

Coexistence of high temperature superconductivity and antiferromagnetic order in a cuprate with multiple hole Fermi pockets

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The intricate relationship between high temperature superconductivity and antiferromagnetic order in cuprates, and the fundamental origin of electron pairing remain open questions. By utilizing high-resolution laser-based spatially-resolved angle-resolved photoemission spectroscopy, we investigate the seven-layer $\text{Bi}_2\text{Sr}_2\text{Ca}_6\text{Cu}_7\text{O}_{18+\delta}$ (Bi2267) and identify a cuprate system that consists of multiple hole Fermi pockets. The observed Fermi pockets exhibit pronounced momentum-, temperature- and Fermi surface-dependent energy gaps. Crucially, high temperature superconductivity with a critical temperature (T_c) of ~ 75 K emerges in a system with multiple Fermi pockets and the presence of strong antiferromagnetic order and correlations. In particular, substantial electron pairing is observed along the Fermi pocket with an energy gap up to ~ 42 meV in lightly-doped CuO_2 planes ($p \sim 0.05$). These findings challenge the conventional understanding of the roles of the nodal and anti-nodal electronic states in driving high temperature superconductivity. They show that superconductivity and antiferromagnetism can coexist in a cuprate with multiple Fermi pockets, offering further insights into the pairing mechanism in cuprate superconductors.

Despite nearly four decades of intensive research, the fundamental mechanism underlying high temperature superconductivity in cuprates remains a major challenge in condensed matter physics^{1,2}. A key debate centers on the nature of the pairing glue, the microscopic interaction responsible for the high superconducting critical

temperature (T_c). In undoped CuO_2 planes, the hybridization between Cu $3d_{x^2-y^2}$ and O $2p$ orbitals generates strong in-plane superexchange interactions, stabilizing long-range antiferromagnetic (AFM) order^{3–6}. In the usual electronic phase diagram, such a long-range AFM order is rapidly suppressed with slight hole doping, followed by the emergence

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of superconductivity at higher doping levels^{2,7}. Within the superconducting doping regime, antiferromagnetic spin fluctuations become pronounced in the underdoped region and persist even into the heavily overdoped region^{8–14}. Theoretical studies have considered the spin fluctuations as a prime candidate of pairing glue in generating high temperature superconductivity^{15–20}. These underscore the intricate interplay between antiferromagnetism and superconductivity in cuprate superconductors.

The extensive studies have also established a general electronic picture of doping the parent Mott insulators of cuprates^{1,21–28}. Upon slight hole doping of the parent compound, the highest occupied electronic states first emerge near the $(\pi/2, \pi/2)$ nodal region of the Brillouin zone^{22–28}. With increasing doping, a Fermi arc develops around the nodal region in the underdoped regime, characterized by coherent quasiparticles along the arc and incoherent electronic states near the $(\pi, 0)$ antinodal region. As the doping further increases, the Fermi arc expands and eventually evolves into a large Fermi surface in the overdoped regime. Such a doping evolution of electronic structure is generally observed in cuprate superconductors and the electronic states around the $(\pi, 0)$ antinodal region are considered to be particularly important in achieving high temperature superconductivity^{19,20,24,29–34}.

In this paper, we report the observation of a unique Fermi surface topology in a seven-layer cuprate $\text{Bi}_2\text{Sr}_2\text{Ca}_6\text{Cu}_7\text{O}_{18+\delta}$ (Bi2267). The observed Fermi surface consists of multiple hole-like Fermi pockets centered at $(\pi/2, \pi/2)$, exhibiting distinct momentum-, temperature- and Fermi surface-dependent energy gaps. This system, comprising multiple hole pockets, reveals the coexistence of high temperature superconductivity, antiferromagnetic order and the pseudogap state.

These findings challenge the well-accepted picture of doping the parent Mott insulators and provide vital insights into the microscopic origin of high temperature superconductivity in cuprate superconductors.

Results

Our angle-resolved photoemission spectroscopy (ARPES) measurements were conducted on a slightly overdoped $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ (Bi2223) sample with a superconducting transition temperature $T_c = 109.0$ K (see Methods and Supplementary Fig. 1). It has been shown from the scanning transmission electron microscopy (STEM) measurements that the Bi2223 samples may contain intergrowth phases of $\text{Bi}_2\text{Sr}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+4+\delta}$ with $n = 1 - 9$ (abbreviated as $\text{Bi}22(n-1)n$)³⁵. Using high-resolution laser-based spatially-resolved ARPES, we successfully identified the multilayer regions on the Bi2223 sample surface (see Methods and Supplementary Fig. 2). The high spatial and energy resolutions of our ARPES setup, combined with excellent system stability, make it possible to systematically collect high quality ARPES data from a fixed spot. While scanning the sample surface, we detected two regions consisting of the seven-layer Bi2267 phase and performed detailed ARPES measurements as reported below.

Figure 1 presents the Fermi surface mappings and constant energy contours measured in the two regions (Region1 and Region2) of the Bi2223 sample surface. The corresponding band structures along different momentum cuts for Region2 are shown in Fig. 2 (the band structures for Region1 are provided in Supplementary Fig. 3). In both regions, four main Fermi surface sheets are clearly observed and

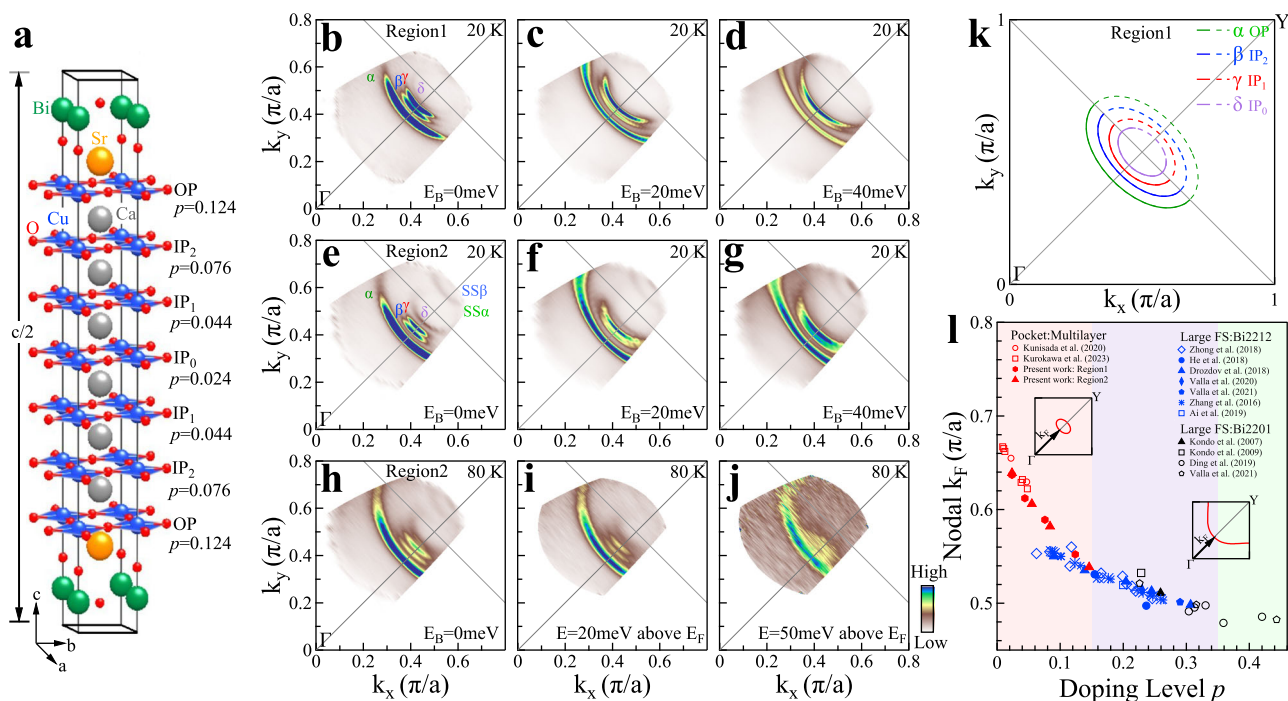


Fig. 1 | Observation of multiple Fermi pockets in Bi2267. **a** Crystal structure of Bi2267. Each half unit cell contains seven adjacent CuO_2 planes with three kinds of inner planes (IP₀, IP₁, IP₂) and one kind of outer planes (OP). The doping level progressively decreases from the outermost to the innermost planes, with values of 0.124, 0.076, 0.044 and 0.024 for the OP, IP₂, IP₁ and IP₀ planes, respectively, deduced from the Fermi pocket areas in Region1. Fermi surface mapping (**b**) and constant energy contours at binding energies of 20 meV (**c**) and 40 meV (**d**) measured at 20 K in Region1. **e–g** Same measurements as (**b–d**) but in Region2. Fermi surface mapping (**h**) and constant energy contours at energies of 20 (**i**) and 50 meV (**j**) above the Fermi level (E_F) measured at 80 K in Region2. Four Fermi surface sheets are identified as α , β , γ and δ along with the superstructure Fermi surface

denoted as SS α and SS β . Detailed analysis is shown in Supplementary Fig. 6. **k** Fermi surface of Bi2267 in Region1. Multiple Fermi pockets are observed labeled as α , β , γ and δ , which are mainly derived from OP, IP₂, IP₁ and IP₀ planes, respectively. On the opposite side of the $(\pi, 0)$ – $(0, \pi)$ line, except for part of the δ Fermi pocket, the observed Fermi pockets are rather weak and are plotted as dashed lines.

l Summarized doping dependence of the nodal Fermi momentum (k_F) in some cuprate superconductors. The nodal Fermi momentum is defined in the upper-left inset for a Fermi pocket and in the bottom-right inset for a large Fermi surface. The data points combine our results with those from the previous measurements of Bi2201, Bi2212 and other multilayer cuprates^{36–47}.

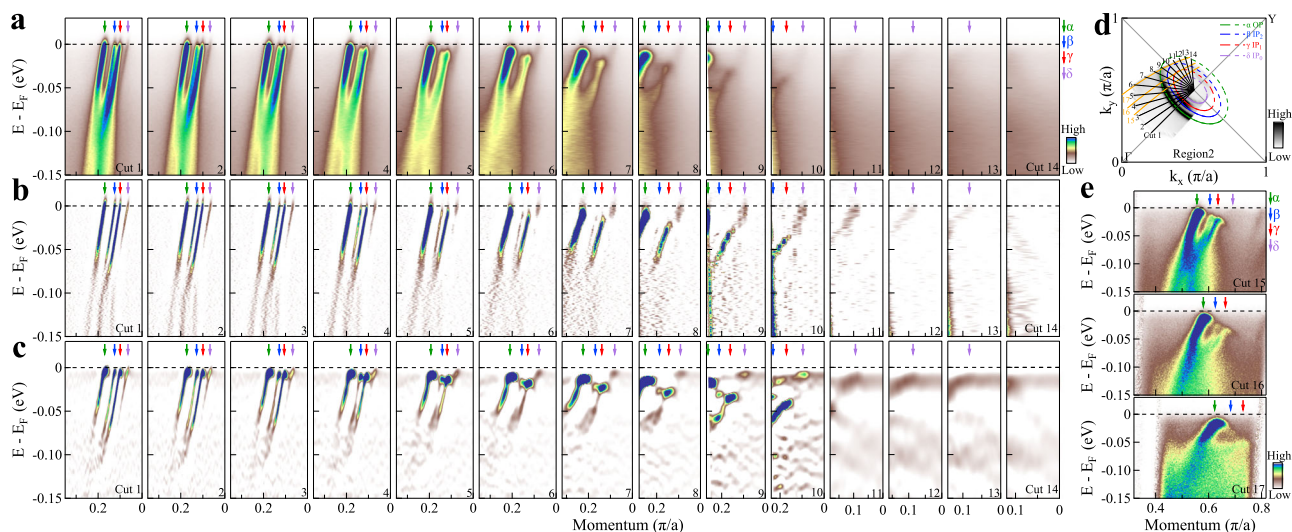


Fig. 2 | Momentum-dependent band structures of Bi2267 measured at 20 K in Region2. **a** Band structures measured along various momentum cuts crossing $(\pi/2, \pi/2)$ point. The locations of these momentum cuts are indicated by black solid lines in **(d)**. Four primary bands are identified and labeled as α , β , γ and δ marked by green, blue, red and lilac arrows, respectively. **b** The corresponding MDC second derivative images from **(a)**. **c** The corresponding EDC second derivative images

from **(a)**. **d** Fermi surface mapping and the multiple Fermi pockets α (green line), β (blue line), γ (red line) and δ (lilac line), with the momentum cuts marked. **e** Band structures measured along the momentum Cuts 15–17; the location of the momentum cuts is shown in **(d)** as orange solid lines. The four primary bands α , β , γ and δ are marked by green, blue, red and lilac arrows, respectively.

labeled as α , β , γ and δ in Fig. 1(b, e). The Fermi surface mapping for Region2 (Fig. 1e) and the corresponding band structures (Fig. 2) reveal a pocket-like Fermi surface for the δ band, with weak spectral weight extending across the $(\pi, 0)$ – $(0, \pi)$ line. The spectral weight of the β and γ Fermi surfaces is primarily concentrated near the nodal region, which can be attributed to a significant anisotropic gap opening, as discussed below. With increasing binding energy, the spectral weight gradually spreads towards the $(\pi, 0)$ – $(0, \pi)$ line, as seen in the constant energy contours (Fig. 1(c, d, f, g)). The full contours of the β and γ Fermi pockets become visible (Fig. 1d) and extend smoothly across the $(\pi, 0)$ – $(0, \pi)$ line. To determine the complete shape of the α Fermi surface, measurements were performed at 80 K (Fig. 1(h–j)), utilizing thermal broadening to probe the spectral weight above the Fermi level. The α Fermi surface is more consistent with a hole pocket around $(\pi/2, \pi/2)$ based on the analyses of the α band structures and the associated constant energy contours, as shown in Supplementary Fig. 4 and Supplementary Fig. 5, and described in Section 4 of Supplementary Information. We note that, due to the experimentally limited momentum range, the exact topology of the α Fermi surface is difficult to be pinned down and further work needs to be done to expand the covered momentum space up to the antinodal region. The possibility of strong AFM correlations, in addition to AFM order, also needs to be further explored as the origin of the hole pocket-like behavior. By analyzing the Fermi surface mappings and the constant energy contours in Fig. 1(b–j), along with the band structures shown in Fig. 2 and Supplementary Fig. 3, the multiple Fermi pockets are determined as shown in Fig. 1k. These pockets exhibit a similar elliptical shape, with a major-to-minor axis ratio of 1.7 ± 0.2 . From the areas of these pockets, the estimated doping levels are 0.124 (0.146), 0.076 (0.084), 0.044 (0.055) and 0.024 (0.024) for the α , β , γ and δ Fermi pockets in Region1 (Region2), respectively. Overall, the electronic structures of Region1 and Region2 are similar, with only slight variations in the doping levels of the observed Fermi pockets. We note that, due to the low photon energy of our laser, we cannot directly measure the electronic states near the $(\pi, 0)$ and $(0, \pi)$ antinodal regions. No low energy electronic states are observed near the antinodal region from the previous ARPES measurements on the five-layer and six-layer cuprates^{36,37}.

Figure 1l shows the nodal Fermi momentum k_F of the observed Fermi pockets as a function of their respective doping levels (red

symbols in Fig. 1l). Here, k_F is defined as the distance between the Γ point and the left edge of the Fermi pocket along the nodal direction (upper-left inset, Fig. 1l). For comparison, we also include data from previous measurements of five-layer and six-layer cuprates^{36,37}. These results show that the nodal Fermi momentum k_F of Fermi pockets decreases monotonically with the increase of the doping level. Additionally, we summarize the distribution of k_F for Bi-based cuprates (Bi2201 and Bi2212) as a function of their doping levels, where a large Fermi surface is typically observed (bottom-right inset, Fig. 1l)^{38–47}. Across a broad doping range ($p = 0.06$ – 0.44), the nodal Fermi momentum of the large Fermi surface decreases monotonically as the doping level increases. The nodal Fermi momentum has a direct one-to-one correspondence to the doping level of the individual CuO_2 plane because the interlayer coupling has minimal effect on the nodal Fermi momentum⁴⁸. As seen in Fig. 1l, the Fermi pocket and the large Fermi surface show distinct doping dependence of the nodal Fermi momentum. It highlights the unique characteristics of the Fermi surface (Fermi pockets) we observed in Bi2267, which are distinct from the large Fermi surface observed in Bi2201 and Bi2212.

The multiple observed Fermi pockets can be mainly attributed to the four inequivalent types of CuO_2 planes in the seven-layer cuprate. It has been shown that, in multilayer cuprates, the hole doping level varies significantly among the CuO_2 planes: it is highest in the outer planes (OPs) due to their proximity to the charge reservoir, decreases progressively toward the inner layers and reaches its lowest value in the innermost plane(s) (IP₀s)^{35,49}. In the underdoped or optimally-doped cuprates, the interlayer coupling is usually weak⁵⁰. In such a case, the formation of a Fermi surface is dominated by a particular type of CuO_2 plane with a given doping level because the weak interlayer coupling has little effect on the Fermi surface topology³⁷. The observation of separate Fermi pockets indicates that there are four types of CuO_2 planes in the measured material and the α , β , γ and δ Fermi pockets are mainly derived from the OP, IP₂, IP₁ and IP₀ CuO_2 planes, respectively. We find that the doping level ratios between adjacent CuO_2 planes are close to 2, with $p(\text{OP})/p(\text{IP}_2) = 1.63$, $p(\text{IP}_2)/p(\text{IP}_1) = 1.73$ and $p(\text{IP}_1)/p(\text{IP}_0) = 1.83$. This trend is consistent with previous observations in multilayer cuprates with an odd number of CuO_2 planes, which feature a single innermost CuO_2 plane^{36,37}. In contrast, in multilayer cuprates with an even number of CuO_2 planes where two

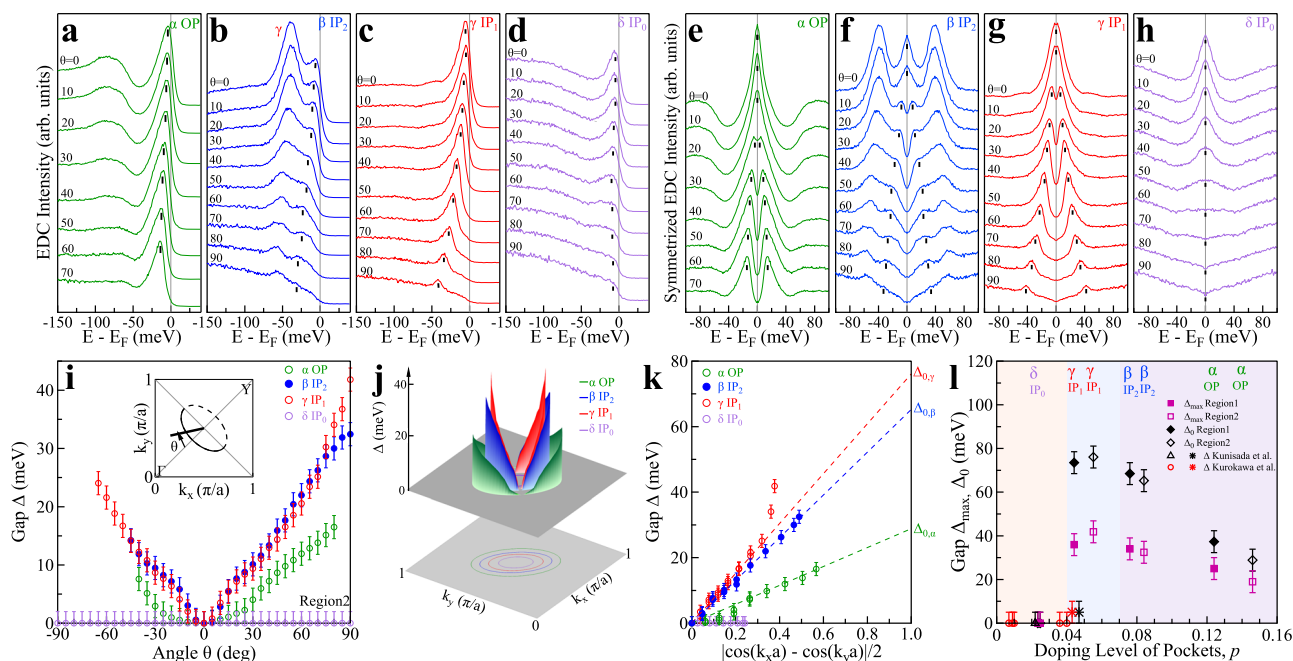


Fig. 3 | Photoemission spectra and the energy gap of Bi2267 along the Fermi pockets measured at 20 K in Region2. EDCs measured along the α (a), β (b), γ (c) and δ (d) Fermi pockets, respectively. The location of the Fermi momentum is defined by the angle θ , as depicted in the inset of (i). The EDC peaks corresponding to α , β , γ and δ bands are marked by ticks in (a–d). e–h Symmetrized EDCs obtained from (a–d). i Momentum-dependent energy gaps along the Fermi pockets obtained from the symmetrized EDCs in (e–h). The maximum gap size at the major vertex is ~ 19 meV, ~ 32.5 meV, ~ 42 meV and 0 for the α , β , γ and δ Fermi pockets, respectively. The angle θ is defined in the insets of (i). j Three-dimensional plot of the energy gaps along the Fermi pockets. k The energy gaps plotted as a function of

$|\cos(k_x a) - \cos(k_y a)|/2$ for the Fermi pockets. The energy gaps along α and β Fermi pockets exhibit a linear dependence, with the extrapolated values $\Delta_{0,\alpha}$ and $\Delta_{0,\beta}$ at the $(0, \pi)$ point. The energy gap along γ Fermi pocket shows a linear dependence near the nodal region with the extrapolated value $\Delta_{0,\gamma}$ at the $(0, \pi)$ point. It deviates from a linear dependence with the momentum moving away from the nodal region. l The distribution of the energy gaps, the maximum energy gap $\Delta_{\max}$ (purple symbols) and the extrapolated energy gap Δ_0 (black symbols), on the Fermi pockets measured in two regions (Region1 and Region2). Error bars indicate the uncertainties in determining the energy gap sizes. The maximum energy gaps observed in the previous work^{36,37} are also included for comparison.

innermost CuO_2 planes (IP_0 s) are present, the doping level of each IP_0 is approximately halved, yielding a ratio of $p(\text{IP}_1)/p(\text{IP}_0) \approx 4$ in the six-layer system³⁷. Our observed doping distribution among the CuO_2 planes aligns with the case of odd number of CuO_2 planes and the region we measured can be attributed to the seven-layer Bi2267, as shown in Fig. 1a.

Figure 2a illustrates the evolution of the band structures as the momentum cuts rotate from the nodal direction (Cut 1) toward the $(\pi, 0)$ – $(0, \pi)$ direction (Cut 10) and beyond (Cut 11–Cut 14) around the $(\pi/2, \pi/2)$ point. To enhance the visibility of the bands, the corresponding MDC and EDC second-derivative images are shown in Fig. 2b and Fig. 2c, respectively. Fig. 2e presents typical band structures along momentum cuts nearly parallel to $(0, 0)$ – (π, π) direction around the major vertices of the Fermi pockets. Four distinct bands, labeled α , β , γ and δ , are clearly identified and highlighted with colored arrows in Fig. 2(a–c) and Fig. 2e. Along the nodal direction (first panel of Fig. 2(a–c)), the four bands are well separated, with the β and γ positioned closer to each other. The α band is strong in Fig. 2(a–c) (Cut 1–Cut 9) and Fig. 2e (Cut 15–Cut 17), shifting to higher binding energy from Cut 1 to Cut 9 due to energy gap opening. The β band is prominent near the nodal region (Cut 1–Cut 4 in Fig. 2(a–c)) but rapidly loses spectral weight as the momentum cuts rotate from Cut 4 to Cut 10, also shifting to higher binding energy due to energy gap opening. Notably, the β band exhibits strong Bogoliubov hybridization with the α back-bending band, which becomes increasingly obvious from Cut 1 to Cut 10. This hybridization, similar to that observed in Bi2223^{51–53}, likely arises from relatively strong interlayer coupling between OP and IP_2 layers as well as the substantial superconducting gap difference between the α and β bands. The γ band remains robust and well-defined from Cut 1 to Cut 10, shifting far below the Fermi level due to

the large energy gap opening. It also exhibits Bogoliubov band hybridization with the β back-bending band, particularly evident in Cut 7–Cut 9 in Fig. 2(a–c). The δ band exhibits notably weaker spectral weight compared to the other three bands, particularly at deep energy levels near the $(\pi, 0)$ – $(0, \pi)$ direction. It crosses the Fermi level all the way from Cut 1 to Cut 13 in Fig. 2(a–c).

To determine the superconducting gap along the Fermi pockets, the original energy distribution curves (EDCs) and the corresponding symmetrized EDCs along the pockets measured at 20 K for Region2 are presented in Fig. 3(a–d) and Fig. 3(e–h), respectively. All the four bands show well-defined quasiparticle peaks near the nodal region ($\theta = 0$) (Fig. 3(a–d)). Strong and clear EDC peaks for the α and γ bands are observed along the Fermi pockets (Fig. 3(a, c)). The β band, being relatively weaker than the γ band and overlapping with it, exhibits a two-peak feature in both the EDCs and symmetrized EDCs, as marked in Fig. 3(b, f). The EDC peaks near the Fermi level, marked by ticks in Fig. 3(b, f), correspond to the β band, while the higher binding energy peaks originate from the γ band. For the δ band, prominent EDC peaks appear near the nodal region ($\theta = 0$) but become rather weak when the Fermi momentum moves towards the major vertex region ($\theta = 90$) (Fig. 3(d, h)). Detailed analysis of the band structure and EDC feature assignment are summarized in Supplementary Fig. 7. The gap size is extracted from the peak positions in the symmetrized EDCs. The superconducting gap along the α , β , γ and δ Fermi pockets, as obtained from the symmetrized EDCs in Fig. 3(e–h), is plotted in Fig. 3i. Figure 3j shows the three-dimensional distribution of the energy gaps along the Fermi pockets, revealing distinct Fermi surface-dependent gap behaviours. While the α , β and γ Fermi pockets exhibit clear gap opening, the δ Fermi pocket remains gapless along the Fermi pocket.

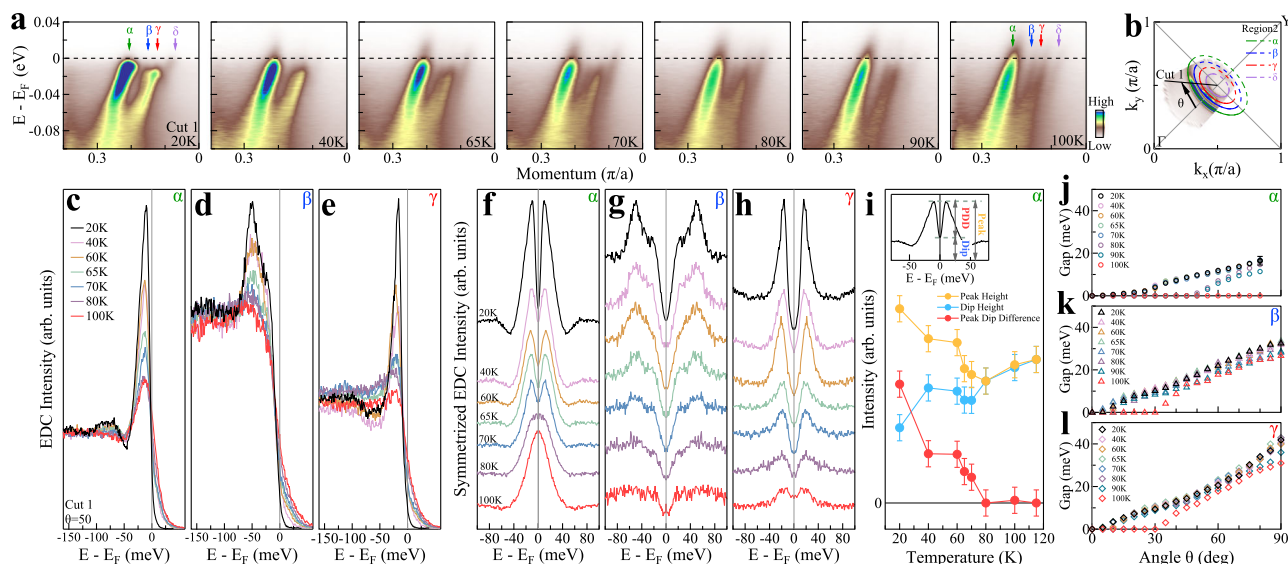


Fig. 4 | Evolution of band structures, photoemission spectra and energy gaps with temperature along a typical momentum cut crossing the Fermi pockets. **a** Band structures measured along the momentum Cut 1 at different temperatures. The location of the momentum Cut 1 is shown in **(b)**. **b** Fermi surface mapping and the Fermi pockets α (green line), β (blue line), γ (red line) and δ (lilac line), with the momentum cut marked and the angle θ defined. **c–e** EDCs measured at different temperatures obtained from **(a)** at the Fermi momenta of the α (**c**), β (**d**) and γ (**e**) bands as marked by the colored arrows in **(a)**. **f–h** The corresponding symmetrized EDCs obtained from **(c–e)**. **i** Temperature dependence of the spectral intensities

extracted from the symmetrized EDCs of the α band in **(f)**. The upper-left inset defines the three spectral intensities: the peak height, the dip height and the peak-dip difference (PDD). The corresponding intensities as a function of temperature are plotted by the orange solid circles (peak height), the blue solid circles (dip height) and the red solid circles (PDD). The error bars reflect uncertainties in determining the spectral intensity. Momentum dependence of the energy gap along the α (**j**), β (**k**) and γ (**l**) Fermi pockets measured at different temperatures. The uncertainties in determining the gap size are marked by the error bar in **(j)**.

The gap opening along the α , β and γ Fermi pockets is highly anisotropic, vanishing along the nodal direction ($\theta = 0$) and reaching its maximum near the major vertex ($\theta = 90$). The momentum-dependence of the gap for the α and β Fermi pockets follows a d-wave form $\Delta = \Delta_0 |\cos(k_x a) - \cos(k_y a)|/2$ as seen in Fig. 3k. The gap along the γ Fermi pocket follows the d-wave form near the nodal region ($\theta = 0 - 70$) but deviates near the major vertex region ($\theta = 70 - 90$). This gap behaviour is similar to that observed in underdoped cuprates⁵⁴. The gap sizes along the α , β and γ Fermi pockets differ significantly. The maximum gap sizes at the major vertex are ~ 19 , ~ 32.5 and ~ 42 meV for the α , β and γ Fermi pockets, respectively (Fig. 3(i–k)). The extrapolated maximum gap sizes Δ_0 , extracted from the d-wave form in Fig. 3k, are ~ 29 , ~ 65 and ~ 76 meV for the α , β and γ Fermi pockets, respectively. The EDC lineshape, momentum-dependence and Fermi surface-dependence of the energy gap along the Fermi pockets are similar from the measurements of Region1, except for slight variations in gap size due to doping differences between the two measured regions (Supplementary Fig. 8). Figure 3l summarizes the maximum energy gap $\Delta_{\max}$ (purple symbols) and the extrapolated energy gap Δ_0 (black symbols) along the Fermi pockets measured in both Region1 and Region2. We note that the energy gaps along the α , β and γ Fermi pockets we observed in our seven-layer Bi2267 are significantly larger than those reported for five-layer and six-layer cuprates^{36,37}. In the previous ARPES measurements on the five-layer and six-layer cuprates^{36,37}, only a small gap (<5 meV) is observed in the IP₁ CuO₂ plane adjacent to OP which is comparable or slightly smaller than the gap in the OP CuO₂ plane. This suggests that the superconducting gap in the IP₁ CuO₂ plane is possibly induced by the OP CuO₂ plane. In our seven-layer cuprate, (1). we observed much larger gap opening (up to 42 meV) in the OP, IP₂ and IP₁ CuO₂ planes; (2). we observed a large gap opening in the IP₁ CuO₂ plane that is not adjacent to OP CuO₂ plane. Its gap size is even larger than that in the IP₂ CuO₂ plane that is adjacent to the OP CuO₂ plane; (3). the gap opening in the inner IP₁ and IP₂ CuO₂ planes is significantly larger than that in the OP CuO₂ plane. These results clearly indicate that the gap opening

in the inner CuO₂ planes is not induced by the outer OP CuO₂ planes. Instead, the gap opening is more intrinsic to a specific CuO₂ plane, which is dictated by electron correlation and magnetic interaction that are dependent on the doping level and the interlayer interactions. The extrapolated energy gap Δ_0 , derived from energy gaps around the nodal region (Fig. 3k), is generally regarded as the intrinsic superconducting gap. For the γ Fermi pocket, this gap can reach ~ 76 meV (Fig. 3l), exceeding that of Bi2223 and representing the largest value reported in any cuprates to date^{54,55}. The large values of $\Delta_{0,\beta}$ and $\Delta_{0,\gamma}$ (Fig. 3l) indicate the strong pairing strength in the β and γ Fermi pockets of Bi2267.

To investigate the temperature dependence of the gap opening and superconductivity in the Fermi pockets, we performed detailed measurements in Region2 over a temperature range of 20 K to 115 K (Fig. 4). Figure 4a shows a typical band structure measured at different temperatures along a momentum cut near the major vertices of the Fermi pockets ($\theta = 50$, Fig. 4b). The four bands, α , β , γ and δ , are clearly resolved and exhibit distinct temperature evolution. The δ band crosses the Fermi level without forming a gap at all measured temperatures. Figure 4(c–e) display the EDCs at different temperatures measured at the Fermi momenta of the α , β and γ bands, respectively, obtained from Fig. 4a. The corresponding symmetrized EDCs are presented in Fig. 4(f–h). Further analyses are shown in Supplementary Fig. 9 and Supplementary Fig. 10. At low temperatures, strong and sharp EDC peaks appear for the α (Fig. 4c) and γ (Fig. 4e) bands, whereas the β band sharpens but does not form well-defined peaks (Fig. 4d).

The α , β and γ Fermi pockets exhibit distinct temperature-dependent behaviors in the energy gap opening and superconducting coherence development. As shown from Fig. 4(f–h), the α band develops an energy gap at low temperatures but the gap closes at 80 K and 100 K (Fig. 4f). In contrast, the gap opening in the β and γ bands persists up to 100 K (Fig. 4(g, h)). Figure 4i presents the temperature dependence of the spectral intensity of the dip at the Fermi level, the EDC peak height and their difference obtained from the symmetrized

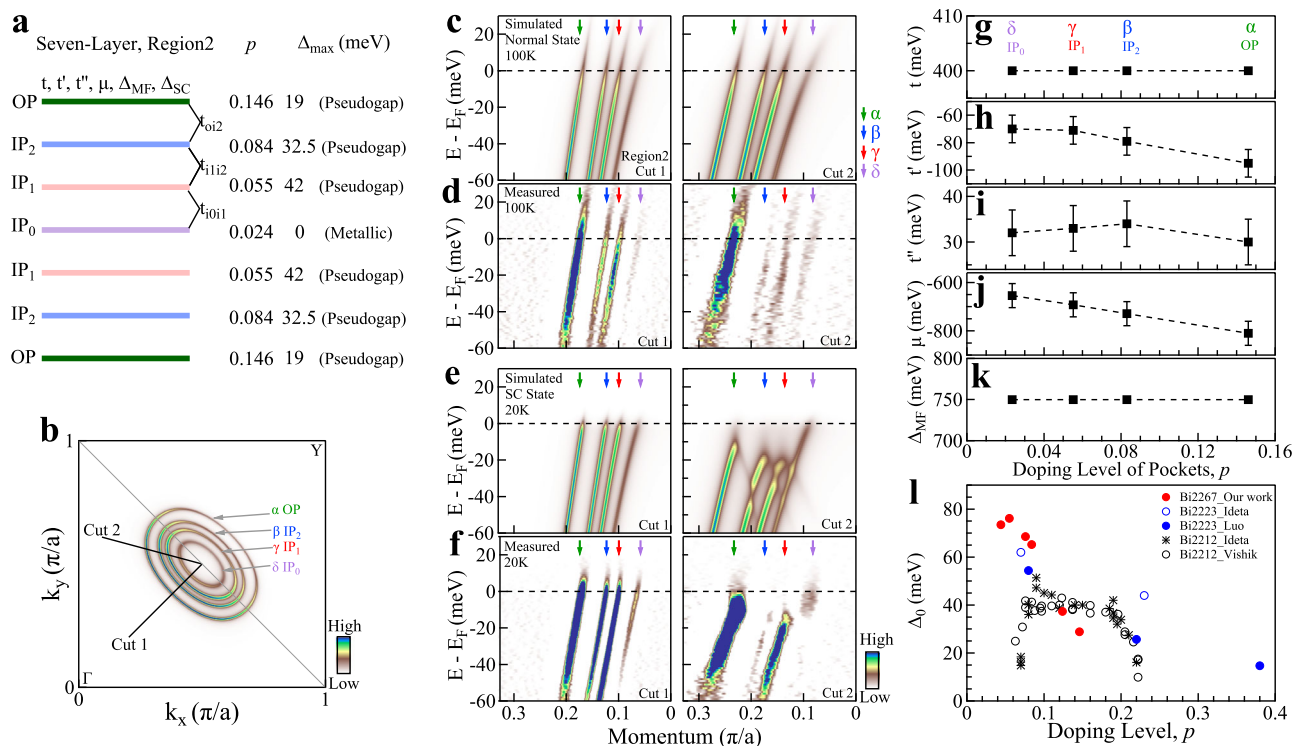


Fig. 5 | Formation of the multiple Fermi pockets in the seven-layer cuprate in terms of the mean field t - U model. **a** Schematic illustration of the seven-layer mean field t - U model including the in-plane parameters $t, t', t'', \mu, \Delta_{\text{MF}}$ and Δ_{SC} , and the interlayer hopping parameters t_{0i2}, t_{1i12} and t_{0i1} . The doping level and maximum energy gap distributions among the seven CuO_2 layers in Bi2267 (Region2) are also shown. **b** Simulated Fermi surface consisting of multiple Fermi pockets. **c-f** Simulated band structures (**c, e**) and their comparison with the measured results (**d, f**). The simulation details are described in Section 9 of the Supplementary Information. **c** Simulated band structures along the momentum Cut 1 (left panel) and Cut 2 (right panel) at 100 K in the normal state. The location of the momentum

cuts is represented by black solid lines in (**b**). Four bands are labeled as α, β, γ and δ marked by green, blue, red and lilac arrows, respectively. **d** Measured band structures along the momentum Cut 1 (left panel) and Cut 2 (right panel) at 100 K in the normal state in Region2. These are MDC second-derivative images. **e, f** Same as (**c, d**) but at 20 K in the superconducting state. **g-k** The extracted parameters t (**g**), t' (**h**), t'' (**i**), μ (**j**) and Δ_{MF} (**k**) obtained by fitting the observed Fermi surface and band structures in Region2 with the mean field t - U model. Error bars in (**g-k**) represent the uncertainties in extracting the fitting parameters. **l** Comparison of the extrapolated energy gap Δ_0 in our Bi2267 with those measured in Bi2212^{54,55,68} and Bi2223^{53,55}.

EDCs for the α band in Fig. 4f. The spectral weight shows little variation with temperature between 80 K and 115 K, while the superconducting coherence peak starts to abruptly develop between 70 K and 80 K. This provides a reliable estimate of the superconducting transition temperature (T_c) of the measured Region2 as 75 ± 5 K^{56,57}. This is consistent with the T_c of 70 ± 5 K for the seven-layer Bi2267 reported in refs. 35,58 and the overall gap measurements along the three Fermi pockets at different temperatures (Fig. 4(j-l)). For the α Fermi surface, below 70 K, the energy gap is present along the entire Fermi pocket except for the nodal direction (Fig. 4j). At 80 K and 90 K, the gap closes around the nodal region, forming a Fermi arc extending to $\theta = 50$. At 100 K, the gap vanishes along the entire Fermi pocket. These observations suggest a superconducting transition temperature T_c between 70 and 80 K, the emergence of a pseudogap between 70 and 100 K and the pseudogap temperature onset T^* of 95 ± 5 K for the α Fermi surface. For the β and γ Fermi pockets, a well-defined Fermi arc appears at 100 K, with a length extending only to $\theta = 30$ (Fig. 4(k, l)). This indicates that the pseudogap temperature T^* of the β and γ Fermi pockets is significantly higher than that of the α Fermi surface. Such a variation in T^* can be attributed to the doping difference of the Fermi pockets with the α Fermi surface being slightly underdoped ($p = 0.146$), while the β ($p = 0.084$) and γ ($p = 0.055$) pockets are heavily underdoped. Notably, this represents a direct observation of pseudogap formation on the Fermi pockets by ARPES in multilayer cuprates.

It has been found that the antiferromagnetic order is strongly enhanced in multilayer cuprates and the strength of the antiferromagnetic order increases with the number of CuO_2 layers (n)⁴⁹.

Our observation of multiple Fermi pockets is consistent with the presence of strong antiferromagnetic order and strong electron correlation in this seven-layer cuprate. The observation that the α Fermi surface is more consistent with the Fermi pocket picture suggests that there remain strong electron correlation and antiferromagnetic order in the outer CuO_2 planes in the seven-layer cuprates. As shown in ref. 49, for the five-layer ($n = 5$) cuprates, the OP layers can be antiferromagnetic in the underdoped sample (see Fig. 6a of ref. 49). Since the region of the long-range antiferromagnetic order increases with the number of CuO_2 planes (n), it is reasonable to expect that the OP layers in the seven-layer ($n = 7$) cuprates may exhibit a static antiferromagnetic order when the doping level is relatively low. The Fermi pockets we observed in Bi2267 are different from the pockets observed in the electron doped cuprates^{59,60}. Their formation cannot be explained in the band folding picture. When the doping level of CuO_2 plane is small, there is no large Fermi surface to start with as required in the band folding picture. Instead, the electron correlation is strong and it is more suitable to be described by the t - U model as presented in detail in the Supplementary Information⁶¹⁻⁶⁵. In this framework, key parameters such as the intralayer electron hopping (t, t' and t''), the chemical potential (μ), the mean field gap (Δ_{MF}) and the superconducting gap (Δ_{SC}) for each layer, along with the interlayer hopping terms (t_{0i2}, t_{1i12} and t_{0i1}), are taken into account (Fig. 5a). The observed Fermi surface (Fig. 2d) and the measured band structures along typical momentum cuts in the normal state (Fig. 5d and Supplementary Fig. 11) for Region2 are well fitted by using the mean field t - U model, with the fitted parameters plotted in Fig. 5(g-k). The

simulated Fermi surface (Fig. 5b) and the corresponding simulated band structures in the normal state (Fig. 5c and Supplementary Fig. 11) obtained by using these parameters in Fig. 5(g–k), are in good agreement with the measured data. Incorporating a set of appropriate superconducting gap and interlayer coupling parameters (listed in the Supplementary Information and Supplementary Table 1), we further simulated the band structures in the superconducting state (Fig. 5e). These simulations also closely match with the measured band structures in Fig. 5f.

The observation of multiple Fermi pockets demonstrates that strong electron correlation and strong antiferromagnetic order are associated with all the seven CuO₂ planes in Bi2267. From the fitted parameters (Fig. 5(g–k)), we find that the multiple Fermi pockets and their associated band structures in the normal state can be described by a similar set of parameters $\{t, t', t'', \Delta_{\text{MF}}\} = \{0.4, -0.079, 0.032, 0.75\}$ and a varying μ . Based on the same band structure (Supplementary Fig. 11b), the formation of the multiple Fermi pockets can be attributed to the shifting of the chemical potential (Supplementary Fig. 11a and Supplementary Fig. 11b). The variation in chemical potential arises from the uneven charge carrier distribution among the four types of CuO₂ planes in Bi2267 (Fig. 5a). Our simulations further indicate that the interlayer coupling has a minimal impact on the Fermi pocket topology and related band structures (Supplementary Fig. 12). As the interlayer coupling strength depends on the doping levels of adjacent CuO₂ planes⁵⁰, and the three inner planes are underdoped or heavily underdoped (Fig. 5a), the interlayer coupling is weak in Bi2267, particularly between the two inner planes. Consequently, the observed Fermi pockets (α , β , γ and δ) in momentum space can be mainly attributed to the four distinct types of CuO₂ planes (OP, IP₂, IP₁ and IP₀) in real space.

Discussion

In Bi2267, the doping level of the IP₁ (IP₂) CuO₂ plane is ~ 0.05 (~ 0.08), which is heavily underdoped. We found that the pairing gap along the IP₁ (IP₂) Fermi pocket can reach $\Delta_{\text{max}} = 42$ meV (~ 33 meV) and the extrapolated gap $\Delta_0 = 76$ meV (~ 66 meV) is even larger than the gap $\Delta_0 \sim 60$ meV for the inner IP CuO₂ plane in Bi2223 that has a doping level of 0.08 (Fig. 5l). The observation of larger gap opening on the IP₁ and IP₂ CuO₂ planes in Bi2267 is consistent with the usual trend that the energy gap size increases with decreasing doping level in cuprates⁶⁶. In cuprate superconductors, the superconducting transition temperature T_c is determined not only by the gap size, but also by the coherence temperature, which increases with increasing doping level⁶⁶. In multilayer cuprates, the pairing strength and the coherence temperature may be realized in different CuO₂ planes, and in this case, the interlayer coupling is also important in determining T_c . In optimally-doped and overdoped Bi2223, the OP CuO₂ planes are heavily overdoped ($p = 0.22$ – 0.30)^{53,55}. In Bi2267, the doping level of the OP CuO₂ planes is $p = 0.12$ – 0.15 . This indicates that the coherence temperature of the OP CuO₂ planes in Bi2267 is lower than that in Bi2223. The coherence temperature difference may account for why the maximum T_c in Bi2223 ($T_c \sim 110$ K) is higher than that in Bi2267 ($T_c \sim 75$ K).

Our findings provide the direct observation of a cuprate exhibiting multiple hole Fermi pockets and demonstrate that such a system can sustain high temperature superconductivity with a T_c as high as ~ 75 K. In previous studies, cuprates have consistently exhibited a large Fermi surface, although the antinodal states may become incoherent in the underdoped regime^{22,24,67}. The antinodal electronic states have been considered essential for high temperature superconductivity^{19,20,24,31–34}. However, our present observation of Fermi pockets only indicates that the nodal region electronic states alone can support high temperature superconductivity, challenging the conventional understanding of the role of antinodal electronic states. Although the observed Fermi pockets are confined near the nodal

region, a large energy gap of up to ~ 42 meV can already open along the Fermi pockets (Fig. 3l), indicative of strong pairing strength that can be realized in the Fermi pockets. In fact, Bi2267 exhibits the strongest pairing strength among all measured cuprate superconductors (Fig. 5l)^{53–55,68}. Furthermore, the α and γ bands exhibit pronounced superconducting coherence peaks at low temperatures (Fig. 3(a,c)), reflecting the establishment of phase coherence in the corresponding CuO₂ planes. These indicate that the Fermi pockets alone meet the requirements for the strong pairing strength and phase coherence to achieve high temperature superconductivity⁶⁶. Our observation of high temperature superconductivity in a cuprate with multiple hole pockets raises a fundamental question: in cuprates with a large Fermi surface, which part of the electronic states, the nodal region or antinodal region, plays the dominant role in generating high temperature superconductivity. Our present findings motivate further theoretical studies to investigate whether and how high temperature superconductivity can be realized through only the nodal region electronic states.

Our present work also provides valuable perspectives on the electronic phase diagram and electron pairing mechanism in cuprates. In the conventional phase diagram of cuprates, high temperature superconductivity emerges only after the suppression of antiferromagnetic order², which implies the competition between superconductivity and the antiferromagnetic order. However, in our Bi2267, we observe that high temperature superconductivity can coexist with the strong antiferromagnetic order. In particular, strong electron pairing with an energy gap as high as ~ 42 meV can be realized in a CuO₂ plane (IP₁) with a doping level of ~ 0.05 and a robust antiferromagnetic order (Fig. 3l). This observation offers deeper insight to understand the pairing mechanism in cuprate superconductors. The previous NMR studies on multilayer cuprates⁴⁹ indicate that the region of the long-range antiferromagnetic order increases with the number of CuO₂ planes (n). For $n = 5$, long-range AFM order can persist up to a doping level of ~ 0.10 , and for our case of $n = 7$, such a doping level is expected to be even higher. The slightly doped CuO₂ planes in the seven-layer cuprate ($p \sim 0.05$) therefore are well within the doping range of the long-range antiferromagnetic order. We found that all the observed γ and δ bands are caused by Fermi pockets; no other features are observed outside of the Fermi pockets. These results also indicate that the IP₁ and IP₀ layers are in the long-range AFM order regime. The electron pairing already occurs in the slightly doped CuO₂ planes ($p \sim 0.05$), which exhibit an antiferromagnetic order without entering the spin fluctuation region yet⁴⁹. These findings indicate that there is already a pairing interaction in the antiferromagnetic state that is distinct from the spin fluctuations^{15–20}. Further investigations are required to clarify how strong electron pairing emerges in the CuO₂ plane with slight doping and robust antiferromagnetic order.

In summary, by using high-resolution laser-based spatially-resolved ARPES, we successfully measured the electronic structure and energy gap of the seven-layer Bi2267. We identified a cuprate system that consists of multiple hole Fermi pockets. The formation of Fermi pockets originates from the strong electron correlation and the enhanced antiferromagnetic order in the multilayer system. Strong electron pairing with an energy gap as high as ~ 42 meV is observed in the CuO₂ plane at a slight doping level of ~ 0.05 where a robust antiferromagnetic order persists. The large energy gap and sharp EDC peaks observed along the Fermi pockets near the nodal region highlight the strong electron pairing strength and phase coherence. Importantly, such a system with multiple hole pockets can sustain high temperature superconductivity with a T_c of ~ 75 K while coexisting with the antiferromagnetic order. These observations provide key insights in understanding the electronic phase diagram and the electron pairing mechanism in high temperature cuprate superconductors.

Methods

Sample growth and preparation

Single crystals of $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ (Bi2223) were grown by the traveling solvent floating zone method^{69,70}. The samples were post-annealed at 550 °C under high oxygen pressure, about 160 atmospheres for 14 days. The obtained samples are slightly overdoped with a T_c of 109.0 K and a transition width $\Delta T \approx 2.0$ K, as seen in Supplementary Fig. 1.

ARPES measurements

ARPES measurements were carried out using our lab-based laser ARPES system equipped with the 6.994 eV vacuum-ultra-violet (VUV) laser and a DA30L hemispherical electron energy analyzer^{71,72}. The energy resolution was set at 1 meV and the angular resolution is approximately 0.3° , corresponding to a momentum resolution of $0.004 \text{ \AA}^{-1}$. The samples were cleaved in situ at a low temperature and measured in vacuum with a base pressure better than 5×10^{-11} Torr at 20 K. The Fermi level is referenced by measuring on a clean polycrystalline gold that is electrically connected to the sample. The laser spot size on the sample was set at $\sim 20 \mu\text{m}$. It has been observed that our Bi2223 single crystal samples contain intergrowth phases of $\text{Bi}_2\text{Sr}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+4+\delta}$ with the number of CuO_2 planes per structural unit $n = 1 - 9$ ³⁵. We first scanned the entire sample surface by performing ARPES measurements on each spot (see Supplementary Fig. 2). Different regions of the sample surface were identified by looking at the number of bands and their distribution measured on each spot. In this work, we selected the region with four bands (Supplementary Fig. 2h) with a size of $\sim 50 \mu\text{m}$ and carried out detailed momentum- and temperature-dependent measurements. The DA30 mode of our electron energy analyzer is used to map the Fermi surface and band structures without moving the sample at different temperatures. From the measured Fermi surface (Fig. 1) and band structures (Fig. 2), we determined that the measured area is Bi2267 as described in the main text.

Data availability

All data are processed by using Igor Pro 8.02 software. All data needed to evaluate the conclusions in the paper are available within the article and its Supplementary Information files. All raw data generated during the current study are available from the corresponding author upon request.

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Author contributions

X.J.Z., L.Z. and X.Y.L. proposed and designed the research. X.Y.L., H.C., Y.W.C. and J.M.S. carried out the ARPES experiments. C.T.L. grew the single crystals. X.Y.L. contributed in sample preparation. H.C., T.M.M., B.L., W.P.Z., N.C., X.L.R., Y.J.S., C.H.Y., J.X.Z., S.J.Z., Z.M.W., F.F.Z., F.Y., Q.J.P., Z.Y.X., G.D.L., X.T.L., H.Q.M., L.Z. and X.J.Z. contributed to the development and maintenance of the ARPES systems and related software development. X.Y.L., Y.H.L. and T.X. contributed to theoretical analysis and discussions. X.Y.L., L.Z. and X.J.Z. analyzed the data and wrote the paper. All authors participated in discussions and comments on the paper.

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Competing interests

The authors declare no competing interests.

Additional information

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