

# Quantitative determination of pairing interactions for high-temperature superconductivity in cuprates

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A profound problem in modern condensed matter physics is discovering and understanding the nature of fluctuations and their coupling to fermions in cuprates, which lead to high-temperature superconductivity and the invariably associated strange metal state. We report the quantitative determination of normal and pairing self-energies, made possible by laser-based angle-resolved photoemission measurements of unprecedented accuracy and stability. Through a precise inversion procedure, both the effective interactions in the attractive d-wave symmetry and the repulsive part in the full symmetry are determined. The latter is nearly angle-independent. Near  $T_c$ , both interactions are nearly independent of frequency and have almost the same magnitude over the complete energy range of up to about 0.4 eV, except for a low-energy feature at around 50 meV that is present only in the repulsive part, which has less than 10% of the total spectral weight. Well below  $T_c$ , they both change similarly, with superconductivity-induced features at low energies. Besides finding the pairing self-energy and the attractive interactions for the first time, these results expose the central paradox of the problem of high  $T_c$ : how the same frequency-independent fluctuations can dominantly scatter at angles  $\pm\pi/2$  in the attractive channel to give d-wave pairing and lead to angle-independent repulsive scattering. The experimental results are compared with available theoretical calculations based on antiferromagnetic fluctuations, the Hubbard model, and quantum-critical fluctuations of the loop-current order.

## INTRODUCTION

Quantitative analysis of very precise tunneling experiments, conducted by McMillan and Rowell (1) using the Eliashberg theory (2, 3), decisively confirmed that the exchange of phonons with fermions is responsible for the conventional s-wave pairing in metals such as Pb. Tunneling experiments integrate over the momentum dependence of many-body effects. This is sufficient for s-wave superconductors because the normal and superconducting properties have the full symmetry of the lattice. For any superconductor, the dependence of the normal self-energy  $\Sigma(\mathbf{k}, \omega)$  on the momentum  $\mathbf{k}$  has the full symmetry of the lattice; however, for high-temperature superconductors such as cuprates, the dependence of the pairing self-energy  $\phi(\mathbf{k}, \omega)$  on  $\mathbf{k}$  has a  $B_{1g}$  or  $d_{x^2-y^2}$  symmetry. Correspondingly, the effective interaction spectrum is characterized by two functions—(i) with the full symmetry of the lattice, which we will call the normal Eliashberg function  $\epsilon_N(\mathbf{k}, \omega)$ , and (ii) with the pairing symmetry, which we will call the pairing Eliashberg function  $\epsilon_P(\mathbf{k}, \omega)$ —rather than by a single function sufficient for s-wave pairing, which is often denoted by  $\alpha^2F(\omega)$  (1). The much more sophisticated angle-resolved photoemission spectroscopy (ARPES) experiments (4, 5) are then required because both the momentum dependence and the frequency dependence of the interactions are necessary to decipher the fundamental physics. The procedure used for determining fluctuations using the

Eliashberg theory remains valid for the pairing mediated by collective fluctuations, even when their high-energy cutoff is comparable to the electronic bandwidth.

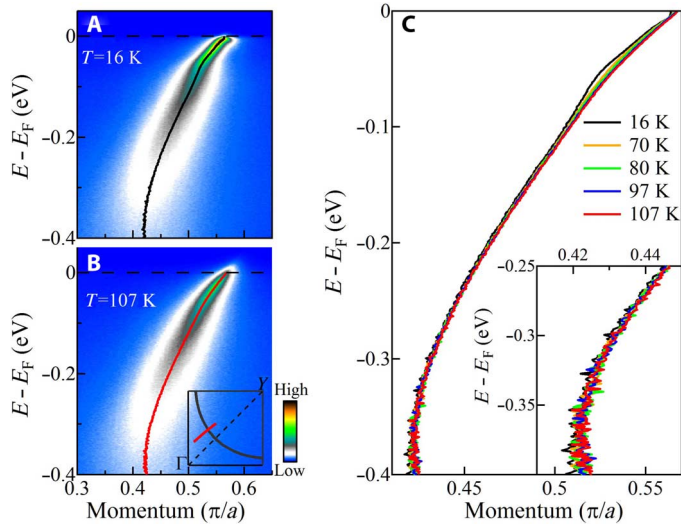
## RESULTS OF ARPES EXPERIMENTS

The procedure for extracting normal and pairing self-energies is described in section SI. The conditions required for ARPES to quantitatively yield the electron self-energies are very demanding. We seek to measure the absolute magnitude of the photoelectron current per unit-photon flux at various temperatures above and below  $T_c$ , at various angles, and across a range of frequencies extending to the upper cutoff of the fluctuations. Requirements on the data, estimates of signal-to-noise errors and systematic errors, partial correction of systematic errors, and limits of the validity of our results and analysis are given in section SII.

We carried out high-resolution laser-ARPES measurements on two  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  (Bi2212) samples—one slightly underdoped with a  $T_c$  of 89 K (UD89 hereafter) and the other overdoped with a  $T_c$  of 82 K (OD82 hereafter)—along various momentum cuts and at various temperatures. Some of the data on UD89 (6) and OD82 (7) (only for energy  $\leq 0.1$  eV below  $T_c$ ) were reported earlier, but without the analysis (and extension to higher energy) essential for deciphering the physics, as presented in this work. Figure 1 shows an example of the measured data of photoemission intensity as a function of momentum and energy, along a momentum cut marked in the inset to Fig. 1B, at temperatures well below  $T_c$  (Fig. 1A) and above  $T_c$  (Fig. 1B) in the UD89 sample. As shown in Fig. 1C, the data at different temperatures overlap each other at high binding energies extremely well, showing

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**Fig. 1. Color representation of the measured photoemission intensity of the UD89 sample along the  $\theta = 35^\circ$  direction. (A)  $T = 16$  K. (B)  $T = 107$  K. (C) Progression of energy-momentum dispersions at temperatures of 16, 70, 80, 97, and 107 K. The inset to (C) presents, on an expanded scale, an illustration of the consistency of the data up to an accuracy of  $5 \times 10^{-3}(\pi/a)$  in the region at high energy where no temperature-dependent corrections to the dispersion are necessary. In section SII, we show the systematic errors in the data when such accuracy is not met and how we correct them.**

their high quality and reproducibility. For some momentum cuts where the dispersions show small drifts with temperature and for other systematic errors, we used a method of data correction (section SII).

The suggested strategy (8) for extracting many-body effects relies on momentum distribution curves (MDCs), which represent the intensity of photoelectrons as a function of momentum  $k_\perp$  perpendicular to the Fermi surface for various fixed energies (for example, across the horizontal cuts in Fig. 1A or Fig. 1B). The two-dimensional momentum  $\mathbf{k}$  is represented by the angle  $\theta$  with respect to the crystalline axis and the magnitude  $k_\perp$  measured from the  $(\pi/a, \pi/a)$  point, as shown in Fig. 2C. In Fig. 2A, we present the measured MDC for UD89 for one of the trajectories across the Fermi surface at five energies  $\omega$  in the normal state above  $T_c$  (red circles) and in the superconducting state at 16 K (blue circles). These MDCs are from more than 5000 such plots taken; the results presented here are derived from the analysis of such plots in the two samples at various temperatures, angles, and energies. The signal-to-noise ratio of the fit in Fig. 2A may be best appreciated in Fig. 2B, where the normalized difference in the measured MDC intensities between 16 and 97 K is compared to the same function calculated from the fits in Fig. 2A. This represents the best results we have obtained; acceptable results are shown for the OD82 sample in fig. S1.

## NORMAL AND PAIRING SELF-ENERGIES

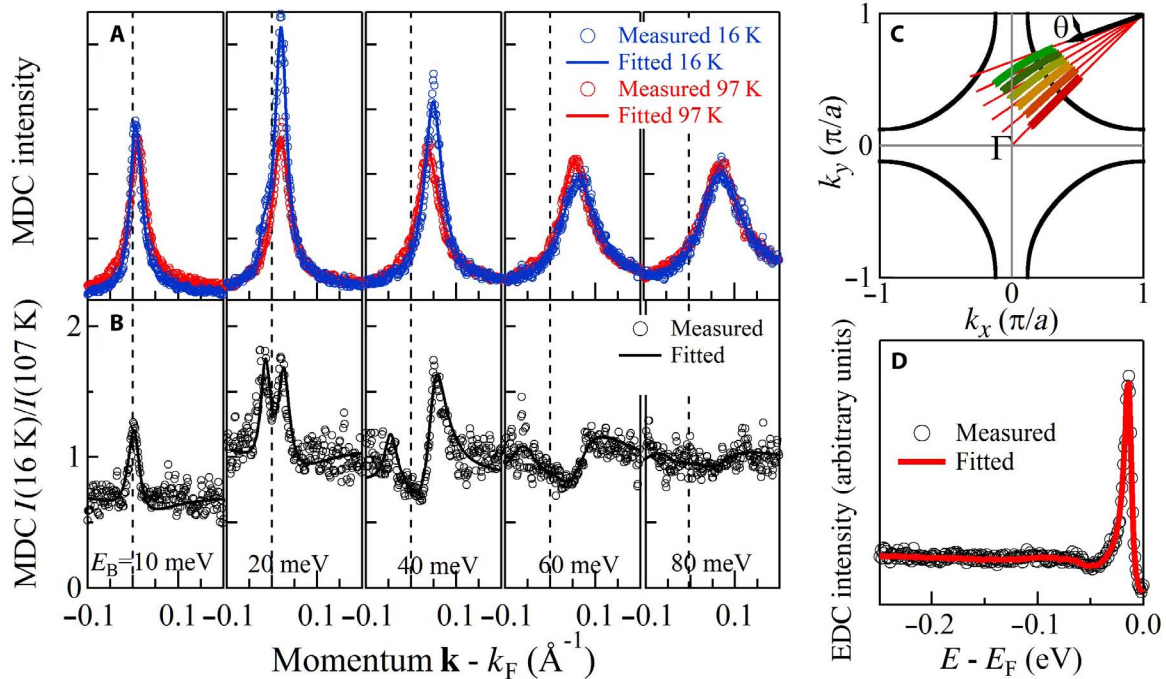
The relation of the measured photoelectron intensity  $I(\mathbf{k}, \omega)$  to the spectral function  $A(\mathbf{k}, \omega)$  is described in the Supplementary Materials, where we also present the procedure for extracting the normal self-energy  $\Sigma(\mathbf{k}, \omega)$  and the pairing self-energy  $\phi(\mathbf{k}, \omega)$  by fitting the MDCs. Representative fits are shown in Fig. 2A. The MDCs in the normal

state in Fig. 2A (red circles) over a wide region of energy and momentum are very well represented by Lorentzians as a function of  $k_\perp$ . This is true precisely (9) only if  $\Sigma(\mathbf{k}, \omega)$  is a function only of  $\theta$  and  $\omega$ . In the superconducting state, we fit the MDCs with  $\phi(\mathbf{k}, \omega)$  depending only on  $\theta$  and  $\omega$ , with almost equally good results, as shown in Fig. 2A (blue circles). As a further measure of confidence in the data and determination of the self-energies, we compare the measured photoemission spectrum [energy distribution curve (EDC)] at a fixed momentum to the EDC calculated from our fit to the MDCs at various energies in Fig. 2D.

We present the self-energies obtained directly from such fits in Fig. 3 so that the signal-to-noise ratio and the limits on the consistency of the data are directly visible. The evolution of the magnitude of the extracted normal and pairing self-energies is shown as a function of energy at various temperatures in Fig. 3 (A and B, respectively) for  $\theta = 20^\circ$  in OD82. The pairing self-energy measured at various  $\theta$  at a temperature of 16 K for the UD89 sample is shown in Fig. 3C. Note that  $\phi(\theta, \omega)$  has been scaled by  $\cos(2\theta)$ . Within the uncertainties in the data, the conclusions are the same if we scale instead by the appropriate d-wave basis for a square lattice  $[\cos(k_x a/\pi) - \cos(k_y a/\pi)]$ .

The self-energies near  $T_c$  are sufficient to deduce the effective interactions leading to the value of  $T_c$ . Near  $T_c$ , the real and imaginary parts of both normal and pairing self-energies are smooth functions of energies up to high energies (and angles). However, as  $T$  decreases below  $T_c$ , one finds in Fig. 3 two low-energy features below  $\sim 75$  meV. It was suggested (10) and experimentally shown (11) that forward scattering (9) from impurities lying in between the  $\text{CuO}_2$  planes produces the low-energy peak at  $\sim 15$  meV in the self-energies at the superconducting state. The other structure at about 65 meV is expected for all cases in which the fluctuations are due to the interactions among the fermions themselves (12, 13) because the opening of the superconducting gap  $\Delta$  diminishes the spectral weight at energies below  $O(2\Delta)$  and piles it up at higher energies. This process occurs in addition to the generalization to d-wave superconductors of the shift of the self-energies by  $\Delta$ , which is well known in phonon-mediated s-wave superconductors. These two superconductivity-induced features are irrelevant in determining the value of  $T_c$ , although they are quite important in determining the temperature dependence of the superconducting gap, which is not our focus in this paper.

In Fig. 3A, except for the low-energy features, the normal (real and imaginary) self-energy  $\Sigma(\theta, \omega)$  is nearly independent of temperature. The imaginary part  $\Sigma_2(\theta, \omega)$  varies linearly with  $\omega$  to a good approximation, as in the normal state. In the same energy range,  $\Sigma_2(\theta, \omega)$  is also nearly independent of  $\theta$ , as already noted by Valla *et al.* (14), Kaminski *et al.* (15), and Bok *et al.* (16). On the other hand, not surprisingly, the real and imaginary parts of the pairing self-energy  $\phi(\theta, \omega)$  in Fig. 3B systematically increase with decreasing temperature below  $T_c$  with saturation at low temperatures. Except for the low-energy features, the imaginary part of  $\phi(\theta, \omega)$  in Fig. 3B is weakly  $\omega$ -dependent up to about 0.2 eV, beyond which the signal-to-noise level does not allow quantitative conclusions. In Fig. 3C, we show that the real and imaginary parts of the pairing self-energy  $\phi(\theta, \omega)$  scaled by  $\cos(2\theta)$  over the energy range up to  $\sim 0.2$  eV are also independent of  $\theta$  up to about  $\pm 10\%$  in absolute value. This is an important check on data and analysis because, in these experiments, this is the quantity deduced with the largest error as it comes from the difference between the spectra below  $T_c$  and the spectra above  $T_c$ . At  $\theta = 20^\circ$ , the bottom of the band from the Fermi energy is  $\sim 0.2$  eV (17), which serves as the



**Fig. 2. Measured MDCs and their fits at five different energies at 97 and 16 K in the UD89 sample along the dark-green trajectory ( $\theta = 20^\circ$ ).** Such data were taken at 1-meV intervals and at all of the trajectories shown in (C). The vertical scale in (A) is the measured photoelectron current for a fixed photon flux (in arbitrary units). It is crucial that this scale remain within the error bars discussed in the text at all temperatures, energies, and momenta, and that any systematic errors be corrected. (B) Normalized difference in measurements taken at the two temperatures in (A) and at the same quantity calculated from the fits in (A). The fits to the MDCs were made according to the procedure described in fig. S1. The normal and pairing self-energies are extracted from such fits, also as described in fig. S1. (D) intends to show the consistency of the fits to the MDCs at different energies; it shows the EDC generated from the MDC within the complete range of energies measured at an angle  $\theta = 20^\circ$  at the Fermi surface at 16 K and compares direct measurements of the EDC at the same point and same temperature.

natural cutoff. Therefore, the complete fluctuation spectra have been accessed for this angle. From the measurements of  $\Sigma(\theta, \omega)$  above  $T_c$  in Bok *et al.* (16) and below  $T_c$  found here, the cutoff energy increases smoothly with increasing  $\theta$  (to  $\sim 0.4$  eV for  $\theta = 45^\circ$ ). It appears reasonable to assume that the pairing self-energy at other angles has the same cutoff as that of the normal self-energy, as it does at  $\theta = 20^\circ$ ; this can be verified by future experiments with better signal-to-noise ratios for  $\phi(\theta, \omega)$  at higher energies,  $\theta$  values closer to  $45^\circ$ , and  $T$  values closer to  $T_c$ .

## NORMAL AND PAIRING ELIASHBERG FUNCTIONS

Important conclusions about the fundamental physics of cuprates can already be reached from the self-energies (Fig. 3), which have been directly extracted from the experimental data. Reaching some other conclusions requires solving the Eliashberg integral equations for anisotropic superconductivity, described in section SIII. In section SIII, we show, using the experimentally obtained normal and pairing self-energies as inputs, that the determination of the Eliashberg functions  $\epsilon_N(\theta, \omega)$  and  $\epsilon_P(\theta, \omega)$  is limited only by the accuracy of the self-energies and by the procedure for solving the integral equations. We will also provide a self-consistency check on the validity of this procedure using the experimental results.

$\epsilon_N(\theta, \omega)$  and  $\epsilon_P(\theta, \omega)$  are deduced from the measured self-energies  $\Sigma(\theta, \omega)$  and  $\phi(\theta, \omega)$  in Fig. 3 through solution of the integral equations using the maximum entropy method (18). To avoid instabilities in the numerical solutions of these equations using such a procedure, we

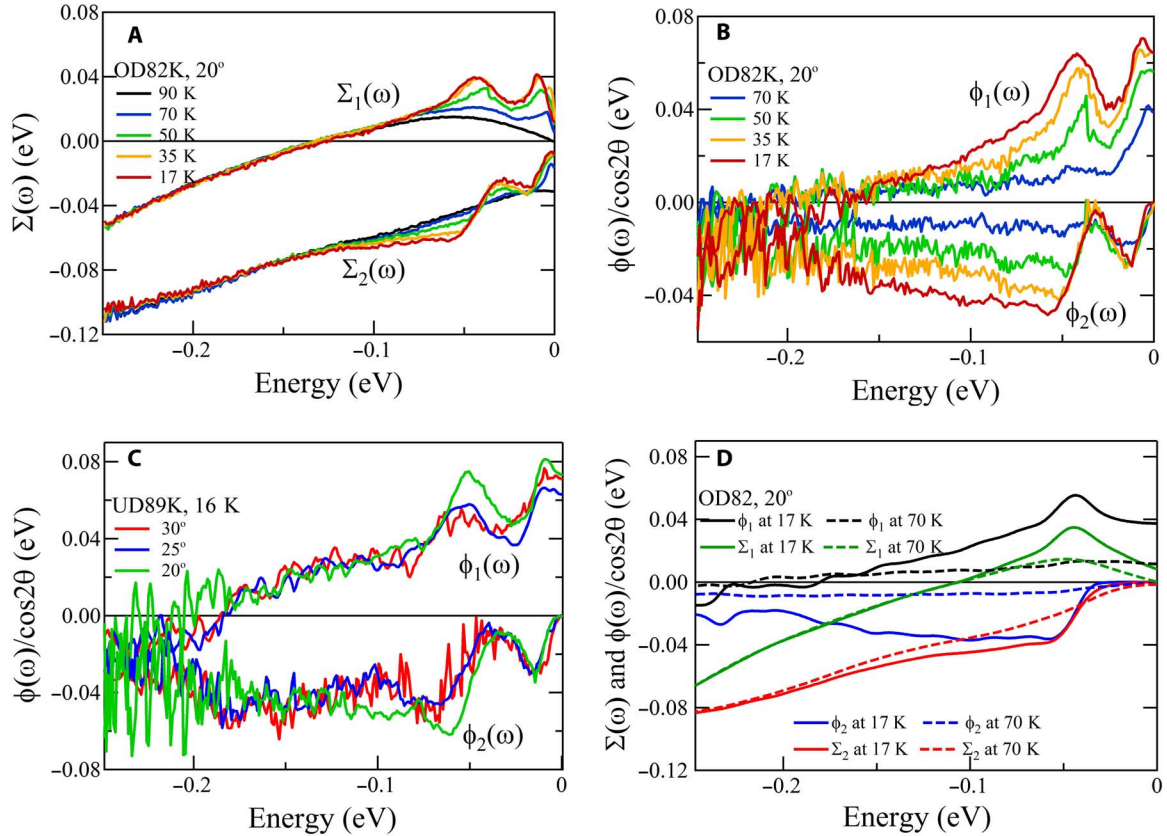
smoothened the raw data of self-energies (such as those shown in Fig. 3, A to C) at each energy by averaging the data in the range of  $\pm 5$  meV around it (as exemplified in Fig. 3D). The results obtained are shown in Fig. 4 for both UD89 and OD82. Despite the smoothing of self-energy, we found weak oscillations of about 10% magnitude in  $\epsilon_N$  and  $\epsilon_P$ , which are artifacts from the maximum entropy method. In Fig. 4, we plot  $\epsilon_N(\theta, \omega)$  and the scaled quantity  $\epsilon_P(\theta, \omega) \equiv \epsilon_P(\theta, \omega)/\cos(2\theta)$ .

Let us start with Fig. 4C, which presents results close to  $T_c$ . The normal-state bump at  $\sim 50$  meV in  $\epsilon_N(\theta, \omega)$  hardly changes for  $T \lesssim T_c$  and is absent in  $\epsilon_P(\theta, \omega)$ . If we ignore the bump,  $\epsilon_P(\theta, \omega) \approx \epsilon_N(\theta, \omega)$  to within 10% accuracy. As  $T$  decreases well below  $T_c$ , both functions develop a peak in the region around 50 to 75 meV, as shown in Fig. 4 (A and B). These are related to the superconductivity-induced features in the self-energies that we have already discussed. The result that  $\epsilon_P(\theta, \omega)$  loses the low-energy feature as  $T \rightarrow T_c$  is highlighted in Fig. 4F, where its evolution with temperature is shown.

For comparison, we also include  $\epsilon_N(\theta, \omega)$ , which was deduced from its self-energy in the normal state at various angles by the same methods used in Fig. 4 (D and E). These results are consistent with earlier deductions (19) from ARPES data along the diagonal ( $\theta = 45^\circ$ ) direction, derived using the same technique and by fitting the measured optical spectra in the normal state (20, 21), which are averages over all angles weighted by their Fermi velocity.

In Fig. 5, we calculate the real and imaginary parts of the pairing self-energy  $\phi(\theta = 20^\circ, \omega)$  from the deduced  $\epsilon_N$  at 70 K (Fig. 4C) and at 35 K





**Fig. 3. Normal and pairing self-energies.** (A and B) The evolution of the extracted normal self-energy (A) and the evolution of the pairing self-energy (B) as a function of temperature directly from the fits to the MDCs in OD82. The normal and pairing self-energies show superconducting gap-induced features at low energies up to about  $3\Delta$  and smoothly vary in energy thereafter up to a cutoff energy. (C) The pairing self-energies in UD89 at 16 K divided by  $\cos(2\theta)$ . Determination of the pairing self-energy has acceptable signal-to-noise ratios up to about 0.2 eV only. The data fall together at the angles shown (up to an accuracy better than 10%) up to about 0.2 eV. (D) The self-energies smoothed over  $\pm 5$  meV (as discussed in the text) and, after removing the impurity, induced features for OD82 at  $T = 17$  K (solid lines) and  $T = 70$  K (dashed lines).

(Fig. 4B) by using the Eliashberg equations and by assuming that  $\epsilon_P = \epsilon_N$ . The calculations (Fig. 5, solid lines) are directly compared with the extracted values (Fig. 5, circles and squares). Because  $\phi \ll \Sigma$  near  $T_c$ ,  $\epsilon_N$  here is determined primarily by  $\Sigma$ . The measured  $\phi$  determines the deduced  $\epsilon_P$  using the Eliashberg equations. Therefore, the success of the comparison depends on both (i)  $\epsilon_N \approx \epsilon_P$  near  $T_c$ , except for the small bump near 50 meV in  $\epsilon_N$ , and (ii) the applicability and mutual consistency of the Eliashberg equations for the normal and pairing self-energies at a similar accuracy.

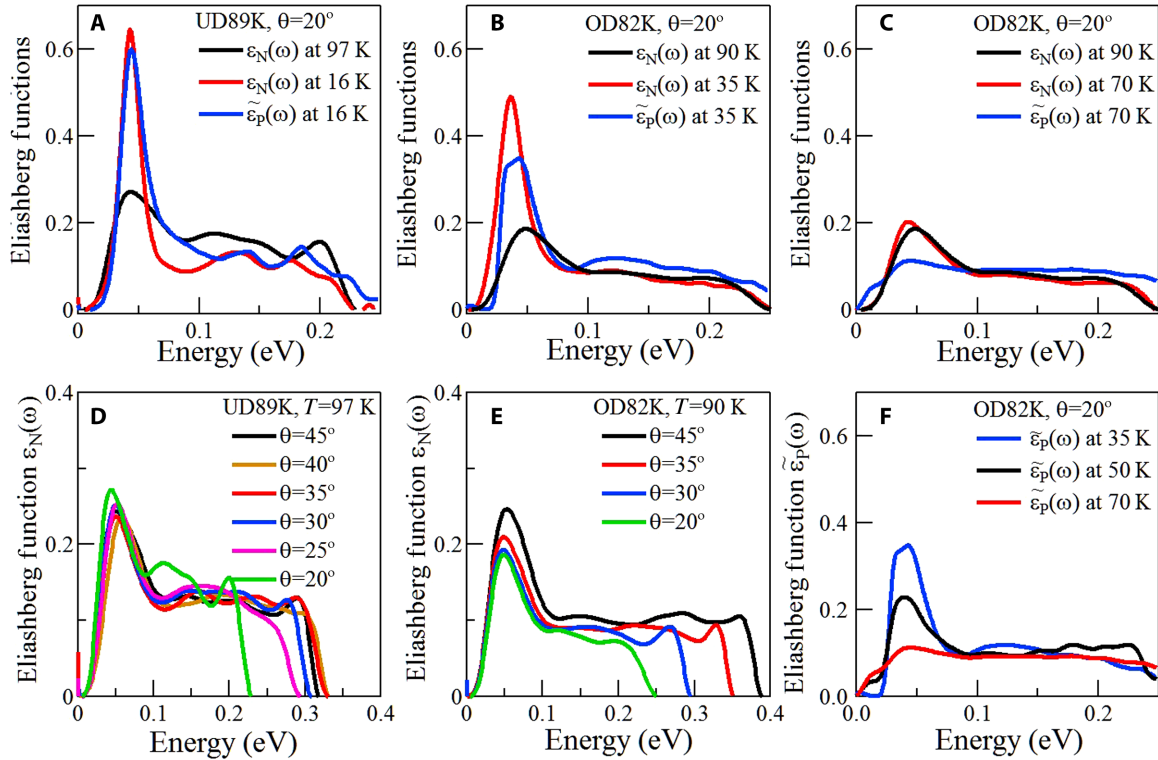
### SALIENT POINTS OF THE EXPERIMENTAL RESULTS AND THEIR IMPLICATIONS

We extracted the electron self-energies in both pairing and full lattice symmetries directly from the ARPES data without adjustable parameters using the procedure described in section SII. We then used the integral equations, which are shown in section SIII, to numerically deduce both the pairing and the normal Eliashberg functions. No theoretical assumptions underlie our results, except that superconductivity is due to generalized BCS (Bardeen-Cooper-Schrieffer) pairing. We summarize here the principal conclusions of our data and analysis.

(A) The experimental results and their fits in Fig. 2 show that the imaginary part of the normal self-energy above is independent of  $\mathbf{k}$  and linear in  $\omega$  to a good approximation, as found earlier (14, 15). The strange metal anomalies (such as the linear-in- $T$  resistivity) and corresponding aspects in optical conductivity follow (9). The real and imaginary parts of both the normal and the pairing self-energies acquire superconductivity-induced features at low energy below  $T_c$ . The pairing self-energy near  $T_c$  is nearly a constant as a function of  $\omega$  up to the upper cutoff and acquires the same superconductivity-induced features below  $T_c$ , as in the normal self-energy.

(B) In comparison with Fig. 4, one finds that, near  $T_c$ ,  $\epsilon_P(\theta, \omega) \approx \epsilon_N(\theta, \omega)$  to within the stated accuracy, except for the small bump near 50 meV in the latter. The part of these (dimensionless) quantities that is nearly a constant as a function of  $\omega$  has the same magnitude as that of the normal state  $\epsilon_N$ , and has a value of about 0.15 for UD89 and a consistently somewhat smaller value for OD82 up to the angle-dependent cutoff, which is about 0.2 eV at 20°, moving continuously to about 0.4 eV at 45°. Additional superconductivity-induced features in both functions are observed as temperature decreases well below  $T_c$ ; within our accuracy, they have the same frequency dependence.

(C) Above  $T_c$ ,  $\epsilon_N(\theta, \omega)$  consists of a low-energy bump at about 50 meV with a half-width of about 10 meV on top of a nearly constant part up to



**Fig. 4. Eliashberg functions: Normal [ $\epsilon_N(\theta, \omega)$ ] and scaled pairing [ $\epsilon_P(\theta, \omega) \equiv \epsilon_P(\theta, \omega)/\cos(2\theta)$ ].** These are calculated using the solution for the Eliashberg equations from the measured self-energies. (A) and (B) compare  $\epsilon_P(20^\circ, \omega)$  and  $\epsilon_N(20^\circ, \omega)$  deep in the superconducting state for the two samples, and the latter also above  $T_c$ . At low temperatures, they are of the same accuracy over the whole frequency range, with a large superconductivity-induced enhanced low-energy peak. (C) Closer to  $T_c$ , the low-energy peak in  $\epsilon_P(\omega)$  disappears. This trend is shown more directly in (F). (D) and (E) give  $\epsilon_N(\theta, \omega)$  for  $T$  above  $T_c$ , showing the increase in cutoff energy with increasing  $\theta$ . The gentle waviness in all of the results represents artifacts of the maximum entropy method for the solution of the Eliashberg equations.

the angle-dependent cutoff,  $\epsilon_P(\theta, \omega)$ , which can be deduced only below  $T_c$ , has no low-energy bump near  $T_c$ . This means that coupling of fermions in the attractive d-wave channel to excitations that appear in the bump does not occur, but that coupling of fermions to such excitations in the s-wave channel does occur. On the other hand, the nearly constant part has a similar magnitude of coupling to fermions in both the s-wave channel and the d-wave channel.

(D) It is well understood (22, 23) that d-wave pairing [that is,  $\phi(\theta, \omega) \propto \cos 2\theta$ ] is favored only when fermions scatter dominantly over angles of  $\pm \pi/2$ . Together with point (C), this exposes the central paradox of d-wave superconductivity in cuprates: the fundamental physics of cuprates requires that the same fluctuations that dominantly scatter at angles  $\pm \pi/2$  in the attractive channel lead to a nearly angle-independent repulsive scattering in the normal channel with the full symmetry of the lattice.

It is reasonable to assume that the bump in the normal state is due to optical phonons of apical oxygens, as has been suggested (24). In an interesting time-resolved conductivity experiment (21) at room temperature, the results were analyzed with fluctuations that could be divided into a peak at around 50 meV and a broad electronic continuum with a cutoff at about 0.4 eV; the bump has a relaxation rate that is much slower than the continuum, indicating independent sources for the two contributions. Our results for  $\epsilon_N$  are consistent with these conclusions with the additional information (through  $\epsilon_P$ ) that the feature around 50 meV has no attractive coupling in the d-wave channel.

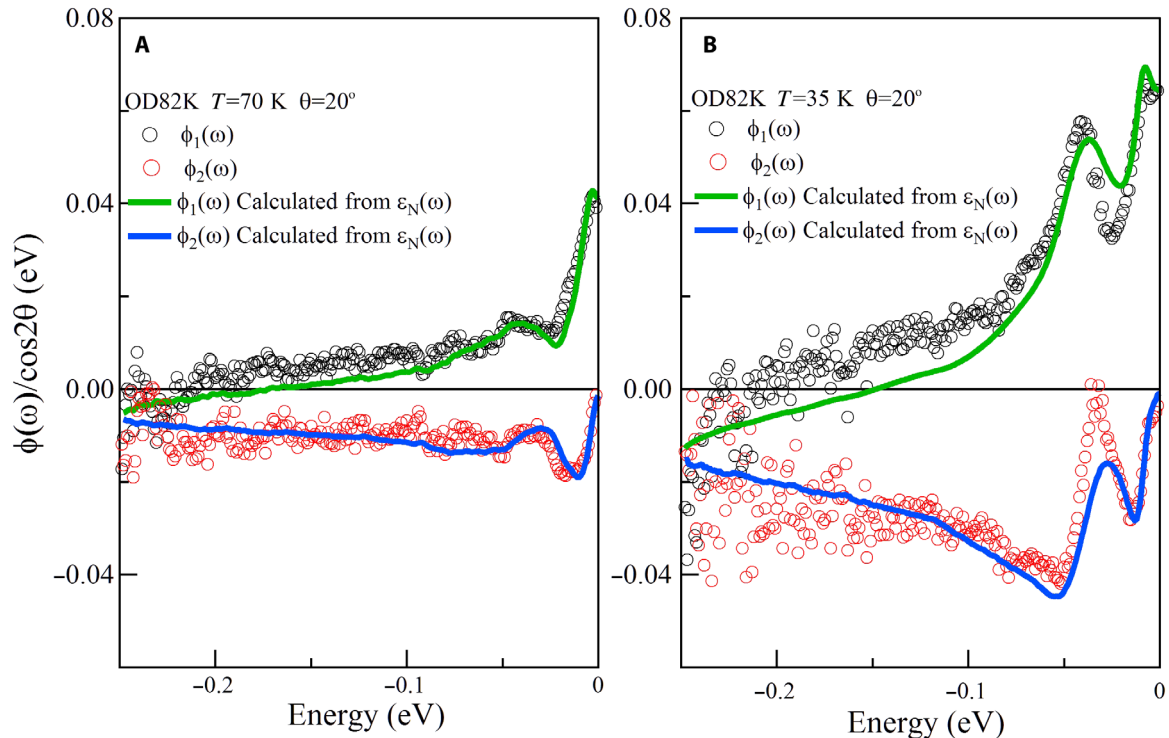
### Determining $T_c$

One may wish to calculate  $T_c$  directly from the deduced Eliashberg functions. Such a check, however, is circular because the Eliashberg functions are obtained from the solution of equations whose linearized version yields  $T_c$ . Thus, if the complete information over the entire Brillouin zone were available, using the Eliashberg function near  $T_c$  back in the linearized Eliashberg equations would give the experimental  $T_c$ . Such an exercise may, however, be taken as a test of several of the steps in extracting the final results from the experiments and as a test of the extrapolations. Using the extrapolations for  $\epsilon_P$  from the angles measured (so that their upper cutoff at other angles is also the same as the measured cutoff of  $\epsilon_N$ ) and extrapolating from the measured angles to all angles, the linearized Eliashberg equations for deductions, using  $\xi(\mathbf{k})$  of Markiewicz *et al.* (25), indicate that  $T_c \approx 135$  K for the UD89 sample and  $T_c \approx 90$  K for the OD82 sample.

We can obtain estimates of the dimensionless coupling constants in the s-wave and d-wave channels by using their approximate expressions (26, 27)

$$\begin{aligned} \lambda_s &\approx \langle \int_0^\infty d\omega \frac{2}{\omega} \epsilon_N(\theta, \omega) \rangle_\theta \\ \lambda_d &\approx \langle \int_0^\infty d\omega \frac{2}{\omega} \cos(2\theta) \epsilon_P(\theta, \omega) \rangle_\theta \end{aligned} \quad (1)$$

Using the result in Fig. 4C—that, near  $T_c$ ,  $\epsilon_N(\theta, \omega) \approx \epsilon_P(\theta, \omega) \approx 0.15$  from about  $T_c$  up to the cutoffs  $\omega_c(\theta)$  and  $\approx 0.15\omega/2T$  for  $\omega \lesssim 2T$ —yields



**Fig. 5. Calculation of pairing self-energies assuming that  $\varepsilon_p = \varepsilon_N$ .** (A and B) The experimentally deduced real and imaginary parts of self-energy (in absolute units) are compared with a calculation of the same quantity from the Eliashberg equations, assuming that  $\varepsilon_p = \varepsilon_N$  for (A)  $T = 70$  K and (B)  $T = 35$  K in the OD82 sample. This agreement occurs only if the Eliashberg equations are applicable for the analysis of the data and for the relation of the two Eliashberg functions.

$\lambda \approx 2\lambda_d \approx 1.2$ . In materials such as cuprates, where pair-breaking due to inelastic scattering is important, these parameters alone do not determine  $T_c$  (27). However, the deduced fluctuations provide an enhancement  $O(\log(\omega_c/T_c))$  of the effective coupling constants, which are crucial factors in the proper determination of  $T_c$  for the kind of fluctuation spectrum deduced.

### Brief comparison of cuprate models with the experimental results

The results in the literature for calculations starting with different physical ideas are compared to the experiments in section SIV. We summarize the comparisons here.

All calculations use adjustable parameters with which features of the experiments, such as  $T_c$ , may be reproduced. The comparison with experimental results must then be performed with respect to the momentum and frequency dependence of the pairing and normal self-energies and Eliashberg functions noted in principal conclusions (A) to (D).

(i) The calculations (28) using measurements (29) of antiferromagnetic spin fluctuations [in  $(\text{La}_{2-x}\text{Sr}_x)\text{CuO}_4$  for  $T/T_c \approx 0.25$ ] in the Eliashberg theory correctly give  $\phi(\theta, \omega)$ , consistent with  $\propto \cos(2\theta)$ . The calculations give a peak in  $\phi(\theta, \omega)$ , reproduced in fig. S3 at about 0.1 eV and nearly zero for the pairing self-energy beyond it. In Figs. 2 and 4 of Hong and Choi (28),  $\Sigma(\omega, \theta)$  is not linear in  $\omega$  and is strongly angle-dependent. To come to conclusions about the applicability of the theory, we should compare these results with the results in Fig. 3, which has a constant part in  $\phi(\theta, \omega)/\cos(2\theta)$  up to the cutoff, a linear part in  $\omega$ , and a nearly angle-independent part in  $\Sigma(\omega, \theta)$  at all temperatures. Because

no measurements are available at higher  $T$ , comparisons near  $T_c$  are not possible.

(ii) From the results of very extensive dynamical mean-field theory calculations on the Hubbard model (30, 31), a value of the nearest-neighbor hopping parameter  $t \approx 0.3$  eV is chosen to obtain nearly the right maximum value of  $T_c \approx t/30$ . Only angle-averaged self-energies are calculated with this technique. The calculations give peaks in the pairing self-energy (reproduced in fig. S4) at energies of about  $0.2t$  and  $t$  (that is, between 0.06 and 0.3 eV), which sharply decrease to zero in between. The imaginary part of such a  $\Sigma(\omega)$  [see Fig. 5 in Gull and Millis (31)] is constant beyond  $\omega \approx 0.2t \approx 0.06$  eV, with a peak below this value. To come to conclusions about the inapplicability of the theory, we must compare these results with the experimental results in Fig. 3, which give, just below  $T_c$ , a linear part in  $\omega$  self-energy and a nearly constant pairing self-energy up to the high-frequency cutoff.

(iii) The spectra of loop-current fluctuations are calculated as the quantum-critical fluctuations of the dissipative quantum XY model. It is proportional to  $\tanh(\omega/2T)$  (32), with a high-frequency cutoff, which fits the deduced Eliashberg functions near  $T_c$  [except for the low-energy bump in  $\varepsilon_N(\omega)$ ]. It leads to the well-known angle and frequency dependence of the measured normal self-energies. In Fig. 5, we have shown, in effect, that it also yields the measured pairing self-energy near  $T_c$  and well below  $T_c$ . In section IV, we recapitulate earlier results (33) about the momentum dependence of matrix elements coupling the fermions to fluctuations of the model so as to give both repulsion in the normal channel and attraction in the pairing channel

with the same frequency-dependent spectra. The physics behind the central paradox thereby follows.

## MATERIALS AND METHODS

Optimally doped and slightly underdoped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  (Bi2212) single crystals were grown by the traveling solvent floating zone method. The overdoped Bi2212 sample was prepared by annealing the optimally doped sample under an oxygen atmosphere. All of the samples were of high quality, exhibiting sharp superconducting transitions with a transition width of  $\sim 2$  K.

Angle-resolved photoemission measurements were carried out in our vacuum ultraviolet laser-based angle-resolved photoemission system (34). The photon energy of the laser was 6.994 eV at a bandwidth of 0.26 meV, and the energy resolution of the electron energy analyzer (Scienta R4000) was set at 0.5 to 1 meV, giving rise to an overall energy resolution of  $\sim 1.0$  meV. The angular resolution was about  $0.3^\circ$ , corresponding to a momentum resolution of  $\sim 0.004 \text{ \AA}^{-1}$  at a photon energy of 6.994 eV. All samples were cleaved in situ and measured in vacuum with a base pressure better than  $5 \times 10^{-11}$  torr. More details about the sample and the experiments can be found in Zhang *et al.* (6, 35) and He *et al.* (7).

## SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at <http://advances.sciencemag.org/cgi/content/full/2/3/e1501329/DC1>

SI. Extraction of normal and pairing self-energies from ARPES in the superconducting state

SI. 1. Single-particle spectral function

SI. 2. Procedure for extracting self-energy

SII. Correction of systematic errors and renormalization of ARPES data

SII. 1. Limits of the validity of results

SIII. Equations for self-energy

SIII. 1. Exact representation of self-energy

SIII. 2. Familiar Eliashberg integral equations for d-wave superconductors

SIV. Comparison of theories with experimental results

Fig. S1. Data corrections that are necessary because of systematic errors attributable to the movement of the sample with change in temperature.

Fig. S2. Exact representation of the normal and pairing self-energies.

Fig. S3. Imaginary part of the pairing self-energy at various angles across the Fermi surface calculated by Hong and Choi (28) from the spin-fluctuation spectra in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  measured by Vignolle *et al.* (29) at optimal doping.

Fig. S4. Imaginary part of the gap function for the Hubbard model calculated by Gull and Millis (30) for various dopings indicated in the plot.

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## Quantitative determination of pairing interactions for high-temperature superconductivity in cuprates

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