Magnetic order in the pseudogap phase of high-\(T_C\) superconductors

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One of the leading issues in high-\(T_C\) superconductors is the origin of the pseudogap phase in underdoped cuprates. Using polarized elastic neutron diffraction, we identify a novel magnetic order in the YBa\(_2\)Cu\(_3\)O\(_{6+x}\) system. The observed magnetic order preserves translational symmetry as proposed for orbital moments in the circulating current theory of the pseudogap state. To date, it is the first direct evidence of an hidden order parameter characterizing the pseudogap phase in high-\(T_C\) cuprates.

In optimally and underdoped regimes, high-\(T_C\) copper oxides superconductors exhibit a pseudogap state\(^2, 3, 4\) with anomalous magnetic\(^5\), thermodynamic\(^6\) and optical\(^7\) properties below a temperature, \(T^*\), large compared to the superconducting transition temperature, \(T_c\). The origin of the pseudogap is a challenging issue as it might eventually lead to identify the superconducting mechanism\(^8\). Two major classes of theoretical models attempt to describe the pseudogap state: in a first case, it represents a precursor \(\Theta_I\) of the superconducting mechanism. In the second approach, the pseudogap is associated with either \(\Theta_{II}\) or the phase \(\Theta_{II'}\) preserving \(TSL\) competing with the SC one. The order parameter, associated with these competing phases may involve charge and spin density waves\(^8\) or orbital circulating currents (CC)\(^11, 12\) competing with the SC one.

Most of these phases break the translation symmetry of the lattice (TSL). Therefore, they may induce charge, nuclear or magnetic superstructures that can be probed by neutron or X-ray diffraction techniques. In contrast, CC phases\(^11, 12\) preserve the TSL as they correspond to 4 or 2 current loops per unit cell (referred as \(\Theta_I\) and \(\Theta_{II}\) phases, respectively). These charge currents could be identified by virtue of the pattern of ordered orbital magnetic moments pointing perpendicularly to the CuO\(_2\) planes. These orbital magnetic moments should be detectable by neutron diffraction. Although the TSL is preserved, the magnetic signature of the CC phase does not reduce to ferromagnetism: the loops are staggered within each unit cell corresponding to a zero magnetic propagation wavevector, \(Q=0\), but with no net magnetization. In neutron diffraction, the magnetic intensity superimposes on the nuclear Bragg peak, meaning that these experiments are very delicate as the magnetic intensity \(\propto M^2\) (\(M\) is the magnetic moment) is expected to be very small as compared to the nuclear Bragg peaks. In order to detect this hidden magnetic response, polarized neutron experiments are then required.

As proposed by C.M. Varma\(^11, 12\), there are two possibilities CC phases preserving TSL. The first (the phase \(\Theta_I\)) has not been detected by polarized elastic neutron scattering experiments\(^19, 20\). Although it is controversial, a recent ARPES measurement observed a dichroic signal in the Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_{6+\delta}\) system consistent with the phase \(\Theta_{II'}\)\(^21\). Here, we have performed polarized elastic neutron scattering experiments to test the magnetic moments of this second CC state, phase \(\Theta_{II'}\), which actually had never been attempted before. We successfully report the first signature of a novel magnetic order in the pseudogap state of YBa\(_2\)Cu\(_3\)O\(_{6+x}\) (YBCO). The pattern of the observed magnetic scattering corresponds to the one expected in the circulating current theory of the pseudogap state with two current loops per CuO\(_2\) unit-cell, phase \(\Theta_{II'}\). Alternatively, a decoration of the unit cell with staggered moments on the oxygen sites could also account for the measurements.

All the polarized neutron diffraction measurements were collected on the 4F1 triple-axis spectrometer at the Laboratoire Léon Brillouin (LLB), Saclay (France). Our polarized neutron diffraction setup is similar to that originally described in \(^22\) with a polarizing incident neutron with at \(E_i = 14.7\) meV obtained with a polarizing supermirror (bender) and with an Heusler analyzer (see also ref. \(^19, 26\) in the context of high-\(T_C\) cuprates). The direction of the neutron spin polarization, \(\mathbf{P}\), at the sample position is selected by a small guide field \(\mathbf{H}\) of the order of 10 G\(^24\). Using that configuration, we monitor for each measured point the neutron scattering intensity in the spin-flip (SF) channel, where the magnetic intensity \(\propto M^2\) is expected, and in the non-spin-flip (NSF) channel which measures the nuclear scattering. To have similar counting statistics on both SF and NSF, we count the SF channel systematically 20 times longer than the NSF. We define the normalized spin-flip intensity as \(I_{norm} = I_{SF}/I_{NSF}\) (inverse of the flipping ratio (FR)). With that setup, a typical flipping ratio, ranging between 40 and 60, is obtained. However, even with that high FR, the SF intensity is massively coming from the NSF nuclear Bragg peak through unavoidable polarization leakage (corresponding to about \(\sim 90-95\%)
of the SF intensity). As a very stable and homogeneous neutron polarization is essential through the data acquisition, all the data have been obtained in a continuous run versus temperature. We prove that method to be efficient enough to see weak magnetic moments (\(\sim 0.05 \mu_B\)) on top of nuclear Bragg peaks, see e.g. the first determination of the A-type antiferromagnetism in Na cobaltate systems \(\text{[22]}\).

We quote the scattering wave vector as \(\mathbf{Q} = (H,K,L)\) in units of the reciprocal lattice vectors, \(a^* \approx b^* = 1.63\ \text{Å}^{-1}\) and \(c^* = 0.53\ \text{Å}^{-1}\). Most of the data have been obtained in a scattering plane where all Bragg peaks like \(\mathbf{Q} = (0,K,L)\) were accessible (in twinned samples, this is indistinguishable from Bragg peaks with \(\mathbf{Q} = (H,0,L)\)). In order to evidence small magnetic moments, measurements have been performed on the weakest nuclear Bragg peaks having the proper symmetry for the CC phase \(\text{[26]}\) (the Bragg peak \(\mathbf{Q} = (0,1,1)\) offers the best compromise).

We have studied 5 different samples (see Table I): 4 samples in the underdoped regime and one in the overdoped regime. In Fig. 1a, we report the raw neutron intensity measured at \(\mathbf{Q} = (0,1,1)\) for the spin flip (SF) channel and for the non-spin-flip (NSF) channel for an underdoped sample \(\text{YBa}_2\text{Cu}_3\text{O}_{6+\delta}\) (sample C). The measurement has been done with a neutron polarization \(\mathbf{P} // \mathbf{Q}\) (see Fig. 1b) where the magnetic scattering is entirely spin-flip \(\text{[14, 22, 23, 24]}\). Between room temperature and a temperature \(T_{mag} \approx 220\text{K}\), the NSF and SF intensities display the same evolution within error bars. Then, for \(T < T_{mag}\), the NSF is essentially flat whereas the SF intensity increases noticeably at low temperature. This behaviour signals the presence of a spontaneous magnetic order below \(T_{mag}\) on top of the nuclear Bragg peaks. In Fig. 1c, we show the normalized magnetic intensity as a function of the temperature for the 4 underdoped samples and the overdoped sample. For the 4 underdoped samples, the magnetic intensity increases at low temperature below a certain temperature \(T_{mag}\) whereas no magnetic signal is observed in the Ca-YBCO overdoped sample (sample E).

We perform further measurements where the neutron polarization is along the complementary directions, as shown in Fig. 1b, either the vertical direction \(\mathbf{P} // \mathbf{z}\), or \(\mathbf{P} \perp \mathbf{Q}\) but still within the horizontal scattering plane. The observance of the polarization selection rule for a

| label | \(x\) | \(T_{onset} \text{[K]}\) | \(T_{mag} \text{[K]}\) | References |
|-------|------|----------------|----------------|------------|
| A     | \(\text{O}_{6.5}(t)\) | ud 54 | 300 ± 10 | \[23\] |
| B     | \(\text{O}_{6.6}(t)\) | ud 61 | 250 ± 20 | \[30\] |
| C     | \(\text{O}_{6.6}(d)\) | ud 64 | 220 ± 20 | \[20\] |
| D     | \(\text{O}_{6.75}(t)\) | ud 78 | 170 ± 30 | - |
| E     | \(\text{Ca}(15%) - \text{O}_{7-x}(t)\) | od 75 | \(\approx 0\) | - |

TABLE I: List of samples utilized in the polarized elastic neutron experiments. The experiments were performed in the \((\text{Y},\text{Ca})_2\text{Cu}_3\text{O}_{6+\delta}\) family for 5 samples from the underdoped (ud) to overdoped (od) part of the cuprates phase diagram. (t) and (d) stands for twinned and detwinned samples, respectively. References are given where the samples have been described in previous neutron scattering studies. In contrast to the other samples, an in-plane magnetic ordering occurred at \(\mathbf{Q} = (1/2,1/2)\) in sample A with \(M \sim 0.05 \mu_B\) at 60 K \(\text{[22]}\).
magnetic order is limited, showing that the magnetic order is characterized as leakage. The observed magnetic peak is resolution $K$ has been taken to remove the effect of the polarization. The difference in temperature between $T=75$ K and $275$ K is discussed. First, we perform a scan along the $L$-direction needed. However, some qualitative aspects can be briefly discussed. Next, we do not perform a detailed and quantitative determination of magnetic structure for which further work is needed. This directly arises from the hierarchy of the observed magnetic intensities (intensity at $L=0$ is larger than at $L=2$, Fig. 2b). Finally, using the observed magnetic cross-section (Fig. 2c) and a weakly momentum dependent form factor, one can deduce a typical magnitude of ordered magnetic moment of $M \simeq 0.05$ to $0.1 \mu_B$ with the moment decreasing with increasing doping in the 4 samples.

As shown in Fig. 2c, the typical cross-section of the magnetic order is $\sim 1–2$ mbarns, i.e. $\sim 10^{-4}$ of the strongest Bragg peaks. This explains why such a magnetic order was not reported before with unpolarized neutron diffraction. Due to these experimental limitations, we do not perform a detailed and quantitative determination of magnetic structure for which further work is needed. However, some qualitative aspects can be briefly discussed. First, we perform a scan along the $L$-direction in the SF channel across the Bragg peak (Fig. 2a) where the difference in temperature between $T=75$ K and $275$ K has been taken to remove the effect of the polarization leakage. The observed magnetic peak is resolution limited, showing that the magnetic order is characterized by long range 3D correlations at $T=75$ K. Second, by looking at other Bragg peaks along $c^*$ (Fig. 2b), we found that the magnetic intensity is not uniformly distributed versus $L$, meaning that i) the magnetic intensity does not arise from the Cu-O chains, and ii) the moments arrangement within a bilayer appears to be mainly parallel. This directly arises from the hierarchy of the observed magnetic intensities (intensity at $L=0$ is larger than at $L=2$, Fig. 2b). Finally, using the observed magnetic cross-section (Fig. 2c) and a weakly momentum dependent form factor, one can deduce a typical magnitude of ordered magnetic moment of $M \simeq 0.05$ to $0.1 \mu_B$ with the moment decreasing with increasing doping in the 4 samples.

Therefore, we observe an unusual magnetic order in a temperature and doping range that cover the range where the pseudogap state is observed in YBCO. Our data do not contradict previous unsuccessful polarized neutron reports as the Bragg spots where the effect is observed are along a direction at $45^\circ$ from the one previously studied. The deduced $T_{mag}$, defined as the change of slope in the normalized intensity $I_{mag}$, decreases with increasing doping (see table I). It matches the pseudogap temperature, $T^*$, of the resistivity data in YBCO as shown on Fig. 3. The occurrence of a magnetic order in this temperature and doping range points towards a magnetic signature of an hidden order parameter associated with the pseudogap state. As all
anomalous physical properties evidencing the pseudogap, the temperature dependence of the magnetic order does not exhibit a marked change at $T_{mag}$. Being on top of nuclear Bragg peaks, that magnetic order does not break TSL, indicating a zero magnetic propagation wavevector, $Q = 0$. As shown in Fig. 11c, no magnetic intensity occurs below $T_{mag}$ at the Bragg peak $Q = (0,0,2)$, ruling out a ferromagnetic order. The absence of breaking of TSL points towards a magnetic pattern of antiparallel magnetic moments within each unit cell. Among the proposed order parameters, only one gives magnetic scattering at $Q = (10L) \equiv (01L)$: it is the orbital moments arising from the circulating current phase with 2 current loops per CuO$_2$ unit cell, $\Theta_{II}$ (Fig. 8b). Another possibility could be a model with collinear spin current loops per CuO$_2$ plaquette as sketched on Fig. 8c. Any other model characterized by a decoration of the CuO$_2$ plaquette would also give rise to a magnetic contribution at the proper Bragg spots.

From our present measurements, one cannot distinguish between these two models. Only a detailed study of magnetic form factors would allow to differentiate the scattering from spin and orbital moments. However, this approach might be hampered by the existence of magnetic form factors associated with flux phases [28]. Alternatively, if considering spin models, one would rather expect moments lying within the CuO$_2$ plaquette as it is the case for copper spins in undoped cuprates. A reason should be found to explain why the moments exhibit an out-of-plane component. Whatever the origin of the observed order, its pattern challenges the single plaquette model. Any other model characterized by a decoration of the unit cell with staggered spin or orbital moments, the symmetry of the observed order corresponds to the one expected in orbital moments emanating from a circulating current state [11] [12].

We are very grateful to C.M. Varma for invaluable encouragement, critics and ideas on these experiments. We also thank B. Keimer, J.-M. Mignot, P. Monceau, L. Pintschovius, and L.-P. Regnault for their support.

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[1] M.R. Norman, D.P. Pines, & C. Kallin, preprint cond-mat/0507031.
[2] M.R. Norman & C. Pépin, Rep. Prog. Phys. 66, 1547 (2003).
[3] T. Timusk, & B. Statt, Rep. Prog. Phys. 62, 61 (1999).
[4] J.L. Tallon & J.W. Loram, Physica C 349, 53 (2001).
[5] H. Alloul et al. Phys. Rev. Lett. 63, 1700 (1989).
[6] T. Ito et al. Phys. Rev. Lett. 70, 3995 (1993).
[7] J.W. Loram et al. Physica C 235-240 134 (1994).
[8] P.A. Lee, Physica C 317–318, 194–204, (1999).
[9] V.J. Emery & S.A. Kivelson, Nature 374, 434 (1995).
[10] J. Orenstein, & A.J. Millis, Science 288, 468 (2000).
[11] C.M. Varma, Phys. Rev. B, 55, 14554 (1997); Phys. Rev. Lett. 83, 3538 (1999); preprint cond-mat/0507214.
[12] M.E. Simon & C.M. Varma, Phys. Rev. Lett. 89, 247003 (2002).
[13] S. Chakravarty et al. Phys. Rev. B 63, 094503 (2001).
[14] C. Castellani et al. Phys. Rev. Lett. 75, 4650 (1995).
[15] H.C. Chen, et al. Phys. Rev. Lett. 93, 187002 (2004).
[16] D. Poilblanc, cond-mat/0505249.
[17] J. Zaanan, et al. Phil. Mag. B, 81, 1485 (2001).
[18] F. Onufrieva, P. Pfeuty, Phys. Rev. Lett., 82, 3136 (1999).
[19] S.H. Lee et al. Phys. Rev. B 60, 10405 (1999).
[20] Ph. Bourges, L.P. Regnault, J.Y. Henry, & C. Marin, Unpublished data (1998).
[21] A. Kaminski, et al., Nature 416, 610 (2002); S. Borisenko, et al., Nature 431, 2 (September 2004); A. Kaminski, et al., ibid.
[22] R.M. Moon et al. Phys. Rev. 181, 920 (1969).
[23] Y. Sidis, et al. Phys. Rev. Lett. 86, 4100 (2001).
[24] Magnetic neutron diffraction always measures magnetic components perpendicular to the scattering wavevector. In a polarized experiment, only the magnetic components perpendicular to the neutron polarization direction contribute to the magnetic enhancement. In a polarized experiment, only the magnetic components perpendicular to the neutron polarization direction contribute to the spin-flip channel [22].
[25] S.P. Bayrakci et al. Phys. Rev. Lett. 94, 15705 (2005).
[26] Bragg magnetic peaks characteristic of the two CC states proposed [12] differ by 45°: main Bragg peaks like $Q = (11L)$ are expected for the state $\Theta_1$ and like $Q = (10L) \equiv (01L)$ for the state $\Theta_{II}$. In both cases, no magnetic contribution occurs on Bragg peaks like $Q = (00L)$.
[27] C. Wu, Y. Zaanan & S.C. Zhang, cond-mat/0505644.
[28] X.G. Wen et al. Phys. Rev. B, 39, 14143 (1989).
[29] V. Hinkov et al., Nature 430, 650 (2004).
[30] L. Pintschovius et al., Phys. Rev. Lett. 89, 037001 (2002).
[31] J.L Tallon et al., Phys. Rev. B, 51, R12911 (1995).