From Antiferromagnetic Order to Static Magnetic Stripes: The Phase Diagram of $(La,Eu)_{2-x}Sr_xCuO_4$

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The magnetic order of $(La,Eu)_{2-x}Sr_xCuO_4$ ($x \leq 0.2$) has been investigated with μSR techniques. In this system a low temperature tetragonal (LTT) structure is present in the entire range of doping and it is possible to follow the evolution from the long range antiferromagnetic state at $x = 0$ to the static magnetic stripes. We find a nonmonotonic change of the Néel temperature with increasing x and the obtained magnetic phase diagram of the LTT phase resembles the generic phase diagram of the cuprates where the superconductivity is replaced by a second antiferromagnetic phase.

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Spatial modulations of the charge density seem to be a generic feature of doped transition metal oxides, such as cuprates, nickelates, and manganites [1–3]. In particular, there is growing evidence that so called stripe correlations of spin and charge are crucial for the understanding of the physics of the cuprate high temperature superconductors (HTSC) [4]. In this class of compounds static spin and charge stripes are inferred from neutron and x-ray diffraction investigations of the low temperature tetragonal (LTT) phase [1] which is observed in rare earth doped $La_{2-x}Sr_xCuO_4$ (LSCO) [5]. It is well known that the small structural differences between the orthorhombic (LTO) phase of LSCO and the LTT phase have a drastic influence on the electronic properties [5]. Superconductivity (SC) is suppressed in the LTT phase and static antiferromagnetism (AF) occurs in compounds with large hole content. It has been argued that the LTT structure provides a pinning potential for the stripe motion and the changes of the electronic properties at the LTO to LTT phase transition are attributed to a change from dynamic to static stripes [1,3].

These observations are further examples which clearly show the proximity of superconductivity and AF in the cuprates. Usually, this proximity is discussed in connection with the generic phase diagram of the HTSC exhibiting a change from an antiferromagnetic to a superconducting ground state with increasing charge carrier content. It is argued that the properties of the HTSC are determined by quantum critical behavior and, moreover, a close connection of AF and SC in terms of a SO(5) theory has been suggested [6].

It is apparent that the LTT phase of doped La_2CuO_4 markedly differs from the generic phase diagram of the HTSC, since static stripe AF is also present for high carrier content. This stripe AF is well established for $x \sim 0.12$ from studies of Nd doped LSCO [1,7,8]. For smaller x yet another structural modification occurs in Nd doped LSCO inhibiting studies of a strongly underdoped LTT phase. However, the LTT phase is present in the entire range of Sr concentrations $0 \leq x \leq 0.25$ when doping LSCO with smaller Eu ions. In these systems it is possible to investigate the change from the commensurate AF at $x = 0$ to the stripe AF at large x. So far, magnetic properties of $(La,Eu)_{2-x}Sr_xCuO_4$ (LESCO) have been studied with magnetization, ESR and NMR measurements [9,10]. In the LTT phase a drastic slowing down of the spin fluctuations is revealed [9], which depends nonmonotonically on the Sr doping and is most pronounced for $x \simeq 1/8$ [11].

In this Letter we present a systematic muon spin rotation (μSR) study of Eu doped LSCO. Our measurements yield two very surprising features: (i) In the LTT phase the magnetic ordering temperature T_N depends nonmonotonically on the hole concentration. (ii) Even more spectacular, the obtained charge carrier concentration dependence $T_N(x)$ for $x \geq 0.05$ is very similar to the superconducting $T_c(x)$ in the LTO phase. Thus, the magnetic phase diagram resembles the generic phase diagram of the HTSC where superconductivity is replaced by a second antiferromagnetic phase.

For our study we have used well characterized, carefully annealed samples with the composition $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$ ($0 \leq x \leq 0.2$) [9,11,12]. For all samples x-ray diffraction data show an LTT structure at low temperature [13] and the LTO → LTT transition temperature $T_{LT} \simeq$ 125 hardly changes as a function of x. As discussed in Ref. [11] SC is destroyed in underdoped LESCO and there is a broad crossover to a superconducting LTT phase for large x which is determined by the decreasing tilt distortion Φ. If Φ is smaller than a critical value $\Phi_c \simeq 3.6°$, there is no significant difference between the superconducting properties of LTO and LTT phases (see also [5]). For the samples studied here this broad crossover occurs at $x \simeq 0.19$. Superconductivity is absent for $x < 0.15$ whereas for higher doping traces of SC are observed in resistivity and susceptibility measurements [11]. Zero-field, longitudinal-field, and transverse-field μSR experiments have been performed at the M13 and M15 spectrometers of TRIUMF, Canada, and at the GPS and LTF spectrometers of Paul Scherrer Institute, Switzerland.

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In Fig. 1 we show representative zero field μSR time spectra of $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$ for Sr concentrations $0 \leq x \leq 0.2$. A striking nonmonotonic Sr concentration dependence of the static magnetic order is apparent. For $x = 0$ and $x = 0.014$ a clear precession of the time evolution of the muon spin polarization is visible which proves the presence of a static magnetic field at the muon site. This precession is absent for higher doping $0.02 \leq x \leq 0.08$. For $x = 0.02$ a strong decay of the muon polarization signals the proximity of a spin freezing, whereas there is no indication for static magnetic order at 10 K for $x = 0.04$ and $x = 0.08$. However, clear signatures of static magnetic order reappear in the μSR spectra for higher Sr concentrations. A strong decay and a precession is visible in the spectrum for $x = 0.1$ at $T = 10$ K and the signatures of magnetic order are even more pronounced for $x = 0.12$ and $x = 0.15$. Further increasing the doping suppresses the magnetic order and for $x = 0.2$ we find only an onset of spin freezing at $T = 8$ K. Note that the two phase behavior reported in Ref. [14] for $x = 0.15$ does not occur in our samples with a larger Eu content, i.e., a more stable LTT phase.

Whereas the data discussed so far show that the electronic properties of the LTT and LTO phases markedly differ at high hole doping, the suppression of magnetic order for small Sr concentrations is very similar. This is illustrated in Fig. 2, where we compare our observations to the findings in LSCO. Neither for the transition temperatures to the long range AF state at small Sr doping $x \leq 0.02$ nor for the concentration range $0.02 \leq x \leq 0.05$ with short range static magnetic order we do observe any significant differences between the LTT and LTO phases. However, drastic differences show up upon further increasing the doping. Whereas the ordering temperature as determined from μSR data decreases monotonically in LSCO, we find an increase of T_N with increasing hole content in LESCO. For a doping of $x = 0.08$ T_N is already twice as large as the ordering temperature inferred from μSR data on LSCO and this difference further increases with increasing x.

We turn now to the analysis of the μSR spectra. For all samples the low T spectra are analyzed without a significant nonmagnetic signal fraction [18]. Comparing the spectra for $x = 0$ and $x \simeq 0.12$ in Fig. 1 reveals two main differences. The precession frequency is apparently much smaller in the doped system showing a smaller magnetic field at the muon site on the one hand. On the other hand there is much stronger damping of the precession in the doped system revealing a substantial spatial inhomogeneity of the sublattice magnetization. A closer inspection of the data reveals that it is impossible to fit the spectra for $x > 0.04$ assuming a single precession frequency. As has been discussed in Ref. [7] a better fit is obtained assuming a superposition of at least two precessing spectra [19]. The larger frequency ν of the two frequencies obtained in this "two spectra analysis" is clearly visible in the Fourier transformed *raw data*. It reflects a well developed peak, whereas the second signal is necessary only in order to describe the asymmetric shape of the frequency peak with a slowly decreasing low-frequency tail (see the Fourier spectrum for $x = 0.18$ in the inset in Fig. 3). In the following we consider only the larger frequency ν, which gives

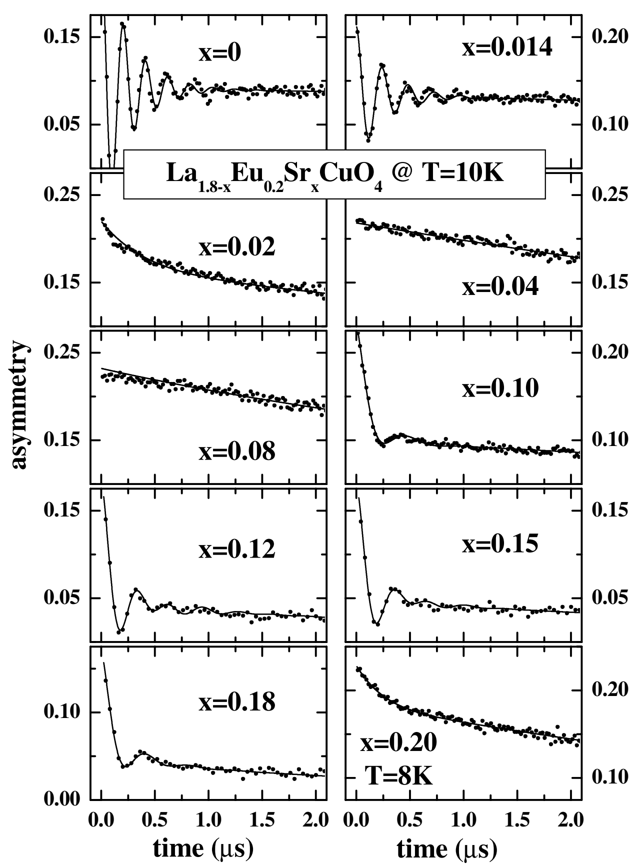

FIG. 1. Zero field μSR spectra at $T = 10$ K for $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$ with $0 \leq x \leq 0.2$. The solid lines are fits to the data (see text).

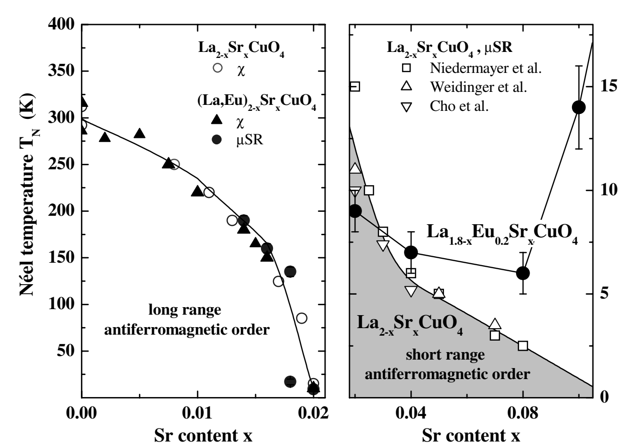

FIG. 2. Magnetic ordering temperatures T_N in lightly hole doped $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$ (filled symbols) and $La_{2-x}Sr_xCuO_4$ (open symbols). Left: Long range Néel state for $x \lesssim 0.02$ as signaled by the susceptibility χ and μSR. Right: Short range order for $0.02 \leq x \leq 0.1$ as revealed from μSR. The data for $La_{2-x}Sr_xCuO_4$ were taken from Refs. [15–17].

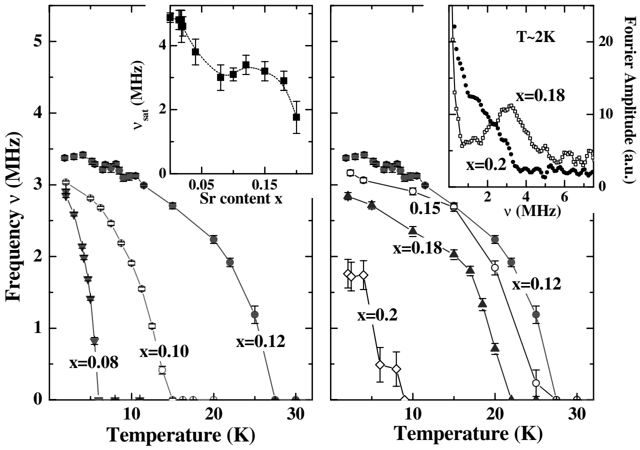

FIG. 3. Temperature dependence of the μSR frequency ν in $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$ for $0.08 \leq x \leq 0.2$. Left inset: Saturation frequency ν_{sat} as a function of x. Right inset: Comparison of the Fourier transformed μSR spectra for $x = 0.18$ and $x = 0.2$ at $T \simeq 2$ K.

an appropriate measure for the magnetic field at the muon site and thus allows the staggered magnetization of the Cu spins to be determined.

The temperature and doping dependence of ν is shown in Fig. 3. For the samples with $0.08 \leq x < 0.18$ the sublattice magnetization follows a typical temperature dependence of an order parameter and does not show any anomalies at low T. In particular, there is no further increase of ν at $T \ll T_N$, which is observed in the Néel state of lightly doped LSCO ($x \leq 0.02$) (see, e.g., [20]). Moreover, our data do not give any evidence for a reorientation of the Cu spins at low T which has been inferred from neutron diffraction data on Nd doped LSCO [21]. Again in contrast to the findings for the LTT phase of Nd doped compounds [7] decoupling experiments in longitudinal fields reveal that the spin system in LESCO becomes static in the time window of μSR. We thus conclude that in Nd doped LSCO both the remaining magnetic dynamics as well as the Cu spin reorientation are due to the magnetic moments of the Nd ions and their interaction with the Cu spins.

For the Eu doped system the frequencies for $T \to 0$ (ν_{sat}) are very similar in the concentration range $0.08 \leq x < 0.18$ and taking into account the different ordering temperatures the data indicate a nearly concentration independent $T = 0$ sublattice magnetization in this range of doping. Though T_N is an order of magnitude smaller than for undoped $(La,Eu)_2CuO_4$ and though there are many mobile holes [12] the values of ν_{sat} amount to about 70% of the frequency found in the Néel state for $x = 0$. These rather large ν_{sat} are qualitatively consistent with the calculations of the magnetic properties of CuO_2 planes with a static stripe order in Ref. [22].

A drastic change of the μSR spectra is found when slightly increasing the hole content from $x = 0.18$ to $x = 0.2$. T_N drops substantially (see Fig. 1) and, in addition, the low T spectra for $x = 0.2$ differ drastically from those found for smaller doping. Using the same fit as for smaller x reveals a reduced $\nu_{sat} \simeq 1.7$ MHz. However, there is not only a reduction of ν_{sat}. For $x = 0.2$ the μSR spectra do not show a clear precession even at 2 K and there is no well defined frequency as displayed in the right inset in Fig. 3. Whereas a clear peak at $\nu \simeq 3$ MHz is visible in the Fourier transformed spectrum for $x = 0.18$, there is only a broad distribution of precession frequencies for $x = 0.2$. It is apparent that the magnetic properties of $La_{1.8-x}Eu_{0.2}Sr_x$-CuO_4 drastically change at a hole doping of $x \sim 0.2$. We stress that in the same range of doping there is also a pronounced change of SC. For samples showing a well developed magnetic order in the μSR data susceptibility measurements exhibit reduced shielding signals (see, e.g., the data for a smaller Eu content $y = 0.17$ in Ref. [11]). We mention that these pronounced changes of AF and SC correlate with the critical tilt angle Φ_c. They are found at $x < 0.2$ in samples with smaller Eu content, i.e., for smaller tilt angle [23]. Our results for $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$ are summarized in the phase diagram in Fig. 4. For all samples we find a low temperature structural transition at $T_{LT} \gtrsim 125$ K and a LTT-like structure at low temperatures [13] whereby the tilt angle Φ decreases monotonically with x. At low Sr doping a strong suppression of the AF is observed, as in the LTO phase. The increase of the ordering temperature for larger x strongly suggests that one has to discriminate clearly between the AF state at very low doping and the static magnetism at moderate and high doping.

It is straightforward to discuss the nonmonotonic behavior of the magnetic order within the stripe scenario. For very small x the Coulomb repulsion prevents the formation of stripes. In this range of doping the suppression of AF

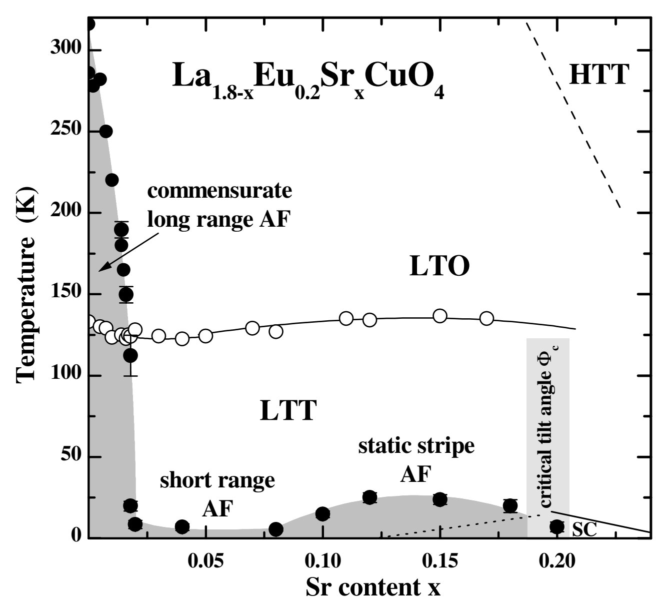

FIG. 4. Phase diagram of $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$. Full and open circles denote the magnetic and the structural (LTO $\to$ LTT) transition temperatures, respectively. The structural transition from the high temperature tetragonal to the LTO phase is indicated by the dashed line. The solid (dotted) line marks the region with strong (weak) diamagnetic signals due to superconductivity (see Ref. [11]).

can be qualitatively explained by the presence of individual holes with large mobility [24] for both LTO and LTT phases. Neutron scattering gives evidence for dynamic stripes along the [100]/[010] directions in the LTO phase of LSCO for Sr contents $x > 0.05$ [25] and in this range of doping we find the increase of T_N in the LTT phase (see Fig. 2) with increasing Sr content. It is thus quite natural to ascribe the increase of T_N with increasing x to the pinning of stripe correlations. The nonmonotonic $T_N(x)$ is thus a consequence of the usual suppression of AF at low doping on the one hand and the formation of static stripes for sufficiently large x on the other hand. It is also straightforward to interpret the disappearance of AF at large x, i.e., at the critical tilt angle, within this scenario. With increasing x the tilt distortion in the LTT phase decreases and thus one expects a "depinning" of stripes, i.e., the disappearance of static stripe AF for large x (see, e.g., [3]). Summarizing this discussion we find that the phase diagram of the LTT phase in Fig. 4 strongly supports the stripe picture, though our μSR data do not give a direct confirmation of a spatially modulated AF in the form of stripes.

We now turn to the most surprising result of our study. Comparing the phase diagrams of pure and Eu doped LSCO yields a striking similarity. The superconducting phase of LSCO seems to be replaced by the stripe AF in LESCO. In the LTO phase superconductivity occurs for $x > 0.05$ and T_c increases with increasing hole content. In the same range of hole doping we observe an increase of the static magnetic order in the LTT phase and even the ordering temperatures T_N compare well with T_c found in the LTO phase, i.e., $T_N^{\mathrm{LTT}}(x) \sim T_c^{\mathrm{LTO}}(x)$. Thus, by changing the structure from LTO to LTT it is possible to switch from SC to AF with a nearly unchanged critical temperature. For $x = 0.15$ this switching at the same critical temperature has also been observed in Nd doped LSCO [7]. Moreover, the phase diagram (Fig. 4) shows that the change from SC to AF does not only occur due to the LTO $\rightarrow$ LTT transition, but also within the LTT phase. For $x \geq 0.20$ the magnetic transition is replaced by bulk SC and again, the observed T_c corresponds to the extrapolated T_N. Further studies of this crossover region are in progress to discriminate the influence of hole doping [26] and tilt distortion [5].

The magnetic phase diagram of the LTT phase with its striking similarity to the generic phase diagram of the HTSC is the main result of our study. Our data clearly show the proximity of AF and SC. Often this proximity is inferred from the generic phase diagram and it is argued that the hole doping determines whether a system is superconducting or magnetic. Our data show that it is possible to switch the entire hole concentration dependent phase diagram from SC to AF whereby the crucial parameter is a small structural deformation of the CuO_2 layers. Moreover, the phase diagram in Fig. 4 suggests that it is necessary to discriminate between the static AF at low doping and the stripe AF for larger x. It is the latter AF phase which is closely related to superconductivity.

Comparing the electronic phase diagrams of the LTO and the LTT phases it seems worthwhile to investigate theoretically, e.g., in terms of a SO(5)-like theory, a switching between antiferromagnetism and superconductivity at constant hole doping. Moreover, from Fig. 4 it is apparent that a proximity of antiferromagnetic and superconducting ground states is not restricted to a single hole doping. In contrast, our data suggest that this proximity is important for a discussion of the electronic/magnetic properties of the cuprates in the entire superconducting region.

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