Tuning competing orders in $La_{2-x}Sr_xCuO_4$ cuprate superconductors by the application of an external magnetic field

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We report the results of a combined muon-spin rotation and neutron-scattering study on $La_{2-x}Sr_xCuO_4$ in the vicinity of the so-called 1/8 anomaly. Application of a magnetic field drives the system toward a magnetically ordered spin-density-wave state, which is fully developed at 1/8 doping. The results are discussed in terms of competition between antiferromagnetic and superconducting order parameters.

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I. INTRODUCTION

Competing order parameters are a central theme in condensed-matter physics. This is especially true for the study of high-temperature superconductors (HTSCs), where superconductivity occurs upon hole doping of an antiferromagnetic Mott insulator. As a consequence of the competition between superconductivity (SC) and antiferromagnetism (AF), different ground states have been identified in the underdoped regime of La-based cuprates. Among those are (i) *d*-wave SC; (ii) a disordered spin-glass-like state, which coexists with SC over a broad range of doping;[1] and (iii) a spin-density-wave (SDW) state with suppressed SC around a specific hole concentration $x \approx 1/8$. This so-called 1/8 anomaly was observed in $La_{15/8}Ba_{1/8}CuO_4$ (Ref. 2) and later in $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ (LNSCO),[3–5] where the effect is concomitant to a structural phase transition from a high-T orthorhombic (HTO) to a low-T tetragonal (LTT) phase.[6] A similar anomaly was found in the $La_{2-x}Sr_xCuO_4$ (LSCO) system at $x \approx 0.115$,[7] however without a structural HTO-LTT transition and without a complete suppression of SC. A stripe model with spatial modulations of spin and charge densities has been suggested to account for the incommensurate magnetic and simultaneous charge order observed in neutron-diffraction experiments on LNSCO.[3–5] In this model dynamic stripe correlations of spins and holes are stabilized for $x \sim 1/8$ in the LTT phase and suppress SC.

Starting from $La_{1.88}Sr_{0.12}CuO_4$, there are several routes to reach the 1/8 state. One way is simply to substitute Sr with Ba (Ref. 8) or La with Nd. An alternative is to introduce pinning centers into the CuO_2 planes. Muon-spin-rotation (μSR) results on Zn-doped $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$ show an enhancement of magnetism in the vicinity of 1/8 doping,[9] suggesting that small amounts of nonmagnetic impurities act as pinning centers for dynamical stripe correlations. The observation of similar magnetic anomalies in Zn substituted Bi-2212 and $YBa_2Cu_3O_{6+x}$ seems to indicate that the 1/8 anomaly is not just a specific feature of the La-based compounds but is a general property of HTSCs.[10,11]

The subtle balance between the competing orders may also be changed by external perturbations such as magnetic fields[12,13] or pressure.[14] For example, it was demonstrated that the static incommensurate (IC) magnetic neutron response for LSCO with doping close to 1/8 is enhanced by the application of a magnetic field perpendicular to the CuO_2 planes.[12,13] The microscopic mechanisms behind the 1/8 anomaly as well as the primary cause of the field effect remain however, poorly understood.

We have therefore performed a systematic study of the competition between the AF and SC order parameters in the vicinity of 1/8 doping. By combining μSR and neutron-diffraction results obtained on the same single crystals, we show that for samples in the 1/8 doping state the AF order is already fully developed in zero field (ZF) and can therefore not be enhanced by the application of an external magnetic field. A field-induced enhancement of the staggered moment is only observed in samples with a reduced ZF magnetic response.

II. METHODS

High quality LSCO with $x=0.105$, $x=0.12$, and $x=0.145$ and LNSCO single crystals were grown by the traveling solvent floating zone method.[15] The Sr content was reassessed by measuring the structural transition from a high-T tetragonal (HTT) to a low-T orthorhombic (LTO) phase, which varies strongly with hole doping.[16,17] The magnetic onset temperature was determined for all samples with both μSR ($T_f^{\mu SR}$) and neutron scattering (T_f^n). The difference between $T_f^{\mu SR}$ and T_f^n is due to the different observation time scales of the two experimental techniques. Table I summarizes the properties of the single crystals used in this study.

TABLE I. Compilation of μSR and neutron-scattering results on La-based compounds. μ_{lo} and μ_{av} denote the local and average Cu moments as determined by μSR and neutron scattering, respectively. $T_f^{\mu SR}$ and T_f^n are the corresponding time scale dependent freezing temperatures. δ is the incommensurability. The values in the last column indicate the structural transition temperatures.

1/8 compound	Nominal doping	T_c onset	μ_{lo}	μ_{av}	$T_f^{\mu SR}$	T_f^n	δ	$T_{LTT\text{-}HTO}$
$La_{2-x-y}Nd_ySr_xCuO_4$	$x=0.12$, $y=0.4$	7 K	$0.35(2)\mu_B$	$0.071(4)\mu_B$	50 K	65 K	0.122(4)	70 K
$La_{2-x}Ba_xCuO_4$	$x=0.125$	5 K[a]	$0.35\mu_B$ [b]		40 K[c]	50 K[a]	0.118[a]	55 K[a]
$La_{2-x}Sr_xCuO_{4+y}$	$x=0.09$, $y=0.03$	40 K[d]	$0.36\mu_B$ [d]		40 K[d]	40 K	0.123[d]	
Compound	**Nominal doping**	T_c	$\mu_{lo}/\mu_{lo}(1/8)$	$\mu_{av}/\mu_{av}(1/8)$	$T_f^{\mu SR}$	T_f^n	δ	$T_{LTO\text{-}HTT}$
$La_{2-x}Sr_xCuO_4$	$x=0.120$	27 ± 1.5 K	0.51(4)	0.33(8)	15 K	30 K	0.125(3)	255 K
$La_{2-x}Sr_xCuO_4$	$x=0.105$	30 ± 1.5 K	0.36(8)	0.22(9)	10 K	25 K	0.108(2)	290 K
$La_{2-x}Sr_xCuO_4$	$x=0.145$	36 ± 1.5 K	<0.014	0			0.13(2)	190 K

[a]Reference 18.
[b]Reference 19.
[c]Reference 20.
[d]Reference 21.

The ZF μSR experiments were performed at the πM3 beamline of the Paul Scherrer Institute (PSI), which provides 100% spin-polarized positive muons. They are implanted into the sample and come to rest at interstitial lattice sites without losing their initial polarization. The spin of the muon then acts as a very sensitive local magnetic probe through its precession in the internal field $\mathbf{B}(\mathbf{r})$. In cuprate materials the muon stopping site is close to an apical oxygen and $\mathbf{B}(\mathbf{r})$ arises from dipolar fields created by the surrounding Cu^{2+} moments.[22] The neutron-scattering experiments were carried out on the cold neutron spectrometers RITA II at PSI, FLEX at the Hahn-Meitner Institute, and IN14 at Institut Laue-Langevin. The experiments were performed with a fixed incoming and final wave vector ($k_f=k_i=1.5$ or 1.9 Å^{-1}). A Be or pyrolytic graphite (PG) filter was installed before the analyzer in order to eliminate higher-order contamination. The samples were mounted in vertical 15 T cryomagnets such that $(Q_h,Q_k,0)$ were accessible. All measurements in an external magnetic field $\mu_0 H$ were performed after field cooling.

III. RESULTS AND DISCUSSION

Incommensurate AF order is observed at $\mathbf{Q}_{IC}=(0.5,0.5\pm\delta,0),(0.5\pm\delta,0.5,0)$ in tetragonal units of $2\pi/a=1.65$ Å^{-1}. The incommensurability δ depends on the Sr content as in Ref. 23. Figure 1 summarizes the results of our elastic neutron-diffraction experiments. Panels (a)–(d) are presented with increasing elastic response in ZF. For $x=0.145$ [Fig. 1(a)], no elastic response is observed in ZF. However, application of $\mu_0 H=13$ T induces an elastic response at $\delta\approx0.13$, confirming a previous report.[24] For $x=0.105$ and $x=0.12$ an elastic response exists already in ZF and an applied magnetic field enhances the magnetic response for $T<T_f^n$ [Figs. 1(b) and 1(c)]. LNSCO shows the strongest ZF response and the absence of a field effect at all T.

Next, we plot in Fig. 1(e) the T dependence of the intensity I at $\mathbf{Q}_{IC}$. The T axis and I axis are normalized to T_f^n and $I(2\text{ K})$ [for LNSCO we used $I(5\text{ K})$ to stay above the Nd ordering temperature]. Due to the absence of a field effect in LNSCO we plot, for simplicity, only the ZF data in this figure. The ZF elastic response of LNSCO exhibits an order-parameter-like T dependence, whereas the ZF response for $x=0.105$ and $x=0.12$ is significantly different and close to a linear T dependence as indicated by the dashed lines. It is remarkable that the application of an external magnetic field changes the T dependence in such a way that the normalized data now also fall on the same order-parameter-like curve.

The superconducting transition temperature is strongly suppressed in LNSCO ($T_c\approx7$ K). We therefore consider this compound to most closely mimic the physics of the so-called 1/8 state. The time evolution of the muon-spin polarization, $A(t)$, exhibits a strongly damped oscillatory behavior that is well described by a Bessel function with a frequency $\nu\approx3.5$ MHz (see Fig. 2). This observation is consistent with the existence of an IC SDW state.[19,25] We stress that similar results are obtained for $La_{15/8}Ba_{1/8}CuO_4$ (Refs. 18, 26, and 27) and for the AF-ordered volume fraction in superoxygenated $La_{2-x}Sr_xCuO_{4+y}$.[21] The latter compound was shown to phase separate into optimally doped SC regions and an AF-ordered phase closely related to the 1/8 state. The characteristic features of the 1/8 state are therefore (i) a strongly suppressed T_c, (ii) the observation of a Bessel-type relaxation with $\nu\approx3.5$ MHz in the μSR time spectra, and (iii) incommensurate SDW order with $\delta\approx0.125$ and the absence of a field effect. The 1/8 state consists, most likely, of static stripes with an associated SDW order, which in turn suppresses the SC order parameter. The 1/8 anomaly is limited to a very narrow doping range and slight variations in the doping level lead to very noticeable changes in the physical properties.

Due to its dipolar character, $\mathbf{B}(\mathbf{r})$ is directly proportional to the ordered Cu^{2+} moment. For LNSCO the internal field at $T=5$ K is found to be 27 mT, which is about 2/3 of the value observed in the undoped compound La_2CuO_4.[28,29] Assuming a value of $0.6\mu_B$ for the Cu^{2+} moment in La_2CuO_4, the internal field corresponds to a local ordered moment μ_{lo}

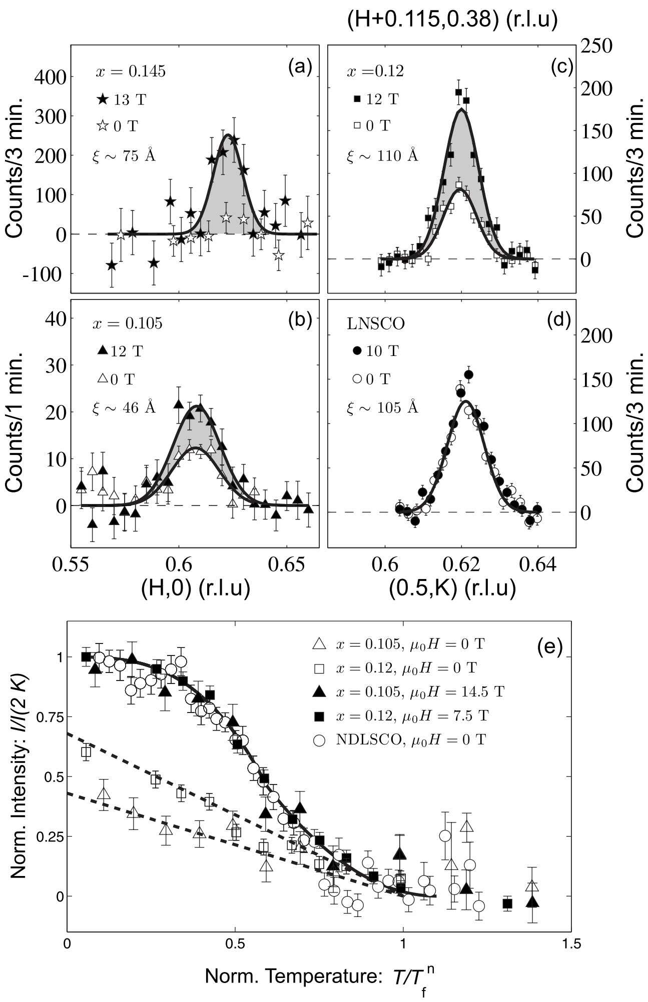

FIG. 1. Q scans around $\mathbf{Q}_{IC}$ performed on (a) $x=0.145$, (b) $x=0.105$, (c) $x=0.12$, and (d) LNSCO. Solid lines are Gaussian fits to the data. Note the different x-axis scales for the left and right panels. The magnetic correlation length ξ is derived from the full width at half maximum (FWHM) of the magnetic peaks. Data in (c) and (d) are resolution limited. (e) T dependences of the intensity at $\mathbf{Q}_{IC}$ for $x=0.105$, $x=0.12$, and LNSCO. Open symbols indicate data taken in ZF, while filled symbols represent data taken in a magnetic field. Lines are guides for the eye.

$\approx 0.36\mu_B$. The average ordered moment estimated from the neutron-diffraction experiments suggests $\mu_{av}=0.07\mu_B$, consistent with a previous report.[4] Note that for a sinusoidal SDW there is a factor of 2 between μ_{lo} and μ_{av}.[19] However, there is still a discrepancy between μ_{lo} estimated by μSR and μ_{av} determined by neutron diffraction (see also Ref. 30). A full knowledge of the microscopic spin topology might be necessary to solve this issue.

When moving away from 1/8 doping, the ordered moment at ZF decreases systematically with decreasing doping. For $x=0.12$, $A(t)$ still exhibits the characteristic Bessel-type oscillation, albeit with a reduced frequency and an increased damping (see Fig. 2). Note that we observe the full muon asymmetry, i.e., all muons experience $\mathbf{B}(\mathbf{r})\neq 0$. This implies that the magnetic order persists throughout the entire volume of the sample, which is a remarkable result for a superconductor with a T_c as high as 27 K. We emphasize that the

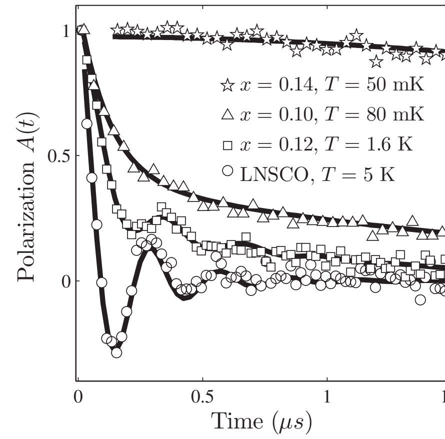

FIG. 2. μSR time spectra obtained in ZF and low temperatures. The solid lines are fits with a Bessel function for LNSCO and $x=0.12$, a simple exponential decay ($x=0.105$), and a Kubo-Toyabe function ($x=0.145$).

magnetic ground state may still be inhomogeneous but the characteristic length scale for this inhomogeneity has to be smaller than $\sim 10-20$ Å, which is the typical range for dipolar fields that originate from AF-ordered moments. In fact, a nanoscale inhomogeneous state consisting of SC droplets and patches of AF correlated regions is a likely candidate for the ground state in a region of the phase diagram where the antiferromagnetic and superconducting phases are very close in energy.[31,32]

The neutron-scattering results confirm the existence of IC magnetic order in ZF and in addition reveal a significant enhancement of the elastic intensity by an applied magnetic field [see Fig. 1(b)].[12] Combining the ZF μSR data with the neutron results, we are able to display the field dependences for the different samples in a common plot (see Fig. 3). It can be inferred from this figure that for $x=0.12$ the applied field tends to restore the magnetism characteristic of 1/8 doping. This suggests that the effect of the field is to drive the system toward the 1/8 ground state.

For $x=0.105$, $A(t)$ does no longer show the features of a Bessel function, but is now well described by a single exponential decay (see Fig. 2). The static nature of $\mathbf{B}(\mathbf{r})$ was verified by longitudinal field experiments. We deduce a static field distribution $\Delta\approx 10$ mT, which is significantly reduced from the 27 mT observed in the 1/8 compounds. Application of $\mu_0H=12$ T doubles the amplitude of the elastic signal [see Fig. 1(b)]. The value characteristic of static stripe order, however, cannot be fully restored in this system.

For the $x=0.145$ compound, $A(t)$ does not exhibit any relaxation due to electronic moments. The slow decay of $A(t)$ is well fitted by a static Kubo-Toyabe function (see Fig. 2), which describes the field distribution $P(\mathbf{B})$ arising from nuclear moments alone.[34] The width of $P(\mathbf{B})$ defines an upper limit for the electronic moments $\mu_{lo}<0.005\mu_B$. Neutron-diffraction studies on this sample show field-induced static AF order resembling that of underdoped compounds for $\mu_0H>\mu_0H_c$ with $\mu_0H_c\approx 7$ T (see Fig. 4). A previous report found $\mu_0H_c\approx 3$ T,[24] which might indicate that the doping level of that sample is slightly lower.[33]

We have also performed a systematic study of the vortex lattice (VL) in $x=0.105$, $x=0.12$, and $x=0.145$ by small-

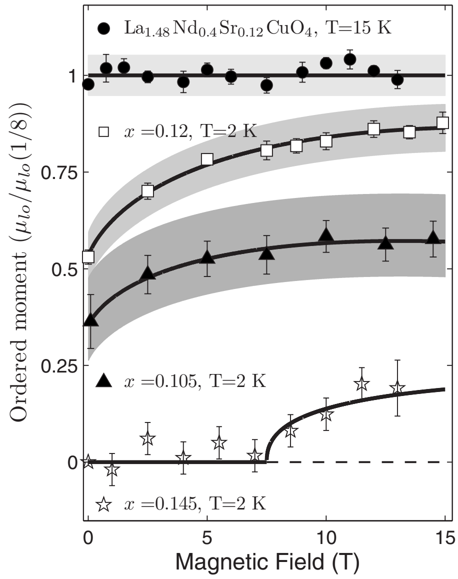

FIG. 3. H dependences of the response at $\mathbf{Q}_{\mathrm{IC}}$ for LSCO $x=0.105$, $x=0.12$, $x=0.145$, and LNSCO. The solid lines are fits to $\mu=\sqrt{I(H)}\propto\sqrt{(H/H_c)\ln(H_c/H)}$ (Ref. 33). Here the ZF moments estimated by μSR are used and the gray colors indicate the error related to the determination of the ZF order moment. The $x=0.145$ data are presented in arbitrary units.

angle neutron scattering. For $x=0.145$, we observe a VL resembling that at optimum doping[35] but only for $\mu_0 H<7$ T. No VL could be detected in $x=0.12$, where the largest elastic field effect is observed. Although vortices might exist in disordered structures, we find it difficult to correlate the elastic field effect with vortex matter physics. Instead, we interpret our data in terms of competing order parameters. Recently, we reported the observation of a single d-wave gap in the angle-resolved photoelectron spectroscopy (ARPES) spectra of the $x=0.145$ sample.[36] The most likely ZF ground state of $x=0.145$ is therefore pure d-wave SC similar to that observed at optimum doping. Application of a magnetic field tunes the system into a different ground state where static IC AF coexists with SC. This ground state resembles that of more strongly underdoped LSCO ($x<0.13$), where static AF and SC orders compete even in ZF. For $x\approx 0.12$, the ordered Cu moment at $\mu_0 H=15$ T is close to that the 1/8 ground state (see Fig. 3). Therefore we argue that the effect of the applied field is to drive the system toward the 1/8 ground state.

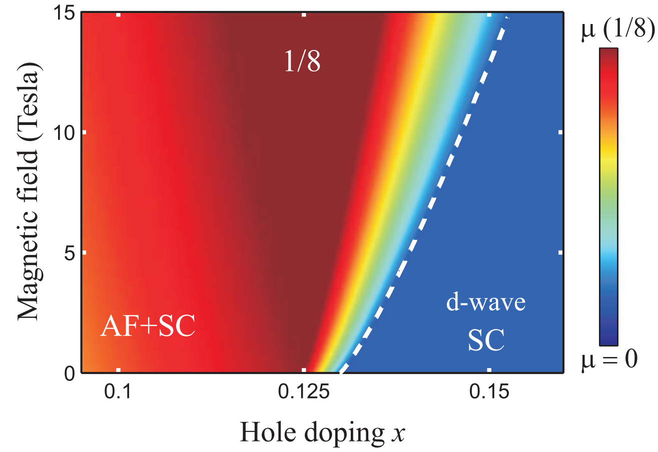

FIG. 4. (Color) Schematic doping-field phase diagram for $La_{2-x}Sr_xCuO_4$. The ordered moment is given in false colors with red (blue) as the maximum (minimum).

IV. CONCLUSIONS

To summarize our combined μSR and neutron-diffraction experiments, we present in Fig. 4 a schematic $\mu_0 H$-x phase diagram, in which the ordered Cu moment is depicted by a false color scheme.[37] The 1/8 state and the pure d-wave SC ground state are pictured as the dark red and the blue regions, respectively. Colors in between represent a state where AF and SC coexist. With the application of a magnetic field, one can tune the pure SC state into the mixed state of AF and SC. At the specific doping $x=0.12$, we found that the field drives the system toward the 1/8 state. The different ground states are therefore very close in energy. Our results clearly support the notion of competing SC and static AF order parameters. The systematics of our data shows that the existence of AF is intrinsic and not due to defects or chemical inhomogeneities. Any suppression of superconductivity either by a change in chemistry or by an external perturbation goes along with a concurrent and systematic enhancement of static magnetism.

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