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LOCAL STRUCTURAL FLUCTUATION IN THE SUPERCONDUCTING STATE OF $La_{2-x}Sr_xCuO_4$

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The local and dynamic structural fluctuation in the superconducting state was found in $La_{1.88}Sr_{0.12}CuO_4$ by transmission electron microscopy. Concretely, the local structural change is detected as a dynamic fluctuation of the local *Pccn*/LTT region in the interior of the LTO domain. The simple discussion indicates that the dynamic fluctuation does not favor superconductivity and results in the slight lowering of Tc.

Key words: $La_{1.88}Sr_{0.12}CuO_4$, transmission electron microscopy, dynamical structural fluctuation, superconducting sate,

I. INTRODUCTION

In $La_{2-x-y}Sr_xNd_yCuO_4$ exhibiting superconductivity, there are low-temperature structural transitions from a low-temperature orthorhombic (LTO) phase to a *Pccn* one, then to a low-temperature tetragonal (LTT) one. The low-temperature transitions are characterized by a deviation of the tilt axis of the CuO_6 octahedron from the <110> direction and accompany a change in spontaneous strain[1-4]. As for physical properties, the LTO phase is superconductive, while the appearance of the *Pccn* and LTT phases results in the large lowering of Tc[4,5]. We so far examined microstructures related to the low-temperature transitions in $La_{1.5}Sr_{0.1}Nd_{0.4}CuO_4$ by transmission electron microscopy. It was then found that, when the temperature is lowered, the *Pccn*/LTT phases are first nucleated only along the twin boundary and then appear with the spotty-shape in the interior of the LTO domain. An interesting feature of the transition is that the microstructure in the large supercooling state is a complex mixture of the LTO and *Pccn*/LTT regions with a dynamic nature[6,7]. This mixture is attributed to the fact that the tilt of the octahedron as an order parameter is not locally coupled to an appropriate spontaneous strain.

The slight lowering of Tc was found in $La_{2-x}Sr_xCuO_4$ with no Nd-substitution, where the low-temperature structural transition has not been reported. On the basis of the above-mentioned facts, dynamic and local structural fluctuation related to the transition can be expected in the LTO superconducting state with the slight Tc-lowering. Then we examined in-situ observation of the local structural fluctuation in the superconducting state of $La_{1.88}Sr_{0.12}CuO_4$ with Tc of about 20K. In this paper, we describe experimental data obtained by the in-situ observation and simply discuss a correlation between the fluctuation and superconductivity.

II. EXPERIMENTAL PROCEDURE

The details of sample preparation were mentioned in our previous paper[6,7]. In-situ observation of the dynamic structural fluctuation in the LTO superconducting state was performed by taking electron diffraction patterns, and bright- and dark-field images, using a transmission electron microscope with a 5um objective aperture and a cooling stage equipped with the liquid He reservoir. In addition, we also use imaging plates as a recording medium in order to avoid a drift of a specimen during exposure. In the present experiment, particularly we took dark field images by using a 100-type spot, which is forbidden for the LTO structure. Because both *Pccn* and LTT structures give rise to the 100-type forbidden spot, these structures can not be distinguished in electron diffraction. We then write the *Pccn*/LTT structure without distinction in this paper.

III. RESULTS AND DISCUSSIONS

$La_{1.88}Sr_{0.12}CuO_4$ was suggested to undergo the LTO to *Pccn*/LTT low-temperature structural transition about 135K from the appearance of the 100-type forbidden spot. Figures 1(a) and 1(b) show an electron diffraction pattern of $La_{1.88}Sr_{0.12}CuO_4$ at 85K and a corresponding dark-field image taken by the 100 forbidden spot, respectively. The electron incidence is parallel to the [001] direction. In the pattern, Fig. 1(a), there are 100-type forbidden spots indicated by an arrow A and 1/2 1/2 0-type superlattice spots by an arrow B in reciprocal space, in addition to fundamental spots due to the LTO structure. In the dark-field image, Fig. 1(b), a bright-line contrast is seen in the dark contrast region. From the comparison with bright field images taken at about 200K in the LTO phase, the bright-line contrast region exist along a twin boundary between two neighboring LTO domains. Because of the 100 dark-field images, the bright-line contrast region is understood to be due to the *Pccn*/LTT phase, which is nucleated along the twin boundary.

In order to examine the evolution of the microstructure, the specimen was cooled down to 12K.

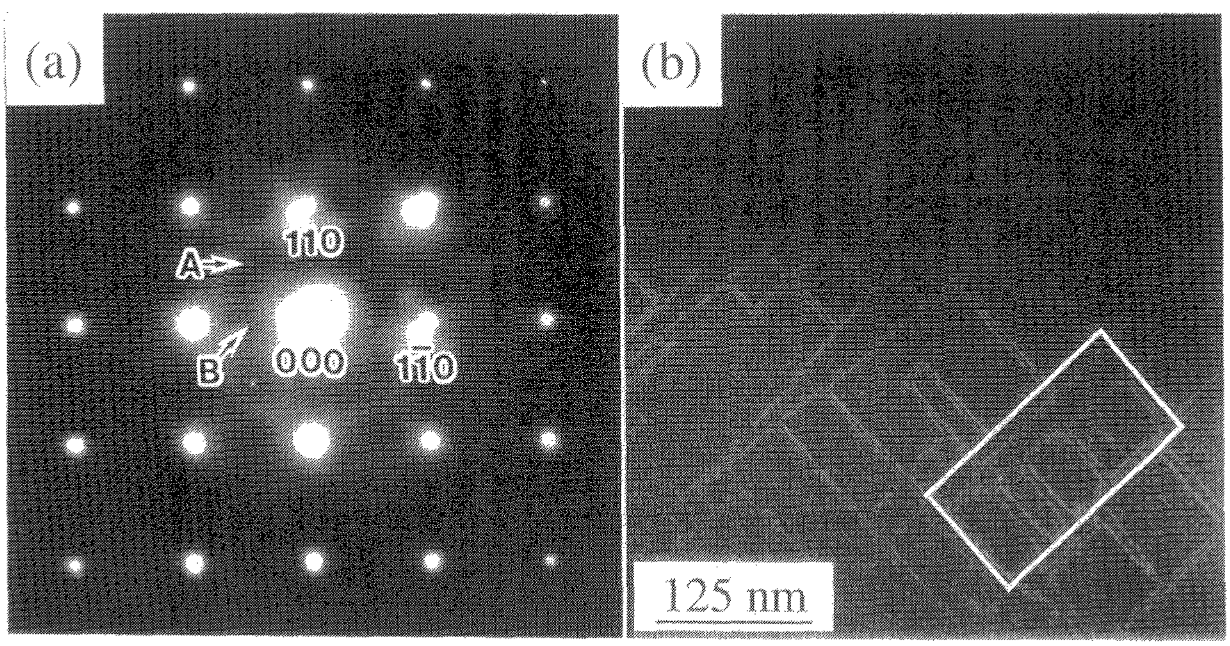

Fig. 1 Electron diffraction pattern of $La_{1.88}Sr_{0.12}CuO_4$ at 85 K, together with a corresponding 100 dark field image.

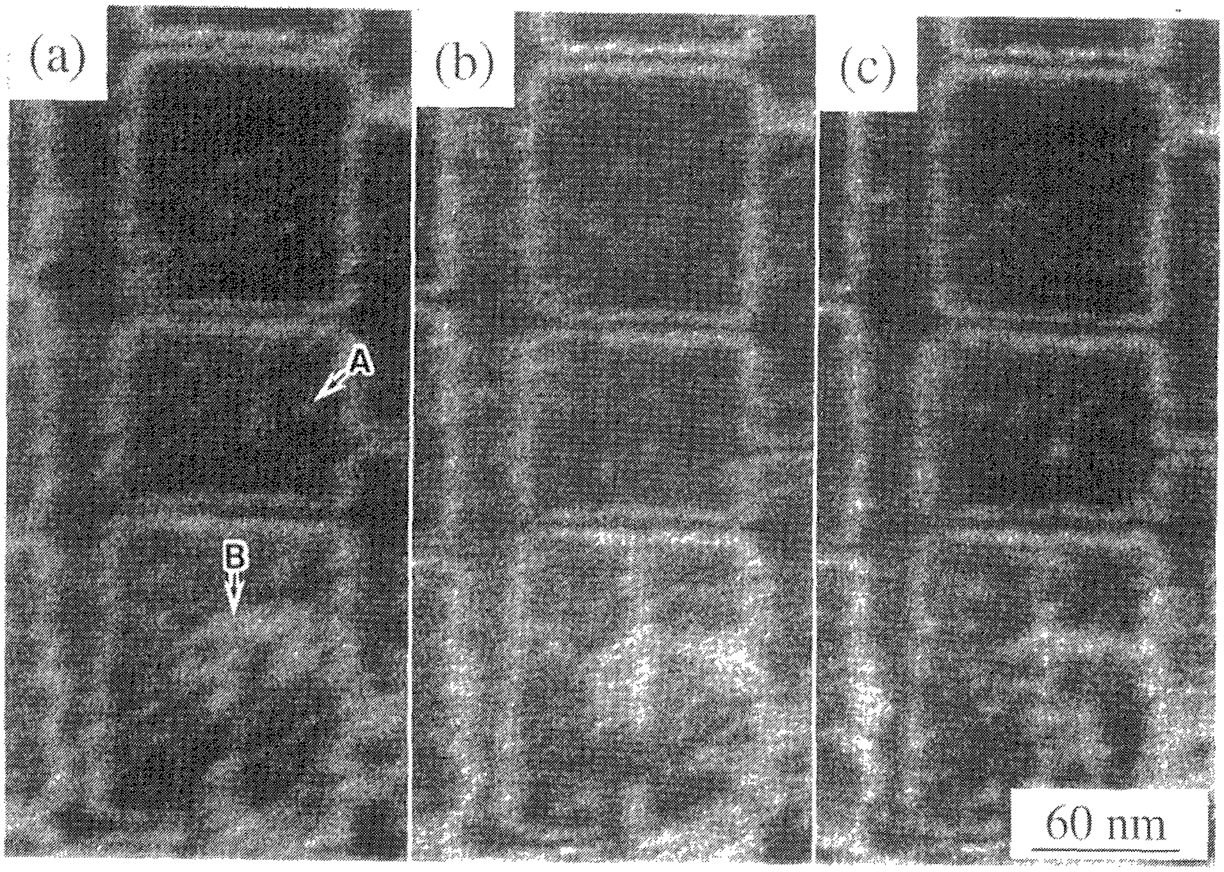

Fig. 2 A series of 100 dark field images of $La_{1.88}Sr_{0.12}CuO_4$ when the specimen was aged at 12K for 2342 sec., 2363 sec., and 2385 sec., respectively.

We first took 100-forbidden dark field images in an exposure time of about 30 sec. The images exhibit only the bright-line contrast along the twin boundary. That is, the features of the images at 12K are the same as those at 85K. Then we took images in a very short exposure time. Figures 2(a), 2(b), and 2(c) show a series of 100 dark field images when the specimen was aged at 12K for 2342 sec., 2363 sec., and 2385 sec., respectively. The exposure time of each image is about 1.5 sec.. In contrast to the images with the exposure time of about 30 sec., there are *Pccn*/LTT regions with a spotty-shape in the interior of the LTO domains in these images. An average size of the *Pccn*/LTT region is about 10 nm, as indicated by an arrow A, and the regions are dynamically nucleated and annihilated in the LTO domain. It is also found that the region with a relatively large size B continuously changes its shape. In other words, the total volume of the *Pccn*/LTT region with the spotty shape is understood to keep constant during the observation at 12K. As expected based on our previous work on the low-temperature structural transition[7], local and dynamic structural fluctuation actually occurs in the superconducting state of $La_{1.88}Sr_{0.12}CuO_4$.

In the present work, the local structural change in the superconducting state could be detected as dynamic fluctuation of the *Pccn*/LTT region with the spotty shape. The fluctuation does not accompany an increase in the total volume fraction, so that the fluctuated state seems to be a kind of the dynamic equilibrium state. Here we simply discuss a correlation between superconductivity and the dynamical structural fluctuation.

The Sr-content dependence of Tc was measured by Momono *et al*[8]. According to their data, Tc exhibits a small dip around x=0.115, which indicates a weak suppression of Tc. The Sr content of the specimen used in the present work is x=0.12 within this content range. In addition, the present work indicates that the local *Pccn*/LTT regions with the spotty shape are structurally fluctuated with time and space. Because the appearance of the *Pccn*/LTT phase results in the lowering of Tc[5], the dynamic *Pccn*/LTT fluctuation should also lead to the weak suppression of Tc. Note that the dynamic fluctuation occurs only in the vicinity of x=0.115, but the *Pccn*/LTT line phase nucleated along the twin boundary is observed in the Sr content range exhibiting no suppression of Tc as well as around x=0.115[8]. That is, the line phase is basically out of relation to physical properties such as superconductivity.

IV. REFERENCES

1. A. R. Moodenbaugh, Y. Xu, M. Suenaga, T. J. Folkerts, and R. N. Shelton, *Phys. Rev.* **B 38**, 4596 (1988).

2. J. D. Axe, A. H. Moudden, D. Hohlwein, D. E. Cox, K. M.Mohanty, A. R. Moodenbaugh, and Y. Xu, *Phys. Rev. Lett.* **62**, 2751 (1989).

3. T. Suzuki and T. Fujita, *J. Phys. Soc. Jpn.* **58**, 1883 (1989).

4. M. K. Crawford, R. L. Harlow, E. M. McCarron, W. E. Farneth, J. D. Axe, H. Chou, and Q. Huang, *Phys. Rev.* **B 44**, 7749 (1991).

5. W. E. Pickett, R. E. Cohen, and K. Krakauer, *Phys. Rev. Lett.* **67**, 228 (1991)

6. Y.Inoue, Y.Horibe, and Y. Koyama, *Phys. Rev.* **B56**, 14176 (1997)

7. Y. Horibe, Y. Inoue, and Y. Koyama, *J. Superconductivity* **10**, 461, (1997)

8. N. Momono, M. Ido, T. Nakano, M. Oda, Y. Okajima, and K. Yamaya, *Physica* **C233**, 395, (1994)

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