

# Energy gaps in the failed high- $T_c$ superconductor $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$

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**A central issue in high- $T_c$  superconductivity is the nature of the normal-state gap (pseudogap)<sup>1</sup> in the underdoped regime and its relationship with superconductivity. Despite persistent efforts, theoretical ideas for the pseudogap evolve around fluctuating superconductivity<sup>2</sup>, competing order<sup>3–8</sup> and spectral weight suppression due to many-body effects<sup>9</sup>. Recently, although some experiments in the superconducting state indicate a distinction between the superconducting gap and pseudogap<sup>10–14</sup>, others in the normal state, either by extrapolation from high-temperature data<sup>15</sup> or directly from  $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$  (LBCO-1/8) at low temperature<sup>16</sup>, suggest the ground-state pseudogap is a single gap of  $d$ -wave<sup>17</sup> form. Here, we report angle-resolved photoemission data from LBCO-1/8, collected with improved experimental conditions, that reveal the ground-state pseudogap has a pronounced deviation from the simple  $d$ -wave form. It contains two distinct components: a  $d$ -wave component within an extended region around the node and the other abruptly enhanced close to the antinode, pointing to a dual nature of the pseudogap in this failed high- $T_c$  superconductor that involves a possible precursor-pairing energy scale around the node and another of different but unknown origin near the antinode.**

The first high- $T_c$  superconductor discovered,  $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$  (LBCO), holds a unique position in the field because of an anomalously strong bulk  $T_c$  suppression near  $x = 1/8$ . Right around this ‘magic’ doping level, scattering experiments by neutrons<sup>18,19</sup> and X-rays<sup>20</sup> find a static spin and charge (stripe) order. By itself, this observation raises a series of intriguing questions: whether the stripe order is a competing order that suppresses the superconductivity in LBCO-1/8; if the answer is positive, which aspect, the pairing strength or the phase coherence, is involved in the  $T_c$  suppression and how this mechanism applies to other dopings or families. For our investigation of the ‘ground-state’ pseudogap, as defined in ref. 16, which ignores the residual superconductivity, its sufficiently high doping yet extremely low bulk  $T_c$  ( $\sim 4\text{K}$ ) makes LBCO-1/8 an ideal system: especially for the small-gap measurement near the node, difficulties due to either the unscreened disorder potential, a problem for extremely low doping, or trivial thermal broadening, a problem above  $T_c$  for higher doping, are circumvented.

Whereas thermal effects require an extrapolation from high-temperature data to obtain the ground-state physics<sup>15</sup>, a direct

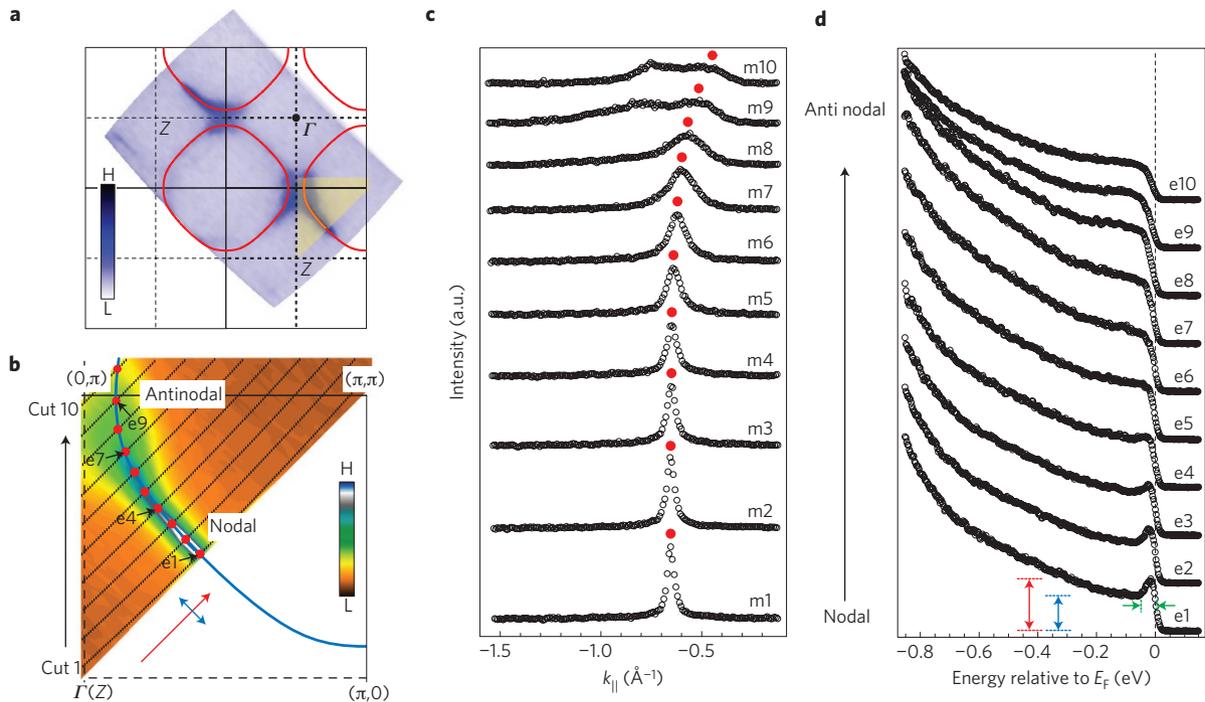
measurement on LBCO-1/8 at low temperature has been made<sup>16</sup>. With experimental resolutions compromised to obtain sufficient signal to noise, a simple  $d$ -wave gap function is reported with no discernible nodal quasi-particles found. Given the importance of this issue, we have carried out an angle-resolved photoemission<sup>21</sup> study of LBCO-1/8 at  $T > T_c$  with much improved resolutions in a measurement geometry favourable for the detection of nodal quasi-particles (see Fig. 1 and Supplementary Information, Section SI). Our data provide two important new insights. First, there is a well-defined nodal quasi-particle peak, suggesting nodal quasi-particles can exist in the stripe-ordered state. Second, there is a rich gap structure, suggesting the pseudogap physics is more elaborate than the simple  $d$ -wave version. In particular, a new kind of pseudogap, which is not smoothly connected to the usual one tied to the antinodal region, can exist in the nodal region when superconductivity is suppressed owing to the loss of phase coherence.

As shown in Fig. 1, there exists a well-defined quasi-particle peak in the energy distribution curves (EDCs) at the Fermi crossing points ( $k_F$ ) around the node. On dispersion towards the antinode, the line shape quickly becomes incoherent. Data taken with different photon energies ( $h\nu$ ) in different Brillouin zones show consistent results (see Supplementary Information, Fig. S1), providing the first unambiguous piece of direct evidence that nodal quasi-particles survive in the stripe-ordered state. This suggests that these two seemingly very different aspects of cuprate phenomenology can be compatible with each other<sup>22</sup>.

The observation of quasi-particle peaks in the EDCs gives a firm foundation for the gap analysis around the node. However, given the small gap size as well as a relatively small peak-background intensity ratio and large quasi-particle peak linewidth ( $\Gamma$ ) (Fig. 1d), quantitative determination of the gap is non-trivial in LBCO-1/8. In the following, we will present an analysis that addresses two important aspects of our data: (1) the gap measured by the leading edge gap (LEG) method, the same as that used in ref. 16, is shown to have two components and cannot fit a simple  $d$ -wave form (Fig. 2); (2) the gap around the node, measured either by the LEG method (Fig. 2) or by a curve fitting procedure commonly used in the field (Fig. 3), is shown to be  $d$ -wave-like with a finite gap slope.

In Fig. 2a, from the systematic shift of the leading-edge midpoint (LEM) along the energy axis, we notice that the LEG keeps increasing from the node towards the antinode with the rate

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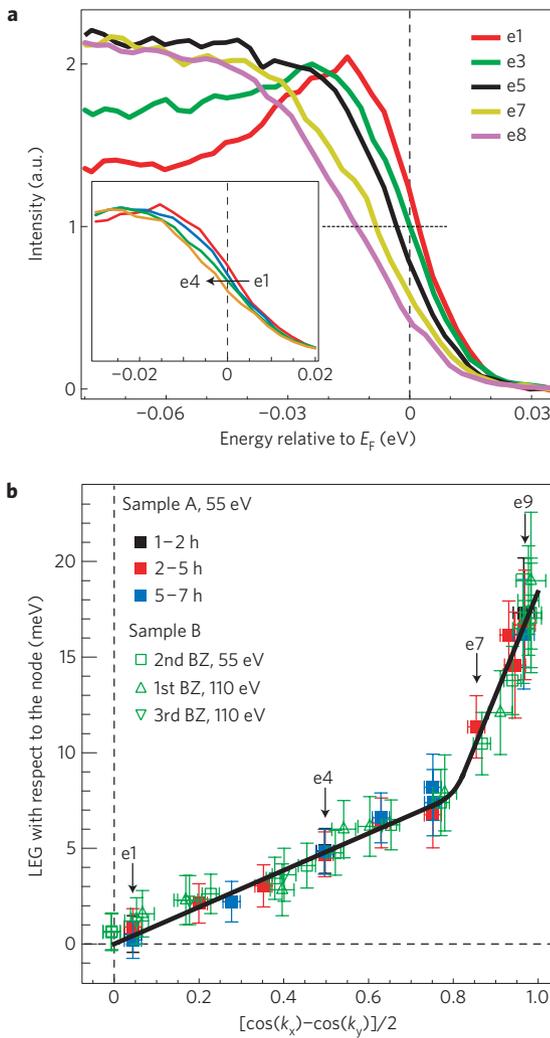
**Figure 1 | Angle-resolved photoemission spectra and the Fermi surface of LBCO-1/8.** All data were taken on sample A with  $h\nu = 55$  eV at  $T = 21 \pm 2$  K (see Supplementary Information, Fig. S1 for data on sample B with  $h\nu = 110$  eV). **a**, A coarse Fermi surface map having a large momentum coverage. **b**, A fine Fermi surface map taken within 2–5 h after sample cleaving, covering the yellow-shaded region mostly in the second Brillouin zone for the detailed spectral analysis. Cuts were taken parallel to the zone diagonal (red arrow) with the polarization of light fixed in plane and orthogonal to the zone diagonal (blue arrow). Each dot of the dotted lines corresponds to an actual sampling momentum position by that cut. The red curves in **a** and blue curves in **b** represent the same tight-binding Fermi surface resulting from a global fit to the data (see Supplementary Information, Fig. S2). **c**, The MDCs at  $E_F$ , m1–m10, of cuts 1–10 from the nodal to the antinodal as shown in **b**.  $k_F$  determined are indicated by red dots (see Supplementary Information, Section SIIA and Fig. S9). Note that  $k_F$  on cut 10 is already past the zone boundary. The parallel momentum coverage of each MDC does not correspond to the length of that cut shown in **b**. The extra peak features in m9 and m10 are due to the Fermi crossings in the adjacent quadrant of the Brillouin zone as shown in **a**. **d**, The EDCs at  $k_F$ , e1–e10, of cuts 1–10, respectively. Red, blue and green arrows indicate the peak intensity, background and peak linewidth in e1, respectively. All spectra are offset vertically for clarity. Labels e1, e4, e7 and e9 in **b** denote the momentum positions of the corresponding EDCs. See Supplementary Information, Section SI for further information.

of increase getting larger close to the antinode. This is more clearly shown in Fig. 2b, where the momentum dependence of the extracted LEG from two selected samples is plotted. Consistent results are obtained at different times after sample cleaving, showing no signs of sample ageing, which could affect small-gap measurements, and in different Brillouin zones with different  $h\nu$  on another sample. They unanimously point to a pronounced deviation of the gap function from the simple  $d$ -wave form. As detailed in Supplementary Information, Section SIIIB,C,SIV, this observation goes beyond the interpretation based on either a simple  $d$ -wave gap under the influence of experimental resolutions (see Supplementary Information, Fig. S10) and the line-shape decoherence towards the antinode (see Supplementary Information, Fig. S11) or a single pairing gap with the inclusion of higher  $d$ -wave gap harmonics<sup>23</sup> (see Supplementary Information, Fig. S7). It naturally reveals a striking characteristic: this normal-state gap has two distinct components, with the one around the node (the nodal gap) exhibiting a simple  $d$ -wave form, that is, a linear dependence with respect to  $[\cos(k_x) - \cos(k_y)]/2$ , along the underlying Fermi surface over a significant momentum range and the other setting in near the antinode (the antinodal gap), which deviates sharply from its nodal counterpart.

Although it is clear that the LEG opens in a  $d$ -wave fashion around the node (Fig. 2a, inset), it is too crude to conclude that the real gap ( $\Delta$ ) function of the system is also simple  $d$ -wave-like because the leading edge shift in principle can be due to the variation in  $\Gamma$  even if  $\Delta$  is fixed<sup>24</sup>. This alternative has to be

explored especially for LBCO-1/8, where  $\Gamma \gg \Delta$  near the node. Thus, we fit the  $E_F$ -symmetrized EDCs at  $k_F$  to a phenomenological model<sup>25</sup> that assumes a self-energy,  $\Sigma(k_F, \omega) = -i\Gamma + (\Delta^2/\omega)$  (Fit Model), where  $\Gamma$  and  $\Delta$  are subject to the fit (Fig. 3a). Because of the small peak–background ratio, the fitting results are affected by the background subtraction. In Fig. 3b, we show the results of  $\Delta$  without any background subtraction, or with a momentum distribution curve (MDC) constant background or an EDC integral background subtraction (see Supplementary Information, Fig. S5). Two general trends of the results are: (1) regardless of the methods for background subtraction, an initial gap slope is robustly defined close to the node where quasi-particle peaks are clearly present in e1–e3; (2) as the peak feature weakens, a large background dependence appears starting from e4. For e4, with more background subtracted from the high-binding-energy side of the peak feature,  $\Delta$  decreases and tends to fall onto the initial gap slope. For the completeness of our analysis, we have also shown in Supplementary Information, Fig. S6 fitting results based on another model that fits the data less well. Although the quantitative gap values are model dependent, its  $d$ -wave form remains robust (see Supplementary Information, Section SIID).

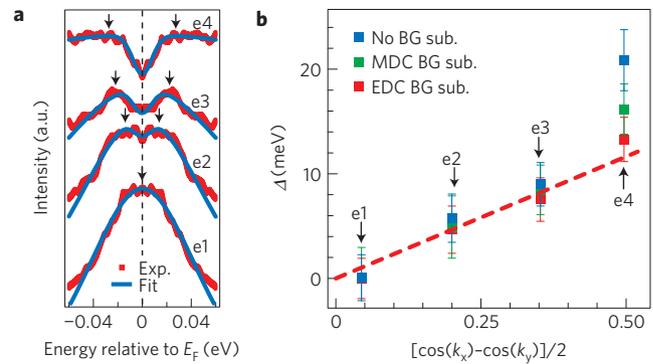
Summarizing the above, both the model-independent (the LEG method) and model-dependent (the curve fitting procedure) gap analysis suggest that the nodal gap opens in a  $d$ -wave fashion with a finite slope. Despite its presence in the normal state, it is highly suggestive that this nodal gap is of a pairing origin based on the following reasons. (1) Its  $d$ -wave form is consistent with the



**Figure 2 | The pseudogap function of LBCO-1/8 by the LEG analysis.**

**a**, Selected EDCs at  $k_F$ , e1, e3, e5, e7 and e8, reproduced from Fig. 1d in an expanded energy scale that are normalized in intensity at the LEM (see Supplementary Information, Figs S3,S4). Inset: e1-e4 similarly normalized and shown in a more expanded energy scale to visualize the LEG opening near the node. **b**, LEG plotted as a function of  $[\cos(k_x) - \cos(k_y)]/2$ , for LBCO-1/8 sample A measured at different times after sample cleaving with  $h\nu = 55$  eV and for sample B measured in different Brillouin zones (BZs) with  $h\nu = 55$  and 110 eV.  $T = 21 \pm 2$  K. The results regarding the LEG function at  $T \sim 20$  K were repeated on sample C (Fig. 4b) and another six samples from different batches of growth (not shown). Labels e1, e4, e7 and e9 denote the momentum positions of the corresponding EDCs in Fig. 1d. The black line is a guide to the eye for the ground-state pseudogap function. The LEG values for cuts at different momentum positions for each sample are referenced to the value from the nodal reference cut taken right after that cut. Error bars are determined by the uncertainty of  $E_F$ ,  $k_F$  and the energy window dependence of the LEG by the LEG analysis (see Supplementary Information, Section SIIB).

generic pairing symmetry of cuprate superconductors, particularly in La-based cuprates, where a similar gap in the nodal region is found to be related to superconductivity<sup>12</sup>. (2) Its gap slope strikingly coincides within error bars with those of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (LSCO)  $x = 0.110$  (ref. 26) and LBCO  $x = 0.083$  (ref. 27) ( $T_c = 26$  K and 23 K, respectively, see Supplementary Information, Fig. S8 for raw data), which are both in the superconducting state (Fig. 4a). Compared with the one analysed using Fit Model in Bi2212 of a similar doping level well below  $T_c$  (Fig. 3c in ref. 11), it

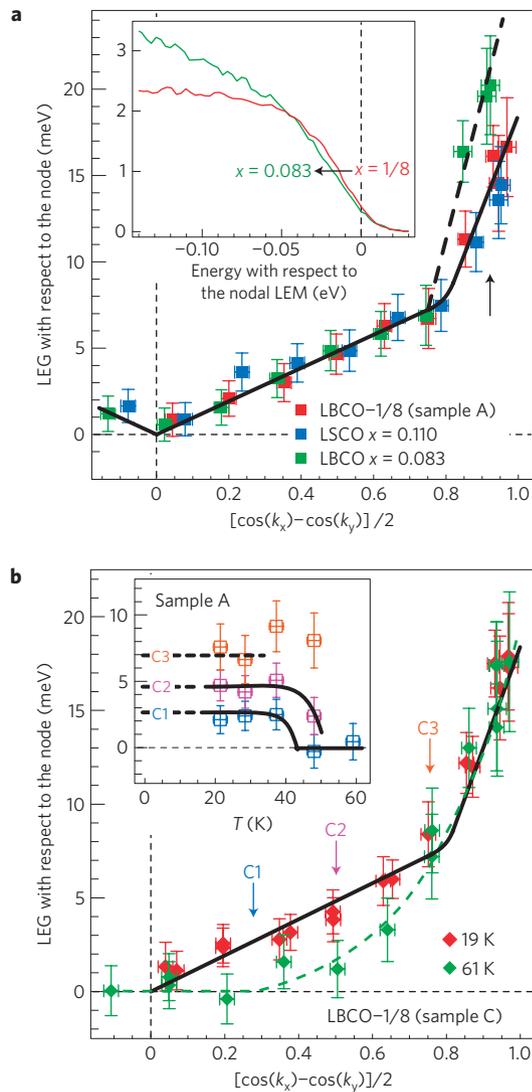


**Figure 3 | The nodal gap analysis by curve fitting.** **a**, The  $E_F$ -symmetrized EDCs near the node, e1-e4, and fit curves by Fit Model. The arrows are very rough guides to the eye for the centroids of possible peak features in the EDCs. **b**,  $\Delta$  from different fits without/with different methods of background subtraction (BG sub.; see Supplementary Information, Section SIIC). Labels e1-e4 denote the momentum positions of the corresponding EDCs in **a**. The dashed line is a guide to the eye showing the initial nodal gap slope by fitting using Fit Model. Error bars are determined by the uncertainty of  $E_F$ ,  $k_F$  and the statistical errors of  $\Delta$  from the fits (with its fitting energy window dependence), but do not include the dependence of background subtraction (see Supplementary Information, Section SIIB). A resolution Gaussian with  $\Delta E = 18$  meV is used for the convolution with the spectral function in the fitting.

exhibits a factor of  $\sim 2$  reduction, reminiscent of the optimal  $T_c$  difference between these two families. (3) Its susceptibility to thermal smearing in contrast to its antinodal counterpart (Fig. 4b) is reminiscent of the cases in other superconducting cuprates<sup>11,12</sup>. The existence of precursor pairing in the normal state of LBCO-1/8 is further supported by recent transport measurements<sup>28</sup>. A precipitous drop in the in-plane resistivity at  $T_{2D} \sim 40$  K, where the concurrent stripe (as a density wave) formation would often result in an increase of resistivity, implies an onset of superconducting fluctuations. Although the coincidence of its partial closing with the simultaneous resistivity increase at  $T = T_{2D}$  on heating (Fig. 4b) suggests the precursor-pairing origin of the nodal gap rather than the conventional density-wave type, understanding the relationship between these two coexisting orders in the system at  $T < T_{2D}$  still poses a challenge.

On the other hand, the pairing strength, as reflected by the slope of the nodal pairing gap, is comparable in LBCO-1/8 and its neighbouring compounds of much higher bulk  $T_c$  values (Fig. 4a). Hence, a natural explanation for the bulk  $T_c$  suppression in LBCO-1/8 is its lack of a global superconducting phase coherence. Interestingly, this loss of superconducting phase coherence coincides with the stabilization of the stripe order at  $x \sim 1/8$ . Intrigued by this, theorists have proposed that the global superconducting phase coherence can be prohibited through the dynamical interlayer decoupling in the system where superconductivity is modulated by the stripe order of some specific configurations<sup>29,30</sup>. Nevertheless, the modulated superconductivity of a non-zero wave vector generally does not produce a  $d$ -wave gap with a point node as observed here. Our finding would put a strong constraint on future theoretical attempts to resolve the microscopic mechanism for the failure of high- $T_c$  superconductivity in LBCO-1/8.

As suggested by its different momentum dependence from its nodal counterpart, the antinodal pseudogap may have a different origin, which has been the subject for intense discussions in the literature<sup>2-9</sup>. Generally, as inferred from Supplementary Information, Fig. S11, a similar antinodal phenomenology as we observed can be achieved either by (1) a true gap opening together



**Figure 4 | The doping and temperature dependence of the LEG function.** **a**, Comparison of the LEG function between LBCO-1/8 (sample A, within 2–5 h after sample cleaving, reproduced from Fig. 2b), LSCO  $x = 0.110$  at  $T = 21 \pm 2$  K and LBCO  $x = 0.083$  at  $T = 19 \pm 2$  K. The dashed line is a guide to the eye for the antinodal gap component of LBCO  $x = 0.083$ . Inset: Comparison between LBCO-1/8 and LBCO  $x = 0.083$  of the near- $E_F$  portion of antinodal EDCs taken at the momentum position roughly indicated by the arrow in **a**, showing the absence of the reported anomaly at  $x \sim 1/8$  in the antinodal pseudogap size<sup>16</sup>. EDCs are normalized in intensity at the LEM and shifted in energy with respect to the nodal LEM of each sample. **b**, The LEG function of LBCO-1/8 (sample C) at  $T = 19 \pm 2$  K is compared with the one at  $T = 61 \pm 2$  K. The dashed green curve is a guide to the eye for the 61 K data. Note that the rapid smearing of the distinction between the two gaps as temperature increases is not captured by the extrapolation scheme used in ref. 15 to obtain the ground-state information based on results at high temperatures. Inset: Detailed temperature dependence in the nodal gap region of sample A at three selected momentum positions, C1–C3, as indicated by arrows of different colours. Note that C3 is close to the crossover position of the two gap components. Solid and dashed lines are guides to the eye. See Supplementary Information, Section SIII A for discussion. The same guidelines in black in **a** and **b** for LBCO-1/8 at low temperature are reproduced from Fig. 2b. All data were taken with  $h\nu = 55$  eV. The same nodal referencing scheme is used for the LEG values as in Fig. 2b. Error bars are determined by the uncertainty of  $E_F$ ,  $k_F$  and the energy window dependence of the LEG by the LEG analysis (see Supplementary Information, Section SIIB).

with a strong quasi-particle scattering (the  $\Delta$ - $\Gamma$  physics) or by (2) a great suppression of the quasi-particle spectral weight (the Z physics). Whereas attempts of scheme (1) working in the weak coupling approach can produce a qualitatively similar quasi-particle gap structure by considering the competition between superconductivity and the charge<sup>6,7</sup>- or/and spin<sup>8,29</sup>-density-wave order, scheme (2) demands a strong-coupling route (for example, ref. 9) where the extended quasi-particle analysis as presented above might break down owing to its incapability of capturing the lowest-lying excitations of a vanishing spectral weight. In any case, new physics other than the nodal-type precursor pairing alone is required to fully capture the essence of the antinodal pseudogap.

In contrast to the notion of a simple  $d$ -wave nodal liquid as the pseudogap ground state that is directly derived from the antinodal pseudogap<sup>15,16</sup>, our observation of an apparent break-up in the gap function suggests a very different picture. It reveals a much richer pseudogap physics with its two aspects manifesting differently in distinct momentum regions, that is, the nodal precursor pairing and the antinodal pseudogap of different but unknown origin. These two aspects might be emphasized differently by different experimental probes in the normal state, which has led to the two extremes of ideas for the pseudogap, in particular, whether it has a direct relationship with pairing. Our results suggest a plausible reconciliation between them.

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## Author contributions

T.S., M.F. and K.Y., T.A. and Y.K. provided and prepared the LSCO  $x = 0.110$ , LBCO-1/8 and LBCO  $x = 0.083$  samples, respectively. S.-K.M., K.T. and N.M. maintained the experimental endstation and ensured its high performance. R.-H.H. and K.T. carried out the experiment with the assistance of S.-K.M., N.M. and T.S. R.-H.H. did and K.T. repeated the data analysis and simulations. R.-H.H. wrote the paper with helpful suggestions and comments by S.-K.M. Z.H. and Z.-X.S. are responsible for project direction, planning and infrastructure.

## Additional information

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