

## UNUSUAL MAGNETIC ORDER IN THE PSEUDOGAP REGION OF THE SUPERCONDUCTOR $\text{HgBa}_2\text{CuO}_{4+\delta}$

Y. Li<sup>1</sup>, V. Balédent<sup>2</sup>, N. Barisic<sup>1</sup>, Y. Cho<sup>1</sup>, B. Fauqué<sup>2</sup>, Y. Sidis<sup>2</sup>, G. Yu<sup>1</sup>, X. Zhao<sup>1</sup>,  
P. Bourges<sup>2</sup>, M. Greven<sup>1</sup>

<sup>1</sup> Department of Applied Physics, Stanford University, Stanford, California 94305, USA

<sup>2</sup> Laboratoire Léon Brillouin, CEA-CNRS, CEA-Saclay, 91191 Gif-sur-Yvette, France

The pseudogap region of the phase diagram is an important unsolved puzzle in the field of high-transition-temperature ( $T_c$ ) superconductivity, characterized by anomalous physical properties below a certain temperature,  $T^*$ [1]. In contrast to the superconducting temperature  $T_c$  which exhibits a dome-like shape, the pseudogap phase is observed only at low doping in the underdoped region of the cuprates phase diagram. There are open questions about the number of distinct phases and the possible presence of a quantum-critical point underneath the superconducting dome. In particular, it has been proposed [1,2] that the pseudogap is characterized by an ‘hidden’ order breaking time reversal symmetry. In addition to this, a separate but perhaps more important issue remains to be settled in the cuprates: are fluctuations of a such ‘hidden’ order, associated with a quantum phase transition, capable of providing the pairing glue and explaining the anomalous normal-state properties?

Recent pioneering polarized neutron diffraction work [3], carried out at the LLB, has established that in underdoped  $\text{YBa}_2\text{Cu}_3\text{O}_{6+d}$  (YBCO) a new form of order sets in below  $T^*$ . This “hidden” magnetic order, presumably due to charge currents circulating around the Cu-O plaquettes[2], appears to be in competition with superconductivity and is found to disappear at higher doping.

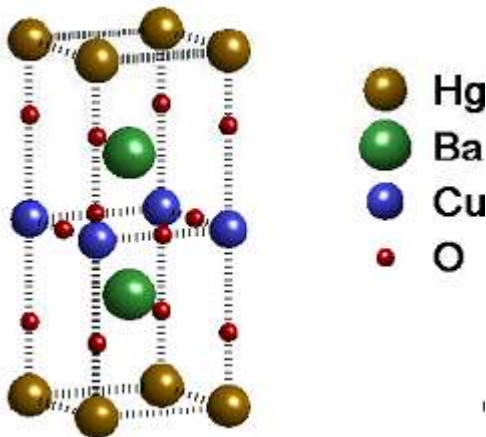


Fig. 1: Crystal structure of  $\text{HgBa}_2\text{CuO}_{4+\delta}$

While the high- $T_c$  superconductors all contain the Cu-O plane as a basic building block, they also exhibit significant structural and chemical differences. YBCO is a double Cu-O layer compound, with orthorhombic symmetry and Cu-O

chains in addition to the Cu-O planes, and a maximum  $T_c$  of about 93 K. A structurally simpler material, with higher  $T_c$ , is the Hg-based compound,  $\text{HgBa}_2\text{CuO}_{4+\delta}$  (Hg1201) (Figure 1): it contains only one Cu-O layer per unit cell, has a tetragonal crystal structure, a wide spacing between the Cu-O planes, and a very high  $T_c$  of 98 K at optimal doping. Quite generally, the Hg-based superconductors are known to have the highest values of  $T_c$  among single-layer, double-layer, etc. cuprates.

We have used polarized neutron diffraction on the triple axis spectrometer 4F1 at the ORPHÉE reactor of the Laboratoire Léon Brillouin [4] to demonstrate in a series of underdoped crystals for the model superconductor Hg1201 that the characteristic temperature  $T^*$  marks the onset of an unusual magnetic order. This was possible by the recent success in growing gram-sized Hg1201 single crystals at Stanford University [4] and the high polarization efficiency of the spectrometer.

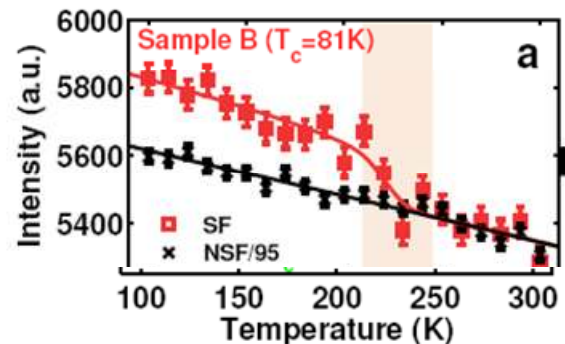


Fig. 2: Spin-flip (SF) and non-spin-flip (NSF) intensity of Bragg peak (101) in one underdoped sample of Hg1201 (from [4]). A magnetic signal is observed below 250 K.

Figure 2 demonstrates the existence of a magnetic component in the spin-flip (SF) geometry for an underdoped sample. Due to the relatively strong intensity from unavoidable nuclear Bragg peak leakage in the SF geometry, the measurement is done at a weak nuclear reflection  $\mathbf{Q}=(101)$ . The neutron polarization was parallel to the momentum transfer,  $\mathbf{P} \parallel \mathbf{Q}$ , a geometry in which all magnetic scattering occurs in the SF channel. The linear slope of the nuclear scattering observed in the non-spin-flip channel can be accounted for by the Debye-Waller factor.

As expected, due to non-zero leakage in the SF geometry, the SF data exhibit a linear nuclear scattering contribution as well. However, the SF data furthermore exhibit an additional component below  $T_{\text{mag}} \sim 250$  K, which we conclude to be of magnetic origin [4]

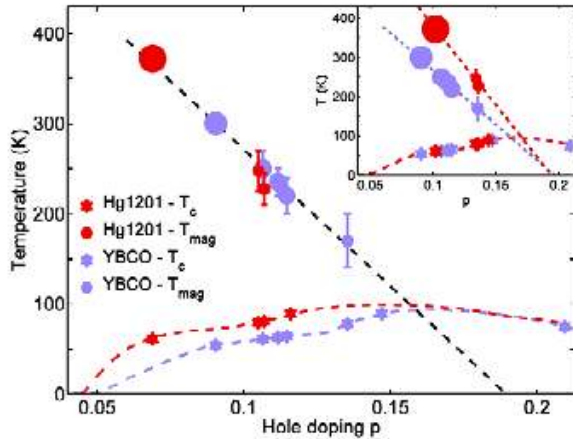


Fig.3: Universal pseudogap phase diagram of Hg1201 and YBCO (from [4]). The temperature  $T_{\text{mag}}$  extracted from the neutron data [2,4] reproduces the doping behaviour of  $T^*$ . The combined data extrapolate to  $T_{\text{mag}} = 0$  K at  $p_c = 0.19$ .

The onset of magnetic order in YBCO has been associated with the pseudogap temperature  $T^*$ , where  $T^*$  is the characteristic temperature obtained from resistivity measurements. Below  $T^*$ , the resistivity is sub linear with temperature. Interestingly, that deviation from linear resistivity scales with the magnetic intensity. Resistivity data for a separate small crystal of Hg1201 with the same  $T_c$  (81 K) follow quite strongly the same trends, suggesting that the observed magnetic and charge properties share the same physical origin.

Together with the recent results for YBCO[2], this observation constitutes a demonstration of the universal existence of such a state as it is shown in the figure 2 [4]. (i) In both cases, the order preserves the translational symmetry of the underlying lattice, unlike conventional antiferromagnetism, which occurs at  $(1/2, 1/2, 0)$  and equivalent reflections; (ii) The magnetic scattering develops below a temperature which coincides with the pseudogap temperature  $T^*$  determined from transport measurements, suggesting that the novel order involves both magnetic and charge degrees of freedom; (iii) The magnetic signal is of comparable strength for the two compounds, it is strongest in very underdoped samples, and the transition appears to be continuous.

The findings appear to rule out theories that regard  $T^*$  as a crossover temperature rather than a phase transition temperature. Instead, the experiments for Hg1201 and YBCO are qualitatively consistent with a true magnetic phase transition, suggesting a novel

state of matter with broken time-reversal symmetry having the symmetry of two counter-circulating charge current loops per  $\text{CuO}_2$  plaquette [3] (Figure 4, left). More specifically, the results suggest a variant of the previously proposed charge current-loop order that involves apical oxygen orbitals [5] (figure 4, right).

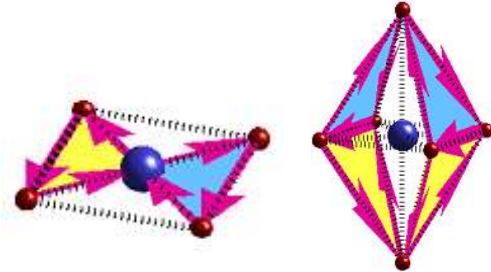


Fig. 4: Simplified schematic illustrations of two circulating current ordered states that break time-reversal symmetry, but preserve translational symmetry. Left) Planar circulating currents phase originally proposed by C.M. Varma [3]. Right) circulating currents involving apical oxygen [5].

The maximum  $T_c$  occurs close to where the experiment fails to discern a magnetic signal. It appears likely that the order competes with the superconductivity with the notion that many of the unusual properties arise from the presence of a quantum-critical point [1,3] at  $p_c = 0.19$  (figure 3). One intriguing possibility is that the fluctuations associated with an underlying quantum critical point are directly responsible for the appearance of superconductivity and the unusual normal state properties, such as the linear resistivity found up to remarkably high temperatures.

#### References:

- [1] M.R. Norman, D. Pines, C. Kallin, Adv. Phys. 54, 71, (2005).
- [2] B. Fauqué *et al*, Phys. Rev. Lett. 96, 197001 (2006).
- [3] C.M. Varma, Phys. Rev. B 73, 155113 (2006).
- [4] Y. Li *et al* Nature, 455, 372 (2008).
- [5] C. Weber *et al*, Phys. Rev. Lett. 102, 017005 (2009).

Contact: [philippe.bourges@cea.fr](mailto:philippe.bourges@cea.fr)