Pseudogap Phase and the Quantum-Critical Point in Copper-Oxide Metals

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A systematic solution of a model for copper oxides reveals a line of transitions $T = T_p(x)$ for x, the doping away from half filling less than a critical value, to a phase with broken time reversal and rotational symmetry. The single-particle spectrum in this phase is calculated to have a gap $\sim[\cos(k_x a/\pi) - \cos(k_y a/\pi)]^2$. The properties in this phase are compared to the properties of the so-called "pseudogap phase" of the copper oxides.

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A schematic phase diagram of the copper-oxide (CuO) metals is shown in Fig. 1. The superconducting region is surrounded by three distinct regions: a region marked (III) with properties characteristic of a Fermi liquid, a region marked (I) in which a Fermi surface is discerned but in which the quasiparticle concept is inapplicable, and a region marked (II), the so-called pseudogap region, in which the concept of a Fermi surface itself is lost.

The topology of Fig. 1 around the superconducting region is that expected around a quantum-critical point in itinerant fermions. The marginal Fermi-liquid (MFL) phenomenology [1] with which many of the unusual properties in region (I) are understood assumes a scale-invariant low energy fluctuation spectrum characteristic of a quantum-critical point at $x = x_c$, the composition near the highest T_c. A region of Fermi liquid is then expected for $x > x_c$ at low temperatures, as in region (III), and a region with a broken symmetry for $x < x_c$ at low temperatures. As in heavy-fermion compounds [2], a region of superconductivity is found at low temperatures peaked in the region around the quantum-critical point. From this point of view, the crucial problem in CuO metals is the symmetry of the phase in the region (II) below $T_p(x)$.

In a systematic theory starting with a general model for CuO [3], the region (II) in Fig. 1 is derived to be a phase in which a fourfold pattern of current flows in the ground state in each unit cell as in Fig. 6 of Ref. [3]. This phase will be referred to as the circulating current (CC) phase. The properties of this phase were not studied in Ref. [3], which was mainly concerned with the fluctuations leading to the MFL properties in region (I) and the superconductivity induced by them.

In the past few years, the properties in region (II) have become much clearer, thanks especially to angle-resolved photoemission (ARPES) [4] and thermodynamic measurements [5,6]. An anisotropic pseudogap in the single-particle spectra begins to develop at $T < T_p(x)$ which is similar to the temperature below which other properties begin to change from the MFL behavior of region (I) [7]. I calculate in this paper that the CC phase has a single-particle spectra with a gap consistent with the observed symmetry, and of the right order of magnitude, and from which the other properties in region (II) follow. I suggest further experiments to confirm this identification. (Numerous ideas already suggested for the pseudogap phase are discussed elsewhere [7].)

The model.—The CC phase is a mean-field solution of a general Hamiltonian in the basis of three orbitals per unit cell, d, p_x, p_y [8]:

$$H = K + H_{\rm int}^{(1)} + H_{\rm int}^{(2)}, \tag{1}$$

$$K = \sum_{\mathbf{k},\sigma} \epsilon_d n_{d\mathbf{k}\sigma} + 2t_{pd} d^{+}_{\mathbf{k},\sigma}[s_x(k) p_{xk\sigma} + s_y(k) p_{yk\sigma}] - 4t_{pp} s_x(k) s_y(k) p^{+}_{xk\sigma} p_{yk\sigma} + \text{H.c.} \tag{2}$$

Here a particular choice of the relative phases of the x and y orbitals in the unit cell has been made, $s_{x,y}(k) = \sin(k_x a/2, k_y a/2)$ and, for later, $c_{x,y}(k) = \cos(k_x a/2, k_y a/2)$ and $s^2_{xy}(k) = \sin^2(k_x a/2) + \sin^2(k_y a/2)$. I consider the local interaction on the Cu and the O orbitals,

$$H_{\rm int}^{(1)} = \sum_{i,\sigma} U_d n_{di\sigma} n_{di-\sigma} + U_p(n_{pxi\sigma} n_{pxi-\sigma} + n_{pyi\sigma} n_{pyi-\sigma}), \tag{3}$$

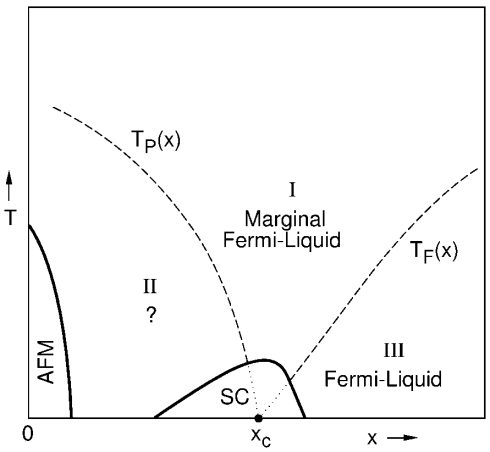

FIG. 1. Generic phase diagram of the cuprates for hole doping. Not shown is a low temperature "insulating phase" in region II due to disorder.

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and the nearest neighbor interaction between the Cu and the O orbitals,

$$H_{\rm int}^{(2)} = 2V \sum_{\mathbf{k}\mathbf{k}'\mathbf{q},\sigma\sigma'} c_x(q) d^+_{\mathbf{k}+\mathbf{q}\sigma} d_{\mathbf{k}\sigma} p^+_{x\mathbf{k}'-\mathbf{q}\sigma'} p_{x\mathbf{k}'\sigma'} + x \to y\,. \tag{4}$$

More general interactions do not change the essential results derived here. Throughout this paper (renormalized) energy difference ϵ_d between the Cu and the O orbitals is taken to be zero. It is important that, in CuO, $\epsilon_d \lesssim O(t_{pd})$. Taking it as zero simplifies the calculations presented; the principal effect of a finite ϵ_d will be mentioned.

The circulating current phase.—First, I derive some results of Ref. [3] in a simpler way by a simpler treatment of the large on-site repulsions in Eq. (3) and calculate the phase diagram of the CC phase. In the limit $(U_d, U_p) \gg (t_{pd}, t_{pp})$, a good mean-field approximation [9] for low density of holes (or electrons) consists in replacing $t_{pd} \to \bar{t}_{pd} = t_{pd}|x|$; $t_{pp} \to \bar{t}_{pp} = t_{pp}|x|$, where $|x|$ is the deviation from half filling in the conduction band: $x > 0$ for holes, and $x < 0$ for electrons. A more general treatment, which does not change any of the essential results, considers separately the average occupation in the oxygen and copper orbitals and renormalizes t_{pd}, t_{pp} accordingly.

A mean-field (nonsuperconducting) order parameter is sought which does not break translational symmetry. This means that the mean-field Hamiltonian is just a change of the coefficients in the kinetic energy operator K. Most of the possible mean-field decompositions of the interactions change only the magnitude of the coefficients while preserving the symmetry. The only interesting mean-field decomposition comes from $H_{\rm int}^{(2)}$ and yields the complex mean-field order parameter,

$$Re^{i\phi} \equiv V/2 \sum_{\mathbf{k}\sigma} s_x(k) \langle d^+_{\mathbf{k}\sigma} p_{x\mathbf{k}\sigma}\rangle - s_y(k) \langle d^+_{\mathbf{k}\sigma} p_{y\mathbf{k}\sigma}\rangle\,. \tag{5}$$

The mean-field Hamiltonian itself is

$$H_{mf} = K - Re^{-i\phi} \sum_{\mathbf{k}\sigma} [s_x(k) d^+_{\mathbf{k}\sigma} p_{x\mathbf{k}\sigma} - s_y(k) d^+_{\mathbf{k}\sigma} p_{y\mathbf{k}\sigma}] + \text{H.c.} + \frac{R^2}{2V}\,. \tag{6}$$

The energy of each of the three bands obtained by diagonalizing Eq. (6) is changed by a finite (R, ϕ). But for any k the trace of the change in energy of the three bands is zero. The change in energy can therefore be expressed purely in terms of the change in energy $\delta\epsilon_{ck}$ of the filled part of the (hole) conduction band. (The expression for $\delta\epsilon_{ck}$ and several other details is given in [10].) So the mean-field values R_0 and ϕ_0 are determined by minimizing

$$\frac{R^2}{2V} - 2\sum_{\mathbf{k}} \delta\epsilon_{c\mathbf{k}}(\phi, R) f(\epsilon_{c\mathbf{k}})\,. \tag{7}$$

From Eq. (7), it is deduced that $R_0 \neq 0$ at $T = 0$ for

$$\frac{2|x|\hat{t}_{pd}}{V} < \sum_{\mathbf{k}<k_F} \left(s^2_{xy}(k) + \frac{8t_{pp}}{t_{pd}} \frac{s^2_x(k) s^2_y(k)}{s^2_{xy}(k)} \right), \tag{8}$$

and ϕ_0 is $\pi/2$ or $-\pi/2$. The symmetry of the transition is therefore of the Ising variety.

For $|x| \ll 1$, Eq. (8) is satisfied only for x less than critical doping $|x_c|$,

$$|x_c| \approx \frac{1}{2}\,(V/t_{pd})\,(0.25 + 0.5 t_{pp}/t_{pd})\,. \tag{9}$$

$|x_c|$ defines the quantum-critical points for both electron and hole dopings. $\theta_0 \equiv R_0/(2\bar{t}_{pd})$ obtained by expanding Eq. (7) to $O(R^4)$ is given to within numerical factors of $O(1)$ by $\theta_0^2 = \bar{t}^2_{pd}(x_c - x)/2[V(t_{pp} + t_{pd})]$. For $\epsilon_d \neq 0$, t^2_{pd} is replaced by $t^2_{pd} + O(\epsilon^2_d)$. With V, t_{pp}, t_{pd}, and ϵ_d of similar magnitude, as for CuO compounds, x_{ec}, x_{hc} are therefore about 0.2. The line of transitions at finite temperatures varies with x as $T_p/E_F \sim |x_c - x|^{1/2}$. This is to be identified with $T_p(x)$ of Fig. 1.

The energy of the (transverse) fluctuations in ϕ about ϕ_0 at long wavelengths is estimated from Eq. (7) to be

$$\Omega_0^0 = O[\theta_0^2(\bar{t}_{pp} + \bar{t}_{pd})/4]\,. \tag{10}$$

The variation of $\Omega^0_{\mathbf{q}}$ with q is estimated to be slow, of $O(qa)^2$.

The eigenvectors of the states in the conduction band to leading order in θ and t_{pp}/t_{pd} are

$$|c_{\mathbf{k}\theta\sigma}\rangle = \frac{1}{N_k}\left[|c_{\mathbf{k}o\sigma}\rangle + i2\sqrt{2}\,\theta_0 \frac{\bar{t}_{pp}}{\bar{t}_{pd}} s_x(k) s_y(k) |a_{\mathbf{k}o\sigma}\rangle \right], \tag{11}$$

where $|a_{\mathbf{k}o}\rangle$ and $|c_{\mathbf{k}o}\rangle$ are the nonbonding and conduction band states of k for $\theta = 0$, and N_k is the normalization factor. The term proportional to θ_0 causes time-reversal breaking. Similarly, expressions can be derived for the other two bands. The three together correspond to a current carrying state with the pattern shown in Fig. 6 of Ref. [3].

The CC phase breaks a rotational invariance of the Cu-O lattice (besides the time-reversal invariance). So lattice defects, such as interstitials, dislocations, or grain boundaries, couple as external fields do to a $d = 2$ Ising order parameter [11]. No thermodynamically sharp transition is therefore possible at $T_p(x)$.

Anisotropic gap in the normal state.—The source of the instability above is interband transitions at $k = 0$. Therefore the relative energies of the three bands shift with nothing special happening at the chemical potential. This solution will now be shown to be modified by the fluctuations which lead to scattering among the states [Eq. (11)] near the chemical potential. The Hamiltonian for such fluctuations, generated by operators $\delta\phi_q$, $\delta\phi^+_q$,

is derived to be

$$H_{\rm fluct} = \sum_{\mathbf{q}} \Omega^0_{\mathbf{q}} \delta\phi^+_{\mathbf{q}} \delta\phi_{\mathbf{q}} + \sum_{\mathbf{k}\mathbf{k}'\sigma} g(\mathbf{k},\mathbf{k}') c^+_{\mathbf{k}'\theta\sigma} c_{\mathbf{k}\theta\sigma} (\delta\phi_{\mathbf{k}-\mathbf{k}'} + \delta\phi^+_{\mathbf{k}'-\mathbf{k}}) + \text{H.c.}, \tag{12}$$

where, to zeroth order in t_{pp}/t_{pd},

$$g(\mathbf{k},\mathbf{k}') = \theta_0 \bar{t}_{pd} [s_x(k) s_x(k') - s_y(k) s_y(k')]/s_{xy}(k). \tag{13}$$

For $\mathbf{k} \to \mathbf{k}'$, this is $\sim[\cos(k_x a) - \cos(k_y a)]$.

Consider now the renormalization of the phase-fluctuation energy through Eq. (12). For small q,

$$\Omega^2(\mathbf{q}) = \Omega^{02}_{\mathbf{q}} - 2\Omega^0_{\mathbf{q}} \bar{t}^2_{pd} \theta^2_0 (1 - q^2/8) \sum{}' [\cos(k_x a) - \cos(k_y a)/s_{xy}]^2/(\epsilon_{\mathbf{k}+\mathbf{q}} - \epsilon_{\mathbf{k}}), \tag{14}$$

with the restriction on the sum that $\mathbf{k} < \mathbf{k}_f$ and $\mathbf{k} + \mathbf{q} > \mathbf{k}_f$. The two terms in (14) have similar magnitudes suggesting that the phase fluctuations can be unstable over a substantial part of momentum space. A way to restore stability is by a condensation of the phase fluctuations.

The condition for stability is derived by the ansatz that $\langle \delta\phi_{\mathbf{q}} \rangle$ and $\langle c^+_{\mathbf{k}+\mathbf{q},\theta\sigma} c_{\mathbf{k}\theta\sigma} \rangle$ are finite for a range of $\mathbf{q}$ around zero [12]. Equation (12) then leads to new eigenstates which are annihilated by

$$\gamma_{k\sigma} = \left(c_{\mathbf{k}\theta\sigma} + \sum_{q \neq 0} u_{k,\mathbf{k}+\mathbf{q}} c_{\mathbf{k}+\mathbf{q}\theta\sigma} \right) \Big/ M_k. \tag{15}$$

M_k is the normalization. It will turn out that $u_{k,\mathbf{k}+\mathbf{q}} \neq 0$ only for states with $|(\epsilon(\mathbf{k}), \epsilon(\mathbf{k}+\mathbf{q})) - \mu| \lesssim \Omega^0_0$. For such states, momentum is not a good quantum number; I label the new states by k to indicate that the *average* momentum of such a state is $\mathbf{k}$. This unusual symmetry breaking is essential to gain phase space for scattering near k_f.

From Eq. (12),

$$\langle \delta\phi_{\mathbf{q}} \rangle = -\frac{1}{\Omega^0_{\mathbf{q}}} \sum{}' g(\mathbf{k},\mathbf{k}+\mathbf{q}) \langle c^+_{\mathbf{k}+\mathbf{q},\theta\sigma} c_{\mathbf{k}\theta\sigma} \rangle. \tag{16}$$

I use the Brillouin-Wigner (BW) self-consistent approximation to get

$$\langle c^+_{\mathbf{k}+\mathbf{q},\theta\sigma} c_{\mathbf{k}\theta\sigma} \rangle = g(\mathbf{k},\mathbf{k}+\mathbf{q}) \langle \delta\phi_{\mathbf{q}} \rangle / (E_k - E_{k+q}), \tag{17}$$

where E'_ks are the new one-particle eigenvalues to be determined. The BW approximation is exact in the limit that the number of states $(\mathbf{k}+\mathbf{q})$ coupled to a given state $\mathbf{k}$ is very large.

Combining Eqs. (16) and (17) yields the self-consistency equation,

$$\Phi(k, k+q) = g(\mathbf{k},\mathbf{k}+\mathbf{q})/\Omega^0_{\mathbf{q}} \sum{}' g(\mathbf{k}',\mathbf{k}'-\mathbf{q})/(E_{k'-q} - E'_k) \Phi(k', k'-q), \tag{18}$$

where $\Phi(k, k+q) = (E_{k+q} - E_k) \langle c^+_{\mathbf{k}+\mathbf{q}\theta\sigma} c_{\mathbf{k}\theta\sigma} \rangle$. In Eq. (18), the sum is restricted to states k, etc., such that $|\epsilon(k) - \mu| \lesssim \Omega^0_0$, due to the retarded interaction.

For a solution of Eq. (18), note that, since $q \to 0$, $g(\mathbf{k},\mathbf{k}+\mathbf{q}) \sim [\cos(k_x a) - \cos(k_y a)]$, for $q \to 0$, $\Phi(\mathbf{k},\mathbf{k}+\mathbf{q}) \sim [\cos(k_x a) - \cos(k_y a)]$ also. Second, $E(k) \approx \epsilon(\mathbf{k})$ for $|\epsilon(\mathbf{k}) - \mu| \gtrsim \Omega^0_0$. A consistent solution for k near k_f is then

$$E_k = \epsilon_{\mathbf{k}} \pm D(k), \qquad k \gtrless k_f, \tag{19}$$

$$D(k) = D_0 [\cos(k_x a) - \cos(k_y a)]^2, \tag{20}$$

as may be verified by substitution. The value of D_0 is evaluated to be of $0[\theta^2_0 \bar{t}^2_{pd} \rho(0)]$.

The gap in the one-particle spectrum at the chemical potential has a magnitude D_0 which can be estimated to be $x(x_c - x) t^2_{pd}(t_{pd} + t_{pp})/(V t_{pp})$ which, at $(x_c - x) \approx 0.05$, and $t_{pd} \sim V \sim t_{pp} \sim 1$ eV, is $\approx$20 meV. This has the right order of magnitude [4]. The differences of (19) from the D-wave BCS single-particle spectrum are significant and have observable consequences discussed below.

The leading decrease in energy due to the modification of the single-particle spectrum is $\sim\theta^2_0$. Therefore, the nature of the transition remains unchanged, at least in mean-field theory; only quantitative changes are introduced in the condition [Eq. (8)] for the occurrence of the CC phase.

Properties in the circulating current phase.—The single-particle density of states in the CC phase calculated from Eq. (19) is equal to $\rho(0)$, the normal density of states for $\omega/D > 1$, and to

$$\rho_{cc}(\omega) = \rho(0) \frac{2}{\pi} \arcsin\left(\left| \frac{\omega}{D} \right|^{1/2} \right), \qquad \left| \frac{\omega}{D} \right| \leq 1. \tag{21}$$

This increases as $|\omega/D|^{1/2}$ for $|\omega/D| \ll 1$ and, unlike the d-wave superconductors (which have a logarithmic singularity at $\omega = \Delta$, the superconducting gap), $\rho_{cc}(\omega)$ is less than $\rho(0)$ at all $|\omega| < D$.

Equation (21) should be compared with the single-particle density of states measured in tunneling [13] and with the specific heat [5] C_v and magnetic susceptibility [6] χ. The former shows a diminution in the single-particle density of states for low energies at $T \lesssim T_p(x)$ but shows no rise above $\rho(0)$ at finite energies until $T \lesssim T_c(x)$, when the characteristic superconducting density of states appears. C_v in the CC phase is predicted $\sim T^{3/2}$ and $\chi \sim T^{1/2}$ for $T \ll T_p(x)$. Because of the intervention of

superconductivity, it is hard to test these power laws accurately. In the measured range [5,6], $T\chi/C_v$ is nearly independent of temperature as predicted here. $\chi(T)$, measured more accurately than $C_v(T)$, can be fit to $T^{1/2}$. One can deduce the continuation to the T dependence below T_c by invoking conservation of entropy on the $C_v(T)$ measurements. $C_v(T) \sim T^{3/2}$ for $T \ll T_p(x)$ is then not inconsistent, while $C_v(T) \sim T^2$ clearly is.

To obtain the spectral function $A(k, \omega)$ measured by ARPES [4], one needs the single-particle self-energy besides Eq. (19). The self-energy differs in an important way from the corresponding calculation for a d-wave superconductor [14] (and is quite different from s-wave superconductors) [15]. In both cases, the bare polarizability $\chi_0(q, \omega)$ is zero for $\omega < D(q)$, [$\Delta(q)$ for the superconductors]. The lowest energy single-particle scattering for momentum q occurs by an intermediate one-particle state near the zero of the gap. Therefore the threshold for single-particle scattering of a state at q is also $D(q)$. For $\omega \gtrsim D(q)$, $\chi_0(q)$ with E_k's given by Eq. (19) is proportional to $\omega/D\epsilon_F$. So the renormalized $\chi(q, \omega)$ for $\omega > D(q)$ is similar to the normal state above $T_p(x)$, i.e., of the marginal Fermi-liquid form. The single-particle self-energy for $(\omega, T) \ll D(q)$ is exponentially small, but for $(\omega, T) \gtrsim D(q)$ it returns to the value $\Sigma_n(\omega, q, T)$ without the pseudogap. By interpolation from the known form at low and high energies,

$$\mathrm{Im}\Sigma(\omega, \mathbf{q}, T) \approx \mathrm{sech}\left(\frac{D(\mathbf{q}, T)}{(\omega^2 + \pi^2 T^2)^{1/2}}\right) \mathrm{Im}\Sigma_n(\omega, \mathbf{q}, T)\,. \tag{22}$$

The spectral function at the Fermi wave vectors $\hat{k}_f$ defined by $E(\hat{k}_f) = \pm D(\hat{k}_f)$, calculated using Eqs. (19) and (22) and the marginal Fermi-liquid form for Σ_n, is plotted in Fig. 2 for a few temperatures. A pseudogap in the direction $\hat{k}_f$ appears below $T \approx D(\hat{k}_f)$ producing the illusion of "Fermi arcs" shrinking as temperature decreases. The line shape in Fig. 2 at low energies, in the pseudogap region, fits the experimental curves [4] well within the experimental resolution. A prediction following from the results of the previous section is that $D \sim (x_c - x)$ and also $\sim T_p(x) - T$.

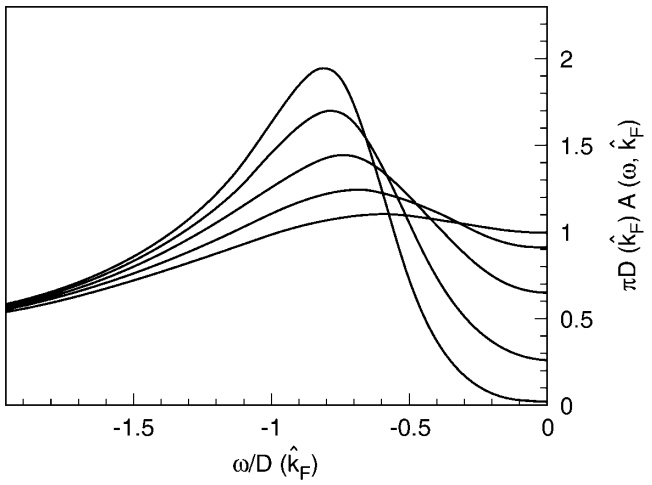

FIG. 2. The spectral function at the Fermi wave vectors as a function of energy below the chemical potential normalized to the gap in the direction of that wave vector $D(\hat{k}_f)$ for temperatures $T = nD(\hat{k}_f)/4\pi$ for $n = 1$, 2, 3, 4, and 5.

The promotion of d-wave or s-wave superconductivity by current fluctuations, depending on the Fermi surface at a given doping, has been derived elsewhere [10]. In conclusion, I note that although I have presented a systematic theory in agreement with the principal experimental results, it is only a mean-field theory. One can be confident of its applicability only if the CC phase is directly observed. It is possible that, in an improved theory, the quasiparticles are topological excitations of fermions bound to comoving current fluctuations.

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