$ appear to meet in a single point. Both the existence of static magnetic order as well as the distribution of the internal fields with a reduced average value can be understood in the picture of stripe correlations.[4]

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