# Muon Spin Relaxation Studies of Magnetic-Field-Induced Effects in High-$T_c$ Superconductors

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Muon spin relaxation measurements in high transverse magnetic fields ($\parallel \hat{c}$) revealed strong field-induced quasistatic magnetism in the underdoped and Eu-doped $(La, Sr)_2CuO_4$ and $La_{1.875}Ba_{0.125}CuO_4$, existing well above $T_c$ and $T_N$. The susceptibility counterpart of Cu spin polarization, derived from the muon spin relaxation rate, exhibits a divergent behavior towards $T \sim 25$ K. No field-induced magnetism was detected in overdoped $La_{1.81}Sr_{0.19}CuO_4$, optimally doped Bi2212, and Zn-doped $YBa_2Cu_3O_7$.

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The interplay between magnetism and superconductivity is one of the central subjects in the study of high-$T_c$ superconductivity [1]. Recently, remarkable effects have been observed in experiments with high external magnetic fields $B_{ext}$ applied parallel to the $c$ axis of single crystal specimens. Lake *et al.* [2] found a substantial increase of inelastic neutron scattering intensity at low energy transfers and incommensurate wave vectors with increasing field in optimally doped $La_{2-x}Sr_xCuO_4$ (LSCO). More dramatic effects have been observed in underdoped LSCO [3,4] and in oxygen-doped $La_2CuO_{4+y}$ (LCO) [5], where static spin correlations are enhanced by $B_{ext}$, as seen by increased elastic intensities of satellite neutron Bragg peaks. NMR experiments in high fields in $Tl_2Ba_2CuO_{6+\delta}$ (Tl2201) [6] and $YBa_2Cu_3O_{7-\delta}$ (YBCO) [7] detected the presence of dynamic antiferromagnetic spin correlations inside vortex cores. There remain, however, many unanswered questions, such as, in which case is the field-induced magnetism static or dynamic? Does it occur exclusively in vortex cores or everywhere in the system? Is this phenomenon generic to all the cuprates or specific to particular series and/or doping ranges? Is the superconducting order parameter affected by the field-induced magnetism?

Muon spin relaxation ($\mu$SR) is a probe well suited to shed some light on these aspects. Following zero-field (ZF) $\mu$SR studies on static magnetism of LSCO and YBCO superconductors [8,9], our recent ZF $\mu$SR results [10] elucidated the coexistence of magnetism and superconductivity in $La_2CuO_{4.11}$ (LCO:4.11) and LSCO with $x = 0.12$ (LSCO:0.12), where static magnetism occurs only in a finite volume fraction, forming nanoscale static spin stripe regions with a radius ~30 Å (comparable to the in-plane coherence length $\xi_{ab}$). In $\mu$SR measurements with low transverse field (TF) ($B_{ext} = 0.2$ T) in $La_{2-x-y}Eu_ySr_xCuO_4$ (LESCO) [11], we found a tradeoff between the superfluid density and the magnetically ordered volume fraction, which indicates that the superconductivity and static spin ordering occur in microscopically intertwined but spatially separate regions.

In this Letter, we report TF-$\mu$SR studies of the effect of high magnetic fields on several cuprate systems, listed in Table I, using single crystal specimens. The values of their transition temperatures for superconducting state ($T_c$) and static magnetic order ($T_N$) in ZF are shown together with the volume fraction $V_{Cu}$ of static Cu moments in ZF determined by ZF-$\mu$SR. References [3,10,12] describe growth and/or characterization of similar (not identical) crystals with the same nominal compositions made by the groups who prepared the present specimens.

Specimens of dimensions ~8 mm × 8 mm × 1 mm are mounted in He gas-flow cryostats with the largest face ($ab$ plane) perpendicular to the muon beam direction $\hat{z}$, along which the external field was applied with an initial muon spin polarization made perpendicular to $\hat{z}$ using a

TABLE I. Single crystal specimens studied in this work.

| Composition | Abbreviation | $T_c$ (K) | $T_N$ (K) | $V_{Cu}$(%) |
|---|---|---|---|---|
| $La_{1.88}Sr_{0.12}CuO_4$ | LSCO:0.12 | 28 | 20 | 40 |
| $La_{1.875}Ba_{0.125}CuO_4$ | LBCO | 2.5 | 40 | 80–100 |
| $La_{1.75}Eu_{0.1}Sr_{0.15}CuO_4$ | LESCO | 20 | 22 | ~50 |
| $La_{1.81}Sr_{0.19}CuO_4$ | LSCO:0.19 | 30 | 0 | 0 |
| $YBa_2(Cu_{2.979}Zn_{0.021})O_7$ | YBCO(Zn) | 80 | 0 | 0 |
| $(Bi, Pb)_2Sr_2CaCu_2O_8$ | Bi2212 | 80 | 0 | 0 |

 

spin rotator. We used the HI-TIME $\mu$SR spectrometer at TRIUMF to detect high-frequency precession signals. Time resolution and positron trajectories restricted the highest available field to $B_{\rm ext} = 6$ T.

Traditionally, the TF-$\mu$SR data in high fields have been analyzed using Fourier transforms and/or rotating reference frame [13,14]. Neither of them, however, clearly display the existence of multicomponent signals. As an alternative method [15], we extracted the amplitude of the signal from very short time intervals (~50 ns) by fitting the precession signal to a simple sinusoidal form $A\cos(\omega t + \phi)$. In such a time domain, the amplitude and frequency are practically constant, and the derived amplitude $A$ for each interval represents the muon spin relaxation function in TF, which we call the "envelope" of the precession signal. Figure 1 shows the time evolution of the envelope in several systems.

A significant increase of the relaxation rate with increasing fields is seen in LSCO:0.12 and LESCO at $T \ll T_c$ [Figs. 1(a) and 1(b); $B_{\rm ext}$ shown in 1(b)], and also at $T > T_c$ and $T > T_N$ as shown for LSCO:0.12 in Fig. 1(a). We obtained similar results in LBCO. The observed relaxation in these systems at $T < T_N$ is due predominantly to magnetism, since their relaxation rates are much higher than those expected from the magnetic field penetration depth $\lambda$. In LSCO:0.12 at $T < T_N$, the envelope shows a clear two-component decay, as demonstrated in the logarithmic plot of Fig. 1(c). In contrast, essentially no field dependence has been observed in LSCO:0.19 [Fig. 1(b)], YBCO(Zn), and $(\mathrm{Bi,Pb})_2\mathrm{Sr}_2\mathrm{CaCu}_2\mathrm{O}_8$ (Bi2212), in which the relaxation can be explained in terms of the penetration depth $\lambda$ in the superconducting state. A slight reduction of the relaxation rate at higher fields in YBCO(Zn) and Bi2212 can be attributed to a small increase of $B_{\rm ext}$ relative to $H_{c2}$ [16].

We fit the observed envelope of LBCO, LESCO, and LSCO:0.12 ($T > 25$ K) with a single exponential decay $\exp(-\Lambda t)$ and derived the muon spin relaxation rate $\Lambda$, as shown in Fig. 2. In the case of the two-component decay of LSCO:0.12 below $T_N$, we fit the envelope with a sum of two exponentially decaying functions. The points in Fig. 2(b) for this region (left of the vertical solid line) correspond to the exponential decay rates of the faster component, i.e., the slope of the broken lines in Fig. 1(c), while the points above $T = 25$ K represent $\Lambda$ in the single-component fit. These single- and two-component exponential fits in LSCO:0.12 and LESCO are displayed by the solid lines in Figs. 1(a) and 1(b), which exhibit a good agreement with the observed data.

In addition to large and field-dependent relaxation rates below $T_N$ due to local field from static Cu moments, we see a significant field-dependent relaxation well above $T_c$ and $T_N$ in LESCO, LSCO:0.12, and LBCO. As shown in the inset figures of Fig. 2, the relaxation rates $\Lambda$ above $T_N$, after correction for background relaxation due to nuclear

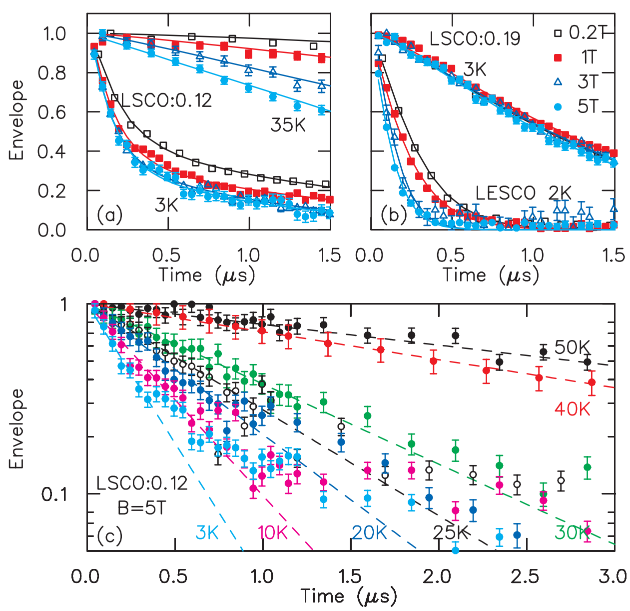


FIG. 1 (color). The envelope of muon spin precession in transverse external fields $B_{\rm ext}$ parallel to the $c$ axis observed in (a) LSCO:0.12 at $T = 3$ K and 35 K; (b) LSCO:0.19 at $T = 3$ K and LESCO at $T = 2$ K; and (c) LSCO:0.12 in $B_{\rm ext} = 5$ T in a logarithmic plot which demonstrates decay in two-component signals below $T \sim 25$ K.

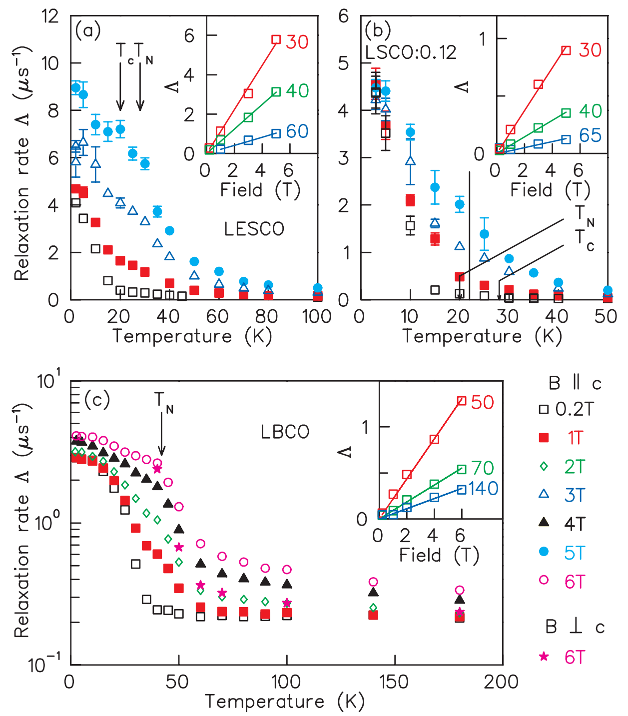


FIG. 2 (color). Muon spin relaxation rate $\Lambda$ in TF-$\mu$SR with $B_{\rm ext}$ applied parallel to the $c$ axis in (a) LESCO, (b) LSCO:0.12, and (c) LBCO. The purple star represents the results in LBCO with $B_{\rm ext} \perp \hat{c}$. Points of $\Lambda$ represent the decay rate of a single-component exponential signal, except for $T < 25$ K in LSCO:0.12 where they represent those of the faster component in a two-component exponential signal. The insets show field dependence of $\Lambda$ at selected temperatures.

dipolar fields and magnet-related inhomogeneous fields, exhibit a linear relation to the external field at a given temperature. In LBCO, we also performed some measurements with crystals cut into a different orientation to have the external field perpendicular to the $c$ axis, whose results are shown by the purple stars in Fig. 2(c). While $\Lambda$ exhibits nearly no dependence on the field orientation below $T_N$, we found a remarkable reduction of the field-induced effect above $T_N$ for $B_{\rm ext} \perp \hat{c}$.

From the slope of the field dependence of $\Lambda$ for $B \parallel c$ axis, such as those shown in the insets of Fig. 2, we obtained $d\Lambda/dB$, which might be called the "susceptibility counterpart" since the field-induced relaxation in these system is likely due to static random fields as discussed later. Figure 3(a) shows $dB/d\Lambda$, or the counterpart of inverse susceptibility, in these three systems as a function of temperature. We find that $dB/d\Lambda$ shows nearly linear temperature dependence with the intersect to the horizontal axis at $T \sim 25$ K common to all the three systems. This is a behavior analogous to inverse dc susceptibility of a ferromagnet having the Curie temperature of 25 K.

Using a SQUID magnetometer, we also measured the dc susceptibility $\chi_{\rm dc}$ of our specimens of LSCO:0.12 and LESCO in an external field $B \parallel \hat{c}$ of 0–5 T. At $T_c < T < 200$ K, the magnetization $M$ exhibits linear variation with $B$, with the field-independent slope $dM/dB$. Figure 3(b) shows the inverse susceptibility $1/\chi_{\rm dc} \equiv dB/dM$ in LSCO:0.12 and LESCO. The large $\chi_{\rm dc}$ in LESCO is due to the Van Vleck term of $Eu^{3+}$. In both systems $1/\chi_{\rm dc}$ is nearly independent of temperature, in a remarkable contrast to the "counterpart from $\mu$SR" in Fig. 3(a).

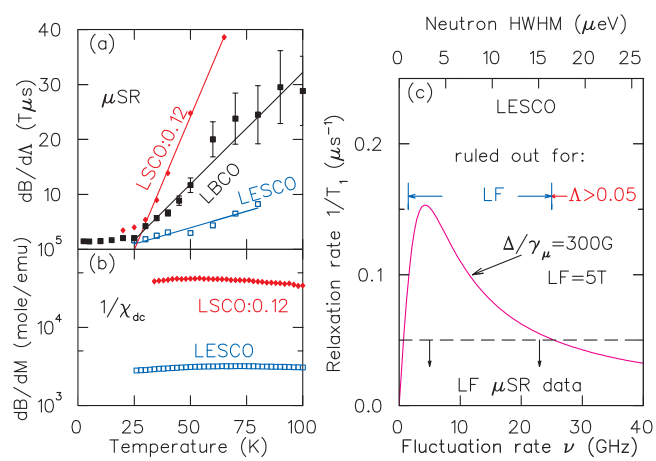


FIG. 3 (color). (a) The inverse susceptibility counterpart $dB/d\Lambda$ derived from the dependence of the relaxation rate $\Lambda$ in TF-$\mu$SR on $B_{\rm ext} \parallel \hat{c} = 0.2$–6 T. (b) The inverse of dc susceptibility $\chi_{\rm dc} \equiv dM/dB$ determined from the magnetization $M$ for $B \parallel \hat{c} = 0.2$–5 T. (c) Expected longitudinal relaxation rate $1/T_1$ in LF-$\mu$SR for Cu spin fluctuations with the rate $\nu$ compared with the LF-$\mu$SR results in LESCO at external field of 5 T. $\nu$ is also displayed with corresponding energy width (HWHM) in neutron studies.

In order to study possible dynamic effects, we have also performed $\mu$SR measurements of LESCO (and LBCO) under longitudinal field (LF) of $B_{\rm ext} \parallel \hat{c} = 5$ T [4 T] at temperatures between 2 and 150 K (of 20–30 K intervals) (2.5 and 160 K; 20 K intervals). We found no relaxation at any temperature, which provides an upper limit of the muon spin relaxation rate $1/T_1 < 0.05\ \mu{\rm s}^{-1}$. When Cu moments of $\sim 0.5\ \mu_B$/Cu become static, the internal magnetic field of 300 G would be created at the muon site in LESCO and LBCO [10,11]. For Cu spin fluctuations with the rate $\nu$, we simulated $1/T_1$ in LF of $B_{\rm ext} = 5$ T assuming a Gaussian random internal field with the width $\Delta/\gamma_\mu = 300$ G, where $\gamma_\mu$ denotes the gyromagnetic ratio of the muon spin. The simulation results in Fig. 3(c) exceed the upper limit of $1/T_1$ observed in LESCO for $\nu = 1$–$25 \times 10^9$/s, ruling out $\nu$ in this region [blue arrows in Fig. 3(c)]. When $\nu$ is significantly larger than $\nu \sim \gamma_\mu B_{\rm ext}$, which gives the $1/T_1$ maximum, $\Lambda$ in TF should be nearly equal to $1/T_1$ in LF, contrary to the observed data $\Lambda \gg 1/T_1$. Consequently, the fast fluctuation $\nu > 25 \times 10^9$/s cannot explain observed results. Combined results in LF and TF now indicate that $\nu > 10^9$/s is incompatible with our data.

Thus, we consider that the observed relaxation above $T_N$ is due to quasistatic random fields induced by the external field. Since this effect does not appear in the uniform susceptibility, it should be related to Cu spin polarization with finite wave vector component $q$, presumably originating from (possibly short ranged) stripe spin correlations which are stabilized below $T_N$. The dependence on the crystal orientation in LBCO rules out "polarization of dilute magnetic impurities" as a possible explanation. Good fit of the envelope to single exponential decay above $T_N$ suggests that the observed phenomenon is not confined to a small volume fraction of the system.

Regarding the two-component signals in LSCO:0.12 below $T_N$, the signal with fast (slow) relaxation must be due to volume fraction with (without) static magnetic order. In Fig. 4(a), we show the amplitude fraction $V_\mu$ of muons involved in the fast relaxation. The volume fraction $V_{\rm Cu}$ of static Cu moments is slightly smaller than $V_\mu$, since muons stopped near the edge, but outside, of the "magnetic islands" are also depolarized. After converting $V_\mu$ into $V_{\rm Cu}$ using Fig. 8 of Ref. [10], we obtained $(1 - V_{\rm Cu}) \equiv V_{\rm sc}$. (Different models for the shape of magnetic regions would yield qualitatively similar behavior of $V_{\rm Cu}$.) If one assumes that the slow relaxation is due to the superconducting region, $V_{\rm sc}$ represents the superconducting volume fraction. Figure 4 shows that both $V_{\rm sc}(T \rightarrow 0)$ and $T_c$ decreases with increasing field, but their ratio is nearly independent of the field. Similar proportionality between $T_c$ and the superconducting volume fraction [17] has been observed in the case of varying $T_c$ with (Cu,Zn) substitution in LSCO systems [18] and increasing Eu concentration in LESCO [11].

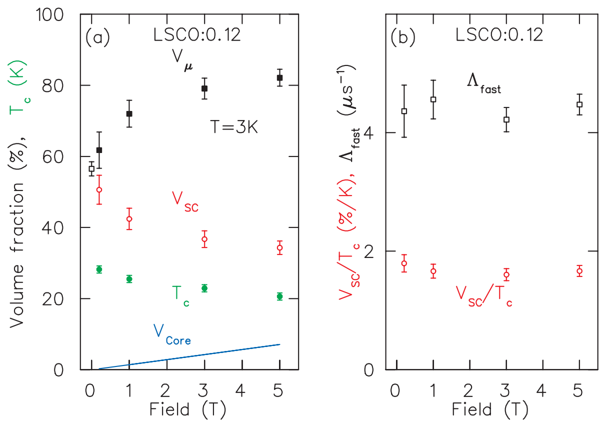


FIG. 4 (color). (a) The volume fractions of the fast muon decay signal $V_\mu$ and superconducting region $V_{sc}$ and $T_c$ in LSCO:0.12, and the core region $V_{core}$ for a superconductor with $\xi_{ab} = 23$ Å. (b) The decay rate $\Lambda_{fast}$ at $T = 3$ K and $V_{sc}/T_c$ in LSCO:0.12.

In LSCO:0.12, the relaxation rate $\Lambda$ of the fast-decay component is nearly independent of the field at $T \leq 5$ K, as shown in Figs. 2(b) and 4(b). This implies that the external field increases the magnetic volume fraction but does not change the moment size. The blue solid line $V_{core}$ in Fig. 4(a) represents volume fraction of the vortex core region for superconductors having the in-plane coherence length $\xi = 23$ Å. We can rule out the "vortex cores with full static magnetism" at $T = 2$–3 K in YBCO:Zn, Bi2212, and LSCO:0.19, in which the observed envelopes in $B_{ext} = 5$–6 T indicate that muon spins involved in such "fast decay" is less than 1% in volume. Static magnetism with much smaller field (~15 G) in the vortex core, proposed for an underdoped YBCO [13], still remains as a possibility in these three systems.

The strong field-dependent effects above and below $T_N$ were observed only in the systems which exhibit static magnetism in ZF in partial or nearly full volume fraction $V_{Cu}$ (see Table I.). Thus, the field-induced relaxation is not generic to all the cuprate systems but is confined to systems having competing magnetic states very close in free energy to their superconducting state. The stronger effect for the field applied perpendicular to the $CuO_2$ planes in LBCO suggests that this phenomenon could be related to suppression of superconductivity caused by the field. Though the superconducting $T_c$'s are rather low in underdoped LSCO:0.12, LBCO, and LESCO systems, the "dynamic superconductivity"[17,19] detected by the Nernst effect [20] might extend up to $T \sim 150$ K ("Nernst region"). The suppression of dynamic superconductivity could favor competing antiferromagnetism [21].

Recently, a very small diamagnetic magnetization $M_{dia}$ was detected in Bi2212 in the Nernst region above $T_c$ [22]. The field and temperature dependences of $M_{dia}$ follow behavior similar to that of $\Lambda$ in $\mu$SR. This suggests a possibility that the observed relaxation above $T_c$ is due to the dynamic supercurrent screening $B_{ext}$. However, if we assume $\Lambda = \alpha M_{dia}$ and obtain the proportionality factor $\alpha$ using the results in Bi2212 near $T = 0$, that factor gives the value of $\Lambda$ more than 10 times smaller than the observed value for a reasonable estimate of $M_{dia}$ above $T_c$ in LSCO and LESCO using the Nernst results. Furthermore, it is difficult [19] to expect the dynamic diamagnetism to have the time scale slower than $t \sim 1$ $\mu$s, required for a flux vortex lattice to produce field inhomogeneities observable in $\mu$SR. These difficulties have to be resolved before the dynamic screening scenario is adopted.

In conclusion, our $\mu$SR measurements in high TF revealed field-induced magnetism in LSCO:0.12, LESCO, and LBCO existing in a wide range of temperatures above and below $T_c$ and $T_N$. This phenomenon is not common to all the cuprate superconductors, but rather confined to those systems having magnetically ordered states closely competing with the superconducting state.

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