Fermi Liquid - Non-Fermi Liquid Transition in the Double Exchange Model

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Abstract

An approximate solution to the double-exchange model is presented. The forced alignment of the conduction-electron spin with the core spin that causes ferromagnetism also removes a large part of the Hilbert space, needed for coherent propagation of electrons carrying spin and charge. As a result, the electron becomes a composite particle and its Green’s function exhibits a two-fluid character: a coherent Fermi-liquid component associated with the ferromagnetically ordered core spins and non-Fermi liquid component associated with the disordered spins. With increasing temperature there is a continuous transfer of spectral weight from the former to the latter until the Fermi liquid component disappears above the ferromagnetic $T_c$. Implications for manganites, which exhibit very large magnetoresistance, are discussed.

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Recent discovery of very large magnetoresistance in $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$ and other manganites \cite{1, 2, 3} has revived interest in the double-exchange model \cite{4}. Approximately in the range $0.2 < x < 0.4$, the low-temperature state is a ferromagnetic metal which undergoes a transition to a paramagnetic state above a critical temperature $T_c$. The important effects originate from the d orbitals of the manganese ions. The $E_g$ orbitals form a conduction band containing $1 - x$ electrons per site. The electron strongly interacts ferromagnetically via the Hund’s rule mechanism with the $S = 3/2$ core spin formed from the three-fold degenerate $T_g$ orbitals. As it hops from one atom to the next, the spin of a conduction electron must remain parallel to the direction of the core spins. The latter will then tend to line up to facilitate coherent propagation, giving rise to metallic ferromagnetism \cite{4, 5}. What is new and surprising is that, associated with the magnetic transition, there appears to be a metal-insulator transition in the manganites and, that it is the high-temperature phase that is insulating. The large increase in resistivity at $T_c$ and the enormous magnetoresistance are clearly linked to this transition, imply extreme sensitivity to external fields. The aim of this work is to see whether such a sensitivity is intrinsic to the double-exchange model.

We consider the following Hamiltonian

$$H = -\sum_{ij\sigma} t_{ij} c_{i\sigma}^{\dagger} c_{j\sigma} - J_H \sum_i \vec{S}_i \cdot \vec{s}_i.$$  

(1)

Here $c_{i\sigma}$ destroys an $E_g$ electron at site $i$ and the first term is then the usual nearest-neighbor hopping Hamiltonian with $t_{ij} = t$. The second term describes the Hund’s rule coupling between the core spin $\vec{S}_i$ and the conduction electron spin density $\vec{s}_i$. We will focus on the $J_H \to \infty$ limit. The nominal two-fold degeneracy of the $E_g$ orbitals could in fact be lifted by a coupling to the lattice. For the most part we ignore the degeneracy, its effect would be discussed later.

The key point is that the very cause of ferromagnetism, namely, that the conduction-electron spin is forced to follow the core-spin direction, removes a large portion of the Hilbert space needed for coherent propagation of electrons carrying spin and charge. Consider the on-site problem, with a classical core spin $\vec{S}_i$. There are two single-electron states with electron spin parallel and antiparallel to $\vec{S}_i$ and energies $\frac{1}{2}J_H S$, respectively. As $J_H \to \infty$,
both the doubly occupied and the antiparallel singly occupied states are projected out. In
the quantum mechanical case, an electron combines with the core spin to form two manifolds
of total spin $S \pm \frac{1}{2}$ with energies $\pm \frac{1}{2} J_H S$ and $\frac{1}{2} J_H (S + 1)$, respectively. Again the $S - \frac{1}{2}$ and
the doubly occupied sectors are projected out. This restriction is more stringent than that
in the infinite-$U$ Hubbard model where only double occupancy is forbidden, and dominates
the physics.

We present approximate solutions that incorporate the projective physics both below and
above the transition by using a functional integral representation similar to the one used by
Millis et al. [6]. We find that the conduction-electron Green’s function is a sum of two
terms: a Fermi-liquid (coherent) component associated with the ferromagnetically ordered
core spin configuration, and a non-Fermi liquid (incoherent) component associated with the
disordered spin configurations. With increasing temperature ($T$), there is a continuous
transfer of spectral weight from the coherent to the incoherent component, as the Fermi
liquid component disappears completely above $T_c$. For $x < 0.5$, the theory yields reasonable
values for $T_c$ and a hole-like Fermi surface. It is also found that the approximate particle-hole
symmetry close to the Fermi surface is violated.

Consider first the on-site problem with a classical $\vec{S}_i$, pointing along some arbitrary
direction. Let $f_i$ and $g_i$ be operators that destroy single electrons with spins parallel and
antiparallel to $\vec{S}_i$. The operators $c_{i\sigma}$ in Eq. (1) are quantized along the global z-axis and
can be represented as a linear combination: $c_{i\sigma} = \frac{1}{\sqrt{2S}} [b_{i\sigma} f_i + \text{sgn$(\sigma)$} b^\dagger_{i-\sigma} g_i]$, where $b_{i\sigma} = r_{i\sigma} e^{i\phi_{i\sigma}}$ are complex numbers which satisfy the normalization condition $\sum_{\sigma} b^\dagger_{i\sigma} b_{i\sigma} = 2S$. If
$\vec{S}_i$ is expressed as $(S, \theta, \phi)$ in spherical coordinates, then we can choose $r_{i\uparrow} = \sqrt{2S} \cos \frac{\theta}{2}$,
$r_{i\downarrow} = \sqrt{2S} \sin \frac{\theta}{2}$ and $\phi = \phi_\downarrow - \phi_\uparrow$. In the quantum case, $b_{i\sigma}$ become time-dependent Bose
fields, then it is convenient to use functional integral techniques. Note that the core spin
variables can be expressed as: $S^+_{i} = b^\dagger_{i\uparrow} b_{i\downarrow}$, $S^z_{i} = \frac{1}{2} (b^\dagger_{i\uparrow} b_{i\uparrow} - b^\dagger_{i\downarrow} b_{i\downarrow})$. This is nothing but the
exact Schwinger boson representation of the core spin [8, 9] – a fact that will be useful later.
As $J_H \rightarrow \infty$, we can ignore the upper (antiparallel) level (i.e. terms containing $g$) so that
the conduction-electron fields are exactly represented by
\[ c_{i\sigma} = \frac{1}{\sqrt{2S}} b_{i\sigma} f_i, \] (2)
where \( c_{i\sigma}, f \) are Grassman field. The partition function can be written as a functional integral with an action given by
\[ A = \int_0^\beta d\tau \left[ \sum_{i\sigma} \left( 1 + \frac{f_i^* f_i}{2S} b_{i\sigma}^* \frac{\partial b_{i\sigma}}{\partial \tau} + (f_i^* \frac{\partial f_i}{\partial \tau} + \mu f_i^* f_i) + \frac{1}{2S} \sum_{ij\sigma} t_{ij} b_{i\sigma}^* b_{j\sigma} f_i^* f_j \right) \right]. \] (3)
The integrals over the Bose fields are to be done subject to constraints \( \sum_{\sigma} b_{i\sigma}^* b_{i\sigma} = 2S \). The action is invariant under the gauge transformation: \( f_j \rightarrow f_j e^{i\psi_j}; \ b_{j\sigma} \rightarrow b_{j\sigma} e^{-i\psi_j} \). Hence at each site, one of the Bose fields, say \( b_{j\uparrow} \), can be chosen to be real. Then, written in terms of the polar variables \((S, \theta, \phi)\), the action becomes identical to the one derived by Millis et al. [6].

It is clear from Eqs. (2) and (3) that, as a consequence of projecting out the antiparallel state, the conduction electron, in essence, has become a composite object. Its charge is carried by a spinless fermion field \( f \), and its spin is wedded to the core spin and is carried by the Bose fields. Thus, the charge of the conduction electron is given by \( n_i = f_i^* f_i \) and its spin by \( \vec{s}_i = \frac{f_i^* f_i}{2S} \vec{S}_i \). The last term in the action describes hopping of a spinless charge characterized by a fluctuating hopping parameter \( t B_{ij}/S \), where \( B_{ij} = \frac{1}{2} \sum_{\sigma} b_{i\sigma}^* b_{i\sigma} \) essentially measures the nearest-neighbor ferromagnetic correlation and has a magnitude \( |B_{ij}| = S[\cos^2(\frac{\theta_i - \theta_j}{2}) - \sin \theta_i \sin \theta_j \cos^2(\frac{\phi_i - \phi_j}{2})] \). This has a maximum value \((= S)\) for ferromagnetic alignment: \( \theta_i = \theta_j; \phi_i = \phi_j \). Eqs. (2) and (3) are very similar with to the corresponding ones for the infinite-\( U \) Hubbard model (or, \( t - J \) model with \( J = 0 \)). Physics is quite different, however [9]. The charge and spin-fields in the present case are not connected by a projective constraint: “spinons” and “holons” can occupy the same site simultaneously.

The absence of such gauge fluctuations is probably the reason why a stable ferromagnetic state can exist in these systems for such large \( x \).

A Hartree-Fock decomposition of (3) yields effective actions that are quadratic in the charge and spin-fields: \( A_{MF} = A_f + A_b + \text{const} \). The factor \((1 + \frac{f_i^* f_i}{2S})\) can be absorbed by shifting \( \mu \) and rescaling the Bose fields so that they correspond to a spin of renormalized
magnitude: $S \rightarrow S_R = S \zeta = S + \frac{1-x}{2}$. This has a very simple meaning: the spin of a given site fluctuates between $S$ (no electron) and $S + \frac{1}{2}$ (one electron), the average value for $1-x$ electrons is $S_R$. The actions are then given by

$$A_b = \int_0^\beta d\tau \left[ \sum_{i \sigma} b_{i \sigma}^* \frac{\partial b_{i \sigma}}{\partial \tau} + \frac{D}{2S_R} \sum_{ij \sigma} t_{ij} b_{i \sigma}^* b_{j \sigma} \right] ,$$

(4)

$$A_f = \int_0^\beta d\tau \left[ \sum_i (f_i^* \frac{\partial f_i}{\partial \tau} - \mu f_i^* f_i) + \frac{B}{S_R} \sum_{ij \sigma} t_{ij} f_i^* f_j \right] ,$$

(5)

where $D = \langle f_i^* f_j \rangle$ is the average “kinetic energy” of charge and $B = < B_{ij} >$ is the average value of short-range ferromagnetic correlation. Thus, although the bare problem has a single energy scale (the bare bandwidth $W = 12t$), the propagation of charge and spin is governed by distinct energy scales as characterized by the hopping parameters $t_f = \frac{B}{S_R}$ and $t_b = \frac{BD}{2S_R}$, respectively. A determination of these energy scales is very important from an experimental standpoint since $W$ can be in the eV range, the experimental $T_c$ is only a few hundred degrees [1 – 3].

A number of results can be obtained independently of the constraints. Since fermion fields are not constrained, $A_f$ simply describes free spinless fermions characterized by a cosine band $\epsilon_f(k) = -2t_f \cos k_x + \cos k_y + \cos k_z$. The parameter $B$ is maximum ($= S_R$) in the ferromagnetic state and decreases with increasing $T$, but must remain finite at $T_c$ since it measures only short-range magnetic correlations. Hence, we come to the conclusions that (1) charge fermions remain “metallic” across the ferromagnetic transition and (2) the charge bandwidth, $W_f = \frac{B}{S_R} W$, equals $W$ at $T = 0$, but decreases with increasing $T$ as $B$ decreases. For fixed $S_R$, there is an obvious symmetry about $x = 0.5$ (quarter filling), so we can restrict our attention to $x < 0.5$. (3) The theory predicts a hole-like Fermi surface in this region. To determine $T_c$, we need to compute $D$ which determines the Bose parameter $t_b$. For $x$ not too large, we use a quadratic approximation for the hole spectrum to obtain

$$D = x - \frac{x(6\pi^2 x)^{2/3}}{10} \left\{ 1 + \frac{5\pi^2}{12} \left( \frac{kT}{t_f \epsilon_F} \right)^2 \right\} ,$$

where $\epsilon_F = (6\pi^2 x)^{2/3}$ is the hole Fermi energy for a band with $t_f = 1$. Thus $D$ is quite small, depends only on $x$ at $T = 0$. For $x = 0.2$, $D \sim 0.1$ at $T = 0$, and decreases with increasing
Therefore, $t_b/t_f = D/(2B) \sim D/(2S_R) << 1$. Hence, $T_c$ must be much smaller than the fermion bandwidth.

The Bose Hartree-Fock problem is identical to that for a spin-$S_R$ Heisenberg ferromagnet, but with a temperature dependent exchange constant $J = tD/(2BS_R)$. For the Heisenberg model, a qualitatively correct solution is obtained both above $\tilde{\omega}_k = \frac{D}{2S_R}$ the transition by imposing the constraint on the average via a Lagrange multiplier $\Lambda$. Then the bosons are free, with a spectrum $\omega_k = 6t_b\gamma_k$, with $\gamma_k = 1 - \frac{1}{3}(\cos k_x + \cos k_y + \cos k_z)$. Long-range magnetic order (along the x direction) appears through a Bose condensation in the $\tilde{k} = 0$ mode $[9]$. The magnetization is the same as the condensate density and is given by:

$$m = S_R - \int \frac{d^3k}{(2\pi)^3} \frac{1}{e^{\beta(\omega_k + \Lambda)} - 1}$$

To obtain $B$ we only need to replace 1 by $\gamma_k$ in the numerator. The equations have been discussed in detail in ref.$[9]$. Briefly, in the ordered regime, $\Lambda = 0$ and $m > 0$. The ground-state is ferromagnetic since at $T = 0$ both integrals vanish giving $m = S_R$ and $B = S_R$. By expanding the Bose spectrum we obtain the spin-wave theory results at low temperatures: $m = S_R - \text{const.} \bar{T}^3$ and $B = S_R - \text{const.} \bar{T}^2$. Above $T_c$, $m = 0$, and $\Lambda \propto \xi^{-2} > 0$, where $\bar{x}$ is the (correlation) length over which spins are ordered.

We have numerically and accurately solved the fermion-boson self-consistency problem (for the exact cosine spectra). Fig 1 shows the critical temperature, $2kT_c/W$, as a function of doping. Also shown are the chemical potential $2\mu_f/W$ and the charge fermion bandwidth $W_f/W = t_f/t$, evaluated at $T_c$. Two aspects need to be stressed. (1) $T_c$ is smaller than the fermion bandwidth by an order of magnitude, confirming the earlier statement that the charge degrees of freedom remain metallic across the ferromagnetic transition. (2) The magnitude of $T_c$ is quite reasonable. Thus for a bare bandwidth of 2 eV, $T_c$ ranges from 20 to 40 meV for $0.14 < x < 0.5$. Of course, fluctuations would bring down $T_c$ somewhat.

If we take a two-fold degenerate level, there are two spinless charge fermions at each site, otherwise the calculation is very similar. This leads to a larger $T_c$ since the parameter $D \sim (1 - x)$ rather than $x$. It also implies an electron-like Fermi surface since there are $\frac{(1-x)}{2}$ electrons per orbital. However, the high-field Hall effect seems to be consistent with
a hole-like Fermi surface [10]. An experimental determination of the Fermi surface would settle this issue.

*Spectral Function:* To gain further insight we have calculated the electron Green’s function. The electron is a composite object carrying both spin and charge, and is therefore a superposition of essentially infinite pairs of charge fermions and spin bosons subject only to momentum and energy conservation: 

\[ c_{k\omega\sigma} = (2S_R N\beta)^{-1/2} \sum_{q\nu} b_{q\nu\sigma} f_{k-q,\omega-\nu}, \]

where \( \omega \) and \( \nu \) are the odd and even Matsubara frequencies. Then, the electron Green’s function is a convolution of the fermion and the Boson Green’s functions [11]. We do a particle-hole transformation on the charge fermions \( h = f^* \) and work with holons, characterized by a spectrum \( \epsilon_k = -6t_f \gamma_k \). The spectral function is given by

\[ A(k,\omega) = \frac{m\pi}{S_R} \delta(\omega+\epsilon_k-\mu) + \frac{1}{8\pi^2 S_R} \int d^3q [f(\epsilon_{q-k}-\mu) + n(\omega_q+\Lambda)] \delta(\omega-\omega_q-\Lambda+\epsilon_{q-k}-\mu). \] (7)

where \( n(x) \) and \( f(x) \) are Bose and Fermi functions. Each term in the integral (sum) describes the propagation of a charge riding on a particular mode of the core spins. As is usual in Bose systems, the bosons separate into a condensate or spin-ordered and an excited or spin-disordered component. The condensate component (first term) clearly constitutes a coherent Fermi liquid. However, this is no ordinary Fermi liquid, since it represents spinless fermions with a spectral weight proportional to the magnetization \( m \), and thus weakens with increasing \( T \) and disappears at \( T_c \), as the spectral weight is continuously transferred to the second term which we call \( A_1(k,\omega) \). We now show that \( A_1 \) is incoherent, i.e., it does not have a Fermi-liquid character and has a number unusual properties.

In the ordered state \( \Lambda = 0 \). Since \( \omega_q > 0 \), it is is easy to see that, at \( T = 0 \), \( A_1 \) is nonzero only for \( \omega > 0 \). This reflects the fact that we can only create (not destroy) an excited boson at \( T = 0 \). Therefore, the approximate particle-hole symmetry near the Fermi surface, a property of ordinary Fermi liquids, is violated. This is a consequence of broken time-reversal invariance and should be observable, perhaps in photoemission and tunneling (density of states) measurements. The density of states is easier to anlayze and is given by

\[ D(\omega) = \frac{m}{2S_R} D_h(\mu-\omega) + \frac{1}{2S_R} \int dz D_h(z) D_h(z+\Lambda+\mu-\omega)[n(z+\Lambda)+f(z+\Lambda-\omega)], \] (8)
where $\omega$ is measured relative to the Fermi surface, and $D_b$ and $D_h$ are the boson and fermion densities of states. The first term is the condensate contribution. Since $D_b(z)$ nonzero only for $z > 0$, we see immediately that, at $T = 0$, the integrand contributes only if $\omega > z > 0$, exhibiting the particle-hole asymmetry. In the region $kT, \omega << \mu$, $D_h$ varies slowly with frequency, and $D_b(\omega) = \text{const.}$. $\Theta(\omega)\omega^{1/2}$ at low frequencies. Then both above and below $T_c$ we obtain, $D(\omega) = D_h(\mu + \Lambda - \omega)[\frac{1}{2} + \frac{1}{2St}\phi(\omega, T)]$, where $\phi$ is the symmetry violating contribution and has the scaling form $\phi(\omega, T) = \omega^{3/2}g(\omega/\Lambda)$. At $T = 0, \phi = \Theta(\omega)(\omega/\omega_m)^{3/2}$, where $\omega_m$ is the maximum boson energy. For $T > 0, \phi$ has the same $\omega^{3/2}$ form for $\omega >> kT$, but acquires a finite value for $\omega \leq 0$, which vanishes exponentially as $T^3/2e^{-\beta|\omega|}$ as $\omega \rightarrow -\infty$.

Precisely, on the Fermi surface ($\omega = 0$), $\phi$ is temperature dependent and scales as $T^3/2$. The spectral function itself can be calculated analytically by using the quadratic approximations for the fermion and boson spectra: $\epsilon_k \approx t_f(Q - k)^2$, $\omega_q \approx t_bq^2$. Let $\Omega \equiv (1 - \frac{t_b}{t_f})(\omega - \Lambda + \epsilon_k - \mu)/\epsilon_k$. Then,

$$ A_1 = \frac{\Theta(1 - \Omega)\Theta(\omega_m - \omega_-)}{16\pi St t_b\beta|Q - k|} \left[ \log \frac{1 + e^{-\beta(\omega_1 + \Lambda - \omega)}}{1 + e^{-\beta(\omega_2 + \Lambda - \omega)}} + \log \frac{1 - e^{-\beta(\omega_1 + \Lambda)}}{1 - e^{-\beta(\omega_2 + \Lambda)}} \right], $$

Note that $\omega_+ > \omega_- \geq 0$. The quasiparticle peak, if it exists, would occur at $\Omega = 0$. At $T = 0$ only the first term in (9) contributes. It has no singularity and exists only for $\omega > \omega_- \geq 0$, exhibiting the lack of p-h symmetry. For $T > 0$, this term just broadens out. The second term in (9) contributes only for $T > 0$ and also has a non-Fermi liquid character. In the ordered region ($\Lambda = 0$) it exhibits a logarithmic singularity $\sim -\log \omega_- \sim -\log \Omega^2$. But as shown in Fig. 2, even this weak singularity disappears for $T > T_c$ i.e., for a finite $\Lambda$ as small as 0.0025W. Therefore, the state above $T_c$ is quite remarkable: the charge of the electron is itinerant and has a Fermi surface, but the spin of the electron is localized over a distance $\xi \sim \Lambda^{-\frac{1}{2}}$. The electron itself does not exist as a well-defined quasiparticle nor does it have a well defined Fermi-surface.
To summarize, the Green’s function is a sum of two terms: one representing a coherent, perfectly polarized, spinless Fermi liquid, and the other an incoherent, non-Fermi liquid of unpolarized electrons. With increasing $T$, there is a continuous transfer of spectral weight from the coherent to the incoherent component. It is not hard to see that such a separation would also occur for higher order Green’s functions. This is consistent with the continuous transfer of spectral weight in the measured optical conductivity from a metallic to an incoherent component [12]. In this two-fluid picture, the dc conductivity is $\sigma = \sigma_c + \sigma_{inc}$, where $\sigma_c = n_c e \mu_c$ is the coherent part, with $n_c$ and $\mu_c$, the density and the mobility of the “coherent” carriers, respectively. There is of course no transport theory for the incoherent electrons—their interaction with the lattice [13] and disorder may be very important. But calculations based on scattering of itinerant electrons from spin fluctuations is clearly not applicable. However, if we accept the two-fluid picture, then, to be consistent with experiments, the incoherent contribution $\sigma_{inc}$ is similar to that for a semiconductor or Anderson-localized insulator. In particular $\mu_{inc} \ll \mu_c$, perhaps by orders of magnitude. Then, the resistivity $\rho \sim 1/\sigma_c$, i.e., is metallic at low temperature. But close to $T_c$, it rapidly goes over to $1/\sigma_{inc}$, simply because $n_c$ vanishes as $T \rightarrow T_c$. The changing $n_c$ also allows $\rho$ to change continuously without violating unitarity. An applied magnetic field has the opposite effect: it transfers electrons from the incoherent component to the metallic component by forcing the core spins to line up, the effect being strongest close to $T_c$. In the insulating regime, introduction of even a small number of coherent electrons would cause a large drop in the resistivity simply because of the very large difference in the mobility. Therefore, a modest field will result in large magnetoresistance as well as a large shift in the position of the resistivity maximum, as observed. This sensitivity to external fields is due to spin-charge separation.

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Figure Captions

Fig. 1. Energy Scales: Plot of $\frac{2kT_c}{W}$, $\frac{2\mu_f}{W}$ and the $W_f/W$ as a function of doping $x$. Here $W = 12t$ = bare bandwidth, $W_f = 12t_f$ = Fermion bandwidth at $T_c$ and $\mu_f$ = is the hole Fermi energy.

Fig. 2. The incoherent part of the spectral function $A_1$ close to the Fermi surface. Below the transition (solid line) it has a weak logarithmic singularity which disappears above $T_c$ for $\Lambda = 0.0025W$ (dotted line). The parameter $G \equiv 2\Lambda/W$. Energies are in units of $W/2$. 