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Study of magneto-resistivity in manganite systems

G C Rout[a]*, N Parhi[b] and S N Behera[cᵥ]

*Condensed Matter Physics Group, Govt. Science College, Chatrapur-761 020, Orissa, India

[b]P G Department of Physics, M P C College (Autonomous), Baripada-757 001, Orissa, India

[c]Institute of Physics, Sachivalaya Marg, Bhubaneswar-751 005, Orissa, India

E-mail : gcr@iopb.res.in

Abstract : We consider here the band Jahn-Teller distortion in dynamic limit and the ferromagnetism in the same itinerant e_g band of the manganese atom in presence of an external field. The electron Green's functions are calculated by Zubarev's technique for the colossal magnetoresistance system in presence of electron-phonon interaction. The Jahn-Teller distortion (e) and the magnetization (m) are calculated self consistently. The resistivity for the system is calculated from the electron relaxation time in Drude model taking into account the phonon frequencies in the Einstein limit. The temperature dependence of magneto-resistance is studied for different lattice coupling parameters and external magnetic field strengths. It is observed that the temperature variation of resistivity explains experimental results qualitatively.

Keywords : Electron-phonon interaction, colossal magnetoresistance, metal-insulator transition.

PACS Nos. : 71.38.-k, 75.47.Gk, 71.30.+h

1. Introduction

$La_{1-x}Sr_xMnO_3$ is recognized to be the simplest among many Colossal magnetoresistance (CMR) manganese oxides [1–4]. The parent material of CMR is the $LaMnO_3$ which is a Mott-insulator. The phase diagram of this system is already complicated, in particular near $x = 0.1$, where several phase boundaries of insulator-metal, lattice symmetry as well as magnetic structures are entangled on the temperature (T)-doping concentration (x) diagram [5]. In other words, if we understand correctly this complexity in the phase diagram, we might find a possible clue to the CMR mechanism in the manganese oxides.

The charge ordering (CO), lattice distortion and magnetic properties are known to couple with each other around $x = 0.125(1/8)$. An unusual phase transition occurs from ferromagnetic metal to insulator transition by either applying magnetic field or lowering temperature [5–7]. On cooling from high temperatures, the crystal symmetry undergoes from pseudo-cubic to orthorhombic at $T_H = 291$ K. Then ferromagnetic long range order occurs below $T_c = 172$ K. The crystal symmetry changes again

from orthorhombic to another pseudo-cubic at $T_L = 145$ K accompanied with the jump in magnetic structure. Simple 3D-isotropic ferromagnetic order appears for $T_L < T < T_c$. Below $T_L = 145$ K, very small antiferromagnetically ordered component appears. The electric resistivity jumps by as much as 10 times at $T_L = 145$ K, the conductivity below T_c is rather metallic. The most fascinating feature is that the resistivity jumps more in enhanced manner under the applied field above T_L. It means that at certain temperatures in this temperature range $T_L < T < T_c$ there appears a striking phenomenon of the large positive magneto-resistance effect in the CMR manganese oxides. Further more, the crystal undergoes the transition to be less distorted below T_L.

It is widely known that the conventional double exchange scenario cannot explain the unique magneto transport phenomena observed in the metallic phase near the Mott-insulator. One of the additional ingredients required is the lattice degree's of freedom in Mn^{3+} ion. The electronic configurations in the ion is represented as $(t_{2g})^3(e_g)^1$, where one electron occupies the doubly

degenerate e_g orbital. Therefore, the ion has the orbital degree's of freedom as well as spin and charge ones.

The unusual properties of the CMR effect are regarded as a cooperative phenomena associated with a structural change due to a tiny atomic displacement, competing with magnetic interactions and charge fluctuations between different valencies of manganese cations. In this respect, Mn^{3+} orbitals show Jahn-Teller (J-T) effect [8] and hence the degeneracy can easily be lifted by lowering the crystal symmetry costing the lattice distortion energies. Therefore it is believed that the CMR should be the result of the local distortion which is often defined as 'polarons'. The itinerary of such polarons gives rise to the conduction. It is well recognized that the J-T interaction plays a crucial role to determine the super exchange interactions in these transition metal oxides. In this communication, we address the magneto-resistivity in presence of dynamic band Jahn-Teller effect.

2. Formalism

We consider here the itinerant e_g electrons of Mn atoms in presence of the induced ferromagnetic order in the same e_g electrons, although the strong Hund's rule coupling aligns the localized spins in the t_{2g} state. In the simplified model under consideration, we assume the existence of a direct Heisenberg type exchange interaction between the e_g electrons to be responsible for induced ferromagnetic state. Furthermore, calculations are performed in the mean field approximation accounting for the magnetic exchange interaction in the Ising limit. Since the e_g band is doubly degenerate a band Jahn-Teller distortion is invoked to remove the degeneracy. An external magnetic field drastically reduces the magnetic state and drives the system from insulating ferromagnetic state to a metallic ferromagnetic state. The aim of the calculation is to study the interplay between the transitions from the paramagnetic to the ferromagnetic state and the structural transition induced by the band Jahn-Teller distortion. The different terms in the model Hamiltonian are detailed below [9,10].

We consider a model system in which the e_g electrons of the Mn atoms interact with the lattice as well as between themselves.

$$H_c = \sum_{k,\sigma} \left(\epsilon_0(k) - \mu - B\sigma \right) \left(c_{1k\sigma}^\dagger c_{1k\sigma} + c_{2k\sigma}^\dagger c_{2k\sigma} \right), \quad (1)$$

where H_c represents the non-interacting electronic energy in a two-fold degenerate (e_g) band with single particle energy $\epsilon_0(k)$ and chemical potential μ. The operators $c_{\alpha k\sigma}^\dagger$ $(c_{\alpha k\sigma})$ being the creation (annihilation) operator of the electron in the band α of spin σ and energy $\epsilon_0(k)$ $(\alpha = 1,2$; is the orbital index). Here, B is the external magnetic field acting in the spin direction σ where $\sigma = +1$ for spin up and $\sigma = -1$ for spin down.

The Heisenberg exchange interaction between the spins of the e_g electrons at the i-th and j-th atomic sites in the Ising limit can be written as

$$H_M = \frac{JM}{2} \sum_{\alpha,k} \left[c_{\alpha k\uparrow}^\dagger c_{\alpha k\uparrow} - c_{\alpha k\downarrow}^\dagger c_{\alpha k\downarrow} \right] \quad (2)$$

where the ferromagnetic order parameter is $M = -J <S>$ and J is the effective interaction energy. The electron hopping is present in a ferromagnetic metallic system. The exchange energy per atom is comparable to the kinetic energy and it attains the magnitude of ~ 1 eV by interacting with all other electrons in the system.

The coupling between the electron density in a degenerate electron band and static elastic strain mode is of the form,

$$H'_{JT} = Ge \sum_{k,\sigma} \left[c_{1k\sigma}^\dagger c_{1k\sigma} - c_{2k\sigma}^\dagger c_{2k\sigma} \right], \quad (3)$$

where G is the strength of the electron lattice interaction in presence of a tetragonal distortion e. Therefore, the interaction of the electrons in the degenerate conduction band with the lattice acts as external perturbation to the free electrons in the band which tries to create a population difference between the two bands. As the population difference increases, the strain builds up in the system resulting in a splitting of the single degenerate band into two with band energies $\epsilon_{1,2}(k) = \epsilon_0(k) \pm Ge$, provided there is a net gain in the electronic energy due to the redistribution of electrons between the split sub bands in comparison to the cost in the elastic energy. Under this situation, there are two non-degenerate bands separated by a gap of magnitude $2Ge$.

The H''_{JT} describes the coupling of phonons to the Jahn-Teller distorted e_g conduction bands

$$H''_{JT} = Ge \sum_{k,q,\sigma} G(q) \left[c_{1k+q\sigma}^\dagger c_{1k\sigma} - c_{2k+q\sigma}^\dagger c_{2k\sigma} \right] A_q \quad (4)$$

with $G(q)$ as the dynamic electron phonon coupling constant and $A_q = b_q + b^\dagger_{-q}$.

$$H_p = \sum \omega_p b^\dagger_q b_q,$$ (5)

where ω_q is the energy of a phonon, $b^\dagger_q$ and b_q the creation and annihilation operators of the phonos.

Hence, the manganite system exhibiting Jahn-Teller static lattice distortion and ferromagnetism in presence of external applied magnetic field can be described by the total hamiltonian H given by

$$H = H_C + H_M + H'_{JT} + H''_{JT} + H_p$$ (6)

3. Calculation of electron Green's functions

The double time single particle electron Green's functions are calculated by equations of motion method of Zubarev [11] for the total Hamiltonian in eq. (6). The electron Green's functions are defined as

$$A_1(k,w) = \left\langle\left\langle c_{1k\uparrow};c^\dagger_{1k\uparrow}\right\rangle\right\rangle_\omega,$$

$$A_2(k,w) = \left\langle\left\langle c_{1k\downarrow};c^\dagger_{1k\downarrow}\right\rangle\right\rangle_\omega,$$

$$A_3(k,w) = \left\langle\left\langle c_{2k\uparrow};c^\dagger_{2k\uparrow}\right\rangle\right\rangle_\omega,$$

$$A_4(k,w) = \left\langle\left\langle c_{2k\downarrow};c^\dagger_{2k\downarrow}\right\rangle\right\rangle_\omega.$$ (7)

The four coupled Green's functions are solved and written

$$A_\alpha(k,\omega) = \frac{1+E_\alpha(0)}{2\pi(\omega - E_\alpha(k) - \Sigma_\alpha(k-q,\omega))},$$ (8)

where

$$E_\alpha(0) = \frac{G(0)[(\omega - E_\alpha(k)) < A_0 > +\omega_0 < B_0 >]}{(\omega - E_\alpha(k))^2 - \omega_0^2}$$ (9)

for $\alpha = 1$–4. Here the terms $G(q)$, A_q, B_q, w_q in eq. (9) are respectively, the electron-phonon coupling, phonon displacement, phonon momentum and phonon energy for phonon wave vector (q).

The self energies Σ_α $(k - q, \omega)$ of the electrons are given by

$$\Sigma_\alpha(k-q,\omega) = \sum_q G^2(q) \cdot \frac{S_\alpha}{(\omega - E_\alpha(k-q))^2 - \omega_q^2}$$ (10)

with $S_\alpha = (\omega - E_\alpha(k-q))(1+2v_q) + \omega_p(1-2n_\alpha)$. Here, v_q and n_α are the Bose-Einstein distribution function and the

electron occupation numbers respectively. They are given by $v_q = \left[e^{\omega_q/k_B T} - 1\right]^{-1}$ and $n_1 = n_{1k-q\uparrow}$, $n_2 = n_{1k-q\downarrow}$, $n_3 = n_{2k-q\uparrow}$, $n_4 = n_{2k-q\downarrow}$.

$$E_1(k-q) = \epsilon_0 (k-q) - \mu - B + Ge + \frac{JM}{}$$

$$E_2(k-q) = \epsilon_0 (k-q) - \mu + B + Ge - \frac{JM}{}$$

$$E_3(k-q) = \epsilon_0 (k-q) - \mu - B - Ge + \frac{JM}{2}$$

$$E_4(k-q) = \epsilon_0 (k-q) - \mu + B - Ge - \frac{JM}{}$$ (11)

4. Calculation of resistivity

The resistivity $\rho(\omega,B)$ of the manganite system can be found from the Drude formula

$$\rho(\omega,B) = \frac{m}{ne^2\tau}$$ (12)

where m and e the mass and charge of the electron, n being the electron number density. The electron relaxation time due to the electron-phonon interaction is given by $\tau^{-1} = \Sigma_\alpha \tau_\alpha^{-1}$. The τ_α can be calculated from the imaginary part of the self-energy of the four Green's functions $A_\alpha(k,\omega)$ with $(\alpha = 1$–$4)$ i.e.

$$\tau_\alpha^{-1} = -\mathrm{Im}\,\Sigma_\alpha(k-q,\omega).$$ (13)

Hence, the resistivity $\rho(\omega, B)$ for frequency ω and external magnetic field B reduces to

$$\rho(\omega,B) = \rho_0 \tau_0 \sum_q G^2(q) \sum_{j=1}^{4} F_j(q),$$ (14)

where

$$F_j(q) = \frac{C_{j1} D_{j2} - D_{j1} C_{j2}}{C_{j2}^2 + D_{j2}^2}$$ (15)

with

$$C_{j1} = [(\omega - E_j(k-q))(1+2v_q) + \omega_q(1-2n_j)],$$

$$D_{j1} = \eta(1+2v_q),$$

$$C_{j2} = (\omega - E_j(k-q))^2 - \omega_q^2 - \eta^2,$$

$$D_{j2} = 2\eta(\omega - E_j(k-q)).$$ (16)

For simplicity, the resistivity is calculated by taking phonons in the Einstein model and n_j's in half filling band condition.

The magneto-resistance of the system is defined as

$$\frac{\Delta\rho}{\rho(0)} = -\frac{\rho(B)-\rho(0)}{\rho(0)}, \qquad (17)$$

where $\rho(B)$ and $\rho(0)$ are the resistivity at magnetic field B and zero magnetic fields respectively. The magneto-resistance is calculated numerically for different model parameters using the self-cosnistent values of magnetization (M) and the static J-T distortion (e) present in e_g band. The expressions for M and e are given below.

The magnetization in the conduction band of the manganite system is defined as

$$M = -Ng_L\mu_B\sum(n_{\alpha\uparrow}-n_{\alpha\downarrow}), \qquad (18)$$

where N, g_L and μ_B are the number of atoms per unit volume, Lande g-factor and the Bohr magneton of the electron of the manganse atome respectively with $\alpha = 1$ to 2.

The occupation numbers $n_{1\uparrow}$, $n_{1\downarrow}$, $n_{2\uparrow}$, $n_{2\downarrow}$ are defined by

$$n_{1\uparrow} = \sum_k \left\langle c_{1k,\uparrow}^\dagger ; c_{1k,\uparrow} \right\rangle,$$

$$n_{1\downarrow} = \sum_k \left\langle c_{1k\downarrow}^\dagger ; c_{1k,\downarrow} \right\rangle,$$

$$n_{2\uparrow} = \sum_k \left\langle c_{2k,\uparrow}^\dagger ; c_{2k,\uparrow} \right\rangle,$$

$$n_{2\downarrow} = \sum_k \left\langle c_{2k\downarrow}^\dagger ; c_{2k,\downarrow} \right\rangle. \qquad (19)$$

For simplicity of calculation, n_α's are calculated taking only static J-T effects neglecting the dynamic part from the Hamiltonian given in eq. (6). The reduced magnetization $m = M/Ng_L\mu_B$ is found to be

$$m = \sum[f(\beta E_1)-f(\beta E_2)+f(\beta E_3)-f(\beta E_4)], \qquad (20)$$

where $\beta = (k_BT)^{-1}$ and the Fermi function in general is

given by $f(y) = 1/(e^y + 1)$. The sum over the momenta of the electrons in the conduction band is replaced by the integration over energy of the electron $\epsilon_0(k)$ with integration limit from $-W/2$ to $+W/2$ as

$$\sum_k = \int_{-W/2}^{+W/2} N(\epsilon_0)d\epsilon_0. \text{ Here, } N(\epsilon_0) \text{ is the model density}$$

of states [12] and $2D$ is the total band width W of the conduction band. To simulate the strong energy dependence of $N(\epsilon_0)$ around the center of the band we take

$$N(\epsilon_0) = N(0)\left[1 - \frac{D^4}{D}\ln\frac{D^4}{\epsilon_0^2}\right] \qquad (21)$$

where $N(0)$ is unperturbed density of state of the free electron and ϵ_0 is the kinetic energy of the conduction band.

The static band Jahn-Teller distortion is given by

$$e = e_0\sum_{k,\sigma}\left[\left\langle c_{1k,\sigma}^\dagger ; c_{1k,\sigma} \right\rangle - \left\langle c_{2k,\sigma}^\dagger ; c_{2k,\sigma} \right\rangle\right], \qquad (22)$$

where e_0 is the static lattice distortion. The reduced lattice distortion ($\tilde{e}$) is given by

$$\tilde{e} = \sum_k\left[\sum_{i=1}^{2} f(\beta E_i) - \sum_{j=3} f(\beta E_j)\right] \qquad (23)$$

The reduced magnetization (m) and the reduced lattice distortion ($\tilde{e}$) are solved self-consistently taking the model parameters of the system. The parameters are scaled with respect to the conduction band width W. They are the

magnetic coupling parameter $g = \dfrac{JN\mu_Bg_L}{W}$ reduced

magnetic field $:b = \dfrac{B}{W}$, the static Jahn-Teller coupling

parameter : $g_1 = \dfrac{G_{e0}}{W}$, dynamic electron-phonon coupling

constant : $g_2 = \dfrac{\tau_0 G^2(0)}{W}$, reduced temperature : $t = \dfrac{k_BT}{W}$,

bare phonon frequency : $p = \dfrac{\omega_q}{W}$ and the renormalized

frequency : $c = \dfrac{\omega}{W}$, life time broadening $s = \dfrac{\eta}{W}$.

5. Results and discussions

In the manganite system $La_{1-x}Sr_xMnO_3$ there occurs a static elastic strain in the electron density in the degenerate e_g electron band. Due to strong Hund's rule coupling the

t_{2g} electrons align ferromagnetically with Curie temperature $T_c \approx 250$ K. The lattice distortion in the form of static Jahn-Teller distortion is expected to suppress or to enhance the Curie temperature and thereby dropping the resistivity below T_c in the concentration range $0.2 \leq x \leq 0.5$. In the present communication, we want to study the effect of J-T distortion on the magnetization, T_c and resistivity. The dimensionless parameters involved in the calculations are the magnetization (m), lattice strain ($\tilde{e}$), static lattice coupling strength (g_1), magnetic coupling strength (g), chemical potential (z), temperature (t), magnetic field (b), phonon vibration energy (ω), phonon wave vector ($\tilde{q}$), electron frequency (c), electron velocity (v), life time broadening (s) and dynamic lattice coupling strength (g_2)

The mean-field magnetization m and the lattice strain e are solved self-consistently and their temperature dependences are shown in the Figure 1. The ferromagnetic
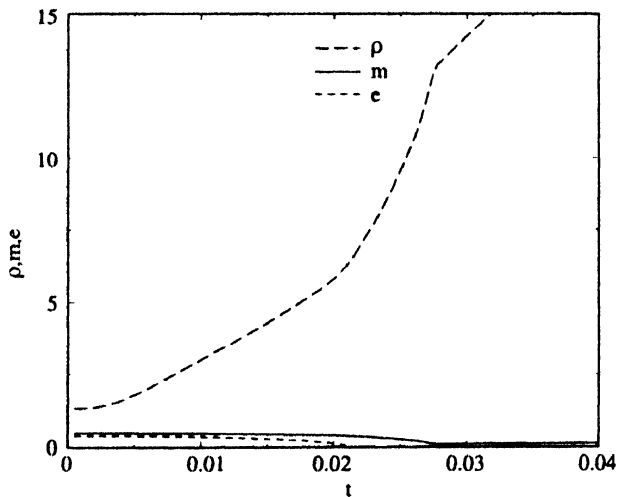

Figure 1. Self-consistent plots of ρ vs t, and $\tilde{e}$ vs t for fixed values of b = 0.0001, g = 0.13995, g_1 = 0.074, p = 0.01, s = 0.02.

(FM) transition temperature is t_c = 0.0275 and the J-T distortion temperature is t_d = 0.02. Here, magnetism exists in distorted phase for $t < t_d$, the pure FM exists for $t_d < t < t_c$ and the paramagnetic phase exists for $t > t_c$. The temperature dependence of the resistivity ρ is shown in the Figure 1. The resistivity decreases with decrease of temperature $t > t_c$ in paramagnetic phase. The decrease of resistivity with decrease of temperature is very rapid in the FM phase $t_d < t < t_c$. Again the resistivity decrease is slower in the distorted phase. The resistivity ρ behavior in manganites shows a close interplay with the magnetization (m). The ρ shows a steep decrease with temperature in accord with the onset of the ferromagnetic magnetization at t_c as observed for $La_{1-x}Sr_xMnO_3$ system [13].

Thus, the observed temperature dependence of the ρ can be interpreted in terms of the carrier scattering by thermal spin-fluctuations. In the distorted phase for $t < t_d$, the magnetization is strongly coupled to the lattice distortion which causes the decrease in spin-fluctuation in the distorted phase. As a result, the ρ shows slight increase with the onset of the lattice distortion. The resistivity remains unaffected by the magnetic coupling g in FM phase and paramagnetic phase [see Figure 2]. However, resistivity shows increase near and below the distortion temperature t_d when the magnetic coupling is increased. The effect of static Jahn-Teller coupling g_1 on the resistivity is shown in Figure 3. The J-T coupling g_1
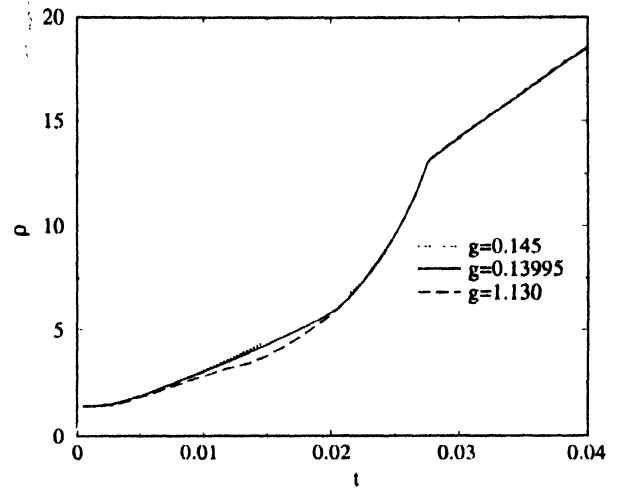

Figure 2. Self-consistent plots of ρ vs t, for fixed values of b = 0.0001, g_1 = 0.074, p = 0.01, s = 0.02 and the different values of g = 0.145, 0.13995, 0.130.
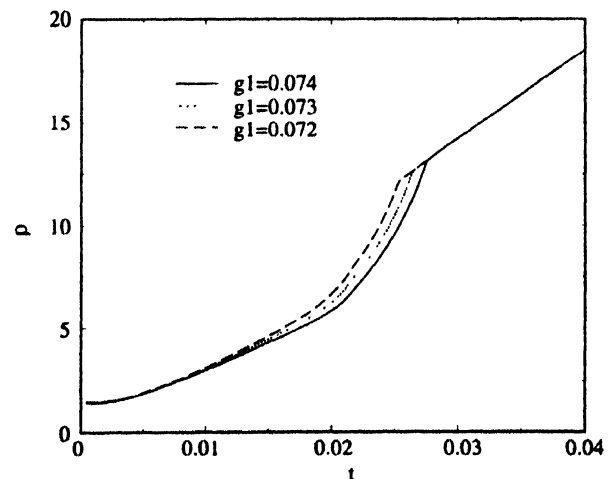

Figure 3. Self-consistent plots of ρ vs. t for fixed values of b = 0.0001, g = 0.13995, p = 0.01, s = 0.02 and the different values of J-T distortion g_1 = 0.074, 0.073, 0.072.

reduces the resistivity in ferromagnetic phase for the temperature range $t_d < t < t_c$. As a result, the system is driven to the ferromagnetic metallic phase.

The external magnetic field suppresses the resistivity in the J-T distorted phase of the system as well as near FM transition temperature t_c as shown in the Figure 4. This agrees qualitatively with experimental observations for the $La_{1-x}Sr_xMnO_3$ compound [1–4,13]. The system stabilizes with maximum doping of $x = 0.20$ as shown in the Figure 5. As doping increases from $x = 0$ to 0.20,
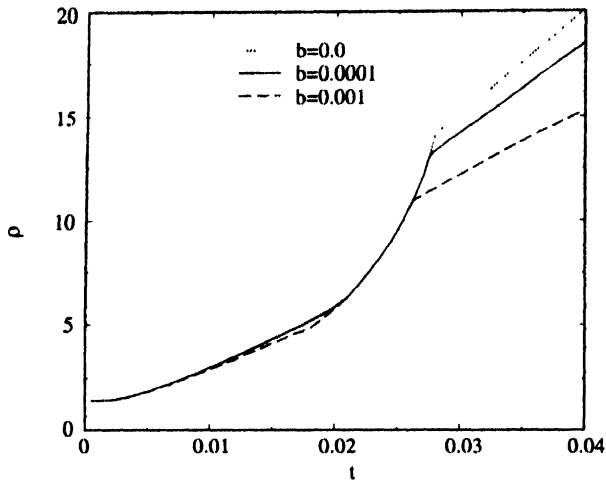

Figure 4. Self-consistent plots of ρ vs t for fixed values of $g = 0.13995$, $g_1 = 0.074$, $p = 0.01$, $s = 0.02$ and the different values of external magnetic field $b = 0.0, 0.0001, 0.001$.
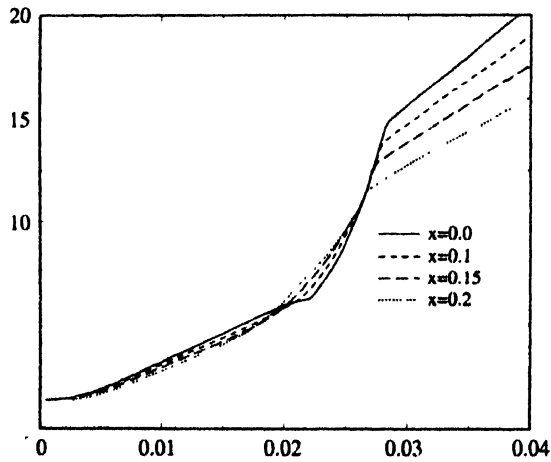

Figure 5. Self-consistent plots of ρ vs t for fixed values of $b = 0.0001$, $g = 0.13995$, $g_1 = 0.074$, $p = 0.01$, $s = 0.02$ and the different values of impurity concentration $x = 0, 0.1, 0.15, 0.2$.

the resistivity is suppressed in the J-T distorted phase for temperatures $t < t_d$. This agrees qualitatively with experimental data on $La_{1-x}Sr_xMnO_3$ [14,15]. The ferromagnetic phase is present in coexistence of lattice distorted phase. In this phase structural phase transition couples to the magnetic transition. In pure FM phase for temperatures $t_d < t < t_c$, the Sr doping increases the resistivity. However, in the paramagnetic phase for $t > t_c$, the resistivity decreases with impurity doping.

The magneto-resistance (MR) means the resistance

change induced by an external magnetic field. The temperature dependence of MR is shown in Figure 6. The MR effect is greater in J-T distorted phase for temperatures $t < t_d$. The higher is the magnetic field, the greater is the MR. The negative MR with magnetic field is observed in the $La_{1-x}Sr_xMnO_3$ system up to an external
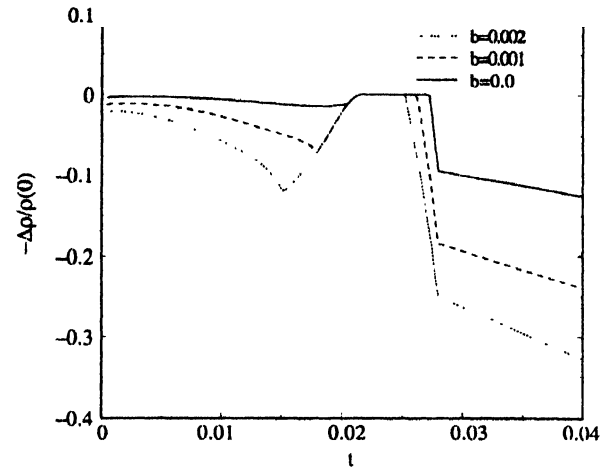

Figure 6. Self-consistent plots of $\frac{\Delta\rho}{\rho(0)}$ vs t for fixed values of $g = 0.13995$, $g_1 = 0.074$, $p = 0.01$, $s = 0.02$ and the different values of external magnetic field $b = 0.00025, 0.001, 0.002$.

field of 15T [3,15]. The magnetic field reduces the resistivity in the distorted phase due to the suppression of the spin-scattering of the e_g carriers which is caused partly by external magnetic field and partly by lattice strain.

6. Conclusion

The manganite system is described by the Hamiltonian consisting by the induced ferromagnetism in the J-T distorted e_g band. The phonon is coupled to the e_g band giving rise to the static and dynamic J-T coupling effect. The lattice strain and the magnetization are calculated self-consistently. The resistivity and the magneto-resistivity are calculated from simple Drude model taking phonon frequencies in the Einstein model. Our model shows a steep decrease of resistance with decrease of temperature at the onset of ferromagnetism. The observed temperature dependence can be explained in terms of the carrier scattering by thermal spin-fluctuations. The large negative magnetoresistivity in the coexistence phase of lattice distortion and magnetization is due to the suppression of the spin scattering of the e_g electrons which is caused partly by external field and partly by lattice strain. Our simple model calculation of resistivity explains the experimental results qualitatively. The present calculation

of magneto-resistivity can be improved by considering the phonon frequencies in Debye model.

References

[1] K Chabara, T Ohono, M Kasai, Y Kanke and Y Kazono *Appl. Phys. Lett.* **62** 780 (1993)

[2] R von Helmolt, J Wecker, B Holzapfel, L Schultz and K Samwer *Phys. Rev. Lett.* **71** 2331 (1993)

[3] Y Tokura, A Urushibara, Y Moritomo, T Arima, A Asamitsu, G Kido and N Furukawa *J. Phys. Soc. Jpn.* **63** 3931 (1994)

[4] S Jin, T H Tiefel, M Mc Cormack, R A Fastnacht, R Ramesh and L H Chen *Science* **264** 413 (1994)

[5] H Kawano, R Kajimoto, K Kubota and H Yoshizuwa *Phys. Rev.* **B53** R14712 (1996)

[6] Y Endoh, K Hirota, Y Murakami, T Fukuda, H Kimura, H Nojiri, K Kaneko, S Ishihara, S Okamoto and S Maekawa (unpublished)

[7] H Nojiri (unpublished)

[8] J Goodenough *Phys. Rev.* **100** 564 (1955); J Kanamori *J. Appl. Phys.* **31** 145 (1960)

[9] N Parhi, G C Rout and S N Behera *Indian J. Phys.* **77A** 153 (2003)

[10] N Parhi, G C Rout and S N Behera *Indian J. Phys.* **77A** 575 (2003)

[11] D N Zubarev *Sov. Phys. Uspekhi* **3** 3 (1960)

[12] H Ghosh, M Mitra, S N Behera and S K Ghatak *Phys. Rev.* **B57** 13414 (1998)

[13] R M Kusters, D A Singleton, D A Keen, R McGreevy and W Hayes *Physica* **B155** 362 (1989)

[14] A Urushibara, Y moritomo, T Arima, A Asamitsu, G Kido and Y Tokura *Phys. Rev.* **B51** 14103 (1995)

[15] Y Moritomo, A Asamitsu and Y Tokura *Phys. Rev.* **B056** 12190 (1997)