Universal versus Material-Dependent Two-Gap Behaviors of the High-T_c Cuprate Superconductors: Angle-Resolved Photoemission Study of $La_{2-x}Sr_xCuO_4$

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We have investigated the doping and temperature dependences of the pseudogap and superconducting gap in the single-layer cuprate $La_{2-x}Sr_xCuO_4$ by angle-resolved photoemission spectroscopy. The results clearly exhibit two distinct energy and temperature scales, namely, the gap around $(\pi, 0)$ of magnitude Δ^* and the gap around the node characterized by the d-wave order parameter Δ_0. In comparison with Bi2212 having higher T_c's, Δ_0 is smaller, while Δ^* and T^* are similar. This result suggests that Δ^* and T^* are approximately material-independent properties of a single CuO_2 plane, in contrast to the material-dependent Δ_0, representing the pairing strength.

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One of the central issues in the studies of high-T_c cuprates is whether the pseudogap is related to the superconductivity or a phenomenon distinct from superconductivity. In the former scenario, possible origins of the pseudogap are preformed Cooper pairs lacking phase coherence [1] or superconducting fluctuations [2]. In the latter scenario, the pseudogap is due to a competing order such as spin density wave, charge density wave, d-density wave [3], etc. It has been well known that the pseudogap in the antinodal $\sim (\pi, 0)$ region increases with underdoping as observed by angle-resolved photoemission spectroscopy (ARPES) [4] and tunneling spectroscopy [5]. However, the energy gap measured by Andreev reflection [6], penetration depth [7], and Raman experiments in B_{2g}-geometry [8,9], which is directly associated with superconductivity, exhibits the opposite trend, that is, the gap decreases with underdoping, suggesting a different origin of the superconducting gap from the antinodal gap.

A recent ARPES study of deeply underdoped $Bi_2Sr_2Ca_{1-x}Y_xCu_2O_8$ (Bi2212) has revealed the presence of two distinct energy gaps between the nodal and antinodal region [10,11]. A similar two-gap behavior has been observed in the optimally doped single-layer cuprate $Bi_2Sr_2CuO_{6+\delta}$ (Bi2201) [12,13] and $La_{2-x}Sr_xCuO_4$ (LSCO) [14,15]. On the other hand, attempts have been made to understand the pseudogap within a single d-wave energy gap [16–19]. From the measurement of the Fermi arc length, Kanigel *et al.* [18] has proposed that the $T = 0$ ground state of the pseudogap state is a nodal liquid which has a single $d_{x^2-y^2}$ gap. In such a single gap picture, preformed Cooper pairs are the most likely origin of the pseudogap.

Since the doping and temperature dependences of the energy gap would reveal the entangled two-gap behavior, we have investigated the energy gap of LSCO by ARPES as a function of doping and temperature. In the present work, the momentum dependence of the gap clearly exhibits two-gap behavior as in the previous studies [10–15]: the pseudogap Δ^* in the antinodal region and the d-wave-like gap Δ_0 around the node. Furthermore, from comparison with those on Bi2212, we have found that the magnitude of the Δ^* and the pseudogap temperature T^* is not appreciably material-dependent, suggesting that the pseudogap is a property of a single CuO_2 plane. On the other hand, the magnitude of the Δ_0, which is proportional to the superconducting gap, is strongly material dependent (CuO_2 layer number dependent) like T_c.

High-quality single crystals of LSCO ($x = 0.03$, 0.07, 0.15) were grown by the traveling-solvent floating-zone method. The critical temperatures (T_c's) of the $x = 0.07$, 0.15 samples were 14 and 39 K, respectively, and the $x = 0.03$ samples were nonsuperconducting. The ARPES measurements were carried out at BL10.0.1 of Advanced Light Source (ALS) and at BL-28A of Photon Factory (PF) using incident photons of linearly polarized 55.5 eV and circularly polarized 55 eV, respectively. SCIENTA R4000 and SES-2002 analyzer were used at ALS and PF, respectively, with the total energy resolution of ~20 meV and momentum resolution of $\sim 0.02\pi/a$, where $a = 3.8$ Å is the lattice constant. The samples were cleaved *in situ* and

measurements were performed from 20 to 155 K. In the measurements at ALS, the electric field vector **E** of the incident photons lies in the CuO_2 plane, rotated by 45° from the Cu-O bond direction, which enhances the dipole matrix elements in the nodal region [20].

Figure 1 illustrates that the quasiparticle (QP) band dispersions are determined by momentum distribution curve (MDC) peak positions. Here, the Fermi momentum k_F is defined by the momentum where the QP dispersion crosses the E_F. To quantify the energy gap size, the leading edge midpoints (LEM's) of the energy distribution curves (EDC's) at k_F shall be used in the present analysis. As shown in Figs. 1(d), 1(e), and 1(f), the LEM's at k_F are shifted toward higher binding energies in going from the node to the antinode, indicating an anisotropic gap opening.

The gap sizes have been evaluated from the shift (Δ_{LEM}) of the LEM of EDC's relative to the node. The angular dependence of the gap for each doping is plotted as a function of the d-wave parameter $|\cos(k_x a) - \cos(k_y a)|/2$ in Fig. 2(a). These plots do not obey the simple straight line expected for the pure d-wave order parameter but has a kink at $|\cos(k_x a) - \cos(k_y a)|/2 \cong 0.7$–$0.9$. Interestingly, the kink occurs near the antiferromagnetic Brillouin-zone (AFM BZ) boundary but not exactly on it, as shown by vertical arrows. Qualitatively the same results have been obtained for the single-layer cuprates Bi2201 [12] and underdoped Bi2212 [10,11]. Note that the gaps for $x = 0.07$ near the node are almost closed above T_c (=14 K), but d-wave-like gap opens below T_c as shown in the inset.

In order to discuss the character of the energy gaps, we define two distinct energy scales Δ^* and Δ_0: Δ^* from Δ_{LEM} closest to $|\cos(k_x a) - \cos(k_y a)|/2 = 1$ and Δ_0 from the extrapolation of the linear dependence near the node toward $|\cos(k_x a) - \cos(k_y a)|/2 = 1$, as indicated in panel (a). Since the Δ_{LEM} is affected by the width of EDC's, the gap magnitude (Δ) is approximately given by 2–3 times Δ_{LEM} [12]. As shown in Fig. 2(b), a relationship $\Delta = 2.2\Delta_{\mathrm{LEM}}$ well explains both the LEM and peak shift in the $x = 0.15$ data and also explains the data for $x = 0.03$ and 0.07. Therefore, we have assumed this relationship for analysis of the Δ^* and Δ_0 as described below.

In Fig. 2(c), the doping dependence of the observed Δ^* and Δ_0 thus deduced are summarized. The doping dependence of Δ^* is quantitatively consistent with various spectroscopic data such as B_{1g}-geometry Raman scattering [9]. $\Delta^* \sim 30$ meV for the $x = 0.15$ sample is consistent with the previous ARPES results, too [14]. On the other hand, Δ_0 remains unchanged in going from $x = 0.15$ to $x = 0.07$ and vanishes in the nonsuperconducting sample $x = 0.03$ [21], similar to the results of the lightly doped Bi2212 [10]. However, vortex-liquid states suggestive of superconducting states were observed in $x = 0.03$ [22]. The present result $\Delta_0 \sim 0$ for $x = 0.03$ may be due to the high tem-

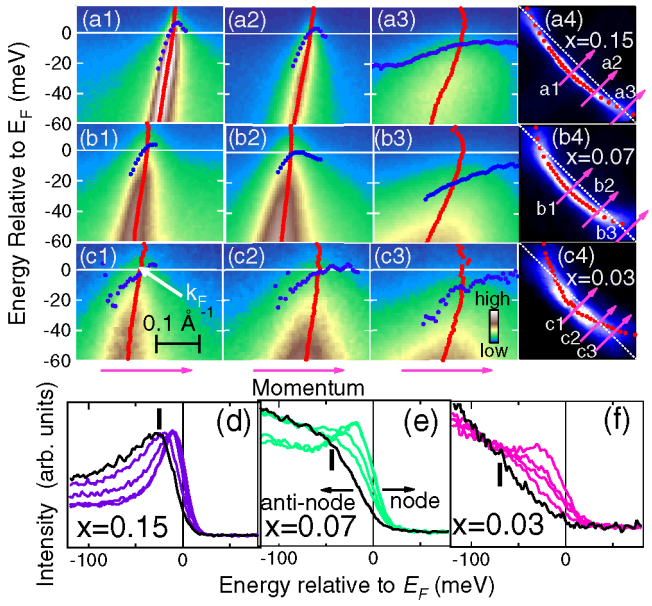

FIG. 1 (color online). ARPES intensity plot of $La_{2-x}Sr_xCuO_4$ (LSCO). (a1)–(a3), (b1)–(b3), and (c1)–(c3): Band image plots in energy-momentum (E-k) space for $x = 0.15$, 0.07 and 0.03, respectively. Energy dispersions determined by MDC's peaks and leading edge midpoints (LEM) are shown by red dots and blue dots, respectively. (a4),(b4), and (c4): Spectral weight mapping at E_F in momentum space for each doping level. Red dots indicate Fermi momenta k_F. White dotted lines indicate the antiferromagnetic Brillouin zone (AMF BZ). (d)–(f): EDC's at k_F for each doping level. Black lines correspond to antinodal EDC's and vertical bars represent energy position of the antinode gap.

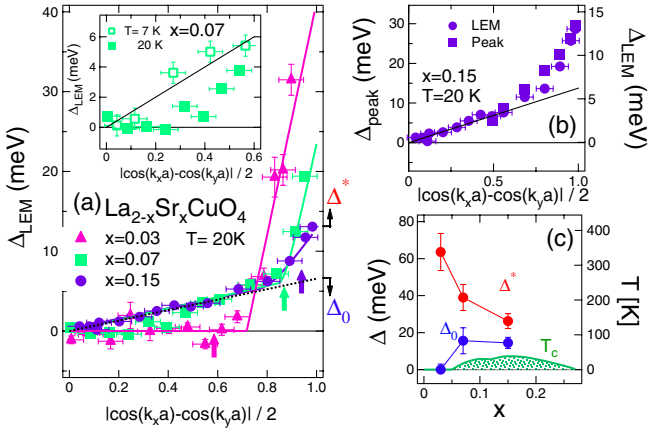

FIG. 2 (color online). Momentum dependence of the energy gap at $T = 20$ K in LSCO with various doping levels. (a): Leading edge midpoints (LEM) Δ_{LEM} relative to that at the node. Vertical arrows represents the boundary of the AFM BZ. Inset shows LEM near the node for $x = 0.07$ below and above T_c (=14 K). (b): Comparison of the peak position (Δ_{peak}) and Δ_{LEM} for $x = 0.15$, indicating the relationship $\Delta_{\mathrm{peak}} \simeq 2.2\Delta_{\mathrm{LEM}}$. (c): Doping dependence of Δ^* and Δ_0 obtained by assuming the relation in panel (b).

perature effects similar to the LEM above T_c near the nodal direction for $x = 0.07$.

The temperature dependence of the gap is shown in Fig. 3. In Figs. 3(a1), 3(a2), 3(a3), and 3(a4), EDC's for $x = 0.15$ exhibit clear shifts of the LEM between the nodal and antinodal directions in the $T = 20$, 55 and 80 K data. In contrast, the LEM at $T = 155$ K show almost no shift between the nodal and antinodal direction, indicating that the gap is closed on the entire Fermi surface. The angular dependence of the Δ_{LEM} for each temperature are plotted in Fig. 3(b). As shown in the inset, Δ^* decreases with increasing temperature and closes at $T^* \sim 150$ K, again consistent with T^* obtained from the angle-integrated photoemission results [15]. Δ_0 also decreases with temperature similar to the decrease of T_c. However Δ_0 seems finite at $T = 55$ K, slightly above T_c. Probably, the gap closes near the node direction [19], although the low energy scales in LSCO did not allow us to resolve it.

Now, let us compare the two-gap energy scales of LSCO with those of other high-T_c cuprates to clarify their relation to T_c. In Fig. 4(a), the doping dependences of Δ^*, Δ_0 and T^* for LSCO and another single-layer cuprate Bi2201 are plotted. In the same manner, those for double-layer Bi2212 which has about twice higher T_c than those of LSCO are plotted in Fig. 4(b). Interestingly, the doping dependences of Δ^* of all these samples approximately scale with T^* following the relationship $2\Delta^*/k_BT^* = 4.3$, reminiscent of the d-wave BCS relationship [23]. Furthermore, these data fall on approximately the same lines for all the compounds irrespective of the different T_c as indicated in Figs. 4(a) and 4(b). Especially, pseudogap temperatures for optimally doped $Bi_2Sr_2Ca_2Cu_3O_{10}$ (Bi2223) [24] and Bi2201 [12] are both $T^* \sim 150$ K, similar to the present result of LSCO $T^* \sim 140$ K, although they have very different T_c's. Therefore, we speculate that Δ^* is a universal property of a single CuO_2 plane and is not much affected by its chemical environment [25]. One possible explanation for the material independence of Δ^* is that its magnitude is determined by J, since the exchange interaction J is almost material independent. A pseudogap originated from antiferromagnetic spin fluctuations [26] or RVB-type spin singlet formation [27] has its origin in J. Indeed, a phenomenological model where the pseudogap Δ^* comes from the RVB gap explains the two characteristic gap energy scales [28,29].

In contrast to Δ^*, the d-wave order parameter Δ_0 of Bi2212 is twice as large as those of LSCO, reminiscent of the difference in the magnitude of T_c. The strong material dependences of Δ_0 mean that Δ_0 is not a property only of a single CuO_2 plane but also influenced by the environment such as the apical oxygens or the block layers and/or the neighboring CuO_2 planes in multilayer cuprates. Namely, the number of CuO_2 layers and the distance of the apical oxygen atoms (in block layers) from the CuO_2 plane are important factors for the superconducting gap and hence T_c. The effect from outside the CuO_2 plane has been modelled using the distant-neighbor hopping parameters t' and t'' [30], which are affected by the p_z orbital of the apical oxygen and the position of the empty Cu $4s$ orbital. In other words, the $(\pi, 0)$ pseudogap does not depend on details of the band structure nor on the parameters t', t'', but only on t and/or J.

If the pseudogap in the antinodal region precludes contribution to the superconductivity and the superconductivity comes mainly from the near-nodal region, the "effective" superconducting gap $\Delta_{\mathrm{sc}} \propto (\text{Fermi arc length}) \times \Delta_0$ rather than Δ_0 would be more directly related to T_c [31]. Here, the Fermi arc is defined by the momentum region

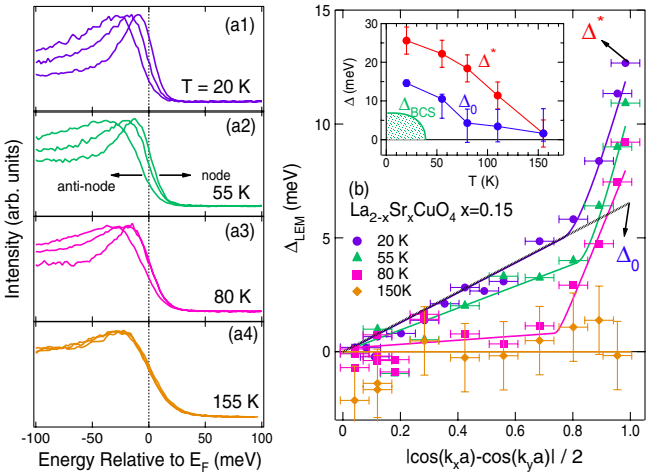

FIG. 3 (color online). Momentum dependence of the energy gap for $x = 0.15$ at various temperatures. (a1)–(a4): EDC's at k_F in the nodal to the antinodal directions. (b): Momentum dependence of Δ_{LEM} along the Fermi surface for $x = 0.15$ at various temperatures. Inset shows the temperature dependence of Δ^* and Δ_0 with the assumption $\Delta = 2.2\Delta_{\mathrm{LEM}}$, as in Fig. 2. The d-wave BCS gap Δ_{BCS} $(=4.3k_BT_c/2)$ is also plotted for comparison.

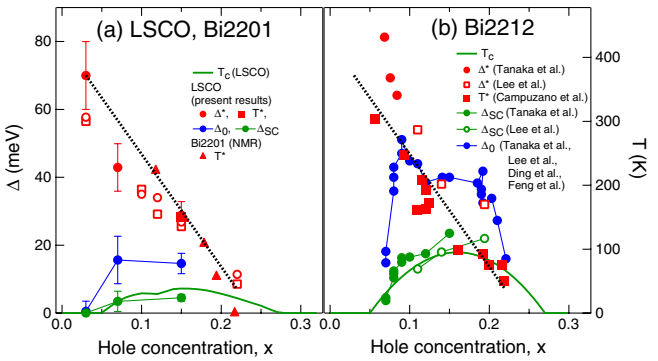

FIG. 4 (color online). Doping dependence of the characteristic energies (Δ^*, Δ_0) and temperatures (T^*, T_c) for single-layer cuprates (LSCO, Bi2201) (a) and double-layer cuprates Bi2212 (b). Gap energies Δ and temperatures T have been scaled as $2\Delta = 4.3k_BT$ in both panels. Parameter values have been taken from NMR results for Bi2201 [33], and ARPES for Bi2212 [4,10,11,34,35].

where the energy gap closes just above T_c. According to the high-resolution ARPES, the arc length for LSCO ($x = 0.15$) is ~30% of the entire Fermi surface [14]. For $x = 0.07$, the arc length is ~20% as seen in the inset of Fig. 2. Using these value for LSCO, the doping dependence of $\Delta_{sc} = (\text{arc length}) \times \Delta_0$ is plotted in Fig. 4(a). In the same manner, Δ_{sc} for Bi2212 were determined by using the Fermi arc length [11] [Fig. 4(b)]. The plotted Δ_{sc} approximately agree with the dome of T_c through the BCS formula $2\Delta_{sc} = 4.3 k_B T_c$. Particularly, the decrease of T_c with underdoping can be ascribed to the reduction of the Fermi arc length together with the Δ_0, which remains nearly constant till $x \sim 0.07$ and then drops. As for the nonsuperconducting $x = 0.03$ sample, the arc length may be too short to produce sufficient carriers for superconductivity or the nodal spectra may have a small gap due to localization as seen in the transport properties [32].

In summary, we have reported an ARPES result of LSCO and have clearly shown a signature of the two distinct energy gap scales, Δ^* and Δ_0. From comparison of the present results with those of other cuprates, we have found that the magnitude of Δ^* is almost material independent, suggesting that the pseudogap is a distinct phenomenon from superconductivity. On the other hand, Δ_0 exhibits a large difference between materials, reflecting the different superconducting properties including the different T_c's. Using the obtained two-gap parameters in conjunction with the Fermi arc picture [31], we have obtained the magnitude of the ''effective'' superconducting gap and consistently explained the doping dependence of T_c in LSCO as well as in Bi2212. The present results enforce the picture of superconductivity on the Fermi arc and clarify how T_c disappears in the underdoped region. Since the observed material dependence of Δ_0 is a crucial factor for the high-T_c superconductivity, the relationship between Δ_0 and other model parameters such as t' and t'', the number of CuO_2 planes, the apical oxygen—Cu distance, and possibly electron-phonon coupling, has to be clarified in future studies.

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