

## Quantum form of the double-exchange interaction

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A quantum form of the Hamiltonian for a double-exchange (DE) system, based on the results of Anderson and Hasegawa's semiclassical treatment, is presented together with several predictions. The magnetization decreases mainly in  $3/2$  and  $5/2$  powers of temperature at low temperature but includes a  $T^3$  term. The Curie temperature, in a rescaled DE coupling energy  $(\sqrt{2}b/2S^2)J_d$  unit, is dependent on hole concentration  $x$  and has highest value that is about  $1/3$  that of ordinary Heisenberg magnet at  $x=0.5$ . The susceptibility at high temperatures lacks a  $1/T^2$  term and has a characteristic temperature in rescaled DE coupling units that is about  $3/4$  that of Heisenberg magnet in molecular-field theory.

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### I. INTRODUCTION

Colossal magnetoresistance (CMR) materials<sup>1</sup> attract considerable attentions due to their potential for applications and their rich physical phenomena for scientific interests. The exotic phenomena arise from the strong coupling among charge, spin, and lattice in which the double exchange (DE)<sup>2</sup> plays an important role. Anderson and Hasegawa<sup>3</sup> studied DE in great detail and showed that the transfer element is proportional to  $\cos(\theta/2)$ , where  $\theta$  is the angle between the neighboring ionic spins. This was confirmed by studies on layered manganites.<sup>4</sup> However, their semiclassical treatment (SCT) is based on the simplest case of only two magnetic ions with one electron traveling between them. In real materials there are a large number of ions and electrons. The hopping of electrons is unavoidably affected by other degrees of freedom, e.g., lattice effect as pointed out by Millis *et al.*<sup>5</sup> To study the interplay among spin, charge, and lattice degrees of freedom dynamically, it is insightful to treat the DE problem in a quantum-mechanical form.

However, there are two features of DE that make it quite different from the usual form of exchange interaction and these make its treatment very difficult. One is the half-angle dependence of the interaction in SCT and the other is the extra degree of freedom allowed by the electron's motion. Kubo and Ohata<sup>6</sup> used a  $s$ - $d$  model to describe the problem and a projector method was introduced to show the equivalence between DE and  $s$ - $d$  model. This projector method is also used by Shen and Wang<sup>7</sup> and Izyumov and Skryabin.<sup>8</sup> Recently, Green *et al.*<sup>9</sup> used a many-body coherent potential approximation to study this  $s$ - $d$  model and showed that there are unphysical behavior in magnetic susceptibility at half-filling and no magnetic transition for any finite spin  $S$ . These indicate that  $s$ - $d$  model is not completely equivalent to Anderson and Hasegawa's semiclassical DE model. As a matter of fact, it compares the SCT and the  $s$ - $d$  Hamiltonian, one may find that there are some hidden factors in the hopping amplitude of the  $s$ - $d$  model that are important to make the  $s$ - $d$  Hamiltonian to be completely equivalent to Anderson and Hasegawa's semiclassical model. For example, Muller-Hartmann and Dagotto<sup>10</sup> (MHD) pointed out that the hopping amplitude for nearest-neighbor sites depends on both the spin values at the two sites and their projections. How-

ever, MHD's treatment puts a strong restriction that magnetic ground state is completely determined by the atomic states of two nearest-neighbor magnetic ions and omits an intermediate case, namely, the motional electron is in neither of the two sites that leads to loss of the explicit motional character (Fermion operators) in their effective Hamiltonian. Therefore, in our opinion, the  $s$ - $d$  model needs further improvement to describe DE system. In this paper, we have extended Anderson and Hasegawa's SCT on DE to obtain a quantum-mechanical Hamiltonian for DE. A temperature-dependent double-time Green function<sup>11-13</sup> solution for the DE magnetic system is obtained. The results show that at low temperatures the magnetization of localized spin expands in the  $T^{3/2}$ ,  $T^{5/2}$ ,  $T^{7/2}$  series which is typical Heisenberg behavior<sup>14-16</sup> but an additional  $T^3$  term is predicted, characterizing the motional and hole-concentration-dependent nature of the DE. The Curie temperature in a rescaled DE coupling unit is hole-concentration dependent and about  $1/3$  that of Heisenberg magnet at  $x=0.5$ . The susceptibility at high temperatures lacks a  $1/T^2$  term and exhibits a ratio of characteristic temperature and rescaled DE coupling constant of about  $3/4$  that of Heisenberg magnet from molecular-field theory.

### II. MODEL

In CMR materials there are two valences of Mn ions that are responsible for their magnetism. One is  $\text{Mn}^{4+}$  that has three localized  $3d$  electrons and spin  $S$ . This localized spin is defined as the *core spin* for the convenience of our discussion. Another is  $\text{Mn}^{3+}$  that also has three localized  $3d$  electrons and an extra  $d$  electron that is motional. The motion of the extra electron between two Mn ion neighbors generates a double-exchange interaction.<sup>2</sup>  $\text{Mn}^{4+}$  is referred to as  $d$  hole with a concentration  $x$  in DE system. We start with the following Hamiltonian for DE system

$$H_{DE} = - \sum_{f,h} \frac{J_d(f,h)}{\sqrt{2}} \left( 1 + b \frac{\mathbf{S}_f \cdot \mathbf{S}_h}{S^2} \right) (c_f^+ c_h - c_f c_h^+) + \mu \sum_f c_f^+ c_f,$$

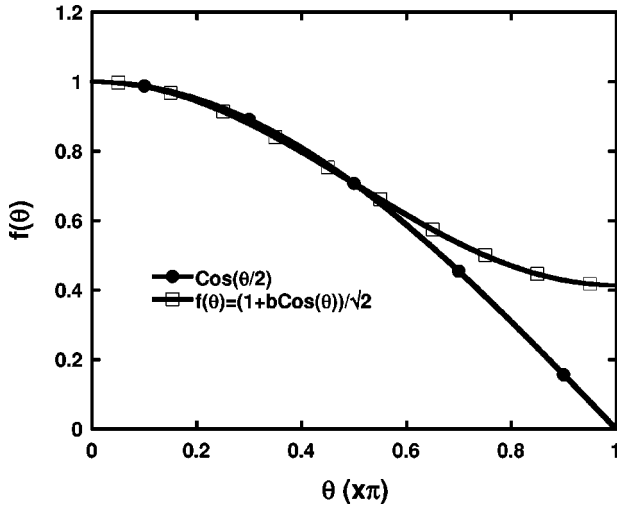


FIG. 1. Comparison between  $\cos \theta/2$  and  $f(\theta) = (1/\sqrt{2})(1 + b \cos \theta)$  with  $b = \sqrt{2} - 1$ .

where the parameter  $J_d(f, h)$  describes the usual hopping between Mn ions,  $b = \sqrt{2} - 1$ ,  $c_f^+$  ( $c_f$ ) is the operator which creates (destroys) a  $d$  electron with spin parallel to the localized core spin at the  $f$ th site, and  $\mu$  is the chemical potential. The addition of the chemical potential term allows the deviation of the occupied state of the motion electron from Anderson and Hasegawa's ideal two-body state<sup>3</sup> and deals with the fluctuation induced dynamical occupation number change of moving electron on a Mn ion.

First we show that this DE Hamiltonian is a good approximation for half-angle dependence in the SCL when the material is in the ferromagnetic (FM) state. In classical limit, the  $(1/\sqrt{2})[1 + b(\mathbf{S}_f \cdot \mathbf{S}_h/S^2)]$  becomes  $(1/\sqrt{2})(1 + b \cos \theta_{fh}) \equiv f(\theta_{fh})$ , while  $f(\theta_{fh})$  fits quite well to  $\cos(\theta_{fh}/2)$  for  $0 \leq \theta_{fh} \leq \pi/2$ , although a discrepancy appears for  $\pi/2 < \theta_{fh} \leq \pi$  as shown in Fig. 1. In SCL,  $\theta_{fh} = 0$  and  $\pi$  represent the material is in FM and antiferromagnetic (AF) states, respectively. Correspondingly, we expect the DE Hamiltonian is a good description for material in FM states. For AF states, the approximation may generate an inaccurate phase diagram border between FM and AF regions.

Next, we show the temperature-dependent Green function solution<sup>11-16</sup> for the DE Hamiltonian. To describe the magnetic properties of a DE system, we also add a Zeeman term in the Hamiltonian

$$H = - \sum_{f,h} \frac{J_d(f,h)}{\sqrt{2}} \left( 1 + b \frac{\mathbf{S}_f \cdot \mathbf{S}_h}{S^2} \right) (c_f^+ c_h - c_f c_h^+) + \mu \sum_f c_f^+ c_f - g \mu_B H \sum_f \left( S_{fz} + \frac{S_{fz}}{2S} c_f^+ c_f \right).$$

The temperature-dependent retard Green function  $\langle\langle A(t); B \rangle\rangle$  is defined<sup>13,14</sup> by

$$\langle\langle A(t); B \rangle\rangle \equiv -i \theta(t) \langle [A(t), B]_{\pm} \rangle,$$

where  $A$  and  $B$  are either Fermi or Bose operators and correspondingly the commutator

$$[A, B]_{\pm} = AB \pm BA; \theta(t) = \begin{cases} 1, & t > 0 \\ 0, & t < 0; \end{cases}$$

and single angular brackets denote an average with respect to the canonical density matrix of the system at temperature  $T$ . The equation of motion of  $\langle\langle A(t); B \rangle\rangle$  in Fourier form is

$$E \langle\langle A; B \rangle\rangle_E = \frac{1}{2\pi} [A, B]_{\pm} + \langle\langle [A(t), H]; B \rangle\rangle_E.$$

The correlation function  $\langle BA(t) \rangle$  can be extracted from the solution of above equation through the relation

$$\langle BA(t) \rangle = \lim_{\epsilon \rightarrow 0} \int_{-\infty}^{\infty} \frac{\langle\langle A; B \rangle\rangle_{\hbar\omega + i\epsilon} - \langle\langle A; B \rangle\rangle_{\hbar\omega + i\epsilon}}{\exp(\hbar\omega/k_B T) - 1} \times e^{-i\omega t} d\omega, \quad (1)$$

$$\langle AB(t) \rangle = \lim_{\epsilon \rightarrow 0} i \int_{-\infty}^{\infty} \frac{\langle\langle A; B \rangle\rangle_{\hbar\omega + i\epsilon} - \langle\langle A; B \rangle\rangle_{\hbar\omega + i\epsilon}}{\exp(\hbar\omega/k_B T) - 1} \times \exp(\hbar\omega/k_B T) e^{-i\omega t} d\omega. \quad (2)$$

In our calculation, we shall need four kinds of correlation functions  $\langle S_{fz}(t) S_{hz}(t) \rangle$ ,  $\langle S_f^+(t) S_h^-(t) \rangle$ ,  $\langle c_f^+(t) c_h(t) \rangle$ , and  $\langle c_f(t) c_h^+(t) \rangle$ , however,  $\langle S_{fz}(t) S_{hz}(t) \rangle$  is only used to decouple the entanglement between the core spin and motional electron. We shall therefore study two Green's functions  $\langle\langle S_f^+(t); e^{w S_{hz}} S_h^-(t') \rangle\rangle$  and  $\langle\langle c_f^+(t); c_h(t') \rangle\rangle$ , where  $w$  is a parameter used to derive magnetization. The equations of motion for these Green's functions are

$$E \langle\langle S_f^+; e^{w S_{hz}} S_h^- \rangle\rangle = \frac{1}{2\pi} [S_f^+, S_h^-] + \langle\langle [S_f^+(t), H]; e^{w S_{hz}} S_h^- \rangle\rangle,$$

$$E \langle\langle c_f^+; c_h \rangle\rangle = \frac{1}{2\pi} \{c_f^+, c_h\} + \langle\langle [c_f^+(t), H]; c_h \rangle\rangle.$$

To conveniently describe solutions of above equations, we define some intermediate parameters and their space-Fourier components, namely,

$$C(f, h) = \langle c_f^+ c_h \rangle - \langle c_f c_h^+ \rangle, G(f, h) = \langle \mathbf{S}_f \cdot \mathbf{S}_h \rangle,$$

$$C(\mathbf{k}) = \frac{1}{N} \sum_{f,h} e^{-\mathbf{k} \cdot (\mathbf{f}-\mathbf{h})} C(f, h),$$

$$G(\mathbf{k}) = \frac{1}{N} \sum_{f,h} e^{-\mathbf{k} \cdot (\mathbf{f}-\mathbf{h})} G(f, h),$$

where  $N$  is the number of magnetic atom in the material and  $\mathbf{f}$  and  $\mathbf{h}$  represent the atom positions  $\mathbf{R}_f$  and  $\mathbf{R}_h$ . We use Callen decoupling<sup>15</sup> to decouple the core spins and Tayablikaff decoupling<sup>12,13</sup> to do the disentanglement among core spin Fermion, and Fermion-Fermion. Some manipulations yields

$$x = \frac{1}{2} - \frac{1}{2N} \sum_{\mathbf{k}} \tanh \left[ \frac{\phi(\mathbf{k}) + E_0^I}{2k_B T} \right], \quad (3)$$

$$C(\mathbf{k}) = \tanh \left[ \frac{\phi(\mathbf{k}) + E_0^I}{2k_B T} \right], \quad (4)$$

$$G(\mathbf{k}) = \langle S_z \rangle^2 + \langle S_z \rangle \coth\left(\frac{E_{\mathbf{k}}^c}{2k_B T}\right), \quad (5)$$

$$\langle S_z \rangle = \frac{(S - \Phi)(1 + \Phi)^{2S+1} + (S + 1 + \Phi)\Phi^{2S+1}}{(1 + \Phi)^{2S+1} - \Phi^{2S+1}}, \quad (6)$$

where

$$E_0^I = -\mu + g\mu_B \mathbf{H} \frac{\langle S_z \rangle}{2S},$$

$$\phi(\mathbf{k}) = \sqrt{2} \left[ J_d(\mathbf{k}) + \frac{b}{S^2} \frac{1}{N} \sum_{\mathbf{k}'} J_d(\mathbf{k} - \mathbf{k}') G(\mathbf{k}') \right],$$

$E_0^I + \phi(\mathbf{k})$

is the energy spectrum of charge wave for motional electrons;

$$\begin{aligned} E_{\mathbf{k}}^c &= \frac{\sqrt{2}b\langle S_z \rangle}{S^2} \frac{1}{N} \sum_{\mathbf{k}'} [J_d(-\mathbf{k}') - J_d(\mathbf{k} - \mathbf{k}')] C(\mathbf{k}') \\ &+ \frac{\sqrt{2}b\langle S_z \rangle^2}{S^4} \frac{1}{N^2} \sum_{\mathbf{k}_1, \mathbf{k}_2} [J_d(-\mathbf{k}_1 + \mathbf{k}_2) - J_d(\mathbf{k} - \mathbf{k}_1 - \mathbf{k}_2)] \\ &\times \varphi(\mathbf{k}_2) C(\mathbf{k}_1) + g\mu_B \mathbf{H} \left(1 + \frac{\langle n \rangle}{2S}\right) \end{aligned}$$

is the spin-wave energy spectrum for core spins,  $\varphi(\mathbf{k}) = 1/\exp(E_{\mathbf{k}}^c/k_B T) - 1$ ,  $\langle n \rangle = \langle c^+ c \rangle = 1 - x$ ,  $\Phi = 1/N \sum_{\mathbf{k}} \phi(\mathbf{k})$ . In deriving Eq. (1), identities

$$x = 1 - \langle c^+ c \rangle = \frac{1}{N} \sum_{\mathbf{k}} \frac{1}{\exp\left[\frac{\phi(\mathbf{k}) + E_0^I}{k_B T}\right] + 1}$$

and

$$1 - 2x = \frac{1}{N} \sum_{\mathbf{k}} \tanh\left[\frac{\phi(\mathbf{k}) + E_0^I}{2k_B T}\right]$$

are used. Equations (3) and (4) come from the space-Fourier components of the correlation functions,  $\langle c_f^+ c_h \rangle$ ,  $\langle c_f c_h^+ \rangle$ , and  $\langle \mathbf{S}_f \cdot \mathbf{S}_h \rangle$  that are obtained through relations (1) and (2). Equation (6) can be obtained following Callen's method.<sup>15</sup> These are coupled equations on parameters  $C(\mathbf{k})$ ,  $G(\mathbf{k})$ , and  $\langle S_z \rangle$ . The  $C(\mathbf{k})$  and  $G(\mathbf{k})$  represent the dynamical character of motional electrons and core spins. The coupling of  $C(\mathbf{k})$  and  $G(\mathbf{k})$  with each other reflects the interplay between the charge wave and spin wave. Our derivation for the coupled equations is not restricted to special crystal structure and nearest neighbor (NN) for the exchange interaction.

### III. RESULTS AND DISCUSSION

To reveal some characteristics of DE systems we shall discuss some interesting asymptotical solution of DE equation. For the sake of simplicity, the material is assumed to be simple cubic and the DE to be with NN. In that case,

$J_d(\mathbf{k}) = \frac{1}{3} J_d(0) [\cos k_x a + \cos k_y a + \cos k_z a]$ ,  $J_d(0) = z J_d$ , where  $a$  is the NN distance and  $z$  is the coordination number. The quantities  $\phi(\mathbf{k})$  and  $E_{\mathbf{k}}^c$  in Eqs. (3)–(5) become

$$\phi(\mathbf{k}) \equiv \sqrt{2} J_d(\mathbf{k}) \left[ 1 + \frac{b}{S^2} \frac{1}{N J_d(0)} \sum_{\mathbf{k}'} J_d(-\mathbf{k}') G(\mathbf{k}') \right], \quad (7)$$

$$\begin{aligned} E_{\mathbf{k}}^c &= 2q \langle S_z \rangle [J_d(0) - J_d(\mathbf{k})] \left[ 1 + \frac{\langle S_z \rangle}{S^2} \frac{1}{N J_d(0)} \right. \\ &\times \left. \sum_{\mathbf{k}'} J_d(\mathbf{k}') \varphi(\mathbf{k}') \right] + g\mu_B \mathbf{H} \left( 1 + \frac{\langle n \rangle}{2S} \right), \quad (8) \end{aligned}$$

where the new quantity  $q$  is

$$q \equiv \frac{\sqrt{2}}{2} \frac{b}{S^2} \frac{1}{N J_d(0)} \sum_{\mathbf{k}} J_d(-\mathbf{k}) C(\mathbf{k}). \quad (9)$$

We define  $\langle S_{tz} \rangle = \langle S_z \rangle [1 + (\langle n \rangle / 2S)]$  which includes the contribution from motional electron. In our calculation  $\langle n \rangle$  has been set to be a constant  $1 - x$ , thus the temperature dependence is mainly from  $\langle S_z \rangle$ . Following the techniques used by Dyson,<sup>14</sup> Callen<sup>15</sup> and Thahir-Khelli and ter Haar,<sup>16</sup> we obtain series solutions for the following three interesting temperature ranges. At low temperatures,

$$\begin{aligned} \langle S_z \rangle &= S - Li_{3/2}(1) \tau^{3/2} - \frac{3}{4} \pi Li_{5/2}(1) \tau^{5/2} - \frac{33}{32} \pi^2 Li_{7/2}(1) \tau^{7/2} \\ &\dots - 3 \left( \frac{b}{bS + b + S} \right)^{3/2} Li_{3/2}(1) \left| Li_{3/2} \left( \frac{x}{x-1} \right) \right| \\ &- O(\tau^4) - \dots, \quad (10) \end{aligned}$$

where  $\tau = 3k_B T / 4\pi z \bar{J}_d S$  following Dyson's notation<sup>14</sup> for simple cubic,  $\bar{J}_d = (\sqrt{2}b/2S^2) J_d$  is rescaled  $J_d$ , and  $Li_n(y) = \sum_{m=1}^{\infty} y^m / m^n$  is a polylogarithm function which becomes the Riemann- $\zeta$  function when  $y=1$ . The polylogarithm function allows us to express the coefficients of temperature series for Fermi distribution in which  $y$  is negative. The feature of the core spin of DE is that it has similar terms  $T^{3/2}$ ,  $T^{5/2}$ ,  $T^{7/2}$ , . . . as that of Heisenberg system and a new term  $T^3$ . This new  $T^3$  term comes from motional character of DE representing a characteristic feature for DE system. On the other hand, the dominant terms in the series of  $\langle S_z \rangle$  are  $3/2$  and  $5/2$  terms indicate the DE system behavior is close to a localized Heisenberg system. This is consistent with the experiments.<sup>17</sup> If the temperature is just below  $T_C$ , we can expand  $G(\mathbf{k})$  and thus  $\langle S_{tz} \rangle$  in powers of  $1/T$ . Since the spectrum in the Fermi distribution does not include  $\langle S_z \rangle$  factor explicitly, the expansion of this part has no meaning. However, we can expand the Bose distribution to express the Curie temperature by a parameter  $q_c$  and using  $\langle S_z \rangle \rightarrow 0$  we obtain

$$\frac{k_B T_C}{\bar{J}_d} = \frac{2z(S+1)}{9F^2(-1)} [(4S+1)F(-1) - (S+1)] q_c \frac{\sqrt{2}S^2}{b}, \quad (11)$$

where we follow Callen's definition  $F(n) \equiv 1/N \sum_{\mathbf{k}} \{ [J_d(0) - J_d(\mathbf{k})]/J_d(0) \}^n$  and  $F(-1)$  has been evaluated by Watson<sup>18</sup> to have a value of 1.51638. The  $k_B T_C/\bar{J}_d$  for DE has a factor  $q_c \sqrt{2}S^2/b$  comparing with that of ordinary Heisenberg system. The  $q_c$  can be determined from two coupled equations derived from the Fermi distribution with independent variables  $q_c$  and  $u$ ,

$$q_c = \frac{\sqrt{2}b}{2S^2} \frac{1}{N} \sum_{\mathbf{k}} \gamma(\mathbf{k}) \tanh \left\{ \frac{\delta}{2q_c} [\gamma(\mathbf{k}) - u] \right\}, \quad (12)$$

$$x = \frac{1}{2} - \frac{1}{2N} \sum_{\mathbf{k}} \tanh \left\{ \frac{\delta}{2q_c} [\gamma(\mathbf{k}) - u] \right\}, \quad (13)$$

where

$$\gamma(\mathbf{k}) = \frac{J_d(\mathbf{k})}{J_d(0)},$$

$$\delta = \frac{3\sqrt{2}F(-1)[(2bS+3S+2b)F(-1) - 2b(S+1)]}{2S(S+1)[(4S+1)F(-1) - (S+1)]},$$

$$u = \frac{3\mu_S F(-1)}{\sqrt{2}[(2bS+3S+2b)F(-1) - 2b(S+1)]J_d(0)}.$$

It is clear from the two equations that  $q_c$  is dependent on the hole concentration  $x$ , as well as the crystalline geometry factor  $F(-1)$  of the materials. The  $x$  dependence of  $q_c$  for simple cubic can be obtained by a numerical method<sup>19</sup> with  $S=3/2$  for CMR manganites. From the above results, we can estimate the Curie temperature  $T_C$ . Figure 2 shows the  $x$  dependence of  $T_C$  with  $J_d \sim 0.2$  eV as used by Millis *et al.*<sup>5</sup> The  $T_C$  has highest value at  $x=0.5$ , and  $k_B T_C/\bar{J}_d$  is about 33% that of ordinary Heisenberg magnet. The calculated  $T_C \sim 1050$  K for  $x=1/3$  is much higher than the experiment  $T_C \sim 250$  K (Ref. 20) for  $\text{La}_{0.67}\text{Ca}_{0.33}\text{MnO}_3$ . However, this is consistent with the theoretical result obtained from SCT by Millis *et al.*<sup>5</sup> who pointed out that a lattice degree of freedom might be added to explain CMR. As a matter of fact, according to band-structure calculations<sup>21</sup> of  $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$  that include the lattice distortion effect,  $J_d \sim 52$  meV and this value gives  $T_C \sim 270$  K for  $x=1/3$ . Note that in our calculation the dynamic effects of lattice and superexchange interactions are not included. These two factors may reduce the calculated  $T_C$ , however the calculated  $T_C \sim 270$  K agrees

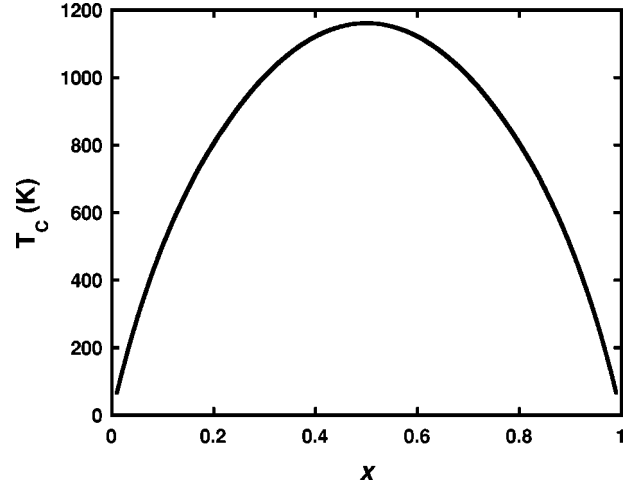


FIG. 2. The hole concentration  $x$  dependence of Curie temperature  $T_C$  with  $S=3/2$ ,  $J_d=0.2$  eV.

with experiments. In the high-temperature region, we obtain a series expansion in inverse powers  $T$  for the susceptibility,

$$\chi_T = \frac{g^2 \mu_B^2 \left(1 + \frac{1-x}{2S}\right)^2}{2k_B T} \left[ 1 + \left(\frac{T_s}{T}\right)^2 + O\left(\frac{1}{T^4}\right) \right], \quad (14)$$

where  $T_s$  is a characteristic temperature given by  $k_B T_s/\bar{J}_d = \sqrt{(2z/3b)S^3(S+1)}$ . We do not find the  $1/T^2$  term in agreement with Anderson and Hasegawa's result,<sup>3</sup> and furthermore,  $k_B T_s/\bar{J}_d$  is about 77% of that of a Heisenberg magnet derived from molecular-field theory<sup>15</sup> for  $S=3/2$  and  $z=6$ .

#### IV. CONCLUDING REMARK

We have investigated the magnetic properties of a DE system by using a quantum form of the Hamiltonian. The results for Curie temperature and the high-temperature series expansion of the susceptibility nicely reproduce those from the semiclassical treatment on DE. The core spin behavior is close to that of Heisenberg magnet at low temperatures and thus may explain the recent experiment.<sup>17</sup> These provide that the Hamiltonian we have obtained is a reasonable description for DE interaction.

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<sup>19</sup>The sum may be replaced by an integral over the first Brillouin zone. The integral can be approximately transformed into Gaussian quadrature summation then we can find numerical solutions for  $q_c$  and  $u$ . The Gaussian quadrature method can be found in

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<sup>20</sup>J.W. Lynn, R.W. Erwin, J.A. Borchers, Q. Huang, A. Santoro, J-L. Peng, and Z.Y. Li, Phys. Rev. Lett. **76**, 4046 (1996).  
<sup>21</sup>W.E. Pickett and D.J. Singh, Phys. Rev. B **53**, 1146 (1996); These authors obtain a total density of states  $D(E_F) \approx 0.85/\text{eV Mn}$  for  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$ . In our calculation, the Fermi spectrum is  $E(k) = \sqrt{2}zJ_d\gamma(k)$ . We assume a cubic Fermi surface at ground state and have

$$\frac{36\sqrt{2}(1-x)}{(2\pi)^3 z J_d (4-x)} \int_0^{\pi(1-x)^{1/3}} \int_0^{\pi(1-x)^{1/3}} \int_0^{\pi(1-x)^{1/3}} dudv \times \frac{1}{\sqrt{\sin^2 \pi(1-x)^{1/3} + \sin^2 u + \sin^2 v}} = D(E_F)$$

with  $x = 1/3$ , from this we obtain  $J_d \approx 52$  meV. The Fermi level argument and the density of states expression can be found in C. Kittel, *Introduction to Solid State Physics* (Wiley, New York 1996), Chap. 6; J. Callaway, *Quantum Theory of the Solid State* (Academic Press, New York, 1991), Chap. 1, respectively.