Two-Dimensional Magnetic and Superconducting Phases in Metal-Insulator La$_{2-x}$Sr$_x$CuO$_4$ Superlattices Measured by Muon-Spin Rotation

A. Suter, E. Morenzoni, T. Prokscha, B. M. Wojek, H. Luetkens, G. Nieuwenhuys, A. Gozar, G. Logvenov, and I. Božović

Phys. Rev. Lett. 106, 237003 — Published 8 June 2011
DOI: 10.1103/PhysRevLett.106.237003
Magnetism in the 2D Limit and Interface Superconductivity in Metal-Insulator \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) Superlattices

A. Suter, E. Morenzoni, T. Prokscha, B. M. Wojek, H. Luetkens, G. Nieuwenhuys, A. Gozar, G. Logvenov, and I. Božovič

1Laboratory forMuon Spin Spectroscopy, Paul Scherrer Institute, 5232 Villigen PSI, Switzerland
2Physik-Institut, Universität Zürich, 8057 Zürich, Switzerland
3Brookhaven National Laboratory, Upton, New York 11973-5000, USA

(Dated: May 3, 2011)

We show, by means of low-energy muon spin rotation measurements, that few-unit-cells thick \( \text{La}_2\text{CuO}_4 \) layers synthesized digitally by molecular beam epitaxy synthesis are antiferromagnetically ordered. Below a thickness of about 5 \( \text{CuO}_2 \) layers the long-range ordered state breaks down, and a magnetic state exists in close proximity (few Å) to high-temperature superconducting layers, without transmitting supercurrents.

By reducing the dimensionality of a solid, its electronic states and physical properties can be drastically modified, but these changes are not easy to predict for strongly correlated electron materials. For example, in thin interfacial layers inside oxide heterostructures a host of electronic states were discovered experimentally – a high-mobility 2D electron gas [1], magnetism [2], quantum Hall effect [3], and interface superconductivity between insulators [4]. In metal-insulator (MI) bilayer \( \text{La}_{3.55}\text{Sr}_{0.45}\text{CuO}_4/\text{La}_2\text{CuO}_4 \) heterostructures (LSCO-LCO), where none of the constituents is superconducting, interface superconductivity with \( T_c \approx 30 \text{ K} \) has been discovered recently [5].

Up to now these studies used probes that are sensitive to charge but tell little about the microscopic magnetic state. For instance although it is known that 1 unit cell of LCO sandwiched between two optimally doped LSCO layers is still insulating [6], one can only speculate about its magnetic state. LCO is assumed to be close to the realization of a spin-1/2 isotropic Heisenberg antiferromagnet on a square lattice (2DHAF), since its in-plane exchange constant, \( J \), is about \( 10^4 \) times larger than any other exchange coupling present. Bulk material shows antiferromagnetic (AF) long-range order (LRO) below a Néel temperature of \( T_N \approx 310 \text{ K} \) [7, 8]. In thin films reducing the thickness results in a decreased \( T_N \) [9], whereas strain seems to play only a minor role [10]. In the 2D limit, at any finite temperature LRO will be destroyed by thermal fluctuations [11, 12]. P.W. Anderson proposed that for the 2DHAF [13] even at \( T = 0 \) quantum fluctuations destroy LRO; instead, a quantum spin-liquid – the resonating valence bond (RVB) state – should form. Chakravarty, Halperin, and Nelson [14] solved the 2DHAF in the long wave limit and arrived at a different picture. The phase diagram is basically controlled by the temperature and the spin stiffness, \( \rho_S \), and only part of the phase diagram is dominated by quantum fluctuations (quantum disordered regime), whereas in the other part the spin correlation length, \( \xi(T) \), grows exponentially by lowering the temperature (renormalized classical regime, RC). Indeed, measurements of \( \xi(T) \) in the paramagnetic phase (\( T > T_N \)) of bulk LCO revealed that it follows the RC behavior [15, 16]. While numerical simulations support the long-wave-limit calculations [17, 18], it has been argued [19–22] that small deviations from the ideal 2DHAF, due to frustrating second-neighbor exchange, charge carrier doping, defects, etc., could reduce \( \rho_S \) and thus enhance the effect of quantum fluctuations, preventing the spins from acquiring LRO.

In this Letter we present a study focusing on the magnetic state of LCO layers within MI LSCO-LCO superlattices (SLs), where the number of \( \text{CuO}_2 \) layers within the LCO stack can be varied to approach the 2D limit. To probe AF order and magnetic fluctuations we used polarized low-energy muons as a local probe. Low-energy muon spin rotation (LE-\( \mu \)SR) [23] can detect superconductivity and/or magnetism, either static or fluctuating, even in ultrathin layers [24]. We show that down to about 5 \( \text{CuO}_2 \) layers LCO acquires LRO at low enough temperatures. Below this thickness, LCO enters a different magnetic state, characterized by short-range correlations, and increased magnetic fluctuations. This indicates a cross-over to a quantum disordered regime in this 2DHAF model system. Furthermore, we show that this magnetic state exists in close spatial proximity to superconducting layers.

We have synthesized and studied a series of samples that contain ultrathin, isolated layers of LCO. The synthesis was carried out by means of an atomic-layer-by-layer molecular beam epitaxy (ALL-MBE) system equipped with in-situ surface science tools. ALL-MBE allows for synthesis of complex heterostructures in which the thickness of individual layers can be controlled down to a single atomic layer [6, 25]. We digitally varied the thickness of LCO layers alternating with metallic \( \text{La}_{1.56}\text{Sr}_{0.44}\text{CuO}_4 \) (LSCO) layers. Counting in 1/2-unit-cell (UC) increments, each of which contains a single \( \text{CuO}_2 \) plane, the investigated SLs have the re-
precess in the internal field, contributions at the muon stopping site and their static and decays) yields information about local magnetic field dis-

zuon decay spectra (typically from a few million muon

from the spontaneous zero-field precession signals (Fig.

In [3LSCO+12LCO] we observe static AF order, evident

narrowing regime [29]).

To detect magnetism we performed μSR experiments

as a function of temperature under zero-field conditions

(ZF). To quantify the magnetic volume fraction and the

robustness of the magnetic state, as well as to character-

ize superconductivity we applied small magnetic fields

parallel and perpendicular to the \(ab\)-planes, always per-

pendicular to the muon spin (“transverse field”, TF). For
each muon spin rotation measurement, a mosaic of four

nominally identical \(1 \times 1 \text{ cm}^2\) samples was used. Fig. 1d

shows the muon stopping distributions as used in the

experiments. The time evolution of the polarization of

the muon ensemble \(A_0 P(t)\), which is obtained form the

muon decay spectra (typically from a few million muon
decays) yields information about local magnetic field dis-

tributions at the muon stopping site and their static and
dynamic properties. In case of AF LRO, muon spins precess

in the internal field, \(B_{\text{int}}\), of the electronic mag-

natic moments with a frequency, \(\nu_\mu = (\gamma_\mu/2\pi) B_{\text{int}}\) (\(\gamma_\mu\)
is the gyromagnetic ratio of the muon), proportional to

the staggered magnetization, and oscillations at this fre-

quency show up in the polarization spectra. The pres-

ence of substantial magnetic disorder (e.g., a frozen spin
glass state) or electronic low-frequency (< 10 MHz) fluc-
tuations leads to a strongly damped \(A_0 P(t)\) due to a

rapid dephasing of the muon spin ensemble. However,
electronic high-frequency fluctuations (> 100 MHz) will

only lead to a weak depolarization of \(A_0 P(t)\) (motional

narrowing regime [29]).

ZF polarization spectra are shown in Figs. 1a and 1b

for [3LSCO+12LCO] and [3LSCO+9LCO], respectively. In

[3LSCO+12LCO] we observe static AF order, evident from

the spontaneous zero-field precession signals (Fig.

1c). The internal field, \(B_{\text{int}} = 39.0(8) \text{ mT}\), is equal to

what is observed in single-phase LCO films [9] and bulk

samples [30], thus showing that the full electron magnetic

moment is present. The magnetic volume fraction esti-
mated from the oscillatory amplitude of \(A_0 P(t)\) shows that

about 1/4 of the film volume is magnetically or-
dered. From this, we can estimate the magnetic layer

thickness to be \(d_{\text{mag}} \approx 3 \text{ nm} (4–5 \text{ CuO}_2 \text{ layers})\), which is

in quantitative agreement with simple model calculations
taking into account Sr interdiffusion and charge redistrib-

ution between the M and I layers [5, 28]. According to

the model, the inner 5 CuO\(_2\) layers have doping levels of

\(x < 0.006\) and lie well within the AF part of the phase

diagram (\(T_N \to 0\) for \(x \gtrsim 0.02\)). The model also pre-
dicts that one or two nominally insulating CuO\(_2\) planes

at the interface will have doping levels corresponding to

FIG. 1. (a)–(c) \(\mu^+\) spin-polarization spectra, \(A_0 P(t)\), for zero

applied magnetic field. (a) \(A_0 P(t)\) for the [3LSCO+12LCO] SL. The

\(T = 40 \text{ K}\) spectrum (upper curve, red online) is

Gaussian like, whereas the \(T = 5 \text{ K}\) (lower curve, blue

online) is more exponential-like, which indicates enhanced

spin dynamics at lower temperatures. (b) \(A_0 P(t)\) for the

[3LSCO+9LCO] SL. Here \(A_0 P(t)\) shows only a very weak

additional exponential contribution at \(T = 5 \text{ K}\) compared to

[3LSCO+12LCO], and no zero-field precession is observable.

For details see the text. (c) enlarged scale and different bin-
ing for [3LSCO+12LCO] showing zero-field precession sig-

als, i.e. a well defined static internal field at the muon

site. The \(T = 40 \text{ K}\) and \(T = 200 \text{ K}\) curves are shifted up

by 0.02 for clarity. (d) The \(\mu^+\) stopping distributions \(n(z)\),

used in the experiments. The yellow stripes represents the

LCO, the green ones the LSCO within the SL, shown for

[3LSCO+12LCO].
the superconducting part of the LSCO phase diagram.

Our measurements of the superconducting properties of these SLs by LE-μSR provide a further independent confirmation of the presence of interface superconductivity and of the charge levels in the SLs. From the absence of Meissner screening of a magnetic field applied parallel to the SL (ab-planes) we infer that no supercurrents flow along the c-axis, consistent with superconductivity being restricted to the interface. The London penetration depth, \( \lambda_L \), was estimated from the increased muon depolarization rate below \( T_c \) when field-cooling the sample in a field applied perpendicular to the ab-planes. Assuming a pancake vortex model [31, 32] which takes into account the layered structure of the SLs, we find \( \lambda_L \approx 350 \text{ nm} \) which is about 1.5 times larger than \( \lambda_L \) of optimally doped bulk LSCO [33]. Using the clean limit relation for the superfluid density \( n_S = \frac{\mu_0}{\pi^2} \frac{n}{\lambda^2} \), we find an averaged superfluid density of about half the value of optimally doped LSCO, again in satisfactory agreement with the charge transfer model.

In contrast to the [3LSCO+12LCO] SL no signs of spontaneous ZF precession — and hence no evidence for static AF LRO — are found down to \( T = 5 \text{ K} \) in [3LSCO+9LCO] and [3LSCO+6LCO]. Fig. 1b shows the ZF time spectra for the [3LSCO+9LCO] sample. At \( T = 40 \text{ K} \), \( A_0 P(t) \) shows a Gaussian depolarization typical for nuclear dipole fields. At \( T = 5 \text{ K} \) an additional very weak exponential component appears. The dash-dotted line shows the expected \( A_0 P(t) \), assuming a doping level \( x \) calculated from the charge redistribution model and using experimental parameters from measurements on single-phase films with the corresponding \( x \) values. Clearly, the predicted and measured spectra differ in two major features: the experimental data show no spontaneous precession and no sign of a fast initial depolarization is visible. The former points to the absence of static LRO, and the latter indicates that even static disordered magnetism, which would lead to a fast depolarization, is significantly suppressed.

To further investigate the magnetic state we estimated the magnetic volume fraction in the samples by measuring \( A_0 P(t) \) in a weak magnetic field applied transverse to the initial muon polarization. In this case \( A_0 P(t) \) can be written as [34]:

\[
A_0 P(t) = A_T \exp[-(\sigma t)^2/2] \cos(\gamma_\mu B_{tot} t + \phi) + A_L e^{-\lambda t} \cos(\phi),
\]

where \( A_T \), \( \sigma \) and \( A_L \), \( \lambda \) are the asymmetries and corresponding depolarization rates, transverse and parallel to the total field \( B_{tot} = |(B_{ext} + B_{int})| \), while \( \phi \) is the detector phase. \( \lambda \) was negligibly small in all measurements. \( A_L \) is a measure of the presence of static magnetism (ordered or disordered) and its volume fraction. For instance, in the case of static magnetic order with an underlying isotropic magnetic field distribution and 100% volume fraction \( A_L/A_0 \) will grow to 1/3. In contrast, \( A_T/A_0 \) would drop to zero in the magnetic phase. In any para- or diamagnetic sample, \( A_L \) will be identically zero at all temperatures. The resulting magnetic layer thicknesses can be estimated from the relation \( d_{mag} \approx (3 + n)/(c/2)(1 - A_T/A_0) \), where \( n = 6, 9, 12 \) depending on the SLs, and \( A_T/A_0 \) is shown in Fig. 2a. The estimated magnetic layer thicknesses for \( n = 6, 9 \) is \( d_{mag} \approx 0.4 - 1 \text{ nm} \) (≈ 1–2 CuO\(_2\) layers), again in agreement with the calculated charge distribution (inner layer doping estimate: [3LSCO+9LCO] \( x < 0.008 \), [3LSCO+6LCO] \( x < 0.025 \)). Since the inner layer doping of the [3LSCO+6LCO] is at the border of the AF region, it will not be discussed here.

For [3LSCO+9LCO] \( A_L \) is drastically reduced, and both \( A_L/A_0 \) and \( 1 - A_T/A_0 \) show only a small deviation from zero below 50 K. This behavior is typical for fast fluctuations where \( A_L \) is vanishing at all temperatures. We ascribe this difference, together with the absence of a ZF precession and the very weak initial drop of \( A_0 P(t) \) (Fig. 1b), to increased fluctuations in ultrathin LCO layers, which prevent the formation of either LRO or a static disordered magnetic state (e.g. spin glass).

These fluctuations are not expected within the RC regime. The following estimate indicates that they are of quantum nature. Within the RC regime \( \xi \) is given as: \( \xi(T)/a = 0.5 \exp(1/y) [1 - y/2 + O(y^2)] \), with \( y \) the in-plane lattice constant, \( y = k_B T/(1.13 J) \), and \( J/k_B \approx 1500 \text{ K} \) for LCO. At \( T \approx 150 \text{ K} \), \( \xi/a > 10^4 \) which should result in a quasi-static magnetic state, i.e. either ZF precession or, in the strongly disordered case, a strong initial depolarization should be observable. Both are absent in the [3LSCO+9LCO] and [3LSCO+6LCO] SLs down to the lowest temperature. The same conclusion is reinforced by the fact that the time-independent component \( A_T/A_0 \) is drastically reduced, when decreasing the number of CuO\(_2\) planes in LCO (Fig. 2b) and by application of increasing magnetic fields (Fig. 2c). Assuming a random static internal field within the CuO\(_2\) planes, the magnetic field dependence of \( A_L(b) \propto 1/(2[1 + b^2]) \) with \( b = B_{ext}/B_{int} \). The expected ratio \( R \equiv A_L(B_{ext,1})/A_L(B_{ext,1}) \) for \( B_{ext,1} = 3 \text{ mT} \), and \( B_{ext,2} = 10 \text{ mT} \) and the measured internal field of \( B_{int} = 39 \text{ mT} \) is \( R = 0.94 \), however, for the [3LSCO+12LCO] a value of \( R = 0.76(1) \) is found (see Fig. 2c). This drastic reduction can only originate from fluctuations and cannot be due to disorder. In order to see if unexpected doping, i.e. deviations from the simple charge-transfer model, could lead to such a strong modification of the magnetic state, we performed the same measurements on 53 nm thick single phase La\(_{1.95}\)Sr\(_{0.05}\)CuO\(_4\) films and find \( R = 0.95(2) \) in the so-called cluster-spin glass phase. This is in excellent agreement with the static model estimate, indicating that the strong reduction of \( R \) in the SLs is due to dimensional effects, i.e. increased magnetic fluctuations, and not due to charge-transfer ef-
FIG. 2. (a) Normalized transverse asymmetry $A_T/A_0$ as function of temperature for the different SLs, obtained from weak transverse field measurements in $B_{\text{ext}} = 10 \text{ mT}$. (b) Normalized longitudinal asymmetry $A_L/A_0$ versus temperature. Note: for non-magnetic samples $A_L \equiv 0$. The shaded area shows the region where the superconducting transition takes place: [3LSCO+9LCO], $T_c = 25.0 \text{ K}$; [3LSCO+12LCO], $T_c = 24.0 \text{ K}$. (c) $A_L/A_0$ versus temperature for different external fields $B_{\text{ext}}$ for the [3LSCO+12LCO] SL, for field cooling (FC) and zero-field cooling (ZFC). The pronounced reduction of $A_L/A_0$ between 3 mT and 10 mT is due to a strong reduction of the spin stiffness compared to bulk LCO.

Another estimate further supports our finding: from the known magnon dispersion $[35]$ in LCO one can put a lower limit on the magnon wavelength that can be thermally excited at $T = 5 \text{ K}$ to about 1 $\mu$m, and this is the length scale on which magnons would destroy static long-range AF order. Experimentally, we see the absence of LRO on the length scale of less than 5 nm (local order on this length scale would lead to ZF muon spin precession or strong damping), this requires the presence of very short-wavelength, high-energy ($\sim 100 \text{ meV}$) AF fluctuations, which at $T = 5 \text{ K}$ can only be of quantum nature.

All these findings show that LCO within these SLs is not in the RC regime (as is the case for bulk LCO for $T > T_N$), i.e. the spin stiffness of the AF state is drastically reduced. Currently we do not know what is the reason for this strong reduction of the spin stiffness, however we can rule out that it is caused by disorder.

Acknowledgments: The μSR experiments were fully performed at the SμS. The work at BNL was supported by the U.S. Department of Energy Project MA-509-MACA.

* andreas.suter@psi.ch
† present address: Max-Planck-Institut für Festkörperforschung, 70569 Stuttgart, Germany

[References]


