

METALLIC STRIPES IN OXYGEN DOPED La_2CuO_4

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We have investigated the charge ordering in an oxygen doped La_2CuO_4 crystal by diffuse high-resolution x-ray diffraction using synchrotron radiation. The micro-strain ϵ in the CuO_2 plane is close to the micro-strain ϵ_0 for the formation of static charge ordered phases. The crystal shows the co-existence of a first phase with an insulating crystal of charge strings and a second superconducting phase of "superstripes". The first insulating phase is due to charges at the critical doping $1/8$ self trapped into a crystal of ordered strings of finite length (~ 60) and a commensurate period of 4 lattice units. The superconducting phase shows diffuse diffraction peaks with short coherence length due to fluctuations of stripes with an incommensurate period of about 4.8 unit cells and length longer than 500 .

1. Introduction

The superconducting phase in cuprate superconductors occurs in a superlattice of metallic quantum stripes, called "superstripes" [1-5], where the hopping between the stripes for a single particle (t_{\perp}) is smaller than for an electron pair ($t_{\perp}^2 N_0$, where N_0 is the density of states at the Fermi level). The fact that the stripes show up clearly in some high- T_c compounds and not in others has been explained recently by revealing that different compounds show very slow or very fast stripe fluctuations and therefore their detection depends on the experimental measuring time. The stripes formation and dynamics (and hence their fluctuations) has been discovered experimentally to be controlled by the elastic strain field ϵ [6-11], due to the lattice mismatch of bcc CuO_2 layer with neighbor rocksalt fcc layers. The "superstripes" appear for an elastic strain larger than a critical value ϵ_c . High- T_c superconductivity shows up around this critical point in a region of micro strain $|\epsilon - \epsilon_c| / \epsilon_c < 0.5$. Static charge order appears at higher strain $\epsilon > \epsilon_0$.

As it has been shown in a recent paper [11] the $\text{La}_2\text{CuO}_{4+\delta}$ compounds, have a high value of micro-strain, therefore they show superconducting superstripes at ($\delta \sim 0.16$, $\epsilon \sim 0.072$) and static charge ordering at ($\delta = 1/8$, $\epsilon \sim 0.072$). By choosing a sample with an effective hole density of $\delta \sim 0.15$ we have found here the presence of domains made of crystals of strings at hole doping $1/8$ that co-exist with macroscopic domains where a superlattice of superstripes appears. Hence we were able to investigate the transformation of an insulating electronic crystal of ordered localized charges into a metallic striped phase. This allowed us to study the distribution of the lengths of the short static strings, very picked at $m = 11\mathbf{a}$ (where \mathbf{a} is the axis of the unit cell), and of the metallic superstripes, more broadened, with lengths going from about $m = 20\mathbf{a}$ to "infinite" ($\mathbf{a}^* \sim 0$) with an average length of $m = 77\mathbf{a}$ (more than 400\AA).

2. Experimental

A flux grown single crystal of $\text{La}_2\text{CuO}_{4+y}$ was doped by electrochemical oxidation. The crystal shows a spinodal macroscopic decomposition into two domains with about equal probability: phase (1) made of macroscopic domains with static antiferromagnetic order below 40 K [12] and a second phase (2) made of superconducting domains with a single superconducting transition $T_c=40$ K, where the hole density is the same as in the optimum Sr doped La_2CuO_4 superconductors with $\delta=0.16$ holes per Cu site [13, 14].

The charge ordering has been studied by temperature dependent diffraction data collected on the crystallography beam-line at the synchrotron radiation facility Elettra at Trieste [15-17]. The X-ray beam emitted by the wiggler source at the 2 GeV electron storage ring was monochromatized by a Si(111) double crystal monochromator. The temperature of the crystal was monitored with an accuracy of ± 1 K. The data was collected with a photon energy of 12.4 KeV ($\lambda=1 \text{ \AA}$), using an imaging plate as a 2D detector. The sample oscillation around the **b** axis was in a range $0 < \phi < 30^\circ$, where ϕ is the angle between the direction of the photon beam and the **a** axis. We have investigated a portion of the reciprocal space up to 0.6 \AA^{-1} momentum transfer, i.e., recording the diffraction spots up to the maximum indexes 3, 3, 19 in the **a***, **b***, **c*** direction respectively.

3. Results and Discussion

Thanks to the high brilliance synchrotron radiation it has been possible to record a large number of weak superstructure spots due to charge ordering around the main peaks of the average structure. Twinning of the crystal was taken into account to index the superstructure peaks. The oxygen ordering has been found to occur in the temperature range 270-330 K, while the charge ordering develops below 190 K [16]. At $T = 100$ K we have found the competition and coexistence of two charge ordered domains.

Fig. 1 shows a section of the x-ray diffraction image (**c***,**b***) around the main peak (0,0,6), revealing global view of the superstructure peaks. The diffraction peaks due to the twinning along the **a*** direction are observable (rectangles).

For further clarification we have selected the peaks belonging to the two different domains in the two axis directions **a*** and **b*** and shown in Fig. 2 and Fig.3.

Different harmonics of the modulation (1st and 2nd) due to the first domain in the two direction **b*** and **a*** are shown in Fig.2b and 3b respectively, revealing 3D long range charge ordering, and narrow (resolution limited) diffraction peaks with wavevector

$$\mathbf{q}_1 = 0.089 (\pm 0.003) \mathbf{a}^*, 0.248 (\pm 0.002) \mathbf{b}^*, 0.495 (\pm 0.005) \mathbf{c}^*.$$

This charge ordered phase shows the formation of a crystal made of charge strings of finite length $11\mathbf{a}$, separated by $\mathbf{R}=4\mathbf{b}$ and doubling of the unit cell along the **c** axis. The profiles of the first harmonic peaks of this domain along the **b*** and **a*** directions are shown (denoted by (1)) in the Fig.4a and Fig.4b respectively. The second harmonics are shown in Fig.5. For this domain we have not been able to separate the modulation due to the oxygen ordering from charge ordering indicating that the static charge ordering has the same modulation as oxygen ordering.

In the second domain, we have been able to separate the modulation due to the 3D oxygen ordering from charge ordering, especially in the second harmonic. In Fig.3a one can distinguish the peaks due to the oxygen ordering (narrow and with a finite **a***

component) from the peaks due to the charge ordering (diffuse and with almost zero a^* component). The metallic superconducting phase shows a pattern of diffuse spots (Fig. 2a and Fig.3a) due to charge ordering with a coherence length of about 350 Å and with a wavevector:

$$\mathbf{q}_2 = 0.013 (\pm 0.003)\mathbf{a}^*, 0.208 (\pm 0.003)\mathbf{b}^*, 0.290 (\pm 0.005)\mathbf{c}^*$$

and a pattern of narrower, resolution limited, diffraction spots, with wavevector:

$$\mathbf{q}_{2\text{oxy}} = 0.037 (\pm 0.001)\mathbf{a}^*, 0.198 (\pm 0.002)\mathbf{b}^*, 0.290 (\pm 0.005)\mathbf{c}^*.$$

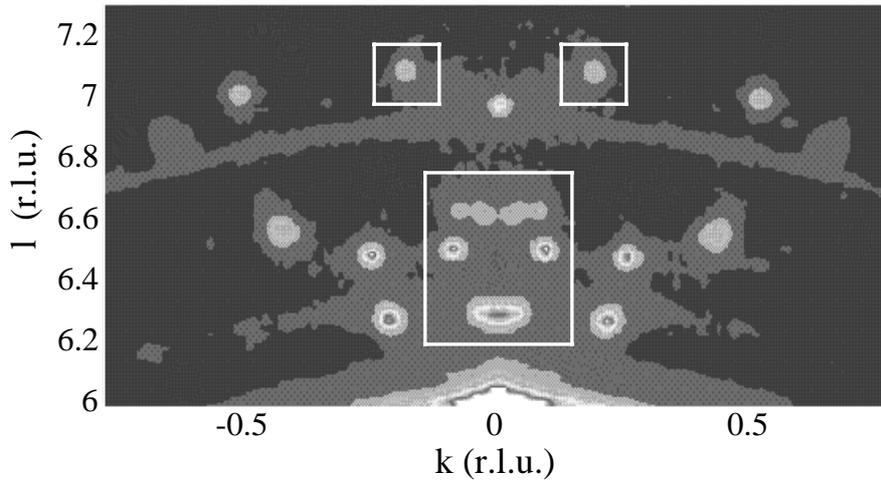


Fig.1: X-ray diffraction pattern of the $\text{La}_2\text{CuO}_{4.1}$ in the (c^*, b^*) plane by an image plate detector. The diffraction peaks along the a^* direction due to the twinning are also observable (enclosed rectangles).

The third harmonic of the \mathbf{q}_2 modulation is also observable (Fig.2a) showing an added evidence of the anharmonicity. This second wavevector \mathbf{q}_2 is associated with in plane superconducting stripe ordering. The length of the stripes in the a^* direction becomes longer than $77\mathbf{a}$ (~ 400 Å), of the same order of magnitude of the coherence length of the modulation. The transverse modulation of the superlattice of stripes along the \mathbf{b} -axis is the same as the one found in superconducting Bi2212, and characterized by diagonal stripes with wavevector of $0.208\mathbf{b}^*$ indicating a separation between the stripes of $R \sim 4.8\mathbf{b}$. The period along the c axis direction is found to be $3.5\mathbf{c}$.

The profiles of the first harmonics of this domain along the b^* and a^* directions are shown (denoted by (2)) in Fig.4a and Fig.4b respectively. In Fig.5 we show the second harmonics where the oxygen modulation, with long coherence length $1/\Gamma = 101\mathbf{a}$ and the charge modulation, with short coherence length $1/\Gamma = 44.5\mathbf{a}$ could be separated.

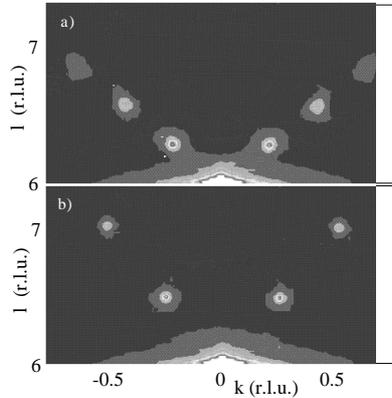


Fig.2: modulation peaks along the b^* direction of the two domains: insulating (panel b) and superconducting (panel a).

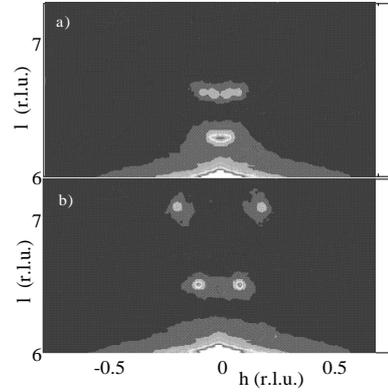


Fig.3: modulation peaks along the a^* direction of the two domains: insulating (panel b) and superconducting (panel a).

In summary this work provides an experimental evidence for the coexistence of two different phases with different hole densities in the same sample ($\text{La}_2\text{CuO}_{4.1}$) showing the coexistence in the same sample of a crystal of strings at doping $1/8$ and a superlattice of superconducting stripes in the high T_c superconducting phase at higher doping, according with the new phase diagram of the doped perovskites[11]. The superconducting phase shows diffuse diffraction peaks with an incommensurate period of about $4.8b$ and stripes with a distribution of lengths broadened around the average value of about 400 \AA . On the other hand, the doped charges at the critical $1/8$ doping are self trapped into a crystal of ordered strings of finite length of about 60 \AA and a commensurate period of 4 lattice units and characterized by sharp

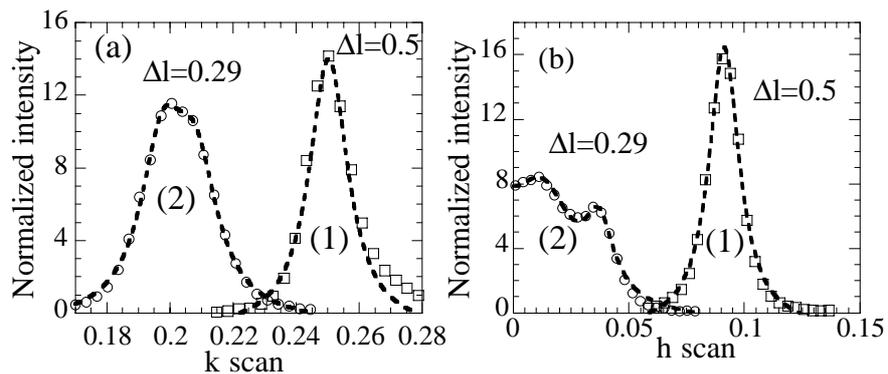


FIGURE 4. Scans along the $Q = (0, k, 6 + \Delta l)$ (panel a) and along the $Q = (h, 0, 6 + \Delta l)$ (panel b) due to the diffuse scattering peaks of stage 3.5 superstructure (2), (open circles), and the resolution limited peaks due to the stage 2 superstructure (1) open squares).

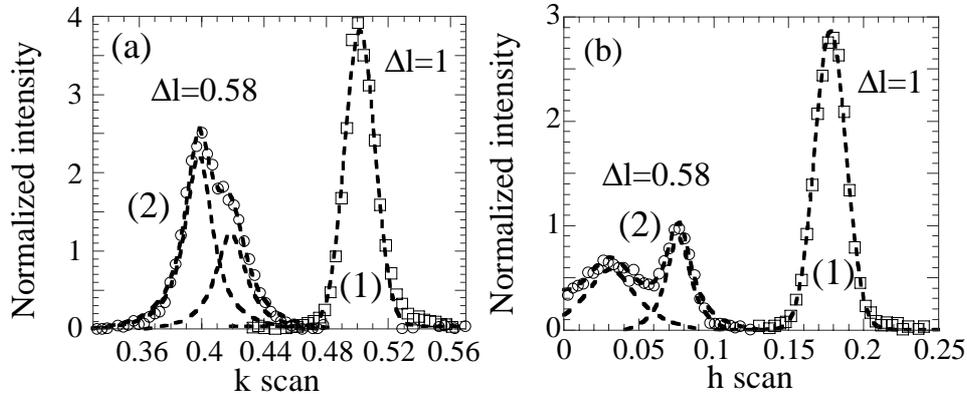


FIGURE 5. Scans along the second harmonic $Q = (0, k, 6 + \Delta l)$ (panel a) and along the $Q = (h, 0, 6 + \Delta l)$ (panel b) due to the diffuse scattering peaks of stage 3.5 superstructure (2) (open circles), and the resolution limited peaks due to the stage 2 superstructure (1) (open squares).

diffraction peaks. In conclusion the present results show coexistence of strings and stripes in the oxygen doped La_2CuO_4 system.

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