Spectroscopic evidence for preformed Cooper pairs in the pseudogap phase of cuprates

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Angle-resolved photoemission on underdoped La$_{1.895}$Sr$_{0.105}$CuO$_4$ reveals that in the pseudogap phase, the dispersion has two branches located above and below the Fermi level with a minimum at the Fermi momentum. This is characteristic of the Bogoliubov dispersion in the superconducting state. We also observe that the superconducting and pseudogaps have the same d-wave form with the same amplitude. Our observations provide direct evidence for preformed Cooper pairs, implying that the pseudogap phase is a precursor to superconductivity.

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In conventional superconductors, the energy gap decreases with increasing temperature and vanishes at $T_c$. For the high temperature cuprates, the situation is more complicated. In the underdoped region, an energy gap, known as the pseudogap, persists above $T_c$, its maximum amplitude remaining unchanged before the gap fills in at a temperature, $T^*$ [1, 2]. Both $T^*$ and the pseudogap increase with reduced doping, whereas $T_c$ decreases. In spite of much effort, there is no consensus on the origin of the pseudogap [3]. One idea is that the energy gap above $T_c$ is indicative of preformed Cooper pairs [4, 5] that only become phase coherent below $T_c$. A competing view is that the pseudogap results from some other order which competes with superconductivity. In this picture, the superconducting gap only exists along gapless Fermi arcs [6, 7, 8, 9, 10], and as in conventional superconductors, it closes at $T_c$. Using angle-resolved photoemission spectroscopy (ARPES), we show the existence of a Bogoliubov-like dispersion in the pseudogap phase of the underdoped cuprate La$_{1.895}$Sr$_{0.105}$CuO$_4$. This provides direct evidence that the pseudogap is a signature of preformed Cooper pairs above $T_c$.

ARPES experiments were carried out at the Surface and Interface Spectroscopy beamline at the Swiss Light Source on single crystals of underdoped La$_{1.895}$Sr$_{0.105}$CuO$_4$ (LSCO) with $T_c=30$K and overdoped Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi2212) with $T_c=65$K, grown using the travel solvent floating method. Circularly polarized light with $h\nu=55$ eV and linearly polarized light with $h\nu=21.2$ eV were used for LSCO and Bi2212, respectively. The energy and angle resolutions were 17 meV and 0.1 - 0.15°. The Fermi level was determined by recording the photoemission spectra from polycrystalline copper on the sample holder. For LSCO, the samples were cleaved in situ by using a specially designed cleaver [10].

In Fig. 1 we show spectra obtained below and above $T_c$ from angle resolved photoemission spectroscopy (ARPES) for underdoped La$_{1.895}$Sr$_{0.105}$CuO$_4$ ($T_c=30$K) near the node (Fig. 1(a), (d)), near the anti-node (Fig. 1(c), (f)), and in between (Fig. 1(b), (e)). Here, the node refers to where the d-wave superconducting gap vanishes, with the anti-node where it is maximal. To determine the Fermi momentum $k_F$ and the energy gap, we symmetrize the spectra along each cut [11], $A(\omega) = I(\omega) + I(-\omega)$, where $I(\omega)$ is the measured intensity at the energy $\omega$. $k_F$ was identified by searching for the minimum gap location of the symmetrized spectra along a given cut in momentum space. The same underlying Fermi surface was obtained by analyzing the spectra in the superconducting state and in the pseudogap phase. The energy gap at a given $k_F$ was determined by fitting the symmetrized spectrum (Fig. 1(g)), with the spectral function calculated from a model self-energy [12] of the form $\Sigma = -i\Gamma + \Delta^2/(\omega + i0^+)$, and then convolved with the instrumental resolution. Like for lightly underdoped La$_{1.855}$Sr$_{0.145}$CuO$_4$ [13], the superconducting gap at $T=12$ K traces out a simple d-wave form with a maximum amplitude of 25 meV at the anti-node (Fig. 1(i)). Above $T_c$ at $T=49$K, there is a small gapless Fermi arc near the node (Fig. 1(h), (j)). Beyond the Fermi arc, a pseudogap is present that follows the same simple d-wave form as the superconducting gap. It is interesting to note that in our spectral fits, the inverse lifetime, $\Gamma_1$, has the same angular anisotropy as the energy gap (Fig. 1(i), (k)). Similar results have been inferred from fits to scanning tunneling...
microscopy data below \( T_c \) \[14\], and photoemission data, both below \( T_c \) \[15\] and also above \( T_c \) in the pseudogap phase \[10\].

Fig. 2 demonstrates the different dispersions in the pseudogap phase along two momentum cuts, one where the spectrum is gapped (cut 1 in Fig. 2(e)) and the other which intersects the gapless Fermi arc (cut 2 in Fig. 2(e)). As the spectra were acquired at \( T = 49K \), there is appreciable thermal population above the Fermi energy (\( E_F \)), which allows us to follow the dispersions through \( E_F \). To trace the dispersion in the vicinity of \( E_F \) (Fig. 2(j)), we divide the raw ARPES spectra by the instrumental resolution broadened Fermi function, and then follow the peak positions of the divided spectra. When the spectral peak is weak and sits on a sloped background (for the lowest two curves in Fig. 2(d)), we first fit the spectrum with polynomials to high precision and then use the second derivative of the fitted curve to determine the peak position. Along cut 2, the spectral peak of the Fermi function divided data continuously moves to higher energies and crosses \( E_F \) at the underlying \( k_F \) (Fig. 2(i)). The dispersion resembles that of a normal metal. On the other hand, the dispersion along cut 1 shows a remarkable difference to that along cut 2. As shown in Fig. 2(c), from top to bottom curves, the spectral peak approaches \( E_F \) before \( k \) reaches the underlying \( k_F \) and then it recedes to higher binding energies and loses spectral weight. Fig. 2(d) shows the Fermi function divided data along cut 1. The dispersion along this cut has two branches, separated by an energy gap, moving in opposite directions; one is above \( E_F \) and the other is below. As \( k \) approaches the underlying \( k_F \) from the occupied side, the spectral weight of the upper branch increases. The spectral peaks of the two branches have approximately the same weight at \( k_F \), and at the same location, the energy gap between the two branches is minimal, giving rise to a flat topped spectrum. All of this is characteristic of the Bogoliubov-like dispersion seen previously below \( T_c \) \[17\] \[18\]. But, away from \( k_F \), the peaks in the lower and upper branches are slightly asymmetric in energy relative to \( E_F \) (Fig. 2(j)). This may result from an asymmetry of the self-energy in the pseudogap phase, which would act to broaden the spectral peaks and shift them to slightly higher binding energy on the occupied side. To illustrate this, in Fig. 2(d), we overlay the spectra obtained in the superconducting state (the thin lines) on those in the pseudogap phase. It can be seen that the peak positions of the Fermi function divided spectra in the superconducting state are at a lower binding energy, and the dispersion is now symmetric relative to \( E_F \) to that of the upper branch determined in the pseudogap phase. It should be mentioned that in the gapped region of the zone in the pseudogap phase, all dispersions obtained from the Fermi function divided spectra show a back bending at \( k_F \) (Fig. 2(j)), one of the characteristics of the Bogoliubov-like dispersion. However, when the energy gap amplitude becomes too large near the anti-node, only the lower branch of the dispersion can...
involve mixtures of electron and hole states, the spectral peak positions. (e) Fermi surface and locations of cuts 1 and 2. The thick line centered at the node indicates the gapless Fermi arc. (f)-(i): the same as (a)-(d), but the spectra are along cut 2 in (e). (j) The dispersions in the gapped region of the zone obtained from the Fermi function divided spectra. The pair of closed circles are the two branches of the dispersion derived from (d) at 49K, the dispersion indicated with open circles is along the same momentum cut shown in Fig. 3(e) on a heavily overdoped Bi2212 sample with $T_c = 65K$ and a maximum superconducting gap, $\Delta_{\text{max}} = 24$ meV, similar to that of underdoped La$_{1.885}$Sr$_{0.115}$CuO$_4$. Above $T_c$, the spectral peak of the Fermi function divided spectra shows a linear dispersion going through $k_F$ (Fig. 3(g), (i)). Below $T_c$ (Fig. 3(b), (d)), the superconducting gap opens up and the linear dispersion of the normal state transforms into the Bogoliubov dispersion of the superconducting state: $E_k = \pm \sqrt{\epsilon_k^2 + \Delta_k^2}$, where $\epsilon_k$ is the energy band dispersion in the normal state and $\Delta_k$ is the gap function. The double branches in the electronic excitation spectra involve mixtures of electron and hole states, the spectral weight in the lower (upper) branch is proportional to $v^2$ $(u^2)$ through the relation $u^2 = 1 - v^2 = \frac{1}{4}(1 + \epsilon_k/E_k)$, where $u$ and $v$ are the coherence factors [17, 18]. At the normal state $k_F$, the lower (upper) branch of the Bogoliubov dispersion reaches its maximum (minimum), and the two branches have equal spectral weight. Remarkably, like in overdoped Bi2212, the two branches of the dispersion along cut 1 in the pseudogap phase of underdoped La$_{1.885}$Sr$_{0.115}$CuO$_4$ possess all of these properties.

To summarize, our main experimental findings are: 1) beyond the gapless Fermi arc, the pseudogap above $T_c$ has the same simple $d$-wave form as the superconducting gap below $T_c$, 2) above $T_c$ there exists a Bogoliubov-like dispersion near the underlying $k_F$ where the spectra are gapped, and 3) the same underlying Fermi surface was obtained both in the superconducting state and in the pseudogap phase. In the pseudogap phase, the low energy electronic excitations along this cut are well defined both in energy and in momentum (Fig. 2(a)-(d)). Thus our experimental results can readily rule out the possibility that the pseudogap in underdoped cuprates only exists in the anti-nodal region, which has been attributed to a competing order such as charge ordering [20]. Our experimental findings are also inconsistent with a wide range of scenarios where the pseudogap originates from some ordering phenomenon associated with a non-zero $Q$ vector [21]. In our ARPES spectra, we do not find any
signatures of additional (shadow) states that are displaced by a non-zero $Q$, neither in the pseudogap phase nor in the superconducting state. It should be mentioned that in the pseudogap phase, the dispersion in the anti-nodal region also bends back at the underlying $k_F$ (Fig. 2(j)). However, due to the large amplitude of the pseudogap near the anti-node, the upper branch of the Bogoliubov-like dispersion is not thermally populated at 49K, and thus cannot be identified by Fermi function division.

Our experimental results support the idea that the pseudogap originates from preformed Cooper pairs for $T > T_c$. However, because the energy gap is larger than the phase stiffness in the pseudogap phase, the preformed pairs have a finite lifetime and can not travel in the crystal coherently. The similarity between the Bogoliubov-like dispersion in the pseudogap phase of underdoped La$_{1.895}$Sr$_{0.105}$CuO$_4$ and the dispersion of Bogoliubov quasiparticles in the superconducting state of heavily overdoped Bi2212 provides direct evidence that the pseudogap in underdoped cuprates arises from pairing of electrons, and thus the pseudogap phase is a state precursor to superconductivity. Because the superconducting and pseudogaps have the same simple $d$-wave form along the underlying Fermi surface beyond the Fermi arc (Fig. 1(ii)), it is difficult to image that there is some $k_F$ located between the anti-node and the end of the Fermi arc at which the pseudogap changes its nature from preformed Cooper pairs near the arc to competing order near the anti-node. It is more reasonable to infer that there is a single pseudogap above $T_c$ which transforms into the superconducting gap below $T_c$.

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