Suppression of the antiferromagnetic pseudogap in the electron-doped high-temperature superconductor Pr$_{1.3-x}$La$_{0.7}$Ce$_x$CuO$_{4+8}$

Masafumi Horio$^1$, Teppei Yoshida$^1$, Hakuto Suzuki$^1$, L.C.C.Ambolode Il$^1$, Atsushi Fujimori$^1$, Daiki Otsuki$^1$, Takashi Mizokawa$^2$, Kanta Ono$^3$, Hiroshi Kumigashira$^3$, Hiroaki Anzai$^4$, Masashi Arita$^4$, Hirofumi Namatame$^4$, Masaki Taniguchi$^5$, Yosuke Mori$^6$, Akira Takahashi$^6$, Tadashi Adachi$^6$, and Yoji Koike$^6$

$^1$Department of Physics, University of Tokyo, Bunkyo-ku, Tokyo 113-0033, Japan
$^2$Graduate School of Frontier Sciences, University of Tokyo, Kashiwa, Chiba 277-0882, Japan
$^3$Photon Factory, KEK, Tsukuba, Ibaraki 305-0001, Japan
$^4$Hiroshima Synchrotron Radiation Center, Hiroshima University, Higashi-Hiroshima, Hiroshima 739-0046, Japan
$^5$Department of Physical Science, Hiroshima University, Higashi-Hiroshima, Hiroshima 739-0046, Japan and $^6$Department of Applied Physics, Tohoku University, Sendai, Miyagi 980-8579, Japan

1 Introduction

In the underdoped region of the electron-doped high-temperature superconductors (e-HTSCs), a large pseudogap of order $\sim 10$ 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 antiferromagnets 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$_x$CuO$_4$ (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$_x$CuO$_4(x = 0.10)$ 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$_x$CuO$_4(x = 0.10)$ with varying annealing conditions.

2 Experiment

Single crystals of Pr$_{1.3-x}$La$_{0.7}$Ce$_x$CuO$_4(x = 0.10)$ 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 $h\nu=55$eV. The samples were cleaved in situ at $T=15$K in an ultra-high vacuum of $2\times10^{-10}$ Torr.
3 Results and Discussion

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 spots" 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 spots" 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 quasiparticle 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:

\[
\varepsilon - \mu = \varepsilon_0 - 2t(\cos k_xa + \cos k_ya) - 4t' \cos k_xa \cos k_ya - 2t''(\cos 2k_xa + \cos 2k_ya).
\]

The doped electron concentration estimated from the area of fitted Fermi surface which denotes the doping level estimated from the area of 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$_x$CuO$_4$ 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 $T_c$ decreases with increasing Ce concentration. On the other hand, the present samples maintain the high $T_c$’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 $T_c$ 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 $T_c$. This result may continuously connected to the superconductivity at zero-doping recently observed in thin film and powder samples of e-HTSCs [7,8].

Acknowledgement

This work was supported by A3 Foresight Program from the Japan Society for the Promotion of Science and a Grant-in-Aid for Scientific Research on Innovative Area "Frontier of Materials, Life and Particle Science Explored by Ultra Slow Muon Microscope".

References


* horio@wyvern.phys.s.u-tokyo.ac.jp