Eur. Phys. J. B **31**, 449–452 (2003)
DOI: 10.1140/epjb/e2003-00053-3

THE EUROPEAN PHYSICAL JOURNAL B

Magnetic anisotropy of an extended double exchange model for layered manganites: A Monte Carlo study

Hongsuk Yi[a] and Sangsan Lee

Supercomputing Center, Korea Institute of Science and Technology Information, Yuseong, PO Box 122, Daejeon 305-600, Korea

Received 1st October 2002
Published online 6 March 2003 –

Abstract. The magnetic ground state properties of layered manganites are investigated on the basis of an anisotropic double exchange model using a Monte Carlo technique. The temperature dependence of magnetization and spin-spin correlations are calculated in a highly anisotropic hopping integral t_c/t_{ab} regime. The ferromagnetic ordering temperature (T_c) is suppressed by introducing t_c/t_{ab}, and eventually a layered ferromagnetic structure appears along the c-axis, but there are block-walls. The significant change of magnetic anisotropy is also observed for the antiferromagnetic superexchange integral J_c/t_{ab}. We discuss the connection of these results to the magnetic anisotropy observed in the $La_{1.4}Sr_{1.6}Mn_2O_7$.

PACS. 75.50.Cc Other ferromagnetic metals and alloys – 75.30.Et Exchange and superexchange interactions – 75.30.-m Intrinsic properties of magnetically ordered materials – 72.10.-d Theory of electronic transport; scattering mechanisms

Rapid Note

1 Introduction

Colossal magnetoresistance (CMR) observed in the perovskite manganites [1] has attracted considerable attentions for last decades, because of their potential usages to magnetic sensor or memory technology. For their immediate applications in the electronic devices, great research efforts have been focused on developing new CMR materials operating at low field. The simplest way of searching for convincing candidates with the low-field magnetoresistance consists perhaps in applying the so-called Ruddlesden-Popper series, $A_{n+1}Mn_nO_{3n+1}$ (A=Rare earth and Alkaline earth). Especially, the $n = 2$ member of $(La,Sr)_3Mn_2O_7$ attracts great attentions, because its unique two dimensional nature [2,3]. The layered perovskite manganite is composed of the magnetic $[MnO_2]$ bilayers with the intervening rock-salt $[A_2O_2]$ block, which can be viewed as an intrinsic array of multilayer junction.

The most notable feature of these materials is the occurrence of a very large anisotropic tunnelling magnetoresistance (TMR) [4,5] and the formation of ferromagnetic domains [6,7]. Recent neutron scattering [8] and muon spin rotation [9] experiments for the doped compound of $La_{1.4}Sr_{1.6}Mn_2O_7$ have shown that three dimensional ferromagnetic (FM) coupling evolves near FM ordering temperature $T_c \sim 90$ K, while for temperature well below T_c the magnetic coupling between the adjacent $[MnO_2]$ bilayers is mostly antiferromagnetic (AF). Kimura *et al.* [10] has observed a remarkable enhancement of the interplane TMR effect under the applied hydrostatic pressure, reflecting the field-induced dimensional crossover of the charge-transport. Such a reduced dimensionality is accompanied by the change of the orbital state from the $d_{x^2-y^2}$ to $d_{3z^2-r^2}$ orbital [11] which is different from the cubic ABO_3 manganites. Moreover, the stabilization of the $d_{x^2-y^2}$ orbital causes the FM double exchange interaction within the MnO_2 sheet as well as the AF superexchange interaction between the adjacent bilayer [12].

In this paper, we report anisotropic effects of magnetic ground state of the extended double exchange model for layered manganites with perovskite structure. In order to include the effects of magnetic fluctuations of the localized t_{2g} spins of Mn atoms, we adopt an unbiased computational method on a three dimensional double exchange system by taking into account the highly anisotropic hopping integral t_c/t_{ab} as well as AF superexchange coupling J_c/t_{ab}. It is found that the temperature dependence of magnetization and spin-spin correlations show a ferromagnetic state at intermediate temperature. At low temperature, a layered ferromagnetic structure within the ab-plane runs along the c-axis but there are block-walls. We reveal that the origin of the anisotropic spin correlation at low temperatures is quite different from that near ferromagnetic ordering temperature.

2 Theoretical description

The basis for the theoretical understanding of the cubic ABO_3 manganites is usually the notion of double

[a] e-mail: hsyi@hpcnet.ne.kr

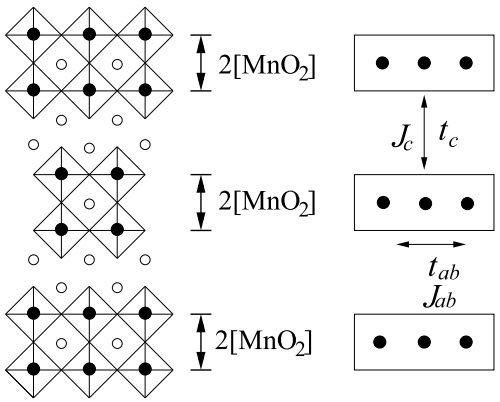

Fig. 1. Schematic structure of $A_3Mn_2O_7$ projected along the [010] direction. The MnO_6 octahedra are connected with a solid line. Solid and open circles represent Mn and A atoms, respectively. Anisotropic hopping (superexchange) integrals t_c (J_c) along c-direction across $[A_2O_2]$ layers and t_{ab} (J_{ab}) in the $[MnO_2]$ bilayers.

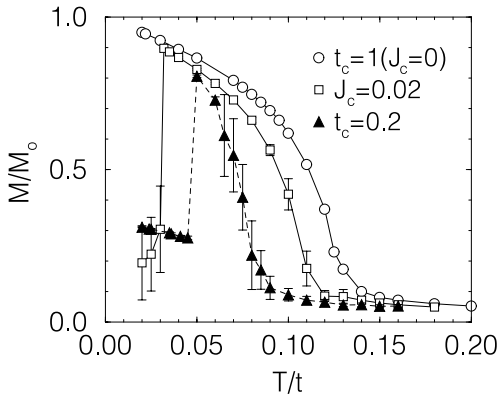

Fig. 2. The temperature dependence of $M = \langle \sum_i S_i^z \rangle$ for different values of hopping integrals t_c/t_{ab} and J_c/t_{ab}.

exchange (DE) [13], which considers the exchanges of electrons between neighboring Mn^{3+} and Mn^{4+} sites with strong on-site Hund's coupling. The hopping amplitude of the electron to the neighboring Mn sites is maximum when the two neighboring t_{2g} spins are parallel, the ferromagnetic metallic state is achieved by gaining the kinetic energy of the conduction electrons. It is straightforward to extend it to an anisotropic layered structure like $La_{2-2x}Sr_{1+2x}Mn_2O_7$ with $x = 0.3$. As shown in the schematic drawing of the $A_3Mn_2O_7$ structure projected along the [010] direction in Figure 1, each plane can be modelled on a $[MnO_2]$ bilayer [17] so that the hopping integral t_c between adjacent $[MnO_2]$ bilayers leads to an effective FM DE coupling of $t_{ll'}{}^c = t\cos(\theta^c_{ll'}/2)$. Besides, we consider the spin disorder scattering within the plane.

Based on these considerations, we propose the following Hamiltonian to describe the spin correlations of the e_g electrons in the layered manganites:

$$\mathcal{H} = -\sum_{\langle ij \rangle}^{\text{intra}} t_{ij}^{ab}\left(c_j^+ c_i + c_i^+ c_j\right) - \sum_{\langle ll' \rangle}^{\text{inter}} t_{ll'}^{c}\left(c_l^+ c_{l'} + c_{l'}^+ c_l\right) + J_{ab}\sum_{\langle ij \rangle}^{\text{intra}} \boldsymbol{S}_i \cdot \boldsymbol{S}_j + J_c \sum_{\langle ll' \rangle}^{\text{inter}} \boldsymbol{S}_l \cdot \boldsymbol{S}_{l'} \quad (1)$$

where $\langle ij \rangle$ and $\langle ll' \rangle$ indicate the nearest neighbor pairs of inplane and interplane sites, respectively. The single-orbital operator c_i^+ creates a spinless conduction electron at site $\boldsymbol{R}_i$. The effective hopping integral can be written as $t_{ij}^{ab} = t_{ab}\cos(\theta_{ij}^{ab}/2)$ and $t_{ll'}^{c} = t_c\cos(\theta_{ll'}^{c}/2)$, where θ_{ij}^{ab} and $\theta_{ll'}^{c}$ are the nearest neighbor relative angles between the Mn^{4+} ions within the ab-plane and the out-of-plane, respectively. J_c and J_{ab} are the nearest neighbor AF interactions between out-of-plane and inplane t_{2g} spins. Here, we consider only a single orbital for the e_g electrons and neglect the effects of the electron-electron correlations. In order to describe the reduce dimensionality of the layered perovskite structure properly, we must adopt the orbital degree of freedom in equation (1). Recent numerical studies, however, show that the single-orbital model is sufficient to understand more complicated magnetic phases such as phase separation [14,15] and magnetic domains [16].

We apply a Monte Carlo integration procedure for the summation over the configuration angles $\{\theta_{\mathbf{i}}, \phi_{\mathbf{i}}\}$ of the t_{2g} spins using a standard Metropolis algorithm [15,18]. The localized t_{2g} spin $\boldsymbol{S_i}$ is treated as a classical spin $\boldsymbol{S_i} = |\boldsymbol{S_i}|(\sin\theta_i\cos\phi_i\hat{x} + \sin\theta_i\sin\phi_i\hat{y} + \cos\theta_i\hat{z})$ with the polar angles $\{\theta_{\mathbf{i}}, \phi_{\mathbf{i}}\}$ characterizing the orientation of the localized spin $\boldsymbol{S}_i$ since the quantum effects can be neglected near room temperature. The azimuthal angle ϕ_i rotates independently since the contribution of the phase factor in the hopping integral to the partition function can be neglected [18]. The actual calculations are performed for three dimensional cubic lattices $L^3 = 6^3$ with periodic boundary conditions in spatial directions. In our Monte Carlo calculation, typically 5×10^3 initial sequences of configurations are discarded for the thermalization processes before the Monte Carlo data collections are made. We actually take $5 \times 10^3 \sim 10^4$ samples for each measurement depending on the numerical accuracies. Unless stated otherwise, we take $t_{ab} = t = 1$ as the unit of energy, and set $J_{ab}/t = 0$ since $d_{x^2-y^2}$ orbital in the $La_{2-2x}Sr_{1+2x}Mn_2O_7$ with $x = 0.3$ facilitates the FM DE interactions within the plane, while interactions between interplanes are dominated by J_c [11]. It means that the layered A-type AF structure prevails over three dimensional FM structure, making a sharp contrast with cubic perovskites like $La_{1-x}Sr_xMnO_3$.

3 Monte Carlo results

Figure 2 shows the temperature (T) dependence of the magnetization, $M(T) = \langle \sum_i S_i^z \rangle$, for $t_c/t_{ab} = 0.2$, 1, and $J_c/t = 0.2$. For the isotropic case marked by open circles, $M(T)$ steeply rises in the vicinity of FM ordering temperature $T_c \approx 0.13t$, and reaches near the saturation value of $M/M_o = 1.0$ with decreasing T. Note that the estimation of $T_c = 0.13t$ corresponds to 300 K if we take

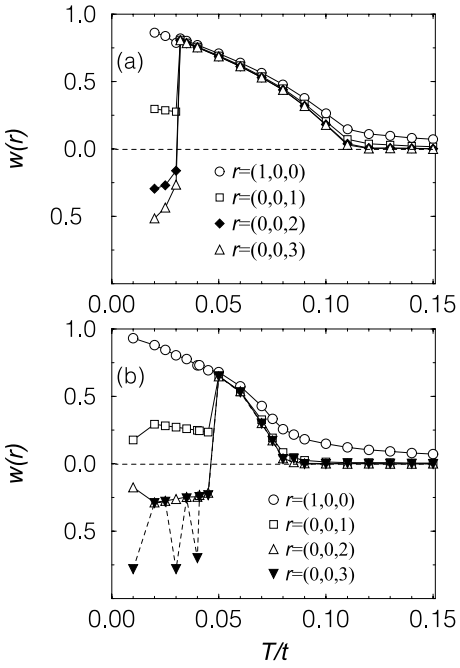

Fig. 3. The temperature dependence of spin-spin correlation function $w(r) = 1/N \sum_i \boldsymbol{S}_i \cdot \boldsymbol{S}_{i+r}$ for four values of $\boldsymbol{r}$ (a) for $J_c/t = 0.02$ and (b) for $t_c/t = 0.2$.

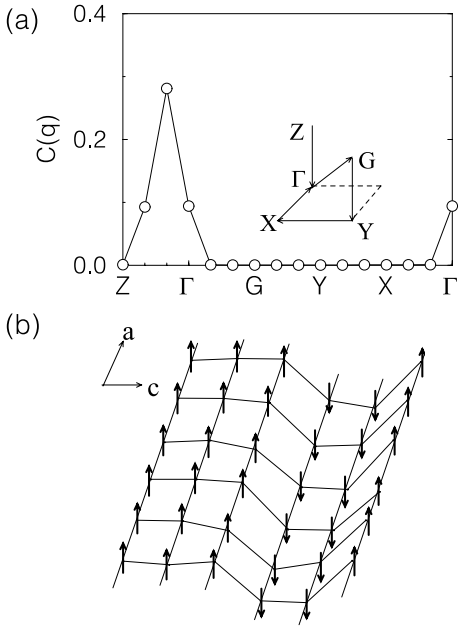

Fig. 4. (a) Spin structure factor $C(q)$ for the localized spins versus momentum for $t_c/t_{ab} = 0.2$ and $T = 0.024$ on a $6\times6\times6$ lattice along the path $Z-\Gamma-G-Y-X-\Gamma$ [$(0,0,\pi)$-$(0,0,0)$-(π,π,π)-$(0,\pi,0)$-$(\pi,0,0)$-$(0,0,0)$]. (b) Snapshot of the localized spin S.

$t = 0.2$ eV [19]. For the case of $J_c/t = 0.02$ and $t_c/t_{ab} = 1$, the applied superexchange coupling greatly reduces the FM ordering temperature in the vicinity of $T_c \approx 0.11$ due to the suppression of the spin disorder scattering. With decreasing T, a sharp drop of $M(T)$ appears near $T^* \approx 0.03$. Even for $t_c/t_{ab} = 0.2$ with $J_c/t = 0$, $M(T)$ marked by triangles begins to develop near $T_c \sim 0.09$ and eventually drops in the vicinity of $T^* \approx 0.05$. The similar behaviors are obtained from the simulation for different values of the t_c/t_{ab} and/or J_c in the highly anisotropic regime. Our finding of the magnetic ground state with the incomplete saturation of M/M_o in the vicinity of T^* is to be an intrinsic properties of the anisotropic double exchange model.

To elucidate the nature of the anisotropic magnetic ground state of the system, we calculate the spin-spin correlation function among the localized t_{2g} spins. As usual, the spin-spin correlation function is defined as

$$w(r) \equiv 1/N \sum_i \boldsymbol{S}_i \cdot \boldsymbol{S}_{i+r}, \tag{2}$$

where N is the number of sites. In Figure 3a, we show T-dependence of anisotropic $w(r)$ for different values of lattice sites $\boldsymbol{r}$, $J_c/t = 0.02$, and $t_c/t_{ab} = 1$. The shortest spin correlation within the ab-plane, *i.e.*, $r = (1,0,0)$, shows a typical ferromagnetic state at low temperature, while the spin correlation along the c-axis shows a drastically drop below $T^* \approx 0.03$ due to the reduced spin scattering in the spin ordered state. Especially, the spin correlations become negative both for $r = (0,0,2)$ and $r = (0,0,3)$. The negative spin correlation is also observed for $t_c/t_{ab} = 0.2$ with $J_c/t = 0$ as shown in Figure 3b. This result implies that the anisotropic hopping integral plays the same role in the magnetic anisotropy of the anisotropic double exchange model as the superexchange interaction.

Fourier transformation of the spin-spin correlation function provides the spin structure factor

$$C(\mathbf{q}) = 1/N \sum_{\mathbf{l},\mathbf{m}} \mathrm{e}^{\mathrm{i}(\mathbf{l}-\mathbf{m})\cdot\mathbf{q}} \langle \boldsymbol{S}_\mathbf{l} \cdot \boldsymbol{S}_\mathbf{m} \rangle, \tag{3}$$

where the momentum $\mathbf{q}$ takes the values $2\pi n/L_r$, with n running from 0 to $L_r - 1$, and L_r denoting the number of sites along the $r = x$, y, or z directions. One of the main results observed here occurs at low temperature where a remarkable magnetic incommensurability appears along the out-of-plane direction with the peak structure developing at $\mathbf{q} = (0,0,\pi/3)$. This behavior is illustrated in Figure 4a where $C(\mathbf{q})$ as a function of momentum wave vector $\mathbf{q} = (q_x, q_y, q_z)$ for $t_c/t_{ab} = 0.2$ at $T = 0.024$ is shown. In our Monte Carlo simulations long-range antiferromagnetic order at $\mathbf{q} = (\pi,\pi,\pi)$ (G-type AF) and at $\mathbf{q} = (0,0,\pi)$ (A-type AF) as well as long-range FM order at $\mathbf{q} = (0,0,0)$ have not been observed as expected. Combining these results with the absence of signals along the $X = (\pi,0,0)$ and $Y = (0,\pi,0)$ directions, we identify the magnetic ground state is a layered ferromagnetic structure within the ab-plane where the symmetry of the spin correlation under lattice $\pi/2$ rotation is conserved, and the neighboring ab-planes are also ferromagnetically coupled but there are block-walls.

A typical spin configuration (snapshot) emerging from our Monte Carlo simulations is shown in Figure 4b at the

same parameters used in Figure 4a. The presence of FM spin domains in the anisotropic double exchange model suggests that tendencies towards magnetic anisotropy in the layered manganites. Similar magnetic anisotropy has been observed in independent runs with different initial conditions for the localized spins. It is observed that the size of magnetic domains in the $La_{1.4}Sr_{1.6}Mn_2O_7$ reaches about 3 μm in the absence of external field [6]. However, the finite size of lattices studied here prevents us from comparing directly with the experimental results.

It is worthwhile to note that the orbital degree of freedom plays a significant role in the magnetic ground state of the layered Mn-oxides [11]. Ishihara *et al.* [12] have suggested that the stabilization of the $d_{x^2-y^2}$ orbital facilitates the FM coupling *via* DE interaction within the $[MnO_2]$ bilayer and causes the AF coupling *via* superexchange interaction between the adjacent bilayers that is differ from the cubic ABO_3 manganites where the $d_{3z^2-r^2}$ and $d_{x^2-y^2}$ orbital are degenerated. Especially, the polarization of the $d_{x^2-y^2}$ orbital in the Mn e_g band enhances the double exchange interactions within a plane, which results in the small value of the t_c/t_{ab}. In this anisotropic regime, the e_g carriers cannot easily hop into the adjacent planes because each FM planes are block-walls along the c-axis. Thus, we speculate that the effects of the increased $d_{x^2-y^2}$ orbital polarization may be implicitly included in the parameter values in our model Hamiltonian.

In summary, we have investigated the anisotropic effects of the extended double exchange model for the layered manganites using the Monte Carlo technique. In the highly anisotropic regime of t_c/t_{ab}, the T-dependence of $M(T)$ and $w(r)$ show a robust ferromagnetic state at the intermediate temperature ($T^* < T < T_c$). With decreasing temperature, the ferromagnetic spin correlation is drastically suppressed due to the reduced spin scattering in the anisotropic spin state. From the analysis of the spin structure factor and snapshot of the localized spins, we found that the layered type ferromagnetic structure within the ab-plane runs along the c direction but there are block-walls. The similar results are obtained for the anisotropic J_c/t_{ab}. Although it is not clear that the orbital polarization plays a significant role in the magnetic anisotropy as suggested in $La_{1.4}Sr_{1.6}Mn_2O_7$, it would be interesting to study the relation between the anisotropic carrier's kinetic energy and the orbital polarization, particularly in their magnetic anisotropy.

We would like to thank N.H. Hur and J. Yu for useful discussions and their valuable contributions.

Rapid Note

References

1. S. Jin, T.H. Tiefel, M. McCormack, R. A. Fastnacht, R. Ramesh, L.H. Chen, Science **264**, 413 (1994)
2. T.G. Perring, G. Aeppli, Y. Moritomo, Y. Tokura, Phys. Rev. Lett. **78**, 3197 (1997); D.N. Argyriou, J.F. Mitchell, J.B. Goodenough, O. Chmaissem, S. Short, J.D. Jorgensen, Phys. Rev. Lett. **78**, 1568 (1997)
3. K. Hirota, Y. Moritomo, H. Fujioka, M. Kubota, H. Yoshizawa, Y. Endoh, J. Phys. Soc. Jpn **67**, 3380 (1998); N. Hur, J.T. Kim, K. Yoo, Y. Park, J. Park, E. Chi, Y. Kwon, Phys. Rev. B **57**, 10 740 (1998)
4. T. Kimura, Y. Tomioka, H. Kuwahara, A. Asamitsu, M. Tamura, Y. Tokura, Science **274**, 1698 (1996)
5. Y. Moritomo, A. Asamitsu, H. Kuwaharaa, Y. Tokura, Nature **380**, 141 (1996)
6. T. Fukumura, H. Sugawara, T. Hasegawa, K. Tanaka, H. Sakaki, T. Kimura, Y. Tokura, Science **284**, 1969 (1999)
7. U. Welp, A. Berger, D.J. Miller, V.K. Vlasko-Vlasov, K.E. Gray, J. F. Mitchell, Phys. Rev. Lett. **83**, 4180 (1999)
8. D.N. Argyriou, J.F. Mitchell, P.G. Radaelli, H.N. Bordallo, D.E. Cox, M. Medarde, J.D. Jorgensen, Phys. Rev. B **59**, 8695 (1999); T.G. Perring, G. Aeppli, T. Kimura, Y. Tokura, M.A. Adams, Phys. Rev. B **58**, R14 693 (1998)
9. R.H. Heffner, D.E. MacLaughlin, G.J. Nieuwenhuys, G.M. Luke, Y. Tokura, Y.J. Uemura, Phys. Rev. Lett. **81**, 1706 (1998)
10. T. Kimura, A. Asamitsu, Y. Tomioka, Y. Tokura, Phys. Rev. Lett. **79**, 3720 (1997)
11. M. Kubota, H. Fujioka, K. Hirota, K. Ohoyama, Y. Moritomo, H. Yoshizawa, Y. Endoh, J. Phys. Soc. Jpn **69**, 1606 (2000)
12. S. Ishihara, S. Okamoto, S. Maekawa, J. Phys. Soc. Jpn **66**, 2965 (1997); R. Maezono, S. Ishihara, N. Nagaosa, Phys. Rev. B **58**, 11583 (1998); K. Hirota, Y. Moritomo, H. Fujioka, M. Kubota, H. Yoshizawa, Y. Endoh, J. Phys. Soc. Jpn **67**, 3380 (1998)
13. C. Zener, Phys. Rev. **82**, 403 (1951); P.W. Anderson, H. Hasegawa, Phys. Rev. **100**, 675 (1955); P.-G. de Gennes, Phys. Rev. **118**, 141 (1960); K. Kubo, N. Ohata, J. Phys. Soc. Jpn **33**, 21 (1972)
14. A. Moreo, S. Yunoki, E. Dagotto, Science **283**, 2034 (1999); A. Moreo, S. Yunoki, E. Dagotto, Phys. Rev. Lett. **83**, 2773 (1999)
15. H. Yi, J. Yu, Phys. Rev. B **58**, 11123 (1998)
16. C. Cuhler, S. Yunoki, A. Moreo, Phys. Rev. Lett. **80**, 845 (1998); E. Dagotto, T. Hotta, A. Moreo, Phys. Rep. **344** 1 (2001), and reference there in
17. Y. Liu, L. Sheng, D. Xing, J. Dong, J. Phys. C **10**, 9747 (1998)
18. S. Yunoki, J. Hu, A. Malvezzi, A. Moreo, N. Furukawa, E. Dagotto, Phys. Rev. Lett. **80**, 845 (1998)
19. H. Yi, N. H. Hur, J. Yu, J. Phys. C **12**, 5453 (2000); H. Yi, N. H. Hur, J. Yu, Phys. Rev. B **61**, 9501 (2000)