Supplementary Figure S1. (a) Layout of the multidetector diffractometer D7 [S1]. (b) Definition of the X' and Y' polarization directions within the scattering plane. $\gamma = 41.6^\circ$ is the angle between the incident wave vector $k_i$ and X', set by the instrument configuration, $2\theta$ is the scattering angle, and $\alpha$ is defined as the angle between the wavevector $Q$ and X'. Reproduced from [S2].

Supplementary Figure S2. L-scan measured at 10 K in the $SF_X$ channel. (a) around (3,0,1) for $SCCO - 8$. (b) around (0,0,1) for $SCCO - 5$. The scattered intensity is described by a Gaussian signal on top of the sloping background (shaded area). All measurements were performed on 4F1.
SUPPLEMENTARY FIGURE 3: TEMPERATURE DEPENDENCE

Supplementary Figure S3. (a) SCCO − 8 : T-dependence of the magnetic signal centered at (3,0,1) as measured in the SFX channel (red symbols), compared with the T-dependence of a background taken at positions shifted by ΔL = ±0.2 away from (3,0,1) (see text). (b-c) SCCO − 5 : (b) Unpolarized neutron measurements of the T-dependence of the scattered intensity at (1,0,1) (blue circles). The departure from a linear T-dependence is shown by the dashed line. The shaded area allows the identification of the background to which an extra magnetic scattering adds at low temperature. (c) T-dependence of the magnetic signal at (3,0,1) as measured in the SFX channel (violet squares) compared to the T-dependence at the background position (3 0 0.8) (gray circles). All measurements were performed on 4F1.

SUPPLEMENTARY FIGURE 4: ABSENCE OF THE MAGNETIC INTENSITY WITHIN THE CHAIN SUBSYSTEM

Supplementary Figure S4. Full magnetic intensity deduced from XYZ-PA (4F1, T=10K) within the chain subsystem along (H,0,0,1) in superspace r.l.u or (H,0,1.43) in ladders r.l.u for SCCO − 5.
SUPPLEMENTARY FIGURE 5: IN-PLANE AND OUT-OF-PLANE MAGNETIC INTENSITY

Supplementary Figure S5. SCCO − 8: Mapping of the magnetic intensities within ladders at T=5 K, as deduced from XYZ-PA carried out using SF data measured on D7: (a) the in-plane intensity $I_{ac}$, (b) the out-of-plane intensity $I_b$. The maps are given in r.l.u of the ladder subsystem and the intensities calibrated in mbarn. The magnetic spots are located by crosses along the ladder scattering ridge along (H,0,1). The blue arrows show the satellite magnetic reflections at (H,0,1,-1) using superspace notations or L∼0.43 in ladders r.l.u. The blue crosses indicate the positions of Nuclear Bragg scattering from the ladders. Red spots around these two regions do not correspond to magnetic scattering but are due to polarization leakage.

SUPPLEMENTARY FIGURE 6: ABSENCE OF CDW

Supplementary Figure S6. SCCO − 5: series of scans performed on 4F1 in the NSF channel at 8 K (red symbols) and 150 K (blue symbols). (a) scans along (3,1,L) crossing both L=1.2 and L=1.14 positions in r.l.u of the ladders, corresponding to $q_{CDW}$ reported for ladders and chains, respectively. (c) K-scans across (3,K,1.14) in r.l.u of the ladders, corresponding to $q_{CDW}$ reported for chains. The increase of the intensity in both large L or K is due scattering of Al from the sample holder. (b,d) Differential intensities 8 K-150 K, from scans reported in (a,c) that show the absence of any CDW-induced structural distortion.
Supplementary Figure S7. (a) Ladder unit cell in SCCO containing Copper sites (full dots) and Oxygen sites (open dots). (b-f) The CuO$_2$ plaquette can be decorated with various magnetic patterns: (b) a spin on Cu site. (c) a spin (or orbital) nematic state, with two sets of staggered spin on O sites as proposed in [S3]. (d) loop current (LC) state $CC - \theta_I$ [S4, S5]. (e) LC state $CC - \theta_{II}$ [S6] showing two possible patterns breaking rotational symmetry along the diagonals: upper part denoted $\epsilon = +1$ and lower part denoted $\epsilon = -1$. (f) LC state $CC - \theta_{III}$ [S7]: lower part (Horizontal) and upper part (vertical) pattern, respectively. For all LC states, the staggered orbital moments are expected to point perpendicular to the circulating currents (ladder plane).

Supplementary Figure S8. (a) Model of antiferromagnetically interacting Cu spins in the first ladder. The question mark and colored arrows show the frustrated Cu spin on the second ladder, suggesting a high degree of frustration between neighboring ladders. (b) L-scan across (1.5,0,L) as measured on 4F1 at 12 K in the SF$_X$ channel.
SUPPLEMENTARY FIGURE 9: MAGNETIC MOMENTS ON THE OXYGEN SITES

Supplementary Figure S9. (a) Magnetic nematic order, with staggered magnetic moments on O sites in each CuO$_2$ plaquette. (b) H-dependence of $|F(Q)|^2$ for a model of magnetic moments on oxygen sites for L=1.

SUPPLEMENTARY FIGURE 10: Q-DEPENDENCE OF LC PATTERNS

Supplementary Figure S10. Q-dependence along (H,0,1): (a) the squared magnetic structure factor $|A_\phi(Q)|^2$ associated with each of the LC patterns: $CC - \theta_I$, $CC - \theta_{II}$ (+, − and average (see text) denoted $+/−$) and $CC - \theta_{III}$ (Horizontal and Vertical). (b) The structure factor $|B_{\phi\phi}(Q)|^2$ coming from the intra-rung correlations: $\phi = 0$ for identical patterns and $\phi = \pi$ for opposite patterns. (c) The structure factor $|C_{\phi\phi}(Q)|^2$ coming from the inter-ladder correlations: $\psi = 0$ for identical patterns and $\psi = \pi$ for opposite patterns. (The intra-rung term, $|B_{\phi}(Q)|^2$ is taken for $\phi = 0$).
**SUPPLEMENTARY FIGURE 11: CC − ΘI LC PATTERS**

Supplementary Figure S11. (a) $CC − \Theta_I$ state [S6]: 4 LCs per $CuO_2$ flowing clockwise (blue triangles) and anticlockwise (red triangles). (b) $H$-dependence of the structure factor for a model of $CC − \Theta_I$ LCs for $L=1$.

**SUPPLEMENTARY FIGURE 12: CC − ΘII LC PATTERS**

Supplementary Figure S12. $CC − \Theta_{II}$ state [S6]: 2 LCs per $CuO_2$ flowing clockwise (blue triangles) and anticlockwise (red triangles) and aligned along a given diagonal. (a) Uncorrelated model: only 2 two Cu sites in the first ladder are decorated with the same $CC − \Theta_{II}$ pattern, whereas the second ladder remains non magnetic. (b) Correlated model: 2 two Cu sites in the first ladder and the two other in the second one are decorated with the same $CC − \Theta_{II}$ pattern (case: in-phase, $\varphi = 0$). (c) Correlated model: 2 two Cu sites in the first ladder exhibit the same LC patterns, while the two other in the second one are characterized by a LC pattern rotated at $\pi/2$ (case: crisscrossed, $\psi = \frac{\pi}{2}$, $\delta = -1$). (d) $|F(Q)|^2$ for the uncorrelated and in-phase case (magenta) and the correlated one, considering 4 distinct cases: in-phase, $\psi = 0$ (black), crisscrossed, $\psi = \pm \frac{\pi}{2}$, $\delta = +1$ green), out-of-phase, $\psi = \pi$ (red), crisscrossed, $\psi = \pm \frac{\pi}{2}$, $\delta = -1$ (blue).
SUPPLEMENTARY FIGURE 13: \( CC - \Theta_{III} \) LC PATTERNS

Supplementary Figure S13. \( CC - \Theta_{III} \) state [S7, S8]: 2 LCs per \( Cu_2O_2 \) flowing clockwise (blue triangles) and anticlockwise (red triangles) and aligned along either the ladder rungs (Horizontal, (a-c)) or the ladder legs (Vertical, (d-f)). (a,d) Uncorrelated model (in-phase intra-rung coupling: \( \varphi = 0 \)): Only 2 two Cu sites in the first ladder are decorated with the same \( CC - \Theta_{III} \) pattern; whereas the second ladder remains non magnetic. (b,e) Correlated model (in-phase inter-ladder coupling: \( \psi = 0 \)): 2 two Cu sites in the first ladder and the two other in the second one are decorated with the same \( CC - \Theta_{III} \) pattern. (c,f) Squared magnetic structure factors: uncorrelated case (red solid line) and correlated case (black dotted line).

SUPPLEMENTARY FIGURE 14: COMPARISON OF MEASURED INTENSITIES AND LC MODELS

Supplementary Figure S14. (a) Q-dependencies of the squared magnetic form factor for oxygen \( f_0^2(Q) \) and copper \( f_{Cu}^2(Q) \). The shaded area indicates the Q-range where the PND study was carried out. (b,c) Q-dependencies of \( (f(Q)^2 |F(Q)|^2) \) along (H,0,1): (b) uncorrelated case with in-phase intra-rung correlation only, (c) correlated case, in-phase inter-ladder correlation. (d,e) Q-dependencies of of \( I_b \) (mbarn) along (H,0,1): (d) uncorrelated case with in-phase intra-rung correlation only, (e) correlated case, in-phase inter-ladder correlation. The fits correspond to different LC patterns: \( CC - \Theta_{III} \) (black), \( CC - \Theta_{III} \) Vertical (magenta) and Horizontal (orange). The solid and dotted lines correspond to fits including the Cu- and O-form factors, respectively.
Supplementary Figure S15. (a) Q-dependencies of the orientation factors $[1 - Q_i^2]$, with $i = \{a, b, c\}$. (b,c) Q-dependencies of $I_{mag}$ along (H,0,1): (b) SCCO-5 data, with fits for the uncorrelated case with in-phase intra-rung correlations only. (c) SCCO-8 data with fits for the the correlated case with in-phase intra-ladder correlations. The fits correspond to different LC patterns: $CC - \theta_{II}$ (black), $CC - \theta_{III}$ Vertical (magenta) and Horizontal (orange). The solid and dotted lines are associated with fits including the Cu- and O-form factors, receptively.
SUPPLEMENTARY NOTE 1: POLARIZED NEUTRON DIFFRACTION (PND) AND XYZ-POLARIZATION ANALYSIS (XYZ-PA)

Neutron diffraction

The PND experiments were carried out on two instruments: the 4F1 triple axis spectrometer (Orphée reactor, Laboratoire Léon Brillouin, Saclay) and the D7 multidetector diffractometer (Institut Laue Langevin, Grenoble). On each instrument, measurements were carried out with a neutron wavevector of 2.57 Å\(^{-1}\) and 2.02 Å\(^{-1}\), respectively.

\(\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_{2y}\text{O}_{41}\) (SCCO-x) exhibits an aperiodic atomic structure with two, ladders (ld) and chains (ch), incommensurate sub-lattices. Within these units, the Bragg peaks need to be indexed in the 4D superspace as \((H,K,L_{ld},L_{ch})\). Both sublattices are incommensurate along the \(c\)-axis with an incommensurability parameter \(\frac{1}{\pi} = \frac{c_{ch}}{c_{ld}} = 1.42\). In the experiments, we used the ladder sub-lattice parameters as a reference: \(H, K\) and \(L\) coordinates are expressed in reduced lattice units (r.l.u) of the ladder (ld) subsystem: \(\frac{2\pi}{a} = 0.55\ \text{Å}^{-1}, \frac{2\pi}{b} = 0.48\ \text{Å}^{-1}\) and \(\frac{2\pi}{c} = 1.61\ \text{Å}^{-1}\), respectively.

The samples were always aligned in the \((H,0,0)/(0,0,L)\) scattering plane, in order to probe the \([a,c]\) plane of the ladders. Within that scattering plane, a transferred momentum \(Q\) of the form \((H,0,L)\) is accessible. Within these units, the Bragg peaks associated with the chains subsystem contribute at \(L_{chains} = 1.42L\) as \(\frac{2\pi}{c_{ch}} = 2.28\ \text{Å}^{-1}\). Additionally, tilting the sample out of the scattering plane using goniometers allowed us to access wavevectors of the form \(Q(H,K,L)\) on 4F1.

Considering a magnetic sample, the magnetic scattering cross-section [S9], \(I_{mag}\), reads:

\[
I_{mag} = \Phi_S N_{cell} r_0^2 f(Q)^2 |F(Q)|^2 m_L^2
\]  

\(\Phi_S\) corresponds to the neutron flux at the sample in \(n/s/\text{barns}\). \(N_{cell}\) is the number of unit cells in the sample. \(r_0\) stands for the neutron magnetic scattering length, \(r_0^2 = 290\ \text{mbarns}\). \(f(Q)\) is to the magnetic form factor and \(|F(Q)|\) the magnetic structure factor. Owing to the dipolar nature of the interaction between neutron spin and the magnetic moments, \(m\), \(I_{mag}\) probes \(m_L\) the magnetic components perpendicular to \(Q\), only.

The squared modulus of \(m_L\) can be expressed using the regular Cartesian coordinates of the lattice:

\[
m_L^2 = m^2 - (m.Q)^2 = \sum_{i,j=a,b,c} (\delta_{ij} - \frac{Q_iQ_j}{Q^2}) m_i m_j
\]  

For a magnetic moment, \(m(\pm m_a,\pm m_b,\pm m_c)\) with \(n\) non zero components, there are \(2^n\) equivalent magnetic domains. The cross-terms \((i \neq j)\) cancel out when summing over all domains, at variance with the diagonal terms \((i = j)\). In the case of the present study with \(Q = (H,0,L)\), \(I_{mag}\) reduces to the sum of in- and out-of plane terms, so that:

\[
I_{mag} = I_{ac} + I_b
\]  

With \(I_{ac} \propto m_{ac}^2 = [(1 - |F_a/Q|^2)m_a^2 + (1 - |F_c/Q|^2)m_c^2]\) and \(I_b \propto m_b^2\) (because \(K = 0\)).

It is also convenient to use the so-called \{XYZ\} referential, where \(X\) is the unitary vector parallel to \(Q\). \(Y\) and \(Z\) are the two unitary vectors orthogonal to \(X\), within the scattering plane and perpendicular to the scattering plane, respectively. So that \(m_L = m_X Y + m_Z Z\), with the in- and out-of (scattering) plane components.

Polarized neutron diffraction setup

On the incoming neutron beam, a bender (polarizing super-mirror) can polarize the neutron spin and a Mezei flipper can adiabatically flip the neutron spin. A pyrolitic graphite filter is further added before the bender to remove high harmonics on 4F1. The neutron spin polarization is maintained using a homogeneous guide field of a few Gauss. The neutron spin polarization direction, \(P\), is controlled on the sample by Helmholtz coils on 4F1 and a quadrupolar assembly on D7 [S1]. On the scattered neutron beam, the final neutron spin polarization is analyzed using either an analyzer made of co-aligned Heusler single crystals (on 4F1) or polarizing benders (on D7) placed in front of the multidetector bank (Supplementary Figure S1.a)

Polarized neutron diffraction cross sections

Polarization analysis of PND allows us to distinguish between the different contributions to the scattered intensity [S9]. For a nuclear scattering, the neutron spin remains unchanged and the scattered intensity is measured in the Non Spin-Flip (NSF) channel. Since the spin polarization of the neutron beam is not perfect, a small amount of the nuclear scattering can nevertheless leak into the Spin-Flip (SF) channel. The ratio between scattered intensities in the NSF and the SF channels is called the flipping ratio FR and characterizes the quality of spin polarization of the neutron beam. For a magnetic scattering, the scattered intensities in each channel strongly depends on \( \mathbf{P} \). Indeed, neutron spins are described using Pauli matrices, whose quantization axis is given by \( \mathbf{P} \). The magnetic intensity \( I_{mag}^{SF}(\mathbf{P}) \propto (\mathbf{m}_\perp \cdot \mathbf{P})^2 \) does not flip the neutron spin and remains in the NSF channel. The remaining magnetic \( I_{mag}^{SF}(\mathbf{P}) \propto (\mathbf{m}_\perp)^2 - (\mathbf{m}_\perp \cdot \mathbf{P})^2 \) appears in the SF channel.

On D7, the direction of polarization \( \mathbf{X} \) is not parallel to \( \mathbf{Q} \) but along a direction \( \mathbf{X}' \) turned by an angle \( \alpha \) (Supplementary Figure S1.b). Therefore, one needs to estimate the scattered intensities in directions of \( \mathbf{P} \) corresponding to unitary vectors \( (\mathbf{X}', \mathbf{Y}', \mathbf{Z}') \), as shown in Supplementary Figure S1.b, given by:

\[
\begin{pmatrix}
X' \\
Y' \\
Z'
\end{pmatrix} = \begin{pmatrix}
cos \alpha & \sin \alpha & 0 \\
-\sin \alpha & \cos \alpha & 0 \\
0 & 0 & 1
\end{pmatrix}
\begin{pmatrix}
X \\
Y \\
Z
\end{pmatrix}
\]

(4)

Then, the full magnetic scattering, \( I_{mag} \), given in Eq. 3, splits in two terms, \( I_{mag}^{SF}(\mathbf{P}) \) and \( I_{mag}^{NSF}(\mathbf{P}) \)

\[
\begin{align*}
I_{mag}^{SF}(\mathbf{X}') &= I_{ac} \cos^2 \alpha + I_b \\
I_{mag}^{NSF}(\mathbf{X}') &= I_{ac} \sin^2 \alpha \\
I_{mag}^{SF}(\mathbf{Y}') &= I_{ac} \sin^2 \alpha + I_b \\
I_{mag}^{NSF}(\mathbf{Y}') &= I_{ac} \cos^2 \alpha \\
I_{mag}^{SF}(\mathbf{Z}') &= I_{ac} \\
I_{mag}^{NSF}(\mathbf{Z}') &= I_b
\end{align*}
\]

(5)

In addition to the polarization magnetic cross-sections of Eq. 5, \( I_{mag}(\mathbf{P}) \), one should consider the nuclear intensity, \( I_{nucl} \) and a background, \( B_g \), in both SF and NSF channels. Both SF and NSF cross-sections read:

\[
\begin{align*}
I_{nucl}^{NSF}(\mathbf{P}) &\sim B_g^{NSF} + I_{nucl}^{NSF}(\mathbf{P}) + I_{mag}^{NSF}(\mathbf{P}) \\
I_{nucl}^{SF}(\mathbf{P}) &\sim B_g^{SF} + I_{mag}^{SF}(\mathbf{P})
\end{align*}
\]

(6)

As discussed in [S10], due to imperfect polarizations, the measured neutron intensities are mixing the cross-sections of Eq. 6. In each channel, it can be actually written,

\[
\begin{align*}
I_{meas}^{NSF}(\mathbf{P}) &\propto B_g^{NSF} + I_{nucl}^{NSF}(\mathbf{P}) + I_{mag}^{NSF}(\mathbf{P}) + \frac{1}{FR(\mathbf{P})} [B_g^{SF} + I_{mag}^{SF}(\mathbf{P})] \\
I_{meas}^{SF}(\mathbf{P}) &\propto B_g^{SF} + I_{mag}^{SF}(\mathbf{P}) + \frac{1}{FR(\mathbf{P})} [B_g^{NSF} + I_{nucl}^{NSF}(\mathbf{P}) + I_{mag}^{NSF}(\mathbf{P})]
\end{align*}
\]

(7)

where \( FR(\mathbf{P}) \) is the polarization dependent flipping ratio of the experiment. For both instruments, 4F1 and D7, \( FR(\mathbf{P}) \) were measured for all relevant scattering angle using a quartz sample. One can deduce the PND cross-sections, Eq. 6, from the measured ones, Eq. 7 [S10]. In the present study, although the nuclear and magnetic scattering preserve the lattice translation symmetry, the short range magnetism (SRM) occurs at \( \mathbf{Q} \) values where there is additional extinction of the nuclear peaks due to the 3D atomic structure. Therefore, the effects of imperfect polarizations of the nuclear term and background terms of Eq. 7 are relatively weak (although sizeable) and readily corrected. It should be emphasized that this is a very different situation from the case of most superconducting cuprates [S2, S3] where the \( \mathbf{q}=0 \) magnetism occurs at the same Bragg position of the nuclear structure.
XYZ-Polarization analysis

We systematically performed a longitudinal XYZ polarization analysis (XYZ-PA), that allows the determination of the full magnetic intensity $I_{mag} = I_{ac} + I_b$ (Eq. 3) from a set of measurements in the SF channel with $P$ along each of the 3 unitary vectors $(X', Y', Z')$. The combination of all those measurements provides access to the in- ($/// ac$) and out-of-($/// b$) scattering plane magnetic scattering and the SF background. Two different situations occur for both instruments:

- **On $D7$, Diffractometer:** $\alpha = 90 - \theta + 41.6^\circ$ (Supplementary Figure S1), with $\theta$ the Bragg angle that depends on $Q$ and the neutron wave length. From $X'$, $Y'$ and $Z'$ measurements, the full measurable magnetic intensity is deduced from Eq. 5 as follows:

  \[
  \begin{aligned}
  I_{ac} &= \frac{[I^{SF}(X') - I^{SF}(Y')]}{[\cos^2 \alpha - \sin^2 \alpha]} \\
  I_b &= \frac{[I^{SF}(X') - I^{SF}(Z')]}{\sin^2 \alpha I_{ac}} \\
  I_{mag} &= \frac{[I^{SF}(X') - I^{SF}(Z')] + g(\alpha)[I^{SF}(X') - I^{SF}(Y')]}{2 \cos^2 \alpha - \sin^2 \alpha} \tag{8}
  \end{aligned}
  \]

  For $I_{mag}$, the second term is weighted by $g(\alpha) = [1 + \sin^2 \alpha]/[\cos^2 \alpha - \sin^2 \alpha]$, which goes to unity for $\alpha=0$.

- **On $4F1$, Triple axis spectrometer:** $\alpha = 0$, one obtains the usual relations:

  \[
  \begin{aligned}
  I_{ac} &= I^{SF}(X) - I^{SF}(Y) \\
  I_b &= I^{SF}(X) - I^{SF}(Z) \\
  I_{mag} &= 2 I^{SF}(X) - I^{SF}(Y) - I^{SF}(Z) \tag{9}
  \end{aligned}
  \]

**SUPPLEMENTARY NOTE 2: L-DEPENDENCE OF THE SHORT RANGE MAGNETISM (SRM)**

Supplementary Figure S2.a shows the SF intensity measured along $(3,0,L)$ for $P//X$ at 10 K on sample $SCCO - 8$. On the L-scan, the magnetic signal at $L=3$, exhibits a Gaussian line-shape and appears on top of a sloping background. The determination of such a sloping background has been confirmed by XYZ-PA. A qualitatively similar type of signal is observed for sample $SCCO - 5$ (Supplementary Figure S2.b).

**SUPPLEMENTARY NOTE 3: TEMPERATURE DEPENDENCE OF THE SHORT RANGE MAGNETISM**

- **$SCCO - 8$:** the T-dependence of the scattered intensity at $(3,0,1)$ was measured in the $SF_X$ channel on $4F1$ (Supplementary Figure S3.a). It displays a net enhancement at low temperature. According to the L-scan across $(3,0,1)$ performed at low temperature (Fig. 2.b in the main text), the SRM signal centered at $L=1$ vanishes at $L=0.8$ and 1.2. Two additional T-dependencies were measured at those L values and averaged to provide the T-dependence of the non magnetic background. The comparison of both T-dependencies indicates that the magnetic signal starts developing below an onset temperature $T_{mag} \approx 80$ K.

- **$SCCO - 5$:** The raw temperature dependencies of the magnetic scattering at $(1,0,1)$ and $(3,0,1)$, were measured on $4F1$ (Supplementary Figure S3.b-c). Using an unpolarized neutron beam, the scattered intensity at $(1,0,1)$ exhibits a linear increase on cooling down to a temperature where an extra enhancement of the intensity becomes visible. Using a polarized neutron beam, the signal at $(3,0,1)$ was measured in $SF_X$ and compared to the scattered intensity at $(3,0,0.8)$, a background position according to measurements in $SCCO - 8$. The comparison between polarized and unpolarized neutron data highlights an onset temperature $T_{mag} \approx 50$ K below which the SRM sets in. XYZ-PA has been also employed at $\sim 100$K at $(0,0,1)$, showing the vanishing of the SRM at high temperature.
SUPPLEMENTARY NOTE 4: ABSENCE OF MAGNETIC SIGNAL WITHIN THE CHAINS

A new magnetic signal is clearly observed at \((H, 0, L)\) with integer values of \(H\) and \(L\), corresponding to the ladder subsystem. However, no magnetic signal is observed for the chain subsystem at Bragg positions \((H, 0, 0, 1)\) using superspace notations, corresponding to \((H, 0, 1, 43)\) in the ladder subsystem units. A scan performed across \((H, 0, 1, 43)\), shown in Supplementary Figure S4, shows the full scattered magnetic intensity as deduced from XYZ-PA (4F1) in SCCO \(-5\) (which complements a similar scan in SCCO \(-8\) in the Fig. 3.a of the main text). In both samples, the XYZ-PA reveals the absence of scattered magnetic intensity at positions corresponding to the chains subsystem.

SUPPLEMENTARY NOTE 5: CALIBRATION IN ABSOLUTE UNITS

We converted the measured intensities in absolute units using a vanadium sample, which allows the determination of the neutron flux at the sample position \(\Phi_S\). A vanadium sample is a pure incoherent scatterer. For PND measurements, \(2/3\) of its intensity shows up in the SF channel \((1/3\) in the NSF channel) and its energy \((\omega)\) integrated intensity reads:

\[
I_{Vana}^{SF} = \Phi_S N_{cell} \frac{2}{3} (d\sigma/d\Omega)^{inc}
\]

Where \((d\sigma/d\Omega)^{inc} = 0.394\) barns stands for the vanadium incoherent cross section. For a vanadium sample mass, \(m_V = 1\) g, and a molar mass, \(M_V = 50.94\) g.mol\(^{-1}\), one obtains \(N_{cell} = 0.6023 \frac{m_{max}}{M_V} = 0.0118\) cells/mol.

The incoherent scattering for vanadium is purely elastic and is described by a Dirac distribution in energy, \(\delta(\omega)\). The measured \(\omega\)-dependence is obtained after convolution by the Gaussian instrumental energy resolution, characterized by a full width at half maximum (FWHM) \(\Delta_\omega\): the measured intensity \(I_{meas}(\omega)\) acquires a Gaussian profile as,

\[
I_{meas}(\omega) = I_{max} e^{-4\ln(2) \frac{\omega^2}{\Delta_\omega^2}}
\]

Integrating over energy, one obtains:

\[
I_{Vana}^{SF} = I_{max} \Delta_\omega \frac{\pi}{\ln(2)}
\]

Taking into account the instrument energy resolution: \(\Delta_\omega = 1.25\) meV for \(k_i = k_f = 2.57\) Å\(^{-1}\) gives \(\Phi_S = 1678\) n/s/barns. This value of \(\Phi_S\) holds for both experiments on SCCO \(-5\) and SCCO \(-8\) on the spectrometer 4F1. A similar procedure using a vanadium standard sample was used for data calibration on D7 [S10].

SUPPLEMENTARY NOTE 6: IN-PLANE AND OUT-OF-PLANE MAGNETIC SCATTERING AMPLITUDES

The in-plane \(I_{ac}\) and out-of-plane \(I_b\) magnetic intensities for SCCO \(-8\), as extracted from XYZ-PA on D7 data using Eq. 8, are shown in Supplementary Figure S5. From these maps, one sees that both magnetic components, in-plane and out-of-plane of the ladder a-c plane, are sizeable. For results obtained on both instruments, Supplementary Table S1 gives a summary of the measured magnetic intensities (as extracted from XYZ-PA) at different reciprocal space positions in SCCO \(-5\) and SCCO \(-8\) and the corresponding \(I_{ac}\) and \(I_b\) intensities in absolute units. The data were also systematically corrected by the quartz flipping ratios following the procedure given above for both 4F1 & D7 (we remind that the procedure is also described in [S10] for D7). Note that, on 4F1, the measured \(Q\)-dependencies were systematically measured for negative and positive H values, and symmetrized by averaging the values of the magnetic intensity. The results in Supplementary Table S1 show that both in-plane \(I_{ac}\) and out-of-plane \(I_b\) magnetic components are not zero, leading systematically to a magnetic moment which is not pointing along a high symmetry direction, but typically is making a tilt with the direction perpendicular to the \(CuO_2\) planes as it is observed in all superconducting cuprates [S2, S3].
SUPPLEMENTARY NOTE 7: ABSENCE OF CHARGE DENSITY WAVE-LIKE INSTABILITY

A charge density wave has been reported in the two-leg ladders system, SCCO-x, from conductivity measurements that established a continuous phase diagram of the doped ladders [S11]. However, a contradictory result has been reported as resonant X-ray diffraction concluded that CDW order dominates at two particular x=0 and x=11 [S12], and is supposed to melt for other x, including the present x = 5 and x=8.

In the pure compound SCCO, both ladders (ld) and chains (ch) display charge density waves (CDW), as reported by RXD and Neutron Diffraction studies [S13, S14]. For CDW_{ld} and CDW_{ch}, the incommensurate propagation wavevector q_{CDW} is given in super-space notations by (H, K, L, L′) with:

- L = L_{integer} ± 0.2 r.l.u within the ladders
- L′ = L′_{integer} ± 0.2 r.l.u, within the chains

Both CDW are characterized by a similar onset [S11]. For SCCO — 5 and SCCO — 8 compositions, optical conductivity measurement [S15, S16] report an onset of charge ordering within the ladders T_{CDW} ∼90K for SCCO — 5 and 10K for SCCO — 8.

In a neutron diffraction experiment, the charge order is detected owing the lattice distortion it induces. To detect the hallmark of a CDW, one has to look in the NSF channel in a PND study. During our PND experiments, we actually did not detect the hallmark of any CDW instability.

Search for charge density wave in the ladders

According to literature [S11], T_{CDW} should be at ∼90 K for the SCCO — 5 sample. We performed two L-scans below and above that temperature. Supplementary Figure S6.a shows two scans along (3, 1, L, 0), measured on 4F1 in the NSF channel at T=8K and 150K. The difference between the two sets of measurements (Supplementary Figure S6.b) exhibits a featureless flat L-dependence only, pointing out the absence of any extra signal at L=1.2. Thus, the signal associated with a CDW_{ld} (if any) falls below the threshold of detection of our measurement.

Search for charge density wave in the chains

Supplementary Figure S6.c shows the same measurement within the chain subsystem, where one expects CDW scattering at L′=0.8, corresponding to L=1.14 using the ladder lattice parameter. Additionally, we collect a K-scan across (3, 1, 0, 0.8), corresponding to (3, 1, 1.14) in ladder notations. The differential intensity between 8K and 150K (Supplementary Figure S6.d) does not reveal any signature of CDW_{ch}.

The absence of evidence for a CDW instability could also originate from the hole redistribution between the chains and the ladders upon Ca-doping, which leads to a change of the chains and ladders nuclear space group symmetry. These space group symmetry changes could affect q_{CDW}.  

<table>
<thead>
<tr>
<th></th>
<th>I(0, 0, 1)</th>
<th>I(1, 0, 1)</th>
<th>I(3, 0, 1)</th>
</tr>
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<tbody>
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<td>4F1</td>
<td>D7</td>
</tr>
<tr>
<td>I_{mag}(mbarn)</td>
<td>0 ± 5</td>
<td>6 ± 4.0</td>
<td>16 ± 10</td>
</tr>
<tr>
<td>SCCO — 8</td>
<td>I_{b}</td>
<td>-1 ± 2</td>
<td>3.1 ± 1.7</td>
</tr>
<tr>
<td></td>
<td>I_{ac}</td>
<td>1 ± 2</td>
<td>2.9 ± 1.8</td>
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<tr>
<td>I_{mag}(mbarn)</td>
<td>11.4 ± 2.9</td>
<td>7.1 ± 2.9</td>
<td>-</td>
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<tr>
<td>SCCO — 5</td>
<td>I_{b}</td>
<td>4 ± 1.7</td>
<td>1.8 ± 1.7</td>
</tr>
<tr>
<td></td>
<td>I_{ac}</td>
<td>7.4 ± 1.7</td>
<td>5.3 ± 1.7</td>
</tr>
</tbody>
</table>

Supplementary Table S1. Summary of the measured magnetic intensities for SCCO — 5 and SCCO — 8 on D7 and 4F1.
In addition, it is worth noticing that $T_{CDW}$ decreases with increasing the Ca content, while the ordering temperature $T_{mag}$ of the new short ranged magnetic signal keeps growing. This may highlight an interesting competition between the $q = 0$ magnetism and the $CDW$, which could affect the development of the magnetic correlations.

SUPPLEMENTARY NOTE 8: MAGNETIC PATTERNS AND RELATED STRUCTURE FACTORS

To describe the observed magnetic intensities, one needs to calculate the momentum dependence of the magnetic cross-sections (Eq. 1) for given magnetic patterns. Eq. 1 contains essentially two terms depending on $Q$: the magnetic form factor, $f(Q)$, which depends on the nature of the magnetic moments and the magnetic structure factor $F(Q)$ which is the Fourier transform of the given magnetic pattern. We discuss in this section several magnetic models and calculate $F(Q)$ for each of them.

Supplementary Figure S7.a shows the ladder unit cell in the a-c plane. The building block is given by a $CuO_2$ square plaquette with a characteristic length of the square lattice, $a_s = c$. Hereafter, all positions in real space are given in units of $a_s$. Note that as $a \sim 3a_s$, the size of ladder unit cell is approximately the size of 3 square plaquettes, although the ladder unit cell contains 4 inequivalent Cu atoms. Indeed, as shown in Supplementary Figure S7.a, the $Cu_4O_6$ unit cell is made of a first $Cu_2O_3$ ladder with two Cu sites on a rung at coordinates of (0,0) and (1,0). The second ladder is obtained by a translation of these coordinates by $(3/2,1/2)$. This gives 4 distinct Cu sites distributed on 2 ladders. Each Cu site is at the center of a $CuO_2$ plaquette, with O sites at $(\pm 1/2,0)$ and $(0, \pm 1/2)$ around the Cu site. Note that there are only 6 distinct oxygen sites, since the two $CuO_2$ plaquettes on the same rung share one oxygen along the rung. Using the $CuO_2$ plaquette as a building block, one can decorate it with various magnetic patterns (Supplementary Figure S7.b-f). The magnetic dipoles can be related to a spin moment on Cu sites, spin or orbital moments on O sites, or orbital moments produced by loop currents (LC) between Cu and O sites or O sites only.

Antiferromagnetic Cu spins

We consider a ladder where Cu spins are coupled antiferromagnetically (Supplementary Figure S8.a). This model comprises antiferromagnetic interactions along the ladder legs and rungs (due to superexchange interaction across the $180^\circ$ oxygen bridge between $Cu$ ions). The existence of an antiferromagnetic order at long range is questionable. Indeed such a spin arrangement within ladders generates a frustration of the interladder magnetic interaction, due to the $90^\circ$ oxygen bridges between neighboring ladders (Supplementary Figure S8.a). In this limit, one would actually expect the individual ladders to be in a non-classical state of singlets on each rung, forming spin dimers [S17]. This corresponds to a non magnetic ground state, without any magnetic static fingerprint observable. Here, our purpose is to look for the origin of a short range magnetism located at bragg positions. We therefore simply ask the question whether or not the Cu spins on a single ladder may lead to magnetic signal observed in neutron diffraction. Thus is at least valid for the isolated ladders limit of $SCCO-5$. As a consequence, one considers a set of independent antiferromagnetic ladders to compute the squared magnetic structure factor present in Eq. 1:

$$|F(Q)|^2 = |4 \sin(\pi L) \sin(\pi H/3)|^2$$  

The two terms describe the antiferromagnetic coupling between 2 Cu spins along the leg and along the rung, respectively. This model breaks the lattice translation symmetry and should give a net magnetic contribution at half integer values of $H$ and $L$ which we did not observe during our experiment in $SCCO - 8$ (Supplementary Figure S8.b). It further rules out any magnetic scattering for integer $H$ or $L$ values which is at odds with our experimental measurements, where the magnetic scattering was observed at $Q$-positions of the form $(H,0,1)$ with integer $H$. One can therefore eliminate a conventional Cu spin antiferromagnetism as the origin of the observed magnetic scattering.
Magnetic moments on oxygen sites

Next, we consider a magnetic nematic model [S3, S18–S20] where O sites within a CuO$_2$ plaquette carry magnetic moments (spin or orbital) pointing in opposite directions for oxygen atoms located either along a or c directions with respect to the Cu site (Supplementary Figure S9). Once the ladders are decorated with such a magnetic nematic patterns, one observes 3 spins on O sites coupled ferromagnetically along a. They are coupled antiferromagnetically with 3 other spins translated by (1/2,1/2). This gives a squared magnetic structure factor, as follows:

$$|F(Q)|^2 = |2 \sin \left( \frac{\pi}{2} \left( \frac{H}{3} + L \right) \right) (1 + 2 \cos(2\pi \left( \frac{H}{3} \right)))|^2$$ (13)

The last term accounts for the ferromagnetic lines with 3 spins and the first term gives the Q-space relationship between neighboring lines with opposite spin directions. Such a magnetic pattern gives an extinction at (3,0,1) (Supplementary Figure S9.b). The model with magnetic moments on O sites (Supplementary Figure S9) then fails to account for the observation of a neighboring lines with opposite spin directions. Such a magnetic pattern gives an extinction at (3,0,1) (Supplementary Figure S9.b). The model with magnetic moments on O sites (Supplementary Figure S9) then fails to account for the observation of a magnetic signal at (3,0,1).

Loop current phases

We now discuss different magnetic patterns based on three distinct loop current models, shown in Supplementary Figure S7.d-f, all preserving the lattice translation symmetry.

Single LC pattern

Each CuO$_2$ square plaquette can be decorated by a single loop current (LC) pattern as shown in Supplementary Figure S7.d-f. For the three LC models, there are 4 different possibilities of putting the LC pattern around a given Cu site by making $\pi/2$ rotations. One can then write down the structure factor of a single LC pattern $F(Q) \equiv A_\phi(Q)$ with $\phi=\{0, \frac{\pi}{2}, \pi, \frac{3\pi}{2}\}$, where $\phi$ denotes the angle of rotation for each pattern. One can conveniently describe all the different situations by introducing a local toroidal moment for a given CuO$_2$ plaquette $i$ : $\Omega_i = \sum_j m_j \times r_j$, with $m_j$ a magnetic moment and $r_j$ its position with respect to the Cu site at the center of the CuO$_2$ plaquette [S21–S23]. Taking each of the four $\phi$ values, $\Omega_i$ is pointing along the diagonal separating the clockwise and anticlockwise LCs. We consider three distinct LC states:

- **CC – $\theta_1$** (Supplementary Figure S7.d): Theoretical works on copper oxide ladders predicted the appearance of a $CC – \theta_1$ phase with LCs in hole-doped SCCO [S4, S5, S24]. On each CuO$_2$ plaquette, there are 4 LCs which generate staggered orbital moment at positions: $(\pm x_0, \pm x_0)$ with respect to a Cu site. $x_0 \sim 0.146$ is the coordinate of the triangle center of mass, where the orbital moment is assumed to be. This state is twofold degenerate with: $A_{\phi\pm\frac{\pi}{2}}(Q) = -A_{\phi}(Q)$. The magnetic structure factor is independent of $\phi$ as:

$$A_{\phi}(Q) = \pm 4 \sin(2\pi x_0 \frac{H}{3}) \sin(2\pi x_0 L)$$ (14)

$|A_{\phi}(Q)|^2$ for $CC – \theta_1$ is shown on Supplementary Figure S10.a (green line).

- **CC – $\theta_{11}$** (Supplementary Figure S7.e): This state [S6, S25] breaks the inversion and time-reversal symmetry. In each square plaquette, it is made of two LCs turning clockwise and anti-clockwise and aligned along one diagonal ($\epsilon = +1$) or the other ($\epsilon = -1$). This state is fourfold degenerate with : $A_{\phi\pm\frac{\pi}{2}}(Q) = -A_{\phi}(Q)$ and $A_{\phi\pm\frac{3\pi}{2}}(Q) = A_{\phi}(Q)$, corresponding to both orientations or domains ($\epsilon = \pm1$). The two LCs produce 2 staggered orbital moments located with respect to the Cu site again at positions: $\pm(x_0, \epsilon x_0)$. The magnetic structure factor reads:

$$A_{\phi}(Q) = 2 \sin(2\pi x_0 (\frac{H}{3} + \epsilon L))$$ (15)

$|A_{\phi}(Q)|^2$ for the two different $CC – \theta_{11}$ orientations ($\epsilon = \pm1$) are shown on Supplementary Figure S10.a (dashed blue lines). The full blue line represents an averaged of both domains with equal population.
The selection rule: the results of the sample SCCO-5 and (iii) to SCCO-8, respectively.

Inter-ladder correlations with the site translated by $(3/2, 1/2)$. Both types of correlations contribute to the structure factor. Therefore, two types of LC correlations should be considered: i) the intra-rung correlations with the site shifted by $(1,0)$ ii) the inter-rung correlations with the site shifted by $(3/2, 1/2)$. Both types of correlations contribute to the structure factor.

**Intra-rung correlations:** For the intra-rung, one considers two patterns on each of both Cu sites of a rung, $A_\phi(Q)$ and $A_{\phi'}(Q)$. Defining $\phi = \phi - \phi'$ and depending on the correlations, the pattern shifted by $(1,0)$ is either identical ($\phi = 0$ and $A_{\phi'}(Q) = A_\phi(Q)$) or opposite ($\phi = \pi$ and $A_{\phi'}(Q) = -A_\phi(Q)$). In general, the intra-rung structure factor, $B_{\phi\phi'}(Q)$, can be written as:

$$B_{\phi\phi'}(Q) = [A_\phi(Q) + A_{\phi'}(Q)]\cos(\frac{\pi H}{3}) + i[A_\phi(Q) - A_{\phi'}(Q)]\sin(\frac{\pi H}{3})$$  \hspace{1cm} (18)

$|B_{\phi\phi'}(Q)|^2$ is shown on Supplementary Figure S10.b for both correlations. Interestingly, this term gives zero structure factor at $(3,0,1)$ for the opposite patterns ($\phi = \pi$) at variance with the experimental results. This implies that both patterns are identical ($\phi = 0$) and the intra-rung structure factor can be always simplified as:

$$B_{\phi}(Q) = 2A_\phi(Q)\cos(\frac{\pi H}{3})$$  \hspace{1cm} (19)

**Inter-ladder correlations:** The case of the inter-ladder is obtained in the same way defining two coupled LCs, $B_\phi(Q)$ and $B_{\phi'}(Q)$, shifted by $(3/2, 1/2)$. Again, defining $\psi = \phi - \phi'$ and depending on the correlations, the pattern shifted by $(3/2,1/2)$ is either identical ($\psi = 0$ and $B_{\phi'}(Q) = B_\phi(Q)$) or opposite ($\psi = \pi$ and $B_{\phi'}(Q) = -B_\phi(Q)$) as for the intra-rung correlations. However, it is also possible that the LC shifted by $(3/2,1/2)$ is aligned along a different diagonal than the one of the first ladder, $\psi = \pm \frac{\pi}{2}$ and $B_{\phi\pm \frac{\pi}{2}}(Q) \neq B_\phi(Q)$. The inter-ladder structure factor can then be written as $C_{\phi\phi'}(Q)$ as:

$$C_{\phi\phi'}(Q) = [B_\phi(Q) + B_{\phi'}(Q)]\cos(\frac{\pi}{2}(H + L)) + i[B_\phi(Q) - B_{\phi'}(Q)]\sin(\frac{\pi}{2}(H + L))$$  \hspace{1cm} (20)

$|C_{\phi\phi'}(Q)|^2$ is shown on Supplementary Figure S10.c for both correlations $\psi = 0$ and $\psi = \pi$. For in-phase ladders, one obtains the selection rule: $H + L = 2n$. The out-of-phase case ($\psi = \pi$) is ruled out by the experiments as it gives zero structure factor at $(3,0,1)$.

Finally, the LCs magnetic structure factor $F(Q)$, present in Eq. 1, corresponds to (i) $A_\phi(Q)$ for an independent single pattern, (ii) $B_\phi(Q)$ for an independent ladder and (iii) $C_{\phi\phi'}(Q)$ for coupled ladders. It is worth to recall that the case (ii) corresponds to the results of the sample SCCO-5 and (iii) to SCCO-8, respectively.
**CC – Θ_I like phase of LCs**

The CC – Θ_I intra-rung pattern can be taken in-phase (φ = 0) as requested but the inter-ladder patterns are necessary out-of-phase as shown in Supplementary Figure S11.a as they share a current link along the diagonal of the inter-ladder small square. This gives the following \( |F(Q)|^2 \):

\[
|F(Q)|^2 = |16 \sin\left(\frac{\pi}{2} (H + L)\right) \cos\left(\frac{\pi}{3} H\right) \sin(2\pi x_0 \frac{H}{3}) |^2
\]

(21)

According to the previous section, the third term (in brackets) corresponds to the CC – Θ_I pattern, the second term accounts for the ordering in-phase within the ladder and the first term the out-of-phase coupling between ladders. Such a structure factor gives rise to magnetic scattering extinction rules that do not account for our experimental observations (Supplementary Figure S11.b). For instance, it prohibits scattering when H and L are both odd, at variance with our observed magnetic scatterings at (1,0,1) and (3,0,1). Even in the case of SCCO-5 (independent ladder), it does not correspond to the results as \( |F(Q)|^2 = 0 \) for (0,0,1) where the magnetic signal is observed.

**CC – Θ_{II} like phases of LCs**

- **Uncorrelated ladders: SCCO – 5**

We first discuss the case of the independent ladders using the CC – Θ_{II} pattern (Supplementary Figure S12.a). Within the ladder unit cell, the first ladder is decorated with in-phase pattern, whereas no LCs occur for the second ladder. \( |F(Q)|^2 \) reduces to a product of the in-phase intra-rung term times the magnetic pattern of Eq. 15:

\[
|F(Q)|^2 = \sum_{\epsilon=\pm 1} \frac{1}{2} |4 \cos(\pi \frac{H}{3}) \sin(2\pi x_0 (\frac{H}{3} + \epsilon L))|^2
\]

(22)

Here, we assumed four possible domains with equal population. This structure factor (shown in Supplementary Figure S12.d) reproduces the SRM along the (H,0,1) line (see Fig. 5.b of the manuscript), while the magnetic correlations along a (perpendicular to the ladders) are confined within a single ladder.

- **Correlated ladders: SCCO – 8**

The magnetic structure factor is now given by Eq. 20. There are three different ways to couple the first and second ladders within the SCCO unit cell, corresponding to different phase shift \( \psi \). The ladders couple in-phase (\( \psi=0 \)) (Supplementary Figure S12.b), out-of-phase (\( \psi=\pi \)) or exhibit a crisscrossed coupling (\( \psi=\pm \frac{\pi}{2} \)) (Supplementary Figure S12.c). For the last case where \( |B_{\psi'}(Q)| \neq |B_{\psi}(Q)| \) in Eq. 20, two different situations are possible to orient the magnetic patterns shifted by (3/2,1/2) denoted (\( \delta = +1 \)) and (\( \delta = -1 \)). Using the toroidal moment formalism (see section above), one can define \( \Omega = \sum \Omega_i \) the effective toroidal moment for the full SCCO unit cell. For the in-phase ladders (\( \psi=0 \)), \( \Omega \) remains along the same diagonal, whereas this vector is null for out-of-phase ladders. Interestingly, for the two crisscrossed cases, \( \Omega \) points either along a rung, i.e the direction \( a (\delta = +1) \) or along a leg, i.e along the direction \( c (\delta = -1) \). The related squared structure factors, \( |F(Q)|^2 = |C_{\psi'}(Q)|^2 \), are:

\[
\psi = 0 : |F(Q)|^2 = \sum_{\epsilon=\pm 1} \frac{1}{2} |8 \cos\left(\frac{\pi}{2} (H + L)\right) \cos(\pi \frac{H}{3}) \sin(2\pi x_0 (\frac{H}{3} + \epsilon L))|^2
\]

(23)

\[
\psi = \pi : |F(Q)|^2 = \sum_{\epsilon=\pm 1} \frac{1}{2} |8 \sin\left(\frac{\pi}{2} (H + L)\right) \cos(\pi \frac{H}{3}) \sin(2\pi x_0 (\frac{H}{3} + \epsilon L))|^2
\]

(24)

\[
\psi = \pm \frac{\pi}{2}, \delta = \pm 1 : |F(Q)|^2 = \left\{ \begin{array}{l}
|2 \sin(2\pi x_0 (\frac{H}{3} + L)) + \delta 2 \sin(2\pi x_0 (\frac{H}{3} - L))|^2 \cos^{\frac{2}{\pi}}(\frac{H}{3})

+ |2 \sin(2\pi x_0 (\frac{H}{3} + L)) - \delta 2 \sin(2\pi x_0 (\frac{H}{3} - L))|^2 \sin^{\frac{2}{\pi}}(\frac{H}{3})
\end{array} \right\}
\]

. \left| 2 \cos\left(\frac{\pi}{2}\right) \right|^2
\]

(25)
Among all $|F(Q)|^2$ (Supplementary Figure S12.d), the squared structure factor for the in-phase case (Supplementary Figure S12.b) is the one which reproduces the main features of our experimental results, namely: i) the absence of scattering at (0,0,1), ii) a scattering at odd H and L, iii) a stronger scattering at H=3 than at H=1. The crisscrossed $CC - \theta_{111}$ with the effective toroidal moment along the ladder ($\psi = \pm \frac{\pi}{2}, \delta = -1$), shown Supplementary Figure S12.c, is also consistent for the observed SRM, even if the difference of intensities between H=3 and H=1 is less pronounced in that case. Such a crisscrossed structure indicate that the effective toroidal moment should be along the ladder. Note that in bilayer cuprates $YBa_2Cu_3O_{6+x}$, the effective toroidal moment is found parallel to the underlying $CuO$ chains [S23].

**SUPPLEMENTARY NOTE 9: MAGNETIC FORM FACTOR**

We have considered various magnetic pattern within the $CuO_2$ plaquette involving spin on the Cu site, or spin/orbital moment on the oxygen sites and orbital moments originating from LCs. Another factor present in Eq. 1 is the magnetic form factor. Two different form factors can be considered here, either the isotropic magnetic Cu-form factor or the oxygen-one (which can be estimated from ref. [S26]). For LC states, electron are delocalized between several Cu and O sites or O sites only, but the exact form factor associated with the induced orbital moment remains unknown. Previous PND measurements in 2D cuprates [S27] suggested that both magnetic Cu- and O-form factor could be used. Supplementary Figure S14.a shows the $Q$-dependencies of $f(Q)^2$ for copper and oxygen. The $Q$-range of interest for our study is indicated by a shaded area, where $f_{Cu}^2(Q)$ varies of 47% against 21% for $f_{O}^2(Q)$. In principle, since electrons are likely to be more delocalized for LCs, the magnetic O-form factor could be best suited to describe a fast decay of LCs magnetic signal as compared to the magnetic Cu-form factor. However, fitting the $Q$-dependence of the magnetic signal with either form factors gives good agreement with our data and the extracted magnetic moment amplitudes are, although different, of the same order of magnitude when considering the $O$ or $Cu$ form factors, as will be shown in the next section, Supplementary Table S2.
SUPPLEMENTARY NOTE 10: MAGNETIC MOMENT AMPLITUDES AS EXTRACTED FROM DATA MODELING

- **Out-of-plane magnetic moment: \( m_b \)**

As discussed above, among the different magnetic patterns of Supplementary Figure S7.b-f, some of the LCs-like phases can describe our experimental data. In principle, being confined in the (a,c) plane, classical LCs produce an orbital moment, \( m_b \equiv m_{LC} \), pointing perpendicular to the LC plane. That corresponds to the magnetic intensity, \( I_b \), that we have extracted from XYZ-PA (Supplementary Table S1). Using \( I_b \propto m_b^2 \), one can deduce the out-of-plane magnetic moment, \( m_b \equiv m_{LC} \). Supplementary Figure S14.d-e show \( I_b \) calibrated in mbarns fitted by different models. The evolution of the magnetic intensity along (H,0,1) is rather different for SCCO-5 (Supplementary Figure S14.d) and SCCO-8 (Supplementary Figure S14.e). No correlation develops between the ladders in the former case and the magnetic scattering remains diffusive. For the latter case, magnetic correlations develop between the ladders. The scattered intensity \( I_b \) can be described as:

\[
\text{SCCO} - 5 : \quad I_b(Q) = r_o^2 f(Q)^2 m_b^2 |A(Q)|^2 |2 \cos(\pi \frac{H}{Q})|^2
\]

\[
\text{SCCO} - 8 : I_b(Q) = r_o^2 f(Q)^2 m_b^2 |A(Q)|^2 |2 \cos(\pi \frac{H}{Q})|^2 |2 \cos(\frac{2\pi}{3}(H + L))|^2
\]

The Figs. S14.b-c show the effect of a \( Q \)-independent magnetic pattern, \( |A(Q)|^2 = constant \), with in-phase coupling within a rung, on the one hand, and, between ladder, on the other hand. That reproduces the main features of the evolution of \( I_b \) along (H,0,1). Next, in the Figs. S14.d-e, we add the evolution of the magnetic pattern of each LC phase, \( |A(Q)|^2 \). Supplementary Figure S14.d-e show the best fits of the PND data with \( |A(Q)|^2 \) corresponding to the different \( CC - \theta_{11} \) and \( CC - \theta_{111} \) LCs. The deduced values of \( m_b \) are listed in supplementary Table S2.

- **In-plane magnetic moment: \( m_{ac} \)**

One can perform the same analysis for the full measured intensities \( I_{mag} \) although, in principle, the classical LCs phase cannot account for the in-plane magnetic intensities reported in Supplementary Table S1. \( I_{mag} \) is proportional to \( m_{ac}^2 = m_a^2 + m_c^2 \), with \( m_{ac}^2 = (1 - |\frac{H}{Q}|^2)m_a^2 + |\frac{H}{Q}|^2 m_c^2 \). As it combines magnetic moments along the ladder and along the rung, \( m_{ac} \) varies with \( Q \) due to the orientation factors. For a H-scan along (H,0,1), the variations of these orientation factors, weighting \( m_a^2 \) and \( m_c^2 \) are given in Supplementary Figure S15. Unfortunately, the set of collected data for both samples remains insufficient to determine independently \( m_a \) and \( m_c \). As a consequence, one further constrains the fits by enforcing \( m_a = m_c \). This simple assumption eliminates the Q-dependence of \( m_{ac} \).

Supplementary Figure S15.b-c show the fit of the PND data for samples \( \text{SCCO} - 5 \) and \( \text{SCCO} - 8 \) using the same functions as in Supplementary Figure S14.d-e. One sees that the same LCs structure factor properly account for the full measured magnetic intensities. The resulting magnetic moments \( m_{ac} \) are listed in Supplementary Table S2. From these results, for both samples, one finds \( m = (m_a, m_b, m_c) \) with typically \( |m| \approx 0.09 \mu_B \) that value depends on the specific LCs pattern considered via \( |A(Q)|^2 \). That gives as well a tilt angle of 55° of the magnetic moment with respect to the \( b \) axis.

<table>
<thead>
<tr>
<th>( f(Q)^2 )</th>
<th>( \text{LC pattern} )</th>
<th>( m_b(x = 8) )</th>
<th>( m_b(x = 5) )</th>
<th>( m_{ac}(x = 8) )</th>
<th>( m_{ac}(x = 5) )</th>
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<td>Cu</td>
<td>( CC - \theta_{11} )</td>
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<td>0.05</td>
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<td>O</td>
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<tr>
<td>Cu</td>
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</table>

Supplementary Table S2. Out-of-plane magnetic moment \( m_b \) (\( \mu_B \)) and measured magnetic moment \( m_{ac} = \sqrt{m_a^2 + m_c^2} \) (\( \mu_B \)) for both samples \( \text{SCCO} - x \). The values are shown for both Cu and O form factors and for the different LCs magnetic patterns. Typical error on the estimation of the moment is 0.01 \( \mu_B \).
SUPPLEMENTARY NOTE 11: ORIGIN OF THE PLANAR COMPONENT

In the original model proposed by C.M. Varma, LCs are confined within the CuO$_2$ planes. This should generate only orbital magnetic moments perpendicular to the ladder planes, which is at variance with the experimental observation where an extra in-plane magnetic scattering is reported. Then, it was suggested that the ground state could not be solely made of one of the four orthogonal $CC - 	heta_{11}$ states, but could rather emerge by their quantum superposition [S21, S22]. Within that framework, the degree of quantum admixture shows up in PND measurements in the form of an extra magnetic scattering that looks like that originating from an effective magnetic planar component. In $Cu_2O_3$ ladders, LCs settle in at lower temperature where quantum effects might be larger than thermal fluctuations. This makes the proposal of quantum effect at the origin of the planar magnetic scattering an interesting scenario.

Alternatively, it was proposed in superconducting cuprates that the planar component arises from LCs running over the CuO$_6$ octahedron [S2, S28–S30]. Indeed, the cuprates, where the $q=0$ magnetism was observed in monolayer and bilayer materials, are all containing CuO$_6$ octahedron and a apical oxygen site. In the CuO$_2$ layers, the Cu site is located at the center of either a CuO$_6$ octahedron or a CuO$_5$ pyramid. It was therefore proposed that LCs could delocalize on opposite edges of O-octahedra or O-pyramids, yielding a magnetic planar component. Whatever is the relevance of such a proposal for superconducting cuprates, it cannot hold for two-leg ladder cuprates, since there is no apical oxygen above the $CuO_2$ plaquette. For that kind of materials, LCs have to remain confined with the $Cu_2O_3$ ladders.

SUPPLEMENTARY REFERENCES:


