Nonmonotonic $d_{x^2-y^2}$ superconducting gap in electron-doped Pr$_{0.89}$LaCe$_{0.11}$CuO$_4$: Evidence of coexisting antiferromagnetism and superconductivity?

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Recent experiments on Pr$_{0.89}$LaCe$_{0.11}$CuO$_4$ observe an anisotropic spin-correlation gap and a nonmonotonic superconducting (SC) gap, which we analyze within the framework of a $t-t'-t''-t'''-U$ model with a $d_{x^2-y^2}$ pairing interaction including a third harmonic contribution. By introducing a realistic broadening of the quasiparticle spectrum to reflect small-angle scattering, our computations explain the experimental observations, especially the presence of a maximum in the leading edge gap in the vicinity of the hot-spots. Our analysis suggests that the material behaves like a two-band superconductor with the $d$-wave third harmonic acting as the interband pairing gap, and that the anti-ferromagnetic (AFM) and SC orders co-exist in a uniform phase.

PACS numbers: 74.72.Dn 74.20.Rp 74.25.Dw 74.25.Jb

Recent Raman scattering$^1$ and ARPES experiments$^2$ on electron-doped Pr$_{0.89}$LaCe$_{0.11}$CuO$_4$ (PLCCO) find a $d_{x^2-y^2}$ superconducting (DSC) gap, which varies non-monotonically along the Fermi surface (FS). The gap increases as one moves away from the nodal direction towards the hot-spot region in the Brillouin zone (BZ) where it attains a maximum value. It then decreases as one approaches the zone boundary along the anti-nodal direction. The involvement of the hot-spots suggests that the bosonic pairing possesses a magnetic origin,$^3$,$^4$ If we assume that the system continues to remain uniform with doping, and recall that the residual gap in a half-filled AFM insulator yields two FS pockets on electron doping, these observations present a striking conundrum: The leading-edge gap is the largest in the momentum region of the hot-spots where there are no segments of the FS and quasiparticle states lie well below the Fermi energy.

In this article, we discuss a relatively simple route for resolving this dilemma and understanding the behavior of the superconducting state of PLCCO. In particular, we consider a uniformly doped system with co-existing AFM and DSC orders, where the quasiparticle spectrum is broadened realistically to reflect the effects of small angle scattering. We thus obtain in the normal state a finite spectral weight at the Fermi energy ($E_F$) throughout the BZ, and when a third harmonic term is included in the pairing interaction, the computed leading edge gap along the FS reproduces the corresponding experimental variations remarkably well. The third harmonic contribution possesses the proper symmetry to couple the DSC order parameter on the two FS pockets to induce a maximum in the leading edge gap around the hot-spots even though the hot-spots lie outside the momentum region of the two FS pockets. Notably, non-monotonic gap variations have been obtained in some earlier calculations,$^3$,$^4$ but we are not aware of a previous study involving a realistic model of electron doping with two FS pockets.

Our study gives insight into the issue of well known asymmetry between the properties of cuprates with electron vs hole doping, which has been a subject of considerable recent debate (see e.g., Ref. 5). There is growing evidence that the normal state of the electron doped cuprates can be described as an AFM metal up to a quantum critical point near optimal doping, and that non-monotonic gap variations, it supports the scenario that the system remains uniform with electron doping not only in the normal but also in the SC state without the intervention of other orders.

We model PLCCO as a uniformly doped spin density wave (SDW) antiferromagnet with $d$-wave superconductivity. At the mean field level, there is long-range AFM order, but when fluctuations are included, the Néel temperature $T_N$ becomes zero (in the absence of interlayer coupling), and the mean-field gap turns into a pseudogap $\Delta^*$ with crossover temperature $T^*$. In the SC doping range the FS consists of a necklace of two types of pockets: an electron-like pocket near the antinodal point ($\pi, 0$), and a hole-like pocket near the ($\pi/2, \pi/2$) nodal point. A pseudogap near the hot-spot separates the two pockets. Our one-band Hubbard model Hamiltonian is

$$H = \sum_{\mathbf{k}, \sigma} \xi_{\mathbf{k}} c^\dagger_{\mathbf{k}, \sigma} c_{\mathbf{k}, \sigma} + U_0 \sum_{\mathbf{k}, \mathbf{k}' } c^\dagger_{\mathbf{k}, \sigma} c^\dagger_{\mathbf{k}+\mathbf{Q}, \sigma} c_{\mathbf{k}', \sigma} c_{\mathbf{k}+\mathbf{Q}, \sigma} + \sum_{\mathbf{k}, \mathbf{k}' } V(\mathbf{k}, \mathbf{k}') c^\dagger_{\mathbf{k}, \sigma} c^\dagger_{\mathbf{k}'+\mathbf{Q}, \sigma} c_{\mathbf{k}'', \sigma} c_{\mathbf{k}'+\mathbf{Q}, \sigma} \tag{1}$$

where $c^\dagger_{\mathbf{k}, \sigma} (c_{\mathbf{k}, \sigma})$ is the electronic creation (destruction) operator with momentum $\mathbf{k}$ and spin $\sigma$. The independent particle dispersion with respect to the chemical potential $E_F$ is given by

$$\xi_{\mathbf{k}} = -2t[c_x(a) + c_y(a)] - 4t'c_x(a)c_y(a) + 2t''[c_x(2a) + c_y(2a)] - 4t'''[c_x(2a)c_y(a) + c_y(2a)c_x(a)] - 4t'''c_x(2a)c_y(2a) - E_F,$$
with \( c_i(\alpha a) = \cos(\alpha \vec{k}_i a) \) and \( a \) is the lattice constant.

The effective on-site Hubbard repulsion \( U_Q \) is taken to be doping-dependent.\[8\]. The Mott gap \( U_Q S \) arises from a finite expectation value of the staggered magnetization \( S \) at the commensurate ordering wave vector \( \vec{Q} = (\pi, \pi) \). The \( d \)-wave superconductivity including first and third harmonics is defined by \[8, 10\].

\[
\Delta_k = \Delta_{ik} + \Delta_{3ik} = \sum_{i} V(\vec{k}, \vec{k'}) \langle c_{i\vec{k}'}^\dagger c_{i\vec{k}'} \rangle \\
= \sum_i \Delta_i g_{ik} = \sum_i g_{ik} V_i \sum_{\vec{k}'} g_{i\vec{k}'} \langle c_{i\vec{k}'}^\dagger c_{i\vec{k}'} \rangle, \quad (3)
\]

with \( i = 1, 3 \) and \( g_{ik} = c_x(i\alpha) - c_y(i\alpha) \).

Hamiltonian of Eq. 1 is diagonalized straightforwardly \[10\]. The resulting quasiparticle dispersion consists of upper and lower Hubbard bands (UHB and LHB), each gapped via the DSC pairing as:

\[
E_k^{U+L+} = -E_k^{U-}(L-) = \sqrt{(\xi_k^+ + E_{0k})^2 + \Delta_k^2}, \quad (4)
\]

where \( E_{0k}^2 = (\xi_k^+)^2 + (U_Q S)^2 \) and \( \xi_k^\pm = (\xi_k^+ \pm \xi_{k+\vec{Q}})/2 \). The SDW magnetization \( S \) per site is evaluated self-consistently at each temperature via the equations,

\[
S = \sum_k \alpha_k \beta_k \left[ [(v_k^+)^2 - (v_k^-)^2] + [(v_k^+)^2 - (u_k^-)^2] f(E_k^{U+}) - [(v_k^-)^2 - (u_k^+)^2] f(E_k^{L+}) \right], \quad (5)
\]

where \( f(E) = 1/(\exp(\beta E) + 1) \) is the fermi function with \( \beta = 1/k_B T \). The expressions for \( \alpha_k, \beta_k, u_k^\pm \) and \( v_k^\pm \) are same as Eq (9) and Eq (14) respectively of Ref. \[10\].

The self-consistent SC-gap equations are

\[
\Delta_i = -V_i \sum_k \Delta_k g_{ik} \left[ \frac{\tanh(\beta E_k^{U+}/2)}{2E_k^{U+}} + \frac{\tanh(\beta E_k^{L+}/2)}{2E_k^{L+}} \right] \quad (6)
\]

The one-particle Green function is

\[
G(\vec{k}, \omega) = \frac{[\omega Z + \xi_k^+ + E_{0k}] f(E_k^{U+})}{(\omega^2 - \Delta_k^2) Z^2 - (\xi_k^+ + E_{0k})^2} + \frac{[\omega Z + \xi_k^- - E_{0k}] f(E_k^{L+})}{(\omega^2 - \Delta_k^2) Z^2 - (\xi_k^- - E_{0k})^2}, \quad (7)
\]

where we have included broadening due to small-angle elastic scattering \[11, 12\] with the associated renormalization factor, \( Z = 1 + i \Gamma_0(\omega) \text{sgn}(\omega)/\sqrt{\omega^2 - \Delta_k^2} \).

\( \Gamma_0(\omega) \) is the normal state scattering rate, discussed below. The corresponding spectral intensity is \( A(\vec{k}, \omega) = -\text{Im}[G(\vec{k}, \omega)]/\pi \).

We fitted the dispersion given by Eq. \[11\] to the ARPES results on PLCCO\[2\] at 30 K to obtain the relevant TB parameters as follows: \( t = 0.12 \text{ eV}, t' = -0.06 \text{ eV}, t'' = 0.034 \text{ eV}, t''' = 0.007 \text{ eV}, t'' = 0.02 \text{ eV}, \) with \( E_F = -0.082 \text{ eV} \) at 11% doping.\[13\] The effective Hubbard parameter is found to be \( U_Q = 4.15t \), which yields a self-consistent value of the magnetization \( S \) of 0.281 from Eq. \[15\].

Fig. 1 clarifies the nature of the quasiparticle spectrum and the FS. As Eq. 4 shows, the non-interacting band is split via the Hubbard interaction into the upper and lower Hubbard bands. As seen from Fig. 1(a), each of these bands is further split by the SC interaction to yield pairs of bands \( E_k^{U\pm} \) (magenta and red) and \( E_k^{L\pm} \) (cyan and green) which lie symmetrically above and below the \( E_F \) line. The spectral weights for various bands (proportional to the vertical width of shading) are seen to vary greatly with \( \vec{k} \). In effect, the spectral weight of the non-interacting band is redistributed between the UHB and the LHB through the AFM order, and it is modified further via the SC order at very small energy scales of a few meV’s. Above the \( E_F \), we see from Fig. 1(a) that most of the spectral intensity resides in the UHB branch \( E_k^{U+} \) (magenta) along the \((\pi, 0) \rightarrow (\pi, \pi) \rightarrow (\pi/2, \pi/2)\) line. At other momenta, the spectral weight generally lies below \( E_F \) and is carried by the \( E_k^{L-} \) band (green).

Fig. 1(b) shows a map of the normal state (30K) FS. This FS is consistent with the experimental results on electron-doped \( \text{Nd}_{0.87}\text{Ce}_{0.13}\text{CuO}_4 \) (NCCO)\[14, 15\], and it can be understood with reference to the band structure of Fig. 1(a). The \( E_k^{U-} \) band (red) gives rise to the \((\pi, 0)-\)centered electron pockets, while the \((\pi/2, \pi/2)-\)centered hole pocket arises from the \( E_k^{L-} \) band (green).
in PLCCO. The computed energy bands in the normal state along the three cuts used in the fitting procedure are compared directly with the corresponding experimental dispersions in panels (a)-(c). The overall agreement is seen to be quite good, although a discrepancy is evident in (a) for the anti-nodal cut in that the computed size of the electron pocket around \((\pi,0)\) is smaller than that indicated by the experimental data.\(^{[16]}\) Notably, due to the presence of the AFM gap, quasiparticle states along the hot spot in (b) lie well below the \(E_F\), leading to a suppression of the spectral weight up to binding energies of about 60 meV. In contrast, along the nodal direction in (c), the quasiparticle band crosses \(E_F\), and as already pointed out, gives rise to the \((\pi/2,\pi/2)\) centered hole pockets.

The spectral intensity computed from the imaginary part of the one-particle Green’s function of Eq. 7 is shown for a series of momenta in the bottom row of Fig. 2. Here the cuts in the top and bottom set of panels correspond to each other. For example, in (e) the five spectra shown refer to the five k-points given by the blue dots in (b) with the lowest spectrum in (e) corresponding to the leftmost dot in (b). The computations assume a \(k\)-independent broadening function of form: \(\Gamma_0(\omega) = C_0[1 + (\omega/\omega_0)^p]\), which is similar to a form which has been proposed for hole-doped cuprates.\(^{[17]}\) Parameter values of \(C_0 = 0.1\) eV, \(\omega_0 = 1.59\) eV, with \(p = 3/2\) reasonably reproduce the experimentally observed broadenings.\(^{[18]}\) We emphasize that, for our purposes, the detailed spectral shape is not so important. The key is the presence of quasiparticle broadening, which allows the development of a finite spectral weight at the \(E_F\) and the formation of the leading edge superconducting gap at all momenta, even though the underlying quasiparticle states lie well below the \(E_F\) at most momenta.

Fig. 3 compares spectral intensities in the vicinity of the \(E_F\) in the normal and the SC state at four different momenta A-D (see insets). We see that in (a)-(c) the midpoint of the leading-edge of the 8 K spectrum (solid lines) is shifted by \(\Delta_{\text{shift}}\) towards higher binding energies with respect to the spectrum at 30 K (dashed lines), while along the nodal direction in (d), this shift vanishes due to the node in the d-wave gap. Experiments\(^2\) find a leading edge gap \(\Delta_{\text{shift}}\) of 2.0 meV along the antinodal direction at the momentum point A \((\pi,0.8)\), which increases to 2.5 meV at the hotspot B\((2.34,1.0)\); \(\Delta_{\text{shift}}\) then decreases to 1.6 meV at C\((1.85,1.23)\) and becomes zero along the nodal direction at the point D\((1.52,1.52)\). The computations of Fig. 3 reproduce this behavior. For this purpose, the superconducting parameters are found to be: \(V_1 = -76\) meV and \(V_3 = 54\) meV, leading to self-consistent values of the gap parameters at 8 K of \(\Delta_1 = 1.25\) meV and \(\Delta_3 = -0.53\) meV from Eq. 8 and 8. These parameter values yield \(T_c = 15\) K, which is in reasonable accord with the experimental value of 26 K. Note that

\[\text{Intensity (arb. units)}\]

\[E_F - E [\text{eV}]\]

\[E_F - E [\text{eV}]\]

\[E_F - E [\text{eV}]\]

\[E_F - E [\text{meV}]\]

\[E_F - E [\text{meV}]\]

\[E_F - E [\text{meV}]\]

\[E_F - E [\text{meV}]\]
a leading edge gap is clearly seen near the hotspot in (b). This gap is a direct consequence of the broadening of the spectrum due to small angle scattering and the associated residual spectral weight at the \(E_F\), even though this spectral weight is quite small due to the presence of a sizable pseudogap in the spectrum.

Fig. 4(a) shows variations in the leading edge shift \(\Delta_{\text{shift}}\) and its non-monotonic and anisotropic nature more clearly. Here the minimum computed value of \(\Delta_{\text{shift}}\) (blue line) is plotted along various directions given by the angle \(\phi\), where \(\phi = 0\) refers to the antinodal direction and \(\phi = 45^\circ\) to the nodal direction. The actual momentum points at which the computed values of \(\Delta_{\text{shift}}\) are plotted, shown in the inset, lie close to the non-interacting FS. The corresponding experimental values (red dots) are in good accord with the theoretical results. In particular, the gap reaches a maximum value of about 2.5 meV around \(\phi = 18^\circ\) near the hot-spot in both theory and experiment. The monotonic d-wave gap obtained by setting \(\Delta_1 = 0\) in the calculations is also shown for reference (dashed line) to highlight the non-monotonic gap variations in the present case.

Our explanation of nonmonotonic gap variations should be distinguished sharply from the notion that one simply observes two different gaps in the experiments, i.e. an SC gap along the pockets and an AFM gap near the hotspots. This alternative picture suffers from the problem that the AFM gap in the normal state of the cuprates is far too large, and moreover, it is not clear how the leading edge gap comes about since there are no states at the \(E_F\) away from the region of the two small FS pockets. Interestingly, the two band model study of Ref. 10 would require an anomalously small value of \(t\) to fit the experimental gap values in PLCCO.

Third harmonic gaps have been found not only in electron-doped PLCCO [2] and NCCO [1], but also in hole-doped (underdoped) \(\text{Bi}_2\text{Sr}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+1}\) (n = 1–3) (BISCO) [20, 21]. Interestingly, the third harmonic contribution in electron-doped cuprates possesses an opposite sign and its size is substantially larger than in the hole-doped case, leading to the nonmonotonic gap variations discussed above. Our study provides insight into this behavior. Separate electron and hole pockets in PLCCO and NCCO are suggestive of two-band superconductivity as in the case of \(\text{MgB}_2\) where interband pairing greatly enhances the \(T_c\). However, the form of our Hamiltonian in Eq. 11 does not involve a separate interband pairing gap. Fig. 1(b) shows that \(\Delta_{3k}\) possesses the correct symmetry and that its size is maximal in the interval between the two pockets just like the leading edge gap \(\Delta_{\text{shift}}\). With this in mind, we propose that the third-harmonic gap \(\Delta_{3k}\) can be looked upon as playing the role of interband pairing gap in PLCCO and NCCO. In fact, the ‘hot spot’ between the two pockets is associated with strong AFM fluctuations in a quasi-two-dimensional system [2, 11], which in turn are responsible for coupling the UHB and the LHB, and thus coupling the phase of the order parameter on the two FS segments.

The fact that the nonmonotonic gap in our model is intimately associated with the AFM pseudogap indicates that superconductivity arises in an AFM background. These results support the picture of electron doped cuprates being uniformly doped AFM metals, even in the superconducting state, rather than being phase separated AFM and SC domains. Note however that when fluctuations are added, the present AFM gap becomes a short-range order pseudogap, with a possible Neel ordering transition at lower temperatures due to interlayer coupling. Open questions remain as to whether superconductivity coexists with this residual 3D Neel order, or whether the onset of superconductivity destroys either the 3D order, or even the 2D \((T=0\text{K})\) long-range AFM order.

We thank Hong Ding for sharing some of his unpublished results with us. This work is supported by the U.S.D.O.E contract DE-AC03-76SF00098 and benefited from the allocation of supercomputer time at NERSC and Northeastern University’s Advanced Scientific Computation Center (ASCC).

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