Temperature Dependence of the Conductivity in (La,Sr)MnO₃

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Abstract

Using the Kondo lattice model with classical spins in infinite dimension, conductivity in the perovskite-type 3$d$ transition-metal oxide (La,Sr)MnO₃ is theoretically studied. Green’s functions as well as spontaneous magnetization are obtained exactly on the Bethe lattice as a function of temperature. Conductivity is calculated from the Kubo formula. Below the Curie temperature, resistivity as a function of magnetization is in a good agreement with the experimental data. Anomalous behaviour in the temperature dependence of the optical conductivity observed in (La,Sr)MnO₃ is also explained.

KEYWORDS: Transition-metal oxide, manganese oxide, double-exchange ferromagnetism, optical conductivity, magnetoresistance, Kondo lattice model, infinite dimensions
The discovery of the high-$T_c$ superconducting cuprates has driven a renaissance in the study of 3$d$ transition metal oxides. One of such materials is the perovskite-type manganese oxides $(R,A)\text{MnO}_3$ with $R$ and $A$ being rare-earth and alkaline-earth ions, respectively. Under appropriate hole doping, the system becomes a ferromagnet which is explained by a double-exchange mechanism.\footnote{The double-exchange mechanism involves overlapping $d$ orbitals of the transition metal ions, which allows the exchange interaction to occur.}

One of the most prominent features in this family of materials is the giant magnetoresistance (MR) with negative sign. Resistivity decreases as the magnetic field is applied. Sharp drop in resistivity is also observed when temperature is lowered below the Curie temperature $T_c$. In La$_{1-x}$Sr$_x$MnO$_3$,\footnote{La$_{1-x}$Sr$_x$MnO$_3$ is a widely studied material in this family due to its rich phase diagram and interesting magnetic and transport properties.} resistivity at $T \gtrsim T_c$ is scaled as a function of field-induced magnetization in the form $\rho/\rho_0 = 1 - C(M_{\text{tot}}/M_s)^2$, where $M_{\text{tot}}$ and $M_s = 4\mu_B$ are the total magnetization and the nominal saturation magnetization, respectively. Here, resistivity $\rho$ is normalized by its zero-field value $\rho_0$. With an appropriate normalization, resistivity below $T_c$ is also scaled in the same form as a function of spontaneous magnetization.

In La$_{1-x}$Sr$_x$MnO$_3$, anomalous behavior in the optical conductivity $\sigma(\omega)$ is also observed.\footnote{Optical conductivity measures the response of a material to an applied electric field as a function of frequency.} At the paramagnetic phase around room temperature, a gap-like structure is observed. Below $T_c$, unconventional temperature dependence in $\sigma(\omega)$ which extends beyond $\omega \sim 2\text{eV}$ is found. Transfer of the spectral weight from the high-energy part at $\omega \gtrsim 1\text{eV}$ to the low-energy part $\omega \ll 1\text{eV}$ is observed as temperature is lowered. This implies that the electronic structure in La$_{1-x}$Sr$_x$MnO$_3$ changes in the energy scale $\sim 1\text{eV}$ by changing temperature in the scale $\sim 10^{-2}\text{eV}$. The result may not be explained from the rigid-band picture. Effects of strong electron correlation in the energy scale $\sim 1\text{eV}$ must be taken into account to understand the properties.

From the theoretical point of view, the Kondo lattice model with ferromagnetic coupling is studied as a canonical model of La$_{1-x}$Sr$_x$MnO$_3$. The Hamiltonian in the classical spin limit is described as

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

where $\vec{m}_i = (m_i^x, m_i^y, m_i^z)$ denote localized spins with $|\vec{m}|^2 = 1$, while $\vec{\sigma}_i$ represent spins of itinerant electrons. Electrons and localized spins correspond to Mn 3$d$ electrons in $e_g$ and $t_{2g}$ orbitals, respectively. Hund’s coupling $J$ is considered to be larger than the electron bandwidth in La$_{1-x}$Sr$_x$MnO$_3$. The author has examined the model in infinite
dimension \( (D = \infty) \) with a Lorentzian density of states (DOS) where the MR data in La\(_{1-x}\)Sr\(_x\)MnO\(_3\) has been well reproduced.

In this paper, we examine the model on a \( D = \infty \) Bethe lattice, where we see a semi-circular DOS (S-DOS) in the noninteracting system \( N_0(\varepsilon) = (2/\pi W) \sqrt{1 - (\varepsilon/W)^2} \). The bandwidth \( W \equiv 1 \) is hereafter taken as a unit of energy. This system has more realistic points than those with Lorentzian DOS (L-DOS): It is composed from short-range hopping terms, and the DOS has band edges. Ferromagnetic phase exists under doping, and \( T_c \) in La\(_{1-x}\)Sr\(_x\)MnO\(_3\) at \( 0.1 < x < 0.25 \) is reproduced quantitatively at \( J = 4.9 \). Here, we study temperature dependence of the conductivity at \( T \ll T_c \) and its relation with spontaneous magnetization.

In an infinite-dimensional lattice system, the model is mapped to a single-cite model interacting with a dynamical field \( \tilde{G}_0 \). Trace over fermion degrees of freedom is evaluated as

\[
\tilde{Z}_f(\vec{m}) = 4 \exp \left( \sum_n \log \det \left[ \frac{(\tilde{G}_0^{-1} + J\vec{m}\vec{\sigma})}{i\omega_n} \right] e^{i\omega_n} \right),
\]

so that the partition function is given by \( \tilde{Z} = \int d\Omega \tilde{Z}_f(\vec{m}) \). Then, Green’s function for the single-cite system is obtained in the form

\[
\tilde{G}(i\omega_n) = \frac{1}{Z} \int d\Omega \tilde{Z}_f(\vec{m}) \left( \tilde{G}_0^{-1}(i\omega_n) + J\vec{m}\vec{\sigma} \right)^{-1}.
\]

Self-energy and Green’s function for the lattice system are given by \( \tilde{\Sigma} = \tilde{G}_0^{-1} - \tilde{G}^{-1} \) and \( G(\epsilon, i\omega_n) = [i\omega_n - (\epsilon - \mu) - \tilde{\Sigma}]^{-1} \), respectively. The Weiss field \( \tilde{G}_0 \) is determined self-consistently as

\[
\tilde{G}_0^{-1} = \left( \int d\epsilon N_0(\epsilon) G(\epsilon, i\omega_n) \right)^{-1} + \tilde{\Sigma}.
\]

The nominal carrier electron number in La\(_{1-x}\)Sr\(_x\)MnO\(_3\) is \( n = 1 - x \). Hereafter we use the hole notation so that the hole concentration is expressed by \( x \). Magnetization is calculated as \( \langle \vec{m} \rangle = \int d\Omega \vec{m} \tilde{Z}_f(\vec{m})/\tilde{Z} \). Conductivity in \( D = \infty \) is calculated from the formula

\[
\sigma(\omega) = \sigma_0 \sum_\sigma \int d\omega' I_\sigma(\omega', \omega' + \omega) \frac{f(\omega') - f(\omega' + \omega)}{\omega},
\]

\[
I_\sigma(\omega_1, \omega_2) = \int N_0(\epsilon) d\epsilon W^2 A_\sigma(\epsilon, \omega_1) A_\sigma(\epsilon, \omega_2),
\]
where \( A_\sigma(\epsilon, \omega) = -\text{Im} G_\sigma(\epsilon, \omega + i\eta)/\pi \) and \( f \) is the Fermi distribution function. The constant \( \sigma_0 \) gives the unit of conductivity. Integration over \( \epsilon \) gives

\[
I_\sigma(\omega_1, \omega_2) = \frac{W^2}{2\pi^2} \text{Re} \left( \frac{\tilde{G}_R(\omega_1)}{G_{R\sigma}(\mu, \omega_1)} - \frac{\tilde{G}_R(\omega_2)}{G_{R\sigma}(\mu, \omega_2)} - \frac{\tilde{G}_A(\omega_1)}{G_{A\sigma}(\mu, \omega_1)} - \frac{\tilde{G}_A(\omega_2)}{G_{A\sigma}(\mu, \omega_2)} \right),
\]

(7)

where \( \tilde{G}_R \) and \( \tilde{G}_A \) (\( G_R \) and \( G_A \)) are the retarded and the advanced Green’s function of the single-cite (lattice) system, respectively. We calculate the resistivity as \( \rho = 1/\sigma(\omega \rightarrow 0) \).

Let us now examine temperature dependence of the resistivity. We choose \( J = 4 \) and \( x = 0.175 \), where \( T_c = 0.022 \). We calculate the resistivity \( \rho(T) \) normalized by the value at \( T_c \), \( \rho_0 = \rho(T_c) \), and the total spontaneous magnetization \( M_{\text{tot}}(T) \) normalized by the nominal saturation magnetization \( M_s \). Since we consider \( t_{2g} \) electrons with \( S = 3/2 \) and itinerant \( e_g \) electrons, we have \( M_{\text{tot}} = \frac{3}{2} \langle m_z \rangle + \frac{1}{2} \langle \hat{\sigma}_z \rangle \) and \( M_s = 2 \). Here, \( z \)-axis is taken to be parallel to the magnetization. We also calculate the resistivity and the total induced magnetization by applying magnetic field at a fixed temperature in the paramagnetic region \( T = 1.01 T_c \) for comparison. In this case, we normalize the resistivity \( \rho(H) \) by its zero-field value \( \rho_0 \).

In Fig. 1, we show \( \rho/\rho_0 \) as a function of \( M_{\text{tot}}/M_s \) for both cases of calculation by changing temperature \( T \) and the external magnetic field \( H \) at the system with the S-DOS. We also show the result for L-DOS in the equal condition, where the resistivity is changed by the magnetic field. The difference between any of the three cases are small in the wide region of \( M_{\text{tot}}/M_s \). Thus, magnetization is the essential thermodynamical variable that determines the nature of the resistivity, since quasi-particle properties such as life-time and DOS are strongly magnetization dependent. Effects of temperature is subsidiary here, because we are in the limit \( T \ll W \). It is analogous to the nature in La\(_{1-x}\)Sr\(_x\)MnO\(_3\) that the resistivity is scaled by the magnetization in a universal way.

We then make a quantitative comparison with the experimental data of La\(_{1-x}\)Sr\(_x\)MnO\(_3\) at \( x = 0.175 \) where \( T_c = 283 \text{K} \). In Fig. 1, we plot the resistivity at \( T = 294 \text{K} \) as a function of induced magnetization normalized by its zero-field value. Temperature dependence of the resistivity below \( T_c \) with an appropriate normalization is also shown as a function of spontaneous magnetization. The experimental result in La\(_{1-x}\)Sr\(_x\)MnO\(_3\) is reproduced quantitatively at \( M_{\text{tot}}/M_s \ll 0.2 \).

Now, optical conductivity is calculated at \( J = 2 \) and \( x = 0.2 \). In Fig. 2, we show \( \sigma(\omega) \) at \( T = 1.05 T_c \), \( 0.5 T_c \) and \( 0.25 T_c \), where \( T_c = 0.019 \). In the paramagnetic phase at
$T = 1.05 T_c$, we see a two-peak structure in $\sigma(\omega)$ at $\omega \sim 0$ and $\omega = 2J$, and a gap in between. As temperature is lowered below $T_c$, a transfer of the weight from the high-energy part $\omega \sim 2J$ to the Drude part $\omega \sim 0$ is clearly observed. The structure of the spectra and its temperature dependence is found to be qualitatively unchanged at $J \gtrsim 1$ in the carrier doped system. The nature of $\sigma(\omega)$ agrees with the experiment in a qualitative way.

The above result is understood from the change of the quasi-particle DOS, which is shown in the inset of Fig. 2. In the paramagnetic phase at $T > T_c$, the DOS equally splits into the lower sub-band and the upper sub-band at $\omega \sim \pm J$, which are the spectra for quasi-particles with spins parallel and anti-parallel to the localized spins, respectively. The origin of the peaks at $\omega \sim 0$ and $\omega \sim 2J$ in $\sigma(\omega)$ is the intra-band and inter-band excitation processes, respectively. We note here that the fermi level is at the lower band in the case of hole doping. Below $T_c$, polarization of localized spins give rise to the change in the probability that an electron on a site has a spin parallel (anti-parallel) to the localized spin on the site. Therefore, the weights of the sub-bands change as shown in the figure. Amplitude of the optical conductivity is roughly proportional to the product of the DOS of particle and hole excitations with the same spin species. Then, as the magnetization increases, the weight of $\sigma(\omega)$ transfers to the low frequency part.

It has been made clear in this paper that the transport properties of the system are intensively influenced by the variation of the magnetic structure because itinerant electrons and localized spins are strongly coupled. Resistivity is universally scaled by the magnetization. Neither temperature nor external magnetic field are essential parameters. Transfer in the weight of the optical conductivity is due to the change in the quasi-particle DOS by the magnetization of localized spins.

In order to compare dynamical processes of the system with experiments, one should also treat the motions of localized spins which are coupled not only to itinerant electrons but also to the environment. Within the current framework, such dynamics of localized spins is replaced by the thermal average over spin fluctuations. The error due to this treatment is proportional to $1/D$ because it is a spatial correlation term, so it vanishes at $D \to \infty$. This replacement is also valid in finite dimensions if the characteristic time-scale of the process is much longer than the thermal relaxation time of the localized spin $\tau_s \gtrsim 1/T$. Therefore, the present approach for the conductivity $\sigma(\omega)$ is justified at $\omega \ll 1/\tau_s$ in the above sense, but may give inaccurate results for $\omega \gtrsim 1/\tau_s$ if we
compare with experiments. We speculate that this is one of the reasons why the optical conductivity spectra at $\omega \sim 2J$ is not reproduced quantitatively while the resistivity is in a good agreement with experiments. In any cases, the qualitative nature of the transfer in the weight of $\sigma(\omega)$ seems to be unchanged; it is mainly determined by the quasi-particle DOS of the initial and the final states.

To summarize, we have calculated the Kondo lattice model with classical spins on a Bethe lattice in $D = \infty$. Temperature dependence of the conductivity is calculated. As a function of the spontaneous magnetization, resistivity is scaled in the same form as in the case of MR. The result agrees with the experimental data not only qualitatively but also in a quantitative way. Anomalous behavior in the optical conductivity of $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ is explained from the change in the quasi-particle DOS. Due to the strong coupling between itinerant electrons and localized spins, transport properties are intensively influenced by the magnetization of the system.

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Figure Captions

Fig. 1:  Resistivity as a function of total spontaneous magnetization (circle). Resistivity as a function of total field-induced magnetization for semi-circular (triangle) and Lorentzian (dashed curve) DOS are also shown. The experimental data measured by changing magnetic field (square) and temperature (solid curve) are taken from ref. [4].

Fig. 2:  Optical conductivity for $J = 2$ and $x = 0.2$. Inset shows the DOS in an arbitrary unit for the up-spin quasi-particle. The down-spin DOS at $\omega$ is equal to the up-spin DOS at $-\omega$ from the symmetry. Temperatures are $T = 1.05T_c$ (solid curves), $T = 0.5T_c$ (dashed curves) and $T = 0.25T_c$ (dotted curves).

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La$_{1-x}$Sr$_x$MnO$_3$
$x=0.175$

$\rho / \rho_0$

$M_{\text{tot}} / M_s$

$T=294K$

$\rho (T)$

$D=\infty \ S=\infty \ KLM$

$J/W = 4$

$S$-DOS (by T)

$L$-DOS (by H)

$S$-DOS (by T)

$L$-DOS (by H)

Fig. 1
Fig. 2