The Spin Waves in the Double Exchange Interaction Systems

Chong Der HU

Department of Physics, National Taiwan University, Taipei, Taiwan, R.O.C.

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We performed a detailed study of the double exchange interaction. Having rearranged Kubo and Ohata's Hamiltonian [J. Phys. Soc. Jpn. **33** (1972) 21], we found that it can be treated perturbatively for the temperature lower than T_C . The spin wave propagator was calculated and the spin stiffness was computed. It agrees reasonably well with experimental results. We believe that the double exchange interaction is the fundamental mechanism in the perovskite manganites.

KEYWORDS: colossal magnetoresistance, manganites, double exchange

§1. Introduction

In the past few years, we have seen a revival of the study of the perovskite manganites which had been known to have the so called colossal magnetoresistance phenomenon (CMR) long time ago.^{1, 2)} The representative compound is $La_{1-x}A_xMnO_3$ where A represents a divalent element. The rich physics of these kind of compounds has induced earnest and sustained research.³⁾ Owing to different doping concentration x , the conduction electron density and the valence and the magnetic moment of manganese ions vary. Combined with lattice distortion and temperature effect, these compounds exhibit many phases under different conditions, such as antiferromagnetic insulator, ferromagnetic insulator, ferromagnetic metal, charge ordering and paramagnetic insulator. The most prominent feature is the coupling between magnetic phase transition and the metal-insulator transition. Zener⁴⁾ had proposed an intuitively appealing mechanism, the double exchange interaction (DE). With the middle oxygen ion acting as medium, the conduction electrons can hop from a Mn^{+3} ion to a Mn^{+4} ion. Due to the on-site Coulomb repulsion, the electron spin must be parallel to the spins of Mn ions (Hund's rules) and thus creating magnetic coupling. This theory is able to explain the coupling between magnetic and transport properties since the ferromagnetic order favors the motions of conduction electrons. However, Millis *et al.*⁵⁾ argued that the only energy scale in DE is at least an order in magnitude larger than T_C . Their calculation using the infinite-dimension Kondo lattice model also showed that DE is inadequate to explain the behavior of resistivity near T_C . They proposed that the Jahn-Teller effect should be considered. There are also experimental evidences indicating that the electron-lattice interaction is important. For example, the isotope effect on the thermoelectric power⁶⁾ and on T_C .⁷⁾ A possibly related issue is the evidence of polaron formation.⁸⁾ Thus raised question of whether DE is the most important mechanism in perovskite manganites. We think it is.

Since it was proposed by Zener, DE had been fur-

ther developed by Anderson and Hasegawa⁹⁾ and De Gennes.¹⁰⁾ Next major progress was made by Kubo and Ohata¹¹⁾ who used projection operators to project out the lowest-energy states according to Hund's rules and thus, simplified the Hamiltonian. It is not until the recently we have seen many theoretical works.¹²⁻¹⁸⁾ From our point of view, it is worthy to analyze DE carefully and make a detailed calculation to see its implication. Then the results can be a base to incorporate the Jahn-Teller effect. Certainly, as said by Müller-Hartmann and Dagotto,¹⁹⁾ it is crucial to have a proper quantum mechanical treatment of the spin system. Therefore, we developed a fully quantum mechanical calculation for DE. We chose to calculate the spin wave propagator at finite temperature because DE is closely related to the magnetic properties of the system and there are experimental data available.^{20, 21)} The analysis of the Hamiltonian is in §2 and the calculation is in §3. Comparison with experiments and conclusion are in §4.

§2. Hamiltonian

In the CMR manganites, the lowest state of a Mn^{+3} ion is the $S = 2$ state for the total spin of four d-electrons, according to the Hund's rules. The $S = 1$ configuration has a higher energy, by as much as $I = 3$ eV, as shown in band-structure calculations.²²⁾ On the other hand, the band width is around 2.4 eV which gives a hopping energy (to be denoted by t) of 0.2 eV. Therefore, the energy difference between the two spin configurations, I , is the largest energy scale. As we are interested in the spin waves, the low-lying excitations, we can restrict ourselves to the lower spin configuration. By doing this we can work in the large I limit and simplify the Hamiltonian. The price to pay is that we have neglected the processes of the order t/I , such as an electron hopping between Mn^{+3} ($S = 1$) and Mn^{+4} sites. Thus, we use the Hamiltonian developed by Kubo and Ohata:¹¹⁾

$$H = -t \sum_{i,j,\alpha,\beta} c_{i\alpha}^\dagger (P_i P_j)_{\alpha\beta} c_{j\beta}. \quad (1)$$

Here and in all the following equations i and j are nearest

neighbors. Considering the effect of the on-site exchange interaction, the projection operator P is used to project out the states in which the spins of holes anti-parallel to the local spins:

$$P_i = \frac{S - \mathbf{S}_i \cdot \boldsymbol{\sigma}}{2S + 1}. \quad (2)$$

Within the Hamiltonian, the manganese ions can be Mn^{+4} , which has spin $3/2$, or Mn^{+3} which has spin $S = 2$. At a Mn^{+4} site, there is a hole whose spin is set to be antiparallel to the local spin $S = 2$ by the projection operator. Equation (1) describes the hoppings of the holes between Mn^{+3} and Mn^{+4} sites. At finite temperatures, the spins of manganese ions are not always aligned, hence, there exist spin-up and spin-down holes. In the process of hopping, the spins of holes can be flipped though the total spin is conserved.

The Hamiltonian can be expanded as

$$H = \frac{-t}{(2S + 1)^2} \sum_{i,j,\alpha,\beta} c_{i\alpha}^\dagger [S^2 + \mathbf{S}_i \cdot \mathbf{S}_j - (SS_{iz} + SS_{jz} - \frac{1}{2}S_{i-}S_{j+} + \frac{1}{2}S_{i+}S_{j-})\sigma_z - (SS_{i-} + SS_{j-} - S_{iz}S_{j-} + S_{i-}S_{jz})\frac{\sigma_+}{2} - (SS_{i+} + SS_{j+} + S_{iz}S_{j+} - S_{i+}S_{jz})\frac{\sigma_-}{2}]_{\alpha\beta} c_{j\beta}. \quad (3)$$

It seems that there is only one energy scale, namely t . (The energy scale of the spin wave is $t/(2S + 1)$.¹¹⁾ However, there are small parameters x and $1/(2S + 1)$. Because of the former, we consider only the terms with one fermion loop. To show that the latter exists, we write the Hamiltonian in the following way:

$$H = H_0 + H_1 + H_2 + H_3 + H_4 \quad (4)$$

where

$$H_0 = -\frac{t}{N(2S + 1)^2} \sum_{i,j,\mathbf{k},\sigma} e^{i\mathbf{k} \cdot \mathbf{R}_{ij}} [(S - \sigma \langle S_z \rangle)^2 c_{\mathbf{k},\sigma}^\dagger c_{\mathbf{k},\sigma} + (S_{iz} + S_{jz} - 2\langle S_z \rangle)(\langle S_z \rangle - \sigma S) n_{\mathbf{k},\sigma}], \quad (5)$$

$$H_1 = -\frac{t}{N(2S + 1)^2} \sum_{i,j,\mathbf{k},\mathbf{q},\sigma} e^{i\mathbf{k} \cdot \mathbf{R}_{ij} + i\mathbf{q} \cdot \mathbf{R}_i} (c_{\mathbf{k}-\mathbf{q},\sigma}^\dagger c_{\mathbf{k},\sigma} - n_{\mathbf{k},\sigma})(S_{iz} + S_{jz} - 2\langle S_z \rangle)(\langle S_z \rangle - \sigma S), \quad (6)$$

$$H_2 = -\frac{t}{N(2S + 1)^2} \sum_{i,j,\mathbf{k},\mathbf{q},\sigma} e^{i\mathbf{k} \cdot \mathbf{R}_{ij} + i\mathbf{q} \cdot \mathbf{R}_i} c_{\mathbf{k}-\mathbf{q},\sigma}^\dagger c_{\mathbf{k},\sigma} (S_{iz} - \langle S_z \rangle)(S_{jz} - \langle S_z \rangle), \quad (7)$$

$$H_3 = -\frac{t}{N(2S + 1)^2} \sum_{i,j,\mathbf{k},\mathbf{q},\sigma} e^{i\mathbf{k} \cdot \mathbf{R}_{ij} + i\mathbf{q} \cdot \mathbf{R}_i} (c_{\mathbf{k}-\mathbf{q},\sigma}^\dagger c_{\mathbf{k},\sigma} S_{i-} S_{j+} + c_{\mathbf{k}-\mathbf{q},\sigma}^\dagger c_{\mathbf{k},\sigma} S_{i+} S_{j-}) \quad (8)$$

and

$$H_4 = -\frac{t}{N(2S + 1)^2} \sum_{i,j,\mathbf{k},\mathbf{q}} e^{i\mathbf{k} \cdot \mathbf{R}_{ij} + i\mathbf{q} \cdot \mathbf{R}_i} [c_{\mathbf{k}-\mathbf{q},\uparrow}^\dagger c_{\mathbf{k},\downarrow} (S_{iz} S_{i-} - S_{i-} S_{jz} - SS_{i-} - SS_{j-}) + c_{\mathbf{k}-\mathbf{q},\downarrow}^\dagger c_{\mathbf{k},\uparrow} (-SS_{i+} - SS_{j+} + S_{i+} S_{jz} - S_{iz} S_{j+})], \quad (9)$$

where $\langle X \rangle$ is the thermal average of X in the ensemble of H_0 , $n_{\mathbf{k}\sigma} = \langle c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} \rangle$ and N is the number of lattice sites. H_0 is the mean-field part of eq. (1). H_1 and H_2 represents the contribution of the quantum fluctuations of S_z . H_3 has the flipping of the local spins and H_4 is the hole spin-flip terms.

Since H_0 contains the most important contribution, we elaborated it in more details. First of all, the spin-up holes and spin-down holes form two bands. Assuming $\langle S_z \rangle > 0$, the spin-down holes are the majority carriers. They have a smaller effective mass (proportional to $(S + \langle S_z \rangle)^{-2}$) and lower energy band bottom. The spin-up holes are immobile, have zero energy at $T = 0$, and $\langle S_z \rangle = S$. But they form a narrow band at low temperature. The bottom of the band is high compared to that of spin-down holes. Hence there are fewer of them. However, as temperature rises, $\langle S_z \rangle$ becomes smaller and two bands move closer to each other. The local spins have discrete energy levels whose spacings are related to the hole energy and abundance. This is our basic picture.

The energy of an hole with spin polarization σ is

$$\varepsilon_{\mathbf{k},\sigma} = -(S - \sigma \langle S_z \rangle)^2 \varepsilon_{\mathbf{k}}. \quad (10)$$

where

$$\varepsilon_{\mathbf{k}} = \frac{t}{(2S + 1)^2} \sum_{n,n.} e^{i\mathbf{k} \cdot \mathbf{R}}. \quad (11)$$

As usual, $\sum_{n,n.}$ sums over nearest neighbors and $\sigma = \pm 1$. If the local spins have $\langle S_z \rangle > 0$, then the spin-down holes have lower energy. H_0 can be solved to give the zeroth order spin wave propagator. This will be given in next section.

It seems that H_{1-4} are of the order S^0 . Actually, their higher order terms are not. For example, the thermal average of the terms first order in H_1 vanish. The second order terms are proportional to

$$\frac{t^2}{(2S + 1)^4} (\langle S_z^2 \rangle - \langle S_z \rangle^2) (S_z - \sigma S)^2.$$

It vanishes in the low temperature limit. At finite temperature, the criterion of our $1/(2S + 1)$ expansion is

$$\langle S_z^2 \rangle - \langle S_z \rangle^2 \leq S.$$

Using the Weiss' mean field theory with spin level spacing ω_0 , this is valid for $k_B T \leq S\omega_0$. Thus, for H_1 , we can have a $1/(2S + 1)$ expansion except for the region near T_C .

Similar argument can be applied to higher order terms and terms with H_2 . H_3 and H_4 are the spin-flip terms. Any physical quantity involving spin-flipping must contain the factor $\langle S_+ S_- \rangle$. It will be shown in next section that $\langle S_+ S_- \rangle$ is proportional to S_z . Thus, the terms second order in H_3 or H_4 are of the order $1/(2S+1)$. Therefore, H_1 , H_2 , H_3 and H_4 can be treated perturbatively. Due to the complexity of the Hamiltonian, we consider only the $O(1/(2S+1))$ terms.

§3. Spin Wave Propagator

We give some details of how to calculate the spin wave propagator in this section. The spin wave in the DE systems had been studied by Furukawa.²³⁾ The reason why we embark on this subject is that we are interested in the finite temperature behavior. Readers will also notice that our approaches are different. In order to investigate the behavior of the spin waves, we calculate the spin wave propagator. Since we are interested in the finite temperature behavior of spin waves, those simplifications such as Holstein-Primakoff transformation will not be applied. Instead, we perform calculation with the expansion of $1/(2S+1)$. The leading term in H is H_0 . It is solved *exactly*. As a result, the zeroth order spin propagator $-\langle \hat{T} S_+(\tau) S_-(\tau') \rangle$ where $\hat{T}$ is the time-ordered operator, can be evaluated.

$$\begin{aligned} D_0(iq_n) &= -\frac{1}{2} \int_{-\beta}^{\beta} d\tau \langle \hat{T} S_+(\tau) S_-(\tau') \rangle e^{iq_n(\tau-\tau')} \\ &= \frac{2\langle S_z \rangle}{iq_n - \omega_0} \end{aligned} \quad (12)$$

where

$$\omega_0 = \frac{2}{N} \sum_{\mathbf{k}, \sigma} f_{\mathbf{k}\sigma} (\langle S_z \rangle - \sigma S) \varepsilon_{\mathbf{k}}, \quad (13)$$

comes from the commutator H_0 and S_+ :

$$[H_0, S_+] = -\omega_0 S_+. \quad (14)$$

and $f_{\mathbf{k}\sigma}$ is the Fermi-Dirac distribution function of holes with spin polarization σ . The derivation of eq. (12) was done by Giovannini *et al.*²⁴⁾ We briefly outline its process. In view of eq. (14), we found

$$S_{\pm}(\tau) = e^{\mp \tau \omega_0} S_{\pm}, \quad (15)$$

and

$$\begin{aligned} D_0(iq_n) &= -\sum_{l,m} \frac{e^{-\beta E_l}}{2(iq_n - \omega_0)} [(e^{-\beta \omega_0} - 1) |(S_-)_{ml}|^2 \\ &\quad + (1 - e^{\beta \omega_0}) |(S_+)_{ml}|^2]. \end{aligned} \quad (16)$$

Since $E_m = E_l + \omega_0$, we reached eq. (12).

In the calculation of higher order terms, we confronted the complexity that the Wick's theorem is not applicable to spin operators.²⁴⁾ As a result, there will be other propagators such as $\langle \hat{T} S_{i+}(\tau) S_{j-}(\tau') S_{mz}(\tau_1) \rangle$, $\langle \hat{T} S_{i+}(\tau) S_{j-}(\tau') S_{mz}(\tau_1) S_{lz}(\tau_2) \rangle$ and $\langle \hat{T} S_{i+}(\tau) S_{j-}(\tau') S_{m+}(\tau_1) S_{l-}(\tau_2) \rangle$. (The time argument of S_z is not irrelevant even though $[H_0, S_z] = 0$. It helps determine the order of spin operators.) They can also be calculated *exactly* in the ensemble of H_0 . The way to calculate these

propagators is make contractions so that the final form contains only S_z .²⁴⁾ If this goal is reached we can evaluate the thermal average because S_z and H_0 are commute. The procedure of computing $\langle \hat{T} S_+(\tau) S_-(\tau') S_z(\tau_1) \rangle$ will be given as an example. Others will follow accordingly. We begin with the following identity:

$$\begin{aligned} &\langle \hat{T} S_{i+}(\tau) S_{j-}(\tau') S_{mz}(\tau_1) \rangle \\ &= \delta_{ij} [(1 - \delta_{im}) \langle \hat{T} S_+(\tau) S_-(\tau') \rangle \langle S_z \rangle \\ &\quad + \delta_{im} \langle \hat{T} S_+(\tau) S_-(\tau') S_z(\tau_1) \rangle]. \end{aligned} \quad (17)$$

The meaning of eq. (17) is that if S_{mz} operates on the same site as $S_+ S_-$, then these three operators cannot be decoupled. Consider $\theta(\tau - \tau') \theta(\tau' - \tau_1) \langle S_+(\tau) S_-(\tau') S_z \rangle$ (the order of spin operators is fixed). It belongs to the second term of eq. (17). One moves S_+ in a cyclic fashion²⁵⁾ and creates various commutators. Once it was moved back to its original position, an additional factor of $\exp(-\beta \omega_0)$ was created from its commutator with $\exp(-\beta H_0)$ from the ensemble average. Thus, we found (using eq. (15))

$$\begin{aligned} &\theta(\tau - \tau') \theta(\tau' - \tau_1) \langle S_+(\tau) S_-(\tau') S_z \rangle \\ &= \theta(\tau - \tau') \theta(\tau' - \tau_1) e^{\omega_0(\tau' - \tau)} \\ &\quad \times \left[\frac{2\langle S_z^2 \rangle}{1 - e^{-\beta \omega_0}} - \frac{2e^{-\beta \omega_0} \langle S_z \rangle}{(1 - e^{-\beta \omega_0})^2} \right] \end{aligned} \quad (18)$$

This way, we found that

$$\begin{aligned} &\langle \hat{T} S_+(\tau) S_-(\tau') S_z(\tau_1) \rangle \\ &= -\mathcal{D}(\tau - \tau') \langle S_z^2 \rangle - 2\langle S_z \rangle e^{\omega_0(\tau' - \tau)} [n_B(n_B + 1) \\ &\quad - n_B \theta(\tau - \tau_1) \theta(\tau_1 - \tau') \\ &\quad + (n_B + 1) \theta(\tau' - \tau_1) \theta(\tau_1 - \tau)] \end{aligned} \quad (19)$$

where

$$\begin{aligned} &\mathcal{D}(\tau_1 - \tau_2) \\ &= -2[n_B \theta(\tau_2 - \tau_1) + (n_B + 1) \theta(\tau_1 - \tau_2)] e^{\omega_0(\tau_2 - \tau_1)} \end{aligned} \quad (20)$$

and

$$n_B = \frac{1}{e^{\beta \omega_0} - 1}. \quad (21)$$

We can calculate $\langle \hat{T} S_+(\tau) S_-(\tau') S_z(\tau_1) S_z(\tau_2) \rangle$ and $\langle \hat{T} S_+(\tau) S_-(\tau') S_+(\tau_1) S_-(\tau_2) \rangle$ the similar way. The results are given in Appendix A.

In general, the form of the spin wave propagator is

$$D(iq_n, \mathbf{q}) = \frac{2\langle S_z \rangle}{iq_n - \omega_0 - \Pi(iq_n, \mathbf{q})}. \quad (22)$$

where $\Pi(iq_n, \mathbf{q})$ is the self-energy. In a perturbative series, we might get the form

$$\begin{aligned} &D(iq_n, \mathbf{q}) \\ &\simeq \frac{2\langle S_z \rangle}{iq_n - \omega_0} - \frac{2\langle S_z \rangle Z'(iq_n)}{iq_n - \omega_0} + \frac{2\langle S_z \rangle A(iq_n, \mathbf{q})}{(iq_n - \omega_0)^2} + \dots \end{aligned}$$

This implies a spin wave propagator of the form

$$D(iq_n, \mathbf{q}) \simeq \frac{2\langle S_z \rangle [1 - Z'(iq_n)]}{iq_n - \omega_0 - A(iq_n, \mathbf{q})}. \quad (23)$$

$1 - Z'(iq_n)$ is the weight of the coherent part of the spin wave and $A(i\omega_0, \mathbf{q})$ gives the spin wave disper-

sion relation and dissipation. We shall calculate $A(iq_n, \mathbf{q})$ and $Z'(iq_n)$ and retain only the terms of the order of $1/(2S + 1)$.

In general, the full spin propagator is given by

$$D(iq_n, \mathbf{q}) = \frac{-1}{2N} \int_{-\beta}^{\beta} d(\tau - \tau') \sum_{i,j} e^{iq_n(\tau - \tau') + i\mathbf{q} \cdot \mathbf{R}_{ij}} \times \langle \hat{T} \exp\{-\int_0^{\beta} d\tau_1 [H_1(\tau_1) + H_2(\tau_1) + H_3(\tau_1) + H_4(\tau_1)]\} S_{i+}(\tau) S_{j-}(\tau') \rangle. \quad (24)$$

Pertubatively, we have

$$\begin{aligned} & \langle \hat{T} \exp\{-\int_0^{\beta} d\tau_1 [H_1(\tau_1) + H_2(\tau_1) + H_3(\tau_1) + H_4(\tau_1)]\} S_{i+}(\tau) S_{j-}(\tau') \rangle \\ & \simeq \langle \hat{T} S_{i+}(\tau) S_{j-}(\tau') \rangle - \langle \hat{T} S_{i+}(\tau) S_{j-}(\tau') \int_0^{\beta} d\tau_1 [H_1(\tau_1) + H_2(\tau_1) + H_3(\tau_1) + H_4(\tau_1)] \rangle \\ & + \frac{1}{2} \langle \hat{T} S_{i+}(\tau) S_{j-}(\tau') \int_0^{\beta} d\tau_1 [H_1(\tau_1) + H_2(\tau_1) + H_3(\tau_1) + H_4(\tau_1)] \cdot \int_0^{\beta} d\tau_2 [H_1(\tau_2) + H_2(\tau_2) + H_3(\tau_2) + H_4(\tau_2)] \rangle. \end{aligned} \quad (25)$$

Only H_3 contributes to the first order term:

$$\sum_{i,j} e^{i\mathbf{q} \cdot \mathbf{R}_{ij}} \langle \hat{T} S_{i+}(\tau) S_{j-}(\tau') H_3(\tau_1) \rangle = -\langle \hat{T} S_+(\tau) S_-(\tau_1) \rangle \langle \hat{T} S_+(\tau_1) S_-(\tau') \rangle \frac{1}{N} \sum_{\mathbf{k}, \sigma} f_{\mathbf{k}, \sigma} \varepsilon_{\mathbf{k} - \mathbf{q}}. \quad (26)$$

The expressions of the second-order terms are quite lengthy. We have

$$\begin{aligned} & \sum_{i,j} e^{i\mathbf{q} \cdot \mathbf{R}_{ij}} \langle \hat{T} S_{i+}(\tau) S_{j-}(\tau') H_1(\tau_1) H_1(\tau_2) \rangle \\ & = \frac{1}{N} \sum_{\mathbf{p}, \sigma, l} e^{i\mathbf{p}l(\tau_1 - \tau_2)} \chi_z(i\mathbf{p}l, \mathbf{p}, \sigma) (\langle S_z \rangle - \sigma S)^2 [\langle \hat{T} S_+(\tau) S_-(\tau') S_z(\tau_1) S_z(\tau_2) \rangle - 2\langle S_z \rangle \langle \hat{T} S_+(\tau) S_-(\tau') S_z(\tau_1) \rangle \\ & + \langle S_z \rangle^2 \langle \hat{T} S_+(\tau) S_-(\tau') \rangle + (\langle S_z^2 \rangle - \langle S_z \rangle^2) \langle \hat{T} S_+(\tau) S_-(\tau') \rangle], \end{aligned} \quad (27)$$

and

$$\begin{aligned} & \sum_{i,j} e^{i\mathbf{q} \cdot \mathbf{R}_{ij}} \langle \hat{T} S_{i+}(\tau) S_{j-}(\tau') H_4(\tau_1) H_4(\tau_2) \rangle \\ & = \sum_{\sigma, l} e^{i\mathbf{p}l(\tau_1 - \tau_2)} \chi_t(i\mathbf{p}l, \mathbf{q}, \sigma) \{ \langle \hat{T} S_+(\tau) S_-(\tau_1) \rangle \langle \hat{T} S_+(\tau_2) S_-(\tau') \rangle + [\langle \hat{T} S_+(\tau) S_-(\tau') S_+(\tau_2) S_-(\tau) \rangle \\ & - \langle \hat{T} S_+(\tau) S_-(\tau_1) \rangle \langle \hat{T} S_+(\tau_2) S_-(\tau') \rangle + \langle \hat{T} S_+(\tau) S_-(\tau') \rangle \langle \hat{T} S_+(\tau_2) S_-(\tau_1) \rangle] \}, \end{aligned} \quad (28)$$

where

$$\chi_z(i\mathbf{p}l, \mathbf{p}, \sigma) = \frac{1}{N} \sum_{\mathbf{k}} (\varepsilon_{\mathbf{k}} + \varepsilon_{\mathbf{k} - \mathbf{p}})^2 \frac{f_{\mathbf{k}, \sigma} - f_{\mathbf{k} - \mathbf{p}, \sigma}}{i\mathbf{p}l - \varepsilon_{\mathbf{k}, \sigma} + \varepsilon_{\mathbf{k} - \mathbf{p}, \sigma}}, \quad (29)$$

and

$$\begin{aligned} \chi_t(i\mathbf{p}l, \mathbf{p}, \sigma) & = \frac{1}{N} \sum_{\mathbf{k}} [(S - \sigma \langle S_z \rangle) \varepsilon_{\mathbf{k}} + (S + \sigma \langle S_z \rangle) \varepsilon_{\mathbf{k} - \mathbf{p}}]^2 \\ & \times \frac{f_{\mathbf{k}, \sigma} - f_{\mathbf{k} - \mathbf{p}, -\sigma}}{i\mathbf{p}l - \varepsilon_{\mathbf{k}, \sigma} + \varepsilon_{\mathbf{k} - \mathbf{p}, -\sigma}}, \end{aligned} \quad (30)$$

come from the hole polarization. The last terms of eqs. (27) and (28) give disconnected diagrams. They will be canceled by the corresponding diagrams in the thermodynamic potential. The term second order in H_2 is higher order in $1/(2S + 1)$ except for

the region near T_C , and can be neglected. The term $\langle \hat{T} S_{i+}(\tau) S_{j-}(\tau') H_1(\tau_1) H_2(\tau_2) \rangle$ are also negligible for the similar reason. Hence, there are only three terms, namely, those in eqs. (26), (27) and (28) need to be considered. Among them, the spin-flip processes in eqs. (26) and (28) give $A(iq_n, \mathbf{q})$ and the S_z related eq. (27) gives $Z'(iq_n)$. The full expressions are in Appendix A. Here, we retaining only the leading terms in eqs. (A.2) and (A.3),

$$\begin{aligned} Z'(iq_n) & = \frac{\beta}{2N \langle S_z \rangle} [\langle S_z^3 \rangle - 2\langle S_z^2 \rangle \langle S_z \rangle + \langle S_z \rangle^3] \\ & \times \sum_{\mathbf{p}, \sigma} (S - \sigma S_z)^2 \chi_z(0, \mathbf{p}, \sigma), \end{aligned} \quad (31)$$

and

$$A(iq_n, \mathbf{q}) = -\frac{2\langle S_z \rangle}{N} \sum_{\mathbf{k}, \sigma} f_{\mathbf{k}, \sigma} \varepsilon_{\mathbf{k}-\mathbf{q}} - \langle S_z \rangle \sum_{\sigma} \chi_t(iq_n, \mathbf{q}, \sigma). \quad (32)$$

Thus, we completed the calculation of the spin propagator. Using the analytical continuation to the real frequency we found the spin wave energy

$$\omega(\mathbf{q}) = \omega_0 - \frac{2\langle S_z \rangle}{N} \sum_{\mathbf{k}, \sigma} f_{\mathbf{k}, \sigma} \varepsilon_{\mathbf{k}-\mathbf{q}} - \langle S_z \rangle \sum_{\sigma} \chi_t(\omega, \mathbf{q}, \sigma). \quad (33)$$

$\omega(\mathbf{q})$ is gapless. At $\mathbf{q} = 0$,

$$\chi_t(\omega, 0, \sigma) = \frac{1}{N} \sum_{\mathbf{k}} (2S\varepsilon_{\mathbf{k}})^2 \frac{f_{\mathbf{k}, \sigma} - f_{\mathbf{k}, -\sigma}}{\omega - \varepsilon_{\mathbf{k}, \sigma} + \varepsilon_{\mathbf{k}, -\sigma}}. \quad (34)$$

In view of eqs. (10) and (11), we found a solution $\omega = 0$ from eq. (33) by substituting eq. (34) into eq. (33). We evaluate the spin stiffness ($\omega = D_S \mathbf{q}^2$) assuming cubic symmetry.

$$D_S = - \sum_{\mathbf{k}, \sigma} \left(\frac{\langle S_z \rangle}{3} + \frac{S^2 + \langle S_z \rangle^2}{12\langle S_z \rangle} - \frac{\sigma S}{2} \right) f_{\mathbf{k}, \sigma} \nabla^2 \varepsilon_{\mathbf{k}} + \sum_{\mathbf{k}, \sigma} \frac{(S^2 - \langle S_z \rangle^2)^2}{24S\langle S_z \rangle^2} \sigma f_{\mathbf{k}, \sigma} \frac{(\nabla \varepsilon_{\mathbf{k}})^2}{\varepsilon_{\mathbf{k}}} \quad (35)$$

It can easily be derived that as $T \rightarrow 0$, $\langle S_z \rangle \rightarrow S$, D_S is reduced to the form given by Kubo and Ohata, which was obtained with Holstein-Primakoff transformation. On the other hand, $D_S \rightarrow O(\langle S_z \rangle)$ as $\langle S_z \rangle \rightarrow 0$ and $T \rightarrow T_C$ (see Appendix B).

§4. Results and Discussion

The goal of this work is computing the spin stiffness. A by-product is that we calculated approximately the Curie temperature. Our calculation is self-consistent. For a given $\langle S_z \rangle$, we can adjust the chemical potential and the hole density so as to get the desired doping concentration. Now $\langle S_z \rangle$ can, in turn, be calculated from the ensemble of H_0 . We can then adjust $\langle S_z \rangle$ to reach self-consistency. At this stage, only H_0 is considered. We can do the computation in the whole temperature range. For a given doping concentration, we found that above certain temperature there is no solution for $\langle S_z \rangle \neq 0$. We deemed this temperature T_C .

The T_C from our calculation on double exchange (DE) alone is at about 800°K for $x = 0.3$. It is three times of the experimental T_C . However, there is superexchange interaction in manganites. To make our result conformable to realistic systems, we added a superexchange term (SE) to our Hamiltonian

$$\frac{J}{2} \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j. \quad (36)$$

A reasonable value for J is $t/35$ where $t = 0.2$ eV since the on-site Coulomb repulsion is around $U \approx 7$ eV and $J \approx t^2/U$.²⁶⁾ Since SE is quite small we can treat it with a mean-field approximation. It modifies ω_0 . Its contribution to D_S was considered to the first order. The standard textbook approach gives the following modification:

cation:

$$\begin{aligned} \omega_0 &\rightarrow \omega_0 - zJ\langle S_z \rangle \\ D_S &\rightarrow D_S - Ja^2\langle S_z \rangle/2 \end{aligned}$$

where z is the coordination number and a is the lattice constant.

The results are shown in Figs. 1–3 in which crosses, the dashed lines and the solid lines represent those at doping $x = 0.4, 0.3$ and 0.2 respectively. In Fig. 1 we showed the averaged local spin. The upper (lower) set were calculated without (with) SE. Though it is not the magnetization, the curves showed approximately where T_C 's are. We have to be cautious because the results do not account for the fluctuations. They are self-consistent in the mean-field level. However, they give an approximate range of T_C . It is lower than those given by eq. (4.12) of Kubo and Ohata¹¹⁾ but are twice of the results of Furukawa.²³⁾ The fact that our result is not too far away from those of ref. 11 is expected though they used a different method to calculate T_C and their eq. (4.12) is an approximation. Furukawa used a infinite-dimension system and therefore, making comparison is difficult. We also noted that SE suppressed magnetization so that it was not saturated as $T \rightarrow 0$. This seems agree with experimental findings. Uhlenbruck *et al.*²⁷⁾ had shown that the magnetic moments of manganese ions increase with applied magnetic field even at very low temperature.

SE reduced the T_C of DE by more than half even though its magnitude is rather small. The reason is that SE reduces $\langle S_z \rangle$. The change in $\langle S_z \rangle$ affects the shape of hole bands and change the numbers of spin-up and spin-down holes. At finite temperature, it does not cost much energy because the holes near Fermi level can have their spins flipped. For the same reason, applying external magnetic field affects T_C , moves it to the other direction. Hence, a little SE can have a large effect on the numbers of spin-up and spin-down holes, which in turn, are the deciding factors of the coupling between local spins. For the same reason large-doping materials are more susceptible to SE. The spins of holes are easier to flip if the chemical potential is higher. In Fig. 2 we show the influence of the superexchange interaction. T_C decreases as J increases. For realistic ratio of $J/t \simeq 0.03$, T_C drops to 350°K. Incorporating fluctuations will further suppress T_C and make it compatible with experimental results. We note here that we did not try to fit experimental T_C . It is meaningless at this stage. Other effects such as the shapes of the bands, Jahn-Teller distortion and orbital degeneracies are not included in our calculation. Different ions, such as La^{+3} and Ca^{+2} , giving different potentials will impede the motion of holes and diminish the coupling. If considered, T_C will be lower. Nevertheless, our T_C is quite close to experimental values. It would not have been the case if we, in view of eq. (1), had inferred that T_C is of the order $xzt/(2S+1)$ due to the strength of spin waves. The spin stiffness softens as temperature rises. Thus, T_C is much lower than above prediction. One may understand it by the following argument: as temperature rises, $\langle S_z \rangle$ becomes smaller. A smaller $\langle S_z \rangle$ means smaller energy difference between

Fig. 1. Averaged local spin versus temperature. The results at doping $x = 0.2, 0.3$ and 0.4 are represented by the solid line, the dashed line and crosses. The lower set were computed with SE included.

Fig. 2. T_C versus J/t . The results at doping $x = 0.2, 0.3$ and 0.4 are represented by the solid line, the dashed line and crosses.

spin-up and spin-down holes and smaller difference in abundances of spin-up and spin-down holes, which in turn, give even smaller difference in the energies of up- and down- local spins and even smaller $\langle S_z \rangle$.

Recent numerical computations²⁸⁻³⁰⁾ gave much lower T_C , typically around $0.1t$. This value has the same order of magnitude as experimental results. Our T_C is too high because it was calculated by mean-field approximation of DE. However, the fluctuation effect alone is unlikely to reduce T_C to such a low value. SE must play some role here. In ref. 30, Yi *et al.* showed that T_C was reduced by half if SE was introduced at the strength $J \simeq 0.3t$. This is compatible with our calculation. This creates another

Fig. 3. D_S versus temperature. The results at doping $x = 0.2, 0.3$ and 0.4 are represented by the dashed line, the solid line and crosses.

problem. if T_C given by refs. 28–30 is correct, adding SE which does exist in manganites, will make it too low. Apparently, more study is in order.

The experimental data on the spin waves are quite consistent.^{20,21)} The manganites have isotropic spin waves. There is no noticeable energy gap. The spin wave stiffness D_S is expected to increase with x . At $x = 1/3$, $D_S \simeq 170 \text{ meV Å}^2$ and decreases with increasing temperature. We show in Fig. 3 D_S versus T . From our calculation, $D_S \simeq 160 \text{ meV Å}^2$ at $x = 0.3$. Also, at $T/T_C = 3/5$, $D_S(T)/D_S(0) \simeq 4/5$ from experiments, similar in Fig. 3. We also noticed that there is significant fluctuation of yet unknown origin near T_C . We did not attempt to make comparison since our calculation is not accurate in this region. By analyzing the DE Hamiltonian, it can be attributed to the fact S_z is not a good quantum number. The quantum fluctuation becomes significant at $T \rightarrow T_C$. It is possible that this is the cause for the deviation between our result and experimental data in the temperature dependence of D_S .

In conclusion, we have studied the DE Hamiltonian in details and found that we can perform the $1/(2S + 1)$ expansion at $T < T_C$. We then calculated the spin wave propagator at finite temperature. The resulting spin wave stiffness was in reasonably good agreement with experimental data. Adding superexchange interaction can reduce the Curie temperature to the range of real systems. We concluded that DE is the fundamental mechanism in perovskite manganites.

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Appendix A

In this appendix, we give expressions of two propagators.

$$\begin{aligned}
& \langle \hat{T} S_+(\tau) S_-(\tau') S_z(\tau_1) S_z(\tau_2) \rangle \\
&= -\langle S_z^3 \rangle \mathcal{D}(\tau - \tau') + 2\langle S_z^2 \rangle e^{\omega_0(\tau_2 - \tau_1)} \{ 2n_B [\xi(\tau', \tau_1, \tau_2, \tau) + \xi(\tau', \tau_2, \tau_1, \tau)] \\
&\quad - 2(n_B + 1) [\xi(\tau, \tau_1, \tau_2, \tau') + \xi(\tau, \tau_2, \tau_1, \tau')] + n_B [\xi(\tau', \tau_1, \tau, \tau_2) + \xi(\tau', \tau_2, \tau, \tau_1) + \xi(\tau_1, \tau', \tau_2, \tau) + \xi(\tau_2, \tau', \tau_1, \tau)] \\
&\quad - (n_B + 1) [\xi(\tau, \tau_1, \tau', \tau_2) + \xi(\tau, \tau_2, \tau', \tau_1) + \xi(\tau_1, \tau, \tau_2, \tau') + \xi(\tau_2, \tau, \tau_1, \tau')] \} \\
&\quad + 2\langle S_z \rangle \{ 2n_B(n_B + 1) [(n_B + 1)\theta(\tau - \tau') + n_B\theta(\tau' - \tau)] + (n_B + 1)^2\theta(\tau - \tau') \\
&\quad - n_B^2\theta(\tau' - \tau) + n_B(n_B + 1) [\xi(\tau', \tau, \tau_1, \tau_2) + \xi(\tau', \tau, \tau_2, \tau_1) + \xi(\tau_1, \tau', \tau, \tau_2) \\
&\quad + \xi(\tau_2, \tau', \tau, \tau_1) + \xi(\tau_1, \tau_2, \tau', \tau) + \xi(\tau_2, \tau_1, \tau', \tau) - \xi(\tau, \tau', \tau_1, \tau_2) \\
&\quad - \xi(\tau, \tau', \tau_2, \tau_1) - \xi(\tau_1, \tau, \tau', \tau_2) - \xi(\tau_2, \tau, \tau', \tau_1) - \xi(\tau_1, \tau_2, \tau, \tau') - \xi(\tau_2, \tau_1, \tau, \tau')] \}. \tag{A.1}
\end{aligned}$$

Its form in frequency representation is less lengthy. However, we will show the Fourier transformed expression after it was multiplied by a term $(1/\beta) \sum_l F(ip_l) e^{ip_l(\tau_1 - \tau_2)}$. It will be useful in evaluating eq. (27).

$$\begin{aligned}
& -\frac{1}{2\beta} \sum_l \int_{-\beta}^{\beta} d(\tau - \tau') e^{ip_l(\tau - \tau')} \int_0^{\beta} d\tau_1 \int_0^{\beta} d\tau_2 e^{ip_l(\tau_1 - \tau_2)} F(ip_l) \langle \hat{T} S_+(\tau) S_-(\tau') S_z(\tau_1) S_z(\tau_2) \rangle \\
&= \sum_l F(ip_l) \{ \delta_{0p_l} [\frac{2\beta\langle S_z^3 \rangle}{iq_n - \omega_0} + \frac{4\langle S_z^2 \rangle}{(iq_n - \omega_0)^2} + n_B(n_B + 1) \frac{4\beta\langle S_z \rangle}{(iq_n - \omega_0)^2}] \\
&\quad + \frac{2\beta\langle S_z \rangle}{p_l} [\frac{2}{iq_n - \omega_0} - \frac{1}{iq_n + ip_l - \omega_0} - \frac{1}{iq_n - ip_l - \omega_0}] \} \tag{A.2}
\end{aligned}$$

Last term gives a disconnected diagram, so it will be canceled. We also have

$$\begin{aligned}
& \langle \hat{T} S_+(\tau) S_-(\tau') S_+(\tau_1) S_-(\tau_2) \rangle \\
&= \langle S_z^2 \rangle [\mathcal{D}(\tau - \tau_2) \mathcal{D}(\tau_1 - \tau') + \mathcal{D}(\tau - \tau') \mathcal{D}(\tau_1 - \tau_2)] \\
&\quad - 2\langle S_z \rangle \{ (n_B + 1/2) [\mathcal{D}(\tau - \tau_2) \mathcal{D}(\tau_1 - \tau') + \mathcal{D}(\tau - \tau') \mathcal{D}(\tau_1 - \tau_2)] \\
&\quad + (n_B + 1) [\xi(\tau, \tau', \tau_1, \tau_2) + \xi(\tau, \tau_2, \tau_1, \tau') + \xi(\tau_1, \tau', \tau, \tau_2) + \xi(\tau_1, \tau_2, \tau, \tau')] \\
&\quad + n_B [\xi(\tau', \tau, \tau_2, \tau_1) + \xi(\tau', \tau_1, \tau_2, \tau) + \xi(\tau_2, \tau, \tau', \tau_1) + \xi(\tau_2, \tau_1, \tau', \tau)] \}. \tag{A.3}
\end{aligned}$$

The terms of the order $\langle S_z \rangle$ will be neglected. For the two terms of the order $\langle S_z \rangle^2$, the second one gives a disconnected diagram. Hence, taking the Fourier transform of the first term and the contribution from the holes, we obtain eq. (28).

Appendix B

To see the behavior of D_S as $\langle S_z \rangle \rightarrow 0$, we note first that

$$f_{\mathbf{k},-} - f_{\mathbf{k},+} = 4S\langle S_z \rangle \varepsilon_{\mathbf{k}} \frac{\partial f_{\mathbf{k},0}}{\partial \varepsilon_{\mathbf{k},0}} + O(S_z^3), \tag{B.1}$$

where

$$\varepsilon_{\mathbf{k},0} = \frac{S^2 \varepsilon_{\mathbf{k}}}{(2S + 1)^2} \tag{B.2}$$

and $f_{\mathbf{k},0}$ is the Fermi function with energy $\varepsilon_{\mathbf{k},0}$. Substituting eq. (B.1) into eq. (33) and integrating by parts, we found that $D_S = O(S_z)$.

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