Magnetic and Transport Properties of the Kondo Lattice Model with Ferromagnetic Exchange Coupling

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Abstract. The Kondo lattice model with Hund’s ferromagnetic spin coupling is investigated as a microscopic model of the perovskite-type 3d manganese oxide $R_{1-x}A_x$MnO$_3$ where $R$ and $A$ are rare earth element and alkaline earth element, respectively. We take the classical spin limit $S = \infty$ for the simplicity of the calculation, since the quantum exchange process seems to be irrelevant in the high temperature paramagnetic phase. Magnetic and transport properties of the system are calculated. In the hole doped systems, ferromagnetic instabilities are observed as the temperature is lowered. The giant magnetoresistance of this model is in excellent agreement with the experimental data of La$_{1-x}$Sr$_x$MnO$_3$.

1 Introduction

Strong correlation in electron systems is one of the unsolved problems in the field of the condensed matter physics which has been studied over several decades. Especially, after the discovery of the high-$T_c$ materials, many theoretical and experimental works have been performed to approach this problem.

One of the groups of materials that the strong correlation effect is observed in its physical properties is the 3$d$ transition-metal oxides with perovskite-like lattice structures $(R, A)$MO$_3$. In these materials, one can control the number of 3$d$ electrons by changing the species of the transition-metal $M$. By choosing the component of $R$ and $A$, rare earth element and alkaline earth element, respectively, we can change electron hopping energy

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and carrier concentration. One of the novel feature in this group of materials is that the carrier doping is possible without destroying the fundamental network of $MO_3$, so that the effect of randomness can be minimized. Prominent spin-charge coupled properties in these materials near the metal-insulator transition due to electron correlation are observed.

As an example, the effective mass of the carrier doped $LaTiO_3$ shows anomalous enhancement [1], which is considered to be the effect of the strong correlation in the vicinity of the Mott transition. From the theoretical point of view, the Hubbard model near half-filling is examined as a simplified model for the doped $LaTiO_3$. The singularity in the charge mass is observed at $n \rightarrow 1$ [2, 3].

Recently, transport properties for filling-controlled single crystals of manganese oxides $(R, A)MnO_3$ have been investigated systematically [4, 5]. In this materials, $3d$ electrons are considered to form both localized spins and itinerant electrons which are coupled each other by Hund’s interaction. Several complex phenomena due to strongly coupled spin, charge and lattice degrees of freedom are observed.

For moderately doped samples of $La_{1-x}Sr_xMnO_3$, there exists a ferromagnetic metal state below $T_c$ due to double exchange mechanism [6, 7, 8], while at $x \sim 0$, the antiferromagnetic insulator phase is observed. At moderately doped region [4], a sharp drop is observed in the resistivity below the magnetic transition temperature $T_c$. It is also reported that the field-induced magnetic moment also increases the conductivity. Namely, the giant magnetoresistance (GMR) with negative sign is observed in this material. In $(Pr, Ca)MnO_3$, the phase transition from a spin glass insulator to a ferromagnetic metal is observed by inducing the magnetic field at low temperatures. Under certain conditions, the change of resistivity is larger than ten orders of magnitude when the external magnetic field is applied [5]. The interesting points of these phenomena are that they show a new type of magnetotransport feature as well as their possibilities to be applied to devices.

In this paper, we study thermodynamical properties of a microscopic model which effectively describes the nature of $(R, A)MnO_3$. One of our aims is to calculate the conductivity and the magnetoresistance in order to compare with the experimental data in $La_{1-x}Sr_xMnO_3$. 

2
2 Model

In a Mn$^{3+}$ ion, $t_{2g}$ orbitals and $e_g$ orbital are considered to be occupied with three and one 3$d$ electrons, respectively. The doped holes enter the band constructed from strongly hybridized Mn $e_g$ and O 2$p$ orbitals. We consider that the $t_{2g}$ electrons are nearly localized and mutually coupled ferromagnetically due to Hund’s rule, so that they form $S = 3/2$ Heisenberg spins. They are also coupled with $e_g$ electrons by Hund’s coupling. Then, as a model Hamiltonian of this system, the Kondo lattice model (KLM) in three dimensions,

$$H = - \sum_{ij,\sigma} t_{ij} \left( c^\dagger_{i\sigma} c_{j\sigma} + h.c. \right) - J \sum_i \vec{\sigma}_i \cdot \vec{S}_i$$

with spin $S = 3/2$ and the Hund’s ferromagnetic coupling $J > 0$ has been proposed [9]. Here, the Pauli matrices $\vec{\sigma}_i = (\sigma_i^x, \sigma_i^y, \sigma_i^z)$ represent the spin of itinerant electrons while $\vec{S}_i$ denotes the localized spin. The Hund’s coupling parameter $J$ is estimated to be larger than the band width $W$ of the itinerant electrons, so that the system is in the strong coupling regime. In order to make a clear distinction from the antiferromagnetically coupled Kondo lattice model, we sometimes refer to this model as the $e$-$t$ model [11], since it is constructed from $e_g$ and $t_{2g}$ electrons. The carrier number of the band electron for $R_{1-x}A_x$MnO$_3$ is considered to be $n \simeq 1 - x$.

Since we consider the case where the localized spin is in a high-spin state with the ferromagnetic coupling, the effect of quantum exchange seems to be unimportant in the paramagnetic phase where the thermal fluctuation of spins is dominant. Thus we consider the infinite high-spin limit $S = \infty$ so that the localized spins are classical rotators. The Hamiltonian is described as

$$H = - \sum_{ij,\sigma} t_{ij} \left( c^\dagger_{i\sigma} c_{j\sigma} + h.c. \right) - J \sum_i \vec{\sigma}_i \cdot \vec{m}_i,$$

where $\vec{m}_i = (m_i^x, m_i^y, m_i^z)$ and $|\vec{m}|^2 = 1$. The partition function is given by

$$Z = \text{Tr}_S \text{Tr}_f \exp[-\beta(H - \mu \hat{N})].$$

Although the Hamiltonian (2) is only constructed from one-body terms for the fermion degrees of freedom, thermodynamic properties of this system are still not easy to obtain since we have to take the trace over all the localized spins in eq. (3). One of the methods
to calculate the thermodynamical quantities is the Monte Carlo method for cluster systems combined with the finite size scaling analysis. Our numerical results will be shown elsewhere. In this paper, we give another approach to obtain the thermodynamic limit of the system.

In order to calculate the thermodynamic properties in a controlled manner, we take the limit of infinite dimension \( D = \infty \). This corresponds to the limit of large coordination number where we may neglect site off-diagonal terms in self-energies and vertex corrections. In this limit, the problem is reduced to the single-site problem coupled with a dynamical mean field \([12]\). In ref. \([13]\), the present author have shown that the one-body Green’s function of the \( D = \infty \) and \( S = \infty \) KLM can be calculated exactly. The Green’s function is calculated exactly as

\[
\tilde{G}(i\omega_n) = \left\langle \left( \tilde{G}_0^{-1}(i\omega_n) + J\vec{m}\vec{\sigma} \right)^{-1} \right\rangle_{\vec{m}},
\]

where \( \left\langle \cdots \right\rangle_{\vec{m}} \) represents the thermal average over \( \vec{m} \). We determine the Weiss field \( \tilde{G}_0 \) self-consistently.

In the paramagnetic phase, we have

\[
\tilde{G}(i\omega_n) = \left\langle \frac{\tilde{G}_0(i\omega_n) - J\vec{m}\vec{\sigma}}{\tilde{G}_0(i\omega_n) - 2 - J^2|\vec{m}|^2} \right\rangle_{\vec{m}} = \frac{\tilde{G}_0(i\omega_n)^{-1}}{\tilde{G}_0(i\omega_n)^{-2} - J^2},
\]

since \( \langle \vec{m} \rangle = \vec{0} \) in the paramagnetic phase. The self-energy is then given by

\[
\Sigma(i\omega_n) = \tilde{G}_0^{-1}(i\omega_n) - \tilde{G}^{-1}(i\omega_n) = J^2\tilde{G}_0(i\omega_n).
\]

From the derivation, it is clear that, as long as we restrict ourselves to the paramagnetic phase, the Green’s function of the present model is the same as that of the system with the Ising spin \( \vec{m} = (0, 0, \pm 1) \). Therefore, the paramagnetic solution has the close relationship between that of the Falicov-Kimball model in \( D = \infty \).

Using the Kubo formula, the optical conductivity is calculated from the equation \([14, 15]\)

\[
\sigma(\omega) = \sigma_0 \sum_\sigma \int d\omega' \int d\epsilon W^2 N_0(\epsilon) \\
\times A_\sigma(\epsilon, \omega') A_\sigma(\epsilon, \omega' + \omega) \frac{f(\omega') - f(\omega' + \omega)}{\omega},
\]

where
where
\[ A_\sigma(\epsilon, \omega') = -\frac{1}{\pi} \text{Im} G_\sigma(\epsilon, \omega' + i\eta) \] (8)
is the spectral function and \( f(\omega) \) is the Fermi distribution function. Here we used the fact that the vertex corrections vanish in the limit of infinite dimension. The constant \( \sigma_0 \sim (e^2 a^2 / \hbar) \cdot (N/V) \) gives the unit of the conductivity where \( a \) is the lattice constant.

### 3 Results at \( D = \infty \)

Calculations are performed using the Lorentzian density of states as well as the semi-circular density of states with the bandwidth \( W \equiv 1 \). We express the induced magnetization by \( M = \langle m_z \rangle \).

It has been shown that the analytical structure of the Green’s function of the KLM in \( D = \infty \) and \( S = \infty \) is the same as that of the Falicov-Kimball model in \( D = \infty \) which has been studied intensively [13]. As a typical example, the imaginary part of the self-energy at the fermi level is finite \( \text{Im} \Sigma(0) \neq 0 \) in the paramagnetic phase [14, 16]. As the magnetic moment is induced, the thermal fluctuation of spins decreases which causes the decrease of the imaginary part of the self-energy.

In Fig. 1, we show the density of states of the up-spin particle at \( J = 4 \) under the magnetic field for various values of the induced magnetization \( M \). At \( M = 0 \), the density of states splits into lower and upper band in the case \( J \gg W \), which has the similar behavior as seen in the Hubbard approximation. For the semi-circular density of states, the gap opens at \( J_c = 0.5W \) [17]. Therefore, we have the metal-insulator transition at half-filling in the strong coupling region. The Kondo resonance peak is not observed in this model. As the local spin is polarized, the weight at lower band increases for up-spin fermion.

In Fig. 2, we show the optical conductivity \( \sigma(\omega) \) at \( n = 0.8 \) for various values of \( M \), which is calculated from eq. (7). At paramagnetic phase \( M = 0 \), we see a peak structure at \( \omega \sim 2J \) as well as the Drude part at \( \omega \sim 0 \). The peak at \( \omega \sim 2J \) corresponds to the excitation process from the lower band to the upper band of the split density of states. As the magnetization is induced, the peak at \( \omega \sim 2J \) diminishes and the Drude part develops. This is explained by the change of the shape in the density of states from the two-peak structure at \( M = 0 \) to the single-peak structure at \( M = 1 \). From the experimental point
of view, the optical conductivity in moderately doped La$_{1-x}$Sr$_x$MnO$_3$ shows the similar behavior when the temperature-induced magnetic moment develops [18].

In Fig. 3, we show the resistivity $\rho$ scaled by its zero-field value $\rho_0$ as a function of $M$. This curve reproduces the experimental data of GMR in La$_{1-x}$Sr$_x$MnO$_3$ [4] excellently. At $M \ll 1$ we see

$$\frac{\rho(M)}{\rho(M = 0)} = 1 - CM^2,$$

(9)

where $C \sim 4$ at $J = 4$ and $n = 0.825$. The value of $C$ decreases as the hole is doped [13], which is in qualitative agreement with the experimental data [18].

4 Discussion

The calculation of the present model in the strong coupling limit $J \gg W$ shows that the quasi-particle excitation is incoherent in the paramagnetic phase since $\text{Im} \Sigma \sim J^2/W$ is satisfied. The above results imply that the system has a short coherence length. Therefore, the effective single-site treatment of the $D = \infty$ system is justified in the strong coupling limit. The time scale of the quantum spin-flip process is estimated by $1/T_K \sim 1/W$ at $J \gg W$, which is relatively longer than the quasi-particle lifetime $1/\text{Im} \Sigma \sim W/J^2$. Then, it is also justified to take the limit of $S = \infty$.

Thus, the GMR in La$_{1-x}$Sr$_x$MnO$_3$ is quantitatively reproduced by the present calculation. In order to understand the glassy behavior in (Pr,Ca)MnO$_3$, where the band width of $e_g$ electrons are relatively narrower, it seems to be necessary to introduce antiferromagnetic exchange coupling between localized spins and impurity potentials which has been neglected in La$_{1-x}$Sr$_x$MnO$_3$, so that the frustration effect takes place. Such microscopic analysis of (Pr,Ca)MnO$_3$ and other related materials will be reported elsewhere.

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Figure Captions
Fig. 1. Density of states of up-spin particle at $J = 4$ under magnetic field.

Fig. 2. Optical conductivity at $J = 4$ $n = 0.8$ for various values of $M$.

Fig. 3. Magnetization dependence of $\rho/\rho_0$ at $\langle n \rangle = 0.8$. 