Electronic structure of Pr$_{2-x}$Ce$_x$CuO$_4$ studied via ARPES and LDA+DMFT+$\Sigma_k$

I. A. Nekrasov, N. S. Pavlov, E. Z. Kuchinskii, and M. V. Sadovskii
Institute for Electrophysics, Russian Academy of Sciences, Ekaterinburg 620016, Russia

Z. V. Pchelkina
Institute for Metal Physics, Russian Academy of Sciences, Ekaterinburg 620219, Russia

V. B. Zabolotnyy, J. Geck, B. Büchner, and S. V. Borisenko
Institute for Solid State Research, IFW-Dresden, P.O.Box 270116, D-01171 Dresden, Germany

D. S. Inosov
Max-Planck-Institute for Solid State Research, Heisenbergstrasse 1, D-70569 Stuttgart, Germany and
Institute for Solid State Research, IFW-Dresden, P.O.Box 270116, D-01171 Dresden, Germany

A. A. Kordyuk
Institute for Solid State Research, IFW-Dresden, P.O.Box 270116, D-01171 Dresden, Germany and
Institute of Metal Physics of National Academy of Sciences of Ukraine, 03142 Kyiv, Ukraine

M. Lambacher and A. Erb
Walther-Meißner-Institut, Bayerische Akademie der Wissenschaften, Walther-Meißner Strasse 8, 85748 Garching, Germany

(Dated: October 28, 2009)

The electron-doped Pr$_{2-x}$Ce$_x$CuO$_4$ (PCCO) compound in the pseudogap regime ($x \approx 0.15$) was investigated using angle-resolved photoemission spectroscopy (ARPES) and the generalized dynamical mean-field theory (DMFT) with the $k$-dependent self-energy (LDA+DMFT+$\Sigma_k$). Model parameters (hopping integral values and local Coulomb interaction strength) for the effective one-band Hubbard model were calculated by the local density approximation (LDA) with numerical renormalization group method (NRG) employed as an “impurity solver” in DMFT computations. An “external” $k$-dependent self-energy $\Sigma_k$ was used to describe interaction of correlated conducting electrons with short-range antiferromagnetic (AFM) pseudogap fluctuations. Both experimental and theoretical spectral functions and Fermi surfaces (FS) were obtained and compared demonstrating good semiquantitative agreement. For both experiment and theory normal state spectra of nearly optimally doped PCCO show clear evidence for a pseudogap state with AFM-like nature. Namely, folding of quasiparticle bands as well as presence of the “hot spots” and “Fermi arcs” were observed.

PACS numbers: 74.72.-h; 74.20.-z; 74.25.Jb; 31.15.A-

I. INTRODUCTION

Many experimental and theoretical papers have been seeking to describe the nature of high-temperature superconductivity (HTSC) in cuprates. In contrast to the normal (Fermi-liquid) metal, HTSC compounds show many abnormal properties for temperatures above the superconducting transition; the normal state pseudogap being a notorious example. The origin of this anomalous state is usually attributed either to superconducting fluctuations (precursor Cooper pairing) or to some order parameter competing with superconductivity, e.g. AFM fluctuations, incommensurate or fluctuating charge density waves (CDW), stripes, etc.

Recently a generalized LDA+DMFT+$\Sigma_k$ computational scheme was proposed to describe the pseudogap state in strongly correlated systems, by accounting for nonlocal AFM (or CDW) fluctuations with short-range order. Its relation to other theoretical DMFT-like approaches to the pseudogap state was discussed e.g. in Ref. 4. Both model computations and those for real systems were done and this approach, for instance, allowed one to describe the experimentally observed partial Fermi surface (FS) “destruction”, which was theoretically studied for hole-doped HTSC prototype system Bi$_2$Sr$_2$CaCu$_2$O$_{8-\delta}$ (Bi2212) and electron-doped one Nd$_{1.85}$Ce$_{0.15}$CuO$_4$ (NCCO). Two-particle properties can also be described by this approach, e.g. calculated optical spectra in the pseudogap state compare well with experimental data for Bi2212 and NCCO.

In this paper we study the electron-doped Pr$_{2-x}$Ce$_x$CuO$_4$ (PCCO) in the pseudogap state ($x = 0.15$) using the generalized LDA+DMFT+$\Sigma_k$ computational scheme and ARPES measurements. We present here both experimental and theoretical quasiparticle spectral functions and Fermi surfaces. These are found to agree well with each other supporting competing order parameter fluctuations as origin of the pseudogap instead of superconducting scenario.
II. COMPUTATIONAL DETAILS

The crystal structure\textsuperscript{12} of Pr\textsubscript{2}CuO\textsubscript{4} has tetragonal symmetry and the space group is I4/\textit{mmm}. Lattice constants\textsuperscript{12} are \textit{a}=\textit{b}=3.962, \textit{c}=12.154 Å. There are two crystallographic types of oxygen atoms in Pr\textsubscript{2}CuO\textsubscript{4}: the first one belongs to CuO\textsubscript{2} layer and the second is located within the Pr atoms. The atomic positions in the elementary cell are as follows: Cu – 2\textit{a}(0,0,0), O\textsubscript{1} – 4\textit{c}(0,0.5,0), Pr – 4\textit{e}(0,0,0.35171), O\textsubscript{2} – 4\textit{d}(0,0.5,0.25).\textsuperscript{12}

In Fig. 1 the crystal structure of Pr\textsubscript{2}CuO\textsubscript{4} is shown. Middle size grey spheres correspond to the copper atoms, a small dark and black spheres are O\textsubscript{1} and O\textsubscript{2} atoms, respectively and a big grey spheres — praseodymium atoms.

Electronic structure of PCCO was investigated within generalized LDA+DMFT+Σ\textsubscript{\textit{k}} scheme.\textsuperscript{5,6,7} This scheme incorporates the density functional theory in local density approximation (LDA)\textsuperscript{13} and the dynamic mean-field theory (DMFT)\textsuperscript{8} with “external” momentum-dependent self-energy Σ\textsubscript{\textit{k}}.\textsuperscript{6}

As a first step the LDA band structure calculation was performed. Using crystal structure data, the electronic band structure was obtained with the linearized muffin-tin orbitals (LMTO) method\textsuperscript{14}. Further hopping integral values were calculated for effective Cu-3\textit{d}(\textit{x}^2−\textit{y}^2) Wannier function within the \textit{N}-th order LMTO (NMTO) framework.\textsuperscript{15} Corresponding hopping integral values are \textit{t} = −0.4385, \textit{t}' = 0.1562, \textit{t}'' = 0.0976. The value of Coulomb interaction on effective Cu-3\textit{d}(\textit{x}^2−\textit{y}^2) orbital \textit{U}=1.1 eV was obtained via constrained LDA computations\textsuperscript{16}. Secondly, the DMFT calculations\textsuperscript{8}, which take the hopping integrals and the Coulomb interaction as input parameters, were performed.

To account for the AFM spin fluctuations, a two-dimensional model of the pseudogap state is applied.\textsuperscript{17} Corresponding \textit{k}-dependent self-energy Σ\textsubscript{\textit{k}}\textsuperscript{1,17} describes nonlocal correlations induced by (quasi) static short-range collective Heisenberg-like AFM spin fluctuations. The quasi static approximation for AFM fluctuations necessarily limits our approach to high - enough temperatures (energies not so close to the Fermi level)\textsuperscript{17}, so that in fact we are unable to judge e.g. on the nature of low temperature (energy) damping in our model. Thus we avoid here possible discussion of whether the damping corresponds to marginal or regular Fermi liquid. Moreover as shown in Ref. 6 for the “hot-spot” corresponding self-energy has essentially non Fermi liquid behavior.

The Σ\textsubscript{\textit{k}} definition contains two important parameters: the pseudogap energy scale (amplitude) Δ, representing the energy scale of fluctuating SDW, and the spatial correlation length ξ. The latter is usually determined from experiment. The Δ value was calculated as described in Ref. 6 and found to be 0.275 eV. The value of correlation length was taken to be 50 lattice constants, in accordance with the typical value obtained in neutron scattering experiments on NCCO\textsuperscript{18}. To solve DMFT equations numerical renormalization group (NRG\textsuperscript{19,20}) was employed as an “impurity solver”. Corresponding temperature of DMFT(NRG) computations was 0.011 eV and electron concentration was \textit{n}=1.145.

III. EXPERIMENTAL DETAILS

Photoemission experiments were performed at UE112-PGM beamline at BESSY using SCIENTA SES100 analyzer. Typical energy and angular resolution for the excitation energy (\textit{h}\textit{ν} = 100 eV) used in this study were 20 meV and 0.2° respectively. Samples of Pr\textsubscript{1.85}Ce\textsubscript{0.15}CuO\textsubscript{4+δ} were grown using traveling solvent floating zone technique and annealed to achieve optimal \textit{T}_c of 25K with transition width of 1K, which resulted
from improved growth conditions. Similarly, the FWHM of X-ray rocking curves was less than 0.08°, signaling high quality of the samples.

For the photoemission measurements, the samples were mounted on a cryomanipulator and cleaved in situ in ultra high vacuum with a base pressure \(\lesssim 1 \times 10^{-10}\) mBar. During the whole experiment, including the temperature cycling, when the sample was heated to room temperature and then cooled back again, no observable aging effects were detected.

IV. RESULTS AND DISCUSSION

Generally speaking, finite temperature and interaction lead to notable life-time effects. Thus, instead of quasi-particle dispersions obtained in usual band structure calculations one has to deal with corresponding spectral function \(A(\omega, \mathbf{k})\):

\[
A(\omega, \mathbf{k}) = -\frac{1}{\pi} \text{Im} G(\omega, \mathbf{k}),
\]

where \(G(\omega, \mathbf{k})\) is the retarded Green’s function obtained via LDA+DMFT+\(\Sigma_\mathbf{k}\) scheme.

Color plots, whose intensity encodes the function values, became a traditional and convenient way of representing these multiple variable functions. Such a color plot of LDA+DMFT+\(\Sigma_\mathbf{k}\) quasiparticle spectral function (1) of copper 3d orbital is presented in Fig. 2. Width of the quasiparticle spectral function in the color plot is inversely proportional to the quasiparticle lifetime. The calculated quasiparticle band dispersion has minimum at \(\Gamma\) point (-1.52 eV) and maximum at \(M\) point (2 eV). It crosses the Fermi level along \(X-M\) as well as \(M-\Gamma\) directions. Because of AFM pseudogap fluctuations, there is a well detectable (quasi) folding of the quasiparticle band, reflected in the formation of the so called “shadow” band, which has its maxima at \(\Gamma\)-point and minima at \(M\)-point. However, because of the short-range nature of the antiferromagnetic order, this does not result in a complete folding, as it would be the case for a long-range AFM order. Namely, quasiparticle band and the “shadow” band are not exactly the same. No real band gap opens at \((\pi/2, \pi/2)\) point. Nevertheless, suppression of the spectral weight is clearly detectable in the vicinity of \(X\) \((\pi, 0)\) point, thus signaling opening of the pseudogap, which in this case can be viewed as a precursor of the real band gap. Splitting takes place between the quasiparticle band and the “shadow” band with the value of about \(2\Delta\).

In Fig. 3 an extended picture of PCCO Fermi surfaces is presented (panel (a) — LDA+DMFT+\(\Sigma_\mathbf{k}\) results, panel (b) — experimental ARPES data). Strictly speaking Fig. 3 is a color map in reciprocal space of the corresponding spectral function plotted at the Fermi level. FS is clearly visible as reminiscence of non-interacting band close to the first Brillouin zone border and around \((\pi/2, \pi/2)\) point (so called Fermi arc), where the quasiparticle band crosses the Fermi level. There is an interesting physical effect of partial “destruction” of the FS observed in the “hot spots”, points that are located at the intersection of the FS and its AFM umklapp replica. This FS “destruction” occurs because of the strong electron scattering on the antiferromagnetic (AFM) spin (pseudogap) fluctuations on the copper atoms. Also the “shadow” FS is visible as it should be for AFM folding. As no long-range order is present in the underdoped phase the “shadow” FS has weaker intensity with respect to FS. Another evidence of presence of electron pock-
ets can be seen in the experimental FS map shown in Fig. 3b. One pocket is centered at point with coordinates $(0,0.8\pi)$ and the other at $(0.8\pi,0)$. Again, in agreement with the theoretical prediction, the pocket “sides” which coincide with originally unreconstructed FS have higher intensity, while the newly appearing “replicas/shadows” have weaker intensity.

The PCCO FS is very similar to that of Nd$_{2-x}$Ce$_x$CuO$_4$ (NCCO), which belongs to the same family of superconductors. The NCCO was recently studied both theoretically and experimentally. Let us compare theoretical (upper line) and experimental (lower line) energy quasiparticle dispersion for most characteristic cuts introduced in Fig. 3 (see Fig. 4). Theoretical data were multiplied by the Fermi function at a temperature of $30K$ and convoluted with a Gaussian to simulate the effects of experimental resolution, with further artificial noise added.

The Cut 1 intersects quasiparticle and “shadow” Fermi surfaces close to the Brillouin zone border. One can find here a “fork”-like structure formed by the damped “shadow” band (-0.5-0 arb. u.) and better defined quasiparticle band (0.5-1 a.u.). This structure corresponds to preformation of FS cylinder around $(\pi,0)$ point. The Cut 2 goes exactly through the “hot spot”. Here we see a strong suppression of the quasiparticle band around the Fermi level as it is also shown in Fig. 2. The Cut 3 crosses the Fermi arc, where we can see a very well defined quasiparticle band. However weak intensity “shadow” band is also present. For the case of long range AFM order and complete folding of electronic structure, FS and its “shadow” should form a closed FS sheet around $(\pi/2,\pi/2)$ point, while in the current case the part of the pocket formed by the “shadow” band is not so well defined in momentum space. As can be seen there is a good correspondence between the calculated and experimental data in terms of the above described behavior, which is also similar to the results reported for Nd$_{2-x}$Ce$_x$CuO$_4$ (NCCO) in our earlier work.

**V. CONCLUSION**

In this work the LDA+DMFT+$\Sigma_k$ was performed for electron-doped Pr$_{2-x}$Ce$_x$CuO$_4$ compound in the pseudogap regime. The LDA+DMFT+$\Sigma_k$ calculation shows that Fermi-liquid behavior is still conserved far away from the “hot-spots”, while the destruction of the Fermi surface observed in the vicinity of “hot spots” is due to strong scattering of correlated electrons on short-range antiferromagnetic (pseudogap) fluctuations. Comparison between experimental ARPES and LDA+DMFT+$\Sigma_k$ data reveals a good semi-quantitative agreement. The experimental and theoretical results obtained once again support the AFM scenario of pseudogap formation not only in hole doped HTSC systems but also in electron doped ones.

We thank Thomas Pruschke for providing us the NRG code. This work is supported by RFBR grants 08-02-00021, 08-02-91200 and RAS programs “Quantum physics of condensed matter” and “Strongly correlated electrons in solids”. IN and ZP are supported by Grants of President of Russia MK-3227.2008.2(ZP) and MK-614.2009.2(IN), Russian Science Support Foundation (IN) and the Dynasty Foundation (ZP). The experimental measurements for this study were possible owing to the financial support of Forschergruppe FOR538 and by the DFG under Grant No. KN393/4.  

---

1 T. Timusk, B. Statt, Rep. Progr. Phys., **62**, 61 (1999); M. V. Sadovskii, Phys. Usp. **44**, 515 (2001); M. V. Sadovskii, in “Strings, branes, lattices, networks, pseudogaps and dust”, Scientific World, Moscow, 2007, p. 357 (in Russian), English version: cond-mat/0408489.

2 V. J. Emery, S. A. Kivelson, Nature **374**, 434 (2002).

3 A. A. Kordyuk, S. V. Borisenko, V. B. Zabolotnyy, R. Schuster, D. S. Inosov, D. V. Bychkovskii, A. I. Luslov, R. Follath, A. Varykhalov, L. Pattkey, and H. Berger, Phys. Rev. B **79**, 020504(R) (2009).

4 E. Z. Kuchinskii, I. A. Nekrasov, Z. I. Pchelkina, M. V. Sadovskii, JETP **104**, 792 (2007); I. A. Nekrasov, E. Z. Kuchinskii, Z. V. Pchelkina and M. V. Sadovskii, Physica C **460–462**, 997 (2007).

5 E. Z. Kuchinskii, I. A. Nekrasov, M. V. Sadovskii, JETP Lett. **82**, 198 (2005).

6 M. V. Sadovskii, I. A. Nekrasov, E. Z. Kuchinskii, Th. Prusiske, V. I. Anisimov, Phys. Rev. B **72**, 155105 (2005).

7 E. Z. Kuchinskii, I. A. Nekrasov, M. V. Sadovskii, Phys. Rev. B **75**, 115102 (2007).

8 A. Georges, G. Kotlier, W. Krauth and M. J. Rozenberg, Rev. Mod. Phys. **68**, 13 (1996).

9 E. E. Kokorina, E. Z. Kuchinskii, I. A. Nekrasov, Z. V. Pchelkina, M. V. Sadovskii, A. Sekiyama, S. Suga, M. Tsunekawa, JETP **107**, 818 (2008); I. A. Nekrasov et al., J. Phys. Chem. Solids **69**, 3269 (2008).

10 A. Damascelli, Z. Hussain, and Zhi-Xun Shen, Rev. Mod. Phys. **75**, 473 (2003).

11 A. A. Kordyuk and S. V. Borisenko, Low Temp. Phys. **32**, 298 (2006).

12 D. E. Cox et al. Phys. Rev. B **40**, 6998 (1989).

13 R. O. Jones and O. Gunnarsson, Rev. Mod. Phys. **61**, 689 (1989).

14 O. K. Andersen, Phys. Rev. B **12**, 3060 (1975); O. K. Andersen and O. Jepsen, Phys. Rev. Lett. **53**, 2571 (1984).

15 O. K. Andersen and T. Saha-Dasgupta, Phys. Rev. B **62**, R16219 (2000); O. K. Andersen et al. Science **285**, 86 (2001); O. K. Andersen, T. Saha-Dasgupta, S. Zhov, Bull. Mater. Sci. **26**, 19 (2003).

16 O. Gunnarsson, O. K. Andersen, O. Jepsen, and J. Zaanen, Phys. Rev. B **39**, 1708 (1989).

17 J. Schmalian, D. Pines, B. Stojkovic, Phys. Rev. B **60**, 667.
E. Z. Kuchinskii, M. V. Sadovskii, JETP 88, 347 (1999).

I. A. Zobkalo et al., Solid State Comm. 80, 921 (1991);
E. M. Motoyama, G. Yu, I. M. Vishik, O. P. Vajk, P. K. Mang, M. Greven, Nature 445, 186 (2007).

K. G. Wilson, Rev. Mod. Phys. 47, 773 (1975);
H. R. Krishna-murthy, J. W. Wilkins, and K. G. Wilson, Phys. Rev. B 21, 1003 (1980); ibid. 21, 1044 (1980).

R. Bulla, A. C. Hewson and Th. Pruschke,

J. Phys.: Condens. Matter 10, 8365 (1998);
M. Lambacher,
Ph.D. thesis, Technische Universität München, 2008.

N. P. Armitage, F. Ronning, D. H. Lu, C. Kim, A. Damascelli, K. M. Shen, D. L. Feng, H. Eisaki, Z.-X. Shen, P. K. Mang, N. Kaneko, M. Greven, Y. Onose, Y. Taguchi, Y. Tokura, Phys. Rev. Lett. 88, 257001 (2002).