Double Degeneracy and Jahn-Teller Effects in CMR Perovskites

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Abstract

Jahn-Teller (JT) electron-phonon coupling effects in the colossal magnetoresistance perovskite compounds $La_{1-x}A_xMnO_3$ are investigated. Electron-electron correlations between two degenerate Mn $e_g$ orbitals are studied in the Gutzwiller approximation. The static JT distortion and antiadiabatic polaron effects are studied in a modified Lang-Firsov approximation. We find that (i) the electron or hole character of the charge carrier depends on the static JT distortion, and (ii) due to the two-component nature of the JT coupling, fluctuations in the JT distortion direction contribute to the charge transport in similar fashion as the local spins.

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The recently rediscovered “colossal magnetoresistance” (CMR) perovskite-based $La_{1-x}A_xMnO_3$ compounds ($A = Ca, Sr, Ba$) exhibit striking magnetic field and temperature dependence of the resistivity\(^1\). Strong correlation between the electrical resistivity and ferromagnetism in these materials\(^2\) was qualitatively explained by Zener’s double exchange (DE) model\(^3\). In this model, the three $t_{2g}$ Mn $d_{xy}, d_{xz}, d_{yz}$ orbitals are completely filled and the electrons are localized; thus their spins can be treated as a localized composite spin $S = 3/2$; the conduction electrons are provided by $e_g$ electrons on the Mn$^{3+}$ ions. Strong Hund’s-rule coupling of strength $J^H \gg t$ (the hopping matrix element) of the electronically active $e_g$ electrons to the localized $t_{2g}$ electrons, results in the motion of electrons spin-polarizing the localized Mn spins and leading to ferromagnetism. An applied magnetic field will polarize the local spins and thus decrease the electrical resistivity. The DE model was subsequently refined by Anderson and Hasegawa\(^4\), and de Gennes\(^5\), and studied more recently\(^6\) to explain topical experiments. However, it now appears that the DE model can not adequately explain various experimental data by itself: Jahn-Teller (JT) electron-phonon coupling also plays an essential role\(^7\)–\(^10\).

Previously, we studied a model of combined DE and JT coupling\(^11\) and found that the JT coupling can substantially reduce the resistivity and the Curie temperature $T_c$. Using this model, we can qualitatively understand the metal-insulator (MI) transition around $T_c$: namely, a polaron localized by the combination of the changing of the spin polarization cloud around it and Anderson localization due to the dramatic increase of spin-disorder scattering near $T_c$. Here we study the DE model with JT coupling using two $e_g$ orbitals at each lattice site. One motivation is to incorporate the doubly-degenerate states to take into account the mobile carrier density correctly and study the electron-electron (e-e) on-site correlation effects. A second motivation is that the doubly-degenerate states act like a pseudospin which couples to a vector (of two phonon modes). For $T$ close to or above $T_c$, a pseudospin-induced “double exchange” effect will then become important (see below).

The Hamiltonian we use consists of the DE part $H^{DE}$ and coupling to the lattice represented by $H^{JT}$. $H^{DE}$ can be derived from the Kondo-lattice Hamiltonian...
\[ H^{DE} = -t_0 \sum_{\langle i,j \rangle} (a_{i\alpha}^\dagger a_{j\alpha} + b_{i\alpha}^\dagger b_{j\alpha} + \text{h.c.}) + V \sum_i n_i^a n_i^b - J_H \sum_i \vec{S}_i \cdot \vec{\sigma}_{\alpha\beta} (a_{i\alpha}^\dagger a_{i\beta} + b_{i\alpha}^\dagger b_{i\beta}), \] (1)

where \( \vec{\sigma} \) is the Pauli matrix and \( \vec{S}_i \) is the local spin of the three \( t_{2g} \) electrons \( (S = 3/2) \). The operators \( a_{i\alpha} \) \( (a_{i\alpha}^\dagger) \) and \( b_{i\alpha} \) \( (b_{i\alpha}^\dagger) \) annihilate (create) an electron at the two \( e_g \) orbitals \( (a \text{ and } b) \) with spin \( \alpha \) at site \( i \).

The second term in Eq.(1) describes the e-e interaction between \( a \) and \( b \) orbitals on the same site. A detailed calculation showed that for the Hund’s-rule coupling \( J_H \gg t_0/2 \), the eigenstates of a two-site systems are approximately formed by electrons all parallel (or all anti-parallel) to the on-site local spins. Based on the counting of single electronic states, it is natural to postulate this will also be true for \( N \)-site systems. So in the limit of \( J_H \gg t_0/2 \), \( H^{DE} \) can be written as

\[ H^{DE} = -t_0 \sum_{\langle i,j \rangle} \gamma_{ij}^S (a_{i\alpha}^\dagger a_{j\alpha} + b_{i\alpha}^\dagger b_{j\alpha} + \text{h.c.}) + V \sum_i n_i^a n_i^b, \] (2)

where we have omitted the spin index of the electrons since the electronic spin is fixed by the local spins: the single-electron states form two bands with a gap \( \sim 2J_H \gg k_B T \). Thus only the lower band is occupied, and it can be assumed there is no double degeneracy of each \( e_g \) orbital. (This assumption is further strengthened if there is a large on-site Hubbard interaction). \( \gamma_{ij}^S \) is the bandwidth renormalization due to the DE mechanism: \( \gamma_{ij}^S = \left< \frac{s_{ij}^{+1/2}}{2S+1} \right> \), where \( s_{ij} \) is the total spin of the subsystem of the two localized spins on sites \( i \) and \( j \) and the electronic spin. The angular brackets \( < \ldots > \) denote a thermal average.

\( H^{JT} \) can be written as

\[ H^{JT} = \lambda_{JT} \sum_i (Q_{1,i} \vec{\tau}_i^z + Q_{2,i} \vec{\tau}_i^x) + \frac{K}{2} \sum_i (Q_{1,i}^2 + Q_{2,i}^2) + \frac{M}{2} (Q_{1,i}^2 + Q_{2,i}^2), \] (3)

where \( Q_1 = (1/\sqrt{6})(2\delta Z - \delta X - \delta Y) \) and \( Q_2 = (1/\sqrt{2})(\delta X - \delta Y) \) are the two local JT distortions, with \( \delta X^i \) the distance change between neighboring \( O^{2-} \) ions along the \( X^i \)-axis. Here we have used pseudospin notation to represent the two degenerate orbitals: \( c_{\uparrow} = a; \ c_{\downarrow} = b \) with \( \bar{\alpha} \) denoting the pseudospin index. Then \( \vec{\tau}_i = \vec{\sigma}_{\bar{\alpha}i}^\dagger c_{\bar{\alpha}i} \). When we define the
distortion vector $\vec{Q}$ by $Q_x = Q_1$ and $Q_z = Q_2$, the JT coupling becomes $\lambda_{JT} \vec{Q} \cdot \vec{\tau}$. This shows clearly that the eigenenergy of the system is independent of the distortion direction $\vec{Q}/|Q|$. If we assume that $|Q|$ is fixed, $\vec{Q}$ can be treated as a local "classical" spin. Then the JT coupling is formally similar to the electron-spin coupling in Eq.(1). In the CMR perovskites, if the JT electron-lattice coupling is close to the adiabatic limit and the coupling is strong (i.e. effective $\lambda_{JT}|Q| \gg t_0/2$), it is thus possible effectively to have additional "double exchange" from the 2-component-doublet JT coupling. However, there are several mechanisms which will stabilize a specific distortion direction, including (1) anharmonic lattice interaction\textsuperscript{20}, (2) higher-order JT interactions\textsuperscript{20}, (3) interactions between local distortions at different sites\textsuperscript{21}. To be specific, we consider only effect (3) here, by introducing an effective near-neighbor "ferromagnetic" coupling. At low $T$, the JT distortion will be homogeneous in a certain direction. However, at higher $T$, fluctuations of the local distortions will induce the additional "double exchange" effects. The local JT distortion will start to be observed for $T \lesssim T_c$\textsuperscript{15}. Since $\vec{Q}$ is a classical variable constrained to the $x - z$ plane instead of a sphere, the fluctuations of $\vec{Q}/|Q|$ will be larger than a corresponding local spin $\vec{S}$. Thus its fluctuation is still significant at $T \lesssim T_c$ even though the Curie temperature $T_c$ is smaller than the "paramagnetic transition temperature" of $\vec{Q}/|Q|$ alone.

The following calculations are restricted to the metallic phase ($T < T_c$). Incorporating the above JT distortion-induced "DE" effect introduces an additional bandwidth reduction factor $\gamma_{JT}$. At low $T$, the distortion direction is "frozen", $\gamma_{JT} \equiv 1$. At $T$ closer to $T_c$, we will approximate the stabilizing force by a "ferromagnetic" coupling between the $\vec{Q}_i$’s. The strength of this coupling is unknown and needs to be determined by (e.g. structural) experiments. After a rotation to make the distortion direction point along the $z$-axis and quantization of $\mathcal{H}^{JT}$, the Hamiltonian components become

$$\begin{align*}
\mathcal{H}^{JT} &= -\sqrt{\epsilon_p} \hbar \omega \sum_{i\alpha} n_{i\alpha} \sigma_{\alpha}^z (B_i^\dagger + B_i) + \hbar \omega \sum_i B_i^\dagger B_i, \\
\mathcal{H}^{DE} &= -t_0 \sum_{\langle i, j \rangle} \gamma_{i,j}^{\alpha} \gamma_{JT} \sum_{\alpha} c_{i\alpha}^\dagger c_{j\alpha} + \text{h.c.}) + V \sum_i n_i \bar{\sigma}_\uparrow n_i \bar{\sigma}_\downarrow. \tag{4}
\end{align*}$$

Here, the electron-phonon coupling strength is $\epsilon_p = (\lambda_{JT})^2/2K$, and $\hbar \omega$ is the phonon
frequency.

We treat the dynamical phonons within the homogeneously modified variational Lang-Firsov approximation using a canonical transformation \( U = e^{-S_1(\{\Delta\})} e^{-S_2(\gamma)} \), where \( S_1 = 1/2\sqrt{\epsilon_p \hbar \omega} \sum_i \Delta (B^\dagger_i - B_i) \) is designed to describe static lattice distortions through the introduction of a (variational) static displacement field \( \Delta \). \( S_2 = -\sqrt{\epsilon_p / \hbar \omega} \sum_i \gamma n_i \sigma^z (B^\dagger_i - B_i) \) describes the antiadiabatic polaron formation, with \( \gamma \) measuring the degree of the polaron effect. After the transformation \( U \) we can average \( \tilde{\mathcal{H}} \) over a squeezed phonon state \( |\Psi_{ph}\rangle = \exp(-\sum_i \tau_i (B_i B^\dagger_i - B^\dagger_i B_i)) |\text{vac}\rangle \) with \( \tau \) a variational parameter. The e-e correlations are approximated by the Gutzwiller approximation. The free energy can then be written as

\[
\mathcal{F} = -\lambda_a (p_a + d) - \lambda_b (p_b + d) + (V + 2\epsilon_p \gamma (2 - \gamma))d \\
- \frac{1}{\beta} \sum_{k, i \in \{a, b\}} \ln[1 + e^{-\beta(\epsilon_k - \mu + \lambda_i)}] - \epsilon_p (2\gamma - \gamma^2)(1 - x) \\
+ \frac{\hbar \omega}{4} (\tau^2 + \tau^{-2}) + \frac{\Delta^2}{4\epsilon_p} + (1 - \gamma)\Delta (p_a - p_b) + \mathcal{F}_s + \mathcal{F}_q
\]

with the electron dispersion relation given by \( \epsilon_k = 2\gamma_s \gamma_j \xi(\gamma)(\cos(k_x) + \cos(k_y) + \cos(k_z)) \) with a polaronic band narrowing factor \( \xi(\gamma) = \exp(-\epsilon_p \gamma^2 \tau^2 / (\hbar \omega)) \). Here we have used the notation \( \uparrow \to a; \downarrow \to b \). \( q_i \) are the discontinuities in the single-particle occupation number at the Fermi surfaces, which give additional band narrowing due to e-e interactions: \( m_i^*/m_0 \sim 1/q_i \). The relations between \( q_i \) and \( p_i \) (probability of occupation of orbital \( i \)) and \( d \) (double occupancy) is

\[
q_i \equiv \left( \frac{[p_i(1 - p_a - p_b - d)]^{1/2} + (p_d)^{1/2}}{p_i + d(1 - p_i - d)} \right)^2,
\]

where \( \bar{a} \equiv b \) and \( \bar{b} \equiv a \). \( x \) is the doping density. Note the crucial distinction between the static JT and antiadiabatic polaron distortions: the double degeneracy of \( e_g \) is lifted by \( (1 - \gamma)\Delta \); in contrast, the antiadiabatic polaron distortion contributes an effective e-e interaction \( 2\gamma (2 - \gamma)\epsilon_p \).

\( \mathcal{F}_s \) and \( \mathcal{F}_q \) in Eq.(5) are free energies from the local spin \( S = 3/2 \) and JT distortion
direction $\vec{Q}/|Q|$, respectively. We used molecular crystal approximations for both $\mathcal{F}_\parallel$ and $\mathcal{F}_\perp$. $\mathcal{F}_\perp$ contributes only at high $T$:

$$\mathcal{F}_\perp = l_{\text{eff}} \langle Q_z \rangle - \log(\nu_{JT})/\beta - 6J_{\text{eff}} \langle Q_z \rangle^2. \tag{7}$$

Here $l_{\text{eff}}$ is the effective field for $\vec{Q}/|Q|$, and $\nu_{JT} = I_0(\beta l_{\text{eff}})$. $\langle Q_z \rangle = I_1(\beta l_{\text{eff}})/I_0(\beta l_{\text{eff}})$. In this molecular crystal approximation, $\gamma_{JT} = \sinh(2\beta l_{\text{eff}})/(\pi \beta l_{\text{eff}} I_0^2(\beta l_{\text{eff}}))$, where $I_n(x)$ is the $n$-th order modified Bessel function.

The mean-field equations at low $T$ ($\gamma_{JT} \equiv 1$) are derived by minimizing the free-energy Eq.(5):

$$\lambda_j - \epsilon_p (1 - \gamma)^2 (p_j - p) = \sum_{i \in \{a,b\}} \int d\epsilon \rho(\epsilon) \left( \frac{\partial q_i}{\partial p_j} \right) f(q_i, \lambda_i, \epsilon) \tag{8}$$

$$1 - x = \sum_{i \in \{a,b\}} \int d\epsilon \rho(\epsilon) f(q_i, \lambda_i, \epsilon) \tag{9}$$

$$\gamma \frac{\tau^2}{h \omega} \sum_{i \in \{a,b\}} \int d\epsilon \rho(\epsilon) (q_i \epsilon) f(q_i, \lambda_i, \epsilon) \tag{10}$$

$$(1 - \frac{1}{\tau}) = \frac{4\epsilon_p \gamma^2}{(h \omega)^2} \sum_{i \in \{a,b\}} \int d\epsilon \rho(\epsilon) (q_i \epsilon) f(q_i, \lambda_i, \epsilon) \tag{11}$$

Here $f(q_i, \lambda_i, \epsilon) = [1 + e^{\beta(q_i \epsilon - \mu + \lambda_i)}]^{-1}$ is the Fermi-distribution function, and $\rho(\epsilon)$ is the electronic density of states. For the dispersion relation $\epsilon_k = 2t_{\text{eff}} (\cos(k_x) + \cos(k_y) + \cos(k_z))$, $\rho(\epsilon) = \frac{1}{2\pi t_{\text{eff}}} \int_0^\infty dx \cos \left( \frac{x \epsilon}{2t_{\text{eff}}} \right) [J_0(x)]^3$, with $J_0(x)$ the zeroth order Bessel function. Below, we evaluate $\Phi(z) = \int_{-\infty}^z dx \rho(x)$, which converges easily and can be accurately approximated with high order Chebyshev polynomials.
Eq. (8)-(13) are solved numerically by iteration. The resistivity is calculated using the same approximation as in Ref. [9], which is comparable to the memory function method [11]. The resistivity is given by:

\[ \rho = \rho_a \rho_b / (\rho_a + \rho_b) \]

where \( \rho_i \propto (m^*)^2 \Delta (\gamma_s \gamma_{JT}) \) with \( m^* \sim 1/t_{eff} \) and

\[
\Delta (\gamma_s \gamma_{JT}) = \langle \left( \frac{s_0 + 1/2}{2S + 1} \right)^2 \cos^2 (\theta_{ij}) \rangle - (\gamma_s \gamma_{JT})^2
\]

with \( \gamma_{JT} \equiv 1 \) at low \( T \): 

\[
\Delta (\gamma_s \gamma_{JT}) = \langle \left( \frac{s_0 + 1/2}{2S + 1} \right)^2 \rangle - (\gamma_s)^2
\]

At higher \( T \), the fluctuation of local JT distortions begins contributing to the resistivity. The self-consistent equation for \( l_{eff} \) can be derived easily from minimizing the free energy, and is similar to Eq. (13) through the substitution: \( h \rightarrow l_{eff}, m \rightarrow \langle \cos \theta \rangle \), and \( \gamma_s \rightarrow \gamma_{JT} \). The resistivity is readily calculated given \( \langle \cos^2 (\theta) \rangle = \frac{1}{2} (1 + I^2_{L} / I^2_{T} (\beta l_{eff})) \).

In Fig. 1, we show the variations of the electron density of each \( e_g \) channel. It can be seen that the character of the carrier is strikingly dependent on the static JT distortion \( \Delta \). For small \( \Delta \) (or \( \Delta = 0 \) when there is no JT coupling), the charge carriers will be electrons, since the electron density in both channels is lower than half-filling. Only with large enough \( \Delta \) does the carrier (in the majority channel) become hole-like. The e-e interaction \( V \) can change only the double occupancy but not the carrier character. (Nagaoka “ferromagnetism” is unlikely here due to the doping \( x \) and moderate \( V \) interaction strength). With large JT distortion, because of the creation of the majority channel, the e-e interaction \( V \) is unimportant for charge transport. Based on this result, if experiments (e.g. Hall transport or thermoelectric power) can detect the hole-character of the charge carriers, this will indirectly indicate the presence of a large static JT distortion, either local or homogeneous. It is indeed observed that the thermoelectric power is positive (hole-like) at \( x = 0.2 \) and negative (electron-like) at \( x = 0.4 \), which is consistent with Fig. 1 in a certain range of electron-phonon coupling and phonon frequency. In our choice of model parameters we have used the hopping matrix element as the energy unit: the temperature unit is calculated with \( t_0 = 1ev \), so if \( t_0 = 0.2ev \), then \( T = 400 \) will be \( T = 80K \). The unit of magnetic field is renormalized by \( H = g\mu_B B \), where \( g \) is the effective Landé \( g \)-factor. Our self-consistent mean-field solution
gives $T_c \sim 2550$ for $x = 0.2$, i.e. $T_c \sim 500K$ with $t_0 = 0.2ev$.

Fig. 2 shows the magnetic-field dependence of the magnetoresistance ratio $\Delta \rho/\rho_0$ in the low-$T$ ($\gamma_{JT} \equiv 1$) region. The global features of the $\Delta \rho/\rho_0 - H$ curves are somewhat similar to the experimental data.\textsuperscript{23} Note that we have not included any additional “residual” resistivity mechanisms (disorder, phonon scattering, etc) which will strongly decrease $\Delta \rho/\rho_0$ at low $T$, and would need to be subtracted from experimental data for comparison with our prediction. From various experiments, $\Delta \rho/\rho_0$ can be as large as 0.8 for moderate to high $T$ around $T_c$, so the spin-fluctuation scattering considered here will then dominate. The temperature dependence of the magnetoresistivity ratio $\Delta \rho/\rho_0$ is shown in Fig. 3. Without the local JT distortion direction fluctuation ($\gamma_{JT} \equiv 1$), the $\Delta \rho/\rho_0 - T$ curve is flat close to $T_c$. At high $T$ ($T < T_C$ and $\gamma_{JT} \neq 1$), the effects due to the local JT distortion fluctuations can make $|\Delta \rho/\rho_0|$ increase with $T$, see Fig. 3, which has also been observed.\textsuperscript{26} Even without self-consistently including higher order JT coupling, our calculations qualitatively agree with the experiments: there is a characteristic temperature $T_p$ ($T_p \sim 1000$ for the parameters we chose). For $T < T_p$, the local JT distortion direction is frozen, the resistivity is then due to fluctuation of local spins and other “residual” scattering mechanisms. For $T_p < T < T_c$, the JT distortion direction fluctuation begins to contribute to the resistivity, and gives correct magnitude and $T$ dependence of $\Delta \rho/\rho_0$.

In conclusion, we have studied the combined effects of JT electron-lattice coupling and the double degeneracy of $e_g$ orbitals in the CMR perovskites in their metallic phase ($T < T_c$). We found that without the static JT distortion, the carriers are electrons. However, with sufficiently large static JT distortion, the carrier can be hole-like. Also, the additional scattering effects of the JT distortion (effective “double exchange” effects of the local JT distortion direction) will be enhanced in the inhomogeneous insulating phase ($T > T_c$) where small magnetic polarons are localized. This will be reported elsewhere.

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FIGURES

FIG. 1. Occupation density variation of each $e_g$ channel with the electron-lattice coupling $\epsilon_p$ at low $T$. $t_0 = 1$, $V = 6.0$, $\hbar\omega = 0.5$, $T = 400$. $\Delta$ increases monotonically with $\epsilon_p$.

FIG. 2. The variation of $\Delta \rho/\rho_0$ with magnetic field at low $T$ ($\gammaJT \equiv 1$). $t_0 = 1$, $\epsilon_p = 2.0$, $V = 6.0$, $\hbar\omega = 0.5$.

FIG. 3. The variation of $\Delta \rho/\rho_0$ with $T$ at low ($\gammaJT \equiv 1$) and high $T$ ($T < T_c$) with $J_{eff} = 0.1$. Parameters are the same as in Fig.2 and $\Delta \rho = \rho(H = 0.03) - \rho_0$. Note that the magnitude of $\Delta \rho/\rho_0$ at low $T$ can not be compared directly with experiments due to “residual” scattering (see text).