

Double exchange and the cause of ferromagnetism in doped manganites

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The coexistence of ferromagnetism and metallic conduction in doped manganites has long been explained by a double exchange model (ferromagnetic Kondo-lattice model) in which the ferromagnetic exchange arises from the carrier hopping. We evaluate the zero-temperature spin stiffness $D(0)$ and the Curie temperature T_C on the basis of the double exchange model using the measured values of the bare bandwidth W and the Hund's rule coupling J_H . The calculated $D(0)$ and T_C values are too small compared with the observed ones even without considering a strong electron-phonon coupling. We thus suggest that the ferromagnetism in doped manganites should not originate from the double exchange interaction. On the other hand, an alternative model based on the d - p exchange and a strong electron-phonon coupling can quantitatively explain the magnetoresistance and T_C values.

The discovery of "colossal" magnetoresistance in thin films of the manganite perovskites $R_{1-x}D_x\text{MnO}_3$ (R = a rare-earth element, and D = a divalent element)^{1,2} has stimulated extensive studies of magnetic, structural and transport properties of these materials.³ The coexistence of ferromagnetism and metallic conduction has long been explained by the double exchange (DE) model,^{4,5} where the effective hopping for the manganese $3d$ conduction electrons varies with the angle between the manganese core electrons due to a strong Hund's coupling. However, Millis *et al.*⁶ proposed that, in addition to the double exchange, a strong electron-phonon interaction arising from a strong Jahn-Teller effect should be involved to explain the basic physics of manganites. In this modified model, the primary cause of the ferromagnetism of doped manganites is still the double exchange interaction.

In the DE model, it is implicitly assumed that doped carriers are Mn e_g electrons. However, electron-energy-loss spectra (EELS),⁷ photoemission spectroscopy,⁸ and an *ab initio* Hartree-Fock band-structure calculation⁹ have shown that the ferromagnetic manganites ($x < 0.4$) are doped charge-transfer insulators with carriers mainly residing on the oxygen orbitals. Thus even the basic assumption in the DE model is not justified.

There are two important parameters in the double exchange model (ferromagnetic Kondo-lattice model), namely, the bare bandwidth W of the e_g bands, and the Hund's rule coupling J_H between e_g and t_{2g} electrons. These parameters are related to an optical transition between the exchange splitted e_g bands,^{10,11} and thus can be reliably determined from optical data. With these unbiased parameters, one can calculate the zero-temperature spin stiffness $D(0)$ and T_C without adjustable parameters. The parameter-free calculations for the two measurable quantities can clearly address whether the DE interaction causes the ferromagnetism of doped manganites.

Here we use the measured values of the bare bandwidth W and the Hund's rule coupling J_H to calculate $D(0)$ and T_C on the basis of the double exchange model. The calculated $D(0)$ and T_C values are too small compared with the observed ones even without taking into account a strong

electron-phonon coupling. On the other hand, an alternative model based on the d - p exchange and a strong electron-phonon coupling can quantitatively explain the magnetoresistance and the T_C values.

Now we start with a Kondo-lattice-type Hamiltonian,¹² which leads to Zener's DE model when $J_H \rightarrow \infty$,

$$H = -\frac{1}{2} \sum_{\langle ij \rangle ab\alpha} t_{ij}^{ab} (d_{ia\alpha}^\dagger d_{jb\alpha} + \text{H.c.}) - J_H \sum_{i\alpha\beta} \vec{S}_i \cdot d_{i\alpha\alpha}^\dagger \vec{\sigma}_{\alpha\beta} d_{i\alpha\beta} + H_{INT}. \quad (1)$$

Here $d_{ia\alpha}^\dagger$ creates an electron in e_g orbital a with spin α , t_{ij}^{ab} is the direction-dependent amplitude for an electron to hop from orbital a to orbital b on a neighboring site, and H_{INT} represents the other interactions. The calculated band structure is well fit by a t_{ij}^{ab} , which involves only nearest-neighbor hopping that is only nonzero for one particular linear combination of orbitals, i.e., $t_{ij}^{ab} \propto W$. Here we still call Eq. (1) two-orbital DE model rather than two-orbital Kondo-lattice model for convenience. The quantum and thermal average of the hopping term in Eq. (1), defines a quantity K :

$$K = (1/6N_{site}) \sum_{\langle ij \rangle ab\alpha} t_{ij}^{ab} \langle d_{ia\alpha}^\dagger d_{jb\alpha} + \text{H.c.} \rangle. \quad (2)$$

The quantity K is related to the optical spectral weight by a familiar sum rule,

$$K = \frac{2a_0}{\pi e^2} \int_0^\infty d\omega \sigma_1(\omega), \quad (3)$$

where σ_1 is the real-part optical conductivity contributed only from the e_g electrons, and a_0 is the lattice constant. The quantity K generally consists of the Drude part K_D , and incoherent part K_I which, in general, involves interband and intraband optical transitions. The Drude part K_D can be related to the plasma frequency Ω_p as

$$K_D = \frac{a_0}{4\pi e^2} (\hbar\Omega_p)^2. \quad (4)$$

On the basis of Eq. (1), Quijada *et al.*¹² showed that, to the order of $1/J_H$, the spin stiffness $D(0)$ is

$$D(0) = \frac{Ka_0^2}{4S^*} \left(1 - \frac{\eta W^2}{16J_H SK} \right), \quad (5)$$

where $S=3/2$, $S^*=S+(1-x)/2$, and $\eta=1.04$ when $H_{INT}=0$. The presence of interactions may change the value of η . A similar result was obtained by Furukawa¹⁰ for a one-orbital DE model using the dynamical mean-field method, but the value of η is doping dependent and less than 1.

From the above equations, one can calculate K and $D(0)$ using realistic values of the bare bandwidth W and the Hund's rule coupling J_H . Both the local-density approximation (LDA) (Ref. 13) and "constrained" LDA (Ref. 14) calculations show that $J_H \approx 1.5$ eV. The calculated J_H value is very close to the atomic values for $3d$ atoms. This is reasonable because J_H is not screened when the ion is put in a solid. The bare bandwidth W cannot be calculated reliably for $3d$ metal based compounds due to a strong correlation effect. Fortunately, the values of both W and J_H can be determined from an optical transition between the exchange splitted e_g bands.^{10,11} The peak position of this optical transition is about $2J_H$, and the width of the peak contains information about the bare bandwidth.^{10,11} From the optical data of Refs. 12 and 15, one finds $J_H \approx 1.6$ eV and $W = 1.6-1.8$ eV by comparing the data with the theoretical predictions.^{10,11} The value of J_H obtained from the optical data is in excellent agreement with the calculated one. This implies that the feature appeared at about 3 eV in the optical data indeed arises from the optical transition between the exchange splitted e_g bands.

The quantity K^0 for the noninteracting two-orbital model can be evaluated when the bare bandwidth W is known. Takahashi and Shiba¹⁶ have calculated the optical conductivity using a tight-binding (TB) approximation of the band structure. From their calculated result for the interband optical conductivity, we evaluate that $K_I^0 = 0.022W$ for $x \sim 0.3$. Since $K_D^0 = 1.2K_I^0$,¹⁶ then $K_D^0 = 0.026W$ and $K^0 = 0.048W$. The LDA calculation for a cubic and undistorted structure shows that¹⁷ $W = 3$ eV and $\hbar\Omega_p^0 = 1.9$ eV. Using Eq. (4) and $\hbar\Omega_p^0 = 1.9$ eV, one yields $K_D^0 = 78.6$ meV. Comparing $W = 3$ eV with $K_D^0 = 78.6$ meV, one readily finds that $K_D^0 = 0.026W$, in remarkably good agreement with that ($K_D^0 = 0.026W$) estimated from the TB approximation. This justifies the relation $K^0 = 0.048W$ obtained from the TB approximation.

When the Hund's coupling J_H is turned on, the quantity K is reduced compared with K^0 . For $J_H = \infty$ and $x = 0.3$, $K = 0.77K^0$.^{11,18} It was also shown that¹¹ the reduction factor (0.77) is basically the same for $J_H \geq W/4$. Using this reduction factor, we have $K = 0.037W$ for $x = 0.3$. With $W = 1.6$ eV, $J_H = 1.6$ eV, and $K = 0.037W$, we yield $D(0) = -25$ mV \AA^2 from Eq. (5). The negative value of $D(0)$ implies that the ferromagnetism is not sustainable with these unbiased parameters. It might be possible to have a small

positive $D(0)$ if one includes higher order terms in Eq. (5). Nevertheless, the theoretical $D(0)$ value is too small compared with the measured ones (160–190 mV \AA^2).^{19–21}

Now we turn to the calculation of T_C for $x = 0.3$. For a one-orbital DE model with $J_H = \infty$, the dynamic mean-field (DMF) calculation shows that $T_C^{MF} = 0.02W/k_B$,¹⁰ while Monte Carlo simulations yield $T_C \leq 0.01W/k_B$.^{22,23} This implies that the DMF calculation overestimates T_C by a factor of at least 2 due to the neglecting of spatial fluctuations. For realistic parameters $W = 1.6$ eV, $J_H = W = 1.6$ eV, the DMF calculation obtains $T_C^{MF} = 0.01W/k_B = 180$ K.¹⁰ Considering the fact that the DMF method can overestimate T_C by a factor of at least 2, one has $T_C \leq 100$ K.

It is essential to consider the two-orbital DE model with inclusion of a strong electron-electron correlation. This two-orbital model has been generally accepted to be more relevant to the physics of manganites. From Monte Carlo simulation on this model with realistic onsite Coulomb interactions and J_H , Sheng and Ting²⁴ find that $T_C \approx 0.005W/k_B$ in the case of an anisotropic spin or orbital configuration. This leads to $T_C \approx 90$ K with $W = 1.6$ eV. Therefore, with the unbiased parameters, both the one- and two-orbital DE models predict T_C values, which are about a factor of 4 smaller than the maximum T_C (~ 400 K) observed in manganites.

The above calculations have not taken into account a strong electron-phonon interaction. It was shown that a strong electron-phonon coupling can lead to a substantial suppression on T_C .⁶ Thus the predicted T_C value of about 100 K from the DE model is an upper limit. Furthermore, the DE model along with a strong electron-phonon interaction predicts no isotope shift of T_C ,⁶ in contradiction with the giant oxygen-isotope shifts observed.^{25,26} All these facts lead us to suggest that the DE interaction is not the primary source of the ferromagnetism in doped manganites.

The question is why the DE model cannot quantitatively explain the ferromagnetism in doped manganites. As mentioned above, the DE model implicitly assumes that doped carriers are Mn e_g electrons, which is not the case according to the electron-energy-loss and photoemission spectra.^{7,8} Furthermore, the "constrained" LDA calculation¹⁴ shows a large on-site Coulomb repulsion of about 8–10 eV, in agreement with the photoemission data.⁸ The simple LDA calculation which ignores the strong correlation effect shows a bare plasma frequency of about 1.3 eV for $x = 0.33$ with a distorted structure determined by neutron scattering.²⁷ The bare plasma frequency calculated is much smaller than the one observed in $\text{Nd}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (3.3 eV).²⁸ The large bare plasma frequency observed in this material is consistent with the fact that doped holes reside mainly on the oxygen orbitals with a large bandwidth. The bare plasma frequency of about 3.3 eV for single conduction band (oxygen band) implies a bare K^0 of about 0.24 eV, which gives an upper limit for the K . The electron-phonon interaction with a coupling constant $\lambda \sim 1$ will reduce the K slightly,^{29,30} but can significantly decrease the Drude weight. Optical data indeed show that the K value for $\text{Nd}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ is about 0.2 eV,¹² while the Drude frequency is about 0.57 eV,³¹ much smaller than the bare plasma frequency.

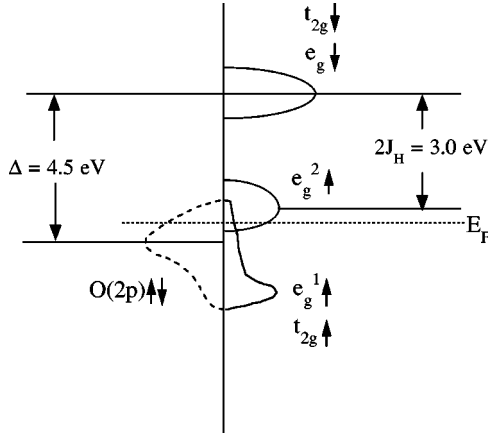


FIG. 1. The schematic band structure for doped manganites ($x < 0.4$) constructed from the LDA + U calculation (Ref. 14). The energy scales are consistent with the optical data (Refs. 12, 31, and 34)

In Fig. 1, we plot a schematic band structure for doped manganites ($x < 0.4$), which can be extracted from the LDA + U calculation. Here we have assumed that the local Jahn-Teller distortions still survive upon doping, but the average magnitude of the distortions decreases, in agreement with the experiments.^{32,33} The doping with a divalent element shifts down the e_g^2 band due to the decrease of the Jahn-Teller distortions. The density of the oxygen holes is equal to x per cell, while the electron carrier density in the majority e_g^2 band is the same as the hole carrier density in the majority e_g^1 band. The electrons in the majority e_g^2 band and holes in the majority e_g^1 band will contribute to the ferromagnetic coupling through double exchange. However, this contribution should be rather small. The current band structure is consistent with the optical transitions at the photon energies of about 1.5 eV, 3.0 eV, and 4.5 eV.^{12,31,34} The optical transition at about 3 eV is related to the transition between the exchange splitted e_g bands shown in Fig. 1.

What is an alternative model for the ferromagnetism in doped manganites? Since doped holes mainly reside on the oxygen orbitals according to EELS and photoemission spectra,^{7,8} there should be a d - p exchange interaction between the oxygen holes and Mn spins. If we consider an oxygen hole (spin 1/2) sitting in between two Mn ions, an exchange interaction between the oxygen and Mn spins (d - p exchange) will lead to a ferromagnetic interaction between Mn spins.³⁵ In this case, the ferromagnetic exchange energy between two Mn spins is $J \propto t_{pd}^4/E_g^3$, where t_{pd} is a hybridization matrix element between the d and p orbitals, and E_g is a charge transfer gap. In addition, the exchange interaction between Mn and oxygen spins is given by $J_{pd} \propto t_{pd}^2/E_g$.³⁵ Using a scaling relation $t_{pd} \propto d^{-4}$ (where d is the bonding length),³⁶ one can see that t_{pd} for the doped manganites ($d = 1.97 \text{ \AA}$) is smaller by 20% than for the cuprate La_2CuO_4 ($d = 1.89 \text{ \AA}$). Furthermore, the charge transfer gap E_g for doped manganites [$\sim 4 \text{ eV}$ (Ref. 34)] is about twice as large as that for La_2CuO_4 [$\sim 2 \text{ eV}$ (Ref. 37)], so one expects that J_{pd} for the former should be smaller by a factor of 2.8 than for the latter where $J_{pd} = 0.17 \text{ eV}$.³⁸ Then J_{pd} is estimated to be about 0.06 eV for doped manganites.

Recently, Alexandrov and Bratkovsky³⁹ have proposed

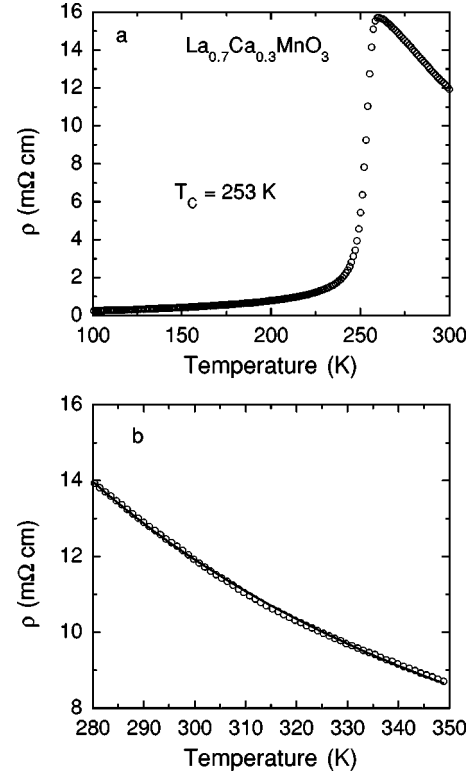


FIG. 2. (a) The temperature dependence of the resistivity of a $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ film over 100–300 K, (b) the resistivity in the paramagnetic state of the film. The metal-insulator crossover temperature T_p is about 260 K and the ferromagnetic transition temperature is about 253 K. The solid line is the fitted curve by Eq. (10).

that, in addition to the d - p exchange interaction, there is a strong electron-phonon interaction that may lead to the formation of small (bi)polaron. In the paramagnetic state, the singlet bipolarons (spin zero) are stable and the ferromagnetic interaction is produced by the thermally excited polarons (spin 1/2). Thus the Curie temperature T_C is self-consistently determined by the polaron density at T_C .

In this model, there are three coupled mean-field equations which can quantitatively explain the observed colossal magnetoresistance (CMR) (Ref. 39) and the isotope shift of T_C .⁴⁰ Here we want to generalize these equations in Ref. 39 to a more realistic case where $T_C \ll W_p/k_B$, where W_p is the polaron bandwidth. For $T_C \leq T \ll W_p/k_B$ and in zero magnetic field, one easily finds that

$$n = 2 \left(\frac{k_B T}{1.05 W_p} \right)^{3/2} \exp(-\Delta/2k_B T) \cosh(J_{pd} S \sigma / 2k_B T), \quad (6)$$

$$m = n \tanh(J_{pd} S \sigma / 2k_B T), \quad (7)$$

$$\sigma = B_S (J_{pd} S m / 4k_B T). \quad (8)$$

Here n is the polaron density per cell, Δ is the bipolaron binding energy, σ and m are the magnetizations of Mn^{3+} ions and holes, respectively, and $B_S(y) = (1 + 1/2S) \coth[(S + 1/2)y] - (1/2S) \coth(y/2)$ is the Brillouin function. The above equations are the same as those in Ref. 39 except that the prefactor of Eq. (6) is different from that of the corre-

sponding Eq. (5) in Ref. 39. Linearizing these equations with respect to m and σ near T_C , and taking $S = 2$, we find that the Curie temperature T_C in zero magnetic field is given by

$$k_B T_C = \frac{J_{pd}^4}{(1.05W_p)^3} \exp(-\Delta/k_B T_C). \quad (9)$$

One should note that Eq. (9) is valid only for the second-order phase transition where the polaron density has no discontinuity at T_C . Furthermore, in order for Eq. (9) to have a physically meaningful solution, it is required that $dT_C/d\Delta < 0$,³⁹ and $k_B T_C \leq J_{pd} \sqrt{x/2}$ (since $n \leq x$). Experimentally, it was found that the ferromagnetic transition is second order when T_C is higher than 250 K.^{41,42} Therefore Eq. (9) should be able to quantitatively explain the T_C value of our $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ film if the equation is really relevant to the ferromagnetism of doped manganites.

Figure 2(a) shows the temperature dependence of the resistivity of our $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ film over 100–300 K. The metal-insulator crossover temperature T_p is about 260 K and the ferromagnetic transition temperature is about 253 K, which corresponds to a temperature where $d\rho/dT$ has a maximum. As shown in Fig. 2(b), the resistivity in the paramagnetic state can be fitted by an equation⁴³

$$\rho = \frac{C}{\sqrt{T}} \exp(E_p/k_B T), \quad (10)$$

where $C = (a_0 h/e^2 \sqrt{k_B}) (1.05W_p)^{1.5}/\hbar \omega_o$, ω_o is the characteristic optical phonon frequency and E_p is an activation energy.

Using $\hbar \omega_o = 74$ meV, a typical value for oxides,⁴⁴ and the value of the fitting parameter $C = 36(4)$ m Ω cm K^{0.5}, we find $W_p = 80(6)$ meV. In addition, the bipolaron binding energy $\Delta = 2E_s$, where E_s is the activation energy deduced from thermoelectric power data.⁴³ For $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ film with a similar T_p , Jaime *et al.*⁴⁵ found that $E_s = 3.80$ meV. Substituting $T_C = 253$ K, $\Delta = 2E_s = 7.6$ meV, and $W_p = 80(6)$ meV into Eq. (9), we get $J_{pd} = 65(4)$ meV, in good agreement with the above estimate. If there were no bipolaron formation and charge ordering in $\text{La}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$, one would have a maximum $T_C = (J_{pd}/k_B) \sqrt{x/2} = 380(24)$ K. Indeed, a maximum $T_C \approx 380$ K has been observed in $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$ with no charge-ordering instability.⁴⁶

In summary, we have provided evidence that the ferromagnetism of doped manganites is not caused by the double exchange interaction. Alternatively, the model based on d - p exchange and strong electron-phonon coupling can quantitatively explain the colossal magnetoresistance,³⁹ the isotope effect,⁴⁰ and the T_C values (this work).

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