Theory of Copper-Oxide Metals

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A model for copper-oxide metals with large local repulsion on copper and Coulomb interaction between ions is solved systematically to get the leading low-energy behavior of physical properties. The one-particle spectra is of a marginal Fermi liquid at the density where mean-field solutions display a valence degeneracy of copper ions. The metallic state is unstable to superconductivity with an electronic energy scale.

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Landau Fermi-liquid theory and associated quasiparticle concepts appear not to be valid in the normal state of copper-oxide metals [1–3]. A useful guide in the pursuit of the theory of these metals and their superconductive instability is that these phenomena do not occur in other transition metal oxides, or sulfides or selenides, etc., either quasi-two-dimensional or three-dimensional. The appropriate model for copper oxides should then have special features rooted in their special chemistry.

At half-filling the copper oxides are antiferromagnetic insulators with the lowest one-electron excitations on Cu ($Cu^{++} \rightarrow Cu^{+}$) and the lowest one-hole excitations on oxygen ($O^{--} \rightarrow O^{-}$). The lowest optically active particle-hole excitation has the charge-transfer character ($Cu^{++}O^{--} \rightarrow Cu^{+}O^{-}$) with an energy ~ 1.8 eV. The ionic contribution to these excitations due to Coulomb interactions, i.e., the difference in Madelung energy on Cu and on oxygen is estimated [4] to be much larger, ~ 10 eV. The principal physical feature of one of the models [5] proposed for copper oxides is that, in this unique situation, long-range ionic interactions, besides the on-site repulsion on the copper d orbitals, play a crucial dynamical role for the low-energy excitations in the metallic state. The model is given by the Hamiltonian

$$H = \sum_{\langle ij\rangle,\sigma} t(d^{+}_{i\sigma} p_{j\sigma} + \text{H.c.}) + \Delta_0 \sum_j (n^d_j - n^p_j) + U \sum_j n^d_{j\uparrow} n^d_{j\downarrow} + V \sum_{\langle ij\rangle} n^d_i n^p_j , \quad (1)$$

where d^{+} creates holes on the copper $d_{x^2-y^2}$ orbitals, p^{+} creates holes on the oxygen p orbitals, and t_{pd} are nearest neighbor hopping integrals. $U \rightarrow \infty$ corresponds to the neglect of Cu^{+++} states. The model cannot be reduced, in the metallic state, to the Hubbard model for $V \gtrsim \Delta_0, t$, where new physics might be expected.

Several mean-field calculations [6,7] (Hartree-Fock, Gutzwiller variational, large N slave boson, etc.) on the model of (1) reveal that, as the density of holes is increased in the metallic state, the energy of the $Cu^{++}O^{--} \rightarrow Cu^{+}O^{-}$ excitation decreases due to the ionic interactions. The energy is *bistable* as a function of the ratio of holes on oxygen to holes on Cu, $\langle n_O/n_{Cu}\rangle$, beyond some density x of holes, defined as $\langle n_{Cu}\rangle + \langle n_O\rangle = 1 + x$. For some concentration x_c two degenerate values of $\langle n_O\rangle/\langle n_{Cu}\rangle$ result. The bistability ends at a critical point near which RPA [6] and slave boson calculations [7] show a superconductive instability promoted by charge-transfer excitations. $\langle n_O\rangle/\langle n_{Cu}\rangle$ is coupled to the uniform density leading to phase separation. Numerical diagonalization of the model [8] for a small number of atoms in the ring geometry gives similar results as well as long-range superconducting correlations.

Here, we consider a more general model by replacing the interactions V by Coulomb interactions between the ions, because, as seen below, precise satisfaction of the screening conditions in the metal is crucial to eliminate phase separation [9] in the model. Their intracell parts are handled by a constraint on the states allowed in a cell. In mean-field calculations on the resulting model, a local valence instability, where Cu^{++} ions with their associated ionic screening and polarization become degenerate with Cu^{+} ions with their associated screening and polarization, again arises for some deviation x from half-filling. Such a degeneracy puts in jeopardy the assumption of adiabaticity as a function of the ionic interactions in Landau theory [10]. Fluctuation near the point of degeneracy have singular low-energy resonances of a form that systematic calculations are possible.

We start by defining a *basis set* by diagonalizing states of a cell i by putting the kinetic energy terms connecting a cell i to its neighbors to zero and keeping the low energy states. We interpret V in (1) as the intracell part of the Coulomb interactions [screened by processes at energies larger than $O(t)$]. We work at an *average* occupation of $(1 + x)$ holes per cell as required by $(1 + x)$ negative charges per unit cell assumed uniformly distributed by imposing a chemical potential μ. The minimum basis set must then include one-hole and two-hole states per unit cell. The zero-hole state $|0\rangle_i$ as well as three or higher hole states are then costly in energy compared to the one-hole and two-hole states by $O(V)$. They are excluded as is due to $U \rightarrow \infty$, the two-hole state on Cu. The allowed low-energy states are the one-hole state on Cu denoted by $d^{+}_{1\sigma i}|0\rangle$, the one-hole state on the linear combination of oxygen orbitals in the cell i that hybridizes with the Cu orbital in cell i denoted by $d^{+}_{2\sigma i}|0\rangle$, and

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the two-hole singlet states $\phi_{1i}^{+}|0\rangle$ and $\phi_{2i}^{+}|0\rangle$, which are, respectively, the singlet with one hole on Cu and the other on the oxygens and with both holes on oxygens [11]. The ϕ_i's should be thought of as hard core boson operators. $d_{2\sigma i}^{+}|0\rangle$ is the lowest of the one-hole oxygen states by virtue of its hybridization to $d_{1\sigma i}^{+}|0\rangle$. The bare operators in (1) expressed in terms of the constrained operators are

$$d_{i\sigma}^{+} = \phi_{1i}^{+} d_{2i-\sigma}; \qquad P_{i\sigma}^{+} = \phi_{1i}^{+} d_{1i\sigma} + \phi_{2i}^{+} d_{2i-\sigma}, \quad (2)$$

where $P_{i\sigma}$ is the linear combination of $P_{i\sigma}$, which hybridizes with $d_{i\sigma}$.

The allowed states in a cell i must fulfill the completeness relation or constraint

$$\psi_i^{+}\psi_i + \phi_i^{+}\phi_i = 1, \quad (3)$$

where $\psi_i = (d_{1\uparrow} d_{1\downarrow} d_{2\uparrow} d_{2\downarrow})_i$ and $\phi_i = (\phi_1 \phi_2)_i$. The kinetic energy in (1) mixes the one-hole and two-hole sectors in the same and neighboring cells. Using (2),

$$H_1 = \sum_{(ij),\sigma} t_{ij} \phi_{1i}^{+} d_{2\sigma i} (d_{1\sigma j}^{+} \phi_{1j} + d_{2\sigma j}^{+} \phi_{2j}) + \text{H.c.} \quad (4)$$

Here both $j = i$ and j nearest neighbors to i are summed; t_{ij} differs from t by numerical constants, which depend on lattice structure. The kinetic energy in (1) also operates on the one-hole states of neighboring cells i and j, creating the disallowed states $|0\rangle_i$ or $d_{1i\sigma}^{+} d_{1i-\sigma}^{+}|0\rangle$. Eliminating such high-energy states leads to a low-energy sub-Hamiltonian, acting on the space of singly occupied cells, which is a straightforward generalization of the Heisenberg Hamiltonian

$$H_2 = -J \sum_{(ij)} \left(\sum_{\sigma,\tau} a_\tau \psi_{i\sigma\tau}^{+} \psi_{j\sigma\tau} \right) \times \left(\sum_{\sigma'\tau'} a_{\tau'} \psi_{j\sigma'\tau'}^{+} \psi_{i\sigma'\tau'} \right), \quad (5a)$$

$$= -J \sum_{(ij)} (1 - \boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j)(1 - \boldsymbol{\tau}_i \mathbf{A} \boldsymbol{\tau}_j). \quad (5b)$$

In (5) $\boldsymbol{\sigma}$'s and $\boldsymbol{\tau}$'s are Pauli matrices in spin and $d_1 - d_2$ space, respectively, $J \approx 0(t^2/V)$. Equation (5a) exhibits the singlet valence bond form suggested by Anderson and collaborators [12]. a_τ and the anisotropy tensor $\mathbf{A}$ arise because the local Hamiltonian of a cell i is in general not rotationally invariant in $\boldsymbol{\tau}$ space. It is very important, however, that at the valence degeneracy point defined below, where the effective local Hamiltonian is diagonal, $\mathbf{A}$ has only a uniaxial anisotropy: $\mathbf{A} \equiv (1, 1, A)$.

The second term in (1) transforms to

$$H_3 = \Delta_0 \sum_i [(n_{1i} - n_{2i}) - 2\phi_{2i}^{+}\phi_{2i}]. \quad (6)$$

The total Hamiltonian is $H = H_1 + H_2 + H_3$. For $\Delta \gg t, J$ and $x = 0$ (half-filling), H reduces to the Heisenberg Hamiltonian $J(-1 + \boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j)$ for low energies.

We now perform a standard mean-field calculation [13] together with fluctuations about it by introducing fields

$$\varepsilon_{ij} = J \sum_{\sigma,\tau} a_\tau \psi_{i\sigma\tau}^{+} \psi_{j\sigma\tau} \equiv \sum_{\sigma} \psi_{i\sigma} (\varepsilon_{ij}^{s} \mathbf{1} + \varepsilon_{ij}^{a} \boldsymbol{\tau}_z) \psi_{j\sigma}, \quad (7a)$$

$$\{\mathbf{M}_i\} = \psi_i^{+} \{\mathbf{m}\} \psi_i. \quad (7b)$$

Here $\{\mathbf{m}\} = \mathbf{1}, \boldsymbol{\tau}, \boldsymbol{\sigma}, \boldsymbol{\sigma}\boldsymbol{\tau}$. Also, we introduce a Lagrange multiplier λ_i to enforce the constraint. We look for a solution with spin singlet in the $(i - j)$ bond as in (7a). The only nonzero mean-field components of $\{\mathrm{M}\}$ found in the mean-field solution are the components of τ, which are taken as T_z and T_x. This requires only that $J \gtrsim tx$. Both (6) and (5b) favor spatially *uniform* T_z and T_x. The free energy has a saddle point at the uniform state $\lambda_i = \lambda_0$, $\varepsilon_{ij} = \varepsilon$ for i, j nearest neighbors, $\phi_{i1} = \phi_1, \phi_{i2} = \phi_2$. The mean-field Hamiltonian for fermions diagonalized to momentum space ($\mathbf{k}$) is

$$H_{mf} = \sum_{\mathbf{k}\sigma} \psi_{\sigma\mathbf{k}}^{+} [(\varepsilon^{s}(\mathbf{k}) - \lambda_0)\mathbf{1} + (h_z(\mathbf{k}) + JAT_z)\boldsymbol{\tau}_z + (h_x(\mathbf{k}) + JT_x)\boldsymbol{\tau}_x] \psi_{\sigma\mathbf{k}}, \quad (8)$$

where $\boldsymbol{\tau}$ is quantized along the x axis and

$$h_z(\mathbf{k}) = -2t_k \phi_1^2, \qquad h_x(\mathbf{k}) = \Delta_0 + t_k \phi_1 \phi_2 + \varepsilon_{\mathbf{k}}^{a}. \quad (9)$$

The mean-field energy may now be minimized with respect to $\lambda_0, \phi_1, \phi_2, T_z, T_x$, and $\varepsilon(k)$. The details of this calculation will be presented elsewhere. The chemical potential μ is fixed so that $\phi_1^2 + \phi_2^2 = x$; λ_0 determines the occupation $\sum_\sigma \langle \psi_\sigma^{+} \psi_\sigma \rangle = 1 - x$. Luttinger's theorem for the physical particles is then satisfied. The coefficients of the "external fields" h_z and h_x are functions of the concentration x. As x increases $\phi_1 \phi_2$ and ϕ_1^2 increase so that at a critical value $x = x_c$ may be found where $h_x(0) = 0$. *At this point the model has rotational symmetry about the z axis.*

Consider next, the fluctuations about the mean-field solution. The fluctuation Hamiltonian is given by the harmonic Hamiltonian of the fields ϕ, and $\{\mathrm{M}\}$ and the linear coupling of the fields to the fermion eigenstates of H_{mf} obtained by expanding (4), (5), and (6) about the mean field. Given the constraint (3), the fluctuations in the density [$m = \mathbf{1}$ in (7)] in a cell from its average value are the fluctuations between the one-hole density $\psi_i^{+}\psi_i$ and the two-hole density $\phi_i^{+}\phi_i$. These couple to the long-range Coulomb (r^{-1}) interactions, and lie near the plasma energy ω_p and can be ignored. Were we to ignore Coulomb interactions, such fluctuations would constitute the zero-sound modes, whose velocity would be suppressed to zero [7] as the charge transfer (τ modes) considered below comes down in energy. This leads to phase separation in models without Coulomb interactions. Note that the density of charge excitations is x per unit cell. So $\omega_p^2 \sim x$.

The other collective modes, evident from the constraint (3), are the internal $d_1 - d_2$ fluctuations, the phase modes of ϕ's, and the internal $\phi_1 - \phi_2$ modes.

Local gauge invariance requires the phase of all operators to be identical. Because of finite $\langle\phi_i\rangle$ and the resultant finite λ_0, the phase modes are massive [7] and can be ignored. A leading order calculation of $\langle\phi_{1i}^{+}(t)\phi_{2j}(0)\rangle$ using H_1 shows that they are proportional to the particle-hole excitation spectra of a Fermi liquid that is broad and incoherent. There are three kinds of $d-d$ modes expected from H_2, Eq. (5). The charge-transfer fluctuation ($\boldsymbol{\tau}$) modes, the spin fluctuation ($\boldsymbol{\sigma}$) modes, and the coupled spin and charge transfer ($\boldsymbol{\sigma\tau}$) modes. Because of finite $\langle\tau\rangle$ in the mean-field calculation, the latter two are mutually coupled, but they are uncoupled to the $\boldsymbol{\tau}$ modes and are in general massive.

Now we calculate the $D_{\alpha\beta}(q,\omega) = \langle\tau_\alpha\tau_\beta\rangle(q,\omega)$ fluctuations by eliminating the linear coupling of τ's to ψ's and ϕ's. The $\langle\tau_z\tau_z\rangle$ modes are always massive because there is a field in the τ_z direction. Consider the point $x = x_c$, where, as mentioned, the model has rotation symmetry about the τ_z axis. Two sets of modes must now be distinguished, the amplitude modes and the phase modes in the $\tau_x - \tau_y$ plane. The amplitude modes (taken to be $\langle\tau_x\tau_x\rangle$) are in general also massive. Because of rotational symmetry, the phase mode $\langle\tau_y\tau_y\rangle$ is required to be massless at $q = 0$. However, this mode is not the mode of a conserved quantity and is therefore damped. Near $x = x_c$ and at temperature zero, the phase mode is

$$D_{yy}(q,\omega) \approx \frac{B^{-1}}{i\omega/\gamma(q) + a^2q^2 + G(x)}, \quad \text{for } \omega \lesssim v_F q. \tag{10}$$

In (10) B is of the order of the bandwidth and a of a lattice constant. $G(x) \to 0$ as $x \to x_c$. The "gap" $G(x) \approx |\Delta_0 + 4tx|/B$. The damping $\gamma(q)$ is given by γ_0 for $v_F q \lesssim \gamma_0$ and by Landau damping $\sim(v_F q)$ for $v_F q \gtrsim \gamma_0$, where γ_0 is due to disorder or inelastic scattering. Collective modes of the type (10) violate Landau's adiabaticity criteria for $d \leq 3$. If we approach x_c from $x > x_c$, where the effective field on τ_x is opposite in sign, the same phenomena occurs. Thus $x = x_c$ is an isolated critical point at $T = 0$.

The physics of the new results may be best discussed by comparing it with the charge transfer instability already found by standard RPA calculations [6] and by auxiliary boson methods [7] followed here on the model of Eq. (1). Detailed numerical calculations were reported which show that the instability is possible for reasonable parameters. The differences here from previous calculations is that the use of Coulomb interaction (a) eliminates phase separation, and (b) the long-range interactions are handled by a constraint, Eq. (3), which operates on both oxygen and copper states rather than copper states alone. The interesting degrees of freedom per unit cell are represented by the pseudo-spin τ, which expresses both charge transfer and current degrees of freedom rather than an Ising degree of freedom representing charge transfer alone. The general Hamiltonian in τ space is a uniaxially anisotropic interaction Hamiltonian with an "external" field in an arbitrary direction. As a function of electron concentration the field aligns with the anisotropy axis at $x = x_c$. At this point we have a massless phase degree of freedom at long wavelengths. The phase diagram is different from the earlier calculations [6,7] in an essential way: Instead of a first order transition ending at a critical point, which must be reached by varying more than one parameter, we have a critical point at $T = 0$ reached only by varying x in the model studied.

The critical mode is at $q = 0$, but it is an internal (breathing) mode of the unit cells. It does not couple to the uniform density, and no third order invariant appears allowed. If there is no significant nesting in the band structure, no mixing of the critical mode with charge density or spin-density modes at finite q occurs.

The interaction of fermions with bosons of the form (10) is being thoroughly investigated [14–16]. The leading term in the self-energy for the one-particle Green's functions calculated by second order perturbation has the marginal Fermi liquid [3] frequency dependence [14–16]

$$\Sigma(\omega) \sim \omega\ln\omega + i\omega\,\mathrm{sgn}\omega, \tag{11}$$

at $x = x_c$, with a negligible momentum dependence for $d = 3$. For uniaxial anisotropy in $d = 3$, (10) should be generalized by $a^2q^2 \to a_z^2q_z^2 + a_\rho^2q_\rho^2$ and $\gamma(q) \approx \gamma_0 + v_{Fz}q_z + v_{F\rho}q_\rho$. Then $\Sigma'(\omega) \sim \omega\ln\omega$ for $\gamma_0 \approx 0$, as well as for the case $v_{Fz}q_z \ll \gamma_0$ provided $a_2^2q_{zc}^2 \geq \omega$. Here q_{zc} is the upper cutoff in q_z. Note that since the stiffness in the collective modes comes from the (screened) Coulomb interactions, a_z and a_ρ are expected to be of comparable magnitude. Thus even with incoherence in the z direction, i.e., typical $v_{Fz}q_z \ll \gamma_0$, a $d = 3$ calculation is appropriate.

Corrections to the second order result (11) are of two kinds, vertex corrections and corrections due to insertions of the renormalized fermion Green's functions: The simplifying feature in the problem is that with (10), the momentum exchanged is much larger than the energy exchanged. Both the corrections are then unimportant [14–16] in $d = 3$ just as in the Migdal argument for electron-phonon interactions. (11) then gives the asymptotically exact dependence of the self-energy.

The dispersion obtained from Eqs. (8) and (9) has the same symmetry as in one-electron calculations. Also the Fermi surface encloses the total number of electrons, so the Luttinger theorem is obeyed. However, as already discussed the number of current *carriers,* or the total metallic spectral weight in optical conductivity is x, consistent with experiments [17]. For $G \neq 0$, $\mathrm{Im}\Sigma(\omega) \sim (\omega/G)^2$ for $\omega \to 0$ characteristic of a Fermi liquid. Thus for $x \neq x_c$, a crossover to a Fermi-liquid behavior for $T \lesssim G$ is predicted.

Because of umklapp scattering (which is implied since the collective modes (11) are interband modes), Eq. (12) implies a momentum transport scattering rate $\sim T$ in $d = 3$. The necessity of umklapps in transport is also

seen from the fact that the observed ratio of the thermal and electrical conductivity obeys the Wiedemann-Franz law. In $YBa_2Cu_3O_7$ the resistivity in the plane ρ_{ab} has a linear temperature dependence; the resistivity perpendicular to the planes ρ_c is very sensitive to defects but for the best samples $\rho_c \simeq c_1 + c_2 T$ down to T_c [18]. Coherent transport thus exists in all directions, which means that a $d = 3$ theory is appropriate in the pure limit. Then the observed resistivity, thermal conductivity, optical conductivity, Raman scattering intensity, and the tunneling conductance near the hole density for the highest T_c follow [3] from Eq. (11). Coherent transport in the c direction is lost in $YBa_2Cu_3O_7$ with small amounts of disorder, and is never observed, for example, in $Bi_2Sr_2CaCu_2O_8$, presumably because the interplane hopping is much smaller. In this case $v_{Fz}q \ll \gamma_0 \ll v_F q$ appears appropriate and only the in-plane component of momentum in the interaction of fermions with the charge-transfer resonances is conserved. As discussed, Eq. (11) remains valid.

Careful resistivity measurements [19] have recently clarified that the linear temperature dependence of ρ_{ab} is observed down to T_c only near the density of highest T_c, with higher temperature dependence at low temperatures not inconsistent with a Fermi liquid around it. (The effects of disorder are also prominent at lower densities.) This is consistent with the developments here; a marginal Fermi-liquid behavior is to be expected only for $T \gtrsim G(x)$ with a crossover to Fermi liquid at lower temperatures.

The charge transfer fluctuations also provide an effective interaction in the particle-particle channel, for particles of momenta $k, k + q$ on the Fermi surface, just as found earlier [6,7],

$$V_{pp}(\mathbf{k}, \mathbf{k} + \mathbf{q}) \sim D(\mathbf{q}, \varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}+\mathbf{q}}). \qquad (12)$$

Since vertex corrections merely renormalize the coupling constant, an Eliashberg theory for superconductivity can be formulated. The absence of pair amplitude on Cu, because $U \approx \infty$, requires that the gap pair wave function $\psi(k) \equiv \sum_\sigma \langle c_{k\sigma} c_{k-\sigma} \rangle$ have $\sum_k \psi(\mathbf{k}) = 0$, where the sum is over the entire Brillouin zone. So simple s-wave pairing is ruled out. The most likely pairing is of a *generalized* s-wave type [6,7], in tetragonal crystals. For generalized s waves the surface at which $\Delta(k)$ goes through zero may or may not intersect the Fermi surface. If it does the density of states in the superconductive state (for pure samples) is linear in energy; if it does not, a BCS-like gap (anisotropic, in general) is expected.

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