Evolution of the pseudogap across the magnet-superconductor phase boundary of $Nd_{2-x}Ce_xCuO_4$

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The doping dependence of electronic states in an electron-doped high-temperature superconductor (HTSC) $Nd_{2-x}Ce_xCuO_4$ was studied by high-resolution angle-resolved photoemission spectroscopy. We observed that the high-energy pseudogap around the hot spot shows an abrupt filling-in at the magnet-superconductor phase boundary, resulting in the unusual reconstruction of the Fermi surface. The magnitude (Δ_{PG}) and the temperature (T^*) at which the pseudogap is filled-in show a close relation to the effective hyperfine coupling energy (J_{eff}) and the spin-correlation length (ξ_{AF}), respectively. These results suggest the magnetic origin of the pseudogap and the unconventional nature of the magnet-superconductor phase transition in electron-doped HTSC.

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High-temperature superconductivity in copper oxides is realized by doping a moderate amount of holes or electrons into the parent Mott insulator. One of the most intriguing targets of research on high-temperature superconductors (HTSC) is how the electronic states evolve from the insulator to the superconductor. For the hole-doped system, the evolution is accompanied by formation of a pseudogap defined as suppression of the electronic states near the Fermi energy (E_F). Although the pseudogap in hole-doped materials has been intensively studied by various experiments,[1,2] the physical origin and the relation to the superconducting (SC) mechanism are still subjects of fierce debate.

Recently, the study of pseudogap in HTSC is extended to electron-doped materials. The optical conductivity in $Nd_{2-x}Ce_xCuO_4$ (NCCO) shows a pseudogaplike feature, whose energy and characteristic temperature decrease with increasing doping.[3–5] Angle-resolved photoemission spectroscopy (ARPES) observed a similar pseudogap with a characteristic momentum dependence.[6,7] These observations have suggested that the origin of pseudogap in electron-doped HTSC may be the antiferromagnetic (AF) electron correlation, and raised a question how the pseudogap interplays with the SC order. This problem has attracted a special attention after the proposal that the magnetic quantum critical point (QCP) may exist in the optimal SC phase of electron-doped HTSC (Refs. 4 and 8–10) as in heavy fermion superconductors.[11] In fact, an optical study suggested that the pseudogap onset temperature goes to zero at the QCP of $x \sim 0.17$ in the SC phase.[4] However, a subsequent optical study[5] reported that the pseudogap gradually fades away with doping, suggesting no direct relation to the QCP. In order to settle the confusion and elucidate the nature of the AF-SC phase transition, it is important to directly observe the evolution of pseudogap as a function of doping and temperature around the AF-SC phase boundary.

In this paper, we report a systematic doping- and temperature-dependent ARPES study on NCCO in the range of the AF to the overdoped SC phase ($x=0.13-0.17$). We observed that the pseudogap at the hot spot shows an abrupt filling-in at the AF-SC phase boundary, while the energy (Δ_{PG}) and the onset temperature (T^*) of pseudogap do not show such a drastic change even on crossing the boundary. We found that Δ_{PG} and T^* show a good correspondence to the effective hyperfine coupling energy (J_{eff}) and the spin-correlation length (ξ_{AF}), respectively. These results suggest the magnetic origin of pseudogap as well as the anomalous nature of the AF-SC phase transition in electron-doped HTSC.

High-quality single crystals of $Nd_{2-x}Ce_xCuO_4$, ($x=0.13$, 0.15, 0.16, 0.17) were grown by the traveling-solvent floating-zone method. Samples of $x=0.13$ and 0.15 show the Néel order below 110 K and 80 K, respectively, and those of $x=0.15$, 0.16, and 0.17 show the superconductivity below 25 K. Details of samples used in this study are described elsewhere.[12] ARPES measurements were performed using a SES-200 spectrometer at Tohoku University and the undulator 4m-NIM beamline at the Synchrotron Radiation Center, Wisconsin. We used the monochromatized He Iα resonance line (21.218 eV) and 22 eV photons to excite photoelectrons. The energy and angular (momentum) resolutions were set at 11 meV and 0.2° (0.01 $Å^{-1}$), respectively. A clean surface of sample for ARPES measurements was obtained by *in situ* cleaving along the (001) plane.

Figure 1(a) shows the experimentally determined Fermi surface (FS) of NCCO for four different samples of $x=0.13-0.17$, obtained by integrating the ARPES intensity over ±20 meV with respect to E_F at 30 K as a function of a two-dimensional wave vector. We find that the FS is circularlike centered at (π,π) for all samples as predicted by the LDA band calculation[13] although there is a characteristic intensity variation along the FS. In the underdoped sample of $x=0.13$, we find a considerable intensity suppression around the hot spot defined as a point where the FS intersects the magnetic zone boundary $(\pi,0)$-$(0,\pi)$. This intensity suppression produces a pseudogap around the hot spot and at the same time causes the fragmentation of FS into two pieces at $(\pi/2,\pi/2)$ and $(\pi,0)$ or $(0,\pi)$, respectively. On increasing the doping from $x=0.13$ to $x=0.16$, the intensity between

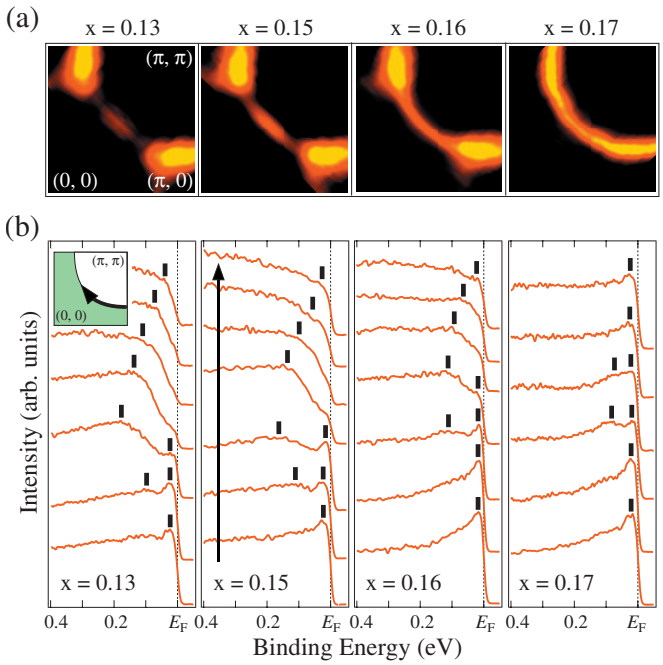

FIG. 1. (Color) (a) Doping dependence of the FS in NCCO, obtained by plotting the ARPES intensity integrated over ±20 meV with respect to EF as a function of a two-dimensional wave vector. The intensity is normalized to that at the highest binding energy (400 meV) and symmetrized with respect to the $(0,0)$-(π,π) line. (b) Doping dependence of a set of ARPES spectra measured at several k points along the FS. The position of quasiparticle peaks and humps in the spectra is denoted by bars.

$(\pi/2,\pi/2)$ and the hot spot rapidly recovers, and finally the two pieces of FS look to be connected at $x=0.17$, resulting in a continuous circular holelike FS as shown in Fig. 1(a). It is noted here that the recovery from the fragmented FS to the continuous FS is achieved by a relatively small additional doping of about 4% from $x=0.13$ to $x=0.17$, suggesting that the electronic structure of NCCO is strongly modified at the AF-SC phase boundary.

To study the doping dependence in more detail, we show in Fig. 1(b) ARPES spectra measured at several Fermi momenta (k_F). In all samples, a large pseudogap of 100–200 meV opens midway between $(\pi,0)$ and $(\pi/2,\pi/2)$. One can also find a sharp quasiparticle (QP) peak at E_F in the momentum region close to the $(\pi,0)$ point. This characteristic k dependence of ARPES spectrum in NCCO is consistent with the previous report,[7] where the QP peak and the high-energy hump are assigned as the upper and lower branches of bands folded by the AF correlation, respectively. Figure 1(b) clearly shows that the magnitude (size) of the pseudogap gradually decreases and at the same time the pseudogap itself is systematically filled-in when the doping level is changed from $x=0.13$ to $x=0.17$.

First, we discuss the doping dependence of the pseudogap energy. Figure 2(a) shows ARPES spectra for four different samples of $x=0.13$–0.17, measured at the k point where the binding energy of the hump structure in ARPES spectrum shows the maximum value for each sample. We define this maximum binding energy of the hump structure as the

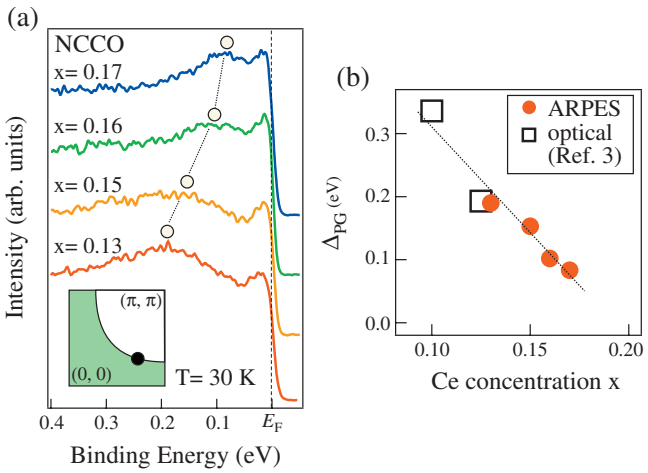

FIG. 2. (Color) (a) Doping dependence of ARPES spectrum measured at the k point on the FS shown by a filled circle in the inset, where the hump structure (open circles) has the maximum binding energy for each sample. (b) Pseudogap energy Δ_{PG} obtained from the present ARPES (red circles) plotted as a function of Ce concentration, compared with those from the optical study (squares) (Ref. 3). The straight line is a guide for the eyes.

pseudogap energy (Δ_{PG}). As clearly seen in Fig. 2(a), the hump structure denoted by open circles gradually approaches E_F with increasing the doping level, suggesting the doping-induced reduction of Δ_{PG}. Figure 2(b) shows the plot of Δ_{PG} as a function of Ce concentration, compared with the optical conductivity experiment.[3] We find that both experimental results are located on one line, showing a linear dependence of Δ_{PG} on the Ce concentration. By extrapolating the linear line in Fig. 2(b), we evaluate the critical doping level where Δ_{PG} vanishes as $x\sim0.2$. These behaviors of Δ_{PG} show a good agreement with those of the magnetic excitations measured by the inelastic neutron scattering (INS), which reported that the effective hyperfine coupling energy (J_{eff}) is linearly reduced by doping and vanishes around $x=0.2$.[14] This agreement between ARPES and INS gives an alternative evidence for the magnetic origin of the pseudogap in NCCO.

Next, we discuss the doping-induced filling behavior of the pseudogap. Figure 3(a) shows the doping dependence of ARPES spectrum measured at the hot spot. As seen in the figure, the pseudogap is gradually filled-in due to the growth of the states inside the gap (in-gap states) on going from $x=0.13$ to $x=0.17$. We have numerically fit the spectra with two Lorentzians at and away from E_F, respectively, multiplied by the Fermi-Dirac function as shown in Fig. 3(a). The Lorentzian away from E_F is asymmetrically broadened to reproduce the high-energy part of spectrum. The asymmetry can be attributed to the enegy-dependent quasiparticle lifetime and the angle-independent background signal seen over $k\geqslant k_F$.[7] It is obvious from Fig. 3(a) that the in-gap states rapidly increase within a very small fraction of doping from $x=0.13$ to $x=0.17$. It is remarked that the pseudogap is still substantially large [about 80 meV, see Fig. 2(a)] even at $x=0.17$ while the in-gap states sufficiently grow up to give the continuous FS at the same doping level as seen in Fig. 1(a). This means that the disappearance of pseudogap is not sim-

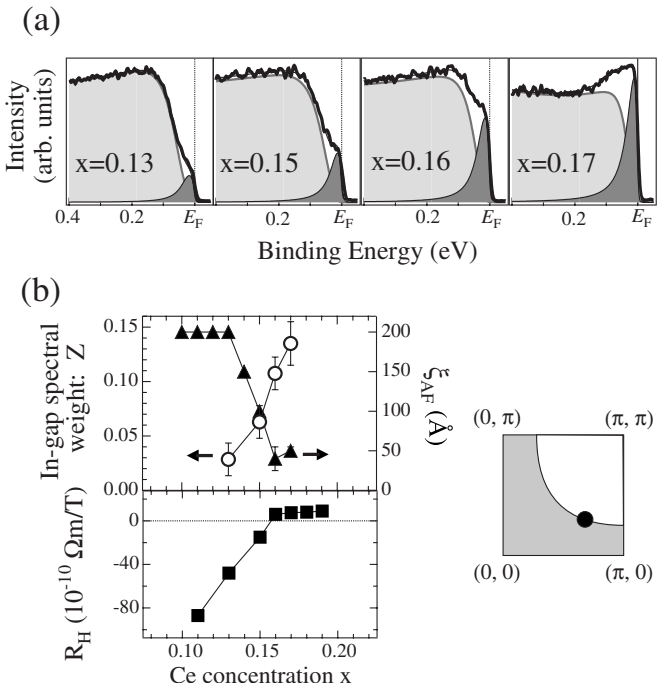

FIG. 3. (a) Doping dependence of ARPES spectrum measured at the hot spot. The spectra are fitted by two Lorentzians at and away from E_F, respectively, multiplied by the Fermi-Dirac function. The Lorentzian away from E_F is asymmetrically broadened to reproduce the high-energy part of the spectrum. (b) The ratio of the spectral weight (Z) of the in-gap states to that of the whole spectrum as a function of doping, compared with the in-plane spin-correlation length (ξ_{AF}) (Ref. 12) and the Hall coefficient (R_H) at low temperature (Ref. 10).

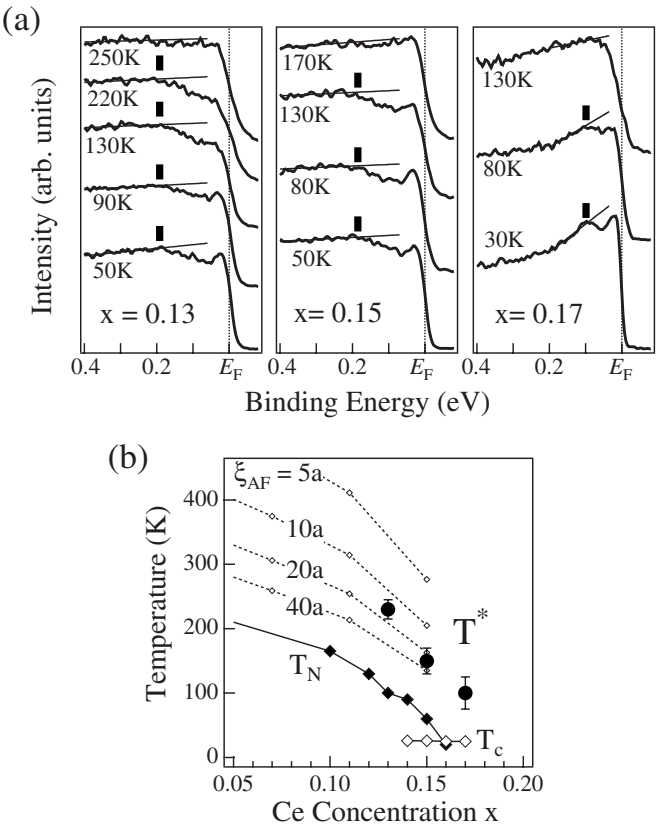

FIG. 4. Temperature dependence of ARPES spectrum for three different samples of $x=0.13$, 0.15, and 0.17, measured at the k point shown in the inset of Fig. 2. A straight line on each spectrum is a linear extrapolation of the high-energy part of the spectrum, showing the onset of the pseudogap. (b) Plot of the onset temperature of the pseudogap as a function of Ce concentration, compared with the contour lines of the spin-correlation length (ξ_{AF}) (Ref. 19). The contour lines are shifted as a whole by $\delta x=-0.03$ to compensate the absence of oxygen-reduction treatment in the as-grown samples used in the neutron scattering (Ref. 19).

ply the diminishment of the pseudogap energy to zero, but accompanied by the gap-filling effect near the AF-SC boundary. We show in Fig. 3(b) the ratio of the spectral weight (Z) of the in-gap states to that of the whole spectrum as a function of doping. Also shown in Fig. 3(b) are the in-plane spin-correlation length (ξ_{AF}) (Ref. 12) and the Hall coefficient (R_H) at low temperature.[10] The rapid increase of Z around $x=0.15$ shows a similar trend as the rapid drop of ξ_{AF} in the same doping range, suggesting that the reduction of the AF spin-correlation length in the CuO_2 plane is closely related to the filling of pseudogap. It is expected that the doping-induced drastic change in the electronic structure near E_F may cause anomalous transport behaviors.[10,15–18] In fact, as seen in Fig. 3(b), the anomalous doping dependence of R_H, namely the monotonic increase with doping and the sudden saturation accompanied with the change of sign at $x=0.16$, shows a qulitative agreement with the doping dependence of Z. This suggests that the formation and/or disappearance of pseudogap strongly affects the transport properties. The previous ARPES study[6] reported that in the lightly doped NCCO ($x\leqslant 0.10$), the FS is basically divided into the electron pocket around $(\pi,0)$ and the small hole pocket at $(\pi/2,\pi/2)$. This two-FS topology has been suspected to explain the negative R_H in the lightly doped NCCO.[15] Here it is expected that on further doping, the spin correlation becomes short ranged, and at a certain doping the continuous holelike FS centered at (π,π) is recovered causing the saturation and the change of sign in R_H. The close relation among Z, ξ_{AF}, and R_H revealed in the present study supports this picture, and indicates that the anomalous transport properties in electron-doped HTSC originate in the decay of the AF spin correlation.

Temperature-dependent measurements give an additional evidence for the close relation between the spin correlation and the pseudogap filling. Figure 4(a) shows the temperature dependence of ARPES spectrum measured at the k point shown in the inset of Fig. 2. In the measurements, we increased the temperature from lowest to highest, and then decreased again to confirm the reopening of the pseudogap. On increasing temperature across T_N (110 K and 80 K for $x=0.13$ and 0.15, respectively), the spectrum first shows no significant change except for the thermal broadening of the quasiparticle peak near E_F. The gap energy is almost constant independent of temperature for all samples. This suggests that while the long-range spin-order disappears at T_N, the interaction to produce the pseudogap remains even above T_N. However, on further increasing temperature, the

pseudogap is suddenly filled-up at around 230 K, 150 K, and 100 K for $x=0.13$, 0.15, and 0.17, respectively, as evident by the disappearance of the remarkable deviation of the spectral curve from the linear extrapolation of the high-energy part superimposed on each spectrum for comparison. Since the simple thermal effect due to the Fermi-Dirac distribution function would rather suppress the spectral weight in this energy region at high temperature, the observed sudden increase of the spectral weight certainly indicates the sudden filling of the pseudogap at each onset temperature (T^*). The observed T^* is plotted as a function of doping in Fig. 4(b), where we also show the contour line of the spin-correlation length (ξ_{AF}) obtained from the neutron scattering[19] for comparison. As seen in the figure, the T^* value decreases with increasing doping as in the previous optical studies.[3,4] It is remarked that the T^* values obtained in the present ARPES are situated in the temperature range where ξ_{AF} becomes about 20 times the lattice constant (3.95 Å). This indicates the importance of the long-range spin-correlation for the formation of the pseudogap in electron-doped HTSC. Here, it is noted that the obtained T^* value at $x=0.17$ is considerably high in contrast to the recent optical study[4] which suggests that T^* goes to zero at $x\sim0.17$. This contradiction between the ARPES and the optical study may be attributed to the momentum-integrating nature of the optical experiment. Since the pseudogap in electron-doped HTSC is strongly anisotropic as seen in Fig. 1(b), the pseudogap structure in the momentum-integrated spectrum may become too broad and vague to trace in the overdoped region.

Finally we discuss the nature of the AF-SC phase transition in NCCO. We observed that the pseudogap in NCCO shows a drastic "filling-in" effect at the doping range of $0.13\leqslant x\leqslant 0.17$, while the magnitude of the pseudogap does not vanish below $x=0.17$. As shown in Fig. 3, the spectral weight is systematically transferred by doping from the pseudogap to the metallic states in a similar trend as the reduction of the spin-correlation length. This may indicate the microscopic phase competition between the static AF order and the SC order under the AF fluctuation, since ARPES gives the spatially integrated electronic states. However, we cannot rule out the possibility of the homogeneous intermediate state at the phase boundary since the diamagnetic susceptibility proportional to the SC volume fraction reaches maximum at $x=0.14$ and does not change up to $x=0.17$.[12] It is required to construct a new theoretical framework describing the unconventional AF-SC phase transition in electron-doped HTSC.

In conclusion, we have performed systematic ARPES measurements on an electron-doped HTSC NCCO and observed the evolution of the pseudogap across the AF-SC phase boundary. We found that the pseudogap at the hot spot undergoes an abrupt filling-in around the AF-SC phase boundary, resulting in the drastic modification of FS to cause the anomalous transport behaviors. The pseudogap energy and the onset temperature are gradually decreased with doping, but do not vanish at the AF-SC phase boundary, keeping substantially large values even in the overdoped region. The present ARPES results suggest the magnetic origin of the pseudogap and the unconventional mechanism of the AF-SC phase transition in electron-doped HTSC.

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