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Pseudogap suppression by competition with superconductivity in La-based cuprates

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We carried out a comprehensive high-resolution angle-resolved photoemission spectroscopy (ARPES) study of the pseudogap interplay with superconductivity in La-based cuprates. The three systems $La_{2-x}Sr_xCuO_4$, $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$, and $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$ display slightly different pseudogap critical points in the temperature versus doping phase diagram. We studied the pseudogap evolution into the superconducting state for doping concentrations just below the critical point. In this setting, near optimal doping for superconductivity and in the presence of the weakest possible pseudogap, we uncover how the pseudogap is partially suppressed inside the superconducting state. This conclusion is based on the direct observation of a reduced pseudogap energy scale and re-emergence of spectral weight suppressed by the pseudogap. Altogether these observations suggest that the pseudogap phenomenon in La-based cuprates is in competition with superconductivity for antinodal spectral weight.

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I. INTRODUCTION

Strange metal behavior [1] and pseudogap physics [2] remain the most challenging problems of the cuprate superconductors. One key characteristic of strange metals is that resistivity scales uninterrupted with thermal excitation energy down to the lowest measurable temperature. In the cuprates, this is observed at a critical doping p^* [3,4]. Above p^*, standard Fermi liquid properties are restored [5], whereas below it, a mysterious pseudogap phenomenon emerges. The pseudogap manifests in multiple experiments. In spectroscopic measurements, the pseudogap is associated with a partial gapping of spectral weight near the Fermi level [6]. Numerous studies attempted to address the nature of the pseudogap [2]. These experiments are typically carried out in the normal state, aiming to connect the pseudogap to either superconducting fluctuations [7], a symmetry breaking order parameter [8–10], or a cross-over phenomenon [11]. There is much less experimental work addressing the pseudogap inside the superconducting state [12]. In the very underdoped regime, photoemission studies point to a competition between the pseudogap phenomenon and superconductivity — with the latter being partially suppressed by the former [13]. It has, however, been difficult to tune or influence the pseudogap, which appears rather insensitive to disorder or magnetic field [14,15]. Theoretical and experimental work suggests that the pseudogap critical point p^* is confined by the van Hove singularity crossing of the Fermi level [16,17].

Tuning the van Hove singularity by hydrostatic pressure is one way to manipulate the pseudogap phenomenon [18]. Another route is to identify interactions with other phases. The pseudogap has been shown to suppress the superconducting order parameter [13]. Much less is known about the reciprocal relation, namely, how superconductivity influences the pseudogap phenomenon. An unsolved problem relates to the interplay between the pseudogap and superconductivity in the regime close to p^*. This issue has been difficult to address since it challenges both temperature and energy resolution limitations of most synchrotron angle-resolved photoemission spectroscopy (ARPES) [19] beamlines.

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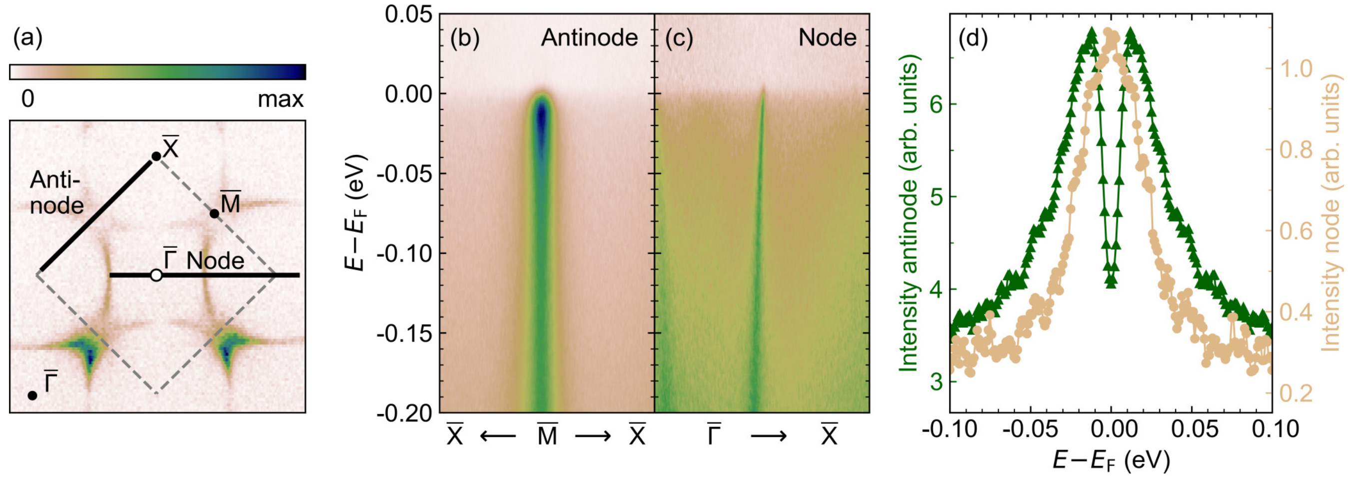

FIG. 1. Photoemission intensities recorded on LSCO $p = 0.145$ in the superconducting state. (a) Fermi surface map, recorded with $h\nu = 160$ eV photons at $T = 7$ K, and integrated ± 16 meV around the Fermi level. The Brillouin zone boundary is indicated by the dashed grey lines and high symmetry points are labeled $\overline{\Gamma}$, $\overline{\mathrm{X}}$, and $\overline{\mathrm{M}}$. Solid black lines indicate nodal and antinodal directions. (b,c) ARPES spectra recorded at $T = 6$ K along the antinodal ($\overline{\mathrm{M}} \to \overline{X}$) and nodal ($\overline{\Gamma} \to \overline{\mathrm{X}}$) directions using $h\nu = 55$ eV photons. (d) Nodal (beige circles) and antinodal (green triangles) symmetrized energy distribution curves (EDCs) at the Fermi momentum, after background subtraction as described in Ref. [20].

Here, we study the pseudogap in the limit $p \to p^*$, where T^* approaches T_c. We chose to examine La-based cuprates in which the pseudogap energy scale is much larger than the superconducting gap amplitude [19,21]. Investigating these compounds, where $\Delta^* \gg \Delta_{sc}$, enabled us to track both, the "pure" pseudogap as well as its interplay with superconductivity. Using state-of-the-art low-temperature and high-energy resolution ARPES beamlines, we explored the evolution of the pseudogap inside the superconducting state of $La_{2-x}Sr_xCuO_4$ (LSCO) $x = p = 0.145$, $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$ (Eu-LSCO) $p = 0.21$, and $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ (Nd-LSCO) $p = 0.20$. We find that the pseudogap amplitude is partially suppressed inside the superconducting state, suggesting a competing interaction. As a defining property of the pseudogap phase, we observe an antinodal spectral weight suppression for $T < T^*$. Below T^*, we identify a third temperature scale $T^\dagger > T_c$, below which antinodal weight partially recovers. Eventually, complete recovery is found for $T \to 0$. This spectral weight recovery is discussed in the context of a triphase competition between superconductivity, charge order, and pseudogap physics.

II. METHODS

Single crystals of LSCO ($p = 0.145$, $T_c = 37$ K [22,23] and $p = 0.12$, $T_c = 27$ K [24]), Nd-LSCO ($p = 0.20$, $T_c = 20$ K) [21], and Eu-LSCO ($p = 0.21$, $T_c = 15$ K) were synthesized by the traveling floating zone method. The critical pseudogap dopings are $p^* \approx 0.19$ [4] for LSCO and $p^* \approx 0.23$ [25,26] for Nd-LSCO and Eu-LSCO. ARPES experiments were carried out at beamline I05 [27] of the Diamond Light Source and the Surface and Interface Spectroscopy (SIS) beamline of Swiss Light Source. Single crystals were mechanically cleaved *in situ* in an ultrahigh vacuum using top posts. Measurements were performed using 55 eV or 160 eV linear-horizontally polarized light at I05 and circularly polarized light at SIS. Dependent on photon energy and instrument, the energy resolution (Gaussian standard deviation) spans the range of 5 to 15 meV.

III. RESULTS

We studied three different La-based compounds (LSCO, Nd-LSCO, and Eu-LSCO). Consistent with existing ARPES literature, the data quality obtained from LSCO and Nd-LSCO crystals [21,29] is comparatively better than that recorded on Eu-LSCO [30]. In Figs. 1(a) to 1(c), we display a Fermi surface map, nodal, and antinodal spectra recorded on LSCO $p = 0.145$. Symmetrized [31] nodal and antinodal energy distribution curves (EDCs) at k_F are shown in Fig. 1(d). These results are directly comparable to a previous study of this compound [22]. The improved data quality stems from higher energy resolution and smaller beam spot. These advances result in a higher signal-to-background ratio that we exploit to study the pseudogap phenomenon. Antinodal spectra recorded on Nd-LSCO $p = 0.20$ are depicted in Fig. 2 for temperatures as indicated. The pseudogap spectra $T_c < T < T^*$ were discussed in a previous publication [21]. Here, we also enter the superconducting state. From the raw energy distribution maps of Nd-LSCO shown in Figs. 2(a) and 2(b), it is directly visible that the spectral gap at 22 K, just above T_c, is larger than that inside the superconducting state. This is further confirmed by analyzing the EDCs at the underlying Fermi momentum k_F — see Figs. 2(c) and 2(d). The symmetrized antinodal EDCs display an effectively smaller gap inside the superconducting state than what is observed for $T \approx T_c$. The results on Eu-LSCO $p = 0.21$ reveal the increase of the antinodal spectral weight inside the superconducting state while no such gain is detectable at the nodal point — see Fig. 2(e). Both observations suggest a weakening of the pseudogap.

The Nd-LSCO and Eu-LSCO compounds (space group 138 [33]) are special because they have additional chemical disorder due to the substitution of neodymium and europium. This substitution stabilizes the so-called low-temperature

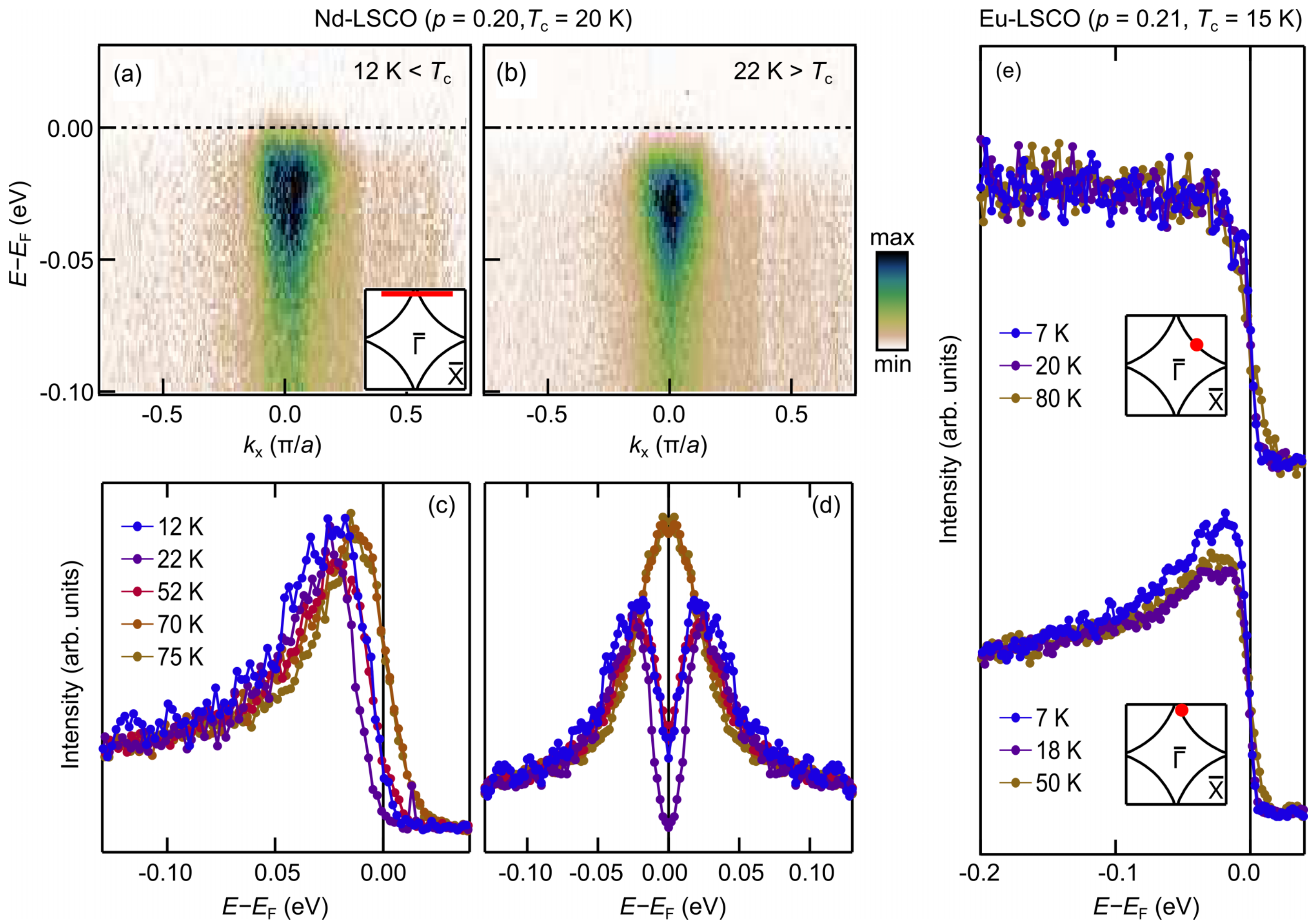

FIG. 2. Temperature dependence of nodal and antinodal spectra in Nd-LSCO ($p = 0.20$) and Eu-LSCO ($p = 0.21$) at $h\nu = 55$ eV. (a,b) Energy distribution maps along the antinodal direction (see inset), recorded on Nd-LSCO [21] for temperatures as indicated. (c,d) Corresponding energy distribution curves (EDCs) and symmetrized EDCs at the underlying Fermi momentum. (e) Temperature-dependent nodal and antinodal spectra recorded on Eu-LSCO. Pseudogap temperatures for Nd-LSCO $p = 0.20$ and Eu-LSCO $p = 0.21$ are $T^* \approx 80$ K and $T^* \approx 75$ K [21,28]. The EDCs in (c)–(e) are normalized to the respective high-energy tail.

tetragonal phase. We, therefore, additionally investigated the LSCO $p = 0.145$ compound that has less chemical disorder and a different crystal structure (space group 64 [34–36]). The larger T_c of this compound allowed to probe deep into the superconducting state. Background-subtracted [20] antinodal EDCs are shown in Fig. 3(a) as a function of temperature. The pseudogap opens at $T^* \approx 162.5$ K. As in all other hole-doped cuprates, the pseudogap manifests itself by a loss of spectral weight near the Fermi level. Upon cooling, the weight loss gradually increases. In Fig. 3(b), the antinodal spectrum at $T = 45$ K, just above the superconducting transition, is shown. We find a pronounced re-emergence of antinodal spectral weight inside the superconducting state, as exemplarily shown by the spectrum taken at $T = 7$ K [see Fig. 3(b)]. This is in strong contrast to the nodal spectra, which are essentially temperature independent [Fig. 3(c)].

Complementary to our observation of spectral weight loss, we find a peculiar temperature dependence of the pseudogap. The vertical dashed lines in Fig. 3(b) indicate the peak positions in the symmetrized antinodal EDCs. Defining the gap amplitude by half the distance between the peaks yields a reduction of the gap amplitude for $T \ll T_c$.

IV. ANALYSIS

A. Spectral weight

A defining property of the pseudogap is the partial suppression of the antinodal spectral weight $I(k_{AN}, \omega)$. We define the integrated spectral weight as $W_i = \int d\omega[I(k_i, \omega) + I(k_i, -\omega)]$ with $i = AN, N$ being antinodal or nodal. Our integration window of the symmetrized EDCs is set to $-0.2 < \omega < 0.2$ eV [37]. The nodal spectral weight W_N is essentially temperature independent [see circles in Fig. 4(a) and Figs. 2(e) and 3(c)]. In contrast, antinodal spectral weights display a significant temperature dependence, as shown by the rectangles in Figs. 4(a) and 4(b). When entering the pseudogap state, W_{AN} is suppressed and gradually diminishes upon cooling. However, for Eu-LSCO, Nd-LSCO, and LSCO, a gradual recovery is observed below a temperature scale $T^\dagger$. In LSCO $p = 0.12$ and 0.145, $T^\dagger \sim 2$–$3\,T_c$ while in Eu-LSCO $p = 0.21$, $T^\dagger$ is closer to T_c. The spectral weight recovery continues inside

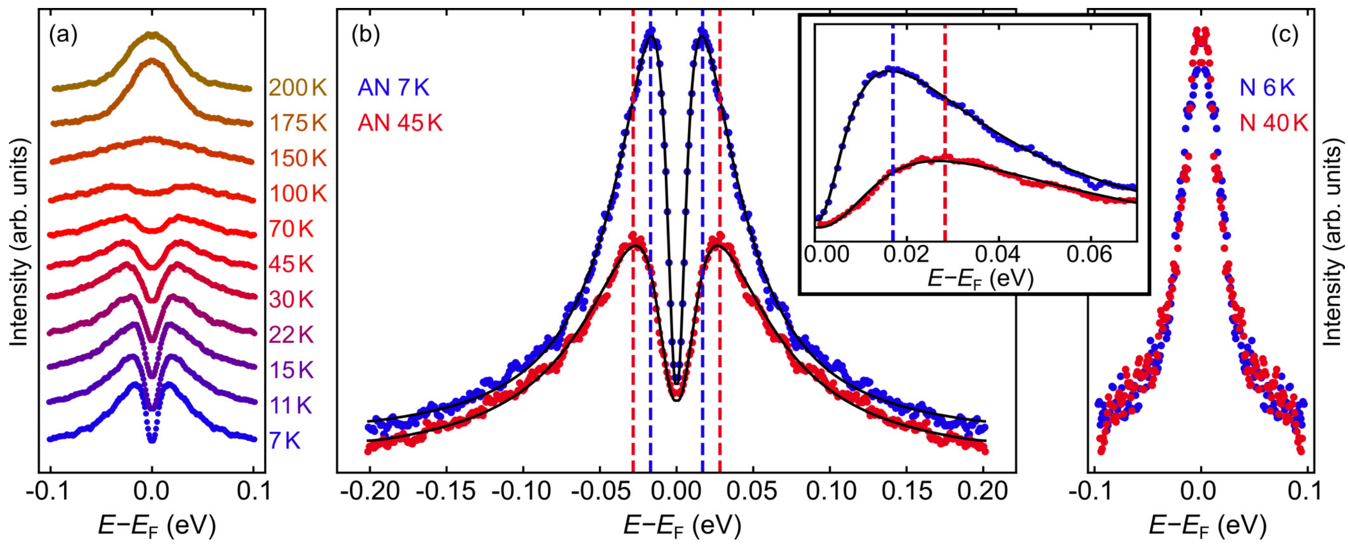

FIG. 3. Nodal and antinodal spectra versus temperature in LSCO $p = 0.145$ ($T_c = 37$ K) at $h\nu = 55$ eV. (a) Symmetrized antinodal spectra for temperatures as indicated. The pseudogap onset temperature of $T^* \approx 162.5$ K, deduced from spectral weight analysis of the EDCs, is consistent with that extracted from transport measurements [28]. (b, c) Comparison of symmetrized antinodal and nodal spectra for $T \ll T_c$ (blue) and $T \gtrsim T_c$ (red). Solid black lines are fits using a phenomenological self-energy function [32] — see text. Vertical dashed lines indicate the peak position. The inset displays a zoom of the low-energy part of the symmetrized EDCs in (b). A background defined using the methodology given in Ref. [20] was subtracted from all spectra.

the superconducting state. Eventually, as $T \to 0$, W_{AN} fully recovers to the level of weight observed above $T \leqslant T^*$.

B. Gap analysis

To extract the amplitude of the antinodal spectral gap, we employ the spectral function $A(k_F, \omega) = -\pi^{-1}\Sigma''/[(\omega - \Sigma')^2 + \Sigma''^2]$ convoluted with a Gaussian distribution to mimic the experimental resolution [32]. Σ' and Σ'' are the real and imaginary parts of the self-energy, respectively. Our problem involves at least a superconducting gap and a pseudogap. Since the nature of the pseudogap is not established, a microscopic understanding of its self-energy is missing. Eliashberg theory, in contrast, describes the self-energy effect associated with superconductivity [38]. If the pseudogap was a precursor to superconductivity, the Eliashberg framework would also apply to the pseudogap state.

There are several experimental indications that the pseudogap is not associated with superconducting fluctuations [6,39]. In Nd-LSCO, for example, there is strong evidence of vanishing of the pseudogap at a quantum critical point inside the superconducting dome [3]. In Nd-LSCO $p = 0.20$, we observe a spectral gap at the superconducting onset $T_c = 20$ K. Mean-field theory yields $2\Delta = \alpha k_B T_c$ where $\alpha = 4.3$ [40] for weakly coupled d-wave superconductors. The gap amplitude of 20 to 25 meV [Fig. 4(c)] implies $\alpha \sim 20$. Although the coupling coefficient α can be larger in the strong coupling limit, this appears unreasonably large. In contrast, assigning the gap to the pseudogap onset temperature ($2\Delta = \alpha k_B T^*$) yields a more reasonable $\alpha \approx 5$. We thus associate the observed spectral gap with the pseudogap phenomenon and assume that the superconducting gap is not directly detectable due to the finite energy resolution. In LSCO, the differentiation of the pseudogap and superconducting gap is less obvious. Around optimal doping, the pseudogap energy scale is smaller than that in Nd-LSCO. At the same time, the superconducting gap is expected to be larger due to its larger T_c. Within the experimental resolution, it was not possible to differentiate these two gaps. The antinodal spectra of LSCO are fitted using a single gap model. Using the phenomenological ansatz [32] $\Sigma'' = -\Gamma =$ constant and $\Sigma' = \Delta^2/\omega$, the spectral weights of LSCO and Nd-LSCO can be parametrized. With this function, a single gap energy scale is extracted as a function of temperature [see Fig. 4(c)]. In the pseudogap, the gap follows roughly an order parameter like $(1 - T/T^*)^{0.5}$ dependence. The temperature dependence is interrupted for $T < T_c$, where the amplitudes of the gaps decrease with decreasing temperature. Thus, when the pseudogap is analyzed, either by spectral weight or gap amplitude, suppression is observed below two different temperature scales. The gap amplitude is partially suppressed upon entering the superconducting state, whereas spectral weight decreases below T^*. Recovery is found below a temperature scale $T^\dagger$ much larger than T_c.

V. DISCUSSION

It is interesting to compare our results on La-based cuprates with previous ARPES studies in $(Bi,Pb)_2(Sr,La)_2CuO_{6+\delta}$ (Bi2201) [13] and $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) [41]. In all systems, the onset of the pseudogap is heralded by the suppression of W_{AN} and the opening of an antinodal spectral gap. This conclusion holds even if slightly different definitions of the integrated spectral weight are employed. Upon further cooling, W_{AN} diminishes, and the pseudogap energy scale increases. However, the three systems react differently upon approaching the superconducting state. In Bi2212, the recovery of W_{AN} has a sharp onset at the superconducting transition [41,42]. For LSCO and Eu-LSCO, the recovery of W_{AN} starts already below a temperature scale $T^\dagger > T_c$ — as

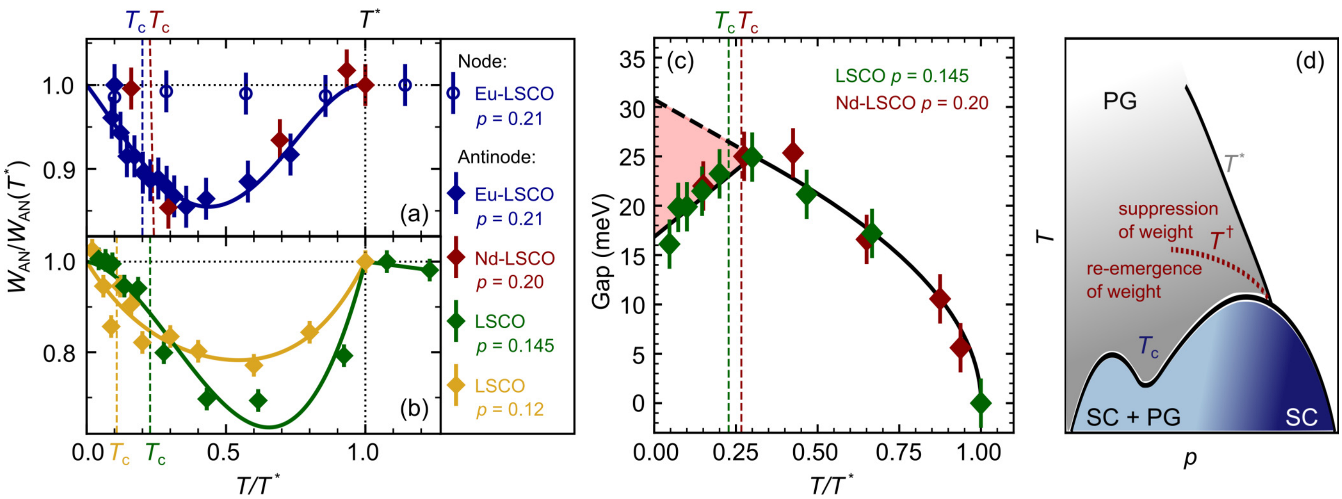

FIG. 4. Antinodal competition between the pseudogap and superconductivity. (a,b) Integrated spectral weight as a function of temperature. Antinodal weights of LSCO, Eu-LSCO, and Nd-LSCO are plotted with rectangles whereas open circles denote nodal weight. Solid lines are guides to the eye. Vertical dashed lines indicate $T = T^*$ and T_c/T^*, respectively. Error bars provide an estimate of the systematic uncertainty. (c) Antinodal spectral gap as a function of temperature for LSCO (this work) and Nd-LSCO [21]. The gap amplitude follows a linear dependency for $T \lesssim T_c$ and can be fitted by an order parameter like $(1 - T/T^*)^{0.5}$ behavior for $T \gtrsim T_c$. The energy resolution defined by the standard deviation of the Fermi step sets the error bars. (d) Phase diagram (temperature versus doping) indicating phase space of different antinodal spectral weight behavior. Outside the pseudogap spectral weight is conserved. We show that there exists a temperature scale $T^\dagger < T^*$ below which spectral weight is partially recovered.

schematically illustrated in Fig. 4(d). If the recovery is interpreted in terms of phase competition with superconductivity, it must involve superconducting fluctuations in the normal state. Although such superconducting fluctuations are known to exist [7,28,39,43,44], it is not obvious that they would impact the pseudogap stronger in La-based cuprates. Another possibility is that the charge ordering [45–48] competes with the pseudogap. Charge order is expected to generate an additional temperature and energy scale. The energy scale was reported in the normal state of $YBa_2Cu_3O_{7-x}$ (YBCO) and Bi2212 [49]. A triphase competition [50,51] between the pseudogap, charge (stripe) order, and superconductivity is likely expressed differently in La- and Bi-based cuprates, explaining the different phenomenology in different cuprate systems.

Inside the superconducting state, the systems also behave differently. Both Bi2201 and Bi2212 display so-called coherence peaks [52–54]. In Bi2212, the coherence peak associated with superconductivity appears at an energy scale smaller than the pseudogap [52]. For Bi2201, on the contrary, the two energy scales are comparable [13]. Certainly, for Nd-LSCO $p = 0.20$, as discussed above, we expect the superconducting energy scale to be much smaller than the pseudogap. This aligns with the fact that no superconducting coherence peak is observed. It is difficult to distinguish the changes in spectral weight associated with superconductivity and the pseudogap state. Generally, superconductivity is expected to redistribute spectral weight from below to above the cooper-pairing energy scale [13]. No net gain or loss of spectral weight is expected from the emergence of superconductivity. The recovery of W_{AN} suggests a competing interaction between superconductivity and the pseudogap state [see Figs. 4(a) and 4(b)]. This interpretation is further reinforced by the observation of a diminishing pseudogap energy scale inside the superconducting state.

VI. CONCLUSION

In summary, we carried out a high-resolution ARPES study of the pseudogap inside the superconducting state. Three La-based systems (Eu-LSCO, Nd-LSCO, and LSCO) were investigated for doping concentrations just below the critical doping p^*, above which the pseudogap phenomenon vanishes. We observe that the total antinodal spectral weight is suppressed below the pseudogap temperature and begins to recover already above the superconducting onset T_c. The pseudogap energy scale grows with decreasing temperature until T_c is reached and is partially suppressed inside the superconducting state. Our results are different from what was reported in single- [13] and bi-layer [41,42] Bi-based cuprates. We interpret this as a tri-interaction between superconductivity, charge order, and pseudogap physics that is expressed differently across material systems.

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