New Hamiltonian and Cooper pair's origin of pseudogap and colossal magnetoresistance in manganites

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Abstract

This paper proposes a new Hamiltonian for manganites, and demonstrates quantitatively that the pseudogap and colossal magnetoresistance of manganites come from Cooper pairs, consisting of two oxygen $2p$ holes in MnO_2 plane."

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1. Introduction

The manganites known as colossal magnetoresistive (CMR) manganese oxides continue to attract considerable attention due to the presence of CMR and pseudogap [1-19]. The pseudogap of manganites was observed for the first time by Dessau *et al.* in 1998. Although there were more than ten mechanisms on the pseudogap and CMR, no one mechanism can give quantitative comparisons between experiments and theory, and no one mechanism connects with Cooper pair. CMR and pseudogap have not yet haven widely accepted explanations. Theory falls behind experiment very far, is still in model stage, phenomenological, and have some obvious mistakes.

The goals in this letter are: (i). To propose a new Hamiltonian for the manganites, based on strong correlation; (ii). To prove the Cooper pair's origin of both pseudogap and CMR of manganites by uniform explanations for the 14 key experimental facts; (iii). To give briefly some applications of the pseudogap at room temperature.

2. New Hamiltonian

A widely used models of manganites are one- and two-orbital, in which just one and two orbitals of e_g are considered, respectively (Eqs. (5.7) and (5.11) in Ref. [9]). However, experiments have observed that the itinerant carriers in manganites are doped oxygen $2p$ holes rather than $3de_g$'s [13]. Dagotto thought [9]: "However, adding the oxygen orbitals to the electronic models complicates enormously the theoretical studies, which are already quite difficult even with only Mn sites."

The basic building block of manganites is the MnO_6 octahedron [7]. These octahedrons share their in-plane oxygen atoms, forming two dimensional MnO_2 planes, and in this plane, many effects occur [7]. The Fermi surface in MnO_2 plane has nesting structure [2]. The Mn^{3+} and Mn^{4+} ions are local spins $S = 2$ and $S = 3/2$, respectively [9]. Therefore, in both lattice and electronic structures the manganites and high-Tc cuprates are the same [11].

For the CuO_2 plane of the high-T_c cuprates, the strong correlated Hamiltonian is [17]

$$\begin{aligned} H &= t_\circ \sum_{i,\alpha,s}(d^+_{is}p_{\alpha s} + h.c) + U_d \sum_i n_{di\uparrow}n_{di\downarrow} + U_p \sum_\alpha n_{p\alpha\uparrow}n_{p\alpha\downarrow} \\ &+ V_\circ \sum_{i,\alpha} n_{di}n_{p\alpha} + \epsilon_d \sum_i n_{di} + \epsilon_p \sum_\alpha n_{p\alpha}, \qquad (2.1) \end{aligned}$$

where d^+_{is} and $p^+_{\alpha s}$ create holes on the Cu:d and O:p orbits at sites i and α with spin $s = 1/2$, respectively, n_{di} ($n_{p\alpha}$) is the number operator of d (p) holes, $n_{di} \equiv n_{di\uparrow} + n_{di\downarrow}$, $n_{p\alpha} \equiv n_{p\alpha\uparrow} + n_{p\alpha\downarrow}$, $-t_\circ$ is the hopping integral for the holes between adjacent Cu:d and O:p orbits, and U_p, U_d, and $V_\circ$ are intra- and interatomic Coulomb repulsion on O:p orbits, Cu:d orbits, and between both orbits, respectively. The site index with an alphabetic letter stands for Cu site and that with a Greek letter stands for the O site in the CuO_2 plane. We use x to stand for the hole number in one unit cell of Cu lattice in the CuO_2 plane. At half-filling, $x = 0$, there exists one e_g hole per Cu site, i.e., Cu^{2+}; and for $x > 0$ extra holes go into O:p orbits and Cu^{2+} is stable under doping. If we take that the local spins are 2 or 3/2, then Eq. (2.1) becomes the strong correlated Hamiltonian of the MnO_2 planes.

By treating the first term as a perturbation, Ref. [20] derived an effective Hamiltonian, Ref. [11] made simplification, and the form is

$$\begin{aligned} H &= -\sum_{i\alpha\beta s} T_{\alpha\beta}p^+_{\alpha s}p_{\beta s} + J_K \sum_{i\alpha\beta ss'} \widehat{\mathbf{S}}_i \cdot \overrightarrow{\sigma}_{ss'}p^+_{\alpha s}p_{\beta s'} + J \sum_{ij(i<j)} \widehat{\mathbf{S}}_i \cdot \widehat{\mathbf{S}}_j \\ &+ H_{hole-phonon} + H_{hole-hole} \\ &\equiv H_{Kinetic} + H_{Kondo} + H_{Heisenberg} + H_{hole-phonon} + H_{hole-hole}, \qquad (2.1)' \end{aligned}$$

where the summation over α and β stands for the oxygen sites around i-th Mn^{3+} site with local spin $S = 2$; $p_{\alpha s}$ annihilates $O_{p\sigma}$ hole with spin s=1/2 at site α; $\widehat{\mathbf{S}}_i$ is the local spin operator of Mn^{3+} at site i; $\overrightarrow{\sigma}$ is Pauli matrix vector; and i and j are the nearest neighbors. $J > 0$ and $J < 0$ are AFM and FM Heisenberg Hamiltonian, respectively.

The Hamiltonian in Eq. $(2.1)'$ has many different points in comparison with the until now widely accepted Hamiltonian of manganites in Eqs. (5.6-11) of Ref. [9]): (i). $-T_{\alpha\beta} \simeq t_{\circ}$ is the hopping integral between Mn^{3+} and O^{2-} rather than the hopping integral t between Mn^{3+} and Mn^{3+} [9]; (ii). Our Kondo term represents the interaction between the $2p$ hole and the local spins ($S \equiv 2$) of Mn^{3+} ion, rather than that between the electron of $3de_g$ and Mn^{4+} ion with $S = 3/2$. The serious mistakes of the previous Hamiltonian are: (a). In the previous Kondo Hamiltonian, J_K is substituted by Huns energy J_H. We think that this is a conceptual mistake. According to the basic definition, the Hund energy can only exist in an isolated atom or ion. However, the $3de_g$ electron in the Kondo term of one- nd two-orbital Hamiltonians is itinerant; (b). By the way, if we do not take our Kondo coupling constant $J_K(\approx 0.2)$ eV, and instead take Hund energy $J_H(\approx 2)$ eV, then our numerical simulations give, for example, the pseudogap is 1000 eV! (c). In Eq. $(2.1)'$, the Kondo Hamiltonian is derived rather than a phenomenological term (Refer to Eq. (5.1) of Ref. [9]); (iii). The third term in Eq. $(2.1)'$ can be both FM ($J < 0$) and AFM ($J > 0$) rather than only AFM and a phenomenological term (Refer to Eq. (5.3) of Ref. [9]); If the manganites are in FM state rather than in AFM state, then the "Double-Exchange" FM model gives this term; (iv). The $H_{hole-phonon}$ represents the coupling between $2p$ hole (rather than $3de_g$ electron [9]) and the Jahn-Teller distortions of the local MnO_6 octahedron. This term should also include the breathing model; (v). $H_{hole-hole}$ represents the Coulomb interaction among the $2p$ holes rather than the $3de_g$ electrons.

It is easy to observe that the first three terms in Eq. $(2.1)'$ can cause an indirect exchange interaction between two itinerant $2p$ holes in MnO_2 plane. This indirect interaction is mediated by two nearest neighbour FM or AFM coupling local spins at sites of Mn^{3+} or Mn^{4+} ions (See Fig. 3.2 in Ref.

[11].). This indirect interaction is called two local spin-mediated interaction (TLSMI), and can cause Cooper pairing in both macroscopic superconductive state and pseudogap state, in which the systems do not have macroscopic superconductivity in the MnO_2 plane of the high-T_c cuprates [11].

The first three terms of Hamiltonians in Eq. (2.1)$'$ and Eq. (3.5) in Ref. [11] are the same. Using more than fifty pages, Ref. [11] derives the mathematical expression of TLSMI, obtains the solutions of BCS gap equation, gives the numerical program to calculate pseudogap and pairing probability of individual carrier. We can use all the formulas in Ref. [11], and just in the stage of numerical calculations substitute the values of related parameters of manganites, given in section 3. For $S = 2$ Ref. [11] gives

$$TLSMI = -A\frac{272(cos\theta)^2N''JJ_K^2}{T^2 + B272 \times 64(cos\theta)^2JJ_K^2/\{1 + C20J_K^2\}}, \quad (2.2)\ (See\ (3.74)\ of\ [11])$$

where $A,\ B,\ ,C$ are constants, N'' is determined by the size of the cluster with magnetic order, for long-range magnetic order the size of cluster is infinite, N'' has maximum value 2, the minimum value is $N'' = 1$, and θ is the angle between two nearest neighbour Mn ions in FM coupling case (In AFM case $\theta \equiv 0$.) [11].

The pseudogap functions are of $p-$ and d-wave symmetry for FM and AFM, and give in Eqs. (3.67-68) and Eq. (3.66), respectively [11].

3. Values of parameters of manganites

Of course, manganites are not the high-T_c cuprates. We have to use the values of related parameters of manganites. Fortunatuly, all the values of parameters of manganites can be found. Eqs. (5.26-5.35) of Ref. [11] show that J_K is a function of $U_d, \Delta_{eff}, V_\circ$. $\Delta_{eff} = 1.8\ eV$ and $U_d = 6.8\ eV$ [13]. The hopping integral t between Mn^{3+} and Mn^{3+} is 0.2 - 1 eV [9]. We take $t_\circ = 0.5$ eV. Of course, if the on-site Coulomb repulsion energy $U_d = 6.8$ eV, then the Coulomb repulsion energy between Mn^{3+} and Cu^{2-}, $V_\circ$, will much less than 6.8 eV. Referring to the CuO_2 plane of high-T_c cuprates, we take $V_\circ = 1.0$ eV [11]. $J = 0.05$ eV [9]. Bandwidth is 1.8 eV [2]. (for reference: the effective mass of itinerant hole is $m_{eff} = 0.3m_e$, m_e is the electron mass, average free path is 1.44 nm, Fermi velocity is 0.38c, $U_d = 7\ eV$ [7]. $m_{eff} = 3.3m_e$, the average free path is 2.5 nm [2].)

4. Numerical results and explanations for experiments

4.1. Pseudogap

(1). Figs. 1 and 2 show the experimental data of the pseudogap versus temperature for $La_{0.625}Ca_{0.375}MnO_3$ and $La_{2-2x}Sr_{1+2x}Mn_2O_7$, respectively.

Explanation: Our theoretical cures in Figs. 1 and 2 can quantitatively explain the data well.

(2). Refs. [16,1] reported that for the sample $La_{2-2x}Sr_{1+2x}Mn_2O_7$ with $x = 0.4$ at $T > 126$ K the pseudogap is observed in the entire Fermi surface, and there is no zero pseudogap anisotropy.

Explanation: The theory in section 2 indicates that in FM state the pseudogap is of p-wave symmetry. Our numerical simulations show that for $La_{2-2x}Sr_{1+2x}Mn_2O_7$ with $x = 0.4$ in FM state the minimum and maximum pseudogap is along the $Mn - O$ bond and 45° away from $Mn - O$ bond directions, respectively. The ratio is $\Delta(Mn - O)/\Delta(45^\circ) = 0.358/0.626 \neq 0$.

(3). Ref. [2] observed that the pseudogap of $La_{2-2x}Sr_{1+2x}Mn_2O_7$ with $x = 0.4$ is of d-wave symmetry at 20 K other than p-wave symmetry.

Explanation: There is phase transition from pure FM to coexistence of

FM and AFM at $15 < T < 40$ K in the MnO_2 plane [9]. Our numerical simulations show that in the same values of parameters the d-wave is much easier to occur than the p-wave. This theoretical result can explain the observed d-wave (other than p-wave) pseudogap at $T = 20$ K.

(4). Many different types of magnetic systems can have pseudogap [11,6].

Explanation: Many quite different types of magnetic systems can have, in principle, the same Hamiltonian structure as the first three terms of Eq. $(2.1)'$ [21,1]. So long as a system has the Hamiltonian liking the first three terms in Eq. (2.1) and there are some appropriate values of parameters, then this system will have the pseudogap certainly.

4.2. CMR

(1). The data on the temperature dependence of CMR for single crystal $La_{1.2}Sr_{1.8}Mn_2O_7$ at different magnetic fields are reported by Refs. [15,16,9,13], which is shown in Fig. 3.

Explanation: According to Refs. [11,20,6], the motion of Cooper pairs in pseudogap states is free but ranom. Similar to the high-Tc cuprates [11,20,6], we refer CMR completely to the formation to Cooper pairs in the pseudogap state as well. In our calculations for the temperature dependence of CMR at 0 and 5 T, we use the expression of resistivity

$$\rho_{ab} = \rho_0 + \rho_1[1 - Q(T)], \tag{4.1}$$

where $Q(T)$ represents the probability of one individual carrier to become one carrier of a Cooper pair, ρ_0 and ρ_1 represent residual resistivity and resistivity without Cooper pairs, respectively. Due to that we study CMR in $La_{1.2}Sr_{1.8}Mn_2O_7$, we neglect the small temperature dependence of rho_1. Refs. [11,6] have given the formula of $Q(T)$ (See Eq. (2.405) in Ref. [11].

According to Eq. (2.2), our numerical calculations need the experimental magnetization curves at H = 0, 5 T, which are given by Refs. [15-17].The better fitting between the data and our numerical result in Fig. 3 indicates that CMR is really completely caused by the formation of Cooper pairs in $La_{1.2}Sr_{1.8}Mn_2O_7$.

(2). The experiments found that the necessary condition occurring CMR is that the CMR manganites are in FM state [8,9].

Explanation: In FM state, the more stronger the applied field is $\Rightarrow$ the more stronger the FM order is $\Rightarrow$ the stronger the magnetization is $\Rightarrow$ the stronger the pairing potential TLSMI in Eq. (2.2) is $\Rightarrow$ the larger the number of Cooper pairs is $\Rightarrow$ the larger the (negative) CMR is.

(3). The magnitude of CMR is a linear function of square of magnetization around Curie temperature, where the magnetization is small [16].

Explanation: Eq. (2.2) indicates that the Cooper pairing potential is a function of square of magnetization, $\propto (cos\theta)^2$. The first order approximation of Taylor expansion in $\propto (cos\theta)^2$ is proportional to $\propto (cos\theta)^2$. The number of Cooper pairs should be dependent on TLSMI (Refer to Eq. (2.405) in Ref. [11].), and, thus, CMR is dependent on $\propto (cos\theta)^2$ linearly.

(4). The data in Fig. 3 show that all the interplane resistivities are equal approximately to the inplane resistivities plus about 39 Ωcm, correspondingly.

Explanation: The Cooper pairs are formed only in the MnO_2 plane. However, the Cooper pairs can tunnel from one MnO_2 plane to another nearest neighbor MnO_2 plane. The tunnel process is of resistance, and does not be sensitive to temperature. Thus, we obtain $\rho_c = \rho_{ab} + 39\ \Omega cm$, where 39 Ωcm is the tunneling resistivity.

(5). The experimental data of resistivity versus temperature at different pressures from 1 atm to 6.0 GPa in sample $La_{0.33}Ca_{0.67}MnO_3$ show that the pressure causes CMR as well as magnetic field [18]. The variations of lattice constants a and b in the MnO_2 plane are: a=5.4610 and 5.4043, b=5.4750 and 5.4324 $\AA$ for 1 atm and 5.87 GPa, respectively [18].

Explanations: The larger the pressure is ⇒ the shorter the lattice constant is ⇒ the larger the value of J in Eq. (2.2) is [19] ⇒ the larger the value of TLSMI is ⇒ the larger the number of Cooper pairs ⇒ the less the resistivity is.

(6). For $La_{0.625}Ca_{0.375}MnO_3$ $\rho(T = T_{M-I} = 120\ K)/\rho(T = 100\ K) = 30$ at a definite band width, and if the band width reduces, then $\rho(T = T_{M-I} = 240\ K)/\rho(T = 100\ K) = 1000$ [3]. Ref. [16] observed the effect of band width on resistivity.

Explanation: Our numerical calculations for $La_{1.2}Sr_{1.8}Mn_2O_7$ are as follows. $\rho(T = 126\ K)/\rho(T = 100\ K) = 18,\ 25,\ 31$ for band width $W = 1.8, 1.5, 1.2$ eV, respectively.

(7). CMR can occur in thallium manganite pyrochlores $(Tl_2^{3+}Mn_2^{4+}O_7)$ without double-exchange [13].

Explanation: Our paring potential of Cooper pair TLSMI in Eq. (2.2) is dependent on J, i. e., Heisenberg interaction, and J_K, i. e., Kondo interaction, and is independent of the double-exchange.

(8). Ref. [13] concluded clearly from experimental facts: "Thus, thallium manganite pyrochlores $(Tl_2^{3+}Mn_2^{4+}O_7)$ has neither mixed valence for a double exchange-magnetic interaction nor a Jahn-Teller cation such as Mn^{3+}, which both are known to play an essence role in CMR perovskite materials."

Explanation: Although our new Hamiltonian in Eq. (2.2) can contain Jahn-Teller interaction, in all the above numerical calculations, which give the quantitative explanations for both the pseudogap and CMR, we do not consider the Jahn-Teller term. This fact points out that Jahn-Teller effect for the pseudogap and CMR is not important.

(9). The data for epitaxial film and polycrystalline (3, 14, 24 μm average grain size) of $LaCaMnO$ are that the small grain size leads to (i). High resistivity. (ii). Small CMR [13].

Explanation: (i). The conduction occurs in MnO_2 plane. Therefore, the less the grain's size is, the larger the tunneling resistivity between two grain's MnO_2 planes is. (ii). Eq. (2.2) indicates the Cooper pairing potential TLSMI is proportional to N''. The less the grain's size is $\Rightarrow$ the less the value of N'' is $\Rightarrow$ the less the value of N'' $\Rightarrow$ the less the TLSMI is $\Rightarrow$ the less the number of Cooper pairs $\Rightarrow$ the less the CMR is.

(10). Although the value of the pseudogap in CMR manganites is much larger than that in the high-T_c cuprates, CMR manganites have not macroscopic superconductivity.

Explanation: Ref. [11] proves that the condition of emergence of macroscopic superconductivity is that Josephson coupling energy $E_J(T)$ between Cooper pairs is large enough and [11]

$$E_J(T) \propto \frac{\Delta(T)}{R_n}. \tag{4.2}$$

For manganites, the resistivity without the Cooper pairs, $\rho_{n,manganites}$, is nearly equal to $100 \times \rho_n$, where ρ_n is the resistivity of high-T_c cuprates in normal state. Although the pseudogap $\Delta_{manganites}$ is nearly equal to $10 \times \Delta_{cuprates}$, due to small $E_J(T)$ the maganites never have macroscopic superconductivity. Along this line, it is not impossible that in future one will discovery room temperature macroscopic superconductivity by making a magnetic material with large pseudogap but small resistivity at room temperature .

5. Conclusions

From the similarities in electronic and lattice structures of CMR manganites with the high-T_c cuprates, we infer the pseudogaps and CMR of manganites are caused by the formation of Cooper pairs. This inference on Cooper pair's existence in CMR manganites are further verified by the explanations for the 15 experimental facts about the pseudogap and CMR. Therefore, this paper concludes: (1). The observed *d*- and *p*-wave symmetry pseudogap in FM and AFM regions, respectively, of CMR manganites come from Cooper pairs; (2). The Cooper pair consists of two oxygen $2p$ holes, and exists in the MnO_2 plane; (3). The new Hamitonian in Eq. $(2.1)'$ should become a starting point of theoretical study on CMR manganites other than many Hamiltonians proposed before this paper [9].

6. Applications and advantages of CMR manganites over high-T_c cuprates and semiconductors

We call any body with pseudogap pseudogapbody. Many applications of pseudogapbodies have been given in Refs. [6,11,22]. This section gives only two examples (Refer to Ref. [6]).

(1). A permanent current in a ring made by $La_{0.625}Ca_{0.375}MnO_3$ can be induced by the change of magnetic flux in the ring at room temperature [22,6]. Set the radius of the ring is 0.5 m, and the radius of rings wire is 3 mm. The induction critical current in this ring is $I_c = 2.94 \times 10^8$ A, the inductance is $L = 4.2 \times 10^{-6}$ H, $B_a = LI_c/\pi 0.5^2 = 1600$ T in the opening of the ring, the maximum magnetic pressure between the two rings and at zero distance is $P_{mag} = 1.006 \times 10^1 1$ Pa, and, the maximum magnetic levitation is

0.806×10^{10} kg, and so big value of B_a can storage energy $10\times^{12}$ J/m^3. If we use the high-T_c cuprates to make the same size ring working at 77 K, then the supercurrent is in the ring's surface with thick 100 nm due to Meissner effect [23], and, thus $I_c = 1.96 \times 10^3 << 2.94 \times 10^8$ A. If the high-T_c cuprates are hard superconductors, then $I_c = 1.96 \times 10^5$ A. It is important to note that in magnetic field the d-wave superconductivity of the hig-T_c cuprates and the p-wave superconductivity of CMR manganites are reduced and increased, respectively.

(2). $La_{0.625}Ca_{0.375}MnO_3$ can be used as transmission line (wire or cable), which's effective resistivity is $\rho_{eff} = 10^{-17}$ Ωcm, if the line length is 1000 m. There is no any heat production caused by the voltage applied on the two ends of the line, because the applied voltage is needed just by the acceleration of the free but random Cooper pairs. This super-low resistivity can be verified if you apply a voltage less than critical value 0.1 mV [6].

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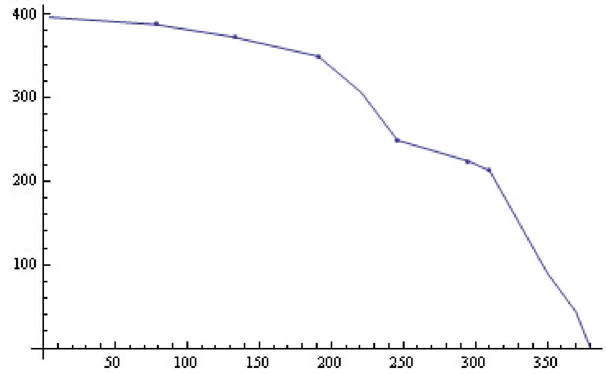

Figure 1: Diagram of pseudogap Δ (meV) versus temperature T (K) for $La_{0.625}Ca_{0.375}MnO_3$. The data come from Ref. [3].

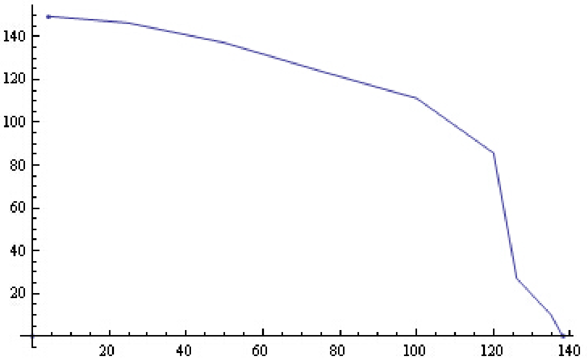

Figure 2: Diagram of pseudogap Δ (meV) versus temperaturen T (K) for $La_{1.2}Sr_{1.8}Mn_2O_7$. The data come from Ref. [5].

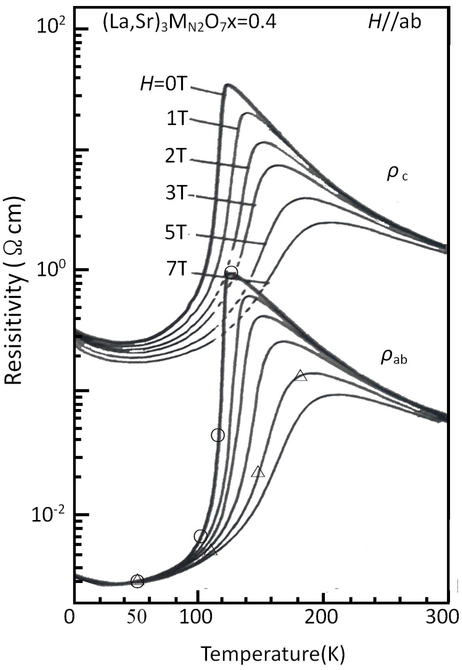

Figure 3: Diagram of temperature dependences of resistivity for single crystal of $La_{1.2}Sr_{1.8}Mn_2O_7$. The experimental curves come from Ref. [15]. An external magnetic field applied parallel to the MnO_2 plane, and no thermal hysteresis are observed [15,16]. ρ_{ab} and ρ_c are the inplane and interplane resistivity. $T_{Curie} = 126$ K at zero field [15,16]. The circles and triangles are our numerical results for 0 and 5 T, respectively.