Kondo Effect in High-T_c Cuprates

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We study the Kondo effect due to the nonmagnetic impurity, e.g., Zn, in high-T_c cuprates based on the spin-charge separated state. In the optimal or overdoped case with the Kondo screening, the resistivity is given by $\rho(T) = \frac{4\hbar}{e^2}\frac{n_{\rm imp}}{1-x} + \frac{\alpha T}{x}$ (x: hole concentration, $n_{\rm imp}$: impurity concentration, α: constant), which is in agreement with experiments. In the underdoped region with the pseudospin gap, an SU(2) formulation predicts that the holon phase shift is related to the formation of the local spin moment, and hence the residual resistivity is given by $\rho_{\rm res} = \frac{4\hbar}{e^2}\frac{n_{\rm imp}}{x}$, which is also consistent with the experiments. The magnetic impurity case, e.g., Ni, is also discussed. [S0031-9007(97)04488-8]

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A Kondo effect is a phenomenon shown by a magnetic impurity put into a metal [1]. High-T_c cuprates offer a unique opportunity to study the Kondo effect in a strongly correlated metal. In the undoped high-T_c cuprates, the valency of a Cu atom is Cu^{2+} (d^9) and the system is a Mott insulator. By the hole doping, the system becomes metallic and shows superconductivity with high T_c. We believe that the Kondo effect in this system is actually observed for the *nonmagnetic* impurity, e.g., Zn, replacing the Cu atom in the conducting plane. The valency of Zn is Zn^{2+} (d^{10}) and compared with the Cu^{2+} case, one electron is trapped by one additional positive charge of the nucleus, which forms a singlet on the Zn site. In the underdoped cuprates with spin gap, it is found experimentally that a local moment of $S = 1/2$ appears on neighboring Cu sites [2–8]. We believe this localized spin is not screened by the conduction electron spins because of the reduced density of states for spins at the Fermi energy E_F in the presence of the spin gap [9,10]. Once the spin gap collapses with the increased hole concentration, the density of states for spins at E_F recovers and also the Kondo screening, i.e., the singlet formation between localized spin and conduction spins, occurs. Associated with the formation of local moments, it is found that the residual resistivity $\rho_{\rm res}$ is very large in high-T_c cuprates. For example, $\rho_{\rm res}$ at 1% Zn doping in $La_{2-x}Sr_xCuO_4$ ($x = 0.15$) amounts to $\sim 100\ \mu\Omega\,{\rm cm}/\%$. This value should be compared with $\rho_{\rm res} = 0.32\ \mu\Omega\,{\rm cm}/\%$ for Zn doping in the Cu metal [11].

The single band t-J model is believed to be the low energy effective model of high-T_c cuprates [12], and the s-wave ($\ell = 0$) scattering is expected to dominate. Then the Friedel sum rule [13] relates the phase shift δ_σ for the s-wave scattering to the difference of the valence Z between the host and impurity atoms and the spin of the impurity S as

$$Z = \frac{1}{\pi}(\delta_\uparrow + \delta_\downarrow), \tag{1}$$

$$2S = \frac{1}{\pi}(\delta_\uparrow - \delta_\downarrow). \tag{2}$$

The residual resistivity in the limit of small $n_{\rm imp}$ is given in two dimensions as

$$\rho_{\rm res} = \frac{2\hbar}{e^2}\frac{n_{\rm imp}}{n}(\sin^2\delta_\uparrow + \sin^2\delta_\downarrow), \tag{3}$$

where $n_{\rm imp}$ is the impurity concentration and n is the carrier concentration [6,11]. Theoretically, it is not a trivial problem whether the carriers are the electrons with the concentration $n = 1 - x$ or the doped holes with $n = x$. The experimentally observed values of $\rho_{\rm res}$ in the underdoped region are quantitatively fitted by Eq. (3) by putting the carrier concentration $n = x$, and the phase shift $\delta_\sigma = \pi/2$ (unitarity limit) [6–8]. Note that this is the largest value theoretically expected from Eq. (3). As the doping proceeds to the optimal and overdoped regions, the residual resistivity decreases and fits the formula Eq. (3) with n being increased to $1 - x$ with $\delta_\sigma = \pi/2$ unchanged [8]. This crossover seems to correspond to the disappearance of the local moment. It is noted that $\rho_{\rm res}$ in the optimal and overdoped regions is consistent with the Fermi liquid picture, where $S = 0$ and $Z = 1$ in Eqs. (1)–(3). However, considering the fact that the conductivity without the impurities is proportional to x and hence is dominated by the hole carriers in the optimal doping region, the residual resistivity corresponding to $n = 1 - x$ is a mystery. Even more unconventional is the underdoped case, where the experimental observation that $n \cong x$ in Eq. (3) means that the phase shift cannot be associated with the electrons. Can one associate the phase shift with the holes? Then what determines that phase shift? These are the questions to which we give solutions below.

As mentioned earlier, the interpretation of carrier concentration as x or $1 - x$ is a complicated issue in high-T_c cuprates. It is commonly agreed that, in optimally doped samples, a large Fermi surface of area $1 - x$ exists. Even

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in underdoped samples there is evidence that the pseudogap is formed out of an underlying large Fermi surface [14]. Nevertheless, the dc conductivity is consistent with x carriers with a scattering rate of $2kT$. This is supported by the analysis of the Drude part ($\omega < 2kT$) of the optical conductivity [15,16] and by the fact that the superfluid density scales with x. The optical conductivity for higher frequency is not Drude-like, and has been fitted by $\omega^{-1+\alpha}$ [17]. The spectral weight when integrated over a large frequency range of order 1 eV becomes insensitive to x. These observations indicate that the carriers are not Fermi liquid, as emphasized by Anderson [18]. The dichotomy between a large Fermi surface of area $1 - x$ and a low energy spectral weight of x is readily accounted for in the resonating valence bond (RVB) theory, using the concept of spin-charge separation [19]. In this view the dc conductivity is dominated by the holons. The formal implementation of these ideas is the slave boson mean field theory and gauge fluctuations [20–22]. Even though Anderson and co-workers have abandoned this approach in favor of the tomographic Luttinger liquid [18], we believe that this line of investigation still has merits, especially in view of recent refinements [23], and we shall base our discussion on this method. Let us first consider the U(1) theory which is applicable to optimally doped and overdoped regions. In this formalism the electron ($C^\dagger_{i\sigma}$) is described as the composite particle of spinon ($f^\dagger_{i\sigma}$) and holon (b_i), i.e., $C^\dagger_{i\sigma} = f^\dagger_{i\sigma} b_i$ with the constraint

$$\sum_\sigma f^\dagger_{i\sigma} f_{i\sigma} + b^\dagger_i b_i = 1 . \tag{4}$$

This constraint is taken care of by introducing the gauge potential a_0 which couples to the left-hand side of Eq. (4). We recall that the resistivity is given by the Ioffe-Larkin composition rule $\rho = \rho^{\text{spinon}} + \rho^{\text{holon}}$ [20]. For the clean case it is dominated by ρ^{holon} which is inversely proportional to x. The residual resistivity ρ_{res} is given as

$$\begin{aligned}\rho_{\text{res}} &= \rho^{\text{spinon}}_{\text{res}} + \rho^{\text{holon}}_{\text{res}} \\ &= \frac{4\hbar}{e^2} n_{\text{imp}} \left[\frac{\sin^2 \delta^{\text{spinon}}}{1 - x} + \frac{\sin^2 \delta^{\text{holon}}}{x} \right]. \end{aligned} \tag{5}$$

Now we calculate the phase shift for spinons and holons. Our discussion is based on the mean field approximation. In the presence of a Zn the static configuration of a_0, together with the Coulomb potential A_0, is determined self-consistently to satisfy both the charge neutrality condition and Eq. (4) on average for each site i, except the Zn site. The spinons and holons feel the potential $a_0 + A_0$ and a_0, respectively, and each particle has its one-particle states which are characterized by their phase shifts. We can relate the phase shifts to the change $\Delta N_{\text{spinon}}, \Delta N_{\text{holon}}$ in the number of particles inside a large sphere including Zn by applying the Friedel sum rule to the spinons and holons separately. We shall see that $\Delta N_{\text{spinon}}, \Delta N_{\text{holon}}$ are determined by the charge neutrality condition and the constraint Eq. (4). The fluctuating part of the gauge field gives an inelastic contribution to the resistivity, which is simply added to the residual one which we are now interested in.

In order to determine ΔN_{spinon} and ΔN_{holon}, we first write down a specific model for the Zn impurity. Relative to Cu(d^9), it has an additional nuclear charge and an additional electron, making it charge neutral and a spin singlet. Thus we can simply remove it from the consideration and treat it as a vacancy in the Cu(d^9) lattice. We model this vacancy by introducing a strong local repulsive potential for spinons and holons, i.e., $H_{\text{Zn}} = V_0(\sum_\sigma f^\dagger_{0\sigma} f_{0\sigma} + b^\dagger_0 b_0)$ with the limit $V_0 \to \infty$. Note that on the Zn site $i = 0$, the right-hand side of Eq. (4) is zero. The introduction of Zn creates a violation of the constraint on a single site and we immediately find that, within a large sphere,

$$\Delta N_{\text{holon}} + \Delta N_{\text{spinon}} = -1 . \tag{6}$$

Next we observe that the number of holes within a large sphere containing N sites is fixed by charge neutrality, i.e., it is fixed to be Nx by the background charge introduced by doping which lies off the Cu(d^9) plane. The introduction of the Zn impurity only excludes the hole from the Zn site but cannot change the total hole number within the large sphere. We then conclude that $\Delta N_{\text{holon}} = 0$ and, using Eq. (6), $\Delta N_{\text{spinon}} = -1$. For δ^{spinon} we can apply Eq. (1) with $Z = \Delta N_{\text{spinon}} = -1$. In the optimally doped and overdoped cases, no local moment appears so that the impurity is nonmagnetic, and we put $S = 0$ in Eq. (2). This gives $\delta^{\text{spinon}} = \delta_0 = \pi/2$ as the nonmagnetic potential scatterer in a Fermi liquid, and $\rho^{\text{spinon}}_{\text{res}} = \frac{4\hbar}{e^2} \frac{n_{\text{imp}}}{1-x}$. For holons, on the other hand, δ^{holon} cannot be replaced by the value at a representative energy such as the Fermi energy. However, the holons are hard-core bosons interacting strongly with the gauge field, which might behave like fermions. This gives $\delta^{\text{holon}} = \pi \Delta N_{\text{holon}} = 0$ and $\rho^{\text{holon}}_{\text{res}} = 0$. In summary, the residual resistivity ρ_{res} is given by

$$\rho_{\text{res}} = \rho^{\text{spinon}}_{\text{res}} = \frac{4\hbar}{e^2} \frac{n_{\text{imp}}}{1 - x} , \tag{7}$$

in agreement with the experiments. As noted before, the appearance of $1 - x$ in ρ_{res} when x appears in the T-dependent part of ρ is highly nontrivial and may be regarded as an important test of the Ioffe-Larkin rule.

Next, we discuss the underdoped regime. From our discussion up to now, it is very difficult to explain the experimental observation. Since the resistivity is proportional to x, we need the holon scattering to be unitary. However, since there is no spin label on the holon, the natural values for δ^{holon} is 0 or π, and in either case $\rho^{\text{holon}}_{\text{res}}$ is zero. We also observe that unlike the overdoped case, where the Kondo scattering is smoothly connected to the strongly overdoped Fermi liquid limit, in the underdoped case we cannot recover the experimental value even if we extrapolate to the zero doping limit, i.e., Néel state. In this case, the unit cell is doubled, and the doped holes form two nonequivalent Fermi pockets near $(\pi/2, \pm\pi/2)$. Let

us assume that a local moment is formed, an assumption which is by no means obvious. By extending Eqs. (1) and (2) to include two pockets and setting $Z = 1$, $S = 1/2$, we find $\delta_\uparrow = \pi/2$ and $\delta_\downarrow = 0$. From Eq. (3) we find $\rho_{\rm res} = \frac{2\hbar}{e^2}\frac{n_{\rm imp}}{x}$, which is still small compared with the experiment by a factor of 2. On the other hand, if no local moment is formed, we find $\delta_\uparrow = \delta_\downarrow = \pi/4$ and $\rho_{\rm res}$ is even smaller. Thus it is apparent that the experimental observation is highly nontrivial to explain.

Recently, it was pointed out that the traditional formulation of the t-J model [which we shall call the U(1) formulation] is inadequate for small doping, because it does not include low lying excitations connected to the SU(2) symmetry which is known to exist exactly at half filling [22]. A new formulation was introduced which includes these fluctuations, and it is believed to be a better starting point for the underdoped region [23]. A feature of the SU(2) formulation is that two bosons (b_1, b_2) which form a SU(2) doublet is introduced. Instead of Eq. (5), the constraint is given by

$$\sum_\sigma f_{i\sigma}^\dagger f_{i\sigma} + b_{1i}^\dagger b_{1i} - b_{2i}^\dagger b_{2i} = 1\,, \tag{8}$$

and the number of vacancy is given by $b_{1i}^\dagger b_{1i} + b_{2i}^\dagger b_{2i}$. We recover the U(1) formulation if the boson isospin doublet (b_{1i}, b_{2i}) is polarized in the z direction, giving $(b_i, 0)$. In contrast, in the SU(2) mean field theory [23], the underdoped normal state is represented by the staggered flux phase, where the constraint is satisfied by $\langle b_1^\dagger b_1\rangle = \langle b_2^\dagger b_2\rangle$, i.e., the isospin is strongly fluctuating. We shall argue that the experiment may be explained using this new formulation. As in the U(1) case, we treat the Zn site as a vacancy by $H_{\rm Zn} = V_0(\sum_\sigma f_{0\sigma}^\dagger f_{0\sigma} + \sum_{\alpha=1,2} b_{\alpha 0}^\dagger b_{\alpha 0})$ with $V_0 \to \infty$. Now we treat all the Cu sites using the SU(2) formulation. Then the discussion on the charge neutrality applies similarly to $\Delta N_{\rm holon}$ as before, and $\Delta N_{\rm holon} = \Delta\sum_i (b_{1i}^\dagger b_{1i} + b_{2i}^\dagger b_{2i}) = 0$. On the other hand, $\Delta N_{\rm spinon} = \Delta(\sum_{i:\text{in the sphere}} f_{i\sigma}^\dagger f_{i\sigma})$ is no longer related to the charge. We then argue that the potential will form a spinon bound state, which is responsible for the local moment. This has been shown to be the case in the spin gap phase if the spinon spectrum has point nodes and a linear density of states [9,10]. Within a large sphere, the formation of the local spin means that, excluding the Zn site, the number of spinons has increased by one. This compensates the spinon repelled from the Zn site so that $\Delta N_{\rm spinon} = 0$. We also note that, in analogy with Eq. (6), $\Delta N_{\rm spinon} + \Delta\sum_i (b_{1i}^\dagger b_{1i} - b_{2i}^\dagger b_{2i}) = -1$. Thus we conclude that $\Delta\sum_i (b_{1i}^\dagger b_{1i} - b_{2i}^\dagger b_{2i}) = -1$. From these considerations, we conclude that

$$\Delta\sum_i (b_{1i}^\dagger b_{1i}) = -\Delta\sum_i (b_{2i}^\dagger b_{2i}) = -\frac{1}{2}\,. \tag{9}$$

In terms of phase shift, we have $\delta_{b_2} = -\delta_{b_1} = \pi/2$, leading to a residual resistivity of

$$\rho_{\rm res} = \frac{4\hbar}{e^2}\frac{n_{\rm imp}}{x}\,, \tag{10}$$

in agreement with experiment.

We note that our argument so far applies to any divalent nonmagnetic impurity, such as Zn, Mg. Let us now consider a magnetic impurity such as Ni. In this case, the Ni is in a d^8 configuration with $S = 1$. In the underdoped case, the argument proceeds as before, except that the additional $S = 1/2$ on the bound state will have strong antiferromagnetic exchange with the $S = 1$, leading to an $S = 1/2$ local moment. The boson counting is the same as before, and we predict Eq. (10) to hold. This is, in fact, the experimental situation [24,25]. In the optimal or overdoped case, we believe the $S = 1$ moment will be partially screened, so that an $S = 1/2$ local moment remains. This can be viewed also as a ferromagnetic Kondo problem, as shown by Khaliullin *et al.* [10]. In this case we find $\delta_\uparrow = \pi, \delta_\downarrow = 0$ or vice versa, and the spinon scattering should be very weak. Indeed, experimental $\rho_{\rm res}$ is much smaller that the Zn doped case in the optimally doped or overdoped case [25].

Finally we discuss the case of a trivalent impurity such as Al. An extra mobile electron is donated to the layer, leaving a positive charge on the impurity site relative to the Cu(d^9). In the U(1) formulation we find $\Delta N_{\rm holon} = -1$ and $\Delta N_{\rm spinon} = 0$. Thus we expect the scattering of the spinon to be nonunitary in general. In the underdoped case, we expect the formation of the $S = 1/2$ local moment. In the SU(2) formulation we find $\Delta N_{\rm spinon} = 0$ and $\Delta\sum_i (b_{1i}^\dagger b_{1i} - b_{2i}^\dagger b_{2i}) = -1$ by the constraint. Combined with $\Delta\sum_i (b_{1i}^\dagger b_{1i} + b_{2i}^\dagger b_{2i}) = -1$, we conclude that the holon scattering is also nonunitary.

To complete the discussion, we have to argue that it is plausible to assign a phase shift to the b_1 and b_2 bosons as if they were fermions. In the SU(2) formulation, the effective Lagrangian describing the holons $h_i = [b_{1i}, b_{2i}]$ in the underdoped spin gap region is given by [23]

$$L = \int dr h^\dagger(r,\tau)\left[\partial_\tau + ia_0^3\tau_3 + iA_0 + \frac{1}{2m}(-i\nabla + \vec{a}^3\tau_3 + \vec{A})^2 - \mu\right]h(r,\tau) + \sum_{q,\omega} a_\mu^3(q,\omega)\Pi_{S\mu\nu}(q,\omega)a_\nu^3(-q,-\omega)\,, \tag{11}$$

where the spinons have been integrated over to give the polarization function Π_S in the effective action for the gauge field. Because the gauge symmetry is broken from SU(2) to U(1) in the staggered flux state, only the a^3 gauge field remains massless. Note also that the Ioffe-Larkin composition rule no longer applies because the

external vector potential A_μ is coupled to h_i with the unit matrix and not with τ_3. The conductivity is then totally determined by that of the holon system. Here we have the two problems, i.e., the strong gauge field fluctuation and the hard-core condition for the holons. We believe that these two are resolved simultaneously by introducing the statistical transmutation of b_1,b_2 to fermions. This is accomplished by introducing the Chern-Simons gauge field a' coupled to b_1,b_2 with the $(+)$ and $(-)$ gauge charges, respectively [26],

$$L = \int dr h^\dagger(r,\tau)\Big[\partial_\tau + i(a_0^3 + a_0')\tau_3 + iA_0 + \frac{1}{2m}(-i\nabla + (\vec{a}^3 + \vec{a}')\tau_3 + \vec{A})^2 - \mu\Big]h(r,\tau) + a^3\Pi_S a^3 + (a^3 + a')\Pi_H(a^3 + a') + a'\Pi_{\rm CS}a'. \tag{12}$$

In the Coulomb gauge, the gauge field has two components as a_0 and $a_1 = a_{\rm transverse}$. In this representation, $\Pi_{\rm CS}$ is given by $(\Pi_{\rm CS})_{01} = (\Pi_{\rm CS})_{10} = cq = \frac{q}{2\theta}$, with the diagonal components being zero. Here θ is the statistical angle, and the bosons are transformed into fermions when $\theta = (2m + 1)\pi$ (m: an integer). We take this choice because the hard-core condition is automatically taken into account even for the noninteracting fermions. Because of the opposite charges of b_1 and b_2, the system remains gauge neutral and also the time reversal symmetry is preserved at the mean field level. Now the holon h_i is coupled to $(a^3 + a')\tau_3$, and we obtain the effective action for $a = a^3 + a'$ by integrating over $a^3 - a'$. Then the gauge flux fluctuation $D = \langle(\nabla \times a)\cdot(\nabla \times a)\rangle$ is given by $D = \frac{1}{\Pi_{H1}+\Pi_{S1}} f$ with the factor f being

$$f = \frac{c^2q^2(\Pi_{H0} + \Pi_{S0}) - \Pi_{H0}\Pi_{S0}\Pi_{S1}}{c^2q^2(\Pi_{H0} + \Pi_{S0}) - \Pi_{H0}\Pi_{H1}\Pi_{S0}\Pi_{S1}/(\Pi_{H0} + \Pi_{S0})}. \tag{13}$$

$\Pi_{\alpha 0}$ and $\Pi_{\alpha 1}$ are the diagonal longitudinal and transverse components of the spinon ($\alpha = S$) and holon ($\alpha = H$) polarization function. It is easy to see that $f < 1$, and this factor represents the reduction of the gauge field fluctuation. The physical picture is that a large part of the original gauge field is canceled by the Chern-Simons gauge field attached to the fermions by an appropriate choice of the integer m. Then it is expected that the hard-core potential and the strong gauge field fluctuations are taken into account in terms of the two-component free fermion theory, and this may justify the phase shift argument given above.

In summary, we have analyzed the Kondo effect in high-T_c cuprates based on the spin-change separated state. The change of the phase shifts $\delta^{\rm holon}$ and $\delta^{\rm spinon}$ for holons and spinons due to the Kondo screening, together with the crossover from SU(2) to U(1) theory, explains the change of the residual resistivity from $\rho_{\rm res} = \frac{4\hbar}{e^2}\frac{n_{\rm imp}}{x}$ to $\rho_{\rm res} = \frac{4\hbar}{e^2}\frac{n_{\rm imp}}{1-x}$ as the hole concentration increases and the local moment disappears. Lastly, we comment on the bipolaronic model for the underdoped cuprates. In this model the charge of the carrier is $2e$ and $n = x/2$ in the underdoped region. This gives $\rho_{\rm res} = \frac{2\hbar}{e^2}\frac{n_{\rm imp}}{x}$, which is half of that expected above. Then the experiments support the existence of the carrier not with change $2e$ but with e.

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