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Small and Large Pseudogaps in High-T_c Superconductors Observed by Ultrahigh-Resolution Photoemission Spectroscopy

T. Sato[a], Y. Naitoh[a], T. Kamiyama[a], T. Takahashi[a], T. Yokoya[b], K. Yamada[c], Y. Endoh[d], and K. Kadowaki[e]

[a]Department of Physics, Tohoku University, Senidai 980-8578, Japan

[b]Institute for Solid State Physics, University of Tokyo, Kashiwa, Chiba 277-8581, Japan

[c]Institute for Chemical Research, Kyoto University, Uji 611-0011, Japan

[d]Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan

[e]Institute of Materials Science, University of Tsukuba, Ibaraki 305-3573, Japan

Two different (small and large) pseudogaps have been observed by ultrahigh-resolution photoemission spectroscopy in optimally doped $Bi_2Sr_2CaCu_2O_8$ and $La_{1.85}Sr_{0.15}CuO_4$. The small pseudogap is smoothly connected from the superconducting gap and closes slightly above T_c, while the large one appears as a remarkable depletion of the density of states in a wider energy range near E_F and persists far above T_c. The small pseudogap is likely associated with pairing and the large one is ascribed to the development of antiferromagnetic correlation.

Pseudogap, observed by various experimental techniques such as angle-resolved photoemission (ARPES) [1,2] and NMR [3], has been a subject of intensive discussion and is now regarded as a key property directly related to the high-T_c mechanism. ARPES has revealed that the pseudogap is smoothly connected from the superconducting gap across T_c with the same $d_{x^2-y^2}$ symmetry and closes at a certain temperature (T^*) higher than T_c [2]. In fact, many thermal and transport properties of underdoped high-T_c superconductors show various anomalies in the "anomalous metallic phase" [3–9] ascribable to opening of the spin/charge(pseudo) gap. However, the temperature at which the anomaly appears and its doping dependence do not necessarily coincide with each other [3–11], causing a confusion in investigating the origin as well as the relation to the superconducting mechanism. In this paper, we report an ultrahigh-resolution (ΔE=7 meV) angle-resolved (ARPES) and angle-integrated (AIPES) photoemission study on $Bi_2Sr_2CaCu_2O_8$ (Bi2212) and $La_{1.85}Sr_{0.15}CuO_4$. We found that there are two different pseudogaps at E_F; one is a "small pseudogap" which is smoothly connected from the superconducting gap and the other is a "large pseudogap" which is seen as a depletion of the density of states (DOS) near E_F and seems not to be directly connected to the superconducting gap.

Single crystals of nearly optimally doped Bi2212 (T_c= 90 K) and LSCO (T_c= 38 K) were grown by the traveling-solvent floating-zone method. Ultrahigh-resolution PES (ΔE=7 meV) measurements were performed using a SCIENTA SES-200 spectrometer with a high-flux discharge lamp. We used the He I α (21.218 eV) line to excite photoelectrons. Crystals were cleaved along (001) plane for ARPES or scraped at (110) plane for AIPES under vacuum of 5×10^{-11} Torr to obtain a clean surface. We have confirmed no angular dependence for the scraped surface. The Fermi level (E_F) of samples was referenced to a gold film evaporated onto the sample substrate and its accuracy is better than 0.5 meV.

Figure 1(a) shows the temperature dependence of ultrahigh-resolution ARPES spectrum of optimally doped Bi2212 measured at the Fermi surface closest to the (π, 0) point. Intensity

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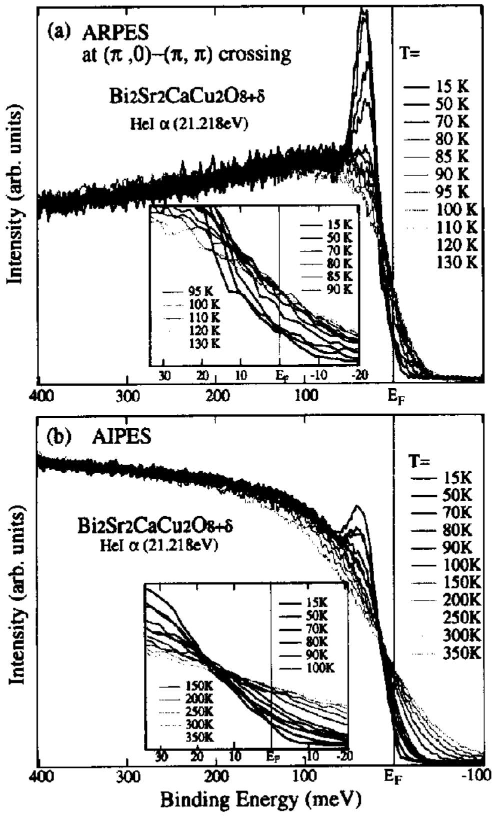

Figure 1. Temperature dependence of (a) angle-resolved and (b) angle-integrated photoemission spectrum of Bi2212 (T_c = 90 K). ARPES spectra were measured at (π, 0)-(π, π) crossing. Inset shows the same in an expanded energy scale.

of spectra is normalized to the area under the curve. Spectra in the superconducting state show a sharp coherent peak with a large leading-edge gap of about 20 meV at E_F. On increasing temperature, the coherent peak gradually loses its spectral weight without changing its energy position relative to E_F, while the leading-edge midpoint gradually approaches E_F, indicating that the superconducting gap gradually closes. Inset shows the expansion of ARPES spectra near E_F, where we find that the spectral intensity at E_F still increases slightly even above T_c but seems to saturate at 110 K-130 K. This suggests existence of a pseudogap above T_c even in optimally doped Bi2212 as in the underdoped samples. Figure 1(b) shows the temperature dependence of ultrahigh-resolution angle-integrated photoemission (AIPES) spectra measured with a scraped crystal surface. It is well known that an ARPES spectrum represents the electronic structure at a particular k point in the Brillouin zone while an AIPES spectrum reflects essentially the density of states (DOS) modulated by the photoionization cross-section and the matrix element effect. Spectra at low temperatures show a leading-edge gap similar to the ARPES spectra, but are significantly broadened probably because the superconducting gap with a $d_{x^2-y^2}$ symmetry is integrated over all the Brillouin zone in AIPES. As the temperature is increased, the coherent peak gradually loses its intensity and disappears around T_c, while the peak position stays always at 35-40 meV as in the ARPES result. The leading-edge gap also gradually disappears as increasing the temperature. In spite of these similarities between ARPES and AIPES, we find some qualitative differences. AIPES spectra show a significant depletion near E_F which starts far away from the coherent peak. The spectral intensity at E_F gradually increases with increasing the temperature, but it does not saturate even at 110 K as in the ARPES case (Fig. 1(a)), rather showing a systematic increase over 200 K.

In order to see more directly the change of the spectral function $A(k,\omega)$ and the DOS free from the Fermi-Dirac distribution function, we have symmetrized both ARPES and AIPES spectra with respect to E_F[12]. Figure 2(a) shows the symmetrized ARPES spectra at the (π, 0)-(π, π) crossing. With increasing temperature, the coherent peak located at 35-40 meV reduces its intensity and at the same time the gap is gradually filled-in. However, the spectrum at 95 K (>T_c = 90 K) still shows suppression of the spectral weight at E_F, indicative of opening of a pseudogap above T_c. We find in Fig. 2(a) that the spectra at 90-100 K clearly show a deviation from the 130 K spectrum near E_F, while the spectra at 110-120 K coincide well with that at 130 K. This suggests that optimally doped Bi2212 has a

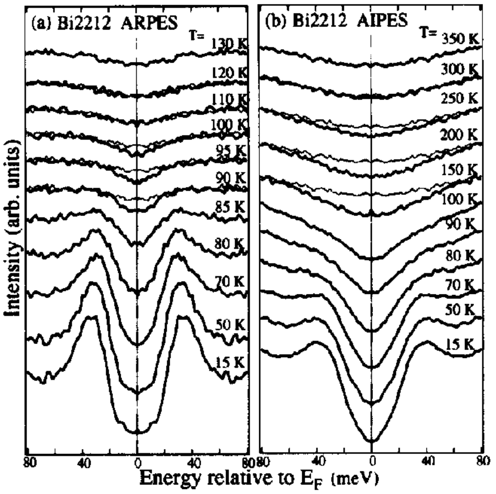

Figure 2. (a) Symmetrized angle-resolved photoemission spectra of optimally doped Bi2212 measured at (π, 0)-(π, π) crossing. Superimposed gray curve is the 130 K spectrum. (b) Symmetrized AIPES photoemission spectra of optimally doped Bi2212. Superimposed gray curve is the 350 K spectrum.

pseudogap above T_c (90 K), which closes around T^* =110 K. In Fig. 2(b), we show the symmetrized AIPES spectra in the same energy scale as in Fig. 2(a). Although the symmetrized spectra of AIPES look similar to those of ARPES, there is a striking difference in the temperature range. The temperature-induced change saturates around T^*=110 K in ARPES while that of AIPES shows a gradual change even over 200 K and saturates around 300 K. We call this saturation temperature in the AIPES spectrum (or DOS) as T_0. The energy scale of the depletion in the DOS near E_F is much larger than that of the superconducting gap (Δ~40 meV), suggesting that there are two different pseudogaps in Bi2212; one is smoothly connected from the superconducting gap across T_c and the other appears as a depletion in the DOS in a wider energy range and is not directly connected to the superconducting gap. We call the former and the latter pseudogap as a "small" and a "large" pseudogap, respectively. We have observed a similar "large pseudogap" with almost the same energy scale (100-150 meV) also in $La_{1.85}Sr_{0.15}CuO_4$ while a small pseudogap has not been well resolved in the experiment [13,14]. The observed smooth evolution from the superconducting gap to the small pseudogap suggests that the small pseudogap observed by the present ARPES on optimally doped Bi2212 is essentially the same as the "pseudogap" observed by ARPES on underdoped samples [1,2]. This means that the phase boundary defined by T^* does not fall on the top of the superconducting phase but a slightly away from it in the overdoped region. In fact, the in-plane resistivity of optimally doped Bi2212 shows a deviation from the T-linear behavior at a temperature higher than T_c (~150 K) [7,8]. A recent NMR study on nearly optimally doped Bi2212 (T_c = 86 K) shows a spin-gap behavior in $1/T_1T$ above T_c (T^* ~130 K)[9], supporting the present ARPES result.

On the other hand, the large pseudogap is not smoothly connected to the superconducting gap nor the small pseudogap. The temperature-induced evolution continues over T^* and appears to saturate around T_0 =300 K. This saturation temperature coincides well with the temperature at which the uniform magnetic susceptibility starts to decrease ($T_{m\chi}$ ~300 K) [8]. This suggests that the large pseudogap is related to the development of antiferromagnetic correlation below $T_{m\chi}$. A recent NMR study on Bi2212 (T_c = 86 K) has reported that the Knight shift starts to deviate from the T-linear dependence at a temperature above 200 K [9]. This may be related to the temperature-induced evolution of the DOS near E_F observed in the present AIPES. Further a recent tunneling spectroscopy on optimally doped Bi2212 [10] has reported that a pseudogap larger than the superconducting gap opens at E_F in the tunneling spectrum even at 300 K. The shape of tunneling spectrum and its temperature dependence are very similar to those of the present AIPES spectrum. It is reasonable to ascribe the gap observed by the tunneling to the large pseudogap because the tunneling spectroscopy essentially probes the DOS as AIPES.

In Fig. 3, we summarize characteristic temper-

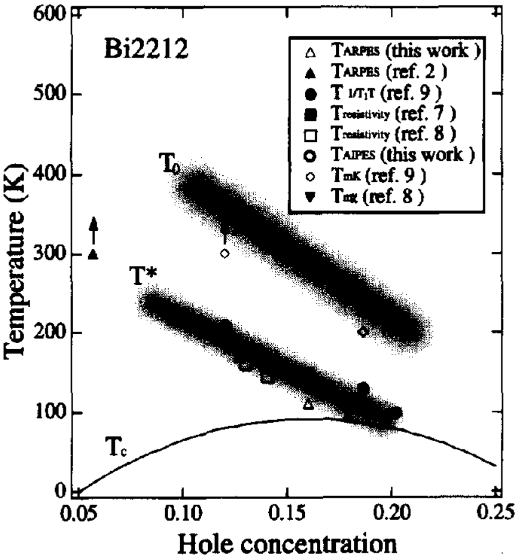

Figure 3. Comparison of characteristic temperatures of various physical properties as a function of doping in Bi2212. Characteristic temperatures of small pseudogap (△ and ▲, ARPES of this work and ref. 2), $1/T_1T$ (●, ref. 9) , and in-plane resistivity (■, ref. 7 and □, ref. 8) are on the same line just above the T_c curve, while those of large pseudogap (○, AIPES of this work), Knight shift (◇, ref. 9), and magnetic susceptibility (▼, ref. 8) form another line far above the T_c and T^* curves. Symbols with an arrow show the lowest bound of the temperature. The hole concentration of samples is determined from the T_c [15].

atures of various physical properties as a function of doping to demonstrate that there are two different pseudogaps in Bi2212. The characteristic temperatures of a small pseudogap (ARPES), $1/T_1T$ (NMR) [9], and the in-plane resistivity [7,8] appear to be on the same line (T^*) just above the T_c curve, while those of the DOS (AIPES), Knight shift (NMR) [9], and the magnetic susceptibility form another line (T_0) far above the T_c and T^* lines.

In conclusion, from the present ultrahigh-resolution photoemission study on optimally doped Bi2212 and LSCO, we found two different pseudogaps at E_F. One is a "small pseudogap" which is smoothly connected from the superconducting gap across T_c and closes slightly above T_c. The other is a "large pseudogap" which appears as a depletion of DOS near E_F in a wider energy range and is not directly connected to the superconducting gap. The temperature-induced filling of large pseudogap saturates far above T_c. These results suggest that the small pseudogap originates in the superconducting paring while the large pseudogap is closely related to the development of the antiferromagnetic correlation.

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