Orissa Journal of Physics
ISSN 0974-8202

Vol. 25, No.1
February 2018
pp. 57-62

Tight Binding Model Study of the Electron Specific Heat of Manganese Oxides; $R_{1-x}A_xMnO_3$

D D SAHOO[1] , S PANDA[2†], and G C ROUT[3]

[1]*Dept. of Physics, Mahanga Puspagiri Mahavidyalaya, Erakana, Podamarai, Cuttack-754206, India.*
[2]*Dept. of Physics, Trident Academy of Technology, Chandaka Industrial Estate, Bhubaneswar-751024, India.*
[3]*Condensed Matter Physics Group, Physics Enclave, Plot No-664/4825, Lane 4A, Shree Vihar, Bhubaneswar-751031, India. Email:gcr@iopb.res.in,Mob:09937981694*
[†]*Corresponding author. Email Id: saswatip7@ gmail.com*

Received: 30.11.2017 ; *Revised : 19.12.2017* ; *Accepted : 2.1.2018*

Abstract: We report here a tight binding model for colossal magneto-resistive (CMR) manganites which includes band Jahn-Teller (JT) distortion in conduction band and antiferromagnetic (AFM) Heisenberg interaction in core band in presence of usual Kubo-Ohata type double exchange (DE) model. The Hamiltonian is solved using Zubarev's Green's function technique and the interplay between lattice strain and AFM parameter is studied. Two jumps appear in the temperature dependent specific heat as observed in experiments.

Keywords: Colossal magneto-resistance; Jahn-Teller effect; Magnetization

PACS: 75.47.Gk; 71.70.Ej; 75.25.Dk

1. Introduction

The doped perovskite manganese oxides of the general formula $R_{1-x}A_xMnO_3$ (R- trivalent rare earth ion, A- divalent alkaline earth ion and x-doping concentration) is a strongly correlated system having varieties of electrical, magnetic and structural phases co-existing and competing with each other [1]. Near the magnetic phase transition temperature, it exhibits an important characteristic known as colossal magneto-resistance (CMR). The above said properties make manganese oxides a potential material for modern electronic devices. The undoped manganese oxide is an antiferromagnetic (AFM) insulator. Upon doping it changes its phase from antiferromagnetic to ferromagnetic and insulator to metallic state. Study of charge, orbital and spin orderings of the

manganites helps in understanding the physical mechanisms behind metal-insulator phase transition and CMR effect. Manganites exhibit charge ordering at lower temperatures due to localization of charges which promote insulating behavior [2], whereas above the charge ordering transition temperature the, system goes to metallic state. Another mechanism which promotes insulating behavior is band Jahn-Teller (JT) distortion, which is a structural distortion. It is observed that charge ordering (CO) gives rise to JT distortion. In order to understand the metal-insulator (MI) phase transition and the observed CMR effect in manganites, inclusion of one or more extra mechanisms like charge ordering or JT distortion is required along with double exchange (DE) mechanism.

Rout and co-workers have studied the doped manganese oxide systems using theoretical model calculations. For a CO manganite in presence of ferromagnetism, they have studied the temperature dependent magnetization, magnetic susceptibility, velocity of sound and ultrasonic attenuation, Raman spectra and tunneling conductance spectra [3-7]. For a AFM–CO manganite system, the MI transition is successfully explained through the study of tunneling conductance and specific heat [8]. Further they have considered JT interaction as an extra mechanism for ferromagnetically ordered manganites and studied the magnetic susceptibility [9] and resistivity [10, 11], which explains the CMR effect near FM transition temperature.

In this paper, we propose a tight binding model for the manganites considering JT interaction as an extra mechanism in the presence of AFM spin fluctuation in the core localized electrons in the usual Kubo-Ohata type DE model. We investigate the effect of elastic constant on the temperature dependent lattice strain, AFM order parameter and specific heat.

2. Formalism

The model Hamiltonian for the Manganite system is written as

$$H = \sum_{\alpha,k,\sigma} \epsilon_{\alpha,k,\sigma}\, C^{\dagger}_{\alpha,k,\sigma} C_{\alpha,k,\sigma} - Ge \sum_{\alpha,k,\sigma} (-1)^{\alpha}\; C^{\dagger}_{\alpha,k,\sigma} C_{\alpha,k,\sigma}$$

$$+J \sum_{\alpha,i} \overrightarrow{\sigma_{\alpha,i}} \,.\, \overrightarrow{S_i} \;+ \sum_{\beta,i,j} J_{\beta}\; (i,j)\, \overrightarrow{S_i} \,.\, \overrightarrow{S_j} + \sum_{k,\sigma} \epsilon_{d,\sigma}\, d^{\dagger}_{k,\sigma} d_{k,\sigma} + \frac{1}{2} Ce^2 \qquad (1)$$

Here $C^{\dagger}_{\alpha,k,\sigma}(C_{\alpha,k,\sigma})$ and $d^{\dagger}_{k,\sigma}(d_{k,\sigma})$ represent the conduction and core electron creation (annihilation) operators respectively. The band dispersion energy is $\epsilon_k = -2t_1\left(cosk_x + cosk_y\right) - 4t_2 cosk_x cosk_y$, t_1 and t_2 being the first and second nearest neighbour hopping integrals. The second term

of the Hamiltonian represents Jahn-Teller (JT) interaction in conduction band with static band JT coupling G and lattice strain e. Here $\alpha = 1,2$ represents the two degenerate e_g orbitals of Mn^{3+} ion. The third term of the Hamiltonian represents the double exchange (DE) interaction between spins of the itinerant e_g and localized t_{2g} electrons with DE coupling J. The fourth term describes the antiferromagnetic coupling between the spins of nearest neighbor (NN) and next nearest neighbor (NNN) t_{2g} electrons. The first and second nearest neighbor spin interactions described by J_β (i, j) respectively for β =1,2. The AFM spin coupling is written as $J_H = J_1(cosk_x + cosk_y) + 2J_2 cosk_x cosk_y$, with J_1 and J_2 being the NN and NNN Heisenberg coupling constants respectively. The fifth term of the Hamiltonian describes the kinetic energy interaction of t_{2g} core electrons of Mn^{3+} ions with core level appearing at ϵ_d with respect to Fermi level ϵ_F=0. The last term of the Hamiltonian represents lattice energy with lattice constant C. The above Hamiltonian is solved using Zubarev's Green's function technique and the coupled Green's functions for e_g and t_{2g} electrons are calculated. The various order parameters are calculated from the correlation functions. The lattice strain produced due to band JT distortion in conduction band is $e = \frac{-G}{C}\sum_{\alpha,k,\sigma}(-1)^\alpha \bar{n}^c_{\alpha,k,\sigma}$, the transverse AFM order parameter in core t_{2g} band is $< S^d >= \sum_{k,\sigma} < d^\dagger_{k,\sigma}\, d_{k,-\sigma} >$ and the induced AFM order parameter in conduction band is $< s^c >= \sum_{\alpha,k,\sigma} < C^\dagger_{\alpha,k,\sigma}\ C_{\alpha,k,-\sigma} >$. The temperature dependent entropy (S) and e_g electron specific heat(C_v) are calculated from the free energy of electrons using formula S= $\left(\frac{\partial F}{\partial T}\right)_{\mu,V}$ and $C_v = T\left(\frac{\partial S}{\partial T}\right)_V$. The physical parameters are scaled with respect to the nearest neighbour hopping integral t_1=0.25eV ≃ 2500K. The reduced parameters are NNN hopping integral $\tilde{t}_2 = \frac{t_2}{t_1}$, DE coupling $g_1 = \frac{J}{t_1}$, static JT coupling g=$\frac{G}{t_1}$, the t_{2g} NN-AFM spin coupling $g_{s1} = \frac{J_1}{t_1}$, the t_{2g} NNN-AFM spin coupling $g_{s2} = \frac{J_2}{t_1}$, reduced lattice constant $c_1 = \frac{C}{t_1}$, reduced temperature t = $\frac{k_B T}{t_1}$, reduced chemical potential $u_m = \frac{\mu}{t_1}$.

3. Results and Discussion

The gap equations for lattice strain e and AFM gap of core electrons $<S^d>$ are solved self-consistently for a given set of physical parameters of manganite systems. The temperature dependent lattice strain is plotted in figure 1(a) for different elastic constants c_1=0.0116 to 0.0128. It is observed that the lattice strain exhibits perfect mean-field behaviour with structural distortion temperature t_s occurring at 0.1 to 0.2. This temperature corresponds to the temperature $T_s = t_s \times t_1$ = 250K to 500 K for t_1=0.25ev = 2500K. The lattice strain is continuously suppressed with increase of elastic constant accompanied by corresponding suppression in t_s. It is to note further that there occurs anomaly at low temperatures for higher elastic constants. There occurs AFM order in the core electron as solved above by the self-consistent approach. Due to Hund's coupling the AFM order of the core electrons is induced in the conduction band and the temperature dependent conduction electron AFM order is shown in figure 1(b).

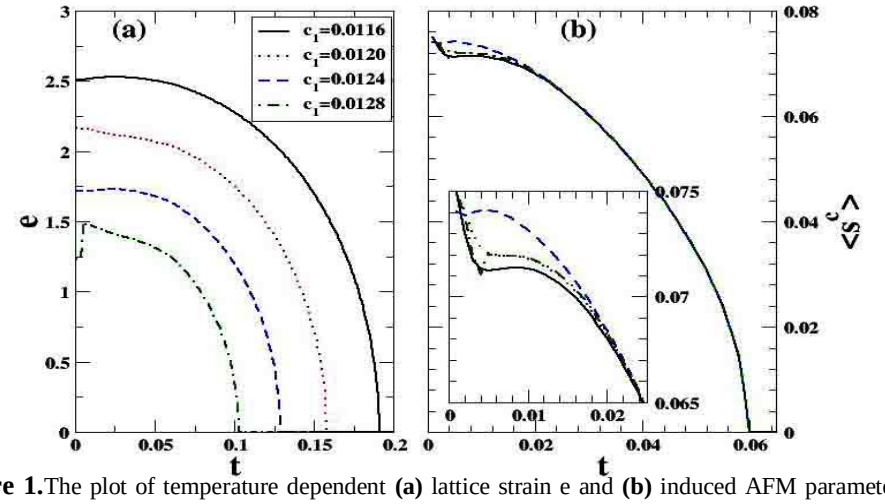

Figure 1. The plot of temperature dependent **(a)** lattice strain e and **(b)** induced AFM parameter $<s^c>$ in conduction band for constant values of g_1=0.5, g=0.185, g_{s1}=0.68, g_{s2}=0.475, t_2=0.0320, u_m=0.2 and different values of elastic constant c_1=0.0116, 0.0120, 0.0124, 0.0128.

The magnitude of the induced AFM order in the conduction band ($<s^c>$) is much reduced in comparison to the core d electron of the manganite system. The $<s^c>$ exhibits mean-field behaviour with Neel temperature occurring at t_N = 0.06 ($T_N \simeq$ 150K) as observed experimentally with T_N =160K for $Pr_{0.6}Ca_{0.4}MnO_3$ system [12] and T_N = 140K for $La_{0.5}Ca_{0.5}MnO_3$ system [13]. However, the elastic constant suppresses the lattice strain and correspondingly enhances the induced $<s^c>$ at low temperatures only. The $<s^c>$ at low temperatures increases with decrease of temperature, then remains nearly constant and then rapidly increases with further

decrease in temperature, exhibiting the low temperature anomaly like that observed in the specific heat measurement of the manganite systems $Pr_{0.6}Ca_{0.4}MnO_3$ [12] and resistivity measurement of the $La_{2/3}Ca_{1/3}MnO_3$ system [14]. Thus we conclude that magnetic energy at low temperatures is enhanced at the cost of lattice energy.

The electron specific heat of the system is numerically calculated from the free energy of the system. The computed temperature dependent specific heat $\widetilde{C_v} = \frac{C_v}{k_B}$ is plotted in figure 2. For a given elastic constant, it exhibits a very

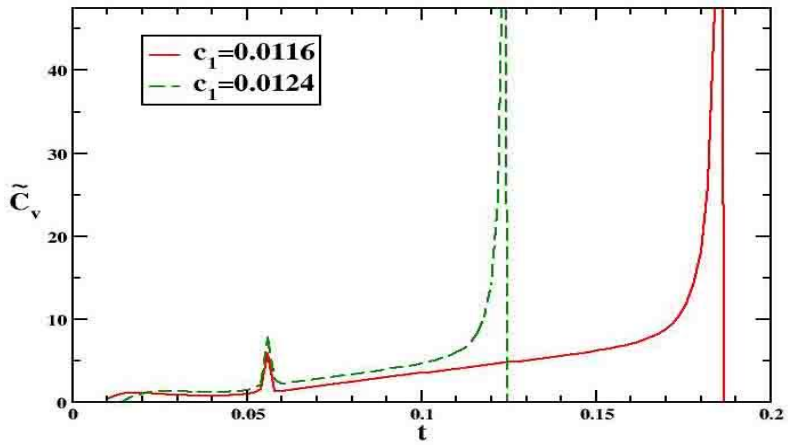

Figure 2. The plot of temperature dependent specific heat for different values of elastic constant c_1=0.0116, 0.0124 and other physical parameters remaining same as in figure 1.

sharp jump at structural distortion temperature t_s. Then it rapidly decreases with decrease of temperature and then monotonously decreases linearly with further decrease of temperature exhibiting metallic character of the manganite system. We observe again a sharp jump at Neel temperature t_N = 0.06 ($T_N \simeq$ 150K) associated with the AFM phase transition of the system. It is to mention that the jump at Neel temperature is much smaller compared to the jump at the robust structural distortion temperature as observed experimentally for the system $Pr_{0.6}Ca_{0.4}MnO_3$ [12]. Below t_N, the specific heat decreases very slowly with decrease of temperature and then exhibits a small flat peak at low temperatures. This peak arises due to the interplay of the elastic energy and AFM as reported by some experimental measurements. The two peak structure in specific heat is reported in specific heat measurements of $Pr_{0.6}Ca_{0.4}MnO_3$ and $La_{0.5}Ca_{0.5}MnO_3$ [12,13] systems. The theoretical investigation of the origin of the anomalous low temperature specific heat is in progress.

4. Conclusion

We have proposed a tight binding model to study the thermal properties of manganite systems taking Heisenberg core electron antiferromagnetic interaction, JT distortion and the DE interaction in the presence of lattice energy. All the calculations are carried out by Zubarev's Green's function technique. We observe low temperature anomaly in lattice strain, induced conduction electron antiferromagnetism and specific heat. Besides this we observe two sharp specific heat jumps, one associated with structural distortion temperature and the other with antiferromagnetic phase transition. The effect of DE coupling, JT coupling, Heisenberg coupling and chemical potential on specific heat will be investigated latter on.

References

[1] J M D Coey and M Jaime, *Rev. Mod. Phys*, **73**, 581, (2001)

[2] James C Loudon, Neil D Mathur and Paul A Midgley, *Nature*, **420**, 797, (2002)

[3] G C Rout, S Panda, and S N Behera, *Physica B*, **404,** 4273, (2009)

[4] G C Rout and S Panda, *Solid State Comm.*, **150**, 613, (2010)

[5] G C Rout and S Panda, *J. Phys. Condens. Matter*, ***21,*** 416001**,** (2009)

[6] G C Rout, S Panda and S N Behera, *J. Phys. Condens. Matter*, **22**, 376003**,** (2010)

[7] S Panda, J K. Kar and G.C. Rout, *J. Magn. Magn. Mater*., **448**, 339, (2018)

[8] S Panda, J K. Kar and G.C. Rout, *Mat. Res. Express*, **3,** 09614, (2016)

[9] G C Rout, N Parhi and S.N Behera, *physica B**, 387,** 259*, (2007)

[10] G C Rout, N Parhi and S.N Behera, *physica B**, 404,** 2315*, (2009)

[11] G C Rout, S Panda, and S N Behera, *J. Phys. Condens. Matter*, **23,** 396001**,** (2011)

[12] M R Lees, O A Petrenko, G Balakrishnan and D M K Paul, *Phys. Rev. B*, **59**, 1298, (1999)

[13] Z Jirak, E Hadova, O Kaman, K Knizek, M Marysko, E Pollert, M Dlouha and S Vratislav, *Phys. Rev. B***, 81,** 024403, (2010)

[14] Y Xu, J Zhang, G Cao, C Jing and S Cao, *Phys. Rev. B*, **73,** 224410, (2006)