# Observation of Magnetic Ordering in Superconducting $La_{2-x}Sr_xCuO_4$ by Muon Spin Rotation

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Muon spin rotation experiments performed on superconducting $La_{2-x}Sr_xCuO_4$ samples clearly show that internal magnetic fields coexist at low temperatures ($T < 2$ K) with superconductivity for $x \leq 0.15$. The magnetic fields in the superconducting state are an order of magnitude smaller than the corresponding fields in undoped $La_2CuO_4$. The data are discussed in the context of a magnetic pairing mechanism in high-$T_c$ superconductors.

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There is growing evidence that magnetic interactions play an important role in high-$T_c$ superconductivity.[1-11] Experimentally it was shown that slight changes in the stoichiometry of the high-$T_c$ compounds cause a transition from superconducting to magnetically ordered phases and vice versa. For example, $La_{2-x}Sr_xCuO_4$ is antiferromagnetic for small $x$ values[3-5] and becomes superconducting for $x \geq 0.07$. Similar effects were found for $YBa_2Cu_3O_x$ as a function of the oxygen content.[6]

Theoretically the close relation between antiferromagnetic ordering and superconductivity was established already in the early stages of high-$T_c$ superconductor research.[7-9] Recently, magnetic fluctuations were introduced explicitly as the cause for the pairing of charge carriers in the superconducting state.[10,11] Aharony and co-workers presented a model in which the energy for the formation of pairs is provided by a local approach of the charge carriers, thereby reducing the frustration introduced by each carrier in the antiferromagnetic ordering. A similar mechanism for pairing was proposed by Schrieffer, Weng, and Zhang[10] in the spin-bag model. In these theories, antiferromagnetic correlations are a necessary condition for the appearance of superconductivity.

In this context the experimental verification of the coexistence of magnetism and superconductivity is very important. Here, we should distinguish between the simple coexistence in systems like GdBaCuO where magnetism and superconductivity are associated with electrons which are more or less spatially separated and the coexistence of magnetism and superconductivity in systems like LaSrCuO where the superconducting and magnetic electrons are intimately related and strongly coupled.

The first indication of magnetic correlations in superconducting $La_{1.85}Sr_{0.15}CuO_4$ was reported by Gutsmiedl, Wolff, and Andres.[12] These authors found a nuclear contribution to the low-temperature specific heat which they attributed at least in part to the magnetic splitting of nuclear ground-state levels in an internal magnetic field. Brewer *et al.*[6] reported the onset of magnetic ordering below the superconducting transition temperature in $YBa_2Cu_3O_{6+x}$ in the concentration range around $x = 0.4$ but no details on the internal magnetic fields were given.

In the present paper we present muon spin rotation ($\mu$SR) results on internal magnetic fields in $La_{2-x}Sr_xCuO_4$ with Sr concentrations at which the samples are superconducting. Since the experiments were performed in zero external field and since the samples were cooled in zero field to avoid flux trapping, any appearance of a magnetic interaction must be due to intrinsic magnetic fields in the samples and thus gives an unambiguous signature of the existence of magnetic moments.

The experiments were performed at the Low Temperature Facility of the Paul-Scherrer-Institut in Switzerland. At this installation, which contains a $^3He$-$^4He$ dilution refrigerator, $\mu$SR experiments at temperatures between 35 mK and 4.2 K can be made. The temperature was measured with a calibrated Ge diode and a calibrated carbon resistor in the mixing chamber, and for the low temperatures with a $^{60}Co$ nuclear thermometer on the sample holder. The samples were tightly pressed onto the Cu cold finger with cork pieces and a Cu frame.

The samples were prepared in the usual way by sintering the appropriate chemicals. The final pellets of 12 mm diam and 1 mm thickness were single phase and had superconducting transition temperatures (midpoint) at 14 K for $x = 0.07$, at 26 K for $x = 0.10$, and 32 K for $x = 0.15$. The Meissner fractions of the three samples determined in an external field of 2 mT were 0.08, 0.22, and 0.23, respectively. These are reasonable values for this type of material.[13] The Meissner fractions represent conservative lower limits of the superconducting volume fractions as trapped flux will reduce the actual signals.[14]

In the $\mu$SR experiment,[15] positive muons are implanted into the sample with an energy of approximately 4 MeV. The stopped muons decay by emitting a positron and two neutrinos. The emission probability of the positrons is anisotropic with respect to the muon spin direction and therefore allows the detection of the spin precession. In the present case only a decay of the asymmetry (no precession) is observed. However, since this decay is much faster than expected for nuclear moments[16] it is a clear indication of the presence of electronic moments in the sample.

Unfortunately, the site of the muon in the $La_2CuO_4$ structure is not known (for a discussion of possible sites see, e.g., Ref. 5). However, in the antiferromagnetic structure of undoped $La_2CuO_4$ a unique $\mu$SR frequency was observed, indicating that all muons stop at equivalent sites. Thus undoped $La_2CuO_4$, for which the magnetic structure is known,[3] can serve as a reference for the relative variation of internal fields in Sr-doped samples observed by the muons.

Representative data for $La_{1.93}Sr_{0.07}CuO_4$ for three different temperatures are displayed in Fig. 1. At the left side the time spectra are presented and on the right side, the corresponding Fourier transforms. The spectrum at 4.2 K represents the situation without an internal field from electronic moments. In this case the asymmetry $R(t)$ is almost constant in the time range shown in Fig. 1. The slight decay of $R(t)$ is due to nuclear moments. The spectra at 35 mK and 2.2 K clearly show a fast decay of the asymmetry corresponding to internal fields much higher than those arising from nuclear moments. Thus at these temperatures clearly internal magnetic fields are present.

In all spectra, the contribution from muons which stopped in the Cu sample holder was subtracted. This part shows practically no variation of the asymmetry in the time range shown. The fraction of this contribution was determined by a fit to the data at longer times (the spectra were measured up to 10 $\mu$s) and at low temperatures where a separation of the fast and slowly decaying part is easily possible.

Spectra for different Sr-concentrations measured at 35 mK are shown in Fig. 2. A clear relaxation, although much slower than for the $x=0.07$ sample, is seen also for $x=0.10$ and 0.15. Thus in these samples clearly magnetic fields due to electronic moments are present.

It should be emphasized that the observed effect is not due to magnetic impurities or to minority phases since essentially all muons (at least more than 70%) which stop in the sample participate in this process. This can be deduced from the observed asymmetry at $t=0$ which reaches the expected value. Thus the observed phenomenon is indeed an intrinsic property of the sample.

For $x=0.07$ and $T \leq 1.2$ K, the spectra were fitted with a Gaussian distribution around a finite frequency ($\nu \approx 1.5$ MHz). Above 1.2 K, the shape changed and an exponentially decaying function was more appropriate.

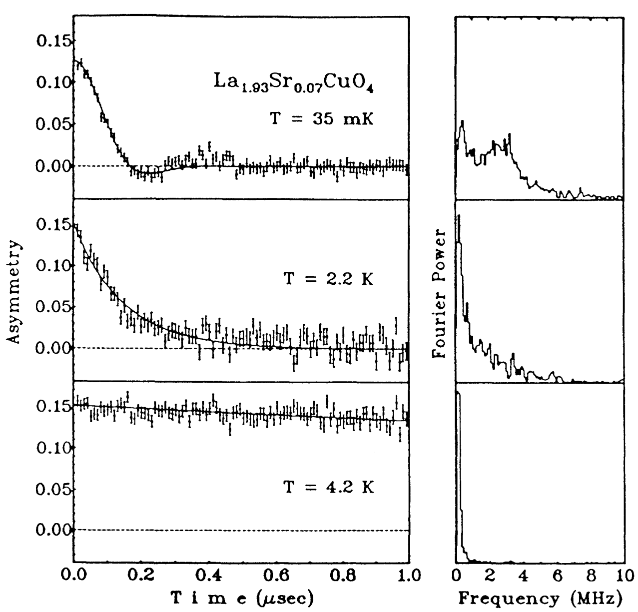


FIG. 1. $\mu$SR spectra and Fourier transforms for $La_{1.93}Sr_{0.07}CuO_4$ at different temperatures. The solid lines are theoretical-fit curves (see text).

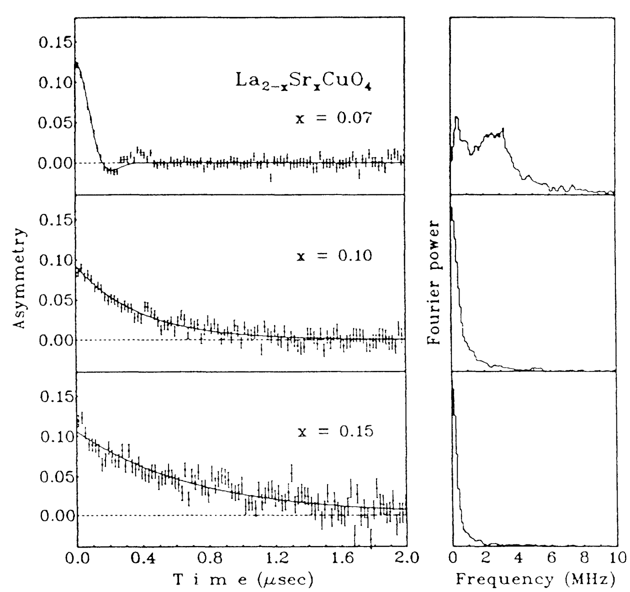


FIG. 2. $\mu$SR spectra and Fourier transforms for $La_{2-x}Sr_xCuO_4$ with different Sr concentrations $x$. The temperature of the samples was 35 mK.

The spectra for $x=0.10$ and 0.15 could all be fitted with a single exponentially decaying function. From the present data we cannot decide whether the observed magnetic fields are static or dynamic, i.e., due to frozen or fluctuating magnetic moments.

As a measure of the characteristic decay time of the anisotropy in Figs. 1 and 2 we take the value $T_e$ at which the anisotropy has fallen to $1/e$ of its value at $t=0$ and relate it to the average magnitudes of the $\mu$SR frequency $\langle|\omega|\rangle$ and the corresponding internal magnetic field $\langle|B_\mu|\rangle$ by

$$1/T_e \approx \langle|\omega|\rangle = \gamma_\mu \langle|B_\mu|\rangle , \qquad (1)$$

where $\gamma_\mu = 851.4$ MHz/T is the gyromagnetic ratio of the muon. In this way all data can be represented on a common graph irrespective of the detailed shapes of the spectra. The results are shown in Fig. 3.

Two features can be seen. (i) The values of $1/T_e$ (and correspondingly the magnitudes of the internal fields) depend strongly on the Sr concentration. At 35 mK, an order of magnitude difference is observed between $1/T_e$ for $x=0.07$ and $1/T_e$ for $x=0.15$. (ii) The $1/T_e$ values are fairly constant below 1 K but fall off steeply at higher temperatures, reaching unmeasurably small values between 2 and 3.5 K. The temperature at which the ordering disappears is weakly dependent on the Sr concentration.

Figure 4 shows the phase diagram derived from these data; also included are the results from an earlier investigation[5] on nonsuperconducting samples ($x \leq 0.05$). In the upper part of Fig. 4 the temperatures $T_N$ and $T_c$ for the transitions to magnetic ordering and to superconductivity are displayed as functions of the Sr concentration. For $x \geq 0.07$ and temperatures below 2–3 K a region with coexisting magnetic ordering and superconductivity is found. In the lower part of the figure we have plotted the magnitude of the observed magnetic fields at 35 mK. It can be seen that the fields are strongly reduced in the superconducting samples compared to undoped $La_2CuO_4$.

It is important to note that such a strong reduction of the fields cannot be explained by assuming that the magnetic moments producing these fields become randomly oriented but keep their magnitudes. Actually, for any deviation of the antiparallel ordering of the moments one would expect an increase of the magnetic fields at the muon site rather than a decrease as observed here. Thus the only explanation for the reduction of $\langle|B_\mu|\rangle$ in the superconducting state is either that the magnetic moments fluctuate even at the lowest temperature measured (35 mK) or that the moments themselves are reduced.

A reduction of the magnetic order parameter in the superconducting state was predicted by Schrieffer, Wen, and Zhang in the spin-bag model.[10] It will be interesting to see whether a reduction of the magnetic moments by a factor of approximately 15, as suggested by the present experiment, is reasonable in this model.

In the ionic picture of Aharony and co-workers[11] the muon is surrounded by either an antiferromagnetic or by a ferromagnetic local ordering of the Cu moments.

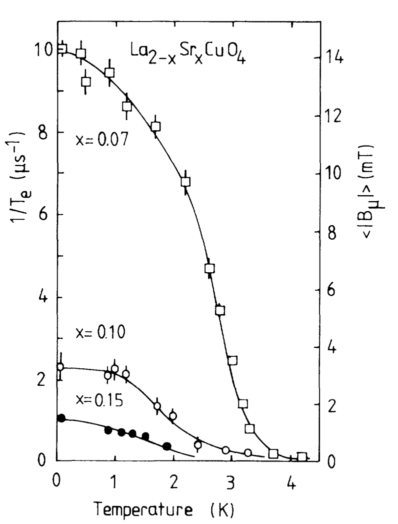


FIG. 3. The decay rate $1/T_e$ of the muon spin polarization as a function of temperature for $La_{2-x}Sr_xCuO_4$ with different Sr concentrations. The scale on the right-hand side refers to the average magnitude of the internal magnetic field calculated from $1/T_e$ via formula (1).

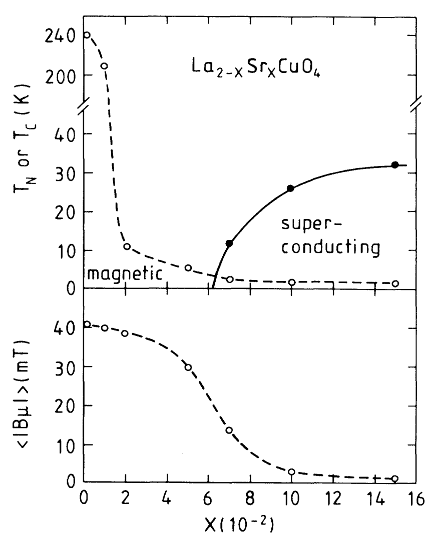


FIG. 4. Phase diagram of $La_{2-x}Sr_xCuO_4$ concerning the magnetic and superconducting properties as a function of the Sr concentration $x$. Upper part, the magnetic ordering temperature $T_N$ and the transition temperature to superconductivity $T_c$. Lower part, the average magnitude of the internal magnetic field at the muon site $\langle|B_\mu|\rangle$ at 35 mK. The lines are a guide to the eye.

However, since the ferromagnetic alignment which is connected with the charge carriers moves rapidly through the lattice (or is in a band state) an averaging effect with a reduction of the order parameter in the same sense as in the spin-bag model can be expected. Thus both models[10,11] are in qualitative agreement with the data.

Finally, we would like to mention that we also investigated the following samples: $YBa_2Cu_3O_7$ ($T_c$ = 90 K), $Bi_2Sr_2CaCu_2O_{9-y}$ ($T_c$ = 85 K), and Ba(Pb,Bi)$O_3$ ($T_c$ = 13 K). In none of these samples did we find a fast depolarization of the muon signal at 35 mK. This does not mean that there are no $Cu^{2+}$ spins in those systems. Rather the spins have no frozen component. However, in oxygen-deficient YBaCuO simultaneous superconductivity and magnetic ordering was reported in Ref. 6.

In summary, the present data give clear evidence for the coexistence of internal magnetic fields and superconductivity in $La_{2-x}Sr_xCuO_4$. In connection with the two-dimensional magnetic fluctuations observed in neutron scattering,[17,18] the present results suggest that magnetic correlations exist in $La_{2-x}Sr_xCuO_4$ in a wide concentration and temperature range, in particular, also in the superconducting region. The present data suggest that the fluctuations slow down as one lowers the temperature, and correlation times on the order of $10^{-8}$ s or longer are reached below 3 K.

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