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Investigation of extended superconducting dome in protect-annealed $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$

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1 Introduction

Electron-doped cuprate superconductors are usually characterized by the *T'* structure (Fig.1 (b) [1]), a much narrower superconducting (SC) dome, and the more robust antiferromagnetic (AF) phase in the phase diagram than their hole-doped counterparts (Fig. 1 (d) [2]). Unlike the *T* structure of the hole-doped $La_{2-x}Sr_xCuO_4$ (LSCO) (Fig.1 (a) [1]), in the *T'* structure, copper atoms are surrounded by four oxygen atoms in the square-planar coordination. Nevertheless, in real systems, some impurity oxygen atoms remain at the apical sites forming a structure shown in Fig. 1 (c). The apical oxygen atoms induce high disorder and are harmful to superconductivity. This leads to insulating as-grown samples, and superconductivity emerges only after annealing [3], which is believed to remove the apical oxygen atoms [2].

The SC state in single crystals of $Pr_{2-y-x}La_yCe_xCuO_4$ with relatively high T_c can be realized for low Ce concentration [4]. Recently, Adachi *et al.* successfully synthesized bulk SC single crystals of $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$ (PLCCO) with x = 0.05, 0.10, and 0.15 ($T_c \sim 27$ K, Fig. 2 (d), pink circles) by utilizing an improved novel “protect annealing” method [5,6]. From angle-resolved photoemission spectroscopy (ARPES) studies of PLCCO after protect annealing, we found that the AF correlation of x = 0.10 samples can be strongly suppressed and the electron content estimated from Fermi surface area can significantly deviate from the Ce doping level (Fig. 1 (e) [6]). This has also been confirmed by the recent study targeting on conventionally annealed $Pr_{1-x}LaCe_xCuO_{4-\delta}$ [7]. Although the oxygen reduction during annealing may induce additional electrons into the system, the generally accepted phase diagram of electron-doped cuprates regards the nominal Ce doping level as the electron concentration. Therefore, the SC dome of electron-doped cuprates may not be that narrow as we see in Fig. 1 (d). In order to systematically investigate the possibly extended SC dome of PLCCO after protect annealing, we have performed ARPES measurements on single crystals of PLCCO with different Ce doping levels and annealing conditions to estimate the electron concentration.

2 Experiment

ARPES measurements were performed at PF BL-28A and UVSOR BL5U with the total energy resolution of ~30 meV. The incident light was circular polarized with 55 eV photon energy and linearly polarized (s) with 60 eV photon energy respectively. The samples were cleaved and measured *in situ* at temperature ≤ 10 K and pressure $\leq 1.5 \times 10^{-10}$ Torr.

Single crystals of $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$ with T_c varying from 28 K to 16 K were synthesized by the traveling-solvent floating-zone method. The basic annealing steps consist of protect annealing, low-temperature annealing, and dynamic annealing. The dynamic annealing separated the annealing processes into a few stages providing time for oxygen atoms to travel from the bulk to the surface,

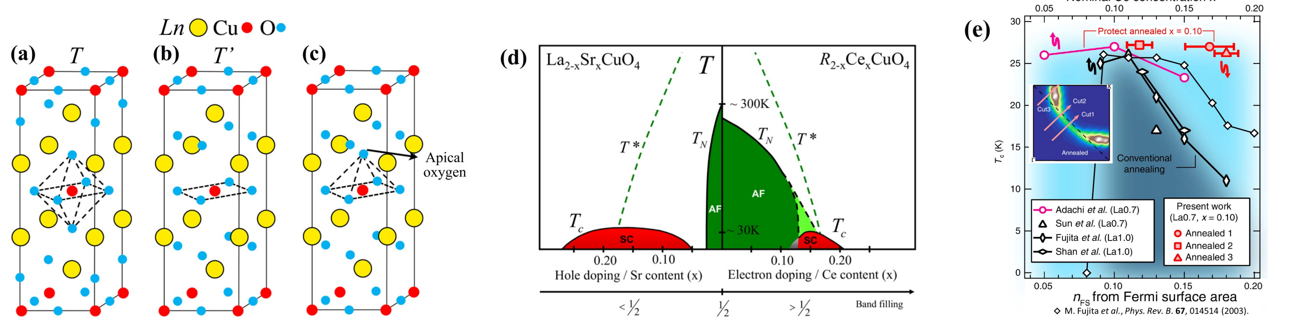

Fig. 1. **(a-b)** *T* and *T'*-type structures [1]. **(c)** *T'*-type structure with the apical oxygen atom. **(d)** Generally accepted phase diagram of hole-doped (left) and electron-doped (right) cuprates, where *R* represents the rare earth elements [2]. **(e)** Fermi surface (inset) and new phase diagram of PLCCO after protect annealing plotted as T_c *vs* n_{FS} [6].

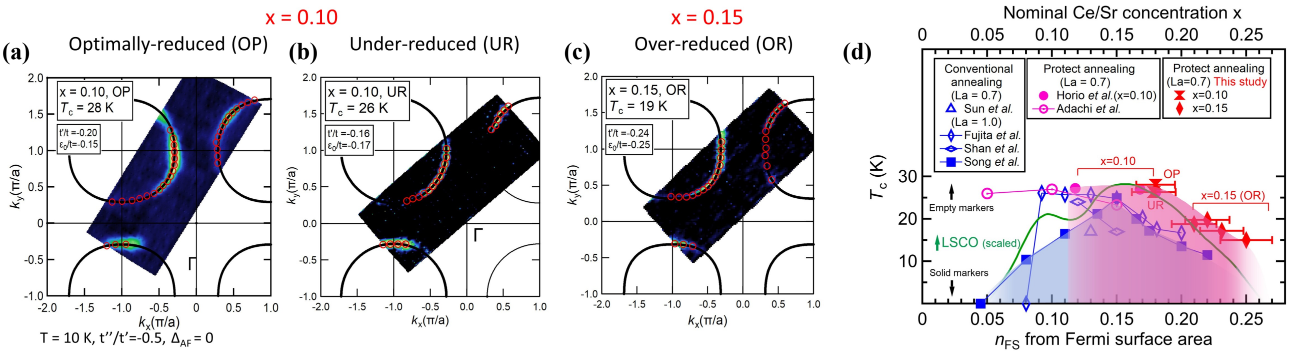

Fig. 2. **(a-c)** Fermi surface intensity plot integrated within ±10 meV around E_F and tight-binding fitting results for x_{Ce} = 0.10 and 0.15 with different reduction conditions. Sample T_c and tight-binding fitting parameters were also indicated. **(d)** Extended SC dome in the new phase diagram of PLCCO (magenta shaded area). Solid markers were plotted as T_c *vs* n_{FS} (electron number estimated from Fermi surfaces) which compose the new phase diagram. Empty markers [6,7,9] were plotted as traditional way: T_c *vs* x_{Ce}. Green curve comes from the data of hole-doped LSCO [12,13] and was plotted against x_{Sr}. In order to compare, LSCO's optimal T_c of 37 K was scaled to PLCCO's optimal T_c of 28 K in our study.

improving the oxygen homogeneity of the samples. To systematically estimate the real electron concentration a series of samples with Ce doping content x = 0.10 and 0.15 as well as different annealing conditions have been measured. We classify the crystals with the same Ce doping level by the reduction status, i.e., under-reduced (UR), optimally reduced (OP), and over-reduced (OR), which can be estimated by the annealing time and the T_c.

According to the Luttinger theorem, i.e., the number of electrons is given by the Fermi surface volume [8], one can use ARPES to accurately measure the electron concentration.

3 Results and Discussion

The Fermi surfaces determined by ARPES in Fig. 2 (a-c) indicate that there is no AF pseudogap on the entire Fermi surfaces for both x = 0.10 and 0.15, i.e., AF correlation was strongly suppressed, unlike those early studies based on the conventionally annealed samples [2] and consistent with the previous study on the strength of the same protect annealing method [6]. In order to calculate the Fermi surface area [10,11], we used the tight-binding model as the fitting function. T_c was plotted against electron content estimated from the Fermi surface area (n_{FS}) in Fig. 2 (d). In the same figure, T_c from some previous studies was also plotted against nominal Ce doping level [6,9]. The current systematic study indicates that n_{FS} can be significantly larger than the Ce doping level and thus reveals an extended SC dome in the new phase diagram of PLCCO after protect annealing (Fig. 2 (d), magenta shaded area). Thanks to the superiorities resulting from the novel protect annealing method [5,6], the SC dome in our study is even more extended than that obtained from the conventionally annealed samples (Fig. 2 (d), blue shaded area [7]). In order to compare present results with those of the hole-doped counterpart, we also reproduced the scaled data from LSCO [12,13] as a green curve in the same figure. This new phase diagram of PLCCO after protect annealing exhibits similar or even more extended SC dome than that of hole-doped LSCO, thus providing a clue for understanding the asymmetry of the phase diagram of cuprate superconductors. Further studies are in process so as to confirm the boundaries of the new phase diagram.

References

[1] Y. Krockenberger, *et al., JPSJ*. **83**, 114602 (2014).
[2] N. P. Armitage *et al., Rev. Mod. Phys*. **82**, 2421 (2010).
[3] Y. Tokura, *et al., Nature* **337**, 345 (1989).
[4] Y. Koike *et al.*, *Jpn. J. Appl. Phys*. **31**, 2721 (1992).
[5] T. Adachi *et al.*, *J. Phys. Soc. Jpn*. **82**, 063713 (2013).
[6] M. Horio *et al.*, *Nat. Commun*. **7**, 10567 (2016).
[7] D. Song *et al.*, *Phys. Rev. Lett*. **118**, 137001 (2017).
[8] J. M. Luttinger *et al., Phys. Rev*. **119**, 1153 (1960).
[9] M. Fujita *et al., Phys. Rev. B*. **67**, 014514 (2003).
[10] M. Hashimoto, PhD Thesis, University of Tokyo (2007).
[11] R. Sensarma, *et al. Phys. Rev. Lett.* **98**, 027004 (2007).
[12] H. Takagi et al., Phys. Rev. B. **40**, 2254–2261 (1989).
[13] N. Momono, et al., J. Phys. Chem. Solids. **59** (1998).

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