

## Low-temperature resistivity in double-exchange systems

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We make a quantum description of the electron low-temperature properties of double-exchange materials. In these systems there is a strong coupling between the core spin and the carriers spin. This large coupling makes the low energy spin waves a combination of ion and electron density spin waves. We also analyze the spin up and down spectral functions of the temperature-dependent quasiparticles of this system. Finally we analyze the low-temperature resistivity of these systems. We find that static hopping amplitude disorder couples with the spin fluctuations and produces a low-temperature resistivity scaling as  $T^{3/2}$  whose magnitude agrees with experimental data.

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Doped perovskite manganites have attracted much attention lately, since they undergo a ferromagnetic-paramagnetic transition accompanied by a metal-insulator transition.<sup>1</sup> The double-exchange (DE) mechanism<sup>2</sup> plays a major role to explain this magnetic transition. In the DE picture, the carriers moving through the lattice are strongly ferromagnetically coupled to the Mn ion spins producing a modulation of the hopping amplitude between neighboring Mn ions.

A big effort has been done to understand the electron transport properties of these materials at temperatures  $T$  near the critical temperature.<sup>3</sup> Nevertheless, the low  $T$  properties of these systems are poorly understood. At low  $T$ , the resistivity  $\rho$  is generally fitted to  $\Delta\rho \sim T^2$ .<sup>4,5</sup> Although the  $T^2$  behavior is similar to that produced by electron-electron interaction, the coefficient of the  $T^2$  term is about 60 times larger than the expected for electron-electron scattering<sup>5</sup> and therefore this mechanism has been ruled out. Another source for this  $T^2$  behavior is single spin wave scattering. However, in DE materials only one spin channel is metallic and single spin wave scattering processes are prohibited. Two spin wave scattering gives a  $T^{9/2}$  increase,<sup>6</sup> clearly slower than the experimental data. In Ref. 7 the  $T$  dependence of the resistivity is attributed to polaron coherent motion. At low  $T$  this process gives a good fit of the resistivity. But this model requires the existence of soft optical modes and polarons at almost zero  $T$ . This situation can arise in low critical  $T$  manganites but is unlikely to happen in high critical  $T$  materials.

Furukawa<sup>8</sup> proposes an unconventional one-magnon scattering in half metals, which gives  $\Delta\rho \sim T^3$ . We believe this dependence is not right, because the inverse of the imaginary part of the electron self-energy and not the transport scattering time is calculated; an appropriated calculation taking into account the fractional loss of forward velocity<sup>9</sup> will give a  $T^{7/2}$  dependence of the resistivity. On the other hand, one magnon scattering is calculated in first order perturbation theory in the Hund's coupling,  $J_H$ , between the electron and ion spins. This coupling is assumed to be infinity in the DE model so that perturbation theory in this parameter is not valid (in particular it would imply a zero lifetime for the carriers). Wang and Zhang<sup>10</sup> assume that the minority spin electron states are localized and obtain  $\Delta\rho \sim T^{3/2}$ , however again in this approach the scattering time is proportional to  $J_H$ . To summarize, a complete understanding of low  $T$  resis-

tivity remains elusive. There is not agreement on the actual power of  $T$  that fits  $\rho$  but it is clear that the magnitude of the increase of  $\rho$  with  $T$  is large.

In this work we study some low  $T$  properties of DE materials. We analyze the spin waves and we find that, to minimize the energy, the ion spin waves (ISW) and the electron spin density waves (ESDW) become coupled in a composite spin wave (CSW) with energy independent of  $J_H$ . We also analyze the spin up and down spectral weights of the  $T$  dependent quasiparticles. These spectral functions do not depend on  $J_H$ . Finally the low  $T$  resistivity of the DE systems is analyzed. We find that the zero  $T$  hopping amplitude disorder is coupled with the low energy magnons in the system and produces a resistivity that increases as  $T^{3/2}$ . The magnitude of this term of the resistivity is in agreement with experimental data.

The Hamiltonian describing Mn oxides is

$$\hat{H} = -t \sum_{i \neq j, \sigma} C_{i, \sigma}^+ C_{j, \sigma} - \frac{J_H}{S} \sum_{i, \sigma, \sigma'} C_{i, \sigma}^+ \boldsymbol{\sigma}_{\sigma, \sigma'} C_{i, \sigma'} \mathbf{S}_i. \quad (1)$$

Here  $C_{i, \sigma}^+$  creates an electron at site  $i$  and with spin  $\sigma$ ,  $t$  is the hopping amplitude between nearest-neighbor sites, and  $\mathbf{S}_i$  is the ion spin at site  $i$ . The ions are located on a single cubic lattice with lattice parameter  $a_0$ . The second term in Eq. (1) describes the ferromagnetic coupling which forces the electron spins to be parallel to the ion spins. The electrons lower their energy by hopping from site to site. And to minimize the kinetic energy, the ion spins become ferromagnetically coupled. The ground state (GS) of the system is half metallic: the system is metallic for one spin orientation but it is insulator for the opposite orientation.

Writing the electrons operators as Bloch operators  $C_{\mathbf{k}\sigma}^+$  and representing the ion spins in terms of Holstein-Primakoff bosons,<sup>11</sup> in first order in the  $1/S$  expansion the Hamiltonian gets the form

$$\begin{aligned} H = & \sum_{\mathbf{k}\sigma} \varepsilon_{\mathbf{k}} C_{\mathbf{k}\sigma}^+ C_{\mathbf{k}\sigma} - J_H \sum_{\mathbf{k}\sigma} \sigma C_{\mathbf{k}\sigma}^+ C_{\mathbf{k}\sigma} \\ & - J_H \sqrt{\frac{2}{SN}} \sum_{\mathbf{q}, \mathbf{k}} (b_{\mathbf{q}}^+ C_{\mathbf{k}\uparrow}^+ C_{\mathbf{k}+\mathbf{q}\downarrow} + b_{\mathbf{q}} C_{\mathbf{k}+\mathbf{q}\downarrow}^+ C_{\mathbf{k}\uparrow}) \\ & + \frac{J_H}{NS} \sum_{\mathbf{k}, \mathbf{q}_1, \mathbf{q}_2, \sigma} \sigma b_{\mathbf{q}_1}^+ b_{\mathbf{q}_2} C_{\mathbf{k}-\mathbf{q}_1, \sigma}^+ C_{\mathbf{k}-\mathbf{q}_2, \sigma}. \quad (2) \end{aligned}$$

Here  $N$  is the number of sites in the system,  $\varepsilon_{\mathbf{k}} = -2t \sum_{\alpha} \cos(k_{\alpha} a_0)$ , is the electron energy spectrum and  $b_{\mathbf{q}}^{\dagger}$  creates an ISW with momentum  $\mathbf{q}$ , which decreases the  $z$  component of the total ion spin by unity. In Eq. (2) the sum of momenta is restricted to the first Brillouin zone. In the DE case ( $J_H \rightarrow \infty$ ), the GS of this Hamiltonian is ferromagnetic, with all the electron spins and core spins parallel, pointing up in the  $z$  direction. The ground state energy per electron is  $E_0 = -J_H + E_{KE}$ , being  $E_{KE} = (1/N_e) \sum_{\mathbf{k}}^{\text{occ}} \varepsilon_{\mathbf{k}}$  and  $N_e$  the number of electrons.

*Composite spin waves.* We are interested in the low energy ( $\sim t$ ) spin excitations of the Hamiltonian (2). A rotation of an electron spin costs an energy of the order of  $J_H$ , and there are not low-lying single particle spin excitations. Also the creation of an ISW costs an energy of the order of  $J_H$ . Therefore, the only low-energy spin excitations are collective modes, such that the energy is minimized by coherently distributing the momentum and the spin loss among a large number of electrons and core spins. The low energy mode will be a linear combination of an ISW and an ESDW, being the latter defined by the operator

$$a_{\mathbf{q}}^{\dagger} = \frac{1}{\sqrt{N_e}} \sum_{\mathbf{k}} C_{\mathbf{k}+\mathbf{q}\downarrow}^{\dagger} C_{\mathbf{k}\uparrow}. \quad (3)$$

The form and the energy of the excitation are obtained by diagonalizing the matrix

$$\begin{pmatrix} \langle |a_{\mathbf{q}}[H, a_{\mathbf{q}}^{\dagger}]| \rangle & \langle |b_{\mathbf{q}}[H, a_{\mathbf{q}}^{\dagger}]| \rangle \\ \langle |a_{\mathbf{q}}[H, b_{\mathbf{q}}^{\dagger}]| \rangle & \langle |b_{\mathbf{q}}[H, b_{\mathbf{q}}^{\dagger}]| \rangle \end{pmatrix}, \quad (4)$$

where the expectation value is obtained in the ferromagnetic GS. Using Hamiltonian (2) and in the  $J_H \rightarrow \infty$  limit the eigenvectors for this matrix are

$$\Lambda^+(\mathbf{q}) = b_{\mathbf{q}}^{\dagger} + \sqrt{\frac{N_e}{2SN}} a_{\mathbf{q}}^{\dagger}, \quad (5)$$

$$\Xi^+(\mathbf{q}) = \sqrt{\frac{N_e}{2SN}} b_{\mathbf{q}}^{\dagger} - a_{\mathbf{q}}^{\dagger}, \quad (6)$$

which correspond to the following energies:

$$\omega(\mathbf{q}) = -\frac{E_{KE}}{3S} \frac{N_e}{N} \sum_{\alpha} \sin^2\left(\frac{q_{\alpha} a_0}{2}\right), \quad (7)$$

$$\omega_1(\mathbf{q}) = 2J_H + J_H \frac{N_e}{NS} + 2\frac{NS}{N_e} \omega(\mathbf{q}). \quad (8)$$

For comparison with phonons these two modes represent acoustic and optical spin waves.  $\omega(\mathbf{q})$  is proportional to  $t$ , and does not depend on the Hund's coupling  $J_H$ . At long wavelengths  $\omega_1(\mathbf{q}) = \rho_s q^2$ , being  $\rho_s = -E_{KE} N_e / (12SN) a_0^2$  the spin stiffness.  $\omega(\mathbf{q})$  is a gapless Goldstone mode reflecting the spontaneous breaking of rotational symmetry. Its quadratic dispersion in  $\mathbf{q}$  reflexes the O(3) symmetry of the underlying Hamiltonian.

The expression obtained for the low energy mode is equal to that obtained previously by using second order perturba-

tion theory in  $1/S$ .<sup>12-15</sup> The operator  $\Lambda^+(\mathbf{q})$  acting on the ferromagnetic GS creates a symmetric combination of an ISW and an ESDW, in such a way that at each place the expectation value of the core spin and the electron spin are parallel; this is the reason why the energy scale of this excitation is  $t$ . We call the excitation created by  $\Lambda^+(\mathbf{q})$  a composite spin wave (CSW).

The operator  $\Xi^+(\mathbf{q})$  creates an antisymmetric combination of an ISW and an ESDW. This collective mode has an energy above the Stoner continuum.

The two modes,  $\Lambda^+(\mathbf{q})$  and  $\Xi^+(\mathbf{q})$ , are equivalent to those found in diluted magnetic semiconductors.<sup>16-18</sup> In semiconductors the coupling between the carriers and the Mn is antiferromagnetic and the high energy mode occurs at an energy below the Stoner continuum.

*Finite temperature quasiparticles.* To eliminate in Eq. (2) the terms linear in the magnon operator, which should not appear in the low energy physics in the  $J_H \rightarrow \infty$  limit, we make a canonical transformation of the Hamiltonian,  $H \rightarrow H' = \exp(-U)H \exp(U)$ , with<sup>14</sup>

$$U = J_H \sqrt{\frac{2}{SN}} \sum_{\mathbf{k}, \mathbf{p}} \left( \frac{C_{\mathbf{k}\uparrow}^{\dagger} C_{\mathbf{k}+\mathbf{p}\downarrow} b_{\mathbf{p}}^{\dagger}}{\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}+\mathbf{p}} + 2J_H} - \text{H.c.} \right). \quad (9)$$

In the infinite Hund's coupling limit and for large core spin, the low energy part of  $H'$  takes the form

$$H' = \sum_{\mathbf{k}} (\varepsilon_{\mathbf{k}} - J_H) C_{\mathbf{k}\uparrow}^{\dagger} C_{\mathbf{k}\uparrow} + \frac{1}{4NS} \sum_{\mathbf{k}, \mathbf{k}', \mathbf{q}} (\varepsilon_{\mathbf{k}} + \varepsilon_{\mathbf{k}'} - 2\varepsilon_{\mathbf{k}+\mathbf{q}}) C_{\mathbf{k}\uparrow}^{\dagger} C_{\mathbf{k}'\uparrow} b_{\mathbf{q}}^{\dagger} b_{\mathbf{k}+\mathbf{q}-\mathbf{k}'}. \quad (10)$$

The transformed spin wave operator is now

$$e^{-U} b_{\mathbf{q}}^{\dagger} e^{+U} \approx b_{\mathbf{q}}^{\dagger} - [U, b_{\mathbf{q}}^{\dagger}] = \Lambda^+(\mathbf{q}), \quad (11)$$

which describes the coherent oscillation of the spins of the core and itinerant electrons and coincides with the operator which creates a CSW. The transformed quasiparticle operator takes the form

$$d_{\mathbf{k}}^{\dagger} \propto C_{\mathbf{k}\uparrow}^{\dagger} - \frac{1}{\sqrt{2NS}} \sum_{\mathbf{p}} b_{\mathbf{p}} C_{\mathbf{k}+\mathbf{p}\downarrow}^{\dagger}, \quad (12)$$

which accounts for the fact that the spin of the carriers follows the core spins. The quasiparticles have a  $T$  dependent spectral weight in spin up and spin down electron states:

$$A_{\uparrow,(\downarrow)}(\mathbf{k}, \omega) = \frac{1}{2\pi} (1 \pm [1 - \delta m(T)]) \delta(\omega - \varepsilon_{\mathbf{k}}). \quad (13)$$

In the above expressions  $\delta m(T) = 1 - M(T)/M(0) \sim T^{3/2}$  and  $M(T)/M(0) = 1 - 1/M(0) \sum_{\mathbf{q}} n_{\mathbf{q}}$  is the relative magnetization suppression due to thermal CSW excitations.

From the spectral function we see that the quasiparticle, which has its spin aligned with the fluctuating ion spins, will, at finite  $T$ , be a spin up state with probability  $[1 - \delta m(T)/2]$  and a spin down state with probability  $\delta m(T)/2$ . The appearance of a spin down shadow band<sup>19</sup> at energies  $-J_H$  is due to the thermal excitation of low energy

long-wavelength CSW. The relative electron spin polarization and ion spin polarization have the same value and scale as  $T^{3/2}$ . Note that the spectral weight does not depend on Hund's coupling  $J_H$ . The weight of the quasiparticles in spin up and spin down electron states is important to explain much of the temperature dependence of the magnetoresistance of magnetic tunnel junctions.

*Low-temperature resistivity: intrinsic scattering.* A CSW modifies the value of the hopping amplitude although it does not modulate spatially the value of  $t$ . The size of the Brillouin zone is not modified by the presence of a CSW. This implies that an electron is not scattered by a single CSW; the electron creation operator evolves continuously from  $C_{\mathbf{k}}^+$  to  $d_{\mathbf{k}}^+$ . The reason for this behavior is that in the  $J_H \rightarrow \infty$  limit, and in the adiabatic approximation, the electron spins follow instantaneously the core spin fluctuations. The adiabatic approach is based in the fact that the core spin fluctuates at frequencies related only to the temperature which is assumed to be much smaller than the hopping amplitude.

The absence of one-magnon scattering in the  $J_H \rightarrow \infty$  limit is clear in the canonical transfer Hamiltonian Eq. (10). The only source of scattering is a two-magnon process<sup>6</sup> which produces an inverse scattering time proportional to  $T^{9/2}$  and therefore a low-temperature resistance also proportional to  $T^{9/2}$ .<sup>6</sup> Moreover, with appropriate parameters this contribution to the electron scattering only predicts an increase of the resistivity of 0.5  $\mu\Omega$  cm at  $T=100$  K, 200 times smaller than the experimental value.<sup>5</sup>

The presence of CSW's also produces self-energies. A quasiparticle with momentum  $\mathbf{k}$  gets a self-energy,

$$\Sigma_{\mathbf{k}} = \frac{1}{2SN} \sum_{\mathbf{q}} n_{\mathbf{q}} (\varepsilon_{\mathbf{k}+\mathbf{q}} - \varepsilon_{\mathbf{k}}) \xrightarrow{T \rightarrow 0} -\frac{1}{12} \frac{N}{S_T} C \left( \frac{KT}{\rho_s} \right)^{5/2} \varepsilon_{\mathbf{k}},$$

where  $C = (1/4\pi^2) \int_0^\infty [u^{3/2}/(e^u - 1)] du \approx 0.045$  is a constant. The thermally activated CSW's reduce the average transfer integral and lead to a decrease of the carriers bandwidth and therefore to a renormalization of the effective mass,  $m^*$  increases as  $\sim T^{5/2}$ . The change in the bandwidth affects the electronic transport properties of the system. By using the Drude formula, a change in the electron mass implies also a change in the resistivity of the system.<sup>20</sup> However for realistic values of the parameters we obtain that at low temperatures the change in the effective mass is very small (less than 0.5 percent at 50 K), and therefore this effect cannot explain the increase in the resistivity found experimentally.

*Low-temperature resistivity: scattering by impurities.* The doped perovskite manganites have a rather high zero temperature residual electronic resistivity. This implies that the scattering of carriers by impurities and imperfections is very strong. We analyze now whether the electron-impurity scattering can produce a temperature dependent scattering time. There are two sources of disorder in the tight-binding formalism, diagonal and nondiagonal. Diagonal disorder appears as fluctuations in the diagonal of the Hamiltonian matrix with respect to its average value, zero in our case. The diagonal disorder is described by a perturbation  $V_d = \sum_{i,\sigma} \varepsilon_i C_{i,\sigma}^+ C_{i,\sigma}$  with  $\sum_i \varepsilon_i = 0$ . This perturbation commute

with the unitary transformation  $U$  and therefore it is not coupled with magnons. Hence, the scattering time coming from this perturbation does not depend on temperature. This approach is valid for weak disorder.

The other kind of disorder in the tight-binding Hamiltonian is the nondiagonal, i.e., deviations of the values of the hopping amplitude with respect to its average value  $t$  ( $\delta t_{ij}$ ). In real perovskites these zero temperature fluctuations in the hopping amplitude are due to Jahn-Teller distortions and to the dispersion in the value of the ionic radius of the doping atoms. The latter effect can produce rather large dispersions in the values of the hopping amplitude.<sup>21</sup> Nondiagonal disorder is described by a perturbation to the tight-binding Hamiltonian

$$V_{nd} = \sum_{i \neq j, \sigma} \delta t_{i,j} C_{i,\sigma}^+ C_{j,\sigma}, \quad (14)$$

with  $\sum \delta t_{i,j} = 0$ . The important point here is that  $V_{nd}$  does not commute with the unitary transformation  $U$ ,

$$V_{nd} \rightarrow V'_{nd} = V_{nd} - [U, V_{nd}], \quad (15)$$

where the second term mix electrons with magnons and has the following form in reciprocal space:

$$\begin{aligned} [U, V_{nd}] = & \frac{1}{2S} \sum \delta t_{i,j} e^{i(\mathbf{k}-\mathbf{k}'+\mathbf{q}-\mathbf{q}')\mathbf{R}_i} \times (2e^{-i\Delta(\mathbf{k}'+\mathbf{q}')} \\ & + 2e^{i\Delta(\mathbf{k}+\mathbf{q})} - e^{-i\Delta\mathbf{k}'} - e^{i\Delta\mathbf{k}} - 2e^{i\Delta(\mathbf{q}-\mathbf{q}'-\mathbf{k}')} \\ & - 2e^{i\Delta(\mathbf{q}-\mathbf{q}'+\mathbf{k})}) C_{\mathbf{k}\uparrow}^+ C_{\mathbf{k}'\uparrow} b_{\mathbf{q}}^+ b_{\mathbf{q}'}. \end{aligned} \quad (16)$$

Here  $\Delta$  runs over the nearest neighbors of the Mn locations  $\mathbf{R}_i$ . In this expression the coupling of the carriers and the magnons through nondiagonal disorder appears clearly. From this equation we evaluate the contribution of  $V_{nd}$  to the temperature dependence of the inverse scattering time. The lowest orders in temperature are

$$\begin{aligned} \frac{1}{\tau_{nd}} = & \frac{2\pi}{\hbar} \frac{18\overline{\delta t^2}}{S^2} \sum n_{\mathbf{q}} (1+n_{\mathbf{q}'}) \\ & \times \delta(\varepsilon_{\mathbf{k}'} - \varepsilon_{\mathbf{k}} + \omega(\mathbf{q}') - \omega(\mathbf{q}')), \end{aligned} \quad (17)$$

where  $\overline{\delta t^2} = (1/6N) \sum_{i,j} (\delta t_{ij})^2$  is a measure of the nondiagonal disorder. Assuming that the residual resistivity is only due to nondiagonal disorder, an estimation of  $\overline{\delta t^2}/t^2 \sim 0.2$  is done for  $\rho_0 \sim 100$   $\mu\Omega$  cm. Low critical  $T$  manganites are more disordered leading to a larger  $\overline{\delta t^2}$  than that for those with large critical  $T$ . Since for small wave vectors the magnon energies are much smaller than the electron energies involved, we can neglect the magnon energies in the delta function and consider that the scattering is nearly elastic,  $\varepsilon_{\mathbf{k}'} \approx \varepsilon_{\mathbf{k}}$ . It is important to note that as we are considering a two magnon process, an electron can emit a magnon without being a hot electron. With this, in the quasielastic approximation, the low-temperature inverse scattering time has the form

$$\frac{1}{\tau_{nd}} = \frac{36\pi}{\hbar} \delta t^2 N(E_F) \left( \frac{\delta m}{S} + (\delta m)^2 \right), \quad (18)$$

$N(E_F)$  being the density of states at the Fermi energy. From the temperature dependence of  $\delta m$  we see that the nondiagonal disorder perturbation  $V_{nd}$  produces an inverse scattering time and therefore a resistance which has terms that increase with temperature as  $T^{3/2}$  and  $T^3$ :

$$\Delta \rho_{nd} = \frac{m^*}{ne^2} \frac{36\pi}{\hbar} \delta t^2 N(E_F) \frac{0.0587}{S^2} \times \left( \left( \frac{a_0^2 T}{\rho_s} \right)^{3/2} + 0.0587 \left( \frac{a_0^2 T}{\rho_s} \right)^3 \right). \quad (19)$$

Here we have used the Drude expression for the electrical resistivity and  $n$  is the carrier density. The two terms arise from  $n_{\mathbf{q}} + n_{\mathbf{q}'} n_{\mathbf{q}'}$  in Eq. 17. The two different powers of  $T$  are directly related with the number of momentum integrations to be done. For realistic values of  $\rho_s$ , the  $T^{3/2}$  term is more important than the  $T^3$  term up to temperatures higher than the critical one. In order to compare with experiments we calculate the magnitude of the  $T^{3/2}$  term. For appropriate values of the parameters,  $t=0.2$  eV,  $x=0.3$ , and  $m^* = 2.5m_0$ , we obtain  $\Delta \rho_{nd} \approx 0.226 \delta t^2 / t^2 T^{3/2} \mu\Omega \text{ cm K}^{-3/2}$ .

Typical experimental values for  $\Delta \rho$  are  $10 \mu\Omega \text{ cm}$  at  $T = 50$  K. Quantitative agreement of the experimental  $\Delta \rho$  and the calculated  $T^{3/2}$  term is achieved with a zero temperature dispersion in the hopping amplitude of  $\delta t^2 / t^2 \sim 0.12$  which is a reasonable value. We therefore conclude that the coupling of the nondiagonal disorder and the spin waves can account for the low-temperature dependence of the electrical resistivity in doped perovskites manganites.

In conclusion, we have studied the low energy and low  $T$  electronic properties of DE systems. We have obtained that the low energy spin excitations are composite spin waves: a linear combination of ion and electrons spin waves. We have also analyzed the spectral function of the  $T$  dependent quasiparticles. Finally, we have studied the low-temperature resistivity of double-exchange systems. We find that scattering by static disorder in the hopping amplitude couples with spin fluctuations and produces a resistivity which increases as  $T^{3/2}$ . The magnitude of this resistivity is in agreement with experimental data.

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