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Suppression of the antiferromagnetic pseudogap in the electron-doped high-temperature superconductor $Pr_{1.3-x}La_{0.7}Ce_xCuO_{4+\delta}$

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1 Introduction

In the underdoped region of the electron-doped high-temperature superconductors (e-HTSCs), a large pseudogap of order ** eV opens due to AF order or strong AF correlation. Angle-resolved photoemission (ARPES) studies have shown a pseudogap opening around the "hot spot", that is, the crossing point between the Fermi surface and the AF Brillouin zone (BZ) boundary [1]. Since the discovery of the e-HTSCs, it has been known that annealing in a reducing atmosphere plays an essential role to realize superconductivity. As-grown samples are non-superconducting antiferromangets and by annealing, the AF phase shrinks, and superconductivity appears [2]. ARPES studies revealed that annealing reduces the intensity of the AF folded bands and the spectral intensity generally increases at E_F [3,4]. Nevertheless, the pseudogap of the antiferromagnetic origin at the "hot spot" has been seen in all the e-HTSCs from underdoped to overdoped region studied so far [5]. Therefore, the AF pseudogap has been considered as a universal phenomenon and the relationship between the antiferromagnetism and superconductivity has been extensively discussed for the e-HTSCs.

In a previous study, Brinkmann *et al.* [6] annealed thin single crystals of $Pr_{2-x}Ce_xCuO_4$ (PCCO) sandwiched by PCCO polycrystals and realized superconductivity with low Ce concentrations down to 4%. Recently superconductivity was found even at zero-doping in thin films and powdered samples of e-HTSCs [7,8]. Inspired by these studies, Adachi *et al.* [9] further improved annealing method, and annealed single crystal of $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$($x$ = 0.10) with the sample surface protected by the powders of the same composition [9]. Although samples of this composition have not shown superconductivity in previous studies [10], their samples showed a T_c as high as 27.0 K. In order to clarify the changes induced by the annealing, we have performed ARPES measurements on single crystals of $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$($x$ = 0.10) with varying annealing conditions.

2 Experiment

Single crystals of $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$($x$ = 0.10) were synthesized by the floating zone method. The actual Ce content was confirmed to be x=0.100 by the inductively coupled plasma (ICP) method. Three kinds of samples were prepared; as-grown, weakly annealed, and annealed samples, among which only the annealed one showed superconductivity and the T_c was 27.0 K. The "weakly annealed sample" was annealed at 650 K for 24 hours, and the "annealed sample" was at 800 K for 24 hours.

ARPES experiment was performed at beamline 28A of Photon Factory with $h\nu$=55eV. The samples were cleaved *in situ* at T=15K in an ultra-high vacuum of 2×10^{-10} *Torr*.

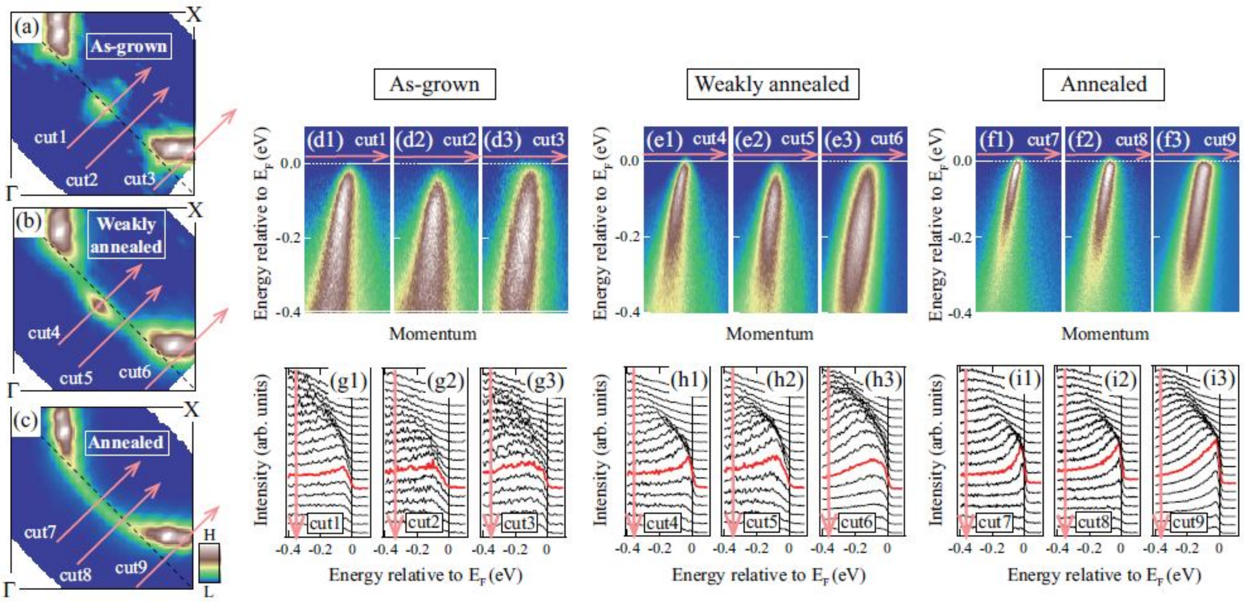

FIG. 1: ARPES spectra of $Pr_{1.2}La_{0.7}Ce_{0.1}CuO_4$. (a)-(c): Fermi surface mapping of as-grown, weakly annealed, and annealed samples, respectively. The intensity is integrated over ~10meV with respect to E_F. The suppressed intensities at the "hot spots", the crossing points between the Fermi surface and the AF Brillouin zone boundary, are fully recovered in the annealed sample. (d1)-(d3), (e1-e3), (f1)-(f3): Intensity plot in energy-momentum space for each sample. The position and the direction of the cuts are indicated in (a)-(c). (g1)-(g3), (h1)-(h3), (i1)-(i3): EDCs plotted along the cut for each sample. The AF pseudogap that is still observed in the weakly-annealed sample is suppressed in the reduction annealed sample.

3 Results and Discussions

In Fig. 1(a)-(c), Fermi surface mappings of as-grown, weakly annealed, and annealed samples are shown. For the as-grown sample, the intensity at the "hot spot" is strongly suppressed due to stabilized AFM in this sample. The intensity partially recovers by weak annealing, but nodal and anti-nodal regions are still disconnected. This means removal of apical oxygen is not enough and the influence of AF correlation still remains strongly. However, in the annealed sample, the suppressed intensity at the "hot spot" is fully recovered, and Fermi surface gets continuous on the entire momentum region. This Fermi surface is completely different from that of the previous studies on superconducting samples with low Ce concentration which looks rather similar to that of the present weakly-annealed sample. Changes induced by reduction annealing is clear also in Figs. 1(d1)-(d3), (e1)-(e3), (f1)-(f3), (g1)-(g3), (h1)-(h3), and (i1)-(i3) where the intensity plot in energy-momentum space and EDCs along the cuts through the node, "hot spot", and anti-node for each sample are shown. For the as-grown and weakly annealed samples, the peak is shifted toward higher binding energies at the "hot spot" and the antinode due to the splitting of the band caused by AF correlation. On the other hand, the annealed sample shows a clear quasi-particle peak on the entire Fermi surface, and the pseudogap is closed.

We have also measured two additional annealed samples and fitted the Fermi surface of the three annealed samples to the tight-binding model:

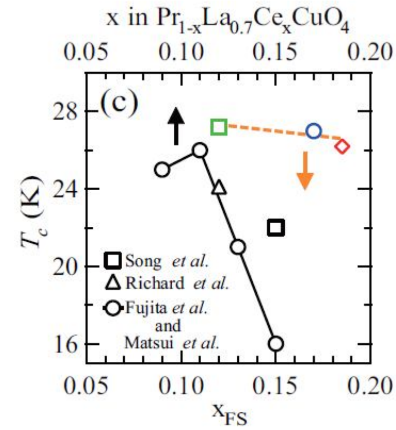

FIG. 2: T_c's of three annealed samples plotted against x_{FS} which denotes the doping level estimated from the area of fitted Fermi surface. For comparison, the T_c's of $Pr_{1-x}LaCexCuO_4$ samples used in the previous neutron scattering and ARPES studies [3,4,11] are also plotted against nominal Ce concentration.

$$\varepsilon - \mu = \varepsilon_0 - 2t(\cos k_x a + \cos k_y a) - 4t' \cos k_x a \cos k_y a - 2t''(\cos 2k_x a + \cos 2k_y a).$$

The doped electron concentration estimated from the area of the fitted Fermi surface x_{FS} was found in the range from $x_{FS} = 0.12$ to 0.185, significantly larger than that is expected from the nominal Ce concentration of x=0.10. Because about 1% oxygen is known to be removed from the sample by annealing, the origin of the additional electron doping cannot be explained at this moment. In

Fig. 2, the T_c's of the three reduction annealed samples are plotted against the electron concentration. In Fig. 2, T_c's of the $Pr_{1-x}LaCe_xCuO_4$ samples which were used in the previous neutron scattering [11] and ARPES studies [3,4,12] are also plotted with respect to the nominal Ce concentration. In the previous studies, the *Tc* decreases with increasing Ce concentration. On the other hand, the present samples maintain the high *Tc*'s compared to all the previous samples up to highest x_{FS}=0.185. This indicates that the suppression of the AF pseudogap rather than high electron doping concentration is essential for the enhancement of *Tc* in the course of reduction annealing.

In conclusion, we have performed ARPES measurements on $Pr_{1.2}La_{0.7}Ce_{0.1}CuO_4$ samples with varying the annealing condition. Samples were annealed by a new method and achieved a T_c as high as 27.0 K. Annealing samples didn't show the signature of the AF pseudogap which is typical for e-HTSCs, indicating that the suppression of AFM is responsible for the enhancement of the *Tc*. This result may continuously connected to the superconductivity at zero-doping recently observed in thin film and powder samples of e-HTSCs [7,8].

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