H1. MAGNETIC ORDER IN THE PSEUDOGAP PHASE OF HIGH-TC SUPERCONDUCTORS

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In the optimally doped and underdoped regimes, high-T_c copper oxide superconductors (SC) exhibit a pseudogap state with anomalous magnetic, transport, thermo-dynamic, and optical properties below a temperature, T^*, which is large in comparison to the super-conducting transition temperature, T_c. The origin of the pseudogap is a challenging issue as it might eventually lead to identify the superconducting mechanism. Two major classes of theoretical models attempt to describe the pseudogap state: in the first case, it represents a precursor of the superconducting *d*-wave gap with preformed pairs below T^*, which would acquire phase coherence below T_c. In a second approach, the pseudogap is associated either with an ordered or with a disordered phase competing with the SC state. The order parameter associated with these competing phases may involve charge- and spin- density waves, or charge currents flowing around the CuO_2 square lattice, such as D-charge density wave (DDW) or orbital circulating currents (CC) as proposed by C. M. Varma [1].

Most of the above 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, the novel CC phases preserve the TSL as they correspond to 4 or 2 current loops per unit cell [1]. The charge currents could be identified by virtue of the pattern of ordered orbital magnetic moments pointing perpendicularly to the CuO_2 planes (*i.e.*, along c^*). 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 which corresponds to a zero magnetic propagation wavevector, $\boldsymbol{Q} = 0$, but with no net magnetization. In neutron diffraction, the magnetic intensity is superimposed on the nuclear Bragg peak, meaning that these experiments are very delicate as the magnetic intensity, proportional to the squared magnetic moment M^2, is expected to be very small as compared to the nuclear Bragg intensity. In order to detect this hidden magnetic response, polarized neutron experiments, which allow us to separate magnetic and nuclear cross sections, are therefore required.

Motivated by this theoretical work, we have studied the possibility of a magnetic order associated to these circulating current phases. Polarized neutron measurements were performed on the triple-axis spectrometer 4F1, using full polarization analysis. We have reported the first successful observation of a magnetic order in the pseudogap state of the cuprate $YBa_2Cu_3O_{6+x}$ [2]. Since this type of phase does not break the translational symmetry, the magnetic contribution occurs only on top of the nuclear Bragg peaks, and we thus measured the magnetic signal on the weakest Bragg peak: the (011) reflection provides the best compromise between the magnetic scattering amplitude and the leakage of the Bragg scattering into the spin-flip channel. For the neutron polarization $\boldsymbol{P} // \boldsymbol{Q}$, where one expects the magnetic scattering to be strongest, the spin-flip (SF) intensity increases notably at low temperature, in contrast of the non-spin-flip (NSF) intensity which is essentially flat (Fig. 1). To assess the reproducibility of our observation, we have studied a large variety of samples from the underdoped part of the phase diagram of the high-T_c compound family $YBa_2Cu_3O_{6+x}$ with T_c ranging from to 54 K in underdoped samples to 75 K in an overdoped sample.

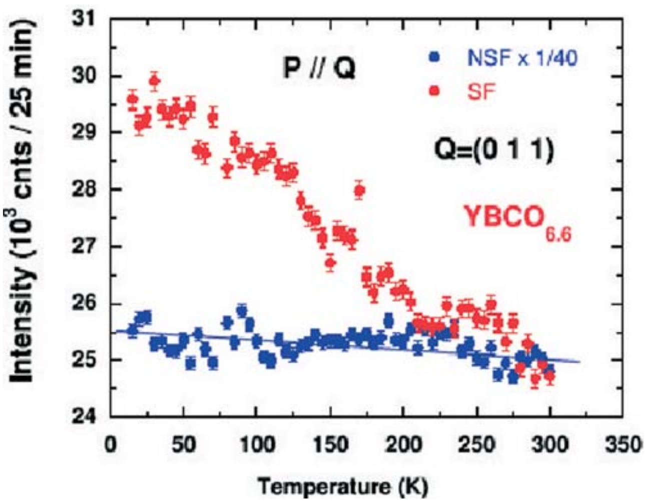

Figure 1. Raw polarized-neutron intensity : the full red points show the spin-flip (SF) scattering and the blue ones the non-spin-flip (NSF) scattering. A signal occurs below ⊕220 K only in the SF channel.

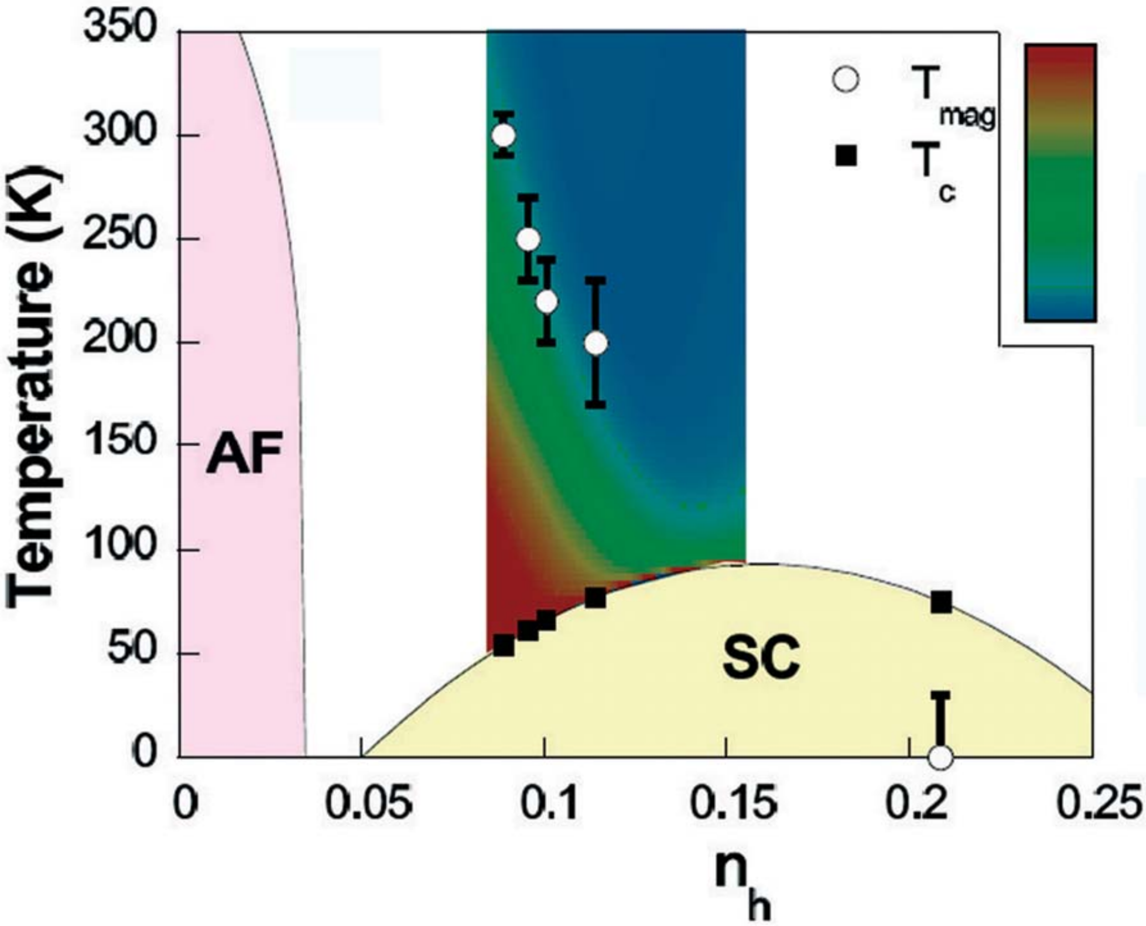

Figure 2. High-T_c phase diagram as a function of the hole doping. The coloured area (blue=0) shows the pseudogap phase as measured by resistivity. T_{mag} is the temperature where the magnetic order occurs.

The results demonstrate the appearance of a magnetic scattering in the SF channel below a temperature T_{mag}, that matches quite well the pseudogap temperature T^* as defined by the resistivity measurement (Fig. 2). The magnitude of the observed signal changes with the neutron polarization as is expected for a magnetic intensity. This definitively rules out any experimental artefact.

The typical cross section of the magnetic order is $\oplus 10^{-4}$ of the strongest Bragg peaks. This explains why such a magnetic order was not detected before using unpolarized neutron diffraction. Using the observed magnetic cross section and assuming a weakly momentum-dependent form factor, one can deduce a typical magnitude for the ordered magnetic moment of about 0.05 to 0.1 μ_B, with a moment decreasing with increasing doping. This is about the magnitude expected from current loops in the CC phase. However, such an orbital moment is expected to lie perpendicular to the CuO_2 planes, whereas, in our case, the moments are not purely along the c* axis. Combining the data for all measured polarizations in the different samples, one can estimate the mean angle between the direction of the moments (assumed to be collinear) and the *c** axis to be _ = 45° ± 20°, valid for all samples. In summary, we have reported the first signature of an unusual magnetic order in several $YBa_2Cu_3O_{6+x}$ samples matching the pseudogap behaviour in underdoped high-Tc cuprates [2]. Such an observation points towards the existence of a hidden order parameter for the pseudogap phase in high-Tc superconductors. Importantly, our experiment reveals a 3D long-range order which does not break the translational symmetry of the lattice and, therefore, implies a decoration of the unit cell with staggered spin or orbital moments. The symmetry of the observed order corresponds to that expected for orbital moments associated with a circulating current state [1].

[1] C.M. Varma, Phys. Rev. B 55, 14554 (1997); *ibid.* **73**, 155113 (2006).
[2] B. Fauqué et al, Phys. Rev. Lett. **96**, 197001 (2006).