

Two-Dimensional Orbital-Like Magnetic Order in the High-Temperature $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ Superconductor

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In high-temperature copper oxide superconductors, a novel magnetic order associated with the pseudogap phase has been identified in two different cuprate families over a wide region of temperature and doping. We report here the observation below 120 K of a similar magnetic ordering in the archetypal cuprate $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO) system for $x = 0.085$. In contrast with the previous reports, the magnetic ordering in LSCO is only short range with an in-plane correlation length of $\sim 10 \text{ \AA}$ and is bidimensional (2D). Such a less pronounced order suggests an interaction with other electronic instabilities. In particular, LSCO also exhibits a strong tendency towards stripes ordering at the expense of the superconducting state.

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The origin of the pseudogap phase remains one of the most animated debates in the high-temperature copper oxide superconductors. Occurring in the normal state and over a wide region of doping, the pseudogap is visible in both magnetic and charge properties of all cuprates superconductors [1,2]. Using polarized neutron diffraction, a novel long range magnetic order has been recently established in two different cuprate families, $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ (YBCO) [3,4] and $\text{HgBa}_2\text{CuO}_{4+\delta}$ (Hg1201) [5]. This observation is of primary importance, since the temperature of this magnetic transition matches the one of the pseudogap regime, providing strong support in favor of a true phase transition [2,6]. While the novel magnetic order also breaks time reversal symmetry, it should be described as a $\mathbf{Q} = 0$ antiferromagnetic order (AFO), i.e., a magnetic order preserving the translation symmetry of the lattice (TSL), but with a staggered magnetic pattern present within each unit cell. This magnetic order has been predicted in the circulating current theory of the pseudogap state [7]. That suggests that this $\mathbf{Q} = 0$ AFO corresponds to an orbital-like order. In this theory, the magnetic moments are indeed associated with two opposite orbital moments per unit cell generated by closed current loops [7]. Recent variational Monte Carlo calculations in the extended Hubbard model show that orbital moments can indeed develop once the apical oxygen orbitals are taken into account [8]. However, the orbital nature of the observed magnetic moments is still an open experimental issue. Alternatively, spin-based models could actually describe the observed magnetic peaks [3].

In the archetypal HTS $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO), the low energy spin excitation spectrum is dominated by incommensurate (IC) spin fluctuations around the planar antiferromagnetic (AF) wave vector at $\mathbf{Q}_{\text{IC}} = \mathbf{Q}_{\text{AF}} \pm (\delta, 0) \equiv \mathbf{Q}_{\text{AF}} \pm (0, \delta)$ [9] with $\mathbf{Q}_{\text{AF}} = (1/2, 1/2)$. In isostructural compounds $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ [10] and

$(\text{La, Nd})_{2-x}(\text{Sr, Ba})_x\text{CuO}_4$ [11], where bulk superconductivity is strongly reduced, spin (SDW) and charge (CDW) density wave orders develop, respectively, at \mathbf{Q}_{IC} and $2\mathbf{Q}_{\text{IC}}$ [12]. Furthermore, IC spin excitations are also observed in strongly underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6.45}$ [13]. These fluctuations indicate that the rotation invariance of the system is spontaneously broken below 150 K. All of these properties can be understood within the charge stripe model [14], for instance.

While the $\mathbf{Q} = 0$ AFO (or magnetic orbital-like order) has been observed in a broad doping and temperature range in YBCO and Hg1201 systems, the static striplike order essentially develops in LSCO either at low doping near the Mott-insulating state or near the locking-in composition of $x = 1/8$ [10,15–17] reaching its maximum temperature of 20 K [15]. We report a study of the $\mathbf{Q} = 0$ AFO in one LSCO sample with 8.5% of Sr. At the difference of YBCO and Hg1201, the observed magnetic order is short-range.

The sample consists of three single crystals (total mass 7 g) obtained by the traveling solvent floating zone method similar to the ones reported by [18], coaligned within less than 1 deg. Most of the data have been obtained in a scattering plane where all Bragg peaks like $\mathbf{Q} = (H, 0, L)$ (in tetragonal notations for which $a = b = 3.82 \text{ \AA}$ and $c = 13.15 \text{ \AA}$) were accessible. The superconducting transition $T_c = 22 \text{ K}$ has been measured by both magnetic susceptibility on a small piece as well as using neutron depolarization on the whole sample, corresponding to a doping level of 8.5% [19]. All the polarized neutron diffraction measurements were collected on the same 4F1 triple-axis spectrometer at the *Laboratoire Léon Brillouin, Saclay, France* as in previous measurements [3–5] with an incident neutron wave vector of $k_i = 2.57 \text{ \AA}^{-1}$. Although the actual symmetry of LSCO is orthorhombic, we are using here tetragonal notations for an easier comparison with other cuprates.

Respecting TSL, the magnetic moments of the $\mathbf{Q} = 0$ antiferromagnetic order scatter neutrons at the same positions as the Bragg peaks of the crystallographic structure. As a result, measurements have to be performed on the weakest nuclear Bragg peak having the proper symmetry for the magnetic phase in order to evidence small moments. As in other cuprates [3–5], the Bragg point $\mathbf{Q} = (1, 0, 1)$ offers the best compromise. Attempts to observe a long range order magnetic component on $\mathbf{Q} = (1, 0, 1)$ was not successful in LSCO. Therefore, the $\mathbf{Q} = 0$ antiferromagnetic long range order present in other cuprates is either absent in LSCO or too weak to be experimentally detected. One can give an upper estimate of less than $0.02\mu_B$ for a 3D ordered $\mathbf{Q} = 0$ antiferromagnetic moment in LSCO, as compared to the measured value of $\sim 0.1\mu_B$ in YBCO_{6,6} for a doping $\sim 10\%$ [3].

As pseudogap properties in LSCO are less accurate than in other cuprates [1], a more disordered state, characterized by finite correlation length, can be actually expected. In case of short-range magnetic order, magnetic intensity would be redistributed in momentum space, making its detection on top of a nuclear Bragg peak almost impossible even with polarized neutron diffraction. In order to look for a broader magnetic signal in Q space, we then measure off the Bragg position but still localized around the same planar wave vectors, respecting the TSL, say $Q_{2D} = (1, 0)$. Figure 1 shows scans in the spin-flip channel for $\mathbf{H} \parallel \mathbf{Q}$ along either the diagonal $(1, -1)$ direction [Fig. 1(a)] or along a^* [Fig. 1(b)] across the wave vector $\mathbf{Q} = (1, 0, L)$ for $L = 0, 0.45$ and 0.5 . In all these scans, a peak is observed at 30 K and vanishes at high temperature. The scans along the diagonal direction show a signal centered at $Q = (1, 0, L)$ with a full width at half maximum (FWHM) of $\Delta_q = 0.11 \pm 0.02$ reduced lattice unit (r.l.u.) independently of L (the resolution FWHM is typically 0.07 r.l.u.). This leads to a correlation length of $\xi_{(1,1,0)} \equiv 2/\Delta_q \approx 11 \pm 2 \text{ \AA}$ after resolution deconvolution. As shown by Fig. 1(b), the scan along a^* is broader, yielding $\xi_{(1,0,0)} = 8 \pm 3 \text{ \AA}$. It is worth noticing that the signal/background ratio is only about 5%. The Fig. 1(c) displays the temperature of the maximum of the signal at $Q = (1, 0)$ for $L = 0.5$, as well as the temperature dependence of the background measured off the peak at $Q = (0.8, 0.2)$. The background exhibits a slope consistent with a Debye-Waller factor, on top of which the magnetic signal shows up on cooling down. The difference between both curves indicates a transition temperature T_{mag} around 120 K, in agreement with high-temperature scans shown on Fig. 1. Passing through T_c , the magnetic intensity does not show any noticeable change [Fig. 1(c)]. Finally, additional Q scans at several L values have been also performed. Figures 2(a) and 2(b) show difference of scans between 30 and 120 K for $L = 0$ and $L = 0.93$. A magnetic peak is observed at any L indicating a quasi L -independent structure factor, as shown by Fig. 2(e), where we report the difference between the neutron inten-

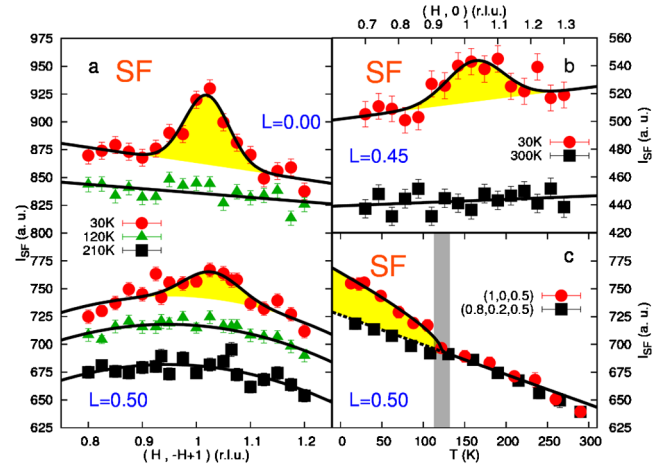


FIG. 1 (color online). Raw data in the spin-flip (SF) channel for a neutron polarization $\mathbf{H} \parallel \mathbf{Q}$. (a) diagonal scans along the $(1, -1)$ direction around $Q = (1, 0, L)$ at different temperatures for $L = 0$ and $L = 0.5$. (b) longitudinal scans along the $(1, 0)$ direction around $Q = (1, 0, 0.45)$. (c) Temperature dependence of neutron intensity (SF) at $Q = (1, 0, 0.5)$ (red circles) and at a background position $Q = (0.8, 0.2, 0.5)$ (black dots). For all figures, yellow areas represent the magnetic signal. The typical counting time is about 1 h per point in order to get sufficient statistics.

sity and the background from the scans measured at different L . The magnetic intensity is present at all measured L values. This implies that the magnetic correlations are basically two-dimensional (2D). The observed magnetic signal is therefore a 2D short-range order, occurring around the same symmetry points as in case of the long range 3D order in YBCO [3,4] and Hg1201 [5], corresponding to orbital-like magnetic order. The magnetic order is found to be static at the energy scale given by the spectrometer energy resolution which is about 1 meV. However, it might be fluctuating at lower energy.

Figure 2(c) shows the difference between 30 and 120 K in the spin-flip channel and $\mathbf{H} \parallel \mathbf{Q}$ for a scan along the a^* axis around $\mathbf{Q} = (0, 0, 3.65)$ which has been chosen such as the modulus of \mathbf{Q} is similar to the wave vector $\mathbf{Q} = (1, 0, 0.5)$. Interestingly, the magnetic signal centered at $(H, K) = (1, 0)$ is absent for $H = K = 0$. This result implies a specific magnetic structure factor which needs more than one magnetic moment per unit cell and a sum of these moments equal to zero within each unit cell, as it is expected for the orbital magnetic order [7].

In a polarized neutron scattering measurements, the scattered intensity associated with a magnetic component simultaneously perpendicular to the momentum \mathbf{Q} and to the polarization direction shows up exclusively in the SF channel. As a result, the magnetic signal has to be purely spin flip for $\mathbf{H} \parallel \mathbf{Q}$. Accordingly, the temperature difference in the non-spin-flip channel for $\mathbf{H} \parallel \mathbf{Q}$ [Fig. 2(d)] shows no intensity peaked at $H = 1$ in contrast to the corresponding SF data [Fig. 1(a)]. Turning the neutron polarization direction, one can further single out the mag-

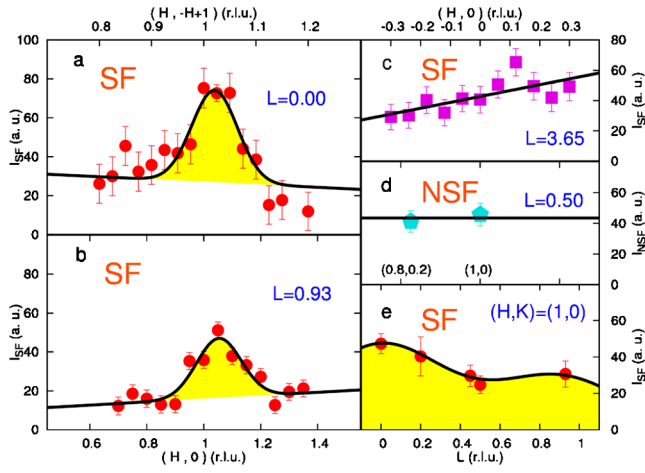


FIG. 2 (color online). Difference of measurements performed at 30 K and 120 K for a neutron polarization $\mathbf{H} \parallel \mathbf{Q}$. (a)–(c) Scans in the spin-flip channel: (a) Around $\mathbf{Q} = (1, 0, 0)$ along a diagonal direction $(1, -1)$, (b) around $\mathbf{Q} = (1, 0, 0.93)$ along a^* . are Gaussian fits, a slight shift of the peak maximum to 1.03 is typically obtained. (c) Around $\mathbf{Q} = (0, 0, 3.65)$ along a^* . (d) Scan in the non-spin-flip channel around $\mathbf{Q} = (1, 0, 0.5)$ along the diagonal direction $(1, -1)$. (e) L dependence of the magnetic intensity (background subtracted) determined from the various Q scans measured across the $\mathbf{Q} = (1, 0, L)$ in the SF channel. The line across the points is a guide to the eye.

netic scattering associated with each magnetic component [3,4]. The intensity measured for each polarization is a sum of a magnetic intensity $I_{H\alpha}$ ($\alpha = \{x, y, z\}$) and a background (BG) signal, which does not depend on the neutron polarization. For only a magnetic scattering, the neutron intensity should obey the following selection rule: $I_{Hx} = I_{Hy} + I_{Hz}$. The Fig. 3(a) shows the polarization analysis of the scan along the diagonal for $L = 0$. The expected relation for a magnetic scattering is observed demonstrating the magnetic nature of this intensity centered at $(H, K) = (1, 0)$. Likewise, the neutron intensity for both polarizations perpendicular to \mathbf{Q} , each sensitive to either M_z or M_y , has the same amplitude. Similar observations were made in the other cuprates [3–5]. This may indicate that the moments are pointing along a direction forming 45° angle between c^* axis and the (a^*, b^*) plane.

In order to provide a more quantitative description of the observed magnetic signal, we report on Fig. 3(b) the temperature dependence of the normalized Q -integrated magnetic structure factor, $S_{\text{mag}} \equiv \int d^3Q I_{\text{mag}}(Q) / \int d^3Q$. First, the magnetic intensity at $Q = (1, 0, 0.5)$ is obtained from the temperature dependence of the magnetic signal after subtraction of the background [both shown on Fig. 1(c)]. The absolute value of S_{mag} is then calculated from these data after integration in Q space of the magnetic signal and after normalization of the magnetic intensity to the nuclear Bragg intensity. As represented in Fig. 3(b), the deduced Q -integrated magnetic intensity, S_{mag} , reaches a value of 1.2 mbarns at the lowest temperature. A magnetic local moment, M_{loc} , can be obtained from S_{mag} . The absolute

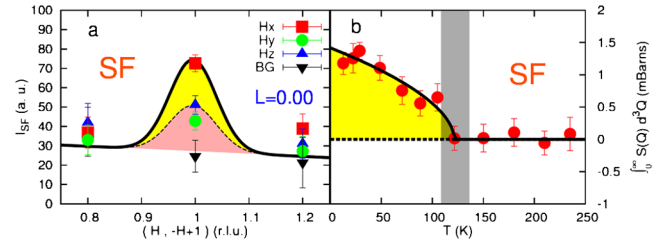


FIG. 3 (color online). (a) Polarization analysis of the difference of scans measured at 30 and 120 K in the spin-flip channel around $\mathbf{Q} = (1, 0, 0)$ along a diagonal direction $(1, -1)$. The neutron polarization is applied successively along three different directions \mathbf{H}_α . The label α correspond to the Cartesian axis $\{x, y, z\}$, so that the x axis is parallel to \mathbf{Q} , while the z axis stands for the direction perpendicular to the scattering plane. (b) Temperature dependence of Q -integrated magnetic intensity, S_{mag} .

value of the magnetic local moment is found around $0.1 \mu_B$ at low temperature. Remarkably, similar amplitude for both the neutron structure factor and the magnetic moment has been reported for the $\mathbf{Q} = 0$ AFO in both YBCO [3,4] and Hg1201 [5].

Here we have demonstrated that the orbital-like order is present in the archetypal LSCO system and is likely to be a generic property of superconducting cuprates. In YBCO and Hg1201, the 3D long range order appears at a temperature T_{mag} matching the pseudogap temperature T^* . In our LSCO sample, the $\mathbf{Q} = 0$ AF state settles in at $T_{\text{mag}} \sim 120$ K. As the doping is quite low (8.5%), a much larger T^* is typically expected from the generic phase diagram of high- T_c cuprates. However, it is worth pointing out that the features associated with the pseudogap temperature are less defined in LSCO than in the other cuprates [1], making it difficult to define T^* in an unequivocal way. Likewise, several anomalies have been reported close to T_{mag} in the specific heat [19,20], the uniform spin susceptibility [19] and the Nernst effect [21] for LSCO samples in the same doping range: their interpretations should be reconsidered in light of our data.

In addition to the reduced value of T_{mag} , the $\mathbf{Q} = 0$ AFO remains 2D and short range in LSCO. This frustration of the $\mathbf{Q} = 0$ AF correlation could result from a competition with another electronic instability, namely, the tendency toward stripes phase known, among cuprates, to occur specifically in the LSCO system. While the observed ordering temperature $T_{\text{mag}} \sim 120$ K is larger than the static stripeslike ordering temperature ever reported in LSCO, *a priori*, there has to be a direct competition between the $\mathbf{Q} = 0$ AFO and the fluctuating stripe phase as these phases are breaking different symmetries. To look for such a connection, we have studied the temperature dependence of the IC spin fluctuations [22,23], usually associated with dynamical stripes at low energy. We observe strong dynamic IC fluctuations but no static IC magnetic peaks have been evidenced down to 1.5 K. Typical scans

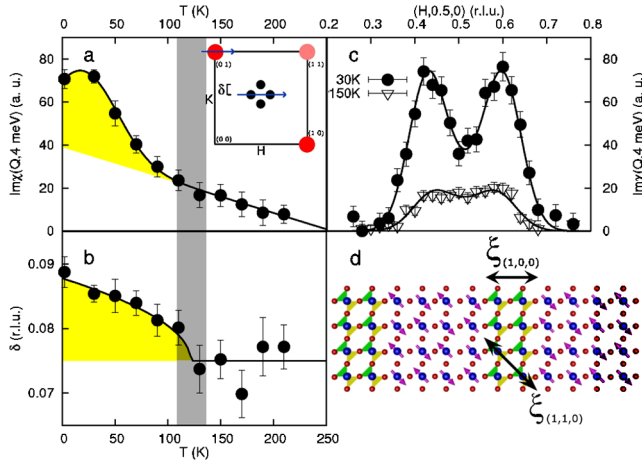


FIG. 4 (color online). Unpolarized inelastic neutron scattering measurements of the IC magnetic fluctuations around $Q_{IC} = Q_{AF} \pm (\delta, 0)$: (a) Temperature dependence of the spin susceptibility at an energy of 4 meV: $\text{Im}\chi(Q_{IC}, \hbar\omega = 4 \text{ meV})$. The inset represents the location of the different magnetic response in the a-b plane: $Q = 0$ AFO and IC spin fluctuations are shown in red and black, respectively. (b) Temperature dependence of the IC parameter δ . In (a) and (b), the vertical dashed line indicates $\sim T_{\text{mag}}$. (c) Typical H -scans across IC spin excitations at $\hbar\omega = 4 \text{ meV}$. The figure shows the imaginary part of the dynamical magnetic susceptibility $\text{Im}\chi(Q, \hbar\omega)$ at 30 K (full circles) and at 150 K (open triangles). (d) Schematic picture of the CuO_2 plane for a hole doping of 1/12 based on the bond centered stripes model discussed in ref. [12].

across the incommensurate peaks at $\hbar\omega = 4 \text{ meV}$ are shown in Fig. 4(c), indicating IC spin excitation at the wave vectors Q_{IC} with $\delta = 0.085 \pm 0.005$ at $T = 30 \text{ K}$ [see Fig. 4(b)]. We discover that the incommensurability parameter δ exhibits a clear enhancement at T_{mag} [Fig. 4(b)] which is accompanied by an increase of the peak magnitude [Fig. 4(a)]. Both behaviors have been actually already reported in stripe ordered Ba doped cuprate [10] around 65 K. Here, we are able to relate these variations with the onset of the $Q = 0$ AFO at T_{mag} , highlighting the relation between both electronic instabilities.

Recently, it has been shown that orbital currents could also develop in doped two-leg spin ladders [24]. Inspired by this work, a simple picture can emerge based on the bond centered stripes model [12]. Hole-poor regions exhibiting fluctuating copper spins [in purple in Fig. 4(d)] are separated by hole-rich regions, depicted as hole doped 2-leg ladders. The $Q = 0$ AFO might develop within the charge stripes: the circulating current phase Θ_{II} [7] is represented for a sake of example. The observed correlation lengths of the $Q = 0$ AFO are shown along the (100) and (110) directions. In order to preserve the lattice translation invariance, magnetically order charge stripes should remain magnetically decoupled from each other. Excluded from the hole-poor AF domains where the spin fluctuations are growing at low temperature, the $Q = 0$ AFO correla-

tion lengths are found limited by the size of the charge stripes [see Fig. 4(d)]. Still within this picture, the absence of 3D $Q = 0$ AFO in LSCO could be explained as stripes alternate directions as one goes from one CuO_2 plane to the next.

Finally, at the same time as dynamical stripes are reported through incommensurate magnetic fluctuations, LSCO exhibits a short range $Q = 0$ AFO (or orbital-like magnetic order) which could be confined within the charge stripes. Having the same symmetry as the long range magnetic order reported in two others cuprates [3–5], the nature of this magnetism, involving both copper and nearest neighbor oxygens, is likely a keystone towards understanding the physics of HTS.

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