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Nonmonotonic pseudogap in high-T_c cuprates

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The mechanism of high-temperature superconductivity has not been resolved for so long because the normal state of cuprates, which exhibits enigmatic pseudogap phenomena, is not yet understood. We performed careful temperature- and momentum-resolved photoemission experiments to show that the depletion of the spectral weight in slightly underdoped cuprate superconductor, usually called the "pseudogap," exhibits an unexpected nonmonotonic temperature dependence: decreases linearly approaching T^* at which it reveals a sharp transition but does not vanish and starts to increase gradually again at higher temperature. The low-temperature behavior of the pseudogap is remarkably similar to one of the incommensurate charge ordering gap in the transition-metal dichalcogenides, while the reopening of the gap at room temperature fits the scenario of temperature-driven metal-insulator transition. This observation suggests that two phenomena, the electronic instability to density-wave formation and the entropy-driven metal-to-insulator crossover, may coexist in the normal state of cuprates.

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In the normal state, the conventional superconductors are metals and the superconductivity emerges as a particle-particle instability, known as Cooper pairing, resulting in a superconducting gap (SG) in the spectrum of one-particle excitations. The superconducting cuprates were found to be different, exhibiting a pseudogap (PG) already in the normal state,[1] and, since its discovery, the PG towers as an impregnable wall on the way to understanding the high-T_c superconductivity,[2] either foreshadowing the SG (Ref. 3) or interfering with it.[4] Although one of the first theories of a pseudogap[5] in metals was developed for one-dimensional charge-density-wave (CDW) systems,[6] it is high-temperature superconductors (HTSCs) where the PG phenomenon has started to play the lead.[1]

The PG in HTSC is considered as a peculiar phenomenon[7] because it is properly anisotropic, resembling the unique d-wave symmetry of the SG, and appears in a solid region in the universal doping-temperature (xT) phase diagram of the cuprates, i.e., closes above T^*.[2,7,8] Recent momentum-resolved experiments[9–12] have shown that the momentum distributions of the SG and PG are actually different from d wave, and one can speak about two gaps coexisting in the superconducting state. This finding, however, does not reveal the PG origin.[12] Although a number of theories of the PG do exist, choosing the true mechanism is, in part, complicated because the PG phase boundaries on xT-phase diagram, especially the crossover temperature T^*, are often ill defined.[2] We performed careful temperature- and momentum-resolved photoemission experiments to show that the PG in cuprates does not vanish at T^*, but exhibits behavior which rules out the possibility to describe it by a single mechanism such as superconducting phase fluctuations[13,14] and suggests that two phenomena, the electronic instability to density-wave formation[15] and the entropy-driven metal-to-insulator crossover, may coexist in the normal state of cuprates,[16] causing the depletion of the spectral weight in the whole temperature range.

The data were collected using the synchrotron radiation ("one-cubed ARPES" beamline at BESSY and SIS beamline at SLS)] and the photoelectron analyzers SES R4000 and SES 100. All the presented spectra are measured on one underdoped sample (x=0.11, with T_c=77 K): $Bi(Pb)_2Sr_2Ca(Tb)Cu_2O_{8+\delta}$ (Tb-BSCCO), but similar results have been obtained also for other Tb-BSCCO and Dy-BSCCO samples of similar doping level. Energy and angular resolutions were better than 10 meV and 0.2°, respectively. The data were collected from a fresh sample surface within about 4 h after cleavage which is much less than the average lifetime ($\sim$12 h at 3×10^{-11} mbar) of the samples we used. This ensures that neither the change in the effective doping concentration on sample surface nor the aging of the surface could affect the data. Nevertheless, we checked the reproducibility of the result at a few temperature points, both cycling the temperature and after 6 h of measurements, and ensured that the change in the leading edge position is within the uncertainty of single measurements and much less than the corresponding values of the observed effect. The key spectra are measured along the cut of the Brillouin zone (BZ) (vertical double-headed arrow on the last panel of Fig. 1) which goes through the hot spots (shown by large points) where, according to many theories, the PG should be largest. The energy distribution curves (EDCs) were integrated over a finite momentum range of (±0.15 Å^{-1}) around k_F. Such an integration ensures that the leading edge midpoint (LEM) of a nongapped spectrum stays at the Fermi level.[14,17]

Figures 1 and 2 present the results of the detailed temperature dependence of the one-particle excitation spectra measured by angle-resolved photoemission spectroscopy (ARPES). The data are measured along the cut (vertical double-headed arrow in the sketch of the Fermi surface in

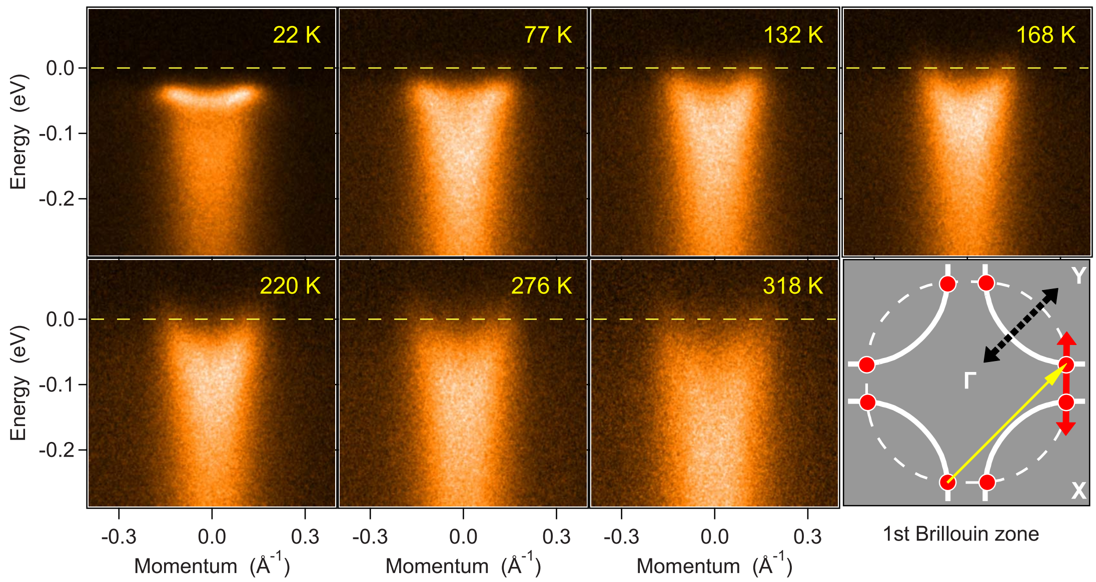

FIG. 1. (Color online) Temperature evolution of the ARPES spectra. Panels in color scale, which represent the "ARPES images" (the momentum distribution of the photoemission intensity) taken along the "hot-spot" cross section (see the last panel), demonstrate the evolution of the quasiparticle spectral weight in a wide temperature range of 20–320 K. All spectra are measured at 50 eV excitation energy at which only the antibonding band is visible. The sketch of the Fermi surface (FS) (white solid lines) illustrates the position of the hot spots (large points), which are the points on the FS connected by the (π,π) vector (thin yellow arrow); the dotted line represents the FS shifted by the (π,π) vector; the solid vertical and dashed diagonal double-headed arrows show the position of the hot-spots crossing and nodal crossing, respectively.

Fig. 1) of the BZ which goes through the hot spots [the spots at the Fermi surface which are connected by the (π,π) vector (see the sketch)] for an underdoped Tb-BSCCO. Figure 1 represents several characteristic spectra taken at different temperatures. Figure 2 contains representation of the same data set as a temperature map (panel a) and as a momentum integrated EDCs measured at different temperatures and compared to each other (panels b and e) as well as to the similar EDCs but measured for each temperature along the nodal direction (panels c, d, f, g). The gap is seen as a shift of the whole spectrum from the Fermi level (Fig. 1) or, more explicitly, as a shift of the LEM of a gapped EDC (Fig. 2). Since the momentum-integrated EDC of the nongapped spectrum is expected to stay at zero binding energy for any temperature,[14,17] as it is observed for the nodal EDCs (Fig. 2, panels c, d, f, g), the finite shift of the LEM is a good empirical measure for a gap of unknown origin. From the temperature map presented in Fig. 2(a) one can easily see an

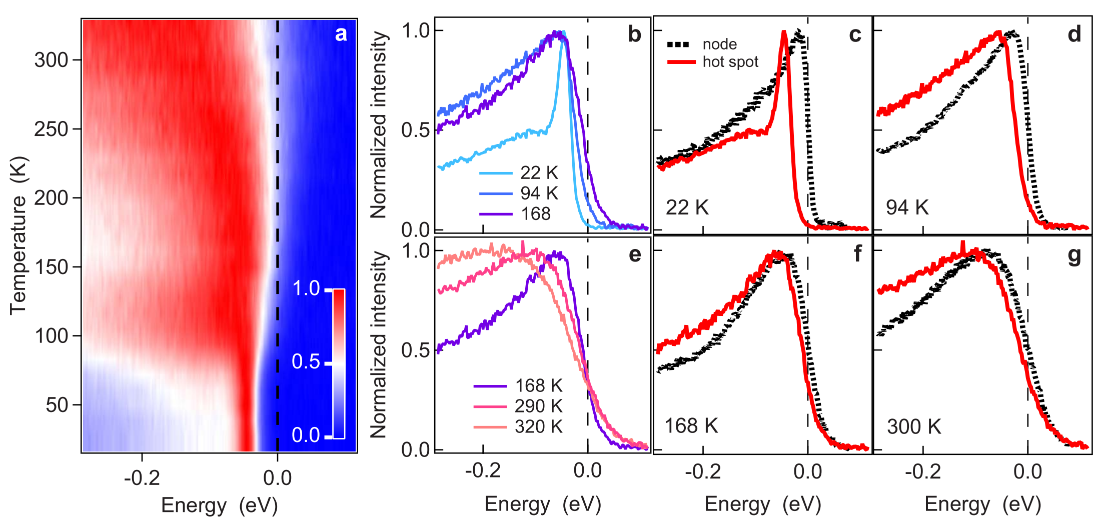

FIG. 2. (Color online) The temperature map. (a) The temperature map which consists of a number of momentum-integrated EDCs measured at different temperatures at a hot spot. Separate EDCs are shown in panels (b–g): as compared to each other (panels b and e) and to the similar EDCs measured each for the same temperature but along the nodal direction (panels c, d, f, g). The gap is seen as a shift of the LEM. In terms of the color scale of panel a, the LEM corresponds to white color close to the Fermi level. All the hot-spot EDCs are integrated in momentum range of ± 0.15 Å^{-1} around k_F.

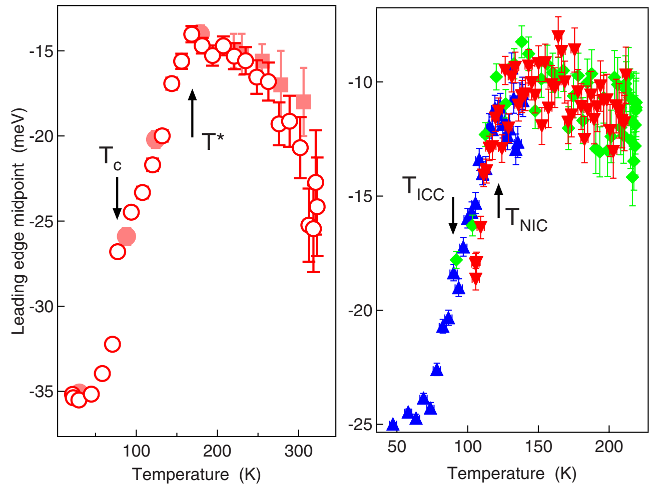

FIG. 3. (Color online) Nonmonotonic gap function. The position of the LEM of the integrated k_F EDCs (averaged for two Fermi crossings) as a function of temperature for an underdoped Tb-BSCCO (left) with T_c=77 K and T^*=170 K is remarkably similar to the pseudogap in a transition-metal dichalcogenide $TaSe_2$ (right) (Ref. 6) with the transitions to the commensurate and incommensurate CDW phases at T_{ICC}=90 K and T_{NIC}=122 K, respectively.

unusual temperature evolution of the gap (in terms of the color scale, the LEM corresponds to the white color): first it decreases with increasing temperature up to about 170 K, then it starts to increase again. It is important to stress that the LEM is not the only quantity which describes the PG phenomenon. The shift of the LEM is also correlated with the width of the spectra as well as with the position of the EDC maxima [see Fig. 2(a)]: the closer the LEM is to the Fermi level, the narrower the spectrum is (compare spectra at 132 K and 220 K to 168 K in Fig. 1). In other words, in the normal state it is at 170 K when the quasiparticles are most coherent.

The left panel of Fig. 3 summarizes the extracted values of LEM for the two hot spots of Tb-BSCCO. The presented temperature dependences allow us to highlight the following PG properties that have not been observed in cuprates before. First, the PG persists over the whole temperature range, even well above T^* ($\sim$200 K) at which it is expected to close for BSCCO at such a doping level.[8,18] Second, the gap value is nonmonotonic, reaching a finite minimum value at about 170 K and increasing on both sides. Third, a sharp change in the slope of the dependence at 170 K suggests a real phase transition at this temperature (which we denote as T^* here). Alternatively, T^* can be defined as the temperature at which the quasiparticles are most coherent. The observed $\Delta(T)$ dependence indicates that the previous understanding of the PG properties was, at least, not complete.

What is remarkable is that the observed nonmonotonic temperature dependence of the gap up to the temperature slightly above T^* reveals one-to-one correspondence to the analogous quantity measured for $TaSe_2$ (Fig. 3, right panel), although there is a notable difference at higher temperature (above 220 K), which we will discuss later. In the range between 100 and 220 K, the correspondence appears in the following: (1) the transitions at T_{NIC} and T^* are similarly sharp (the derivative $d\Delta/dT$ has a finite jump); (2) the gap decreases linearly from T_c to T^* like from T_{ICC} to T_{NIC}; and (3) the gap does not close at T^* and T_{NIC}.

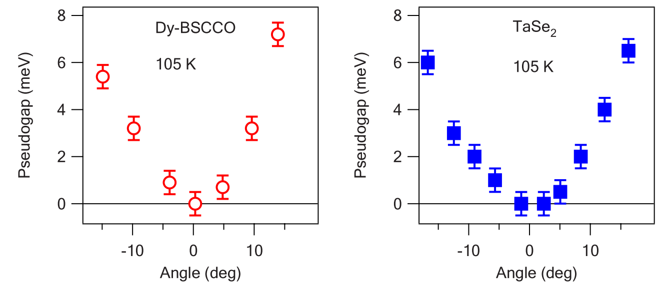

FIG. 4. (Color online) Momentum anisotropy of the pseudogap. The dependences of the gap on the Fermi surface as a function of angle in Dy-BSCCO in the pseudogap state (left) and in $TaSe_2$ in the incommensurate CDW state (right) are almost identical.

It is important to stress that the discovered correspondence in temperature evolution of the gap in BSCCO and $TaSe_2$ completes the overall similarity of the spectral functions of these two classes of compounds. The depletion of the spectral weight at the Fermi level is really partial and changes smoothly with binding energy; there is neither bending nor backfolding of the experimental dispersion (the Bogoliubov-type dispersion predicted by the BCS theory) in the PG state as it is seen in the superconducting state of cuprates[12,19] or in the commensurate CDW state of $TaSe_2$ (Ref. 15) (to compare the superconducting and PG ARPES spectra of BSCCO, see Ref. 20). Finally, the strong d-wave-like anisotropy of the PG cannot be considered peculiar to HTSC since similar dependence has been observed for $TaSe_2$ (Ref. 15) that is illustrated by Fig. 4. This allows one to conclude that the cuprates and the dichalcogenides in the PG state reveal virtually identical spectra of one-particle excitations as a function of energy, momentum, and now temperature.

Fortunately, the physical reason for the formation of the spectral function in question is well known for $TaSe_2$—it is the incommensurate CDW state. According to neutron-scattering experiments,[21] $TaSe_2$ at T_{NIC}=122 K undergoes the transition into the incommensurate CDW state which turns to the commensurate CDW at T_{ICC}=90 K. Therefore, between T_{ICC} and T_{NIC}, the observed gap is the incommensurate band gap (IBG), the spectroscopic appearance of which is not expected to be trivial and has been addressed theoretically only recently.[22] The fortunate situation with $TaSe_2$ is that one can conclude from direct ARPES experiment that the IBG opens linearly with lowering T, as it is seen from the left panel of Fig. 3.[15] A similar scenario can be valid for the cuprates, with the only difference that the superconductivity happens before the charge ordering becomes commensurate and the high-T_c compounds, due to superconducting renormalization of the electronic structure, escape the commensurate charge ordering. There is one known exception to this trend, the $La_{2-x}Ba_xCuO_4$ (LBCO) compound with 1/8 doping level,[23] where the charge ordering suppresses the superconductivity completely.[14]

To advocate the discussed analogy we note that there is a number of observations of the charge or spin ordering in the hole-doped cuprates: the "checkerboard" pattern by tunneling[24] and ARPES (Ref. 25) experiments, the spatial modulation of spin/charge density by neutron scattering,[23] and the charge-density modulation by x-ray scattering.[26] Very recently, the idea of density-wave phase has been supported by careful measurements of the Hall effect.[27]

The finite gap above T^* in BSCCO, such as the one above T_{NIC} in $TaSe_2$, can be partially explained either by the fluctuating incommensurate charge ordering[5] or by momentum-dependent scattering.[28] On the other hand, a gradual but robust increase in the gap in BSCCO above 250 K implies a certain difference between cuprates and dichalcogenides and finds a natural explanation in terms of metal-to-insulator crossover which is predicted to be a generic feature of the phase diagram of strongly correlated electrons at high temperatures.[16] This scenario is in agreement with recent observation of coherent-incoherent crossover in $La_{2-x}Sr_xCuO_4$ (LSCO).[29]

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