Search for the existence of circulating currents in high-$T_c$ superconductors using the polarized neutron scattering technique

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We review experimental attempts using polarized neutron scattering technique to reveal the existence in high temperature superconductors of a long-range ordered state characterized by the spontaneous appearance of current loops. We draw a particular attention to our recent results (B. Fauquê et al., Phys. Rev. Lett. 96, 197001 (2006)) that, up to now, can be explained only by the theory of circulating current proposed by C.M. Varma.

Since the discovery of high-$T_c$ copper oxide superconductors, a large variety of theoretical models has been proposed to capture the intrinsic microscopic nature of CuO$_2$ planes, where superconductivity develops. Several models postulate the existence of circulating currents (or current loops) in the CuO$_2$ planes that could be responsible for the exotic electronic properties of these materials, in particular the existence of the mysterious "pseudo-gap" phase. In underdoped high-$T_c$ copper oxide superconductors, this phase is notable from its departure from the behavior of conventional metal and it has been proposed that it could correspond to a long-range ordered phase associated with a new state of matter, that could either co-exist or compete with superconductivity.

In the staggered flux phase model $^8$ or $d$-wave charge density wave (DDW) model $^9,^{10,11,12}$, CuO$_2$ planes are characterized by the appearance of staggered circulating currents flowing into square copper plaquettes (Fig.1a). In contrast to these models, where both translation invariance and time reversal symmetry are broken, the time reversal symmetry is broken in the circulating current (CC) phase proposed by C.M. Varma, but translation invariance is preserved $^{13,14,15,16}$. The order parameter of CC-phase is made of a set of staggered circulating currents embedded in a given Cu square plaquette. Each current flows along a triangular path involving a copper site and two of its nearest neighbor oxygen sites (Fig.1b-d). Two distinct currents patterns have been proposed: the $\Theta_I$-phase made of 4 triangular loops (Fig.1b) or the $\Theta_{II}$-phase made of 2 triangular loops (Fig.1c) forming a "butterfly" centered on Cu site (Fig.1c-d).

Whatever its origin, the array of circulating currents in the CuO$_2$ planes generates a magnetic field distribution: for the staggered flux phase or the DDW phase, one may speak about antiferromagnetic (AF) orbital magnetism (with a prop-
agitation wave vector at \( \mathbf{q} = (\pi, \pi) \) and \( \mathbf{q} = 0 \) AF-orbital magnetism in the case of CC-phases. Neutron scattering technique can be used to probe the existence of circulating currents in high-\( T_c \) copper oxide superconductors. Neutron spin does indeed interact with any internal magnetic field. The magnetic intensity for scattering neutron is written:

\[
\frac{d\sigma}{d\Omega}|_{i\rightarrow f} \propto |<f|\sigma \mathbf{B}(\mathbf{q})|i>|^2
\]

where \( \sigma \) is the neutron spin and \(|i>| \) and \(|f>| \) are the initial and final states of the neutron. \( \mathbf{B}(\mathbf{q}) \) describes the Fourier transform of the magnetic field distribution at the wave vector \( \mathbf{q} \). When the internal magnetic field originates from the circulating currents \[8,12\], \( \mathbf{B}(\mathbf{q}) \) can be expressed in terms of the Fourier transform of the current density \( \mathbf{j}(\mathbf{q}) \), i.e. \( \mathbf{B}(\mathbf{q}) \propto \frac{i|\mathbf{q} \times \mathbf{a}|}{q^2} \). The neutron scattering cross section for polarized neutron then reads \[17\]:

\[
\frac{d\sigma}{d\Omega}|_{i\rightarrow f} \propto |<f|\mathbf{j}(\mathbf{q}) \times \mathbf{a}|i>|^2
\]

The neutron scattering intensity in the spin flip (SF) channel, \( I \), has to fulfill a polarization selection rule equivalent of the one for spin moments which in absence of chirality can be written:

\[
I_{P//\hat{a}} = I_{P//\hat{z}} + I_{P//\hat{a} \perp}
\]

where the unitary vectors \( \hat{a} \) and \( \hat{a} \perp \) are respectively parallel and perpendicular to wave vector \( \mathbf{q} \) in the scattering plane, and \( \hat{z} \) is perpendicular the scattering plane (Fig. 4d). For unpolarized neutron and taking into account the charge conservation constraint, the cross-section takes the simple form:

\[
\frac{d\sigma}{d\Omega} \propto \frac{<j(\mathbf{q})>^2}{q^2}
\]

The description of the current density could be quite complicated due to the nonzero size of the Wannier function, the mixing of different orbital, and many body effects when the current becomes substantial \[8,12\]. For sake of simplicity, it can be useful to associate an effective orbital magnetic moment to a given current loop. The moment is located at the center of the current loop and is aligned perpendicular to the CuO\(_2\) planes, owing to the planar confinement of the currents. Therefore, in contrast to a spin moment on Cu site, the effective orbital moment should be much more spread in real space. As a consequence, one may expect circulating currents to give rise to a neutron scattered magnetic intensity decreasing faster than the squared Cu magnetic form factor, \( F_{\text{Cu}}^2(\mathbf{q}) \), at large wave vector in agreement with Eq. 4.

Let us consider, at first, the case of AF orbital magnetism. There were several observations of an AF ordering in the underdoped regime of superconducting YBa\(_2\)Cu\(_3\)O\(_{6+x}\) \[18,19,20\]. When observed, this phase usually develops well above \( T_c \) (close to room temperature) and displays an ordered moment of about 0.05-0.02 \( \mu_B \). The polarization neutron analysis reveals that the AF phase is dominated by moments aligned in the CuO\(_2\) plane, as in the insulating AF parent compound. Considering, on the one hand, that other high quality single crystals with a similar doping level do not show a similar order \[21\] and, on the other hand, that impurity substitution out of the CuO planes (in the CuO chains) could induce an AF order at \( \sim 300 \text{K} \) in optimally doped samples \( T_c=93 \text{K} \) \[22\], it is likely that the observed AF order is not a generic property of the underdoped state and may be either remnant of the AF insulating state or induced by impurities or defects in the CuO chains.

Beyond this AF spin order, Mook \textit{et al.} \[21,22\] reported polarized neutron experiments suggesting the existence of a weak AF quasi-2D order, potentially characterized by: (i) magnetic moments perpendicular to the CuO\(_2\) plane, (ii) a decrease of the neutron scattering intensity at large wave vector larger that \( F_{\text{Cu}}^2(q) \). These experiments can be viewed as providing evidence in favor of the DDW model. However, it is worth pointing out that the estimated magnitude of the ordered moment is \( \sim 0.0025 \mu_B \), i.e close to the experimental threshold of detection \[22\]. Furthermore, the small number of studied Bragg reflections was not sufficient to get solid conclusions concerning the evolution of the magnetic signal at large wave vector. Finally, the lack of reproducibility of the neutron scattering data severely
questions their relevance for cuprates in general. In addition, the study of the charge excitation spectrum by angle resolved photo-emission do not show any indication of a doubling of the unit cell, resulting from breaking of the translation symmetry expected for the DDW phase. Therefore, there are no compelling evidences of circulating currents breaking translation invariance and leading to an orbital AF state.

Concerning the two possibles CC phases preserving translation symmetry proposed by C.M. Varma [13,14,15,16], the \( \Theta_{I} \)-phase was not detected by earlier polarized elastic neutron scattering experiments [24,25]. The \( \Theta_{II} \)-phase has been later on proposed [15] to account for an angle resolved photoemission experiment [26]. Using circularly polarized photons, A. Kaminski et al. [26] reported a dichroic signal in the Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_{8+\delta} \) system indicating a time reversal breaking symmetry in the pseudo-gap state.

Recently, this \( \Theta_{II} \)-phase has been searched with polarized neutron scattering using uniaxial polarization analysis [27]. In contrast to the earlier inconclusive attempts, experimental evidences in favor of the existence of the CC \( \Theta_{II} \)-phase were found [27]: a magnetic order, hidden in the pseudo-gap regime of high-\( T_c \) superconductors, has been observed. The study performed by Fauqué et al. [27] covers a large part of the phase diagram of high-\( T_c \) cuprates and show a remarkable reproducibility of the neutron results, that was missing in the previous studies devoted to DDW phase. Five different samples of different origin were indeed studied: 4 YBa\(_2\)Cu\(_3\)O\(_{6+x} \) samples in the underdoped (UD : \( T_c < T_{c,\text{max}} \)) regime and one Y\(_{0.85}\)Ca\(_{0.15}\)Ba\(_2\)Cu\(_3\)O\(_{6+x} \) in the overdoped regime (\( T_c > T_{c,\text{max}} \)). Hereafter, the samples are labeled as: UD54 (\( x=0.55, \ T_c=54 \) K), UD61 (\( x=0.6 - T_c=61 \) K), UD64 (\( x=0.6 - T_c=64 \) K), UD68 (\( x=0.75 - T_c=68 \) K), OD75 (\( x=1 - T_c=75 \) K). Polarized neutron data have been obtained on the triple axis spectrometer 4F1 at the Orphéée reactor (Saclay) (see Ref. [27] for the description of the experimental set up).

In contrast to the DDW phase or the flux phase, the CC-phase does not break translation invariance and the magnetic scattering intensity appears at the same wave vector as the nuclear scattering intensity. For the CC-phase \( \Theta_{II} \), both intensities are superimposed since interference terms with the nuclear structure factor cancel out due the existence of magnetic domains. The use of polarized neutron is then essential to identify the magnetic scattering generated by the circulating currents. Most of the data of Ref. [27] were obtained in a scattering plane \((010)/(001)\) (Fig. 4). In order to evidence small magnetic intensity, measurements have to be performed on the weakest nuclear Bragg peaks where the magnetic scattering is expected: the Bragg peak \( q=(0,1,1) \) offers the best compromise. So far, only the polarized neutron data in the normal state are reliable. The effects specific to the superconducting state such as the evolution of the magnetic response associated with the circulating current through \( T_c \) remains to be settled carefully.

Figure 2.a shows the raw neutron intensity measured at \( q=(0,1,1) \) for SF and NSF channel with \( P//q \) (sample UD64), for which the magnetic scattering is entirely spin-flip and given by Eq. 4. When cooling down from room temperature, the NSF and SF intensities display the same evolution within error bars, until \( T<T_{mag} \), where the SF intensity increases noticeably, whereas the SF intensity remains flat. This behavior signals the presence of a spontaneous magnetic order below \( T_{mag} \). The normalized magnetic intensity \( I_{mag} \) (see Ref. [27]) is \( \sim 1 \) mbarn (Fig 3a), i.e. \(~\)
YBa of the low temperature signal. Moreover, the ob-
unambiguously demonstrates the magnetic origin
signal follows the expected selection rule. That
shown in Fig.4.a-c, indicates that the observed
rule given by Eq.3. The polarization analysis,
netic, it should follow the polarization selection
pseudo-gap phase [4] (Fig. 3.b).

gap temperature, T
ing doping and matches quite well the pseudo-
the hole doping
duced
The unusual magnetic order in a tempera-
ture and doping range that cover the range
where the pseudo-gap state is observed in
(Y, Ca)Ba2Cu3O6+x. For the 4 underdoped sam-
ple, the full normalized magnetic intensity sys-
tematically increases below a certain temperature
Tmag whereas no magnetic signal is observed in
the overdoped sample (OD75) (Fig. 3). The de-

erved magnetic peak at q=(0,1,1) is resolution
limited along the (001) direction, showing that
the magnetic order is characterized by long range
3D correlations (at least at T=75 K)[27]. The
polarized neutron data of Ref. [27] therefore pro-
vide compelling evidences of the existence of a
magnetic order hidden in the pseudo-gap state of
high-Tc cuprate superconductors. But can this
magnetic order be ascribed to the Θ11I-phase?

For comparison of the neutron scattering data
with what one could expect from the Θ11I-phase,
one can express the magnetic intensity either in
term of orbital magnetic moments (Ref. [27]) or
in term of the Fourier transform of the current
density: both approaches being equivalent at a
qualitative level. Here, we propose to consider
the second approach. Since the Θ11I-phase can be
viewed as an array of butterfly triangular circu-
lating currents centered on each Cu site (Fig 1.c),
the Fourier transform of the current distribution
takes the general form:

\[ j(q) \propto \sum_{G} \delta_{G} \beta(q) \]

\[ \times (\cos(\frac{H}{2})A_{10}(q)\hat{e}_x - \cos(\frac{K}{2})A_{01}(q)\hat{e}_y + \cos(\frac{H-K}{2})A_{1-1}(q)(\hat{e}_x - \hat{e}_y)) \]  

with \( \hat{e}_i \) the unitary vector in Cartesian coor-
dinates and \( G \) vectors of the Bravais lattice.
According to this expression, the neutron in-
tensity could be different from zero only for
wave vectors along \( q=\{(0,K,L),(H,0,L),(H,H,L)\} \).
Furthermore, since the current loops are stag-
gered in each Cu plaque, there is no uniform
magnetic field in the CuO2 planes, this implies
that the neutron scattering intensity vanishes for
\( q=(0,0,L) \). The magnetic signal is indeed exper-
imentally observed at \( q=(0,1,L) \) (Fig. 4) and
vanishes at \( q=(0,0,2) \) (Fig. 4a).

When the circulating currents are modeled
with a set of thin wires along which flow a cur-
rent with a given intensity, one obtains for the
\( A_{ij} \) terms in Eq. 5

\[ A_{ij}(q) \propto \sin(\pi (iH + jK))/((iH + jK)^2) \]

The terms \( A_{ij} \) become ei-
ther 1 or 0 at a Bragg position. The term \( \beta(q) \)
incorporate the dimpling of CuO planes and, for instance, does not be ruled out on pure experimental grounds. Since YBa$_2$Cu$_3$O$_{6+x}$ is a bilayer system, current loops in the 2 CuO planes could be either in phase ($\psi = 0$) or out-of-phase ($\psi = \pi/2$), so that $\beta(q) \propto \cos(\pi Ld/c + \psi)$ ($d = 3.3$ Å stands for the distance between the planes within the bilayer.) Finally, for the scattering plane (010)/(001), the neutron cross-sections, given by Eqs. 2-5, can be written for the circulating current model shown Figs. 4.a-c as

$$I_{P//\hat{q}} \propto \frac{\cos^2(\pi Ld/c + \psi)}{q^2} \quad (6)$$

$$I_{P//\hat{q} \perp} = I_{P//\hat{z}} \quad (7)$$

$$I_{P//\hat{q} \perp} = 0 \quad (8)$$

The L dependence of the magnetic Bragg intensity measured in the SF channel for $P//\hat{q}$ and $q=(01L)$ is in agreement with Eq. (6) with $\psi = 0$ (Fig. 4.c). One may speculate that this in-phase configuration could favor the tunneling of charge carriers along the c axis. However, in Fig. 4.a-c, the magnetic signal for $I_{P//\hat{q}}$ and for $I_{P//\hat{z}}$ do not simply match and a magnetic signal occurs also for $P//\hat{q} \perp$, in contrast to what is expected in Eq. (7). This discrepancy could be related to the fact that the CC picture of Figs. 4.c-d assumes perfect CuO$_2$ planes and, for instance, does not incorporate the dimpling of CuO$_2$ planes in the YBa$_2$Cu$_3$O$_{6+x}$ system. Further, spin degrees of freedom might also play a role in producing in-plane magnetic moments (necessary to account for a non-zero signal for $P//\hat{q} \perp$), by spin-orbit scattering as proposed in Ref. [28] for the CC-phase.

Considering the reported neutron data [27], the observed magnetic order is qualitatively consistent with the existence of a circulating current state proposed by C.M. Varma to account for the pseudo-gap [13,14,15,16]. The neutron data nevertheless suggest that a more realistic description of the CuO$_2$ should be included, in addition to spin degrees of freedom. It is also important to emphasize that, if, on the one hand, the neutron data support the CC-phase model, on the other hand, alternative interpretations cannot be ruled out considering the limited amount of data available.

Figure 4. Temperature dependencies of the normalized magnetic intensity, measured in the SF channel at $q=(011)$ in sample UD61: a) for $P//\hat{q}$, b) for $P//\hat{z}$, c) for $P//\hat{q} \perp$ d) Sketch of the scattering plane showing the three polarization directions discussed here. e) Sample UD54: L-dependence of the normalized magnetic intensity measured at $T = 70$ K and $q=(011)$ as compared to $\frac{\cos^2(\pi Ld/c)}{q^2}$ (solid line) and $\frac{1}{q}$ (dashed line).

At that stage, one could consider, as an example, a ferromagnetic order with magnetic Cu moment aligned perpendicular to the CuO$_2$ planes. The magnetic intensity at $q=(002)$ should vanish as well, whereas a signal should be observed at the Bragg reflection $q=(01L)$, as observed experimentally. Furthermore, the L-dependence reported in Fig. 4.c is dominated by the cosine square term, corresponding to "ferromagnetically" coupled planes. Since the studied Bragg reflections are at rather small $|q|$, one cannot distinguish between an evolution at larger vector controlled by a term $\sim \frac{1}{q^2}$ for circulating currents or by a term $\sim (1 - \frac{1}{2q^2})F_C^2(q)$ for ferromagnetic spins (for instance). However, this cannot account for the magnetic intensity observed for $P//\hat{q} \perp$ (Fig. 4.a-c). Another spin model could be a magnetic order involving staggered spins on oxygen sites which could also account for the observed magnetic scattering (see Ref. [27]). Thus, a spin order cannot be ruled out on pure experimental grounds.
The alternative spin orders have nevertheless two main drawbacks: (i) they should have typically been detected by local probe measurements such as NMR or μSR, (ii) no theoretical model has been developed, so far, suggesting this could be relevant for cuprates.

It is clear that more work is needed to determine the exact magnetic structure factor and the evolution of the magnetic intensity at larger wave vector. Further experiments will require a study of the magnetic signal with different experimental set-up: (i) by using also a polarizing mirror to analyze the final polarization (ii) by performing the 3D polarization analysis \[29\] (using CRYOPAD for instance). Another advantage of CRYOPAD is the suppression of the magnetic guide field at the sample position which allows to study the magnetic response in the superconducting state. Further, an external magnetic field could be applied since it should couple to spins, but should have a minor effect (if any) on the circulating currents.

As a conclusion, we have seen that the neutron polarized scattering is a unique technique that has been used to search for the existence of circulating currents in cuprates yielding tiny magnetic field distribution to which neutron spin can couple. While the search for flux phase and DDW phase remains unsuccessful, there are now experimental evidences of a magnetic order hidden in the pseudo-gap state of high-\(T_c\) superconducting cuprates, supporting the circulating current state proposed by C.M. Varma in his theory of the pseudo-gap phase.

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