Field-induced electronic phase separation in a cuprate high temperature superconductor

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We present a combined neutron diffraction (ND) and high-field muon spin rotation (μSR) study of the magnetic and superconducting phases of the high-temperature superconductor La1−xSr0.66CuO4+y (Tc = 38 K). We observe a linear dependence of the ND signal from the modulated antiferromagnetic order (m-AFM) on the applied field. The magnetic volume fraction measured with μSR increases linearly from 0% to ~40% with applied magnetic field up to 8 T. This allows us to conclude, in contrast to earlier field-dependent neutron diffusion studies, that the long-range m-AFM regions are induced by an applied field, and that their ordered magnetic moment remains constant.

Magnetic fluctuations are at the heart of research of high-temperature superconductivity (SC), being a prime candidate for the SC pairing glue [1]. Static magnetism on the other hand seems to be in competition with SC but the exact relation between modulated antiferromagnetic (m-AFM) correlations and SC remains controversial.

Besides inducing SC, hole doping suppresses commensurate AFM order of the parent compound and introduces m-AFM correlations, sometimes concurrent with charge modulations, commonly referred to as magnetic and charge “stripes”, respectively [2, 3]. There is experimental evidence that m-AFM co-exists with SC in the La-based cuprates with the 214 composition of La2−xMxCuO4, where the holes are introduced via the doping by M-atoms, being Sr or Ba [2, 4–6], but it remains unsolved if the coexistence is microscopic or not.

Another route for introducing holes is by controlling the oxygen content, which is the method used for YBa2Cu3O6+δ (YBCO) and La2CuO4+y (LCOO) [7]. Unlike doping by chemical substitution, the oxygen dopants remain mobile in LCOO down to temperatures around 200 K [8–10]. When oxygen dopants are allowed to anneal at ambient temperatures, the holes can distribute in a way that favours long-range ordered m-AFM regions with a period of 8 unit cells [11] and support connected SC regions to give an optimal Tc [10]. In co-doped La2−xSrxCuO4+y (LSCO), holes are introduced both by chemical substitution and by intercalating extra oxygen. So far, m-AFM order is found in all oxygen (co-)doped samples when they are optimally oxygen-doped achieving Tc ≈ 40 K [11–14]. In contrast, oxygen-stoichiometric LSCO shows m-AFM order only in the underdoped region, x < 0.135 [15–17].

In oxygen-stoichiometric La214 cuprates, m-AFM order is most prominent at dopings just around x = 0.125, accompanied by a suppression of Tc [18–20]. Here the intensity of the m-AFM neutron diffraction (ND) signal is enhanced by an applied magnetic field [16, 21, 22]. At higher doping levels, m-AFM order is absent, but can be induced with a magnetic field [21, 23–25].

The field-driven enhancement of the magnetic ND signal is generally interpreted as a field-induced increase of the ordered moment [16, 21, 26]. However, muon spin rotation (μSR) has revealed that many La214 cuprates are electronically phase separated into an optimally doped SC phase and a magnetic phase [12, 14, 17]. An inverse correlation exists between the volumes of these two phases across the Sr-doping range of LSCO [12, 13].

In this work, we study the magnetic stripes in the co-doped cuprate La1.94Sr0.06CuO4+y with Tc = 38 K. In contrast to other LSCO compounds, zero field data shows no static magnetism. We show that an applied magnetic field induces static magnetic order, increasing the magnetic volume fraction (Vm) linearly from 0% at 0 T to 37(3)% at 8 T, see Fig. 1. At the same time, the intensity of the ND signal from the m-AFM order also increases linearly with an applied magnetic field. The combination of these observations implies that the
Field-induced ND signal is caused by an increase in the magnetic volume fraction, and not by an increase in the ordered magnetic moment as was concluded in earlier studies. The insights gained from the combination of high-field μSR and ND allow us to propose a coherent framework for understanding the field-induced magnetic order in La-214 superconductors.

A 5 g single crystal was grown by the traveling solvent floating zone method and was oxygenated through a wet-chemical technique [12] obtaining a single transition temperature of $T_c = 38$ K. Magnetisation measurements were performed both before and after oxidation, see the supplementary material (SM) [27]. To ensure consistency between results obtained from all experiments, the sample was always field cooled and we adopted a slow cooling procedure to enable the mobile excess-oxygen to find an optimal configuration in the crystal structure before freezing [9].

The magnetic susceptibility was measured with a vibrating sample magnetometer in a Physical Properties Measurement System set-up at the Technical University of Denmark. ND was performed at the cold triple axis spectrometers RITA-II at Paul Scherrer Institute (PSI), Switzerland [28, 29] and ThALES at Institute Laue-Langevin (ILL), France [30, 31]. In both ND experiments, the sample was mounted in a 14.9 T cryomagnet with $c$-axis along the field. The incoherent energy resolution was $\sim 0.2$ meV (FWHM). The crystal was mounted to allowed access to wave vectors $(h,k,l) = (2\pi/a, 2\pi/b, 0)$ enabling us to measure the signal from the m-AFM, which appears as a quartet of weak peaks around the antiferromagnetic point $Q_{AFM} = (1,0,0)$ [32]. The μSR measurements were carried out at the general purpose spectrometer GPS [33] and the high transverse field spectrometer HAL9500 [34], both located at PSI, using a smaller piece of the same sample and with the

magnetic field applied along its $c$-axis. See SM [27] for details on the experimental techniques.

A selection of raw ND data is presented in Fig. 2 (a). A small signal is present at 2 K in zero field with counts slightly above the 40 K background, see SM [27]. We fitted the data using Gaussian peaks on a sloping background. Due to the weak signal, not all parameters were fitted simultaneously. The peak center and width show no visible dependence on the magnetic field, and we have thus fixed these to the values obtained at 14.9 T.

The m-AFM signal is present at all four scattering vectors $Q_{m-AFM} = (1 \pm \delta_h, \pm \delta_k, 0)$ with $\delta_h \approx \delta_k = 0.124(2)$, and corresponds to a period-8 AFM modulation along the Cu-O bonds. The full width at half maximum (FWHM) of the peak is $2w = 0.019(2)$ r.l.u. This value is within errors the same as the intrinsic instrument resolution and gives a conservative lower bound on the size of the m-AFM regions through the correlation length, $\xi = 1/w > 50$ Å [35]. The m-AFM regions are thus fairly large at all applied fields.

We have further measured the m-AFM peak at $Q_{m-AFM} = (1 \pm \delta_h, \pm \delta_k, l)$, for $2 \leq l \leq 4$, and find the intensity to be independent of $l$, see SM [27]. This demonstrates that the field-induced m-AFM order is two-dimensional, different from the zero-field m-AFM phase in LSCO where short-ranged magnetic correlations along the $c$-axis were found for $x = 0.10$ [36] and $x = 0.12$ [37].

We have searched for charge stripes of the type observed in LSCO with $x \sim 0.12$ using the exact same X-ray diffraction methods at BW5, DORIS [38] as in Refs. 4 and 5. No signal was found in our sample, see SM [27].

![FIG. 1. The low temperature (<5 K) field dependence of the ND intensity from the m-AFM phase (blue) and the magnetic volume fraction ($V_m$) measured with μSR (gray).](image-url)

![FIG. 2. (a): ND data measured at RITA-II at 1.6 K. The data is collected by scanning along $q = (h00)$ across the point $Q = (1.125, -0.125, 0)$, counting between 7 and 18 min. per point. Each data set is offset by a multiple of 1.5. Solid lines are Gaussian fits as described in the text. (b-d): Examples of μSR asymmetry data in applied magnetic fields measured at HAL9500. Solid lines are two-component fits and dashed envelopes highlight the difference in the fitted relaxation rates.](image-url)
Fig. 2 (b-d) show a selection of μSR asymmetry spectra in a rotating reference frame (RRF) chosen such that the very high muon spin precession frequencies at high fields are scaled down for better visualization of the data. The RRF frequency is 130 MHz at 1 T and 1080 MHz at 8 T. In panel (b), data taken at 50 K shows an undamped oscillation of the muon spin asymmetry that is well described by a model including a single Gaussian component with a frequency corresponding to the external magnetic field of 1 T. This behaviour is expected for a sample in the paramagnetic state. At 5 K and 1 T in panel (c), the spectrum shows a clear decay of the muon spin asymmetry, which is dominated by one oscillating component with Gaussian relaxation. In panel (d) at 8 T and 5 K, the time evolution of the muon spin asymmetry is best described using two distinct components corresponding to the magnetic and non-magnetic regions in the sample, respectively. We show below that the non-magnetic regions of the sample are superconducting. The pronounced rapid decay of the muon asymmetry at early times is modelled by a Gaussian oscillation with an enhanced relaxation rate consistent with muons stopping in magnetically ordered regions. The magnetic and the SC components have rotation frequencies \( \omega_m \) and \( \omega_{SC} \), and Gaussian decay parameters \( \sigma_m \) and \( \sigma_{SC} \). For details on the fitting function see SM [27].

Fig. 3 shows the parameters extracted from fits to the ND and μSR data, along with parameters from magnetisation measurements. In panel (a), we display the temperature dependence of the integrated intensity of the ND peak measured at ThALES. The magnetic ordering temperature is determined to \( T_N = 39(1) \) K, for both 6 T and 12 T.

Fig. 3 (b) displays \( V_m \), as a function of temperature extracted from the μSR data. The fast relaxation associated with magnetic order vanishes at 40(1) K. Panel (c) shows that the relaxation rate \( \sigma_{SC} \) of the SC regions takes a constant value 0.47 \( \mu s^{-1} \) at higher temperatures. On cooling below 32 K, \( \sigma_{SC} \) reaches a value of 0.95(5) \( \mu s^{-1} \). In panel (d) the rotation frequency of the muons in the SC regions is seen to be constant at high temperature, with a value that corresponds to the external magnetic field. The small negative shift of \( \omega_{SC} \) below 38 K together with the increase of \( \sigma_{SC} \) appearing below \( T_c \) is typical for SC and can be understood in terms of a broadening of the field distribution due to the formation of a flux line lattice within the SC state [39]. We thus ascribe this non-magnetic component at low temperatures to muons stopping in SC regions of the sample. There is no evidence of a third component with non-SC or non-magnetic properties.

The temperature dependence of the sample magnetization was measured with a vibrating sample magnetometer (VSM) by cooling in a magnetic field applied along the c-axis. We measured for 7 different field values in the range 0.1–9 T and found \( T_c \) as a function of applied field.

The transition from the normal to the SC state broadens with applied field. We observe that \( T_c \) decreases from the zero-field value 38(2) K with fields up to \( \sim 5 \) T after which it stabilizes at a value of \( \sim 34 \) K. Raw data and details on the fitting procedure are shown in SM [27].

The magnetic signal appears, within error, at the same temperature independent of applied field (6 T, 8 T, and 12 T) measured with both ND and μSR, see Fig. 3 (a-b). The SC transition \( (T_c) \) found with magnetization measurements coincides with the onset temperature found by μSR at 8 T as seen in Fig. 3 (c) compared to (c-d). At low fields, the SC and magnetic phases have the same transition temperature, but at higher fields the two transition temperatures differ from each other, as \( T_N \) remains unchanged while \( T_c \) is suppressed with field. This result is consistent with findings in LSCO with \( x = 0.10 \) and \( T_N \sim T_c (H = 0) = 30 \) K [16].

The ND signal at low temperature increases linearly with magnetic field, see Fig. 1. Theoretically, the measured intensity is proportional to the magnetic volume.
fraction, $V_m$, multiplied by the square of the component of the ordered moment perpendicular to the scattering vector [40]. At the same time, our $\mu$SR experiments show a pure SC phase at zero field and a linear increase of a $V_m$ with applied field. Combining the two techniques, constituting a local probe and a coherent probe, we conclude that the observed increase in the ND signal is caused by an increase in $V_m$, and not by an increase of the ordered moment.

In zero field, a small ND signal is present, while no static magnetism is observed with $\mu$SR. This small discrepancy can be explained by the difference in time scale between the two techniques [22, 41, 42], which is further described in SM [27].

While the concomitance of $T_N$ and $T_c$ at low fields might be seen as indication of one correlated phase, we interpret the different on-set temperatures at higher field values as a signature of two competing phases, magnetic and SC, with almost degenerate free energy at zero fields. The application of a magnetic field tips the balance in the energy landscape, suppressing $T_c$ and favoring the formation of regions with quasi-static m-AFM order.

Extrapolating our $\mu$SR results to higher fields in Fig. 1, $V_m$ reaches 100% at $B_{sat} \sim 23$ T, suppressing the SC phase completely. This suppression is also seen in the diamagnetic response of the magnetization measurements with applied field. A crude extrapolation of the fall in diamagnetic response to higher fields is consistent with a complete suppression of SC at 23 T, see SM [27].

Previous ND investigations of LSCO in samples that exhibit 100% magnetic volume fraction in zero field [21, 37] found a field-enhanced intensity which was ascribed to an increase in the ordered moment of pre-existing magnetic order. A similar interpretation is presented for LCOO in Ref. 26. In contrast, the linear increase in the magnetic scattering in our sample intensity can be fully accounted for by the linear increase in the magnetic phase fraction derived from the muon signal, leaving no evidence for an increase in the local moments.

This raises an apparent contradiction. While the phenomenology of the field-enhanced scattering in the two systems seem incompatible with a single mechanism, it seems unlikely that the same effect in two such similar systems is fundamentally different. Below we present an interpretation of the available data which allows for a single description favoring the phase fraction description.

There are only a few studies of the field dependent magnetic scattering that include both muon and neutron data. This makes it difficult to construct a direct connection between phase fractions and the magnetic scattering intensity. An exception is the combined ND and muon study of LSCO samples of $x = 0.105$, 0.12, and 0.145 in Ref. 21. Of particular note is the LSCO sample with $x = 0.12$ which shows significant field enhanced scattering but near 100% magnetic phase fraction even in zero field. However, muons do not measure local moments directly, but the local magnetic field which is influenced by moments arrayed over a range of around 10-20 Å. Ref. 21 notes that the magnetic structure for the $x = 0.12$ sample may be inhomogenous at this length scale.

Thus, a possible connected picture emerges. In the LSCO sample reported here, phase separation creates large magnetic and SC regions, and the application of a magnetic field increases the magnetic fraction in a straightforward manner. In the LSCO sample with $x = 0.12$, a frustrated tendency towards phase separation leads to inhomogeneity on a sub-nanometer scale. Application of a field again favors formation of the striped magnetic region, but in this case it “fills in the gaps” creating a more homogeneous phase and larger local fields at the muon sites.

It would be interesting to contemplate if a similar explanation of a field-induced shift in balance between the volume of two competing volume fractions could be relevant also for the superconductor YBa$_2$Cu$_3$O$_{6.45}$, where $T_N \sim T_c$ and a linear field-dependence of the magnetic neutron diffraction signal is observed [43].

In conclusion, we have found that La$_{2-x}$Sr$_x$CuO$_{4+y}$ with $x = 0.06$ shows no magnetic order at low temperature, and that the application of a small magnetic field induces long-range m-AFM ordered regions. The volume of these regions is proportional to the applied field, while the ordered magnetic moment remains constant. These findings are in contrast to the interpretation of earlier data on field-induced magnetism in oxygen-stoichiometric LSCO samples. This makes it relevant to re-investigate with $\mu$SR whether the field-enhanced m-AFM signal in the other La-214 cuprates is due to an increase of the ordered moment, as previously concluded, or rather caused by an increased magnetic volume fraction. The answer to this is highly relevant for the understanding of the interplay between magnetism and SC in the cuprate superconductors.

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Supplementary material: Field-induced electronic phase separation in a cuprate high temperature superconductor

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SAMPLE SYNTHESIS AND CHARACTERIZATION

A large single crystal was grown at the Department of Energy Conversion and Storage, Technical University of Denmark, in a mirror furnace using the traveling solvent floating zone method. After cutting, we obtained a 20 mm long cylinder with a diameter of 10 mm and a mass of \(m = 5107\) mg. The nominal Sr content of the material (from the mixing of powders preceding the growth of the single crystal) was \(x = 0.06\).

We measured the magnetic response of the as-grown sample and found a diamagnetic signal below \(\sim 6\) K, as shown in Fig. 1. This value of the critical temperature is consistent with other Sr doped samples with \(x \sim 0.06\) [1, 2].

The sample was oxygenated at University of Connecticut cut through a wet-chemical technique for several months. Prior to oxygenation the sample was cut to a smaller size (5 g), and the magnetisation data has not been corrected to account for the change related to the sample shape or size. The absolute values of the two magnetization curves in Fig. 1 can therefore not be compared directly.

The \(\text{La}_{1.94}\text{Sr}_{0.06}\text{CuO}_{4+y}\) crystal has \(a \simeq b = 5.33\) Å, and \(c = 13.15\) Å in orthorhombic notation.

OXYGEN MOBILITY AND COOLING RATES

Excess oxygen has been shown to be mobile in this family of compounds, and e.g. quench cooling of the sample from 200 K or 300 K yields strongly different low-temperature properties than slow cooling [3, 4]. For this reason, all experiments in the main paper and here were performed using a slow cooling rate (1 K/min or lower) from room temperature to 100 K in order to provide consistency between measurements, and to make sure the mobile excess-oxygen find an optimal configuration in the crystal structure before freezing. The sample was heated to at least 50 K (above \(T_c\) and \(T_N\)) before any change of magnetic field value and was therefore field cooled into the superconducting state in all experiments (neutron diffraction, muon spin rotation, and magnetisation measurements).

We note in passing that the magnetic properties of the sample measured with neutrons does not change over time on a scale of years as the data from RITAII and ThALES (see Fig. 1 in the main text) are in complete agreement, although taken in 2010 and 2016, respectively. This indicates that there is no significant change of the oxygen content, despite storing the sample in air at ambient temperature.
MAGNETISATION MEASUREMENTS AND THE SUPERCONDUCTING PHASE

In Fig. 2, we display the measured magnetisation. The VSM measurements provide insight into the magnetic response of the sample, which at low temperatures is dominated by diamagnetism due to the superconducting phase of the sample for all applied fields in this study \((B \leq 9 \, \text{T})\). The precise determination of a single \(T_c\) is made difficult by thermal fluctuations \([5]\) that broaden the transition from the normal state to the superconducting state. We therefore introduce \(T_{c,1}\) and \(T_{c,2}\) as shown in Fig. 2, where the crossing points of the sloping lines with the high temperature constant and the low temperature constants as \(T_{c,1}\) and \(T_{c,2}\) respectively, as done in previous studies \([6]\). \(T_{c,1}\) is referred to as \(T_c\) in the main text as this is our estimate of the onset of SC.

The two characteristic temperatures are extracted for each field and plotted in the phase diagram shown in Fig. 3. Below the \(T_{c,2}(H)\) line the diamagnetic signal is saturated which we interpret as the manifestation of a fully developed vortex lattice state. Above this temperature, an intermediate regime which we denote vortex gas marks a region where magnetism decreases approximately linearly with temperature. In this region, the diamagnetic response is not yet fully developed. The upper phase boundary \((T_{c,1}(H))\) marks the transition to a normal state.

It is clear from the raw data in Fig. 2 that the diamagnetic response becomes weaker with applied field. The value of the intersection between the fit to the sloping section and the second axis \((T = 0 \, \text{K})\) approaches zero as the applied field is increased. A plot of this is shown in Fig. 4. If we make a crude extrapolation of the intersection values to higher fields, it is reasonable to think that the diamagnetic effect disappears (or is rather small) around 23 T consistent with a magnetic volume fraction \((V_m)\) of the sample is 100% found with \(\mu\)SR (see Fig. 1 in the main text). The dependence of the intersections is not linear on single nor double logarithmic scale.

In Fig. 5 we show the value of the low temperature magnetisation normalised to the applied magnetic field. The normalised magnetisation is identical to the field-average (or effective) susceptibility, \(\chi_{\text{eff}}\). We observe that the absolute value of \(\chi_{\text{eff}}\) decreases with applied field and can almost be described with a power law; a straight line on the double-logarithmic plot in Fig. 5. The reduction of the diamagnetic signal with applied field is due to a combination of two effects; firstly, a reduction of the superconducting volume and secondly, an increasing pinning strength, \(\text{e.g.}\) from defects preventing pinned magnetic flux to exit the sample, thus reducing the dia-
MUON SPIN ROTATION

With the \( \mu \)SR technique the time-dependent decay of a spin-polarized ensemble of muons is measured. The muons enter the sample and reside at one unique interstitial lattice site which makes the \( \mu \)SR technique a local probe that is sensitive to magnetic fields down to 10 \( \mu \)T inside the sample \([8, 9]\). The positive muons are initially completely spin polarized, and after implantation they precess in the local magnetic field with a gyromagnetic ratio of \( \gamma_\mu = 2\pi \times 135.5 \text{ MHz T}^{-1} \). Muons are unstable particles, and decay into a positron and a neutrino-antineutrino pair within microseconds. The positron is emitted preferentially along the direction of the muon spin, and the recorded decay positron count rate thus monitors the spin precession corresponding to the magnetic field strength at the muon site. The \( \mu \)SR spectra obtained in zero field conditions cover a time window of about \( 10^{-6} - 10^{-9} \) s, which means that magnetic fluctuations faster than this are averaged to zero, while magnetic fluctuations slower than this are perceived as being static. The average muon implantation depth is of the order 100 \( \mu \)m, and hence the magnetic and superconducting properties measured with muons are representative of the bulk sample.

The HAL9500-spectrometer has 8 detectors in front of the sample position and 8 detectors behind. The muon precession phase, normalization-, and background counts of each detector are fitted separately with the model key parameters asymmetry, relaxation rates, and oscillation frequency shared between all detectors. The positron count rate in each detector is therefore fitted to the following function

\[
N_i(t) = N_{i0} \exp\left(-t/\tau_\mu\right) (1 + A(t)) + N_{bgi},
\]

(1)

where \( N_i(t) \) is the number of positrons detected at time \( t \) in detector \( i \), and \( N_{i0} \) and \( N_{bgi} \) are the initial number of positrons and the background count in the detector, respectively. \( \tau_\mu \) is the average muon life time of 2.2 \( \mu \)s and from the decaying asymmetry function, \( A(t) \), physical parameters such as strength and distribution of the magnetic field within the sample can be extracted from fits to the models presented below. In this work we used the unique possibility of the HAL9500 instrument to apply a magnetic field up to 8 T (high-field) to the sample.

A zero field experiment was performed on the GPS...
spectrometer. We used the forward (in front of sample) and backward (behind the sample) detector with respect to the muon beam direction to derive the muon spin asymmetry defined as

$$A(t) = \frac{N^I(t) - \alpha N^b(t)}{N^I(t) + \alpha N^b(t)},$$  

(2)

where $\alpha$ is a scaling factor to account for different efficiency and solid angle of the two detectors.

Zero field $\mu$SR results

Fig. 6 shows a ZF-$\mu$SR spectrum measured at 5 K. The data are fitted by a Gaussian Kubo-Toyabe-function (KT)

$$A(t) = \frac{1}{3} + \frac{2}{3}(1 - \sigma^2 t^2) \exp(-\sigma^2 t^2/2).$$  

(3)

From this we find $\sigma = 0.168(1) \mu m^{-1} \sim \gamma_\mu \Delta$, where $\Delta$ is the second moment of the magnetic field distribution at the muon site [10].

A Gaussian KT function models the relaxation of muons stopping in a non-magnetic environment with static and randomly oriented nuclear moments. There is a slight deviation between data and model for long decay times ($> 6 \mu s$), which indicates that the moments may be fluctuating slightly or that the system is not completely disordered, in agreement with the ND data shown in the main paper.

**FIG. 6.** $\mu$SR spectrum recorded under zero field conditions at 5 K. The red line is the result from a Gaussian Kubo-Toyabe-function fitted to the data.

High transverse field $\mu$SR results

The high transverse field $\mu$SR data has been fitted with a two-component model with the following function represented in a rotating reference frame;

$$A(t) = V_m \exp \left( -\sigma_m^2 t^2 / 2 \right) \cos(\omega_m t + \phi^I)$$
$$+ (1 - V_m) \exp \left( -\sigma_{SC}^2 t^2 / 2 \right) \cos(\omega_{SC} t + \phi^I),$$  

(4)

where $\omega_m$ and $\omega_{SC}$ are the Larmor frequencies of the muon spin and $\sigma_m$ and $\sigma_{SC}$ are the second moments of the magnetic field distribution at the muon site for two distinct regions of the sample.

The magnetically ordered regions are denoted with subscript $m$, and the superconducting regions are denoted with the subscript SC. The volume fraction of the sample with ordered electronic moments is parameterized by $V_m$. $\phi^I$ is the initial phase of the muon spin orientation determined by the detector geometry.

A selection of raw data and fits shown in rotating reference frames, can be found in Fig. 2 (b-d) of the main text. We performed a temperature dependence study with an applied field of 8 T, and a field dependence study performed at 5 K. The resulting parameters, not found in the main text, are presented in Fig. 7 and 8.

In Fig. 7 the relaxation rate of the magnetic regions of the sample are shown as a function of temperature. The relaxation rate decreases as a function of temperature and reaches zero at $T_N$. The rotation frequency in the magnetic regions, $\omega_m$, is fixed to 1084.6304(8) MHz and was found by fitting the 50 K data. The found value is slightly above the Larmor frequency of the muon in an external magnetic field of 8 T (1084.3104 MHz). The other three fitting parameters ($\sigma_{SC}$, $\omega_{SC}$, and $V_m$) can be found in the main text in Fig. 3 (b-d).

The rotation frequency in the magnetic and superconducting regions, $\omega_m$ and $\omega_{SC}$, as a function of applied magnetic field both has a linear dependence of field with a slope of 135.5388 MHz/T as seen in Fig. 8 (a) and (c).

**FIG. 7.** The relaxation rate of the magnetic regions of the sample as a function of temperature (see eq. (4)). The sample was field cooled in an applied field of 8 T.
FIG. 8. Parameters found from the two-component fits (eq. (4)) as a function of applied field. $V_m$ as a function of field can be found in the main text in Fig. 1. The sample was heated and field cooled for each value of the field. The magnetic rotation frequency is found to be about 0.2 MHz above the superconducting frequency (not visible on a scale from 0-1000 MHz).

The relaxation rate of the magnetic fraction, $\sigma_m$, is constant at around 4.5 $\mu$s$^{-1}$ and has a slight upturn at low magnetic fields (Fig. 8 (b)). The superconducting relaxation rate is constant around a value of 1 $\mu$s$^{-1}$ and does not seem to follow $\sigma(H)/\sigma(H = 0) = 1 - K\sqrt{H}$ as described in Ref. [11]. However the statistical errors assigned to the data from the $\chi^2$-fits are clearly not representative of the true error of the values of the relaxation rates. It is possible that our data quality is not adequate to determine whether or not there is a small reduction of the superconducting relaxation rate as a function of applied field.

**NEUTRON DIFFRACTION**

Neutron diffraction was performed at the cold-neutron triple axis spectrometers RITA-II at the Paul Scherrer Institute (PSI), Switzerland and ThALES at Institut Laue-Langevin (ILL), France [12]. With a constant final energy of 5.0 meV, the elastic energy resolution of the two experiments was close to 0.2 meV, while the $q$-resolution was around 0.025 r.l.u. FWHM at ThALES and around 0.015 r.l.u. FWHM at RITA-II.

RITA-II was configured in the monochromatic imaging mode [13, 14], giving an effective collimation of open-80'-40'-open. No collimation was used on ThALES, but the analyzer geometry was unfocused (flat) and boron containing shielding was mounted to cover all but the central analyzer crystal, in order to enhance the ratio of signal to noise. For both experiments, a Be-filter was placed between sample and analyzer to filter out scattering from second-order scattering.

In the RITA-II data from 2010 shown in Fig 2 of the main text, the level of the background, on which the magnetic peaks reside, is seen to change upon application of a field. In contrast, no field dependence is found of the background in the ThALES data from 2016. This could be explained by at least two factors. a) We performed a few re-adjustments during the week-long RITA-II experiment to attempt to maintain perfect alignment. In contrast, in the ThALES experiment, we performed no realignment during the much faster measurements, but only checked the crystal orientation before and after the full measurement series. b) In the RITA-II experiment, the peak scans were performed along the $hk$-direction in reciprocal space, which requires a small adjustment of scattering angle (A4) for each point of the scan. In contrast, in the ThALES experiment we performed the scans as pure sample-rotation (A3) scans. Here, the scan direction is along a (slightly) curved path in reciprocal space, but there is no change in the scattering angle (A4). Since we modified the experimental procedure in two ways, we cannot tell which of the two factors was decisive for obtaining the more constant background in the ThALES experiments. Either way, we conclude that the changing background in the RITA-II experiment is an artefact of the experimental procedure.

**Inter-planar magnetic correlations**

To measure the inter-planar magnetic correlations along the $l$-direction the sample was mounted with the $c$-axis in the scattering plane. The magnetic field was applied along $l$ in a horizontal field magnet with a maximum field strength of 5.5 T. The experiment was carried out at the RITA-II spectrometer with the same configuration as described above. The sample was oriented so that the (1.125$h$,0.125$h$,0) direction was contained in the scattering plane together with (0,0,$l$). Sample rotation (A3) scans were performed above $T_N$ and subtracted from the low temperature data at 2.3 K. Fig. 9 shows the final $l$-dependence obtained by binning amplitudes found from Gaussian fits to the background subtracted data, along $l$ with a step size of 0.3 r.l.u.
FIG. 9. Neutron diffraction intensities of the m-AFM peak along the l-direction measured at 2.3 K.

Even with a counting time of 48 minutes per point it was not possible to gather enough statistics to obtain a reliable position and width of the m-AFM peak from the measured data. This is largely due to mismatch between the instrument resolution with poor out of plane resolution and the rod-like signal along l from the m-AFM scattering. In contrast, the resolution matches the rod-like signals when the sample is aligned with a and b in the scattering plane [15]. The signal from the m-AFM domains is also weak as the applied field of 5.5 T only induces magnetic order in a small fraction of the sample (~20%).

The data shows a field-induced m-AFM signal but no intensity modulation along l. We therefore conclude that there is no significant correlation along the c-direction of the field-induced m-AFM order. In LSCO, we previously found l-modulations with a period of 2 [16]. It is however also possible that any modulation of the neutron signal from a single (magnetic) domain could be washed out, and thus not observed, due to a scattering signal from another magnetic domain (e.g. due to twinning in the crystal) with large intensity at l-values which had low scattering intensity in the first domain [16].

**Structural modulation: Staging**

Apart from paving the way for the electronic phase separation in the m-AFM and superconducting phases, the excess oxygen also causes anti-phase boundaries of the CuO₆ octahedra tilt pattern along the c-axis, usually denoted as "staging" due to the similarity of intercalated (staged) graphite [17]. The periodicity of the anti-phase tilt patterns can be observed as (pairs of) superstructure peaks when scanning through Bmab allowed peaks along l, as shown in Fig. 10. Previously, it has been shown that the (main) staging peak position shifts closer to the

![Image](image-url)
our sample is positioned at the cross-over point between annealed (O-doping) and quenched (Sr-doping) doping where the local doping levels are so even that no spatially separated domains of doping levels large enough to induce magnetism. A systematic study of this effect could be interesting.

Raw data and fits

Fig. 11 shows raw data of sample rotation scans measured at ThALES. The background points have been counted for 30 seconds/point and the peak for 4.5 minutes/point for the 0 T data. The high temperature data is a combination of several temperatures and fields in the range 6-12 T and 40-50 K to gain an even better statistics. The data has been fitted with a Gaussian with a sloping background and the centers and widths are locked to the values found from a Gaussian fit to all data (all field values) measured at 2.9 K combined. The resulting fit from the measurement at 2.9 K and 12 T has been plotted to show how large this signal is compared to the signal from the 0 T data. The high field fit also acts as a visual representation of the locked width and center of the fits.

There are ten points in the background on each side of the a-AFM peak position so even if the individual points have a large uncertainty the determination of the sloping background from the points is rather accurate. The low temperature (0 T and 2.9 K) data points in the peak lie systematically and significantly above the high temperature data points, indicating that there is a small signal present in the sample in zero field when measured with neutrons.

TIME-SCALES FOR DIFFERENT PROBES

In Fig. 1 in the main text we show that a small neutron signal is present, while no static magnetism is observed with μSR in zero applied filed. This small discrepancy may be due to the difference in time scale of the two techniques. Cold neutron measurements probe a timescale of $\sim 10^{-11}$ s and can not distinguish fluctuations on longer timescales from static magnetism. In contrast, high transverse field μSR measurements probe a timescale of $\sim 10^{-7}$ s. Magnetic moments fluctuating on a time scale between these two values would thus be seen as static by the neutrons, but as fluctuating by the muons. The difference between the data obtained by the two techniques may thus be reconciled if the m-AFM is not truly static, but rather slowly fluctuating.

X-RAY STUDIES OF CHARGE ORDER

We performed two hard X-ray diffraction experiments at the BW5 beamline at HASYLAB, Deutsches Elektronen-Synchrotron, Hamburg, Germany. Here we looked for a signature of the charge-density wave (CDW), observed in LSCO samples with dopings close to $x \lesssim 1/8$ at $(h \pm 0.25, h \pm 0.25, l)_O$ [20]. The sample was cut into a thin slice ($d = 1.4$ mm) in order to reduce the absorption, meaning that 10 % of the incoming beam was transmitted through the sample.

There are ten points in the background on each side of the a-AFM peak position so even if the individual points have a large uncertainty the determination of the sloping background from the points is rather accurate. The low temperature (0 T and 2.9 K) data points in the peak lie systematically and significantly above the high temperature data points, indicating that there is a small signal present in the sample in zero field when measured with neutrons.
In the first experiment, March 2011, we searched for a field-induced signal at $(\pm0.25, \pm0.25, l)_{O}$ for $l$-values of 8, 8.25, 8.5, 9.5, 11.5, and 12.5. Hints of a weak signal was seen at $(\pm0.25, \pm0.25, 8.25)_{O}$, but at no other $l$-values. A second, and more focused experiment was performed in November 2011. As it can be seen in Fig. 12, the findings from March were not reproduced, even with unusually long counting times of 7.5 min/point. We therefore conclude that there is no signature of field-induced CDW signal in our sample when probed with hard X-ray diffraction.

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