Theory of the Marginal-Fermi-Liquid Spectrum and Pairing in a Local Copper Oxide Model

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We solve for the properties of an impurity model abstracted from a model of copper oxide superconductors. At mixed valence, provided that Friedel's screening condition is satisfied, the marginal-Fermi-liquid spectrum is obtained for the spin and charge susceptibilities. The frequency dependence of the self-energies is $\Sigma(\omega)\sim\omega\ln\omega+i\omega$. This critical point is surrounded by two different Fermi-liquid phases. A simplified two-impurity version of this model is also addressed, and presents similar features, including a logarithmic divergence for coherent pair susceptibility.

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The quasiparticle concept and Landau Fermi-liquid theory have been cornerstones of condensed matter physics. The normal state properties [1] of the high-T_c materials appear not to conform to this framework. In an attempt to find the unifying features in the diverse observed anomalies, a phenomenological form for the spectral function for spin and charge fluctuations was proposed [2]. For $\omega\ll v_F q$, this marginal-Fermi-liquid (MFL) spectrum has the form

$$\mathrm{Im}\chi_{\rho,\sigma}(\mathbf{q},\omega)\sim\begin{cases}-N(0)\dfrac{\omega}{T}, & \omega\ll T,\\ -N(0), & T\ll\omega\ll\omega_c,\end{cases}\tag{1}$$

where ω_c is a cut-off energy. It is a scale invariant spectrum, as in the fluctuation regime of a quantum critical point. One of its consequences is that the quasiparticle residue vanishes logarithmically, $z^{-1}\sim 1+\lambda\ln(\omega_c/x)$ where $x=\max(|\omega|,T)$.

An important point about the MFL spectrum is that the singularities are in the frequency dependence; the momentum dependence is assumed smooth. It is then of interest to study strongly correlated models of an impurity embedded in a Fermi gas, where exact answers may be obtained. If the (q independent) singularities in such a model do not depend on any special symmetries which are lost in the lattice problem, they are likely to be relevant to the lattice problem. One way to see that the singularities in the single impurity problem do not get modified in any essential way is to study the stability to perturbations, including the coupling to another impurity.

An impurity model abstracted from a model of copper oxides was solved recently by Wilson's numerical renormalization group [3]. The spectrum at the mixed-valence point of impurity [such as occurs in the copper-oxide metals, where Cu fluctuates between $Cu^+(S=0)$ and $Cu^{++}(S=\frac{1}{2})$] is consistent with the MFL form. Here we obtain and extend the results analytically gaining significant new insights, and also study the effect of the perturbation from another impurity.

The impurity model studied bears the same relationship to a proposed copper oxide model [4] as the Anderson or Wolff model for a local moment bears to the Hubbard model. We consider

$$\mathcal{H}=\sum_{k,\sigma,l}\varepsilon_{kl}c^\dagger_{k\sigma l}c_{k\sigma l}+\varepsilon_d n_d+Un_{d\uparrow}n_{d\downarrow}+t\sum_{k,\sigma}(d^\dagger_\sigma c_{k\sigma 0}+\mathrm{H.c.})+\sum_{k,k',l}V_{kk'l}(n_d-\tfrac{1}{2})\left(\sum_\sigma c^\dagger_{k\sigma l}c_{k'\sigma l}-1\right).\tag{2}$$

d is the local orbital, which as required by symmetry hybridizes only with one point-group channel ($l=0$). This will be referred to as the hybridizing channel. Other channels, the screening channels, have only the ionic interaction V_l. Given finite t and large U/t, it does not matter asymptotically whether or not one keeps V_0 finite. We will take $V_0=0$. In fact, in the Wolff model, there is no separate d orbital. One must identify ε_d and U as the local level energy of the hybridizing orbital and the local repulsion at the special site.

It is convenient to change the representation to real space operators, specifically to those operating on radial shell orbitals, defined by Wilson [5]. The Hamiltonian in such an effective one-dimensional representation is $\mathcal{H}=\mathcal{H}_0+\mathcal{H}_{\mathrm{KIN}}$ where

$$\begin{aligned}\mathcal{H}_0&=\varepsilon_d(n_d-\tfrac{1}{2})+Un_{d\uparrow}n_{d\downarrow}+t\sum_\sigma(d^\dagger_\sigma h_\sigma+h^\dagger_\sigma d_\sigma)+\sum_{l,\sigma}V_l(n_d-\tfrac{1}{2})(s^\dagger_{l\sigma}s_{l\sigma}-\tfrac{1}{2}),\\ \mathcal{H}_{\mathrm{KIN}}&=\sum_{n,\sigma}(t_h h^\dagger_{n\sigma}h_{(n+1)\sigma}+t_s s^\dagger_{nl\sigma}s_{(n+1)l\sigma}+\mathrm{H.c.}),\end{aligned}\tag{3}$$

where n is the shell index, and h_σ and $s_{l\sigma}$ simply denote the hybridizing and screening electrons at the 0th shell. The screening channels may be expressed in terms of a single spinless fermion operator. Indeed, we can express the original

screening fermions operator $s_{l\sigma}$ in terms of boson fields [6]:

$$s_{l\sigma}(x)=\frac{1}{\sqrt{\Omega}}e^{-i\phi_{l\sigma}(x)}. \tag{4}$$

Then, the screening density at the impurity site is

$$\sum_{\sigma,l}(s_{l\sigma}^{\dagger}s_{l\sigma}-\tfrac{1}{2})=\frac{1}{\pi}\sum_{\sigma,l}\frac{\partial\phi_{l\sigma}}{\partial x}. \tag{5}$$

Now define $\phi_s=N^{-1/2}\sum_{\sigma,l}\phi_{l\sigma}$, as well as $(N-1)$ other orthogonal new fields, such that the transformation to the new fields is a unitary canonical transformation. The kinetic energy is invariant under this transformation, and the interacting term involves only the total charge ϕ_s. We now reexpress ϕ_s in terms of a fermion operator s through a transformation similar to (4). The screening interacting term then is

$$\hat{V}(n_d-\tfrac{1}{2})(s^{\dagger}s-\tfrac{1}{2}),\quad \hat{V}=\sqrt{N}V, \tag{6}$$

where N is the number of screening channels, assumed all to have the same interaction V.

The physical idea in the proposed route to the breakdown of the Fermi-liquid and quasiparticle concepts followed here is that for ionic interactions $\hat{V}$ above a critical value, local multiparticle resonances form at the chemical potential, drawing weight of the one-particle or hole spectra from higher energies on the scale of the charge transfer gap in the insulating state. The multiparticle resonances cannot be constructed perturbatively, putting in jeopardy the Landau correspondence between the low-lying excitations of the interacting and noninteracting fermions. To form the low energy resonances, we consider $U,\hat{V}\gg t$. Actually, we will take $U\to+\infty$ (only Cu^{+} and Cu^{++} allowed). We can now diagonalize the spectrum of the impurity. The two lowest states are

$$\eta^{\dagger}|0\rangle=|0,1\rangle,\ \text{with energy } E_{\eta}=-\frac{\hat{V}}{4}-\frac{\varepsilon_d}{2}, \tag{7}$$

$$\zeta_{\sigma}^{\dagger}|0\rangle=|\sigma,0\rangle,\ \text{with energy } E_{\zeta}=-\left[\left(\frac{\varepsilon_d}{2}-\frac{\hat{V}}{4}\right)^2+t^2\right]^{1/2},$$

where in the bras, the first number is the charge and spin of the impurity plus the hybridizing channel and the second, the charge in the screening channel. Other states are separated by energies of at least $\hat{V}$. The states in (7) satisfy the important physical requirement of the Friedel screening sum rule. In the model of Eq. (2), this is achieved by having N channels, each with the small phase shift $1/\pi N$, so that bosonization, valid only for potentials $V\ll t$ is legitimate.

We now perturbatively turn on t_s and t_h and generate an effective Hamiltonian in the subspace of the two states (7). We must also satisfy the constraint

$$\sum_{\sigma}\zeta_{\sigma}^{\dagger}\zeta_{\sigma}+\eta^{\dagger}\eta=1,\ \text{or}\ \sum_{\sigma}\zeta_{\sigma}^{\dagger}\zeta_{\sigma}\le 1. \tag{8}$$

The effective Hamiltonian so generated is

$$\mathcal{H}=\mathcal{H}_{\mathrm{KIN}}+\varepsilon\sum_{\sigma}\zeta_{\sigma}^{\dagger}\zeta_{\sigma}+\hat{t}\sum_{\sigma}(\zeta_{\sigma}^{\dagger}\eta s^{\dagger}h_{\sigma}+\mathrm{H.c.})+J\zeta^{\dagger}\sigma\zeta\cdot h^{\dagger}\sigma h+\left(\sum_{\sigma}\zeta_{\sigma}^{\dagger}\zeta_{\sigma}-\tfrac{1}{2}\right)\left[V_s(s^{\dagger}s-\tfrac{1}{2})+V_h\left(\sum_{\sigma}h_{\sigma}^{\dagger}h_{\sigma}-1\right)\right]. \tag{9}$$

$\mathcal{H}_{\mathrm{KIN}}$ is the kinetic energy of the hybridizing and the screening electrons plus, in principle, the contribution of the now uncoupled $(N-1)$ other combinations of the original screening electrons, where $\varepsilon=E_{\zeta}-E_{\eta}$, and the new coupling constants are

$$V_s\simeq\frac{2t_s^2}{\varepsilon+\hat{V}},\quad \hat{t}\simeq\frac{2\hat{t}_h t_s\hat{V}}{\hat{V}^2+\varepsilon^2},\quad J\simeq V_h\simeq\frac{2\hat{t}_h^2}{\varepsilon+V}, \tag{10}$$

with $\hat{t}_h=t_h^2/(\varepsilon+\hat{V})$. The direct hybridization of the local orbitals has disappeared in (9). It is replaced by the term proportional to $\hat{t}$, which imposes the readjustment in the Fermi sea of the hybridizing and screening electrons to go from one multiparticle resonance to another. Now, it is seen that the operators multiplying $\hat{t}$, J, V_s, and V_h are all marginal, and the terms in the kinetic energy connecting the impurity sites to farther and farther shells are successively irrelevant. The marginality of $\hat{t}$ is crucial to the subsequent developments. It is connected with the fact that there is one operator each in the screening and the hybridizing channel in the operators multiplying $\hat{t}$, which is in turn due to the obedience of the Friedel screening requirement by the impurity basis of Eq. (7). If the screening is less than this so that hybridization is relevant, we would end up with a Fermi liquid, and if more than this, the corresponding term would be irrelevant like in one of the models of Ref. [3], leading to singularities too strong for the Cu-O problem.

It is convenient to introduce the bosonic field ϕ_{σ} associated with h_{σ} by a relation similar to (4) and rotate $\mathcal{H}$ in Eq. (9) by $\mathcal{U}=\mathcal{T}_{\uparrow}\mathcal{T}_{\downarrow}$ with

$$\mathcal{T}_{\sigma}=\exp[i(\zeta_{\sigma}^{\dagger}\zeta_{\sigma}-\tfrac{1}{2})\phi_{\sigma}(0)],\quad [\mathcal{T}_{\sigma},\mathcal{T}_{-\sigma}]=0. \tag{11}$$

The Hamiltonian then transforms to $\mathcal{H}=\mathcal{H}_0+\mathcal{H}'$, with

$$\mathcal{H}_0=\mathcal{H}_{\mathrm{KIN}}+\varepsilon\left(\sum_{\sigma}\zeta_{\sigma}^{\dagger}\zeta_{\sigma}-\tfrac{1}{2}\right)+t\left(s^{\dagger}\eta\sum_{\sigma}\zeta_{\sigma}^{\dagger}+\mathrm{H.c.}\right)+J\sum_{\sigma}\zeta_{\sigma}^{\dagger}\zeta_{-\sigma}, \tag{12}$$

$$\mathcal{H}'\approx V_{\rho}\rho_h\left(\sum_{\sigma}\zeta_{\sigma}^{\dagger}\zeta_{\sigma}-\tfrac{1}{2}\right)+V_{\sigma}\sigma_h\sum_{\sigma}\sigma\zeta_{\sigma}^{\dagger}\zeta_{\sigma}. \tag{13}$$

Here, $\mathcal{H}'$ contains the longitudinal scattering terms, including those generated from $\mathcal{H}_{\mathrm{KIN}}$ by the rotation (11). ρ_h and σ_h are the charge and longitudinal spin densities of the hybridizing channel. In $\mathcal{H}_0$, only one linear combination $\alpha=(\zeta_{\uparrow}+\zeta_{\downarrow})/\sqrt{2}$ is now coupled to the conduction electrons. The other combination $\beta=(\zeta_{\uparrow}-\zeta_{\downarrow})/\sqrt{2}$ is free [except for the constraint of Eq. (8)]. $\mathcal{H}'$ gives

corrections to physical properties which are at most of order $\omega \ln\omega + O((\omega \ln\omega)^2)$, as checked *a posteriori*.

We now derive the physical properties of $\mathcal{H}_0$, taking exactly into account the constraint of single occupancy at the impurity site and then consider the perturbative corrections due to $\mathcal{H}'$. The potential part of $\mathcal{H}$ is now written in terms of α and β:

$$\mathcal{V} = (\varepsilon + J)(\alpha^\dagger \alpha - \tfrac{1}{2}) + (\varepsilon - J)\beta^\dagger \beta + t(s^\dagger \alpha^\dagger + \alpha s) . \quad (14)$$

We also allow $\varepsilon = \varepsilon_0 + \lambda(T)$ in (14) to depend on temperature, with $\lambda(T) \to 0$ as $T \to 0$. This is justified below. In (14), we have dropped η, ensuring the constraint in the form $n_\alpha + \eta_\beta \leq 1$. The matrix elements of $\mathcal{H}$ are of course identical whatever form is chosen for the constraint in (8), as can be readily checked. The Hamiltonian (14) conserves n_β. In the subspace $n_\beta = 0$, the constraint is satisfied with an unconstrained α. So, the Hamiltonian is a simple quadratic one in s and α. In the subspace $n_\beta = 1$, $n_\alpha = 0$ and the Hamiltonian is diagonal. Physical quantities can therefore be computed by taking the trace on both $n_\beta = 0,1$ subspaces. One then has $\langle n_\beta \rangle = Z^{-1} \exp(-\beta F_1)$ with $Z = \exp(-\beta F_0) + \exp(-\beta F_1)$, where $F_{0,1}$ are the free energies associated with the Hamiltonian in subspaces $n_\beta = 0,1$ minus the free energy of the free conduction electrons. At $T = 0$, one finds

$$F_1 = (\varepsilon_0 - 3J)/2 ,$$

$$F_0 \approx -\frac{|\varepsilon_0 + J|}{2} - \frac{\Gamma}{2\pi} \ln\left(1 + \frac{W^2}{\Gamma^2 + (\varepsilon_0 + J)^2}\right) \quad (15)$$

for a square density of states (DOS), where W is the bandwidth and $\Gamma \sim \pi \rho t^2$ is the α-level width.

If the system is exponentially frozen in either one of the subspaces $n_\beta = 0,1$, one obtains two different Fermi-liquid states at low temperature. To reach the critical point, one must allow fluctuations between them. This requires that the ground state energies in the two subspaces are identical, i.e., $F_0 = F_1$, which always has a solution provided $J > (\Gamma/4\pi) \ln(1 + W^2/\Gamma^2)$ is antiferromagnetic. We note in passing that in the present problem, the sign of t is arbitrary, but not that of J in the (x,y) plane, due to the presence of t. The above condition does not determine $\langle n_\beta \rangle(T)$. In fact, the critical point occurs at the mixed-valence point, where the states $n_\zeta = 0$ and $n_\zeta = 1$ are degenerate and have equal weight, so that $\langle n_\zeta \rangle = \langle n_\alpha + n_\beta \rangle = \frac{1}{2} + O(\lambda, T)$. $\langle n_\alpha \rangle$ is calculated from the α Green's function, which is easily shown to be

$$G_\alpha(\omega) = G_\alpha^{(0)}(\omega)(1 - \langle n_\beta \rangle) = \frac{1 - \langle n_\beta \rangle}{i\omega + i\Gamma \,\mathrm{sgn}(\omega) - \lambda} , \quad (16)$$

where in (16), $G_\alpha^{(0)}(\tau)$ is the Green's function of α without the constraint, and ω is a Matsubara frequency. The conditions $F_0 = F_1$ and $\langle n_\zeta \rangle = \frac{1}{2}$ are simultaneously satisfied at $\varepsilon_0 = -J$, and yield $\lambda(T) \approx \Gamma \exp[-\beta\lambda(T)/2] \approx 2T \ln(\Gamma/T)$, as well as the relation $J = (\Gamma/4\pi) \ln(1 + W^2/\Gamma^2)$. In the limit $T \to 0$, we then have $\langle n_\alpha \rangle \to \frac{1}{2}$ and $\langle n_\beta \rangle \sim (T/\Gamma) \ln(\Gamma/T)$. The temperature dependences of λ and $\langle n_\beta \rangle$ are associated with a frequency-dependent phase shift of the conduction electrons at the critical point, $\delta(\omega) \sim (\omega/\Gamma) \ln(\Gamma/\omega)$, unlike the analytic phase shift for a Fermi liquid. This arises from $\mathcal{H}'$ as seen below. Note that the treatment of the constraint in mean field at this level or in the original Hamiltonian (9) gives rise to a similar behavior for n_β and λ [7]. In this approach, λ plays the role of a Lagrange multiplier enforcing the constraint.

In the present impurity problem, the critical point is reached only by fine-tuning parameters, as was already encountered for instance in the case of the two-Kondo impurity problem [8,9]. If the MFL singularity survives in the lattice problem, one can expect that the singularity in the free energy at the mixed-valence point would pin the chemical potential near it for a finite range of the parameters [3].

The free energy can be computed exactly and has the form $-T \ln\{\exp(-\beta F_\alpha) + \exp[-\beta(F_{\mathrm{KIN}} + \lambda/2)]\}$ up to a constant term, where F_α is the free energy of the quadratic Hamiltonian involving α and s. The impurity charge and longitudinal spin susceptibilities $\chi_{\sigma,\rho}$ can now be calculated at the mixed-valence critical point, using the relations $\sigma_z = (\alpha^\dagger \beta + \beta^\dagger \alpha)$ and $\rho = (\alpha^\dagger \alpha + \beta^\dagger \beta - \frac{1}{2})$ and

$$\chi_m(T) = \int_0^\beta \langle T_\tau m(\tau) m(0) \rangle d\tau , \quad m = \sigma_z, \rho . \quad (17)$$

The charge susceptibility can be also computed from the free energy, but the Kubo formula turns out to be simpler to deal with. For this, one uses $G_\alpha(\tau)$ of Eq. (16), and the fact that n_β commutes with $\mathcal{H}_0$. At zero frequency, the singular part of χ_ρ is proportional to $\beta n_\beta (1 - n_\beta) \sim \Gamma^{-1} \ln(\Gamma/T)$, i.e., a MFL-like susceptibility. The spin susceptibility is more delicate to compute. In the associated Kubo formula, it is not possible to use the Wick's theorem even though the Green's function of α is known exactly. However, noting that the α particles are hybridized to the conduction electrons, χ_σ is exactly related to the absorption spectra in an x-ray problem with a Lorentzian DOS [10] in the conduction band and a scattering term $U' n_\alpha n_\beta$, with an infinite U'. This term enforces the constraint between n_α and n_β. An antibound state is created at a large energy of order U', associated with a phase shift π. Physically, at the secondary threshold [11] at the chemical potential, the exponent is expected to be 0. In that case, we find that in terms of the time variable, and for large τ (but $\tau < \beta/2$), $\langle T_\tau m(\tau) m(0) \rangle \sim 1/\tau$, with an essentially symmetric behavior with respect to $\beta/2$. After integrating over τ, one then finds that χ_σ is proportional to $\Gamma^{-1} \ln(\Gamma/T)$, and thus behaves similarly to χ_ρ. The same calculation yields $\langle T_\tau \beta^\dagger(\tau) \beta(0) \rangle \sim 1/\tau$.

We now study the effect of the perturbation $\mathcal{H}'$. One can now justify the introduction of $\lambda(T)$ by the fact that we indeed find that the correction to the real part of the local impurity self-energy is to the leading order of the form $\omega \ln\omega$, and that the frequency dependence of $\chi_{\rho,\sigma}$ is

of the MFL form. Finally, this also results in a $T^2\ln(T)$ singularity in the free energy, or a singular specific heat $C(T)/T \sim \ln(\Gamma/T)$, which diverges in the same way as already found in the two-channel or two-impurity Kondo problems, for which the susceptibilities also present a MFL form [8,9].

Next consider the conduction electron self-energy. The fixed point Hamiltonian alone yields that the Green's function of h_σ is of the free-fermion form. A perturbative calculation in $\mathcal{H}'$ also yields a correction to the $\omega=0$ self-energy

$$\Sigma_h(\omega) \approx (\lambda_\rho+\lambda_s)[\omega\ln\omega + i\omega\,\mathrm{sgn}(\omega)] + O((\omega\ln\omega)^2) \tag{18}$$

with λ_ρ and λ_s of order 1. This is in contrast to the typical $\omega^{1/2}$ self-energy found at the critical points of other impurity problems [8]. The fact that the successive terms are not higher powers of $\ln\omega$ times ω as in the x-ray problem is due to the constraint and the related fact that the β level does not sit at $\omega=0$. Similarly, the effective screening electrons yield a self-energy which is constant due to the hybridizing term, plus a term of the same form as (18). It is reasonable to expect that the individual screening channels of the starting Hamiltonian (2) also have a $\omega\ln\omega$ self-energy, even if it seems hard to invert the transformation of Eqs. (4)–(6). This self-energy calculation also justifies the treatment of $\mathcal{H}'$ perturbatively. The pairing susceptibility $\langle T_\tau h_\sigma^\dagger h_{-\sigma}^\dagger(\tau) h_{-\sigma} h_\sigma(0)\rangle_\omega$ can also be perturbatively calculated and has a $\ln(\omega,T)$ divergence.

Next, we very briefly consider the problem of two identical Cu impurities in the metal at sites a and b. Quite generally, we can expand in appropriate local symmetry channels about a and b to get a Hamiltonian which also includes some transfer terms:

$$\mathcal{H} = \mathcal{H}_a + \mathcal{H}_b + \sum_{n,\sigma} t_{n,h} h_{n\sigma}^{a\dagger} h_{n\sigma}^{b} + \sum_n t_{n,s} s_n^{a\dagger} s_n^b + \text{H.c.}, \tag{19}$$

where $\mathcal{H}_{a,b}$ are of the form given by Eq. (3), with all operators having the extra label a or b. To keep the model tractable, we only consider the case of a single screening channel. In a model with more channels and spin in the screening channels, one must also consider a mixing term between the hybridizing channel at one site and the screening channels at the other, which prevent us from cleanly doing the transformations (4)–(6). In a simplified model with a single screening channel, the bosonization can be performed using the same steps as for the one-impurity case, without affecting the transfer part. The conclusions of a similar study as above are that the transfer Hamiltonian does not destroy the critical point, now characterized by the mixed-valence condition $J \sim F_0 - 2F_1 > 0$ where $F_{0,1}$ are the ground state energies in the $n_\beta = n_{\beta_a} + n_{\beta_b} = 0,1$ subspaces. The principal reason is that, apart from the constraints, $\beta_{a,b}$ remain decoupled from the other operators and from each other. We find that the local as well as the cross susceptibilities are of the MFL form. In particular, the cross pairing susceptibility, reflecting the phase coherence of pairs between the two sites is also logarithmically singular.

It is interesting to note here that a mean-field treatment [7] of the constraint (8) on the (prebosonized) Hamiltonian (9) yields essentially the same answers as obtained here and earlier [3].

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