# Photoemission Studies of the Pseudogap Regime in the High $T_c$ Cuprate Phase Diagram

**P.D. Johnson[1], H.B Yang[1], J.D. Rameau[1], G.D. Gu[1], T.E. Kidd[2], H. Claus[3] and D. Hinks[3]**

[1] Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory, Upton, NY
[2] Physics Department, University of Northern Iowa, Iowa
[3] Materials Science Division, Argonne National Laboratory

E-mail: pdj@lbnl.gov

**Abstract**. By examining thermally excited states above the Fermi level in angle resolved photoemission spectroscopy, we show that the disconnected Fermi arcs observed in the pseudogap state of the cuprate phase diagram are actually components of fully enclosed hole pockets. The spectral weight of these pockets is vanishingly small at the Luttinger surface, coincident with the antiferromagnetic zone-boundary formed by the underlying spin lattice, creating the illusion of the Fermi arcs. The area of the pockets as measured in this study is consistent with the doping level, and hence the carrier density, of the samples measured. Furthermore, the shape and area of the pockets is well reproduced by phenomenological models of the pseudogap phase that reflect its proximity to a Mott insulator.

## 1. Introduction

High $T_c$ superconductivity remains a challenge for the materials science community. Both the cuprates [1] and the more recently discovered Fe-based superconductors [2] present new and exciting phenomena that do not lend themselves to orthodox treatment. In the case of the cuprates, the superconducting phase emerges from a Mott insulator upon selective doping with charge carriers. However, in the case of the Fe-based materials, the superconductivity emerges from an underlying metallic state. It is therefore important to obtain a correct understanding of the normal states from which high temperature superconductivity emerges. Recognizing this, the pseudogap phase of the underdoped cuprates has been heavily studied. This region of the phase diagram presents its own challenges, including a gap in the spectral response even in the normal state [3], and a Fermi surface that seemingly consists of disconnected Fermi arcs.[4] The presence of this gap, or pseudogap, has promoted considerable discussion as to whether there exists some form of pairing of the carriers at temperatures above that at which the long range phase coherence associated with superconductivity develops [5], or whether there exists another ground state that competes with the superconductivity.[6,7] The disconnected Fermi arcs present another puzzle. Indeed, to date, there is no precedent in any other condensed matter system for such a phenomenon. Two candidate descriptions of the arc have emerged. In one picture superconducting fluctuations above Tc result in a d-wave gap with the finite arc reflecting closure of the gap in the nodal region due to classical thermal fluctuations.[8] In the other picture, as discussed later, the arcs represent

one side of a hole-pocket associated with the proximity to the Mott insulating state.[9-13] These are commonly referred to as one-gap or two-gap models, respectively.[14]

Quantum oscillation experiments appear to indicate the presence of just such small Fermi pockets in the underdoped regime and a large Fermi surface in the overdoped regime.[15] Within the quantum oscillation community, there have been several different models proposed for the nature of the pockets. However it is important to recognize that those experiments are carried out in high magnetic fields. A recent study has reported that such high magnetic fields can in themselves lead to a spurious reconstruction of the Fermi surface resulting in the formation of pockets.[16] In the present paper we discuss, in more detail, our recent Angle-Resolved Photoemission Spectroscopy (ARPES) studies of the underdoped Bi2212 cuprates, with a particular emphasis on the possibility of nodal hole pockets. As discussed in detail elsewhere, the existence of such pockets is consistent with the fact that cuprates are strongly correlated systems or doped antiferromagnetic Mott insulators.

## 2. Experimental Procedure

Photoemission, by its nature, requires an electron in the initial state. Thus, it is seen as one of the primary probes of the occupied electronic states below the chemical potential. However, techniques have been developed to enable some level of study of the states above the chemical potential, $E_F$. These include the recognition that the occupation probability of the states immediately below and above $E_F$ is determined by the finite width in energy of the Fermi-Dirac (FD) distribution function. Thus, access is provided to states above $E_F$ simply by dividing the ARPES spectrum by the FD function.[17,18] Another technique that has become very prevalent with studies of the high $T_c$ materials is the so-called symmetrization method.[4] Here one effectively forces a particle-hole symmetry onto the system and symmetrizes the data around the chemical potential. In the studies reported here, the spectra are analyzed by first removing any broadening due to experimental resolution, using the so called Lucy-Richardson technique,[19] and then dividing by the appropriate temperature dependent Fermi-Dirac distribution. As discussed elsewhere,[19] rather than simply dividing by a resolution broadened FD function, it is important to follow such a procedure, if one wants to avoid analysis induced binding energy shifts and false intensity variations in high resolution data.

The photoemission studies reported in this paper were carried out on underdoped cuprate samples, both Ca doped and oxygen deficient varieties. The Ca-rich crystal was grown from a rod with $Bi_{2.1}Sr_{1.4}Ca_{1.5}Cu_2O_{8+\delta}$ composition using an arc-image furnace with a flowing 20% O2-Ar gas mixture. The maximum $T_c$ was 80K for Ca doped samples. The sample was then annealed at 700$^o$C giving a 45K Tc with a transition width of ~2K. The oxygen-deficient $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) crystals were produced by annealing optimally doped Bi2212 crystals, at 450$^o$C to 650$^o$C for 3–15 days. The spectra shown in this paper were all recorded on beam line U13UB at the NSLS using a Scienta SES2002 electron spectrometer. Each spectrum was recorded in the pulse-counting mode with an energy and angular resolution of 15 meV and 0.1$^o$ respectively.

## 3. Results

Figure 1 shows spectra obtained after the analysis described above from the optimally doped Bi2212 ($T_c$=91K) and underdoped Bi2212 ($T_c$=65K) both in the normal state.[20] In both cases, the spectra were initially recorded with a photon energy of 16.5 eV and at a temperature of 140K. The spectra, after analysis, from the optimally doped materials show bands moving clearly through $E_F$ as one moves away from the nodal direction. This is consistent with the loss of superconductivity in the normal state. However the spectra, after analysis, from the underdoped material show a gap in the spectral function that moves to deeper binding energies as one moves away from the nodal direction. The latter gap is consistent with the findings of phenomenological models of the pseudogap phase such as the so-called YRZ model, which predict the presence of Fermi hole pockets with x-dependent area in the pseudogap phase.[10-12] The predicted band dispersions from the YRZ model [10] are shown in fig. 2 for a doping level of p = 0.1 or $T_c$ ~ 65K. The figure shows the calculated spectral function $A(\underline{k},\omega)$ moving away from the nodal direction using the expression $A(\underline{k},\omega) = -(1/\pi)\mathrm{Im}\, G(\underline{k},\omega)$

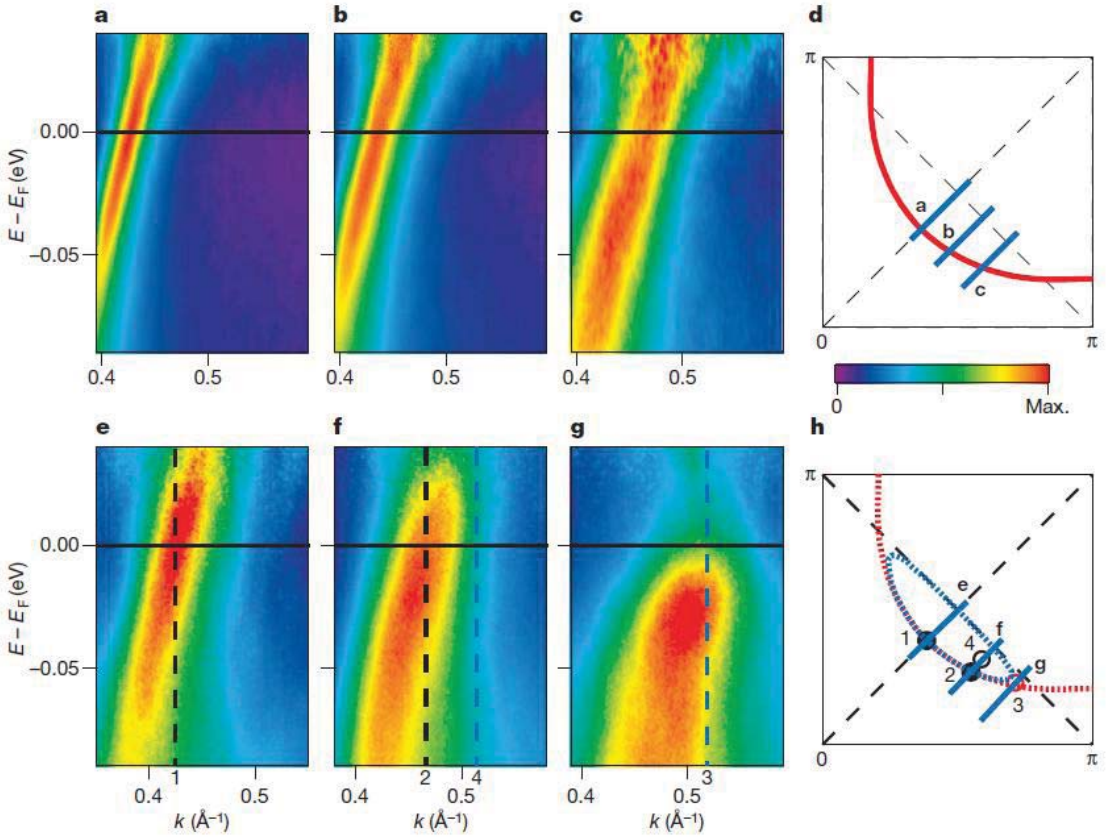


Fig 1. Reproduced from ref. [20], spectra, after analysis as described in the text, obtained as a function of angle α away from the nodal direction for optimally doped Bi2212 (upper panels) and the underdoped Bi2212 (lower panels) $T_c$ = 65K. The location of the different cuts in momentum space are indicated in the schematic diagrams on the right.

where the Green's function G(k,ω) is given by the form

$$G(k,\omega) = \frac{g_t}{\omega - \xi(k) - \Delta_R^2/[\omega + \xi_o(k)] + i\Gamma} + G_{inc} \qquad (1)$$

with the various terms defined in ref. [10]. We have introduced a broadening term Γ to allow for finite lifetime effects. We note here that the expression

$$\Delta_R^2/[\omega + \xi_o(\underline{k})] \qquad (2)$$

is effectively a real self-energy term reflecting a hole moving through the background of paired singlets characterizing the RVB state.[21] It is this term that is responsible for the vanishing back side of the pocket. Each band in fig. 2 is characterized by two Fermi-surface crossings, one corresponding to a pole in the Green's function and one corresponding to a zero value. Within the context of the YRZ ansatz, the latter zeros occur wherever

$$\xi_0(\underline{k}) = -2t(\cos k_x + \cos k_y) = 0 \qquad (3)$$

Here t is the nearest neighbor hopping term and x is the doping level. Examination of equation 3 shows that it is satisfied along the magnetic zone boundary running from (π,0) to (0,π). As can be seen in fig. 2(b), the two Fermi surface crossings associated with the dispersing bands map out the boundary of a hole pocket with an area that approximately scales with the doping level.

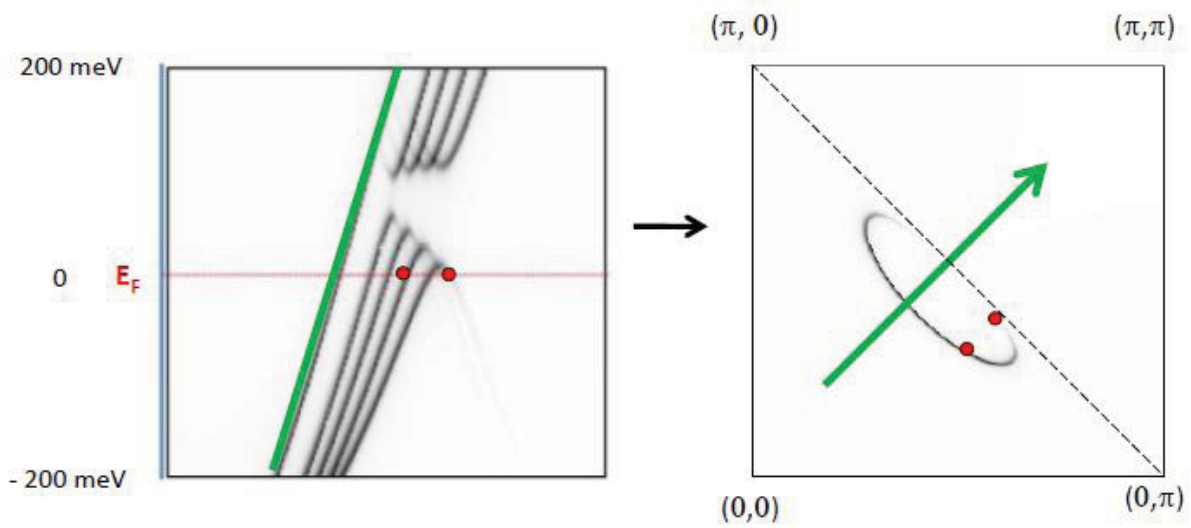


Fig 2. Dispersions calculated within the framework of the YRZ model on moving away from the nodal direction as defined by the green dispersing band. The red points indicate the Fermi surface crossings. The right panel shows the pocket defined by the crossings. The diagonal dashed line in the right panel defines the Luttinger surface which in this case coincides with the anti-ferromagnetic zone boundary.

That such a pocket can exist with one side's FS crossings defined by the poles of the Green's function and the backside defined by its zeros, is, as noted above, consistent with the observation that these materials are close to the Mott insulating state. Fig. 3 shows more finely detailed data recorded from the end of the arc region of a $T_c$ = 65K sample.[22] The figure shows both the raw data and the same data after analysis as described earlier. Fig. 3(o) holds the key to the analysis. It is assumed that the dispersion of a given band will follow that implied by fig. 2. Then, experimentally, one makes two key observations, point A, the initial FS crossing and, point B, the maximum travel of the band before loss of intensity. Then, based again on fig. 2, we make the assumption that a third point, C, defines a second Fermi surface crossing or "Ghost" crossing. The k-space symmetry, or bending back, of the band reflected across X as shown in the figure is consistent with the pair-like structure of the pseudogap assumed by the YRZ model.

Fig. 4(a) shows the results of such an analysis for three samples of different doping levels as indicated. Fig 4(b) shows the results of calculations using the YRZ ansatz for comparison. The agreement between experiment and calculation for the FS is quite remarkable. Again we note that other theoretical approaches that recognize the strongly correlated nature of the cuprates will also produce pockets. The inset to figure 4(a) shows the area of the pockets as a function of doping, versus the nominal doping level deduced from the Presland formula relating doping to $T_c$.[23] It would appear that at higher doping levels there is some deviation from the 1:1 correspondence observed at lower levels. It is unclear whether this represents a breakdown of the Luttinger sum rule concept [24] or the formation of electron pockets in the anti-nodal region. The latter would not be observable in the experiment because of the presence of the pseudogap. However, we also note that the presence of electron pockets at doping levels much below optimum is inconsistent with the results of the YRZ ansatz.[10] We note that the pockets reported in the present study are in relatively good agreement with the area bounded by the locus of superconducting

Bogoliubov band minima $k_B(E)$ extracted from spectroscopic imaging scanning tunneling spectroscopy (SISTS) studies of a sample with a similar doping level.[25]

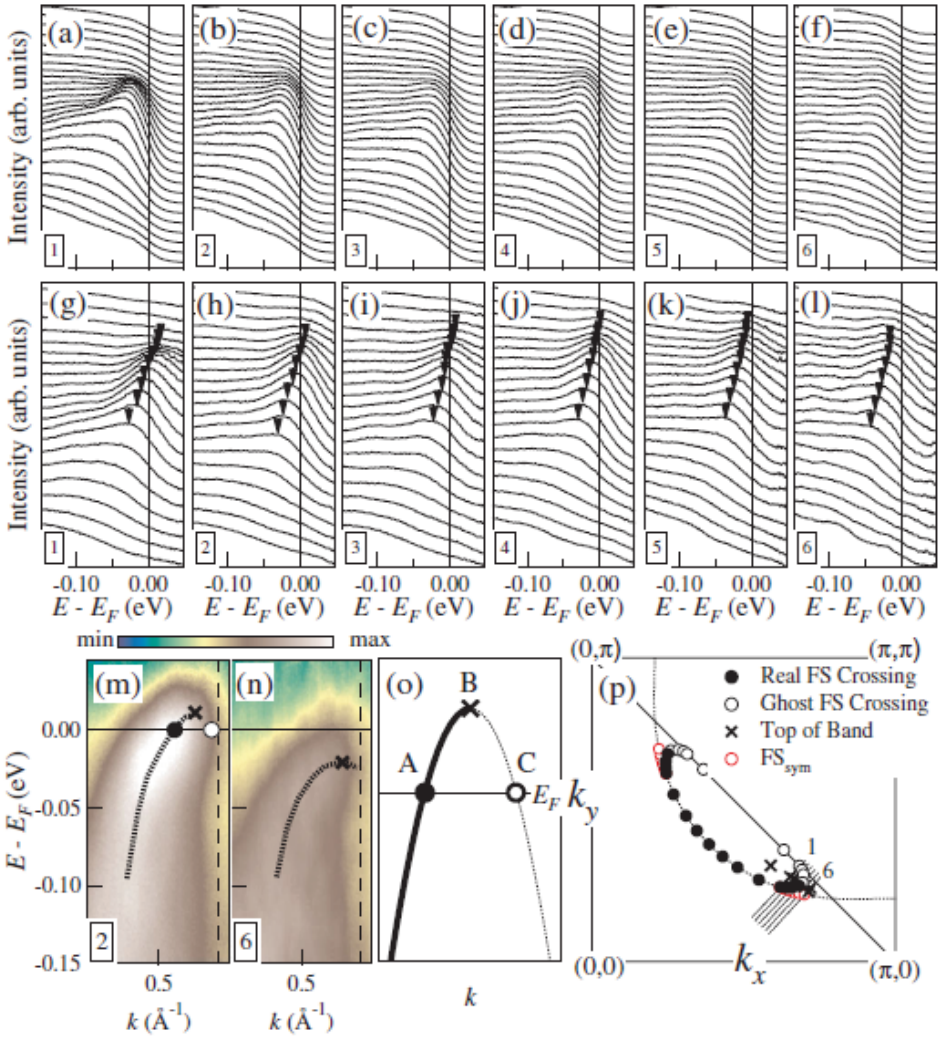


Fig 3. The upper panels (a-f) show spectra as recoded in the vicinity of the end of the arc. The center panels (g-i) show the same spectra following analysis as described in the text. Panel (m) shows a spectrum on the arc and (n) a spectrum off the arc. Panel (o) shows the basis of the analysis that results in the pocket shown in panel (p). Reproduced from Ref. [20].

## 4. Discussion

The calculated Fermi pockets in fig. 4(b) reflect different values of the pseudogap parameter $\Delta_R$ where

$$\Delta_R = \Delta_0(cosk_x - cosk_y) \qquad (4)$$

represents a pseudogap having the same underlying symmetry as the superconducting gap for these materials. In fig. 5 we plot those values as a function of doping. The plot suggests a critical point in the vicinity of x=0.2 doping at which point the Fermi surface switches from being doping-dependent nodal

Fermi pockets to a large Fermi surface centered on the (π, π) point. Such a transition should be evident in, for instance, transport measurements and indeed it is, as found in the studies of Tallon et al.[26]

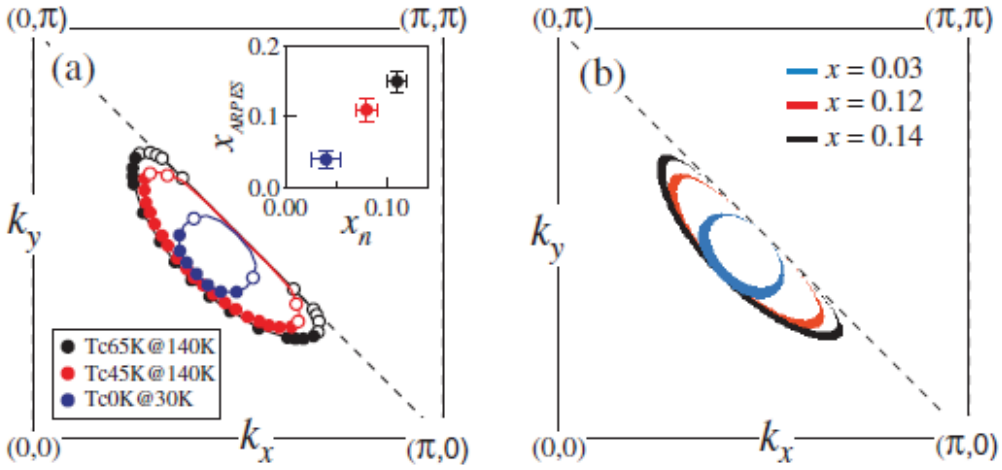


Fig. 4. (a) Experimentally pockets determined in the manner outlined in figure 3 for a series of doping levels. (b) Nodal Fermi pockets calculated within the framework of the YRZ ansatz for the same doping levels. Reproduced from ref [20].

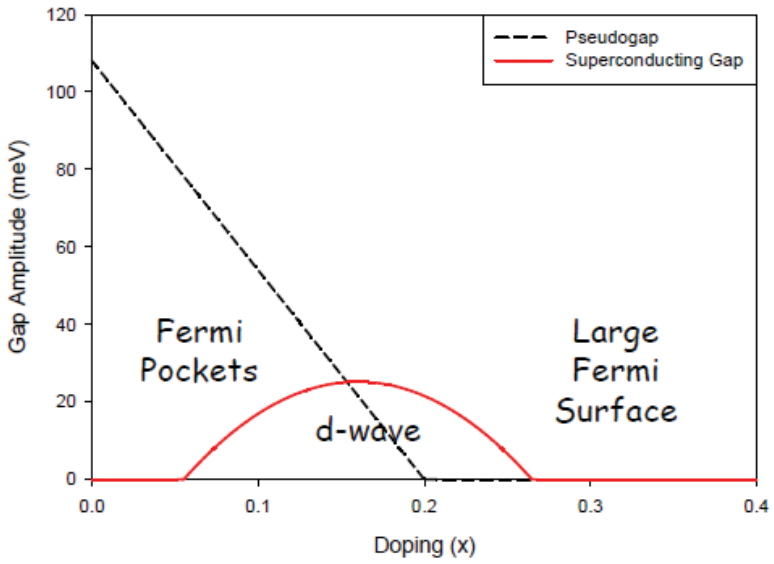


Fig. 5 Plot of the pseudogap parameter $\Delta_0$ as a function of doping x. The plot would indicate the presence of a possible critical point in the vicinity of 0.2 as suggested in ref [10].

The studies and analysis reported in this paper provide strong support for the possibility that the disconnected Fermi arcs observed in the Bi2212 system represent one side of nodal hole pockets with area scaling in proportion to the doping level. As noted earlier, the development of such pockets is entirely consistent with the fact that the cuprates emerge from the Mott-insulating state.

Although not reproduced here, we have shown elsewhere that in the anti-nodal region the spectral response in the normal state reveals a gap symmetric with respect to the chemical potential. A symmetric gap is an indication of some form of pairing. What exactly is the nature of the pairing? While some authors have suggested the possibility of pre-formed Cooper pairs,[8] others have suggested some form of singlet pairing.[21] Here we note again that the symmetric gap is observed only along the copper-oxygen bond directions. The gap is not symmetric away from these directions, implying that the pairing is not showing the full d-wave symmetry observed for the superconducting state as it exists below $T_c$. This would suggest that the pairing observed near the zone boundary is in fact real-space singlet pairing reflecting the underlying spin lattice of the $CuO_2$ planes.

**Acknowledgements**
The authors would like to thank Seamus Davis, Mike Norman, Philip Phillips, Maurice Rice, John Tranquada, Alexei Tsvelik, Subir Sachdev, Tonica Valla, and Ali Yazdani for useful discussions. The work at Brookhaven is supported in part by the U.S. DOE under Contract No. DE-AC02-98CH10886 and in part by the Center for Emergent Superconductivity (CES), an Energy Frontier Research Center funded by the U.S. DOE, Office of Basic Energy Sciences. The work at Argonne is partially supported by the U.S. DOE under Contract No. DE-AC02-06CH11357 and partially by the same CES. T. E. Kidd acknowledges support from the Iowa Office of Energy Independence Grant No. 09-IPF-11.

**References:**

[1] J.G. Bednorz and K.A. Muller, Zeitschrift fur Physik B – Condensed Matter, 64, 189 (1986)
[2] Y. Kamihara, T. Watanabe, M. Hirano, H. Hosono, JACS 130, 3296 (2008)
[3] T. Timusk and B. Statt, Rep. Prog. Phys. 62, 61 (1999)
[4] M. R. Norman et al., Nature (London) 392, 157 (1998).
[5] V. J. Emery and S. A. Kivelson, Nature (London) 374, 434 (1995).
[6] A.V. Chubukov and D. K. Morr, Phys. Rep. 288, 355 (1997).
[7] S. Chakravarty, C. Nayak, and S. Tewari, Phys. Rev. B 68, 100504 (2003).
[8] M.R. Norman,, A. Kanigel, M. Randeria, U. Chatterjee, and J.C. Campuzano, Rev. B 76, 174501 (2007)
[9] T.D. Stanescu, G. Kotliar, Phys Rev B, 74, 125110, (2006).
[10] K.Y. Yang, T. M. Rice, and F. C. Zhang, Phys. Rev. B 73, 174501 (2006).
[11] K.Y. Yang et al., Europhys. Lett. 86, 37 002 (2009)
[12] S. Sachdev, M. Metlitski, M. Punk, J. Phys.-Condensed Matter, 24, 294205 (2012)
[13] Seungmin Hong and Philip Phillips, arXiv:1110.0440 (2012)
[14] S Hufner, M A Hossain, A Damascelli and G A Sawatzky, Rep. Prog. Phys. **71** 062501 (2008)
[15] Doiron-Leyraud et al., Nature (London) 447, 565 (2007)
[16] Wu, T. et al. Nature 477, 191194 (2011).
[17] Kisker, E. et al., Phys. Rev. Lett. 58, 1784–1787 (1987).
[18] Matsui, H. et al., Phys. Rev. Lett. 90, 217002 (2003).
[19] J. D. Rameau, H.-B. Yang, and P. D. Johnson, J. Electron Spectrosc. Relat. Phenom. 181, 35 (2010).
[20] H.-B. Yang et al., Nature (London) 456, 77 (2008).
[21] P.W. Anderson, Science, 235, 1196 (1987)
[22] H.-B. Yang et al., Phys. Rev. Lett., 107, 047003 (2011)
[23] M.R. Presland, J.L. Tallon, R.G. Buckley, R.S. Liu and N.E. Flower, Physica C 176, 95 (1991)
[24] K.B. Dave, P.W. Phillips, C.L. Kane, arXiv:1207.4201 (2012)
[25] Y. Kohsaka et al., Nature (London) 454, 1072 (2008).
[26] J. L. Tallon, J. W. Loram, J. R. Cooper, C. Panagopoulos, and C. Bernhard, Phys Rev B, 68, 180501 (2003).